Environmental radioactivity in the ground water at the Gran Sasso National Laboratory (Italy): a possible contribution to the variation of the neutron flux background

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Abstract The possible sources analyzed up to now for the neutron flux background at the Gran Sasso National Laboratory are the natural radioactivity in the rock, the concrete, as well as the induced part coming from inter-action of cosmic ray muons with the rock or the detector material itself. Water was considered only as moderator in concrete, due to its variable concentration and its radio-activity as additional source for neutron flux modulation. Therefore, the water–rock interaction and its spatial-temporal variation induced by hydrological pattern of the Gran Sasso aquifer are taken into account.

Keywords Environmental radioactivity · Ground water · Neutron flux

Introduction

The neutron flux background plays a key-role in several research activities for Neutrino Physics and Dark Matter detection in underground environment. The neutron sources considered at deep underground are (α, n) reactions on light elements (e.g., Li, F, Na, etc.), spontaneous fission, mainly of ^{238}U [1], and those induced by cosmic ray muons. The Gran Sasso National Laboratory—National Institute of Nuclear Physics (LNGS-INFN) is located inside the largest aquifer of central Italy. The natural radioactivity in rock and materials used for the internal structures of the LNGS-INFN has been studied in detail [2, 3], and the specific activities of natural radionuclides are known with high accuracy for the characterization of neutron background at the LNGS-INFN [3]. Moreover, numerical simulations [1] and neutron flux measurements were carried out inside the LNGS-INFN [3–9].

Nevertheless, the contribution of the natural radioactivity in the ground water and its spatial-temporal variations induced by the water–rock interaction and the hydrological properties of the Gran Sasso aquifer have never been considered. The measurements of the neutron flux made during the years at the LNGS-INFN revealed differences of orders of magnitude [1], but it is difficult to correlate them with the aquifer properties at that time.

Within the framework of the INFN scientific program ERMES (Environmental Radioactivity Monitoring for Earth Sciences) environmental radioactivity measurements were performed inside the LNGS-INFN: particularly, radon [10, 11], radiocarbon [12], and tritium [13]. These measurements have shown the existence of different chemical–physical and fluid dynamical characteristics in the ground water.

The measurements of the neutron flux [3–9] and the numerical simulations [1] have been performed in time intervals not exceeding the temporal and spatial variations of environmental radioactivity detected by ERMES. The results of the ground water radioactivity monitoring show temporal modulations induced by ground water variations due to percolation effect related to snow melting, and spike-like events due to rainfall and/or diffusive processes through the structural discontinuities [14].

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Therefore, this work is a preliminary study focusing on the importance of ground water radioactivity as additional source for neutron flux variation.

Experimental

Sampling and chemical analysis

The ground water samples were collected weekly in three sites located inside the LNGS-INFN underground laboratories (Fig. 1). One liter of each samples were stored in cleaned and rinsed polyethylene bottles after 5 min of water flushing at maximum flow.

The pH measurements were performed using the pH meter with an accuracy of ±0.1 pH units [15].

The conductivity of the water samples was measured with a conductivity meter MC 226 from Mettler Toledo® [15] with a relative accuracy of 1%.

The hardness of water samples was determined by an Ethylenediaminetetraacetic (EDTA) titrimetric method [15] with a relative accuracy of 1%. Mg(EDTA) salts and Calmagite as indicator were used to improve the performance.

Sample treatment and multi-element determination by ICP-MS

The water samples were diluted 10 times with deionized water and acidified with 2.5% of nitric acid to stabilize trace elements in the sample. During sampling handling reagents of trace elements analysis grade (HNO_3 super pure by Carlo Erba® Reagenti), ultra pure water (produced by Millipore MilliQ®-Element), plastic containers, and ancillary equipment were used because long-lived radionuclides Th and U at trace level had to be measured. Inductively Coupled Plasma-Mass Spectrometer (ICP-MS: quadrupole mass spectrometer from Agilent® Technologies, model 7500a) is successfully used in several applications for water quality and multi-element measurements of trace impurities in different types of water samples: drinking water, sea/lake water, waste water, and ground water [16].

This instrument was not equipped with a collision or reaction cell but a Babbington nebulizer was installed. The tuning of the instrumentation was optimized in order to reach high sensitivity, stable signal, and low background.

The concentration values for all elements were determined in quantitative mode using an external calibration curve, because the matrix effect of the ten times diluted ground water samples was negligible. During the measurements a multi-element solution was used as internal standard to correct for possible instability and drift of the ICP-MS device. The calibration curve of light mass elements (Na, Mg, K, and Ca) was normalized using ^{45}Sc while Th and U responses were corrected using ^{209}Bi. The calcium concentration was determined on the isotopes ^{43}Ca and ^{44}Ca, because the ^{40}Ca signal, the most abundant one is interfered from ^{40}Ar in this type of instrumentation. Regarding magnesium, two isotopes (^{24}Mg and ^{25}Mg) were acquired to have a double check.

The uncertainty of the measurements was in general less than 5%, which is reasonable for ICP-MS [16]. The uncertainty for K concentration values was worse than for the other elements, estimated as 30%. This was due to the high background signal at the mass of K due to the well-known interference by $^{38}Ar^1H$ on ^{39}K, ^{40}Ar on ^{40}K, and $^{40}Ar^1H$ on ^{41}K [17].

Fig. 1 The schematic view of the Gran Sasso National Laboratory. The overthrust fault (gray line) and the measurement sites (E1, E3, and E4) are shown. The analyzed ground water flows in tubes introduced in the rock up to a depth ranging from 3 m to 6 m. In the site E3 there are two sampling point: E3 parallel to the fault in North direction, and E3dx orthogonal to the fault in E4 direction
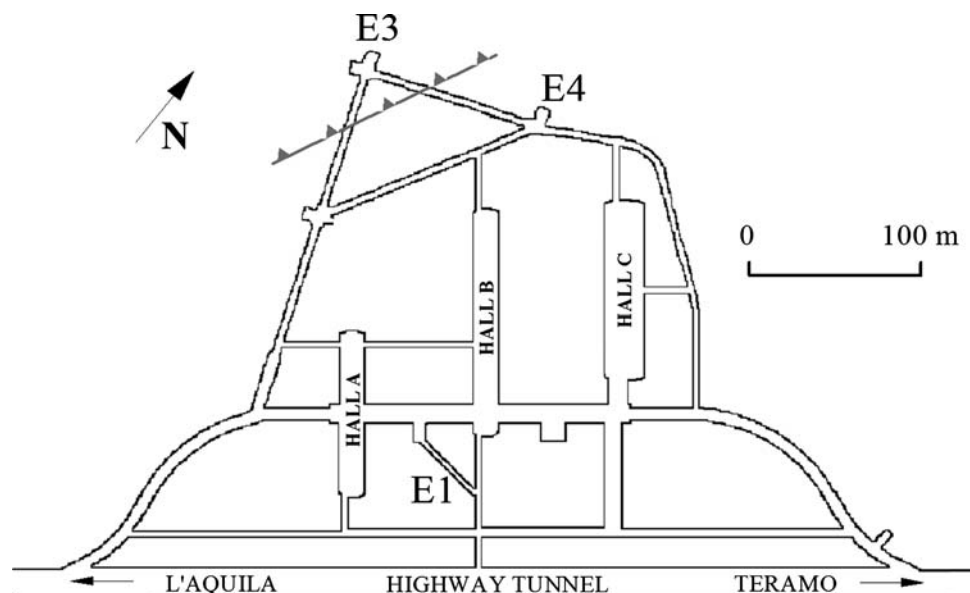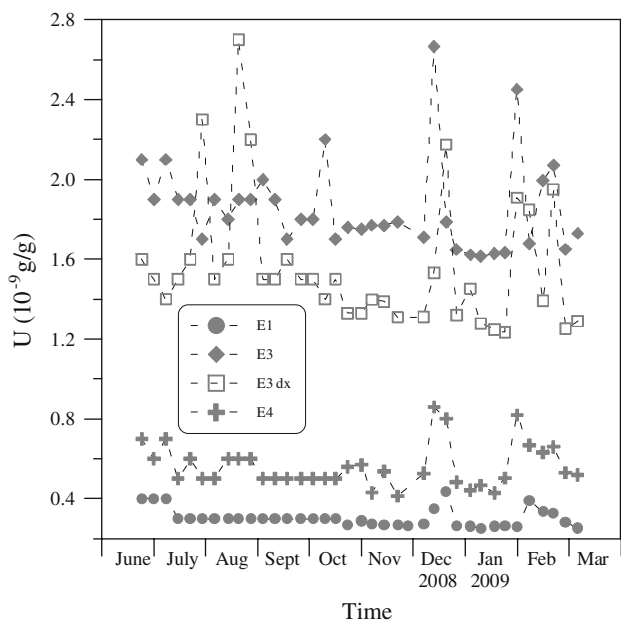
Fig. 2 The uranium concentration of the ground water sampled at E1, E3, E3dx, and E4. The relative accuracy is about 5%

Results and discussion

Figure 2 shows the uranium concentration in the ground water measured from June 2008 to March 2009 in the three different sites at the LNGS-INFN. Figures 3 and 4 show sodium, magnesium, potassium, and calcium concentrations and the chemical–physical parameters for the same water samples. The data emphasizes two different groups of water: (1) E1, E4 and (2) E3, E3dx. Moreover, the analysis of some samples on stable isotopes (δ^2H, δ^{18}O), ^{14}C and ^{3}H confirm this grouping (Rozansky K, private communication). While the second group showed clear U variations with slightly depleted stable isotopes in water (about 0.25‰ in δ^{18}O and 2.1‰ in δ^2H), the first group showed much lower U levels. The highest ^{14}C concentration was found at site E4 (71.7 pMC, percent modern carbon) and the lowest at site E3 (57.1 pMC), documenting a longer ground water-rock interaction time in the latter case. Tritium levels were in the range between 6 and 13 TU (Tritium Unit), higher than the current tritium content in rainfall of the Gran Sasso region, indicating a transit times of groundwater of the order of some decades.

Fig. 3 The sodium, magnesium, calcium, and potassium concentrations of the ground water sampled at E1, E3, E3dx, and E4. The relative accuracy is about 5% for sodium, magnesium, and calcium, and 30% for potassium
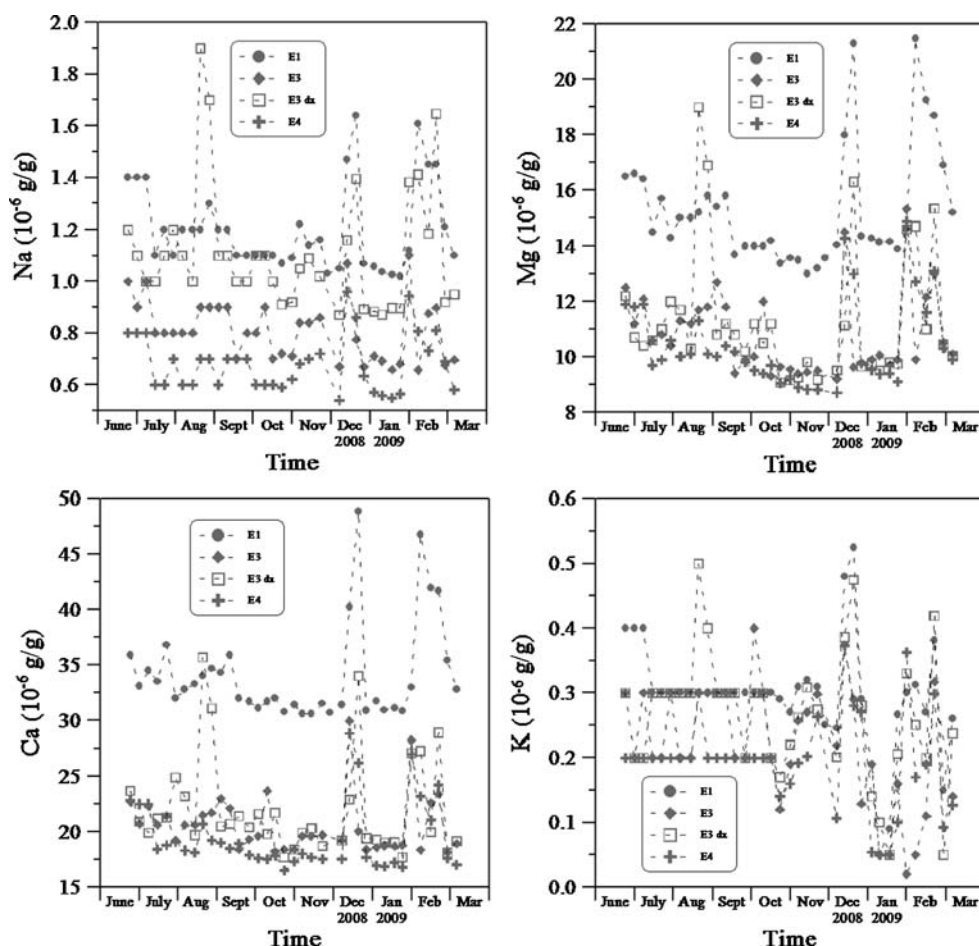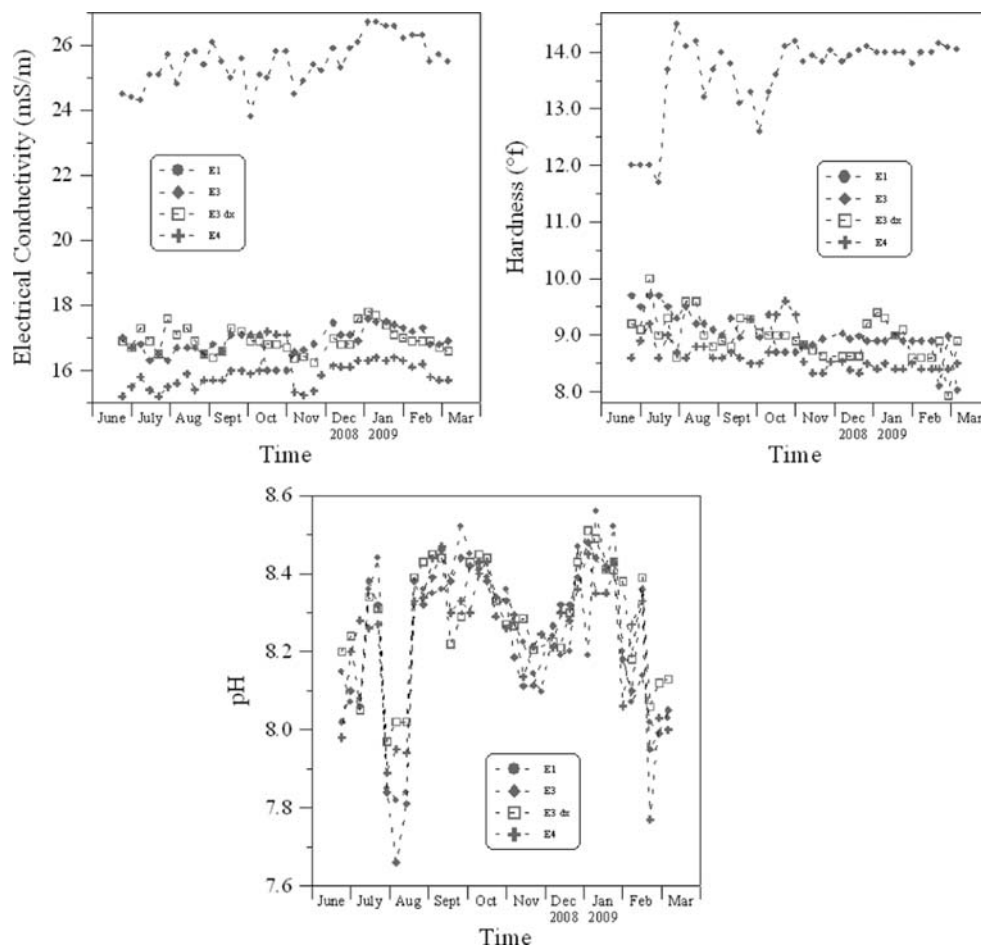
Fig. 4 The chemical-physical parameters (electrical conductivity, hardness, and pH) of the ground water sampled at E1, E3, E3dx, and E4. The relative accuracy is about 1% for electrical conductivity and hardness. The pH accuracy is ±0.1 pH units

The hydrological and hydro-geological settings have been monitored and studied during the construction of the highway tunnel and LNGS-INFN [18]. These studies have emphasized a complex structure of the aquifer due to geological and structural discontinuities, induced by the distensive tectonic phase that characterizes the Gran Sasso area. The presence of semi-permeable and impermeable structures near these discontinuities produces hydrostatic gradients as well as different hydrologic and geochemical properties in the various compartments of the aquifer. In particular, the main overthrust fault is separating water masses belonging to two distinct creeks: [11] the first one in which are embedded the main laboratories, flows in well-drained cretaceous formations, while the latter is lying within not-drained and poorly permeable dolomitic formations (Fig. 1).

The high peaks in the uranium concentration (Fig. 2) were induced prevalently by geodynamical processes through the fault and hydrological pattern due to the high permeability of the cretaceous limestone forming of the Gran Sasso massif. The water–rock interaction and ground water geochemistry modulate these variations.

The thorium concentration could not be determined and gave only upper detection limits. This can be understood as thorium compounds are generally very poorly soluble in water.

The characterization of the neutron flux background at the LNGS-INFN was done until now considering as possible sources: (1) the radioactivity in the rock and the concrete, and (2) the induced radioactivity from cosmic ray muons interaction with rock or detector itself [3–9]. Water was considered only as neutron moderator, due to its presence in the concrete [1]. Therefore, only an expected steady-state neutron background was evaluated.

Nevertheless, the radioactivity in the ground water showed spatial-temporal variations at the LNGS-INFN; for a better characterization of neutron flux background new piece of information should be considered. In particular, the fluid dynamics properties in fractured porous rock play an important role to define the uranium pattern in the ground water.

Conclusions

The neutron flux background at the LNGS-INFN is an important issue for rare events research activities as e.g., Neutrino Physics and Dark Matter search. The studies on

neutrons performed until now are not conclusive and the characterization of this background requires new measurements and analyses of the natural radioactivity in water over a suitable time–space range. The spatial-temporal variations of ground water radioactivity could induce a neutron flux modulation due to water–rock interaction and hydrological properties of the Gran Sasso aquifer. This work shows that there are two types of water with different uranium concentrations at the LNGS-INFN. Further neutron monitoring activities and numerical simulations will be scheduled to better characterize the neutron flux background at the LNGS-INFN.

References

1. Wulandari H, Jochum J, Rau W, von Feilitzsch F (2004) Astropart Phys 22:313
2. Esposito A, Pelliccioni M (1985) Nucl Sci J 22:291
3. Bellotti E (1988) Nucl Instr Meth 264:1
4. Rindi A, Celani F, Lindozzi M, Miozzia S (1988) Nucl Instr Meth A 272:871
5. Aleksan R, Bouchez J, Cribier M, Kajfasz E, Pichard B, Pierre F, Poinsignon J, Spiro M, Thomas JF (1989) Nucl Instr Meth A 274:203
6. Belli P, Bernabei R, D'Angelo S, De Pascale MP, Paoulizi L, Santonico R, Taborgna N, Iucci N, Villoresi G (1989) Nuovo Cimento 101A:959
7. Cribier M, Pichard B, Soirat JP, Spiro M, Stolarczyk T, Tao C, Vignaud D, Belli P, D'Angelo S, Wink R (1995) Astropart Phys 4:23
8. Arneodo F, Borio-Di-Tigliole A, Cavanna F, Cesana A, Chen Y, Dolfini R, Nardo R, Piano-Mortari G, Rappoldi A, Raselli GL, Rossella M, Rossi C, Tatananni M, Terrani M (1999) Nuovo Cimento 112A:819
9. Menghetti H (2006) Nuovo Cimento 29C:345
10. Plastino W, Bella F (2001) Geophys Res Lett 28:2675
11. Plastino W (2006) In: Povinec PP, Sanchez-Cabeza JA (eds) Radionuclides in the environment. Amsterdam, Elsevier, p 646
12. Plastino W, Kaihola L, Bartolomei P, Bella F (2001) Radiocarbon 43:157
13. Plastino W, Chereji I, Cuna S, De Felice P, Kaihola L, Lupsa N, Balas G, Mirel V, Berdea P, Baciu C (2007) Radiat Meas 42:68
14. Caputo M, Plastino W (2004) Geophys J Int 158:385
15. Eaton AD, Clesceri LS, Rice EW, Greenberg AE, Franson MH (eds) (2005) Standard methods for the examination of water and wastewater. American Public Health Association, American Water Works Association, Water Environment Federation, Washington, DC, p 1368
16. Becker JS (2007) Inorganic mass spectrometry: principles and applications. Wiley, New York, p 514
17. May TW, Wiedmeyer RH (1988) J Anal At Spectrom 19:150
18. Lunardi P, Catalano P (2006) Gran Sasso: il Traforo autostradale. Gangemi Editore, Roma, p 272