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Research Article / Araştırma Makalesi

EFFECT OF TiO_2 AND Ag NANOPARTICLES ON MIGRATION BEHAVIOR OF LEACHATE CONTAMINANTS IN AEROBIC BIOREACTOR LANDFILLS

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ABSTRACT

Nowadays, depending on the increase of usage of TiO_2 and Ag nanoparticles in consumer products, it is expected that they will reach to sanitary landfills at the end of their useful life. Also, both Ag and TiO_2 nanoparticles have a potential to migrate from porous media to groundwater according to the environmental conditions such as pH, ionic strength, concentration of the nanoparticles, and properties of the liner system. Thus, it is an important issue how the impacts of these nanoparticles on migration behaviors of contaminants from landfill leachate to the groundwater. In this study, three pilot-scale aerobic landfill bioreactors were simultaneously operated for a period of 375 days. Three reactors were loaded with solid wastes representing Istanbul municipal solid wastes. In groundwater samples, variations of pH, Cl^-, conductivity, alkalinity, chemical oxygen demand, total organic carbon, total nitrogen were investigated. The results of this study showed that migration behavior of leachate contaminants have been very different in the presence of Ag nanoparticles in aerobic bioreactor landfills. It can be concluded that Ag nanoparticles can increase transport of leachate contaminants from landfill leachate to groundwater.
Keywords: Aerobic landfill, leachate, contaminant transport, TiO_2 and Ag nanoparticles.

AEROBİK BİYOREAKTÖR DÜZENLİ DEPO SAHALARINDA SIZINTI SUYU KİRLETİCİLERİNİN TAŞINIMI ÜZERİNE TiO_2 ve Ag NANOPARTİKÜLLERİN ETKİLERİ

ÖZ

Günümüzde, TiO_2 ve Ag nanopartiküllerin tüketici ürünlerinde kullanımının artışına bağlı olarak, bu ürünlerin faydalı kullanım ömürleri sonunda düzenli depolama sahalarına ulaşması beklenmektedir. pH, iyonik kuvvet, nanopartikül konsantrasyonu gibi çevresel koşullar ile taban sistem özelliklerine göre TiO_2 ve Ag nanopartiküller gözenekli ortamdan yeraltısuyuna geçiş potansiyeline sahiptirler. Bu nedenle, düzenli depolama sahası sızıntı suyundaki kirleticilerin yeraltısuyuna geçişini bu nanopartiküllerin nasıl etkileyeceği önemli bir konudur. Bu çalışmada, üç adet pilot ölçekli aerobik biyoreaktörler eş zamanlı olarak 375 gün işletilmiştir. Reaktörler İstanbul iline ait kentsel katı atıklarını temsil eden katı atıklarla yüklenmiştir. Yer altı suyu numunelerinde pH, Cl^-, iletkenlik, alkalinite, kimyasal oksijen ihtiyacı, toplam organik karbon ve toplam azot değişimleri incelenmiştir. Çalışma sonucunda Ag nanopartikül içeren aerobik biyoreaktör depo sahalarında sızıntı suyu kirleticilerinin taşınımı çok farklı gerçekleşmiştir. Ag nanopartiküllerin, sızıntı suyu kirleticilerinin yeraltısuyuna taşınımını arttırdığı sonucuna varılabilir.
Anahtar Sözcükler: Aerobik depo sahası, sızıntı suyu, kirletici taşınımı, TiO_2 ve Ag nanopartiküller.

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1. INTRODUCTION

Nanoparticles have large surface areas, quantum effects and biological reactivities, shapes and sizes, deformability, durability, tendency to aggregate and optical sensitivity [1]. Among all nanoparticles, the use of TiO_2 in the production of daily and commercial products can improve properties such as hardness, scrub resistance, photo-catalytic activity and UV protection to products including TiO_2 nanoparticles [2]. TiO_2 nanoparticles are one of the most used with up to 10,000 t/year in the production of consumption products [3]. For instance, TiO_2 nanoparticles are blended in sunscreen lotions, paints, cosmetics, coatings and cleaning agents, plastics because of their capacity to absorb and reflect UV light and scrub resistance, [5, 6]. Moreover, Ag nanoparticles are one of the other most commonly used nanomaterials in scientific, industrial and medical applications electronics such as photography, as catalysts and antimicrobial agents as well as daily life products (cosmetics, plastics, clothes etc.) because of its strong antimicrobial activity [4, 5]. Therefore, these nanoproducts introduce into waste stream and they will inevitable disposed of in solid waste landfills at the end of their useful life [1], [4-10]. Landfilling is still the dominant technology in solid waste management. Landfills are typically provided with layers of nearly impermeable material to prevent contamination of surronding soil, surface and groundwater by leachate generated from landfills [11]. Leachate from solid-waste landfills contains a large number of organic, inorganic and hazardous compounds including nanoparticles, heavy metals [12], halogenated compounds and pesticides [13,14]. Sorensen et al., (2015) stated that emerging contaminants in urban groundwater include engineered nano-materials [15]. Landfills must be designed in a way to prevent contamination of the surrounding environment. Hence, impermeability layer such as geomembrane and geosynthetic clay liner should be placed at the base of landfill and over the waste top lift, respectively [16].

In leachate contaminant transport through landfill clay liners the following problems can be mentioned: (1) advective and dispersive transport of contaminants through defects in the geomembrane and clay liner [14] (2) diffusive transport of organic contaminants through non-defective composite liners [17, 18].

Sanitary landfilling is commonly used as a final disposal method for municipal solid waste in developing countries due to the commercial and technological advantages [19, 20]. However, landfill aeration is performed as a new method in the stabilization of municipal solid waste by researchers because of the increasing environmental problems such as water, soil and air pollution and its advantages such as faster degradation of waste, enhancing leachate quality and decreasing gas emission [21-23]. In addition, when landfill is operated as bioreactor, solid waste stabilization can be accelerated and controlled and optimizated in terms of the addition leachate and other liquid. Many studies used aerobic landfill bioreactors as lab, pilot and field scale in literature have been recorded in recently [20-25].

The object of the work was to investigate of the effects of TiO_2 and Ag nanoparticles on migration behavior of landfill leachate contaminants through landfill clay liner in aerobic bioreactor landfills. During 375 days of storage in aerobic bioreactor landfills, the variations of pH, Cl^- (chloride), conductivity, alkalinity, chemical oxygen demand (COD), total organic carbon (TOC), total nitrogen (TN) are investigated.

2. MATERIALS AND METHODS

2.1. Feedstock and Nanoparticles

Municipal solid waste (MSW) in aerobic bioreactor landfills were obtained from the Odayeri Sanitary Landfill to represent the properties of solid wastes in İstanbul, Turkey. Each reactors were loaded with about 900 kg municipal solid waste including 62% organic, 12% paper, 2% glass, 1% metals, 8% plastic, 3% textile, and 12% ash and others [26]. TiO_2 and Ag nanoparticles were purchased from Sigma-Aldrich (product code 637254 for TiO_2 nanoparticles at anatase and 576832 for Ag nanoparticles) form <25 nm and <100 nm particle size, respectively. Nanoparticles were added to each reactor, except for the control reactor, at a ratio of 100 mg/kg wet weight during the filling operation of the reactors. The operational conditions, amount of added solid waste and nanoparticles to reactors are summarized in Table 1. The minimum, mean and maximum concentrations of contaminants of leachate generated from AC, AT and AA reactors were given in Table 2.

2.2. Reactor Setup and Operation

Three pilot-scale aerobic bioreactor landfills (AC, AT and AA) were simultaneously operated for a period of 375 days. MSW feedstock were fed into the bioreactors.

A detailed schematic of the experimental set-up is depicted in Figure 1. The inner diameter of the reactors was 80 cm. The reactors were comprised of two parts with heights of 300 cm and 50 cm. The upper part was the main landfill reactor and the bottom part was consisted in order to model the landfill liner and the groundwater system. The landfill liner system was consisted of two compacted clay layers (10 + 10 cm) situated under a 0.2 cm high density polyethylene (HDPE) geomembrane. The clay material and geomembrane used in this study were obtained from Komurcuoda Sanitary Landfill in Istanbul, Turkey. The total area of geomembrane defects was about 1.0% of the surface area of the reactor.

Distilled water simulating groundwater was placed (80 L) at the bottom of the reactors. A drainage layer and a perforated leachate collection pipe were used for leachate collection at the bottom of the upper part of landfill reactors. The reactors were also equipped with leachate discharge valves, leachate recirculation system and aeration system, and landfill gas collection pipes. Temperature probes were located at 60 cm and 120 cm depths from the top of each reactor in order to measure temperature variations. Air was introduced from the bottom of the solid waste and passed through an upward direction by the help of the perforated aeration pipes O_2, CO_2 and CH_4 contents of the generated gas was measured in order to control whether there is enough O_2 for aerobic degradation. Experimental studies were conducted at room temperature (20-25 °C).

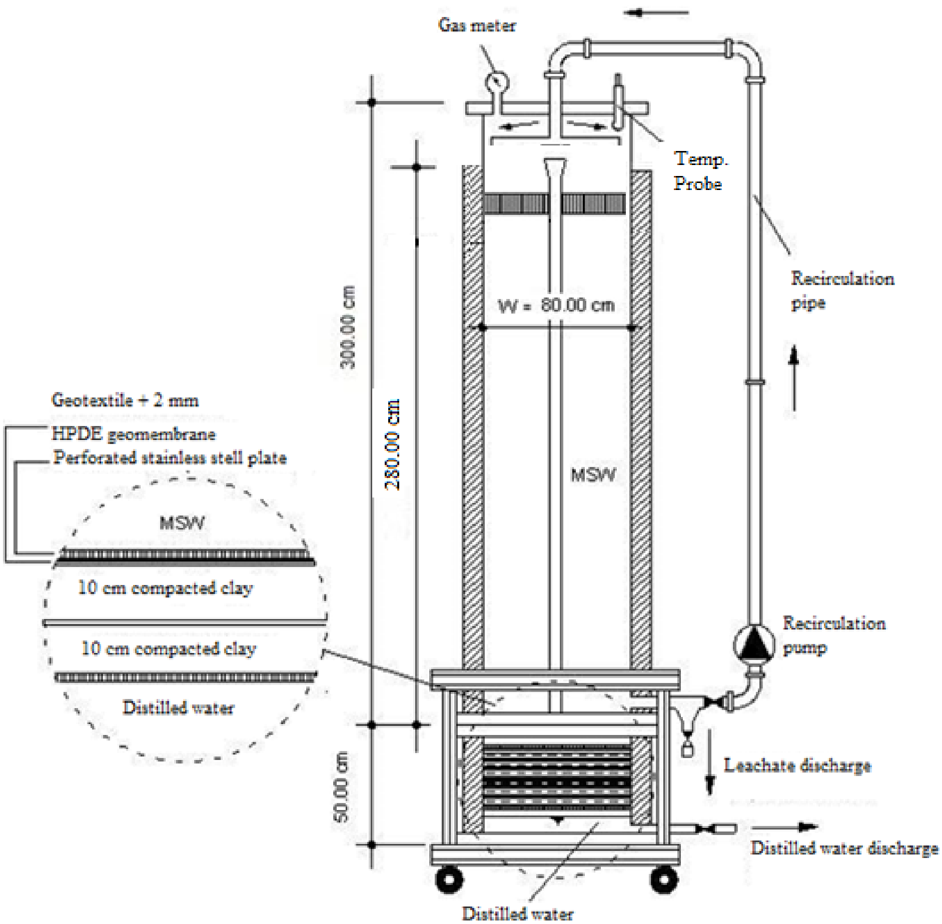

Figure 1. A detailed schematic of the experimental set-up.

Table 1. The operational conditions of reactors and the amount of added MSW, TiO_2 and Ag nanoparticles

Reactors	Operation Conditions	The amount of disposed solid waste (kg)	The amount of added nanoparticles (g)
AC	aerobic control bioreactor	892.610	-
AT	aerobic bioreactor operated with TiO_2 nanoparticles addition	817.990	81.8
AA	aerobic bioreactor operated with Ag nanoparticles addition	844.630	84.4

Table 2. The characterization of leachate generated from AC, AT and AA reactors

Parameters	Reactors								
	AC			AT			AA		
	min	mean	max	min	mean	max	min	mean	max
pH	4.78±0.07	6.85±0.04	7.73±0.05	7.05±0.03	7.65±0.05	8.29±0.06	6.77±0.05	7.67±0.07	8.02±0.03
Alkalinity (mg/L)	9000±101	17850±125	32100±153	2292±43	6171±95	13200±35	5170±75	8911±92	15400±40
Cl^- (mg/L)	2769±50	4028±55	6822±65	2834±45	3442±47	4004±52	3000±75	3587±55	4567±65
Conductivity (mS/cm)	19.6±0.2	30.3±0.4	35.4±0.3	11.7±0.1	17±0.15	23.6±0.3	12.7±0.2	18.4±0.1	26.4±0.2
COD (mg/L)	5000±35	59054±250	104000±500	1281±25	4954±32	13000±27	1764±22	8051±40	27500±35
TOC (mg/L)	2750±20	22101±65	35000±100	1000±28	2214±30	5000±52	1280±18	3276±25	7800±55
TKN (mg/L)	437±55	993±85	2073±100	117±28	667±50	1341±72	468±65	977±83	1720±66

2.3. Analytical Procedure

The leachate samples were collected at 1 week intervals and groundwater samples were collected (1 L) once a month during about 375 days. Leachate and groundwater samples were characterized for pH, alkalinity, conductivity, Cl^-, chemical oxgyen demand (COD) following Standart Methods for Examination of Water and Wastewater [27]. Total Organic Carbon (TOC) and Total Kjeldahl Nitrogen (TKN) contents of leachate and Total Nitrogen (TN) contents of groundwater samples were determined based on the thermal oxidation method at high temperature by using Hachlange, IL 550 TOC-TN model apparatus. Also, gas analyses (CH_4, CO_2 and O_2) were carried out using a GeoTech GA2000 Plus Model gas-measuring device.

The analyses of conventional parameters (pH, alkalinity, Cl^-, conductivity) in leachate samples started on day 7 and contiuned an average weekly period until the end of the experimental study. In addition, analyses of compounds in model groundwater samples were carried out on a monthly basis. Moreover, migration of leachate contaminants (particularly Cl^-, organic contents) through landfill clay liner to the model groundwater was considered as a measure of contaminant transport percentage as follows:

$$\text{Transport percentage (\%)} = \left[1 - \frac{(C_L)_m - (C_{MG})_m}{(C_L)_m}\right] \times 100 \quad (1)$$

where $(C_L)_m$ and $(C_{MG})_m$ are the mean concentrations of contaminants in the leachate and model groundwater determined during landfilling period.

Groundwater values reported are from about day 90 to day 375 as the migration of contaminants from landfill leachate to the model groundwater. Because groundwater measured more than 90 days ago is similar to distilled water.

3. RESULTS AND DISCUSSION

3.1. The Variations of pH and alkalinity

The variations of pH and alkalinity values in AC, AT and AA reactors were given in Figure 2a and 2b, respectively. pH values of groundwater samples collected from AC, AT and AA reactors were in the range of 4.3 – 5.3, 4.9 – 5.6 and 5.7 – 5.8, respectively. There was no significant change in pH values of groundwater samples of reactors during 375 days of operation in the reactor systems. However, pH values of groundwater samples collected from AA reactor were higher than samples collected from AC and AT reactors. Yang et al., (2013) [28] reported similar results in landfill leachate treated with Ag nanoparticles in their lab-scale study indicating the effects of Ag nanoparticles on methanogenic activities during anaerobic digestion of solid waste. The results show a slight decrease continuing with an increase in the rest of the study. Thus, the obtained results are in accordance with the results of the literature studies.This can be explained by the effect of Ag nanoparticles existing in AA reactor. Because Ag nanoparticles can facilitate to migration of contaminants in clay liner systems. There are very few studies subjected to the effects of Ag nanoparticles on pH of an aquatic environment in literature. Besides, it was mostly focused on on the effect of pH on the aggregation characteristics of nanoparticles, Al-Badawi, et al., (2010) [29] reported the occurrence of Ag nanoparticles aggregation and settling under acidic pH and/or high ionic strength.

As seen in Figure 2b, the alkalinity in AC and AT reactors remained low (30 mg $CaCO_3$/L) during experimental period. The alkalinity in reactor AA increased from 123 mg $CaCO_3$/L to 268 mg $CaCO_3$/L on day 200, then decreased to 170 mg $CaCO_3$/L on day 200 and remained stable until the end of the experimental period. Based on the experimental findings, transport percentage of alkalinity from landfill leachate to model groundwater was determined to be about 0.0712%, 0.33%, 1.915% in AC, AT and AA reactors, respectively. Literature studies still did not explain the influence of Ag nanoparticles on alkalinity. According to the obtained results, the alkalinity of the gorundwater samples taken from AA reactor seems higher than the other two reactors, but the alkalinity values are quite low when compared with leachate alkalinity values (around 200 mg/L for AA, and around 20 mg/L for AC and AT reactors). Thus, the transport mechanism of alaklinity can only be explained by the transportation of organic and inorganic pollutants some of which can cause an increase in alkalinity values.

3.2. The Variations in Cl^- Concentration and Conductivity

Figure 2c and 2d shows variations of electrical conductivity and Cl^- values in AC, AT and AA reactors, respectively. The electrical conductivity and Cl^- values of the first sample (about 100 days of operation time) collected from AA reactor were about 400 µS/cm and 50 mg/L, respectively. As seen from Figure 2c and 2d, maximum conductivity and Cl^- values of groundwater collected from AA reactor were around 775 µS/cm and 100 mg/L, respectively. As for AC and AT reactors, the average electrical conductivity and Cl^- values of groundwater samples collected were remained stable at about 40 µS/cm and 5 mg/L over 375 days, respectively. The electrical conductivity and Cl^- values of groundwater samples of AA reactor were about 20 fold higher than those at other reactors. Also, transport percentages of Cl^- were 0.122%, 0.141% and 2.385% in AC, AT and AA reactors, respectively. It can be concluded that this increase can be explained by the migration of leachate through the liner systems to groundwater in AA reactor.

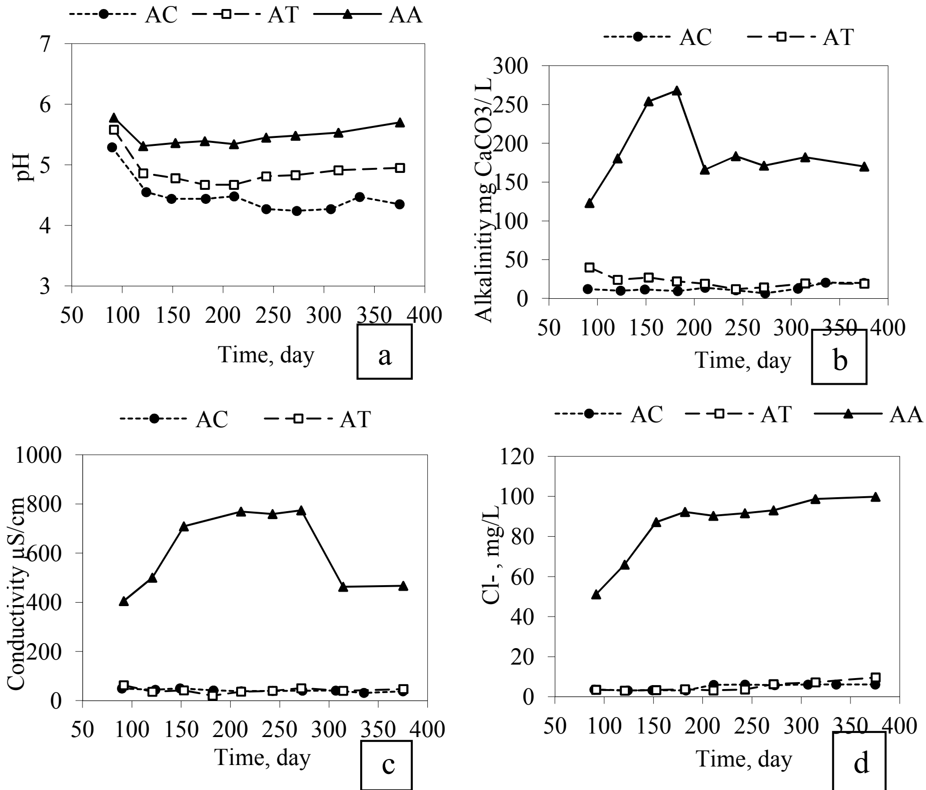

Figure 2. Variations of pH (a), alkalinity (b), conductivity (c) and Cl^- (d) values of ground water during experimental study

3.3 The Variations of Organic Contents

Figure 3a and 3b shows that the maximum COD and TOC concentrations of groundwater samples of AC reactor were about 40 and 13 mg/L, respectively. Increase in COD and TOC concentrations of groundwater samples were observed in AT and AA reactors until days 200. The maximum COD concentrations of groundwater samples of AT and AA reactors were 430 and 770 mg/L, respectively. Similarly, the maximum TOC concentrations of groundwater samples of AT and AA reactors were about 120 and 220 mg/L, respectively. COD and TOC concentrations of groundwater samples of AT and AA reactors were higher compared with AC reactor. It can be concluded that TiO_2 and Ag nanoparticles can be effective on migration of organic components of leachate to groundwater in aerobic bioreactor landfills. Fortner et al., 2012 [30] described that clay minerals are related to the fate and transport processes of nanoparticles along with their interactions with low permeability barriers for solid waste landfill.

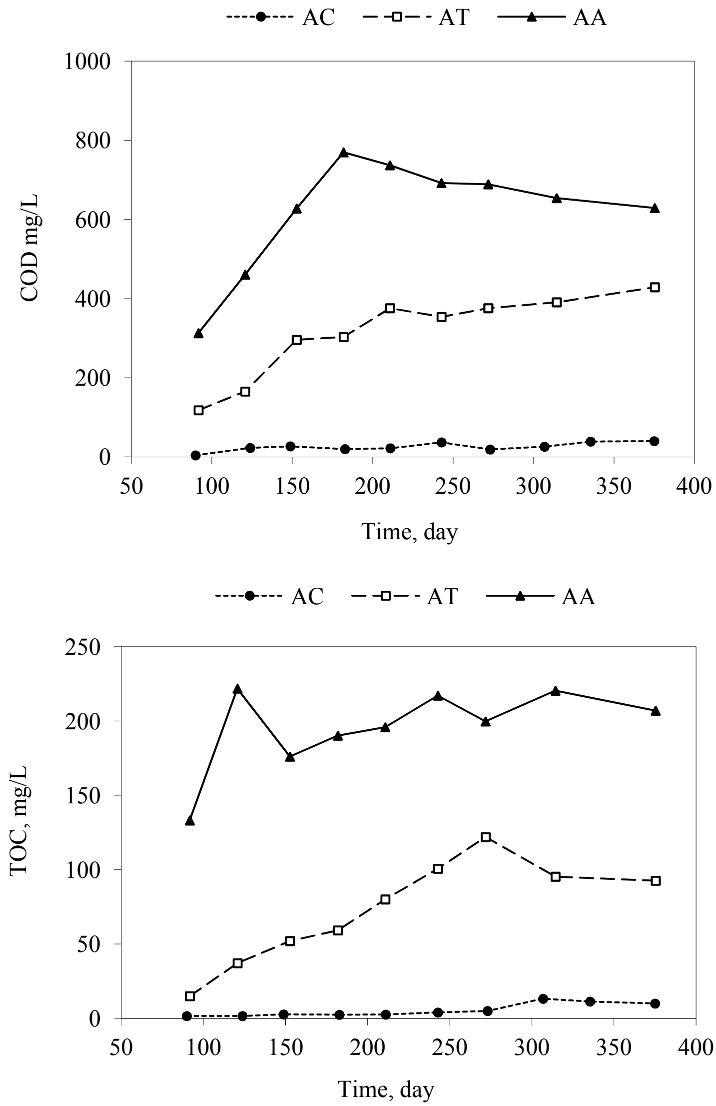

Figure 3. Variations of COD (a) and TOC (b) concentrations of groundwater during experimental study.

3.4. The Variations of TN

Many studies are reported that nitrogen removal rapidly occure during aerobic waste stabilization [24, 31-34]. Hence, it is expected that the nitrogen content of groundwater also decrease depending on decreasing of nitrogen content of leachate during aerobic stabilization. The average TN concentrations of the groundwater samples collected from AC, AT and AA reactors and transport ratios of TN in the reactors were around 1, 1.5, 2 mg/L (Figure 4.) and 0.1, 0.2 and 0.2%, respectively. As seen in Figure 4, TN values in reactors was no a sigfinicant change. It can be concluded that there has no a impact of TiO_2 and Ag nanoparticles on nitrogen migration in clay liner of aerobic bioreactor landfill.

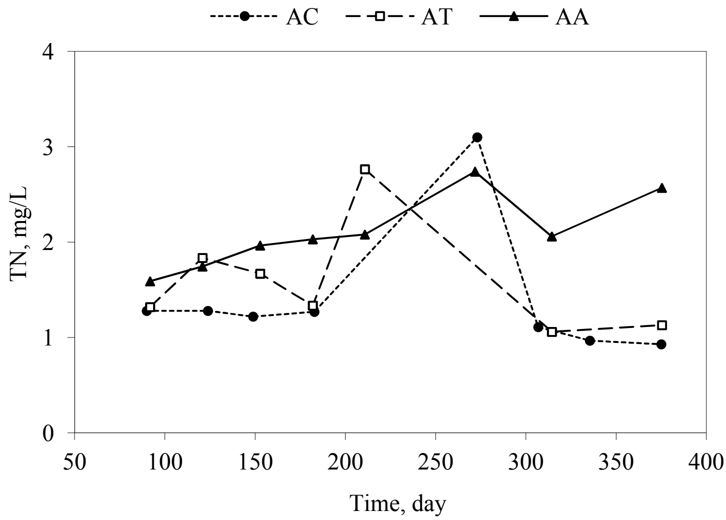

Figure 4. Variations of TN concentrations of groundwater during experimental study

4. CONCLUSIONS

The effects of TiO_2 and Ag nanoparticles on migration behavior of contaminants from leachate to groundwater were investigated in aerobic reactors including TiO_2 and Ag nanoparticles. The values of pH, alkalinity, Cl^-, conductivity and COD parameters in groundwater of reactor including Ag nanoparticles have been higher than the other reactors. Therefore, it can be inferred that Ag nanoparticles can increase the transport ratio of leachate contaminants from leachate to groundwater in aerobic bioreactor landfills. In addition, only COD value in groundwater of reactor including TiO_2 nanoparticles have been higher than control reactor and so it can be concluded that TiO_2 nanoparticles can increase the transport ratio of organic content of leachate from leachate to groundwater in aerobic bioreactor landfills. Although leachate contaminants can reach to the groundwater in trace concentrations, potential risks of these contaminants cannot be ignored due to their adverse effects on non-target organisms in the ecosystem. Therefore, more strict environmental strategies supported by legislations and regulations are needed for nanowaste management system and a resilient control of the migration of various leachate contaminants.

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