Agnieszka Gontaszewska, Andrzej Kraiński
Instytut Budownictwa, Uniwersytet Zielonogórski

IMPACT OF SEWAGE SLUDGE LAND APPLICATION ON GROUNDWATER'S QUALITY

WPŁYW OSADÓW ŚCIEKOWYCH UŻYWANYCH W ROLNICTWIE NA JAKOŚĆ WÓD PODZIEMNYCH

Key words: sewage sludge, groundwater quality, pollution, sludge exploitation.

Summary: Sewage sludge is nowadays treated as a fertiliser and applied to agriculture. It can result in soil and groundwater quality. The aim of the study is to describe an example of sewage sludge application and its influence on groundwater composition in West Poland. Groundwater sample from four piezometers was examined each three months and compared with hydrogeochemical background. The investigations, carried out while fertilising the land with the sludge, have revealed a considerable increase in the concentration of some ions (e.g. $N\text{-}NH_4$ by 200%, $N\text{-}NO_3$ by 1203,5%, SO_4 by 142,8% and Zn by 383,1%) as well as an inconsiderable in pH of groundwater. The authors noticed also the decrease in the concentration of heavy metals ions that may be caused by the improvement in sorption features of the ground and reduction of mobility of heavy metals ions because of precipitation. Little thickness of an unsaturated zone (8.8 m.) is not effective protection against the infiltration of the pollution from the surface. The time of the vertical infiltration into the aquifer has been estimated at t=14.4 days.

Słowa kluczowe: osady ściekowe, wody podziemne, zanieczyszczenie, wykorzystanie osadów.

Streszczenie: Osady ściekowe są obecnie stosowane w rolnictwie jako wartościowy nawóz, co może jednak powodować zmiany w jakości gleb i wód gruntowych. Celem pracy jest opis jednego z przypadków stosowania osadów ściekowych w rolnictwie i jego wpływ na skład wód podziemnych. Próbki wód podziemnych były pobierane co kwartał od początku używania osadów ściekowych. Zanotowano wzrost zawartości niektórych jonów (m.in. $N\text{-}NH_4$ o 200%, $N\text{-}NO_3$ o 1203,5%, SO_4 o 142,8% oraz Zn o 383,1%). Zanotowano także obniżenie zawartości niektórych metali ciężkich, co mogło zostać spowodowane polepszeniem się własności sorpcyjnych gleby i obniżeniem mobilności jonów metali ciężkich. Czas infiltracji do warstwy wodonośnej określono na około 14,4 doby.

INTRODUCTION

Sewage sludge is an unwanted and troublesome side-product of water treatment processes. More and more sewage sludge is produced along with human life improvement. The disposal of it is a huge environmental problem. A solution to it can be agricultural application of sewage sludge. There is no doubt, that sewage sludge is a kind of fertiliser increasing crop productions, but it can contain some harmful substances, like heavy metals or trace elements. So it may exert also negative influence on soil and groundwater quality [Brady, 1984].

The aim of this study is to describe an example of sewage sludge land application from Poland. The paper presents the analysis of the relation between the underground water quality and the composition of the sludge.

METHODS AND MATERIALS

The described sludge originates from Gubin-Guben wastewater treatment plant. This plant purifies municipal wastewater from two border cities: Gubin, Poland and Guben, Germany. The sludge of the wastewater treatment plant is used in land fertilising in the area of the town of Brzozów, West Poland (Fig. 1).

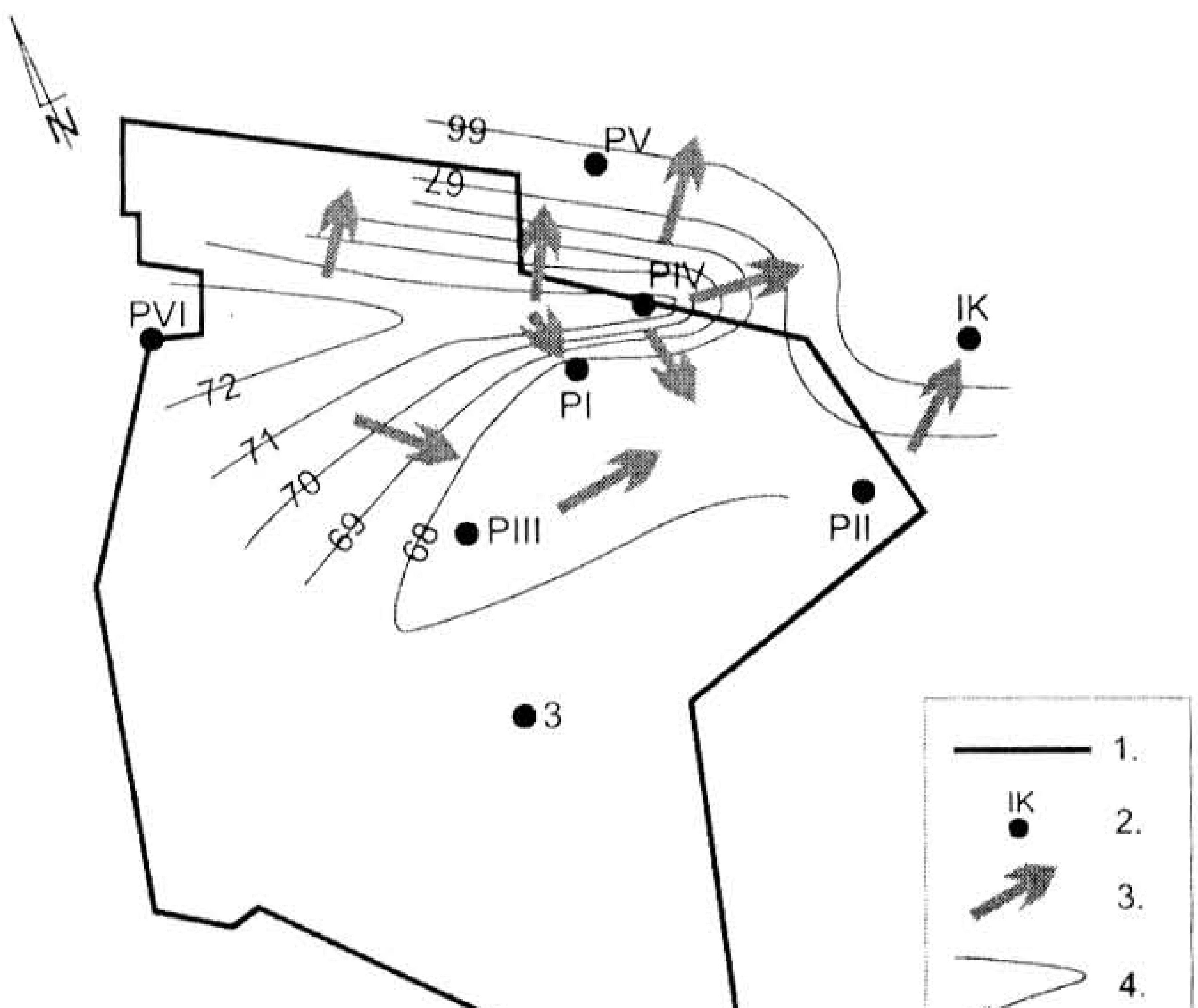

Fig. 1. Situation map of the region of sludge application; Explanation: 1- region's boundary; 2 – point of water uptake; 3 – groundwater flow; 4 – hydroisohypse

In order to evaluate the influence of the agricultural application of the sludge some geological research has been carried out as well as a local monitoring network has been established to observe its influence on the environment. The compositions of groundwater and sewage sludge are analysed each three months.

First, the sewage sludge has been sterilised with calcium carbonate in the amount of 40kg $CaCO_3/m^3$ and then applied as a fertiliser. The amount of $CaCO_3$ is sufficient to the full disinfection of the sewage sludge. The volume of the applied sewage sludge conformed the Decree of Ministry of Environment [Decree...] and amounted 26,6 t of 10%-moisture sludge per a hectare (10,0 t of dry matter) per 5 years. The sewage sludge components have been examined in a treatment plant laboratory. The tests have been carried out according to Decree of Ministry of Environment [Decree...], e.g. the content of heavy metals was examined by spectrometry of atomic absorption.

The composition of groundwater and the sludge have been analysed each three months. Before sampling, each piezometer was cleaned by double or treble pumping. Groundwater samples were delivered to laboratory in 24 hours. The results of the 2001-2002 research are presented in tab. 1.

Tab. 1. The results of the examination of the sludge of the Gubin-Guben sewage-treatment plant

indicator	unit	14.02.01	29.05.01	15.09.01	11.12.01	26.03.02	31.05.02	10.07.02	mean values	typical range
humidity	%	72,6	69,2	73,4	68,8	69,9	48,3	69,4	67,40	-
organic matter	%	48,2	47,0	47,4	36,0	43,4	75,0	49,9	49,56	50,62
reaction	pH	12,1	12,1	10,6	11,9	11,9	10,5	11,3	11,49	6,5-7,5
nitrogen	%	2,39	2,65	2,75	3,00	3,40	4,20	4,7	3,30	3,53
phosphorus	% P_2O_5	1,10	1,3	1,11	1,28	1,50	2,05	2,45	1,54	3,01
calcium	% CaO	6,3	6,1	11,1	6,3	6,20	7,9	8,4	7,47	3,31
potassium	%K_2O			0,05			1,20	1,20	0,82	0,4
magnesium	%MgO.	0,54	0,6		0,9	0,78	0,9	1,10	0,80	0,72
zinc	mgZn/kg	167	208	1170	688	414	38	187	410	1350
copper	mgCu/kg	95	97	31	250	290	197	81	149	147
lead	mgPb/kg	22	45	200	85	83	60	14	73	47,5
chromium	mgCr/kg	8,2	6,7	13	15	42	23	13	17	175
nickel	mgNi/kg	9,3	11	24	21	22	22	7	17	30
cadmium	mgCd/kg	1,0	0,8	4	8	2	1	1	2,5	3,3

The values of particular physicochemical parameters vary considerably, e.g. the content of zinc ranges from 38-117-mg Zn/kg of dry matter. pH of the sludge ranges from 10,5 to 12,1. Typical values given in the latest column are average values of the listed features for 156 Polish sewage treatment plants (years 1993-2000).

INVESTIGATED AREA

The geological structure of the region of the sludge application has been recognised in details to the depth of 15 m under the ground level. There are sand deposits in the subsoil which are glacial outwash of Weichsel glaciation in origin - GIII+1 on Fig. 2 and Fig. 3 [Cepek et al., 1994]. The thickness of the outwash series exceeds 15 m It consists mainly of medium and coarse sands and locally fine sand and sand gravel mix. In the southern part of the region there are loamy sands of the thickness up to 3m down from the surface of the area.

At the bottom of the fluvioglacial deposits occurs glacial till Saale glaciation in origin – GIVmax on Fig. 2 and Fig. 3 [Linder, 1992]. The top surface partly of the glacial drift is very irregular (Fig. 2, Fig. 3). This is probably partly the result of the glaciotectonic distortions connected with the presence of the glacier front of Brandenburger phase in the northern part of the region.

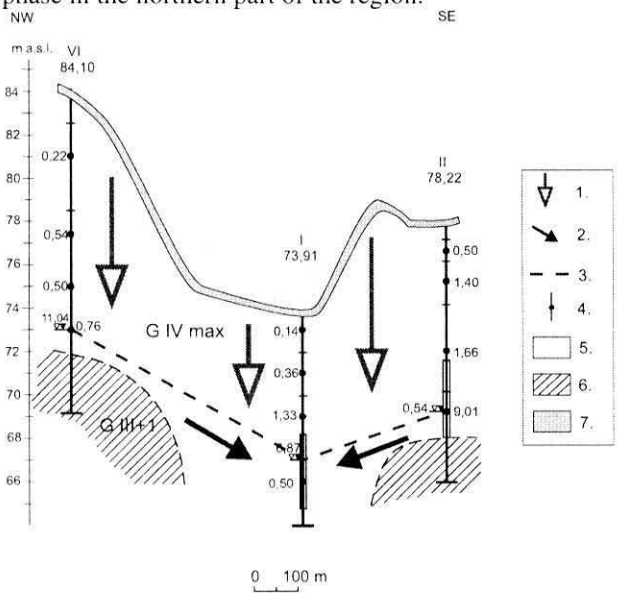

Fig.2. Geological Section I – I; Explanations: 1 – infiltration of pollution in the unsaturated zone; 2 – water flow in the water bearing layer; 3 – underground water level; 4 – filtration coefficient (m/h); 5 – sands and gravel; 6 – till; 7 – soil

A single aquifer of the Quaternary level occurs in the subsoil of the described area. The water level stabilises at the depth 0,91-12,81 m under the ground level. The falls of the water level vary (Fig. 1), which results from the elevation of the loamy subsoil in the line of the piezometers IV-VI. The thickness of the unsaturated zone south from the piezometers IV-VI increases from about 6-7m to nearly 11-13 m. (Fig. 3) with the mean value m=8,81m for the investigated area. The values of the filtration coefficient oscillate in the range from k=0,075 m/h up to k=1,98 m/h, with the mean value k=0,63 m/h (for n=24). It is also characteristic for the majority of piezometers that the values of the filtration coefficient increase with depth.

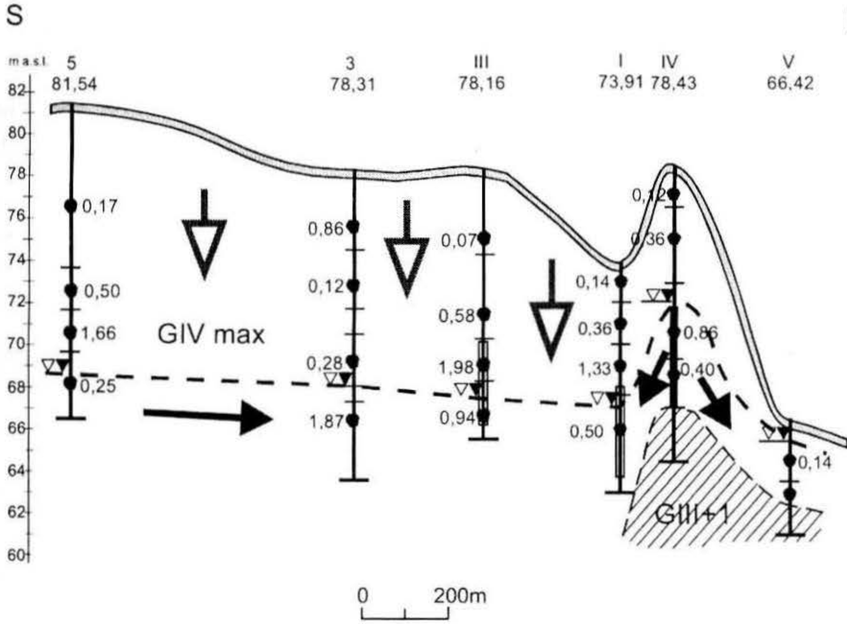

Fig. 3. Geological Section II – II. Explanations – see fig. 2

The thickness of the unsaturated zone in the recognised (northern) part of the area amounts from about 1m (holes II and VI) up to several meters (holes 3 and IV). The values of the filtration coefficient amount from k=0,14m/h to k=1,87m/h. The mean value k=0,68m/h (for n=11) is congruent with the calculated value of the filtration coefficient for the unsaturated zone. Periodical variations of the water level depend exclusively on the meteorological conditions (rainfalls) and amounted from 0,49m (hole III) to 1,88m (I). Assuming the above mean values of the parameters of the unsaturated zone, the time of vertical infiltration through the zone can be calculated (Macioszczyk, 1999) (time of the shift of pollution from the sludge to the groundwater level) which means that chemical compounds can be shifted from sludge to water bearing layer within two weeks:

$$t_a = \frac{m_a \cdot w_o}{\sqrt[3]{\omega^2 \cdot k'}}$$

where:

t_a – time of vertical infiltration through the unsaturated zone
m_a – thickness of unsaturated zone [m]

w_o – volumetric humidity [-]
ω – annual efficient infiltration [-]
k' – coefficient of vertical filtration in unsaturated zone [m/d]

RESULTS

The data derived from piezometers PI, PII, PIII and the dug well (IK) have been used for the analysis of variation in the chemical composition of groundwater resulting from sludge fertilisation.

The received mean values from the piezometers and the dug well have been compared to the values derived from piezometers PVI. The features of the groundwater derived from the piezometer PVI have been assumed to be the hydrogeochemical background (so, the groundwater composition corresponds to the one before sludge fertilisation). Piezometer PVI was realised in 2002. Thus, the credibility of the comparison is considerably weakened since there has been only one measurement for the piezometer and so the hydrogeochemical background may be not well illustrated.

Tab. 2 Statement of the mean values of indicators in the water of the local monitoring network

	P I	P II	P III	IK	*mean values*	*P VI*	*increase (%)*
pH	6,6	6,8	6,8	6,9	***6,8***	***6,7***	***1,2***
color	22,3	11,0	22,8	20,0	***19,0***	***20***	***-5,2***
oxidization	3,8	3,7	2,9	4,2	***3,7***	***6,5***	***-78,1***
basis	1,7	2,4	1,8	2,6	***2,1***	***4,8***	***-126,3***
Ca	80,7	88,3	76,5	102,0	***86,9***	***80,1***	***7,8***
Mg	11,5	17,1	11,6	14,9	***13,8***	***18,2***	***-32,0***
Fe	2,84	1,98	3,35	0,28	***2,12***	***0,61***	***71,2***
Mn	0,36	0,89	0,55	0,17	***0,49***	***0,15***	***69,4***
N-NH4	0,64	0,78	0,58	0,26	***0,57***	***0,19***	***66,4***
N-NO2	0,06	0,09	0,08	0,08	***0,08***	***0,35***	***-358,1***
N-NO3	17,06	12,21	12,19	17,45	***14,73***	***1,13***	***92,3***
PO	0,11	0,08	0,13	0,52	***0,21***	***0,13***	***38,2***
Cl	34,1	34,2	26,4	34,1	***32,2***	***38***	***-18,0***
SO_4	84,1	97,0	87,1	120,0	***97,1***	***40***	***58,8***
K	17,8	13,2	6,6	20,5	***14,5***	***9,8***	***32,6***
Na	8,8	11,6	7,1	13,8	***10,3***	***22,6***	***-118,8***
detergents	0,035	0,038	0,023	0,015	***0,028***	***0,001***	***96,4***
Zn	0,416	0,358	0,091	0,508	***0,343***	***0,071***	***79,3***
Cu	0,013	0,012	0,009	0,013	***0,012***	***0,028***	***-138,1***
Pb	0,014	0,010	0,011	0,014	***0,012***	***0,026***	***-112,7***
Cr	0,008	0,007	0,005	0,006	***0,007***	***0,008***	***-23,1***
Ni	0,013	0,013	0,009	0,011	***0,011***	***0,020***	***-76,9***
Cd	0,001	0,002	0,001	0,002	***0,002***	***0,002***	***-21,5***

The comparison of the derived values reveals that:

- pH of groundwater has increased from 6,7 to 6,8;
- oxidising abilities of water has decreased from 6,5 to 3,5 mg O_2/dm^3;
- the content of ammonia nitrogen has increased from 0,19-0,60 mg NH_4/dm^3;
- the content of sulphate has increased from 40 to 90 mg SO_4/dm^3;
- the content of detergents has increased from0,0015-0,0320 mg/dm^3;
- the content of zinc has increased from0,07-0,30 mg Zn/dm^3.

But there is a slight decrease in the number of heavy metal ions:

- the content of copper has decreased from 0,028 to 0,012 mg Cu/dm^3;
- the content of lead has decreased from 0,026 to 0,012 mg Pb/dm^3;
- the content of chromium has decreased from 0,008 to 0,007 mg Cr/dm^3;
- the content of nickel has decreased from 0,020 to 0,011 mg Ni/dm^3.

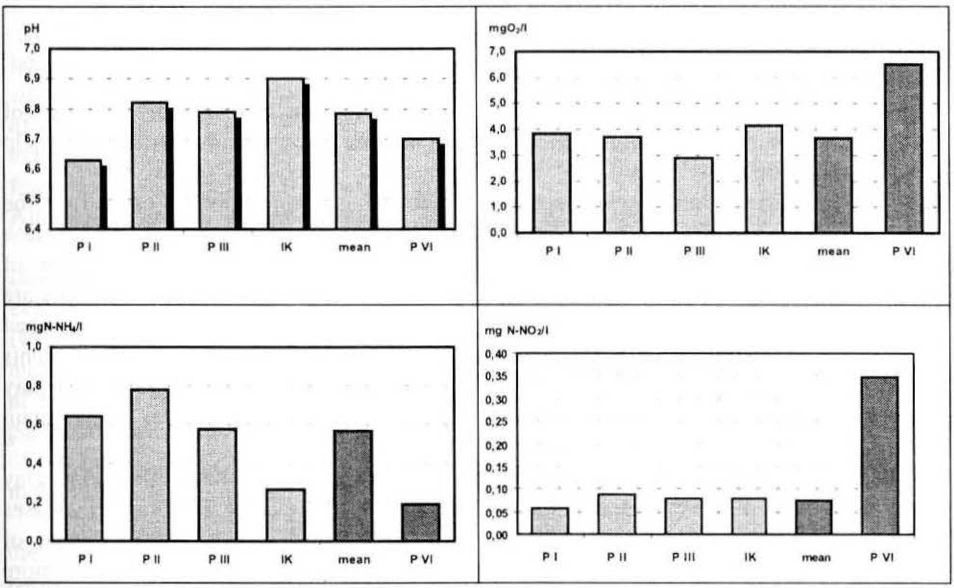

Fig. 4. Comparison of mean value of some parameters in piezometers and well with hydrogeochemical background

DISCUSSION

The data from Tab.2 demonstrate deterioration in groundwater quality within the investigated area. The authors have noticed increase in the amount of SO_4, N-NO_3, Zn, Mn, Fe, K and detergents. The applied sewage sludge is the only possible contamination source. Also high correlation coefficients (up to 0,94) calculated for some groundwater's ingredients (Tab. 3) indicate one, common source of pollution.

Tab. 3. Values of the correlation coefficient (r) between the selected ions in the local monitoring network, (det – detergents)

ions	PI	PI + PIII	PI + PII + PIII + testwell IK
$Cl-SO_4$	0,92	0,70	0,60
NH_4-NO_2	0,87	0,69	0,58
NH_4-det	0,45	0,27	0,39
NH_4-Ni	0,74	0,68	0,51
NH_4-Cu	0,94	0,84	0,71
NO_2-Cu	0,91	0,84	0,62
Cu-Ni	0,65	0,60	0,49
Cr-Ni	0,77	0,66	0,59
Zn-Cl	-	0,65	0,45

Because of the short time of vertical filtration (about 2 weeks) the elements of sludge may be quickly transferred from ground surface into groundwater with the help of infiltrating rain.

The decrease in number of heavy metal ions has been probably caused by the improvement in sorption features of the ground because of the increase in pH reaction [Kabata-Pendias, Pendias, 1992; Sukreeyapongse et al., 2002; Tan, 2000]. Sorption of soils in our climate consists mainly in cation sorption. Absorbents of heavy metals are clay mineral, zeolites, metal hydroxides and oxides and organic matter. In the described case only clay minerals and metal hydroxides and oxides may be treated as absorbents in the whole aquifer and organic matter in soil. Because of a small amount of clay minerals in the outwash deposits, the organic matter in the soil layer is the main absorbent.

Cation exchange capacity of organic matter amounts 150-400 cmol(+)/kg, clay minerals10-150 cmol(+)/kg, and sands 1-10 cmol(+)/kg. The capacity usually increases with the increase in pH [Brady, 1984; Sikora, Budek, 1996; Tan, 2000] because of releasing H^+ ions from sorptive complex. For pH≥7 exchangeable metal ions are more than 80% of the exchange complex of soil colloids.

Ni, Cd, Cu and Pb ions are quickly absorbed by clay minerals and organic matter [Bojkowska, Sokołowska, 1992; Kabata-Pendias, Pendias, 1992; Macioszczyk, Dobrzyński, 2002]. Also in the studied case the heavy metals delivered with sludge may be bound in the soil's exchange complex. The high pH of sewage sludge (average 11.5) raised the soil's pH as well as in the cation exchange capacity. However, the exchange capacity of the investigated soil (a podzoil formed from sands with low content of humus) is very small and may not be sufficient for the next sewage sludge application [Tan, 2000]. Also the sands of the outwash deposits are of a low exchange capacity because of an inconsiderable content of clay fractions [Bagińska, 1994].

The abatement of heavy metals content could be also caused by the precipitation of insoluble compounds in groundwater. The sewage sludge fertilising has resulted in an increasing amount of other components, e.g. SO_4 (by 142%) and CO_3. Some of the heavy metal ions (Ni, Pb, Cr) create compounds with these anions. Both nickel carbonate and lead carbonate and sulphate are practically insoluble compounds [Macioszczyk, Dobrzyński, 2002]. In spite of high content of $CaCO_3$ in sewage sludge (average 7,47%) there is only little increase in CO_3 in groundwater (8,5%). CO_3 might have been bound in insoluble compounds, e.g. with heavy metals. Chromium does not remain in groundwater. It precipitates as a suspension or bonds with phosphates and hydroxides [Kabata-Pendias, Pendias, 1992]. Zinc was the only heavy metal ion which content has increased, probably because of low solubility of its compounds and almost lack of sorption by clay minerals. Zinc is one of the most active ions and is also easily accessible for plants.

CONCLUSIONS

The carried out analysis reveals that the examined sewage sludge applied to the land has a negative influence on groundwater quality. A repeated increase in contents of some ions e.g. zinc, ammonia nitrogen and nitrate nitrogen ions as well as the contents of detergents has been recorded. The increased content of ammonia nitrogen exceeds even the standards for drinking water. The pollution with heavy metals has not been found to be hazardous, which may be initially attributed to the high pH of the sludge. Although, it can be just time-delayed, until the soil's sorptive capacity is depleted.

The paper presents only a part of the results of the examinations, which has been carried out for two years now. The short time of the observations does not allow a full evaluation of the influence of the land application of sludge on groundwater quality. However, a considerable influence of high pH of sludge on the content of ions of heavy metals in groundwater is observed, since the high pH results in a decrease in the number of heavy metal ions. The variations in the water quality within the mean values of parameters are considerable and may exceed 100%.

It may also be concluded that after the depletion of the sorption capacity of the soil in the unsaturated zone the composition of water will deteriorate.

The essential limitation of the presented analysis is the lack of the reliable data on the hydrogeochemical background and the values of the natural variations of the parameters within a year and a decade of years.

REFERENCES

BAGIŃSKA B., 1994: Sorptive properties of outwash deposits and its effect on groundwater chemistry on agriculture region. (in Polish) Przegląd Geologiczny, 2.

BOJKOWSKA I., SOKOŁOWSKA G., 1992: Forms of Cd, Pb and Zn occurrence in Polish contemporary alluvial deposits (in Polish). Przegląd Geologiczny, 6.

BRADY N.C., 1984: The Nature and Properties of Soils. Macmillan Pub. Com., New York.

CEPEK A., HELLWIG D., NOWEL W., 1994: Zur Gliederung des Saale-Komplexes im Niederlausitzer Braunkohlerzeier. Brandenburgische Geowissenschaftliche Beiträge 1.

Decree of Ministry of Environment about municipal sewage sludge from 27. August 2002

FYTIANOS K., CHARANTONI E., VOUNDRIAS E., 1998: Leaching of heavy metals from municipal sewage sludge. Environ. Int. 4 (24).

JEONG CH. H., 2001: Effect of land use and urbanisation on hydrochemistry and contamination of groundwater from Taejon, Korea, Journal of Hydrology 253.

KABATA-PENDIAS A., PENDIAS H., 1992: Trace elements in soils and plants. CRC Press, Boca Raton, Florida.

LINDNER L., 1992: Quaternary (in Polish). Wyd. PAE, Warsaw.

MACIOSZCZYK A., DOBRZYŃSKI D., 2002: Hydrogeochemistry of active exchange groundwater zone. Wyd. Naukowe PWN, Warsaw.

MACIOSZCZYK T., 1999: Time of water vertical infiltrating as indicator of aquifers shielding. Przegląd Geologiczny 8.

MORENO J., PEREZ A., ALIAGA A., HERNANDEZ T., 2003: The ecological dose of nickel in a semiarid soil amended with sewage sludge related to the unadmended soil. Water, Air and Soil Pollution 143.

NYAMANGARA J., MZEZEWA J., 1999: The effect of long-term sewage sludge application on Zn, Cu, Ni and Pb levels in a clay loam soil under pasture grass in Zimbabwe, Agriculture, Ecosystems and Environment 73.

REUTER G.J., SAUNDERS W.R., DALTON R.I., ALTHOFF W.F., 1983: An emergency hydrogeologic evaluation of a chemical dump site. Ground Water 21(5)

RYDIN E., 1996: Experimental studies simulating potential phosphorus release from municipal sewage sludge. Wat. Res. 7 (30).

SIKORA W., BUDEK L., 1996: Mineralogical composition and sorptive properties of some common clayey rock from Poland (in Polish). Przegląd Geologiczny 6.

SINTIC A., 2001: Entwicklung der Klärschlammentsorgung in europäischen Raum. Wasserwirtschaft, Abwasser, Abfall 8 (48).

SUKREEYAPONGSE O., HOLM P., STROBEL B., PANICHSAKPATANA S., MAGID J., HANSEN H., 2002: pH-dependent release of Cd, Cu and Pb from natural and sludge-amended soils. J. Environ. Qual. 31.

TAN K.H., 2000: Environmental soil science, Marcel Dekker Inc, New York.

WALPER G., 2002: Landwirtschaftliche Klärschlammverwertung in Hessen. Wasserwirtschaft, Abwasser, Abfall 4 (49).

WANG M-J., 1997: Land application of sewage sludge in China. The science of the total Environment 197.

WIEBUSCH B., COVERDALE S., 2002: Klärschlammverwertung und –entsorgung bei der Northumbrian Water im Nordostern Englands. Wasserwirtschaft, Abwasser, Abfall 1 (49).