

Metal concentrations of simulated aerobic and anaerobic pilot scale landfill reactors

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Abstract

Leachate and solid waste samples from aerobic and anaerobic simulated landfill reactors operated with and without leachate recirculation were characterized in terms of metals such as Fe, Ca, K, Na, Cd, Cr, Cu, Pb, Ni, and Zn. Metal concentrations of aerobic landfill reactor leachate samples are always below the regulation limits. The higher concentrations in anaerobic landfill leachate samples decreased to regulation limits after the landfill becomes methanogenic. The effect of leachate recirculation is determined in anaerobic landfills more clearly than aerobic landfills. Metal precipitation resulted in a decrease in leachate metal content and an increase in solid waste metal content as expected. Result of the study show that the metal content of landfill leachate samples is not a major concern for both aerobic and anaerobic landfills.

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1. Introduction

Sanitary landfill leachate is the most complicated and costly wastewater to treat due to its high content of organic and inorganic pollutants. One of the most common groups of contaminants in landfill leachate is heavy metals such as chromium (Cr), nickel (Ni), zinc (Zn), copper (Cu), cadmium (Cd), lead (Pb), and iron (Fe). Landfill leachate might contain heavy metals in considerable concentrations among many other constituents. Heavy metals may constitute an environmental problem, if the leachate migrates into surface water or groundwater, or a treatment issue where the leachate is collected and treated prior to discharge. Thus, during recent decades, monitoring of heavy metals in landfill leachate has commonly been prescribed by the authorities and routinely performed by landfill operators [1]. In the acidic conditions of a landfill, however, metals such as cadmium, copper, iron, manganese, and lead can dissolve and migrate with leachate. Other chemical reactions in a landfill

can also change the state of metals, allowing them to attach to other particles and travel with leachate. Four processes have been reported to control heavy metal concentrations in landfill leachate such as complexation, oxidation–reduction reactions, sorption and precipitation [2].

There have been a large number of studies in the literature that have reported metal concentrations of landfill leachate [3–9]. Revans et al. [4], used six experimental columns filled with heavy metal rich municipal solid wastes and investigated the long-term fate of metals in landfills. The aim of the study was to determine if there would be a flash of heavy metals if the landfills were turned from anaerobic to aerobic. It is concluded from the study that as the environment changes from methanogenic to oxic, the speciation of the metals may change but the aqueous concentrations in leachate are not anticipated to increase significantly. Flayhammar and Hakansson [5], studied the mobilization of heavy metals in partly stabilized MSW during the oxidation with air. They found that the pH buffer capacity of partly degraded MSW was high enough to maintain alkaline leachates during the experiments. Therefore, the risk for unexpected high concentrations and mobilization rates of heavy metals during the oxidation of the most reactive phases of sulfides and organic matter is low. Jensen and Christensen [6] used leachate samples collected from four Danish landfills and separated them

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into size fractions in order to obtain information about size distribution of colloids and associated heavy metals using filters with pore sizes 1.2, 0.40 and 0.001 μm . The concentrations ranges of heavy metals were found to be Cd 0.2–3.6 $\mu\text{g/L}$; Ni 28–84 $\mu\text{g/L}$; Zn 85–5310 $\mu\text{g/L}$; Cu 2–34 $\mu\text{g/L}$; Cr 0–188 $\mu\text{g/L}$; and Pb 0–16 $\mu\text{g/L}$. Kjeldsen and Christophersen [7], reported the leachate composition of 106 old Danish landfills. Average metal concentrations were determined as Cd 6.8 $\mu\text{g/L}$; Ni 130 $\mu\text{g/L}$; Zn 670 $\mu\text{g/L}$; Cu 70 $\mu\text{g/L}$; Cr 76 $\mu\text{g/L}$; and Pb 70 $\mu\text{g/L}$. They have concluded that the leachate concentrations in general decreased with the age of landfill and the leachate concentrations were lower than other similar studies. Kjeldsen et al. [8] made a detailed review about the change of landfill leachate composition with waste age. They have reported wide variations in heavy metal concentrations from different landfills and concluded that metal concentrations were low. However, concentrations of heavy metals may vary depending on the differences in waste composition and landfill technology. Kalyuzhnyi and Gladchenko [9] reported average heavy metal concentrations of a Russian landfill in their study. The concentrations determined as Fe 4.08–170.4; Zn 0.86–28.8; Cu 0.04–0.24; Pb 0.038–0.1; and Cd 0.0008–0.024 mg/L . It is concluded that the concentrations were very variable and there was a noticeable difference between summer and winter values.

All of the studies in the literature are about the metal concentrations of anaerobic traditional or bioreactor landfills. There are only a few but not adequate studies in the literature about the metal concentrations in aerobic landfills. In this study, the change of metal concentrations of aerobic and anaerobic landfill leachate and solid waste samples are investigated by four landfill reactors. Metals such as Ca, K, Na, Fe, Cd, Cr, Ni, Pb, Zn, and Cu concentrations in leachate originating from four reactors were monitored and compared. The objectives of this study are: (i) to give comparatively heavy metal levels in leachate and solid waste samples from simulated aerobic and anaerobic reactors, (ii) to determine the effect of leachate recirculation on heavy metal emissions, (iii) to determine the mechanisms controlling the change of heavy metals in leachate.

2. Materials and methods

2.1. Experimental system

The laboratory-scale landfill reactors, which were constructed from 0.5 cm polypropylene and used in this study are shown in Fig. 1. Aeration pipes are used only in aerobic landfill reactors. The inner diameter and height of the reactors were 50 and 200 cm, respectively. A second layer with the diame-

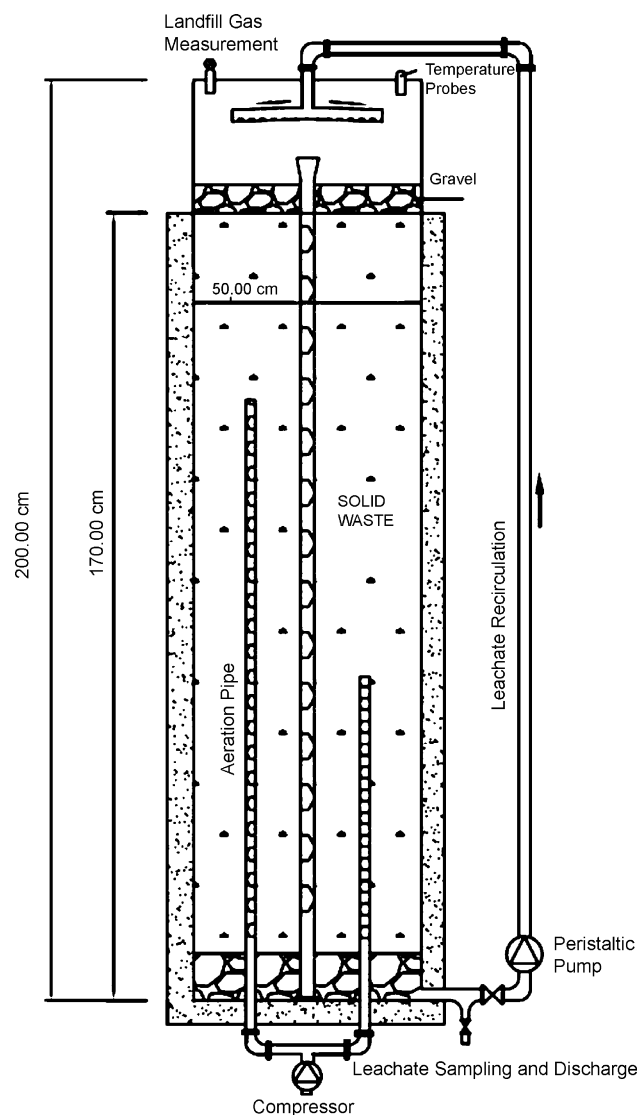


Fig. 1. Aerobic and anaerobic landfill reactors.

ter of 60 cm was constructed around the reactors and the blank between these two layers was filled with heat isolation material to prevent temperature redistribution between the reactors and the surrounding environment [10].

The lower part of the reactors consists of 15 cm gravel drainage with a perforated pipe, which has 2.5 cm diameter inserted to collect and discharge the generated leachate. Leachate collection was realized by opening the discharge valve on a daily basis at the beginning of the experiment, and at 1- or 2-week intervals for the following period. Leachate samples were collected while discharging leachate from the landfill reactors

Table 1
Operational conditions used in the reactors to simulate different landfill conditions

Column	Operating condition	Refuse (kg)	Airflow (L/min/kg waste)	Water flow (L/day/m ³ waste)
A1	Aerobic with leachate recirculation	179	0.084	0.35
A2	Aerobic dry	174	0.086	–
AN1	Anaerobic with leachate recirculation	173	–	0.21
AN2	Traditional landfill	175	–	–

and kept at 4 °C in plastic bottles. The quantity of discharged leachate for each reactor was measured and then stored in a refrigerator to use for recirculation [10].

Each column was used to simulate a different landfill concept and the operational conditions used are given in Table 1. The waste samples obtained from Odayeri sanitary landfill in Istanbul city, and the average composition of solid wastes landfilled at Odayeri landfill is 44% organic (food and yard wastes), 8% paper, 6% glass, 6% metals, 5% plastic, 5% textile, 9% nylon, 8% diaper, and 9% ash and others [11].

2.2. Experimental procedure

The aeration was achieved by a compressor that was connected to the aeration pipes at the bottom of aerobic reactors.

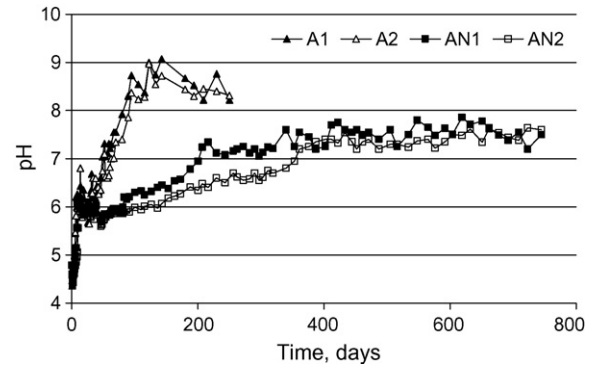


Fig. 2. The change of pH in aerobic and anaerobic landfill reactors.

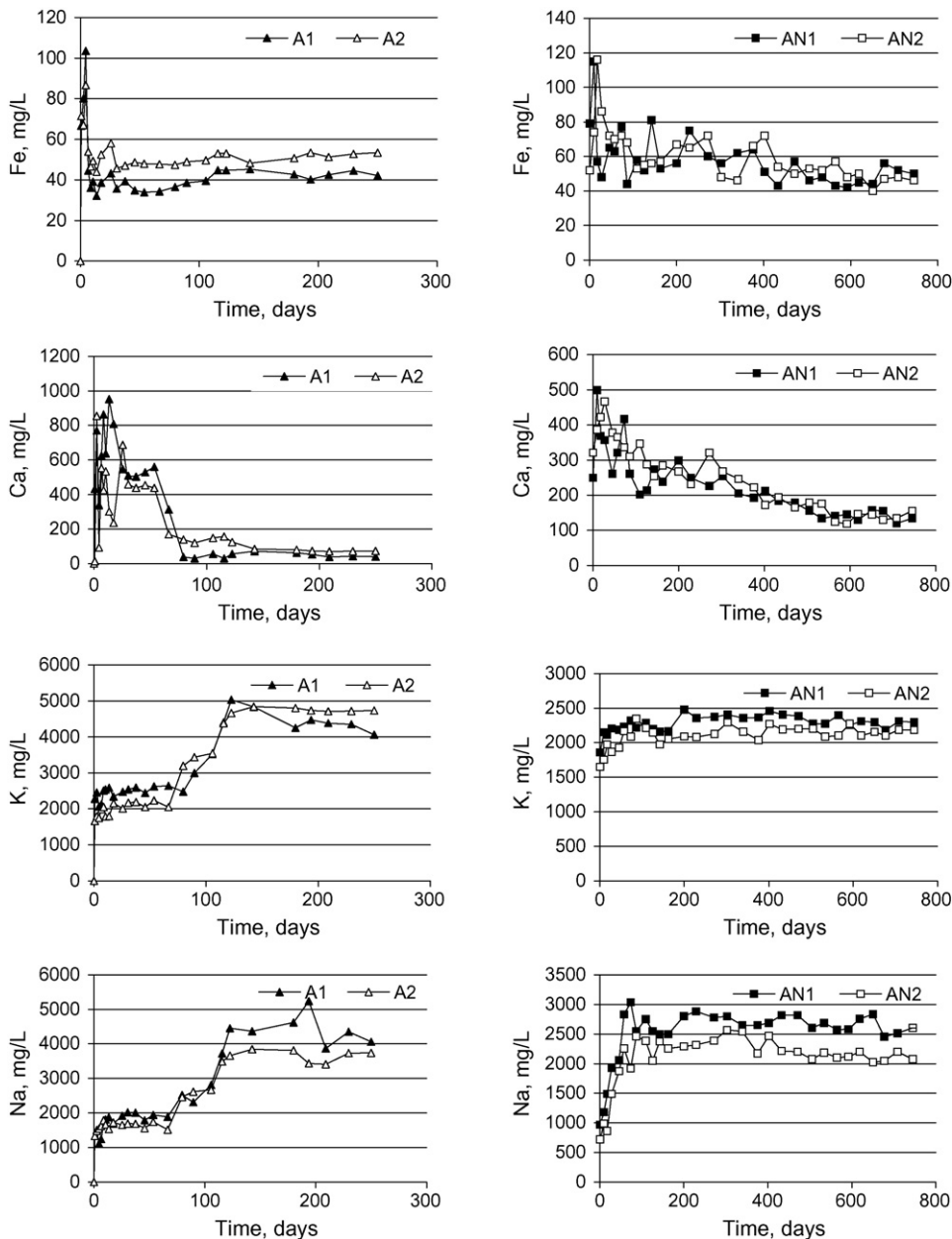


Fig. 3. The changes in Fe, Ca, K, and Na concentrations of leachate samples.

Air was introduced at the bottom of the waste and passed through the waste in an upward direction by the help of the perforated aeration pipes with 60 and 120 cm length in each aerobic reactor [12].

Leachate was recirculated using a peristaltic pump located at the top of the recirculated reactors. The recirculated leachate quantity was low at the beginning of the study. After reaching to methanogenic phase, the recirculated leachate quantity increased in AN1 reactor.

2.3. Analytical methods

The digestion of leachate and solid waste samples were realized in a microwave furnace (ETHOS 1600). The quantity of the samples used for digestion was 5 mL for leachate samples and

1 g for solid waste samples. Solid waste samples were dried at 103 °C for 24 h and ground to pass a 1.5 mm screen. Leachate and solid waste samples were put in a teflon vessel which is resistant to pressure and digested with 6 mL HNO₃, 3 mL HCl, and 0.25 mL H₂O₂. After the digestion process, the samples were filtered and the volume of the samples was completed to 100 mL. Metal concentrations determined by atomic absorption spectrophotometer (Perkin-Elmer Simaa 6000 Model).

3. Results and discussion

3.1. Leachate pH

pH is considered to be the most significant parameter affecting metal concentrations in landfill leachate. Metal dissolution

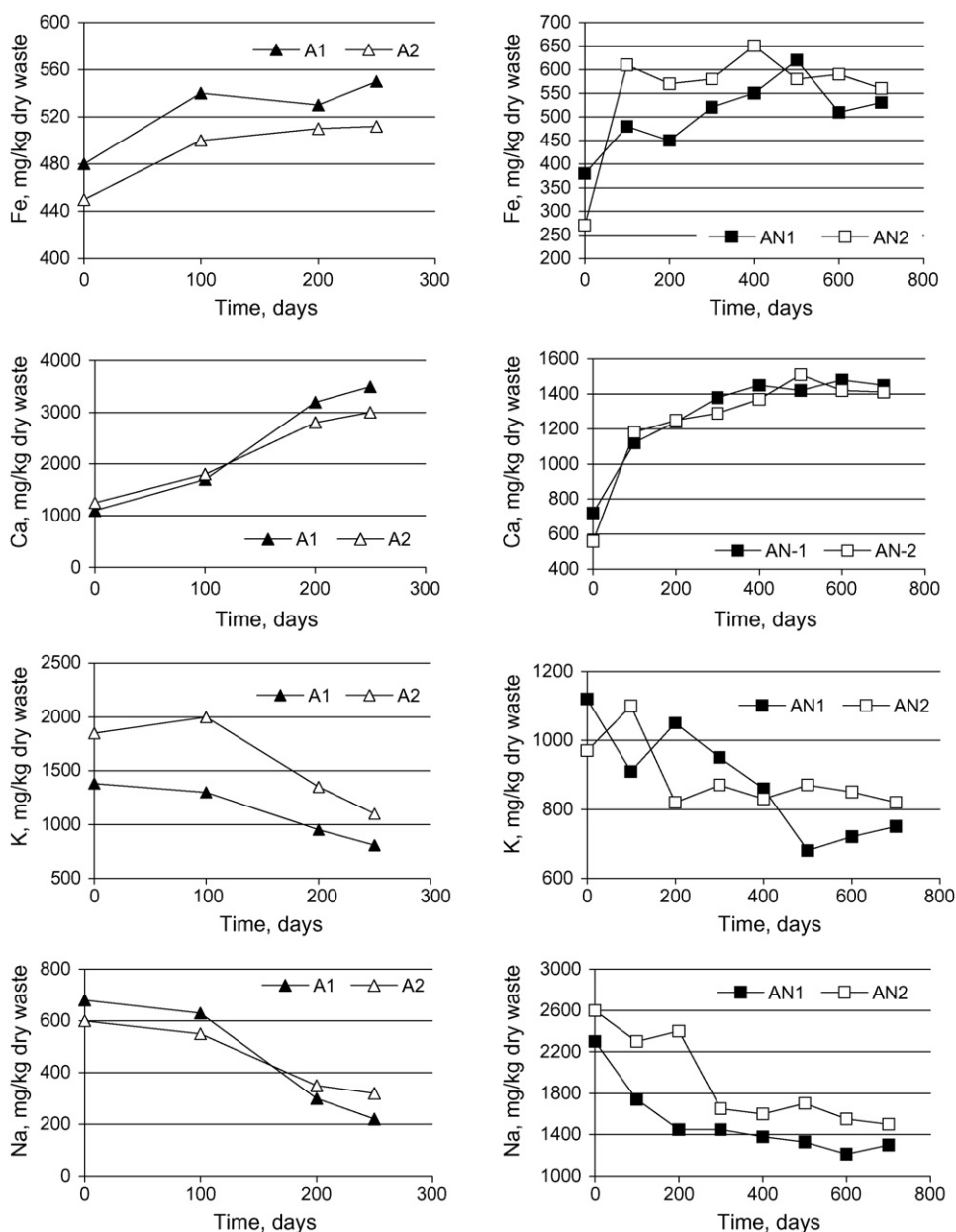


Fig. 4. The changes in Fe, Ca, K, and Na concentrations of solid waste samples.

is enhanced at low pH which is one of the characteristics of young landfill leachate. Solid wastes contain soils and organic matter that have significant sorptive capacity especially at high pH values [8,13].

The pH curves (Fig. 2) show that, pH values were in the range of 4–6 in the first 30 days of degradation in all reactors. After day 30, pH values began to increase and reached to 8 after day 100 in aerobic reactors. After that, no considerable change was observed in pH of leachate from aerobic landfill reactors and measured between 8 and 9. At day 100, the pH values were 6.3 and 6.0 in AN1 and AN2 reactors, respectively. On day 250, when aerobic landfilling operation is finished, pH of the leachate from AN1 and AN2 reactors were 7.2 and 6.7, respectively. These results show that when aerobic degradation of solid wastes completed, the AN1 reactor reaches to optimal pH values for anaerobic degradation, indicating the rapid degradation of solid wastes in aerobic conditions. These results are in accordance with the data stated by Cossu et al., Ishigaki et al., and Nakasaki et al. [14–16].

3.2. Metals—iron, calcium, sodium and potassium

The changes in Fe, Ca, K, and Na concentrations are plotted in Fig. 3. According to a statistical evaluation by Kyle-

forfs and Lagerkvist [17], the concentrations of metals are expected to reduce as the leachate changes from acidogenic to methanogenic, concurrent with a decrease in redox potential and an increase in pH. This expected decreasing trend is obvious in the Fe and Ca and but less defined in the Na and K, because of the sorption and precipitation that occur at higher pH values are effective for Fe and Ca, and have a minor effect for Na and K [2].

Fe concentrations of all the reactors were in the same range at the beginning of the landfilling operation. The concentrations decreased rapidly from 100 to 40 and 50 mg/L after 30 days for A1 and A2 reactors, respectively, while it is decreased to 50 and 60 mg/L in AN1 and AN2 reactors after 250 days, respectively.

Ca, has a tendency of complex formation with HCO_3^- and dissolved organic compounds and dissolution/precipitation reactions involving calcite (CaCO_3) and maybe siderite (FeCO_3) [2]. Thus, the precipitation process is closely linked to the dissolved carbonate compounds. As a result of the precipitation process in aerobic landfill leachate that has high alkalinity values ($\sim 10,000$ mg/L CaCO_3), Ca concentrations are decreased below 100 mg/L after day 100. The slow increase in the pH and the slowly generation of the precipitation causes a slow decrease in Ca concentrations of AN1 and AN2 reactors. Ca concentrations decreased around 100 mg/L after 600 days of landfilling in anaerobic reactors.

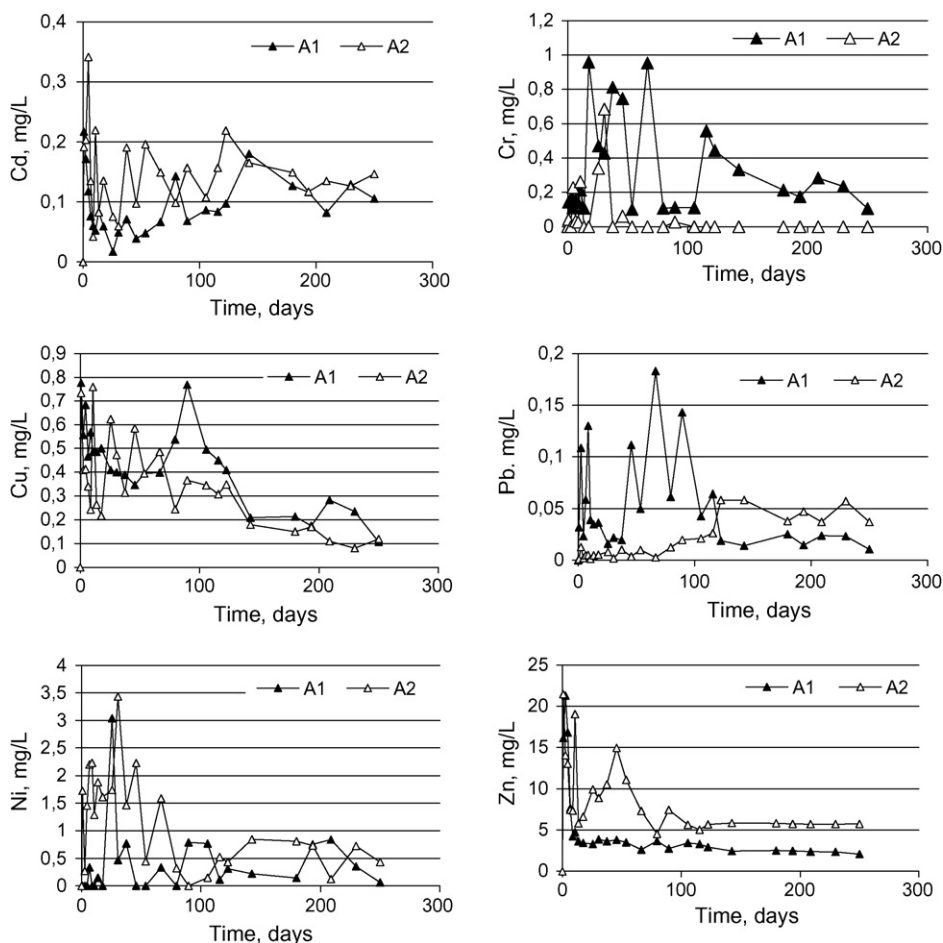


Fig. 5. The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in aerobic landfill leachate samples.

Na and K concentrations of leachate samples are in the same trend with chloride (Cl^-) concentrations [12]. This situation is confirmed by Kimmel and Braids [18] who showed a linear relationship between Na and Cl^- concentrations in water samples obtained from a leachate plume at different distances from a landfill.

K concentrations determined as 2000 and 2500 mg/L for A1 and A2 reactors, respectively, at the beginning of the landfilling. After 90 days, K concentrations reached to 5000 mg/L, and on day 250, determined as 4000 and 4700 mg/L, respectively. At the beginning of the anaerobic landfill operation, K concentrations of leachate samples were 1900 and 1700 mg/L for AN1 and AN2 reactors, respectively. There was no considerable change in both AN1 and AN2 reactors after 200 days of operations and K concentrations are determined around 2300 and 2100 mg/L, respectively, during the operation period. The same trend for Na concentrations in both aerobic and anaerobic landfill reactors can be clearly seen from Fig. 3.

The same metals are determined in solid wastes samples at the beginning and after filling of the waste to landfill reactors. Fe, Ca, K, and Na concentrations determined in solid waste samples are shown in Fig. 4.

At the beginning of the operation, Fe concentrations in A1 and A2 landfill reactors determined as 480 and 450 mg/kg, and after 100 days of aerobic landfilling, Fe concentrations were 540 and 500 mg/kg, respectively. At the end product of aero-

bic landfill reactors the Fe concentrations determined to be 550 and 512 mg/kg, respectively, almost the same as the 100 days results. The same trend is observed in Fe concentrations of the solid waste samples taken from AN1 and AN2 reactors. The concentrations determined for AN1 and AN2 reactors at the beginning and after 700 days of operations are 380; 530 and 270; 560 mg/kg, respectively. The reason of the increase in Fe concentration in solid waste samples can be explained by the precipitation process.

Ca concentrations were 1200 mg/kg in solid waste samples of aerobic reactors at the beginning of the landfilling. As a result of the precipitation process, the concentrations are increased oppositely to the trend determined in leachate samples. After 250 days, Ca concentrations are determined as 3500 and 3000 mg/kg, respectively. The same but slowly generated trend is determined in anaerobic reactors. Ca concentrations were 720 and 560 mg/kg at the beginning while the concentrations are determined as 1450 and 1410 mg/kg for AN1 and AN2 reactors, respectively, on day 700.

K and Na concentrations are decreased in solid waste samples, oppositely to the increasing trend in leachate samples. The reason for this situation is the high solubility and lower complex formation and precipitation affiliation of these elements [19]. K concentrations decreased from 1400, 1850, 1120 and 970 mg/kg to 800, 1100, 750, and 820 mg/kg in A1, A2, AN1, and AN2 reactors, respectively. Similarly, Na concentrations decreased to 220,

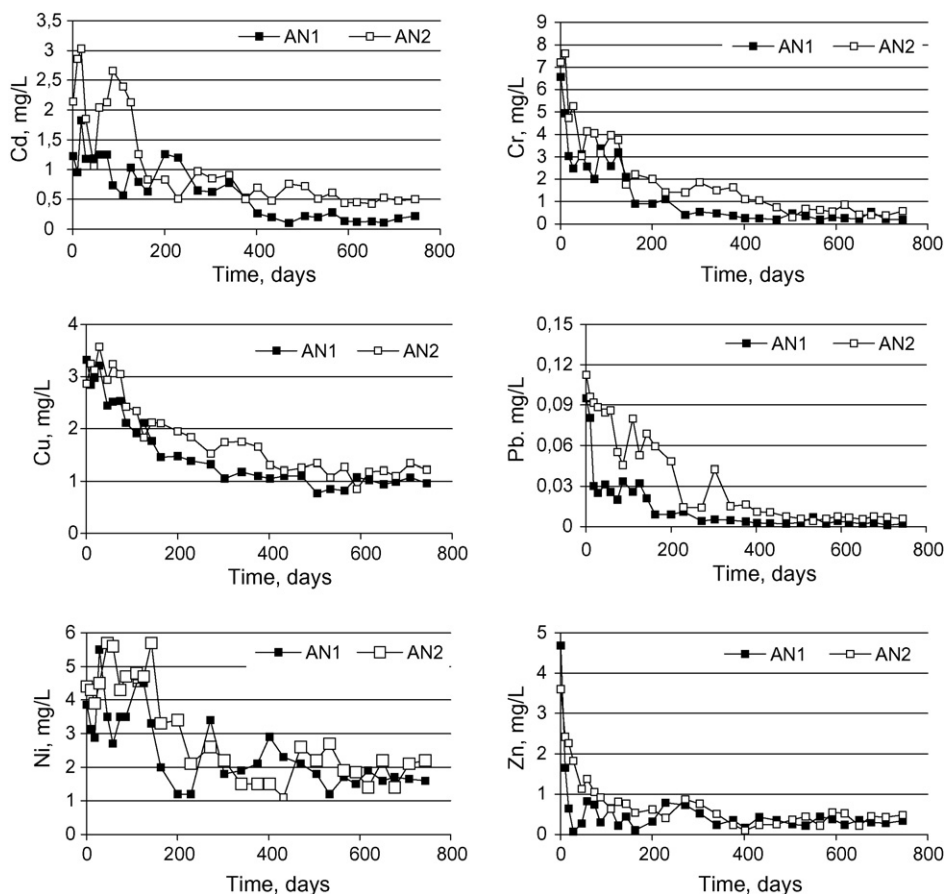


Fig. 6. The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in anaerobic landfill leachate samples.

320, 1300, 1500 mg/kg from 680, 600, 2300, and 2600 mg/kg, respectively.

3.3. Heavy metals—cadmium, chromium, copper, lead, nickel, and zinc

The main processes for the low metal concentrations in landfill leachate are sorption and precipitation. Solid wastes contain soils and organic matter, which have a significant sorptive capacity, especially at neutral to high pH values prevailing in methanogenic leachate [13]. Additionally, solubilities of the metals with sulfides and carbonates is low and sulfide precipitation is often cited as an explanation for low concentrations of heavy metals in leachate. In general, sulfide precipitation is expected to dominate heavy metal attenuation compared with complexation agents [20]. Cr is an exception to this because it does not form an insoluble sulfide precipitate. It tends to form insoluble precipitates with hydroxide [4,8].

Cd, Cr, Cu, Pb, Ni, and Zn concentrations of leachate samples collected from aerobic and anaerobic landfill reactors are given in Figs. 5 and 6, respectively.

Cd, and Pb concentrations were below 0.5 mg/L and Cr and Cu concentrations are determined below 1 mg/L in both A1 and A2 reactors. Pb concentrations determined below 0.5 mg/L in

anaerobic reactors. Cd concentrations decreased below 0.5 mg/L after approximately 400 days in AN1 reactor. Cr concentrations were 6 and 8 mg/L at the beginning of the operation in AN1 and AN2 reactors, respectively. Cr decreased below 0.5 mg/L after 350 days in AN1 reactor, and after 600 days in AN2 reactor. Cu concentrations of leachate samples were around 3 mg/L at the beginning for both AN1 and AN2 reactors. The concentrations decreased around 1 mg/L after 300 days in AN1 reactor, and after 400 days in AN2 reactor.

Maximum Ni concentrations are determined as 3 and 3.5 mg/L in A1 and A2 reactors, respectively. The concentrations decreased below 0.5 mg/L in A1 reactor, and below 1 mg/L in A2 reactor after 100 days of operation. Maximum Ni concentrations were 4.5 and 6.7 mg/L in AN1 and AN2 reactors, respectively. Ni concentrations determined to be around 2 mg/L after approximately 400 days of operation in both AN1 and AN2 reactors.

Zn concentrations in all reactors are determined around 20 mg/L at the beginning of the landfilling operation. Zn concentrations decreased rapidly to 3 mg/L after 15 days and stayed at this level during the operation period. In A2 reactor, Zn concentrations decreased to 5 mg/L after 90 days of operation and stayed at this level. Zn concentrations decreased below 1 mg/L in AN1 and AN2 reactors after 20 and 75 days, respectively.

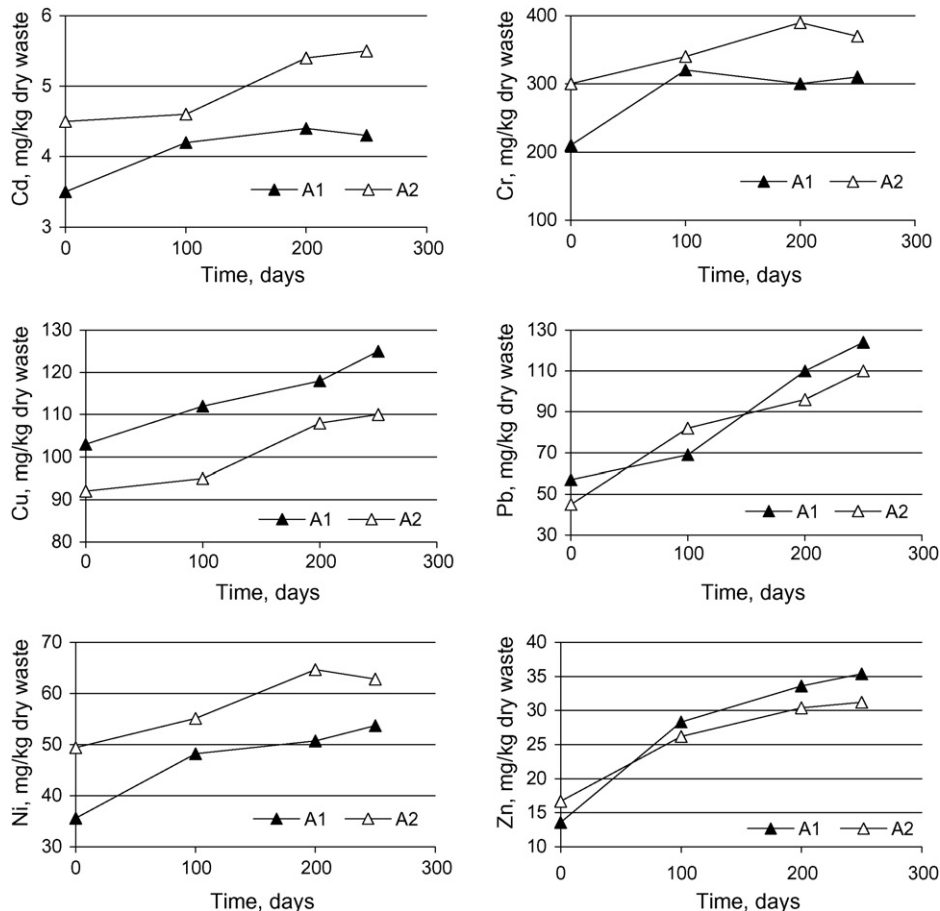


Fig. 7. The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in aerobic landfill solid waste samples.

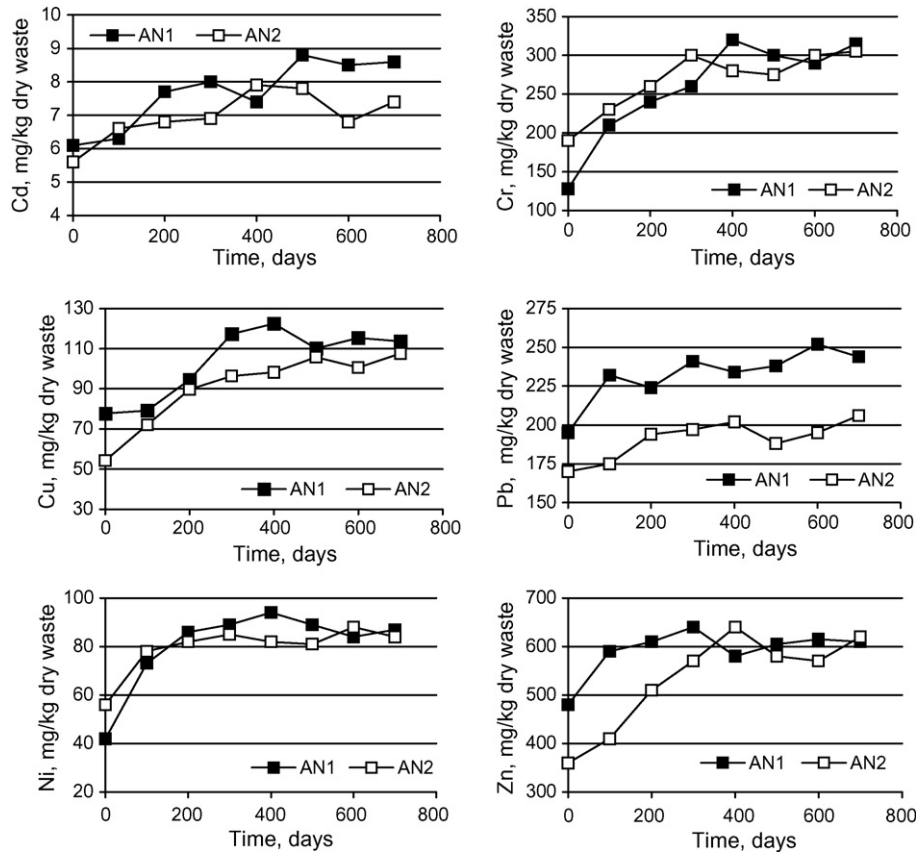


Fig. 8. The changes in Cd, Cr, Cu, Pb, Ni, and Zn concentrations in anaerobic landfill solid waste samples.

Heavy metal concentrations of solid waste samples taken from aerobic and anaerobic landfill reactors are shown in Figs. 7 and 8, respectively. All heavy metal concentrations are increased in solid waste samples. Fang and Wong [21], determined similar results in their studies and suggested that the increase in metal concentrations generates as a result of the mass loss in landfills.

Heavy metal concentration of leachate is a function of pH and carbonates. Heavy metal concentrations increase at low pH values and decreases when carbonate species increase. The pH of the leachate generated from both aerobic and anaerobic landfills is in the acidic ranges at the first stages of the degradation. Then, leachate pH values increase to neutral values. The change of the pH during the degradation in landfills also effects the metal concentrations of leachate. pH is a critical parameter when determining the solubility of metals and heavy metals have minimum solubility in the range of 7–10.

The discharge limits to sewer systems determined by Water Contamination Control Regulation of Turkish Government is given in Table 2. Aerobic landfill leachate samples are below the regulation limits in all periods of degradation. Pb and Zn concentrations are below the limits in anaerobic landfill reactors. All other metals decreased below limits in both AN1 and AN2 reactors after approximately 200 days of operation. The decrease is generated rapidly in AN1 reactor as a result of the positive effect of leachate recirculation on anaerobic degradation.

Table 2

Wastewater discharge limits to sewer systems in Turkish regulations

Parameter (mg/L)	Discharge limit
Cadmium (Cd)	2
Chromium (Cr)	5
Copper (Cu)	2
Lead (Pb)	3
Nickel (Ni)	5
Zinc (Zn)	10

4. Conclusions

pH values of the leachate samples are between 4.5 and 6.0 at the first stages of the degradation in both aerobic and anaerobic landfills. As a result of leachate recirculation the degradation rates increase and the pH reaches to neutral values earlier in leachate recirculated reactors. Thus, high pH values results in low metal concentrations in leachate samples because of the low solubility.

Metal precipitation in landfill body resulted in an increase in heavy metal content of solid waste samples. Thus, the increase in metal content of solid waste samples can be explained by precipitation of heavy metals with sulfides and carbonates. Occasionally, phosphates and hydroxides can also precipitate metals. Hydroxide precipitates form at or above neutral pH

values, which typically generates in methanogenic landfill leachate samples.

The ultimate conclusion of all studies is that heavy metal concentrations in landfill leachate are not a major concern. All metals in both aerobic and anaerobic landfill leachate samples are below regulation limit or reach to these limits after a short time. Solid wastes stabilize rapidly in aerobic landfills besides lower metal concentrations. Also, leachate recirculation effects the time to reach regulation limits in the case of heavy metals besides its effect on degradation rates. The effect of leachate recirculation can be seen more clearly in anaerobic landfills.

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