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Chemical composition, sources, and ecological effect of organic phosphorus in water ecosystems: a review

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Abstract

Phosphorus (P) affects the water–air CO_2 flux through primary productivity, and affects the changes of carbon cycle and ecological environment in the global world. Organic phosphorus (Po) is an important P component in water environments. Various processes control the formation and transformation of Po and outbreaks of algal blooms. Here, research topics on Po in global water over the past 50 years are systematically reviewed to understand the progress in the (i) Po pretreatment techniques in various media, (ii) technical methods and qualitative and quantitative research on chemical composition of Po and bioavailability, (iii) source analysis of and factors affecting Po in different media of water environments and biogeochemical processes, (iv) interactions among Po, organic matter, and minerals, and their environmental behaviors, and (v) quantification of material exchanges at the sediment–water interface, interfacial processes, and ecological effects. Finally, the future research directives regarding Po in water environments are discussed. The findings provided an important scientific basis to formulate and revise global standards for water nutrients and a better understanding of water eutrophication and its control.

Highlights

- Significant evidence was obtained for the bioavailability of organic phosphorus in water ecosystems.
- The environmental and biogeochemical behaviors of organic phosphorus in different media were reviewed.
- Quantitative mechanistic model was proposed for organic phosphorus migration and transformation in lakes.

Keywords Organic phosphorous, Carbon cycle, Phosphorus-31 nuclear magnetic resonance, Enzymatic hydrolysis, Organic matter, Eutrophication

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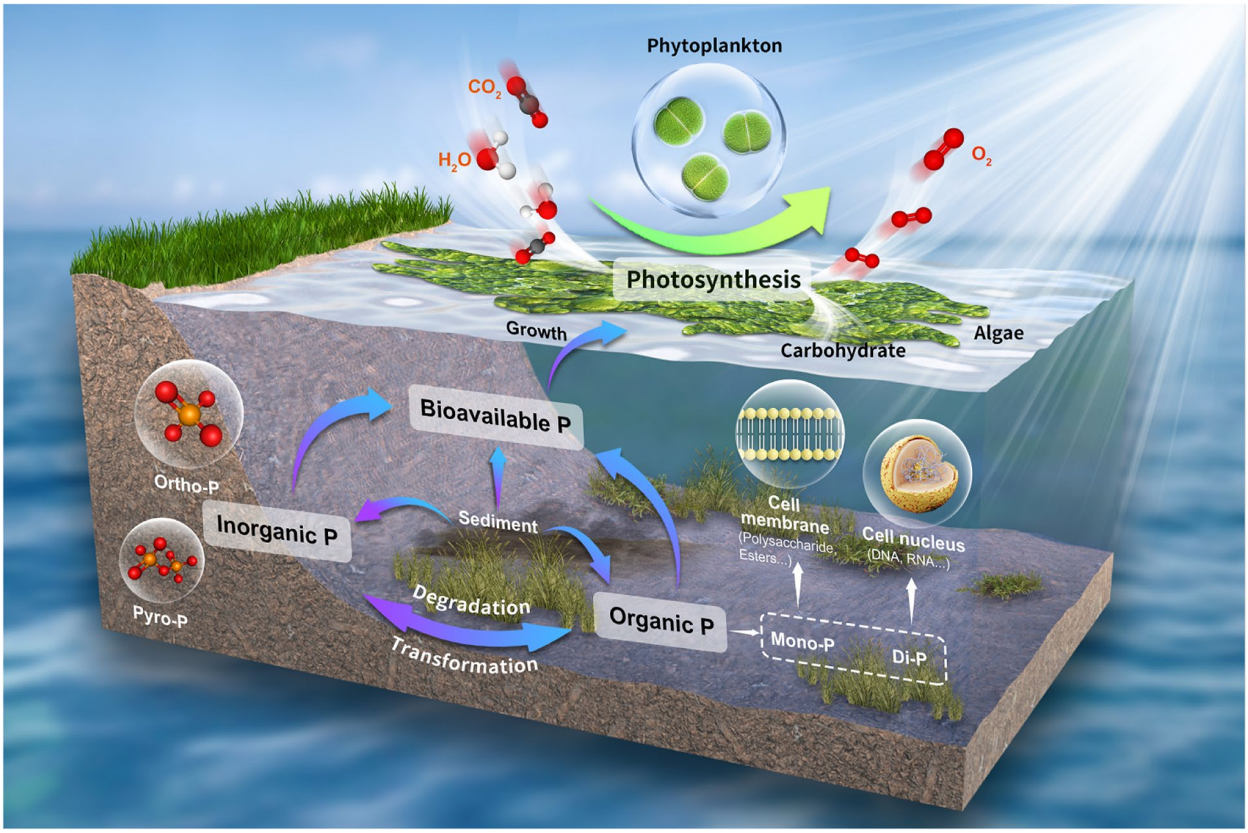
Graphical Abstract



1 Introduction

Under multiple impacts of climate change, urbanization and industrial development, lakes, rivers, sea and reservoir around the globe are facing serious issues of water shortage, and deterioration and increased vulnerability of aquatic environments (Sohail et al. 2022; Søndergaard et al. 2003; Liu et al. 2016a, b; Stackpoole et al. 2019). Phosphorus (P) is one of the basic nutrients of aquatic life. Its distribution, content and geochemical cycle directly affect the primary productivity of aquatic plants and the quantity of plankton, and further affect the global carbon cycle and even the global climate (Falkowski et al. 2000; Zhang et al. 2001; Wu et al. 2022a, b).

In the ocean or lake, P is an important limiting element, and the changes of O_2 and CO_2 in the atmosphere are caused by the changes of phosphorus. Carbon is fixed in organic matter by plankton photosynthesis, and phosphorus plays a leading role in the reproduction of plankton and the whole biological cycle (Hu et al. 2022a). Even for tiny microorganisms, they still promote the global carbon cycle (Malone et al. 1990; Wu et al. 2022a, b). Meanwhile, P is a key element of water eutrophication, not only essential to sustain life, but also an important biogenic element that when presented in excess can cause outbreaks of cyanobacterial blooms (Dillon et al. 2005; Reinhard et al. 2017; Zhu et al. 2020; Jia et al. 2022). Moreover, it forms the backbone of DNA and is a major component involved in important life processes, such as energy transfer in cell membranes (McIntyre et al. 2020). Lacustrine eutrophication, which is affected by multiple compounded factors and complex pollution sources, can threaten the quality of lacustrine environments along with drinking water quality (Søndergaard et al. 2003; Liu et al. 2016a, b; Stackpoole et al. 2019). Waterbodies have been severely polluted by P, and water eutrophication has occurred frequently. Therefore, controlling the P load in water environments is a primary issue that should be addressed to maintain the quality of lacustrine environments (Wu et al. 2022a, b).

Presently, investments are being made in treating water eutrophication in China and overseas. Particularly, strategies to control exogenous pollution, such as establishing discharge standards for sewage and wastewater, improvements in equipment and processes, technological upgrades, and deep purification of P-containing industrial wastewater, have been emphasized. In 2000, following the increased investments in scientific research, the comprehensive eutrophication index of typical lakes reached an inflection point such that total P in lakes had been well controlled (Li et al. 2019a, b, c). However, severity and frequency of cyanobacterial bloom outbreaks did not reduce.

Surface water contains both inorganic and organic phosphorus (Po). Using "inorganic P" as the key search term, in the CiteSpace® program, 45,000 articles and reports were retrieved from the Web of Science Core Collection published as of June 18, 2022. Organisms in water environments directly utilize the soluble orthophosphate (Ortho-P) released from inorganic P. Multiple studies have assessed the spatiotemporal distribution of its content and forms, and interfacial migration conversion, and regeneration mechanisms, and estimated its endogenous load and strategies to control water eutrophication (Hakulinen et al. 2005; Li et al. 2009; Mackey et al. 2019). Po is an important component of various media including water, particulate matter, sediments, algae, and aquatic plants (Cade-Menun et al. 2006; Ji et al. 2017; Bai et al. 2017; Yuan et al. 2020; Ni et al. 2022). Po accounts for 21–60% of total amounts of P in sediments (Zhang et al. 2008; Ding et al. 2010), 15.5–72.4% of total P in aquatic plants and algae (Feng et al. 2016a; b), and more than 50% of total P in particulate matter (Feng et al. 2020). It is also a potential source of bioavailable P that causes frequent outbreaks of cyanobacterial blooms and a key factor in maintaining the eutrophic state of surface waters. Therefore, the biogeochemical processes of Po, e.g., migration and conversion patterns, and techniques for its reduction have gained increasing attention; However, studying mechanisms responsible for water eutrophication and the techniques for controlling water pollution at a global scale is difficult (Yuan et al. 2020; De Brabandere et al. 2008; Ni et al. 2019a).

In oceans and lakes, due to the complex composition of Po and limited technical means for its detection, quantification of fluxes of Po is often underestimated compared to inorganic P (De Brabandere et al. 2008; Zhu et al. 2013; Ma et al. 2022a, b). Here, research on Po in the surface water multi-media over the past 50 years was reviewed to better understand sources and dynamics of P and blooms of hazardous algae in waterbodies to effectively control water eutrophication that affects quality of aquatic environments. The current research trends and common topics were reviewed, along with new methods, techniques, and theoretical models, with an aim to systematically sort and summarize the main findings, for example, on the chemical structure and bioavailability of Po and constructive solutions and directives for future research on Po.

2 Research history and leading-edge topics on Po in various media of global water environments over the past 50 years

On October 31th, 2022, 8,504 articles were retrieved from the Web of Science Core Collection using the keywords "lake / river / sea or ocean / reservoir / water / surface water" and "organic phosphorus / organophosphorus / Po," but excluding the search term "pesticide". The number of published articles on Po in lakes, rivers, sea and reservoirs accounted for 36.8%, 33.7%, 20.5% and 9.1%, respectively (Fig. 1a). More than 70% of the articles published have studied Po in lakes and rivers (Fig. 1a). The earliest study on Po in surface water, which was published in 1975, characterized soluble Po using various enzymes (Herbes et al. 1975). Figure 1a shows that 1–2 articles on Po were published annually during 1976–1990. Subsequently, international literature on Po increased rapidly after 1991. However, Chinese literature on this topic was relatively less, with articles published only after 2000, followed by a steadily increasing trend. Organic phosphorus has been studied in more than 30 countries. Among them, China and the United States accounted for 75.5% of all countries' Po research, where the main keywords were eutrophication, theoretical models, heavy metals, organic chemicals, plankton, water quality, and bacteria. With the increase of time, Po was combined with eutrophication and water quality (Fig. 1b).

According to the statistics of the study sites of Po in lakes and rivers, research on Po was mainly conducted in North America, China, Europe, Africa, South America, and Oceania, especially in China and the United States. It can be seen that the study sites of lakes and rivers in China were mostly located along the Yellow River and Yangtze River in the east, especially in the southeast. The study of lakes and rivers in the United States mainly focused on the Great Lakes in the Northeast and Mississippi River in the Southeast (Fig. 2). 1570 articles focused on lakes and rivers in North America, accounting for 38.1% of the total research. More than 50% of studies of lakes in North American focused on lakes in the North Temperate zone south of the border between the United States and Canada, and 5.5% of them focused on Lake Erie. This was followed by 1302 research articles on lake Po in China, accounting for 32.8% of the total. Po in Tai Lake accounted for 7.4% of the research in China. In Europe, the Po of high latitude cold zone lakes in Norway, Denmark and Finland was studied, accounting for 16.5% of the total research in European lakes, while

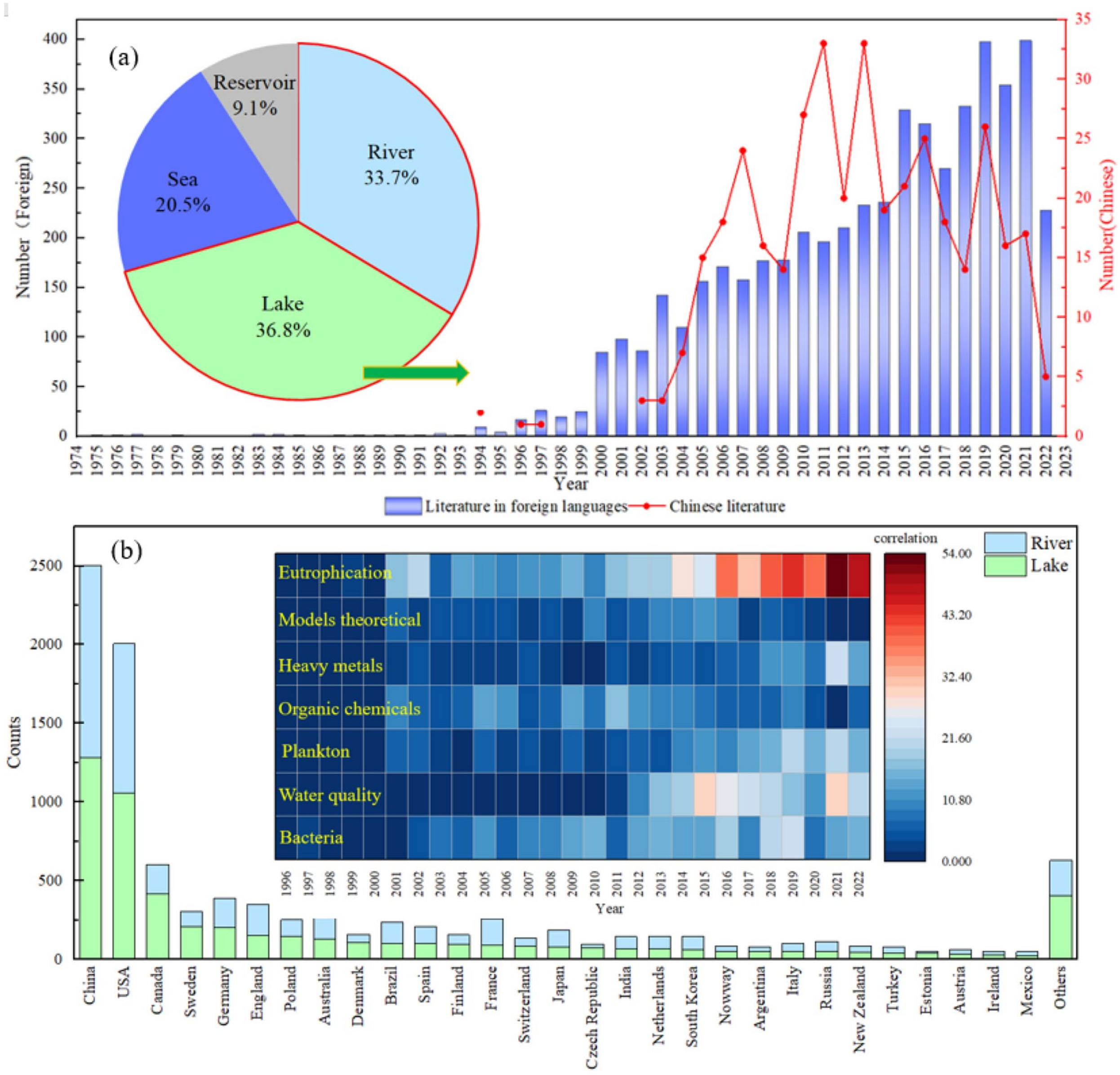


Fig. 1 Distribution and research of organic phosphorus in the main countries and regions. (**a** The change of the published numbers of Po over time and its distribution in various surface waters; **b** The distribution of published Po papers in lakes and rivers and the trend of Po research over time in the top 30 countries with the most research. The data collection time range was from January 1st 1975 to October 31th 2022)

the research on the temperate lakes along the Mediterranean coast accounted for 15.7% of the research in European lakes. In South America and Africa, the impacts of Po on climate in subtropical lakes were studied, accounting for 56.7% of the research on the lakes in the region.

The connections between the articles were analyzed using the visualization retrieval function of CiteSpace® (Panahi et al. 2020; Chen et al. 2012). CiteSpace® was used to analyze the keywords of the related articles at an interval of 1 year. The node type was "keyword" and the selection standard was "TOP 5.0%" with 25 being the maximum number of items selected for each interval. The top 10 keywords in decreasing order of identification frequency were "phosphorous," "nitrogen," "water," "organic matter," "phytoplankton," "sediment," "water quality," "eutrophication," "carbon," and "dynamics" (Fig. 3). This indicated that studies on Po in the surface water focused on eutrophication and environmental behaviors of nitrogen (N), P, and organic matter, and were related to carbon and its dynamics. Meanwhile, previous studies in global

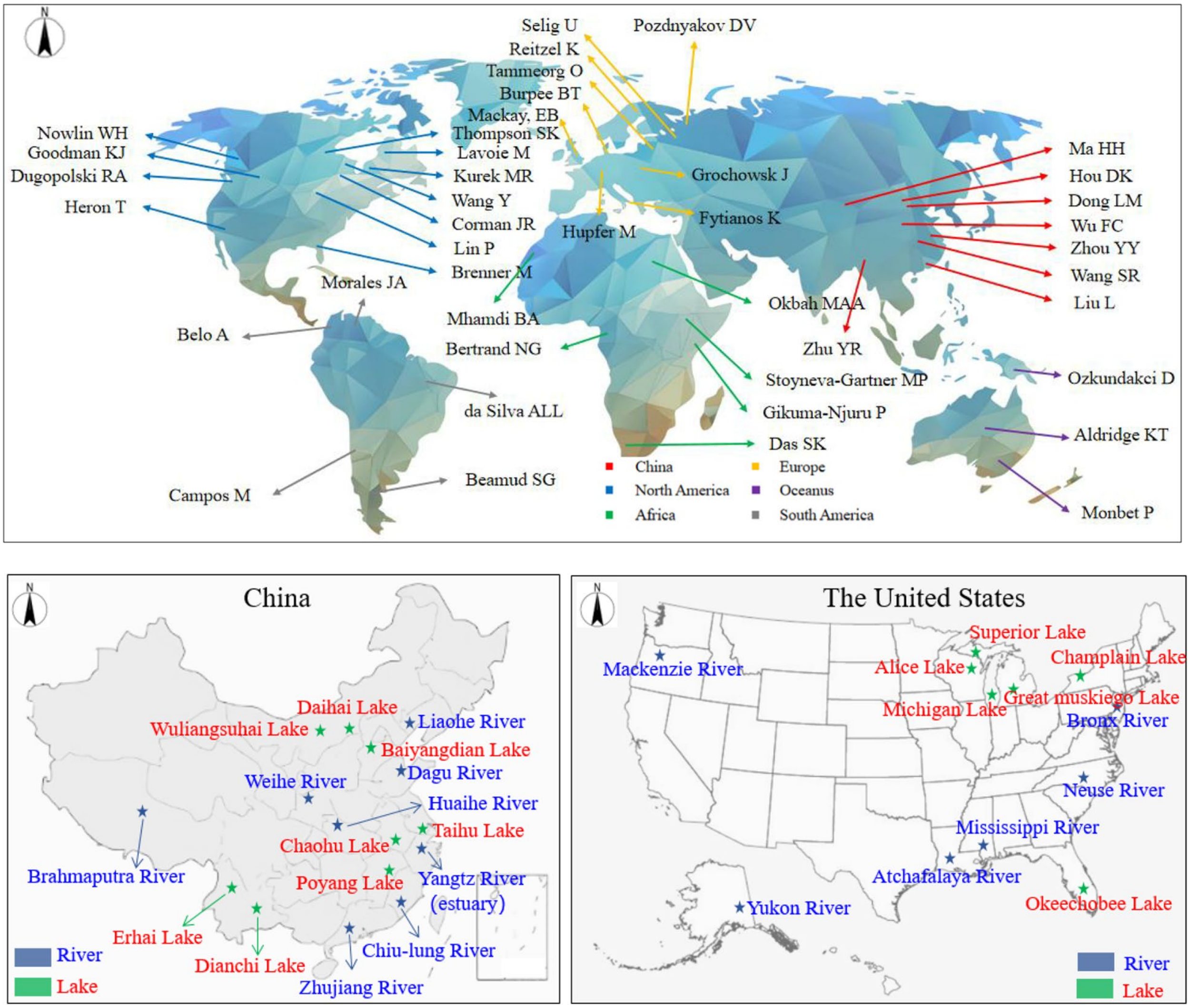


Fig. 2 Distribution of key locations for research on Po in global water and the corresponding main authors

lakes, rivers, sea, and reservoirs, based on the keywords and their connection of Po can be summarized into six categories, which were phosphorus form, research medium, environmental factors, connection with organisms, eutrophication research, and mechnism study, respectively (Fig. 3). On this basis, the study focused on the review of Po pre-treatment techniques in various media, determination of Po chemical composition and bioavailability, source analysis, factors affecting Po, and interactions of Po with other environmental components such as organic matter and minerals. Association with organic matter and other minerals influences Po's characteristics and bioavailability. We believe that the study makes a significant contribution to the water quality of P because the findings provide a scientific basis for formulating strategies to reduce P pollution in the water ecosystems, avoid water eutrophication, and enhance water quality, by formulating more effective strategies for the management of water ecosystems.

3 Research progress on the pretreatment techniques of Po in the multi-media

Analyzing pretreatment techniques for quantification of forms of Po, including enrichment, concentration, extraction, purification, and separation, in the multi-media including waterbodies, sediments, and algae, is important to accurately characterize the chemical characteristics of Po. Techniques for pretreatment and characterization differ depending on media. For example, low Po contents in lacustrine waterbodies must be initially enriched and concentrated before extraction and separation. Furthermore, when Po exists in sediments in a solid state, it must be extracted and enriched before components can be characterized and quantified. Acquiring

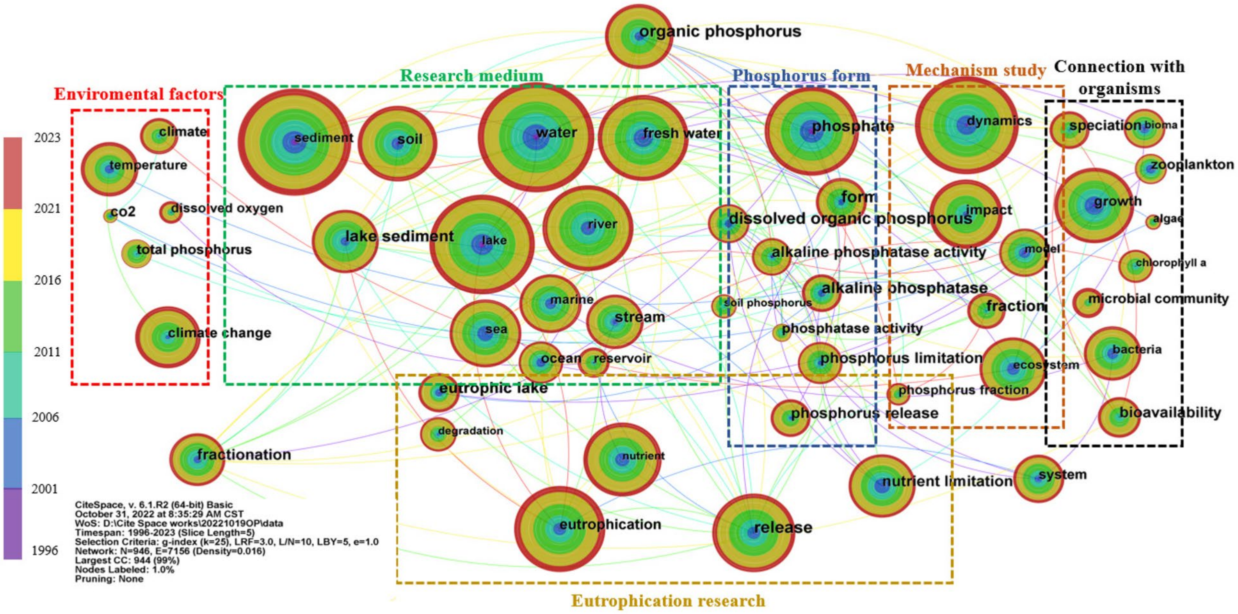


Fig. 3 Study on the key words and their connection of Po in global lakes, rivers, sea and reservoirs (The larger the circle, the more research has been done in that area. The six dashed boxes were shown from left to right and from top to bottom: environmental factors, research medium, phosphorus form, mechnism study, connection with organisms, and eutrophication research. The lines between the key words represent their connections and research) (The articles were published from January 1996 to February 2023)

Po after extraction and purification is technically difficult while pretreating biological cell structures, such as algae and aquatic plants (Feng et al. 2018). Additionally, to improve spectral resolution and accurately characterize the chemical characteristics of Po (Ding et al. 2010; Zhu et al. 2015a, b; Ma et al. 2022a, b), methods to reduce the interference of paramagnetic metal ions, such as iron (Fe), manganese (Mn), nickel (Ni), cobalt (Co), and copper (Cu), when using phosphorus-31 nuclear magnetic resonance (^{31}P NMR) spectroscopy, must be determined.

Presently, NaOH-ethylenediaminetetraacetic acid (EDTA) is commonly used to extract Po from aquatic environments. However, the extraction rate and spectral resolution can be further improved by eliminating paramagnetic ions, such as Fe and Mn (Feng et al. 2018; Jarosch et al. 2015). 8-hydroxyquinoline precipitation can be used to effectively remove 83–91% and 66–78% of Fe and Mn from the NaOH-EDTA extract, respectively (Ding et al. 2010). Additionally, concentrations of extractants used, extraction ratio, and oscillation duration vary depending on media. Different extraction methods have been compared that use various inorganic and organic acids, chelating agents, and alkaline solutions, which have been used individually or in combination, at various temperatures and pH (SI. Table 1). Some studies found that Ca- and Al-bound P could be easily extracted when pH was < 2. However, only Ca-bound P dissolved well when pH was slightly higher, while Al-bound P dissolved well under alkaline conditions (Luyckx et al. 2020). A mixed extractant comprising HCl, H_2SO_4, and oxalic acid effectively balanced the amount of P, small amounts of metals and operational feasibility (Luyckx et al. 2020).

Pretreatment techniques suitable for extraction of Po from multi-media in the surface water of global world were established by exploring and optimizing results of previous studies combined with characteristics of various media. The earliest techniques for extraction of Po date back to 1997, when ultrafiltration was used to study Crystal Lake in the United States (Fig. 4) (Nanny and Minear 1997). Subsequently, membrane-based reverse osmosis was used to analyze waters of the Pacific Ocean, Atlantic Ocean, and 16 oceans of the North Sea for P (Kolowith et al. 2001). However, the operation of these two methods was complex, and extraction membranes were easily clogged. Subsequently, as an improvement, freeze-drying was used to extract Po from the Pee Dee River in the eastern United States. However, the process was time consuming and strong paramagnetic ion interferences were observed (Cade-Menun et al. 2006; Toor et al. 2003).

In 2009, Reitzel et al. (2009) extracted Po from five lakes in Denmark using an Al salt precipitation method, which was also used for extraction of Po from Lake Tai, China (Bai et al. 2014). In 2020, a team led by academician Fengchang Wu combined the Al salt extraction method with NMR spectroscopy and increased

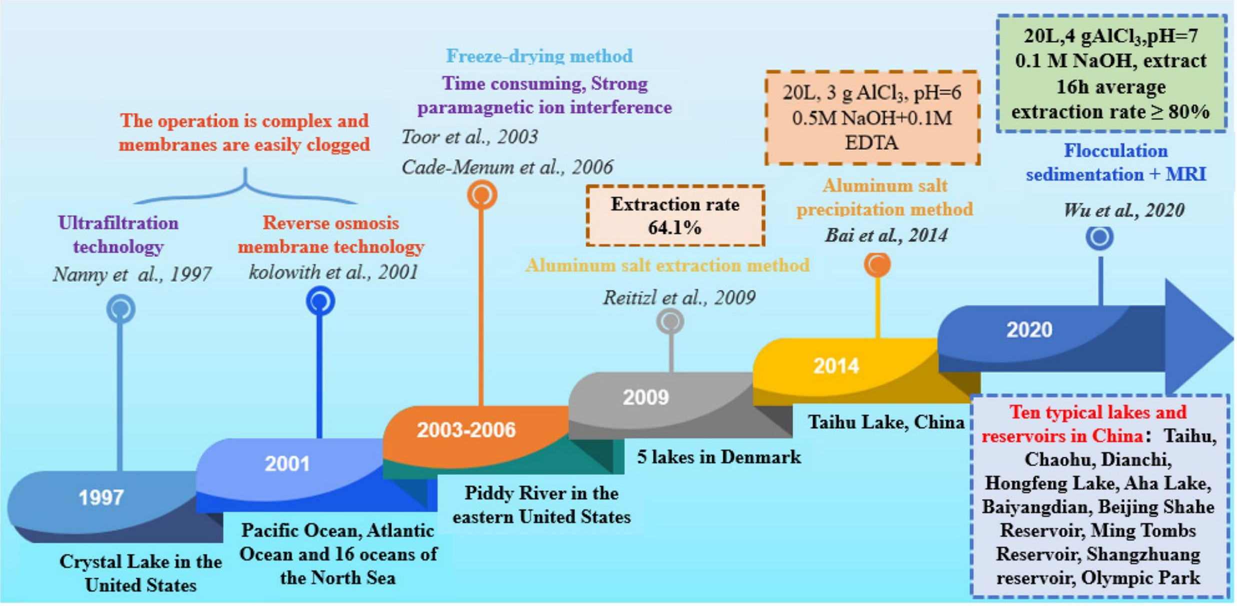


Fig. 4 Multi-level optimization of extraction techniques for Po in global lakes

the mean rate of extraction of Po to > 80% (Feng et al. 2020; Zhang et al. 2019). Moreover, the team proposed not only new techniques for flocculation, sedimentation, enrichment, extraction, and characterization of Po from 20 waterbodies with a large volumetric capacity by solution ^{31}P NMR, but also optimal conditions for the flocculant, extractant, and pH used, as well as duration of extraction. Based on this, the first batch of compositional data for dissolved P (DP) and particulate P (PP) in 10 typical lakes and reservoirs in China was developed (Feng et al. 2020; Zhang et al. 2019). Furthermore, Feng et al. (2016a; 2018). established a new technique for enrichment, concentration, and extraction of Po from algae and aquatic plants with an efficiency of extraction of 95% by enzyme hydrolysis and solution ^{31}P NMR technology. They further proposed the optimal extractant, optimal delay time, and detection time, which produced high-resolution NMR spectra without any superimposed spectral peaks.

In addition, NaOH-EDTA is considered as the best and most stable extractant of Po, and thus, has been adopted in several studies (Bahureksa et al. 2021; Amelung et al. 2017; Doolette et al. 2018). Zhu et al. (2018) established a new technique for multi-level continuous extraction and efficient identification of Po in sediments, which addressed the problem of strong interferences by paramagnetic metals, such as Fe and Mn. Moreover, they discontinued the use of Ortho-P as a reference displacement compound. Through experimentation and repeated comparisons and verifications, they proposed a new standard for the chemical shift of P during NMR spectroscopy based on the characteristic peak of phosphomonoester (Mono-P), which was key to accurately characterize constituents of Po.

4 Research progress on the chemical composition and structural characteristics of Po in the multi-media of surface water

4.1 Core techniques for characterizing the compositional structure of Po

Solid- and liquid-state ^{31}P NMR spectroscopy, Fourier transform infrared spectroscopy (FT-IR), ultraviolet (UV) spectroscopy, enzymatic hydrolysis, and Fourier transform–ion cyclotron resonance–mass spectrometry (FT–ICR–MS) were developed to characterize the chemical composition of Po in the surface water environments. Among these technologies, liquid-state ^{31}P NMR spectroscopy has been widely used to characterize the chemical components of Po in the multi-media (Feng et al. 2016a; b; Yuan et al. 2019; Liu et al. 2017; Copetti et al. 2019), because it has several advantages, such as the requirements of a small sample volume and simple pretreatment method and the fact that it does not damage the sample structure. Moreover, this technique can precisely separate Po components, which include inorganic P and Po. Inorganic P includes orthophosphate, pyrophosphate, and polyphosphate, and Po includes Mono-P, phosphodiester (Di-P), and phosphonate.

Mono-P, the main component of Po, is widely distributed in various water environmental media and accounts for approximately 90% and 80% on average of the total Po in aquatic plants and algae, respectively (Feng et al. 2016a; b). A combination of ^{31}P NMR spectroscopy and enzymatic hydrolysis can provide a new research approach to identify components of Po and their bioavailability in aquatic plants and algae (Feng et al. 2018; Liu et al. 2016a, b). Additionally, it can assist in analyzing the chemical characteristics of Po in waterbodies and particulate matter (Bai et al. 2017; Feng et al. 2020; Zhang et al. 2019). Moreover, this technique plays a key role in studying the composition of Po and its behavior in sediments during eutrophication (Yuan et al. 2020; Zhu et al. 2013; Ni et al. 2019b). The Mono-P content in such sediments is 7.9–78.3 mg/kg, dry mass; that is, it is the main component of Po in sediments (Zhu et al. 2015a, b; Bai et al. 2009; Cade-Menun 2005; Markarov et al. 2002).

The biochemical and eco-environmental functions of Mono-P differ depending on the media. The specific Mono-P components in the natural environment are α and β-glyceride, inositol phosphate, nucleotides, glucose 6-phosphate, glucose 1-phosphate, choline phosphate, and some decomposition products of ribonucleic acid (Smernik and Dougherty 2007; Doolette et al. 2009; He et al. 2011). Generally, the proportion of unknown Mono-P components to total P is 9.7–24.2% (Feng et al. 2016a; Doolette et al. 2009). Because of the overlapping signal peaks of some similar phosphate groups, identifying Mono-P components is difficult through liquid-state ^{31}P NMR spectroscopy. Previously, before deconvolution analysis was used to identify and characterize the structures of most chemical components of Mono-P, standard samples were added to correct the chemical shift of Mono-P during ^{31}P NMR spectroscopy (Feng et al. 2018). However, some spectral peaks of Mono-P still overlapped, so that not all the components could be analyzed based solely on standard spiking experiments. To address this problem, Mono-P in soil samples was determined using high-resolution two-dimensional hydrogen P NMR spectroscopy (2D^{1}H-^{31}P NMR), which identified the NMR absorption signals on two independent frequency coordinates, H and P spectra. Signals of chemical shifts can form a 2D NMR plane map, making characterization of the molecular structure of complex Mono-P components more intuitive, clear, and reliable (Vestergren et al. 2012).

Presently, most qualitative and quantitative analyses of Mono-P in the environmental media have considered the compound as a whole, and improvements are needed to characterize the molecular structure of each component of the Mono-P (Ni et al. 2019a; Zhu et al. 2018). To obtain accurate and reliable results from the qualitative and quantitative analyses of Mono-P in multi-media, such as waterbodies, sediments, soils, and algae, 2D^{1}H-^{31}P NMR spectroscopy was applied together with pretreatment techniques that have large extraction volumes, high resolutions, and do not damage sample structures (Vestergren et al. 2012). To date, the application of such a high-end technology to characterize the Po structure in the surface water environments has not been reported. Widespread application of this technology in the future will provide reliable and accurate characterization and identification of the compositional structure of Po in various environmental media, thus laying the foundation for further research on the interfacial behaviors and migration and conversion processes of Mono-P in multi-media.

In recent years, FT–ICR–MS, as an emerging high-end technology in the field of environmental sciences, has been used to accurately determine the mass, calculate the molecular formulas, and estimate the structures of Po in environmental samples, natural organic matter, proteins, and other biological macromolecules (Michael 2019; Miranda et al. 2020). During FT–ICR–MS, ion cyclotron motion is used to obtain current signals that can assist in determining the molecular charge-to-mass ratio (m/z) of dissolved organic matter (DOM) and dissolved organic phosphorus (Bai et al. 2009; Cade-Menun 2005; Markarov et al. 2002; Smernik and Dougherty 2007). This allows characterization of the molecular compositions of substances. Two visualization methods have been used while analyzing the FT–ICR–MS data: van Krevelen (VK) diagram and Kendrick-analogous mass difference network (KAMDN) diagrams (Yong et al. 2022).

The VK (or paradigm) diagram is a scatter plot of the molecular formulas of all samples, with molecular O/C and H/C values as the abscissa and ordinate, respectively (SI. Figure 2; Li et al. 2022a; Gao et al. 2021; Yang et al. 2022). Molecules in the organic matter, such as moieties including CHO, CHNO, CHOS, CHNOS, CHOP, are categorized by different diagram types according to their different O/C and H/C ratios (Liu et al. 2020; Qi et al. 2022). Patterns of these molecules can also be analyzed based on their occurrence frequency (She et al. 2021). For the KAMDN grid diagram, the mass errors between the actual and standard samples and the differential molecular units between the homologous molecular formulas are considered as the nodes and node connections, respectively. The diagram represents connections between homologous molecular formulas; additionally, it can be used to determine the molecular structure of organic matter and predict its function by analyzing the pendant groups in the homologous molecules of organic matter (Young et al. 2022; Tziotis et al. 2011). However, identification of Po is still challenging because of the

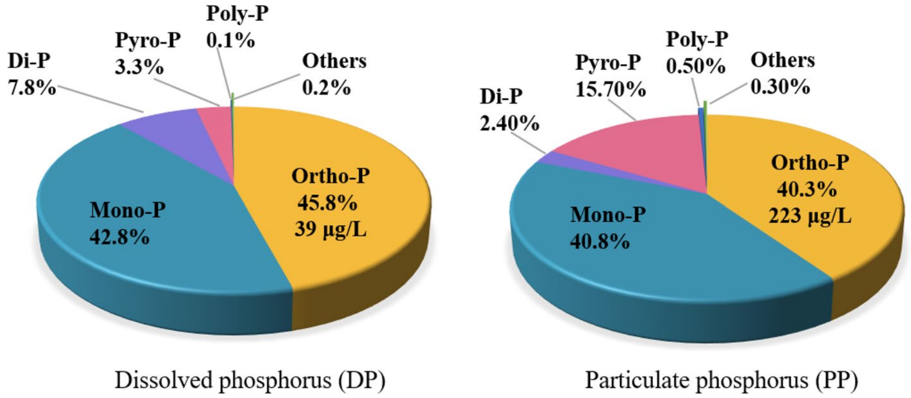


Fig. 5 Distribution of the composition, characteristics, and differences between DP and PP in 10 typical lakes and reservoirs in China (Ortho-P: orthophosphate; Mono-P: phosphomonoester; Di-P: phosphodiester; Pyro-P: pyrophosphate; Poly-P: polyphosphate)

close mass between 12C + 35Cl and 16O + 31P (delta mass = 0.18 mDa) (Fu et al. 2020; Li et al. 2022b; Qi et al. 2022), suggesting that the monoisotopic peak assigned to CHOP formula can also be assigned to a CHOCl formula within a typical mass error (e.g. 1.0 ppm). If the monoisotopic peak is of enough intensity, we can determine the true positive formula based on 13C and 37Cl isotopic pattern. However, there is still no satisfactory formula assignment method for low-intensity monoisotopic peaks. Moreover, there is still a paucity of linking FT ICR-MS results with ^{31}P-NMR results. These weaknesses are all the future directions, and this topic needs to be further studied and expanded.

4.2 Qualitative and quantitative characterization of the chemical composition and structure of Po

Po includes Mono-P, phosphodiesters, and phosphonate. Both DP and PP contain high amounts of Mono-P, but DP contains more phosphodiester molecules than PP (Feng et al. 2020). Moreover, Ortho-P and Mono-P are predominant in both DP and PP, cumulatively accounting for more than 40% of the total P (Feng et al. 2020). The amount of pyrophosphate in PP is five times that in DP (Fig. 5). Ten freshwater lakes with different trophic status have varying compositional structure and distribution of DP and PP as follows: moderately eutrophic lakes (mainly Ortho-P and Mono-P) > light eutrophic lakes (mainly Mono-P) > mesotrophic lakes (mainly Ortho-P) (Zhang et al. 2019). In moderately eutrophic (heavily polluted) and light eutrophic (moderately polluted) lakes, the concentration of Mono-P is high (Feng et al. 2020), and the unstable Mono-P may be hydrolyzed to Ortho-P under the action of alkaline phosphatase. At the same time, Ortho-P is the most direct source of nutrients for algae and phytoplankton, and excessive Ortho-P leads to algal blooms in water ecosystems. The results indicated that Ortho-P and Mono-P should be emphasized in eutrophic lakes, especially in heavily polluted lakes. After completely considering the variations in lake and reservoir regions and trophic types, it is recommended that lakes and reservoirs be suitably classified to ensure effective implementation of control strategies for eutrophication and that water quality standards be formulated by region.

Algae and aquatic plants contain 22 forms of P with Mono-P and phosphodiesters being predominant in Po, along with some glycerides and adenylate (Fig. 6). Mono-P accounts for 80% and 90% of Po in algae and aquatic plants, respectively (Feng et al. 2018). Similar amounts of Po have been detected in particulate matter and algae, indicating that algal residues are the main source of lacustrine particulate matter (Figs. 5 and 6). Interestingly, the Po components vary substantially among organs of aquatic plants. Specifically, leaves contain more fatty acids and aromatic substances compared with rhizomes (Liu et al. 2018). Furthermore, Liu et al. (2018) detected phytate P for the first time in *Nymphoides peltatum*, a phytoplankton. Phytate P, a cyclic compound containing six phosphate groups, chelates with divalent and trivalent metal elements, such as Ca, Mg, Zn, and Fe, to form insoluble compounds. Previously, only phytate P was detected in sediments, but its source was unknown; however, presently, it is speculated that phytate P might have originated from certain types of plants (Liu et al. 2018).

4.3 Comparative analysis of the compositional structure of Po in various environmental media

Mass concentrations of Po and P components were compared in the different environmental media. The mass concentration of Po in algae was 154,000 times that in DP, 5 times that in aquatic plants, and 95 times that in sediments. Moreover, the mass concentration of Mono-P in algae was 174,000 times that in DP and PP, 6 times

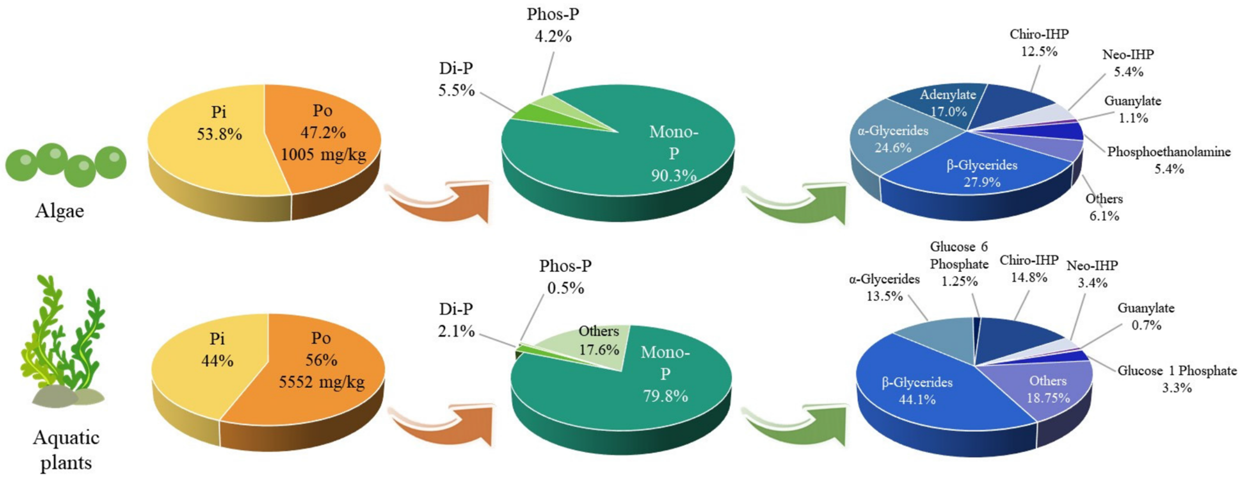


Fig. 6 Composition and structural characteristics of Po in algae and aquatic plants of lakes and their differences (Pi: Inorganic phosphorus; Po: organic phosphorus; Ortho-P: orthophosphate; Mono-P: phosphomonoester; Di-P: phosphodiester; Pyro-P: pyrophosphate; Poly-P: polyphosphate; Phos-P: phosphonate; Chiro-IHP: Chiro-Inositol phosphate; neo-IHP: neo- Inositol phosphate)

that in aquatic plants, and 108 times that in sediments (Fig. 7). Further, the proportion of Po to total P in the five environmental media was 7–56%. Therefore, comprehensively comparing the compositional structure and degradation of P in various environmental media can assist in understanding the sources of endogenous P pollution and provide an important scientific basis for using Po to address aquatic pollution and restore waterbodies.

5 Research progress on the bioavailability of Po in the environments

5.1 Bioavailability of Po in algae and aquatic plants

Assessments of bioavailability of algal Mono-P in lakes led to the detection of 17 P components through algal enzyme culture, of which 15 were Po components. Moreover, 54.6% of the algal Po (mean 3,033 mg/kg, dry mass) could be hydrolyzed by enzymes to release Ortho-P (Feng et al. 2018). Mono-P can increase Microcystis biomass and lipid productivity and promote metabolic activities of *Chlorella*. This is due to the increase in ATP synthase, which is involved in photosynthesis, thereby accelerating the accumulation of intracellular lipids (Chu et al. 2019). Furthermore, the degradation of active Mono-P and phosphodiester promotes algal growth, thus sustaining eutrophication (Ni et al. 2019b).

Mono-P and pyrophosphates are degraded under the actions of various enzymes (alkaline phosphatase, phosphodiesterase, phytase). Active Mono-P is predominant in algae. Approximately 55% and 58% of Po in algae and aquatic plants, respectively, is converted to Ortho-P. The release rate of Ortho-P resulting from algal decomposition is approximately four times that from aquatic plants. Simultaneously, Mono-P and polyphosphate of algal tend to accumulate in the cells of organisms and provide energy for cellular metabolism. Moreover, the bioavailability of these P components is large when algae decompose, which provides important evidence for the direct and rapid supply of nutrients for algal blooms (Feng et al. 2016a; b; 2018). Therefore, the urgent need to manage endogenous Po while addressing water pollution increases the difficulty in implementing suitable management strategies.

5.2 Bioavailability of Po in sediments

Significant progress has been made in the application of liquid-state ^{31}P NMR spectroscopy to study the bioavailability of Po in lacustrine sediments during enzymatic hydrolysis and photodegradation. For example, alkaline phosphatase, which is widely distributed in lakes, was found to improve the degradation rate of phosphomonoester in lacustrine sediments (Zhu et al. 2018). Moreover, photodegradation of Po can potentially replenish P in shallow lakes during the suspended release of sediments (Scigelova et al. 2011). Furthermore, when compositions and enzymatic hydrolysis characteristics of Po in the sediments of key lakes in 10 different regions in the world were analyzed, it was found that the eutrophication of lakes in China and Sweden was more serious. The concentration of Ortho-P ranged from 50.2% to 81.8% of TP in sediments, and the concentration of phosphomonoester ranged from 11.6% to 29.5% of TP (Torres et al. 2014; Zhang et al. 2013a, b; Reitzel et al. 2007; Zhang et al. 2015; Shinohara et al. 2012; Özkundakci et al. 2014; Ma et al. 2022a, b; Giles et al. 2013; Zhang et al. 2013a, b). However, the concentration of phosphomonoester ranged from 48.1% to 65.6% of TP

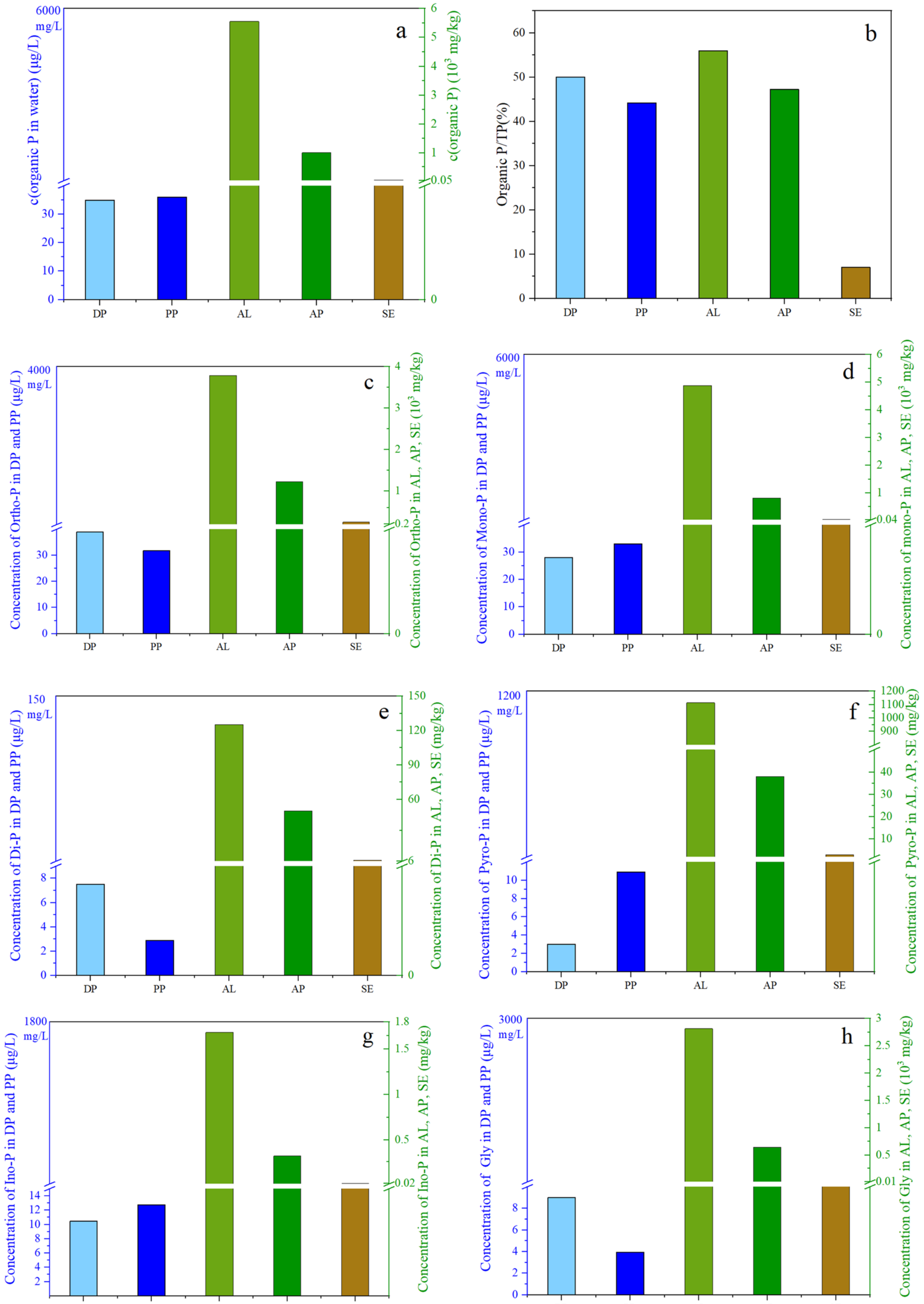


Fig. 7 Comparison of the compositional structure of Po in various environmental media in lakes and their differences (DP: dissolved phosphorus; PP: particulate phosphorus; AL: algae; AP: aquatic plants; SE: sediments; Po: organophosphorus; TP: total phosphorus; Mono-P: phosphomonoester; Di-P: phosphodiester; Pyro-P: pyrophosphate; Gly: glyceride; Ino-P: inositol phosphrus)

in less eutrophicated lakes (i.e., Champlain Lake) (Torres et al. 2014; Zhang et al. 2013a, b; Reitzel et al. 2007; Zhang et al. 2015; Shinohara et al. 2012; Özkundakci et al. 2014; Ma et al. 2022a, b; Giles et al. 2013; Zhang et al. 2013a, b). According to the measurement results, the total Po content in sediments was 7–284 mg/L, which was greater in eutrophic lakes, such as Lake Dian, than in sediments, although 70.8% of the Po was bioavailable. Notably, sediments were an important endogenous source of P for lakes and thus, their contribution could not be underestimated because the total P content in sediments is large and Po does not exist as an inert substance (Fig. 8).

Changes in profiles in sediments indicated that the distribution of Po in sediments was closely related to the eutrophication history of the lacustrine waterbody. Furthermore, the overall degradation rate increased with increasing burial depth, with active Mono-P and phosphodiesters being largely degraded. This indicated that sediments might serve as a continuous source of available P to the overlying waterbody. Simultaneously, the Po content in sediments that was hydrolyzable by enzymes increased continuously during the eutrophication stage, which might be one of the important factors responsible for a sustained P source for cyanobacterial outbreaks (Zhu et al. 2018).

6 Research progress on the source analysis of Po and its biogeochemical processes in the environments

6.1 Tracing sources of Po in the environments

In recent years, phosphate oxygen isotope ($\delta^{18}O_p$) has been extensively used as a geochemical tracer to determine sources and cycles of P in nature (Elsbury et al. 2009; Defforey and Paytan 2018). Phosphates originated from various sources have significantly different oxygen isotopic compositions. Exchanges between phosphate and oxygen in water occur under the influence of enzymatic catalysis before equilibrium fractionation is reached. $\delta^{18}O_p$ indicates the degree and rate of P bioavailability (Yuan et al. 2019; von Sperber and Kries 2014) and can be used for comprehensively understanding the mechanism and kinetics of enzyme-catalyzed reactions (Chilakala et al. 2019). During degradation, Mono-P might combine with exogenous oxygen to form phosphate. Hence, the influence of exogenous oxygen and isotopic fractionation should be considered while analyzing $\delta^{18}O_p$. Therefore, the key when using the $\delta^{18}O_p$ technique for tracing the source of Mono-P is to identify the characteristics of oxygen isotopic fractionation under the UV, photocatalytic and microbial degradation and other paths (Jaisi et al. 2017; Wang et al. 2022).

In various media, Mono-P undergo chemical degradation through various pathways. For example, its degradation is accelerated under the action of ultraviolet light. Moreover, alkaline phosphatases from different sources have been used to catalyze degradation of various phosphomonoester substrates, such as α-D-glucose-1-phosphate, β-glyceride, and 5-AMP, in oxygen isotope-labeled water. The slope of the $\delta^{18}O_p$ and $\delta^{18}O_w$ linear fitting lines for the degradation of the different substrates to phosphate was 0.23–0.2878 (Liang and Blake 2006). Thus, the use of both $\delta^{18}O_p$ and enzymatic hydrolysis could lead to a better analysis of the source characteristics of Mono-P in the various media of environments.

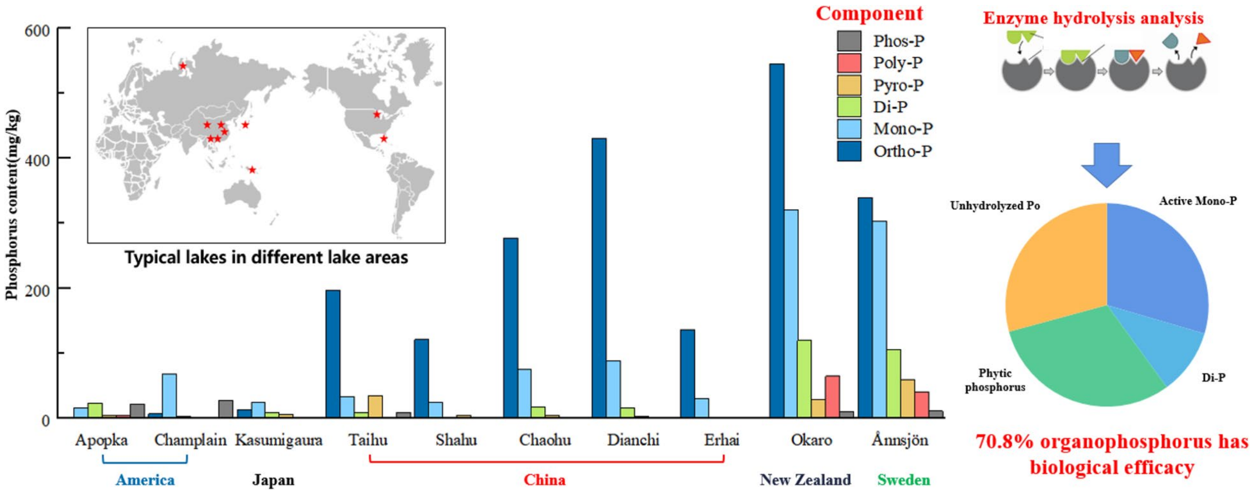


Fig. 8 Composition and structural characteristics of Po in sediments and its bioavailability (Data from Zhu et al. 2018; Torres et al. 2014; Zhang et al. 2013a, b; Reitzel et al. 2007; Zhang et al. 2015; Shinohara et al. 2012; Özkundakci et al. 2014; Ma et al. 2022a, b; Giles et al. 2013; Zhang et al. 2013a, b)

6.2 Biogeochemical behaviors of Po at the interfaces of multi-media

The waterbody-sediment and waterbody-algae interfaces are sensitive to changes in the physicochemical properties of nutrients, and reflect biopositive effects. Conversion, circulation, and exchange of nutrients between the interfaces actively occur in these areas. Interfacial effects and migration and conversion of constituents of P have been published, which have focused on the binding form of inorganic P and its interfacial migration activities (Fan 2019). However, the interfacial migration and conversion behaviors of Mono-P in the waterbodies, sediments, and algae media have been rarely studied. The interfacial behaviors of Po in environments include oxidation–reduction, adsorption–desorption, suspension–subsidence, and degradation–complexation. These behaviors result in strong interactions among organic matter, minerals, heavy metals, and toxic and harmful particulate matter in environments. The surrounding environmental conditions also considerably influence the interfacial behaviors. For example, a conceptual model of sediment P behavior showed obvious P desorption when salinity exceeded 6 ppt, with the desorption of Po being greater than that of inorganic P (Tye et al. 2018).

Degradation of Po is the most important P regeneration pathway (Tye et al. 2018). HCl-P is dominant when sediments are under high salinity stress, but NaOH-P dominates during sediment dredging (Copetti et al. 2019). The main NaOH-P and HCl-P sources are anthropogenic inputs, whereas both in situ biological and anthropogenic inputs are important sources of Po components (Copetti et al. 2019). Moreover, sediment dredging facilitates release of Po into its overlying waterbody (Copetti et al. 2019), and the release of P components is also accelerated under the influence of extracellular enzymes, resulting in the formation of an anaerobic environment in waterbodies (Li et al. 2016).

During the sediment resuspension process, the steady-state concentration of hydroxyl radicals (OH) increases with the increasing nutrient levels. In other words, the greater the level of eutrophication, the stronger the steady-state concentration of OH produced during sediment resuspension. DOM, nitrates, and Fe^{3+} are main photosensitizers that generate OH, leading to the release of additional soluble phosphates, which in turn accelerates eutrophication (Guo et al. 2020). Additionally, microorganisms promote the chemical conversion (degradation and mineralization) of Po in the water environments. Genetic diversity and gene expression enable the microorganisms to adapt to harsh environments and participate in P cycling under various environmental conditions (Mackey et al. 2019). Moreover, in the presence of suitable biochemical pathways to cleave the molecularly stable carbon–P bonds, Po can serve as an alternative source of inorganic phosphates (Ulrich et al. 2018).

7 Factors affecting the migration and conversion of Po in the water environments and its ecological effect

7.1 Main factors affecting the chemical composition, migration, and conversion of Po

The compositional structure, migration, conversion, and interfacial behaviors of Po in water environments are complex and affected by multiple factors (Guo et al. 2020). For example, the chemical properties of Po in sediments and its interfacial behaviors are affected by terrestrial input and runoff characteristics. The relationship between Po and environmental conditions has been analyzed using redundancy analysis and the geochemical indicator method, and based on the spatial distribution of Po in sediments on a 4,000-year temporal scale; the findings indicated that the gradient of weather conditions presented by latitudinal zonality affected Po accumulation through precipitation and temperature (Lü et al. 2018).

Furthermore, migration and conversion of Po is related to seasonality. For example, Po in the Grand River, Ontario Canada continuously provided bioavailable P rather than inorganic Ortho-P to lacustrine waterbodies in winter and spring (Shinohara et al. 2018). Moreover, frozen lakes in the Northern Hemisphere always warmed the fastest in the immediate or following month after the melting of ice (Li et al. 2022b). Endophytic algal debris is the main contributor to sediment Po (P_o) pollution, with humic-bound Po (HA-Po) and humic-acid-bound Po (FA-Po) serving as active Po sources in the sediments (Ma et al. 2022b; Ni et al. 2019a). In a study on the differences in the Po compositional structure between lakes in the eastern China plains and in the Yunnan-Guizhou Plateau, it was found that lakes with higher trophic levels exhibited greater degrees of humification and larger molecular weight of Po compounds; these caused a certain resistance to biodegradation, thereby reducing the risk of Po in sediments being released. Moreover, large amounts of alkaline phosphatase in lakes could degrade active Po, providing important nutrients for algal growth (Ni et al. 2019a; b). The vigorous growth of algae led to the increase of the concentration of chlorophyll-a, and it was positively correlated with the concentration of Mono-P in DP and PP, indicating that Mono-P had bioavailability (Zhang et al. 2019). Previous study also found that pH can control the synthesis of pyrophosphate and polyphosphate in water ecosystems (Ni et al. 2020). It has been shown that the environmental factors which influence polyphosphate storage are the phosphate concentration, light intensity and temperature (Brown and Shilton 2014). Mono-P/PP showed a positive correlation with chlorophyll-a, TP and trophic state index, which suggested that particulate Mono-P possessed a potential contribution to lake eutrophication (Zhang et al. 2019; Feng et al. 2020). In addition, bacteria and phytoplankton were also important factors affecting P (Faithfull et al. 2011).

After comprehensively understanding the effects of multiple factors on the migration and conversion of Po, structural equation modeling (SEM), an important statistical method for systematically analyzing large data, was used to further study the water environments (Li et al. 2019a, b, c; McArdle et al. 2003; Gao et al. 2022). Studies on eutrophication have indicted the role of several factors (such as water depth) that affect P accumulation and other nutrients and the outbreaks of algal blooms; moreover, in some cases, individual factors, which are not completely independent, influence mutually (Qin et al. 2020). Thus, using only statistical methods to analyze the effects of multiple factors on nutrient accumulation or algal bloom outbreaks is not sufficiently reliable. However, complete data under natural conditions are lacking, making it difficult to obtain the requisite independent variables.

Structural equation modeling based on the systemic and causality theories should be used to quantitatively describe the response relationship among multiple factors, such as Po migration and conversion behaviors, and algal bloom outbreaks, along with the mutual influence of various environmental factors. More importantly, the variables in water environments are hierarchically related based on trophic and systemic levels. It can analyze such multi-level data, build models, and simultaneously identify potential correlations between factors, which can assist in analyzing the potential migration and conversion pathways of Po in the water environments. Moreover, it can be combined with indoor simulations and field investigations to further understand the mechanisms responsible for the outbreak of algal blooms, and to predict and estimate the contribution rate of phosphomonoester to endogenous P in various environments, thereby providing a scientific basis for the management and control of water eutrophication (Kenney et al. 2009; Proffitt 2017; Kreiling et al. 2020).

8 Research progress on the interrelationship between Po and organic matter and minerals

Statistical analysis of large data sets on Po and environmental factors indicated that morphology of Po was closely related to sources of organic matter (Hu et al. 2022b). When organic matter is derived from algae, bioavailability of Po is greater, which in turn favors degradation and recycling of Po. However, Po derived from terrestrial and aquatic plants has relatively poor bioavailability and tends to be buried in sediments. Based on experimental studies, possible mechanisms for the interactions between Po and organic matter in sediments have been proposed (Zhu et al. 2018). One mechanism includes the formation of macromolecular Po compounds through covalent bonds with organic matter, while the second mechanism involves the formation of complexes with metal ions.

The combination of Po with organic matter reduces Po bioavailability and promotes its rapid burial and early diagenesis. In particular, Po strongly interacts with humic acid of macromolecular organic matter, which explains why terrestrial and aquatic plants are more likely to be deposited and buried than algae. Influences of minerals on P components varied greatly, which resulted in a 10% and 90% reduction in the bioavailability of phosphomonoester and phytate P, respectively, thereby increasing their preservation and burial time in sediments (Zhu et al. 2018). Therefore, minerals considerably affect the migration and conversion behaviors of Po, and greatly limit the bioavailability of Po sourced from terrestrial and aquatic plants. A combination of solid- and liquid-state NMR reveals that several Po compounds bound to metals and other minerals could not be extracted from suspended and surface sediments.

Po generally undergoes photolysis; however, it is hydrolyzed under alkaline conditions. Unstable Po is significantly positively correlated with organic matter and has a large potential for release. Po, polyphosphate, and pyrophosphate, which were predominant in the cyanobacteria of Lake Dian, were released into the waterbody through chemical and biodegradation, which exacerbated eutrophication (Li et al. 2019a, b, c; Xie et al. 2019; Jin et al. 2019). Po potentially increases the bioavailability of P (Kleber et al. 2021; Qu et al. 2019; Ji et al. 2022). Mono-P, the main component of Po, significantly correlates with HA-P_o and strongly interacts with minerals. The rapid decomposition of phosphodiesters increases accumulation of Mono-P. Simultaneously, conversion of Po to dissolved reactive phosphorus accelerates under the action of alkaline phosphatase, especially high algal biomass where metabolic processes (hydrolysis and mineralization) occur more rapidly (Yuan et al. 2020). Other than humic substances, minerals also inhibit Po adsorption, especially phytate-like P, which is not easily hydrolyzed by enzymes. Thus, the interactions of Po with organic matter and minerals in sediments significantly influence the biogeochemical cycle of Po (Zhu et al. 2018).

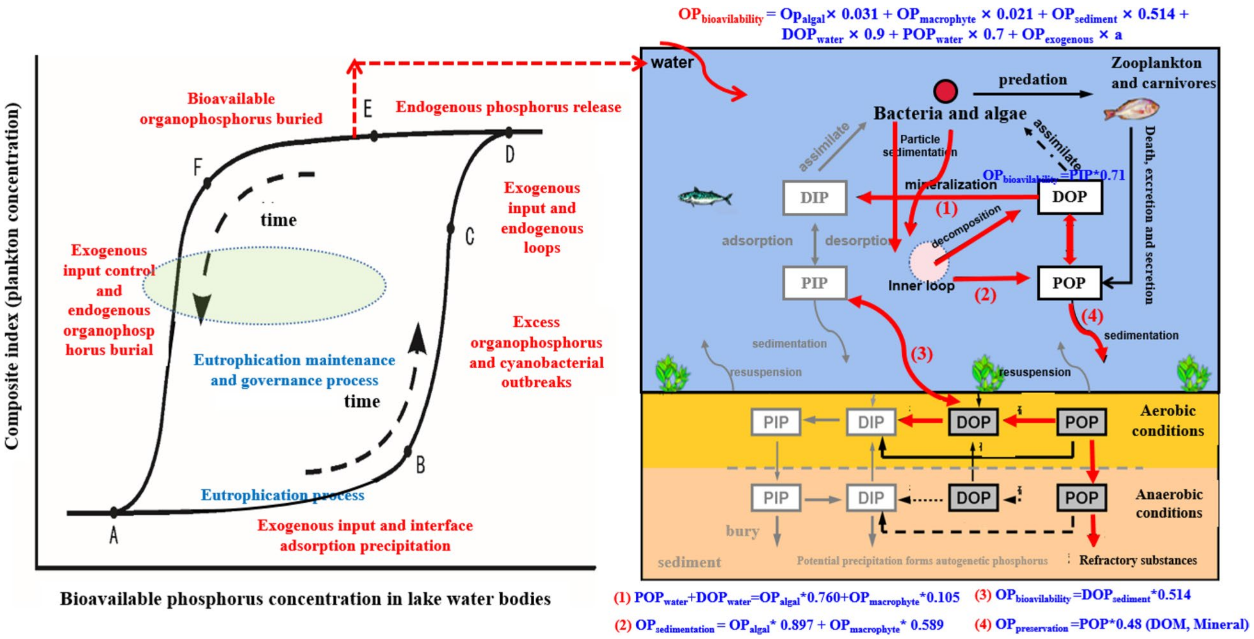


Fig. 9 Theoretical model for the biogeochemistry of Po in lakes

8.1 Model for the biogeochemical cycle of Po in the environments

A quantitative model of the typical biogeochemical cycle of Po in lakes was proposed based on previous studies on Po in lakes, such as Lakes Tai, Chao, and Dian, in China and previous conceptual models (Fig. 9) (Feng et al. 2020; Ma et al. 2022a, b; Zhang et al. 2019; Zhu et al. 2018; Liu et al. 2018). The scheme on the right side is the theoretical model of the biogeochemical cycle of P proposed by previous scholars (Fig. 9). The red arrows in the map indicate new paths for the quantitative conversion of Po, as proposed by the research team led by Fengchang Wu. The model was developed to (i) determine the proportion of algae and aquatic plants that died and get buried, and the factors influencing the proportion of these biological samples that can be converted to increase Po bioavailability, (ii) to quantify DP and PP and determine the key parameters influencing them, and (iii) to improve key influencing parameters, such as conversion path and ratio, for Po bioavailability and organic matter and minerals. To date, this is a relatively complete theoretical model of Po biogeochemistry in lakes that can quantitatively characterize Po migration and conversion behaviors. The model serves as an important reference to understand the current global P biogeochemical cycle and improve other internationally accepted theories of the cycle.

Contributions of Po, derived from different media, to cyanobacterial blooms in Lake Tai have been quantitatively evaluated based on the theoretical model, sediments, algae, and aquatic plant data of the lake, and the corresponding results of simulation experiments (Fig. 10) (Zhu et al. 2013, 2018; Feng et al. 2018; Zhang et al. 2019). The findings indicated that the Po derived from algae, sediment, aquatic plants, and PP and DP in the water body contributed 76%, 13.4%, 10.5%, and < 0.1% to cyanobacterial blooms, respectively. Moreover, 48.7% of Po in the sediment tended to be buried for a long time. These results indicated that Po plays a critical role in the eutrophication of lakes. Accordingly, a relationship between key Po indicators and the level of lacustrine eutrophication was proposed. Thus, as the importance and contribution of Po in controlling pollution in lakes is far more than that gleaned from previous knowledge, understanding of this aspect should be prioritized on an urgent basis.

9 Conclusion and future prospects

The review on understanding the chemical composition, sources, and environmental behavior of Po in various media revealed the following key aspects: (i) In terms of the research paradigm, a novel technical system for characterizing Po components and bioavailability in different environmental media was proposed. (ii) A model for the biogeochemical cycle of Po in lakes was established. (iii) The results of the previous studies provided key references to control endogenous pollution and cyanobacterial bloom outbreaks. Several studies reported that Po is largely abundant and is mostly utilized by organisms.

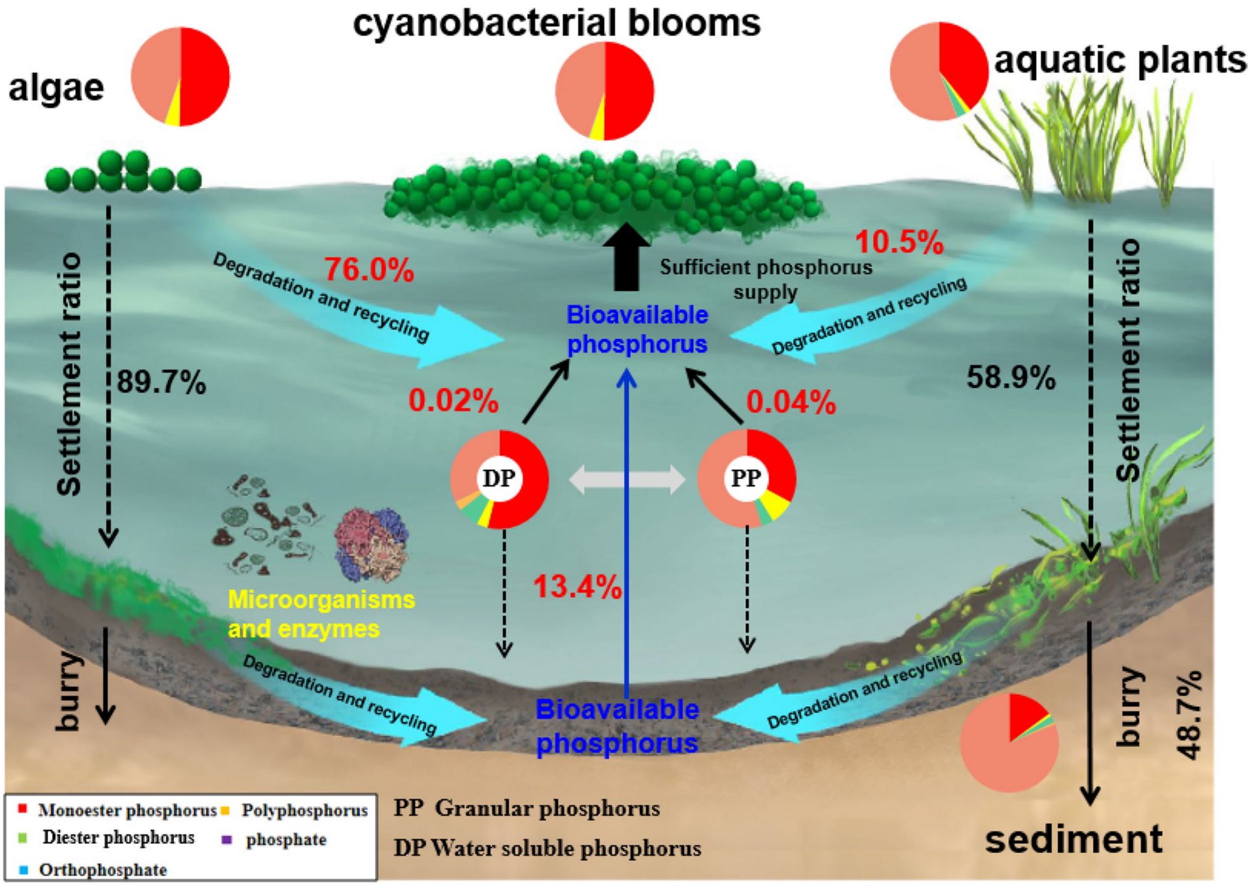


Fig. 10 Contribution of Po in different environmental media to algal bloom outbreaks in lakes and the corresponding cyclic processes

The results of the quantitative cycle model verified these findings. (iv) Regarding meeting the social requirements, the current standards only considered total and inorganic P but not the role, variations, and effects of Po. Furthermore, to control exogenous pollution more effectively, the control of endogenous pollution should be strengthened as well. The challenges of controlling endogenous pollution should be focused upon and accordingly, targeted control strategies should be adopted, especially in the aquatic ecosystem of cold and arid regions. There is still a lack of research on Po. Overall, research on Po is important to manage water environments and ensure efficient decision-making on pollution control. Undoubtedly, current research on Po is still developing. Further studies considering a higher number of waterbody, longer study periods, and additional field experimental conditions are still needed to verify the mechanisms and processes pertaining to Po. Finally, under the context of global climate change, further in-depth research is required to understand its impact on the Po cyclical response and other related aspects.

Abbreviations

P	Phosphorus
Po	Organic phosphorus
N	Nitrogen
Fe	Iron
Mn	Manganese
Ni	Nickel
Co	Cobalt
Cu	Copper
^{31}P NMR	Phosphorus-31 nuclear magnetic resonance
EDTA	NaOH-ethylenediaminetetraacetic acid
DP	Dissolved phosphorus
PP	Particulate phosphorus
FT-IR	Fourier transform infrared spectroscopy
UV	Ultraviolet
FT–ICR–MS	Fourier transform–ion cyclotron resonance–mass spectrometry
2D^{1}H-^{31}P NMR	Two-dimensional hydrogen P NMR spectroscopy
DOM	Dissolved organic matter
VK	Van Krevelen
KAMDN	Kendrick-analogous mass difference network
δ^{18}Op	Phosphate oxygen isotope
OH	Hydroxyl radicals
Po	Sediment Po
HA-Po	Humic-bound Po
FA-Po	Humic-acid-bound Po
SEM	Structural equation modeling
Ortho-P	Ortho-P

Mono-P	Phosphomonoester
Di-P	Phosphodiester
Pyro-P	Pyrophosphate
Poly-P	Polyphosphate
Pi	Inorganic phosphorus
Phos-P	Phosphonate
Chiro-IHP	Chiro-Inositol phosphate
neo-IHP	Neo- Inositol phosphate
AL	Algae
AP	Aquatic plants
SE	Sediments
TP	Total phosphorus
Gly	Glyceride
Ino-P	Inositol phosphorus

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1007/s44246-023-00038-4.

Additional file 1:

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Authors' contributions

W.F. wrote the manuscript, T.W. and Y.Z. analyzed the data and developed the graphical representations, F.S. modified the manuscript, J.P.G. modified and improved the manuscript language, and F.W. modified the manuscript, provided experimental platform and constructive guidance. All authors discussed the results and reviewed the manuscript. The authors read and approved the final manuscript.

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Availability of data and materials

The datasets used or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests

For Editorial board members.

Fengchang Wu is an editorial board member for Carbon Research and was not involved in the editorial review, or the decision to publish, this article. All authors declare that there are no competing interests.

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