

Article

Areal Distribution of Ammonium Contamination of Soil-Water Environment in the Vicinity of Old Municipal Landfill Site with Vertical Barrier

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Abstract: The content of the paper is focused on determining the influence of an old municipal landfill site on the pollution of soil and groundwater by ammonium. The assessment of the influence was conducted on piezometric recording basis, laboratory tests and site investigation, which gave information on contamination level and direction of pollutants migration. Based on the groundwater monitoring results, several maps of groundwater level changes were created. Moreover, mapping of ammonium distribution and migration paths within Lubna Landfill surroundings was also provided. The monitoring data show improvement of water quality in almost every piezometer after only a few years from when groundwater protection system was installed at the site. It indicates that reduction of ammonium within the vicinity of the landfill is continuously progressing in time. On the basis of the results obtained, the magnitude of variability in pollutant migration and changes in concentration, as well as efficiency of the vertical barrier were assessed.

Keywords: ammonium; sanitary landfill; groundwater monitoring; pollution

1. Introduction

Landfilling is the oldest and the most common form of removal and disposal of waste. There are many reactions and transformations (chemical, biological and physical) that occur within waste that result in formation of a number of harmful chemicals and substances. The composition of leachate is affected significantly by the type of waste disposed, the degree of compaction, and water content [1–4]. If a landfill’s waste has a high content of organic fraction, leachate is characterized by a higher oxygen content and higher concentration of ammonium and organic nitrogen.

During landfill stabilization, the processes of hydrolysis and partial degradation of macromolecular humic substances predominate. As a result of these reactions, low molecular fulvic and humic acids occur. A degradation of humic substances containing organic nitrogen leads to the increase of ammonium concentration in the leachate, which is released by deamination and ammonification processes [5]. In general, nitrogen is found in leachate at high concentrations [6]. Leachate migrating into the landfill foundations determines a hazard to the groundwater environment and adjacent areas [7,8]. As leachate enters the soil, it can cause long-term or even permanent contamination of an aquifer. Soil and groundwater mostly absorb soluble inorganic compounds in the form of ions, for example Ca^{2+} , Cl^- , Fe^{3+} , K^+ , Mg^{2+} , Mn^{2+} , Na^+ , NH_4^+ , HCO_3^- , NO_3^- , SO_4^{2-} , and a number of organic and mineral substances [9,10].

Nitrogen can be found in a groundwater environment as dissolved organic nitrogen (DON), nitrate (NO_3^-), nitrite (NO_2^-) or as ammonium ions (NH_4^+). The most common nitrogen compound found in groundwater is NO_3^- , but in strongly reducing environments NH_4^+ can be the dominant form [11]. NH_4^+ is found in the groundwater naturally as a result of anaerobic decomposition of organic material. In addition to the reduction zone, where intense decomposition of organic matter occurs, there are also temporary oxidation zones nearby landfill sites (Figure 1).

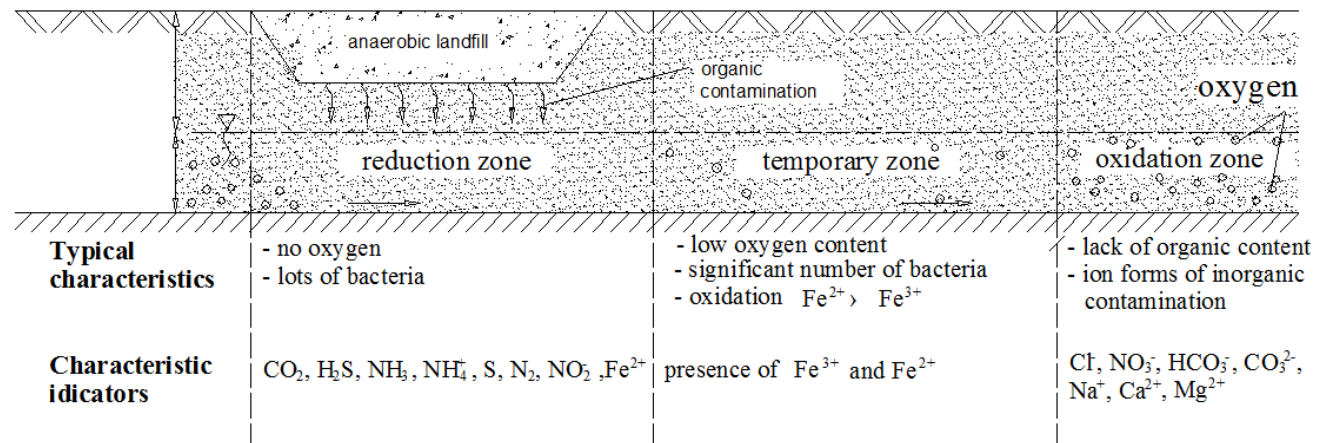


Figure 1. Pattern of contamination zones migration within landfill’s subsoil.

NH_4^+ is also commonly found in groundwater as the consequence of anthropogenic activities, mostly due to leaching from organic waste disposal, leaking sewage systems or the spreading of fertilizers [11,12]. High levels of NH_4^+ in an environment indicate that groundwater is contaminated by anthropogenic sources and may contain pesticides, pathogens or other inadvisable substances [13,14]. The potential nitrogen transformation processes that may commonly occur in landfill environment are:

Ammonification, nitrification, denitrification, immobilization, sorption, volatilization and anaerobic ammonium oxidation (Figure 2) [15].

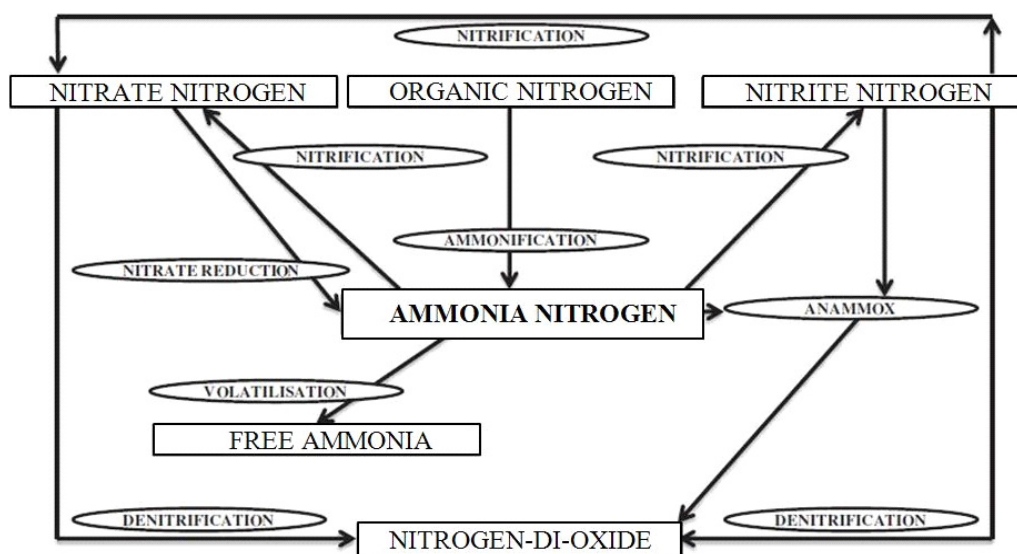


Figure 2. Potential nitrogen transformation pathways/nitrogen cycle that may occur in landfill environment.

One of the major environmental problems associated with landfilling municipal solid wastes is connected with the discharge of leachate into the environment, which may cause serious pollution to groundwater aquifers and surface waters. The leachate from municipal solid waste landfills contains large concentrations of ammonium and organic substances, which results in several environmental problems, for example it can contribute towards eutrophication and acidification and is toxic to many organisms [15–18]. Monitoring programs generally concern sampling of water and ground surrounding the landfill in order to determine characteristic contaminant indicators, such as ammonium, chlorides or heavy metals concentration. This approach is not always effective, especially when background levels of many chemical indicators are high. Accordingly, there is a real need for the application of the methods that can positively identify leachate as the source of contamination in a monitored environment. Many scientific studies prove that the isotopic approach can face this problem more effectively, and owing to its ability of tracing leachate in the environment, can offer more information than traditional techniques. The most commonly used tracers, like ^2H , ^3H , ^{15}N , ^{13}C , ^{34}S , ^{18}O , ^{11}B , and $^{87}\text{Sr}/^{86}\text{Sr}$, may also give information about the occurrence of leachate pollution if standard chemical pollution parameters do not show any peculiar anomaly or yield ambiguous results. For instance, nitrogen isotopes can be used to identify sources of nitrates and ammonium in groundwater or tritium, owing to its sensitivity and stability, can be used as a leachate pollution tracer. The use of tracers provides a better understanding of the leachate production and attenuation mechanisms and confirms whether contamination comes from a municipal landfill or from other local sources.

Ammonia (NH_3) and organic-N, collectively known as total Kjeldahl nitrogen (TKN), represents a high percentage of the total soluble nitrogen compounds in leachate [19]. Due to the anaerobic condition within landfills, concentrations of nitrite and nitrate are typically low. Nitrate is very mobile and is closely monitored for health reasons. It may also indicate whether or not a landfill has entered

its anaerobic stage. In the anaerobic stage, anaerobic decomposition dominates and the entire landfill is in a chemically reducing state that results in more ammonia than nitrate or nitrite. If the NH_3 content is too high (e.g., greater than 1000 mg/L), nitrification may be inhibited [20].

Ammonia is water-soluble and is, because of that, deposited on land and vegetation very quickly. Ammonia may also react with acid gases or small particles and is in that case deposited much more slowly and, because of this, potentially transported over much longer distances before deposition. The roots of vegetation take up deposited ammonia, it may also be oxidized to nitrate by bacteria in the soil and the nitrate is then taken up by vegetation. This process contributes towards soil acidification. Low concentrations of ammonia lead to positive manuring effect, whereas too high concentration results in toxicity. The proportion of ammonium in the form of ammonium nitrogen depends on the temperature, pH, ammonia release and ionic strength. When the pH is higher, more ammonia is produced and at lower pH values more ammonium is produced instead. When the pH is approximately 8 at the temperature 180 °C, ammonia is approximately 10% of the total ammonium and ammonia is only 1% when the pH is 7.5 and the temperature is 18 °C. It is mainly ammonia that causes toxic effects on aquatic organisms [21].

2. Materials and Methods

2.1. Study Area

The Lubna Landfill is located to the northeast side of Warsaw, within 1 km distance from a village and approximately 30 km south from the city of Warsaw. The area of the landfill belongs to Góra Kalwaria district, however the northeast corner of the landfill is next to the boundary of Konstancin-Jeziorna district [22].

The nearest western surrounding area of the landfill is recognized as wasteland with grassland plants. Further from the landfill, there are forest, grasslands and arable fields. The woodland is a legally protected landscape. The location map is presented in Figure 3. Within the landfill environment there are also former clay-pits, from where clay was extracted to supply several local brick factories (all of have been closed for several years). Some of the clay-pits are filled with water and some were illegally filled with waste of different composition, including technological compounds (information obtained from citizens). There are also several ‘wild’ waste dumps located on the wasteland, which could have significantly affected the quality of the first level of groundwater.

Lubna Landfill (embankment type) was established in 1978, when no environmental protection system applied. It covers area of approximately 22 ha, and it is almost 60 m high. Since 1996, remediation works have been carried out on the landfill. They include: cut-off wall around the landfill (constructed in 1998), leachate drainage system (constructed in 1997 and 1998), berms (constructed successively), degassing and mineral capping system. The cut off-wall of 0.6 m thickness and length of 2002.8 m was made of ready-mixed bentonite. The depth of the cut-off-wall depends on the geological conditions (5.5–17 m). It was installed approximately 2.0 below impermeable soil layers (Figure 4). The landfill was closed in 2011.

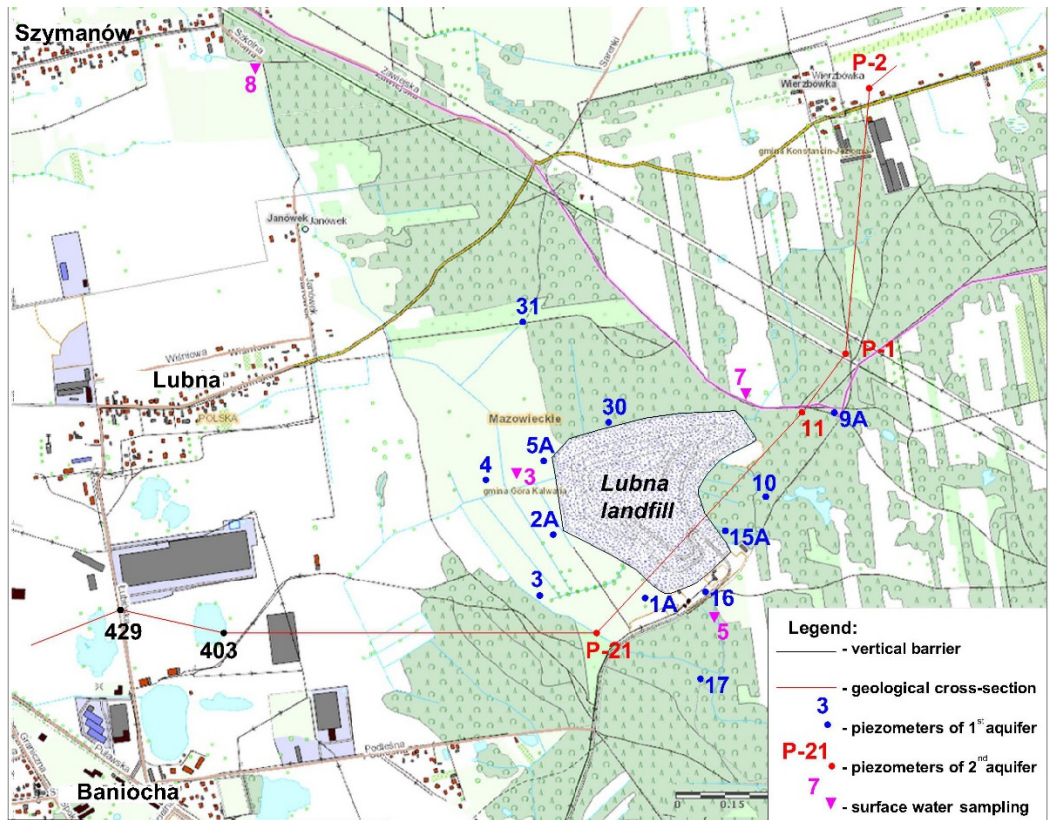


Figure 3. Groundwater monitoring for Lubna Landfill, with geological cross-section.

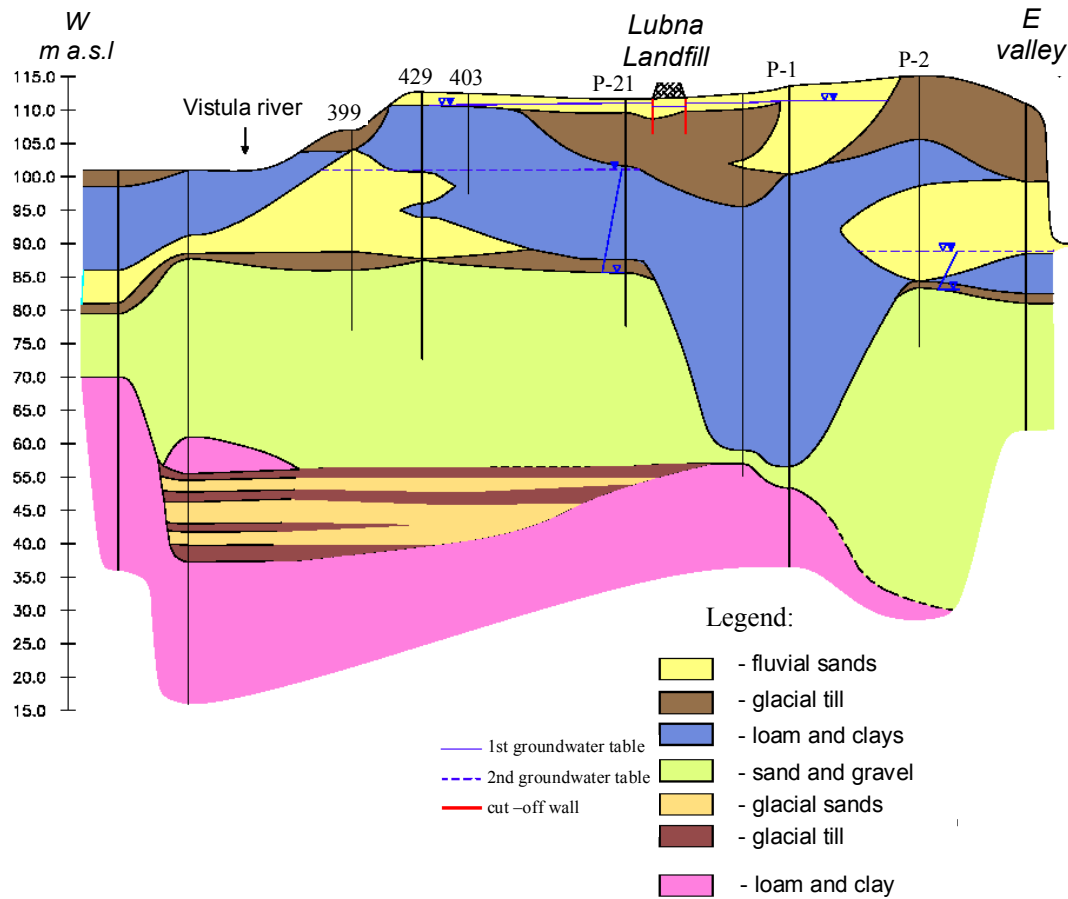


Figure 4. Geological cross-section of Lubna Landfill subsoil.

2.2. Morphology and Geological Structure

The landfill is located within the boundaries of Warsaw Plain, zone of plateau where Vistula flood plain occurs. There are two major geomorphologic formations distinguished: post glacial denudated and non-denudated plateau, and river valleys. It is covered by boulder clays, with several small valleys occurring. The denuded plateau spreads out over the area where no boulder clay layers were observed. The altitude of the area is 105–110 m a.s.l. In the center of the plateau, there is a zone of the fluvioglacial water flows. In locations of previous glacial water flow, the oxbow lake formations could be observed. Nowadays there are several basins filled with organic deposits.

The investigated geological structure of the landfill area proves that the ground is stratigraphic and genetically diverse (Figure 4). The older subsoil (at the depth of 80 m) consists of varicolored clays, loams and sands (Tertiary, Pliocene) and is covered by boulder clays (Quaternary, Pleistocene), consisting of firm sandy clays with lime cobbles, at depth of 60 m. Then there is a 5 m thick complex of sandy-gravel deposits formed during the masovian interglacial. This layer is covered by series of mid-polish glaciation deposits consisting of (from the top) thin layer (3 m) of maximum stadial boulder clay (firm sandy clays), then it transforms into thick complex of marginal lacustrine and varved clays (alternately stratified clays, loams, and silty sands) of the total thickness of 15–20 m (maximum stadial). The upper part forms a complex consisting of unconsolidated boulder clays (sandy clays, clayey sands) covered with the discontinuous fluvioglacial sandy topsoil layer of various thicknesses (0.5–15 m). A topsoil layer consists of loams with peat deposits occurring locally.

2.3. Hydrography and Hydrogeological Conditions

The surface runoff of the landfill area is collected by ditch No. 2. The water from the ditch falls into a narrow watercourse, which directs it to Vistula River. Within the scope of the landfill remediation plan, dated on 1996, the leachate from a landfill was supposed to be collected by ditch No. 2, however due to the lack of the water treatment expected effects, the leachate was being transported to the liquid municipal waste treatment plant.

On the south part of the landfill there is ditch No. R. It drains water into the near waterway. Ditch No. R goes through woodlands, and has not been maintained properly for many years. Now it is water filled only during freshets (snow melting, long-term precipitation). The distance from the landfill to Vistula River is approximately 4 km (to east), and to the nearest small watercourse, it is 6.5 km distance (northwest).

On the investigated area there are two quaternary aquifers. Moreover, within formations separating these two levels, the saturated clayey and sandy interbeddings of a small thickness occurs. For detailed geological structure of the landfill subsoil please see Figure 4.

First level of groundwater table is located within topsoil consisting of fluvioglacial sands. The drilled depth of groundwater table falls in the range of 0.1 to 1.8 m. The groundwater flow is determined by infiltration of rainfall and by the local drainage. The analyzed aquifer was contaminated by the landfill leachate and contaminants, washed out from rainfall water, all coming from infiltration through the waste disposed on the landfill from 1978 to 1998 (before the cut-off wall construction). After the end of the cut-off wall construction in June 1998, a process of leachate infiltration through the first aquifer and to ditches was successfully eliminated. The existing drainage, leachate

recirculation system [23], network of ditches and the cut-off wall bypassing the landfill resulted in significant changes of the groundwater flow direction and its velocity, comparing it to the primary hydroisohypse lay-out.

The readings from piezometers allowed the determination of the groundwater flow direction: northwest direction (ditch No. 2) and southeast direction (ditch No. R). Depending on the first aquifer groundwater table level the direction of flow could be changing periodically. During the high groundwater level conditions, water accumulates at the surface forming local ponds. The hydraulic conductivity of first aquifer soil is as follows:

- fine sands and silty sands— $k = 3 \times 10^{-5}$ m/s; and
- medium and coarse sands— $k = 2 \times 10^{-4}$ m/s.

The second (main) aquifer is located within a layer of masovian interglacial saturated sands of 40 m thickness, and is a primary usable level (Quaternary). It is being explored through the deep well. It is separated from the first aquifer by two layers of boulder clay: medium stadial of hydraulic conductivity $k = 2.7 \times 10^{-9}$ m/s, and maximum stadial of hydraulic conductivity $k = 3.7 \times 10^{-10}$ m/s [24]. These forms are moderately permeable and impermeable. The second subsoil deposits (underlying boulder clays), separating the usable groundwater table and the ground surface, consist of marginal fluvioglacial deposits, with thin interbeddings of firm clays, silts, and silty sands (genetically-varved clay).

The isolating layers of silty sands, in deeper parts, are connected to the usable level. The vertical migration through this complex of deposits could be described as a long-term percolation process. It is a practically impermeable layer protecting the usable level against the contaminants infiltrating from surface. The aquifer connected to the usable level consists of medium sands of $k = 2 \times 10^{-4} - 6 \times 10^{-4}$ m/s. The water from excavated wells was used for land farming purposes [25].

The runoff of the groundwater of this level is directed to Vistula Valley (to the northeast direction). The approximate velocity of infiltration within this layer, for the gradient of 4.2‰, equals $v = 8.4 \times 10^{-2}$ m/s. The yield of the well in this section is 30–75 m³/h. The groundwater table is located at the depth of 10–11 m (piezometer no P21-inflow direction) and at the depth of 24.5–27 m (piezometer P-1 and P-2- outflow direction).

2.4. Sampling Methodology

For the purpose of quality assessment of the first aquifer groundwater and also the surface water, since 1994 there has been a monitoring system maintained at the Lubna Landfill site [24]. Local observations allowed recording groundwater quality changes during the reclamation works and also afterwards. The monitoring system was also used to assess the efficiency and quality of all engineering activity focused on remediation of the contaminated site [26]. Due to some piezometers being damaged during the vertical barrier construction (in 1998), some of them had to be replaced with new instruments installed at the site. The final monitoring system consisted of 12 shallow piezometers, which have been providing records since 1994. There are also 4 wells located near ditches that have been observed for the last 18 years (Figure 3).

Another important aspect of groundwater monitoring concerns the problem of landfills impacting agricultural lands in terms of nitrogenous compounds contamination. Studies on this issue were conducted in rural areas, located to the east direction of the Lubna Landfill.

Quality tests of surface and groundwater samples were carried out focusing on 15 selected pollution indicators. These were: pH, ammonium, PTEs (Cu, Zn, Pb, Cd, Cr, Hg), electrolytic conductivity, total organic carbon (TOC), polycyclic aromatic hydrocarbon (PAH), chemical oxygen demand (COD), biochemical oxygen demand (BOD), chlorides and sulfates. The physicochemical analyses of samples were accompanied with measurements of groundwater level changes in piezometers and wells, and also the flow of water through the investigation ditch when flooded. In addition, measurements of the rainfall were carried out as part of monitoring program for the landfill. The average value of a rainfall for analyzed area was 556 mm, the minimum was at level 530 mm and maximum at level 620 mm per year. The concentration of specific indicators shows stronger connection with the groundwater level (supplied by rainfall and thawing snow). According to monitoring data, groundwater level in all piezometers shows seasonal pattern. The highest levels are recorded in March and April and the lowest in August and September. It should be noted that in accordance to high groundwater level, lower concentration of indicators is noted and conversely.

Since 2006, the measurements were collected once every six months, and during a period from 2007 to 2010 the frequency was doubled (time schedule adopted according to Polish Standards). Due to the landfill closing, the measurements after 2011 were again limited to every six months. The observation network of the first aquifer groundwater within the landfill site is presented in Figure 5.

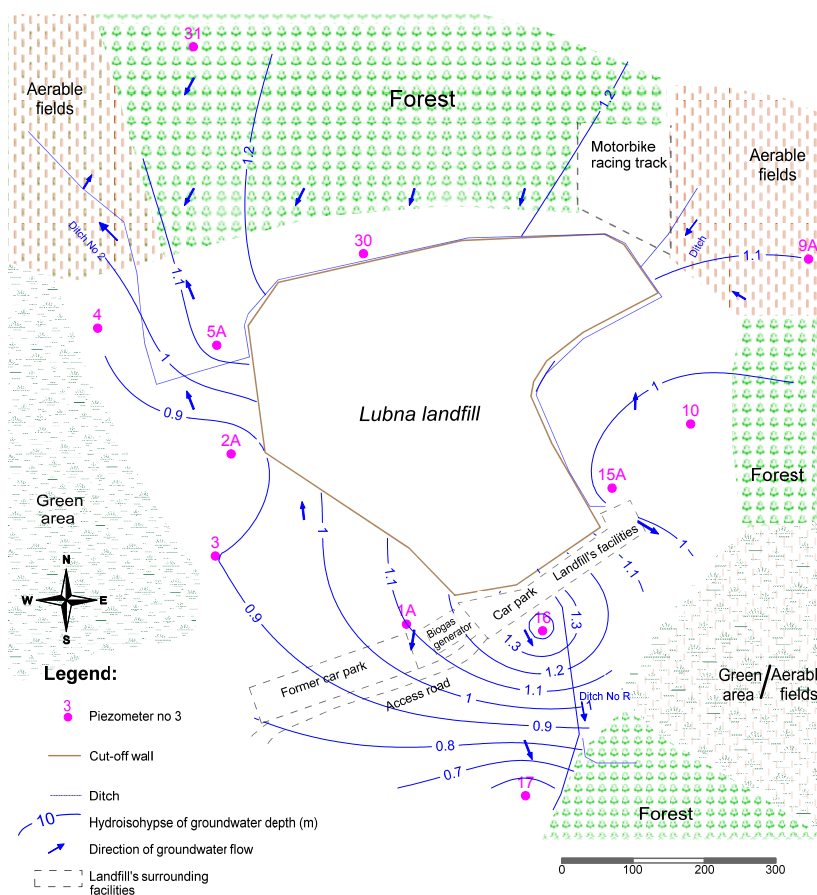


Figure 5. The groundwater depth contour map for Lubna Landfill surroundings (2014).

It consists of 3 piezometers located on the east side of the landfill (9A, 10, 15A), then within the zone of groundwater discharge from the landfill there are 7 piezometers, 3 on the south (1, 16, 17) and four on the west side of the landfill (2A, 3, 4, 5A). The last two instruments are located to the north, where the ground water inflow was defined, these being piezometers Nos. 30 and 31.

To avoid any misreading during the physical and chemical analyses of samples, all of them were collected with great care to eliminate potential errors. It is estimated that 30% of readings are biased due to inappropriate method of sampling and transport of samples [27]. Up to 2004, the adopted sampling procedure was the one recommended in the Polish Standards. Since, procedures were change in accordance to ISO 56673, [28] and ISO 5667-11 [29]. Before sampling, the top water in each piezometer was carefully pumped out. The calculations of pumping time were based on volume of the water in each piezometer and the efficiency of the pump. To assure the appropriateness of sampling, each pumping was accompanied with simultaneous measurement of pH, temperature and electric conductivity. The samples were collected into smoked glass bottles, and when transported kept between 1 °C and 50 °C. The measurement of ammonium was commenced straight after the sample is collected, if the time interval exceeded 4 h, the sample was stabilized with the use of sulfate acid. Between 1994 and 2004, ammonium measurements were carried out by direct nesslerization method, recommended in the Polish Standards. Nowadays, the spectrophotometry method has been used, according to European Standard EN ISO 11732 [30], with the use of Spectrophotometer UV-VIS DR 6000 (Figure 6).

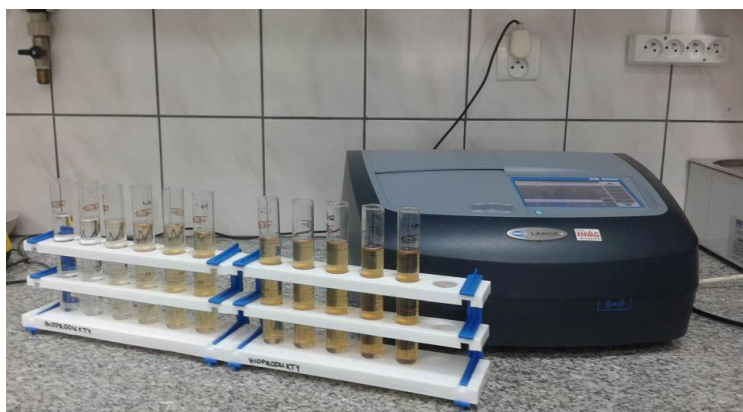


Figure 6. Experimental set-up to determine ammonium in groundwater samples.

3. Results and Discussion

Before reclamation works were introduced at the landfill site, the main two pathways for contaminants migration, including ammonium, were: surface runoff reaching open waterways and infiltrating through the waste body into groundwater. The assessment of the groundwater quality within the landfill site was based on comparison of ammonium contamination levels measured for collected samples with levels required in Polish Standards (Standards adopted from EC Directive) (Table 1).

Table 1. Ammonium concentration changes in time for selected piezometers.

No. Piezometer	Concentration of Ammonium [mg NH ₄ -N/L]				
	Date of Sampling				
	25 November 1996	02 November 1998	03 December 2003	12 November 2008	30 May 2014
1A	3.0	0.8	1.2	0.16	0.05
3	589	588	175	79.2	4.21
4	24.3	3.9	2.3	3.9	3.96
5A	14	2.9	1.2	0.66	0.60
9A	11	0.2	0.3	0.16	0.14
10	18	0.2	0.2	0.4	0.08
11/16	22	1.6	1.4	0.85	2.42

Notes: Directive 2000/60/EC [31] established a framework for community action of water policy and Directive 2006/118/EC [32] on the protection of groundwater against pollution and deterioration: good groundwater chemical class (≤ 1.5 mg/L NH₄-N); poor groundwater chemical class (> 1.5 mg/L NH₄-N).

As stated in Table 1, it is clear that ammonium concentrations exceeded the allowable standard value of 1.5 mg/L. At worst, the concentration reached 589 mg/L (piezometer No. 3 on 26 November 1996). The contamination level decrease continued with time, however some periodical deviations occurred (please see Table 1 and Figure 7).

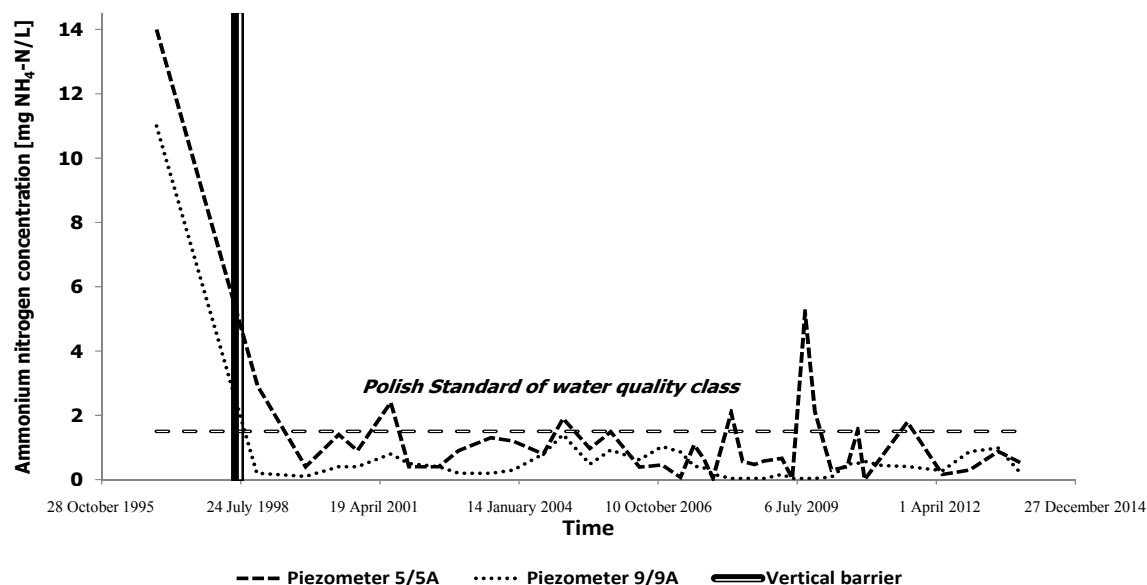


Figure 7. Changes of NH_4^+ concentration on Lubna Landfill in piezometers 5A and 9A.

A significant decrease of concentration could be observed after the construction of the vertical barrier and leachate drainage system. Progressing improvement of water quality was observed in almost every piezometer after only a few years since the groundwater protection system was installed.

Statistical evaluation was performed for monitoring data obtained from seven selected piezometers after the vertical barrier closure (Table 2).

Table 2. Statistical evaluation and summary of ammonium concentrations [mgNH₄-N/L] in the groundwater samples for selected piezometers (after vertical barrier closure).

No. of Piezometer	Amount of Samples	Min. Value	Max. Value	Mean	Median	Skewness	Std. Dev.	% above Directive Limit
1A	38	0.04	2.46	0.56	0.30	1.51	0.60	7.89
3	36	27.30	589	150.54	78.70	2.02	147.76	100.00
4	38	0.07	57.30	6.39	2.22	3.19	12.73	63.16
5A	39	0.04	5.20	1.06	0.66	2.11	1.03	23.08
9A	39	0.04	1.40	0.41	0.40	0.97	0.34	0.00
10	39	0.04	3.72	0.91	0.56	1.85	0.97	17.95
11/16	38	0.10	5.59	1.51	1.22	1.35	1.29	42.11

The evaluation included the determination of minimum, maximum, mean, median and skewness values of ammonium concentration in tested samples. Additionally, standard deviation and, expressed as a percentage, exceeding values from the directive [32] were determined. Comparing the mean and median, it can be concluded that, in almost all of monitoring piezometers, distribution is asymmetrical. The exception is the piezometer No. 9A, where these values are nearly equal. Considering the

coefficient of skewness, it can be noted that its value is greater than one in almost all piezometers, except piezometer No. 9A, which proves that the distribution of results is right-skewed.

The results indicate that mean values of NH_4^+ concentration exceed the directive limit in only three piezometers. This case particularly shows a significant improvement of water quality in the rest of piezometers in comparison with the initial state (before the vertical barrier construction).

Nevertheless, at some of piezometer's locations only slow improvement could be observed. The assumption is that in this particular location there could be additional external sources of contamination identified, not necessarily caused by the landfill itself. The source could be located outside the vertical barrier, so in this case it will be flushed out in time by groundwater flowing through the first aquifer. Another theory is that probable external source could be the clay-pit where waste was initially disposed.

The maps of ammonium distribution in time clearly show that before barrier installation, the readings from all instruments were giving the range of contamination from 3 to 589 $\text{mg NH}_4^+/\text{L}$ (Figure 8).

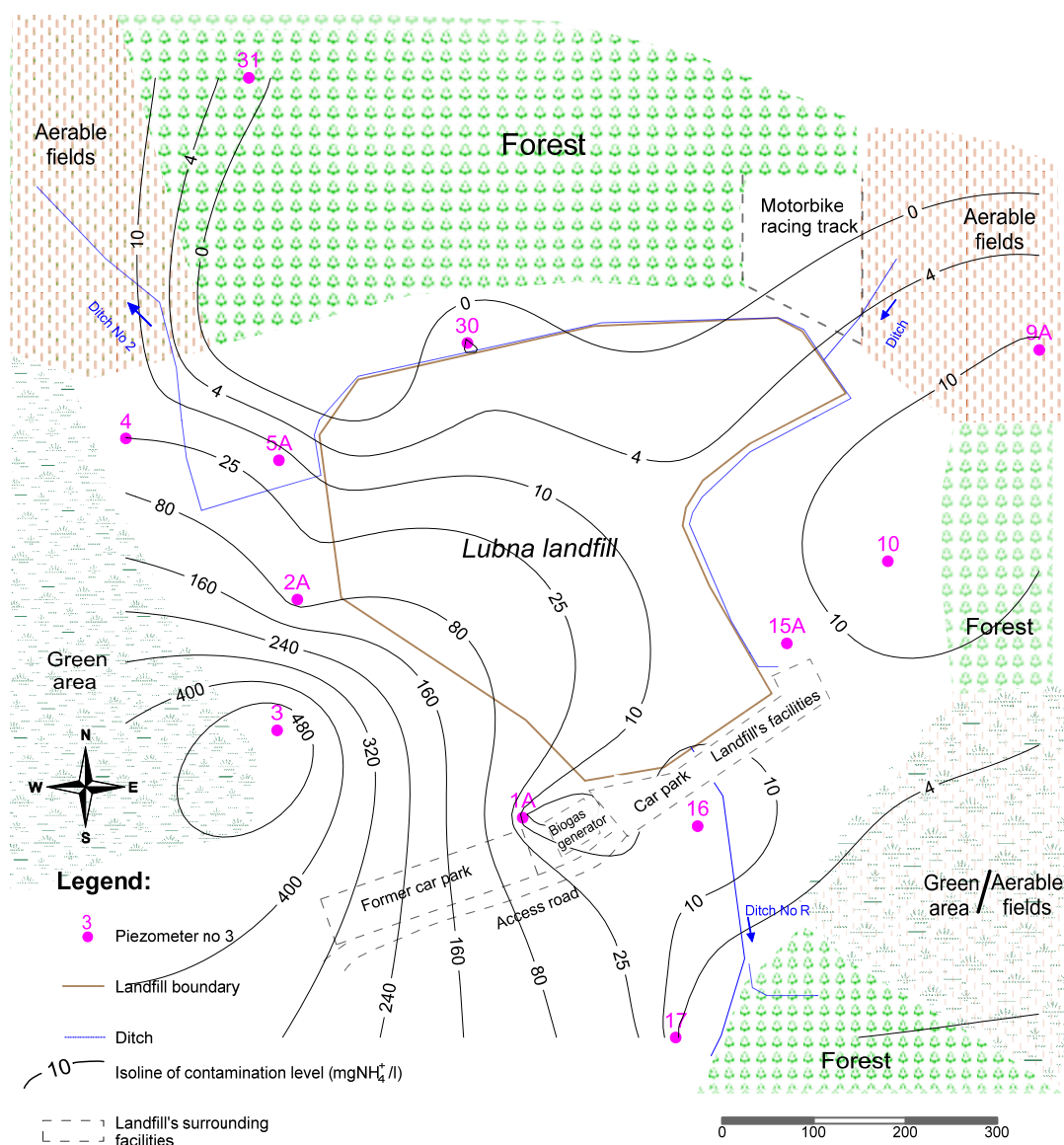


Figure 8. Distribution map of ammonium contamination within Lubna Landfill surroundings (1996), before vertical barrier construction.

The exceptions are piezometers No. 30 and No. 31 located in the water inflow zone. This basically confirms the hazardous environmental impact of the landfill (Figure 9). At five and 15 years after vertical barrier construction (Figures 9 and 10, respectively) raised concentration of ammonium was recorded only in piezometers located in the groundwater discharge zone on west side of the landfill (piezometers Nos. 1A, 2A, 3, and 4). However, initial readings for this location gave the highest contamination levels. The reduction of ammonium within the vicinity of the landfill is slow but continuously progressing in time. The concentration of ammonium for the past 10 years (Figures 9 and 10) has decreased as much as 10 times (piezometer No. 2A).

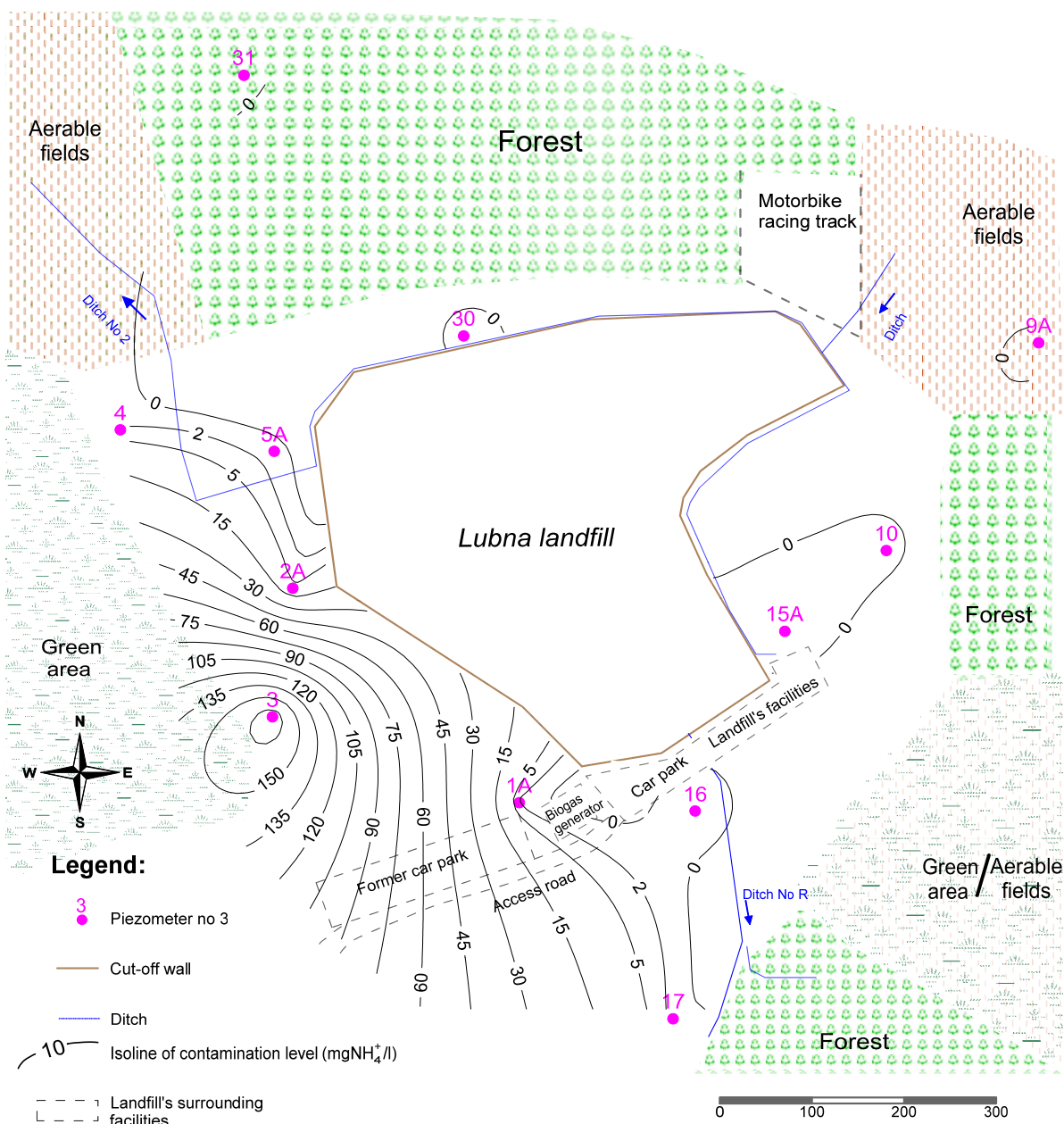


Figure 9. Distribution map of ammonium contamination within Lubna Landfill surroundings (2003), five years after vertical barrier construction.

Due to construction of the vertical barrier and drainage ditches, the initial direction of groundwater flow was diverted. At present, there are two main discharge water paths. The first is influenced by

ditch No. 2 (north-west direction) and the second by ditch R (south-east direction), as shown in Figure 5. Ammonium nitrogen migration covers the path of groundwater flow within the area of the landfill; however, it is limited by the processes of sorption (Figures 9 and 10).

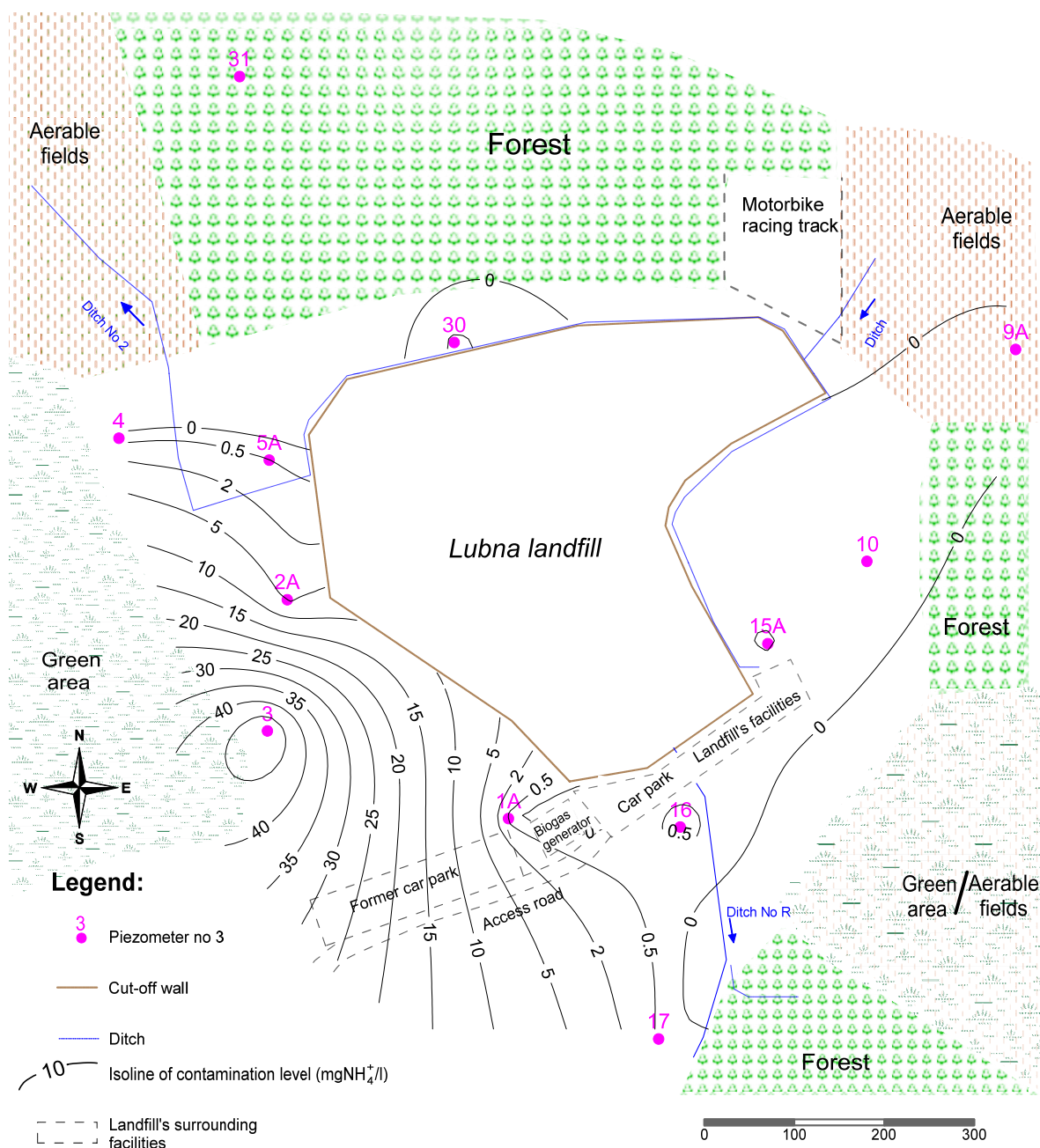


Figure 10. Distribution map of ammonium nitrogen contamination within Lubna Landfill surroundings (2014), 15 years after vertical barrier construction.

4. Conclusions

Monitoring data, collected since 1996, has proven that the primary source of ammonium contamination of groundwater was undoubtedly the municipal waste landfill itself, but also the surrounding so-called “wild waste dumps”. Systematically collected monitoring data show that construction of the vertical barrier significantly improved water quality, which is predominantly reflected in slow and systematic

process of ammonium contamination reduction (even several dozen times). This is observed in almost every piezometer after only a few years since the closure of vertical barrier. Moreover, it is worth noting that this reduction is also supported by natural processes, such as self-purification of the soil-water environment. According to monitoring data, there is also a seasonal pattern for groundwater level changes in all piezometers. The highest levels have always been recorded in March and April and the lowest in August and September. In other words, the higher the groundwater level, the lower concentration of indicators, and *vice versa*.

It must be emphasized that discharge of leachate containing large concentrations of ammonium and organic substances results in several environmental problems, for example eutrophication and serious pollution to groundwater and surface water as well. The monitoring data results presented in this paper clearly indicate efficiency of vertical barrier application and its crucial role in protecting the soil-water environment from contaminants migration and the risks associated with landfills. The recorded values for NH_4^+ still exceed standard requirement in just three piezometers (external pollution sources assumed). However, compared to the initial environment condition, for complete monitoring network (Table 1), a significant improvement of water quality could be observed.

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Author Contributions

Eugeniusz Koda conceived the subject of the paper, literature review and provided the expertise on the research, designed the experimental, laboratory and field work. Piotr Osiński prepared the manuscript, contributed to writing, figure preparation and modeling part. Anna Sieczka was responsible for experimental data analyses, literature review, and writing the conclusions of the research. Dorota Wychowaniak contributed to statistical analyses part and prepared the data set of the research, which will be precisely discussed in a doctoral dissertation.

Conflicts of Interest

The authors declare no conflict of interest.

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