# OPTIMISATION OF N REMOVAL IN LANDFILL LEACHATES TREATMENT WITH MEMBRANE BIOREACTOR: PILOT PLANT AND FULL SCALE STUDIES

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SUMMARY: Biological treatment of landfill leachates was followed during a year in a membrane bioreactor (MBR) facility located in east Luxembourg. Good removal performances were observed but at high aerated volumes ratio. A pilot scale MBR was put into operation, treating the same leachates to demonstrate that similar performances could be reached with lower aeration volumes (lower cost). Results show that a removal of 99.4% of ammonia could be maintained with an aeration volume decrease from more than 75% to near 5% of the total volume. TN removal was also enhanced with no external carbon addition. Simulations with activated sludge model nº1 were performed with good predictions when aeration was high. When aeration was lowered, ASM1 predictions failed, so an adapted model for the particular case of nitrogen removal was tested. The adapted model that includes two-step nitrification and anammox activity shows better results, demonstrating the potential of these processes in landfill leachates biological treatment.

## 1. INTRODUCTION

Landfill constitutes the most common technique used worldwide for waste disposal (Salem et al., 2008). Upon its economic advantages, it minimizes the negative impacts on the environment and allows waste to decompose under controlled conditions. The landfill is in simple terms a big reactor, impermeable (watertight) at the bottom and semi-impermeable at the top, where crouched and compacted wastes are disposed for biochemical transformation until stabilization. The main effluents of this kind of bioreactors are leachates and biogas. The leachates, which treatment is the focus of this research work, are defined as the aqueous effluent generated as a consequence of rainwater percolation through wastes, biochemical processes in waste cells and the inherent water content of waste itself (Renou et al., 2008). It's a highly polluted liquid with variable composition that threats surface and groundwater if directly discharged (Bloor and Banks, 2003. It can contain an important amount of organic matter; biodegradable but also refractory compounds (humic and fulvic acids) (Vasel and Jupsin, 2003); high ammonia nitrogen concentrations; some heavy metals; and numerous other substances that can be considered as pollutants. Another associated difficulty are the enormous variations in composition and flows, that depends on many parameters such as time of disposal (age of the landfill); quantity of

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precipitation; temperatures; waste type and composition; disposal technique; etc. To reduce pollution content in this wastewaters, divers treatments exist going from physical/chemical techniques to biological treatments and several combinations of them (Laitinen et al., 2006; Bohdziewicz et al., 2008). Nowadays, the membrane separation techniques associated with an activated sludge in a membrane bioreactor (MBR) allows excellent removal performances, and it's a common technique used in real facilities (Praet et al., 2000). The sludge is composed of autotrophic bacteria that consume ammonia nitrogen and use it for growth (nitrification), producing nitrite and nitrate. Heterotrophic bacteria are also present consuming organic matter with nitrite, nitrate (denitrification) or oxygen as electron acceptor depending on the applied conditions. Aeration, that supplies the oxygen needed for nitrification is generally applied in excess in order to completely eliminate ammonia, despite the high cost associated. Furthermore, with high aeration, denitrification is inhibited and high amounts of nitrates are not consumed. This may lead to an effluent with high nitrate concentration that is now focused by European disposal restrictions. Indeed, nitrates may contribute to the eutrophication of aquatic environment (Jokela et al., 2002).

The objective of this research work is to show that it is possible to maintain good removal performances with lower aerated volumes in a membrane bioreactor treating landfill leachates. Energy used in excessive aeration can be saved. Denitrification, particularly endogenic is enhanced as well as other strain activities as anammox bacteria. Hence, outlet nitrate concentration is reduced. To this purpose, a pilot MBR was built and put into operation. The results obtained were contrasted with the luxembourgish full scale facility data. Furthermore, simulations with ASM1 model and a nitrogen removal adapted model, including two-step nitrification and anammox bacteria were performed to clarify the processes involved and their role in leachate treatment by membrane bioreactors.

## 2. MATERIALS AND METHODS

### 2.1 Muertendall MBR

The leachates used in this study come from Muertendall´s MSW sanitary landfill located in East Luxembourg, which operates since 1984. The facility is operated by the SIGRE syndicate (Syndicat Intercommunal pour la gestion des déchets ménagers, encombrants et assimilés en provenance des communes de la région de Grevenmacher, Remich et Echternach) and receives the household waste from 25 municipalities (50000 inhabitants) corrresponding to 11.5% of Luxembourg´s population. The bottom geomembrane which collects the leachates was installed between 1995 and 1998 and a membrane bioreactor for the on-site treatment is in operation since 2005. MBR configuration comprises an anoxic volume of $42m^3$, followed by two aerated tanks equivalent to $130m^3$. The aerated zone corresponds to a 75.6% of the total volume. Membranes are used in a sidestream configuration. The flows of leachates treated are variable. Often, values close to $25m^3$ per day are reported but they can rise up to $100m^3$. The leacheates feed an (MBR) activated sludge with an average VSS concentration of 14.2g/l. The hydraulic residence time varies also from less than 8,6 days to values near to 2 days. In average, 1.8mg/l of dissolved oxygen is measured in the aerated tanks. The MBR is located in an insulated structure that allows rather constant temperature conditions. Eventually, an external carbon source is added in order to boost denitrification, pH can be controlled that way through alkalinity (Melcer et al., 2003). However, this pH control is not used frequently because raw leachates have a rather high alkalinity (average 47.5meq/l).

## 2.2 Pilot MBR

The pilot MBR is composed of three PVC tanks. The first tank has a volume of 238 liters and operates under anoxic conditions. A mechanical artefact is installed to provide mixing of the sludge with the incoming leachates and the occasional external carbon source addition. The second (238 liters) tank is aerated by means of two disc diffusers (Passavant Intech, Roeflex®) and alimented by an electric air pump. The third tank of 25.5 liters is where the membrane filtration occurs (Zenon, ZeeWeed®-10, 0.93m$^2$ with a mean pore diameter of 0.1μm). Air is also pumped below the membranes to decrease the clogging effect. The pilot configuration is therefore an anoxic/aerated/membrane. A fourth tank is necessary to store treated water for the membrane backwashing (B-S tank). No significant bacterial activity is supposed to be present in this tank where the treated leachates samples are taken for laboratory analysis. The recycle is guarantied by two magnetic pumps (Iwaki, MD-6-230GS). The filtration and backwash are performed by the same diaphragm pump (Shurflo®, 75420-17) by means of an electronic valve system. Trans-membrane pressure and sludge flows are continually recorded by Endress-Hauser equipment (Promag 50, Delta bar).

## 2.3 Measuring campaign

Two measuring campaigns were performed, the first during January, February and March 2009, and the second between August and November 2009. Samples were collected once per week including influent, and effluent. The influent and effluent samples were analysed for ammonium nitrogen ($NH_4^+$-N); nitrites ($NO_2^-$-N); nitrates ($NO_3^-$-N); filtered and non-filtered chemical oxygen demand (COD); 7 day biochemical oxygen demand ($BOD_7$); total nitrogen; temperature; pH; and alkalinity. BOD tests were performed with and without addition of a nitrification inhibitor (allylthiourea, ATU). The suspended solids; volatiles suspended solids; electrical conductivity; pH; and temperature were also measured in the sludge. All analyses were made via standard methods. In general, samples were analysed just after they were taken in the interest of eliminating time related interferences.

## 2.4 Simulations with ASM1

Simulations were performed using the wastewater treatment simulator WEST®. ASM1 model was chosen to perform initial simulations, even if some trials were also executed with ASM3. Similar results were found with both models so it was decided to continue with the simplest one (ASM1). The calibration procedure applied included oxygen transfer characterization and a tracing test for hydrodynamics validation. Further details of the simulation work could be found in (Galleguillos et al., 2011). The characterization used to define the leachate composition in terms of ASM partitioning of material was also presented formerly (Galleguillos and Vasel 2011). It was based on a physical-chemical method combined with BOD analysis for the COD fractions and on standard analysis for nitrogen forms.

## 2.5 Simulations with autotrophic removal model (ASM1e)

A special adaptation of the ASM1 model for autotrophic removal (Van Hulle, 2005) that incorporates nitritation by ammonia oxidizing bacteria and nitratation by nitrite oxidizing bacteria and Anammox bacteria was used, particularly for the second part of the experiment in which aeration is reduced. Heterotrophic activity is also included in the model as it is often the case in nitrogen removal bioreactors. Bacterial growth was modelled according to ASM1 as well as decay. Endogenous respiration was not considered because there are not clearly documented models for ammonium and nitrite oxidizers and Anammox. The model is composed of eleven processes: Hydrolysis of entrapped organics, growth and decay of ammonia oxidizers, growth and decay of nitrite oxidizers, growth and decay of anammox bacteria, and growth and decay of heterotrophic biomass, including anoxic growth on nitrates and on nitrites. Default parameters were used and temperature dependency was eliminated because in our case, constant temperature was kept close to 20ºC during all the experiment. For further details about the model, the components, the representation matrix and default parameters refer to (Van Hulle, 2005).

## 3. RESULTS

### 3.1 Muertendall MBR

Raw leachates contain 281mgN-$NH_4^+$/l in average with a slightly increase during summer months (Figure 1). Even with values up to 700mgN/l, a 99.98% removal of N-$NH_4^+$ is maintained. Ammonia nitrogen is well transformed to nitrate; however, considering the total nitrogen removal in Figure 2, clearly denitrification is not achieved. An average removal of 43.6% of total nitrogen reveals that nitrogen is leaving the system as nitrates, instead of being transformed into nitrogen gas through denitrification. This could be partly explained by the lack of biodegradable carbon source in leachates (Wiszniowski et al., 2007) necessary for the heterotrophic bacteria to grow or the lack of anoxic conditions in which bacteria consume nitrates instead of oxygen as electron acceptor.

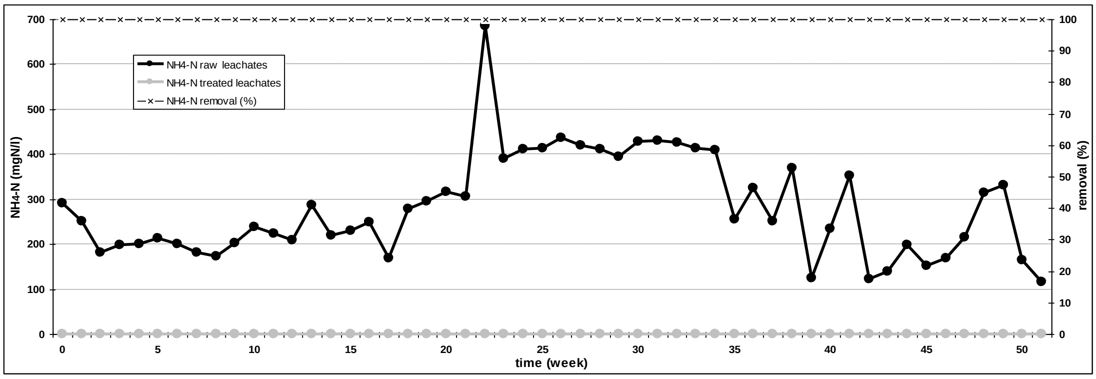


Figure 1. Inlet and outlet ammonia of Muertendall´s MBR with percentage of removal.

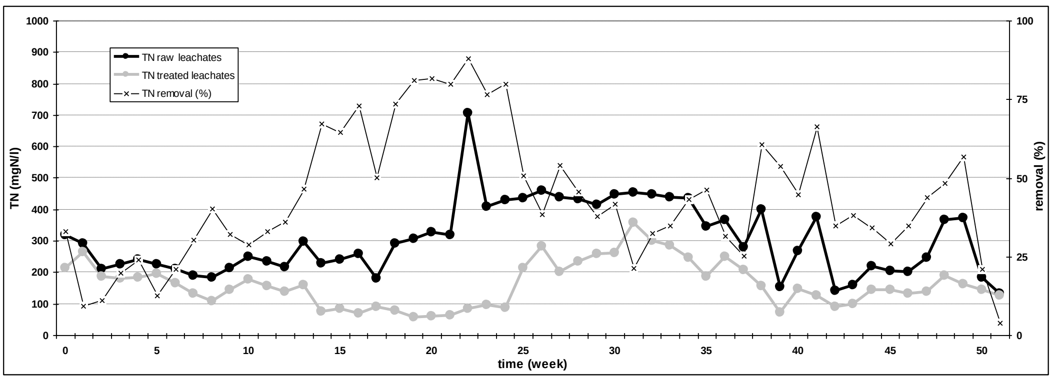


Figure 2. Inlet and outlet TN of Muertendall´s MBR with percentage of removal (TN calculated as ammonia+nitrites+nitrates).

Periods of high total nitrogen removal are present, contrasting with general low nitrogen removal. A possible explanation could be the external carbon source addition; however, there is no consistent data about this because carbon addition was only performed to control pH through alkalinity and not properly followed with flows and concentration data.

Inlet COD presents concentrations in a range from 1000mg/l to more than 3500mg/l also with a slightly increase during summer months (Figure 3). Figure 4 shows leachates rapidely biodegradable COD aproximated with BOD5 analysis. Clearly, the COD present in raw leachates is mainly non biodegradable or at least slowly biodegradable (ratio $BOD_5$/COD = 0.3 in average). Nevertheless, a 58.5% COD removal is achieved, meaning that part of the "recalcitrant" fraction is consumed (Vasel and Jupsin, 2003), the outlet COD (645.44mgCOD/l in average) contains thus, low biodegradability ($BOD_5$ = 4.7mgBOD/l in average) respecting disposal restrictions. Occasional formation of foam was observed in aerated tanks representing extra operational costs to control it.

Based on mass balances, specific ammonia uptake rate (sAUR) and specific nitrate uptake rate (sNUR) were computed. Average sAUR found was 9.5mgNH4+-N·(d)-1·(gVSS)-1 and sNUR was 13.9mgNO3--N·(d)-1·(gVSS)-1.

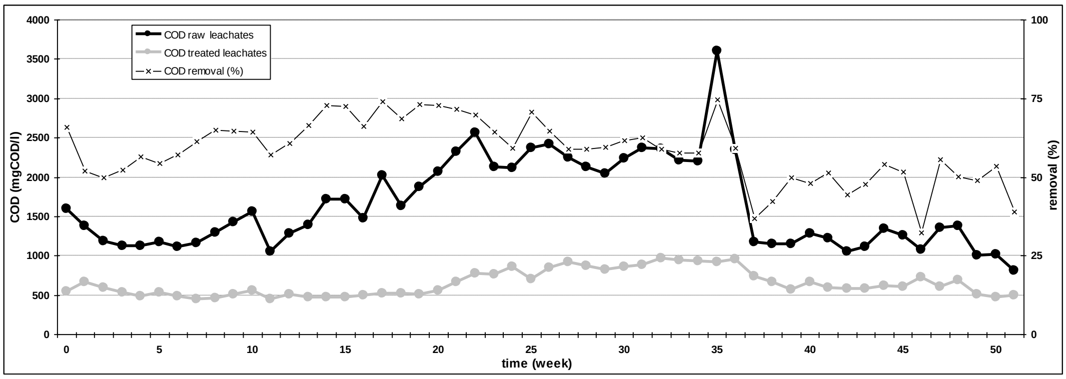


Figure 3. Inlet and outlet COD of Muertendall´s MBR with percentage of removal.

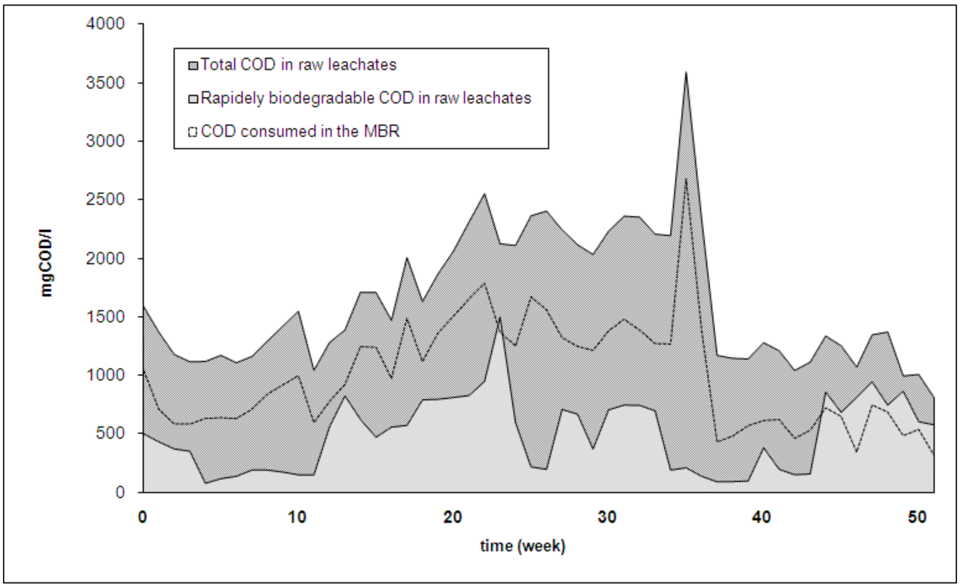


Figure 4. Inlet total COD in raw leachates, with biodegradable COD and consumed COD in MBR.

### 3.2 Pilotl MBR

During the first measurement campaign, the pilot was operated with an aerated volume corresponding to 52.5% of the total volume. The influent flow rate was fixed to 80 liters per day (3.58 liters per hour per square meter of membrane ($l{\cdot}h^{-1}{\cdot}m^{-2}$)). The sludge recycles ratio=3, thus 240 liters per day. This recycles flow ensures the recirculation of nitrates and the homogenisation of the mixed liquor micro-organism concentration in the system. No sludge extraction was performed, except for sample analysis. Volatile suspended solids increased from 2.3g/l at beginning to more than 10g/l during the second campaign. Ammonium nitrogen (Figure 5) was consumed almost completely (99%), contrasting with the case of total nitrogen. A 32.6% total nitrogen removal was measured. An external carbon source was added in order to boost denitrification. The theoretical optimum COD/N ratio needed is close to 4 but very dependent on the COD and N considered and on the conditions applied to the studied system (Komorowska-Kaufman and Klaczynski, 2005). In this case, acetic acid is used; 75ml were added daily starting on day 47 conducting to increase the COD/N ratio to values closer to 5. Total nitrogen removal increased rapidly to 81.7%. In the case of COD removal, a 35.4% was achieved, slightly increased until 48.4% during acetic acid injection (COD of external source not included).

Considering aerated VSS, average sAUR found was 16.9mgNH4+-N·(d)-1·(gVSS)-1. Specific nitrate uptake rate considering VSS in anoxic conditions was 9.4mgNO3--N·(d)-1·(gVSS)-1. Both values are rather similar to those measured on the real facility. After acetic acid injection, sAUR decreased to 10.6mgNH4+-N·(d)-1·(gVSS)-1and sNUR rised to 10.9mgNO3--N·(d)-1·(gVSS)-1 showing even more similarities.

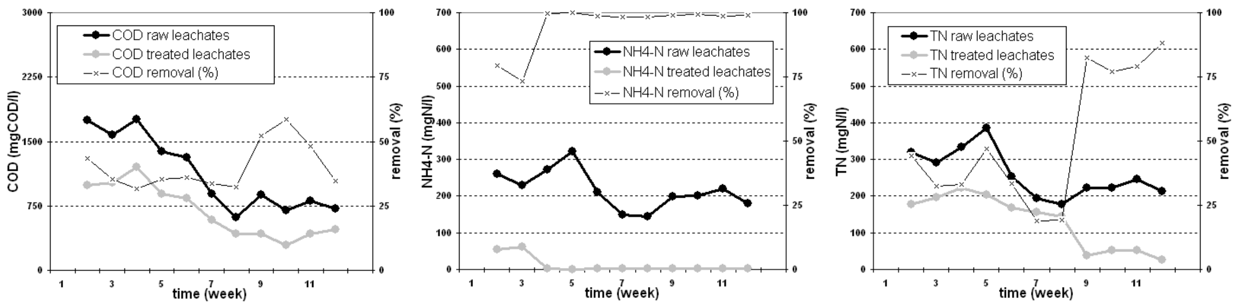


Figure 5. COD, NH4+ and TN in the raw and treated leachates with removal percentage during the first measuring campaign.

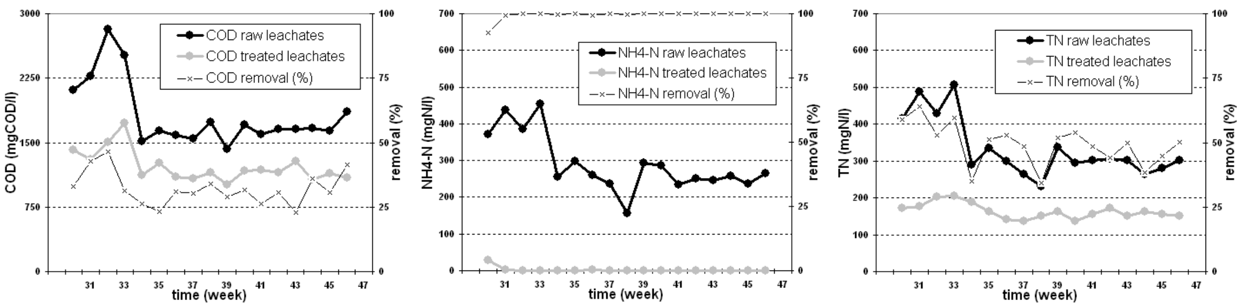


Figure 6. COD, NH4+ and TN in the raw and treated leachates with removal percentage during the second measuring campaign.

During the second measuring campaign, all operational parameters were maintained except the aerated volume which was decreased even more, reaching a 5.1% of total volume and the external carbon source was stopped. In these conditions, ammonia nitrogen continues to be consumed almost completely with 99.4% removal. TN removal increases, reaching a 49.3%, meaning that fewer nitrates are leaving the system. COD removal measured is 32.1% in average, slightly lower than during the first campaign. Average sAUR found was 94.7mgNH4+-N·(d)-1·(gVSS)-1, and sNUR equal to 3mgNO3--N·(d)-1·(gVSS)-1 .

### 3.3 Simulations

The results of simulation with ASM1 and ASM1e of outlet ammonia are shown in Figure 7. For the period before day 100, that is to say with large aeration volume, both models show similar results. During the second part of the experiment in which aeration volume is reduced, the ASM1e model represents better the outlet ammonia concentration.

According to the ASM1 model in which nitrification is modelled as one step and anammox bacteria is absent, dissolved oxygen concentration is not high enough to ensure that ammonia will be consumed by the considered autotrophic bacteria. This is why ASM1 simulation shows concentrations close to 100mgN/l in treated leachates. The ASM1e simulation shows important amounts of ammonia leaving the system from day 210 to 250, but values obtained are anyway lower than those calculated with ASM1. From day 250, outlet ammonia is almost absent consequently, to the analytically measured data. The incorporation of nitrification in two steps, and the ammonia consumption by anammox bacteria are important processes to consider because they may have an important role in low aerated bioreactors.

The results of simulations and the analytical measurement of the outlet total nitrogen (calculated as the sum of nitrates, nitrites and ammonia nitrogen) are presented in Figure 8. Both models follow in a satisfactory way the tendency of the measured values but the ASM1e is more accurate, particularly for the second part in which aeration is reduced. Again, the effect of ammonia consumption by anammox bacteria could be responsible for the differences. Furthermore, it is possible that the incorporation of ammonia oxidizers separated from nitrites oxidizers (two-step nitrification) can lead to a nitrate shunt. In this situation, oxygen needs can be reduced by 25% and the reduction of nitrite to nitrogen gas requires 40% less carbon sources (Henze et al., 2009).

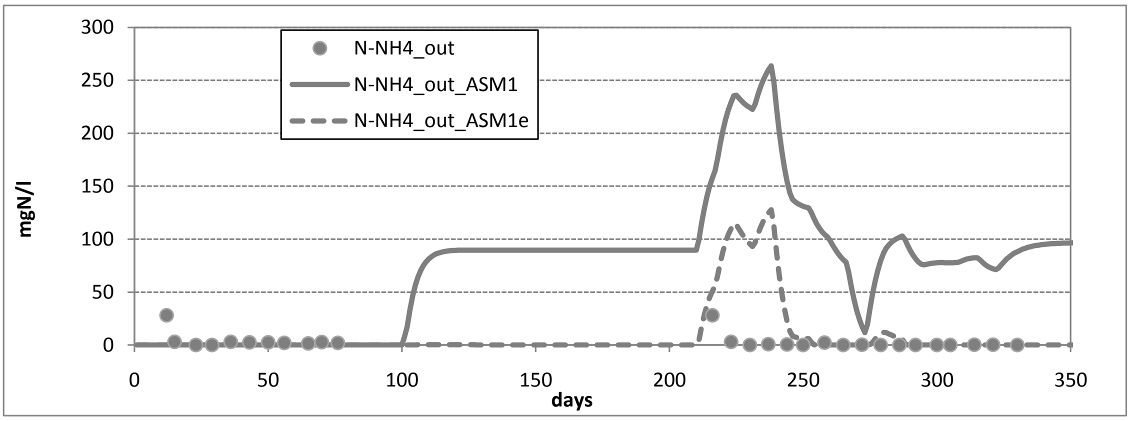


Figure 1. Simulation of outlet ammonia nitrogen with ASM1 and ASM1e.

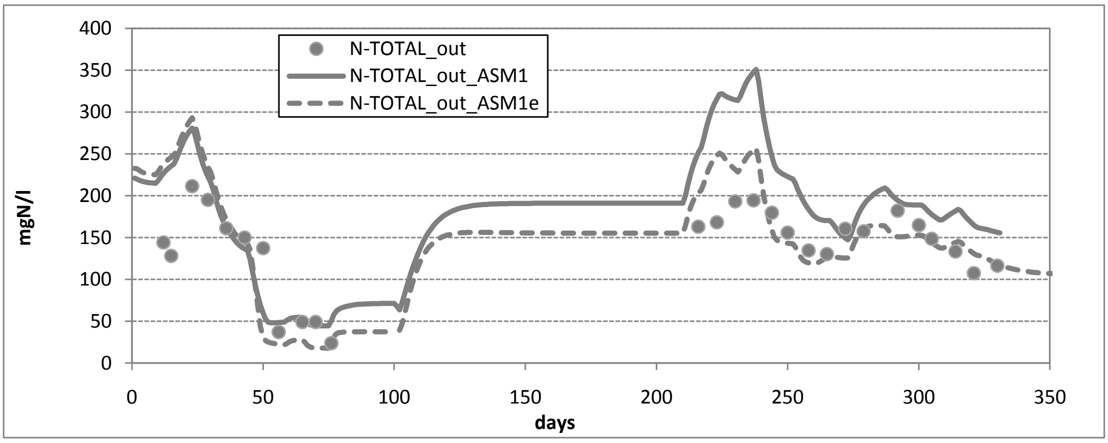


Figure 2 Simulation of outlet TN with ASM1 and ASM1e (TN is calculated as ammonia+nitrites+nitrates).

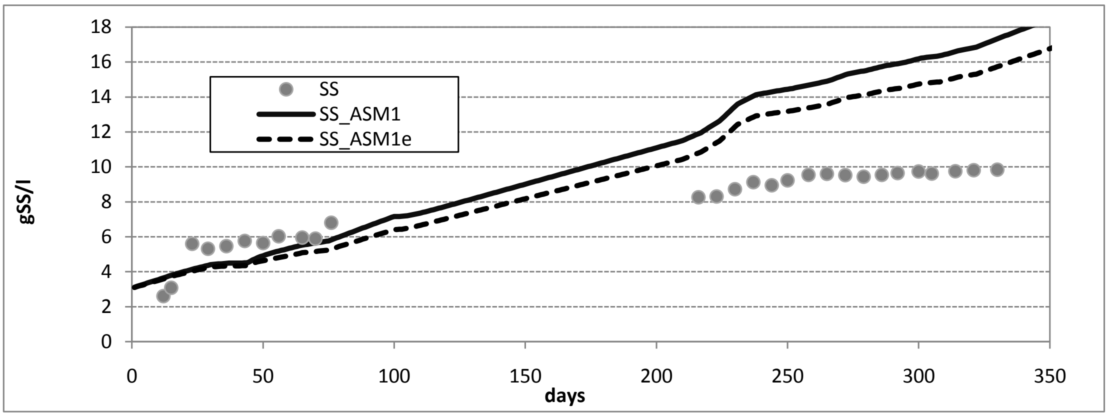


Figure 3 Simulation of suspended solids with ASM1 and ASM1e.

Additionally, the nitritation based process has the advantage of producing less sludge (approximately 40% less) (Henze et al., 2009). This can be appreciated in Figure 9 in which simulated and measured suspended solids (SS) are plotted. ASM1e produces less SS than ASM1. However, it must be noticed that both models overestimate the values obtained by analytical measurement. Clearly, some important processes must be missing in the simulation of SS and so in the overall model. According to both models, SS is mostly composed of particulate inert organics which concentration in the bioreactor increases, particularly by biomass decay but also by inlet leachates content. This model assumption appears to be false and some consumption must be taking place. It is possible that due to very high sludge age obtained in MBRs, the microorganisms able to degrade refractory COD compounds are able to grow in the reactor (Vasel and Jupsin, 2003).

## 4. DISCUSSION

Activated sludges are adaptable micro-organism communities that canl consume several substances present in wastewater, depending on conditions applied. Consumption performances will change accordingly to micro-organism behaviour which depends on many different operational parameters. In our case, bacterial communities are primary responsible for pollutant consumption, particularly, autotrophic bacteria that consume the ammonia nitrogen using oxygen. As mentioned before, ammonia nitrogen is transformed into nitrite, and then to nitrate through nitrification. Taking a look into Muertendall´s facility performances, a 99.9% of $NH_4^+$ removal reveals that the aeration is applied in excess. Indeed, the primary treatment objective is to reduce ammonia concentration, so excess aeration is well applied. Nevertheless, the problem related, is that ammonia reduction by nitrification leads to nitrate formation. The outlet nitrates concentration of the Muertendall facility reaches 161.5mgN/l in average, a high value that could be avoided.

Heterotrophic bacteria are the second important bacterial group present in the sludge. It consumes organic matter (measured as COD), and oxygen or nitrates as electron acceptor depending on conditions applied. When excess oxygen is used, nitrates will practically not be consumed, a situation that may explain low TN removal (43.6%). A low biodegradable carbon in leachates must be considered also, taking into consideration the occasional external carbon addition.

With the MBR pilot, excellent ammonia removal was maintained with an aeration volume kept near 52.5% of the total volume. However, TN removal was lower. Lack of biodegradability of leachate´s COD was a plausible explanation, which was tested and demonstrated by external carbon addition in the anoxic tank that boosted TN and COD removal. Afterwards, it was decided to eliminate external carbon source addition.

During the final campaign, a 99.4% of NH4+ removal was obtained, even with a much lower aerated volume of 5.1%. At the same time, the anoxic volume was larger and nitrate consumption was enhanced. COD removal, however, returns to lower values. sAUR computed values suggest that ammonia oxidizing bacteria consume more nitrogen under these conditions but the presence of other bacterial groups as anaerobic ammonium oxidising bacteria (anammox) could be responsible for ammonia consumption as well (Ganigué et al., 2007). Furthermore, partial nitrification may give some answers concerning lower needs of aeration, and lower carbon consumption (Iacopozzi et al., 2007). Another advantage of small aerated ratio is the elimination of foam, problem also detected in the Muertendall station.

It must be underlined that low aeration may have some influence over membrane filtration (clogging effect)(Defrance et al., 2000), but this issue was not considered within this study, so further research is needed to clarify this aspect. At least we did not observe any difference during the tests.

Table 1. Removal performances of MBR treating landfill leachates under different aerated volumes.

| | $NH_4^+$-N removal: | COD removal: | Total nitrogen removal: |
|---|---|---|---|
| Aerated volume: 75.6%<br>With external carbon source | 99.9% | 58.5% | 43.6% |
| Aerated volume: 52.5%<br>Without external carbon source | 99% | 35.4% | 32.6% |
| Aerated volume: 52.5%<br>With external carbon source | 99.9% | 48.4% | 81.7% |
| Aerated volume: 5.1%<br>Without external carbon source | 99.4% | 32.1% | 49.3% |

## 5. CONCLUSIONS

Muertendall´s MBR which is operated with a large aerated volume (75.6%), presents an excellent removal of ammonia nitrogen. Nevertheless, total nitrogen removals are low, indicating high nitrate concentrations contained in treated wastewater. In the case of organic matter, good removal performances are observed, considering the low biodegradability of leachates.

With the MBR pilot, the aeration volume was first reduced to 52.5%. Under these conditions, excellent ammonia nitrogen removal was maintained, but total nitrogen and COD removal decreases. To increase denitrification, an external carbon source was added. Total nitrogen removal rapidly increases to values up to 81.7%, as well as COD removal that rises up to 48.5%.

During the final part of the study, no external carbon source was added and the aeration volume was decreased to reach a 5.1% of total volume. Under these conditions, Ammonia nitrogen is still consumed over 99%, and TN removal increases to values higher than the ones found in the full scale facility. The aeration, with its associated costs can be lowered, with similar nitrogen removal performances. In the case of COD removal, a slightly decrease was observed and must be considered. External carbon source addition under these low aerated conditions can be tested.

Simulations performed with ASM1 and the nitrogen removal adapted model ASM1e enhances the active role of different autotrophic strains. Particularly, the inclusion of ammonia oxidizers (separated from nitrites oxidizers) and anammox bacteria which appear to be necessary in low aeration conditions. Also, the high sludge age encountered in MBR allows the growth of micro-organisms capable of consuming refractory compounds so they could be considered when modeling these processes.

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