## VILNIUS UNIVERSITY NATURE RESEARCH CENTRE

GINTARĖ SLAVINSKIENĖ

# GROUNDWATER QUALITY CHANGE THROUGHOUT THE POLLUTION FILTRATION PROCESS IN LITHUANIAN MUNICIPAL WASTE LANDFILLS

Summary of doctoral dissertation Physical sciences, Geology (05P)

Vilnius, 2016

This doctoral dissertation was written at Vilnius University in the Nature Research Centre at the Institute of Geology and Geography in 2010-2015.

**Scientific supervisor** – dr. Arūnas Jurevičius (Nature Research Centre at the Institute of Geology and Geography, Physical Sciences, Geology – 05P)

Scientific consultant – dr. Jurga Arustienė (Lithuanian Geological Survey, Physical Sciences, Geology – 05P)

## The dissertation will be defended at the Council of Geology of Vilnius University:

**Chairman** – dr. Kęstutis Jokšas (Nature Research Centre at the Institute of Geology and Geography, Physical Sciences, Geology – 05P).

### Members:

dr. Rimantė Zinkutė (Nature Research Centre at the Institute of Geology and Geography, Physical Sciences, Geology – 05P),

prof. habil. dr. Jonas Mažeika (Nature Research Centre at the Institute of Geology and Geography, Physical Sciences, Geology – 05P),

dr. Jonas Satkūnas (Lithuanian Geological Survey, Physical Sciences, Geology – 05P), dr. Rein Vaikmäe (Institute of Geology at Tallinn University of Technology, Physical Sciences, Geology – 05P).

The public defence of the dissertation is to be held in the meeting of the Council of Geology of Vilnius University at 3 p.m. on 27 May 2016 in the Nature Research Centre.

Address: Akademijos Str. 2, Vilnius, Lithuania.

The summary of the dissertation was sent out to relevant institutions on 28 April 2016.

The dissertation is available at the libraries of Vilnius University, Nature Research Centre and in the Website of Vilnius University: <u>www.vu.lt/lt/naujienos/ivykiu-kalendorius</u>

## VILNIAUS UNIVERSITETAS GAMTOS TYRIMŲ CENTRAS

GINTARĖ SLAVINSKIENĖ

## POŽEMINIO VANDENS KOKYBĖS KAITA LIETUVOS BUITINIŲ ATLIEKŲ SĄVARTYNŲ TERŠALŲ FILTRACIJOS PROCESE

Daktaro disertacijos santrauka Fiziniai mokslai, geologija (05 P)

Vilnius, 2016

Disertacija rengta 2010–2015 metais Gamtos tyrimų centro Geologijos ir geografijos institute.

**Mokslinis vadovas** – dr. Arūnas Jurevičius (Gamtos tyrimų centro Geologijos Geografijos institutas, fiziniai mokslai, geologija – 05P)

**Mokslinė konsultantė** – dr. Jurga Arustienė (Lietuvos geologijos tarnyba, fiziniai mokslai, geologija – 05P)

### Disertacija ginama Vilniaus universiteto Geologijos mokslo krypties taryboje:

**Pirmininkas** – dr. Kęstutis Jokšas (Gamtos tyrimų centro Geologijos Geografijos institutas, fiziniai mokslai, geologija – 05P).

#### Nariai:

dr. Rimantė Zinkutė (Gamtos tyrimų centro Geologijos Geografijos institutas, fiziniai mokslai, geologija – 05P),

prof. habil. dr. Jonas Mažeika (Gamtos tyrimų centro Geologijos Geografijos institutas, fiziniai mokslai, geologija – 05P),

dr. Jonas Satkūnas (Lietuvos geologijos tarnyba, fiziniai mokslai, geologija-05P),

dr. Rein Vaikmäe (Talino technologijos universiteto Geologijos institutas, fiziniai mokslai, geologija – 05P).

Disertacija bus ginama viešame Geologijos mokslo krypties posėdyje 2016 m. gegužės mėn. 27 d. 15 val., Gamtos tyrimų centre.

Adresas: Akademijos g. 2, Vilnius, Lietuva.

Disertacijos santrauka išsiuntinėta 2016 m. balandžio mėn. 28 d.

Su disertacija galima susipažinti Vilniaus universiteto, Gamtos tyrimų centro bibliotekose ir VU interneto svetainėje adresu: <u>www.vu.lt/lt/naujienos/ivykiu-kalendorius</u>

## **ABBREVIATIONS**

C-closed hydrogeological system

 $COD_{Mn}$  – permanganate oxidation

O – open hydrogeological system

SC – semi-closed hydrogeological system

SO – semi-open hydrogeological system

TDS - total dissolved solids

### **TERMS AND CONCEPTS**

- Landfill means a waste disposal site for the deposit of the waste onto or into land for safe storage and control of waste.
- Municipal waste means waste from households, as well as other waste which, because of its nature or composition, is similar to waste from household.
- Leachate means any liquid percolating through the deposited waste and emitted from or contained within a landfill.
- Classification indicators means a hydrogeological system, the volume of waste, and hydrochemical characteristics of the most intensive groundwater pollution zones.
- Typical landfill means a landfill which characterises the landfills installed in the same hydrogeological systems with similar (or equivalent) waste volumes and the hydrochemical characteristics of the most polluted groundwater.
- Hydrogeological system means water conductive and low-conductive upper unconfined and confined hydraulically related aquifers, which determine dispersion characteristics of surface pollutants under ground.

#### **INTRODUCTION**

**The relevance of the research.** The processes of sorption, crystallisation, dilution and cation exchange occurring in the rock-water system form conditions that geochemically prevent migration of pollutants. The forecasting of groundwater pollution is virtually impossible without the assessment of complex geochemical processes.

In groundwater, anions, cations, molecules can migrate in the form of simple ions or compounds. One chemical element may form a number of migration forms whose balance is sensitive to environmental changes (Diliūnas et al. 2009; Appelo, Postma 2005). In groundwater, these compounds of hydrochemical components have different thermodynamic parameters, which determine their distribution and migration potential (Appelo, Postma, 2005).

Both in Lithuania and all over the world, the lack of eco-awareness and economic reasons have contributed to the many landfills that were constructed without taking into account environmental requirements. Such landfills, which in fact are concentrated sources of pollution, pose a potential threat to groundwater safety, but at the same time are a favourable object of research in order to assess the patterns of groundwater self-cleaning processes.

At present, considerable research is being conducted into topics related to the problems of pollutant dispersion in the environs of landfills. Previous research emphasises the heterogeneity of landfills in terms of waste volume, heap size and leachate contamination. Nevertheless, there are hardly any studies focusing on the values and characterisation of the above-mentioned indicators that determine pollutant dispersion in landfills. It should also be noted that throughout literature the following pattern can be found in many cases: if a comparative analysis of pollution dispersion is performed in landfills located in different hydrogeological systems, landfills with different characteristics are selected as research objects. This may result in inaccurate patterns of pollution dispersion.

The thesis analyses the variety of indicators that determine pollution dispersion in landfills. Additionally, the analysis aims to characterise representative groups of these indicators and identify options for selecting representative landfills. A thermodynamic model facilitated the identification of migration forms of hydrochemical components in different groundwater pollution zones, which will in turn enable the assessment of patterns of pollution dispersion in landfills located in different hydrogeological systems. The results should be relevant to environmental protection and engineering professionals as well as scientists.

The thesis aims to classify non-operational municipal waste landfills. In addition, it seeks to assess the patterns of groundwater chemical composition changes during the filtration process in municipal waste landfills located in different hydrogeological systems.

#### The research tasks are to:

- 1. Identify representative types of hydrogeological systems, waste volume and intensive groundwater pollution zones of non-operational municipal waste landfills.
- 2. Draw a classification map of landfills and assess the selection possibilities of typical landfills that are located in different hydrogeological systems in order to analyse groundwater pollution dispersion.

- 3. To identify impact zones of different levels of typical landfills located in different hydrogeological systems on groundwater and to identify the distances within which the impact of the landfill on groundwater still remains.
- 4. To assess trends in changes of the migration forms of hydrochemical components and the groundwater saturation with minerals in the filtration of the polluted groundwater in the typical landfills located in different hydrogeological systems.

### The following theses are to be defended:

- 1. Landfill sites are located without taking into account the type of the hydrogeological system, which determine dispersion of pollutants into groundwater. Landfills are characterised by a great variety of waste volumes and the hydrochemical characteristics of pollutants dispersed in groundwater.
- 2. The classification map of landfills was drawn up to facilitate selection of landfills with similar volumes of waste and similar hydrochemical characteristics of the most intensive groundwater pollution zones in each identified hydrogeological system.
- 3. The more confined the layer of the aquifer, the shorter the distance, within which the impact of the landfill pollution on groundwater still remains.
- 4. With the decrease of the landfill pollution dispersion in groundwater, the carbonate balance shift towards carbonate removal takes place. This manifests in the reduction of the percentage of migration forms of calcium, magnesium, manganese, nickel and lead carbonate and in the increase of the percentage of ionic migration forms of these hydrochemical components; and also in the decrease of saturation of water with carbonate minerals. Percentage changes in the migration forms are the smallest in open hydrogeological systems.
- 5. With the reduction of the intensity of pollution dispersion in groundwater from landfills, the concentration of chlorides, which determines the reduction of the percentage of migration forms of cadmium chloride and the increase of migration forms of ions, is reduced. Percentage changes in the migration forms are the largest in open hydrogeological systems.
- 6. With the reduction of the intensity of pollution dispersion in groundwater from landfills, reduction of sulphates takes place. This manifests in the reduction of the percentage of vitriolic migration forms of calcium, magnesium, manganese, lead and nickel. Percentage changes in the vitriolic migration forms in open, semi-open and closed hydrogeological systems are very small and similar.

#### The novelty of the research

For the first time in Lithuania, the present doctoral thesis has established the characterisation of municipal waste landfills in terms of the prevailing type of hydrogeological systems, waste volume and hydrochemical characteristics of the intensive groundwater pollution zone. The methodology of selecting typical landfills has been developed. Moreover, the patterns of pollution dispersion in landfills located in different hydrogeological systems but with similar hydrochemical and waste volume characteristics have been identified for the first time.

#### The relevance of the research

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 in order to assess the patterns of landfill pollution dispersion across different hydrogeological systems. This classification will also be an important tool in assessing new locations for landfills. The research relies on a practical implementation of cluster analysis, which can also be used for handling different types of data. The study revealed the physico-chemical processes taking place in the hydrogeological environment of municipal waste landfills that change the chemical composition of groundwater. Additionally, the migration forms of chemical components and the tendencies of groundwater to dissolve or precipitate individual minerals in different hydrogeological systems and pollution zones were calculated. This data can be used for the forecasting of pollution dispersion in groundwater in landfills that are located in similar hydrogeological systems. Moreover, the results of the analysis facilitate the selection of prevention measures in order to limit pollution dispersion.

#### **Approbation of the research results**

The results of the dissertation have been presented in 4 scientific articles, 3 international and 2 national conferences.

#### The structure of the dissertation

The dissertation consists of the introduction, 4 chapters, results and conclusions and the list of the author's publications. The total corpus of the dissertation is 112 pages, 13 formulas, 32 figures, 14 tables and 105 bibliographical sources.

#### Acknowledgments

I would like to express my sincere gratitude to my supervisor Dr. Arūnas Jurevičius for his dedication, valuable advice and methodological recommendations. I would also like to thank my scientific consultant Dr. Jurga Arustienė for consulting me and providing access to the data necessary for this research. Thanks also to Danutė Karvelienė for her support and technical assistance during the preparation of this dissertation, and to mapping specialist Neringa Mačiulevičiūtė-Turlienė for advice on developing the landfill classification map. I am also indebted to Audrius Indriulionis for his assistance in mastering the methods of mathematical statistics. Last but not least, I would like to thank my husband Aurimas and other family members for their warm encouragement, patience and belief in the significance of this research.

#### 2. THE OBJECTS OF THE RESEARCH

The objects and indicators of classification. 923 non-operational Lithuanian municipal waste landfills were selected for the classification (Fig. 1). The landfills were classified based on the following indicators: 1) hydrogeological systems in which they are located; 2) the size of the waste volume; 3) hydrochemical characteristics of the intensive groundwater pollution zone, i.e. total dissolved solids (TDS) and the water class based on the prevailing anions. The first indicator was used to classify 923 landfills and the second and third indicators were used to classify 142 landfills.



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

The analysis objects of pollution dispersion in groundwater. Three typical landfills located in hydrogeological systems with different levels of buffer constrains were selected for the analysis (Fig. 2). The non-operational landfill of Kariotiškės is located in Rykantai village, Lentvaris township (Trakai district municipality). Sandy sediments of fluvioglacial origin prevail in the environs of the landfill. The non-operational landfill of Alytus is located in Takniškiai village, Alovė township (Alytus district municipality). The biggest part of the household waste area is located on the fine and ultra fine grain sand layers of fluvioglacial origin. The edges of the landfill are dominated by moraine deposits: loam or sandy loam with gravel and rare sand lenses. The non-operational landfill of Zabieliškis is located in Zabieliškis village, Pelėdnagiai township (Kėdainiai municipality district). The geological section of the neighbourhood of the landfill mainly consists of loam and sandy loam.



Figure 2. Situational map of the typical landfills.

### **3. METHODOLOGY**

#### 3.1. Methods and indicator data of landfill classification

#### The location data of landfills

The sites of non-operational municipal waste landfills were identified based on their coordinates, which together with other landfill inventory data have been stored by the Lithuanian Geological Survey since 1998 (Lithuanian Geological Survey, 2015).

#### Identification of hydrogeological systems in which landfills are located

The types of hydrogeological systems where the landfills are located were established on the basis of active groundwater circulation zone vulnerability map (Baltrūnas et al., 1998). Such maps are often used as a methodological tool to assess the risk of anthropogenic pollution to groundwater (Kazakis, Voudouris, 2015). The reasoning behind groundwater vulnerability maps is based on the idea that part of the geological deposits, depending on their filtration parameters, are more conducive to groundwater pollution. The analyzed territory is divided into districts that characterise the borders of different hydrogeological systems (Duarte et al., 2015). MapInfo 9.0 software was used to adapt the vulnerability map of the active groundwater circulation area to the use of LKS-94 coordinate system. According to the coordinates of the landfills, the map visualises the sites of the landfills and their typical geological and hydrogeological conditions.

#### Identification of the landfill waste volume

The calculations of landfill waste volume were conducted according to the truncated pyramid volume formula (3.1), whose components were obtained by assessing the dimensional parameters of the landfill environment (natural terrain or quarry). The

dimensions were assessed using the services of Lithuanian spatial information portal (Lithuanian spatial information portal, 2015).

$$V_{\text{vid}} = \frac{1}{3} H_{\text{vid}} \left( S_{\text{pagr}} + S_{\text{virš}} + \sqrt{S_{\text{pagr}} \cdot S_{\text{virš}}} \right)$$
(3.1)

where  $S_{pagr}$  – represents the landfill base area;  $S_{virš}$  – is the top of the area;  $H_{vid}$  – represents the average height of the landfill;  $V_{vid}$  – average volume of the landfill (Sokolovas, 2005).

The hierarchical cluster analysis algorithm was used to classify the waste volume.

## Identification of hydrochemical characteristics of the most intensive groundwater pollution zones

<u>Hydrochemical data and its sample.</u> The study used the data of groundwater monitoring hydrochemical analyses, conducted in 142 landfills in 1997-2013 (Lithuanian Geological Survey, 2015). The characterisation of the most intensive groundwater pollution zones relied on the charts of TDS distribution in landfills and selected sets of analyses with the maximum TDS values. The study used data of a total of 320 analyses. The reliability of hydrochemical analyses was assessed according to the electrical neutrality principle (3.2) (Appelo, Postma, 2005). The average concentrations of the following hydrochemical components: pH, Eh, CO<sub>2</sub>, TDS, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, COD<sub>Mn</sub>, Pb, Cd, Ni, Cr, Zn of the selected analyses were calculated in the next stage. The results were used to 1) classify the prevailing TDS values in the most intensive groundwater pollution zones, 2) assess the chemical damage done to groundwater based on the TDS classes, 3) identify and classify water classes based on the prevailing anions typical of the most intensive groundwater pollution zones.

$$E.N.(\%) = \frac{\sum cations - \sum anions}{\sum cations + \sum anions} \times 100\%$$
(3.2)

where *E.N.* is electrical neutrality, %;  $\Sigma$  cations and  $\Sigma$  anions is the sum of cations and anions respectively, expressed in mg equivalents.

<u>Identification and classification of water classes based on the prevailing anions.</u> In order to determine the water classes in the most intensive groundwater pollution zones, anion concentrations of  $HCO_3^-$ ,  $SO_4^{2-}$ ,  $Cl^-$ ,  $NO_3^-$ ,  $NO_2^-$  were expressed in percentage-equivalent form (%). Concrete water classes were defined based on these calculations. Only those ions whose percentage equivalent parts exceeded 20% of the equivalent value were recorded in the water class expression (Appelo, Postma, 2005). Groups of water classes were identified based on the prevailing anions in factual water classes.

<u>Classification of TDS values and assessment of chemical damage of identified</u> <u>groundwater classes.</u> The hierarchical cluster analysis algorithm was used to classify the TDS values of the most intensive groundwater pollution zones. Total pollution index Zvwas used to assess the classification groups of groundwater chemical pollution and the intensity of pollution (3.3). The following total pollution indices were also calculated:  $Zv_{bendr}$  of general hydrochemical components (HCO<sub>3</sub><sup>-</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>),  $Zv_{biog}$  of biogenic hydrochemical components (NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>) and  $Zv_{metal}$  of heavy metals (Pb, Cd, Ni, Cr, Zn). The background values typical of the chemical composition of groundwater in sandy (f III) and clay (g III) sediments were used for the analysis (Baltrūnas, 2004).

$$Zv = \sum K_k - (n-1) \tag{3.3}$$

where Zv represents the total pollution index;  $C_i$  is the actual concentration of a chemical element;  $C_f$  is the background concentration of a chemical element; n represents the number of hydrochemical components.

#### **Identification of typical landfills**

A landfill classification map was drawn in order to select typical landfills. The map was drawn at 1:1 000 000 scale using MapInfo 9.0 software. Districts of hydrogeological systems were highlighted by a respective map area colour. District boundaries of were marked using the vulnerability map of the active groundwater circulation zone (Baltrūnas et al., 1998). The size of waste volume was marked using the selected conventional sign of a proportional size. TDS groups typical of the most intensive groundwater classes based on the prevailing anions were marked by the edging of a conventional sign. In total, the map depicts 923 landfills. Since 142 landfills were classified according to the hydrochemical characteristics of the most intensive groundwater pollution zone, the remaining 781 landfills were labeled as landfills that were classified only according to the type of the hydrogeological systems.

#### The hierarchical cluster analysis algorithm

Hierarchical cluster analysis is a reliable and frequently used method for the synthesis of hydrochemical data (Singh ir kt., 2008; Thakur ir kt., 2015; Vasantkavigar ir kt., 2013). The algorithm divides the landfills into clusters in such a way that the variables are as comparable as possible, and the differences among clusters are as significant as possible.

## **3.2.** The analytical data and methods of pollution dispersion in the groundwater of typical landfills

#### Establishing landfill impact zones in groundwater

<u>Hydrodynamic and hydrochemical data</u>. Landfill impact zones in groundwater were established based on the groundwater monitoring data that is collected in the Lithuanian Geological Survey groundwater information system (Lithuanian Geological Survey, 2015) (Table 1).

Landfill	Representative period of research	Borehole number	Analysed hydrochemical components	Number of analyses	Number of hydrometric measurements
Kariotiškės	2000–2009 m.	28766, 28768, 28769, 28770, 28771, 35158, 36795, 38041	pH, Eh, TDS, CO <sub>2</sub> , Cl <sup>-</sup> , SO <sub>4</sub> <sup>2</sup> ,	119	100
Alytus	2000–2010 m.	28830, 28832, 28833, 28834, 31150, 40989	$HCO_3^{-}, Na^+, K^+, Ca^{2+}, Mg^{2+}, MH_4^+, NO_2^{-}, NH_4^+, NO_2^{-}, NO_2^{-}, Cr$	142	105
Zabieliškis	2012–2013 m.	32323, 32324, 32325, 46097, 46098, 46099	Ni, Pb, Cd Mn	98	24

**Table 1.** Landfill impact zones in groundwater were established based on the groundwater monitoring hydrodynamic and hydrochemical data.

<u>Hierarchical cluster analysis of hydrochemical data.</u> The differentiation of the landfill impact zone levels in groundwater was determined using the hierarchical cluster analysis algorithm. The main macrocomponents and TDS were chosen as hydrochemical variables:  $Cl^-$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Na^+$ ,  $K^+$ ,  $HCO_3^-$ ,  $SO_4^{2-}$ ,  $NH_4^+$ . Permanganate index  $COD_{Mn}$  that characterises the concentration levels of organic matter in groundwater was also one of the indicators. The average concentration levels of the above-mentioned hydrochemical components established in boreholes throughout the representative period were used for the hierarchical cluster analysis.

Evaluation of the chemical damage and pollution intensity in the groundwater of the landfill impact zones. In the next stage, the average concentration levels of hydrochemical components pH, Eh, TDS, CO<sub>2</sub>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, COD<sub>Mn</sub>, Cr, Ni, Pb, Cd, Mn that are typical of classification groups of the impact zones were calculated. The size ratio of these concentration levels and background concentration levels of the hydrochemical components in groundwater were estimated. Furthermore, the total pollution indices were calculated using Formula 3.3.: (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>), *Zv*<sub>biog</sub> (NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>), *Zv*<sub>metal</sub> (Cr, Ni, Pb, Cd, Mn).

<u>Graphical representation of the landfill impact zones and establishing the distance</u> <u>within which the effect of the landfill impact remains.</u> According to the hydrodynamic data grid and cluster analysis results, graphic program Corel Draw 12 derived the boundaries of the landfill impact zones. Based on the assessment results of the groundwater chemical damage, the groundwater zone with a very low landfill impact was established. The approximate distance from the landfill to the boundaries of this zone was calculated.

## Thermodynamic calculations of mineral saturation in groundwater and migration forms of hydrochemical components using computer program PHREEQC

The average concentration levels of hydrochemical variables (i.e. pH, Eh, Cl<sup>-</sup>, SO<sub>4</sub><sup>2</sup>, HCO<sub>3</sub><sup>-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, Cr, Ni, Pb, Cd Mn) that are typical of the landfill impact zones were used in the calculations as the input data. The computer software program calculated the values of the ion activity coefficient (*f*), the ion activity concentration (*a*), the molar ion concentration (*C*), and the solution ionic strength (*I*) that are typical of migration forms of hydrochemical components in different landfill impact zones. Furthermore, the values of the mineral saturation in groundwater indicator (*SI*) that characterise the balance of chemical reactions were also estimated. The percentage numbers of migration forms of all hydrochemical components in relation to the total molar concentration of each component were estimated. The saturation index is expressed by Formula 3.4 (Appelo, Postma, 2005).

$$SI = \lg \frac{IAP}{K}$$
(3.4)

where *SI* represents the saturation index; *IAP* is the multiplication of ions which will be formed in the solution; *K* represents the constant of the reaction equilibrium.

In theory, it is assumed that:

- 1. When SI > 0, the solution is saturated, rebound hypertension (mineral crystallisation) takes place.
- 2. When SI = 0, the equilibrium in the water-rock system takes place.
- 3. When SI < 0, a direct reaction takes place, i. e. minerals dissolve.

Due to the measurement and calculation errors, value SI = 0 is unlikely. Therefore, when assessing the results of the saturation index, an error range  $(\pm)$  in terms of the theoretical value of the exact equilibrium state was introduced. It was assumed that the equilibrium in the water-rock system takes place when *SI* is within  $\pm 0.3$  (i.e. the error of values *IAP* and *K* is  $\pm 30\%$ ).

#### 4. RESULTS AND DISCUSSION

#### 4.1. Results of the assessment of classification indicators

#### Hydrogeological systems in which landfills are located

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). According to the vulnerability of the active groundwater zone in terms of anthropogenic pollution, Lithuanian territory is divided into four representative districts. They are characterised by different geological and hydrogeological characteristics, such as natural insularity and filtration properties (Fig. 3): I. Very high vulnerability areas: open hydrogeological systems. They consist of loose water-permeable rock. Homogeneous aquifers prevail. II. High vulnerability areas: semi-open hydrogeological systems. The clayeyness of these sediments exceeds 50%. They are characterised by heterogeneous layers. III. Medium vulnerability areas: semiclosed hydrogeological systems. The clayeyness of these sediments exceeds 50%. They are characterised by heterogeneous layers. IV. Low vulnerability areas: closed hydrogeological systems. They consist of not less than 90% of clay and rock layers. These hydrogeological systems are characterized by homogeneous layers of low permeability.



**Figure 3.** Types of hydrogeological systems. Hydrogeological systems: I – open, simple filtration; II – semi-open, complex filtration; III – semi-closed, complex filtration; IV – closed, highly complex filtration (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.

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.

According to Bagchi, an incorrect understanding of the pollutant behaviour in a hydrogeological environment determined the fact that the majority of old landfill sites are located regardless of hydrogeological systems (Bagchi, 1994). This was corroborated by my research results: more than 70% of the landfills are located in open and semi-open hydrogeological systems that are conducive for pollution. Previous research confirms the tendency to ignore the vulnerability of hydrogeological systems when selecting the location of a landfill. Specifically, Assmuth and Strandberg (1992) studied the hydrogeological conditions in 43 landfill sites and found that most of the formations surrounding the landfill sites are dominated by sandy sediments. Antonova et al.'s study (2014) identified that small (between 0.3 and 4.0 hectares) landfills in clayey and sandy sediments and large (between 10 and 65 hectares) landfills in sandy karst sediments.

The above-mentioned tendency to ignore the vulnerability of hydrogeological systems is reflected in the great number of studies focusing on pollution dispersion in landfills of different age and size. Andersen et al. (1991), Lyngkilde and Christensen (1992), Kjeldsen et al. (2002) studied the migration processes of pollutants in ten-year old landfills of 10 ha that are located in sandy sediments. Kehew and Passero (1990) conducted research into pollution dispersion in landfills in sandy sediments that were approximately 20 years old and whose size was between 10 and 55 hectares. Therefore, the results and analysis of similar studies suggest that old closed landfills, regardless of their size and age, were located without taking into account the insulation and filtration properties of their hydrogeological systems.

#### The landfill waste volume

The minimum and maximum landfill average waste volume was 1,000 m<sup>3</sup> and 1,530,907.2 m<sup>3</sup> respectively (Fig. 4A). The calculated quartiles of 25% and 75% amounted to 150,784.5 m<sup>3</sup> and 1,941,271.2 m<sup>3</sup> respectively; the median and mean amounted to 694,841.7 m<sup>3</sup> and 1,423,319.4 m<sup>3</sup>.



**Figure 4.** The distribution of average waste volume in 142 landfills (A), dendrogram of average waste volume (B), chart of the average volume and area of the waste heap in different clusters (C).

The cluster analysis of waste volume identified nine clusters (Fig. 4B). Clusters C6 and C7 include individual landfills with a waste volume between 1,600,000 and 2,000,000 m<sup>3</sup>. Cluster C8 includes landfills with a waste volume of 3,000,000 to 3,500,000 m<sup>3</sup>. Cluster C9 includes landfills with the highest waste volume as well as individual landfills with a waste volume larger than 4,000,000 m<sup>3</sup>. Landfills belonging to clusters C6–C9 are former landfills of district regions and the main landfills of the biggest Lithuanian cities (i.e. Vilnius, Kaunas, Klaipėda, Šiauliai, Panevėžys). Taking into account this aspect and the fact that these clusters are composed of individual landfills with the largest waste volume of the whole sample, clusters C6–C9 clusters were merged into representative waste volume group V (Fig. 4C).

The landfills of cluster C4 have a waste volume that ranges from 320,000 to  $450,000 \text{ m}^3$ . The landfills of cluster C5 have a waste volume that ranges from 550,000 to  $760,000 \text{ m}^3$ . These clusters cover a relatively small number of landfills (8%) and include the landfills of the biggest cities and former district regions. In terms of the size of their waste volume, clusters C4 and C5 are quite different from the remaining clusters C1–C3. In the light of these considerations, clusters C4–C5 were merged into representative waste volume group IV (Fig. 4C).

Clusters C3 and C2 include landfills of regional towns of different sizes. Cluster C3 covers 11% of the landfills, which have a waste volume that ranges from 110,000 to 260,000 m<sup>3</sup>. Meanwhile, about 23% of the landfills fall under cluster C2 and have a waste volume ranging from 40,000 to 100,000 m<sup>3</sup>. Therefore, clusters C3 and C2 are characterised by an intermediate size of waste volume and could be considered as separate representative waste volume groups III and II (Fig. 4C).

About 49% of all the landfills are part of cluster C1. The waste volume in these landfills varies between 1,000 and 40,000 m<sup>3</sup>. Cluster C1 is characterized by landfills with the smallest waste volume across the sample and covers landfills of rural areas. Cluster C1 represents representative group I (Fig. 4C).

Studies with a focus on the amount of municipal waste in regions/cities with a different population size established that the amount of waste produced is proportional to the population size of the region or city (Mor et al., 2006). This aspect is also reflected in the results of the present thesis. Specifically, the landfills with the largest waste volume are located near the country's largest cities, while the ones with the lowest waste volume are located in villages.

Throughout literature, cases of municipal landfill waste volume classification are found only in isolated cases; additionally, most of such studies do not provide experimental justification for classification. According to Antonova et al., the category of small landfills encompasses landfills with a waste volume and area ranging between 0.3 and 4.0 hectares (< 100,000 m<sup>3</sup>), the category of average waste volume encompasses landfills with a waste volume and area ranging between 4.0 and 10 hectares (100,000 m<sup>3</sup>) and the category of high waste volume encompasses landfills with a waste volume and area ranging between 10 and 65 hectares (1,000,000 m<sup>3</sup>) (Antonova et al., 2014). Analogies and similarities are clear across the results of these scarce studies. It can be concluded that the experimentally determined limits of waste volume reliably differentiate the size of waste volume typical of the closed municipal waste landfills in the sample.

#### Hydrochemical characteristics of the most intensive groundwater pollution zones

<u>Water classes based on the prevailing anions and their classification groups.</u> 11 water classes typical of the most intensive groundwater pollution zones were identified; 8 out of 11 water classes were typical of individual landfills (Table 2). The largest percentage of landfills centres around water classes HCO<sub>3</sub> (48%), HCO<sub>3</sub>–Cl (27%) and HCO<sub>3</sub>–SO<sub>4</sub> (11%), which define the main groups of water classes based on the prevailing anions. Water class group HCO<sub>3</sub> incorporates two water classes and covers 48.6% of landfills. Water class group HCO<sub>3</sub>–Cl incorporates five water classes and covers 34.5% of landfills. Water class group HCO<sub>3</sub>–SO<sub>4</sub> combines four water classes and covers 16.9% of landfills. As can be seen, the most intensive groundwater pollution zones of the investigated landfills are dominated by water classes that are either bicarbonate or bicarbonate and have an increased concentration level of chloride.

Classifica-	Water classes based on	Num- ber of		Concentration, % ekv, range limits							
tion group the preva	the prevailing anions	land- fill	Cl-	SO4 <sup>2-</sup>	HCO3 <sup>-</sup>	CO3 <sup>2-</sup>	NO <sub>2</sub> -	NO <sub>3</sub> -			
HCO	HCO <sub>3</sub>	68	0.5-19	1.5-19	32-98	0.02-0.3	0-0.8	0-16.5			
HCO3	HCO <sub>3</sub> –NO <sub>3</sub>	1	8.6	16.8	46.3	0.0	0.0	28.4			
HCO3–Cl	HCO <sub>3</sub> –Cl	39	21-48	0.1-18	37-78	0-0.54	0-0.14	0-10.21			
	Cl	1	79.3	2.6	18.1	0.0	0.0	0.0			
	Cl-HCO <sub>3</sub>	6	44-63	2-17	32-39	0.02-0.04	0-0.05	0-2.43			
	HCO <sub>3</sub> ClNO <sub>3</sub>	1	27.9	5.4	41.3	0.0	0.0	25.4			
	HCO <sub>3</sub> Cl-SO <sub>4</sub>	2	22-32	21-31	37-37	0.03-0.04	0-0.01	0.07-0.08			
	HCO <sub>3</sub> –SO <sub>4</sub>	20	3-19	20-40	29-74	0.02-0.09	0-3.1	0-2			
	HCO <sub>3</sub> –SO <sub>4</sub> –NO <sub>2</sub>	1	4.3	20.0	46.8	0.0	21.8	1.1			
пс03-504	SO <sub>4</sub> –HCO <sub>3</sub>	2	10-18	37-45	35-45	0.03-0.07	0-0.1	0.02-18.7			
-	SO <sub>4</sub> -HCO <sub>3</sub> -Cl	1	21.3	39.0	38.2	0.0	0.1	1.3			

Table 2. Water classes and their groups based on the prevailing anions.

<u>Average TDS values and their classification groups.</u> The minimum and maximum average concentration of TDS was 453.8 mg/l and 30,700 mg/l respectively. The calculated quartiles of 25% and 75% amounted to 1,121 mg/l and 3,736 mg/l respectively; the median and mean amounted to 2,047 mg/l and 3,380 mg/l respectively (Fig. 5A). Previous research has shown that the concentration of TDS in the groundwater of wells that are located close to the waste heap and in the leachate most often varies between 300 and 20,000 mg/l and between 2,000 and 60,000 mg/l respectively (El-Hames et al., 2013; Mor et al., 2006; Christensen et al., 2001). It can be concluded that the average concentration levels of TDS and the sample of 142 landfills are representative in terms of size.



**Figure 5.** Chart of the average TDS distribution in 142 landfills (A), dendrogram of the average TDS concentration levels (B), chart of average values of the TDS distribution across different classes (C).

Different types of classification of TDS levels 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 3,000 mg/l. The following limit concentration levels, i.e. > 1,000 mg/l, > 1,500 mg/l, > 2,000 mg/l, > 3,000 mg/l, > 10,000 mg/l, are defined as unsuitable for use and dangerous for natural environment (FEPA, 1991; WHO, 2004; Sen, 2014; Hiscock, Bense 2014). The TDS levels of background concentration vary from 220 to 680 mg/l in different genetic types of aquifer (Baltrūnas, 2004).

The cluster analysis distinguished six clusters (Fig. 5B). Cluster groups C6 and C5 are characterised by TDS levels that are lower than 1,600 mg/l and 3,000 mg/l respectively. Taking into account the types of classification indicated in different

normative acts and the background concentration limits typical of different genetic types, it can be concluded that the groundwater of the landfills belonging to these clusters is clean and the least dangerous for natural environment. As a result, the clusters can be joined in one group, whose rounded TDS levels range from 400 to 3,000 mg/l (Fig. 5C). The concentration levels of TDS typical of the landfills in clusters C4 and C1 are higher than 10,000 mg/l and mostly exceed the natural background typical of groundwater. This groundwater is therefore detrimental to the environment and human health. In view of this, clusters C4 and C1 groups can also be joined in one group, whose rounded TDS levels range from 20,000 to 32,000 mg/l (Fig. 5C). The concentration levels of TDS typical of the landfills in clusters C2 and C3 occupy an intermediate position between clean, safe and highly mineralized, contaminated groundwater. Thus, cluster C2 is characterized by TDS levels whose rounded values vary from 3,000 to 5,000 mg/l. Meanwhile, cluster C3 is characterized by high TDS levels whose rounded values vary from 5,000 to 20,000 mg/l (Fig. 5C).

The chemical damage of groundwater in terms of TDS classification groups. Groundwater of very high TDS concentration levels (20,000-32,000 mg/l). This group comprises 2.1% of the investigated landfills. The landfills are located in open and semiopen hydrogeological systems. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of very high TDS concentration levels in landfills located in sandy sediments amount to Zvbendr ~ 2,044,  $Zv_{biog} \sim 21,207$  and  $Zv_{metal} \sim 242$  respectively. Groundwater of high TDS concentration levels (5,000-20,000 mg/l). This group includes about 14% of all the investigated landfill sites. The landfills of this group are located in open, semi-open, semi-closed and closed hydrogeological systems. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in sandy sediments amount to  $Zv_{bendr} \sim 491$ ,  $Zv_{biog} \sim$ 3,464 and  $Zv_{metal} \sim 68$  respectively. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in clayey sediments amount to Zvbendr ~ 163, Zvbiog ~ 1,079, Zvmetal ~ 47 respectively. Groundwater of medium TDS concentration levels (3,000-5,000 mg/l). This group includes about 15.4% of all the investigated landfills, which are located in open, semi-open, semi-closed and closed hydrogeological systems. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in sandy sediments amount to  $Zv_{bendr} \sim 216$ ,  $Zv_{biog} \sim$ 697,  $Zv_{metal} \sim 14$  respectively. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in clayey sediments amount to Zvbendr ~ 125, Zvbiog ~ 442, Zvmetal ~ 14 respectively. Groundwater of low TDS concentration levels (< 3,000 mg/l). This group includes about 68.3% of all the investigated landfill sites. The landfills are located in open, semi-open, semi-closed and closed hydrogeological systems. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in sandy sediments on average amount to  $Zv_{bendr} \sim 30$ ,  $Zv_{biog} \sim 21$ ,  $Zv_{metal} \sim 6$  respectively. The total pollution index of general hydrochemical components, biogenic hydrochemical components and metals in the groundwater of landfills located in clayey sediments on average amount to  $Zv_{bendr} \sim 13$ ,  $Zv_{biog} \sim 9$ ,  $Zv_{metal} \sim 0,1$  respectively.

## 4.2. Options of typical landfill selection

The assessment results of classification indicators revealed a wide heterogeneity of landfills in terms of waste volume, groundwater pollution and hydrogeological systems. Without categorising the values of the indicators into representative groups, the selection of typical landfills would be virtually impossible. Figure 6 shows the classification map of landfills.



Figure 6. Classification map of municipal waste landfills.

According to the classification of the three indicators visualised on the map, 31 types of landfills with the same classification indicator characteristics were identified (Fig. 6). Only three types of landfills are located in all the investigated hydrogeological systems. They characterise landfills with a minimum waste volume whose most intensive groundwater pollution zones are typical of low chemical damage. Landfills with the maximum volume waste and a high level of groundwater chemical damage are

the most suitable ones for exercises on pollution dispersion. However, the following aspects complicate the comparability of classification indicators: 1) as can be seen in the classification map (Fig. 6), these types of landfills are usually located only in 1-3 investigated hydrogeological systems; 2) landfills located in closed hydrogeological systems comprise a very small percentage of all landfills; 3) in order to solve exercises on pollution dispersion, hydrochemical data of landfill leachate, a sufficient sample of hydrochemical data of groundwater and a favourable groundwater hydrodynamic situation are needed.

Because of the heterogeneity of the groundwater monitoring programs conducted in Lithuania and the lack of leachate collection facilities in the majority of old closed landfills, hydrochemical data of landfill leachate in most landfills is non-existing. In addition, hydrochemical data of groundwater is not sufficient. These aspects complicate the selection of typical landfills in different hydrogeological systems with identical classification indicators that also fulfil other requirements. However, the classification indicators. Table 3 examplifies the procedure of typical landfill selection in different hydrogeologial systems. Selected landfills are characterised by identical groundwater chemical damage and water classes based on the prevailing anions, similar waste volume. The present doctoral thesis investigated the selected landfills as research objects in order to identify the patterns of pollution dispersion in groundwater.

Classification indicators		Name of the typical landfill						
		Kariotiškės	Alytus	Zabieliškis				
Type of a hydrogeological systems		0	O SO					
Group of waste volume, m <sup>3</sup>		>1,000,000	>1,000,000 300,000-1,000,000 1					
Factual w	vaste volume, m <sup>3</sup>	3,500,000	370,044	182,000				
	Group of TDS concentration, mg/l	5,000-20,000	5,000–20,000	5,000-20,000				
Hydrochemical characteristics of the intensive groundwater pollution zone	Factual TDS concentration, mg/l	6,440	5,000	5,668				
	Group of water classes based on the prevailing anions	HCO <sub>3</sub> +Cl	HCO <sub>3</sub>	HCO <sub>3</sub>				
	Factual water class based on the prevailing anions	HCO <sub>3</sub> +Cl	HCO <sub>3</sub>	HCO <sub>3</sub>				

<b>Table 3.</b> Typical landfills and their classification indicators.
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\* Hydrogeological systems: O-open, SO-semi-open, C-closed; TDS-total dissolved solids.

## **4.3.** The assessment results of pollution dispersion in the groundwater of typical landfills

#### The landfill impact zones in groundwater and their hydrochemical characteristics

After the cluster analysis of 10 hydrochemical indicators of the investigated groundwater boreholes in the typical landfills, three cluster groups were established (Fig. 7 and 8). The cluster groups were given appropriate names corresponding to the landfill impact zones: *the most intensive groundwater pollution zone* (the pollution zone that coincides with or is closest to the waste heap), *polluted* groundwater zone (intermediate pollution zone) and *the least polluted* groundwater zone (the pollution zone that is most distant from the waste heap).



**Figure 7.** The impact zones dendrograms of the landfills of Kariotiškės (A,), Alytus (B) and Zabieliškis (C) on groundwater.

The following total pollution indices are typical of *the most intensive groundwater pollution zones, polluted groundwater zones* and *the least polluted groundwater zones* respectively:  $Zv_{bendr} \sim 39-157$ ,  $Zv_{biog} \sim 45-511$ ,  $Zv_{metal} \sim 16-23$ ;  $Zv_{bendr} \sim 21-23$ ,  $Zv_{biog} \sim 3-38$ ,  $Zv_{metal} \sim 10-12$ ;  $Zv_{bendr} \sim 5$ ,  $Zv_{biog} \sim 1-2$ ,  $Zv_{metal} \sim 0.1-9$ . Based on the concentration coefficients and total pollution index in the established landfill impact zones, it can be noted that there is a very weak or no general hydrochemical component and metal pollution in the least polluted groundwater zone. The distance within which the impact of the landfill pollution with hydrochemical components in open hydrogeological systems is ~ 200-300 m, in semi-open systems ~ 100-200 m and in closed systems ~ 50-100 m (Fig. 8).



**Figure 8.** The impact zones scheme of the landfills of Kariotiškės (A,), Alytus (B) and Zabieliškis (C) on groundwater.

## Thermodynamic calculation results of migration forms of hydrochemical components and mineral saturation in groundwater

Regardless of the hydrogeological systems and groundwater pollution intensity, calcium, magnesium and manganese migrate mainly in the form of  $Ca^{+2}$ ,  $Mg^{+2}$  and  $Mn^{+2}$  (77–95%) (Tables 4 and 5). A small part of these hydrochemical components can be found in carbonate  $CaHCO_3^+$ ,  $MgHCO_3^+$ ,  $MnHCO_3^+$  compounds (3–16%); an even lower part of these components can be found in sulfate  $CaSO_4^0$ ,  $MgSO_4^0$ ,  $MnSO_4^0$  compounds (1–8%). Regardless of the hydrogeological systems and groundwater pollution intensity, lead mainly migrates in carbonate PbHCO<sub>3</sub><sup>+</sup>, PbCO<sub>3</sub><sup>0</sup> compounds (86–91%), but sometimes it can be found in ionic Pb<sup>+2</sup> form (5–10%). Regardless of the

hydrogeological systems, in the most intensive groundwater pollution zones the biggest part of nickel migrates in carbonate NiHCO<sub>3</sub><sup>+</sup>, NiCO<sub>3</sub><sup>0</sup> compounds (48–57%), a slightly lower part of nickel can be found in ionic Ni<sup>2+</sup>form (37–47%), and a yet smaller part in sulfate NiSO<sub>4</sub><sup>0</sup> form (2–5%). In the polluted and least polluted groundwater zones and under changed thermodynamic conditions, the largest part of nickel migrates in ionic nickel Ni<sup>+2</sup> form (49–61%), while a slightly smaller part in carbonate NiHCO<sub>3</sub><sup>+</sup>, NiCO<sub>3</sub><sup>0</sup> compounds (30–47%), and a yet smaller part in sulfate form (1–8%). Regardless of the hydrogeological systems and groundwater pollution intensity, the biggest part of cadmium migrates in ionic Cd<sup>+2</sup> form (66–90%), while smaller parts in chloride CdCl<sup>+</sup> (3–24%) and CdSO<sub>4</sub><sup>0</sup>, CdCO<sub>3</sub><sup>0</sup> (1–7%) forms. Sodium, potassium, chlorine and chromium migrate exclusively in Cl<sup>-</sup>, Na<sup>+</sup>, K<sup>+</sup>, Cr<sup>+2</sup> forms (98–100%).

C		Migration forms % in terms of the general content of a hydrochemical component								
onent	Migration	gration Landfill of Kariotiškės			L	andfill of Aly	Landfill of Zabieliškis (C)			
onent	form	(0)			(SO)					
		1	2	3	1	2	3	1	2	3
	HCO3 <sup>-</sup>	60.3	73.8	80.7	78.4	80.7	85.1	79.8	85	85.9
CO <sub>3</sub>	H <sub>2</sub> CO <sub>3</sub>	34.9	21.9	15.6	17.3	15.6	12.1	15.2	10.5	10.9
	CaHCO <sub>3</sub> <sup>+</sup>	3.9	3.4	2.8	2.6	2.7	2.1	3.5	3.1	2.1
	Ca <sup>+2</sup>	83.3	85.8	89.5	77.4	86	91.4	80	83.6	88.8
Ca	CaHCO <sub>3</sub> <sup>+</sup>	12.0	10.7	8.7	15.8	9.1	5.9	10.9	9.9	6.5
	CaSO <sub>4</sub> <sup>0</sup>	4.6	3.3	1.5	6.3	4.6	2.3	8.4	6.0	4.3
	$Mg^{+2}$	88.9	90.8	93.6	84.6	90.7	94.6	86.1	88.8	92.5
Mg	MgHCO <sub>3</sub> <sup>+</sup>	7.1	6.3	5.0	9.6	5.3	3.4	7.2	5.0	3.6
	MgSO4 <sup>0</sup> 3.9 2.7 1		1.2	5.5	3.8	1.9	6.5	5.9	3.7	
	SO4 <sup>-2</sup>	64.8	70.2	73.7	71.3	73.6	78.7	70.4	72.5	77.3
$SO_4$	CaSO <sub>4</sub> <sup>0</sup>	25.6	22.6	19.7	14.1	18.3	15.6	19.1	17.9	15.1
	MgSO <sub>4</sub> <sup>0</sup>	8.7	6.8	6.5	10.3	7.5	5.5	8.1	8.5	7.2
					dolomite Cal		calcite (	CaCO3,		
ar		dolomite CaMg(CO <sub>3</sub> ) <sub>2</sub> ,		calcite CaCO <sub>3</sub>			aragonite			
vate	coturated					J	03			
vbu	saturateu	c	alcite Ca	CO3	aragonita					
ino;							dolomite CaMg(CO <sub>3</sub> ) <sub>2</sub>			
18					cueos					
n i							dolomita			calcite
atic		ibrium aragonite CaCO <sub>3</sub>				$C_{2}M_{q}(CO_{2})_{2}$			CaCO <sub>3</sub> ,	
tur	in equilibrium					calcite CaCO <sub>3</sub>			arago-	
l sa							culotte cucoj		n	
era						aragor	ite CaCO <sub>3</sub>			CaCO <sub>3</sub>
Min										
~	non-saturated									

Table 4. Migration forms of the main macrocomponents and mineral saturation in groundwater

\* Landfill impact zones: 1: the most intensive pollution zone, 2: polluted zone, 3: the least polluted zone; hydrogeological systems: O-open, SO-semi-open, C-closed.

The thermodynamic calculations of mineral saturation in groundwater showed that the vast majority of minerals are not saturated in groundwater across all the landfill impact zones (*SI* ranges from -0.4 to -87.6). When in contact with these minerals, groundwater tends to dissolve these minerals. For the description of the process changes taking place in the landfill impact zones in groundwater, more significant are the minerals that are not saturated, in equilibrium or whose saturation changes from no saturation to equilibrium in groundwater. These minerals, depending on their saturation index, may be prone to fall out of groundwater (deposit in the layer), be in equilibrium in groundwater or to be dissolved. Their saturation index ranges from -1.1 to 2.0. Mainly, these are calcium, magnesium and manganese carbonates that are very sensitive to changes in the hydrochemical environment, as well as chromium oxide and hydroxide (Tables 4 and 5).

		Migration forms % in terms of the general content of a hydrochemical component								
Compo nent	Migration form	Landfill of Kariotiškės (O)			Landfill of Alytus (SO)			Landfill of Zabieliškis (C)		
		1	2	3	1	2	3	1	2	3
	Mn <sup>+2</sup>	85.7	87.2	90.2	81.1	87.6	92.2	83.8	86.0	90.1
Mn	MnHCO <sub>3</sub> <sup>+</sup>	11.0	10.4	8.7	14.5	9.0	6.0	10.1	9.7	6.6
	MnSO <sub>4</sub> <sup>0</sup>	3.0	2.3	1.1	4.2	3.3	1.7	5.6	4.2	3.2
	Cd <sup>+2</sup>	69.0	83.1	89.9	70.6	85.2	89.6	66.4	73.2	84.5
Cd	$CdCl^+$	23.6	8.8	2.7	15.1	4.2	2.9	19.9	13.1	5.3
Cu	$CdSO_4^0$	3.9	3.2	1.5	5.9	4.6	2.3	7.1	5.4	4.2
	CdCO <sub>3</sub> <sup>0</sup>	1.6	3.4	4.6	6	4.7	4.3	4.4	6.9	5.0
	NiHCO <sub>3</sub> <sup>+</sup> ; NiCO <sub>3</sub> <sup>0</sup>	49.8	47.1	42.3	56.6	41.2	29.7	48.2	43.1	32.7
Ni	Ni <sup>+2</sup>	47.5	51	56.8	37.5	53.8	65	46	49.0	61.1
	NiSO4 <sup>0</sup>	2.3	1.7	0.8	5.2	4.9	5.1	5.0	7.5	6.0
Dh	PbHCO <sub>3</sub> <sup>+</sup> ; PbCO <sub>3</sub> <sup>0</sup>	87.2	88.8	88.8	90.8	88.8	86.2	88.5	89.5	87.3
10	Pb <sup>+2</sup>	9.5	8.4	8.2	4.6	7.6	9.6	6.1	5.2	8.1
	saturated									
ation in tter	in equilibrium			rhodochrosite MnCO3			rhodochrosite MnCO <sub>3</sub>			
Mineral satur: groundwa	non-saturated	rhodochrosite MnCO <sub>3</sub>					rhodochro site MnCO3			rhodochr osite MnCO3

Table 5. Migration forms of the main metals and mineral saturation in groundwater

\* Landfill impact zones: 1: the most intensive pollution zone, 2: polluted zone, 3: the least polluted zone; hydrogeological systems: O-open, SO-semi-open, C-closed.

## The importance of physico-chemical processes in the filtration process of landfill groundwater pollution. <u>Inorganic macrocomponents</u>

Carbonate equilibrium. Intensive decomposition of organic matter takes place in the waste heap area, which leads to the formation of carbon dioxide  $(CO_2)$  gas.  $CO_2$ combines with water to form H<sub>2</sub>CO<sub>3</sub> acid, which dissolves carbonates. These processes increase the concentration levels of calcium, magnesium and hydrogen carbonates and groundwater becomes saturated with respect to carbonate minerals (Baedecker, Back, 1979; Kehew, Passero, 1990). The decrease in the amount of organic matter in water leads to the decrease of the concentration of carbon dioxide causing a shift in the carbonate equilibrium and carbonates to fall out. These processes determine the change trends of ionic and carbonate migration forms of hydrochemical components as the groundwater pollution intensity changes. The study found that the average concentration levels of hydrocarbonates are about 48-78% higher in the most intensive groundwater pollution zone than in the least polluted zone. The percentage change of the average hydrocarbonate concentration levels in semi-open and closed hydrogeological systems is about 12–30% higher than that in open hydrogeological systems. The average concentration levels of calcium and magnesium decrease by about 55-77% as the groundwater pollution intensity decreases. The changes in the concentration levels of calcium and magnesium across the different hydrogeological systems are insignificant and do not follow a clear change pattern: the concentration levels of calcium change by 1-5%; the comparison of open and closed hydrogeological systems differs in terms of the magnesium concentration levels by 7%. The change in semi-open hydrogeological systems is about 17–24% higher than that in open and closed hydrogeological systems.

The saturation index calculations showed that calcium and magnesium carbonate minerals (dolomite CaMg(CO<sub>3</sub>)<sub>2</sub>, calcite and aragonite CaCO<sub>3</sub>) are found in a saturated state and equilibrium in groundwater (Table 4). The groundwater of *the most intensive pollution zones* is saturated by dolomite and calcite, while the groundwater of semi-open and closed hydrogeological systems is also saturated by aragonite. Aragonite can be found in equilibrium in the groundwater of open hydrogeological systems. Across all the hydrogeological systems, the groundwater of polluted zones is saturated by dolomite and calcite. Saturated aragonite can be found only in closed hydrogeological systems, while it is in equilibrium in the groundwater of open and semi-open hydrogeological systems. The groundwater of the *least polluted zones* is in equilibrium with calcite and aragonite across all hydrogeological systems. The groundwater of open and closed hydrogeological systems is weakly saturated by dolomite minerals, while in semi-open hydrogeological systems is weakly saturated by dolomite minerals, while in semi-open hydrogeological systems this mineral is in equilibrium.

The calculation results of the migration forms of calcium and magnesium reflect the carbonate equilibrium changes in the groundwater of the landfill impact zones. Specifically, as the groundwater pollution intensity decreases, the percentage amount of calcium and magnesium carbonate forms in all the analyzed landfills decreases too. At the same time, as the carbonates are removed from the groundwater, the ionic forms of these hydrochemical components increase. The percentage part of calcium and magnesium carbonate forms amounted to 7–16% in the most intensive pollution zone, 5–11% in polluted zones and 3–9% in the least polluted zones (Table 4). The percentage part of ionic forms comprised 77–89%, 84–91% and 89–95% in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 4). As the pollution intensity decreased, the decrease of the percentage part of Ca and Mg carbonate forms in groundwater comprised 2–3%, 6–10% and 4% in open, semi-open and closed hydrogeological systems respectively. The increase of the percentage part of ionic forms in groundwater comprised 5–6%, 10–14% and 6–9% in open, semi-open and closed hydrogeological systems respectively.

<u>Ammonification and nitrification.</u> Nitrification occurs as the groundwater filtration takes places from the most intensive pollution zone in the direction of the least polluted zone. Nitrification determines the decrease of the ammonium ion concentration levels and the increase in the nitrate concentration levels. It was noted that this process occurs differently across different hydrogeological systems. In open and semi-open hydrogeological systems, as the pollution intensity decreases, the ammonium ion average concentration levels decrease by 98.5–99.9%, while the nitrate average concentration levels increase by 65–95%. However, in closed hydrogeological systems, nitrification is not that clear. In the most intensive pollution zone, both the average concentration levels of ammonium ions and nitrate were similar. As the pollution intensity decreases, the ammonium ion average concentration levels decrease from 37.3 mg/l in the most intensive groundwater pollution zone to 1.5 mg/l in polluted zones (96%) and increase to 2.6 mg/l (42%) in the least polluted groundwater zone.

<u>Sulfate reduction.</u> Sulfates can be characterized by high concentration level changes in the leachate pollution dispersion in groundwater. Experimental and laboratory studies have shown that due to sulfate reduction and complexation with free iron ions or

heavy metals, mostly low sulfate concentration levels prevail in the environs of landfills (Islam, Singhal, 2004; Harris et al., 2006). High sulfate concentration levels in groundwater are usually established where specific waste is accumulated (Kjeldsen et al., 2002). While specific waste is not accumulated in the investigated landfills, the decrease in the sulfate concentration levels is mainly due to the reduction and dilution of sulfates. As the pollution intensity decreases, the sulfate average concentration levels in ground water decrease by 74–85%. The decrease in sulfate average concentration levels in open and semi-open hydrogeological systems is approximately 9–11% higher than in closed hydrogeological systems. This is due to a more prominent effect of sulfate-reducing microorganisms in open and semi–open hydrogeological systems.

The effects of the sulfate reduction processes are visible on the changes in the migration forms of calcium and magnesium sulfates. As the pollution intensity decreases, the sulfate forms of the above-mentioned hydrochemical components are reduced by 3–4% (Table 4).

Sorption, ion exchange and dilution. Most of the potassium, sodium and chlorine compounds have a very high solubility. Potassium has a high sorption energy and is actively involved in cations exchange reactions; it is one of the main reasons that determines the decrease of the potassium concentration levels in the pollutant filtration flow direction (Kimmel, Braids, 1980). Compared with other cations, sodium has the lowest sorption energy. Chloride is a conservative hydrochemical component that is not involved in geochemical processes. Its migration front coincides with the groundwater migration front. As the pollution intensity decreases, the average concentration levels of potassium, sodium and chloride in the groundwater of the landfill impact zones decrease by 75-98%, 85-93%, 83-93% respectively. As the pollution intensity decreases, the decrease in the chloride average concentration levels in open hydrogeological systems is approximately 4-10% higher than in the groundwater of semi-open and closed hydrogeological systems. As the pollution intensity decreases, the decrease in the sodium and potassium average concentration levels is practically the same as the difference is only about 1%. In semi-open hydrogeological systems this decrease is about 10% higher than in open and closed hydrogeological systems.

In the groundwater of the landfill impact zones, potassium, sodium and chlorides migrate in exclusively ionic forms. They do not form complex compounds of varying solubility with other hydrochemical components partly because their concentration level changes in groundwater can mainly be determined by processes such as sorption, ion exchange between solid and liquid phases, and dilution.

## The importance of physico-chemical processes in the filtration process of landfill groundwater pollution. <u>Metals</u>

Sorption, complexation, carbonate equilibrium. Changes in the concentration levels during the filtration process depend on the changes of migration forms, formation of other chemical compounds, sorption and desorption on the rock surface, dispersion etc. All these processes either intensify or inhibit the spreading of metals with groundwater (Diliūnas et al., 2009; Abu-Rukah, Abu-Aljarayesh, 2002). Due to the aforementioned physico-chemical processes, most of the metal concentration levels, with the exception of somewhat higher concentration levels of manganese, in the groundwater of the investigated landfills are low. As the pollution intensity decreases, the chromium, nickel, manganese and lead average concentration levels in the groundwater of the investigated hydrogeological systems decrease by 20–92%. As the pollution intensity decreases, the

decrease in the manganese average concentration levels in semi-open and closed hydrogeological systems is approximately 34–37% higher than in open hydrogeological systems. As the pollution intensity decreases, the decrease in the chromium average concentration levels in open and semi-open hydrogeological systems is approximately 40% higher than in closed hydrogeological systems. As the pollution intensity decreases, the decrease in the nickel average concentration levels in open and semi-open hydrogeological systems is approximately 14–27% higher than in the groundwater of closed hydrogeological systems. As the pollution intensity decrease in the lead average concentration levels in open and closed hydrogeological systems is approximately 20% higher than in semi-open hydrogeological systems. The cadmium average concentration levels are practically identical in the most intensive pollution zone and in the least polluted zones (0.0003 to 0.0004 mg/l).

The calculation results show that the changes in the migration forms of manganese, nickel and lead as well as macrocomponents such as calcium and magnesium are largely determined by the carbonate equilibrium change during the filtration of groundwater pollution from the most intensive pollution zone in the direction of the least polluted groundwater zone. As the pollution intensity decreases, *manganese* carbonate forms decrease and the percentage parts of ionic forms increase across all the investigated landfills. Manganese carbonate (MnHCO<sub>3</sub><sup>+</sup>) accounted for 10–14%, 9–10% and 7–9% in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 5). The percentage part of ionic Mn<sup>+2</sup> accounted for 81–86%, 86–88%, 90–92% in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 5). As the pollution intensity decreased, the percentage part of manganese carbonate forms in the groundwater of open hydrogeological systems accounted for 2%, 8% and 4% in open, semi-open and closed hydrogeological systems respectively. The increase of the percentage part of ionic forms accounted for 5%, 11% and 6% in open, semi-open and closed hydrogeological systems respectively.

As the pollution intensity decreases, *nickel* carbonate forms decrease and the number of ionic forms increases. The percentage part of nickel carbonate forms  $(NiHCO_3^+, NiCO_3^0)$  comprised 48–57%, 41–47% and 30–42% in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 5). The percentage part of ionic Ni<sup>+2</sup> forms amounted to 37–47%, 49–54% and 57–65% % in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 5). As the pollution intensity decreases, the decrease in the percentage part of nickel carbonate forms accounted for 7%, 27% and about 15% in open, semi-open and closed hydrogeological systems respectively. As the pollution intensity decreases, the increase in the percentage part of nickel Ni<sup>+2</sup> ionic forms amounted to 9%, 28% and about 15% in open, semi-open and closed hydrogeological systems respectively.

In open hydrogeological systems, as the pollution intensity and the percentage number of  $Pb^{+2}$  ionic forms decrease, *lead* carbonate (PbHCO<sub>3</sub><sup>+</sup>) forms increase. Lead carbonate forms amounted to 87% and 89% in the most intensive pollution zone, and polluted zones and the least polluted zones respectively (Table 5). The percentage part of ionic forms accounted for 10% and 8% in the most intensive pollution zone, and polluted zones and the least polluted zones respectively. In semi-open and closed hydrogeological systems, as the pollution intensity decreases, lead carbonate forms decrease and ionic forms increase. Lead carbonate forms accounted for 89–91%, 89–90% and 86–87% in the most intensive pollution zone, polluted zones and the least polluted zones and the least forms accounted for 89–91%, 89–90% and 86–87% in

respectively. The percentage part of ionic forms amounted to 6–5%, 5–8% and 8–10% in the most intensive pollution zone, polluted zones and the least polluted zones respectively (Table 5). In the groundwater of open hydrogeological systems, as the pollution intensity decreased, the increase in the percentage part of lead carbonate forms amounted to 2%, while its decrease in semi-open and closed hydrogeological systems accounted for 5% and 1% respectively. In the groundwater of open hydrogeological systems, as the pollution intensity decreased, the decrease in the percentage part of lead ionic forms amounted to 1%, whereas its decrease in semi-open and closed hydrogeological systems accounted for 5% and about 2% respectively.

Manganese, nickel and lead carbonate minerals (i.e. rhodochrosite MnCO<sub>3</sub>, cerussite PbCO<sub>3</sub> and NiCO<sub>3</sub>) are not in a saturated state in the groundwater of all the investigated landfill impact zones and do not tend to fall into the sediment, even though their saturation index increases as the pollution intensity increases in the majority of cases. Manganese carbonate rhodochrosite (MnCO<sub>3</sub>) is in equilibrium in the groundwater of the most intensive pollution and polluted zones in semi-open and closed hydrogeological systems, while in other areas it is in a non-satured state and groundwater tends to dissolve carbonate minerals (Table 5). The groundwater of all the landfill impact zones is not saturated by lead and nickel carbonates (cerussite PbCO<sub>3</sub>, NiCO<sub>3</sub>) (SI varies from -0.45 to -6.32). These patterns may be due to very low concentration levels of manganese, lead and nickel in groundwater, which are about 10<sup>4</sup>-10<sup>7</sup> times lower than the concentration levels of macrocomponents. As a result, the influence of the carbonate system on the metal migration is significantly lower than that of the macrocomponent migration. In conclusion, as the pollution intensity decreases, the decrease in the lead, nickel and manganese concentration levels are determined by sorption processes.

The calculation of migration forms shows that *cadmium* migrates mainly in ionic and chloride forms in the groundwater of the landfill impact (Table 5). The interchange of these forms is determined by the changes in the chloride concentration levels as the groundwater pollution intensity decreases. In the groundwater of the most intensive pollution zones, where the chloride concentration levels amount to 155-242 mg/l, CdCl<sup>+</sup> migration form and ionic form consists of 15-24% and 66-71% of the total component content respectively. As the pollution intensity decreases, the chloride concentration levels and CdCl<sup>+</sup> migration forms decrease to 18-36 mg/l and 3-5% respectively, while Cd<sup>+2</sup> ionic migration forms increase to 84–90%. As the pollution intensity decreases, the percentage part of cadmium chloride CdCl<sup>+</sup> forms decreased by 21%, 12% and about 15% in open, semi-open and closed hydrogeological systems respectively. As the pollution intensity decreases, the percentage part of cadmium Cd<sup>+2</sup> ionic forms increased by 21%, 19% and about 18% in open, semi-open and closed hydrogeological systems respectively. In summary, as the groundwater pollution and chloride concentration levels decrease, CdCl<sup>+</sup> complex decomposes and part of cadmium transfers into Cd<sup>+2</sup> ionic form. Since cadmium mainly migrates only in ionic and soluble chloride migration forms, a slight decrease in its concentration levels as the pollution intensity decreases is determined, similar to the above-described metals, by sorption processes.

<u>Sulfate reduction</u>. The effects of the sulfate reduction processes are visible on the changes in the migration forms of manganese, nickel and cadmium sulfate. As the pollution intensity decreases, the sulfate forms of the above-mentioned hydrochemical components are reduced by 1–4% in groundwater (Table 5).

## CONCLUSIONS

- 1. Landfills are located in open, semi-open, semi-closed and closed hydrogeological systems. Landfills located in open hydrogeological systems account for the largest percentage (52%) of the landfills and those located in semi-closed and closed hydrogeological systems account for the smallest percentage (10–11%) of the landfills. A total of 26% of the landfills are located in semi-open hydrogeological systems
- 2. According to the volume of waste, landfills can be divided into five categories: very small landfills (< 40,000 m<sup>3</sup>), small landfills (40,000–100,000 m<sup>3</sup>), medium landfills (100,000–300,000 m<sup>3</sup>), large landfills (300,000–1,000,000 m<sup>3</sup>) and very large landfills (> 1,000,000 m<sup>3</sup>). Very small landfills make up the majority (49%) of the landfills. The percentage of small, medium, large and very large landfills is 23%, 16%, 8% and 4% respectively.
- 3. The total dissolved solid (TDS) concentration levels in the groundwater of most intensive groundwater pollution zones may be divided into four groups: low TDS level (400–3,000 mg/l), medium (TDS 3,000–5,000 mg/l), high (TDS 5,000–20, 000 mg/l), and very high (TDS 20,000–32,000 mg/l). Landfills with low TDS levels in the groundwater of most intensive pollution zones make up the majority (68%) of such landfills. Landfills with medium, high and very high TDS levels in the groundwater of intensive pollution zones account for 15%, 14% and 2% respectively.
- 4. According to the prevailing anionic composition, the groundwater of most intensive pollution zones can be divided into three classes: HCO<sub>3</sub>, HCO<sub>3</sub>–Cl, and HCO<sub>3</sub>–SO<sub>4</sub>. Landfills with groundwater class HCO<sub>3</sub> according to the prevailing anionic composition dominate in most intensive pollution zones (49%). The percentage of landfills with class HCO<sub>3</sub>–Cl and HCO<sub>3</sub>–SO<sub>4</sub> groundwater in intensive pollution zones accounts for 35% and 17% respectively.
- 5. The classification map of landfills was drawn up to facilitate selection of landfills with similar volumes of waste and similar hydrochemical characteristics of the most intensive groundwater pollution zones in each identified hydrogeological system.
- The distance within which the impact of the landfill pollution with hydrochemical components in open hydrogeological systems is ~ 200–300 m, in semi-open systems ~ 100–200 m and in closed systems ~ 50–100 m.
- 7. With the decrease of the landfill pollution dispersion in groundwater, the carbonate balance shift towards carbonate removal takes place. This manifests in the reduction of the percentage of migration forms of calcium, magnesium, manganese, nickel and lead carbonate and in the increase of the percentage of ionic migration forms of these hydrochemical components. Intensity of changes to the percentage part of these migration forms in open hydrogeological systems is 3–14% lower than that in semi-open and closed hydrogeological systems.
- 8. With the reduction of the intensity of pollution dispersion from landfills changes to the carbonate balance reduces the groundwater saturation with carbonate minerals. Groundwater is saturated or balanced with macro-component calcium and magnesium carbonates in the most intensive groundwater pollution zones or polluted zones; it is balanced in the least polluted areas; it is balanced in the least

polluted zones; it is not saturated or it is balanced with manganese carbonate in the most intensive groundwater pollution zones or polluted zones; it is not saturated in the least polluted zones; and it is not saturated with microcomponent lead and nickel carbonates in all pollution zones.

- 9. With the reduction of the intensity of pollution dispersion in groundwater from landfills, the concentration of chlorides, which determines the reduction of the percentage of migration forms of cadmium chloride and the increase of migration forms of ions, is reduced. Changes to the percentage part of migration forms of cadmium chloride and ion in open hydrogeological systems is 2–9% higher than in semi-open and closed hydrogeological systems.
- 10. With the reduction of the intensity of pollution dispersion in groundwater from landfills, reduction of sulphates takes place. This manifests in the reduction of the percentage of vitriolic migration forms of calcium, magnesium, manganese, lead and nickel. Changes to the percentage part of vitriolic migration forms in open, semi-open and closed hydrogeological systems is similar and amounts to 1–4%.

## SANTRAUKA

**Darbo tikslas** – atlikti uždarytų ir neuždarytų nebeveikiančių komunalinių atliekų sąvartynų klasifikavimą ir skirtingose hidrogeologinėse sistemose įrengtuose tipiniuose sąvartynuose įvertinti požeminio vandens cheminės sudėties kaitos dėsningumus filtruojantis komunalinių atliekų sąvartynų teršalams.

### Darbo uždaviniai:

- 1. Nustatyti uždarytiems ir neuždarytiems nebeveikiantiems komunalinių atliekų sąvartynams būdingus hidrogeologinių sistemų, atliekų tūrio ir intensyviausiai teršiamų gruntinio vandens zonų hidrocheminių charakteristikų tipus.
- 2. Sudaryti sąvartynų klasifikavimo žemėlapį ir įvertinti tipinių sąvartynų, įrengtų skirtingose hidrogeologinėse sistemose, parinkimo galimybes taršos sklaidos gruntiniame vandenyje analizei.
- 3. Tipinių sąvartynų, įrengtų skirtingose hidrogeologinėse sistemose, gruntiniame vandenyje išskirti skirtingo lygmens sąvartynų poveikio zonas ir nustatyti atstumus, kuriuose dar išlieka sąvartynų poveikis gruntiniam vandeniui.
- 4. Tipiniuose sąvartynuose, įrengtuose skirtingose hidrogeologinėse sistemose, įvertinti hidrocheminių komponentų migracinių formų ir gruntinio vandens įsotinimo mineralais kaitos tendencijas vykstant užteršto gruntinio vandens filtracijos procesui.

## Ginamieji teiginiai:

- 1. Sąvartynai yra įrengti neatsižvelgiant į hidrogeologinės sistemos, lemiančios teršalų sklaidą gruntiniame vandenyje, tipą. Sąvartynams būdinga didelė atliekų tūrio ir produkuojamos taršos sklaidos gruntiniame vandenyje hidrocheminių charakteristikų įvairovė.
- 2. Sudarytas sąvartynų tipizacijos žemėlapis kiekvienoje išskirtoje hidrogeologinėje sistemoje leidžia lengviau pasirinkti panašaus atliekų tūrio sąvartynus su panašiomis intensyviausiai teršiamos gruntinio vandens zonos hidrocheminėmis charakteristikomis.
- 3. Atstumas, kuriame dar išlieka sąvartynų taršos poveikis gruntiniam vandeniui, didėjant vandeningo sluoksnio uždarumui, trumpėja.
- 4. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje vyksta karbonatinės pusiausvyros poslinkis karbonatų iškritimo kryptimi, pasireiškiantis kalcio, magnio, mangano, nikelio ir švino karbonatinių migracinių formų procentinės dalies mažėjimu, šių hidrocheminių komponentų joninių migracinių formų procentinės dalies didėjimu ir gruntinio vandens įsotinimo karbonatiniais mineralais mažėjimu. Atvirose hidrogeologinėse sistemose migracinių formų procentinių dalių pokyčiai mažiausi.
- 5. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje mažėja chloridų koncentracija, lemianti kadmio chloridinių migracinių formų procentinės dalies mažėjimą ir joninių migracinių formų didėjimą. Atvirose hidrogeologinėse sistemose migracinių formų procentinių dalių pokyčiai didžiausi.
- 6. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje vyksta sulfatų redukcijos procesai, mažinantys kalcio, magnio, mangano, švino, nikelio sulfatinių migracinių formų procentines dalis. Atvirose, pusiau atvirose ir

uždarose hidrogeologinėse sistemose sulfatinių migracinių formų procentinių dalių pokytis labai mažas ir tarpusavyje panašus.

**Darbo mokslinis naujumas.** Pirmą kartą Lietuvoje atliktas komunalinių atliekų sąvartynų klasifikavimas pagal sąvartynų aplinkoje vyraujantį hidrogeologinės sistemos tipą, atliekų tūrį ir intensyviausiai teršiamos gruntinio vandens zonos hidrochemines charakteristikas. Parengta tipinių sąvartynų išskyrimo metodika. Taip pat pirmą kartą nustatyti taršos sklaidos dėsningumai skirtingose hidrogeologinėse sistemose įrengtuose sąvartynuose, turinčiuose panašias hidrochemines ir atliekų tūrio charakteristikas.

Mokslinė ir praktinė darbo reikšmė. Klasifikavimui reikia didelės imties reprezentatyvios ir patikimos pradinių duomenų informacijos, tačiau duomenų grupavimas turėtų palengvinti išsirinkti tipinius sąvartynus, įrengtus skirtingose hidrogeologinėse sistemose. Sudarytas Lietuvos komunalinių atliekų sąvartynų tipizacijos žemėlapis gali būti naudingas įvertinant konkrečios vietos tinkamumą sąvartynams įrengti. Darbe yra pateikiamas klasterinės analizės praktinis pritaikymas, kuris gali būti naudojamas kaip pavyzdys labai skirtingiems duomenims susisteminti. Termodinaminio modeliavimo rezultatai gali būti panaudojami sąvartynų, įrengiamų panašiose hidrogeologinėse sistemose, taršos sklaidai požeminiame vandenyje prognozuoti. Taip pat gauti tyrimo rezultatai palengvina taršos sklaidos ribojimo ir prevencijos priemonių parinkimą.

## Išvados:

- Sąvartynai yra įrengti atvirose, pusiau atvirose, pusiau uždarose ir uždarose hidrogeologinėse sistemose. Didžiausią procentinį kiekį sudaro sąvartynai, įrengti atvirose hidrogeologinėse sistemose (52%), mažiausią – pusiau uždarose ir uždarose hidrogeologinėse sistemose (10–11%). Pusiau atvirose hidrogeologinėse sistemose sąvartynų procentinis kiekis sudaro 26%.
- 2. Pagal atliekų tūrį sąvartynus galima suskirstyti į 5 grupes: labai maži sąvartynai (< 40 000 m<sup>3</sup>), maži sąvartynai (40 000–100 000 m<sup>3</sup>), vidutinio dydžio sąvartynai (100 000–300 000 m<sup>3</sup>), dideli sąvartynai (300 000–1 000 000 m<sup>3</sup>) ir labai dideli sąvartynai (> 1 000 000 m<sup>3</sup>). Vyrauja labai mažų tūrių sąvartynai (49%). Mažų, vidutinių, didelių ir labai didelių tūrių sąvartynų procentinis kiekis atitinkamai sudaro 23%, 16%, 8% ir 4%.
- 3. Intensyviausiai teršiamų zonų gruntinis vanduo pagal mineralizacijos dydį gali būti suskirstytas į 4 grupes: mažos mineralizacijos (400–3000 mg/l), vidutinės mineralizacijos (3000–5000 mg/l), didelės mineralizacijos (5000–20 000 mg/l) ir labai didelės mineralizacijos (20 000–32 000 mg/l). Vyrauja sąvartynai (68%), kurių intensyviausiai teršiamos zonos gruntinis vanduo yra mažos mineralizacijos. Sąvartynų, kurių intensyviausiai teršiamos zonos gruntinis vanduo yra vidutinės, didelės ir labai didelės mineralizacijos, procentinis kiekis yra atitinkamai 15%, 14% ir 2%.
- 4. Intensyviausiai teršiamų zonų gruntinis vanduo pagal vyraujančią anijoninę sudėtį gali būti suskirstytas į 3 klases: HCO<sub>3</sub>, HCO<sub>3</sub>–Cl, HCO<sub>3</sub>–SO<sub>4</sub>. Vyrauja sąvartynai (49%), kurių intensyviausiai teršiamos zonos gruntinio vandens klasė pagal vyraujančią anijoninę sudėtį yra HCO<sub>3</sub>. Sąvartynų procentinis kiekis, kurių intensyviausiai teršiamos zonos gruntinio vandens klasės pagal vyraujančią anijoninę sudėtį yra HCO<sub>3</sub>–SO<sub>4</sub>, yra atitinkamai 35% ir 17%.

- 5. Sudarytas sąvartynų tipizacijos žemėlapis palengvina kiekvienoje išskirtoje hidrogeologinėje sistemoje pasirinkti panašaus atliekų tūrio sąvartynus su panašiomis intensyviausiai teršiamos gruntinio vandens zonos hidrocheminėmis charakteristikomis.
- 6. Atstumas, kuriame dar išlieka sąvartynų taršos poveikis gruntiniam vandeniui atvirose hidrogeologinėse sistemose siekia ~ 200-300 m, pusiau atvirose ~ 100-200 m ir uždarose ~ 50-100 m.
- 7. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje vyksta karbonatinės pusiausvyros poslinkis karbonatų iškritimo kryptimi, pasireiškiantis kalcio, magnio, mangano, nikelio ir švino karbonatinių migracinių formų procentinės dalies mažėjimu ir šių hidrocheminių komponentų joninių migracinių formų procentinės dalies didėjimu. Šių migracinių formų procentinių dalių pokyčių intensyvumas atvirose hidrogeologinėse sistemose yra 3–14% mažesnis negu pusiau atvirose ir uždarose hidrogeologinėse sistemose.
- 8. Karbonatinės pusiausvyros pokyčiai mažėjant sąvartynų taršos sklaidos intensyvumui mažina gruntinio vandens įsotinimą karbonatiniais mineralais. Gruntinis vanduo makrokomponentų kalcio ir magnio karbonatais intensyviai teršiamose ir teršiamose zonose yra įsotintas arba pusiausvyroje, mažiausiai teršiamose zonose pusiausvyroje; mangano karbonatu intensyviausiai teršiamose ir teršiamose zonose yra pusiausvyroje arba neįsotintas, o mažiausiai teršiamose zonose neįsotintas; mikrokomponentų švino ir nikelio karbonatais visose taršos zonose yra neįsotintas.
- 9. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje mažėja chloridų koncentracija, lemianti kadmio chloridinių migracinių formų procentinės dalies mažėjimą ir joninių migracinių formų didėjimą. Atvirose hidrogeologinėse sistemose kadmio chloridinių ir joninių migracinių formų procentinių dalių pokyčiai yra 2–9% didesni negu pusiau atvirose ir uždarose hidrogeologinėse sistemose.
- 10. Mažėjant sąvartynų taršos sklaidos intensyvumui gruntiniame vandenyje vyksta sulfatų redukcijos procesai, mažinantys kalcio, magnio, mangano, švino ir nikelių sulfatinių migracinių formų procentines dalis. Atvirose, pusiau atvirose ir uždarose hidrogeologinėse sistemose sulfatinių migracinių formų procentinės dalies sumažėjimo dydis yra panašus ir sudaro tik 1–4%.

## LIST OF PUBLICATIONS

Scientific paper published in impact-factor journals referred in the Thomson Reuters Web of Science (WoS) database:

Slavinskienė, G., Jurevičius, A. 2016. Groundwater vulnerability to pollution dispersion of landfills in Lithuania. *Baltica*, 29 (1) (in press).

Other publication in peer-reviewed journals:

**Slavinskienė, G. 2011**. Methodical principles of assessment of pollution spread in groundwater on complex hidrogeologic conditions. *Proceedings of the 14th Conference for Junior Researchers "Science – Future of Lithuania"*, 120–126. [in Lithuanian]

Slavinskienė, G., Jurevičius, A. 2015. Evalution of attenuation of inorganic macrocomponents in the landfill environment under different hydrogeological conditions. *Proceedings of the 18th Conference for Junior Researchers "Science – Future of Lithuania"*, 86–93. [in Lithuanian]

Slavinskienė, G., Jurevičius, A. 2016. Speciation of heavy metals in landfill leachatepolluted groundwater: homogeneous sandy aquifer. Universal Journal of Geoscience, 4(1), 8–14.

## **CURRICULUM VITAE**

Name	Gintarė Slavinskienė
Data and place of birth	21 October 1984, Mažeikiai, Lithuania
Adress	Institute of Geology and Geography Nature Research Centre Akademijos Str. 2, LT-08412 Vilnius, Lithuania Phone: +37061545792 E-mail: gintare@geo.lt
Education	
2010-2015	PhD student in Nature Research Centre, Institute of Geology and Geography
2007–2009	Master's degree in Geology (Faculty of Natural Sciences, Vilnius University)
2003–2007	Bachelor's degree in Geography (Faculty of Natural Sciences, Vilnius University)
Work experience	
2008–till now	Senior engineer at the Sector of Climate and Water systems, Nature Research Centre at the Institute of Geology and Geography
Scientific interests	Groundwater pollution, thermodynamic modeling