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Urban point sources of nutrients were the leading cause for the historical spread of hypoxia across European lakes

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Enhanced phosphorus export from land into streams and lakes is a primary factor driving the expansion of deep-water hypoxia in lakes during the Anthropocene. However, the interplay of regional scale environmental stressors and the lack of long-term instrumental data often impede analyses attempting to associate changes in land cover with downstream aquatic responses. Herein we performed a synthesis of data that link paleolimnological reconstructions of lake bottom-water oxygenation to changes in land cover/use and climate over the last 300 years in order to evaluate whether the spread of hypoxia in European lakes was primarily associated with enhanced phosphorus exports from either growing urbanization, intensified agriculture or climatic change. We showed that hypoxia started spreading in European lakes around CE 1850 and was greatly accelerated after CE 1900. Socio-economic changes in Europe beginning in CE 1850 resulted in widespread urbanization as well as a larger and more intensively cultivated surface area. However, our analysis of temporal trends demonstrated that the onset and intensification of lacustrine hypoxia were more strongly related to the growth of urban areas than to changes in agricultural areas and the application of fertilizers. These results suggest that anthropogenically-triggered hypoxia in European lakes were primarily caused by enhanced phosphorus discharges from urban point sources. To date, there have been no signs of sustained recovery of bottom water oxygenation in lakes following the enactment of European water legislation in the 1970s to 1980s, and the subsequent decrease in domestic phosphorus consumption.

Anthropocene | lake hypoxia | land cover/uses | meta-analysis | varved sediment

Introduction

Changes in land cover and land use have been identified as important drivers of phosphorus (P) transfers from terrestrial to aquatic systems, resulting in significant impacts on water resources (1–3). In post-World War II Europe, changes in land cover, land use and P utilization caused widespread eutrophication of freshwaters (3). Elevated rates of P release from point sources to surface water bodies increased in step with population increases, with the novel use of P in domestic detergents and with enhanced connectivity of households to sewage systems that generated concentrated effluents (4). The intensification of agriculture and drastic increased use of fertilizers from industrial and manure sources resulted in elevated P concentrations in runoff from diffuse sources (4). These trends have now metastasized from Europe and North America to most nations, which explains the almost global development of eutrophication problems in surface waters (1).

Much of our understanding regarding the interactions among changes in land cover/use, climate and lake eutrophication comes from detailed studies of individual lakes (5), modeling exercises (1), and/or regional-scale syntheses of instrumental data (6,7); these studies are largely based on relatively short time series (8). Depending on the multitudinous local differences in catchment and lake morphology, river transport capacity, climate, geology and regional trajectories in socioeconomic development, the responses of lakes to surrounding land changes can differ greatly in intensity, modalities and kinetics (9–12). Multiple sites need to be investigated in order to quantify a regional trend as well as evaluate local to regional heterogeneities. Only a few studies have interpreted the long-term trajectories of lakes (based on >100-year lake records) in terms of eutrophication on a regional scale by analyzing trends in nutrient and dissolved CO₂ concentrations (13, 14), carbon burial rates (15), cyanobacterial dominance (16) and hypoxia development (17). However, none of these studies considered the temporal dynamics of land cover and use, and only a few studies (16, 17) considered modern land cover. Our current lack of knowledge of the effects arising from cumulative

Significance

Using a compilation of data arising from over 1,500 European watersheds, we have identified the relative role of different drivers in initiating hypolimnetic hypoxia, a critical indicator of lake health. In particular, our regional synthesis of laminated lake sediments indicated a significant acceleration in the spread of lacustrine hypoxia in the 1900s, which occurred well before the general use of commercial fertilizers in the mid-20th century and the onset of supraregional climate warming in the 1970s. The spread of hypoxia was best explained by urban expansion and the associated intensification of anthropogenic point sources of phosphorus, whereby changes in life style increased the discharge of nutrients from treated and raw sewage, and ultimately led to enhanced lacustrine biological productivity.

Reserved for Publication Footnotes

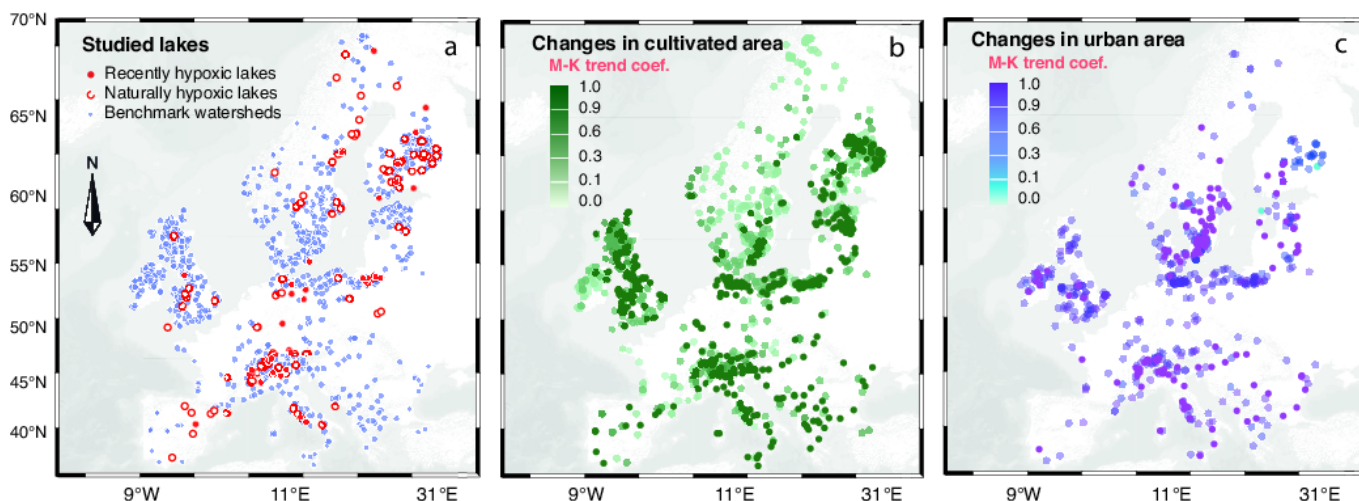


Fig. 1. Location of the 1,607 study sites and changes in land cover over the last 300 years (CE 1700-2000). (a) 51 recently hypoxic lakes (red dots), 97 naturally hypoxic lakes (white dots), and 1,459 benchmark watersheds (blue dots) comprised of 769 from the Lake-Core Database and 690 randomly-selected European lakes from the GLWD database. (b-c) Increases in cultivated areas (%) and urban areas (%) for the last 300 years were observed in all the watersheds according to a Mann-Kendall test, where a higher coefficient indicates a stronger increase (69).

environmental pressures present the potential for a serious underestimation of the long-term impacts of land use changes and hinder our ability to identify the relative importance of P sources to lake ecosystems (18).

Recent progress in land use science has provided an insightful large-scale perspective spanning centuries to millennia (19–22). Additionally, European high-resolution datasets (23, 24) allow for investigations to be conducted at the scale of individual lake watersheds. The present study relies on existing datasets of changes in land cover at the watershed scale (HILDA model (24)), climate data (UDEL model (25)) and a database on the historical onset of hypoxia in lakes (17) to (i) reconstruct the European dynamic of lacustrine hypoxia during the Anthropocene, and (ii) decipher whether P from diffuse sources (agriculture) or point sources (urbanization) is responsible for the spread of lacustrine hypoxia in Europe.

One widely-studied response of lakes to eutrophication is bottom water hypoxia ($[O_2] < 2 \text{ mg L}^{-1}$). Bottom water hypoxia in lakes is detrimental not only for the biota that would normally inhabit oxic aquatic and benthic environments, but also facilitates biogeochemical reactions that generate methane and further mobilize pollutants from previously-accumulated sediment, including P (26-28). Hypoxia can develop naturally, but more often is the result of: (i) cultural eutrophication which enhances biomass production and ultimately its decomposition through microbial oxygen respiration (29-31), and (ii) rising mean temperatures which decrease oxygen solubility in water (32), stimulate microbial oxygen respiration (30), and/or strengthen thermal stratification (33, 34). Among these forcing mechanisms, recent paleolimnological studies identified excess P availability, and not climate, as the main driver for the onset of lacustrine hypoxia during the Anthropocene (17, 35). These studies used the presence and environmental signals of varved (i.e. annually-laminated) sediments in lakes distributed in the French Alps and worldwide to assess the long-term dynamics of hypoxia. Indeed, hypoxic conditions are recorded in lake sediments by virtue of preserved laminations after crossing a critical threshold in bottom water oxygenation that prevent macrobenthic bioturbation in the deeper parts of basins (35). The onset of sustained lamination (including varves) in modern lake sediments is an unambiguous and independent proxy for the timing of hypoxic, anoxic (i.e. complete absence of oxygen), or even euxinic (i.e. sulfidic) bottom water conditions on a regional scale. The well-defined geochronology

of lacustrine varves provides forensic evidence to quantify the timing, prevalence and causes of aquatic regime shifts (17).

Additive mixed-effect models (36) were used to analyze temporal trends and to depict differences among groups of watersheds in Europe: (i) 51 watersheds with lakes recording recent hypoxia onset; (ii) 97 watersheds with lakes recording natural hypoxia; (iii) 769 benchmark watersheds extracted from the *Lake Core Database* (37); and, (iv) 690 benchmark watersheds from the *Global Lakes and Wetlands Database* (GLWD) (38). Lakes of the GLWD have been selected randomly in Europe to represent various gradients of human pressure, climate conditions, land cover and land use.

Results

Our sampling captured the wide ranges of lake morphometric properties, catchment sizes, modern human activities and climatic conditions that are spread across Europe (Figs. 1a, S1; Table S1). General trends in land cover change in Europe during the last 300 years corresponded to increases in the percentages of urban and cultivated areas, albeit some regions were more affected than others (Fig. 1b-c).

Based on our analyses (see Methods), we found that the fraction of lakes recording hypoxia in Europe increased over the past 300 years, from an initial annual rate of $0.06 \pm 0.004 \text{ \% a}^{-1}$ (Pearson's test, $p < 0.0001$) between CE 1850 and 1900 to rates of $0.20 \pm 0.01 \text{ \% a}^{-1}$ between 1900 and 2000 CE ($p < 0.0001$; Fig. 2a). In total, we found that 51 lakes shifted to hypoxia during the last 300 years (Table S2). The catchments of these 51 lakes with recent hypoxia onset had higher percentages of both cultivated and urban areas in CE 2000 than the benchmark watersheds (Fig. S2). Furthermore, most of the lakes with recent hypoxia onset were low elevation sites (48/51 were situated between sea level and the 1000 m above). We also found that the patterns of historical change in land cover and land use for these 51 lakes were best described by nonlinear (i.e. additive mixed-effect) models; urbanized areas increased sharply at the end of the 19th century (from 0.02 % in CE 1700 to 4.1 % in CE 2000), whereas the proportion of cultivated lands have expanded more gradually since the early 18th century (from 7.8 % in 1700 to 23.4 % in 2000) and occurred well before the first spread of hypoxia (Fig. 2b-c). More than half of the 51 lakes shifted to hypoxia before the introduction of fertilizers in Europe in the middle of the 20th century. Climate warming, as well as changes in precipitations,

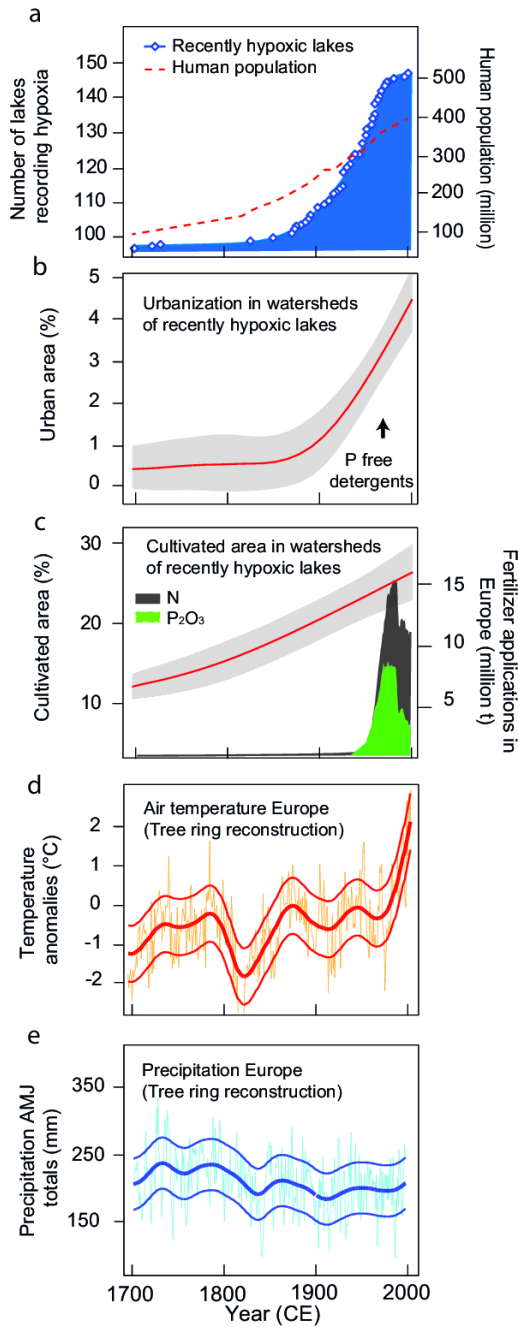


Fig. 2. Trends in the prevalence of lake hypoxia and urbanization as well as observed climate change dynamics during the past 300 years in Europe. (a) Spread of lacustrine bottom water hypoxia shown as a cumulative number of lakes (blue curve) based on the onset of varve deposition in lake sediments from the 51 lakes subset, and the human population in these watersheds (red dashed curve). (b) Percentages of urban and (c) agriculturally-cultivated areas in watersheds of the 51 lakes that shifted to hypoxia during the last 300 years. In (b, c), temporal trends and 95% confidence intervals were calculated according to centennial land use data and the additive mixed-effect model (AMM). Black arrow in (b) indicates early European water legislation in the 1970s and 1980s (70). Dark grey and green shaded peaks in (c) indicate the respective nitrogen (N) and phosphate (P_2O_5) fertilizer applications in the European Union since the 1950s (71). European trends in (d) air temperature and (e) April-May-June (AMJ) precipitation reconstructed from tree rings (72).

is also an unlikely primary driver for the onset of hypoxia as the

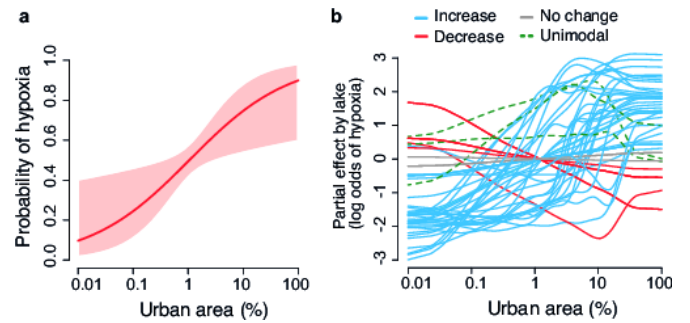


Fig. 3. Probability of hypoxia onset increased as a function of urban area (%) in the 51 lake subset. The logistic GAMM showed that the probability of a hypoxia onset in lakes increased as the proportion of urban area increased over the last 300 years (a). The random smooth logistic GAMM further detected that the vast majority of lakes experienced an increase in probability of hypoxia as urban land cover increased but that the timing of the onset varied among lakes (b).

Table 1.

Random slope logistic GAMM	edf	Ref.df	Chi.sq	p-value	Signif.
s(Urban area)	34.0	46.0	118.6	6.3e-14	***
s(Cultivated area)	1.9	2.3	3.5	0.24	
s(Pastured area)	3.3	4.1	5.2	0.27	

Random smooth logistic GAMM	edf	Ref.df	Chi.sq	p-value	Signif.
s(logUrban, Lake)	65.4	204	200.2	<2e-16	***

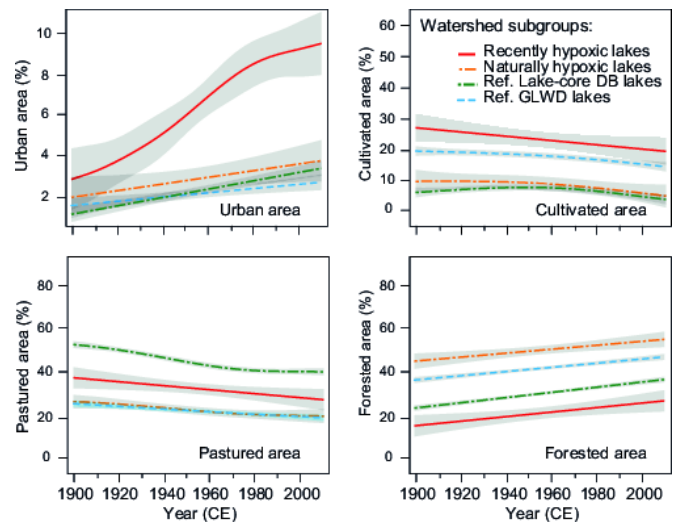


Fig. 4. One hundred year trends for land cover in Europe based on an additive mixed-effect model (AMM), grouping watersheds according to their history of downstream lake hypoxia or reference source. Trends in Europe represent decadal percentages of urban, cultivated, grassland and forested areas. Note the higher increase in urbanization for the recently hypoxic sites during the last 110 years compared to the reference sites. Grey bands indicate 95% confidence intervals of the predicted means based on the AMM.

main warming signal in the air-temperature record post-dates the initial spread of hypoxia (Fig. 2d-e).

Our statistical analyses support the conclusion that urban point sources were the leading driver for the onset of hypoxia.

Using a general additive mixed model (i.e. GAMM) we found that the probability of hypoxia onset in our 51 lake subset increased as the proportion of urban area increased over the last 300 years ($p < 0.0001$), but was unrelated to the changes in cultivated and pastured land area ($p > 0.1$) (Fig. 3; Table 1; $R^2 = 0.23$). A common observation across the lakes with hypoxia developing only recently is the acceleration of urbanization around CE 1900 that coincided with the onset of hypoxia (Fig. S3). However, the timing of hypoxia onset was quite variable across lakes (Fig. 3b). The varve records showed no evidence of a sustained return to improved oxygenated conditions, despite many efforts of remediation (Fig. 2a).

Centennial trends in land cover for the 51 watersheds differed notably from trends in the 97 study watersheds recording natural hypoxia (i.e. sites with sustained varves for >300 years; see Methods) and the 1,459 benchmark watersheds (Fig. 4). The rate of expansion of urban areas was significantly higher in watersheds associated with recent hypolimnetic hypoxia than in other European watersheds (Table 1, Fig. 4). To the contrary, the rate of changes in cultivated and pastured areas are similar in watersheds with recent hypoxic lakes and other European watersheds, although the absolute magnitude of cultivated land was generally higher around the sites with recent hypoxia. M-K tests indicated that hypoxia onsets were preceded by centennial increases in urban (45 % of the sites), cultivated (95 % of the sites) and pastured areas (80 % of the sites). However, during the transition toward hypoxia (± 20 years centred on the time of the onset), urban areas expanded in 71 % of the sites, whereas cultivated and pastured areas were decreasing in 61 % and 74 % of the sites, respectively (Tables S3, S4). Collectively, these findings suggest that urban point sources of nutrients were the leading factor explaining the spread of lacustrine hypoxia in Europe over the last few centuries. The prevailing importance of urban point sources of nutrients as the preeminent trigger towards the spread of hypoxia was also validated by M-K and AMM analyses of decadal-scale landscape and climatic reconstructions spanning the period between CE 1900-2010 (24; Figs. 4, S3, S4). Finally, basin-scale analyses of modern characteristics confirmed the prevailing importance of local human activities on the presence of hypoxia (Fig. S5).

Discussion

This regional-scale analysis of paleolimnological records adds to the growing evidence that modern human activities are a widespread force in shaping the structure and functioning of inland waters (13, 39–42). Our previous paper (17) demonstrated that the spread of lacustrine hypoxia at the global scale was predominantly the result of non-descript human impact. However, the current study specifically pinpoints urban point sources of nutrients as the main forcing mechanism within Europe. Based on earlier water quality studies and paleoecological data, it is known that algal blooms decreased water transparency for most lakes in Europe starting in the middle to late 19th century (37). The eutrophication phase was often more pronounced beginning CE ~ 1950 (37, 43), but the development of widespread hypolimnetic hypoxia has largely predated the more visible effects of eutrophication in the epilimnion (this study). As such, the spread of hypolimnetic hypoxia can be considered as an early warning of eutrophication, caused by enhanced sediment and organic matter fluxes towards bottom waters. The hypolimnion acts as an integrator of processes taking place over the entire water column.

It is generally well accepted that contemporary freshwater eutrophication is predominantly caused by diffuse P sources, principally from agriculture (2), in developed nations (i.e. nations having very high human development in 2014 according to the UN Human Development Index). In contrast, the situation in developing nations is mixed and includes diffuse sources of P

and domestic point sources (18). However, our analysis of longer-term trends in Europe (Fig. 2) provides an important historical perspective, whereby intensive fertilization of agricultural soils and associated diffuse sources of P and N increased through the middle of the 20th century largely post-dated the initial spread of lake hypoxia (Fig. 2a, c) (2, 18). As such, diffuse sources of P appear to have had a subordinate role compared to point sources for most of the last 300 years, and were not decisive for the onset of lacustrine hypoxia in most of the studied lakes. However, nutrient arising from agricultural areas likely had some effect as the long-term M-K tests demonstrated that hypoxia onsets were preceded by increases in cultivated and pastured areas, as well as urban areas. Overall, we suggest that lakes have suffered a slow loss of resilience as a result of both point and diffuse P inputs over time until a disproportionate increase from urban point sources tipped the balance towards hypoxia.

In present-days Europe and North America, domestic sewage and industrial waste water mostly receive an efficient treatment, including P removal prior to discharging effluents into lakes (44). However, the situation around the end of the 19th century was quite different as urban waste waters with increasing P content were directly discharged into waterways (44) and began affecting downstream aquatic ecosystems. The problem was fuelled by urban expansion, a growing population, an accelerating economy during the industrial revolution, the rising standard of living, and novel domestic and industrial uses of P (45). The first P-containing detergents were introduced around the end of the 19th century and soon enjoyed wide acceptance (45). All of these developments were synchronous with the rapid spread of lake hypoxia.

Importantly, our study shows that lakes with recent hypoxia shifted abruptly and irreversibly to an alternate stable state. For instance, among the lakes considered in this study, three peri-alpine lakes (Geneva, Bourget, Annecy, Fig. S6) that were previously oxygenated over the last millennia shifted to hypolimnetic hypoxia between CE 1930 and CE 1950 following a slight P increase (i.e. with enrichments of only $\sim 8\text{--}10 \mu\text{g P L}^{-1}$; (35, 46–47). This illustrates that even a small increase in P availability can stimulate enough primary productivity to trigger hypoxia without generating algal blooms (as blooms were only observed after CE 1950). Likewise, the temporal trend of oxygenation in European lakes (Fig. 2a) shows a slowing down of rate of increase, but no turning off of hypoxia after the 1980s, despite the implementation of restoration programs and successful controls on nutrient influx. The crossing of critical thresholds of nutrient loading appeared to have abruptly and irreversibly shifted lacustrine ecosystems from one state to another (48). Imported P, both from watersheds (external load) and remobilized from lake sediments (internal load) can explain the stability of hypoxia over the last ca. 30 to 40 years. P loads from watersheds to downstream lakes initially accumulate in lake sediments, but later may be remobilized from sediments into overlying waters under hypoxic conditions. P-rich sediments have been identified as the key factor in sustaining hypoxia (49, 50). For instance, the accumulation of organic matter during eutrophic conditions and the subsequent diagenetic release of P from near-surface sediments is known to cause lakes to remain in a eutrophic state even if the external input of P has diminished (1). In addition, a reduced ability of ecosystems to remove nitrogen *via* denitrification and anaerobic ammonium oxidation may be related to hypoxia and could lead to accelerated eutrophication (49). Finally, an increase in water temperature could also decrease the threshold of P concentrations sustaining hypoxia, with more intense stratification, reduced solubility of oxygen at higher water temperatures and enhanced metabolic rates in warmer bottom waters (51).

Unfortunately, the lack of past land cover data at a sufficiently high spatial resolution in other regions prevents from

expanding this work globally. Nonetheless, the observed regime shifts to new stable hypoxic conditions highlight the challenges for developing countries facing persistent diffuse P emissions, growing P demands together with changes in lifestyle (e.g., diet shifts), and expanding urban areas (including the development of megacities and peri-urbanization). Moreover, wastewater from sewage and industry is often untreated and may be the primary contributor towards eutrophication (52). For example, only 35 % of wastewater in Asia and <1 % in Africa were treated in CE 2005 (52). Without implementation of wastewater treatment of P in point and mixed sources, the future of lakes in these regions will likely result in prevalent hypoxic hypolimnetic conditions, degraded water quality and the necessity for decade-long restoration efforts.

In conclusion, our analyses of varved sediment records indicate that nutrient point sources from growing urban areas were the leading driver for the onset of hypoxia in the hypolimnion of downstream lakes. Point and diffuse sources have always both contributed to the total supply of nutrient inputs to lakes, but with varying intensities over time and space. Our results show that urban point sources of P were the dominant driver of lake eutrophication in European lowland systems during the Anthropocene. During the last few decades, the relative contribution of diffuse P sources has progressively become a major cause of modern freshwater eutrophication in developed countries, as point sources have been reduced and fertilizer use has increased. The lack of re-oxygenation of the hypolimnion evident from our analyses highlight the importance of the history and legacy of past land uses, and the need for long-term strategies to maintain and restore water quality in modern lake ecosystems.

Materials and Methods

Reconstructing the dynamics of hypoxia

The sediment textures of many lakes offer a simple proxy for the oxygenation history of bottom waters (53-55). Indeed, the appearance of laminated sediment on top of homogeneous sediment indicates that annual oxygenation conditions fell below a critical threshold in both duration and concentration (35, 56, 57), hence recording the die-out of macrobenthos and the end of its related bioturbation (Fig. 57, 54, 58, 59). If laminations are proven to reflect annual cycles of sedimentation, they offer the additional advantage that the shift from well-oxygenated to at least seasonal hypoxic hypolimnetic conditions can often be precisely dated by counting varves from the sediment/water interface down-core (54). The Varves Working Group of PAGES (VWG) has intensively investigated varved lakes over the last decade (54, 60, 61), enabling the assembly of a large dataset of lake hypoxia (17). In Europe, 148 varved sediment records were referenced in the global compilation of the VWG (17) and indicated that the European dynamics of lacustrine hypoxia encompassed: (i) a period of relatively undisturbed conditions prior to CE 1850 serving as a pre-industrialized baseline reference; (ii) a period of major changes during the early industrialization of western countries and the following "Great Acceleration" phase of the so-called Anthropocene (42); and, (iii) the initiation of European lake restoration programs since the 1970s. Land use changes in watersheds of recently varved lakes have been compared to a set of 97 naturally varved lakes in order to dismiss any sampling bias related to morphometric properties. Preservation of laminated sediments usually indicates that lakes have strong hypoxia; however, strong seasonal hypoxia may not systematically develop laminations, notably due to the absence of contrasting seasonal sedimentation, or as a consequence of wind causing sediment resuspension. Our data matrix does not attempt to include all lakes with hypoxia but instead includes a conservative and large selection of well-characterized lakes with laminated sediments to provide a statistically sound and relevant basis for constraining the dynamics of hypoxia in Europe.

Paleolimnological data

A literature search was conducted in April 2014 (17) and updated in June 2015 using the ISI Web of Science database and Google Scholar with different combinations of the following keywords: 'varve' and 'lake', and 'lamin' and 'lake sediment'. The search yielded 148 relevant European lakes that contain laminated or varved sediments. Descriptions and data on varved sites, sediments and dating methods are available in (17) and references

therein. The original chronologies were expressed in Common Era (CE) calendar years. Laminated lacustrine sites had to satisfy several conditions in order to be included in this synthesis. Accepted sites (i) contained a varved or well-preserved laminated sedimentary sequence, (ii) featured a published age-model relying on varve counting and/ or radiometric dating, and (iii) the lakes' sediment texture had to be explicitly described or illustrated by pictures outlining the laminated intervals. The timing of the first onset of hypoxia was obtained for each lake by examining all relevant published varve data. Where time intervals could not be dated precisely with the help of published data, corresponding authors were contacted and asked for advice. The water depth for each lake was collected and used to verify that lake level fluctuations were not the cause of changes in preservation conditions of the varves. Descriptions and data for lake sites were compiled in this study (Table S1).

Land use and climate data

Modern data and temporal changes in land use and climate during the last 300 years were analyzed for 1,607 watersheds. Hydrological basins of each site were calculated using the flow accumulation and flow direction rasters made available from HydroSHEDS together with lake perimeters and areas, using the Global Lakes and Wetlands Database from the World Wildlife Foundation (38). The following variables were extracted from modeled areas using the geographic information system ArcGIS: (i) modern site characteristics; (ii) past land use from CE 1900 to 2010 at decadal steps and with a 1 km² spatial resolution and (iii) past land use from CE 1700 to 2000 with centennial resolution. Mean local temperatures, precipitation, population densities (62), changes in urban area, cultivated, pastured and forest areas (24) as well as past human population densities (63) were extracted from modeled areas for each watershed.

Numerical analysis

An additive mixed-effect model (AMM) framework generated using the *mgcv* library in R (64) was used to describe the general nonlinear trends in land uses over the last three centuries. It was anticipated from (17) that watersheds with recently hypoxic lakes would contain an environmental signal reflecting a more urbanized and agriculturally-cultivated landscape compared to watersheds serving as benchmarks as well as naturally hypoxic lakes. Thus, the relationships among urban, cultivated, pastured and forested areas were evaluated for the four watershed categories of this study. Confidence intervals were derived using the standard errors produced by the *predict.gam* function in R (65), with type = 'response' specified in the model (*mgcv* library (66)).

Multiple regression analyses were conducted to identify the main drivers of hypoxia onsets. For each recently hypoxic lake ($n = 51$), we created a binomial time series indicating whether the first hypoxic event had occurred or not at each date of the land cover data (i.e. 1700, 1800, 1900, 1910, ... , 2010). To test the relative importance of the different land cover types, we then ran a logistic general additive mixed-effect models (GAMM) using the binomial time series as the response variable, the percentage of urban, cultivated and pastured areas as fixed effect explanatory variables, and lake ID as the random effect (testing a random slope and intercept for each lake). To further test whether the smooth term varied among lakes, we tested a random smooth logistic GAMM, which not only allowed the slope to vary among lakes but also the shape of the nonlinear relationship. All GAMMs were fit using the *bam* function of the *itsadug* package in R (67).

Non-parametric Mann-Kendall (M-K) tests for monotonic trends were used to quantify trends of land use for each of the 1,607 watershed time series within the past 300 years. This analysis was based on the Kendall rank correlation coefficient and was conducted using the Kendall library (68). A positive score shows a monotonically increasing trend, whereas a negative value shows a monotonically decreasing trend (69). For each site, M-K tests were run for two time windows to identify the potential effects of slow and fast land cover changes on the hypoxia onset: (i) we anticipated that fast changes in the land cover would show an effect within a short period of time (± 20 years) centered on the time of the onset to be consistent with the uncertainties of reconstructions, and (ii) slow changes in the land cover would show an effect over a longer period of time (~ 200 years) preceding the onset of hypoxia to be consistent with the long-term history and potential legacy effects of past land changes in Europe.

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cation of lakes and flowing waters with particular reference to nitrogen and phosphorus as factors in eutrophication. (OECD, Paris, France).

1. Carpenter SR (2005) Eutrophication of aquatic ecosystems: Bistability and soil phosphorus. *Proc Natl Acad Sci USA* 102(29):10002-10005.
2. Carpenter SR, et al. (1998) Nonpoint pollution of surface waters with phosphorus and nitrogen. *Ecol Appl* 8(3):559-568.
3. Vollenweider RA (1968) *Water management research. Scientific fundamentals of the eutrophication of lakes and flowing waters with particular reference to nitrogen and phosphorus as factors in eutrophication.* (OECD, Paris, France).
4. Garnier J, et al. (2015) Phosphorus budget in the water-agro-food system at nested scales in two contrasted regions of the world (ASEAN-8 and EU-27). *Glob Biogeochem Cycles* 29(9):1348-1368.

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5. Patterson RT, Dalby A, Kumar A, Henderson LA, Boudreau REA (2002) Arcellaceans (thecamoebians) as indicators of land-use change: settlement history of the Swan Lake area, Ontario as a case study. *J Paleolimnol* 28(3):297–316.

6. Taranu ZE, Gregory-Eaves I (2008) Quantifying relationships among phosphorus, agriculture, and lake depth at an inter-regional scale. *Ecosystems* 11(5):715–725.

7. O'Reilly CM, et al. (2015) Rapid and highly variable warming of lake surface waters around the globe. *Geophys Res Lett* 42(24):2015GL066235.

8. Anneville O, Gammeter S, Straile D (2005) Phosphorus decrease and climate variability: mediators of synchrony in phytoplankton changes among European peri-alpine lakes. *Freshw Biol* 50(10):1731–1746.

9. Ramstack J, Fritz S, Engstrom D (2004) Twentieth century water quality trends in Minnesota lakes compared with presettlement variability. *Pap Earth Atmospheric Sci*. Available at: <http://digitalcommons.unl.edu/geosciencefacpub/43>.

10. Fraterrigo JM, Downing JA (2008) The Influence of Land Use on Lake Nutrients Varies with Watershed Transport Capacity. *Ecosystems* 11(7):1021–1034.

11. Pham SV, Leavitt PR, McGowan S, Peres-Neto P (2008) Spatial variability of climate and land-use effects on lakes of the northern Great Plains. *Limnol Oceanogr* 53(2):728–742.

12. Perga ME, et al. (2015) High-resolution paleolimnology opens new management perspectives for lakes adaptation to climate warming. *Front Ecol Evol* 3:72. doi: 10.3389/fevo.2015.00072

13. Keatley BE, Bennett EM, MacDonald GK, Taranu ZE, Gregory-Eaves I (2011) Land-use legacies are important determinants of lake eutrophication in the Anthropocene. *PLoS ONE* 6(1):e15913.

14. Anderson NJ, Bennion H, Lotter AF (2014) Lake eutrophication and its implications for organic carbon sequestration in Europe. *Glob Change Biol* 20(9):2741–2751.

15. Perga M-E, et al. (2016) A century of human-driven changes in the carbon dioxide concentration of lakes. *Glob Biogeochem Cycles* 30(2):2015GB005286.

16. Taranu ZE, et al. (2015) Acceleration of cyanobacterial dominance in north temperate-subarctic lakes during the Anthropocene. *Ecol Lett* 18(4):375–384.

17. Jenny J-P, et al. (2016) Global spread of hypoxia in freshwater ecosystems during the last three centuries is caused by rising local human pressure. *Glob Change Biol* 22(4):1481–1489

18. Bennett EM, Carpenter SR, Caraco NF (2001) Human impact on erodable phosphorus and eutrophication: A global perspective increasing accumulation of phosphorus in soil threatens rivers, lakes, and coastal oceans with eutrophication. *BioScience* 51(3):227–234.

19. Pongratz J, Reick C, Raddatz T, Claussen M (2008) A reconstruction of global agricultural areas and land cover for the last millennium. *Glob Biogeochem Cycles* 22(3):GB3018.

20. Verburg PH, et al. (2015) Land system science and sustainable development of the earth system: A global land project perspective. *Anthropocene*. doi:10.1016/j.ancene.2015.09.004.

21. Vliet J van, et al. (2015) Meta-studies in land use science: Current coverage and prospects. *Ambio* 45(1):15–28.

22. Klein Goldewijk K, Beusen A, van Drecht G, de Vos M (2011) The HYDE 3.1 spatially explicit database of human-induced global land-use change over the past 12,000 years. *Glob Ecol Biogeogr* 20(1):73–86.

23. Fuchs R, Herold M, Verburg PH, Clevers JGPW (2013) A high-resolution and harmonized model approach for reconstructing and analysing historic land changes in Europe. *Biogeosciences* 10(3):1543–1559.

24. Fuchs R, Herold M, Verburg PH, Clevers JGPW, Eberle J (2015) Gross changes in reconstructions of historic land cover/use for Europe between 1900 and 2010. *Glob Change Biol* 21(1):299–313.

25. Willmott CJ, Robeson SM (1995) Climatologically aided interpolation (CAI) of terrestrial air temperature. *Int J Climatol* 15(2):221–229.

26. Frossard V, et al. (2013) Chironomid assemblages in cores from multiple water depths reflect oxygen-driven changes in a deep French lake over the last 150 years. *J Paleolimnol* 50(3):257–273.

27. R A Mah, D M Ward, L Baresi, Glass and TL (1977) Biogenesis of methane. *Annu Rev Microbiol* 31(1):309–341.

28. Wahlen M (1993) The Global methane cycle. *Annu Rev Earth Planet Sci* 21(1):407–426.

29. Nixon SW (1995) Coastal marine eutrophication: a definition, social causes, and future concerns. *Ophelia* 41:199–219.

30. Müller B, Bryant LD, Matzinger A, Wüest A (2012) Hypolimnetic oxygen depletion in eutrophic lakes. *Environ Sci Technol* 46(18):9964–9971.

31. Conley DJ, et al. (2009) Hypoxia-related processes in the Baltic Sea. *Environ Sci Technol* 43(10):3412–3420.

32. Deutsch C, Brix H, Ito T, Frenzel H, Thompson L (2011) Climate-forced variability of ocean hypoxia. *Science* 333(6040):336–339.

33. Straile D, Jöhnk K, Rosksnecht H (2003) Complex effects of winter warming on the physicochemical characteristics of a deep lake. *Limnol Oceanogr* 48(4):1432–1438.

34. Coma R, et al. (2009) Global warming-enhanced stratification and mass mortality events in the Mediterranean. *Proc Natl Acad Sci* 106(15):6176–6181.

35. Jenny J-P, et al. (2014) Inherited hypoxia: A new challenge for reoligotrophicated lakes under global warming. *Glob Biogeochem Cycles* 28(19):1944–19224.

36. Simpson GL, Anderson NJ (2009) Deciphering the effect of climate change and separating the influence of confounding factors in sediment core records using additive models *Limnology and Oceanography* 56(6):2529–2541

37. Battarbee RW, et al. (2010) A paleolimnological meta-database for assessing the ecological status of lakes. *J Paleolimnol* 45(4):405–414.

38. Lehner B, Döll P (2004) Development and validation of a global database of lakes, reservoirs and wetlands. *J Hydrol* 296(1–4):1–22.

39. Wolfe AP, et al. (2013) Stratigraphic expressions of the Holocene–Anthropocene transition revealed in sediments from remote lakes. *Earth-Sci Rev* 116(0):17–34.

40. Crutzen PJ (2002) Geology of mankind. *Nature* 415(6867):23–23.

41. Crutzen PJ, Steffen W (2003) How long have we been in the Anthropocene Era? *Clim Change* 61(3):251–257.

42. Steffen W, Crutzen PJ, McNeill JR (2007) The Anthropocene: are humans now overwhelming

the great forces of nature. *AMBIO J Hum Environ* 36(8):614–621.

43. Vallentyne JR (1974) *The Algal Bowl: Environment Canada, vol. 22. Miscellaneous Special Publication, Ottawa, Ontario*.

44. Selman M, Greenhalgh S, Diaz RJ, Sugg Z (2015) Water quality: eutrophication and hypoxia. Available at: http://pdf.wri.org/eutrophication_and_hypoxia_in_coastal_areas.pdf.

45. Ashley K, Cordell D, Mavinic D (2011) A brief history of phosphorus: From the philosopher's stone to nutrient recovery and reuse. *Chemosphere* 84(6):737–746.

46. Jenny J-P, et al. (2013) A spatiotemporal investigation of varved sediments highlights the dynamics of hypolimnetic hypoxia in a large hard-water lake over the last 150 years. *Limnol Oceanogr* 58(4):1395–1408.

47. Berthon V, et al. (2013) Trophic history of French sub-alpine lakes over the last ~150 years: phosphorus reconstruction and assessment of taphonomic biases. *J Limnol* 72(3):e34.

48. Barnosky AD, et al. (2012) Approaching a state shift in Earth's biosphere. *Nature* 486(7401):52–58.

49. Conley DJ, Carstensen J, Vaquer-Sunyer R, Duarte CM (2009) Ecosystem thresholds with hypoxia. *Hydrobiologia* 629(1):21–29.

50. Colen CV, et al. (2012) Organism-sediment interactions govern post-hypoxia recovery of ecosystem functioning. *PLOS ONE* 7(11):e49795.

51. Meire L, Soetaert KER, Meysman FJR (2013) Impact of global change on coastal oxygen dynamics and risk of hypoxia. *Biogeosciences* 10(4):2633–2653.

52. Howarth R, Ramakrishna K (2005) Nutrient Management. *Millennium Ecosystem Assessment (MA)* (Washington, DC: Island Press.). K. Chopra, R. Leemans, P. Kumar, and H. Simons. Ecosystems and Human Wellbeing: Policy Responses.

53. O'Sullivan PE (1983) Annually-laminated lake sediments and the study of Quaternary environmental changes – a review. *Quat Sci Rev* 1(4):245–313.

54. Zolitschka B, Francus P, Ojala AEK, Schimmelmann A (2015) Varves in lake sediments – a review. *Quat Sci Rev* 117:1–41.

55. Lotter AF, Sturm M, Teranes JL, Wehrli B (1997) Varve formation since 1885 and high-resolution varve analyses in hypertrophic Baldeggersee (Switzerland). *Aquat Sci* 59(4):304–325.

56. Rabalais NN, Turner RE, Union AG (2001) *Coastal hypoxia: consequences for living resources and ecosystems* (American Geophysical Union).

57. Cicchetti G, et al. (2006) Relationships between near-bottom dissolved oxygen and sediment profile camera measures. *J Mar Syst* 62(3–4):124–141.

58. Tylmann W, Zolitschka B, Enters D, Ohlendorf C (2013) Laminated lake sediments in north-east Poland: distribution, preconditions for formation and potential for paleoenvironmental investigation. *J Paleolimnol* 50(4):487–503.

59. Christensen CJ, Gorsline DS, Hammond DE, Lund SP (1994) Non-annual laminations and expansion of anoxic basin-floor conditions in Santa Monica Basin, California Borderland, over the past four centuries. *Mar Geol* 116(3–4):399–418.

60. Francus P, Ridge JC, Johnson MD (2013) The rise of varves. *GFF* 135(3–4):229–230.

61. Ojala AEK, Francus P, Zolitschka B, Besonen M, Lamoureux SF (2012) Characteristics of sedimentary varve chronologies – A review. *Quat Sci Rev* 43:45–60.

62. Center for International Earth Science Information Network - CIESIN - Columbia University, Centro Internacional de Agricultura Tropical - CIAT (2005) Gridded population of the world, Version 3 (GPWv3): population density grid. Available at: <http://dx.doi.org/10.7927/H-4XK8CG2>.

63. Ellis EC, Klein Goldewijk K, Siebert S, Lightman D, Ramankutty N (2010) Anthropogenic transformation of the biomes, 1700 to 2000. *Glob Ecol Biogeogr* 19(5):589–606.

64. Wood S (2006) *Generalized additive models: an introduction with R* (Chapman and Hall/CRC Press).

65. R Development Core Team (2008) *R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria*. Available at: URL <http://www.R-project.org>.

66. Wood SN (2011) Fast stable restricted maximum likelihood and marginal likelihood estimation of semiparametric generalized linear models. *J R Stat Soc Ser B Stat Methodol* 73(1):3–36.

67. Rij J van, Wieling M, Baayen RH, Rijn H van (2016) *itsadug: Interpreting Time Series and Autocorrelated Data Using GAMMs* Available at: <https://cran.r-project.org/web/packages/itsadug/index.html> [Accessed July 5, 2016].

68. McLeod AI (2011) *Kendall: Kendall Rank Correlation and Correlation and Mann-Kendall trend test, R package version 2.2*. Available at: <http://cran.rproject.org/package=Kendall>. Last accessed 5 February 2015.

69. Legendre P, Legendre LFJ (2012) *Numerical Ecology, Volume 24, Third Edition* (Elsevier, Amsterdam), 3 edition.

70. Sands P, Galizzi P (2006) Council Directive 76/160/EEC of 8 December 1975 concerning the quality of bathing water (*OJ L 031 05.02.1976 p. 1*). *Documents in European Community Environmental Law* (Cambridge University Press). Second edition Available at: <http://dx.doi.org/10.1017/CBO9780511610851.053>.

71. European Commission (2010) *The EU Nitrates Directive*. [Online] Available at: <http://ec.europa.eu/environment/pubs/pdf/factsheets/nitrates.pdf> [Accessed 22 1 2012].

72. Büntgen U, et al. (2011) 2500 Years of European climate variability and human susceptibility. *Science* 331(6017):578–582.

73. De Vicente I, Cattaneo K, Cruz-Pizarro L, Brauer A, Guilizzoni P (2006) Sedimentary Phosphate Fractions Related to Calcite Precipitation in an Eutrophic Hardwater Lake (Lake Alserio, Northern Italy). *Journal of Paleolimnol* 35(1):55–64.

74. Meriläinen JJ, Hynynen J, Palomäki A, Mäntykoski K, Witick A (2003) Environmental history of an urban lake: a paleolimnological study of Lake Jyväsjärvi, Finland. *Journal of Paleolimnol* 30(4):387–406.

75. Garibaldi L, Mezzanotte V, Brizzio MC, Rogora M, Mosello R (1999) The trophic evolution of Lake Isèo as related to its holomixis. *Journal of Limnology* 58(1):10–19.

76. Renberg I (1986) Photographic demonstration of the annual nature of a varve type common in N. Swedish lake sediments. *Hydrobiologia* 140(1):93–95.

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