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Variations in physical and chemical parameters of landfill leachates over time

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ABSTRACT

Landfill leachates contain a number of micro-pollutants such as, among other, heavy metal compounds, polycyclic aromatic hydrocarbons (PAHs) and volatile petroleum-derived compounds. Research of physical and chemical parameters of landfill leachates was performed at an active municipal waste landfill during 10-year period. Samples of leachate for testing were taken from piezometers installed in two places, namely sealed bottom of a new section and reclaimed unsealed landfill section. Variations in leachate physical and chemical parameters were observed during landfill operation (pH, chemical oxygen demand (COD), ammonium nitrogen, sulphates, total phosphorus and chloride anion contents). The highest COD values were noted between the fifth and seventh year of research. Elimination of phosphorus compounds proceeded also during the research work. Compounds of Pb, Cu, Cd, Cr, Mn and Zn were found in landfill leachates. Leachates taken from the old unsealed landfill section demonstrated higher concentrations of metals than leachates taken from the sealed part. In the case of PAH, the highest concentrations were noted for acenaphthylene and fluorene as well as benzo(a)pyrene. The share of aromatic hydrocarbons in analysed volatile petroleum-derived compounds was 77%. The highest concentration in tested leachates was noted for benzene. The objective of the research work was to define initial properties of landfill leachates needed for performance of permanent landfill monitoring and evaluation of its impact on the ground and water environment.

Keywords: Sealed and unsealed parts of landfill; Landfill leachates; Migration; PAH; Volatile hydrocarbons; Micro-pollutants; Heavy metals

1. Introduction

Mineral and organic pollutants from landfill leachates featuring definitely high migration properties that flow into underground water are considered toxic and even carcinogenic. The source of such pollutants can also be decommissioned municipal waste landfills. Such landfills should be provided with permanent standard monitoring facilities. In 2016, 53 landfills were reclaimed in Poland [1]. Monitoring of such objects is often incomplete, and even not performed. Various attempts for assessment of hazards, particularly assessment of pollutants flowing into underground water, which may come from landfill leachates, have been made [2–4]. In that respect, research work performed at the real sites, as well as model research simulating probable composition of leachates and migration of pollutants to the soil substratum and underground water has been attempted. In the case of an unsealed landfill, removal of pollutants contained in leachates proceeds partly in the landfill substratum (aeration zone) and in the water-bearing layer (saturation zone) [1]. Elimination of such pollutants in landfill substratum occurs mostly due to mechanisms that initiate, among other, reactions of biodegradation, adsorption, interaction and ion exchange, precipitation and filtration [5]. Those processes depend mostly on the type of soil, content of organic substance and argillaceous minerals therein and on biochemical activity in such medium [6,7]. In the water-bearing layer,

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process of dilution and mixing of polluted water (leachates) with pure underground water occurs [8,9]. Flow velocity of such water depends on the water permeability coefficient and hydraulic gradient. Also processes of sorption, interaction and ion exchange as well as precipitation can occur there.

Oxygen content in pores of the porous (permeable) soil is gradually depleted developing finally an anaerobic environment. At the same time, the bed compaction and colmatation increase due to neutralisation and precipitation of some compounds contained in leachate. This causes reduction of medium porosity, and consequently, increased duration of leachate flow through the filtration bed. Furthermore, pores get blocked to the ever increasing extent with liquid displacing the gaseous phase. Such change of conditions has impact on the share of particular types of purification, therefore, on the final effect of impurities removal [10,11]. Gradual reduction of porosity has, no doubt, impact on increase of mechanical purification of infiltrating leachate. If such medium contains argillaceous minerals, the possibility of water evaporation will decrease, but, at the same time, the role of sorption and ion exchange will increase [2]. The oxidation processes will be less intense, and anaerobic conditions will emerge. New products, resulting from leachate precipitation through the tested bed, will be trapped, to the ever increasing extent, in pores. In biological purification operation, if any, processes in which anaerobic bacteria dominate over aerobic bacteria activity will prevail.

The objective of this research was analysis of physical and chemical composition of landfill leachates containing heavy metals and petroleum-derived compounds, including polycyclic aromatic hydrocarbons (PAHs) and volatile petroleum-derived compounds. Testing of these compounds and variations of their contents in landfill leachates during 10 years was performed in one local functioning municipal waste landfill located in Middle Pomerania. This research work starts constant monitoring of landfill leachates, which can migrate to underground water causing its pollution.

2. Materials and methods

2.1. Materials

Research on physical and chemical composition was performed on samples of leachates coming from municipal waste landfills located in Middle Pomerania. The research works were performed during the period of 2007–2016 with systematic increase of their scope.

Leachate samples for testing of petroleum-derived compounds, heavy metals and other physical and chemical parameters were taken using a special pump (Toń-2) from well-like boreholes located at the landfill site. Those wells were located in two places: sealed bottom of a new section and reclaimed unsealed landfill section. The section having sealed landfill bottom was put in operation in 2012. At that time, selective waste collection was introduced in the area from which the waste came; this had, in a short time, a positive impact on quality and quantity of generated leachates. Leachate samples to be tested were placed in a thermally insulated container and immediately transported to the test laboratory located approximately 10 km away from the landfill. This allowed for maintaining of stable leachate temperature (approximately 22°C) during sampling as well as during sample transport. Slightly elevated leachate temperature indicated occurrence of permanent biochemical transformation processes of pollutants (particularly the organic fraction) contained in leachate [12]. These research have been used to conduct permanent monitoring of the landfill. Leachates to be tested were taken during the May–September period, at least twice per month. To proceed with statistical analysis and making of graphs, averaged values from particular research periods were used.

It was noted that landfill leachates have, among other, considerable concentration of chloride, ammonium and sulphate ions as well as calcium, organic nitrogen, heavy metal compounds, volatile acids and phenols, PAHs and petroleum-derived compounds [8,11,13,14]. In the past years, a number of technical measures including waste compaction, application of protective soil layers, degassing of landfills, reclamation of particular sectors and deposition of partly segregated waste were performed thus leading to considerable limitation of pollution in landfill leachates. Nevertheless, those pollutants still exist in landfill leachates in quantities qualifying them to the micro-pollutants group [8,15,16].

2.2. Analytical methods

Tests on the primary leachate chemical components including chemical oxygen demand (COD), biological oxygen demand (BOD_5), chlorides, phosphorus and nitrogen compounds, pH reaction and conductivity were performed in accordance with applicable Polish legislature (Regulation of the Minister of Environment of 18 November 2014 (Journal of Laws 2014 item 1800) on conditions to be complied with during discharge of wastewater into water or ground and on substances that are particularly dangerous for aquatic environment) [17]. Leachate samples taken for testing of heavy metal contents were tested in atomic absorption spectrometry (iCE 3500Z Thermo Scientific SOLAAR) spectrometer. Samples of leachates for testing were mineralised using a mixture of acids (65% HNO_3 and 30% H_2O_2) using microwave energy (Milestone 1200 Mega apparatus).

Samples of landfill leachates to be tested for content of PAHs and selected petroleum-derived compounds content were taken directly from storage reservoirs using 'Toń-2' bucket. The 'purge and trap' technique was used for determination of volatile petroleum-derived compounds, which were determined in a gas chromatograph (HP 5890) provided with flame and ionising detector as well as purge and trap injector. Non-polar HP1 column was used to separate the mixture of analytes. In such conditions, the method sensitivity level was 0.01 mg L^{-1}. Analytes recovery level was minimum 80%.

Extraction of the tested PAHs was performed using C_{18} (octadecyl) sorbent-filled columns and solid-phase extraction apparatus. The eluate was then condensed in the Kuderna-Danish apparatus. The final analysis was initially performed in gas chromatograph (HP 5890) provided with flame ionization detector and split–splitless injector, whereas during the period of 2013–2016 a non-polar column with 5% phenyl phase HP5 was used to separate the mixture of analytes. In such conditions, the method sensitivity level was 0.01 mg L^{-1}. Analytes recovery level was minimum 80%.

Taking into account the so-called 'US EPA list', the following substances were determined: naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene and indeno(1,2,3-c,d)pyrene. In Poland, it is recommended to determine six PAHs for classification of surface and underground water, that is, pyrene, benzo(b)fluoranthene, benzo(k) fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene and indeno(1,2,3-c,d)pyrene. As representatives of volatile petroleum-derived compounds concentrations of benzene, toluene, ethylbenzene, xylene, isooctane, nonane and undecane were determined.

2.3. Statistical analysis

For statistical analysis of test results, Microsoft Excel software of 2007 version (licence No. 43370567) and STATISTICA of StatSoft version 8.0 (licence No. AXAP712D715618FA-Y) were used. The scope of applied statistical tools comprised the non-linear regression issues and selected statistics available in the basic statistics and tables module of STATISTICA [18].

3. Results and discussion

Monitoring of composition and transformations proceeding in landfill leachates allows for design of special leachate treatment plants and for forecasting of migration of pollutants contained in leachates to underground water. It is then possible to take actions to limit the negative impact of leachates on such water. Examples of impact of landfill leachates on water and ground environment are given in numerous papers. Increased concentration of mineral (heavy metals) and organic compounds (aromatic and aliphatic hydrocarbons) was noted in underground water at the researched municipal waste landfill site [1,12,19].

Selected elements of authors' own research are illustrated in Fig. 1. As it appears from those research works, the processes of biodegradation of pollutants contained in leachates proceeded more actively in the initial period of landfill operation. During the first year of landfill operation, significantly higher concentration of nitrogen V than ammonium ions was noted. The redox potential featured positive values, which indicated oxidising conditions existing in the tested leachates. This condition was maintained fairly stable during the next year although, after a short period (after 2 years of operation), conditions in this environment changed from aerobic to anaerobic, which was indicated by, among other, much higher concentration of ammonium ions than nitrogen V ions and negative redox potential. Over time, pH of landfill leachates changed (from acid, during the initial years of landfill operation) to slightly alkaline in the subsequent years. During the initial period of landfill operation, the most intensely occurring processes of biochemical degradation of organic substance contained in the leachates were observed [16]. Biochemical activity indicator, expressed by BOD_5/COD ratio varied during the research work between 0.81 and 0.72. Other authors noted similar research results [8,19]. After 5 years of landfill operation, those values were maintained within the range of 0.23–0.25, which absolutely indicates certain stabilisation of biochemical processes in the leachates. This originates from the fact that landfill leachates contained, at that time, considerably high amount of hardly biodegradable substances, which were subjected to further biodegradation in the subsequent years. It is quite possible that part of organic substance was absorbed by argillaceous minerals (illite occurs there) or humus compounds [2]. Considerable reduction of phosphate ions concentration was also noted, which could indicate that said ions generated hardly soluble chemical compounds. Contents of sulphate VI, potassium, nitrate V and nitrate III ions decreased considerably. Concentration of chloride ions was maintained at similar level [1]. It appears from the analysis of particular relationships illustrated in Fig. 1, that increased concentration of chloride ions occurs mostly in the range between 137 and 5,800 mg Cl^- L^{-1}, whereas for ammonium nitrogen, within the range from approximately 12 to 790 mg $N–NH_4^+$ L^{-1}. It can be assumed that some amount of chlorides occurring in landfill leachates may be present in the form of ammonium chloride. Variable, although high, values of conductivity were also noted; they varied, during long-term research work, mostly from 3,000 to 13,000 µS cm^{-1}. Increase of this parameter results from mineralisation processes occurring in the tested leachates.

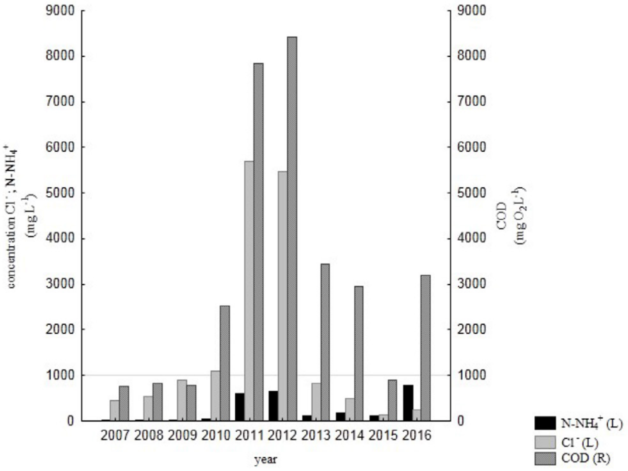

Fig. 1. Changes in selected leachate chemical parameters.

After approximately 10 years of landfill operation, the following was found: increase of total nitrogen and sulphates, pH value, total solids and COD_{Cr} as well as chlorides concentration. This means that non-segregated wastes, including street sweepings, were still deposited in the landfill, particularly during the winter and spring period, being the main source of chlorine compounds [8]. High temperature values, particularly during spring and summer periods, promoted water evaporation processes, therefore, leachate thickening. They caused also high leachate conductivity indicating, however, decreasing trends in the subsequent years. Elimination of phosphorus compounds was still noted probably through phosphorus transformation into a solid phase and/or sedimentation as well as binding by leachate-occurring sorbents, mainly argillaceous minerals and humus compounds [1]. Landfill leachates in the initial years of landfill operation had very intense colour, which was caused, to a major extent, by humus substance content. The research work performed during the period of 2014–2016 indicated that said leachates had straw colour of low intensity, which originated mainly from occurrence of humic acids. At the same time, less and less organic substance has been deposited at the landfill during recent years as such substance is being separated already at the collection stage or during municipal waste treatment in the waste segregation station located at the landfill site. The separated fractions of organic waste were dispatched to compost windrows.

The statistic analysis results are illustrated in particular graphs (Figs. 3–5 and 7–9). Correlations between selected physical and chemical parameters of landfill leachates were also indicated. Examples of correlations are shown in Figs. 2–5; they illustrate categorised graphs of dispersion together with the regression line. As it appears from those graphs, ammonium nitrogen content varied from 20 to 700 mg N–NH_4^+ L^{-1}, which is partly correlated with COD_{Cr} indicator within the limits from approximately 900 to 3,000 mgO_2 L^{-1} (Fig. 2). Ammonium nitrogen content has impact on total solids in landfill leachates (Fig. 3), likewise on chlorine concentration (Fig. 4). Correlation coefficients (*r*) are kept within the range of 0.96–0.99. It appears from the collective histogram (Fig. 5) that landfill leachates feature mainly alkaline reaction (pH from 7.2 to 8.0) [10]. At the initial period of waste deposition in the landfill, the leachates show slightly acid reaction (pH from 6.2 to 6.8), which is specific for the initial phase of organic matter decomposition.

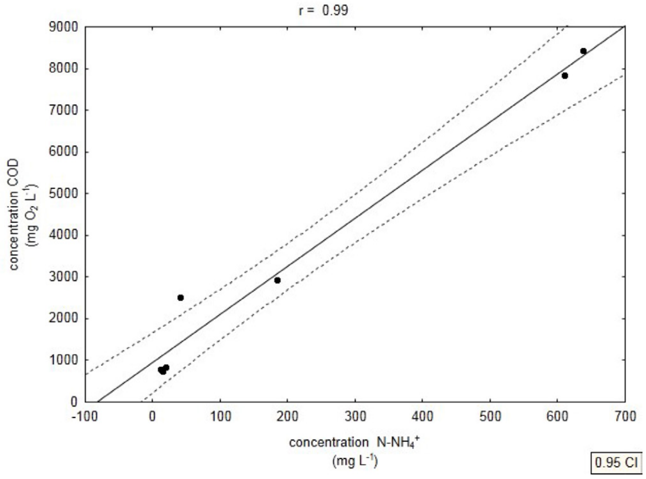

Fig. 2. Categorised dispersion graph together with COD and ammonium nitrogen regression line.

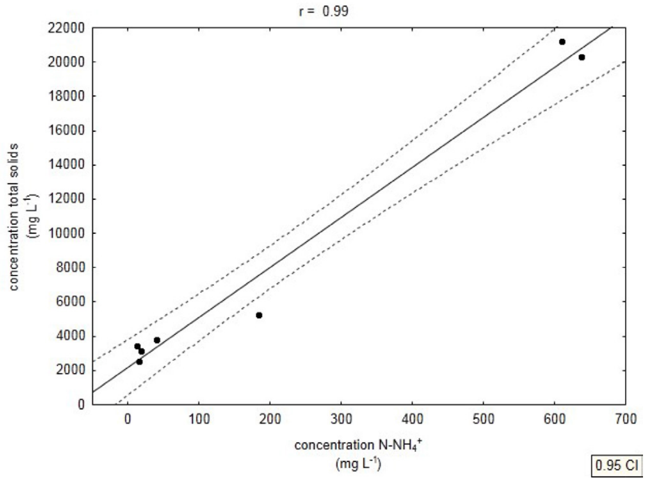

Fig. 3. Categorised dispersion graph together with total solids and ammonium nitrogen regression line.

Heavy metals content in tested landfill leachates did not exceed admissible values defined in the regulation of the Minister of the Environment of 18 November 2014 (Journal of Laws 2014 item 1800) on conditions that must be complied with during introduction of wastewater to waters or to the ground and on substances that are particularly dangerous for the aquatic environment [17].

Average lead content in the tested leachate was 0.01 mg L^{-1}. The highest Pb concentration was noted in spring season and it was 0.15 mg L^{-1}, whereas the lowest value was noted in the summer season 0.01 mg L^{-1}. Copper content in the analogous period varied from 0.02 to 0.32 mg L^{-1}. The average value of this element content in leachate was 0.08 mg L^{-1}. The average concentrations in tested leachates were for cadmium 0.03 mg L^{-1}

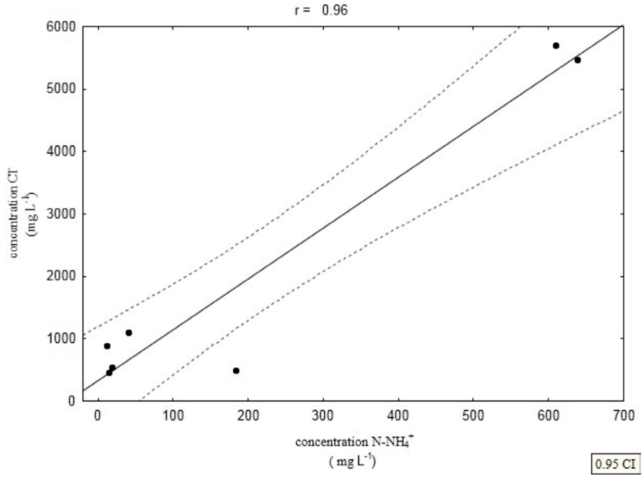

Fig. 4. Categorised dispersion graph together with chlorides and ammonium nitrogen regression line.

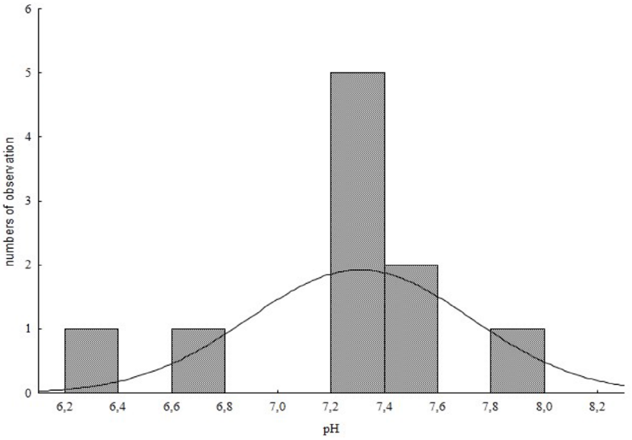

Fig. 5. 2007–2016 leachate pH collective histogram.

and chromium 0.04 mg L^{-1}. The highest concentrations of zinc (average value, 0.41 mg L^{-1}) and manganese (average value, 1.00 mg L^{-1}) were noted in tested leachate taken from the old unsealed part of the waste landfill. Averaged heavy metal concentrations in leachate taken for testing from the sealed and unsealed sectors are illustrated in Fig. 6. Contents of tested metals in landfill leachates depended greatly on weather conditions (average rainfall rate was 550 mm m^{-2}). The leachates taken from the old part featured higher values of concentrations than those taken from the sealed part.

Occurrence of both volatile petroleum-derived compounds and PAHs was determined in landfill leachate samples. Test results for the above compounds are illustrated in Figs. 7–9, which show, in form of histograms, number of findings for each group of chemical compounds with consideration of their sum.

In the case of PAHs, the highest concentrations were noted for acenaphthylene and fluorene having three condensed aromatic rings and for benzo(a)anthracene made of four condensed aromatic rings. In three samples, concentration of naphthalene and benzo(g,h,i)perylene was below the method's sensitivity. The sum of concentration of 16 PAHs was within the range from 100.00 to 2,200.00 mg L^{-1}. The most frequently noted sum value occurred within the limits from 100.00 to 500.00 mg L^{-1} (Fig. 7). Also sum of six PAHs was determined, and its value remained within the range from 2.65 to 1,400 40 mg L^{-1} and mostly remained within the limits from 2 to 200 mg L^{-1} (Fig. 8). Percentage of PAHs in the sum of 16 PAHs, depending on the number of condensed aromatic rings, was analysed (Fig. 10). Compounds having three and four condensed aromatic rings make more than

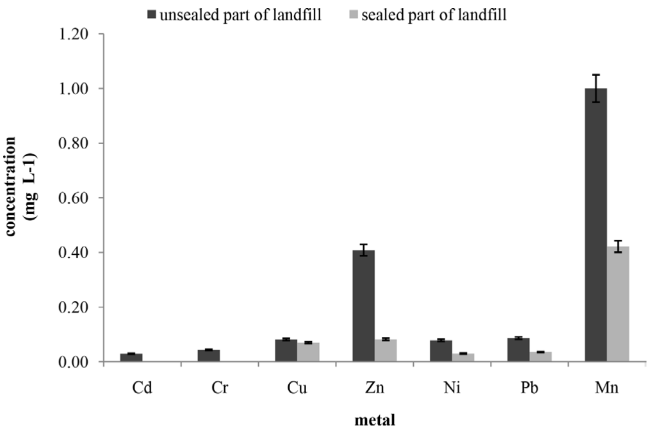

Fig. 6. Average content of tested metals in landfill leachates taken from sealed and unsealed parts of waste landfill.

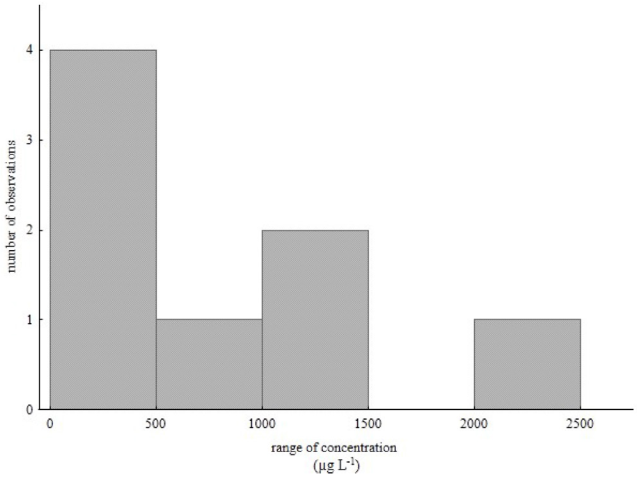

Fig. 7. Histogram: sum of 16 PAHs.

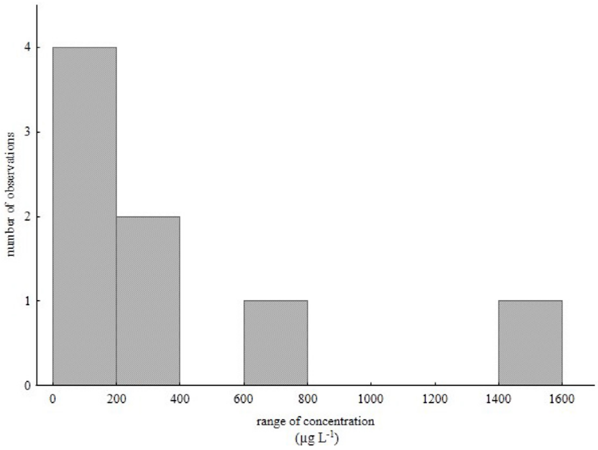

Fig. 8. Histogram: sum of 6 PAHs.

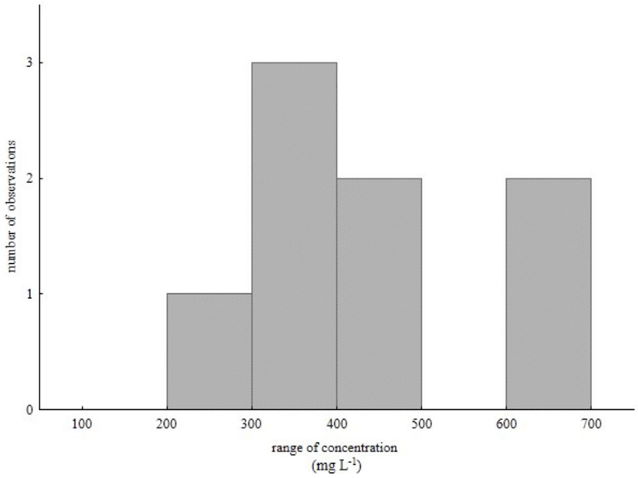

Fig. 9. Histogram: sum of volatile hydrocarbons.

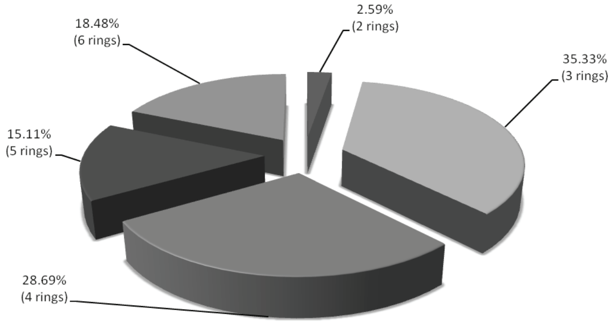

Fig. 10. Percentage of polycyclic aromatic hydrocarbons (sum of 16 PAHs).

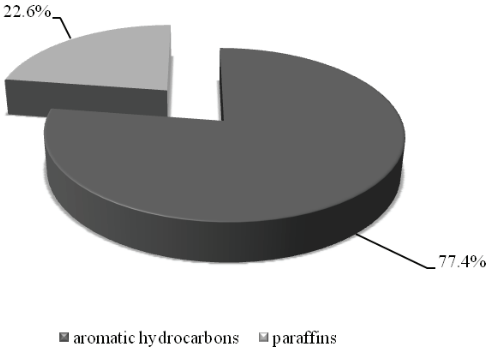

Fig. 11. Percentage of volatile petroleum-derived compounds in landfill leachate.

60% of all PAHs determined in landfill leachate samples. Approximately 30% share of hydrocarbons having five and six condensed aromatic rings results rather from their highest hydrophobic properties than from biodegradation. It appears from the research work performed by Siebielska [20] that in anaerobic conditions number of condensed aromatic rings have no significant influence, in statistical terms, on biodegradation speed. The most profound carcinogenic properties from all determined PAHs have benzo(a)pyrene and then benzo(g,h,i)perylene as well as benzo(b)fluoranthene [11,14,15].

In landfill leachate samples, the highest concentration of benzene and nonane was determined; it was in excess of 190 mg L^{-1} in the case of benzene and in excess of 180 mg L^{-1} in the case of the other micro-pollutant. The lowest concentration noted for undecane was below 80 mg L^{-1}. The sum of volatile petroleum-derived compounds varied from 204.52 to 661.00 mg L^{-1} (Fig. 9). The most frequently noted sum varied from 300.00 to 400.00 mg L^{-1} (Fig. 11). Aromatic hydrocarbons share in the analysed volatile petroleum-derived compounds was in excess of 77% (Fig. 11), which could result from better solubility of those compounds in water. As far as volatile aromatic compounds are concerned, the highest concentration was featured by benzene. It constituted more than 30% of all determined volatile aromatic hydrocarbons in the tested landfill leachates.

4. Conclusions

It appears from performed landfill leachate tests that the leachates are subject to permanent transformation. Environmental conditions, physical and chemical composition of waste containing considerable amounts of organic and mineral substances, technical operations applied at the landfill site and existing sealing of the landfill substratum have big impact on those changes [10,11,15]. During landfill operation, concentrations of heavy metals decreased, which resulted from activities taken up for waste separation. Nevertheless, landfill leachates contained a number of organic micro-pollutants including, PAHs and volatile petroleum-derived hydrocarbons and heavy metals. Percentage of PAHs (16 PAHs) varied depending on the number of condensed rings. Compounds having three and four condensed aromatic rings made more than 60% of all PAHs. Approximately 30% share belonged to hydrocarbons with five and six condensed aromatic rings, which probably originates from their higher hydrophobic properties and, to a lesser extent, from biodegradation. It appears from research works conducted by various authors [11,15,20] that in anaerobic conditions, the number of condensed aromatic rings has no significant statistic impact on biodegradation speed. From among determined PAHs, the highest danger of underground water pollution comes from benzo(a)pyrene and then benzo(g,h,i)perylene and benzo(b)fluoranthene. Their percentage in the entire group 16 PAHs is, on average, 17.35%.

Increased concentration of benzene (>190 mg L^{-1}) and nonane (>180 mg L^{-1}) was also noted. The lowest concentration was determined for undecane (<80 mg L^{-1}), whereas the sum of volatile petroleum-derived compounds varied from 204.52 to 661.00 mg L^{-1}. The highest concentration in the volatile aromatic compounds group had benzene, showing clear carcinogenic properties. Its share in the tested landfill leachates was in excess of 30% of all determined volatile aromatic hydrocarbons.

The above has been confirmed by results of long-term research on the hazard of pollution of underground water with heavy metals and hydrocarbons, also carcinogenic ones, migrating from landfills together with rainfall infiltrating into underground water. Monitoring research of landfill leachates containing toxic groups of mineral and organic compounds can be taken into consideration in designing of special treatment stations for such leachates. They can also be used in forecasting of the phenomena of migration of such pollutants to underground water, particularly at all waste landfills without sealed landfill bottom.

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