CO-TREATMENT OF MUNICIPAL LANDFILL LEACHATE WITH DAIRY WASTEWATER IN MEMBRANE BIOREACTOR

Abstract: The aim of the study was to determine the effectiveness of leachates from municipal landfill co-treatment with the dairy wastewater in an aerobic membrane bioreactor. It was working in MSBR (sequential membrane bioreactor) systems twice daily and was equipped with the immersed membrane module installed inside what enabled its back-washing performance. The system was working. The concentration of activated sludge in the membrane bioreactor was equal to 4.0 g/dm$^3$. However, the sludge load was at the level of 0.06 g COD/(g d.m. · d). The oxygen concentration was at the level of 3.0 g O$_2$/m$^3$. The share of leachate was varied in a range of 5 to 15 % vol. The evaluation of the effectiveness of the treatment process was based on the change of parameters characterizing the crude sewage and treated sewage. All analysis was carried out according to standards. Following parameters were determined: COD, BOD$_5$, TOC and concentrations of phosphate phosphorus, total nitrogen and ammonium nitrogen. Chemical analysis is often not enough to define the degree of wastewater treatment. It was used toxicological research to determine the effect on the environment. Toxicity of wastewaters was measured using biotests with Vibrio fischeri and Daphnia magna. The results revealed that the volume of leachate in the treated mixture should not exceed 10 % vol. The following conclusion can be drawn from the present research - co-treated wastewater was not toxic. Landfill co-treatment with the dairy wastewater impacts on the effectiveness of biological wastewater treatment. Leachate includes substances which have low susceptibility to biodegradation; on the other hand, dairy wastewaters provide a lot of organic compounds, which can help to treat them.

Keywords: dairy wastewater, landfill leachate, membrane bioreactor, activated sludge

Introduction

The aim of the presented study was to determine the impact of dairy wastewater on the effectiveness of the municipal landfill leachates and co-treatment in the aerobic membrane bioreactor. In Poland, the most common and cheapest way to dispose of waste of is to store it on the surface. The municipal waste landfill is a source of leachate that is harmful to the environment. Their treatment is costly and moreover is based on complex technology. The characteristic factors are especially: a very high concentration of toxic ammoniacal nitrogen lasting for a long time in a landfill and high values of chemical oxygen demand (COD) and biochemical oxygen demand (BOD$_5$) [1-5]. It was recognized more than 200 organic contaminants, consisting of cyclic and aromatic hydrocarbons, bicyclic compounds, benzene derivatives, linear and cyclic ethers, alcohols, acids and esters, ketones, phenols, furans, organic nitrogen compound, silica, phosphorus, sulfur, and hazardous toxic as chlorobenzene, dichlorobenzene, xylene, toluene, styrene, naphthalene,
absorbable organic halogens (AOX) [2, 5]. According to the Regulation of the Minister of the Environment of 18 November 2014 [6] on types of detailed requirements about location, construction, exploitation and closure, which should be associated with appropriate types of landfill leachate the wastewater should be recognized, cumulated and subjected to treatment to the extent enabling their acceptance at the sewage treatment plant or discharge into waters or the ground [7].

The results of previous research conducted by the author of this article including on the co-treatment of leachate with municipal wastewater in the aerobic membrane bioreactor, which worked in the SBR system, the best proportion share of leachate in the wastewater mixture was at the level of 5 % vol. [8, 9]. Because dairy wastewater is biodegradable landfill product which requires additional disposal, can be successfully co-treated with municipal landfill leachate [10-16]. The presence of dairy wastewater can provide a high degree of biodegradability of leachate in the feed. This solution should ensure the increase of the proportion of leachate volume in the feed (with dairy wastewater) and improve the efficiency of treatment.

A number of reports in the literature associated with the possibility of municipal leachate treatment, published in major scientific journals has increased 15 times for the last 15 years. A lot of research considers the problem of leachate treatment by using physicochemical and biological methods, but only a few of them can show the effectiveness of treatment (to reduce wastewater toxicity [5, 17-21]. Because of the potential danger of toxic components of industrial wastewater effect on the environment, the implementation of toxicity testing was compulsory. Matejczyk et al. [5] evaluated the risk posed the leachates from 22 municipal solid waste landfill sites in Southern Poland using chemical and ecotoxicological assays. They found that the leachate toxicity could not be explained by the presence of trace levels of the contaminants detected by chemical analysis. The study thus concluded that the environmental risk posed by landfills should be evaluated by integrated chemical, ecotoxicological and microbiological monitoring program [5]. Bioindication method allows the assessment of the environment or properties of environmental factor, in particular, using biological indicators (plants or animals). Because, it can be supposed that between the leachate components and in this case dairy wastewater can be observed the interaction processes such as addition, synergism/potentiation or antagonism, it was decided to carry out toxicity tests [22, 23].

**Materials and methods**

The substrate of this study was leachate generated at the municipal landfill in Tychy-Urbanowice and dairy wastewater from District Dairy Cooperative in Bierun. It was found that the values of contaminants indicators which characterized particular samples of dairy wastewater differed from each other, what was caused by the technological process carried on during their sampling. Part of the samples originated directly from the production while the rest was collected from tanks and milk processing devices. It was stored at 4 °C. A sampling of raw and purified wastewater was carried out according to the Regulation of the Minister of the Environment [7]. Activated sludge from Municipal Wastewater Treatment Plant in Gliwice was used. The values of selected physicochemical parameters of landfill leachate and dairy wastewater were shown in Table 1.
Co-treatment of municipal landfill leachate with dairy wastewater in membrane bioreactor

Table 1

| Parameter | Unit            | Leachate | Dairy wastewater |
|-----------|-----------------|----------|------------------|
| COD       | [mg O₂/dm³]    | 3040-4300| 3700-14000       |
| BOD₅      | [mg O₂/dm³]    | 250-380  | 400-2700         |
| TOC       | [mg C/dm³]     | 260-600  | 200-4000         |
| N₅ₒ      | [mg N/dm³]     | 350-530  | 60-270           |
| N-NH₄⁺    | [mg N-NH₄⁺/dm³]| 500-1000 | 3.0-30           |
| N-NO₃⁻    | [mg N-NO₃⁻/dm³]| 2.5-8.0  | 19-87            |
| P-PO₄³⁻   | [mg P-PO₄³⁻/dm³]| 14-22   | 29-125           |

Biological process co-treatment of leachate dairy wastewater was carried out in a membrane bioreactor of 15 dm³ vol. installed inside the membrane module (Fig. 1). The capillary membranes were made of polyethersulfone (cut-off 80 kDa). The construction of the module ensured back-washing performance. Inserted the sensors into the reaction chamber to measure wastewater level, oxygen concentration and temperature.

![Fig. 1. The scheme of the experimental set-up: 1 - raw wastewater tank, 2 - aerobic-anaerobic chamber, 3 - capillary membrane module, 4 - purified wastewater tank, 5 - manometer](image)

At first, the cycle of research was started from adapting activated sludge microorganisms to the aerobic biodegradation of pollutants appearing in dairy wastewater. This process was carried out for each kind of wastewater differing to a percentage share of leachate. The concentration of the activated sludge was equal to 4.0 g/dm³, and the oxygen concentration was at the level of 3 mg/dm³. The share amount of leachate was changed over the range of 5 to 15 % vol. Moreover, the constant sludge load was roughly 0.06 g COD/(g d.m. · d). The membrane bioreactor acted as SBR in two cycles per day. The filling and stirring phases have been carrying out for 4 h, aeration - 7 h, wastewater sedimentation and clarification - 1 h. Sludge age was 25 h.

Assessment criteria (degree of wastewater treatment) was the change value of pollution indicators which was characterized by the crude sewage and the treated sewage. Defined following markings: COD, BOD₅, TOC, N₅ₒ, N-NO₃⁻, N-NH₄⁺, P-PO₄³⁻. To determine the various forms of carbon used carbon analyzers multi N/C (Analytic Jena Company). Concentrations of total nitrogen, ammonium, nitrate, phosphate phosphorus and COD were determined according to the methodology specified by Merck, and BOD₅ using the
respirometric method with a set of equipment WTW OXI Top. Total Cd, Ni, Pb, Zn, Cr and Cu concentrations in the leachate were determined by atomic absorption spectroscopy (AAS) SpectrAA 880 VARIAN.

Toxicity assessment was carried out by using F Daphtoxit MicroBioTests Inc. Magna production, according to standards OECD Guideline 202 and ISO 6341. The test was carried out by making use of the dormant eggs of the crustaceans *Daphnia magna*.

Five dilutions of dairy wastewater mixture with 10 % vol. (100, 50, 25, 12.5, 6.25 %) were prepared. Wastewater sample (1:1 dilution) had standard fodder, which was prepared according to the manufacturer's instructions. Their toxicity was assessed in four reps by indicating 5 animals to every test hole, filled wastewater with various dilutions. Incubation took place at 22 °C, and there was no light. The test reaction was stopped the movement of crustacean, which was observed 24 and 48 hours after incubation in relation to a control sample with no mixture. Another indicator organism to toxicity assessment was luminescent bacterium *Vibrio fischeri*. Toxicity was assessed based on Microtox® results.

Degree of toxicity was evaluated based on the change of emitted intensity light by bacteria. The experiment was performed in accordance with the WET (Whole Effluent Toxicity) MicrotoxOmni system. The same wastewater dilutions were prepared as for the acute toxicity determination of *Daphnia magna*. The result of the test was the *EC*₅₀ value, i.e. the concentration of the sample resulting in a reduction of luminescence by 50 %. The *EC*₅₀ value was determined by graphical interpolation of the result (percentage effect on the "concentration logarithm/mortality" scale). To determine toxicity, the *EC*₅₀ results were converted to *TU* toxicity units [24]:

\[ TU = \left( \frac{1}{EC_{50}} \right) 100 \% \]  

where *EC*₅₀ - concentration of the test sample that produces 50 % toxic effect.

Classification of wastewater toxicity is presented in Table 2.

| *TU* | Classification                          |
|------|----------------------------------------|
| < 0.4 | No acute toxicity - none of the tests showed a toxic effect |
| 0.4 < *TU* < 1 | Low acute toxicity - at least one test showed a test effect |
| 1 < *TU* < 10 | Acute toxicity - at least one test showed a test effect |
| 10 < *TU* < 100 | High acute toxicity - at least one test showed a test effect |
| *TU* > 100 | Very high acute toxicity - at least one test showed a test effect |

**Table 2**

**Results and discussion**

**Physicochemical analysis of co-treated wastewater**

As aforementioned, the values of physicochemical indicators characterizing mixtures of dairy wastewater with of leachate were different in time, but every time they were maintained at a constant level both sludge loading *COD* and the leachate percentage. The physicochemical parameters of dairy wastewater were changed, and it was connected with taking theirs after various production processes. The mean values of pollutants characterized by the leaches the mixture with the dairy wastewater were shown in Table 3. Values of pollutants in the wastewater in membrane bioreactor were shown in Table 4.
Table 3

The values of pollutants characterized by the leachates of the mixture with the dairy wastewater

| Share leachate [% vol.] | Parameter [mg/dm$^3$] | COD | BOD$_5$ | TOC | N$_{tot}$ | N-NH$_4^+$ | N-NO$_3^-$ | P-PO$_4^{3-}$ |
|-------------------------|------------------------|-----|---------|-----|---------|----------|-----------|-----------|
|                         |                        | 5   | 6545    | 2100| 1454    | 159      | 31        | 34        | 78        |
|                         |                        | 10  | 7100    | 1750| 1590    | 231      | 34        | 14        | 34.5      |
|                         |                        | 15  | 4045    | 1500| 642     | 125      | 30        | 9.1       | 42        |

Table 4

Effectiveness of the leachates co-treatment in the membrane bioreactor depending on the volumetric their share

| Share leachate [% vol.] | Parameter [mg/dm$^3$] | COD | BOD$_5$ | TOC | N$_{tot}$ | N-NH$_4^+$ | N-NO$_3^-$ | P-PO$_4^{3-}$ |
|-------------------------|------------------------|-----|---------|-----|---------|----------|-----------|-----------|
|                         |                        | 5   | 106     | 5   | 22      | 5        | 1.5       | 1.5       | 3.0       |
|                         |                        | 10  | 120     | 7   | 25      | 4.5      | 1.5       | 1.5       | 2.8       |
|                         |                        | 15  | 180     | 10  | 64      | 7.0      | 1.8       | 3.0       | 3.0       |

It was found that with the increase of the share of dairy effluent in wastewater, in the range of value 5-10 % vol. the content of organic compounds in the outflow of the membrane bioreactor increases. COD treated wastewater at 10 % vol. the share of leachate was 120 mg O$_2$/dm$^3$. Degree of removal, in this case, was equal to 98.3 %. However, for the share of leachate at the level of 15 % the concentration of chemical oxygen demand of treated wastewater was higher - 180 mg O$_2$/dm$^3$ what exceeded the normalized value (125 mg O$_2$/dm$^3$). Reduction of BOD$_5$ concentration was high for all of the leachate and changed over the range of 95.5-98.3 %. It decreased concentration BOD$_5$ to 5 mg O$_2$/dm$^3$ for 5 % vol. share of leachate to 7 mg O$_2$/dm$^3$ for 10% vol. and to 10 mg O$_2$/dm$^3$ for 15 % vol.

The research results indicate that the degree of removal of organic carbon significantly decreased with the share of leachate at the level of 15 % vol. The concentration of total organic carbon for this leachate, the content was reduced from 642 mg/dm$^3$ and in case of raw sewage to 64 mg/dm$^3$. It was an equal high degree of removal of the pollutant load at the level of 90 %. However, the concentration of organic carbon exceeded standards which should have wastewater before drainage to surface water. Moreover, the shares of leachate in a treated mixture were at the level of 5 % by and 10 % vol. allowed to decrease in the concentration of organic compounds expressed as TOC level at 22 and 25 mg/dm$^3$.

Considering the values of organic pollution indicators characterizing treated wastewater, the most advantageous percentage of leachate in the dairy wastewater mixture was their share at the level of 10 % vol.

Let’s focus on the content of nitrate-nitrogen in the effluent from membrane bioreactor. The main problem during the co-treatment of effluents with municipal wastes was the reduction of nitrate-nitrogen to the normalized value, i.e. 30 mg/dm$^3$ [7-9]. Its amount in treated wastewater exceeded twice the permissible value. Therefore, an attempt to co-treat the leachate with dairy wastewater was made. Considering nitrate nitrogen, this significantly improved the efficiency of this biogen reduction.

The concentration of N-NO$_3^-$ did not exceed 3.0 mg/dm$^3$ for all tested percentage of leachate. The degree of removal this biogen was from 95.5 to 67 % (Fig. 3). In association with that, the total nitrogen was removed with greater efficiency too. Its concentration in
the effluent for 10 % vol. leachate was equal to 4.5 mg N/dm$^3$, and for 15 % vol. was equal to 7 mg N/dm$^3$. This corresponded to levels of removal of 98 and 94.4 % respectively. It was claimed that the amount of leachates in a mixture with dairy wastewater do not affect the effectiveness of N-NH$_4$ removal. The degree of removal of this parameter was at the level of 95 %. During the nitrification, the concentration of ammonium nitrogen did not exceed 3 mg N-NH$_4$/dm$^3$.

But there was a problem with the removal of phosphorus, which amount of phosphorus in bioreactor exceed standards. According to Regulation of the Minister of the Environment of 28 January 2009, the concentration of total phosphorus should not exceed 2 mg/dm$^3$. The concentration of P-PO$_4^{3–}$ in treated wastewater for 5 % vol. was at the level of 3.0 mg/dm$^3$, 10 % vol. - 2.8 mg/dm$^3$ and 15 % - 3.0 mg/dm$^3$. It is really difficult to remove phosphorus and nitrogen simultaneously. In this study, in the first phase of MSBR operation, wastewater contained significant amounts of nitrate nitrogen and ammonium nitrogen, which could make difficult their dephosphatation in the anaerobic part of the cycle. In general, phosphorus is well removed (at short sludge age), and it allows to intense the denitrification, but it makes difficult the process of nitrification. Probably, to improve the effectiveness of phosphorus removal should be modified cycle of MSBR working. On the other hand, it can be used the additional process of additional chemical precipitation of phosphorus, using recurrently or in case of observing the change of wastewater parameters. The technology of chemical precipitation of phosphorus is commonly used in municipal wastewater treatment plants. It is a great addition to biological processes, increasing the degree of phosphorus removal, at a relatively low investment costs and easy implementation [25].

**Toxicity tests**

The next step of the study was a toxicological analysis of wastewater co-treatment before and after biological treatment. Because being susceptible to toxic substances can be different in case of groups of species or organisms, so two organisms were tested.

Results of toxicity of raw leachate, dairy wastewater, and mixtures were shown in Table 5.

Based on these results it can be concluded that both municipal landfill raw leachate and dairy wastewater are toxic to *Daphnia magna*. In both cases, all organisms were killed. Moreover, at first, it was diluted twice and then was diluted four times, but no organisms survived the high dose of toxicant, which was in the sample. With a six-fold dilution of leachate, 75 % of shellfish lived after one day. The next day, however, only one living *Daphnia* was noticed from among the initial 20.

The analysis of the toxicity of leachate using the *Daphnia magna* bioindicator was also carried out by Przypadek [26]. He analysed the leachate from the landfill in Stary Sacz. Based on the conducted studies, it showed that inhibition of physiological processes concerned 90 % of individuals (in samples collected in January) and 100 % (in July). This confirmed the conclusion that leachates from municipal landfill belong to harmful and toxic substances for humans and the environment [26].

In the case of dairy wastewaters, the first day recorded 4 dead organisms (80 %), and another day their amount increased to 100 %. Authors were comparing the highest dilution of leachate and dairy wastewater, as result leachate was less toxic.
Co-treatment of municipal landfill leachate with dairy wastewater in membrane bioreactor

Table 5
Toxicity of leachate, dairy wastewater and mixtures for *Daphnia magna*

| Subject of research | Dilution [%] | Number of immobilized *Daphnia* after 48 h | Inhibition [%] |
|---------------------|--------------|-------------------------------------------|----------------|
| Leachate            | 100          | 20                                        | 100            |
|                     | 50           | 20                                        | 100            |
|                     | 25           | 20                                        | 100            |
|                     | 12.5         | 19                                        | 95             |
|                     | 6.25         | 0                                         | 0              |
| Dairy wastewater    | 100          | 20                                        | 100            |
|                     | 50           | 20                                        | 100            |
|                     | 25           | 20                                        | 100            |
|                     | 12.5         | 20                                        | 100            |
|                     | 6.25         | 7                                         | 35             |
| Dairy wastewater + 5 % leachate | 100 | 20 | 100 |
|                     | 50           | 20                                        | 100            |
|                     | 25           | 20                                        | 100            |
|                     | 12.5         | 14                                        | 70             |
|                     | 6.25         | 4                                         | 20             |
| Dairy wastewater + 10 % leachate | 100 | 20 | 100 |
|                     | 50           | 20                                        | 100            |
|                     | 25           | 20                                        | 100            |
|                     | 12.5         | 15                                        | 75             |
|                     | 6.25         | 4                                         | 20             |
| Dairy wastewater + 15 % leachate | 100 | 20 | 100 |
|                     | 50           | 20                                        | 100            |
|                     | 25           | 20                                        | 100            |
|                     | 12.5         | 17                                        | 85             |
|                     | 6.25         | 3                                         | 15             |

Focused on *Vibrio fischeri* bacteria, it was proved that inhibition of luminescence decreased with increasing wastewater dilution. Figure 2 shows a comparison between the degree of inhibition of bioluminescence after 15 min and raw leachate, dairy wastewater, and their mixtures.

Fig. 2. Effectiveness of inhibition of luminescence of dairy wastewater, leachate and their mixtures
Toxicological research shows that the mix with different proportion of leachate volume was less toxic than each of the substrates separately, both to *Daphnia magna* and *Vibrio fischeri*. At 5 % vol. the luminescence inhibition was at the level of 41.78 %, while the toxicity of dairy wastewater was equal to 95.3 %, and of leachate was equal to 64.35 %. Toxicity testing of mixtures was repeated three times to confirm obtained results. The only degree of luminescence inhibition of mixture (15 % vol.) increased by 1.39 %, but it was still smaller than dairy wastewater. It can be supposed that antagonism was caused by the interaction of the components of leachate and dairy wastewaters. Table 6 shows the toxicity of wastewater which was converted to toxic units (TU).

**Table 6**

| Type of sample                      | Toxic units - *TU* |
|------------------------------------|--------------------|
|                                    | *Vibrio fischeri* | *Daphnia magna* |
| Landfill leachate                  | 1                  | 18.62            |
| Dairy wastewater                   | 7.76               | 26.30            |
| Dairy wastewater 5 % vol. leachate | 0.82               | 13.27            |
| Dairy wastewater 10 % vol. leachate| 1.12               | 13.80            |
| Dairy wastewater 15 % vol. leachate| 1.82               | 15.31            |

Based on the results obtained, the toxicity of dairy wastewater and leachate to *Vibrio fischeri* was in the range of 1-10 (according to the Persoone classification), which allowed to define it as toxic. Only leachate with 5 % vol. was defined as moderately toxic. It was observed that more sensitive to pollutants is *Daphnia magna*. The *TU* was in the range of 10-100, which confirmed that wastewater is very toxic.

Based on the obtained results of physicochemical analyses and literature reports, it can be assumed that the toxicity of the tested wastewater mixtures was caused by a high content of toxic ammonium nitrogen in the leachate from the municipal waste landfill [1-4, 27-29].

Pivato and Gaspari performed leachate toxicity tests using bioluminescent *Vibrio fischeri* bacteria. Their studies confirmed that leachate toxicity depends mainly on the concentration of ammonia. Toxicity was lower in sustainable landfills where ammonia decomposed [28].

The literature on the subject can also find research on the impact of heavy metals on the intensity of the toxic effect of leachate [30-33]. For this reason, in this study, the content of heavy metals in leachate was analysed. It was found that the toxic effects of heavy metals present in landfill leachate could be excluded, as their concentrations were very low. The concentrations of individual metals were at the following levels: Pb - 19.3 μg/dm³, Cd - 0.63 μg/dm³, Cu - 38.4 μg/dm³, Ni - 141.1 μg/dm³, Cr - 310 μg/dm³, Zn - 320 μg/dm³, not exceeding the norm values.

In the case of dairy wastewater, the most likely a toxic effect had detergents and disinfectants in them.

Toxicity testing of treated wastewater showed that according to classification, it could be determined as non-toxic, both *Daphnia magna* and *Vibrio fischeri*. Taking into account the *Daphnia magna*, all organisms survived in a bioreactor. Treated wastewater also stimulated the luminescence of *Vibrio fischeri* (Fig. 3).
In the case of wastewater co-treatment at the level of 5 % vol., the value of bioluminescence inhibition was changed over a range of 51-58 % (during longer exploitation). Therefore, in the case of treated dairy wastewater at the level of 10 % vol., after 5 minutes, luminescence inhibition was equal to −13.95 %, and after 15 minutes decreased to −17.47 %. It was also observed that at the same time, the share of leachate increases, and simulation decreases. This can be explained by the fact of staying in the refractive compounds, which included the leachate.

**Conclusion**

The following conclusions can be drawn from the present study:

- The results revealed that the volume of leachate in the treated mixture should not exceed 10 % vol. The concentration of COD, BOD₅ and TOC in the mix of treated wastewater were equal to 120, 7 and 25 mg/dm³, and so the values were lower than standard types.

- Introduction dairy wastewater to treated leachate had a positive impact on the removal of nitrate nitrogen. Its concentration in treated wastewater was at a level of 1.5 mg/dm³ (5 % and 10 % vol.) however, at the level of 15 % vol. was double. The amount of share leachates in the mixture with the dairy wastewater did not reduce on the effectiveness of removal of N-NH₄⁺, which was equal to 95 %.

- The phosphorus was not sufficiently removed from the tested wastewater; it exceeded the permissible values. Concentration of P-PO₄³⁻ in treated wastewater (5 % vol.) was at the level of 3.0 mg/dm³, 2.8 mg/dm³ (10 % vol.), 3.0 mg/dm³ (15 % vol.). To improve the effectiveness of phosphate removal, the cycle of the MSBR reactor should be modified by a combination of anaerobic and aerobic reaction phases.

- Conducted toxicological studies have shown that in the biological process of municipal landfill leachate and dairy wastewater treatment, was determined total removal of high toxicity to all organisms.

- At 5 % vol. of leachate participation, the inhibition of luminescence was on the level of 41.78 % while the toxicity of the dairy wastewater itself was 95.3 %, and leachate
It can, therefore, be assumed that due to the interaction of components of leachates and dairy wastewater, there was an interaction that weakened the toxic effect, and thus the impact of antagonism.

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Co-treatment of municipal landfill leachate with dairy wastewater in membrane bioreactor

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