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Quantifying Sediment Accumulation and Accretion using ^{210}Pb and ^{137}Cs

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PURPOSE: This Dredging Operations and Environmental Research (DOER) Program Technical Note (TN) describes the use of the radionuclides ^{210}Pb and ^{137}Cs to measure rates of sediment accumulation and accretion and provides recommendations for best practices in field collection and laboratory analysis for U.S. Army Corps of Engineers (USACE) projects.

BACKGROUND: The ability to quantify the flux of sediment to an area and the corresponding change in bed or surface elevation is useful in both sediment dynamic and hydrodynamic studies. Rates of accretion and accumulation can be used to determine past rates of contaminant and nutrient burial, constrain sediment budgets, identify areas experiencing long-term sedimentation, evaluate site-wide variability in sedimentation, and validate sediment transport models. Numerous radionuclides can be employed as tracers or can provide age control in a broad range of geophysical studies. This TN focuses on the quantification of sediment accumulation and accretion rates using lead-210 (^{210}Pb ; half-life = 22.2 years) and cesium-137 (^{137}Cs ; half-life = 30.7 years) methods. These methods have been successfully applied in a variety of terrestrial, coastal, and marine environments (Baskaran and Naidu 1995; Baskaran et al. 2015; Boyd et al. 2017). Estuarine sediment budgets, reservoir infilling, and engineering methods to increase wetland sedimentation are common focuses of USACE projects which can utilize radionuclide-based rates of sediment accumulation and accretion. The DOER Program supports ongoing research to evaluate the accretion dynamics of thin-layer placement and restored marshes.

Sediment Accumulation and Accretion Rates. Sediment accumulation (*mass/area/time*) occurs when sediments are transported by a fluid (air or water) and subsequently deposited due to a reduction in the fluid's transport ability. Frictional forces reduce velocity and turbulence below a threshold allowing deposition of sediment. Sediment accumulation can also be initiated by flocculation, physical trapping, and bioaggregation of sediments. Rates of sediment accumulation are useful in determining sediment fluxes to an area and for determining fluxes of particles and contaminants adsorbed (e.g., Moser and Bopp 2001) to imported sediments used in geophysical and biogeochemical studies. Sediment accretion (*distance/time*) is the change in the vertical elevation of a surface over time due to the accumulation of sediment. The accretion rate is a component of elevation change where elevation change is the sum of accretion and subsidence or uplift.

A variety of methods exist to measure rates of accumulation and accretion. The selection of a method should be based on the study area and the time scale of accumulation as well as a careful evaluation of the project goals. These time scales of ^{210}Pb and ^{137}Cs methods average inter-annual and annual sedimentary events and are useful in historical contaminant burial, sea-level rise, and sediment budget studies. The half-lives of the radionuclides ^{210}Pb and ^{137}Cs allow for

the determination of rates over the range of approximately 10 to 100 years. These radionuclides can be used in depositional environments where they accumulate through direct or indirect atmospheric inputs. Additionally, their activity can be quantified via non-destructive gamma ray spectrometry. The geochemical cycle and measurement of ^{210}Pb and ^{137}Cs are discussed further.

Models of ^{210}Pb and ^{137}Cs Accumulation. The radionuclide ^{210}Pb occurs naturally and is a member of the uranium-238 (^{238}U) decay series. Figure 1 depicts the production and transport of ^{210}Pb . Radium-226 (^{226}Ra), a ^{238}U progeny, decays to a gas, radon-222 (^{222}Rn), some of which permeates from the substrate to the atmosphere. In the atmosphere, ^{222}Rn decays through a series of short-lived daughters (half-lives < 1 hour) to ^{210}Pb . There, ^{210}Pb is scavenged by aerosols and deposited on the surface via wet and dry deposition.

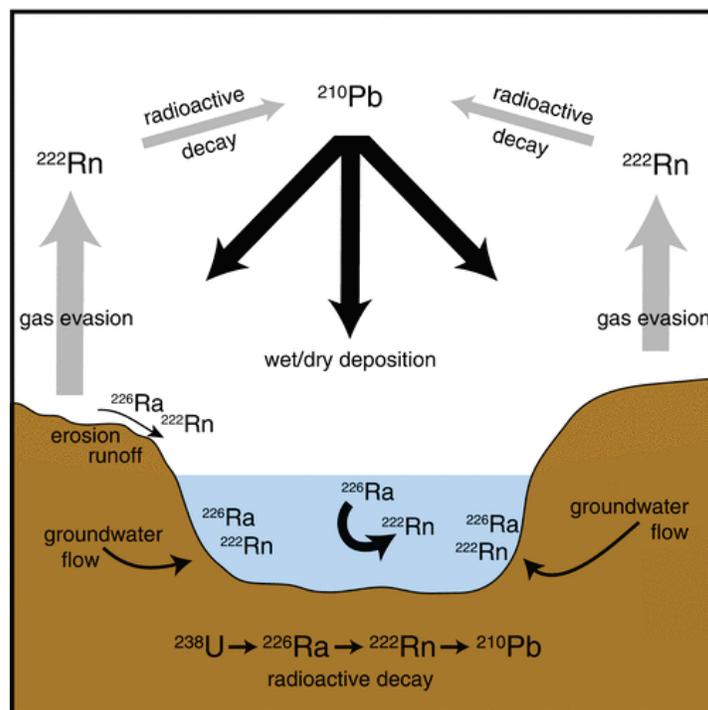


Figure 1. Conceptual depiction of geochemical cycle of ^{210}Pb . Reproduced with permission from Swarzenski (2015).

This atmospherically sourced ^{210}Pb contributes to an *excess* inventory in surface soils and sediments. Excess ^{210}Pb is indistinguishable from the ^{210}Pb generated in the soil, known as supported or in situ ^{210}Pb . The supported ^{210}Pb is determined indirectly through proxy or background measurements. The excess ^{210}Pb can then be calculated as the difference between the total ^{210}Pb and supported ^{210}Pb . The excess ^{210}Pb concentration profile and the known decay rate provide the basis for calculation of accretion rates (Figure 2).

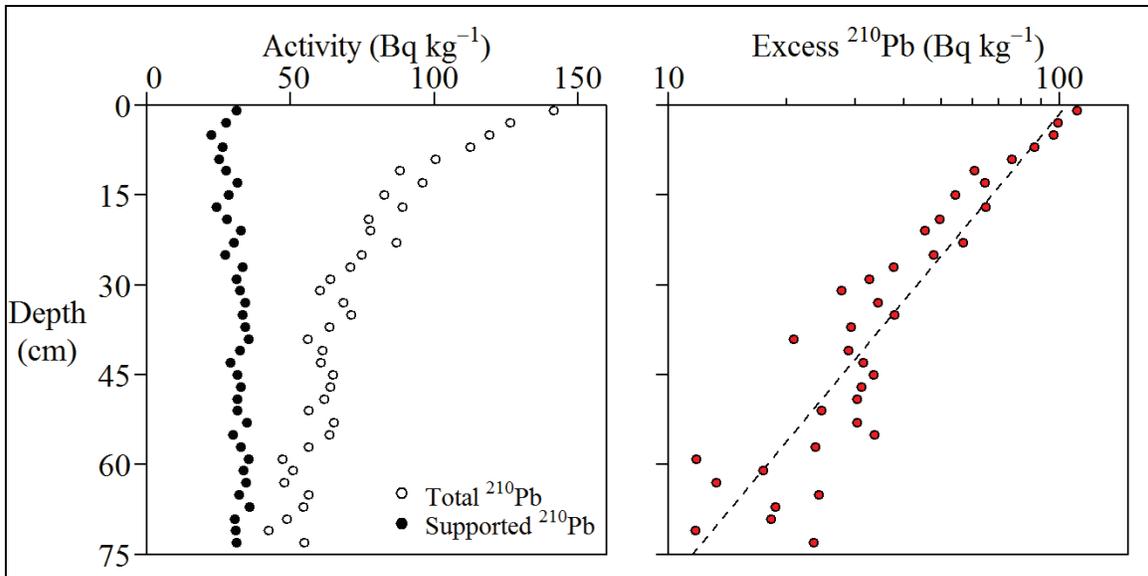


Figure 2. Profiles of total and supported ²¹⁰Pb (left) and excess ²¹⁰Pb (right) activity in Bequerels (Bq; 1 decay per second) per kilogram measured in a marsh sediment core collected in 2012. The difference of the total and supported ²¹⁰Pb is the excess ²¹⁰Pb. This excess ²¹⁰Pb is used to date sediments and determine accumulation rates. The least squares slope, change in the excess ²¹⁰Pb activity with depth, is related to the rate of burial.

A variety of ²¹⁰Pb dating models have been developed to determine an age-depth relationship from excess ²¹⁰Pb activity. Some such models are the Constant Flux- Constant Supply, Constant Rate of Supply, and Constant Initial Concentration. These models are similar in that they are based on the deposition of atmospheric ²¹⁰Pb but differ in the assumptions of the mechanism of ²¹⁰Pb delivery. Careful evaluation is needed to determine which, if any, models are valid for a given environment or site. Further information on the assumptions and application of these models can be found in Robbins (1978).

Anthropogenic ¹³⁷Cs is produced during the ²³⁵U fission reaction and is the most commonly used isotope for recent geochronology due to its well-defined input history. The application of ¹³⁷Cs employs the record of thermonuclear weapons testing fallout and its subsequent accumulation on surface sediments (Figure 3). Testing of weapons capable of injecting radioactive debris into the stratosphere began in 1952 with the “Ivy Mike” test on Enewetak Atoll. Such testing peaked in 1963 followed by a rapid decline due to the adoption of the Test Ban Treaty of 1963 (known as the “Partial Test Ban Treaty”) by the United States, United Kingdom, and Union of Soviet Socialist Republics (U.S.-U.K.-U.S.S.R. 1963). Widespread Northern Hemisphere atmospheric weapons testing was continued by non-party nations until finally ceasing in 1980.

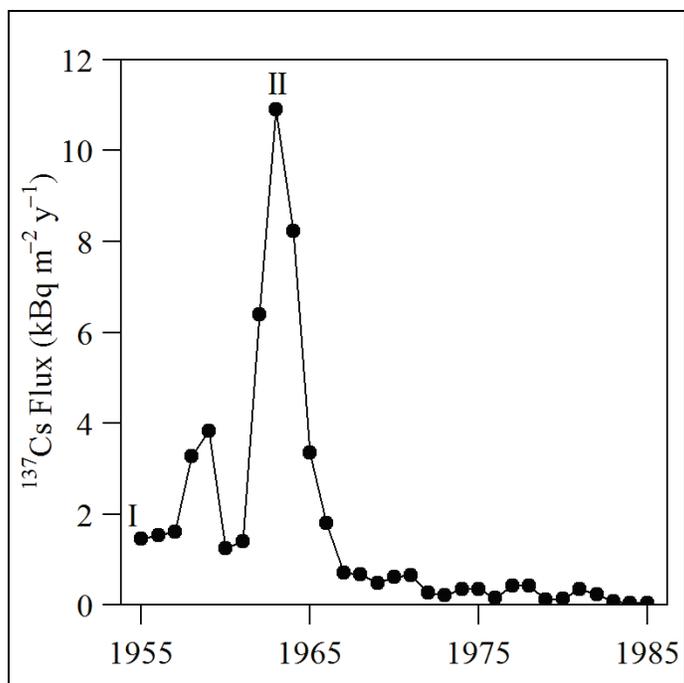


Figure 3. The 1955–1985 record of ^{137}Cs fallout at New York, NY, USA. The fallout record was inferred from the ^{90}Sr fallout measured by Environmental Measurements Laboratory (1999). The main events used in ^{137}Cs chronology are (I) 1954 onset of Northern Hemisphere thermonuclear weapons testing fallout and (II) peak weapons testing fallout. This fallout record is used to provide age control to sediment and soil deposits.

The 1954 onset and 1963 peak fallout of weapons testing can often be identified in a continuous sediment record and used to calculate rates of sediment accumulation and accretion (Figure 4). As ^{137}Cs decays to effective extinction in the early 2100s, another fallout-sourced radionuclide, ^{241}Am , will likely be utilized as a direct replacement (Boyd and Sommerfield 2017). This gamma emitting radionuclide ingrows from decay of ^{241}Pu that was deposited concurrently with ^{137}Cs in nuclear fallout, albeit in lower concentrations (~17% of ^{137}Cs activity).

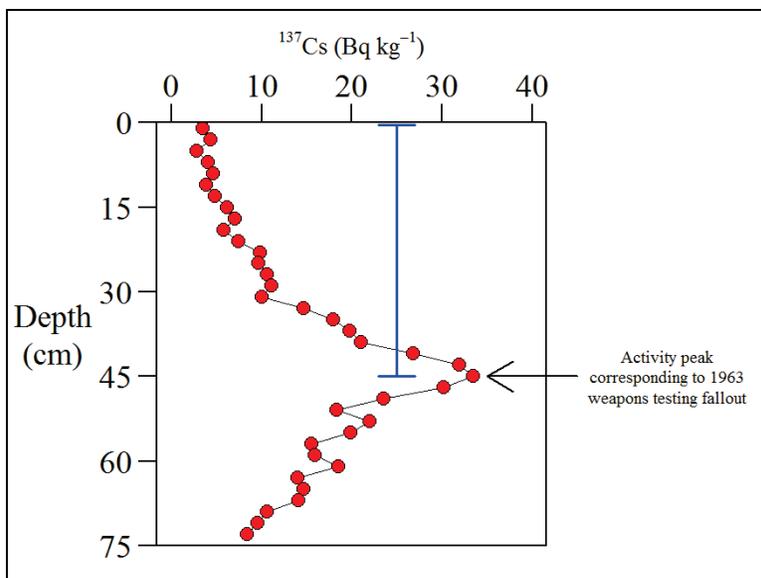


Figure 4. A ¹³⁷Cs profile measured in a salt marsh sediment core collected in 2012. Identification of the 1963 weapons peak at 45 centimeters (cm) depth equates to an accretion rate of 0.92 cm year⁻¹. The product of the sediment mass inventory (g cm⁻²) and the accretion rate yields a mass accumulation rate (g cm⁻² y⁻¹).

Radionuclide Measurement. Gamma rays, which are high-energy photons, can be emitted through radioactive decay, and their energy can be used to identify the nuclide of origin. Gamma spectrometry involves the detection and quantification of these photons to determine the concentration of radionuclides present in the sample (Gilmore 2008). ²¹⁰Pb has a 46.5 keV gamma-emission during beta decay to ²¹⁰Bi. The 661.7 keV gamma ray associated with ¹³⁷Cs is emitted when the daughter ^{137m}Ba decays to ¹³⁷Ba. While other methods of quantifying radioactivity exist, gamma spectrometry is a non-destructive method requiring no chemical preparation capable of simultaneous measuring multiple radionuclides. Thus, gamma spectrometry allows for the use of multiple radionuclide tracers in a given study, if applicable. Gamma spectroscopy is commonly accomplished using a high purity germanium detector. Figure 5 shows the gamma spectroscopy system at the U.S. Army Engineer Research and Development Center in Vicksburg, MS, that is available for radionuclide analysis. This ultra-low background system utilizes a 70 millimeter (mm) diameter planar crystal that allows for measurement of low-level radioactivity from a variety of different sample types and geometries.

Figure 6 shows an example gamma spectrum with count peaks highlighted for ²¹⁰Pb and ¹³⁷Cs. Additionally, samples prepared for gamma spectrometric analysis have the capability of being reanalyzed as well as used in further analysis such as those for stable isotopes, contaminants, or nutrients.



Figure 5. The gamma spectroscopy system operated at the ERDC. This system utilizes a 70 mm planar germanium detector, an ultra-low background Pb shield, and a liquid nitrogen recycling system allowing for continuous, high-resolution analysis of low-activity samples.

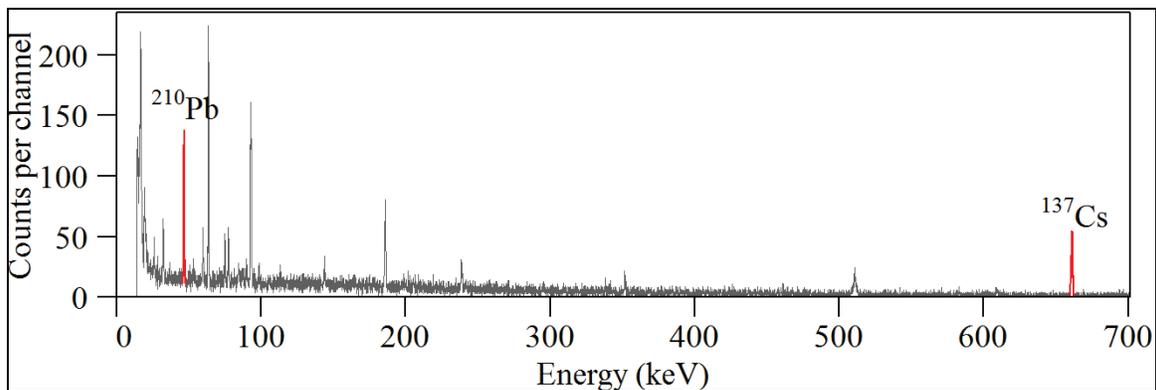


Figure 6. An example spectrum resulting from gamma spectroscopy showing photon counts detected for ^{210}Pb and ^{137}Cs (shown in red) over a 24-hour analysis. The area of the count peak is used to determine the radionuclide activity. This peak area is directly related to the radionuclide activity of the sample.

APPLICATION: Ensuring confidence in radionuclide-based rates begins during sample collection, where retrieval of a core representative of the sediment record is essential. Coring methodology is dependent on substrate and environment. Some field reconnaissance or knowledge of field conditions is helpful in determining sampling protocol. Additionally, preliminary gamma spectroscopy can determine if collection procedures will yield adequate sample mass and if modifications to sampling protocol are necessary (i.e., larger diameter sediment cores). The use of ^{210}Pb and ^{137}Cs dating models together, when applicable, provides cross validation and additional confidence in derived rates.

Radionuclide dating can be used at a fine spatial scale in estuaries, rivers, lakes, or coastal regions to quantify sedimentation for local features within a given water body. The following examples illustrate the usefulness and constraints of the described radionuclide methods in USACE activities in this variety of environments.

Determination of Sediment Accumulation Rates in Estuaries. Rates of sediment accumulation in bays, mudflats, and tidal wetlands can be determined via ^{137}Cs and ^{210}Pb radionuclides (Olsen et al. 1981; Bartholdy and Madsen 1985; Boyd et al. 2017). These sedimentation rates can be used to constrain estimates of the amount of material being retained in an estuary (Kuzyk et al. 2009). Subaqueous sediment accumulation and accretion rates can be calculated via ^{210}Pb and ^{137}Cs methods where repeat bathymetric survey data to measure vertical change are not available or possible (Sommerfield et al. 1995).

Contaminant and Nutrient Transport and Burial. Cesium-137 and ^{210}Pb adsorb to particles and can be used as tracers of sediment, nutrients, and contaminants. These radionuclides can even be used as a proxy for certain contaminants (Santschi 1989). Historic rates of nutrient burial generated from ^{137}Cs methods were used to estimate the ability of restored marsh land to sequester nutrients (Andrews et al. 2008). Radionuclide-based rates have been used to measure carbon burial potential of coastal marshes (Unger et al. 2016). Cesium-137 was used to calibrate a contaminant transport model for the Great Lakes (Robbins 1985). Radionuclide geochronology can be used to determine the burial rates of contaminants (Heim and Schwarzbauer 2013). The presence of legacy radionuclides in surface sediments can indicate mobility of particle-associated contaminants due to bioturbation (Bradshaw et al. 2006).

Rates of Reservoir Infilling. Reduction in capacity of reservoirs is a primary concern of water resource and flood managers. One specific problem is generating rates of sedimentation to use in models and for management decisions (Randle et al. 2013). Generation of accumulation rates from ^{137}Cs and ^{210}Pb profiles have been shown to be an effective method of measuring reservoir infilling (McCall et al. 1984; Baskaran et al. 2015). Radionuclide rates have been used to identify sediment accumulation patterns related to depth in small lakes (Evans and Rigler 1983). The ^{137}Cs and ^{210}Pb dating models have been applied successfully in lakes (Krishnaswamy et al. 1971) where the sediment inventory of the radionuclides can be used to identify sediment depocenters, or *hot spots* of deposition (Crusius and Anderson 1995). These depocenters occur due to focusing of sediments which results in elevated radionuclide concentrations. Radionuclide inventories, the total ^{137}Cs or ^{210}Pb contained in the sediments, can be used to identify depocenters, thereby further elucidating depositional dynamics in lake and reservoir environments. The shape of radionuclide profiles can be used to determine when

sedimentation is due to erosion from the water shed versus reworking of sediment from within the catchment (Zhang and Walling 2005).

Determination of Wetland Viability. Comparison of rates of sediment accretion and sea-level rise can be used to identify wetland locations with an accretionary deficit. These locations are the most likely to benefit from anthropogenic nourishment, including dredged material placement (e.g., Boyd and Sommerfield 2016). Additionally, ^{137}Cs - and ^{210}Pb -based sediment accumulation rates can be used to identify wetland sites that may have a sediment deficit relative to neighboring wetlands that may otherwise be similar in appearance or ecological metrics. While not a current practice, radionuclide-based accumulation and accretion rates can be used to monitor recovery and determine the long-term efficacy of wetland restoration projects using dredge material.

SUMMARY: As demonstrated by previous work, ^{210}Pb and ^{137}Cs are useful tools for quantifying sediment accumulation and accretion in a variety of environments. Gamma spectrometry allows for the simultaneous measurement of these radionuclides while preserving the sample material for further analysis. Care must be taken in collection of representative cores and selection of applicable dating models. Radionuclide chronology can be used to determine rates of sediment accumulation and accretion for various sediment management and beneficial use applications.

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