



Effect of low molecular weight organic acids on phosphorus adsorption by ferric-alum water treatment residuals

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ABSTRACT

Effects of low molecular weight organic acids (LMWOAs; citric acid, oxalic acid and tartaric acid) on phosphorus (P) adsorption by ferric-alum water treatment residuals (FARs) were studied. Both batch and column experiments indicated that the effects of LMWOAs on P adsorption were closely related to adsorption time. Initially, all acids presented inhibitory function on P adsorption. The inhibition became weaker with time, eventually promoting P adsorption for citric acid and tartaric acid. In the column experiment with a 61-day duration, high P adsorption rates (>55%) were observed for the test groups containing citric acid and tartaric acid. Interestingly, higher pH likely enhanced P adsorption with the effects of LMWOAs and a distinct relationship between LMWOAs' effects on P adsorption and their concentrations was not observed. Moreover, fractionation of the adsorbed P from the FARs demonstrated that oxalic acid reduced P adsorption capacity, while citric acid and tartaric acid increased. Based on the forms of Fe and Al existing in the FARs and Fourier transform infrared spectroscopy analyses, LMWOAs can promote P adsorption through activating crystalline Fe/Al and preventing crystallization of amorphous Fe/Al to increase P adsorption sites, and can also inhibit P adsorption by competition with adsorption sites.

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1. Introduction

Water treatment residuals (WTRs) have been widely studied because of their strong phosphorus (P) adsorption capability [1–6]. Therefore, to improve the recycling of P adsorption material, some researchers have also focused on factors that can affect P adsorption by WTRs. For example, particle size, pH, adsorption time, temperature and P species have been reported to affect P adsorption on WTRs [7,8]. Among them, pH showed the greatest influence, affecting both the amount of P adsorption and the processes of P adsorption [9]. In addition, P adsorption capability of WTRs was not affected by the aging of WTRs [10,11]. Even under different redox systems, WTRs still performed with high P adsorption ability [12]. More attention should be paid to the effects of environmental factors on P adsorption by WTRs during their application.

Low molecular weight organic acids (LMWOAs) are distributed widely in nature. They are mainly derived from the decomposition of organic matter and secretion of plant roots and microbes, and they always include oxalic acid, citric acid, tartaric acid, malic acid, succinic acid, salicylic acid, p-hydroxybenzoic acid and maleic acid, among others. The concentrations of LMWOAs are usually low in

natural environment [13,14]. The facts that LMWOAs have adverse effects on P adsorption by soil and can also increase the amount of exchangeable Al from soil have been reported [15–17]. However, in some special circumstances, they can exist at high levels, which is detrimental to the stability of P in the environment. In wetlands or soils where WTRs are reused, LMWOAs produced by plants and microbes during their metabolism may affect the process of P adsorption, but little attention has been paid to these aspects. Therefore, the effects of LMWOAs on P adsorption were studied in detail in this work, which could supply the basis for constructed wetlands or soil remediation.

In this study, batch experiments and column experiments based on ferric-alum water treatment residuals (FARs) were employed to study the characteristics of P adsorption and how they were affected by citric acid, oxalic acid and tartaric acid. Moreover, the mechanisms of P adsorption perturbation by LMWOAs were also inferred by analyzing structures and components of FARs in column experiments.

2. Materials and methods

2.1. Materials

The FARs were taken from the dewatering workshop of the 9th Water Supply Plant of Beijing, China, in April 2010. Samples were

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air-dried and ground to pass through a sieve in order to provide test adsorbent with a diameter less than 1 mm (it was denoted as Raw-FARs). Citric acid, oxalic acid and tartaric acid were selected in this work, and their concentrations were determined according to the previous studies [15–18].

2.2. Batch experiments

Samples of 0.5 g were put into a series of 50 ml polyethylene bottles, and 25 ml solutions containing 0.01 M KCl, 500 mg/L P (KH_2PO_4) and a certain concentration of LMWOs were then added. All samples were kept at a specified pH and shaken for 2, 5 and 10 days. Then, the samples were immediately centrifuged at $5000 \times g$ for 10 min and filtered through 0.45 μm Millipore filter paper. The P concentration was analyzed by the ammonium molybdate spectrometry method. P adsorption without LMWOs was also run as a control group. Triplicate samples were taken and the result was represented as their average. The standard error deviation was within 5% in the batch experiments. Ippolito et al. suggested that a 10-day shaking period should be used in batch tests to ensure complete P adsorption by water treatment residuals (WTRs) [19]. This suggestion was adopted in the present study though the shaking time is commonly set up to 2 days in many batch tests related to P adsorption by WTRs [5,9].

The effect of LMWOs concentrations on P adsorption was studied at pH 7. Concentrations of LMWOs were kept at 3, 6, 9 or 25 mmol/L. Meanwhile, the pH values of 5, 7 and 9 were also tested with 6 mmol/L LMWOs.

2.3. Column experiments

Four columns (i.d.: 25 mm, height: 500 mm) were packed with 100 g of FARs with approximately 3 cm quartz sand at the bottom of these columns to prevent loss of FARs. On one hand, three columns were arranged to determine the effects of LMWOs on P adsorption by the FARs. A synthetic solution (pH 7) containing 50 mg of P/L (as KH_2PO_4) [20] and 6 mmol/L LMWOs was supplied continuously at a rate of approximately 720 ml/day using a constant-head feeding tank. Water in the storage tank was renewed every day. On the other hand, the rest one column was employed as a control group. Influent and effluent were sampled every day during the first 15 days and then collected every two days for the remaining 46 days. Both dissolved reactive P (DRP) and total reactive P (TRP) were determined. All tests were repeated twice and the average values were reported. The standard error deviation was within 1%.

2.4. Residuals analyses

After the column experiments, all FARs were dried at 105°C and mixed together for further analysis. Total Fe and Al contents of the samples were determined using inductively coupled emission spectroscopy (ICP-AES, ULTIMA, JY) according to USEPA-METHOD 3050B [21]. Oxalate-extractable P, Fe and Al were determined by ICP-AES after extracting in darkness at a 1:60 solid:solution ratio [2]. A sequential extraction procedure, which divides P into Al-bound P (Al-P), Fe-bound P (Fe-P), occluded-P (O-P) and Ca-bound P (Ca-P), was used for the extraction of inorganic P after adsorption [22]. Fourier transform infrared spectroscopy (FTIR; Nexus 670, Nicolet) was used to examine the structure of the FARs.

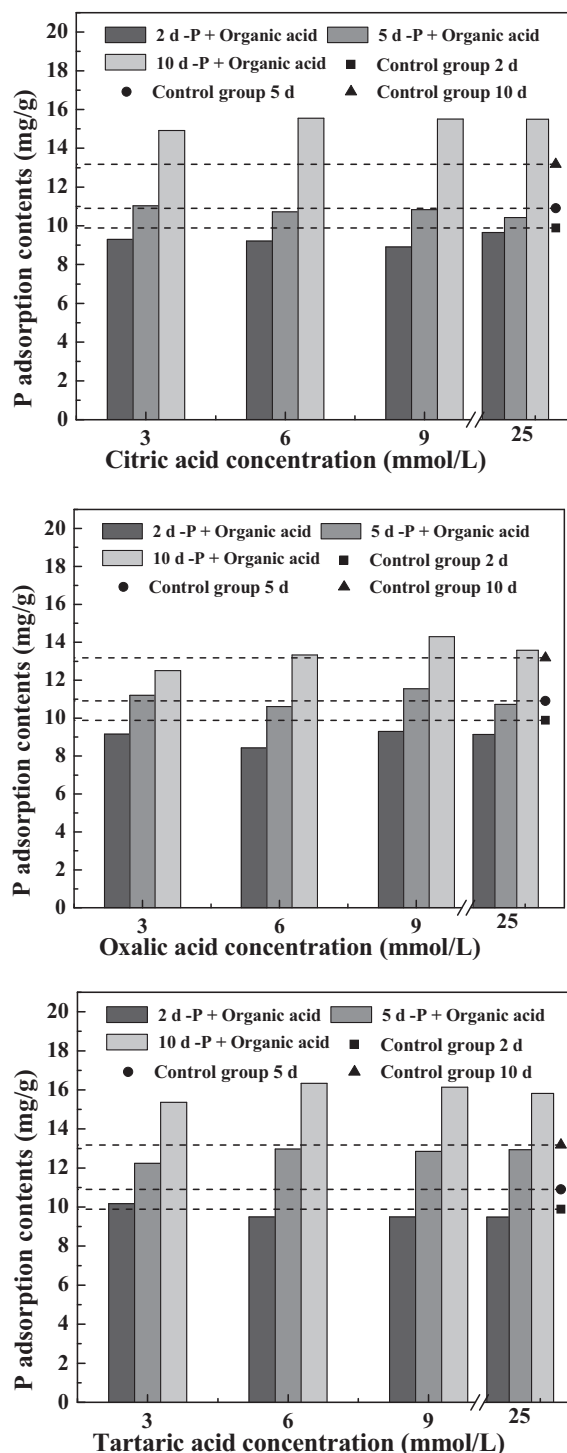


Fig. 1. Effects of LMWOs on P adsorption under different concentrations.

3. Results

3.1. Batch experiments

3.1.1. Effect of LMWOs under different concentrations

The results of the effect of LMWOs on P adsorption under different concentrations are shown in Fig. 1. Compared with the control group, citric acid and oxalic acid showed a strong inhibitory action while tartaric acid did a weak inhibition within the first 2 days. Furthermore, the inhibition effects of citric acid and oxalic acid became weaker while tartaric acid promoted P adsorption

at the 5th day. Both citric acid and tartaric acid were favorable for P adsorption at the 10th day, whereas oxalic acid still kept the inhibition. More than 2.18–3.05 mg/g P had been adsorbed by the FARs with tartaric acid. No distinct relationship between the actions and concentrations of LMWOAs was observed in the experiments, even at the concentration of 25 mmol/L. However, much higher concentrations (i.e. >25 mmol/L) probably lead to different results, whereas such high concentrations would diverge from the real situation.

3.1.2. Effect of LMWOAs under different pH

The results of the effect of LMWOAs on P adsorption under different pH values are shown in Fig. 2. Similar trends for P adsorption with and without LMWOAs were observed under different pH values though lower pH values were more favorable to P adsorption. Compared with the control group, LMWOAs showed inhibitory action at the 2nd day, in which tartaric acid presented the weakest inhibition (Fig. 2c). As pH increased, the inhibitory action became weaker for all acids. However, the effect of LMWOAs changed from inhibition to promotion with pH increasing at the 5th and 10th day. The strongest promotion function by tartaric acid was observed, followed by citric acid (Fig. 2a) and oxalic acid (Fig. 2b). Therefore, the inhibition of LMWOAs on P adsorption by FARs weakened with increasing pH.

3.2. Column experiments

The results of column experiments are illustrated in Fig. 3. There was a similar distribution pattern between DRP and TRP (data not shown) in outflow. Compared with the control group, all acids exhibited different inhibitory actions on P adsorption during the initial stage (Fig. 3a). At the 9th day (Fig. 3b), however, inhibition by citric acid and tartaric acid was transformed into promotion. In the citric acid group, P removal rate declined slowly after the 9th day, but the rate rose again at the 21st day. The P removal rate was increased in the tartaric acid group on the 9th day, and ascended from 72.23% (on the 9th day) to 93.20% (on the 23rd day). Over the whole experiment (61 days), both the citric acid group and the tartaric acid group maintained a high removal rate of P (>55%), while the control group saturated. Additionally, the oxalic acid group showed an inhibitory action on P adsorption until the 33rd day, while no obvious promotion or inhibition was observed since then. Moreover, out of all the groups, only the citric acid group was blocked on the 41st day. Therefore, when FARs are applied as P adsorption material in different environments, attention should be paid to the problem of blocking.

Consequently, the effects of citric acid, oxalic acid and tartaric acid on P adsorption by FARs were closely related to adsorption time. Citric acid and tartaric acid mainly promoted P adsorption, especially tartaric acid, while oxalic acid inhibited P adsorption.

3.3. Residuals analysis

Extraction results of different forms of P are shown in Fig. 4. It can be seen that FARs in the tartaric acid group adsorbed the largest amount of P, followed by the citric acid group, the control group and the oxalic acid group. Compared with the control group, it can be found that the FARs in citric acid group and tartaric acid group adsorbed more than 3.65 and 3.98 mg/g P, respectively, while it was decreased by 1.17 mg/g in the oxalic acid group. These results indicated that citric acid and tartaric acid promoted P adsorption while oxalic acid inhibited it. Furthermore, Fe–P and Al–P were found to be the main forms existing in FARs after adsorption. Compared with the contents of Fe–P and Al–P in the control group, both species were more in the tartaric acid and citric acid groups but lower in the oxalic acid group. Moreover, the amounts of total inorganic P

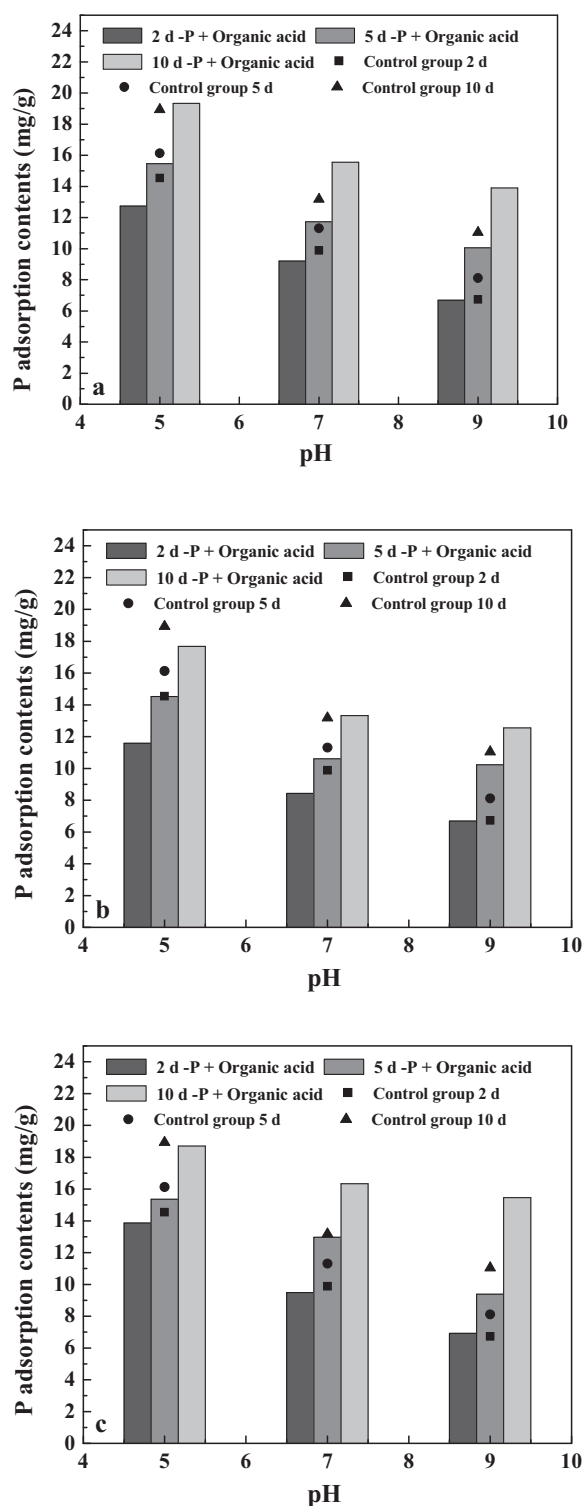


Fig. 2. Effect of LMWOAs on P adsorption under different pH conditions. (a) Citric acid, (b) oxalic acid and (c) tartaric acid.

from sequential extraction and oxalate-extractable P were not significantly different, which indicated that most of the adsorbed P existed in FARs were amorphous forms.

The existence of both Al and Fe in the residuals resulted in significantly higher P adsorption capability. Therefore, the effects of LMWOAs on the forms of Al and Fe were also investigated. Levels of different forms of Al and Fe of FARs in different groups are displayed in Fig. 5. In all groups, approximately $92.08 \pm 3.65\%$ of total Al was

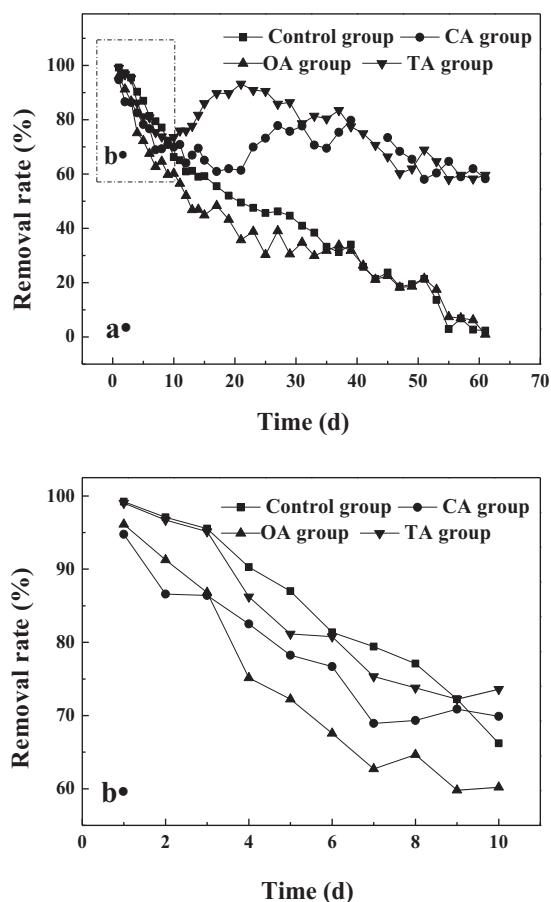


Fig. 3. Removal efficiency of DRP in the experimental columns. (a) During the entire 61 days and (b) in the first 10 days. The CA group, OX group and TA group are citric acid group, oxalic acid group and tartaric acid group, respectively. The CA group was blocked on the 41st day, and was stopped for 2 days.

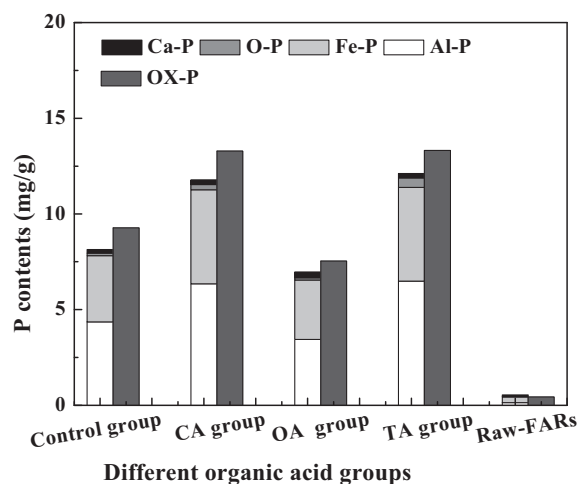


Fig. 4. Extraction results of different forms of P. OX-P is oxalate-extractable P. The CA group, OX group and TA group are citric acid group, oxalic acid group and tartaric acid group, respectively.

oxalate-extractable, while only $63.91 \pm 3.53\%$ of total Fe could be extracted by oxalate. In addition, total amounts of Al in Raw-FARs were 3.14 and 3.89 mg/g higher than those in the citric acid and tartaric acid groups, respectively. Meanwhile, total amounts of Fe in Raw-FARs were 3.03 and 7.65 mg/g higher than those in the citric acid and the tartaric acid groups, respectively. However, no visible

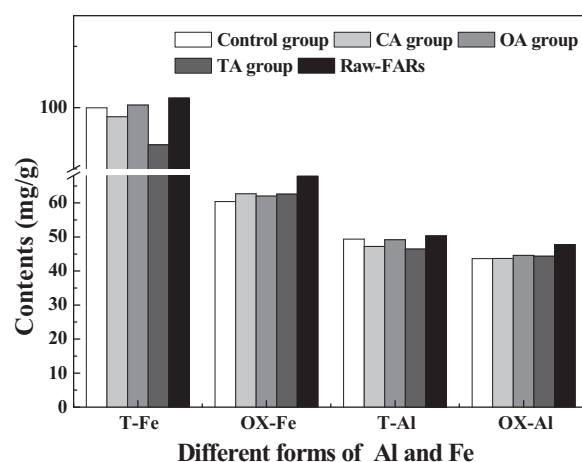


Fig. 5. Contents of different forms of Fe and Al in different groups. The CA group, OX group and TA group are citric acid group, oxalic acid group and tartaric acid group, respectively. OX is oxalate-extractable; T-Fe/Al is the total contents of Fe/Al.

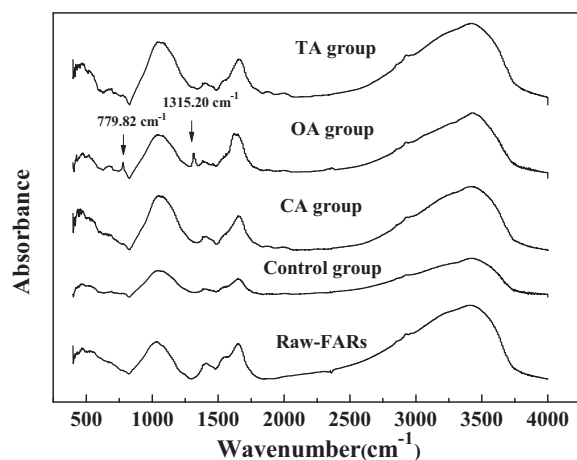


Fig. 6. Results of FTIR analysis of FARS in different groups. The CA group, OX group and TA group are citric acid group, oxalic acid group and tartaric acid group, respectively.

differences in total Fe or total Al were observed among Raw-FARs and the FARS in the oxalic acid group and the control group. These results indicated that citric acid and tartaric acid could promote the loss of Fe and Al from FARS. Additionally, the amounts of oxalate-extractable Fe and Al in all groups were lower than in Raw-FARs. And the amounts of oxalate-extractable Fe and Al in the citric acid, oxalic acid and tartaric acid groups were higher than those in the control group.

Results of FTIR analysis of FARS in different groups are shown in Fig. 6. For the FARS with citric acid or tartaric acid, the patterns of the FTIR spectra after adsorption were similar to those before adsorption, thus the citric acid and tartaric acid had the weak ability of competing for adsorption sites with P. However, for the FARS with oxalic acid, the FTIR pattern presented two specific peaks at 1315.20 cm⁻¹ and 779.82 cm⁻¹ after adsorption. The peaks at 1317 cm⁻¹ represented oxalate corresponding to the C=O stretching frequencies [23]. The peak 780 cm⁻¹ was also considered as a characteristic peak of oxalate [24]. Thus the two peaks at 1315.20 cm⁻¹ and 779.82 cm⁻¹ presented oxalate after adsorption, indicating that oxalic acid strongly competed with P for adsorption sites in the FARS.

4. Discussion

The ability of $\text{Al}(\text{OH})_3$ to adsorb P in FARs can be reduced due to its crystallization in lake water [25]. In addition, amorphous Fe can also crystallize under these some conditions (pH 6, 2–6 weeks under 40–60 °C) [26]. Compared with Fe and Al hydroxides, FARs might have different but similar characteristics in structure and P adsorption mechanism. Agyin-Birikorang and O'Connor [27] found that incubation of fresh Al-WTR was more effective than field drying for stabilizing the most reactive Al in Al-WTR. Therefore, crystallization of reactive Fe/Al caused the amounts of oxalate-extractable Fe/Al to decrease in all groups (similar to wet incubation), which were below the values observed for Raw-FARs (similar to field drying).

LMWOAs not only can activate crystal Fe/Al [28,29] but also can inhibit the crystallization of amorphous Fe/Al [30,31]. On one hand, LMWOAs can bind with amorphous Fe/Al to form Fe/Al complex compound which inhibit crystallization of Fe and Al in the FARs; on the other hand, LMWOAs can change the crystal Fe and Al in FARs into the amorphous forms through acidization [30,32]. As a result, crystallization of Fe and Al in the FARs weakened in the LMWOA groups, causing the amounts of oxalate-extractable Fe/Al in the control group to be lower than those in LMWOA groups. Moreover, the loss of Fe/Al in the citric acid and tartaric acid groups might result from the strong acidization effects.

Previous studies showed that the larger amount of oxalate-extractable-Fe/Al in WTRs was responsible for the stronger P adsorption ability [33]. As was shown in batch and column experiments, the inhibitory actions of LMWOAs only occurred in the initial stage of P adsorption. The reason for this phenomenon might be that more P adsorption sites were occupied by the LMWOAs [15,16] than that increased by activation effects of LMWOAs on Fe/Al in FARs at the beginning [28]. Oxalic acid had the most obvious inhibitory action because it has the strongest ability to compete with P for adsorption sites (Fig. 6). However, activation of crystal Fe/Al increased with time, which in turn increased the number of adsorption sites for P. Therefore, oxalic acid showed neither inhibition nor promotion (after the 33rd day). For citric acid and tartaric acid, however, little Fe/Al was lost, but the total amount of amorphous Fe/Al was higher than that in the control group, which helped to add adsorption sites for P. Furthermore, citric acid and tartaric acid only weakly competed for adsorption sites (Fig. 6), and thus, both groups had an increasing trend of higher P adsorption.

Results of the inorganic P sequential extraction of FARs showed that all LMWOAs in this study simultaneously inhibited or promoted the combination of P with Fe and Al in FARs. Therefore, it was deduced that LMWOAs should show similar effects on P adsorption for other WTRs (Fe-WTRs or Al-WTRs).

The mechanism of LMWOAs' effects on P adsorption by FARs could be divided into two aspects: (1) inhibition caused by competition for adsorption sites and (2) promotion due to the activation of crystalline Fe/Al and the prevention of the crystallization of Fe/Al in FARs. The dominant action depended on the properties of the specific LMWOAs. In this study, oxalic acid could inhibit P adsorption through a stronger ability to compete for adsorption sites. However, citric acid and tartaric acid could improve P adsorption because of their stronger abilities for activation and weak ability to compete for adsorption sites. Furthermore, the fact that higher pH values were more beneficial for Al activation was demonstrated in the study of Li and Xu [17] and Kodama and Schnitzer [34], which might explain the stronger promotion of LMWOAs under high pH conditions.

However, on one hand, Hu et al. argued that organic acids were not helpful for P adsorption by some variable charge soils [35]. Qualls et al. found that natural dissolved organic matter could inhibit P adsorption by ferric chloride and aluminum sulfate [36].

On the other hand, Borggaard et al. considered that organic acids could inhibit the aging process of Al in some sandy soil, resulting in high maximum P adsorption capacities [37]. Gerke found that humic substances could initially inhibit P adsorption by poorly ordered Fe-oxide, whereas the inhibition became weaker with time and finally changed to promotion of P adsorption [38]. These divergences likely arose from different experimental conditions such as adsorption time, adsorbents and organic matters. Gerke found that after 55 day humic substances were beneficial to the adsorption [38]. The favorable effects of LMWOAs on P adsorption in this study were presented after 5 days in batch tests and 9 days in column experiments. Nevertheless, the adsorption time set in previous studies was no more than 5 days. It was not long enough to make the promotion functions of organic acids happen fully and only the inhibition effects can be observed. Moreover, adsorbents such as FARs, contain some crystal Fe and Al. These Fe and Al can be transformed to the amorphous forms by LMWOAs, causing the adsorption sites to increase. But if adsorbents had no crystal but amorphous Fe and Al (e.g. ferric and aluminum salts), there would have been no adsorption sites that could be increased by organic matters. As a result, organic matters could only occupy the P adsorption sites, presenting the inhibition function. Finally, different types of organic acids could express various effects on P adsorption by the FARs owing to their specified chemical characteristics. Further work is needed to clarify these effects in detail.

5. Conclusion

This work was designed to study the effects of LMWOAs on P adsorption by the FARs. Results revealed that the effects of LMWOAs changed from inhibition to promotion with increase of adsorption time. With pH rising, the inhibitory action of LMWOAs on P adsorption weakened and was also gradually changed to promotion. No distinct relationship between the actions and concentrations of LMWOAs was observed. LMWOAs could inhibit or promote the combination of P with Fe and Al in the FARs. The inhibitory action could rise from competition of adsorption sites by LMWOAs while the promotion action could result from the inhibition of crystallization of amorphous Fe/Al by LMWOAs. The results could be significant to the recycling and reusing of the FARs.

Acknowledgments

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References

- [1] G.K. Hausteine, T.C. Daniel, D.M. Miller, P.A. Moore, R.W. McNew Jr., Aluminum-containing residuals influence high-phosphorus soils and runoff water quality, *J. Environ. Qual.* 29 (2000) 1954–1959.
- [2] M.K. Gibbons, G.A. Gagnon, Understanding removal of phosphate or arsenate onto water treatment residual solids, *J. Hazard. Mater.* 186 (2011) 1916–1923.
- [3] Y.Q. Zhao, A.O. Babatunde, Y.S. Hu, J.L.G. Kumar, X.H. Zhao, Pilot field-scale demonstration of a novel alum sludge-based constructed wetland system for enhanced wastewater treatment, *Process Biochem.* 46 (2011) 278–283.
- [4] W.H. Park, Integrated constructed wetland systems employing alum sludge and oyster shells as filter media for P removal, *Ecol. Eng.* 35 (2009) 1275–1282.
- [5] M.M. Mortula, G.A. Gagnon, Alum residuals as a low technology for phosphorus removal from aquaculture processing water, *Aquacult. Eng.* 36 (2007) 233–238.
- [6] A.O. Babatunde, Y.Q. Zhao, Equilibrium and kinetic analysis of phosphorus adsorption from aqueous solution using waste alum sludge, *J. Hazard. Mater.* 184 (2010) 746–752.
- [7] Y. Yang, Y.Q. Zhao, A.O. Babatunde, Y.X. Ren, Y. Han, Characteristics and mechanisms of phosphate adsorption on dewatered alum sludge, *Sep. Purif. Technol.* 51 (2006) 193–200.
- [8] M. Razali, Y.Q. Zhao, M. Bruen, Effectiveness of a drinking-water treatment sludge in removing different phosphorus species from aqueous solution, *Sep. Purif. Technol.* 55 (2007) 300–306.

- [9] Y. Yang, D. Tomlinson, S. Kennedy, Y.Q. Zhao, Dewatered alum sludge: a potential adsorbent for phosphorus removal, *Water Sci. Technol.* 54 (2006) 207–213.
- [10] S. Agyin-Birikorang, G.A. O'Connor, Lability of drinking water treatment residuals (WTR) immobilized phosphorus: aging and pH effects, *J. Environ. Qual.* 36 (2007) 1076–1085.
- [11] Y. Yang, Y.Q. Zhao, P. Kearney, Influence of ageing on the structure and phosphate adsorption capacity of dewatered alum sludge, *Chem. Eng. J.* 145 (2008) 276–284.
- [12] I.W. Oliver, C.D. Grant, R.S. Murray, Assessing effects of aerobic and anaerobic conditions on phosphorus sorption and retention capacity of water treatment residuals, *J. Environ. Manage.* 92 (2011) 960–966.
- [13] D.L. Jones, Organic acids in the rhizosphere—a critical review, *Plant Soil* 205 (1998) 25–44.
- [14] B.W. Strobel, Influence of vegetation on low-molecular-weight carboxylic acids in soil solution—a review, *Geoderma* 99 (2001) 169–198.
- [15] N.S. Bolan, R. Naidu, S. Mahimairaja, S. Baskaran, Influence of low-molecular-weight organic acids on the solubilization of phosphates, *Biol. Fertil. Soils* 18 (1994) 311–319.
- [16] S. Staunton, F. Leprince, Effect of pH and some organic anions on the solubility of soil phosphate: implications for P bioavailability, *Eur. J. Soil Sci.* 47 (1996) 231–239.
- [17] J.Y. Li, R.K. Xu, G.L. Ji, Dissolution of aluminum in variably charged soils as affected by low-molecular-weight organic acids, *Pedosphere* 15 (2005) 484–490.
- [18] K. Kpombekou-A., M.A. Tabatabai, Effect of low-molecular weight organic acids on phosphorus release and phytoavailability of phosphorus in phosphate rocks added to soils, *Agric. Ecosyst. Environ.* 100 (2003) 275–284.
- [19] J.A. Ippolito, K.A. Barbarick, H.A. Elliott, Drinking water treatment residuals: a review of recent uses, *J. Environ. Qual.* 40 (2011) 1–12.
- [20] D.C. Seoa, J.S. Chob, H.J. Lee, J.S. Heo, Phosphorus retention capacity of filter media for estimating the longevity of constructed wetland, *Water Res.* 39 (2005) 2445–2457.
- [21] USEPA, Method 3050B: Acid Digestion of Sediment, Sludge and Soils, second ed., Governmental Printing Office, Washington, DC, 1996.
- [22] A.E. Cox, J.J. Camberato, B.R. Smith, Phosphate availability and inorganic transformation in an alum sludge-affected soil, *J. Environ. Qual.* 26 (1997) 1393–1398.
- [23] M.A. Gabal, A.A. El-Bellihi, S.S. Ata-Allah, Effect of calcination temperature on Co(II) oxalate dihydrate–iron (II) oxalate dihydrate mixture: DTA-TG, XRD, Mössbauer, FT-IR and SEM studies (Part II), *Mater. Chem. Phys.* 81 (2003) 84–92.
- [24] F. Pinzari, M. Zotti, A.D. Mico, P. Calvini, Biodegradation of inorganic components in paper documents: formation of calcium oxalate crystals as a consequence of *Aspergillus terreus* Thom growth, *Int. Biodeterior. Biodegrad.* 64 (2010) 499–505.
- [25] J. Berkowitz, M.A. Anderson, C. Amrhein, Influence of aging on phosphorus sorption to alum floc in lake water, *Water Res.* 40 (2006) 911–916.
- [26] R.G. Ford, P.M. Bertch, K.J. Farley, Changes in transition and heavy metal partitioning during hydrous iron oxide aging, *Environ. Sci. Technol.* 31 (1997) 2028–2033.
- [27] S. Agyin-Birikorang, G.A. O'Connor, Aging effects on reactivity of an aluminum-based drinking-water treatment residual as a soil amendment, *Sci. Total Environ.* 407 (2007) 826–834.
- [28] J.Y. Li, R.K. Xu, S.C. Xiao, G.L. Ji, Effect of low-molecular-weight organic anions on exchangeable aluminum capacity of variable charge soils, *J. Colloid Interface Sci.* 284 (2005) 393–399.
- [29] W.P. Miller, L.W. Zelazny, D.C. Martens, Dissolution of synthetic crystalline and noncrystalline iron oxides by organic acids, *Geoderma* 37 (1986) 1–13.
- [30] R.M. Cornell, U. Schwertmann, Influence of organic anions on the crystallization of ferrihydrite, *Clays Clay Miner.* 27 (1979) 402–410.
- [31] K.F.N.K. Kwong, P.M. Huang, Influence of citric acid on the hydrolytic reactions of aluminum, *Soil Sci. Soc. Am. J.* 41 (1977) 692–697.
- [32] S.A. Welch, W.J. Ullman, The effect of organic acids on plagioclase dissolution rates and stoichiometry, *Geochim. Cosmochim. Acta* 57 (1993) 2725–2736.
- [33] E.A. Dayton, N.T. Basta, A method for determining the phosphorus sorption capacity and amorphous aluminum of aluminum-based drinking water treatment residuals, *J. Environ. Qual.* 34 (2005) 1112–1118.
- [34] H. Kodama, M. Schnitzer, Effect of fulvic acid on the crystallization of aluminum hydroxides, *Geoderma* 24 (1980) 195–205.
- [35] H.Q. Hu, J.Z. He, X.Y. Li, F. Liu, Effect of several organic acids on phosphate adsorption by variable charge soils of central China, *Environ. Int.* 26 (2001) 353–358.
- [36] R.G. Qualls, L.J. Sherwood, C.J. Richardson, Effect of natural dissolved organic carbon on phosphate removal by ferric chloride and aluminum sulfate treatment of wetland waters, *Water Resour. Res.* 45 (2009) W09414.
- [37] O.K. Borggaard, S.S. Jørgensen, J.P. Moberg, B. Raben-Lange, Influence of organic matter on phosphate adsorption by aluminium and iron oxides in sandy soils, *J. Soil Sci.* 41 (1990) 443–449.
- [38] J. Gerke, Phosphate adsorption by humic/Fe-oxide mixtures aged at pH 4 and 7 and by poorly ordered Fe-oxide, *Geoderma* 59 (1993) 279–288.