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

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11 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 12 measuring OC sequestration rates in wetland sediments. ¹³⁷Cs radioisotope dating is relatively simple, with calculations 13 14 based on a single point representing the onset (1954) or peak (1963) of the ¹³⁷Cs fallout. ²¹⁰Pb radioisotope dating is more 15 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 16 depth of sediment cores need to be screened, analyzed, and interpreted carefully to estimate OC sequestration rates with high 17 18 precision. To this end, we propose a decision framework for screening ¹³⁷Cs and ²¹⁰Pb profiles into high- and low-quality 19 sediment profiles, and we compare dating using the 1954 and 1963 time-markers, i.e., the rates of sedimentation and, consequently, OC sequestration over the past ~60 years. Our findings suggest that ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration 20 21 rates are comparable, especially when using the 1963 (vs. 1954) time-marker.

22 1 Introduction

Wetlands in agricultural landscapes serve a crucial role in providing habitat for wildlife, regulating climate, improving water quality, and preventing reducing floods. Moreover, these wetlands have the potential to sequester organic carbon (OC).

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25 making them) (Bridgham et al., 2006; Nahlik and Fennessey, 2016; Bansal et al., 2023). Accounting for the balance between

the sequestration and emission of carbon can help establish wetlands as essential candidates to be for natural climate solutions
 by offsetting carbon emissions- (Hambäck et al., 2023). These wetlands embedded in agricultural landscapes are recognized

28 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 critical 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.

41 There are several ways to estimate the potential of wetlands to store OC (Bansal et al., 2023). One of these methods is 42 radiometric dating, which can estimate calculate the OC storage rates of wetlands over periods of 10 to \geq 1,000 years. 43 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). The estimation 44 of Estimating OC sequestration rates involves building an age-depth profile or model of ¹³⁷Cs and ²¹⁰Pb from sediment cores 45 that demonstrate the relationship between the depth of sediment layers and their corresponding age. Since the inorganic 46 radioisotopes (137Cs and 210Pb) strongly bind with the soil particles once in contact, the radioisotopes can act as an efficient 47 48 tracer for investigating OC sequestration rates (Ritchie and McHenry, 1990; Craft and Casey, 2000). These characteristics 49 allow for accurate tracking of carbon movement within ecosystems, thereby enabling the extraction of detailed information 50 about carbon sequestration dynamics in wetlands.

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The characteristics of ¹³⁷Cs and ²¹⁰Pb to estimate wetland OC sequestration rates are presented in Table 1. ¹³⁷Cs is an artificial radioisotope whichthat was formed due toproduced 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 60 61 (e.g., Bernal and Mitsch, 2012). ¹³⁷Cs dating assumes constant sedimentation rates measured since 1954 and 1963. or 1963. 62 In using the two time-markers for 137 Cs, we do not expect the sedimentation rates to be equal, but we do expect them to be 63 similar. The onset and the peak of 137 Cs activity at 661.6 keV can be used to mark 1954 and 1963, respectively. These time-64 markers (1954 and 1963) can be used to date sediment layers (Pennington et al., 1973; Ritchie and McHenry, 1990; DeLaune 65 et al., 2003) and consequently the OC sequestration rates. ¹³⁷Cs has an additional time-marker for Europe in 1986 due to the 66 Chernobyl nuclear accident and for Japan in 2011 due to the Fukushima Daiichi nuclear accident (Foucher et al., 2021), 67 indicating that OC sequestration estimates can be derived for different timescales. In the Americas, we do not see evidence 68 of the 1986 or 2011 ¹³⁷Cs peak, which is observed in Europe and Japan, respectively, so we did not need to use other 69 radioisotope techniques (e.g., ²³⁹⁺²⁴⁰Pu) to distinguish the 1986 or 2011 ¹³⁷Cs peak from the 1963 ¹³⁷Cs peak. ¹³⁷Cs dating 70 requires a gamma spectrometer to estimate OC sequestration rates. Sample preparation for gamma analysis involves drying, 71 weighing, disaggregating, homogenizing, and sieving (Bansal et al., 2023). Samples vary from 1 to 1,500 g, with smaller 72 samples associated with higher uncertainties and, therefore, requiring longer times to analyze. Gamma analysis counting 73 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). ¹³⁷C dating calculations are less complicated than ²¹⁰Pb, with little modelling knowledge or 74 expertise needed (Breithaupt et al., 2018).; and 24 to 48 h in Kamula et al., 2017). ¹³⁷Cs dating provides a simple result (an 75 76 average sedimentation rate), while ²¹⁰Pb dating provides a more complex result (using a supply rate model to reveal trends in 77 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 78 79 measurable when ¹³⁷Cs is no longer measurable.

Unlike ¹³⁷Cs, ²¹⁰Pb is a naturally occurring radionuclide of derived from ²³⁸U and deposits atmospherically from the decay of 81 82 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 83 of OC sequestration rates in wetlands (Craft and Richardson, 1998; Craft and Casey, 2000; Craft et al., 2018; Creed et al., 84 2022). ²¹⁰Pb activity can be measured using gamma (observed at 46.5 keV) and alpha spectrometry (destructive) (Walling 85 and He, 1999; Bellucci et al., 2007). Traditional alpha analysis requires 0.2-0.5 g of sample and additional sample 86 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) 87 activity is measured, assuming both ²¹⁰Pb and ²¹⁰Po are in a secular equilibrium. ²¹⁰Pb activity is calculated by comparing 88 ²¹⁰Po activity against the known activity of ²⁰⁹Po (isotope tracer). In alpha analysis, the additional time required for sample 89 90 preparation is compensated for 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 91

92 supported ²¹⁰Pb. ²¹⁰Pb_{ex} is used to determine the sediment accumulation rate. 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 93 atmospheric radon-222 (²²²Rn), which deposits ²¹⁰Pb onto the sediment surface from the air. Unsupported ²¹⁰Pb activity 94 95 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, 96 97 consequently, OC sequestration rates in wetlands. Some models used for ²¹⁰Pb dating are (1) constant flux-constant 98 sedimentation (CFCS) model, (2) constant rate of supply (CRS) model, and (3) constant initial concentration (CIC) model (Appleby and Oldfield, 1978; Sanchez Cabeza and Ruiz Fernandez, 2012; Kamula et al., 2017).). Both ¹³⁷Cs and ²¹⁰Pb 99 provide suitable time-markers and a longer time horizon compared to direct measurements using the time-marker of horizons 100 101 (2-10 years) to study sediment accretion and, subsequently, OC sequestration rates in wetlands (Bernal and Mitsch, 2013; 102 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. 103

104	Table 1: Characteristics of	¹³⁷ Cs and unsupported ²	²¹⁰ Pb (²¹⁰ Pbex)	dating to estima	te sedimentation	rates in wetland
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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	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 sedimentationsediment 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))

The combined use of ¹³⁷Cs and ²¹⁰Pb 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., 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.

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This The main objective of this research paper compares is to explore the use of 137 Cs- and 210 Pb to estimate recent OC 115 sequestration rates in intactundisturbed (i.e., not directly impaired by human activities) freshwater mineral soil 116 117 wetlands temperate inland wetland soils located on agricultural landscapes. Sediment cores were screened (to remove Here, we aim to (1) categorize ¹³⁷Cs or ²¹⁰Pb profiles with evidence of vertical mixing), into high- and then low-quality via a 118 119 decision framework, (2) apply the remaining profiles were used decision framework to estimate OC sequestration rates using. (3) use 1963 and 1954 time-markers to compare the ¹³⁷Cs-or- and ²¹⁰Pb-radioisotope dating. The based OC sequestration 120 121 rates based on ¹³⁷Cs vs. ²¹⁰Pb and based on the onset (1954) or peak (1963) to get a better understanding of the sediment history, and (4) select the best approach for ¹³⁷Cs radioisotope activity were then compared and ²¹⁰Pb to estimate the OC 122 sequestration rates with highest precision. This study helps reduce uncertainty in studies that rely on ¹³⁷Cs or ²¹⁰Pb 123 124 radioisotope dating.

125 2 Methods

126 2.1 Sediment core collection

127 Triplicate sediment cores were collected from 30 intact freshwater mineral soil wetlands undisturbed temperate inland 128 wetland soils in agricultural landscapes across southern Canada (Supplementary-Fig. 1). A summary of the physical 129 characteristics of these wetlands can be found in Supplementary Table 1. These wetlands were intactundisturbed, with no 130 known history of cultivation. The sediment cores were extracted from the center of the wetland, constituting the open-water 131 area. A Watermark Universal Corer (inner diameter of 6.8 cm) and or VibeCore Mini with poly core tubes (inner diameter of 132 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 133 for compacted sediment cores. Shallow (15 to 90 cm) sediment cores were sectioned into 1- or 2-cm increments. Deeper (> 134 90 cm) sediment cores were sectioned into 5-cm increments. The sediment cores were stored at -5 $^{\circ}$ C for further processing 135 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].

142 **2.2** Generation of ¹³⁷Cs and ²¹⁰Pb profiles

143 Sediment core increments were weighed (wet mass), dried, weighed again (dry mass), disaggregated, homogenized, and 144 sieved. The increments were sieved to remove gravel (> 2 mm); radioisotopes do not bind on the gravel, and gravel does not 145 contain OC; therefore, removing 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 and ²¹⁰Pb activity, ¹³⁷Cs analysis was performed using a gamma 146 spectrometer, and ²¹⁰Pb analysis was performed using both gamma and alpha spectrometers to increase throughput rates. The 147 148 gamma analysis was conducted using the high-purity germanium detectors, e.g., Broad Energy Germanium detectors 149 (BE6530) and high resolution Small Anode Germanium well detectors (GSW275L) (Mirion Technologies, Inc., Atlanta, 150 GA, USA). The alpha analysis was conducted using ORTEC® alpha spectrometer (AMETEK® Advanced Measurement 151 Technology, TN, USA). Both radioisotope analyses were performed at the Landscape Dynamics Laboratory, University of 152 Manitoba. Winnipeg, Canada. Although the underlying principles of gamma and alpha analysis differ, each focuses on 153 quantifying the decay of ²¹⁰Pb, generating comparable results (Zaborska et al., 2007). Measurement accuracy of gamma 154 detectors is ensured by assessing the counting errors with reference materials within the same geometry as the sample (e.g., 155 petri dish). Detection error was < 10% with a counting time of up to 24 h. Furthermore, Landscape Dynamics Laboratory 156 undergoes regular Proficiency Testing through the International Atomic Reference Material Agency (IARMA) and 157 previously through the International Atomic Energy Agency (IAEA) to ensure acceptable accuracy and precision of 158 analytical results using gamma spectroscopy.

159 2.3 Screening of ¹³⁷Cs and ²¹⁰Pb profiles

160 Sediment cores were screened to remove profiles with evidence of vertical mixing, and then the remaining profiles were used 161 to estimate OC sequestration rates using ¹³⁷Cs or ²¹⁰Pb radioisotope dating. The actual ¹³⁷Cs peak can vary from the expected 162 peak, increasing uncertainty in ¹³⁷Cs dating (Drexler et al., 2018). ¹³⁷Cs peaks can be "noisy" or "disturbed"; i.e., flattened, 163 broadened, truncated, mixed, fluctuating (Drexler et al., 2018), or one-sided where the 137 Cs peaks appear at the surface of 164 the sediment core (indicating no or little sedimentation since 1963). Such noise in ¹³⁷Cs peaks can be caused by diffusion, disturbance, removal, and receiving ¹³⁷Cs enriched sediments (Anderson et al., 1987; Milan et al., 1995; Jagercikova et al., 165 $\frac{2015}{10}$. The magnitude and shape of the ¹³⁷Cs peaks observed in the sediments can be affected by the atmospheric deposition 166 167 rate of ¹³⁷Cs, which is obviously affected by the number and magnitude of emission events and the weather conditions 168 following these events (UNSCEAR, 2000). The magnitude and shape of these peaks are also impacted by the movement of 169 water and sediment within each wetland's catchment during the peaks' development (Milan et al., 1995; Zarrinabadi et al., 170 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 171 can cause an upward and downward mixing of the ¹³⁷Cs in the profile, resulting in peak attenuation (Robbins et al., 1977). 172

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 $cores_7$ (30 wetlands x 3 replicates = 90), 79 were suitable (complete and datable) for ¹³⁷Cs dating and 47 were suitable for ²¹⁰Pb dating. SuitabilityOnly 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 the peak of ¹³⁷Cs activity. The suitability of ²¹⁰Pb profiles for dating was assessed evaluated by determining the exponential decline in ²¹⁰Pb activity with depth until background levels are reached.

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185 *Classification of the selected* ¹³⁷Cs *profiles:* The 79 suitable ¹³⁷Cs profiles were then classified into high- and low-quality
 186 using the following steps (Fig. 1a2a):

- The ¹³⁷C depth profile and the shape of the peak were assessed. The presence of aA 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).
- When analyzing sediment samples, a clear peak in ¹³⁷Cs activity didn't always exist- (e.g. Fig 4a). If a peak was 191 2. 192 absent, which could have resulted from sediment influxes with very high or very low ¹³⁷Cs activity levels, the total ¹³⁷Cs activity of the entire profile was examined. If the cumulative ¹³⁷Cs inventory value for the entire profile was \geq 193 194 500 Bq m⁻², then the ¹³⁷Cs profile was considered as-high-quality. Conversely, if the cumulative ¹³⁷Cs inventory 195 value for the entire profile was < 500 Bg m⁻², then the 137Cs profile was considered as low-quality. The cutoff cumulative ¹³⁷Cs inventory value of 500 Bg m⁻² was established by assessing the ¹³⁷Cs reference inventory value, 196 197 the value of ¹³⁷Cs present in a non-eroded system with an undisturbed profile. The ¹³⁷Cs reference inventory value 198 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). 199 The ¹³⁷Cs reference inventory is a catchment-wide reference value and not specific to the wetland center; thus, the 200 201 cumulative ¹³⁷Cs inventory value of 500 Bq m⁻² was viewed as a conservative indicator of the suitability of the ¹³⁷Cs 202 profiles. Ideally, reference sites are large, open, level, non-eroded areas, usually in forage or grassland since the 203 1950s, and within 10 km of the site of interest. In this study, it was impossible to identify a suitable reference site 204 near every wetland; it is usually difficult to find reference sites in agricultural landscapes. However, we could locate

205	reference sites used in other studies within 50 km except from nine wetlands in SK (51° N and 104° W), which
206	were ~150 km from the reference site. Although this was not considered ideal, it was considered acceptable.
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208	Classification of the selected ²¹⁰ Pb profiles: The 47 suitable ²¹⁰ Pb profiles were classified into high- and low-quality profiles
209	based on the following steps (Fig. 1b2b):
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211	1. 210 Pb activity were plotted with a log-transformed 210 Pb _{ex} against mass depth (g cm ⁻²).
212	2. A linear regression analysis was performed (where the slope is used to derive the mass or sediment accumulation
213	rate in g cm ^{-2} yr ^{-1}).
214	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 ,
215	then the ²¹⁰ Pb profile was classified as high-quality- (e.g., Fig. 3a).
216	4. If either of the normality and constant variance tests were not passed, had with an $R^2 \le 0.5$, or a p-value ≥ 0.05 , then
217	the ²¹⁰ Pb profile was considered low-quality- (e.g., Fig. 3b).
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219	The ²¹⁰ Pb profiles were also classified based on using a two-step piecewise linear regression model to capture recent shifts in
220	OC sequestration rates. However, no significant improvement was observed. Consequently, ²¹⁰ Pb-based OC sequestration
221	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-
222	quality profiles. Increasing the cut-off R ² value may produce better profiles to be selected chosen for the study-but. Still, it
223	can reduce the number of available sediment cores for the study and potentially ignore the natural variability and significant
224	events occurring in the real environment.



Figure 12: Classification of high- and low-quality ¹³⁷Cs and ²¹⁰Pb profiles outlining the decision frameworks for screening (a) ¹³⁷Cs profiles and (b) ²¹⁰Pb profiles.

229 **2.4 OCOrganic carbon stocks and sequestration rates**

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 or mass accumulation and OC
 sequestration.

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For ¹³⁷Cs dating, sediment accumulation and OC sequestration rates (Mg ha⁻¹ yr⁻¹) were estimated using the cumulative sum of sediment or OC (Mg ha⁻¹) from the surface to the depth corresponding to the time-markers of ¹³⁷Cs of each core and dividing by the number of years from the time-marker to the years the samples were collected. Unit conversion is applied to 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 elongated peak with a sharp rise after the onset of the ¹³⁷Cs peak was considered the 1963 peak instead of the peak with the highest activity in the profile.

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For ²¹⁰Pb dating, mass or sediment accumulation and OC sequestration rates were estimated using the Constant Flux and Constant Sedimentation (CFCS) model (Sanchez-Cabeza and Ruiz-Fernandez, 2012_{5} ; 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 relationship of ²¹⁰Pb_{ex} with mass depth and converts ²¹⁰Pb_{ex} to the mass or 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 increments represented by the time-marker (e.g., 1963).

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249 OC stocks for the 1954 and 1963 time-markers were calculated by multiplying the OC content per unit mass of soil (g;). 250 Here, OC content was calculated from OC concentration in %(%) measured by loss-on-ignition (LOI) method, (Kolthoff 251 and Sandell, 1952) by the mass of sediment for each section interval and specific depth interval per unit area (g cm⁻²) down 252 the profile to the respective time-marker. OC (%) was calculated by multiplying organic matter (%) by LOI with 0.58, 253 assuming 58% of the organic matter is carbon. Despite the broad applicability, simplicity in measurement techniques, and 254 cost-effectiveness, the LOI approach is associated with some limitations, such as the ignition of non-organic particles at high 255 temperatures or the use of a conventional conversion factor (Pribyl, 2010; Hoogsteen et al., 2015), which can result in over-256 estimation of OC content.

257 2.5 Statistical analysis

258 Statistical analyses were conducted using used sediment cores where both with ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration rates 259 were available, (number of sediment cores (n) = 44). The ¹³⁷Cs- and ²¹⁰Pb-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 to compare the distribution of ¹³⁷Cs- and ²¹⁰Pb-based OC sequestration against the
1:1 line.
The sample datasets included:
D1, all suitable ¹³⁷Cs and ²¹⁰Pb profiles with OC sequestration rates estimated since 1954 (number of sediment cores)

- (n)n = 44).
- D2, all suitable ¹³⁷Cs and ²¹⁰Pb 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).
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272 A O-O plot was calculated for each dataset. The x- and y- coordinates of a point in a O-O plot corresponded to the $\mathbf{p}^{\text{th}}\mathbf{p}a^{\text{th}}$ 273 percentiles of the two OC sequestration rate estimates that were being compared in the plot. Here, p = (k - 0.5) / n, where n 274 is the sample size and k = 1, ..., n (Jain et al., 2007). To assess whether the two OC sequestration rate estimates are similar. 275 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 276 sequestration rate estimates are similar. If the points were distributed in a straight line and close to a 1:1 line, then it 277 suggested that the two estimates came from the same distribution. In contrast, if the points were not distributed in a straight 278 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 279 plots were generated in Microsoft Excel (Microsoft 365, Version 2402, Redmond, WA).

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281 Since interpreting the Q-Q plot through a visual inspection can be subjective to human perception, we compared the 137 Cs-282 and ²¹⁰Pb-based OC sequestration rate estimates using a distance sampling model. A distance sampling model captures how 283 the detectability of objects from the observer (walking along a straight line) decreases with the increase in the object-to-284 observer distance. If the objects are closely distributed along the observer's path of the observer (i.e., if points of the Q-Q 285 plot were closely distributed along the 1:1 line), then the distribution of the distances is expected to be a half normal 286 distribution. The Cramer-von Mises test was used to estimate whether the distances (q1, q2, ..., qn) from the points to the 287 1:1 line were from a half-normal distribution. Given a set of distance samples (g1, g2, ..., gn) and a detection function, the 288 Cramer-von Mises test builds a model that fits the distance sampling data to the detection function (for details on modelling, 289 see Miller et al., 2019). A half-normal key is commonly used as a detection function, corresponding to a half-normal 290 distribution's shape.

292 The Cramer-von Mises test produced a p-value and Akaike's Information Criterion (AIC) as its test statistic. A p-value larger 293 than the significant level (p = 0.05) indicated that the likelihood of points being observed closer to the 1:1 line is high, and 294 that the likelihood probability decreases as the distances increase. This provided evidence of the points being closely 295 distributed along the 1:1 line. The AIC was used to rank the distance sampling models, which are built by the Cramer-von 296 Mises test, from best to worst (e.g., Burnham and Anderson, 2003); a small AIC value indicates a good fit to the half-normal 297 key and thus provides evidence that the points are close to the 1:1 line (Miller et al., 2019). The distance sampling Cramer-298 von Mises test was computed using the "distance" package in R version 4.0.3 (Miller and Clark-Wolfe, 2023; R Core Team, 299 2023).

300 **3** Results

301 3.1 High- and low-quality ¹³⁷Cs and ²¹⁰Pb profiles

302 Of the 79 suitable ¹³⁷Cs profiles, 62 (78%) were classified as being of high-quality. Of the 62 high-quality ¹³⁷Cs profiles, 303 61% had clear and distinct peaks, with a smooth rise and decline, while. In contrast, the remaining 39% had noise—either 304 one-sided peaks or disturbed peaks; (e.g., Fig. 4). Of the 62 high-quality ¹³⁷Cs profiles, 4 (6.5%) were repositioned to capture 305 the ¹³⁷Cs enriched sediments post 1963- (e.g., ¹³⁷Cs profile of S-LO-I-W4-T2-CW-R2 in Supplementary Fig. 2a). In these 306 profiles, which had a cumulative 137 Cs inventory value > 1.200 Bg m⁻², the depth that corresponded to 137 Cs cumulative 307 inventory value of \sim 500 Bg 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 308 309 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 310 311 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 312 313 value < 500 Bq m⁻² because the 1963 peak was clear, distinct, and elongated with two-to-three points on both sides of the 314 peak- (e.g., ¹³⁷Cs profile of M-OA-I-W4-T2-CW-R2 in Supplementary Fig. 7b). One ¹³⁷Cs profile was considered high-315 quality despite showing marginal quality to the set criteria in the decision framework, where the peak profile had good shape 316 with several points on the downside of the peak and one point on the other side and had a cumulative ¹³⁷Cs inventory value 317 of 499 Bg m⁻². One ¹³⁷Cs profile was classified as low-quality despite a cumulative ¹³⁷Cs inventory value > 500 Bg 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 318 319 Fig. 12b).

- 321 Of the 47 ²¹⁰Pb profiles, 40 (85%) were classified as high-quality.
- 322
- 323 There were 44 sediment cores with both ¹³⁷Cs and ²¹⁰Pb suitable profiles available. Of these, 30 were categorized as high-
- β 24 quality ¹³⁷Cs and high-quality ²¹⁰Pb (Fig. 2a), 63a), six were categorized as high-quality ¹³⁷Cs and low-quality ²¹⁰Pb (Fig. 2a), 63a).
- $\frac{2b}{7}$, $\frac{7}{2b}$, seven were categorized classified as low-quality ¹³⁷Cs and high-quality ²¹⁰Pb (Fig. $\frac{3a}{4a}$), and $\frac{1}{2}$ one was categorized
- as low-quality ¹³⁷Cs and low-quality ²¹⁰Pb (Fig. $\frac{3b}{4}b$). (See Supplementary Figs. $\frac{21}{10}$ to $\frac{13}{12}$ for ¹³⁷Cs and ²¹⁰Pb profiles in all
- 327 study wetlands.)





Figure 2: 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 (g cm⁻²): (a) high-quality ¹³⁷Cs and high-quality ²¹⁰Pb (SK-A3 T1); (b) high-quality ¹³⁷Cs and
 low-quality ²¹⁰Pb (MB-4 T3).



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) lowhigh-quality ¹³⁷Cs and high-quality ²¹⁰Pb (SK-A1-T3); andS-LO-I-W3-T1-CW-R1); (b) lowhigh-quality ¹³⁷Cs and low-quality ²¹⁰Pb (SK-A6-T1M-OA-I-W4-T3-CW-R3).



342 **3.2** ¹³⁷Cs vs. ²¹⁰Pb derived OC-organic carbon sequestration rates

The comparability of ¹³⁷Cs vs. ²¹⁰Pb 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.

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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. 45).



Figure 45: 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. 4e5c) and D4 (i.e., high-quality ¹³⁷Cs and ²¹⁰Pb profiles using the 1963 time-marker; Fig. 4d5d) 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. 4a5a) and D3 (i.e., high-quality ¹³⁷Cs and ²¹⁰Pb profiles using the 1954 time-marker; Fig. 4b5b).
- 357

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. 4a5a); s = 0.62, i = 0.03 for D3 (Fig. 4b5b); s = 0.68, i = 0.29 for D2 (Fig. 4e5c); and s = 0.76, i = 0.20 for D4 (Fig. 4d5d). D2 and D4 had regression lines and slopes closer to the 1:1 line but intercepts further from the origin compared to than D1 and D3.

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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).

367 **3.3** Sediment accumulation, OC 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 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.

372	Table 2: Sedimentation accumulation, OC stocks, and sequestration rates of intactundisturbed wetlands estimated using high-
373	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)

- TheBased on the 1954 time-marker, the total sediment accumulation-based on the 1954 time-marker ranged from 214-1,727
 Mg ha⁻¹ using ¹³⁷Cs dating and 111-1,014 Mg ha⁻¹ using ²¹⁰Pb dating. In contrast, the total sediment accumulation based on
 the 1963 time-marker was lower, ranging from 56-1272 Mg ha⁻¹ using ¹³⁷Cs and 95-874 Mg ha⁻¹ using ²¹⁰Pb dating.
- The ¹³⁷Cs--derived mean OC sequestration rate was almost two times larger, at 1.02 Mg ha-1 yr-1 using the 1954 timemarker 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⁻¹ for 1954 and 35 Mg ha⁻¹ for 1963 (Table 2).
- 382

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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TheFigure 3 and Supplementary Figs. 1 to 12 present the depth distributions of 137Cs and 210Pb activity (along with the
 linear plot of log-transformed 210Pbex against mass depth 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.

391 4 Discussion

This study compared ¹³⁷Cs and ²¹⁰Pb dating for OC estimates in wetlands that were intactundisturbed (i.e., without direct impact human activities) since both radioisotopes dating are known to provide reliable estimatesforecasts for recent OC sequestration rates (i.e., post--1954, which coincides with the onset of ¹³⁷Cs atmospheric deposition) (Drexler et al₇., 2018; Creed et al., 2022).

396

This study highlights some of the advantages and disadvantages of using ¹³⁷Cs vs. ²¹⁰Pb dating. For example, the significantly-smaller number of suitable ²¹⁰Pb 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 ²¹⁰Pb dating is more prone to disturbance than ¹³⁷Cs (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. 42). The larger number of sediment cores using ¹³⁷Cs dating can be beneficial in accurately 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 405 406 pulse that occurred in 1954 and 1963. At the 1963 peak, the activity is declining declined with time because of two factors: 407 (1) peak natural radioactive decay, with the 137 Cs 30-year half-life reducing the peak size over time, and (2) peak attenuation 408 due to physical, chemical, or biological reasons (Drexler et al. 2018). The declining ¹³⁷Cs activity limits its applicability as a 409 radioisotope dating tool; however, recent studies have reported adequate ¹³⁷Cs reference inventories for Canadian landscapes 410 (Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Li et al., 2008; Mabit et al., 2014; Zarrinabadi et al., 2023). In 411 addition, the use of ¹³⁷Cs inventory for dating to complement the peak has addressed the potential inadequacies that could be 412 attributed to declining peak resolution with the passage of time. ¹³⁷Cs dating is advantageous for its simplicity in pre- and 413 post-processing of samples and the presence of additional time-markers in other regions (Breithaupt et al., 2018; Foucher et 414 al., 2021). For example, there are additional time-markers corresponding correspond to the 1986 Chernobyl nuclear plant 415 accident and 2011 Fukushima accident, although. However, their effect is less felthas yet to be recorded in North America 416 due to the substantial distance from the source. Recognizing that there may be regional or local variation in peaks, we used 417 non-eroded ¹³⁷Cs reference sites to deal with regional variation in deposition. We also used multiple sampling sites within 418 wetlands to assess local variation in deposition. Further, we looked for evidence from Chernobyl and Fukushima nuclear 419 events in our data but found none (data not shown).

421 Further, we looked for evidence from Chernobyl and Fukushima nuclear events in our data but found none (data not shown). 422 ¹³⁷Cs dating is best suited for where the total OC is sequestered since a fixed time-marker (1954 onset or 1963 peak) or the 423 average OC sequestration rate over that period is needed. In contrast, the atmospheric deposition of ²¹⁰Pb is continuous and, 424 therefore, not limited in its applicability as a radioisotope dating tool. ²¹⁰Pb dating is best suited for where variable OC 425 sequestration rates are needed over a longer more extended period (earlier than 1954). ²¹⁰Pb dating is advantageous because 426 its calculations are based on multiple points and can provide several time markers associated with progressive OC 427 sequestration rates derived using a constant rate of supply model —including the 1954 onset and 1963 peak of ¹³⁷Cs 428 activity—improving the precision of the OC sequestration rates. This precision enables estimating OC sequestration rates 429 when wetlands are not intactundisturbed (history of drainage or at different ages since restoration) and intactundisturbed (no 430 history of drainage).

Table 3: Advantages The advantages and disadvantages of using 137Cs and unsupported 210Pb (210Pbex) to estimate wetland 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. 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		
dating	¹³⁷ Cs	²¹⁰ Pbex
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). 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 Eukushima) 	 Requires full profile of ²¹⁰Pb to do the calculations, if the sediment core disturbed then it cannot be used to estimate OC sequestration rates. Sensitive to vertical mobilization of sediments, but not as much as ¹³⁷Cs. The alpha method is destructive, and therefore the sample is not available for re-use or re-
	 Sometimes the peak is not distinct. Can be estimated using only one reliable method i.e., Gamma detection. Can run only one sample at a time on a single detector. 	 The alpha method requires extra precaution using hydrochloric acid for digesting, heating spiking with ²⁰⁹Po tracer (i.e., analysts come in direct contact with radioactive material ²⁰⁹Po and hot acid).
	 Sensitive to vertical mobilization of sediments. Sensitive to declining ¹³⁷Cs inventory due to radioactive decay. Sensitive to changes in redox potential. More sensitive to biological and chemical activity compared to ²¹⁰Pb (e.g., ¹³⁷Cs can be taken up by plants instead of sodium or potassium, and ¹³⁷Cs is soluble and therefore subject to mobility into solution then moving up and down the core. 	 The alpha method takes more time per sample (i.e., overnight digest followed by at least 48 on the alpha counter), and is more labor intensive i.e., digest, engraving coins, plating transferring into ensemble, etc.). The alpha method requires more technical expertise for post processing of the data. Uncertainty of ²¹⁰Pb_{ex} results derived from gamma analysis can be higher than alpha.

434 **4.1** Challenges in interpreting the ¹³⁷Cs peak

435 A potential weakness of 137 Cs radioisotope dating arises from the challenges in interpreting the disturbed 1963 peak. The 436 noise in the 1963 peak in wetlands on agricultural landscapes can be due to the redistribution of sediments since wetlands are 437 susceptible to receiving a large mass of sediments resulting from various erosional processes due to their positioning within 438 the landscape (Lobb et al., 2011; Zarrinabadi et al., 2023). Soil erosion resulting from wind, water, and/or tillage can lead to 439 higher or lower ¹³⁷Cs levels (Li et al., 2010; Foucher et al., 2021; Zarrinabadi et al., 2023) in wetlands in agricultural 440 landscapes. If ¹³⁷Cs enriched soil from the surrounding landscape gets deposited on top of the wetland's original soil layer-in the wetland, then, it can lead to an increased increase the ¹³⁷Cs inventory value (Walling and Quine, 1991; Li et al., 2010). 441 442 The magnitude of 137 Cs enrichment depends on whether sediment comes from surface or sub-surface layers. (Li et al., 2010; 443 Lal, 2020). For example, if the wetland receives 137 Cs enriched topsoil post–1963-then, the 137 Cs inventory would be higher 444 than the ¹³⁷Cs depleted subsoil.

445

446 The screening of 137 Cs profiles (Fig. <u>1a</u>2a) considered the redistribution of sediments within the landscape-and. It 447 demonstrated that the difficulty in disturbed ¹³⁷Cs profiles¹³⁷C profile interpretation can be reduced by investigating the 448 cumulative $\frac{137}{\text{Cs}}$ inventory value. A cutoff cumulative $\frac{137}{\text{Cs}}$ inventory value can be useful in excluding help exclude 449 questionable profiles. The range of 137 Cs reference inventory values from previous erosion studies within the study area (e.g., 450 Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al., 2023) can help in establishing and setting the 451 cutoff cumulative ¹³⁷Cs inventory value. The mean ¹³⁷Cs reference inventory values in the four provinces of Canada where 452 our wetland sites are located are were utilized in this instance. The mean ¹³⁷Cs reference inventory value estimated to be 453 1,684 Bq m⁻² (coefficient of variation (CV) = 49%) for AB, three AB wetland sites (53° N and 113° W) (Zarrinabadi et al. 454 2023), 989 Bq m⁻² (CV = 20%) for seven SK wetland sites (51° N and 107° W) (Sutherland, 1991), 1,008 Bq m⁻² (CV = 455 $\frac{20.5\%}{10.5\%}$ for SK. 17.9%) for nine SK wetland sites (51° N and 104° W) (Sutherland, 1991), 1.430 Bg m⁻² (CV = 8.6%) for five 456 MB, and wetland sites (50° N and 100° W) (Zarrinabadi et al. 2023), 1, $\frac{273}{447}$ Bg m⁻² (CV = $\frac{158}{880}$) for three ON 457 (Sutherland, 1991; wetland sites (43.3° N and 80.3° W) (Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al. 2023) and 458 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 459 ¹³⁷Cs reference inventory values were decay-corrected to 2021 for comparability. The cutoff cumulative ¹³⁷Cs inventory 460 value for this study was selected by checking the minimum ¹³⁷Cs reference inventory value of the local region; i.e., 546 Bq 461 m⁻² (using values reported in Sutherland, 1991; Kachanoski and Von Bertoldi, 1996; Zarrinabadi et al. 2023). Hence, any 462 137 Cs inventory value less than 500 Bg m⁻² was considered questionable and classified as low-quality. Additionally, > 75% of 463 137 C profiles had a cumulative 137 Cs inventory value of > 500 Bg m⁻², indicating that theour wetland sites' 137Cs reference 464 inventory value for our wetland sites is most likely around 500 Bq m⁻².

Variation Variations in both the ¹³⁷Cs peak types (e.g., distinct, broadened, fluctuating, etc.) and in ¹³⁷Cs inventory values in 466 467 this study suggested that the ¹³⁷Cs profiles were impacted by various regional erosional processes in the surrounding 468 agricultural landscape. Recent evidence suggests that there may be an outward movement of sediment and ¹³⁷Cs from the 469 center of the wetlands to the riparian area (Zarrinabadi et al., 2023), suggesting that the base ¹³⁷Cs inventory value observed 470 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. ¹³⁷CsA ¹³⁷C inventory of a sediment core can further help in the assignment 471 472 of $assign the {}^{137}Cs$ peak. For example, the {}^{137}Cs peak was repositioned in disturbed sediment cores with higher {}^{137}Cs 473 inventory, where the first discernable peak after the sharp rise from the onset of 137 Cs activity and exceeding or around the 474 reference value was assumed to be the original ¹³⁷Cs peak. ²³⁹⁺²⁴⁰Pu isotopes, like ¹³⁷Cs, are a product of nuclear testing and 475 can be used to identify the peak of 137 Cs. Future research will use $^{239+240}$ Pu to replace 137 Cs as 137 Cs levels diminish.

476 **4.2** Challenges in interpreting ¹³⁷Cs and ²¹⁰Pb profiles

477 Mobilization of ¹³⁷Cs and ²¹⁰Pb in the sediment often occurs in wetlands. Vertical mixing of ¹³⁷Cs within sediments can be 478 caused by remobilization and redistribution by wind and water, ice movement and inversion, disturbance by animals, and 479 disturbance by humans that ditch and drain the wetlands, till through the wetland when it is dry_{τ} and let cattle access them for water which cause disturbances to the bottom sediments (Anderson Robbins et al., 1987; Lobb et al., 1995) Wilan 480 481 et al., 1995; Jagercikova Takahashi et al., 2015). Vertical mixing affects the profile by attenuating the peak upward and 482 downward (which we addressed using the ¹³⁷Cs inventory value and not just the peak when assessing the profile). Horizontal 483 mixing of ¹³⁷Cs dating within sediment occurs by physical movement of sediments into or out of the wetland, causing 484 uneven distribution of the OC content, where accumulation may be high at the edges of open water of the wetland (Lobb et 485 al., 2011; Zarrinabadi et al., 2023). This heterogeneity can be caused by horizonal the horizontal focusing of sediments in 486 sub-basins within a wetland, i.e., multiple center points. Sampling multiple sediment cores from individual wetlands can help 487 capture the heterogeneity within the wetland. HSuppose the 137Cs activity of most of the sediment cores from an individual 488 particular wetland are is noisy with a higher inventory value, then (e.g., 137Cs profile of S-LO-I-W4-T2-CW-R2 in 489 Supplementary Fig. 2a). In that case, the impact by erosional processes can be deduced with higher certainty because the. 490 The higher observed inventory value could be a result offrom the movement of enriched material via erosion/lateral flow to 491 the center of the wetland, therefore increasing the quantity number of ¹³⁷Cs from the value that would be expected if no new 492 enriched material was introduced via erosion/lateral flow. In this study, the assumption of no substantial downward mixing 493 of 137 Cs was supported by (1) sampling three cores from each wetland, (2) assessing the sharpness of the rise of the peaks (a 494 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. 495

496 **4.3** ¹³⁷Cs vs. ²¹⁰Pb derived OC sequestration rates and stocks

497 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 in-closer proximity 498 499 withto the 1:1-line, suggesting compatibility between 137 Cs- and 210 Pb-based estimates- (Fig. 5c and 5d). Conversely, the 500 ¹³⁷Cs-²¹⁰Pb O-O plot of the 1954 OC sequestration rates showed more deviation from the 1:1 line; ¹³⁷Cs-based OC 501 sequestration rates were more dispersed and were higher than the 210 Pb-based OC sequestration rates, (Fig. 5a and 5b). The 502 mean OC sequestration rates in Table 2 further verify the comparability of OC sequestration rates using the 1963 time-503 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 504 sequestration rate is $0.67 \text{ Mg ha}^{-1} \text{ yr}^{-1}$). Providing better sequestration rate estimates has consequences for estimating OC 505 506 stocks with an improved degree of accuracy, which may provide policymakers with better tools to make informed carbon 507 management decisions supported with data.

508

509 To put our findings ininto practice and in the widerbroader OC sequestration perspective, we consider a scenario where two 510 independent studies were performed using 137Cs and 210Pb (with the CFCS model) at the same exact locations. If the cores 511 were not selected based on the criteria we used to selectchoose high-quality profiles, then these two studies' OC 512 sequestration rate estimates are likely to disagree. However, we know and have demonstrated through our findings that they 513 are linearly dependent, and the equation of our linear regression lines may be used to transform one estimate to the other. 514 Consequently, if the cores were selected based on the criteria of our selection criteria, then one can expect the OC 515 sequestration rate estimates to have similar values, which alleviates the interpretation challenges of having two different 516 estimates from two independent studies. This observation may help with consistency when disagreements in estimates are 517 observed. Another practical application of our findings may be in data augmentation. For example, if we have ²¹⁰Pb data for 518 a set of locations and 137 Cs data for other locations, the linear regression equation could be used to transform 210 Pb data to 519 augment ¹³⁷Cs data, and vice versa. This can help data-driven modelling approaches where, whereas larger datasets help-to 520 achieve robust modelling tools. Similarly, because OC stocks can be derived from sequestration rates for specific years, 521 estimates derived using one radioisotope can be used to estimate OC from a dataset derived from another estimate, further 522 contributing to the augmentation of the corresponding OC stock data.

523

Based on the results of this study, we recommend (1) using high-quality 137Cs and 210Pb profiles to estimate OC

525 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,

527 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.

532 **5** Conclusions

Information regarding OC sequestration rates within freshwater mineral soil wetlandstemperate 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.

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538 While estimations based on the onset of ¹³⁷Cs in 1954 or its peak in 1963 were reasonable, estimates anchored to the 1963 539 peak of ¹³⁷Cs exhibited closer alignment with those derived from ²¹⁰Pb data (using the CFCS model). These findings suggest 540 that estimates derived from both ¹³⁷Cs and ²¹⁰Pb radioisotope dating methods are interchangeable and reasonably comparable 541 when utilizing the 1963 ¹³⁷Cs time-marker.

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The combined use of Combining ¹³⁷Cs and ²¹⁰Pb tracers provides a comprehensive assessment of sedimentation rates. While one tracer offers an average rate of sedimentation rate 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. 547 Code and data availability. The R code for the distance sampling modelling along with the data to run the code is available 548 at https://doi.org/10.5281/zenodo.10951658. The organic carbon (OC) sequestration rates data used to check the 549 comparability of the radioisotope profiles can be found in the Supplement. The radioisotope profiles used for screening can 550 be foundare in the paper and Supplement. Other The paper and Supplement present other relevant data to support our 551 conclusion-are presented in the paper and/or Supplement.

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Author contributions. The authors' contributions of authors 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.

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

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