

Article

Groundwater and Human Health Risk Assessment in the Vicinity of a Municipal Waste Landfill in Tychy, Poland

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Abstract: Groundwater quality and human health assessment in the vicinity of landfills can be performed with the use of numerous index methods. The aim of this paper is to present the results of the Environmental Risk Assessment (ERA) and Health Risk Assessment (HRA) in the vicinity of a municipal landfill complex for monitoring data from 1995, 2003, 2010, and 2021 and in the context of average statistical data about Poles. The calculations take into account an extended range of parameters, including sulphates, chlorides, and ammonium ions. The calculation results for the Horizontal ratio indicate that it should not be used for all parameters. This was mainly reflected in the low sulfate content of the water monitored by a piezometer directly below the old landfill. Other indicators, reaching as high as around 2000 (the Nemerow Pollution Index) or approx. 18,000 (the enrichment factor), confirm the negative impact of the landfill. The Hazard Index values reached almost 700, which would indicate a high risk to human health when consuming water with similar parameters. Overall, the results illustrate that using the selected indices to assess groundwater risk can be a valuable method for supporting long-term observations of groundwater quality, which can be used to make predictions using artificial intelligence methods.

Keywords: landfill; hydrogeology; risk; pollution indices; public health; Tychy; Poland



Citation: Dąbrowska, D.; Witkowski, A.J. Groundwater and Human Health Risk Assessment in the Vicinity of a Municipal Waste Landfill in Tychy, Poland. *Appl. Sci.* **2022**, *12*, 12898. <https://doi.org/10.3390/app122412898>

Academic Editors: Cheng-Yu Ku and Mauro Marini

Received: 27 October 2022

Accepted: 13 December 2022

Published: 15 December 2022

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1. Introduction

For economic reasons, a landfill is one of the most common ways of neutralizing municipal waste [1,2]. However, it is a real environmental problem due to the amount of deposited waste, migration of pollutants, and the possibility of fires [3–5]. Due to economic development and population growth, waste production continues to increase [6]. About 1/3 of the approximately 2 billion tons of municipal waste produced annually is inadequately managed [7]. According to a forecast by the World Bank, in the next three years, the global production of solid waste in cities will increase to as much as 6,100,000 tons per day [8,9]. Groundwater risk assessment is an effective way to protect groundwater resources and should be performed on the basis of the results of groundwater monitoring. The question remains, however, what methods can be used to support monitoring studies so that risk assessment is as effective as possible.

The current legal conditions regulate the design, construction, and operation of waste landfills, as well as groundwater monitoring systems and leachate drainage systems (Journal of Laws 2021, item 673, Regulation of the Minister of Climate and Environment of March 19, 2021, amending the ordinance on landfills). However, the proposed range of the parameters and the number of observation points in the context of a reliable risk assessment for groundwater and human health may not be sufficient [10]. The minimum range of field and laboratory tests is limited to specific EC, pH, total organic carbon (TOC), Cu^{2+} , Zn^{2+} , Pb^{2+} , Hg^{2+} , Cd^{2+} , Cr^{6+} , and PAHs.

The assessment of metal contamination in groundwater is one of the more widely discussed topics [11–15], especially in the case of landfills in developing countries [16]. Additionally, the impact of the metal pollution of groundwater on human health has been

assessed in many papers [17–19] due to the fact that even small concentrations of metals have negative and long-lasting effects. Negative effects on human health due to the type of metal and the amount of pollution can manifest as neurological problems, kidney disease, and an increased risk of cancer [20–22].

In the case of municipal landfills, it is necessary to pay attention to the monitoring of the main ions in groundwater [23,24]. Chloride content in groundwater which does not exceed 250 mg/L is not regarded as an impurity [25]. However, groundwater and soil may contain increased chloride and sulphate concentrations as a result of the migration of pollutants from landfill leachate [26,27]. In the case of water in the vicinity of landfills, the values of chlorides can reach 2000 mg/L [28].

There are a number of methods which can be used to assess the risk of groundwater pollution, such as estimation methods, parameter methods, ranking methods, hydrogeochemical modeling, and artificial neural networks. In the case of areas where the constant monitoring of changes in physicochemical parameters in groundwater is carried out, artificial intelligence methods can be used to assess the water quality and risk along with providing a forecast of changes [29,30]. However, in areas where monitoring is carried out in accordance with the legal requirements or with an extended range of parameters, methods using indicators can be applied. There are a number of indicators that may be helpful in assessing the quality of groundwater in the vicinity of pollution sources [31–36]. Many of the proposed indicators have been specifically designed to assess the metal content of water. Examples of these include the heavy metal pollution index and the metal pollution index [37–39]. In studies devoted to the impact of landfill leachate, it is extremely important to pay attention to the selection of the parameters for the assessment of water quality [40,41]. However, the formulae used to calculate the indicators are constructed in such a way that in almost every case it is possible to change the range of the parameters and calculate the indicator based on another set [42].

The aim of this paper is to present the results of the Environmental Risk Assessment (ERA) and Health Risk Assessment (HRA) in the vicinity of a municipal landfill complex in Tychy (Southern Poland) using groundwater monitoring data from 1995–2021. A set of data including metals (Pb, Cd, Ni, Cu, Fe, and Zn) and showing the standard approach to the subject of pollution indicators as well as a set of selected parameters (chlorides, sulphates, and NH_4) was used in this study in order to emphasize the significance of the risks that are ignored when assessing the quality of groundwater based on legal regulations. The values obtained in this work were also compared to other studies conducted in Poland and to examples of the use of the discussed indicators in the case of landfills around the world.

2. Materials and Methods

2.1. Study Area

The municipal waste landfill in Tychy is divided into two parts: a closed and reclaimed landfill (I), and the currently operating landfill sites ((II), (III)) along with the surrounding infrastructure. It covers an area of 12.7 ha (Figure 1).

The disused landfill, with an area of 3.5 ha, was used before 1988 as a landfill for building materials and later transformed into a municipal landfill. The landfill was closed due to the lack of an appropriate lining system to prevent the migration of pollutants [10]. The active part of the landfill consists of two sites, which were built in the years 1994–2004.

The landfill complex has been monitored since 1995. Originally, the observation network monitoring the Quaternary aquifer consisted of 14 piezometers (P1–P14) located at the front of the old and new municipal landfills. In the following years, this network was systematically rebuilt. In 2000, 2 piezometers (P15 and P16) were made, located north of the landfills in the zone of groundwater inflow (up gradient). In the following years, the expansion of the active system and the accompanying infrastructure resulted in the liquidation of some of the piezometers (P5, P6, P7, P8, P11, P12, P13, and P14) and the drilling of 2 new single piezometers (P5 and P18), as well as two pairs of nested piezometers measuring the upper (P17 and P19) and lower parts of the aquifer (P19 and P19A) (Figure 1).

In 2011, 3 piezometers were removed: P11, P12, and P13. In 2017, the P8 piezometer was destroyed [43]. Currently, the observation network for local groundwater monitoring of these landfills consists of 14 piezometers: P1, P2, P3, P4, P5', P9, P10, P15, P16, P17, P17A, P18, P19, and P19A (Figure 1).

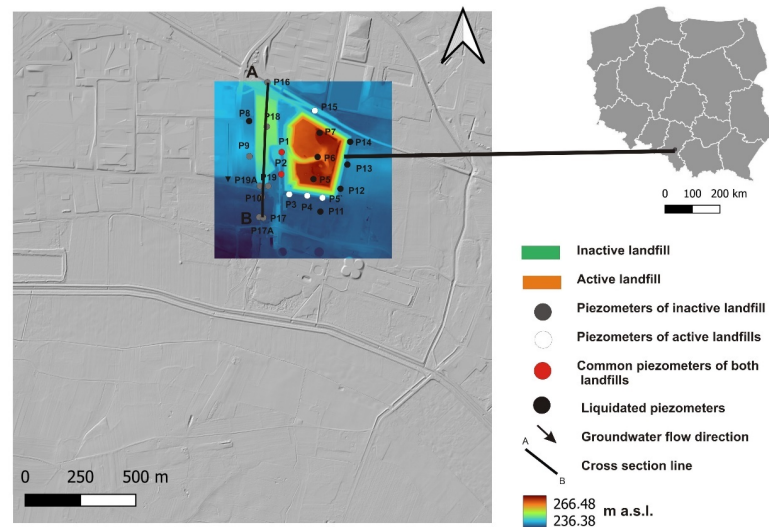


Figure 1. Location of the research area along with the observation network system.

The municipal landfills are located within the Fore-Carpathian depression in the central part of the Upper Silesian Coal Basin. The upper part of the Upper Carboniferous in this region contains layers from the Upper Silesian sandstone series. Locally, on the Upper Carboniferous, there are eroded patches of Triassic sediments [44]. Carboniferous and Triassic sediment are covered by thick (up to 80 m) Miocene clay formations. On the Miocene, there are Quaternary sediments (sands, gravels, and clays) with a thickness below 17.0 m [45] (Figure 2).

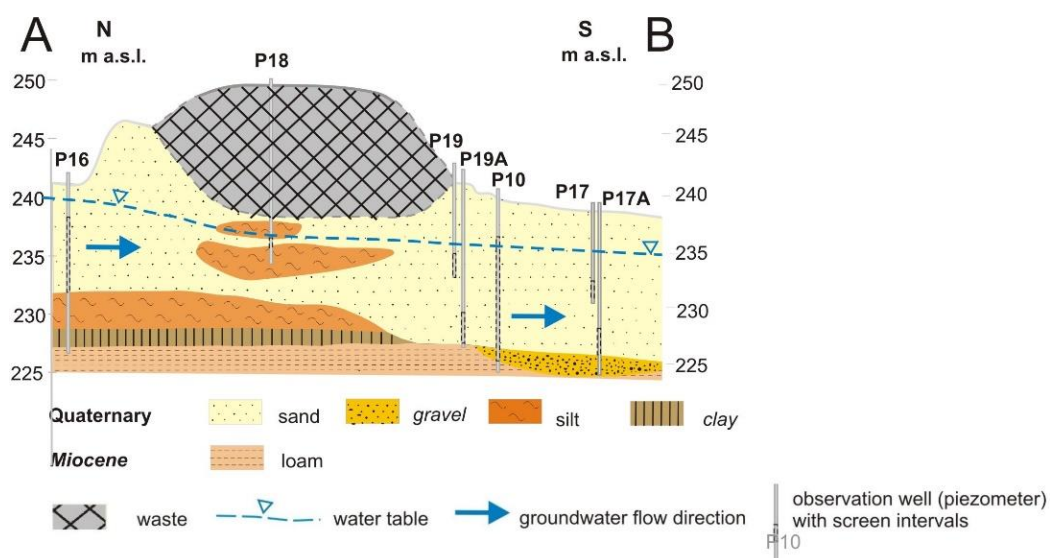


Figure 2. Geological cross-section of the study area (based on [10]).

The hydrogeological profile consists of three aquifers—Quaternary (Pleistocene), Triassic (locally), and Carboniferous. Locally, groundwater exists in the interbedding of the Miocene sediments. The Quaternary aquifer is locally separated by poorly permeable sediments. The lower aquifers are protected by very poorly permeable Miocene layers of a thickness of 80 m [46]. The groundwater flows south in the research area (Figure 3).

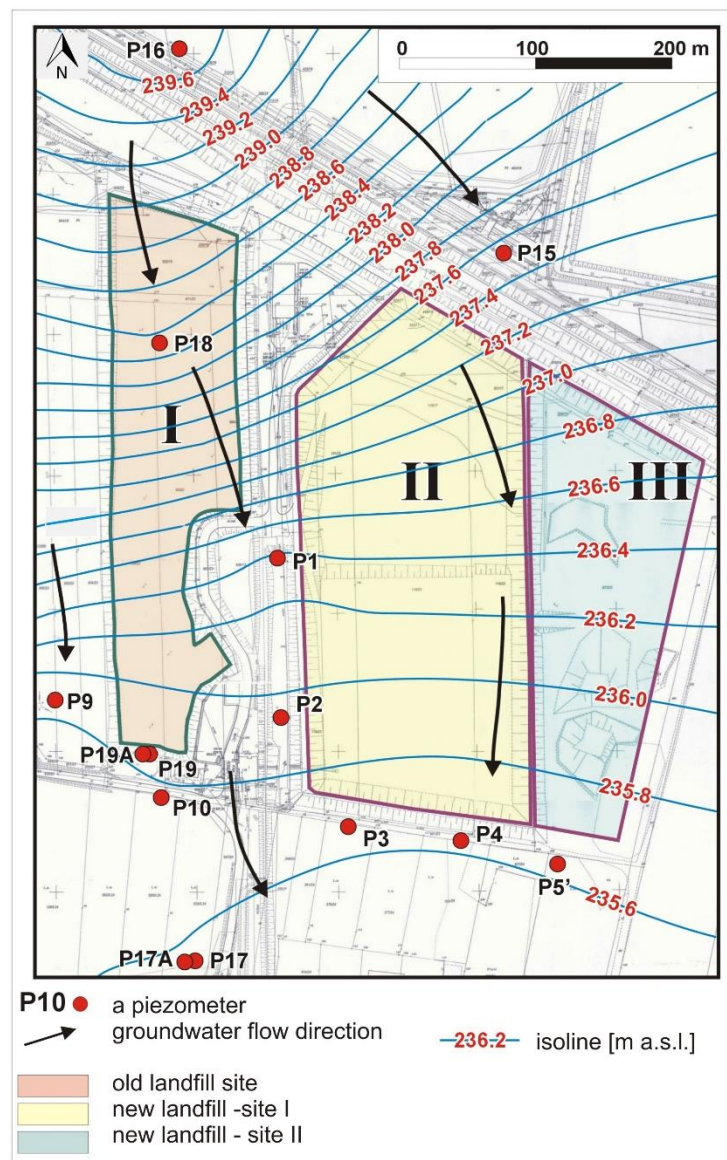


Figure 3. Contour map of the study area (based on [44]).

2.2. Methodology

The Environmental Risk Assessment (ERA) methodology was used to evaluate the environmental risks associated with the chosen metals and other pollution components in the groundwater in the vicinity of the municipal landfills in Tychy. Data from the monitoring of groundwater quality near pollution sources from 1995–2021 were analyzed. The years 1995, 2003, 2010, and 2021 were selected from this period. The choice of the years for which the assessment was carried out was not accidental:

- 1995 was the year that the monitoring system started;
- 2003 was the last year before the activation of the first site of the new landfill;
- 2010 was the year with the largest number of monitoring points near the landfills;
- 2021 was the last year for which year-round research is available.

The following parameters were used in the analysis: Pb, Cd, Ni, Cu, Fe, Zn, and the content of chlorides, sulphates, and NH_4 . The research took into account the data from all existing piezometers located in the observation network around the inactive waste landfill in a given year. The results for the individual pairs of P17/P17A and P19/P19A nested piezometers were both presented. Three indicators were calculated under the ERA, i.e., the Horizontal ratio [17], the Nemerow Pollution, and the enrichment factor [47].

Due to the fact that the long-term monitoring of groundwater quality in the vicinity of an active landfill does not indicate water pollution, and that the landfill has a liner system, the analyses did not take into account the data from piezometers belonging to the observation network of this facility.

The values of individual components in the water were assessed using the Horizontal ratio (H_R) in the direction of groundwater flow at a certain distance from the P16 piezometer (located up stream) to the individual piezometers from the observation network of the closed landfill, i.e., P1, P2, P8, P9, P10, P17, P18, and P19 piezometers.

The Horizontal ratio was calculated using the following formula:

$$H_R = \frac{C_{fA}}{C_{fB}} \tag{1}$$

where:

C_{fA} —is the concentration of the parameter on site A (in this case, the value from the P16 piezometer).

C_{fB} —is the concentration of the parameter on site B.

An index value greater than 1 for a single parameter suggests that the water would be treated after passing through the landfill, an index value of less than 1 for a single parameter suggests an impact of landfill leachate on groundwater, and a value of 1 suggests no impact. The article takes into account the total values of the index. If more parameters are taken into account, each result should be multiplied by the number of parameters.

The Nemerow Pollution Index (NPI) is a measure that takes into account the relationship between the measured value of a given parameter and the limit value [48]. The value of this index was calculated using the formula:

$$NPI = \frac{C_i}{L_i} \tag{2}$$

where:

C_i —is the measured value of the i th parameter.

L_i —is the allowable limit of the i th parameter.

The values for groundwater quality class III were adopted as the limit value for the good state of water quality on the basis of the Regulation of the Minister of Maritime Economy and Inland Navigation of 11 October 2019, on the criteria and method of assessing the state of groundwater bodies (Journal of Laws 2019 item 2148). As L_i in the formula above, the following values were adopted (Table 1):

Table 1. Allowable limits of particular parameters.

Parameter	L_i Value [mg/L]
Pb	0.100
Cd	0.005
Ni	0.020
Cu	0.200
Fe	5.000
Zn	1.000
Cl	250.000
SO4	250.000
NH4	1.500

An index value < 1 suggests low contamination, a value between 1 and 3 suggests moderate contamination, a value between 3 and 6 indicates considerable contamination, and a value > 6 indicates very high contamination [49].

The enrichment factor (EF) is another measure that takes into account the relationship between the measured concentration of a given parameter, but this time in relation to the

concentration of the immobile parameter. This factor was calculated using the following formula:

$$EF = \frac{\frac{CF_m}{c_{mi}}}{\frac{CF_b}{c_{bi}}} \quad (3)$$

where:

numerator—is the ratio of heavy metal to immobile element in the analyzed sample.

denominator—is the ratio of heavy metal to immobile element in the background sample.

An index value < 2 suggests minimal enrichment, values between 2 and 5 indicate moderate enrichment, values between 5 and 20 suggest significant enrichment, values between 20 and 40 represent very high enrichment, and a value greater than 40 suggests extremely high enrichment. In various studies, the immobile parameter was taken into account, for example, Al, Fe, Me, Mn, Sc, and Ti [50,51]. Iron was used as the immobile parameter [52] in this study. The choice of iron as an immobile component was dictated by the fact that it was measured in all piezometers throughout the observation period.

The second part of the analysis included the determination of health risk assessment. The risk assessment methodology was prepared based on the guidelines of the US Environmental Protection Agency [53]. For this purpose, the formula for calculating the average daily dose through the ingesting pathway (ADD) [54] was used. ADD was calculated from the following formula:

$$ADD = \frac{C \cdot ING \cdot EF \cdot ED}{BW \cdot AT} \cdot 10^{-6} \quad (4)$$

where:

C—is the concentration of the particular parameter in water ($\mu\text{g}/\text{L}$).

ING—is the ingestion rate (mL/day).

EF—is the exposure frequency in days/years.

ED—is the exposure duration in years.

BW—is the body weight (kg).

AT—is the average exposure time in days.

It should be noted that for the analyzed area of research, only an analysis of drinking water was carried out. Statistical data for Poland was used in the analysis. It was assumed that the inhabitants of the city of Tychy would use only groundwater from this area. The data for the P16 piezometer (located at the inflow of water to the landfill) and for the P9 and P10 piezometers (located at the outflow) were taken into account. The choice of these piezometers was dictated by the fact that, in their technical design, the filter is present on the entire thickness of the aquifer [10].

The input data for the calculation were assumed to be an average female weight of 70 kg and an average male weight of 85 kg [55]. It was also assumed that a woman drinks an average of 1.6 L of water, and a man 2 L per day. These numbers were averaged, and the ING value was 1800 mL and the BW value was 77.5 kg. It was assumed that a person lives an average of 80 years, consumes water with these properties, and drinks it 365 days a year.

The above equation was used to calculate the A Hazard Quotient (HQ). This is an indicator that uses the relationship between ADD and Reference Dose (RfD), i.e., the maximum permissible daily exposure level for humans [56].

$$HQ = \frac{ADD}{RfD} \quad (5)$$

RfD values were determined based on the Integrated Risk Information System (IRIS) of the U.S. EPA. Such data do not exist for all the parameters described. There are no such values for chlorides and sulfates. The remaining values are summarized in Table 2.

Table 2. RfD values for analyzed parameters [56].

Parameter	RfD [mg/kg/day]
Pb	14
Cd	0.5
Ni	20
Cu	40
Fe	0.7
Zn	300
NH4	0.1

Values of this indicator which do not exceed 1 prove that there is no risk. Cumulative risk resulting from exposure to several parameters is found by adding the HQ values to obtain a Hazard Index HI [57].

All the monitoring results taken into account were taken from annual reports. Chemical analyses were performed in accredited laboratories. Measurement uncertainty was determined for each measurement, and the percentage error was calculated for each analysis, taking into account the ion balance.

The described research methods have been used by various research centers around the world [47–52]. Some of the presented formulae have only been applied to sediments or soils.

3. Results and Discussion

The newest results of the monitoring carried out since 1995 indicate that the closed municipal waste landfill in Tychy remains a source of groundwater pollution. Electrical conductivity (EC) is a parameter used to indirectly assess the mineralization of water. In shallow groundwater exposed to anthropogenic pollution, an EC value of over 1000 $\mu\text{S}/\text{cm}$ indicates water pollution [58]. Leakage from municipal landfills has an EC of up to approx. 20,000 $\mu\text{S}/\text{cm}$. The EC of the tested samples in 2021 was within a wide range, from 543 $\mu\text{S}/\text{cm}$ (in October in P2) to 29,900 $\mu\text{S}/\text{cm}$ (in June in P18) [44]. In 2021, the water with the lowest mineralization was recorded in the P2 piezometer (366–396 mg/L), located east of the landfill, and in P16 (707–623 mg/L), located north of the landfill in the water inflow zone. The most highly mineralized water (>2000 mg/L) occurred under the landfill (P18) and in its southern foreland in piezometers P19A and P17A (measuring the bottom of the aquifer), in P17 (measuring the top of the aquifer), and in P10, on water outflow.

The highest pH in 2021 was found in the most heavily polluted water under the landfill (in the P18 piezometer—7.94) and in its immediate vicinity in the direction of water outflow. The lowest pH values were found east of the landfill (5.67 in P2). Current research results indicate the permanent or periodic occurrence of transitional conditions in water in the vast majority of the tested piezometers, taking into account the results of Eh. In the analyzed water, the Eh value ranged from 49 mvolts in P19 and 52 mvolts in P19A, to 339 mvolts in P2, and up to 312 mvolts in P16.

The total organic carbon (TOC) content in uncontaminated groundwater usually does not exceed 10 mg/L [59]. In the studied water, these values range from 2.9–3.4 mg C/L (in P2) and 5.1–5.9 mg C/L (in P16) to 1400–1917 mg C/L in P18. High concentrations of TOC were also found in P19A (81–100 mg C/L), in P19 (50–53 mg C/L), in P10 (up to 44–48 mg C/L), in P17A (up to 36–43 mg C/L), and in P1 (32–35 mg C/L).

The results of research from 2021 in the area clearly show increased concentrations of ammonium ions (NH_4^+). The content of this ion varied from <0.13 mg/L periodically in the P16 piezometer to an extremely high value of 2282 mg/L in the water sampled from the P18 piezometer. Increased concentrations of these ions were recorded in practically all the tested piezometers. High values were recorded in P10 (89–142 mg/L), in P1 (19–56 mg/L), and in nested piezometers monitoring deep parts of the aquifer, i.e., in P17A (146–152 mg/L) and in P19A (89–226 mg/L). Lower concentrations were recorded in P16 and in the P2 piezometer.

The nitrate content was significantly lower and ranged from <0.89 mg/L in P1, P10, P18, P19, and P19A to 56–62 mg/L in P17, and up to 55–62 mg/L in P2. High levels of nitrates were recorded in the water in the P16 piezometer (20 mg/L). Extremely high levels of these ions were found in piezometers representing oxidizing groundwater conditions: in P2 (55–62 mg/L) and in P17 (56–62 mg/L). The highest Eh values, a high content of sulphates, and the lowest TOC content and amount of ammonium ions were also found in these piezometers.

Chloride content in the tested water varied from 32mg/L in P16 and 42 mg/L in P2 to 4760 mg/L in P18. Concentrations above 1000 mg/L were not found in any piezometer (apart from P18). Values greater than 100 mg/L were recorded in the vast majority of the piezometers (except for P2, P16, and P19). In four of the piezometers, apart from the already mentioned P18 (in P1, P10, P17, and P17A), the chloride content exceeded 500 mg/L.

The content of sulphates in the monitored water varied within a range from <10.0mg/L in P18 and P19, to 168–190 mg/L in P1, and 170 mg/L in P17. Anomalously elevated values (over 100 mg/L) were also found in the P2, P16, and P17A piezometers.

In the monitored groundwater, apart from nickel and occasionally cadmium and lead, practically no significantly elevated concentrations of heavy metals were found. The vast majority of measurements of these metals (except nickel) indicated their very low concentrations (practically below the limits of their quantification). However, elevated nickel concentrations were still recorded (up to 0.69–0.75 mg/L in P18).

In the monitored water, significantly increased iron content (up to 36–74 mg/L) was also found in P1. Additionally, iron content greater than 10mg/L was found in the P18, P19, and P19A piezometers.

Increased manganese content was found in practically all the piezometers except for P16 (0.016–0.028 mg/L) and P18 (0.051–0.041 mg/L), and the maximum concentrations reached 6.0–8.6 mg/L in P1 and 7.3 mg/L in P10.

The results of physicochemical analyses from 2021 were taken into account in the overall water quality assessment based on the Regulation of the Minister of Maritime Economy and Inland Navigation of 11 October 2019, on the criteria and method of assessing the state of groundwater bodies. The results of the analyses made it possible to estimate that these are bodies of water of a poor chemical state (V class and in one case IV). Detailed information is provided in Table 3.

Table 3. Water quality classes determined on the basis of data from 2021.

Piezometer	Class	Factors Determining Belonging to the Class
P1	V	pH, EC, TOC, NH ₄ ⁺ , Fe, Mn, K, Cl ⁻
P2	V	pH, Mn
P9	V	pH, NH ₄ ⁺ , K, Mn
P10	V	EC, TOC, NH ₄ ⁺ , Na, K, Mn, B, Cl ⁻ , HCO ₃ ⁻
P16	IV	pH
P17	V	EC, TOC, Na, Cl ⁻
P17A	V	EC, TOC, NH ₄ ⁺ , Na, K, Mn, HCO ₃ ⁻ , Cl ⁻
P18	V	EC, TOC, NH ₄ ⁺ , Na, K, Fe, B, Ni, Cd, Cl ⁻ , HCO ₃
P19	V	TOC, NH ₄ ⁺ , K, Fe, Mn
P19A	V	EC, TOC, NH ₄ ⁺ , Na, K, Fe, B, HCO ₃ ⁻

Class IV is water of medium quality (the values of the water quality indicators increased as a result of natural processes and a weak anthropogenic impact), while Class V is water of poor quality (the values of the water quality indicators confirm an anthropogenic impact).

The results of the chemical analysis for the chosen four years in all of the samples are illustrated in Table 4. Concentrations of Pb, Cd, Ni, Cu, Fe, and Zn are given in µg/L, and those of Cl, SO₄, and NH₄ in mg/L. The table presents the averaged data for the selected four years for which the values of the individual indices were calculated. The average values of the individual parameters were calculated on the basis of three (in 1995) or two values measured in the piezometers in particular years.

Table 4. Chemical analysis results in 1995, 2003, 2010, and 2021.

	Year	Pb µg/L	Cd µg/L	Ni µg/L	Cu µg/L	Fe µg/L	Zn µg/L	Cl mg/L	SO ₄ mg/L	NH ₄ mg/L
P1	1995	40.0	4.0	50.0	10.0	4460.0	60.0	124.0	133.7	0.2
	2003	20.0	5.0	130.0	15.0	5860.0	90.0	376.0	195.0	1.4
	2010	10.0	1.0	4.0	4.0	34,450.0	5.0	260.0	220.0	27.5
	2021	4.0	5.0	48.0	4.0	55,000.0	32.0	772.0	179.0	56
P2	1995	410.0	5.0	37.0	18.0	2.0	87.0	106.3	165.7	0.0
	2003	20.0	5.0	27.0	20.0	64.0	13.0	95.5	129.4	0.0
	2010	10.0	1.0	4.0	6.0	177.0	5.0	125.4	165.0	13.8
	2021	4.0	5.0	17.0	7.0	250.0	45.0	42.0	115.0	1.8
P8	1995	20.0	5.0	10.0	7.0	31,100.0	60.0	41.2	128.0	1.3
	2003	20.0	5.0	10.0	20.0	22,010.0	10.0	35.5	116.4	0.1
	2010	10.0	1.0	4.0	4.0	13,300.0	39.0	87.2	120.0	0.4
	2021	-	-	-	-	-	-	-	-	-
P9	1995	20.0	5.0	40.0	10.0	54.0	65.0	88.0	92.3	0.3
	2003	20.0	5.0	15.0	18.0	243.0	12.0	129.7	87.0	0.1
	2010	10.0	1.0	5.0	5.0	170.0	36.0	65.6	130.0	18.9
	2021	4.0	5.0	48.0	7.0	7850.0	37.0	195.0	84.0	20.0
P10	1995	60.0	39.0	65.0	10.0	1070.0	89.0	769.7	271.3	41.4
	2003	20.0	5.0	68.0	30.0	22,600.0	13.0	783.8	143.8	282.9
	2010	10.0	1.0	4.0	4.0	860.0	8.0	341.0	65.0	79.6
	2021	4.0	5.0	41.0	4.0	8160.0	42.0	571.0	85.0	115.5
P16	1995	-	-	-	-	-	-	-	-	-
	2003	20.0	3.0	24.0	18.0	81.0	15.0	24.3	94.9	0.3
	2010	10.0	1.0	4.0	4.0	30.0	15.0	22.6	80.0	0.4
	2021	4.0	5.0	18.0	4.0	150.0	32.0	33.0	150.0	0.2
P17	1995	-	-	-	-	-	-	-	-	-
	2003	-	-	-	-	-	-	-	-	-
	2010	10.0	1.0	4.0	4.0	15,880.0	5.0	64.4	115.0	1.4
	2021	4.0	5.0	22.0	15.0	170.0	19.0	911.0	170.0	1.1
P17A	1995	-	-	-	-	-	-	-	-	-
	2003	-	-	-	-	-	-	-	-	-
	2010	10.0	1.0	13.0	4.0	994.0	5.0	967.5	76.0	311.8
	2021	4.0	5.0	70.0	7.0	4800.0	27.0	867.0	120.0	138.5
P18	1995	-	-	-	-	-	-	-	-	-
	2003	-	-	-	-	-	-	-	-	-
	2010	5.0	0.0	356.0	23.0	6170.0	361.0	5420.0	250.0	2994.0
	2021	4.0	13.0	720.0	120.0	18,500.0	1750.0	4485.0	10.0	2141.0
P19	1995	-	-	-	-	-	-	-	-	-
	2003	-	-	-	-	-	-	-	-	-
	2010	10.0	1.0	4.0	4.0	3525.0	5.0	135.5	14.5	20.2
	2021	4.0	5.0	13.0	6.0	17,000.0	28.0	53.0	10.0	16.5
P19A	1995	-	-	-	-	-	-	-	-	-
	2003	-	-	-	-	-	-	-	-	-
	2010	10.0	1.0	7.0	4.0	1345.0	5.0	553.0	97.0	1054.6
	2021	4.0	5.0	29.0	4.0	18,320.0	66.0	448.0	56.0	157.5

All indicator values presented in Figures 4–7 were calculated as sums of individual indicator values for individual parameters.

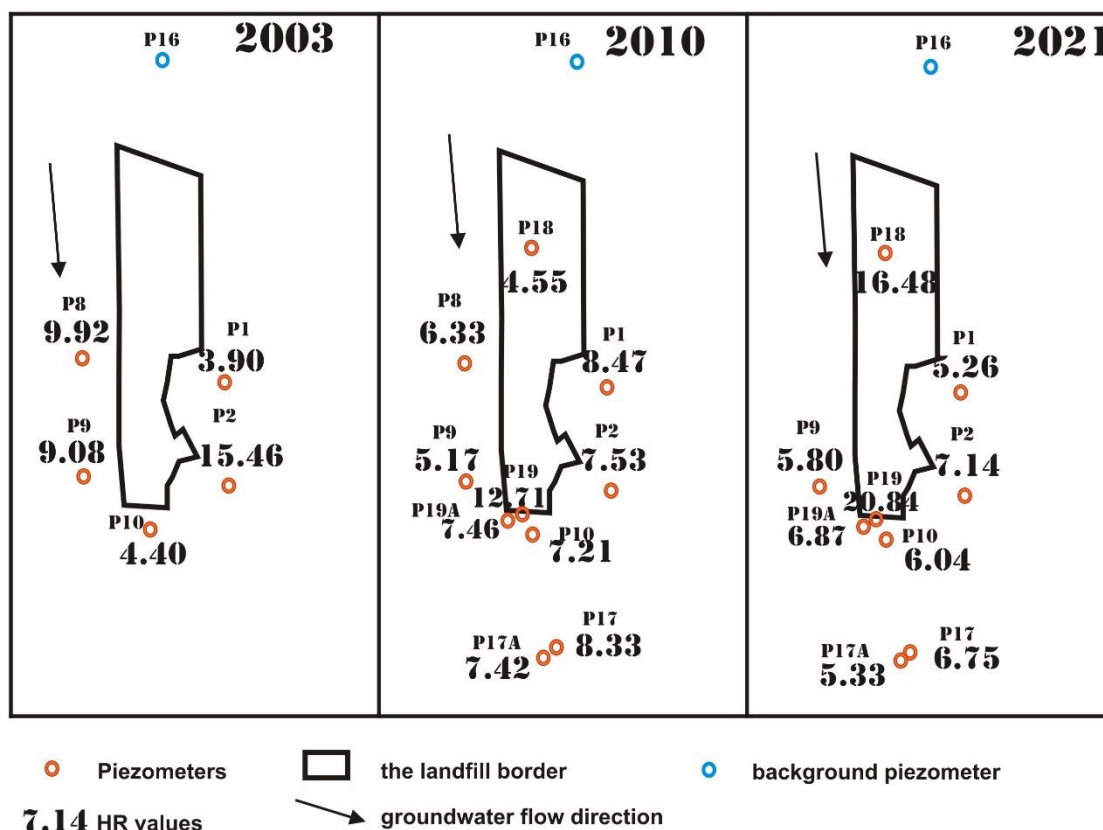


Figure 4. HR index values in years 2003, 2010, and 2021.

The values obtained for the HR index varied over the years. In 1995, it was not possible to calculate this index due to the absence of the P16 piezometer.

The values of the index calculated for the year 2003 would indicate that the water in all the piezometers was more polluted at the beginning of the system than after flowing under the landfill. This would suggest that the landfill had no impact on the water quality in the analyzed area, which is an incorrect conclusion when considering the groundwater monitoring results (Figure 4). The total (very high) value of the index was due to the identical (low) values of heavy metals in the water both in the P16 piezometer and in other piezometers belonging to the observation network, as well as in the increased concentration of chromium, which was as much as 12 times higher than the values measured in other piezometers.

However, the values of this index for 2010 indicate the impact of the landfill on groundwater. In one case (P19 piezometer), the value of the index was higher than 10, which is related to the low content of sulphates in the water of this piezometer.

The values obtained for the HR index indicate the negative impact of the landfill. However, as in the previous period, they were affected by the very low concentration of sulphates in the water of the P18 piezometer and the P19 piezometer. As a result, for this single component alone, the total index value increased to over 16, while the value of 15 was for the sulphates alone (Appendix A).

So far, this indicator has not been used for groundwater in Poland. In the case of studies conducted in the area of other landfills [17], higher values of the HR index were obtained. Values for a single component reached 2.5. This value was calculated for copper. The total value of the indicator obtained by the authors of the article on the landfill in Nigeria was about 10.

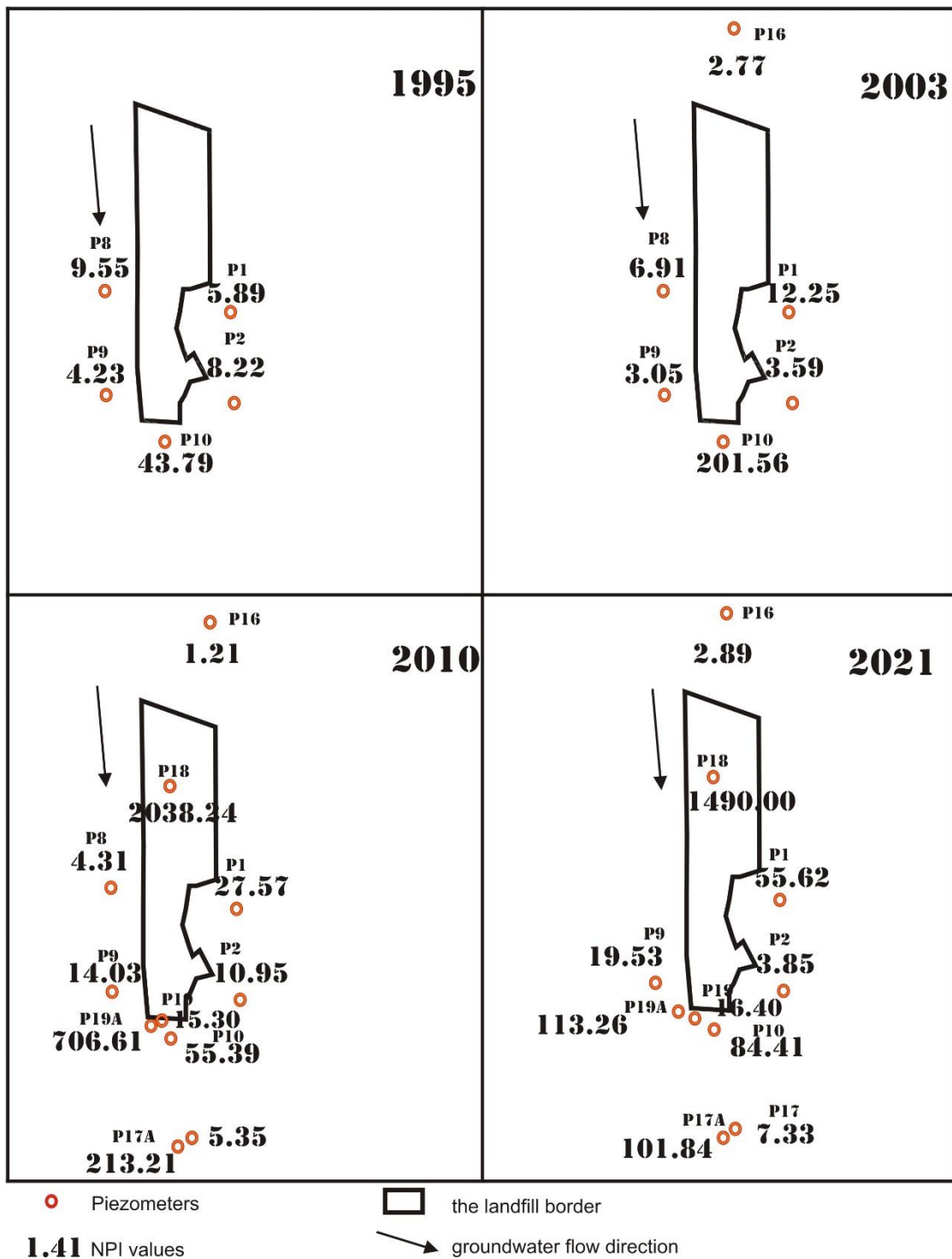


Figure 5. Nemerow Pollution Index values in 1995, 2003, 2010, and 2021.

The second selected index is the Nemerow Pollution Index. This indicator was calculated for 1995, 2003, 2010, and 2021 (Figure 5). The structure of the indicator, despite its simplicity, allows the degree of groundwater pollution in this area to be determined. The NPI values obtained for 1995 indicate that the downstream water of the piezometers was the most polluted. The calculated value for the P10 piezometer was more than five times that of the P2 piezometer.

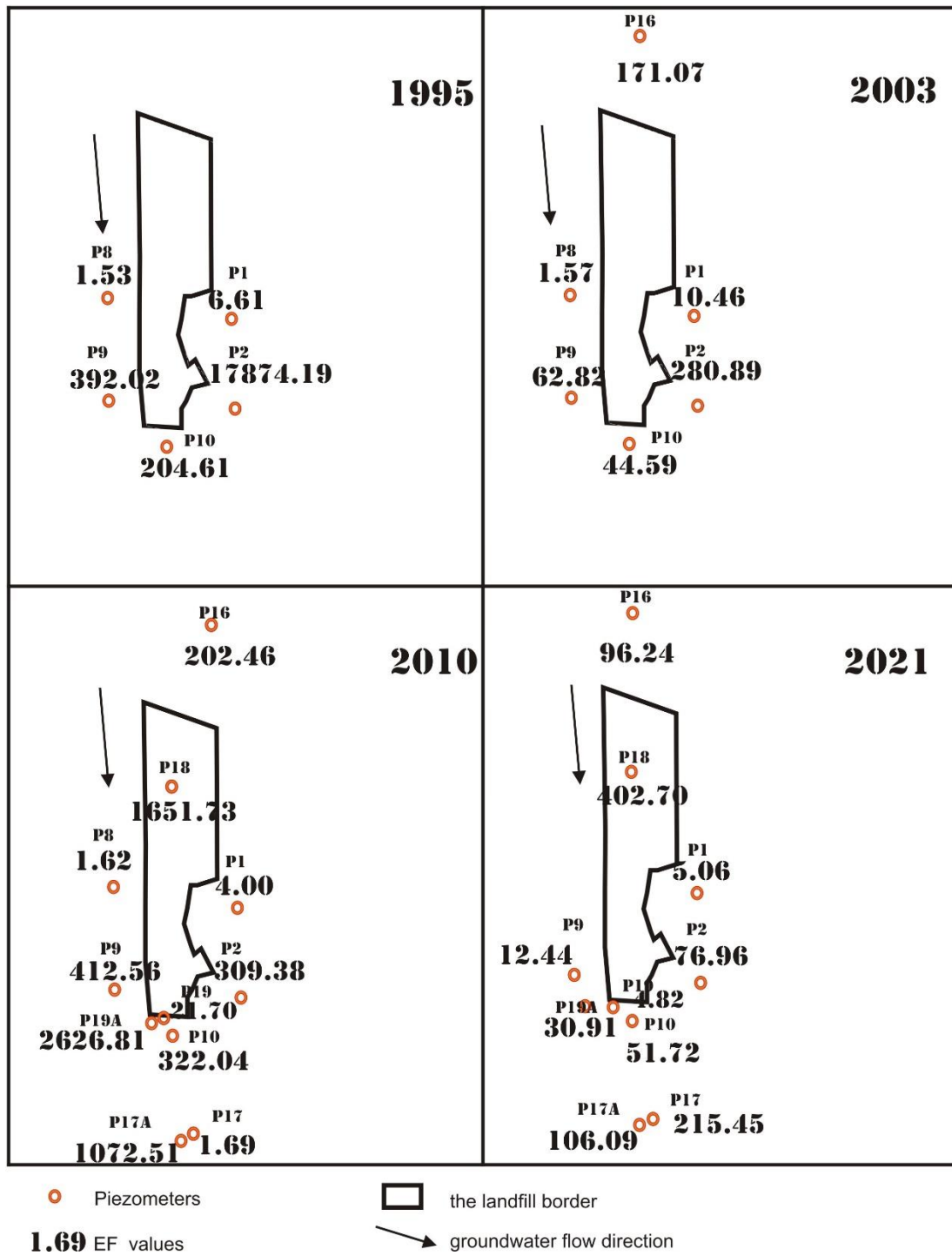


Figure 6. EF values in 1995, 2003, 2010, and 2021.

The following years in which the index was calculated made it possible to indicate the differences between the upstream piezometer (P16) and other localized piezometers. The year 2003 confirmed the further negative impact of the closed landfill. During this year, the value of the index for P10 was over 50 times higher than for the P2 piezometer.

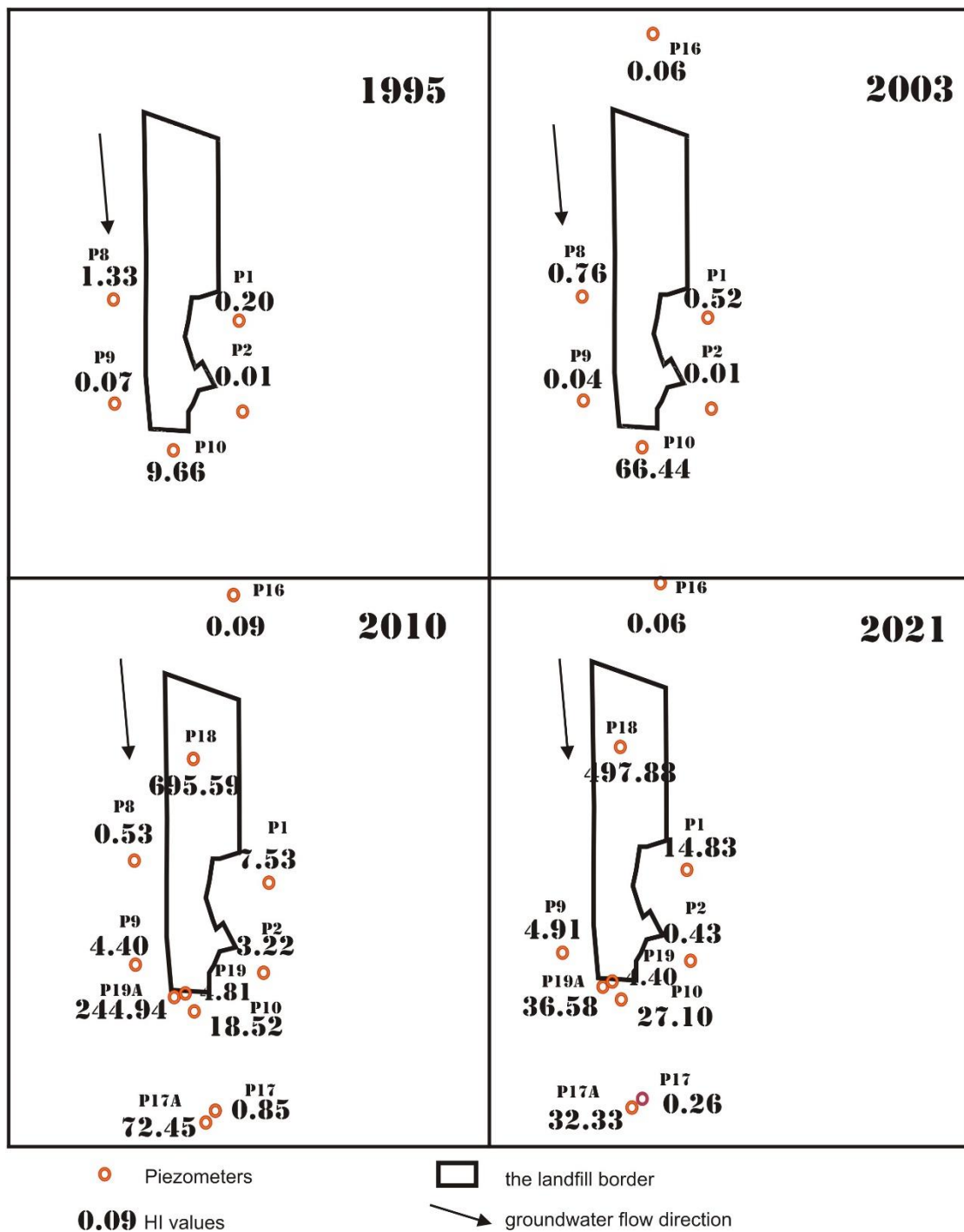


Figure 7. HI values in 1995, 2003, 2010, and 2021.

In 2010 and 2021, it was possible to determine the pollution both in the upper and lower parts of the aquifer due to measurements from the nest piezometers. The P18 piezometer also operated in the same years. The results for both years are very different. The values of the index suggest that the quality of water flowing into the landfill has deteriorated (the NPI value in this period doubled in P16). At the same time, the results for all the piezometers, except for the P18 piezometer and for the nest piezometers monitoring the lower part of the aquifer, deteriorated.

In 2010 and 2021, it was possible to determine the pollution both in the upper and lower parts of the aquifer due to measurements from the nest piezometers. The P18

piezometer also operated in the same years. The results for both years are very different. The values of the index suggest that the quality of water flowing into the landfill has deteriorated (the NPI value in this period doubled in P16). At the same time, the results for all the piezometers, except for the P1 and P18 piezometers and for the nest piezometers monitoring the lower part of the aquifer, deteriorated. A big change took place within the P19A piezometer. The value decreased by about six times between 2010 and 2021. At the same time, for the P17A piezometer, the index value was about two times lower.

Compared to other publications where the NPI was used [60–62], the obtained values are higher. This applies primarily to the water in piezometers P10, P17A, P18, and P19A. In the cited works, the index values for a single component rarely exceeded 1 when the main ions were not analyzed. In one work [61], values as high as those found in the landfill in Tychy were obtained for the ammonium ion. The Nemerow index was used in the work [63], which concerns a landfill located in north-eastern Poland (Podlasie Province). However, the authors of the paper used these other parameters to assess the value of the indicator: pH, electrolytic conductivity, total suspension, chemical oxygen demand, biochemical oxygen demand, total organic carbon, dissolved organic carbon, total nitrogen, total dissolved nitrogen, total phosphorus, and orthophosphates. In this article, high index values were obtained for chemical oxygen demand (about 20) and total organic carbon (5.81).

The enrichment factor (EF) values varied considerably in the individual measurement years. In 1995 and 2003, the values obtained in most of the piezometers are comparable. Very high values were obtained in the P2 piezometer—about 18,000 in 1995 and almost 300 in 2003 (Figure 6). Within these two measurement years, the value of the indicators for water in the P9 piezometer decreased by six times, by more than four times for the P10 piezometer, and by more than 60 times for the P2 piezometer.

Typically high values of this index were calculated for the P18 piezometer. It is surprising, however, that the value of the EF index in the P19A piezometer was higher than in P18 in 2010. Another surprising result is the higher HF value in the P17 piezometer than in P17A. Chlorides had a large share of the increased value of the index for the P17 piezometer in 2021. Chlorides contributed approximately 50% to the final index value. Another component that increases the values of this index for most parameters is the ammonium ion.

The enrichment factor has mainly been used for soil contamination [64–66]. For groundwater, it was used in one study [67]. The values of this indicator ranged from –61.2 to 43.42, suggesting a low risk. The index values for a single component in this article exceeded 4000. It is not possible to compare the obtained values of this indicator with that of other groundwater in the area of landfills in Poland, because it was only used to assess the quality of sediments.

The health risk assessment is one of the most important elements in the risk assessment in the vicinity of landfills. The Hazard Index (HI) is an effective tool for calculating the non-carcinogenic health risk in different intake forms [60]. The results calculated for non-carcinogenic health risks for people in the study area indicate the highest risk in the event of consumption of water from the area of the P18 piezometer and piezometers collecting water from the lower part of the aquifer (P17A and P19A), as well as from the area of the P10 piezometer (Figure 7).

It is worth noting that this index was calculated without taking into account sulphates or chlorides. In this situation, the high values of the index were mainly determined by the concentrations of ammonium, iron, and cadmium. For example, these three parameters constitute up to 99.9% of the index value in the P10 piezometer. The ammonium ion caused 99.97% of the high value of the indicator for the P18 piezometer. This proves that there is little risk from metals in this area.

HQ indices and the total value of indices (HI) have been used many times in various research centers [38,68–71]. The HQ indices varied widely, from about 0.001 to about 30 [72]. The authors identified a high risk of contamination with lead and copper. The total values

of the indicator reached about 90. These values were definitely lower than those obtained in the area of the described landfill in Tychy.

Much more often, health risk assessment is performed based on the quality of water or the emission of pollutants generated from waste. In Poland, a human health risk assessment was carried out in the vicinity of landfills, but it was a study based on the measurement of atmospheric pollution by gases generated as a result of a waste fire [73]. In addition, an assessment of human health was also carried out on the basis of data on the quality of groundwater, but in selected cities [74]. Health was also assessed on the basis of microbiological risk, e.g., in the work of Podlasek et al. [75].

4. Conclusions

In this study, 32 groundwater samples from 1995, 2003, 2010, and 2021 were analyzed for various physicochemical parameters to assess their quality using four indices, i.e., the Horizontal ratio (HR), the Nemerow Pollution Index (NPI), the enrichment factor (EF), and the Hazard Index (HI). The calculations include not only metals such as Pb, Cd, Ni, Cu, Fe, and Zn, but also chlorides, sulphates, and NH_4 . The values of the three additional parameters had a significant impact on the final values of the indicators.

Despite the fact that most of the existing indices are very helpful in assessing the risk to groundwater and human health in the vicinity of pollution sources, the improper selection of background parameters or piezometers as a reference can lead to misinterpretation of the situation. The long-term monitoring of groundwater in the vicinity of the closed landfill in Tychy clearly indicates the migration of leachate from the landfill and the contamination of groundwater in this area. However, if the results for HR are taken into account, it appears that the landfill is not the main pollution source. Performing the isolines with the use of software using algorithms indicates that the greatest pollution is generated in the P19 region.

In this study, the NPI turned out to be the least resistant to the choice of background, indicator parameter, or reference point. In this case, it seems most reasonable to use legal acts to determine the relationship between the individual parameter values measured in the piezometers and the limit values for groundwater quality class III.

The assessment of human health, even on the basis of general statistical data on Poles, indicates that consuming groundwater from this region exposes drinkers to the harmful effects of substances contained within it. This indicator also suggests that the high value depends primarily on the concentration of chloride and ammonium ions. From the point of view of the use of monitored groundwater as drinking water, the most important problem is the water quality in front of the landfill, i.e., in the outflow zone, e.g., in the vicinity of the P17 and P17A piezometers, which are located approximately 200 m from the landfill. In this context, it is worth mentioning the vertical differentiation of the flow of pollutants. Taking into account the HI index, the water from the shallow piezometer P17 indicates no risk (index < 1), and that of the deeper one (P17A) indicates high risk (index of several dozen). In this case, if we only had a single shallow piezometer (P17), we could conclude that the groundwater quality in this observation well is not negatively affected by the nearby landfill and could even be used as drinking water.

Due to the advantages of the selected proposed indices in groundwater risk assessment, it is suggested that the NPI index and artificial neural network methods be used for long-term observations in future studies on other landfills.

Author Contributions: Conceptualization, D.D. and A.J.W.; methodology, D.D.; writing—original draft preparation, D.D. and A.J.W.; writing—review and editing, D.D. and A.J.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Table A1. Partial values of HR, NPI and EF indices.

HR Index	Year	Pb	Cd	Ni	Cu	Fe	Zn	Cl	SO ₄	NH ₄	Total
P1	2003	1.00	0.60	0.18	1.20	0.01	0.17	0.06	0.49	0.19	3.9
	2010	1.00	2.00	1.00	1.00	0.00	3.00	0.09	0.36	0.01	8.46
	2021	1.00	1.00	0.38	1.00	0.00	1.00	0.04	0.84	0.00	5.26
P2	2003	1.00	0.60	0.89	0.90	1.27	1.15	0.25	0.73	8.67	15.46
	2010	1.00	1.00	1.00	0.67	0.17	3.00	0.18	0.48	0.03	7.53
	2021	1.00	1.00	1.06	0.57	0.60	0.71	0.79	1.30	0.11	7.14
P8	2003	1.00	0.60	2.40	0.90	0.00	1.50	0.69	0.82	2.02	9.93
	2010	1.00	1.00	1.00	1.00	0.00	0.38	0.26	0.67	1.01	6.32
P9	2003	1.00	0.60	1.60	1.00	0.33	1.25	0.19	1.09	2.02	9.08
	2010	1.00	1.00	0.80	0.80	0.18	0.42	0.34	0.62	0.02	5.18
	2021	1.00	1.00	0.38	0.57	0.02	0.86	0.17	1.79	0.01	5.8
P10	2003	1.00	0.60	0.35	0.60	0.00	1.15	0.03	0.66	0.00	4.39
	2010	1.00	1.00	1.00	1.00	0.03	1.88	0.07	1.23	0.00	7.21
	2021	1.00	1.00	0.44	1.00	0.02	0.76	0.06	1.76	0.00	6.04
P17	2010	1.00	1.00	1.00	1.00	0.00	3.00	0.35	0.70	0.28	8.33
	2021	1.00	1.00	0.82	0.27	0.88	1.68	0.04	0.88	0.18	6.75
P17A	2010	1.00	1.00	0.31	1.00	0.03	3.00	0.02	1.05	0.00	7.41
	2021	1.00	1.00	0.26	0.57	0.03	1.19	0.04	1.25	0.00	5.34
P18	2010	2.00	n/a	0.01	0.17	0.00	0.04	0.00	0.32	0.00	2.54
	2021	1.00	0.38	0.03	0.03	0.01	0.02	0.01	15.00	0.00	16.48
P19	2010	1.00	1.00	1.00	1.00	0.01	3.00	0.17	5.52	0.02	12.72
	2021	1.00	1.00	1.38	0.67	0.01	1.14	0.62	15.00	0.01	20.83
P19A	2010	1.00	1.00	0.57	1.00	0.02	3.00	0.04	0.82	0.00	7.45
	2021	1.00	1.00	0.62	1.00	0.01	0.48	0.07	2.68	0.00	6.86
NPI Index	Year	Pb	Cd	Ni	Cu	Fe	Zn	Cl	SO ₄	NH ₄	Total
P1	1995	0.40	0.80	2.50	0.05	0.89	0.06	0.50	0.53	0.16	5.89
	2003	0.20	1.00	6.50	0.08	1.17	0.09	1.50	0.78	0.93	12.25
	2010	0.10	0.10	0.20	0.02	6.89	0.01	1.04	0.88	18.33	27.57
	2021	0.04	1.00	2.40	0.02	11.00	0.03	3.09	0.72	37.33	55.63

Table A1. Cont.

HR Index	Year	Pb	Cd	Ni	Cu	Fe	Zn	Cl	SO ₄	NH ₄	Total
P2	1995	4.10	1.00	1.85	0.09	0.00	0.09	0.43	0.66	0.01	8.23
	2003	0.20	1.00	1.35	0.10	0.01	0.01	0.38	0.52	0.02	3.59
	2010	0.10	0.20	0.20	0.03	0.04	0.01	0.50	0.66	9.22	10.96
	2021	0.04	1.00	0.85	0.04	0.05	0.05	0.17	0.46	1.20	3.86
P8	1995	0.20	1.00	0.50	0.04	6.22	0.06	0.16	0.51	0.85	9.54
	2003	0.20	1.00	0.50	0.10	4.40	0.01	0.14	0.47	0.09	6.91
	2010	0.10	0.20	0.20	0.02	2.66	0.04	0.35	0.48	0.26	4.31
P9	1995	0.20	1.00	2.00	0.05	0.01	0.07	0.35	0.37	0.19	4.24
	2003	0.20	1.00	0.75	0.09	0.05	0.01	0.52	0.35	0.09	3.06
	2010	0.10	0.20	0.25	0.03	0.03	0.04	0.26	0.52	12.60	14.03
	2021	0.04	1.00	2.40	0.04	1.57	0.04	0.78	0.34	13.33	19.54
P10	1995	0.60	7.80	3.25	0.05	0.21	0.09	3.08	1.09	27.62	43.79
	2003	0.20	1.00	3.40	0.15	4.52	0.01	3.14	0.58	188.57	201.57
	2010	0.10	0.20	0.20	0.02	0.17	0.01	1.36	0.26	53.07	55.39
	2021	0.04	1.00	2.05	0.02	1.63	0.04	2.28	0.34	77.00	84.4
P16	2003	0.20	0.60	1.20	1.20	0.09	0.02	0.02	0.10	0.38	3.81
	2010	0.10	0.20	0.20	0.20	0.02	0.01	0.02	0.09	0.32	1.16
	2021	0.04	1.00	0.00	0.90	0.02	0.03	0.03	0.13	0.60	2.75
P17	2010	0.10	0.20	0.20	0.02	3.18	0.01	0.26	0.46	0.93	5.36
	2021	0.04	1.00	1.10	0.08	0.03	0.02	3.64	0.68	0.73	7.32
P17A	2010	0.10	0.20	0.65	0.02	0.20	0.01	3.87	0.30	207.87	213.22
	2021	0.04	1.00	3.50	0.04	0.96	0.03	3.47	0.48	92.33	101.85
P18	2010	0.05	0.00	17.80	0.12	1.23	0.36	21.68	1.00	1996.00	2038.24
	2021	0.04	2.60	36.00	0.60	3.70	1.75	17.94	0.04	1427.33	1490
P19	2010	0.10	0.20	0.20	0.02	0.71	0.01	0.54	0.06	13.47	15.31
	2021	0.04	1.00	0.65	0.03	3.40	0.03	0.21	0.04	11.00	16.4
P19A	2010	0.10	0.20	0.35	0.02	0.27	0.01	2.21	0.39	703.07	706.62
	2021	0.04	1.00	1.45	0.02	3.66	0.07	1.79	0.22	105.00	113.25
EF Index	Year	Pb	Cd	Ni	Cu	Fe	Zn	Cl	SO ₄	NH ₄	Total
P1	1995	0.45	0.90	2.80	0.06	1.00	0.07	0.56	0.60	0.18	6.62
	2003	0.17	0.85	5.55	0.06	1.00	0.08	1.28	0.67	0.80	10.46
	2010	0.01	0.01	0.03	0.00	1.00	0.00	0.15	0.13	2.66	3.99
	2021	0.00	0.09	0.22	0.00	1.00	0.00	0.28	0.07	3.39	5.05
P2	1995	8913.04	2173.91	4021.74	195.65	1.00	189.13	924.61	1440.61	14.49	17,874.18
	2003	15.63	78.13	105.47	7.81	1.00	1.02	29.84	40.44	1.56	280.9
	2010	2.82	5.65	5.65	0.85	1.00	0.14	14.17	18.64	260.45	309.37
	2021	0.80	20.00	17.00	0.70	1.00	0.90	3.36	9.20	24.00	76.96

Table A1. Cont.

HR Index	Year	Pb	Cd	Ni	Cu	Fe	Zn	Cl	SO ₄	NH ₄	Total
P8	1995	0.03	0.16	0.08	0.01	1.00	0.01	0.03	0.08	0.14	1.54
	2003	0.05	0.23	0.11	0.02	1.00	0.00	0.03	0.11	0.02	1.57
	2010	0.04	0.08	0.08	0.01	1.00	0.01	0.13	0.18	0.10	1.63
P9	1995	18.52	92.59	185.19	4.63	1.00	6.02	32.59	34.20	17.28	392.02
	2003	4.12	20.58	15.43	1.85	1.00	0.25	10.67	7.16	1.77	62.83
	2010	2.94	5.88	7.35	0.74	1.00	1.06	7.71	15.29	370.59	412.56
	2021	0.03	0.64	1.53	0.02	1.00	0.02	0.50	0.21	8.49	12.44
P10	1995	2.80	36.45	15.19	0.23	1.00	0.42	14.39	5.07	129.07	204.62
	2003	0.04	0.22	0.75	0.03	1.00	0.00	0.69	0.13	41.72	44.58
	2010	0.58	1.16	1.16	0.12	1.00	0.05	7.93	1.51	308.53	322.04
	2021	0.02	0.61	1.26	0.01	1.00	0.03	1.40	0.21	47.18	51.72
P16	2003	12.35	37.04	74.07	5.56	1.00	0.93	6.00	23.43	10.70	171.08
	2010	16.67	33.33	33.33	3.33	1.00	2.50	15.07	53.33	43.89	202.45
	2021	1.33	33.33	30.00	0.67	1.00	1.07	4.40	20.00	4.44	96.24
P17	2010	0.03	0.06	0.06	0.01	1.00	0.00	0.08	0.14	0.29	1.67
	2021	1.18	29.41	32.35	2.21	1.00	0.56	107.18	20.00	21.57	215.46
P17A	2010	0.50	1.01	3.27	0.10	1.00	0.03	19.47	1.53	1045.61	1072.52
	2021	0.04	1.04	3.65	0.04	1.00	0.03	3.61	0.50	96.18	106.09
P18	2010	0.04	0.00	14.42	0.09	1.00	0.29	17.57	0.81	1617.50	1651.72
	2021	0.01	0.70	9.73	0.16	1.00	0.47	4.85	0.01	385.77	402.7
P19	2010	0.14	0.28	0.28	0.03	1.00	0.01	0.77	0.08	19.10	21.69
	2021	0.01	0.29	0.19	0.01	1.00	0.01	0.06	0.01	3.24	4.82
P19A	2010	0.37	0.74	1.30	0.07	1.00	0.02	8.22	1.44	2613.63	2626.79
	2021	0.01	0.27	0.40	0.01	1.00	0.02	0.49	0.06	28.66	30.92

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