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Jerzy J. MAŁECKI

A proposal of hydrochemical classification of groundwaters anthropogenetically transformed

The more intensive transformations of chemical composition of groundwaters due to an influence of anthropogenic factors bring about such case that the most of existing hydrochemical classifications, elaborated for waters of natural eomposition, could not characterize fully the polluted waters. Classifying such water kinds it should be enlarged an amount of interpreted elements with a content of nitrate and potassium ions, which in many cases occur in higher concentrations than the main components. The presented here attempt of modification of this classification has based on the principles of the classifieation of Szczukariew-Prikloński, one of the most applied systems, constructed according very clear scheme. It has based on lowering the limit of interpreted ions till 10% mval and increasing up to four the number of basic anions (HCO_3^-, Cl^-, NO_3^-) and cations (Ca^{2+}, Mg^{2+}, Na^+, K^+), determining the hydrochemical type of water. These eight ions make 225 possible combinations, theoretically enlarging to this number the amount of distinguished classes. The valuation of such modified classification was made on homogenous hydrochemical material from area of the Biebrza Depression.

The most commonly applied criterion of appreciation of the chemical composition of groundwaters is the classification based on definited hydrochemical classes. Variability of ehemical composition and multitude of aims, for which the groundwaters are exploited, have induced a generation of many classification systems. Within them were distinguished: systems of first order — mathematical, using definited limits of content of single hydrochemical elements; systems of seeond order — genetie, based on mutual relations between these elements; and classifications, joining both two systems (A. Macioszczyk, 1987). It eould be admited that how many scientists so many classification systems. They are very often modified, adopted to actually discussed scientific problem. T. Paćes (1983), distinguishing vertical hydroehemical zones, has used two criteria: changes of redox potential and of composition and proportions of gases, dissolved in water. The nearsurface zone eharaeterizes with dominance of O_2-CO_2-N_2 and Eh value about +0.4 V; the transitional zone has dominance of N_2-CO_2 and Eh is about 0.0 V but in the deepest zone Eh value is about -0.3 V and gas associations are CH_4-N_2 and CH_4-N_2-H_2S. The examples of other classifications, submited to exploitation aims of discussed waters, are systems applied in balneology and medieine.

For classification of medicinal waters were applicated the so-called pharmacodynamic eoefficients. They are estimated on basis of content of solid and gas components and water temperature. Another criterion is a comparison of osmotic pressure of analyzed water with similar pressure of some physiologieal fluids (J. Dowgiałło et al., 1969). Such classifieation types, undoubtedly significant for valuation of water properties, practically do not explain a genesis of water chemical composition.

Among a dozen or so of most commonly applied hydrochemical classifications, most of them based on six main, prevailing ions (HCO_3^-, SO_4^{2-}, Cl^-, Ca^{2+}, Mg^{2+}, Na^+). Only few ones inelude more interpreted components as, for instance, the classification of C. Palmer, discussing on valuation of salinity range also the anions of strong acids (NO_3^-, J^-, Br^-) and cations with various alkalinity, in alkalies group — K^+ and Li^+, and among heavy metals — Fe^{2+} and Mn^{2+}. Such classification, regarding mutual relations between ions, is an example of genetic one but it is less used in praxis because its usefulness for valuation genesis of chemical composition of groundwaters is illusive (Z. Pazdro, B. Kozerski, 1990).

Currently is often used the classification of Monition, which within the triangle--rhombic diagram except of basie six ions also includes concentrations of NO_3^- and K^+ but it regards them as sums of Cl^- + NO_3^- and Na^+ + K^+. It makes impossible, in valuation of anthropogenic influences, an interpretation of eoncentrations of these ions as independent hydrochemical indicators.

The more intensive transformations of chemical composition of groundwaters under influence of anthropogenic factors cause that most of existing hydrochemical classification systems, elaborated for waters of natural composition, could not truely characterize the polluted waters. Attempting classifying such waters the interpretation should be enlarged with content of such ions as nitrate and potassium ones. Their concentrations, particulary in groundwaters, in many cases resulted from anthropogenic influences. Nitrates, although they are included to minor water components (W. S. Samarina, 1977), sometimes occur with concentrations higher than main ones.

An incerase of nitrate compounds within groundwaters is a phenomenon commonly noticed by many scientists . E. Zasadowska (*fide* J. Burchard et al., 1988) in the Toruń-Wrzosy district has found concentrations of 30, 40 and up to 60 mg N-NO_3/dm^3. In regions of Łódź and Bełchatów J. Burchard, U. Hereźniak-Ciotowa (*op. cit.*) have noticed in waters exploited as drinkable ones the nitrates concentrations up to 90 and 120 mg N-NO_3/dm^3. Also from groundwaters of the Wrocław province D. Góralczyk (*op. cit.*) has described considerable content of nitrates, up to 86 mg N-NO_3/dm^3. In northern part of the Kurpie Outwash Fan B. Bagińska (1989) has defined the upper limit of the hydrochemical background at value of 25 mg N-NO_3/dm^3 also noticing there the point anomalies with values up to 93 mg N-NO_3/dm^3.

Problems connected with the environment oversaturation with nitrogen are known also from other countries. In Hungary it is noticed constant increase of nitrate amount — in average about 60 mg $N-NO_3/dm^3$ (A. Pólik, 1987). A. S. Kleczkowski (1984) indicated that in regions of middle and north-western India the nitrogen content in shallow groundwaters has achieved value of 250 mg $N-NO_3/dm^3$. On the agricultural areas such high nitrogen concentration could be connected with usage of too large doses of fertilizers in relation to the sum of mineral components, absorbed by plants.

On municipal regions the leakages from sewage systems, unproper water-sewage management, particulary on peripherial areas of cities without sewerage, and pollutions, cumulated in past, are main reasons of content increase of these compounds.

In the Białystok agglomeration within waters of first horizon mainly the nitrogen compounds had commonly content, overpassing the sanitary norms. Amount of nitrogen from nitrates was between 20 and 30 mg $N-NO_3/dm^3$ (J. Małecki, 1989).

T. Przedecki and S. Sztromajer (1975), carring on their studies on the Łódź area, came to similar opinions about forms of pollutions cumulation. They have found that recently occuring there contamination of groundwaters was resulted from cumulation during several tens of years of pollutions, coming from textile industry.

Many scientists have proved that the nitrogen compound toxically infuence on organisms. It was found that due to constant usage of water with higher content of nitrates were observed cancers of alimentary canal. Occurrence of stomach cancer among people could be correlated with increased concentrations of nitrates and nitrites in exploited well waters (J. Fiszer et al., 1976). In that light all hydrochemical classifications, attempting also to illustrate the stage of anthropogenic transformations of groundwaters and valuation of their economic usability but not regarding the nitrogen compounds, seem to be incomplete.

Nitrogen in nitrate form $N-NO_3^-$ obviously occurs in groundwaters, achieving commonly maximum concentration in shallow groundwaters. In zone of oxidizing conditions it becames the last stage of biodegradation of organic matter. Intensively migrating it undergoes in limited range to adsorption. It is also found in waters of reducing type (J. D. Hem, 1985) and is a necessible element of hydrochemical classification of waters strongly anthropogenetically transformed.

Second element, according to author needed for classification of polluted waters, is potassium ion. Similary as nitrogen it could be supplied to groundwaters due to overdosing of mineral fertilization and from industrial and vital contaminations. Within groundwaters of the Białystok agglomeration, in zones of organic matter pollutions, author has found potassium concentration up to 230 mg/dm^3. In agricultural region (central part of the Podhale Depression) anomalous values of potassium content, up to 58.2 mg/dm^3, indicated also areas of polluted waters (J. Małecki, 1987).

For including the potassium content into classification scheme of waters insists also the fact that within low mineralized waters of active exchange zone are noticed some regularities of concentration distribution of alkalic elements. Potassium makes from dozen up to several tens of percent of sodium content; all deviation from this pattern could be an additional indicator, confirming the anthropogenic influences.

The attempt of division modification, considering the concentration of nitrogen and potassium ions, was based on principles of classification of Szczukariew-Prikłońki, one of most often applied division, with very clear scheme. Proposed here changes relate to lowering the limit of percent content of ions from 20 to 10% mval content and increasing the number of interpreted ions, determining the hydrochemical type of water. In such version this classification bases on four anions (HCO_3^-, SO_4^{2-}, Cl^-, NO_3^-) and cations (Ca^{2+}, Mg^{2+}, Na^+, K^+). They offer 225 of possible combinations, theoretically enlarging to that number the amount of definited classes (Tab.1).

Valuation of such modified classification was carried on homogenous hydrochemical material from area of the Biebrza Depression. The aquifers there, of variable thickness from several up to 80 m, have irregular spatial distribution of beds. In central part of this area beds are joined forming common hydraulic system. Chemical analyses inform about composition of groundwaters from area about 1300 km^2. This region is mainly agricultural and significant part of depression belongs to the protected landscape park. The most important menances for groundwaters are towns: Ełk, Prostki, Grajewo and Szczuczyn.

Discussion of an application of this division, elaborated mainly to classify the waters transformed anthropogenetically, has based on materials from an area with relatively small scale of anthropopression. Such apparent discrepancy results from the fact that content limits of interpreted ions were such fixed that also on areas with small scale anthropogenic transformations of groundwaters the mentioned changes could be indicated in regional analyses.

Interpreting chemical composition of groundwaters from the Biebrza Depression according to principles of Szczukariew-Prikłoński method (6 main ions, 49 classes, limit at 20% mval) it was found that they had very constant composition, with dominated types: HCO_3-Ca-Mg (78% of data) and HCO_3-Ca (12% of data — Figs. 1, 2). Nextly were enlarged number of interpreted classes up to 225, including within classification system also nitrate anion and potassium cation. This modified classification was elaborated in such way that any hydrochemical class had its own field on the 3-dimensions diagram, illustrating occured frequences of distinguished classes. To get the most clear and simple such diagram and to keep some consequency in distinguishing the new classes also new classification numbers were introduced, other than in division of Szczukariew-Prikłoński, based on 6 ions (HCO_3^-, SO_4^{2-}, Cl^-, Ca^{2+}, Mg^{2+}, Na^+). In modified classification these ions form the hydrochemical classes with numbers belonging to the first area (I), which corners were determinated by class numbers: 1-7-91-97 (Tab. 1). The remaining field was divided for three areas:

— II area, which corners are defined by class numbers 8–15–98–105; they are hydrochemical classes in which except of main 6 ions is included also the NO_3^- ion ,

— III area, which corners are determined by class numbers 106–112–221–217; they are classes in which except of 6 main ions is regarded also ion K^+,

— IV area, which corners are defined by class numbers 113–120–218–225; they are classes in which for classification are also used — except of main 6 ions — NO_3^- and K^+ ions.

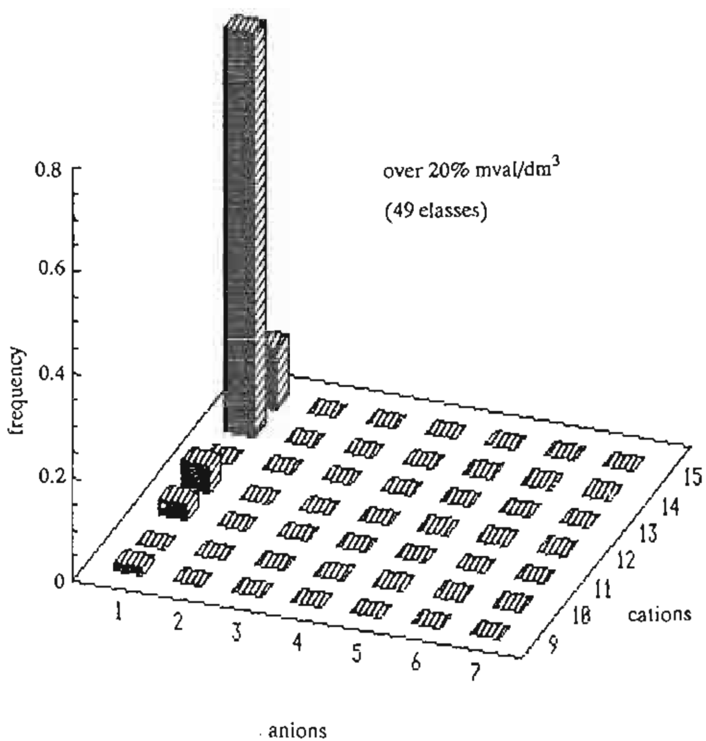
Distinguishing hydrochemical types of groundwaters of the Biebrza Depression, basing on such modified classification and with maintaining hitherto used interpreta-

Table 1

The modified classification scheme

Prevailing ions	1 HCO_3^-	2 HCO_3^- SO_4^{2-}	3 SO_4^{2-}	4 HCO_3^- Cl^-	5 HCO_3^- SO_4^{2-} Cl^-	6 Cl^- SO_4^{2-}	7 Cl^-	8 HCO_3^- NO_3^-	9 HCO_3^- SO_4^{2-} NO_3^-	10 SO_4^{2-} NO_3^-	11 HCO_3^- Cl^- NO_3^-	12 HCO_3^- SO_4^{2-} Cl^- NO_3^-	13 Cl^- SO_4^{2-} NO_3^-	14 Cl^- NO_3^-	15 NO_3^-
	I area							II area							
15 Ca^{2+}	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
14 Ca^{2+}, Mg^{2+}	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30
13 Mg^{2+}	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45
12 Ca^{2+}, Na^+	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60
11 Ca^{2+}, Mg^{2+}, Na^+	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75
10 Mg^{2+}, Na^+	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90
9 Na^+	91	92	93	94	95	96	97	98	99	100	101	102	103	104	105
	III area							IV area							
8 Ca^{2+}, K^+	106	107	108	109	110	111	112	113	114	115	116	117	118	119	120
7 Ca^{2+}, Mg^{2+}, K^+	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135
6 Mg^{2+}, K^+	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150
5 Ca^{2+}, Na^+, K^+	151	152	153	154	155	156	157	158	159	160	161	162	163	164	165
4 Ca^{2+}, Mg^{2+}, Na^+, K^+	166	167	168	169	170	171	172	173	174	175	176	177	178	179	180
3 Mg^{2+}, Na^+, K^+	181	182	183	184	185	186	187	188	189	190	191	192	193	194	195
2 Na^+, K^+	196	197	198	199	200	201	202	203	204	205	206	207	208	209	210
1 K^+	211	212	213	214	215	216	217	218	219	220	221	222	223	224	225

[— hydrochemical water types with possible anomalous concentrations of potassium


over 20% mval/dm3
(49 classes)
frequency
0.8
0.6
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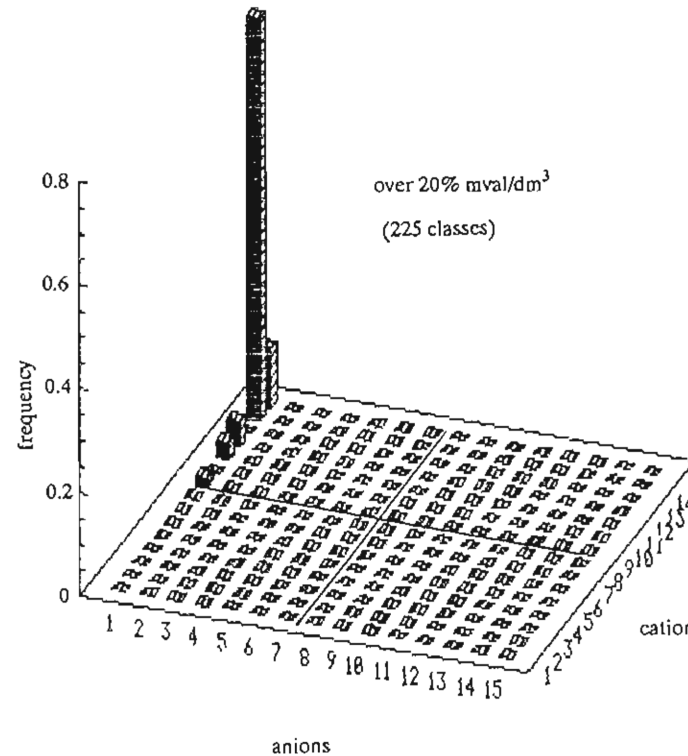
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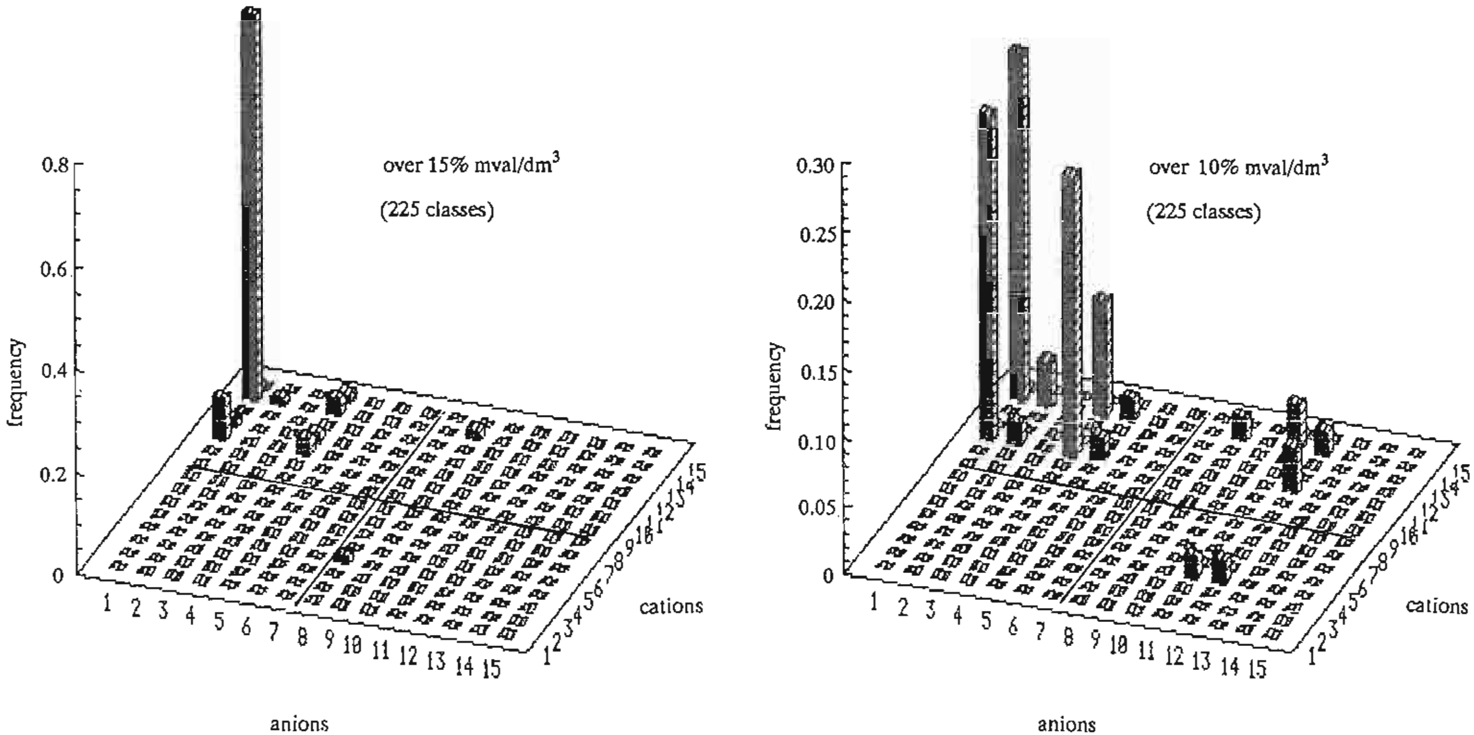

Fig. 1. 3-dimension diagrams of occurrence frequency of hydrochemical classes of groundwaters from the Biebrza Depression at fixed interpretation limit (n+60).
Diagramy przestrzenne frekwencji występowania klas hydrochemicznych w zależności od ustalonej granicy interpretacyjnej dla wód podziemnych Kotliny Biebrzańskiej (n=60)

tion limit of 20% mval, no changes have been found (Fig. 1). The amount of definited classes and frequency of their occurrences were analogous as in case of unmodified classification of Szczukariew-Prikłoński.

Some differences have resulted after lowering the limit of interpreted ions. At limit value of 15% mval (Figs. 1, 2) within the I area have occured new hydrochemical types of waters but with minute frequency from 2 up to 9%, which allowed to omit them in regional interpretations. The prevailing water type (74% of data) were HCO_3-Ca-Mg waters. But such limit changing allowed to distinguish classes of waters with anomalous high content of nitrates. In the II area has occured water of no. 24 of hydrochemical class and in the IV area — water of class no. 173 (Fig. 2). But participation of these waters in whole number of analyses was minimal (4% of data). It indicated sporadically occuring point anomalies on studied region.

Further lowering of limit of interpreted ions to value of 10% mval (Figs. 1, 2) have involved fundamental change in amount and frequency of occurrences of distinguished hydrochemical classes. In the I area have been indicated three dominant water types: no. 16, 61 and 64. For regional interpretation were regarded only these types, which frequency of occurrence overpassed 20% of whole data (Tab. 2).

Valuating the chemical composition of groundwaters from the Biebrza Depression according to discussed above principles, once more it was confirmed that main hydrochemical type are HCO_3-Ca-Mg waters (frequency is 25%) and is documented increase content of sodium and chloride ions and small — of sulphate one. In areas II and IV have occured more hydrochemical classes, containing nitrate ion. Minimal number of these classes does not allow to precise any regional conclusions and it indicates only some point anomalies. Occurrence of hydrochemical types no. 176 and 177, which include also potassium ion, has not indicated its anomalous content. According to author the finding of hydrochemical types in area with corners determinated by class no. 106-120-136-150 and 211-225 (Tab. 1) could inform about such anomalous values. In remaining parts of areas III and IV, where potassium and sodium ions are classified together, is lack of data for such opinion.

Further lowering of limit of interpreted ions to value of 5% mval/dm^3 has caused decreasing of class amount and equalizing of their percent content (Fig. 2). Constructed in that way classes represent the polyions waters that made unable to indicate the prevailing ions, particulary at regional valuations. It seems that the fixed limit of over 10% for studied hydrochemical material is the optimal one.

Interpreting the chemical composition of waters with such modified method, except of possibilities of documenting anomalous concentrations of nitrate and potassium ions, the other macrocomponents can be better characterized.

In groundwaters from the Biebrza Depression it was found, except of dominant HCO_3^-, Ca^{2+} and Mg^{2+} ions also significant content of Cl^- and Na^+ ions but only sporadically were documented hydrochemical water type with higher content of subsurface SO_4^{2-} ion (Tab. 2).

Some difficulty in hydrochemical interpretations could be large number of — 225 distinguished waters types. This number is calculated experimentally but really many of such definited types are not existed in natural conditions and they are limited only

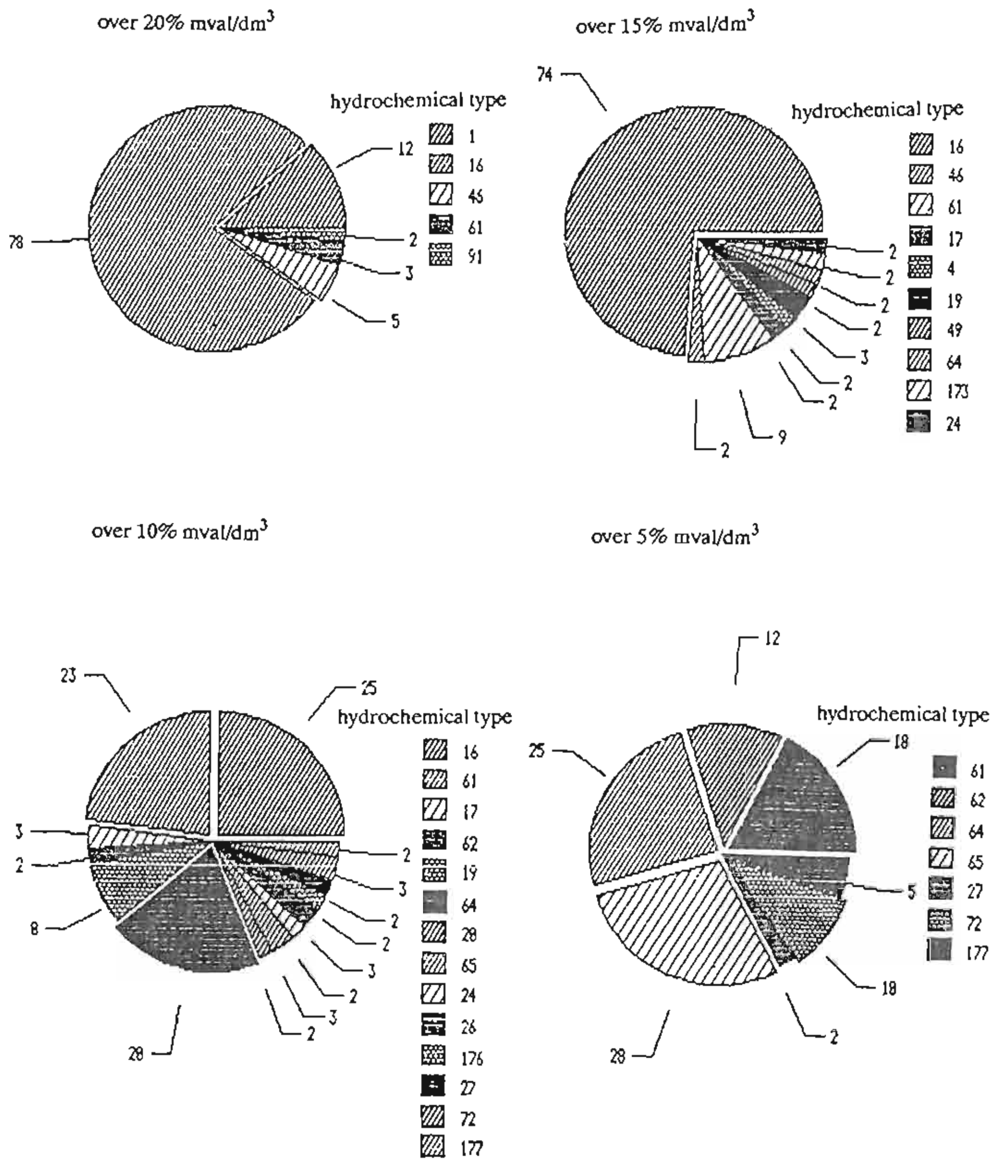

Fig. 2. Circular diagrams of percent content of hydrochemical classes of groundwaters from the Biebrza Depression at fixed interpretation limit (n=60).

Diagramy kołowe procentowego udziału klas hydrochemicznych w zależności od ustalonej granicy interpretacyjnej dla wód podziemnych Kotliny Biebrzańskiej (n=60)

Table 2

The hydrochemical types of groundwaters from the Biebrza Depression according to modified classification

Prevailing ions	1 HCO_3^-	2 HCO_3^- SO_4^{2-}	3 SO_4^{2-}	4 HCO_3^- Cl^-	5 HCO_3^- SO_4^{2-} Cl^-	6 Cl^- SO_4^{2-}	7 Cl^-	8 HCO_3^- NO_3^-	9 HCO_3^- SO_4^{2-} NO_3^-	10 SO_4^{2-} NO_3^-	11 HCO_3^- Cl^- NO_3^-	12 HCO_3^- SO_4^{2-} Cl^- NO_3^-	13 Cl^- SO_4^{2-} NO_3^-	14 Cl^- NO_3^-	15 NO_3^-
	I area							II area							
15 Ca^{2+}	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
14 $Ca2^+$, Mg^{2+}	**16**	17	18	19	20	21	22	23	24	25	26	27	28	29	30
13 Mg^{2+}	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45
12 Ca^{2+}, Na^+	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60
11 Ca^{2+}, Mg^{2+}, Na^+	**61**	62	63	**64**	65	66	67	68	69	70	71	72	73	74	75
10 Mg^{2+}, Na^+	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90
9 Na^+	91	92	93	94	95	96	97	98	99	100	101	102	103	104	105
	III area							IV area							
8 Ca^{2+}, K^+	106	107	108	109	110	111	112	113	114	115	116	117	118	119	120
7 Ca^{2+}, Mg^{2+}, K^+	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135
6 Mg^{2+}, K^+	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150
5 Ca^{2+}, Na^+, K^+	151	152	153	154	155	156	157	158	159	160	161	162	163	164	165
4 Ca^{2+}, Mg^{2+}, Na^+, K^+	166	167	168	169	170	171	172	173	174	175	176	177	178	179	180
3 Mg^{2+}, Na^+, K^+	181	182	183	184	185	186	187	188	189	190	191	192	193	194	195
2 Na^+, K^+	196	197	198	199	200	201	202	203	204	205	206	207	208	209	210
1 K^+	211	212	213	214	215	216	217	218	219	220	221	222	223	224	225

16, 61, 64 — hydrochemical classes, including over 20% of samples;
17, 19, 20, 62, 65, 24, 26, 27, 72, 176, 177 — other distinguished hydrochemical classes, 2-8% of samples

to several tens. Actually elaborated new classification systems of groundwaters, due to computer technics and easiness of data calculations, base on immense amount of classes. P. I. Stuyfzand (1991), elaborating his classifications with hierarchic pattern (main types, types, subtypes and classes) has achieved theoretically 3168 classes. Despite of so large amount of classes this classification very clearly documented changes of hydrochemical types of waters (salinity) within shore zone of the North Sea at Dutch coasts.

Author knows that proposed here classification is not all the universal one. It mainly applicates to waters transformed anthropogenetically, particulary in areas with intensive agriculture. Interpretation is in some way hindered due to omiting of other pollution indicators, both of macro- and microscale waters composition and of their general mineralization, but it was necessary to maintain the scheme clarity.

Valuation of such modified method of Szczukariew-Prikłoński, based only on results of chemical analyses of waters from one studied area, does not allow to formulate very definite conclusions. But could be stated that this method enables quick and with relatively small expense of work determining the stage of anthropogenic transformations of chemical composition of groundwaters and to illustrate in spatial form any regional differentiation of their composition. Undoubtedly the presented method needs further studies, particulary of the problem whether determining of limit of 10% mval for interpreted ions is proper or not. Such investigations should be carried on for groundwaters from various hydrochemical environments and with different stage of anthropogenic transformation.

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Katedra Ochrony Środowiska
i Zasobów Naturalnych
Wydziału Geologii
Uniwersytetu Warszawskiego
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Jerzy J. MAŁECKI

PROPOZYCJA HYDROCHEMICZNEJ KLASYFIKACJI WÓD PODZIEMNYCH PRZEOBRAŻONYCH ANTROPOGENICZNIE

S t r e s z c z e n i e

Coraz silniejsze przeobrażenia składu chemicznego wód podziemnych pod wpływem czynników antropogenicznych sprawiają, że większość istniejących klasyfikacji hydrochemicznych, konstruowanych dla wód o składzie naturalnym, nie może w pełni charakteryzować wód zanieczyszczonych. Przy klasyfikacji tego typu wód należałoby rozszerzyć ilość interpretowanych składników o zawartość jonów azotanowego i potasowego, które w wielu przypadkach występują w stężeniach przewyższających składniki główne. Próbę modyfikacji oparto na założeniach klasyfikacji Szczukariewa-Prikłońskiego, jednej z najczęściej stosowanych metod, skonstruowanej na przejrzystym schemacie. Polegała ona na obniżeniu granicy interpretowanych jonów do 10% mval oraz zwiększeniu do czterech podstawowych anionów (HCO_3^-, SO_4^{2-}, Cl^-, NO_3^-) i kationów (Ca^{2+}, Mg^{2+}, Na^+, K^+) decydujących o typie hydrochemicznym wody. Tych osiem jonów daje 225 możliwych kombinacji, teoretycznie zwiększając do tej liczby ilość typowanych klas. Ocenę funkcjonowania tak zmodyfikowanej klasyfikacji przeprowadzono na jednorodnym materiale hydrochemicznym z obszaru Kotliny Biebrzańskiej.