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Short Report

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Impact of chemical composition of groundwater/drinking water on health status of inhabitants in the Slovak Republic and proposal of limit values for the influential elements

(Action A4: „Linking of environmental and health indicators")

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Abstract

This work aims to evaluate the impact of chemical composition of groundwater/drinking on health status of inhabitants in the Slovak Republic. Primary data consist of the Slovak national database of groundwater analyses (20,339 chemical analyses, 34 chemical elements/compounds) and data on health status and demographic growth of Slovak population expressed in the form of health indicators (HI). 14 HI were evaluated including life expectancy at birth, potential years of lost life, relative and standardized mortality for the most common causes of deaths in the Slovak Republic: cardiovascular and oncological diseases, diseases of gastrointestinal and respiratory system. The chemical and health data were unified in the same form and expressed as the mean values for each of 2,883 municipalities within the Slovak Republic for further analysis. Pearson and Spearman correlation as well as method of artificial neural network (ANN) was used as mathematic method for environmental and health data analysis. Based on the results of calculations through ANN, the most significant chemical elements having influence on evaluated HI were identified together with their limit values. The following chemical elements/parameters in the groundwater were defined as the most significant: Ca+Mg (mmol l^{-1}), Ca, Mg, TDS, HCO_3 and SO_4. The most significant relationship between HI and chemical composition of groundwater was documented for Ca+Mg (mmol.l^{-1}), Ca and Mg. The following limit values were set for these most significant groundwater chemicals/parameters: Ca+Mg 2.9 – 6,1 mmol.l^{-1}, Ca 78 – 155 mg l^{-1} a Mg 28 – 54 mg l^{-1}. At these concentration ranges the health status of Slovak population is the most favourable and the life expectancy is the highest. These limit values are about twice higher in comparison with the current Slovak valid guideline values for the drinking water.

Key words

Groundwater, health status of inhabitants, Ca, Mg, artificial neural network

1. INTRODUCTION

Chemical elements can occur in the environment (water, soils, air) in relation to human health in deficit or excess contents. Each chemical element can be a medicine or a poison; it depends on the dose (Paracelsus). There are three main exposure routes of chemical elements from the environment to human organism: ingestion, inhalation and dermal contact (Selinus et al. 2005). In natural, non-contaminated environment, ingestion of food and water is considered to be the main exposure route of chemical elements to humans. While chemical elements soluble in drinking water occur mainly in the ionic form and are directly bioavailable to human organism, those found in soils can enter organism mainly vicariously through the food chain. The input of chemical elements from soils to human organism through the food chain and potential manifestation of health effects depends on many factors, mainly their bioavailability (Brümmer 1986; NRC 2003; Kabata-Pendias & Mukherjee; 2007) and speciation (McGeer et al. 2004; Chojnacka et al. 2005; Peralta-Videa et al. 2009).

In current world the foodstuffs are of global origin. Majority of them come from various regions of the world and their variability and various chemical compositions are warranted. Only a small amount of consumed food, mainly locally grown vegetable and fruit, is of local origin and its chemical composition reflects geological structure and therefore geochemical background of regions where people live. In case of drinking water the situation is different. People use for drinking and cooking water from the same source, of the same chemical composition for long term period, often during lifetime, until they move to live in other region. The variability of chemical composition of water is not such high compared to foodstuff. It is practically the same for long term period if originates from the same source. In case of deficit or excess of certain chemical elements, mainly those essential for humans in drinking water, health effects can occur due to long term water consumption. Health effects of classic contaminants found in drinking water, e.g. potentially toxic elements of nitrates are well recognized and well documented (Smith et al. 1992; Morales-Suarez-Varela et al. 1995; Järup et al. 1998; Fryzek et al. 2001; Ward et al. 2005; Mitchell et al. 2011). Due to their known adverse effects, these contaminants are strictly limited in drinking water guidelines. Influence of other, mainly essential elements (e.g. Ca, Mg, K) on human health is not currently well recognized and that is why they are not limited in such guidelines (e.g. WHO drinking water guideline, WHO 2011) or are limited only as recommended values (e.g. Slovak guideline for water used for human consumption, Anon 2010). There are many works documenting increased incidence / mortality for cardiovascular diseases associated to deficit Ca and mg contents in drinking water (Rylander et al., 1991; Yang et al., 1997a; Rubenowitz et al., 1999; Nerbrand et al., 2003; Kousa et al., 2006; Maksimović et al., 2010). Several studies can be found in world literature that link deficit Ca and Mg contents to increased mortality for oncological diseases (Yang et al., 1997b; 1999a, b, c, 2000; Rapant et al., 2015; Rapant et al., 2016).

This paper deals with evaluation of relatively wide scale of chemical elements in groundwater/drinking water in relation to health status of Slovak population. Various indicators of health status and demographic growth of population are linked to contents of chemical elements/compounds in groundwater, so called environmental indicators (EI). Therefore, we evaluate impact of groundwater originated from various geological environments, of various genesis reflected in variable chemical composition, on mortality for the most common causes of deaths with potential association to chemical elements in drinking water, namely: cardiovascular and oncological diseases, diseases of gastrointestinal tract and respiratory system and life expectancy. We use several mathematical and statistical methods (Pearson and Spearman correlations, artificial neural network) for linking data on chemical composition of groundwater with various causes of deaths. Method of neural network was used for derivation of limit values for chemical elements in groundwater to define levels at which the health status of population is most favourable. This approach was used e.g. in works Rapant et al. (2015; 2016).

2. MATERIAL

2.1 Environmental indicators

Environmental indicators (EI) represent chemical elements/compounds/parameters analysed, measured and monitored in the environment, that can effect biota or humans (Rapant et al. 2010). In our work we evaluated 34 environmental indicators, mainly anorganic components of chemical composition of groundwater including all common macroelements, trace elements and basic parameters of natural radioactivity. We do not assess organic pollutants because there are no available data collected at recquired density across the whole territory of the Slovak Republic.

The data source for groundwater chemical composition comes from national environmental-geochemical mapping, mainly the *Geochemical Atlas of the Groundwater* and environmental-geochemical maps of Slovak regions (Vrana et al., 1997; Rapant et al., 1999). These were complemented in particular by data from national groundwater monitoring, hydrogeochemical maps and other regional and local geochemical works (SHMU www.shmu.sk/en; Kordík et al. 2000). Our database thus includes virtually all sources of groundwater used for bulk supply of drinking water. The total number of chemical analyses of groundwater was 20,339. These include chemical analyses of the water since 1991, when the modern environmental-geochemical mapping of the Slovak Republic was started *under the IGCP 360 Geochemical Correlation Programme* (Darnley et al. 1995). The density of the groundwater samples was about one sample per 2.5 sq. km.

Groundwater is the most important source of drinking water for most of the population in Slovakia; coveringr approximately 90 % of inhabitants (Klinda &Lieskovská 2010). Approximately 10 % of the Slovak population uses water from individual wells for drinking and cooking purposes. About 50 % of the population is supplied with drinking water from local water companies using local water resources with a low discharge (less than 10 l s^{-1}) captured and distributed to water supply pipes in the vicinity of the villages. Only in southern Slovakia (especially in the Quaternary sediments) is the population supplied from large water resources that are distributed across distances of 50-100 km. In this work we consider groundwater and drinking water as one. However, we are aware of some inaccuracies, which may limit our results. But the size of the dataset (more than 20,000 chemical analyses, more than 30 set chemicals), reduces the uncertainties to a large extent. We were not able to assess the proportion of different bottled water that people consume.

We transformed the data on water chemical composition (EI) into a form compatible with the data on health indicators to give one value for each administrative-territorial unit of the Slovak Republic at municipal level (2,883 municipalities). Based on the input analytical data we have compiled surface distribution of each evaluated chemical element/compound in the form of so called pixel maps (1 pixel represents surface 1 sq km), using MapInfo Professional 9.0 software. An average elemental concentration for each pixel (grid cell) was computed through the common inverse distance interpolation gridding method based on averaging 10 samples that are the nearest to the pixel centre. The average value for chemicals for specific administration units (villages, districts and Slovak Republic) was then calculated as the arithmetic mean value of each pixel falling under the administration units. They are available in table as well as in map form on the website of the project Geohealth www.geology.sk/geohealth.

The set of evaluated chemicals in the groundwater (EI) with respective mean values for the Slovak Republic is reviewed in Table 1 (Rapant et al. 2014a).

Table 1 Characteristics of chemical composition of the groundwater in the Slovak Republic (mean values).

GROUNDWATER (n=20,339)												
pH	**TDS**	**COD_{Mn}**	**Ca+Mg**	**Li**	**Na**	**K**	**Ca**	**Mg**	**Sr**	**Fe**	**Mn**	**NH_4**
7.33	629.75	2.18	3.5	0.019	20.34	11.10	93.56	28.29	0.36	0.17	0.12	0.10
F	**Cl**	**SO_4**	**NO_2**	**NO_3**	**PO_4**	**HCO_3**	**SiO_2**	**Cr**	**Cu**	**Zn**	**As**	**Cd**
0.13	32.96	79.32	0.11	38.76	0.20	303.85	18.21	0.0013	0.0026	0.2673	0.0019	0.0010
Se	**Pb**	**Hg**	**Ba**	**Al**	**Sb**	**^{222}Rn**	**^{226}Ra**					
0.0010	0.0014	0.0001	0.0747	0.0297	0.0009	14.46	0.053					

Note: Data except of pH in mg l^{-1}, Ca+Mg in mmol l^{-1}, ^{222}Rn and ^{226}Ra in Bq l^{-1}

2.2 Health indicators

Health status of Slovak population is evaluated based on health indicators – indicators of demographich growth and health status of inhabitants. Health indicator (HI) is variable that expresses health status of inhabitants in society through direct measure or observation (Last 2001). In our work, we use for evaluation of impact of chemical composition of groundwater on health status of population, dataset of 14 HI reviewed in Table 2 together with description of methods of their calculations. For data evaluation we have selected basic demographic indicators, namely life expectancy at birth and potential years of lost life for all causes of deaths. Further, we evaluate 4 the most significant causes of deaths in Slovakia that can be potentially associated with the environment as influencing factor, including cardiovascular diseases (CVD), oncological diseases (OD), diseases of gastrointestinal tract and diseases of respiratory system. Health indicators are expressed in the form of relative or indirect age-standardized mortality for selected diagnoses in accordance to international classification of diseases ((ICD, 10th revision, www.who.int/classifications/icd/en/).

Table 2 Evaluated health indicators of the Slovak Republic

Health Indicator	Description of indicator	Method of calculation	Units	Mean SR*
Demographic indicators describing age structure of municipalities				
DOZ	life expectancy at birth – population	cumulative calculation of all years of life during lifetime / No. of living persons at the beginning of the year	years	72.60
Premature mortality				
PYLL100	potential years of lost life	100,000 x [the sum of the years of people up to the age of nearly 65 years (deaths at age between 1 to 64 years) / number of inhabitants]	years	4033.00
Relative mortality for selected cause of death				
ReC00-C97	malignant neoplasms	100,000 x [No. of deaths for selected cause / number of inhabitants]	No. of deaths per 100,000 inhabitants	212.79
ReI00-I99	diseases of the circulatory system			531.05
ReJ00-J99	diseases of respiratory system			58.08
ReK00-K93	diseases of the digestive system			45.83
Standardized mortality for selected cause of death				
SMRC00-C97	malignant neoplasms	indirect age-standardized mortality rate of inhabitants to the Slovak standard (19 age groups)	%	100
SMRI00-I99	diseases of the circulatory system			100
SMRJ00-J99	diseases of respiratory system			100
SMRK00-K93	diseases of the digestive system			100
Potential years of lost life for selected cause of death				
PYLLC00-C97	malignant neoplasms	100,000 x [the sum of the years of people up to the age of nearly 65 years (deaths at age between 1 to 64 years) / number of inhabitants]	years	1005.20
PYLLI00-I99	diseases of the circulatory system			866.19
PYLLJ00-J99	diseases of respiratory system			172.69
PYLLK00-K93	diseases of the digestive system			334.80

Note: Health indicators are classified according to International classification of diseases (ICD), 10th revision (http://www.who.int/classifications/icd/en/), * mean for the Slovak Republic for the period 1994 – 2003, data re-calculated according to the number of inhabitants in the Slovak municipalities

The data source was the database of the Statistical Office of the Slovak Republic (www.statistics.sk). All health indicators were calculated as a cumulative function for the years 1994 to 2003, i.e. for a ten-year period, when all cases were summed up and all numbers of inhabitants were taken as persons-per-years (number of inhabitants as of December 31 in a pertinent year) for each territorial unit assessed. Calculation methodology and standardization of health indicators was carried out according to recommendations of WHO and other authors such as Beaglehole et al. (1993); Jeníček (1995); Last (2001), Bencko et al (2003a; b). Our data thus represent average values for ten-year period for each municipality of the 2,883 Slovak municipalities. Map visualization of health indicators is presented for potential years of lost life (PYLL) in Fig. 1. Other evaluated health indicators are available online at www.geology.sk/geohealth.

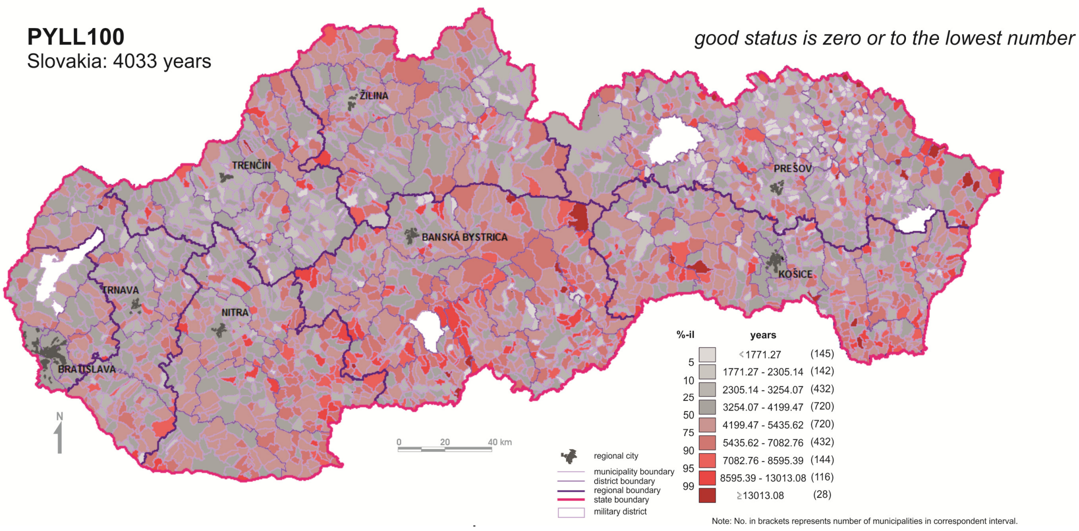

Fig. 1 Potencial years of lost life in the Slovak Republic at levels of municipalities

3. METHODS

3.1 Division of environmental and health indicators according to geological structure

The geological structure of the Slovak Republic is rather complicated. It is characterized by alteration of rocks with various geneses, ages and therefore various mineralogical and petrographic characteristics and variable geochemical backgrounds (Kohút et al. 1999). Finally it is reflected in the variable chemical composition of groundwater/drinking water that as we suppose can have variuos influences on health status of resident population.

The geological structure of the Slovak Republic has been divided in work of Rapant et al. (2014a) into eight main units, which are categorized as follows:

1. Paleogene Flysch: mainly sandstones, shales, claystones,
2. Carbonatic-silicate Mesozoic and Paleogene: mainly marl, marly limestones, dolomites, sandstones and shales,
3. Carbonatic Mesozoic and basal Paleogene: mainly limestones, dolomites, carboniferous conglomerates,
4. Neogene: mainly clays, claystones, conglomerates, sands, gravels,
5. Quaternary: mainly gravel, sand, clay, rock fragments,
6. Crystalline: mostly granites, gneisses and migmatites,
7. Paleozoic: mostly metasediments, metavolcanics,
8. Neovolcanic rocks: mainly andesites, basalts and their volcanoclastics.

The geological units were, from the point of view of health indicators, ordered (from the most favourable to the most unfavourable) as follows: Paleogene Flysch, Carbonatic-silicate Mesozoic and Carbonatic Mesozoic and basal Paleogene, Neogene, Quaternary, Crystalline, Paleozoic, Neovolcanic rocks. Generally, the most favourable rock environments for human health were defined carbonatic rocks and the most unfavourable silicate rocks.

Subsequently, we divided our data by chemical composition of groundwater – environmental indicators as well as indicators of health status – health indicators into partial datasets according to the geological units and analysed through the following statistical methods characterized below.

3.2 Statistical analysis

Statistical analysis of relationship between EI and HI was based on standard methods of data correlation, including linear (Pearson) correlation coefficient and non parametric (Spearman) order correlation coefficient. Statistical significance of calculated correlation coefficients were evaluated based on level of significance P as follows: $P < 0.05$ – verified dependence (+), $P < 0.01$ – high dependence (++), $P < 0.001$ – very high dependence (+++).

3.3 Neural network analysis

Investigation of the relationships between two different variables is the domain of statistics. However, the selection of appropriate statistical methods to link two databases requires a correct choice to measure relevant interdependency and relationships. Correlation coefficients express the intensity of stochastic dependence between two variables, demonstrating dependant relationships between measured attributes. Classical Pearson correlation coefficients express the degree of simple linear dependence of two variables. Spearman correlation coefficients are a measure of monotonic dependence. Our data were not normally distributed, but unevenly distributed, and often spoiled by errors, being incomplete, and exhibiting high variability.

Our data have all the attributes of common life. It would be incorrect to assume the existence of a functional relationship. Classical methods of regression analysis may not explore the situation fully in its complexity and may lead to wrong conclusions. Complex situations merit complex analytical approaches. Therefore, for the analysis of relationships between chemical composition of the groundwater and particular health indicators we used artificial intelligence - artificial neural networks (ANN). Detailed characteristics of ANN and principles and methods of calculations for linking EI and HI are described in work Rapant et al. (2015).

The order of effects of the chemicals in groundwater on HI was determined from the value of the sensitivity coefficient s_r. HI are influenced by those chemical elements for which the sensitivity coefficient is greater than one. In order to identify the influential chemicals from the point of view of the groundwater chemical composition, 200 networks were calculated. Selecting 200 networks has proven to be fully satisfactory, because for the next networks the value of correlation coefficient do not increase, but stagnated or declined.

Despite satisfactory performance (reliability) of the network, the impact of various environmental indicators was relatively low and was different for each created network. Therefore, the most influential chemicals were ordered based on median values of sensitivity rate ($s_{r)}$ of 50 of the best networks with the highest correlation coefficient. This approach iwas used e.g. by Opitz and Shavlik (1996); Han et al. (2011); Kourentzes et al. (2014) and Rapant et al. (2015).

Based on the median values of s_r (from the best 50 networks) calculated for each chemical element we assess the influence of the chemical element on particular health indicator. The influence increases with the increase of s_r value. Chemical elements with s_r<1 are defined as not influential on HI. Statistical significance of calculated coefficients s_r is characterized by coefficient of determination R^2. Statistical significance of s_r increases with the increase of R^2.

The results of calculations of ANN necessary for determination of shape of dependence between EI and HI were verified through the method of decils. The approach was as follows. The range of concentrations of each element evaluated in groundwater was divided into deciles. In the next step, we found the centroid of the points of which the x-coordinate belonged in individual deciles. Subsequently we found the second degree polynomial that ran a straight line through the centre of deciles 2 to 9. The conformity for the very influential elements was excellent. In the decline of the element influence the consensus exists, but the similarity decreases (Rapant et al. 2015).

The ANN method is very usefull also for derivation of limit values for chemical elements in groundwater in relation to particular HI, it can derive content levels for evaluated chemicals at which HI have more favourable values. We define two types of limit values, including limit (critical) values and optimal limit values. Limit (critical) contents represent the intersection of model curve of chemical content with average value of HI. Optimal limit contents represent, in case of curve shape (parabola) intersection with average value of HI ± standard deviation of HI. In case of straight line shape we define limit values based on intersection with 40% value of health indicator. In the case that the model curve of the chemical content does not intersect the average value of HI we were not able to determine the limit values. For many chemical elements limit value (upper or lower limit) does not exist. It means that increasing or decreasing content of chemical elements in groundwater does not have any influence on health status. The way of derivation of limit values for EI is shown in Fig. 2. We use empirical Bayesian balanced average as average value of HI instead of real values of health indicators. The advantage of this approach is that it takes into account the number of inhabitants in single municipalities (Chaikaew, et al. 2009; Chen et al. 2008; Rapant et al. 2016).

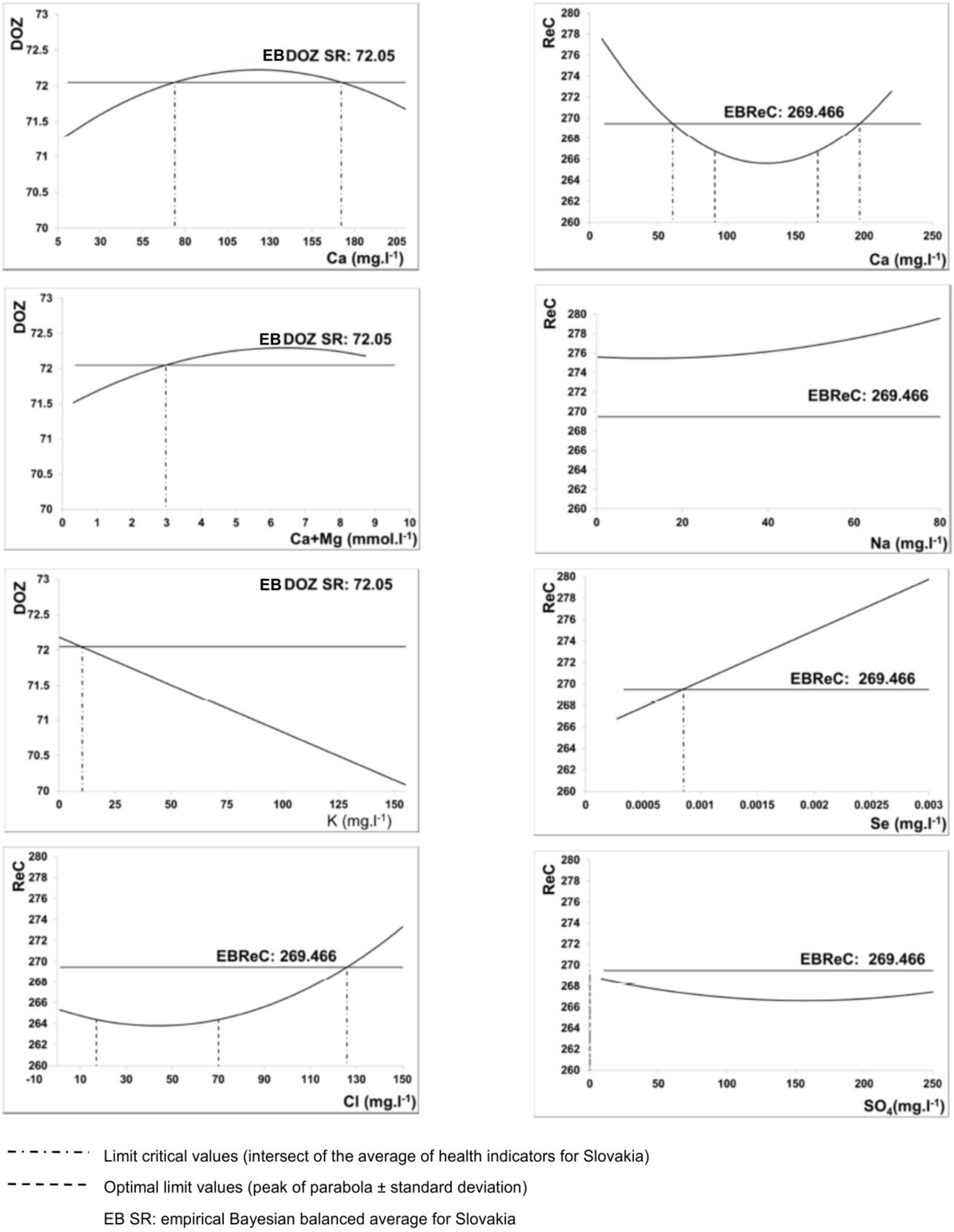

Fig. 2 Derivation of limit values for environmental indicators in relation to their influence on health indicators

4. RESULTS

Mean values of evaluated HI for two the most favourable geological units (Paleogene Flysch - 1, Carbonatic-silicate Mesozoic and Paleogene - 2) and two the most unfavourable geological units (Neovolcanic rocks - 8 and Paleozoic - 7) in relation to health indicators are rewieved in Table 3 together with the mean values for the Slovak Republic as well as two selected districts of the SR (Krupina and Bardejov). Krupina district is entirely built up by the rock environment of

Neovolcanics (in terms of HI, it is the least favourable geological environment) and the resident population is supplied only by drinking water from local groundwater sources of this district. The Bardejov district is entirely built up by Flysch Paleogene rocks (in terms of HI it is the most favourable geological environment) and the resident population is supplied from local groundwater sources of this district. The mean contents of 10 the most influential chemical elements/parameters on HI according to ANN together with two typical potentially toxic elements (PTE) – arsenic and lead are shown for the above mentioned units in Table 4.

Selected results of linear and Spearman correlations between EI and HI for geological environment as a whole are summarized in Table 5.

In Tables 6 and 7 we summarize the results of the calculations from the ANN. Table 6 provide a review of the results of sensitivity coefficients for the most influential chemical elements/parameters in the groundwater and evaluated health indicators, together with the order of influence for single elements. In Table 7 the results of calculations of ANN for life expectancy (DOZ) are shown including derived limit values for the most influential chemical elements/parameters in the groundwater. All results of ANN calculations for evaluated HI are published on the website www.geology.sk/geohealth.

Table 3 Mean values for health indicators in selected areas of the Slovak Republic

Geological unit/district	1	2	7	8	Krupina	Bardejov	SR
Health indicator	n = 727	n = 154	n = 100	n = 309	n = 36	n = 86	
DOZ	73.69	72.75	71.47	71.11	69.95	74.07	72.60
PYLL100	3874.38	3985.46	4360.96	4586.18	5609.07	3140.73	4033.00
ReC00-C97	177.99	195.96	209.46	236.28	243.23	175.32	212.79
ReI00-I99	463.32	505.07	569.73	638.78	889.20	492.82	531.05
ReJ00-J99	54.42	57.44	70.21	81.98	81.11	26.62	58.08
ReK00-K93	34.22	42.40	41.39	66.88	75.68	25.39	45.83
SMRC00-97	95.03	95.18	101.78	102.91	99.73	91.20	100
SMRI00-I99	100.03	98.86	111.73	108.50	131.06	100.71	100
SMRJ00-J99	109.39	100.61	124.81	126.34	116.33	50.50	100
SMRK00-K93	84.31	94.23	94.92	130.61	150.20	62.63	100
PYLLC00-C97	909.88	921.47	1053.42	1097.32	1121.6	808.8	1005.20
PYLLI00-I99	831.99	937.66	1052.18	1050.95	1518.2	779.9	866.19
PYLLJ00-J99	229.74	146.28	274.92	202.67	259.2	231.1	172.69
PYLLK00-K93	287.97	340.66	369.48	491.26	693.29	211.84	334.8

Note: 1 – Paleogene Flysch, 2 – Carbonatic-silicate Mesozoic and Paleogene, 7 – Paleozoic, 8 – Neovolcanic rocks, SR – mean for the Slovak Republic, n = number of municipalities in evaluated geological unit/district

Table 4 Mean values for selected chemical elements/parameters in groundwater in selected areas of the Slovak Republic

Geological unit/district	1	2	7	8	Krupina	Bardejov	SR
Parameter	n = 727	n = 154	n = 100	n = 309	n = 36	n = 86	
TDS [mg l^{-1}]	524.64	586.79	302.27	439.73	362.34	484.79	629.75
Ca+Mg [mmol l^{-1}]	3.02	3.45	1.68	2.11	1.58	2.75	3.50
Na [mg l^{-1}]	12.74	12.79	8.53	16.09	13.12	10.34	20.34
Ca [mg l^{-1}]	88.53	99.86	43.15	56.13	42.01	80.75	93.56
Mg [mg l^{-1}]	19.67	23.27	14.70	17.14	12.96	17.98	28.29
Cl [mg l^{-1}]	17.14	21.24	13.18	21.66	13.81	13.77	32.96
SO_4 [mg l^{-1}]	62.72	65.38	45.65	49.70	22.42	44.96	79.32
NO_3 [mg l^{-1}]	16.19	21.72	18.02	26.44	16.49	14.84	38.76
HCO_3 [mg l^{-1}]	287.65	323.63	138.29	191.51	174.23	282.12	303.85
As [mg l^{-1}]	0.00079	0.00135	0.00863	0.00241	0.0018	0.00114	0.00192
Se [mg l^{-1}]	0.00068	0.00074	0.00063	0.00086	0.0006	0.00068	0.00097
Pb [mg l^{-1}]	0.00125	0.00121	0.00142	0.00134	0.0018	0.00094	0.00136

Note: 1 – Paleogene Flysch, 2 – Carbonatic-silicate Mesozoic and Paleogene, 7 – Paleozoic, 8 – Neovolcanic rocks, SR – mean for the Slovak Republic, n = number of municipalities in evaluated geological unit/district

Table 5 Pearson and Spearman correlation between EI and HI for geological environment as a whole

Parameter	**Linear correlation**			**Spearman correlation**		
	r	**P**	**significance**	**R**	**P**	**significance**
Ca+Mg & DOZ	0.140	0.000	+++	0.181	0.000	+++
NO_3 & DOZ	-0.021	0.392	-	0.069	0.005	++
As & DOZ	0.020	0.411	-	-0.078	0.001	++
Ca+Mg & PYLL100	-0.130	0.000	+++	-0.187	0.000	+++
NO_3 & PYLL100	-0.001	0.960	-	-0.077	0.002	++
As & PYLL100	-0.017	0.484	-	0.083	0.001	+++
Ca+Mg & ReC00-C97	-0.085	0.000	+++	-0.134	0.000	+++
NO_3 & ReC00-C97	-0.050	0.043	+	-0.112	0.000	+++
As & ReC00-C97	-0.001	0.960	-	0.080	0.001	++
Ca+Mg & ReI00-I99	-0.083	0.001	+++	-0.151	0.000	+++
NO_3 & ReI00-I99	-0.031	0.198	-	-0.092	0.000	+++
As & ReI00-I99	-0.013	0.586	-	0.030	0.224	-
Ca+Mg & ReJ00-J99	-0.108	0.000	+++	-0.138	0.000	+++
NO_3 & ReJ00-J99	-0.057	0.020	+	-0.111	0.000	+++
As & ReJ00-J99	-0.003	0.912	-	0.090	0.000	+++
Ca+Mg & ReK00-K93	-0.049	0.047	+	-0.119	0.000	+++
NO_3 & ReK00-K93	0.075	0.002	++	-0.038	0.116	-
As & ReK00-K93	0.001	0.959	-	0.171	0.000	+++
Ca+Mg & SMRC00-C97	-0.033	0.175	-	-0.038	0.119	-
NO_3 & SMRC00-C97	0.012	0.618	-	-0.004	0.861	-
As & SMRC00-C97	0.006	0.798	-	0.086	0.000	+++
Ca+Mg & SMRI00-I99	-0.023	0.351	-	-0.046	0.061	-
NO_3 & SMRI00-I99	0.077	0.002	++	0.052	0.034	+
As & SMRI00-I99	-0.014	0.578	-	0.039	0.112	-
Ca+Mg & SMRJ00-J99	-0.066	0.007	++	-0.084	0.001	+++
NO_3 & SMRJ00-J99	-0.010	0.693	-	-0.056	0.023	+
As & SMRJ00-J99	0.004	0.871	-	0.081	0.001	+++
Ca+Mg & SMRK00-K93	-0.039	0.112	-	-0.088	0.000	+++
NO_3 & SMRK00-K93	0.105	0.000	+++	0.007	0.780	-
As & SMRK00-K93	0.018	0.456	-	0.168	0.000	+++
Ca+Mg & PYLLC00-C97	-0.079	0.001	++	-0.095	0.000	+++
NO_3 & PYLLC00-C97	-0.028	0.258	-	-0.042	0.086	-
As & PYLLC00-C97	-0.001	0.971	-	0.106	0.000	+++
Ca+Mg & PYLLI00-I99	-0.084	0.001	+++	-0.121	0.000	+++
NO_3 & PYLLI00-I99	0.042	0.083	-	0.002	0.929	-
As & PYLLI00-I99	-0.020	0.421	-	0.091	0.000	+++
Ca+Mg & PYLLJ00-J99	-0.025	0.302	-	-0.079	0.001	++
NO_3 & PYLLJ00-J99	0.009	0.715	-	0.004	0.856	-
As & PYLLJ00-J99	-0.006	0.806	-	0.058	0.018	+
Ca+Mg & PYLLK00-K93	-0.041	0.092	-	-0.079	0.001	++
NO_3 & PYLLK00-K93	0.079	0.001	++	0.006	0.800	-
As & PYLLK00-K93	0.028	0.248	-	0.156	0.000	+++

Note: r – Pearson correlation coefficient, R – Spearman correlation coefficient, P – value; level of significance = 0.05 – verified dependence (+), P = 0.01 – high dependence (++), P = 0.001 – very high dependence (+++)

Table 6 Coefficients of sensitivity and order of influence for 10 the most influential elements/parameters in groundwater in relation to health indicators according to calculations through ANN

Parameter	1		2		3		4		5		6		7		8		9		10		11		12		13		14		xP
	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	s_r	P	
Ca+Mg	1.419	1	1.115	1	1.027	3	1.370	1	1.590	1	1.057	1	1.003	3	1.677	1	1.001	6	1.180	1	1.044	1	1.046	1	1.003	4	1.169	1	1.92
Mg	1.153	3	1.027	3	1.005	8	1.150	3	1.255	3	1.009	7	1.004	1	1.291	3	1.002	4	1.065	3	1.004	4	1.002	6	1.004	3	1.063	3	3.92
Ca	1.246	2	1.048	2	1.013	4	1.211	2	1.346	2	1.015	5	1.003	2	1.387	2	1.003	3	1.108	2	1.008	3	1.006	3	1.004	2	1.100	2	2.62
TDS	1.086	4	1.003	5	1.074	1	1.053	4	1.008	4	1.015	6	1.001	8	1.018	4	1.016	1	1.051	4	1.016	2	1.002	7	1.010	1	1.028	4	3.92
HCO_3	1.012	8	1.013	4	1.034	2	1.026	5	1.005	5	1.023	4	1.002	4	1.006	5	1.005	2	1.028	5	1.002	5	1.010	2	1.003	6	1.012	5	4.38
SO_4	1.004	9	1.002	7	1.0094	5	1.009	7	1.001	8	1.006	8	1.001	10	1.001	10	1.001	5	1.006	8	1.001	10	1.003	5	1.001	7	1.003	9	7.77
Cl	1.003	11	1.002	9	1.007	6	1.027	6	1.001	9	1.029	2	1.001	5	1.001	11	1.000	13	1.021	6	1.002	6	1.002	8	1.001	8	1.003	8	7.85
NO_3	1.003	10	1.001	11	1.006	7	1.004	8	1.001	10	1.003	9	1.001	11	1.002	6	1.001	8	1.004	9	1.001	8	1.001	11	1.001	11	1.001	10	9.31
SiO_2	1.002	13	1.002	8	1.001	12	1.003	10	1.000	17	1.027	3	1.001	6	1.001	14	1.000	11	1.014	7	1.000	13	1.001	9	1.000	21	1.008	6	10.77
Na	1.0434	7	1.001	12	1.003	9	1.002	9	1.000	16	1.002	12	1.001	12	1.001	13	1.000	14	1.001	13	1.001	7	1.003	4	1.001	9	1.000	19	11.31
K	1.0732	6	1.000	15	1.000	17	1.001	12	1.000	20	1.000	17	1.001	20	1.001	15	1.000	16	1.000	17	1.000	14	1.001	10	1.000	13	1.000	28	16.00

Note: s_r – coefficient of sensitivity, P – order of influence, xP – arithmetic mean of order of influence for all evaluated health indicators, 1 – DOZ, 2 – PYLL100, 3 – ReC00-C97, 4 – ReI00-I99, 5 - ReJ00-J99, 6 - ReK00-K93, 7 - SMRC00-C97, 8 - SMRI00-I99, 9 - SMRJ00-J99, 10 - SMRK00-K93, 11 - PYLLC00-C97, 12 - PYLLI00-I99, 13 - PYLLJ00-J99, 14 - PYLLK00-K93

Table 7 Results of calculations of ANN and derived limit values for 10 the most influential chemical elements/parameters in groundwater of the Slovak Republic in relation to DOZ

Order	Parameter	s_r	R^2	Limit content		Optimal content		Evaluated function of dependence	Contents*	
				LL	UL	LL	UL		min	max
1	**Ca+Mg**	1.419	0.997	2.98	does not exist	does not exist	does not exist	concave parabola	0.35	7.97
2	**Ca**	1.246	0.975	73.95	172.21	does not exist	does not exist	concave parabola	9.83	201.01
3	**Mg**	1.152	0.975	18.13	does not exist	does not exist	does not exist	concave parabola	2.45	97.75
4	**TDS**	1.086	0.899	358.46	does not exist	does not exist	does not exist	concave parabola	87.30	1412.30
5	**COD_{Mn}**	1.081	0.994	does not exist	2.27	does not exist	does not exist	straight line	0.75	7.48
6	**K**	1.073	0.964	does not exist	9.85	does not exist	does not exist	straight line	0.27	153.15
7	**Na**	1.043	0.977	does not exist	24.07	does not exist	does not exist	concave parabola	0.71	119.69
8	**HCO_3**	1.012	0.993	250.79	does not exist	does not exist	does not exist	concave parabola	16.57	592.05
9	**SO_4**	1.003	0.522	31.42	185.32	does not exist	does not exist	concave parabola	9.38	319.50
10	**NO_3**	1.003	0.832	does not exist	71.45	does not exist	does not exist	concave parabola	1.33	227.09

Note: s_r – coefficient of sensitivity, R^2 – coefficient of determination, LL – lower limit, UL – upper limit, *minimum – maximum contents of chemical elements/parameters in groundwater of the Slovak Republic (units in mg l^{-1}, Ca+Mg in mmol l^{-1})

5. DISCUSSION

Comparison of HI values in geological units/districts

Based on comparison of HI in single geological units (Table 3, significant differences in life expectancy, potential years of lost life and also mortality for particular causes of deaths are documented. Carbonatic geological units (Paleogene Flysch - 1, Carbonatic-silicate Mesozoic and Paleogene - 2) are characterized with significantly more favourable levels of practically all health indicators compared to silicate geological units (Neovolcanic rocks - 8 and Paleozoic - 7). For example, life expectancy in Paleogene Flysch is more than 2.5 years higher (DOZ=73.69 years) than in geological unit of Neovolcanic rocks (DOZ=71.11 years). Similar situation can be observed in case of potential years of lost life (PYLL100). Its level is about 20% lower in Paleogene Flysch (3874.38 years), it means more favourable than in Neovolcanic rocks (4586.18 years). The differences in relative and standardized mortality for cardiovascular and oncological diseases and diseases of gastrointestinal and respiratory systems between these two geological units move in major cases in range 20 – 100% to the detriment of silicate geological units. Even higher differences in levels of HI are observed when comparing two districts, namely Bardejov district (the most favourable geological environment – Paleogene Flysch) and Krupina district (the most unfavourable geological environment – Neovolcanic rocks). Following are the differences to the detriment of the Krupina district: lower life expectancy more than 4 years, PYLL100 worse more than about 90%, relative mortality for diseases of gastrointestinal and respiratory systems more than 3 times higher compared to Bardejov district. Similar is the situation in case of other HI. More significant difference in the levels of health indicators in two evaluated districts compared to differences documented between single geological units can be attributed to the fact, that silicate rock environment is aquiferous to a lesser extent and the resident population in this area is often supplied by drinking water from more distant, dominantly carbonatic units (with markedly higher Ca and Mg groundwater contents), which have generally much greater water-bearing capacity. Therefore, we attribute these differencse in health indicators documented between carbonatic and silicate geological units and both districts mainly to different levels of Ca, Mg contents and water "hardness" (Ca+Mg).

The contents of these three chemical elements/parameters in groundwater/drinking water are significantly higher in carbonatic geological units than in silicate geological units (Table 4). We did not observe significant differences between the contents of other chemicals within evaluated geological units.

Statistical analysis

From the results of linear and Spearman correlations (Table 5) credible conclusions cannot be made. Our variables (EI and HI) do not have normal distribution and analysed dependences are generally not linear, often even monotonous. That is why we do not find achieved results reliable. Correlation coefficients are in both correlations very low and they range in more than 90% between levels $\pm < 0.1$. However, a very important fact is that the correlation coefficients for Ca, Mg, water hardness and evaluated health indicators (except of health indicator DOZ) show almost in all cases (also in case of other HI not reviewed in Table 5) negative values dominantly at levels with statistical very high significance. This fact indicates deterioration in values of all evaluated HI for mortality at low (deficit) Ca, Mg contents and water hardness of groundwater/drinking water in the Slovak Republic. The reverse situation is documented for health indicator DOZ (life expectancy) for which we observe positive values of correlation coefficients in case of Spearman as well as linear correlation, at levels with statistically very high significance. This clearly indicates the trend that at increased levels of Ca and Mg contents and water hardness human life extends (life expectancy is higher).

Neural networks

We find the results obtained through ANN calculations more representative because they are able to eliminate inhomogeneity of statistical datasets. Based on the obtained results we document Ca+Mg, Ca, Mg, MIN, HCO_3 a SO_4 as the most influential elements/parameters of chemical composition of groundwater for single HI (Table 6, 7). These 6 parameters are found in the group of the first 10 most influential EI in all evaluated health indicators. Other EI – Cl, NO_3, SiO_2, Na and K were ranked among 10 most influential parameters only in the case of some evaluated health indicators. Their averaged influence (xP) on HI is relatively low (7.85 – 16) and their mean levels of sensitivity coefficient are low (s_r <1.01). Three groups of chemical elements/parameters among the most influential EI on HI can be clearly identified. The first group contains Ca, Mg and Ca+Mg. We attribute to these three EI the highest influence on HI. They show the highest levels of s_r. The second group of EI (TDS and HCO_3) is considered to have only stochastic relationship (influence) to HI. It is demonstrated by the fact that the chemical composition of groundwater in the Slovak Republic is mainly of Ca-Mg-HCO_3 character. TDS and HCO_3 can be generally seen as indicators of Ca and Mg groundwater content. HCO_3 is the commonest anion in groundwater in the Slovak Republic and its concentrations are associated mainly with Ca and Mg cations (mineralization of water due to dissolution of carbonates). Similarly values of groundwater mineralization (TDS) depend mainly on the Ca and Mg contents (the most represented cations) and HCO_3 (the most represented anion) content in groundwater of the Slovak Republic (Rapant et al. 1996). The third group of the influential elements includes SO_4, Cl and NO_3. These three parameters are typical of anthropogenic groundwater contamination in the Slovak Republic. Their influence is, based on levels of sensitivity coefficients s_r, markedly lower (mainly about one order) compared to the influence of Ca, Mg and Ca+Mg. In the case of these three parameters the important fact is that their increased contents in groundwater of the Slovak Republic due to anthropogenic contamination are accompanied mainly by increased contents of Ca and Mg, which were documented in this study as the most influential parameters in relation to HI. Therefore mentioned anions do not have significant influence on health status of Slovak population. This statement does not deny any potential negative effects of nitrates, chlorides and sulphates on human health at all. All of them can have significant adverse health effects at contents locally increased in particular groundwater sources. Such highly contaminated groundwater sources are not used for drinking purposes and therefore we do not consider the influence of these three parameters in groundwater as significantly determinant for health status of population within the whole territory of the Slovak Republic. Very important fact is that all potentially toxic elements, As, Pb, Hg, Zn, Sb and other, have very low influence or are characterized as not influential for HI. Their sensitivity coefficients are in major cases lower than 1 or they are very low (s_r < 1.01). This finding is fully in accordance with our current knowledge on low impact of potentially toxic elements on health status of population in contaminated abandoned mining areas present in the Slovak Republic (Rapant et al., 2014b).

Based on the results of ANN calculations as the most influential for evaluated HI were clearly identified Ca, Mg and water hardness (Ca+Mg). Other evaluated EI are found to be less influential or with stochastic relationship to HI and therefore we will not discuss them further.

Calcium and magnesium are important intracellular cations, which are significantly involved in many enzymatic systems.. They are essential for processes of hematopoiesis and for the proper functioning of the heart and also in the prevention of oncological diseases (Bencko et al. 2011). The significance of both elements in drinking water for cardiovascular diseases has been described several times in the literature, mainly the association of the deficit Ca and Mg contents and increased incidence or mortality for CVD (Rubenowitz et al., 1998; Nerbrand et al., 2003; Kousa et al., 2006; Maksimović et al., 2010). On the other hand, there are only few works documenting association between the deficit Ca and Mg contents and mortality for OD, in Taiwan (napr. Yang et al. 1997b; 1999a, b, c, 2000), Japan (Sakamoto et al., 1997) and Slovakia (Rapant et al. 2014a; Rapant et al. 2016). We were not able to find any reference in the world literature dealing with increased incidence or mortality for diseases of gastrointestinal or respiratory system and deficit Ca and Mg contents in drinking water. Only one Russian ecological study describes significantly higher incidence of stomach and duodenal ulcer related to soft water with water hardness less than 1.5 mmol l^{-1} (Lutai, 1992 in Kožíšek, 2003). Generally, the epidemiologic studies show that the influence of Ca and Mg on the increased occurrence of oncological diseases is ambiguous. Some of the studies suggest an increased incidence of these diseases (cancer of breast, prostate, stomach and digestive tract) at raised Ca or Mg contents in human tissues and fluids, while some report exactly the opposite results (Rodriguez et al. 2003; Larsson et al. 2006; Ahn et al. 2007; Lin et al. 2007; Butler et al. 2010). In our study, we observe the highest differences just in mortality for diseases of gastrointestinal and respiratory systems. When comparing two districts, Krupina and Bardejov, we can see three times higher (worse) levels of these HI in the Krupina district than in Bardejov.

Mortality for CVD, OD and diseases of gastrointestinal and respiratory systems represent about 80 – 85% causes of deaths in Slovakia (NHIC 2015). Increased mortality for these causes of deaths in silicate geological units (volcanic rocks, granitoids, crystalline shists) and in the Krupina district (volcanic environment) is strongly reflected in demographic indicators, including life expectancy (DOZ) as well as potential years of lost life (PYLL100). The most markedly can be this difference seen in case on comparison of two discussed districts. The difference in life expectancy is almost 5 years and in potential years of lost life is more than 100% to the detriment of the Krupina district. From the mentioned above it is evident that deficit Ca and Mg contents or water hardness are significantly reflected in all main causes of deaths in Slovakia, including cardiovascular, oncological diseases and also diseases of gastrointestinal and respiratory systems. And vicerversa at their raised contents the life expectancy is higher (human life is longer).

Proposal of limit values

The most important output of our work is the definition of limit values for evaluated EI, at which we document the lowest mortality for evaluated causes of death or the human life is the longest. We review the limit values for evaluated HI together with recommended (not obligatory) values defined by the Slovak guideline for drinking water for comparison in Table 8. If we have to take into account the importance of particular HI, we will characterize life expectancy and potential years of lost life as the most significant. They reflect all other health indicators. Further, these are followed by other significant HI, such as mortality for cardiovascular diseases (about 48% of all causes of deaths) and mortality for oncological diseases ((about 25% of all causes of deaths). Mortality for diseases of gastrointestinal system (about 6% of all causes of deaths) and respiratory system (about 5% of all causes of deaths) have the lower level of significance. We cannot define levels mathematically. For about half of HI the limnit value does not exist or cannot be defined (Table 8). The absence of limit values means that increasing or decreasing content of chemical element does not have influence on health indicator. In definition of limit values we have to take into account also the potential adverse health effects of very hard water. One of the potential health effects of hard water that should be mentioned is the formation of urinary stones. However, some epidemiological studies did not confirm this relationship (Singh et al. 1993; Kohri et al. 1993). Currently there is no direct evidence that increased water hardness can cause adverse health effects (Kožíšek, 2003), except of extremely high Mg water contents (hundreds of mg l^{-1}) that cause diarrheal diseases. Among other adverse effects of hard water belong senzoric properties of water – unfavourable taste, formation of coatings on surface of coffee or tea and loss of aromatic substances from food and beverages by binding on Ca carbonate. From the technological point of view even hard water is not favourable because of crust formation; on the other hand soft water has corrosive abilities.

Optimal water hardness from the point of view of human health is hardly determinable. Most of authors recommend as the most favourable values for Mg minimum 20 – 30 mg l^{-1}, for Ca 40 – 80 mg.l^{-1} and for water hardness 2 – 4 mmol l^{-1} (Rosborg ed. 2015).

After taking into account our calculations and all other known facts we propose the following limit values for Ca, Mg and water hardness for water used for public supply as well as bottled drinking water in the Slovak Republic. For water used for drinking water public supply we propose recommended values for Ca > 50 mg l^{-1}, for Mg > 25 mg l^{-1} and for water hardness (Ca+Mg) > 2 mmol l^{-1}. For bottled drinking water we propose limit values for Ca > 60 mg l^{-1}, for Mg > 30 mg l^{-1} and water hardness (Ca+Mg) > 2.5 mmol l^{-1}. In case of Ca, we propose lower limit value compared to that derived by ANN calculations in this study, due to generally reviewed fact that the potential health effects are more likely associated to Mg in drinking water compared to Ca (Catling et al. 2005). Both elements almost always occur together in Slovak groundwater, mainly in the Ca/Mg ratio 2:1. Therefore we are not able to evaluate health effects of Ca and Mg separately. In case of all three parameters (Ca, Mg, and water hardness) we do not find reasonable and necessary to define upper limit values. Water with increased hardness above 5 mmol l^{-1}, or with Ca contents above 180 mg l^{-1} and Mg contents above 50 mg l^{-1} practically do not occur in the territory of the Slovak Republic and generally they are not used for drinking purposes. In addition, for the majority of evaluated health indicators the upper limit boundary does not exist, it means that for high contents of these parameters no adverse health effect on humans were documented.

The limit values proposed in this study are in comparison with the recommended values defined in the Slovak guideline for drinking water (Table 8, 9) about two times higher. Following this fact we recommend to increase existing limits for these parameters to reach lower level of mortality for the most common causes of deaths and higher lifetime.

Table 8 Our derived limit values for Ca, Mg contents and water hardness (Ca+Mg) for single health indicators

Health indicator	Order	Element	Limit content		Optimal content		Contents*	
			LL	UL	LL	UL	min	max
DOZ	1	**Ca+Mg**	2.98	does not exist	not defined	does not exist	0.35	7.97
	2	**Ca**	73.95	172.21	85.56	160.60	9.83	201.01
	3	**Mg**	18.13	does not exist	does not exist	does not exist	2.45	97.75
PYLL100	1	**Ca+Mg**	2.87	6.67	3.21	6.33	0.35	7.97
	2	**Ca**	79.40	169.74	87.05	162.09	9.83	201.01
	3	**Mg**	20.44	83.24	33.82	69.87	2.45	97.75
ReC00-C97	3	**Ca+Mg**	1.73	5.85	2.23	5.34	0.35	7.97
	4	**Ca**	60.56	196.84	91.18	166.21	9.83	201.01
	8	**Mg**	25.66	35.83	12.72	48.77	2.45	97.75
ReI00-I99	1	**Ca+Mg**	2.90	9.10	4.40	7.60	0.35	7.97
	2	**Ca**	does not exist	89.40	does not exist	does not exist	9.83	201.01
	3	**Mg**	24.30	95.80	42.00	78.10	2.45	97.75
ReJ00-J99	1	**Ca+Mg**	3.20	11.67	5.88	8.99	0.35	7.97
	2	**Ca**	93.08	does not exist	does not exist	does not exist	9.83	201.01
	3	**Mg**	28.63	does not exist	83.99	120.05	2.45	97.75
ReK00-K93	1	**Ca+Mg**	does not exist	4.08	0.41	3.53	0.35	7.97
	4	**Ca**	17.74	127.58	35.14	110.18	9.83	201.01
	7	**Mg**	does not exist	33.54	does not exist	10.65	2.45	97.75
SMRC00-C97	2	**Ca+Mg**	does not exist	4.17	does not exist	does not exist	0.35	7.97
	3	**Ca**	104.07	does not exist	does not exist	does not exist	9.83	201.01
	1	**Mg**	does not exist	33.50	does not exist	does not exist	2.45	97.75
SMRI00-I99	1	**Ca+Mg**	not defined	not defined	not defined	not defined	0.35	7.97
	2	**Ca**	not defined	not defined	not defined	not defined	9.83	201.01
	3	**Mg**	does not exist	65.85	does not exist	does not exist	2.45	97.75
SMRJ00-J99	6	**Ca+Mg**	3.27	does not exist	does not exist	does not exist	0.35	7.97
	3	**Ca**	90.03	does not exist	does not exist	does not exist	9.83	201.01
	4	**Mg**	25.81	does not exist	does not exist	does not exist	2.45	97.75
SMRK00-K93	1	**Ca+Mg**	0.99	2.16	0.99	2.16	0.35	7.97
	2	**Ca**	not defined	not defined	not defined	not defined	9.83	201.01
	3	**Mg**	does not exist	29.67	does not exist	does not exist	2.45	97.75
PYLLC00-C97	1	**Ca+Mg**	not defined	not defined	not defined	not defined	0.35	7.97
	3	**Ca**	93.17	194.91	106.52	181.56	9.83	201.01
	4	**Mg**	not defined	not defined	not defined	not defined	2.45	97.75
PYLLI00-I99	1	**Ca+Mg**	5.70	8.88	5.73	8.85	0.35	7.97
	3	**Ca**	150.76	does not exist	164.04	does not exist	9.83	201.01
	3	**Mg**	56.20	82.78	56.20	82.78	2.45	97.75
PYLLJ00-J99	4	**Ca+Mg**	does not exist	4.06	does not exist	does not exist	0.35	7.97
	2	**Ca**	does not exist	121.18	does not exist	does not exist	9.83	201.01
	3	**Mg**	does not exist	47.63	does not exist	does not exist	2.45	97.75
PYLLK00-K93	1	**Ca+Mg**	does not exist	4.84	0.73	3.84	0.35	7.97
	2	**Ca**	17.58	173.05	57.80	132.83	9.83	201.01
	3	**Mg**	does not exist	37.27	does not exist	does not exist	2.45	97.75
Mean values		**Ca+Mg**	**2.95**	**6.15**	**2.95**	**5.83**	**0.35**	**7.97**
		Ca	**78.03**	**155.61**	**89.61**	**152.29**	**9.83**	**201.01**
		Mg	**28.45**	**54.51**	**48.33**	**79.91**	**2.45**	**97.75**
Limit values defined by Slovak guideline for drinking water (Anon 2010)			***Ca > 30 mg l^{-1}***		***Mg 10 – 30 mg l^{-1}***		***Ca+Mg 1.1 – 5.0 mmol l^{-1}***	

Table 9 Proposed limit values for drinking and bottled water

	Recommended values	
Parameter	Drinking water for public supply	Bottled drinking water
Ca+Mg	2 – 5 mmol l^{-1}	2.5 – 5 mmol l^{-1}
Ca	50 – 180 mg l^{-1}	60 – 180 mg l^{-1}

Mg	25 – 50 mg l^{-1}	30 – 60 mg l^{-1}

Impact of other than environmental factors

In conclusion, we would like to mention some other factors that can be characterized as confounding for our results. The mortality for oncological diseases depends except of discussed aspects of chemical composition of groundwater also by a series of other factors, mainly e.g. eating habits, lifestyle, air pollution, socio-economic conditions etc. Such data are not available for particular Slovak municipalities but are available only for selected areas and districts.

Therefore, below we provide a review of available information concerning the two discussed districts of the Slovak Republic – Krupina (with the most unfavourable geological environment) and Bardejov (with the most favourable geological environment), where we document the highest differences in level of mortality for discussed causes of death as well as life expectancy (Rapant et al. 2015).

Both Krupina and Bardejov represent typical rural districts situated in mountain areas of the Slovak Republic. Average altitude of municipalities in Bardejov district is 351 m above sea level and in Krupina district 303 m above sea level. Population in both districts lives mainly in family houses. Most residents grow vegetables and fruit in their gardens for their own consumption. Regarding air quality we can state that neither in the two districts nor in their surroundings there is any significant source of air pollution (e.g. chemical industry, coal power plant etc.). Level of air pollution in both districts is low and that is why no local station for monitoring of air quality is situated there (Anon 2015). Soil contamination is a similar case as an index of environmental risk from soil contamination shows very low levels ($I_{ER}<1$) in both districts. Moreover, we also document very low levels of environmental risk of soil contamination in both evaluated districts (Rapant et al. 2004; Rapant et al. 2008).

A higher rate of the Gypsy population characterized by significantly worse socio-economic level, health status and also life expectancy in comparison with other Slovak population is probably a very important confounding factor. In Bardejov district a total number of inhabitants of Gypsy nationality is approximately two times higher than that in Krupina district (Table 10).

Other important socio-economic factors that could have some impact on HI include registered level of unemployment rate as well as average salary of local residents. Both factors show more unfavourable levels in Bardejov district compared the the Krupina district.

From among other health determinants influencing human health positively or negatively, available data on lifestyle and health-care are reviewed in Table 10. Based on the comparison of the reviewed data we can conclude that there are no significant differences between the listed health determinants in both discussed districts. On the other hand, slightly but not significantly better values for these factors can be observed in Krupina district, where significantly worse health status and shorter life expectancy were reported.

Table 10 List of selected socio-economic, health-care and lifestyle characteristics for Krupina and Bardejov districts compared with the Slovak Republic

Socio-economic characteristics[a]	Krupina	Bardejov	SR
Level of registered unemployment (% of population)	16.95	19.6	*12.29*
Average nominal monthly salary in Euro	694	614	*957*
Rate of gypsy nationality (% of population)	2.1 - 4	4.1 - 8	2
Health-care characteristics[b]			
No. of physicians posts per 10,000 population - adults (age 18+ years)	4.36	3.40	*4.32*
No. of physicians posts per 10,000 population - children and adolescents (age 0-17 years)	6.86	7.44	*9.87*
Lifestyle characteristics[c, d]			
Regular physical activity in average (% of population)	45	39.5	*58.5*
Regular eating habits (% of population)	75	49	*68*
Smoking (% of population)	25	43	*19.5*
Excessive alcohol intake (% of population)	9.8	11	*6.8*

Note: [a]Statistical office of the Slovak Republic (www.statistics.sk), [b]NHIC 2013, [c]Data source for Krupina district: Kosmovský et al., 2015, [d]Data source for Bardejov district and the Slovak Republic: EHES – European Health Examination Survey (www.ehes.info), SR – Slovak Republic

6. CONCLUSION

Based on the achieved results we can conclude that health status together with life expectancy of population in the Slovak Republic is significantly influenced by chemical composition of groundwater/drinking water, mainly by the Ca, Mg contents and water hardness (Ca+Mg). Mortality for the main causes of deaths including cardiovascular and oncological diseases and also diseases of gastrointestinal and respiratory system is markedly lower at concentration ranges of discussed parameters in groundwater as follows: Ca 78 – 155 mg l^{-1}, Mg 28 – 54 mg l^{-1} and water hardness (Ca+Mg) 2.9 – 6.1 mmol.l^{-1}. Worse health status and lower life expectancy are observed at low, deficit contents of these parameters in groundwater/drinking water. Our derived limit values are about 2 times higher compared to limits defined within the Slovak guideline for drinking water. We propose to increase them in case of drinking water used from public supply at following concentrations: Ca>50 mg.l l^{-1}, Mg >25 mg.l l^{-1} and Ca+Mg >2 mmol l^{-1}. For drinking bottled water we recommend following limit values: Ca>60 mg l^{-1}, Mg>30 mg.l l^{-1} and Ca+Mg pre >2.5 mmol l^{-1}. Based on achieved results we recommend to World Health Organization to make revision for definition of drinking water quality standards for Ca and Mg contents or water hardness (Ca+Mg).

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