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High-resolution historical records from Pettaquamscutt River basin sediments: 1. ²¹⁰Pb and varve chronologies validate record of ¹³⁷Cs released by the Chernobyl accident

Ana Lúcia Lima,^{1,*} J. Bradford Hubeny,² Christopher M. Reddy,¹ John W. King,² Konrad A. Hughen,¹ and Timothy I. Eglinton¹

¹Department of Marine Chemistry and Geochemistry, Woods Hole Oceanographic Institution, Woods Hole, MA 02543, USA ²Graduate School of Oceanography, University of Rhode Island, Narragansett, RI 02882, USA

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Abstract—Cesium-137 derived from the explosion of the Chernobyl reactor in 1986 was preserved in anoxic sediments from a coastal environment in southern Rhode Island. Although the radioactive plume was detected in surface air samples at several locations in the United States, this is the first known record of a Chernobyl 137 Cs peak in sediments from North America. The inventory of Chernobyl 137 Cs that was preserved in the Pettaquamscutt River is small compared to European counterparts and should only be detectable for the next 15–20 yr. However, the presence of two 137 Cs peaks (1963 and 1987) identifies a well-dated segment of the sediment column that could be exploited in understanding the decomposition and preservation of terrestrial and aquatic organic matter. Different methods for calculating the 210 Pb chronology were also evaluated in this study and checked against independent varve counting. The end result is a detailed chronology of a site well suited for reconstruction of historical records of environmental change. *Copyright* © 2005 Elsevier Ltd

1. INTRODUCTION

On April 26, 1986, a flaw in design and a series of operator actions caused the Chernobyl-4 reactor to explode during a test to determine how long the turbines would spin after a loss of electrical power supply. The initial release of fission products to the atmosphere was followed by a second explosion, which allowed air to flow into the core and caused the graphite moderator to burst into flames (Hohenemser et al., 1986; Mould, 2000). The nine-day fire that followed was responsible for the main release of radioactivity into the environment. Roughly all of the xenon gas, 20% of the cesium and iodine, and $\sim 5\%$ of the remaining radioactive material in the reactor was set free by the accident, accounting for a release of more than 8×10^{18} Bq (1 becquerel = 1 disintegration per second) of fission products into the atmosphere (Mould, 2000) (http:// www.world.nuclear.org). While most of the released material was deposited close to the site of the accident in northern Ukraine, southern Belarus and Russia's Bryansk region (Stone, 2001), strong winds carried a plume towards Finland and Sweden (ApSimon and Wilson, 1986; Mould, 2000). By May 2, the plume had reached the UK and Japan, and by May 6, Canada and the United States (Ayoama et al., 1986; Smith and Clark, 1986; Mould, 2000). Even though ¹³⁷Cs was detected in the atmosphere in several regions of the United States following the accident (Larsen et al., 1986; Feely et al., 1988; Holloway and Liu, 1988), no record of a clear Chernobyl ¹³⁷Cs peak is observed in sediments from Florida (Robbins et al., 2000), Massachusetts (Spliethoff and Hemond, 1996), California (Fuller et al., 1999) or other locations in the country (Van Metre et al., 1997). However, a discernible Chernobyl ¹³⁷Cs peak in varved sediments from Nicolay Lake, Cornwall Island in the Arctic Ocean (Lamoureux, 1999) implies that insufficient depth resolution and/or bioturbation may explain the absence of the Chernobyl peak in some of the aforementioned sites.

Significant levels of ¹³⁷Cs first appeared in the atmosphere in the early 1950s as a result of above ground nuclear weapons testing. The number of nuclear detonations reached its highest in 1962, resulting in a maximum in ¹³⁷Cs fallout the following year. As a consequence of the nuclear weapons Limited Test Ban Treaty instated in 1963 (Carter and Moghissi, 1977), little radioactive fallout was observed in the late 1960s and 1970s in the northern hemisphere. Because ¹³⁷Cs deposition reflects the history of nuclear tests, this artificial radionuclide is commonly used as a chronostratigraphic marker to constrain dating records (Anderson et al., 1988; Ritchie and McHenry, 1990; Spliethoff and Hemond, 1996; Appleby, 2001). Following the 1986 Chernobyl accident, the atmospheric concentration of 137 Cs in Europe remained ~4-times higher than the 1963 levels for several months (Cambray et al., 1987) and a number of investigations reported the presence of Chernobyl-derived ¹³⁷Cs in sediment traps from the Black Sea and a lake in Switzerland (Buesseler et al., 1987; Wieland et al., 1993) and in surficial sediments from Denmark (Ehlers et al., 1993), Netherlands (Zwolsman et al., 1993), Switzerland (Dominik and Span, 1992; Gunten et al., 1997; Albrecht et al., 1998) and UK (Gevao et al., 1997). At these locations, elevated activities of ¹³⁷Cs imply that the 1986 peak can serve as a valuable sedimentary marker for several decades (137 Cs half-life = 30.2 yr).

As part of a study to develop historical records of combustion (Lima et al., 2003), we collected sediment cores from an estuarine anoxic basin site in southern Rhode Island and generated detailed ²¹⁰Pb, ¹³⁷Cs, and varve chronologies. The anoxic nature of this environment inhibits bioturbation, creating undisturbed laminations that are ideal for sediment dating. Here, the varve chronology is used to validate the dates generated by ²¹⁰Pb models, and not the other way around. We revisit the ²¹⁰Pb models used for dating recent sediments and show that ¹³⁷Cs derived from the Chernobyl accident was

^{*} Author to whom correspondence should be addressed (alima @whoi.edu).



Fig. 1. Map showing the boundaries of the watershed of the Pettaquamscutt River (RI) (dotted line) and the location of the site of sediment freeze-core collection. Modified after Lima et al. (2003).

preserved in sediments from a site in the Northeastern United States.

2. EXPERIMENTAL

2.1. Study Area

The Pettaquamscutt River, also known as the Narrow River, is located in Washington County, southern Rhode Island (Fig. 1). This estuary is ~9.7 km long, ranges from 100 to 700 m in width (Boothroyd, 1991) and has a small (35 km²) drainage area (Orr and Gaines, 1973). The Pettaquamscutt can be morphologically divided into two basins and a channel. The upper basin is 13.5 m deep at its maximum and receives input of freshwater from the Gilbert Stuart Stream. The lower basin is deeper (19.5 m), has a larger area and is confined to the north by a shallow sill (less than 1 m deep) and to the south by a long narrow channel that connects it to its salt-water source, Rhode Island Sound. The bottom waters and sediments of the upper and lower basins are permanently anoxic, mostly due to a stable salinity-dominated stratification of the water column (Gaines and Pilson, 1972). As a result, no bioturbation of the surficial sediments is observed and annually laminated layers are well preserved (Fig. 2).

2.2. Sampling

Freeze-cores were collected in the deepest part of the lower basin (Fig. 1) in April 1999 (Lima et al., 2003). Unlike gravity coring, which can disturb the surficial sediments and lead to compaction of sediment layers, freeze coring allows for recovery of intact sediment-water interfaces (Shapiro, 1958). Consequently, this sampling technique is ideal for high-resolution records of aquatic sediments, and especially those containing high amounts of siliceous tests that render sediments flocculant (Koide et al., 1973). Before lowering into the water, the aluminum corer ($30 \times 8 \times 165$ cm) was filled with a slurry of dry ice

and methanol. The corer was lowered to approximately 2 m above the sediment-water interface, allowed to drop into the sediment and left there for $\sim 10-15$ min, so that a thick slab of sediment froze onto the metal surface of the corer. After collection, the sediment slabs were separated from the corer, wrapped in aluminum foil, kept in dry ice, and transported back to the laboratory where they were stored in a chest freezer (-18°C). X-radiographs of the frozen slabs collected in 1999 showed laminated sediments and confirmed the absence of benthic animal burrows (Fig. 2). Here, we report results from the slab that showed the most distinct and the highest number of laminations on the X-radiographs. It is noteworthy that the cores did not show different number of laminations, the X-radiographs did. When we were choosing the cores, we were not looking at thin-sections, but at X-radiographs taken at a medical facility. Because the X-ray machine was not dedicated to X-raying sediments and the cores varied in thickness, different numbers of laminations were observable in each core. However, the sand layers deposited by the 1954 and 1938 hurricanes were distinct in every X-ray slide.

Before sampling the core, a 10-cm-wide subsection of the slab was cut lengthwise to be made into thin sections for subsequent varve counting (²¹⁰Pb measurements and varve counting were conducted on the same sediment slab). The frozen sediment was subsequently sliced using a compact tile saw equipped with a diamond wafering blade, while the slab was kept frozen by regular applications of liquid nitrogen. The sediment-water interface was sectioned at 1 cm, while the remaining of the core was sliced at 0.5 cm intervals. The samples were placed in precombusted glass-jars, air-dried, homogenized with a mortar and pestle, and stored until radiometric measurements and geochemical analyses were performed. The deepest portion of the lower basin of the Pettaquamscutt River has been sampled repeatedly over the years. In this paper we also refer to data obtained for freeze-cores collected in 1987, 2000, and 2003.



Fig. 2. Composite image of 20^{th} century laminations from the lower basin of the Pettaquamscutt River. Laminae deposited by historical hurricanes (1954, 1938) are marked with arrows and help constrain the varve chronology. The width of the composite has been exaggerated $2\times$; scale bar = 1 cm.

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