SPATIAL ANALYSIS OF ENVIRONMENTAL AND ANTHROPOGENIC INFLUENCES ON NITRATE CONCENTRATION IN SHALLOW GROUNDWATERS. CASE STUDY: RUSCOVA VALLEY, MARAMUREŞ MOUNTAINS (ROMANIA)

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Key-words: nitrate, shallow groundwater pollution, spatial analysis, Ruscova Valley, Maramureş Mountains.

Abstract. In many rural areas of Romania groundwaters stand out as the main sources for water supply. As a result, the assessment of the shallow aquifer of Ruscova Valley (Maramureş Mountains) is an important research direction since the nitrates are considered some of the major causes of deteriorating water quality, with influences on the human health. The outcomes of this paper relied on data collected from field surveys (water samples) combined with Geographical Information Systems (GIS) and statistical assessment of the existing and derived spatial data. In addition to nitrate concentration, determined in 26 water samples, several parameters were measured during the field surveys: water *pH*, temperature, depth to water and altitude of the sample point. Moreover, several environmental and anthropogenic potential explanatory factors for nitrate concentration were analysed: depth to water, land use/land cover, lithology, soil texture, number of households and population density. In order to reveal the relationship between the nitrate concentration and this explanatory factors the authors used a multiple linear regression (MLR). Spatial and statistical analysis were realised for three buffering zones (100 m, 200 m, 300 m) created around each water sample in order to identify and quantify the role of the explanatory factors at various scales. Finally, through regression analysis it was possible to determine which datasets explain better the nitrate concentration in the shallow groundwater.

1. INTRODUCTION

In comparison to surface water, groundwater has been considered as an important source of water supply due to its relatively low susceptibility to pollution and its large storage capacity (U.S. Environmental Protection Agency (USEPA), 1996). Diffuse nitrate pollution of groundwater is currently considered one of the major causes of deteriorating water quality (Knapp, 2005), thus becoming a widespread environmental issue affecting all countries regardless of their level of economic development (Tagma *et al.*, 2009).

Nitrate groundwater pollution is currently increasing and often recognized to be a threat to health of living beings which depend to this type of water resources. Because of their solubility and low binding capacity, nitrates' capacity to migrate to shallow aquifers and groundwater favours their persistence in polluted water until consumed by plants or other organisms. Excessive amounts of nitrate can cause a blood disorder-methaemoglobinaemia, commonly known as infant cyanosis or "blue-baby syndrome" which leads to a reduction of the oxygen caring-capacity of bloodstream. The most exposed to this disease are infants under 1 year of age. In epidemiological studies, derivation methaemoglobinaemia was not reported in infants in the areas where drinking-water consistently contained less than 50 mg/L (World Health Organization (WHO), 2004). Moreover, the coupling of nitrates with amines, through the action of bacteria in the digestive tract, may result in the formation of nitrosamines, potential related to cancer risk (Bell, 1998).

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The natural nitrate concentration in groundwater under aerobic conditions is a few milligrams per litre and depends strongly on soil type and on geological settings. Nitrate can reach both surface water and groundwater as a consequence of agricultural activity (including excess application of inorganic nitrogenous fertilizers and manures) from wastewater treatment and from oxidation of nitrogenous waste products in human and animal excreta, including septic tanks (WHO, 2011).

According to the WHO (2011) guidelines for nitrate, the maximum acceptable concentration of nitrate for potable water is 50 mg/L for a short-term exposure, and represents the reference values in the development of national standards. In few states, i.e. Italy, a limit of 10 mg/L NO_3^- has been recommended for drinking water destined to infants. Also, the USEPA (1995) has established a drinking-water standard of 10 mg/L nitrate-nitrogen (equivalent to 10 ppm nitrate-nitrogen or 45 ppm nitrate) based on the human health risks due to nitrate consumption. The Nitrates Directive 91/676/EEC (European Commission, 1991), developed in response to the European Union's concern about the environmental and health implications of this phenomenon and the EU Water Framework Directive 2000/60/EC (European Commission, 2000), stressed the need to reduce water pollution caused by nitrates from agricultural sources to 50 mg/L nitrate concentrations in water bodies. The same threshold was established by the Romanian Government Decision no. 964/2000 (Romanian Government, 2000) for the approval of the Action Plan for the water protection against pollution with nitrates from agriculture sources and by the Law 458/2002, modified by Law 311/2004 (Romanian Government, 2004) on the quality of drinking water.

2. STUDY-AREA

The study-area is located in the Maramureş Mountains, including the lower part of Ruscova Valley and its main tributaries (e.g. Repedea, Cvasniţa) (Fig. 1). Furthermore, the area is part of Maramureş Mountains Natural Park (Category V IUCN – Protected Landscape-Natural Park), overlapping, in terms of Park zoning, the sustainable development and sustainable management areas.

The study-area covers 1,230 ha with altitudes decreasing from 620 m to 410 m. Generally, the relief is represented by floodplain and fragmented terraces (formed in the Cvasniţa – Ruscova and Ruscova – Vişeu confluence areas). In terms of geological features, the area corresponding to Poienile de sub Munte and Ruscova Depressions was formed within the Paleogene Gulf (Posea *et al.*, 1980). Largely, the rock types belong to sedimentary formations (including sands, gravels, sandstones, conglomerates and marls), covered by fluvisols, dystric and eutric cambisols. After the hydrogeological map of the Geological Institute of Romania, 1969, 1:1,000,000 scale, the considered shallow aquifer lies in the aquifers systems hosted within the sandstones and marls (Eastern Carpathian Flysch). According to the Climatic Regionalization (Bogdan, 2002), the area displays a moderate continental climate with Atlantic humid influences. The multi-annual air temperature means vary between 5°…6°C in the eastern part and 7°…8°C in the extreme west of the area. As in the case of temperature, the quantity of precipitation is unevenly distributed within the area. The multi-annual mean is estimated between 1,000 – 1,100 mm in the eastern part and around 900 mm in the Ruscova – Vişeu confluence area.

There are three administrative-territorial units (Poienile de sub Munte, Repedea, Ruscova) with about 21,000 inhabitants (2014) (http://statistici.insse.ro/shop/). The main land-use category is represented by agricultural land, including arable lands and orchards, frequently developed near the built-up areas. The households are spread close to the main roads, and in the central parts of localities. Agriculture and animal breeding are the most significant activities carried out in this area.

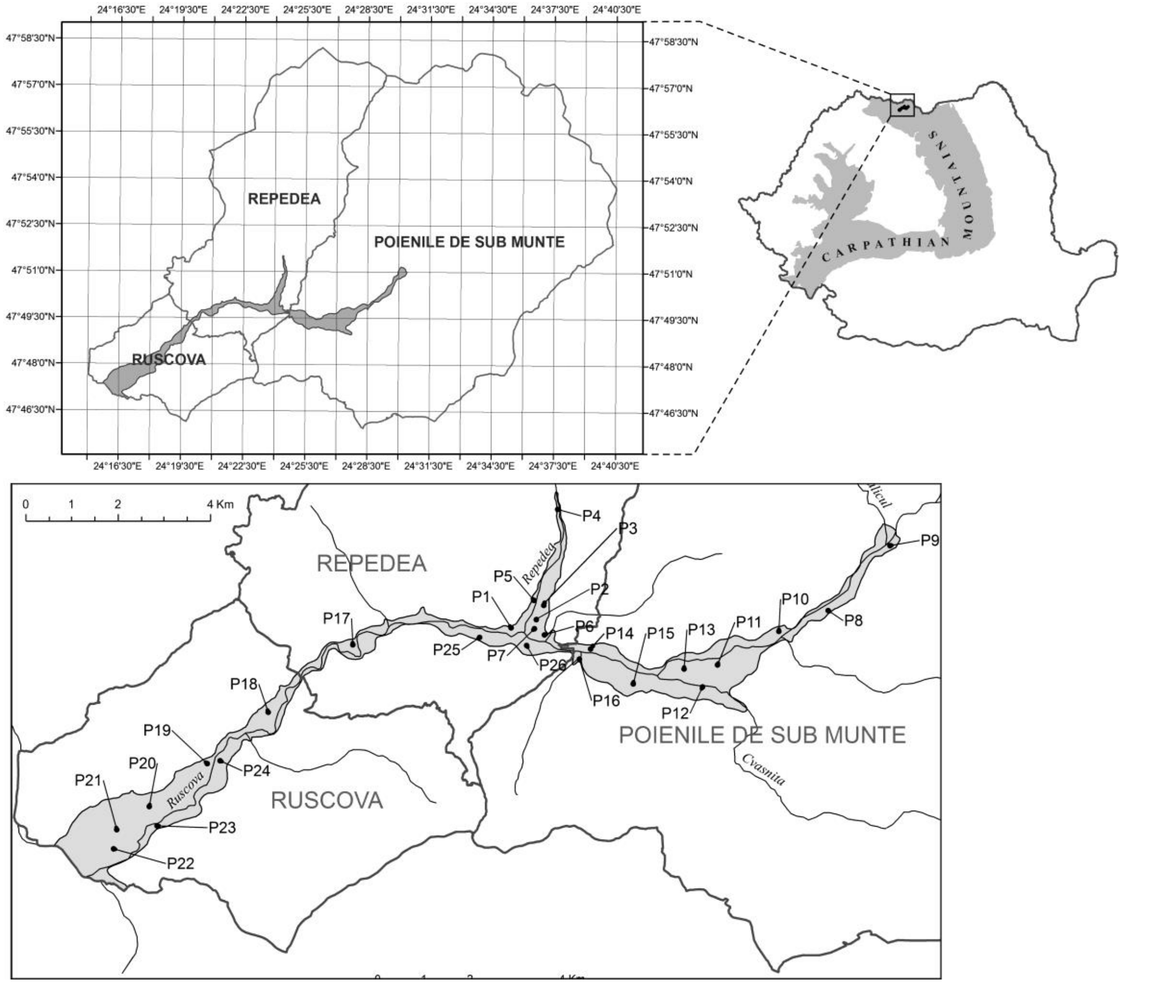

Fig. 1 – Location of the study-area.

In the study-area, groundwaters still represents the main drinking water sources. Although there is connection to water supply system, local communities often use the individual wells linked to shallow groundwater.

3. METHODS

3.1. Field sampling

In the spring of 2014, in Poienile de sub Munte, Repedea and Ruscova localities the nitrates content was determined *in situ* in 26 water samples from domestic wells. Additionally, several parameters were measured in the field: depth to water, water *pH*, water temperature and sample altitude (Table 1). Nitrate concentration was determined using nitrate-test strips *(MQuant^{TM} Nitrate Test, Cat. No. 1.10020.0001, E. Merck, Darmstadt, Germany)*, a quick analysis instrument based on the visual comparison of the reaction zone of the test strip with the colour scale indicated on the box. Water *pH* and temperature were determined using the *WTW Multi 340i* handheld measuring instrument. The position of each water sample site was recorded using a GPS system.

Table 1

The measured indicators for water samples

Sample code	Location	NO_3^- (mg/L)	Depth to water (m)	Temperature (°C)	pH	Altitude (m)
P1	Repedea	40	3.5	12	7.5	500
P2		70	4.0	8	7.9	509
P3		10	6.0	11	7.5	512
P4		15	12.0	12	7.7	547
P5		30	7.0	8	7.8	511
P6		20	3.0	15	7.7	505
P7		40	7.0	16	7.2	504
P17		50	4.0	9	7.8	476
P25		<10	8.0	10	7.9	496
P26		<10	5.0	10	7.8	506
P8	Poienile de sub Munte	10	6.0	9	7.8	579
P9		25	7.0	9	7.7	604
P10		30	8.0	12	7.1	570
P11		15	12.0	12	7.5	549
P12		<10	15.0	10	7.6	545
P13		<10	16.0	13	7.8	539
P14		15	8.0	9	7.7	514
P15		<10	4.0	9	7.4	529
P16		<10	12.0	8	8.1	522
P18	Ruscova	20	7.0	10	7.9	457
P19		40	10.0	9	7.4	441
P20		50	12.0	10	8.1	434
P21		25	18.0	11	7.8	426
P22		70	18.0	11	7.3	424
P23		<10	18.0	11	7.8	422
P24		40	5.0	10	7.8	443

3.2. Spatial and statistical analysis

Environmental and anthropogenic explanatory factors

Given the geographical conditions, human activities and land use, the current findings revealed that in the study-area the shallow groundwater is polluted especially as a result of farming activities and human waste.

In order to assess the nitrate concentration in the shallow groundwater and its relation with the explanatory factors several spatial data, including both environmental and anthropogenic factors, were used: depth to water, slope declivity, land use/land cover, lithology, soil texture, number of households and population density (Table 2). The data was considered according to 100 m, 200 m, and 300 m buffers surrounding each well. The resulted data were analysed using statistical regression analysis. Different multiple linear regression models were created for each buffer depending on size. The results were used to determine how each explanatory factor can influence the nitrate concentration in the shallow groundwater at different scale.

Depth to water (DW) was determined by measurements of local water level in the shallow wells/hand pumps. *Land use/land cover* data derived from the orthophoto images, scale 1:5,000, classified into four classes according to their potential influence on nitrate concentration: areas mostly occupied by *built-up areas* (BA), areas mostly occupied by *arable lands* (AL), areas mostly occupied by *grasslands* (GRASS) and *others* (OTH), including forests, shrubs, water courses, alluvial deposits and bare soils. *The slope declivity* (SD) was determined by TIN (Triangulated Irregular Network) surface model created by counter lines extracted from the topographic map, 1:25,000 scale. The *population density* (PD) was calculated based on TEMPO Online database, at built-up areas level. The *number of households* (HN) was extracted and calculated based on orthophoto images, scale 1:5,000. This indicator is important since includes isolated houses or stables outside the built-up areas (within arable lands and/or grasslands). The data referring to *lithology* (LITHO) and *soil texture* (TEXT) was extracted from the geological and soil maps, respectively (scale 1:200,000). In order to carry out the spatial and statistical analysis, these maps were coded according to their classes (Table 3).

Table 2

Explanatory factors used to assess the nitrate concentration in the shallow groundwater

Data layer	**Source / comments**
Depth to water, in meters (DW)	terrain records; measurements of water level in shallow wells/hand pumps
Slope declivity, in degrees (SD)	TIN (Triangulated Irregular Network) – surface model; derived from the topographic map, scale 1:25,000
Land use/land cover	derived from the orthophoto images, scale 1:5,000; divided in 4 categories
Lithology features (LITHO)	extracted from the geological map, scale 1:200,000 (Geological Institute of Romania); divided in 5 categories
Soil texture (TEXT)	extracted from the soilo map, scale 1:200,000 (National Research & Development Institute for Pedology, Agrochemistry and Environment Protection); divided in 3 categories
Number of households in the defined area (NH)	derived from the orthophoto images, scale 1:5,000
Population density, number/defined area (PD)	Calculated by National Institute of Statistics; reported at built-areas level (LAU 2)

Table 3

Lithology and soil texture categories extracted from the geological and soil maps

Description	**Code**
Phyllites, sericitous-chloritous schists	LITHO1
Gravels, sands and loess-like deposits	LITHO2
Micaschists, Paragneisses	LITHO3
Sandstones, marly clays, marls, sandstones (flysch)	LITHO4
Bituminous shaly clays and siliceous sandstones	LITHO5
Varied texture	TEXT1
Loamy	TEXT2
Loamy-sand…loamy-clay	TEXT3

Quantifying spatial data

The spatial analysis of the environmental and anthropogenic influences on nitrate concentration in shallow groundwater was undertaken for three buffering zones (Fig. 2), shaped around each water sample: buffer = 100 m (3.14 ha), buffer = 200 m (12.56 ha) and buffer = 300 m (28.26 ha). This approach is aimed at identifying and quantifying the influence of the explanatory factors around each water sample at various scales. Furthermore, the buffer zones were chosen in order to determine which datasets explain better the nitrate concentration in the shallow groundwater through regression analysis.

Excepting the depth to water, which remained the same, all explanatory factors were calculated using zonal statistics account. Thus, the percentage of each category of land use, lithology and soil texture was determined depending on the buffer area. *The slope declivity mean* was calculated as Weighted Arithmetic Mean within each specific buffer. *The number of households* was determined depending on the number of units within each specific buffer. Similarly was determined the *relative population density* in each specific buffer, depending on the total number of habitants and built-up areas within each locality in the study-area.

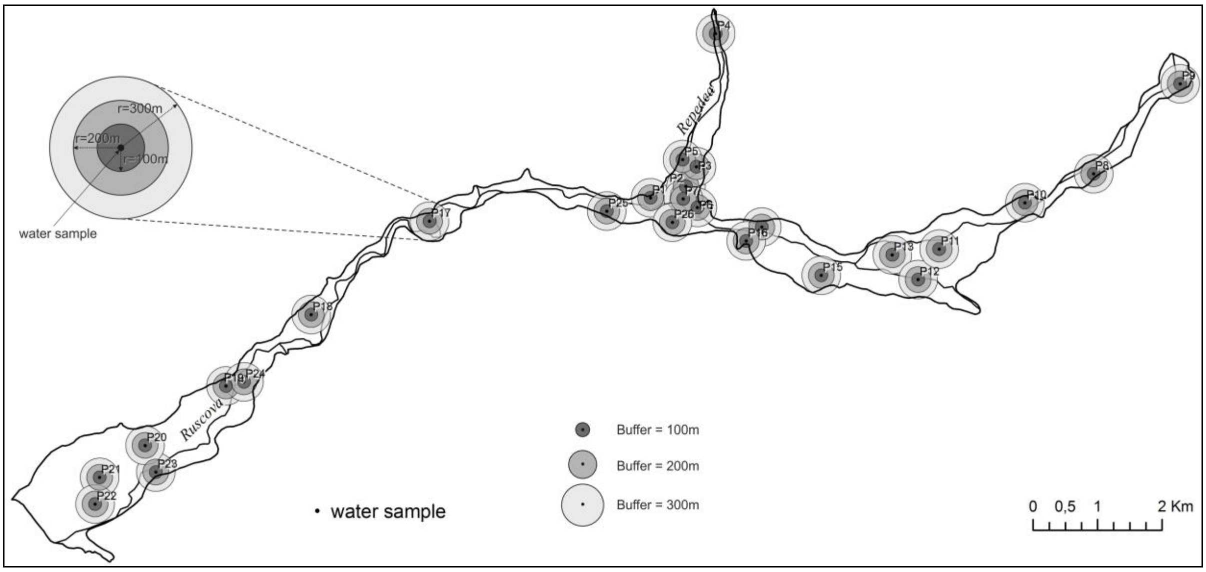

Fig. 2 – Water samples with 100 m, 200 m and 300 m buffers.

Statistical analysis

Data normalization. For the statistical analysis, all data (including nitrate concentration) were normalized into the range 0…1. Normalization was done by *Min-Max linear transformation* of the input data in order to achieve similar data range (Eq. 1).

$$N = (X - X_{min})/(X_{max} - X_{min}) \quad (1)$$

where N = normalized data (0…1); X = the dataset; X_{min} = minimum value from the dataset; X_{max} = maximum value from the dataset.

Multiple linear regression models. For estimating the correlation between the independent variables and the nitrate concentration in shallow groundwater the multiple linear regressions (MLR), performed using SPSS statistical software package (Statistical Package for the Social Sciences), was applied. The aim of this paper was not to obtain a model which describes nitrate level in the study-area, but to test the importance of different factors on nitrate pollution. For this reason, all the selected explanatory factors were included in this analysis.

NO_3^- values were used as dependent variable and DW, SD, BU, AL, GRASS, OTH, LITHO1, LITHO2, LITHO3, LITHO4, LITHO5, TEXT1, TEXT2, TEXT3, NH, PD as independent variables. MLR was performed using backward method (stepwise elimination) until only those which were significant ($p < 0.05$) were maintained in the models. Moreover, models were created for each of the three different buffer sizes defined for the current study.

The standardized beta coefficients *(ß)* were used by determining the relative influence of the analysed independent variables on nitrate concentration. For statistical tests, the following interpretation of p values was used: $p < 0.05$ = significant, $p < 0.01$ = highly significant, $p < 0.001$ = extremely significant, $p > 0.05$ = not significant.

4. RESULTS AND DISSCUTION

4.1. The water sample indicators

The samples show that the shallow groundwater temperature during the sampling campaign varied between 8 and 16°C, with highest values within populated areas. The *pH* values fall into the neutral-alkaline range, with highest values in P16, P20, P2 and P18. Generally, the nitrate concentration values are below 50 mg/L, ranging between <10 and 70 mg/L: 17 samples exceeding 10 mg/L and two 70 mg/L (Table 2).

The highest nitrate concentration (≥50 mg/L) was identified in individual fountains (P2 and P17 – Repedea) and public fountains (P20 and P22 – Ruscova). Here, depth to water varies between –6 and –8 meters. According to the specific buffers, the environmental conditions show sedimentary rocks (gravels, sands) covered by soils with varied textures. The foremost land use categories are represented by built-up areas and arable lands.

Lowest values (≤10 mg/L) were identified in wells/hand pumps in Paleogene rocks (sandstones, marls, marly clays) mainly covered by soils with loamy and loamy-sand…loamy-clay texture. Here, the main land use/land cover categories are grasslands and forests or shrubs.

4.2. Spatial variability of nitrate concentration

The spatial variability of nitrate concentrations in the shallow groundwater of the study-area was obtained using a simple interpolation method: IDW (Inverse Distance Weighted). The map (Fig. 3) indicates increasing values from upstream to downstream of the Ruscova Valley, where the built-up areas and arable lands are more extended. The lowest values were spatialized in the eastern half (approx. 10-20 mg/L), with a minimum in Poienile de Sub Munte locality. The highest values of nitrate concentration are specific for Ruscova locaity, with maximum values near the Ruscova–Vişeu rivers confluence area (50-70 mg/L).

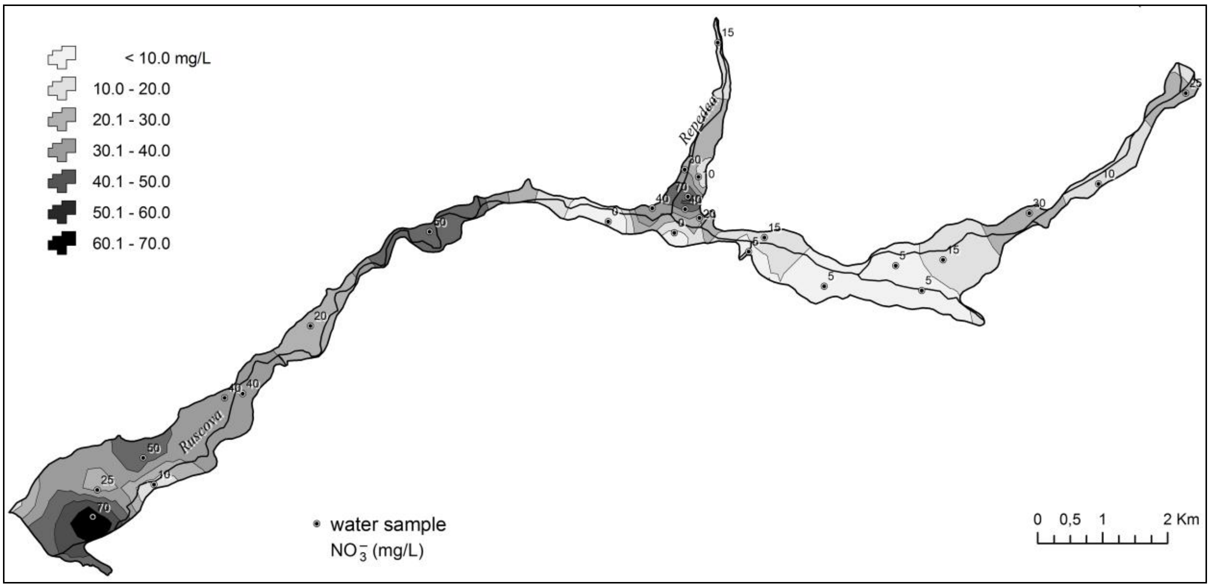

Fig. 3 – NO_3^-concentration in the shallow groundwater.

Generally, the mean NO_3^- value within the study-area, calculated as *Weighted Arithmetic Mean*, is 30.0 mg/L, with differences between localities: 17.0 mg/L in Poienile de sub Munte, 33.0 mg/L in Repedea and 42.0 mg/L in Ruscova.

4.3. The influence of the analysed explanatory factors on nitrate concentration

In this study-area, the content of nitrates in the shallow groundwater can be explained only relying on both environmental and anthropogenic factors. The different values resulted from the regression models applied for the three different buffer zones explained how different variables could become statistically significant at various scales (Table 4).

Table 4

Results of multiple linear regressions (coefficients *ß)* at different scale

Independent variables	buffer zone = 100 m	buffer zone = 200 m	buffer zone = 300 m
DW	^{x}	^{x}	^{x}
TOPO	^{x}	^{x}	0.738^*
LITHOcateg.1	^{x}	2.538^{***}	^{x}
LITHOcateg.2	^{x}	4.254^{***}	^{x}
LITHOcateg.3	-0.67^*	2.320^{***}	^{x}
LITHOcateg.4	^{x}	3.023^{***}	^{x}
LITHOcateg.6	^{x}	^{x}	^{x}
STcateg.1	^{x}	^{x}	^{x}
STcateg.2	^{x}	^{x}	-0.759^*
STcateg.3	^{x}	^{x}	-0.955^{**}
BU	14.576^{**}	^{x}	1.115^*
AL	20.668^{**}	^{x}	^{x}
GRASS	14.734^{**}	^{x}	^{x}
OTH	^{x}	^{x}	^{x}
PD	-2.198^*	^{x}	0.894^*
NH	^{x}	^{x}	^{x}

*** $p < 0.001$ = extremely significant; ** $p < 0.01$ = highly significant; * $p < 0.05$ =significant; ^{x} $p > 0.05$ = statistical not significant

In relation to the number and location of the water samples, the spatial data used and the particularities of the study-area, the multiple regression coefficients indicate the following:

1) The best results in terms of statistical significance were obtained only when all sixteen independents variables were used concurrently in the models. Multiple linear regression results which involved environmental or anthropogenic explanatory factors alone are not statistically significant ($p > 0.05$) for each independent variable. These can explain that the shallow groundwater pollution is a complex process, which depends on both environmental and anthropic factors;

2) At minimum local scale (buffer zone = 100 m), built-up areas and agricultural activities have a strong influence on nitrate concentration (BU = 14.58, AL = 20.67; GRASS = 14.73). These can be explained because the analysed water samples are generally located within populated areas, where these land use categories prevail. The regression coefficient of PD (-2.198) indicates a low inverse relationship between population density and nitrate concentration explained by the reduced number of inhabitants within specific area (mean = 94.2);

3) At medium scale, buffer zone = 200 m, agricultural activities (AL = 10.71) and build-up areas (BU = 6.28) have a strong influence on the nitrate concentration. Also, a direct influence of lithology on nitrate contents was noticed. Thus, the regression coefficients indicated a decreasing in NO_3^- values depending to the rock's permeability;

4) At maximum scale used in present study (buffer = 300 m), the highest values from the dataset indicated a direct influence of BU (1.12) and PD (0.89) on NO_3^- concentration. This can be explained by the extent of the built-up area and the high number of inhabitants in the analysed buffer (mean = 394.2). Moreover, the SD has direct influences on nitrate contents, indicating that NO_3^- tended to be high when the mean slope gradient increased. This can be explained by the topographic characteristics which are favouring the nitrate accumulation.

5) The DW (depth to water), LITHO5 (bituminous shaly clays and siliceous sandstones), TEXT1 (soil varied texture), OTH (forests, shrubs, water courses, alluvial deposits and bare soils) and NH (number of households) are not statistically significant ($p > 0.05$) for all buffer sizes. This means that these independent variables cannot explain nitrate concentration in the shallow groundwater at these defined scales.

6) Finally, the regression coefficients indicate changes in the number of explanatory factors and their influence on nitrate concentration, depending on buffer zones. In other words, anthropic factors tended to be more important as the distance to wells/hand pump decreased. Thus, the results highlighted that the model associated with 100 m buffer displayed land use as the explanatory factor with the strongest influence. As the distance increases, the environmental factors tend to become more important. The results for 200 m and 300 m buffer indicated the importance of soil texture and especially lithology which proved to be statistically highly significant ($p < 0.001$).

4.4. Uncertainties of data

Based on the researches carried out for the current study and the resulted outcomes, some limitations and assumptions have to be pointed out:

The number and homogeneity of water samples. The dataset include only 26 water samples, inhomogeneously distributed over a surface of 1,230 ha. This can be explained because in some cases the access to the private wells/hand pump was restricted. Also, for certain households, the water sources are represented by artesian springs, located in the nearby slopes.

The determination method of nitrate concentration. Nitrate concentration in the water samples was determined using nitrate-test strips. In this case, in comparison with the laboratory results, these values are considered approximate.

The period of the vegetation period when the samples were determined. Nitrate concentrations in the shallow groundwater can vary substantially depending on vegetation season. Groundwater pollution studies indicate that trees and plants may temporarily store large quantities of nutrients during the growing season for release to aquifers during subsequent nongrowing season (Walker, 1973). Thus, we can deduct that during the nongrowing vegetation period the nitrate concentration in the shallow groundwater can be slightly higher.

Therefore, the current approach should be used as a preliminary study concerning the nitrate concentration in the shallow groundwater in relation to its main explanatory factors.

5. CONCLUSIONS

The current study refers to the assessment of the shallow ground water quality within a rural area located in Maramureş Mountains focused on the nitrate concentration and their environmental and anthropogenic explanatory factors.

The datasets were analysed according to three buffer zones in order to identify and quantify the role of the explanatory factors at various scales, in the buffer zones set around each water sample. Furthermore, multiple linear regressions were used to test the importance of these explanatory factors on nitrate pollution.

In the Ruscova Valley only in few analysed water samples the nitrate concentration values were below 10 mg/L. Frequently, the nitrates concentrations were 10-30 mg/L or higher, indicating the pollution of shallow groundwater due to anthropic activities (including application of fertilizers, human and animal waste, septic tanks). As a result, within the study-area, the nitrate concentration varies, registering lower values in the eastern half (10-20 mg/L), and higher values near the Ruscova–Vişeu rivers confluence area (50-70 mg/L).

According to the statistical results, the shallow groundwater pollution represents a complex process which depends on both environmental and anthropogenic factors. The greatest correlation coefficients (max. 14.734), with *p* statistically significant (< 0.05) were obtained when the regression models involve all sixteen independent variables. Therefore, regression models applied at buffer zones indicate that anthropic factors tended to be most important close to wells/hand pumps. However, as the distance increase, the environmental factors tend to become more significant.

Given that the increase of nitrates concentration in shallow groundwater may be also influenced/triggered by other factors, a future study aimed at including the quantitative data concerning fertilizers used in agriculture, the number of livestock farms, the number and proximity to stables, the number and location of latrines and septic tanks etc. which can explain better the nitrate pollution in this area.

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