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Absorption of phosphorus from wastewater by aged refuse excavated from municipal solid waste landfill

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Abstract: Municipal solid waste(refuse) landfill stabilizes as the refuse degrades. After years of biodegradation, the refuse in the landfill becomes stabilized and aged, which may vary with the local climate, humidity, and composition of refuse placed. In this work, it is found that the refuse with an age of over 8 years at Shanghai Refuse Landfill has been significantly stabilized and sufficiently aged and is thus suitable for excavation. The 8-year old aged refuse is mechanically screened, and the fine fractions of refuse(aged refuse) with a diameter less than 2 cm are then used as a biological absorbent for removal of both inorganic and organic phosphorus in livestock wastewater and prepared aqueous solution. It is proved that the aged refuse is very effective for the quantitative removal of both types of phosphorus. The absorption mechanism is proposed. It is considered that phosphorus is firstly absorbed onto the surface of the aged refuse and then used as a substrate for the growth of microorganisms which ultimately leave the aged refuse as sludge.

Keywords: refuse; phosphorus; landfill; livestock wastewater; biological absorption

Introduction

Viewing municipal solid waste landfill as a bioreactor has been widely accepted(Bookter, 1982; Reninhart, 2002; Zhao, 2001; 2002a; 2002b). The biodegradable wastes, such as food origin wastes, backyard wastes, papers, etc., will biodegrade slowly but effectively, under the anaerobic landfill conditions(Zhao, 2001; 2002a). As a result, the wastes in the landfill become stabilized and aged. The mineralization extent of the aged wastes may be a function of composition, fraction of biodegradable components, moistures of the wastes, and the climatic conditions of the landfill located (Zhao, 2001). Depending on the local climatic conditions and components of the wastes, the wastes in landfill may greatly stabilized or mineralized, after placement of 8—15 years. For instance, it had been found that the wastes in Shanghai MSW Landfill was substantially stabilized after 8 years of placement, and the wastes thus excavated generated no leachate on drying and released slight odor (Zhao, 2002c). Hence, the wastes excavated from a 8-year old landfill compartment has been well characterized and can be used for a variety of purposes, as one of example reported in this paper.

There are over 100 sanitary MSW landfills in China that are still operational. In addition, over 2000 dumping sites that had ever serviced as final disposal sites for MSW in hundreds of cities in China. It is roughly estimated that around 100 millions tons of wastes that are buried in these landfills and dumping sites. This quantity should be much larger when considering the wastes in the world. Many dumping sites are actually surrounded by the commercial and residential development areas and should be removed or rehabilitated, as the cities expand. Meanwhile, it is more and more difficult to sit new landfills for most cities in China, as the public awareness for the landfill hazard raise. Many attacks from the local habitants to the officers and managers responsible for the landfills occur, even during the period of sits selection. In the southern territory of China, where nearly every inch land has been cultivated carefully for agricultural and industrial uses, sitting for a landfill is so difficult that many cities have to cancel the original planning of landfill and to construct incineration plant.

It is considered that the recycling of existing closed landfills should be extremely significant and practical so that the life span of the landfills can be extended. In our previous work, the excavation of aged wastes from landfill and then refilling of fresh wastes into the excavated space left was practiced. It was found that around 80% of the aged wastes (8-years old) could be recycled, recovering the plastics, glass, fine fraction of organic materials, after screening and separation. Another 20% aged wastes were found to be larger size of stone and non-degradable rubber and polymer products such as automobile covers, bulky electronic devices, which should be refilled into the landfill compartments. Hence, the life span of landfill can be substantially extended, when these 80% of recyclable aged waste can be recycled.

In the 80% of recyclable waste, some 20% are plastic and glass products, which may be readily recycled, with a relatively good market, though the quality of these recyclables have deteriorated. The remaining aged wastes are humus-riched and compost-like fine organic wastes, which should receive more attention to find their recycling uses.

One of research work done at our laboratory is to use the aged waste as biofilter materials to treat leachate (Zhao, 2002a), sewage (Guo, 2002), and livestock wastewater (Shao, 2002). In these innovative research work, it has

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found that the biofilter made of the aged organic waste can be used for the effective treatment of a variety of wastewater, including MSW landfill leachate, sewage, livestock wastewater, and so on. It is also found that the aged waste can be used for the effective bioabsorption of both inorganic and organic phosphorus, as reported in this paper.

Phosphorus, present as both inorganic and organic forms in many types of wastewaters, must be removed deeply from the wastewaters(Bollag, 1983). The regulatory standard for total phosphorus in the waters discharged into water receivers is 0.1—0.5 mg/L. Traditional wastewater treatment processes, such as activated sludge method, anaerobic and aerobic biological methods, are generally difficult to meet the requirements for the phosphorus removal (Choi, 2002; Heijnen, 1992; Hogrefe, 1986; Keek, 1993). Meanwhile, the phosphorus removed from the wastewater is actually transferred into the sludge while the wastewater is biologically treated. The sludge thus generated contains over 98% of water and look bulky. The presence of phosphorus in the sludge makes its further treatment more difficult(Kalynzhnyi, 2002; Luthy, 1997; Mihelcic, 1993).

Ideal method for phosphorus removal from wastewater seems to collect the phosphorus in a condensed phase which can hold a high content of phosphorus, using membrane and airlift reactors(Tijhuis, 1994; Wong, 1989). Methods such as chemical precipitation, physical absorption, biological conversion, etc., should be included.

Chemical precipitation of phosphorus may result to generation of a bulky sludge. Moreover, traditional Ca and Mg series precipitates can not remove the phosphorus to the regulatory set values of less than 0.5 mg/L, useless the precipitates of calcium or magnesium phosphates can precipitate as crystals by addition of crystal seeds and aging for days.

Physical absorption for phosphorus removal from wastewater has been studied widely. Unfortunately, most absorbents are not regenerable, which limit the uses of these absorbents. Biological removal of phosphorus should be a prior option for the treatment of wastewater containing phosphorus. In our preliminary research, it was found that the fine aged fractions of wastes excavated from a 8-year old landfill compartment had unexpected biological absorption capacity for phosphorus present in the aqueous solutions or real livestock wastewater containing phosphorus. The research results are reported in this paper, attempting to find out another use for the wastes.

1 Experimental

1.1 Chemicals and solutions and livestock wastewater used

All chemicals used are of analytical grade. Inorganic phosphorus is analyzed colorimetrically using molybdenum blue method. The livestock wastewater, including influent and effluent, is oxidized firstly by nitric acid before analysis.

The aqueous solution containing phosphorus is prepared by dissolving a given quantity of Na_3PO_4 in distilled water.

The real livestock wastewater is taken from a livestock farm in Shanghai, and its quality is shown in Table 1. It should be pointed that the livestock wastewater given in Table 1 is sampled from the effluent discharging point after the raw livestock wastewater has been treated anaerobic and aerobic processes. It has been proved that both phosphorus and ammonium-N can not be removed to the required values using the biological processes. Using the solvent extraction method (Zhao, 1999), it was found that the proportion of organic phosphorus in the wastewater was around 20% of the total phosphorus.

Table 1 The quality of livestock wastewater used

Items	Values	Items	Values
pH	7.2—8.5	SS, mg/L	83—407
COD_{Cr}, mg/L	197.6—2419.84	BOD_5, mg/L	102—459
NH_3-N, mg/L	260.68—1140.2	Total N, mg/L	512.7—1669.08
NO_3-N, mg/L	2.09—44.2	NO_2-N, mg/L	0.017—8.97
Total P, mg/L	24.2—102.03	DO, mg/L	1.22—2.4
Color	40—200		

1.2 Preparation of the fine aged wastes

The preliminary treatment for the aged wastes has been described in detail in another paper(Zhao, 2002a; 2002c). The wastes with 8-years old excavated from Shanghai MSW Landfill were directly screened by a rotary drum screen with definite and uniform punched holes of 15 mm in diameter. Around 50%—60% of the excavated wastes can pass through the holes, which is referred to as the fine aged fractions, or simplified as the aged refuse, in this paper. The aged refuse less than 15 mm are then ground into powder with less than 3 mm in diameter, which is used as the absorbent for the removal of phosphorus from aqueous solution and livestock wastewater.

1.3 Absorption of phosphorus

A given quantity of the fine aged refuse thus obtained is added into a flask containing a given volume of aqueous solution or livestock wastewater. Mix the absorption system in a mechanical vibrator with a frequency of 110 r/min at room temperature, unless otherwise indicated. All the data provided in this paper are at least in duplicate, and most are in triplicate.

Long-term absorption of phosphorus: A biofilter is made using 75 kg of the fine aged refuse, with an inner diameter of 30 cm and height of 100 cm. The livestock wastewater passed through the biofilter for 6 months at a hydraulic load of 25 L/d, by spraying over the surface of the fine aged refuse, with 4 h of wastewater introduction and 20 h of drying. The operation is repeated in the next day and so on, imitating the treatment process for sewage. The effluent collected in the bottom of the biofilter is then analyzed. The operational conditions have been optimized in our previous research work

(Shao, 2002). The biofilter can work well for years, without blockage and deterioration of the treatment of efficiencies for the pollutant removal.

2 Results

2.1 Equilibrium time for phosphorus absorption from prepared aqueous solutions

Table 2 shows the equilibrium time required for the absorption of phosphorus from the prepared aqueous solution containing 50 mgP/L, using the aged refuse. It can be seen that the equilibrium can be reached after around 15 h of solid-liquid contact. Meanwhile, it can be also seen that the absorption is very effective, with a removal of 99% of P from the aqueous solution.

Table 2 Effect of time on the absorption of phosphorus using the aged refuse

Initial P in the aqueous solutions: 50 mg/L; refuse/solution ratio: 2 g/50 ml

Time, h	0	3	6	9	12	15	18	21	24
P left in the aqueous solutions	50.00	38.45	35.03	21.89	8.98	0.60	0.58	0.50	0.51
P absorbed, %	0.0	20.0	29.9	56.2	82.0	98.8	98.8	99.0	99.0

2.2 Effect of aged refuse/P ratios on the absorption

Table 3 shows the relationship between the aged refuse weight and the phosphorus removal from prepared aqueous solutions containing varied phosphorus concentrations, at a equilibrium contacting time of solid/liquid system of 18 h. It can be obviously seen that the maximum removal of phosphorus from the aqueous solutions is around 98.4%—98.7%, and the removal increases as the quantity of aged refuse used increases up to the maximum removal of phosphorus. The maximum absorption capacity of the aged refuse for phosphorus is found to be around 2.46—2.62 mg of P in 1 g of the aged refuse used, regardless of the initial phosphorus concentrations in the aqueous solutions. Increase of the aged refuse quantity will lead to the decrease of P absorbed in a given quantity of the aged refuse.

As the highest removal percentage of phosphorus from the aqueous solutions is around 98%—99%, the phosphorus left in the effluent after absorption is increased as the initial concentrations of phosphorus in aqueous solutions increase, from 0.65 mg/L at initial 50 mg/L P to 1.8 mg/L P at initial 120 mg/L P, respectively.

2.3 Effect of pH on the absorption of P in the aged refuse

To 300 ml solution containing 50 mg/L P, add 3 g of the aged refuse, adjust pH using NaOH or HCl solution to a given value, stir with a magnetic stirrer for 15 h and then analyze. The results are given in Table 4. It can be seen that the phosphorus removals keep at the maximum values of around 98%—99% at a pH range of 5—9. Beyond this pH value range, the removal will decrease. In general, the pH of real organic wastewater falls into this range. Obviously, the biological absorption can work well at a wide range of pH, indicating the adaptability of the aged refuse to the removal of phosphorus.

Table 3 Relationship between aged-refuse quantity and phosphorus removed Aqueous solutions: 50 ml

Quantity of the aged refuse, g	0.5	1	1.5	2	3	4	5
P concentration in aqueous solution	50 mg/L						
P removal, %	56.60	98.40	98.70	98.70	98.70	98.70	98.70
P absorbed/aged refuse, mg/g	2.83	2.46	1.64	1.23	0.82	0.61	0.49
P concentration in aqueous solution	80 mg/L						
P removal, %	30.40	60.90	98.40	98.40	98.40	98.40	98.40
P absorbed/aged refuse, mg/g	2.43	2.43	2.62	1.97	1.31	0.99	0.79
P concentration in aqueous solution	100 mg/L						
P removal, %	27.40	55.70	71.20	98.40	98.40	98.40	98.40
P absorbed/aged refuse, mg/g	2.74	2.78	2.38	2.46	1.64	1.23	0.98
P concentration in aqueous solution	120 mg/L						
P removal, %	25.70	43.20	65.40	95.20	98.50	98.50	98.50
P absorbed/aged refuse, mg/g	3.08	2.59	2.62	2.86	1.97	1.48	1.18

Table 4 Relationship between pH and phosphorus removal by aged refuse

pH	1	2	3	4	5	6	
P removal, %	0	23.40	94.30	95.70	98.70	98.20	
pH	7	8	9	10	11	12	13
P removal, %	98.40	98.70	98.70	92.60	90.70	68.70	46.90

2.4 Long-term absorption using the refuse-based-biofilter

Fig.1 shows the removal efficiency of total P from the livestock wastewater using the biofilter. Obviously the total P can be removed very effectively, with an average removal of 99%, nearly regardless of the influent total P. When the influent total P ranges from 20—100 mg/L, the corresponding effluent total P will be reduced to 0.1—2.3 mg/L, depending on the influent total P concentrations used. When the influent total P are lower than 40 mg/L, the effluent total P will be lower than 0.5 mg/L, the allowed discharge standard for the wastewater in China.

The biofilter is being continued for treatment of the

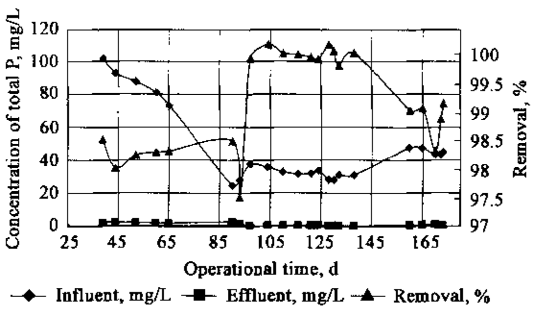

Fig.1 Removal of total P from the wastewater and its dependence on the initial concentrations in the influents

livestock wastewater and reflect the same removal capability to phosphorus and COD as shown below.

2.5 Elution of the phosphorus in the aged refuse

In order to justify the removal mechanism of phosphorus in the aged refuse, the aged refuse in the biofilter is eluted with tap water after the biofilter is stopped for use. The results are shown in Table 5. The total phosphorus in the washing water is 0.14 mg/L. From Table 5 it can be seen that the phosphorus in the elution solution is the same as that in the washing solution until day 22. On the day 23, the phosphorus in the washing water is removed considerably by the biofilter so that the phosphorus concentration in the effluent is lower than in the washing water.

Hence, the phosphorus that removed from the influents should be transferred as insoluble forms or as the substrate of the bacteria that become sludge after metabolism. However, it seems impossible for the phosphorus to form insoluble chemicals such as calcium phosphate or magnesium phosphate as the aged refuse has stayed in the landfill for at least 8 years and the soluble forms of calcium and magnesium should be eluted away by the leachate and precipitation. The only way for the phosphorus removal is that the phosphorus in the influent is biologically absorbed when it passes through the biofilter and then leaves the system in the form of sludge which can be collected readily at the bottom of the container used for the collection of effluent.

Table 5 Elution of P from the biofilter using water

The total P in the washing water: 0.14 mg/L

Days for elution	The first day	Day 3	Day 7	Day 10	Day 23
P in the elution solution, mg/L	0.14	0.14	0.14	0.14	0.04

2.6 Bacteria population determination in the biofilter

The population of bacteria in the biofilter is counted with conventional method and the results are shown in Table 6. Compared the bacteria population in the fresh aged refuse and the aged refuse sampled from the biofilter having used for the removal of phosphorus in the livestock wastewater, the population of *Heterobacter*, *Nitrobacter*, *Nitrosomonas*, and *Denitrobacter* increase by 1—4 order of magnitude, while that of *Coliform* seems unchanged. Hence, the contact of wastewater with the aged refuse can favor the growth of bacteria, which also partly explain why the aged refuse biofilter acts as a biological reactor and the phosphorus is removed biologically.

Table 6 Bacteria population determination in the fresh aged refuse and the aged refuse in the biofilter after contacting with livestock wastewater (number/g of aged refuse in dry basis)

Types of bacteria	*Heterobacter*	*Coliform*	*Nitrobacter*	*Nitrosomonas*	*Denitrobacter*
Fresh aged refuse	4.5×10^6	1.28×10^5	3.38×10^3	6.08×10^4	4.73×10
Used aged refuse in the biofilter	9.6×10^7	1.15×10^5	2.44×10^4	6.41×10^7	1.92×10^4

2.7 Isotherm for the phosphorus absorption

To 1 g of aged refuse, add 50 ml of livestock wastewater containing phosphorus of 50 mg/L, 60 mg/L, 70 mg/L, 80 mg/L, 90 mg/L, 100 mg/L, 110 mg/L and 120 mg/L respectively with pH around 7, shake at a mechanical shaker at 15℃, analyze the supernatant after 15 h contact. Calculate the equilibrium P(C_e, mg/L) concentrations in the supernatant and the equilibrium absorption quantity of phosphorus(q_e, mgP/g aged refuse). The results are shown in Fig.2. It can be seen that the curve of C_e against q_e is a straight line. By simple simulation using the data in Fig.2, the following equation can be thus obtained:

$C_e/q_e = 0.3055C_e + 1.0952$, with correlation coefficient of $R = 0.9912$,

where C_e is the phosphorus in the aqueous solution after absorption equilibrium (mgP/L), q_e is the phosphorus absorbed onto the aged refuse after equilibrium(mgP/g aged refuse).

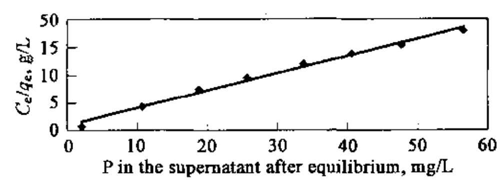

Fig.2 Isotherm of phosphorus bioabsorption by the aged refuse

2.8 Equilibrium time for the removal of COD from the livestock wastewater using the fresh aged refuse

To 10 g of the aged refuse, add 0.3 L livestock wastewater with COD concentration of 647 mg/L, shake it at a mechanical shaker at 15℃, sample once in 3 h and determine COD. The results are shown in Table 7. Obviously, at least 15 h will be required for the contacting equilibrium to reach. That may mean that the retention time of the wastewater in the biofilter should be longer than 15 h so that the maximum COD can be removed. Around 90% of COD can be removed, which has also been justified by a long-term test using the biofilter in another work (Shao, 2002).

Table 7 Relationship between the COD concentration in the livestock wastewater and contacting time with the aged refuse

Initial COD concentration: 647 mg/L

Contacting time, h	3	6	9	12	15	18	21	24
COD removed by the aged refuse, mg COD/g aged refuse	8.2	11.5	13.6	15.7	17.5	17.5	17.6	17.5
COD in the sampled effluent, mg/L	373.3	263.4	193.5	123.6	63.5	62.8	60.1	62.2

3 Discussion

It has well proved that phosphorus in refuse landfill leachate is always lower than 1—4 mg/L, much lower than in that in municipal wastewater. The release of pollutants, including phosphorus, from the refuse placed in landfill may be quite complex. As soon as refuse is placed in a landfill, phosphorus should be released partly. The route for the phosphorus released from the refuse may be present in the leachate, in the sludge settled down to the bottom of container(such as holding cell), or may return to the refuse mass again because of the biological and chemical absorption.

It is difficult to balance phosphorus in refuse among leachate, sludge and decomposed residue in the landfill. In this research, it has ever tried to collect and determine the phosphorus present in the effluent, in the used aged refuse in the long-term test column, and in the sludge settled down to the bottom in the container used to collect the effluent discharged from the biofilter. It is indeed true that quite a part of phosphorus is present in the bottom sludge in the container. However, it is nearly impossible to get quantitative relationships for the phosphorus distributions in the medium concerned. The phosphorus may be absorbed in the aged refuse and then released after days later as sludge. Perhaps isotope test may be a good way to trace the phosphorus routing among the effluent, sludge and the aged refuse in the column.

Huge quantity of aged refuse is available in the world. The resource is inexhaustible. According to the research results obtained in this work, 1 g of the aged refuse can remove around 2.62 mg of P from wastewater on a daily basis, and also, the bioabsorption will continue for a long term period and the absorption of aged refuse will never reach saturation. In this case, a biofilter with 1000 kg of aged refuse can remove 2.62 kg of phosphorus on a daily basis, which is also dependent on the organic pollutants such as COD and BOD presence in the wastewater. In general, the phosphorus concentrations in livestock wastewater may be 100 mg/L. In order to treat 1000 L such livestock wastewater, at least 30—40 kg of aged refuse is required, with around 98% removal. In the presence of COD and BOD, the quantity of aged refuse required is much larger, as proved in the long-term test(Shao, 2002). Practically, 1 ton of the aged refuse can only treat 300 L livestock wastewater with a quality shown in Table 1, with COD, BOD and phosphorus removals of around 90%, 85% and 98% respectively.

4 Conclusions

The aged refuse excavated from a 8-year refuse landfill can be used for an effective removal of phosphorus from prepared aqueous solution and real livestock wastewater. 1 g of the aged refuse can biologically absorbed 2.62 mg of phosphorus from aqueous solution. The absorption capacity may be reduced if COD and BOD is presence in the wastewater. It is proved that the phosphorus is removed by the aged refuse in a biological mechanism. Considering the availability of huge quantity of aged refuse in the world, this work has shown perspectives of the uses of aged refuse.

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