Anthropocene 48 (2024) 100452

Contents lists available at ScienceDirect

Anthropocene

journal homepage: www.elsevier.com/locate/ancene

The environmental consequences of rapid urbanization in central Florida reconstructed with high resolution ^{241}Am and ^{210}Pb dating in lake sediments

Paul W. Volante[a,*], James M. Kaste[a], Troy Clift[b], Matthew N. Waters[b]

[a] Geology Department, William & Mary, Williamsburg, USA
[b] Department of Crop, Soil, and Environmental Sciences, Auburn University, Auburn, USA

ARTICLE INFO

Dataset link: Revised Lake Bonny Dataset - Volante et al.

Keywords:
Sediment dating
Radionuclides
Pollution
Heavy metals
Florida
Urbanization

ABSTRACT

Throughout the 20th century, Florida was one of the fastest growing states in the US, putting unique environmental stress on the region. Accurately dated lake sediments can provide invaluable records of environmental change that extend beyond monitoring records. Here, we analyze profiles of americium-241 (^{241}Am), cesium-137 (^{137}Cs), lead (Pb), zinc (Zn), and uranium-series radionuclides in Lake Bonny in Lakeland, Florida. The ^{241}Am peak is sharp in the sediment profile, while the ^{137}Cs peak is broader and spread evenly across two layers. The measured ^{137}Cs inventory of ~413 Bq/m^2 is less than half of the expected inventory from atmospheric deposition (accounting for decay since deposition), indicating significant losses. The reliability of ^{137}Cs as a chronological tool can be complicated in environments with low quantities of 2:1 clays and low available potassium (K), characteristic of Florida and the U.S. southeast. Using a piecewise constant rate of supply ^{210}Pb model verified by ^{241}Am, we reconstruct sedimentation and chemical change in this lake. Highest sedimentation rates in the lake occur during decades of peak population growth in the mid-20th century. Uranium (U) and radium-226 (^{226}Ra) inputs to the lake reach a maximum in the 1960s, consistent with expansion of local phosphate mines and elevated groundwater pumping during that time in response to drought conditions. Total Pb in the sedimentary record captures the rise and fall of the use of leaded gasoline, but Zn inputs to the lake remain nearly two orders of magnitude above background levels in the last decade. Our high-resolution chronology of the lake reveals regional impacts on water and lake quality in central Florida during a period of rapid population growth.

1. Introduction

Humans have measurably impacted earth systems for thousands of years, but these changes intensified dramatically since the industrial revolution beginning around 250 years ago (Dong et al., 2021). Lake sediments record changes to the ecology, climate, air and water chemistry, hydrologic flows, and predominant geomorphic processes in a region over time (Pirrone et al., 1998; Zhang and Walling, 2005; Routh et al., 2007; Balascio et al., 2019). The analysis of gamma-ray emitting radionuclides via gamma spectrometry in sediment cores is a robust technique that can be used to contextualize chronologies of environmental change (Appleby et al., 1986 and Bruel and Sabatier, 2020). Dating models produced via gamma spectrometry can be precise enough to resolve chemical changes on annual to decadal time scales (Appleby and Oldfield, 1978). Three of the most widely used radionuclides in gamma spectrometry for creating age models of recent sediments include naturally occurring ^{210}Pb and anthropogenic ^{137}Cs and ^{241}Am released from atmospheric atomic weapon detonations primarily during 1952–1963. Dating models using ^{210}Pb ($t_{1/2}$ = 22.2 years), which is produced naturally in the atmosphere and in the earth's crust from continuous decay of radon, rely on the determination of 'excess' ^{210}Pb in sediments. The main source of excess ^{210}Pb in lake sediments is fallout from the atmosphere. This fallout enters the lake system directly via wet/dry deposition on to the lake surface, or indirectly through runoff from the surrounding lake catchment. Excess ^{210}Pb is measured by the extent to which total ^{210}Pb exceeds the "supported" ^{210}Pb that is produced by in situ decay of ^{226}Ra found within the sediments. The constant rate of supply (CRS) model assumes that the flux of excess ^{210}Pb to the catchment is constant over time and that the sedimentation rate can freely change (Goldberg, 1963; Appleby and Oldfield, 1978; Swarzenski,

* Corresponding author.
E-mail address: pwvolante@gmail.com (P.W. Volante).

https://doi.org/10.1016/j.ancene.2024.100452
Received 11 May 2024; Received in revised form 28 August 2024; Accepted 14 October 2024
Available online 23 October 2024

2014). ^{210}Pb dating models need to be corroborated with an independent tracer such as ^{137}Cs or ^{241}Am to confirm that the above assumptions have been met (Appleby, 2001).

The layer of a sediment core with the highest activity of ^{137}Cs and ^{241}Am is widely used as a reference point to the year with maximum fallout (1963 CE) to validate ^{210}Pb dating models (Beck and Bennett, 2002). ^{137}Cs ($t_{1/2}$ = 30.1 y) is a direct fission product of uranium-235, whereas ^{241}Am ($t_{1/2}$ = 432 y) is a decay product of plutonium-241 (^{241}Pu; $t_{1/2}$ = 14 y). ^{137}Cs has been more widely used as a dating proxy than ^{241}Am because its atomic abundance is orders of magnitude higher, which combined with a shorter half-life, produces a strong and easily observed gamma emission at 662 keV compared to the gamma emission of ^{241}Am at 59.5 keV. While ^{137}Cs is more abundant than ^{241}Am, two half-lives have elapsed since maximum deposition in 1963, so ^{137}Cs activities in sediments and soils are approaching less than a quarter of the initial amount deposited during the atomic weapon test period (Drexler et al., 2018). As a decay product of ^{241}Pu, ^{241}Am is actively in-growing in sediments and soils and will reach a maximum activity in 2037 (Appleby et al., 1991).

Both ^{137}Cs and ^{241}Am bind to organic matter and secondary minerals with contrasting affinities. ^{137}Cs has been observed to become permanently trapped in the interlayer of 2:1 clays like illite (Fuller et al., 2015). However, in environments with highly weathered clays such as bauxite or gibbsite, ^{137}Cs is found to be geochemically mobile (Drexler et al., 2018), whereas ^{241}Am appears to fix more permanently to soil and sediment particles in a wide range of environments (Brezonik and Engstrom, 1998; Olid et al., 2008; Hansson et al., 2014). Additionally, it is well established that vegetation has the tendency to uptake ^{137}Cs because plant root receptors can mistake the $^{137}Cs^{+}$ cation for K^{+}, given that these cations have a +1 oxidation state and similar atomic radii (Dahlman et al., 1975; Zhu and Smolders, 2000). This is especially common in environments with low soil K (Kaste et al., 2021). Vegetation uptake of ^{241}Am is near zero and Am only exists as a +3 cation in the near-surface environment (Popplewell, 1984; Iurian et al., 2015). In regions with low 2:1 clay compositions and high uptake, ^{137}Cs is likely to be geochemically mobile, while ^{241}Am tends to be more immobile and thus more reliable as a dating tool.

During the early and mid-20th century, Florida experienced rapid population growth from 753,000 in 1910 to nearly 5 million in 1960 (US Census Data). Substantial increases in population, related construction, and stresses on water supplies result in environmental degradation and pollution. In Florida, common examples include eutrophication of lakes, measured in Lake Okeechobee (Brezonik and Engstrom, 1998) and Lake Apopka (Schelske et al., 2005) and deposition of Pb and other heavy metals (Kamenov et al., 2009; Escobar et al., 2013) among other types of pollution. Reliable sediment dating techniques are vital to understand the timing of historical pollution to measure changes in response to regulations and conservation efforts. However, the south-eastern region of the United States is an area that contains highly weathered clays, low available soil K, and abundant vegetation in its wetlands, lakes, and marshes. In these environments, ^{241}Am or other dating techniques may be necessary to verify ^{137}Cs or ^{210}Pb based dating models. We analyzed the gamma emitting radionuclides in a sediment core from a Lake Bonny, a eutrophic lake in Lakeland, FL, USA, to understand the chemical history of the lake and evaluate the reliability of ^{210}Pb, ^{137}Cs, and ^{241}Am dating models in a region with highly weathered clays, dense vegetation, and abundant cations that could desorb ^{137}Cs from sediment.

2. Methods

2.1. Study site and sample collection

Lake Bonny is a small (0.822 km^2), eutrophic, shallow (max depth 3.7 m) lake surrounded by the city of Lakeland, Polk County, Florida (28°2′5.31"N; 81°55'52.24"W) within the Peace River-Saddle Creek Watershed. The lake lies within the Lakeland/Bone Valley Upland, which consists of sandhills and many solution depression lakes. This region is covered in phosphatic and clayey sands from the Miocene-Pliocene Bone Valley Member of the Peace River Formation in the Hawthorn Group (Griffith et al., 1997). Lake Bonny is connected to the much larger Lake Parker to the north. Lake Bonny and Lake Parker drain into the Saddle Creek, Lake Hancock, the Peace River and finally the Gulf of Mexico.

A core was extracted from the deepest portion of the lake on July 15th, 2020 (Fig. 1) using a piston corer (Fisher et al., 1992). The core was sectioned in the field at 4-cm intervals and stored on ice in Whirl-Pak bags. Sections were frozen, freeze-dried, and ground with a mortar and pestle. Dry bulk density was measured, and then organic matter concentration was determined by weight loss on ignition (LOI) after combustion at 550°C for 3 hours. Total Pb, phosphorous (P), Zn, and other metals in each dried section were measured using an ARL 3560AES ICP analyzer following acid digestion using standard EPA methods (Griffith et al., 1997).

2.2. Gamma spectrometry

The activities of ^{241}Am, ^{137}Cs, ^{210}Pb, and other U-series radionuclides were measured via a lead and copper shielded Canberra (Mirion Industries) ‘broad energy’ intrinsic germanium detector. The efficiency calibration of the system for measuring U-series radionuclides was obtained by measuring the Canadian Certified Reference Materials Project BL- 4a ore, which has 0.1248 % U certified in equilibrium with all ^{238}U and ^{235}U daughters (Steger and Smith, 1985). The ^{235}U/^{238}U activity ratio can be assumed to be 0.04605 (Murray et al., 1987). The efficiency for ^{137}Cs and ^{241}Am measurements was determined via a certified solution containing those radionuclides from Eckert & Ziegler. Calibration standards of BL-4a or the E&Z solution were measured in the exact same geometry as the unknown sediment samples. For lower energy gamma emissions (<100 keV), including ^{241}Am and ^{210}Pb, a point-source transmission correction was applied using the self-attenuation equation reported by Cutshall et al. (1983). This compares the transmission rate of gamma rays from an enriched point source through the samples and standards to an empty container to correct for self-attenuation for the calibration process and sample activity determination.

Sediment from each core slice was packed into 50 mm diameter Falcon® ‘Tight-Fit Lid’ 12 mL polystyrene containers and weighed and sealed with paraffin wax to trap ^{222}Rn for the determination of ^{226}Ra via the gamma emissions of its short-lived granddaughters ^{214}Pb (352 keV) and ^{214}Bi (609 keV). The samples remained sealed for >3 weeks to allow saturation of the radon daughters (^{214}Pb, ^{214}Bi) in the container with respect to ^{226}Ra before gamma analysis. Total ^{210}Pb is determined via its attenuation-corrected 46 keV gamma emission (Cutshall et al., 1983) and ^{241}Am via its attenuation-corrected 59.5 keV emission. To determine ^{226}Ra activity in each layer, we use a combination of radon daughter emissions (^{214}Pb, ^{214}Bi) and the direct gamma emission of ^{226}Ra at 186 keV corrected for the calculated ^{235}U interference using the 63.3 keV photon (^{238}U) and the ^{235}U/^{238}U activity ratio of 0.04605 (Dowdall et al., 2004; Murray et al., 1987). In most layers, the ^{226}Ra values determined indirectly using ^{214}Pb and ^{214}Bi were identical to the direct interference-corrected determination by the 186 keV ^{226}Ra emission. In a few cases, the ^{214}Pb and ^{214}Bi values were slightly lower, which we attribute to radon leakage from cracks in the wax, in which case we use the value from the direct 186 keV line. We define excess ^{210}Pb in the sediment by subtracting the ^{226}Ra activity from the total ^{210}Pb measured in each layer. In layers below 110 centimeters of depth in the core, ^{210}Pb and ^{226}Ra were statistically indistinguishable, which confirms our calibration approach. Samples were counted for 3–5 days to reduce the counting uncertainty below 8 %. The detection limit for ^{210}Pb for a 5 g sediment sample counted for 240k seconds is approximately 5.5 Bq/kg.

Fig. 1. Bathymetric and satellite image maps of Lake Bonny, Lakeland Florida (28°02'31.9"N 81°55'45.5"W). Contours are in feet. Bathymetric map is modified from (Florida LAKEWATCH, 2003). Satellite imagery from Google Maps, 2024.

2.3. ^{210}Pb piecewise CRS model

The constant rate of supply (CRS) ^{210}Pb dating model is a technique commonly used to determine the age of sediments in a core. The CRS model assumes that ^{210}Pb deposition to sediment is constant over time while sedimentation rates can vary (Goldberg, 1963; Appleby and Oldfield, 1978; Swarzenski, 2014). In this study, we used the open-source package serac (ShortlivEd RAdionuclide Chronology) to create a piecewise CRS ^{210}Pb model (Bruel and Sabatier, 2020) in R (v. 4.2.1, R Core Team, 2022). A piecewise CRS model differs from a CRS model in that it allows the user to enter a known age with a corresponding depth to get a more accurate age model. We assigned the layer with the greatest ^{241}Am activity (52–56 cm) to the year 1963. Information such as dry bulk density, core slice thickness, and activity of excess ^{210}Pb are entered into a.txt file that is then accessed by the serac package to create the model.

3. Results

3.1. Total Pb, excess ^{210}Pb, and Zn

At the base of the core, extractable Pb concentrations range between

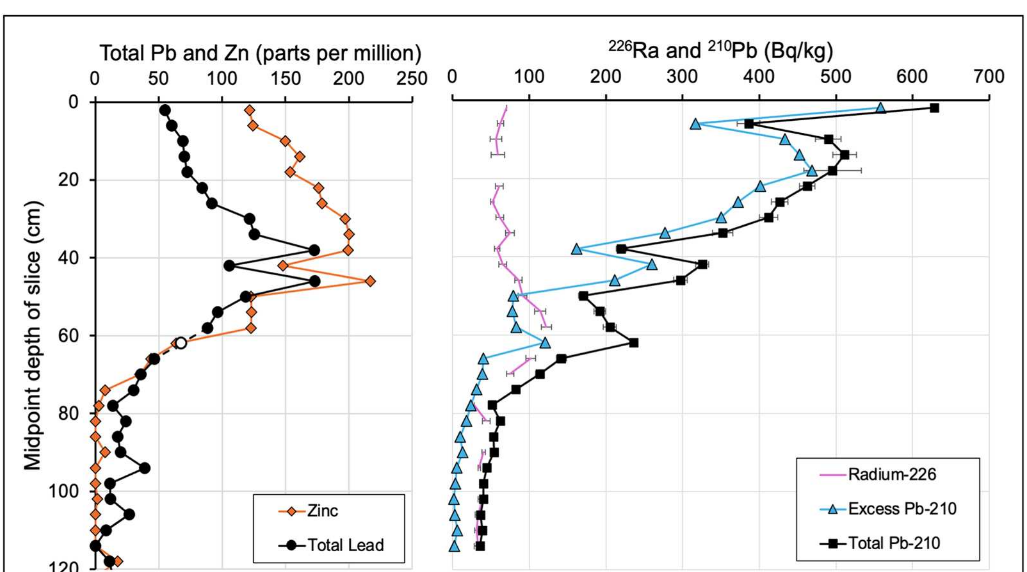

Fig. 2. Total Pb, Zn, ^{226}Ra, excess ^{210}Pb and total ^{210}Pb in the Lake Bonny core. A white dot indicates data is unavailable for that core slice and represents an average value of the layer above and below the missing layer data. The error bars for ^{226}Ra and total ^{210}Pb represent 2σ uncertainties.

5 and 12 ppm, which is near crustal average and reasonably represents the natural background (Lima et al., 2005). Pb concentrations gradually rise to around 25 ppm near 110 cm depth, then rise sharply between the 76–36 cm layers to a maximum of 173 ppm in the 44–48 cm and 36–40 cm layers (Fig. 2). Above this, Pb concentrations decline to near 50 ppm at the surface. The total excess ^{210}Pb inventory in the core is 8059 Bq/m^2. At depths below 110 cm in the core, ^{210}Pb is indistinguishable from ^{226}Ra. Above 110 cm, excess ^{210}Pb activity climbs nearly logarithmically, reaching a maximum of 588 Bq/kg at the surface. Zn is very low in the bottom half of the core (<10 ppm) but jumps to 35 ppm at 70 cm depth, rising to a peak of 217 ppm at 46 cm depth where Pb is also high. Unlike Pb, Zn remains elevated over 120 ppm in the upper layers.

3.2. Activities of ^{137}Cs and ^{241}Am

From the top of the core, ^{137}Cs activities are ~9 Bq/kg until the 44–48 cm layer (Fig. 3). After this layer, ^{137}Cs activities increase to the maximum of 18.2 Bq/kg in both the 48–52 cm and 52–56 cm layers. ^{137}Cs activities decline sharply down to 1.9 Bq/kg in the 68–72 cm layer. After this layer, ^{137}Cs activities are only intermittently detectable. The total inventory of ^{137}Cs we measured in the core is 413 Bq/m^2. ^{241}Am activities are mostly undetectable until the 32–36 cm layer, which contains an activity of 1.0 Bq/kg. ^{241}Am activities do not begin to increase significantly until the 48–52 cm layer with an activity of 2.6 Bq/kg. The 52–56 cm layer contains the highest ^{241}Am activity of 3.42 Bq/kg, which drops to 0.2 Bq/kg in the 64–68 cm layer before becoming undetectable in the following layers. The total inventory of ^{241}Am we measured in the core is 44.4 Bq/m^2.

3.3. ^{226}Ra activity and phosphorous

^{226}Ra activities are lowest in the period from 1867 to 1916, ranging from 28.0 to 43.8 Bq/kg (Fig. 4). ^{226}Ra activities increase steeply to a maximum of 122.1 Bq/kg in 1958. The ^{226}Ra activities then decrease to 57.8 Bq/kg in 1991. The ^{226}Ra activities remain around 60 Bq/kg up through 2020. P concentrations mirror ^{226}Ra activities. P concentrations are close 2000 ppm prior to 1930 but rise considerably up to just above 5000 ppm in 1958. P concentrations then gradually fall to between 2000 and 3500 ppm in the period after the peak.

3.4. Mass accumulation rates in Lake Bonny

The serac model estimates that the average sedimentation rate for 0–100 cm of depth is ~1.0 cm/yr. The total mass accumulation rate (MAR, units g/cm^2/yr) of each layer can be obtained by multiplying the sedimentation rate (cm/yr) by the dry bulk density (g/cm^3) for each layer. Furthermore, the MAR can be split into the organic and mineral MAR rates using the LOI results (Fig. 5).

Total MAR gradually rose from 0.056 g/cm^2/yr in 1903–0.082 g/cm^2/yr in 1941. The MAR was turbulent in the following 50-year period. The total MAR dropped to 0.039 g/cm^2/yr in 1953, rose to 0.083 g/cm^2/yr in 1963, fell once more to 0.028 g/cm^2/yr around 1980. From 1995 onwards, the total MAR stabilized around 0.04 g/cm^2/yr.

The organic MAR contribution is generally greater than the mineral MAR. However, from 1916 to 1928 and again from 1963 to 1991, the mineral MAR was greater than the organic MAR. In the period since 1991, the organic MAR has outpaced the mineral MAR.

4. Discussion

4.1. Comparison of ^{210}Pb, ^{241}Am, and ^{137}Cs profiles

The peak ^{241}Am activity of 3.2 Bq/kg occurs in the 52–56 cm layer, while the peak ^{137}Cs activity of 18.2 Bq/kg is spread equally across both the 48–52 and 52–56 cm layers (Fig. 3). The ^{241}Am peak has a full width at half maximum (FWHM) approximately half that of ^{137}Cs, which indicates a reduced geochemical mobility (Fig. 3). After the ^{137}Cs peak at 52–56 cm, ^{137}Cs activities plateau around 10 Bq/kg from this depth up to around 10 cm, whereas ^{241}Am activities decline quickly to <5 Bq/kg

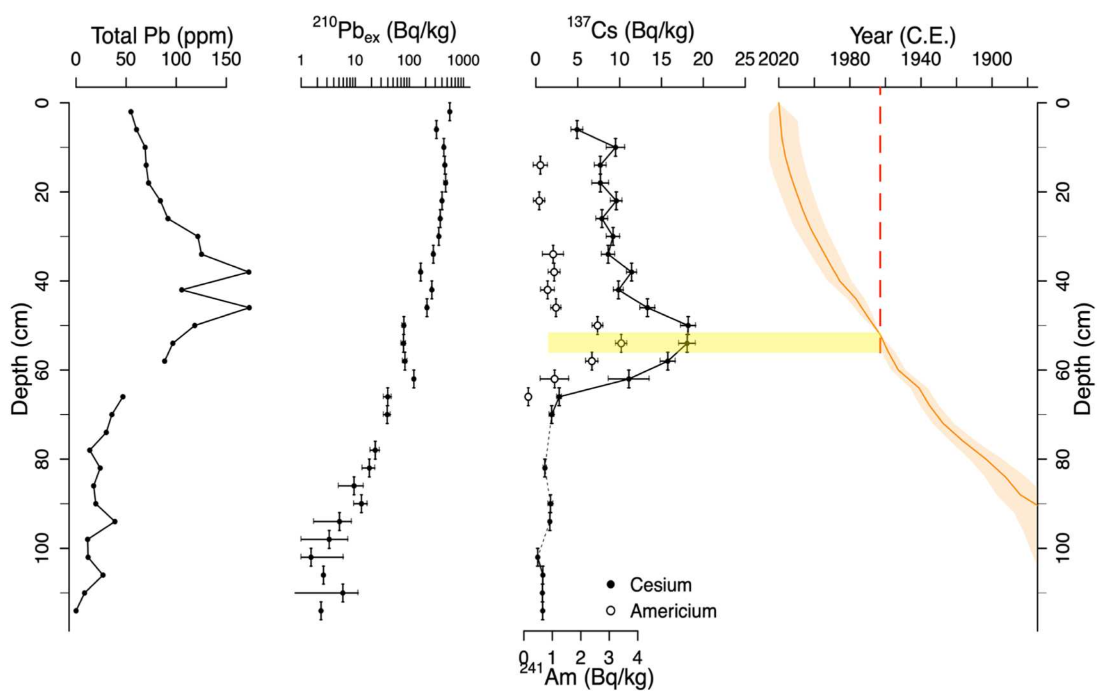

Fig. 3. Total Pb, ^{210}Pb$_{ex}$, ^{241}Am, and ^{137}Cs activity profiles in Lake Bonny. The highlighted region represents the 4 cm layer that contains peak ^{241}Am fallout. The dashed line symbolizes the year of peak fallout deposition, 1963 CE. Vertical error bars represent the 4 cm height of a sample. Horizontal error bars for ^{210}Pb$_{ex}$, ^{241}Am, and ^{137}Cs activities are 2σ uncertainties. The orange band surrounding the piecewise CRS model is the 1σ uncertainty in the age estimate. This figure was generated through the serac modelling package (Bruel and Sabatier, 2020).

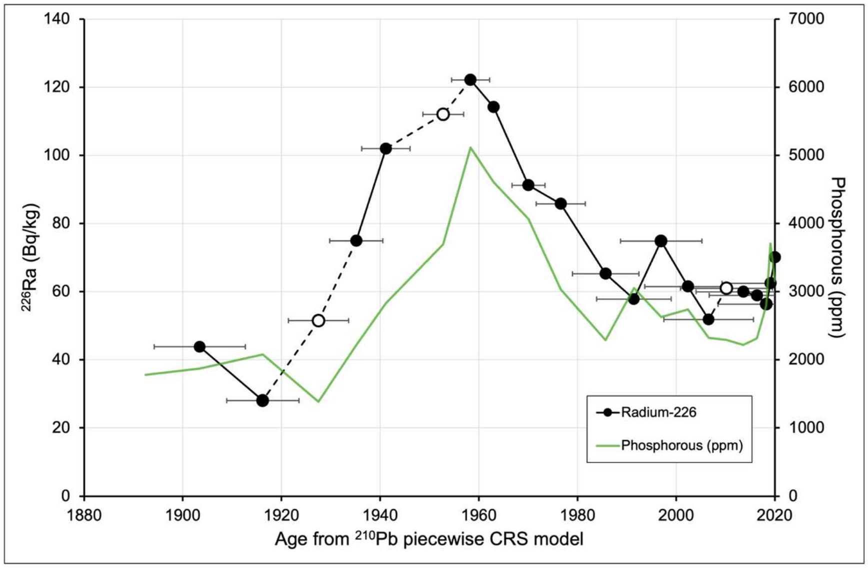

Fig. 4. ^{226}Ra (Bq/kg) and phosphorous with age from ^{210}Pb piecewise CRS model. A white dot indicates data is unavailable for that core slice and represents an average value of the layer above and below the missing layer data. Horizontal error bars represent 1σ age uncertainty estimates.

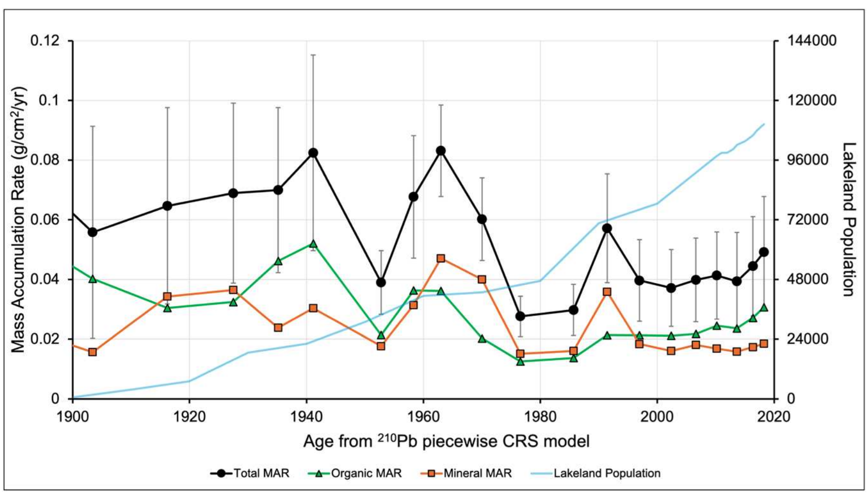

Fig. 5. Mass accumulation rates (MAR) in Lake Bonny. MAR were determined using dry bulk density of a core slice and the number of years contained in that slice (from the CRS model). The organic contribution was calculated via LOI.

from the peak at 44–48 cm layer up to the surface. It is interesting to note that ^{137}Cs activities remain around 10 Bq/kg for a long period after the peak and do not decrease significantly until 4–8 cm in depth. This could indicate that ^{137}Cs in Lake Bonny has remobilized and diffused into layers above the layer of maximum ^{137}Cs deposition. Some ^{137}Cs in layers above the peak could also be explained by the release of ^{137}Cs from the surrounding watershed (Davis et al., 1984). Due to the eutrophic state of Lake Bonny, it is probable that K^+ facilitates the desorption of Cs from exchange surfaces on organic and mineral sediment (Iurian et al., 2015; Drexler et al., 2018)

4.2. Pb and Zn pollution

Chemical changes in the Lake Bonny core give insights into anthropogenic activities in the 20th century in the Lakeland area. In the deeper sections of the sediment core, there are small total Pb peaks of 27 and 39 ppm that are dated to 1845 and 1868 respectively. The city of Lakeland was formally created in 1884, although indigenous people had already been living in the area that would become Polk County since at least the 1700 s (Covington, 1968; Mulligan, 2008). These early peaks in lead are consistent with Civil-War era smelting that have been identified in cores from Virginia (Balascio et al., 2019), New Jersey (Kemp et al., 2012), and Rhode Island (Lima et al., 2005).

After 1900, total Pb increased steadily to a maximum of 173 ppm that lasted from approximately 1974 to 1990 in the Lake Bonny core. Relevant potential sources of Pb in the early half of the 20th century in this setting include lead paint, lead arsenate pesticides, phosphate mine waste, and leaded gasoline. Since the early 1900s, residential and industrial structures have existed around Lake Bonny, so lead paints could have contributed to the total Pb observed. Citrus has commonly been grown in Lakeland since the early 1900s. Lead arsenate, a pesticide first applied to citrus in 1893 in Florida, could have contributed to total Pb as well (Miller et al., 1933). The application of lead arsenate on citrus was halted in 1927 but allowed once again between 1929 and 1933 in Florida (Harding, 1945; Escobar et al., 2013). Debris from phosphate mining is known to contain significant concentrations of Pb, so it is possible that dust or runoff from the mines that border Lake Bonny or the connected Lake Parker increased total Pb in Lake Bonny beginning in the early 1900s (Krekeler et al., 2008). The burning of fossil fuels, included leaded gasoline, is likely a significant source of Pb to Lake Bonny over the 20th century.

Leaded gasoline usage peaked in the US between the 1970s and mid 1980s (Nriagu, 1990). In the Lake Bonny core, there is a peak in total lead (Fig. 2) that occurs in the 36–40 cm and 44–48 cm layers, which corresponds to an age range between 1974 and 1990. This is consistent with 1974 leaded gasoline peak observed in the east coast US (Nriagu, 1990; Graney et al., 1995) as well as with other studies that measured total Pb in Florida lakes. Escobar et al. (2013) determined that the total Pb maximum occurred around 1990 in a core from Little Lake Bonnet and in 1982 in Little Lake Jackson. Schottler and Engstrom (2006) determined that the total Pb peak was located at a depth range corresponding to 1970–1990 in Lake Okeechobee. In these cases, atmospheric deposition of leaded gasoline was concluded to be the most significant source of Pb.

While regulations targeting leaded gasoline clearly reduced Pb inputs into Lake Bonny in the late 20th century, Zn concentrations remain very high in the sediments throughout the 20th and 21st centuries. Zn concentrations from the 44–48 cm layer (~1960) to the surface range from 120 to 217, which is over an order of magnitude higher than background levels at the base of the core. The Zn concentration in all sediments deposited in Lake Bonny since ~1950 are consistently above the consensus 'threshold effects level' for aquatic ecosystems of 121 ppm (Fig. 2) (MacDonald et al., 2000). The initial rise in Zn pollution observed in the sediments in early 1900s is consistent with regional pollution from coal combustion that also contributed Pb to the Lake (Sarkar et al., 2015). However, accelerated Zn deposition indicated by concentrations peaking near 200 ppm in the 1950s is almost certainly a consequence of population growth and urbanization. Zinc is widely known to be a pollution problem in urbanizing areas from tire and brake wear (Councell et al., 2004, Lopez et al., 2023), roofing materials (Chang et al., 2004), and commercial sunscreen (Chatzigianni et al., 2022). Nearly all automobile tires contain 1–2 % zinc oxide, which are introduced to soils and waters as the tires break down. Roofing shingle manufacturers have recently been adding zinc and copper to asphalt shingles because these metals kill moss and lichens, increasing shingle lifetime. The effects of the built environment surrounding Lake Bonny are recorded by sedimentary Zn.

4.3. Mass accumulation rates and population

Increases in MAR can be correlated to increases in population and the construction and development projects to accommodate population increases. The loosening of soil during construction and creation of more impervious surfaces can both increase the amount of sediment that washes into lakes and catchments. From 1900–2020, the population of Lakeland increased from 600 to 114,000 people (Fig. 5; US Census Data). MAR, while somewhat variable, appears to have three phases. There is an early stage of stability, followed by volatility, and a return to stability in recent years.

From 1903–1941, the MAR is relatively stable. However, from 1941 to 1991, the MAR is volatile and varies by almost 0.05 $g/cm^2/yr$. From 1991 to present, the MAR is mostly stable once again. While population did increase from 1903 to 1941, the population growth is more gradual than the other periods, resulting in gradual MAR increases. From 1941–1991, the population of Lakeland triples. Maps from Brenner et al., (1993) show that urban developments more than doubled from 1944 to 1975. Additionally, phosphate mining in the Lakeland area reached its climax around the early 1970s (Stewart, 1966; Robertson, 1973; Brenner et al., 1993). This rapid urbanization paired with greater phosphate mining likely explain the volatile MAR from 1941 to 1991. Although the population of Lakeland has continued increasing at a healthy rate since 1991, it is likely that the state of developments has reached a more mature state as undeveloped land becomes scarcer, resulting in a more stable MAR.

4.4. Elevated ^{226}Ra around 1960

^{226}Ra activities in the Lake Bonny Core in the 1960s are close to double the ^{226}Ra activities prior to 1940 and after 2000. Significantly elevated ^{226}Ra activities mid-core have been observed in a variety of lakes in the Lakeland area including nearby Lake Parker (Brenner et al., 1997). Increases in ^{226}Ra activities around the depth corresponding to the early 1960s could result from the deposition of sediment associated with phosphate mining. Separately or in combination with this explanation, the mixing of Lake Bonny with Floridan aquifer rich in uranium-238 (^{238}U) around the 1960s could have increased ^{226}Ra activities in the Lake Bonny as well.

In the mid-20th century, phosphate mining was prevalent around Lakeland, FL and in surrounding areas. By 1975, phosphate mines were widespread in Lakeland, FL and located adjacent to Lake Parker which is connected to Lake Bonny. It has long been recognized that uranium, the main source of radium, forms strong complexes with phosphate (e.g., Hobday and Galloway, 1999). Phosphate mining debris is known to contain ^{238}U, which generate ^{226}Ra and other radionuclides via decay (Roessler et al., 1979; Barišić et al., 1992). Brenner et al. (1997) analyzed ^{226}Ra and total phosphorous (P) in Lake Parker and found a strong correlation in ^{226}Ra activities and total P concentration and noted that the peak ^{226}Ra activity occurred between 1963 and 1979 in Lake Hollingsworth (another lake close to Lake Bonny). They argue that increased delivery of sediment containing radium and P resulted from heightened construction and phosphate mining in the Lakeland area. P concentrations in Lake Bonny correlate strongly with ^{226}Ra activities, which suggests a common source (Fig. 4). Debris from the nearby phosphate mines that entered either Lake Parker or Lake Bonny in the form of runoff or dust would have increased amounts of both ^{226}Ra and P. Due to the highly connected nature of lakes in Florida, nutrients like P and other aqueous species that entered Lake Parker would have mixed with Lake Bonny and vice versa (Schelske et al., 2005; Clift and Waters, 2024). Both the timing and increase in ^{226}Ra and P quantities agree with previous work that studied ^{226}Ra activities in Lake Parker and Lake Hollingsworth and changes in land use in Lakeland by Brenner et al. (1993) and Brenner et al. (1997).

An additional explanation for the increased ^{226}Ra activity in Lake Bonny could be due to groundwater management practices that took place during the mid-20th century in southwest Florida near Tampa. From 1961–1971 rainfall was below the average of the prior 30 years for all but 2 years in this 10-year period in southwest Florida (Stewart and Hughes, 1974). This fact, combined with construction that disrupted water recharge, meant that several lakes fell below their typical water levels. To remedy this, water from the Floridan aquifer was pumped into these lakes to maintain the lake surface water level. One of these lakes is Round Lake, 50 km west of Lake Bonny near Tampa. Brenner et al., (2000) reported that the water pumped from the Floridan aquifer contained elevated levels of ^{226}Ra compared to the surface water in Round Lake. The Floridan aquifer is in contact with the Hawthorn group, which contains carbonate-fluorapatite that contains small amounts of ^{238}U that is released upon weathering (Upchurch and Randazzo, 1997). ^{238}U that subsequently decays into ^{226}Ra resulted in elevated ^{226}Ra concentrations in Round Lake throughout the duration of groundwater augmentations, which has continued at least up until 2011 (Dimova and Burnett, 2011).

While it is unclear if similar groundwater augmentation practices occurred in Lake Bonny or the area closer to Lakeland, groundwater pumpage from the Floridan aquifer in the Lakeland region increased substantially from 1950 to 1970. This coincides with when ^{226}Ra activities rose to their highest levels. A Florida Geological Survey report (Robertson, 1973) contains annual water pumpage volumes from 4 aquifers including the Floridan aquifer (the Floridan aquifer is stated to be the major source of water in the study area) in the Lakeland ridge area of Polk County from 1950 to 1970. The water pumpage is broken down into categories such as municipal pumpage in Lakeland, total municipal pumpage (which includes the smaller cities of Bartow and Mulberry) industrial pumpage, and total pumpage across all categories. Additional Lakeland municipal water pumpage data from 1928 to 1950 was obtained from Stewart, (1966). Fig. 6 shows a comparison of the ^{226}Ra activities observed in Lake Bonny with the serac age model as well as Lakeland municipal pumpage and total pumpage. ^{226}Ra activities and aquifer pumpage rise contemporaneously. A greater reliance on water from the Floridan aquifer could have increased ^{226}Ra levels in Lake Bonny and surrounding lakes even if groundwater augmentation did not occur because water pumped from the aquifer would have inevitably made its way into surface waters (e.g. lakes) after it was used for industrial processes, irrigation, municipal use, etc. While the rises in ^{226}Ra activities and aquifer pumpage rates are offset, it is likely that inputs of sediment from phosphate mining began elevating ^{226}Ra activities prior to increases in aquifer pumpage rates.

There is a strong correlation between the increase in ^{226}Ra activity, P concentration, phosphate mining activity, and total pumpage in the Lakeland area. The expansion of phosphate mining around Lake Bonny between 1944 and 1975 likely resulted in greater inputs of phosphate mining debris that increased activities of ^{226}Ra and P concentrations. Population growth, drought conditions, and increasing water demand from industries resulted in heightened water pumpage in the 1950s-1970s in the Lakeland region. Water pumped from the Floridan aquifer that entered groundwater or drained directly into lakes could have raised ^{226}Ra activities in Lake Bonny during this time. After peak phosphate mining levels in the 1970s in the Lakeland area, the focus of phosphate mining began moving south of Lakeland (Robertson, 1973). Also, by the end of the 1970s, drought conditions had relented. The reduction in phosphate mining activity in Lakeland and cooling demands on aquifer pumping likely resulted in the decrease in ^{226}Ra activities after 1960 that continued into the 2000s.

4.5. *^{241}Am and ^{137}Cs expected and measured inventories*

One way of assessing the geochemical mobility of ^{241}Am and ^{137}Cs in Lake Bonny is to compare the total measured inventory of each radionuclide (Bq/m^2) to expected inventories from deposition during the atomic weapon test period. Expected ^{137}Cs inventories in Polk County were calculated using county-scale ^{137}Cs deposition activities (Bq/m^2) from Simon et al., (2004). The ^{137}Cs deposition for each year of the atomic weapon test year is provided, so these values can be decay corrected to the year of coring (2020) and then summed to get the total expected ^{137}Cs inventory for Polk County of 900 Bq/m^2.

The total ^{137}Cs activity we measured (413 Bq/m^2) in Lake Bonny is only half of the expected 900 Bq/m^2 accounting for decay since deposition in the 1951–1972 period. This would suggest that ^{137}Cs is being exported from the Lake Bonny system. The Hawthorn group underlying Lake Bonny contains only minor amounts of smectite or other 2:1 clays capable of binding ^{137}Cs, so it is logical that ^{137}Cs would be mobile in this environment. Additionally, due to the eutrophic state of Lake Bonny, K^+ cations likely compete for adsorption onto organic or mineral matter in the lake (Iurian et al., 2015; Yin et al., 2017). Lake Bonny is connected to Lake Parker, Saddle Creek, Lake Hancock, the Peace River, and finally the Gulf of Mexico (City of Lakeland Lakes & Stormwater, 2010). It is possible that over time Cs^+ ions have been exported from this system to downstream lakes or the Gulf of Mexico. In the nearby Lake Hollingsworth, Whitmore et al. (1996) found no discernable ^{137}Cs peak and argued that the lack of clays and continued release of ^{137}Cs from watershed vegetation resulted in a smeared ^{137}Cs profile. In Lake Okeechobee a ^{241}Am peak is clearly observable, whereas the ^{137}Cs peak has diffused substantially (Brezonik and Engstrom, 1998). Other Florida lakes often contain smeared ^{137}Cs peaks that demonstrate high geochemical mobility.

Robbins et al., (2000) quantified the activities of ^{137}Cs, ^{241}Am and several other radionuclides in the Florida Bay. They found ^{137}Cs activities of 750 Bq/m^2 (Table 1) in a core taken from an unvegetated mudbank. This value, while closer to the expected inventory of 900 Bq/m^2, is higher than the activity of ^{137}Cs found in Lake Bonny. The authors note the Florida Bay does contain small amounts of smectite that could immobilize ^{137}Cs (Manker and Griffin, 1971). It is possible that the Florida Bay has a greater ^{137}Cs activity than Lake Bonny because a much larger watershed drains into the Florida Bay compared to Lake Bonny. Dissolved Cs^+ carried away from inland Florida could accumulate in the Florida Bay over time.

In the northeastern US, chemical weathering of soils is less extensive than the south, resulting in higher amounts of clay in the A horizon of soils (Smith et al., 2014). The preservation of more 2:1 clays results in the measured ^{137}Cs inventory more closely matching the predicted ^{137}Cs inventory in sites New Hampshire and Vermont (Table 1). New Jersey has total clay weight percentages that are lower than the northeast, but

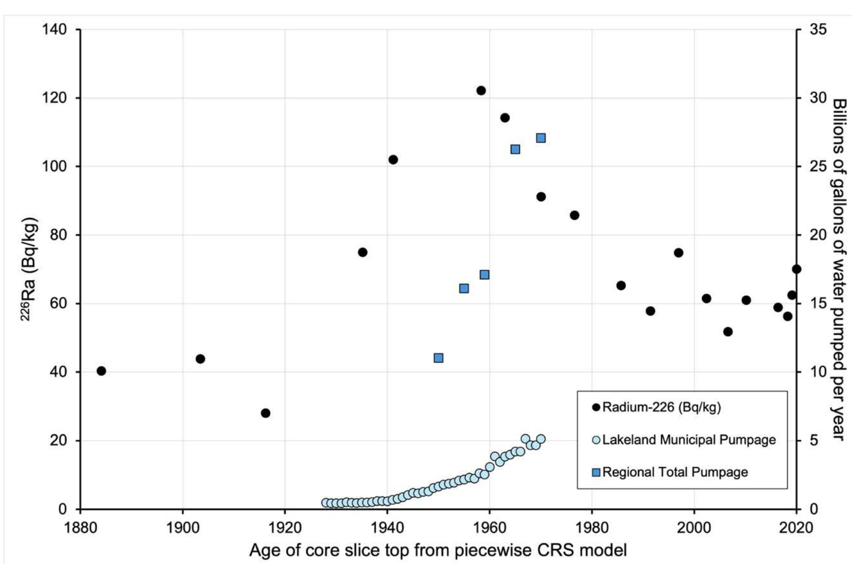

Fig. 6. Measured ^{226}Ra and historical groundwater pumpage over time via the ^{210}Pb CRS model. Municipal pumpage refers to water usage by households for daily activities. Total pumpage includes municipal pumpage and pumpage for agriculture, phosphate mining, and other industrial activities. Total pumpage and Lakeland municipal pumpage for 1950–1970 are from Robertson, (1973), whereas Lakeland municipal pumpage from 1928 to 1950 is from Stewart, (1966). The WebPlotDigiziter (Rohatgi, 2022) tool was used to convert water pumpage figures from Stewart, (1966) and Robertson, (1973) into data points.

Table 1
Compilation of measured ^{137}Cs and ^{241}Am inventories and their predicted values from sites spanning the northern to southern regions of the US east coast. Site coordinates for soil/sediment cores are provided where available. Predicted inventories were not provided in all studies. Two entries in one row indicate two cores that were taken in close proximity to one another. Note that a ^{241}Am inventory was not explicitly provided in Robbins et al. (2000) and had to be calculated. The ^{137}Cs inventory measured in Russell Bank was converted into Bq/m^2, divided by sum of the Cs/Pu ratios provided in Table 4 of Robbins et al. (2000) and then multiplied by the sum of the Am/Pu ratios also provided in Table 4 to yield a ^{241}Am inventory in Bq/m^2.

Location	^{137}Cs Measured (Predicted ^{137}Cs in Bq/m2)	^{241}Am Measured (Predicted ^{241}Am in Bq/m2)	Site Description	Reference
Camels Hump, VT (44°19'N; 72°53'W)	-	55.7	Undisturbed, acidic spodosol soils developed primarily on glacial till	Kaste et al., (2011)
Ducktrap, ME (44°16'N; 69°1'W)	-	35	Undisturbed, acidic spodosol soils developed primarily on glacial till	Kaste et al., (2011)
Moosilauke, NH (44°0'N; 71°50'W)	-	41	Undisturbed, acidic spodosol soils developed primarily on glacial till	Kaste et al., (2011)
Androscoggin River, NH	1437 (1550) 1301 (1550)	28.1 (30) 27.5 (30)	Spodosol soil derived from granite outwash till, beech and oak hardwood forest	Landis et al., (2016)
Woodstock, VT	1678 (1550) 1338 (1550)	30.9 (30) 29.9 (30)	Inceptisol soils from both a white pine dominated forest and perennial grass pasture	Landis et al., (2016)
Barnegat Bay, NJ (39°47'57.49"N; 74°06'05.92"W) and (40°01'51.80"N; 74°05'09.39"W)	594 (1800) 647 (1800)	25.7 (29) 8.6 (29)	Back-barrier lagoon-type estuary	(Boyd and Sommerfield, 2017)
Delaware Bay, DE (39°14'25.89"N; 75°06'12.80"W)	643 (1800)	15.5 (29)	Marsh separated from tidal wetlands by a narrow barrier beach in the DE Bay Estuary	(Boyd and Sommerfield, 2017)
Lake Bonny, Lakeland, FL (28°2'5.31"N; 81°55'52.24"W)	413 (900)	44.4	Shallow, eutrophic lake in an urbanized setting. Located in a karst terrain and underlain by phosphatic clayey sands	This paper
Russell Bank (Core19C) (25°02'N; 80°45'W)	750 (900)	55.6	An unvegetated mudbank in the Florida Bay	Robbins et al., (2000)

greater than Florida and the south. Despite this, measured ^{137}Cs values are about one third of the predicted inventory of 1800 Bq/m^2. The cores taken from New Jersey were taken from an estuary environment, so it is likely that lower clay compositions and competition from other dissolved cations contributed to the mobility and desorption of ^{137}Cs cations (Boyd and Sommerfield, 2017; Zucker et al., 1984; Martin et al., 1994).

While there is not a predicted ^{241}Am inventory for Lakeland or the work from Robbins et al., (2000), the measured ^{241}Am activity in the Lake Bonny core (44.4 Bq/m^2) is reasonably close to the measured activity of ^{241}Am in the Florida Bay core (55.6 Bq/m^2; Table 1). In the cores from Boyd and Sommerfield, (2017), measured ^{241}Am is close to the predicted value in one Barnegat Bay core, while only one third of the ^{137}Cs is accounted for. However, in the second Barnegat Bay core, both radionuclides are only about one third of their predicted amount. The authors did not speculate about why this is the case. Overall, ^{241}Am measured inventories more closely approximate their predicted inventory, while measured ^{137}Cs inventories more often fall short of their predicted inventories. Low clay compositions, uptake by vegetation, and competition from other cations are common causes for ^{137}Cs inventories that fall short of their predicted values.

5. Conclusions

This study demonstrates that ^{241}Am is preferable for verifying a ^{210}Pb piecewise CRS model in a eutrophic lake with low amounts of 2:1 clays. The ^{241}Am peak has a FWHM about half that of the ^{137}Cs peak. The ^{137}Cs peak is spread across two layers, while the ^{241}Am is focused in one layer. Additionally, the layers above the ^{137}Cs contain significant activities of ^{137}Cs up until the surface layers suggesting continued diffusion of ^{137}Cs over time. The total measured ^{137}Cs inventory (448 Bq/m^2) in Lake Bonny is about half of the expected inventory of 900 Bq/m^2 from historical deposition (Simon et al., 2004). Since Lake Bonny is eutrophic, it is probable that competition from K^+ cations caused ^{137}Cs cations to desorb from negatively charged sites on mineral and organic matter. These ^{137}C cations could be exported out of the Lake Bonny system to downstream water bodies including the Gulf of Mexico.

This study illustrates how radionuclides can record chemical changes in lacustrine systems due to anthropogenic activities. The atmospheric lead peak from leaded gasoline is present, although other sources of Pb such as lead paint, lead arsenate fertilizers, and phosphate mining debris likely contributed to total Pb as well. Elevated Zn levels that continue into the present are likely related to the breakdown of products containing Zn, such as tires and roofing shingles. Trends in total MAR appear to correlate with population growth and sediment inputs from phosphate mining. Elevated ^{226}Ra activities (~120 Bq/kg) in Lake Bonny around 1960 likely result from a combination of phosphate mining and water pumpage from the Floridan Aquifer, which both increased in intensity between the 1950s and 1970s evidenced by historical records of land use and aquifer pumpage in the Lakeland area (Stewart, 1966; Robertson, 1973; Brenner et al., 1993). ^{226}Ra activities have declined in Lake Bonny sediments since the 1960s and stabilized around 60 Bq/kg in the years since the peak. This is likely tied to phosphate production moving away from the study region and reduced Floridan aquifer pumping demands after drought conditions relented.

This analysis of sediments from Lake Bonny demonstrates that ^{241}Am peak is a reliable dating tool that can serve as a reference point for recent sediments and to verify ^{210}Pb dating models. In regions that possess low amounts of 2:1 clays, high rates of vegetative uptake, and competition from other cations, ^{241}Am profiles will likely yield more accurate dating models than those using ^{137}Cs profiles.

CRediT authorship contribution statement

Matt N Waters: Writing – review & editing, Resources, Methodology, Investigation, Funding acquisition, Data curation. **Troy Clift:** Writing – review & editing, Methodology, Formal analysis, Data curation. **James M Kaste:** Writing – review & editing, Validation, Software, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Paul William Volante:** Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We thank members of the South Florida Water Management District for input. This work was supported by an honors fellowship from the Charles Center at William & Mary to PWV, NSF Instrumentation & Facilities Grant 2203667 to JMK, and from NSF Grant 1941568 to MNW.

Data Availability

We shared our data through a Mendeley Data link
Revised Lake Bonny Dataset - Volante et al. (Mendeley Data)

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