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Is Landfill Leachate a Potential Source of Nitrogen for Plant Growth?

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Abstract. Landfill leachate contains a substantially high concentration of ammonical-nitrogen which could be regarded as an alternative nitrogen source for plants. However, leachate is typically saline and has elevated concentrations of various toxic pollutants, which are potential barriers in its use for soil irrigation. Retardation of growth as a result of phytotoxicity has been successfully avoided by applying diluted leachate, the concentration of which is obtained by bioassays using seed germination data. However, pollution risk and nitrogen saturation are still major concerns, which must be fully studied in the application of leachate for the rehabilitation of closed landfills.

Keywords: Landfill leachate, toxicity, irrigation, nitrogen, trees

1. Introduction

Landfilling is the most common method in dealing with municipal solid wastes worldwide, and leachate produced from landfills as a result of water percolating through or emerging from the buried waste is a major environmental concern. Landfill leachate is a very complex high-strength wastewater which contains suspended and dissolved materials removed from the decomposing waste in the landfill body. It consists of soluble organic and inorganic constituents. It is highly toxic and has detrimental effects on the environment.

Modern strategic landfills usually have greater filling capacity and longer life span, but larger landfills imply greater environmental hazards. Although most of them have impermeable bottom linings and associated devices to contain pollutants generated within the buried waste, and piping system for leachate to be collected and subsequently treated, on-site or off-site, the strength and volume of leachate are problematic. Information on the chemical properties and toxicity of leachates helps understand their chemical constituents, possible pollution impact, potential mitigation measures and effective regulatory means [1-9].

1.1. Chemical properties

Typically landfill leachate has slightly alkaline pH. Its conductivity and salinity are high, indicating elevated concentrations of ions and salts that are present. This is common as large amount of inorganic compounds from the buried wastes dissolved in the infiltrating water and attributed to the high ionic strength.

Landfill leachate is generally characterized by extremely high concentrations of biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), total and ammoniacal-nitrogen, anions (e.g. Cl^- and SO_4^{2-}), cations (e.g. Na^+ and K^+) and heavy metals (e.g. Cd, Cu and Zn). It also contains a variety of trace organics. Toxic organics such as halogenated aliphatics (e.g. chloroform and dichloromethane), aromatic hydrocarbons (e.g. benzene, toluene, ethylbenzene and xylene), phenolics and many others were found in various studies [4, 10, 11]. However, due to the highly complex nature of landfill leachate, it is difficult and impossible to identify all the organics present, especially those that are water soluble. It has been mentioned that as much as 90% of the organic matter in a leachate sample is very soluble and cannot be identified by most conventional analyses for specific organic compounds [12].

Chemical properties of leachate vary with factors such as climate, waste composition, landfill age and season. Tropical and subtropical landfills take shorter time to become methanogenic as reflected by the

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alkanility of the leachate of operating landfills [13]. Leachate from operating landfills has lower oxidation reduction potential (ORP) values because of the highly anaerobic conditions that prevail. Leachate strength largely declines with time, and fluctuates between seasons. The higher level of rainfall in summer will end up in stronger leachate as more pollutants are washed out, but there is the likelihood that the increase in quantity of pollutants generated is cancelled out by the dilution effect [9]. There is significant difference in the strength of leachates from landfills of different ages, particular between operating and postclosure landfills, with the pollutant concentrations decline quickly once the landfill operation stops. Even though the concentrations of most chemical constituents are relatively much lower in older sites, their values may still exceed the maximum discharge limit, and this applies particularly to total nitrogen allowed for effluent discharge to wastewater treatment facilities.

1.2. Ecotoxicity

Ecotoxicity studies on landfill leachate supplement chemical data in assessing the ecological impacts of the leachate. These provide a comprehensive picture to include the effects of all constituents present and the possible interactions between chemical species present in the complex wastewater. Selection of bioassays is important, and usually a battery of multi-species tests is used to include different trophic levels. Results obtained demonstrate the extremely high toxicity of landfill leachate to various organisms from different ecosystems and that leachate can be mutagenic or even carcinogenic research [14-17].

Different compounds present in leachate have been proposed to be the toxicant responsible for leachate toxicity. Ammoniacal-nitrogen and organic compounds were proposed to be the factors governing the toxicity of leachates [9, 18], though the toxicity of other landfill associated factors such as alkalinity, chloride and heavy metals are also suggested [19, 20]. However, as landfill leachate is so chemically complex, extensive chemical analyses are required to find out the major toxicant present. Moreover, evidence obtained is derived from statistical analyses on the data from chemical analyses and toxicity bioassays, which is not sufficiently confirmative. Toxicity identification evaluation (TIE) which is developed by the US EPA and adopts various manipulation and fractionation techniques in combination with toxicity test has been recently used. Graduated pH adjustment, C18 solid phase extraction, oxidant reduction, zeolite addition and EDTA chelation, alone or in combination, and mass balance method subsequently conducted confirm the toxicity of ammonia and non-polar organics in landfill leachate. Completion identification of all toxicants was nevertheless difficult due to the complexity of landfill leachate and the limitations of TIE.

Landfill leachate, with such an ample supply of ammonical-nitrogen, has been suggested to be an alternative source of nitrogen which is the most important and often limiting plant nutrient. Leachate application to soil would be a form of irrigation water especially in water deficit areas and dry seasons [20, 21]. However, different plant species respond differently to leachate irrigation. It is not known if the presence of other pollutants in leachate is a constraint to leachate irrigation for plant growth.

1.3. Phytotoxicity

The outcomes of leachate irrigation trial have been controversial [22-26]. This can be attributed to the high spatial and temporal variations in the composition of landfill leachate, complexity in composition, difficulties in predicting the toxicity of the individual constituents and consequently an arbitrary decision in the dilution factor and irrigation schedule. Germination of *Brassica chinensis* (Chinese white cabbage) and *Lolium perenne* (perennial ryegrass) seeds has been proven to be very good surrogate model for the rapid evaluation of phytotoxicity [27]. The dilution rate was determined based on the dose response relationship of the seeds. Choosing the correct leachate dilution level is essential to safeguard the plants from toxicity damages and ensure healthy plant growth [21, 27].

2. Leachate Application to Soil

2.1. Plant performance

Selecting the right plants for irrigation by leachate is as well important. Various tree species were evaluated using the leachate application rate determined by seed assay. None of the 19 species exhibited retarded growth after 90 days when compared with the water irrigated control (Figure 1) [28]. The increments in tree height, standing leaf number and basal diameter were higher in the leachate-irrigated seedlings. The robust seed germination bioassay has served as a reliable surrogated model for finding the most effective concentration (dilution) for leachate irrigation.

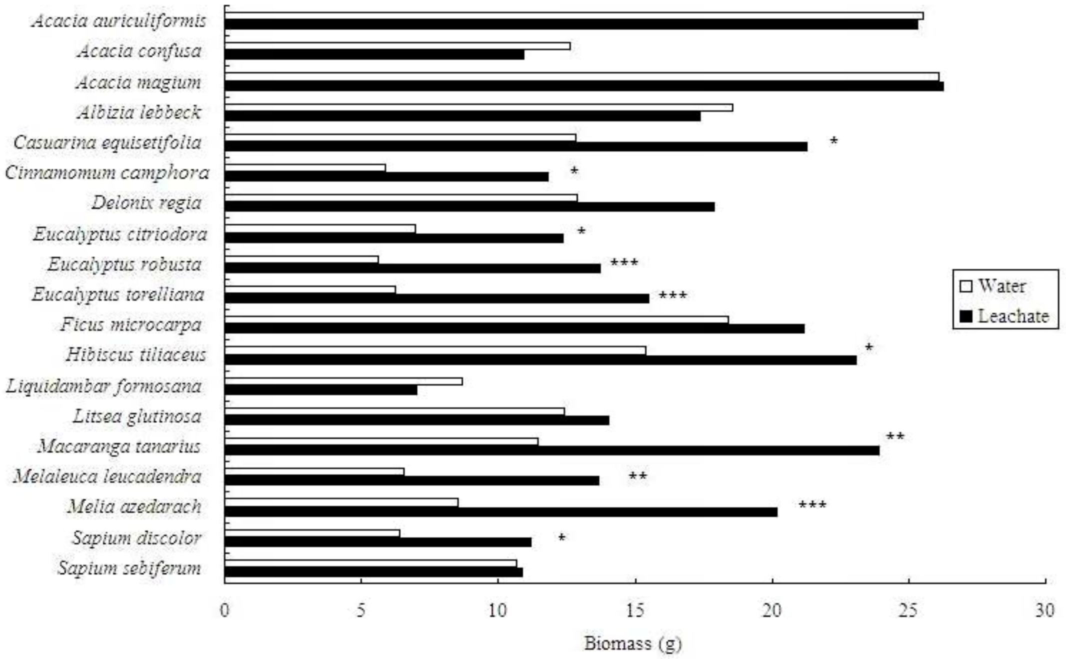

Figure 1 Biomass of seedlings of 19 tree species harvested after 90-day irrigation with water and diluted leachate (*: $p < 0.05$; **: $p < 0.01$; ***: $p < 0.001$ by t-test).

Nitrogen accumulation relying solely on symbiotic fixation and atmospheric deposition often go at a pace which limits ecological succession. The process is accelerated when there is external nitrogen input and more biomass formed with higher tissue nitrogen content. Most species exhibited increase in foliar nitrogen content under leachate irrigation (Figure 2). The NHx-N originated from leachate after being assimilated by plants would return to the soil as litter and become soil nitrogen capital [29].

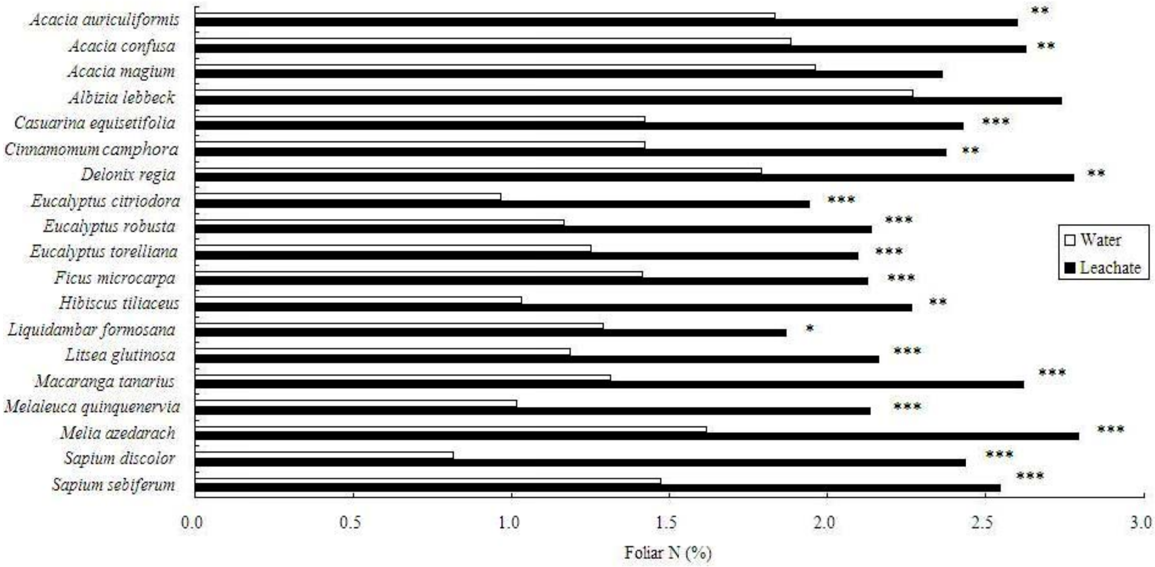

Figure 2 Foliar nitrogen contents of different tree species after 90 days of irrigation (*: $p < 0.05$; **: $p < 0.01$; ***: $p < 0.001$).

2.2. Effects on soil

In addition to supporting plant growth, it is essential to maintain a healthy edaphic environment. This includes the capacity of soil to function to sustain biological productivity, maintain environmental quality and promote plant, animal and human health [30]. The effect of prolonged application on plant productivity and soil biology was investigated by a 40-week leachate irrigation experiment, using soil columns 60 cm in length and landfill leachate diluted to the EC50 level (5% v/v) [28]. The electrical conductivity (EC) of landfill leachate (24 mS cm^{-1}) was much higher than that of saline water (0.75 - 2.25 mS cm^{-1}) (Landon, 1991) (31). Leachate application dramatically increased the EC of the soil after 40 weeks (Figure 3). Leachate enriched the soil with Cl^-, most of which was retained in the upper layer of the soil. After 20 weeks, the soil was saturated with Cl^- at about 250 mg kg^{-1}, a level that was marginal for plant injury [26].

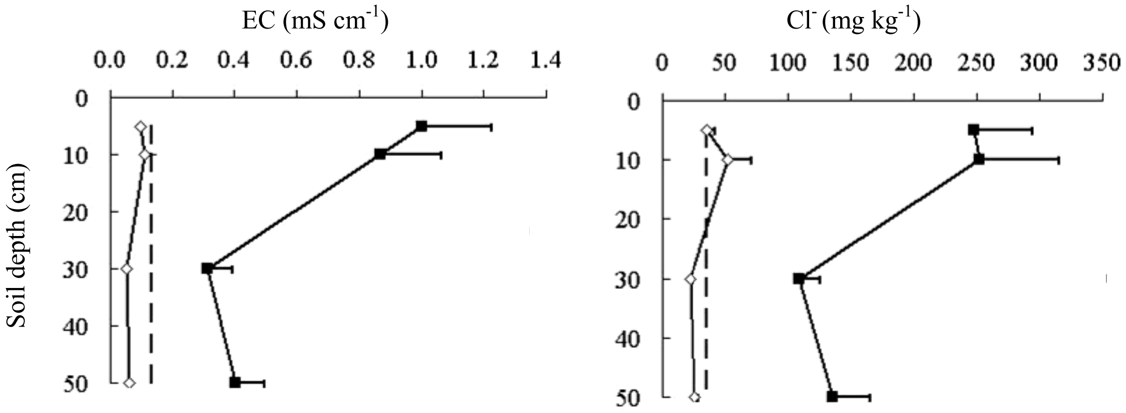

Figure 3 Electrical conductivity (EC) and chloride at different soil depths after irrigation with water (◇) or leachate (■) for 40 weeks. Broken lines represent the initial soil levels before irrigation. Error bars show the standard deviation of 5 replicates.

2.2.1. Salination

Besides minimizing the salt input by proper dilution of leachate, the impact of salinity can be reduced if salts are eluted away from the rooting zone. An excess amount of water, in addition to evapotranspiration, is added to leach salts out of the rooting zone to maintain an acceptable salt content, a concept known as leaching requirement [32]. The volume of irrigation can be calculated based on the evapotranspiration, salinity of the diluted leachate and crop tolerance. If the leaching requirement exceeds the upper limit of water input to soil, alternating irrigation with diluted leachate and water, or only diluted leachate in rainy seasons (usually the growing seasons) can help to control soil salt content. Bowman *et al.* reported the feasibility of alternating irrigation with leachate and water on grasses (*Cynodon dactylon* and *Pennisetum clandestinum*) to mitigate the problem of soil salination [33]. Leachate applied at 50% (one leachate irrigation followed by one watering) resulted in yield reduction, but the yield was better than water treatment when the frequency of leachate application was reduced to 20%. Compared with continuous leachate application, alternating irrigation may be a better option. Keeping the nitrogen input rate, both irrigation plans would add the same amount of salts to soil. However, the alternating irrigation plan can leach away the salts more efficiently. In other words, more salts can be leached out using smaller volumes of water. Thus, the hydraulic loading can be reduced. The irrigation practice can be further improved with the consideration of leachate phytotoxicity, nitrogen requirement and leaching requirement to avoid excessive application of nitrogen and salts. Moreover, planting salt-tolerant species (e.g. *Hibiscus tiliaceus*) can lower the adverse impact to vegetation in episodic drought conditions due to hot dry weather or a breakdown of the irrigation system.

2.2.2. Acidification

It has long been reported that the high contents of ammonium and base-forming cations such as Ca^{2+} and Mg^{2+} in leachate led to an increase in the pH of soil and soil percolate [34]. In contrast, marked decrease in pH was observed in soil subject to prolonged leachate irrigation. The effect of alkaline leachate on soil pH could be counteracted by the exchangeable and residual acidity of the soil, as well as the H^+ ions produced in microbial nitrification. Theoretically, 4 moles of hydrogen are produced for each mole of NHx-N oxidized by *Nitrosomonas*. In field conditions, the net acidity contributed by fertilizer was about 0.08 kg H^+ for each kg of NHx-N applied (equivalent to 1.12 mol H^+ per mol of NHx-N) [35]. Deviation from the stoichiometry of the empirical reaction may be attributed to loss of NHx-N by leaching, volatilization and plant uptake.

Plant uptake of NO_3^- releases bicarbonate ions (HCO_3^-) to maintain the electrical neutrality at the rhizophere and at the same time counteracts the acidity produced in nitrification. However, NO_3^- is very susceptible to leaching loss. Leaching, rather than root uptake of NO_3^-, primarily determines the degree of acidification. It is anticipated that if leachate irrigation continues, nitrification together with the leaching of NOx-N and base-forming cations would further reduce soil pH.

3. Fate of Nutrients in Soil

3.1. Nitrogen

Nitrogen is usually the limiting element in unfertilized ecosystems. The amount of available nitrogen in soil is small, while the quantity removed annually by plants is comparatively large. It should not be surprising that leachate irrigation remarkably increases soil NHx-N content by up to 4 times. However, landfill leachate which contained less than 1 mg NOx-N L^{-1} could increase soil NOx-N by 25 times after 40 weeks (Figure 4) [21]. In energy terms, assimilation of NH_4^+ is more efficient than NO_3^-, since NO_3^- requires reduction to NH_4^+ before incorporation into the synthesis of amino acids. Nitrification helps reduce the harmful effects of the leachate applied, as NO_3^- is much less toxic to aquatic life when compared with NHx.

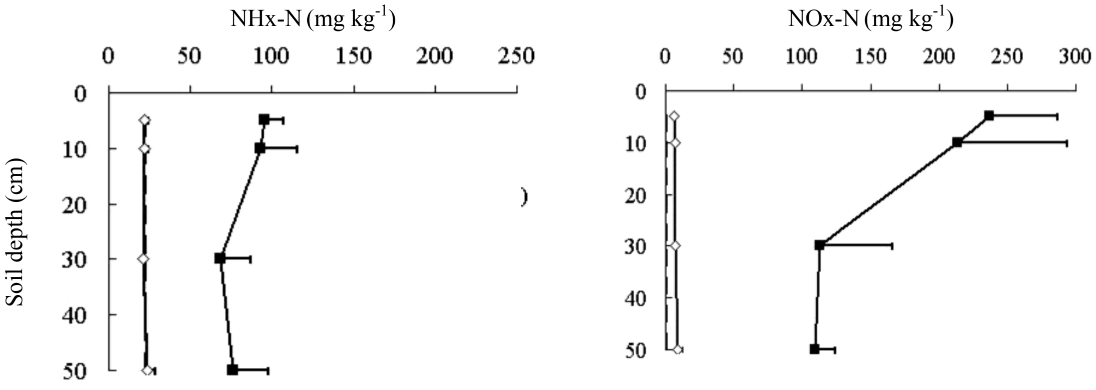

Figure 4 NHx-N and NOx-N contents at different soil depths after irrigation with water (◇) or leachate (■) for 40 weeks. Broken lines represent the initial nitrogen contents. Error bars show the standard deviation of 5 replicates.

Transformation of NH_4^+ to NO_3^- may lead to eutrophication and hypoxia in nearby water, as anionic NO_3^- has very low retention, and is relatively mobile in soil [29, 36, 37]. However, the impact of NO_3^- pollution is less serious in modern landfills since most of them are equipped with drainage and treatment systems to handle surplus moisture in the final soil cover. Besides proper runoff collection, the risk of NO_3^- pollution can be mitigated by determining the leachate application rate carefully to prevent excessive supply of nitrogen.

3.2. Phosphorus

Leachates are usually low in phosphorus, having the highest total phosphorus content of only 30.3 mg L^{-1}. The major form that existed in the leachate was orthophosphate (PO_4^{3-}). Phosphorus is essentially immobile in soil and the landfill body. Under alkaline conditions, PO_4^{3-} ions quickly react with Ca to form insoluble tricalcium phosphate ($Ca_3(PO_4)_2$). It further reacts to form compounds which are thousands of times less soluble [38]. Less than 0.01% of phosphorus in soil exists as soluble forms [36]. Therefore, foliar phosphorus contents were relatively lower with leachate than those with water (Figure 5). Phosphorus amendment with leachate irrigation may be costly yet ineffective. Phosphorus addition up to 87 kg P ha^{-1} did not further boost plant growth when compared with leachate treatment in a 90-day experiment (Table 1).

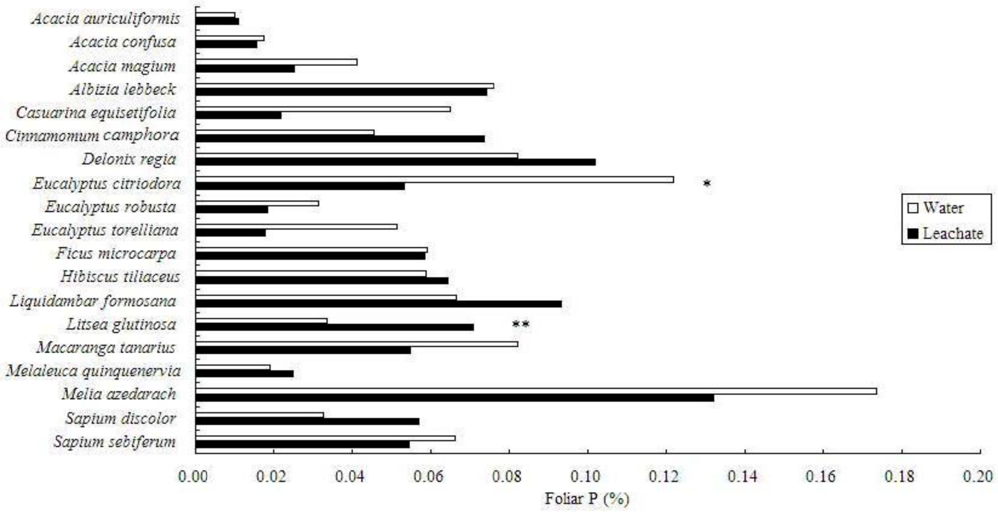

Figure 5 Foliar phosphorus contents of different tree species after 90 days of irrigation (*: $p < 0.05$; **: $p < 0.01$).

Table 1 Growth performance (% height increment) of *Eucalyptus citriodora*, *Hibiscus tiliaceus* and *Sapium discolor* after 90-day irrigation under different treatments. Different letters in a column represent a significant difference at p = 0.05 between different treatments of the same species according to Tukey's HSD test.

Species	Treatment	P added (kg P ha^{-1})	Height increment (%)
Eucalyptus citriodora	Water	0.00	5.69 ± 4.44 b
	Chemical fertilizer	43.1	31.2 ± 14.4 ab
	Leachate	0.00	39.1 ± 15.1 a
	Leachate + 0.5P	21.6	31.5 ± 12.3 ab
	Leachate + 1P	43.1	31.6 ± 18.9 ab
	Leachate + 2P	86.2	25.5 ± 18.6 ab
Hibiscus tiliaceus	Water	0.00	15.8 ± 8.63 b
	Chemical fertilizer	43.1	28.5 ± 10.0 ab
	Leachate	0.00	28.5 ± 5.68 ab
	Leachate + 0.5P	21.6	38.8 ± 22.8 ab
	Leachate + 1P	43.1	58.6 ± 24.3 a
	Leachate + 2P	86.2	39.5 ± 27.3 ab
Sapium discolor	Water	0.00	4.67 ± 6.86
	Chemical fertilizer	43.1	10.1 ± 3.93
	Leachate	0.00	9.93 ± 8.52
	Leachate + 0.5P	21.6	23.7 ± 12.9
	Leachate + 1P	43.1	16.5 ± 12.6
	Leachate + 2P	86.2	18.3 ± 10.3

4. Microbial Activity

Microbiological and biochemical properties of soil are sensitive and early indicators of ecological stress [39, 40]. As a result, soil enzyme activities could act as an integrated index of the soil biological status [41]. Soil enzymes are mostly extracellular, which are originated from microbes, root exudates or decomposition of roots and microflora. They are produced for the degradation of large molecules for plant and microbial absorption, and catalyse the rate determining soil functions such as organic decomposition, nutrient cycling and xenobiotic detoxification [42]. Intracellular enzymes like dehydrogenase and catalase are enzymes which are only found within viable cells, because their degradation in soil is rapid once they are released due to cell lyses. Hence, intracellular enzyme activities are a good indicator of the soil microbial activities.

4.1. Dehydrogenase

Dehydrogenases are a group of endocellular oxidoreductase which catalyze the dehydrogenation process. Dehydrogenase activity gives a measure of soil oxidative power, the number of viable microbial cells and microbial activities [43, 44]. It had been proven to be linearly correlated with microbial biomass [45]. Li and

Zhao showed a decrease in dehydrogenase activity with an increase in NHx-N concentration in synthetic wastewater [46]. Dehydrogenase activity increased in both the leachate treatment and the control with water irrigation (Figure 6) [28]. Higher nutrient and water contents have favourable effects for bacterial population growth. The constituents in the leachate seem not to affect the survival of soil microorganisms.

4.2. Phosphatase

In soil, a considerable part of phosphorus was bound as organic phosphoric acid ester in the form of phytanic acid and phytin. Plant phosphorus uptake requires mineralization of organic phosphorus to orthophosphate. Phosphatases are a group of enzymes that hydrolyze the ester and anhydride of phosphoric acid to generate phosphate. There was generally higher activity for the leachate-irrigated soil, which indicates leachate would stimulate the phosphatase activity as it provided more nutrients for microbial growth (Figure 6) [28]. In addition, leachate irrigation promoted plant growth which increased the amount of enzyme originated from plants. Acid phosphatase activity was much higher than alkaline phosphatase as the soil pH was acidic (Figure 4.6) which was near to the optimum pH of the acid phosphatase [47].

4.3. Urease

Urease catalyses the hydrolysis of urea to NH_3 and CO_2, and plays an important role in nitrogen mineralization [44]. Ureases are extracellular enzymes which are adsorbed onto the clay surface or humic acid molecules. They originate from plant litter, living or dead roots, microorganisms and animals [48]. Contrastingly, a negative correlation was observed between soil NHX-N content and the urease activity (R=-0.254, df=158, p<0.01), implying the inhibition of nitrogen mineralization by leachate irrigation [28]. Yet, the plant should receive ample supply of available nitrogen from leachate irrigation, and outweigh the reduced nitrogen supply from mineralization.

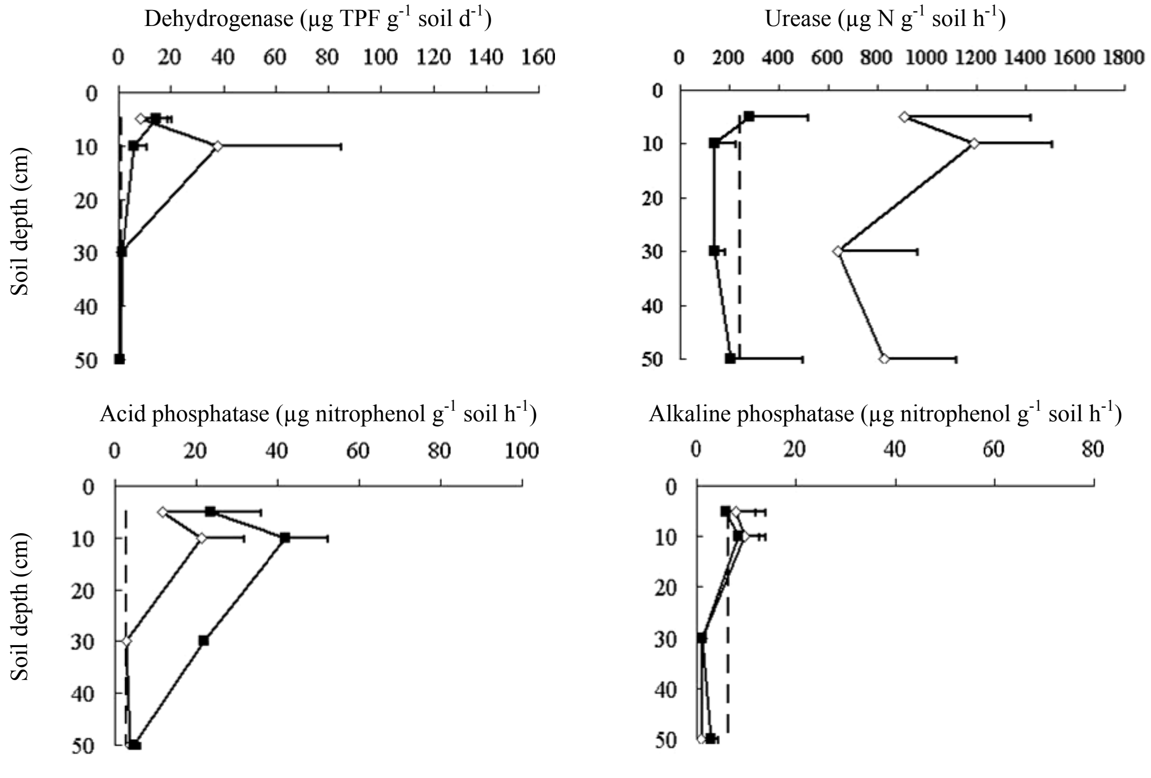
Dehydrogenase (μg TPF g^{-1} soil d^{-1})
Urease (μg N g^{-1} soil h^{-1})
Acid phosphatase (μg nitrophenol g^{-1} soil h^{-1})
Alkaline phosphatase (μg nitrophenol g^{-1} soil h^{-1})

Figure 6 Enzyme activities at different soil depths after irrigation with water (◇) or leachate (■) for 40 weeks. Broken lines represent the initial soil enzyme activities. Error bars show the standard deviation of 5 replicates.

5. Conclusions

Terrestrial ecosystems are often nitrogen-limited, either because of the low total reserve or low availability. Forest litter adds nitrogen back to the soil and form a closed cycle in which the annual rate of nitrogen uptake per unit area is limited by and approximately balances the annual nitrogen return to the forest floor [49]. About 95% of the nitrogen that cycle annually within the pedosphere interacts solely within the soil, plant and soil microbes [50]. This closed cycle differs from the open cycle of wetlands and fertilized croplands where primary production is mainly supported by external nitrogen supply. The addition of nitrogen by fertilizer application is likely to increase vegetative growth and the accumulation of nitrogen in both biomass and soil, hence the speed of ecological succession.

Leachate irrigation seems to be an attractive alternative of nitrogen and water source. However, toxicity and salt damage are major threats, though this could be reduced by appropriate dilution of leachate. On the other hand, it should be addressed that high soil nitrogen level and the massive leaching of NOx-N may result in not only the risk of water pollution but also the concern about nitrogen saturation which may impair the ecological succession of the recipient habitat. Nitrogen saturation occurs when the primary production is not increase further by increased nitrogen supply. Nitrification of the surplus nitrogen and subsequent leaching may lead to soil acidification. The loss of nitrogen may exceed the input over a rather long period of time. Moreover, excessive foliar nitrogen may disturb the nutrient balance and increase the sensitivity to frost, drought, pests and pathogens [51, 52]. Generally young, rapid-growing and well-nourished plants are more likely to suffer from attack by pests. A high content of amino acids in the plants may result in a more severe attack by sucking parasites [53]. Change in the vulnerability of plants, together with the soil acidification, may finally lead to the decline of forest.

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