Journal of Hazardous Materials 160 (2008) 554–558

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Studies on land application of sewage sludge and its limiting factors

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ARTICLE INFO

Article history:
Received 26 September 2007
Received in revised form 8 March 2008
Accepted 10 March 2008
Available online 20 March 2008

Keywords:
Sewage sludge
Land application
Grass
Soil
Heavy metal

ABSTRACT

Field experiments were conducted to study the effect of sewage sludge application on the heavy metal content in soils and grasses. The sewage sludge was obtained from Northern Shenyang Wastewater Treatment Plant, China, and applied at 0, 15, 30, 60, 120 and 150 t ha^{-1}. Native grasses *Zoysia japonica* and *Poa annua* were chosen as experimental plants. The experimental results showed that nutrient content of the soil, especially organic matter, was increased after sewage sludge application. The grass biomass was increased and the grass growing season was longer. Heavy metal concentrations in the soil also increased; however, the Zn content did not exceed the stringent Chinese environmental quality standard for soil. Pb and Cu did not exceed the standard for B grade soil, but Cd concentration in soil amended by sewage sludge has exceeded the B grade standard. Therefore, it is suggested that the sewage sludge produced from the wastewater treatment plant should not be applied to farmland, for which B grade soil or better is required. The sludge is suitable for application to forestry and grasslands or nurseries where food chain contamination with cadmium is not a concern.

1. Introduction

Increasing urbanization and industrialization have resulted in a dramatic increase in the volume of wastewater produced worldwide. The increasing numbers of plants for the treatment of wastewater have brought about an increase in the generation of sewage sludge. Large volumes of sludge need to be disposed of or treated in some manner. One way of sludge disposal is its application on land, and this has become common practice [1]. The application of sewage sludge on agricultural soils has been widespread in many countries around the world and this has been shown to improve soil properties and increase plant productivity. In the European community, over 30% of sewage sludge is used as fertilizer in agriculture. For example in UK, Gove et al. [2,3] reported increasing use of enhanced treated sewage sludge in agriculture.

Agricultural land application appears to be a logical and reasonable use of sewage sludge, since it may improve many soil properties, such as pH and contents of organic matter and nutrients [4–6]. In Belgium, 57% of the sludge is applied to land. In France, 60% of the sewage sludge is used for land application [7].

Sewage sludge is effective as a fertilizer, increases dry matter yield of many crops [8], and can also improve soil physical properties such as porosity, aggregate stability, bulk density, and water retention and movement [9]. Although raw sewage sludge contains valuable nutrients such as nitrogen, phosphorus, organic matter, and essential trace elements, it also contains various toxins, especially heavy metals, which cause harm to soil–plant system and further might pose a serious risk to human health [10,11]. The advantages and disadvantages caused by land application of sewage sludge have attracted the attention of environmental authorities, the public and scientists [12].

China is now in a stage of accelerating development resulting in rapid industrialization and urbanization. According to the statistics data of Ministry of Construction of the People's Republic of China, there are 427 wastewater treatment plants in 2000 increased rapidly to 708 ones in 2005. Therefore, the sewage sludge increased correspondingly, now up to 1.0×10^8 t of sewage sludge is produced annually. There has been an increasing tendency in recent years to use sewage sludge as fertilizers in agriculture. The agricultural areas of land application of sewage sludge, usually in the suburbs of cities, involves nearly all the main food crops and vegetables in China such as rice, wheat, corn, millet, soybean, rye, taro, Chinese cabbage, cucumber, tomato, lettuce, hot pepper, cauliflower, and cabbage [13,14].

Recently, environmental awareness has been intensified, the utilization and treatment of sewage sludge is one of major environmental concerns throughout in China. In order to reduce the chances of contaminants such as heavy metals entering the human food chain. Therefore, more extensive research is needed to determine which crops and plants can be planted in the sludge-amended soils and how much loading of sewage sludge can be accepted in practice. So far there are some studies dealing with responses of

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doi:10.1016/j.jhazmat.2008.03.046

Table 1
Contents of nutrients and heavy metals in sewage sludge from Northern Shenyang Wastewater Treatment Plant

Analyze	Value	Standard[a]
pH	6.73	
Organic matter	35.6%	
Total N	2.26%	
Total P	1.15%	
Total K	820 mg kg^{-1}	
Cd	5.0	5.0
Pb	255	300
Cu	170	250
Zn	290	500

Unit for Cd, Pb, Cu and Zn as mg kg^{-1}.
[a] Maximum limit specified by National Standard GB4284-84 for harmful substances in agricultural fertilizers.

the soil–grass system to the application of sewage sludge [7,15,16]. The objectives of this study were to identify the limiting factors of sewage sludge land application, to find out and design optimum loading rates of sewage sludge, and to screen out suitable grass species that can efficiently absorb and accumulate heavy metals in land amended with sewage sludge. The research findings would indicate a sustainable and safe pathway to treat and reuse the sewage sludge produced from the Northern Shenyang Wastewater Treatment Plant, China.

2. Materials and methods

2.1. Sewage sludge component

Northern Shenyang Wastewater Treatment Plant (referred to hereafter as “the Plant”) services more than 1 million people in northern and western suburbs of Shenyang City and receives a mixed sewage from domestic, commercial and industrial sources. The Plant provides an essential public health service to treat about 24% of Shenyang’s sewage, or about 400×10^3 t/day, and produces 120 t/day of sludge containing 70% water or 13×10^3 t/a. The nutrition components and heavy metal content of sewage sludge collected from the Plant are shown in Table 1.

2.2. Field experiment design

Each plot area of 10 m^2 (2 m × 5 m) for field experimental was designed. There were two kind of native grasses and seven levels of loading rates of sewage sludge applications in the experiments with three replicates. There were 42 plots totally that occupied about 612 m^2 including the buffer area between experimental plots. Field experiments were started in May 2000. The field plots were located on vacant land in the north part of the Plant. The research sites for the experiments are meadow brown soils, which are typical soils in Shenyang areas. The basic physical and chemical properties of the soil in the experimental field were measured as shown in Table 2. The entire experimental field was ploughed and fine tilled to remove all existing grasses before sewage sludge was spread onto the plots at sludge loading rates of 0, 15, 30, 60, 90, 120 and 150 tonne dry matter per hectare (t ha^{-1}); the sludge was tilled into the top 150 mm of soil in the recommended way for local farmers. Each treatment had three replicates. Three weeks after sewage application, plots were planted out with grass as described below in Section 2.3. After 4–16 months, grass samples were harvested for analysis while soil samples were analyzed after 16 months.

2.3. Experimental plants and sampling design

Native grasses *Zoysia japonica* and *Poa annua* were chosen for the field experiment; the grass seeds were planted on 23 May 2000 in field plots with sewage sludge applied on 2 May 2000. The grass (*Z. japonica*) was harvested 120 days after sowing; grass samples were collected from five squares 0.3 m × 0.3 m within 2 m × 5 m plot and then mixing up evenly. After harvesting, grass samples were washed with tap water and rinsed twice with deionised water to remove any attached particles. Each grass tissue sample was put into a brown paper envelope and then into ovens at 80 °C for drying to constant weight. The grass samples (*Z. japonica* and *P. annua*) were collected and pre-treated in the same way in October of the subsequent year.

At the end of the trial, the surface (0–15 cm) soil samples were collected from each plot by taking five points using a soil borer (size 5 cm diameter) in each plot (2 m × 5 m) and then mixed up evenly. These soil samples were dried at 105 °C and ground to powder (through 100 mesh/in.) then stored in brown paper envelope for analysis.

2.4. Chemical and statistical analyses

All analyses were carried out in accordance with standard methods of China (GB/T17140-1997). The organic matter content of sewage sludge was analyzed by the potassium dichromate method. Total phosphorus (TP) was determined by molybdenum antimony colorimetry after digestion by HNO_3–$HClO_4$–HF. Total potassium (TK) was determined by flame emission spectrometry after HNO_3–$HClO_4$–HF digestion, and nitrogen was determined by micro-kjeldahl method. The concentrations of Cd, Pb, Cu and Zn were determined by flame atomic absorption spectrometry using a Hitachi model 180-80 AAS spectrometer. Analytical quality control was verified using an environment standard substance for heavy metals from Environmental Monitoring Station of China. Statistical analysis was performed using SPSS 10.1. Means and standard errors are presented for all data.

3. Results and discussion

3.1. Effect of sewage sludge application on the grass biomass

The measure of grass biomasses in Table 3 showed that the biomasses of grass were increased obviously due to sewage sludge application (Table 3). The biomass of *Z. Japonica* was increased by 64–316% in the sludge treatments compared with the control (significant difference, $P < 0.01$). The nutrient amounts were significantly elevated in soil after sewage sludge application. Sewage sludge as fertilizer supplied abundant nutrients. Sludge application

Table 2
Basic physical and chemical properties of soil in experimental field

OM (%)	TN (%)	TP (%)	pH	Heavy metals mg kg^{-1}			
				Cd	Pb	Cu	Zn
2.410 ± 0.096	0.196 ± 0.016	0.154 ± 0.019	6.450 ± 0.075	0.277 ± 0.024	28.256 ± 2.391	27.682 ± 3.398	27.110 ± 2.438

Data are reported as mean ± S.D.

Table 3
Effects of sewage sludge application on biomass of grasses (kg/plot)

Sewage sludge loading rate t ha^{-1}	*Zoysia japonica* biomass		*Poa annua* biomass
	2000	2001	2001
Control	2.58^a ± 0.12	2.26^a ± 0.17	1.27^a ± 0.14
15	4.25^c ± 0.39	2.78^b ± 0.54	1.84^c ± 0.13
30	5.59^c ± 0.59	2.13 ± 0.19	1.33 ± 0.03
60	5.92^c ± 0.30	2.73^b ± 0.24	1.69^c ± 0.10
90	5.89^c ± 0.30	2.07 ± 0.09	1.59^b ± 0.02
120	9.12^c ± 0.71	2.51^b ± 0.20	1.10 ± 0.15
150	10.74^c ± 1.85	1.99 ± 0.16	1.39 ± 0.06

Data are reported as mean ± S.D. Values with different superscripts have significant differences as follows: a *vs.* b are significantly different at P = 0.05; a *vs.* c are significant different at P = 0.01.

alters the chemical and physical properties of the soil, which may affect plant nutrient balance. In the next year (16 months after the applications of sewage sludge), the biomass of *Z. japonica* was also increased as compared to control (significant difference, P < 0.05). It is thus clear that the residual effects of sewage sludge application were reduced.

In 2001, the biomasses of both grasses are the highest at the sewage sludge treatment rates of 15 and 60 t ha^{-1}. *Z. japonica* biomass is increased by 23.07% and 21.04% respectively compared with the control (significant difference P < 0.05), and *P. annua* biomass was increased by 44.65% and 32.91% compared with control (significant difference P < 0.01). The biomass of *Z. japonica* is higher than *P. annua*, and since *Z. japonica* is resistant to drought and was observed to have flourishing roots, it may be a suitable grass species for the greenbelt of the city.

3.2. Soil nutrient content change by sewage sludge application

Soil organic matter plays an essential role in the cycle of nutrient (N, P, K), and affects the sustainability of soil fertility. Therefore, the organic matter content can be an important indicator of soil fertility. After one growth season following the sewage sludge application, soil samples were collected and the nutrient content was determined. The results are shown in Table 4.

The results in Table 4 show that the organic matter increases from 12.79 to 80.8% compared to the control as the sewage sludge loading rates are increased from 15 to 150 t ha^{-1}. The organic matter amounts increased significantly in upper 15 cm layer of the soil. Total nitrogen (TN) content was only significantly different compared to the control (P < 0.05) at sewage sludge loading rate of 150 t ha^{-1}. TP contents in soil with sewage sludge loading rates of 15, 30 and 90 t ha^{-1} were increased 30.2%, 190.5% and 31.8% respectively compared with the control respectively.

Table 4
Effect of sewage sludge application on nutrients in soil

Sewage sludge loading rate (t ha^{-1})	OM (%)	TN (%)	TP (%)
Control	3.44^a ± 0.19	0.20 ± 0.01	0.13 ± 0.02
15	3.99^b ± 0.35	0.20 ± 0.02	0.16^b ± 0.01
30	3.88^b ± 0.16	0.22 ± 0.01	0.37^c ± 0.01
60	6.08^c ± 0.27	0.24 ± 0.02	0.14 ± 0.01
90	6.01^c ± 0.45	0.23 ± 0.01	0.17^b ± 0.02
120	5.89^c ± 0.15	0.23 ± 0.02	0.09 ± 0.02
150	6.22^c ± 0.22	0.26^b ± 0.01	0.13 ± 0.02

Data are reported as mean ± S.D. Values with different superscripts have significant differences as follows: a *vs.* b are significantly different at P = 0.05; a *vs.* c is significant different at P = 0.01.

Table 5
Effects of sewage sludge application on the concentration of heavy metals in the soil mg kg^{-1}

Treatment t ha^{-1}	Cd	Pb	Cu	Zn
Soil with *Zoysia japonica* planted				
Control	0.39^a ± 0.04	28.36^a ± 2.14	27.01^a ± 2.58	26.30^a ± 2.39
15	0.42^b ± 0.01	44.05^c ± 2.30	35.03^c ± 3.03	36.80^c ± 4.03
30	0.42^b ± 0.03	49.11^c ± 2.04	41.26^c ± 2.20	38.58^c ± 2.02
60	0.41^b ± 0.02	42.40^c ± 2.88	37.73^c ± 2.19	43.36^c ± 4.19
90	0.56^b ± 0.02	37.80^c ± 1.86	37.56^c ± 2.21	39.49^c ± 2.82
120	0.43^b ± 0.03	37.68^c ± 2.08	36.69^c ± 3.48	33.64^c ± 2.51
150	0.43^b ± 0.01	36.53^b ± 1.81	30.59^b ± 4.68	31.08^b ± 0.89
Soil with *Poa annua* planted				
Control	0.23^a ± 0.01	28.16^a ± 4.95	28.35^a ± 2.59	27.92^a ± 2.30
15	0.36^b ± 0.01	43.47^c ± 2.55	31.90^b ± 2.94	35.74^b ± 2.90
30	0.52^b ± 0.06	39.42^c ± 5.83	32.11^b ± 1.89	41.11^c ± 2.94
60	0.48^b ± 0.03	37.26^c ± 2.33	30.64^b ± 2.70	37.41^b ± 1.56
90	0.41^b ± 0.02	38.59^c ± 3.21	29.52 ± 1.74	41.79^c ± 3.80
120	0.40^b ± 0.02	39.93^c ± 2.93	30.86^b ± 2.11	34.46^b ± 3.51
150	0.43^b ± 0.01	41.30^c ± 1.99	36.33^c ± 2.15	35.18^b ± 1.45

Data are reported as mean ± S.D. Values with different superscripts have significant differences as follows: a *vs.* b is significantly different at P = 0.05; a *vs.* c is significant different at P = 0.01.

3.3. Heavy metal concentration in the soil amended by sewage sludge application

Sludge contains relatively high levels of organic matter and plant nutrients, which have the beneficial advantage as soil conditioner and fertilizer. However, there are potential environmental impacts of sludge application in terms of soil and groundwater contamination. Usually the heavy metals in soils and plants are the priority factors to be considered for the land application of sewage sludge. The heavy metals contents in the sludge-amended soil of this study are shown in Table 5.

When sewage sludge was applied to soil, Cd, Pb, Cu and Zn concentrations in the soil ranged from 0.3 to 0.6, 36 to 50, 30 to 42 and 31 to 43 mg kg^{-1} respectively. For comparison the Cd, Pb, Cu and Zn concentrations in control soils are only 0.23–0.39, 28.16–28.36, 27.01–28.35 and 26.30–27.92 mg kg^{-1} respectively. The heavy metals concentrations in soil are obviously increased compared to the untreated control. This demonstrated that sewage sludge application may have the risk of soil contamination, if application technology is not appropriate and site condition is not suitable.

The Cd concentration exceeds the environmental quality standard for grade B soils (Grade B applies to farmland, vegetable land, tea land, fruit land and grazing land), but it does not exceed the standard for Grade C soils (Grade C soils are applicable for forestry land and land with higher absorption capacity). Pb and Cu concentrations in soil are lower than the grade B standard for soils and Zn concentration even meets the grade A environmental qual-

Table 6
Environmental quality standard for soil in China (GB15618-1995; metal concentrations mg kg^{-1})

Metal	Soil grade				
	A	B			C
		pH < 6.5	pH 6.5–7.5	pH > 7.5	
Cd	0.20	0.30	0.30	0.60	1.0
Cu farm	35	50	100	100	400
Cu garden	–	150	200	200	400
Pb	35	250	300	350	500
Zn	100	200	250	300	500

Grade A: natural conservation area, drinking water catchments, tea garden.
Grade B: farmland, vegetable land, tea land, fruit land and grazing land.
Grade C: forestry land and the land with higher absorption capacity.

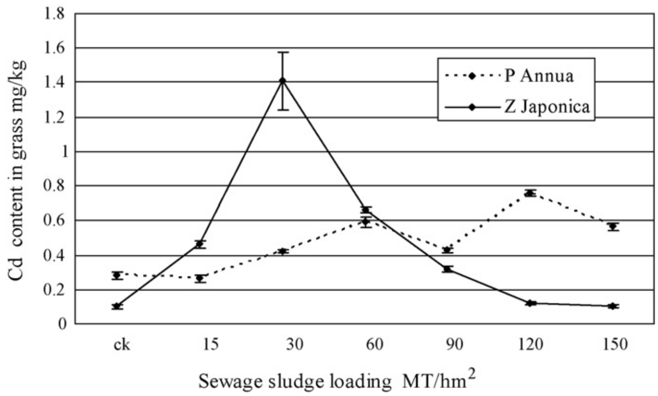

Fig. 1. Cd content of grasses grown in soils with different sewage sludge application rates.

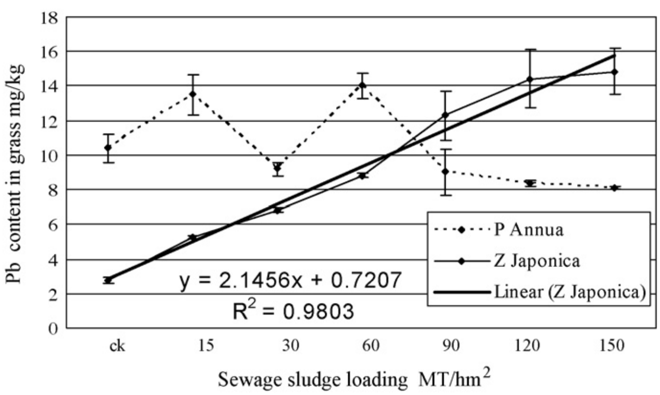

Fig. 2. Pb content of grasses grown in soils with different sewage sludge application rates.

ity standard (Grade A soils apply to natural conservation areas, drinking water catchments, tea garden). For reference, the concentration limits for the various soil grades are shown in Table 6 [extracted from GB15618-1995]. To sum up, it can be seen that Cd is the restricting element for the land application of the sewage sludge produced by the Plant. According to the field experiments the application of the sewage sludge should no longer be applied to the farmland, vegetable, tea, fruit and grazing lands, and only be applied to the forestry and grassland.

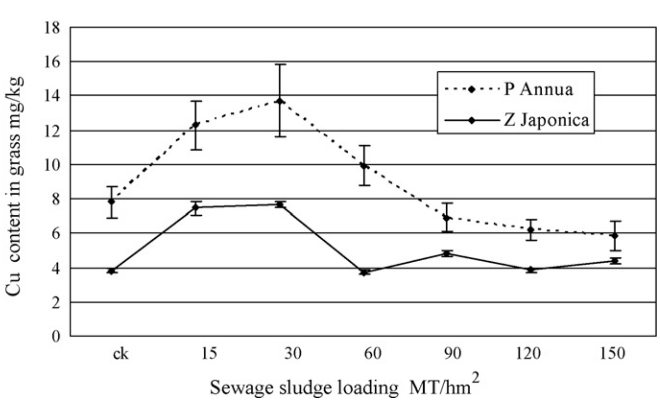

Fig. 3. Cu content of grasses grown in soils with different sewage sludge application rates.

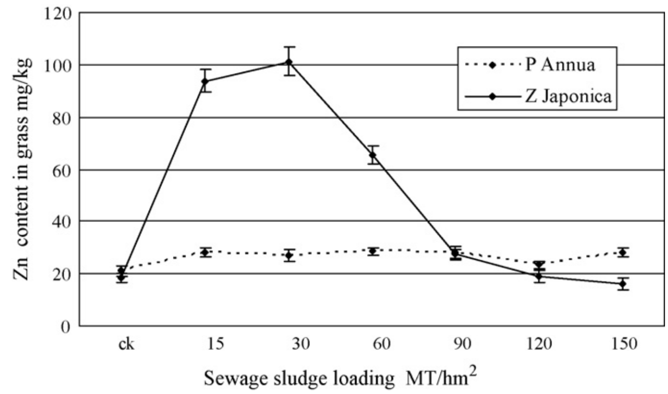

Fig. 4. Zn content of grasses grown in soils with different sewage sludge application rates.

3.4. Heavy metals content of grass tissues of P. annua and Z. japonica

The heavy metal contents of the two grass species are shown in Figs. 1–4. Fig. 1 shows that Cd content in *Z. japonica* tissue is different with sewage sludge loading rates. At the treatment of loading rates at 15, 30, 60 and 90 t ha^{-1}, Cd levels are significantly higher than control. The highest content of Cd in *Z. japonicai* tissue is found at sludge loading rates of 30 t ha^{-1}. The Cd content of *P. annua* rises with an increase of sludge loading rate. In general the Cd content of *P. annua* is higher than the control (significant difference) at all sludge treatment levels.

The concentration of Pb (Fig. 2) in *Z. japonica* tissue increased with sewage sludge loading rates (significant difference compared with control) and it is obvious that *Z. japonica* has the stronger ability to take up Pb. The uptake of Pb by *Z. japonica* has a linear relationship with sewage sludge loading rate (R^2 = 0.9803). *Z. japonica* is therefore a suitable plant for remediation in Pb polluted soil.

The Cu uptake of both *Z. japonica* and *P. annua* increased with sewage sludge loading rates in range of 15–30 t ha^{-1} and the uptake of Cu was significantly higher than control ($P < 0.05$). The Cu uptake was less with sewage sludge loading rate in range of 60–150 t ha^{-1}. It is obvious that grass easily absorbs and accumulates Cu from sewage sludge loading rates of 15–30 t ha^{-1}, as shown in Fig. 3.

The Zn uptake of *Z. japonica* is higher at sludge loading rates of 15, 30 and 60 t ha^{-1}. Zn uptake is decreased from a maximum at 30 t ha^{-1} treatments. The Zn uptake by *P. annua* is very small and only increased slightly with increase of sewage sludge loading rate as shown in Fig. 4.

4. Conclusion

The growth of *Z. japonica* and *P. annua* was well promoted in soil amended by sewage sludge application. Total biomass of the grasses rises with the increase of sewage sludge loading rates and biomass of *Z. japonica* is higher than *P. annua*. In addition the grass growing season was longer. Soil nutrient contents are increased with the sewage sludge application. Organic matter content in soil with planted *Z. japonica* is increased 12.79–80.80%. However, the contents of N and P were increased only slightly in the soil. In range of 5–30 t ha^{-1} of sewage sludge loading rate, *Z. japonica* accumulates greater amounts of Cd and Zn than it does at higher loading rates. In contrast *P. annua* accumulates greater amounts of Pb and Cu. The Pb uptake of *Z. japonica* has positive linear relationship with sewage sludge loading rate (R^2 = 0.9803).

Heavy metal concentration in soil increased with sewage sludge application. The ratio of Cd concentration increase in soil is higher than those for Pb, Cu and Zn. The cadmium concentration in this field experiment is above the permissible levels for environmental quality standard for agricultural soil; in contrast the concentrations of Pb, Cu and Zn are within the environmental quality standard. According to the characteristics and components of the sewage sludge produced in Northern Shenyang Wastewater Treatment Plant and the performance of field experiment, it is can be seen that Cd is the limiting factor for this kind of sewage sludge for application to the farmland, vegetable, tea, fruit and grazing lands. It is suggested that this type of sewage sludge could be applied to the forestry and grasslands or nurseries in order to avoid food chain contamination of cadmium. The characteristics and components of sewage sludge are very various and soil type, site condition and plant species as well. Therefore, the study on the limited factors case by case is prerequisite for sewage sludge land application and for realizing the resource reuse.

Acknowledgements

The research presented here was in part supported by the Project 'Pollution control for industry effluent liquid and sediment discharge' of the Chinese Academy of Sciences, and was in part also supported by Northern Shenyang Wastewater Treatment Plant, China.

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