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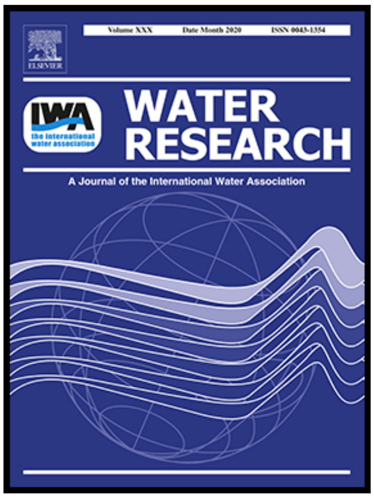

Eutrophication exacerbated organic pollution in lakes across China during the 1980s-2010s

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Highlights

- Lake organic pollution across China during the 1980s-2010s was investigated.
- Organic pollution levels of lakes were high in the north and low in the south.
- Eutrophication and salinization were the key factors varying organic pollution.
- Eutrophication exacerbated lake organic pollution during the 1980s-2010s.
- Lake expansion had dilution effects on organic pollution in saline lakes.

Eutrophication exacerbated organic pollution in lakes across China during the 1980s-2010s

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Abstract: Lakes are vital sources of drinking water and essential habitats for humans and various other living organisms. However, many lakes face organic pollution due to anthropogenic disturbance and climatic influence, and the spatiotemporal changes of organic pollution in lakes over a large area are still unclear. Based on three monitoring datasets of chemical oxygen demand (COD) in 390 lakes, this study demonstrated the apparent spatiotemporal differences of organic pollution in lakes during the 1980s-2010s and the effects of water eutrophication and salinization. Throughout China, lake organic pollution showed a general spatial trend of being more severe in the north compared to the south. This pattern is reflected in the positive linear correlations between *in-situ* COD concentrations and lake latitude, observed in both the 1980s ($p < 0.05$) and the 2010s ($p < 0.01$). In terms of spatial differences, the influence of total nitrogen concentrations increased from 0.27% in the 1980s to 35.24% in the 2010s. Moreover, with increasing human activity, 78.31% of the studied lakes ($N = 83$) showed increasing COD concentrations during the 1980s-2010s. In addition, the logarithmic dissolved organic carbon concentrations were linearly correlated with log water conductivities (Pearson's $r = 0.49$, $p < 0.01$), suggesting that lake expansion would attenuate organic pollution in saline lakes through dilution effects. These results are valuable for understanding the spatiotemporal dynamics of organic pollution and are crucial for effective management of organic pollution in different lakes.

Keywords: Organic pollution; Chemical oxygen demand; Spatiotemporal changes; Water eutrophication; Lake expansion

1 Introduction

Lakes play an essential role in supporting drinking water for human society, providing habitats for plants and animals, and reducing water pollutants through biogeochemical processes (NIGLAS, 2019; Zhang et al., 2023). However, due to anthropogenic disturbance and climate change, many lakes are facing organic pollution that degrades water quality, affects the health of species in the food chain, and threatens the safety of lake ecosystems (Gullian-Klanian et al., 2021; Nosrati et al., 2012). Thus, grasping the spatiotemporal dynamics of lake organic pollution is essential for maintaining the health of lake ecosystems, securing water resources, and guiding effective pollution control and management strategies for lakes.

The commonly used metrics of organic pollution in lakes are chemical oxygen demand (COD) and total organic carbon (TOC). Of these, COD determines organic pollution levels using the amount of oxidant required to break down organic matter, and potassium permanganate is often used as the oxidant (Laszakovits et al., 2020). The International Organization for Standardization recommends the use of COD to measure organic matter concentrations, and the Chinese government stipulates that good lake waters should have COD concentrations of < 6.0 mg/L (MEEC, 2002). Being different from COD, TOC concentrations determine organic pollution levels using the weight of organic carbon atoms per unit volume, which is measured by converting organic matter into carbon dioxide (Artifon et al., 2019). In comparison, TOC can better reflect the total organic matter content, but has a higher measurement cost (Knap et al., 1996); in addition, COD concentration is often linearly correlated with TOC concentration (Jiao et al., 2021). As a result, COD has become one of the standard indicators widely used to indicate organic pollution in lakes and as a surrogate for TOC concentration (Han and Ma, 2015; Jiao et al., 2021; MEEC, 2002).

Organic pollution in lakes is regulated by several anthropogenic and natural factors. Human activity not only introduces anthropogenic organic byproducts into lakes, but also elevates water eutrophication levels and increase autochthonous organic matter from algal production (Artifon et al., 2019; Baines and Pace, 1991). For lakes in the Changjiang Delta, China, the amount of dissolved organic matter is strongly determined by eutrophication levels in the lakes and the percentages of artificial surface areas in the lake basins (Liu et al., 2020a). Similarly, natural factors influence organic pollution in lakes by transporting terrigenous organic matter into lakes and influencing the production and transformations of organic matter in lakes (Nosrati et al., 2012). In addition, natural factors also influence organic pollution in lakes by altering water evapoconcentration and dilution effects (Butturini et al., 2022; Nosrati et al., 2012). Due to water evapoconcentration, saline lakes in China (30.0 mg/L) usually have higher dissolved organic carbon (DOC) concentrations than freshwater lakes (8.1 mg/L) (Song et al., 2018a). In contrast, precipitation has low DOC concentrations and has been reported to dilute organic pollution in Chinese lakes (Liu et al., 2021a; Safieddine and Heald, 2017). Furthermore, due to the spatial variability of various factors, we need to know the organic pollution in different lakes over a large area.

Over the past decades, global lakes have been changing under the pressure of increasing human activity and climate warming. Algal blooms have become more common in lakes, with 8.8% of global lakes experiencing an increase in bloom risk between the 1980s and 2010s (Ho et al., 2019). In addition, the water volume of many lakes (79.38%) has also increased, and one of the five hotspots for volume increase is located on the Tibetan Plateau (Luo et al., 2022). Along with changes in algal blooms and water volumes, satellite-based monitoring of eutrophic lakes along the Changjiang River in China has shown that both DOC and particulate organic carbon (POC) concentrations fluctuate obviously over different seasons (Liu et al., 2023, 2021b). In these cases, organic pollution in lakes should vary temporally in the context of global change, and understanding their long-term changes can provide valuable insights for improving lake water quality.

Across the vast territory of China, there are thousands of lakes influenced by various anthropogenic and/or natural factors (NIGLAS, 2019); moreover, many lakes show obvious changes in various influencing factors during the 1980s-2010s (Zhang et al., 2019). Therefore, here we attempt to investigate the spatiotemporal changes and underlying forces influencing the organic pollution of lakes across China during the 1980s-2010s. We have three objectives: (1) to reveal the spatial pattern of organic pollution in Chinese lakes; (2) to identify the changes in organic pollution during the 1980s-2010s; (3) to assess the effects of different driving forces. Due to the lack of TOC concentration in the 1980s, we used COD to indicate the level of organic pollution. This study is very valuable for understanding and controlling organic pollution in different lakes in the context of a changing world.

2 Materials and Methods

2.1 Research area

China contains 2,693 lakes that are larger than 1 km^2 in size (NIGLAS, 2019), and we focus on 390 Chinese lakes where measurements were taken (Fig. 1). The lakes studied cover a total area of 65,850 km^2, representing 88.5% of the total area of China's lakes (NIGLAS, 2019). Biogeochemical processes in lakes are strongly influenced by environmental characteristics, which present great spatial differences across China. Comparatively, population density is high in the east; altitude indicated by digital elevation model (DEM) is higher in the west; precipitation and vegetation coverage are high in the southwest; and cropland is mainly distributed in the northeast (Liu et al., 2020). According to the topographic and ecological conditions, China is divided into five limnetic zones: the Inner Mongolia-Xinjiang Lake zone (IMXL); the Tibetan Plateau Lake zone (TPL); the Yunnan-Guizhou Plateau Lake zone (YGPL); the Northeast Plain and Mountain Lake zone (NPML); and the Eastern Plain Lake zone (EPL) (NIGLAS, 2019). During the 1980s-2010s period, precipitation and glacier melting led to lake expansion in the TPL (Zhang et al., 2019), while anthropogenic pollution led to lake eutrophication in the EPL (NIGLAS, 2019). Therefore, Chinese lakes are ideal areas to study changes in organic pollution in a changing world.

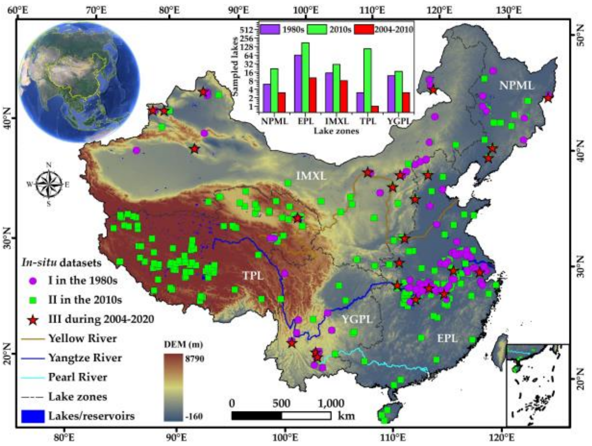

Fig. 1. The sampled lakes in the 1980s (N = 102), in the 2010s (N = 346), and during 2004-2020 (N = 25). The inset histogram shows the number of sampled lakes in the five lake zones.

2.2 *In-situ* data collection

2.2.1 Field investigation

This study used three datasets of *in-situ* measurements acquired via field sampling or automatic monitoring. Three *in-situ* datasets are included (Fig. 1, (Supplementary Table S1). ① Dataset I contains lake-based mean COD, total nitrogen (TN), total phosphorus (TP), chlorophyll-a (Chl-a) concentrations, conductivities, and water transparency as indicated by Secchi-disk depth (SDD) for 102 lakes, calculated by averaging three measurements obtained from the National Lake Survey in the 1980s. ② Dataset II contains COD, TN, TP, Chl-a, DOC concentrations, excitation-emission matrix fluorescence spectra of colored dissolved organic matter (CDOM-EEMs), conductivity, and SDD values at 1795 stations for 346 lakes in the 2010s (Dataset II), which were collected in this study. ③ Dataset III contains time series of COD concentrations for 25 lakes during 2004-2020. Among them, weekly COD values at representative stations for 18 lakes during 2004-2020 were automatically monitored by the Ministry of Ecology and

Environment, China (MEEC, Supplementary Table S3). In addition, annual mean COD concentrations for seven lakes were obtained from the literature.

When doing the field samplings, water conductivities were measured with a *YSI* multi-parameter probe; SDD was recorded with a Secchi disk on the sun-shielded side of the sampling vessel; and surface waters (0 – 30 cm) were synchronously collected to measure COD, TN, and TP concentrations (NIGLAS, 2019; Wang and Dou, 1998). In addition, surface waters were also filtrated through filter membranes with pore sizes of 0.7 mm (*Whatman* GF/F filter), 0.45 mm (pre-combusted GF/F filter), and 0.22 mm (*Millipore* polycarbonate filter); then, the filter membranes and liquids were collected as Chl-a and DOC/CDOM samples (Knap et al., 1996; Mueller et al., 2003). COD, TN, TP, and Chl-a samples were stored frozen at -18°C, while DOC and CDOM samples were kept refrigerated at 4°C.

2.2.2 Laboratory measurement

Upon return to the laboratory, all samples were measured immediately. COD levels were determined by the potassium permanganate boiling method (APHA, 1995). TN and TP concentrations were measured through spectrophotometric analysis (Mueller et al., 2003). Chl-a concentrations were measured via the spectrophotometric technique (Mueller et al., 2003). DOC concentrations were quantified through the high-temperature combustion method (Knap et al., 1996). CDOM-EEMs were obtained using a *Hitachi* F-7000 spectrometer with excitation wavelengths ranging from 200 to 450 nm (5-nm increments) and emission wavelengths ranging from 250 to 600 nm (1-nm increments). Then, the excitation and emission spectra were calibrated according to the *Hitachi Corporation's* guidelines (Murphy et al., 2010).

2.2.3 PARAFAC analysis of CDOM-EEMs

CDOM-EEMs offer a comprehensive insight into the sources and composition of dissolved organic matter, but the signals of different components are superimposed. To identify the composition of dissolved organic matter, parallel factor analysis (PARAFAC) has been utilized to decompose CDOM-EEMs into distinct components (Murphy et al., 2013). After excluding 29 samples with measurement errors identified through an intercomparison of all corrected CDOM-EEMs (N = 593, Section 2.2.2), a four-component model was developed and validated using split-half and random initialization analyses with the drEEM toolbox. The four CDOM components were matched for their spectral characteristics by referencing the online OpenFluor database, a repository of fluorescence spectra (https://openfluor.lablicate.com/).

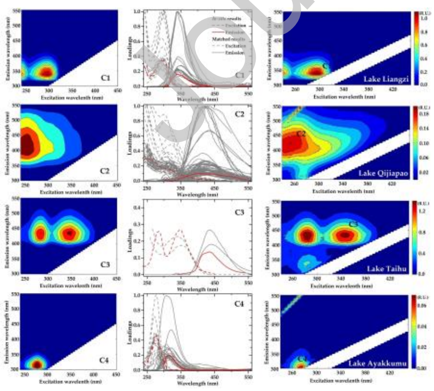

Fig. 2. The PARAFAC results and the representative CDOM-EEMs examples. The left figures are the four CDOM fluorescence components. The middle figures are the comparisons with the OpenFluor database. The right figures are the typical CDOM-EEMs examples for the four CDOM components in the deep europhic Lake Liangzi (114°34′E, 30°14′N) in the EPL, the

brackish Lake Qijiapao (124°17′E, 46°49′N) in the NPML, the shallow eutrophic Lake Taihu (120°05′E, 31°13′N) in the EPL, and the saline Lake Ayakkumu (89°18′E, 37°32′N) in the IMXL.

With Tucker congruence coefficients of > 0.95, we found 26, 78, 3, and 21 matches for the four CDOM fluorescence components from the OpenFluor database (Fig. 2). Component C1 has its peak at the excitation / emission wavelengths of 235(290) / 350 nm and is identified as an autochthonous tryptophan-like substance (Cawley et al., 2012). Component C2 reaches its maximum at 260 /400 nm and is associated with allochthonous humic-like substances (Limberger et al., 2019). Component C3 shows peaks at 280(350) / 440 nm and is characterized as an allochthonous terrestrial humic-like substance (Gullian-Klanian et al., 2021). Component C4 peaks at 275 / 310 nm, indicating an autochthonous tyrosine-like substance (Yang et al., 2019).

2.3 Lake basin delineation

This study determined basins for the 390 lakes investigated in the 1980s and/or 2010s (Section 2.2.1). This were involved three steps. First, water flow directions were calculated from the DEM data (Supplementary Table S1) by utilizing the D8 method (O'Callaghan and Mark, 1984), and rivers flowing into lakes were visually selected by overlaying the flow direction results with the river network of the HydroRIVERS dataset (Lehner and Grill, 2013). Second, basin boundaries of the selected rivers flowing into lakes were automatically delineated from the flow accumulations using a published software with a user-friendly graphical interface (Xie et al., 2022). Third, all inflowing river basins for a given lake were merged and manually corrected using the HydroBASINS dataset (Lehner and Grill, 2013). Supplementary Fig. S1 shows the basins for the lakes studied. Note that the basin boundaries of some lakes overlap.

2.4 Basin data extraction

The lake basin boundaries were used to calculate the arithmetic mean values in both the 1980s and 2010s of seven basin properties, that have been reported to have influences on organic pollution in lakes (Song et al., 2018). Due to limitations in data availability, some basin characteristics only had data available in certain years. For the basin data used (Supplementary Table S1), the 1-km gridded data of population density and land cover in both 1990 and 2015 were obtained from the Resource and Environmental Science Data Platform (RESDP, https://www.resdc.cn/). Moreover, the 1-km gridded data of annual mean wind speed, mean air temperature, total precipitation, and mean normalized difference vegetation index (NDVI, 8-km) in both the 1980s and 2010s were also obtained from the RESDP. In addition, the 1-km gridded data of soil organic carbon (SOC) content in both the 1980s and 2015 were provided by the national Tibetan Plateau Data Center (https://www.tpdc.ac.cn/home/). Using these original data, we first calculated the grided mean values for the 1980s and 2010s, and then calculated the arithmetic average values of the different lake basins.

2.5 Statistical analysis techniques

In this study, all statistical analyses were conducted using SPSS 22.0 software. Correlation regressions were employed to explore the correlations between different indicators or the effects of different drivers on organic pollution in lakes. Independent samples *t*-tests were used to determine whether the differences in lake COD concentrations (basin properties) in the 1980s and 2010s were significant. In addition, the general linear model (GLM) was applied to assess the contributions of different drivers to spatial changes in lake COD concentrations. The GLM approach, which utilize an information-theoretic framework and Akaike's information criterion, helps to minimize model selection bias through stepwise regressions (Tong et al., 2017). In particular, the statistical results were considered significant at a *p*-value of less than 0.05 (two-tailed test).

3 Results

3.1 Environmental differences in China's lakes

Chinese lakes were characterized by high eutrophication in the southeast. In the 2010s, lakes in the southeastern YGPL, NPML, and EPL zones had mean Chl-a concentrations of 81.08 ± 87.16 μg/L, 40.55 ± 59.86 μg/L, and 31.26 ± 34.23 μg/L, respectively (Fig. 3a). In comparison, lakes in the northwestern IMXL and TPL zones had mean Chl-a concentrations of only 5.25 ± 25.42 μg/L and 0.88 ± 0.97 μg/L, respectively. In addition, lakes in the southeastern EPL zone also had higher TN (1.49 ± 1.01 vs. 1.17 ± 1.09 mg/L) and TP (1.12 ± 0.26 vs. 0.09 ± 0.36 mg/L) concentrations than lakes in the northwestern TPL zone. Overall, lake eutrophication was spatially consistent with the geographical distribution of population density and cropland, with high levels in southeastern China (Fig. 3a, Supplementary Figs. S2a-b).

Lakes with low eutrophication in northwestern China generally showed high salinization levels, as indicated by high water conductivities. In the 2010s, all sampled lakes in the northwestern IMXL (*N* = 15) and TPL (*N* = 72) zones had mean *in-situ* water conductivities as high as 23.66 ± 44.42 ms/cm and 23.0 ± 32.2 ms/cm, respectively (Fig. 3b). However, lakes in the southeastern YGPL, NPML, and EPL zones had water conductivities of only 0.45 ± 0.22 ms/cm, 1.04 ± 1.13 ms/cm, and 0.33 ± 0.26 ms/cm, respectively. Lakes with high water conductivities were mainly distributed in the arid areas with low precipitation and/or high air temperature (Fig. 3b, Supplementary Figs. S2c-d). However, some lakes fed by glacial meltwater with low conductivities were also found in arid areas. For example, Lake Kanas (48.81°N, 87.01°E) in the arid IMXL zone had a very low conductivity of only 0.059 ms/cm.

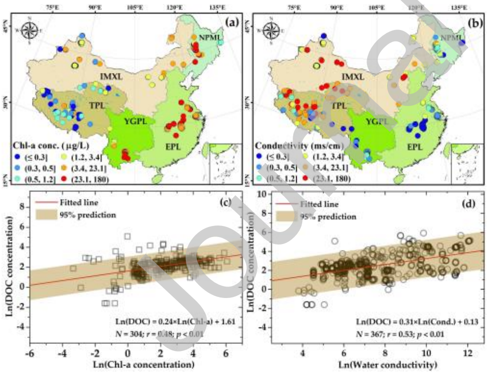

Fig. 3. The environmental characteristics of lakes throughout China in the 2010s. The geographical distributions of (a) *in-situ* Chl-a concentrations and (b) water conductivities. The linear relationships (c) between log DOC concentrations and log Chl-a concentrations and (d) between log DOC concentrations and log water conductivities.

3.2 Spatiotemporal changes in lake COD concentrations

Along with the spatial variations in environmental characteristics, *in-situ* lake COD concentrations showed a general spatial trend of being higher in the north compared to the south of China (Figs. 4a-b). Lake-based average COD concentrations were positively linearly correlated with lake latitude in both the 1980s (N = 102, r = 0.41, p < 0.05) and the 2010s (N = 346, r = 0.45, p < 0.01). To be specific, the two northern IMXL and NPML zones had mean COD concentrations of 9.42 ± 5.55 mg/L and 7.81 ± 6.24 mg/L in the 1980s and 2010s, respectively (Figs. 4a-b); in comparison, the three southern TPL, YGPL, and EPL zones had mean COD concentrations of 5.65 ± 7.08 mg/L and 4.79 ± 2.76 mg/L in the 1980s and 2010s, respectively. In addition, we also found that the southern YGPL and EPL zones with high population density had high COD concentrations in some lakes (> 6.0 mg/L), which accounted for 21.88% and 26.59% of the lakes investigated in the 1980s and 2010s, respectively (Figs. 4a-b).

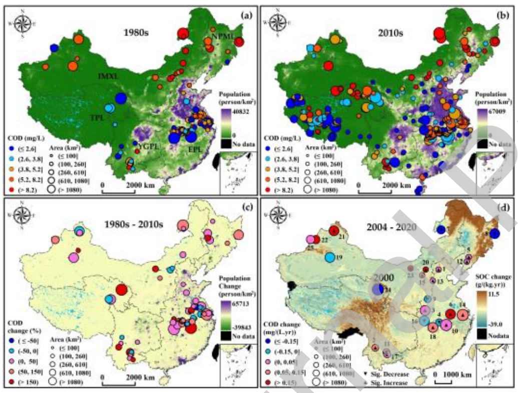

Fig. 4. The spatiotemporal variations of the lake-based mean COD concentrations. Mean COD concentrations in the (a) 1980s (N = 102) and (b) 2010s (N = 346). (c) Changes in COD concentrations between the 1980s and 2010s (N = 83, Supplementary Table S2). (d) Change rates of COD concentration during 2004-2020 (N = 25, Supplementary Fig. S3).

Along with the intensification of human activity as indicated by increasing population density (Figs. 4a-b), most lakes showed increasing COD concentrations between the 1980s and the 2010s. For lakes with measurements in both eras (N = 83, Supplementary Table S2), lake COD concentration in the 2010s was significantly higher than in the 1980s (t-test, p < 0.05), and 78.31% of lakes showed increased

COD concentrations (Fig. 4c). Furthermore, for the 25 lakes with *in-situ* time series data during 2004-2020, 18 (72%) showed increased COD concentrations and 16 (64%) witnessed significant increases (p < 0.05, Supplementary Table S3). In addition, attention should also be paid to lakes Qinghai, Hulun, and Xingkai, which showed decreasing COD concentrations and expanding areas during 2004-2020 (Fig. 4d). These changes indicate that water eutrophication caused by anthropogenic pollution and lake expansion caused by climate change have important impacts on organic pollution in Chinese lakes.

3.3 Relationships between COD and environmental factors

COD concentrations in the 2010s were strongly related to environmental factors, especially water conductivities and eutrophication levels. On the one hand, organic matter accumulated in lakes together with salinity. Compared to freshwater lakes (N = 229) with conductivities < 1,000 μs/cm, saline lakes (N = 93) had higher mean COD concentrations in the 2010s (6.23 ± 4.85 mg/L vs. 4.86 ± 2.96 mg/L). Furthermore, a significant linear correlation between log COD concentrations and log conductivities was observed for saline lakes (r = 0.49, Fig. 5a). On the other hand, water eutrophication increased organic pollution. Lakes with Chl-a concentrations below 10 μg/L (N = 199) had COD concentrations of 4.38 ± 3.55 mg/L, while those with Chl-a concentrations above 10 μg/L (N = 114) had COD concentrations of 6.42 ± 3.01 mg/L. Furthermore, significant positive correlations were found between log COD concentrations and log Chl-a concentrations (r = 0.33, Fig. 5b), log TN concentrations (r = 0.56, Fig. 5c), and log TP concentrations (r = 0.61, Fig. 5d) during the 2010s. In addition, COD concentrations were not significantly correlated with conductivities and all three eutrophication factors in the 1980s (Fig. 5), highlighting instead the increasingly important impacts of water eutrophication and lake expansion on organic pollution in Chinese lakes during the 1980s-2010s.

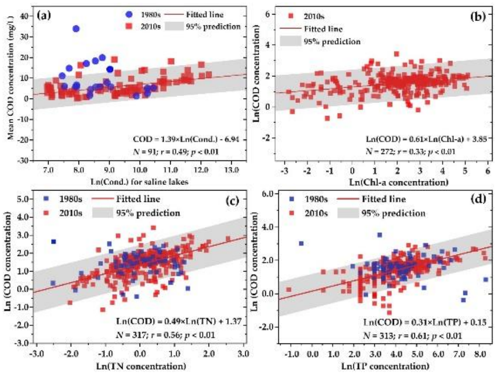

Fig. 5. Effects of environmental characteristics on COD concentrations in lakes. The linear relationships between log COD concentrations and (c) log water conductivities for saline lakes (> 1,000 μs/cm) (Mayer et al., 2005), (d) log Chl-a, (e) log TN, and (f) log TP concentrations in the 2010s. *In-situ* data from the 1980s are also shown (Supplementary Table S1).

4 Discussion

4.1 Drivers of changes in COD concentrations

Based on the GLM method, we quantified the relative contributions of eleven anthropogenic (TN and TP concentrations in lakes; population density and cropland coverage percentages in basins) and natural (conductivity and SDD in lakes; wind speed, air temperature, precipitation, NDVI, and SOC in basins) factors to the spatial variation of COD concentrations (Sections 2.5, 3.2). The total contribution of anthropogenic factors was only 24.69% in the 1980s and increased to 40.22% in the 2010s (Fig. 6a). Thereinto, TN concentration had an increasing contribution from 0.27% to 35.24% during the 1980s-2010s. In addition, natural precipitation and air temperature contributed more to the spatial variation of COD concentrations in the 2010s (41.14%) than in the 1980s (4.26%). The increasing contributions of TN concentration, precipitation, and air temperature indicated that eutrophication and climate change played an important role in the changes of COD concentrations in lakes during the 1980s-2010s.

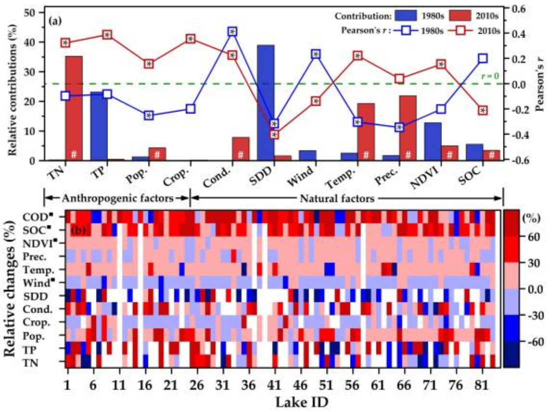

Fig. 6. Potential factors influencing the spatiotemporal changes in lake organic pollution during the 1980s-2010s. (a) The relative influence of different drivers on spatial variations. The symbols "#" indicate the significant contribution factors ($p < 0.05$). *r* values between COD concentrations and different indicators are also shown, and the symbols "*" indicate the significant correlation factors ($p < 0.05$). (b) Relative changes of different factors for different lakes (Supplementary Table S2). The symbols "■" indicate the significantly changed factors (*t*-test, $p < 0.05$, Section 2.5). Pop: population density; Crop.: cropland coverage percentage; Cond.: conductivity; Temp.: air temperature; Prec.: precipitation.

4.1.1 Water eutrophication

Human-induced water eutrophication elevated COD concentrations in Chinese lakes. Spatially, water eutrophication mainly influenced lakes in the two eastern EPL and NPML zones, characterized by high Chl-a concentrations and population density (Fig. 3a, Figs. 4a-b). Liu et al. (2020a) reported that eutrophication increased lake DOC concentrations in the EPL zone, and Zhu et al. (2019) reported that nutrient levels were the critical drivers that altered lake DOC composition in the NPML zone. Temporally, for the 83 lakes investigated in both the 1980s and 2010s, intensifying human activity, as indicated by increasing population density, raised TN and TP concentrations in 53.06% and 58% of the lakes, respectively (Fig. 6b). As a result, 88.94% of freshwater lakes in China showed an increasing frequency of algal blooms during the 1980s-2010s (Hou et al., 2022). Meanwhile, half of the saline lakes in the remote TPL zone have also experienced an increase in Chl-a concentrations due to increased nutrient inputs (Pi et al., 2021). Therefore, we believe that water eutrophication has been the key driver for elevating COD concentrations in both freshwater and saline lakes in China during the 1980s-2010s (Figs. 4c-d). Furthermore, *in-situ* data from the 2010s revealed a significant positive relationship between log-transformed Chl-a and DOC concentrations (Fig. 3c).

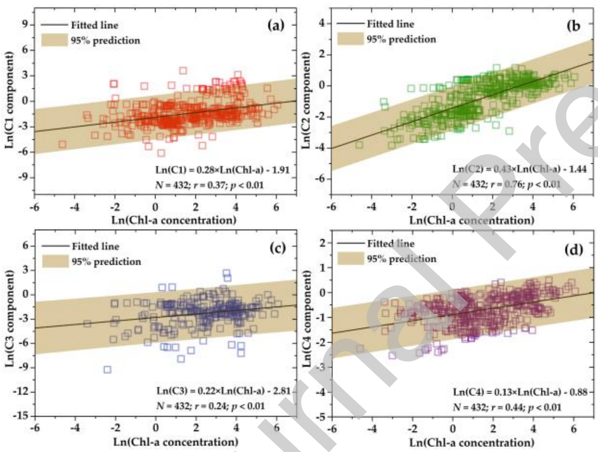

Fig. 7. Effects of water eutrophication on organic pollution. (a - d) The linear relationships between log Chl-a and log the intensities of the four CDOM components (Section 2.2.3).

Water eutrophication increases not only autochthonous organic matter via phytoplankton production (Baines and Pace, 1991; Zhang et al., 2010) but also anthropogenic organic byproducts derived from human activities (Bauer et al., 2013; Peleato et al., 2017). For Chinese lakes, eutrophication-induced algal proliferation released a large amount of autochthonous dissolved organic matter through degradation and secretion processes, as evidenced by the significant correlations between Chl-a and levels of autochthonous components C1 (r = 0.37, Fig. 7a) and C4 (r = 0.44, Fig. 7d). Furthermore, algae themselves are also a type of autochthonous particulate organic matter (Artifon et al., 2019). In addition, organic byproducts of human activity also entered the lakes along with nutrients, as indicated by the significant correlations between Chl-a and the levels of the allochthonous CDOM components C2 (r = 0.76, Fig. 7b) and C3 (r = 0.24, Fig. 7c). In these cases, due to algal proliferation and wastewater discharge, increasing COD or DOC concentrations have been found in several typical Chinese lakes during 2000-2020, such as eutrophic Lake Taihu (Liu et al., 2021b), saline Lake Daihai (Liang, 2021), and polluted Lake Wulungu (Han and Ma, 2015).

4.1.2 Lake expansion

Saline lakes are typically tail-end waters, and terrigenous refractory organic matter accumulates in lakes along with water evaporation (Bianchi, 2011). In addition, saline lakes typically exhibit higher levels of saturated and oxidized DOC than freshwater lakes (Butturini et al., 2022). Here we also identified a significant relationship between log DOC concentrations and log water conductivities in the 2010s (r = 0.53, Fig. 3d). Furthermore, many lakes with increased COD levels during the 1980s-2010s also showed increased conductivities (Fig. 6b).

However, due to increasing precipitation and/or glacial melting, many saline lakes in the northwestern arid/semi-arid IMXL and TPL zones experienced water level ascension and area expansion during the 1980s-2010s (Li et al., 2022; Yao et al., 2015). Compared with saline lakes, rain and glacial waters are freshwater and have low DOC concentrations of < 1.0 mg/L in northwestern China (Safieddine and Heald, 2017). Therefore, many saline lakes in the TPL zone are experiencing an apparent decline in water salinity (Song et al., 2022). For example, the COD concentration in the expanding Lake Qinghai, the largest saline lake in China, has declined (Li et al., 2021b). Namely, lake expansion would attenuate organic pollution in saline lakes, which could be due to the dilution effects of precipitation.

Expansion of saline lakes would dilute allochthonous/terrestrial organic matter accumulated in the lakes with water salinization (Chen et al., 2022; Li et al., 2021). For saline lakes with conductivities of > 25 ms/cm in the 2010s, decreasing salinity led to significant decreases in both allochthonous / terrestrial humic CDOM components C2 (r = 0.42, Fig. 8b) and C3 (r = 0.87, Fig. 8c), which were positively related to water conductivities. However, lake expansion had different effects on the intensities of the autochthonous CDOM components C1 (r = 0.44, Fig. 8a) and C4 (r = -0.46, Fig. 8d), and the effects were significant (p < 0.01). Possible reasons include: ① similar to allochthonous components C2 and C3, lake expansion diluted the autochthonous component C1; ② together with river inputs, a large amount of nutrients were transported into saline lakes and promoted the production of autochthonous component C4 by stimulating algal proliferation (Liu et al., 2021c). It has been reported that many saline lakes in the remote TPL zone have increasing nutrient levels (Wu et al., 2021) and Chl-a concentrations (Pi et al., 2021) due to urbanization and economic development.

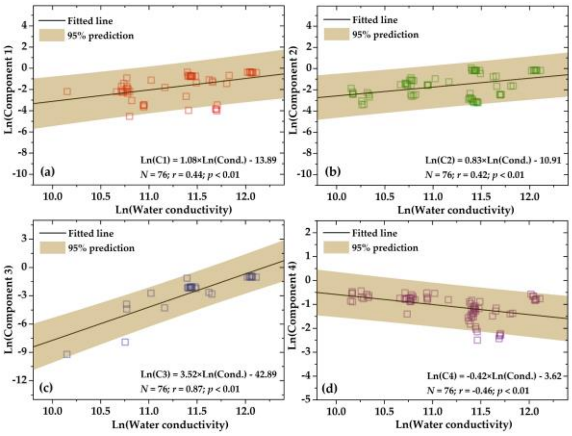

Fig. 8. Effects of lake expansion on organic pollution. (a - d) The linear relationships between log conductivities and log intensities of the four CDOM components (Section 2.2.3) for lakes with conductivities of > 25 ms/cm in the 2010s.

4.1.3 Basin characteristics

Basin characteristics, including NDVI, air temperature, SOC, and precipitation, might exacerbate organic pollution in Chinese lakes during the 1980s-2010s by increasing the input of riverine organic matter. First, higher NDVI in the 2010s (Fig. 6b) could indicate more terrestrial input of organic matter produced by the decomposition of fallen leaves. Su et al. (2023) reported that vegetation restoration elevated SOC content in the Loess Plateau, a typical area with increasing NDVI in China. Second, higher air temperature in the 2010s (Fig. 6b) could lead to faster decomposition rates of fallen leaves. For U.S. rivers entering the ocean, Tian et al. (2013) reported that rising air temperature could elevate DOC concentrations by 0.48 mg/(L·°C). Third, higher SOC in the 2010s (Fig. 6b) could signify more organic matter that could be flushed into lakes. Nosrati et al. (2012) found that increasing SOC elevated riverine DOC concentrations in a semi-arid basin in Iran. Finally, increased precipitation in the 2010s (Fig. 6b) could flush more terrestrial organic matter into lakes. Globally, riverine organic matter fluxes are typically determined by water discharge (Bauer et al., 2013; Nosrati et al., 2012).

4.2 Limitations of using COD to indicate organic pollution

Due to the lack of large-scale and long-term measurements of organic carbon, this study adopted COD data to reflect the spatiotemporal changes of organic pollution in Chinese lakes during the 1980s-2010s. Globally, lakes, rivers, and/or oceans generally show positive relationships between the concentrations of COD and DOC, the existing form of approximately 90% organic carbon (Jiao et al., 2021). For different Chinese lakes, Song et al. (2018a) reported a linear fitting formula between DOC concentrations and COD measured by the Potassium dichromate method. *In-situ* COD concentrations in this study were also related to DOC concentrations, but the correlations varied between different lake zones (Fig. 9). The fitting slopes between log DOC and log COD concentrations of the northwestern IMXL, NPML, and TPL zones (Fig. 9b-c) had higher values than those of the southeastern EPL zone (Fig. 9d). Therefore, there are limitations in using the COD metric to indicate organic pollution in different lake types.

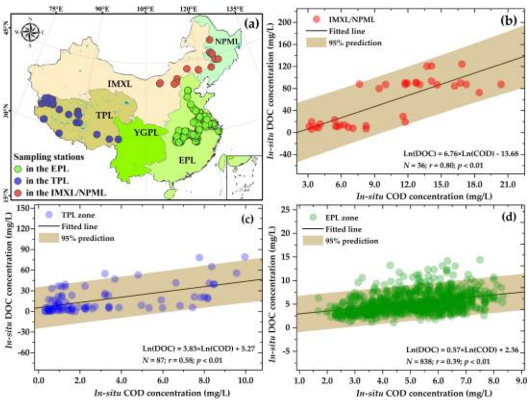

Fig. 9. Relationships between *in-situ* DOC and COD concentrations. (a) Sampling stations with synchronous *in-situ* DOC and COD concentrations in the 2010s (Section 2.1). (b) The linear relationships between *in-situ* DOC and COD concentrations for (b) the NPML and IMXL zones, (c) the TPL zone, and (d) the EPL zone, separately.

There are two main reasons for the limitations of using COD to indicate organic pollution. First, COD concentrations only reflect the content of organic matter that can be reduced by potassium permanganate; however, some complex organic matter such as lipids and polyaromatic hydrocarbons are not sensitive to potassium permanganate (Walter, 1961). Secondly, different types of organic matter have different consumption rates of potassium permanganate. For example, both organic acids and alcohols can be reduced by potassium permanganate, but the partially oxidized organic acids consume less potassium permanganate (Walter, 1961). In these cases, we recommend using COD only to indicate the general spatial pattern of organic pollution in different lakes and the temporal changes in organic pollution in a particular lake, as was done in this study.

5 Conclusions

Based on COD measurements in 390 Chinese lakes, this study revealed the spatiotemporal changes of organic pollution in China's lakes during the 1980s-2010s. Across China, organic pollution was generally worse in northern lakes and better in southern lakes, as indicated by the significant positive linear relationships between COD concentrations and lake latitude (1980s: r = 0.22; 2010s: r = 0.35). Water eutrophication and salinization jointly determined the spatial pattern of organic pollution, and anthropogenic factors had a total relative contribution of

40.22% in the 2010s. Along with the intensification of human activity, 78.31% of the studied lakes showed increasing COD concentrations during the 1980s-2010s. However, the expansion of lakes due to climate change resulted in decreasing COD concentrations in saline lakes in the northwestern lakes. These findings are of great value for understanding and controlling organic pollution in different lakes over a large area along with environmental changes.

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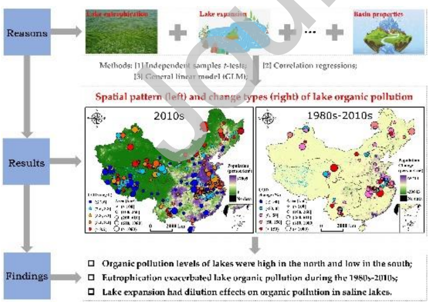
Graphical Abstract



Declaration of interests

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

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: