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CHANGES IN THE CONTENT OF SELECTED HEAVY METALS IN GROUNDWATER EXPOSED TO THE IMPACT OF A MUNICIPAL LANDFILL SITE*

Agata Szymańska-Pulikowska

Institute of Environmental Engineering
Wrocław University of Environmental and Life Sciences

Abstract

Landfilling is the most popular method of waste treatment in Poland. Old and usually uninsulated landfills constitute a major source of contaminants, which are leached by precipitation waters. As rainwater percolates through a landfill, suspended and dissolved components, waste decomposition products and various microorganisms are leached. Given the lack of any protection against leachate contacting groundwater, contaminants spread over a distance ranging from dozens of meters to several kilometers.

The purpose of this paper was to determine the degree of contamination of leachate and groundwater with heavy metals (manganese, iron, nickel, chromium, zinc, copper, lead and cadmium) in the surroundings of the Maślice municipal landfill site in Wrocław. The landfill was created without prior insulation of its base, and it has accumulated *ca* 2.4 mln m^3 of waste in over thirty years of its use. Studies on the groundwater were carried out from 1995 to 2008, and the examination of the leachate was concluded in 2004.

The analysis of the results shows that the groundwater flowing to the research object is already contaminated. While water percolates through the uninsulated base of the landfill, its quality continues to deteriorate. A large quantity of accumulated waste contributes to the severe contamination of the groundwater.

Leachate from the insulated part of the landfill displayed properties that correspond to "old" landfills. Inside such landfills, the processes occurring during the methanogenic phase of waste decomposition are abundant. Concentrations of heavy metals found during

dr hab. inż. Agata Szymańska-Pulikowska, Institute of Environmental Engineering, Wrocław University of Environmental and Life Sciences, Pl. Grunwaldzki 24, 50-363 Wrocław, Poland, phone: 71 320 55 52, e-mail: agata.szymanska-pulikowska@up.wroc.pl

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the research (except some characteristic values of iron and chromium) were within the limits that allow discharge to sewage facilities, surface waters or soil.

Changes in the content of analyzed heavy metals in the leachate and groundwater give a reason to suppose that in the years to come the amount of leached contamination may grow. Thus, it is necessary to continue the research – even in a scope broader than required by regulations concerning the monitoring of landfills.

Key words: municipal waste, landfilling, groundwater, leachate, contamination, heavy metals.

ZMIANY ZAWARTOŚCI WYBRANYCH METALI CIĘŻKICH W WODACH PODZIEMNYCH NARAŻONYCH NA ODDZIAŁYWANIE SKŁADOWISKA ODPADÓW KOMUNALNYCH

Abstrakt

Składowanie jest najbardziej rozpowszechnioną formą unieszkodliwiania odpadów w Polsce. Stare, często nieuszczelnione składowiska stanowią poważne źródło zanieczyszczeń, wymywanych przez wody opadowe. Przesiąkanie wody przez złoże odpadów powoduje wymywanie do wód gruntowych zawieszonych i rozpuszczonych składników, substancji powstających w trakcie rozkładu odpadów oraz rozmaitych mikroorganizmów. Brak zabezpieczeń uniemożliwiających kontakt wód odciekowych z podziemnymi powoduje rozprzestrzenianie się zanieczyszczeń, których zasięg może wynosić od kilkudziesięciu metrów do kilku kilometrów.

Celem pracy było określenie zanieczyszczenia wybranymi metalami ciężkimi (mangan, żelazo, nikiel, chrom, cynk, miedź, ołów i kadm) wód odciekowych i podziemnych na terenach otaczających składowisko odpadów komunalnych „Maślice” we Wrocławiu. Powstało ono bez wcześniejszego uszczelnienia podłoża, a w ciągu ponad trzydziestu lat eksploatacji zgromadzono na nim ok. 2,4 mln m^3 odpadów. Badania wód podziemnych prowadzono w latach 1995-2008, badania odcieków zakończono w 2004 roku.

W badaniach wykazano, że już wody podziemne dopływające do składowiska są zanieczyszczone. Podczas przepływu przez nieuszczelnione podłoże składowiska następuje dodatkowe pogorszenie ich jakości. Duża ilość nagromadzonych odpadów jest przyczyną poważnego zanieczyszczenia wód podziemnych.

Wody odciekowe z uszczelnionej części składowiska wykazywały właściwości odpowiadające „starym” składowiskom, w których wnętrzu dominują procesy związane z fazą metanogenną rozkładu odpadów. Stwierdzone w trakcie badań stężenia metali ciężkich (oprócz niektórych wartości charakterystycznych dla żelaza i chromu) mieściły się w granicach pozwalających zarówno na odprowadzanie do kanalizacji, jak i do wód lub do ziemi.

Zmiany zawartości analizowanych metali ciężkich w wodach podziemnych i odciekowych pozwalają przypuszczać, że w kolejnych latach ilość wymywanych zanieczyszczeń może się zwiększać. W związku z tym konieczne jest kontynuowanie badań, nawet w zakresie szerszym od wymaganego przez przepisy dotyczące monitoringu składowisk.

Słowa kluczowe: odpady komunalne, składowanie, wody podziemne, wody odciekowe, zanieczyszczenie, metale ciężkie.

INTRODUCTION

Landfilling is the most popular method of waste treatment in Poland. In 2009 alone, the amount of municipal waste dumped grew by 7,859,000 tons (Environment 2010), whereas its aggregate quantity in landfills is difficult to estimate. Old and usually uninsulated landfills constitute a major source of contaminants, which are leached by precipitation waters. As rainwater percolates through a landfill, suspended and dissolved components, waste decomposition products and various microorganisms are leached. The composition of produced leachate depends on the diversity and composition of the waste, degree of its decomposition, water content, type of environmental management, etc. Depending on the amount of precipitation and the method of waste concentration, the resultant leachate can equal 10% up to 60% of the annual precipitation (SZYC 2003). The time elapsing between rainfall and production of leachate can be 1 to 2 months (BENDZ et al. 1997). Leachate generated in the early phase of waste decomposition (acetogenesis) is characterized by a high content of organic acids, which increases the solubility of heavy metals. In the later phase (methanogenesis), leachate has neutral to alkaline pH (organic acids are decomposed to methane and carbon dioxide), which limits the solubility and migration of metals to leachate. Apart from the above properties, leachate can contain various other constituents, such as pesticides, polycyclic aromatic hydrocarbons (PAHs) and chloroorganic compounds (WILLIAMS 2002, ÖMAN, JUNESTEDT 2008, KULIKOWSKA 2009). When unused or past sell-by date medications are disposed to municipal waste containers, they are released into the environment. Studies have shown the presence of steroids, vitamins and sulfonamides in leachate from municipal landfills. Such substances as ibuprofen, naproxen, carbamazepine penetrate into groundwater (EGGEN et al. 2010). It has not been verified whether there is an indirect relationship between the content of such metals as nickel or cadmium and the toxicity of leachate. However, recurrence of organic contamination has contributed to its elevated toxicity. In order to remove the metaloorganic bonds of heavy metals, it is necessary to apply complex physicochemical purification methods (ÇEÇEN, GÜRSOY 2000, OLIVERO-VERBEL et al. 2008). These methods are recommended particularly in the case of leachate from long-exploited landfills, while leachate from "young" objects may be purified by biological methods (WILLIAMS 2002, KOC-JURCZYK, JURCZYK 2007, RENOU et al. 2008, KULIKOWSKA 2009).

The risk of heavy metal migration is linked to some other factors apart from the pH of leachate. At landfills in the vicinity of Prague (one operating and one closed), only arsenium, selenium and rubidium were found in the dissolved and mobile form of leachate (MATURA et al. 2010). The other analyzed metals (iron, aluminum, nickel, chromium, zinc, copper and lead) partly created inorganic colloidal bonds with carbonates or components of clay. Between 20% to almost 100% of iron, aluminum and lead were bound in this form. The occurrence of colloidal heavy metal bonds restricts their migration to groundwater. Research conducted at three uninsulated and long exploited landfills (Augsburg, Munich, Gallenbach) has shown that a change in hydrochemical conditions (on the border between the bed of the landfill and the area where groundwater is found) constitutes an effective barrier which stops metals in a colloidal form and reduces their leaching from a landfill (BAUMANN et al. 2006). Similar effects have been observed when leachate contacts surface waters. In oxidating conditions, metals were immobilized in carbonate and organic complexes (ETTLER et al. 2006). Given the lack of any protection against leachate contacting groundwater, contaminants spread over a distance ranging from dozens of meters to several kilometers (WILLIAMS 2002).

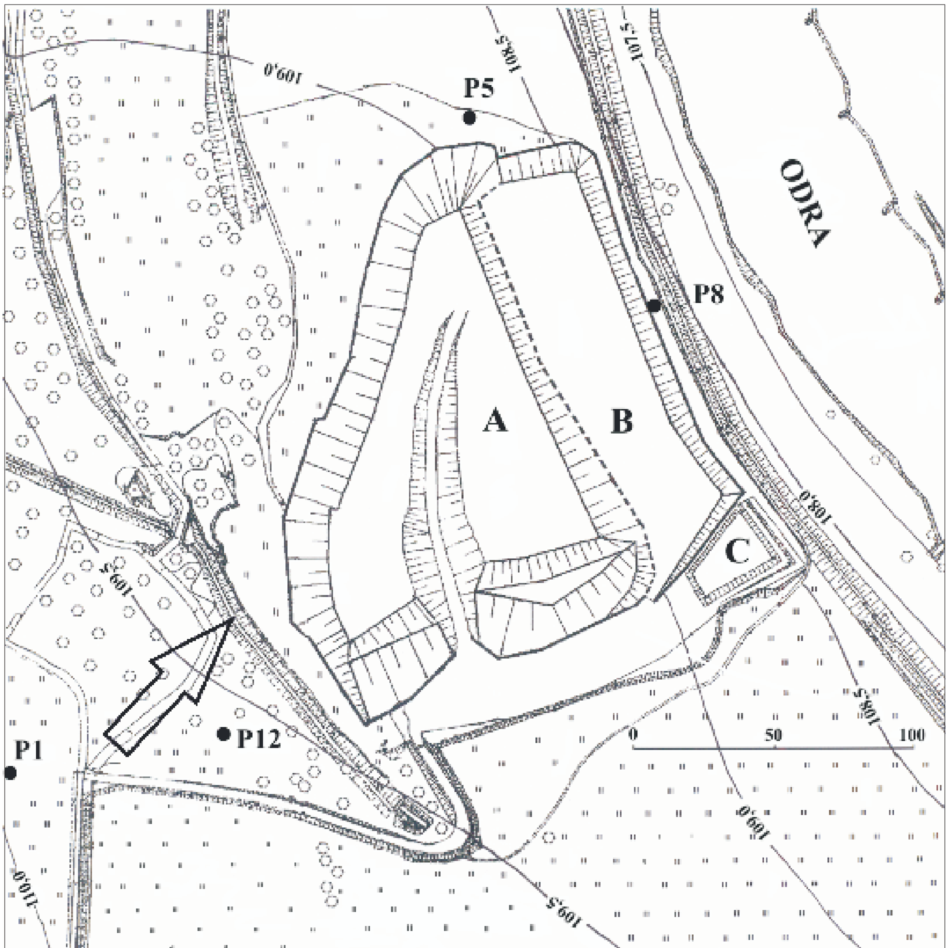
The rules for monitoring the environment in the surroundings of different types of landfills were specified in the Regulations of The Minister of Environment of 9 December 2002, concerning the range, time, method and conditions of monitoring landfill sites. The act defines the obligatory range of assays for every element of the environment (groundwater, surface water, leachate and landfill gas). Monitoring must take place from the moment the location is selected (the pre-exploitation phase), throughout the whole period of exploitation and for 30 years after the decision to close it down.

The purpose of this paper was to determine the contamination of leachate and groundwater with heavy metals (manganese, iron, nickel, chromium, zinc, copper, lead and cadmium) in the surroundings of the Maślice municipal landfill site in Wrocław.

MATERIAL AND METHODS

The object of this research was the municipal landfill site in Maślice, located in Kozia Street in the north-western part of Wrocław (Figure 1). It was built in the late 1960s to fill up an excavation pit remaining after sand deposits had been removed. In the first phase of its use, the waste was deposited into the excavation pit without any prior insulation.

After the excavation area had been filled up, an aboveground part of the landfill started to form. In the 1990s, the facility was expanded by building a new, insulated section and a ground reservoir (from which leachate was channeled) equipped with a monitoring system for the groundwater (a network of piezometers) and a pair of car scales. The area of the “old” section was *ca* 7 ha, while the area of the “new” section was *ca* 2 ha. The use of the latter part began in 1994 and all dumped waste was directed there for several years. When a deposit almost as high as the layer of waste in the old section had been formed, the dumping of waste continued throughout the whole available area, both in the insulated and uninsulated sections.



A – the old section of the landfill **B** – the new section of the landfill **C** – leachate reservoir	**P1, P12, P8, P5** – piezometers – 109,5 –, – 109,0 – hydroisohipsis → the direction of groundwater flow

Figure 1. The location of sample collection sites

The landfill functioned in this manner until the turn of 1999 and 2000. Eventually, the heap of waste reached a height of 50 meters containing nearly 2.4 million m^3 of waste. Rehabilitation works included for example stabilization of the waste body, installation of an outer cover and a screen isolating groundwater, as well as a system for removal of gas from the landfill. In 2004, the reservoir for leachate was bulldozed.

The location of the landfill is advantageous, mainly because it is isolated from adjacent terrains. To the west, partly north and south, it is surrounded by a fairly wide and ecologically varied stretch of woodland, which runs over to arable fields and meadows. Housing estates are about 800-1000 meters away from the facility. To the south of the landfill, the area is covered by allotments and industrial plants. Apart from these objects, the south-eastern side of the landfill site is bordered by meadows with narrow belts of shrubs, bushes and single trees. The Odra River flows 50-100 meters to the east of the site.

In the base of the landfill there are tertiary and quaternary (Pleistocene and Holocene) deposits. Tertiary deposits (clays, dusty clays, compact silty clays; local dust and sandy dust) lie at an average depth of 4.0-6.0 meters and they are 100-120 meters thick. Above them, there are quaternary deposits, which originate from the river (gravels, clay sand-gravel mixes and sands) with silty sand lenses. The total thickness of quaternary deposits ranges from 3.5 to 9.5 meters. One permanent aquifer has been found in the deposits. Its whose water table near the landfill is at a depth of 3-5 meters. Groundwater flows away towards the Odra riverbed, where the nature of drainage is in question (Figure 1).

The paper presents the results of analyses of the content of selected heavy metals (manganese, iron, nickel, chromium, zinc, copper, lead and cadmium) in groundwater (flowing to and away from the landfill) as well as in leachate. The studies of the chemical composition of the groundwater were carried out from 1995 to 2008; the studies of the leachate were finished in 2004, when the reservoir was bulldozed.

Research samples were taken 3 or 4 times a year. Leachate samples were collected from the reservoir, and groundwater samples came from four piezometers located on the sides of the inflow (2) and outflow (2) below the landfill (Figure 1). Before groundwater samples were taken, the stagnant water in the piezometer well was pumped out twice. The content of manganese, iron, nickel, chromium, zinc, copper, lead and cadmium in the samples was determined by means of atomic emission spectrometry, with excitation in inductively coupled plasma (ICP-AES). All the determinations were made at the Centre for Environmental Quality Sciences, the Institute of Soil Science and Environmental Protection at the Wrocław University of Environmental and Life Sciences. The evaluation of significance of the differences between groundwater and leachate from the landfill was made on the basis of a one-way analysis of variance (test F) at the significance level of p=0.05. Statistical analysis of the research results was carried out with Statistica 9 software.

RESULTS AND DISCUSSION

Table 1 shows characteristic values (the average, standard deviation and coefficient of variation) of concentrations of analyzed heavy metals in the groundwater flowing to the landfill. The results show that from 1995 to

Table 1

Characteristic values (average, standard deviation, coefficient of variation) of heavy metal concentrations in groundwater flowing to the landfill

Specification	n	Average	SD	V
Mn (mg dm^{-3})	106	1.909	2.315	121.3
Fe (mg dm^{-3})	106	41.09	54.87	133.5
Ni (mg dm^{-3})	98	0.235	0.212	90.20
Cr (mg dm^{-3})	98	0.092	0.181	196.3
Zn (mg dm^{-3})	98	0.688	1.218	177.0
Cu (mg dm^{-3})	98	0.046	0.032	70.02
Pb (mg dm^{-3})	98	0.021	0.026	123.9
Cd (mg dm^{-3})	98	0.002	0.002	144.2

SD – standard deviation, V – coefficient of variation, n – sample dimension

2008, groundwater flowing into the landfill could not be classified as natural. Average values often exceeded the ranges acknowledged as geochemical background, which proved the existence of a source (or sources) of contamination already flowing out from the landfill. Although the scope of research did not provide explicit identification of the cause of this phenomenon, the low pH of the analyzed waters and high content of sulphates (SZYMAŃSKA-PULIKOWSKA 2009) in conjunction with higher than normal concentrations of heavy metals may lead to a conclusion that biochemical oxidation of metal sulfides occurs in this area. This results in a considerable decrease of the pH and an increase in the heavy metal content of groundwater. Values deemed natural were most significantly exceeded for the concentrations of iron, manganese and nickel. The situation looked slightly better for lead and cadmium. Compared to the levels specified in the Regulation of the Minister of Environment of 23 July 2008 (on the criteria and methods of assessment of groundwater conditions), the values shown in Table 1 indicate severe contamination as well as high variability in the properties of water flowing to the landfill at Maślice.

Table 2 shows characteristic values (average, standard deviation, coefficient of variation) of the concentrations of selected heavy metals in water leaching from the Maślice municipal landfill (landfill leachate). The average contents of the analyzed constituents in leachate from the Maślice landfill were relatively low. Compared to concentrations of a typical methanogenic phase, the average values for the analyzed water were lower in the case of iron, copper, zinc, cadmium and lead and similar for nickel. It was only the average content of chromium that was markedly higher than the value reported in the literature (WILLIAMS 2002). All the average values determined for the leachate from the Maślice landfill exceeded corresponding values ob-

Table 2

Characteristic values (average, standard deviation, coefficient of variation) of heavy metal concentrations in landfill leachate

Specification	n	Average	SD	V
Mn (mg dm^{-3})	33	1.842	3.670	199.2
Fe (mg dm^{-3})	33	18.38	36.98	201.1
Ni (mg dm^{-3})	33	0.176	0.079	44.58
Cr (mg dm^{-3})	33	0.254	0.133	52.23
Zn (mg dm^{-3})	33	0.382	0.334	87.30
Cu (mg dm^{-3})	33	0.093	0.075	80.79
Pb (mg dm^{-3})	33	0.037	0.028	76.79
Cd (mg dm^{-3})	33	0.003	0.003	127.7

SD – standard deviation, V – coefficient of variation, n – sample dimension

tained during analyses of twelve Swedish landfills exploited for a period of several to 50 years (ÖMAN, JUNESTEDT 2008).

When compared with the results obtained for Polish landfills, the samples taken in Wrocław contained from 3- to 4-fold more chromium and copper and about 30% more zinc. Cadmium was the only metal whose concentration was three-fold lower than in the leachate from the Wysieka landfill, which has been exploited for a few years (KULIKOWSKA, KLIMIUK 2008). Analyses of leachate from old sections of another municipal landfill (MELLER et al. 2001) also revealed twice as much lead. The concentrations of zinc and cadmium were similar. Leachate from Maślice, on the other hand, contained markedly more manganese and iron (nine- and two-fold, respectively) and approximately 25% more nickel.

The extreme variability of the results reported in the literature enable us to conclude that inside the landfill at Maślice the dominant phase is the methanogenic one, when heavy metal concentrations in leachate decline. Similar changes were also observed during research conducted at other landfills, where most contaminants leached from "old" waste were organic and inorganic compounds. This poses a grave threat to the environment and yet is often neglected while planning the scope of monitoring studies (KULIKOWSKA, KLIMIUK 2008, ÖMAN, JUNESTEDT 2008, RENOU et al. 2008, SZYMAŃSKA-PULIKOWSKA 2008, 2008a).

The scale of heavy metal contamination of the analyzed leachate can also be determined through a comparison of their concentration with the levels detected in sewage discharged to sewage facilities or to surface water or soil. Among standardized constituents, the average concentrations of nickel, chromium, zinc, copper, lead and cadmium fall into the range of values that allow their channeling to both sewage facilities and surface water or soil. Iron was an exception because its content exceeded the level acceptable for discharging untreated leachate to the environment.

Table 3 shows characteristic values (average, standard deviation, coefficient of variation) of concentrations of selected heavy metals in waters flowing away below the Maślice municipal landfill.

Table 3

Characteristic values (average, standard deviation, coefficient of variation) of heavy metal concentrations in groundwater flowing away below the landfill

Specification	n	Average	SD	V
Mn (mg dm^{-3})	103	3.366	6.789	201.7
Fe (mg dm^{-3})	106	31.33	26.25	83.79
Ni (mg dm^{-3})	96	0.362	0.568	157.1
Cr (mg dm^{-3})	96	0.955	1.251	131.1
Zn (mg dm^{-3})	95	0.406	0.503	123.9
Cu (mg dm^{-3})	95	0.175	0.246	140.9
Pb (mg dm^{-3})	96	0.049	0.080	164.3
Cd (mg dm^{-3})	96	0.004	0.008	179.0

SD – standard deviation, V – coefficient of variation, n – sample dimension

The average heavy metal concentrations in water exposed to contact with the uninsulated base of the landfill differed from the corresponding values in water flowing to the facility. In most cases (manganese, chromium, nickel, copper, lead and cadmium), the average concentrations were significantly higher (even several-fold) due to the inflowing contaminants. The increase is noticeable despite the presence of reducing conditions in the landfill base, which encourage the precipitation of insoluble forms of the analyzed elements (SZYMAŃSKA-PULIKOWSKA 2009). Compared to the water flowing to the landfill, the water flowing away from the landfill contained on average less of just two elements: iron and zinc. The data presented in Table 3 also shows greater variability in the content of metals in outflowing water than in water flowing to the landfill (except iron, chromium and zinc). The water flowing away from the landfill must be classified as poor quality, mainly due to high concentrations of manganese, iron, nickel and chromium. The average heavy metal content in groundwater was even higher than the corresponding values in leachate, which proves continuous inflow from the uninsulated part of the landfill. Large accumulation of deposited waste creates a risk of emitting large quantities of contaminants. However, research conducted within the surroundings of small municipal landfills has not revealed such bad contamination of groundwater. The values considered natural can be exceeded only in isolated cases or for certain metals (STRUK-SOKOŁOWSKA et al. 2005).

Table 4 presents a range of concentrations (non-outlier values) of heavy metals marked in groundwater flowing away below the landfill and in leachate. Evaluation of the significance of differences between the average values (presented in Tables 2 and 3) was made by means of one-dimensional analysis of variance (test F) at the level of significance p=0.05. The measures of positions presented in the chart illustrate a greater range of fluctuations of the content of nearly all the analyzed constituents (except zinc) in the water flowing away below the landfill. The values characterizing the leachate were more balanced. Significant differences between the average concentrations

Table 4

The range of concentrations and significance of differences between average contents of heavy metals in groundwater flowing away below the landfill and in leachate

Specification	Range*		Test F
	groundwater flowing away below the landfill	leachate	
Mn (mg dm^{-3})	0.7-5.75	0.04-2.47	1.516
Fe (mg dm^{-3})	21.16-41.1	0.8-30.9	4.973**
Ni (mg dm^{-3})	0.135-0.589	0.142-0.223	3.474
Cr (mg dm^{-3})	0.462-1.28	0.17-0.3	10.26**
Zn (mg dm^{-3})	0.207-0.606	0.153-0.582	0.064
Cu (mg dm^{-3})	0.085-0.27	0.086-0.137	3.488
Pb (mg dm^{-3})	0.017-0.077	0.018-0.056	0.703
Cd (mg dm^{-3})	0.0013-0.007	0.0004-0.0035	1.477

* non-outlier values
** the marked differences are significant at level of p=0.05

of metals in the leachate and groundwater were only found for iron and chromium. The research revealed maximum values of concentrations of selected heavy metals (except for zinc and copper) in groundwater exposed to the influence of the landfill, thus leaving no other option but to classify the groundwater as the worst quality grade of ordinary groundwater. In the case of leachate, maximum concentrations of iron and chromium prohibited its discharge to the environment, whereas no values found in the research have made it impossible to discharge it to sewage facilities. The ranges of cadmium, lead and copper shown in Table 4 were similar to those found during the studies on large, uninsulated landfills in Germany (Augsburg, Munich, Gallenbach). Concentrations of chromium, iron, manganese and nickel in the surroundings of the landfill at Maślice were higher, and only the amounts of zinc were lower than in analogous German facilities (BAUMANN et al. 2006).

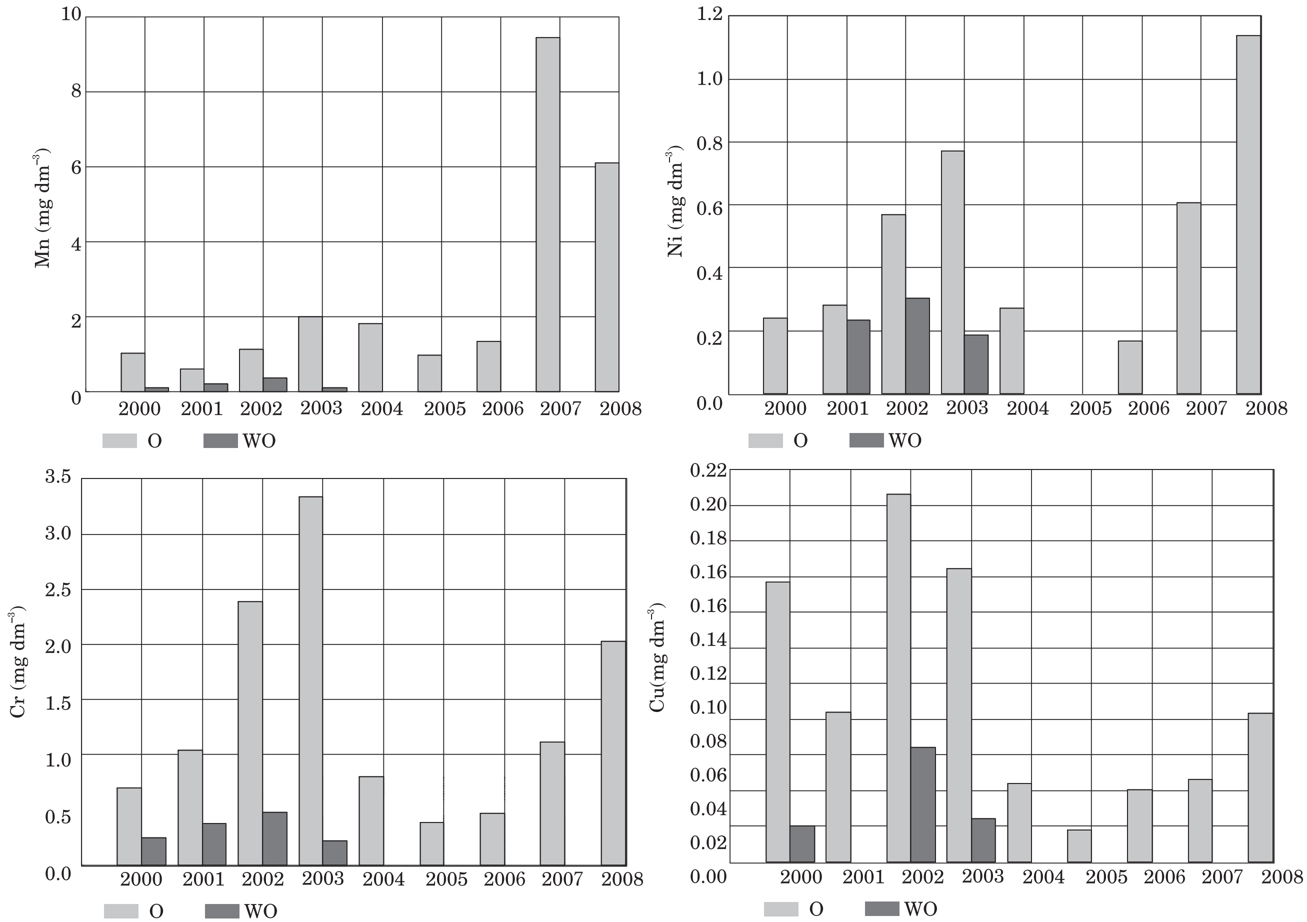


Fig. 2. The average concentrations of manganese, nickel, chromium and copper in leachate (WO) and groundwater flowing away below the landfill (O) in successive years of research

Figure 2 shows a comparison of average annual concentrations of manganese, nickel, chromium and copper that occur in leachate and groundwater flowing away below the landfill, after its closure. From the year 2000 to 2003 (when it was possible to calculate the average values) leachate was characterized by significantly lower contents of the above constituents. Both in leachate and in the water flowing away below the landfill until 2003-2004, a rise in the concentration of manganese, nickel and chromium was observable as well as a persistently high copper content. Afterwards, the concentrations of these metals declined, although in the successive years (2006-2008) their content in groundwater rose again, which was probably caused by the leaching of metals from the waste accumulated in the landfill in the final years of exploitation.

The picture of changes in the properties of leachate occurring in time is not always clear. Research conducted in the first eight years of exploitation of the municipal landfill in Sianów has shown an increasing concentration of heavy metals in leachate in the first 3 to 4 years of exploitation (SZYMAŃSKI et al. 2007). Particularly high concentrations were found for zinc, iron, manganese, copper, lead and nickel. The concentrations of chromium and cadmium were considerably lower. During twelve years of research of leachate from Dyer Boulevard Landfill (Florida, USA), it was observed that with time the content of chromium and nickel was decreasing, while the concentrations of iron and manganese were increasing. In other cases, many research results indicated concentrations below detection levels (STATOM et al. 2004). The occurrence of higher concentrations of some contaminants in groundwater under the impact of the Maślice landfill, after a short-lasting decline, is a signal that the area requires constant monitoring, and the emission of contamination may last for many years to come

CONCLUSIONS

1. The groundwater in the whole area surrounding the Maślice municipal landfill is of poor chemical quality. The persistent inflow of contaminants from the uninsulated section adds to its evident deterioration.

2. The analyzed leachate showed properties corresponding to “old” landfills, in which processes related to the methanogenic phase of waste decomposition dominate. Concentrations of heavy metals found during the research (except some values characteristic of iron and chromium) were within the limits that allow discharging leachate to sewage facilities, surface water or soil.

3. Changes in the concentration of the analyzed constituents in groundwater and leachate which occur after a landfill is closed down suggest that

the amount of leached contaminants will grow in the following years. That is why it is necessary to continue research, even in a wider scope than required by existing regulations concerning the monitoring of landfills:

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