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Activities and geochronology of ^{137}Cs in lake sediments resulting from sediment resuspension

Gerald Matisoff

Department of Earth, Environmental and Planetary Sciences, Case Western Reserve University, Cleveland, OH, USA 44106-7216

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ABSTRACT

In lakes with a large surface area to watershed ratio ^{137}Cs delivery is primarily by direct atmospheric fallout to the lake surface, where its activity in the sediments has been used to estimate the exposure to organisms and sediment mass deposition rates. Comparison of ^{137}Cs in the historical atmospheric fallout record with ^{137}Cs activity profiles in sediment cores reveals that although the general features of a maxima in the fallout deposition can be matched to activity peaks in the core, the general shape of the ^{137}Cs profile is not an exact replica of the fallout history. Instead, the sediment reflects post-depositional processes such as resuspension, bioturbation, partitioning of ^{137}Cs between the sediment solids and the pore fluids, and molecular diffusion of ^{137}Cs through the pore fluids. Presented here is a model that couples these processes to a system time averaging (STA) model that accounts for the time history of ^{137}Cs fallout and the particle residence time in the water column or in the 'active' surface sediment subject to resuspension. Sediment profiles are examined by comparing reasonable ranges of each of the coefficients of each of these major processes and by applying the model to cores collected from two large, shallow lakes, Lake Erie (USA/Canada) and Lake Winnipeg (Canada). The results indicate that the STA model with molecular diffusion and sediment resuspension best describes the data from these large, shallow lakes.

1. Introduction

^{137}Cs has been detected in measurable quantities in sediments since the atmospheric nuclear weapons tests of the 1950s resulted in global stratospheric fallout. More recently, the accident at Chernobyl in 1986 distributed ^{137}Cs over much of Europe and the accident at Fukushima in 2011 had a more localized ^{137}Cs release. ^{137}Cs becomes strongly bound to particles and is delivered by direct deposition or by erosion and transport into receiving waters such as lakes. In-lake processes and resuspension of bottom sediment retain ^{137}Cs in the water column and in near surface sediment where organisms may be exposed to it for extended periods of time, so techniques are needed to calculate the ^{137}Cs activity in these settings as both a function of time and depth in the sediment.

Work in the 1960s and early 1970s led to the realization that vertical profiles of ^{137}Cs in lake sediments were related to the time history of radioactive fallout in the air so that sedimentation rates could be determined from these profiles (Ravera, 1961; Pickering et al., 1966; Schreiber et al., 1968; Ritchie et al., 1970; Krishnaswami et al., 1971). ^{137}Cs was first measurably detected in the early 1950s, so sediment at depth in a core with ^{137}Cs activity at the limit of detection has sometimes been assumed to be deposited in 1954 (Pennington et al., 1976; Ritchie and McHenry, 1990) although downward transport and modern low activities increase the uncertainty of that measurement. Atmospheric fallout in the northern hemisphere peaked in 1962–1963 so the ^{137}Cs peak activity in the sediment profile is assumed to represent 1963 (Krishnaswami and Lal, 1978), although because the residence time of the fallout in the atmosphere is about 1 year (Lal and Rama, 1966; Ritchie et al., 1973; respectively) this peak activity layer is sometimes assumed to represent 1964 (Ritchie and McHenry, 1990). Other ^{137}Cs activity horizons in the sediment profile have also been used as time stratigraphic markers (secondary activity peaks in 1958 and 1971), although these activity horizons are not always present and their use may actually be in error because post-depositional mixing can move sediment downward (Robbins and Edgington, 1975). In some locations, fallout from Chernobyl enables a 1986 time horizon to also be identified (Erlinger et al., 2008; Blebea-apostu et al., 2012). Using these ^{137}Cs activity layers as

E-mail address: gerald.matisoff@case.edu.

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stratigraphic markers enables sedimentation rates (cm y^{-1} or g cm^{-2} y^{-1}) to be calculated as the sediment depth (cm or g cm^{-2}, respectively) divided by the number of years between core collection and the year of the peak fallout. Identification of more than one time horizon enables calculation of sedimentation rates for multiple time intervals.

Comparison of ^{137}Cs in the historical atmospheric fallout record with ^{137}Cs activity profiles in sediment cores reveals that although the general features of a maximum in the fallout deposition can be matched to an activity peak in the core, the general shape of the ^{137}Cs profile is not an exact replica of the fallout history. Instead, most ^{137}Cs activity profiles in cores display moderate activities in the upper portion of the cores even though there is no fallout currently, one broad activity maximum spread out over several centimeters of the core if the core does not record Chernobyl fallout or two broad activity maxima if Chernobyl fallout is captured by the core, and a long downward decrease in activity deeper in the profile than the lower activity maximum. There are a number of explanations that have been put forward to account for these observations, including the time history of ^{137}Cs derived from the watershed (Davis et al., 1984; He et al., 1996), post-depositional remobilization by bioturbation or physical mixing (Robbins and Edgington, 1975) or sediment resuspension (Robbins et al., 2000; Schloesser et al., 2014), sorption kinetics of ^{137}Cs to the clay minerals in the sediment (Comans et al., 1989; Smith and Comans, 1996), and partitioning of ^{137}Cs between the solid phase and the dissolved phase coupled with molecular diffusion of ^{137}Cs through the pore fluids (Davis et al., 1984; He et al., 1996). Particularly noteworthy is that the shape of the ^{137}Cs profile in the sediment provides information not only about the time history of radioactive fallout but also has integrated information about the watershed, in-lake, and post-burial processes that have occurred following atmospheric deposition. Proper interpretation of the ^{137}Cs profiles requires a model of not only one of the processes described above, but of all the multiple processes that have affected the depositional history and redistribution of ^{137}Cs in a sediment column. In a system such as a reservoir or small lake with a small hydraulic residence time and a large (drainage basin)/(lake area) ratio the watershed may contribute in a major way to the sedimentary load of the isotope. In contrast, in large lakes such as the Great Lakes, very little ^{137}Cs originates from the watershed and with the majority of ^{137}Cs reaching sediments from direct atmospheric transfer any isotope originating from the watershed will be integrated and be indistinguishable from the atmospheric component (McCall et al., 1984). Presented here is an exploration of these processes in large lakes where the majority of ^{137}Cs delivered to the lake is from direct atmospheric deposition. Here a diagenetic model is used that includes post-depositional bioturbation, physical mixing, partitioning of ^{137}Cs between the sediment solids and the pore fluids, and molecular diffusion of ^{137}Cs through the pore fluids. This diagenetic model is coupled to a system time averaging (STA) model that accounts for the time history of ^{137}Cs fallout and the particle residence time in the water column or in the 'active' surface sediment subject to resuspension. Sediment profiles are examined by comparing reasonable ranges of each of the coefficients of each of these major processes and by applying the model to cores collected from two large, shallow lakes subject to frequent bottom sediment resuspension, Lake Erie (USA/Canada) and Lake Winnipeg (Canada).

2. Methods

2.1. Study areas

Lake Erie is the shallowest and smallest of the Laurentian Great Lakes in North America. Station 91 M is located at 41.8410° N, 082.9163° W in the shallow western basin and has a water depth of 10.3 m (Fig. 1). The water column occasionally stratifies for up to a few days, but it is frequently thoroughly mixed. The Western Basin has a hydraulic residence time of about 50 days. Station 43 is located in the central basin at 41.7882° N, 081.9455° W and has a water depth of 22.4 m (Fig. 1). The water column at this deeper water site undergoes annual thermal stratification, but also undergoes spring and fall overturn. The hydraulic residence time for the lake is 2.6 years. Sediments had a median particle diameter of about 10 μm and consist of about 41% quartz, 32% clay (mostly illite), 17% alkali feldspar, 6% dolomite and 4% organic carbon.

Lake Winnipeg is divided into a shallow South Basin (10% volume; mean depth = 9 m; water residence time = 1.3 y), connected by a narrow, deep channel to a larger, deeper North Basin (81% volume; water residence time = 4.3 y). Station 500 was located at 50.79472 N, 96.73306 W in the South Basin at a water depth of 10.6 m. The water column occasionally stratifies for up to a few days, but it is frequently thoroughly mixed. Station 505 was located at 53.39638 N, 98.47500 W in the North Basin at a water depth of 16.5 m (Fig. 1). The water column at this site undergoes annual thermal stratification, but also mixes regularly and experiences spring and fall overturn. The majority of the hydrologic loading is delivered to the South Basin by two major tributaries, the Winnipeg River and Red Rivers, and the largest external particulate load is delivered from the Red River. Sediments averaged 40% silt (mostly quartz and feldspar) and 60% clay (mostly clay minerals) with about 3% organic carbon and minor amounts of dolomite (Simpson et al., 2003).

2.2. Core collection

In Lake Winnipeg, 30 cm or longer cores 8.65 cm diameter were collected from the MV Namao from both the South Basin (Station 500) and the North Basin (Station 505) in Sept/Oct 2013. The cores were sliced at 1 cm intervals on board the ship, kept at 4 °C, and then subsequently freeze-dried for analysis. In Lake Erie, 6.7 cm diameter cores 55 cm long were collected using a Benthos corer from the EPA ship R/V Lake Guardian from both the Western Basin (Station 91M) and the Central Basin (Station 43) in June 2013. Upon retrieval the cores were immediately capped and stored vertically at 4 °C until transferred to the laboratory where they were sectioned at 1 cm intervals, dried at 60 °C, ground and analyzed.

2.3. Gamma spectroscopy

Ten to fifteen grams of ground sediments were placed into 47-mm polystyrene petri dishes for analysis by gamma spectroscopy for ^{137}Cs (661.7 keV) (Larsen and Cutshall, 1981). We used either an EG&G N-type HPGe gamma detector or a Canberra LEGe detector for the radionuclide analyses. Samples were counted for 22–24 h to decrease the counting errors (~10–20%) associated with small samples and low radionuclide activities. Standards were prepared using the same geometries as the samples. Counting efficiencies were established using two Nycomed Amersham® mixed radionuclide standards (QCY44 & RBZ44). All measured counts were corrected for background levels, detector and geometry efficiencies, branching ratios and decay, and were then divided by their respective sample weights and reported as activity per gram. Activities were decay corrected to the date of collection. Quality assurance was established using the National Bureau of Standards Columbia River Sediment (SRM 4350B) with a reported ^{137}Cs activity (±1 S.D.) of 0.029 ± 0.002 Bq g^{-1}. Measured activities of ^{137}Cs on the EGG detector averaged 0.031 ± 0.003 and on the Canberra detector averaged 0.028 ± 0.004. Uncertainty was calculated with

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