Anthropogenic modification of phosphorus sequestration in lake sediments during the Holocene: A global perspective

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Abstract

Human activity has fundamentally altered the global phosphorus (P) cycle. Yet our understanding of when and how humans influenced the P cycle has been limited by the scarcity of long-term P sequestration records, particularly outside Europe and North America. Lake sediments provide a unique archive of past P burial rates and allow the human-mediated disruption of the global P cycle to be examined. We compiled the first global-scale and continentally resolved reconstruction of lake-wide Holocene P burial rates using 108 lakes from around the world. In Europe, lake P burial rates started to increase noticeably after ~4000 calendar years before 1950 CE (cal BP), whereas the increase occurred later in China

(~2000 cal BP) and in North America (~550 cal BP), which is most likely related to different histories of population growth, land-use and associated soil erosion intensities. Anthropogenic soil erosion explains ~86% of the observed changes in global lake P burial rates in pre-industrial times. We also provide the first long-term estimates of the global lake P sink over the Holocene (~2686 Tg P). We estimate that the global mean lake sediment P sequestration since 1850 CE (100 cal BP) is ~1.54 Tg P yr^{-1}, representing approximately a six-fold increase above the mean pre-industrial value (~0.24 Tg P yr^{-1}; 11,500 to 100 cal BP) and around a ten-fold increase above the Early-Middle Holocene low-disturbance baseline of 0.16 Tg P yr^{-1}. This study suggests that human activities have been affecting the global P cycle for millennia, with substantial alteration after industrial times (1850 CE).

Key words: Phosphorus; Lake sediments; Holocene; Human impacts; Land use

1. Introduction

Phosphorus (P) is an essential element for life on Earth and is a critical nutrient for plant growth and food production (Cordell et al., 2009; Alewell et al., 2020). Recent human activities have substantially altered the global terrestrial P cycle (Ashley et al., 2011; Chen et al., 2016; Yuan et al., 2018), and a safe planetary boundary for P has been exceeded (Steffen et al., 2015), risking future food production (Alewell et al., 2020). Furthermore, the resulting excessive anthropogenic P loading to global freshwaters has exacerbated lake eutrophication, negatively affecting biodiversity and health of aquatic ecosystems (Smil, 2000; Jenny et al., 2019). Although it is not known when the Earth shifted from nature-dominated to human-dominated, inferring pre-disturbance P processes is critical to understanding the trajectory of the global P cycle over centennial to multi-millennial timescales.

Lake sediment cores are valuable archives for assessing temporal dynamics of terrestrial P cycling because of the strong linkage between catchment P loading and sediment P burial (Boyle, 2002; Moyle et al., 2021a; Moyle and Boyle, 2021). Therefore, records of P accumulation in sediments provide estimates of the timing, magnitude, and extent of anthropogenic disturbance to catchment-scale P cycles.

Characterizing the temporal variability of lake P burial rates across broad geographic areas is critical for understanding the global P cycle (Anderson et al., 2020). Although anthropogenic drivers of changes in sediment P records have been investigated intensively, most studies have focused on limited spatial scales (individual lakes) or relatively short time periods (typically the last one or two centuries) (Wang et al., 2021; Bhattacharya et al., 2022). Furthermore, most relevant publications report only sediment P concentrations instead of basin-wide P burial rates. Sediment P accumulation rates from single cores are likely to be biased by sediment focusing in the lake (Scholtysik et al., 2022) and, thus, may not represent the pattern of lake-wide mean sediment P sequestration. Previous studies of long-term P burial largely focused on North America and Europe (Moyle et al., 2021a), which highlights gaps in a global coverage of P records. This is particularly true for China, which has a long history of agricultural P use (Ashley et al., 2011). Several studies have shown that early anthropogenic deforestation and farming in Europe started to have detectable impacts on lake P burial rates and P cycle millennia ago (Boyle et al., 2015; Klamt et al., 2021; Moyle et al., 2021a), and similar patterns are likely for lake sediments in China.

To date, a truly global synthesis of lake P records on centennial to millennial timescales is missing, and there is also a lack of a well-constrained and long-term global estimate of total lake P sequestration. In this study, we substantially extend an existing meta-analysis of Holocene lake-sediment P burial (Moyle et al., 2021a), presenting records from 108 lakes on six continents (Fig. 1). This dataset represents the largest and most widely distributed coverage of lake-wide P burial rates so far. We explore patterns of lake P burial rates during the Holocene at global and continental scales and relate these to early and recent anthropogenic disturbances (e.g., land use, soil erosion, P fertilizer inputs, and sewage disposal).

Furthermore, for the first time we produce a global estimate of lake P sequestration throughout the Holocene and propose incorporating this long-term record into Earth System Models (ESMs).

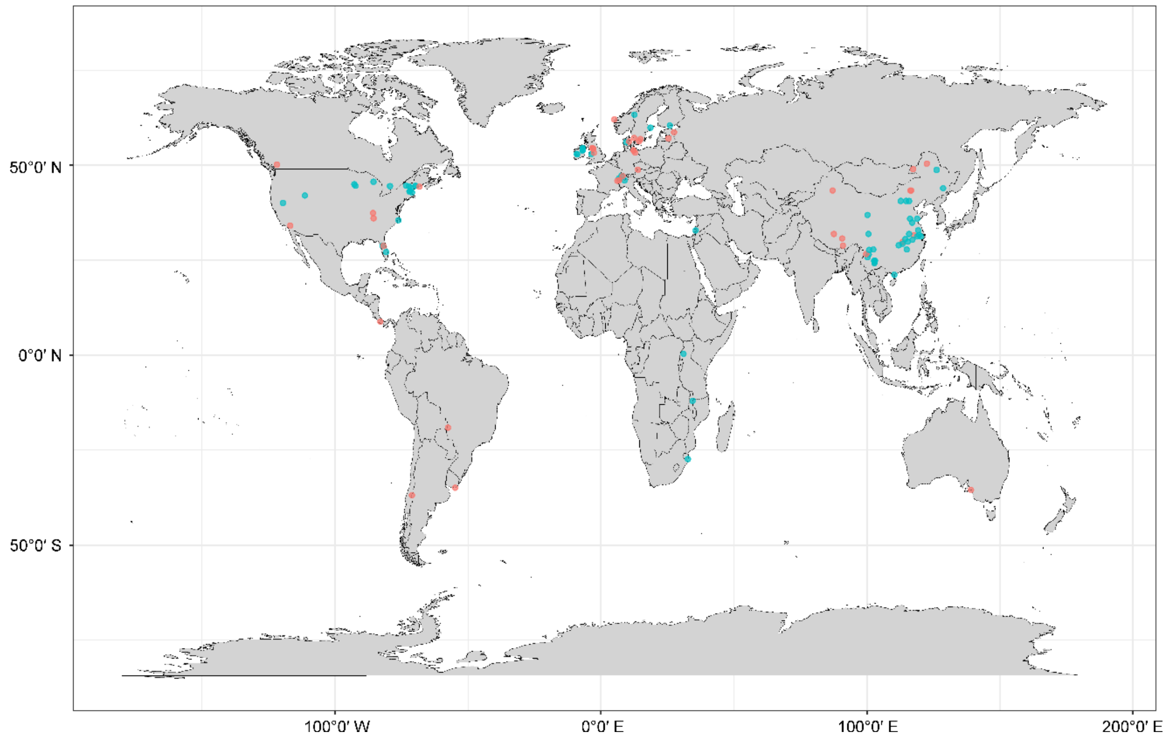

Fig. 1. Global distribution of sediment P records from 108 lakes in the study. The pink color indicates lakes with long-term P records (> 200 years) and the blue color with more recent P records (since 1850 CE).

2. Materials and methods

2.1. Data synthesis

Our database consists of sediment-P data from 108 lakes in North America, Europe, China, West Asia, Africa, South America, and Australia and were derived from published sources (Fig. 1; Supplementary Table S1). The five critical criteria during the site-selection process were the availability of (i) chronologies (AMS ^{14}C dates for records longer than 200 years and ^{210}Pb and /or ^{137}Cs data for records less than 200 years), (ii) P concentrations or P burial rates in sediment cores, (iii) location of the lake, (iv) mean water depth of the lake, and (v) water depth of the coring site. The largest number of lakes in this study is from China (41), followed by Europe (34), North America (25), South Africa (3), South America (3), West Asia (1), and Australia (1). Recalibration of existing ^{14}C ages was performed using OxCal 4.0 (Ramsey, 2009). Calibration of ^{14}C ages in lakes of the Northern

Hemisphere was performed using the IntCal20 calibration curve (cal BP, 2σ probability, Reimer et al., 2020). The Southern Hemisphere calibration curve SHCal04 (McCormac et al., 2004) was used for ^{14}C ages of lakes in the Southern Hemisphere. Short P records (covering the past 100–200 years) were from North America, Europe, China, and Africa, and long P records (covering time periods longer than the past 200 years) were from North America, Europe, China, South America, and Australia. Although the total dataset does not include all studies of lake sediment P undertaken to date, it represents the largest and the longest temporal synthesis of lake P burial rates to date.

2.2. Sediment-inferred mean lake-wide P burial rates

A simple process model (Moyle and Boyle, 2021) was applied to published records of sediment-core P concentrations or sediment-core P burial rates to calculate sediment-inferred mean lake-wide P burial rates (Lsed; m^{-2} yr^{-1}) for the Holocene (i.e., the last 11,500 calendar years before present, where present is defined as 1950 CE). Lsed considers sediment focusing effects within a lake basin, and for this study we assume that sediment focusing intensity remains constant through a lake's history. As Lsed represents the lake-wide mean P burial rate, it enables comparison of P records between different sites (Moyle and Boyle, 2021; Moyle et al., 2021a). For the purpose of this study, the P retention coefficient, defined as the proportion of P supplied that is retained in the lake sediments, was not considered mainly because these values are not generally available for most of the lakes. Also, according to Moyle et al. (2021a), temporal changes in lake P retention coefficients during the Holocene are relatively small compared with between-site differences. Therefore, P net sequestration rates in most of our sediment records can still provide reliable information about long-term variation of P loading from the catchment to lakes.

In our dataset, Holocene records of Lsed for 24 lakes in North America and Europe (Table S1) were collected from the database of Moyle et al. (2021b). For other lakes, Lsed was calculated using the equations in Moyle and Boyle (2021): Lsed $= L_{core} \times Z_{mean} / Z_{core}$; where L_{core} = P burial rate of the sediment core (mg m^{-2} yr^{-1});

L_{core} is calculated based on sediment P concentrations (Pcon, mg g^{-1}) and core mass accumulation rates (MAR, g m^{-2} yr^{-1}) given by: L_{core} = Pcon × MAR; Z_{mean} = mean water depth of the lake (m); Z_{core} = water depth of the coring site (m). If MAR was not reported in the original publications, it was calculated based on sediment accumulation rates (SAR, cm yr^{-1}) from the age-depth model and the dry bulk density (DBD, g cm^{-3}) of the sediment core, as given by: MAR = SAR × DBD ×10^4. In studies for which DBD was not available, it can be estimated reliably by the following methods:

1) Water contents (W, g/g) of sediments were preferentially used. In cases for which W was reported, DBD is given by Moyle and Boyle (2021):

$$DBD = (1-W) / (W/d_w + (1-W)/d)$$

Where d_w = Water Density, taken as 1.0 g cm^{-3}, and d = Minerogenic Sediment Density, taken as 2.7 g cm^{-3}.

2) If water contents were not reported but loss on ignition (LOI, wt.%; at 550°C) for the core sediment was reported, DBD is given by Moyle and Boyle (2021):

$$DBD = 2.13 \times LOI^{-0.682}$$

3) If neither water contents nor LOI were reported but total organic carbon (TOC, wt.%) data were available, DBD is estimated based on the following formula (Avnimelech et al., 2001):

$$DBD = 1.665 \times (TOC)^{-0.887} \ (TOC > 6\%)$$

$$DBD = 1.776 - 0.363 \times \ln(10 \times TOC) \ (TOC \leq 6\%).$$

For the site-specific geochemical parameters used to apply the model, refer to Table S1.

2.3. Estimation of global lake P burial rates using biomes

Following the approach of Anderson et al. (2020), we used global biomes as a basis for identifying coherent landscape types and to scale up Lsed from individual lake records to the global estimate of Holocene P sequestration. Firstly, the 108 lakes were assigned to global biomes based on their locations within The Nature

Conservancy's Terrestrial Ecoregion polygons, following Olson et al. (2001). Then, the median of Lsed in each biome for three time-intervals (Early and Middle Holocene 11,500-4000 cal BP, Late Holocene before pre-industrial times 4000-100 cal BP, and modern times since 100 cal BP) was multiplied by lake areas from the biome, following Anderson et al. (2020) in which the known area of the world's 22 largest lakes and the global reservoir lake areas are not considered. We calculated global average Lsed as the sum of Lsed from all biomes. Although the distribution of the 108 lakes across the different biomes is not homogenous, the dataset over the last 11,500 years covers the majority (11 of 15) of the global biomes (Fig. S1).

2.4. Statistical analysis

The statistical analysis was performed using R software (R CoreTeam, 2021). To get matching ages to sediment P data, other records (e.g., SAR, MAR, DBD, LOI, TOC, and W data) were interpolated in R using approxfun (method = "linear") in "stats" package. The Lsed data were $\log_{10}$ transformed prior to smoothed-trend analyses to better represent the overall change patterns. Then, the trends of $\log_{10}$ (Lsed) were determined by fitting a simple generalized additive model (GAM) via gam() function from the "mgcv" package (Wood, 2017) with REML smoothness selection (using method = "REML") and the number of basis functions (k), following the approach in Simpson (2018). We tested the number of basis functions (k) required for the GAM models to sufficiently estimate the fitted trends.

Mean and median values of Lsed were calculated for each of the 100-year bins to reflect the variation of Lsed at centennial scales during the Holocene. In a similar way, mean and median values of Lsed were calculated for each of the 20-year bins from 1850 CE to 2010 CE and the 10-year bins from 2010 CE to 2020 CE. The breakpoints were detected on the mean curves of the 100-year bins for global and continental data using the R package "BreakPoints" (Hurtado et al., 2020).

A GAM was applied to estimate the long-term impact of climate change (using records of global temperature anomalies (MarcFott et al., 2013) and atmospheric CO_2) and anthropogenic land-use disturbance (using records of global soil erosion as indicated by SAR (Waters et al., 2016) and global cropland areas (Mottl et al., 2021)) on the global Lsed values. The R package “mgcv” (Wood, 2017) was used for calculating the GAM. The dataset for the GAM was calculated from the binned mean values of every 100-year interval during the Holocene. We found a heavy leverage of the data point at 50 cal BP (0-100 cal BP range) with the highest P value and cropland area. Therefore, the time window between 11,450 and 100 cal BP was selected for the modelling. The GAM formula to assess controls on Lsed is a function of changes in SAR, global cropland areas (Cropland), global temperature anomalies (Temp), and atmospheric CO_2 during the Holocene: Lsed ~ s(SAR) + s(Cropland) + s(Temp) + s(CO_2). P-value results are informative about the contribution of SAR, Cropland, temperature, and atmospheric CO_2 to Lsed.

3. Results and discussion

3.1. Changes in lake P burial rates during the Holocene

Despite differences among continents, global and continental trajectories during the Holocene show a similar pattern: the Lsed records all show unprecedented high values after ~100 cal BP (after ~1850 CE; Fig. 2a; Fig. 3). This global-scale pattern is consistent with the mean breakpoint year at 1900 CE (range 1850-1950 CE) in the global Lsed synthesis (Supplementary Fig. S2), which coincides with recent definitions of the Anthropocene (Waters et al., 2016). In the last 100 to 200 years, intensive farming practices and industrialization have caused notable land-use changes, which are responsible for intensified global soil erosion, P leaching from agricultural soils, and P accumulation in lakes (Ruttenberg, 2003; Klein and Ramankutty, 2004; Kemp et al., 2020; Zhang et al., 2022). The sharp increases in Lsed values globally and continentally in the past 100–200 years (Fig. 2a; Fig.3) undoubtedly were related to anthropogenic P transport from terrestrial to aquatic systems (Yuan et al., 2018) via fertilizer use, P loss from agricultural systems via soil erosion, and P discharge from sewage effluents . We estimate that the mean global lake P burial rate since 1850 CE (100 cal BP) was ~1.54 Tg P yr^{-1} (Table S2), representing a ~six-fold increase above the mean pre-industrial value (~0.24 Tg P yr^{-1}; 11,500 to 100 cal BP). As such, our results suggest that because of unprecedented anthropogenic activities around the globe during the past 100-200 years, a fundamental shift in the global P cycle occurred, consistent with previous studies (Yuan et al., 2018; Alewell et al., 2020).

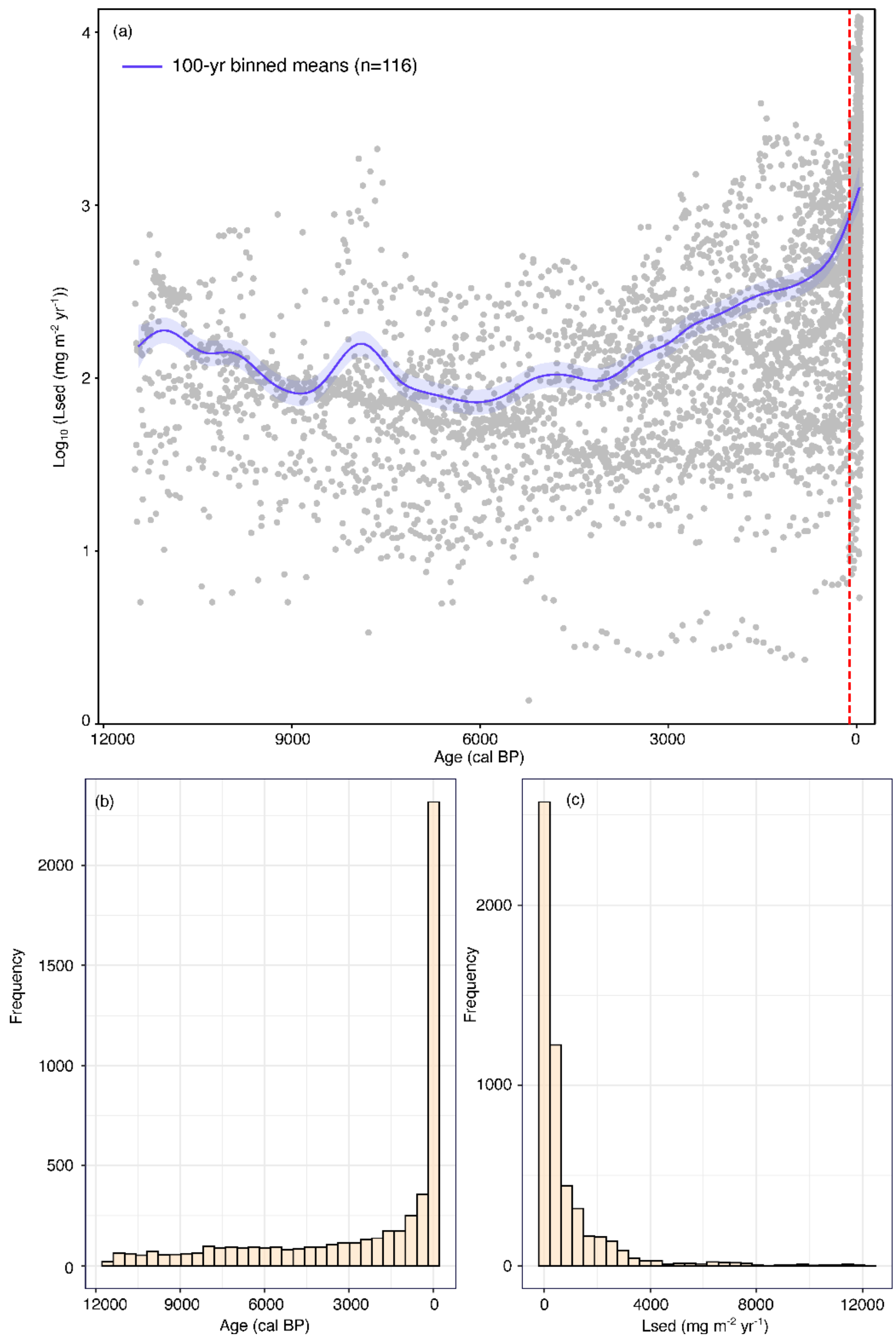

Fig. 2. (a) Generalized additive model (GAM) based trends fitted to the means of P data in 100-yr bins (blue curve, basis dimension k=30, samples n=116) in 108 lakes during the

Holocene, with 95% confidence intervals on the predicted means (blue shaded envelopes); the blue fit is the result of a GAM with restricted maximum likelihood (REML) smoothness selection; the data of Lsed (shown as grey points) were $\log_{10}$ transformed. The vertical dashed red line represents the breakpoint at 50 cal BP (i.e., mean breakpoint year, minimum-maximum range 0-100 cal BP). (b) Histogram (bins = 30) showing the frequency of 5365 calibrated ^{14}C ages. (c) Histogram (bins = 30) showing the frequency of values of Lsed.

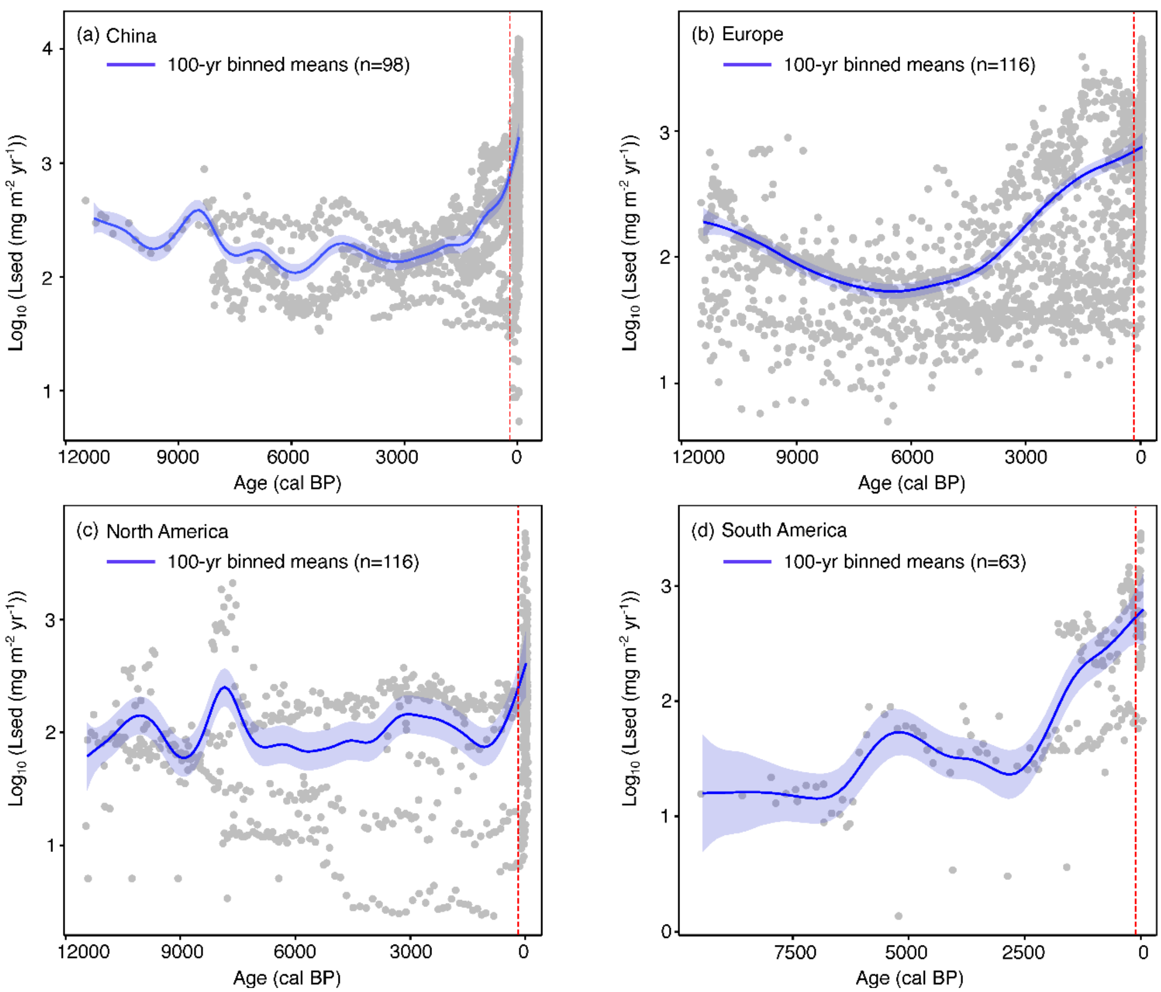

Fig. 3. Generalized additive model (GAM) based trends fitted to the means of P data in 100-yr bins (blue curves) from lakes in (a) China, (b) Europe, (c) North America, and (d) South America during the Holocene, with 95% confidence intervals on the predicted means (blue shaded envelopes); the blue fits are the results of a GAM with restricted maximum likelihood (REML) smoothness selection (basis dimension k=30). The vertical

dashed red line represents the breakpoint at 50 cal BP (i.e., mean breakpoint year, minimum-maximum range 0-100 cal BP) for each continent; the data of Lsed (shown as grey points) were $\log_{10}$ transformed.

In the Early Holocene, Lsed records for the global average, China, Europe, and North America all show relatively high values followed by decreasing trends towards the Middle Holocene (Fig. 2a; Fig. 3a-c). This phenomenon can be mostly explained by elevated catchment P supply from base-rich rejuvenated soils after deglaciation in the Early Holocene, which was mainly attributed to natural soil evolution driven by climate amelioration rather than human activities. Boyle et al. (2013), Boyle et al. (2015) and Moyle et al. (2021a) also found high soil P export associated with recently deglaciated landscapes. For P records of China, the declining trend from the Early Holocene (11,500-8000 cal BP) to the Middle and Late Holocene (Fig. 3a) mirrors the changes in soil erosion intensity in China (Fig. 4a) that were controlled predominantly by the Asian summer monsoon (Zhang et al., 2022). In South America, limited data points in the Early and Middle Holocene (~9500-5000 cal BP; n=25; Fig. 3d) result in large uncertainties and thus the pattern is not discussed.

Although lake P burial rates among different continents all show maximal values in the modern period compared to the last 11,500 years, there are notable differences in the pre-industrial (before ~1850 CE) temporal evolution of P records (Fig. 3). The most obvious difference is in the timing of the first visible increase in P records. For instance, the P curve for European lakes began to increase substantially after the onset of the Late Holocene around 4500-4000 years ago (beginning of the Bronze Age in Europe; Fig. 3b). However, the first increases were later in China (~2000 cal BP) and North America (~550 cal BP) (Fig. 3). In landscapes that experienced early deforestation and farming, anthropogenic landscape disturbance enhanced catchment P yield and sediment P sequestration in lakes during the Middle and Late Holocene (Boyle et al., 2015; Klamt et al., 2021). This is because P loss through catchment soil erosion was the dominant contributor to lake P inputs (Carpenter et al., 1998; Yuan et al., 2018; Alewell et al., 2020). During the Middle and Late Holocene, Lsed records for the global average, Europe, and North America showed patterns consistent with their respective anthropogenic soil erosion time-series (Fig. 4a). This indicates that the different patterns of enhanced Lsed in the Late Holocene among these continents are most likely attributed to differing soil erosion intensities associated with distinct continental land use histories.

In Europe, the first increases in Lsed (~4500-4000 cal BP; Fig. 4a) are coincident with accelerated soil erosion caused by deforestation and intensive land use (Boyle et al., 2015; Moyle et al., 2021a). In North America, the notable rise in sediment P burial rates occurred later than in Europe (after ~550 cal BP, Fig. 3c). This record shows co-variations with enhanced rates of soil erosion in North America (Fig. 4a) which was very likely attributable to European colonization (Jenny et al., 2019; Kemp et al., 2020). Although elevated sediment P burial rates occurred in some lakes of North America during the Early Holocene (~11,000-7500 cal BP), data for this period are sparse (n=133, Fig. 3c).

In China, the first noticeable increase in P burial rates (~2000 cal BP) was later than the early record of soil erosion (Fig. 4a; Zhang et al., 2022). This can be explained by the fact that most of the long-term lake P records from China selected for this study are distributed in the middle and lower reaches of the Yangtze River Basin (east-central China) and in southwest China (Fig. 1). During the Bronze Age and early Iron Age (~4000-2000 cal BP), however, the economic center and the most populated areas of China were in northern and central China (Ashley et al., 2011; Li et al., 2021). It is therefore reasonable to infer that before ~2000 cal BP, human disturbances in lake catchments of east-central and southwest China remained relatively low (Hosner et al., 2016). Consequently, P loading from croplands, through manure application and soil erosion, was probably limited. P burial rates in Chinese lakes increased strongly after ~2000 cal BP, particularly after ~1500 cal BP (Fig. 3a and 4), broadly coincident with increased rice agriculture (mainly in southern and southwestern China; Klein Goldewijk and Ramankutty, 2004) and more rapid increases in soil erosion (Zhang et al., 2022), population growth (Li et al., 2009), and expansion of cropland areas in ancient China (Fig. 4b; Klein Goldewijk et al., 2017). This finding indicates increasing anthropogenic impacts on lake P burial after 2000-1500 cal BP, which is supported by archaeological evidence documenting a large population migration from northern China to central-eastern China after the Sui Dynasty (~1500-1400 cal BP; Li et al., 2021). China has a long history of using P-rich animal manure and human excreta as fertilizer (dating back to ~4000-5000 years ago; Ashley et al., 2011). Therefore, it is probable that anthropogenic land-use and soil erosion intensified across the country around 4000 years ago (Fig. 4a; Zhang et al., 2022). Although the site coverage does not represent the whole continent (Fig. 1a), initial anthropogenic impacts on the P cycle in China may also have occurred early in the Late Holocene, ~4000 cal BP in regions with dense Bronze Age and Iron Age populations. As predicted, this mirrors the pattern of early P burial increases seen across Europe. However, further investigations, with more lake-sediment P records over multi-millennial intervals across China, are needed to better support this argument.

The Holocene record of soil erosion intensity in South America is not available, yet the modelled P burial rates increased continuously over time during the Holocene (Fig. 3d). Nevertheless, relatively high Lsed values during pre-industrial times coincide with early-anthropogenic land use during agricultural expansion (5000 BP to 1500 CE) and the colonial period (1500 to 1800s CE) in South America (Armesto et al., 2010). This phenomenon implies detectable early-anthropogenic impacts on terrestrial P cycling in South America.

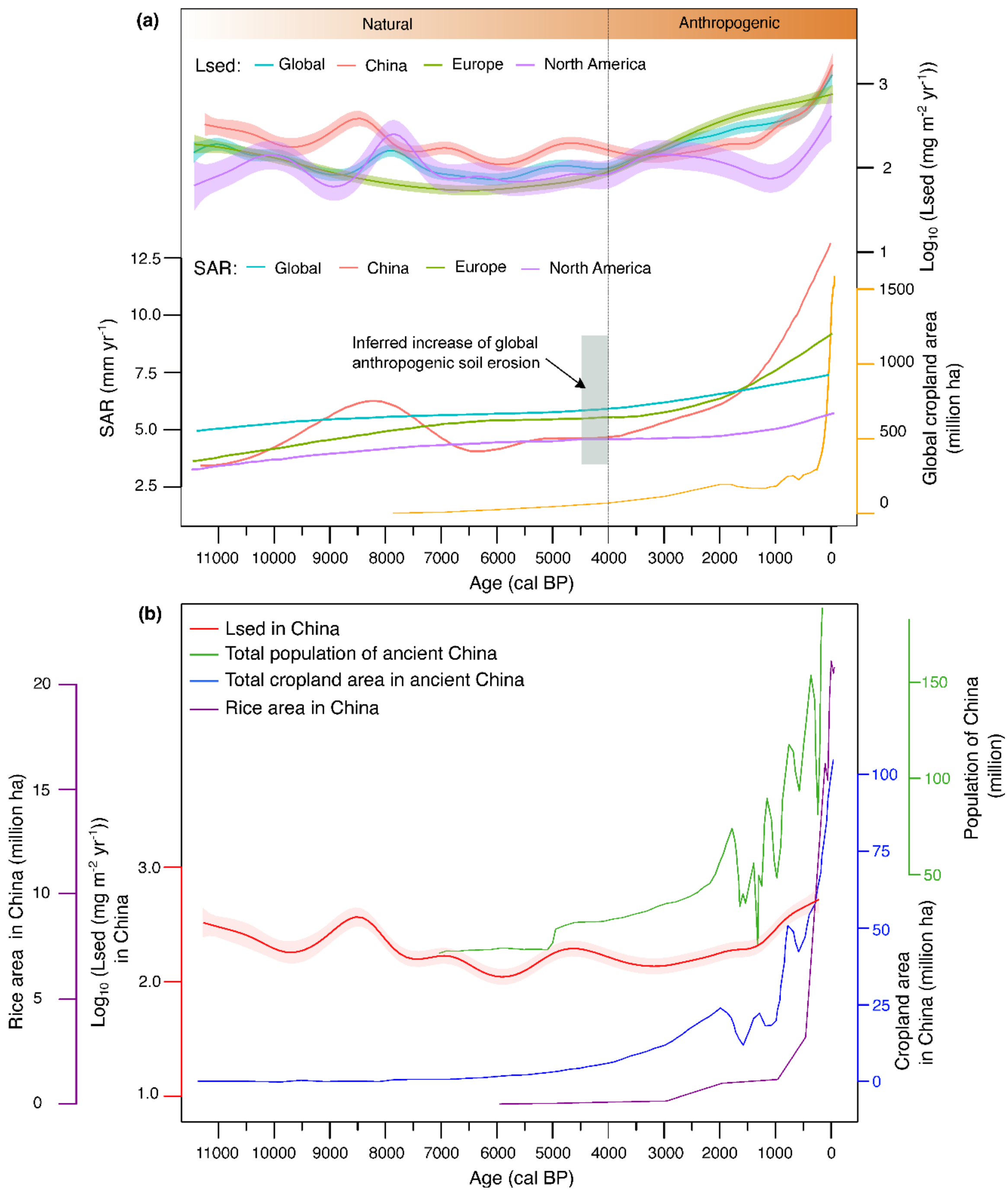

Fig. 4. (a) Comparison of Lsed (GAM-fitted trends of 100-yr binned means), soil erosion intensity (reconstructed from lake sediment accumulation rates, SAR) at global (for 632 sites, Jenny et al., 2019, SAR data n=3980) and continental levels (Jenny et al., 2019; Zhang et al., 2022), and global cropland area during the Holocene (Klein Goldewijk et al., 2017); the global synthesis of Lsed values(this study) is represented by 108 lakes; the rectangle and vertical grey line indicate the timing of early increases in anthropogenic soil

erosion globally (Jenny et al., 2019). (b) Comparisons of the GAM-fitted trend of Lsed values in Chinese lakes between 11,500 cal BP and 150 cal BP with estimated population (Li et al., 2009), total cropland area (Klein Goldewijk et al., 2017), and rice area estimates (Li et al., 2009). Note that in (a) and (b) the dataset of Lsed is log_{10}-transformed and the fits of Lsed are the result of GAM with REML-based smoothness, with 95% confidence intervals on predicted means (shaded envelopes).

3.2. Early anthropogenic impacts on global lake P burial rates since the Late Holocene

The global pattern of Lsed values (Fig. 2a) is biased towards Europe and China (Fig. 3a, b) because these areas represent a large portion of sites in our compilation (69%; Fig. 1). Nonetheless, the general trend for increasing global Lsed values since ~4500-4000 cal BP (Fig. 4a) tracks early increases in global soil erosion intensity (proxy of SAR; Fig. 4a; Jenny et al., 2019), global cropland area (Fig. 4a; Li et al., 2009), and deforestation in lake watersheds at global levels (Mottl et al., 2021). A GAM fit to global soil erosion intensity (Jenny et al., 2019) explains ~86 % of the variance in global average Lsed values from the Early Holocene to 1850 CE ($R^2_{adj} = 0.85$). The predictor of soil erosion intensity is significant (p-value < 2e–16), whereas the contributions of climate variables (air temperature and atmospheric CO_2) and global cropland areas to Lsed values are not significant (p-values are 0.73, 0.55, and 0.18, respectively). Therefore, statistical analyses support the inference that anthropogenic landscape disturbance and associated intensified soil erosion was the leading driver for intensified global lake P burial rates in pre-industrial periods.

The mean global Lsed between ~4000 and 100 cal BP was significantly higher than (almost double) that before ~4000 cal BP (Fig. 5), which indicates that global P input to freshwater since the beginning of the Late Holocene increased over natural background levels. Whereas anthropogenic disruption of the global P cycle since the 20th century is well documented (Liu et al., 2016; Yuan et al., 2018; Scholz and Wellmer, 2019), only a handful of studies of European lakes have recognized early anthropogenic impacts (Boyle et al., 2015; Klamt et al., 2021; Moyle et al., 2021a). Our results reveal that human activities began to interfere with the global terrestrial P cycle from the Middle-Late Holocene, suggesting this pattern is spatially much wider than previously recognized. The profound early anthropogenic impact on the P cycle is particularly observed in Europe and China, where there are long histories of agricultural land-use and landscape disturbance (Klein Goldewijk et al., 2017). Our findings support those of Moyle et al. (2021a) who revealed substantial impacts of human activities on terrestrial P cycling from ~6000 cal BP in lowland Europe.

3.3. Global and continent-scale lake P burial rates in recent times

Although continental increases in lake P loading started millennia ago, the most substantial increase in lake P loading occurred in the last 100 to 200 years (Fig. 2a; Fig. S2), coinciding with intensive farming practices and industrialization. Over this period global P burial has increased consistently through time (Fig. 6). The substantial increase in anthropogenic P loading over the past century has enhanced P concentrations in both global lake waters and lake sediments (Sharpley et al., 2013; Moyle et al., 2021a). This study shows that lake P burial records on a global scale match well with the historical use of mineral P fertilizers from the late 19th century onward (Fig. S3a, b), and confirms the effects of anthropogenic P use on lake P burial rates on a decadal timescale.

Consistent with the global average, lake sediment P accumulation records from all continents show systematic increases from 1850 CE to 1950 CE (Fig. 6). However, since 1950 CE, China and Europe show an accelerating trend, whereas the records of North America and South America have declined, and Africa has increased steadily. These distinct patterns of P sequestration across the continents are likely to be a consequence of different patterns of variable landscape history (Moyle et al. 2021a) and/or lake morphological and hydrological factors, though sampling bias i.e., small sample sizes (n= 68 for Africa and n=34 for South America) may also explain the pattern. China has the highest mean lake P burial rate (~2000 mg P m^{-2} $year^{-1}$; Fig. 6), which may reflect high P fertilizer use and runoff. In China, the steady increase in Lsed values since the 1950s mirrors the intensified P fertilizer input and the calculated P loss into freshwaters (Fig. S3a, c). This result is consistent with previous studies that showed intense P fertilizer input constituted the highest contribution to sediment P enrichments in China over the past century (Li et al., 2015; Lu and Tian, 2016).

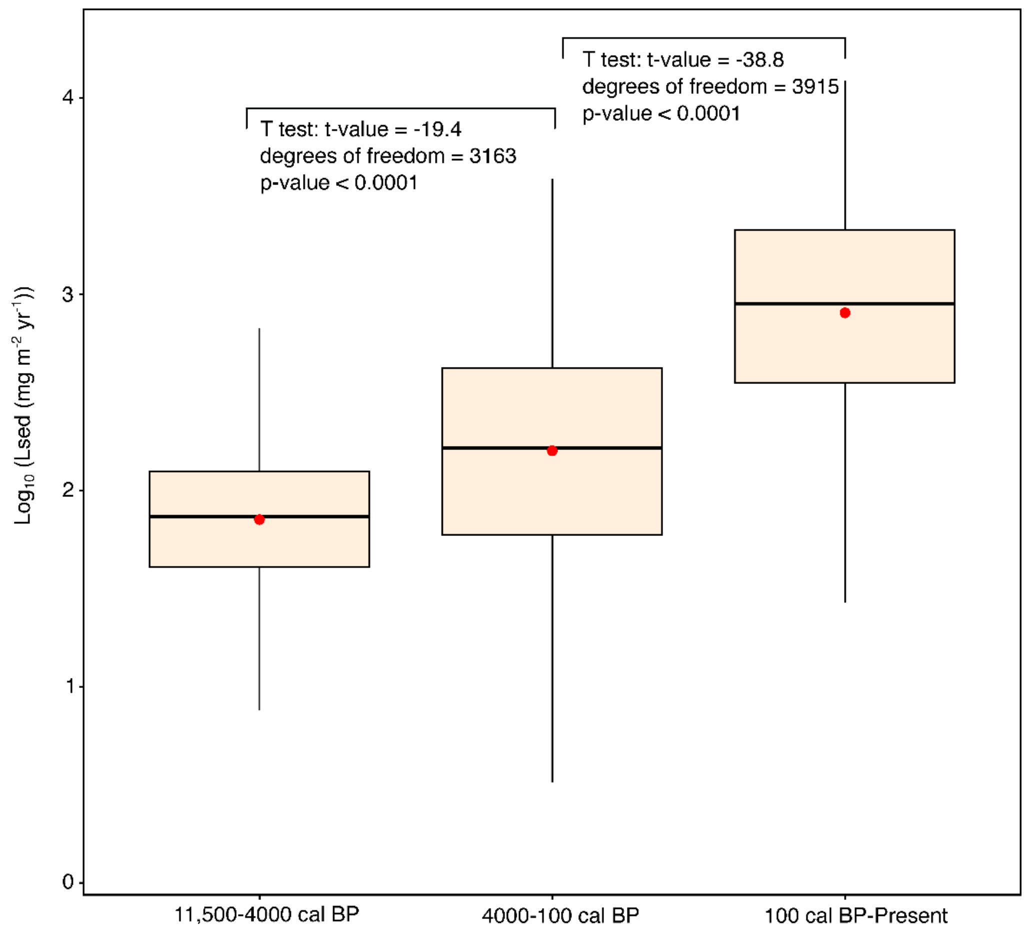

Fig. 5. Boxplots showing the global synthesis of Lsed from three time-intervals: Early and Middle Holocene (11,500 to 4000 cal BP), Late Holocene (4000 to 100 cal BP), and recent times (from 100 cal BP to present). The red dot and the black horizontal line in each boxplot indicate mean and median value of the interval, respectively. The dataset of Lsed was $\log_{10}$-transformed.

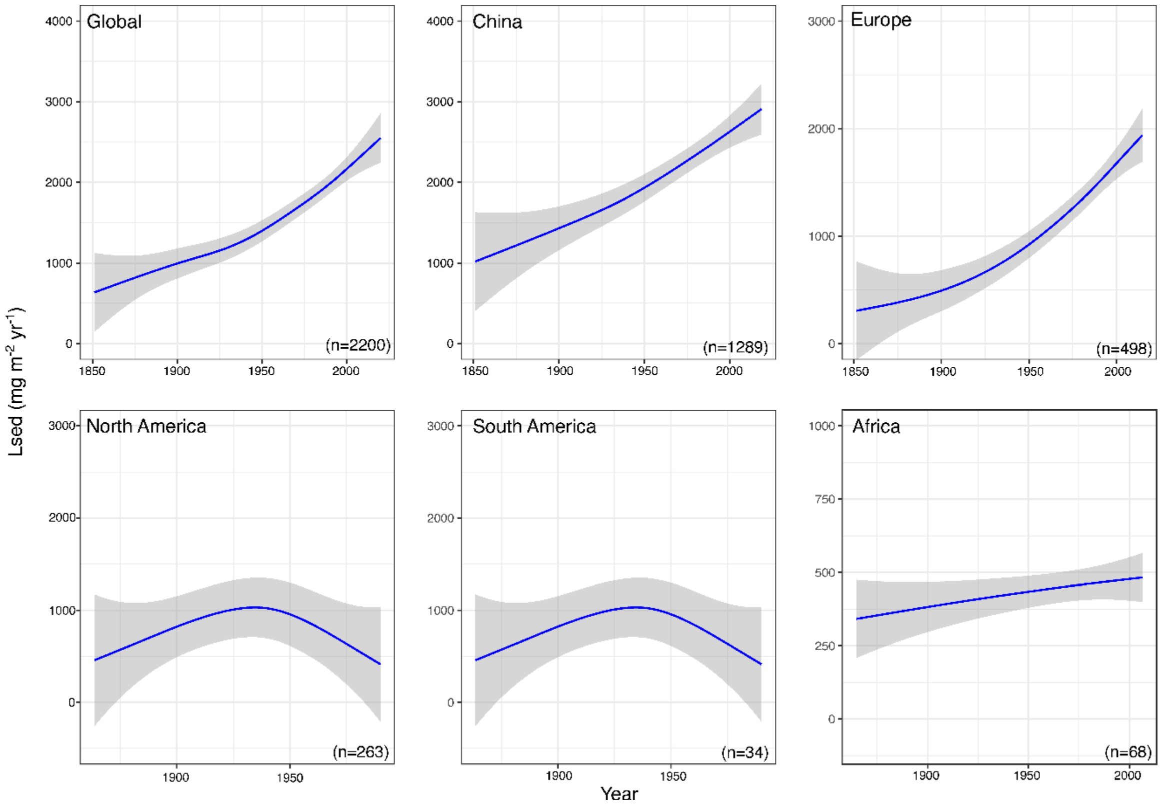

Fig. 6. Comparative Lsed records for selected continents since 1850 CE as GAM curves with REML smoothness selection (k=10) and shaded 95% confidence intervals.

3.4. Estimates of total global P sequestration rates by lake sediments

Our dataset enables estimates of modern and Holocene global P sequestration rates in lake sediments (Table S2), which provides insights into previously poorly constrained long-term global transport and deposition of P. The global average lake sediment P sequestration rate in the Early and Middle Holocene (before ~4000 cal BP), i.e., with limited human impacts, is ~0.16 Tg P yr^{-1}. This doubled between ~4000 cal BP and 1850 CE to an average of ~0.32 Tg P yr^{-1} (Table S2), and increased ten-fold after 1850 CE to an average of ~1.54 Tg P yr^{-1}. This result emphasizes the extent to which human activity has caused global lake P inputs and sequestration to accelerate above the natural background. Our estimate of the global lake sediment P sequestration rate of ~1.54 Tg P yr^{-1} for recent times is of the same order of magnitude as the value of P retention in freshwaters (5.4 ± 3.2

Tg P yr^{-1}) reported by Yuan et al. (2018). Although this study is the first global review of long-term lake P records, and therefore represents the best estimates of lake P sequestration to date, we speculate that our estimate of ~1.54 Tg P yr^{-1} likely underestimates current rates of global P sequestration in lakes, considering the number of lakes represented here and the widely reported increases in P supply over the period 1850 CE to present. Burial in lake sediments represents one, if not the most important, mechanism for retaining P in freshwater systems as a whole.

Quantifying the fate of P in freshwater ecosystems is important for validating ESMs that attempt to model biogeochemical cycles (Lacroix et al., 2020). Yet most global ESMs do not account for human impacts on terrestrial and freshwater P cycles during the Holocene (Reed et al., 2015). Missing or low-resolution long records are an inherent problem that precludes accurate estimation of the global lake sediment P sink throughout most of the Holocene period. Here, we provide a conservative estimate of ~2686 Tg P for the global lake sediment P sink (in all biomes) over the Holocene (Table S2). As not all biomes are represented in this study, it is not possible to make conclusive statements on the contribution of the different biomes to the global P sink, however this study represents the best estimate of the global Holocene lake sediment P sink to date. This sequestered P pool is permanently stored by lake sediments, removing it from the P cycle of terrestrial ecosystems. This significant P sink has not been considered in the framework of ESMs on centennial to millennial timescales. Incorporating global lake sediment P sequestration rates and total P sinks during the Holocene (~2686 Tg P) into the framework of ESMs will improve our understanding of P cycling in the environment and its responses to global climate change.

Our database with long-term records is biased towards, or over-represents, the temperate and boreal continents (Fig. 1a) in the Northern Hemisphere. The Southern Hemisphere and high latitudes of Europe, Asia, and North America are underrepresented, leading to a significant data gap. This highlights the importance of a broad geographic distribution of lake P records and illustrates the need to target underrepresented areas in future studies. Our study also supports previous findings about the imbalanced distribution of paleolimnological study sites (Smil, 2000; Mendonça et al., 2017; Dubois et al., 2018; Escobar et al., 2020). The lack of data from some of the major lakes in these underrepresented areas also affects our calculated global lake P sink, however this value still represents the most comprehensive global estimate to date. Future research should be directed towards improving this dataset with more well-dated and long-term P records from underrepresented areas.

4. Conclusions

Using lake sediment P records from 108 lakes across the globe, we present the most comprehensive study of anthropogenic modification of P sequestration in lake sediments over the Holocene. Consistent with the widely recognized human perturbation of the global P cycle from the industrial period (1850 CE) to the present day, we show the highest lake sediment P burial rates occurred over the last 100-200 years, coinciding with an expansion in population, increased sewage inputs, agricultural intensification, and the use of chemical fertilizers. This highlights a significant perturbation of the natural global P cycle during this period. Crucially, our results reveal that human activity has impacted the global terrestrial P cycle from the Late Holocene (~4000 cal BP), showing that not only do anthropogenic impacts on the global P cycle extend over millennial timescales, but that this pattern is spatially much wider than previously recognized. We find the timings of early increases in P mobilization differed among Europe, Asia [China], and North America, largely related to different land-use practices.

Combining the lake sediment records from our global dataset, we provide the estimates of the total Holocene P sequestration in lake sediments (~2686 Tg P), representing a substantial global P sink. Incorporating this value into the framework of ESMs will improve our understanding of long-term P cycling and its responses to global climate change. However, despite being the most comprehensive study on long-term global lake sediment P burial to date, our database is biased towards the Northern Hemisphere. We therefore reiterate the need for future research to be directed towards collecting well-dated and long-term lake sediment P records from underrepresented areas.

CRediT authorship contribution statement

Luyao Tu: Conceptualization, Methodology, Formal analysis, Writing-Original Draft, Visualization, Data Curation, Funding acquisition. **Madeleine Moyle:** Methodology, Validation, Writing-Review & Editing. **John F. Boyle:** Methodology, Validation, Writing-Review & Editing. **Paul D. Zander:** Methodology, Validation, Writing-Review & Editing. **Tao Huang:** Validation, Visualization, Writing-Review & Editing. **Lize Meng:** Resources, Writing-Review & Editing. **Changchun Huang:** Supervision, Validation, Writing-Review & Editing. **Martin Grosjean:** Methodology, Supervision, Validation, Funding acquisition, Writing-Review & Editing. **Xin Zhou:** Supervision, Funding acquisition, Validation, Writing-Review & Editing.

Declaration of Competing Interest

The authors declare that they have no competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary Information

Supplementary information will be available in the online version of the paper.

Data availability

The data will be available at Mendeley Data Repository.

Acknowledgements

This work was supported by the Swiss National Science Foundation (grant number 200020_204220), National Science Foundation of China (grant number 211410B32208), Natural Science Research of Jiangsu Higher Education

Institutions of China (grant number 211410B52211), the National Key R&D Program of China (grant number 2022YFF0801101), and a research grant from Nanjing Normal University. We thank the authors who contributed their data for the purpose of this work. We thank Dr. Mark Brenner and an anonymous reviewer for their constructive and thoughtful comments.

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