

Article

The Long-Term Effects of Dredging on Chromium Pollution in the Sediment of Meiliang Bay, Lake Taihu, China

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Abstract: Sediment dredging is a common remediation tool for polluted water bodies. However, the long-term effects of dredging on chromium (Cr) contamination remain unclear. This study was conducted to evaluate the long-term effects of sediment dredging on Cr contamination in Lake Taihu, six years after dredging was performed. In this study, high-resolution equilibrium dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) sampling techniques were used for sampling total dissolved Cr and DGT-labile Cr(VI) at the sediment water interface. The results show that the vertical averaged concentrations of total dissolved Cr in summer ($112.6 \pm 28.8 \mu\text{g/L}$) and winter ($115.3 \pm 29.9 \mu\text{g/L}$) in the non-dredged site were above the fisheries water quality standard (AEPC, 2002). They were 38% lower in overlying water and 20% lower in sediment pore water in the dredged site in winter, while in summer the reduction was not evident. The concentration of total dissolved Cr in the dredged site was significantly higher in spring and autumn than those in the non-dredged site, which was probably caused by the large rainfall and river discharge during the two seasons. The vertically averaged concentrations of DGT-labile Cr(VI) in both the non-dredged and dredged sites did not exceed the drinking water quality standard requirements (WHO, 1993). Modeling of DGT-induced fluxes from sediment into overlying water showed a higher response time (T_c) and lower adsorption rate (k_1) and desorption rate (k_{-1}) in the dredged site except in summer, indicating that sediment dredging decreased Cr mobility in sediments. Overall, these results confirm that sediment dredging decreased the risk of Cr contamination in winter in Lake Taihu.

Keywords: sediment dredging; chromium; diffusive gradients in thin films (DGT); high-resolution equilibrium dialysis (HR-Peeper)



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1. Introduction

Water pollution is a worldwide environmental problem that threatens sustainable social development and human health [1,2]. Water contamination mainly includes heavy metals/metalloids, persistent organic pollutants (POPs), and nutrients, with most of these contaminants ultimately accumulating in sediments and becoming potential internal pollution sources [3]. Chromium (Cr) is a dominant heavy metal contaminant, which mainly originates from industrial and urban sources [4]. Cr(VI) is highly toxic [5] and threatens the security of some aquatic ecosystems. Thus, the removal of Cr from sediments is essential for the long-term restoration of the aquatic environment.

Among the various techniques utilized for the remediation of sediments, sediment dredging is one of the most commonly applied and effective methods for the removal of contaminants from the sediment of aquatic environments [6]. Theoretically, sediment dredging can alleviate Cr contamination, removing the source of internal pollution. Many

studies have reported the successful control of Cr contamination in aquatic systems using sediment dredging [7]. However, negative effects have also been reported. For example, Jiang et al. [8] found that the weak acid extractable Cr was higher by 40–83% after dredging, while De Jonge et al. [9] found that Cr from stable sulfide precipitates can be released due to the strong oxidation conditions created during sediment dredging. Therefore, further studies are required to comprehensively assess dredging effects in the field. Furthermore, most relevant studies had been implemented over short time-periods, typically from weeks to months, with few studies paying attention to long-term dredging efficacy.

Commonly, in natural environments, Cr exists in two forms: Cr(VI), which is highly soluble and toxic, and Cr(III), which is hardly mobile and less toxic [4]. However, Cr(III) may form stable and soluble chelated complexes with dissolved organic matter [10,11]. Therefore, judging the effects of dredging in terms of total sediment Cr concentrations and using traditional speciation methods does not provide a comprehensive assessment. When investigating redox-sensitive sediment constituents such as Cr, passive sampling techniques are the most accurate methods for determining concentrations (USEPA, 2001). Diffusive gradients in thin films (DGT) and High-resolution equilibrium dialysis (HR-Peeper) are widely used for in situ high-resolution measurement of labile and dissolved analytes in sediments, respectively [12,13]. The zirconium/zirconium oxide based DGT has been found to exhibit high selectivity for Cr(VI) [14,15]. Therefore, the combined use of HR-Peeper and DGT techniques could be most effective for evaluating the risk of Cr in sediments and the long-term effects of dredging.

Lake Taihu is the third-largest freshwater lake in China [16] and is situated in the Yangtze River Delta, one of the most developed regions of China [17]. Lake Taihu plays an important role in flood control, tourism and recreation, shipping, aquaculture, and water supply, providing drinking water for several cities in the surrounding region [17]. However, large volumes of pollutants have been discharged into the lake due to anthropogenic activities related to industrial and economic development, resulting in deterioration of water quality, eutrophication, and the increased occurrence of harmful cyanobacterial blooms. Meiliang Bay is an important harbor of Lake Taihu and is the main freshwater source for Wuxi City, despite suffering from severe levels of heavy metal contamination [18]. Environmental sediment dredging is generally aimed at removing surface sediments rich in nitrogen, phosphorus, heavy metals/metalloids, and organic pollutants, in order to improve the water ecological environment. The Water Resources Department of Jiangsu Province issued “Notice on The Opinions on Accelerating the Implementation of The Ecological Dredging Project in Taihu Lake” in 2008. Sediment dredging was implemented in the Meiliang Bay of Lake Taihu from 2009 to 2011 with an average dredging depth of 30 cm. The sampling site in this study was dredged in 2011 [19]. Removing high concentration of heavy metals/metalloids sediments, such as Cr, especially bioavailable Cr(VI), was one of the key goals of this ecological dredging project [4]. Therefore, we conducted this study to investigate the long-term impact of sediment dredging on Cr pollution in Meiliang Bay.

2. Materials and Methods

2.1. Study Site and Sampling

Meiliang Bay is located in the north of Lake Taihu with a surface area of about 100 km² and an average depth of 2.3 m. The discharge of municipal wastewater and industrial sewage into the bay has resulted in serious levels of water pollution, with studies reporting the highest levels of heavy metal pollution in this area of Lake Taihu [20]. The sampling sites selected in this study were located in two adjacent regions of Meiliang Bay. The first site (31°31′33″ N, 120°12′33″ E) was dredged in 2011 with a pre-dredge water depth of 2.6–3.2 m and post-dredge water depth of 2.8–4.5 m. The second site (31°26′18″ N, 120°11′12″ E) was not dredged prior to sample collection with water depth of 2.3–3.2 m. Before dredging, the concentrations of total sediment Cr and physical characteristics were

similar between the two sampling sites (Figure S1 and Table S1). The physical characteristics of lake water in the two sampling sites after dredging are shown in Table S2.

Sediment cores were collected quarterly, in spring (April 2016), summer (July 2016), autumn (October 2016), and winter (January 2017). At each sampling event, six sediment cores were collected from both sampling sites, with a diameter of 9 cm and a length of 30 cm. Cores were returned to the laboratory and kept still for one night allowing any disturbed and resuspended sediment to settle.

The HR-Peeper probe was constructed of Perspex plates (20 cm) with 30 equally spaced 200 μ L chambers, and the chambers were filled with deionized water and covered with a 0.45 μ m cellulose nitrate membrane. The ZrO-Chelex DGT probe was composed of a ZrO-Chelex binding gel, an agarose diffusive gel, and a Durapore[®] PVDF filter membrane. Total dissolved Cr and labile Cr(VI) were collected at a 4 mm vertical resolution using HR-Peeper and ZrO-Chelex DGT probes, which were prepared as reported by Xu et al. [13] and Wang et al. [15], respectively. The HR-Peeper collected overlying water and sediment pore water samples. During the preparation of the HR-Peeper probe devices, the chambers were first soaked in deionized water and then covered with Durapore[®] PVDF membranes. Following this, a plastic window was fixed to the membrane and the probe was tightened with screws. Then, the probe was placed in deionized water and aerated with N₂ for over 16 h prior to deployment. The DGT uptake of Cr(VI) is independent of solution pH (4.15–9.58) [15]. The assembled DGT devices were also aerated with N₂ for over 16 h.

In three of the six cores, an HR-Peeper probe and a ZrO-Chelex DGT probe were inserted into each core to determine the concentration of total dissolved Cr and labile Cr(VI) at the sediment water interface. The operational procedure for HR-Peeper and DGT techniques were described in detail by Xu et al. [13]. The other three sediment cores were cut with a cutting ring to form sections with a height of 1 cm. The 0–1, 3–4, and 9–10 cm sediment sections were packed into sealed bags and freeze-dried. Then, the sediment sections were ground and passed through a 200-mesh sieve for the determination of total sediment Cr and Cr fractions in sediments. Cutting of sediment cores and grounding of sediment sections were carried out in an anaerobic glove box (Erlab, Captair pyramid). Cr concentrations were determined only on the fine fraction passing through a 200-mesh sieve. The total sediment Cr content of sediments samples was extracted following the method of Bettinelli et al. [21], while the European Community Bureau of Reference (BCR) method was used to sequentially extract Cr fractions in sediments [22].

2.2. Sample Analyses

The exposure time of the HR-Peeper in the sediments was at least 48 h to ensure the equilibrium status was achieved between the concentrations of dissolved substances in the overlying water and pore water and the chamber water. After retrieval of the HR-Peeper from the sediment cores, the surface of the filter membrane was cleaned sequentially using wet filter papers and deionized water. To prevent oxidation of the pore water samples, two plastic membranes were placed on the surfaces of the filter membranes on the two sides of each probe. The overlying water and pore water samples were immediately collected using a pipette with a volume of about 200 μ L [13]. The ZrO-Chelex DGT probes were inserted into the sediment cores and incubated for 24 h. Finally, the DGTs were carefully removed out of the cores. For the DGT-labile element Cr(VI), the ZrO-Chelex binding gels were sliced into 4 mm sections by using a cutter made of stacking ceramic blades. Each slice was placed into a 2.0 mL centrifuge tube. The 0.2 M NaOH–0.5 M H₂O₂ solution was used to elute Cr (VI) in the binding gel [15]. The Cr concentrations of all the samples were determined by central laboratory of Jiangsu Academy of Agricultural Sciences using an inductively coupled plasma source mass spectrometer (ICP-MS, NexION 300, PerkinElmer). In the case of ICP-MS detection technique, the isotope (m/z 52) was applied to the measurement because of interference (⁴⁰Ar + ¹²C), and the collision cell/dynamic reaction cell technology was applied (KED/helium (He)).

2.3. Statistical Analyses

One-way analysis of variance (one-way ANOVA) was performed using SPSS v.22 software (IBM SPSS, Chicago, IL, USA). All figures were plotted using Origin Pro 2017 (OriginLab, Northampton, MA, USA).

The DGT-Induced Fluxes in Sediments (DIFS) model was used in this study, with the main parameters calculated using Equations (1)–(4) as follows [23]:

$$R = \frac{C_{DGT}}{C_{sol}} \quad (1)$$

$$K_d = \frac{C_s}{C_{sol}} = \frac{F_1}{C_{sol}} \quad (2)$$

$$k_{-1} = \frac{1}{T_c(1 + K_d P_c)} \quad (3)$$

$$k_1 = \frac{k_{-1}}{K_d P_c} \quad (4)$$

where the input parameter of R was calculated as the ratio of DGT-labile Cr(VI) concentration (C_{DGT} , $\mu\text{g/L}$) to the soluble Cr concentration (C_{sol} , $\mu\text{g/L}$) (Equation (1)). The Cr distribution coefficient between the solid and dissolved phases (K_d , cm^3/g) was calculated as the ratio of weak acid extractable Cr (F_1 , $\mu\text{g/L}$) to soluble Cr concentration (C_{sol}) (Equation (2)). In parameter estimation mode, an experimental DGT R value is passed to the program. If the response time (T_c , s) is to be fitted, DIFS makes an initial guess, computes a solution as just described, compares the predicted R with the experimental R in the data file, and then refines the estimate of T_c . This process continues until the R values agree within the tolerances, or until no further convergence is possible [23], referring to the time required for the perturbed system to reach 63% of its equilibrium position. Then, the desorption (k_{-1} , /s) and adsorption (k_1 , /s) rate constants for Cr in the sediments were calculated via Equations (3) and (4), respectively, where P_c (g/cm^3) refers to the particle concentration of sediments.

3. Results and Discussion

3.1. The Total Dissolved Cr and DGT-Labile Cr(VI) Concentration in Overlying Water and Sediments in the Non-Dredged and Dredged Sites

Figures 1 and 2 show the vertical distributions and average concentrations of total dissolved Cr and DGT-labile Cr(VI), respectively. The total dissolved Cr and DGT-labile Cr(VI) concentrations in the overlying water and sediments fluctuated marginally with depth, across a depth range of 8 to -100 mm, at a resolution of 4.0 mm. Therefore, an average of different depths (vertically ranging from 8 to 0 mm for overlying water and from 0 to -100 mm for sediment) was used to explore the impact of dredging on the total dissolved Cr and DGT-labile Cr(VI) concentrations. The evident seasonal variability of the total dissolved Cr and DGT-labile Cr(VI) is mainly owing to the seasonal cyanobacterial blooms and Mn redox [24]. In summer, the high concentration of total dissolved Cr was primarily caused by the complexation of Cr(III) with dissolved organic matter (DOM). The DOC complexed with Cr(III) was mainly produced by the cyanobacterial blooms in summer. In winter, the high concentrations of total dissolved Cr and DGT-labile Cr(VI) in aerobic sediments were primarily caused by reoxidation of Cr(III) by Mn(III/IV) oxides, as evidenced by the lowest concentrations of dissolved and DGT-labile Mn(II) [24].

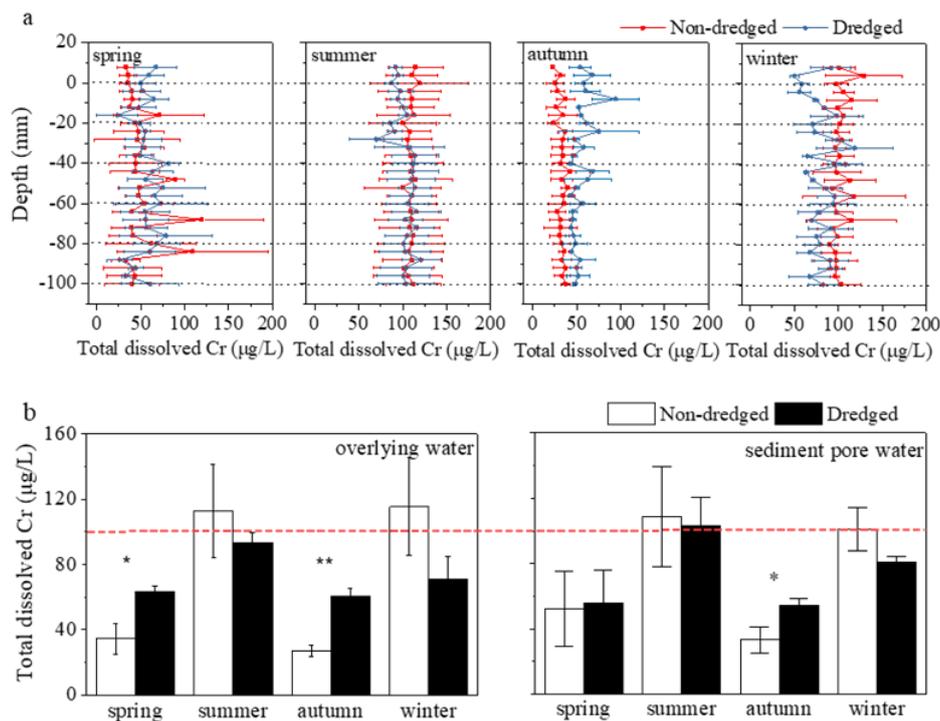


Figure 1. The vertical changes (a) and average (b) of total dissolved Cr concentration extracted by HR-Peeper in sediment and overlying water (overlying water: from 8 to 0mm depth averaged; sediment pore water: 0 to –100 mm averaged). Significant difference between the Cr concentrations of non-dredged and dredged sediments is indicated by * at 0.05 level and ** at 0.01 level. The red line indicates the limiting concentration (100 µg/L) for the fisheries water quality standards (AEPC, 2002).

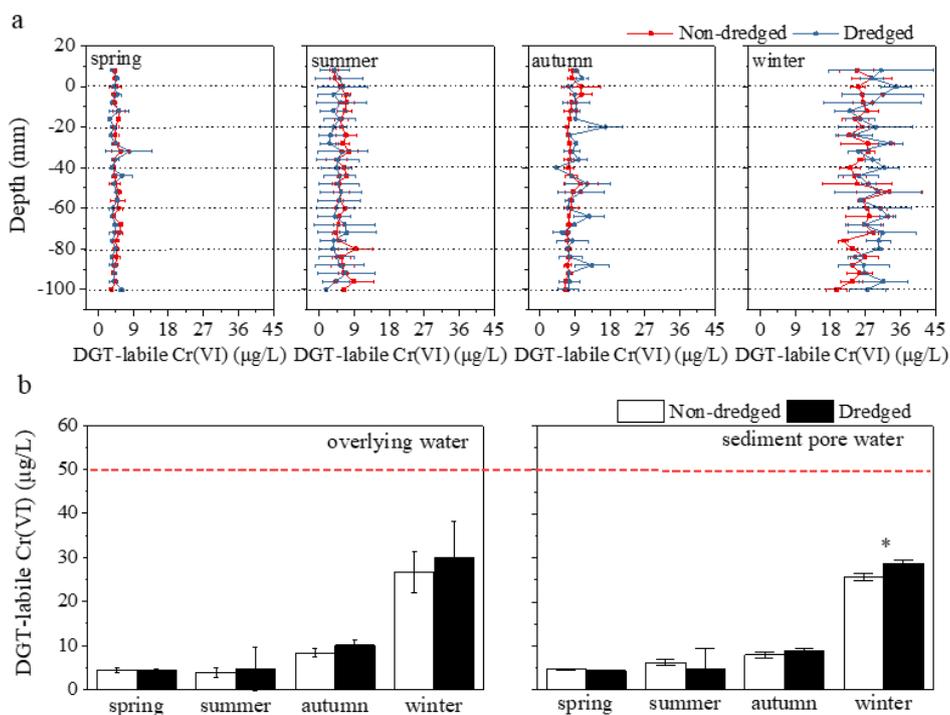


Figure 2. The vertical changes (a) and average (b) of DGT-labile Cr(VI) at sediment and overlying water (overlying water: from 8 to 0 mm depth averaged; sediment pore water: from 0 to –100 mm averaged). Significant difference between the DGT-labile Cr(VI) concentrations of non-dredged and dredged sediments is indicated by * at 0.05 level. The red line indicates the limiting concentration (50 µg/L) for the drinking water quality standards for Cr(VI) (WHO, 1993).

In the overlying water, compared with the non-dredged site, the average amount of total dissolved Cr in the dredged site was 84% higher in spring and 125% higher in autumn, while 17% lower in summer and 38% lower in winter. The trend of variation of total dissolved Cr in the sediments of dredged sites compared with the non-dredged sites was consistent with the trend observed in the overlying water. The total dissolved Cr concentration in sediments of the dredged site was 7% higher in spring and 63% higher in autumn, while 5% lower in summer and 20% lower in winter, compared with the non-dredged sediments.

Both the internal source of sediments and the external source of rivers could affect the concentrations of total dissolved Cr at the sediment–water interface investigated in this study. The sediment in shallow Lake Taihu was commonly and frequently resuspended and redeposited, probably providing a generally homogeneous surface sediment and overlying water [25]. Previous studies indicate the concentrations of Cr could significantly increase in the suspended particulate matter with increases in wind speed and duration time in Lake Taihu [26]. Besides, Cr was more likely to be adsorbed to suspended sediment in the overlying water compared with other heavy metals/metalloids [27]. Compared with the non-dredged site, the dredged site was nearer the developed city of Wuxi (Figure S2). With the rapid development of local economy, approximately $5 \times 10^8 \text{ m}^3$ wastewater has been discharged into the dredged lake zone by Liangxi River, which runs through the city (Taihu Basin Authority of Ministry of Water Resources, <http://www.tba.gov.cn/slbthlyglj/>). Thus, the concentration of total dissolved Cr of the dredged site was more affected by external source compared with the non-dredged site.

Storm water runoff is recognized as the main mechanism of transportation of external heavy metals into fluvial system [28]. The seasonal change in external Cr source could be mainly affected by rainfall. Mn, Pb, Cr, and Ni were major contributors to high values of the comprehensive pollution index of rivers after rain [29]. In autumn, the rainfall was 238.3 mm, which was 294.5% higher compared with the average year (Figure 3), even larger than the rainfall in summer. The large increase of total dissolved Cr concentration (63% in sediment and 125% in overlying water) in autumn in the dredged site could be caused by this high amount and intensity of rainfall. When we sampled in spring, it had entered the rainy season. The contribution of river discharge to the total dissolved Cr could not be ignored, resulting in an increase of 7% in sediment and 84% in overlying water. In the flood season of summer, the high concentration of total dissolved Cr was primarily caused by the complexation of Cr(III) with dissolved organic matter which was mainly produced by cyanobacterial blooms [24]. Both the dredged and non-dredged sites were suffering from serious cyanobacterial blooms in summer [30]. Thus, there was not an evident difference in the concentrations of total dissolved Cr between the two sites in summer. Winter is the typical dry season (Figure 3), and the internal sediment source contribute more to the concentration of total dissolved Cr. The beneficial effect of sediment dredging was observed with a decrease (20% in sediment and 38% in overlying water) in winter. Sediment dredging might also decrease the sediment release of Cr in the other seasons, while this effect was masked by the increase of rainfall, especially in spring and autumn.

Compared with the non-dredged site, the concentration of total dissolved Cr in the overlying water and sediment of the dredged site were both higher in spring and autumn, while being lower in summer and winter (Figure 1). The total dissolved Cr concentrations in overlying water in spring and autumn in both the non-dredged and dredged sites were well below 100 $\mu\text{g/L}$, which is the limit value of the fisheries water quality standard (AEPC, 2002) (Table 1). In the non-dredged site, the total dissolved Cr concentration in the overlying water in summer ($112.6 \pm 28.8 \mu\text{g/L}$) and winter ($115.3 \pm 29.9 \mu\text{g/L}$) exceeded the limit value of the fisheries water quality standard for total dissolved Cr (AEPC, 2002). However, the total dissolved Cr concentration in overlying water in the dredged site was $71.0 \pm 27.5 \mu\text{g/L}$ in winter (Figure 1), which was well below the 100 $\mu\text{g/L}$ limit value. This indicates that dredging could effectively reduce the risk posed by total dissolved Cr pollution in the winter periods.

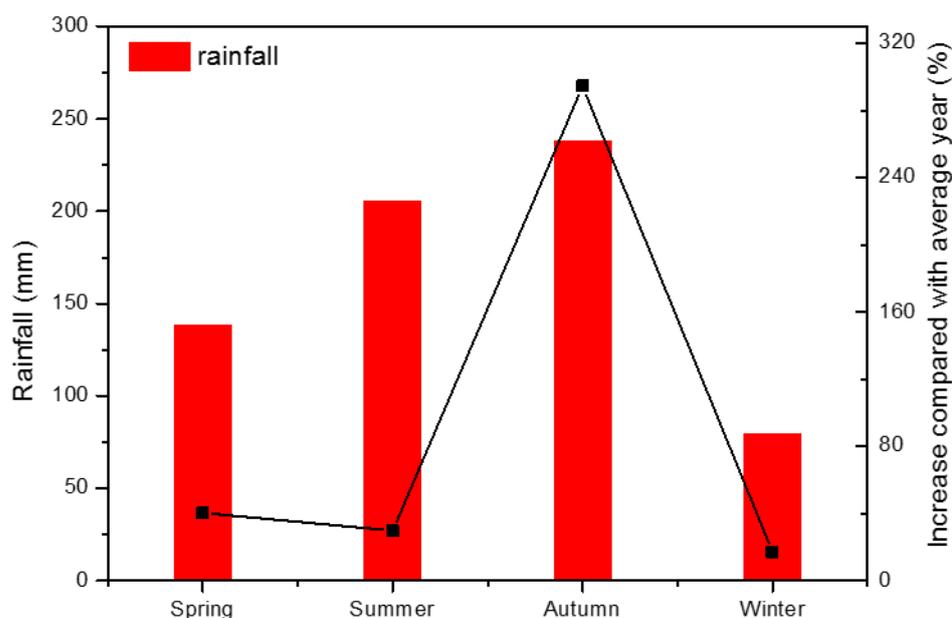


Figure 3. The seasonal fluctuation in amount of rainfall (bars) and increase compared with average year during the sampling time in this study (Taihu Basin Authority of Ministry of Water Resources, <http://www.tba.gov.cn/slbthlyglj/>).

Table 1. The environmental quality standards for Cr.

Quality Standards	Parameters	Criterion Value
1	Total dissolved Cr	100 µg/L
2	Cr(VI)	50 µg/L
3	Total sediment Cr	150 mg/kg

Note: 1 refers to the fisheries water quality standards for total dissolved Cr set by the Administration of Environmental Protection of China (AEPC, 2002); 2 refers to the drinking water quality standards for Cr(VI) set by World Health Organization (WHO, 1993); and 3 refers to the marine sediment quality standards of grade II set by State Oceanic Administration of China (SOA, 2002).

In the overlying water, compared with the non-dredged site, the average amounts of DGT-labile Cr(VI) in the dredged site was reduced by 2% in spring, while being 18% higher in summer, 21% higher in autumn, and 12% higher in winter. Compared with those in the non-dredged site, the DGT-labile Cr(VI) concentration in sediments of the dredged site was 5% lower in spring and 24% lower in summer, while 13% higher in autumn and 12% higher in winter. The concentrations of DGT-labile Cr(VI) in winter were highest in both the non-dredged (25.6 ± 6.1 µg/L) and dredged (28.7 ± 7.3 µg/L) sites, compared with the other seasons (Figure 2). Although winter measurements in the dredged sites were higher than those in the non-dredged site, measured values were all less than 50 µg/L, which is the limit value of the drinking water quality standards for Cr(VI) (WHO, 1993), indicating that dredging has little effect on the risk posed by DGT-labile Cr(VI).

3.2. Total Sediment Cr Concentrations and Fractions in the Non-Dredged and Dredged Sediments

In the non-dredged and dredged sediments, concentrations of total sediment Cr varied 59.3–75.8 and 58.8–138.7 mg/kg, respectively (Figure 4). In both the non-dredged and dredged sediments, the magnitude of all four Cr fractions was ranked in the following order: weak acid extractable Cr < reducible Cr < oxidizable Cr < residual Cr. The weak acid extractable Cr, reducible Cr, oxidizable Cr, and residual Cr fractions in the non-dredged sediment varied 0.25–0.66, 1.65–2.28, 7.55–9.01, and 48.8–65.4 mg/kg, respectively. The concentrations of weak acid extractable Cr, reducible Cr, oxidizable Cr and residual Cr fractions in the dredged sediments ranged 0.23–0.70, 1.93–3.61, 7.28–9.77, and 47.0–127.7 mg/kg, respectively (Figure 4).

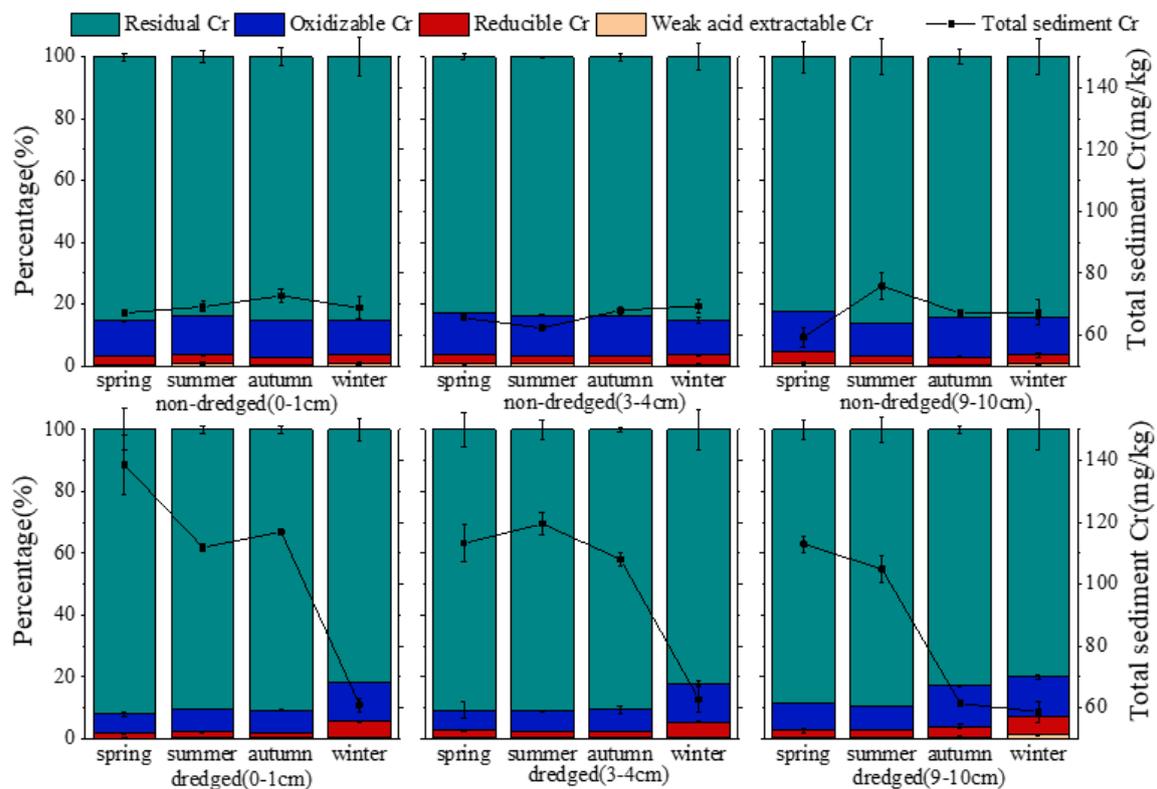


Figure 4. Seasonal percentage of changes of weak acid-soluble, reducible, oxidizable, residual fractions, and total sediment Cr at 0–1, 3–4, and 9–10 cm sediment depths. Significant difference between the Cr concentrations of non-dredged and dredged sediments is indicated by * at 0.05 level.

The total sediment Cr concentration, which was mostly composed of residual Cr, was higher after dredging, except in winter, possibly due to the slow degradation of DOM enriched Cr in deeper sediments [31] and additional inputs [32] such as the decomposition of algae and sewage discharge. Due to the low bioavailability of residual Cr, the risk of Cr contamination did not increase with the increase in residual Cr or total sediment Cr. The ranges of weak acid extractable (0.25–0.66 mg/kg) and oxidizable Cr (7.55–9.01 mg/kg) fractions in the non-dredged sediment was similar to those (0.23–0.70 and 7.28–9.77 mg/kg) in dredged sediments. Although the range of reducible Cr in the dredged sediment (1.93–3.61 mg/kg) was higher than in the non-dredged sediment (1.65–2.28 mg/kg), the total sediment Cr concentration was less than 150 mg/kg, which is the limit value for marine sediment quality standards of grade II (SOA, 2002). Overall, these findings indicate that there is no significant risk presented by total sediment Cr concentrations or the four extracted Cr fractions due to dredging.

3.3. The Kinetics of Cr Mobilization

The sediment Cr at the depths of 0–1 and 3–4 cm was more ready to move into overlying water than that at the depth of 9–10 cm. Therefore, the depths of 0–1 and 3–4 cm were selected to analyze the kinetics of Cr mobilization in non-dredged and dredged sediments using the DIFS model (Table 2). The R values calculated using Equation (1) were similar or smaller in the dredged site (0.05–0.15) compared to those of the non-dredged site (0.06–0.33), except in winter. The R values computed from the data in autumn and winter (0.12–0.50) were larger than those in spring and summer (0.05–0.11). This result indicates that Cr was more easily resupplied from the solid sediments in autumn and winter. A greater K_d value depicts a higher ability to preserve Cr in the sediment solids [33]. The K_d values were similar in the dredged site (3.46–9.09 cm³/g) and the non-dredged site (3.41–8.07 cm³/g) except in autumn, reflecting similar sediment Cr retention capabilities overall. The values

of response time T_c were similar or greater in the dredged site ($0.05\text{--}1.00 \times 10^7$ s) compared with the non-dredged site ($0.01\text{--}1.00 \times 10^7$ s), indicating that a similar or longer period was needed before reaching equilibrium [34]. The k_1 and k_{-1} values in the dredged site (7.29×10^{-9} to $3.67/s$ and 2.36×10^{-8} to $7.31/s$) were similar or smaller than those in the non-dredged site (6.73×10^{-9} to $10.90/s$ and 2.28×10^{-8} to $29.75/s$), indicating similar or slower desorption and adsorption processes, while also being consistent with the observed changes in T_c .

Table 2. Parameters of Cr from DIFS model at depths of 0–1 and 3–4 cm in the non-dredged and dredged sediments.

Depth	Parameters	Spring		Summer		Autumn		Winter		
		Non-Dredged	Dredged	Nondredged	Dredged	Nondredged	Dredged	Nondredged	Dredged	
0–1 cm	In put	R	0.11	0.07	0.06	0.06	0.33	0.12	0.24	0.50
	Out put	K_d (cm^3/g)	6.55	4.07	5.88	5.63	9.04	4.23	4.74	4.76
		T_c (s)	5.48×10^4	1.00×10^7	1.00×10^7	1.00×10^7	0.09	2.69×10^4	0.01	2.89
		k_1 (/s)	1.02×10^{-6}	1.28×10^{-8}	6.73×10^{-9}	7.27×10^{-9}	0.36	4.44×10^{-6}	10.90	0.03
		k_{-1} (/s)	3.83×10^{-6}	2.99×10^{-8}	2.28×10^{-8}	2.36×10^{-8}	1.88	1.08×10^{-5}	29.75	0.09
3–4 cm	In put	R	0.1	0.09	0.06	0.05	0.23	0.15	0.26	0.29
	Out put	K_d (cm^3/g)	8.07	9.09	4.03	4.06	12.65	6.81	3.41	3.46
		T_c (s)	1.14×10^5	3.29×10^5	1.00×10^7	1.00×10^7	1.21	1.26×10^4	0.04	0.05
		k_1 (/s)	3.33×10^{-7}	9.32×10^{-8}	1.3×10^{-8}	1.28×10^{-8}	0.01	4.11×10^{-6}	4.34	3.67
		k_{-1} (/s)	1.55×10^{-6}	4.88×10^{-7}	3.01×10^{-8}	3.00×10^{-8}	0.10	1.61×10^{-5}	8.54	7.31

These results show a higher response time (T_c), lower adsorption rate (k_1), and desorption rate (k_{-1}) in the dredged site except in summer, indicating that sediment dredging decreased Cr mobility in sediments six years after dredging.

4. Conclusions

The current study focused on the long-term effects of dredging on Cr contamination in sediments of Lake Taihu (China). Dredging successfully lowered the concentrations of total dissolved Cr in winter, ensuring they were well below the limit value of the fisheries water quality standard (AEPC, 2002), while in the non-dredged site their concentrations were beyond the limit value in summer and winter. The concentration of total dissolved Cr in the dredged site was significantly higher in spring ($63.3 \pm 17.7 \mu\text{g/L}$) and autumn ($60.8 \pm 16.2 \mu\text{g/L}$), which were probably caused by the large rainfall and discharge during the two seasons. The concentration of total dissolved Cr between the two sites was not evidently different in summer, which might be caused by the similar intensity of cyanobacterial blooms there. DGT-labile Cr(VI) and total sediment Cr levels were higher after dredging. However, the higher risk posed by Cr contamination was negligible, with the concentrations of DGT-labile Cr(VI) and total sediment Cr in sediments being well below the limit values for the drinking water quality standard (WHO, 1993) and the marine sediment quality standards of grade II (SOA, 2002), respectively. Dredging suppressed the kinetic remobilization of Cr from solid sediments except in summer, prolonging the equilibration period and reducing desorption and adsorption processes, as shown by the higher T_c and lower k_1 and k_{-1} values. Overall, these findings confirm that sediment dredging decreased the risk of Cr contamination in winter in Lake Taihu six years after sediment dredging.

Supplementary Materials: The following are available online at <https://www.mdpi.com/2073-4441/13/3/327/s1>, Figure S1. The sampling site locations and the Cr concentrations in overlying water and sediment in March 2010 (the Cr concentrations were referred to Jiang et al., 2012), Figure S2. Map of Lake Taihu (left) and location of discharge pipes of Liangxi River, dredged sites, and un-dredged sites (right), Table S1. The physicochemical characteristics of overlying water and sediment before dredging in July 2010 (Fan et al., 2016), Table S2. The physicochemical characteristics of overlying water after dredging in 2016, Table S3. The characteristics of conditions on sample sites after dredging in 2016.

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References

1. Lau, S.; Lane, S. Biological and chemical factors influencing shallow lake eutrophication: A long-term study. *Sci. Total. Environ.* **2002**, *288*, 167–181. [[CrossRef](#)]
2. Moore, J.W.; Schindler, D.E.; Scheuerell, M.D.; Smith, D.; Frodge, J. Lake Eutrophication at the Urban Fringe, Seattle Region, USA. *Ambio* **2003**, *32*, 13–18. [[CrossRef](#)] [[PubMed](#)]
3. Liu, C.; Shao, S.; Shen, Q.; Fan, C.; Zhou, Q.; Yin, H.; Xu, F. Use of multi-objective dredging for remediation of contaminated sediments: A case study of a typical heavily polluted confluence area in China. *Environ. Sci. Pollut. Res.* **2015**, *22*, 17839–17849. [[CrossRef](#)]
4. Gorny, J.; Billon, G.; Noiriell, C.; Dumoulin, D.; Lesven, L.; Madé, B. Chromium behavior in aquatic environments: A review. *Environ. Rev.* **2016**, *24*, 503–516. [[CrossRef](#)]
5. Besser, J.M.; Brumbaugh, W.G.; Kemble, N.E.; May, T.W.; Ingersoll, C.G. Effects of Sediment Characteristics on the Toxicity of Chromium(III) and Chromium(VI) to the Amphipod, *Hyalella azteca*. *Environ. Sci. Technol.* **2004**, *38*, 6210–6216. [[CrossRef](#)]
6. Yu, J.; Fan, C.; Zhong, J.; Zhang, L.; Zhang, L.; Wang, C.; Yao, X. Effects of sediment dredging on nitrogen cycling in Lake Taihu, China: Insight from mass balance based on a 2-year field study. *Environ. Sci. Pollut. Res.* **2015**, *23*, 3871–3883. [[CrossRef](#)]
7. Mao, Z.-G.; Gu, X.-H.; Lu, X.-M.; Zeng, Q.; Gu, X.; Li, X. Pollution distribution and potential ecological risk assessment of heavy metals in sediments from the different eastern dredging regions of Lake Taihu. *Huanjing Kexue* **2014**, *35*, 186–193.
8. Jiang, X.; Shi, Z.; Liu, F.; Chen, C.; Jin, X. Effects of Dredging on Speciation and Bio-Toxicity of Heavy Metals in the Surface Sediment from Meiliang Bay, Taihu Lake. *Res. Environ. Sci.* **2010**, *23*, 1151–1157.
9. De Jonge, M.; Teuchies, J.; Meire, P.; Blust, R.; Bervoets, L. The impact of increased oxygen conditions on metal-contaminated sediments part I: Effects on redox status, sediment geochemistry and metal bioavailability. *Water Res.* **2012**, *46*, 2205–2214. [[CrossRef](#)] [[PubMed](#)]
10. Gustafsson, J.P.; Persson, I.; Oromieh, A.G.; Van Schaik, J.W.J.; Sjöstedt, C.; Kleja, D.B. Chromium(III) Complexation to Natural Organic Matter: Mechanisms and Modeling. *Environ. Sci. Technol.* **2014**, *48*, 1753–1761. [[CrossRef](#)] [[PubMed](#)]
11. Puzon, G.J.; Tokala, R.K.; Zhang, H.; Yonge, D.; Peyton, B.M.; Xun, L. Mobility and recalcitrance of organo-chromium(III) complexes. *Chemosphere* **2008**, *70*, 2054–2059. [[CrossRef](#)] [[PubMed](#)]
12. Ding, S.; Xu, D.; Sun, Q.; Yin, H.; Zhang, C. Measurement of Dissolved Reactive Phosphorus Using the Diffusive Gradients in Thin Films Technique with a High-Capacity Binding Phase. *Environ. Sci. Technol.* **2010**, *44*, 8169–8174. [[CrossRef](#)]
13. Xu, D.; Wu, W.; Ding, S.; Sun, Q.; Zhang, C. A high-resolution dialysis technique for rapid determination of dissolved reactive phosphate and ferrous iron in pore water of sediments. *Sci. Total. Environ.* **2012**, *421*, 245–252. [[CrossRef](#)] [[PubMed](#)]
14. Devillers, D.; Buzier, R.; Simon, S.; Charriau, A.; Guibaud, G. Simultaneous measurement of Cr(III) and Cr(VI) in freshwaters with a single Diffusive Gradients in Thin Films device. *Talanta* **2016**, *154*, 533–538. [[CrossRef](#)] [[PubMed](#)]
15. Wang, Y.; Ding, S.; Shi, L.; Gong, M.; Xu, S.; Zhang, C. Simultaneous measurements of cations and anions using diffusive gradients in thin films with a ZrO-Chex mixed binding layer. *Anal. Chim. Acta* **2017**, *972*, 1–11. [[CrossRef](#)] [[PubMed](#)]
16. Chengxin, F.; Shang, J.; Lu, X.; Fan, C. Effects of sludge dredging on the prevention and control of algae-caused black bloom in Taihu Lake, China. *J. Environ. Sci.* **2013**, *25*, 430–440. [[CrossRef](#)]
17. Qin, B.; Xu, P.; Wu, Q.; Luo, L.; Zhang, Y. Environmental issues of Lake Taihu, China. *Hydrobiol.* **2007**, *581*, 3–14. [[CrossRef](#)]
18. Wenchuan, Q.; Dickman, M.; Sumin, W. Multivariate analysis of heavy metal and nutrient concentrations in sediments of Taihu Lake, China*. *Hydrobiol.* **2001**, *450*, 83–89. [[CrossRef](#)]

19. Wang, Z.; Li, D.; Qin, H.; Li, Y. An integrated method for removal of harmful cyanobacterial blooms in eutrophic lakes. *Environ. Pollut.* **2012**, *160*, 34–41. [[CrossRef](#)]
20. Sun, Q.; Ding, S.; Chen, M.; Gao, S.; Lu, G.; Wu, Y.; Gong, M.; Wang, D.; Wang, Y. Long-term effectiveness of sediment dredging on controlling the contamination of arsenic, selenium, and antimony. *Environ. Pollut.* **2019**, *245*, 725–734. [[CrossRef](#)]
21. Bettinelli, M.; Beone, G.; Spezia, S.; Baffi, C. Determination of heavy metals in soils and sediments by microwave-assisted digestion and inductively coupled plasma optical emission spectrometry analysis. *Anal. Chim. Acta* **2000**, *424*, 289–296. [[CrossRef](#)]
22. Rauret, G.; López-Sánchez, J.F.; Sahuquillo, A.; Rubio, R.; Davidson, C.; Ure, A.; Quevauviller, P. Improvement of the BCR three step sequential extraction procedure prior to the certification of new sediment and soil reference materials. *J. Environ. Monit.* **1999**, *1*, 57–61. [[CrossRef](#)]
23. Harper, M.P.; Davison, W.; Tych, W. DIFS—a modelling and simulation tool for DGT induced trace metal remobilisation in sediments and soils. *Environ. Model. Softw.* **2000**, *15*, 55–66. [[CrossRef](#)]
24. Fan, X.; Ding, S.; Chen, M.; Gao, S.; Fu, Z.; Gong, M.; Tsang, D.C.; Wang, Y.; Zhang, C. Peak Chromium Pollution in Summer and Winter Caused by High Mobility of Chromium in Sediment of a Eutrophic Lake: In Situ Evidence from High Spatiotemporal Sampling. *Environ. Sci. Technol.* **2019**, *53*, 4755–4764. [[CrossRef](#)] [[PubMed](#)]
25. Jalil, A.; Li, Y.; Zhang, K.; Gao, X.; Wang, W.; Khan, H.O.S.; Pan, B.; Ali, S.; Acharya, K. Wind-induced hydrodynamic changes impact on sediment resuspension for large, shallow Lake Taihu, China. *Int. J. Sediment. Res.* **2019**, *34*, 205–215. [[CrossRef](#)]
26. Zheng, S.; Wang, P.; Wang, C.; Hou, J.; Qian, J. Distribution of metals in water and suspended particulate matter during the resuspension processes in Taihu Lake sediment, China. *Quat. Int.* **2013**, *286*, 94–102. [[CrossRef](#)]
27. Wang, H.; Zhou, Y.; Wang, X. Transport Dynamics of Cr and Zn Between Deposited Sediment and Overlying Water. *CLEAN Soil Air Water* **2016**, *44*, 1453–1460. [[CrossRef](#)]
28. Villanueva, J.D.; Granger, D.; Binet, G.; Litrico, X.; Huneau, F.; Peyraube, N.; Le Coustumer, P. Labile trace metal contribution of the runoff collector to a semi-urban river. *Environ. Sci. Pollut. Res.* **2016**, *23*, 11298–11311. [[CrossRef](#)]
29. Zhang, L.; Zhao, B.; Xu, G.; Guan, Y. Characterizing fluvial heavy metal pollutions under different rainfall conditions: Implication for aquatic environment protection. *Sci. Total. Environ.* **2018**, *635*, 1495–1506. [[CrossRef](#)] [[PubMed](#)]
30. Qin, B.; Yang, G.; Ma, J.; Wu, T.; Li, W.; Liu, L.; Deng, J.; Zhou, J. Spatiotemporal Changes of Cyanobacterial Bloom in Large Shallow Eutrophic Lake Taihu, China. *Front. Microbiol.* **2018**, *9*, 451. [[CrossRef](#)]
31. Kikuchi, T.; Tanaka, S. Biological Removal and Recovery of Toxic Heavy Metals in Water Environment. *Crit. Rev. Environ. Sci. Technol.* **2012**, *42*, 1007–1057. [[CrossRef](#)]
32. Chen, M.; Ding, S.; Gao, S.; Fu, Z.; Tang, W.; Wu, Y.; Gong, M.; Wang, D.; Wang, Y. Efficacy of dredging engineering as a means to remove heavy metals from lake sediments. *Sci. Total. Environ.* **2019**, *665*, 181–190. [[CrossRef](#)] [[PubMed](#)]
33. Zhou, A.; Tang, H.; Wang, D. Phosphorus adsorption on natural sediments: Modeling and effects of pH and sediment composition. *Water Res.* **2005**, *39*, 1245–1254. [[CrossRef](#)] [[PubMed](#)]
34. Harper, M.P.; Davison, W.; Zhang, H.; Tych, W. Kinetics of metal exchange between solids and solutions in sediments and soils interpreted from DGT measured fluxes. *Geochim. Cosmochim. Acta* **1998**, *62*, 2757–2770. [[CrossRef](#)]