Jin-Yong Lee Jeong-Yong Cheon Hyung-Pyo Kwon Hee-Sung Yoon Seong-Sun Lee Jong-Ho Kim Joung-Ku Park Chang-Gyun Kim

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J.-Y. Lee  $(\boxtimes) \cdot$  J.-Y. Cheon  $\cdot$  H.-P. Kwon GeoGreen21 Co. Ltd., 4th Floor, SEK Building, 1687-22, Pongchon6-dong, Gwanak-gu, Seoul 151812, South Korea E-mail: hydrolee@empal.com Tel.: +82-2-8759491 Fax: +82-2-8759498

H.-S. Yoon · S.-S. Lee School of Earth and Environmental Sciences, Seoul National University, Seoul 151747, South Korea

J.-H. Kim · J.-K. Park Department of Soil and Groundwater, Environmental Management Corporation, Incheon 404170, South Korea

C.-G. Kim

Department of Environmental and Civil Engineering, Inha University, Incheon 402751, South Korea

Abstract Attenuation characteristics of landfill leachate were examined for two uncontrolled landfills in Korea. The two landfills containing municipal wastes without appropriate bottom liner and leachate treatment system have different landfill age, waste volume, and most importantly different hydrogeologic settings. One landfill (Cheonan landfill) is situated in an open flat area while the other (Wonju landfill) is located in a valley. Variations of various parameters including dissolved organic carbon (DOC), dissolved oxygen (DO), alkalinity, pH, electrical conductivity (EC), redox potential (ORP), ammonia (NH<sub>3</sub>), nitrate  $(NO_3^-)$ , sulfate  $(SO_4^{2-})$ , and chloride (Cl<sup>-</sup>) were examined along groundwater flow path. All these parameters were analyzed every month for a year. In the interior of the landfills, typical anaerobic conditions revealed by low DO and NO<sub>3</sub> concentrations, negative ORP values, high NH<sub>3</sub>, alkalinity, and Cl<sup>-</sup> concentrations were observed. Generally, higher levels of contaminants (DOC, NH<sub>3</sub>, and Cl<sup>-</sup>) were detected in the dry season while they were greatly lowered in the wet season. Significantly, large decrease of Cl<sup>-</sup> concentration in the wet season indicates that the dilution or mixing is one of dominant attenuation mechanisms of leachate. But detailed variation behaviors in the two land-

fills are different and they were largely dependent on permeability of surface and subsurface layers. The intermediately permeable surface of the landfills receives part of direct rainfall infiltration but most rainwater is lost to fast runoff. The practically impermeable surface of clayey silt (paddy field) at immediately adjacent to the Cheonan landfill boundary prevented direct rainwater infiltration and hence redox condition of the ground waters were largely affected by that of the upper landfill and the less permeable materials beneath the paddy fields prohibited dispersion of the landfill leachate into down gradient area. In the Wonju landfill, there are three different permeability divisions, the landfill region, the sandy open field and the paddy field. Roles of the landfill and paddy regions are very similar to those at the Cheonan. The very permeable sandy field receiving a large amount of rainwater infiltration plays a key role in controlling redox condition of the down gradient area and contaminant migration. This paper reports details of the attenuation and redox conditions of the landfill leachates at the two uncontrolled landfills.

Keywords Uncontrolled landfill · Leachate · Redox condition · Attenuation · Dilution · Permeability

# Attenuation of landfill leachate at two uncontrolled landfills

## Introduction

A great number of uncontrolled landfills without appropriate bottom liners and leachate collection systems have been wide spread in Korea. Both limits of environmental budgets and shortages of administrative measures have enforced them to be left abandoned or untreated. However, as the economy of the country enhanced, a public grievance has arisen concerning groundwater contamination by leachates emanating from the uncontrolled landfills. According to data of the Ministry of Environment of Korea (MOE), there exist more than 1,170 landfills with varying sizes and ages throughout the country at which land filling was completed (MOE 2003), and only 232 landfills (19.8%) have some treatment facilities for leachates on site or the leachates are transported to other places for special treatment (Lee et al. 2006, submitted). They were mostly formed before 1995. But landfills recently constructed have reliable bottom liners and leachate collection/ treatment systems that prevent migration of leachate plume into the surrounding ecosystems (aquifers). Few environmental incidences are reported from these landfills. Consequently, interests of relevant environmental authorities have been focused on the relatively older uncontrolled landfills.

Leachate composition and levels of contaminants of concern vary much with landfill age, waste composition, and hydrogeologic conditions of the landfills (Qasim and Chiang 1994). Heavy metals (Cd, Cu, As, Pb, and  $Cr^{6+}$ ), ammonium, and nitrate  $(NO_3)$  from the uncontrolled landfills pose a major environmental threat to human health in the country. Especially in the landfills mainly containing municipal wastes (not industrial origin), high levels of ammonium and  $NO_3^-$  are most problematic. Chronic drinking of NO<sub>3</sub> contaminated groundwater can cause methemoglobinemia (blue baby disease) in infants (Bouwer 1990; Knobeloch et al. 2000; Majumdar 2003), and high level of ammonium is toxic to many organisms and contributes to eutrophication and acidification (Lind and Nordh 2004). Under reduced (anaerobic) conditions that developed within the landfills, ammonium is dominantly abundant rather than  $NO_3^-$  and its concentration can remain significantly high for a very long period of time (Hutchings et al. 1998; Pivato and Raga 2006).

Leachate plumes may be attenuated by natural mechanisms such as dilution, dispersion, sorption, and biodegradation (Christensen et al. 1994; Bjerg et al. 1995). With the help of the above processes, levels of contaminants of concern may be lower than those that would be simply expected. Ammonium attenuation in soils and groundwater is predominantly due to cationic exchange and/or nitrification (biological oxidation) processes (Buss et al. 2004). NO<sub>3</sub><sup>-</sup>, chloride (Cl<sup>-</sup>), hard-

ness, and sulfate  $(SO_4^{2-})$  in the leachate are not attenuated by soil but are attenuated only by dilution (Bagchi 1990; Qasim and Chiang 1994). A natural low permeability hydrogeologic layer (clayey silt or clay materials) can provide an effective hydraulic barrier for the leachate plume migration.

This study focused on natural attenuation of the leachate contaminants at two uncontrolled landfills which have some different landfill conditions such as landfill size, landfill age, and most importantly hydrogeological settings. For this, monthly field investigations (January–December of 2005) and subsequent field measurement and laboratory analysis of relevant physical/chemical parameters were performed. The results are described in this paper.

#### **Materials and methods**

### Site characteristics

The two landfills of this study are located in Cheonan and Wonju, about 100 km east and south from Seoul, respectively (see inset map in Fig. 1). The Cheonan landfill is located on the ground surface in a relatively flat landscape surrounded by paddy fields (Fig. 1a). The landfill depth is about 4 m and the cover soil thickness is 1-3 m. The landfill covers approximately 6,000 m<sup>2</sup> and contains about 41,000 m<sup>3</sup> of municipal wastes (Table 1). The landfill received wastes in the period 1991–1998. Neither bottom liners nor leachate collection systems exist. Average annual precipitation of the area for past 10 years (1995-2004) is 1,332 mm. Over 60% of total precipitation occurs in the wet season (June-August) (Lee and Lee 2000). A total of 12 monitoring wells were newly installed at an average depth of 6 m (Fig. 1b). Full length was screened except for first 1–3 m below ground surface.

The Wonju landfill is located in a valley surrounded by relatively steep mountains with small pine trees (Fig. 2a). The landfill depth is approximately 50 m and the thickness of the cover soil is only 1.5 m. The landfill received waste in the period 1982-1995. The landfill covers 81,000 m<sup>2</sup> and contains 322,000 m<sup>3</sup> of household wastes (see Table 1). No bottom liners exist for the landfill while there is a concrete barrier wall at lower boundary of the landfill and a leachate storage system (settlement of suspended solids) for seepage water from the barrier wall (Fig. 2b). However, the leachate settlement system has not been operated and was abandoned. Lower part of the landfill was left as barren fields with some woods and plants. The mean annual precipitation of the area is 1,353 mm. Below the landfill, there is a sandy open field in which some kinds of vegetables have been partly grown. In more down gradient area, rice

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**Fig. 1** Location of the Cheonan landfill showing also layout of the monitoring wells and transect A–A' along mean groundwater flow path. Inset map shows locations of Cheonan and Wonju cities

fields and vegetable patches exist in which some groundwater use wells are found. A total of 14 monitoring wells were newly installed at depths of 3–6 m in this landfill area (see Fig. 2b).

In the Cheonan landfill, hydrogeologic layers vary much with location. In upper area of the landfill (at which household wastes are partially found), the first 3 m is sandy fill soil and under the layer, there exists sandy red soil (Fig. 3a). In central part of the landfill, wastes layer mostly exist below first 1–3 m of the fill soil. In paddy fields, first 2 m are soils composed of clayey silt. The fine-grained materials are for water

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Landfills	Cheonan landfill	Wonju landfill	
Location	Cheonan city	Wonju city	
Period of landfilling	1991–December 1998	May 1982–August 1995	
Landfill type	General landfilling	Valley landfilling	
Landfilling volume	$41,000 \text{ m}^3$	$322,000 \text{ m}^3$	
Waste type	Household wastes	Household wastes	
Landfilling area	$6,000 \text{ m}^2$	$81,000 \text{ m}^2$	
Landfilling depth	~4 m	$\sim 50 \text{ m}$	
Thickness of cover soil	1 m	1.5 m	
Landfill land use	Bare or barren land	Partly public sports facility	

Fig. 2 Location of the Wonju landfill showing also layout of the monitoring wells and transect B–B' along mean groundwater flow path



conservation of rice cultivation. Below the silt layer, sandy soils are encountered. Weathered layers of gneiss occur at depths below 4 m. The water levels are located 1–4 m below ground surface. The annual water level fluctuation was on average 1.23 m. The general direction of groundwater flow is southwesterly. In the landfill, the water levels were rather high and groundwater flow diverges from the landfill. The monitoring wells, CW1–CW3 (in and upper area of the landfill; screened interval = 3–6 m depth), showed large hydraulic conductivity with an order of  $10^{-2}$  cm/s while the monitoring wells, CW4–CW7 (down gradient of the landfill; screened interval = 2–6 m depth), had small values of hydraulic conductivity in order of  $10^{-5}$  cm/s. The large variance of the hydraulic conductivity indicates that the subsurface is heterogeneously formed in

the area; that is, lateral extension of each hydrogeologic layer is limited.

The subsurface cross-section in the Wonju landfill is shown in Fig. 3b. In the immediate bottom of the barrier wall (WW1 and WW2), the subsurface is very irregular. Sandy soil or boulder layer is found at first 1– 2 m depth. Below the depth, weathered layer or fresh granite is encountered. Downward from the monitoring wells, WW3, the hydrogeologic stratification is very simple. Three main layers including sandy soil, weathered rock, and fresh granite were found even though thickness of each layer varies. From WW3 to WW8, the sandy soil layer thickens and bottom of the layer generally becomes deeper. Furthermore, depths at which the weathered and fresh granites are being encountered are larger. Groundwater flow occurs

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**Fig. 3** Simplified hydrogeological sections of the two landfills (**a** Cheonan, **b** Wonju). The figure is adopted from Lee et al. (2006, submitted)



mainly in the sandy soil and weathered rock layers. In the area below the sandy open field (paddy fields), clayey silt layer exists to depths of first 1-2 m. Below the silt layer, sandy soil is encountered to a depth of about 7 m.

Generally, groundwater flows toward the residential area and the stream (north-west direction), but the water level contours are semicircular, indicating converging flow because the landfill and the study area are located in the valley surrounded by mountain and hills (see Fig. 2b). The water levels are located 1-2 m below ground surface except for WW0 (4-6 m below ground surface). The annual water level fluctuation is 0.3-1 m. Cracks with noticeable width are found at some parts of the concrete barrier wall. The monitoring wells, WW5 and WW6, showed large hydraulic conductivities with orders of  $10^{-3}$  and  $10^{-2}$  cm/s while the other monitoring wells had smaller values of hydraulic conductivity in order of  $10^{-4}$  cm/s. The hydraulic conductivity around the monitoring wells, WW12 and WW13, is in order of  $10^{-5}$  cm/s.

#### Groundwater monitoring

Groundwater samplings for field measurement and laboratory analysis were regularly conducted every month. A volume of groundwater was obtained by a low rate peristaltic pump. Prior to sampling, pH, dissolved oxygen (DO), water temperature, redox potential (ORP), and electrical conductivity (EC) were measured using a multi-probe device (YSI 556). Redox sensitive parameters including dissolved iron (Fe<sup>2+</sup>), manganese  $(Mn^{2+})$ , sulfide  $(S^{2-})$ , and ammonia  $(NH_3)$  were determined in the field using a spectrophotometer (DR2010, HACH, Pittsburgh, PA, USA). The groundwater samples were then analyzed in the laboratory for  $Cl^{-}$ ,  $NO_{3}^{-}$ , nitrite,  $SO_4^{2-}$ , alkalinity, and dissolved organic carbon (DOC). Samples for laboratory analysis of anions were collected in 60 ml HDPE bottles through a 0.45 µm syringe filter. Water samples of 125 ml were also collected for laboratory analysis of alkalinity. Water samples for DOC analysis were directly collected from the undisturbed water stream in 40 ml glass amber vials. All

samples were preserved at 4°C immediately after sampling. Ionic constituents ( $NO_3^-$ ,  $NO_2^-$ ,  $SO_4^{--}$ , and  $CI^-$ ) were analyzed by IC (DX–120, DIONEX, CA, USA). The DOCs for filtered samples was analyzed by TC Analyzer (Phoenix8000, Dohrmann, CO, USA). Alkalinity was determined by potentiometric titration Gran plots for graphical determination of the end point (Stumm and Morgan 1996; Drever 1997).

# **Results and discussion**

#### Water levels and water temperatures

The water levels in the two landfills correspond well to rainfall (Fig. 4). The water levels were high in the wet season (June–September) while they were low in the dry season (December–March). The effect of large amount of precipitation on the water levels in the wet season lasted for additional 3–4 months (up to December). The water level fluctuation of each well was restricted within 1.5 m at both landfills. Groundwater temperatures of shallow wells (high water tables) are mostly dependent on ambient air temperatures (solar radiation). In this country, the air temperatures are the highest in summer (July–August) while they are the lowest in winter (December–January; Lee and Hahn 2006). Mean groundwater temperatures were 17–17.5°C, and their maximums were 27–29°C (Table 2). Generally, ground-

**Fig. 4** Groundwater levels and monthly precipitation for 2001 (**a** Cheonan, **b** Wonju)

water temperatures were high within the landfill but they were relatively low outside of the landfill. The highest groundwater temperatures resulted mainly from high ambient air temperatures in the summer and the fermentation heat during decomposition of organic wastes. At both landfills, the minimum temperatures occurred in December while the maximum temperatures were recorded in July–August, which corresponds well to the air temperatures.

Dissolved oxygen and electrical conductivity

The DO in the Cheonan landfill ranged from 0.2 to 5.6 mg/l. Depletion of the DO (<1 mg/l) mostly occurred within the landfill although a slight increase was observed in the wet season (Fig. 5). The low DO concentration was due to decomposition of the organic wastes in the landfill (Lyngkilde and Christensen 1992). A large increase of DO concentration was observed down gradient from the well CW4, which is immediately outside of the landfill lower boundary. Generally, DO concentrations of the down gradient wells were sustained above 2 mg/l throughout the year. In the Wonju landfill, DO concentrations showed a gradual simple increase trend along the flow path (with distance from the landfill). Although there are no monitoring wells within the landfill, the leachate escaping from the concrete barrier indicated DO depletion (< 1 mg/l). The



Table 2 Summary of groundwater temperatures (unit: °C)

Location	Mean	Median	Maximum	Minimum	SD	Range
Cheonan	17.5	16.6	29.0	5.8	6.4	23.2
Wonju	17.0	16.4	27.0	3.2	6.6	23.8

increase of DO level in the wet season was also detected in this landfill.

The EC is one of the important indicators for groundwater contamination by landfill leachate (Fig. 6). As expected, the ground waters showed very high EC values within the landfill (especially for CW1 well), which ranged from 1,729 to 13,040  $\mu$ s/cm (average = 7,621  $\mu$ s/cm). The values largely decreased in CW2 and CW3 wells in the order of 2,000  $\mu$ s/cm. The ECs again largely dropped at the down gradient well, CW4, to the order of 400  $\mu$ s/cm. This level was maintained in the down gradient wells. In consideration of background EC levels (250–300  $\mu$ s/cm) above the landfill, the ground waters at the down gradient wells appeared least affected by the landfill leachate. Meanwhile, a large drop of ECs was observed in the wet season at both in and out of the landfill. In the Wonju landfill, a



Fig. 5 Dissolved oxygen (DO) of ground waters in the two landfills



Fig. 6 Electrical conductivities of ground waters in the two landfills

gradual decrease of EC levels was detected with distance from the landfill. At immediate down gradient of the landfill (WW2), EC levels ranged from 1,102 to 4,010  $\mu$ s/ cm (average = 2,874  $\mu$ s/cm). From WW3 to WW6, the EC levels were similar and they largely dropped at WW8. At a distance of 158 m from the landfill (WW13), EC values were the most similar to background levels. At this point, the effect of the landfill leachate appeared minimal.

#### pH and oxidation-reduction potential

The pHs of the ground waters at the two landfills were slightly acidic or near neutral (Table 3). Leachates of the landfills were most acidic and the mean pHs of ground waters were 6.2 and 6.4 for Cheonan and Wonju landfills, respectively. In the wet season, variation of the pHs among the monitoring wells was greatest (Lee et al.

Table 3 Values of pH measured at two uncontrolled landfills

Location	Mean	Median	Maximum	Minimum	SD	Range
Cheonan	6.2	6.3	7.3	5.0	0.5	2.3
Wonju	6.4	6.4	7.0	5.1	0.4	1.9

2006, submitted). But within the monitoring scale of the areas, any significant increase of the pHs was not detected with distance from the landfills.

The ORP is a useful indicator representing redox condition of ground waters (Fig. 7). Within the Cheonan landfill, the ORP values were always negative (-8 to -150 mV) throughout the monitoring year, which indicated anaerobic condition of the landfill. Outside of the landfill, the ORP levels drastically increased up to 328 mV. This increase matches well with higher values of DO concentrations in the down gradient.

Most interestingly, somewhat contrary to expectation, ORP levels were generally lowered in the wet season although they increased slightly at some wells within the landfill. Addition of rainfall with more oxic condition to the existing ground waters would elevate the ORP levels (Lee et al. 2001a) but the reverse was observed in the down gradient wells. The decrease of the ORP levels may be derived from outer expansion of the anaerobic condition of the landfill interior by accelerated groundwater flow in the wet season. The newly recharged waters in the wet season would push out the existing anaerobic ground waters into the outside of the landfill. In the Wonju landfill, variation of the ORP levels was generally continuous without large increase or decrease with distance. Also within the landfill, the ORPs were negative (inferred from the seeped leachate from the concrete barrier). In outside of the landfill, the ORP levels were generally high (50–350 mV; see Fig. 7). In some measurements in the wet season, similar phenomena (lowering of ORP levels in the wet season) were observed at the down gradient wells. But this decrease of the ORP in the wet season was confined to immediate adjacent wells to the landfill.

Dissolved organic carbon and alkalinity

The DOC indicates dissolved organic matter in ground waters, which can be used for substrates of biological degradation, and it may also influence the behavior of many other contaminants including toxic heavy metals (Christensen et al. 1996; Drever 1997). Within the Cheonan landfill, values of DOC were varying highly from 6.9 to 618.1 mg/l (Fig. 8). The highest values occurred in the dry season while the lowest were in the wet season. The drastic decrease of DOC in the wet season was due to mixing of a large amount of infiltrated rainwater with





Fig. 7 Variation of redox potential (*ORP*) along transect in the two landfills

Fig. 8 Variation of dissolved organic carbon (*DOC*) along mean groundwater flow path (each transect) of each landfill

Cheonan

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existing ground waters. Outside of the landfill, DOC levels largely decreased to below 10 mg/l, which was similar to background level. In the Wonju landfill, concentrations of DOC ranged between 15.4 and 266.1 mg/l at the immediate down gradient well (WW2) of the landfill. A large drop of DOC level also occurred in the wet season. In this landfill, decrease of DOC with distance was rather monotonous. Relatively high concentration of DOC was maintained with distance.

Alkalinity would be increased with decomposition of organic matter by microbes in the landfill (Qasim and Chiang 1994). In the interior of the Cheonan landfill, levels of alkalinity were very high (average 1,333.2 mg/l as  $CaCO_3$ ) and they decreased along flow path to about 60 mg/l (Fig. 9). Excess of the alkalinity relative to calcium (Ca) is likely to be derived from the biodegradation of organic matter (Borden et al. 1995; Basberg et al. 1998; Lee et al. 2001b). The alkalinity values in the Wonju landfill ranged between 46.0 and 1,425.0 mg/l. As expected, the nearest well (WW2) to the landfill showed the highest values of alkalinity (average 793.2 mg/l). Large decrease in the wet season occurred only in the immediate adjacent wells to the landfill. Considering the distances to the landfills, high alkalinity was sustained at a longer distance in the Wonju landfill.

### Ammonia and nitrate

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The NH<sub>3</sub> is the most abundant form of nitrogen in the landfill under reduced condition (Hutchings et al. 1998). But in its form, migration is relatively limited. In oxic condition, it would be readily converted to  $NO_3^-$ , which is much mobile. Both forms of nitrogen are the main environmental concerns in landfill areas in Korea (Chung and Lee 1998; Kim et al. 2001). In the interior of the Cheonan landfill, the ammonia-nitrogen  $(NH_3-N)$ was greatly enriched up to 860.0 mg/l (Fig. 10), which is 1,720 times greater than the Korean drinking water standard (0.5 mg/l NH<sub>3</sub>–N). But these values drastically decreased outside of the landfill and they were all within the standard (< 0.5 mg/l). Within a distance of 10 m, NH<sub>3</sub>-N concentration dropped to below the drinking water standard. In the Wonju landfill, immediately down gradient well, WW2 showed NH<sub>3</sub>-N concentrations of 18.7-191.0 mg/l. Nevertheless, of a large increase of NH<sub>3</sub>-N at WW4 (~62 m from the landfill), level of NH<sub>3</sub>-N generally decreased along flow path. Compared with NH<sub>3</sub>–N levels of the Cheonan, they were relatively high even at substantial distances (75-115 m), which were greater than the drinking water standard.





Perimeter of the landfil

Fig. 9 Variation of alkalinity along mean groundwater flow path

Fig. 10 Variation of NH<sub>3</sub>-N along mean groundwater flow path

In the interior of the Cheonan landfill,  $NO_3^-$  was mostly depleted in the dry season ( $< 1 \text{ mg/l } NO_3^--N$ ) while a large increase was detected in heavy rain period (Fig. 11). Consumption of  $NO_3^-$  as electron acceptor for biodegradation would lower the  $NO_3^-$  level (Lyngkilde and Christensen 1992; Lee et al. 2001b). Further or additional degradation of organic matter using replenished  $NO_3^-$  also resulted in lowered levels of  $NO_3^--N$  in some down gradient wells, CW2–CW3. The lowered  $NO_3^$ concentration was not largely recovered even at outside of the landfill except for the increase in the wet season. All the values of  $NO_3^-$  within and outside of the landfill were below the Korean drinking water standard (10 mg/l  $NO_3^--N$ ) except for the single measurement for CW1.

In the Wonju landfill, a very different behavior of  $NO_3^-$  was observed. At WW2 (immediately down gradient of the landfill),  $NO_3^-$  concentrations were lowered in the dry season (<3 mg/l  $NO_3^-$ -N). However, they drastically increased up to 219.7 mg/l in the down gradient well, WW4 (~62 m from the landfill), which is 21 times greater than the drinking water standard. The largest increase of  $NO_3^-$  levels occurred in the rainy season. The large increase of  $NO_3^-$  concentration appeared not to be attributed to nitrification alone of the NH<sub>3</sub> derived from the upper area under aerobic condition (see Figs. 5, 7) because levels of  $NH_3^-N$  (more re-



Fig. 11 Variation of NO<sub>3</sub>-N along mean groundwater flow path

duced form of nitrogen) were mostly also elevated at this point. Low activity of denitrification microbes would not produce additional reduced forms of nitrogen at the point (Kim et al. 2001), which is also supported by lowered alkalinity (see Fig. 9). Consequently, other sources of both nitrogen forms ( $NH_3$  and  $NO_3^-$ ) were probably expected. The possible sources of additional nitrogen were production of NH<sub>3</sub> and subsequently NO<sub>3</sub> by biological decay of surface plants such as soybeans, corns, and sesame (Galloway 1998) or migration of nitrogen from surrounding pine forests (Gundersen et al. 1998). The high concentrations of  $NO_3^-$  largely decreased again with distance from this point. The lowered levels of  $NO_3^-$  were very similar to those at immediate down gradient well (WW2) of the landfill. Meanwhile, an intermediate form of nitrogen, NO<sub>3</sub>, NO<sub>2</sub>, was mostly below 1 mg/l.

#### Sulfate and chloride

In the dry season,  $SO_4^{2-}$  concentrations within the Cheonan landfill were nearly depleted (Fig. 12), which were the results of anaerobic biodegradation of organic matter. But in the wet season,  $SO_4^{2-}$  concentrations were



Fig. 12 Variation of  $SO_4^{2-}$  concentration along mean groundwater flow path

highly elevated. Outside of the landfill, the lowered concentrations gradually increased with distance. But these values were much lower than the Korean drinking water standard (200 mg/l). High level of  $SO_4^{2-}$  and low concentration of  $S^{2-}$  indicated no  $SO_4^{2-}$  reduction in the down gradient wells. In the Wonju landfill, variation of  $SO_4^{2-}$  concentration was very simple, a general increase with distance. At this site,  $S^{2-}$  was only detected below 0.6 mg/l.

The Cl<sup>-</sup> concentration may represent the extent of leachate contamination and also indicate mixing or dilution effect by newly infiltrated rainwater because the Cl<sup>-</sup> is most conservative (Qasim and Chiang 1994). The Cl<sup>-</sup> concentrations were greatly dependent on the season (more accurately on precipitation amount). In the dry season, the Cl<sup>-</sup> concentrations were very high with the Cheonan landfill (CW1) and they were above 1,000 mg/l(Fig. 13). These high values drastically decreased to about 20 mg/l or less in the wet season. In down gradient wells, decrease of Cl<sup>-</sup> concentrations was also great although the decreasing ratios were lessened. Hence, dilution by heavy rainfall would make a significant contribution in leachate attenuation or in subsequent evolution or biochemical reaction of existing ground waters with replenishment of new ground waters with additional electron acceptors. The lessened



Fig. 13 Variation of Cl<sup>-</sup> concentration along mean groundwater flow path

decreasing ratios of Cl<sup>-</sup> concentration in the down gradient wells may indicate that the mixing effect is derived from upper area (actually impermeable clay silt layer covers the surface of the down gradient area, preventing direct rainfall infiltration; see discussion in next section). Meanwhile, except for the CW1 well, all the monitoring wells showed Cl<sup>-</sup> concentrations within the drinking water standard (250 mg/l).

In the Wonju landfill, Cl<sup>-</sup> concentrations ranged from 127.2 to 410.5 mg/l at the least down gradient well, WW2. The concentrations gradually decreased with distance. At about 80 m distance from the landfill, Cl<sup>-</sup> concentrations effectively decreased below the standard value (250 mg/l) in the dry season. In the wet season, a large decrease of Cl<sup>-</sup> concentration was also detected in this landfill. But the enlarged decreasing ratio (or mixing ratio) of Cl<sup>-</sup> concentration may reflect direct rainfall infiltration through permeable upper surface soil layer or additional recharge from surrounding area as well as new ground waters from up gradient area. Meanwhile, substantial increase of Cl<sup>-</sup> at 62 m distance (WW4) indicated additional source of Cl<sup>-</sup>. The unexpected increase of Cl<sup>-</sup> concentration at this monitoring well was coincident with the abnormaly high concentrations of  $NO_3^-$  and  $NH_3$ –N. But the origin of the additional Cl<sup>-</sup> is not known at the moment.

#### Conceptual redox setting

In the above, spatial and temporal variations of various parameters were described and discussed. In this section, based on the hydrogeologic setting of the landfills and the observed variation behaviors of the parameters, a simple but most feasible conceptual schematic is suggested to explain best the observed phenomena (Fig. 14). But all the explanations are not necessarily satisfactory.

In the Cheonan landfill, there exist two distinctive hydrogeologic divisions, which are the main landfill region and the down gradient paddy fields. Surface of the upper landfill is intermediately permeable or relatively less permeable (Fig. 14a). When rainfall occurs (especially in the wet season) most of the rainwater is lost to runoff but a small portion infiltrates into the subsurface of the landfill. In the dry season, anaerobic condition would be dominantly prevailing. But in the wet season, aerobic condition forms temporarily but it rapidly dissipates to the down gradient area. The surface of the paddy field is clayey silt. It is hard for rainwater to infiltrate directly into this layer. So, the temporal redox condition is largely affected by that of the upper area (landfill). The less permeable subsurface materials retard migration of some contaminants of concern ( $NH_3$ ,  $NO_3^-$ , and Cl<sup>-</sup>) into the down gradient area.

In contrast, the Wonju landfill area has three distinctive divisions including the main landfill region, the



Fig. 14 Simple conceptual schematics illustrating variation of some relevant parameters along groundwater flow path

sandy open field, and the paddy field. The redox condition of the landfill interior is not different from that of the Cheonan landfill. However, the open sandy field played a significant role in controlling the redox condition in this area. The very permeable surface layer facilitated direct infiltration of rainwater into subsurface in the wet season. The infiltrated rainwater would elevate levels of DO, ORP, and  $NO_3^-$ . The highly aerobic condition may affect the redox condition of more down gradient area (the paddy field). The highly permeable subsurface also caused rapid groundwater flow (J. Y. Lee et al. 2006, submitted) and maintained high levels of contaminants (NH<sub>3</sub>, NO<sub>3</sub>, and Cl<sup>-</sup>) even at some substantial distances.

#### **Summary and conclusion**

In this study, attenuation characteristics of the leachate at two uncontrolled landfills were evaluated along groundwater flow path. The two landfills are much different with respect to age, volume, and hydrogeologic condition but they contain same kind of municipal wastes. The Cheonan landfill is situated in flat open area while the Wonju landfill is located in the valley. Based on the hydrogeologic investigation and regular monitoring of groundwater quality, the following main conclusions were drawn.

In the interior of the landfills, typical anaerobic condition prevailed, accompanied with highest DOC, NH<sub>3</sub>, and Cl<sup>-</sup> concentrations. Generally, levels of contaminants of concern were high in the dry season while they were low in the wet season. Drastic decrease of Cl<sup>-</sup> concentration in the wet season indicates that dilution and mixing are one of the major mechanisms of leachate attenuation.

The impermeable surface layer of the down gradient paddy field in the Cheonan landfill prevented direct rainfall infiltration. Hence, the redox condition was greatly affected by that of the up gradient landfill area. Furthermore, the less permeable subsurface of the paddy field restricted migration of contaminants along the groundwater flow path.

The permeable sandy open field located between the Wonju landfill and the paddy field played an important role in controlling redox condition and contaminant migration below the open field. A large amount of rainwater infiltration through the permeable surface layer caused a great dilution and mixing of leachate but sustained high levels of  $NO_3^-$  by supplying additional oxygen. The rapid groundwater flow also maintained high levels of contaminants throughout the open field.

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#### References

- Bagchi A (1990) Design, construction and monitoring of sanitary landfills. Wiley, New York
- Basberg L, Banks D, Sæther OM (1998) Redox processes in groundwater impacted by landfill leachate. Aquat Geochem 4:253–272
- Bjerg PL, Rügge K, Pedersen JK, Christensen TH (1995) Distribution of redoxsensitive groundwater quality parameters down gradient of a landfill (Grindsted, Denmark). Environ Sci Technol 29:1387–1394
- Borden RC, Gomez CA, Becker MT (1995) Geochemical indicators of intrinsic bioremediation. Ground Water 33:180– 189
- Bouwer H (1990) Agricultural chemicals and groundwater quality. J Soil Water Conserv 45:184–189
- Buss SR, Herbert AW, Morgan P, Thornton SF, Smith JWN (2004) A review of ammonium attenuation in soil and groundwater. Q J Eng Geol Hydrogeol 37:347–359

- Christensen JB, Jensen DL, Christensen TH (1996) Effect of dissolved organic carbon on the mobility of cadmium, nickel and zinc in leachate polluted groundwater. Water Res 30:3037–3049
- Christensen TH, Kjeldsen P, Albrechtsen H-J, Nielsen PH, Bjerg PL, Holm PE (1994) Attenuation of landfill leachate pollutants in aquifers. Crit Rev Environ Sci Technol 24:119–202
- Chung SY, Lee KK (1998) Groundwater environment of the Okmyong waste landfill in the Pohang city. J Korean Soc Groundwater Environ (in Korean) 5:223–232
- Drever JI (1997) The geochemistry of natural waters: Surface and groundwater environments. Prentice Hall, Upper Saddle River, NJ
- Galloway JN (1998) The global nitrogen cycle: changes and consequences. Environ Pollut 102:15–24
- Gundersen P, Callesen I, De Vries W (1998) Nitrate leaching in forest ecosystems is related to forest floor C/N ratios. Environ Pollut 102:403–408

- Hutchings HH, Hammel JE, Osiensky JL (1998) Nitrogen leaching from unlined cull onion landfills. J Environ Qual 27:1254–1260
- Kim DW, Park SW, Lee JY, Lee PK (2001) A study on the extent of the pollution of an illegal landfill. J KoSSGE (in Korean) 6:31–38
- Knobeloch L, Salna B, Hogan A, Postle J, Anderson H (2000) Blue babies and nitrate-contaminated well water. Environ Health Perspect 108:675–678
- Lee JY, Lee KK (2000) Use of hydrologic time series data for identification of recharge mechanism in a fractured bedrock aquifer system. J Hydrol 229:190–201
- Lee JY, Cheon JY, Lee KK, Lee SY, Lee MH (2001a) Factors affecting the distribution of hydrocarbon contaminants and hydrogeochemical parameters in a shallow sand aquifer. J Contam Hydrol 50:139–158
- Lee JY, Cheon JY, Lee KK, Lee SY, Lee MH (2001b) Statistical evaluation of geochemical parameter distribution in a ground water system contaminated with petroleum hydrocarbons. J Environ Qual 35:1548–1563
- Lee JY, Hahn JS (2006) Characterization of groundwater temperature obtained from the Korean national groundwater monitoring stations: implications for heat pumps. J Hydrol (in press)

- Lind S, Nordh M (2004) Ammonium removal from landfill leachate by use of ashes. MS Thesis, KTH-Royal Institute of Technology, Stockholm, Sweden
- Lyngkilde J, Christensen TH (1992) Fate of organic contaminants in the redox zones of a landfill leachate pollution plume (Vejen, Denmark). J Contam Hydrol 10:291–307
- Majumdar D (2003) The blue baby syndrome: nitrate poisoning in humans. Resonance 8:20–30
- MOE (Ministry of Environment, Korea) (2003) Status report on policy of environment conservation. MOE, Seoul, Korea
- Pivato A, Raga R (2006) Tests for the evaluation of ammonium attenuation in MSW landfill leachate by adsorption into bentonite in a landfill liner. Waste Manag 26:123–132
- Qasim SR, Chiang W (1994) Sanitary landfill leachate. CRC Press, Boca Raton, FL
- Stumm W, Morgan JJ (1996) Aquatic chemistry, 3rd edn. Wiley, New York