

REMOVAL OF NITROGEN COMPOUNDS FROM LEACHATE FROM LANDFILLS FOR NON-HAZARDOUS WASTE

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ABSTRACT: The leachate from landfills usually contains high concentrations of ammonia, organic and toxic compounds, heavy metals. Due to the synergism of various factors it is difficult to predict the quantity and composition of landfill leachates. NH_4^+ - N is basic and long-term pollutant and cause acute toxicity. The purification of leachate with high ammonia content is one of the most difficult tasks in wastewater treatment worldwide. Several laboratory tests were conducted in order to establish optimal technological parameters for the processes in a treatment plant of removal of various contaminants for leachate from landfills for non-hazardous waste. There was performed an analysis of the processes in order to their optimization through model technological solutions implemented in a laboratory. There was established the influence of different leachate quantities from landfills for non-hazardous waste in the composition of treated wastewaters into the biological stage of technological scheme with preliminary denitrification and SBRs on the removal rates of nitrogen compounds and other pollutants from waters.

KEY WORDS: landfill leachate, non-hazardous waste, wastewater, biological nitrogen removal

1. INTRODUCTION

The leachates from landfills for non-hazardous wastes are a serious environmental problem worldwide. Due to the specifics of leachate genesis it typically contains high concentrations of ammonia, organic compounds, toxic compounds and heavy metals. The composition of the leachate varies as its characteristics may change over time due to the waste decomposition in landfills.

Among all leachate components NH_4^+ -N is the basic long-term pollutant and cause acute toxicity. The concentration of NH_4^+ -N is the main reason for the necessity of purification. Also, COD, BOD, heavy metals

and other substances must be reduced during treatment of leachate [2].

The most widely used methods in the world for the treatment of leachate with a high content of organic pollutants are biological methods for purification.

The treatment of leachate through biological methods is one of the most difficult problems associated with the wastewater treatment. While nitrification is usually easily feasible with more than 95% of ammonia removal through an exclusive application of the biological treatment, the reduction in COD is a significantly bigger challenge. The efficiency of COD reduction/removal ranges

from 90% to nearly 20%, depending on the characteristics of the leachate (origin and age). Due to the specifics of leachate and in order of more effective treatment through biological methods, the practice is its mixing with domestic wastewater, thus it may be achieved more favorable COD to BOD ratios in the mixed flow. The current researches having in mind this regard.

2. MATERIALS AND METHODS

The laboratory installation that was used for the presented experiments consisted of a denitrifying bioreactor and two consecutive SBRs (Figure 1) at a provided total residence time 11 days.

The denitrifying reactor (DN) was with a fixed biomass and an upward flow. 1100 g of natural occurred zeolite, clinoptilolite with 2.5 - 5.0 mm size fraction, was used as biomass carrier in the reactor for denitrification. The carrier was characterized with the following composition, %: SiO₂ - 67.96, Al₂O₃ - 11.23, Fe₂O₃ - 0.83, K₂O - 2.85, Na₂O - 0.74, CaO - 3.01, MgO - 0.06, TiO₂ - 0.90. The cation exchange capacity and exchangeable cations in meq/100g zeolite were respectively CEC - 112.75, K⁺ - 33.88,

Na⁺ - 21.01, Ca²⁺ - 63.48, Mg²⁺ - 2.68. The liquid phase of the bioreactor had volume of 850 cm³. The reactor for denitrification was inoculated with 40 ml enrichment mixed culture of denitrifying bacteria, obtained from activated sludge.

The two SBRs (SBR1 and SBR2) were cylindrical vessels with an identical design and equal volumes of 2400 cm³ each. They were inoculated with 200 ml activated sludge for wastewater treatment of ethanol distillery plant. The concentration of activated sludge in the two reactors, operating in batch modes, was maintained in the range of 5.1 to 6.9 g/l.

The SBRs cycle of operation was two hours, repeated 12 times per day (180 min/d), with a duration of aeration in SBR1 of 30 min and in SBR2 of 60 min per every two hours. The recirculation of water from SBR1 in the denitrifying reactor was carried out through a peristaltic pump.

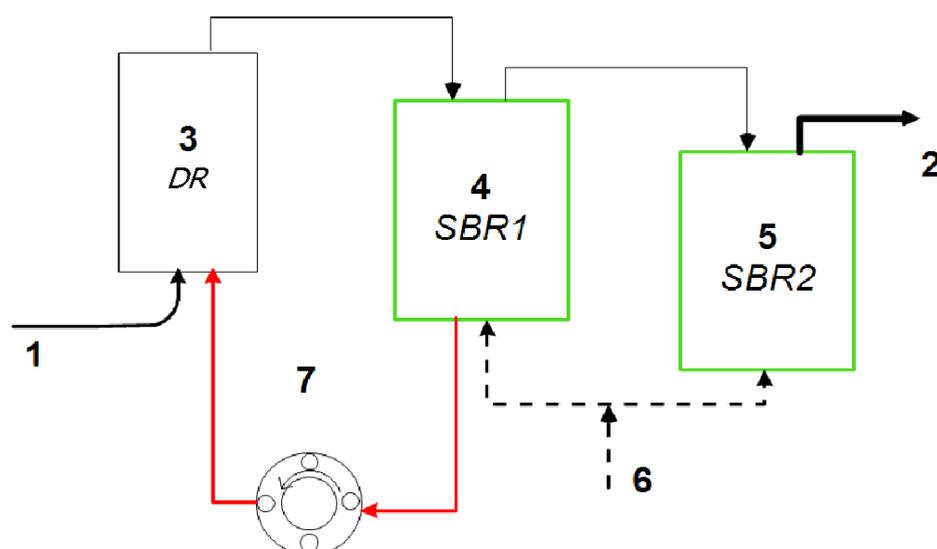


Figure 1. Laboratory installation with preliminary denitrification and two consecutive SBRs for wastewater treatment.

1 – feeding solution; 2 – effluent; 3 – denitrifying reactor DR; 4 – SBR1,
5 – SBR2 ; 6 - aeration; 7 – recirculating pump

The leachate used in these studies was characterized by the following chemical composition (in mg/l): NH_4^+ - 740, NO_3^- - 3.5, $\text{NO}_2^- < 0.003$, Cl^- - 851, PO_4^{3-} - 3.2, SO_4^{2-} - 105, BOD_5 - 185 and COD - 766. The leachate was added in concentrations respectively 2, 3 and 4% to wastewater from the production of bioethanol, the chemical composition of which was respectively (in mg/l): COD - 1088, organic acids (in the form of CH_3COOH) - 262, NH_4^+ - 23.1, NO_3^- - 0.59, $\text{NO}_2^- < 0.003$, PO_4^{3-} - 2.44.

Two series of studies were conducted, as in the first leachate in the above concentrations was added to wastewater diluted with distilled water (50:50). The second part of the experiment was performed again with 2, 3 and 4% of leachate, but without dilution of the wastewater from bioethanol production.

The pH of the influents and in SBR1 was adjusted in the range of 7.6 - 7.8 by addition a solution of 10% NaOH.

3. ANALYTICAL METHODS

The sampling points in the laboratory installation were located in each of the three facilities. After reaching a balance in the system and when changing the treated solution, there were measured the parameters pH, nitrates, nitrites, ammonium, phosphates and permanganate oxidation.

The pH was measured using pH electrode (VWR) and pH meter HANNA HI 9021. The ammonium concentration was measured by the Nessler method. The nitrate concentration was measured by sodium salicylate method. The nitrite concentration was determined by N(1-naphthyl) ethylenediamine dihydrochloride method. The phosphate concentration was determined by the molybdenum-blue ascorbic acid method. The concentration of organic compounds was determined by measuring its oxidation (by KMnO_4).

4. RESULTS AND DISCUSSION

In the present study for investigation the influence of the quantity of leachate and the ratio COD to $\text{NH}_4^+\text{-N}$ on the degree of

removal of nitrogen compounds from water, the used leachate is added in increasing percentages (respectively 2, 3 and 4%) initially to waste water from the production of bioethanol 50% diluted with distilled water. The aim of the stepwise increase in the concentration of leachate was the adaptation of microorganisms in the formed on the zeolite in the reactor for denitrification and in the sludge in both SBRs to the peculiarities of its chemical composition.

The values of the measured parameters of water in the sampling points, obtained from the two series of experiments (input solution diluted to 50% or 100% of WW and leachate of 2, 3 and 4%), are presented in Table 1.

From the data presented in Table 1 is obvious that when adding of leachate to diluted to 50% wastewater, the values of permanganate oxidation decreased from 177.7 - 174.1 mg/l to 20.7 - 5.1 mg/l at the three added quantities of leachate (84.5 - 97%).

In this series of experiments, the concentration of ammonium nitrogen in the effluents of the installation increased with the increasing of amount of added leachate (0.002 mg/l at 2%, 1.43 mg/l at 3% and 1.47 mg/l at 4% - Figure 1). The concentration of nitrates in the effluent in the treatment of 2, 3 and 4% of leachate was in the range 1.60 - 2.38 mg/l, with the lowest values were measured in the variant with 2% of leachate. The denitrification rate was 0.17, 0.27 and 0.26 mg/l.h respectively for 2, 3 and 4% of leachate in the incoming waters. From these results it can be judged that the process of denitrification is not limited.

From these experiments it was found that the highest degree of nitrogen removal (98.58%) was reached at 2% of leachate in the incoming waters, at 3% - the removal of nitrogen is 95.59%, and at 4% - 92.67%.

The input phosphate concentration was in the range of 9.19 to 12.01, and in the effluents - from 6.04 to 9.11 mg/l at all three tested concentrations of leachate. From these results it can be judged that the removal of phosphates was not achieved.

Table 1. Values of the measured parameters of waters in the sampling points at variants with 2, 3 and 4% of leachate in diluted to 50% and non-diluted wastewater.

Composition incoming water	Sampling Points	pH	PO mgO ₂ /l	NO ₃ ⁻ -N, mg/l	NH ₄ ⁺ -N, mg/l	NO ₂ ⁻ -N, mg/l	PO ₄ ³⁻ , mg/l
2% leachate in 50% wastewater	Influent	7.64±0.07	174.72	0.001	32.6	< 0.001	9.19
	DR	7.68±0.1	31.92	0.071	15.01	0.17	9.79
	SBR1	7.60±0.09	13.16	1.25	1.69	0.94	8.11
	SBR2	7.67±0.1	20.72	1.602	0.002	< 0.001	6.04
3% leachate in 50% wastewater	Influent	7.7±0.07	177.66	< 0.001	49.22	< 0.001	10.39
	DR	7.65±0.08	11.63	< 0.001	15.7	0.04	10.27
	SBR1	7.71±0.03	7.27	2.29	3.28	0.167	10.14
	SBR2	7.66±0.08	5.11	2.43	1.43	0.04	9.07
4% leachate in 50% wastewater	Influent	7.65±0.08	176.14	< 0.001	65.46	< 0.001	12.01
	DR	7.65±0.09	16.16	0.03	14.85	0.79	11.67
	SBR1	7.75±0.05	10.77	3,74	3.51	0.36	9.69
	SBR2	7.64±0.05	15.07	2.38	1.74	0.07	8.57
2% leachate in wastewater 100%	Influent	7.68±0.02	265.44	0.01	34.26	0.06	9.11
	Influent	7.66±0.1	22.68	0.11	7.37	0.29	10.02
	SBR1	7.62±0.07	12.88	2.46	0.8	0.5	9.67
	SBR2	7.6±0.1	24.08	1.31	0.012	0.38	9.08
3% leachate in wastewater 100%	Influent	7.71±0.08	240.78	0.003	49.45	< 0.001	10.9
	DR	7.67±0.05	21.82	0.077	14.08	< 0.001	10.03
	SBR1	7.73±0.02	13.47	1.98	1.75	0.039	9.12
	SBR2	7.68±0.05	13.2	2.03	1.19	< 0.001	8.52
4% leachate in wastewater 100%	Influent	7.67±0.1	256.94	0.039	68.42	< 0.001	12.54
	DR	7.63±0.08	34.74	< 0.001	20.95	< 0.001	10.83
	SBR1	7.77±0.05	18.85	2.61	5.29	0.06	9.54
	SBR2	7.67±0.08	15.89	2.08	1.66	0.099	9.5

To establish the influence of the ratio COD:NH₄⁺-N on the removal of nitrogen compounds from water was carried out a second series of experiments at which 2, 3 and 4% of leachate was added to undiluted wastewater from the production of bioethanol.

The data presented in Table 1 shows that the values of permanganate oxidation in the input solution (from 256.9 to 240,8 mg/l) decreased to ranges of 13.2 – 24.1 mg/l for the

three added quantities of leachate (89.9-94.8%).

The outflows from the laboratory installation in the treatment of 2, 3 and 4% of leachate contained ammonium nitrogen in concentrations of 0.012, 1.19 and 1.66 mg/l (Figure 2). From the results it can be said that increasing the amount of leachate at the inlet leads to an increase of ammonium nitrogen at the outlet.

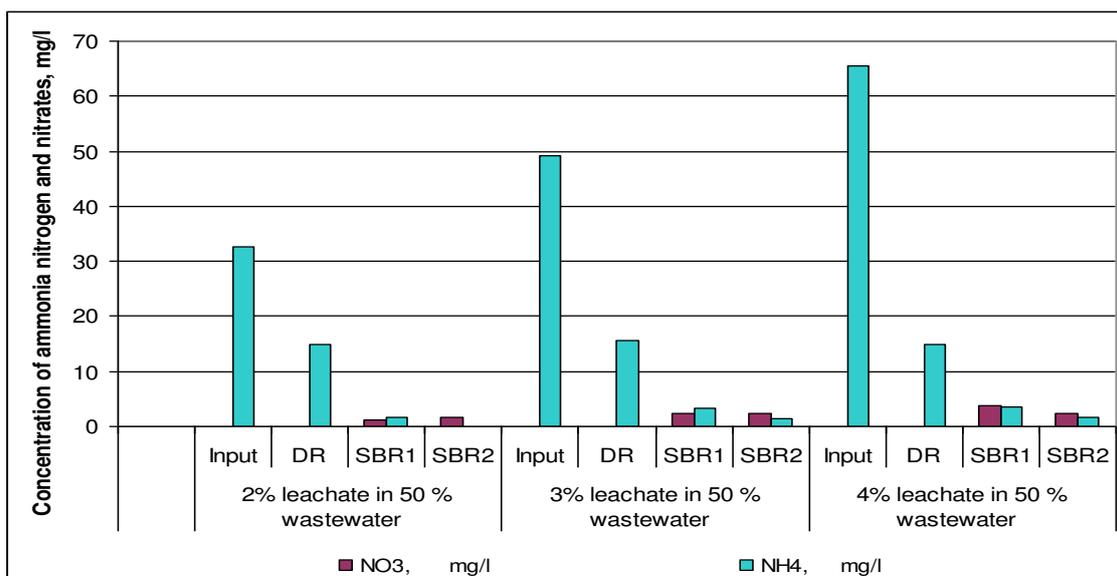


Figure 2. Concentrations of ammonium nitrogen and nitrates for 2, 3 and 4% of leachate at diluted to 50% influents (wastewater from the production of bioethanol).

For the concentration of nitrates in effluent and in the second series of experiments was observed that same dependence as those for the concentration of ammonium nitrogen (Figure 3). The increase of the leachate quantities at the inlet raised the concentration of nitrates in effluent (1.33 mg/l for 2%, 2.03 mg/l for 3% and 2.08 mg/l for 4% of leachate). The concentration of nitrates

at the outlet of the anaerobic reactor was relatively low. It was found that the rate of denitrification process increased with the increase of leachate content. For 2% it was 0.14 mg/l.h, and for 3 and 4% - 0.23 and 2.23 mg/l.h. The estimated rate of nitrogen removal for 2% of leachate in the incoming solution is 99.89%, and for 3% and 4% respectively 96.39 and 96.69%.

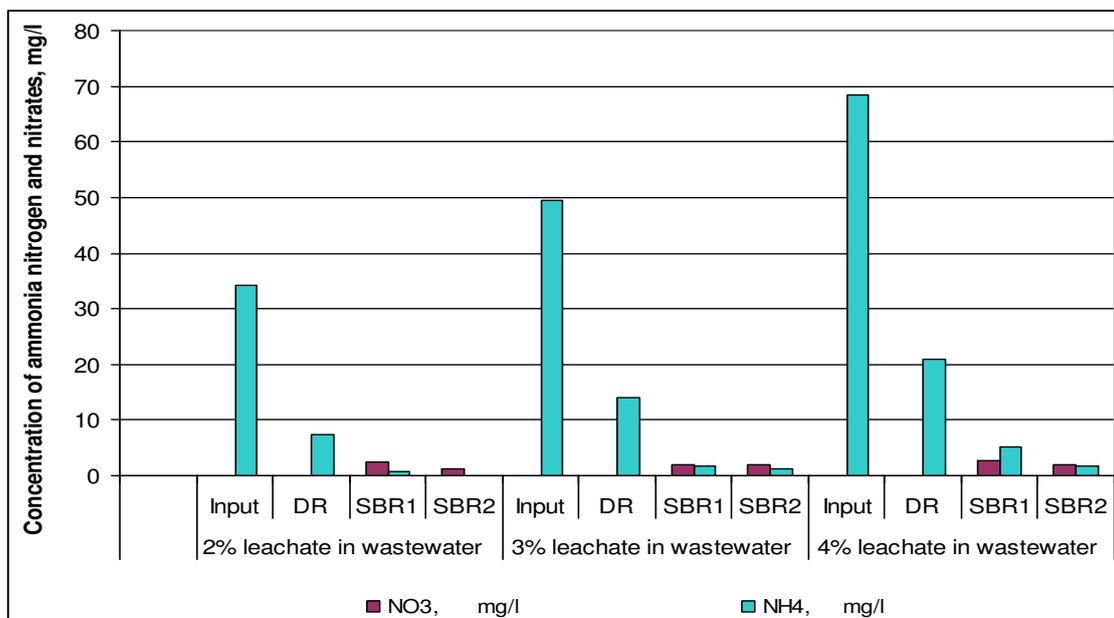


Figure 3. Concentrations of ammonium nitrogen and nitrates for 2, 3 and 4% of leachate at undiluted influent (wastewater from the production of bioethanol)

The results in Table 1 show that the concentration of phosphates in the influent were 9.11 and 10.9 and 12.54 mg/l for 2, 3 and 4% of leachate. In the effluent their concentration was reduced slightly and was in range (8,52 - 9,5 mg/l).

5. CONCLUSION

From the obtained results the following conclusions could be drawn, determining the relevance of the investigated biotechnological methods for leachate treatment.

In both series of experiments it was carried out an efficient reduction of COD. The values of permanganate oxidation of effluents from the laboratory when using diluted and undiluted wastewater from the production of bioethanol were respectively 20.72 and 24.44 mg/l. These rates showed that the concentration of activated sludge in the two SBRs and the operational mode allowed the achievement of an effective oxidation of large quantities of easily degradable organic compounds.

The increased ratio COD to N did not affect the course of nitrification. In both experiments with initial concentrations of ammonium nitrogen in the range 32.6 – 34.3 mg/l the concentration of ammonium nitrogen in the effluent decreased to 0.002 – 0.012 mg/l.

The same conclusion can be made and for the process of denitrification. For the both studied ratios of COD:N, the concentrations of nitrates at the outlet were in the range 1.602-1.31 mg/l, as the obtained values are almost equal and within the error of the used methodology.

The influent solution for the three percentages of leachate contained no nitrites. Higher concentrations of nitrites in some of the experiments were measured after the denitrifying reactor or after first SBR reactor. At the outlet the concentration of nitrites was 0 – 0.07 mg/l. This result shows that at the selected operational mode in the second SBR also occurs optimally the process of anammox at which nitrites and ammonium ions are

converted directly into final products molecular nitrogen and water.

With the increase of the leachate quantities in the influent, the rate of nitrogen removal was reduced.

6. REFERENCES

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