

## The historical dependency of organic carbon burial efficiency

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### Abstract

Many studies have viewed lakes as quasi-static systems with regard to the rate of organic carbon (OC) burial, assuming that the dominant control on BE is sediment mineralization. However, in systems undergoing eutrophication or oligotrophication (i.e., altered nutrient loading), or climatic forcing, the changes in primary production will vary on both longer (> 10 yr) and shorter (seasonal) timescales, influencing the rate of OC accumulation and subsequent permanent burial. Here, we consider the extent to which permanent OC burial reflects changing production in a deep monomictic lake (Rostherne Mere, UK) that has been culturally eutrophied (present TP > 200  $\mu\text{g L}^{-1}$ ), but has undergone recent reductions in nutrient loading. We compare multi-year dynamics of OC fluxes using sediment traps to longer-term burial rates estimated from two  $^{210}\text{Pb}$ -dated sediment cores. The recent sediment record demonstrates that most of the autochthonous OC is preserved (~95% of OC captured in the deep trap and 86% of the NEP in the contemporary system), contrary to widely held assumptions that this more labile, algal-dominated OC component is not well preserved in lake sediments. A revised method for calculating BE for lakes which have undergone changes in primary productivity in recent decades is developed, which reduces some of problems inherent in existing approaches using historical sediment records averaged over the last 25–150 yr. We suggest that an appreciation of lakes in all biomes as ecosystems responding dynamically to recent human impact and climate change (for example) can improve up-scaled regional and global estimates of lake OC burial.

Despite covering only a small portion of the earth's land surface (~3.7%; Verpoorter et al. 2014), lakes are now recognized as key sites for the transformation and storage of considerable amounts of carbon (C) derived from either in-lake production or transfer from the catchment (Cole et al. 2007; Catalan et al. 2016). A portion of the organic carbon (OC) that settles to the bottom of a lake will be mineralized and either recycled or degassed as  $\text{CO}_2$ , or potentially undergo methanogenesis and degassed as  $\text{CH}_4$  (Fahrner et al. 2008), and the remainder will be buried. Burial of this OC in lake sediments can be considered as removal of atmospheric or terrestrial C from the active pool over geological timescales. Estimates of global C burial by lakes are between 0.02 Pg C  $\text{yr}^{-1}$  and 0.07 Pg C  $\text{yr}^{-1}$ , with most lakes burying between 4.5 g C  $\text{m}^{-2} \text{yr}^{-1}$  and 14 g C  $\text{m}^{-2} \text{yr}^{-1}$  (Tranvik et al. 2009), although rates are considerably higher in agriculturally dominated landscapes of Europe (~60–100 g C  $\text{m}^{-2} \text{yr}^{-1}$ ; Anderson et al. 2014) and North America (~7–554 g C  $\text{m}^{-2} \text{yr}^{-1}$ ; Heathcote and Downing 2012; Anderson et al. 2013; Clow

et al. 2015). Furthermore, many previous estimates based on lake sediment cores have not been corrected for the effects of sediment focusing, which will lead to overestimation when up-scaled (Buffam et al. 2011; Engstrom and Rose 2013; Anderson et al. 2014). Clearly, understanding sedimentation processes in lakes, and the extent to which lakes preserve OC in their sediments (i.e., burial efficiency [BE]), is key to improving the accuracy of such estimates and clarifying the role of lakes in regional and global C cycling.

Approaches to estimating OC BE have received considerable attention in both limnology and oceanography (Alin and Johnson 2007; Sobek et al. 2009; Anderson et al. 2014) yet within limnology this attention is still sparse without much agreement on a standard method. Formalized as the ratio between the rate of OC burial and gross sedimentation at the sediment surface (Sobek et al. 2009), studies of BE have been largely concerned with the factors controlling OC preservation. Production has generally been assumed to be constant over time, a concept largely derived from the marine literature (Hedges et al. 1999; Burdige 2007). While some drivers of OC preservation are relatively well understood, such as oxygen exposure time, with hypoxic or anoxic conditions in the hypolimnion reducing the OC decomposition rate (Laskov et al. 2002; Sobek et al. 2014), other key factors are still to be fully constrained, for instance the

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influence of temperature on decomposition rates (Sobek et al. 2009; Kothawala et al. 2014). For example, while Gudas et al. (2010) have argued that warmer water temperatures result in more mineralization and reduced OC burial, Anderson et al. (2013) demonstrated little climatic effect on C burial rates across a temperature gradient in 116 Minnesotan lakes. In a study of lakes from West Greenland, Sobek et al. (2014) similarly found little effect of temperature on burial efficiency. The source and type of OC has also been recognized as a potential control of OC BE, with terrestrially derived OC often assumed to be refractory, and autochthonous OC, labile (Sobek et al. 2009). Other drivers have also been argued to have a bearing on OC BE, such as basin morphometry (Ferland et al. 2012), changes in sunlight (Cory et al. 2014; Koehler et al. 2014; Tranvik 2014), sediment flocculation (Von Wachenfeldt et al. 2008; Sobek et al. 2009), OC molecular properties (Kellerman et al. 2015), and mineral sorption (Maerki et al. 2006).

Much of the current understanding of processes of lake OC burial (especially preservation) is based on studies from boreal lakes (Sobek et al. 2005), where production (OC input) is generally assumed to be constant in the recent past (steady-state conditions). In agricultural landscapes, long-term changes to land use intensity have led to the disruption of regional nutrient cycling with increased erosion, transportation, and deposition of sediment from tilled agriculture (Clow et al. 2015), leading to the widespread development of freshwater eutrophication influencing autochthonous OC production and hence OC burial (Anderson et al. 2013; Dietz et al. 2015). While the factors controlling OC preservation are clearly important for OC BE, the implicit assumption that lake OC production is in a (quasi-) steady state in most systems is invalid, given the multiple stressors that lakes are subject to in all biomes (tropical, temperate, and boreal) (Leavitt et al. 2009; Williamson et al. 2009). Many culturally impacted lakes are in a state of continual adjustment to increasing or decreasing nutrient loading (i.e., inputs of OC, whether terrestrial or aquatic, have changed over recent decades), and thus may influence OC burial rates together with those factors (i.e., temperature,  $O_2$ ) influencing preservation.

Here, we utilize long-term (5-yr) high temporal resolution (2–4 weeks) sediment trap observations of OC flux in a strongly stratified, nutrient-rich lake that is recovering from cultural eutrophication (Rostherne Mere, UK), to generate estimates of OC flux to the sediment surface at seasonal, annual, and sub-decadal scales (Douglas et al. 2002). Although traps (in the same way as sediment cores), are susceptible to focusing effects (i.e., over-trapping), trap fluxes can be corrected by comparison to monitored records of net ecosystem production (NEP) at the lake (Scott 2014), and to contemporary OC accumulation measured from a well-dated, focusing-corrected sediment core. Furthermore, we compare the long-term high-resolution trap monitoring data to this sediment record of OC burial over the last ~150 yr to

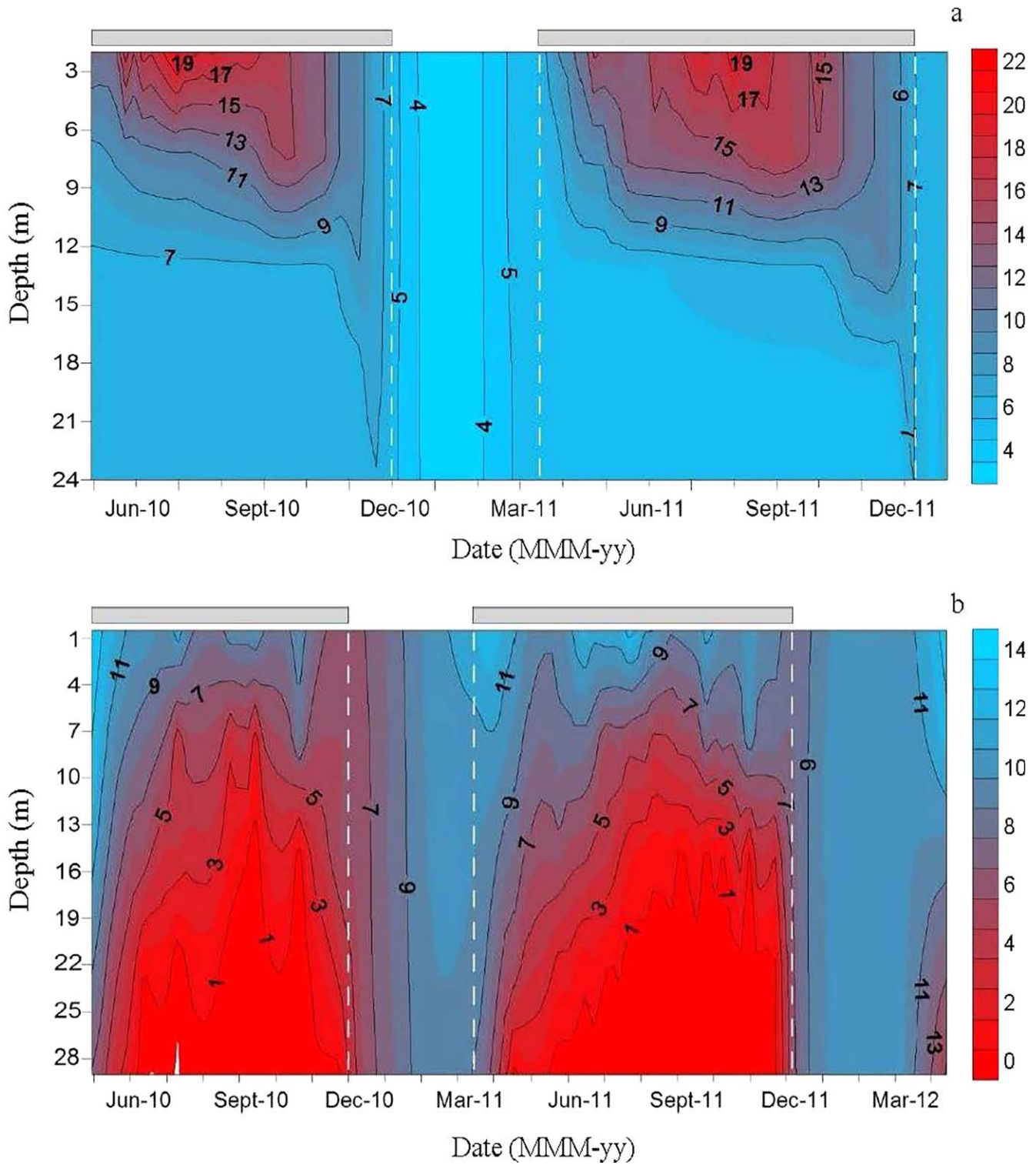
examine historical patterns of OC burial and apply different methods to calculate OC BE at Rostherne Mere. Given the widespread occurrence of (seasonal) hypolimnetic anoxia among lakes globally (Kalff 2002), Rostherne Mere provides an important test of the widely held assumption that labile OC is rapidly mineralized after sedimentation in such lakes. This distinction is critical in determining to what degree historical trends in OC burial are driven by changes to terrestrial OC supply and sedimentary OC preservation (the latter controlled by oxygen exposure time, temperature, and OC lability), rather than long-term changes in autochthonous production. This study has broad implications for studies of OC BE and preservation in such dynamic, culturally impacted lakes and other systems where direct and indirect impacts of human activity have altered autochthonous OC production and/or terrestrial OC inputs in the recent past.

## Methodology

### Study site

Rostherne Mere (53°20'N, 2°24'W) is a relatively large (48.7 ha) and deep (maximum depth ~30 m; mean depth 13.6 m) lake within the Shropshire-Cheshire Meres (Reynolds 1979; Carvalho et al. 1995). It is a kettle basin lake, with one significant inflowing stream to the west, numerous small stream-fed inflows and a single surface outflow. The lake is monomictic with an estimated water retention time of 1.6–2.4 yr (Moss et al. 2005). The lake hosts an automated water quality monitoring station located at a central buoy, being part of the UK Lake Ecological Observatory Network (UKLEON) project ([see http://www.ceh.ac.uk/our-science/projects/uk-lake-ecological-observatory-network-ukleon](http://www.ceh.ac.uk/our-science/projects/uk-lake-ecological-observatory-network-ukleon)).

The lake remains hyper-eutrophic ( $> 100 \mu\text{g P L}^{-1}$ ; Carlson 1977), in large part due to anthropogenic P loading from a sewage treatment works beginning in the 1930s and ending in 1991. Cyanobacteria are the dominant primary producers in the lake, forming blooms in the summer, largely *Aphanizomenon flos-aquae* and *Microcystis aeruginosa* (Reynolds 1979; Moss et al. 2005). The stable summer stratification (average thermocline depth April to November = 10 m; Fig. 1a), leads to an anoxic hypolimnion (Fig. 1b) developing over summer as a result of organic matter decomposition rapidly utilizing the oxygen in the lower water column (Scott 2014). The epilimnion, however, remains well oxygenated and nutrient dynamics are mainly influenced by biological uptake, replenishment from inflows, and entrainment of hypolimnetic waters during periods of windy weather (Krivtsov et al. 2001a). Despite the removal of point-source sewage effluent, the lake is still hyper-eutrophic ( $216 \pm 88 \mu\text{g P L}^{-1}$ ; Scott 2014) with high levels of P mobilized from the sediments below the anoxic hypolimnion ( $7\text{--}11 \text{ mg m}^{-2} \text{ d}$ ; Krivtsov et al. 2001b) causing substantial internal loading and limiting the rate of ecological recovery (Krivtsov and Sigg 2005; Moss et al. 2005).



**Fig. 1.** The seasonal cycle of stratification at Rostherne Mere during 2010 and 2012 (adapted from Scott 2014). Periods of lake stratification are shown with gray bars and dashed lines. (a) Depth-time plot of temperature (°C) and (b) depth-time plot of dissolved oxygen (mg L<sup>-1</sup>) between May 2010 to December 2011.



### Sediment collection

Sediment trapping using both open tube and sequencing traps was carried out at Rostherne Mere from April 2010 to March 2015. Open tube sediment traps (KC Denmark, Silkeborg, Denmark; older variation of <http://www.kc-denmark.dk/products/sediment-trap-station/sediment-trap-station-oe80-mm-tubes.aspx>), comprising four clear plastic tubes (450 mm length/72 mm internal diameter, 1 : 6.3 trapping ratio, 0.016 m<sup>2</sup> trapping area per 4 tubes), were deployed at 10 m and 25 m depth in the central, deepest part of the lake (~30 m). Some early tube trap collections (shallow trap April 2010 to June 2010; deep trap April 2010 to May 2011) used a funnel (to maximize absolute catch for a separate project) that resulted in a lower material capture per unit area (Bloesch and Burns 1980). As a result, a series of calibration traps (paired traps with and without funnel) were deployed and a lake-specific calibration factor (with:without funnel = 3.13) was calculated to correct tube trap collections on a unit area basis. Technicap PPS 4/3 automatic sequencing traps (1310 mm length/252 mm internal diameter, 1 : 5.1 trapping ratio, 0.05 m<sup>2</sup> trapping area; <http://www.technicap.com/images/product/pps-4-3.pdf>) were also deployed at 10 m and 25 m water depths, sequentially opening into 12 individual 250 mL HDPE bottles, each representing a 2-week collection period (except in January and February longer collection periods of up to 4 weeks were used). The traps were reset every 6 months as dictated by the trapping interval used, with trap sediment kept cool, dark, and sealed during transport to the laboratory where it was stored frozen prior to analysis.

A 112 cm long sediment core (RM-LIV-2011, hereafter the RML core) was collected at 26 m water depth in September 2011 using a Livingstone piston corer (Wright 1967). The sealed core was transported vertically to the laboratory, and stored vertically in a dark cold room at 5°C, prior to extrusion at 1 cm intervals for the upper 50 cm of sediment, and then at 0.5 cm intervals for the remainder of the core.

Water column oxygen concentration and temperature were measured at 1 m intervals approximately every 3 weeks between May 2010 and April 2012 using a YSI 6600 V2 multi-parameter sonde (Scott 2014). Hourly NEP estimates of the pelagic epilimnion at 1 m depth at the UKLEON buoy were calculated for 2011–2012 by the free-oxygen method using daytime water column dissolved oxygen calculations (Odum 1956). Precipitation data from the closest reliable Meteorological Office station (Shawbury, Shropshire, UK; situated 64 km south-west from Rostherne Mere) were utilized to generate 30-yr monthly and annual means for comparison to the study period monthly and annual means. Average daily wind speed and surface water temperatures were taken from the UKLEON buoy.

### Sediment analysis

All trap and core samples were freeze-dried prior to analysis. For all samples organic matter (OM) was determined using

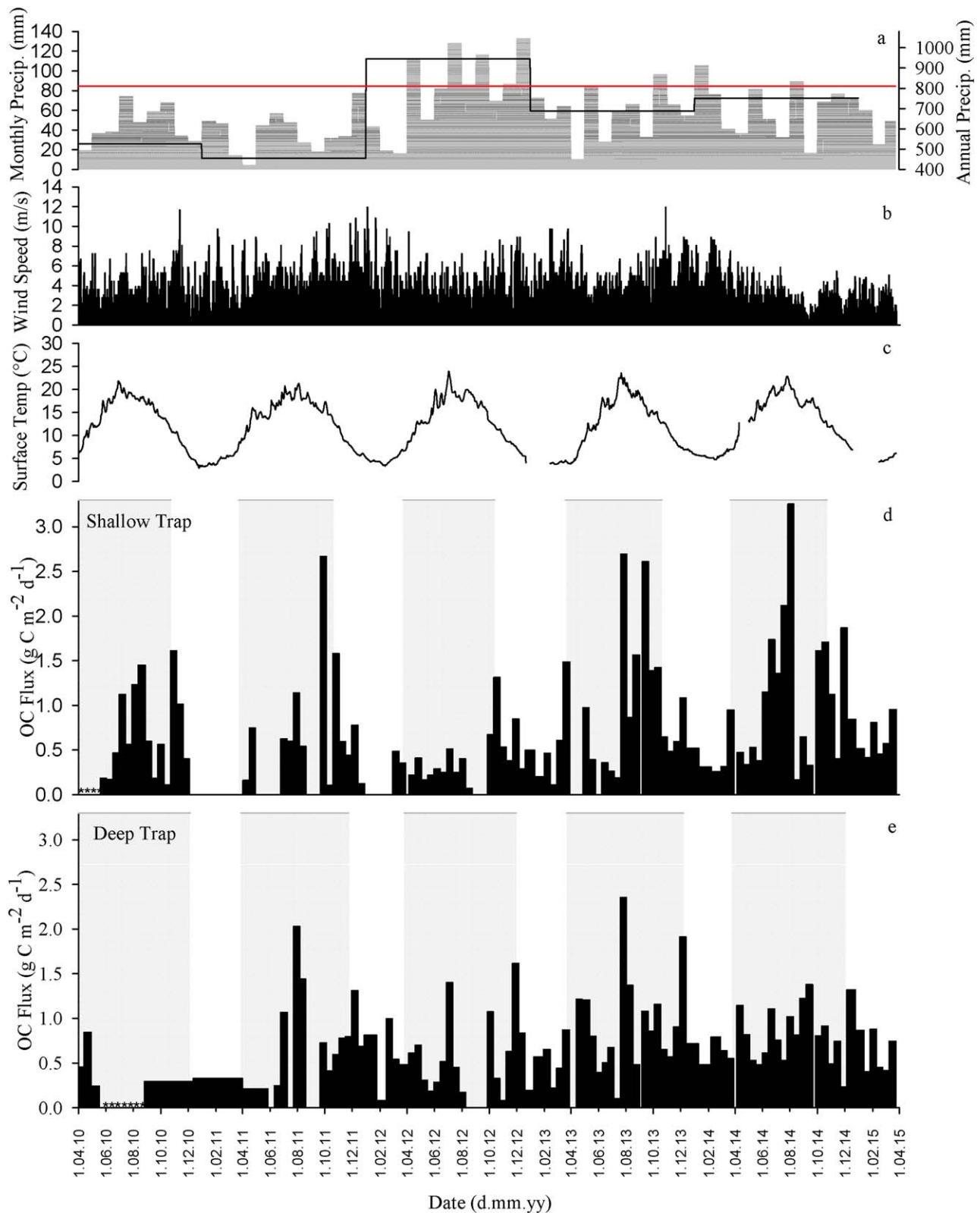
sequential loss-on-ignition, where OM was calculated by weight-loss after 3 h at 550°C (Dean 1974). Percentage OC was calculated from %OM using a lake-specific conversion factor (%OC = %OM \* 0.56) estimated from analysis of 20 sediment samples with a range of %OM (14–65%) with total OC determined via mass-spectrometry elemental analysis.

Freeze dried and homogenized core samples were analyzed via alpha spectrometry for <sup>210</sup>Pb activity to determine chronology and sediment accumulation rates according to the constant rate of supply (CRS) model with confidence intervals calculated by first-order error analysis of counting uncertainty (Appleby 2001).

### Data analysis

Preliminary analysis of the sediment trap OC flux showed a high winter sediment collection, especially in the deep (25 m) trap (Fig. 2d,e), despite little to no primary production during this time, as confirmed by high-resolution monitoring of NEP (Scott 2014). Rostherne Mere has an intensely managed catchment, with restricted land use and agricultural activity, so it can be assumed the terrestrial OC contribution is minimal and the NEP is the dominant OC source. Therefore, the high winter collection in the deep sediment trap implies over trapping is a significant issue within Rostherne Mere, as expected from its morphometry and stratification pattern (Hilton 1985). Seasonal over trapping was corrected through the removal of the winter sediment collection (1<sup>st</sup> November–15<sup>th</sup> March, as determined by comparison to the NEP analysis, i.e., the period when net production was ~0) from the calculated annual collection totals. Furthermore, a correction value of +10% was added to offset the OC loss rate in the sediment traps as a result of mineralization in the traps themselves. The 10% correction factor is an arbitrary number taken from other similar studies that have previously suggested the figure to be a suitable correction with minimal error (Bloesch and Burns 1980; Horppila and Nurminen 2005). This approach was independently verified by comparison to the 2011–2012 NEP (Scott 2014) with the corrected trap fluxes fitting within the expected range of NEP values, assuming the terrestrial OC component within this study is minimal. Subsequently the corrected values were summarized as annual totals (1<sup>st</sup> April to 31<sup>st</sup> March; Table 1) and compared between years and trap depths (Table 2).

Loss rates of OC in the water column were calculated by the difference between each sampling depth (NEP at ~0.5 m, traps at 10 m and 25 m; Fig. 3). Both trap and core corrected data were used to calculate BE according to two published methods (Fig. 3). Alin and Johnson (2007) proposed burial efficiency (here denoted BE<sub>25</sub>) as the ratio of OC burial (the mean OC mass accumulation of sediments between 10 yr and 25 yr in age; see below) to NEP. Alternatively, Sobek et al. (2009) describes burial efficiency (here denoted BE<sub>150</sub>) as the ratio of OC burial (the mean OC mass accumulation of sediments between 25 yr and 150 yr in age) to the OC



**Fig. 2.** Rostherne Mere OC flux ( $\text{g C m}^{-2} \text{d}^{-1}$ ) captured in sediment traps (uncorrected for over-trapping or mineralization losses) at a depth of 25 m (e) and 10 m (d) overlaid with timings of stratification (gray bars stratified period) for respective depths. Daily average surface water temperature (c), daily average wind speed (b), and monthly (shaded gray area against the left y-axis) and annual total precipitation (single black line against right y-axis) with 30-yr average (single red line against right y-axis) plotted (a) for comparison to changes in trapped OC flux. \* = missing data.

**Table 1.** Uncorrected and corrected OC flux rates ( $\text{g C m}^{-2} \text{yr}^{-1}$ ) for limnological years (1<sup>st</sup> April to 31<sup>st</sup> March) 2010–2015. Data from 1<sup>st</sup> November to 15<sup>th</sup> March have been removed to correct for over trapping of resuspended particles and a 10% trap OC mineralization factor has been applied (see text for details). Net ecosystem production (NEP) for 2011–2012 is estimated from the free-oxygen method (Scott 2014).

Year	Uncorrected shallow trap	Corrected shallow trap	Uncorrected deep trap	Corrected deep trap	NEP
2010–2011	225.15	167.69			
2011–2012	157.72	133.74	208.52	122.56	135.6 ± 91
2012–2013	154.09	106.81	183.00	112.98	
2013–2014	306.14	279.78	313.39	227.01	
2014–2015	355.61	280.58	287.93	215.36	
Total means (2011–2015)	<b>243.39</b>	<b>200.23</b>	<b>248.21</b>	<b>169.48</b>	
Total means (2010–2015)	<b>239.74</b>	<b>193.72</b>			

**Table 2.** Uncorrected and corrected trap OC flux ratios highlighting the changes applied with correction methods and the loss rate between sediment traps. Note the outlying 2012 ratio highlighting the impact of the annual variability from extreme meteorological behavior.

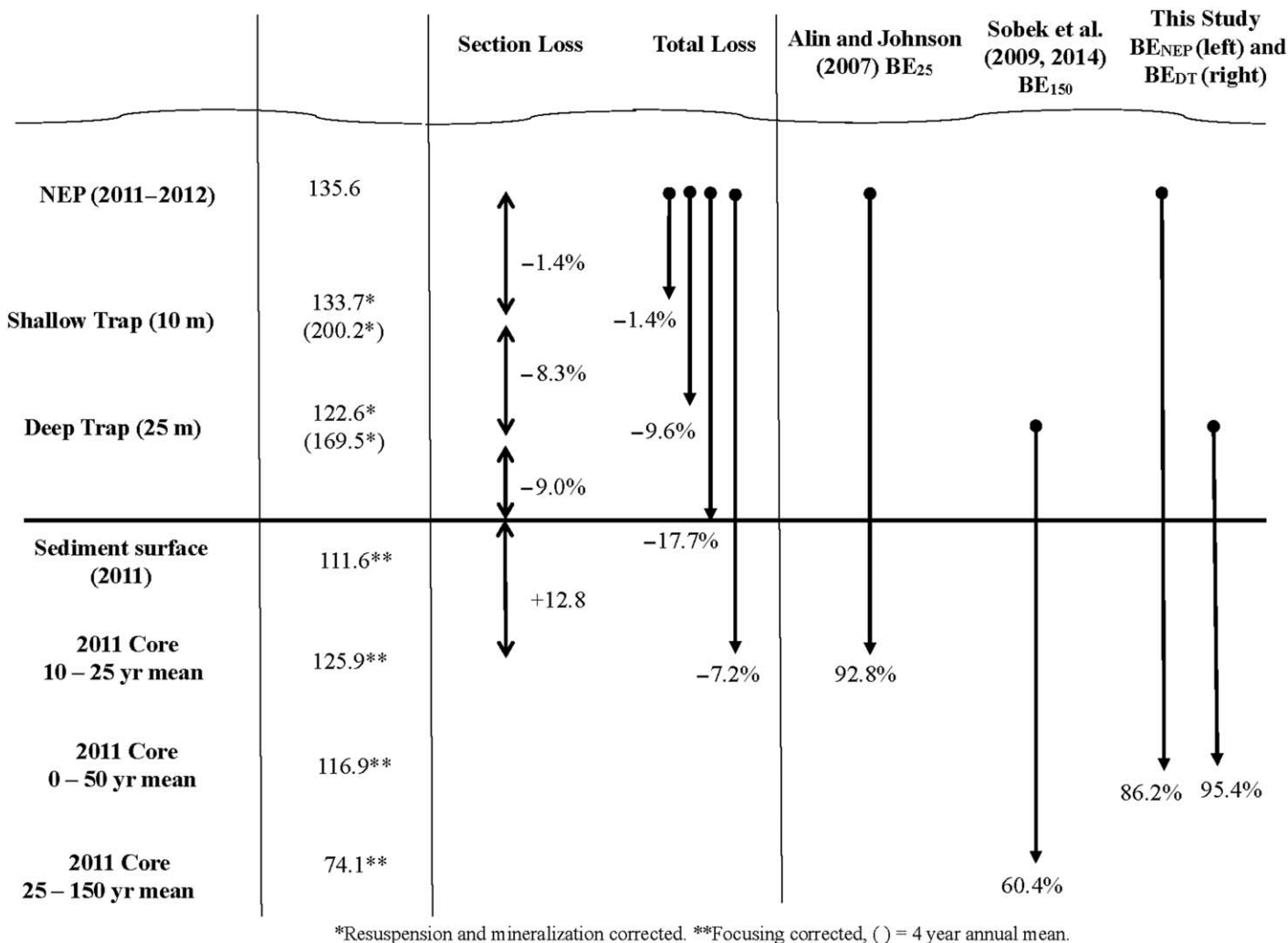
Year	Shallow corrected/shallow uncorrected	Deep corrected/deep uncorrected	Deep uncorrected/shallow uncorrected	Deep corrected/shallow corrected
2010–2011	0.74			
2011–2012	0.85	0.59	1.32	0.92
2012–2013	0.69	0.62	1.19	1.06
2013–2014	0.91	0.72	1.02	0.81
2014–2015	0.79	0.75	0.81	0.77
Total means (2011–2015)	<b>0.81</b>	<b>0.67</b>	<b>1.09</b>	<b>0.89</b>
Total means (2010–2015)	<b>0.80</b>			

delivery to the sediment surface (deep trap capture). The literature on calculating OCBE is sparse, with an agreement on a standard method elusive. Therefore, due to limitations found with both these methods when applied to Rostherne Mere due to its recent, and ongoing, changes in production over the timescales used in calculations of  $\text{BE}_{25}$  and  $\text{BE}_{150}$ , a revised method of calculating BE is proposed using sediments deposited over the last 50 yr.

The use of sediment cores provides a long term perspective on C burial rates. Sediment core OC accumulation rates were estimated by multiplying bulk sediment accumulation rates ( $\text{g dry matter cm}^{-2} \text{yr}^{-1}$ , derived from  $^{210}\text{Pb}$  dating) by the OM% and the OM/OC conversion factor (see above) and are reported as  $\text{g C m}^{-2} \text{yr}^{-1}$ . Significant problems can arise with this approach for whole-basin upscaling if sediment focusing is not considered (Anderson et al. 2014), due to pelagic accumulation often overestimating whole-lake accumulation. Therefore, core sediment accumulation rates were focusing-corrected (by a factor of 0.7) using the ratio of the expected unsupported flux of  $^{210}\text{Pb}$  ( $9.82 \text{ pCi cm}^{-2}$ ) to that

found in the core (Anderson et al. 2013, 2014). After deposition into the bottom sediments, the OC mineralization rate slows exponentially toward zero with increasing depth below the sediment-water interface (Middelburg 1989). Sediments younger than 5 yr have the highest rate of decay, dropping dramatically after  $\sim 10$  yr (Thomsen et al. 2004; Galman et al. 2008). Therefore, the most recent 10 yr of accumulation ( $< 5$  cm sediment core depth,  $^{210}\text{Pb}$  dated age  $\sim \text{AD } 2001 \pm 2.04$  yr) was removed from flux calculations to avoid the influence of incomplete mineralization (Galman et al. 2008; Heathcote et al. 2015).

For comparison to the RML core, a previous core taken from the central deepest part of Rostherne Mere in 1977 (Livingstone 1979) using a Mackereth 1 m mini-corer (Mackereth 1969) was used (hereafter referred to as the LIV77 core). A loess smoother was fitted to the LIV77 core data to reduce noise and enable clearer comparison to the RML core. Additionally, we compare the historical record of Rostherne Mere with nine Danish lakes (all with independently dated  $^{210}\text{Pb}$  records and focusing corrected; Anderson et al. 2014). These



**Fig. 3.** Rostherne Mere OC from NEP (2011–2012), trap data (corrected 2011–2012 and 4-yr annual mean in brackets), and historical sediment core accumulation rate, showing the estimated losses through the water column and into the surface sediment. Various methods for calculating burial efficiency are shown (BE<sub>25</sub> [NEP to 10–25 yr mean of core], Alin and Johnson 2007; BE<sub>150</sub> [deep trap to 25–150 yr mean of core], Sobek et al. 2009; BE<sub>NEP</sub> [NEP to 0–50 yr mean of core], this study; BE<sub>DT</sub> [deep trap to 0–50 yr mean of core], this study), highlighting the difference in the methods. Values shown are g C m<sup>-2</sup> yr<sup>-1</sup>.

Danish lakes have been undergoing nutrient reduction over a similar time period to Rostherne Mere, as part of a wider national policy to reduce nutrient loading to surface waters (European Commission 2012). A loess smoother was fitted through the aggregate Danish core data to reduce noise and highlight the trend, with a  $p = 0.95$  confidence envelope calculated in R (using ggplot2 software; Wickham 2009).

**Results**

During the study, total annual rainfall at Rostherne Mere (Fig. 2a) was below the 30-yr average (study period = 676.7 mm yr<sup>-1</sup>; 30-yr annual mean = 810.2 mm),

with the exception of 2012 where the total annual rainfall (1054.6 mm) was 30% higher than the 30-yr average. This was largely due to an unusually wet summer, with a 55% higher June to September total rainfall in 2012 (413.2 mm) compared to 30-yr mean (267.1 mm). Daily wind speed was variable (daily average range 0–11.9 m s<sup>-1</sup>) with slight increases over the winter months, as expected in this location (Fig. 2b). Surface water temperature reflects air temperature (range ~3°C to 24°C; see Figs. 1a, 2c).

The shallow and deep trap uncorrected OC flux shows high capture is not restricted to summer, although the stratified and productive summer period commonly had higher catches than the winter and spring (Fig. 2d,e), with greater



fluxes recorded in 2013–2015 than earlier years (Table 1). Despite being suspended only 15 m apart in the water column, the two traps sometimes showed large differences in individual 2-weekly catch, as observed in other multiple trap studies (Moschen et al. 2009). At times, simultaneous peaks in collection in both traps indicate rapid settling (e.g., early August 2013; Fig. 2d,e), while other periods are characterized by a slow downward flux of particles (e.g., August 2011 and August 2014; Fig. 2d,e). This variation in settling rate results in the signal in the shallow trap being blurred in the deep trap on a 2-weekly timescale (e.g., summer 2012; Fig. 2d,e). Across the study the uncorrected winter OC collection mean is similar between the shallow trap and deep trap (5-yr average  $243.4 \text{ g C m}^{-2} \text{ yr}^{-1}$  and  $248.2 \text{ g C m}^{-2} \text{ yr}^{-1}$ , respectively; Table 1), with the deep trap collecting more in 3 of the 4 yr (2011–2014; Table 2). This is typical where intermittent complete mixing (ICM) dominates sedimentation processes (Hilton 1985), resuspending unconsolidated organic matter into the water column during the mixed period. After correction for both ICM (i.e., removing the winter collection) and a fixed 10% mineralization loss in the collecting bottles, the shallow trap mean value was  $200.2 \text{ g C m}^{-2} \text{ yr}^{-1}$ , and  $169.5 \text{ g C m}^{-2} \text{ yr}^{-1}$  in the deep trap over the study period (Table 1). Calculated NEP from high-frequency monitoring in 2011–2012 ( $135.6 \text{ g C m}^{-2} \text{ yr}^{-1}$ ; Scott 2014) differs by <5% with the corrected trap catch in 2011–2012 for both shallow and deep traps, supporting the approach taken to adjust trap catch here. The difference between the corrected shallow trap flux in 2011–2012 ( $\sim 134 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) and the surface sediment accumulation rate (from the core collected in September 2011;  $\sim 112 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) is  $22.1 \text{ g C m}^{-2} \text{ yr}^{-1}$ , which is close to the C efflux as calculated from lake profile  $\text{CO}_2$  measurements in 2011–2012 ( $33.5 \text{ g C m}^{-2} \text{ yr}^{-1}$ ; Scott 2014). We therefore conclude that corrected trap flux is a good estimate of NEP (shallow trap) and OC delivered to the surface sediments (deep trap) for calculation of BE, assuming the terrestrial OC contribution is minimal as previously suggested.

The average OC loss through the water column in 2011–2012 was 1.4% (lake surface to shallow trap at 10 m), 8.3% (shallow trap to deep trap at 25 m) and 9.0% (deep trap to surface sediment at 26 m) with about 10% of NEP lost through the water column to 25 m, and a further  $\sim 8\%$  at the sediment surface (Fig. 3). Using the Alin and Johnson (2007) method comparing NEP in 2011–2012 to average sediment accumulation rate from the previous 10–25 yr,  $\text{BE}_{25}$  is estimated at 92.8% (Fig. 3). Alternatively, the Sobek et al. (2009) method, comparing deep trap flux to average sediment accumulation rate from the previous 25–150 yr, gives an estimate of  $\text{BE}_{150}$  as 60.4%, or two-thirds of the Alin and Johnson (2007) method (Fig. 3).

The burial rate from the RML core (focusing-corrected; Fig. 4) shows an increase in OC burial from  $24 \text{ g C m}^{-2} \text{ yr}^{-1}$  in 1900 to  $138 \text{ g C m}^{-2} \text{ yr}^{-1}$  in the late 1980s (a sevenfold

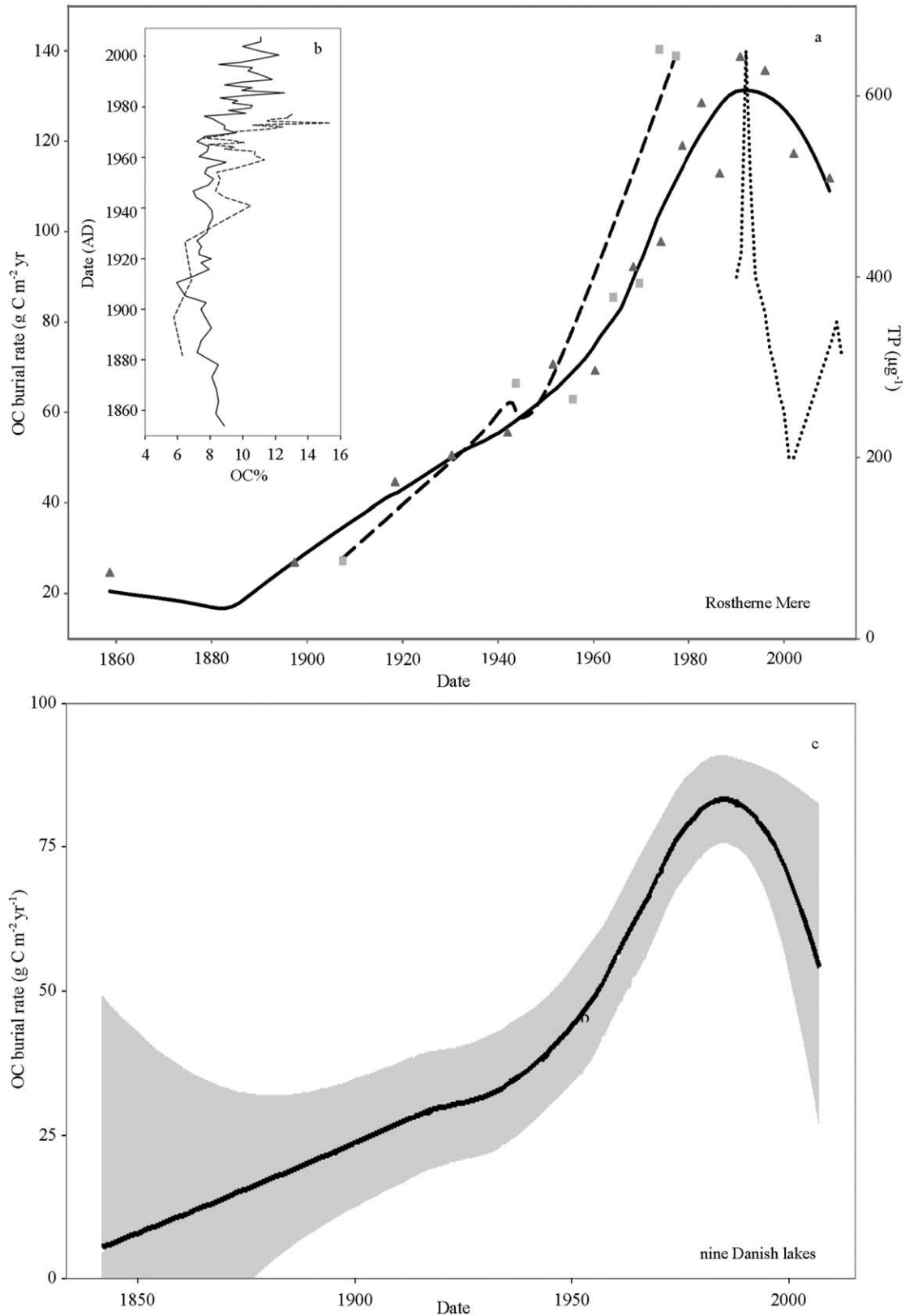
increase). Both the temporal pattern and burial rates of the RML core are very similar to the focusing corrected LIV77 core (Fig. 4a), demonstrating the consistency of the deep water sediment archive across the lake. Small discrepancies between the two cores are likely due to differing core locations and depths (Anderson 1990) as well as the incomplete mineralization in the upper part of the 1977 core compared to sediments this age in the 2011 core (Fig. 4b). Since the 1990s, the RML core OC accumulation rate has declined to approximately  $110 \text{ g C m}^{-2} \text{ yr}^{-1}$ . The temporal pattern of OC burial at Rostherne Mere corresponds to the historical record of intensification of eutrophication over the last century associated with sewage treatment works development, and the recent diversion of effluent from the inflowing stream in 1991, initiating a gradual recovery (Moss et al. 2005). This recent decline in sedimentary OC accumulation rate shows good agreement with the decline in the measured TP concentrations from a maximum of  $400\text{--}600 \mu\text{g P L}^{-1}$  at the peak of eutrophication in the late 1980s, to approximately  $200\text{--}300 \mu\text{g P L}^{-1}$  in recent years (Fig. 4a). Although the exact timing differs slightly and accumulation rates are lower, changes over the 19<sup>th</sup> and 20<sup>th</sup> centuries at Rostherne Mere (with nutrient enrichment followed by reduction) are mirrored in many Danish lakes (Fig. 4c) which have undergone similar experiences of human impact and recent management over the last 100–150 yr. Trajectories of change in lake OC production and burial are clearly shared across industrial and post-industrial landscapes across Europe.

## Discussion

### Inter-annual variability in OC dynamics

Culturally impacted lakes are not only prone to longer term changes ( $> 10$  yr) in OC burial potential (due to varying nutrient loading), but also show short term fluctuations on inter-annual timescales (Reynolds and Reynolds 1985; Gibson et al. 2000). At Rostherne Mere, for example, trap fluxes were atypically low in 2012–2013, only  $106.81 \text{ g C m}^{-2} \text{ yr}^{-1}$  in the shallow trap, which is 50% lower compared to the mean of the other 4 yr (Table 1). Although there are several episodes of negligible trap catch over the 5-yr record in either or occasionally both traps, consistently and unusually low flux was most evident during the summer of 2012, with trap catch negligible in both traps in September 2012 (Fig. 2d,e). The reason for this seasonal anomaly can most likely be attributed to extreme meteorological conditions. Rainfall was 66.2% higher in September 2012 (the wettest summer period for the UK since 1912) and 30.2% higher in total for the year 2012–2013, compared to the 30-yr average (Fig. 2a). The exceptional hydrological conditions of that summer will have resulted in a combination of factors limiting algal growth. Light for photosynthesis would have been reduced by greater cloud cover (Brooks and Zastrow 2002) and increased turbidity from greater inflows of turbid flood





**Fig. 4.** (a) Loess smoothed focusing corrected OC burial rate from core RML (solid line, dark gray triangles, collected in 2011) and LIV77 core (dashed line, light gray squares; collected in 1977), compared with mean surface water annual TP concentration (dotted line). (b) OC% down core profiles for RML (solid line) and LIV77 (dashed line) cores. (c) Aggregated sedimentary records of nine Danish lakes over the last ~ 150 yr showing 20<sup>th</sup> century eutrophication and oligotrophication (taken from Anderson et al. 2014). Lake records have been independently <sup>210</sup>Pb-dated and focusing-corrected. A loess smoother with 0.95 confidence envelope has been fitted through the data.

water (mean minerogenic fraction in shallow sediment trap was 9.1% higher [56.3%] than the study period mean [47.2%]), while reduced lake water residence time would decrease phytoplankton standing crop by outflow washout (Reynolds et al. 1982; Cross et al. 2014). Together, this would reduce total algal production in the lake, and hence reduce OC burial potential.

In recent years (2013–2015), the sediment trap total collections have shown an increase in total yield (Table 1), aligning to an increase in TP levels (Fig. 4a) and the impact of climatic variability mentioned previously. This rise suggests a short term increase in production despite a longer-term trend to oligotrophication and recovery. These seasonal and inter-annual fluctuations highlight the importance of combining trap and sediment core studies allowing the variability in OC dynamics (production, sedimentation, and burial) to be assessed across a range of temporal scales, from seasonal to decadal, and emphasize the benefits of sediment trap studies that last more than one limnological cycle (Kulbe et al. 2006; Moschen et al. 2006).

#### Preservation controls on OC burial efficiency

Rostherne Mere is very efficient at storing OC, with an estimated BE of between 60% and 93%, as calculated using the Sobek et al. (2009) and Alin and Johnson (2007) methods, respectively ( $BE_{150}$  and  $BE_{25}$ ; Fig. 3). This falls within the upper end of BE reported in a range of other lakes with and without focusing-correction, with ~31% in two lakes in West Greenland ( $BE_{150}$  method used for Lake SS4 and Lake SS8; Sobek et al. 2014), 23.2–26.1% ( $BE_{150}$  method) and 44.7% ( $BE_{25}$  method) in eutrophic Baldeggersee, Switzerland (Teranes and Bernasconi 2000; Muller et al. 2012) (not focusing corrected). Brothers et al. (2013) reported ~100% efficiency in Kleiner Gollinsee, Germany (deep trap to uncorrected sediment surface accumulation), yet here application of the  $BE_{150}$  method gives a 28.5% efficiency. Previously, discussion of variability in OC BE between lakes has focussed on the processes driving preservation, with consensus that the dominant controls are oxygen exposure time, temperature and the dominant OC type (i.e., labile autochthonous vs. refractory allochthonous carbon) (Calvert et al. 1991; Sobek et al. 2014).

When considering the controls on OC preservation in Rostherne Mere, the long, stable periods of stratification and associated hypolimnetic anoxia are a key factor. The high levels of production in the lake lead to increased oxygen consumption rate in the hypolimnion following the sedimentation of the spring algal bloom (Rippey and Mcsorley 2009), with rapid deoxygenation of the hypolimnion (within 4–6 weeks after stratification; Fig. 1b) and low OC mineralization rates in the deeper water column and at the sediment surface in the profundal zone (Laskov et al. 2002; Sobek et al. 2014). Once the available dissolved  $O_2$  is depleted (within a few weeks of stratification; Scott 2014; Fig. 1b),

denitrification, methanogenesis, and manganese reduction will be stimulated (Davison and Woof 1984; Thomsen et al. 2004; Fahrner et al. 2008). However, within Rostherne Mere the redox sequence observed in other systems may only reach the initial stages due to a lack of available electrons (Davison and Woof 1984; Davison et al. 1985), adding to the high BE potential of the lake. While research in marine systems has questioned the role of anoxia in promoting high OM preservation (Calvert et al. 1991), anaerobic respiration of OM is generally less efficient than aerobic (Sobek et al. 2009). Additionally, given the year-round low temperature (~6°C; Fig. 1a) in the hypolimnion of Rostherne Mere (and other mid- and higher latitude stratifying lakes), kinetic rates affecting biogeochemical and biological processes involved in OC respiration and diagenesis will also be reduced, enhancing OM preservation (Tison and Pope 1980).

Interestingly, the OC loss rates in the water column at Rostherne Mere are seen to increase with depth, with the representative loss rates of the warmer oxygenated epilimnion (NEP to shallow trap, Fig. 3) being lower than the colder anoxic deeper water column sections (shallow trap to sediment surface, Fig. 3). Preliminary results from in-trap decomposition experiments using Rostherne seston (Radbourne, unpubl. data) show little mineralization in a sealed container during the first 7 d after sedimentation. Therefore, we propose a week is enough time for sedimenting particles to be deposited in the shallow trap relatively intact (thus only a 1.4% loss found, Fig. 3), whereas, the particles sedimenting to the deep trap will take longer to be captured. The deep trap seston thus includes organic matter that has been partially mineralized within the water column during sedimentation, explaining reduced deep trap flux of OM despite colder and less oxic ambient waters at this depth.

Finally, autochthonous OC derived from algal production is generally regarded as labile compared to more refractory, allochthonous (terrestrial) OC. Sobek et al. (2009) found that BE was one-third that in lakes where OC was predominantly composed of autochthonous matter (mean  $BE_{150} = 22\%$ ) compared to lakes in which OC was composed predominantly of allochthonous inputs (mean  $BE_{150} = 66\%$ ). However, some eutrophic lakes that are dominated by autochthonous production, such as Rostherne Mere (this study), Baldeggersee (Teranes and Bernasconi 2000), and Kleiner Gollinsee (Brothers et al. 2013), can be highly efficient OC sinks, implying that OM source may not be a major control in all lakes. For example, previous work at Rostherne Mere has shown that the preservation of non-siliceous algae is excellent, potentially leading to higher burial rates with increasing production (Livingstone and Cambray 1978).

#### Quantifying OC burial efficiency

Recently, there has been considerable focus on quantifying burial rate to estimate the global role of lakes in removing C from the active carbon pool (Tranvik et al. 2009). Two

methods for calculating OC BE were employed in this study; Alin and Johnson (2007) compared recent (previous 10–25 yr) OC accumulation rate against NEP ( $BE_{25}$ ; Fig. 3), and Sobek et al. (2009) who used the long term mean OC accumulation rate (25–150 yr) and the delivery to the sediment surface (here the deep trap capture;  $BE_{150}$ , Fig. 3). When applied to Rostherne Mere using focusing-corrected core RML, the  $BE_{25}$  method gives an estimated efficiency of 92.8%, and  $BE_{150}$  60.4% (Fig. 3). This discrepancy of 32.4% (i.e., about half the  $BE_{150}$  value) highlights inadequacies in using either method in systems where OC production has not been constant. This indeed will be the case in many post-industrial landscapes where nutrient loading issues are now being addressed (such as under the EU Water Framework Directive; e.g., Denmark, Fig. 4c), in regions where production is increasing with progressive nutrient enrichment, and where terrestrial OC loading is increasing due to landscape or climate change, as in boreal regions (Evans et al. 2005; Monteith et al. 2007). For example, it is reported that Baldeggersee's OC delivery to the sediment surface (as net export from the epilimnion is very similar to deep trap collection; Muller et al. 2012) is between  $90 \text{ g C m}^{-2} \text{ yr}^{-1}$  and  $103.5 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Teranes and Bernasconi 2000; Muller et al. 2012), which implies steady burial of  $24.3\text{--}40.1 \text{ C m}^{-2} \text{ yr}^{-1}$ , given a BE of 23–45% ( $BE_{150}$  and  $BE_{25}$ , respectively) over recent decades. However, since the 1960s there has been a burial of  $\sim 50 \text{ g C m}^{-2} \text{ yr}^{-1}$ , rising to  $> 75 \text{ g C m}^{-2} \text{ yr}^{-1}$  in the 1990s (Teranes and Bernasconi 2000), and although some continued mineralization would be expected in the 1990s sediments, this demonstrates the methodological mismatch of comparing contemporary productivity with historical accumulation (during lower productivity).

There are two main differences between the two methods; the calculation of inputs (denominator in BE ratio) being either from NEP ( $BE_{25}$ ) or the deep trap catches ( $BE_{150}$ , i.e., surface sediment) and the choice of numerator in the BE ratio for the sediment core historical mean (10–25 yr or 25–150 yr;  $BE_{25}$  and  $BE_{150}$  respectively). At Rostherne Mere, it is the choice of historical record (BE numerator) that drives the burial efficiency value due to the changing production of the lake influencing the core mean value (Fig. 3), with the input (BE denominator) being very similar, as little OC is lost within the water column, although this will not be the case for all lakes. However, OC loss rates during sedimentation down the water column reported from eutrophic Swiss lakes up to 90 m deep have also been shown to be relatively minor, as here (Muller et al. 2012; Fig. 3).

Application of the Sobek et al. (2009) method ( $BE_{150}$ ) to lakes undergoing change in production (i.e., due to changes in nutrient loading, such as its reduction or redirection) will result in an unrepresentative BE, due to the changing OC accumulation rate (Fig. 4a,b). As the method uses average OC burial between 25 yr and 150 yr in age, this may include sediments that were deposited under very different

ecological or trophic conditions. In much of NW Europe and North America, lakes in agricultural landscapes over the last 100–150 yr have experienced progressive nutrient loading, greater allochthonous OC inputs and increasing eutrophication (Teranes and Bernasconi 2000; Brothers et al. 2013; Anderson et al. 2014; Clow et al. 2015; Heathcote et al. 2015). In Europe, for example, average OC burial rates have increased by a factor of 2.2 over the last 100–150 yr, with a significant rise in hyper-eutrophic lakes from a mean of  $59 \text{ g C m}^{-2} \text{ yr}^{-1}$  pre-1950 to  $\sim 100 \text{ g C m}^{-2} \text{ yr}^{-1}$  post-1950 (Smith 2003; Anderson et al. 2014).

Given widespread cultural eutrophication across the globe, it is unsurprising that similarly sharp increases in OC burial rates have been found elsewhere. Comparable patterns are found in nine Danish lakes (Fig. 4c), and although not corrected for sediment focusing, similar relative increases in OC burial have been reported from lakes in Mexico (threefold increase) and Germany (fourfold increase) in the modern period (Brothers et al. 2013; Carnero-Bravo et al. 2015). At Baldeggersee, Teranes and Bernasconi (2000), found an increase in OC burial rates, rising from  $15 \text{ g C m}^{-2} \text{ yr}^{-1}$  pre-1960 to  $103.5 \text{ g C m}^{-2} \text{ yr}^{-1}$  in 1995–1996. At Rostherne Mere a similar pattern emerges; with severe cultural eutrophication accelerating post-1900 resulting in a sevenfold rise in OC burial during the 20<sup>th</sup> century (Fig. 4a). Therefore, the application of the Sobek et al. (2009) method ( $BE_{150}$ ) to lakes undergoing change in production will underestimate burial efficiency by comparing contemporary nutrient-enhanced production with largely pre-impact burial rates, when production was commensurately lower too. Further, it is likely that preservation of OC in historical periods was in fact lower than in the contemporary system as the speed and severity of hypolimnetic deoxygenation will have increased with cultural eutrophication over the last 100–150 yr (exceeding any marginal increase in mineralization from warming hypolimnia in the last  $\sim 50$  yr; Dokulil et al. 2006), leading to greater underestimation using the  $BE_{150}$  method.

Recent oligotrophication is leading to reductions in production in many lakes as nutrient loading is controlled and reduced, as seen at Rostherne Mere (Fig. 4a) and in Denmark (Fig. 4c). At both Rostherne Mere and the nine recovering Danish lakes, a decline in OC burial rate begins following a reduction in nutrient loading (Fig. 4a,c). Comparison of the LIV77 core taken at Rostherne Mere in 1977 confirms the long-term pattern found in the RML core from 2011, and agrees closely with the focusing-corrected values (Fig. 4a), suggesting that mineralization losses do not continue after permanent deposition. Indeed, comparison of the burial rate and OC% for the two cores (Fig. 4a,b) suggests that mineralization is largely complete after  $\sim 10$  yr, in agreement with recent studies on lake sediments (Galman et al. 2008). While there will be some variability expected even from cores collected in close proximity (Rippey et al. 2008), the good agreement between these cores supports the approach of

using sediment focusing (independently applied to both  $^{210}\text{Pb}$ -dated cores) to estimate a basin-mean value from a single core. Moreover, the LIV77 core OC data also support the contention that OM mineralization effectively ceases  $\sim 10$  yr after deep water sedimentation in such lakes: OC burial rates in the LIV77 and RML cores from the 1960s are essentially the same (within methodological and within-basin variability), while OC%, initially higher in the uppermost section of the LIV77 core, falls to similar values in both cores by  $\sim 1965$  (Fig. 4b), as expected if OM mineralization was still incomplete at that time (Fig. 4b). The fall in productivity following reduced nutrient loading seen at Rostherne Mere and the nine Danish lakes is mirrored in the pattern of OC burial rate in their lake sediment records, despite mineralization processes in the uppermost sediments, in agreement with simulations of OC burial under various models of mineralization (Heathcote et al. 2015).

It is clear that the OC burial rate in lakes undergoing recent changes in nutrient loading will respond dynamically to changes in both production and preservation environment. However, as decomposition of organic matter continues after deposition onto the lake bed and during incorporation into the lake sediment record, the use of recent sediments (possibly  $< 25$  yr and certainly  $< 10$  yr in age; Galman et al. 2008) should be avoided due to potential continued diagenesis after deposition (Sobek et al. 2009). However, comparing contemporary production with pre-20<sup>th</sup> century sediment records is also problematic. Consequently, application of the Alin and Johnson (2007) method that uses a more recent time period of OC accumulation (10–25 yr mean) may result in a more realistic estimate for lakes where major changes in production have occurred over  $\sim 25$  yr, and have since stabilized. At Rostherne Mere and other lakes undergoing recent recovery over this timescale (cf. Danish lakes; Fig. 4c), it is clear from the trends of the OC burial rates (Fig. 4a,b) that recent diagenesis does not remove the signal of changes in recent lake production recorded in the sediment record. From Fig. 3, comparison of the 2011 sediment surface OC burial rate from the core ( $112 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) to the mean from 1985 to 2000 ( $126 \text{ g C m}^{-2} \text{ yr}^{-1}$ ; i.e., the  $\text{BE}_{25}$  method) shows a 12.8% higher historical burial rate, and generates a paradoxical BE over 100%. In this case, it is happenstance that the  $\text{BE}_{25}$  method included the period of maximum lake production, pre-sewage diversion, at this site. Nonetheless, this highlights the problems inherent in any such historical approach to estimating contemporary OC sedimentary dynamics in systems undergoing recent change.

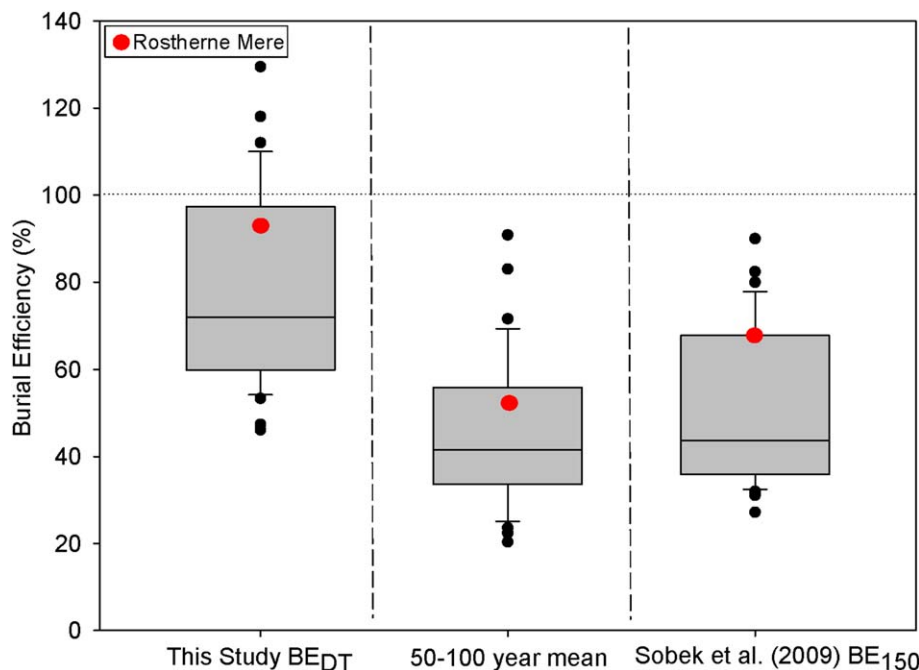
#### Updated OC burial method: $\text{BE}_{\text{DT}}$ and $\text{BE}_{\text{NEP}}$

This study suggests the current methods for estimating OC BE may be inappropriate when applied to lakes that have recently undergone, or are undergoing, changes in their trophic status and production due to anthropogenic

impacts (such as nutrient loading) or global change drivers (such as climate change) (Evans et al. 2005; Monteith et al. 2007). The implications for errors in up-scaling lake OC burial rates for regional and biome-scale C cycling without an effective methodology are substantial (Heathcote et al. 2015).

The fundamental issue with both methods discussed here ( $\text{BE}_{25}$  and  $\text{BE}_{150}$ ) is the calculation of a contemporary OC burial rate via a historical sediment mean OC burial value that will either underestimate BE in increasingly productive lakes (e.g., those becoming more nutrient enriched) or overestimate it in lakes that are recovering from eutrophication (Fig. 5). Therefore, here we propose adapting previous approaches for assessing the OC burial in lakes that are in a state of trophic flux, by reducing the historical dependency in the sediment core mean value by using the 0 to 50-yr mean (Fig. 3; labelled  $\text{BE}_{\text{DT}}$  and  $\text{BE}_{\text{NEP}}$ ), using delivery of OC to surface sediment as estimated from a deep trap or surface sediment accumulation ( $\text{BE}_{\text{DT}}$ ) or epilimnetic export of OC as estimated by NEP respectively ( $\text{BE}_{\text{NEP}}$ ). Using this time frame will reduce the historical dependency inherent in the  $\text{BE}_{150}$  method and capture the most recent lowered accumulation rates found in recovering lakes, addressing issues of the  $\text{BE}_{25}$  method. This new approach using surface sediment accumulation ( $\text{BE}_{\text{DT}}$ ) was applied to 36 stratifying ( $> 10$  m maximum depth) European lakes known to have been impacted by nutrient enrichment over the last 100–150 yr, extracted from the dataset of Anderson et al. (2014; Fig. 5). All these lakes have independently dated, focusing-corrected sediment records, with delivery to the sediment surface estimated from surface sediment accumulation rate. We argue that  $\text{BE}_{\text{DT}}$  values are better estimates of true, current BE than those methods using historical sediment data from earlier (pre- or early impact) periods, with mean  $\text{BE}_{\text{DT}} \sim 75\%$  compared to  $\sim 40\%$  using either sediment records from 50–100 yr or 25–150 yr ago (Sobek et al. 2009; Fig. 5). For example, Kleiner Gollinsee is estimated to have an efficiency of 28.5% using the  $\text{BE}_{150}$  method, inexplicably low compared to BE of  $\sim 100\%$  calculated from deep trap flux to sediment surface accumulation (Brothers et al. 2013). However, the  $\text{BE}_{\text{DT}}$  method proposed gives BE as 70.7%, fitting within the range of other eutrophic lakes (Fig. 5), and representing a more realistic value for contemporary BE. Similarly, Baldegersee (Teranes and Bernasconi 2000; Muller et al. 2012) is calculated with a BE of 41.8–48.1% ( $\text{BE}_{\text{DT}}$  and  $\text{BE}_{\text{NEP}}$ , respectively), compared to  $\text{BE}_{150}$  of  $\sim 23\%$ . While these are relatively low figures compared to other such lakes (Fig. 5), this may be due to over-trapping within the sediment traps as the reported net export from the epilimnion in Baldegersee in 1996 was  $90 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Muller et al. 2012), compared to delivery to the sediment surface (via deep trap collection) of  $103.5 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Teranes and Bernasconi 2000). Over-trapping will underestimate BE (by overestimating





**Fig. 5.** Burial efficiency (BE%) for 36 stratifying European lakes (> 10 m) that have been impacted by recent nutrient enrichment (data from Anderson et al. 2014). Three methods for BE% are calculated; this study's BE<sub>DT</sub> (last 50 yr of accumulation), 50 to 100-yr sediment core mean representing the early/pre-impact BE, and the BE<sub>150</sub> method (Sobek et al. 2009). All cores are <sup>210</sup>Pb-dated and focusing-corrected. Rostherne Mere's BE is marked with a red dot. Lakes > 100% BE are those with net oligotrophic recovery in the last 50 yr. See text for details.

production), which highlights the need for effective trap corrections, as mentioned previously.

Rostherne Mere's BE using this updated method is estimated as ~95% (BE<sub>DT</sub>; Fig. 3), and fits within the range of this larger dataset (Fig. 5). Some lakes are shown to have a BE<sub>DT</sub> > 100% (Fig. 5), which are those that have undergone rapid oligotrophication at some point over the last 50 yr, resulting in a higher mean organic carbon accumulation rate (OCAR) in the sediment core compared to the current sediment surface OCAR. To account for this, individual adjustments of the BE<sub>DT</sub> or BE<sub>NEP</sub> methods can be made to quantify the rate of trophic recovery over different time scales, by adjusting the sediment core mean OCAR date range. This adjustment will generate an improved representation of the sediment core OCAR mean, enabling a more realistic BE to be calculated.

This study has focused on a lake system dominated by autochthonous production and minimal terrestrial OC inputs. However, most of the world's lakes are boreal systems (Tranvik et al. 2009), and those with significant peatland, forest and organic rich soil in their catchments typically have high loadings of terrestrial DOC (Jonsson et al. 2001; Sobek et al. 2007; Ferland et al. 2012), while temperate lowland lakes, such as Rostherne Mere, have much less (due to both the lack of such sources of

terrestrial OC and the agriculture practiced in the catchment). The BE<sub>NEP</sub> method proposed in this study relies on autochthonous contribution representing the majority of total OC inputs to the lake, as such it will be susceptible to substantial overestimation of the total OC load. Therefore, research design for lake systems with a high terrestrial contribution must also consider that the BE<sub>NEP</sub> may not be a suitable method to use without adding the estimate for the terrestrial OC component. This can be done through direct field study of major inflows and lake DOC and POC pools or utilizing the literature to estimate load of OC. Below, we estimate terrestrial OC loadings to Rostherne Mere as a worked example of method alteration for systems with high terrestrial loading.

The allochthonous loading of DOC from Rostherne's catchment can be estimated from major river inflow and outflow concentrations from 2011 to 2012 (Scott 2014; 8 mg L<sup>-1</sup> DOC) and the inflow and outflow volumes measured in 2016 (Radbourne, unpubl. data) and up-scaled to account for missing sources (Carvalho et al. 1995), plus a small amount released from the lake sediment (Scott 2014), giving a total DOC loading of 3.67 g C m<sup>-2</sup> if all DOC was sedimented to the lake floor. Terrestrial POC (TPOC) input is more difficult to estimate but there is consensus that loadings are less than for DOC (Worrall and Moody 2014; Barry et al. 2016). Even

if catchment losses are set as high as 50% of values for DOC, given significant mineralization in transit to and while in the lake (Worrall and Moody 2014), we conservatively estimate a TPOC loading of  $6.88 \text{ g C m}^{-2}$  to the lake floor. Combining the DOC and TPOC values we estimate that terrestrial OC may account for  $10.55 \text{ g C m}^{-2} \text{ yr}^{-1}$ . This figure likely overestimates the contribution of terrestrial OC to the sediment traps and lake floor at Rostherne Mere as the lake is third in a chain in its catchment, which (although both smaller than Rostherne) would act both as sinks for TPOC and provide further opportunities for mineralization. However, even if terrestrial OC loading was this significant at Rostherne Mere, there would only be a minor effect on the calculation of  $BE_{NEP}$ , as adding the terrestrial OC loading of  $10.55 \text{ g C m}^{-2} \text{ yr}^{-1}$  to the NEP of  $135.6 \text{ g C m}^{-2} \text{ yr}^{-1}$  would change the BE from 86.2% to 80.0%.

The  $BE_{DT}$  method already accounts for terrestrial inputs as the deep trap collection will include all in-lake and terrestrial OC contributions. However, there must be a consideration of the relative importance of terrestrial OC inputs against resuspension in trap collections, especially during the winter high flow events (which are considered the dominant periods of terrestrial OC loading, as catchment runoff is at its highest). In this study, the removal of the winter collection corrected for resuspension in the deep trap, a suitable correction in systems with low terrestrial OC inputs and high winter resuspension, like Rostherne Mere, as evident in the shallow and deep trap correction comparison (see Table 2). Removal of this inter collection would also therefore preferentially remove terrestrial OC inputs and therefore underestimate the total OC input. To account for this discrepancy, an addition of the estimated terrestrial OC input during this winter period (as discussed above, based on catchment loading) could be included in the calculation.

It is clear this updated approach will contain some error due to the continued diagenesis of OC in the upper sediment before permanent incorporation into the sediment archive (Galman et al. 2008) and thus (under steady-state production) will on some level overestimate the true burial efficiency. However, as mentioned above, this may be only a minor issue in deep seasonally hypoxic lakes, such as Rostherne Mere (this study), Baldeggersee (Teranes and Bernasconi 2000) and Kleiner Gollinsee (Brothers et al. 2013), where OC mineralization rates are already suppressed. In other lake systems (e.g., warmer, shallower, less prone to stratification) this overestimation may be larger and needs further examination. While no method in lakes that are changing rapidly will be perfect, the approach proposed here does at least recognize the role of recent lake ecosystem history and attempt to take this into account, and can improve the estimation of lake OC burial efficiency over previous approaches.

## Conclusions and implications

Much of the current literature on the role of lakes in global C cycling assumes constant OC burial rates, but it is clear from the present study and others (Heathcote and Downing 2012; Anderson et al. 2013, 2014) that OC burial rates have fluctuated historically, increasing in lakes as they have become more eutrophic, but also declining following recovery (Fig. 4). It is evident that OC BE is subject to both the controls of preservation as well as production, and varies over a range of timescales, from seasonal to multi-annual and over longer timescales (decadal and centennial). Productive, stratifying lakes with seasonally anoxic hypolimnia also demonstrate that autochthonous OC, although labile, can be well preserved and buried in lake sediments over long time periods (Livingstone and Cambray 1978; Livingstone and Reynolds 1981).

Future work requires the wider utilization of long term lake monitoring programmes to understand further the extent to which seasonal, inter-annual and multi-annual variability and changing external stressors, such as nutrient loading and climate change, will have upon OC dynamics in lakes, such as increasing terrestrial OC loading through hydrological and land use change. A greater appreciation of the variable nature of OC burial rates will improve our understanding of C cycling in the large (and growing) number of impacted, non-steady state lakes and give greater confidence to up-scaling models that estimate the role of lakes as important regional and global sinks of OC. Furthermore, changes in autochthonous production are a key control on historical patterns of OC burial and need to be considered for a deeper understanding and evaluation of the role of lakes in global C dynamics.

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### Conflict of Interest

None declared.

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