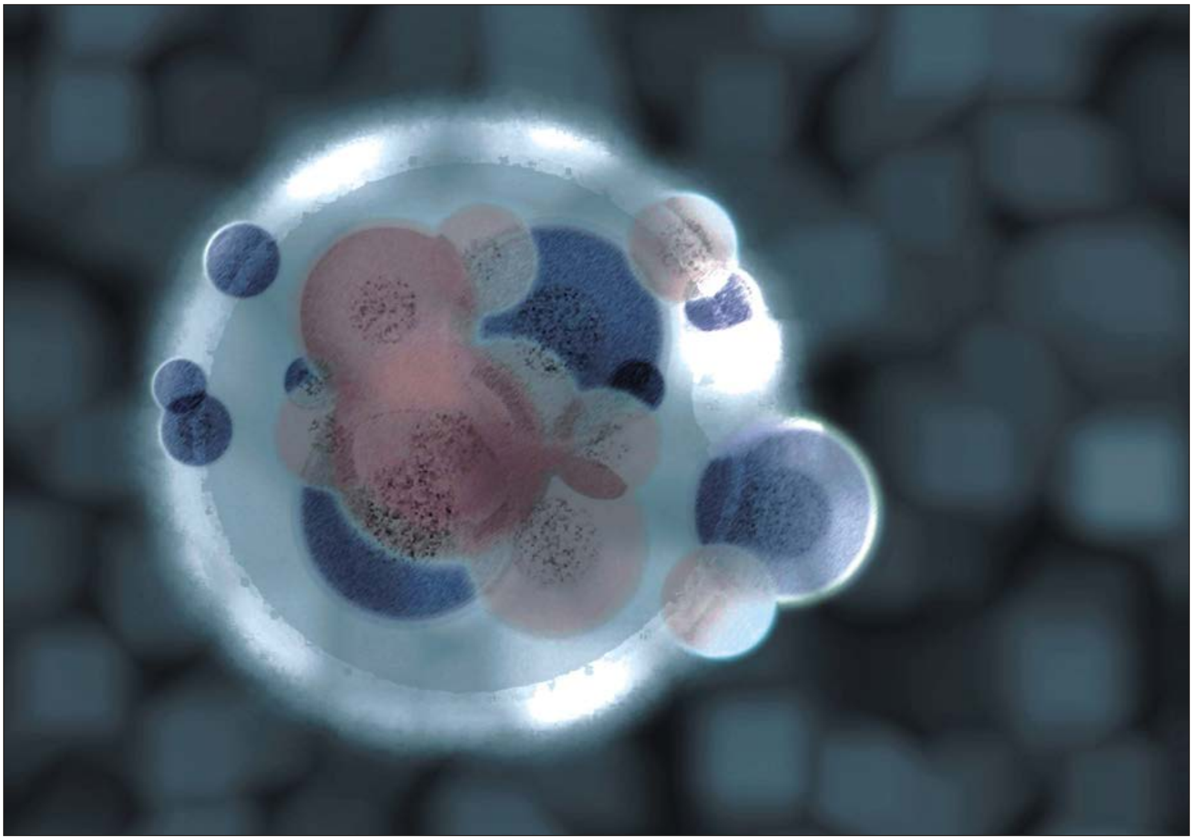

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Removal of Ammonia from Landfill Leachate in a Two-Stage Biofiltration Process

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Abstract: The efficiency of nitrification and denitrification in a two-stage biological filter in the removal of ammonia from sanitary landfill leachate, using plastic media, was investigated. A pilot-scale biological filter constituted of leachate from Weltevreden sanitary landfill was built. During the first 25 weeks of the investigation no external carbon source was added. To facilitate denitrification methanol was added to the pilot-scale. The first 9 weeks of the study was characterised by the raw leachate that had an average pH value of 8.89±0.13, an ammonia-nitrogen concentration of 450±20 mg N L^{-1} and a COD concentration of 1600±116 mg O_2 L^{-1}. The change in raw leachate characteristics resulted in the decrease of pH and alkalinity which necessitated the addition of soda-ash (Na_2CO_3) to achieve complete nitrification in the first stage of the pilot-scale. A decrease in ammonia-nitrogen was noted from 200±10 to 2±2 mg N L^{-1} and a 99% ammonia removal was therefore recorded after Stage 1. In Stage 2, denitrification took place after the addition of the methanol at a methanol:nitrate ratio of 1.23:1. The nitrite and nitrate were 0.20±0.02 and 13.67±0.58 mg N L^{-1}, respectively, at the end of the experiment which was acceptable according to the general authorisation from the Department of Water Affairs and Forestry (DWAF). The COD removal was very poor since only 36% was removed at end of the experimental period; which meant that 64% was non-biodegradable. Ammonia-nitrogen from landfill leachate can be removed by a two-stage biofiltration process.

Key words: Ammonia, biological filter, landfill leachate, nitrification

INTRODUCTION

Most of the Municipal Solid Waste (MSW) generated in South Africa each year is placed in landfills. South Africa has 750 operating landfill sites (ENVIRO-FILL, 2004). Schoeman *et al*. (2003) reported that there are approximately eight strong leachate-producing landfill sites in South Africa and many more weak leachate-producing sites. Decay of the waste material can give rise to increased levels of heavy metals and poisonous chemical substances, which leach into the surrounding soil and contaminate groundwater, rivers and streams. Leachate characteristics vary not only because of the different kinds of waste present, but also according to the age of the landfill.

Leachate is a toxic waste which could pollute ground water supplies and soils, if the landfills are not lined with clay and/or a geomembrane. It also contains high concentrations (200-4000 mg N L^{-1}) of ammonia and poorly degradable organic compounds (Geens *et al*., 2000). Ammonia concentration remains high throughout all the stages of landfill waste degradation. The high concentration of ammonia-nitrogen (>10 mg N L^{-1}) can create numerous problems for the environment, such as eutrophication of surface water because nitrogen is an essential growth nutrient for plant material. Leachate can contaminate groundwater if the landfill site is not lined. When leachate is not treated before discharged or when it leaches into the soil, depending on the geological formation, it can contaminate surface water when entering streams, it affects aquatic animals due to the depletion of dissolved oxygen in receiving water and when ammonia is biologically oxidised to nitrite in the presence of oxygen (WEF, 1998). Therefore, in general, landfill leachate should be treated to the required standards for discharge (e.g., ammonia should be less than 3 mg N L^{-1}), whether in the vicinity of a domestic sewer, a natural water course, land or tidal water (DOE, 1995). The options for the treatment of landfill leachate for removal of ammonia are numerous and vary in design and operational complexity (Strachan *et al*., 2000). The options include complex physical-chemical methods and/or biological treatment methods.

Biological nitrification and denitrification are the traditional methods used to treat nitrogenous wastes such as municipal sewage, agricultural and landfill leachate (Shiskowski and Mavinic, 1998). The removal of nitrogen by biological nitrification/denitrification is a two-step

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process. In the first step, ammonia is converted aerobically to nitrate (nitrification) and in the second step nitrates are converted to nitrogen gas (denitrification) (Metcalf and Eddy, 1991).

Nitrification is the biological oxidation of ammonia to nitrate by autotrophic bacteria. These bacteria derive energy from the oxidation reaction and utilise inorganic compounds as their principal source of food for cell synthesis. There are two specific genera of autotrophic bacteria involved in nitrification, namely the *Nitrosomonas* species and *Nitrobacter* species. The growth rate for *Nitrosomonas* species is lower than that for *Nitrobacter* species, which means that the rate of conversion of ammonia to nitrate by *Nitrosomonas* species is much slower than that of nitrite to nitrate by *Nitrobacter* species (Metcalf and Eddy, 1991).

Biological denitrification is the reduction of nitrate to nitrogen gas by facultative heterotrophic organisms that require carbon as a source of food (Metcalf and Eddy, 1991). The nitrified liquor is usually deficient in organic carbon and a low carbon source level can limit the overall biological denitrification process and enough organic carbon sources must be provided for proper denitrification (Ra *et al*., 2000). A sufficient carbon:nitrogen ratio of at least 2:1 is necessary to complete denitrification reaction in natural systems (Metcalf and Eddy, 1991).

Nitrification and denitrification can be obtained in biological filters with plastic and rock media. But plastic media has high surface contact areas than rock media, so that and larger quantities of active microorganisms can be immobilised, thus a higher organic loading can be applied, while still achieving good nitrification (Metcalf and Eddy, 1991). Another factor favouring plastic medium filter is its high ventilation which permits higher oxygen transfer. The objective of this investigation was to evaluate a two-stage biofiltration process by using microorganisms forming a biofilm on plastic media to remove ammonia from sanitary landfill leachate. This process consisted of an upper part for nitrification and a lower part for denitrification. The efficiency of this process in reducing COD was also investigated.

MATERIALS AND METHODS

Landfill leachate: The leachate used in the study was collected from Weltevreden Sanitary landfill site which is located next to Main Reef Road in Brakpan. The site, established in 1995, is a class D site and about 18 000-20 000 metric tons of domestic waste are disposed monthly to the general waste section (G:B$^+$) of the site. Leachate produced at the site was approximately 10 Ml per year (Ekurhuleni Metropolitan Council, 2004). The annual rainfall that was received at the site during the time of study was 500 mm (Ekurhuleni Metropolitan Council, 2004). Leachate was collected in drums and transported to the laboratory.

Laboratory scale reactor set-up: A polyvinyl chloride (PVC) laboratory scale reactor had the following dimensions: 330 mm wide and 3000 mm long. There were two stages in the reactor; the first stage for nitrification (upper part of the reactor) and the second stage for denitrification (the bottom part). Both stages were packed with plastic trickling filter medium. Between the two sections of the reactor a collection point was inserted as shown in a schematic representation of the two-stage biological filter (Fig. 1). The reactor was operated in a down-flow mode to allow air to contact with the void spaces in the nitrification stage.

Nitrification stage: The length of the nitrification stage in the biofilter was 1700 mm. This stage was packed with two 300×300×300 mm Terbo-Pac plastic medium type TBX 2700 (minimum organic load of >800 g BOD_5/m^3/day) on top of three 300×300×300 mm Terbo-Pac plastic media type TBX 1900 (minimum organic load of >300 g BOD_5/m^3/day), designed specifically to promote nitrification. The air for aeration in this stage was supplied by means of an air pump at a flow rate of 55 l/min through a grit of perspex tubing which was placed at the bottom of Stage 1.

Denitrification stage: The length of the denitrification stage in the biofilter was 1000 mm. The denitrification stage was packed with two 300×300×300 mm Terbo-Pac

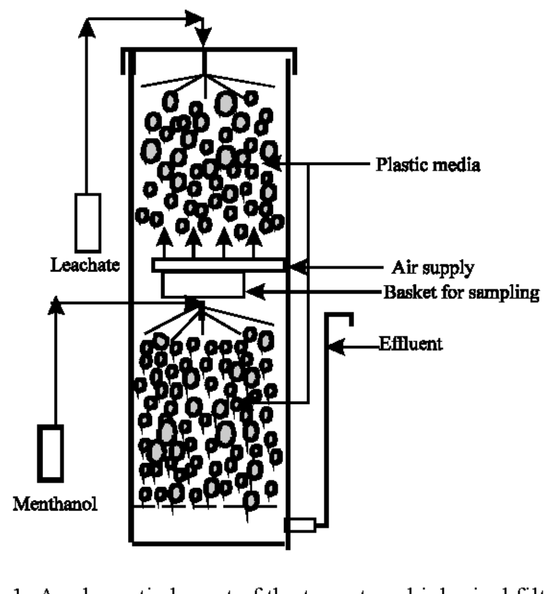

Fig. 1: A schematic layout of the two-stage biological filter

plastic medium type TBX 2700 (minimum organic load of >800 g $BOD_5/m^3/d$). This stage was submerged, to allow anaerobic conditions for denitrification. Effluent from the bottom was collected in a container situated on the same level on the top of the second stage. The detention time of the second stage of the reactor was 17 days.

Operational condition of the biological filter: The reactor was operated in two phases for a period of twelve months. During the first phase which lasted for eight months, no external carbon source was added to the process. However, during the second phase methanol was added. Leachate was fed to the top of the reactor at a dosing rate of 6 L day^{-1}. For the first four weeks of operation, raw leachate was seeded with primary settling tank effluent from a domestic waste water treatment plant. (Daspoort sewage treatment plant, Pretoria). Only raw leachate was fed into the reactor after four weeks, but due to a deficiency of phosphorous in the landfill leachates, 2.4 mg P L^{-1} in the form of KH_2PO_4 was added to the feed water to ensure biological growth. Micro-organisms requires a COD:P ratio of 100:1 as reported by previous investigators (Metcalf and Eddy, 1991).

The concentration of the leachate varied during the trial. After 10 weeks of operation the concentrations changed but it remained constant for the last 37 weeks. Due to the lower concentration of the parameters in the leachate, the pH value was also low (6.35±0.18). Thus, after 17 weeks of the reactor operation, sodium carbonate was also dosed (2 g L^{-1} Na_2CO_3) into the feed to increase the pH and alkalinity. The pH was controlled between 7.5 to 8.5 to ensure that the conditions in the reactor were optimal for nitrification. After 25 weeks of operation of the reactor there was still no denitrification in the second section and methanol was added. Diluted methanol was dosed intermittently (72 mL day^{-1}) at a concentration of 15.8 g L^{-1} into the denitrification stage, with the aim to add sufficient carbon source for denitrification to be complete.

Sampling and analysis: The leachate was collected monthly and was analysed every time when a fresh batch was obtained from the waste site, for the following parameters: ammonia nitrogen (NH_4-N), total Kjeldahl nitrogen (TKN), COD, nitrate, nitrite, alkalinity, phosphate, total suspended solids, conductivity and pH. Samples were collected from both effluents at the end of the nitrification and denitrification stages. The pH of the influent, effluents and the volume throughput were determined on a daily basis, while other parameters such as: ammonium nitrogen (NH_4-N), Total Kjeldahl Nitrogen (TKN), COD, nitrate, nitrite, alkalinity, phosphate and suspended solids were analysed on a weekly basis. The NH_4-N, TKN, COD, nitrite, alkalinity, suspended solids and phosphate were analysed according to standard methods (APHA *et al*., 1998) and the nitrate was analysed according to the salicylate method (Miller and Widermann, 1955).

RESULTS AND DISCUSSION

Leachate quality: For period 1 the average concentrations for the various parameters were about twice those of period 2. In all cases, the NH_3^+ and COD were higher than the required limit of 3 mg N L^{-1} of NH_3^+ and 75 mg L^{-1} of COD set by the Department of Water Affairs and Forestry (DWAF, 1999). The relatively high pH values (8.89±0.13) for period 1 were a result of the high average alkalinity (3456±389 mg L^{-1}, as $CaCO_3$) in the leachate samples. This is typical of old landfill leachate. In period 2 the average pH value was 6.35±0.18 while the average alkalinity was 784±29 mg L^{-1}, as $CaCO_3$ (Table 1).

The total rainfall at Weltevreden sanitary landfill site during the first three months of period 1 was 231 mm and for period 2, was 506 mm. During the last months of period 1 the landfill received heavy rainfall of about 148 mm, which could have an effect on the quality of leachate collected after these months. However, the concentrations of different parameters in the leachate remained lower for the rest of the experimental period 2, although the rainfall decreased. Due to the decrease of rainfall in period 2, it can be assumed that rainfall had no influence on the quality of leachate.

Nitrification efficiency: The decrease in ammonia after the first week of the reactors' operation was noticeable. The ammonia-nitrogen values remained relatively low from the first Week until the tenth week (Fig. 2). The ammonia-nitrogen decreased from an average of 450±20 mg N L^{-1} to an average of 30±3 mg N L^{-1}, which showed an ammonia removal of 84%. This suggest that complete oxidation of ammonia took place. After 10 weeks of

Table 1: Average composition of influent leachate

	Period 1			Period 2		
Parameters	Min.	Max.	Mean	Min.	Max.	Mean
pH-value	8.62	9.1	8.89	6.22	6.7	6.35
Ammonia-N (mg L^{-1})	444	482	450	158	239	200
TKN (mg L^{-1})	450	500	500	163	270	250
COD (mg L^{-1})	1300	1610	1600	400	904	500
Alkalinity (mg $CaCO_3$ L^{-1})	1742	3798	3456	380	811	784
Nitrate-N (mg L^{-1})	0.01	2.0	2.0	1.00	10.00	3.0
Nitrite-N (mg L^{-1})	0.01	0.01	0.01	0.01	1.0	0.01
Phosphate (mg P L^{-1})	2.0	10.0	5.0	12.0	32	10.0
Conductivity (mS m^{-1})	12.5	13.2	13.3	8.54	9.48	9.36
Suspended solid (mg L^{-1})	10.0	30.0	20.2	138	228	118

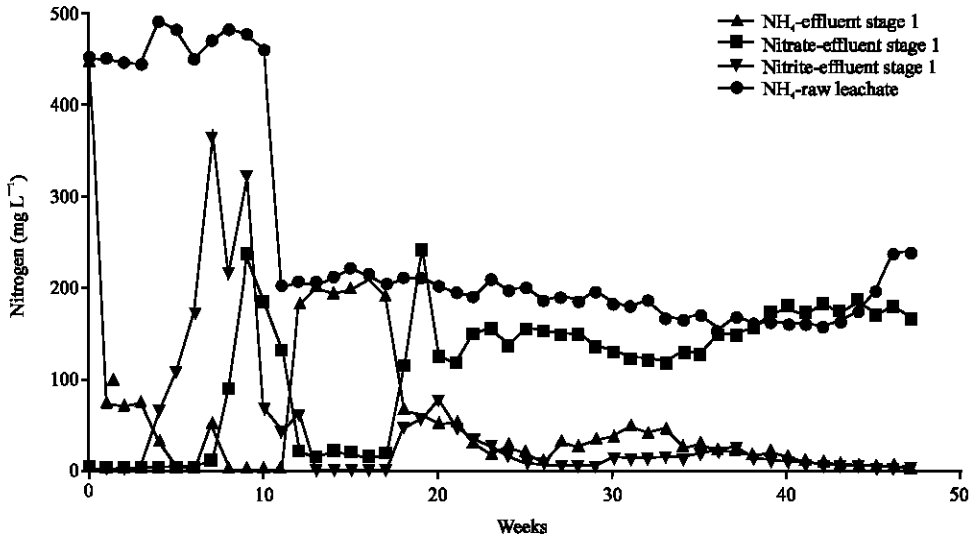

Fig. 2: Performance of the biofilter after Stage 1 with regards to ammonia, nitrite and nitrate-nitrogen

operation the quality of the leachate changed, whereafter the concentration of ammonia in the leachate had an average of 200±10 mg NH_3-N L^{-1}. This sudden change in the composition of the leachate caused an upset of the biological reactor and complete nitrification ceased between weeks 11-18. The ceasing of complete nitrification was probably due to the lower alkalinity in the raw leachate and the lower pH value between weeks 10 to 19. To be able to explain this ceasing, the pH and alkalinity values were compared. For the first 10 weeks, the average pH value was 8.89±0.13 and alkalinity was 3456±389 mg $CaCO_3$ L^{-1}, which changed to an average pH value of 6.35±0.18 and alkalinity of 784±29 mg $CaCO_3$ L^{-1} after 10 weeks. Similar observations were also noted by Brond and Sund (1994) who stated that oxidation of ammonia was inhibited at low pH values. To increase the alkalinity, 2 g L^{-1} of Na_2CO_3 was added to the raw leachate after week 17 and the pH was maintained between 7.5 and 8.5. Thereafter nitrification improved remarkably. The ammonia in the effluent stabilized at an average of 2.0±1.0 mg N L^{-1}. Thus, the average ammonia removal was 99%.

As showed in Fig. 2, initially the nitrite and nitrate concentrations fluctuated for the first 10 weeks, because the quality of the leachate changed and thereafter an increase in nitrite and nitrate concentrations ware noted. The high nitrite concentration (365 mg N L^{-1}) was beneficial because it shortened the denitrification process, which implied that less carbon would be required in the second stage (Alleman, 1985). The nitrate increased to a concentration of 235 mg N L^{-1} after nine weeks, while the ammonia concentration decreased to less than 1.0 mg N L^{-1}.

The reactor stabilised at week 36 until the end of the reactor's operation with nitrate concentration of 136.33±13.44 mg N L^{-1}. Ammonia concentration continued to decrease to an average of 2.0±1.0 mg N L^{-1} at the end of Stage 1, which was acceptable according to the general limit of 3 mg N L^{-1} (DWAF, 1999), while nitrite also decreased to an average of 4.0±1.0 mg N L^{-1} and nitrate concentration was at the steady state of an average 168±12 mg N L^{-1}.

Denitrification efficiency: During the first 25 weeks no external carbon was added to evaluate the ability of the plant to utilize energy obtained from a further reduction of COD and a possible utilisation of energy released due to endogenous respiration by organisms attached to the media in the second stage. Figure 3 and 4 shows the COD concentrations, nitrite and nitrate concentrations at the end of Stages 1 and 2, respectively. No denitrification took place during this stage (Fig. 4).

During the first 24 weeks, the growth (heterotrophic) was very low in Stage 2, because of the non-biodegradable COD. The performance of the COD removal in Stage 2 was also monitored as indicated in Fig. 3. During the first 4 weeks the COD concentration decreased from an average concentration of 1015±364 mg O_2 L^{-1} from influent in Stage 1 to 721±10 mg O_2 L^{-1} at the effluent of Stage 2. The COD concentration in the effluent of Stage 2 increased again between weeks 7 and 12 to an average concentration of 1248±223 mg O_2 L^{-1} due to an increase in the influent from Stage 1.

The COD concentration in the influent from Stage 1 into Stage 2 decreased to an average concentration of 345±26 mg O_2 L^{-1}, which also caused a decrease in the

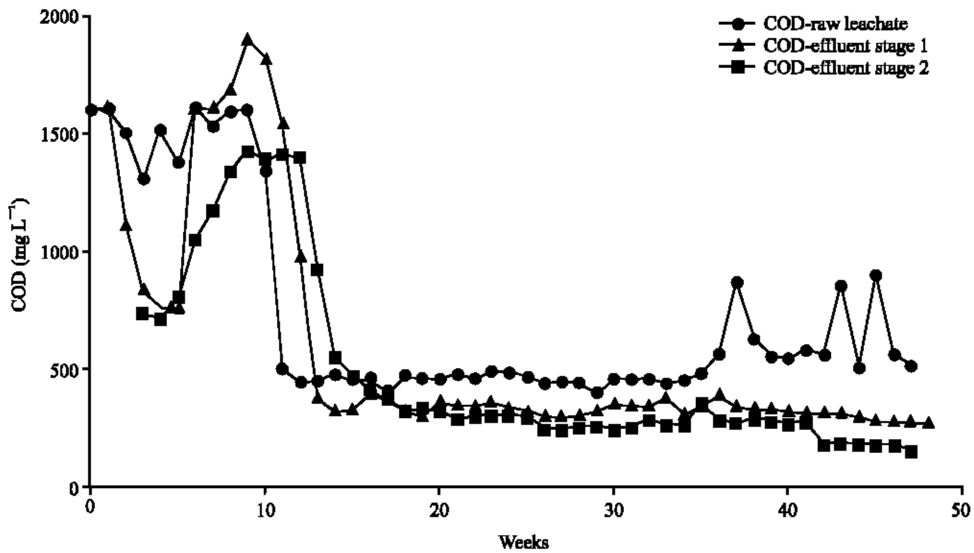

Fig. 3: Performance of the biological filter with regards to COD removal

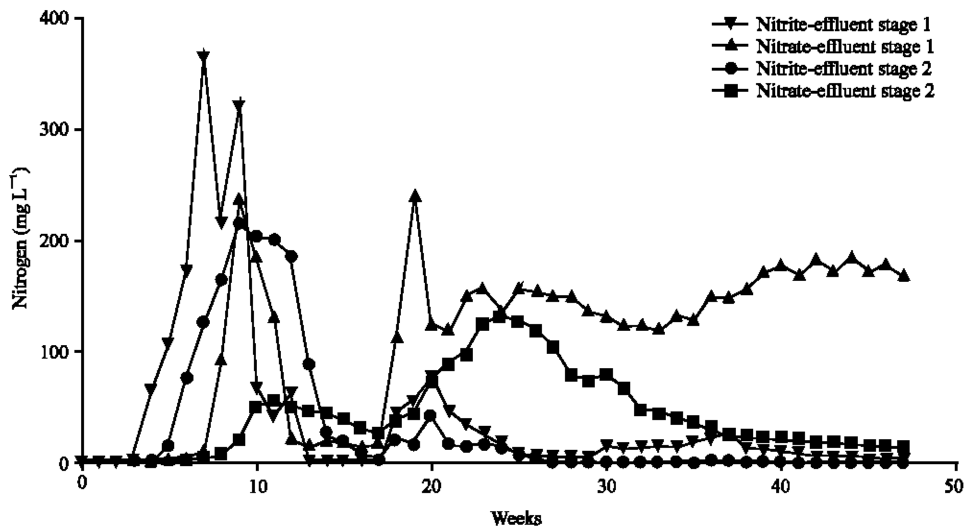

Fig. 4: Performance of the biological filter with regards to nitrite and nitrate removal after stages 1 and 2, respectively

effluent from Stage 2 to an average concentration of 312±16 mg O_2 L^{-1} onwards. The results showed that COD/N ratio of 0.48 in the leachate was usually too low to allow denitrification of such a high nitrate concentration using the biodegradable COD in effluent from Stage 1. A sufficient COD/N ratio of at least 3.7 was required to complete the denitrification reaction (Carrera *et al*., 2003).

From week 25 onwards methanol was added (15.8 g L^{-1}) as an additional source of carbon into Stage 2. According to the literature, biological denitrification is known to occur through heterotrophic bacteria using an available carbon source. Methanol is widely used and Water Pollution Control Federation (WPCF, 1983) has shown that, in general, a ratio of methanol:nitrate-nitrogen of 3:1 should enable complete denitrification.

After the addition of methanol, the denitrification process started and nitrate reduction increased immediately. From week 37 onwards the process progressed steadily and the average nitrate concentration in the effluent from Stage 2 was 19±3 mg N L^{-1} and for the last 3 weeks, it was 14±1 mg N L^{-1}, while the nitrite was 0.2±0.2 mg L^{-1}. The nitrate and nitrite concentration at the end of the reactor's operation was acceptable according to the general limit of 15 mg N L^{-1} from set by DWAF (1999). The denitrification process took place after the addition of methanol and ratio of methanol/nitrate-nitrogen was 1.23:1 which was lower than the theoretical ratio of 3:1 by WPCF (1983).

Unfortunately 64% of the COD seemed to be non-biodegradable and pass out in the effluent. The results were not acceptable according to the limit of 75 mg L^{-1} COD set by DWAF (DWAF, 1998) for the receiving water bodies.

CONCLUSION

The two-stage biological filter with a plastic medium proved to be effective in removing ammonia-nitrogen from sanitary landfill leachate. The ammonia-nitrogen was 2±2 mg N L^{-1} at the end of the trial, which was within the specifications that are considered biologically suitable for fish and other aquatic life. The results of this study demonstrated that nitrification and denitrification did occur in the biological filter.

The COD removal was very poor and the results have shown that only 36% was removed at the end of the trial, reasoning that 64% was non-biodegradable. As the COD concentration in the effluent was still too high, it is recommended that other processes be investigated to reduce the COD in the leachate.

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