1 TREATMENT OF LEACHATES IN HIGH-TECH SYSTEMS

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INTRODUCTION

Leachates are complex wastewaters, which often contain high concentrations of ammonium ions, refractory organic compounds as well as heavy metals. The composition of the leachate is a function of the kind of waste that has been deposited at the landfill. Furthermore, substances created during microbial and abiotic degradation processes of the waste will be eluted from the landfill. It is therefore difficult to determine the chemical composition of a leachate especially in regard to organic substances. Leachates from different landfills differ in composition. No single treatment process is therefore suitable for all leachates. The process has to be optimised for the specific leachate, which will be treated.

KEYWORDS

Leachate treatment, microbial processes, physical/chemical processes.

COMBINED TREATMENT OF LEACHATE AND MUNICIPAL WASTEWATER

Leachates from many landfills are being treated together with municipal wastewater at the sewage treatment plant (Linde 1995, Viviani and Torregrossa 1997). This solution is however being questioned due to e.g. the high ammonium ion content of many leachates. Many sewage treatment plants have during recent years got stricter limits concerning treatment in regard to nitrogen. Leachates and municipal wastewater do furthermore differ in regard to content of organic substances. There might be a risk for that some compounds in the leachate can disturb the processes in the municipal wastewater treatment plant or that some compounds will pass the treatment process unaffected. There is also a discussion concerning the risk for heavy metals from leachates accumulating in the sewage sludge. It is important that the loads in regard to both biochemical oxygen demand (BOD) and nitrogen are reasonable in comparison to the capacity of the treatment plant and that enough nutrients are added, especially phosphorous, since nitrogen in most cases is available in access amounts. Many of the problems reported with co-treatment of leachates and municipal wastewaters might depend on overloading of the sewage treatment plant (Ahnert and Ehrig 1992).

MICROBIAL PROCESSES

The microbial methods can be divided into aerobic, anoxic (nitrate is used as electron acceptor) and anaerobic processes. A combination of these processes is in most cases needed for an efficient treatment of leachates.

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Aerobic processes

Nitrification is an efficient way to reduce the ammonium ion concentration in many leachates since they often contain relatively low concentrations of BOD. A high concentration of BOD involves a risk for nitrifying bacteria being out competed. The reduction of the ammonium concentration is important since ammonia is a toxic compound and ammonium is a fertilizer. A number of studies have been performed on nitrification of leachates (Knox 1987, Morling and Johansson 1989, Spengel and Dzombak 1991, Welander et al. 1997, 1998). Aerobic processes can furthermore be efficient for removal of many organic compounds. Aerobic processes can be built on many different designs e.g. suspended growth processes or the biofilm concept. Examples of suspended growth systems are the activated sludge process or aerated lagoons (Knox 1985, Robinson 1992). Processes built on the biofilm concept include e.g. trickling filters and the suspended carrier process (Knox 1985, Welander and Henrysson 1998). The carriers are kept in suspension through aeration in the latter process.

Suspended growth processes

Most studies on the activated sludge process have been performed at room temperature (Swedish EPA 1992). Furthermore, they have often been performed on leachates coming from landfills, which were in the acidogenic phase. These leachates contain high concentrations of easily degradable organic compounds such as fatty acids (Millot et al. 1987). Activated sludge processes are not so well evaluated in regard to treatment of leachates coming from more stabilized landfills containing low concentrations of BOD. Aerated lagoons have been studied in full-scale for instance in Great –Britain (Robinson and Grantham 1988, Robinson 1992). Chemical oxygen demand (COD) removals of between 83 and 97% or more have been obtained. The leachates used in most of these studies came from landfills in the acidogenic phase.

Attached growth processes

The microorganisms are growing on some carrier material in the biofilm processes. An advantage with these kinds of processes is that they are less sensitive to toxicity than suspended growth processes. Furthermore, the risk for loss of biomass due to poor separability is decreased in biofilm processes since the biomass is growing on a material. Disadvantages with the biofilm processes include the higher oxygen concentration needed in comparison to the suspended growth systems. Furthermore, the cost of the carrier material is of course a disadvantage. Results have shown nitrification of leachates in a trickling filter to be less temperature dependent than nitrification in an activated sludge processes (Knox 1985). Nitrification of leachate in a suspended carrier process at low temperatures has also been shown to be a competitive process (Welander et al. 1997,1998). The nitrification rate at $5^{\circ}C$ was approximately 77% of the rate at $20^{\circ}C$. These results can be explained by the fact that the solubility of oxygen in water increases as temperature decreases (Okey and Albertson 1989), which is important since the activity of the microorganisms in many biofilms is limited by the oxygen concentration available. An increase in the oxygen concentration and a lower specific activity of the microorganisms allow oxygen to penetrate more deeply into the biofilm, activating a greater number of microorganisms. An advantage with the suspended carrier process in comparison to other biofilm processes is the good mixing of the reactor content, which is achieved, minimizing the risk for liquid's short-circuiting and clogging of the media with biomass or other solids. A good mixing also ensures an efficient mass transfer, which is very important. The main disadvantage with the process is its strong oxygen dependence which has been shown to be approximately first order and not half-order as for other biofilm processes (Harrëmoes 1982, Hem et al. 1994).

Anoxic processes

An anoxic process (nitrate is used as electron acceptor) is needed if the nitrate ion concentration is to be reduced. A carbon source and a phosphorous source has to be added in most cases due to the fact that the leachates often contain low concentrations of phosphorous and BOD.

Anaerobic processes

Sulphate reducing bacteria can be used to reduce the concentrations of many heavy metals in different wastewaters. Sulphate is reduced to sulphide and the metals precipitates as metal sulphides. The precipitate can then be separated from the water phase. Anaerobic processes are also suitable for removal of some organic compounds. There is however in many cases a need for further treatment before the leachate can be released into a recipient. Anaerobic process design is built on the same concepts (suspended growth system or attached growth system) as the aerobic processes.

Suspended growth processes

Studies of leachate treatment in different anaerobic suspended growth systems such as lagoons, digesters and upflow anaerobic sludge blanket (UASB) reactors have shown the removal of organic compounds from acidogenic leachates to be relatively efficient (Boyle and Ham 1974, Mennerich 1987, Cameron and Koch 1989, Rumpf and Ferguson 1990, Blakey et al. 1992, Jans et al. 1992, Garcia et al. 1996). The study of Blakey et al. for example, concerning the treatment of acidogenic leachate in batch lagoons, showed there to be a COD removal of more than 80% at a temperature of 258C or 108C. The hydraulic retention time (HRT) needed was longer at 108C than at 258C however.

Treatment of leachates in digesters has yielded highly varying results. A COD removal of 22-96% has been reported in three studies (Bayle and Ham 1974, Cameron and Koch 1980, Méndez et al. 1989). The lowest removals were usually obtained when leachates from old landfills were treated or, alternatively, when treatment was performed at a low temperature (108C).

Treatment of landfill leachate in continuous or batch upflow anaerobic sludge reactors decreased the microtox® toxicity. The treated leachate did however not reach a quality suitable for direct disposal into the sewage treatment plant (Kennedy and Lentz 2000). A further treatment step such as e.g. membrane filtration would be required (Kennedy and Lentz 2000).

Attached growth processes

The COD removal during the treatment of leachates in upflow anaerobic filters has been reported to vary from 56-95% (Henry et al. 1987, Wu et al.1988, Méndez et al. 1989). Both methanogenic and acidogenic leachates were studied. The advantages of biofilm processes in comparison to suspended growth processes mentioned in connection with aerobic treatment also apply to anaerobic treatment. The weaker temperature dependence could be an exception. The potential for better retention of microorganisms in biofilm processes than in suspended growth systems could be of special importance in anaerobic processes, since anaerobic microorganisms grow so slowly.

A combination of aerobic and anoxic or anaerobic processes

Different process designs have been used for the combined aerobic and anoxic treatment of leachates including sequence batch reactors (SBR) and the suspended carrier process

(Hoshomi et al. 1989, Welander et al. 1998). The combination is needed in many cases in order to reduce the nitrogen content of the leachate. Ammonium is oxidized to nitrate in the aerobic stage and nitrate is reduced to nitrogen gas in the anoxic stage.

The sequential anaerobic-aerobic treatment of a medium-strength municipal landfill leachate by Kettunen et al. 1997 resulted in a COD removal of 80-90% and a nitrification of 65->99%. The anaerobic treatment was performed in a pilot scale UASB reactor, whereas the aerobic treatment was performed in a laboratory-scale activated-sludge reactor. The temperature was 13-28ßC in the anaerobic stage and 5-11°C in the aerobic stage. A comparison of sequential anaerobic-aerobic treatment with aerobic treatment alone showed removal of nitrogen in the aerobic treatment alone to mainly be due to assimilation. This poses a risk since it means that the ammonium may be released again when the sludge is used as landfilling.

PHYSICAL PROCESSES

Reverse osmosis

Reverse osmosis is a process, which has been studied in full-scale in Germany, the Netherlands and Switzerland (Woelders 1994, Linde 1995). This process has in many cases been shown to be efficient in regard to removal of organic compounds from leachates. The COD concentration has in some cases been reduced with over 99% (Woelders 1994, Linde 1995, Peters 1997). Reverse osmosis is an efficient method for removal of humic-like substance while it is less efficient for removal of fatty acids. The method is therefore more suitable for treatment of leachates coming from landfills in the methanogenic phase than from landfills in the acidogenic phase (De Walle and Chian 1977). The largest disadvantage with reverse osmosis is the concentrate, which is created during filtration. This concentrate has to be taken car of somehow either by evaporation or landfilling. Landfilling of the concentrate has been questioned since the knowledge about how the concentrate affects the processes of the landfill is scarce. The water balance over the landfill is affected by landfilling of the concentrate, which can create problems (Woelders 1994). Other disadvantages with reverse osmosis are that the membranes can be clogged due to calcium carbonate and that small molecules such as ammonium ions and some chlorinated organic compounds are not so efficiently removed by this method (Collivinarelli et al. 1993, Linde 1995, Heyer et al. 1998). Therefore, the process has to be built in two steps or combined with a microbial treatment in many cases in order to obtain an efficient removal of ammonium ions. It might also be necessary to adjust the pH of the leachate before treatment in order to avoid clogging of the membranes.

Adsorption to activated carbon

Adsorption to activated carbon is a process, which has been used to remove some hydrophobic organic compounds from wastewaters. This process has been shown to efficiently remove some aromatic compounds (Rennerfelt and Ulmgren 1975). The carbon used can be in a granular form or in the form of a powder. An advantage with granular carbon is that it can be regenerated (Pinker 1997). Adsorption to activated carbon has been shown to be most efficient for humic compounds with a molecular weight between 100 and 10000. Fatty acids and humic compounds, which are heavier, adsorbs badly.

Precipitation

Precipitation is commonly used for removal of phosphorous from municipal wastewaters. This method has however also been shown to be useful for the reduction of the concentration of some organic compounds (Rennerfelt and Ulmgren 1975). A study has therefore been performed on precipitation of stabilized leachates with iron chloride (Amokrane et al. 1997). A COD removal of 55% was obtained. Another study compared precipitation of municipal wastewater alone with precipitation of a mixture of leachate and municipal sewage (Ettala 1989). The results showed that the concentration of aluminium sulphate had to be doubled when the leachate content was increased from 0 to 5%. The experiments did also show that the amount of leachate should not be above 1-2%.

Chemical or electrochemical oxidation

Chemicals that have been used for oxidation of organic compounds in leachates include e.g. Fentons reagent (hydrogen peroxide and iron), ozone and sodium hypochlorite (Huang and Fillos 1992, Anagiotou et al. 1993, Banerjee and O´toole 1995, Gau and Chang 1996). Some organic compounds are totally degraded in this kind of processes, which is an advantage. Furthermore, the compounds, which are not totally degraded, might be degraded to more bioavailable substances after oxidation (Huang and Fillos 1992, Anagiotou et al. 1993). There is however a risk for oxidation of certain organic compounds leading to production of substances, which are even more toxic than the original ones. The risk is especially large when chlorinated reagents are used (e.g. sodium hypochlorite) since chlorinated compounds can be produced during the oxidation process (Banerjee and O`toole 1995). COD and total organic carbon (TOC) removals between 22 and 78% have been reported (Huang and Fillos 1992, Anagiotou et al. 1993, Loizidou et al. 1993).

There are also other oxidation methods, which have been tested on leachates such as electrochemical oxidation and radiation (Yamazaki et al. 1981, Cossu et al. 1997, Tsai et al. 1997). Electrochemical oxidation has been shown to be a competitive method for oxidation of organic compounds as well as ammonium in leachates containing high concentrations of refractory organic substances (Cossu et al. 1997). COD removals of between 30 and 50% were obtained when two leachates from a deposit in Taiwan were treated (Tsai et al. 1997). Yamazaki et al. 1981 used Y-radiation from a Co^{60} lamp in order to treat a leachate. The leachate was aerated during the treatment. The treatment efficiency was clearly depending on the aeration intensity. A TOC removal of 80% was obtained at an aeration intensity of 400 ml/min and a dose of 32 Mrad.

Evaporation processes

A full-scale evaporation process has been built at a sanitary landfill in Finland (Ettala 1997). The approach is to let leachate evaporate on a thin plastic film. The energy consumption of the process is relatively low since only small temperature and pressure differences are utilized. Due to the low-pressure difference, fans instead of compressors can be used. The capacity of the process is 130 m^3/d. The COD_{cr} removal is more than 87%, and the NH_4^+-N removal 99.9%. The high ammonium removal is accomplished through adjusting the pH of the leachate to 4, prior to treatment. The leachate is pre-treated in a sand filter. Two disadvantages of the evaporation process are that a concentrate is created, one that needs to be taken care of, and that the low pH has to be adjusted before the treated leachate is discharged.

Two other evaporation processes have been developed in the US and in Italy, respectively (Cross et al. 1997), both processes using landfill gas as the energy source. Most of the organic substances are stripped from the leachate and transferred to the vapour phase. The vapour phase might need to be further treated by e.g. thermal oxidation.

A COMBINATION OF DIFFERENT METHODS

A combination of microbial and physical/chemical methods is needed in many cases in order to get an efficient treatment process for municipal landfill leachates in regard to nitrogen containing compounds, refractory organic substances as well as heavy metals.

Some studies have been performed on a combination of adsorption to activated carbon and microbial treatment (Copa and Meidl 1986, Lankford and Eckenfelder 1990, Horan et al. 1997). The retention time of the organic compounds in the process is increased since they are adsorbed onto the carbon. This increases the chances for the microorganisms to be able to degrade the pollutants. Furthermore, the nitrification can be favoured since some toxic compounds might be adsorbed onto the carbon (Lankford and Eckenfelder 1990, Horan et al. 1997). Another study of a process consisting of an aerobic treatment (nitrification), precipitation-flocculation and adsorption onto activated carbon showed that a TOC removal of 96% was obtained when a methanogenic leachate was treated (Welander and Henrysson 1998b). The TOC concentration of the untreated leachate was around 150 mg/l.

A combination of processes that have been tested is the activated sludge process and adsorption onto activated carbon. Powdered activated carbon (PAC) has then been added to the activated sludge process, which either can be performed in a continuous or batch mode (Copa and Meidl 1986, Aktas-Ozgur and Cecen-Ferhan 2001). The COD removal was increased when PAC was added in the last study due to mainly the none-biodegradable fraction being removed. The nitrification was also enhanced when PAC was added. However, the second step of the nitrification was inhibited (all the batches contained high amounts of NO_2^--N). Another combination of process, which has been tested, is nano-filtration of pre-treated leachate (Trebouet et al. 2001). The pre-treatment consisted of several steps (pH-modification, pre-filtration and coagulation with $FeCl_3$). The results showed that nano-filtration is sufficient for elimination of refractory chemical oxygen demand.

A process consisting of aerobic/anaerobic treatment followed by a rotating contactor, flocculation-sedimentation and reverse osmosis was studied in regard to removal of organic material. The overall efficiency in regard to removal of organics was about 98% (Park et al. 2001). Most of the organic material (95%) was found in a fraction with a molecular weight less than 500. The high molecular weight substances were efficiently removed by reverse osmosis. The final effluent showed an unexpectedly high oxygen demand according to the authors (Park et al. 2001).

The combination of a sophisticated membrane bioreactor and reverse osmosis has been shown to remove more than 99% of the total nitrogen (Kjeldahl-nitrogen 500 mg/l in the inlet) (Cornelissen et al. 2000). Furthermore, the COD concentration was decreased from between 3000 and 7000 mg/l to below detection limit. The BOD/COD ratio of the leachate was 0.35.

CONCLUSIONS

It has been shown that processes consisting of different combinations of methods can be efficient for treatment of leachates from municipal landfills. The evaluation of the treatment efficiencies in regard to organic compounds has however in most cases been performed through measurements of parameters such as COD, BOD and TOC. These parameters do not say anything about the toxicity of the compounds which are removed respectively left in the leachates. Neither do they give any information about the ability of the compounds to accumulate in living tissues. An evaluation of the treatment efficiency in regard to removal of organic compounds should therefore include some other parameters such as the hydrophobicity/hydrophilicity of the compounds and the molecular weight. These parameters are important factors for the ability of the compound to accumulate in living organisms. These kind of analyses is of importance since it is impossible to identify every remaining compound in treated leachates.

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