1 Technical Note: Comparison of radiometric techniques for estimating

recent organic carbon sequestration rates in inland wetland soils

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- 10 **Abstract.** For wetlands to serve as natural climate solutions, accurate estimates of organic carbon (OC) sequestration rates in
- wetland sediments are needed. Dating using cesium-137 (137Cs) and lead-210 (210Pb) radioisotopes is commonly used for
- measuring OC sequestration rates in wetland sediments. ¹³⁷Cs radioisotope dating is relatively simple, with calculations
- based on a single point representing the onset (1954) or peak (1963) of the ¹³⁷Cs fallout. ²¹⁰Pb radioisotope dating is more
- 14 complex as the calculations are based on multiple points. Here, we show that reliable dating of sediment cores collected from
- wetlands can be achieved using either ¹³⁷Cs or ²¹⁰Pb dating or their combination. However, ¹³⁷Cs and ²¹⁰Pb profiles along the
- depth of sediment cores need to be screened, analyzed, and interpreted carefully to estimate OC sequestration rates with high
- precision. To this end, we propose a decision framework for screening ¹³⁷Cs and ²¹⁰Pb profiles into high- and low-quality
- sediment profiles, and we compare dating using the 1954 and 1963 time-markers, i.e., the rates of sedimentation and,
- 19 consequently, OC sequestration over the past ~60 years. Our findings suggest that ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration
- 20 rates are comparable, especially when using the 1963 (vs. 1954) time-marker.

1 Introduction

- Wetlands in agricultural landscapes serve a crucial role in providing habitat for wildlife, regulating climate, improving water
- 23 quality, and reducing floods. Moreover, these wetlands have the potential to sequester organic carbon (OC) (Bridgham et al.,
- 24 2006; Nahlik and Fennessey, 2016; Bansal et al., 2023). Accounting for the balance between the sequestration and emission
- 25 of carbon can help establish wetlands as essential candidates for natural climate solutions by offsetting carbon emissions
- 26 (Hambäck et al., 2023). These wetlands embedded in agricultural landscapes are recognized as temperate inland wetland
- soils. The global carbon stock of temperate inland wetland soils is estimated to be 46 Pg C to 2 m depth, and Canada's
- temperate inland wetland soils are estimated to contain 4.6 Pg C (Bridgham et al., 2006). Compared to peatlands, the rapid

rate of OC sequestration and the more considerable spatial extent of temperate inland wetland soils can help contribute significantly to regional or national carbon sequestration (Bridgham et al., 2006; Nahlik and Fennessey, 2016).

Canada encompasses around 25% of the world's wetlands, with an area of approximately 1.29 million square kilometers, which accounts for 13% of the country's terrestrial area (Environment and Climate Change Canada, 2016), highlighting the global importance of these wetlands. Unfortunately, there is minimal data on the OC sequestration rates in these wetlands. To estimate the OC sequestration potential of these wetlands, it is essential to establish precise measurements to quantify wetland OC sequestration, develop strategies to promote conservation and restoration efforts, incorporate carbon credits in the carbon markets, and validate the wetland-based ecosystem services.

There are several ways to estimate the potential of wetlands to store OC (Bansal et al., 2023). One of these methods is radiometric dating, which can calculate the OC storage rates of wetlands over periods of 10 to ≥ 1,000 years. Frequently used radioisotopes for radiometric dating are cesium-137 (¹³⁷Cs) and lead-210 (²¹⁰Pb), which can be used to estimate relatively recent (up to the last 100 years) OC sequestration rates (Villa and Bernal, 2018). Estimating OC sequestration rates involves building an age-depth profile or model of ¹³⁷Cs and ²¹⁰Pb from sediment cores that demonstrate the relationship between the depth of sediment layers and their corresponding age. Since the inorganic radioisotopes (¹³⁷Cs and ²¹⁰Pb) strongly bind with the soil particles once in contact, the radioisotopes can act as an efficient tracer for investigating OC sequestration rates (Ritchie and McHenry, 1990; Craft and Casey, 2000). These characteristics allow for accurate tracking of carbon movement within ecosystems, thereby enabling the extraction of detailed information about carbon sequestration dynamics in wetlands.

The characteristics of ¹³⁷Cs and ²¹⁰Pb to estimate wetland OC sequestration rates are presented in Table 1. ¹³⁷Cs is an artificial radioisotope that was produced during thermonuclear bomb testing in the 1950s and 1960s, with the onset of atmospheric deposition in 1954 and a peak in 1963 (Ritchie and McHenry, 1990). The testing caused radioactive uranium to decay, and, as a result, ¹³⁷Cs isotope was released into the atmosphere, which was then deposited around the globe. Although there may be challenges in applying our study to some parts of the world, the information is generally applicable and valuable for consideration in all regions. We encourage others to customize this approach further for use in other regions where Cs deposition histories vary.

 ¹³⁷Cs has a half-life of 30.17 years, which can be used to estimate the last ~50-70 years of OC sequestration rates in wetlands (e.g., Bernal and Mitsch, 2012). ¹³⁷Cs dating assumes constant sedimentation rates measured since 1954 or 1963. In using the two time-markers for ¹³⁷Cs, we do not expect the sedimentation rates to be equal, but we do expect them to be similar. The onset and the peak of ¹³⁷Cs activity at 661.6 keV can be used to mark 1954 and 1963, respectively. These time-markers

(1954 and 1963) can date sediment layers (Pennington et al., 1973; Ritchie and McHenry, 1990; DeLaune et al., 2003) and consequently the OC sequestration rates. ¹³⁷Cs has an additional time-marker for Europe in 1986 due to the Chernobyl nuclear accident and for Japan in 2011 due to the Fukushima Daiichi nuclear accident (Foucher et al., 2021), indicating that OC sequestration estimates can be derived for different timescales. In the Americas, we do not see evidence of the 1986 or 2011 ¹³⁷Cs peak, which is observed in Europe and Japan, respectively, so we did not need to use other radioisotope techniques (e.g., ²³⁹⁺²⁴⁰Pu) to distinguish the 1986 or 2011 ¹³⁷Cs peak from the 1963 ¹³⁷Cs peak. ¹³⁷Cs dating requires a gamma spectrometer to estimate OC sequestration rates. Sample preparation for gamma analysis involves drying, weighing, disaggregating, homogenizing, and sieving (Bansal et al., 2023). Samples vary from 1 to 1,500 g, with smaller samples associated with higher uncertainties and, therefore, requiring longer times to analyze. Gamma analysis counting times range from 4 to 48 h for each sample (e.g., 4 to 12 h in Li et al., 2007; 12 to 24 h in Zarrinabadi et al., 2023; and 24 to 48 h in Kamula et al., 2017). ¹³⁷Cs dating provides a simple result (an average sedimentation rate), while ²¹⁰Pb dating provides a more complex result (using a supply rate model to reveal trends in sedimentation rates). Plutonium (Pu) may replace ¹³⁷Cs in the future due to concerns of half-life and persistence as a dating tool. In essence, ²³⁹⁺²⁴⁰Pu has the same source and deposition mechanism as ¹³⁷Cs. Its longer half-life will make its peak measurable when ¹³⁷Cs is no longer measurable.

Unlike ¹³⁷Cs, ²¹⁰Pb is a naturally occurring radionuclide derived from ²³⁸U and deposits atmospherically from the decay of radium-226 (²²⁶Ra) (Walling and He, 1999). ²¹⁰Pb has a half-life of 22.3 years and is used to estimate the last 10-150 years of OC sequestration rates in wetlands (Craft and Richardson, 1998; Craft and Casey, 2000; Craft et al., 2018; Creed et al., 2022). ²¹⁰Pb activity can be measured using gamma (observed at 46.5 keV) and alpha spectrometry (destructive) (Walling and He, 1999; Bellucci et al., 2007). Traditional alpha analysis requires 0.2-0.5 g of sample and additional sample preparation involving leaching with hydrochloric and nitric acid and electroplating (up to 24 h for sample preparation) (Bansal et al., 2023). Alpha analysis can be considered an indirect method for ²¹⁰Pb dating where polonium-210 (²¹⁰Po) activity is measured, assuming both ²¹⁰Pb and ²¹⁰Po are in a secular equilibrium. ²¹⁰Pb activity is calculated by comparing ²¹⁰Po activity against the known activity of ²⁰⁹Po (isotope tracer). In alpha analysis, the additional time required for sample preparation is compensated by running multiple samples simultaneously (Bansal et al., 2023). Gamma and alpha spectrometry of ²¹⁰Pb provides the total ²¹⁰Pb activity, which incorporates unsupported (or excess) ²¹⁰Pb (²¹⁰Pb_{ex}) and supported ²¹⁰Pb. ²¹⁰Pb_{ex} is used to determine the mass or sediment accumulation rate. Supported ²¹⁰Pb is derived from the natural decay of ²²⁶Ra in the sediment, while unsupported ²¹⁰Pb comes from the decay of atmospheric radon-222 (²²²Rn), which deposits ²¹⁰Pb onto the sediment surface from the air. Unsupported ²¹⁰Pb activity decreases over time due to radioactive decay, unlike supported ²¹⁰Pb (Appleby and Oldfieldz, 1983). The choice of model used in ²¹⁰Pb dating can reflect constant and variable sedimentation rates (Sanchez-Cabeza and Ruiz-Fernandez, 2012) and, consequently, OC sequestration rates in wetlands. Some models used for ²¹⁰Pb dating are (1) constant flux-constant sedimentation (CFCS)

model, (2) constant rate of supply (CRS) model, and (3) constant initial concentration (CIC) model (Appleby and Oldfield, 1978). Both ¹³⁷Cs and ²¹⁰Pb provide suitable time-markers and a longer time horizon compared to direct measurements using the time-marker of horizons (2-10 years) to study sediment accretion and, subsequently, OC sequestration rates in wetlands (Bernal and Mitsch, 2013; Villa and Bernal, 2018). In this study, we compared the average OC sequestration rate derived from ¹³⁷Cs temporal markers with the progressive OC sequestration rates derived using a constant rate of supply model applied to ²¹⁰Pb.

Table 1: Characteristics of ¹³⁷Cs and unsupported ²¹⁰Pb (²¹⁰Pb_{ex}) dating to estimate sedimentation rates in wetlands.

Method of radiometric dating	¹³⁷ Cs	$^{210}\mathrm{Pb}_{\mathrm{ex}}$
Type of radioisotope	Artificial (atmospheric deposition 1954 – 1963).	Natural.
Half life	30.17 years.	22.3 years.
Time-marker	1954 (onset) and 1963 (peak).	Recent (10-20 years) to a maximum of 50-150 years.
Radiometric Technique	Gamma spectrometry (nondestructive).	Gamma (nondestructive) and alpha spectrometry (destructive).
Pre-processing	Drying, weighing, disaggregating, homogenizing, and sieving.	For gamma analysis, drying, weighing, disaggregating, homogenizing, and sieving prior to analysis on a gamma counter.
		For alpha analysis, leaching with hydrochloric and nitric acid and electroplating of ²¹⁰ Po which constitutes allowing the digested and therefore extracted ²¹⁰ Po isotope solution to settle on silver coins overnight before measuring the ²¹⁰ Po (known tracer) and ²¹⁰ Po activity (sample) next morning through the alpha counter/ensemble.
Sample size	Minimum 1 g (larger sample size has higher certainty).	1 to 5 g for gamma spectrometry, 0.2 to 0.5 g for alpha spectrometry.
Time requirement for	48 h for each sample for gamma spectrometry.	48 h for each sample for gamma spectrometry.
radiometric dating		48 to 72 h for multiple samples plus sample preparation time per multiple samples.
Output	A single average sedimentation rate.	Variable sedimentation rate.
Estimation approach	Onset of ¹³⁷ Cs activity represents 1954 and highest peak of ¹³⁷ Cs activity represents 1963, observed at 661.6 keV.	Activity of ²¹⁰ Pb is observed at 46.5 keV. Excess ²¹⁰ Pb is used to determine the vertical accretion.
Complexity in estimation	Simple; estimated by using time-marker of onset or peak ¹³⁷ Cs activity and associated sediment accumulation.	More complex; estimated by one of several models to estimate sedimentation rate. Most common models are (1) constant flux—constant sedimentation model, (2) constant rate of supply model, and (3) constant initial concentration model (Appleby and Oldfield, 1978)

The combined use of ¹³⁷Cs and ²¹⁰Pb may improve the accuracy of the dating estimation (Drexler et al., 2018; Creed et al., 2022). The more detailed assessment accrues a higher cost and time requirement, and the need for specialized equipment and technical expertise to conduct laboratory and data analyses may constrain the research efforts (Bansal et al., 2023). Furthermore, factors such as timescales, analytical complexity in interpreting radioisotope profiles (e.g., ¹³⁷Cs peak clarity), variability in atmospheric deposition, and mobilization of radioisotopes can contribute to uncertainty (Drexler et al., 2018; Loder and Finkelstein, 2020; Zhang et al., 2021; Bansal et al., 2023) and limit the applicability of one radioisotope over the

other. Therefore, it is essential to consider the advantages and potential challenges of using radioisotopes before designing research studies.

The main objective of this research paper is to explore the use of ¹³⁷Cs and ²¹⁰Pb to estimate recent OC sequestration rates in undisturbed (i.e., not directly impaired by human activities) temperate inland wetland soils located on agricultural landscapes. Here, we aim to (1) categorize ¹³⁷Cs or ²¹⁰Pb profiles into high- and low-quality via a decision framework, (2) apply the decision framework to estimate OC sequestration rates, (3) use 1963 and 1954 time-markers to compare the ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration rates to get a better understanding of the sediment history, and (4) select the best approach for ¹³⁷Cs and ²¹⁰Pb to estimate the OC sequestration rates with highest precision. This study helps reduce uncertainty in studies that rely on ¹³⁷Cs or ²¹⁰Pb radioisotope dating.

2 Methods

2.1 Sediment core collection

Triplicate sediment cores were collected from 30 undisturbed temperate inland wetland soils in agricultural landscapes across southern Canada (Fig. 1). A summary of the physical characteristics of these wetlands can be found in Supplementary Table 1. These wetlands were undisturbed, with no known history of cultivation. The sediment cores were extracted from the center of the wetland, constituting the open-water area. A Watermark Universal Corer (inner diameter of 6.8 cm) or VibeCore Mini with poly core tubes (inner diameter of 7.6 cm) were used to collect most of the sediment cores. A JMP BackSaver Soil sampler (inner diameter of 3.8 cm) was used for compacted sediment cores. Shallow (15 to 90 cm) sediment cores were sectioned into 1- or 2-cm increments. Deeper (> 90 cm) sediment cores were sectioned into 5-cm increments. The sediment cores were stored at -5 °C for further processing at the laboratory.

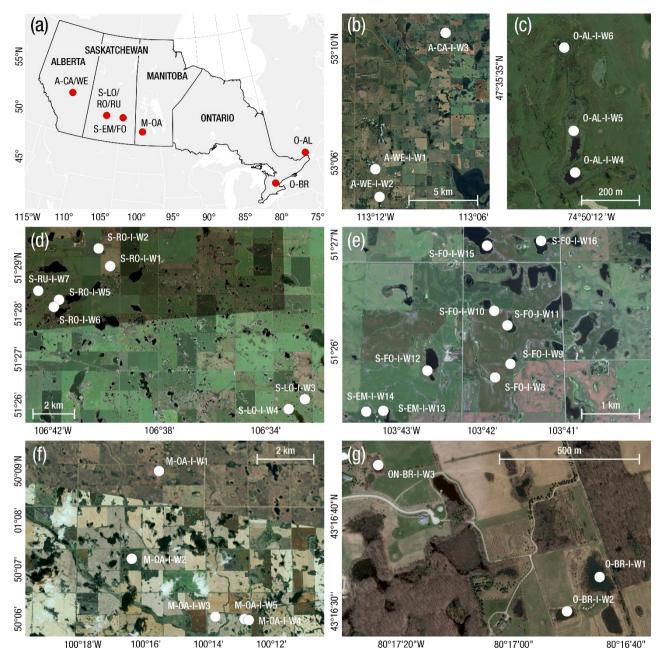


Figure 1: (a) Study area situated in four provinces of Canada; (b) three wetland sites in Alberta (AB); (c) three wetland sites in Ontario (ON); (d) seven wetland sites in Saskatchewan (SK); (e) nine wetland sites in Saskatchewan (SK); (f) five wetland sites in Manitoba (MB); and (g) three wetland sites in Ontario (ON). Figures (b)-(g) are based on the sampling locations of wetlands used in this study reproduced using Google Earth Images [(b) and (c) ©2024 Airbus; (d), (e), and (g) ©2024 Maxar Technologies; (f) ©2024 Airbus and Maxar Technologies].

2.2 Generation of ¹³⁷Cs and ²¹⁰Pb profiles

Sediment core increments were weighed (wet mass), dried, weighed again (dry mass), disaggregated, homogenized, and sieved. The increments were sieved to remove gravel (> 2 mm); radioisotopes do not bind on the gravel, and gravel does not contain OC; therefore, eliminating gravel improves the estimate of radioisotopes and OC. The increments were counted at 661.6 keV for ¹³⁷Cs activity and 46.5 keV for ²¹⁰Pb activity. ¹³⁷Cs analysis was performed using a gamma spectrometer, and ²¹⁰Pb analysis was performed using both gamma and alpha spectrometers to increase throughput rates. The gamma analysis was conducted using the high-purity germanium detectors; e.g., Broad Energy Germanium detectors (BE6530) and Small Anode Germanium well detectors (GSW275L) (Mirion Technologies, Inc., Atlanta, GA, USA). The alpha analysis was conducted using ORTEC® alpha spectrometer (AMETEK® Advanced Measurement Technology, TN, USA). Both radioisotope analyses were performed at the Landscape Dynamics Laboratory, University of Manitoba, Winnipeg, Canada. Although the underlying principles of gamma and alpha analysis differ, each focuses on quantifying the decay of ²¹⁰Pb, generating comparable results (Zaborska et al., 2007). Measurement accuracy of gamma detectors is ensured by assessing the counting errors with reference materials within the same geometry as the sample (e.g., petri dish). Detection error was < 10% with a counting time of up to 24 h. Furthermore, Landscape Dynamics Laboratory undergoes regular Proficiency Testing through the International Atomic Reference Material Agency (IARMA) and previously through the International Atomic Energy Agency (IAEA) to ensure acceptable accuracy and precision of analytical results using gamma spectroscopy.

2.3 Screening of ¹³⁷Cs and ²¹⁰Pb profiles

Sediment cores were screened to remove profiles with evidence of vertical mixing, and then the remaining profiles were used to estimate OC sequestration rates using ¹³⁷Cs or ²¹⁰Pb radioisotope dating. The actual ¹³⁷Cs peak can vary from the expected peak, increasing uncertainty in ¹³⁷Cs dating (Drexler et al., 2018). ¹³⁷Cs peaks can be "noisy" or "disturbed"; i.e., flattened, broadened, truncated, mixed, fluctuating (Drexler et al., 2018), or one-sided where the ¹³⁷Cs peaks appear at the surface of the sediment core (indicating no or little sedimentation since 1963). The magnitude and shape of the ¹³⁷Cs peaks observed in the sediments can be affected by the atmospheric deposition rate of ¹³⁷Cs, which is obviously affected by the number and magnitude of emission events and the weather conditions following these events (UNSCEAR, 2000). The magnitude and shape of these peaks are also impacted by the movement of water and sediment within each wetland's catchment during the peaks' development (Milan et al., 1995; Zarrinabadi et al., 2023). Here, changes in the shape of the peaks are caused by the upward and downward movement of the sediment within the sediment profile (the movement of ¹³⁷Cs through diffusion (Klaminder et al., 2012) is presumed negligible). Bioturbation can cause an upward and downward mixing of the ¹³⁷Cs in the profile, resulting in peak attenuation (Robbins et al., 1977). Even wave action during the period of atmospheric deposition will have a similar attenuation effect (Andersen et al., 2000; Zarrinabadi et al., 2023). Following peak atmospheric deposition, soil erosion and the accumulation of sediment will deliver sediments to the top of the profile, and those sediments

may be higher or lower in concentration depending on the degree of preferential sediment transport and the associated enrichment or depletion of ¹³⁷Cs in the added sediment (Zarrinabadi et al., 2023). Such noise in ¹³⁷Cs peaks needs careful interpretation to avoid over- or under-estimating the OC sequestration rates.

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Selecting suitable cores: Of the 90 sediment cores (30 wetlands x 3 replicates = 90), 79 were suitable (complete and datable) for ¹³⁷Cs dating and 47 for ²¹⁰Pb dating. Only some replicates from the same wetland were ideal for interpretation or further screening. The suitability of ¹³⁷Cs profiles for dating was assessed by zero activity before the onset and peak of ¹³⁷Cs activity. The suitability of ²¹⁰Pb profiles for dating was evaluated by determining the exponential decline in ²¹⁰Pb activity with depth until background levels are reached.

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Classification of the selected ¹³⁷Cs profiles: The 79 suitable ¹³⁷Cs profiles were then classified into high- and low-quality using the following steps (Fig. 2a):

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- 1. The ¹³⁷C depth profile and the shape of the peak were assessed. A clear and distinct peak associated with several points on both sides of the peak verified the ¹³⁷Cs depth profile as high-quality (e.g., Fig. 3a).
- 2. When analyzing sediment samples, a clear peak in ¹³⁷Cs activity didn't always exist (e.g. Fig 4a). If a peak was absent, which could have resulted from sediment influxes with very high or very low ¹³⁷Cs activity levels, the total 137 Cs activity of the entire profile was examined. If the cumulative 137 Cs inventory value for the entire profile was \geq 500 Bq m⁻², then the ¹³⁷Cs profile was considered high-quality. Conversely, if the cumulative ¹³⁷Cs inventory value for the entire profile was < 500 Bq m⁻², the ¹³⁷Cs profile was considered low-quality. The cutoff cumulative ¹³⁷Cs inventory value of 500 Bq m⁻² was established by assessing the ¹³⁷Cs reference inventory value, the value of ¹³⁷Cs present in a non-eroded system with an undisturbed profile. The ¹³⁷Cs reference inventory value differs from region to region (Owens and Walling, 1996), and the most proximal regional value was used to select the cutoff ¹³⁷Cs inventory value (Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al., 2023). The ¹³⁷Cs reference inventory is a catchment-wide reference value and not specific to the wetland center; thus, the cumulative ¹³⁷Cs inventory value of 500 Bg m⁻² was viewed as a conservative indicator of the suitability of the ¹³⁷Cs profiles. Ideally, reference sites are large, open, level, non-eroded areas, usually in forage or grassland since the 1950s, and within 10 km of the site of interest. In this study, it was impossible to identify a suitable reference site near every wetland; it is usually difficult to find reference sites in agricultural landscapes. However, we could locate reference sites used in other studies within 50 km except from nine wetlands in SK (51° N and 104° W), which were ~150 km from the reference site. Although this was not considered ideal, it was considered acceptable.

Classification of the selected ²¹⁰Pb profiles: The 47 suitable ²¹⁰Pb profiles were classified into high- and low-quality profiles based on the following steps (Fig. 2b):

- 1. ²¹⁰Pb activity were plotted with a log-transformed ²¹⁰Pb_{ex} against mass depth (g cm⁻²).
- 2. A linear regression analysis was performed (where the slope is used to derive the mass or sediment accumulation rate in g cm⁻² yr⁻¹).
 - 3. If the linear regression passed both normality and constant variance tests and had an $R^2 > 0.5$ and a p-value < 0.05, then the 210 Pb profile was classified as high-quality (e.g., Fig. 3a).
 - 4. If either normality and constant variance tests were not passed, with an $R^2 \le 0.5$ or a p-value ≥ 0.05 , then the ^{210}Pb profile was considered low-quality (e.g., Fig. 3b).

The ^{210}Pb profiles were also classified using a two-step piecewise linear regression model to capture recent shifts in OC sequestration rates. However, no significant improvement was observed. Consequently, ^{210}Pb -based OC sequestration rates were derived from the linear regression line. An $R^2 > 0.5$ was selected as the cut-off for selecting high-quality over low-quality profiles. Increasing the cut-off R^2 value may produce better profiles to be chosen for the study. Still, it can reduce the number of available sediment cores and potentially ignore the natural variability and significant events occurring in the real environment.

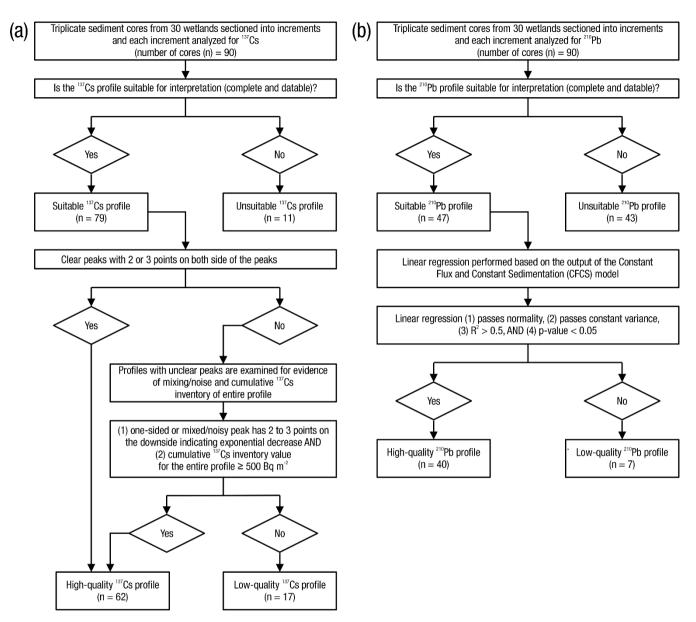


Figure 2: Classification of high- and low-quality ¹³⁷Cs and ²¹⁰Pb profiles outlining the decision frameworks for screening (a) ¹³⁷Cs and (b) ²¹⁰Pb profiles.

217 2.4 Organic carbon stocks and sequestration rates 218 Radioisotope activity measurements were utilized to assign two time-markers, one for 1954 and the other for 1963, in the 219 sediment cores. Sediment radioisotope dating was used to calculate the rates of sediment or mass accumulation and OC 220 sequestration. 221 For ¹³⁷Cs dating, sediment accumulation and OC sequestration rates (Mg ha⁻¹ yr⁻¹) were estimated using the cumulative sum 222 of sediment or OC (Mg ha⁻¹) from the surface to the depth corresponding to the time-markers of ¹³⁷Cs of each core and 223 224 dividing by the number of years from the time-marker to the years the samples were collected. Unit conversion is applied to 225 report the OC sequestration rate estimates in Mg ha⁻¹ yr⁻¹ from g cm⁻² yr⁻¹ for easy standardization and comparability with other studies. For ¹³⁷Cs profiles with noisy peaks and comparatively larger cumulative ¹³⁷Cs inventory values, the first 226 elongated peak with a sharp rise after the onset of the ¹³⁷Cs peak was considered the 1963 peak instead of the peak with the 227 228 highest activity in the profile. 229 230 For ²¹⁰Pb dating, mass or sediment accumulation and OC sequestration rates were estimated using the Constant Flux and 231 Constant Sedimentation (CFCS) model (Sanchez-Cabeza and Ruiz-Fernandez, 2012; Kamula et al., 2017), Here, ²¹⁰Pb_{ex} was estimated by subtracting ²²⁶Ra activity (186 keV) from the total ²¹⁰Pb activity. The CFCS model uses the log-linear 232 relationship of ²¹⁰Pb_{ex} with mass depth and converts ²¹⁰Pb_{ex} to the mass or sediment accumulation rate and, consequently, the 233 234 OC sequestration rate. The OC stock was estimated by taking the cumulative sum of OC (Mg ha⁻¹) from the surface of each 235 sediment core to the depth increments represented by the time-marker (e.g., 1963). 236 237 OC stocks for the 1954 and 1963 time-markers were calculated by multiplying the OC content per unit mass of soil (g). 238 Here, OC content was calculated from OC concentration (%) measured by loss-on-ignition (LOI) method (Kolthoff and 239 Sandell, 1952; Dean, 1974) by the mass of sediment for each section interval and specific depth interval per unit area (g cm 240 2) down the profile to the respective time-marker. OC (%) was calculated by multiplying organic matter (%) by LOI with

over-estimation of OC content.

Statistical analysis

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- 246 Statistical analyses used sediment cores with ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration rates available (number of sediment
- 247 cores (n) = 44). The 137 Cs—and 210 Pb-based estimates of OC sequestration rates were compared using a quantile-quantile (Q-

0.58, assuming 58% of the organic matter is carbon. Despite the broad applicability, simplicity in measurement techniques,

and cost-effectiveness, the LOI approach is associated with some limitations, such as the ignition of non-organic particles at

high temperatures or the use of a conventional conversion factor (Pribyl, 2010; Hoogsteen et al., 2015), which can result in

Q) plot. First, the comparison was done via assessment of the Q–Q plots. Four sample datasets were used to construct Q-Q plots to compare the distribution of ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration against the 1:1 line.

The sample datasets included:

- D1, all suitable 137 Cs and 210 Pb profiles with OC sequestration rates estimated since 1954 (n = 44).
- D2, all suitable 137 Cs and 210 Pb profiles with OC sequestration rates estimated since 1963 (n = 44).
 - D3, high-quality 137 Cs and 210 Pb profiles with OC sequestration rates estimated since 1954 (n = 30).
 - D4, high-quality 137 Cs and 210 Pb profiles with OC sequestration rates estimated since 1963 (n = 30).

A Q-Q plot was calculated for each dataset. The x- and y- coordinates of a point in a Q-Q plot corresponded to the path percentiles of the two OC sequestration rate estimates being compared in the plot. Here, p = (k - 0.5) / n, where n is the sample size and k = 1, ..., n (Jain et al., 2007). The distribution of the points on the Q-Q plot was compared against the y = x (1:1) line to assess whether the two OC sequestration rate estimates are similar. If the points were distributed in a straight line and close to a 1:1 line, then it suggested that the two estimates came from the same distribution. In contrast, if the points were not distributed in a straight line or deviated from the 1:1 line, then it suggested that the two estimates did not come from the same distribution. The Q-Q plots were generated in Microsoft Excel (Microsoft 365, Version 2402, Redmond, WA).

Since interpreting the Q-Q plot through a visual inspection can be subjective to human perception, we compared the ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration rate estimates using a distance sampling model. A distance sampling model captures how the detectability of objects from the observer (walking along a straight line) decreases with the increase in the object-to-observer distance. If the objects are closely distributed along the observer's path (i.e., if points of the Q-Q plot were closely distributed along the 1:1 line), then the distribution of the distances is expected to be a half normal distribution. The Cramervon Mises test was used to estimate whether the distances (q1, q2, ..., qn) from the points to the 1:1 line were from a half-normal distribution. Given a set of distance samples (q1, q2, ..., qn) and a detection function, the Cramer-von Mises test builds a model that fits the distance sampling data to the detection function (for details on modelling, see Miller et al., 2019). A half-normal key is commonly used as a detection function, corresponding to a half-normal distribution's shape.

The Cramer-von Mises test produced a p-value and Akaike's Information Criterion (AIC) as its test statistic. A p-value larger than the significant level (p = 0.05) indicated that the likelihood of points being observed closer to the 1:1 line is high and

that the probability decreases as the distances increase. This provided evidence of the points being closely distributed along the 1:1 line. The AIC was used to rank the distance sampling models, which are built by the Cramer-von Mises test, from best to worst (e.g., Burnham and Anderson, 2003); a small AIC value indicates a good fit to the half-normal key and thus provides evidence that the points are close to the 1:1 line (Miller et al., 2019). The distance sampling Cramer-von Mises test was computed using the "distance" package in R version 4.0.3 (Miller and Clark-Wolfe, 2023; R Core Team, 2023).

3 Results

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3.1 High- and low-quality ¹³⁷Cs and ²¹⁰Pb profiles

Of the 79 suitable ¹³⁷Cs profiles, 62 (78%) were classified as high-quality. Of the 62 high-quality ¹³⁷Cs profiles, 61% had clear and distinct peaks, with a smooth rise and decline. In contrast, the remaining 39% had noise—either one-sided peaks or disturbed peaks (e.g., Fig. 4). Of the 62 high-quality ¹³⁷Cs profiles, 4 (6.5%) were repositioned to capture the ¹³⁷Cs enriched sediments post 1963 (e.g., ¹³⁷Cs profile of S-LO-I-W4-T2-CW-R2 in Supplementary Fig. 2a). In these profiles, which had a cumulative ¹³⁷Cs inventory value > 1,200 Bg m⁻², the depth that corresponded to ¹³⁷Cs cumulative inventory value of ~500 Bq m⁻² was considered as the 1963 time-marker. The high total quantities of ¹³⁷Cs profile inventories can be attributed to receiving ¹³⁷Cs enriched sediments from the surrounding landscape. Sediments that have undergone substantial preferential detachment and entrainment on their pathway into a wetland can have very high concentrations of ¹³⁷Cs and, when interlayered with sediments that are not so enriched, can generate multiple ¹³⁷Cs peaks in the sediment profile peaks after 1963. These observed multiple peaks are local and not regional, ruling out the association with Chernobyl and Fukushima events. Two ¹³⁷Cs profiles were considered high-quality despite a cumulative ¹³⁷Cs inventory value < 500 Bg m⁻² because the 1963 peak was clear, distinct, and elongated with two-to-three points on both sides of the peak (e.g., ¹³⁷Cs profile of M-OA-I-W4-T2-CW-R2 in Supplementary Fig. 7b). One ¹³⁷Cs profile was considered high-quality despite showing marginal quality to the set criteria in the decision framework, where the peak profile had good shape with several points on the downside of the peak and one point on the other side and had a cumulative ¹³⁷Cs inventory value of 499 Bq m⁻². One ¹³⁷Cs profile was classified as low-quality despite a cumulative ¹³⁷Cs inventory value > 500 Bq m⁻² because the peak was highly fluctuating and not discernible (e.g., ¹³⁷Cs profile of O-AL-I-W6-T1-CW-R1 in Supplementary Fig. 12b).

Of the 47 ²¹⁰Pb profiles, 40 (85%) were classified as high-quality.

There were 44 sediment cores with both ¹³⁷Cs and ²¹⁰Pb suitable profiles available. Of these, 30 were categorized as high-quality ¹³⁷Cs and high-quality ²¹⁰Pb (Fig. 3a), six were categorized as high-quality ¹³⁷Cs and low-quality ²¹⁰Pb (Fig. 3b),

seven were classified as low-quality ¹³⁷Cs and high-quality ²¹⁰Pb (Fig. 4a), and one was categorized as low-quality ¹³⁷Cs and low-quality ²¹⁰Pb (Fig. 4b). (See Supplementary Figs. 1 to 12 for ¹³⁷Cs and ²¹⁰Pb profiles in all study wetlands.)

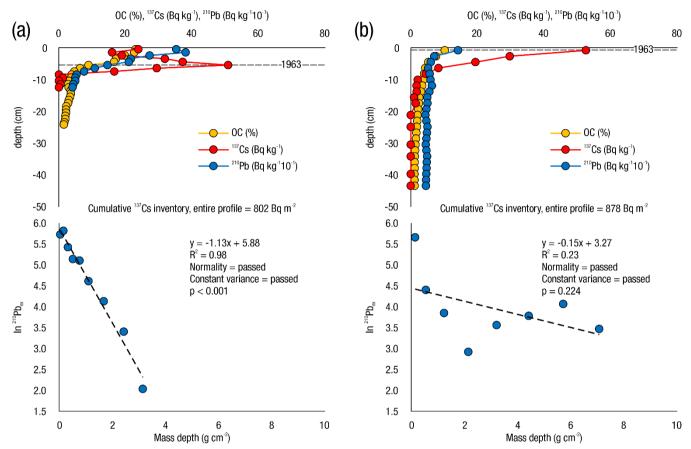


Figure 3: Examples of ¹³⁷Cs and ²¹⁰Pb classifications showing OC (%), ¹³⁷Cs (Bq kg⁻¹), and ²¹⁰Pb (Bq kg⁻¹) depth profiles and plots of log-transformed ²¹⁰Pb_{ex} against mass depth (g cm⁻²): (a) high-quality ¹³⁷Cs and high-quality ²¹⁰Pb (S-LO-I-W3-T1-CW-R1); (b) high-quality ¹³⁷Cs and low-quality ²¹⁰Pb (M-OA-I-W4-T3-CW-R3).

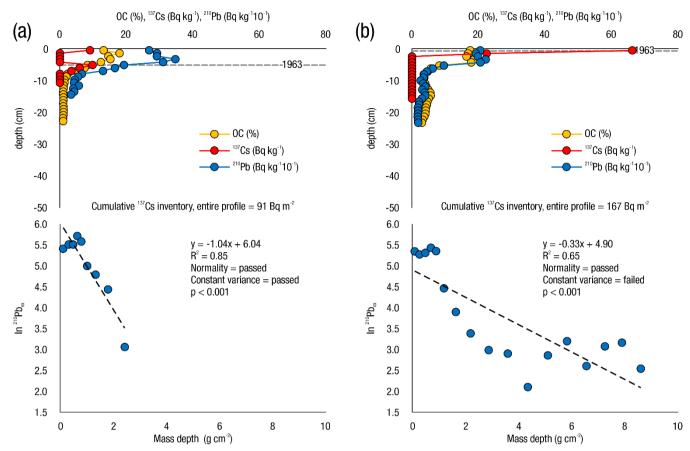


Figure 4: Examples of ¹³⁷Cs and ²¹⁰Pb classifications showing OC (%), ¹³⁷Cs (Bq kg⁻¹), and ²¹⁰Pb (Bq kg⁻¹) depth profiles and plots of log-transformed ²¹⁰Pb_{ex} against mass depth (g cm⁻²): (a) low-quality ¹³⁷Cs and high-quality ²¹⁰Pb (S-RO-I-W1-T3-CW-R3); and (b) low-quality ¹³⁷Cs and low-quality ²¹⁰Pb (S-RO-I-W6-T1-CW-R1).

3.2 137Cs vs. 210Pb derived organic carbon sequestration rates

For each of the four datasets (D1-D4), the points on the Q-Q plot were distributed in a straight line, showing a linear relationship between the two estimates being compared ($R^2 > 0.95$, p-value < 0.001) (Fig. 5).

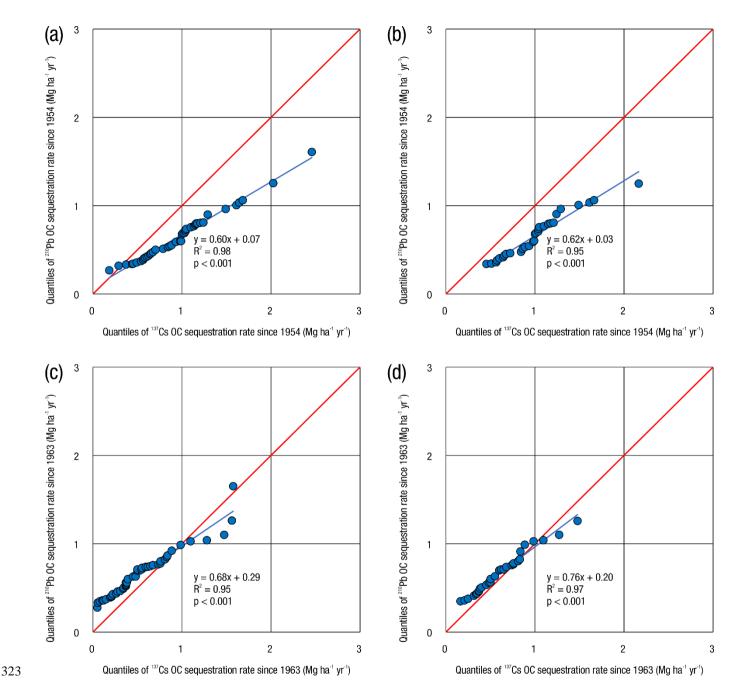


Figure 5: Q-Q plot of ¹³⁷Cs- vs. ²¹⁰Pb-based organic carbon (OC) sequestration rates using (a) all suitable ¹³⁷Cs and ²¹⁰Pb profiles estimated since 1954 (D1), (b) high-quality ¹³⁷Cs and ²¹⁰Pb profiles estimated since 1954 (D3), (c) all suitable ¹³⁷Cs and ²¹⁰Pb profiles estimated since 1963 (D2), and (d) high-quality ¹³⁷Cs and high-quality ²¹⁰Pb profiles estimated since 1963 (D4).

Visual inspection of the Q-Q plots showed that the points for D2 (i.e., all suitable ¹³⁷Cs and ²¹⁰Pb profiles using the 1963 time-marker; Fig. 5c) and D4 (i.e., high-quality ¹³⁷Cs and ²¹⁰Pb profiles using the 1963 time-marker; Fig. 5d) were distributed more closely along the 1:1 line compared to that of D1 (i.e., all suitable ¹³⁷Cs and ²¹⁰Pb profiles using the 1954 time-marker; Fig. 5a) and D3 (i.e., high-quality ¹³⁷Cs and ²¹⁰Pb profiles using the 1954 time-marker; Fig. 5b).

An intercept closer to 0 and a slope closer to 1 indicated good alignment of the regression line to the 1:1 line. The slope (s) and intercept (i) of the regression lines were: s = 0.60, i = 0.07 for D1 (Fig. 5a); s = 0.62, i = 0.03 for D3 (Fig. 5b); s = 0.68, i = 0.29 for D2 (Fig. 5c); and s = 0.76, i = 0.20 for D4 (Fig. 5d). D2 and D4 had regression lines and slopes closer to the 1:1 line but intercepts further from the origin than D1 and D3.

The Cramer-von Mises test was used to build distance sampling models using the point-to-1:1-line distances computed from the Q-Q plots. Models built with the D4 dataset produced the best-fit model (i.e., p-value > 0.05, AIC = -114). Models built with the D1, D2, and D3 datasets had weaker p-values (p-value < 0.05) and can be ranked based on lower AIC scores (AIC = -116 for D2, AIC = -54 for D1, and AIC = -34 for D3).

3.3 Sediment accumulation, organic carbon sequestration rates and stocks

The 30 sediment cores (68% of all the suitable ¹³⁷Cs and ²¹⁰Pb profiles) with high-quality ¹³⁷Cs and ²¹⁰Pb profiles were used to calculate mass or sediment accumulation rates, OC sequestration rates, and OC stocks (Table 2). OC sequestration rates based on ¹³⁷Cs and ²¹⁰Pb dating estimated since 1954 and 1963 of 44 suitable sediment cores (where both ¹³⁷Cs and ²¹⁰Pb profiles were available) are presented in Supplementary Table 2.

Table 2: Sedimentation accumulation, OC stocks, and sequestration rates of undisturbed wetlands estimated using high-quality ¹³⁷Cs and high-quality ²¹⁰Pb profiles.

Type of radiometric dating	¹³⁷ Cs		²¹⁰ Pb		
Time-marker	1954	1963	1954	1963	
Range of accumulated sediment (Mg ha ⁻¹)	214-1,727	56-1,272	111-1,014	95-874	
Mean (standard deviation) stock of OC (Mg ha ⁻¹)	66 (29)	35 (19)	43 (18)	38 (15)	
Mean (standard deviation) rate of OC sequestration (Mg ha ⁻¹ yr ⁻¹)	1.02 (0.44)	0.63 (0.34)	0.67 (0.27)	0.68 (0.26)	

- Based on the 1954 time-marker, the total sediment accumulation ranged from 214-1,727 Mg ha⁻¹ using ¹³⁷Cs dating and 111-
- 350 1,014 Mg ha⁻¹ using ²¹⁰Pb dating. In contrast, the total sediment accumulation based on the 1963 time-marker was lower,
- 351 ranging from 56-1272 Mg ha⁻¹ using ¹³⁷Cs and 95-874 Mg ha⁻¹ using ²¹⁰Pb dating.

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- 353 The ¹³⁷Cs-derived mean OC sequestration rate was almost two times larger, at 1.02 Mg ha-1 yr-1 using the 1954 time marker
- compared to 0.63 Mg ha-1 yr-1 using the 1963 time marker. The corresponding ¹³⁷Cs-based mean OC stocks were 66 Mg ha
- 355 ¹ for 1954 and 35 Mg ha⁻¹ for 1963 (Table 2).

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- 357 The ²¹⁰Pb-derived mean OC sequestration rate was similar at 0.67 Mg ha⁻¹ yr⁻¹ using the 1954 time-marker compared to 0.68
- Mg ha⁻¹ yr⁻¹ using the 1963 time-marker. ²¹⁰Pb-based OC sequestration rates show minimal variation since they were
- calculated using the same sedimentation rate. The corresponding mean OC stocks were 43 Mg ha⁻¹ for the 1954 time-marker
- and 38 Mg ha⁻¹ for the 1963 time-marker, with a variable depth.

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- Figure 3 and Supplementary Figs. 1 to 12 present the depth distributions of ¹³⁷Cs and ²¹⁰Pb activity (along with the linear plot
- of log-transformed $^{210}\text{Pb}_{ex}$ against mass depth in g cm⁻²) of all suitable profiles (n = 44) where both radioisotope profiles are
- 364 available.

4 Discussion

- This study compared ¹³⁷Cs and ²¹⁰Pb dating for OC estimates in wetlands that were undisturbed (i.e., without direct impact
- 367 human activities) since both radioisotopes dating are known to provide reliable forecasts for recent OC sequestration rates
- 368 (i.e., post-1954, which coincides with the onset of ¹³⁷Cs atmospheric deposition) (Drexler et al., 2018; Creed et al., 2022).

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- This study highlights some advantages and disadvantages of using ¹³⁷Cs vs. ²¹⁰Pb dating. For example, the smaller number of
- suitable 210 Pb profiles (47/90 = 52%) due to the lack of a complete decay profile (following the CFCS model as described in
- Sanchez-Cabeza and Ruiz-Fernandez, 2012) indicates that 210 Pb dating is more prone to disturbance than 137 Cs (79/90 =
- 88%). For ¹³⁷Cs, even if the sediment core is disturbed, estimation of OC sequestration rates may be possible with careful
- interpretation (e.g., see Fig. 2). The larger number of sediment cores using ¹³⁷Cs dating can be beneficial in accurately
- 375 representing the heterogeneity of OC sequestration rates as it provides a larger dataset (a 36% gain compared to ²¹⁰Pb).

- Other advantages and disadvantages of ¹³⁷Cs vs. ²¹⁰Pb radioisotope dating are presented in Table 3. ¹³⁷Cs deposition was a
- pulse that occurred in 1954 and 1963. At the 1963 peak, the activity declined with time because of two factors; (1) peak

natural radioactive decay, with the ¹³⁷Cs 30-year half-life reducing the peak size over time, and (2) peak attenuation due to physical, chemical, or biological reasons (Drexler et al. 2018). The declining ¹³⁷Cs activity limits its applicability as a radioisotope dating tool; however, recent studies have reported adequate ¹³⁷Cs reference inventories for Canadian landscapes (Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Li et al., 2008; Mabit et al., 2014; Zarrinabadi et al., 2023). In addition, the use of ¹³⁷Cs inventory for dating to complement the peak has addressed the potential inadequacies that could be attributed to declining peak resolution with time. ¹³⁷Cs dating is advantageous for its simplicity in pre- and post-processing of samples and the presence of additional time-markers in other regions (Breithaupt et al., 2018; Foucher et al., 2021). For example, additional time-markers correspond to the 1986 Chernobyl nuclear plant accident and 2011 Fukushima accident. However, their effect has yet to be recorded in North America due to the substantial distance from the source. Recognizing that there may be regional or local variation in peaks, we used non-eroded ¹³⁷Cs reference sites to deal with regional variation in deposition. We also used multiple sampling sites within wetlands to assess local variation in deposition. Further, we looked for evidence from Chernobyl and Fukushima nuclear events in our data but found none (data not shown).

Further, we looked for evidence from Chernobyl and Fukushima nuclear events in our data but found none (data not shown).

137Cs dating is best suited for where the total OC is sequestered since a fixed time-marker (1954 onset or 1963 peak) or the average OC sequestration rate is needed. In contrast, the atmospheric deposition of ²¹⁰Pb is continuous and, therefore, not limited in its applicability as a radioisotope dating tool. ²¹⁰Pb dating is best suited for where variable OC sequestration rates are needed over a more extended period (earlier than 1954). ²¹⁰Pb dating is advantageous because its calculations are based on multiple points associated with progressive OC sequestration rates derived using a constant rate of supply model — including the 1954 onset and 1963 peak of ¹³⁷Cs activity—improving the precision of the OC sequestration rates. This precision enables estimating OC sequestration rates when wetlands are not undisturbed (history of drainage or at different ages since restoration) and undisturbed (no history of drainage).

Table 3: The advantages and disadvantages of using 137 Cs and unsupported 210 Pb (210 Pbex) to estimate wetland organic carbon (OC) sequestration rates.

Method of radiometric		
dating	$^{137}\mathrm{Cs}$	$^{210}\mathrm{Pb}_{\mathrm{ex}}$
Advantages •	Calculations are based on single points representing the peak (1963) and onset (1954) of the fallout. There are additional time-markers for Europe (1986 due to the Chernobyl nuclear accident) and Japan (2011 due to Fukushima Daiichi nuclear accident). Sedimentation peak may still be evident allowing estimation of OC sequestration rate even if parts of the sediment core are disturbed. Sedimentation rate can be estimated using gamma detection, which is non-destructive, so sample can be re-analyzed or used for other analyses. Less sample preparation time for gamma analysis. After the ¹³⁷ Cs activity is measured, post- processing of data is less challenging.	 Calculations are based on multiple points as there is continuous atmospheric deposition. Sedimentation rate can be estimated using two reliable methods i.e., both alpha and gamma detection. Less sample preparation time for gamma analysis compared to alpha. Gamma analysis is non-destructive, so samples can be re-analyzed for other analyses compared to alpha. Can run multiple samples at a time on a single detector in alpha method.

Method of				
radiometric		¹³⁷ Cs		$^{210}{ m Pb}_{ m ex}$
dating				
Disadvantages	•	Risk of mixing of restored and drained states when estimating OC sequestration rates due to	•	Requires full profile of ²¹⁰ Pb to do the calculations, if the sediment core disturbed
		specificity of the 1954 and 1963 time-markers		then it cannot be used to estimate OC
		(e.g., if drained and restored after 1963).		sequestration rates.
	•	Declining atmospheric deposition and declining	•	Sensitive to vertical mobilization of
		inventory due to radioactive decay (i.e., with no		sediments, but not as much as ¹³⁷ Cs.
		more nuclear testing, atmospheric deposition only	•	The alpha method is destructive, and therefore
		comes from recent accidental releases from		the sample is not available for re-use or re-
		Chernobyl and Fukushima).		analysis.
	•	Sometimes the peak is not distinct.	•	The alpha method requires extra precaution
	•	Can be estimated using only one reliable method		using hydrochloric acid for digesting, heating,
		i.e., Gamma detection.		spiking with ²⁰⁹ Po tracer (i.e., analysts come
	•	Can run only one sample at a time on a single		in direct contact with radioactive material
		detector.		²⁰⁹ Po and hot acid).
	•	Sensitive to vertical mobilization of sediments.	•	The alpha method takes more time per sample
	•	Sensitive to declining ¹³⁷ Cs inventory due to		(i.e., overnight digest followed by at least 48 h
		radioactive decay.		on the alpha counter), and is more labor
	•	Sensitive to changes in redox potential.		intensive i.e., digest, engraving coins, plating,
		More sensitive to biological and chemical activity		transferring into ensemble, etc.).
	•	compared to ²¹⁰ Pb (e.g., ¹³⁷ Cs can be taken up by	•	The alpha method requires more technical
		plants instead of sodium or potassium, and ¹³⁷ Cs		expertise for post processing of the data.
		is soluble and therefore subject to mobility into	•	Uncertainty of ²¹⁰ Pb _{ex} results derived from
		solution then moving up and down the core.		gamma analysis can be higher than alpha.
		solution then moving up and down the core.		

4.1 Challenges in interpreting the ¹³⁷Cs peak

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Bq m^{-2} .

A potential weakness of ¹³⁷Cs radioisotope dating arises from the challenges in interpreting the disturbed 1963 peak. The noise in the 1963 peak in wetlands on agricultural landscapes can be due to the redistribution of sediments since wetlands are susceptible to receiving a large mass of sediments resulting from various erosional processes due to their positioning within the landscape (Lobb et al., 2011; Zarrinabadi et al., 2023). Soil erosion resulting from wind, water, and tillage can lead to higher or lower ¹³⁷Cs levels (Li et al., 2010; Foucher et al., 2021; Zarrinabadi et al., 2023) in wetlands in agricultural landscapes, If ¹³⁷Cs enriched soil from the surrounding landscape gets deposited on top of the wetland's original soil layer, it can increase the ¹³⁷Cs inventory value (Walling and Ouine, 1991; Li et al., 2010). The magnitude of ¹³⁷Cs enrichment depends on whether sediment comes from surface or sub-surface layers (Li et al., 2010; Lal, 2020). For example, if the wetland receives ¹³⁷Cs enriched topsoil post-1963, the ¹³⁷Cs inventory would be higher than the ¹³⁷Cs depleted subsoil. The screening of ¹³⁷Cs profiles (Fig. 2a) considered the redistribution of sediments within the landscape. It demonstrated that the difficulty in disturbed ¹³⁷C profile interpretation can be reduced by investigating the cumulative ¹³⁷C inventory value. A cutoff cumulative ¹³⁷Cs inventory value can help exclude questionable profiles. The range of ¹³⁷Cs reference inventory values from previous erosion studies within the study area (e.g., Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al., 2023) can help in establishing and setting the cutoff cumulative ¹³⁷Cs inventory value. The mean ¹³⁷Cs reference inventory values in the four provinces of Canada where our wetland sites are located were utilized in this instance. The mean ¹³⁷Cs reference inventory value estimated to be 1,684 Bq m⁻² (coefficient of variation (CV) = 49%) for three AB wetland sites (53° N and 113° W) (Zarrinabadi et al. 2023), 989 Bg m⁻² (CV = 20%) for seven SK wetland sites (51° N and 107° W) (Sutherland, 1991), 1,008 Bg m^2 (CV = 17.9%) for nine SK wetland sites (51° N and 104° W) (Sutherland, 1991), 1,430 Bg m^{-2} (CV = 8.6%) for five MB wetland sites (50° N and 100° W) (Zarrinabadi et al. 2023), 1,447 Bq m^{-2} (CV = 8.8%) for three ON wetland sites (43.3° N and 80.3° W) (Kachanoski and Von Bertoldi, 1996) and 1,534 Bq m⁻² (CV = 1.7%) for three ON wetland sites (45.6° N and 74.8° W) (Kachanoski and Von Bertoldi, 1996). The ¹³⁷Cs reference inventory values were decay-corrected to 2021 for comparability. The cutoff cumulative ¹³⁷Cs inventory value for this study was selected by checking the minimum ¹³⁷Cs reference inventory value of the local region, i.e., 546 Bq m⁻² (using values reported in Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al. 2023). Hence, any ¹³⁷Cs inventory value less than 500 Bq m⁻² was considered questionable and low-quality. Additionally, > 75% of ¹³⁷C profiles had a cumulative ¹³⁷Cs

inventory value of > 500 Bg m⁻², indicating that our wetland sites' ¹³⁷Cs reference inventory value is most likely around 500

Variations in the ¹³⁷Cs peak types (e.g., distinct, broadened, fluctuating, etc.) and in ¹³⁷Cs inventory values in this study suggested that the ¹³⁷Cs profiles were impacted by various regional erosional processes in the surrounding agricultural landscape. Recent evidence suggests that there may be an outward movement of sediment and ¹³⁷Cs from the center of the wetlands to the riparian area (Zarrinabadi et al., 2023), suggesting that the base ¹³⁷Cs inventory value observed in the center of wetlands from atmospheric deposition in the 1950s-1960s could be less than that of the non-eroded reference ¹³⁷Cs values from the surrounding catchment. A ¹³⁷C inventory of a sediment core can further help assign the ¹³⁷Cs peak. For example, the ¹³⁷Cs peak was repositioned in disturbed sediment cores with higher ¹³⁷Cs inventory, where the first discernable peak after the sharp rise from the onset of ¹³⁷Cs activity and exceeding or around the reference value was assumed to be the original ¹³⁷Cs peak. ²³⁹⁺²⁴⁰Pu isotopes, like ¹³⁷Cs, are a product of nuclear testing and can be used to identify the peak of ¹³⁷Cs. Future research will use ²³⁹⁺²⁴⁰Pu to replace ¹³⁷Cs as ¹³⁷Cs levels diminish.

4.2 Challenges in interpreting ¹³⁷Cs and ²¹⁰Pb profiles

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Mobilization of ¹³⁷Cs and ²¹⁰Pb in the sediment often occurs in wetlands. Vertical mixing of ¹³⁷Cs within sediments can be caused by remobilization and redistribution by wind and water, ice movement and inversion, disturbance by animals, and disturbance by humans that ditch and drain the wetlands till through the wetland when it is dry and let cattle access them for water which causes disturbances to the bottom sediments (Robbins et al., 1977; Milan et al., 1995; Takahashi et al., 2015). Vertical mixing affects the profile by attenuating the peak upward and downward (which we addressed using the ¹³⁷Cs inventory value and not just the peak when assessing the profile). Horizontal mixing of ¹³⁷Cs dating within sediment occurs by physical movement of sediments into or out of the wetland, causing uneven distribution of the OC content, where accumulation may be high at the edges of open water of the wetland (Lobb et al., 2011; Zarrinabadi et al., 2023). This heterogeneity can be caused by the horizontal focusing of sediments in sub-basins within a wetland, i.e., multiple center points. Sampling multiple sediment cores from individual wetlands can help capture the heterogeneity within the wetland. Suppose the 137Cs activity of most of the sediment cores from a particular wetland is noisy with a higher inventory value (e.g., ¹³⁷Cs profile of S-LO-I-W4-T2-CW-R2 in Supplementary Fig. 2a). In that case, the impact by erosional processes can be deduced with higher certainty. The higher observed inventory value could result from the movement of enriched material via erosion/lateral flow to the center of the wetland, increasing the number of ¹³⁷Cs. In this study, the assumption of no substantial downward mixing of ¹³⁷Cs was supported by (1) sampling three cores from each wetland, (2) assessing the sharpness of the rise of the peaks (a sharp rise means negligible mixing), (3) examining the cumulative ¹³⁷Cs inventory value and validating against the known reference level, (4) classifying ¹³⁷Cs profiles, and (5) corroborating with ²¹⁰Pb dating.

4.3 ¹³⁷Cs vs. ²¹⁰Pb derived OC sequestration rates and stocks

 137Cs radioisotope dating using the 1954 or 1963 time-markers gives reasonable estimates of OC sequestration rates as compared to ²¹⁰Pb radioisotope dating. The ¹³⁷Cs-²¹⁰Pb Q-Q plot of the 1963 OC sequestration rates is closer to the 1:1-line, suggesting compatibility between ¹³⁷Cs- and ²¹⁰Pb-based estimates (Fig. 5c and 5d). Conversely, the ¹³⁷Cs-²¹⁰Pb Q-Q plot of the 1954 OC sequestration rates showed more deviation from the 1:1 line; ¹³⁷Cs-based OC sequestration rates were more dispersed and were higher than the ²¹⁰Pb-based OC sequestration rates (Fig. 5a and 5b). The mean OC sequestration rates in Table 2 further verify the comparability of OC sequestration rates using the 1963 time-marker (mean ¹³⁷Cs OC sequestration rate is 0.63 Mg ha⁻¹ yr⁻¹ while mean ²¹⁰Pb OC sequestration rate is 0.68 Mg ha⁻¹ yr⁻¹). The dispersion using the 1954 time-marker (mean ¹³⁷Cs OC sequestration rate is 1.02 Mg ha⁻¹ yr⁻¹ while mean ²¹⁰Pb OC sequestration rate is 0.67 Mg ha⁻¹ yr⁻¹). Providing better sequestration rate estimates has consequences for estimating OC stocks with an improved degree of accuracy, which may provide policymakers with better tools to make informed carbon management decisions supported with data.

To put our findings into practice and in the broader OC sequestration perspective, we consider a scenario where two independent studies were performed using ¹³⁷Cs and ²¹⁰Pb (with the CFCS model) at the exact locations. If the cores were not selected based on the criteria we used to choose high-quality profiles, then these two studies' OC sequestration rate estimates are likely to disagree. However, we know and have demonstrated through our findings that they are linearly dependent, and the equation of our linear regression lines may be used to transform one estimate to the other. Consequently, if the cores were selected based on our selection criteria, then one can expect the OC sequestration rate estimates to have similar values, which alleviates the interpretation challenges of having two different estimates from two independent studies. This observation may help with consistency when disagreements in estimates are observed. Another practical application of our findings may be in data augmentation. For example, if we have ²¹⁰Pb data for a set of locations and ¹³⁷Cs data for other locations, the linear regression equation could transform ²¹⁰Pb data to augment ¹³⁷Cs data, and vice versa. This can help data-driven modelling approaches, whereas larger datasets help achieve robust modelling tools. Similarly, because OC stocks can be derived from sequestration rates for specific years, estimates derived using one radioisotope can be used to estimate OC from a dataset derived from another estimate, further contributing to the augmentation of the corresponding OC stock data.

Based on the results of this study, we recommend (1) using high-quality ¹³⁷Cs and ²¹⁰Pb profiles to estimate OC sequestration rates, (2) interpreting 137Cs profiles from agricultural landscapes carefully from the perspective of redistribution of sediments, (3) using both ¹³⁷Cs and ²¹⁰Pb to compare and validate estimates if logistic approves. However, in case where one had to choose between ¹³⁷Cs and ²¹⁰Pb we recommend (1) For ¹³⁷Cs: use 1963 time-markers to estimate OC sequestration rates (compared to 1954) since it is found to be most comparable with ²¹⁰Pb dating techniques (CFCS model), (2) For ²¹⁰Pb

(CFCS model): OC sequestration rates from present to 1963 can be estimated with highest precision since we corroborated the estimates with ¹³⁷Cs. However, we cannot comment on the precision of ²¹⁰Pb-based OC sequestration rate estimation before 1963 based on the scope of this study.

5 Conclusions

Information regarding OC sequestration rates within temperate inland wetland soils is crucial for evaluating the potential of these ecosystems to serve as natural climate solutions. Radiometric dating using ¹³⁷Cs and ²¹⁰Pb presents a valuable tool for estimating the recent OC sequestration potential of wetlands. Notably, a robust 1:1 linear correlation has been observed between ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration rates in high-quality sediment profiles.

While estimations based on the onset of ¹³⁷Cs in 1954 or its peak in 1963 were reasonable, estimates anchored to the 1963 peak of ¹³⁷Cs exhibited closer alignment with those derived from ²¹⁰Pb data (using the CFCS model). These findings suggest that estimates derived from ¹³⁷Cs and ²¹⁰Pb radioisotope dating methods are interchangeable and reasonably comparable when utilizing the 1963 ¹³⁷Cs time-marker.

Combining ¹³⁷Cs and ²¹⁰Pb tracers provides a comprehensive assessment of sedimentation rates. While one tracer offers an average sedimentation rate over 60 years, the other provides a temporal trend over the same period. This interchangeability enables more thorough evaluations of the average sedimentation rate in wetlands, which is crucial for leveraging them as natural climate solutions.

Code and data availability. The R code for the distance sampling modelling along with the data to run the code is available at https://doi.org/10.5281/zenodo.10951658. The organic carbon (OC) sequestration rates data used to check the comparability of the radioisotope profiles can be found in the Supplement. These sequestration rate data and the geographical locations, years of sampling, and additional information about the sediment cores are available at https://doi.org/10.5281/zenodo.13696300. The radioisotope profiles used for screening are in the paper and Supplement. The paper and Supplement present other relevant data to support our conclusion.

Author contributions. The authors' contributions are as follows: PM: methodology, field and lab analysis, statistical analysis and modelling, writing; IFC: conceptualization, methodology, field and lab analysis, editing, supervision; CGT: conceptualization, editing, supervision; EE: methodology, field and lab analysis, editing; and DAL: methodology, field and lab analysis, editing.

Competing interests. The authors declare that they have no conflict of interest.

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