Improving Removal Efficiency of Organic Matters by Adding Phosphorus in Drinking Water Biofiltration Treatment

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Objective To investigate phosphorus limitation and its effect on the removal efficiency of organic matters in drinking water biological treatment. **Methods** Bacterial growth potential (BGP) method and a pair of parallel pilot-scale biofilters were used for the two objectives, respectively. **Results** The addition of phosphorus could substantially increase the BGPs of the water samples and the effect was stronger than that of the addition of carbon. When nothing was added into the influents, both COD_{Mn} removals of the parallel biofilters (BF1 and BF2) were about 15%. When phosphate was added into its influent, BF1 performed a COD_{Mn} removal, 6.02 percentage points higher than the control filter (BF2) and its effluent had a higher biological stability. When the addition dose was <20 g·L^{-1}, no phosphorus pollution would occur and there was a good linear relationship between the microbial utilization of phosphorus and the removal efficiency of organic matters. **Conclusions** Phosphorus was a limiting nutrient and its limitation was stronger than that of carbon. The addition of phosphate was a practical way to improve the removal efficiency of organic matters in drinking water biological treatment.

Key words: Drinking water; Biofiltration; Phosphorus; Limiting nutrient; Organic matters; Biological stability

INTRODUCTION

Pollution of raw water is a common problem for the water plants in China[1] and almost half of the cities and towns in this country are confronted with the embarrassed situation that no clear water sources are available[2]. Organic matters are the main pollutants in the sources[1]. The bioavailable organic matters in drinking water may cause bacterial regrowth in distribution system from which a series of problems are derived such as the pipe corrosion and epidemiological risks[3-5]. In a conventional treatment process, i.e. coagulation, sedimentation, filtration and disinfection, a larger dose of disinfectant is usually taken to control the bacterial regrowth, but this may lead to another serious trouble, i.e. the decline of the genetically toxicological safety because humic substances, the main component of natural organic matters (NOM), can react with the disinfectant as the precursor of the disinfection byproducts (DBPs), and consequently the levels of the mutagenic or carcinogenic DBPs such as trihalomethanes (THMs) and haloacetic acids (HAAs) are increased[6-8].

Biofiltration is often undertaken now as an enhanced process. But for the low concentration

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and biodegradability of the substrates, the removal rate of organic matters indicated as COD_{Mn} is rather low, often below 20%-30%[1, 9, 10]. If ozonation is applied proceeding biofiltration, the removal rate of organic matters will be significantly increased[11-13], but most water plants in China can not afford its high expense, so new ways to lower its cost should be found.

The nutrient levels in source water are usually so low that, even polluted, the microbes in biofilters are in an oligotrophic environment. Carbon was usually considered to be the most important limiting nutrient. The widely accepted concepts and measurements of BDOC and AOC are established on this hypothesis[14, 15]. But recent researches revealed that phosphorus might play a limiting role, too. Miettinen *et al.*[3, 16] found that natural waters in the northern hemisphere generally had a high content of organic carbon and the microbial growth in drinking water in Finland was highly regulated not only by organic carbon but also by the availability of phosphorus; Nishijima *et al.*[17] found that the biodegradation rate of glucose by biological activated carbon (BAC) was much higher if phosphorus was absorbed by BAC or added into the influent; Sathasivan *et al.*[18] reported the evident phosphorus limitation on the bacterial regrowth in the distribution systems of Tokyo.

In this study, the limiting effect of phosphorus in the biofiltration process was demonstrated using the bacterial growth potential (BGP) method which was simple and could find the limiting nutrient quickly. It was also demonstrated that the addition of phosphate could improve the removal efficiency of organic matters and the biological stability of the effluent in the biofiltration for drinking water treatment.

MATERIALS AND METHODS

The Pilot-scale Biofiltration Process

Two parallel GAC-sand dual media biofilters (BF1 and BF2, Fig. 1) were set up in a water plant located in Huai River basin in the east of China. Both of the filters were cylinders of 3.0 m in height and 0.15 m in internal diameter. The water levels were 2.5 m high from the bottoms of the reactors. The media in them were 0.9 m high in total with 0.55 m granular activated carbon (GAC) and 0.35 m sand, respectively. A hydraulic loading of 7.5-8.5 $m \cdot h^{-1}$, corresponding to an empty bed contact time (EBCT) of 6.4-7.2 min, and a backwashing cycle of 24 h were performed by both reactors.

The study was divided into two stages. At the first stage, no nutrients were added into each reactor; at the second stage, KH_2PO_4-P was added into BF1 to investigate the effect of phosphorus on the removal of organic matters of the reactor, which was realized by preparing a solution of KH_2PO_4 at certain concentration in a tank and controlling its flow (Fig.1.12). The influent added with phosphate could be sampled from a sampling port (Fig.1.4) and the exact PO_4^{3-}-P concentration could be determined. BF2 was the control filter and no phosphorus was added. The addition amount of phosphate was referred to the difference of the PO_4^{3-}-P concentration between the influent with and without the addition of phosphate.

The effluent of the full-scale sedimentation tank of the plant was used in this study as the influent. The water quality is shown in Table 1.

When this study was carried out, the two reactors had been in performance for nearly 8 months, and steady removals of various pollutants had been reached, so the biofilm developed very well and was under a pseudo-steady state, i.e. the newly reproduced biomass was equal to the declined one.

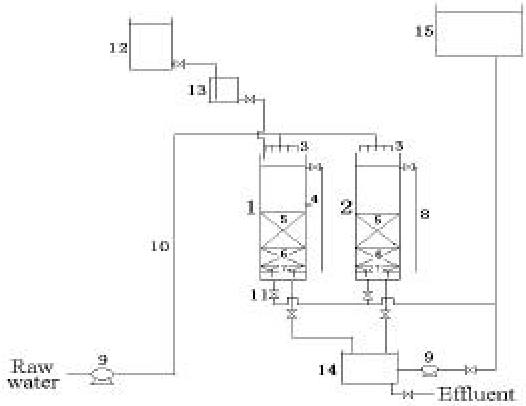

FIG.1. Scheme of the pilot-scale biological filters. 1. Biological filter1 (BF1); 2. Biological filter2 (BF2); 3. Water distributor; 4. Sampling port; 5. Granular activated carbon; 6. Sand; 7. Grave layer; 8. Overflow pipe; 9. Magnetic lifting pump; 10. Distribution pipe; 11. Valve; 12. Phosphate-adding tank; 13. Constant-level tank; 14. Effluent collector; 15. Backwashing water tank

TABLE 1

Quality of the Influent (2001.7.3-2001.10.12)

	COD_{Mn} (mg·L^{-1})	NH_4^+-N (mg·L^{-1})	NO_2^--N (mg·L^{-1})	PO_4^{3-}-P (g·L^{-1})	Turbidity (ntu)	Temperature (℃)	DO (mg·L^{-1})	pH
Maximum	5.40	0.30	0.07	32.00	5.94	31.50	8.66	8.20
Minimum	2.25	0.07	0.00	8.83	1.73	19.70	4.01	7.74
Average	3.88	0.18	0.01	15.01	3.51	27.15	6.75	7.91

Analysis Method

All the water quality indexes mentioned in Table 1 were measured using Chinese national standard methods[19, 20].

Bacterial Growth Potential (BGP) Method

Bacterial growth potential (BGP) was used to assess the phosphorus limitation and the biological stability of the samples and was measured mainly as the method described by Sathasivan and Ohgaki[18]. In their method, an expensive instrument, epifluorescence microscope, was used for the direct total microbial counting, while in this study, the heterotrophic plate counting (HPC) was taken for the microbial counting, so the measurement could be accomplished under basic laboratory equipment. Another change lied in the dose of phosphate in determining BGP(All) and BGP(P); in their methods, about 300 g·L^{-1} KH_2PO_4-P (1 320 g·L^{-1} KH_2PO_4) was required, while this dose was only 50 g·L^{-1} KH_2PO_4-P in ours, for the latter was high enough to reveal the phosphorus limitation and convenient to compare the limiting effect of this element with that of carbon.

Glassware Preparation

As the levels of available nutrients for the microbes in the samples were very low, all the glassware should be treated by the following approaches to be free from pollution: (i) dipping in detergent overnight, (ii) washing by tap water and pure water, respectively, (iii) dipping in diluted acid (HCl) solution overnight, (iv) washing by tap water and pure water, respectively, (v) heating overnight at 210℃.

Sample Preparation

A 1 000 mL sampling bottle was used to collect enough water sample, and different inorganic nutrients or $C_6H_{12}O_6$ were added into the bottle at the doses showed in Table 2 according to different requirements. BGP(All) meant the bacterial growth potential of the sample into which various inorganic nutrients including phosphorus were added. BGP(P) and BGP(C) meant the bacterial growth potentials of the samples into which 50 g • L^{-1} KH_2PO_4-P and 20 mg • L^{-1} $C_6H_{12}O_6$ were added, respectively. BGP(n) meant the bacterial growth potential of the sample into which no nutrient was added. The samples were transferred into 18 mm×150 mm test tubes, 10 mL per tube. For each result, at least one parallel was necessary. All the test tubes were sterilized by autoclaving at 121℃, 15 min and then stored at 4℃.

TABLE 2

Nutrients Added When Different BGPs Were Measured

BGP	Compound	Concentration (g • L^{-1})
BGP(All)	KNO_3	1011.0
	KH_2PO_4 (KH_2PO_4-P)	219.4 (50.0)
	Na_2SO_4	450.0
	$CaCl_2 \cdot 2H_2O$	185.0
	$MgCl_2 \cdot 6H_2O$	415.0
	$FeCl_3 \cdot 6H_2O$	245.0
	$CoCl_2 \cdot 6H_2O$	20.4
	$CuCl_2 \cdot 2H_2O$	27.1
	$MnSO_4 \cdot 5H_2O$	1109.5
	$ZnCl_2$	10.6
	$(NH_4)_6Mo_7O_{24} \cdot 4H_2O$	1.0
BGP(P)	KH_2PO_4 (KH_2PO_4-P)	219.4 (50.0)
BGP(C)	$C_6H_{12}O_6$	20 000

Inoculation, Incubation and Microbial Counting

The effluent of the full-scale sedimentation tank was collected into several 18 mm×150 mm test tubes at 10mL per tube as the inoculum simultaneous with sampling. The test tubes were then incubated at 20℃ for 5 days to ensure that the limiting nutrients in the water had been reduced to the minimum, other nutrients had been reduced to the possible minimum and the microbes had been in the stationary phase and adopted to the water. The incubated inoculum was transferred into the test tubes with water samples at a dose of 2 mL per tube. The inoculated tubes were incubated at 20℃ for 5 days, then the biomass in the samples were counted with heterotrophic plate counting (HPC) and BGP was expressed as the result of HPC with CFU • mL^{-1} as the unit.

RESULT

Demonstration of Phosphorus Limitation and Comparison of the Limitation of Phosphorus and Carbon

Effects of phosphorus and the other inorganic elements on the BGPs of the influent were compared (Fig. 2). After the addition of phosphorus and other inorganic nutrients mentioned in Table 2, BGP(All) could rise from 4.07×10^3 CFU • mL^{-1} (BGP(n)) to 6.40×10^3 or 6.78×10^3 CFU • mL^{-1}. However, BGP(P) could rise to 6.67×10^3 CFU • mL^{-1} when phosphorus alone was added. There was no significant difference between BGP(All) and BGP(P), which suggested that the levels of all the inorganic elements except phosphorus were high enough in the influent and no one but phosphorus should have limiting effect on the microbial growth and metabolism in the biological process treating the influent.

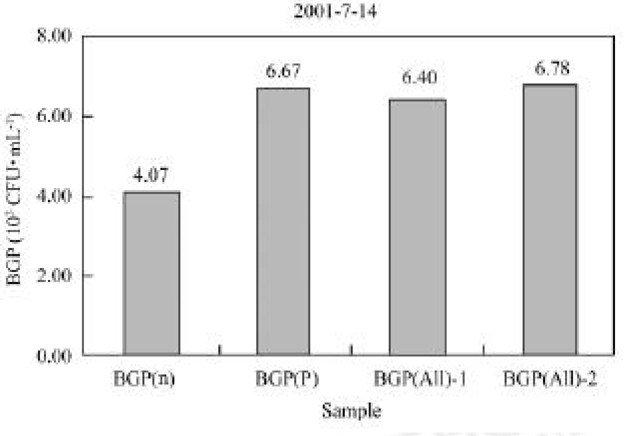

FIG. 2. Comparison of BGP(n), BGP(P) and BGP(All).

More details of phosphorus limitation could be seen in Fig. 3. During the six determinations from July 2001 to October 2001, BGP(P) was 54% higher than BGP(n) on average with the highest increase of 64% and the lowest increase of 41%, respectively.

Carbon was also a limiting nutrient just as it was considered to be (Fig. 4). BGP(C)s were 43% higher than BGP(n) in four determinations on average. But compared with that of phosphorus, its limitation seemed weaker. The average of two BGP(P)s measured simultaneously was about 12% higher than that of BGP(C)s.

Effect of the Addition of Phosphate on the Removal Efficiency of Organic Matters and the Biological Stability of the Effluent

At the first stage of the study, no nutrients were added into the influent, and no significant difference was observed between the removal efficiency of organic matters of BF1 and BF2 (Fig. 5). The average COD_{Mn} concentration of the influent was 3.40 mg • L^{-1}, and the effluents of BF1 and BF2 were 2.91 mg • L^{-1} and 2.85 mg • L^{-1} on average, respectively. The removal curves of the two reactors interwove together, and the average removals were 14.13% and 16.49%, respectively. BF2 removed a little more organic matters than BF1, 2.36 percentage points higher in removal.

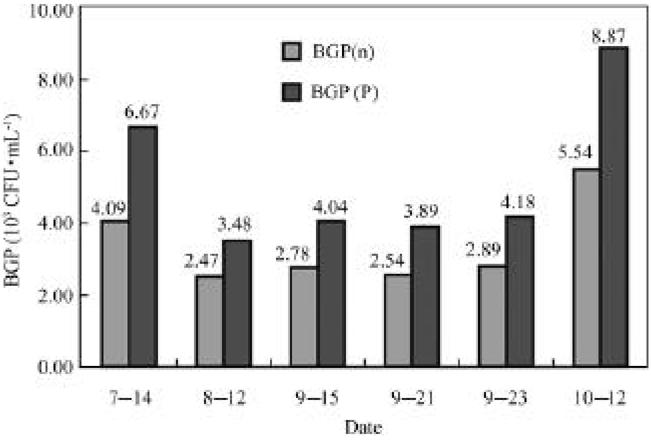

FIG. 3. Limiting effect of phosphorus on BGP.

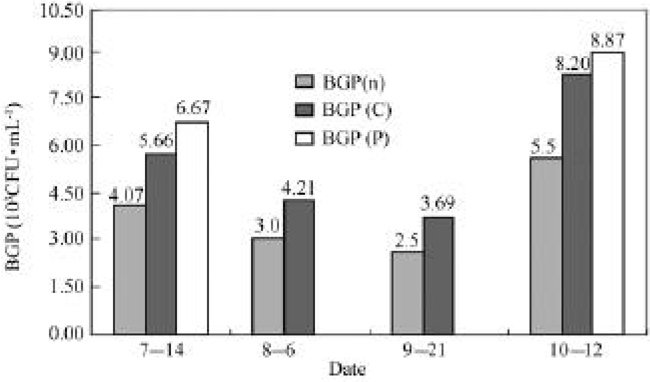

FIG. 4. Carbon limitation and comparison of the limitation of phosphorus and carbon.

At the second stage of the study, the removal of organic matters of BF1 was obviously increased with KH_2PO_4-P added into its influent (Fig. 6). It could be seen in Fig. 6 that almost all the points in the removal rate curve of BF1 were higher than those of BF2. At this stage, the average COD_{Mn} concentration of the influent was 4.24 mg • L^{-1}; the concentrations of the effluents of BF1 and BF2 were 3.36 mg • L^{-1} and 3.61 mg • L^{-1}, respectively, and 0.25 mg • L^{-1} COD_{Mn} was removed by BF1 more than by BF2. The removal rate of BF2 was only 14.54%, while that of BF1 was 20.56%, 6.02 percentage points higher than that of BF2.

Effect of the addition of phosphorus on the biological stability of the effluent of the biofilter was shown in Fig. 7. The BGP(n)s of the influent and the two effluents were measured simultaneously for several times. The sequence of the BGP(n) averages was that the influent (4.26×10^3 CFU • mL^{-1}) > BF2 effluent (2.08×10^3 CFU • mL^{-1}) > BF1 effluent (1.11×10^3 CFU • mL^{-1}), which indicated that the effluent of BF1 had the highest biological stability.

Phosphorus Levels in the Effluents

The effect of the addition of phosphate on the PO_4^{3-}-P concentration of the effluent was investigated (Table 3).

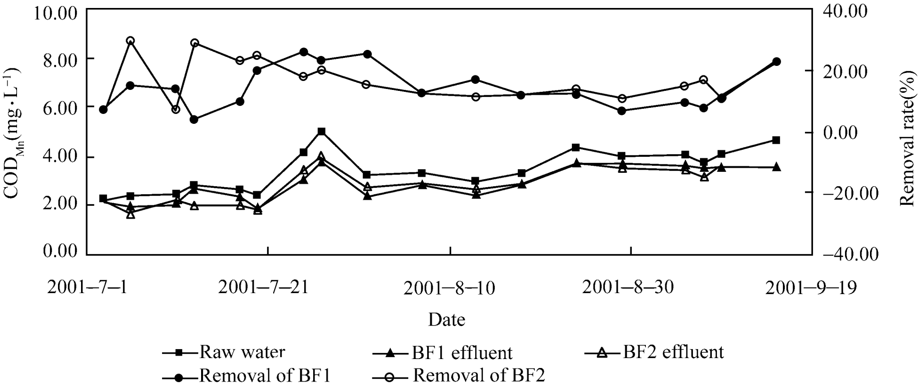

FIG. 5. Removals of COD_{Mn} by the two reactors with no phosphate added.

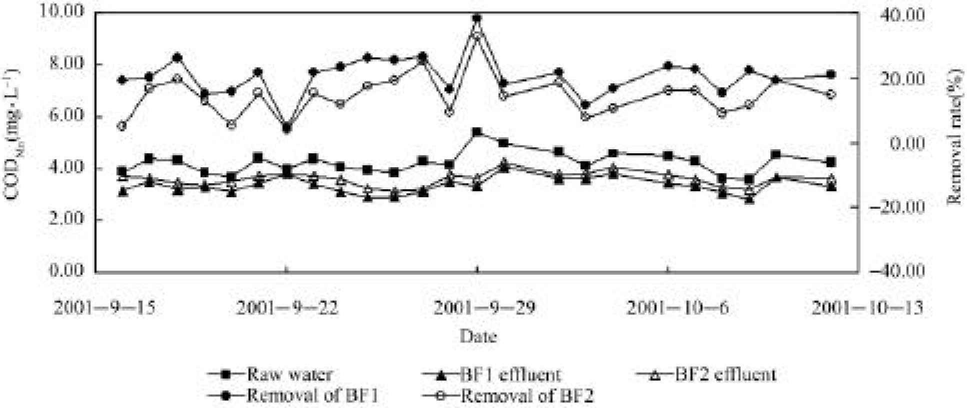

FIG. 6. Removals of COD_{Mn} by the two reactors with phosphate added into BF1.

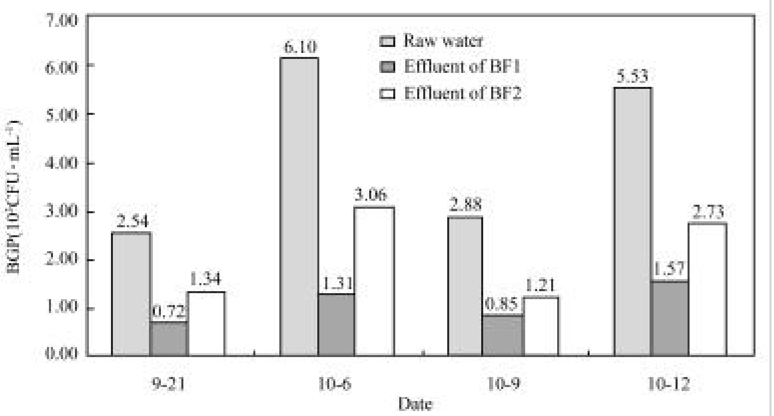

FIG. 7. Effect of phosphate addition on the biological stability of the BF1 effluent.

When no phosphate was added, the average PO_4^{3-}-P concentration of the influent was

20.70 g • L^{-1}, those of the effluents of BF1 and BF2 were 9.33 g • L^{-1} and 11.25 g • L^{-1}, respectively (Table 5), the difference was -1.92 g • L^{-1}. The media in BF1 was irregular-shaped GAC, and that of BF2 was column-shaped GAC, the former was more suitable for the microbial attachment and biofilm formation, the biomass in BF1 was larger than that of BF2 (unpublished data) and more phosphorus for assimilation metabolism was needed in BF1, so more PO_4^{3-} -P was removed by BF1.

TABLE 3

Utilization of PO_4^{3-} -P by BF1 and BF2

		Concentration of PO_4^{3-} -P（ g • L^{-1}）					Removal amounts of PO_4^{3-} -P (g • L^{-1})		
		Influent of BF1[a]	Influent of BF2[a]	Addition amount of KH_2PO_4	Effluent of BF1	Effluent of BF2	BF1	BF2	Difference
1st phase	Average	20.70		0	9.33	11.25	11.36	9.44	1.92
2nd phase	Maximum	148.49	19.92	136.71	79.10	11.78	69.39	11.84	62.74
	Minimum	24.20	8.83	8.81	9.34	1.10	9.91	1.48	1.29
	Average	44.74	13.35	31.38	23.77	6.23	20.97	7.12	13.85
	Average II[b]	27.45	13.69	13.76	12.88	5.90	14.57	7.79	6.78

Note. a: In second phase, KH_2PO_4 was added into the influent of BF1, while nothing was added into the influent of BF2; b: Referred to the average when the phosphate addition amount was less than 20 g • L^{-1}.

After the addition of phosphate, much more of phosphate was removed by BF1 (Table 3). The average addition amount of phosphate to the influent reached 31.38 g • L^{-1}, a rather high level, which was due to the addition amount of 20-140 g • L^{-1} in several days in order to investigate the dose-reaction relationship of the microbial utilization of phosphorus to the removal of organic matters of the biological filter. BF1 could remove 20.97 g •L^{-1} PO_4^{3-} - P under such a condition, but the PO_4^{3-} - P concentration in the effluent was still 23.77 g • L^{-1}, which was higher than 13.35 g • L^{-1}, that of the influent. In fact, the addition amount was less than 20 g •L^{-1} in most days, if the days when the addition amount exceeded 20 g •L^{-1} were excluded, the average phosphate removal of BF1 was 14.57 g • L^{-1}, the concentration in effluent was 12.88 g • L^{-1}, still higher than that of the control filter, but less than 13.69 g • L^{-1}, the concentration of the influent. The results suggested that if only the phosphate addition dose was well controlled, there should be no concern over phosphorus pollution in the effluent of the reactor.

The Dose-reaction Relationships of the Microbial Utilization of Phosphorus and the Increase of COD_{Mn} Removal Efficiency

When the phosphate addition amounts were <20 g • L^{-1}, the differences of the removal amounts of PO_4^{3-} -P of the two reactors were between 4 and 12 g • L^{-1}. A good linear relationship occurred between this difference, i.e. the microbial utilization of phosphorus, and the removal of the organic matters of the reactor (Fig. 8). The correlation coefficients of the differences of the PO_4^{3-} -P removal amounts to the differences of COD_{Mn} removal amounts and removal efficiencies between the two reactors were 0.8287 and 0.8420, respectively. The meanings of the two slopes of the curves in Fig. 8 were that when one more microgram PO_4^{3-} - P was removed by BF1 than by BF2, the COD_{Mn} removal amount and removal efficiency of BF1 were 0.0553 mg • L^{-1} (55.3 g • L^{-1}) and 1.43 percentage points higher than those of BF2, respectively.

But when the phosphate addition amounts were greater than 20 g • L^{-1}, the differences of the removal amounts of PO_4^{3-} -P of the two reactors were >12 g • L^{-1}. The good linear relationship between the phosphorus utilization and the COD_{Mn} removal disappeared, and as a result the microbial utilization of phosphate would no longer promote the removal of organic matters.

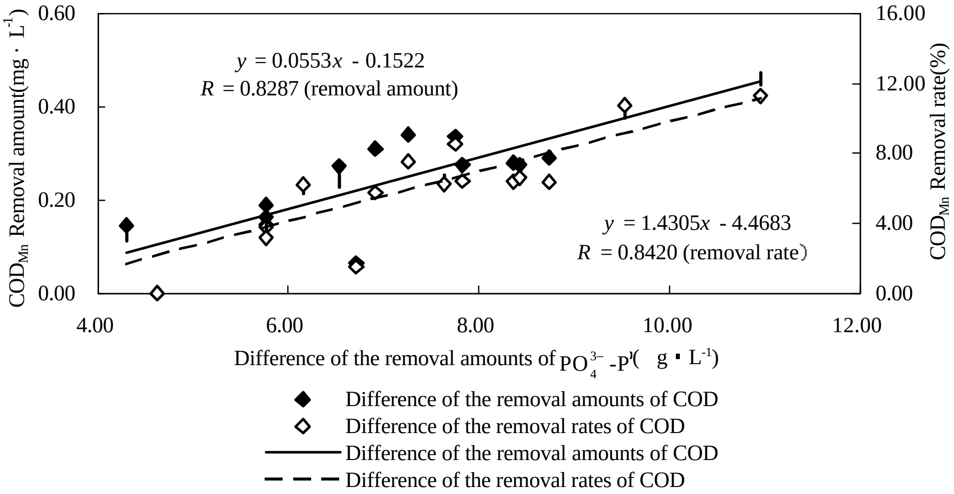

FIG. 8. Relationship of the differences of PO_4^{3-} -P removal amounts (4-12 g • L^{-1}) to the COD_{Mn} removal amounts and removal rates of BF1.

DISCUSSION

In other studies, the phosphorus levels were much lower, which seldom exceeded 5 g • L^{-1}[3, 21, 17, 18], so it seemed more "reasonable" that phosphorus had limiting effect on the microbial growth and metabolism. While in this study, though the raw water had also been treated by coagulation and sedimentation, the average phosphorus level of the influent was of a much higher value, 15.01 g • L^{-1} (Table 1). The reasons that phosphorus still played a limiting role under such a high concentration might be mainly due to the higher level of organic matters. In the studies mentioned above, TOC concentration was between 0.7-3.1 mg • L^{-1} or COD_{Mn} concentration was under 2.0 mg • L^{-1}. The main components of those organic matters were natural organic matters (NOM) because the sources were not polluted; while, the source for the plant where this study was undertaken was polluted by domestic sewage. The average COD_{Mn} concentration was 3.88 mg • L^{-1} and the maximum was 5.40 mg • L^{-1}. The main component of natural organic matters (NOM) were humic substances which had a relatively poorer biodegradability than domestic sewage[1]. So the higher concentration and biodegradability of the samples had a higher level of bioavailable organic matters which led to a higher phosphorus demand since a certain ratio of C:P was required in the microbial growth. Another reason might be that just as the microbes could only utilize the "bioavailable" organic matters, and not all the phosphorus in water could be taken directly by the microbes. In fact, most of the total phosphorus is associated with particulates, only a small part can be used directly by the microbes[22], so the real concentration of the "bioavailable" phosphorus in this study should be less than 15.01 g • L^{-1}.

It is widely accepted that a certain ratio of C:P is needed in microbial growth, though the ratio itself is contentious. If C P=100 1[23, 24] or 100 1.7–2[18] was proper, then it could be drawn that BGP(C) should be greater than BGP(P) since the dose of carbon and phosphorus added into the sample were 8 mg • L^{-1} (20 mg • L^{-1} $C_6H_{12}O_6$) and 50 g • L^{-1}, respectively, and the C P was 100 0.625, which was far more than 100 1–2. But the fact was on the contrary, so the conclusion was that phosphorus was a more insufficient nutrient in the samples and its limitation was stronger than that of carbon.

Up to now, there is serious lack of the references on the direct application of phosphorus limitation to improve the removal of organic matters. These papers were restricted in using a sole carbon source or an artificial cocktail as the organic matters are in a laboratory-scale process[17]. While in this study, the scale was larger, each of the biofilter produced 3.37 m^3 water • day^{-1} and the influent was the actual water in field. Though the difference between the removal rates of the phosphate-added BF1 and the control filter was only 6.02 percentage points, the removals of themselves are rather small, if expressed in another way, the removal rate of BF1 (20.56%) was 41.40% higher than that of BF2 (14.54%), which was a substantial increase.

The organic matters removed by the biological filter were mainly the biodegradable fractions of the total organic matters in the influent. Usually, the biodegradable organic matters presented as BDOC just occupied 15%-35% of TOC in the source water in China[1]. Moreover, as demonstrated above, phosphorus had a strong limiting effect in the biological treatment for the influent, the microbial growth was attributed to the level of the limiting nutrient. Only the concentration of the most important limiting nutrient could be reduced to the minimum. So there must be some of the bioavailable organic matters which could not be utilized by the microbes for phosphorus limitation. For these reasons, both of the COD_{Mn} removal rates of the two reactors at the first stage were rather low, only about 15%, respectively.

After the phosphate was added into the influent, the limiting effect of phosphorus would be weakened and then disappear, and as a result carbon become the predominant limiting nutrient, the part of organic matters that could not be utilized by the microbes because of the phosphorus limitation now might be utilized, so the utilization of carbon could approach its possible maximum and the removal of the organic matters was increased substantially. Furthermore, as Fig. 7 showed, after the treatment of the two reactors, both BGP(n)s of the effluents of BF1 and BF2 were far less than that of the influent, and meanwhile, the BGP(n) of BF1 effluent was obviously lower than that of BF2 effluent, i.e. the level of the organic matters that the microbes could utilize in the effluent of BF1 was less than that in the effluent of BF2, the potential to support the bacterial growth of the effluent of BF1 was reduced, namely, the biological stability of the effluent of BF1 was increased by the addition of phosphate into the influent.

When the addition dose of phosphate did not surpass a limit which was about 20 g • L^{-1} in this study, phosphorus was still a limiting nutrient and the addition could promote the utilization of organic matters of the reactor, but when this limit was broken, the role of phosphorus as the most important nutrient would be replaced by carbon, the microbial growth and metabolism will be dependent on the levels of the biodegradable organic matters, and then the addition of phosphate to the influent could only improve the removal of phosphate itself by the reactor, but will not help to remove organic matters.

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