Technical Note: Comparison of radiometric techniques for estimating recent organic carbon sequestration rates in freshwater mineral soil wetlands

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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 (^{137}Cs) and lead-210 (^{210}Pb) radioisotopes is commonly used for measuring OC sequestration rates in wetland sediments. ^{137}Cs radioisotope dating is relatively simple, with calculations based on a single point representing the onset (1954) or peak (1963) of the ^{137}Cs fallout. ^{210}Pb radioisotope dating is more 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 ^{137}Cs or ^{210}Pb dating, or their combination. However, ^{137}Cs and ^{210}Pb 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 ^{137}Cs and ^{210}Pb profiles into high- and low-quality sediment profiles, and we compare dating using the 1954 and 1963 time-markers. Our findings suggest that ^{137}Cs- and ^{210}Pb-based OC sequestration 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 quality, and preventing floods. Moreover, these wetlands have the potential to sequester organic carbon (OC), making them candidates to be natural climate solutions by offsetting carbon emissions. To estimate the OC sequestration potential, it is critical 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 estimate the OC storage rates of wetlands over periods of 10 to ≥ 1,000 years. Frequently used radioisotopes for radiometric dating are cesium-137 (^{137}Cs) and lead-210 (^{210}Pb), which can be used to estimate relatively

recent (up to the last 100 years) OC sequestration rates (Villa and Bernal, 2018). The estimation of OC sequestration rates involves building an age-depth profile or model of ^{137}Cs and ^{210}Pb from sediment cores that demonstrate the relationship between the depth of sediment layers and their corresponding age. Since the inorganic radioisotopes (^{137}Cs and ^{210}Pb) strongly bind with the soil particles once in contact, the radioisotopes can act as an efficient tracer for investigating OC

35 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 ^{137}Cs and ^{210}Pb to estimate wetland OC sequestration rates are presented in Table 1. ^{137}Cs is an

40 artificial radioisotope which was formed due to 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, ^{137}Cs isotope was released into the atmosphere, which was then deposited around the globe. ^{137}Cs 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). ^{137}Cs dating assumes constant sedimentation rates measured since 1954 and 1963. The onset and

45 the peak of ^{137}Cs activity at 661.6 keV can be used to mark 1954 and 1963, respectively. These time-markers (1954 and 1963) can be used to date sediment layers (Pennington et al., 1973; Ritchie and McHenry, 1990; DeLaune et al., 2003) and consequently the OC sequestration rates. ^{137}Cs 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. ^{137}Cs dating requires a gamma spectrometer to estimate

50 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). ^{137}C dating calculations are less complicated than ^{210}Pb, with little modelling knowledge or expertise needed (Breithaupt et al., 2018).

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Unlike ^{137}Cs, ^{210}Pb is a naturally occurring radionuclide of ^{238}U and deposits atmospherically from the decay of ^{226}Ra (Walling and He, 1999). ^{210}Pb 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). ^{210}Pb activity can be measured using gamma (observed at 46.5 keV) and alpha spectrometry (destructive) (Walling and He, 1999; Bellucci

60 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 ^{210}Pb dating where polonium-210 (^{210}Po) activity is measured, assuming both ^{210}Pb

and ^{210}Po are in a secular equilibrium. ^{210}Pb activity is calculated by comparing ^{210}Po activity against the known activity of ^{209}Po (isotope tracer). In alpha analysis, the additional time required for sample preparation is compensated for by running

65   multiple samples simultaneously (Bansal et al., 2023). Gamma and alpha spectrometry of ^{210}Pb provides the total ^{210}Pb activity, which incorporates unsupported (or excess) ^{210}Pb (^{210}Pb$_{ex}$) and supported ^{210}Pb. ^{210}Pb$_{ex}$ is used to determine the sediment accumulation rate. The choice of model used in ^{210}Pb 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 ^{210}Pb dating are (1) constant flux-constant sedimentation (CFCS) model, (2) constant rate of supply (CRS) model, and (3)

70   constant initial concentration (CIC) model (Appleby and Oldfield, 1978; Sanchez-Cabeza and Ruiz-Fernandez, 2012; Kamula et al., 2017). Both ^{137}Cs and ^{210}Pb 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).

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

Method of radiometric dating	^{137}Cs	^{210}Pb$_{ex}$
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 radiometric dating	48 h for each sample for gamma spectrometry.	48 h for each sample for gamma spectrometry. 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 sedimentation 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; Sanchez-Cabeza and Ruiz-Fernandez; Kamula et al., 2017)

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The combined use of ^{137}Cs and ^{210}Pb may improve the accuracy of the dating estimation, according to Drexler et al. (2018) and 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., ^{137}Cs peak clarity), variability in atmospheric deposition, and mobilization of radioisotopes can contribute to

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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.

85   This research paper compares the use of ^{137}Cs- and ^{210}Pb to estimate recent OC sequestration rates in intact (i.e., not directly impaired by human activities) freshwater mineral soil wetlands located on agricultural landscapes. 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 ^{137}Cs or ^{210}Pb radioisotope dating. The OC sequestration rates based on ^{137}Cs vs. ^{210}Pb and based on the onset (1954) or peak (1963) of ^{137}Cs radioisotope activity were then compared. This study helps reduce uncertainty in

90   studies that rely on ^{137}Cs or ^{210}Pb radioisotope dating.

2   Methods

2.1   Sediment core collection

Triplicate sediment cores were collected from 30 intact freshwater mineral soil wetlands in agricultural landscapes across southern Canada (Supplementary Fig. 1). A summary of physical characteristics of these wetlands can be found in

95   Supplementary Table 1. These wetlands were intact, 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) and 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

100   increments. The sediment cores were stored at -5 °C for further processing at the laboratory.

2.2   Generation of ^{137}Cs and ^{210}Pb 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, removing gravel improves the estimate of radioisotopes and OC. The increments were counted at

105   661.6 keV for ^{137}Cs activity and 46.5 keV for and ^{210}Pb activity. ^{137}Cs analysis was performed using a gamma spectrometer, and ^{210}Pb analysis was performed using both gamma and alpha spectrometers to increase throughput rates. The gamma analysis was conducted using the high-purity germanium detectors, Broad Energy Germanium detectors (BE6530) and high-resolution 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).

110   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 ^{210}Pb, generating comparable results (Zaborska et al., 2007).

2.3 Screening of ^{137}Cs and ^{210}Pb profiles

The actual ^{137}Cs peak can vary from the expected peak, increasing uncertainty in ^{137}Cs dating (Drexler et al., 2018). ^{137}Cs
115   peaks can be "noisy" or "disturbed"; i.e., flattened, broadened, truncated, mixed, fluctuating (Drexler et al., 2018), or one-sided where the ^{137}Cs peaks appear at the surface of the sediment core (indicating no or little sedimentation since 1963). Such noise in ^{137}Cs peaks can be caused by diffusion, disturbance, removal, and receiving ^{137}Cs enriched sediments (Anderson et al., 1987; Milan et al., 1995; Jagercikova et al., 2015). Such noise in ^{137}Cs 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, 79 were suitable (complete and datable) for ^{137}Cs dating and 47 were suitable for ^{210}Pb dating. Suitability for ^{137}Cs profiles for dating was assessed by zero activity before the onset and the peak of ^{137}Cs activity. The suitability of ^{210}Pb profiles for dating was assessed by determining the exponential decline in ^{210}Pb activity with depth until background levels are reached.

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Classification of the selected ^{137}Cs profiles: The 79 suitable ^{137}Cs profiles were then classified into high- and low-quality using the following steps (Fig. 1a):

1. The ^{137}C depth profile and the shape of the peak were assessed. The presence of a clear and distinct peak associated
130   with several points on both sides of the peak verified the ^{137}Cs depth profile as high-quality.

2. When analyzing sediment samples, a clear peak in ^{137}Cs activity didn't always exist. If a peak was absent, which could have resulted from sediment influxes with very high or very low ^{137}Cs 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 ^{137}Cs profile was considered as high-quality. Conversely, if the cumulative ^{137}Cs inventory value for the
135   entire profile was < 500 Bq m^{-2}, then the ^{137}Cs profile was considered as low-quality. The cutoff cumulative ^{137}Cs inventory value of 500 Bq m^{-2} was established by assessing the ^{137}Cs reference inventory value, the value of ^{137}Cs present in a non-eroded system with an undisturbed profile. The ^{137}Cs reference inventory value differs from region to region (Owens and Walling, 1996), and the most proximal regional value was used to select the cutoff ^{137}Cs inventory value (Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al., 2023). The ^{137}Cs
140   reference inventory is a catchment-wide reference value and not specific to the wetland center; thus, the cumulative ^{137}Cs inventory value of 500 Bq m^{-2} was viewed as a conservative indicator of the suitability of the ^{137}Cs profiles.

Classification of the selected ^{210}Pb *profiles:* The 47 suitable ^{210}Pb profiles were classified into high- and low-quality profiles based on the following steps (Fig. 1b):

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1. ^{210}Pb activity were plotted with a log-transformed ^{210}Pb$_{ex}$ against mass (g cm^{-2}).

2. A linear regression analysis was performed (where the slope is used to derive the 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.

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4. If either of the normality and constant variance tests were not passed, had an $R^2 \leq 0.5$, or a p-value ≥ 0.05, then the ^{210}Pb profile was considered low-quality.

The ^{210}Pb profiles were also classified based on a two-step piecewise linear regression model to capture recent shifts in OC

155 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 selected for the study but can reduce the number of available sediment cores for the study and potentially ignore the natural variability and significant events occurring in the real environment.
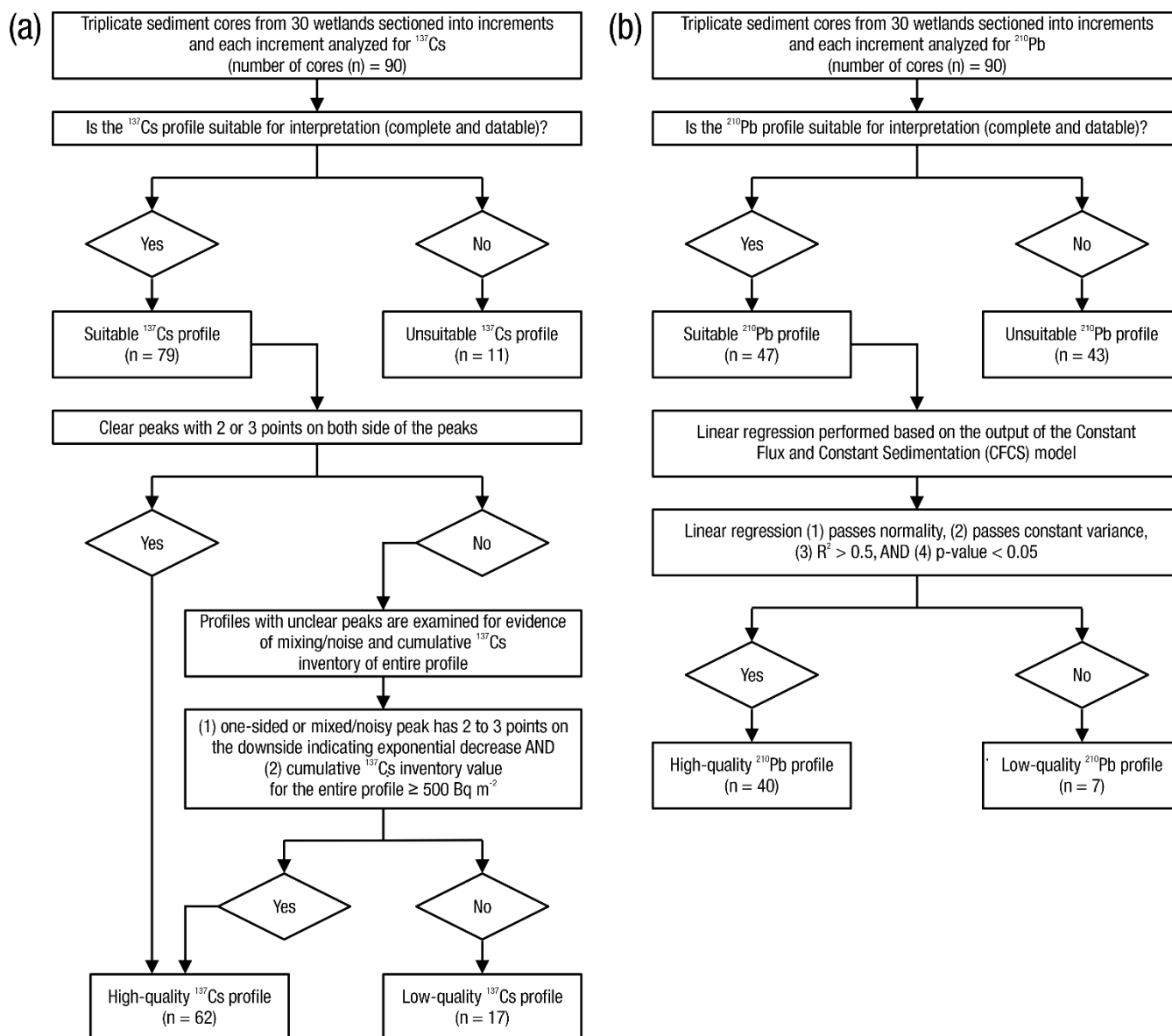
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Figure 1: Classification of high- and low-quality ^{137}Cs and ^{210}Pb profiles outlining the decision frameworks for screening (a) ^{137}Cs profiles and (b) ^{210}Pb profiles.

2.4   OC stocks and sequestration rates

165   Radioisotope activity measurements were utilized to assign two time-markers, one for 1954 and the other for 1963, in the sediment cores. Sediment radioisotope dating was used to calculate the rates of sediment accumulation and OC sequestration.

For ^{137}Cs dating, sediment accumulation and OC sequestration rates (Mg ha^{-1} yr^{-1}) were estimated using the cumulative sum

170   of sediment or OC (Mg ha^{-1}) from the surface to the depth corresponding to the time-markers of ^{137}Cs of each core and dividing by the number of years from the time-marker to the years the samples were collected. For ^{137}Cs profiles with noisy peaks and comparatively larger cumulative ^{137}Cs inventory values, the first elongated peak with a sharp rise after the onset of the ^{137}Cs peak was considered the 1963 peak instead of the peak with the highest activity in the profile.

175   For ^{210}Pb dating, sediment accumulation and OC sequestration rates were estimated using the Constant Flux and Constant Sedimentation (CFCS) model (Sanchez-Cabeza and Ruiz-Fernandez, 2012, Kamula et al., 2017). Here, ^{210}Pb$_{ex}$ was estimated by subtracting ^{226}Ra activity (186 keV) from the total ^{210}Pb activity. The CFCS model uses the log-linear relationship of ^{210}Pb$_{ex}$ with depth and converts ^{210}Pb$_{ex}$ to the sediment accumulation rate and, consequently, the OC sequestration rate. The OC stock was estimated by taking the cumulative sum of OC (Mg ha^{-1}) from the surface of each sediment core to the depth

180   increments represented by the time-marker (e.g., 1963).

OC stocks for the 1954 and 1963 time-markers were calculated by multiplying the OC content per unit mass of soil (g; OC content was calculated from OC concentration in % measured by loss-on-ignition method, Kolthoff and Sandell, 1952) by the mass of sediment for each section interval and specific depth interval per unit area (g cm^{-2}) down the profile to the

185   respective time-marker.

2.5   Statistical analysis

Statistical analyses were conducted using sediment cores where both ^{137}Cs and ^{210}Pb-based OC sequestration rates were available. The ^{137}Cs- and ^{210}Pb-based estimates of OC sequestration rates were compared using a quantile-quantile (Q-Q) plot. First, the comparison was done via assessment of the Q–Q plots. Four sample datasets were used to construct Q-Q plots

190   to compare the distribution of ^{137}Cs and ^{210}Pb-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 (number of sediment cores
195   (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).

200   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 pth percentiles of the two OC sequestration rate estimates that were 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). To assess whether the two OC sequestration rate estimates are similar, the distribution of the points on the Q-Q plot was compared against the $y = x$ (1:1) line. 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

205   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 ^{137}Cs-

210   and ^{210}Pb-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 path of the observer (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 Cramer-von Mises test was used to estimate whether the distances (q1, q2, …, qn) from the points to the 1:1 line were from a

215   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

220   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 likelihood 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

225 was computed using the "distance" package in R version 4.0.3 (Miller and Clark-Wolfe, 2023; R Core Team, 2023).

3 Results

3.1 High- and low-quality ^{137}Cs and ^{210}Pb profiles

Of the 79 suitable ^{137}Cs profiles, 62 (78%) were classified as being of high-quality. Of the 62 high-quality ^{137}Cs profiles, 61% had clear and distinct peaks, with a smooth rise and decline, while the remaining 39% had noise—either one-sided

230 peaks or disturbed peaks. Of the 62 high-quality ^{137}Cs profiles, 4 (6.5%) were repositioned to capture the ^{137}Cs enriched sediments post 1963. In these profiles, which had a cumulative ^{137}Cs inventory value > 1,200 Bq m^{-2}, the depth that corresponded to ^{137}Cs cumulative inventory value of ~500 Bq m^{-2} was considered as the 1963 time-marker. Two ^{137}Cs profiles were considered high-quality despite a cumulative ^{137}Cs inventory value < 500 Bq m^{-2} because the 1963 peak was clear, distinct, and elongated with two-to-three points on both sides of the peak. One ^{137}Cs profile was considered high-

235 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 ^{137}Cs inventory value of 499 Bq m^{-2}. One ^{137}Cs profile was classified as low-quality despite a cumulative ^{137}Cs inventory value > 500 Bq m^{-2} because the peak was highly fluctuating and not discernible.

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

There were 44 sediment cores with both ^{137}Cs and ^{210}Pb suitable profiles available. Of these, 30 were categorized as high-quality ^{137}Cs and high-quality ^{210}Pb (Fig. 2a), 6 were categorized as high-quality ^{137}Cs and low-quality ^{210}Pb (Fig. 2b), 7 were categorized as low-quality ^{137}Cs and high-quality ^{210}Pb (Fig. 3a), and 1 was categorized as low-quality ^{137}Cs and low-

245 quality ^{210}Pb (Fig. 3b). (See Supplementary Figs. 2 to 13 for ^{137}Cs and ^{210}Pb profiles in all study wetlands.)
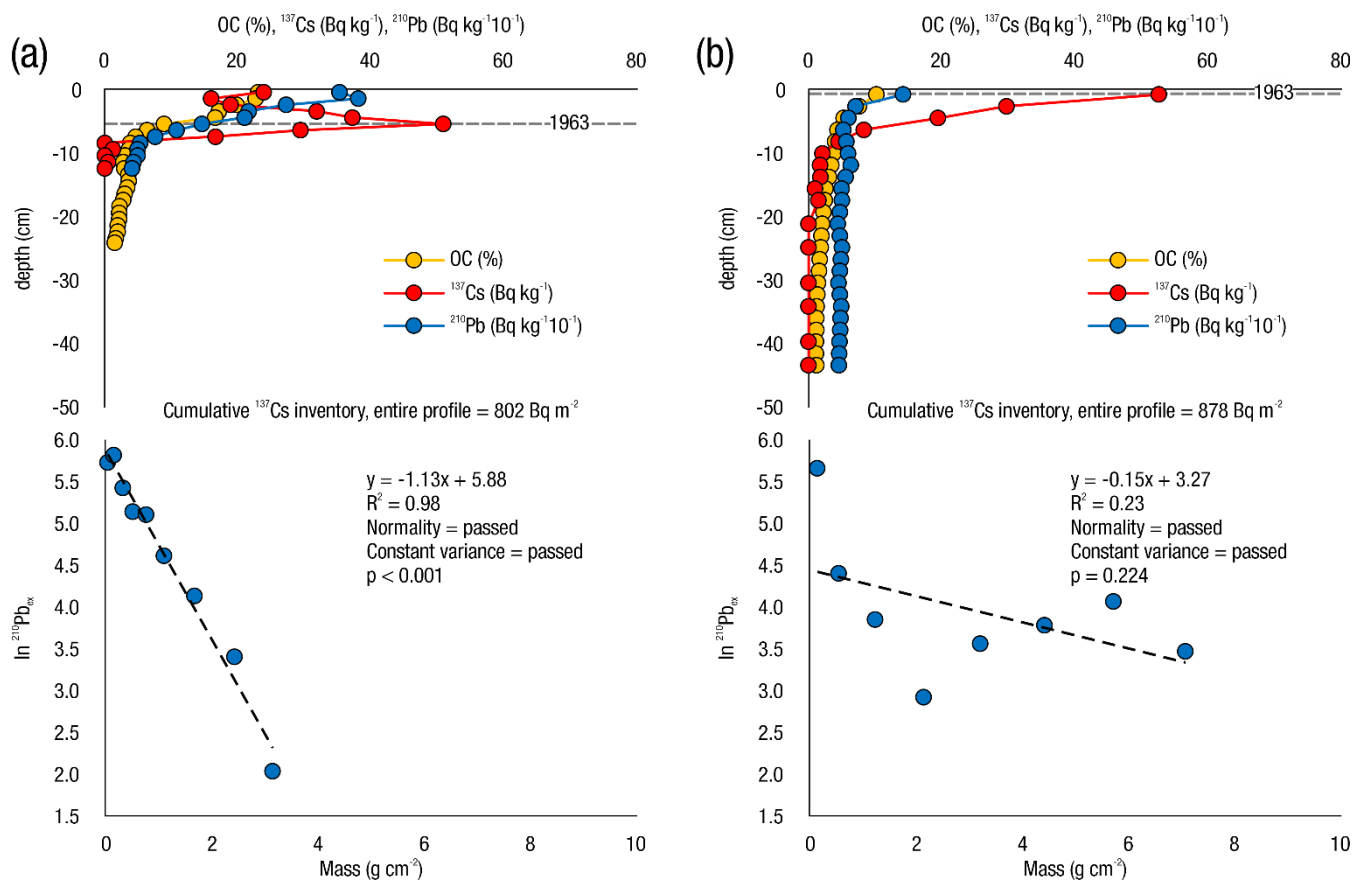
Figure 2: Examples of ^{137}Cs and ^{210}Pb classifications showing OC (%), ^{137}Cs (Bq kg^{-1}), and ^{210}Pb (Bq kg^{-1}) depth profiles and plots of log-transformed ^{210}Pb$_{ex}$ against mass (g cm^{-2}): (a) high-quality ^{137}Cs and high-quality ^{210}Pb (SK-A3 T1); (b) high-quality ^{137}Cs and low-quality ^{210}Pb (MB-4 T3).
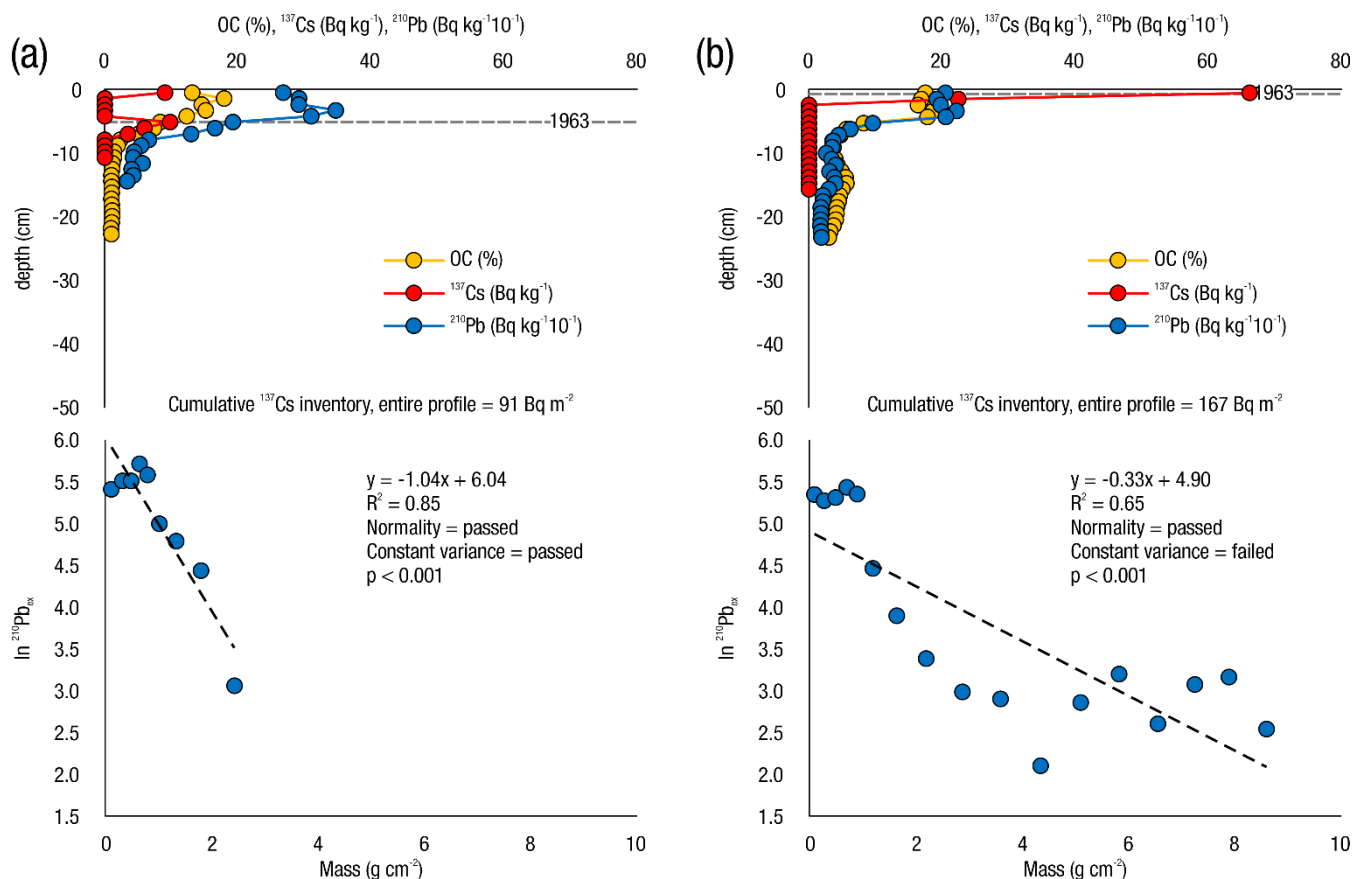
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Figure 3: Examples of ^{137}Cs and ^{210}Pb classifications showing OC (%), ^{137}Cs (Bq kg^{-1}), and ^{210}Pb (Bq kg^{-1}) depth profiles and plots of log-transformed ^{210}Pb$_{ex}$ against mass (g cm^{-2}): (a) low-quality ^{137}Cs and high-quality ^{210}Pb (SK-A1 T3); and (b) low-quality ^{137}Cs and low-quality ^{210}Pb (SK-A6 T1).

255 **3.2 ^{137}Cs vs. ^{210}Pb derived OC sequestration rates**

The comparability of ^{137}Cs vs. ^{210}Pb derived OC sequestration rates was investigated through both visual inspection of the Q-Q plots and the Cramer-von Mises test which assigned significance of the distance of the points from the 1:1 line assessed with p-value and the AIC.

260   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. 4).
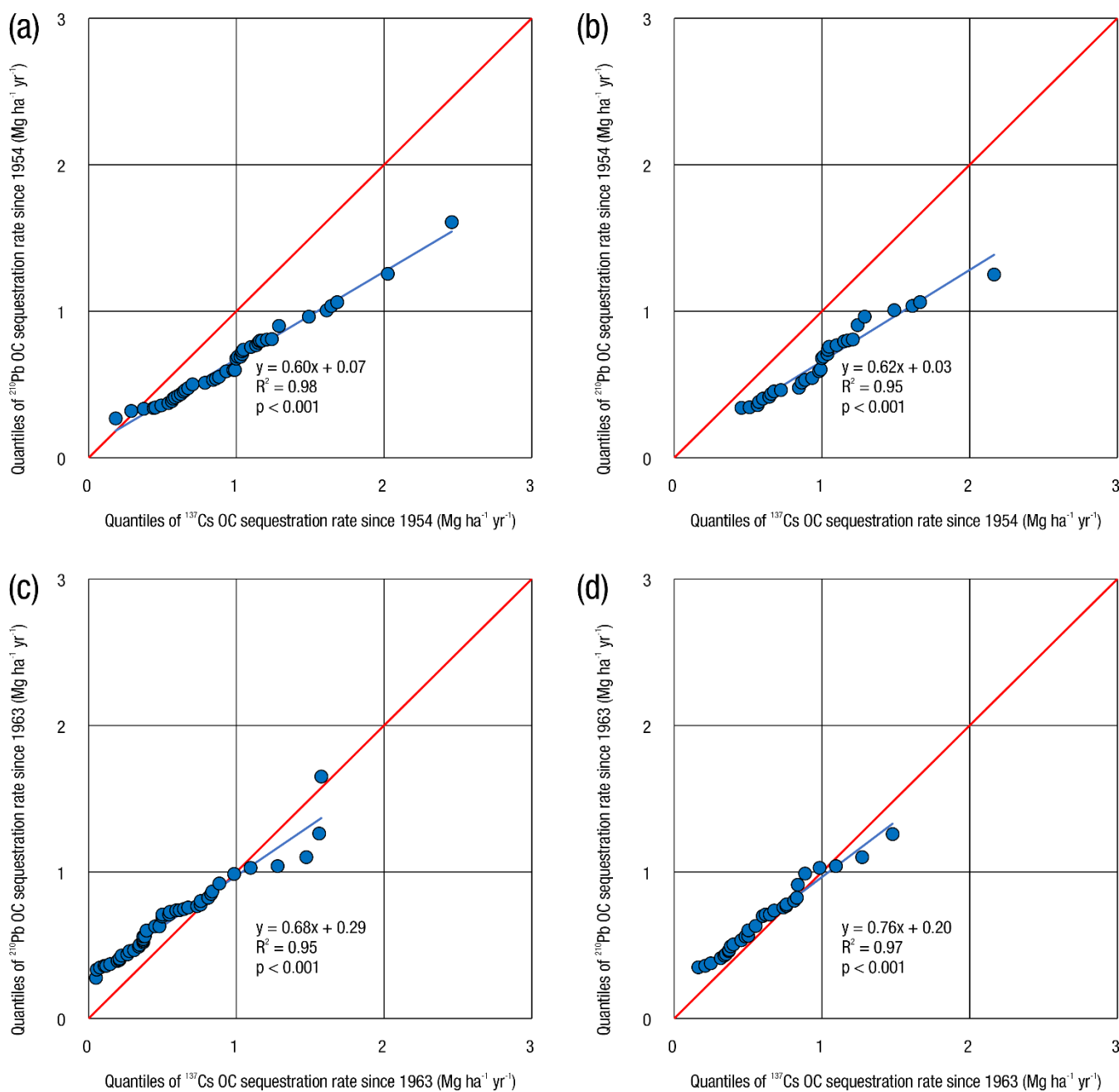
Figure 4: Q-Q plot of ^{137}Cs- vs. ^{210}Pb-based organic carbon (OC) sequestration rates using (a) all suitable ^{137}Cs and ^{210}Pb profiles estimated since 1954 (D1), (b) high-quality ^{137}Cs and ^{210}Pb profiles estimated since 1954 (D3), (c) all suitable ^{137}Cs and ^{210}Pb profiles estimated since 1963 (D2), and (d) high-quality ^{137}Cs and high-quality ^{210}Pb profiles estimated since 1963 (D4).

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Visual inspection of the Q-Q plots showed that the points for D2 (all suitable ^{137}Cs and ^{210}Pb profiles using the 1963 time-marker; Fig. 4c) and D4 (high-quality ^{137}Cs and ^{210}Pb profiles using the 1963 time-marker; Fig. 4d) were distributed more closely along the 1:1 line compared to that of D1 (all suitable ^{137}Cs and ^{210}Pb profiles using the 1954 time-marker; Fig. 4a) and D3 (high-quality ^{137}Cs and ^{210}Pb profiles using the 1954 time-marker; Fig. 4b).

An intercept closer to 0 and 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. 4a); s = 0.62, i = 0.03 for D3 (Fig. 4b); s = 0.68, i = 0.29 for D2 (Fig. 4c); and s = 0.76, i = 0.20 for D4 (Fig. 4d). D2 and D4 had regression lines and slopes closer to the 1:1 line but intercepts further from the origin compared to 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, OC sequestration rates and stocks

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

Table 2: Sedimentation accumulation, OC stocks, and sequestration rates of intact wetlands estimated using high-quality ^{137}Cs and high-quality ^{210}Pb 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)

The total sediment accumulation based on the 1954 time-marker ranged from 214-1,727 Mg ha^{-1} using ^{137}Cs dating and 111-1,014 Mg ha^{-1} using ^{210}Pb dating. In contrast, the total sediment accumulation based on the 1963 time-marker was lower,

290   ranging from 56-1272 Mg ha^{-1} using ^{137}Cs and 95-874 Mg ha^{-1} using ^{210}Pb dating.

The ^{137}Cs 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 ^{137}Cs-based mean OC stocks were 66 Mg ha^{-1} for 1954 and 35 Mg ha^{-1} for 1963 (Table 2).

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The ^{210}Pb-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. ^{210}Pb-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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The depth distributions of ^{137}Cs and ^{210}Pb activity (along with the linear plot of log-transformed ^{210}Pb$_{ex}$ against mass in g cm^{-2}) of all suitable profiles (n = 44) where both radioisotope profiles are available are presented in Fig. 2 and Supplementary Figs. 2 to 13.

4   Discussion

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

This study highlights some of the advantages and disadvantages of using ^{137}Cs vs. ^{210}Pb dating. For example, the

310   significantly smaller number of suitable ^{210}Pb profiles (47/90 = 52%) due to lack of a complete decay profile indicates that ^{210}Pb dating is more prone to disturbance than ^{137}Cs (79/90 = 88%). For ^{137}Cs, even if the sediment core is disturbed, estimation of OC sequestration rates may be possible with careful interpretation (e.g., see Fig. 1). The larger number of sediment cores using ^{137}Cs dating can be beneficial in accurately representing the heterogeneity of OC sequestration rates as it provides a larger dataset (a 36% gain compared to ^{210}Pb).

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

natural radioactive decay, with the [137]Cs 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 ^{137}Cs activity limits its applicability as a

320 radioisotope dating tool; however, recent studies have reported adequate ^{137}Cs 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 ^{137}Cs inventory for dating to complement the peak has addressed the potential inadequacies that could be attributed to declining peak resolution with the passage of time. [137]Cs 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

325 al., 2021). For example, there are additional time-markers corresponding to the 1986 Chernobyl nuclear plant accident and 2011 Fukushima accident, although their effect is less felt in North America due to the substantial distance from the source.[137]Cs dating is best suited for where the total OC sequestered since a fixed time-marker (1954 onset or 1963 peak) or the average OC sequestration rate over that period is needed. In contrast, the atmospheric deposition of ^{210}Pb is continuous and therefore not limited in its applicability as a radioisotope dating tool. [210]Pb dating is best suited for where variable OC

330 sequestration rates are needed over a longer period (earlier than 1954). [210]Pb dating is advantageous because its calculations are based on multiple points and can provide several time-markers—including the 1954 onset and 1963 peak of ^{137}Cs activity—improving the precision of the OC sequestration rates. This precision enables estimating OC sequestration rates when wetlands are not intact (history of drainage or at different ages since restoration) and intact (no history of drainage).

Table 3: Advantages and disadvantages of using ^{137}Cs and unsupported ^{210}Pb (^{210}Pb$_{ex}$) to estimate organic carbon (OC) sequestration rates in wetlands.

Method of radiometric dating	^{137}Cs	^{210}Pb$_{ex}$
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. • Can run multiple samples at a time on a single detector.

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

4.1 Challenges in interpreting the ^{137}Cs peak

A potential weakness of ^{137}Cs 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 redistribution of sediments since wetlands are

340   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/or tillage can lead to higher or lower ^{137}Cs levels (Li et al., 2010; Foucher et al., 2021; Zarrinabadi et al., 2023) in wetlands in agricultural landscapes. If ^{137}Cs enriched soil from the surrounding landscape gets deposited on top of the original soil layer in the wetland, then it can lead to an increased ^{137}Cs inventory value (Walling and Quine, 1991; Li et al., 2010). The magnitude of

345   ^{137}Cs enrichment depends on whether sediment comes from surface or sub-surface layers. For example, if the wetland receives ^{137}Cs enriched topsoil post 1963 then the ^{137}Cs inventory would be higher than the ^{137}Cs depleted subsoil.

The screening of ^{137}Cs profiles (Fig. 1a) considered the redistribution of sediments within the landscape and demonstrated that the difficulty in disturbed ^{137}Cs profiles interpretation can be reduced by investigating the cumulative ^{137}Cs inventory

350   value. A cutoff cumulative ^{137}Cs inventory value can be useful in excluding questionable profiles. The range of ^{137}Cs 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 ^{137}Cs inventory value. The mean ^{137}Cs reference inventory values in the four provinces of Canada where our wetland sites are located are 1,684 Bq m^{-2} (coefficient of variation (CV) = 49%) for AB, 1,008 Bq m^{-2} (CV = 20.5%) for SK, 1,430 Bq m^{-2} (CV = 8.6%) for MB, and

355   1,273 Bq m^{-2} (CV = 15%) for ON (Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al. 2023). The ^{137}Cs reference inventory values were decay-corrected to 2021 for comparability. The cutoff cumulative ^{137}Cs inventory value for this study was selected by checking the minimum ^{137}Cs 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 ^{137}Cs inventory value less than 500 Bq m^{-2} was considered questionable and classified as low-quality. Additionally, > 75% of

360   ^{137}C profiles had a cumulative ^{137}Cs inventory value of > 500 Bq m^{-2}, indicating that the ^{137}Cs reference inventory value for our wetland sites is most likely around 500 Bq m^{-2}.

Variation in both the ^{137}Cs peak types (e.g., distinct, broadened, fluctuating, etc.) and in ^{137}Cs inventory values in this study suggested that the ^{137}Cs profiles were impacted by various regional erosional processes in the surrounding agricultural

365   landscape. Recent evidence suggests that there may be an outward movement of sediment and ^{137}Cs from the center of the wetlands to the riparian area (Zarrinabadi et al., 2023), suggesting that the base ^{137}Cs 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 ^{137}Cs values

from the surrounding catchment. ^{137}Cs inventory of a sediment core can further help in the assignment of the ^{137}Cs peak. For example, the ^{137}Cs peak was repositioned in disturbed sediment cores with higher ^{137}Cs inventory where the first discernable

370 peak after the sharp rise from the onset of ^{137}Cs activity and exceeding or around the reference value was assumed to be the original ^{137}Cs peak.

4.2   Challenges in interpreting ^{137}Cs and ^{210}Pb profiles

Mobilization of ^{137}Cs and ^{210}Pb in the sediment often occurs in wetlands. Vertical mixing of ^{137}Cs within sediments can be caused by remobilization and redistribution by wind and water, ice movement and inversion, disturbance by animals, and

375 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 cause disturbances to the bottom sediments (Anderson et al., 1987; Lobb et al., 1995; Milan et al., 1995; Jagercikova et al., 2015). Vertical mixing affects the profile by attenuating the peak upward and downward (which we addressed using the ^{137}Cs inventory value and not just the peak when assessing the profile). Horizontal mixing of ^{137}Cs dating within sediment occurs by physical movement of sediments into or out of the wetland causing uneven distribution of

380 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 horizonal 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. If the ^{137}Cs activity of most of the sediment cores from an individual wetland are noisy with a higher inventory value, then the impact by erosional processes can be deduced with higher certainty because the higher observed inventory

385 value could be a result of movement of enriched material to the center of the wetland, therefore increasing the quantity of ^{137}Cs from the value that would be expected if no new enriched material was introduced via erosion/lateral flow. In this study, the assumption of no substantial downward mixing of ^{137}Cs 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 ^{137}Cs inventory value and validating against the known reference level, (4) classifying ^{137}Cs profiles, and (5)

390 corroborating with ^{210}Pb dating.

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

^{137}Cs radioisotope dating using the 1954 or 1963 time-markers gives reasonable estimates of OC sequestration rates as compared to ^{210}Pb radioisotope dating. The ^{137}Cs-^{210}Pb Q-Q plot of the 1963 OC sequestration rates is in closer proximity with the 1:1-line, suggesting compatibility between ^{137}Cs- and ^{210}Pb-based estimates. Conversely, the ^{137}Cs-^{210}Pb Q-Q plot of

395 the 1954 OC sequestration rates showed more deviation from the 1:1 line; ^{137}Cs-based OC sequestration rates were more dispersed and were higher than the ^{210}Pb-based OC sequestration rates. Providing better sequestration rate estimates has

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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.

400   To put our findings in practice and in the wider OC sequestration perspective, we consider a scenario where two independent studies were performed using ^{137}Cs and ^{210}Pb (with CFCS model) at the same locations. If the cores were not selected based on the criteria we used to select 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

405   based on the criteria of our selection, 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 ^{210}Pb data for a set of locations and ^{137}Cs data for other locations, the linear regression equation could be used to transform ^{210}Pb data to augment ^{137}Cs data, and vice versa. This can help data driven

410   modelling approaches where larger datasets help to 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.

5   Conclusions

Information regarding OC sequestration rates within freshwater mineral soil wetlands is crucial for evaluating the potential

415   of these ecosystems to serve as natural climate solutions. Radiometric dating presents a valuable tool for estimating the OC sequestration potential of wetlands. Notably, a robust 1:1 linear correlation has been observed between ^{137}Cs- and ^{210}Pb-based OC sequestration rates in high-quality sediment profiles.

While estimations based on the onset of ^{137}Cs in 1954 or its peak in 1963 were reasonable, estimates anchored to the 1963

420   peak of ^{137}Cs exhibited closer alignment with those derived from ^{210}Pb data (using CFCS model). These findings suggest that estimates derived from both ^{137}Cs and ^{210}Pb radioisotope dating methods are interchangeable and reasonably comparable when utilizing the 1963 ^{137}Cs time-marker.

The combined use of ^{137}Cs and ^{210}Pb tracers provides a comprehensive assessment of sedimentation rates. While one tracer

425   offers an average rate of sedimentation over a period exceeding 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

430   comparability of the radioisotope profiles can be found in the Supplement. The radioisotope profiles used for screening can be found in the paper and Supplement. Other relevant data to support our conclusion are presented in the paper and/or Supplement.

Author contributions. The contributions of authors are as follows, PM: methodology, field and lab analysis, statistical

435   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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