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ASSESSING LAND SUSCEPTIBILITY FOR POSSIBLE GROUNDWATER POLLUTION DUE TO LEACHING – A CASE STUDY ON ROMANIA

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Abstract: Pollution can occur in any environment, but the way the pollutants spread over depends on the environmental features. Some pollutants remain more or less confined to the contamination site, others do not. This paper proposes a territory zoning showing lands susceptibility for possible groundwater pollution through leaching in Romania, if pollution occurs. The method uses a GIS technique and takes into account soil permeability and texture, relief type, depth to groundwater and climatic water deficit. Six land susceptibility classes were obtained for the whole territory. The highest vulnerability to groundwater pollution was assessed for the most permeable sandy soils, or near - river soils, whereas the most resilient environment was assessed for the low permeable clayey soils. This land classification is aimed at drawing attention to stakeholders in order to rank and take the most appropriate measures to prevent and control pollution, if occurring. The regions that are most vulnerable to pollution should be managed with more care than the most resilient regions. If global warming continues, more severe rainfall events are expected to happen, thus enhancing the leaching of pollutants toward groundwater, specifically for the most vulnerable classes.

Keywords: zoning; soil texture; hydraulic conductivity; landforms; climatic water deficit

1. INTRODUCTION

Industry and agriculture contribute to economic progress but they also have negative consequences, such as the environmental pollution of air, soil, surface waters and groundwater that occurs mainly through infiltrations through the soil surface.

The main factor responsible for environmental pollution is its source, either diffuse or punctual, and the outcome of the pollutant depends on pollution magnitude and the nature of the polluted environment.

Spilled crude oil as well as the by-products of iron and steel production and urban trash are among the most severe pollutants. In many countries, benzene, toluene and other hydrocarbons were found in groundwater as reported by Senthil Kumar et al., (2017) for India, Riyadh et al., (2018) for Qatar, and Ifelebuegu et al., (2017) for Nigeria. Such pollutants found in groundwater represent a serious risk of neurologic, liver, kidney, heart and cancer diseases (USEPA, 2005; Williams et al., 2006; WHO, 2008; Yang, et al., 2017; Popek, 2018).

In agriculture, the application of some pesticides or essential fertilizers like nitrate nitrogen, potassium and phosphorous compounds excessively on the soil surface or poorly correlated with crop development or rainy periods also pollutes the environment, including groundwater, mainly by nutrient leakage (Davidson & Kanter, 2014; Musacchio et al., 2019; Durukan et al., 2020; Kakar et al., 2020).

The flow of water and pollutants through the soil toward groundwater is a complex process primarily governed by gravity and soil water suction, as previously mentioned by Biggar & Nielsen (1967), Catt (1991), Paltineanu (2001), Kostecki et al., (2005), Lăcătușu et al., (2019a, 2019b), Zhao et al., (2009), Chaozi et al., (2019), Domnariu et al., (2020), Lacatusu et al., (2021), and Paltineanu et al., (2021).

Water content, direction of flow and hydrologic regime of the soils resulting from the climate type (precipitation, reference evapotranspiration, and irrigation application), soil characteristics and vegetation water uptake of the crops and wild flora represent other important factors governing the transport of water and pollutants through the soils.

In addition to soil texture, another main factor determining the magnitude of soil permeability is soil porosity, especially macroporosity. These two properties combined with other soil properties (e.g. humus or carbon content, Na^{+} content, Ca^{+2} content, etc.) govern the flow of pollutants through soils, eventually ending up in the water. Soil permeability for some solutions or suspensions in the soils generally differs from soil water permeability (Kostecki et al., 2005). Previous studies discussed environmental aspects such as soil-plant-water-climate relationships (climate characteristics, soils types and their properties, crop water requirements, etc.) for some parts of the territory of Romania (Paltineanu et al., 2000, 2002, 2017, 2020, Paltineanu & Chitu, 2020).

The land forms also have a great importance, because high-altitude mountains, platforms or sloped hills normally favor runoff toward surface waters compared to leaching, while low-altitude fields like flood plains or low terraces favor accumulation of pollutants. Groundwater depth also depends on the relief type; a deep-water table hampers or prevents pollutant movement from reaching groundwater, while a shallow water table favors its quick transport to itself.

Hydraulic conductivity of the geological deposits underneath the soils presents a wide range depending on their nature. Some sedimentary deposits like sands or gravel favor a quick pollutant transport to groundwater, while massive and unbroken rocks or clay and compact marl deposits impede them.

Some papers emphasized the role of soil texture, climate and relief on the movement of pollutants through the soils (Paltineanu, 2001; Kostecki et al., 2005, Domnariu et al., 2020, Lăcătușu et al., 2021, Paltineanu et al., 2021). However, the soils show a different behavior concerning the movement of pollutants on their way toward groundwater, and there is a need to classify and evaluate the territory according to some natural or environmental factors.

The purpose of this paper is to evaluate land susceptibility for possible groundwater pollution caused by leakage depending on soil permeability, landforms, depth to groundwater and climatic water deficit for the territory of Romania, if pollution events involving fluid or water-driven substances occur, in order to establish priorities for both prevention and control.

2. MATERIAL AND METHODS

The material in this study is represented by the entire land covering Romania, with a special focus on soils. Soil texture involving the particle size distribution over the entire soil depth was used to estimate soil permeability with saturated hydraulic conductivity (Ks) values as reported by Canarache (1990), or derived from the relationships reported by Paltineanu (1998) and Paltineanu et al., (2003). Thus, soil texture is the key natural factor that can either impede (e.g. when clayey) or favor (when sandy) pollutant movement through the soil toward groundwater.

Because this paper focuses on land susceptibility to leaching based on the permeability and texture of the soils, the following considerations stress the differences between, for instance, three soil types possessing various physical and chemical properties: 1) a sandy *eutric psamosol* according to Sistemul Român de Taxonomie a Solurilor (Florea et al., 2012) or *Eutric Arenosol*, symbol AR-eu, according to World Reference Base for soil resources (2014), located at Potelu-Ianca, Olt county, 2) a loamy *cambic chernoziom* or *Haplic Chernozem*, CH-ha from Grindu, Ialomița county, and 3) a loamy-clayey *chernic-argic faeoziom* or *Luvic-Chernic Phaeozem*, PH-ch-lv, from Drăgănești, Teleorman county, all sites from southern Romania.

The clay content, humus content and saturated hydraulic conductivity (Ks) values for these three soil types are presented in Figs. 1a, 1b and 1c. Soil clay content is very low (on average 0.5%) for AR-eu, being moderate for CH-ha (20%) and relatively high (47.4%) for PH-ch-lv. The humus content shows a similar pattern, whereas Ks differs substantially from a highly-permeable sandy soil (AR-eu), where its harmonic weighted mean is higher than 200 mm/h, to a slowly-permeable soil (PH-ch-lv) with Ks mean values of about 2 mm/h and where there are soil subhorizons with even lower Ks values that govern water flow within soils. CH-ha soil presented intermediate Ks values (17.1 mm/h). Details for the properties of these soils were previously reported by Domnariu et al., (2020), Lăcătuşu et al., (2021) and Paltineanu et al., (2021). Nevertheless, at the country level, soil permeability and texture range is much greater than the already mentioned soils, determining a large diversity of situations.

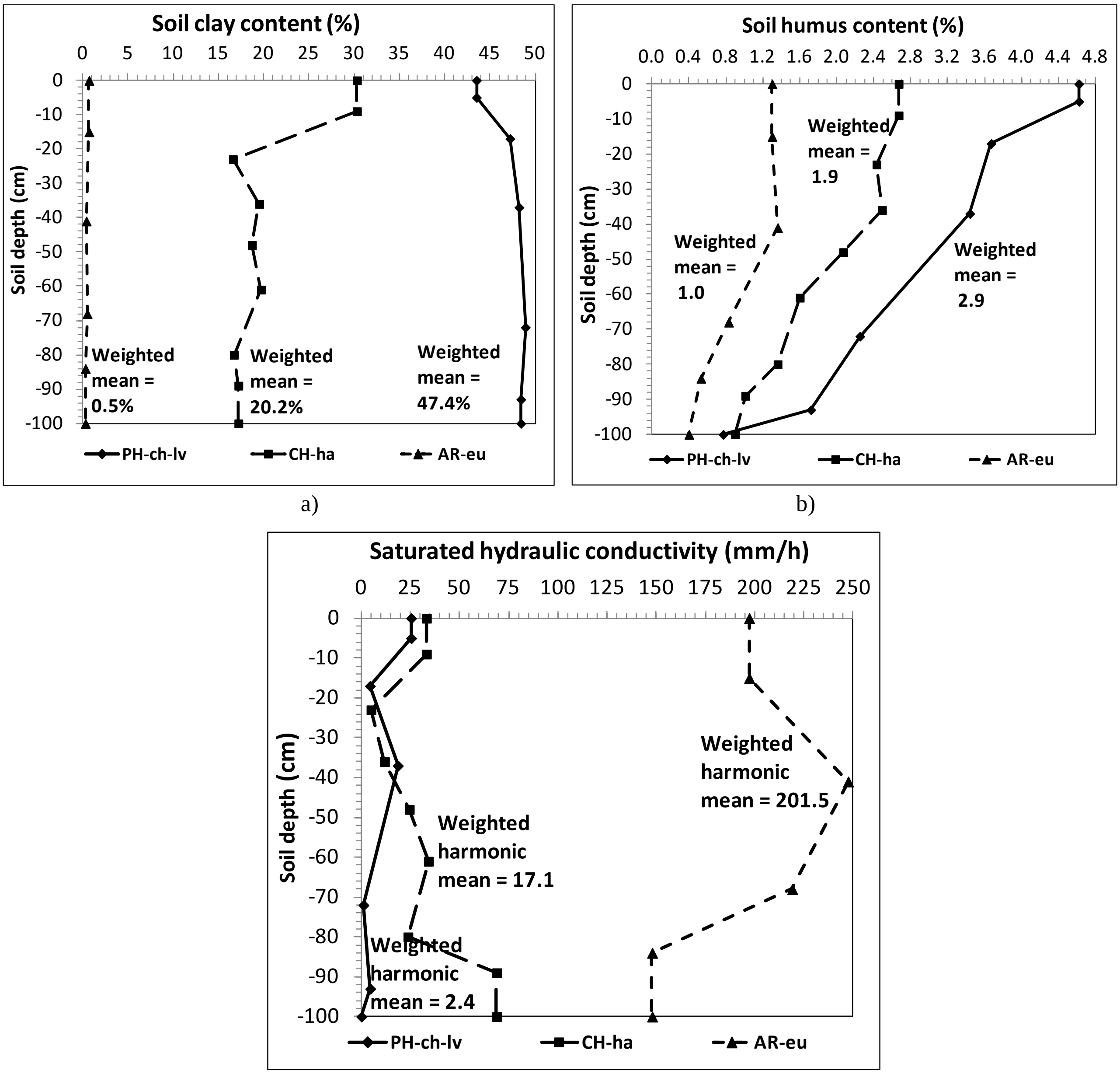
The method used in the present paper has been elaborated as based on an “expert system” method that synthesizes the above considerations, with priority given to soil texture and permeability as resulted from the previous published studies. On the soils map of Romania (Soil databases, Harta solurilor României scara 1: 200000, Archive of National Research and Development Institute for Soil Science, Agrochemistry and Environment - ICPA Bucharest) soil textures, presented as attributes belonging to

SIGSTAR-200 Databases for Soil Resources, were grouped using GIS methods to form susceptibility classes to groundwater pollution due to leaching for the whole country. Other databases used were Profisol and Monitoring, as well as Land Cover Classification System – LCCS, SRTM30 and the river network database.

3. RESULTS AND DISCUSSION

The zoning of the entire Romanian territory achieved by grouping its areas of various landforms, geology and soils to obtain classes of land susceptibility for possible groundwater pollution due to leaching resulted in the establishment of six such classes plus a water body class, described below, Fig. 2.

Class 1 is represented by difficult-to-access lands that might be affected by *accidental pollution*. Ks values generally range between 2 and 15 mm/h. Such areas include shallow or poorly developed soils like rendzinas, lithosols, podzols, humusiosols, compact rocks on the Carpathian Mountain peaks or sloped hills, or even limnisols and swamps. The climate of the class is specifically cold and wet, with climatic water values in excess (positive values, Fig. 3). Runoff and drainage are the main hydrological characteristics for this area, but pollution might occur through various human activities made in e.g. mines,



a) b) c)

Figure 1. Depth distribution of the clay content (a), humus content (b) and saturated hydraulic conductivity (c) for three soils: a sandy, eutric psamosol (AR-eu), a loamy, cambic chernozem (CH-ha) and a loamy-clayey, chernic-argic phaeozem (PH-ch-lv)

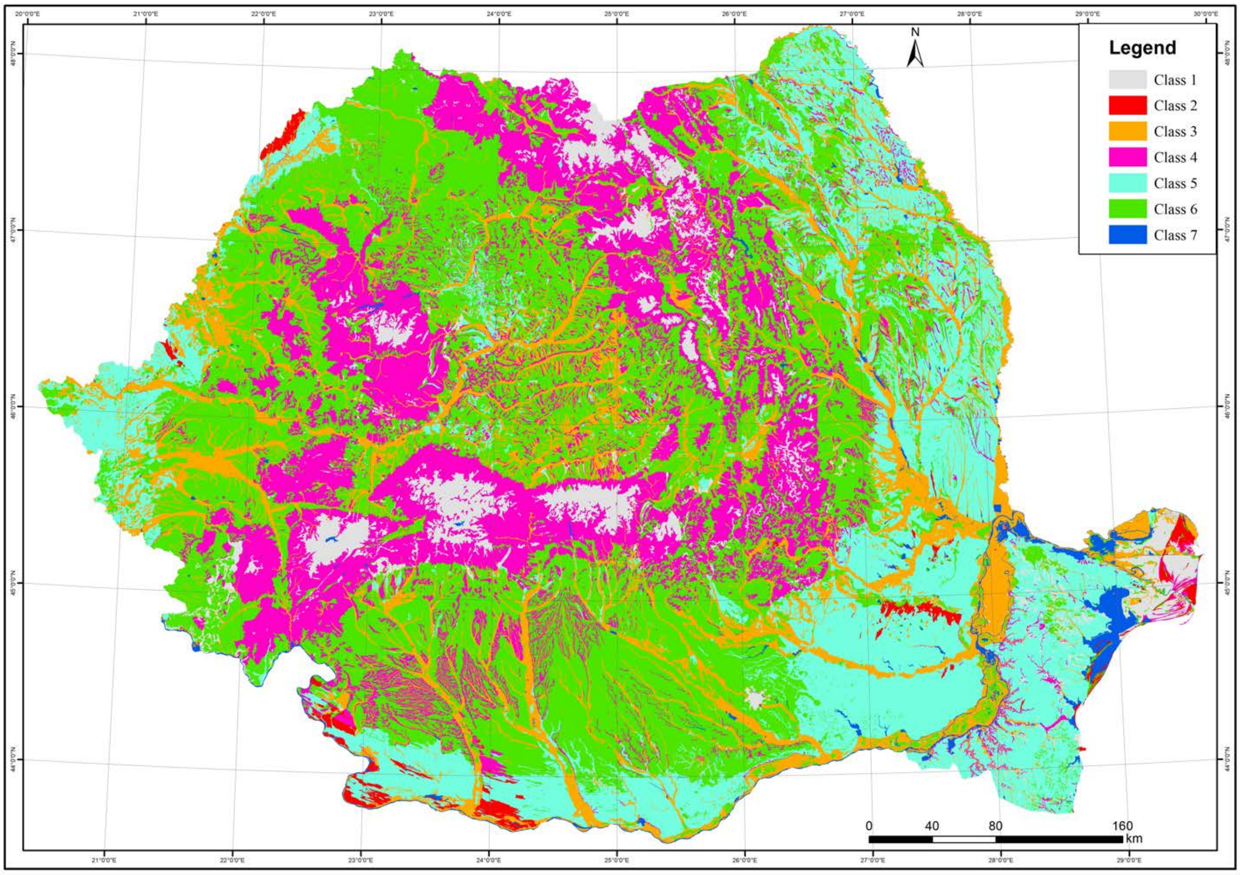


Figure 2. Land susceptibility map of Romania for possible groundwater pollution due to leaching. The legend means: class 1 represents difficult-to-access lands that might be affected by accidental pollution, class 2 encompasses lands that are extremely highly susceptible to pollution by leaching due to excessively high permeability of the soil and geological deposits underneath, class 3 includes lands that are highly susceptible to pollution by leaching due to the shallow water table and the high-to-moderate soil permeability, class 4 is formed by areas situated in mountainous or high-altitude regions and includes lands that are highly-to-moderately susceptible to pollution by leaching due to the moderately permeable soils developed over coarse or loamy texture, class 5 encompasses plains and low platforms that are moderately susceptible to pollution by leaching due to a moderate permeability, class 6 includes lands that are slightly susceptible to pollution by leaching due to low soil permeability, and class 7 represents water bodies.

and the resulted polluted water could flow toward the regions situated below. The total area of this class is 1,202,550.96 ha (representing 5.04% from the total national area).

Class 2 encompasses lands that are *extremely highly susceptible to pollution* by leaching due to excessively high permeability of the soil and geological deposits underneath. This class includes psamosols and other similar soils developed on sandy geological deposits, such as some sandy chernozems. Ks varies roughly between 100 and 300 mm/h. The main areas are located in the southern part of Oltenia region and in the north-western part (Carei, Valea lui Mihai, etc.) of the country, as well as along the flood plains of some rivers. The climate generally shows a high climatic water deficit, showing an average of around -200 to -250 mm annually in Oltenia and Carei regions, Fig. 3. Because of the semi-arid or dry climate of the regions and of the low water storage properties of the soils, irrigation application is needed for crops, and irrigation water, if in excess, might contribute to the groundwater pollution with nutrients. From the climate view point only heavy and rare rain events might produce severe leaching toward groundwater if pollution is also present.

For instance, to completely leach an amount of 200 kg/ha $NH_4NO_3^-$ beyond the root system depth of the majority of crops or plants in southern Oltenia, only an amount of 280 mm of water from rain or irrigation application is needed under a very wet soil water content regime (Domnariu et al., 2020). Such events do not occur annually, but might occur, for instance, once in 5 to 10 years. Other macronutrients like K and P can also be leached, but compared with nitrate, these leached losses are much lower as reported by Anami et al., (2008), Erickson et al., (2005), Kolahchi & Jalali

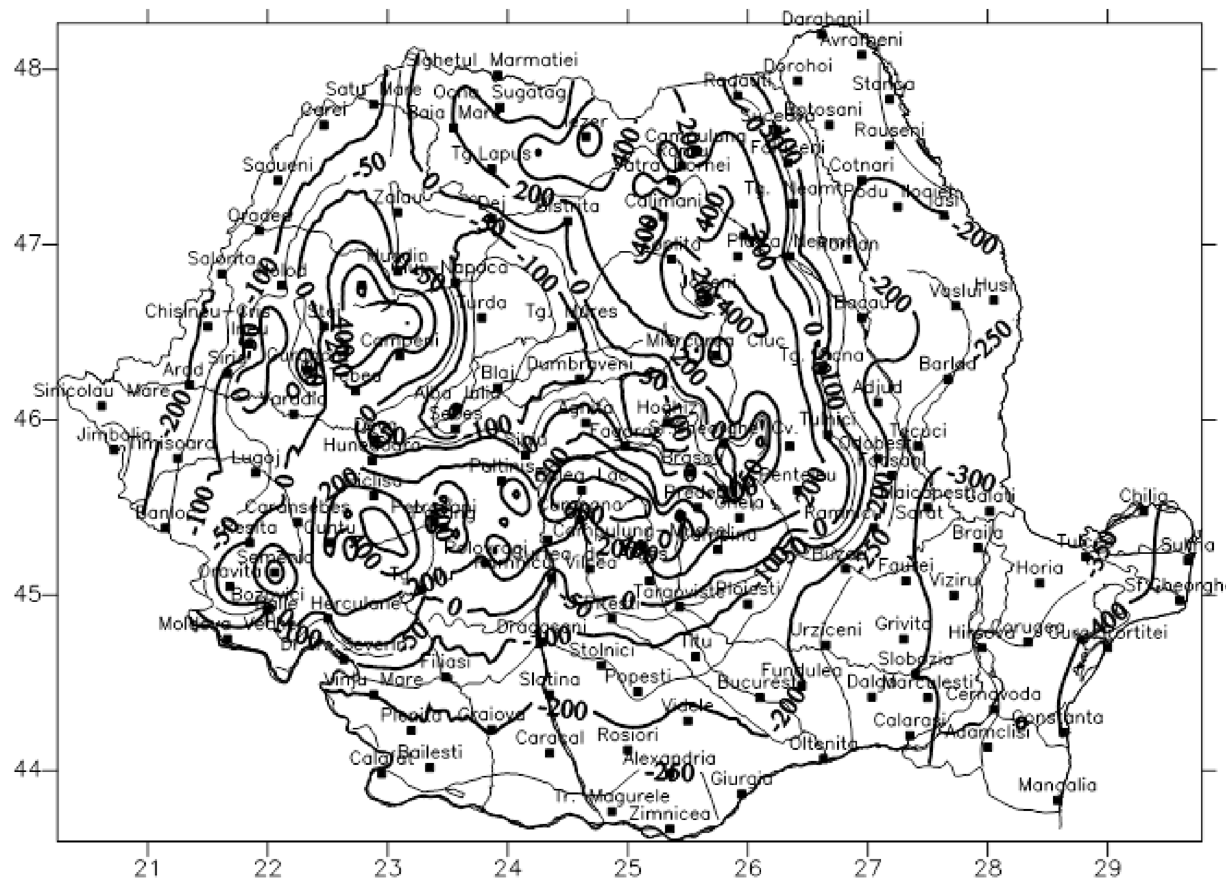


Figure 3. Annual climatic water deficit (mm) as a difference between precipitation and Penman-Monteith reference evapotranspiration in Romania (after Paltineanu et al., 2007)

(2007), Jalali & Jalali (2017), Lăcătușu et al., (2019a, 2019b), Paltineanu et al., (2020, 2021).

Not only nutrients, but also other pollutants like petroleum-derived components such as benzene, toluene, PAHs and TPHs, or other fluid or dissolved pollutants can easily be leached through such soils toward groundwater (Al-Sarawi et al., (1998); Marr et al., (2006), Edwin-Wosu & Albert (2010), Williams et al., (2006), WHO (2008), Yang et al. (2017), Popek (2018), Lăcătuşu et al., (2021)). The total land class area is low, only 214,394.98 ha (0.90%).

Class 3 includes lands that are *highly susceptible to pollution* by leaching due to the shallow water table and the high-to-moderate soil permeability. The soils generally follow the rivers or were formed and developed on alluvial geological deposits. Such soils have a coarse, loamy-sandy, sandy-loamy, loamy or a contrast texture: alluviosols, eutricambosols, gleyic chernozems, gleyic phaeozems, gleysols, etc. Ks values vary within a relatively narrow range (1 to 15 mm/h).

There is water excess within the profile of these soils during a long period of the year, and this excess is attributed to their positions in the overall relief. Because these territories are spread over a great part of Romania, their climate is variable.

Within the soils along the rivers from the mountain and high-elevation platforms and hills having a wet climate, any pollution accidents might have serious consequences for the groundwater and surface waters alike. Nevertheless, many towns and villages are situated within such areas, and polluted substances of domestic origin might also be carried to surface waters and rivers downstream.

Prevention of pollution should be carefully addressed for this class of lands. The total area reaches 3,542,726.49 ha (14.86%).

Class 4 is formed by areas situated in mountainous or high-altitude regions and includes lands that are *highly-to-moderately susceptible to pollution* by leaching due to the moderately permeable soils developed over coarse or loamy texture; however, the groundwater depth is usually large and the slopes essentially favor runoff, not leaching. The soils are represented by eutricambosols, districambosols, rendzinas, antrosols, regosols, prepodzols, luvosols, nigrosols, andosols, etc. Ks presents values ranging from 1 to 12 mm/h. The climate is wet, and pollution might affect essentially surface waters as opposed to seepage to groundwater.

As for class 1, runoff and drainage prevail, and pollution could affect not only the regions themselves, but also the ones located downward.

The area of class 4 is 4,817,666.03 ha (20.21%).

Class 5 encompasses plains and low platforms that are *moderately susceptible to pollution* by leaching due to a moderate permeability. The soils are mainly represented by the best soils from Romania: kastanozems, chernozems, phaeozems, eutricambosols, and to a low extent some luvosols. Ks ranges from about 1 to 10 mm/h.

These territories occupy vast areas in Dobrogea, Baragan, the southern part of the Danube Plain and Moldova, the Western Tisa Plain, where there is a high climate water deficit (-200 to -450 mm annually), favoring the ascendant water flux in the soils due to evaporation, and where irrigation water requirement is high. Because of semi-arid climate characteristics of the regions, irrigation application is necessary, and crop water requirements usually reach about 200 to 300 mm affecting the hydrological soil balance. Prevention should consider nutrient losses by leaching when irrigation is applied in excess in these very fertile agriculture lands.

Comparing for instance the transport in sandy soils with chernozems, for a nitrate leaching loss of 200 kg/ha $NH_4NO_3^-$ applied on the agricultural fields, a much greater amount of precipitation (400 mm) should occur (or combined with irrigation application) in order to remove it from the soil when wet (Paltineanu et al., 2020, 2021, Domnariu et al., 2020). Any kind of pollution can have disastrous consequences for both groundwater and soils. This class has an area of 4,890,169.45 ha (20.51%).

Class 6 includes lands that are *slightly susceptible to pollution* by leaching due to low soil permeability. The soils were developed on clayey-loamy, loamy-clayey or clayey deposits and possess Bt horizons showing a clay content value usually of more than 40%, and frequently between 50 and 60%, with very low water permeability. Such soils mainly are vertosols, vertic luvosols, preluvosols, stagnosols, argic phaeozems, and some argic chernozems. Ks shows very low values, varying from 0.1 to 2.5 mm/h, yet prevailing Ks values between 0.1 and 0.5 mm/h. The nature of the clayey minerals frequently belongs to the smectite-like ones (montmorillonite), with strong swallow-shrink properties, and such characteristics have the main contribution to the almost impermeable Bt horizon.

These soils occupy large areas in the central part of the Danube Plain (Câmpia Piteștilor, Câmpia Găvanu-Burdea, important parts of Olt and Teleorman counties), and also partially in Banat, Satu Mare, Transylvania and Moldova. The climatic water deficit for these soils is not high, and there is a relative balance between precipitation and reference evapotranspiration for a large part of the class territory.

However, during some droughty summer periods, especially in July and August, some crops need irrigation application and the soils are cracked in most of the cases. The leaching of fertilizers or other pollutants from such soils can occur especially through macropores and cracks during these dry periods even if the magnitude of groundwater pollution is low (Li & Ghodrati, 1994; Paltineanu, 2001; Djodjic et al., 2004; Alfaro et al., 2006; Kurunc et al., 2011, Andersson et al., 2013).

If pollution occurs, either industrially or agriculturally, the pollutants usually remain in the upper part of the soils, because the downward water flux is very low. However, when stagnogleyzation occurs during the wet and cold periods, specifically in winters and springs, there is a higher risk of ponding and runoff toward surface waters instead of leaching to groundwater (Paltineanu et al., 2000; Lăcătușu et al., 2019a; Singh et al., 2020). If such heavy-clay soils are polluted with oil-derived products, some of them would decrease soil permeability and, in the light of global warming this situation would additionally affect the environment during the droughty periods as also reported by Wei et al., (2020). Actions should not be less preventative concerning pollution for this class.

If pollution happens, the soil will primarily be endangered. Nevertheless, groundwater can also be polluted in these areas, especially in villages if the preventive anti-pollution measures are not followed (unsealed water wells, oil wells and stables, lack of impenetrable toilets a.s.o.). Still, groundwater moves even through such regions and there is a real possibility of pollution even from upstream. The area reaches as much as 8,855,913.14 ha (37.14%), being the largest class.

Class 7 represent water bodies across the country, about 318,060.51 ha (1.33%).

Totaling the class land areas resulted in a value of 23,841,481.56 ha, varying by 1,781.56 ha (0.007%) from the entire official area of Romania, i.e. 23,839,700.00 ha. This small very error at the country scale is acceptable for this kind of investigation and is due to areas used for construction or some unproductive land uses.

Human activity produces both positive outcomes and pollution. Any environment might be polluted with various substances from industry, agriculture, urban or domestic trash, etc., and the magnitude of pollution depends on the scale of the error of pollution management or accident.

Some pollutants remain more or less confined to the contamination site, e.g. heavy metals, coal dust, etc., others like soluble substances do not.

The downward water flux in soils greatly depends on the climate. Thus, the climatic water deficit represents mean annual values, and it makes sense if long periods are considered. However, in the classes encompassing highly permeable soils and high climatic water deficits, pollution of groundwater could only occur after extreme rainfall events, maybe once in 5-10 years. If the global changes continue at the same rate, extreme events i.e. higher precipitation with a more torrential character will rise in frequency and will enhance leaching of both fertilizers and other pollutants toward groundwater, specifically for the classes 2, 3 and 5.

As more detailed results will be obtained in the future with regard to the penetration of pollutants into the environment represented by soil/subsoil/geological deposits toward groundwater, this classification might be improved.

The present classification was undertaken to make stakeholders aware at both the country level and local level in order to rank and take proper measures to prevent and control pollution. The most vulnerable regions to pollution should be managed with more care than the most resilient ones.

4. CONCLUSIONS

A territory zoning showing lands susceptibility for possible groundwater pollution due to leaching in Romania was proposed in the present paper if pollution occurs, taking primarily into account soil permeability and texture, then landforms, depth to groundwater and climatic water deficit.

Six land susceptibility classes were found. The highest vulnerability to possible groundwater pollution was assessed for the most permeable soils, mainly sandy soils, or for near - river soils, whereas the most resilient environment was assessed for the low permeable clayey soils. Thus, more than 36% of the Romanian territory (classes 2, 3 and 5) is within a relatively high-risk area of possible groundwater pollution due to leaching, and only about 37% (class 6) present more resilient features.

If global warming continues, higher precipitation will enhance leaching of pollutants toward groundwater, specifically for the most vulnerable classes: 2, 3 and 5. The most vulnerable regions to pollution should be managed with more care than the most resilient ones.

The present land classification was carried out to aware the stakeholders to rank and take the best measures to prevent and control pollution.

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