Laboratory Scale and Pilot Study of the Treatment of Municipal Landfill Leachate

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The aim of the study was to investigate the use of the biological treatment with activated sludge (with a native and specially selected mixed culture of microorganisms) coupled with physical and chemical adsorption on activated carbon (GAC) for leachate from municipal landfills. The major goal was to evaluate the process performance and determine the nitrification and denitrification efficiency and reduction of organic pollutants. With the biological process good and stable nitrification efficiency of up to 99 % was achieved, while the organic carbon removal and denitrification efficiency was quite low (30 % of COD and N_{tot} reduction on average). With adsorption on activated carbon, carbonaceous removal efficiency was quite good and stable up to 75 %. We can conclude that the combination of the biological treatment followed by adsorption on activated carbon can meet the legal requirements for treated effluent quality.

Key words:
Landfill leachate, nitrification, denitrification, activated carbon adsorption, selected microorganisms

Introduction

Nowadays landfill leachate is often treated together with municipal sewage in municipal treatment plants. However, stricter regulations for nitrogen discharge and the growing concern over the potential effect of recalcitrant leachate constituents on the biological treatment stage, have led to increased demands for the separate treatment of landfill leachate. The methods that have been studied and employed for leachate treatment include suspended growth systems such as aerated lagoons, the conventional activated sludge process, and sequencing batch reactors.[1,2] As these methods have been found to suffer from different limitations, there has been a continuing search for other alternatives.

Leachate quality is primarily a function of landfill age and the degree of waste stabilisation. However, the characteristics of leachate are also affected by many site specifics such as waste composition, moisture availability and climate. In young landfills, containing large amounts of readily biodegradable organic matter, rapid anaerobic fermentation (acidogenic phase) of this matter takes place, resulting in volatile fatty acids. As a landfill matures it enters in the methanogenic phase, and volatile fatty acids are converted to biogas i.e. carbon dioxide and methane. Refractory compounds dominate the content of organic matter in the leachate.

Municipal landfill leachate contains a high concentration of organic pollution expressed as COD, TOC and BOD, a high concentration of nitrogen compounds mostly in reduced form as TKN and NH_4–N as well as chloroorganics (expressed as AOX), and a high concentration of inorganic salts (expressed as conductivity). A wide variety of heavy metals as well as inorganic and organic compounds can be also found in leachate. However, the concentration of these constituents is generally on the order of micrograms per litre. It has to be noted, that it is impossible to measure all chemicals due to technical and economical reasons. Several studies confirmed the potential toxicological risk and accumulation potential of untreated landfill leachate.[1,2]

The quality of leachate directly affects different leachate treatment alternatives. Because of the variability of leachate quality, the prediction of leachate characteristics and treatability as a function of time has been quite difficult. As a result, neither biological treatment, nor physical/chemical treatment processes separately are able to achieve high treatment efficiencies.[2–6] A combination of both types of treatment is the most effective process train for the treatment of leachate.

The aim of the study was to investigate the possibility of using a pre-denitrification system with activated sludge in suspended form for biological nitrogen removal from landfill leachate at a

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landfill under realistic conditions (variations in temperature and leachate composition) with the goal of obtaining data that could be used in a full-scale process.

In view of the results from previous tests and studies,[5] our company has decided to perform a pilot plant study on continuous wastewater treatment with activated sludge, which was suggested as the most appropriate. The aim of the tests was to investigate and confirm the suggested technology in biological ammonia and COD removal from landfill leachate and the suitability of an external carbon source in the denitrification process. After biological treatment the leachate is further treated in an activated carbon adsorber, elevating the water quality of the final leachate effluent to a permissible values for safe discharge. Such a combined biological and physical/chemical treatment was found to be highly efficient and may offer an extractive alternative in dealing with the high variability of pollutant content in landfill leachate.[2,4,6]

Materials and methods

Tests were carried out between July 2000 and August 2001 and covered all seasonal fluctuations in weather changes. The entire project was divided into three different stages:

1) a test on a biological treatment pilot plant located at a landfill,

2) laboratory tests of physical/chemical adsorption on activated carbon, and

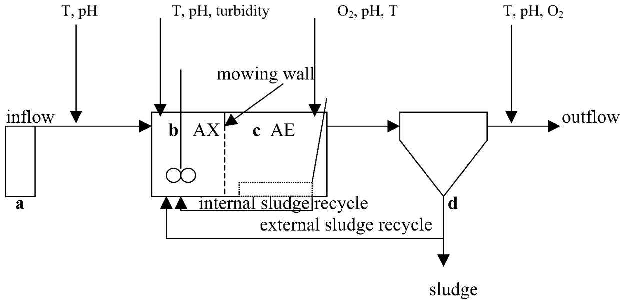
3) laboratory tests of a simulation pilot plant operation with a specially selected mixed culture of micro-organisms as a parallel test.

Pilot units (experimental set-up and operating conditions)

The municipal landfill leachate study was performed on a semi-industrial wastewater pilot plant for biological treatment with single sludge in suspended form as a continuous process (Fig. 1).

The total volume of the plant was 5.3 m^3. The treatment plant consisted of a 1.5 m^3 equalisation basin, two small basins (0.3 m^3) for taking inflow and outflow samples, a 2.6 m^3 reaction basin, and a 0.9 m^3 settling tank. The reaction tank was equipped with a stirrer, aeration system and, wall, that could be shifted to different positions. The system conditions were controlled with on-line sensors for measuring flow, oxygen concentration, pH, temperature and turbidity.

The pilot plant tests were divided into four different phases:



Legend: a – equalization basin, b – AX anoxic part with mixing device, c – aerated part of reaction basin, d – settling basin

Fig. 1 – *Plan of the pilot plant for biological leachate treatment*

– Phase 0 – system adaptation: the adaptation period was approximately one month.

– Phase 1 – nitrification:

– Phase 2 – nitrification and denitrification: the total volume of the reaction basin was split into two parts with a moving wall at a ratio AX:AE = 45 : 55 and connected with the internal sludge recycle.

– Phase 3 – nitrification and denitrification with external carbon sources: acetic acid was chosen to lower the high pH in the process.

Analyses and measurements were performed according to standard methods[7] and ISO methods.[8,9]

Laboratory tests of activated carbon adsorption

The activated carbon test was carried out in laboratory batch adsorber equipped with a mechanical stirrer and constant temperature regulation device.[10] The Freundlich adsorption isotherm was determined with "Carbotech" activated carbon in granular form. The experimental conditions for determining isotherm values were pH 7.0 and adsorption material dosage ranging from 1 to 5 g/l, a temperature of 20 °C and, according to the adsorption kinetics results, a pseudo-equilibrium time of 24 hours. Test efficiency was determined by COD, TOC and colour according to prescribed standard analytical methods.[7]

Laboratory tests as parallel tests with a specially selected mixed culture of micro-organisms

Three different tests of biological degradability were carried out at the same time as the batch tests:

– (A) an aerobic degradation test in the presence of native culture;

– (B) an aerobic degradation test in the presence of activated sludge from the pilot plant; and

– (C) an aerobic degradation test in the presence of specially selected mixed culture nitrification and denitrification bacteria composed of seven seeds of bacteria; four autotrophic and three heterotrophic.

All tests were performed after previous adaptation to the test procedure conditions (T = 18–22 °C, 3–6 mgO_2/l, and X = 5.5–6 g/l in $V_{reactors}$ = 5 l). The step test results were confirmed by the continuous process with the same phase as in the pilot plant.

Results and discussion

Pilot plant tests

Characterisation of the leachate used in the treatability tests is given in Table 1.

BOD_5 and COD concentration appear to remain low (less than 1500 mg/l) for the duration of experiments, most likely due to dilution and stimulation of methanogenesis in the landfill. The BOD_5/COD relation is between 0.2 and 0.3, which is much lower than in municipal wastewater.[2,4] Concentrations of COD and NH_4–N lay more or less in the same range. The ratio between N and C is very inappropriate because the NH_4–N concentration is much higher than the concentration of easily degradable carbon.[2,4,6]

A review of the analyses results for the entire duration of the pilot treatment plant test showed an average of 30 % COD (present in leachate – endogenous COD) reduction. Such a reduction was detected in all three phases, irrespective of the changing conditions in the system or inflow loading (Fig. 2). The COD removal efficiency cannot be fully attributed to either carbon aerobic elimination or to carbon elimination for the requirements of denitrification.

On the basis of the results achieved, the chemical complexity of leachate COD for biological processes can be confirmed as well as the fact that the biological process with activated sludge has insufficient ability for a major decomposition of the complex compounds present in the leachate. The great complexity of organic compounds also caused a high selection of sludge for the autotrophic process or for nitrification of ammonia nitrogen.

The high autotrophic process was also confirmed by the concentration of nitrate nitrogen in the outflow, which lies in the same range as ammonia and organic nitrogen concentration in the inflow. The results show that there was almost no denitrification process present. Sludge is not only highly active for the autotrophic process, but has a large capacity for nitrification at greater hydraulic loads as well. Nitrification remains stable even at very low temperatures (lower than 10 °C) in winter-time (Fig. 3, Table 2).[11,12]

Table 1 – *Composition of leachate in inflow during testing*

	Max.	Min.	Average.
pH	8.6	7.0	7.7
COD (mg/l)	2600	400	926
BOD_5 (mg/l)	950	100	230
NH_4–N (mg/l)	800	140	380
NO_3–N (mg/l)	4.6	0.3	3.0
NO_2–N (mg/l)	1	0.01	0.03
N_{tot} (mg/l)	900	180	400
SO_4^{2-} (mg/l)	365	172	273
B (mg/l)	28	9	19
Cl^- (mg/l)	1100	452	760

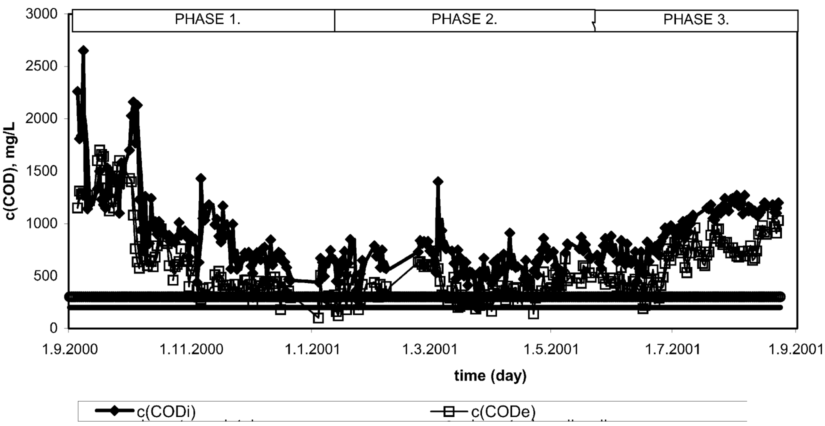


Fig. 2 – *COD in influent and effluent and COD removal efficiency*

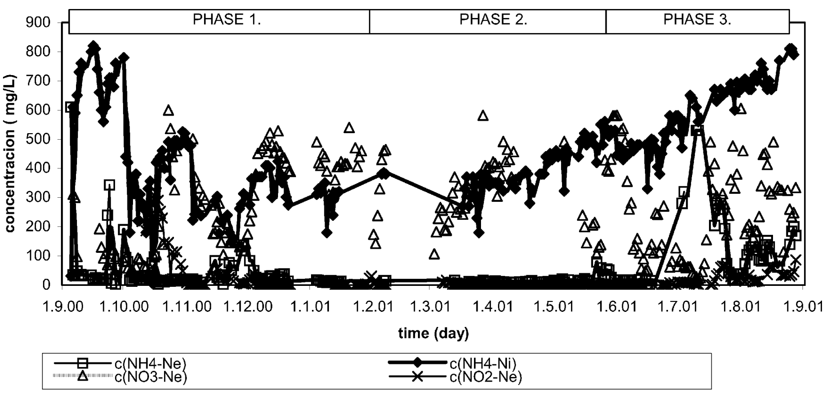


Fig. 3 – *Concentration of NH_4–N in influent and effluent and NO_3–N and NO_2–N in effluent during testing*

Table 2 – *Results for different test phases of the pilot plant leachate treatment*

	Phase 1	Phase 2	Phase 3
Q (l/h)	60/120/180	60/90/120	120/70/60
COD reduction (%)	30	30	30
Nitrification (%)	92	90	85
Denitrification (%)	0	min.	30-(70)
SVI (ml/g)	150/750	300	350
X (g/l)	4,0	4.8	9,0
Bv,c (kgCOD/m^{3}dan)	changing	0.2	0.2
Bv,$_N$ (kgNH_4–N/m^{3}dan)	0.5	0.2	0.5
HRT (h)	45/22/15	45/30/22	22/40/45

The results of volumetric loading (Fig. 4) show two levels, depending on the leachate inflow. Because the concentration in the outflow did not change, it can be concluded that the volume of the plant is not a limiting factor. The optimal volumetric loading related to nitrification is up to 0.5 kg NH_4–N/m^3 per day and is rather low than high according to literature reviews.[11,13,14]

In some cases high concentrations of nitrite in the outflow could be detected due to the presence of inhibitory compounds or due to inhibition caused by high ammonia concentrations in the inflow.[15] Although high concentrations of nitrite were detected at the start, possibly caused by the adaptation of the activated sludge or by the present inhibitors, the nitrite concentration was almost always below 0.01 mg/l, which indicates a stable process. A considerable disturbance in the process appeared at the end of June. The disturbance was accompanied by increased nitrite concentration. The results of further tests show that there was no inhibition in the system, but that it was run to the very limit of stability and was thus destabilised even during the smallest disturbances.

Due to the complex structures of organic compounds in leachate, the denitrification process may be implemented only with the addition of external carbon – methanol or acetic acid. Acetic acid was chosen because of the high buffer capacity of the system, with pH even exceeding 8. Under those conditions inhibition could be expected.[11,15]

The quantity of the acid added gradually rose from C:N = 1 : 1 to 3.5 : 1. After about three weeks, an increase in nitrification was observed (Fig. 3). The quantity of the added external carbon was probably too high, and nitrification becomes unstable. The reason for this may be due to the altered sludge composition. The numbers of autotrophic microorganisms became higher due to the higher growth rate. On the other hand, the numbers of heterotrophic microorganisms became lower (with a lower growth rate) due to regular sludge removal.[11]

The lowest concentration of N_{tot} in the outflow was less than 100 mg/l during the stable period of the denitrification process.

Biomass (Fig.5) showed great differences in its concentration, characteristics and composition. The rise in sludge concentration was the highest during the phase when acetic acid was being added. In certain periods deteriorated settling properties appeared due to the altered sludge structure, when a larger quantity of filamentous structures of microorganisms began to appear, causing sludge washout (SVI between 50 and 300 ml/g).[4,13] The reasons for

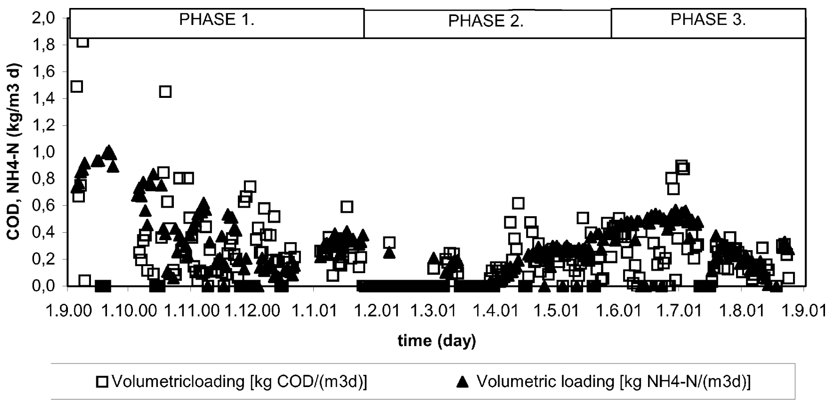


Fig. 4 – *Volumetric loading for COD and NH_4–N during testing*

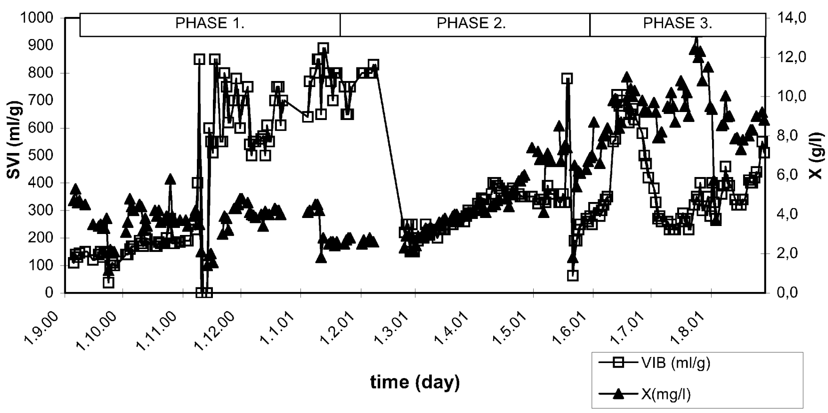


Fig. 5 – *Changes in sludge concentration and sludge volume index during the pilot plant experiment*

this cannot be explained either with changes in inflow, inflow composition or differences in sludge age. With a lower inflow and hydraulic rate the process could not be stabilised, and thus $FeCl_3$ began to be added.[15]

The results of COD reduction (Fig. 2) and nitrification (Fig. 3), showed almost identical values with 4.5 g/l and 8 g/l of activated sludge concentration. As the process efficiency showed no major advantages, the system may operate with a lower biomass concentration (4.5–6.5 mg/l).

Laboratory tests of activated carbon adsorption

In view of the fact that during the all tests the COD removal efficiency was quite low and remained unchanged, despite changes in operating conditions, the activated carbon adsorption test was performed to improve organic pollution mitigation. GAC is considered one of the most effective adsorbent, especially for those substances containing refractory organic compounds that persist in the environment and resist biodegradation.

The results of the batch activated carbon adsorption tests present a good option for removing part of the COD in raw leachate and in the effluent after biological treatment. The effect achieved depends on COD concentration; the greater the COD in the beginning, the greater the effect in the end. Competition phenomena give rise to different adsorption behaviour of COD in complex multicomponent mixtures like leachate or effluents. It can be concluded that: COD reduction after 120 min of contact time with 3.0 g/l of activated carbon is between 540–460 mg/l for the inflow in the pilot plant (untreated leachate) and between 500–270 mg/l for effluent (biological treated leachate). With certain optimisation of the test conditions a reduction in the initial COD of up to 75 % in the inflow and the outflow was reached, the residual COD concentration lying between 270 and 120 mg/l – what could be expected as limiting concentration. Those data also show highly unfavourable tendency at low COD concentration ranges. The adsorption capacity at the residual (equilibrium) concentration C_e = 120 mg/l is 0.22 mgCOD/mgGAC according to the Freundlich isotherm. Test results are presented in Table 3 and Fig. 6.

Laboratory tests as parallel tests with a specially selected mixed culture of micro organisms

A parallel laboratory test of the biological degradability of landfill leachate with a specially selected mixed culture of micro organisms was performed for seven weeks at the Faculty of Food and Biotechnology in Zagreb. The main purpose of the test was to compare the activity of different micro organisms in the process of oxidation, nitrification and denitrification or process conditions and, consequently, better settling properties of activated sludge i.e. a specially selected mixed culture.

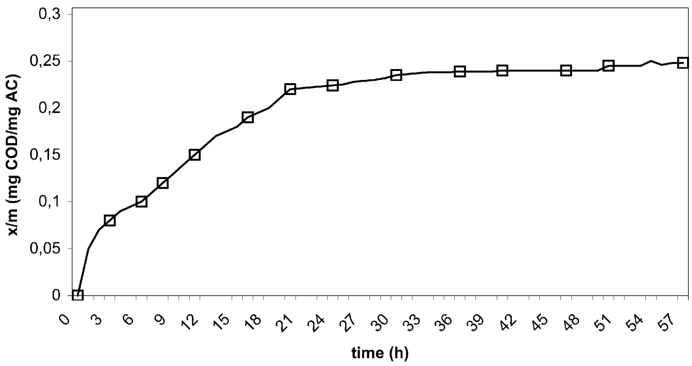


Fig. 6 – *Effect of contact time on the removal of COD from leachate in batch adsorber (with 3 g/l GAC)*

Table 3 – *Results of activated carbon tests*

Contact time (min)	Reduction COD (%)	Reduction DOC (%)	Reduction AOX (%)
30	3–9	/	/
60	8–26	/	/
90	9–40	/	/
120	15–50	8–40	/
1440 (24 h)	up to 75	up to 65	30

Due to the different scale of testing (laboratory and pilot plant scale), in laboratory conditions, in 2L bioreactors, the activity of three different types of biomasses on original wastewater was tested: native culture, activated sludge from pilot plant and selected mixed culture. The results[16] of the repeated tests revealed that a native culture gave similar results with leachate quality changes: COD reduction in all repetitions was 100 mgO_2/l. With activated sludge from the pilot plant COD reduction was slightly smaller, the achieved effect being 30–45 %. All reductions, however, may be caused by a native culture, while only high activity for the autotrophic nitrification process may be attributed to the activated sludge. The best results were achieved with a selected mixed culture, i.e. 47–67 % of COD reduction. With each repeated test a better effect was achieved, irrespective of the fact that the composition of leachate was changing, i.e. worsening. The selected mixed culture shows the ability of heterotrophic nitrification and biological oxidation with a greater effect than the activated sludge. The pH value was also monitored during testing, and lay in a range between 7.8 and 8.4. These values, however, are not optimum for the heterotrophic nitrification process.[3]

The nitrification test with a selected mixed culture as a continuation treatment of the outflow from previous tests, was performed next. We sought to determine the ability of selected mixed culture bacteria for the denitrification process and whether a reduction or incorporation of organic substances had taken place.

The results showed that in the denitrification process COD and therefore NO_3–N reduction was very small. This confirms that COD reduction in nitrification was caused by biodegradation and not by accumulation in the cell structure. The COD and NO_3–N reduction during testing shows, that the relation between C and N must be between 1.5 and 2.

Additionally, a test of raw leachate used as an external carbon source was performed, during which raw leachate was added at a ratio of 3:1 directly into the system after denitrification was accomplished. A selected mixed culture shows great ability to use raw leachate as an external carbon source.

For a better comparison, the step test results were further confirmed by the continuation process tests with the same phase sequences as in the pilot plant test and with the same microorganisms as used in the step tests.

The results in Table 4 show that the selected mixed culture has the ability to produce a slightly better COD reduction. It also has the ability to use carbon from leachate for partial denitrification. The used biomass displays good settling properties. Retention times for complete process bio-oxidation, nitrification and denitrification were a little longer than those obtained during pilot plant testing and the operation processes in the system were more stable.

Since the volumes of laboratory bioreactors and pilot plant are different (laboratory bioreactors 2L and pilot plant 2,6 m^3), it is not possible to compare the achieved results accurately, but the comments in Table 5 are the indication of the activity of selected mixed culture in comparison with conventional activated sludge.

Conclusions

This project demonstrates that nitrification could be achieved with a pre-denitrification activated sludge system yielding high rates of ammonia

Table 4 – *Comparison of results for the continuous process for all three samples*

Sample no.	I		II		III	
parameter	inflow	outflow	inflow	outflow	inflow	outflow
COD (mg/l)	620	255	487	268	780	250
NH_4–N (mg/l)	285	≤1	386	≤1	415	≤1
NO_3–N (mg/l)	5	268	6	389	9	389
pH	7.6	8.2	7.88	8.4	7.9	8.0
HRT (h)	24		28		28	
X (g/l)	5.5		5.5		6	

Table 5 – *Comparison results of all performed tests*

	Laboratory test	Pilot test
Reduction of COD	outflow COD concentration average 250 mg/L, with active carbon adsorption less than 200 mg/L	outflow COD concentration average 500 mg/L, with active carbon adsorption less than 270 mg/L
Nitrification	complete	complete
Denitrification	partly with COD from leachate, complete with addition of acetic acid or methanol	minimal from COD from leachate, partly with acetic acid addition
Process parameters	for 1/3 longer HRT, lower oxygen concentration and biomass needed, biomass (sludge) is stable	shorter HRT, higher oxygen concentration and biomass, biomass with bad settling properties, wash out appears

removal from municipal landfill leachate. The results show that nitrification is feasible at temperature as low as 10 °C. Permit-discharged limits for outflow into the sewer system can be reached with tested technology and good stability for parameter NH_4–N, i.e. c(NH_4–N) < 50 mg/l. However, sludge settle-ability was poor (SVI was between 300 and 750 ml/g) and the reduction of organic substances reached only 30 % on average. The low COD removal achieved corresponds to the high COD to BOD ratio of the untreated leachate. The poor biodegradability of the organic content is typical for leachate from landfills in the methanogenic phase, for which most biodegradable organic matter is already converted to biogas, already in the landfill. The greatest recommended inflow to the pilot plant, where a stable nitrification and denitrification process can be expected, is about 60 l/h or a retention time of 44 hours. Maximal removal rates were 0.2 kgCOD/m^3 per day and 0.5 kgNH_4–N/m^3 per day. A similar system can be implemented on the basis of laboratory tests with selected micro organisms and even better efficiency and sludge settle-ability could be expected.

The dosage of external carbon source (methanol, acetic acid or raw leachate) to the anoxic stage should certain be the most critical parameter to control in a full scale process, especially when the nitrogen content in leachate show great variations. Too low a dose in relation to the nitrate to be denitrified immediately results in decreased nitrogen removal, while an over dosage results in sludge bulking, which leads in destabilisation of the nitrification process.

A series of tests carried out in batch adsorber with activated carbon show that at 24 h of contact time up to 75 % of COD pollutants can be adsorbed and provide an effluent that meets law limits for discharge into sewage system. Discharge low limits to surface water are not met with tested treatment technologies, and additional post-treatment technologies including reverse osmosis or ozone oxidation, are required.

The results of this work show that the applied process requires a very exact process and operating conditions. Treatment prior to the biological process should be considered due to the great changes in composition and hydraulic rate. The results also provided an important insight into the prediction of future trends in leachate quality and the design and operation of leachate management facilities.

List of Symbols

AE – aerobic stage

AOX – adsorbable organic halogenides, mg l^{-1}

AX – anoxic stage

BOD – biological oxygen demand, mgO_2 l^{-1}

Bv,c – Volumetric Loading according to COD, kgCOD m^{-3} dan^{-1}

$Bv_{,N}$ – Volumetric Loading according to NH_4–N, kgNH_4–N m^{-3} dan^{-1}

COD – chemical oxygen demand, mgO_2 l^{-1}

C_e – equilibrium COD concentration, mg l^{-1}

DOC – dissolved organic carbon, mg l^{-1}

GAC – Granular Activated Carbon

HRT – Hydraulic Retention Time, h

NH_4–N – concentration of ammonia nitrogen, mgN l^{-1}

NO_2–N – concentration of nitrite nitrogen, mgN l^{-1}

NO_3–N – concentration of nitrate nitrogen, mgN l^{-1}

Q – inflow, l h^{-1}

SVI – Sludge Volume Index, ml g^{-1}

X – Sludge Concentration, g ml^{-1}

x/m – Adsorptive Capacity, mg COD g^{-1} AC

i – influent

e – effluent

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