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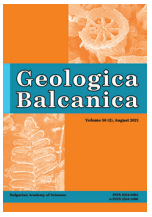

Review of investigations into the vadose zone's variable state of saturation in connection with the assessment of radon potential in Bulgaria

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Abstract. Natural radon (^{222}Rn) is a radioactive noble gas that occurs as the immediate decay product of radium (^{226}Ra), part of the ^{238}U family, in the lithosphere. Radon is driven by advection and diffusion with soil gas throughout connected and water-unsaturated pores and/or cracks in permeable rocks and soils. The aim of the present study is to do a review of the existing so far research activities in Bulgaria in connection with the observation and/or evaluation of the degree of water saturation of the near-surface layer, and on that base to distinguish the up-to-date achievements in regards to the radon potential in situ evaluation. Due to this review, the studies in Bulgaria concerning moisture dynamics in the near-surface layers can be divided mainly into two groups. The first one investigates the hydraulic characteristics (parameters) of soils in the vadose zone. Based on that, conclusions or computer simulations for the saturation degree estimation can be drawn. The other group includes in situ observations by sensors on the change of moisture with time. The results of these studies may serve as a base for more precise moisture dynamics assessment at sites with specific radon potential tendencies.

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INTRODUCTION

Natural radon (^{222}Rn) is a radioactive noble gas that occurs as the immediate decay product of radium (^{226}Ra), part of the ^{238}U family, in the geological environment (lithosphere). It is also connected with the decay of natural uranium, found to varying degrees in a wide range of rocks and soils, and in building materials. Different types of rocks and soils possess different ^{226}Ra content, therefore radon concentrations vary in the near surface field. Radon has high mobility and is driven by diffusion and convection with soil gas throughout connected and water-unsaturated pores and/or cracks in permeable rocks and soils - processes described in several review articles (*e.g.*, Etiope and Martinelli, 2002; Appleton and Miles, 2010). In this regard, the physical characteristics of soil's layers, especially its permeability, can also affect the flow of ^{222}Rn (Porstendörfer, 1994; Nazaroff *et al*., 1998). There-

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fore, the radon potential of an area, which describes the probability of high indoor radon concentration, could be dependent on not only the geological make-up as a constant source of radon but also the changes of the saturation state of the ground. Thus, there is a need for systematic review of the existing studies concerning the moisture dynamics in the near-surface geoenvironment from the viewpoint of radon potential impact.

In respect to the radon potential and moisture content of the medium, certain relationships have been observed (*e.g.*, Pinault and Bauron, 1996). In accordance with the large field area relationships, two tendencies have been reported (Sakoda *et al*., 2011). The first one represents results that concern the increase in radon potential with the increase in moisture content of the media (Arvela *et al*., 2016). In this case, the geological medium is represented by granites and similar magmatic rocks, *i.e.*, potentially fractured rocks. The second tendency shows that the increase in water content in sedimentary rocks and soils leads to a decrease in radon potential, *i.e.*, so-called "screening effect" (Jönsson, 2001; Sakoda *et al*., 2011).

In Bulgaria, studies concerning the measurement of radon in particular geomorphological units and/or evaluation of the radon potential, applying the different approaches over the whole territory, are being done nowadays (Ivanova *et al*., 2019; Antonov *et al*., 2020; Nojarov *et al*., 2020; Turek *et al*., 2020). At the moment, there is a lack of detailed investigation of the near-surface hydrogeology from the viewpoint of radon potential and moisture content tendencies and relationships. An initial step in that direction was made by Valchev *et al*. (2020). The aim of the present study is to review the existing so far research activities in Bulgaria in connection with the observation and/or evaluation of the degree of water saturation of the near-surface layer, and on that base to distinguish the up-to-date achievements in regards to the radon potential *in situ* evaluation.

THEORETICAL REMARKS

For radon transport, especially for long distance migration into soils or sedimentary rocks, gas permeability should be considered as the dominant parameter because advection is generally more important than diffusion (Neznal *et al*., 2004; Neznal and Neznal, 2005; Barnet *et al*., 2008, 2019; Lucchetti *et al*., 2019).

In the unsaturated zone, liquid and gas advective fluxes are described by Darcy's law, assuming there is no interaction between those fluids. Thus, the presence of both fluids decreases the cross-sectional area available for the flow of each fluid, and its permeability, with respect to that fluid, decreases (Benavente *et al*., 2019). Hence, soil water content directly affects gas permeability, and therefore the radon transport and its eventual presence near the ground surface, especially the weather factors (mainly rainfall), have been considered (Fig. 1).

More theoretical notes about gas and water permeability, moisture content, including hydraulic functions and parameterization, could be found in Schanz *et al*. (2011), Nguyen-Tuan *et al*. (2014), and Benavente *et al*. (2019). In addition, the latter research proposed a methodology that estimates gas permeability based on soil texture (by using the so-called pedotransfer function (PTF) analysis based on translation of basic soil data into hydraulic properties) and water content with direct application of the radon studies.

REVIEW OF BULGARIAN RESEARCH ACTIVITIES

In Bulgaria, hydrogeological and hydraulic investigations of the near-surface medium have been in progress since the middle of the last century. However, they have to be summarized and analyzed from the viewpoint of radon potential investigations. Special attention is given to particular studies that allow implementation of their results for the assessment of soils' saturation state directly or by modeling. The studies concerning moisture dynamics in the near-surface layers may mainly be divided into two groups: the first one investigates the hydraulic characteristics (parameters) of the soils in the vadose zone. Based on that, some conclusions or even computer simulations for the saturation degree estimation can be performed. The other group includes *in situ* observations (with sensors) on moisture variations in time.

The first published investigations dedicated to the first group of studies were done by Spassov (1966, 1969). In them, the problems of the approximate calculation of the natural movement of moisture in the zone of aeration based on three methods – tensiometric, centrifugation and adsorption (Spassov, 1966), and of the nature of the total soil moisture potential (Spassov, 1969) were discussed. The latter specifically concerns loess and argillaceous-sandy soils. It is not of a surprise as loess soils, being mostly an unsaturated zone, cover about 13% of the Bulgarian territory. The most recent investigations

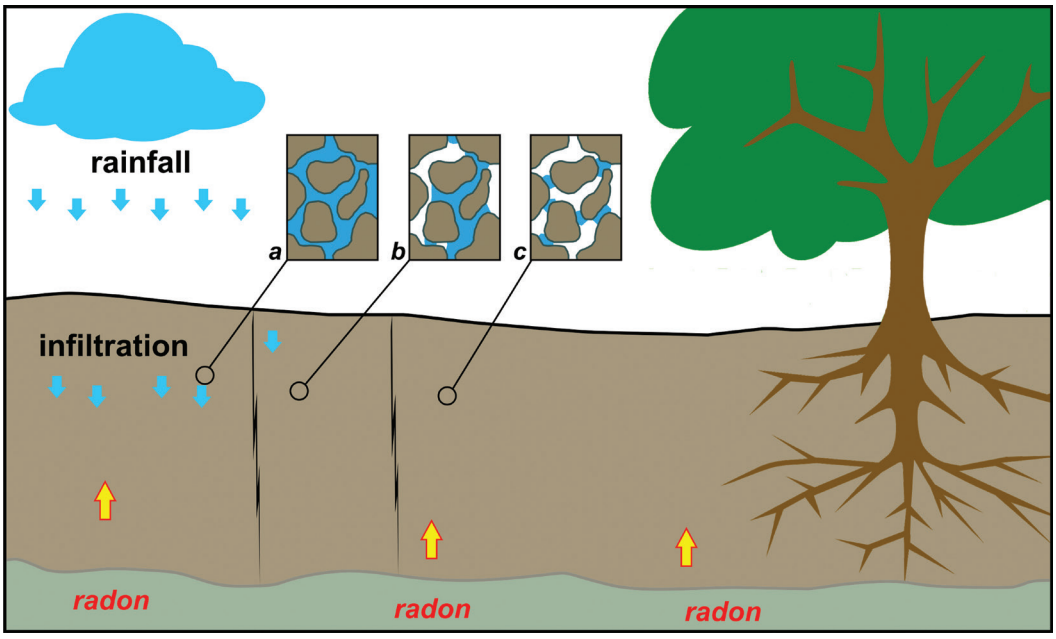

Fig. 1. Schematic spatial-temporal dependence of water and gas phases in unsaturated zone: *a*) zone of full water saturation, *b*) zone of temporally partial water saturation, *c*) steadily unsaturated zone.

connected to or describing the hydraulic characteristics of these soils are connected with the *in situ* field-scale soil hydraulic parameters derivated from all unsaturated layers in an area close to Kozloduy NPP by using large-scale infiltrometer tests (*e.g.*, Mallants *et al*., 2007; Antonov *et al*., 2012). The second group concerns real-time observations on the moisture and matrix (suction) potential of the loess complex (Gerginov *et al*. 2018; Antonov *et al*. 2018a). In these studies, results having been obtained by sensors installed at three levels below the ground, tensiometers and sensors for soil moisture content are discussed.

The first group of studies also includes implementation of PTF's hydraulic characteristics as parameterization of the hydraulic characteristics of Bulgarian soils using the model of Van Genuchten, which is based on seven texture classes (Rousseva, 1997, 2001). Also to the first group belong computer modeling studies, implementing the pedotransfer function's hydraulic characteristics with regard to flow and pollutant transport in the loess complex (Stoyanov, 2009, 2012), the Ogosta River floodplain (Benderev *et al*., 2015; Antonov *et al*., 2018b; 2019), and central Bulgaria (Stoyanov, 2008, 2018). For the loess sediment investigations, estimation of the eventual transport of several radionuclides has been made. In the Ogosta River floodplain, the simulation results showed complete water saturation of the floodplain deposits at some of the arsenic polluted spots, while only the upper layers were saturated at other sites. Similarly to the above-mentioned loess sediment case, it was implemented an approach for simulations of contaminant transport in the research works for central Bulgaria. Connected with observations of moisture dynamics at the upper part of the soil profile are the studies presented by Kolev (2016) and Kotsev *et al*. (2020), discussing the surface-groundwater interaction at the region of Valche Dere, SW Bulgaria, and at the region of Chiprovtsi, NW Bulgaria, respectively.

Apart from these two groups, the data from several regional studies (*i.e.*., Tcherkezova, 2015; Sapundzhiev, 2016; Berov *et al*., 2017; Stoynev and Ivanov, 2019) could serve as a preliminary indicator of the possibility of near-surface field saturation. Tcherkezova (2015) examined the geomorphology (geomorphometric variables) and the possibility of long-term soil saturation near river valleys. Sapundzhiev (2016) evaluated the soil texture (physical clay per cent) on Bulgarian territory. The higher the clay content in soil, the more susceptible it is to saturation. Berov *et al*. (2017) and Stoynev and Ivanov (2019) summarized and analyzed the liquefaction events of water-saturated sediments for a long-term period in the history of Bulgaria.

CONCLUSION

A review of the existing articles on moisture dynamics in Bulgaria show that main area of investigation is confined to North Bulgaria due to the widespread distribution of loess deposits. There are studies using computer models, which are based on a local floodplain soils' hydraulic characteristics that allow estimation of the daily or weekly variations of the soil saturation state. In addition, some regional works on soils susceptible to liquefaction, due to complete or almost complete saturation, exist. All these studies may serve as a base for further detailed investigation of the moisture dynamics at sites with specific radon potential tendencies.

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