



Removal of heavy metals from compost leachate using an anaerobic consecutive system of anaerobic migrating blanket reactor-anaerobic sludge bed reactor

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ABSTRACT

The current work studied the removal of heavy metals (HMs) in the anaerobic migrating blanket reactor (AMBR) and anaerobic sludge bed reactor (ASBR) installed consecutively. In this work, the organic loading rate (OLR) was adjusted to vary from 1.8 to 19.7 g·COD/L·d at 11 runs for a period of 280 d. After the preparation of the samples, the Series Optima Perkin Elmer 4000 ICP-OE 5 was utilized to measure the level of heavy metals including Cr, Zn, Ni, Cu, Cd, and Pb. The minimum and maximum rates of heavy metal removal were 86.18% for Cu and 94.5% for Cd. In the studied AMBR-ASBR consecutive system, the observed order of heavy metal removal efficiency was as follows: Cd > Pb > Ni > Cr > Zn > Cu. The consecutive AMBR-ASBR system is an efficient system for the removal of heavy metals, and it has high efficiency of 87.3% ± 4.63%. Increasing the OLR to 10.1 improved the removal efficiency of copper, chromium, and cadmium, and also enhanced the removal efficiency of zinc up to 19.7. Investigation of the heavy metal concentration in the sludge showed that most of the metals removed from the leachate had accumulated in the sludge and biomass. Therefore, biosorption and adsorption of metals on the biomass was the main process in the removal of heavy metals from compost leachate. Moreover, the results of this study showed that heavy metals in compost leachate can be effectively removed in a sequential AMBR-ASBR reactor by the biosorption process.

Keywords: Compost leachate; Anaerobic migrating blanket reactor; Anaerobic sludge; Bed reactor; heavy metals

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1. Introduction

The activities of human communities and industries have led to the widespread production of waste and resulted in environmental pollution, and with the growth of communities, the amount of pollution is constantly increasing [1,2]. The changes in demographic, economic, and consumption patterns have increased the volume of waste produced in different communities [3,4]. By increasing the amounts of solid waste, it is necessary to develop, and promote solid waste management methods such as composting. The weakness in the management of waste and its entry into the environment has harmful effects on humans and the environment [5,6].

Waste composting is a method in which microorganisms decompose organic matter; during this process, a liquid called leachate exits from the bottom of the reactor [6]. The leachate is a smelly, dark brown liquid leaking from waste due to various biological and chemical interactions; it contains high amounts of suspended solids and organic and mineral solutions [7]. The composition of the leachate produced from waste varies from site to site, depending on the waste matrix, the hydrological conditions, the method of utilization of disposal tools, and climatic conditions. However, in general, leachate has high levels of organic contamination, ammonia, heavy metals, halogenated hydrocarbons, and inorganic salts [8,9]. In a review study conducted by Roy et al. [10], regarding the quality and quantity of the contaminants in compost leachate, the average chemical oxygen demand (COD) in MSW was equal to 48 g-O₂/L. The average biochemical oxygen demand (BOD₅)/COD ratio was also reported to be around 0.33. Moreover, the average total Kjeldahl nitrogen (TKN) and the average concentration of total phosphorus were about 2180 mg-N/L, and 242 mg-P/L, respectively. Based on the previously published studies, the average concentration of heavy metals in leachate from MSW compost is 0.36 mg-Cd/L, 0.61 mg-Cu/L, 1.06 mg-Ni/L, 0.64 mg-Pb/L, and 6.93 mg-Zn/L.

In places where waste is dumped above the static water level, the chemical and biological contaminants present in the leachate move vertically and rapidly depending on the soil properties, causing severe groundwater contamination. The movement of chemical solvents is faster than biological pollutants. Bacteriological contaminants cannot move more than 1 m in the soil. In addition, suspended solids lose their permeability at high distances [7].

Many types of researches on leachate have shown that the penetration of the leachate into the soil increases the amount of lead, chromium, cobalt, and absorbable nickel in the soil. It also reduces soil pH, which in turn increases the amount of absorbable iron, copper, zinc, and manganese. Due to the acidic pH of the leachate and the dissolution of a part of the soil, the amount of calcium dissolved in the soil increases, and the concentration of sodium and the soluble anions increases, which elevates soil salinity [11,12]. In general, the methods used for the management and treatment of leachate can be divided into two general treatment groups, including leachate treatment mixing with urban wastewater or leachate treatment separately [13]. However, due to the specifications of the leachate, it is necessary to treat it separately [14].

Traditional treatment methods such as flocculation, coagulation, settling, and air stripping are usually expensive with regard to initial plant investment, energy requirements, and repeated use of additional chemical substances. Other procedures such as active carbon adsorption, and reverse osmosis only transfer the contamination from leachate into another media and the environmental problem is remaining. Recently, advanced oxidation processes (AOPs), for example, UV/FeII+H₂O₂, UV/H₂O₂, UV/O₃, UV/TiO₂ have been suggested as an efficient solution for the mineralization of recalcitrant organics in landfill leachate. However, the application of these treatment methods is not economically justifiable for large-scale effluents [15,16].

Although the mentioned methods are more effective in removing and destroying organic compounds, their ability to remove heavy metals is not significant. AOP methods also focus more on the removal of biodegradable organic compounds and have a low ability to remove heavy metals [10]. In addition to organic compounds, nitrogen, and phosphorus, heavy metals are pollutants that are present in leachate in amounts above the allowable values [17,18]. Heavy metals exist in the waste leachate usually in the form of mineral complexes, organic complexes, and free ions. Organic and inorganic compounds are the main chemical forms of heavy metals found in the leachate; hence, a major part of heavy metals is colloid-bounds [19]. Since 1970, concerns have been raised regarding the toxic effects of heavy metals on the health of individuals as well as on aquatic ecosystems [18]. The uncontrolled entry of heavy metals into the environment can cause adverse health effects such as cancer, reduced growth, nervous system damage, and death. Contact with some heavy metals such as mercury and lead can cause autoimmunity diseases, in which the body's immune system acts against the cells of the body and causes them to degrade [20]. Heavy metals disrupt the biological processes of wastewater treatment and reduce the amount of biogas production in bio-processes; Zn and Hg, Cr, Ni, and Cd have the highest level of toxicity in these processes [21–23].

Due to the presence of excess heavy metals and toxic organic compounds with low molecular weight and high concentrations of COD and BOD₅ in the leachate, and also the presence of phytotoxic compounds such as ammonia, the treatment of these wastes with conventional aerobic wastewater treatment processes is not recommended, because it kills bacteria in aerobic processes. Therefore, the treatment of the mentioned wastewaters requires separate facilities [10].

Among the biological processes, the anaerobic migrating blanket reactor (AMBR) is a process with a high loading capability, short hydraulic life, constant flow, and uncomplicated design. In addition, anaerobic sludge bed reactor (ASBR) is a highly flexible process, which is highly capable of controlling the microbial population and has an independent biologically active retention time apart from the hydraulic retention time [24]. The use of an AMBR-ASBR series reactor with different organic loading rates (OLRs) in the removal of heavy metals from compost leachate as a new approach was investigated in this study. The main purpose of this work was to investigate the efficiency of AMBR-ASBR anaerobic system in the elimination of heavy metals from compost leachate.

2. Materials and method

2.1. Experimental design and seeding

In this study, the two consecutive processes of AMBR and ASBR on a pilot scale were used to remove heavy metals from compost waste leachate. The schematic of the reactors and their accessories are shown in Fig. 1. The samples entering the reactors was real leachate which collected from the leachate ponds in a composting plant in Isfahan. The leachate characteristics are presented in Table 2.

The characteristics of the investigated compost leachate was typical, and it was consistent with the leachate properties in similar processes in previous studies [10].

After examining the quality of the unprocessed leachate, the real leachate of the composting plant was diluted with a minimum loading of 1 g-COD/L·d and it was introduced into the AMBR reactor using a peristaltic pump with a flow rate of 1 L/d (Etatron Co., Italy). The output of the first reactor was fed into the ASBR reactor using another peristaltic pump. An electronic processor called the programmable logic controller (PLC) manufactured by Omron Corporation, Japan, was used to properly and accurately control the operation of the pumps and mixers used in the AMBR reactor. Furthermore, after analyzing the COD:N: P ratio of the raw leachate, if necessary, the required micronutrients were used for the enhancement of the biological activity of anaerobic reactors; in addition, ammonium chloride (NH₄CL) and potassium di-hydrogen phosphate (KH₂PO₄) was used to supply the nitrogen and phosphorus needed for the prepared reactor. Anaerobic digesting sludge obtained from urban wastewater treatment plants was used for the primary seeding of the anaerobic system. The sludge characteristics are provided in Table 3. The AMBR reactor was made of rectangular transparent Plexiglas plates with a thickness of 6 mm, with useful length, width, and height

of 43, 10, and 23.5 cm, respectively, and with a useful volume of 10 L. In this reactor, the gas space was set to be 7 cm. As shown in Fig. 1, the reactor has four spaces with equal volumes of 2.5 L. The inlet leachate first entered space 1 and then, passing through the designed baffles, it entered spaces 2, 3, and 4, respectively. The baffles were designed by placing two panels adhering to the floor and hanging from the ceiling at a distance of 1 cm apart from each other. The distance between the hanged baffle to the bottom of the reactor was 8 cm. To mix materials in the reactors, we used four LANDA reactors with a tunable timer, with an rpm of 80, a run time of 10 s, and a shutdown time of 15 min. The mixer in the last space was turned off to avoid biomass flocs from escaping. A temperature control chamber (hot water bath) and an element were used to regulate the reactor temperature at 35°C ± 1°C. The ASBR reactor (with a diameter of 10 cm and a height of 40 cm) was designed and built. Specifications and components of this reactor are also shown in Fig. 1. A two-wall reactor equipped with hot water bath, thermocouple, and thermostat was used to control the temperature at

Table 1
Characterization of leachate taken from from Isfahan Composting Plant

Raw leachate	Range	Average
COD (g/L)	80–110	95.5
BOD ₅ (g/L)	49–69.5	55.2
TSS (g/L)	14–17	15.5
TDS (g/L)	28–31.5	29.6
TKN (g/L)	1.8–2.8	2.3
TP (g/L)	0.25–0.35	0.28
EC (mS/cm)	30–37.5	33.5
pH	3.5–5.5	4.4

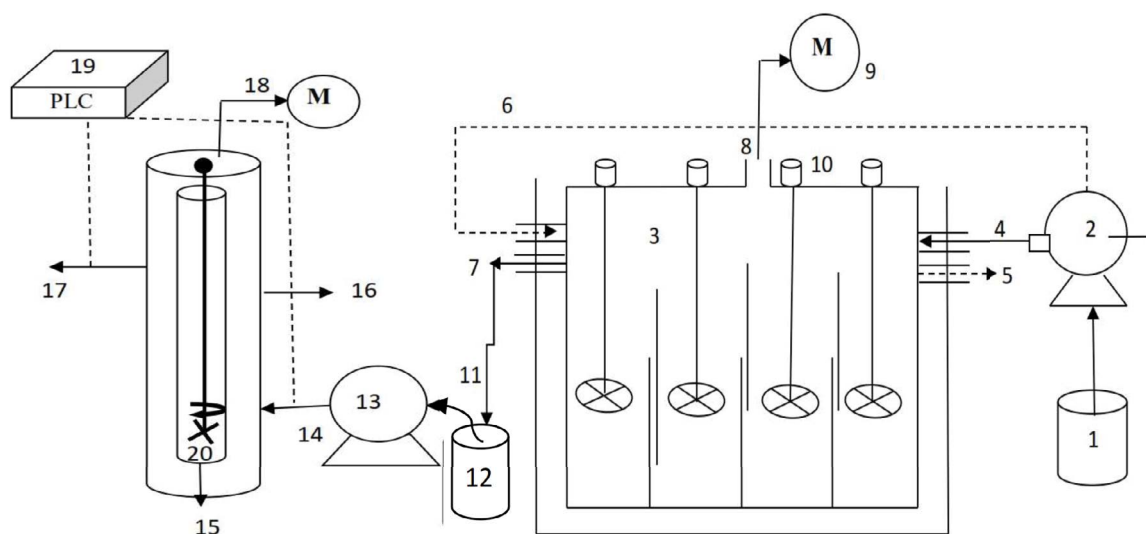


Fig. 1. AMBR and ASBR schematics (1. AMBR feeding tank, 2,13. Feeding pump, 3. AMBR reactor, 4,6. AMBR influent, 5,7. AMBR effluent, 8. Biogas output, 9,18. Gas meter, 10,20. Mixers, 11. Inlet to ASBR feeding tank, 12. ASBR feeding tank, 14. ASBR influent, 15. Sludge sampling valve, 16. Sampling valve, 17. ASBR effluent, 19. PLC).

Table 2
Characteristics of wastewater treatment plant anaerobic digestion sludge

Parameters	Amounts
TSS, mg/L	35,500
VSS, mg/L	26,650
VSS/TSS	0.75
pH	7.55

35°C as a mean to maintain the optimal conditions for anaerobic microorganisms. During the run time, environmental parameters such as temperature and pH were controlled. A precision instrumentation system (PLC) was used for controlling the performance of input and output pumps, temperature, and reaction time (mixing).

2.2. Reactor setup

Because of the high load of organic matter in the leachate, it was first diluted to a large extent and then the dilution factor was reduced over time. The maximum dilution factor was applied when the reactor started. The AMBR reactor launch was done over a period of 40 d. On the 21st day, the mean loading was 0.5 g-COD/L·d while on the 22nd day it was 0.75 g-COD/L·d. At the end of the mentioned period, COD removal performance exceeded 75%. After two runs of reactor setup and achieving the proper COD removal output, the reactors were operated with a loading of 1 g-COD/L·d.

2.3. Operation conditions

Reactors were loaded with diluted leachates for eight rounds of operation, and in the 9th round, the actual leachate without dilution was introduced into the reactors. In the 10th and 11th rounds of operation, the reactor performance was measured by doubling the input flow rate and increasing the amount of organic loading. A complete cycle of ASBR process lasted for 24 h and had four steps including filling for 4 min; reaction for 23 h and 8 min; settling for 30 min, and decanting for 18 min.

2.4. Analytical methods

The Series Optima Perk in Elmer 4000 ICP-OE 5 was applied to measure the concentration levels of heavy metals of Cr, Zn, Ni, Cu, Cd, and Pb. The concentration of each of the heavy metals was measured by using 0.5 g of the sample within 20 mL of the mixture of hydrochloric acid, nitric acid, and hydrofluoric acid (1:1:2) [25,26]. During the experiments, the parameters of SVI, MLSS, VSS, DO, pH, and temperature were monitored for the proper control of the system. In order to measure various parameters we used the methods illustrated in the standard methods for the evaluation of water and wastewater [27].

3. Results and discussion

In this study, the concentration of heavy metals in the inlet and outlet of each AMBR and ASBR system in different OLRs was measured. Based on the obtained results, it was observed that the AMBR reactor has an average efficiency of 40.43 ± 8.53 in the removal of heavy metals in different OLRs. The mean removal efficiency in the ASBR reactor was 78.57 ± 8.75 in different OLRs. Based on findings, although the concentration of heavy metals at the inlet of the AMBR reactor was higher than the ASBR reactor, the average removal efficiency in the ASBR reactor was higher than the efficiency in the AMBR reactor. Previous studies on the removal of heavy metals from acid mine drainage using an ASBR reactor showed that the removal efficiency of heavy metals such as Fe, Cu, and Zn was between 72% and 99% [28]. The other study showed that he removal efficiency of heavy metals from compost leachate in AMBR reactor was in the range of 42%–55% for Ni, Zn, Cr, Cd, Pb, and Cu which is consistent with the results of the present study [29].

Examination of the total average removal efficiency of heavy metals in the AMBR-ASBR series reactor for all studied heavy metals was 87.3 ± 4.63 in different OLRs. This finding showed that the removal efficiency in the AMBR-ASBR series reactor was higher than any of the reactors separately and was able to significantly reduce the amount of heavy metals in the effluent. According to the results presented in Table 3, the removal efficiency of the investigated heavy metals including Cd, Pb, Ni, Cr, Zn, and Cu in the

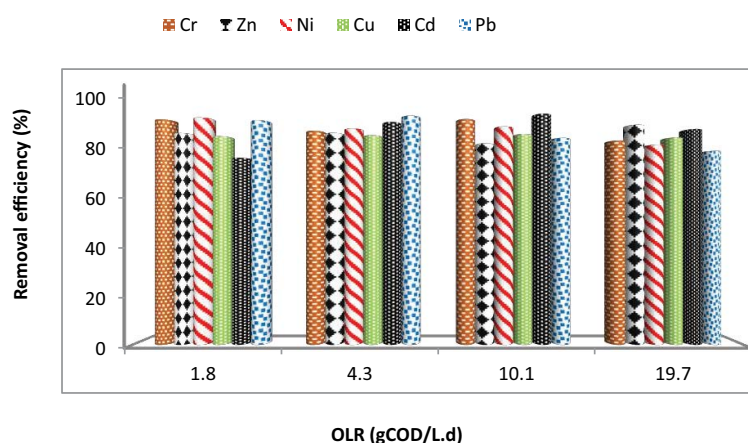


Fig. 2. Efficiency of heavy metals removal in AMBR-ASBR consecutive system.

AMBR-ASBR system was equal to 94.5, 93.7, 92.96, 92.91, 92.3, and 86.18, respectively. Fig. 2 shows the removal efficiency of heavy metals in the AMBR-ASBR series system in different OLRs. As shown in Fig. 2, the maximum removal efficiencies for the heavy metals Cu, Zn, Cr, Ni, Pb, and Cd were obtained in OLRs 10.1, 19.7, 10.1, 1.8, 4.3, 10.1, respectively. Based on the findings, copper, zinc, chromium, and cadmium had higher removal rates at higher OLRs. According to previous studies, the rate of soluble metals removal by biological processes ranges from 50% to 98%, relay upon the initial concentration of metals, concentration of solids in the biological reactor, and the solid retention time (SRT) [30].

Fig. 3 shows the concentration of heavy metals removed in different OLRs and the amount of the metals adsorbed on the sludge that was produced in the consecutive AMBR-ASBR system. As shown in Fig. 3, the concentrations of all the studied heavy metals, except for Zn, in the sludge were higher than the amounts removed in the AMBR-ASBR system; it can be justified because during the utilization of the system the amount of the sludge discharged from the system is almost zero.

With the production of biomass in the AMBR-ASBR continuous reactor and the removal of biomass by sludge

from the process, the amount of heavy metals in the effluent decreases. Determining the amount of heavy metals in the sludge accumulated in the reactor indicates the removal of metals by adsorption on the biomass. It also indicates that biosorption is the dominant mechanism. Therefore, it can be concluded that, the process of metals adsorption on the produced biomass or biosorption is the main effective process resulting in the removal of heavy metals in the AMBR-ASBR system.

The results of study conducted by Xie et al. [17], showed that the most important determinant processes in changing the concentration of heavy metals dissolved in anaerobic bioreactors for Zn, Cu, Ni, Pb and Cd metals is the adsorption process, and the complexation process for Cr. Studies have shown that metabolic-independent biosorption mechanism is the main mechanism for removing metals by biological process. The biological process of heavy metals removal is consistent with the Freundlich isotherm model. Freundlich isotherm is usually applied to state the properties of the adsorption process [30,31]. Biosorption is defined as the ability of bio-materials to form bonds, such as bonding toxic metals onto the surface of the membrane or cell wall in equilibrium reactions. Recent studies have shown

Table 3
Efficiency of heavy metals removal in AMBR-ASBR consecutive system

Heavy metals	OLR (g·COD/L·d)	HMs concentration in AMBR (mg/L)		HMs concentration in ASBR (mg/L)		HMs removal efficiency (%) in series 2 system	Removed HMs concentration in series 2 system (mg/L)	HMs concentration in the sludge of series 2 system (mg/L)
		In	Out	In	Out			
Cr	1.8	0.15 ± 0.01	0.11 ± 0.01	0.11 ± 0.01	0.010 ± 0.00	92.15	0.140 ± 0.01	0.20 ± 0.01
	4.3	0.80 ± 0.06	0.54 ± 0.04	0.54 ± 0.04	0.100 ± 0.00	87.50	0.700 ± 0.06	0.86 ± 0.06
	10.1	2.60 ± 0.15	1.38 ± 0.12	1.38 ± 0.12	0.200 ± 0.01	92.30	2.400 ± .21	2.60 ± 0.15
	19.7	2.00 ± 0.11	1.24 ± 0.07	1.24 ± 0.07	0.330 ± 0.02	83.50	1.670 ± 0.13	2.20 ± 0.17
Zn	1.8	0.42 ± 0.02	0.31 ± 0.02	0.31 ± 0.02	0.057 ± 0.00	86.58	0.360 ± 0.02	0.39 ± 0.02
	4.3	1.50 ± 0.09	0.93 ± 0.07	0.93 ± 0.07	0.200 ± 0.01	86.66	1.300 ± 0.09	1.05 ± 0.09
	10.1	4.20 ± 0.16	2.35 ± 0.12	2.35 ± 0.12	0.740 ± 0.05	82.38	3.460 ± 0.20	2.75 ± 0.22
Ni	19.7	5.00 ± 0.31	3.20 ± 0.16	3.20 ± 0.16	0.500 ± 0.04	90.00	4.50 ± 0.22	3.13 ± 0.18
	1.8	0.15 ± 0.06	0.09 ± 0.00	0.09 ± 0.00	0.011 ± 0.00	92.96	0.145 ± 0.01	0.12 ± 0.00
	4.3	0.78 ± 0.04	0.44 ± 0.03	0.44 ± 0.03	0.090 ± 0.00	88.46	0.690 ± 0.05	0.55 ± 0.03
	10.1	2.80 ± 0.10	1.43 ± 0.05	1.43 ± 0.05	0.300 ± 0.02	89.28	2.50 ± 0.20	2.87 ± 0.14
Cu	19.7	1.98 ± 0.09	1.07 ± 0.08	1.07 ± 0.08	0.360 ± 0.02	81.81	1.620 ± 0.12	1.43 ± 0.11
	1.8	0.10 ± 0.01	0.06 ± 0.00	0.06 ± 0.00	0.015 ± 0.00	85.29	0.080 ± 0.00	0.08 ± 0.00
	4.3	0.47 ± 0.02	0.27 ± 0.02	0.27 ± 0.02	0.068 ± 0.00	85.68	0.400 ± 0.03	0.45 ± 0.04
Cd	10.1	1.52 ± 0.04	0.79 ± 0.04	0.79 ± 0.04	0.210 ± 0.01	86.18	1.310 ± 0.07	1.85 ± 0.09
	19.7	2.30 ± 0.09	1.08 ± 0.07	1.08 ± 0.07	0.350 ± 0.03	84.78	1.950 ± 0.09	2.65 ± 0.21
	1.8	0.05 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	0.010 ± 0.00	76.47	0.030 ± 0.00	0.04 ± 0.00
	4.3	0.22 ± 0.01	0.15 ± 0.01	0.15 ± 0.01	0.020 ± 0.00	91.11	0.200 ± 0.01	0.12 ± 0.00
Pb	10.1	0.60 ± 0.02	0.35 ± 0.02	0.35 ± 0.02	0.030 ± 0.00	94.50	0.560 ± 0.03	1.31 ± 0.10
	19.7	0.85 ± 0.01	0.54 ± 0.04	0.54 ± 0.04	0.100 ± 0.00	88.23	0.750 ± 0.03	1.70 ± 0.10
	1.8	0.12 ± 0.01	0.07 ± 0.00	0.07 ± 0.00	0.010 ± 0.00	91.83	0.110 ± 0.00	0.09 ± 0.00
Pb	4.3	0.44 ± 0.02	0.24 ± 0.01	0.24 ± 0.01	0.020 ± 0.00	93.70	0.410 ± 0.03	0.60 ± 0.05
	10.1	1.30 ± 0.06	0.59 ± 0.03	0.59 ± 0.03	0.200 ± 0.01	84.61	1.100 ± 0.09	1.51 ± 0.10
	19.7	1.45 ± 0.04	0.75 ± 0.06	0.75 ± 0.06	0.300 ± 0.02	79.31	1.150 ± 0.08	1.63 ± 0.09

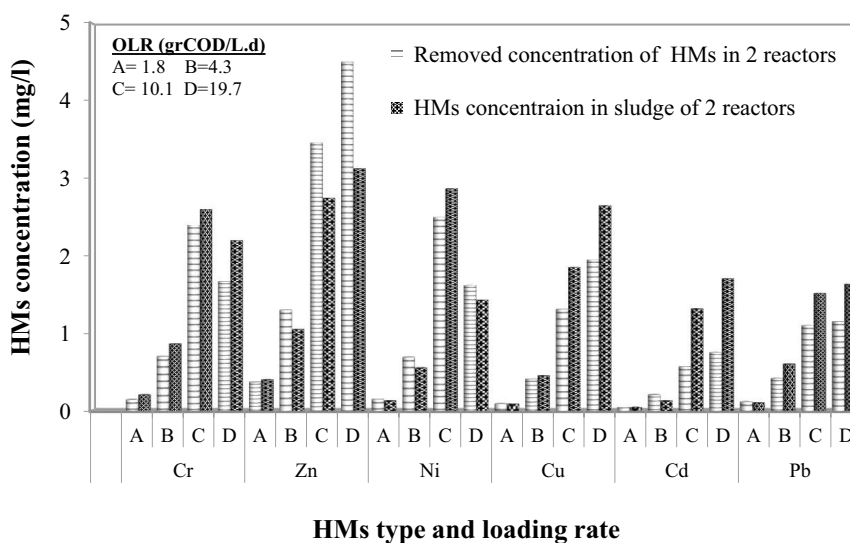


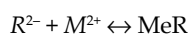
Fig. 3. Removed concentration of heavy metals in AMBR-ASBR consecutive system, and heavy metals concentration in produced sludge.

that biosorption is the result of the interaction between metal ions and functional groups that exist on the biological polymers of the cell wall of non-living organisms [32].

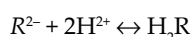
Hence, biosorption can be considered as a type of adsorption process. Biological adsorbents are biologically active substances that are capable of bonding with metals and organic compounds. The biological adsorbents can be of microbial, fungi, marine grass, herbal, or animal origin [33]. Due to the structural diversity of biological materials, the mechanism of biosorption is very complex, and it has a combination of processes including ion exchange, physical adsorption, bonding with the adsorbent surface, and fine surface deposition [33–35]. In the past, it was assumed that physical adsorption is the dominant mechanism of this process, but recent studies [36–38] have shown that this process is similar to the process of ion exchange with functional groups (amine groups, carboxyl, phosphates, sulfates, and hydroxyls) supplied by the cell wall, which is often made of polysaccharides, proteins, and lipids [39]. As a result, biosorbents can be considered weak acidic cation exchangers [40,41]. By releasing protons and metal cations from biomass, other metal cations are bond onto the cell surface. As a result, biomass acts as an organic polyelectrolyte. In this process, metal ions compete with protons over the binding sites [39].

Biosorption is affected by two factors of the biomass surface properties and physicochemical parameters of the solution (pH, ionic strength, temperature, biomass concentration, and the presence of organic and inorganic ligands in the solution) [42].

In the process of biosorption, the following equilibrium reaction occurs in the solution:



At low pH, protons compete for over binding sites:



Biosorption can be modeled by measuring the ratio between the maximum adsorption capacity in a multi-metal system to the capacity in a single metal system at different pHs [43]. In multi-metal systems, some cations compete with each other over binding sites [44]. In such a condition, the internal competition (competition between similar cations) and the competition between metal cations and protons are the same [43]. The performance of biosorption in multi-metal systems depends on various factors: the number and type of metal ions competing over binding sites, metal combinations, and concentration and type of biosorbent [44].

4. Conclusion

This study investigated the removal of heavy metals of Cr, Zn, Ni, Cu, Cd, and Pb from the waste leachate (collected from the Isfahan Composting Plant) in the anaerobic migrating blanket reactor (AMBR) and anaerobic sludge bed reactor (ASBR) in a 280-d period. The removal efficiency for all the metals studied in the AMBR-ASBR consecutive reactor was above 80%; the minimum and maximum rates of heavy metal removal were 86.18% for Cu and 94.5% for Cd, respectively. The results of the study showed that by increasing OLR, the efficiency of heavy metal removal is increased, and simultaneously the concentration of metals in the sludge or produced biomass is increased. Thus, it is concluded that heavy metals adsorption on the produced biomass or biosorption is the dominant elimination process that occurred in the consecutive system of AMBR-ASBR. Biosorption is a combination of ion exchange, physical adsorption, complex with the adsorbent surface, and fine surface deposition. The studied consecutive reactors can remove heavy metals with a high efficiency of 80% in wastes with high organic loading, such as compost leachate. It is suggested for future studies investigate the type of microorganisms present in the sludge of these bioreactors.

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