**ORIGINAL ARTICLE** 



# Physicochemical indicators of the influence of a lined municipal landfill on groundwater quality: a case study from Poland

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#### Abstract

The paper aims to determine the most significant physicochemical indicators of the effects that a lined landfill in southern Poland has had on groundwater quality. The results of the tests of groundwater and leachate water from the landfill for the period 2009 to 2016 were subjected to a detailed statistical analysis based on the 10 physicochemical parameters. A factor analysis was conducted considering the European Union and national requirements for landfills using analytical and statistical tools. The leachate contamination indicators from a landfill were analysed to reveal their interaction with the groundwater. The assessment indicated that there was an elevated and statistically significantly higher electrical conductivity and copper and total organic carbon concentrations in groundwater hydraulically downgradient of the landfill. The assessment also indicated that there was a trend of increasing concentrations of some chemical constituents in groundwater. The adverse effects of the landfill were due to the deposited amount of waste exceeding 10 Mg per day. The impact was noticeable despite low and decreasing concentrations of heavy metals (Hg, Cu, Cd, Pb and Zn) in the leachate. The deterioration of the chemical state of the groundwater in the landfill vicinity could result in the extended time of pollutant migration or mass transport in the irrigated soil medium due to the limited efficiency of the leachate intake system or sealing screen after more than 20 years of landfill operation.

Keywords Municipal waste · Landfill · Water · Impact · Leachate

### Introduction

In the twenty-first century, landfills around the world continue to be an important part of waste management. In Poland, which is ranked sixth among the European Union countries in terms of population, 278 landfills were in use at the end of 2019 (Statistics Poland 2019). Their locations, construction details and method of operation are subject to regulation in the country. The regulation and management of landfills in Poland also consider the hydrological and geotechnical conditions for the construction and operation of these facilities to minimise the potential negative effects of stored waste on the environment (Przydatek 2019b). The existence of such facilities results from the fact that the storage process remains one of the preferred solutions in the waste management hierarchy. Despite the application of the latest technical solutions, landfills as engineering facilities still pose a serious threat to the aquatic environment (Han et al. 2016). This is because chemical processes in the mass of deposited waste can produce landfill gas and leachate that contains environmentally harmful concentrations of some dissolved chemical constituents (Abd El-Salam and Abu-Zuid 2015; Przydatek and Kanownik 2019).

The chemical composition of leachate water is determined by the composition of the waste, the way it is used, the age of the landfill, the availability of oxygen, the hydrogeological conditions and the ambient temperature and precipitation (Ilies and Mavinic 2001; Chen 2006; Longe and Balogun 2009; Regadío et al. 2013; Przydatek 2019c). Sari et al. (2013) divided leachates into young, middle-aged and mature, which is considered important in the choice of technology to neutralise them because, due to its toxic content,

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leachates pose a threat to surface water, groundwater and the soil (Barbieri et al. 2014; Naveen et al. 2017).

Some researchers (Gworek et al. 2016; Przydatek 2019c) have classified leachates as one of the main sources of heavy metal transfer to the environment. Heavy metals are problematic because of their negative effect on the environment. The migration of leachates from landfills poses a high risk to groundwater resources if not adequately protected (Patil et al. 2013). The risk of groundwater contamination by leachates from landfills requires that their composition be identified at each stage of operation and, in particular, exploitation, as groundwater is usually considered to be of good natural quality due to its geological environment (MacDonald and Calow 2009; Przydatek and Kanownik 2019). The control of the municipal landfill site remains an important aspect due to the potential threat of leachates to the surrounding water environment as a result of changes in quality (Xie et al. 2015).

The monitoring of leachates from landfills provides early identification of the risk, which may contribute to reducing the damage to both people and the environment (Salem et al. 2008). Therefore, one of the important activities aimed at recognising the environmental effects of landfilled waste with different physical and chemical properties following the guidelines of Council Directive 1999/31/EC of 26 April 1999 is to comply with environmental laws and regulations

concerning the monitoring of pollution in the area of landfills, considering the water environment, including the leachate water composition (Przydatek and Kanownik 2019). When assessing the effects of landfill sites on the aquatic environment, the use of carefully selected statistical tools and variables can be very helpful, as confirmed by some researchers (Aziz et al. 2018; Farzaneh et al. 2021; Przydatek 2021).

The appropriate operation of the landfill and, as a result, maintaining a low level of leachate water in the waste deposit is a basic requirement for water protection in the vicinity of landfills. This is important because groundwater is the main source of water supply in many urban and rural areas (Singh et al. 2016).

The study aimed to assess the impact of an lined municipal waste landfill in Poland on the physicochemical condition of groundwater in its vicinity using selected statistical tools. The landfill site that was investigated was operational during the period 2009 to 2016.

### Characteristics and geological setting of the landfill

The selected municipal landfill has been operating since 1985 in the area of city Tarnow and is one of the largest in southern Poland N:  $50^{\circ} 02' 35'' \text{ E}: 21^{\circ} 01'55''$  (Fig. 1). The land surface in the area of the landfill is gently undulating



Fig. 1 Location of landfill site

and sloping with an average slope of 5% to the east and south east. The range of the landfill site elevation is between 251.35 and 257.22 m asl.

The landfill is located in the area of the pre-Carpathian subsidence filled with the Tertiary Krakowickie Clay, on which Quaternary formations have settled. The Tertiary clay has a thickness exceeding 2000 m and is deposited in the area of the landfill at a depth of 2–5 m (Przydatek 2019b). Hirschberg (1993) showed that the capacity of clays restricts the development of leachate plumes in clay. The Quaternary formations are formed by Pleistocene glacial accumulation sediments formed as till clay and Holocene sediments formed as clays, sandy clays and locally occurring noncohesive sediments. There is one aquifer in the area of the landfill. It is at the quaternary groundwater level associated with a small interlayer and the occurrence of silty-loam sediment that overlies the Krakowicki Clay.

The direction of the groundwater flow was from northwest to southeast. The landfill under study covers an area of almost 9 ha, consisting of six sectors, including one currently in operation. Sectors I–IV were in operation until the end of 2009, and Sector V stopped service in 2018 and is undergoing reclamation. Sector VI of the landfill has been in use since 2019 (Fig. 1).

Sectors II and III are sealed on the underside with only a natural barrier made of loams and clays and on the top, they are sealed with a geomembrane. Further, Sectors I, IV, V and VI have a 2.0-mm-thick HDPE geomembrane seal on the bottom of the basin, which is on a natural layer of a geological barrier consisting of a layer of loam and sandy clay. The geomembrane is protected against mechanical damage by a geotextile with a layer of 0.5-m-thick gravel.

To protect the soil and water environment, leachates from four inactive sectors and one active sector are taken by siphons with a diameter of Ø 225 mm and a collecting drain with a diameter of Ø 315 mm, from where they go into a tank with a usable capacity of 336 m<sup>3</sup>. This tank is equipped with two open chambers. The first chamber is a settling tank and the second is equipped with a grate for leachate aeration. The leachates captured on the landfill site after the treatment process go to the sewage treatment plants through the sanitary sewage system.

Over 30,000 Mg of municipal waste with a daily quantity exceeding 10 Mg was deposited at the analysed landfill site during the eight years under study. Recently, residues from the recovery of raw material waste have been deposited as a result of secondary segregation conducted in the waste processing plant adjacent to the analysed facility. This method of waste management has helped reduce the amount of waste deposited in the landfill facility, with particularly for organic waste.

Waste materials at the storage site are placed both below and above ground with heavy equipment. During operation, waste is systematically poured over and a layer of inert waste and after the last layer is built, it is covered with humus at a thickness of 0.15 m, which is treated as an initial element of reclamation (Przydatek 2019b).

The landfill sectors are actively degassed using 76 wells. The landfill gas is taken cumulatively in one station in an active way with recovery in the form of cogenerated energy (Przydatek 2019b). According to Faitli et al. (2015), disposal of landfill gas with this energy recovery is a significant priority. Following the structural classification developed by Aziz et al. (2018), landfill construction indicates that such a facility can be classified as anaerobic.

### **Materials and methods**

The test results of the physicochemical parameters of groundwater and leachate samples, meteorological data and the number of leachate samples collected over a period of eight years were used to conduct a statistical analysis and to draw conclusions aimed at demonstrating principal factors that influence groundwater quality in in the immediate vicinity of a municipal landfill (Przydatek and Kanownik 2019).

### Assessment of the landfill water balance

In this paper, the quarterly volume of leachate and the pluviothermal conditions (quarterly sum of precipitation and average quarterly air temperature) were determined. Meteorological data were obtained from the meteorological station of the Institute of Meteorology and Water Management (N:  $50^{\circ} 1'59''$ , E:  $20^{\circ} 59' 4''$ ) located near the landfill.

### Physicochemical methodology of water tests

The electrical conductivity of groundwater and landfill leachate samples was determined in the field after the collection of each sample using a portable multifunctional meter. This device was calibrated each time before the tests were conducted. Each result was taken based on the average of three measurements (Przydatek 2019a).

The samples for metal analysis were filtered to  $0.45 \,\mu\text{m}$  in the field and placed in acid-washed bottles. The water samples were placed in sterilised plastic containers and delivered on the same day to an accredited testing laboratory for according to accredited quality and reliability methods. Then the collected samples were transported to the laboratory under the required cooling conditions (APHA 2007). The minimum volume of samples was 500 ml.

The samples of groundwater from piezometers located in the surround of the landfill site and the samples of the leachate gathered in the collection well at the landfill site were collected once a quarter from 2009 to 2016. Groundwater for laboratory tests was collected after the water from the piezometers was pumped out. The water was taken from four piezometers, including one piezometer (P-1) located hydraulically upgradient of the landfill which corresponds to the requirements (Regulation ME 2013). It is the reference level (the so-called background) for water quality comparisons for three piezometers (P-2, P-3 and P-4) located near the landfill on the groundwater outflow side, which were used to whether leachate from the landfill facility had caused groundwater contamination (Fig. 1). Raw leachate samples from the tank were taken from the drainage located at the bottom of the basin of individual sections of the landfill (including from one active cell).

The suite of chemical parameters that were analysed in groundwater and leachate samples included three general physicochemical parameters: pH, total organic carbon (TOC) and electrical conductivity (EC); six inorganic elements: cadmium (Cd), hexavalent chromium (Cr), copper (Cu), lead (Pb), mercury (Hg) and zinc (Zn); and one group of organic chemical constituents: polycyclic aromatic hydrocarbons (PAHs). Some researchers (Baun and Christensen 2004; Przydatek and Kanownik 2019) have also included Cd,  $Cr^{+6}$ , Cu, Hg, Pb and Zn as typical heavy metal elements contained in leachates.

In an accredited chemical laboratory, the concentrations of  $Cr^{+6}$  Cd, Cu, Hg, PAH and TOC were determined. The concentrations of heavy metals Cd,  $Cr^{+6}$  Cu, were determined using Atomic Absorption Spectroscopy (AAS). Moreover, the concentrations of TOC and PAHs were determined. For the TOC determination, a spectrophotometer was used to measure the absorbance value, while the PAHs were determined using the HPLC method with fluorescence detection after liquid–liquid extraction. In the examined samples, Hg was determined using the CVAAS technique (Almeida et al. 2016).

The laboratory analyses were conducted twice and repeated only when the limit values were exceeded. The results were outside the calibration curve or the abnormal results were obtained for a given matrix. The content of the demonstrated chemical contamination of the leachate was determined to the nearest  $\mu g/L$  (Przydatek 2019a).

The results of the tests on the water leaking from the landfill have been compared with the values included in the Regulation of the Minister of Maritime Economy and Inland Navigation (MMEIN) of 12 July 2019 on substances particularly harmful to the water environment and the conditions to be met when discharging wastewater into water or soil (Regulation MMEIN 2019a).

The quality of groundwater in the piezometers was determined following the Regulation of the Minister of Maritime Economy and Inland Navigation of 11 October 2019 on the criteria and method of assessment of the status of groundwater bodies (Regulation MMEIN 2019b). In addition, the results of the tested waters were compared with the limit values specified by WHO (2017) for drinking water. Han et al. (2016) analysed the impact of a landfill on the aquatic environment, using national and international standards to assess the potential for negative effects on environmental receptors.

The determination of whether concentrations of specific contaminants in groundwater and leachate samples from the landfill exceeded the relevant water quality guideline limits was based on the arithmetic mean value for each tested physicochemical parameter (Przydatek 2019a).

#### Statistical methods of the analysis results

#### Groundwater and leachate samples

For the tested indicators of leachate contamination, the following statistical parameters were determined: the minimum and maximum values, arithmetic mean and standard deviation values (Atta et al. 2015; Przydatek and Kanownik 2019). For the calculation of the values of statistical parameters, the result of the measurement at half of the limit of quantification was used where the values of the water indicators in a given sample were below the limit of quantification defined by a multiple of the detection rate. The collected results of the physicochemical parameters of groundwater samples from piezometers were used for statistical analysis to draw conclusions assessing the influence of the lined landfill on groundwater quality near the landfill facility. The nonparametric Kruskal-Wallis test and multiple comparisons of mean ranks were used to estimate the significance of the differences in the concentrations of the examined groundwater indicators upgradient and downgradient of the municipal landfill. Nonparametric tests were applied due to the lack of a normal distribution of most of the analysed physicochemical indicators according to the results of the Shapiro-Wilk test and the inequality of the variance determined using the Fisher-Snedecor test. For groundwater physicochemical indicators, significant differences between piezometers, extreme values, median and quartile spacing are presented in box plots (Figs. 2, 3).

To determine the effect of the municipal landfill on the physicochemical state of groundwater, the correlation between the physicochemical parameters of the leachate and groundwater samples from downgradient of the landfill were determined (Przydatek and Kanownik 2019). Correlation analysis is a preliminary descriptive technique to estimate the degree of association between the variables involved (Mor et al. 2006). Whether the number of leachate samples and pluviothermal factors influenced the physicochemical state of the groundwater in the piezometers was also checked. For this purpose, correlation coefficients were determined for data with a normal distribution, using



Fig. 2 Differences of values physicochemical indicators

Pearson's linear correlation coefficient method. If the condition of a normal distribution was not met, the Spearman's rank method was used, where Spearman's R correlation coefficient is the nonparametric equivalent of Pearson's coefficient. A rank correlation reveals any monotonous dependence (also nonlinear). As in the case of parametric correlation, Spearman's R correlation coefficient measures the strength of the dependence between variables, but in this case, a quantitative scale with a normal distribution is no longer required (Jiang et al. 2019; Przydatek and Kanownik 2019).

The analysis of the time trend of physicochemical indicators in the leachate from the landfill and groundwater was carried out using the nonparametric Mann–Kendall test. This test is often used in the analyses of climate change (Sa'adi et al. 2019), hydrology (Ali et al. 2019; Książek et al. 2019) and water quality (Kanownik et al. 2019). The hypothesis on the lack of a data trend on the basis of a nonparametric correlation coefficient is verified. The nonparametric equivalent of the correlation coefficient used in the Mann–Kendall test is the rank correlation coefficient of a data string and a sequence of corresponding time moments, called the Kendall tau coefficient. A positive  $\tau_c$  indicates a growing tendency, while a negative  $\tau_c$  suggests a decreasing trend. For this study, a change in the significance level of  $\alpha = 0.05$  was assumed as a statistically significant increasing or decreasing trend. Statistical analyses were conducted in Statistica 12 by StatSoft (StatSoft Inc. USA; 12 for Windows).

### Quality analysis of leachate and groundwater samples and pluviothermal indicators in the landfill site region

The average quarterly volume of leachate that was produced from the landfill from 2009 to 2016 was  $5.655 \text{ m}^3$ . The average quarterly precipitation was 189.6 mm and the quarterly air temperature during the research period averaged 9.7 °C (Table 1).

Piezometer

Piezometer

4000

3000

2000

1000

2000

4000





Fig. 3 Correlation between piezometers below landfill site

8000

10000

Landfill leachate

12000

14000

-2

16000

18000

P-3 🔪 P-4

6000

 Table 1
 Quarterly amount of leachates from the landfill site and basic

 meteorological data (quarterly sum of precipitation and average air
 temperature for the quarter)

Parameters	Min–max	Average	Standard deviation
Amount of the leachates (m <sup>3</sup> )	743–10,231	5655	3048
Precipitation (mm)	64.5–518.4	189.6	176.9
Ambient temperature (°C)	-1.1-22.3	9.7	7.06

During the 8 year study period, the average concentration of the tested substances that particularly were harmful to the water environment in the water leached from a municipal landfill site was several times lower than the maximum average normative limit values (Regulation MMEIN 2019a). The leachate pH reached high values, ranging from 7.5 to 9.0, with an average of 8.2. The concentration of Hg in water samples was generally less than 5  $\mu$ g/L, with a maximum value of 30  $\mu$ gl.

The maximum concentration of Cd was 36  $\mu$ gl, with a maximum value of 200  $\mu$ g/l (Table 2). Examined leachates

from the landfill site did not meet the requirements for the discharge of wastewater into water bodies or to ground due to the high concentrations of TOC. The minimum concentration of the TOC was 152 mg/L and the maximum was 1730 mg/L. The average value over the eight years was 679 mg/L, which was more than 22 times higher than the maximum permitted value of 30 mg/L. Leachates from the landfill were characterised by high specific electrical conductivity ranging from 2.721 to 17.170  $\mu$ S/cm. The concentrations of the tested heavy metals were very low. That same low concentration of heavy metals was demonstrated by Przydatek and Kanownik (2019). The highest concentrations were the following: Zn was 0.42 mg/L, hexavalent Cr was 0.15 mg/L, Cu was 0.06 mg/L and Pb was 0.33 mg/L.

The analysis of the groundwater quality in piezometers indicated that most of the determined physicochemical elements meet the standards of very good water quality at Class I (Table 3). During the 8 year testing period of the groundwater samples collected upgradient of the landfill from piezometer P-1, only the concentration of TOC was higher than the limit value permitted for Class I, which qualified the Table 2 Statistical parameters describing the pollution indicators in the leachates from the municipal solid-waste landfill site and admissible values

Pollution indicators	Min–max	Average	Standard devia- tion	The highest admissible values in accordance with Regulation MMEIN (2019a)
Substance especially harmful to the aqu	atic environment			
Mercury (µg/L)	0.07-4.4	0.43	0.77	30
Cadmium (µg/L)	<1–36	11	13	200
Other indicators				
pH	7.5–9.0	8.2	-	6.5–9.0
TOC (mg/L)	152-1730	679	377	30
Zinc (mg/L)	< 0.03-0.42	0.12	0.07	2
Hexavalent chromium (mg/L)	< 0.01-0.15	0.018	0.026	0.5
Copper (mg/L)	< 0.005 - 0.06	0.027	0.017	0.5
Lead (mg/L)	0.003-0.33	0.06	0.08	0.5
Electrical conductivity (µS/cm)	2721-17,170	10,711	3158	_
PAH (µg/L)	< 0.017-4.42	0.31	0.79	-

Table 3 An average values of physicochemical indicators and groundwater quality class in the piezometer above the landfill site

Physicochemical indicators	Piezometer		Limit value in cl	asses (Regi	ulation M	MEIN <mark>2(</mark>	)19b)	WHO (2017)
	P-1		Hydrochemical	Ι	II	III	IV	
	Min–max	Average	background					
pН	6.8–9.5	7.6	6.5-8.5	6.5–9.5			<6.5 or>9.5	_
TOC (mg/L)	<1-17.2	6.3	1–10	5	10	10	20	_
Electrical conductivity (µS/cm)	218-870	643	200-700	700	2500	2500	3000	-
Hexavalent chromium (mg/L)	< 0.01	0.006	0.0001-0.01	0.01	0.05	0.05	0.1	0.05
Zinc (mg/L)	< 0.01-0.05	0.022	0.005 - 0.05	0.05	0.5	1	2	_
Cadmium (µg/L)	< 0.2-<1	0.19	0.1-0.5	1	3	5	10	3
Copper (mg/L)	< 0.002-0.015	0.003	0.001-0.02	0.01	0.05	0.2	0.5	2
Lead (mg/L)	< 0.002-0.029	0.004	0.001-0.01	0.01	0.025	0.1	0.1	0.01
Mercury (µg/L)	< 0.05-1.1	0.11	0.05-1	1	1	1	5	6
PAH (µg/L)	< 0.017-< 0.1	0.017	0.001-0.1	0.1	0.2	0.3	0.5	-

water as good quality water (Table 3). The average specific electrical conductivity was close to the limit value (700 µS/ cm) for Class I, which was 643 µS/cm. By contrast, groundwater from downgradient of the landfill that was sampled from piezometer P-2 was classified as good quality water for Class II due to the high average specific electrical conductivity, which exceeded the limit value by 22 µS/cm. The other tested physicochemical parameters were at the level of very good quality water. In piezometers P-3 and P-4, the water was more polluted and of much lower quality. When compared with piezometer P-1, the concentration of TOC increased more than three times, which qualifies the water at Class V (i.e. poor quality water), where the values of the physicochemical elements confirm the significant influence of human activity. The maximum electrical conductivity value (8.190 µS/cm) was significantly higher in P3,

with the mean of this indicator at 1.757 and 1.282  $\mu$ S/cm in piezometers P-3 and P-4. The mean Cu concentration was 0.011 mg/L in the piezometer P-3, which exceeded the limit value for very good quality water (Class I), classifying the water as Class II. The values of other physicochemical elements were at a low level and did not exceed the limit values of groundwater quality Class I (Table 4). Groundwater quality testing showed that the lead concentration sporadically exceeded the limit value of 0.01 mg/L for drinking water (WHO 2017).

#### **Comparative statistical analysis**

The comparative statistical analysis of physicochemical indicators in groundwater carried out using the nonparametric Kruskal–Wallis test revealed that, out of the 10 analysis

Table 4 $An$	average values o	of physicoche.	mical indi	cators and grou	undwater qual	ity class in the	piezometers b	elow the landfill site				
Phys-	Piezometer							Limit value in classes	(Regulation MI	MEIN 2019b)		WHO (2017)
indicators	P-2 F	-3	P-4	P-2	P-3		P-4	Hydro- I	Ξ	Π	IV	
	Min-max			Average				chemical background				
Hd	6.5–8.8	6.3–8.0	6.6-	8.77.56		7.22	7.27	6.5-8.5 6.5-9.5			< 6.5 or > 9.5	1
TOC (mg/L)	<1-19.9	<1-380	3.7-4	1.34.8		29.6	20.1	1-10 5	10	10	20	I
Electrical conduc- tivity (μS/ cm)	515-856	1084-8190	160-2-	410 <i>7</i> 22		1757	1282	200-700 700	2500	2500	3000	I
Hexavalent chromium (mg/L)	< 0.01	<0.01	< 0.01	0.005		0.005	0.005	0.00001- 0.01 0.01	0.05	0.05	0.1	0.05
Zinc (mg/L)	< 0.003- 0.25 0	<0.007- 1.13	<0.010 0.069	⊢ 0.029		0.026	0.021	0.005-0.05 0.05	0.5	1	7	I
Cadmium (µg/L)	< 0.1-0.9	< 0.2–2	< 0.2 - <	1.0 0.20		0.34	0.19	0.1–0.5 1	б	Ś	10	c
Copper (mg/L)	< 0.002- 0.023 0	<0.002- 0.083	<0.002 0.021	- 0.0031		0.0110	0.0059	0.001 -0.02 0.01	0.05	0.2	0.5	2
Lead (mg/L)	< 0.002- 0.01 0	<0.002- 0.030	<0.002 0.021	- 0.0030		0.0053	0.0029	0.001-0.01 0.01	0.025	0.1	0.1	0.01
Mercury (µg/L)	< 0.05-0.6	<0.05-<0.9	< 0.05-<	2.50.059		0.089	0.216	0.05–1 1	1	1	5	9
PAH (µg/L)	<0.002-<0.1	<0.002-<0.1	<0.002-<	<0.10.015		0.016	0.017	0.001-0.1 0.1	0.2	0.3	0.5	

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indicators, four (pH, TOC, electrical conductivity and Cd) differed statistically significantly between piezometers at the level of significance of  $\alpha$ =0.001 (Table 5). The values of the physicochemical elements in the groundwater do not differ between the piezometer (P-1) located upgradient of the land-fill and the piezometer (P-2) downgradient of the landfill. The lack of difference in the physicochemical composition of the groundwater between these piezometers may have resulted from the location of piezometer P-2, which was in the central part of the landfill edge (Fig. 1). This may have caused the groundwater that flows directly through footprint of the landfill to bypass this piezometer.

However, differences in the values of physicochemical parameters in the groundwater were found between piezometers P-1 and P-2 and piezometers P-3 and P-4. The pH values of the water below the landfill in piezometers P-3 and P-4 were statistically significantly lower than in the groundwater at the inflow in piezometers P-1 and P-2. However, the concentration of TOC, the value of the specific electrical conductivity and the concentration of Cu were statistically significantly higher in the groundwater downgradient of the landfill (Fig. 2). These physicochemical parameters, in addition to the pH, also indicated that leachate from the landfill had caused the deterioration of groundwater quality.

To recognise the effect of the landfill on the physicochemical composition of the groundwater, a correlation analysis was conducted between the water in the piezometers downgradient of the landfill (P-2, P-3 and P-4) and leachate samples from the landfill. A statistically significant correlation (positive correlation) between the groundwater in piezometer P-2 and the leachates was demonstrated for heavy metals (i.e. Cd and Hg concentrations and a negative correlation for PAHs; Table 6). The groundwater in piezometer P-3 was found to be statistically significantly correlated with the leachate for concentrations of TOC, Zn, Cu, PAHs and EC. In turn, the groundwater in the P-4 piezometer was significantly positively correlated with the leachate for all tested general parameters (pH, TOC and electrical conductivity) and for Zn, Pb and Hg (Fig. 3). The correlation of PAH concentrations exhibits an increase in leachates from the landfill causing a decrease in its concentration in the groundwater in piezometers below the landfill. According to Rosik-Dulewska et al. (2007), the PAH concentration decreases with time.

The analysis of the effect of the volume of leachates and pluviothermal factors (sum of the quarterly precipitation and average quarterly air temperature) on the physicochemical state of groundwater in piezometers revealed that only the air temperature has a statistically significant effect on the concentration of TOC in piezometer P-4. Statistical analysis of groundwater quality indicated a statistically significant upward trend of various concentrations and, in the case of leachate, a downward trend (Table 7). For the groundwater upgradient of the landfill (P-1), an increasing trend was noticeable for four indicators-TOC, EC, Cu and Pb concentrations. On the other hand, downgradient of the landfill, five indicators-pH, TOC, EC, Cu and Pbexhibited increasing trends. The concentration of PAHs in the examined groundwater tended to decrease with the groundwater flow. The study of the leachate composition showed a downward trend of the concentrations for four indicator-Cd, Cu, Pb, Zn-while the PAH concentration increased. The different trends may be due to the extended time of migration of pollutants or mass transport in the irrigated soil medium (Przydatek and Kanownik, 2019).

Physicochemical indicators	Piezometer				Results of Kru	ıskal–Wallis test
	P-1	P-2	P-3	P-4	Test value	Probability test ( <i>p</i> )
	Median					
рН	7.50 <sup>3; 4 a</sup>	7.50 <sup>3; 4</sup>	7.24 <sup>1; 2</sup>	7.20 <sup>1; 2</sup>	17.5	<0.001 <sup>b</sup>
TOC (mg/L)	<i>4.6</i> <sup>3; 4</sup>	<i>3</i> .6 <sup>3; 4</sup>	<i>13.9</i> <sup>1; 2</sup>	20.1 <sup>1;2</sup>	52.9	< 0.001
Electrical conductivity (µS/cm)	<i>633</i> <sup>3; 4</sup>	725 <sup>3; 4</sup>	1.381 <sup>1; 2</sup>	1,296 <sup>1; 2</sup>	76.2	< 0.001
Hexavalent chromium (mg/L)	0.005	0.005	0.005	0.005	2.77	0.43
Zinc (mg/L)	0.025	0.025	0.025	0.025	0.88	0.83
Cadmium (µg/L)	0.15	0.15	0.15	0.15	3.44	0.33
Copper (mg/L)	0.003 <sup>3; 4</sup>	0.003 <sup>3; 4</sup>	0.007 <sup>1; 2</sup>	0.005 <sup>1; 2</sup>	29.0	< 0.001
Lead (mg/L)	0.002	0.002	0.002	0.002	1.90	0.59
Mercury (µg/L)	0.025	0.025	0.025	0.025	1.51	0.68
PAH (µg/L)	0.009	0.009	0.009	0.013	0.32	0.96

Table 5 Comparison of physicochemical indicators values between piezometers using the nonparametric Kruskal-Wallis test

<sup>a</sup>The digits in the upper index mean a piezometer in which the parameter values are significantly different for multiple (two-sided) comparisons of the average ranks

<sup>b</sup>Statistical values in italic mean statistically significant differences at p < 0.001

 Table 6
 Correlation dependence

 of physicochemical indicators

 between water in piezometers

 P-2, P-3 and P-4 and leachates

 from the landfill site

Physicochemical indicators	Piezometer	R spearman	t (n-2)	Test probability (p)
рН	P-2	0.327	1.83	0.08
	P-3	-0.012	-0.053	0.96
	P-4	0.39	2.27	$0.03^{a}$
TOC (mg/L)	P-2	0.10	0.512	0.61
	P-3	0.51	2.52	0.02
	P-4	0.45	2.7	0.01
Electrical conductivity (µS/cm)	P-2	0.018	0.095	0.92
	P-3	0.52	2.74	0.021
	P-4	0.36	2.07	0.048
Hexavalent chromium (mg/L)	P-2	_		
	P-3	_		
	P-4	_		
Zinc (mg/L)	P-2	0.17	0.93	0.40
	P-3	0.57	3.00	0.007
	P-4	0.49	2.97	0.006
Cadmium (µg/L)	P-2	0.12	0.68	0.50
	P-3	-0.03	-0.14	0.89
	P-4	0.33	1.91	0.07
Copper (mg/L)	P-2	-0.32	-1.80	0.08
	P-3	-0.44	-2.58	0.02
	P-4	-0.031	-0.137	0.89
Lead (mg/L)	P-2	0.055	0.291	0.77
	P-3	0.097	0.423	0.68
	P-4	0.153	0.821	0.42
Mercury (µg/L)	P-2	0.521	3.23	0.003
	P-3	0.309	1.42	0.17
	P-4	0.500	3.06	0.005
PAH (µg/L)	P-2	-0.69	-5.05	< 0.001
	P-3	-0.456	-2.23	0.038
	P-4	-0.575	- 3.72	< 0.001

<sup>a</sup>Italic value of statistics means that the relationship is statistically significant at p < 0.05

 Table 7
 Time trends of examined parameters quality of leachate and groundwater

Physicochemical indicators	Leachate	Piezo	Piezometer				
		P-1	P-2	P-3	P-4		
рН				1	1		
TOC (mg/L)		<b>↑</b>	Î				
Electrical conductivity (µS/cm)		<b>↑</b>	<b>↑</b>	$\downarrow$			
Hexavalent chromium (mg/L)							
Zinc (mg/L)	$\downarrow$	$\downarrow$		$\downarrow$	$\downarrow$		
Cadmium (µg/L)	$\downarrow$						
Copper (mg/L)	$\downarrow$	<b>↑</b>	<b>↑</b>	1	1		
Lead (mg/L)	$\downarrow$						
Mercury (µg/L)		<b>↑</b>	Î	<b>↑</b>	1		
PAH (µg/L)	↑	↓	↓	$\downarrow$	↓		

A statistically significant upward  $(\uparrow)$  and downward  $(\downarrow)$  trend

# Discussion

Leachate is still one of the most serious dangers in landfills (Przydatek 2019a). The analysis of the composition of leachate water from a landfill that has been in use for more than 20 years with a waste accumulation of more than 10 Mg per day exhibited high variability over time.

In the raw leachate samples from the tested municipal landfill, pH peaked at 9.0 with an average of 8.2. This indicates that the leachate comes from a mature landfill, which has been in use for over ten years (Nájera-Aguilar et al., 2019) and is in the methanogenic phase (Wdowczyk and Szymańska-Pulikowska 2019). In turn, low concentrations of Hg at 5  $\mu$ g/L and Cd at 36  $\mu$ g/L were recorded in the investigated leachate.

Similarly, low Hg concentrations in tested leachate samples were found by Talalaj (2013). Przydatek and Kanownik (2019) also found a low Cd concentration (mean 1.05 µg/L),

which was considered a factor in the deterioration of the groundwater quality below the landfill. In addition, concentrations of other heavy metals (e.g. Zn, Cu and Pb) in the tested leachates were low with a simultaneous downward trend. The highest concentration of Zn was 0.42 mg/L, which differed from that found by Boateng et al. (2019) in Africa. Kanownik and Policht-Latawiec (2016) considered the low concentration of heavy metals (i.e.  $Cr^{+6}$ , Ni, and Pb), to be a characteristic of old municipal landfills. Abiriga et al. (2020) identified the age of waste as the most significant factor in groundwater contamination in the vicinity of a landfill.

Significant deterioration of leachate quality was caused by an increased concentration of TOC at an average value of 679 mg/L. Gellert (2000) demonstrated that this indicator at concentrations between 1.44 and 74.4 mg/L is toxic. Another tested indicator, electrical conductivity as a measure of salinity in the tested leachates, reached high values in the range of 2.721 to 17.170  $\mu$ S/cm. Slightly lower maximum electrical conductivity values in the tested leachates in Serbia were obtained by Tenodi et al. (2020).

The deterioration of the groundwater quality at the outflow was primarily influenced by the high average electrical conductivity values of 1.757 and 1.282 µS/cm and the average Cu concentration of 0.011 mg/L. Gupta and Paulraj (2017) classified Cu as a particularly toxic element, which was demonstrated based on a statistical analysis. The high value of specific electrical conductivity in groundwater (Brindha et al. 2014; Maiti et al. 2016; Deshmukh and Aher 2016) has been linked by some researchers to the effect of landfill leachate. The deterioration of groundwater quality near the analysed landfill site was also significantly affected by a more than threefold increase in TOC concentration at the outflow and by the increased value of this indicator, which generally affected their classification as the worst, at Class V. Huan-Jung et al. (2006) also discovered high concentrations of both electrical conductivity and TOC based on the studies of water in the area of landfills.

A noticeable increase in the concentration of PAHs in the tested leachates is related to anthropogenic sources, particularly from the incomplete combustion of fuels (Malakahmad et al. 2016). Many compounds from the PAH group exhibit carcinogenic properties and a strong mutagenic effect (Kubiak 2013). However, a noticeable decrease in the concentration of PAHs in groundwater below the landfill results from the degradation of its compounds in the landfill and volatilisation by landfill gas (Talalaj and Biedka 2016).

The deterioration of water quality below the landfill was also indicated by significantly higher concentrations of physical and chemical parameters, particularly with specific electrical conductivity, TOC and Cu, which had noticeable uptrends. Koda et al. (2017) also showed high concentrations of TOC in groundwater contaminated in the area of the landfill through contact with organic substances. These

waters were also significantly correlated with the leachate based on electrical conductivity, Cu and TOC indications. A significant correlation between these indicators and groundwater pollution in the landfill area was demonstrated by Aderemi et al. (2011) and Krčmar et al. (2018). Both Zn and Hg could also affect water pollution. Przydatek showed the deterioration of water quality in the landfill area as a result of elevated Zn concentrations by comparison with natural background levels (2021). The strongest correlation between the chemical composition of leachate samples and groundwater occurred in groundwater immediately downgradient of the landfill site (Boateng et al. 2019). This confirms that the spatial extent of groundwater hazard through the source of contamination from a landfill is intrinsically linked to the penetration of the liquid phase and the composition of leachates (Przydatek 2012). Przydatek and Kanownik (2019), based on an integrated model of water environment hazard with the use of electrical conductivity, TOC and Cd indicators, demonstrated that the course of the negative effect of a landfill site on groundwater and, indirectly, on surface water depends on local geological and hydrogeological conditions and processes taking place in the groundwater environment.

Another factor rarely used to identify the effect on the processes in a waste deposit is the ambient temperature. Research in this area was also conducted by Rowe and Arnepalli (2008), but they did not reach a binding conclusion in this respect.

A study of leachates from a landfill that has been in use for 80 years has shown that an interaction occurs between leachates and groundwater. Some researchers (e.g. Han et al. 2016) found that the most intense groundwater pollution occurs near landfills that are less than 20 years old. The reason for the negative influence could be the limited capacity of leachate treatment systems to treat the produced leachate. This appears to be especially the case after a period of over 20 years of use, which Rowe (2005) also noted.

Another reason for the deterioration could be the leachate leakage through the geomembrane due to manufacturing and construction defects or vapour diffusion through the lining (Pantini et al. 2014; Grugnaletti et al. 2016). The leakage could also cause negative impact on aquatic environments near the landfill, which can be affected by leachate, even from lined landfill facilities. The above-mentioned factors may also result from unintentional failures at the landfill (Ciuła et al. 2020).

The demonstrated pollutants migrating from the landfill to the groundwater may be present in the immediate vicinity or even up to hundreds of metres away (Abiriga et al. 2020). Koda (2009) demonstrated that the migration of pollutants depends on geological conditions and may favour the transport of water and the substances it carries or may stop the transport of water from the landfill site altogether.

# Conclusions

Based on an examines of the leachate composition and groundwater quality near a lined landfill during eight years of its use, the following conclusions were drawn:

- The analysis of the leachate composition and groundwater quality revealed a significant time variability of the physicochemical composition.
- Significant factors influencing the deterioration of the groundwater quality downgradient of the landfill included: (1) more than threefold increase in TOC concentration, which generally qualified the water as being of poor quality; (2) a high electrical conductivity value reaching 8.190 µS/cm; (3) a high Cu concentration, which were statistically significantly higher than in groundwater hydraulically upgradient of the landfill site; and (4) upward concentration trend in groundwater from inflow to outflow in concentrations of EC, TOC and Cu.
- A significant correlation between TOC, electrical conductivity, Zn, Cu, Pb, Hg and PAHs between leachate samples from the landfill and groundwater in the area is the result of the movement of leaching of contaminants into groundwater in the aeration zone.
- The progressing ageing process of the landfilled waste was indicated by low concentrations of heavy metals (Hg, Cu, Cd, Pb and Zn) and their decreasing trend.

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# Declarations

Conflict of interest Not applicable.

Ethics approval Not applicable.

Consent to Participate Not applicable.

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