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EFFECT OF MUNICIPAL LANDFILL LEACHATE ON THE CONTENT OF NITROGEN FORMS IN UNDERGROUND AND SURFACE WATERS

WPLYW ODCIEKÓW ZE SKŁADOWISKA ODPADÓW KOMUNALNYCH NA ZAWARTOŚĆ FORM AZOTU W WODACH PODZIEMNYCH I POWIERZCHNIOWYCH

Abstract: The study has been carried out on the grounds of a municipal landfill in Brodnica. Water samples were collected from 6 sites in different locations. It has been demonstrated that the values of total nitrogen concentrations as well as mineral forms of this element in leachates, ground and surface water were highly varied. In the landfill leachate, the total nitrogen content reached 216.7 mg N dm⁻³, of which 91.6 % consisted of ammonia nitrogen, 2.9 % was nitrate(V) nitrogen and 0.08 % was nitrate(III) nitrogen.

The quality of deep groundwater sampled from an observation borehole drilled on a site where groundwater flows towards the landfill basin corresponded to water purity class I, but fell to class V for groundwater collected from a piezometer situated behind the landfill basin, where the water flows away from it. The total nitrogen and mineral nitrogen forms in groundwater depended on a sampling site. Further away from the landfill cap, concentrations of total and ammonia nitrogen were lower. Considering the above parameters, the quality of groundwater sampled from a piezometer closer to the landfill was within class V, but water samples taken from a piezometer situated further away from the landfill were assigned class II or III. No negative influence of the landfill on the quality of surface waters has been observed as over 94 % of the determinations of biogenic substances did not exceed the threshold values established for water purity class I.

Keywords: landfill, forms of nitrogen, groundwater, surface water

One of the most serious problems caused by building and operating a municipal waste landfill is the migration of leachate to ground, surface and even confined groundwater. Leachate appears as a result of rainwater seeping downwards through the landfill. A much smaller amount of leachate originates from liquids brought to the landfill with waste or derived from the breakdown of organic matter. In some cases,

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increased quantities of leachate can be attributed to surface and groundwater reaching the landfill [1–7].

Most landfills in Poland are inadequately located and operated. Frequently, the hydrogeological or geotechnical conditions of the sites where landfills were created had not been tested, which means that under a certain combination of hydrogeological conditions migration of leachate in water bearing soil layers may take place over considerable distances unless adequate prevention measures are place.

The aim of this study has been to analyze the potential hazard caused by leachate occurring at an active municipal landfill, which can penetrate the water and soil environment. This paper presents the effects of leachate on pollution of ground and surface waters with nitrogen compounds.

Material and methods

The study has been performed on the grounds of an active municipal water landfill in Brodnica, which belongs to the Przedsiębiorstwo Gospodarki Komunalnej sp. z o.o. (Municipal Services Company Ltd.). The landfill is located 350 m off the Drweca River and 900 m away from the secondary protection zone of a municipal water intake point. To the north, the landfill grounds border with a wastewater treatment plant and west of the landfill there is an animal asylum. There are screens isolating the landfill cap from the groundwater and limiting the migration of pollutants in soil, which have been constructed to control the negative influence of the landfill on environment [8].

In order to determine the effect of leachate on the quality of underground (ground and deep ground) and surface water, measurements of the water table were made along with the physicochemical assays of the leachate and water. Water samples for analyses were taken according to the Polish norms: PN-ISO 5667-11:2004; PN-76/C-04620 and PN-88/C-04632. The samples were collected once every four months from six points set at different locations. The sampled water was tested for the total nitrogen content, ammonia nitrogen (N-NH₄), nitrate(V) nitrogen (N-NO₃) and nitrate(III) nitrogen (N-NO₂). The laboratory analyses were made according to the analytical protocols contained in the Polish norms (PN-82/C-04576).

The samples of leachate were collected from the pump station located behind the landfill basin. Samples of groundwater were taken from four piezometers situated around the landfill basin. Confined groundwater was sampled from piezometer P1 (a model observation borehole for determination the hydrochemical background), drilled on a site where groundwater flows towards the landfill, and from piezometer P2, situated where water flows away from the dump. Samples of groundwater were taken from piezometer P3, which was located near a narrow-gauge rail embankment and from piezometer P4, about 8 meters away from the leachate pump station. Both piezometers were drilled on sites where water flows away from the landfill.

The results were processed statistically using the software programme Statistica version 6 (StatSoft Inc. 2001). The least significant differences (LSD) were determined at the level of significance $p = 0.05$.

The evaluation of the quality of water has been made according to the criteria specified in an appendix to the Ordinance of the Minister for Environment of 11th February 2004 on the classification for presentation of the state of surface and groundwater, methods for monitoring water, interpretation of the results and presentation of the state of water (Journal of Laws, No. 32, item 284) [9] and Ordinance of the Minister of the Environment of 14th July 2006 on execution of duties laid on industrial sewage suppliers and conditions for disposal of sewage to sewage facilities (Journal of Law, No. 136, items 963 and 964) [10].

Results and discussion

Many authors [1, 3–5, 7, 11–14] report that effluents from municipal landfills are highly varied in their characteristics. The chemical composition of leachate as well as its amount depend on the age of a landfill, waste disposal technologies, type and fragmentation of waste, amount of water infiltrating cells of waste and methods applied for land reclamation.

Total, nitrate(V), nitrate(III) and ammonia nitrogen are perceived as biogenic parameters in the light of the Ordinance of the Minister for Environment of 11th February 2004 [9]. Many authors [2, 6, 15–17] suggest that the concentration of nitrogen and its mineral forms in landfill leachate is typically high and varies in time.

In Poland, most active landfills are stabilized and therefore the leachate originating from such landfills possesses small levels of organic compounds but very high concentrations of nitrogen compounds.

The leachate management is one of the most difficult tasks that the Municipal Services Company in Brodnica has to deal with. The reason is that the leachate from the landfill in Brodnica contains a very high load of pollutants whereas the quality requirements imposed on treated wastewater are extremely strict.

In the analyzed leachate, the concentrations of biogenic parameters such as the total nitrogen and mineral forms of nitrogen were much lower compared with leachate from other landfills operated for comparably long periods of time [18–22].

The average concentration of total nitrogen reached $216.70 \text{ mg N dm}^{-3}$, of which 91.6 % consisted of ammonia nitrogen (Fig. 1). The highest total nitrogen content was

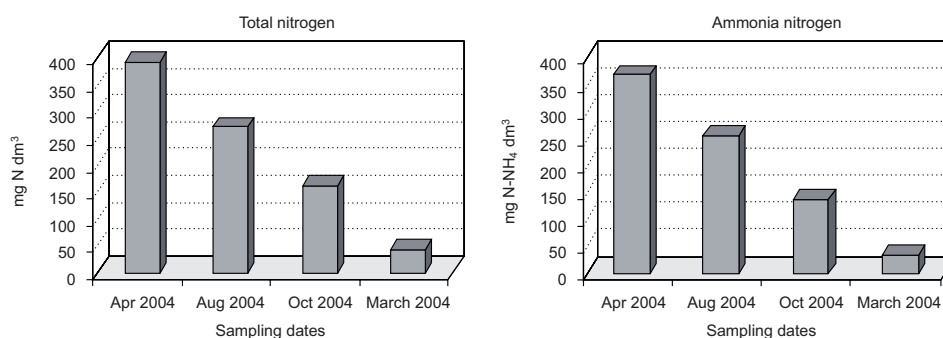


Fig. 1. Total and ammonia nitrogen concentrations in landfill leachate

determined in June 2004, and the lowest occurred in March 2005. Statistical differences in the concentrations of total nitrogen between the sampling dates proved to be highly significant (Table 1). During our study, the total nitrogen concentration decreased considerably, from 392.16 to 42.33 mg N dm⁻³, which could be attributed to the extensive dilution of leachate that took place when the landfill zone II was connected to a system of draining pipes. Another reason was denitrification occurring in the landfill.

Table 1

Statistical calculations for nitrogen forms

Parameter Indices	LSD _{0.05}	Standard deviation	Standard error
Leachate			
Total nitrogen	19.46	149.55	74.77
Ammonia nitrogen	19.15	144.86	72.43
Nitrate(V) nitrogen	3.52	4.90	2.45
Nitrate(III) nitrogen	0.41	0.07	0.03
Groundwater			
Total nitrogen	6.65	12.68	6.55
Ammonia nitrogen	1.83	1.41	0.70
Nitrate(V) nitrogen	6.42	9.09	6.04
Nitrate(III) nitrogen	0.24	0.01	0.01
Deep groundwater			
Total nitrogen	4.47	4.72	2.50
Ammonia nitrogen	1.90	1.56	0.79
Nitrate(V) nitrogen	0.57	0.09	0.05
Nitrate(III) nitrogen	0.25	0.02	0.01
Surface water			
Total nitrogen	3.11	6.28	0.99
Ammonia nitrogen	1.70	1.99	0.12
Nitrate(V) nitrogen	0.47	0.10	0.03
Nitrate(III) nitrogen	0.18	0.01	0.01

Several researchers [2, 3, 16, 23] reported that among the mineral forms of nitrogen, the highest contribution to total nitrogen is made by ammonia nitrogen, which in leachate from stabilized landfills can reach over 2.000 mg N-NH₄ dm⁻³. On new landfills, such a high concentration of ammonia nitrogen is caused mainly by the presence of organic acids, which inhibit nitrification. In turn, ammonia nitrogen in leachate from stabilized landfills originates predominantly from processes of hydrolysis and fermentation of biodegradable organic compounds, which contain proteins.

In the leachate analyzed an external laboratory, there were highly significant differences in values of ammonia nitrogen (Fig. 1, Table 1). The average concentration of this nitrogen form reached 198.40 mg N-NH₄ dm⁻³. Analogously to total nitrogen,

the concentration of ammonia nitrogen, which ranged between 368.55–33.85 mg N-NH₄ dm⁻³, fell drastically over the study period. The highest level of ammonia nitrogen occurred in June 2004 and the lowest – in March 2005.

The average concentration of nitrate(V) nitrogen in the landfill leachate sampled from a collecting well was small (5.69 mg N-NO₃ dm⁻³) but, like total and ammonia nitrogen, highly variable (0.67–11.48 mg N-NO₃ dm⁻³) (Fig. 2, Table 1). Differences in the concentrations of nitrate(V) nitrogen determined between the leachate collected in August 2004 and March 2005 were non-significant, whereas those between the other sampling dates proved to be statistically significant. The highest concentration of nitrates(V) was observed in June and the lowest in August 2004.

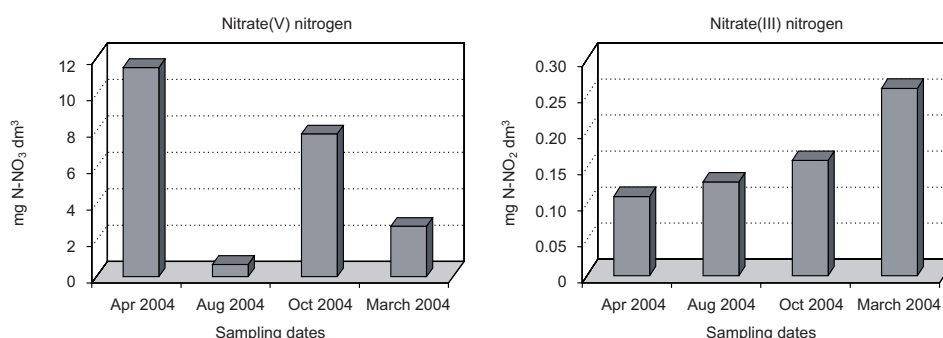


Fig. 2. Nitrate(V) and nitrate(III) nitrogen concentrations in landfill leachate

Our analysis of the fluctuations of nitrate(III) nitrogen shows that the concentration of this nitrogen form continued to increase considerably in time, which indicates that nitrification was at play (Fig. 2). The concentration of ammonia nitrogen over the whole time period decreased largely and eventually fell down to 33.85 mg N-NH₄ dm⁻³.

The influence of a landfill on natural environment can be examined by testing concentrations of pollutants in ground and surface water. The extent of such an influence is measured as a distance from the landfill cap to the line around the landfill where the determined values of pollutants equal those characteristic of the hydrogeochemical background [24–27].

Suchy et al [5] report that waste landfills are usually situated over the surface or close to the surface of the earth, which means they enter the natural water circulation system. Atmospheric precipitations penetrate from the surface land into the landfill masses and seep through the dumped waste, transporting leached pollutants to groundwaters. In Poland, an average 1.000 m³ of water passes through 1 ha of land each year (100 mm/year).

The scale of pollution caused by leachate migrating to ground and surface water can be assessed by observing the quality of water via a network of boreholes (piezometers) or through assays of water in wells near a given landfill [5, 15, 23, 28].

During our investigations, the deep groundwater samples from model piezometer P1 situated outside the landfill basin, where water flows towards the landfill, showed a significant increase in total nitrogen, which was within the range of 0.47–11.04 mg N

dm^{-3} (Fig. 3, Table 1). Such quantities of nitrogen correspond to water purity class IV. The highest values occurred in March 2005 and the lowest – in August 2004. The differences between the values of the total nitrogen content in water sampled in October 2004 and in August 2004 or March 2005 were non-significant, unlike the differences between the determinations obtained in water samples collected in August 2004 and March 2005, which were highly significant. High concentrations of total nitrogen in water collected from the model piezometer are not a measure of the quality of waters originating from the landfill, but imply pollution from other sources in the area where the borehole was drilled.

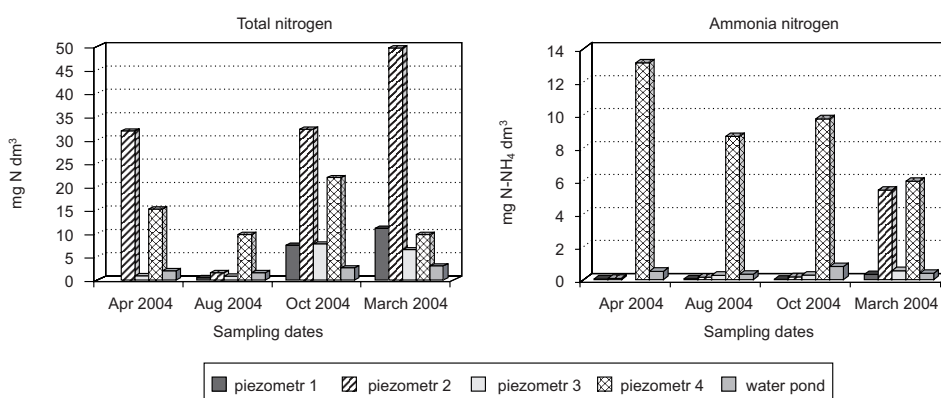


Fig. 3. Total nitrogen and ammonia nitrogen concentration in ground and surface water

The confined groundwater sampled from piezometer P2, located to capture water flowing away from the landfill, was classified as water purity class V according to the total nitrogen content. However, it cannot be stated firmly that such poor quality of water is caused solely by the proximity to the landfill. Several assays imply contamination from other local sources (the animal asylum). Besides, among the parameters most highly exceeded at this site there are the ones which were determined to be much lower in groundwater collected from a piezometer on the landfill itself.

The average value of the total nitrogen concentration in groundwater sampled from piezometer P3, located behind the narrow-gauge rail embankment was $4.99 \text{ mg N dm}^{-3}$, which is assigned to water purity class III, whereas in the water samples obtained from piezometer P4, located behind the landfill basin, the analogous determination equalled to $14.13 \text{ mg N dm}^{-3}$, which allocated the water to class V. In the former case, the highest values of total nitrogen were observed in March 2005, and the lowest ones occurred in August 2004. In the latter case, the highest total nitrogen content appeared in October 2004, dropping to its lowest level in August 2004 and March 2005.

The value of total nitrogen in surface water collected from a water pond ranged around $2.30 \text{ mg N dm}^{-3}$ and that of ammonia nitrogen was at a level of $0.50 \text{ mg N-NH}_4 \text{ dm}^{-3}$, which means that the pond water belonged to water purity class I (Fig. 3). The concentrations of nitrate(V) and nitrate(III) nitrogen were likewise low (Fig. 4).

The level of ammonia nitrogen in deep groundwater collected from model piezometer P1 ranged between 0.02–0.31 mg N-NH₄ dm⁻³, which corresponds to water purity class I (Fig. 3, Table 1). Much higher concentrations of this form of nitrogen were determined in deep groundwater collected from piezometer P2, located on the animal asylum grounds, where the highest values were recorded in March 2005 and the lowest ones appeared in June 2004. An evident increase in the ammonia nitrogen concentration, which ranged within 0.08–5.43 mg N-NH₄ dm⁻³, was found. The average value of the concentration of this nitrogen form was 1.46 mg N-NH₄ dm⁻³, which allocates the tested water to class V. The above suggests that confined groundwater sampled around the landfill does not meet the requirements set for potable and household use waters. The parameter which disqualifies them in that case is the amount of ammonia (over 2-fold above the threshold).

In the groundwater sampled from piezometer P3, the average value of ammonia nitrogen was 0.36 mg N-NH₄ dm⁻³, which classify this water to water purity class II. In the groundwater taken from piezometer P4, the analogous value was 9.39 mg N-NH₄ dm⁻³, meaning that it belonged to class V. In both cases, the highest values were determined in March 2005 and the lowest ones – in August 2004. The high values of ammonia concentrations noticed in observation well P4 suggest the reducing character of the process occurring in the local groundwater.

The concentration of nitrate(V) nitrogen in deep groundwater collected from piezometers P1 and P2 was within the range of 0.36–43.95 mg N-NO₃ dm⁻³ (Fig. 4, Table 1). The average value of this nitrogen form in water collected from model piezometer P1 was 6.03 mg N-NO₃ dm⁻³, which allocated it to class II. In the water sampled from piezometer P2 the determined value was 26.99 mg N-NO₃ dm⁻³, which makes it belong to class V. In the water sampled from piezometer P1, the highest values were recorded in March 2005 and the lowest ones in August 2004. A reverse situation occurred in the case of water collected from piezometer P2.

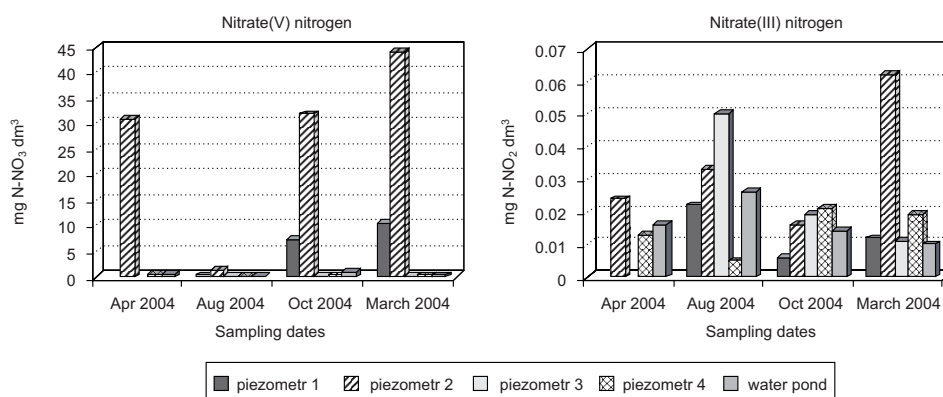


Fig. 4. Nitrate(V) and nitrate(III) nitrogen concentration in ground and surface water

The groundwater sampled from piezometer P3 showed variation in the content of nitrate(V) nitrogen, ranging from 0.06 to 0.15 mg N-NO₃ dm⁻³. In the water obtained

from piezometer P4, the analogous determinations varied from 0.24 to 0.54 mg N-NO₃ dm⁻³. Such values enabled assigning water purity class I. In both cases, the differences in the content of nitrate(V) nitrogen between the sampling dates proved to be non-significant.

The concentration of nitrate(III) nitrogen found in the groundwater collected from model piezometer classify it as belonging to class I. Its level in the deep groundwater sampled from piezometer P2 allocated it to class II (Fig. 4). No significant differences in the concentrations of nitrate(III) nitrogen between the sampling dates occurred at either of the piezometers.

In the groundwater collected from piezometer P3, the average concentration of nitrate(III) nitrogen was 0.027 mg N-NO₂ dm⁻³, which is a value ascribed to water purity class II. In the water collected from piezometer P4, the analogous value was even lower, 0.0145 mg N-NO₂ dm⁻³, which corresponds to water purity class I. The highest levels of nitrate(III) nitrogen were observed in October and the lowest – in August 2004.

Conclusions

1. The analyzed leachate contained, on average, 216.7 mg N dm⁻³, of which 91.6 % consisted of ammonia nitrogen, 2.9 % was made up of nitrate(V) nitrogen and 0.08 % was nitrate(III) nitrogen.

2. The quality of deep groundwater collected from a piezometer drilled on a site where water flows towards the landfill basin corresponded to water purity class I. However, the water samples collected from a piezometer catching water flowing away from the landfill proved to belong to class V.

3. The concentration of total nitrogen and its mineral forms in groundwater depended on a sampling site. The further from the landfill cap, the lower the concentrations of total and ammonia nitrogen. Considering these as water quality parameters, the groundwater sampled from a piezometer closer to the landfill was allocated to class V, whereas the samples taken from a piezometer further away from the dump were belonged to water purity class II or III.

4. The landfill in Brodnica does not have an adverse effect on the quality of surface water, as over 94 % of the determinations of biogenic parameters did not exceed the threshold levels established for water purity class I (very good water quality).

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WPLYW ODCIEKÓW ZE SKŁADOWISKA ODPADÓW KOMUNALNYCH NA ZAWARTOŚĆ FORM AZOTU W WODACH POWIERZCHNIOWYCH I PODZIEMNYCH

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Abstrakt: Badania przeprowadzono na terenie składowiska odpadów komunalnych w Brodnicy. Próbkę pobierano z 6 różnie usytuowanych punktów. Wykazano, że wartości stężeń azotu ogółem oraz mineralnych form tego składnika w odciekach, wodach podziemnych i powierzchniowych były bardzo zróżnicowane. W odciekach składowiskowych średnia wartość stężenia azotu ogółem kształtowała się na poziomie 216.7 mg N dm⁻³, z czego 91.6 % stanowił azot amonowy, 2.9 % azot azotanowy(V) oraz 0.08 % azot azotanowy(III). Jakość wód w głębszych pobieranych z otworu obserwacyjnego usytuowanego od strony napływu wód podziemnych w kierunku niecki składowiska odpowiadała I klasie jakości wód, natomiast pobieranych z piezometru usytuowanego za niecką składowiska na kierunku odpływu wód ze składowiska – V klasie. Wartość stężenia azotu ogółem i jego mineralnych form w wodach gruntowych była uzależniona od miejsca pobierania próbek. Im dalej od czaszy składowiska, tym wartości stężenia azotu ogółem i amonowego były mniejsze. Biorąc pod uwagę wymienione parametry, jakość wód gruntowych pobieranych z piezometru usytuowanego bliżej składowiska odpowiadała V klasie, natomiast pobieranych z piezometru położonego w dalszej odległości od składowiska – II lub III klasie. Nie stwierdzono negatywnego oddziaływania składowiska odpadów na jakość wód powierzchniowych, bowiem ponad 94 % oznaczeń wskaźników biogenych nie przekroczyło wartości granicznych dla I klasy jakości wód.

Słowa kluczowe: składowisko odpadów, formy azotu, odcieki, wody podziemne i powierzchniowe