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The influence of landfills located in different hydrogeological systems on Lithuanian groundwater quality

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Abstract The article presents data on influence of non-operational landfills located in different hydrogeological systems on Lithuania groundwater quality. The landfills are located in open, semi-open, semi-closed and closed hydrogeological systems. The prevailing values of total dissolved solids (TDS) in the most intensive groundwater pollution zones of landfills can be divided into four TDS levels: low (400–3000 mg/l), moderate (TDS 3000–5000 mg/l), high (TDS 5000–20000 mg/l) and very high (TDS 20000–32000 mg/l). Groundwater with a low TDS level prevails in the most intensive groundwater pollution zones of landfills in open and semi-open hydrogeological systems. Groundwater with high and very high TDS levels characterizes about 3% of landfills in open and semi-open hydrogeological systems. The landfills in semi-closed and closed hydrogeological systems are also characterised by groundwater with a low TDS level. No landfills with a very high TDS level were found in semi-closed and closed hydrogeological systems.

Keywords • *landfills* • *classification* • *hydrogeological systems* • *pollution* • *hierarchical cluster analysis*

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INTRODUCTION

Despite increasing environmental awareness and the amount of recycled or incinerated waste, waste disposal in landfills remains the most widely used method in many countries (Awaz 2015; Ashraf *et al*. 2013). A comparison of the years 2000 and 2015 across the European Union showed that the amount of municipal waste disposed of in landfills fell by 10–50% (Eurostat 2015). However, in countries such as Bulgaria, Croatia, Lithuania, Latvia and Turkey, more than 80% of the waste is still disposed of in landfills (Eurostat 2015). According inventory data of various countries in Europe there are about 150000 to 500000 non-operational and still operating municipal waste landfills with an average size of about 8000 m^2 and with a capacity of 30–50 billion m^3 of waste (Wagner, Raymond 2015; Jones *et al*. 2013; Krook *et al.* 2012). Although modern landfill technology has been used in the European Union and the United States since 1980, the number of old non-operational landfills without a leachate collection system comprises a large percentage of the above-mentioned landfills (Zhang *et al*. 2015).

Municipal waste landfills are characterized by a wide variety of waste. The landfill age and the physicochemical processes taking place inside the waste create a complex composition of leachate consisting of four pollutant groups: 1) dissolved organic matter; 2) inorganic macrocomponents; 3) heavy metals; 4) petroleum hydrocarbons (Bjerg *et al*. 2011; Christensen *et al*. 2001; Zhang *et al*. 2015, Kjeldsen *et al.* 2002; Longe, Enekwechi 2007; Barella *et al*. 2013). These four groups of pollutants are associated with research into groundwater circulation and its self-cleaning processes. The concentrations of chlorides, ammonia, nitrates and iron often exceed the permissible norms in permeable, sandy sediments (Lee *et al*. 2007; Şimşek *et al*. 2008; Assmuth, Strandberg 1993). The harmfulness of chemical components for the environment is much lower in low permeable sediments (Milosevic *et al*. 2013; Thomsen *et al*. 2012; Bellezoni *et al*. 2014; Assmuth, Strandberg 1993).

The studies analysing the natural self-cleaning processes of groundwater highlight the fact that pollutant migration depends primarily on the geological structure of the aquifer, which consists of high and low permeability sediments (Bjerg *et al*. 2011). Clayey rocks create geochemical barriers that block and transform many chemical components dissolved in water (Cozzarelli *et al*. 2011; Milosevic *et al*. 2013). Homogeneous sandy aqueous layers are characterized by rapid pollutant migration; the decrease in concentrations due to sorption processes is insignificant, as the most important process is that of dilution (Bagchi 1994; Christensen *et al*. 2001).

According results of our research the hydrogeological conditions, the volume of waste and the chemical composition of groundwater and leachate primarily determine the pollution migration in landfills. According to the characteristics of selected items in different studies, we can conclude that landfills show a wide variety of the above-mentioned indicators. Nevertheless, we did not find any studies that investigated the range of values of parameters determining pollution migration; in addition, there is virtually no research dealing with the problem of typifying these indicators. The classification of landfills could be particularly useful in the analysis of pollution migration. It has been observed that, if a comparative analysis of pollution migration is carried out in landfills located in different hydrogeological conditions, landfills with different characteristics are selected for research. However, it is obvious that the results are more reliable and accurate if the initial data have comparable or identical values. Classification requires a large representative and reliable sample of the original data information, but it is also supposed to facilitate the choice of comparable landfills for the assessment of the patterns of landfill pollution migration in different hydrogeological conditions. Another important aspect of classification is that data can be described qualitatively based on its specific features. Assessing the position of the waste volume size or pollution risk level of a specific landfill in the sample of values of the above-mentioned indicators specific to the landfills is also important from a practical perspective.

This research aimed at classifying non-operational landfills in terms of the indicators of pollution migration and establishing the possibilities of pollution migration in landfills located in different hydrogeological systems. The landfill classification was conducted on the basis of: 1) hydrogeological systems on which the landfills are located, 2) the prevailing TDS level in the most intensive groundwater pollution zone.

The landfill impact area in general can be divided into three hydrochemical zones based on the different levels of landfill impact: the most intensive groundwater pollution zone, the polluted groundwater zone and the least polluted groundwater zone (Brun *et al*. 2002). The most intensive groundwater pollution zone practically coincides with the waste-covered zone or is closest to this zone. In the case where groundwater is influenced by landfill, the hydrochemical composition of the groundwater in this zone is most affected by the landfill leachate, and is characterized by the highest concentration levels of total dissolved solids, organic matter and heavy metals (Bjerg *et al*. 2011; Christensen *et al*. 2001). Therefore this pollution zone has been selected as a suitable object for assessment of the pollution risk level of landfills.

MATERIAL AND METHODS

Study area and sites

923 non-operational municipal waste landfills located across the territory of Lithuania were selected for this research. (Fig. 1). The locations of non-operational municipal waste landfills were identified according to coordinates that, together with other landfill inventory data, have been collected by the Lithuanian Geological Survey since 1998 (Lithuanian Geological Survey 2015). The assessment of the groundwater quality of the most intensive pollution zone was conducted in 142 closed landfills. The prevailing types of hydrogeological systems were identified in 923 landfills. Analysed landfills are installed without the protective diaphragm and other modern security measures.

The Lithuanian territory is dominated by sandy and clayey Quaternary sedimentary covers of glacial origin, whose thickness varies from 10 to 100–200 m. Under the Quaternary sediment lie sandy, clayey and carbonate Pre-Quartenary sediments: in the southeastern part of Lithuania they lie in the depth of 250–500 m and in the western part of Lithuania in the depth of 2000–3000 m. The thickness of the active groundwater exchange zone is 200–300 m (Grigelis *et al*. 1994).

The area investigated was limited to the groundwater aquifer that is situated in the upper part of the sedimentary cover (till 25 metres deep) and is particularly sensitive to surface pollution. Groundwater can be found in sandy and gravel, alluvial, fluvioglacial deposits with bordering clayey layers. The hydraulic conductivity of sandy and gravel sediments reaches 100–300 m/d. In the great part of Lithuania the groundwater table is about at 3–5 meters depth, in southern and eastern Lithuania--about at 5–10 meters depth (Juodkazis 2003).

The vulnerability of the active water exchange zone through anthropogenic pollution across the ter-

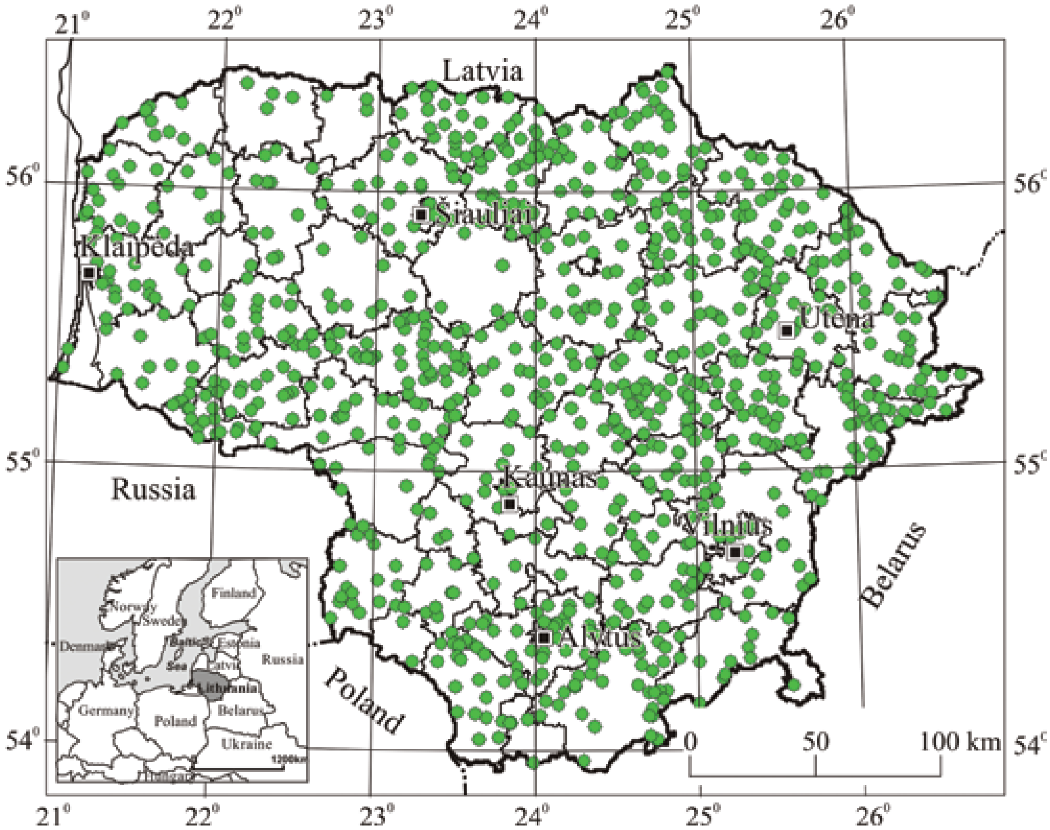

Fig. 1 Situational map of non-operational municipal waste landfills

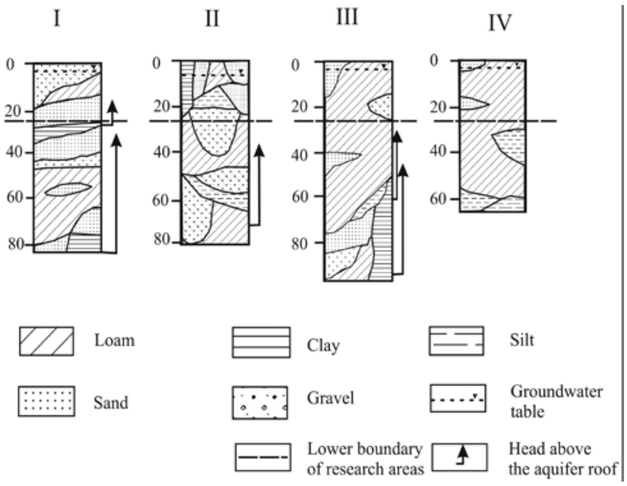

Fig. 2 Types of hydrogeological systems: I – open, simple filtration; II – semi-open, complex filtration; III – semi-closed, complex filtration; IV – closed, highly complex filtration. Hydrogeological profiles by Baltrūnas et al. 1998

ritory of Lithuania is influenced by the different geological and hydrogeological characteristics such as isolation and natural filtration properties in the upper part of the sedimentary column. The characteristics depend on the ratio between sandy and clayey layers. On the basis of these characteristics the Lithuanian territory is divided into four representative districts (Baltrūnas *et al.* 1998) (Fig. 2): I. Very high vulnerability areas: open hydrogeological systems. They consist of loose water-permeable rock. Homogeneous sandy and gravel aquifers prevail. The infiltration intensity is 4–6 l/s km^2. II. High vulnerability areas: semi-open hydrogeological systems. According to the data of hydrogeological profiles clay layers make more than 40–50% of this sedimentary column. They are characterised by heterogeneous layers. The hydraulic conductivity of clayey sediments exceeds 0.005 m/d. The infiltration intensity is 2–3 l/s km^2. III. Medium vulnerability areas: semi-closed hydrogeological systems. Clay layers make more than 60% of this sedimentary column. They are characterised by heterogeneous layers. The hydraulic conductivity of clayey sediments ranges from 0.001 to 0.005 m/d. The infiltration intensity is 1–2 l/s km^2. IV. Low vulnerability areas: closed hydrogeological systems. They consist of not less than 90% of clay and rock layers. These hydrogeological systems are characterised by homogeneous layers of low permeability. The hydraulic conductivity of clayey sediments does not exceed 0.001 m/d. The infiltration intensity does not exceed 1 l/s km^2 (Baltrūnas *et al.* 1998).

The lithology of prevailing sediments in hydrogeological systems determines their inherent filtration properties: the increasing amount of clay particles decreases the potential of pollution filtration, i.e. pollutant filtration becomes more complex.

The types of hydrogeological systems where the landfills are located were established on the basis of

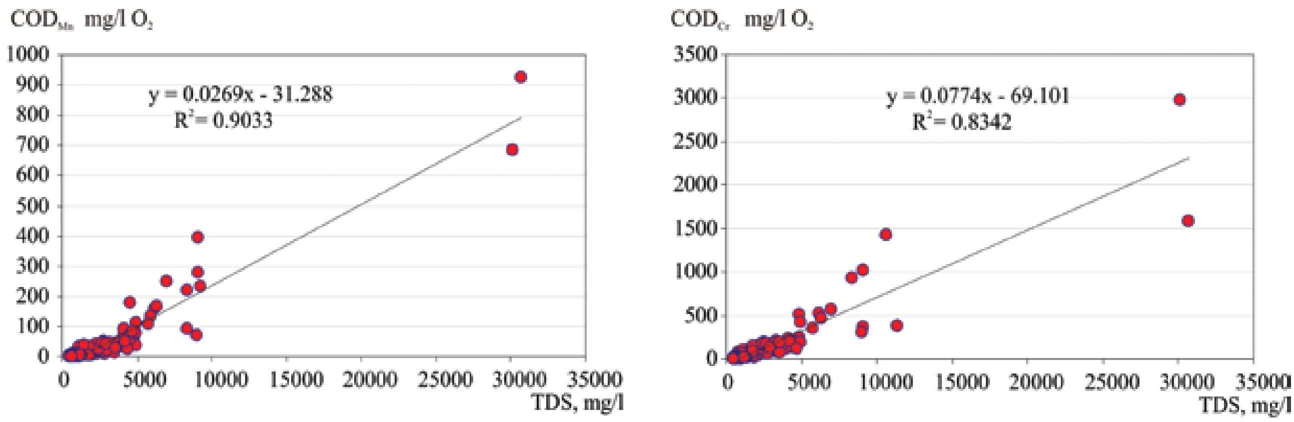

Fig. 3 The dependence of COD_{Mn} and COD_{Cr} on TDS in the most intensive groundwater pollution zone

an active groundwater circulation zone vulnerability map which was compiled by Baltrūnas *et al.* 1998. This map is composed and boundaries of vulnerability area are identified on the basis of Lithuanian hydrogeological, hydrological and geomorphologic conditions.

Identification of hydrochemical characteristics of the most intensive groundwater pollution zones

Groundwater samples were taken throughout 1997–2013 from groundwater observation wells. The depth and diameter of the wells reached till 25 m and 50–100 mm respectively. The well screens contained gravel filler whose thickness was > 20 mm. Well casings and filters were made of chemically inert polyvinyl chloride (PVC). The well mouth was surrounded by a mechanical impact-resistant cover which protected the well from accidental contamination. The sample wells were installed in the waste-covered area of the landfills or next to it in the direction of groundwater flow. While taking groundwater samples, the wells were cleaned using special bailers: in this way, 10 volumes of groundwater that the well could contain were pumped out. Before immersion in the next borehole, the bailer was washed using clean water. The samples were transferred into vessels that had been prepared in a laboratory and preserved cold and dark transported to the laboratory within 24 hours (LST ISO 5667-11:1998[1]).

As mentioned in the Introduction, landfill pollution consists of four groups of pollutants. Examination of the chemical composition of the most intensive groundwater pollution zones of the Lithuanian municipal waste landfills showed that the concentration levels of inorganic macrocomponents and dissolved organic matter have significantly increased. Similarly, they considerably exceed the natural background and limit concentrations. Concentrations of TDS and dissolved organic matter, i.e. COD_{Mn} and $COD_{Cr,}$ characterise those components. A good relation between these variables (Fig. 3) allows choosing one of them to identify the level of pollution. Having the largest sample, TDS was selected for the purposes of the identification of the pollution level.

For hydrochemical characterization of the most intensive groundwater pollution zone, diagrams of the distribution of total mineralization values in the landfill wells were made, and analyses with the maximum values of total mineralization have been selected. 1–3 observation wells were set up in individual landfills in the most intensive groundwater pollution zone. Each landfill and observation well produced from 1 to 9 samples. In total, 320 groundwater samples were taken from 193 observation wells. In the next stage, mean concentrations of the chemical components of selected analyses have been calculated. Oxidation-reduction potential (Eh), pH, CO_2, total dissolved solids (TDS), Cl^-, SO_4^{2-}, HCO_3^-, Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, NO_3^-, NO_2^-, COD_{Cr}, COD_{Mn}, Pb, Cd, Ni, Cr, Zn chemical components have been statistically assessed. Eh and pH are measured directly in the borehole. The results were used to classify the prevailing average TDS values in the most intensive groundwater pollution zones and assess the chemical damage done to groundwater based on the TDS classifications groups.

The reliability of each chemical analysis was assessed using the principle of electrical neutrality (Eq. 1). The results of the chemical analysis were considered reliable if the error did not exceed 5% (Apello, Postma 2005).

$$E.B.(\%) = \frac{\sum cations - \sum anions}{\sum cations + \sum anions} \times 100\% \quad (1)$$

where E.B. is electrical balance, %; Σcations and Σanions is the sum of cations and anions respectively, expressed in mg equivalents.

[1] LST ISO 5667-11:1998. Water quality. Sampling. Part 11: Guidance on sampling of ground waters. [In Lithuanian].

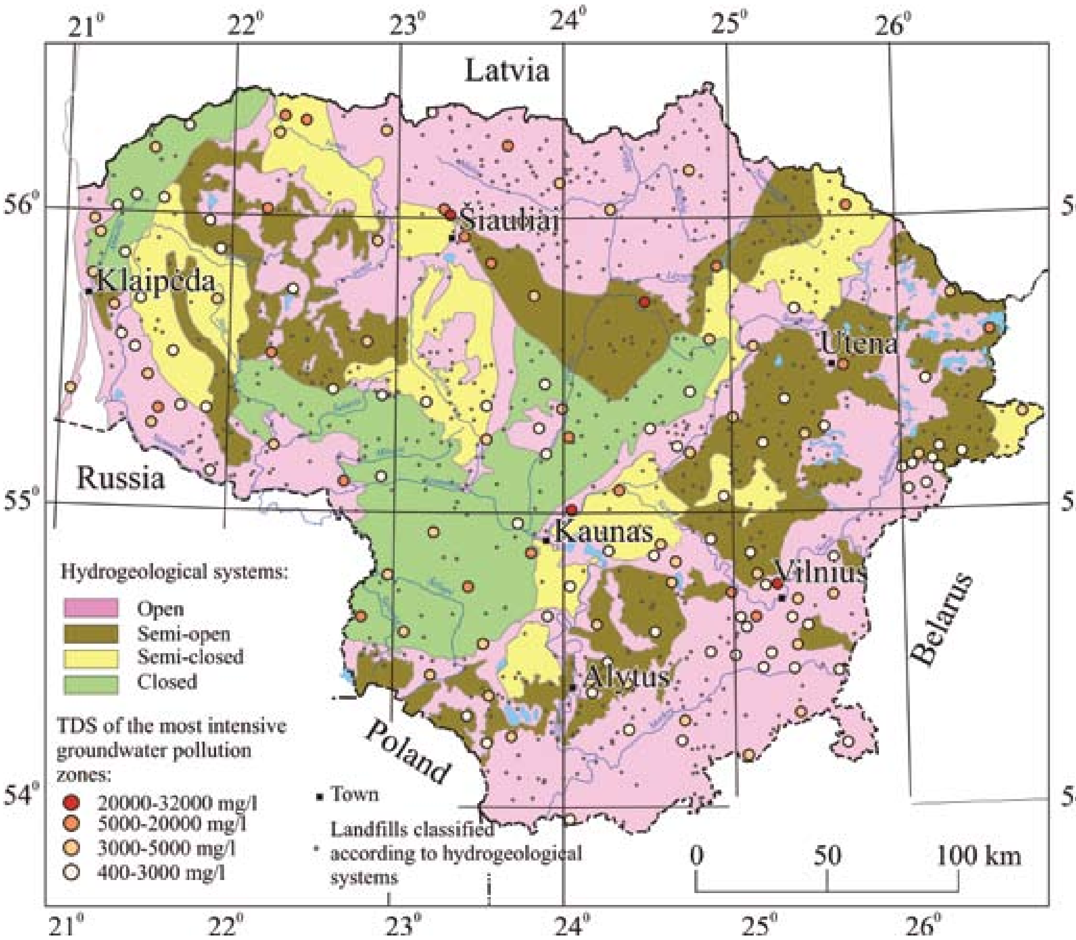

Fig. 4 Classification map of Lithuanian municipal waste landfills. Hydrogeological systems boundaries by Baltrūnas *et al*. 1998.

Classification of average TDS values and assessment of chemical damage of identified groundwater classifications groups

The hierarchical cluster analysis algorithm was used to classify the average TDS values of the most intensive groundwater pollution zones. Hierarchical cluster analysis is a reliable and frequently used method for the synthesis of hydrochemical data (Singh *et al*. 2008; Thakur *et al*. 2015; Vasanthavigar *et al*. 2013). The Euclidean metric method was used to assess the similarities between the objects, (Eq. 2). Ward's method was used to assess similarity of clusters (Eq. 3). Cluster analysis was performed using a Microsoft-Excel @ add-in module XLSTAT 2015.1.01.

$$D(X,Y)=\sqrt{\sum_{i=1}^{n}(x_i-y_i)^2} \quad (2),$$

$$d(U,V)=\left\|\overline{U}-\overline{V}\right\|^2/(1/n_U+1/n_v) \quad (3)$$

where x_i represents the value of attribute *i* of object *X*; y_i is the value of attribute i of object *Y*; *n* represents the number of measured items; *U, V* are clusters (Čekanavičius, Murauskas 2002).

The chemical damage of groundwater in terms of TDS classification groups were assessed by calculating the concentrations of the above-mentioned chemical components in relation to the size of: 1) limit concentrations of chemical components specified in environmental normative documents in the groundwater discharged into the natural environment in the area of low vulnerability to pollution (The Order of the Minister of Environment of the Republic of Lithuania 2007[2], 2008[3]); 2) background concentration values of chemical components in the groundwater in sandy (f III) and clayey (g III) sediments (Diliūnas *et al.* 2004). The indices of aquifers used according to the hydrogeological stratification in Lithuania are as follows: Upper Pleistocene fluvioglacial sediments (f III), Upper Pleistocene glacial sediments (g III) (Description of Lithuanian Quaternary stratigraphy scheme 2009[4]). The prevailing chemical type of

[2] The order of the Minister of Environment of the Republic of Lithuania, 2007. Wastewater management regulation No. D1-515, issued on 8 October, 2007. *State News* 110–4522, 1–15. [In Lithuanian]

[3] The order of the Minister of Environment of the Republic of Lithuania, 2008. On approval of environmental requirements to the management of chemically-polluted areas No. D1-230, issued on 30 April, 2008. *State News* 53–1987, 1–17. [In Lithuanian].

[4] Lithuanian Quaternary Stratigraphy Chart, 2009. Lithuanian Geological Survey under the Ministry of the Environment, Order No. 1-86, 17 June 2009. *Valstybės žinios 74-3055*, 1–5 [In Lithuanian].

groundwater was established using the Piper diagram. The water class expression included only those ions whose percentages exceeded 20% of the equivalent value (Appelo, Postma 2005).

RESULTS

Landfills distribution in hydrogeological systems

The locations of Lithuanian municipal waste landfills on the active groundwater circulation zone vulnerability map (Baltrūnas *et al*. 1998) facilitated the development of the classification map of Lithuanian municipal waste landfills (Fig. 4). Hydrogeological systems areas are marked using colours. Symbols of different colours denote the landfills and the TDS concentration level of the most intensive groundwater pollution zone. Chosen information display method allows map as clearly as possible to read.

Landfills of a varying pollution size are more or less equally spread across the territory of Lithuania. However, their distribution in different hydrogeological systems is different. It was discovered that the investigated landfills are located in four hydrogeological systems: 52.2% of the landfills are located in open hydrogeological systems. Semi-open and semi-closed hydrogeological systems comprise 26.4% and 10.3% of the landfills respectively; 11% of the landfills are located in closed hydrogeological systems (Fig. 4).

General hydrochemistry of intensive pollution groundwater zones

The results show that the coefficient of variation (CV) values of the majority of chemical components range from 1.3 to 4.5 and characterize the significant instability of established concentrations (Table 1, Fig. 5). First of all, this is related to significant differences in the concentrations of organic substances in groundwater and carbonate equilibrium processes. In cases of large quantities of organic matter, organic matter destruction takes place, emitting CO_2. Carbon dioxide combines with water to form carbonic acid, which dissolves carbonate (Fetter 1999). This results in higher alkaline element concentrations and an alkaline groundwater. The above-mentioned elements suggest that landfill pollution with organic matter is widely varying. The differences in means and medians of chemical components characterize significant differences in hydrochemical situations and the ex-

Table 1 Statistical indicators of average concentrations of prevailing chemical components in the most intensive groundwater pollution zones

Chemical components	Units	Threshold values	Natural background fIII/gIII[3)]	Number of landfills	Statistical indicators of average concentrations of chemical components							
					Minimum	Maximum	25% Q1	75% Q3	Median	Mean	Std.dev	CV
pH	pH units	6,5-8,8[1)]	7.54/6.94	138	6.1	8.3	7.0	7.4	7.3	7.3	0.3	0.0
Eh	mV		245/205	13	28.0	345.0	122.8	197.3	174.8	176.8	93.4	0.5
CO_2	mg/l			97	12.0	3649.5	68.0	233.8	100.0	225.8	403.4	1.8
TDS	mg/l	2000[1)]	378/563	142	453.8	30700.0	1121.4	3736.0	2046.5	3385.1	4437.5	1.6
Cl^-	mg/l	500[2)]	14.8/23	142	2.3	3954.8	29.4	548.8	118.5	436.6	716.2	2.8
SO_4^{2-}	mg/l	1000[2)]	24.8/31.5	142	2.3	2157.0	32.5	229.3	104.3	226.3	375.5	2.8
HCO_3^-	mg/l		210/375	142	231.0	17680.0	633.9	1805.8	985.5	1763.6	2469.1	1.3
Ca^{2+}	mg/l		60.9/69	142	24.9	944.0	148.3	308.5	206.5	244.6	139.2	4.5
Mg^{2+}	mg/l		19.6/34.8	142	6.7	438.0	35.8	119.0	65.8	86.8	70.4	1.4
Na^+	mg/l		7/17.4	142	2.6	3011.5	23.1	315.9	81.8	276.5	490.9	0.6
K^+	mg/l		1.6/4.1	142	1.1	2759.0	13.6	177.3	55.0	205.2	426.3	0.8
NH_4^-	mg/l	6.43[1)]	0.14/0.88	142	0.01	4333.0	0.4	115.1	9.0	191.4	539.3	1.8
NO_3^-	mg/l	100[1)]	2.69/2.77	140	0.01	2753.0	0.9	112.8	10.9	120.0	341.0	2.1
NO_2^-	mg/l	1.5[1)]	0.03/0.03	138	0.002	213.0	0.01	2.4	0.1	5.3	23.7	1.3
COD_{Mn}	mg/l O_2		2.58/3.05	138	0.5	2500.0	8.2	77.5	23.3	118.3	313.3	2.6
COD_{Cr}	mg/l O_2	125[1)]		113	2.4	7365.0	25.7	303.3	133.5	431.8	1069.4	2.5
Pb	mg/l	0.075[2)]	0.003/0.003	114	0.001	0.72	0.003	0.08	0.01	0.06	0.12	1.9
Cd	mg/l	0.006[2)]	0.0005/0.001	102	0.0001	0.04	0.0003	0.0003	0.0003	0.0015	0.01	3.4
Ni	mg/l	0.1[2)]	0.003/0.01	115	0.001	0.99	0.01	0.05	0.02	0.07	0.15	2.0
Cr	mg/l	0.1[2)]	0.007/0.012	88	0.001	0.63	0.003	0.02	0.01	0.04	0.11	2.4
Zn	mg/l	1[2)]	0.031/0.06	108	0.001	7.80	0.03	0.07	0.04	0.20	0.81	2.6

Chemical components: TDS–total dissolved solids, COD_{Mn}–permanganate oxidation, COD_{Cr}–dichromate oxidation; Std. dev–standard deviation; CV–variation coefficient. Legislative acts: [1)] The order of the Minister of Environment of the Republic of Lithuania, 2007. Wastewater management regulation No. D1-515, issued on 8 October, 2007. *State News* 110–4522, 1–15. [In Lithuanian]. [2)] The order of the Minister of Environment of the Republic of Lithuania, 2008. On approval of environmental requirements to the management of chemically-polluted areas No. D1-230, issued on 30 April, 2008. *State News* 53–1987, 1–17. [In Lithuanian]. [3)] Natural background by Diliūnas *et al*. 2004.

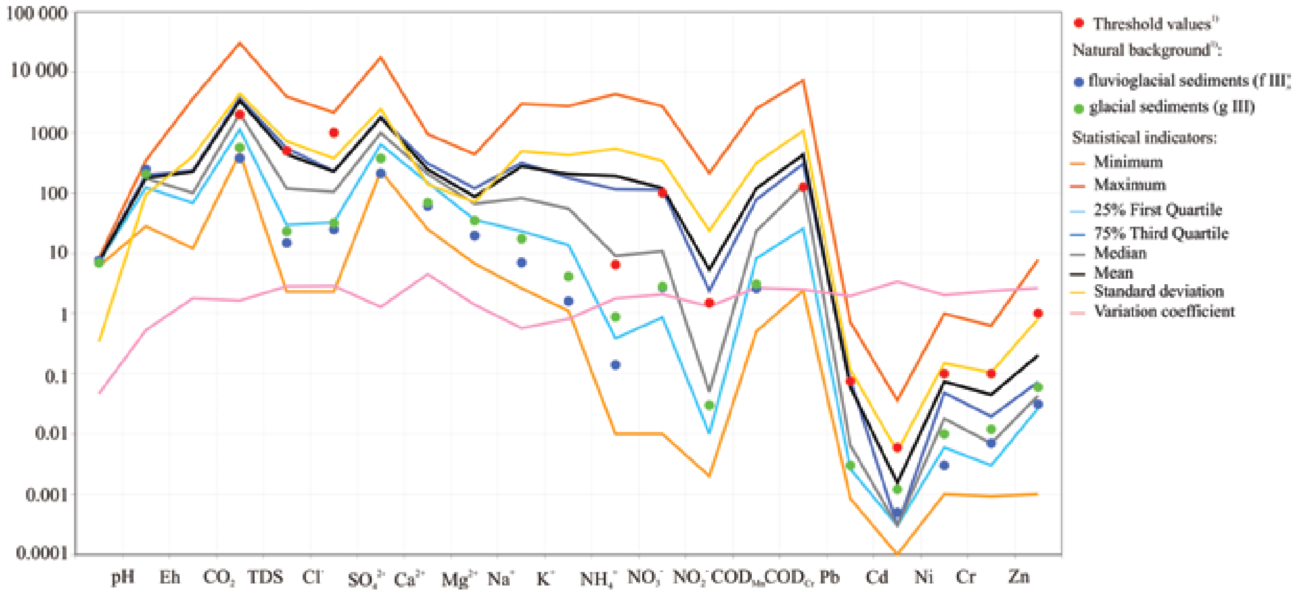

Fig. 5 Descriptive statistics of the chemical components of most intensive groundwater pollution zones (all parameters are measured in mg/l, except Eh (mV), pH (pH units), COD_{Mn} (mg/l O_2), COD_{Cr} (mg/l O_2). [1)] Citation of Legislative acts and Natural background in Table 1

tremes in the sample. The largest differences among these components are typical to TDS, Na^+, K^+, NH_4^+, NO_3^-, Cl^-, SO_4^{2-}, HCO_3^- and COD (Table 1, Fig. 5).

Average TDS values and their classification groups

The minimum and maximum average TDS concentration is 453.8 mg/l and 30700 mg/l respectively, which means that the amplitude of values is 30426 mg/l (Fig. 6). The calculated quartiles of 25% and 75% amounted to 1121.4 mg/l and 3736.0 mg/l respectively, the median and mean amounted to 2046.5 mg/l and 3379.6 mg/l respectively (Table 1).

The cluster analysis identified six clusters (Fig. 7). The landfills that are part of clusters C1 and C4 have very high (>20000 mg/l) average TDS concentration values in the most intensive groundwater pollution zones (Table 2). The average TDS concentration values of these clusters are about three times higher than those of the landfills in cluster C2 and about 15 times higher than those of the landfills in clusters C3, C5 and C6. About 2% of the landfills are characterized by maximum concentration values of TDS in the most intensive groundwater pollution zones.

The landfills in cluster C2 have average TDS concentration values in the most intensive groundwater pollution zones ranging from 5000 to 11000 mg/l (Table 2). Such values are characteristic of 13% of the investigated landfills. The landfills of cluster C3 show average TDS concentration values in the most inten-

Table 2 Statistical summary of average TDS concentration values in clusters

Cluster	Number of landfills	TDS concentration values (mg/l)			
		Minimum	Maximum	Mean	Median
C6	62	454	1541	1025	1048
C5	35	1621	2941	2319	2294
C3	23	3141	5000	4037	3975
C2	19	5668	10615	8318	8733
C4	1	23450	23450	23450	23450
C1	2	30151	30700	30426	30426

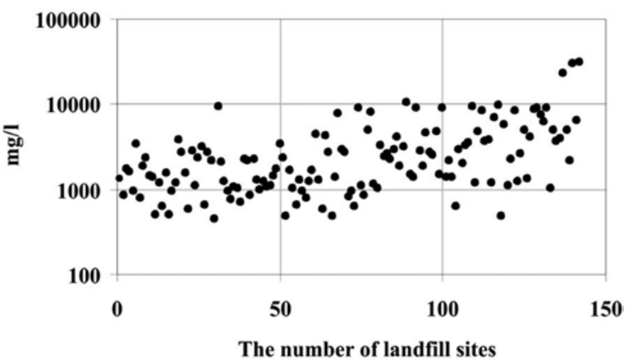

Fig. 6 The distribution of average TDS concentration values in the most intensive groundwater pollution zones in 142 landfill sites

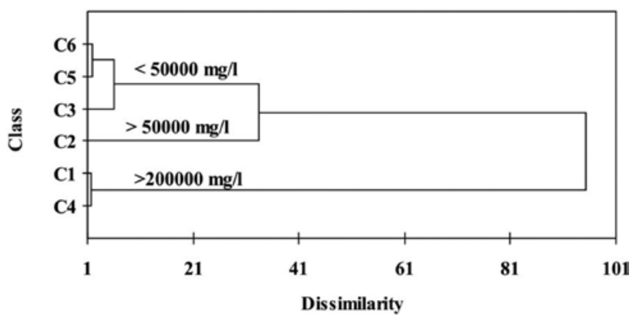

Fig. 7 The dendrogram of average TDS concentration values typical of most intensive groundwater pollution zones

sive groundwater pollution zones that range between 3000 and 5000 mg/l. 16% of the investigated landfills are part of this cluster. The landfills that are part of clusters C5 and C6 are characterized by the lowest average TDS concentration values, i.e. from 1700 to 3000 mg/l in cluster C5 and from 6400 to 1600 mg/l in cluster C6 (Table 2). These clusters cover 25% and 44% of the landfills respectively.

Different types of TDS concentration values classification can be found in normative acts. The concentration limits of groundwater that is clean, suitable for drinking and household needs, and safe for the environment range from <200 to 3000 mg/l. The following limit concentration levels, i.e. >1000 mg/l, >1500 mg/l, >2000 mg/l, >3000 mg/l, >10000 mg/l, are defined as unsuitable for use and dangerous for the natural environment (FEPA 1991; WHO 2004; Sen 2014; Hiscock, Bense 2014). The TDS values of background concentration vary from 220 to 680 mg/l in different genetic types of aquifer (Diliūnas *et al.* 2004).

Clusters C6 and C5 are characterized by TDS concentration values in the most intensive groundwater pollution zones that are lower than 1600 mg/l and 3000 mg/l respectively. Taking into account the classifications indicated in different normative acts and the background concentration values typical of different genetic types, we can conclude that the groundwater of the landfills belonging to these clusters is the least dangerous for the natural environment. As a result, the clusters can be combined into Group I (low mineralisation), where average TDS concentration values range from 400 to 3000 mg/l (Table 2, Figs. 7, 8). The concentration values of TDS typical of the landfills in clusters C4 and C1 are higher than 10000 mg/l and mostly exceed the natural background typical of groundwater (Table 2, Fig. 7, 8). This groundwater is therefore dangerous for the environment and human health. In view of this, clusters C4 and C1 can also be combined into Group IV (very high mineralisation), whose average TDS concentration values range from 20000 to 32000 mg/l (Table 2, Figs. 7, 8). The concentration values of TDS typical of the landfills in clusters C2 and C3 occupy an intermediate position between clean, safe, and highly mineralized, harmful contaminated groundwater. Thus, cluster C3 is characterized by TDS concentration, whose approximated values vary from 3000 to 5000 mg/l (Group II: moderate mineralisation). Meanwhile, cluster C2 is characterized by high TDS concentration, whose approximated values vary from 5000 to 20 000 mg/l (Group III: high mineralisation) (Table 2, Figs. 7, 8).

Table 3 Mean concentrations of the chemical components of TDS classification groups

Chemical components	IV (n=3)		III (n=20)				II (n=22)				I (n=97)			
	O	SO	O	SO	SC	C	O	SO	SC	C	O	SO	SC	C
pH	8.0	8.3	7.6	7.6	7.3	7.5	7.2	7.0	7.0	7.2	7.3	7.2	7.3	7.0
Eh			147	75				193			176	248	272	
CO_2	667		101	423	495	158	238	412	309	234	87	85	85	147
TDS	30426	23450	8977	8334	9274	6929	3621	3939	4664	3866	1343	1125	1384	1014
Cl^-	3881	2178	1211	1489	530	585	354	590	647	899	40	70	95	53
SO_4^{2-}	141	1697	47	301	17	596	191	281	42	93	69	90	160	104
HCO_3^-	17411	12812	4291	2860	6096	3135	1930	1669	2674	2260	756	591	675	659
Ca^{2+}	302	439	220	358	491	473	277	387	332	310	179	219	210	187
Mg^{2+}	134	105	154	174	146	188	119	161	104	138	42	52	57	38
Na^+	2628	3012	1026	664	541	680	311	293	277	459	43	28	58	17
K^+	2142	2087	716	77	380	242	205	168	76	413	34	14	23	7
NH_4^+	3806	1958	493	456	957	530	139	42	473	139	4	1	1	1
NO_3^-	183	1199	1	142	12	278	16	0.4	165	1	9	15	15	1
NO_2^-	20	2	2	1	4	7	2	1	3	1	0.01	0.01	0.4	0.0
COD_{Mn}	1054	189	305	151	247	148	72	132	82	36	14	10	14	8
COD_{Cr}	3017	355	843	416	704	486	207	160	497	409	45	39	37	42
Pb	0.2	0.1	0.1	0.01	0.002	0.11	0.01	0.003	0.041	0.047	0.01	0.01	0.003	0.001
Cd	0.02		0.0004	0.0003	0.0003	0.00	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003	0.0003
Ni	0.6	0.3	0.2	0.02	0.2	0.1	0.04	0.023	0.005	0.03	0.01	0.01	0.02	0.01
Cr	0.3	0.04	0.1	0.1	0.2	0.2	0.01	0.005	0.001		0.01	0.00	0.01	0.01
Zn		0.1	0.04	0.1	0.04	0.01	0.1	0.1	0.01	0.1	0.04	0.1	0.03	0.1

TDS classification groups: Group IV: very high mineralisation, Group III: high mineralisation, Group II: moderate mineralisation, Group I: low mineralisation; hydrogeological systems: O–open, SO–semi-open, SC–semi-closed, C–closed; chemical components: TDS–total dissolved solids, COD_{Mn}–permanganate oxidation; COD_{Cr}–dichromate oxidation; n-number of landfill sites; all parameters are measured in mg/l, except Eh (mV), pH (pH units), COD_{Mn} (mg/l O_2), COD_{Cr} (mg/l O_2).

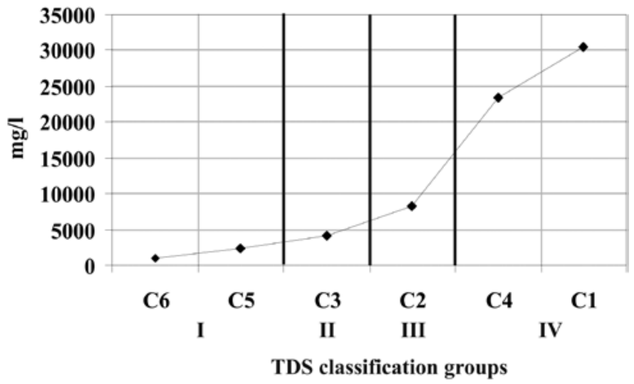

Fig. 8 Mean concentrations of total dissolved solids in the clusters determined from hierarchical cluster analysis

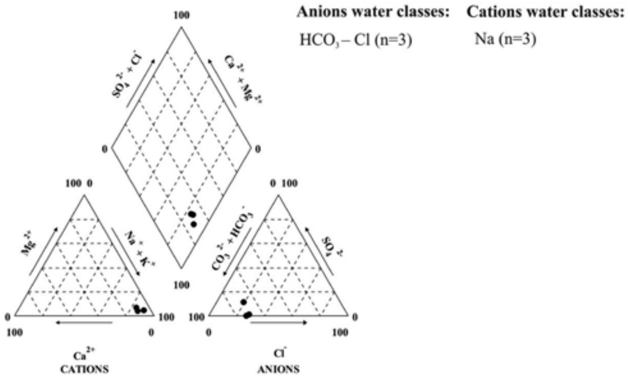

Fig. 9 Piper diagram characterises groundwater chemical types of landfills belonging to TDS classification Group IV

The chemical damage of groundwater in terms of TDS classification groups

The classification map of landfills in Lithuania shows that landfills with different groundwater chemical demage are located in hydrogeological systems of different vulnerability (Fig. 4, Table 3).

Very highly mineralised groundwater (TDS 20000–32000 mg/l, classification Group IV). This cluster group comprises about 2% of the investigated landfill sites. The landfills are located in open and semi-open hydrogeological systems (see Table 3, Fig. 4). The TDS concentration values in the most intensive groundwater pollution zone of this group exceed the natural background values and environmental safety limit concentration by factors of 80 and about 15 respectively. Water type HCO_3–Cl–Na prevails (Fig. 9). This is comparable to the results of water classes and types found in other studies dealing with the groundwater zones most affected by leachate. The most common water types referred to in other studies include Na–Cl–HCO_3, Na–HCO_3–Cl, Na–HCO_3 (Şimşek *et al.* 2008), Na–Cl, Mg–HCO_3, Cl–SO_4 (Han *et al.* 2014). The concentration of chloride and sodium in clean groundwater is about 10 mg/l; therefore, high concentration values of these chemical components are linked to anthropogenic pollution and very high TDS concentration in groundwater (Berkowitz *et al.* 2008).

A weak alkaline environment prevails in the most intensive groundwater pollution zones; pH ranges from 8.0 to 8.3 pH units. The data for the values of the oxidation-reduction potential is not available. From the high organic material (COD_{Cr} and COD_{Mn}) and high carbon dioxide concentrations, we can come to a conclusion about low oxygen content and intensive organic material destruction processes. Values of chemical oxygen demand (COD_{Cr}) that exceed the limits by a factor of 8–24, and COD_{Mn} values that exceed the natural background by a factor of about 341 prevail. Due to the low oxygen concentration, conditions conducive for the reproduction of ammonifying and denitrifying bacteria prevail. Prevailing ammonium concentration values exceed the limits and the natural background by factors of 407–592 and 23419 respectively. Nitrate concentration values exceed the natural background and limit concentration by factors of about 136 and 2–8 respectively.

Prevailing chloride concentration values exceed the limit and the natural groundwater background by factors of 257 and about 6–8 respectively. Sulphates exceed the natural groundwater background concentration values by up to 10 times. The ratio between the limit concentration values and the factual concentrations of sulphates is insignificant. Prevailing concentration values of hydrocarbonates, calcium, magnesium, sodium and potassium exceed the natural groundwater background by factors of about 82, 7, 5, 418 and 1304 respectively. The concentration values of lead and cadmium, nickel and chromium exceed the natural groundwater background by factors of about 40, 160 and 30. The ratio between the prevailing factual concentrations and limit concentration is about 2–6 times.

Highly mineralised groundwater (TDS 5000–20000 mg/ l, classification Group III). This group includes about 14% of all the investigated landfill sites. Landfills are located in open, semi-open, semi-closed and closed hydrogeological systems (Table 3, Fig. 4). The TDS concentration values in the most intensive groundwater pollution zone of this group of landfill exceed the natural background and limit concentration by factors of 22 (f III)/16 (g III) and about 4 respectively. Similarly to very highly mineralised groundwater, water class HCO_3–Cl based on the anion composition is most common (Fig. 10). According to the predominant cations, the most typical were those water classes where 20% of the percentage equivalent was exceeded by Na^+, Ca^{2+}, K^+, Mg^{2+} cations, which was not typical of very highly mineralised groundwater.

A neutral, weak alkaline environment prevails in the most intensive groundwater pollution zones; the pH ranges from 7.3 to 7.7 pH units. The oxidation-reduction potential is between 75 and 174.8 mV. Prevailing concentration values of CO_2 range between 97 and 615 mg/l and are about 5 times lower than in very highly mineralised groundwater. The values of these components suggest that both the concentration of organic material and the amount of carbon dioxide resulting from the destruction of organic material that increase the alkalinity of the groundwater are slightly lower than in very highly mineralised groundwater.

The prevailing concentration of COD_{Cr} exceeds the concentration limits by a factor of 7; permanganate index values exceed the background concentrations by factors of 114 in sandy sediments and 51 in clay sediments. Prevailing ammonium concentration values exceed the limits and the natural background by factors of 55–109 and 3519 (f III)/698 (g III) respectively. Nitrate concentration values exceed the natural background by a factor of about 27 (f III)/56 (g III). The ratio between the environmental concentration limits and the prevailing factual concentrations of nitrates is insignificant.

The prevailing concentration of chlorides exceeds the natural background and the concentration limits by factors of about 87 (f III)/25 (g III) and 2–3 respectively. The concentration values of sulphates exceed the natural background on average by a factor of 6 in sandy sediments, while the ratio size in clay sediments is insignificant. The ratio between the environmental concentration limits and the prevailing factual concentrations of sulphates is insignificant. The prevailing concentration values of hydrocarbonates, calcium, magnesium, sodium and potassium exceed the natural background by factors of about 18 (f III)/11 (g III), 4 (f III)/7 (g III), 8 (f III)/5 (g III), 107 (f III)/35 (g III) and 329 (f III)/91 (g III) respectively. The concentration values of lead, nickel and chromium exceed the natural background by factors of 8 (f III)/26 (g III), 48 (f III)/18 (g III) and 19 (f III)/17 (g III) respectively. The ratio between the actual concentration values of zinc and background concentrations is insignificant. Heavy metal concentrations exceeded the limits by a factor of about 2–3.

Moderately mineralised groundwater (TDS 3000–5000 mg/ l, classification Group II). This group includes about 15% of all the investigated landfill sites. The landfills are located in open, semi-open, semi-closed and closed hydrogeological systems (see Table 3, Fig. 4). The concentration values of total dissolved solids in the most intensive groundwater pollution zone of this group of landfills exceed the natural background and the concentration limits by factors of 10 (f III)/8 (g III) and about 2 respectively. Based on the anion composition, water class HCO_3–Cl and HCO_3 is the most common. According to the predominant cation composition, the most typical water classes are those where 20% of percentage equivalent is exceeded by Na^+, Ca^{2+}, Mg^{2+}, K^+ cations. The largest percentage equivalent part in the identified water classes consists of Na^+ and Ca^{2+} cations (Fig. 11).

A neutral environment prevails in the most intensive groundwater pollution zones; the pH ranges from 7.0 to 7.4 pH units. The oxidation-reduction potential is about 200 mV. The concentration values of CO_2 ranges between about 140 and 390 mg/l. The prevailing concentration of COD_{Cr} exceeds the limit by a factor of up to 2; permanganate index values exceed the background concentration by a factor of 28 (f III)/19 (g III). Prevailing ammonium concentration values exceed the natural background and concentration limits by factors of 714 (f III)/157 (g III) and 7–33 respectively. Nitrate concentration values exceed the natural background by about 3 times in sandy sediments; in clayey sediments the ratio size is insignificant. The ratio between the values of concentration limits and the prevailing factual concentrations of nitrates is insignificant. Nitrate concentration values exceed the limit by a factor of 2–5.

The prevailing concentration of chlorides exceeds the natural background and the limit concentration by factors of about 34 (f III)/39 (g III) and 1–2 respec-

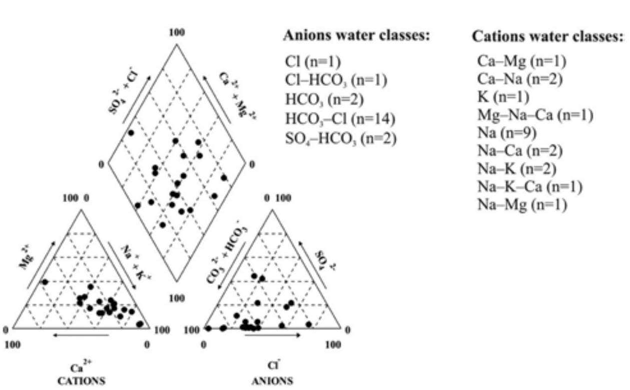

Fig. 10 Piper diagram characterises groundwater chemical types of landfills belonging to TDS classification Group III

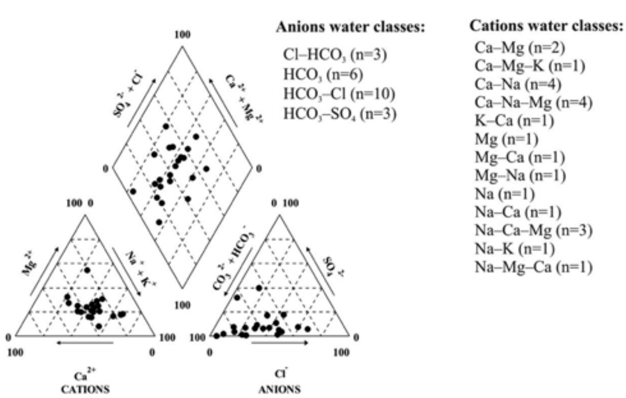

Fig. 11 Piper diagram characterises groundwater chemical types of landfills belonging to TDS classification Group II

tively. The concentration values of sulphates exceed the natural background approximately by a factor of 8 in sandy sediments, while the ratio size in clayey sediments is insignificant. The ratio between the background concentration values and the prevailing factual concentrations of sulphates is insignificant. The prevailing concentration values of hydrocarbonates, calcium, magnesium, sodium and potassium exceed the natural background by factors of about 9 (f III)/6 (g III), 5 (f III)/4 (g III), 6 (f III)/4 (g III), 42 (f III)/25 (g III) and 128 (f III)/20 (g III) respectively.

The concentration values of lead exceed the natural background by a factor of 2 (f III)/14 (g III). The concentration values of nickel and zinc exceed the natural background by factors of 8 and 17 respectively in sandy sediments. In clayey sediments, the ratio of the factual concentrations values and background concentrations of these chemical components is insignificant. The concentration values of cadmium and chromium are close to the natural background. The prevailing concentrations of heavy metals did not exceed the limit concentrations.

Low mineralised groundwater (TDS 400–3000 mg/ l, classification Group I). This group includes about 68% of all the investigated landfill sites. The landfills are located in open, semi-open, semi-closed and closed hydrogeological systems (see Table 3, Fig. 4). The TDS concentration values in the most intensive groundwater pollution zone of this group of landfill sites exceed the natural background by a factor of 3 (f III)/2 (g III). Based on the anion composition, water class HCO_3 is most common. According to the predominant cation composition, the most typical water classes are those where 20% of the percentage equivalent is composed of Ca^{2+} and Mg^{2+} cations (Fig. 12). Clean water is characterized by HCO_3–Ca–Mg type of water (Jamshidzadeh, Mirbagheri 2011); therefore, the hydrochemical composition of these landfills is similar to that of clean groundwater.

A neutral environment prevails in the most intensive groundwater pollution zones; the pH ranges from 7.0 to 7.4 pH units. The potential of the oxidation-reduction potential is about 350 mV. The concentration level of CO_2 and oxygen is 89 mg/l and up to 9 mg/l respectively. These parameters represent relatively good oxidative conditions in the most intensive groundwater pollution zones. This is corroborated by considerably smaller quantities of organic matter than in landfills with a higher pollution level.

The ratio between prevailing values of COD_{Cr} and background concentrations is insignificant. Permanganate index values exceed the background concentration by a factor of 5 (f III)/4 (g III). Prevailing ammonium concentration values in sandy sediments exceed the background concentrations by a factor of about 11, while the ratio in clayey sediments is insignificant. Prevailing NH_4 concentration values exceeds the limit by a factor of about 2–9. Nitrate concentrations values exceed the background and limit concentrations by factors of 4 (f III)/3 (g III) and 2–3 respectively.

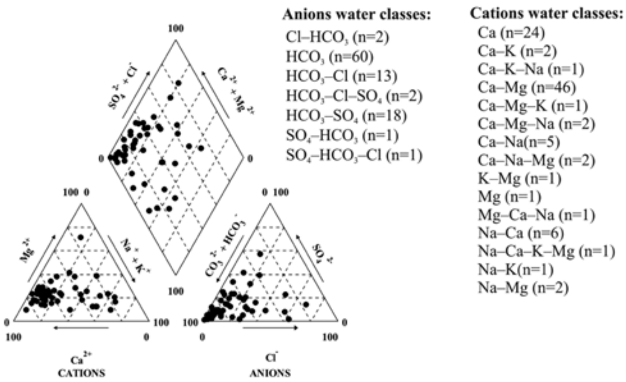

Fig. 12 Piper diagram characterises groundwater chemical types of landfills belonging to TDS classification Group I

The prevailing concentration values of chlorides and sulphates exceed the natural background by factors of only 4 (f III)/3 (g III) and 3 (f III)/4 (g III) respectively. The prevailing concentration values of hydrocarbonates, calcium, magnesium, sodium and potassium exceed the natural background by factors of about 4 (f III)/2 (g III), 3 (f III)/3 (g III), 2 (f III)/1 (g III), 6 (f III)/2 (g III) and 15 (f III)/3 (g III) respectively. The concentration values of heavy metals exceed only the natural background. The ratio between the concentration values of lead, cadmium and chromium and their background concentrations is not significant. The concentration values of nickel and zinc are significant in sandy sediments only. The above-mentioned concentration values exceed the background concentrations by factors of 4 and 14 respectively.

DISCUSSION

The classification of landfills and the classification map of landfills demonstrate the possibilities of pollution migration in landfills located in different hydrogeological systems. The study stated that the landfills are installed regardless of the vulnerability of the hydrogeological system. More than 70% of the landfills have been located in the open and semi-open systems with hydrogeological conditions conducive for pollution. The obtained results confirmed by of other studies, that focus hydrological conditions of landfills (Bagchi 1994; Assmuth, Strandberg 1992; Antonova *et al.* 2012; Lyngkilde, Christensen 1992; Kjeldsen 2002; Kehew, Passero 1990).

When evaluating the risk of groundwater pollution other authors also often rely on the TDS indicator – the concentration of total dissolved solids in water (El

Maghraby 2015; Nas, Berktay 2010). A considerable number of studies refer to TDS concentration values that are found in groundwater wells located close to landfill sites (ranging between 300 and 20000 mg/l) and TDS concentration values that are found in leachate (ranging between 2000 to 60000 mg/l) (El-Hames *et al*. 2013; Mor *et al*. 2005; Christensen *et al*. 2001). Therefore it can be said that during the research evaluated the average TDS concentration values of intensive pollution groundwater zones are reliable for pollution level identification. The TDS concentration values was measured in 142 landfills located in various hydrogeological systems, making it a representative research sample. The analysis of groundwater samples of TDS classification groups clearly shows that highly polluted groundwater with high concentration levels of TDS also contains higher concentration levels of inorganic macrocomponents, dissolved organic matter and heavy metals.

The classification map of landfills has revealed different distribution trends of landfills with varying degrees of pollution across hydrogeological systems. In about 70% of the landfills located in open hydrogeological systems systems, low mineralisation levels of groundwater prevail in the most intensive groundwater pollution zones, while moderately and highly mineralised groundwater prevails in 16% and 11% of the landfills respectively. Very highly mineralised groundwater can be found in about 3% of the landfills. Low mineralised groundwater in the most intensive groundwater pollution zones also prevails in the landfills installed in semi-open hydrogeological systems (~ 70% of landfills). Moderate and high mineralisation of groundwater characterises 13% and 13% of the landfills respectively. Very high mineralisation of groundwater was detected in 3% of the landfills. Semi-closed hydrogeological systems consist of ~ 71% landfills with a low mineralisation level of groundwater in the most intensive groundwater pollution zones. 12% and 18% of the landfills are characterised by moderate and high mineralisation levels of groundwater respectively. No landfills with prevailing very high levels of mineralisation were identified in this category. In the most intensive groundwater pollution zones of closed hydrogeological systems, ~ 53%, 20% and 26% of the landfills had a low, moderate and high level of mineralisation respectively. No landfills with a very high level of mineralisation were found.

CONCLUSIONS

Non-operational municipal waste landfills are located in open, semi-open, semi-closed and closed hydrogeological systems. The greatest number of landfills is located in open hydrogeological systems. Four TDS groups prevail in the most intensive groundwater pollution zones: low mineralisation (400–3000 mg/l), moderate mineralisation (3000–5000 mg/l), high mineralisation (5000–20000 mg/l) and very high mineralisation (20000–32000 mg/l). Low mineralised groundwater prevails.

Landfills installed in open and semi-open hydrogeological systems are characterised by low mineralised groundwater. The groundwater of this group is defined by concentrations of chemical components that are close to the natural background and do not exceed limit concentrations (except for nitrogen compounds); therefore, pollution migration is potentially unharmful. Landfills with high or very high levels of mineralisation in the most intensive groundwater pollution zones comprise the smallest percentage (~ 3%) of the landfills in open and semi-open hydrogeological systems. Highly and very highly mineralised groundwater contains chemical components that significantly exceed limit concentrations and the natural background and create favourable conditions for pollution migration in sandy sediments.

The groundwater in landfills installed in semi-closed and closed hydrogeological systems is characterised by a low mineralisation level. No landfills with a very high mineralisation level were found in this group. This means that clayey sediments are not conducive to pollution migration. This classification of landfills is particularly practical useful in the analysis of pollution migration in different hydrogeological systems.

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