

LANDFILL LEACHATE MONITORING: APPLICATIONS OF STABLE ISOTOPES

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Introduction

The most significant impact of a landfill on the surrounding environment has been consistently identified as arising from the leachate (Farquhar 1989; CAE 1992; Mulvey 1997). Therefore, environmental monitoring programmes for landfills tend to focus on the detection of leachate impacts, particularly any contamination of ground and surface water (Lee and Jones 1991; DCC 1994). Conventional monitoring programmes involve periodic sampling of ground and surface waters surrounding a landfill, for chemical indicators of contamination, such as ammonium, chloride, and heavy metal levels. However, this conventional approach may not be effective in environments where the background levels of many of the chemical indicators are already high, such as wetland areas or sites with contamination that pre-dates the landfill. There is a need for an indicator that can positively identify leachate as the source of contamination in a monitored environment. Stable isotope analysis is being increasingly used in studies of environmental pollution as a means of pinpointing pollution sources (Peterson and Fry 1987; Macko and Ostrom 1994). The relatively distinct isotopic ratios or signatures of substances, created through biogeochemical processes, offer great potential for tracing contaminants such as leachate in the environment¹.

Overview of stable isotopes

Elements (e.g. nitrogen and carbon) can be composed of either heavy or light atoms. These different forms of the same element are called isotopes. *Stable* isotopes, as opposed to *unstable* isotopes, do not radioactively decay. The ratio of heavy to light isotopes in a given element varies depending on the biogeochemical reactions affecting that element. A simple rule of thumb for understanding the partitioning of heavy and light isotopes states that: The lighter isotopes of an element tend to react more easily during biogeochemical reactions, resulting in lighter products and heavier reactants (Fritz and Fontes 1980; Kendall 1998; Criss 1999).

For instance, a common biogeochemical reaction in the environment is nitrification ($NH_4^+ \rightarrow NO_3^-$). Nitrogen has two isotopes (^{14}N and ^{15}N) and during the nitrification process the lighter isotope (^{14}N) detaches easily from the ammonium molecule (NH_4^+) and proceeds to form the nitrate molecule (NO_3^-). This causes the reacted ammonium molecule to become

¹ It should be noted that a contaminant is a foreign substance introduced to an environment, whereas a pollutant is a foreign substance introduced to an environment and resulting in deleterious effects (Ketchum, 1972). Landfill operators and associated monitoring agents accept that generally a degree of localised contamination from leachate will occur around a landfill site and can be tolerated, whereas pollution is unacceptable (Campbell, 1993).

heavier or more *enriched* (i.e. more ^{15}N than ^{14}N) and the produced nitrate molecule to become lighter or more *depleted* (i.e. more ^{14}N than ^{15}N).

Those isotopes, which tend to be the focus of most environmental applications of natural, stable isotopes (Fritz and Fontes, 1980) and are referred to as the *environmental isotopes* are presented in Table I.

Element	Isotopes
Hydrogen	^1H , ^2H , ^3H
Carbon	^{12}C , ^{13}C , ^{14}C
Nitrogen	^{14}N , ^{15}N
Oxygen	^{16}O , ^{18}O
Sulphur	^{32}S , ^{34}S

Table I: The environmental isotopes.

Stable isotopes in landfill studies

In order for stable isotopes to be successfully used to identify leachate contamination in the environment, a distinct isotopic signature (i.e. the ratio of light to heavy isotopes) needs to be identified. If an isotopic signature can be established for leachate, then the following questions could be addressed:

- Is leachate currently exiting the landfill site and, if so, how much is exiting?
- If leachate is exiting, then what is the extent and nature of its interaction with the surrounding environment?

To date, relatively little research appears to have been undertaken on stable isotopic ratios of landfill leachate or leachate contamination of the environment. Fritz, Bryan *et al.* (1994) applied ^{34}S and tritium isotopes to “delineate the nature, flow path, and flow rate” of leachate from a landfill site. Highly enriched ^{34}S found in groundwater monitoring wells confirmed leachate contamination, and the source of the enriched sulphates was traced to gypsum-based plasterboard buried in the landfill. Tritium data has been used to determine the age of leachate being intercepted by monitoring wells, by comparing results for monitoring well samples with historical records of tritium levels in local precipitation (Fritz, Bryan *et al.*, 1994).

Hackley, Liu *et al.* (1996) described the environmental isotopic characteristics of landfill leachates and gases, and suggested that isotopic analysis of leachate could be a powerful technique for ascertaining landfill contamination. In particular, they provided evidence that methanogenesis² occurring in landfills leads to unique isotopic signatures for ^2H and ^{13}C identifiable in leachate samples.

² Methanogenesis is an anaerobic bacterial process that produces methane (CH_4) and is active during the methanogenic phase of landfill development, which generally begins 15 years after of the initial deposition of waste in a landfill.

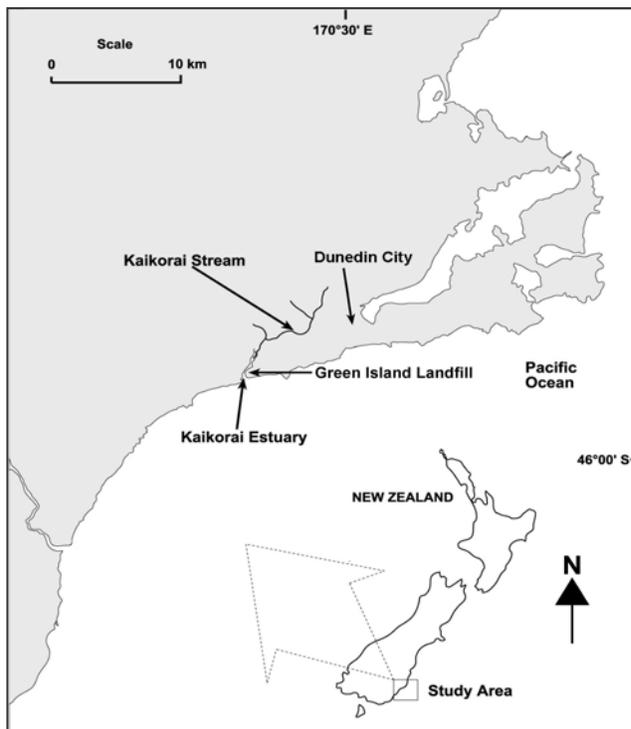
Rank, Papesch *et al.* (1995) obtained a 7-8 year data set of leachate samples to examine the chronological development of the isotopic content of leachate starting with the first waste deposition at a landfill site. Their research focussed on the ^2H and ^3H isotope contents of water contained within the landfill and at the landfill-liner interface. The study concluded that elevated ^3H concentrations (due to leachate contamination by ^3H sources, such as the luminescent dials of watches, in the waste) are likely to be characteristic of most young landfills. Furthermore, there was a strong enrichment of ^2H in leachate and a depletion of ^2H in methane at the onset of the methanogenic phase. The ^{13}C of dissolved inorganic carbon was also observed to increase with the onset of methanogenesis (Rank, Papesch *et al.*, 1995) and an unusually high ^{13}C for dissolved inorganic carbon was observed in groundwater wells suspected of leachate contamination (Arneth, 1988).

The focus of the present study is to investigate the ^{13}C and ^{15}N isotopes in components of landfill leachate to determine whether or not biogeochemical processes within the landfill environment produce a distinct isotopic composition.

The present study

Methods

Leachate, ground and surface water, and vegetation samples were periodically collected from the Green Island Landfill (GILF) and surrounding Kaikorai Estuary, Dunedin, New Zealand (refer to Figure 1) from August 2000 to May 2001. Leachate and groundwater samples were collected from monitoring wells located around the perimeter of GILF. Surface water and vegetation samples were collected from the Kaikorai Stream. Upstream samples were collected at sites approximately 400 m upstream from the GILF and downstream samples were collected at the southernmost limit of the GILF on the Kaikorai Stream.



The landfill lies on reclaimed wetland area within the Kaikorai Estuary. The Kaikorai Stream forms the northern and north-western limits of the landfill. Another active landfill, operated by Envirowaste, is located opposite the GILF across the Kaikorai Stream and also within the bounds of the estuary.

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Figure 1: Location of Green Island Landfill.

Water and leachate samples were analysed for ^{13}C of their dissolved inorganic carbon (CO_2) component and ^{15}N of their nitrate and ammonium components. Vegetation samples were analysed for both ^{13}C and ^{15}N of their particulate organic matter (POM) component.

Results are expressed as a ratio of the heavier to lighter isotope in a sample compared with that same ratio in a suitable standard. This is referred to as a δ (delta) value and uses units of per mil (‰)(Peterson and Fry, 1987).

Results and discussion

The results of isotopic analysis for the nitrate (NO_3^-), ammonium (NH_4^+), and dissolved inorganic carbon (CO_2) components of upstream, downstream, and leachate samples from GILF are summarised in Table II³. These results suggest a general trend of enriched $\delta^{15}\text{N-NH}_4^+$ and depleted $\delta^{15}\text{N-NO}_3^-$ for landfill leachate, and depleted $\delta^{15}\text{N-NH}_4^+$ and enriched $\delta^{15}\text{N-NO}_3^-$ for stream sites. Downstream sites appear to have $\delta^{15}\text{N}$ characteristics approaching those of leachate, with higher $\delta^{15}\text{N-NH}_4^+$ and lower $\delta^{15}\text{N-NO}_3^-$ values than upstream sites.

Site	Average $\delta^{15}\text{N-NO}_3^-$ (‰)	Average $\delta^{15}\text{N-NH}_4^+$ (‰)	Average $\delta^{13}\text{C-CO}_2$ (‰)
Upstream	2.14 ± 3.44	-2.71 ± 2.93	-15.09 ± 0.02
Downstream	-4.87 ± 2.42	7.63 ± 3.18	20.18 ± 0.14
Leachate (from monitoring wells)	-4.54 ± 2.05	18.68 ± 8.88	16.11 ± 0.23

Table II: Summary of results for isotopic analysis of the nitrate, ammonium, and dissolved inorganic components of upstream, downstream, and leachate samples from GILF.

A number of reasons may account for these trends. As organic solid waste decays in the landfill organic nitrogen is converted to ammonium, which causes only a small change in the ratios of light to heavy N isotopes (i.e. $\delta^{15}\text{N-NH}_4^+ \approx \delta^{15}\text{N-organic matter} \approx -5$ to $+2$ ‰). Although landfills are generally considered anaerobic environments, the heterogeneous mixture of wastes can create air pockets, and rainwater also provides an oxygen input. Oxidising conditions in the landfill may cause two reactions: 1) volatilisation (NH_4^+ converted to N_2), and 2) nitrification. The former reaction leaves NH_4^+ highly enriched in ^{15}N , whereas in the latter reaction, ammonium is converted to nitrate, which results in a depletion of $\delta^{15}\text{N-NO}_3^-$ (Mariotti, Lancelot *et al.*, 1984). The more prevalent reducing conditions in a landfill may cause reduction of nitrate to ammonium, and consequently lead to enhanced $\delta^{15}\text{N-NH}_4^+$ values (i.e. the reverse of nitrification), and lower $\delta^{15}\text{N-NO}_3^-$. This sequence of biogeochemical reactions would explain why depleted $\delta^{15}\text{N-NO}_3^-$ values and highly enriched $\delta^{15}\text{N-NH}_4^+$ values were observed in leachate samples from GILF.

³ For details of additional isotopic and ancillary data analyses, undertaken during the present study, please refer to: *Landfill leachate monitoring: applications of stable isotopes* (unpublished MSc thesis, J.C. North, 2001).

The $\delta^{15}\text{N}$ values at downstream sites may reflect the influence of leachate entering the Kaikorai Stream, or an additional source of N with inputs occurring between upstream and downstream sites. $\delta^{15}\text{N-NO}_3^-$ values at the downstream sites do not reflect a marine influence, as might be expected since these sites are closest to the mouth of the Kaikorai Estuary. Marine $\delta^{15}\text{N-NO}_3^-$ tends to be enriched (7 to 10 ‰), particularly with respect to fresh water (Libes, 1992), whereas the downstream sites have a depleted $\delta^{15}\text{N-NO}_3^-$ value compared to upstream values.

Stream water CO_2 can range in $\delta^{13}\text{C}$ values from -5 to -25 ‰ (Hackley, Liu *et al.*, 1996; Kendall *et al.*, 1995), whereas the $\delta^{13}\text{C}$ values for CO_2 of leachate can be up to $+20$ ‰ (Hackley, Liu *et al.*, 1996). Enrichment of leachate $^{13}\text{C- CO}_2$ is caused by methane-producing bacteria, which preferentially use the lighter ^{12}C to form CH_4 .

The $\delta^{13}\text{C- CO}_2$ value for upstream sites (-15.09 ‰) lies within the expected range of stream water $\delta^{13}\text{C}$ value. Downstream sites have an enriched $^{13}\text{C- CO}_2$ value (20.18 ‰), which suggests that a source of enriched CO_2 is entering the Kaikorai Stream at some point after the upstream sampling area. This is not a source of marine origin, since the oceanic CO_2 pool has a $\delta^{13}\text{C- CO}_2$ of approximately 0 ‰. The most likely source is $\delta^{13}\text{C- CO}_2$ enriched leachate since leachate samples exhibit highly enriched $\delta^{13}\text{C- CO}_2$ values (16.11 ± 0.23 ‰).

Table III summarises mean $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values obtained from plant material collected from sampling sites along the Kaikorai Stream. The mean value for plant material (*Arrhenatherum elatius*) $\delta^{15}\text{N}$ became increasingly enriched in the sequence: upstream site (mean $\delta^{15}\text{N} = 5.42 \pm 1.54$ ‰), to downstream site (mean $\delta^{15}\text{N} = 10.28 \pm 2.93$ ‰). Plant material mean $\delta^{13}\text{C}$ was most depleted at the upstream site (mean = -30.71 ± 1.03 ‰) and most enriched at the midstream site (mean = -28.09 ± 0.79 ‰). The distributions of all $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values are presented graphically in Figure 2.

SITE	Mean $\delta^{15}\text{N}$ (‰)	Mean $\delta^{13}\text{C}$ (‰)
Upstream	5.42 ± 1.54	-30.71 ± 1.03
Midstream	8.72 ± 1.84	-28.09 ± 0.79
Downstream	10.28 ± 2.93	-28.88 ± 1.91

Table III: Summary of results for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analyses of plant matter from the Kaikorai Stream.

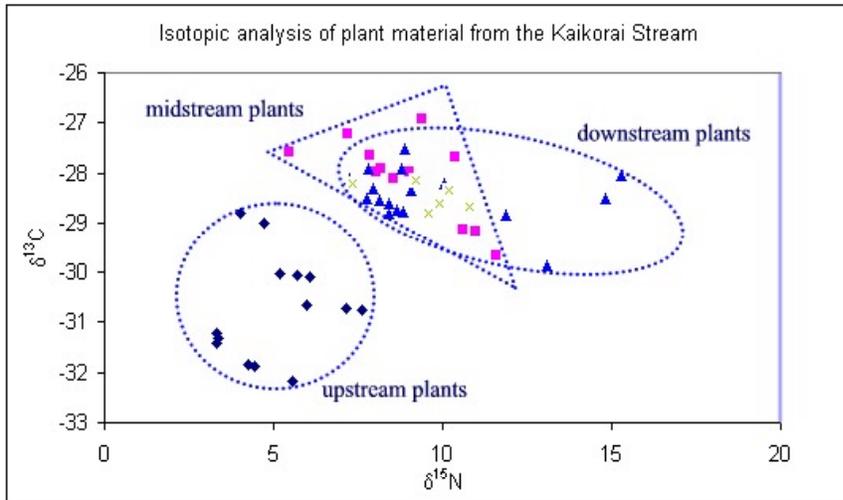


Figure 2: Distribution of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values (in ‰) for plant material taken from sites on the Kaikorai Stream. Results are grouped into upstream, midstream and downstream plants.

$\delta^{15}\text{N}$ of plant material becomes progressively more enriched from upstream sites to downstream sites (refer to Figure 3). $\delta^{13}\text{C}$ values for plant matter do not show a downstream trend, although overall the midstream and downstream sites are more enriched in ^{13}C (-28.88 to -28.09 ‰) than the upstream sites (-30.71 ‰). This suggests that an enriched ^{15}N and ^{13}C source is contributing to the isotopic enrichment of plant nutrients downstream. The potential sources include: 1) increasing marine/estuarine influence downstream; 2) an unidentified ^{15}N and ^{13}C -enriched nutrient source active along the Kaikorai Stream; and 3) leachate inputs to the Kaikorai Stream.

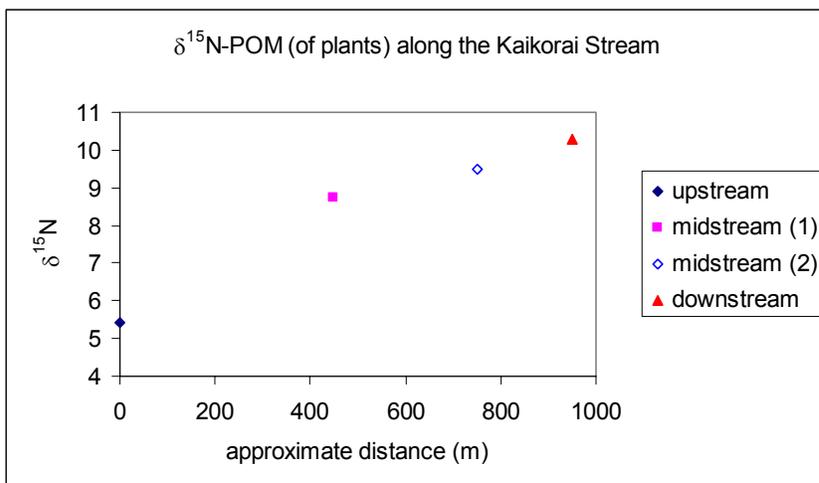


Figure 3: Plant material $\delta^{15}\text{N}$ values (‰) plotted against approximate distance along the Kaikorai Stream, starting at the upstream sampling area.

The results for flora from downstream sites (10.28 ± 2.93 ‰) indicate the influence of an enriched ^{15}N nutrient source, which may be estuarine ammonium. Estuarine $\delta^{13}\text{C}-\text{CO}_2$ values range from -29 to +2 ‰, with more negative values in fresh waters and more positive values in seawater (Coffin, Cifuentes *et al.*, 1994). Therefore, it is possible that more positive estuarine $\delta^{13}\text{C}-\text{CO}_2$ could influence the more positive plant $\delta^{13}\text{C}$ at downstream sites. However the $\delta^{13}\text{C}-\text{CO}_2$ values do not entirely support this hypothesis. The downstream site

exhibits an enriched ^{13}C - CO_2 value (20.18 ‰), which is neither from a source of marine origin (oceanic CO_2 pool is approximately 0 ‰) nor from an estuarine source ($\delta^{13}\text{C}$ - CO_2 ranges from -29 to +2 ‰ (Coffin, Cifuentes *et al.*, 1994)) (refer to Figure 4).

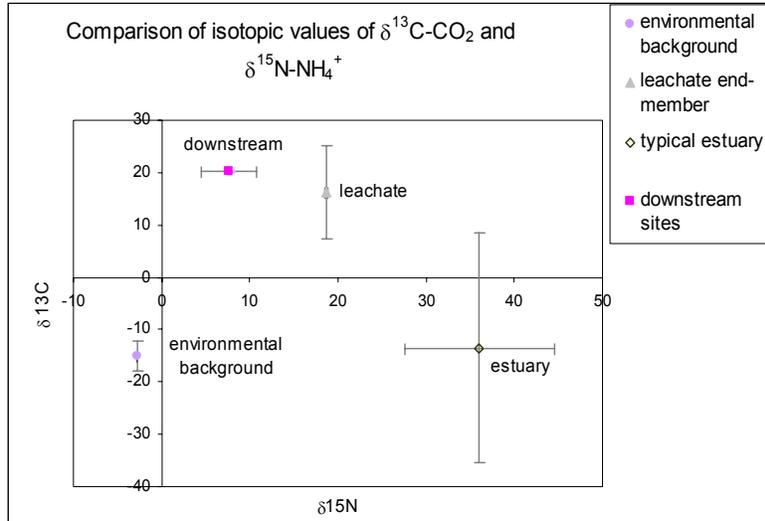


Figure 4: Environmental background (upstream) $\delta^{13}\text{C}\text{-CO}_2$ and $\delta^{15}\text{N}\text{-NH}_4^+$ values for the Kaikorai Stream compared to those of downstream sites, leachate, and typical values of an estuary during spring, summer, and fall months (Velinsky, Pennock *et al.*, 1989). The isotopic composition of downstream sites does not appear to be influenced by the estuary, but does appear to be influenced by leachate.

The second option would require further investigation. Between upstream and downstream sites the main activity is essentially solid waste disposal in landfills on both sides of the Kaikorai Stream. However, livestock occasionally graze on grass verges along the Kaikorai Stream, and there is also a large waterfowl population in the Kaikorai Estuary. Animal and poultry manure ranges in $\delta^{15}\text{N}$ value from 10 – 25 ‰ (Wassenaar, 1995; Kendall, 1998), which could account for the enriched $\delta^{15}\text{N}$ value for downstream sites if enough quantity of manure entered the stream. However, the $\delta^{13}\text{C}$ value for ruminant effluent would be expected to reflect that of their diet (Wahlen, 1994), which is typically plant material with an average $\delta^{13}\text{C}$ value of -27 ‰ (Lajtha and Marshall, 1994). If ruminant effluent were a significant nutrient source in the Kaikorai Stream, then a lighter $\delta^{13}\text{C}\text{-CO}_2$ would be expected than the enriched value observed in the present study (20.18 ‰). Therefore the $\delta^{13}\text{C}$ data does not implicate livestock as a source of nutrients in the Kaikorai Stream.

The third option suggests that leachate is contributing an enriched $\delta^{15}\text{N}$ nutrient source to downstream flora. Since leachate has a very high concentration of ammonium (approximately 740 + mg/l $\text{N}\text{-NH}_4^+$), plants exposed to leachate as a nutrient source would probably preferentially uptake ammonium. Results of $\delta^{15}\text{N}\text{-NH}_4^+$ analysis show that leachate $\delta^{15}\text{N}\text{-NH}_4^+$ values average approximately 19 ‰, which represents a significantly enriched $\delta^{15}\text{N}$ nutrient source. Furthermore, $\delta^{13}\text{C}$ values for CO_2 of leachate can be up to +20 ‰ (Hackley, Liu *et al.*, 1996), which is supported by results of the present study (16.11 ± 0.23 ‰), and reflected in both the enriched $\delta^{13}\text{C}\text{-CO}_2$ value (20.18 ‰) and the higher plant $\delta^{13}\text{C}$ value at downstream sites compared to upstream sites. Therefore, leachate is the likely source of additional downstream plant nutrients in the Kaikorai Stream, although further research is

required to determine the extent of leachate contamination, and whether or not leachate is actually polluting the Kaikorai Stream and Estuary.

It should be emphasised that there is no conclusive evidence that GILF is leaking leachate into the Kaikorai Stream. Other sources of leachate might include the Envirowaste landfill, which is also situated within the Kaikorai Estuary, or an additional (unknown) influence.

Conclusions

The investigation of ^{13}C and ^{15}N isotopic characteristics of leachate at GILF revealed an exciting potential for their use as an environmental monitoring tool. Using $\delta^{13}\text{C-CO}_2$ and $^{15}\text{N-NH}_4^+$ values, a distinct isotopic signature was determined for leachate, consisting of highly enriched $^{13}\text{C-CO}_2$ and highly enriched $^{15}\text{N-NH}_4$. In combination with conventional monitored chemical parameters such as ammonium concentrations, isotopic ratios of ^{13}C and ^{15}N enabled the positive identification of leachate in the Kaikorai Estuary. In particular, ^{13}C and ^{15}N analysis of plant matter from the Kaikorai Stream revealed leachate as a probable source of nutrients for such downstream flora. The source of leachate is not certain: either of the two landfills (GILF and the Envirowaste facility) could be contributing to the contamination.

A number of further studies could assist in the positive identification of the leachate source, including a measurement of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in plant material along transects between the two landfills. Enriched ^{13}C and ^{15}N in plant material may be indicative of nutrient uptake from leachate. Therefore, if ^{13}C and ^{15}N enrichment in plants decreased from GILF to the Envirowaste site, it would suggest that leachate influence was more likely to be due to GILF. Conversely, if ^{13}C and ^{15}N enrichment increased from GILF towards the Envirowaste site, then leachate from the Envirowaste site would appear to be the main source of nutrients contributing to plants.

Most importantly, the impact of leachate on the receiving environment needs to be determined. This would likely involve an estimate of the quantity of leachate entering the Kaikorai Estuary, the contribution of leachate to estuarine food-webs, and notable differences in biodiversity, ecological health, and pollutant load of organisms between the Kaikorai Estuary and a carefully selected control estuary. The Kaikorai Estuary may be able to buffer leachate contamination without incurring any negative environmental impacts. However, a full assessment of both potential short- and long-term impacts from chronic exposure to landfill leachate in this area should be made before it could be dismissed as a non-pollutant.

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