1	How much organic carbon have UK lakes stored in the Holocene?
2	A preliminary estimate
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15	Abstract:
16	Temperate lake sediments store a substantial amount of organic carbon (OC) over millennia.
17	Despite the importance of quantifying terrestrial carbon budgets for nature-based solutions for
18	climate change mitigation, the long-term accumulation of OC in European temperate lakes is
19	poorly constrained. In this study, we analyzed 30 lake sediment records to generate a preliminary
20	first-order estimate of Holocene OC accumulation rate (OCAR) and OC storage in UK lakes. We
21	also examined the environmental variables that influence OCAR and produced synthesized
22	Holocene records of %OC and z-scores of log-transformed OCAR and sediment accumulation rate
23	(SAR) at 500-year resolution. Based on our estimation, we report an average Holocene OCAR of

 7.4 ± 5.5 g C m⁻² yr⁻¹ and a Holocene total OC storage of 0.24 ± 0.18 Pg C in UK lakes. Apart 24 25 from latitude, no relationship was found between the average Holocene OCAR and the various 26 environmental variables (i.e. temperature, precipitation, surface area, catchment area, depth, 27 altitude and geology type). During the Holocene, OCAR closely resembles variations in SAR, whereas the increase in %OC is largely explained by the warming climate. Early Holocene 28 29 variations in OCAR are primarily climate-driven, whereas the anthropogenic impact on the landscape exerted a predominant influence on OC burial during the middle-late Holocene. Our 30 31 results improve the current understanding of terrestrial carbon budgets in the UK and demonstrate 32 the under-appreciated importance of lakes as long-term OC stores as compared to ponds and woodlands. 33

34

35 Keywords:

36 Organic carbon burial, Terrestrial carbon budgets, Lake sediments, Temperate lakes, Holocene,
37 UK, Paleolimnology

38

39 Introduction

In the context of anthropogenic climate change, there is an unparalleled need to achieve carbon
neutrality by this century. Nature-based solutions (NbS) comprise a crucial component of climate
change mitigation due to the large amount of carbon stored in natural and managed ecosystems.
NbS has the potential to mitigate CO₂ emission at a rate of 10 Gt CO₂ yr⁻¹ and, in the long-term,
reduce peak warming by 0.1-0.3°C depending on the different warming scenarios (Girardin et al.,
2021). Quantifying terrestrial carbon budgets is conducive to implementing NbS and could help

guide more targeted and informed land management decisions (Cole et al., 2007; Gregg et al., 2021; Keith et al., 2021).

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49 Despite only constituting 3.7% of the Earth's non-glaciated land surface area (Verpoorter et al., 50 2014), lakes are highly spatially variable and play a critical role in the global carbon cycle (Einsele 51 et al., 2001; Prairie, 2008; Tranvik et al., 2009; Buffam et al., 2011; Radbourne et al., 2017). Lake ecosystems are sites where active processing of carbon (e.g. carbon production, transformation, 52 53 burial, and evasion) takes place (Cole et al., 2007; Prairie, 2008; Tranvik et al., 2009). Allochthonous input of organic matter from catchments contributes to the lake dissolved organic 54 carbon (OC) or dissolved organic matter pool, which can be assimilated by heterotrophs as an 55 56 energy source (Prairie, 2008; Hanson et al., 2015). Dissolved inorganic carbon, mainly in the form of CO₂, is consumed during photosynthetic assimilation by heterotrophs and oxidised into organic 57 carbon in both dissolved and particulate forms (McGowan et al., 2016). OC may be reduced back 58 59 into CO₂ and CH₄ through respiration. Particulate carbon suspended in the water column may sink to the lake bottom and contribute to OC burial in lake sediments (Alin and Johnson, 2007). 60

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62 Although lakes mostly behave as net carbon sources and are super-saturated in CO₂ in the surface 63 waters (Cole et al., 1994; Tranvik et al., 2009), lake sediments can store a large amount of OC in 64 the long-term due to their high OC burial efficiency, rapid sediment accumulation rates and high 65 aquatic productivity (Cole et al., 2007; Sobek et al., 2009). Some of the deeper and larger lakes of 66 tectonic origin could persist in the landscape for millions of years, and lake sediments in glaciated 67 terrains could accumulate for more than 10,000 years (Alin and Johnson, 2007; Cole, 2013). In 68 contrast, many smaller lakes are short-lived or artificially constructed (i.e. reservoirs). Globally,

69 inland waters (including lakes, rivers, streams and reservoirs) receive carbon at the magnitude estimated at 2.9 Pg C yr⁻¹, amongst which 1.4 Pg C yr⁻¹ are evaded into the atmosphere, 0.9 Pg C 70 yr⁻¹ are transported to the sea, and the remaining 0.6 Pg C yr⁻¹ are buried to sediments (Tranvik et 71 72 al., 2009). The total amount of carbon storage in lake sediments during the Holocene is estimated at 400-800 Pg C (Cole et al., 2007), and the long-term OC accumulation rate (OCAR) ranges 73 between 4.5-14 g C m⁻² yr⁻¹ (Dean and Gorham, 1998; Stallard, 1998; Cole et al., 2007). Recently, 74 OCAR in lake sediments has increased due to cultural eutrophication (Anderson et al., 2014; 75 Anderson et al., 2020). Carbon burial is expected to continue to increase, driven by climate change, 76 77 increased runoff and eutrophication (Tranvik et al., 2009).

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79 Lake sediments document spatially and temporally integrated information. Hence, 80 paleolimnological approaches are well-suited for quantifying OC accumulation and storage in lakes and may offer critical insights for understanding changes in the terrestrial carbon cycle in the 81 82 future (McGowan et al., 2016). One widely adopted method uses loss-on-ignition 550°C (%LOI550) data to estimate %OC in lake sediments (Pajunen, 2000; Anderson et al., 2014) or directly makes 83 84 use of downcore total organic carbon (TOC) data measured using an elemental analyzer 85 (McGowan et al., 2016). To date, many studies have adopted a paleolimnological approach for 86 quantifying the amount of OC storage and OCAR in lakes at different spatial and temporal scales 87 (e.g. Dean and Gorham, 1998; Kastowski et al., 2011; Anderson et al., 2020). However, there is a 88 bias towards Holocene scale estimates on boreal and arctic lakes within Europe (e.g. Kortelainen 89 et al., 2004; Anderson et al., 2009; Chmiel et al., 2015), whereas temperate lakes, which are large organic matter stores, receive less attention (Scott, 2014). Temperate lakes cover ~25% of the 90

European lake surface area but contribute to ~35% of European lakes' total carbon accumulation rate (Scott, 2014).

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94 The UK is home to 43,738 water bodies covering 213,911 ha in Great Britain (Hughes et al., 2004) 95 and 1,668 water bodies covering 62,600 ha in Northern Ireland (Gibson et al., 1994), altogether 96 constituting around 1% of the total UK surface area. There are few systematic inventories of OCAR in the UK. Scott (2014) examined the OCAR of two lakes in the Shropshire-Cheshire 97 meres region, and Anderson et al. (2014) included a number of UK lakes in their OCAR estimation 98 99 for European lakes. Both studies focused on the last 100-150 years. Only a few UK lakes were 100 included in the estimation of Holocene OCAR in European lakes (Kastowski et al., 2011). Thus, 101 there is no clear overview of Holocene OCAR and OC storage estimates in the UK. There are 102 extensive lake sediment records in the UK covering part or the complete sequence of the Holocene (e.g. Bennett et al., 1992; Fossitt, 1996; Watkins et al., 2007), offering an excellent opportunity to 103 104 conduct a Holocene OC inventory.

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Here, we present a first-order preliminary estimate for OCAR and OC storage in UK lakes across the Holocene by adopting a paleolimnological approach. In addition, we investigate the temporal variations in OCAR, sediment accumulation rate (SAR) and %OC during the Holocene and examine the variables influencing OCAR in UK lakes (i.e. temperature, precipitation, surface area, catchment area, depth, altitude, latitude and geology type). By comparing the OCAR in UK lakes to estimates from lakes in different regions and various habitat types in the UK, we evaluate the relative importance of lake sediments as an OC store in the UK.

114 Methods

115 Study sites and data collection

The selection of lake sediment cores was based on four main criteria: (1) the sediment record covers at least 3 kyr during the Holocene; (2) a %LOI₅₅₀, TOC, or organic matter (OM) record exists; (3) a minimum of three radiocarbon dates to ensure a relatively reliable age control; and (4) extant lakes.

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121 A wide range of search terms was employed to identify relevant studies. These include "lake", "loch", "llyn", "lough", "Holocene", "Britain", "England", "Scotland", "Wales", "Northern 122 Ireland", "organic matter", "LOI", "TOC", "vegetation", and "pollen". We are aware that this 123 124 search was not exhaustive, and additional sediment records must exist in earlier publications or 125 remain unpublished. However, in contrast to other Holocene estimates for lakes (e.g. Anderson et 126 al., 2009, n=11; Chmiel et al., 2015, n=7), fjords (e.g. Smeaton et al., 2017, n=5) and peatlands (e.g. Ratcliffe et al., 2018, n=12), the number of lakes included is sufficient for a preliminary first-127 128 order estimate.

129

A total of 30 lake sediment records across the UK were selected (Figure 1). This includes three new lake sediment records (Loweswater, Loch na Claise and Loch an Aigeil) that were analyzed for %LOI₅₅₀ following Heiri et al. (2001), which entails burning at 550°C for 4 hours and calculating using Equation (1). %LOI₅₅₀ data for the 27 published records were generated using a temperature range between 450-550°C and furnace times of 1-12 hours (references are in SI). Scottish lakes (n=19) comprise most of the selected records, followed by lakes in England (n=6), Wales (n=3) and Northern Ireland (n=2). The proportional representation of lakes from each country is roughly consistent with the spatial distribution of lakes in the UK, dominated by lakes
in Scotland (57%) and England (36%) in terms of absolute numbers (Gibson et al., 1994; Hughes
et al., 2004).

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141
$$\% LOI_{550} = 100 \left(\frac{(DW_{105} - DW_{550})}{DW_{105}} \right)$$

142

(Equation 1)

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144 Chronology

To obtain Holocene temporal trends in OCAR, SAR and %OC, radiocarbon dates for all 30 lake records were (re)calibrated with the IntCal20 curve (Reimer et al., 2020) using Bayesian age-depth modelling in the Bacon package (version 2.5.8) (Blaauw and Christen, 2011) in R (see Figures S1-S30 for the new age models and Tables S1-S3 for radiocarbon ages from the three new records). The 95% confidence intervals from the Bacon age-depth models were used as conservative age uncertainties. The maximum age uncertainties across all interpolated depth layers ranged from 289 years (Lochan Uaine) to 1,705 years (Loch na Claise), with 77% of the lake records having maximum age uncertainties below 1,000 years.

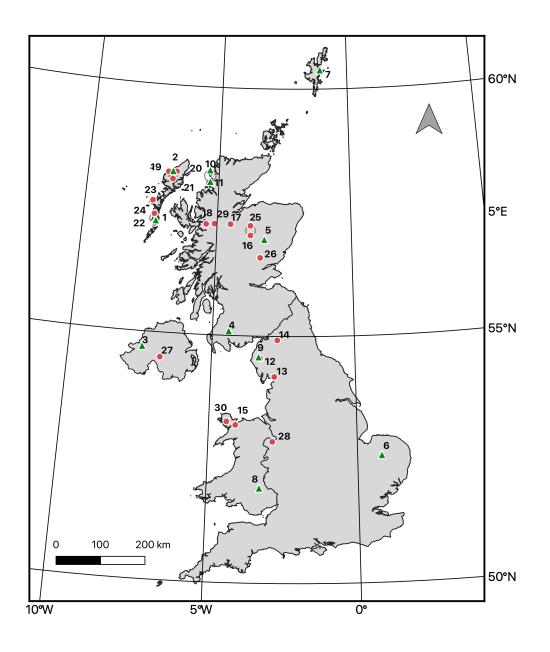


Figure 1. Overview map showing the 30 study sites. Green triangles (n=11) represent lakes with reported dry bulk density data (required to calculate organic carbon accumulation rate); red circles (n=19) represent lakes lacking dry bulk density data. List of lake names, published reference sources and further information associated with each lake are given in the supplementary table.

158 Calculation of average Holocene OCAR and OC storage

159	%LOI550 was multiplied by a factor of 0.469 to convert it into %OC based on previous comparisons
160	of %LOI $_{550}$ and %OC (Dean, 1974). Most of the published literature (n=19) did not report data on
161	dry bulk density (DBD) (Figure 1), but DBD and SAR are required to calculate dry mass
162	accumulation rate (DMAR), which is essential in the calculation of OCAR (see below). Following
163	Dean and Gorham (1998), an empirical relationship between %LOI550 and DBD was established
164	using data from the 11 lakes with DBD reported (Figure 1) (Equation 2), which has $r^2=0.6$ and
165	$p < 2.2*10^{-16}$. This %LOI ₅₅₀ -DBD relationship is highly similar to existing empirical relationships
166	(Dean and Gorham 1998; Moyle et al. 2021) (see Figure S31).

- 167
- 168
- $DBD = 2.4355 \% LOI_{550}^{-0.827}$ (Equation 2)
- 170

This relationship was applied to the estimation of DBD for the 19 lakes. OCAR (g C m⁻² yr⁻¹) was
determined by multiplying DMAR (g cm⁻² yr⁻¹) by %OC and a conversion factor of 10000
(Anderson et al., 2014).

174

To account for the higher-than-average SAR occurring at the deepest part of the lake (Engstrom and Rose, 2013; Anderson et al., 2014), a sediment focusing factor based on the ratio of sediment ²¹⁰Pb flux to regional atmospheric ²¹⁰Pb flux was applied. Due to the paucity of sediment ²¹⁰Pb flux data for most study sites, we employ a regional average focusing factor of 1.78 based on data from 90 UK lakes since 1850 CE (Handong Yang, unpublished data). Sediment focusing varies over time as the pattern of deposition evolves and lake morphometry or size changes (Engstrom and Rose, 2013). However, we assume that the focusing factor remains constant over the Holocene 182 for ease of estimation. The focusing-corrected average Holocene OCAR in each lake sediment record was then averaged to estimate the average focusing-corrected Holocene OCAR in UK lakes. 183 184 The total Holocene OC storage in UK lakes was calculated by multiplying the average focusing-185 corrected Holocene OCAR by 11,650 years and a total UK lake area of 276,511 ha (Gibson et al., 186 1994; Hughes et al., 2004). Standard deviations were calculated for the OCAR in each sediment record and propagated to obtain uncertainty of the average Holocene OCAR in UK lakes. We 187 recognize that there are compounding error sources associated with each step of the estimation, 188 189 but the reported error values represent a conservative estimate of the uncertainty, which means 190 that the true value lies within the range provided.

191

192 Examining controlling factors on OCAR

193 Pearson's product-moment correlation was performed to determine the relationship between the 194 average Holocene OCAR from each sediment record and various modern environmental variables at each site, assuming that OCAR and these variables have remained constant over time. Data for 195 196 surface area, catchment area, altitude, mean depth, maximum depth, geology type (categorized 197 into low, moderate and high alkalinity groups) and latitude were obtained from the UK Lakes 198 Portal (UK Centre of Hydrology and Ecology, 2016) and the original publications; the maximum 199 annual temperature and mean annual precipitation between 1961-2020 at the nearest weather 200 station for each site were obtained from the Met Office (2022). Spearman's rank correlation 201 coefficient was performed to analyze the relationship between OCAR and the three geology types.

202

203 Average UK Holocene temporal trends

204 The OCAR, SAR and %OC data for each lake were linearly interpolated between neighbouring 205 data points to the nearest 500-year since the sampling resolution of all sediment cores lies below 206 500-year. The OCAR and SAR data were log-transformed for normality (hereafter referred to as 207 OCAR_{log} and SAR_{log}). For each sediment record, z-scores of OCAR_{log} and SAR_{log} were calculated 208 by subtracting the mean and dividing by the standard deviation. Synthesized OCAR_{log} and SAR_{log} 209 z-scores were produced by taking the average z-scores for all lakes at each resampled age (i.e. one 210 average value every 500 years). Pearson's product-moment correlation was performed to assess 211 the relationship between the synthesized OCAR_{log} z-score and the synthesized SAR_{log} z-score 212 and %OC.

213

214 **Results**

215 Average Holocene OCAR and OC storage estimates

The average focussing-corrected Holocene OCAR in UK lakes is 7.4 ± 5.5 g C m⁻² yr⁻¹ (uncorrected OCAR: 13.2 ± 9.9 g C m⁻² yr⁻¹), with values ranging from 2.5 ± 0.9 g C m⁻² yr⁻¹ for Loch Buailaval Beag to 20.3 ± 8.9 g C m⁻² yr⁻¹ for Lough Catherine (Figure 2). The area-weighted OCAR is 21 ± 15 Gg C yr⁻¹ (rounded to 2 significant figures). During the Holocene, the total OC storage in UK lakes is approximately 0.24 ± 0.18 Pg C with areal OC storage of 86.5 ± 64.6 Kg C m⁻².

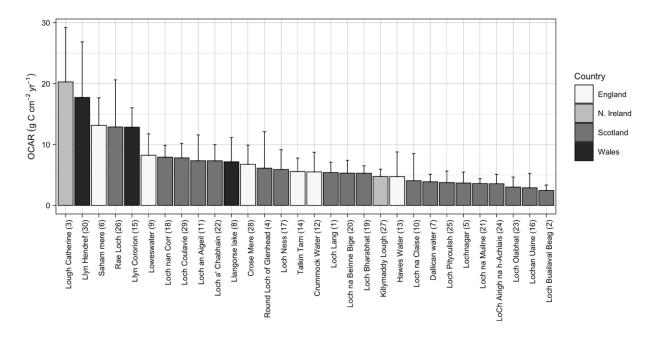


Figure 2. Focusing-corrected average Holocene organic carbon accumulation rate (OCAR) (g C $m^{-2} yr^{-1}$) in each lake sediment record.

222

226 Relationship between OCAR and environmental variables

The only variable that significantly correlates with the average Holocene OCAR from each sediment record is latitude (r=-0.5, $p=3.2*10^{-3}$). No significant correlations were found between OCAR and any other of the variables tested (see Supplementary Information).

230

231 Average UK Holocene temporal trends

232 Since 12 cal kyr BP, %OC has increased rapidly from ca. 6.8 to 17.3%, stabilizing after 8 cal kyr

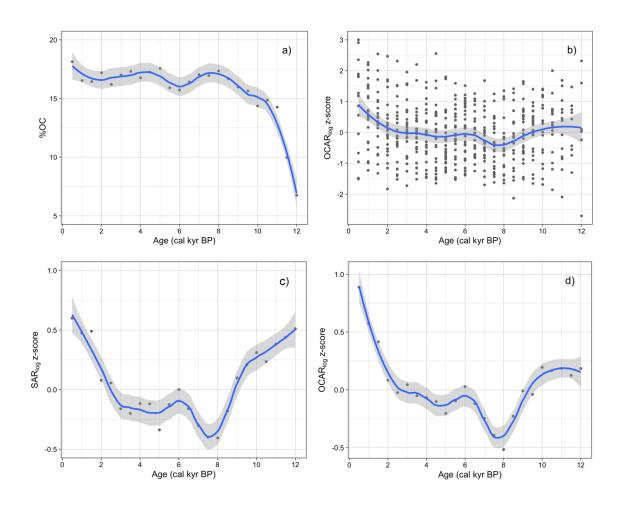
BP around 16-17% with a gradual increase (Figure 3a) to present-day values of 18.1%. The

234 OCAR_{log} z-score remains stable between 12-10 cal kyr BP and subsequently declines between 10-

- 235 8 cal kyr BP (Figure 3b,d). In contrast, the SAR_{log} z-score displays a more extended decline
- between 12-8 cal kyr BP (Figure 3c). Both records display a long-term increase from 8 cal kyr BP
- to the present, interrupted by a temporary decline between 6-5 cal kyr BP. After 3 cal kyr BP,

OCAR_{log} z-score lies predominantly above the average (i.e. when z-score=0), which is greater than
the early and middle Holocene.

240



241

Figure 3. Average Holocene temporal trends for 30 UK lakes with LOESS smoothing (95%
confidence interval, span=0.4) at 500-year resolution: (a) percentage organic carbon (%OC); (b)
raw log-transformed organic carbon accumulation rate (OCAR_{log}) z-score showing data points
from all sediment records; (c) synthesized log-transformed sediment accumulation rate (SAR_{log})
z-score; (d) synthesized OCAR_{log} z-score.

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A very strong correlation exists between the OCAR_{log} and SAR_{log} z-scores (r=0.9, p=1.8*10⁻⁸). An r² of 0.8 suggests that SAR_{log} can explain up to 80% of the variation in OCAR_{log}. On the contrary, %OC displays no relationship with the OCAR_{log} z-score (r=-0.2, p=0.5).

252 **Discussion**

253 Average Holocene OCAR and OC storage estimates

The total Holocene OC storage for UK lakes is estimated to be 0.24 ± 0.18 Pg C (areal OC storage: 254 255 $86.5 \pm 64.6 \text{ Kg C m}^{-2}$), equivalent to $0.88 \pm 0.66 \text{ Pg CO}_2$. The magnitude of the average Holocene OCAR of UK lakes at 7.4 \pm 5.5 g C m⁻² yr⁻¹ is in good agreement with the global range for lakes 256 between 4.5-14 g C m⁻² yr⁻¹ (Dean and Gorham, 1998; Stallard, 1998; Cole et al., 2007), and is 257 higher than the average Holocene OCAR estimates for lakes in Europe (5.2 g C m⁻² yr⁻¹) 258 (Kastowski et al., 2011), southwest Greenland (6 g C m⁻² yr⁻¹) (Anderson et al., 2009), Finland 259 (1.6 g C m⁻² yr⁻¹) (Kortelainen et al., 2004), and Sweden (3.2 g C m⁻² yr⁻¹) (Chmiel et al., 2015). 260 Variations in OCAR in Europe are likely due to region-specific lake characteristics. For example, 261 lower temperatures in Finland may suppress primary production, thus leading to lower OCAR in 262 263 boreal lakes (Kortelainen et al., 2004). In contrast, the higher OCAR in southwest Greenland may 264 be explained by the selection of very small lakes in the study (range: 0.05-0.73 km²) (Anderson et 265 al., 2009) compared to the selection of larger Finnish lakes (average area: 122 km²) (Kortelainen 266 et al., 2004) because OCAR is often found to be inversely related with lake area (Mulholland and 267 Elwood, 1982; Pajunen, 2000).

268

Within habitats of the UK, the Holocene OCAR of lakes $(7.4 \pm 5.5 \text{ g C m}^{-2} \text{ yr}^{-1})$ is higher than the modern estimates for broadleaf and mixed woodlands (4.2 g C m⁻² yr⁻¹, range: 0.7-12 g C m⁻² yr⁻¹), coniferous woodlands (4.9 g C m⁻² yr⁻¹, range: 0.8-11.7 g C m⁻² yr⁻¹) and grasslands (2.2 g C m⁻² yr⁻¹) (Downing et al., 2008), but lower than the Holocene estimates for fjords (8.3 ± 2.0 g C m⁻² yr⁻¹) (Smeaton et al., 2017), peatlands (17.8 ± 4.9 g C m⁻² yr⁻¹) (Ratcliffe et al., 2018), the modern estimate for ponds aged between 18-20 years ($142 \pm 19 \text{ g C m}^{-2} \text{ yr}^{-1}$) (Taylor et al., 2019) and millennial-scale estimates for seagrasses ($138 \pm 38 \text{ g C m}^{-2} \text{ yr}^{-1}$) and salt marshes ($218 \pm 24 \text{ g C m}^{-2} \text{ yr}^{-1}$) (Mcleod et al., 2011) (Figure 4a). The area-weighted OCAR for UK lakes ($21 \pm 15 \text{ Gg C yr}^{-1}$) is lower than all UK habitats apart from seagrasses (Figure 4b) due to the small total surface area of lakes in the UK compared with other habitats.

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280 In comparison to most terrestrial habitats, vegetated coastal habitats exhibit very high OCARs owing to their high efficiency in trapping sediments from both riverine and oceanic sources, 281 282 facilitated by their complex ecosystem structure (Mcleod et al., 2011). Peatlands and ponds also 283 exhibit higher OCARs than lakes. Peatlands are highly efficient OC sinks due to active plant 284 growth (Loisel et al., 2014). Ponds are generally very productive and are effective in trapping and 285 preserving carbon in sediments (Downing et al., 2006; Cole et al., 2007). The higher OCAR in lakes compared to woodlands and grasslands is due to lakes' higher OC preservation rate (Cole et 286 287 al., 2007).

288

289 The OC turnover rates in ponds or forest biomass last for decades to centuries (Cole, 2013). The 290 long-term storage of OC can only occur in soils once forests reach maturity, and yet, the long-term 291 OC burial in soil is much limited compared to its instantaneous carbon assimilation rate due to 292 periodic burning and/or high decay rates (Prairie, 2008). Moreover, despite having very high 293 OCAR, ponds are ephemeral features of the landscape that is more susceptible to in-filling and 294 desiccation compared to lakes. In contrast, lake sediments, peatlands and coastal habitats can store 295 OC for millennia (Cole et al., 2007; Prairie, 2008; Mcleod et al., 2011; Ratcliffe et al., 2018; Taylor 296 et al., 2019). In particular, large and deep lakes in active tectonic regions can store OC for up to

millions of years (e.g. Lake Malawi storing 5,500 Pg C) (Alin and Johnson, 2007). Consequently,
although the average Holocene OCAR of lakes is modest in contrast to various habitat types in the
UK, the capacity of lakes to act as a long-term carbon store cannot be overlooked.

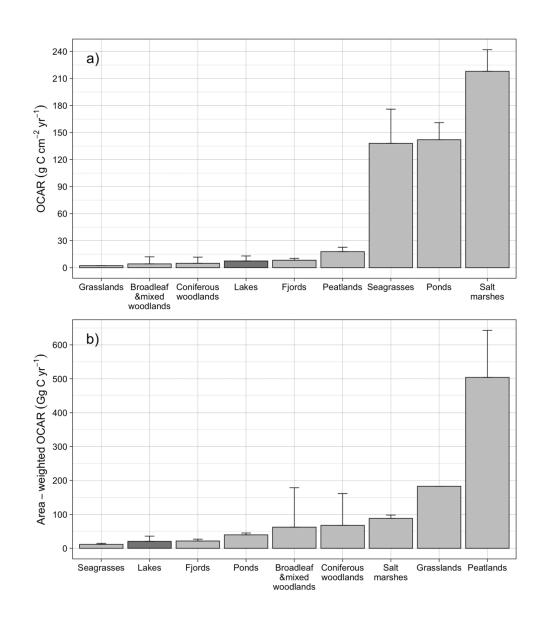


Figure 3. Comparison of (a) average Holocene organic carbon accumulation rate (OCAR) and (b)

- area-weighted Holocene OCAR in a range of habitats in the UK. Sources of OCAR data include
 Mcleod et al. (2011) for seagrasses and salt marshes; Smeaton et al. (2017) for fjords; Taylor et al.
- 305 (2019) for ponds; Downing et al. (2008) for woodlands and grasslands and Ratcliffe et al. (2018)
- 306 for peatlands. Data for habitat area used for calculating area-weighted Holocene OCAR was

obtained from Gibson et al. (1994) and Hughes et al. (2004) for lakes; Taylor et al. (2019) for
ponds; Carey et al. (2008) for woodlands, grasslands and peatlands; Smeaton et al. (2017) for
fjords; Phelan et al. (2011) for salt marshes and Green et al. (2021) for seagrasses.

311 The estimation of Holocene OC storage and accumulation rate in UK lakes provides an important 312 constraint on the regional terrestrial carbon budget. Using the past as an analogue, our results are 313 highly relevant for understanding how carbon budgets in lakes will change in the future (Hanson 314 et al., 2015; McGowan et al., 2016). Moreover, despite the promising potential of NbS for climate 315 change mitigation, there is currently a bias towards carbon flows (e.g. sequestration) and an 316 underappreciation of carbon storage in carbon accounting (Keith et al., 2021). While both processes are essential for the ecosystem service of climate regulation, the overlooking of carbon 317 318 storage hampers current NbS efforts by providing incomprehensive information about the 319 mitigation benefits, which may result in perverse outcomes in NbS implementation (Keith et al., 320 2021). Hence, by quantifying the long-term carbon storage in lake sediments in the UK, we provide 321 critical information that may contribute to a more comprehensive carbon accounting of terrestrial 322 aquatic systems beneficial for facilitating better informed NbS practices and associated land management decisions. Despite the long-term mitigation benefits NbS may offer, we stress that 323 324 these benefits are limited compared to decarbonisation efforts (Girardin et al. 2021), therefore, 325 NbS should not act as a distraction from emission reduction activities in the near term.

326

327 Controlling factors on OCAR

Previous studies have identified a wide range of environmental variables controlling variations in
the average Holocene OCAR from each lake record. The main factors include depth (Anderson et
al., 2014), surface area (Mulholland and Elwood, 1982; Pajunen, 2000; Downing et al., 2008), lake
morphometry (Ferland et al., 2014), precipitation (Tranvik et al., 2009), temperature (Gudasz et

332 al., 2015; Rantala et al., 2016), nutrients (Anderson et al., 2020), oxygen exposure time and 333 mineralization (Sobek et al., 2009). Here, we found a moderate relationship between OCAR and 334 latitude, which may suggest an indirect influence of cooling as latitude increases (Kastowski et al., 335 2011). The absence of a relationship between OCAR and the environmental variables we tested 336 may suggest the relationships found in previous studies may not apply universally. Moreover, none 337 of the variables alone shape variations in OCAR between lakes, but rather, a combination of many 338 site-specific factors. However, it is beyond the scope of this study to explain variations between 339 lakes. Besides, with over 67% of the studied lakes ≤ 10 ha, the absence of a relationship between OCAR and the surface area might be due to the bias towards very small lakes with variable OCAR 340 yet narrow environmental gradients (Kastowski et al., 2014). 341

342

343 Average UK Holocene temporal trends

344 The strong positive correlation between the synthesized OCAR_{log} and SAR_{log} z-scores indicates 345 that variations in OCAR_{log} are largely driven by SAR_{log}, and %OC of the sediment has not changed markedly during the Holocene. The increase in %OC during the early Holocene (12-8 cal kyr BP) 346 347 likely resulted from the increasing annual temperature in Europe (Figure 5a,b) (Davis et al., 2003; Lang et al., 2010), which facilitated the switch from late glacial minerogenic sediment to organic 348 349 gyttja as catchment soil developed (Bennett et al., 1990; Snowball and Thompson, 1990; Edwards 350 and Whittington, 1998). The decrease in the synthesized OCAR_{log} and SAR_{log} z-scores prior to 8 cal kyr BP can be explained by the progressive establishment of woodlands fostered by the 351 increasingly warm climate, which stabilizes the soils and reduces runoff and erosion (Figure 5a,f,g) 352 (Bennett et al., 1992; Fossitt, 1996; Watkins et al., 2007). 353

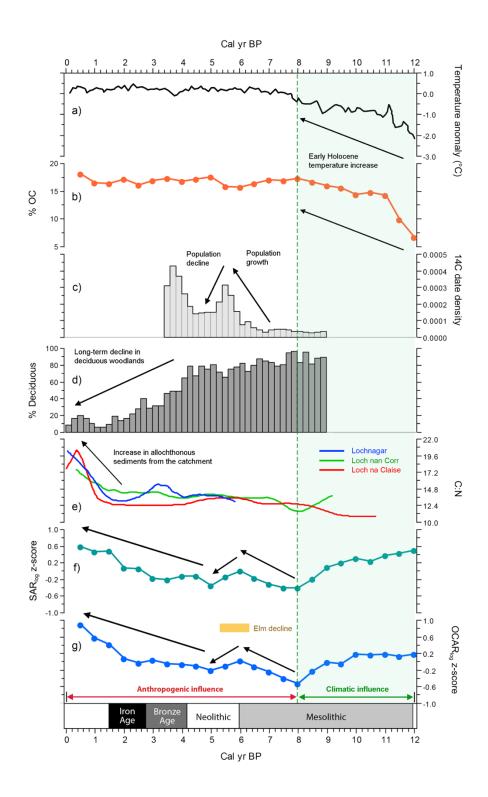


Figure 5. Dominant drivers of Holocene organic carbon accumulation rate (OCAR) and sediment
 accumulation rate (SAR) variations in 30 UK lakes (see Figure 1). (a) pollen-reconstructed annual
 temperature record for Europe (Davis et al., 2003); (b) percentage organic carbon (%OC) at 500 year resolution; (c) radiocarbon (¹⁴C) date probability density functions from archaeological sites

between 9-3.4 cal kyr BP (Woodbridge et al., 2014); (d) pollen-inferred deciduous woodland 359 change since 9 cal kyr BP (Woodbridge et al., 2014); (e) C:N ratios from three study sites (blue: 360 Lochnagar; green: Loch nan Corr; red: Loch na Claise) with LOESS-smoothing (span=0.2) 361 (Dalton et al., 2005; Mackie et al., 2007; Matthews, 2022); (f-g) synthesized log-transformed SAR 362 363 (SAR_{log}) and OCAR $(OCAR_{log})$ z-scores at 500-year resolution. The yellow bar above (g) represents the time range for the elm decline between 6,347-5,281 cal kyr BP based on statistical 364 365 analyses of radiocarbon dates (Parker et al., 2002). The period-based chronology for pre-historic 366 Britain was obtained from Pollard (2008). The green shading separates out intervals where climate 367 exerts more influence on OCAR and SAR variations from periods where anthropogenic impact on 368 the landscape dominates (see text for details).

369

Since 8 cal kyr BP, the long-term increase in the synthesized OCAR_{log} and SAR_{log} z-scores suggests increasing anthropogenic disturbance on the landscape (Figure 5f,g). The initial population increase in Britain started from ca. 7.6 cal kyr BP during the Late Mesolithic, followed by a long-term population growth trend, indicated by the increase in ¹⁴C date densities from archaeological evidence and the sustained decline in deciduous woodlands (Figure 5c,d) (Woodbridge et al., 2014). On the contrary, %OC has remained relatively stable since 8 cal kyr BP, resembling the stabilized trend in temperature (Figure 5b).

377

378 The local maxima in the synthesized OCAR_{log} and SAR_{log} z-scores between 6.5-5.5 cal kyr BP coincides with the widely documented elm decline on the British Isles (6,347–5,281 cal yr BP) 379 (Figure 5f,g) for which human activity is one of the hypothesized causes (Parker et al., 2002). The 380 381 elevated sedimentation and OC burial may have been driven by the transition from Mesolithic 382 hunter-gatherers to Neolithic agriculture in the British Isles between 6.4-6 cal kyr BP (Woodbridge et al., 2014) and the substantial population growth in Britain between 6.1-5.4 cal kyr BP (Collard 383 384 et al., 2010) (Figure 5c), both increasing anthropogenic pressure on the landscape. The increase in the synthesized z-scores is interrupted by a decline towards lower values in both records between 385 5.5-4.5 cal kyr BP. This interval coincides with reduced ¹⁴C date density between 5.3-4.4 cal kyr, 386

indicating reduced Neolithic impact on the landscape, which leads to the re-establishment of
woodlands (Figure 5c,d,f,g) (Woodbridge et al., 2014).

389

Since 5-4 cal kyr BP, the continued increase in the synthesized $OCAR_{log}$ and SAR_{log} z-scores 390 391 suggests enhanced soil erosion, which might be attributed to anthropogenic activities (Van Vliet-392 Lanoë et al., 1992; Edwards and Whittington, 2001). The gradual increase in both z-scores between 4-3 cal kyr BP coincides with the British Bronze Age (2,200-800 BC or 4,150-2,750 BP), whilst 393 the accelerated increase since ca. 3-2.5 cal kyr BP coincides with the British Iron Age (800 BC-43 394 395 CE/500 CE or 2,750-1,907/1,450 BP) (Pollard, 2008). Changes in OCAR and SAR are consistent 396 with the long-term increase in C:N at Loch na Claise, Loch nan Corr and Lochnagar, with an 397 accelerated increase over the last 2 kyr (Figure 5e) (Dalton et al., 2005; Mackie et al., 2007; 398 Matthews, 2022). These suggest an increase in allochthonous sediment input linked to human activities. However, there are limited records of C:N from the studied lakes to help further 399 400 discriminate between allochthonous and autochthonous sources of OC. Compared to natural 401 conditions, anthropogenic sediment delivery can be 5-10 times higher (Dearing and Jones, 2003). 402 Increased land-use pressure, including deforestation, agriculture, and grazing, have all exacerbated 403 soil erosion and increased allochthonous carbon input to lakes (Jones et al., 1985; Beales, 1980; Bennett et al., 1992; Edwards and Whittington, 1998; Mulder, 1999). Soil erosion would also 404 405 elevate nutrient supply into lakes, increasing autochthonous OC accumulation due to increased 406 aquatic productivity.

407

408 Conclusion

409 This study represents a preliminary first-order estimate of Holocene OCAR and OC storage in UK 410 lakes, and we acknowledge a number of limitations, namely, the assumption of a non-411 varying %LOI550-DBD relationship, spatial and temporal extrapolations of OCAR, uncertainties 412 of radiocarbon chronologies and issues of linear interpolation, which could be improved with 413 analyses of new dated sediment cores. Although the OCAR of lakes is modest compared to various 414 coastal and terrestrial habitats in the UK, our results demonstrate the importance of lakes as long-415 term OC stores as compared to ponds and woodlands. While more lake sediment records could be 416 incorporated into the estimation for better spatiotemporal representation of UK lakes, our estimates 417 based on available data improve the current understanding of terrestrial carbon storage in the UK 418 and OC accumulation in European temperate lakes. Our results may contribute important 419 information for understanding how lake carbon budgets would change in the future, as well as for implementing a more comprehensive carbon accounting for better-informed Nature-based 420 421 Solutions and land management practices.

422

Moreover, our results indicate that the first-order response of lake OCAR was to climate amelioration during the early Holocene, initiating organic matter production in terrestrial landscapes and its burial in lake sediments. The second major driver of OCAR since the middle Holocene has been the anthropogenic impact on the landscape, which has resulted in enhanced transport and erosion of OC from the deforested landscapes into lakes and also increased aquatic productivity due to a greater nutrient supply. Further work using geochemical analyses is required to distinguish the type or source of OC that reaches lakes during the middle-late Holocene.

430

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437

438 **References**

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How much organic carbon have UK lakes stored in the Holocene?

A preliminary estimate

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Supplementary information contents:

This file contains age models for the 30 lake sediment records included in this study (Figures S1-S30). These age models were produced using rbacon (Blauuw and Christen, 2011) in R with the radiocarbon dates (re)calibrated against the IntCal 20 curve (Reimer et al. 2020). Please refer to the original publications for the radiocarbon dates. Figure S31 shows the comparison between empirical loss on ignition-dry bulk density relationships derived from this study and previous publications. Tables S1-S3 show the radiocarbon dates for the three new sediment records (Loweswater, Loch an Aigeil and Loch na Claise).

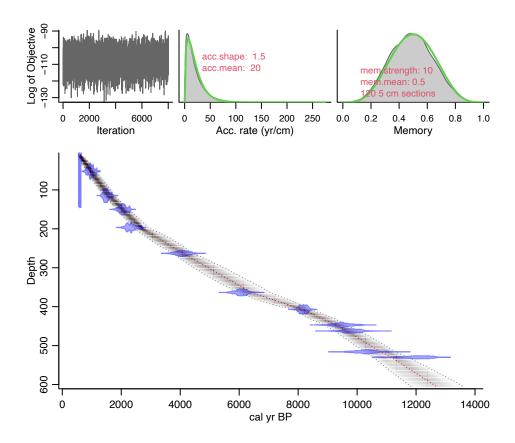


Figure S1. Revised age model for Crose Mere.

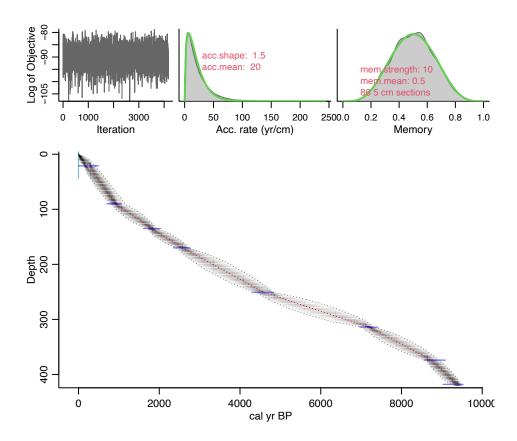


Figure S2. Revised age model for Crummock Water.

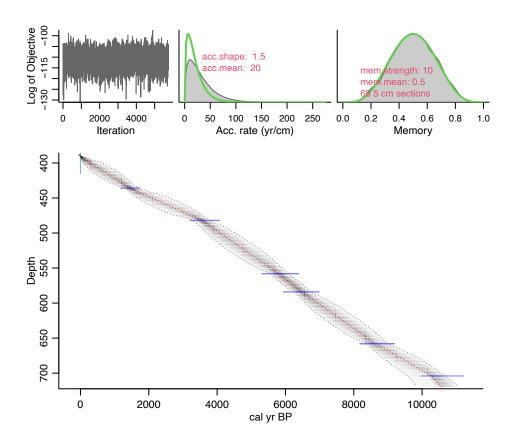


Figure S3. Revised age model for Dallican Water.

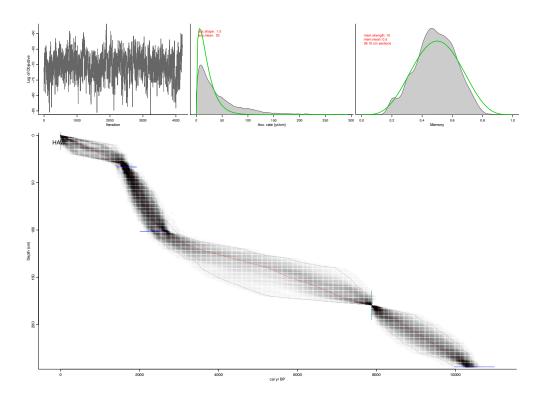


Figure S4. Revised age model for Hawes Water.

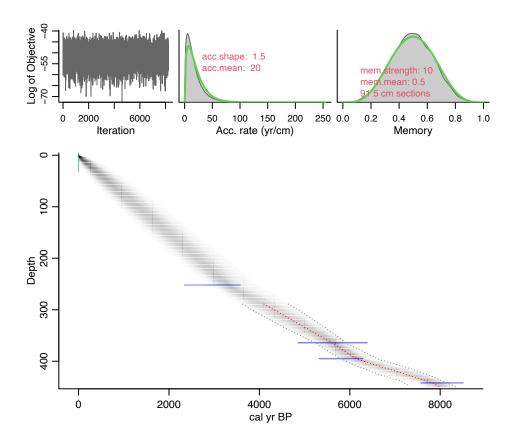


Figure S5. Revised age model for Killymaddy Lough.

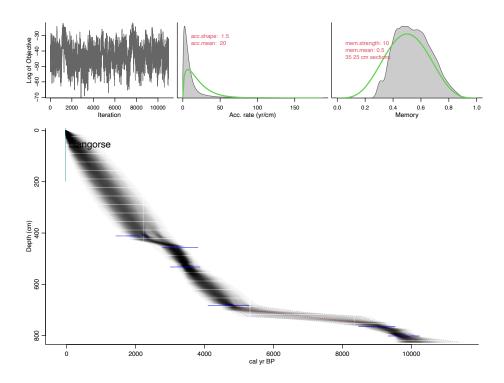


Figure S6. Revised age model for Llangorse Lake.

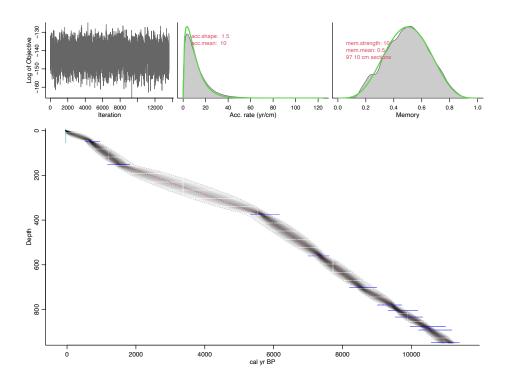


Figure S7. Revised age model for Llyn Cororion.

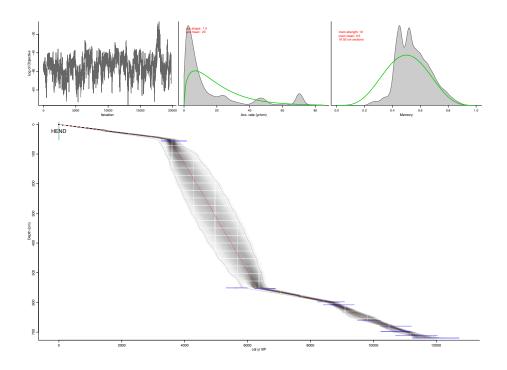


Figure S8. Revised age model for Llyn Hendref.

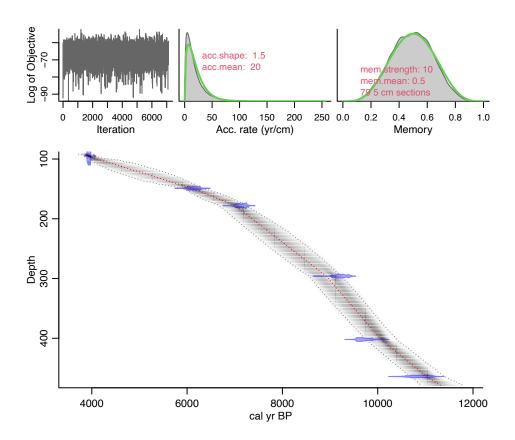


Figure S9. Revised age model for Loch a' Chabhain.

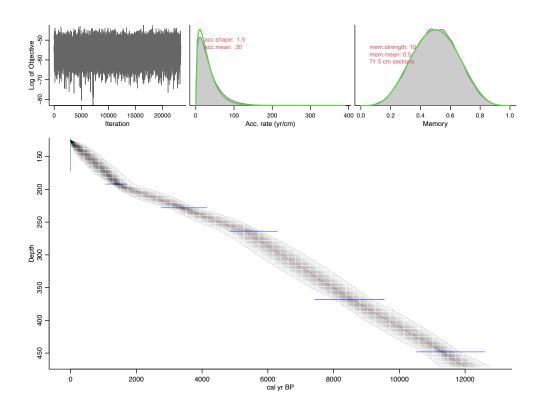


Figure S10. Revised age model for Loch Airigh na h-Achlais.

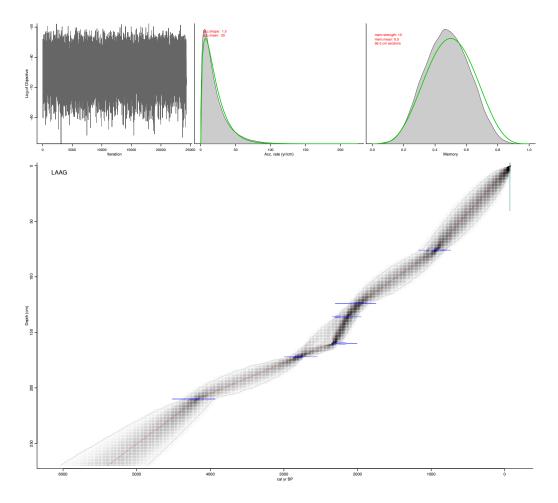


Figure S11. Age model for Loch an Aigeil.

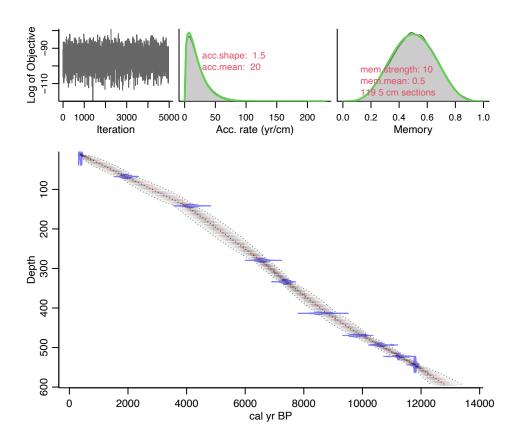


Figure S12. Revised age model for Loch Bharabhat.

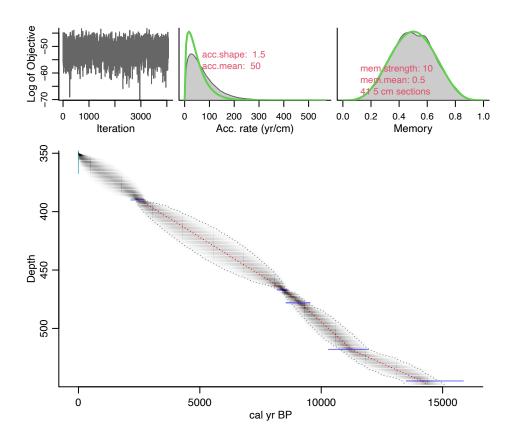


Figure S13. Revised age model for Loch Buailaval Beag.

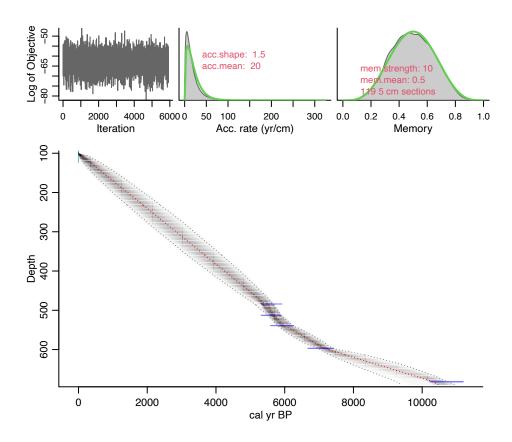


Figure S14. Revised age model for Loch Coulavie.

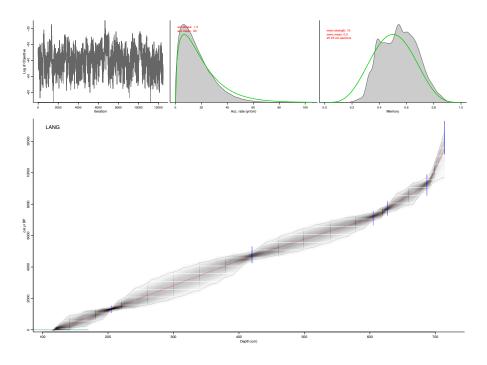


Figure S15. Revised age model for Loch Lang.

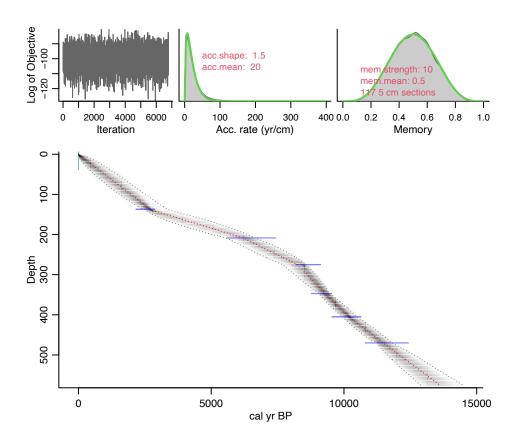


Figure S16. Revised age model for Loch na Beinne Bige.

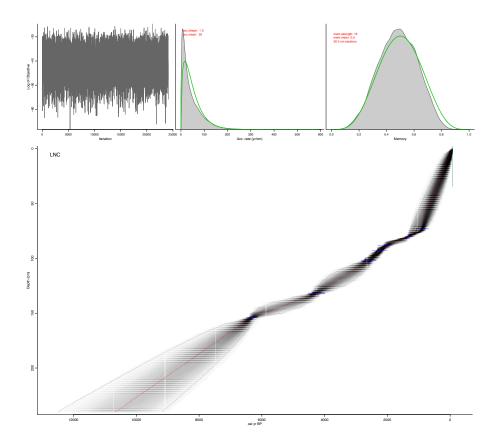


Figure S17. Age model for Loch na Claise.

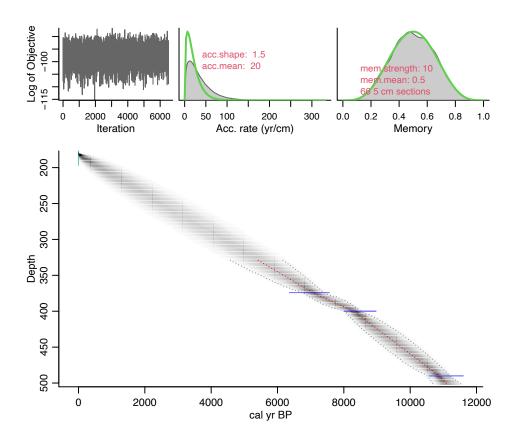


Figure S18. Revised age model for Loch na Muilne.

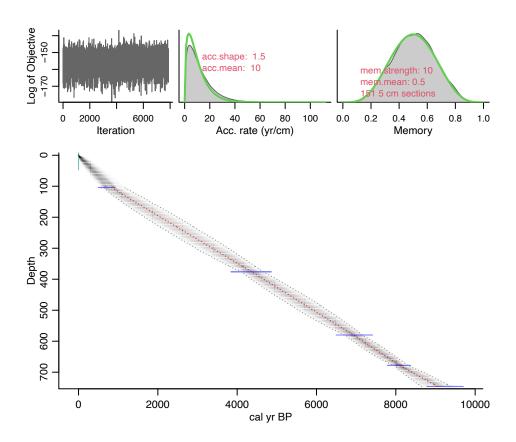


Figure S19. Revised age model for Loch nan Corr.

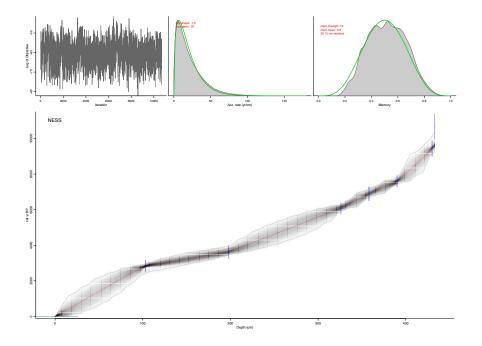


Figure S20. Revised age model for Loch Ness.

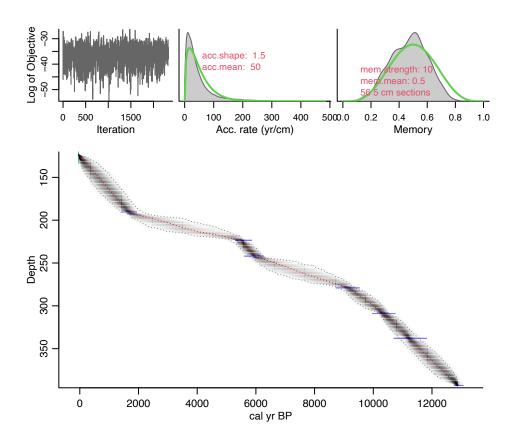


Figure S21. Revised age model for Loch Olabhat.

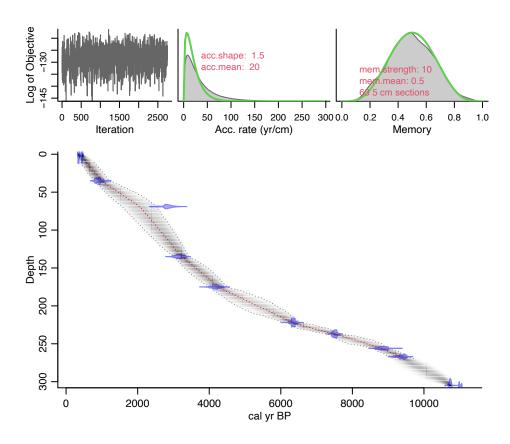


Figure S22. Revised age model for Loch Pityoulish.

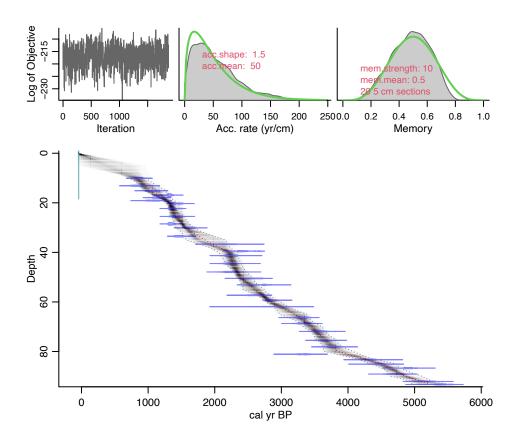


Figure S23. Revised age model for Lochan Uaine.

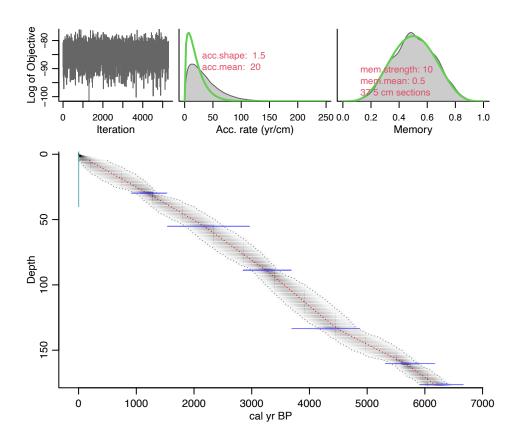


Figure S24. Revised age model for Lochnagar.

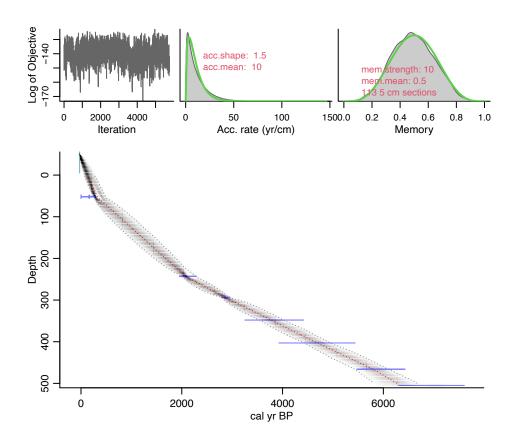


Figure S25. Revised age model for Lough Catherine.

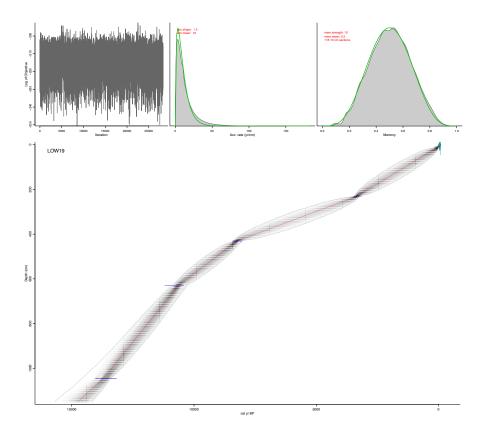


Figure S26. Mixed age model for Loweswater, including both ¹⁴C and ²¹⁰Pb dates.

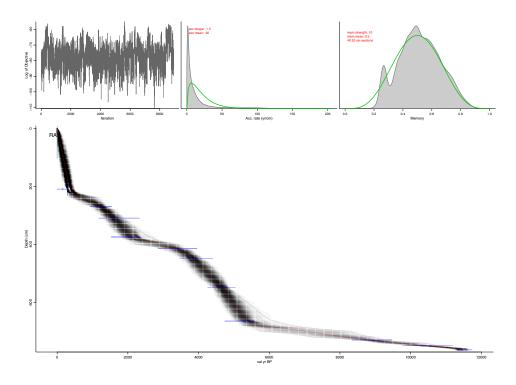


Figure S27. Revised age model for Rae Loch.

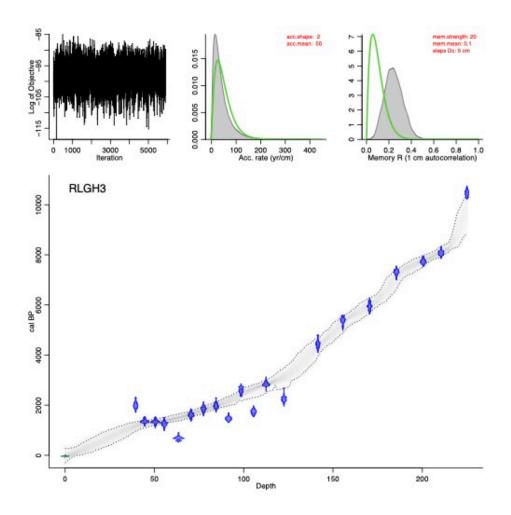


Figure S28. Revised age model for Round Loch of Glenhead.

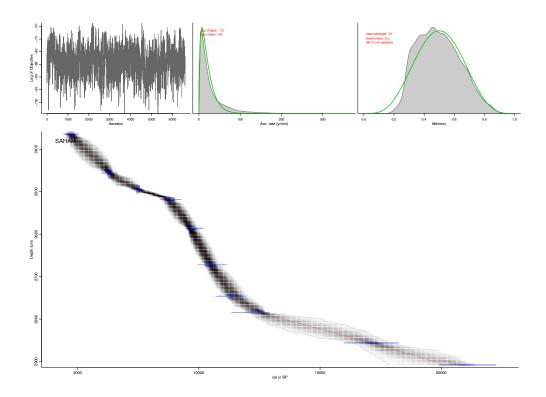


Figure S29. Revised age model for Saham Mere.

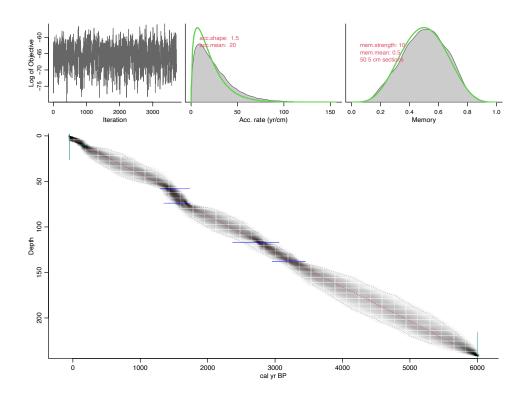


Figure S30. Revised age model for Talkin Tarn.

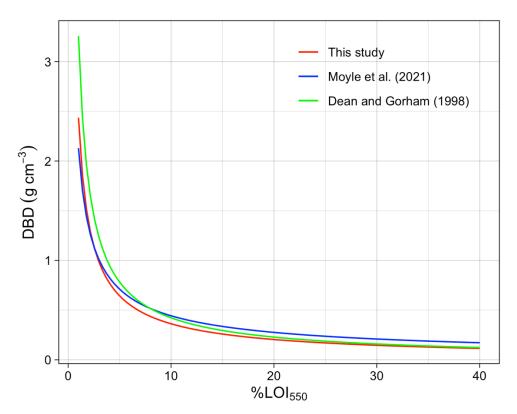


Figure S31. Comparison of empirical loss on ignition (%LOI₅₅₀) - dry bulk density (DBD) relationships derived from this study (DBD=2.4355*%LOI₅₅₀-0.827, r²=0.6, p< $2.2*10^{-16}$), Moyle et al., (2021) (DBD=2.13*%LOI₅₅₀-0.682) and Dean and Gorham (1998) (DBD=1.665*%OC-0.887). For ease of comparison, %OC in the Dean and Gorham (1998) relationship was multiplied by 0.469 and converted into %LOI₅₅₀ following Dean (1974) for ease of comparison.

Sample ID	Lab ID	Depth (cm)	¹⁴ C age (year BP)	¹⁴ C error (year)
LOW19 230-231	Poz-140857	230.5	3125	30
LOW19 430-431	Poz-140900	430.5	7470	40
LOW19 630-631	Poz-140901	630.5	9490	50
LOW19 830-832	Poz-140902	831	18520	140
LOW19 1044-1047	Poz-135822	1045.5	11690	80

Table S1. Radiocarbon (¹⁴C) dates for Loweswater.

 Table S2. Radiocarbon (¹⁴C) dates for Loch an Aigeil (Matthews 2022).

Sample ID	Lab ID	Depth (cm)	¹⁴ C age (year BP)	¹⁴ C error (year)
LAAG_008 (75-76cm)	SUERC-98655	75.5	1017	37
LAAG_007 (124cm)	SUERC-98651	124	2057	35
LAAG4-136cm	BETA-609264	136	2180	30
LAAG4-160cm	BETA-609265	160	2240	30
LAAG_003 (172cm)	SUERC-98656	172	2690	37
LAAG_006 (210cm)	SUERC-98650	210	3836	38

Table S3. Radiocarbon (¹⁴C) dates for Loch na Claise (Matthews 2022).

Sample ID	Lab ID	Depth (cm)	¹⁴ C age (year BP)	¹⁴ C error (year)
LNC_010 (73cm)	SUERC-98648	73	961	37
LNC19-80cm	BETA-609263	80	1420	30
LNC_011 (88cm)	SUERC-98649	88	2073	35
LNC_009 (93cm)	SUERC-98647	93	2264	35
LNC19_008 (102cm)	SUERC-98646	102	2539	35
LNC19_007 (132cm)	SUERC-98645	132	3876	36
LNC19_006 (153-154cm)	SUERC-98641	154	5524	35

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