



A method for determining the treatment dosage of drinking water treatment residuals for effective phosphorus immobilization in sediments



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ABSTRACT

Drinking water treatment residuals (WTRs), non-hazardous by-products generated during treatment of drinking water, can be reused to immobilize phosphorus (P) in sediments for eutrophication control. Prior to application, it is necessary to determine the treatment dosage of WTR to immobilize P in sediments. In the present study, the dosage of Al/Fe based WTRs required to effectively immobilize P in lake sediments was determined in a laboratory setting as a function of the oxalate-extractable Al (Al_{ox}) and Fe (Fe_{ox}) content of the WTRs by completely mixing Al/Fe based WTRs with Lake Baiyangdian sediments. The results showed that the capacity for immobilization of mobile P (P_m) in sediments by Al_{ox} and Fe_{ox} in Al/Fe based WTRs ($(Al_{ox} + Fe_{ox})_{WTR}$) can be described by the equation: $(Al_{ox} + Fe_{ox})_{WTR} = 83 * P_m - 40$ (where Al_{ox} , Fe_{ox} and P_m are expressed in $\mu mol g^{-1}$). This equation was successfully used to determine the respective quantities of three different Al/Fe based WTRs required to immobilize P in sediments from Lake Chaohu, Lake Taihu, the Changjiang River, the Haihe River, the Pear River and the Yellow River. Using the equation derived herein, the required Al/Fe based WTR application dose can easily be determined where the P_m content of the sediment and the Al_{ox} and Fe_{ox} content of the WTR are known.

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1. Introduction

Excessive phosphorus (P) in water bodies (e.g. lakes, rivers, etc.) can lead to eutrophication, and internal loading from P-enriched sediments is one of the primary origins of excessive P (Haggard and Soerens, 2006; Cyr et al., 2009; Jin et al., 2013). At present, *in situ* P immobilization is a comparatively new technology for remediating P-enriched sediments (Paller and Knox, 2010). *In situ* P immobilization utilizes chemically reactive materials to sequester P in non-labile forms within sediments, thereby reducing internal

P loading in water bodies (Yuan et al., 2009; Egemose et al., 2010). The selection of materials with high P sorption capacity and low environmental toxicity is critical to the successful application of *in situ* P immobilization technology (Paller and Knox, 2010). Materials such as Fe and Al salts are often used due to their high P immobilization capacities (Hansen et al., 2003; Reitzel et al., 2005, 2006). Despite their effectiveness, a low-cost alternative to Fe and Al salts is sought to overcome financial constraints to widespread use, particularly where the application of a large quantity of P immobilizing material is required.

Drinking water treatment residuals (WTRs) are ubiquitous, non-hazardous by-products generated by drinking water treatment plants (Babatunde and Zhao, 2007; Ippolito et al., 2011). They are primarily comprised of Fe/Al hydroxides because Fe and Al salts are routinely used to remove suspended solids and humic substances from raw water. The Fe and Al in WTRs are primarily amorphous and exhibit high P adsorption capacity (Li et al., 2013). It has been demonstrated that WTRs can be used as soil amendments to mitigate off-site P pollution (Agyin-Birikorang et al., 2009) or as media for constructed wetlands to remove excessive P in wastewater (Park, 2009; Zhao et al., 2009). Nevertheless, WTRs are generally regarded as waste materials (Babatunde and Zhao, 2007). Therefore, the development of novel productive uses for WTRs is of

Abbreviations: Al_{ox} , oxalate-extractable Al; BD-P, $Na_2S_2O_4/NaHCO_3$ extractable P; BJ1-WTR and BJ2-WTR, WTR from Beijing City No. 9 Waterworks; Fe_{ox} , oxalate-extractable Fe; HCl-P, HCl extractable P; HZ-WTR, WTR from Hanzhou City Tap Water Plant; ICP-AES, inductively coupled plasma atomic emission spectroscopy; Lake B, Lake Baiyangdian; Lake C, Lake Chaohu; Lake T, Lake Taihu; NaOH-P, NaOH extractable P; NH_4Cl -P, NH_4Cl extractable P; P, phosphorus; P_m , mobile P; River C, Changjiang River; River H, Haihe River; River P, Pear River; River Y, Yellow River; TP, total P; WTRs, drinking water treatment residuals.

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particular interest. Recently, the authors verified that Al/Fe based WTRs can be used to effectively immobilize P in sediments (Wang et al., 2012a). The investigation also showed that amendment of sediments with Al/Fe based WTRs did not detrimentally affect bacterial diversity or abundance. Thus, Al/Fe based WTRs may be ideal materials for sediment P immobilization to reduce internal P loading in water bodies. The use of recycled Al/Fe based WTRs as a substitute for Fe and Al salts for eutrophication control could also enhance the economic feasibility of *in situ* sediment P immobilization.

Prior to widespread application of Al/Fe based WTRs, it is necessary to determine the required dosage of the WTR for effective sediment P immobilization. The recommended treatment for P-enriched lake sediments is the addition of Al (usually as alum) with the application dose based on alkalinity such that the applied dose of Al results in a mixture with pH 6–6.3, which is optimum for creating P-sorbing Al(OH)₃ flocs (Cooke et al., 2005; Rydin and Welch, 1998). Despite their high Al content, this method of determining dosage is not applicable to Al/Fe based WTRs due to the variable P immobilization capacity of Al/Fe based WTRs depending on their source and physico-chemical characteristics (Ippolito et al., 2011). Previous studies have shown that the P sorption capacity of Al/Fe based WTRs is closely related to their oxalate-extractable Al and Fe (Al_{ox} and Fe_{ox}) content (Dayton and Basta, 2005). Therefore, it may be possible to determine the required dosage of Al/Fe based WTRs for P immobilization in enriched sediments based upon the Fe_{ox} and Al_{ox} content of the WTR used and the quantity of mobile P (P_m) in the sediments.

In the present study, a mathematical equation to estimate the dosage of Al/Fe based WTR required to immobilize sediment P_m was derived based on the Al_{ox} and Fe_{ox} content of each WTR using P-enriched sediments from Lake Baiyangdian (Lake B). The equation was validated using several different Al/Fe based WTRs at the respective determined dosages to immobilize P_m in sediments from Lake Chaohu (Lake C), Lake Taihu (Lake T), the Changjiang River (River C), the Haihe River (River H), the Pear River (River P) and the Yellow River (River Y). The results reported herein are based on laboratory investigations; thus, additional work is necessary to verify these results in *in situ* field trials. However, the results of the present study provide a simple means of estimating the quantity of Al/Fe based WTRs required to reduce internal P loading and control eutrophication in water bodies with P-enriched sediments.

2. Materials and method

2.1. Sampler collection

Drinking water treatment residuals were collected from Beijing City No. 9 Waterworks (BJ1- and BJ2-WTRs) and the Hanzhou City Tap Water Plant (HZ-WTR) in China. At the Beijing City No. 9 Waterworks, Al and Fe salts were used as coagulants to purify raw influent water whereas the Hanzhou City Tap Water Plant used only Al salts. The BJ1-WTR was collected in April 2010; BJ2-WTR and HZ-WTR were collected in April 2012. The Al/Fe based WTRs were air-dried, ground and sieved to a diameter <1 mm. Sediments were collected from Lake B (38°53' N, 115°59' E), Lake C (31°35' N, 117°51' N), Lake T (31°24' N, 120°60' E), River C (31°24' N, 121°44' E), River H (38°59' N, 117°43' E), River P (22°48' N, 113°37' E) and River Y (34°54' N, 113°40' E) between May 2011 and April 2012. One sediment sample was obtained from the upper 10 cm of the sediments at each location using a Petersen grab sampler. The sediment samples were passed through a 1.8 mm screen to remove impurities, homogenized by stirring and stored in acid-cleaned polyethylene flasks at 4.0 °C.

Additional details concerning WTR and sediment collection sites are provided in the Supplemental Information (Fig. S1).

2.2. Experimental procedure

Immobilization of P_m in Lake B sediments was examined by thoroughly mixing each Al/Fe based WTR with the wet sediments at rates of approximately 0%, 1%, 2%, 4%, 6%, 8%, 10%, 12%, 14%, 16% and 20% (w/w) on a dry weight basis. The WTR/sediment mixtures were placed into a series of brown glass bottles, which were capped and incubated in a constant-temperature culture chamber at 15 °C without aeration or further mixing (Wang et al., 2012a). After incubation, the WTR/sediment mixtures were freeze-dried, ground and passed through a standard 100-mesh (0.15 mm) sieve prior to further testing. The contents of Fe_{ox}, Al_{ox} and P_m in the Lake B sediment and in the WTR/sediment mixtures were determined and a mathematical equation derived to express the relationship between the decreased P_m and the increased Al_{ox} and Fe_{ox} content of sediments amended with Al/Fe based WTRs. Because the Al_{ox} and Fe_{ox} content of each WTR/sediment mixture was primarily attributable to the Al/Fe based WTR added to the sediments, the derived equation largely reflects the capacity of Al_{ox} and Fe_{ox} in the WTRs to immobilize P_m in sediments.

The equation derived using sediments from Lake B was validated using sediments from Lake C, Lake T, River C, River P and River Y. First, the Al_{ox} and Fe_{ox} content of each Al/Fe based WTR and the P_m content of each sediment sample were determined. The quantity of each Al/Fe based WTR required to immobilize P_m in each sediment was determined based on the measured quantity of P_m in each sediment and the Al_{ox} and Fe_{ox} content of the WTR. The WTRs were then mixed with each sediment at the calculated dosage by stirring and the mixtures were incubated at 15 °C for 10 d. After incubation, the P_m in each sediment sample was analyzed to evaluate the effectiveness of Al/Fe WTR addition for immobilization of P_m in the sediments.

2.3. Sample analysis

The total contents of Al and Fe in the WTRs were determined according to Method 3050B (USEPA, 1996). The total P contents of the WTRs and sediments were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES, ULTIMA, JY, France) after the solids were digested in an HNO₃–HF–HClO₄ mixture. The Al_{ox} and Fe_{ox} contents of Al/Fe WTRs, sediments, and WTR-amended sediments were determined by ICP-AES after the solids were extracted in the dark at a 1:60 solid:solution ratio (g mL⁻¹) using a buffered solution 0.2 M ammonium oxalate and 0.2 M oxalic acid adjusted to pH 3 (Shang and Zelazny, 2008).

Sediment P was fractionated according to the method described by Hupfer et al. (1995). Briefly, 1.0 g subsamples of each sediment were sequentially extracted with 25 mL of 1.0 M NH₄Cl for 4 h (NH₄Cl-P), 0.11 M Na₂S₂O₄/0.11 M NaHCO₃ (BD) for 2 h (BD-P), 1.0 M NaOH for 16 h (NaOH-P) and 0.5 M HCl for 16 h (HCl-P), all at 25 °C. Solutions from the BD extraction were stored overnight at 4.0 °C prior to P analysis to oxidize residual reagent (Pilgrim et al., 2007). In each extraction step, the solid residues were separated from the supernatants by centrifugation. Supernatants were filtered using 0.45 μm cellulose acetate membranes, and the P in each solution was quantified using the molybdenum blue technique (Murphy and Riley, 1962). Extractable P in sediments is operationally defined as the sum of the NH₄Cl-P, BD-P, NaOH-P and HCl-P fractions. Of these fractions, NH₄Cl-P is considered immediately available or loosely sorbed. Phosphorus within the BD-P fraction is comprised of P bound to surfaces of redox-sensitive Fe and Mn oxides and (oxy)hydroxides. The NaOH-P fraction contains the

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