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DISCRIMINATION OF CONTAMINATED SOURCES AT LANDFILL SITE AT PREDAJNÁ, SLOVAKIA

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Abstract: Groundwater pollution caused by point contamination sources is a common issue in hydrogeology. The combination of different hydrogeochemical methods represents a suitable tool for evaluation of contaminated sites. The study site is located in Central Slovakia, and is contaminated by acid tar, which leaked from the designated landfills Predajná 1 and 2. Through hydrogeochemical analysis relationships among the monitored parameters from the site were determined. The data were classified into several groups, and sampling results were spatially interpreted. Analyses revealed the dissolution of carbonates as the dominant process involved in the formation of the chemical composition of groundwater. The results indicated both inorganic and organic pollution at the study site. The inorganic pollution was evidenced by high values of EC and TDS, Na^+, Cl^-, and SO_4^{2-} ions, while organic pollution was represented by TOC and surfactants. The results also indicated that the pollution distribution is likely significantly influenced by the fissure to karst-fissure permeability of the environment.

Keywords: Groundwater pollution; Contaminated Site; Hydrogeochemistry; Slovakia

1. INTRODUCTION

Groundwater pollution by anthropogenic activities is currently a significant problem threatening drinking water resources (Fajčíková et al., 2014; Fetter et al., 2018; Barloková & Ilavský, 2018). Especially in populated areas, the rock environment and groundwater are polluted mainly from industry, landfills and other anthropogenic activities, which have a negative impact on the environment (Wagh et al., 2014).

Solid wastes which are disposed of in landfills are subjected to various physico-chemical processes while the leachate is formed. Leachate varies in composition depending on landfill type, the age of waste or the geological settings. Landfill leachate is characterised as a water-based solution of four groups: dissolved organic compounds (alcohol, acids), inorganic macrocomponents (common cations and anions), heavy metals (Pb, Ni) and xenobiotic organic compounds (Christensen et al., 2001; Naveen et al., 2018). The landfill leachate present in the environment is subjected to changes due to processes of natural attenuation. Attenuation is a passive remediation comprising processes which degrade or remove contaminants from groundwater or soil by physical, physico-chemical and microbial processes (Lv et al., 2018).

Fetter et al. (2018) defines hydrogeological assessment of environmental burdens as the process of determining existing groundwater contamination, threats to groundwater quality and the consequent risk to the beneficial use of groundwater. Understanding the processes and relationships that affect pollution transport is challenging, and therefore the interpretation of the data obtained requires a broad knowledge and selection of appropriate graphical and numerical techniques

(Hounslow, 1995; Kazakis et al., 2017).

The Predajná landfills used to dump industrial waste are long-term sources of environmental pollution. In the 1960s and 1970s, waste from acid oil refining (known as acid tars) was exported from Petrochema Dubová refinery to landfills above the village of Predajná and Lopej. Acid tars are a mixture of sulphonic acids, mineral oil, sulphonates, chlorides, free sulphuric acid and water. Acid tar leaks from both landfills were recorded in the 1970s. In the years 1978 - 1979, leakage of the material was observed by decreasing water level at the Predajná 2 landfill by 2.04 m, representing a volume of acid tars of approximately 27,000 m^3. Geological and hydrogeological surveys have confirmed the pollution of the rock environment and groundwater by acid tars in the vicinity of landfills Pollution was also observed on samples of drilling cores at a depth of more than 100 m at a distance of 2.9 km west of the landfill Predajná 2. Since 1984, acid tar waste has not been disposed of at the landfill (Jelínek et al., 2019). From 2015 to the present, the landfills have been monitored by the State Geological Institute of Dionýz Štúr (SGIDŠ).

The purpose of this study is to investigate the groundwater quality affected by contaminated site and draw attention to the suitability and effectiveness of the use of hydrogeochemical methods in the assessment of groundwater pollution in the field of contaminated sites in the vicinity of the Predajná Village in Slovakia.

2. MATERIAL AND METHODS

2.1. Study area

The study area is situated near the villages of Predajná and Lopej in Central Slovakia and comprised of two landfills and their immediate surroundings. The studied site is bordered on the west by the Jaseniansky Brook, on the east by the Veľký Grapeľ Brook, and the south by the Hron River (Fig. 1, Fig. 2).

The area of interest is built by Mesozoic rocks and Quaternary sediments. The Mesozoic bedrock is formed by the Šuňava Formation represented by sandstones and clayey shales, the Gutenstein Formation composed of limestones, dolomites, and Ramsau Dolomites of Hronicum complex. The dolomites are partly weathered and fissured. The Ramsau Dolomites are overlaying the Gutenstein Limestones. The Quaternary sediments are characterized by anthropogenic, fluvial, deluvial-proluvial, glacial and glaciofluvial deposits represented by gravels, sands, sandy gravels and deluvial sediments represented by clayey and clayey silts. The sedimentary cover is mainly represented by fluvial or terrace sediments of the Hron river and glacial deposits (Polák, 2003).

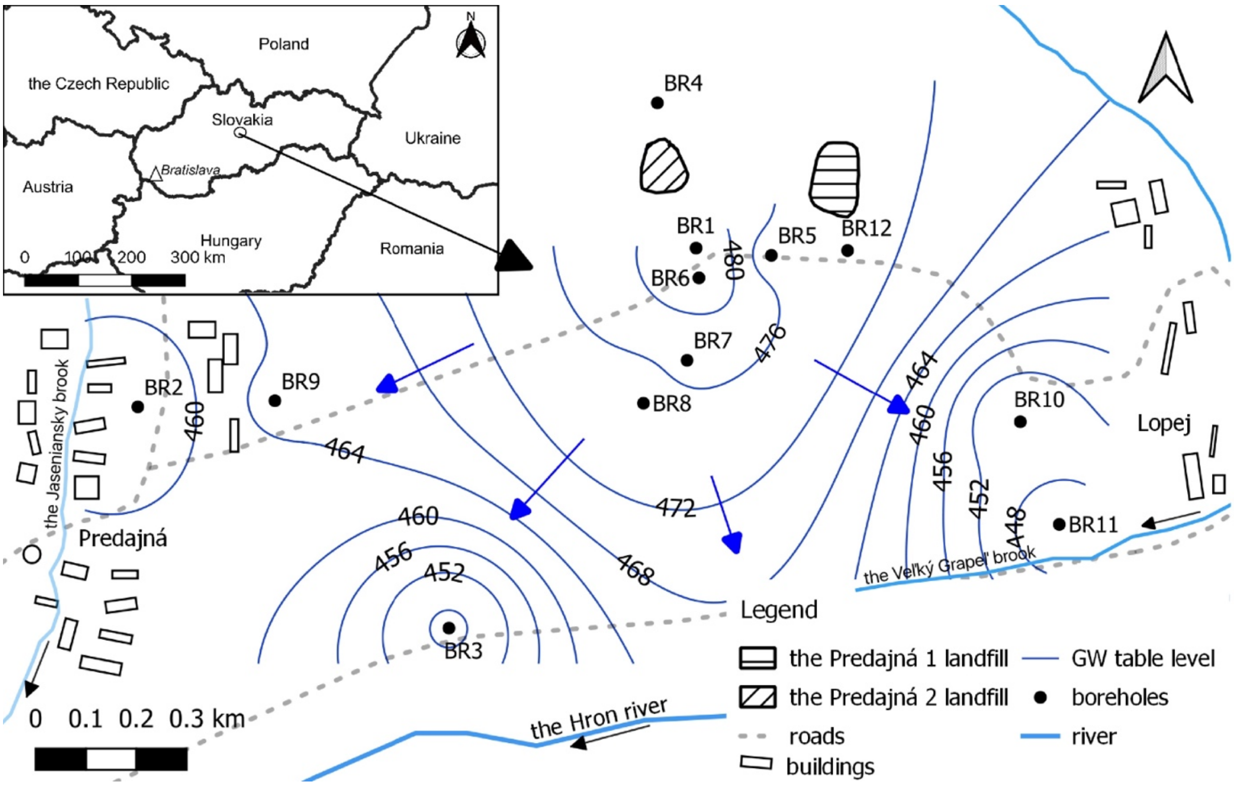

Figure 1. The map of the studied site, sampled boreholes BR1 - BR12 and flow pattern

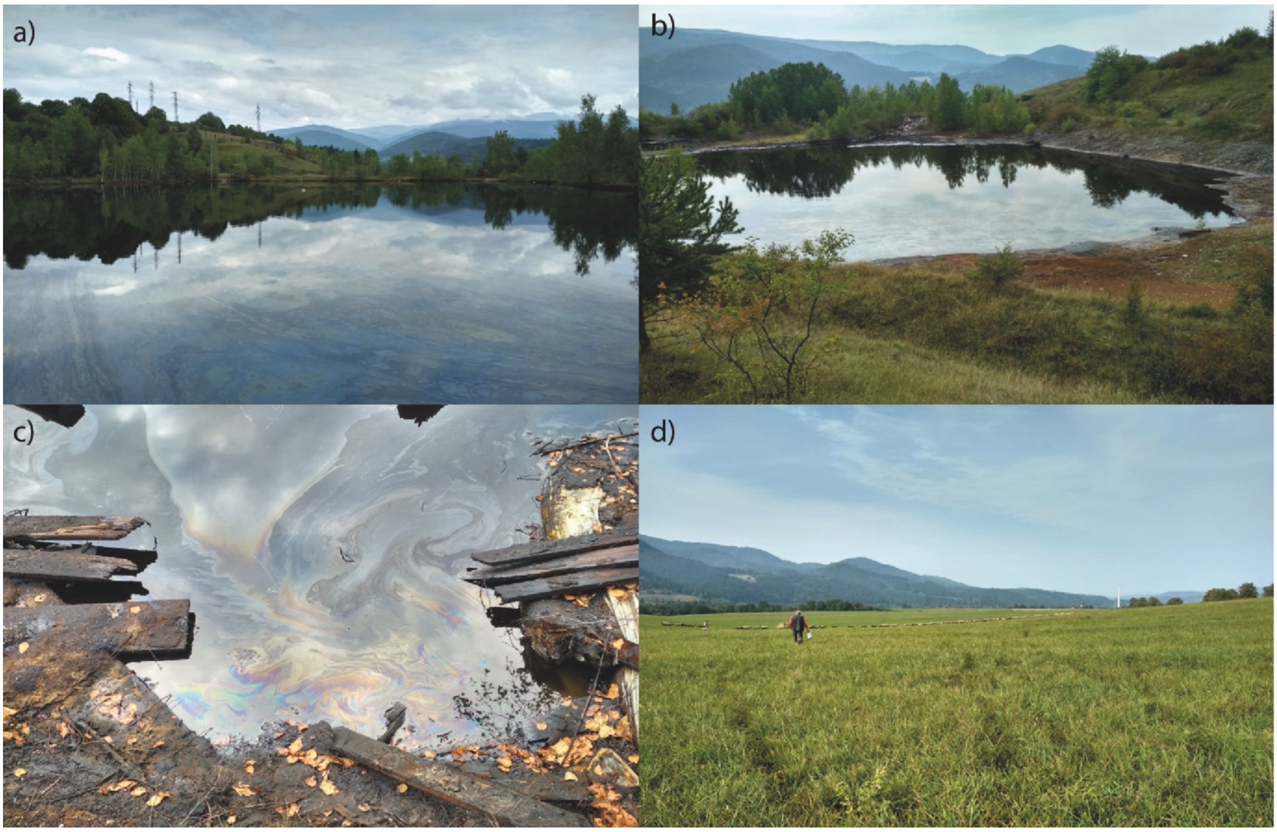

Figure 2. a) the Predajná 1 landfill, b) the Predajná 2 landfill, c) coal tar mixed with rainwater at the Predajná 1 landfill, d) surroundings south of the landfills

Lower Triassic carbonates that comprise limestones and dolomites, form principal groundwater aquifer in the study area. In the Mesozoic carbonate complex, deep groundwater circulation is presumed. The groundwater table in the area of landfills was recorded at a depth of 32 m - 39 m because the landfills are located on the hill. Thus, the groundwater is not in direct connection with the landfill. In the Quaternary fluvial gravels, shallow groundwater circulation is assumed. Groundwater flows in the direction N-S, N-SE, N-SW, respectively (Fig. 1). The Quaternary sediments are characterized by intergranular permeability; on the contrary, a characteristic fissure-karst permeability was determined for Mesozoic rocks. The hydraulic conductivity in gravel is 10^{-4} m/s, in gravelly silt in the range of 10^{-5} m/s to 10^{-9} m/s, and in fluvial and proluvial clayey or silty sediments in the range of 10^{-8} to 10^{-9} m/s (Polák, 2003; Jelínek et al., 2019; Kordík et al., 2019). Due to the presence of caverns, faults and other karst formations, it is difficult to determine accumulation of escaped pollution. It is believed that it has reached considerable depths in the rock environment. The chemical composition of groundwater is significantly influenced by the petrographic-mineralogical conditions of the area as well as anthropogenic activity (landfilling of industrial waste and agricultural use of the area).

2.2. Groundwater sampling and hydrogeochemical analysis

The study processed data obtained from monitoring carried out in November 2019. The sampled area and situation of the twelve boreholes BR1 - BR12, which were used for monitoring, is shown in Figure 1. The borehole BR4 serves as a reference borehole, i.e. it is located upgradient above the Predajná 2 landfill. Prior to groundwater sampling, groundwater table levels were measured with a Solinst 107 TLC portable water level meter. Hydrogeochemical data obtained in the field were: temperature, pH, electrical conductivity (EC), dissolved oxygen, oxygen saturation, and redox potential (Eh). The water samples were collected using Gigant pump. The water from each borehole was sampled into glass bottles with a volume of 1 l and 2 l and into plastic bottles with a volume of 1 l. The samples were analysed in laboratory using spectrophotometry (Fe^{2+}, surfactants), AES-ICP (Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Mn^{2+}), IEC (Cl^{-}, SO_4^{2-}), HTCO (TOC), AAS (As). Data on pH, EC, Na^{+}, Ca^{2+}, Mg^{2+}, K^{+}, Cl^{-}, HCO_3^{-}, SO_4^{2-}, water's hardness, surfactants, total dissolved solids (TDS) and TOC were used for analyses. TOC expresses the total content of organic substances in water; it is also used as an indicator of organic pollution (Pitter, 2015).

Aiming to determine the origin of the chemical composition of groundwater, thermodynamic calculations were carried out (Appelo & Postma, 2005; Haviarová et al., 2019) and the Mg/Ca ratio was determined. The state of water saturation for calcite and dolomite was assessed based on the saturation index (*SI*) (Pačes, 1983; Slavinskienė et al., 2018). Saturation indices for selected minerals were obtained by PHREEQC Interactive 3.3.7 software using phreeqc.dat database (Parkhurst & Appelo, 2013). The Mg/Ca ratio clarifies the proportion of dolomites and limestones forming the chemical composition of water (Pitter, 2015; Fľaková et al., 2010; Bodiš et al., 2017). The partial pressure of CO_2 (p_{CO2}) is an important indicator in the evaluation of carbonate systems and increased values also indicate degradation of organic contamination (Christensen et al., 2001).

2.2.1. Data processing

Within the descriptive statistics, which was used to obtain an overview of the examined data, the characteristics of the position (maximum, minimum, mean), and the attributes of variability (standard deviation) were measured. The values were compared with drinking water quality indicators and their limits given by Decree no. 247/2017 Coll. and with ID and IT provided by Directive no. 1/2015-7, where ID is the limit value of the concentration of a pollutant determined in soil, rock environment and groundwater, and its exceeding may endanger human health. Exceeding IT presupposes a high probability of endangering human health and the environment. The program QGIS 3 was used for map images.

3. RESULTS AND DISCUSSION

3.1. Descriptive statistic

The general statistical characteristics of the investigated physicochemical parameters of groundwater are summarized in Table 1. In total, 12 groundwater samples were examined. The minimum pH (6.62) exceeded the ID given by Directive no. 1/2015-7. pH values indicate weakly acidic to alkaline waters (Fľaková et al., 2010). Background concentration of above-mentioned ions in studied area according to the Geochemical Atlas of Slovakia (Rapant et al., 1996) are 62.12 mg/L (Ca^{2+}), 21. 40 mg/L (Mg^{2+},), 11.0 mg/L (Na^{+},), 3.9 mg/l (K^{+}), 64.36 mg/L (SO_4^{2-}), 61.34 mg/L (Cl^{-}), 83.59 mg/L (HCO_3^{-}), 0.005 mg/L (Fe), 0.025 mg/L (Mn) and 18 µg/L (As). The maximum values of pH, EC, TOC, Cl^{-}, TDS and surfactants exceeded the ID or IT given by Directive no. 1/2015-7. TOC mean values also exceed ID or IT criterion. Limits set by Decree no. 247/2017 Coll. were exceeded for EC and TOC and for Na^{+}, SO_4^{2-}, Cl^{-}, Fe, Mn and As ions. Elevated As concentrations, which were observed only in western part of area of interest, are due to ore mines located in nearby Lopej village. The data indicate contamination of the investigated site and failure to meet the requirements for drinking water.

3.2. Spatial trends

The fissure-karst environment in which the landfill is located predetermines the pollution transport. A comparison of the most significant inorganic pollution indicators at the study site (Na^{+}, Cl^{-}, SO_4^{2-}) pointed out the most polluted borehole – BR3 (Fig. 3). The coal tar was probably accumulated in the vicinity of this borehole due to preferential flow in the fissure-karst system. Boreholes located west of the landfill are only slightly affected by the pollution. Based on the surfactants and TOC the organic pollution is accumulated mainly in the immediate vicinity of the landfill. Two main directions of contamination flow were determined based on the maps – southeast (BR11) and south of landfill (BR3, BR7) (Fig. 3). The south and southeast contamination transport is the same as the general direction of groundwater flow (Fig. 1).

The contaminant / chloride ratio was used to verify the possibility of natural attenuation because Cl^{-} is a conservative tracer and its concentration is affected mainly by dispersion. Contamination in this calculation is represented by Na^{+}, SO_4^{2-}, TOC. The contaminant / chloride ratio has a decreasing tendency in N-S and N-SE directions from the landfill (Table 2). Exceptions are boreholes BR7, BR3, BR11 and partly the BR8 borehole, in which the leaked pollution likely accumulated probably due to preferential flow zones in the carbonate aquifer. The outcomes point out the possibility of natural attenuation (Christensen et al., 2001; Fetter et al., 2018). Grisey & Aleya (2016) affirm the leachate plumes are of limited extent owing to natural processes of attenuation. Sorption of oil substances on the grain surface, settling of oil tar in fissures, filling of the carbonate cracks and fissures by oil tar, and biodegradation, has probably taken place during the movement of oil tars and polluted groundwater in the groundwater flow direction.

3.3. Hydrogeochemical characteristics and speciation calculation

The $Ca-HCO_3$ and $Ca-Mg-HCO_3$ are principal ground water types at the study site. Values of TDS

Table 1. Overview of basic statistical measurements and comparison with the limits of drinking water quality indicators specified in Decree No. 247/2017 Coll. and ID and IT criterion for groundwater given by Directive no. 1/2015-7

	Min	**Max**	**Mean**	**SD**	**247/2017 Coll.**	**ID**	**IT**
pH	6.62	7.71	7.2	0.3	6.5 – 9.5	6 - 6.65 and 8.5 – 9	< 6 and > 9
Eh	-259.4	273.2	76	198.44			
EC	51.5	674	136.1	173.48	125	200	300
Ca^{2+}	47.4	254	123.4	65.46	> 30		
Mg^{2+}	12.8	108	37.46	26.82	10 – 30		
Na^{+}	3.3	1530	152.02	437.24	200		
K^{+}	0.9	33.3	6.22	9.74			
HCO_3^{-}	267	85400	432.58	155.7			
SO_4^{2-}	9.44	3275	382.65	918.84	250		
Cl^{-}	1.21	263	32.1	73.41	250	150	200
Fe	0	0.72	0.14	0.22	0.2		
As	0.0006	0.05	0.009	0.015	0.01	0.05	0.1
Mn^{2+}	0	2.68	0.68	1.24	0.05		
Hardness	171.08	1079.05	462.4	266.83			
Surfactants	0.03	0.55	0.16	0.15		0.25	0.5
TOC	0.5	46.3	7.96	12.49	3	2	5
TDS	443.19	5780.67	1193.71	1490.35		2000	3000

Note: EC [mS/m], Eh [mV], hardness, pH [-], As [μg/L] and others [mg/L].

Table 2. The contaminant/chloride ratio indicating the natural attenuation

Direction	Borehole	Na/Cl	SO_4/Cl	TOC/Cl
N-S	BR1	4.12	69.01	5.41
	BR6	0.84	0.27	0.79
	BR7	7.14	10.42	10.07
	BR8	1.32	1.41	3.21
	BR3	8.97	4.6	0.04
N-SE	BR12	0.74	5.1	7.35
	BR10	0.75	2.42	0.46
	BR11	28.11	9.03	1.49

ranged from 443 to 5,780 mg/L (Table 3). High mineralization in some boreholes indicates groundwater pollution. Common groundwater mineralization in Mesozoic structures around the villages of Predajná and Lopej, according to the Geochemical Atlas of Slovakia (Rapant et al., 1996) is around 200 – 450 mg/L. The values of the Mg/Ca ratio ranged from 0.3 to 0.7, thus corresponding to waters from dolomitic limestones or mixed circuits (Fľaková et al., 2010). The partial pressure p_{CO2} in groundwater reached the value of $9.52 \cdot 10^{-4} – 1.53 \cdot 10^{-2}$ MPa. Elevated values of p_{CO2} especially in boreholes BR12, BR1, and BR4, close to landfills indicate degradation of organic matter. The lowest pH was measured in these boreholes.

Degradation of organic matter leads to an increase in p_{CO2} as a result of which the pH decreases. The results of speciation calculations (Table 3) show in all boreholes the equilibrium in the calcite-dolomite system (Pačes, 1983; Slavinskienė et al., 2018). The water is undersaturated with respect to gypsum although elevated sulphate concentrations have been reported in many boreholes. The water from the BR4 and BR8 boreholes is at equilibrium with siderite and rhodochrosite. The equilibrium with a rhodochrosite was observed on water from BR10 and BR11 boreholes. The water from the rest of the boreholes is undersaturated with respect to rhodochrosite and siderite. Low concentrations of Fe and Mn were observed in groundwater indicating the lack of Fe and Mn oxides in carbonate rocks.

4. CONCLUSION

This study reveals the state of groundwater quality in the vicinity of contaminated site in Central Slovakia and points to the suitability of using a combination of different hydrogeochemical analyses to assess groundwater pollution. Leaks of acid tar material were recorded from the Predajná 1 and Predajná 2 landfills in Central Slovakia, together with increased concentrations of SO_4^{2-}, Cl^{-}, and Na^{+} ions, surfactants, and TOC. Hydrogeochemical calculations pointed to the dissolution of carbonates as the dominant process of the formation of groundwater chemistry. The contaminant/chloride ratio, where SO_4^{2-}, Na^{+}, TOC represented contaminants, revealed the possibility of ongoing attenuation processes. However, certain boreholes (BR3, BR7, BR11) had elevated ratio values indicating accumulation of pollution in their vicinity. Higher p_{CO2} pointed out the ongoing degradation processes nearby landfills.

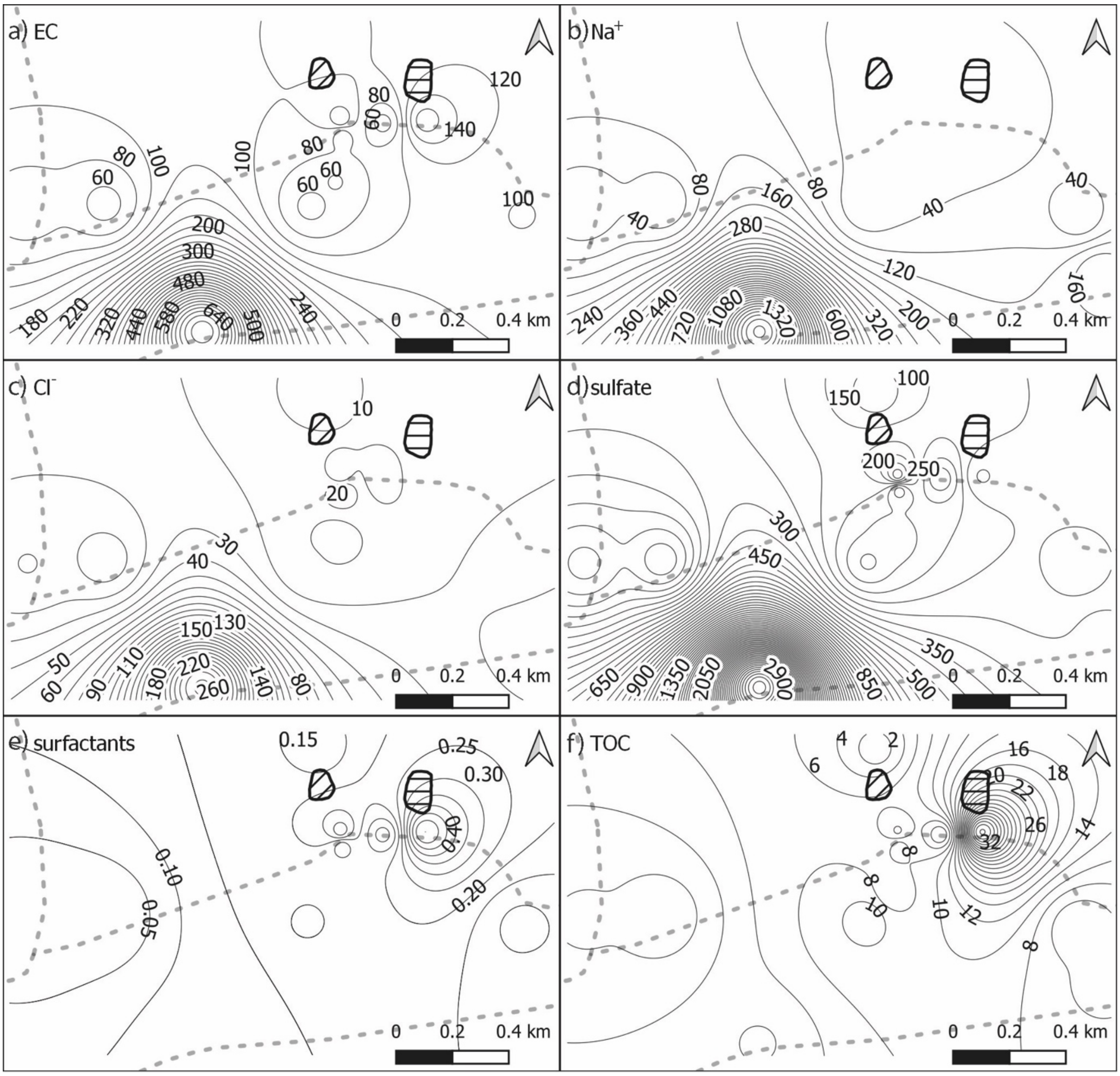

Figure 3. The spatial distribution of main contaminants in the vicinity of landfills Predajná 1 and Predajná 2: (a) EC, (b) Na^+, (c) Cl^-, (d) SO_4^{2-}, I surfactants, (f) TOC

Table 3. Basic hydrogeochemical indicators and selected results of speciation calculations for boreholes BR1 – BR12

Borehole	TDS	Mg/Ca	pH	p_{CO2}	SI				
	(mg/L)	(-)	(-)	(MPa)	calcite	dolomite	gypsum	siderite	rhodochrosite
BR1	1347	0.46	6,96	4.1×10^{-3}	0.32	0.43	-0.84	-1.98	-1.64
BR2	582	0.56	7,19	2×10^{-3}	0.32	0.52	-2.04	-	-
BR3	5780	0.36	7,35	1.44×10^{-3}	0.27	0.18	-0.40	-1.67	1.01
BR4	815	0.62	7,11	3.27×10^{-3}	0.37	0.66	-1.65	-0.36	-0.55
BR5	460	0.63	7,5	8.83×10^{-4}	0.4	0.74	-2.64	-1.35	-1.14
BR6	577	0.28	7,21	1.84×10^{-3}	0.3	0.19	-2.10	-1.25	-1.08
BR7	499	0.3	7,17	1.71×10^{-3}	0.17	-0.06	-1.78	-1.60	-1.44
BR8	443	0.34	7,36	9.52×10^{-4}	0.19	0.05	-2.00	-0.43	0.24
BR9	443	0.61	7,38	1.02×10^{-3}	0.24	0.39	-2.34	-1.82	-
BR10	850	0.52	6,81	5.36×10^{-3}	0.03	-0.09	-1.23	-1.71	0.68
BR11	943	0.45	7,71	6.28×10^{-4}	0.4	0.56	-1.47	-1.90	0.93
BR12	1578	0.7	6,62	1.54×10^{-2}	0.28	0.53	-1.00	-1.19	-0.25

Selected hydrogeochemical analyses are suitable methods for processing data from the monitoring of contaminated sites. The results offer a more precise insight into the state of the selected environmental burden, the changes and relationships between the monitored objects and the physicochemical processes that take place in the waters. The results of this study suggest that combination of various hydrogeochemical analysis can provide valuable information about contamination at studied sites. Taken together, these findings highlighted a role of abovementioned methods that can be used as the preliminary step to building conceptual site models.

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