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Lake eutrophication and its implications for organic carbon sequestration in Europe

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Abstract

The eutrophication of lowland lakes in Europe by excess nitrogen (N) and phosphorus (P) is severe because of the long history of land-cover change and agricultural intensification. The ecological and socio-economic effects of eutrophication are well understood but its effect on organic carbon (OC) sequestration by lakes and its change over time has not been determined. Here we compile data from ~90 culturally-impacted European lakes (~60% are eutrophic, Total P [TP] >30 µg P l^{-1}) and determine the extent to which OC burial rates have increased over the past 100 to 150 years. The average focussing corrected, OC accumulation rate (C AR_{FC}) for the period 1950–1990 was ~60 g C m^{-2} yr^{-1}, and for lakes with >100 µg TP l^{-1} the average was ~100 g C m^{-2} yr^{-1}. The ratio of post-1950 to 1900–1950 C AR is low (~1.5) indicating that C accumulation rates have been high throughout the 20th century. Compared to background estimates of OC burial (~5–10 g C m^{-2} yr^{-1}), contemporary rates have

increased by at least four to five fold. The statistical relationship between C AR_{FC} and TP derived from this study (r^2 = 0.5) can be used to estimate OC burial at sites lacking estimates of sediment C burial. The implications of eutrophication, diagenesis, lake morphometry and sediment focussing as controls of OC burial rates are considered. A conservative interpretation of the results of the present study suggests that lowland European meso- to eutrophic lakes with >30 μg TP l^{-1} had OC burial rates in excess of 50 g C m^{-2} yr^{-1} over the past century, indicating that previous estimates of regional lake OC burial have seriously underestimated their contribution to European carbon sequestration. Enhanced OC burial by lakes is one positive side-effect of the otherwise negative impact of the anthropogenic disruption of nutrient cycles.

Introduction

The eutrophication of lowland lakes is a widespread problem globally but is particularly serious in densely populated areas of Europe and North America (Smith, 2003). The disruption of the biogeochemical cycles of nitrogen (N) and phosphorus (P) has increased nutrient loadings on lakes and has profound effects on the biological structure (notably loss of macrophytes) and on ecosystem productivity (Jeppesen *et al.*, 2000) with associated substantial socio-economic costs (loss of amenity value, increased water treatment costs; (Pretty *et al.*, 2003)). While the negative effects of nutrient enrichment are considerable, one possible positive aspect in terms of regional carbon (C) budgets and environmental sustainability is the increased rate of organic C burial by lakes as a function of increased lake productivity. As atmospheric CO_2 concentration continues to increase and terrestrial sequestration declines, for example due to forest maturation, other biosphere sinks will become increasingly important: a possible role of lakes as C sinks is only now being evaluated (Buffam *et al.*, 2011, Ferland *et al.*, 2012).

A significant role for lakes in the terrestrial C cycle has recently been proposed, particularly in relation to their capacity to process terrestrially-derived C (Tranvik *et al.*, 2009). As a result of the input of dissolved organic carbon (DOC), many lakes are considered net heterotrophic, i.e. ecosystem respiration is greater than production, certainly at DOC concentrations >10 mg C l^{-1} (Algesten *et al.*, 2004). Although lakes emit considerable amounts of CO_2 into the atmosphere, they are also important as long-term sinks for C, particularly in lake-rich landscapes where there is a considerable C-pool in lake sediments (Anderson *et al.*, 2009, Ferland *et al.*, 2012, Kortelainen *et al.*, 2004).

Burial of organic carbon in lakes is a function of primary productivity in the lake itself, inputs from the catchment as DOC, particulate organic C (POC), and loss processes such as respiration within the water column and in sediments as well as downstream transfer (Cole *et al.*, 2007, Sobek *et al.*, 2009). Other factors that can influence organic matter preservation and which can vary by region and lake type are bulk sediment accumulation rates, hypolimnetic anoxia, temperature and faunal density. The extent to which eutrophication has increased organic C-burial by European lakes has not been addressed systematically although it is an implicit assumption in many palaeolimnological studies (Battarbee *et al.*, 2012).

Estimates of C burial rates vary considerably although Tranvik *et al.* (2009) considered the global average to be 10–15 g C m^{-2} yr^{-1}. An early estimate of the global mean (5 g C m^{-2} yr^{-1}) (Stallard, 1998) is comparable to that found in Finnish and Arctic lakes during the Holocene (Anderson *et al.*, 2009, Kortelainen *et al.*, 2004) but more recent estimates suggest that land-use change and eutrophication have increased burial rates in more overtly cultural landscapes (>60 g C m^{-2} yr^{-1}) (Anderson *et al.*, 2013, Heathcote & Downing, 2012). Battin *et al.* (2009) highlighted the need for a better estimate of carbon accumulation by aquatic ecosystems. Moreover, there has not been an explicit consideration of recent temporal change in mean organic C burial rates in response to cultural eutrophication in Europe. Here we compile data from ~90 European lakes (Table 1) and determine the extent to which organic carbon burial rates have increased over the past 100 to 150 years in relation to nutrient enrichment. We show that for European meso- to eutrophic lakes organic C burial rates reflect P availability and are considerably higher than previously thought, which has clear implications for the role of lakes in estimates of regional C sequestration.

Material and Methods

Study sites

The data from 93 lakes used in this analysis are mainly from national environmental agency and European Union projects that were explicitly addressing effects of cultural eutrophication (Anderson, 1997, Bennion *et al.*, 2011a, Bennion *et al.*, 2004, Bennion & Simpson, 2011, Bennion *et al.*, 2011b) and as a result are not a random selection of lakes in Europe. Importantly, the lakes do, however, cover a range of lake typologies, areas, maximum and mean depths, and nutrient (TP) concentrations (Table 1) (see Supplementary Information Table 1 for details).

The lakes are mainly within the temperate climatic zone, located in 11 countries across Europe from Norway to the Alps, spanning a latitudinal range from 44.22 to 60.77° N. The majority of the lakes are softwater systems with the exception of some of the lakes situated in carbonate bedrock catchments in Central Europe. Most of the lakes lie in lowland (< 200 m asl) catchments, with the exception of several Swiss lakes and a small number of higher elevation sites in France and Slovenia. The catchments are largely productive with nutrient loading from either point sources such as sewage treatment works and/or diffuse sources from agriculture. Several lakes have also experienced catchment erosion as a result of forestry activity (planting and felling). The dataset covers a wide range of lake surface areas but sites are mostly < 5 km^2 with the exception of some very large lakes in Switzerland and Scotland, and Lake Mjøsa in Norway (Table 1). Similarly, lakes span a broad range in maximum water depths from 1.5 to 453 m. The oligotrophic lakes tend to be the deepest while the nutrient-rich sites are generally shallow with maximum depths < 20 m. Importantly, for the purposes of this study, the lakes cover a wide range of TP concentrations from <5 to 350 μg l^{-1} (annual mean), spanning the full trophic gradient from oligotrophic to hypertrophic conditions (Table 1). When classified based on the OECD boundaries for annual mean TP there are 11 oligotrophic, 27 mesotrophic, 36 eutrophic, and 19 hypertrophic lakes.

With the exception of five large Scottish lochs and Upton Broad in Norfolk, all of the study lakes have experienced some degree of ecological turnover in their fossil diatom records indicative of eutrophication, (Anderson, 1997, Bennion *et al.*, 2001, Bennion *et al.*, 2004, Lotter, 1998). Palaeolimnological studies show that these lakes have seen marked shifts in their diatom assemblages since ~1850 typically from one characterised by dominance of *Cyclotella* taxa associated with nutrient-poor waters, through *Asterionella formosa*, *Fragilaria crotonensis,* and *Aulacoseira* spp. associated with mesotrophic waters, to high relative abundance of *Stephanodiscus* and *Cyclostephanos* taxa associated with nutrient-rich conditions in the most enriched sites (Hörnström, 1981, Willén, 1991).

Methods

Sediment cores were taken primarily with piston corers, but in some of the oligotrophic lakes a gravity corer was used. Full details of the coring methods used in individual studies can be found in source reports and papers. Cores were processed using standard methodology (see (Anderson, 1997, Bennion *et al.*, 2004, Lotter, 1998) and only briefly outlined here. Cores were either sectioned in the field or returned to the laboratory for sectioning, samples were stored cold until analysis for dry weight (overnight at 105° C) and organic matter (OM: loss-on-ignition at 550 °C for 2-hours) (Heiri *et al.*, 2001). With the exclusion of the Swiss where varve counting was used (Lotter *et al.*, 1997), all cores were dated using ^{210}Pb determined using gamma counting (Appleby *et al.*, 1986). Dry mass accumulation rates (DMAR) and chronologies were determined using the constant rate of supply model, which is applicable to lakes with increases in bulk sedimentation rate due to catchment disturbance and eutrophication. Full details of the dating models are given in the source papers and reports. Organic content was converted to organic carbon (OC) for each lake by multiplying the percentage of OM by a factor of 0.468 (Dean, 1974). The OC accumulation rate (C AR) was calculated by multiplying the DMAR by the estimated OC content. In our study we did not include the inorganic carbon that is present as carbonates in some of the hardwater lakes.

A single core taken in the central, deepest part of a lake will generally over-estimate the basin wide average sedimentation rate due to the process of sediment focussing. This problem is best addressed by multiple coring (Engstrom & Rose, 2013, Rippey *et al.*, 2008) but this is an extremely labour intensive approach and cannot realistically be applied to regional studies of more than 10 lakes (for a notable exception see Kortelain et al. 2004). An alternative approach, increasingly used in pollution studies, is to use the ratio of the mean sediment ^{210}Pb flux to the regional atmospheric ^{210}Pb flux to derive a focussing factor (Engstrom & Rose, 2013). This approach assumes that the atmospheric flux is well prescribed, which is not always true, but it can be approximated by assuming that it is proportional to rainfall, around 100 Bq m^{-2} yr^{-1} per ~1000 mm yr^{-1} of precipitation. This approach was adopted here to calculate a lake specific focussing factor (based on ^{210}Pb flux data and approximate average annual rainfall for each lake), which was subsequently used to provide a focussing-corrected C AR (C AR_{FC}). ^{210}Pb flux data were not available for 15 lakes and therefore the C ARs for these lakes were not focussing corrected, and these sites were excluded from all but the increase ratio calculations (see below).

Decomposition of organic matter continues after deposition onto lake sediments (Sobek *et al.*, 2009) but the rate decays exponentially with most of the C mineralization taking place within 10 years of initial deposition (Galman *et al.*, 2008). In practical terms this means that estimates of organic C burial that include surficial sediments will over-estimate the long-term burial rate. To evaluate this problem we compared three estimates of recent C-burial: first, the uppermost 10 years (as determined by ^{210}Pb-dating), second the post-1950 mean with the surface 10-yrs excluded and finally, the post-1950 mean with the most recent 20-yrs excluded from the period mean. The average C AR was then calculated for each lake for three time periods: pre-1900, 1900–1950 and post-1950 (with the uppermost sediment excluded). Increase ratios were simply calculated as the ratios of mean C AR values for all lakes (i.e. including those without focussing correction as the latter is a lake specific constant): post-1950:pre-1900 and post-1950:1900–1950. In this study, 1950 was used as the dividing date for these time periods as the results of the diatom analyses (e.g. Anderson, 1997; Bennion *et al.,* 2004; Lotter, 1998, 2001) indicate that most of these lakes underwent rapid and severe eutrophication after this time, primarily as a result of the intensification of agriculture and the expansion of phosphate fertilizer use in Europe (Foy & Withers, 1995). As sedimentation rates in lakes vary considerably and can be very high in eutrophic systems (>1 cm yr^{-1}) (Rose *et al.*, 2011) some of the sediment cores used in this study do not include sediment older than ~1900 A.D., occasionally only covering ~50 years. Mean organic C AR are reported for OECD TP categories (see Study sites above) rather than geographic region as coverage is not uniform across Europe (see Table 1).

Results

Carbon accumulation rates derived from individual lake sediment cores range from 2–3 g C m^{-2} yr^{-1} in oligotrophic Loch Maree (Scotland) and Lake Mjøsa (Norway) in the late 1800s, to values >300 g C m^{-2} yr^{-1} in a number of eutrophic lakes after 1950 A.D. Mean C AR_{FC} of all lakes increased from ~17 g C m^{-2} yr^{-1} in the 19th century to ~40 g C m^{-2} yr^{-1} in 1900–950 and to ~60 g C m^{-2} yr^{-1} post-1950 (Fig. 1) although the number of lakes where it was possible to calculate pre-1900 A.D. rates was low (n = 35). The mean C AR_{FC} was lowest for meso-oligotrophic lakes (< 30 µg TP L^{-1}) with values of 22 and ~33 g C m^{-2} yr^{-1} for pre and post-1950 periods, respectively. The mean C AR_{FC} were somewhat higher for eutrophic lakes (30–100 µg TP L^{-1}) with values of 50 and ~73 g C m^{-2} yr^{-1} for pre and post-1950 periods, respectively, and were highest for hypertrophic lakes (>100 µg TP L^{-1}) with values of 59 and ~100 g C m^{-2} yr^{-1} for the pre and post-1950 periods, respectively (Fig. 2). The comparison of the 1950–1990 mean organic C burial rates after removal of the 10 and 20 years values prior to the coring date suggest that the values used here (Figs 1–3) differ by approximately a 5% and 10% (See Supplementary Information Table 1 for details).

The organic C burial rate increased (i.e. the ratio of post-1950 to 1900–1950 C AR) during the 20th century on average by a factor of 1.65x (all lakes; Fig. 3) and the ratio was similar (~1.6) to that for lakes placed into OECD TP categories (cf. Fig. 2). The C burial rate increased on average by a factor of 2.2 (all lakes) if the post-1950 period is compared with the pre-1900 period. The recent 10-yr C AR_{FC} at each lake is correlated with both contemporary TP (r^2 = 0.5) and maximum water depth (r^2 = 0.45) (Fig. 4). The mean C AR_{FC} for the period 1950–1990 at each lake is also correlated with contemporary in-lake TP concentration (r^2 = 0.47).

Discussion

Lakes are increasingly seen as playing an important role in the terrestrial C-cycle (Tranvik *et al*. 2009) and although the long-term storage of OC is documented, there has been limited consideration of rates of change in the organic C-burial rates (Anderson *et al.*, 2013, Heathcote & Downing, 2012): limnologists and biogeochemists have tended to see lakes as static in terms of the C-burial rate. The role of lakes in the terrestrial C cycle has focussed mainly on CO_2 emissions (with emphasis on terrestrial DOC-loads and lake heterotrophy), C mineralization processes in sediments and to a lesser extent, whole-lake C-budgets (Jonsson *et al.*, 2007, Sobek *et al.*, 2003, Sobek *et al.*, 2006, Sobek *et al.*, 2005). Many estimates of lake C burial are derived from mass balance budgets and the burial component is usually derived as the difference between total inputs and losses, an approach which can involve accumulated errors (Sobek *et al.*, 2006). Sediment cores provide an alternative, comparative approach and extend the temporal perspective.

C burial rates and temporal variability

It is clear from the present and other studies (Anderson *et al.*, 2013, Heathcote & Downing, 2012) that organic C burial rates have increased in lowland lakes in the 20th century (Figs 1–3). The highest C AR_{FC} rates recorded in this study were >200 g C m^{-2} yr^{-1} comparable to those observed in two North American studies that focussed on lake eutrophication (127–

144 g C m^{-2} yr^{-1}) (Anderson *et al.*, 2013, Heathcote & Downing, 2012). The mean rate determined by Heathcote & Downing (2012) was 88 g C m^{-2} yr^{-1} (n = 8), higher than the mean rate observed in this study (~60 g C m^{-2} yr^{-1}) for the post-1950 period (n = 78 lakes) and also that of Anderson *et al*. (2013) for agricultural-impacted lakes in central-southern Minnesota (~50 g C m^{-2} yr^{-1}) (n = 116). Hypertrophic European lakes in this study, however, have a mean C AR_{FC} of ~100 g C m^{-2} yr^{-1} (Fig. 2).

Process considerations: sediment focussing, post-depositional mineralization and burial efficiency

Although there is considerable interpretative power associated with using sediment cores to derive past C-burial rates and their temporal variability, there are some significant problems with the approach: sediment focussing, variable burial efficiency and post-depositional mineralization. Lake depth influences sediment focussing (see below) but also affects burial efficiency. Contemporary C AR_{FC} declines with increasing water depth (Fig. 4b) suggesting a possible role for enhanced mineralization in the water column; however, in the present study larger lakes also have lower nutrient TP concentrations (r = –0.58) as has been observed elsewhere (Nürnberg 1996 and references therein) making it difficult to disentangle the effect of production versus loss processes.

Sediment focussing represents a major problem for quantitative palaeolimnology and whole-lake element budgets (Engstrom & Rose, 2013, Molot & Dillon, 1996). The uncorrected mean (1950–1990) C AR observed in individual study lakes ranged from ~7 to >700 g C m^{-2} yr^{-1}, focussing correction reduces these lake specific rates to ~3 and 300 g C m^{-2} yr^{-1} respectively. The ^{210}Pb flux correction method used in this study (Engstrom & Rose,

2013) reduced the C AR by ~45% on average. For a discussion of the problems of sediment focussing relevant to contemporary C-budgets see Buffam *et al*. (2011).

Although lake sediments are efficient at burying OC, a variable and sometimes considerable fraction will be mineralized in the surficial sediments prior to long-term burial (Sobek *et al.*, 2009). These high values are derived from sediments younger than 5–10 years and as has been demonstrated, maximum OC mineralization rates occur in sediments younger than 5 years but then drop dramatically after ~10 years (Galman *et al.*, 2008, Hamilton-Taylor *et al.*, 1984, Teranes & Bernasconi, 2000). The C AR_{FC} from Skanderborg in Denmark illustrates this clearly: burial rates in the surficial sediments are ~3.5 times the average post-1950 C AR (Fig. 5; see also Loch Maree, Fig. 6). Burial efficiency varies with a range of factors but the source (nature and type) of the OM (i.e. terrestrial vs aquatic), the sedimentation rate and the time OM is exposed to oxygen during burial are the most important (Sobek *et al.*, 2009).

Sediment mineralization is time dependent, largely controlled by OM contact with O_2. Sedimentation rates increase as eutrophication proceeds (Rose *et al.*, 2011), which increases the rate at which fresh OM is removed from the reactive surface sediments, enhancing the C burial efficiency (Sobek *et al.*, 2009). Models of mineralization rates in sediments, which emphasize respiratory losses, use a steady state approach with a constant OM input. This is clearly an invalid assumption for lakes undergoing eutrophication as productivity will have increased resulting in a greater C burial rate (see trends in Figs 5–7), it is unclear to what extent the microbial processing of OM input can keep up with the input and offset the higher burial rates. Excluding the surficial reactive sediments younger than ~10 and 20 yrs from the post-1950–1990 period mean had only a small influence on the estimated rates, reducing them by 5 and 10% respectively (See Table S1). In a study of C-burial in 116 Minnesotan lakes using a similar approach, Anderson *et al*. (2013) found a ~10% reduction.

At Frederiksborg Slotssø (Denmark) the post-1950 uncorrected C-burial rate is ~146 g C m^{-2} yr^{-1} but with focussing correction this was reduced to ~58 g C m^{-2} yr^{-1} (Fig. 5). Andersen *et al.* (1979) estimated gross primary productivity to be 560 g C m^{-2} yr^{-1} in this lake with an organic flux to the sediments of 154 g C m^{-2} yr^{-1} with sediment/benthic respiration resulting in a permanent burial of ~24 g C m^{-2} yr^{-1}. Given the two contrasting approaches, these estimates of carbon burial (24 versus 58 g C m^{-2} yr^{-1}) are reasonably similar.

While post-burial mineralization rates are high and consume a significant proportion of the OC input (Sobek *et al*., 2009), it is equally clear that the temporal trends in C AR reported here from a range of lake types cannot be explained solely in terms of mineralization losses over time (Figs 5–6). Moreover, the lakes compiled in this study cover a range of types (maximum depth ranges from < 10 to >400 m) and limnological conditions (Table 1) including lakes with severe seasonal, hypolimnetic anoxia. The organic C burial rates with focussing correction reported here are, therefore, a reasonable approximation of long-term (i.e. decadal) burial rates. For eutrophic lakes, at least, post-burial mineralization losses are relatively low after ~10 years. The extent to which progressive eutrophication and anoxia will increase methanogensis and CH_4 release is beyond the scope of this study (Bastviken *et al.*, 2004).

Drivers of eutrophication in Europe

Although a focus of intense research in the 1970s, eutrophication has not dropped off the environmental agenda, in part because although point-sources are now well controlled in many countries, the problem of diffuse nutrient loading on lakes has re-ignited issues of farming sustainability (Carpenter *et al.*, 1998), as has the question of what ultimately is the controlling nutrient (N or P) (Schindler, 2012). The problem of excessive nutrient use continues to represent a major ecological and water quality threat for many areas (Smith *et al.*, 1999).

The TP-C AR_{FC} relationship (Fig. 4) and the similar timing of regional C AR and European N and P fertilizer increase (Fig. 7) strongly suggest cultural eutrophication as the primary driver of the increased C burial rates. The eutrophication of Europe's surface waters brought about by intensive use of lowland landscapes for human settlement, industry, and agriculture is well documented (Foy *et al.*, 2003). Although there are examples of lakes that first began to be enriched by human activity many centuries and even millennia ago (Bradshaw *et al.*, 2005, Fritz, 1989), an analysis of palaeoecological data from ~100 European lakes indicates that eutrophication for most lakes in Europe occurred from the middle and late 19th century with often a more pronounced eutrophication phase from ~1950 A.D. (Battarbee *et al.*, 2011). For the presently eutrophic and hypertrophic lakes in this study, C AR_{FC} were typically < 40 g C m^{-2} yr^{-1} prior to 1950 but have experienced a marked and continual increase since this time to peak values of ~100 g C m^{-2} yr^{-1} in the 1990s (Figs 2 and 7). This temporal pattern is coincident with the increased use of fertilizers in Europe since 1950, from ~1 x 10^6 tonnes P to a peak of ~6 x 10^6 tonnes P in the 1980s (Fig. 7). The temporal pattern of changing P fertilizer use (as opposed to that of Nitrogen fertilizer) and C AR (Fig. 7) provides some support for the inference that P inputs and hence increased primary productivity determine C burial in many European lakes. This inference is also supported by the comparison of mean C AR_{FC} increases across the OECD trophic status classes such that oligotrophic and mesotrophic lakes have the lowest rates (~30 g C m^{-2} yr^{-1} post-1950) and hypertrophic lakes the highest (~100 g C m^{-2} yr^{-1} post-1950) (Fig. 2).

In this study, C AR_{FC} are correlated with in-lake TP concentration (r^2 =0.5, n = 79; Fig. 4). The fit to TP is, however, poorer than observed in a similar study of Minnesotan lakes (r^2 = 0.60, n = 116; Anderson *et al.*, 2013). Some of the scatter will be due to comparing the contemporary in-lake TP concentration (often only a limited number of samples) with decadal C AR. Moreover, C AR is not only a function of in-lake nutrient availability (and hence aquatic productivity) it also represents inputs of terrestrial C and loss processes, notably burial of soil OC (Quinton *et al.*, 2010). Although soil erosion tends to be severe with the initial forest clearance, there is substantial topsoil losses with the intensification of contemporary agricultural (Alström & Bergman, 1988). While lakes are very efficient at burying terrestrial C, lateral transfer of soil organic matter brings nutrients that can increase aquatic primary productivity (Quinton *et al.*, 2010, Smith *et al.*, 1999). Another problem is the lack of good atmospheric ^{210}Pb flux data to correct the sediment flux; in the Minnesota study by Anderson *et al.* (2013), the atmospheric ^{210}Pb flux was measured, here it is only estimated. Other factors will be related to variable hydrological retention time and some lakes may be N-limited. In broad terms, however, the positive relationship between in-lake TP and C AR_{FC} (Fig. 4) demonstrates the effect of eutrophication on C burial.

Over the past few decades, a significant improvement in water quality in Europe has resulted from reductions in point source pollution via improved wastewater treatment, reduced volumes of industrial effluents and reduced or banned phosphate content in detergents (European_Environment_Agency, 2012). The downturn in C AR_{FC} for enriched lakes over the past two decades, coincident with the decline in fertiliser use, suggests that an effect on C AR is already being seen (Fig. 7).

The “recovery” trends in C AR seen at some lakes provide further support for the idea that C accumulation rates derived from sediment cores are a faithful approximation of changing burial rates, i.e. the temporal change recorded cannot be dismissed as artefacts of post-burial mineralization (see above). Comparing C AR_{FC} with long-term trends of epilimnetic TP concentration in specific lakes confirms this view (Fig. 5). Denmark, as with many parts of Europe, has undergone substantial improvements in waste water treatment, both from point sources and more diffuse sources in agricultural landscapes (Kronvang *et al.*, 1993). Ravn Sø and Knud Sø are deep (> 25 m), monomictic lakes where point-source nutrient redirection and catchment management plans have substantially reduced TP loadings on the lakes (Fig. 5). At both sites there is clear reduction in the C AR_{FC} following nutrient reduction in the late-1970s (Fig. 5) and the timing agrees quite well with the oligtrophication of the lakes. Such responses are not observed at all lakes with nutrient loading reduction, possibly due to the effects of internal nutrient loading (cf Skanderborg; Fig. 5). The lack of a direct response to nutrient reduction may also be, in part, a function of sedimentation processes that vary among lakes (auto- and allochthonous sources C-inputs; resuspension hypolimnetic anoxia, mean depth).

The River Basin Management Plans, produced for the European Union Water Framework Directive (WFD), reveal that diffuse pollution from agriculture is a significant pressure in approximately one third of European lakes, and is one of the chief causes of 44 % or almost

6500 lakes failing to achieve good ecological status or potential (EEA 2012). There is a strong regional pattern in the data with the highest pollution pressures and consequently worst ecological status reported in north-western Europe and the lowest pressures and best ecological status in Austria and Scandinavia where the population density and share of agricultural land are relatively low. For example, the total area-specific load of nitrogen (kg N ha^{-1} yr^{-1}) in north-western European catchments is more than double that in the Nordic countries and Baltic States, with a similar pattern observed for phosphorus. The EEA (2012) state that while wastewater treatment must continue to play a critical role in the protection of Europe's water, increased attention must be given to agricultural sources of nutrients if good water quality and ecological status is to be achieved. Although there are considerable environmental problems associated with eutrophication (water treatment, biodiversity loss, ecological change, loss of amenity value) (Pretty *et al.*, 2003, Smith *et al.*, 1999), it is clear that enhanced OC sequestration is one of the few positive aspects of the problem: eutrophic lakes are sequestering more OC than at any other time in their history. The elevated burial rates observed in this study (Fig. 2) are similar to those observed in most other culturally-impacted sites (Anderson *et al.*, 2013, Heathcote & Downing, 2012).

A detailed analysis of the effect of climate change on lake eutrophication and C burial is complex (e.g. catchment hydrology, nutrient losses, thermal stratification, anoxia, internal P-release (Battarbee & Bennion, 2012)) and is beyond the scope of this study. However, Gudasz *et al*. (2010) have suggested that warming will increase mineralisation of organic matter in littoral sediments but stronger and longer periods of stratification may enhance hypolimnetic anoxia and hence reduce respiratory losses. In contrast, warming may also promote longer ice free seasons (and more productivity if nutrients permit), greater methanogenesis, and reduced burial efficiency (Sobek *et al.*, 2009). Eutrophication coupled with climate change has also resulted in enhanced growth of cyanobacteria and they may be more labile than other algae during sedimentation and initial burial (Dong *et al.*, 2012). However, Anderson *et al*. (2013) could find only a very minor climate effect on C burial after the removal of nutrient gradients in a study of Minnesotan lakes.

Not all of the lakes in this study have agricultural catchments. Therefore the high C AR_{FC} can reflect other factors, for example, sedimentation of terrestrial carbon from forested catchments and input of soil POC due to catchment disturbance (Jonsson *et al.*, 2007). This situation is illustrated by C AR_{FC} in large Scottish lochs: these relatively remote, deep lakes often have low in-lake TP concentrations (<10 μg P L^{-1}) but C AR_{FC} >20 g C m^{-2} yr^{-1} (e.g. Lochs Awe, Eck and Lubnaig) (Fig. 6). These C burial rates are low in the context of the majority of lakes in this study but are still higher than the global average proposed by Tranvik *et al.* (2009). These lakes have been impacted by agro-forestry practices where catchment ploughing and fertilization of seedlings results in altered hydrology, increased lateral transfer of both DOC/POC as well as nutrient runoff. Loch Maree (Fig. 6) has some of the lowest C AR_{FC} (2–4 g C m^{-2} yr^{-1}) with only slight increases in burial rates from 1840 A.D. to present. This lake has an unproductive catchment and the diatom assemblage has changed little over the past 150 years (Bennion *et al.* 2004). Our data indicate, therefore, that C ARs have remained relatively stable some sites where enrichment has not occurred. Moreover, depending on catchment bedrock and groundwater input leading to elevated DIC in relation to DOC, the amount of inorganic carbon (as $CaCO_3$) in hardwater lakes may also represent a substantial sink of carbon in these lakes (Finlay *et al.*, 2009, Mueller *et al.*, 2006).

Climate, DOC and nutrient loadings have all been changing simultaneously throughout NW Europe over recent decades (Clark *et al*. 2010) so there may be complex effects on OC burial in lakes. For example, the impact of this DOC increase on OC flocculation rates in lakes is presently unknown. There is, moreover, an interaction among between DOC, light and productivity; for example, Nurnberg & Shaw (1998) demonstrated a positive relationship between DOC and TP but high DOC can restrict primary productivity through its impact on in-lake light climate. Unfortunately, estimates of DOC concentration were not available for all the lakes used in the present study, so it was not possible to consider its effect on OC burial relative to that of changing nutrients and/or lake morphometry. There was, however, only a very weak statistical effect of DOC on OC burial (r^2 = 0.08) in a comparable study in Minnesota (Anderson *et al*. 2013).

Implications for regional upscaling

Kastowski *et al.* (2011) and Luysseart *et al*. (2012) estimated total annual C burial by European lakes to be 1.25 x 10^{12} g C and 26 x 10^{12} g C, respectively. The results of the present study would suggest that these are probably underestimates of contemporary C-sequestration by European lakes. Luyssaert *et al*. (2012) concluded that in Europe lakes are second to forests in sequestering C; but echoing Battin *et al.* (2009) they state that “C-burial estimates are based on very little observational evidence” (Luyssaert *et al*. 2012, *their* suppl. info). Luysseart *et al*. (2012) used the long-term global average C-rates cited by Tranvik *et al*. (2009) together with the lake area estimates from Saarnio *et al*. (2009) and hence essentially ignored small lakes. As has been shown here and in other studies, the high burial rates associated with small lakes can be important. The regression relationship for in-lake TP-C AR_{FC} derived from this study is quite robust (r^2 = 0.50) and could be used to estimate C burial at lakes with no sediment C burial data.

Kastowski *et al*. (2011) claimed that their results indicate that the C AR of small European lakes is significantly lower than global estimates but as they excluded recent time periods in their study there seems little basis for this inference. As shown here, C-burial by European lakes increased in the 20th century (Figs 2, 6) as the result of eutrophication and rates are substantially greater than the global average. Kastowski *et al*. (2011) also ignored sediment focussing and so their rates and the associated upscaling should be treated with circumspection.

The rates from lowland eutrophic lakes in this study (Figs 1-3) would suggest that the rates cited by Tranvik *et al.* (2009) and used in the Luyssaert et al. (2012) study are probably too low for contemporary lake ecosystems in Europe and hence the lake contribution to regional C-budgets is being underestimated. The mean post-1950 C burial rate observed in this study for all lakes (~60 g C m^{-2} yr^{-1}) is ~4–5 x higher than the global average given by Tranvik *et al*. (2009). Although this European average rate is greater than previously estimated it is not surprising given the extent of lake eutrophication globally (Schindler, 2012, Smith *et al.*, 1999). Anderson *et al.* (2013) reported similar findings for post-European settlement C burial in Minnesotan lakes, the average post-1950 C AR is about 3x the pre-settlement rate. A conservative interpretation of the results of this study would be that meso-eutrophic lakes with >30 μg TP l^{-1} will have had long-term, decadal C burial rates of ~30 g C m^{-2} yr^{-1} during much of the 20th Century (Fig. 7), suggesting that the annual C-sequestration by European lakes is about 3x that estimated by Luyssaert *et al*. (2012). The increase ratio (post-1950:1900–1950; Fig. 3) of around ~1.5x also suggests that the major increases in C AR above quasi-“natural” or background rates occurred before 1900.

Lakes that have undergone eutrophication are primarily burying autochthonous carbon, i.e. CO_2 drawdown directly from the atmosphere (Hanson *et al.*, 2004, Staehr *et al.*, 2012). However, one important aspect not considered in detail in this study is the lateral transport of terrestrial [soil] C (Quinton *et al.*, 2010). Although on one level this is irrelevant as OC buried by lakes, regardless of source, is effectively removed from the C-cycle unlike the temporary storage of carbon in catchment soils; soil erosion shortens soil-C turnover time (Quinton *et al.*, 2010). Lateral movement of soil organic matter also acts as a nutrient transfer (N, P) which can drive further increases in lake productivity (eutrophication) as well as enhancing the burial of soil C. Organic carbon burial rates have increased substantially during the 20th century, by more than 5x in some lakes. These high C burial rates are likely to be maintained in the near future due to the rise of the P-saturation in agricultural catchments.

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Figure legends

Fig. 1 Box plots of mean C AR_{FC} [organic C accumulation rate with focussing correction] (for all lakes) for three time periods: pre-1900, 1900–1950 and post-1950 (see text for details). The thick horizontal line is the mean.

Fig. 2 Box plots of mean C AR_{FC} by TP class (oligo-mesotrophic: <30 µg TP l^{-1}; eutrophic 30-100 µg TP $l^{-1}$and hypertrophic >100 µg TP l^{-1}) and for two time periods within each class: 1900–1950 and post-1950 (see Figure 1 for details of the box plots). Abbreviation as Figure 1.

Fig. 3 Box plots of comparative C AR increase ratios for all lakes for two time periods: post-1950 to pre-1900 and post-1950 to 1900–1950 (see Figure 1 for details of the box plots).

Fig. 4 a. Surface mean C AR_{FC} versus contemporary in-lake epilimnetic TP concentration; Log[C AR] = 0.533[logTP] + 0.844; R^2 = 0.5 (p<0.001). B. Surface mean C AR_{FC} versus water depth: Log[C AR] = -0.483[logWater Depth] + 2.1388; R^2 = 0.45 (p<0.001). Abbreviation as Figure 1.

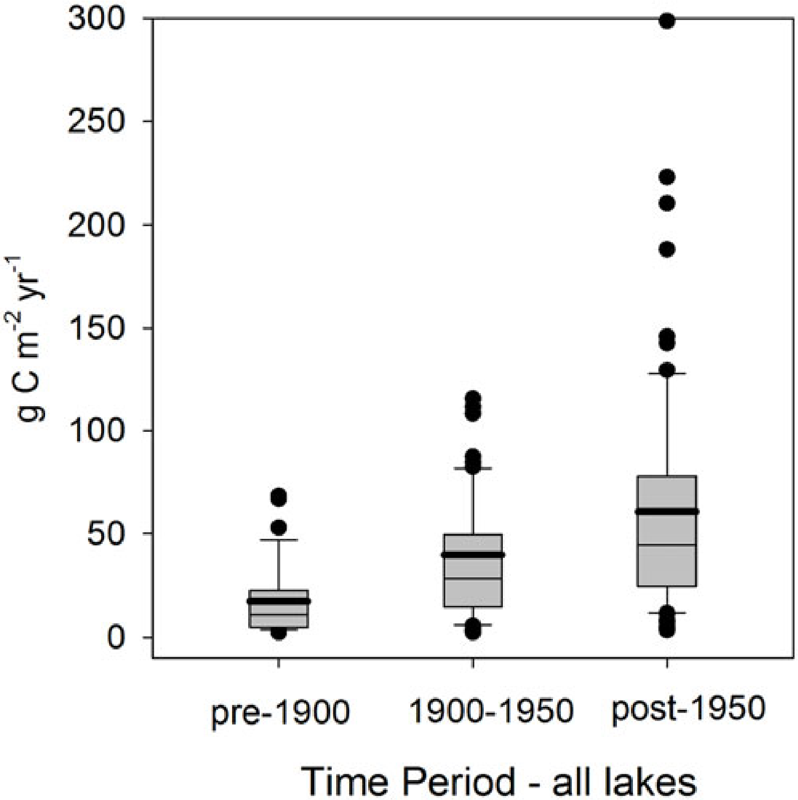
Fig. 5 C AR_{FC} for selected Danish lakes. Nb. variable scales are used for the C AR for individual lakes. For Ravn and Knud Sø contemporary monitoring data of epilimnetic TP (µg TP l^{-1}) are also shown to illustrate the reduction of in-lake TP associated with nutrient management strategies. Chronologies are derived from ^{210}Pb. Abbreviation as Figure 1.

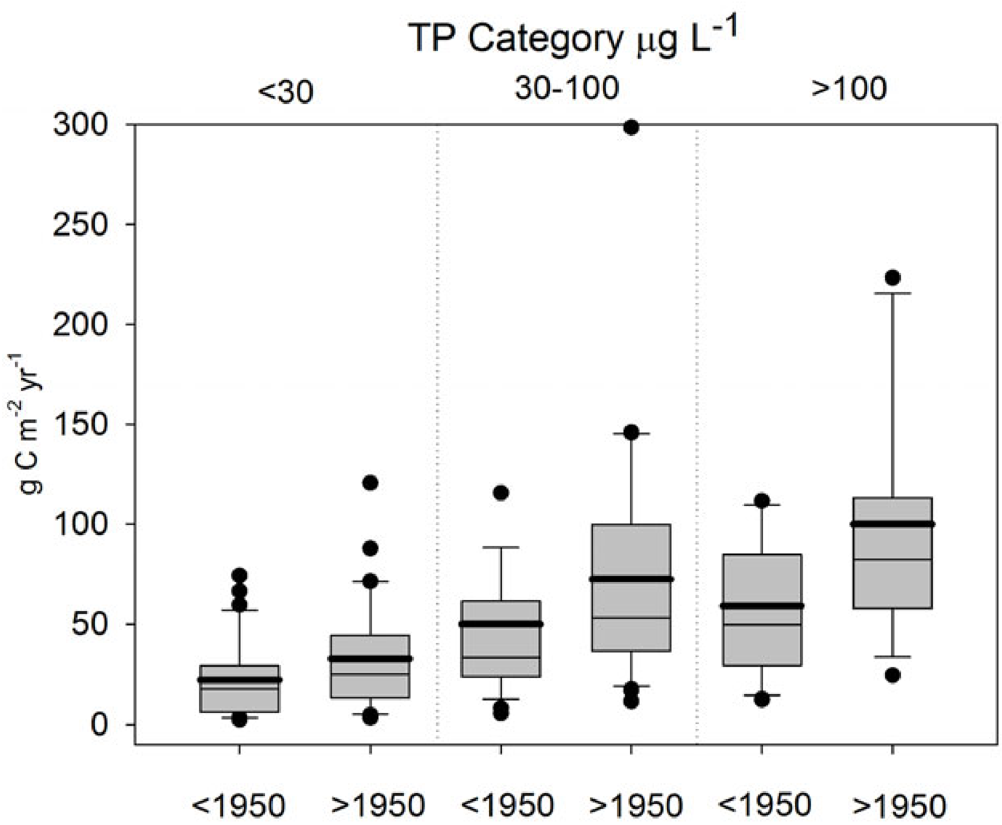
Fig. 6 C AR_{FC} for selected Scottish lochs. Nb. variable scales are used for the C AR for individual lakes. Chronologies are derived from ^{210}Pb. Abbreviation as Figure 1.

Fig. 7 A. C AR_{FC} for all lakes (n=55) in the eutrophic-hypertrophic category (i.e. > 30 µg TP l^{-1}) plotted as individual data points with a LOESS (locally weighted smoothing) smoother (0.2 span) fitted to identify the main trends in the data. The age of individual data points are derived from ^{210}Pb. B. The LOESS smoother from plot A (left) re-plotted (NB. the expanded scale for C AR) together with total European (EU-15) P and N fertilizer use ('000 tonnes) from ~1920 (source: www.fertilizer.org) to illustrate the increased C AR associated with increased fertilizer use after ~1950. The grey dash is only used as an indication of the trend in P fertilizer use from its first production in the 1850s. Regression analysis of the C AR trends derived from the LOESS smoother indicate a doubling of the average C burial increase rate from 0.4 to 1 g C m^{-2} yr^{-1} after 1950. Other abbreviations as Figure 1.

Table 1 Summary of the dataset, with lakes grouped according to OECD trophic classification for TP. The mean is given along with minimum and maximum values in parentheses.

TP class ug L^{-1}	N	countries	latitude	longitude	altitude m asl	lake area km^2	max depth m	current mean TP µg L^{-1}
< 10 Oligotrophic	11	Norway, Scotland, Switzerland, The Netherlands	47.017 to 60.765	-5.608 to 11.032	(-1) 74 (434)	(0.7) 58 (362)	(25) 126 (453)	(4) 5.6 (10)
11 to 30 Mesotrophic	27	Denmark, England, France, Northern Ireland/Republic of Ireland, Norway, Scotland, Slovenia, Switzerland, Wales	46.46 to 60.27	-8.08 to 14.1	(2) 203 (2339)	(0.02) 3.95 (35)	(1.6) 25 (110)	(11) 21 (30)
31 to 100 Eutrophic	36	Denmark, England, France, Northern Ireland/Republic of Ireland, Norway, Scotland, Slovenia, Switzerland	44.22 to 59.44	-8.25 to 13.83	(2) 225 (1768)	(0.02) 6.326 (109)	(-1.5) 15 (69)	(31) 60 (93)
> 100 Hypertrophic	19	Denmark, England, Northern Ireland, Scotland, Switzerland, The Netherlands, Wales	45.97 to 57.16	-3.93 to 12.22	(2) 100 (463)	(0.013) 2.04 (17)	(1.5) 14 (66)	(104) 286 (357)
Whole dataset	93	Denmark, England, France, Northern Ireland/Republic of Ireland, Norway, Scotland, Slovenia, Switzerland, The Netherlands, Wales	44.22 to 60.765	-8.25 to 14.1	(-1) 210 (2339)	(0.013) 15.65 (362)	(1.5) 37 (453)	(4) 80 (357)


300
250
200
150
100
50
0
g C m-2 yr-1
pre-1900
1900-1950
post-1950
Time Period - all lakes


TP Category μg L-1
<30
30-100
>100
300
250
200
150
100
50
0
g C m-2 yr-1
<1950
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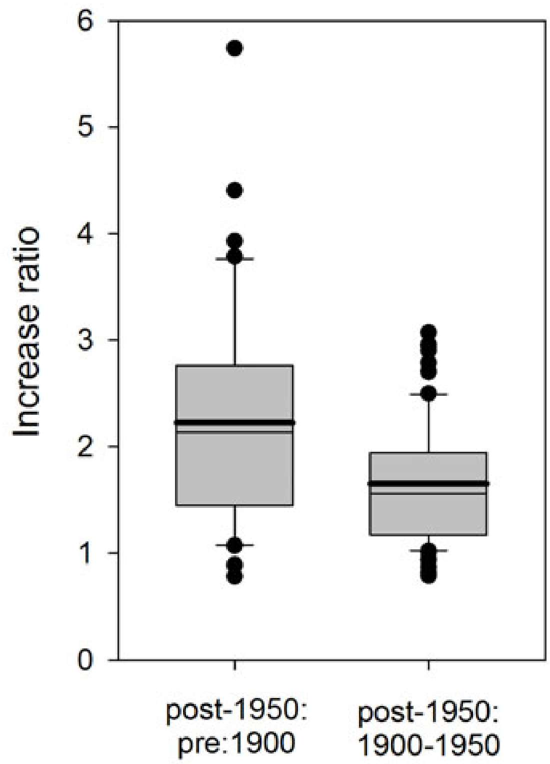
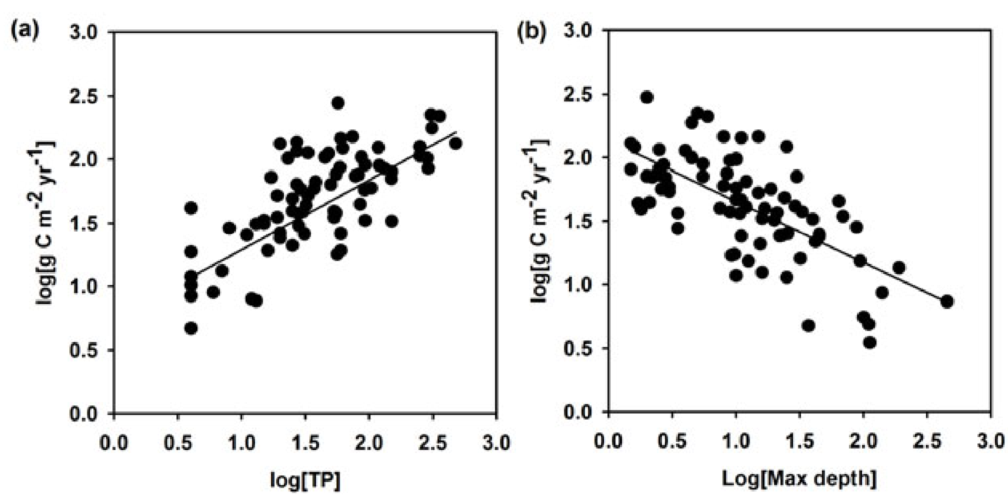
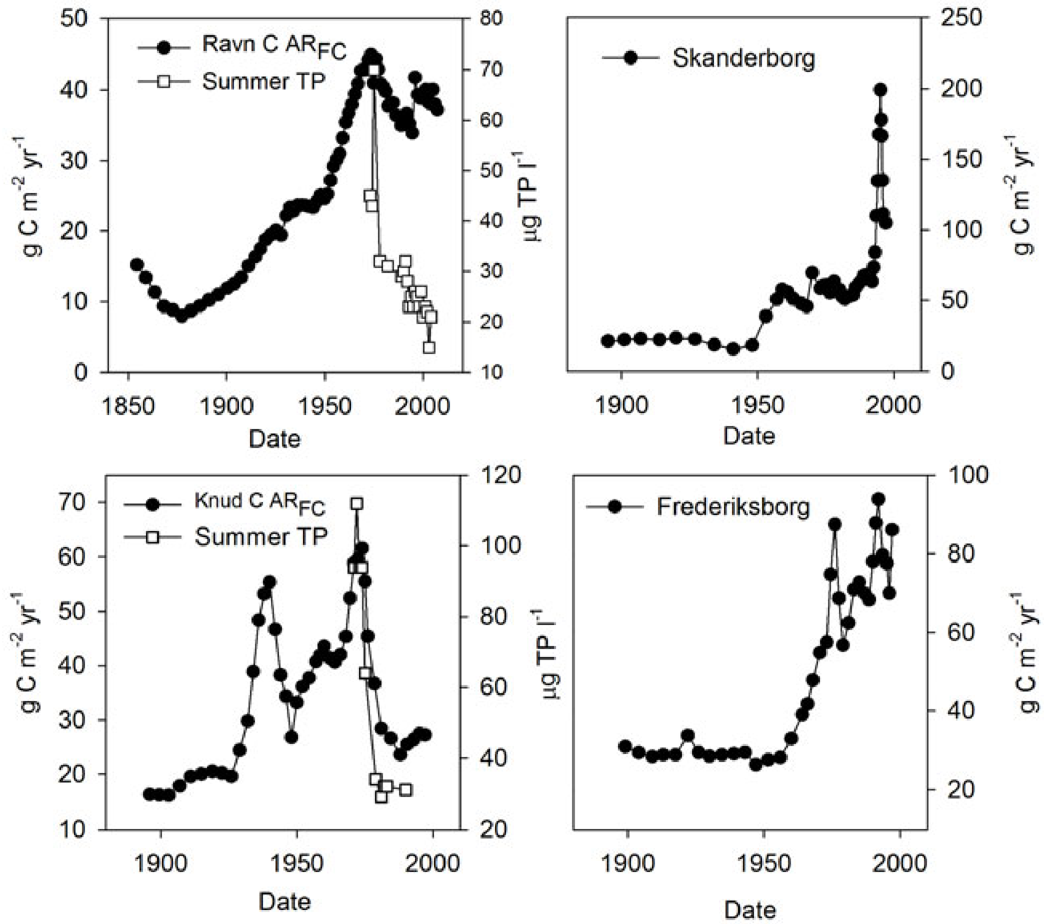
<1950
>1950
<1950
>1950



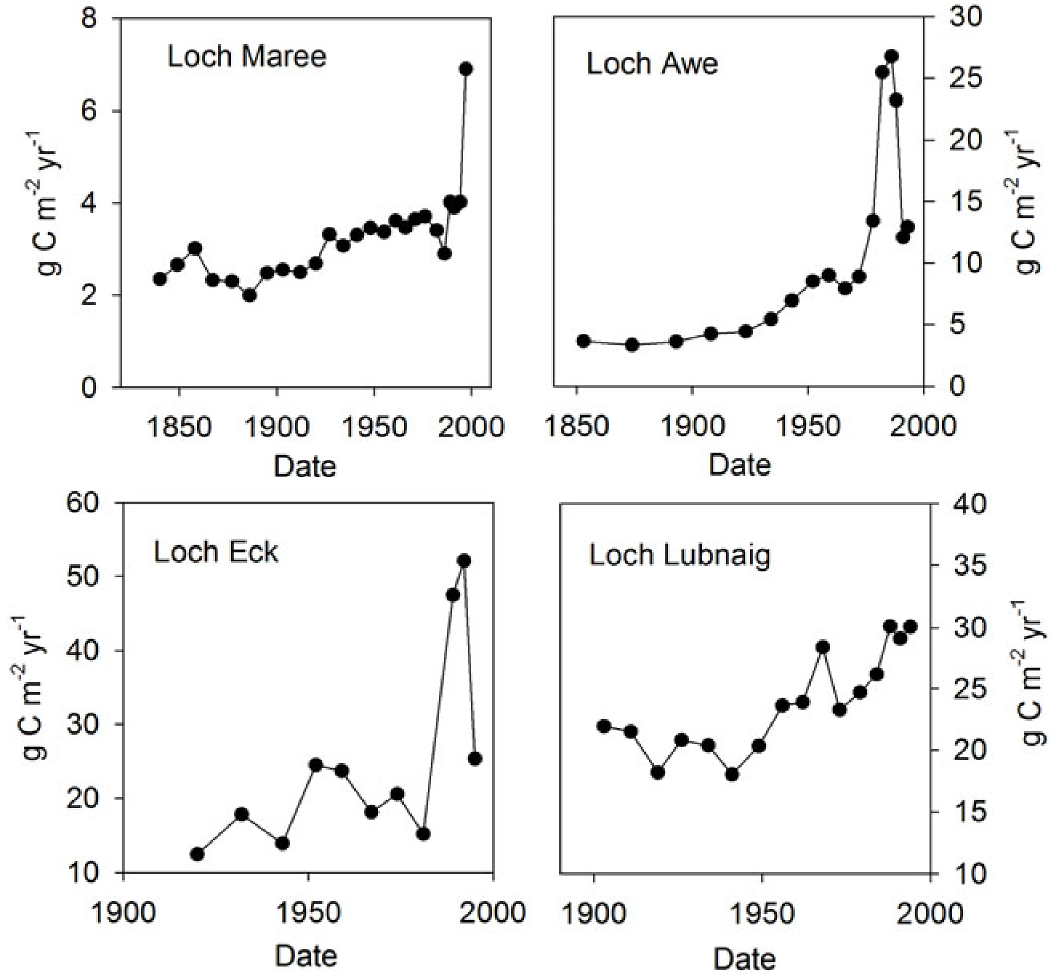
Increase ratio
0
1
2
3
4
5
6
post-1950:
pre:1900
post-1950:
1900-1950

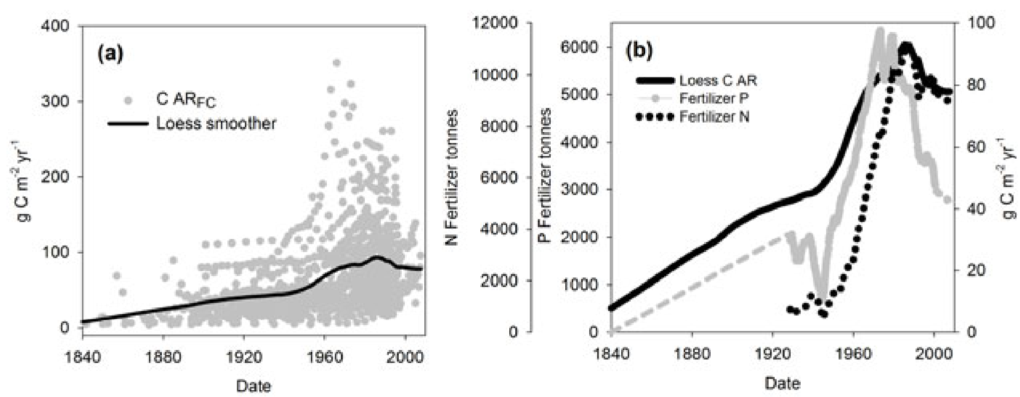


(a)
(b)
log[g C m-2 yr-1]
log[TP]
Log[Max depth]
0.0
0.5
1.0
1.5
2.0
2.5
3.0



Ravn C AR_{FC}
Summer TP
g C m^{-2} yr^{-1}
µg TP l^{-1}
Date
Skanderborg
g C m^{-2} yr^{-1}
Date
Knud C AR_{FC}
Summer TP
g C m^{-2} yr^{-1}
µg TP l^{-1}
Date
Frederiksborg
g C m^{-2} yr^{-1}
Date


Loch Maree
Loch Awe
Loch Eck
Loch Lubnaig
g C m-2 yr-1
Date


(a)
C ARFC
Loess smoother
g C m-2 yr-1
Date
(b)
Loess C AR
Fertilizer P
Fertilizer N
N Fertilizer tonnes
P Fertilizer tonnes