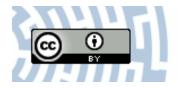


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Citation style: Dąbrowska Dominika, Witkowski Andrzej Jarosław, Sołtysiak Marek. (2018). Application of pollution indices for the assessment of the negative impact of a municipal landfill on groundwater (Tychy, southern Poland). "Geological Quarterly" (Vol. 62, iss. 3 (2018) s. 496-508), doi 10.7306/gq.1420



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Application of pollution indices for the spatiotemporal assessment of negative impact of a municipal landfill on groundwater (Tychy, southern Poland)

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Dąbrowska, D., Witkowski, J.A., Sołtysiak, M., 2018. Application of pollution indices for the spatiotemporal assessment of negative impact of a municipal landfill on groundwater (Tychy, southern Poland). Geological Quarterly, **62** (3): 496–508, doi: 10.7306/gq.1420

Associate editor: Stanisław Wołkowicz

Risk assessment plays an essential role in the protection of groundwater resources, especially in the regions of potential contamination sources. The present research was aimed at assessing the risk of groundwater contamination in the vicinity of the Tychy-Urbanowice municipal landfill systems (southern Poland), using the contamination index tool. Groundwater samples from the Quaternary Aquifer were collected from 22 piezometers located near the former landfill site (currently closed) and the new sealed landfill site during a monitoring period from 1995 to 2015. Extremely high values (1700) of the groundwater contamination index were observed in a piezometer that captures water beneath the abandoned landfill. Very high index values (154 or 216) were also noticed in piezometers located in the groundwater outflow from the landfill site. Results of interpolation for individual piezometers, which capture only the upper or lower part of an aquifer, are unrepresentative. The interpolation for the mean values of the contamination index increased the groundwater risk assessment.

Key words: hydrogeology, groundwater risk assessment, contamination index, municipal landfill site, Tychy-Urbanowice.

INTRODUCTION

Unlined landfills, located on permeable and vulnerable ground, are usually a dangerous source of groundwater contamination. Pollution indicators characteristic of municipal waste landfills include, among others, increased electrical conductivity (EC) and higher concentrations of Cl⁻, SO₄²⁻, N, B⁺, Cr⁶⁺, Cu²⁺, Zn²⁺, Pb²⁺, Hg²⁺, Co³⁺ and Ni²⁺ (Clark and Piskin, 1977; Bojakowska, 1994; Slack et al., 2005). The current legal arrangements, i.e. Council Directive 1999/31/EC of 26 April 1999 on waste landfill, and the Polish Regulation of the Minister of the Environment of 30 April 2013 (Journal of Laws, No. 523) on landfill, impose an obligation to conduct groundwater monitoring around waste landfill, as well as specify the minimum number of observation points and their location in the hydrodynamic system (at least one observation point at the groundwater inflow into the landfill, and at least two monitoring points located at the outflow), and the sampling frequency and minimum range of the field and laboratory tests in relation to specific EC, pH, total organic carbon (TOC), Cu²⁺, Zn²⁺, Pb²⁺, Hg²⁺, Cd²⁺, Cr⁺⁶ and polycyclic aromatic hydrocarbons (PAHs).

In the area of pollution sources, it is necessary to conduct a reliable groundwater quality monitoring that allows both a pro-

per assessment of the impact of the parameters on the ground-water and the development of a plan to counteract the negative effects of pollution (Hakanson, 1980; Grath et al., 2001; Jousma and Roelofsen, 2004; Nielsen, 2006; Quevauviller et al., 2009). Given that the regulations governing the groundwater monitoring around landfills have been undergoing transformations for many years, along with several changes in both the exploitation of local groundwater monitoring networks (LGWMNs) and in their structure (e.g., several optimizations, replacements of contractors carrying out field and laboratory work, with the criterion being the lowest offer), a comprehensive assessment of the impact of waste landfills on groundwater over protracted time intervals is often impossible.

The assessment of the impact of pollution source on ground-water can be carried out with the use of predictive models (Li et al., 2012), monitoring or geophysical tests (Singh et al., 2008), research based on an isotopic multi-tracer approach, and laboratory tests (Weber et al., 2002; Soltysiak, 2007, 2009). The most commonly used method is an analysis of temporal and spatial variability of the individual components of groundwater, based on the results of monitoring tests (Vilomet et al., 2001, 2003).

A synthetic assessment of the impact of pollution source on groundwater in a specific area can be carried out using pollution indices (Soltan, 1999; Singh et al., 2015). The most common measures determining the level of groundwater pollution comprise the heavy metal pollution index (Mohan et al., 1996), metal pollution index (Tamasi and Cini, 2004), Nemerow index comprehensive evaluation method, and C_{fi} contamination index (Backman et al., 1998).

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The water quality index (Brown, 1970) and the application of the inverse distance-weighted method to spatial interpolation (Selvam et al., 2015) use the information on water quality trends and compare to the general public or to the policy makers. Therefore, it was decided to use the $C_{\rm d}$ contamination index (Backman et al., 1998) to assess the groundwater chemical composition changes in the area of Tychy-Urbanowice municipal landfills.

This index allows the possibility to assess comprehensively both spatial and temporal changes in contamination based on a single cumulative index ($C_{\rm d}$), and the partial indices calculated for the individual parameters ($C_{\rm f}$) included in the cumulative index. This is ensured by calculating the index value for individual piezometers and then interpolating the obtained values for the analysed area of research. Such maps can be made for different years of operation of the monitoring network. An important argument for using this method, which is dimensionless, is that it makes it easy to compare different water components.

The aim of the study was to present changes in the values of contamination index in selected years for the operation of the monitoring network and to assess the negative impact of the old landfill on groundwater. Additionally, the lack of piezometers at the inflow of groundwater to the landfill and the lack of data from the location of the landfill site affect the reliable assessment of groundwater quality in the region was also considered. For this purpose, the index calculation was made via a simulation of the existence of the aforementioned observation points, assuming the interpolation of contour line distribution in 1995. Moreover, the interpolation of the contamination index was carried out based on the results from nested piezometers, under three different scenarios (respectively, the results from the piezometers installed in the upper part of the aquifer, in its lower part, and the mean results from two pairs of the piezometer nest).

In order to calculate the index of groundwater contamination, five parameters were utilized: EC, SO₄²⁻, Cl⁻ and HCO₃⁻, and the sum of N-NO₃, N-NO₂ and N-NH₄.

STUDY AREA AND DATA

The study deals with groundwater in the neighbourhood of an unlined municipal landfill located in the southern part of the town of Tychy, in the Urbanowice District, southern Poland. The waste used to be stored in an old sand excavation pit, directly on natural ground. Since 1984, 0.6 million tons of municipal waste, dominated by food waste, paper, organic and inorganic residue and glass (Rosik-Dulewska, 2005), have been stored over an area of 3.5 ha. The thickness of deposited waste varied between 6 and 12 m. Due to the negative impact on groundwater, the landfill was abandoned in 1995 and has been systematically monitored since then. The landfill's reclamation started in 1996 and was completed in 1997 (Witkowski, 2015). The top of the landfill, with an area of ~35,000 m², was covered with a plastic film and soil.

Between 1994 and 2004, two new lined sites were designated near the abandoned landfill. Both new sites had separate leachate drainage systems directed by a common collector to the neighbouring waste water treatment plant (Dąbrowska, 2012). The area housing the entire complex of landfills, along with the accompanying facilities, covers 12.7 ha (Fig. 1; Dąbrowska et al., 2016). There are also other pollution sources in this region – a sewage treatment plant located to the south of the described landfills and a landfill owned by one of the automotive concerns located north-east of the landfills.

Groundwater in the area of these landfills has been monitored for more than 20 years. The original Local Groundwater Monitoring Network (LGWMN) consisting of 14 piezometers (from P1 to P14; Fig. 1) was constructed in 1995. In subsequent years, the network has been supplemented with seven new piezometers:

- piezometers P15, P16 were installed in the upgradient zone of the landfill area;
- piezometer P18 was placed on the top of the abandoned landfill:
- two pairs of nested piezometers (P17 and P17A, and P19 and P19A) were located in separate boreholes, with P17 and P19 measuring the top, and P17A and P19A measuring the bottom of the aguifer.

At the same time, seven piezometers were systematically liquidated (P5, P6, P7, P11, P12, P13 and P14; Fig. 1). The sampling frequency was also changed. Currently, 15 piezometers, including the two pairs of nested piezometers, is being sampled in the area of the complex of landfills (Dąbrowska et al., 2016). Since 1998, the groundwater has been monitored twice a year in the old landfill area. At present, the sampling is continued on piezometers P1, P2, P8, P9, P10, P16, P17, P17A, P18, P19 and P19A. The active landfill is monitored quarterly, currently through the sampling of piezometers P1, P2, P3, P4, P5 and P15.

The monitored complex of landfills is located in the central part of the Upper Silesian Coal Basin, within the Fore-Carpathian depression filled with Miocene sandstone formations. The Quaternary formations occurring in the vicinity of the landfills are represented by Pleistocene fluvial deposits: variously grained sands, rarely gravels and clays. The thickness of the deposits is in the range from 12.5 to 17.0 m (Witkowski and Żurek, 2007).

In the region of the complex of landfills, there are three aquifers of Quaternary, Triassic and Carboniferous origin. Directly beneath the landfills, there is the shallow sandy Quaternary Aquifer, which is ~10 m thick and practically not insulated from the ground surface. Locally, this aquifer is divided by poorly permeable clay-dust formations in two separate, but hydrodynamically connected layers. The two-layer system occurring within the Quaternary Aquifer is very important from the point of view of the spatial distribution of groundwater pollution in the area of the monitored landfills. The Quaternary Aquifer is insulated from the lower-lying water-bearing horizons (Triassic and Carboniferous) by a complex of poorly permeable Miocene clay formations with a thickness of ~80 m (Fig. 2). Groundwater from the Quaternary Aquifer generally flows southward (Fig. 2) towards the Gostynia River, which is the local discharge base for 95% of groundwater flowing out from the landfill area (Sitek et al., 2010). The water table within this aguifer is mostly unconfined; according to the measurement results from 2015, it is located at a depth between ~2.8 m near the P15 piezometer and ~8.3 m near the P8 piezometer (Witkowski, 2015). Locally, the water table is confined; the static water level in the P18 piezometer, located on the top of the inactive landfill, is at a depth of 12.3 m (Fig. 2). The seasonal fluctuations of the groundwater table are small, 30 cm on average.

METHODS

To evaluate the variability of temporal and spatial changes in chemical composition of groundwater in the area of the complex of the municipal waste landfills in Tychy-Urbanowice, the

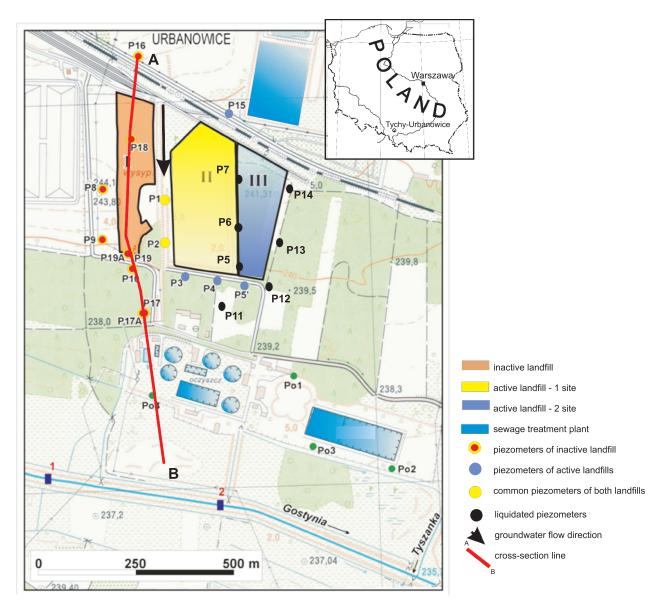


Fig. 1. The system of landfills in Tychy-Urbanowice with the groundwater monitoring network

Cd contamination index was used. This index was first introduced by Backman et al. (1998), and is a measure of the amount of groundwater pollution in relation to the natural hydrochemical background (Backman et al., 1998; Dragon, 2004).

The groundwater contamination index (C_d) in the area of the described parameters has been calculated based on the elemental formula (Bhuiyan et al., 2016):

$$C_{d} = \sum_{C_{Ni}}^{n} C_{fi}$$
 [1

where:

$$C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1$$
 [2]

in which $C_{\rm fi}$ is the contamination index for the i-th parameter; $C_{\rm Ai}$ is the analytical value for the i-th parameter; $C_{\rm Ni}$ is the upper range of the values of the natural hydrochemical background.

In order to select parameters for calculating the contamination index, principal component analysis (PCA) (Abdi and Williams, 2010) was adopted and carried out in respect of the following 14 parameters: 1) SO_4^{2-} , 2) NO_4^{4-} , 3) EC, 4) CI^- , 5) NO_2^{-} , 6) HCO_3^{-} , 7) NO_3^{-} , 8) N, 9) Zn^{2+} , 10) Cd^{2+} , 11) Cu^{2+} , 12) Pb^{2+} , 13) Fe^{2+} , 14) Cr^{6+} . Due to the availability of data (resulting from changing legal regulations, and various monitoring teams), the choice of other parameters, such as TOC, was impossible. In the current research area, the first seven parameters were chosen taking into account results of PCA analysis, because they explain the highest variance in the data set. However, because the range of the parameter values changed both in time and in the samples from particular piezometers, the final contamination index was calculated based on the following parameters: specific EC, SO₄², Cl⁻ and HCO₃, and the total of N-NO₃, N-NO₂ and N-NH₄. What posed a serious problem was the changing number of points over time in the monitoring network, and the irregular frequency of their sampling.

The values of hydrochemical background of groundwater was determined as the mean arithmetic values of the measurements of physicochemical properties, carried out on 15 samples

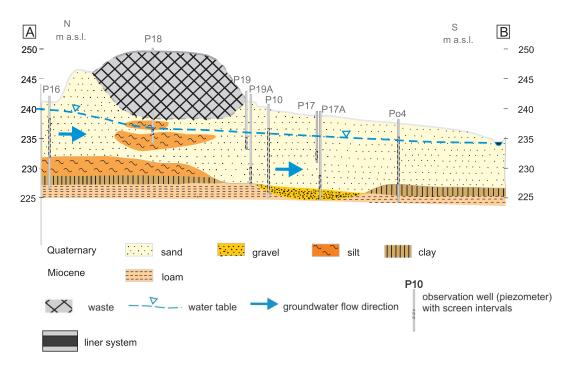


Fig. 2. Schematic hydrogeological cross-section A-B from Figure 1

collected between 1995 and 2000 from the P12 piezometer. It is among the most representative piezometers due to the construction and position in relation to the hydrodynamic system. The analysis of the hydrodynamic system for the study area shows that contaminated water did not flow into this piezometer (Witkowski, 2001). A choice of 15 samples was dictated by the fact that modernization of the monitoring network and changes in the chemical composition of groundwater started in 2000. The tested values of individual parameters have normal distribution (based on Lilliefors test) with the significance correction value >0.2. Finally, the following values of the background were adopted to calculate the contamination index: EC: 240.1 μ S/cm; total sum of N-NH4+N-NO2+N-NO3: 2.7 mg/dm³; CI: 10.3 mg/dm³; SO42-: 66.9 mg/dm³; and HCO3: 38.5 mg/dm³.

The presentation of the indices on the map enabled the assessment of changes in the chemical composition of groundwater in the area of the examined parameters. Maps presenting the distribution of the contamination index for the individual parameters also allow for the instant identification of higher concentrations of the pollutants and their possible spreading (Stigter et al., 2006). The contamination index value increases with the increasing concentration of individual parameters in groundwater. In the case of heavily transformed areas, the water pollution index involves high values. It is understood that, for highly industrialized areas, the index value exceeds 3 (Brraich and Jangu, 2015). The calculation of the indices for each of the five selected parameters ($C_{\rm fi}$) and the total contamination index ($C_{\rm d}$) has been made based on the results of groundwater monitoring carried out between 1995 and 2015.

To interpolate the index values, the ordinary kriging method was employed (Boissonnat and Cazals, 2001). Interpolations of the contamination indices were first made using only the kriging method in the *Surfer* software (Golden Software v14, 2017). After that, the results were corrected taking into consideration hydrogeological knowledge, and fitted to the actual hydrodynamical conditions in the analysed area. The correction concerned the areas for which the algorithm would indicate other

indicator values than those obtained from the research. However, the used method was the most representative among tested triangulation, radial base function, and the nearest neighborhood algorithm.

To illustrate the data representativeness, three variants of the contamination index distribution in the zone of the groundwater outflow from the landfill area have been presented: in the upper part of the aquifer, in the bottom part of the aquifer, and when the results from the two pairs of piezometer nests are involved.

RESULTS

The calculation of the partial contamination index ($C_{\rm fi}$) for EC, SO_4^{2-} , Cl⁻ and HCO $_3^{-}$, and the total of N-NO $_3$, N-NO $_2$ and N-NH $_4$ (N $_{\rm in}$) for the samples taken around the complex of the landfills in 1995–2015, confirmed the negative impact of the old landfill on groundwater. The values of partial contamination indices for these parameters vary widely from –1.0 to 1139.88 (Table 1). These are also the highest and most varied values for N $_{\rm in}$, while the lowest values and also less variable are noticed for sulphates (from –0.98 to 9.1). All groundwater contamination index scores computed for EC, SO_4^{2-} , Cl⁻, HCO $_3^{-}$ and sum of N-NO $_3$, N-NO $_2$, N-NH $_4$ (N $_{\rm in}$) are presented in Appendix 1*.

Significant spatial differences for particular indices have been noticed. The values of contamination indices are up to several hundred times higher for the groundwater in the piezometers of the observation network of the old landfill (P10, P17, P17A, P18, P19, P19A) than those of the new landfill (P3, P4, P5', P11–P14; Appendix 1). This is particularly true for the index of the total amount of mineral forms of nitrogen (N_{in}), whose values ranged from -1 in P12 (monitoring the new landfill; no impact) to 1,139.88 in P18 (monitoring the water beneath the old unlined landfill), and 519.38 in P19A (monitoring the water at the bottom of the aquifer in the southern foreland of the old

landfill, i.e. the water outflow zone). Significantly higher values of the N_{in} contamination index were also observed in other piezometers monitoring the zone of groundwater outflow from the old landfill area (P10: 114.86 – Appendix 1; and P17A: 92.55; Fig. 3A). Nitrogen compounds are a characteristic indicator of the impact of municipal landfill sites on groundwater.

One of the basic parameters indicating groundwater pollution is EC. The background value for this parameter near the landfill was determined at 240 μ S/cm. Directly below the landfill, EC ranged from 8.556 to 37.500 μ S/cm, whereby the contamination index value for this parameter was in the range of 34.63–155.17 (an average of 128.36). High values of this partial contamination index were also determined for samples taken from the southern front of the landfill, i.e. from piezometers P10, P19A and P17A (maxima of 42.06, 22.89 and 27.67, respectively; Appendix 1 and Fig. 3B).

The EC value primarily depends on both the concentration of the main ions and the load of organic compounds. In this paper, only the first factor was taken into account. A higher concentration of chloride ions is an indicator of pollutants near the municipal landfill. The highest concentrations of chlorides were found in samples from the P18 piezometer, up to 6,630 mg/dm³, while the background concentration is ~10 mg/dm³. This state is reflected in the values of the contamination index calculated for chlorides, which range from 390 to 640 with a mean value of 487. The increased values of the chloride index were also determined in samples taken from the southern front of the landfill, i.e. from piezometers P19A, P10 and P17A (Appendix 1 and Fig. 3C). A similar situation was documented for bicarbonates, whose elevated concentrations (18060, 2721 and 2879) and high contamination index values (468, 67, 74) were also found in water samples taken from piezometers P18, P19A and P17A (Fig. 3D).

Extremely high values (EC - 155, N_{in} - 1140, Cl - 640, HCO₃ - 468) of almost all of the analysed contamination indices (excluding SO₄²⁻) were noted in the waters beneath the landfills (P18). The Eh mean value measured in this piezometer in 2015 was 66 (Witkowski, 2015). These waters prove a reducing environment, wherein the processes of denitrification and sulphate removal could take place. It is evidenced by both the relatively low value of the contamination index for sulphates (max. 2.74) and the very high value of the index for the total amount of mineral forms of nitrogen (max. 1139.88), which is determined by the extremely high content of ammonia nitrogen (to 3,091.126 mg/dm³) despite low concentrations of nitrates (5.69 mg/dm³).

The results of the measurements performed during this period showed no significant changes in the groundwater condition in the area, suggesting the absence of the impact of pollution sources on the groundwater monitored by this piezometer. Piezometers P16 and P15, located to the north of the landfills, in the groundwater inflow zone, were drilled later in the period concerned (no earlier than 2000).

The maximum, minimum and mean values of the partial contamination indices for individual parameters ($C_{\rm fi}$) of the analysed period are presented in Table 1. For the chosen piezometers located along the flow line (including nested piezometers installed within the upper or lower zones of the aquifer), these values are presented in Figure 3. The corresponding values for the total index ($C_{\rm d}$) for all piezometers are shown in Table 2, while the spatial distribution of the total values of the contamination indices is presented on multivariate maps (Figs. 4–6).

The distribution of analysed partial indices in relation to the distance from the landfill and the depth in nested piezometers is also noteworthy. It is evident that there are generally higher values of all indices for deeper piezometers (except for sulphates in P17A) (Fig. 3A-E). Furthermore, an increasing trend in the indices for EC, chlorides and sulphates, with regard to the distance from the landfill for both shallow (P19 and P17) and deeper (P19A and P17A) piezometers is presented (Fig. 3C-E). The same trend is observed for bicarbonates in deeper piezometers (P19A and P17A), but there is a slight decreasing trend in the shallow ones (P19 and P17). A different situation is presented in respect of the mineral form of nitrogen (Fig. 3A), where a decreasing trend with increasing distance from the landfill is presented in both shallow (P19 and P17) and deeper (P19A and P17A) piezometers (Fig. 3A). Such distribution of this index resulted in ammonification and denitrification processes occurring in the landfill (P18), denitrification and ammonification processes in the deep part of the aquifer (P19A and P17A), and nitrification processes occurring in the unsaturated zone and in the upper part of the aquifer, observed with an increasing distance from the landfill (P19 and P17).

Between 1995 and 2015, the value of the total contamination index of the groundwater (C_d) ranged from –1.85 in P6 (currently liquidated) to 2,336.64 in P18 (monitoring the water beneath the old landfill) (Table 2). The mean values of the contamination index for the piezometers monitoring groundwater in the area of the new landfill did not exceed 38, while those for the inactive landfill (at the side of the P18 piezometer) were 311.84 (in P19A).

Table 1

Extreme and mean values of the partial contamination indices (Cfi) for selected components in 1995–2015 for piezometers involved in the monitoring network in Tychy

Component	Max C _{fi}	Min C _{fi}	Mean C _{fi}	Number of samples	
EC	155.17	-0.99	9.21	951	
N _{in}	1139.88	-1	51.26	527	
Cl	640.82	-0.8	36.32	697	
SO ₄ ²⁻	9.1	-0.98	0.74	697	
HCO ₃	467.73	-1	24.94	817	

^{*} Supplementary data associated with this article can be found, in the online version, at doi: 10.7306/gq.1420

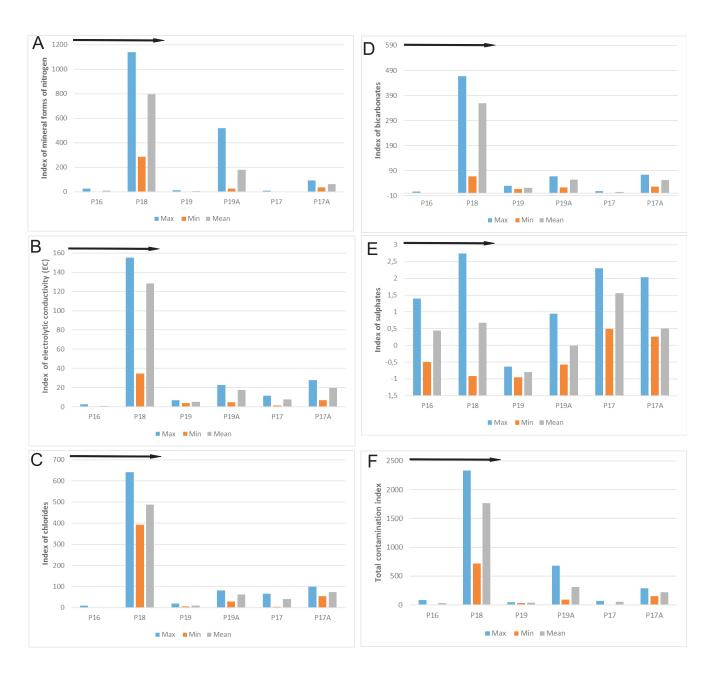


Fig. 3A–F – changes in the values of partial contamination indices (C_{fi}) and total contamination index (C_d) in selected piezometers located along the groundwater pathway, 1995–2015, Tychy-Urbanowice landfills, southern Poland

As with the previously mentioned partial indices, the highest values of the total contamination index (C_d) were presented in the groundwater beneath the old landfill (P18) and in the zone of water outflow from the area of the landfills, particularly in the nested piezometers monitoring the lower part of the aquifer (P17A and P19A) (Fig. 3F above, Table 2). It should be noted that the mean value of the contamination index for the P18 piezometer (1767.47) is almost six times higher than the highest mean value for the other piezometers (311.84 in P19A) (Table 2). The lowest values of the index were observed for the piezometers installed to the east (P13) and to the south (P3, P4, P5, P11) of the new landfill and within the groundwater inflow into the area from the north (P15).

INTERPRETATION

A complex analysis of Figure 3A–E shows that the contamination plume from the landfill moves to the south, and then plunges down in line with the distance from the landfill. The increasing trend among nested piezometers P19, P19A and P17, P17A, located along the groundwater flow direction, indicate natural attenuation processes occurring in the landfill, leading to a reduction in the pollution load produced by the monitored landfill. For example, the presented highest contamination index for sulphates in the shallow piezometer P17 (Fig. 3E) could be the result of sulphide oxidation or an additional source of sulphates in that area (infiltration of contaminated water from surface runoff).

Table 2

Extreme and mean values of the contamination index (C_d), 1995–2015, Tychy-Urbanowice landfills, southern Poland

Piezometer	C _d min value	Year of detection	C _d max value	Year of detection	C _d mean value	Number of samples
P1	12.03	1995	71.01	2015	37.44	36
P2	6.66	2001	48.03	2008	19.85	36
P3	0.02	2005	25.53	2001	9.10	37
P4	-0.65	2000	44.40	2005	9.67	37
P5	-1.73	2000	1.72	1995	0.08	12
P5`	-1.26	2004	14.00	2013	3.59	6
P6	-1.85	1999	3.64	1996	1.73	8
P7	2.89	1996	13.18	1998	8.22	13
P8	2.72	2002	14.51	2011	6.50	36
P9	1.56	2001	81.33	2014	27.14	36
P10	50.54	1996	329.27	2000	153.69	38
P11	-1.36	2001	2.75	2003	-0.04	40
P12	-1.62	1998	4.82	1996	1.22	20
P13	1.89	2003	12.65	1998	8.56	20
P14	7.04	2000	12.82	1998	9.57	14
P15	-0.05	2005	15.51	2003	7.44	11
P16	5.20	2000	82.31	2006	31.47	39
P17	5.29	2010	70.20	2012	54.98	16
P17A	147.8	2015	285.60	2010	215.65	16
P18	720.08	2007	2336.64	2010	1767.47	16
P19	32.45	2014	46.17	2012	38.55	16
P19A	88.85	2008	682.31	2011	311.84	16

The high value of the contamination index for the P16 piezometer, installed at the groundwater inflow into the landfill area, results from local groundwater contamination due to nitrogen compounds and sulphates, associated with both intense agriculture and a large animal shelter located nearby.

It should be noted that spatial distribution of the total contamination index of groundwater (C_d) is mostly influenced by the partial index for N_{in} . Both indices show the same pattern of spatial distribution (Fig. 3A, F).

All calculations of the contamination index were performed for the average annual values of $C_{\rm d}$ at each piezometer, which is associated with a more representative spatial distribution of the index. The temporal changes in the spatial distribution of the contamination index values in the selected five years (1995, 2003, 2007, 2010 and 2015) are shown in Figures 4 and 5. These years were chosen purposely for the following reasons:

- 1995 was the year of monitoring start, with the old landfill being the only one that existed at that time;
- 2003 was the last year before the activation of the first site of the new landfill, while modification of the observation network was also carried out that year;
- 2007 is when the construction of the second site of the new landfill was started, while another modification to the monitoring network was made;
- 2010 saw the largest number of monitoring points before the last reorganization;

 2015 was the final year of the research based on the current/last modification of the observation network.

It should be noted, however, that the number of piezometers used for the interpretation of spatial variability of the pollution index was changeable throughout these years. The numbers of active piezometers in the particular years were as follows: in 1995 - 14 (P1 to P14); in 2003 - 14 (P1-P4, P5', P8-P16); in 2007 - 16 (P1-P4, P5', P8-P13, P15-P17, P17A, P18); in 2010 – 18 (P1–P4, P5', P8–P13, P15–P17, P17A, P18, P19 and P19A); and, in 2015, after the last modification, the observation network comprised 15 piezometers (P1-P4, P5', P8-P10, P15-P17, P17A, P18, P19, P19A). Therefore, a reliable determination of the temporal variation of the spatial distribution of the index in the study area is rather difficult. Especially problematic was the building up of a reliable picture of the situation during the initial period, i.e. in 1995, when the monitoring network was smaller and did not include piezometers P15 and P16 (groundwater inflow zone), piezometer P18 (in the landfill area), nor nested piezometers P17, P17A and P19, P19A (in the groundwater outflow zone from the landfill site).

Against this backdrop, an attempt was made to verify the representativeness of the adopted method of interpolation for the results of the chemical analyses of 1995. The analysis was performed for three different variants of the spatial distribution of the pollution index, as illustrated in Figure 4.

In the first variant (Fig. 4A), the map of the distribution of the groundwater contamination index around the described param-

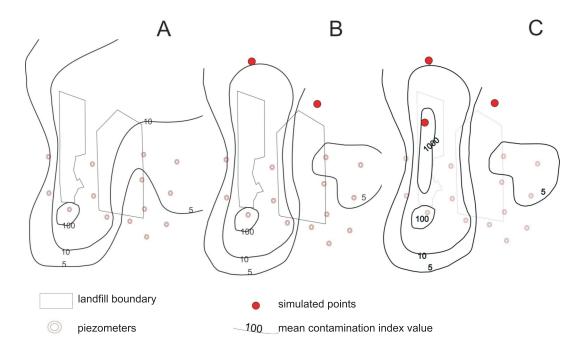


Fig. 4. Spatial changes of the contamination index value (C_d) in 1995 for different configurations of the monitoring network, Tychy-Urbanowice landfills, southern Poland

A – interpolation for the observation network existing in 1995; **B** – interpolation with a simulation of the existence of two piezometers located in the water inflow zone; **C** – interpolation with a simulation of the existence of two piezometers as in the Figure 4B and a monitoring point at the top of the disused landfill

eters was made solely by applying the interpolation method (kriging) to the results from the existing observation network, consisting of 14 piezometers. The resulting pattern of the index distribution indicates the eastward spreading of the pollution cloud. However, this direction is inconsistent with the groundwater flow direction in this region (north to south). Another argument for the rejection of such an interpolation is the fact that water samples taken from the piezometers, which belong to the observation network of the new landfill (P3-P5, P11-P14), do not indicate the impact of the landfill. Moreover, the data from piezometer P12 were used to determine the hydrochemical background for this area. The distribution of the contamination index, as presented in Figure 4A, also emphasizes that the highest values of the contamination index are related to the groundwater outflow from the disused landfill, i.e. in the area of piezometer P10. This could mean that the maximum concentrations of pollutants in the pollution plume, as generated by the disused landfill, shifted from the landfill site to the south, possibly indicating a reduction in pollutant emissions from these parameters. Unfortunately, after having installed new piezometers in both the water inflow zone (P15 and P16) and the outflow zone (P17, P17A, P19 and P19A), and within the landfill area (P18), the subsequent research indicated a further negative impact of the landfill on groundwater and extreme concentrations of pollutants under the landfill. In this situation, the distribution obtained for 1995 (Fig. 4A) should be regarded as unreliable. In order to authenticate the spatial distribution of the analysed index, a simulation of the existence of two piezometers located in the water inflow zone, i.e. P15 and P16 (Fig. 4B), was carried out. In this simulation, the points at the groundwater inflow were allocated the values of the contamination indices calculated using a three-period moving average method, based on the available chemical analyses for the P15 and P16 piezometers. The obtained values of contamination indices for P15 and P16 were 2.47 and 8.19, respectively.

This procedure resulted in a slight shift in the index values >5 towards the north-west and, in turn, their covering of the northeastern part of the old landfill and the northwestern part of the new landfill (Fig. 4B). The arrangement of the index isoclines in the southern part did not change. Its highest values were still related to the P10 piezometer installed outside the landfill in the water outflow to the south. A more reliable interpretation of the distribution of the groundwater pollution index near the landfills in 1995 is shown in Figure 4C. The distribution of the contamination index around the old landfill is also presented based on monitoring data from 1995; the distributions are shown in Figure 4B. Additionally, a monitoring point at the top of the disused landfill was simulated (P18). This point was allocated a value of 1,100.85, which was obtained using a moving average method that was analogous to that used for P15 and P16. This procedure resulted in a completely different spatial distribution of the contamination index in the disused landfill area and its southern foreland (Fig. 4C). The highest index values were related to the groundwater beneath the landfill (in P18), whereas the overall spatial arrangement of the index pointed to the hydrodynamic arrangement of the groundwater, thus indicating that the negative impact of the landfill conformed to the groundwater flow direction, i.e. from north to south.

Similar problems arising from the variable number of piezometers emerged in the interpretation of the spatial distribution of the contamination index in the years of 2003, 2007, 2010 and 2015 (Fig. 5). However, in those years, the maps of the spatial distribution of index C_d were drawn up based on interpolation, taking into account the hydrogeological conditions, especially groundwater flow direction. The interpolation of the spatial distribution of the contamination index for 2007 (Fig. 5B)

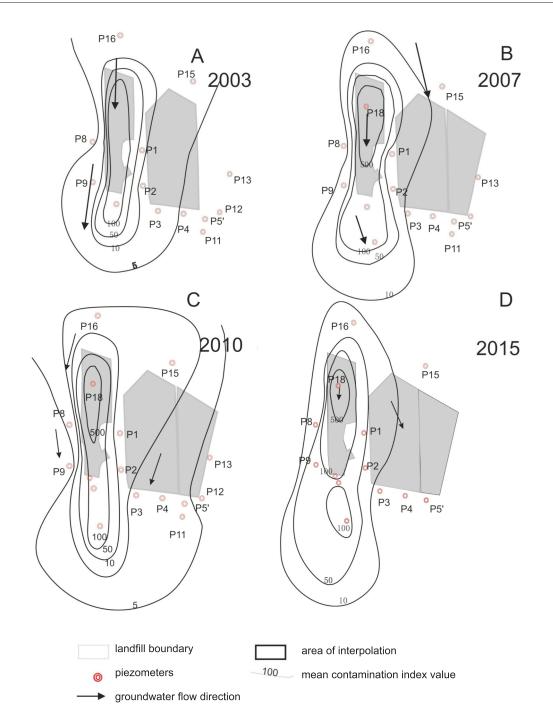


Fig. 5. Temporal changes in the spatial distribution of the groundwater contamination index in the region of the Tychy-Urbanowice landfills, southern Poland, for piezometers existing

has been based on the mean value for active nested piezometers P17 and P17A. For 2010 (Fig. 5C) and 2015 (Fig. 5D), the mean value for each of the two pairs of active nested piezometers (P19, P19A and P17, P17A) has been applied.

The spatial distribution of the contamination index indicated that the greatest alteration of the chemical composition of tested waters (Fig. 5) occurred directly under the inactive landfill and in its immediate southern vicinity.

Worth noting is that the contamination index values at all the piezometers have been very changeable over time (Table 2 and Fig. 5). Significant changes in the pollution index value in the analysed period were observed at piezometers surrounding

the old landfill from the west (P9, P8) and east (P1, P2). The most visible changes were observed at piezometers P9 and P1, which yielded the highest contamination index values of 81.33 and 71.01, respectively (Table 2 and Fig. 5). This may be associated with a periodic change in the groundwater flow direction and the presence of two water-bearing layers in the profile of these piezometers.

Figure 5 shows a general increasing trend in the index values, which is particularly well-visible at the southern discharge zone, where the range of the contour line 10 is shifted further to the south and currently occupies a significantly larger area (Fig. 5D). However, in the last five years, a reverse trend has been

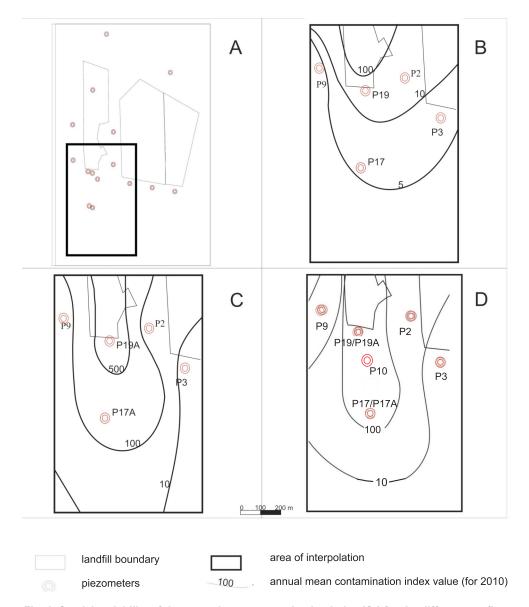


Fig. 6. Spatial variability of the groundwater contamination index (C_d) for the different configurations of nested piezometers, Tychy-Urbanowice landfills, southern Poland

A – detailed study area; B – based on indices for the shallow piezometers P17 and P19; C – based on indices for the deep piezometers P17A and P19A; D – based on average index values for both pairs of nested piezometers

noticed at the piezometers located immediately south of the landfill, especially at piezometer P10. The results of the analyses of the nested piezometers (P17/P17A and P19/P19A) are highly noteworthy (Fig. 5). The concentration of individual parameters in the water within the piezometers, which respectively monitor the upper (P17, P19) and the lower parts of the aquifer (P17A, P19A), is significantly different. In 2010, the annual mean value of the contamination index was 50 times higher at P17A than P17, and 18 times higher at P19A than P19. Five years later, the discrepancy between P17A and P17 decreased to a factor of about two, whereas, in the case of P19A and P19, the index value was only five times higher in the lower part of the aquifer. This resulted in a decrease in the average contamination index calculated for both pairs of the nested piezometers, from 350 to 102 in P19/P19A, and from 145.5 to 114.5 in P17/P17A (Fig. 5C, D). In the same five-year period, the contamination index for piezometer P10 decreased from 115 to 92. This resulted in a different spatial distribution of the index in 2015 (Fig. 5D), which confirms the validity of the aforementioned suggestions concerning natural attenuation processes in the landfill.

Of particular interest is the high temporal variability in the contamination index for piezometer P17 compared to other nested piezometers (Table 2). The reason for this may be the high groundwater vulnerability in this area (thin and very permeable unsaturated zone) and changeable rainwater infiltration. The minimum annual mean value of the contamination index was recorded in the extremely wet year of 2010, when intense recharge caused the contamination dilution in the upper part of the aquifer.

Diversified values from the adjacent nested piezometers negatively affected the functioning of the interpolation algorithm resulting in a less reliable picture of the distribution of the contamination index value. Therefore, the spatial distribution of the contamination index in the southern discharge area was drawn for the following three options:

- based on only the shallow piezometers, i.e. P17 and P19 (Fig. 6B);
- based on only the deep piezometers, i.e. P17A and P19A (Fig. 6C);
- based on the mean values from both pairs of piezometers (Fig. 6D).

These three options show different spatial distributions in the annual mean contamination index values calculated for 2010 (when there was the largest number of monitoring points and before the last network reorganization).

Analysis of these three different depictions of the index values' distribution (Fig. 6B-D) suggests that the first two (Fig. 6B, C), when considered separately, are also unrepresentative of a reliable assessment of the general impact of the landfill on the groundwater status. After having considered only the values from shallow piezometers (P19 and P17), the obtained distribution of the contamination index indicates a limited and rather small impact of the landfill on groundwater. This may also suggest that the spread of the contaminant plume is limited by natural attenuation processes taking place in the foreland of the landfill (Fig. 6B). The opposite situation occurs when the data from the deep piezometers (P17A and P19A) are taken into account (Fig. 6C). The spatial distribution of the C_d index points to a more significant and a wider range of impacts of the landfill. The reason for such values of the C_d index value is the migration of contaminants, which are further away from the landfill, through the bottom part of the aquifer. The spatial distribution of the mean values of the index (Fig. 6D) presents a picture of the range of average groundwater contamination levels within the aquifer profile, which corresponds well with the data from other piezometers with a long screen (P2, P3, P9, P10; Fig. 5C) and is somewhat reliable in the case of a thin homogeneous aquifer.

IMPLICATIONS

The contamination index is a simple and useful tool to determine synthetically the transformation of the chemical composition of water. Its simple formula of calculation allows involving simultaneously the parameters that are incomparable with each other, e.g. conductivity and concentrations. An important condition for the calculation of reliable values of the contamination index involves the properly determined values of the natural groundwater chemical background in the study area. However, it is not a simple task, in areas subjected to the negative and overlapping impact of different contamination sources. If the reliable values of the chemical background for the calculation of the contamination index cannot be determined, the highest acceptable concentrations of the individual parameters for drinking water can be applied. The second highly important aspect of the reliable index calculation is the application of an appropriate set of parameters that are relevant to specific environmental conditions and the types of pollutant generated by monitored parameters. An effective solution in this regard involves the application of principal component analysis (PCA) (Abdi and Williams, 2010). For the analysed landfill area, the PCA of 14 parameters was carried out, which resulted in the selection of seven essential parameters (SO₄²⁻, NH₄⁺, EC, Cl⁻, NO₂⁻, HCO₃⁻, NO₃) that mostly influence the chemical composition and the groundwater condition in the investigated area. Both these aspects of reliable contamination index calculations have been fully taken into account in this paper.

Another important issue discussed in this paper is the reliable interpretation of the spatial and temporal changes of the index. A properly designed groundwater observation network and a properly chosen method to interpret the results provide the basis for reaching accurate conclusions about the impact of pollution sources on groundwater. Of key importance is the knowledge of hydrogeological conditions, especially the groundwater flow direction and the location of the upstream and downstream zones in relation to the monitored landfill. It is important to assess the groundwater composition, alongside the reliable values of the contamination index, in the confirmed uncontaminated upgradient zone of the considered source of pollution. The next step should be the proper interpolation of the spatial distribution of the index in the groundwater downgradient areas. The contour maps of the spatial variability of the C_d index, constructed for the whole area, with no adequate number of piezometers and without taking into account the hydrodynamic conditions, may be unrepresentative, as illustrated in the example of the area of the landfill complex in Tychy (Figs. 4 and 5). The preliminary interpretation of the spatial distribution of the results can also be verified by later data. This was confirmed by the example of the landfill complex in Tychy, for which the conditions from 1995 were analysed (Fig. 4A-C). It was possible to determine the most probable and realistic impact of the landfill on the groundwater status only when the hydrogeological conditions and the subsequent results were taken into account, and the values of the contamination index in the simulated points were determined.

Due to the variable number of piezometers, the same problem occurred with the reliable assessment of the temporal changes in the spatial distribution of the groundwater contamination index (Figs. 4 and 5). The analysis of changes in the calculated indices for selected years (1995, 2003, 2007 and 2015) showed a variable pattern of their spatial distribution.

Extremely high values of the total contamination index of groundwater (C_d) were observed in the P18 piezometer that captures water beneath the abandoned landfill. The index values varied from 720.08 in 2007 to 2.336 in 2010, indicating leachate from the landfill rather than groundwater. Very high index values were also noticed in piezometers located at the groundwater outflow from the landfill site, for example, the C_d value in the P10 piezometer was 50 in 1996 and 329 in 2000. A similar situation occurred in the case of the the deep-nested P19A piezometer. The contamination index value for this piezometer was 88.85 in 2008 and 682.31 in 2011. Groundwater contamination in the P18 piezometer was so high (e.g., EC = $37.500 \mu S/cm$) that even the lowest value of the contamination index for the P18 piezometer (720.08) was higher than the highest value (682.31) for the aforementioned P19A deep piezometer located at the southern edge of the landfill (Figs. 1 and 2).

Analysis of temporal changes of contamination index values shows no visible negative or positive trends in active (P3, P4, P5') and recently abandoned (P11, P12, P13, P14) piezometers monitoring the new active landfill. This may indicate no negative impact of the new active landfill on groundwater. Some different trends were observed in piezometers that monitored the old landfill. Despite the periodical (seasonal) changes, general increasing trends were noticed in old piezometers surrounding the old landfill from the west (P9, P8) and east (P1, P2). Until the period of 2010-2012, the same increasing trend was also observed in new (drilled in 2006-2007) nested piezometers. Recently, the reverse trend has been observed in these piezometers. Interesting changes were found in the old piezometer P10 located south of the old landfill, in the groundwater outflow zone. In this piezometer, an increasing trend in contamination index values was observed from 1997. It changed to a decreasing one in 2001, continuing to date.

CONCLUSIONS

- 1. The calculated contamination index values and the temporal and spatial variability observed around the old landfill indicate:
- significant negative impact of this landfill on groundwater;
- decreasing negative impact in the southern outflow zone over the last 5 years;
- significant vertical changes in the contamination index values in the southern outflow zone;
- recent faster movement of the contamination plume than the contaminant delivery to the groundwater from the landfill;
- appearance of natural attenuation processes within the old landfill and in its outflow zone;
- significant vertical and horizontal changes in the contamination plume extent (due to seasonal changes in both groundwater flow directions and groundwater recharge).
- 2. Reliable assessment of the spatial distribution of the contamination index is determined by the differences in piezometer construction.
- 3. Reliable groundwater risk assessment, which employs the contamination index and contour maps, should be sup-

- ported by hydrogeological knowledge along with a properly designed groundwater monitoring network and a properly selected method to interpret results.
- 4. More representative results of groundwater risk assessment should be obtained for a systematically sampled, homogeneous regular monitoring network that consists of the same types of piezometer: either single piezometers with a long screen capturing water from the entire aquifer layer, or nested piezometers capturing water from different depths.
- 5. It is recommended to reduce the amount of landfill mass encapsulated within the closed landfill, to remove its hazardous material, or to reclaim the area, but in a way that limits the negative impact of leachates on groundwater.

Acknowledgements. The publication has been financed from the funds of the Leading National Research Centre (KNOW) received by the Centre for Polar Studies of the University of Silesia, Poland. We greatly appreciate the helpful comments and suggestions by Prof. I. Nyambe, Dr. D. Porowska and Anonymous Reviewer.

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