

Combined Treatment of Leachate from Sanitary Landfill and Municipal Wastewater by Activated Sludge

A. Del Borghi, L. Binaghi, A. Converti*, and M. Del Borghi

Department of Chemical and Process Engineering “G.B. Bonino”,
Genoa University, Via Opera Pia 15, 16145, Genoa, Italy

*E-mail: converti@unige.it

Original scientific paper

Received: October 9, 2002

Accepted: June 1, 2003

Two leachates from sanitary landfill, characterized by different biodegradability, were mixed with municipal wastewater in different proportions and aerobically treated by activated sludge. The overall removals of chemical oxygen demand (COD) and ammonia as well as the fraction of hardly-biodegradable substances were investigated to select the best operating conditions for the simultaneous removal of these pollutants. Preliminary batch tests were carried out to point out the optimum leachate/mixture volume ratio ($\Psi_{L/M}$) consistent with the biological treatment (0.10). Using this proportion, the performance of a bench-scale activated sludge system was checked under continuous working conditions at different hydraulic retention times (τ_{HRTs}), namely 2, 4 and 8 h. The removal yields of COD and ammonia (Y_{COD} and $Y_{NH_4^+}$) were studied either during the start-up or after reaching stationary conditions. The results obtained under steady-state conditions demonstrated that the organic substances were effectively removed ($Y_{COD} > 0.65$) at $\tau_{HRT} = 8$ h, while the best results were obtained for ammonia at $\tau_{HRT} = 2$ h. Sludges produced by the continuous system were then used in batch activity tests to check the degradation improvement consequent to their acclimation to leachate/mixture volume ratios in the range 0.05–0.2. The best performance was obtained with sludge produced at $\Psi_{L/M} \leq 0.15$ and $\tau_{HRT} = 4$ h, that ensured, for both COD and NH_4^+ , removal yields of $Y = 0.67$ – 0.76 and removal rates of $r = 103$ – 130 and 35 – 63 $mg\ l^{-1}\ h^{-1}$, respectively.

Keywords:

Activated sludges, COD and ammonia removals, aerobic treatment, kinetics, leachate from sanitary landfill

Introduction

The control of leachate produced in municipal solid wastes (MSWs) landfill is one of the most difficult problems to be solved during landfill management due to the leachate's high content in hardly-biodegradable organics as well as inorganics. Chemical composition of leachate is influenced by several factors, including waste composition, landfill operation, and weather. In addition, it varies in the long run because of depletion of various MSW components as well as change in chemical environment (e.g. redox-potential, pH, and ionic strength).¹ The mass concentrations of a number of inorganic pollutants (ammonia, chlorides, sulphates, some trace elements, etc.) seem to depend mainly on the liquid/solid ratio,^{2–4} while the landfill age appears to be the main factor influencing, both, the chemical (COD) and the biological (BOD) oxygen demands.⁵

The control of leachate is considered to be one of the most urgent environmental issues, because it can leak to the surrounding environment in the absence of adequate control facilities. Because of its high content in refractory and toxic organic and in-

organic pollutants, combinations of physico-chemical and biological methods have been using up to now to treat landfill leachate. However, they are recognized as being scarcely effective and too expensive either in terms of investment or running costs,⁶ therefore alternative collection and treatment methodologies are required.

The combined treatment of leachate along with municipal wastewater could constitute an economically-feasible alternative in which the degradation of organic pollutants would be favoured by dilution and adaptation ability of the activated sludge. Since leachate treatment in municipal wastewater purification plants is expected to reduce the abatement capacity, the traditional plant design standards could be unsuitable in this case. To correctly design a new plant, accurate bench-scale experimentation could then be useful to establish the optimum ratio of wastewater to leachate flowrates as well as to investigate the biodegradation kinetics.

Although many studies were performed on the co-treatment of landfill leachate and domestic sewage,^{7–10} the main scientific and technical problems have not yet been solved. Among these, it would be

necessary to determine the efficiency of the biological treatment of old leachate, of leachate from pre-treated waste, characterized by $\zeta_{\text{BOD}/\text{COD}}$ ratios < 0.3, and of high nitrogen content leachate, as well as to optimize the denitrification stage, for which external source of carbon is usually needed. It has been reported that the organic content of leachate with high starting COD and $\zeta_{\text{BOD}/\text{COD}}$ ratio (0.6–0.8) can be reduced by 90 % or more within a period of 2–5 years,¹¹ while more than 50–100 years are necessary to obtain comparable abatements for ammonia.^{5,12}

The leachate BOD/COD ratio progressively decreases during the landfill life, thereby affecting the efficiency of COD removal by activated sludge. Keenan et al.⁸ reported COD removals ranging from 90 to 98 % for leachates having BOD/COD ratios in the range 0.4–0.8, while no more than 35 % reduction was obtained at $\zeta_{\text{BOD}/\text{COD}} < 0.3$.^{5,13} As a consequence, simple aerobic treatments appear to be suitable for the treatment of leachates produced in young sanitary landfills, whereas those from old landfills would need combination of biological oxidation and physico-chemical treatments.

In this work, old and young leachates from different sections of an urban landfill and municipal wastewater have been mixed in different proportions, and treated in a bench-scale activated sludge unit, in order to select the best operating conditions to perform such a combined purification process in a real-scale plant.

Materials and methods

Leachate/wastewater mixtures preparation

The leachates used in this work came from a municipal sanitary landfill located in Monte Scarpino, a valley close to the urban area of Genoa (Italy). The landfill is divided in two different storing areas: the older part (S1), containing more than $8 \cdot 10^6$ m³ MSW, and the younger one (S2), located in the upper part of the valley, with total capacity of $2 \cdot 10^6$ m³ MSW.¹⁴ Samples of leachates from both areas were taken at the discharging point situated in the lower part of the valley. The mixture employed for tests contained leachates from S1 and S2 in the ratio $\Psi = 9:1$, which was representative of the different leachate productions of the two landfill sections. Tables 1 and 2 list the physico-chemical characteristics of both leachates, which showed a large variability depending on the weather conditions. In order to obtain a relatively constant starting composition, 50 l samples of the same batch of concentrated mixture, having $\gamma_{\text{COD}} = 1278 \pm 20$ mg l⁻¹, $\gamma_{\text{NH}_4^+} = 1020 \pm 18$ mg l⁻¹, volatile suspended solids (γ_{VSS}) =

Table 1 – Physico-chemical characteristics of S1 leachate

Parameter	Unit	Range	Mean
pH		7.8–8.5	8.0
γ_{COD}	mg l ⁻¹	966–1784	1090
γ_{BOD_5}	mg l ⁻¹	110–600	248
γ_{VSS}	mg l ⁻¹	10–190	53
$\gamma_{\text{NH}_4^+}$	mg l ⁻¹	715–1259	947
$\gamma_{\text{NO}_3^-}$	mg l ⁻¹	0.21–4.5	0.75
$\gamma_{\text{NO}_2^-}$	mg l ⁻¹	0.02–0.43	0.15
Electric conductance	mS	4500–8200	6256
γ_{Al}	mg l ⁻¹	0.40–1.4	0.87
γ_{Cd}	mg l ⁻¹	nd	nd
γ_{Cr}	mg l ⁻¹	0.09–0.22	0.84
γ_{Fe}	mg l ⁻¹	4.9–7.7	5.7
γ_{Hg}	mg l ⁻¹	nd	nd
γ_{Ni}	mg l ⁻¹	0.10–0.15	0.14
γ_{Pb}	mg l ⁻¹	nd	nd
γ_{Cu}	mg l ⁻¹	0.01–0.05	0.02
γ_{Zn}	mg l ⁻¹	nd	nd
γ_{Cl}	mg l ⁻¹	923–1390	1145
Aldehydes, γ	mg l ⁻¹	0.02–3.9	1.3
Surfactant, γ	mg l ⁻¹	0.18–0.92	0.47
Mineral oils, γ	mg l ⁻¹	41–146	79
γ_{Fats}	mg l ⁻¹	107–289	160
$\gamma_{\text{SO}_4^{=}}$	mg l ⁻¹	9–25	19
$\gamma_{\text{SO}_3^{=}}$	mg l ⁻¹	0.06–1.2	0.73
$\gamma_{\text{H}_2\text{S}}$	mg l ⁻¹	0.01–0.6	0.04
Total $\gamma_{\text{p,t}}$	mg l ⁻¹	0.48–0.61	0.54

nd: not detected

59 ± 3 mg l⁻¹ and pH 8.0 ± 0.5 , were stored at -20 °C and defrosted before their use.

Either wastewater or sludge used for the combined treatment, came from the Genoa depuration plant of Punta Vagno. The average composition of wastewater was: $\gamma_{\text{COD}} = 200$ – 260 mg l⁻¹, $\gamma_{\text{NH}_4^+} = 40$ – 50 mg l⁻¹ and pH 7.0.

Standard methods were used for COD, NH_4^+ , VSS, NO_2^- , NO_3^- and total Kjeldahl nitrogen determinations.¹⁵ Microbial population grown in the reactors was periodically analysed by qualitative microscopic examination of the sludge.

Experimental set-up and operating conditions

Seven stirred and aerated 5.0 l-glass tanks were seeded with sludge up to starting $\gamma_{\text{VSS}} = 3,500$ mg

Table 2 – Physico-chemical characteristics of S2 leachate

Parameter	Unit	Range	Mean
pH		7.2–8.7	8.0
γ_{COD}	mg l ⁻¹	2525–3879	2974
γ_{BOD_5}	mg l ⁻¹	500–1800	1335
γ_{VSS}	mg l ⁻¹	50–580	119
$\gamma_{\text{NH}_4^+}$	mg l ⁻¹	512–2229	1678
$\gamma_{\text{NO}_3^-}$	mg l ⁻¹	0.10–3.2	0.85
$\gamma_{\text{NO}_2^-}$	mg l ⁻¹	0.01–3.0	0.54
Electric conductance	mS	7140–8180	7730
γ_{Al}	mg l ⁻¹	nd	nd
γ_{Cd}	mg l ⁻¹	0.01–0.1	0.07
γ_{Cr}	mg l ⁻¹	0.20–1.1	0.43
γ_{Fe}	mg l ⁻¹	4.4–22	9.4
γ_{Hg}	mg l ⁻¹	nd	nd
γ_{Ni}	mg l ⁻¹	nd	nd
γ_{Pb}	mg l ⁻¹	0.11–0.89	0.34
γ_{Cu}	mg l ⁻¹	0.03–1.3	0.19
γ_{Zn}	mg l ⁻¹	0.30–1.0	0.44
γ_{Cl}	mg l ⁻¹	244–3144	1286.78
Aldehydes, γ	mg l ⁻¹	0.21–8.0	4.8
Surfactant, γ	mg l ⁻¹	0.17–1.1	0.55
Mineral oils, γ	mg l ⁻¹	3.0–427	149
γ_{Fats}	mg l ⁻¹	4.0–686	220
$\gamma_{\text{SO}_4^{2-}}$	mg l ⁻¹	23–61	46
$\gamma_{\text{SO}_3^{2-}}$	mg l ⁻¹	0.51–3.0	1.4
$\gamma_{\text{H}_2\text{S}}$	mg l ⁻¹	0.01–0.9	0.2
Total P, $\gamma_{\text{p,t}}$	mg l ⁻¹	0.65–0.98	0.83

nd: not detected

l⁻¹ and used in parallel at room temperature (20 ± 1 °C) for batch tests. Samples were taken at constant time intervals and analysed for COD, NH_4^+ , NO_2^- and NO_3^- concentrations. The pH was automatically regulated at 7.5 ± 0.2 by a pH control module Setric G.I., Set 7, provided with a peristaltic pump injecting a stream of 1.0 mol l⁻¹ NaOH solution.

Preliminary batch tests were carried out using fresh sludge coming directly from the municipal wastewater treatment plant and mixtures of leachate and wastewater. The leachate volume ratio in the mixture ($\Psi_{\text{L/M}}$) was increased from 0.01 to 0.20, corresponding to starting γ_{COD} and $\gamma_{\text{NH}_4^+}$ concentrations of 250–450 mg l⁻¹ and 55–240 mg l⁻¹, respectively.

Final batch tests were also performed using acclimated sludge from the bench-scale continuous

activated sludge system and mixtures of leachate and wastewater at $\Psi_{\text{L/M}}$ increasing from 0.05 to 0.20, corresponding to starting γ_{COD} and $\gamma_{\text{NH}_4^+}$ concentrations of 270–450 mg l⁻¹ and 80–240 mg l⁻¹, respectively.

A 2.0 l-aerated tank, equipped with an air pump and connected with a 0.85 l settling tank, was used as an activated sludge unit with sludge recycle for continuous tests. It has been employed to verify the actual possibility of biologically treating leachate/wastewater mixtures under continuous working conditions as well as to obtain acclimated sludge. It was seeded with sludge up to starting $\gamma_{\text{VSS}} = 3,500$ mg l⁻¹ and filled up only with wastewater. The reactor was continuously fed by means of a peristaltic pump (Munipuls 2, Gilson) regulated according to the selected hydraulic retention time (τ_{HRT}), namely 2, 4 and 8 h, corresponding to flow rates decreasing from $Q = 1.0$ to 0.25 l h⁻¹. After 15 working days, during which the reactor was fed only with wastewater, leachate was progressively added to the feed up to $\Psi_{\text{L/M}} = 0.10$ (within one week) until steady-state conditions were achieved. The behaviours of COD, NH_4^+ , VSS, NO_3^- and NO_2^- concentrations versus time were investigated. The age of the sludge during continuous tests was kept at 9.3 d by removing daily a fraction of the reactor content.

Results

Preliminary batch tests

Preliminary batch experiments were carried out to investigate the effect on sludge activity of leachate addition into municipal wastewater treatment plant as well as to determine the maximum volume ratio of leachate in the mixture without provoking valuable microbial activity loss.

Final COD removal was always quite low and remained practically constant (~ 0.45) up to $\Psi_{\text{L/M}} = 0.15$, whereas it strongly decreased to 0.25 at $\Psi_{\text{L/M}} = 0.20$ (Fig. 1). At the same time, ammonia removal was always unsatisfactory, gradually decreasing from 0.17 to 0.09 with increasing the leachate volume ratio in the feed from 0.01 to 0.20. The mean COD removal rate, calculated from the beginning to the end of each run, was hardly affected (68–78 mg l⁻¹ h⁻¹) by the leachate/mixture volume ratio up to 0.15, but fell beyond this value (Fig. 2). Less marked effects were evident for NH_4^+ removal rate, that proved to be scarcely influenced by the volume ratio and showed a maximum value (38 mg l⁻¹ h⁻¹) at $\Psi_{\text{L/M}} = 0.05$.

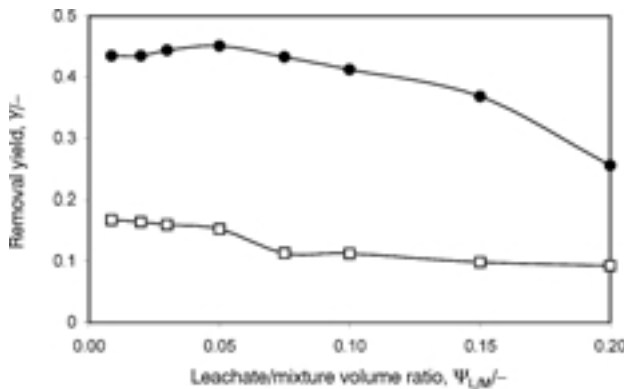


Fig. 1 – Preliminary batch tests with sludge from wastewater treatment plant. Average COD (●) and ammonia (□) removal yields obtained at different leachate/wastewater volume ratios

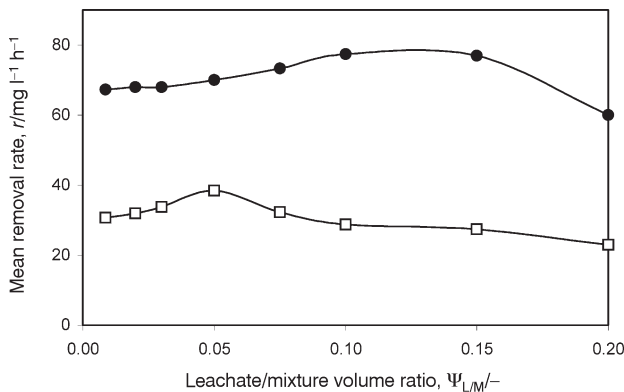


Fig. 2 – Preliminary batch tests with sludge from wastewater treatment plant. Average COD (●) and ammonia (□) removal rates obtained at different leachate/wastewater volume ratios

Continuous tests

To enhance the unsatisfactory COD and NH₄⁺ removal activities of the fresh sludge, leachate/wastewater mixtures were continuously treated in a single stage (without separation of COD removal, nitrification and denitrification) at variable hydraulic retention time (τ_{HRT}), namely 2, 4 and 8 h. To get a progressive adaptation of biomass to leachate as well as to minimize possible inhibition phenomena, the reactor was initially fed only with wastewater and then with mixtures containing leachate in proportion increasing up to 0.10.

As expected, COD removal increased with residence time (Fig. 3), whereas NH₄⁺ removal decreased (Fig. 4). The reason of such a behaviour should be searched, under the carbon source shortage conditions tested in this study, in the complex interplay between the heterotrophic metabolism, which is at the basis of organic pollutant degradation, and the autotrophic one, that depends on it and is responsible for nitrogen removal. Biomass con-

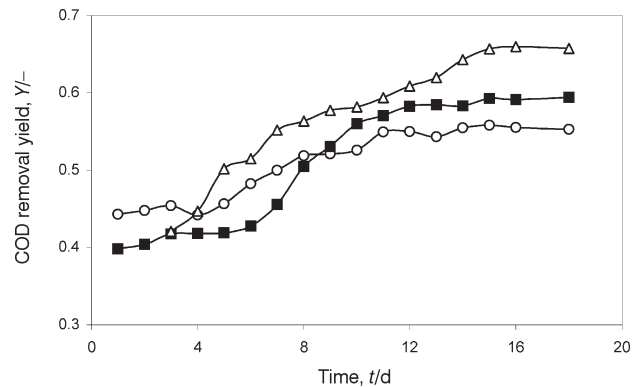


Fig. 3 – Behaviour of COD removal yield during continuous treatment of leachate/wastewater mixtures with volume ratio increasing up to 0.1. Hydraulic retention time (h): (○) 2; (■) 4; (◇) 8

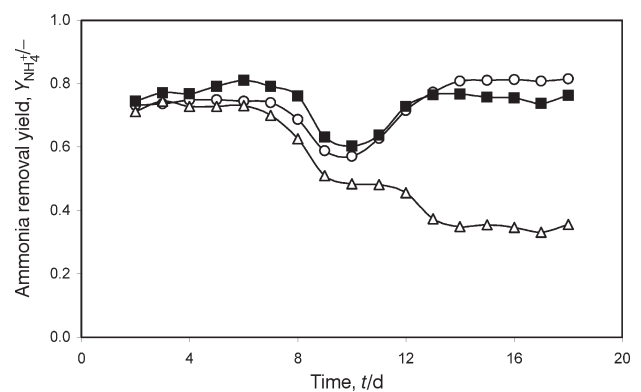


Fig. 4 – Behaviour of ammonia removal yield during continuous treatment of leachate/wastewater mixtures with volume ratio increasing up to 0.1. Hydraulic retention time (h): (○) 2; (■) 4; (◇) 8

centration, expressed as volatile suspended solids in the mixed liquor (MLVSS), reached a maximum threshold of about 4,500 mg l⁻¹ at $\tau_{\text{HRT}} = 4$ h, and decreased notably either under conditions of endogenous metabolism ($\tau_{\text{HRT}} = 8$ h; $\gamma_{\text{MLVSS}} = 1,000$ mg l⁻¹) or close to wash-out ($\tau_{\text{HRT}} = 2$ h; $\gamma_{\text{MLVSS}} = 3,300$ mg l⁻¹).

Sludge activity tests

Sludges from the above continuous tests were recovered by centrifugation and used in batch activity tests on leachate/wastewater mixtures at volume ratios varying from 0.05 to 0.20 (chemical composition in Table 3). COD and NH₄⁺ removals remained relatively constant for $\Psi_{\text{L/M}}$ up to 0.15, while they strongly decreased beyond this threshold (Table 4).

When biomass grown at $\tau_{\text{HRT}} = 8$ d and $\Psi_{\text{L/M}} \leq 0.15$ was used, the COD removal was unexpectedly low ($Y_{\text{COD}} = 0.45\text{--}0.48$), if compared with that obtained under continuous steady-state conditions ($Y_{\text{COD}} > 0.65$). The sludge produced at $\tau_{\text{HRT}} = 4$ h

Table 3 – Chemical composition of leachate/wastewater mixtures with different volume ratios ($\Psi_{L/M}$) used in batch activity tests

$\Psi_{L/M}$, l l ⁻¹	0.05	0.10	0.15	0.20
γ_{COD} , mg l ⁻¹	280	333	391	440
$\gamma_{\text{NH}_4^+}$, mg l ⁻¹	94	142	189	238
γ_{TKN} , mg l ⁻¹	154	236	314	398
$\zeta_{\text{TKN/COD}}$, mg mg ⁻¹	0.55	0.71	0.80	0.90

Table 4 – COD and ammonia removal yields obtained by batch activity tests at variable leachate volume ratio ($\Psi_{L/M}$) and using sludges acclimated at different residence times

$\Psi_{L/M}$, l l ⁻¹	0.05	0.10	0.15	0.20
$\tau_{\text{HRT}} = 2$ h				
Y_{COD} (–)	0.49	0.54	0.47	0.38
$Y_{\text{NH}_4^+}$ (–)	0.88	0.87	0.84	0.77
$\tau_{\text{HRT}} = 4$ h				
Y_{COD} (–)	0.74	0.76	0.67	0.42
$Y_{\text{NH}_4^+}$ (–)	0.76	0.71	0.67	0.57
$\tau_{\text{HRT}} = 8$ h				
Y_{COD} (–)	0.45	0.48	0.45	0.37
$Y_{\text{NH}_4^+}$ (–)	0.45	0.43	0.42	0.35

proved to be the most effective in COD degradation ($Y_{\text{COD}} = 0.67$ – 0.76), while showing an intermediate behaviour with respect to nitrogen removal ($Y_{\text{NH}_4^+} = 0.67$ – 0.76). The sludge produced at $\tau_{\text{HRT}} = 2$ h showed notable decrease in Y_{COD} (0.47 – 0.54), the highest $Y_{\text{NH}_4^+}$ value (0.84 – 0.88), and progressive disappearance of nitrite from the culture medium.

Likewise the COD removal yield, the COD removal rate increased using sludge continuously acclimated only under optimal conditions ($\tau_{\text{HRT}} = 4$ h), i.e. in the absence of any endogenous metabolism ($\tau_{\text{HRT}} = 8$ h) or inhibiting NH_4^+ levels ($\tau_{\text{HRT}} = 2$ h). But the most interesting result of sludge acclimation was the remarkable improvement of ammonia removal, already reported by *Diamadopoulos* et al.⁹ and *Kapetanios* et al.¹⁶ In fact, at $\tau_{\text{HRT}} = 4$ h, this yield increased from 0.10 – 0.17 (Fig. 1) to 0.67 – 0.76 (Table 4) and the rate from 30 – 39 mg l⁻¹ h⁻¹ (Fig. 2) to 35 – 63 mg l⁻¹ h⁻¹ (Table 5).

Discussion

The results of preliminary batch tests are in good agreement with those obtained by *Diamado-*

Table 5 – Average COD and ammonia removal rates obtained by batch activity tests at variable leachate volume ratio ($\Psi_{L/M}$) and using sludges acclimated at different residence times

$\Psi_{L/M}$, l l ⁻¹	0.05	0.10	0.15	0.20
$\tau = 2$ h				
r_{COD} , mg l ⁻¹ h ⁻¹	68.7	89.9	92.3	82.9
$r_{\text{NH}_4^+}$, mg l ⁻¹ h ⁻¹	41.3	62.1	79.4	91.4
$\tau = 4$ h				
r_{COD} , mg l ⁻¹ h ⁻¹	103.0	126.5	130.4	93.5
$r_{\text{NH}_4^+}$, mg l ⁻¹ h ⁻¹	35.5	50.7	62.9	67.5
$\tau = 8$ h				
r_{COD} , mg l ⁻¹ h ⁻¹	63.1	80.4	87.8	81.0
$r_{\text{NH}_4^+}$, mg l ⁻¹ h ⁻¹	21.1	30.8	40.2	44.0

poulos et al.,⁹ *Çeçen* and *Çakiroglu*,¹⁰ and *Chang* et al.,¹⁷ who suggested $\Psi_{L/M} = 0.10$ as the optimum condition for a combined aerobic treatment of these wastes.

The mean COD removal rate fell beyond $\Psi_{L/M} = 0.05$, likely due to some inhibitory effect exerted by substances contained in the leachate; on the other hand NH_4^+ removal rate proved to be scarcely influenced by the volume ratio. The observed higher rates of COD rather than NH_4^+ removals are consistent with the well-known slowness of lithoautotrophic metabolism, leading to NO_3^- formation, with respect to the heterotrophic one responsible for both COD and NO_3^- consumptions.^{18,19} So, the production of NO_3^- became the limiting factor of the whole nitrogen reduction, while most of the carbon was being consumed by the aerobic microflora.

Likewise preliminary batch tests, the results obtained using sludge preliminarily adapted to different hydraulic retention times demonstrate that the yields of, both, COD and NH_4^+ removal remained relatively constant for $\Psi_{L/M}$ up to 0.15, while decreasing beyond this threshold. Considering that leachate is characterized by higher NH_4^+/COD ratio (0.80) than wastewater (0.20), this effect was likely due to excess free ammonia²⁰ rather than to excess toxic carbon sources in the most concentrated mixture. For this reason, comparison will be made only among yield values referring to non-inhibitory conditions.

The results of Table 4 put in evidence a poor NH_4^+ removal, using sludge previously acclimated to $\tau_{\text{HRT}} = 8$ h. It is possible that a strong inhibition took place when this sludge, previously adapted to the endogenous metabolism, met excess carbon source conditions during batch tests. The reduced

carbon dioxide development, due to aerobic metabolism, could have caused deceleration of the autotrophic metabolism responsible for nitrification/denitrification. Since the metabolism of nitrating microflora (*Nitrobacter* sp.) depends on the nitriting one (*Nitrosomonas* sp.), such a carbon source shortage could have favoured the development of the former metabolic type with respect to the latter, as suggested by nitrite accumulation above inhibiting levels ($\gamma = 0.3 \text{ mg l}^{-1}$) (results not shown). Besides, the generalized absence of nitrate in the medium under every conditions tested would be consistent with the well-known quicker heterotrophic metabolism of denitrifying bacteria (*Pseudomonas* sp., *Thiobacillus* sp., etc.) with respect to the autotrophic ones.^{18,19} These effects on the whole could have been responsible for the unsatisfactory ammonia removal ($Y_{\text{NH}_4^+} = 0.43\text{--}0.45$), that appeared to be limited by the carbon dioxide formation rate, i.e. by the aerobic metabolism.

The particularly low COD removals should be ascribed, on the other hand, to the peculiar composition of this sludge. Its microscopic examination did in fact reveal it was particularly rich in *Nitrosomonas* sp. and poor in *Nitrobacter* sp., if compared with sludge obtained at shorter residence times.

Such inhibiting effects only apparently disagree with the results obtained under continuous working conditions (Fig. 3 and 4), which are often preferred just to minimize excess substrate inhibitions. In other words, the best yields were achieved for COD continuous removal at $\tau_{\text{HRT}} = 8 \text{ h}$, because the mass concentration of organic substances within the reactor was well below the inhibiting levels experienced by sludge during batch activity tests.

The low Y_{COD} and high $Y_{\text{NH}_4^+}$ values obtained with sludge acclimated to $\tau_{\text{HRT}} = 2 \text{ h}$ (Table 4) were likely due to continuous acclimation to excess organic carbon conditions and predominance of species belonging to *Nitrobacter* rather than *Nitrosomonas* genus. The large availability of carbon dioxide from aerobic respiration could have stimulated the nitrating activity to such an extent, that the nitriting stage became the factor controlling NH_4^+ removal. On the other hand, the sludge produced at $\tau_{\text{HRT}} = 4 \text{ h}$ proved to be the most effective in COD degradation, while showing an intermediate behaviour with respect to nitrogen removal. The higher availability of organic pollutants under these continuous working conditions could have stimulated the aerobic respiration so that the heterotrophic microflora shifted from a situation of endogenous metabolism to conditions of growth limitation by the retention time. The consequent increase in carbon dioxide formation rate could have accelerated the growth of autotrophic microflora, with particu-

lar concern to the nitrating one. Microscopic examination of the composition of this sludge demonstrated comparable nitriting and nitrating populations, which would justify its higher capacity of NH_4^+ removal in batch activity tests. This hypothesis would also be consistent with the relatively constant concentration of NO_2^- detected in the medium at non-inhibitory level (0.1 mg l^{-1}).

In conclusion, the results of this work demonstrate that old leachate can be effectively treated in a single stage, provided that it is preliminarily diluted with young leachate. The best proportion for leachate removal along with wastewater was $\Psi_{\text{L/M}} = 0.10$. This optimum value, which confirms the observations of *Diamadopoulos et al.*,⁹ *Çeçen and Çakiroglu*,¹⁰ and *Chang et al.*,¹⁷ could be due to a stimulating effect of leachate components at low ratios as well as to the occurrence of inhibiting phenomena due to free ammonia at high leachate levels.^{20,21} Comparison among literature data is difficult because of the wide variability in leachate composition and age, reactor configuration, operating conditions, and so on. Nevertheless, the yields obtained in this work for continuous COD and ammonia removals compare with those obtained for raw and old landfill leachates with aerated ponds, activated sludges or combined chemical/biological techniques,^{3,7,13,22} but are lower than those reported for anaerobic^{23–26} or combined anaerobic/anoxic/aerobic treatments²⁷ or using selected autotrophic denitrifying biomass.²⁸ Finally the acclimation of sludge to $\tau_{\text{HRT}} = 4 \text{ h}$ seemed to be the optimum compromise to maximize COD and NH_4^+ removal yields and rates.

Nomenclature

- γ_{BOD} – biological oxygen demand, mg l^{-1}
- γ_{COD} – chemical oxygen demand, mg l^{-1}
- $\gamma_{\text{NH}_4^+}$ – ammonia concentration, mg l^{-1}
- τ_{HRT} – hydraulic retention time, h
- $\Psi_{\text{L/M}}$ – leachate/mixture volume ratio, dimensionless
- γ_{MLVSS} – volatile suspended solids in the mixed liquor, mg l^{-1}
- MSW – municipal solid waste
- r_{COD} – COD removal rate, $\text{mg l}^{-1} \text{ h}^{-1}$
- $r_{\text{NH}_4^+}$ – ammonia removal rate, $\text{mg l}^{-1} \text{ h}^{-1}$
- γ_{TKN} – total nitrogen (Kjeldahl), mg l^{-1}
- γ_{VSS} – volatile suspended solids, mg l^{-1}
- Y_{COD} – yield of COD removal, dimensionless
- $Y_{\text{NH}_4^+}$ – yield of ammonia removal, dimensionless
- ζ – mass ratio
- Q – volume flow rate, l h^{-1}

References

1. Borzacconi, L., López, I., Anido, C., *Wat. Sci. Technol.* **36(6/7)** (1997) 479.
2. Rees, J. F., *J. Chem. Technol. Biotechnol.* **30** (1980) 458.
3. Pohland, F. G., Harper, S. R., Critical review and summary of leachate and gas production from landfill, EPA 600-2-86-073. Cincinnati, Ohio: US EPA Hazardous Waste Engineering Research Laboratory (1985).
4. Farquhar, G. J., *Can. J. Civ. Eng.* **16** (1989) 317.
5. El-Fadel, M., Bou-Zeid, E., Chahine, W., Alayli B., *Waste Manag.* **22** (2002) 269.
6. Baumgarten, G., Seyfried, C. F., *Wat. Sci. Technol.* **34(7/8)** (1996) 445.
7. Robinson, H. D., Maris, P. J., *Wat. Res.* **17** (1983) 1537.
8. Keenan, J. D., Steiner, R. L., Fungaroli, A. A., *J. Wat. Pollut. Control Fed.* **56** (1984) 27.
9. Diamadopoulos, E., Samaras, P., Dabou, X., Sakellariopoulos, G. P., *Wat. Sci. Technol.* **36(2/3)** (1997) 61.
10. Çeçen, F., Çakiroglu, D., *Biotechnol. Lett.* **23** (2001) 821.
11. Chen, P. H., *Environ. Int.* **22** (1996) 225.
12. Hjelmar, O., Johannessen, L. M., Knox, K., Ehrig, H. J., Flyvbjerg, J., Winther, P., Christensen T. H., In T. H. Christensen, R. Cossu, R. Stegmann (ed.), Proceedings Sardinia 95, 5th International Landfill Symposium, Vol. 1, Cagliari, Italy: Environmental Sanitary Engineering Centre (1995) pp. 243–262.
13. Papadopoulos, A., Fatta, D., Loizidou, M., *J. Environ. Sci. Health* **33** (1998) A651.
14. Blengino, A. M., Manassero, M., Rossello, A., Spanna, C., In M. Kamon (ed.) Proceedings of the 2nd International Congress on Environmental Geotechnics, Rotterdam, The Netherlands: Balkem (1996) pp. 629–634.
15. APHA, In L. S. Clesceri, S. E. Greenberg, R. R. Trussel (ed.) Standard Methods for the Examination of Water and Wastewater, 17th edition. Washington, DC: AWWA, WPCF, APHA (1990).
16. Kapetanios, E., Papadopoulos, A., Loizidou, M., Haralambous, K. J., In T. H. Christensen, R. Cossu, R. Stegmann (ed.) Proceedings Sardinia 95, 5th International Landfill Symposium, Vol. 1, Cagliari, Italy: Environmental Sanitary Engineering Centre (1995) pp. 551–558.
17. Chang, D., Kim, S. Y., Hur, J. M., Chung, T. H., Choi, J. G., In T. H. Christensen, R. Cossu, R. Stegmann (ed.), Proceedings Sardinia 95, 5th International Landfill Symposium, Vol. 1, Cagliari, Italy: Environmental Sanitary Engineering Centre (1995) pp. 499–510.
18. Brock, T. D., Madigan, M. D., Martinko, J. M., Parker, J., *Microbiologia*, Milan, Italy: Citta Studi (1995).
19. Schlegel, H. G., *Microbiologia*, 1st edition, Bologna, Italy: Zanichelli (1996).
20. Aktas, O., Çeçen, F., *Biotechnol. Lett.* **23** (2001) 1607.
21. Hwang, B-H., Hwang, K-Y., Choi, E-S., Choi, D-K., Jung, J-Y., *Biotechnol. Lett.* **22** (2000) 1287.
22. Robinson, H. D., Grantham, G., *Wat. Res.* **22** (1986) 733.
23. Henry, J. G., Prasad, D., Young, H., *Wat. Res.* **21** (1987) 1395.
24. Kennedy, K. J., Hamoda, M. F., Guiot, S. G., *J. Wat. Pollut. Control Fed.* **60** (1988) 1675.
25. Chang, J. E., *Wat. Sci. Technol.* **21(4/5)** (1989) 133.
26. Keenan, P. J., Iza, J., Switzenbaum, M. S., *Wat. Environ. Res.* **65** (1993) 181.
27. Borzacconi, L., Ottonello, G., Castelló, E., Palaez, H., Gazzola, A., Vinas, M., *Wat. Sci. Technol.* **40(8)** (1999) 145.
28. Koenig A., Liu, L. H., *Wat. Sci. Technol.* **34(5/6)** (1996) 469.

