Sediment pollution characteristics and *in situ* control in a deep drinking water reservoir

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ARTICLE INFO

ABSTRACT

Sediment pollution characteristics, *in situ* sediment release potential, and *in situ* inhibition of sediment release were investigated in a drinking water reservoir. Results showed that organic carbon (OC), total nitrogen (TN), and total phosphorus (TP) in sediments increased from the reservoir mouth to the main reservoir. Fraction analysis indicated that nitrogen in ion exchangeable form and NaOH-extractable P (Fe/Al-P) accounted for 43% and 26% of TN and TP in sediments of the main reservoir. The Risk Assessment Code for metal elements showed that Fe and Mn posed high to very high risk. The results of the *in situ* reactor experiment in the main reservoir showed the same trends as those observed in the natural state of the reservoir in 2011 and 2012; the maximum concentrations of total OC, TN, TP, Fe, and Mn reached 4.42 mg/L, 3.33 mg/L, 0.22 mg/L, 2.56 mg/L, and 0.61 mg/L, respectively. An *in situ* sediment release inhibition technology, the water-lifting aerator, was utilized in the reservoir. The results of operating the water-lifting aerator indicated that sediment release was successfully inhibited and that OC, TN, TP, Fe, and Mn in surface sediment could be reduced by 13.25%, 15.23%, 14.10%, 5.32%, and 3.94%, respectively.

Introduction

Various pollutants adsorb to sediments that accumulate at the bottom of reservoirs. These sediments accumulate over long periods of time and can act as new pollutant sources to the overlying water (Sun et al., 2009; Młynarczyk et al., 2013). The forms and types of pollutants usually vary in different sediments because bonding forces vary with respect to grain size (Ruttenberg, 1992; Bo et al., 2003), which results in different release times and release potentials.

As is well known, nitrogen and phosphorus are the most common macronutrients for phytoplankton growth. Furthermore, the nitrogen and phosphorus transport processes between sediments and overlying water play a crucial role in the aquatic ecological environment (Huang et al., 2015). Meanwhile, heavy metal pollution is an environmental crisis that accompanies rapid economic development in many countries. Metal elements constitute an environmental threat by creating serious human health hazards and affecting ecological food chains (Souza and Wasserman, 2015; Xuelu and Chen-Tung Arthur, 2012). To clarify the transport processes, first we should have a clear understanding of the nitrogen, phosphorus, and metal element fractions and release potentials in sediments. During the stratification period, sediment release in Grand Lake (Nikolai et al., 2014) yielded total nitrogen (TN) and total phosphorus (TP) concentrations in the hypolimnion of 1.1 mg/L and 0.17 mg/L, respectively. Soluble Fe and Mn also reached problematic

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levels during the summer stratification in Carvins Cove Reservoir (Nikolai et al., 2014). Thus, sediments act as a source of pollution.

Many laboratory studies have reported on sediment release of N, P, Fe, and Mn (Wang et al., 2014; Wu and Hua, 2014; Zhang et al., 2014). Considering the sediment form changes as well as pressure and temperature variations that occur in real settings, laboratory release experiments are not a reliable reflection of sediment release in reservoirs. In the present study, a new *in situ* reactor was used to study the release characteristics of pollutants in a canyon-shaped deep-water reservoir. Using the intact sediment and true pressure, the *in situ* reactor was used to produce more credible data. Of the *in situ* sediment remediation strategies, sediment capping has been applied in Lake Taihu (Xu et al., 2012) for internal P control, but this method could not be applied in the Jinpen Reservoir for deposition carried by runoff. Additives that were used in Suzhou Creek (Qian et al., 2009) successfully stabilized the metal elements, but this may also cause new problems in drinking water reservoirs, such as increased iron from additives containing ferrihydrite. Dredging for sediment remediation is always toxic to phytoplankton and autotrophic bacteria (Nayar et al., 2004). In this study, sediment release control using water-lifting aerators (WLAs) was shown to be clean and environmentally safe. We also demonstrated significant results: concentrations of organic carbon (OC), TN, TP, Fe, and Mn in the sediment were reduced during the operation of WLAs. The goal of this study was to understand the pollution status and release potential of reservoir sediments and to improve the ability of WLAs to control the release of internal sediments.

1. Materials and methods

1.1. Sampling sites

Jinpen Reservoir (34°42′–34°13′N; 107°43′–108°24′E) is the main drinking water reservoir of Xi'an City in Shaanxi Province, China. Jinpen Reservoir is located at the foot of the Qinling Mountains, 90 km from Xi'an City, as shown in Fig. 1. It is a canyon-shaped deep-water reservoir, and the main reservoir length extends 3.5 km. The total capacity of the reservoir is 2.0×10^8 m^3. When full, the surface area of the reservoir is 4.55 km^2. The main function of the reservoir is urban water supply; agricultural irrigation, power generation, and flood control are accessory functions.

The Heihe River, which originates in the Qinling Mountains, is the main water supply for Jinpen Reservoir. The river is 91.2 km long with a catchment area of 1418 km^2, and the catchment is largely undeveloped and consists primarily of forested mountains. Along the reservoir, six representative sampling sites were chosen. S1 (33°58′26.91″E, 108°08′50.02″N, depth 1 m) was located at the reservoir mouth; S2 (34°00′16.7″E, 108°10′38.67″N, depth 5 m) and S3 (34°01′14.24″E, 108°10′58.1″N, depth 20 m) were upstream from the reservoir; S4 (34°01′58.17″E, 108°11′13.78″N, depth 40 m) was near the middle of the reservoir; S5 (34°02′35.95″E, 108°11′52.83″N, depth 65 m) and S6 (34°02′42.20″E 108°12′22.76″N 70, depth 70 m) represented the main reservoir.

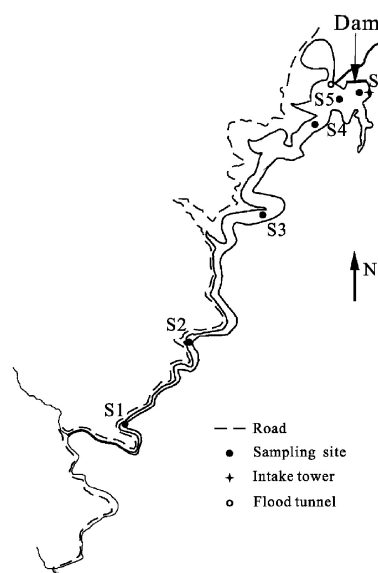

Fig. 1 – Position of Jinpen Reservoir and sampling sites (S1–S6).

1.2. Field work

Sediment samples were collected using a core sampler (HL-CN, Hengling technology Ltd., Corp., China; Ni and Wang, 2015). Sediment columns that showed no evidence of disturbance were retained. One centimeter segments of the sediment columns were collected, transferred to pre-cleaned polythene bags, and sealed. Sediment samples were kept at –4°C until laboratory processing (Zahra et al., 2013).

The *in situ* reactor utilized is a proprietary device specially designed for the investigation of migration and transformation of substances at the sediment–water interface, and its schematic diagram is shown in Fig. 2. The reactor is a stainless steel cylinder with its bottom open. The *in situ* reactor was installed at the water–sediment interface using a windlass with a wire rope, and the lower part of the reactor was embedded into the sediment by its own weight. The water in the reactor was separated from the surrounding water, and dissolved oxygen (DO) consumption and sediment release was monitored by collecting water samples through the outflow nozzle using a long pipe (up to 70 m), as shown. An air bag was installed inside the reactor to maintain internal and external pressure balance. The water samples were pumped into pre-cleaned polythene bottles every 48 hr. Before laboratory analysis, the water samples were cooled to
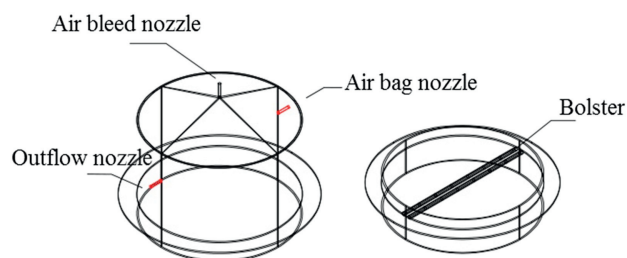

Fig. 2 – Schematic diagram of the *in situ* reactor.

Table 1 – Physical and chemical compositions of six investigated surface sediment samples.

	S1	S2	S3	S4	S5	S6
Silt (%)	26.1	55.4	75.2	79.6	88.5	89.2
Clay (%)	8.6	9.2	10.4	14.3	10.0	8.7
Sand (%)	65.3	35.4	14.4	6.1	1.5	2.1
Median particle size (μm)	21.4	15.5	11.2	10.3	5.9	6.2
Moisture content (%)	65.4	74.2	83.5	82.9	79.8	82.9
Loss on ignition (%)	5.68	4.22	7.24	6.89	8.12	8.29

4°C. A DS5 (HACH, USA) device was attached to the bolster to determine DO concentrations *in situ*.

Water samples in the reservoir were collected every 5 m vertically, and were collected every week under normal conditions and every 2 days while the WLAs were operating. All water samples were also stored immediately until the laboratory analysis could be carried out.

1.3. Analytic methods

Large objects including stones, pieces of brick, and branches were manually removed from sediment samples. Then sediment samples were oven dried, ground, homogenized, and sieved through a 1 mm mesh. The organic matter content was estimated from the weight loss on ignition (LOI%) at 550°C for 5 hr (Młynarczyk et al., 2013). The proportion of sand, silt, and clay was calculated to determine the soil textural class using the Bouyoucos hydrometer method. To determine the grain size distribution, fresh sediment samples were first treated with H_2O_2 (10% volume) to remove organic matter and were later disaggregated and dispersed ultrasonically with pyrophosphate (López et al., 2015). Beckman-Coulter Ls230 laser particle size analysis equipment was used to perform the granulometric determination.

Nitrogen in ion exchangeable form (IEF-N), nitrogen in weak acid exchangeable form (WAEF-N), nitrogen in strong alkali exchangeable form (SAEF-N), and nitrogen in strong oxidation extractable form (SOEF-N) were extracted using the methods described by Yao et al. (2015). P fractions were determined using the SMT protocol, as described in a previous study (Ruban et al., 1999). The operationally defined scheme was composed of five steps: NaOH-extractable P (Fe/Al-P), HCl-extractable P (Ca-P), organic P (OP), inorganic P (IP), and concentrated HCl-extractable P. The TP concentration in sediments was determined by treating the sample at 450°C, followed by HCl extraction (Wang et al., 2012).

The sequential extraction procedure reported by Tessier et al. (1979) was used to obtain information about the fractions of Fe and Mn in sediment. This scheme extracts the sediment-bound metals into the following five fractions: exchangeable (F1), carbonate-bound (F2), Fe-Mn oxide-bound (F3), organic matter bound (F4) and residual form (F5). The supernatant solution from each extraction phase that contained Fe and Mn was analyzed using an inductively coupled plasma optical emission spectrometer (ICP-OES; PerkinElmer, Wellesley, MA, USA). The sediment samples were digested with a $HCl–HNO_3–HF–HClO_4$ mixture (Jie et al., 2014; Dhanakumar et al., 2014).

Determination methods for TN, TP, Fe, Mn, total organic carbon (TOC), and DO in water were performed as reported by Huang et al. (2014).

2. Results and discussion

2.1. Sediment pollution characteristics

2.1.1. Physical and chemical characteristics of sediment
Based on particle size, sediment samples, as well as soils, can be separated into different fractions: sand (0.05–2 mm), silt (0.002–0.05 mm), and clay (<0.002 mm) (Młynarczyk et al., 2013). Due to increased surface area, smaller particles are more reactive (Cade-Menun, 2005). As shown in Table 1, particle size analysis indicated that the majority of surface sediments (except at S1) had high proportions of silt, ranging from 55.4% to 89.2%. This indicated that compared to the reservoir mouth, the surface sediments in the main reservoir were more likely to be a pollution source.
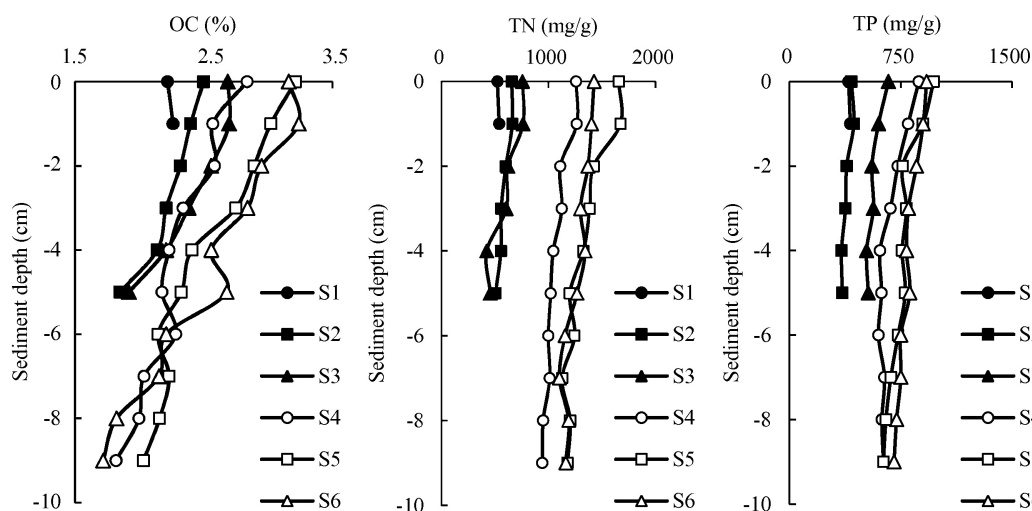
Fig. 3 – Spatial distributions of OC, TN, and TP. OC: organic carbon; TN: total nitrogen; TP: total phosphorus.
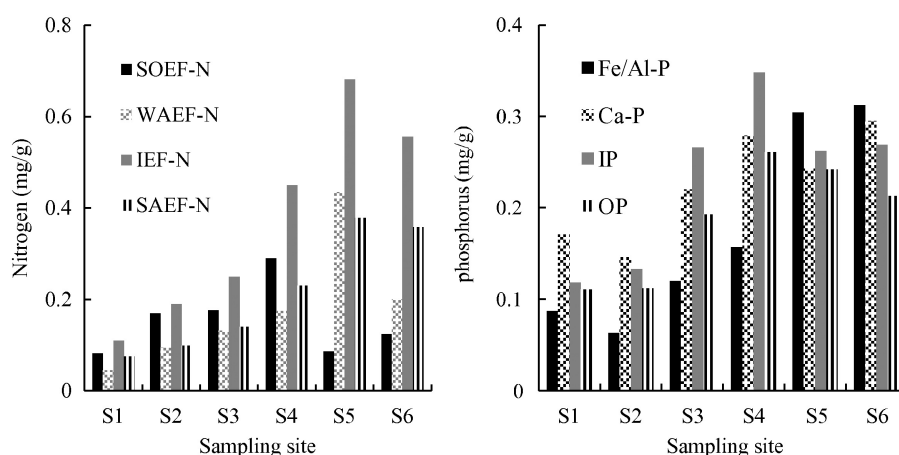
Fig. 4 – N and P fraction concentrations of surface sediments (SOEF-N: N in strong oxidation extractable form, WAEF-N: N in weak acid exchangeable form, IEF-N: N in ion exchangeable form, SAEF-N: N in strong alkali exchangeable form, Fe/Al-P: NaOH-extractable P, Ca-P: HCl-extractable P, IP: inorganic P, OP: organic P).

The median particle size of investigated surface sediments from S1 to S6 showed a declining trend, from 21.4 to 6.2 μm. At S1, the flow velocity was much greater than the flow velocity in the main reservoir. As such, large particles settled first in the reservoir mouth, and then small particles settled in the main reservoir. At S6, the proportion of small particles (silt) accounted for nearly 90% of the sediment.

The quality and quantity of OC (as LOI) in the surface sediment depended significantly on the sediment deposited, especially its granulation (Cade-Menun, 2005).

2.1.2. Horizontal and vertical variation and partitioning of OC, TN, and TP

The spatial distributions of OC, TN, and TP concentrations at varying depths and positions are shown in Fig. 3. Concentrations of OC, TN, and TP showed similar trends from the reservoir mouth to the main reservoir. The concentrations increased gradually, and the degree of pollution in surface sediments was always greater than in lower sediments.

The percentage of OC in surface sediments ranged from 2.22% to 3.16%. The overall horizontal distribution showed the following variation pattern: main reservoir (S5, S6) > middle reaches (S4) > upstream reservoir (S2, S3) > reservoir mouth (S1), with averages of 3.02% ± 0.21%, 2.59% ± 0.09% and 2.30% ± 0.10%, respectively. TN and TP concentrations in surface sediments were 520–1670 mg/kg and 400–970 mg/kg, respectively. The distribution patterns of TN and TP were consistent with the distribution of OC along the entire reservoir.

Vertical distributions of OC, TN, and TP in surface sediments were greater in the surface sediments than they were in the sub-surface sediments. Their concentrations in the lower part of the sediments showed no obvious trend. The concentrations started to increase at 3 cm below the surface in the sediment cores.

Decreased water velocity due to the construction and operation of Jinpen Reservoir led to nutrient accumulation in the sediment. This may be the primary explanation for the horizontal distributions of OC, TN, and TP. The vertical distributions indicted that in the early years of or prior to the construction of Jinpen Reservoir, OC, TN, and TP sources were

natural inputs. Increasing concentrations in the surface sediments resulted from the construction and operation of Jinpen Reservoir. On the other hand, the increased pollutant concentrations could also be attributed to the rapid intensification of agricultural activities during this time period. It is clear that the endogenous pollution load of Jinpen Reservoir accelerated. The
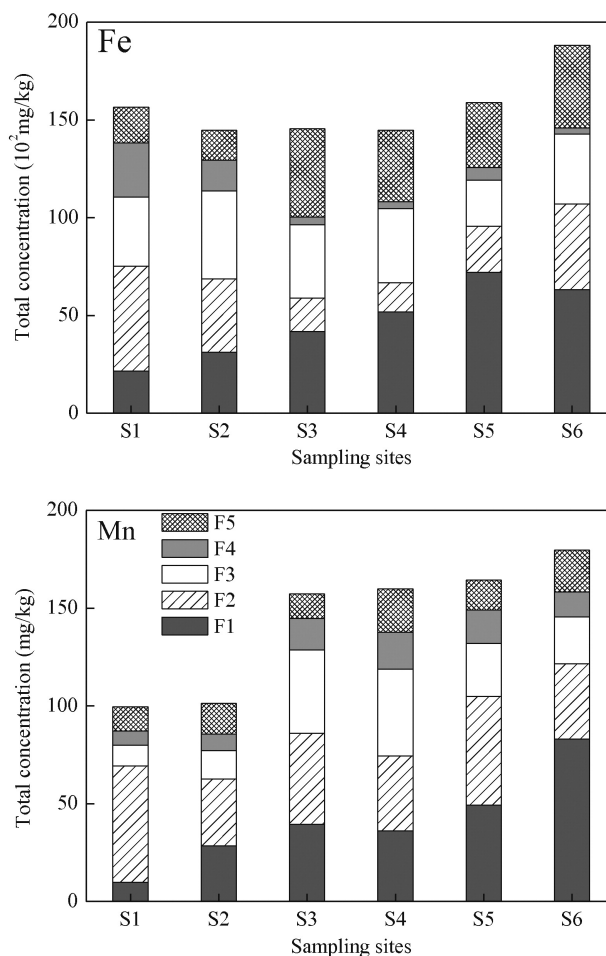
Fig. 5 – Fe and Mn fractions of surface sediments in S1 to S6.

Table 2 – Risk Assessment Code (RAC) of metals in sampling sites.

RAC	Criteria: metals in F1 and F2 fractions	Metals and the risk category in sampling sites
No risk	<1%	–
Low risk	1%–10%	–
Medium risk	11%–30%	–
High risk	31%–50%	Mn (S4) Fe (S1, S2, S3, S4)
Very high risk	>50%	Mn (S1, S2, S3, S5, S6) Fe (S5, S6)

high concentrations of TN and TP in surface sediments could have acted as sources of N and P to the overlying water during the summer stratification period (Ni and Wang, 2015). In light of accelerated reservoir eutrophication and severe ecological degradation, a new technology (WLAs) that controls endogenous pollution release was applied in Jinpen Reservoir. This technology can successfully mitigate the endogenous pollution problem.

N fractions and P fractions were determined, and the concentrations are shown in Fig. 4. Of the four different nitrogen fractions, IEF-N was the most active and the weakest in combination with sediment. As such, if the sediment environment was to shift to aerobic conditions, IEF-N would be easily released into the overlying water. From S1 to S6, IEF-N concentrations increased gradually, and in the main reservoir IEF-N accounted for 43.1% of TN. This means that when the reservoir hypolimnion experiences aerobic conditions, sediment pollution will be the most significant pollution source.

As for P in the sediment, Fe/Al-P is the most active and the main fraction that participates in sediment release. As shown in Fig. 4, nearly all four fractions increased from S1 to S6. At S5 and S6, Fe/Al-P accounted for 26.4% and 26.2%, respectively. The percentages of Fe/Al-P in Jinpen Reservoir were higher than the percentages in Sulejow Reservoir (Trojanowska and Izydorczyk, 2010) and Goczalkowice Reservoir (Młynarczyk et al., 2013).

2.1.3. Horizontal variation of Fe and Mn concentrations in surface sediment

The five fractions and cumulative heavy metal concentrations are shown in Fig. 5. Average concentrations of Fe and Mn (sum of five geochemical fractions) in the main reservoir were 15,880 and 164.4 mg/kg, respectively. The abundance of Fe was greater than that of Mn. Among the six sampling sites, S5 and S6 samples contained higher concentrations of metal elements, implying the influence of storm runoff and sedimentation.

The sequential extraction procedure used to determine partitioning of Fe and Mn revealed that F1 concentrations

increased gradually from S1 to S6. The F1 fraction was the most active, which indicated that in the main reservoir Fe and Mn were likely to be released from the sediment. F5 concentrations of the metal elements were not the highest fraction, and this was inconsistent with F5 concentrations in reservoirs of the Delta region of the Cauvery River (Dhanakumar et al., 2014). Notable portions of Mn associated with F2 fractions suggested the affinity of Mn to the carbonate phase.

The Risk Assessment Code (RAC) developed by Perin et al. (1985) is widely used among risk assessment tools. The RAC is based on the percentage of metal associated with F1 and F2 fractions of sediments. As per RAC classification, the sum of F1- and F2-bound fractions is considered to pose no risk to the environment if it is less than 1% of the total metal concentration. Meanwhile, values ranging from 1%–10%, 11%–30%, 31%–50%, and above 50% indicate low, medium, high, and very high risk, respectively (Dhanakumar et al., 2014). In the study of Jinpen Reservoir sediment, Fe levels at all six sites were characterized as high or very high risk, as shown in Table 2. With the exception of S4, Mn concentrations at the other five sites were also characterized as high or very high risk. That is to say, Fe and Mn would have high release risks if the redox condition of the water–sediment interface changed, for example, stratification in the summer.

2.1.4. Sediment pollution from 2011 to 2015

Sediment pollution of Jinpen Reservoir showed an increasing trend over time. From 2011 to 2015, OC, TN, TP, Fe, and Mn in surface sediments increased by 36.4%, 23.3%, 33.0%, 58.7%, and 28.4%, respectively (Table 3). Jinpen Reservoir is a canyon-shaped reservoir, and the sediment pollution source is runoff inflow. However, as a deep stratified reservoir, the anaerobic condition of the bottom water lasts 4 to 5 months (Zhou et al., 2015). Field observations (Table 5) showed that pollutant concentrations in the bottom waters were greater than those of the surface water. As such, sediment release potentials should be explored.

2.2. *In situ reactor experiment*

2.2.1. DO consumption

DO consumption in water and sediment was determined *in situ* at S4, S5, and S6. As shown in Fig. 6a, DO in the *in situ* reactors at S4, S5, and S6 was depleted after 408 hr, 336 hr, and 264 hr. The sum of overlying water and sediment DO consumption rates decreased in the following order: S6 (0.591 mg/L/day) > S5 (0.480 mg/L/day) > S4 (0.419 mg/L/day). DO consumption of the overlying water is shown in Fig. 6b. The DO consumption rate of the overlying water varied in the following sequence: S6 (0.132 mg/L/day) > S5 (0.122 mg/L/day) > S4

Table 3 – The changes in OC, TN, TP, Fe, and Mn concentrations in surface sediments in the main reservoir (S5 and S6) from 2011 to 2015.

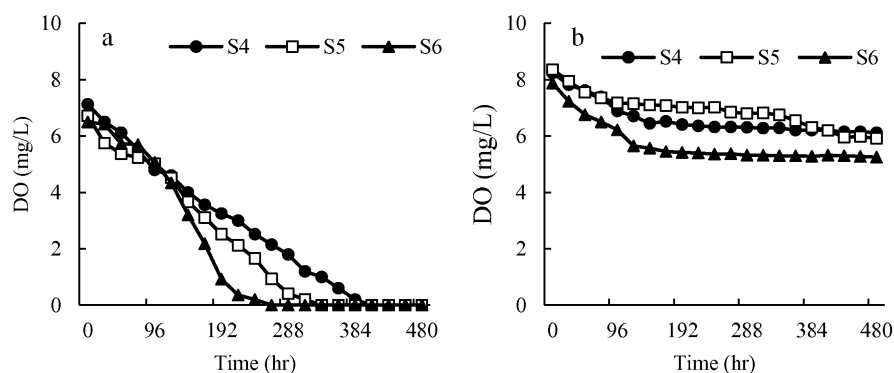| Year/indicator	OC (%)	TN (mg/kg)	TP (mg/kg)	Fe ($\times 10^2$ mg/kg)	Mn (mg/kg)
2011	2.25 ± 0.15	1174 ± 99	694 ± 54	109 ± 9	134 ± 20
2012	2.56 ± 0.12	1269 ± 48	736 ± 62	122 ± 11	142 ± 11
2013	2.75 ± 0.21	1344 ± 105	777 ± 46	139 ± 12	158 ± 15
2014	2.84 ± 0.23	1406 ± 124	847 ± 55	147 ± 10	170 ± 15
2015	3.07 ± 0.18	1447 ± 92	923 ± 69	173 ± 15	172 ± 16

Fig. 6 – Dissolved oxygen (DO) consumption: (a) water and sediment DO consumption (b) overlying water DO consumption.

(0.106 mg/L/day). Accordingly, sediment DO consumption rates at S4, S5, and S6 were 0.313, 0.358, and 0.459 mg/L/day, respectively. These results indicate that the maximum DO consumption rate of water and sediment was in the main reservoir. Along the inflow direction, pollution load increased gradually.

2.2.2. Sediment release
Concentrations of TN, TP, Fe, Mn, and TOC in the overlying water in the in situ reactors were also determined (Fig. 7). TN concentrations of the overlying water in the in situ reactors started to increase as DO was consumed. After 20 days, TN concentrations at S4, S5, and S6 increased from 1.42, 1.52, and 1.51 mg/L to 2.51, 3.33, and 3.05 mg/L, respectively. TP concentrations showed the same trend as TN, and TP maximum

concentrations reached 0.165, 0.202, and 0.224 mg/L. Fe and Mn were also dramatically released from the sediment into the overlying water. At the end of the experiment, Fe concentrations were 0.65, 2.30, and 2.56 mg/L and Mn concentrations were 0.41, 0.61, and 0.52 mg/L at S4, S5, and S6, respectively. TOC concentrations also increased by 53.5%, 74.5%, and 70.8% with respect to initial TOC concentrations in the water.

The experimental results indicated that under anaerobic conditions, pollutants would be released into the overlying water. For example, at S5, compared to initial concentrations, TN, TP, Fe, Mn, and TOC increased by 1.2, 7.1, 9.8, 23.4, and 1.75 times, respectively. Wang et al. (2013) concluded that endogenous pollution is an important contributor to increasing nutrient concentrations and could prompt the outbreak of algal blooms. Acute toxicity tests (Guzmán et al.,
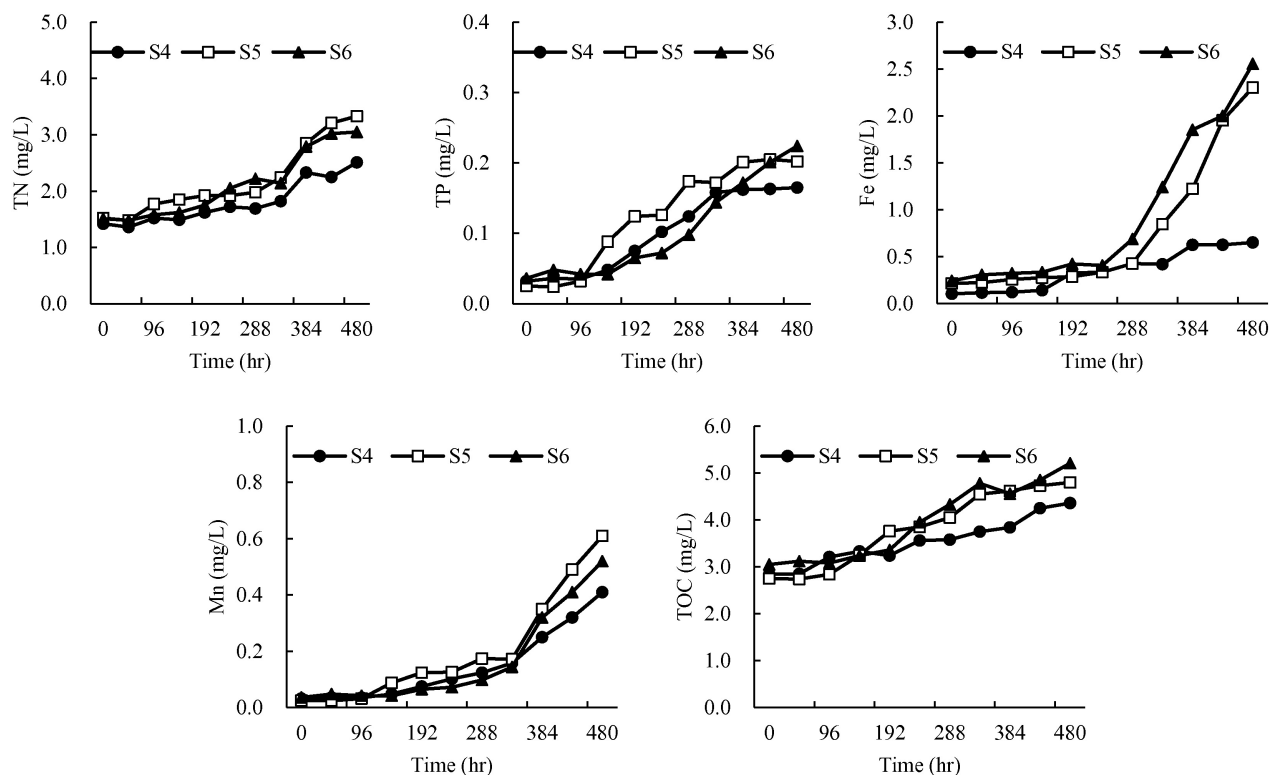

Fig. 7 – Variation of TN, TP, Fe, Mn, and TOC concentrations in the in situ reactors at S4, S5, and S6.
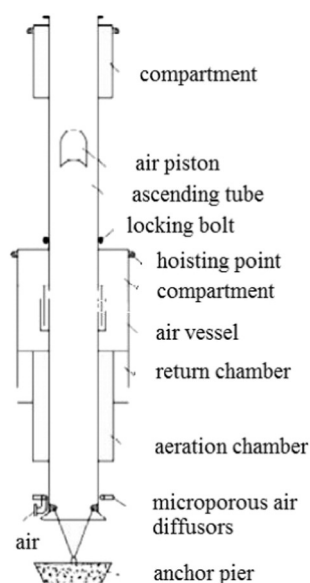
Fig. 8 – The diagram of water-lifting aerator.

2010) indicated that overdosed Fe (>1.0 mg/L) in water could be considered very toxic. Because of the problems caused by N, P, Fe, and Mn in water, *in situ* sediment release control must be taken into consideration. If not, water quality deterioration will occur during the stratification period.

2.3. *In situ sediment pollution control*

Faced with the problems caused by sediment DO consumption and sediment release, a new mixing and oxygenating device called a WLA was designed for *in situ* sediment release control and water quality improvement. It has proved to be

an effective technology, especially in deep and stratified reservoirs (Ma et al., 2015). The diagram of a WLA is shown in Fig. 8. Eight WLAs were installed in Jinpen Reservoir in 2013. From 2013 to 2015, the WLAs were utilized because pollutants started to be released into the overlying water (mainly during September or October).

Before 2013, Jinpen Reservoir was in its natural state; sediment and water quality were unaffected by human disturbance. During the stratification period, the bottom water was in an anaerobic condition. TN, TP, Fe, Mn, and TOC concentrations in the bottom water in 2012 were as high as 2.95, 0.178, 1.78, 0.43, and 4.42 mg/L, respectively. After the natural mixing, however, concentrations of TN, TP, Fe, Mn, and TOC in the surface water increased. This was caused by mixing of the upper and lower portions of the water column.

The mixing function of the WLAs can break up water stratification and increase the concentration of DO in the bottom water. From 2013 to 2015, eight WLAs ran for about 1 month to promote bottom water oxygenation and artificial forced mixing. Operation of the WLAs started at the beginning of the sediment release (when DO content was 0 mg/L in the bottom water, mainly August or September) and ended at the beginning of the mixing period (when the temperature difference of surface water and bottom water was less than 3°C). During the operation of the WLAs in 2015, temperature and DO concentrations varied as shown in Fig. 9. From the beginning of their operation, the temperature of the surface water decreased gradually and temperature of the bottom water increased gradually. After approximately 1 month, the temperature throughout the water column was almost consistent, and the stratification was eliminated. Simultaneously, DO in the bottom water increased to more than 6 mg/L.

During the operation of the WLAs in 2015, OC, TN, TP, Fe, and Mn concentrations in surface sediments at S6 were
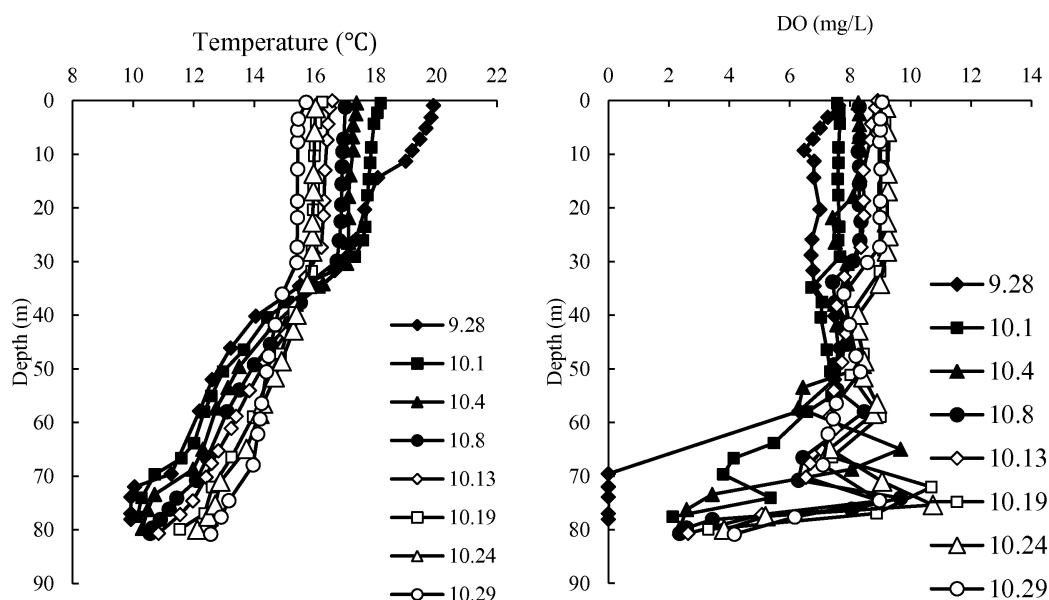

Fig. 9 – Variation of temperature and DO concentration during the operation of WLAs in 2015. DO: dissolved oxygen; WLAs: water-lifting aerators.

Table 4 – The changes of OC, TN, TP, Fe, and Mn concentrations in surface sediment at S6 during the operation of water-lifting aerators (WLAs) in 2015, from triplicate measurements.

Date	OC (%)	TN (mg/kg)	TP (mg/kg)	Fe ($\times 10^2$ mg/kg)	Mn (mg/kg)
2015-09-24	3.17 ± 0.18	1477 ± 92	929 ± 69	169 ± 11	177 ± 14
2015-10-02	2.85 ± 0.09	1328 ± 56	888 ± 72	159 ± 15	168 ± 11
2015-10-13	2.94 ± 0.10	1402 ± 64	802 ± 45	162 ± 9	169 ± 15
2015-10-24	2.81 ± 0.12	1314 ± 66	824 ± 14	158 ± 11	172 ± 12
2015-11-01	2.75 ± 0.14	1252 ± 101	798 ± 24	160 ± 15	170 ± 10

observed five times. As shown in Table 4, following the operation of the WLAs, pollutant concentrations in the surface sediments were reduced. OC, TN, TP, Fe, and Mn concentrations decreased from 3.17%, 1477 mg/kg, 929 mg/kg, 16,900 mg/kg, and 177 mg/kg to 2.75%, 1252 mg/kg, 798 mg/kg, 16,000 mg/kg, and 170 mg/kg. The reduction rates were 13.25%, 15.23%, 14.10%, 5.32%, and 3.94%, respectively. It is clear that after about 1 month of WLA operation, the degree of sediment pollution decreased. A similar study conducted by Zhou et al. (2016) found similar results. The increasing activity of denitrifying bacteria in the surface sediment was the main reason for OC, TN, and TP reduction. The removal efficiency of Fe and Mn in the sediment was not very good.

In the reservoir's natural state, sediment release continued throughout the stratification period. For example, TN increased to 2.45 and 2.95 mg/L in 2011 and 2012 (Table 5), respectively. In the *in situ* reactor, TN increased to 3.05 mg/L (Fig. 7) after 480 hr.

When TN concentrations increased to 1.65, 1.75, and 1.92 mg/L in 2013, 2014, and 2015, respectively, the WLAs began operation. With the increase of DO in the bottom water (Fig. 9), sediment release was limited. After the operation of the WLAs, TN concentration decreased to 1.20, 1.30, and 1.2 mg/L in 2013, 2014, and 2015, respectively. Concentrations of TOC, TN, TP, Fe, and Mn in water were also decreased by 19.6%, 35.0%, 63.2%, 49.3%, and 38.9%. The removal of TOC, N, and P was primarily due to increasing bacterial activity (Zhou et al., 2016). The Fe and Mn control mechanism was to supply DO to the hypolimnion, which is consistent with a previous study (Gantzer et al., 2009). Obviously, sediment release was successfully controlled, and water quality was also improved.

3. Conclusions

Results of this study demonstrate that concentrations of OC, TN, and TP in surface sediments were greater than those in the lower sediments in the main reservoir and in the upstream section. From the reservoir mouth to the main reservoir, OC, TN, and TP concentrations in the sediment showed an increasing trend. In the main reservoir, IEF-N accounted for 43% of the nitrogen fraction, and Fe/Al-P accounted for 26% of the phosphorus fraction. The RAC of metal elements indicated that Fe and Mn in the main reservoir were at high or very high risk levels.

The *in situ* reactor experiment in the main reservoir indicated high release concentrations of TN, TP, Fe, Mn, and TOC. The concentrations reached 3.33, 0.22, 2.56, 0.61 and 5.21 mg/L, respectively. These results were consistent with observations in the natural state in 2011 and 2012.

The operation of WLAs from 2013 to 2015 has been proved to efficiently inhibit sediment release. The operation of WLAs in 2015 resulted in reductions in surface sediment concentrations of OC, TN, TP, Fe, and Mn by 13.25%, 15.23%, 14.10%, 5.32%, and 3.94%, respectively. Concentrations of TOC, TN, TP, Fe, and Mn in overlying water were also reduced by 19.6%, 35.0%, 63.2%, 49.3%, and 38.9%, respectively. WLAs are powerful technologies for controlling the *in situ* release of pollutants in sediments.

Table 5 – Water quality variation at S6 before the reservoir was mixed and after the reservoir was mixed from 2011 to 2015.

Date	TOC (mg/L)		TN (mg/L)		TP (mg/L)		Fe (mg/L)		Mn (mg/L)	
	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom	Surface	Bottom
2011-09-22[b]	3.22	4.15	1.47	2.45	0.019	0.142	0.16	0.85	0.06	0.28
2011-12-28[a]	3.33	3.62	1.63	1.89	0.022	0.056	0.18	0.28	0.09	0.15
2012-09-02[b]	3.14	4.42	1.28	2.95	0.024	0.178	0.15	1.78	0.05	0.41
2012-12-25[a]	3.45	3.65	1.35	1.72	0.032	0.058	0.22	0.39	0.12	0.23
2013-09-12[b]	2.95	3.62	1.32	1.65	0.028	0.104	0.18	0.43	0.05	0.15
2013-10-19[a]	2.85	3.12	1.21	1.22	0.022	0.056	0.16	0.21	0.04	0.06
2014-10-12[b]	3.02	3.67	2.02	1.75	0.021	0.076	0.12	0.37	0.05	0.25
2014-11-02[a]	2.75	3.05	1.32	1.25	0.018	0.022	0.11	0.15	0.05	0.08
2015-09-24[b]	2.88	3.56	1.68	1.92	0.024	0.112	0.12	0.57	0.05	0.13
2015-10-30[a]	2.68	3.22	1.12	1.22	0.018	0.032	0.13	0.22	0.05	0.06

"b" means before the reservoir was mixed and "a" means after the reservoir was mixed. In 2011 and 2012, "mix" refers to natural mixing. In 2013, 2014, and 2015, "mix" refers to the artificial forced mixing by WLAs.

REFERENCES

Bo, M.H., Ming, S.J., Xia, L.X., Mao, Y.H., 2003. Nitrogen forms and their functions in recycling of the Bohai Sea sediments. Geochimica 32 (1), 48–54.

Cade-Menun, B.J., 2005. Characterizing phosphorus in environmental and agricultural samples by 31P nuclear magnetic resonance spectroscopy. Talanta 66 (2), 359–371.

Dhanakumar, S., Solaraj, G., Mohanraj, R., 2014. Heavy metal partitioning in sediments and bioaccumulation in commercial fish species of three major reservoirs of river Cauvery delta region, India. Ecotoxicol. Environ. Saf. 113C (113C), 145–151.

Gantzer, P.A., Bryant, L.D., Little, J.C., 2009. Controlling soluble iron and manganese in a water-supply reservoir using hypolimnetic oxygenation. Water Res. 43 (5), 1285–1294.

Guzmán, F.T., González, F.J.A., Martínez, R.R., 2010. Implementing *Lecane quadridentata* acute toxicity tests to assess the toxic effects of selected metals (Al, Fe and Zn). Ecotoxicol. Environ. Saf. 73 (3), 287–295.

Huang, T., Xuan, L., Rijnaarts, H., Grotenhuis, T., Ma, W., Xin, S., Xu, J., 2014. Effects of storm runoff on the thermal regime and water quality of a deep, stratified reservoir in a temperate monsoon zone, in Northwest China. Sci. Total Environ. 485–486 (3), 820–827.

Huang, L., Fang, H., Reible, D., 2015. Mathematical model for interactions and transport of phosphorus and sediment in the Three Gorges Reservoir. Water Res. 85, 393–403.

Jie, F., Zhao, C., Luo, Y., Liu, C., Kyzas, G.Z., Yin, L., Zhao, D., An, S., Zhu, H., 2014. Heavy metals in surface sediments of the Jialu River, China: their relations to environmental factors. J. Hazard. Mater. 270 (3), 102–109.

López, P., López-Tarazón, J.A., Casas-Ruiz, J.P., Pompeo, M., Ordoñez, J., Muñoz, I., 2015. Sediment size distribution and composition in a reservoir affected by severe water level fluctuations. Sci. Total Environ.

Ma, W., Huang, T., Li, X., 2015. Study of the application of the water-lifting aerators to improve the water quality of a stratified, eutrophicated reservoir. Ecol. Eng. 83, 281–290.

Młynarczyk, N., Bartoszek, M., Polak, J., Sułkowski, W.W., 2013. Forms of phosphorus in sediments from the Goczałkowice Reservoir. Appl. Geochem. 37 (37), 87–93.

Nayar, S., Goh, B., Chou, L., 2004. Environmental impact of heavy metals from dredged and resuspended sediments on phytoplankton and bacteria assessed in in situ mesocosms. Ecotoxicol. Environ. Saf. 59, 349–369.

Ni, Z., Wang, S., 2015. Historical accumulation and environmental risk of nitrogen and phosphorus in sediments of Erhai Lake, Southwest China. Ecol. Eng. 79, 42–53.

Nikolai, S.J., Dzialowski, A.R., Nikolai, S.J., Dzialowski, A.R., 2014. Effects of internal phosphorus loading on nutrient limitation in a eutrophic reservoir. Limnologica 49, 33–41.

Perin, G., Craboledda, L., Lucchese, M., Cirillo, R., Dotta, L., Zanette, M.L., Orio, A.A., 1985. Heavy metal speciation in the sediments of Northern Adriatic Sea—a new approach for environmental toxicity determination. In: Lekkas, T.D. (Ed.), Heavy Metal in the Environment. CEP Consultants, Edinburgh, pp. 454–456.

Qian, G., Chen, W., Lin, T., Chui, P., 2009. In-situ stabilization of Pb, Zn, Cu, Cd and Ni in the multi-contaminated sediments with ferrihydrite and apatite composite additives. J. Hazard. Mater. 170, 1093–1100.

Ruban, V., Brigault, S., Demare, D., Philippe, A.M., 1999. An investigation of the origin and mobility of phosphorus in freshwater sediments from Bort-Les-Orgues Reservoir, France. J. Environ. Monit. 1 (4), 403–407.

Ruttenberg, K.C., 1992. Development of a sequential extraction method for different forms of phosphorus in marine sediments. Limnol. Oceanogr. 37 (7), 1460–1482.

Souza, V.A., Wasserman, J.C., 2015. Distribution of heavy metals in sediments of a tropical reservoir in Brazil: sources and fate. J. S. Am. Earth Sci. 63, 208–216.

Sun, S.J., Huang, S.L., Sun, X.M., Wen, W., 2009. Phosphorus fractions and its release in the sediments of Haihe River, China. J. Environ. Sci. (China) 21 (3), 291–295 (in Chinese).

Tessier, A., Campbell, P.G.C., Bisson, M., Chem, A., 1979. Sequential extraction procedure for the speciation of particulate trace metals. Anal. Chem. 51 (7), 844–851.

Trojanowska, A.A., Izydorczyk, K., 2010. Phosphorus fractions transformation in sediments before and after cyanobacterial bloom: implications for reduction of eutrophication symptoms in dam reservoir. Water Air Soil Pollut. 211 (1–4), 287–298.

Wang, S., Li, X.J., Yang, S., Jin, X., Yi, W., 2012. Effects of organic matter and submerged macrophytes on variations of alkaline phosphatase activity and phosphorus fractions in lake sediment. J. Environ. Manag. 113 (1), 355–360.

Wang, L., Liu, L., Zheng, B., 2013. Eutrophication development and its key regulating factors in a water-supply reservoir in North China. J. Environ. Sci. 25 (5), 962–970.

Wang, L., Yuan, X., Zhong, H., Wang, H., Wu, Z., Chen, X., Zeng, G., 2014. Release behavior of heavy metals during treatment of dredged sediment by microwave-assisted hydrogen peroxide oxidation. Chem. Eng. J. 258 (258), 334–340.

Wu, D., Hua, Z., 2014. The effect of vegetation on sediment resuspension and phosphorus release under hydrodynamic disturbance in shallow lakes. Ecol. Eng. 69 (69), 55–62.

Xu, D., Ding, S., Sun, Q., Zhong, J., Wu, W., Jia, F., 2012. Evaluation of in situ capping with clean soils to control phosphate release from sediments. Sci. Total Environ. 438, 334–341.

Xuelu, G., Chen-Tung Arthur, C., 2012. Heavy metal pollution status in surface sediments of the coastal Bohai Bay. Water Res. 46 (6), 1901–1911.

Yao, X., Wang, S., Jiao, L., Yan, C., Jin, X., 2015. Resolving the influence of nitrogen abundances on sediment organic matter in macrophyte-dominated lakes, using fluorescence spectroscopy. J. Environ. Sci. 27 (1), 197–206.

Zahra, A., Hashmi, M.Z., Malik, R.N., Ahmed, Z., 2013. Enrichment and geo-accumulation of heavy metals and risk assessment of sediments of the Kurang Nallah-feeding tributary of the Rawal Lake Reservoir, Pakistan. Sci. Total Environ. 470–471c (2), 925–933.

Zhang, L., Wang, S., Wu, Z., 2014. Coupling effect of pH and dissolved oxygen in water column on nitrogen release at water–sediment interface of Erhai Lake, China. Estuar. Coast. Shelf Sci. 149, 178–186.

Zhou, Z., Huang, T., Ma, W., Li, Y., Zeng, K., 2015. Impacts of water quality variation and rainfall runoff on Jinpen Reservoir, in Northwest China. Water Sci. Eng. 8 (4), 301–308.

Zhou, S., Huang, T., Zhang, H., Zeng, M., Liu, F., Bai, S., Shi, J., Qiu, X., Yang, X., 2016. Nitrogen removal characteristics of enhanced in situ indigenous aerobic denitrification bacteria for micro-polluted reservoir source water. Bioresour. Technol. 201, 195–207.