LANDFILL LEACHATE TREATMENT: CASE STUDIES

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ABSTRACT

Three case studies of domestic landfill leachates treatment are reported in order to connect biological and physico-chemical processes effectiveness to leachate characterisation and key contaminants treatability.

KEY WORDS: Landfill leachate, characterisation, biological treatment, physico-chemical treatment

INTRODUCTION

Today, the European Union generates 2,140 million tons of waste per year. France's share in that total amounts to 580 million tons per year, consisting of the following broad categories: agricultural wastes 400 Mt/yr, industrial wastes (inert, non-toxic, toxic) 150 Mt/yr, municipal wastes 30 Mt/yr.

An official study of the destination of collected wastes shows that landfilling is the option chosen for 62% of municipal wastes and 57% of industrial wastes, in contrast to reuse by recycling, incineration or composting, which applies to a mere 35% of municipal wastes and 36% of industrial wastes. Keeping pace with both regulatory and technical trends, landfill centres and the more recent storage sites, must allow wastes to be managed rationally in terms of environmental impact and operating costs. In France, landfill sites are classified as Class I for ultimate and stabilised toxic industrial wastes or Class II for domestic and non-toxic industrial wastes.

Wastes cause two types of pollution, which correspond to the migration into the natural environment of:

- leachates, defined as water that has percolated through the wastes (rainwater or groundwater seepage), a source of soil and groundwater contamination,
- biogas produced by the fermentation of organic matter, a source of air pollution.

With regard to leachates, controlling the pollutant loading means reducing its quantity by containing or treating the waste to comply with certain discharge characteristics which are compatible with the receptor medium (river, sea, municipal treatment plant).

STATE-OF-THE-ART: LEACHATES CHARACTERISATION AND TREATMENT

In addition to the type of waste involved (industrial, municipal), leachate characteristics are determined by the siting, the design and the mode of operation of the landfill, and also depend on its evolution through time.

Organic and inorganic contaminants of landfill leachate are released from the waste due to successive biological, chemical and physical processes. Several studies deal with the biological decomposition of solid waste Classing the landfill to a bioreactor makes it possible to relate the leachate composition to the activity of micro-organisms. Basically, three phases of decomposition are distinguished for domestic landfills occurring within twenty years (2-4). In the first stage following waste deposit, initial aerobic phase rapidly consumes the confined oxygen and water infiltration enhances acetogenic fermentation producing leachate characterised by high BOD, COD and ammoniacal nitrogen contents. Volatile fatty acids (VFA) are the main components of the organic matter released, besides the lower pH solubilises metals. Gradually, the methanogenic phase of decomposition starts and consumes the simple organic compounds resulting from acetogenic process to produce biogas. In that stage, the leachate composition represents the dynamic equilibrium between the two microbiological mechanisms with lower BOD and COD values while the ammonia concentration remains high. Dissolved inorganic materials are continuously released. With landfill ageing, waste stabilisation takes place. As the volatile fatty acids leachate content decreases parallel to the BOD/COD ratio, the leachate organic matter is made up of high molecular weight humic and fulvic-like material (HMW).

Though leachate Composition may vary widely within these stages three types of leachates can be defined according to landfill age (5) (Table 1).

Table 1: Leachates classification (5)

leachate type	Young	Intermediate	Stabilised
Landfill age yr	<5	5-10	>10
pH	<6.5	7	>7.5
COD g/l	>20	3-15	<2
BOD/COD	>0.3	0.1 - 0.3	<0.1
TOC/COD	0.3	-	0.4
Organic matter	70 - 90 % VFA	20 - 30 % VFA	HMW
Nitrogen	100 - 2000 mg/l TKN		
Metals g/l	2	<2	<2

Moreover, extensive studies have investigated the characterisation and stability of organic matter in landfill leachates (6-10). From these data, the knowledge of the organic matter composition leads the treatment choice towards biological processes for young landfill leachates and towards physicochemical processes for aged ones.

Many investigations have been carried out to prove the biological treatment suitability for young landfill leachates. Because of operational facilities on site, the aerated lagooning technique was first selected and shown very effective for leachates with high ratio BOD/COD in order to reach 90% COD and 95% BOD_5 removals (1113). Beyond standard aerobic treatments which benefit lies in residence time reduction compared to lagooning, recent studies imply anaerobic technology to achieve higher reaction rates (14-16). Except for volatile fatty acids degradation, both aerobic and anaerobic treatments efficiencies are limited because of the same recalcitrant molecules in part identified as simple organics and natural macromolecules like lignins, tannins, humic materials, carbohydrates (17, 18). Consequently, as the volatile fatty acids fraction decreases with landfill age, the biological treatments become ineffective in organic matter disposal from stabilised landfill leachates. Interestingly, although ammonia nitrogen conversion to nitrate is somewhat more difficult, nitrification may occur successfully during aerobic biological removal of organic material from young or aged landfill leachates (19, 20). This phenomenon is advantageously turned to account in nitrification-denitrification sequence for extensive nitrogen removal (21).

Since the first studies about leachate characterisation, attempts have been made to connect the organic matter composition to biological and physico-chemical processes efficiency (7). Conventional physico-chemical treatment technology includes chemical precipitation, coagulation-flocculation activated carbon adsorption, chemical oxidation and separation by membranes.

Chemical precipitation for leachate treatment mainly makes use of lime because of its low cost and availability with regard to the high dose required. From the mentioned results, raw leachates from medium aged fills or pretreated biological leachates with BOD/COD ratio of 0.1-0.5 provide the best effectiveness of lime precipitation not only for colour and metals removals but also for COD reduction due to a predominant typical molecular weight fraction (7, 22-24). Provided that lime dose (1.4-6.4 g/l) is sufficient to raise the pH value above 10, precipitation combined with aerobic process is able to lower COD value by 30 to 70% (22, 23).

In the same way, coagulation-flocculation process with ferric chloride or alum is better applied to remove high molecular weight compounds that represent a large part of the organic matter in raw stabilised leachates or biologically pretreated ones (BOD/COD<0.1)(20, 22, 23, 25). From literature results, the optimum coagulation-flocculation is obtained with ferric chloride (1-4.8 g/l) under controlled acid pH (4.3-4.9); in these conditions, the treatment ensures considerable reduction of suspended solids content and gives about 60% COD removal (20, 23, 25).

Activated carbon adsorption was investigated on the one hand, as an alternative treatment method to chemical precipitation for raw or biologically pretreated young landfill leachates, on the other hand as polishing treatment combined with coagulation-flocculation for stabilised leachates (7, 20, 25). Because adsorptive capacities increase while the volatile fatty acid fraction decreases, activated carbon adsorption is shown more effective in treating stabilised or biological pretreated young leachate (7). However, even if substantial reductions of COD are observed for raw or chloride ferric pretreated leachate from old landfill, high carbon loadings (respectively 6 and 1.5 g/l) required make this process implementation prohibitive (25).

Last years, concerning chemical oxidation, numerous studies have explored the ozonation suitability for, leachate treatment. Direct ozonation (0.4-1.3 g O_3/g COD) is shown able to provide excellent colour removal with limited COD reduction (25-44%)

when applied both to young and aged leachates (22, 26, 27). Recent developments and applications involve advanced oxidation processes (AOP) based on ozone activation by hydrogen peroxide, UV-light, heterogeneous catalyst to produce HO° radicals known as more efficient oxidant, with the aim to lower the ozone dose and to enhance organic matter degradation (28-34) (Table 2). Despite operating factors variations in matter of mass transfer retention time, pH as well as lack of initial leachate characterisation the below-mentioned results among others prove the advanced ozonation effectiveness for COD removal from leachates whose organic material is mainly non-biodegradable. Ozonation is not so appropriate for young landfill leachate because major part of the COD is made up of the fatty acids refractory to ozone attack (7).

Table 2: Ozonation results

Ref.	Leachate type	Initial COD mg/l	AOP dosage g/g	Ozone dosage g/g COD	COD removal %
28	young	881	O_3/H_2O_2 0.7 O_3	1.4 1.4	40 43
29	young MBR	2500	O_3/H_2O_2 0.4 O_3	2.8 4.3	94 70
30	biological	1250	O_3/UV O_3	3 3	90 75
31	-	1700	O_3/UV O_3	1.5 1.5	94 36
32	biological	750	O_3/UV O_3	0.7	70
33	stabilised	2300	O_3/UV or O_3 O_3/H_2O_2 0.3 O_3	1.5 1.5 1.5	60 92 89
34	stabilised biological	1400	O_3/catalyst O_3	1.5 1.5	89 63

Besides ozonation, emerging technologies for leachate treatment involve membrane separation process subsequent to biological pretreatment in order to avoid severe membrane fouling. For stabilised domestic waste landfills, biological treatment combined with reverse osmosis produces a permeate conform to regulations together with a residual concentrate representing about 20% of the raw leachate (7, 35). The coupling of microfiltration membrane bioreactor with reverse osmosis further allows to overcome process limitations due to fouling by residual organic compounds or suspended solids. This combination achieves full purification of aged landfill leachates (36).

The case studies of Class II landfills hereafter reported show that the treatment line is defined for each particular case to reflect the specific characteristics of the leachate in accordance with regulatory values for final discharge.

LEACHATES CHARACTERISATION, TREATMENT OBJECTIVES

This study concerns three different landfills located in France at Lapeyrouse-Fossat (LF), Saint-Etienne (SE) and Bagnols-en-Forêt (BEF) and whose typical average characteristics and regulatory requirements are respectively given in tables 3 to 5.

Landfill leachates are collected in a lagoon. Besides,' chemical coagulation with ferric chloride at pH 5.5 to 6 combined with flocculation with anionic polymer and subsequent flotation is applied to SE leachate. This pretreatment ensures substantial removals of COD (48%), BOD_5 (58%) and TKN (10%) while the SS content decreases to 35 mg/l, sole value unaffected by high raw leachate quality variations (37).

Table 3: Lapeyrouse-Fossat leachate 30 m^3/d

LF leachate	Raw	Standards
pH	8.2	5.5-8.5
BOD_5 mg/l	135	30
COD mg/l	880	100
BOD_5/COD	0.15	
TKN mg/l	310	30
$N-NH_4^+$ mg/l	300	
Suspended solids mg/l	160	100

Table 4: Saint-Etienne leachate 20 m^3/h

SE leachate	Raw	Physico-chemical	Standards
pH	8.3	6.4	5.5 - 8.5
BOD_5 mg/l	170	70 (max. 100)	30
COD mg/l	1400	730 (max. 1050)	150
BOD_5/COD	0.12	0608	
TKN mg/l	710	640 (max. 780)	20
$N-NH_4^+$ mg/l	690	610 (max. 760)	-
$N-NO_3^-$ mg/l	1.7		50
Suspended solids mg/l	270	35	30

Table 5: Bagnols-en-Forêt leachate 100 m^3/d

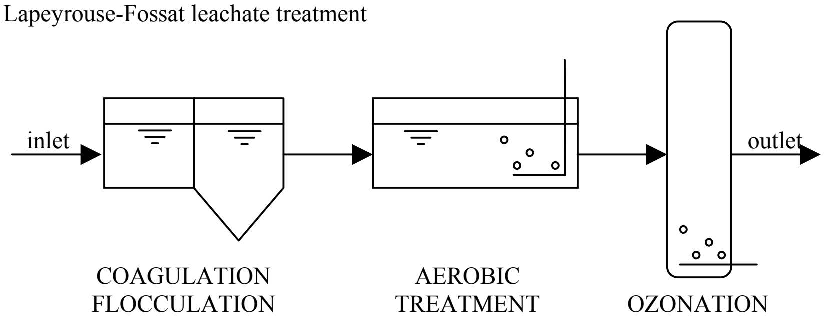
BEF leachate	Raw	Standards
pH	8	5.5-8.5
BOD_5 mg/l	500	30
COD mg/l	2000	90
BOD_5/COD	0.25	
TKN mg/l	400	10
$N\text{-}NH_4^+$ mg/l		300
Suspended solids mg/l	200	30
Metals (Al^{3+}) mg/l	8.5 (3.25)	15 (0.1)

MATERIALS AND METHODS

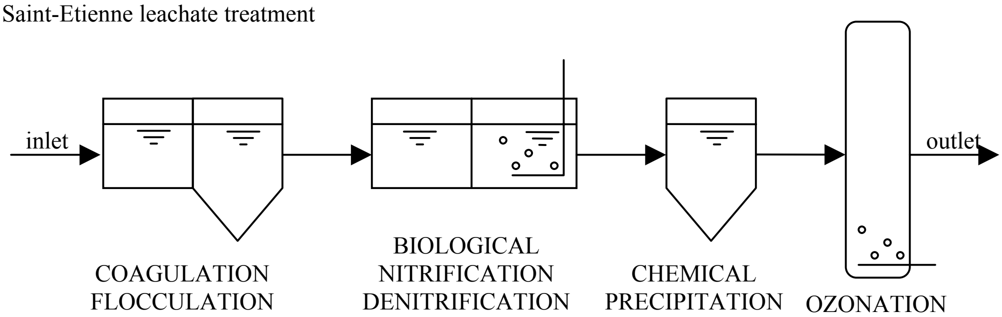
In order to meet the standards established for each leachate, different treatment lines which start-up results are presented, have been designed according to laboratory tests.

- Treatment lines:

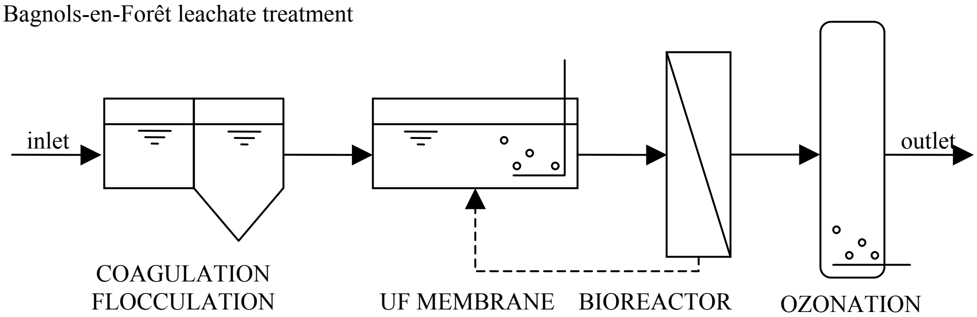
Lapeyrouse-Fossat leachate treatment



Saint-Etienne leachate treatment



Bagnols-en-Forêt leachate treatment



- Analytical methods:

Leachate samples were analysed according to AFNOR standardised methods.

RESULTS AND DISCUSSION

According to literature, conventional parameters classify the BEF leachate as intermediate and the LF and SE ones rather as stabilised. The higher BOD_5/COD (0.25) and lower TKN/COD (0.2) ratios observed for the BEF leachate are consistent with a significant volatile fatty acids content. For the LF and SE leachates respectively, the BOD_5/COD value decrease (from 0.15 to 0.12), parallel to the TKN/COD ratio increase (from 0.35 to 0.50), indicates higher and higher non-biodegradable organics content. In every case, main contribution to TKN nitrogen concentration is ammonia: 75% for the BEF leachate, 97% for LF and SE ones. For easier comparison, figures 1 to 3 depict the gradual relative variations of the parameters bound by regulatory requirements during the different treatment lines.

- Coagulation-flocculation

Coagulation-flocculation process with ferric chloride is applied at first to every raw leachate with similar operating conditions. The results clearly show that, over the expected consequent suspended solids content reduction (60-80%), the organic matter removal extent in matter of COD highly depends on the BOD_5/COD value. The best COD removal and inferred BOD_5 one are obtained from the stabilised leachates probably due to coprecipitation of coagulant flocs with high molecular weight compounds.

- Biological treatment

Although biological treatment technologies used are quite- different, crude efficiencies comparison highlights the processes suitability for ammonia nitrogen removal. In any case, nitrification occurs successfully to reach high TKN conversion varying from 85 to 98%. Simultaneously, while the BOD_5/COD ratio becomes lower from 0.25 for the BEF leachate to 0.13 and 0.10 respectively for the aged LF and SE leachates, residual COD rapidly increases from 240 (BEF leachate) to 520 mg/l (SE leachate) because of resulting accumulation of high molecular weight organics. For SE leachate, denitrification stage complements the nitrogen removal over 95%. Compared to the two others more conventional technologies used, the ultrafiltration membrane bioreactor provides complete suspended solids retention.

- Chemical precipitation

To take advantage of the substantial magnesium concentration (300 mg/l), lime precipitation is applied to biological SE leachate. The magnesium oxide coagulation effect removes the whole of the inorganic carbon together with 27% of the residual COD. This organic matter fraction, assumed to be made up of specific molecular weight range compounds, is responsible for a high part of the leachate colour.

- Ozonation

The treatment lines end with ozonation in order to meet the standards. Considering the three cases, results show that the relation between ozone consumption and COD removal is linear. Over full discoloration, final ozonation process is able to reach at least the COD reduction required, i.e. 89% for the SE and LF leachates and 96% for the BEF one with transferred ozone dose of 2.5g/g COD. Moreover, kinetic studies point out that COD and TOC removals are closely connected. Resulting final TOC values such as 50 mg/l prove that organic matter oxidation occurs up to strong mineralisation.

Figure 1: Results for BEF leachate treatment

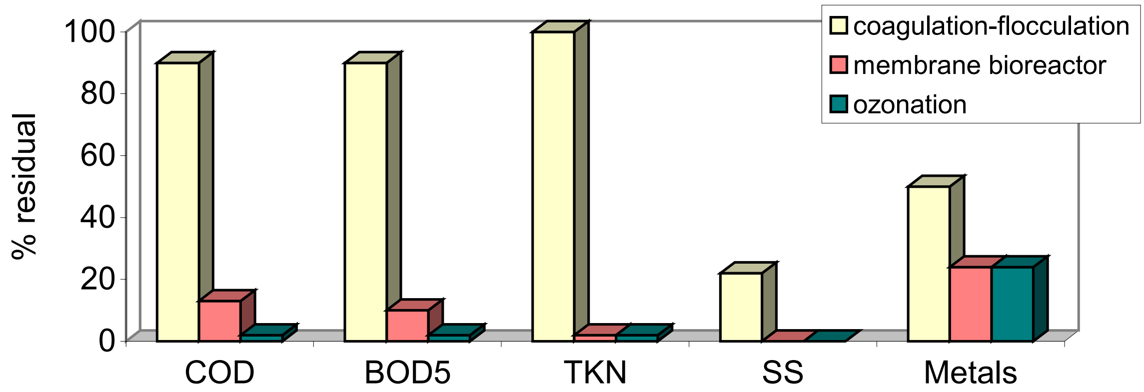


Figure 2: Results for LF leachate treatment

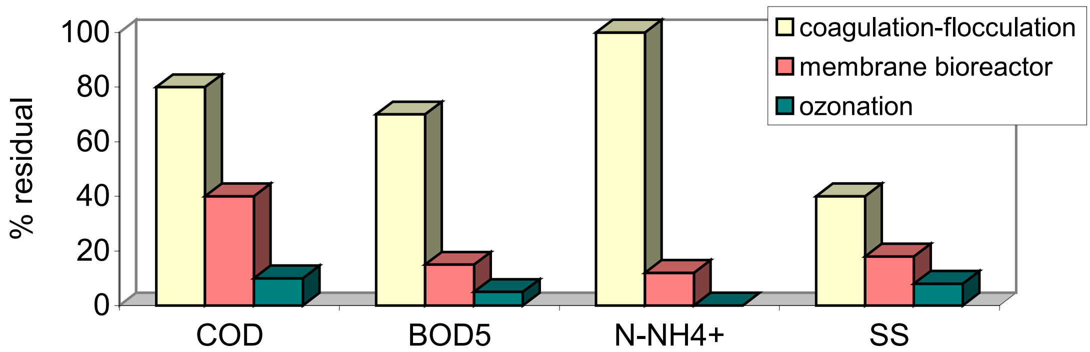
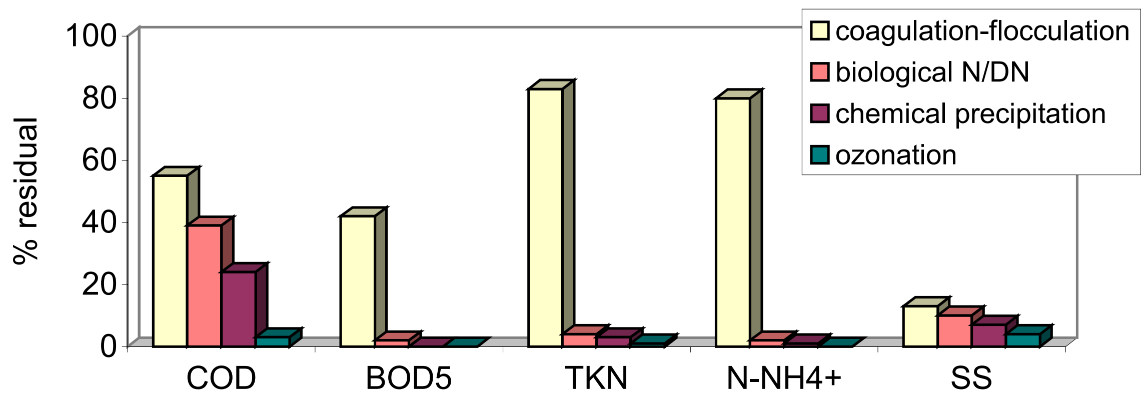


Figure 3: Results for SE leachate treatment



CONCLUSION

This study shows that broad knowledge of leachate organic matter composition and stability related to available treatment methods effectiveness determines appropriate treatment lines in order to meet regulatory requirements for discharge. The case studies here reported display the suitability of biological process rightly combined with physico-chemical ones for intermediate to aged landfill leachate treatment. With regard to conventional parameters (COD, BOD_5, TKN, SS*)* behaviour, relative efficiencies of coagulation - flocculation, biological treatment and chemical precipitation are connected to the specific nature of affected contaminants. Subsequent to this common pretreatments line, final ozonation once achieves substantial mineralisation of residual organic matter without apparent reactivity limit.

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