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LANDFILL LEACHATE AMMONIA REMOVAL IN SUBMERGED AERATED FILTER: A FULL SCALE STUDY UNDER NON-ALKALINIZATION CONDITION

Ricardo Figueira Bidone * and Claudio Fernando Mahler

COPPE/UFRJ, Federal University of Rio de Janeiro, COPPE/Civil Engineering Program, Rio de Janeiro/RJ, Brazil

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ABSTRACT

This study presents the performance of submerged aerated filter technology in landfill leachate treatment. The main objective was to evaluate ammonia removal. The investigation proposes the filter, without the use of chemical agents to alter the pH, as an applicable mechanism for nitrogen removal in the treatment. The research was developed in real-scale. The efficiencies obtained, for a hydraulic retention time of 56 days, were: 83% removal of ammoniacal nitrogen (influent concentration of 2,653 mg/L) and 78% removal of total Kjeldahl nitrogen (influent of 3,058 mg/L). The pH variation range was from 8.3 to 9.3. The alkalinity showed a reduction from 12,338 mg/L to 4,015 mg/L. The dissolved oxygen concentration ranged from 0 to 8.3 mg/L. Additionally, the efficiencies in removal organic matter were: 35% removal of COD (influent of 5,022 mg/L), 45% BOD removal (influent of 2,400 mg/L) and 40% TOC removal (influent of 2,141 mg/L). The expulsion of ammonia was great. The oxidation of organic matter, even after the establishment of a very favorable aerobic condition and despite the long liquid retention time, encountered difficulty in the extremely varied and complex composition of the leachate. The research hypothesis was positively confirmed: there is no need to alkalize the medium to obtain high ammonia expulsion in the leachate. The removal, in terms of load, was quite significant: 2.6 tons.

1. INTRODUCTION

The concentration of ammoniacal nitrogen in landfill leachate generally reaches high values. In Chinese leachate, values of up to 3,100 mg/L were recorded (Ma et al., 2022). In Brazil, the leachate generated in the Bandeirantes and São João landfills, both in the state of São Paulo, presented concentrations of 2,390 mg/L and 2,870 mg/L, respectively (Mahler et al., 2020).

Ammoniacal nitrogen is composed of a gaseous fraction (NH_3) and an ion (ammonium, NH_4^+). The concentrations of each form depend on the pH of the liquid medium. For pH close to 9.2, there is 50% NH_3-N and 50% NH_4^+-N; for pH below 9.2, NH_3-N concentrations fall and NH_4^+-N rise proportionally; for pH greater than 9.2, NH_3-N concentrations rise and NH_4^+-N fall proportionally (Bidone, 2018). This behavior of chemical forms as a function of the variation in the pH can be seen in Figure 1.

The air stripping (or volatilization) of ammonia is a consequence of the forced introduction of air into the liquid medium. In this case, the physical effect of replacing, in the liquid mass, one gas (NH_3) with another (air) prevails. The classic methodology consists of raising the pH of the leachate to values close to 11 or higher (Ferraz et al., 2013), favoring the formation of ammoniacal nitrogen in the form of NH_3.

Alkalinization typically occurs under the action of the following agents: calcium hydroxide, $Ca(OH)_2$, and sodium hydroxide, NaOH (Gomes et al., 2023). After the appropriate pH condition has been established, the mechanical introduction of air into the liquid medium takes place, determining that, through the action of circulation of this mass of air, the expulsion or dragging of NH_3 occurs.

The process is usually carried out in towers or lagoons (Souto and Povinelli, 2007). The positive aspect of this technology is the possibility of obtaining ammonia removal to the desired level. The efficiency in removing ammona will basically depend on the amounts of alkalinizer and the hydraulic detention time used. One of the negative aspects is the need to adjust the pH to discharge the final treated effluent.

Calixto et al. (2021) carried out alkalinization of leachate. The study used calcium hydroxide and sodium hydroxide as alkalizing agents. Both agents satisfactorily raised the pH. There was a reduction in color (clarification of the leachate). They concluded that alkalinization is, in-

* Corresponding author:
Ricardo Figueira Bidone
email: ricardo.bidone@coc.ufrj.br

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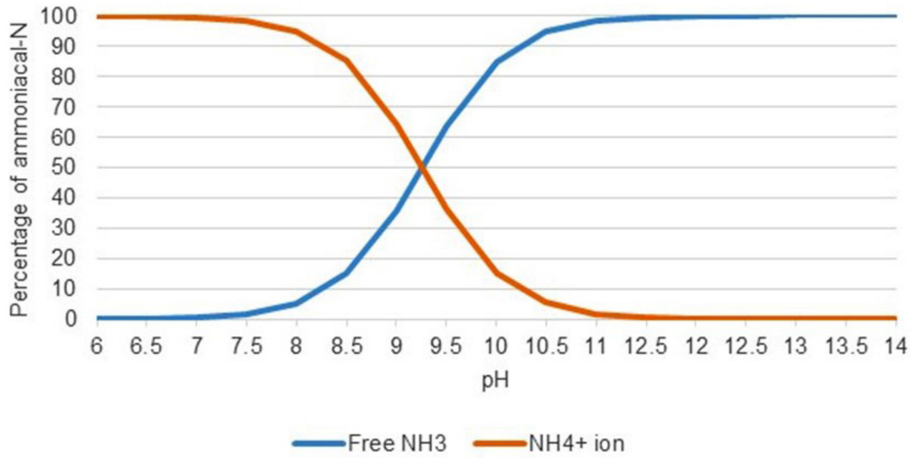

FIGURE 1: Proportion of NH_3 and NH_4+ as a function of pH value variations (Bidone, 2018).

deed, an option to be considered for pre-treatment, especially when preceding stages that perform better at high pH.

Frederice (2019) studied air stripping removal of ammonia in leachate. The research was conducted on a laboratory scale. Two towers built with 150 mm diameter. The air stripping towers had a total height of 4 m; 3 m of which were filled with polypropylene. For NH_3-N initial concentrations of 2,874 mg/L and 2,415 mg/L, have been removed up to 83.4 and 85.5% of the ammonia, respectively, with pH equal to 11.4.

The subject of this study is the removal of ammonia from landfill leachate. The proposed technology is the submerged aerated filter. The main objective is to verify the level of efficiency of the filter in removing this form of nitrogen. The research hypothesis is that there is no need to alkalinize the leachate to achieve high efficiencies. The data were generated on a real scale. The study materials are non-replicable.

The proposed ammonia removal is a clean technology. It does not use chemicals and does not propose dilution of leachate (which would require a huge volume of water). In addition, the electrical energy used to drive (operate) the aeration system is generated within the landfill itself: there is an energy generation unit on site that uses the biogas obtained from the decomposition of waste disposed of in the landfill. Finally, it provides the opportunity to obtain a large quantity of the nutrient nitrogen. The study takes place at a time when Brazil is seeking to become a world reference in sustainability (Bidone et al., 2025).

The Submerged Aerated Filter (SAF) was built to be part of a continuous flow process train comprising: Upflow Anaerobic Fixed Bed Filter → SAF → Constructed Wetland. It is anticipated that significant biological activity will develop on the surfaces of the aerated filter stone, over time. The initial construction of the SAF offered a one-off opportunity to carry out a single batch study of ammonia stripping at full scale in the absence of significant biological activity. This study will inform future interpretation of the mass balance of nitrogen removal from the leachate when the full process train is operational and biological activity has developed.

2. MATERIALS AND METHODS

2.1 Materials

2.1.1 General description

The landfill from which the leachate was obtained is located in the municipality of Minas do Leão, in the state of Rio Grande do Sul, in the extreme south of Brazil. The state capital, Porto Alegre, is 80 km from the landfill. The landfill receives 50,000 tons of domestic solid waste from 150 municipalities per month.

The treatment of raw leachate is carried out in a system composed of two upflow anaerobic filters, operated in series, followed by a submerged aerated filter (unit in which this study was developed) and two constructed wetlands.

2.1.2 Submerged aerated filter features

The main characteristics of the submerged aerated filter, the treatment unit analyzed in this study, are:

- Useful volume (discounting the volume of the support medium): 1,213 m^3;
- Base area: 628 m^2;
- Liquid surface area: 1,200 m^2;
- Useful height: 2.7 m;
- Support medium (filter bed): gravel with the smallest dimension between 38 mm and 76 mm;
- Pipes for leachate distribution: in steel; 150 mm in diameter, perforated along the entire length;
- Number of blowers: 4 units (of 25 HP);
- Air distribution pipes: 4 in number; in stainless steel. Each of them, 150 mm in diameter, at the beginning of the aeration system (with reductions to 100 mm and 75 mm in diameter) and 50 mm in diameter at the end of the system (next to the bubble diffusers);
- Total installed power: 100 HP;
- Number of air diffusors (course bubble type): 360 units;
- Air flow through the tank: 117 m^3/min;
- Air flow per diffusor: 0.325 m^3/min;
- Hydraulic retention time for the batch: 56 days.

Table 1 presents a summary of the aeration conditions of the experiment.

TABLE 1: Summary of the aeration conditions of the experiment.

Item	Value	Units
Aeration rate	117	m^3/min
Filter working volume (in integer values)	1,200	m^3
Specific aeration rate ([117 x 60] ÷ 1,200) ([117 x 60 x 24 x 56 days] ÷ 1,200m^3)	5.85 7,862	(m^3 air/m^3 leachate)/hour m^3 air/m^3 leachate
Aeration power (4 x 25 HP motors)	100 (~75)	HP (kW)
Specific aeration energy ([56d x 24h x 75kW] ÷ 1200m^3)	84	kWh/m^3 leachate

In Figure 2, it is possible to observe a daily service front of the landfill. In Figures 3, 4 and 5 it is possible to observe, in detail, the construction of the submerged aerated filter and its operation.

2.2 Methods

This article evaluates ammonia removal, under non-alkalinization condition, in a submerged aerated filter. The efficiencies obtained in the filter were calculated by comparing the effluent concentrations and the influent concentration (value verified on Day of sample 0), at the same collection point. The test results for Day of sample 0 constitute the raw leachate. This means that: the liquid introduced into the filter, for the purpose of this study, did not undergo any prior treatment. The concentrations mentioned are presented in Table 3, in the Results.

The control variables of the experiment, those whose analysis allows answering the research hypothesis, were pH and ammoniacal nitrogen (ammoniacal-N). The leachate characterization variables were organic matter (BOD, COD and TOC), dissolved oxygen (DO), total Kjeldahal nitrogen (TKN), nitrate (NO_3^-) and alkalinity.

The filter operated in batch mode. The purpose of the batch mode was to determine the number of days required to achieve significant ammonia removal and, during this period, the maximum pH value that the liquid could reach without alkalinization.

From day 42 onwards, the possible removal began to show a tendency to stabilize. The pH also began to vary very little. To confirm this trend, observation of the experiment was continued until 56 days (eight full weeks) had been completed.

2.2.1 Physicochemical analysis

Table 2 presents the analytical methodology used in each physical-chemical analysis. The frequency and point of collection of the test samples are also included in the table. These analyses were performed by the Laboratory of Chemical Analysis Laborquímica Ltd. and followed the provisions of the Standard Methods for the Examination of Water and Wastewater (APHA, 2017).

3. RESULTS AND DISCUSSION

3.1 Influent and effluent concentrations

Table 3 presents the results of the experiment. All data generated for this article are listed there.

3.2 pH and alkalinity

The raw leachate had a pH of 8.3 (alkaline). On the 14th day of the experiment the pH had already reached 9.2. This

FIGURE 2: A daily service front of the landfill.

FIGURE 3: (a) Excavation and preparation of the base to begin construction of the filter; (b) start of installation of the waterproofing membrane; (c) welding of the membrane; (d) image of the progress of the waterproofing work; (e) preparation of forms for concreting the leachate pipe support; (f) spreading of gravel at the base of the filter.

FIGURE 4: (a) Leachate distribution piping and concrete beams (to support the air piping); (b) spreading of gravel over the liquid distribution piping; (c) image of the blower house; (d) view of the air distribution piping installation assembly; (e) installation of the last sections of the air piping; (f) detail of the bubble diffuser.

FIGURE 5: (a) Air distribution test and blower test; (b) filling the filter bed with gravel, after the successful aeration test; (c) another view of the air distribution piping, now with the filter completely filled with gravel; (d) view of the first author of this article, above the filter bed of the filter; (e) immediately after the beginning of operation at the filter.

TABLE 2: Analysis, frequency, collection point and method.

Variable	Frequency	Collection point	Methodology
pH	Weekly	Filter output	Direct Potentiometry
Ammoniacal-N ($mgN.L^{-1}$)	Weekly	Filter output	Titrimetry with prior distillation
TKN ($mgN.L^{-1}$)	Weekly	Filter output	Titrimetry with prior distillation
Nitrate ($mgNO_3^-.L^{-1}$)	Weekly	Filter output	Ultraviolet Spectrophotometric
DO ($mgO_2.L^{-1}$)	Weekly	Filter output	Titrimetry
Total alkalinity ($mgCaCO_3.L^{-1}$)	Weekly	Filter output	Neutralization titrimetry
$BOD_{5,20}$ ($mgO_2.L^{-1}$)	Weekly	Filter output	Membrane electrode potentiometry
COD ($mgO_2.L^{-1}$)	Weekly	Filter output	Open reflux dichromatometry
TOC ($mgC.L^{-1}$)	Weekly	Filter output	Titrimetry – permanganimetry

TABLE 3: Results of the analyses conducted during the batch process (results in mg/L, with the exception of pH).

Day of sample	pH	BOD	COD	TOC	DO	Ammoniacal-N	TKN	NO_3^-	Alkal.
0	8.3	2,400	5,022	2,141	0	2,653	3,058	0	12,338
7	8.9	1,800	4,247	1,750	6.3	2,734	3,035	0.5	12,101
14	9.2	1,600	3,940	1,602	8	2,124	2,558	6.7	11,233
21	9.2	1,500	3,885	1,621	7.2	2,102	2,161	28.3	10,560
28	9.4	2,048	5,120	2,181	9	1,389	1,668	28.3	9,987
35	9.3	1,700	3,740	1,595	7.9	1,147	1,411	18.7	9,576
42	9.5	1,600	3,361	1,378	8.4	654	1,176	10.4	5,348
49	9.3	1,250	3,293	1,517	8.4	645	796	8.6	4,413
56	9.3	1,310	3,276	1,275	8.4	460	658	15.9	4,015

increase occurred naturally/spontaneously (without the addition of alkalizing agents to the liquid medium). Values close to this (9.3, 9.4 and 9.5) were achieved and maintained consistently until the 56th day of the experiment. Aeration of the raw leachate, maintained for 56 days, induced an ion/gas imbalance ($NH_4^+ \rightarrow NH_3 + H^+$). This shift in equilibrium to the right in the equation favored the formation of NH_3 and allowed its subsequent dragging out of the liquid.

Alkalinity fell consistently over the 56 days, from 12,338 mg/L to 4,015 mg/L. It should be noted that a significant portion of the measured alkalinity is due to the presence of dissolved ammoniacal nitrogen. Therefore, the removal of NH_3 inevitably results in a reduction in alkalinity. It is worth mentioning that, even to a small extent, the release of H^+ ions, which occurs due to the small nitrification observed, also results in a reduction in alkalinity.

The pH values vary on a logarithmic scale and alkalinity on a decimal scale. Therefore, any increase in pH, even a small one, results in a not so small decrease in alkalinity.

The increase in pH observed in this research demonstrated that there is no need to use an alkalizing agent to reach a value of up to 9.5. This finding is quite important. Note that it was possible to remove 83% of ammoniacal nitrogen in the pH variation range of the experiment (between 8.3 and 9.3). This level of removal is quite significant.

The "traditional" ammonia removal would have consumed a large amount of alkalizing agent to obtain this percentage of removal. Worse still: the pH, through the "traditional" ammonia removal route, would have been raised to values above 11. In Brazil, the emission standard for the discharge of treated final effluent requires a pH close to neutrality. In this case, even more chemical agents would be consumed, this time to reduce the pH of the treated leachate.

It is important to address the hydraulic retention time in this experiment. "Traditional" ammonia removal, conducted in towers, aims to remove NH_3 in a short period of time (order of magnitude of hours). This is only possible because the pH is quite high (to values above 11) due to the use of a large amount of chemical agent, quickly making a lot of ammonia available for expulsion. It should be noted that the methodology studied in this article, in turn, demands a long liquid retention time (tens of days) because it advocates that no chemical agents are consumed. As a consequence of this, the maximum pH observed was equal to 9.5.

Therefore, there is no doubt that it is possible to raise pH without alkalinization by chemical agents and that it can, in fact, be associated with high ammonia removal. Certainly, depending on the other units that will make up the leachate treatment plants, this discovery, verified on a real scale, is useful for the treatment of industrial effluents.

3.3 Ammoniacal-N, TKN and nitrate

It is important, preliminarily, to mention that the standards and conditions for effluent discharge in Brazil are set forth in CONAMA Resolution 430/2011. The maximum permitted concentration for ammoniacal- N is 20 mg/L. Dilutions of leachate are not permitted.

On the 42nd day of the batch, ammoniacal-N removal had already reached 75%. On the last day (56th), removal reached 83%. The close relationship between ammonia removal, pH and alkalinity is indisputable. The increase in pH stimulated the formation of NH_3-N in the liquid, enabling its removal (by the action of air) and the removal of alkalinity (verified by the enormous reduction in its concentration).

These results allow the following interpretations: (1) the ammoniacal-N dragging trend was consistent and uncontroversial, given the regularity of the decline in concentrations; (2) for a hydraulic retention time of 56 days, under the aeration conditions of the experiment, the ammoniacal-N removal exceeded 80%, without the need for alkalinization of the leachate.

TKN showed a very similar decay behavior to ammoniacal-N. As expected, since TKN is the sum of organic nitrogen and ammoniacal-N. Its removal reached 78%.

Figure 6 shows the downward trends in ammoniacal-N and TKN. It also shows the rise in pH. The values are presented as a function of the 56 days of SAF operation.

Nitrate was generated in small quantities, considering the high availability of ammonia and the completely aerobic condition established in the liquid. However, it is important to note that there was some nitrification. This is not the subject of this article. The study of nitrification would involve the discussion of issues related to the active biomass in the filter and, in the case of leachate (extremely concentrated in terms of ammonia), the limiting concentrations of the pollutant. In this article, we focus on ammonia drag.

Regarding the 20 mg/L of ammoniacal-N, which is the effluent emission standard, it is worth noting that the landfill leachate treatment plant has two wetlands built downstream of the aerated filter. This is where the final stage of leachate treatment occurs. The macrophyte used in the wetlands is Typha subulata, a plant that avidly assimilates nitrogen. The design of this phytoremediation (treatment using wetlands) is another reason for not using chemical agents to remove ammonia. The performance of the macrophytes is not to be hindered/impaired. In the wetlands, the following are also expected: the physical effect of sedimentation of nitrogenous organic matter (because

TABLE 4: Ammoniacal-N removed charge.

Item	Value	Units
Ammoniacal-N removed, in terms of concentration (2,653 - 460)	2,193	mg/L
[2,193 mg/L ÷ 1,000,000 = 0.002193 kg/L]		
Filter working volume (in integer values)	1,200	m^3
[1,200 m³ x 1,000 L/m³ = 1,200,000 L]		
Removed charge ([0.002193 kg/L x 1,200,000 L]	2,631.6	kg of Ammoniacal-N

the wetlands are quite long and the flow velocity is low) and metabolization of soluble ammonium by the plants. The system, as a whole, will certainly generate treated leachate with an ammoniacal-N concentration of less than 20 mg/L.

Finally, regarding NH_3, it is important to emphasize that the gas extracted from the filter is not released into the atmosphere. It is recovered. The filter design included a cover for the unit. Under this cover there is a pipe that captures the NH_3 that was expelled from the liquid mass. The photographic record in this article concerns the first days of operation and does not show the construction phase of the unit's cover.

3.4 Ammoniacal-N removed charge

The removal of ammoniacal-N reached 83%. This percentage, in itself, is already interesting. However, considering the very significant volume of the submerged aerated filter, how much would this removal represent in terms of charge? Table 4 shows the calculation of the ammoniacal-N charge removed.

The amount removed simply exceeds 2.6 tons of ammoniacal-N. It is very important to remember that all this ammonia can be converted into fertilizer. Therefore, it is no exaggeration to say that this result is extraordinary. There is hardly a more sustainable cycle than this: from landfill leachate (a liquid byproduct of solid waste from human activity), the macronutrient essential for food production is made available in this magnitude. Dos Santos et al. (2020)

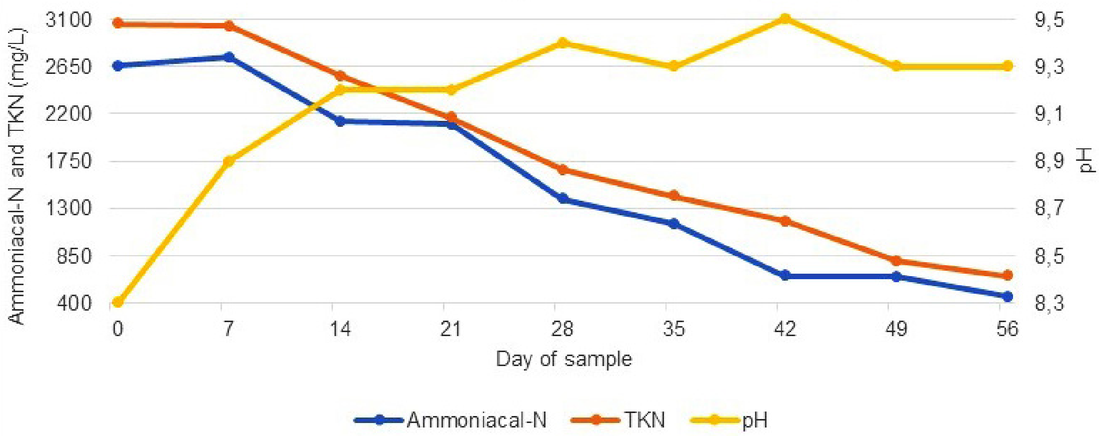

FIGURE 6: Ammoniacal-N, TKN and pH as a function of day of sample.

successfully investigated the recovery of ammonia-N from the chemical absorption of ammonia in phosphoric acid, resulting in ammonium phosphate (a chemical compound widely used to induce plant growth).

3.5 BOD, COD and TOC

The removal of organic matter was monitored during the 56 days of experimentation. It can be observed that BOD, COD and TOC showed a downward trend. The removals reached, respectively, for COD, TOC and BOD: 35%, 40% and 45%.

The removals obtained in terms of BOD, COD and TOC were not greater, possibly because the highly biodegradable organic matter was quickly stabilized (in the first days of the experiment). It is quite noticeable that the substrate became refractory to biological treatment. Only this would justify that, in the presence of so much air and for such a long hydraulic retention time, the oxidation of the organic matter was so limited. The pH of the raw leachate was equal to 8.3. It should be noted that, under these conditions, in the first days of the experiment, the concentration of ammoniacal-N, although considered high, did not seem to completely inhibit the process. However, as the pH increased, even though the ammoniacal-N continued to decay, the liquid medium conditions for the biota became unequivocally more unfavorable. Furthermore, the substrate removal rate, in terms of organic matter, is expected to be higher at the beginning of the process.

3.6 DO

The dissolved oxygen quickly provided the environment with a fully aerobic condition. This condition was maintained throughout the experiment. On the 28th day, the concentration reached the maximum value verified during the experiment: 9 mgO_2/L.

The air supply to the aerated filter was calculated so that it operates in continuous flow. This is the normal operating regime of the landfill. However, for the research hypothesis of this article (non-alkalinization of the leachate, naturally raising the pH and leading to a high ammonia removal) and taking advantage of the beginning of the filter operation, the batch experiment was evaluated as a unique opportunity.

It is true that the hydraulic retention time of the batch was high. This is because the batch would only be completed when a satisfactory level of ammonia removal was reached; this number of days was one of the research questions. The maximum value to be reached by the pH, without alkalinization by a chemical agent, was also a research question. The removal level established as satisfactory had been defined as 75%. This percentage was reached on the 42nd day of the batch.

Aeration was only maintained on the following days in order to ensure that the trend of the results would not be modified.

4. CONCLUSIONS

Thus, objectively, the number of days required to achieve a significant reduction (75%) of ammonia was equal to 42. During this period, the maximum pH value was equal to 9.5. Maintaining the batch for 56 days confirmed the stability of the ammonia reduction trend (whose total removal, in the entire period of the experiment, was equal to 83%).

The research hypothesis was positively confirmed. There is no need to alkalize the medium to obtain high ammonia expulsion in the leachate.

Any liquid that presents high concentrations of ammonia nitrogen could use an ammonia removal unit without alkalinization for subsequent recovery of the ammonia. The submerged aerated filter is a technology that deserves to be evaluated as part of a treatment system.

The removal of ammoniacal-N, in terms of charge, was quite significant: 2.6 tons. Only observation on a real scale allows verification of this magnitude.

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