HYDROGEOCHEMICAL PERSPECTIVE TO ASSESS GROUNDWATER CONTAMINATION WITHIN THE AQUIFER BELOW THE LANDFILL SITE

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ABSTRACT

An investigation was carried out at Astrolabe Park Landfill, an unlined landfill located in the Botany Sands Aquifer, Southern Sydney. The primary objective of this research is to assess the vertical and horizontal trends of chemical constituent in the leachate plume within the groundwater. Two bundled-piezometers along flowpath were sampled to achieve changes downgradient the landfill. Data analysis has demonstrated that the centre of the leachate plume is located at the shallow depth between 3 to 6 meters below the ground surface. Five hydrogeochemical processes are well confirmed to affect the concentration of the chemical constituents within the leachate plume such as dissolution-precipitation, reduction, ion-exchange, mixing as well as adsorption processes are active in the contaminated groundwater. Moreover, the occurrence of organic clay/peat layer is responsible to decrease most of the chemical constituent at some depths. Along the flowpath, some internal and external factors change the chemical composition downgradient the landfill such as dilution and dispersion, grass watering, the occurrence of continuous layer of pyrite-bearing and nitrogen-based fertilizers on the golf course around the landfill site.

Keywords: groundwater, contamination, landfill, hydrogeochemical processes

INTRODUCTION

In urban area, where the space for disposal waste is limited, landfill could be located in the same aquifer as that being used for groundwater pumping to provide drinking supplies. Consequently, rainfall will infiltrate through the landfill and creates leachate plume, which will move away from the site and contaminates groundwater. This investigation examines the changes of chemical constituents within the groundwater below the landfill site to describe the vertical and horizontal distribution of the leachate plume as well as to identify the hydrogeochemical processes responsible to the changes of chemical composition downgradient the landfill.

LANDFILL PROCESSES

Landfills are used to dispose of a large quantity of material, coming from municipal repudiate such as household waste, industrial waste containing toxic organic substances, as well as plant material and building wreckage Zanoni (1972). Landfill wastes will react with water coming from rainfall infiltration, aquifer, and any liquids already present in the landfill including groundwater in saturated zone to produce what is known as leachate. Landfill processes and reactions occur in a physical environment that may be saturated, partly saturated, or even unsaturated where the interstices filled by gasses such as oxygen, carbon dioxide, nitrogen, and methane or vaporized organics. Table 1 illustrates the main processes within the landfill.

Saturation phase Stages present Biochemical condition
Unsaturated Solids, gases Aerobic, oxidizing
Partly saturated Solids, liquids, gases Aerobic, oxidizing
Saturated Solid, liquids, dissolved gases Anaerobic, reducing

Table 1. Saturation stages, phases, and landfill processes

The extent and direction of the leachate plume is characterized primarily by hydrogeological conditions such as porosity, hydraulic conductivity, geology of the aquifer, hydraulic gradient (slope) as well as hydrogeochemical processes and leachate composition itself (Roberts, 1996).

SITE DESCRIPTION

The research has been carried out at Astrolabe Park Landfill, southern Sydney occupies and area of approximately 4.8 hectares. This park is bounded by residential area to the northeast, and by several golf courses and sport facilities, and the fairways to the west, south, and east. Also, in very close proximity to the west, Astrolabe Park margin of the Lachland Ponds System is located. Figure 1 illustrates the park situation as well as sampling position.

Lee (1994) pointed out that the water table within the landfill site remained practically unvarying on a seasonal basis in contrast to adjacent areas and attributed this to the occurrence of a perched aquifer. Besides that, Robert investigation (1996) measured the groundwater level from 18 piezometers within the site reported that the water table ranging from 0.90 meter to 4.96 meter below the ground surface. Generally, local groundwater flow direction is from west towards the ponds systems. Next, Jankowski and Knight (1991) indicated that the regional flow direction is southwest towards Ponds No. 5 and 6 under a hydraulic gradient of 0.005. Also, they concluded that shallow groundwater consequently discharges to the ponds system, while the deep groundwater associate to the regional groundwater system.

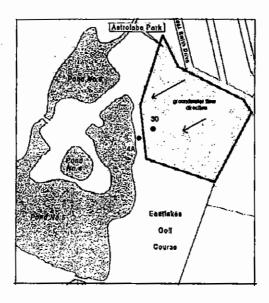


Figure 1. Sampling position

AQUIFER CONDITION

The geology condition of Astrolabe Park was approached by using data research conducted either at Astrolabe Park or its adjacent area such as UNSW David Phillips Fields. Investigation by Webb and Watson (1979) at David Phillips Fields defined a primarily fine to medium grained quartz sand containing two different layers of organic clay with the maximum thickness of 3 meters at the depth of approximately 7-10 meters and 17 meters, respectively, and overlying the Hawkesbury Sandstone bedrock material.

Jankowski and Knight (1991) classified three sub-aquifer system within the site and described as follows: (i) unconfined sandy aquifer, 3 meters depth to 7.7 m; (ii) leaky sandy aquifer, 7.7 m to 17 m; and (iii) semi-confined sandy aquifer, 17 to 30 m depth. The peaty clay layers which role as a confining layer contributes to form leaky aquifer at the depth of approximately 7 to 17 meters.

METHODOLOGY

The field sample collections were carried out at two multi-level piezometers along flowpath, which has codes of 30 and 4A. Spatially, piezometer 30 is located very close to the landfill, whereas the piezometer 4A is in close proximity to the Ponds No.5 being downgradient the landfill as illustrated in Figure 1.

In field, Geopump II peristaltic pump was used in case of groundwater sampling from multi-level piezometer. Next, the discharge water was initially diverted into 2 litre of beaker containing EC, Eh, pH, and DO probes. After few minutes, the reading values of these parameters will stabilize and sampling is possible to be commenced and recorded. Some major ion of groundwater can be also analysed directly in the field by using two *Hach-2010 Direct Reading Spectrophotometer* to determine iron-ferrous (Fe²⁺), sulfide (S²), nitrogen-ammonia (NH₄⁺), nitrate (NO³⁻) and phosphate (PO₄⁻)

Laboratory analysis was conducted in order to determine the concentration of major ion, total sulphur, and total ion using ICP-AES instrument. Besides that, ICP-MS was used to determine concentrations of some trace element and metal. Titration method was also conducted to determine the chloride concentration. Finally, every chemical constituent determination must be based on the principle that any water sample should be in a state of electrolytic neutrality, means that the sum of all cation species concentration must be equal to the sum of all anion species in terms of miliequivalent per litre (meq/l). Accordingly, charge balance errors (CBE) should be conducted to check the accuracy that should be less than 5%.

VERTICAL HYDROGEOCHEMICAL TREND

The analytical results derived from samples in two piezometers are used in the next section to exemplify the vertical trends and changes in the concentration of major ion, carbonate species, redox species, heavy metals as well as physical properties such as fluid EC, pH, and DO.

Groundwater Types, EC and TDS

Alekin (1970) introduced the classification of groundwater type according to the anion and cation composition, calculated in meg/l. In piezometer 30A where located downgradient to the landfill site, the contaminant plume is situated at the shallow depth (8.0 m below the ground surface) and most samples have groundwater type of HCO₃-Ca and HCO₃-Ca-Na. In contrast, groundwater types in piezometer 4A show a remarkable difference with depth indicating quite complicated chemical processes within the leachate plume. Groundwater type dominated by bicarbonate ion with some variation magnesium, sodium and calcium below the depth of 4 meters changes to groundwater dominated by bicarbonate and calcium until reaching the depth of 8 meters and groundwater domination in chloride, sulphate, and sodium ions after the depth of 8 meters. This variation in this bore shows contaminated groundwater on the top and fresh water below. To sum up, groundwater contamination is characterized by the presence of bicarbonate, calcium and sodium with the groundwater type of HCO₃-Ca and HCO₃-Ca-Na and depends on the ion capacity exchange of the geologic material and the concentration of these two cations. In addition, the present of peat layer at the depth of approximately 6 to 8 meters gives the impression to the rapid decreasing of some ions and determine the changes on groundwater type.

The high value of EC and TDS indicates the present of leachate plume. The value of EC and TDS in piezometer 30 reaches a peak at the depth of approximately 6 meters below the ground surface with the relatively high TDC/EC ratio (>0.85) and probably due to the result of high bicarbonate level within the leachate plume. Furthermore, relatively same condition present in piezometer 4A with the high ratio of TDS/EC corresponds to the occurrence of the leachate plume.

Carbonate System Parameters and Metals

Evaluating the values of pH, CO₂, and HCO₃ can assess the primary parameters related to the carbonate system at Astrolabe Park Landfill. Result of vertical distribution of carbonate parameters shows that the high concentration of these parameter are associated with the occurrence of the leachate plume, mostly at the shallow depth below 6 meters. However, concentration of carbon dioxide in piezometer 30 peaks at the depth of 12 meters below the ground surface. It can be concluded that the high concentration of CO₂, and HCO₃ is not only due to the landfill processes such as redox reaction and acid fermentation, but also as a result of dissolution processes within the aquifer matrix, which is active in the certain depth.

The presence of heavy metals at some parts of the aquifer can be explained by remobilisation of heavy metals in water column under anaerobic condition (Fetter, 1998). As the high value of TDS and negative value of Eh, heavy metals ca be remobilised from sediment to groundwater. The concentration of Strontium (Sr²⁺), Boron (B), Zinc (Zn²⁺), Manganese (Mn²⁺), Aluminium (Al²⁺), and Copper (Cu²⁺) clearly designated that the leachate plume occurs at the shallow depth below 7 meters below the ground surface and always related to the value of pH. Furthermore, the value of some ions reduced rapidly to be almost zero near the bedrock layer suggested that the groundwater is uncontaminated with respect to metals or mixed with regional groundwater.

Redox Couples

Groundwater contaminated usually dominated by strongly-reducing environment as a result of reduction processes. At Astrolabe Park landfill, some reduced species such as S²⁻, NH₄⁺, and Fe²⁺ are present in reasonably high concentration in groundwater. Vertical profiles of field Eh versus the redox couples SO₄²⁻/S²⁻, NO₃⁻/NH₄⁺, andn Fe³⁺/Fe²⁺ show that most of reduced species as well as Eh values concentrated at the shallow depth and peak at 7 meters below the ground surface. Also, some positive correlation of contaminant characteristics are well present in Piezometer 4A, confirms that the decreasing concentration of S² is always corresponding to the increasing concentration of sulphate.

NH₄⁺ peaks at a depth of approximately 5-7 meters below the ground surface and then drops rapidly as a result of NO₃⁻ reduction. Below the leachate plume, NO₃⁻ concentration increases and the concentration of NH₄⁺ decreases as the leachate mixes with fresh, oxygenated groundwater. Besides that, concentration of Fe²⁺ peaks at the shallow depth and may be confirmed that the iron within the plume is due to the leaching of disposal waste material especially in Piezometer 30. In Piezometer 4A, concentration

of Fe3⁺ is fairly high and possibly attributable to the dissolution-reduction of ferric oxides in the "Waterloo Rock" layer.

HYDROGEOCHEMICAL PROCESSES

To discuss hydrogeochemical processes active in this site, some methods was used to fully achieve the hydrochemical processes within the piezometer 30 and 4A. The first one is by using some modeling software such as NETPATH (Plummer et. al., 1991) and WATEQ-4F (Ball and Nordstrom, 1991) to calculate the saturation indices of some relevant minerals to determine their affinity for dissolution or precipitation processes. The second is to observe the concentration changes of some major ions with respect to a conservative element such as Cl to obtain the ion exchange processes. Bivariate plots of these ions to assess whether the ion exchange processes takes place will support this second method. The third method will examine the changes in redox condition including the stability fields for different redox couple such as Fe^{2+}/Fe^{3+} , NH_4^+/NO_3 , and S^2^-/SO_4^{2-} . In this project, the Eh value for redox couples is calculated using Nernst Equation and WATEQ4F (Ball and Nordstrom, 1991) modeling software.

The calculation of saturation indices gives an understanding that all samples are

Dissolution - Precipitation

still in undersaturated condition with respect to calcite and dolomite. Here, high concentration levels of calcium and bicarbonate (between 3 and 6 meter below ground surface) are most likely as a result of dissolution processes of shell debris present in silt and clayey sands layers of Botany Sands aguifer. Also, this process has an effect on pH values at these depths. Moreover, the combination of dissolution processes producing enhanced bicarbonate within the plume and combined with redox reactions acquires bicarbonate complicates of the top of the plume (Jankowski and Acworth, 1997). Most samples has already supersaturated with respect to siderite, goethite, and pyrite (especially at the depth above 6 meters below ground surface, in case of siderite). This condition implies that this groundwater can be categorized to be included in the central zone of the leachate plume. Besides that, another sources of major ions such as feldspars weathering can supply some elements, however, it seems that it only contributes by small amount since the Botany Sands aquifer is predominantly comprises by quartz sands. Figure 2 illustrates the relationship between saturation indices with respect to some minerals and Total Dissolved Salts for all samples. By looking at these figures, it gives the impression that the saturation indices increase with the increasing of TDS. Even, some samples are supersaturated with respect to siderite and goethite indicates that these samples are located in the centre of the plume. In short, the saturation indices greater than zero (with high TDS) suggested a tendency for precipitation processes to оссиг.

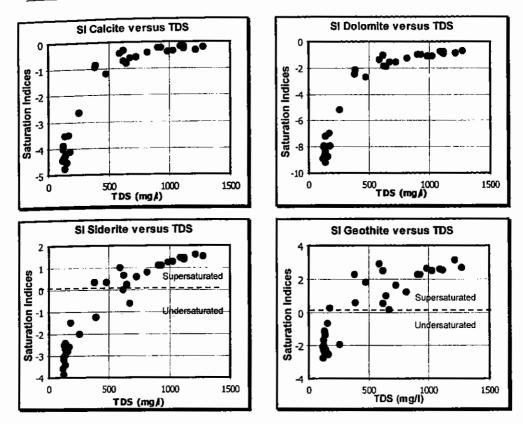


Figure 2.Saturation indices of some minerals versus TDS

Ion Exchange

It is clearly shown that the concentration of calcium in the two piezometers declines after the existence of clay layer at the depth of approximately 6 meters below the ground surface. However, another ions have not changed significantly by the presence this clay layer. On the other hand, another cations have very small increasing of their concentration after that depth. These phenomena can probably be explained by determining the ion exchange processes within those depths. The bivariate plot between Na⁺ + K⁺ and Cl⁻ in Figure 3 indicates that most of the groundwater sample from piezometer 4A and 30 places above the 1:1 line (note that rainwater is located in this line). It means that ion exchange process of Ca²⁺ for Na⁺ is responsible to this case. Accordingly, more Na⁺ is released to groundwater and the concentration of Ca²⁺ decreases. This situation is present within the clay layer between the depths of approximately 6 and 8 meters below the ground surface and just located slightly below the centre of the leachate plume.

However, the calcium ion still exist as a dominant ion within the groundwater as a result of dissolution processes of calcium carbonate (shell) material as discussed in

the previous section. Beside that, the increased concentration of potassium in the leachate plume can be explained by the same way as sodium but its concentration values are much lower than sodium. On the other hand, samples that lay below the 1:1 line are product of ion exchange process between Na⁺ and other cations, which decreases the Na⁺ concentration.

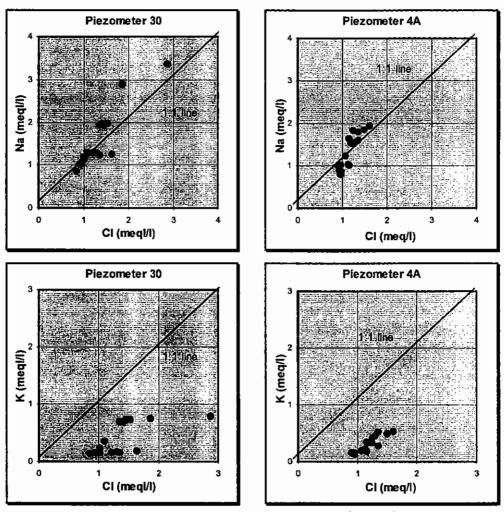


Figure 3. Bivariate plot of Cl versus Na and K

Furthermore, Figure 4 illustrates the ion exchange processes by plotting another cations such as magnesium and calcium against chloride. From these plot, it can be concluded that sample that has Mg:Cl ionic ratio less than 1 (located below the 1:1 line)

or Ca:Cl ratio is greater than 1 (above the 1:1 line) are by some degree the product of ion exchange processes within the clay layers. Moreover, the groundwater sample, which have excess of Na⁺ and Ca²⁺ ions with respect to Cl, usually accompanied by the decreasing concentration of Mg²⁺. So that, the increasing concentration of Na⁺ must be as a result of ion exchange process with Mg²⁺ since there are no significant sodium source within the aquifer matrix.

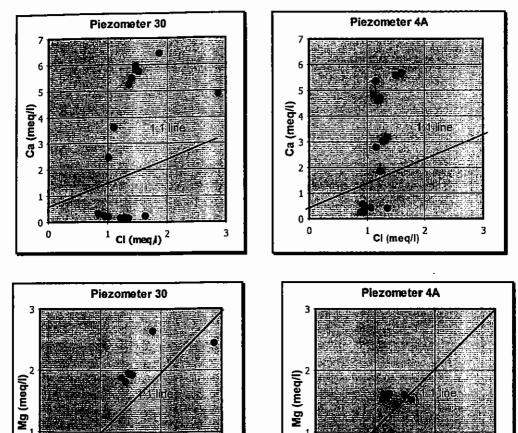


Figure 4. Bivariate plot of Cl⁻ versus Mg²⁺ and Cl⁻ versus Ca²⁺

1 CI (meq#)

1 CI (meq./) 2

Redox Processes

The redox condition within the leachate plume also can be assesses by looking at the vertical distribution of sulfide, iron (II), and nitrate. First of all, the concentration of SO4²⁻ peaks slightly above the depth of 6 to 7 meters and then decrease rapidly to get its minimum value at the depth of 11 meters below ground surface. However, after this depth as oxygen started to generate, the SO4²⁻ concentration goes up again. In opposition, the concentration of S²⁻ peaks at the depth of between 7 to 9 meters below ground surface. This condition implies the reducing environment within the plume. In short, the concentration of SO4²⁻ within the center of leachate plume is very low and above/below this centre, the concentration generates to background level. Next, the presence of NH₄⁺ in the leachate plume indicates that NO₃ has been reduced to produce NH₄⁺ in groundwater. From this reducing environment, significant concentration of NH₄⁺ has been produced and this implies the reduction processes to occur. Lastly, the distribution of Fe³⁺ within the plume indicates that the reducing environment occurs at the depth of approximately 3 to 6 meters below ground surface. In this depth, the Fe²⁺ concentration reaches its uppermost concentration.

Mixing and Adsorption

The variation of groundwater type found at piezometer 4A may imply the mixing between contaminated and deep groundwater from regional flow system. This fact is supported by vertical equipotential map within the site based on the field measurement as shown in Figure 5.

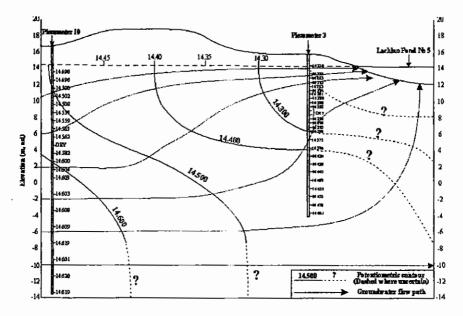


Figure 5. Vertical equipotential map in the vicinity of Pond No. 5

Adsorption process usually affects some trace element such as heavy metals. Ray and Chan (1986) revealed that organic materials might attenuate metal concentrations According to data in Piezometers 4A and 30, all metals concentration reduced rapidly after the occurrence of clay layer at the depth of approximately 6 meters below the ground surface. It can be concluded that probably the adsorption process presents within this layer and then significantly reduced the metals concentrations.

SPATIAL DISTRIBUTION ALONG THE FLOWPATH

The changing of some chemical constituents along the flowpath is to determine the contaminant transport migration characteristics and its spatial distributions. The change of HCO₃ and some redox couples (S²/SO₄² and NH₄⁺/NO₃) accompanied by some parameters such as Eh, pH, and EC are determined by comparing them at the start and end of the flowpath. This discussion is based in the cross section between piezometer 30 and 4A after the depths of the sampling position were adjusted to the real elevation above sea level.

HCO₃ and EC

Along the flowpath, the peak value of HCO_3 declined from 847 mg/l to 622 mg/l (approximately 27% decrease). Beside that, the elevation of the greatest concentration between two piezometers, about 50 m downgradient shows a significant change where the peak value was lowered by more than 4 meters. Next, EC has the similar pattern of change compare to that the HCO_3 . Maximum EC value also decreases from 1429 μ S/cm to 1060 μ S/cm, with the lowering of its peak value elevation by above 4 meters as shown in Figure 6.

Some factors may be accountable to the significant variation and spatial distribution between piezometers 30 and 4A. First of all, considering the location of the piezometer 30 that is to be found within the landfill and piezometer 4A that is positioned approximately 50 meters away downgradient the landfill, it can be predicted that the EC and HCO₃ values at piezometer 4A must be lower than piezometer 30. As a result, the EC and HCO₃ values decrease. Here, the dilution of the leachate plume from both vertical and horizontal dispersion occurs along the flowpath. Next, the human activity factor around the golf course, such as grass watering will result in the increasing of water infiltration, especially at Piezometer 4A. This factor as well as the difference density between fresh and contaminated water may be responsible to the lowering elevation of central mass of the leachate plume.

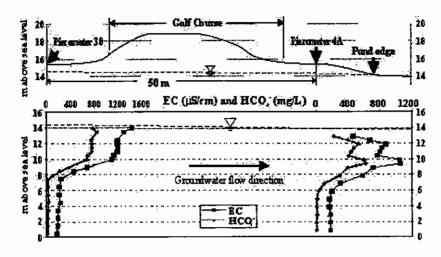


Figure 6. The spatial distribution of HCO₃ - EC along the flowpath

Eh and pH

Along the flowpath, the value of Eh decreases significantly, means that piezometer 4A has more reducing condition compare to piezometer 30 (Figure 7). Moreover, the Eh values at piezometer 30 reach their maximum values between the elevation of approximately 8.5 and 7.5 meter asl, while the Eh at Piezometer 4A peaks at the shallower elevation compare to that at piezometer 30. This condition might due to the variation of different rates of microbiological activities within the sediment along the flowpath. On the contrary, the pH values show an increasing trend along the flowpath, with some variation within their profiles. At piezometer 30, pH values peaks at the elevation of about 9 meter above sea level, and decrease sharply afterwards. Also, according to Figure 6.28, it seems that the pH values are strongly constrained between the value of 6.4 and 6.7. The similar condition occurs at piezometer 4A, where the pH values are still constrained by relatively similar range in the contaminated zone. Here, pH attains their highest values at the elevation of 10 meter asl and then goes down gradually until 4 meter asl.

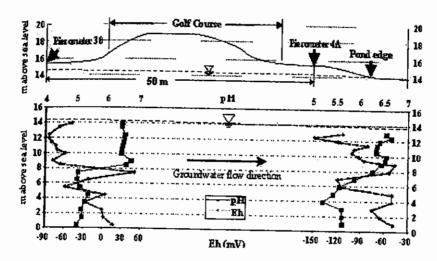


Figure 7. The spatial distribution of pH - Eh along the flowpath

S2- and SO42-

The concentration of S^2 of does not show a significant change along the flowpath, since most of the samples confirmed lower than 1 mg/l. However, the maximum value of S^2 is lowered at piezometer 4A by approximately 1-meter depth. Moreover, the peak value of S^2 is not located within the center of the leachate plume as defined by another parameters, but it observed immediately occurs below the plume.

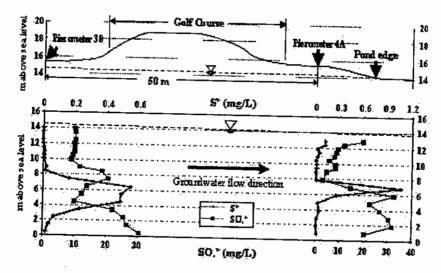


Figure 8. The spatial distribution of S2- and SO42- along the flowpath

NH4+ and NO3

The ideal landfill characteristic of redox reactions is well confirmed by NH₄⁺ and NO₃ concentrations. First of all, the concentration of NO₃ increases downgradient the landfill, whereas the concentration of NH₄⁺ goes down. Moreover, it also observed that NO₃ demonstrates a near absence of their concentration within the leachate plume at Piezometer 30, while NH₄⁺ has the highest concentration within the plume. Similarly, Piezometer 4A experiences the same trend as Piezometer 30. However, the maximum value of NH₄⁺ decreases along the flowpath by approximately 10 mg/l. Besides that, the zone of highest NH₄⁺ dropped by approximately 1 meter along the flowpath. Next, an interesting phenomenon occurs at Piezometer 4A, since the elevated concentration of NO₃ found at the shallow depth above the plume center. It probably comes about as a result of nitrogen-based fertilizers activities on the golf course.

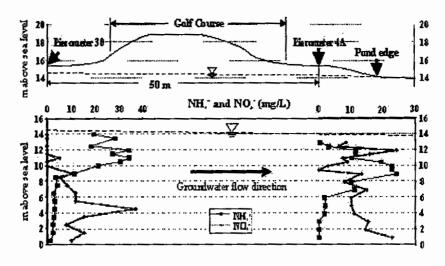


Figure 9. The spatial distribution of NH₄⁺ and NO₃ along the flowpath

CONCLUSION

The most dominant ion within the leachate plume is bicarbonate and calcium. In addition, the groundwater type of HCO₃-Ca is dominant in the plume centre between the depth of approximately 3 and 6 meter. Ion HCO₃ and Ca²⁺ are possibly derived from carbon dioxide by landfill processes as well as dissolution of shell material within the aquifer matrix. In general, groundwater type at Piezometers 30 and 4A doesn't show something remarkable different, except deep groundwater found at Piezometer 4A that

shows a complicated result implies the mixing between contaminated and deep groundwater in regional flow system.

Five hydrochemical processes are well confirmed to affect the concentration of the chemical constituents within the leachate plume. First of all, dissolution of waste material and carbonate material within the aquifer matrix most likely increase the major ion concentration as well as some minor elements. Also, the centre of the plume confirmed that negative saturation indices with respect to dolomite and calcite to occur, confirmed that the dissolution process exists in this zone. Moreover, positive saturation indices with respect to some minerals such as siderite, goethite and pyrite imply that precipitation process also presents in the leachate plume.

Along the flowpath, the peak value of HCO₃ declined significantly and the elevation of utmost concentration between two piezometers, about 50 m downgradient shows a considerable change where the peak value was lowered by more than 4 meters. Similarly, EC also experiences the same trend as compared to HCO₃ trend. In short, it can be assumed that dispersion and mixing with fresh regional groundwater perhaps responsible in this case. The concentration of SO₄² in the two piezometers demonstrates a decreasing trend within the leachate plume and elevated concentrations both below and above the plume by up to 30 mg/l. Also, it confirmed that the maximum value of sulfate increases by more than 10 mg/l along the flowpath. On the contrary, the highest concentrations of S² were observed immediately 1 meter below the plume centre. The concentration of NO₃ raises downgradient the landfill, whereas the concentration of NH₄⁺ goes down. Also, an elevated concentration of NO₃ is found at the shallow depth above the plume center at piezometer 4A, probably as a result of nitrogen-based fertilizers activities on the golf course.

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