

Chemical waste disposal site as a source of groundwater contamination in the multi-AQUIFER system of Upper Silesia, Poland

ANDRZEJ J. WITKOWSKI, ANDRZEJ KOWALCZYK, HANNA RUBIN and KRYSZTYN RUBIN

University of Silesia, Faculty of Earth Sciences, ul. Będzińska 60, 41-200 Sosnowiec, Poland;
Phone: +48 32 3689283, Fax: +48 32 2916888; E-mail: awitkows@us.edu.pl

Abstract. At present the region of Tarnowskie Góry is the best-known area in Poland because of the negative impact of industrial waste disposal sites on groundwater quality in the Quaternary and Triassic multi-aquifer system. An increase of concentrations of B, Ba, Sr, SO₄, Cl in groundwater of both the aquifers have been noted. The high concentrations of boron (up to 116 mg/dm³) observed in the Triassic (the Muschelkalk) carbonate aquifer are particularly alarming, because this aquifer is the most important source of potable water for the region of Tarnowskie Góry.

A groundwater-flow model and a solute-transport model were developed for this multi-aquifer system (four aquifers – two Quaternary and two Triassic, separated by three aquitards). Both the Quaternary aquifers are of the transit type, being a source of recharge of lower Triassic aquifers. The horizontal flow in the Triassic aquifers predominates. Results of groundwater monitoring and numerical modelling revealed a significant differentiation of boron migration intensity within the Quaternary and Triassic aquifers, depending on water flow direction. A limited lateral migration of boron within the Quaternary aquifer and practically unlimited migration within the Triassic ones have been observed. Numerical model simulations showed that important big potable-water intakes, being situated at a distance of about 5-9 km NW downgradient from the waste disposal sites, are safe.

Keywords: industrial wastes, boron, contamination, multiple aquifers

Introduction

The region of Tarnowskie Góry constitutes a perfect example of negative environmental impact caused by casually locating waste site that took into account no environmental considerations. In the considered area, the industrial waste disposal sites of the former chemical plant are biggest in Poland. This situation, together with the naturally high groundwater vulnerability to pollution, has resulted in a progressing degradation of water quality in the Quaternary and Triassic aquifers. Locating the wastes in the watershed area has additionally complicated the situation, leading to multidirectional contamination spreading (Fig. 1).

The high concentration of boron is perceived as particularly dangerous since it reached 240 mg/dm³ in the Quaternary aquifer, and 116 mg/dm³ in the Triassic one. This critical situation resulted in closing of many water intakes situated nearby. Therefore a complex remediation of this area together with relocation of wastes have been performed.

In order to assess the current and perspective spreading of boron in the groundwater of the analysed multi-layered flow system a numerical modelling has been applied. Groundwater flow and single component (for boron) solute transport numerical models of the four-layer system was worked out (Kowalczyk et al., 2003).

Waste disposal site

The region of the Chemical Works "Tarnowskie Góry", which are at the final stage of liquidation process, was an area where for many years the diverse industrial activities took place. The list of these activities encompasses: silver and lead ore mining (from the 12th century), an iron milling (the second half of the 19th century), manufacture of silk paper (1892-1919), and production of paints and chemicals (1921-1995). But the Chemical Works "Tarnowskie Góry" that have manufactured over 30 various inorganic chemicals for over 75 years, among them mainly barium, boron, zinc, copper, and strontium, proved to be particularly environmentally hazardous. The used obsolete technologies generated large amounts of production wastes. The wastes together with the sludge from a sewage treatment plant were stored directly on the ground without any security means that could prevent the exfiltration of leachates. In this way there came into being the waste dumps containing dangerous compounds of barium, boron, strontium, zinc, and copper. The dumps covered an area of over 27 ha and their mass was estimated at circa 1.4 million tonnes (Fig. 1).

An analysis of selected samples of the wastes showed that they have diverse chemical compositions with domination of sulphates (up to 49600 mg/kg d.m.), calcium (up to 31170 mg/kg d.m.) and magnesium (up

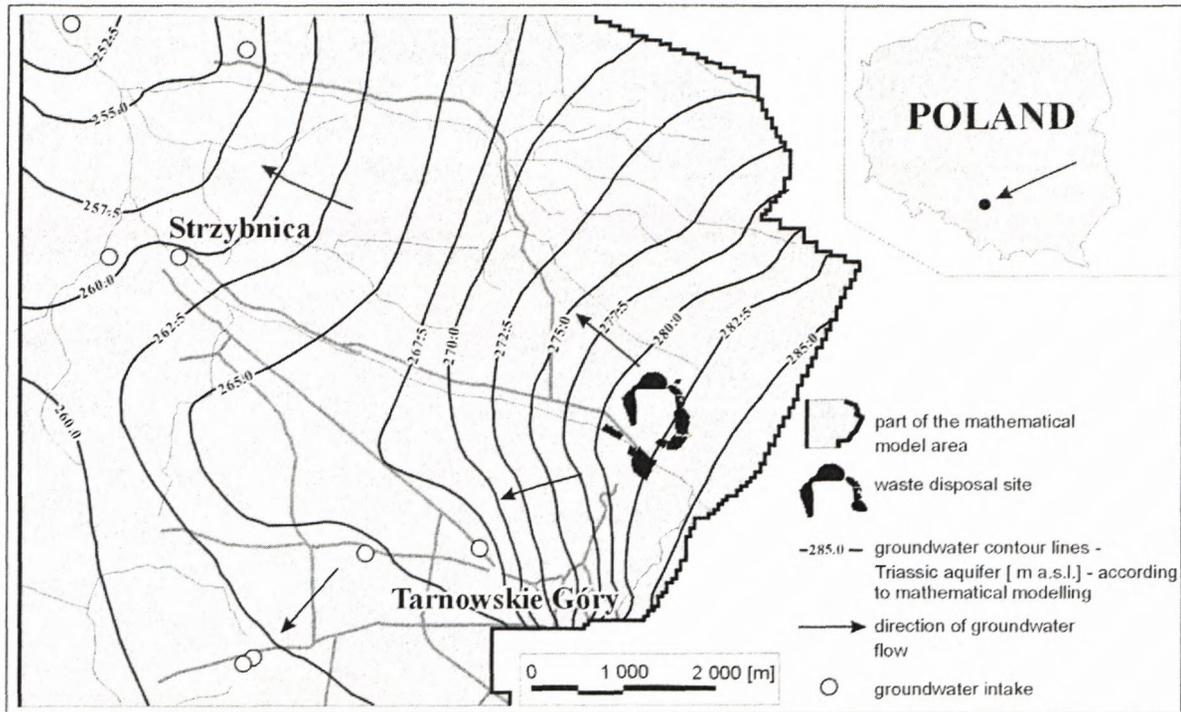


Fig.1 Location of the studied area and groundwater features

to 10840 mg/kg d.m.). Boron content ranged from 65.3 to 2216 mg/kg d.m., barium content from 81.9 to 443.6 mg/kg d.m., strontium content from 1646.7 to 8576.0 mg/kg d.m., copper from 93.9 to 9556 mg/kg d.m., zinc from 305.9 to 9500 mg/kg d.m. and arsenic from 95.3 to 901.8 mg/kg d.m. (Rubin, 1999).

The wastes were characterized by diverse but generally high active porosity (7-50 %) that decreased with their age. The lower porosity features the earlier stored wastes. Until recent the wastes did not constitute any essential barrier to the vertically infiltrating water (the hydraulic conductivity of investigated wastes ranged from $6.25 \cdot 10^{-8}$ to $6.4 \cdot 10^{-7}$ m/s) (Rubin, 1999). Moreover the karst-fissure character of rocks and their secondary permeability increase, caused by the many centuries of mining activity, facilitated the pollutants generated by the wastes to spread within the carbonate karst-fissured aquifer. Conducted dynamic leaching tests on selected waste samples showed a very high leachability of boron (up to 634 mg/l) and a lower one of strontium (up to 30 mg/l) and arsenic (up to 5.2 mg/l). Some samples also showed a high leachability of sulphates (up to 4989 mg/l) and chlorides (up to 1517 mg/l; Rubin, 1999).

The Chemical Works ceased its production activity in 1995 and was subjected to a liquidation process. In the mid-2000s implementation of a project of complex neutralization of the wastes together with polluted-land reclamation have begun. The scope of work planned within the project included construction of the Central Waste Disposal Facility (CWDF) on an area of 14.11 ha and a storage capacity of about 1.5 million m^3 and relocation of the whole of wastes and polluted ground from below the waste dumps to the facility. CWDF is a modern facility that satisfies all ecological requirements. It is an above-

ground-ranging object (after closing of 17 m in height above ground surface). It has 5 cells with sealed bottom and draining systems.

Groundwater occurrence and monitoring

The Tarnowskie Góry region is located in the southern part of Poland (Fig. 1) and belongs to the Silesian-Cracow Monocline consisting of the Triassic formation discordantly overlying folded and faulted Paleozoic base. The Triassic formation is covered by Quaternary deposits of various lithology and thickness. There are Quaternary and Triassic aquifers in the hydrogeological profile. Thickness of these aquifers varies from a few to more than 50 m and from 40 to 150 m respectively. The Quaternary aquifer most often consists of two water-bearing horizons with discontinuous spread. Hydraulic conductivity of the Quaternary aquifer varies from 0.1 m/24 h up to 15 m/24 h. The water of this aquifer is polluted and is not used. The most important and abundant is the Triassic karst-fissured carbonate aquifer (dolomites and limestones with marl interbeddings). Hydraulic conductivity of the Triassic aquifer varies from 1 m/24 h to 16 m/24 h and its effective porosity is estimated between 0.04-0.06. In places both the aquifers form four separated water-bearing horizons: two within the Quaternary and two within the Triassic (Muschelkalk and Roethian; Fig. 2). The Quaternary aquifer is discharged by the Drama and Stoła rivers in local flow systems. In shallower part of the Triassic aquifer (Muschelkalk) both local and regional flow systems have been formed while in the deeper one (Roethian) only regional flow system has developed. The groundwater flows out of the watershed area in the waste disposal sites vicinity to the NWW and SWW (Fig. 1). Velocity of ground-

waterflows in the Quaternary aquifer ranges from 0.001 to 1 m/24 h, whereas in the Triassic aquifer from 0.1 to 3 m/24 h.

The groundwater quality monitoring network in this area has been subjected to multiple modifications since 1990 when it was brought into existence. Unfortunately

the mentioned sites have been systematically monitored only since 1999 (Rubin and Witekowski, 2002). Current groundwater quality monitoring network consists of 45 observation wells which monitor the Quaternary (20 wells) and the Triassic (25 wells) aquifers (Fig. 3). The wells are 4 to 28 metres deep, in case of the Quaternary

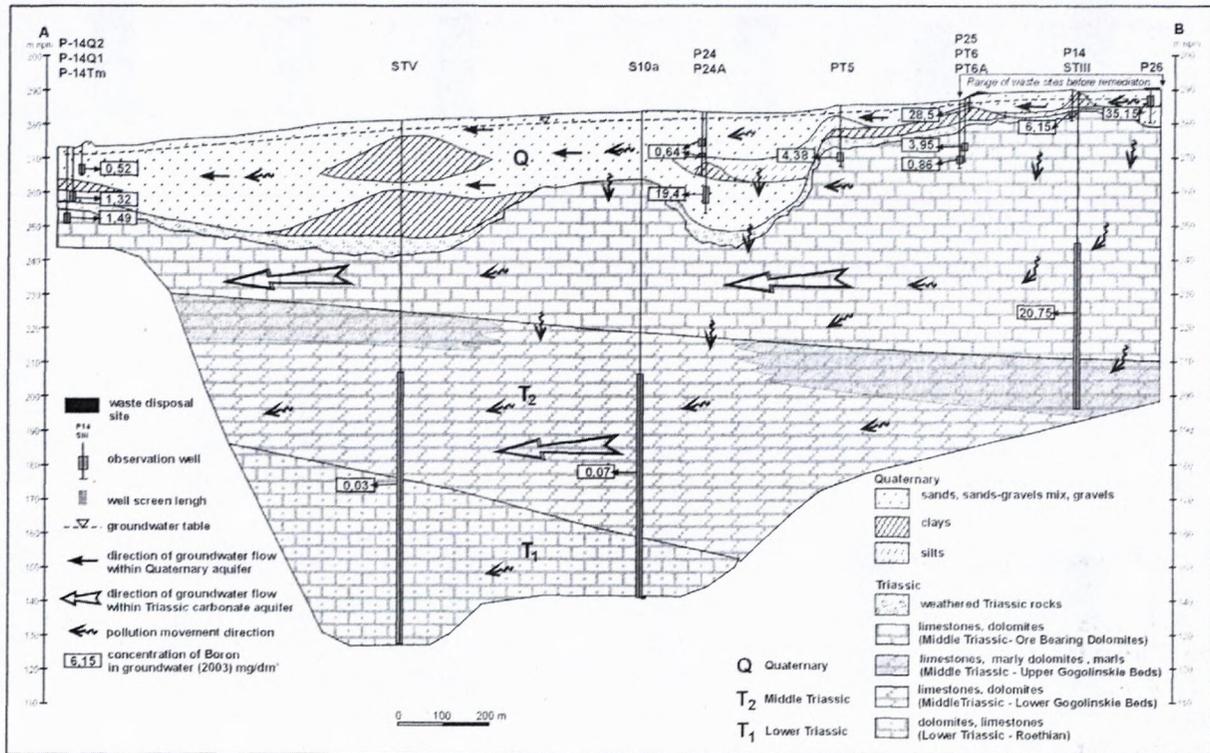


Fig. 2. Hydrogeochemical cross-section.

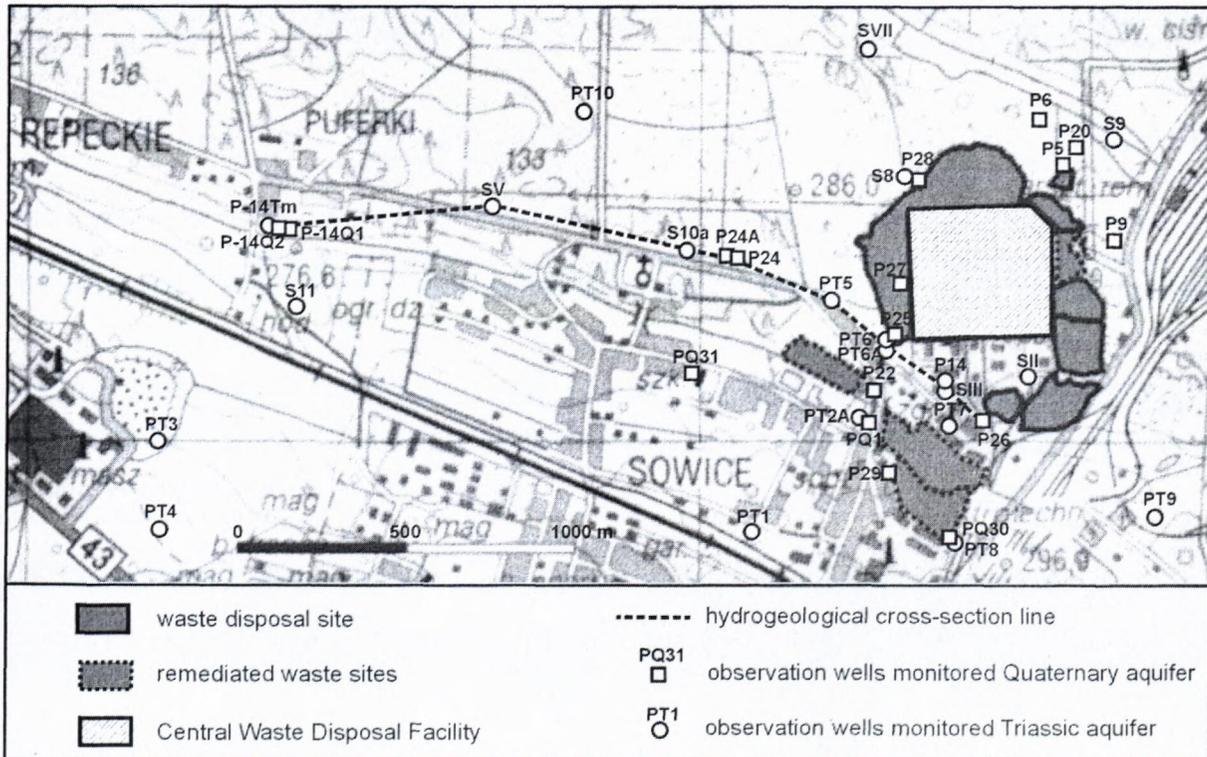


Fig. 3. Groundwater quality monitoring network.

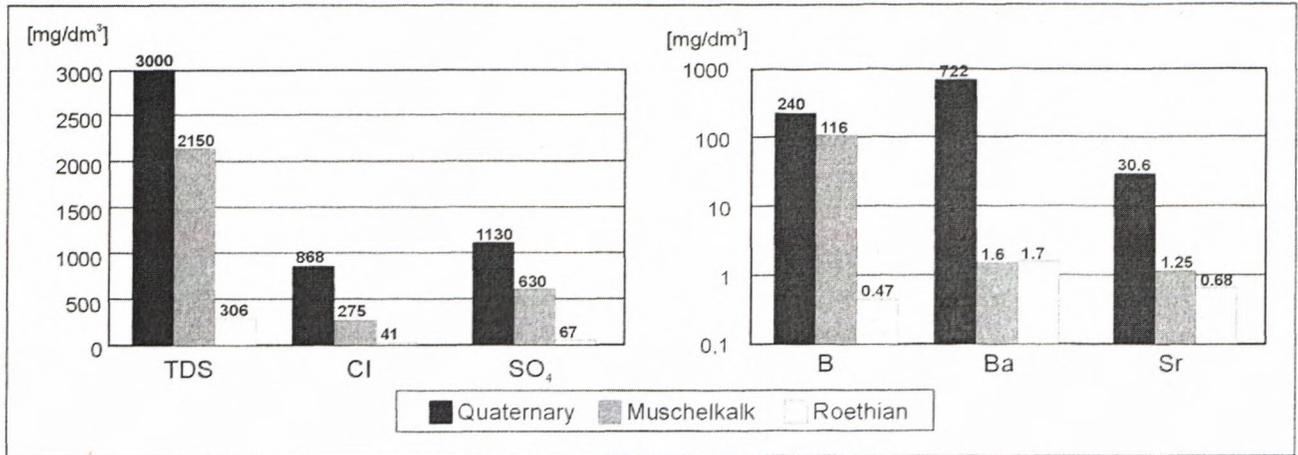


Fig. 4. Maximum concentration of selected groundwater pollution indicators.

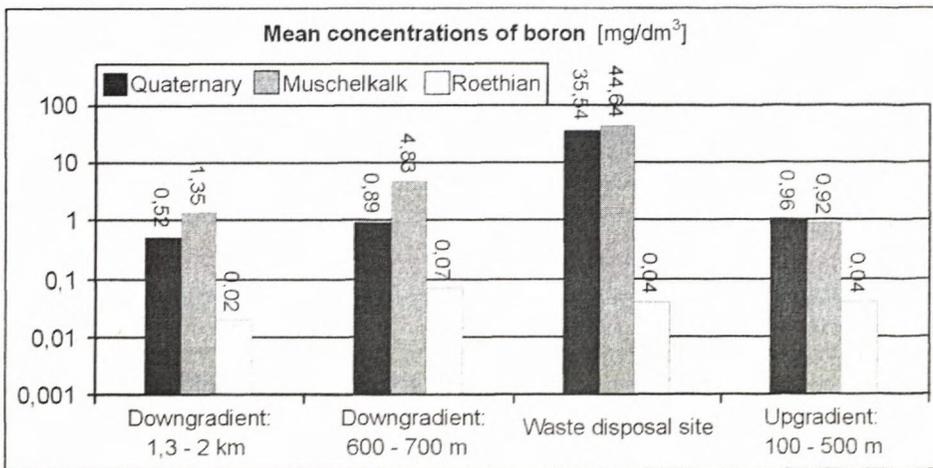


Fig. 5. Changes of boron concentration along selected groundwater flow pathways

aquifer, and 15 to 155 metres deep, in the case of Triassic one. There are only 5 groups of nested observation wells. Each of the groups consists of 2 to 3 wells (altogether 12 wells).

Field measurements encompassed the determination of temperature, pH, specific conductance and oxidation-reduction potential Eh. The range of chemical indicators determined by the laboratory tests has been changed. Currently 24 indicators are determined: TDS, COD, N-NH₄, N-NO₃, Cl, SO₄, HCO₃, Ca, Mg, Na, K, Al, As, Ba, B, Cd, Cr, Cu, Fe, Sr, Zn, detergents (only for the Quaternary aquifer), trichloroethene and tetrachloroethene (only for the Triassic aquifer).

Modelling

A groundwater-flow model and a solute-transport model were developed for the multi-aquifer system. The groundwater-flow system was simulated in three dimensions using the MODFLOW-96 computer code (McDonald and Harbaugh, 1988; Pollock, 1994; Zheng and Wang, 1999). MODFLOW was used in combination with MODPATH in order to determine the migration route and time of a contaminant from a waste disposal site to the existing well fields. MT3D was used in the solute-transport model. The model cell size is 50 x 50 m so the

models were discretized into 231 rows and 113 columns. The models were vertically discretized into four layers (two Quaternary and two Triassic ones) separated by three aquitards. The models were calibrated for present period by means of the trial-and-error methodology using water-level data and boron-concentration data.

It was assumed that the current spatial distribution of boron concentration in the system under consideration is a result of its infiltration from the wastes taking place for the last 70 years. The simulated boron concentrations for that period stay in acceptable agreement with the measured data.

Groundwater quality

Groundwater quality in the Quaternary aquifer is very differentiated but generally it is very bad. Highly contaminated water is observed in the area of waste site: TDS – up to about 3000 mg/dm³, Cl – up to 868 mg/dm³, SO₄ – up to 1130 mg/dm³, B – up to 240 mg/dm³, Ba – up to 722 mg/dm³, Sr – up to 30.6 mg/dm³ (Fig. 4). Temperature of the groundwater varies seasonally between 5 °C (in winter) and 15.5 °C (in summer).

In the waste disposal area in the Quaternary aquifer groundwater in comparison to the water of upgradient zone a 60-time increase of the average boron concentra-

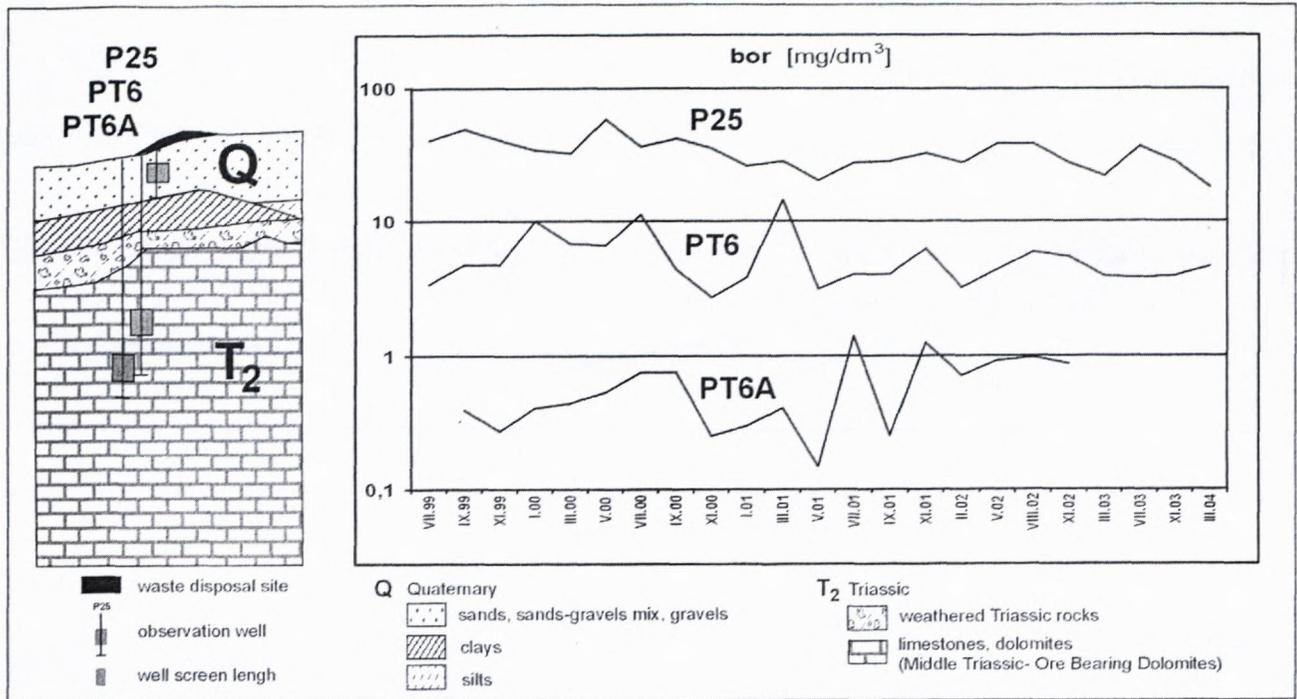


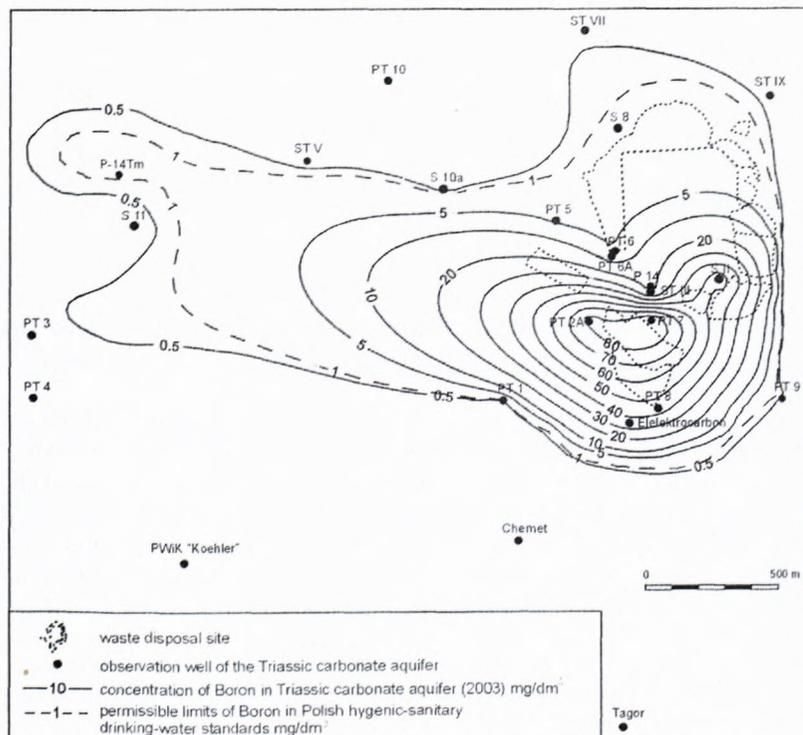
Fig. 6. Changes of boron concentration in groundwater in selected nested piezometers.

tion and a 26-time increase of the average strontium concentration have been observed (Fig. 5). The Quaternary aquifer groundwater in that area also shows increased concentrations of zinc, aluminium, cadmium, manganese, nickel, led, iron, ammonia and nitrates.

Generally similar groundwater quality of Quaternary aquifer in upgradient areas as well as downgradient ones (at a distance of about 2 km from the waste sites) is observed. It indicates a limited lateral migration of contaminants within the Quaternary aquifer.

Within the **Muschelkalk aquifer** the chemical variability of groundwater is connected with location of observation points referred to the disposed waste body and its location in the aquifer profile. Groundwater of this aquifer is generally less contaminated as compared with the Quaternary one (Fig. 4). Significantly contaminated water (locally even more than the one in the Quaternary aquifer) was noticed only in the top part of the Muschelkalk in the area of waste disposal sites and adjacent downgradient areas (up to about 1 km from the sites; TDS – up to 2150 mg/dm³, Cl – up to 275 mg/dm³, SO₄ – up to 630 mg/dm³, B – up to 116 mg/dm³, Ba – up to 1.6 mg/dm³, Sr – up to 1.25 mg/dm³; Rubin and Witkowski, 2003). In the waste disposal site area there have also been observed increased concentrations of ammonia, nitrates, manganese, iron, zinc, cadmium and nickel.

Water of better quality was observed at the base parts of the Muschelkalk aquifer in the area of the considered sites and at a distance of up to about 2 km downgradient from them (TDS – up to 694 mg/dm³, Cl – up to 78.8 mg/dm³, SO₄ – up to 127 mg/dm³, B – up to 1.49 mg/dm³, Ba – up to 0.1 mg/dm³, Sr – up to 0.327 mg/dm³). Temperature of groundwater within the Muschelkalk aquifer is more stable and varies from 10 °C to 12 °C.



Water of better quality was observed at the base parts of the Muschelkalk aquifer in the area of the considered sites and at a distance of up to about 2 km downgradient from them (TDS – up to 694 mg/dm³, Cl – up to 78.8 mg/dm³, SO₄ – up to 127 mg/dm³, B – up to 1.49 mg/dm³, Ba – up to 0.1 mg/dm³, Sr – up to 0.327 mg/dm³). Temperature of groundwater within the Muschelkalk aquifer is more stable and varies from 10 °C to 12 °C.

Fig. 7. Spatial distribution of the boron concentration in the Triassic aquifer (based on monitoring data – 2003)

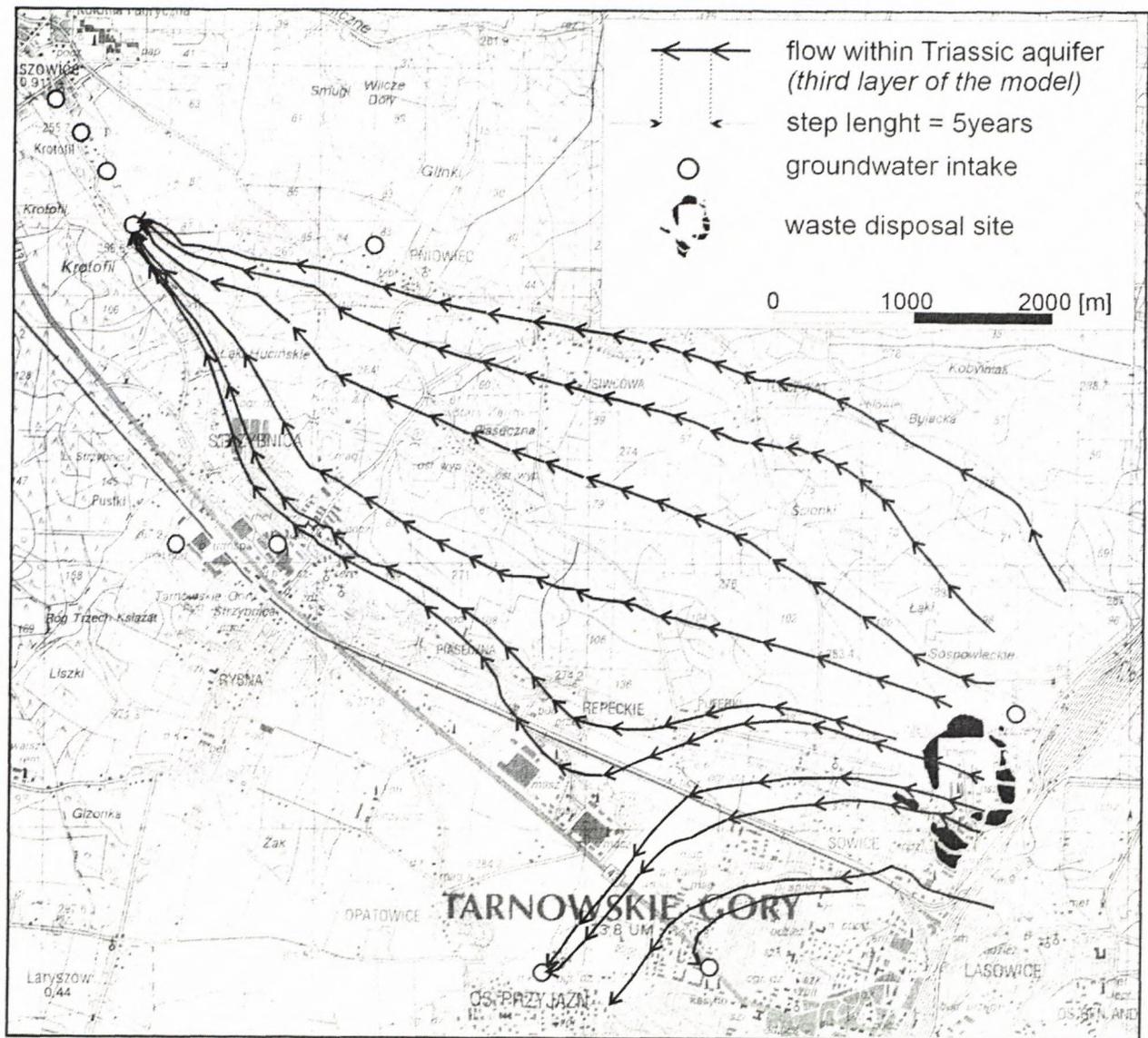


Fig. 8. Groundwater flow pathways according to the numerical modelling.

Groundwater of the fourth, lowest, aquifer (**Roethian**) have been practically uncontaminated by the considered facility (TDS – up to 306 mg/dm^3 , Cl – up to 41 mg/dm^3 , SO_4 – up to 67 mg/dm^3 , B – up to 0.47 mg/dm^3 ; Fig. 4). The groundwater has a slightly lower and stable temperature ranging from 9 to 10°C .

Vertical and horizontal differentiation of the boron concentration in groundwater of the considered multi-aquifer system is observed. Maximum content of H_3BO_3^0 (major easily migrating boron speciation) in groundwater of Quaternary and Triassic (Muschelkalk) aquifers occurs in the waste dump area (Fig. 5). However the maximum content of boron in groundwater of deepest Roethian aquifer is observed in the area of observation well S10a (about 0.7 km W downgradient from the waste disposal sites) where Muschelkalk and Roethian aquifers are probably hydraulically connected (Fig. 2). Vertical differentiation of boron content in

groundwater is very well noticed in nested piezometers (Fig. 6). The mean concentrations of boron in the area of nested piezometers P25, PT6 and PT6A have varied from 0.86 mg/dm^3 (in the deeper part of the Muschelkalk aquifer – piezometer PT6A) up to 28.5 mg/dm^3 (in the shallow Quaternary aquifer – piezometer P25; Fig. 6).

In the course of the five years from 1999 to 2003 a general improvement of groundwater quality of the Quaternary aquifer and some relative stabilization of the groundwater quality of the Triassic aquifer were observed (Fig. 6). The current spatial distribution of the boron concentration within the Triassic aquifer based on monitoring data is shown in Fig. 7.

The hydrodynamic system of the Triassic aquifer shows a practically unlimited boron migration along some privileged flow ways in the NW and SW directions (Fig. 8).

Conclusions

The waste disposal sites under consideration have caused a significant groundwater contamination within the Quaternary and Triassic aquifers. Results of groundwater monitoring as well as numerical modelling indicate that an intensive downward groundwater flow within the first Quaternary aquifer predominates in the area of the waste disposal sites. A downward and locally horizontal flow was observed within the lower Quaternary aquifer. Both the Quaternary aquifers are of the transit type, being a source of recharge of lower Triassic aquifers. The horizontal flow in the Triassic aquifers is predominant while within the upper one (Muschelkalk) some weak downward component is also observed. The natural regional groundwater flow pattern is modified by active wells and old mine workings.

A significant differentiation in migration intensity of boron has been observed within the Quaternary and Triassic aquifers depending on water flow direction. Results of hydrodynamic modelling indicated a practically unlimited water outflow within the Triassic aquifers, which would enable the easily migrating boron to spread along privileged flow pathways without limits. However a limited lateral migration of contaminants within the Quaternary aquifer has been observed. Results of groundwater quality monitoring and simulated groundwater flow and advective transport of boron showed that groundwater of the Triassic aquifers discharging by wells located about 2.5–3 km SW downgradient from the waste disposal sites is possible to be contaminated in 25–30 years. The important, big water intakes situated at a distance of about 5–9 km NW downgradient from the sites are practically safe. Travel

time of advecting boron from the waste site to that well field is estimated at about 90 years (Fig. 8).

Current remediation works and natural attenuation processes should result in general improvement of groundwater quality in all aquifers.

References

- Kowalczyk, A., Lewandowski, J., Rubin, K., Witkowski, A., Wojtal, G. and Wróbel, J., 2003: Numerical modelling of contamination transport in the area of Tarnowskich Gór. *Intergeo, Sosnowiec*, 59 p. (Unpublished report.)
- Mc Donald, M. G. and Harbaugh, A. W., 1988: A modular three-dimensional finite-difference ground-water flow model. U. S. Geological Survey Techniques of Water Resources Investigations, Book 6, 586 p.
- Rubin, H., 1999: Application of selected leaching tests of contaminants from wastes for groundwater contamination risk assessment in the area of waste sites of Chemical Factory "Tarnowskie Góry". Unpublished PhD. Thesis. University of Silesia, Sosnowiec, 161 p. (In Polish.)
- Rubin, H. and Witkowski, A. J., 2002: The local groundwater quality monitoring system in the area of industrial landfills of the chemical plant „Tarnowskie Góry. In: Rubin, H., Rubin, K. and Witkowski, A. J. (eds.): Groundwater quality and vulnerability, *Prace Wydziału Nauk o Ziemi UŚ*, No 22, Sosnowiec, 177–189. (In Polish.)
- Rubin, H. and Witkowski, A. J., 2003: Impact of wastes disposal from chemical factory in Tarnowskie Góry on groundwater based on monitoring data results. In: Kozerski, B. and Jaworska-Szule, B. (eds.): *Współczesne problemy hydrogeologii*, Vol XI, Gdańsk, 61–468. (In Polish.)
- Pollock, D. W., 1994: User's Guide for MODPATH/ MODPATH-PILOT, Version 3: A Particle tracking post-processing package for modflow. The U. S. Geological Survey finite-difference ground-water flow model, U.S. Geological Survey Open-File Report 94-464.
- Zheng, C. and Wang, P. P., 1999: MT3DMS: A modular three-dimensional multi-species model for simulation of advection, dispersion and chemical reactions of contaminants in groundwater systems. Documentation and user's guide, SERDP-99 – 1, U.S. Army Engineer Research and Development Center, Vicksburg.