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

Jean-Philippe Jenny[a,b,c,1], Alexandre Normandeau[d], Pierre Francus[a,b], Zofia Ecaterina Taranu[e,f], Irene Gregory-Eaves[e,g], François Lapointe[a,b], Josue Jautzy[h], Antti E. K. Ojala[i], Jean-Marcel Dorioz[j], Arndt Schimmelmann[k], and Bernd Zolitschka[l]

[a]Centre-Eau Terre Environnement, Institut National de la Recherche Scientifique, Quebec, QC, Canada G1K 9A9; [b]Centre de Recherche en Géochimie et Géodynamique (GEOTOP) Research Center, Montreal, QC, Canada H3C 3P8; [c]Max Planck Institute for Biogeochemistry, 07745 Jena, Germany; [d]Geological Survey of Canada, Bedford Institute of Oceanography, Dartmouth, NS, Canada B2Y 4A2; [e]Department of Biology, McGill University, Montreal, QC, Canada H3A 1B1; [f]Departement des Sciences Biologiques, Université de Montréal, Montreal, QC, Canada H2V 2S9; [g]Groupe de Recherche Interuniversitaire en Limnology, McGill University, Montreal, QC, Canada H3A 1B1; [h]Department of Earth Sciences, University of Ottawa, Ottawa, ON, Canada K1N6N5; [i]Geological Survey of Finland, 02151 Espoo, Finland; [j]Institut National de la Recherche Agronomique UMR 42 Centre Alpin de Recherche sur les Réseaux Trophiques des Écosystèmes Limniques (CARRTEL), Université de Savoie, 74203 Thonon-les-bains Cedex, France; [k]Department of Geological Sciences, Indiana University, Bloomington, IN 47405; and [l]Arbeitsgruppe Geomorphologie und Polarforschung (GEOPOLAR), Institute of Geography, University of Bremen, D-28359 Bremen, Germany

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Enhanced phosphorus (P) 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 past 300 years to evaluate whether the spread of hypoxia in European lakes was primarily associated with enhanced P exports from 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. Socioeconomic 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 was primarily caused by enhanced P 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 P consumption.

Anthropocene | lake hypoxia | land cover/uses | meta-analysis | varves

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 between 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 to quantify a regional trend, as well as to 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 CO_2 concentrations (13, 14), carbon burial rates (15), cyanobacterial dominance (16), and hypoxia development (17). However, only one of these studies (13) 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 environmental pressures presents the potential for serious underestimation of the long-term impacts of land use changes and hinders 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 [Historic Land

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 lifestyle increased the discharge of nutrients from treated and raw sewage, and ultimately led to enhanced lacustrine biological productivity.

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[1]To whom correspondence should be addressed. Email: jjenny@bgc-jena.mpg.de.

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Dynamics Assessment model (HILDA v2; ref. 24)], climate data [University of Delaware Air Temperature & Precipitation (UDEL model; ref. 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 ($[O_2] < 2\ \mathrm{mg{\cdot}L^{-1}}$). 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 is more often 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 (AMMs) (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 (Fig. 1*A*, Fig. S1, and Table S1). General trends in land cover change in Europe during the past 300 years corresponded to increases in the percentages of urban and cultivated areas, albeit some regions were more affected than others (Fig. 1 *B* and *C*).

Based on our analyses (*Materials and 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 ± 0.004% per year (a^{-1}) (Pearson's test, $p < 0.0001$) between CE 1850 and 1900 to rates of 0.20 ± 0.01% a^{-1} between CE 1900 and 2000 ($p < 0.0001$) (Fig. 2*A*). In total, we found that 51 lakes shifted to hypoxia during the past 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 of 51 were situated between sea level and 1,000 m above sea level). 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 has expanded more gradually since the early 18th century (from 7.8% in CE 1700 to 23.4% in CE 2000) and occurred well before the first spread of hypoxia (Fig. 2 *B* and *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 precipitation, is also an unlikely primary driver for the onset of hypoxia because the main warming signal in the air-temperature record postdates the initial spread of hypoxia (Fig. 2 *D* and *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 (GAMM), we found that the probability of hypoxia onset in our 51-lake subset increased as the proportion of urban area increased over the past 300 years ($p < 0.0001$), but was unrelated to the changes in cultivated and pastured land area ($p > 0.1$) (Fig. 3 and 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. 3*B*).

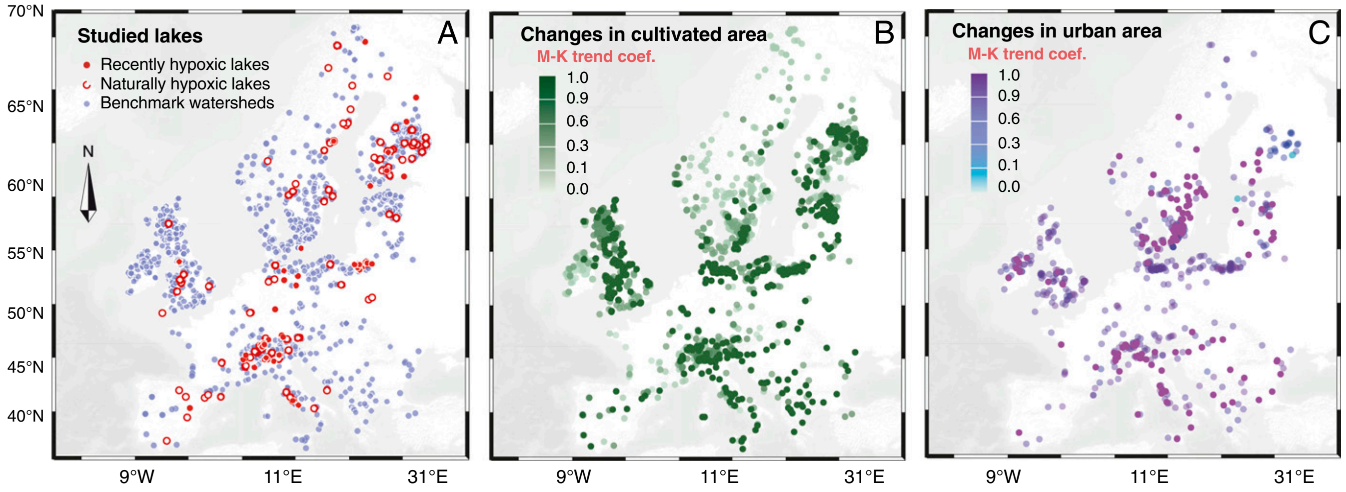

Fig. 1. Location of the 1,607 study sites and changes in land cover over the past 300 years (CE 1700–2000). (*A*) Fifty-one recently hypoxic lakes, 97 naturally hypoxic lakes, and 1,459 benchmark watersheds composed of 769 lakes from the Lake-Core Database and 690 randomly selected European lakes from the GLWD database. (*B* and *C*) Increases in cultivated areas (%) and urban areas (%) for the past 300 years were observed in all of the watersheds according to an M-K test, where a higher coefficient indicates a stronger increase (69).

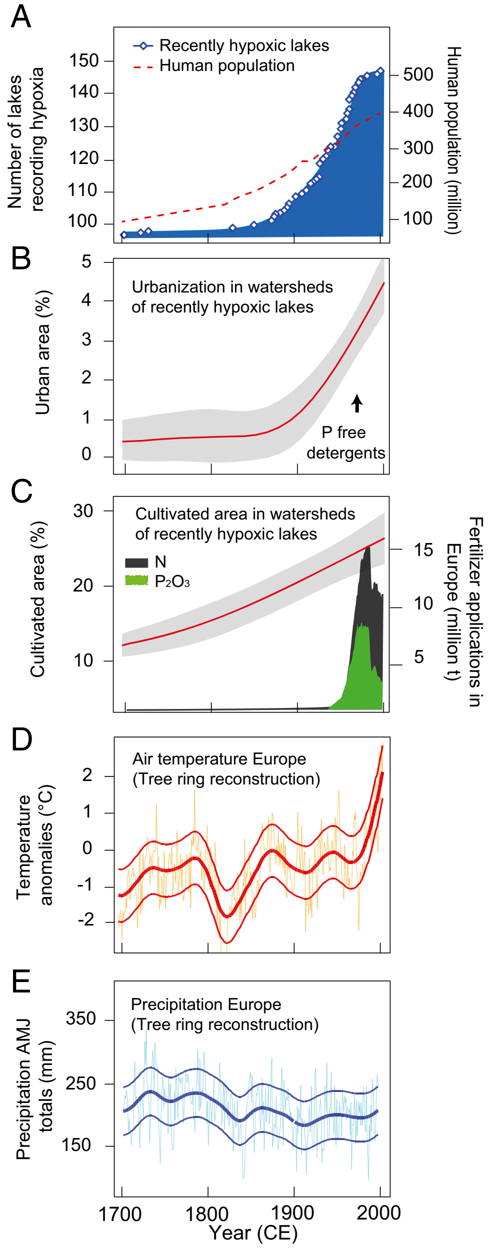

Fig. 2. Trends in the prevalence of lake hypoxia and urbanization, as well as observed climate change dynamics, during the past 300 years in Europe. (*A*) Spread of lacustrine bottom-water hypoxia shown as a cumulative number of lakes (blue curve) based the onset of varve deposition in lake sediments from the 51-lake subset and the human population in these watersheds (red dashed curve). Percentages of urban (*B*) and agriculturally cultivated (*C*) areas in watersheds of the 51 lakes that shifted to hypoxia. In *B* and *C*, temporal trends and 95% confidence intervals were calculated according to centennial land use data and an AMM. The black arrow in *B* indicates early European water nutrient mitigation legislation in the 1970s and 1980s (70). The dark gray 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 air temperature (*D*) and April/May/June (AMJ) precipitation (*E*) are reconstructed from tree rings (72).

Our records showed no evidence of a sustained return to improved oxygenated conditions, despite many efforts at remediation (Fig. 2*A*).

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 laminae for >300 years) (*Materials and 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 (Fig. 4 and Table 1). To the contrary, the rates 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. Mann–Kendall (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 (80% of the sites) areas. However, during the transition toward hypoxia (±20 years centered 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 and 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 past few centuries. The prevailing importance of urban point sources of nutrients as the preeminent trigger toward 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 and 2010 (24) (Fig. 4 and Figs. S3 and 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 function 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 nondescript 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 in 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 an early warning of eutrophication, caused by enhanced sediment and organic matter fluxes toward 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 United Nations 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 nitrogen increased through the middle of the 20th century, largely postdating the initial spread of lake hypoxia (2, 18) (Fig. 2 *A* and *C*). As such, diffuse sources of P appear to have had a subordinate role compared with point sources for most of the past 300 years, and were not decisive for the onset of lacustrine hypoxia in most of the studied lakes. However, nutrients arising from agricultural areas likely had some effect, because 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 toward hypoxia.

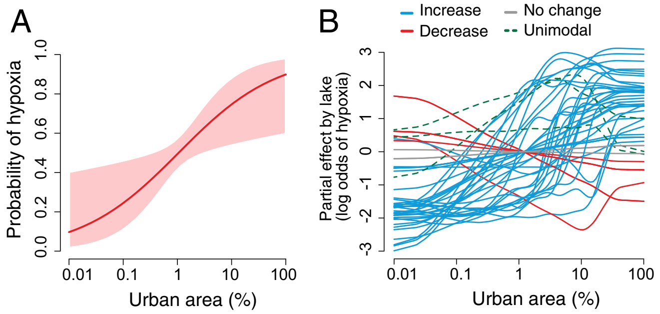

Fig. 3. Probability of hypoxia onset increased as a function of urban area (%) in the 51-lake subset. (*A*) Logistic GAMM showed that the probability of hypoxia onset in lakes increased as the proportion of urban area increased over the past 300 years. (*B*) Random smooth logistic GAMM further detected that the vast majority of lakes experienced an increase in the probability of hypoxia as urban land cover increased, but that the timing of the onset varied among lakes.

In present-day Europe and North America, domestic sewage and industrial waste water mostly receive an efficient treatment, including P removal, before discharging effluents into lakes (44). However, the situation around the end of the 19th century was quite different, when urban waste waters with increasing P content were directly discharged into waterways (44) and began affecting downstream aquatic ecosystems. The problem was fueled 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 perialpine lakes (Lakes Geneva, Bourget, and Annecy) (Fig. S6) that were previously oxygenated over the past millennia shifted to hypolimnic hypoxia between CE 1930 and CE 1950 following a slight P increase (i.e., with enrichments of only ~8–10 μg TP L^{-1}; refs. 35, 46, 47). This finding illustrates that even a small increase in P availability can stimulate enough primary productivity to trigger hypoxia without generating algal blooms (because blooms were only observed after CE 1950). Likewise, the temporal trend of oxygenation in European lakes (Fig. 2*A*) shows a slowing down of the 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 past *ca.* 30–40 years. P loads from watersheds to downstream lakes initially accumulate in lake sediments, but may later 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 the 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).

Table 1. Results of the multiple regression models

Logistic GAMM	edf	Ref. df	χ^2	Probability value	Significance
Random slope					
s(urban area)	34.0	46.0	118.6	6.3×10^{-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					
s(logurban,lake)	65.4	204	200.2	$<2 \times 10^{-16}$	***

A logistic GAMM showed that the probability of hypoxia onset in lakes increased as the proportion of urban area increased over the past 300 years, but was unrelated to the changes in cultivated and pastured land area [adjusted R^2_{adj} (R^2_{ad}) = 0.227]. By incorporating a term that accounted for differences in the trajectory of urban land use among lakes, our logistic GAMM was able to explain a much larger proportion of the variation in the timing of hypoxia onset (R^2_{adj} = 0.396). ***p = 0.001. edf, effective degree of freedom; Ref., reference degree of freedom.

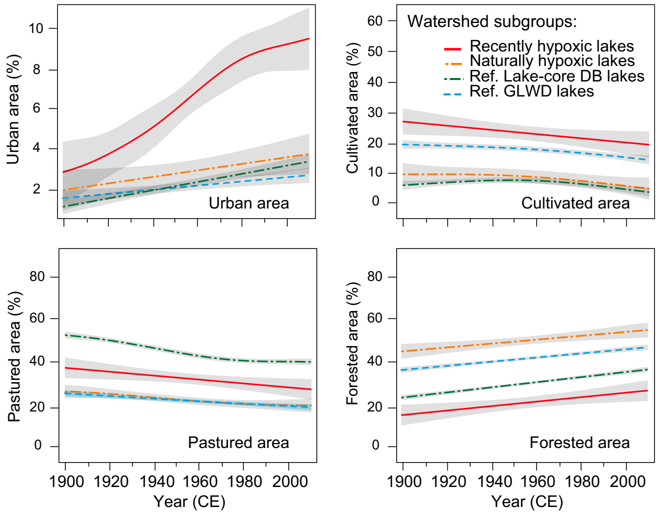

Fig. 4. One-hundred–year trends for land cover in Europe based on an AMM, grouping watersheds according to their history of downstream lake hypoxia or benchmark (Ref.) source. Trends in Europe represent decadal percentages of urban, cultivated, grassland, and forested areas. Note the higher increase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

Unfortunately, the lack of past land cover data at a sufficiently high spatial resolution in other regions prevents us from expanding this work globally. Nonetheless, the observed regime shifts to new stable hypoxic conditions highlights the challenges for developing countries facing persistent diffuse P emissions and growing P demands, together with changes in lifestyle (e.g., diet shifts) and expanding urban areas (including the development of megacities and periurbanization). Moreover, wastewater from sewage and industry is often untreated and may be the primary contributor toward 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 conditions 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 laminated 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 past 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 reoxygenation of the hypolimnion evident from our analyses highlights 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 (54, 58, 59) (Fig. S7). 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 hypolimnic conditions can often be precisely dated by counting varves from the sediment/water interface down-core (54). The Varves Working Group (VWG) of Past Global Changes has intensively investigated varved lakes over the past 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 before CE 1850 serving as a preindustrialized 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 with a set of 97 naturally varved lakes 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.

Numerical Analysis. An AMM framework generated using the *mgcv* library in R (62) was used to describe the general nonlinear trends in land use over the past three centuries. It was anticipated from the study by Jenny et al. (17) that watersheds with recently hypoxic lakes would contain an environmental signal reflecting a more urbanized and agriculturally cultivated landscape compared with watersheds serving as benchmarks, as well as naturally hypoxic lakes. Thus, the relationships between urban, cultivated, pastured, and forested areas were evaluated for the four watershed categories of this study. Confidence intervals were derived using the SEs produced by the *predict.gam* function in R (63), with type = "response" specified in the model [*mgcv* library (64)]. Multiple regression analyses and nonparametric M-K tests for monotonic trends were conducted to identify the main drivers of hypoxia onsets (*SI Materials and Methods*).

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