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A 150-year river water quality record shows reductions in phosphorus loads but not in algal growth potential

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Eutrophication and proliferation of nuisance and harmful algal blooms are a major cause of water-quality impairment globally. Here, we analyze the world's longest continuous (150-year) river water-quality dataset for the River Thames, U.K. (including biological oxygen demand, chloride, phosphorus and silica), to explore the impacts of urbanization, wastewater discharges and agricultural intensification. Over the last 40 years, improvements in wastewater treatment and agricultural management have reduced phosphorus loads by ~80%. However, this has been insufficient to curtail river algal blooms because nutrient concentrations remain above limiting levels. Over the last 50–60 years, rising water temperatures have increased the number of days with water temperatures favourable for diatom blooms in March–April and for cyanobacterial growth in July–August. These results highlight the challenges of eutrophication management in a warming climate and a strategic need to redouble efforts in further reducing nutrient emissions to control nuisance and increasingly harmful algal blooms.

Eutrophication and proliferation of nuisance and harmful algal blooms, linked to excess nutrients from agriculture and wastewater, are a cause of global water-quality impairment^{1,2}. Here, we provide a new perspective on historical eutrophication trajectories, by analysing a hitherto unpublished 150-year dataset of monthly water-quality measurements for the River Thames at Teddington (West London, U.K.): the world's longest continuous river water-quality record³. These data include: chloride (Cl⁻, a conservative tracer of wastewater) and biological oxygen demand (BOD, a measure of organic-matter contamination linked to water body hypoxia), measured since 1868; and two key nutrients: phosphorus (P), and silica (SiO₂, required for diatom growth), measured since 1936. Auxiliary data, including fertilizer P applications, population and a reconstructed daily water temperature record (see Methods), along with the historical nitrate record (which has already been published³), were combined to provide unique insights into the 150-year eutrophication history of this iconic river. Due to the U.K.'s early urbanization and agricultural intensification^{3,4}, the Thames eutrophication trajectory offers a template for understanding anthropogenic pressures on river systems: it exemplifies the multiple drivers that have converged to impair river water quality, and the challenges for remediation, that are representative of myriad world rivers subject to intensifying eutrophication pressures. This analysis is of particular importance and timeliness given the growing contemporary water quality crisis in U.K. rivers,

arising from concerns about serious pollution incidents from discharges of untreated sewage^{5–8}. This paper therefore also provides a new historical benchmark against which to assess current and evolving river water quality challenges in the U.K., and around the world, that increasingly threaten water security.

Results and discussion

Impacts of urbanization and agricultural intensification

Time series, showing trends in concentrations of Cl⁻, BOD and total reactive phosphorus (TRP, see “Methods”) for the River Thames at Teddington, relative to population growth, are displayed in Fig. 1; the origin and nature of this dataset are outlined in the “Methods” and Supplementary Notes. Between 1920 and 2020, the trend in Cl⁻ concentrations closely tracked population growth, reflecting urbanization and increasing wastewater discharges (Fig. 1a; Supplementary Fig. S1a). Municipal wastewater is enriched with Cl⁻ relative to river water, and is widely used as a conservative (non-reactive) tracer of wastewater in temperate climates such as in the U.K., where road salt for de-icing is a negligible fluvial Cl⁻ source^{9–11}. In the 1880s, Cl⁻ concentrations increased with increasing river flows (Fig. 2 and Supplementary Fig. S2a), reflecting nonpoint-source delivery of Cl⁻ in wastewater, from runoff during rainfall events; in the late 19th century municipal wastewater was disposed of by land application at ‘sewage farms’¹². From the

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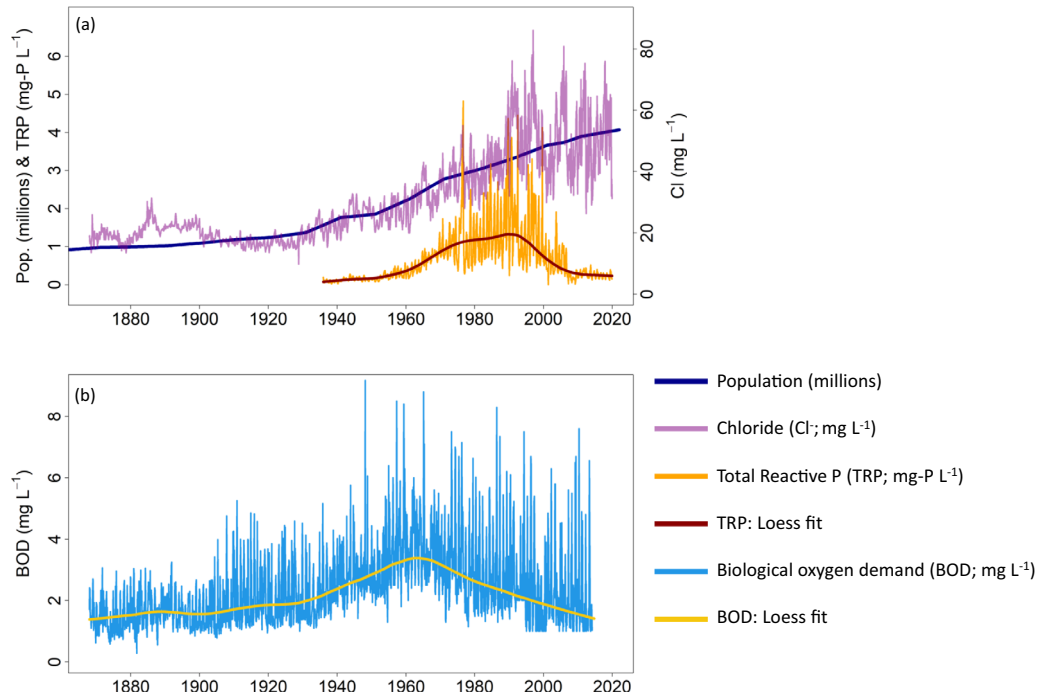


Fig. 1 | Timeseries of chloride, total reactive phosphorus and biological oxygen demand in the River Thames at Teddington, U.K. a Monthly chloride (Cl) concentrations (1868–2020) with the annual population of the Thames catchment superimposed, and monthly total reactive phosphorus (TRP) concentrations (1936–2020) with a Loess (locally estimated scatterplot smoothing) trend line; and **b** monthly biological oxygen demand (BOD) (1868–2020), with a Loess trend line.

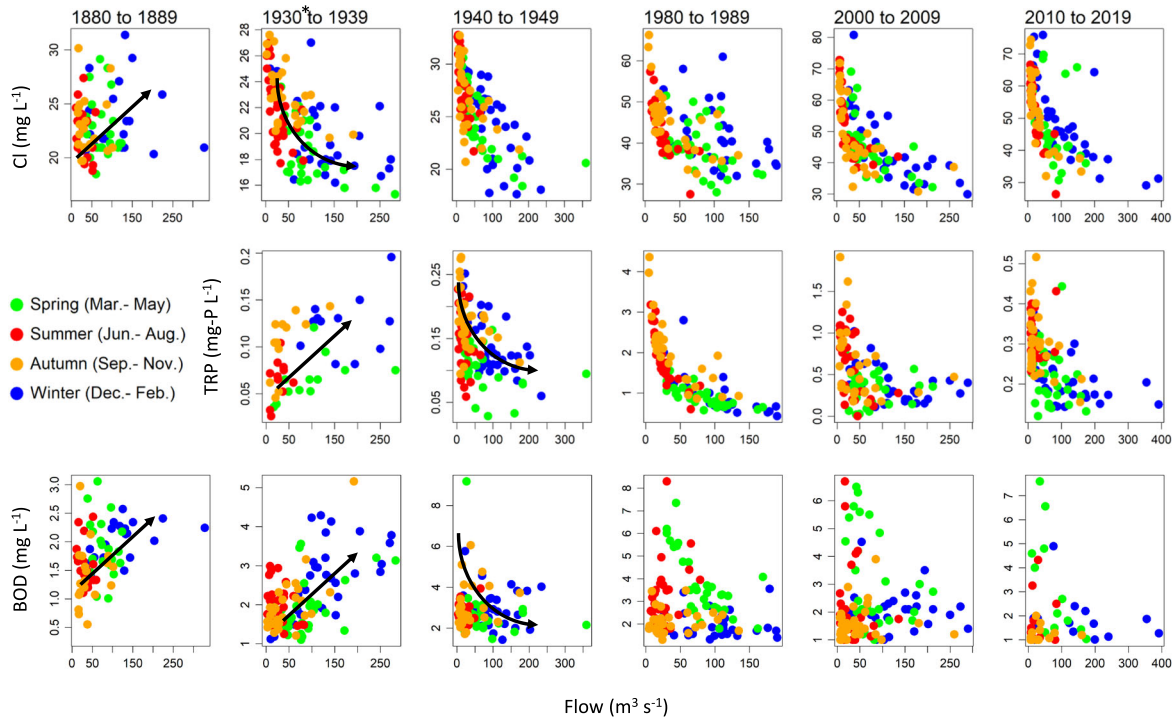


Fig. 2 | Flow-dependency of chloride, total reactive phosphorus and biological oxygen demand in the River Thames. Scatterplots showing the relationships with river flow ($\text{m}^3 \text{s}^{-1}$) for: chloride (Cl), total reactive phosphorus (TRP), and biological oxygen demand (BOD), for selected decades. Symbol colours denote the season of sampling. The black arrows denote changes in flow dependency: from increases in concentration with increasing flow, to dilution with increasing flow. *Note that TRP measurements began in 1936. A full series of decadal concentration vs flow scatterplots for Cl, TRP and BOD are provided in Supplementary Fig. S2.

1900s to 1920s, the Cl and flow relationship transitioned towards a dilution pattern with increasing flow (Supplementary Fig. S2a), corresponding with major shifts in wastewater management: the construction of wastewater treatment plants in the U.K. in the early decades of the 20th century that

started to provide primary and secondary treatment of wastewater from major urban areas¹². Final (treated) effluent from wastewater treatment plants is discharged semi-continuously as ‘point’ sources into the river, resulting in highest concentrations of Cl⁻ at lowest river flows and dilution

with increasing river flows¹¹. This dilution of Cl⁻ with increasing flow, that corresponds with a dominant point (wastewater) source of Cl⁻, has persisted since the 1920s (Fig. 2 and Supplementary Fig. S2a).

TRP measurements began in 1936, but it was not until the 1940s that the TRP-flow relationship switched to a dilution pattern, consistent with a change from nonpoint to a predominantly point-source signal of P from wastewater effluent (Fig. 2 and Supplementary Fig. S2b). The change from a positive to a negative relationship with flow for TRP therefore lagged behind that of Cl⁻, by around 10 years. This decadal lag in response indicates that, during the 1930s, legacy P stores^{13,14}, retained in soils and sediments, continued to be mobilised as nonpoint sources during rainfall events. Since the 1940s, TRP concentrations have persistently shown point-source dilution with flow (Fig. 2 and Supplementary Fig. S2b). TRP concentrations tracked population and Cl⁻ until 1990 (Fig. 1a and Supplementary Fig. S1b), reflecting the dominant control of wastewater on TRP concentrations in the River Thames^{15,16}. After 1990, the TRP and Cl⁻ timeseries diverged as a result of EU Urban Wastewater Treatment Directive¹⁵ mandating large-scale introduction of enhanced tertiary wastewater treatment and reducing P relative to Cl⁻ in final treated effluent (Fig. 1a and Supplementary Fig. S1b). As Cl⁻ is chemically conservative, river Cl⁻ concentrations were unaffected by the changes in wastewater treatment and continued to increase with population, whereas river TRP concentrations fell after 1990 in response to tertiary treatment reducing P discharged in the final effluent.

Median low-flow TRP concentrations (the 'low-flow' threshold was defined here as the 15th percentile of monthly flows) increased from 0.047 mg-P L⁻¹ (1930s) to 2.83 mg-P L⁻¹ (1980s); the largest low-flow TRP concentration increases occurred between the 1960s and 1980s (~1 mg-P L⁻¹ per decade), reflecting increasing wastewater P discharges (Fig. 3a). Although TRP concentrations closely tracked population growth and urban

wastewater inputs between 1936 and 1990 (Fig. 1a and Supplementary Figs. S1b, S2b), the imprint of agricultural intensification on P losses is also evident through changes in high-flow mobilisation of nonpoint P sources within the catchment (Fig. 3b). Median high-flow TRP concentrations (the 'high-flow' threshold was defined here as the 85th percentile of monthly flows) increased from 0.098 mg-P L⁻¹ (1930s) to 0.656 mg-P L⁻¹ (1980s). The greatest increases in high-flow TRP mobilisation occurred in the 1960s (Fig. 3b), reflecting a large-scale increase in nonpoint-source P mobilisation, in response to the build-up of soil P from fertilizer and manure applications⁴ (Fig. 4), along with increases in artificial subsurface (tile) drainage in the Thames catchment between 1950 and 1970¹⁷. This convergence of increases in nonpoint P source availability and enhanced hydrological connectivity through tile drainage¹⁸, likely accelerated the transport and delivery of P from agricultural land to the River Thames during high-flow events in the 1960s.

The greatest rate of increase in BOD occurred during periods of rapid population growth between 1930 and 1965, with BOD closely tracking Cl⁻ concentrations during this time (Fig. 1 and Supplementary Fig. S1c), reflecting contributions of organic matter contamination from urban wastewater. Similar to TRP, BOD concentrations increased with flow until the 1940s, before transitioning towards a point-source dilution pattern, with highest BOD concentrations during lower flows, particularly in spring and summer (Fig. 2 and Supplementary Fig. S2c).

A mixed picture of successes and challenges in water-quality management

The time series of BOD and Cl⁻ diverged after the early 1960s (Fig. 1 and Supplementary Fig. S1c): Cl⁻ concentrations continued to rise with increased urban wastewater discharges, whereas BOD declined, reflecting expansion in the use of activated sludge treatment in the 1960s that reduced organic

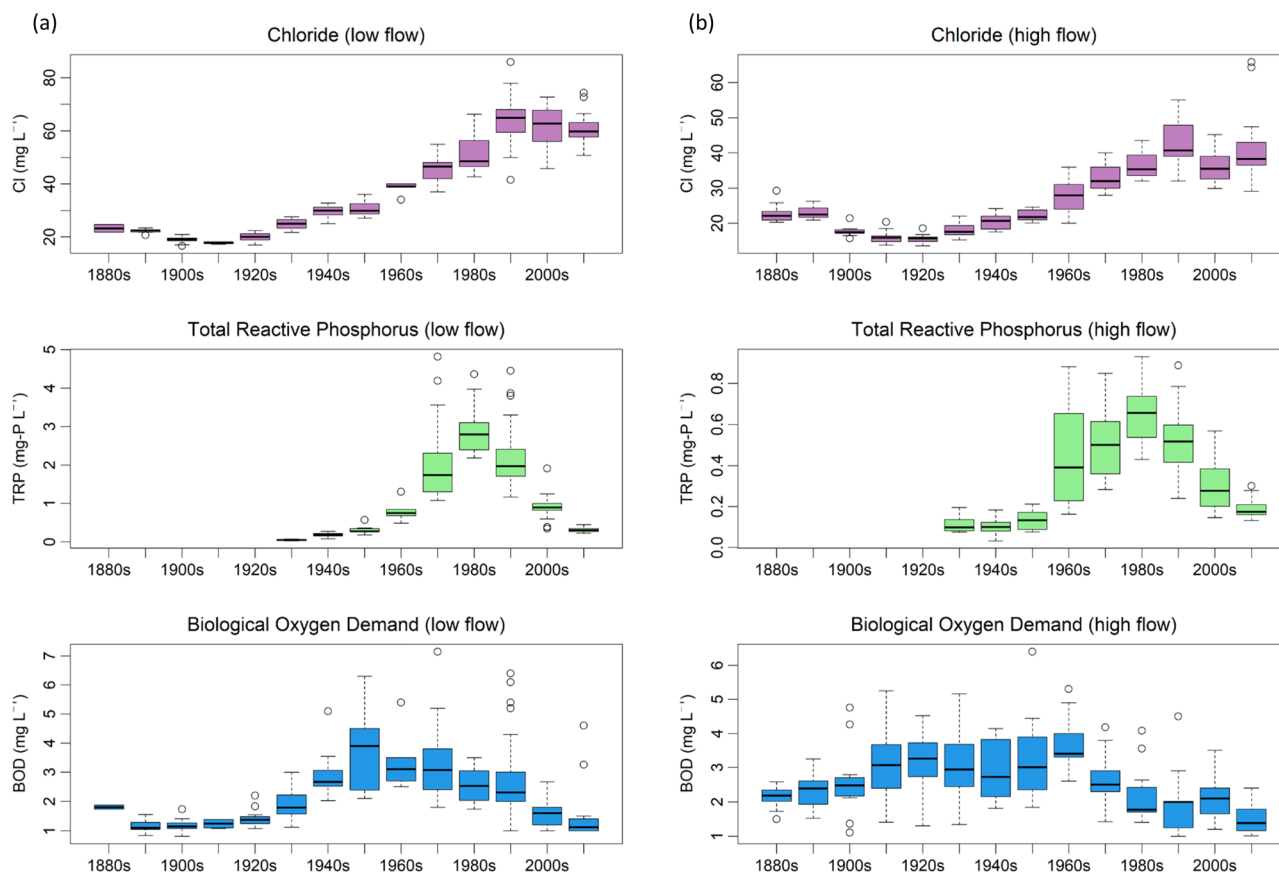


Fig. 3 | Low-flow and high-flow concentrations of chloride, total reactive phosphorus and biological oxygen demand in the River Thames from the 1880s to the 2010s. a Low-flow concentrations: here, the low-flow threshold was defined as the 15th percentile of monthly flows; and **b** high-flow concentrations: here, the high-

flow threshold was defined as the 85th percentile of monthly flows. Boxplot elements: centre line, median; box limits, upper and lower quartiles; whiskers, 1.5 x inter-quartile range; points, outliers.

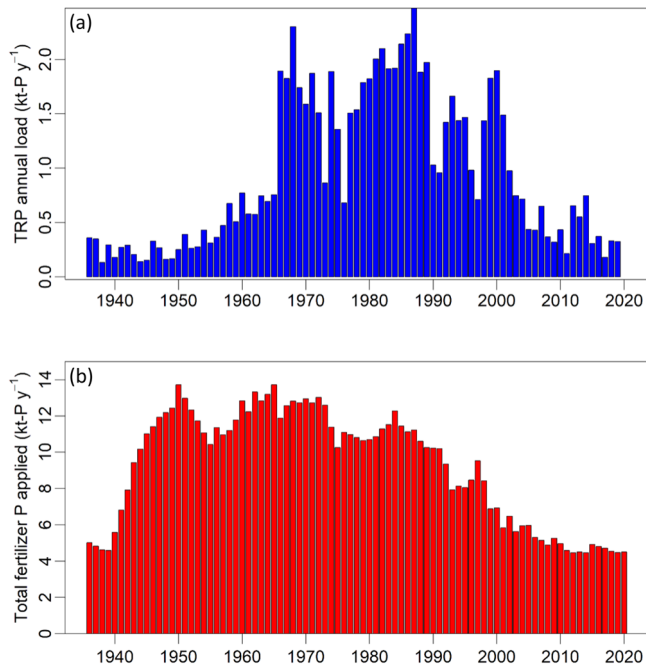


Fig. 4 | Total reactive phosphorus loads in the River Thames and total fertilizer phosphorus applied to agricultural land in the Thames catchment (1936 to 2020). **a** Annual total reactive phosphorus (TRP) loads (kt-P y^{-1}) for the River Thames at Teddington, U.K.; and **b** total fertilizer P (inorganic fertilizer P + manure P; kt-P y^{-1}) applied to agricultural land in the Thames catchment.

matter loading and enhanced nitrification of ammoniacal nitrogen to nitrate¹². Median low-flow BOD declined by 71%, from 3.90 mg L^{-1} (1950s) to 1.13 mg L^{-1} (2010s) (Fig. 3a). Despite these baseline BOD reductions, after the 1950s, the monthly time series (Fig. 1b) reveal an increase in high-BOD spikes above 5 mg L^{-1} (the BOD water-quality standard for high-alkalinity lowland rivers¹⁹), that typically occurred at lower flows in spring and summer (Fig. 2 and Supplementary Fig. 2c).

Median low-flow TRP concentrations declined by 89%, from 2.83 mg-P L^{-1} (1980s) to 0.307 mg-P L^{-1} (2010s) (Fig. 3a), corresponding with the adoption of the Urban Wastewater Treatment Directive in 1991 that required tertiary P removal at wastewater treatment plants serving >10 K population equivalents and discharging to sensitive freshwaters²⁰. Impacts of reduced fertilizer P applications (Fig. 4), along with agri-environment schemes to minimise soil erosion and reduce agricultural nonpoint-source P losses, resulted in reductions in high-flow TRP concentrations (Fig. 3b): median high-flow TRP concentrations declined by 73%, from the peak of 0.650 mg-P L^{-1} (1980s) to 0.174 mg-P L^{-1} (2010s).

The combined effects of these point- and nonpoint-source P mitigation measures in the Thames catchment since the 1980s resulted in an 82% reduction in median annual TRP loads: from 1.99 kt-P y^{-1} (1980s) to 0.353 kt-P y^{-1} (2010s) (Fig. 4). However, this remarkable reduction in TRP loads has had little impact on the within-river eutrophication risk: throughout the 2010s, the median river TRP concentration was 0.264 mg-P L^{-1} (range 0.120 to 0.516 mg-P L^{-1}), i.e., exceeding a threshold concentration of TRP (0.05 mg-P L^{-1}) below which nuisance algal growth in U.K. lowland rivers is expected to become P-limited^{21,22} and exceeding a TRP standard of 0.098 mg-P L^{-1} to achieve ‘Good Ecological Status’ in the River Thames^{19,23}.

Throughout the 150-year record, nitrate concentrations have also remained well above an ecologically-limiting threshold of ~0.4 mg-N L^{-1} ^{121,22}; median nitrate concentrations ranged from 2.13 mg-N L^{-1} (1870s) to 7.82 mg-N L^{-1} (2010s; Supplementary Fig. S3a). Since the 1980s, these high nitrate concentrations have been sustained by groundwater legacy nitrate sources^{3,24} which cannot easily be abated and are expected to persist in the coming decades^{3,24,25}. This persistent overabundance of both P and N (high

ambient concentrations above limiting levels) continues to sustain river algal blooms and contributes to the ongoing water quality and ecological impairment within the River Thames.

The Redfield ratio²⁶ (16 N:1 P) is widely used as a reference ‘optimum’ ratio for primary production across diverse aquatic ecosystems, including algae in streams and rivers^{22,27}, and deviations from the Redfield ratio can indicate potential for nutrient limitation in rivers^{22,28,29}. Molar N:P ratios during the period of highest P concentrations, from the 1970s to early 1990s, were close to the Redfield ratio (median molar N:P ratio in the 1970s was 14.5) (Supplementary Fig. S3b). Since the mid-1990s, River Thames N:P ratios have exceeded the Redfield ratio (median molar N:P in the 2010s was 53), and this indicates that reducing P inputs to achieve lower, ecologically-limiting concentrations may offer the greatest opportunity to curtail algal growth and reduce river eutrophication pressures. However, there remain concerns about the impacts of nitrate on eutrophication in downstream receiving water bodies, including lakes and, in the case of the River Thames, estuarine and coastal waters, where there can be N-limitation or dual N and P co-limitation^{30,31}. Reducing phosphorus inputs can potentially also impact the export of reactive nitrogen from lakes³². These concerns provide a strong rationale for dual reductions of N as well as P to manage eutrophication in downstream receiving water bodies^{31,33}.

Blooming algae: diatom growth and silica depletion

Diatoms are the dominant phytoplankton in the River Thames and give rise to pronounced spring algal blooms when river-water chlorophyll-a reaches maximum concentrations³⁴. During these diatom blooms, dissolved reactive silica (SiO₂) is assimilated to form the diatom outer shells (siliceous frustules), resulting in depletion in river-water SiO₂ concentrations³⁵, and a negative relationship between chlorophyll-a and SiO₂ concentrations (Fig. 5a). Chlorophyll-a concentrations have only been measured since the 1970s in the River Thames at Teddington, at varying sampling frequency and with data gaps (see Supplementary Fig. S5), whereas SiO₂ has been measured continuously, on a monthly basis, in the Thames since 1936 (Fig. 5b); thus, SiO₂ depletion is used here as a long-term indicator (proxy) of diatom blooms³⁶. Here, a diatom ‘bloom’ was defined as SiO₂ depletion to concentrations below 4.1 mg L^{-1} ; this corresponds with the 100 $\mu g L^{-1}$ chlorophyll-a concentration threshold that indicates the onset of algal blooms in the lower River Thames³⁷ (Fig. 5a).

Phytoplankton blooms are highly dynamic, responding to changes in physio-chemical conditions on daily and sub-daily timescales³⁴ and monthly monitoring is insufficient to characterize these dynamics. Recent higher-resolution monitoring has shown that, with an overabundance of P and N in the River Thames, water temperature during the spring is a key driver of diatom blooms in the River Thames^{34,38,39}, among other multiple controls on bloom dynamics, including light⁴⁰. In many rivers, flow and flushing rate also provides a key control on phytoplankton blooms; however, flow in the lower River Thames is highly regulated by locks^{41–43}. Moreover, while there is large interannual variability in river flow, analyses have shown little evidence for any long-term trends in river flow, floods or droughts related to climate change since 1880^{41,44}. Here, we focus on the potential impacts of changes in water temperature on algal blooms. There was a pronounced range in water temperature when spring diatom blooms occurred (i.e., when SiO₂ concentrations were depleted below 4.1 mg L^{-1} during March through June), as shown in Fig. 5c as a bivariate kernel density (2D probability) distribution⁴⁵. From this, we identified an ecological range in water temperature of 9.6 to 18.25 °C that is favourable for spring diatom blooms in the lower River Thames at Teddington. This range is consistent with estimates of the ecological range in water temperatures favourable for diatom blooms at other locations within the River Thames^{34,40}.

Warming waters: increasing the potential for nuisance and harmful algal blooms

Water and air temperature datasets were used to reconstruct a daily river water temperature record for the Thames at Teddington from 1870 to 2020 (see ‘Methods’). The daily river water temperatures were then used to

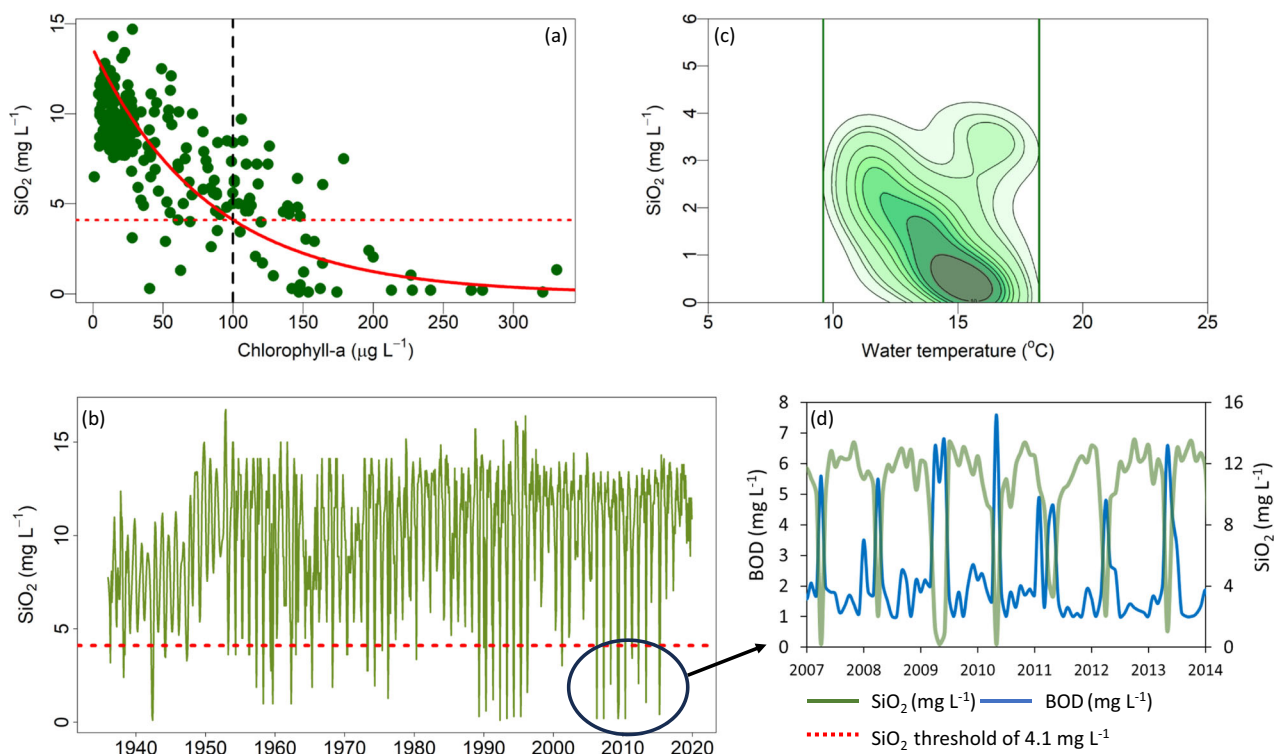


Fig. 5 | Silica, chlorophyll-a, water temperature and biological oxygen demand to explore diatom blooms. **a** Relationship between dissolved reactive silica (SiO_2) and chlorophyll-a in the River Thames (1974–2020) showing the threshold SiO_2 concentration (dashed red line, 4.1 mg L^{-1}) that corresponds with the $100 \mu\text{g L}^{-1}$ chlorophyll-a water-quality threshold (dashed black line) for algal blooms in the lower River Thames³⁷; **b** time series of SiO_2 from 1936 to 2020, showing the 4.1 mg L^{-1} SiO_2 threshold as a red dashed line; **c** bivariate kernel density plot⁴⁵ showing the probability distributions for the relationship between SiO_2 and water

temperature during diatom blooms, for samples collected during spring (March through June) when SiO_2 concentrations were depleted below the 4.1 mg L^{-1} threshold. The vertical lines denote the ecological temperature range favourable for diatom blooms ($9.6\text{--}18.25 \text{ }^\circ\text{C}$); i.e., the upper and lower threshold water temperatures bounding the ‘window’ of diatom blooms (90% probability of SiO_2 depletion below 4.1 mg L^{-1}); **d** timeseries of SiO_2 (green) and biological oxygen demand (BOD; blue) from 2007 to 2014, showing the synchronicity between silica depletion (diatom blooms) and spikes in BOD.

calculate the number of days, per month, when the water temperature was favourable for diatom blooms, i.e., within the range $9.6\text{--}18.25 \text{ }^\circ\text{C}$ (Fig. 6a). Seasonal phytoplankton succession in the River Thames follows a similar pattern to temperate eutrophic lakes: from dominance of nuisance diatoms in spring, with growth in potentially harmful cyanobacterial groups in late summer⁴⁶. A similar analysis was then undertaken to assess the potential for growth of cyanobacteria that dominate at higher water temperatures ($>20 \text{ }^\circ\text{C}$)^{47,48} (Fig. 6b).

The results show that, over the last 50–60 years, warming water temperatures in the River Thames have resulted in conditions more favourable to diatom blooms in March and April, and more favourable for cyanobacteria growth in July and August. Over the last 50 years, the number of days from March 1st through April 30th with water temperatures favourable for diatom blooms has increased by ~ 15 days: from a median of 20 days in the 1970s to a median of 35 days in the 2010s (Fig. 6a). During this time, the most rapid increase in the number of days in March and April with favourable water temperatures for diatom blooms occurred between the 1980s and 1990s: an increase of ~ 12 days, from a median of 21 days in the 1980s to a median of 33 days in the 1990s (Fig. 6a). There was no corresponding change in the number of days with favourable water temperatures for diatom blooms in May. However, rising water temperatures in June are increasingly exceeding $18.25 \text{ }^\circ\text{C}$ and therefore becoming too warm for diatom blooms: the number of favourable water temperature days for diatom blooms in June has declined by ~ 8 days: from a median of 24 days in the 1970s to a median of 16 days by the 2010s (Supplementary Fig. S4). Over the last 60 years, the median number of days from July 1st through August 31st with favourable water temperatures for cyanobacterial blooms has risen by an average of ~ 22 days: from a median of 9 days in the 1960s to a median of 31 days in the 2010s (Fig. 6b).

Reductions in phosphorus loads but not in algal growth potential

A period of high magnitude and frequency of SiO_2 depletion below 4.1 mg L^{-1} , indicative of intense diatom blooms, occurred between 2005 and 2016 (Fig. 5b and Supplementary Fig. S5), despite TRP concentrations having been reduced to levels not seen since the 1950s (Fig. 1a). The BOD time series provides further independent corroborating evidence of persistent eutrophication risk during this time (Fig. 1b): despite the improvements in wastewater treatment addressing gross organic pollution from wastewater and reducing P concentrations, the incidence of high BOD spikes ($>5 \text{ mg L}^{-1}$) increased after the 1950s. Further, there has been a marked shift in the seasonality and hydrological distribution of these BOD spikes. Prior to 1950, the highest BOD concentrations occurred during high flows, particularly in winter, indicating runoff-driven mobilization of organic matter contamination (Fig. 2, Supplementary Fig. S2c). Since the 1950s and 1960s, the highest BOD concentrations have shifted to low-flows in spring and summer (Fig. 2, S2c). These high-BOD spikes now correspond directly with periods of SiO_2 depletion (Fig. 5d). This indicates that the BOD spikes may now be primarily linked to microbial degradation of diatom blooms, and that these blooms are a continuing driver of oxygen depletion in the River Thames.

Consequently, despite the remarkable successes in reducing P loads in the River Thames, owing to substantial investments in reducing wastewater and agricultural P emissions over the last 40 years⁴⁹ (Figs. 1a, 4a), there have been no corresponding reductions in algal growth potential, as demonstrated by the SiO_2 record, which indicates a persistence of spring diatom blooms (Fig. 5b, d). The reductions in nutrient emissions have not been sufficient to reduce river nutrient concentrations to limiting levels and, over the last 50–60 years, there has been an increase in the number of days with water temperatures favourable for diatom blooms in March and April, and an increase in number of days favourable for cyanobacteria blooms in July and August.

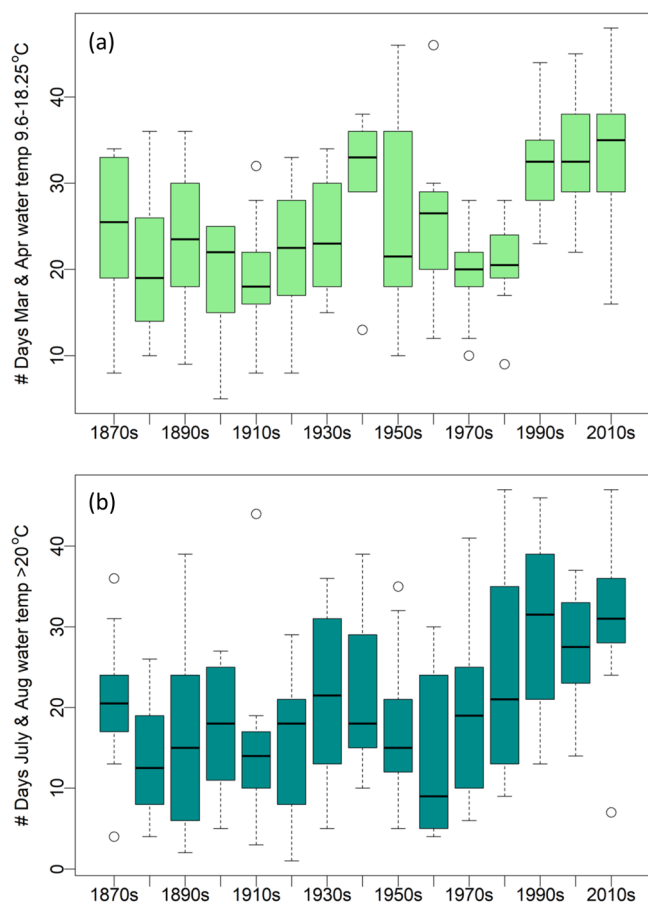


Fig. 6 | Number of days with favourable river water temperatures for diatom and cyanobacteria blooms from the 1870s to the 2010s. Boxplots showing (a) the number of days from March 1st through April 30th, when water temperatures in the River Thames were within the ecological range favourable for diatom blooms (9.6–18.25 °C); and b the number of days from July 1st through August 31st, when water temperatures were >20 °C (i.e., favourable for cyanobacteria growth)^{47,48}. These graphs are based on the 150-year reconstructed water temperature timeseries for the River Thames (see “Methods”). Boxplot elements: centre line, median; box limits, upper and lower quartiles; whiskers, 1.5 x interquartile range; points, outliers.

Conclusions

The world’s longest continuous river water quality record provides unique insights into the history of eutrophication, by documenting the impacts of 150 years of social, economic and environmental change through the lens of river-water chemistry and quality. Although remarkable successes in reducing P loads in the lower River Thames (by ~80%) have been achieved through the combined effects of both point and nonpoint P source management, these management measures have not yet reduced P concentrations to limiting levels that would curtail phytoplankton blooms. Nonetheless, owing to the depletion of P relative to N, the continuing multi-decadal legacy of N from groundwater in the Thames catchment^{3,24} and, given the increasing nutrient pressures linked to climate change⁵⁰, reducing P concentrations to limiting levels continues to offer the best long-term prospect of curtailing river algal blooms in the Thames. However, dual P and N controls may be needed to reduce eutrophication in downstream estuarine and coastal waters^{30,31}. And, although planting of riparian trees has been proposed as a means of reducing algal growth in rivers through shading^{51,52}, such opportunities are more limited in large lowland rivers like the Thames, owing to the large channel width relative to the marginal zone of shading cover.

Achieving the reductions in P loads and concentrations needed to control river algal blooms will be challenging, especially given the legacy of P that has accumulated over the last century^{41,53}. More than 40% of U.K. soils now have soil P levels above the agronomic optimum as a result of long-term

application of mineral P fertilisers and P-rich animal manures and slurries, increasing the risks of nonpoint source P losses⁵⁴. In areas of high P-loss risk, improvements in nutrient management⁵⁵ along with carefully-targeted agricultural conservation practices^{56,57} will be needed to minimise nonpoint source P losses. However, further reducing wastewater effluent P emissions will be also pivotal in lowering ambient P concentrations and reducing eutrophication risk in the River Thames. Wastewater effluent discharges remain responsible for widespread chronic P impairment of water quality in U.K. lowland rivers^{15,20}. As shown in this paper, wastewater effluent continues to contribute the highest P concentrations during the lowest flows, especially in summer in the River Thames, a period of higher risk for cyanobacterial blooms. Although there have been dramatic improvements in baseline P concentrations (since the 1980s) and BOD (since the 1960s), as a result of improvements in wastewater treatment, there is now increasing concern about pollution incidents associated with discharges of untreated sewage to U.K. rivers, many of which are currently unreported^{7,58,59}. These untreated sewage discharges occur not only during rainfall events (from combined sewer overflows and where the capacity of the treatment works is exceeded), but also increasingly during drier weather conditions due to infrastructure failures^{6,7}. These pollution incidents have been attributed to the converging effects of long-term underinvestment in wastewater infrastructure, and expansion in housing and commercial developments, putting pressure on already underperforming wastewater treatment plants, along with greater intensity of rainfall events^{5,59}. The ecological and human health impacts are of growing public concern^{59,60}. The long-term monthly water-quality data presented here are not of sufficient temporal resolution to adequately capture individual sewage pollution incidents that may occur over the duration of a few hours to days, but these data do demonstrate the long-term chronic and cumulative effects of wastewater discharges on water-quality impairment at the catchment scale. The effects of poorly treated wastewater will undoubtedly contribute to the challenges in reducing P concentrations in the River Thames to levels that could limit algal blooms.

Some of the largest diatom blooms (using silica depletion as a proxy) have occurred since 2005, despite river P loads during this time being ~80% lower than at their peak in the mid-1980s. In spite of these dramatic P load reductions, river P concentrations remain above limiting levels. There is a continuing overabundance of P (and N) to support algal blooms and, over the last 50–60 years, rising water temperatures in the River Thames have resulted in an increasing number of days with water temperatures favourable for diatom blooms in March and April and for growth of cyanobacteria in July and August. The lessons from this remarkable 150-year record of river water quality measurements resonate globally: as climate change intensifies, further tightening nutrient emissions will likely be our most effective lever to control the nuisance and harmful algal blooms that increasingly threaten our water quality and water security.

Methods

Total P fertilizer and manure P application data for the Thames catchment

Applications of P (as inorganic fertilizers and livestock manure) to agricultural land within the Thames catchment were calculated on an annual basis, from 1936 to 2020, using the methods of Worrall et al.⁶¹. Annual agricultural census returns⁶² were compiled for each parish within the Thames catchment from 1936 to 1989. In 1989, the U.K. government moved to annual, national-scale reporting with reporting for supra-parish units in 1990, 1995 and 1999. From 2000 to present, there was annual reporting, but only for supra-parish units. Data from parishes and supra-parish units were then combined to derive land use and livestock headage numbers within the Thames catchment. Manure P applications were calculated from the livestock headage data, according to the methods of Johnes et al.⁶³. The inorganic P fertilizer inputs were estimated using the agricultural land use data and fertilizer use data. The Fertiliser Manufacturers Association and the Environment Agency of England and Wales have published annual surveys of the use of inorganic fertilizers since 1962⁶⁴. To convert national-scale annual total P fertilizer data into inputs for the Thames catchment, for each year, the

recommended rates of P application from the U.K. Fertiliser Best Practice manual were used to scale the total annual P fertilizer use to the average that would be applied for each land-use type, for each year, in the Thames catchment. For the years prior to 1962, inorganic P fertilizer inputs were estimated using data from Mattikalli and Richards⁶⁵. Data were reported for 'arable' and 'grassland' in 1943, 1950, 1957 and 1962⁶⁵, and linear interpolation was used to estimate values for the intermediate years. Before 1943, fertilizer inputs were estimated by linear interpolation, decreasing backward through time until they equalled a value expected from organic manures based on evidence from export coefficient models⁶⁶.

Population data

Population census returns were available for every English county for every decade from 1841, with additional projected numbers from 2001 to 2020⁶⁷. The population of the Thames catchment was estimated as a weighted proportion of the area of each county within the catchment. Linear interpolation between census years was used to estimate the Thames catchment population in each year of the study.

River flow data

Mean daily flow records were downloaded from the U.K. National River Flow Archive, for the Thames at Kingston (gauging station number 39001)⁶⁸, which is the lowest gauging station on the River Thames, and less than 3 km upstream from the Teddington water quality monitoring site. Flow data at this gauging station are available from 1883 to the present day.

Meteorological and river water temperature data

Daily rainfall and air temperature records have been maintained at Oxford since the 18th century^{69,70}. Mean daily air temperature data were combined with water temperature data (available from 1974 from the U.K. Harmonized Monitoring Scheme database⁷¹ and the Environment Agency of England and Wales Water Quality Data Archive⁷²; see water quality data below), to derive seasonal regression relationships between water temperature and air temperature. These regression relationships were then applied to the daily air temperature data from 1868, to derive a daily water temperature record for the River Thames from 1868 to 2020.

Water quality data

The following historical water quality datasets were compiled:

- Chloride (Cl; mg L⁻¹): a continuous monthly record from 1868 to 2020;
- Biological oxygen demand (BOD; mg L⁻¹): a continuous monthly record from 1868 to 2014;
- Total reactive phosphorus (TRP; mg L⁻¹, a measure of molybdate-reactive P on an unfiltered water sample⁷³): a continuous monthly record from 1936 to 2020;
- Silica (SiO₂; mg L⁻¹): a continuous monthly record from 1936 to 2020.

The water samples were all collected at the same point on the River Thames but different methods of analyses were used at different times and by different organisations:

- Grand Junction Water Company, 1868–1901;
- London Metropolitan Water Board, 1902–1977;
- Thames Water Authority, 1978–1988;
- National Rivers Authority, 1989–1996;
- Environment Agency of England and Wales, 1996 to 2020.

The datasets prior to 1974 were transcribed from paper records. Hamlin⁷⁴ provides a guide to the historical analytical water chemistry techniques used, and the first verified water quality measurements were in 1867–1868 for the River Thames. A summary of the analytical methods used for each water quality analyte are provided in Supplementary Tables S1–S4. Datasets from 1974 to 2013 were accessed from the U.K. Harmonized Monitoring Scheme database⁷¹ and the datasets after 2013 were accessed from the Environment Agency of England and Wales Water Quality Data Archive⁷². Throughout the period of water quality records, samples of river

water were measured several times a week; however, data were reported differently and raw data were not always available. Between November 1881 and December 1938 measurements were made several times a week and these values were averaged on a monthly time step. Between January 1939 and December 1952 the data were given as quarterly averages and so to estimate monthly data, a cubic spline was fitted to the data for the subsequent period and then used to predict the monthly data given the spline fitted to the quarterly summaries: actual sampling was still each weekday. From January 1953 to March 1974 measurements were made on weekdays and again summarised on a monthly basis. After March 1974, the Thames at Teddington was included in the Harmonised Monitoring Scheme (HMS)^{75,76} and, as such, sampling came into line with national reporting standards at a monthly time step.

The analytical methods for harmonising water-quality data, for the HMS sites was prescribed in DoE⁷⁷ and outlined in Simpson⁷⁵. Current procedures are outlined and controlled by the U.K. Government's Standing Committee of Analysts⁷⁸. Working back from the most recent HMS water-quality records, periods of overlap between historical methods for individual analytes were used for baseline data harmonization and correction.

Data presentation and analysis

The R software environment for statistical computing and graphics⁷⁹ was used for data presentation and analysis. Locally estimated scatterplot smoothing (LOESS) was undertaken in R using the `loess()` (Local Polynomial Regression Fitting) function, and bivariate kernel density plots were created using the 'ks' package⁴⁵.

Data availability

The data presented in this paper are available at the University of Bristol data repository, data.bris, at: <https://doi.org/10.5523/bris.3lgkzhzn24qs2qp65dimo8tan>.

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Author contributions

H.P.J.: Conceptualization; formal data analysis; writing the original manuscript; N.J.K.H.: Conceptualization; transcribing of archival material and provision of historical water quality data records; and contributions to writing the manuscript; F.W.: Conceptualization, collation of historical land-use data and provision of the total fertilizer and manure phosphorus applications dataset; and contributions to writing the manuscript; T.P.B.: Conceptualization; collation of historical meteorological data and river water temperature records; and contributions to writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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