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The desorption mechanism of dissolved organic matter on pollutants and the change of biodiversity during sediment dredging

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ABSTRACT

Sediment dredging is an effective means to control the endogenous pollution of lakes, which could significantly change the concentration and composition of organic matter, especially dissolved organic matter (DOM) in the lake. DOM is particularly important for the release of endogenous pollutants, which will inevitably bring an impact on aquatic biodiversity. Nevertheless, in recent research little attention has been paid to the desorption mechanism of DOM on pollutants and the change of biodiversity during dredging. This study investigated the physicochemical properties of DOM in the sediment by taking a sediment dredging project in Dianchi Lake in China for example. The correlations of DOM properties with the desorption behavior of nitrogen (N), phosphorus (P), cadmium (Cd), lead (Pb) and the biodiversity of aquatic organisms were analyzed. The results show that the aromaticity and humification of DOM were improved after dredging, and the high molecular weight DOM was degraded into low molecular weight substance. The desorption amount of N, P and heavy metals (Cd, Pb) were decreased as the pH values increased. Moreover, NH₄-N promoted the release of Pb²⁺ from DOM, while the release of PO₄³-P was inhibited. Correlation analysis shows that the physicochemical properties of DOM exactly affected the release of N, P, Cd and Pb. It was easier to desorb pollutants with low aromaticity and humification of DOM, leading to a decrease in the diversity of aquatic organisms. This study identified the desorption mechanism of endogenous pollutants in DOM and the ecological risk to aquatic organisms, providing a theoretical basis for the prevention and control of water pollution.

1. Introduction

Lake eutrophication, an environmental problem faced by many countries, is mainly aroused by exogenous and endogenous pollutants. When exogenous pollution is effectively controlled, bottom sediment as the source and sink of pollutants in a lake (Rydin and Brunberg, 1998; Wang et al., 2017), can still release pollutants through convection, molecular diffusion and resuspension, resulting in water quality deterioration and algae bloom (He et al., 2015; Huang et al., 2014). Given that, an environmentally friendly means of lake eutrophication control-sediment dredging emerged. It mainly uses dredgers to dig out the sediment and transport it to the destination for reasonable and effective disposal onshore (Barbosa and de Almeida, 2001). A previous study found that dredging may be a measure to address seriously oxygen

depletion in some hyper-eutrophication lakes (Jing et al., 2013), and can continue declines in environmental contaminants at the basin wide level (Gustavson et al., 2008). Recently, dialogues over the sustainability of dredging practices have risen together with its popularity, highlighting the need for research in assessing its sustainability based on its pollutants control (Manap and Voulvoulis, 2015).

Sediment dredging has been used in several lakes like Taihu Lake in China (Wang et al., 2017) and Sweden's Trummen Lake (Cronberg, 1982), significantly reduced the organic matter, nitrogen (N) and phosphorus (P) in sediments. On the one hand, some scholars have found a decrease in the biomass of phytoplankton in dredged areas, while the biomass of benthic organisms increased in un-dredged areas (Jing et al., 2019). It was also found sediment dredging can promote the change in zooplankton community structure, and reduce the diversity and density of benthic organisms significantly (Zhang et al., 2010). On

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Abbreviations

BIX Biological index Cd Cadmium

Dissolved organic matter DOM DO Dissolved oxygen Electric conductivity EC

Fulvic acid FA FI Fluorescence index Humic acid HA ніх Humification index

Nitrogen Ν

ORP Oxidation reduction potential

P **Phosphorus** Lead

Рb

TOC Total organic carbon TD

The turbidity TN Total nitrogen ΤP Total phosphorus UP water Ultrapure water

the other hand, the physicochemical properties of any residual sediment could be changed by dredging, such as water content, particle size and porosity (Robinson et al., 2005), which are closely related to the transportation of pollutants (Manap and Voulvoulis, 2015). Sediment will release pollutants that bound on sediment particle surfaces and interior matrices when sediments are disturbed (Burton, 2002). As a result, the morphology, composition and structure of matrices in the sediments could be changed greatly by dredging, which means the release of pollutants such as N and P may be affected, and further affect the biodiversity of aquatic organism.

Dissolved organic matter (DOM) act as the main components of sediments' matrices, is an important organic carbon pool in ecosystems with complex composition and structure, and plays multiple roles in the environment (Zhang et al., 2021). It can affect the transformation and distribution of heavy metals in sediment, as well as in the bioavailability and biogeochemical processes of nutrients such as N and P (Lin et al., 2021; Mayora et al., 2018). The correlation between the metal concentration and DOM content in the sediments has been shown by various research teams (Lin et al., 2016; Soares et al., 1999; Yu et al., 2001). The carboxyl and phenolic groups in DOM usually act as weak and strong metal binding sites, respectively (Hur and Lee, 2011). When heavy metals are combined with DOM, the adsorption capacity of sediment for metal ions will be decreased. More metals will be distributed to the water in organic complexes due to the redistribution process of dissolved heavy metals (Ranysivek and Jekel, 2006). The anions generated by the dissociation of DOM could compete for the absorptive sites with phosphate on the surface of sediments, resulting in a decrease in phosphate adsorption (Xie et al., 2019). In addition, the inherent cycle of biological elements can be altered by DOM during dredging, which would, in turn, affect the living environment of benthic organisms and plankton (Tian et al., 2017). Some scholars collected sediments from Donggian Lake to compose sediment-water microcosms for evaluating the effects of dredging on nitrogen cycling (Jing et al., 2013). However, previous studies tended to focus on the release of pollutants in sediment and its effect on biodiversity, but did not thoroughly study the role of DOM in sediment dredging. Therefore, the insights on the mechanisms of pollutant release caused by changes of organic matter composition and on the effect of such changes on aquatic biodiversity during dredging were critical for water quality control.

Dianchi Lake is a typical eutrophic lake in China, and a sediment dredging project has been carried out. Therefore, this study aims to investigate the desorption mechanism of dissolved organic matter on

pollutants and the change of biodiversity during sediment dredging. Firstly, the physicochemical properties of sediment were analyzed to explore the influence of dredging. Secondly, the interaction between sediment's DOM and pollutants, as well as the relationship between DOM and biodiversity were studied. Finally, the effect of sediment dredging on DOM release pollutants and biodiversity was examined. In general, these explorations can provide information about the theoretical basis for the prevention and control of water pollution.

2. Materials and methods

2.1. Sampling sites and water quality indexes

Sediment dredging was carried out from January 2013 to April 2019 in the estuary of the Northern Waihai of Dianchi Lake near Kunming City in China. A total area of 8.955 million m² with 5.039 million m³ sand was dredged.

Three 50 cm core sediments were collected by a columnar sediment sampler, and sixteen surface sediment samples (5–10 cm) were collected by a Peterson XDB0201 sampler (Beijing Zhonghui Tiancheng Technology Co., Ltd, China). Sixteen water samples were collected in August 2020. These sampling sites are located in Northern Waihai (S1, S2, S3, S4, S5, S6, S7, S8), Baofeng Bay (S9, S10, S11, S12) and Baoxiang River (S13, S14, S15, S16) (See Fig. 1). Each sampling site was positioned by a FBD30 Global Positioning System (Beijing TATO Technology Co., Ltd, China) (See Table S1). Samples from these sixteen sites were divided into two parts (dredged and un-dredged) according to the dredging project. Thereinto, S7 and S8 as the un-dredged sites were contrasted with S1, S2, S3, S4, S5 and S6. The un-dredged site S16 was contrasted with S9, S10, S11, S12, S13, S14 and S15.

The pH, dissolved oxygen (DO), electric conductivity (EC) and oxidation-reduction potential (ORP) of the water were measured in situ by a 221 portable pH meter (Shanghai Bant Instrument Co., Ltd, China). The sediment samples were centrifuged at 5000 rpm by a L550 centrifuge (Cence Centrifuge Instrument Co., Ltd, China) in the laboratory. The particle size distribution of sediments was determined by laser particle size analysis using a Mastersizer-2000. TN was measured by the method of alkaline potassium persulfate digestion UV-VIS using a TU1810 double beam ultraviolet spectrophotometer. A 752 UV/VIS spectrometer was used to measure TP and TD. Heavy metals were quantified by the flame atomic absorption (TAS-990 Atomic absorption spectrophotometer). Dichromate titration was used to determine COD.

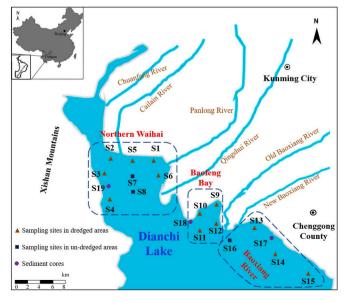


Fig. 1. Locations of sampling sites in Dianchi Lake.

2.2. Biodiversity indexes

The qualitative and quantitative analysis of plankton samples was conducted with the present methods published by Wei et al. (2002). Species identification was based on morphology under a light microscope and zoobenthos identifications were based on standard methods (Thorp and Covich, 2009). The counts were used to calculate biodiversity indexes using the methods detailed in Text S1.

2.3. DOM characterization

The surface sediment samples were dried naturally in a cool place, and then were ground to less than 100 mesh. The extraction and purification of DOM in sediments were based on the base-dissolving acidification method provided by the International Humus Substances Society (IHSS). The HA (molecular weight >1000Da) was obtained by using the SP132638-1 m 1000 Da regenerated cellulose dialysis membranes (Shanghai Yuanye Bio-Technology Co., Ltd, China) treated for 24 h.

The DOM was characterized by using three-dimensional fluorescence spectra (F-7000, Hitachi Limited, Japan) and UV–vis spectra (UV-2600, Shimadzu, Japan). A total organic carbon analyzer (Elementar Vario TOC cube, Elementar, Germany) was used to determine the TOC in DOM. Its C, H, N and S contents were determined by a Vario EI cube element analyzer (Elementar, Germany). These characterizations were used to compare and calculate the properties of different DOM through the indicators detailed in Text S2.

2.4. Desorption experiments

The process of the desorption experiments is diagrammed in Fig. S1. DOM (15 mL) was fully mixed with 15 mL solutions of target pollutants $(NH_4^+-N, NO_3^--N, PO_4^{3-}-P, Cd^{2+})$ for 12 h. The DOM extracted from each surface sediment sample was repeated according to the above experiment process. The concentration of DOM was 90 mgC L⁻¹. The concentration of NH₄⁺-N, NO₃⁻-N and PO₄³-P was 100 mg L⁻¹, and the Cd²⁺ and Pb²⁺ were 20 mg L⁻¹. All of the solutions were made up of ultrapure water (UP water), and their pH was adjusted to 9 (± 0.1) with 0.1 M NaOH or HCl. The desorption experiments were performed in 10 cm regenerated cellulose dialysis bags (Shanghai Yuanye Bio-Technology Co., Ltd, China) placed in containers with 300 mL UP water. Each control experiment contained only corresponding pollutants. The heavy metals took longer to reach desorption equilibrium, and the length of those experiments was extended to 72 h, whereas others reached equilibrium after 24 h. NH₄-N and NO₃-N were measured by the methods of Nessler's reagent spectrophotometry and standard spectrophotometric method with thymol respectively. For orthophosphate, Mo-Sb antiluminosity was used. Flame atomic absorption spectrometry (FAAS) (TAS-990, Beijing Purkay General Instrument Co., Ltd, China) was used for heavy metals.

Sampling site S7 was selected as a model sample in this study. The dosages of DOM and pollutants were the same with the above experiments. Six experiment groups were set for each pollutant, and the pH of the mixture and external solution was adjusted to 4, 5, 6, 7, 8 and 9 (± 0.1). Control experiments contained only one pollutant.

In desorption experiment A, six experimental groups were also set. The concentration of DOM was 135 mgC $\rm L^{-1}$, while the NH $_4^+$ -N and Pb $^{2+}$ were 150 mg $\rm L^{-1}$ and 30 mg $\rm L^{-1}$. After 12 h of mixing, the pH of the mixture and external solution was adjusted to 4, 5, 6, 7, 8 and 9 (\pm 0.1).

In desorption experiment B, the same procedure was used with PO_4^{3-} P and Pb^{2+} . In desorption experiments A and B, the DOM was extracted from S7.

The amount of pollutant desorbed from DOM is calculated as

$$q_t = \frac{c_{out} \times V_{out}}{c_{DOM} \times V_{DOM}} \tag{1}$$

where $q_{\rm t}$ (mg mgC⁻¹) is the amount of pollutant desorbed from DOM at time t (min), $c_{\rm out}$ (mg L⁻¹) is the concentration of external solution at time t (min), $V_{\rm out}$ (mL) is the volume of external solution, $c_{\rm DOM}$ (mgC L⁻¹) is the concentration of DOM and $V_{\rm DOM}$ (mL) is the volume of DOM.

Origin Pro 2020 software was used to fit quasi-first-order, quasi-second-order and Elovich kinetic models. These models were described detailed in Text S3. Pearson correlation analysis was performed, and correlation heat maps were drawn with the aid of Origin Pro (2020) software.

3. Results and discussion

3.1. The quality of water in Dianchi Lake

As is shown in Table S2, the water turbidity (TD) decreased and the transparency increased in the dredged areas compared with those in the un-dredged areas, which also proves that dredging reduced the suspended matter content like Zhang's study (Zhang et al., 2010). The dredging presumably removed a great quantity of mineral ions in the sediment, which could break the dynamic balance between the sediment and the overlying water. Therefore, the water electric conductivity (EC) was decreased after dredging. The oxidation-reduction potential (ORP) in the dredged areas of Baofeng Bay and the Baoxiang River was lower than that in the un-dredged area. However, similar to Zhang's observations, the dredging project had no significant impact on the pH (Zhang et al., 2010). There may be natural buffering substances in the lake, such as HCO_3^- and HPO_4^{2-} (Li et al., 2012).

The indexes of interstitial water are presented in Table S3. Any Cr, Cu, Zn or As was below detectable limits, but total nitrogen (TN), total phosphorus (TP), cadmium (Cd) and lead (Pb) were all detected. The concentration of TN, TP and TOC were higher than that in the undredged areas, which means dredging can release DOM from the sediment and can further desorb pollutants such as N, P, Cd and Pb.

3.2. Physicochemical properties of the sediment

Fig. 2 shows the particle size distributions of the surface sediments from the different sampling sites and Table S4 classifies the particle sizes. The range of particle size at a site was generally less than 250 μm and the largest proportion was silt (59% on average). Most of the samples from dredged areas showed a smaller fraction of particles less than 64 μm than those from the un-dredged ones. That also conforms to Zhang's findings (Zhang et al., 2019). The smaller the particle size of the sediment is, the stronger the adsorption and binding ability of heavy metals will be (Soares et al., 1999). Thus, the dredging project has a great impact on the properties of sediments, which can further affect the transport and transformation of pollutants.

The pollutants in the surface sediments are shown in detail in Table S5, and the pollutants in three columnar samples are shown in Table S6. The concentrations of pollutants in sediments increased with depth before stabilizing. Table S5 shows the TP and TN concentrations increased notably after dredging. The dredging depth of 50 cm was much less than the thickness of the sediment layer, so the middle sediment became surface sediment after the dredging. That led to the high concentrations of TP and TN observed in the sediment.

In Northern Waihai, the concentration of organic matter in the dredged areas was lower than that in the un-dredged areas, which is strikingly different with that at Baofeng Bay and the Baoxiang River. Dredging reduced the concentration of organic matter in the sediment significantly, but the particle size that is greater than 64 μm accounted for about 31.51% of the total (see Table S4) at sampling site S16, showing there contained more sand grains and fewer clay grains, may cause the little organic matter in the sediment. Sampling site S16 was in an estuary where large amounts of sand can accumulate. The concentration of organic matter at Baofeng Bay and the Baoxiang River was

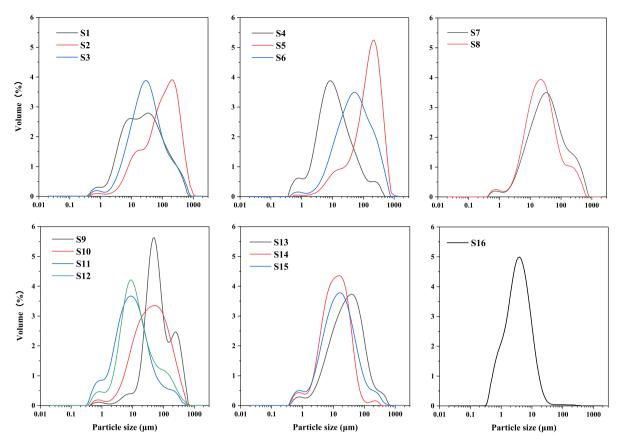


Fig. 2. Particle size distributions of the sediment samples from Dianchi Lake. S1, S2, S3, S4, S5, S6, S7, S8 were the sampling sites in Northern Waihai, S9, S10, S11, S12 were the sampling sites in Baofeng Bay and S13, S14, S15, S16 were the sampling sites in Baoxiang River. Thereinto, S7 and S8 as the un-dredged sites were contrasted with S1, S2, S3, S4, S5 and S6. The un-dredged site S16 between Baofeng Bay and Baoxiang River was contrasted with S9, S10, S11, S12, S13, S14 and S15.

lower than that in Northern Waihai. Much of the organic matter is the remains of algae and aquatic plants that have sunk to the bottom (Li et al., 2018). Southwest winds prevail at Dianchi Lake all year round. That would tend to concentrate organic matter in Northern Waihai, because massive phytoplankton converges due to the action of hydraulic, topographic and wind conditions (Fang et al., 2014).

3.3. The characteristics of DOM

Figure S2 presents the UV–visible spectra of the DOM at pH 7 at a DOM concentration of 5 mgC $\rm L^{-1}$. The SUVA₂₅₄ values ranged from 0.035 to 0.133 and the H/C ratio varied from 0.124 to 0.147 (Table 1).

In Northern Waihai, SUVA $_{254}$ in most of the dredged areas were larger than those in the un-dredged areas, but the H/C ratios were lower than those in the un-dredged ones. The SUVA $_{254}$ values, H/C ratio, FI (0.699–1.417) and HIX (3.112–22.339) indicated that the aromaticity and humification of DOM had been increased after dredging, which means that the DOM in the surface sediment before dredging was mainly newly formed. Dredging helps to remove cenozoic sediments effectively, which are most likely to release pollutants (Yuan et al., 2014). The E_2/E_3 values ranged from 2.422 to 2.935 (all less than 3.5), indicating that the main component of DOM extracted from sediments was HA. The values of E_2/E_4 listed in Table 1 ranged from 4.083 to 5.667. The E_2/E_3 and E_2/E_4 values in the dredged areas were larger than those in the

Table 1The physicochemical properties of DOM.

Area	Sampling site	C/N	H/C	FI	BIX	HIX	Abs_{230}	Abs ₂₈₅	$SUVA_{254}$	E_2/E_3	E_2/E_4
Northern Waihai	S1	8.973	0.138	0.741	0.327	12.117	0.867	0.591	0.070	2.507	4.488
	S2	8.910	0.136	0.710	0.365	7.958	0.770	0.499	0.059	2.570	4.759
	S3	8.965	0.135	0.775	0.390	8.408	0.508	0.325	0.039	2.601	4.702
	S4	8.163	0.140	0.791	0.364	6.609	0.745	0.488	0.058	2.573	4.378
	S5	8.506	0.124	0.699	0.276	22.339	1.699	1.121	0.133	2.734	4.997
	S6	9.022	0.141	0.788	0.427	4.772	0.526	0.328	0.039	2.567	4.382
	S7	8.732	0.146	0.731	0.387	6.607	0.703	0.438	0.052	2.550	4.436
	S8	7.941	0.146	0.763	0.382	7.507	0.660	0.421	0.050	2.512	4.286
Baofeng Bay	S9	7.454	0.139	0.846	0.731	5.084	0.635	0.396	0.047	2.757	5.066
	S10	8.340	0.147	0.806	0.451	3.112	0.470	0.290	0.035	2.422	4.083
	S11	8.450	0.146	0.831	0.387	6.418	0.550	0.351	0.042	2.602	4.443
	S12	8.034	0.144	0.875	0.428	5.820	0.470	0.294	0.035	2.628	4.438
Baoxiang River	S13	8.843	0.146	1.050	0.579	4.113	0.552	0.324	0.039	2.935	5.667
	S14	7.891	0.139	0.760	0.429	4.590	0.557	0.353	0.042	2.628	4.361
	S15	8.860	0.129	0.743	0.359	8.191	0.809	0.521	0.062	2.602	4.638
	S16	7.543	0.139	1.417	0.544	4.593	0.682	0.398	0.047	2.877	5.640

un-dredged areas, indicating that DOM degraded into low molecular weight substances and the degree of polymerization was low after the dredging (Korshin et al., 1997).

At Baofeng Bay and the Baoxiang River, the values of SUVA $_{254}$, E_2/E_3 and E_2/E_4 in the majority of dredged areas were lower than those in undredged areas, but the H/C values were higher. Baofeng Bay and the Baoxiang River were located in the estuary where a large number of inhabitants gathered. Their domestic sewage and industrial wastewater drain into those areas, altering the composition of the DOM in that part of the lake (Tang et al., 2020). This could give rise to the decline of the humification degree of DOM and the accumulation of cenozoic sediments. Sampling sites S13 and S15 were close to the coast and far from

the estuary. Therefore, the physicochemical properties of the DOM were similar to those in Northern Waihai.

Three-dimensional fluorescence EEM spectroscopy was applied to characterize the DOM extracted from each sediment sample. Each EEM gave spectral information about the chemical compositions of DOM samples. One peak was readily identified from EEM fluorescence spectra of the DOM (Fig. S3). This peak has been described as HA peaks (Chai et al., 2012).

3.4. Desorption dynamics

Pseudo-first order, pseudo-second order and Elovich kinetics were

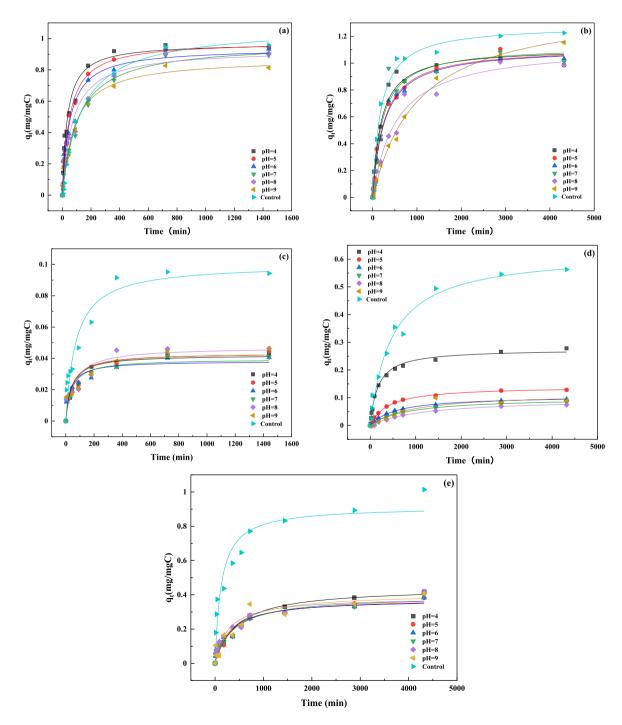


Fig. 3. Desorption curves of pollutants at different pH: (a) NH₄⁺-N, (b) PO₃⁴-P, (c) NO₃⁻-N, (d) Cd²⁺, (e) Pb²⁺.

used to fit the pollutant release process, and the correlation coefficients (R²) (Table S7) indicated that the pseudo-second order model described the desorption well. That means the kind of desorption involved the coordination reactions of carboxyl and hydroxyl groups with the target pollutants (Ho and McKay, 1999). The desorption of NH₄⁺-N, PO₄³-P, NO_3^- -N, Cd^{2+} and Pb^{2+} from the DOM collected from S7 is shown in Fig. 3, the curves indicate the model fit. Desorption is a complex dynamic process with an initial fast phase and a final slow phase, which may be explained by the density functional theory used in previous studies (Gong et al., 2021; Pari et al., 2017). Gong et al. (2021) found DOM can be adsorbed more strongly on the surfaces of α -Fe₂O₃ (001) due to its phenol, carboxyl, amide, and quinone groups, and using density functional theory to unravel atomic/molecular mechanisms during the adsorption process. Furthermore, the process of DOM desorbed ionic contaminants can be well explained by pseudo-first order. Pollutants that were physically absorbed in DOM can be desorbed in a fast initial reaction, while the final slow phase mainly released the pollutants combined with energy-poor covalent bonds or high-energy bonds (He et al., 2015). For NH₄⁺-N and NO₃⁻-N, the fast reaction step was completed within 100 min, whereas the slow reaction step took approximately 300 min. The desorption reached equilibrium in 400 min (Fig. 3a and c). $PO_4^{3-}P$, Cd^{2+} and Pb^{2+} release reached equilibrium in 1500 min (Fig. 3b, d and e).

The state of DOM will change significantly under different pH conditions, and the pollutants will also present different ion forms (Pace et al., 2012). DOM is mainly in the form of positively charged hydrogen ions when pH is less than 7, and it is negatively charged under alkaline conditions (Islam et al., 2020). The desorption capacities of DOM on NH_4^+ -N increased as the pH decreased because of electrostatic repulsion

between the DOM and NH_4^+ . As for the heavy metals Pb and Cd, the desorption capacities of DOM increased with decreasing pH, because the metals' water-soluble forms and exchangeable state are Pb^{2+} , Pb $(OH)^+$ and Cd^{2+} under acidic conditions, and thus can release more Pb^{2+} and Cd^{2+} . This is consistent with the results obtained by Gao's group (Gao et al., 2003). The release rate of PO_4^{3-} -P was faster at pH below 7 and the release quantity was greater at pH above 8. The reason is the electrostatic repulsion between DOM and PO_4^{3-} -P in alkalinity conditions (Schneider and Haderlein, 2016). The amount of P released is of great importance for studying its risk in Dianchi Lake, where the pH of the water is nearly 9. There is also similar strong electrostatic repulsion between DOM and NO_3^- -N, which leads to a strong desorption capacity of DOM on NO_3^- -N (Islam et al., 2020).

Fig. 4 shows the desorption dynamic curves from experiments A (DOM, NH $_4^+$ -N and Pb $^{2+}$) and B (DOM, PO $_4^3$ -P and Pb $^{2+}$). The fitting results are presented in Table S8 and S9. Pseudo-second order kinetics again described the desorption well, including an initial fast phase and a final slow phase. The varying tendencies of NH $_4^+$ -N, PO $_4^3$ -P, and Pb $^{2+}$ under the impact of pH were previously reported. However, the amounts of NH $_4^+$ -N, PO $_4^3$ -P and Pb $^{2+}$ released in the joint desorption experiments were greater than those in the separate desorption experiments.

In joint desorption experiment A, the q_e of Pb^{2+} (from 1.1 to 1.3 mg mgC⁻¹) were higher than those in the separate desorption experiments where they were between 0.3 and 0.4 mg mgC⁻¹ (Figs. 4b and 3e). As for NH₄⁺-N, the q_e ranged from 0.7 to 1.2 mg mgC⁻¹ (Fig. 4a) in joint desorption experiment A, similar to the separate desorption experiments (0.7–0.9 mg mgC⁻¹ (Fig. 3a)). The Pb^{2+} competes for the adsorption sites with NH₄⁺, reducing the number of available binding sites. Meanwhile, there is also ion exchange during the adsorption (Nasrin et al.,

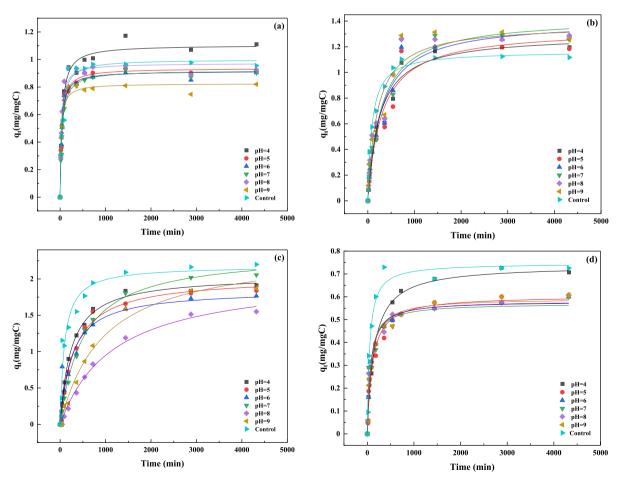


Fig. 4. Desorption curves of pollutants at different pH. Experiment A: (a) NH₄-N, (b) Pb²⁺. Experiment B: (c) PO₄³-P, (d) Pb²⁺.

2019), so the q_e of both Pb^{2+} and NH_4^+ were much higher than those in the separate desorption experiments.

In joint desorption experiment B, the q_e of Pb^{2+} ranged from 0.5 to 0.7 mg mgC $^{-1}$ (Fig. 4d), higher than those in the separate desorption experiments (0.3–0.4 mg mgC $^{-1}$ (Fig. 3e)). The q_e of PO_4^{3-} in joint desorption experiment B (1.5–2.2 mg mgC $^{-1}$ (Fig. 4c)) was also higher than those in the separate experiments (ranging from 0.9 to 1.2 mg mgC $^{-1}$ (Fig. 3b)). Under acidic conditions, the protonation of amido and carboxyl leads to the aggregation of DOM, exhibiting the characteristics of a colloid (Jones and Bryan, 1998). That gives DOM a large specific surface area that can firmly absorb Pb^{2+} and reduce the concentration of Pb^{2+} in the external solution. DOM is negatively charged and much of the Pb^{2+} becomes precipitates at high pH values. As a result, Pb^{2+} had small q_e in both the separate and joint experiments under the action of static electricity and complexation.

As a consequence, it can be found that the q_e of Pb^{2+} in joint experiment A (1.1–1.3 mg mgC $^{-1}$) was higher than that in joint experiment B (0.5–0.7 mg mgC $^{-1}$ (Fig. 4b and d)), showing that NH_+^4 -N promoted the release of Pb^{2+} and inhibited the release of PO_4^3 -P.

3.5. Desorption and DOM properties

Figures S4 to S8 present the desorption dynamic curves of pollutants in DOM from each sample. Fig. 5 shows the correlations between pollutants desorption parameters (NH $_{+}^{+}$ -N (q_{e1} , k_1), PO $_{-}^{3}$ -P (q_{e2} , k_2), NO $_{-}^{3}$ -N (q_{e3} , k_3), Cd $^{2+}$ (q_{e4} , k_4) and Pb $^{2+}$ (q_{e5} , k_5)) and the physicochemical properties of DOM. Parameters of pseudo-first order, pseudo-second order and Elovich desorption kinetics models at pH 9 are shown in Tables S10 to S14.

The q_{e1} showed a moderate positive correlation with BIX (0.48), and a moderate negative correlation with HIX (-0.46). The k_1 had a strong positive relationship with H/C (0.53) and a moderate positive relationship with E $_2$ /E $_3$ ratio (-0.38). A high H/C ratio means that DOM is highly saturated, and a low HIX value indicates little DOM humification. They could lead to fewer adsorbable sites and higher q_{e1} and k_1 (Drexel et al., 2002). The q_{e2} showed only a moderate negative relationship with C/N ratio, whose association degree was -0.43. The k_2 was largely positively associated with the C/N ratio (0.51), and moderately

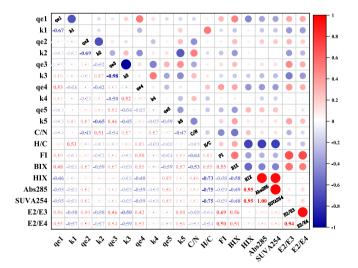


Fig. 5. Correlations between desorption and DOM properties. Blue and red indicate negative and positive correlations, respectively. The size of the circle is proportional to the strength of the correlation. q_{e1} , q_{e2} , q_{e3} , q_{e4} and q_{e5} represent the amounts of NH₄⁺-N, PO₃³-P, NO₃³-N, Cd²⁺ and Pb²⁺ desorbed at equilibration, respectively. k_1 , k_2 , k_3 , k_4 , and k_5 represent the release rate constants of NH₄⁴-N, PO₃³-P, NO₃³-N, Cd²⁺ and Pb²⁺, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

negatively correlated with BIX (-0.39). These indicate that low DOM humification can increase k2 and decrease qe2. There was a medium positive correlation (0.46) between qe3 and E2/E3. As for k3, it had a strong negative relationship with E2/E3 with the correlation degree -0.50, indicating that lower molecular weight and polymerization degree of DOM can release NO₃-N more easily. The main reaction mechanism between NO₃-N and DOM is substitution and adsorption. DOM with a low degree of humification has little carbonyl, hydroxyl and acyl, which could increase the desorption amount of NO3-N (Holmboe and Kristensen, 2002). The qe4 had a moderate positive correlation with FI (0.38), BIX (0.35) and the E_2/E_3 ratio (0.42), while it showed a moderate negative correlation with HIX (-0.40), Abs $_{285}$ (-0.39) and SUVA $_{254}$ (-0.39). However, there was no significant correlation between k_4 and physicochemical properties of DOM. The qe5 and BIX were moderately negatively correlated (-0.39). The k₅ had a moderate negative correlation with the C/N ratio (-0.47), but a moderate positive relationship with BIX (0.37) and the E₂/E₃ ratio (0.38). Perhaps different heavy metals had diverse adsorption sites and characteristics, leading to the differences in the binding ability to DOM (Wang et al., 2021b). Cu and DOM are bound in the form of Cu-DOM-Cu while Pb is combined with DOM in the form of Pb-DOM (Zhao et al., 2018).

These correlations showed that different pollutants had different responses to the physicochemical properties of DOM. Therefore, the influence of those differences on biodiversity should be further explored.

3.6. Biodiversity

The species and quantity of plankton at each sampling site are summarized in Table S15, and the biodiversity indexes for the three regions are presented in Table 2.

Thirty-one genera of phytoplankton were observed under a light microscope. They belonged to six phyla: *Chlorophyta*, *Bacillariophyta*, *Cyanophata*, *Pyrrophyta*, *Euglenophyta* and *Cryptophyta*. The *Chlorophyta* had 14 genera represented, taking up 45.2% of the total species found.

It can be seen from Table 2 that the Shannon-Wiener index (H') and Simpson index (D) in dredged areas were higher than those in undredged areas in Northern Waihai and Baofeng Bay. In the Baoxiang River area, H', D and the Margalef index (M) in dredged areas were basically higher than those in un-dredged areas. The biodiversity dominance index (B) shows the proportion of dominant species, which was higher in dredged areas than in un-dredged areas. All the samples showed that *Microcystis* occupied the absolute dominance among the planktonic algae. The Pielou index (J) in dredged areas was larger than that in un-dredged areas, indicating that sediment dredging had a significant effect on plankton diversity. This is consistent with Zhang's findings (Zhang et al., 2010).

In summary, plankton diversity was generally extremely low, but it was higher in the dredged areas. Sediment dredging can improve ecological diversity.

Table S16 presents similar information for benthic organisms. Six genera of benthic organisms were identified, which belonged to the *Mollusca* and the *Annelida*. The *Mollusca* were represented by 5 genera which took up 83.3% of the species. *Paraprososthenia costata* had absolute superiority in benthic biomass.

Table 2 shows that H', D and M in Northern Waihai were all basically the same, with each index in dredged areas significantly higher than that in un-dredged areas. These indexes were almost the same in the Baoxiang River and Baofeng Bay areas, but each biodiversity index in dredged areas was significantly lower than that in the un-dredged areas. The B and J were generally higher in dredged areas than those in the undredged areas.

So, biodiversity was greater in dredged areas in Northern Waihai but was lower further south. Biodiversity of benthos was generally low everywhere, and no living animal except *Limnodrilus* species was observed at any site. It can be speculated that the damage to the living environment of benthic animals was not only caused by the sediment in

Table 2 Biodiversity indexes.

Area	Sampling site	Plankton biodiversity index	Benthic diversity indexes												
		Abundance (\times 10 ⁴ CFU L ⁻¹)	S	H′	D	J	M	В	Biomass (g m ⁻³)	S	H′	D	J	M	В
Northern	S1	34,924.73	9	1.27	0.48	0.40	0.76	0.68	141.54	1	0.00	0.00	_	0.00	1.00
Waihai	S2	32,156.61	14	1.46	0.58	0.38	1.25	0.50	1946.21	3	1.28	0.55	0.81	0.26	0.55
	S3	24,658.23	14	1.55	0.60	0.41	1.29	0.45	0.00	0	0.00	_	_	_	0.00
	S4	22,531.26	16	1.38	0.53	0.35	1.50	0.61	672.33	2	0.30	0.10	0.30	0.15	0.95
	S5	15,885.20	14	1.70	0.63	0.45	1.34	0.51	1203.11	3	1.40	0.60	0.88	0.28	0.47
	S6	11,502.56	19	1.57	0.56	0.37	1.93	0.59	141.55	2	0.81	0.38	0.81	0.20	0.75
	S7	20,937.35	15	1.41	0.56	0.36	1.41	0.49	636.95	2	0.31	0.10	0.31	0.15	0.94
	S8	14,059.52	14	1.43	0.55	0.38	1.36	0.56	0.00	0	0.00	_	_	_	0.00
Baofeng Bay	S9	20,919.31	14	1.46	0.58	0.38	1.31	0.50	141.54	2	1.00	0.50	1.00	0.20	0.50
	S10	12,509.36	8	1.25	0.53	0.42	0.74	0.58	176.93	1	0.00	0.00	_	0.00	1.00
	S11	8015.99	11	1.62	0.58	0.47	1.11	0.56	141.54	2	1.00	0.50	1.00	0.20	0.50
	S12	11,512.84	16	1.59	0.59	0.40	1.60	0.50	176.93	2	0.97	0.48	0.97	0.19	0.60
Baoxiang River	S13	25,134.60	14	1.24	0.52	0.33	1.28	0.59	636.94	1	0.00	0.00	_	0.00	1.00
	S14	11,188.41	16	1.71	0.61	0.43	1.61	0.48	743.10	3	1.47	0.61	0.93	0.30	0.52
	S15	18,151.20	15	1.50	0.57	0.38	1.43	0.48	3467.80	4	1.06	0.41	0.53	0.37	0.73
	S16	27,279.56	15	1.48	0.58	0.38	1.37	0.46	318.48	4	1.89	0.72	0.95	0.52	0.33

S: the number of species or genus; H': Shannon-Wiener index; D: Simpson index; J: Pielou index; M: Margalef index; B: Berger-Parker index; CFU: Colony-Forming Units.

the lakes but also by the anoxic state of the water column caused by bloom, which led to the widespread death of benthic animals (Wear and Gardner, 2001).

3.7. Diversity and DOM properties

Fig. 6 relates the physicochemical properties of the DOM with the plankton diversity indexes. H/C ratio had a moderate negative relationship with H′ (-0.43) and D (-0.45), respectively. The E_2/E_3 ratio and S were weak positive correlated (0.30). BIX and the E_2/E_4 ratio had a moderate negative relationship with J (-0.35 and -0.31 respectively). HIX had a moderate positive correlation with H′ and J, the association degrees were 0.31 and 0.39, respectively. The remaining properties of DOM had no significant correlation with plankton diversity indexes. These results indicate that the lower the aromaticity and humification of DOM, the easier to desorb N, P and other pollutants. This was consistent with the analysis results in previous parts 3.3 and 3.5.

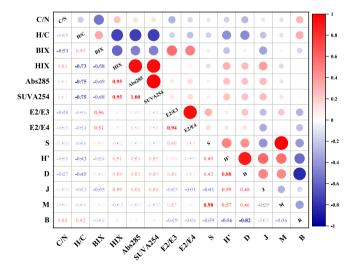


Fig. 6. Correlations between DOM properties and plankton diversity. S is the number of species, H' is the Shannon-Wiener index, D is the Simpson index, J is the Pielou index, M is the Margalef index, B is the Berger-Parker index. Blue and red indicate negative and positive correlations, respectively. The size of the circle is proportional to the strength of the correlation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The release of N, P and other pollutants seems to decrease the number of species present and the diversity of plankton, but its density and uniformity increased. Excessive nutrient levels seem to reduce plankton's biodiversity and simplify its communities (Wang et al., 2021a).

The correlations of the benthic diversity with DOM properties are presented in Fig. 7. The C/N ratio had moderate negative relationships with H', D and J, but a moderate positive correlation with B. The H/C ratio had a strong negative correlation with S (-0.63), and week negative correlations with M, H' and D. BIX showed a moderate negative relationship with B and a moderate positive relationship with J. The E₂/E₃ ratio had significant positive correlations with H' (0.71), D (0.67), J (0.52) and M (0.68), while there was a strong negative correlation (-0.75) between E₂/E₃ and B. Similarly, the E₂/E₄ had strong positive relationships with S (0.57), H (0.68), D (0.60) and M (0.73), and had a strong negative relationship with B (-0.67).

Therefore, the physicochemical properties of sediment's DOM

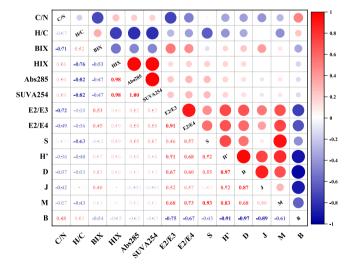


Fig. 7. Correlations between DOM properties and benthic diversity. S is the number of species, H' is the Shannon-Wiener index, D is the Simpson index, J is the Pielou index, M is the Margalef index, B is the Berger-Parker index. Blue and red indicate negative and positive correlations, respectively. The size of the circle is proportional to the strength of the correlation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

influence the diversity of both plankton and benthic organisms, but especially benthic organism. Some scholars suggest that benthic organisms are sensitive to the bottom environment because they live at the bottom of the water column (Knehtl et al., 2021). Furthermore, benthic organisms are more sensitive to nutrient levels than zooplankton (Wang et al., 2021a). They thus better represent the bottom environment.

4. Conclusions

Sediment dredging can significantly change the physicochemical properties of the sediments and further affect the concentration and composition of DOM. Dredging can increase the aromaticity and humification degree of DOM in the sediments, and high molecular weight substances were transformed into low molecular weight substances after dredging. The desorption of all the pollutants studied had an initial fast and a subsequent slow phase. Pseudo-second order kinetics can describe the desorption well. The desorption experiments show that the NH₄-N promoted the release of Pb²⁺ from DOM, while the release of PO₄³⁻-P was inhibited. The properties of DOM can significantly influence the release of N, P and heavy metals. The lower the aromaticity and humification degree of DOM, the easier the release of N, P and heavy metals. This can lead to a decrease in the number and diversity of aquatic organisms, though their density and uniformity may increase. These findings provide new information about the effects of DOM in dredging projects on the lake ecosystem and a theoretical basis for the prevention and control of water pollution.

Declaration of competing interest

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.envres.2022.113574.

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