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The Influence of Anthropogenic Contamination on the Groundwater Chemistry of a Semi-confined Aquifer (The Wielkopolska Buried Valley Aquifer, Poland)

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Abstract The problem of the identification of the influence of anthropogenic contamination on the groundwater chemistry of a semi-confined buried valley aquifer is presented. It has been shown that the symptoms of groundwater chemistry transformation due to anthropogenic contamination appear, even though confined conditions occur. Statistical techniques have been used for the identification of this problem. Factor analysis has been used to recognize the most vulnerable parts of the aquifer. Based on the factor scores, data sets characterizing natural and pollutant groundwater chemistry were distinguished. The comparison of the "natural" groundwater chemistry with the whole data set (by means of the correlation analysis and regression analysis) enabled the identification of the components which are the most sensitive to anthropogenic impact. The changes of groundwater chemistry over time identify the intensity of the anthropogenic impact.

Keywords Groundwater contamination · Groundwater chemistry
Wielkopolska Buried Valley aquifer · Poland

1 Introduction

In the recent years, there has been an apparent tendency for the quality of groundwater to deteriorate, which has been caused by anthropogenic contamination. The most intensive contamination occurs on rapidly urbanized areas where the intensive exploitation of groundwater for industrial and domestic purposes takes place, thus contributing to a high downward gradient. These conditions may accelerate the migration of contaminants from the land surface to the aquifer (Lawrence et al. 2000; Jeong 2001). In regions where the intensive exploitation of groundwater does not take place, contamination may take places when a natural downward gradient occurs. The contamination of groundwater may appear

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even if the aquifer is overlain by a thick confining layer (Simpkins and Bradbury 1992). This is particularly the case in long-term sources of contamination influence, which date back even as far as previous centuries (e.g. influence of unsewered urban and rural areas), where contaminants may appear in deep groundwater.

Within the Polish Lowland area, symptoms of the contamination of deep confined aquifers, e.g. buried valleys, are still more often visible (Gorski 1989; Paczynski 2002). Very often the endangered aquifers are these, which are the most attractive and unique for domestic purposes or food industry demands.

The widely used tools for the identification of groundwater contamination are statistical techniques, particularly multivariate statistics such as factor analysis (Grande et al. 1996; Subbarao et al. 1995; Abu-Jaber et al. 1997). Factor analysis (FA) enables both the classification of groups of data set and hydrochemical facies investigation and also the interpretation their origin (Dalton and Upchurch 1978; Lawrence and Upchurch 1983).

The methodology of FA application for the investigation of the groundwater chemistry origin of the Wielkopolska Buried Valley aquifer (Poland) is presented in the work of Dragon (2006). The present paper focuses on the interpretation of the influence of contamination on groundwater chemistry.

The objectives of this paper are: (1) to demonstrate the impact of contamination on the groundwater chemistry of a semi-confined aquifer (covered with a thick glacial tills complex); (2) the identification of the groundwater components, which are the most sensitive to contamination; (3) the identification of areas with the most noticeably contaminated groundwater.

2 Study Area

2.1 Hydrogeological Setting

The research was conducted in the Wielkopolska Buried Valley aquifer (WBV), covering an area of approximately 1,000 km^2 (Fig. 1).

The thickness of water bearing sediments ranges from 20 to 50 m. These sediments are mainly composed of sand and gravel (Fig. 2). In the bottom part mainly fluvioglacial gravels and coarse sands occur (hydraulic conductivity ranges from k=1.9E-4 to k= 1.0E-3 m/s), while the upper part of the aquifer is composed of medium and fine sands of fluvial genesis (hydraulic conductivity from k=8E-5 to k=5.5E-4, Dabrowski 1990). The wells tapping the WBV aquifer have screens installed mainly in the lower part of the aquifer. The confining layer is composed of glacial tills. The thickness of the aquitard exceeds 50 m in the till plain regions and decreases to less then 20 m, where intertill aquifers and glacial troughs occur (Fig. 3). In the area of the Warta river valley, the aquitard does not occur. The layer of Tertiary clays occurs in the aquifer bottom and isolates the WBV aquifer from the Tertiary aquifers.

The main recharge area is located in the region of the Lwówek-Rakoniewice Rampart, where the groundwater flows westward to the Obra valley and eastward to the Warta valley. The principal source of the recharge is the percolation of groundwater through glacial tills and upper intertill aquifers. The recharge by the inflow from the intertill aquifers, located to the north of the WBV aquifer, also takes place.

The WBV aquifer is characterized by variable vulnerability. The regions of glacial troughs and the areas where intertill aquifers occur (mainly in the region of the Lwówek-

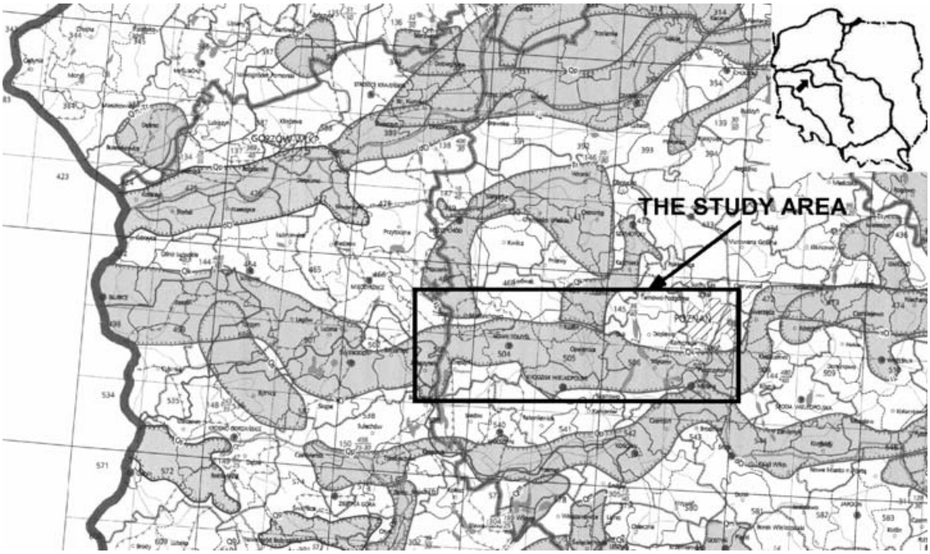


Fig. 1 Location scheme. The areas of the Major Groundwater Basins (MGWB) occurrence are marked (after Kleczkowski, 1990)

Rakoniewice Rampart are the most vulnerable ones, owing to the fact that the aquitard thickness decreases there to less than 30 m (Fig. 3). The intertill aquifers facilitate the migration of contaminants, particularly in the regions of downward gradient appearance (Dragon 2004).

2.2 Land Use Pattern and Urbanization

The examined area is used for various purposes. The eastern part of this region (to the east of Grodzisk Wielkopolski) is dominated by agriculture, while in both the central (recharge area of WBV aquifer) and the western part forested lands prevail. Additionally rural and urban built-up areas occur. There are 5 towns (5,000÷15,000 inhabitants) located in the investigated region.

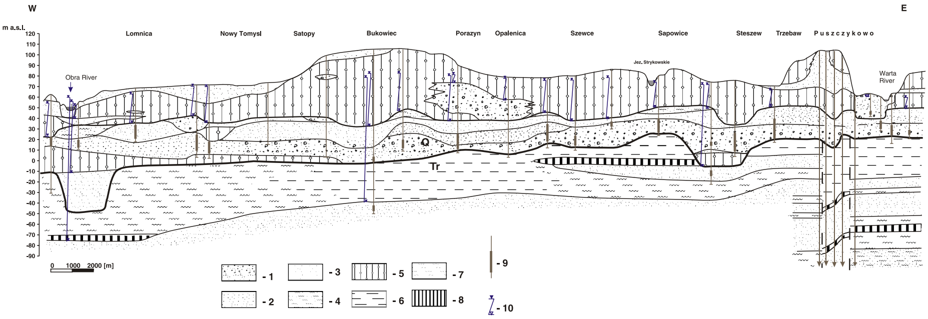


Fig. 2 Hydrogeological cross-section. *1* – coarse sand and gravel; *2* – medium sand; *3* – fine sand; *4* – silty sand; *5* – till; *6* – clay; *7* – silt; *8* – brown coal; *9* – the well screen; *10* – groundwater level; *Q* – Quaternary; *Tr* – Tertiary

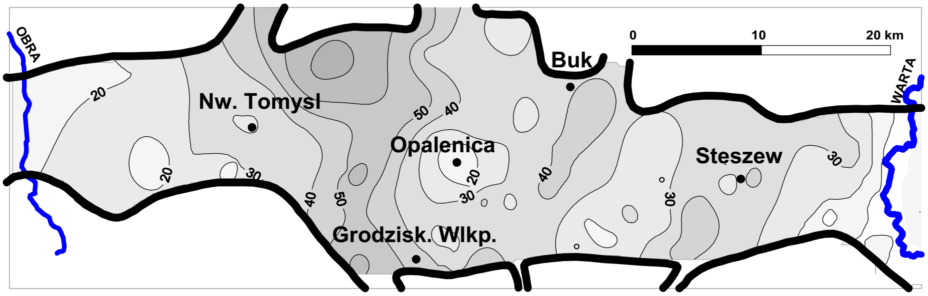


Fig. 3 The thickness of the Wielkopolska Buried Valley aquitard (in meters)

Sources of pollution typical for the whole of Great Poland are present in the studied area. The most significant source of pollution is that of untreated sewage from both rural and urban lands – a long-lasting problem that has still not been solved. Another most important source of contamination, connected with agriculture, are livestock farms. Livestock manure is in most cases spread on the fields. The main risk as regards concerning cultivated land is the excessive usage of fertilizers.

3 Materials and Methods

3.1 Sample Collection and Data Analysis

In order to investigate the groundwater chemistry of the WBV aquifer, the results of a physico-chemical analyses of water sampled from 61 drilled wells were used (Dragon 2006). Only continuously pumped productive wells were chosen for the analyses, which helped to obtain representative samples. The wells chosen were continuously pumped for at least several hours prior to examination. In the region of high well density (urban areas) the most representative wells were chosen evenly localized on the map, which facilitates a spatial interpretation of the FA results and water chemistry variability.

Some parameters were measured in the field using flow-through cell (electrical conductivity, pH and temperature) and by titration (alkalinity). The odour and colour of water were measured as well. The water samples were collected in polyethylene containers and on the same day transported to the Adam Mickiewicz University – Department of Water and Soil Analysis Laboratory in Poznan. Samples for Fe and Mn analyses were acidified by 20% HCL and for nutrients chloroform was used. Before completion of the analysis the samples were stored in the laboratory refrigerator.

The list of parameters measured in the laboratory with detection limit and precision (in brackets) are listed below: oxygen consumption – 0,7 mgO_2/L (5%), N–NH_4 – 0,04 mg/L (10%), N–NO_3 0,1 mg/L (20%), Na and K – 0,1 mg/L (5%), Cl and Mg – 5 mg/L (5%), SO_4 – 10 mg/L (5%), Fe 0,001 mg/L (10%), Mn – 0,003 mg/L (10%), Ca – 2 mg/L (5%), TH – 0,36 mval/L (5%), TDS (determined by evaporating) – 1 mg/L (5%). For quality control measures the ionic error balance was calculated. The calculated error does not exceed 3%.

The determination of the chemical parameters in the laboratory was performed with use of an ionic chromatograph Dionex DX-120 (anions) and atomic absorption spectrometer

Perkin Elmer Analyst 300 (cations). The samples were prepared using a microwave mineraliser Merck MW 500.

Moreover, 225 archival physico-chemical analyses performed during the pumping tests in wells that were built in the period between 1957 and 1998 were also analysed.

3.2 Data Processing

The factor analysis method was used for the identification of the hydrogeochemical processes controlling groundwater chemistry. As the extraction technique the principal component analysis was applied. Prior to analysis, the initial data set was standardized by criteria presented by Davis (1973). Subsequently, the factor loadings matrix was rotated to an orthogonal simple structure according to *Varimax* rotation. The high factor loadings were obtained for the variables correlated in the factor (close to 1 or −1) and low factor loadings (close to 0) for the remaining variables (Davis 1973). To establish the number of factors the Kaiser criterion was applied. Those factors which best describe the variance of the analysed data (eigenvalue >1) and which can be reasonably interoperated were accepted for further analysis (Drever 1997).

Two groups of hydrogeochemical processes (geogenic and anthropogenic) were determined. The regions characterized by the most noticeable anthropogenic transformation of groundwater chemistry were identified. Based on the factor scores, subsets characterising natural and pollutant groundwater chemistry were established. It was shown that these subsets are characterized by significant differences in groundwater chemistry. The comparison of these two subsets (made by means of correlation analysis and regression analysis) enabled the identification of components that clearly reflect the influence of anthropogenic pollution. The comparison of groundwater chemistry from the period when the wells were being constructed and the current ground water chemistry indicated the tendencies of groundwater chemistry transformation caused by anthropogenic pollution. The comparison of the chemistry variation in different ranges of the aquitard thickness allowed the most vulnerable parts of the aquifer to be identified.

The testing of the normality of data distribution was the preliminary step of the statistical analysis, performed mainly by the graphical method (Rigg 1989; StatSoft, Inc. 2003). The majority of parameters analysed had a non-normal type of distribution (Table 1). The principal component analysis was used as an extraction technique, because the data does not need to be normally distributed and it is the applicable method for environmental studies (Abrahams 1972; Brown 1998; Stetzenbach et al. 2001). For the same reason the correlation analysis was used with the aid of graphical methods (Reimann and Filzmoser 1999).

4 Results and Discussion

The results of FA of the data presented in Table 1 indicated the hydrogeochemical processes controlling groundwater chemistry. Four factors, which describe two groups of hydrogeochemical processes (geogenic and anthropogenic), were extracted. The first two factors are the most important (Fig. 4), which explain the 41.2 and 16.7% (respectively) of variance. The detailed interpretation of factors is presented in the work of Dragon (2006). Here the interpretation and further analysis of the first factor, connected with the influence of contamination on groundwater chemistry is presented.

Table 1 Statistical characteristics of the parameters used for factor analysis calculation

Parameter		Min.	Max.	Average	Median	Standard deviation	Skewness
Colour	mg Pt/L	0	80	25	20	15.5	1,8
PH		7.00	7.95	7.50	7.50	0.2	1,4
Oxygen consumption	mg O_2/L	1.3	7.0	3.1	3.0	1.3	1,2
Total hardness	mval/L	2.2	10.7	5.8	5.8	1.7	0,3
Alkalinity (HCO_3)	mval/L	1.7	6.9	5.0	5.2	1.3	−0,55
TDS	mg/L	179	672	385	372	110	0,5
Cl	mg/L	7	84	23	15	18.6	1,8
SO_4	mg/L	<10	175	42	33	35.8	1,6
N–NH_4	mg/L	0.04	0.6	0.19	0.14	0.13	0,6
Fe	mg/L	0.05	13.1	4.1	3.8	2.4	1,2
Mn	mg/L	<0.003	0.57	0.17	0.16	0.09	1,9
Na	mg/L	4.4	22.2	11.0	10.4	4.0	3,8
K	mg/L	0.9	6.8	3.0	2.9	1.2	3,3

Frequency *n*=61

Factor 1 contains the highest factor loadings of Cl, SO_4, high factor loadings (>0.7) of TH and TDS and low factor loadings for the remaining parameters. The relatively high (negative) factor loading includes also the thickness of the aquitard. This observation suggests that the concentrations of these parameters are higher in the regions of relatively small aquitard thickness and vice versa.

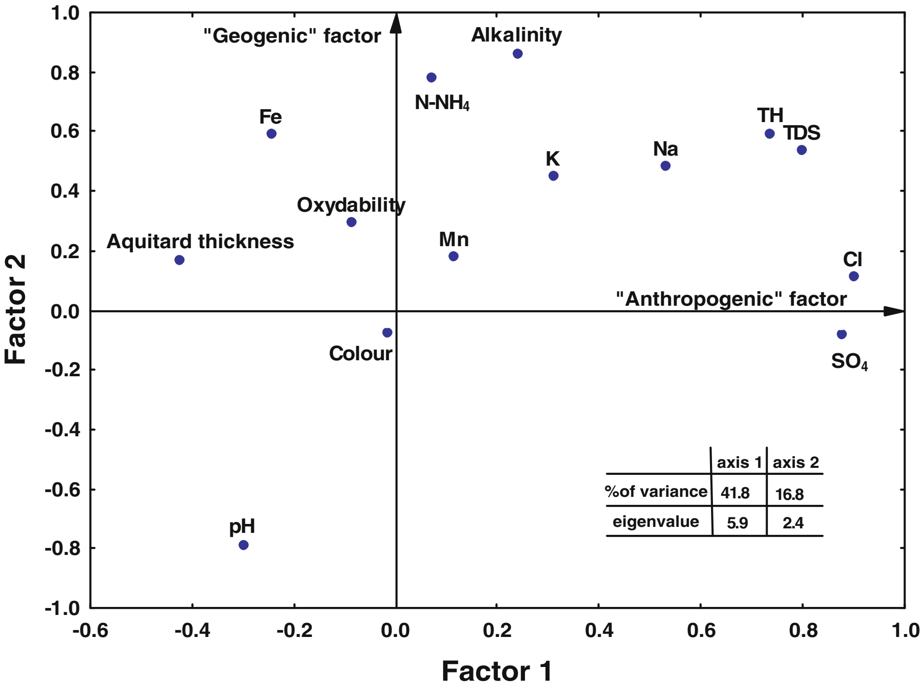


Fig. 4 The plot of factor loadings for the first two factors

Factor 2 contains the highest (>0.7) factor loadings of N–NH_4, alkalinity and pH (negative). The TDS, TH, Fe and K have high factor loadings as well (Fig. 4). The contribution of these parameters reflects organic matter decay in an anaerobic environment. The influence of this process provides a more effective solution of geological environment components (Gorski 1989). The high factor loadings of TH (i.e. Ca and Mg compounds), alkalinity and TDS reflect the Ca–HCO_3 and Ca–Mg–HCO_3 water types characteristic for the WBV aquifer. High (negative) factor loading of pH reflects the influence of acid–base factors on groundwater chemistry (Dragon 2003). The structure of this factor reflects natural hydrogeochemical processes, therefore this factor is called "geogenic."

The highest factor scores of factor 1 occur in the regions where it was observed that the high concentration of the parameters grouped in factor 1 increases with time (Fig. 5). This factor is associated with the impact of the contamination from the land surface and thus it is called "anthropogenic" (Dragon 2006). The factor score values can be treated as contamination indicators. The regions of high factor score values point to the most noticeable anthropogenic transformations of groundwater chemistry and to the high vulnerability of the WBV aquifer. The natural hydrochemical background of Cl (estimated on the basis of the data obtained during the pumping tests) did not exceed 20 mg/L. At present, the concentration of Cl in these regions exceeds 80 mg/L.

The influence of anthropogenic contamination is observed in both urban and rural areas. The most distinct effect were noted in the eastern part of the study area, an area of intense farming and where unsewered rural land of long-term influence occurs. This is also the area in which the thickness of the aquitard that is less than 25 m. In the vicinity of Opalencia (recharge area) where the thickness of the aquitard does not exceed 20 m there is no evidence of anthropogenic contamination due to the nature of the land use – dominantly forest. In the area of intense agricultural development, where the thickness of the aquitard is greater, anthropogenic contamination is also absent.

The factor scores of factor 1 ("anthropogenic" factor) were applied to distinguish the subsets of chemical analyses characterizing respectively "natural" and contaminated groundwater. The distinguished subset of the factor scores <0 characterize "natural" groundwater chemistry, whereas the remaining part of the data set characterizes contaminated groundwater. Afterwards, a comparison of the "natural" groundwater chemistry with the whole data set was performed, using the correlation and regression analyses.

The analysis clearly indicates that the relations between variables performed for the whole data set are different from those that characterize the "natural" groundwater data set.

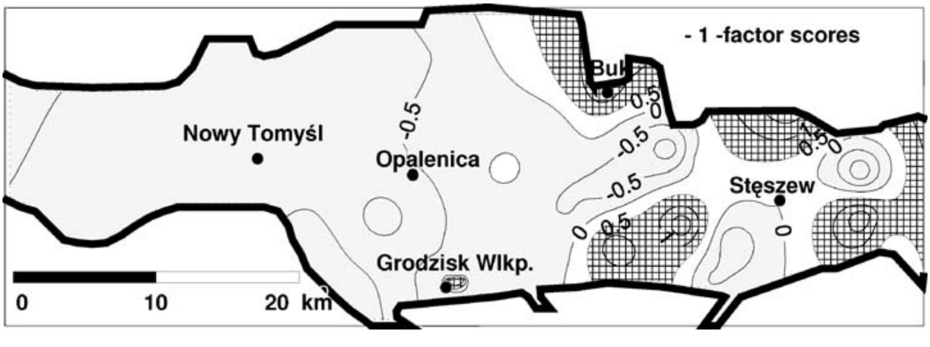


Fig. 5 The distribution of the factor scores ("anthropogenic" factor). The areas of factor scores >0,5 occurrence are marked

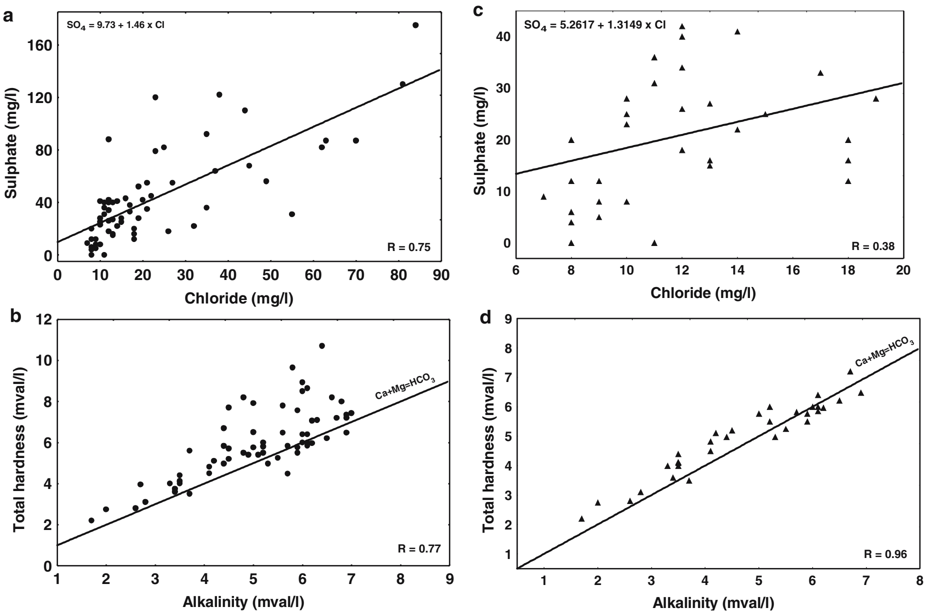


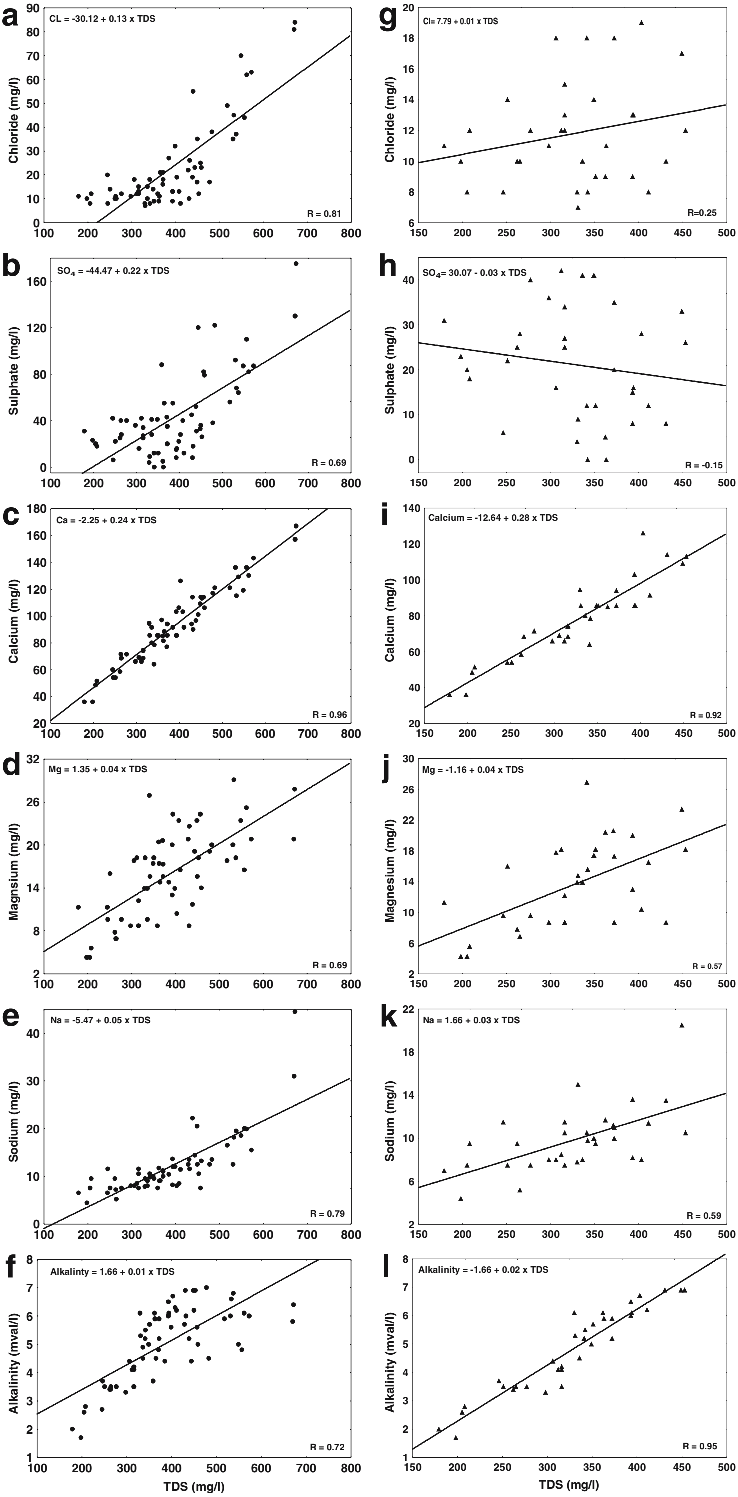
Fig. 6 The scatter diagrams of some hydrochemical parameters (whole data set – *left graphs*, n=61; “natural” groundwater – *right graphs*, n=33; α=0,05)

The strong correlation between chloride and sulphate (Fig. 6a) for the whole data set is notable. Both components are strongly connected with TDS (Fig. 7a,b), as well as with sodium concentration (Fig. 7e). Sodium concentration correlates with chloride as well (data not presented, R=0,7, α=0,05), whereas the lack of correlation between chloride and sulphate (Fig. 6c) and both components with TDS (Fig. 7g,h) is characteristic of the “natural” groundwater. The strong correlation between calcium, magnesium and alkalinity with TDS is apparent for the whole data set (Fig. 7c,d,f) as well as for the “natural” groundwater (Fig. 7i–k).

The plot of TH vs. alkalinity (reflecting HCO_3 concentration) indicates, that the vast majority of the data points fall above the equal line ($Ca+Mg=HCO_3$), which implies a surplus of TH (i.e. Ca and Mg) with relation to alkalinity (i.e. HCO_3) for whole data set (Fig. 6b). Higher TH (relative to alkalinity) is characteristic of the data points with high chloride and sulphate concentrations. This observation may suggest the occurrence of non-carbonate hardness balanced by high sulphate concentration, which is caused by anthropogenic contamination. For the “natural” groundwater chemistry TH and alkalinity are balanced and most of the data points fall near the equal line $Ca+Mg=HCO_3$ (Fig. 6d).

The results illustrate that from among the major groundwater components the anthropogenic input is reflected mainly by concentrations of chloride and sulphide. The

Fig. 7 The scatter diagrams of TDS and major ions (whole data set – *left graphs*, n=61; “natural” ▶ groundwater – *right graphs*). R – correlation coefficient; the equation of regression in upper left part of each graphs are presented; n=33; α=0,05


a
CL = -30.12 + 0.13 x TDS
R = 0.81
Chloride (mg/l)
g
Cl= 7.79 + 0.01 x TDS
R=0.25
Chloride (mg/l)
b
SO$_4$ = -44.47 + 0.22 x TDS
R = 0.69
Sulphate (mg/l)
h
SO$_4$= 30.07 - 0.03 x TDS
R = -0.15
Sulphate (mg/l)
c
Ca = -2.25 + 0.24 x TDS
R = 0.96
Calcium (mg/l)
i
Calcium = -12.64 + 0.28 x TDS
R = 0.92
Calcium (mg/l)
d
Mg = 1.35 + 0.04 x TDS
R = 0.69
Magnsium (mg/l)
j
Mg = -1.16 + 0.04 x TDS
R = 0.57
Magnesium (mg/l)
e
Na = -5.47 + 0.05 x TDS
R = 0.79
Sodium (mg/l)
k
Na = 1.66 + 0.03 x TDS
R = 0.59
Sodium (mg/l)
f
Alkalinty = 1.66 + 0.01 x TDS
R = 0.72
Alkalinity (mval/l)
l
Alkalinity = -1.66 + 0.02 x TDS
R = 0.95
Alkalinity (mval/l)
TDS (mg/l)
TDS (mg/l)

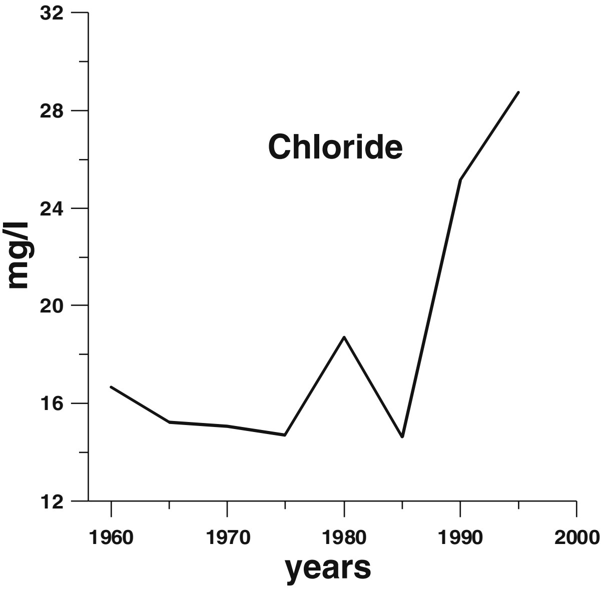
sodium concentration, as well as TH and TDS, are great indicators, sensitive to anthropogenic input. The concentrations of the remaining components are controlled mainly by "natural" (geogenic) hydrogeochemical processes. The strong relation of calcium, magnesium and alkalinity with TDS (in both data sets) reflects Ca–HCO_3 and Ca–Mg–HCO_3 water types. However, the analysis of the scatter diagrams confirms that the participation of calcium in TDS is high relative to magnesium (Ca–HCO_3 water type dominates in the WBV aquifer).

Chlorides and sulphates are the most sensitive indicators of anthropogenic input. In natural groundwater composition these components occur at low concentrations. Low concentrations of these components are also typical for comparable unpolluted confined aquifers in the Great Poland region (Gorski 1989). In this instance the concentration increase can be directly connected with anthropogenic contamination. This finding supports the suggestion of other researchers that chlorides and sulphates are the principal products of urbanization that alter groundwater chemistry (Eisen and Anderson 1979; Gorski 1989). The evidence of groundwater contamination from the land surface shows indirectly that in contaminated parts of the aquifer (mainly in the regions of relative small thickness of the aquitard) preferential flow paths exist (caused probably by the sedimentary heterogeneities of glacial tills). The semi-confined conditions allow reasonably conservative species (e.g. Cl and SO_4) to reach the aquifer. Moreover, the contamination evidence in deep aquifer leads to the conclusion that in these parts of the aquifer rather a semi-confined than confined condition occurs (in a different way to that mentioned earlier in the literature (e.g. Gorski 1989).

The concentrations of other major components are controlled by both anthropogenic and geogenic hydrogeochemical processes, thus the interpretation of their origin is more complicated. The results of factor analysis illustrate this finding (Fig. 4). The TDS, TH and sodium are included in both "anthropogenic" and "geogenic" factors.

The analysis of the variability of groundwater chemistry with time shows the intensity of anthropogenic contamination. The change of the average chloride concentration over time

Fig. 8 The variability of chloride concentration in time on the basis of the archival data (from pumping tests data, *n*=225)



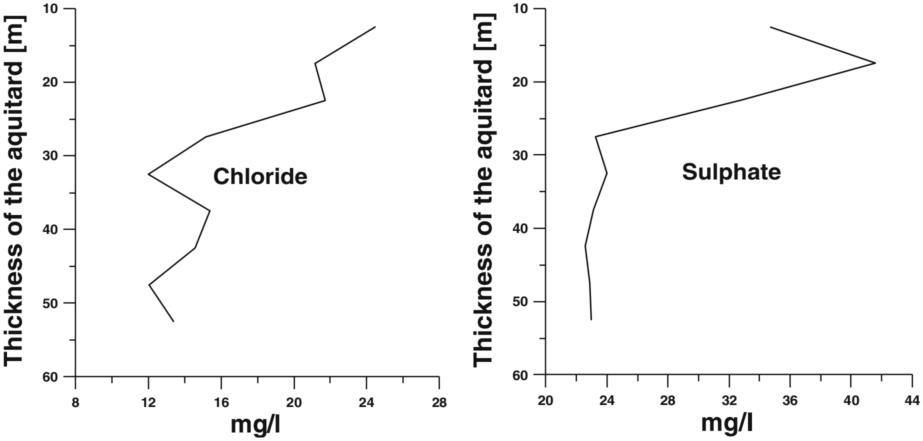


Fig. 9 The variability of the chloride and sulphate concentrations in different ranges of the aquitard thickness (on the basis of archival data from pumping tests, *n*=225)

is a clear illustration of these processes. The tendency for chloride concentration to increase is evident (Fig. 8).

This tendency for concentrations to increase over time is not obvious in the case of the remaining components, even though such trends are clear at individual wells (Dragon 2004). This suggests that the natural processes and the influence of anthropogenic contamination overlap. High concentrations of sulphate, TH and TDS in some regions are the result of natural hydrogeochemical zoning (without the influence of anthropogenic influence).

The spatial variability of groundwater chemistry caused by anthropogenic contamination is connected with the aquifer vulnerability. The concentrations of contamination indicators are higher in regions of small aquitard thickness and vice versa (Fig. 9). This finding is supported by the results of the factor analysis. The highest factor scores of the factor 1 occur in regions of small aquitard thickness (compare Fig. 3 with Fig. 5).

The changes in chloride and sulphate concentrations, caused by the anthropogenic input from the land surface, for the thickness of the aquitard smaller than 30–35 m, are visible (Fig. 9). Below this depth the concentration changes are negligible. The anomalies of chloride concentration between 35 and 45 m of aquitard thickness are visible on the graph. This may be connected with an increase of the contamination impact over time. Such a range of aquitard thickness occurs in the most urbanized areas. The long-term influence of urban areas, dating back to earlier centuries has been noted. Moreover, groundwater exploitation for industrial and domestic purposes, intensive in the most urbanized areas (which caused high drowndown) increases the migration of contaminants from the land surface.

5 Conclusions

The results of the factor analysis prove that the influence of anthropogenic contamination is one of the most important factors controlling the groundwater chemical composition of the Wielkopolska Buried Valley aquifer. The areas with the strongest impact of anthropogenic contamination were identified. These areas represent the most vulnerable parts of the WBV aquifer. Based on the factor scores, the data sets characterising "natural" and pollutant groundwater chemistry were distinguished. The comparison of "natural" groundwater with the whole data set, performed by means of correlation analysis and regression analysis, indicated the components that are the most sensitive to anthropogenic input – i.e. chloride and sulphate. The sodium concentration, TDS and TH are also sensitive to contamination.

The changes of groundwater chemistry over time indicate the intensity of contamination. The changes of the chloride concentration over time show this tendency unquestionably. The nature of groundwater chemistry changes indicates that the main contamination sources come from the unsewered urban and rural areas. Their influence, dating back to earlier centuries, was noted.

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