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Comparison of heavy metal removal efficiencies in four activated sludge processes

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Abstract: The removal efficiencies of heavy metals (As, Cr, Cu, Ni, Pb and Zn) were investigated in the 17 operating municipal wastewater treatment plants (WWTPs) and compared with those in four main activated sludge processes. Significant differences of heavy metal removal efficiencies were observed among four activated sludge processes. The removal efficiency for As (75.5%) in the oxidation ditch (OD) process is significantly higher than that in the conventional activated sludge (CAS) process (38.6%) or sequencing batch reactor (SBR) process (51.4%). The mean removal efficiencies for Cu and Ni in the OD process are 90.5% and 46.7%, respectively, while low mean removal efficiencies are observed for Cu (69.9%) and Ni (16.5%), respectively, in the SBR process. The removal efficiencies for Cu and Ni in the OD process are significantly higher than those in the anaerobic-anoxic-oxic (A^2-O) process. These results highlight the differences of removal efficiencies for heavy metals in different processes and should be considered when selecting a wastewater treatment process.

Key words: wastewater; heavy metal; removal efficiency; treatment process; activated sludge processes

1 Introduction

Wastewater irrigation has resulted in heavy metal pollution of farmland soil [1−3]. Wastewater treatment plants (WWTPs) receive wastewater that contains a mixture of nutrients and organic, inorganic micro-pollutants. In WWTPs, wastewater is treated to reduce the concentrations of these compounds to minimize their impact on the environment [4−7]. Most biological wastewater treatment processes mainly remove organic matter, and the removal of heavy metals is considered a side-benefit [8]. In biological WWTPs, the metal removal efficiency depends on the metal species and concentration, the reactivity of the available biopolymers or biomass, and the composition of other wastewater components [9−10].

There is significant difference between different biological wastewater treatment processes because of the different biological and physicochemical conditions and the operating conditions of the WWTP. HASHIMOTO et al [11] evaluated the estrogenic activities of wastewaters collected from 28 full-scale wastewater treatment plants in Japan and compared the natural estrogen removal efficiencies between the conventional activated sludge process and the oxidation ditch process. Plants with a high solids or hydraulic retention times also had high estrogenic activity removal. The results also provided some information about the removal efficiencies of other pollutants in different WWTPs. However, the mechanisms for the removal of heavy metals are different from the removal of organic pollutants in wastewater. Heavy metals are removed from wastewater through bacterial absorption, surface adsorption by bacteria or particles, and co-precipitation with inorganic salts [12]. Therefore, it is difficult to analyze the removal efficiency for heavy metals by different WWTPs from the removal efficiency of organic pollutants using different WWTPs.

Some studies have reported that biological WWTPs can remove heavy metals from wastewater [13−16]. The removal efficiencies of Zn^{2+} and Cu^{2+} in a lab-scale sequencing batch reactor (SBR) process reached 87.0% and 84.9%, respectively [17]. Some studies have also investigated the removal efficiencies of heavy metals through operating municipal WWTPs. Removal efficiencies above 75% were achieved for total Cr and Cu in two municipal biological WWTPs of Bursa (Turkey) operated under five-stage Bardenpho process [15]. KARVELAS et al [8] indicated that the WWTP in

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Thessaloniki removed 58.2% of Cu, 50.0% of Cr, 44.2% of Ni, 30.8% of Pb, 54.5% of Cd and 42.5% of Zn from wastewater using activated sludge process. In Bursa (Turkey), a WWTP using an activated sludge process removed 94.8% of Cr, 71.7% of Cu, 47.0% of Ni, 64.3% of Pb and 71.9% Zn from wastewater [18]. It had been reported that the metal removal efficiencies are affected by both metal ion speciation and concentration [19−20]. However, there are only a few studies on the removal efficiency of heavy metals at WWTPs using different processes.

In this work, the changes in heavy metal (As, Cr, Cu, Ni, Pb and Zn) concentrations during the wastewater treatment process were investigated in the 17 operating municipal WWTPs in Beijing. We studied the removal efficiencies of heavy metals in the municipal WWTPs and compared the heavy metal removal efficiencies of the four main activated sludge treatment processes used in the 17 municipal WWTPs. We have also discussed the relationships between the wastewater treatment processes and the heavy metal removal efficiencies.

2 Materials and methods

2.1 Description of WWTPs

There are four main types of activated sludge process employed by municipal WWTPs in Beijing: the conventional activated sludge (CAS) process, the oxidation ditch (OD) process, the anaerobic-anoxic-oxic process (A^2-O), and the sequencing batch reactor (SBR) process. There were 17 full-scale municipal WWTPs in Beijing (Fig. 1): three WWTPs using the CAS process; six using the OD process; four using the A^2-O process; and four WWTPs using the SBR process (Table 1).

2.2 Sample collection

Samples of the influent, effluent and sewage sludge were collected. At each sampling site, three water

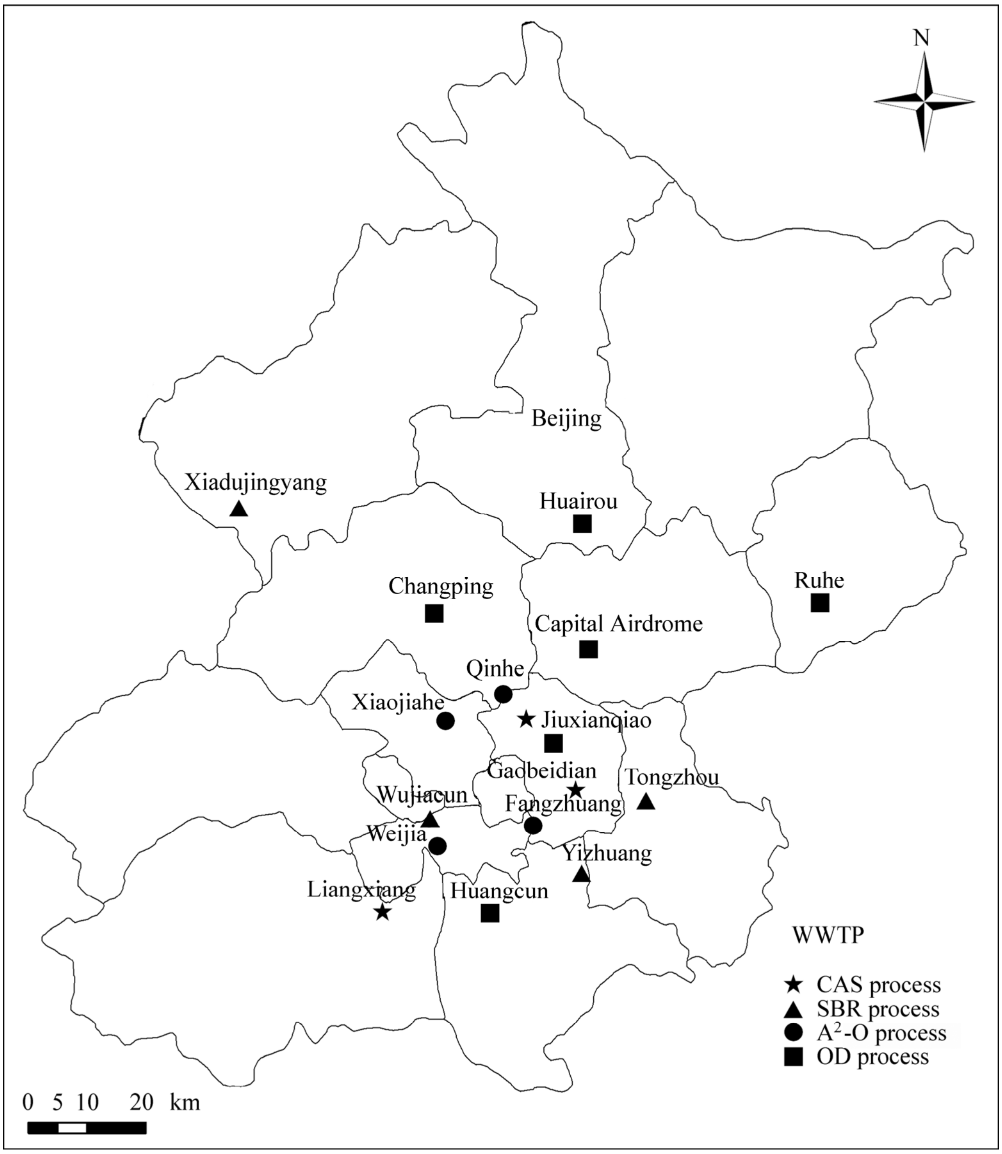

Fig. 1 Locations of investigated wastewater treatment plants in Beijing city, China

Table 1 Location and types of treatment processes in Beijing WWTPs

Location	WWTP	Treatment process	Capacity/ $(10^4 m^3 \cdot d^{-1})$	Sewage sludge$^{a)}$/ $(t \cdot d^{-1})$
Chaoyang district	Gaobeidian	CAS Process	100	500−600
	Jiuxianqiao	OD Process	20	250−300
	Beixiaohe	CAS Process	10	70−80
Haidian district	Qinhe	A^2-O Process	20	90−100
	Xiaojiahe	A^2-O Process	2	15−16
Fengtai district	Weijia	A^2-O Process	10	50−70
	Wujiacun	SBR Process	8	100
	Fangzhuang	A^2-O Process	4	30−40
Daxing district	Huangcun	OD Process	5	70
	Yizhuang	SBR Process	5	40
Shunyi district	Capital Airdrome	OD Process	8	10−15
Changping district	Changping	OD Process	5.4	30−50
Tongzhou district	Tangzhou	SBR Process	4.5	50
Pinggu district	Pinru Ruhe	OD Process	4	40
Huairou district	Huairou	OD Process	3.5	30−40
Yanqing county	Xiadujingyang	SBR Process	3.0	25−35
Fangshan district	Liangxiang	CAS Process	2.7	1.5−2.0

a) Water content is 80%

samples were collected at 1 h intervals and mixed before analysis. The influent samples were collected at locations before being affected by any recycle flow from the sludge treatment facilities. Effluent samples were collected at the outlet from the secondary clarifier. To ensure that the samples of sewage sludge were representative, sewage sludge was collected by taking subsamples from various points in the storage pile and then bulking them together. The collected wastewater and sludge samples were placed in 1.5 L plastic bottles, acidified with 15 mL of nitric acid (HNO_3), and stored at 4 °C [21].

2.3 Analysis of heavy metals

The sewage sludge samples were dried using a freeze dryer (FD-18 Detianyou Co., Beijing, China), ground and passed through a 0.149 mm nylon sieve, and then digested with HNO_3 in accordance with USEPA method using a Microwave Digestion System (Mars 5, CEM. PyNN Co., NC, USA) [22]. The wastewater samples were also digested with nitric acid according to standard methods [21]. The digested extracts were analyzed for Cr, Ni, and Pb by inductively coupled plasma mass spectrometry (ICP-MS, Perkin Elmer SCIEX ELAN DRC-e, Massachusetts, USA). An inductively coupled plasma-optical emission spectrometer (ICP-OES, Perkin Elmer, optima 5300DV, MA, USA) was used to measure the Cu and Zn concentrations in the digests. The As concentrations in water and sewage sludge were determined with an atomic fluorescence spectrometer (AFS 2202, Haiguang Co., Beijing, China).

Quality control samples, including certified liquid samples (catalogue number GSBZ50005−88, GSB07−1187—2000, GSB07−1182—2000, GSB07−1186—2000, GSB07−1183—2000 and GSB07−1188—2000, National Standard Reference Materials Center, China) and sewage sludge samples (catalogue number RTC-CRM055, National Standard Reference Materials Center, China) were analyzed to ensure the accuracy of all analyses. The certified samples were used to check analytical accuracy, and the recovery rates ranged between 1% and 10%.

2.4 Calculation of removal efficiencies

The heavy metal removal efficiencies were calculated from the ratios of the effluent to influent heavy metal concentrations [8, 23]. The removal efficiency ε for each heavy metal in the WWTP was calculated using the following equation:

$$\varepsilon = \frac{C_{\mathrm{i}} - C_{\mathrm{e}}}{C_{\mathrm{i}}} \tag{1}$$

where C_{i} is the heavy metal concentration in influent and C_{e} is the heavy metal concentration in effluent.

All statistical analyses were performed using Microsoft Excel 2003 and SPSS V11.3 (SPSS Corporation). The locations of the investigated WWTPs in Beijing were mapped using the ArcGIS V9.1 (ESRI Corporation).

3 Results and discusssion

3.1 Heavy metals in influent, effluent and sewage sludge of WWTPs

The heavy metal concentrations of the influent wastewater (Fig. 2) in Beijing were not significantly different from the concentrations in influent wastewater from other cites, as all the influent samples were mainly municipal wastewater. The relative abundances of the metals in the influent samples from Beijing were in the order of Cr<As<Pb<Ni<Cu<Zn, which is consistent with the results observed in other cities [8, 19]. The heavy metal concentrations in the effluent decreased significantly after the treatment through the WWTPs. The heavy metal concentrations in the effluent did not exceed the values specified in the standards for irrigation water quality (GB 5084 — 2005) in China. The

concentrations of As, Cr, Cu, Ni, Pb and Zn in sewage sludge were respectively 2388, 19577, 9366, 2003, 4881 and 7167 times the heavy metal concentrations in the influent wastewater, indicating that the majority of Cr, Cu, Pb and Zn in wastewater streams was removed and mainly accumulated in the sewage sludge (Fig. 2).

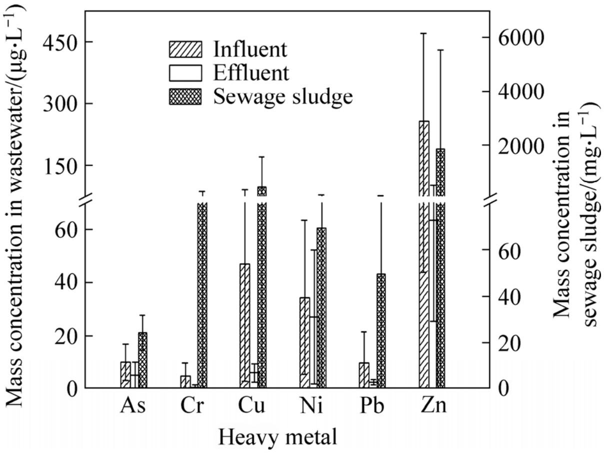

Fig. 2 Concentrations of heavy metals in influent, effluent and sewage sludge in WWTPs

3.2 Removal efficiencies of different heavy metals

The heavy metal removal efficiencies in the WWTPs of Beijing are shown in Fig. 3. The statistical result indicated that the WWTPs removed 70.2%−79.7% of Cr, Cu, Pb and Zn from influent, on average, and the mean removal efficiencies of Cr and Cu were 79.7% and 78.9%, respectively. However, the mean removal efficiencies for As and Ni were only 61.0% and 30.0%, respectively.

Heavy metal removal efficiency depends on the metal species and concentration in biological wastewater treatment plants. ÜSTÜN [18] indicated that heavy metal removal efficiencies were directly proportional to metal influent concentrations by 1-year trial. Howerver, other conditions such as operating parameters, and physical, chemical, and biological factors may also affect the removal efficiency. For example heavy metal removal by activated sludge processes is dependent on dissolved organic matter and pH [24].

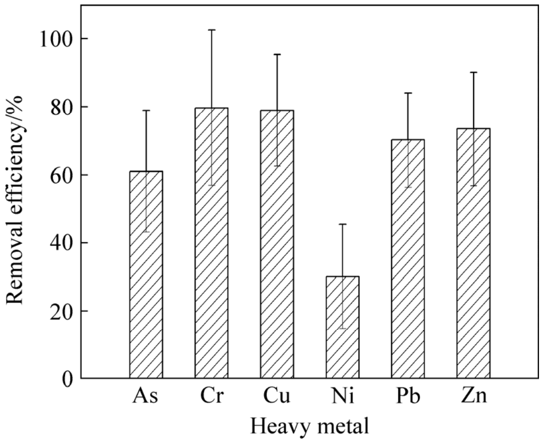

Fig. 3 Removal efficiencies for heavy metals in Beijing WWTPs (*n*=17), China

The removal efficiency of Ni (30.0%) was the lowest among all the heavy metals. Nickel is more mobile than other heavy metals in wastewater [25], and is therefore more difficult to remove by adsorption onto bacteria or particles. CHANPIWAT et al [12] previously reported an average Ni removal efficiency of only 18.6% from six municipal WWWPs using an activated sludge process, with lower removal efficiencies of other heavy metals also reported. In another work, the activated sludge process removed 47.0% of Ni from wastewater in urban WWTPs from east Bursa, but the removal efficiency of Ni was still much lower than the removal efficiencies of Cr (94.8%), Cu (71.7%), Pb (64.3%) and Zn (71.9%) [18]. Nickel is mainly (80%−93%) present in the dissolved phase, in contrast with Cu, Cr, Pb and Zn, which are mostly (75%−95%) associated with the particulate phase [8]. Surface adsorption by bacteria or particles is key mechanism for the removal of heavy metals from wastewater in WWTPs [10].

The mean removal efficiency of As was 61.0% in the WWTPs which is lower than the removal efficiencies of Cu, Cr, Pb and Zn. The predominant forms of N, P and As are as anions in aqueous solution, and all three anions exhibit similar chemical behavior [26]. Previous studies have indicated that there is a lot of phosphate (PO_4^{3-}) and nitrate (NO_3^-) in wastewater, with the concentrations of PO_4^{3-} and NO_3^- approximately 2000 and 300 times the As concentration, respectively, in influent wastewater [27−29]. The PO_4^{3-}, NO_3^-, arsenate (AsO_4^{3-}) and arsenite (AsO_3^{3-}) ions have similar chemical properties, and are competitively adsorbed onto solid surfaces [30−31]. Thus, the competing ions (PO_4^{3-} and NO_3^-) in the wastewater may competitively adsorb onto the activated sludge surface, and therefore reduce the removal efficiency of As in municipal WWTPs.

3.3 Correlation analysis of heavy metal concentrations in influent and effluent wastewaters

The As and Ni concentrations in the influent were positively correlated with the effluent concentrations, but there was no statistical correlation between influent and effluent concentrations for the other heavy metals (Table 2). The lower removal efficiency of Ni in the WWTPs meant that 70.0% of the influent Ni was present in the effluent. However, the Cr, Cu, Pb and Zn concentrations in effluent were low relative to the influent concentrations because of their high removal efficiencies in the WWTPs.

Theoretically, there should be positive correlation between the effluent and influent concentrations for all

Table 2 Correlation coefficients between heavy metal concentrations in influent and effluent (n=17)

As	Cr	Cu	Ni	Pb	Zn
0.501*$^{a)}$	0.122	0.183	0.953**	0.165	0.336

$^{a)}$ There is a significant correlation between heavy metal concentration in influent and effluent.

heavy metals, but random errors (such as analytical and sampling errors) can reduce the accuracy of the heavy metal effluent concentrations [32−33]. This error can then affect the correlation between the influent and effluent concentrations, especially where there is a high removal efficiency for the metal. Therefore, significant positive correlations between influent and effluent were observed for Ni and As, both of which had low removal efficiencies, and the correlation coefficient for Ni was higher than that for As. There was no significant relationship between the influent and effluent concentrations for other heavy metals.

3.4 Comparison of removal efficiencies between different processes

OD process has the highest removal efficiencies for heavy metals in influent (Fig. 4). Table 3 shows that most of heavy metals in the influent were not significantly different for WWTPs with four different activated sludge processes. The heavy metal concentrations in the effluent were significantly lower than the influent concentrations after treatment through the OD process, with removal efficiencies for As, Cr, Cu and Ni of 75.5%, 91.8%, 90.5% and 46.7%, respectively. The OD process was more effective for removing As than the CAS process; and the removal efficiencies of As, Cu and Ni in the OD process were significantly higher than in the SBR process. HASHIMOTO et al [11] found that the oxidation ditch process removed 62%−98% (median 90%) of the estrogenic activity in wastewater, higher than the CAS process, which had removal efficiencies of 46%−90% (median 70%), indicating that the OD process has a high removal efficiency for many types of pollutants in wastewater.

The OD process was more effective for removing heavy metals from municipal wastewater when compared with the CAS and SBR processes because of the high liquor suspended solids (MLSS) concentration used in the OD process. Heavy metals in wastewater are typically removed through bacterial absorption, surface adsorption by bacteria or particles, and co-precipitation with inorganic salts [12, 34−36]. The MLSS concentration

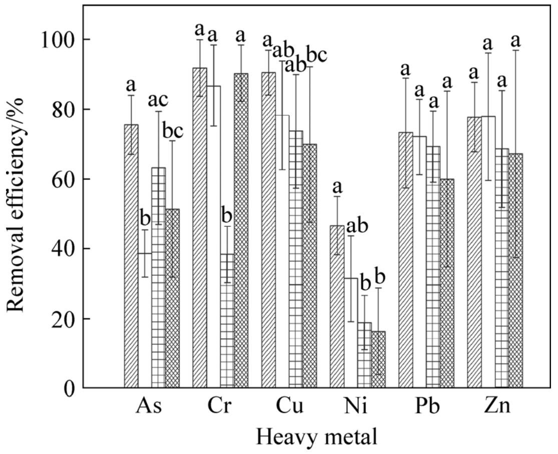

Fig. 4 Heavy metal removal efficiencies of different wastewater treatment processes (Values with different letters indicate that the heavy metal removal efficiencies are significantly different at ρ=0.05 level among different wastewater treatment processes)

Table 3 Concentrations of heavy metals in influent, effluent of different wastewater treatment processes

Element	Sample	Mass concentration of heavy metals/(μg·L^{-1})			
		A^2-O process (n=4)	CAS process (n=3)	OD process (n=6)	SBR process (n=4)
As	Influent	8.29±3.91$^{1)}$ a	5.70±3.18 a	16.9±6.24 a	9.54±0.86 a
	Effluent	3.58±1.27	3.49±2.36	5.00±5.04	5.26±3.00
Cr	Influent	2.19±1.53 a	3.48±7.15 a	6.69±6.36 a	1.69±3.05 a
	Effluent	0.82±0.82	0.278±0.06	1.01±1.07	0.20±0.86
Cu	Influent	20.1±8.68 a$^{2)}$	26.6±51.1 ab	60.5±34.0 bc	24.4±79.1 ac
	Effluent	4.52±2.09	4.09±0.41	6.52±4.30	6.30±3.45
Ni	Influent	16.9±5.34 a	18.8±2.34 ab	43.4±28.8 bc	38.1±70.2 ac
	Effluent	13.7±2.90	12.8±2.26	25.7±33.3	31.6±50.9
Pb	Influent	7.95±4.14 a	7.21±6.80 ab	14.4±17.9 ab	3.48±1.16 bc
	Effluent	2.34±1.23	1.92±0.96	3.10±0.58	2.81±0.06
Zn	Influent	281±336 a	237±289 a	215±117 a	141±39.2 a
	Effluent	78.4±49.4	34.8±12.7	50.2±22.0	69.0±56.9

Note: 1) Data are mean value; ± is the standard deviation. 2) Values with different letters indicate that the heavy metal concentrations in influent are significantly different at p = 0.05 level among different treatment processes.

is an important index to describe the content of active microflora in drainage engineering [37−39]. Increasing the MLSS concentration can improve the bacterial absorption and surface adsorption abilities of the sludge [17]. The MLSS concentration in the OD process (2500−4500 mg/L) is typically higher than that in the CAS (1500−2500 mg/L), SBR (1500−3000 mg/L) process or A^2-O process (2000−4000 mg/L) [40]. Consequently, the removal efficiencies for heavy metals in the OD process were also higher than those in the other processes.

4 Conclusions

1) The operating municipal WWTPs in Beijing exhibit high mean removal efficiencies for Cr, Cu, Pb and Zn from wastewater of 79.7%, 78.9%, 70.2% and 73.5%, respectively. The mean removal efficiencies of Ni (30.0%) and As (61.0%) are lower than the other heavy metals in all treatment plants.

2) The removal efficiencies for As, Cu and Ni in the OD process are also significantly higher than those in the SBR process, with the lowest removal efficiency for Ni in the SBR process, averaging only 16.5%. The removal efficiencies for As in the OD process averaged 75.5%, which is more efficient than the CAS process and A^2-O process.

3) The OD process has high metal removal efficiencies during the treatment of municipal wastewater and should be considered when selecting a wastewater treatment process.

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