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GROUNDWATER QUALITY AND MIGRATION OF POLLUTANTS IN THE MULTI-AQUIFER SYSTEM OF THE FORMER CHEMICAL WORKS "TARNOWSKIE GÓRY" AREA

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Abstract. An increase of B, Ba, Sr, SO_4, and Cl concentrations in groundwater of the multi-aquifer system has been noted within the waste dump area of the former chemical works "Tarnowskie Góry". Very high boron concentrations in the Quaternary aquifer (up to 240 mg/dm^3) and in the Muschelkalk one (up to 116 mg/dm^3) have been observed. In order to assess current and perspective spreading of boron in the groundwater of the analysed multi-aquifer system, a groundwater flow model (four aquifers: two Quaternary and two Triassic ones, separated by three aquitards) and a solution-transport model were developed.
The hydrogeochemical modelling has shown that the share of contaminated Muschelkalk water was equal to 12–22% of the total flow in the Roethian aquifer. A significant differentiation in the intensity of boron migration has been observed within the Quaternary and Triassic aquifers depending on water flow direction. Numerical model simulations have shown that groundwater of the Triassic aquifers, discharged by wells located about 2.5–3 km SW from the waste disposal sites, could be contaminated. The important, large water intakes, situated at a distance of about 5–9 km NW from the sites, are practically safe.

Key words: industrial wastes, multiple aquifer, boron, contamination, modelling.

Abstrakt. W rejonie zwałowisk likwidowanych Zakładów Chemicznych „Tarnowskie Góry" stwierdzono zanieczyszczenie wód wielowarstwowego systemu wodonośnego, wyrażające się podwyższonymi stężeniami m.in. boru, baru, strontu, siarczanów i chlorków. Za szczególnie niepokojące uznano bardzo wysokie stężenia boru w poziomach wodonośnych czwartorzędu (do 240 mg/dm^3) i triasu (wapień muszlowy) (do 116 mg/dm^3).
Dla oceny aktualnego i perspektywicznego rozprzestrzeniania się boru w analizownym wielowarstwowym systemie wodonośnym wykonano badania modelowe, obejmujące model numeryczny warunków hydrodynamicznych (cztery warstwy przepuszczalne: dwie w czwartorzędzie i dwie w triasie, przedzielone 3 warstwami półprzepuszczalnymi) i model migracyjny (dla boru).
W celu określenia udziału zanieczyszczonych wód wapienia muszlowego w całkowitym przepływie wód poziomu retu wykonano także modelowanie hydrogeochemiczne. Wyniki modelowania hydrogeochemicznego wskazują, iż w obszarze oddziaływania składowisk Zakładów Chemicznych udział zanieczyszczonych wód wapienia muszlowego w strumieniu przepływu wynosi w poziomie retu od 12 do 22%. Stwierdzono także wyraźne zróżnicowanie intensywności migracji boru w obrębie czwartorzędowego i triasowego piętra wodonośnego, w zależności od kierunków przepływu wód. Wyniki symulacji transportu boru wskazują na możliwość zanieczyszczenia wód w ujęciach zlokalizowanych w odległości 2–3 km na SW od zwałowisk oraz praktyczny brak tego zagrożenia dla ujęć położonych w odległości 5–9 km na NW od tych składowisk.

S³owa kluczowe: odpady przemysłowe, wielowarstwowy system wodonośny, zanieczyszczenie, bor, modelowanie.

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INTRODUCTION

Due to the casual locating of a waste site with no environmental considerations taken into account, the Tarnowskie Góry region represents a perfect example of a negative environmental impact. The casuality of localisation and the naturally high groundwater pollution vulnerability have 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 spreading of the contamination (Fig. 1).

The high concentration of boron is perceived as particularly dangerous since it reached 240 mg/dm^3 in the Quaternary aquifer, and 116 mg/dm^3 in the Triassic one. This critical situation resulted in a closure of many water intakes. Therefore, a complex remediation of the area, together with relocation of wastes has been performed.

In order to assess a current and perspective boron spreading in the groundwater of the analysed multi-layered flow system, numerical modelling has been applied. Numerical models of the four-layers system were worked out for a groundwater flow and a single component (for boron) solution transport (Kowalczyk *et al*., 2003). The hydrogeochemical modelling has been used for estimation of the contaminated water share in the Muschelkalk aquifer and in the total flow of the Roethian one (Rubin, Samborska, 2005).

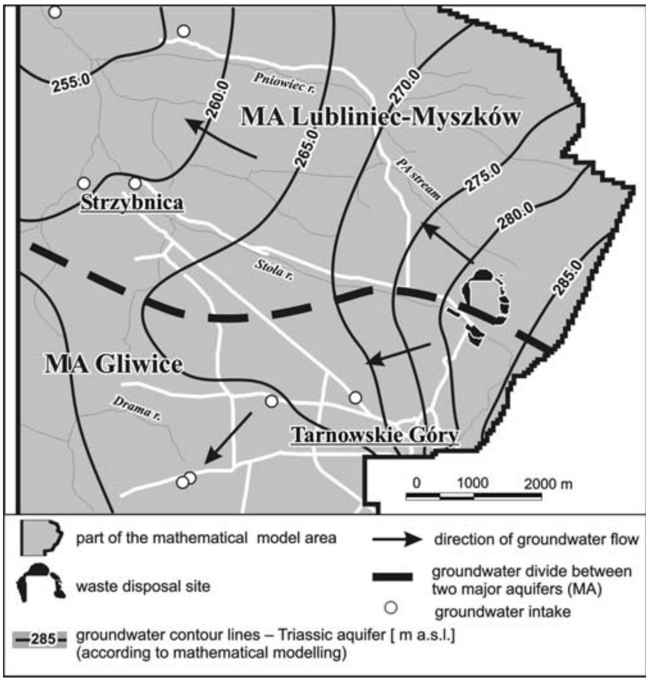

Fig. 1. Localisation of the study area and groundwater features

WASTE DISPOSAL SITE

The region of the Chemical Works "Tarnowskie Góry", was an area where diverse industrial activities took place for many years. That concerns a silver and lead ore mining (from the 12th century), iron milling (the second half of the 19th century), manufacture of silk paper (1892–1919), and production of paints and chemicals (1921–1995). The Chemical Works "Tarnowskie Góry" that have manufactured over 30 various inorganic chemicals for over 75 years, among them barium, boron, zinc, copper, and strontium, have proved to be extremely environmentally hazardous.

The obsolete technologies used there, 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, the waste dumps containing dangerous compounds of barium, boron, strontium, zinc, and copper have been developed. The dumps covered an area of over 27 ha, and their mass was estimated at about 1.4 million tonnes (Fig. 1).

Analyses of the selected samples of wastes have revealed their diverse chemical compositions with a dominance of sulphates (up to 49.600 mg/kg d.m.), calcium (up to 31.170 mg/kg d.m.) and magnesium (up to 10.840 mg/kg d.m.). The boron content ranged from 65.3 to 2.216 mg/kg d.m., barium – from 81.9 to 443.6 mg/kg d.m., strontium – from 1.646.7 to 8.576.0 mg/kg d.m., copper – from 93.9 to 9.556 mg/kg d.m., zinc – from 305.9 to 9.500 mg/kg d.m., and arsenic from 95.3 to 901.8 mg/kg d.m. (Rubin, 1999).

The wastes were characterised by a diverse but generally high active porosity (7–50%) that decreased with their age. The lower porosity features, the earlier stored wastes. Yet, the wastes did not constitute any essential barrier to the vertically infiltrating water (the hydraulic conductivity of the investigated wastes ranged from 6.25×10^{-8} to 6.4×10^{-7} m/s) (Rubin,1999). Moreover, the karst-fissure character of the rocks and their secondary permeability has increased because of many centuries of mining activity. It has facilitated the pollutants generated by the wastes to spread within the carbonate karst-fissured aquifer. The dynamic leaching tests conducted on the 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 showed also a high leachability of sulphates (up to 4989 mg/l) and chlorides (up to 1517 mg/l) (Rubin, 1999).

The Chemical Works have ceased their production activity in 1995, and were subjected to a liquidation process. In mid-2000, the implementation of a project of the complex neutralisation of the wastes, together with polluted-land reclamation, has begun. The scope of the work planned a construction of the Central Waste Disposal Facility (CWDF) at the area of 14.11 ha, with a storage capacity of about 1.5 million m^3, and relocation of all wastes and the polluted ground from beneath the waste dumps to the facility. CWDF is a modern facility that satisfies all ecological requirements. It will be an above-ground-ranging object (17 m above the ground surface after completion). It has five cells with sealed bottom and draining systems.

HYDROGEOLOGICAL SETTING

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 the folded and faulted Palaeozoic basement. The Quaternary deposits of various lithology and thickness cover the Triassic formation. There are Quaternary and Triassic aquifers in the hydrogeological profile. The thickness of these aquifers varies from a few to more than 50 m, and from 40 to 150 m, respectively. Most frequently, the Quaternary aquifer consists of two water-bearing horizons with discontinuos extension. The hydraulic conductivity of the Quaternary aquifer varies from 0.1 m/24 h up to 15 m/24 h. This aquifer water is polluted and not utilised.

The Triassic karst-fissured carbonate aquifer (dolomites and limestones with marls interbeddings) is the most important and abundant. The hydraulic conductivity of this 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 deposits and two within the Triassic (Muschelkalk and Roethian) deposits (Fig. 2). The Quaternary aquifer is discharged by the Drama and Stoła rivers in local flow systems. In the shallower part of the Triassic aquifer (Muschelkalk), both local and regional flow systems have been formed, while in the deeper part (Roethian) – only regional ones. Groundwater flows out the watershed area to the NWW and SWW into the vicinity of the waste disposal sites (Fig. 1). A velocity of the groundwater flows 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.

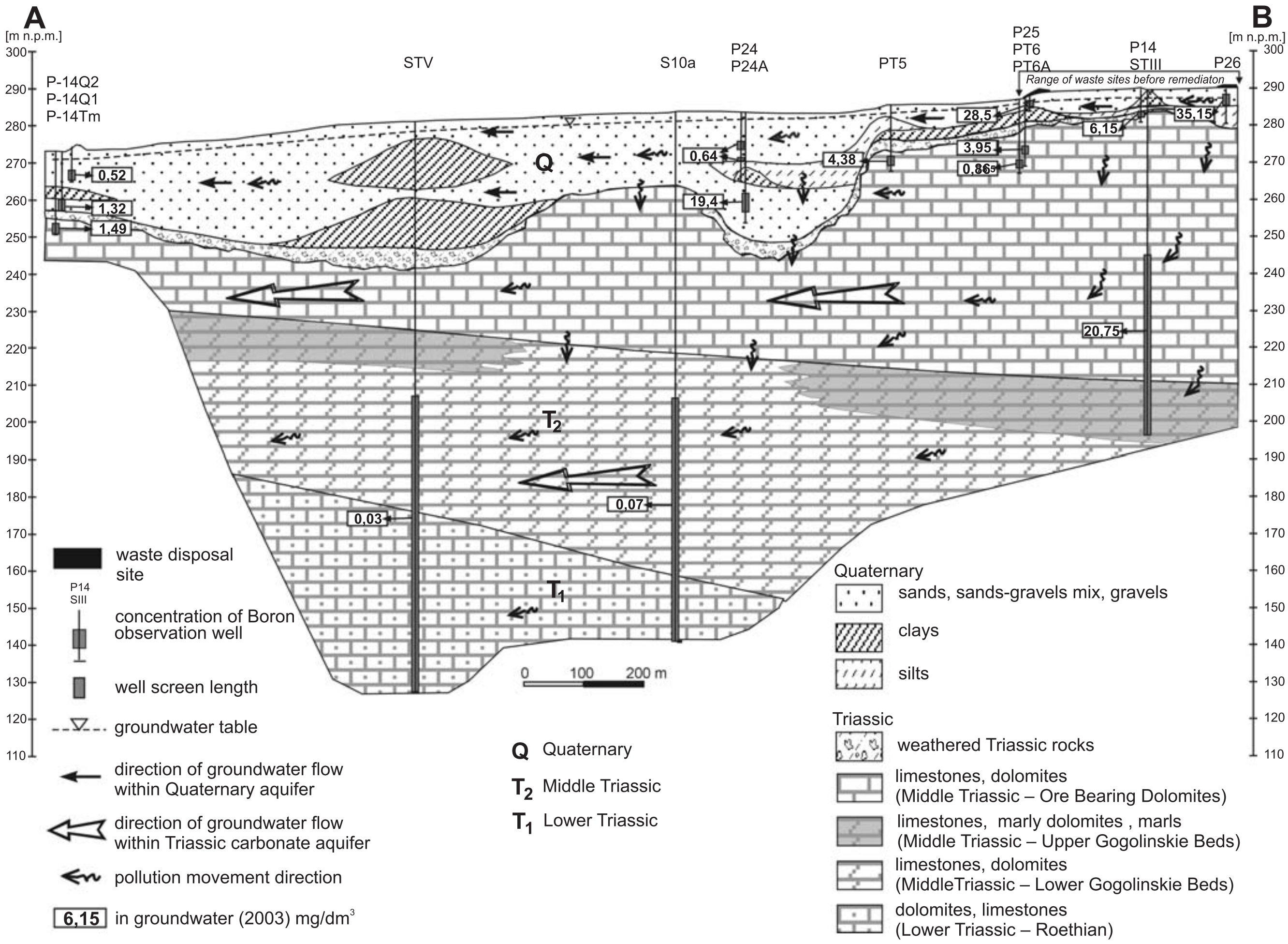

Fig. 2. Hydrogeochemical cross-section

GROUNDWATER QUALITY MONITORING

Since 1990 when it was brought into existence, the groundwater quality-monitoring network has been subjected multiple modifications in the discussed area . Unfortunately, the above mentioned sites have been systematically monitored only since 1999 (Rubin, Witkowski, 2002). The 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 from 4 to 28 metres deep in the Quaternary aquifer and from 15 to 155 metres deep in the Triassic one. There are only five groups of nested observation wells. Each of the group consists of 2 to 3 wells (altogether 12 wells).

Field measurements covered the determination of temperature, pH, specific conductance, and oxidation-reduction potential Eh. The range of chemical indicators determined at the laboratory tests and sampling frequency have been changing over the time. Currently, 24 indicators are determined: TDS, COD, $N\text{-}NH_4$, $N\text{-}NO_3$, Cl, SO_4, HCO_3, Ca, Mg, Na, K, Al, As, Ba, B, Cd, Cr, Cu, Fe, Sr, Zn, detergents (only for the Quaternary aquifer), trichloroethene, and tetrachloroethene (for the Triassic aquifer, only).

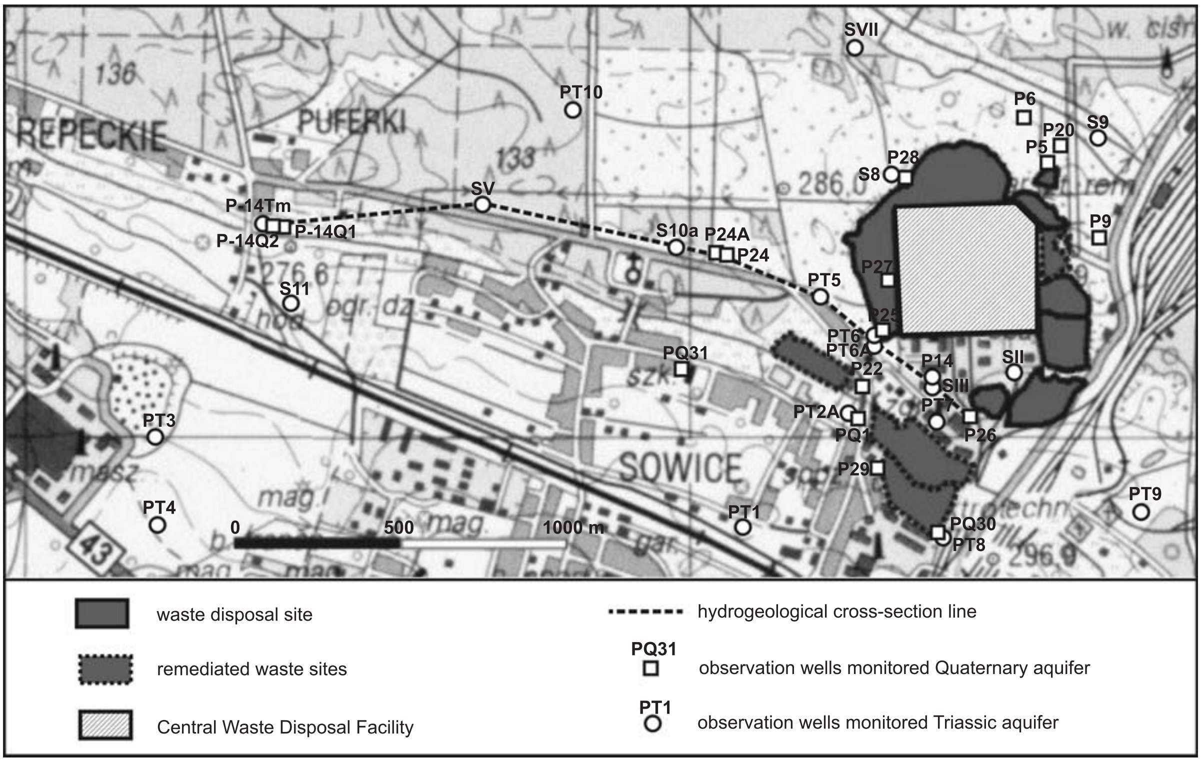

Fig. 3. Groundwater quality monitoring network

GROUNDWATER QUALITY

A groundwater quality in **the Quaternary aquifer** is differentiated but generally very bad. The highly contaminated water is observed in the waste site area showing the following characteristics: TDS – up to about 3.000 mg/dm^3, Cl – up to 868 mg/dm^3, SO_4 – up to 1.130 mg/dm^3, B – up to 240 mg/dm^3, Ba – up to 722 mg/dm^3, and Sr – up to 30.6 mg/dm^3 (Fig. 4). The groundwater temperature varies seasonally between 5°C (in winter) and 15.5°C (in summer).

In the waste disposal area, the average boron concentration has increased 60-times groundwater in the Quaternary aquifer, and the average strontium concentration 26-times, in comparison with the upgradient zone water (Fig. 5). The Quaternary aquifer groundwater shows also in that area increased concentrations of zinc, aluminium, cadmium, manganese, nickel, led, iron, ammonia, and nitrates. Generally, a similar groundwater quality of Quaternary aquifer is observed in upgradient areas as well as in downgradient ones (at a distance of about 2 km from the waste sites). That suggests a limited lateral migration of contaminants within the Quaternary aquifer.

Within **the Muschelkalk aquifer,** the groundwater's chemical variability depends on the location of the observation points with respect to the location of the disposed waste body,

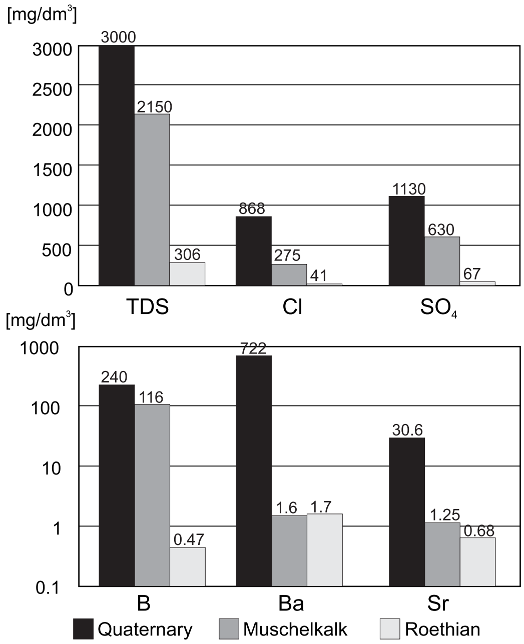

Fig. 4. Maximum concentration of selected indicators of groundwater pollution

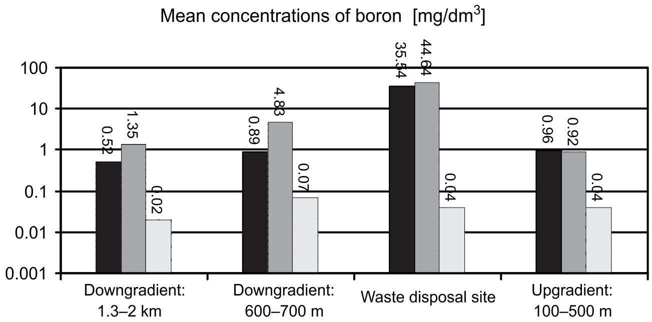

Fig. 5. Changes of boron concentration along the selected groundwater flow pathways

Explanations as in Figure 4

and its position in the aquifer's profile. The groundwater of this aquifer is generally less contaminated as compared to the Quaternary one (Fig. 4). Significantly, the contaminated water (locally even more than water in the Quaternary aquifer) was noticed in the upper part of the Muschelkalk, only within the waste disposal sites and in adjacent downgradient areas (up to about 1 km from the sites). TDS was there up to 2.150 mg/dm^3, Cl – up to 275 mg/dm^3, SO_4 – up to 630 mg/dm^3, B – up to 116 mg/dm^3, Ba – up to 1.6 mg/dm^3, and Sr – up to 1.25 mg/dm^3 (Rubin, Witkowski, 2003). Increased concentrations of ammonia, nitrates, manganese, iron, zinc, cadmium, and nickel have been also observed in the area of the waste disposal site.

The water of a better quality was observed in bottom parts of the Muschelkalk aquifer, in the area of the considered sites, and at a distance of up to about 2 km downgradient from the sites (TDS – up to 694 mg/dm^3, Cl – up to 78, 8 mg/dm^3, SO_4 – up to 127 mg/dm^3, B – up to 1, 49 mg/dm^3, Ba – up to 0.1 mg/dm^3, and Sr – up to 0.327 mg/dm^3). The groundwater temperature within the Muschelkalk aquifer is more stable and varies from 10 to 12°C.

The groundwater of the fourth, the lowest, aquifer (**Roethian**) has been practically uncontaminated by the described 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). It has a slightly lower and stable temperature, ranging from 9 to 10°C.

A vertical and horizontal differentiation of the boron concentration is observed in the groundwater of the considered multi-aquifer system. The maximum content of $H_3BO_3^0$ (major easily migrating boron speciation) in groundwater of the Quaternary and Triassic (Muschelkalk) aquifers, occurs in the waste dump area (Fig. 5). However, the maximum groundwater content of boron of the deepest Roethian aquifer is observed in the area of an 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). The vertical differentiation of the groundwater boron content is very well noticed in the nested piezometers (Fig. 6). The mean concentrations of boron in the

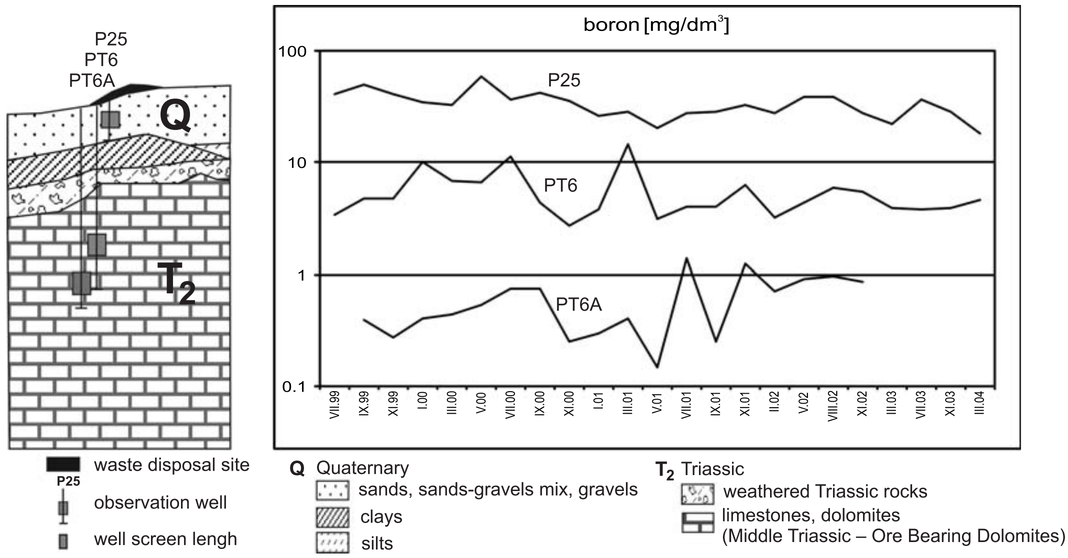

Fig. 6. Changes of boron concentration in groundwater in the selected nested piezometers

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 five-year-period, from 1999 to 2003, a general improvement of the groundwater quality of the Quaternary aquifer and some relative stabilisation 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 Figure 7.

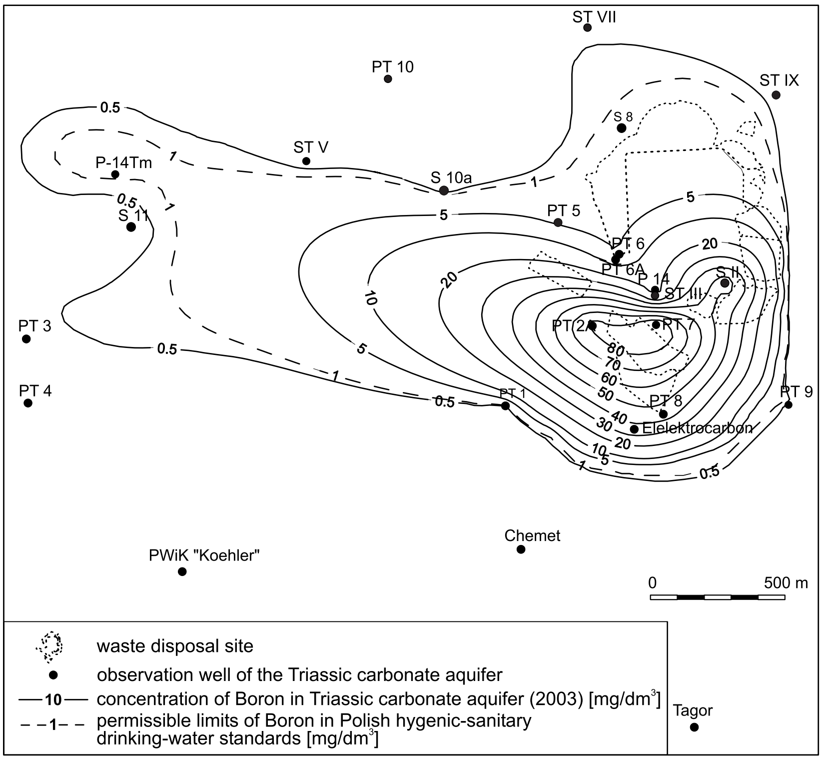

Fig. 7. Spatial distribution of the boron concentration in the Triassic aquifer (based on the monitoring data, 2003)

MODELLING

A groundwater-flow model and a solution-transport model were developed for the multi-aquifer system. A groundwater-flow system was simulated in three dimensions using the MODFLOW-96 computer code (McDonald, Harbaugh, 1988; Pollock, 1994; Zheng, Wang, 1999). MODFLOW was used in a 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 solution–transport model. The model cell size was 50 × 50 m; therefore, the models were subdivided into the 231 rows and 113 columns. They were also vertically divided into four layers (two Quaternary and two Triassic) 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 the boron concentration in the system under consideration was a result of its infiltration from the wastes, which have accumulated in the last 70 years. The simulated boron concentrations for that period were in acceptable agreement with the measured data.

A hydrogeochemical modelling was also performed in order to recognise in detail the distribution of the contamination in the multilayer aquifer system, and to determine the share of the contaminated Muschelkalk water in the whole Roethian aquifer horizon (Rubin, Samborska, 2005). The following programmes have been used: NETPATH (Plummer *et al.*, 1994) and PHREEQC (Parkhurst, Appelo, 1999). This modelling was performed for a flow starting from the waste disposal area and ending at the large water intakes in the Triassic carbonate formation. The current line was selected on the base of the hydrodynamic modelling (Kowalczyk *et al.*, 2003) (Fig. 8).

A reverse modelling of the processes taking place during the groundwater flow along the selected current line was performed with the assumption that the mixture of water from well SIII (the Muschelkalk horizon) and well S10a (the Roethian horizon) constitutes initial solution, and water from well SV (the Roethian horizon) constitutes the final solution (Rubin, Samborska, 2005). In the model, it was assumed that on the way of the flow, carbonate minerals like calcite, dolomite, and siderite as well as barium sulphate (barite) undergo dissolution.

In the conceptual model, the possibility of an ion exchange between ions of magnesium, potassium, and sodium was taken into account because of the large differences of concentration of these components and impossibility of connecting the parameters with aluminosilicate phases in the situation of lack of silicon content determinations in the water. The conceptual model also assumed a possibility of the application of other phases, like goethite and smithsonite as well as to enable mass balance for chlorides and sulphates, halite, and epsomite in the modelling. The boron was not taken into account in the model, as it was a characteristic contamination indicator and because of no possibility of connecting the component with existing mineral phases (Rubin, Samborska, 2005).

RESULTS OF MODELLING

The water table layout and directions, speeds and time of flows in the investigated aquiferous formation and in its individual layers were analysed in the simulation tests, performed with the help of a mathematical model. The investigations showed that the Quaternary aquifers were drained by surface watercourses, and they constituted a shallow water circulation system.

Results of groundwater monitoring as well as numerical modelling point out that an intensive downward groundwater flow predominates within the first Quaternary aquifer in the area of the waste disposal sites. Within the lower Quaternary aquifer, downward and locally horizontal flows were observed. The both Quaternary aquifers were of the transit type, being a source of the lower Triassic aquifers recharge. The horizontal flow in the Triassic aquifers was predominant while within the upper one (Muschelkalk), some weak downward component was also observed. The mentioned vertical component enabled recharge of the deepest Roethian aquifer, and could lead to groundwater mixing of the both Triassic aquifers.

In order to apply hydrogeochemical modelling as a method of assessing the mixing of anthropogenically contaminated groundwater, it was indispensable to develop an appropriate conceptual model. In the situation of the incomplete knowledge of elements, indispensable for that conception, an achievement of a highly authoritative result is limited. Yet, application of two different programmes to the modelling gave a relatively small quantity of solutions, which were characterised by high convergence of results.

Based on the developed models (3 models based on the NETPATH programme and 6 models – on the PHREEQC programme), it may be assumed that the share of the contaminated Muschelkalk water in the Roethian horizon flow ranges from 12 to 22% in the area of influence of the waste disposal site of the chemical works. Models representative for the particular water mixing proportions have been prepared. For each mixing proportion, one of three models, in which both calcite and dolomite are dissolved, was selected to make a presentation. The results of the hydrogeochemical modelling may be applied to verify a model of the hydrodynamic, multi-layer aquiferous system.

The results of hydrodynamic modelling practically indicated an unlimited water flow within the Triassic aquifers,

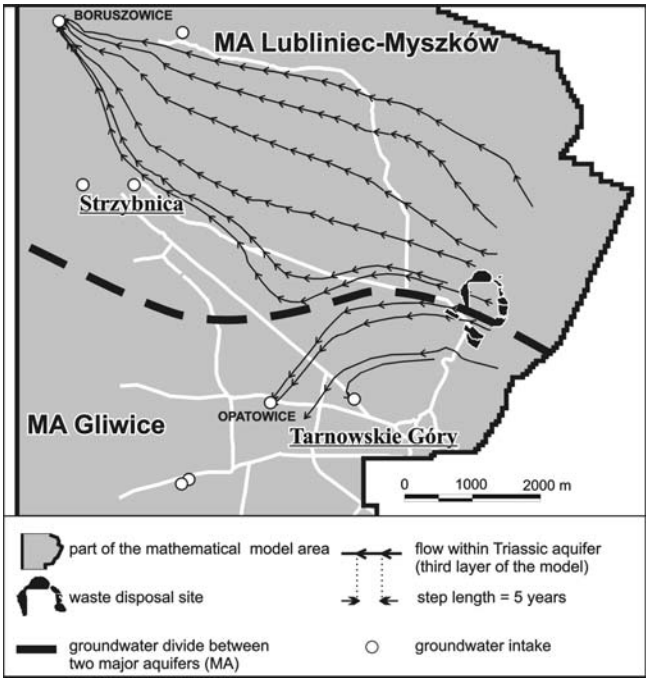

Fig. 8. Groundwater flow pathways according to the numerical modelling

however, the natural regional groundwater flow pattern is modified by active wells and old mine workings. The outflow pattern of the Triassic aquifer water from the waste disposal area shows that NW and N directions dominate with a slight SW component (Fig. 8) (Witkowski *et al*., 2005a). These are directions, in which the intakes exploiting groundwater from the Triassic formation are situated (Boruszowice, Opatowice). The simulation results indicate that with the current water circulation, the water flowing from the waste disposal area does not recharge the Staszic intake, where presently no increased boron and barium contents are registered.

Another essential goal of the performed modelling tests was to make a simulation of the boron migration in the investigated multi-layer aquiferous system. The modelling was expected to show current and future spreading of boron concentration in the groundwater system. Its first phase consisted of reconstructing the current spatial distribution of boron concentration by means of monitoring investigations (for the year 2002). In its second phase, a forecast of boron spreading within the analysed system, up to the year 2030, was drafted. The simulated current boron concentrations within the Muschelkalk aquifer stay in acceptable agreement with the measured data.

A significant differentiation of the boron migration intensity has been observed within the Quaternary and Triassic aquifers, depending on water flow direction. The above mentioned, practically unlimited water outflow within the Triassic aquifers, 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 was noticed.

The results of groundwater quality monitoring and simulated groundwater flow, and advective boron transport suggested that groundwater of the Triassic aquifers, discharged by wells located about 2.5–3 km SW downgradient from the waste disposal sites, could possibly be contaminated in 25–30 years (Witkowski *et al.*, 2005b). The important, large 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 wells field was estimated for about 90 years (Fig. 8).

CONCLUSIONS AND FINAL REMARKS

The waste disposal sites under consideration have caused a significant groundwater contamination in the Quaternary and Triassic aquifers. The current state of groundwater chemistry in this region is a result of many years of negative impact of those sites. The authoritative assessment of the present groundwater quality and migration of contaminants in the entire multi-aquifer formation complex was possible due to the comprehensive investigations – both the on-site groundwater monitoring and the multi-aspect mathematical modelling, including a groundwater-flow model and a solution-transport model, as well as the hydrogeochemical modelling.

The investigation results proved a limited spatial range of the contamination migration within the Quaternary aquifers, and indicated some general improvement of the groundwater quality of these aquifers. However, the reported contamination of the significant Triassic aquifers constitutes an essential problem. It mainly concerns the Muschelkalk aquifer where the important groundwater contamination was reported from the vicinity of the waste disposal sites.

An important and at the same time optimistic fact is that the groundwater of the lower Triassic aquifer (Roethian) is practically uncontaminated. With its large water abundance and with only a slight component of the inflow of water from the upper Muschelkalk aquifer, the hazard that quality of the groundwater will be negatively influenced by the contaminants migrating from the near-surface zone is very low. Another optimistic fact there is a relative stabilisation of the groundwater quality within the Muschelkalk aquifer. Processes of the natural attenuation and remediation work should result in fastening the groundwater quality improvement in all the aquifers.

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