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Original paper

Evolution of chemical and biological characteristics of Glina Landfill between August 2017 and June 2019. Biochimical evolution of Glina Landfill

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Abstract

The present research aims to characterize from biochemical point of view leachate samples from the urban waste storage facility located in Glina, Ilfov County. The measurements were focused on the concentrations in nitrates, nitrites, ammonium, phosphorus and other characteristics like pH, biochemical oxygen consumption and chemical oxygen consumption on samples taken at the entrance and the exit of the aeration lagoon.

The methods used in the laboratory analysis for the leachate samples were in accordance with the methodology applied for wastewater treatment.

The high CBO_5 yield, between 62% and 78%, confirmed the ability of micro-organisms present in leachate to decompose pollutants, to absorb the decomposed material and finally to eliminate them from the leachate in a high rate.

Concerning the chemical consumption of oxygen, there was also high efficiency, i.e. values between 54% and 71%, which evolved simultaneously with the CBO_5 variation.

Keywords Waste facility management, leachate, pollutants removal, microbiological purification.

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Introduction

The *Glina landfill*, Ilfov County, was established in the 1977 years on the former Ochiul Boului pond, Popeşti-Leordeni commune. It was administered by the City of Bucharest until 1998. After the 1977 earthquake, the Ochiul Boului pond became the dumpsite of Bucharest, the largest landfill of the Capital.

The Glina landfill has a total area of 119.2 ha and a total storage volume of 17.5 mil m^3. The area occupied by the non-compliant warehouse is 37 ha and the area available for extension is 75.4 ha. The area managed between 2003 and 2005 was 2.54 ha, and between 2005 and 2018 it was 16 ha.

At present ECOREC Ltd, a company established in 2001, manages this landfill (the largest in Romania).

Aerobic microorganisms are commonly used for purifying most of the predominantly organic wastewater – carbon, nitrogen or phosphorus compounds – and for stabilizing certain sludge categories.

Leachate composition and properties depend on the environmental conditions (temperature, precipitations), landfill age and maturity, waste characteristics and composition, landfill operating technology (GALIT I. & al [5]).

During the ageing process, deposits pass through different stages of organic waste degradation, starting from aerobic to anaerobic decomposition. Therefore, the leachate properties such as Chemical Oxygen Consumption (CCO), Biochemical Oxygen Consumption (CBO), Ammonium (NH3) or pH change according to the internal and environmental conditions of waste decomposition.

Biodegradability can be estimated by the CBO/CCO ratio, environmental conditions, temperature, pH and the absence of inhibitors that have major effects on the bacteria. The following chemical bonds can be biologically decomposed: carbon bonds, nitrogen bonds (ammonia nitrogen NH4-N, among others), AOX relations (BODEA T. & al [1]).

The biological processes for leachate treatment are based on the biochemical reactions depending on the metabolism of some bacteria, fungi and other lower microorganisms, especially protozoa which are found in wastewater with organic load and form biomass (BOROŞ M.N. & al [2]).

In the present research we have investigated the ability of anaerobic microorganisms to remove pollutants in the stationary state of the leachate (in the aeration lagoon) during monitoring 15 days, and we started from the presumption that the excess of negatively charged hydroxyl may compete with the inorganic constituents available in the leachate for the decomposition of the pollutants.

Generally, the main parameters of the biologically treated leachate, resulting from the municipal waste dumps treated in aeration lagoons, are the following: CCO, CBO_5, the CBO_5/CCO ratio, pH, suspension matters, ammonia, nitrates and nitrites, phosphorus and heavy metals content.

The aeration lagoons are basins that are usually dug in the ground and operate without recirculating solids, which is a major difference from urban wastewater treatment sludge systems.

The purpose of introducing the aeration lagoon is to reduce the leachate pollutant load by microbiological decomposition, increase the efficiency of the treatment system by introducing a biological treatment step before reverse osmosis and at the same time the lagoon can perform the role of buffering basin for maximum leachate flow rates periods.

The performance of the lagoons can be characterized firstly by reducing the concentration of pollutants during the leachate stationing period in the lagoon. Thus, based on the ***efficiency percentage*** or ***efficiency index (I_e)***, it can be defined as a ratio between the difference of concentrations at the entrance to (C_i) and at the exit from (C_e) the lagoon and the concentration at the leachate entrance (C_i) to the aeration lagoon:

$$\mathbf{I_e = (C_i - C_e) / C_i}$$

Efficiency shows the capacity of the lagoon to remove the various types of pollutants and consequently what other methods are necessary for the total leachate treatment under safe conditions for the system and the environment.

Material and Methods

The material used in the research was the leachate from cell 2 of the Glina sanitary facility. The samples were collected quarterly between August 2017 – June 2019 from the entry and exit points, and from the aeration lagoon every 2 months, respectively.

The samples were taken in sterile containers and transported to the laboratory on the same day for biochemical analysis and microbial loading quantification. When the analysis could not be performed on the same day, the samples were kept maximum 24 hours under refrigeration conditions (4ºC).

Biochemical analysis methods

In the case of the materials in suspension (mg / l), the determination method was based on filtering and drying; the principle of the method consisted in separating the suspended particles by filtration, drying in the oven at 105°C and weighing.

Regarding the nitrate (NO_3^-, mg / l), the determination method was spectrometric with sulfosalicylic acid and the method's principle was based on measuring the intensity of the yellow coloring complex, sodium nitro salicylate consisting in salicylic acid and nitrates. Maximum absorption was at λ = 415 nm. Nitrate ions interacted with sodium salicylate in the sulfuric acid environment, forming 3-nitrosalicylic and 5-nitrosalicylic acids, whose salts had a yellow color.

Regarding the nitrogen (NO_2^-, mg / l), the determination method was by molecular absorption spectrometry and the method's principle was the following: determinations were usually made against a reference sample, by comparison, contained in a cell of the same size as the one in which the sample for analyze was found. The reference sample usually contained the solvent and constituents of the sample, except for the species whose absorbance we measured. With such a reference solution in the cell, the transmitted radiation intensity represented the intensity of the incident radiation minus the one lost through diffusion, reflection and any absorption due to other constituents.

Regarding ammonia (NH_4^+ mg / l), the determination method was the manual spectrometric method and the method's principle consisted in the reaction of ammonium ions, in basic medium, with potassium tetraiodomercurate (K2 [HgI4]) forming a complex (oximercurammonium iodide) of yellow-brown colour.

Regarding total phosphorus (mg / l), the determination method was spectrophotometric with ammonium molybdate and the method's principle was the following: the phosphate anion reacted with ammonium molybdate in acidic medium, and ammonium phosphomolybdate was formed, the resulting ammonium phosphomolybdate formed under the action to a reducer, a blue complex known as molybdenum blue. The color intensity of the complex was proportional to the phosphate concentration.

For the CBO5 (mg / l), was employed the method by dilution and contribution of allylthiourea; the method's principle was the following: the determination of the oxygen consumed for five days by microorganisms in water by the difference between the amount of oxygen found in the water sample immediately and five days after sampling.

Regarding CCOCr (mg / l), the determination method was based on the potassium dichromate and the method's principle consisted in the following: the CCO-Cr value was determined directly against a wavelength of a control sample prepared and treated as the samples to be analyzed but containing only distillate water and oxidizing mixture in the same proportions to a laboratory spectrophotometer.

Results and Discussions

Regarding biochemical oxygen consumption, Figure 1 shows the results of the laboratory analysis performed on the leachate samples taken at the entrance and the exit of the aeration lagoon. At the lagoon entrance, the Biochemical Oxygen Consumption had values between 20.0845 mgO2 / l (February 2019) to 15.844 mgO2 / l (October 2017).

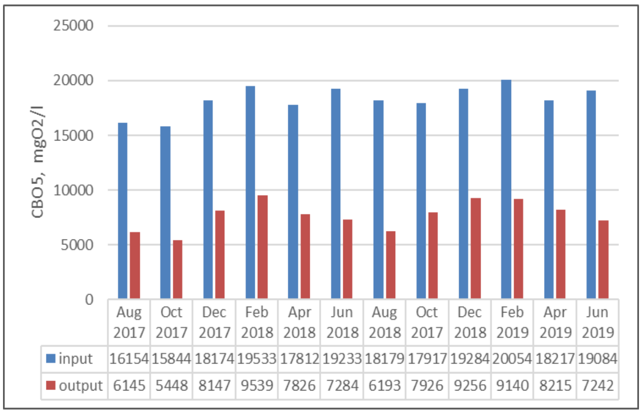

Figure 1. Variation of Biochemical Oxygen Consumption CBO of leachate from Glina ecological landfill.

At the exit point the concentration of leachate in CBO5 recorded values within the range of 5448 mgO2/l (October 2017) the minimum and 9534 mgO2/l the maximum value recorded in February 2018, a value corresponding to the minimum retention of 9.550 mgO2/l and a 52% minimum retention yield, respectively.

During storage by the time of storage cell closure, waste went through degradation stages of the organic components specific to the aerobic phase. After storage cell closure (final coverage) the dominant phenomena were specific to anaerobic decomposition. Therefore, properties such as Chemical Oxygen Consumption (CCO), Biochemical Oxygen Consumption (CBO), Total Oxygen Consumption (COT), pH and other characteristics changed depending on the storage stage of the fermentable waste and on the microbial component of the biocenosis of the leachate, respectively (STOICA R.M. & al [15]).

From the analysis presented graphically in Figure 2, we observe the time variation of the CCO determined by the irregular rhythm of biodegradation; a slow rate of biodegradation could result in increased CCO over time. The figure shows that CCO decreased during the same period with the decrease of the content in nitrates and with the return of the pH to values of 7.1-7.4, recording a much more consistent decrease at the exit point of the lagoon, with the yield reaching a maximum of 69%, so the commissioning of the aeration lagoon had a high efficiency for the CCO.

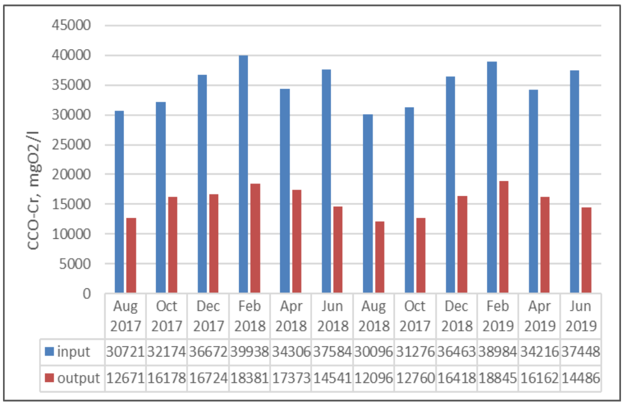

Figure 2. Variation of CCO Chemical Oxygen Consumption of leachate from the Glina ecological landfill.

Figure 2 shows that CCO had a continuous increase in August 2017 – February 2018, i.e. from 30,721 mgO2/l to 39,938 mgO2/l at the lagoon entrance. After February 2017 the CCO values varied between 29,784 and 37,904 mgO2/l.

The analysis of the samples taken at the lagoon exit recorded the following values: the maximum concentration was 18,845 mgO2/l in February 2019 and the minimum was obtained in August, i.e. 12,096 mgO2/l, and the retention was between 18,153 and 23,734 mgO2/l. The values resulted in a yield between 59 and 68% on CCO removal.

HALIM A.A. & al (2009) [6] studied the feasibility of lagoon for treating phenolic compounds, as well as organic matter; a reduction of 55-64% of CCO and 80-88% of phenol was achieved.

When leachate from landfills had a CBO / CCO ratio greater than 0.25, it might have been subjected to normal biological treatment in bioreactors or in lagoons, and ponds with aerobic or anaerobic bioepurification.

DUMITRU M.A. (2019) [3] stated that biological processes were proved to be very effective in removing organic matter and nitrogen from immature leachate when the CBO / CCO ratio was higher than 0.5.

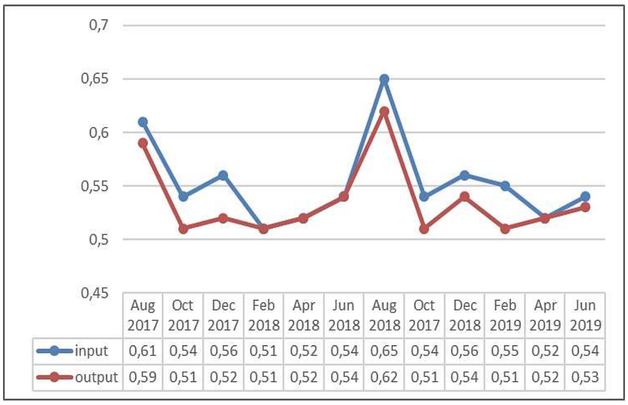

Figure 3. Variation of CBO / CCO ratio in leachate from *Glina landfill* (August 2017 – June 2019).

From the analysis presented in Figure 3, it appears that the leachate from the *Glina landfill* with a CBO / CCO ratio greater than 0.5 was very well suited for biological treatment even in the colder months (February and December) when the climatic conditions reduced the performance of lagoon purification.

Regarding this report, (GARBO F. & al [4]) showed that the leachate was poorly biodegraded at a CBO / COD ratio below 0:20 and also showed that the leachate was stable and difficult to further degrade biologically.

The content in *nitrates* and *nitrites* highlighted by laboratory measurements on samples taken at the entrance in the aeration lagoon ranged between 84.9 mg/l and 94.8 mg/l in the coldest periods of the year (February and December respectively) and between 31,9 mg/l and 50.7 mg/l (Figure 4) at the exit of the aeration lagoon.

The highest retention was recorded in August 2018, i.e. 62.9 mg / l, which implied an intense activity of the microorganisms present in the lagoon leachate during the summer period.

At the exit from the aeration lagoon, the lowest nitrate content corresponded to August 2019 (25.4 mg/l) and October 2017 (29.5 mg/l), minimums for which we considered the following explanations:

- pH had a value of 7.4, which was very close to neutral, i.e. optimal for the activity and life of microorganisms;

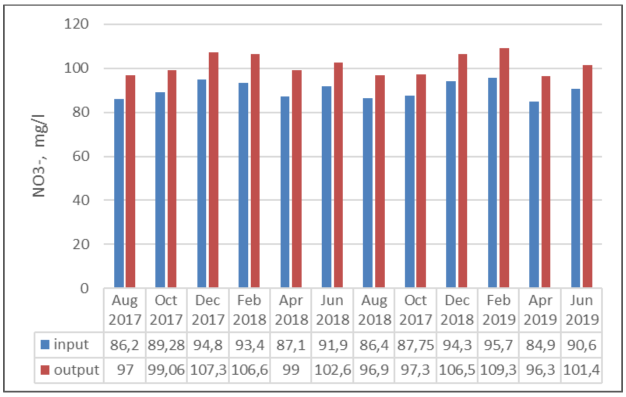

Figure 4. Nitrate and nitrite content of leachate from *Glina landfill* (August 2017 – June 2019).

- the average temperature of the environment was 21°C, which also corresponded to the optimal development of the bacteria;

- spraying the landfill to maintain optimum humidity (50-65%) for the processes of decomposition of fermentable waste from the warehouse.

The same effect was also found regarding the content in ***nitrites***, where the minimum values recorded at the entrance in the aeration lagoon were between 9.5 and 9.8 mg/l registered in October 2017 and 2018 respectively and at the exit from the lagoon the minimum values were 4.8 mg/l (October 2017) and 5.4 mg/l (April 2019).

Mendoza F. (2017) [10] used anaerobic and aerobic lagoons located near landfills for biological treatment of the landfill leachate. Reductions of over 70% of N, P and Fe were obtained in the system, which resulted in the dilution of the leachate.

The presence of a significant amount of ***ammonium*** in leachate – between 602 mg/l (October 2017) and 857 mg/l (February 2018) indicated a degradation of soluble nitrogen, largely due to the decomposition of waste in the landfill. Consequently, NH3-N concentration rised with the increasing age of the landfill, mainly due to hydrolysis and fermentation of nitrogenous substances, which were fractions of biodegradable substrate in the landfill.

The higher concentration of NH3-N enhanced algal development and promoted eutrophication due to the decreased dissolved oxygen content. Moreover, nitrification also led to an increase in algae, thus decreasing the performance of the biological treatment system. Consequently, the accelerated eutrophication promoted by the depletion of dissolved oxygen resulted in the increased toxicity of the living organisms present in the leachate (POZNYAK T. & al [12]).

The biochemical activity of the microorganisms that degraded the organic matter from the leachate led to the decrease in the ammonium content between 41% and 63% in August 2018 – June 2019 (three years after the commissioning of the aeration lagoon).

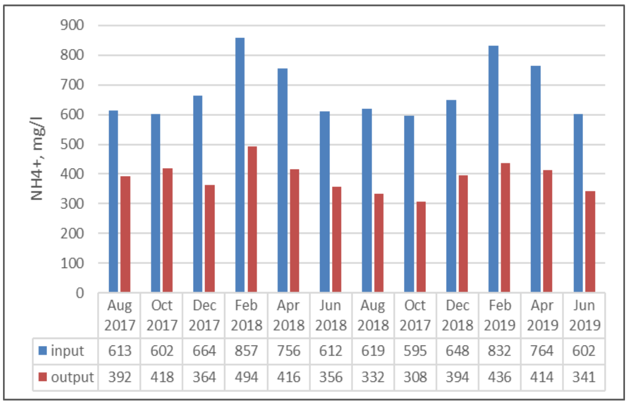

Figure 5. Ammonium content of leachate from *Glina landfill* (August 2017 – June 2019).

In February 2019 and April 2019, an increase of ammonia at the entrance point in the aeration lagoon was noticed, due to the reduction of biomass activity caused by the decrease in the ambient temperature (Figure 5).

Regarding the values of the ammonium concentration at the exit from the aeration lagoon, the experimental determinations showed the following results:

- the maximum concentration was recorded in February 2018 when a concentration of 494 mg/l was recorded;
- the minimum concentration was found in October 2018, i.e. 308 mg/l;
- the retention of ammonia in the aeration lagoon was between 294 mg/l and 363 mg/l, which meant a lower yield of the lagoon in the retention of ammonium.

Iordache O. & al (2017), listed ammoniacal nitrogen as a major toxic substance for such living organisms as *Salmo gairdnieri* and *Oncorhynchus nerka*. This fact was established by various toxicity analysis using bioanalysis, which explains the low yield in retaining this pollutant (IORDACHE O. & al [8]).

The applications of biological processes in leachate purification are: elimination of organic substances measured in CBO, CCO, CBO / CCO ratio, nitrification, denitrification, phosphorus removal, sludge stabilization. Organic nitrogen and phosphorus are converted to ammonia and phosphate.

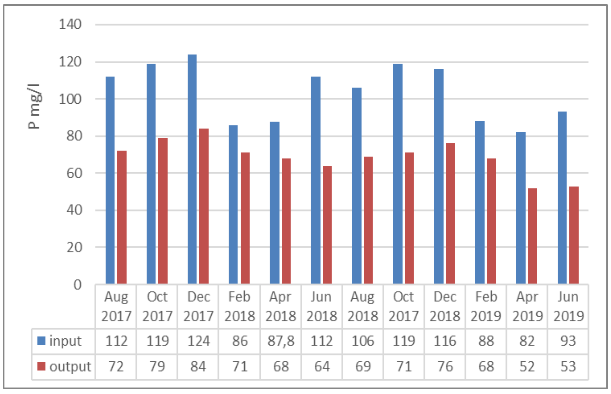

Figure 6. Variation in total phosphorus content of leachate from *Glina landfill* (August 2017 – June 2019).

Figure 6 shows an increase in the total phosphorus content from 82 mg/l to a maximum of 119 mg/l in October 2018, i.e. 42 months after the commissioning of the aeration lagoon, the values measured at the entry point in the lagoon. At the exit point from the lagoon concentrations between 52 mg/l minimum were recorded in April 2019, and 84 mg/l maximum were recorded in December 2017.

The results obtained on the leachate from the Glina landfill were in agreement with the data presented by (KOC-JURCZYK J. & al [9]) in their research.

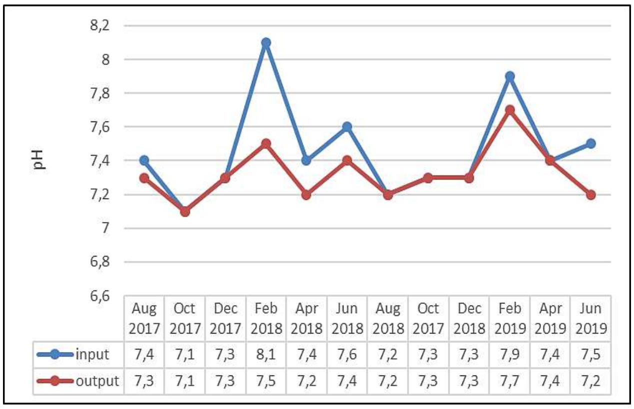

Figure 7. pH variation in leachate from *Glina landfill* (August 2017 – June 2019).

As a primary effect observed in the experimental determinations, the biochemical activity of the micro-organisms present in the aeration lagoon resulted in the *change of the pH* which decreased by 1.9 units from 8.1 in February 2018 to 7.1 in April 2019 (Figure 6).

The pH decrease was due to the activity of the organisms in the leachate which was more intense when the leachate pH was neutral.

The data obtained in these investigations were in agreement with the activity reported by (ISMAIL T. & al [7]), who observed that the yield of the biodegradation plant increased by the optimal dosing of pH and by (ÖZGÜR A. & al [11]). Significant reductions were recorded between April and August 2016 with regard to the nitrogen content compounds (nitrates, nitrates and ammonia).

Conclusions

As seen from the experimental results, the ***Biochemical Oxygen Consumption*** decreased by 26% throughout this time period. The decrease in the ***Chemical Oxygen Consumption*** was much more consistent, as it reached a maximum of 38% after the aeration lagoon became operational.

Also, the ***nitrate*** content established by laboratory measurements showed an increasing efficiency in the biological treatment after the exploitation of the aeration lagoon particularly during the hot periods, which implied an intense activity of the microorganisms present in the leachate during that period.

The measurements made on the nitrite concentration showed an intense activity of the microorganisms present in the lagoon leachate during the summer period, and a decrease in their activity in the winter months.

The presence of a significant amount of ***ammonium*** (1.322 mg/l) in the leachate indicated the degradation of the soluble nitrogen due to waste decomposition.

The analysis resulted in an increase in the amount of ***total phosphorus***, as the results from the two experimental stages (before and after the insertion of the aeration lagoon) were consistent.

As a first effect observed in the experimental determinations, the biochemical activity of the microorganisms present in the aeration lagoon recorded a reduced pH variation that was kept close to the neutral value from 7.8 (February 2017) to 7.1 (June 2019). The values determined a high intensity of biological degradation of the leachate.

The large amount of polluted elements present in the leachate often required the combination of several treatment forms, in order to obtain a satisfactory result because the quantity of leachate and its degree of impurity were dependent on: the type of deposited waste, the age of leachate, the meteorological characteristics of the location area, the quality insulation from the landfill surface.

Previous researches conducted by (PANȚER Z. & al [13] [14]) on the same facility found that there were no major differences in pollutant concentration and retention efficiency of the aeration lagoon. After the first two years of storage operation, the pollutants showed no significant variations and the pH value showed a slightly alkaline medium with decreasing tendency towards neutral values.

The biological treatment of the leachate is a simple and especially viable method in terms of cost effectiveness. The advantages of this method mainly consist in the efficiency of treating leachate with high concentrations of organic substances, nitrogen or CBO, in the case of a high CBO/CCO ratio, usually encountered in the young leachate.

The effluent discharged from the aeration lagoon is inserted into the treatment plant by reverse osmosis with a very low pollutant load, which facilitates the operation of the station at the highest parameters in parallel with the reduction of the treatment costs.

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