



## Uranium groundwater anomalies and L'Aquila earthquake, 6th April 2009 (Italy)

Wolfgango Plastino<sup>a,b,\*</sup>, Pavel P. Povinec<sup>c</sup>, Gaetano De Luca<sup>d,e</sup>, Carlo Doglioni<sup>f</sup>, Stefano Nisi<sup>e</sup>, Luca Ioannucci<sup>e</sup>, Marco Balata<sup>e</sup>, Matthias Laubenstein<sup>e</sup>, Francesco Bella<sup>a</sup>, Eugenio Coccia<sup>e</sup>

<sup>a</sup> Department of Physics, University of Roma Tre, Via della Vasca Navale, 84, I-00146 Rome, Italy

<sup>b</sup> National Institute of Nuclear Physics, Section of Roma Tre, Via della Vasca Navale, 84, I-00146 Rome, Italy

<sup>c</sup> Department of Nuclear Physics and Biophysics, Comenius University, Mlynská dolina F1, SK-84248 Bratislava, Slovakia

<sup>d</sup> National Civil Protection Department, Rome, Via Vitorchiano, 2, I-00189 Rome, Italy

<sup>e</sup> National Institute of Nuclear Physics, Gran Sasso National Laboratory, S.S. 17/bis km 18+910, I-67010 Assergi (AQ), Italy

<sup>f</sup> Department of Earth Sciences, University of Roma "La Sapienza", Piazzale Aldo Moro, 5, I-00185 Rome, Italy

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### ABSTRACT

Monitoring of chemical and physical groundwater parameters has been carried out worldwide in seismogenic areas with the aim to test possible correlations between their spatial and temporal variations and strain processes. Uranium (U) groundwater anomalies were observed during the preparation phases of the recent L'Aquila earthquake of 6th April 2009 in the cataclastic rocks near the overthrust fault crossing the deep underground Gran Sasso National Laboratory. The results suggest that U may be used as a potential strain indicator of geodynamic processes occurring before the seismic swarm and the main earthquake shock. Moreover, this justifies the different radon patterns before and after the main shock: the radon releases during and after the earthquake are much more during the preparatory period because the process does not include only the microfracturing induced by stress–strain activation, but also radon increases accompanying groundwater U anomalies.

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### 1. Introduction

Earthquake prediction is widely recognized as being among the most challenging scientific problems, both due to its societal relevance and to the intrinsic complexity of the problem, and it has been analyzed with several controversial discussions, debates, and reviews (e.g. Geller et al., 1997; Wyss, 1997; *Nature Debates*, 1999; Keilis-Borok and Soloviev, 2003; Peresan et al., 2005).

Radon ( $^{222}\text{Rn}$ ) as a possible candidate for earthquake's precursor has been studied for a long period, but there is no clear evidence that it is really a good precursor. It has been suggested as one of several possible early signals, and its groundwater anomalies associated with earthquakes and water–rock interactions were detected in several seismogenic areas worldwide indicating possible transport of radon through microfractures or the crustal gas fluxes along active faults (Ulomov and Mavashev, 1967; Scholz et al., 1973; Wakita et al., 1980, 1989; Hauksson, 1981; Monnin and Seidel, 1992; Virk and Singh, 1994; Igarashi et al., 1995; King et al.,

1995; Plastino and Bella, 2001; Richon et al., 2003; Plastino, 2006; Kawada et al., 2007). The physical processes associated with radon groundwater anomalies are based on changes of radon emanation rates occurring due to strain signal near the earthquake's nucleation point. Particularly, it is unclear its behaviour before, during and after the main shock, considering the consolidated scheme for radon release due to stress–strain processes in the rock.

In the geological environment, the radon groundwater concentration depends on the isotopic abundance of its parent radionuclides ( $^{238}\text{U}$  and  $^{226}\text{Ra}$ ), and on their geochemical patterns with reference to environmental redox and pH characteristics. The geodynamic processes induced by earthquakes can modify radon migration patterns in groundwater as a potential indicator of strain. However, to predict the activity of radon in fractured lithologies is difficult and the measurement of radon concentration does not uniquely characterize the rock deformation or the chemical inhomogeneity (Torgersen et al., 1990), as well as its relationship with the transient crustal strain signals from 'aseismic' fault slip near the earthquake's nucleation point (Roeloffs, 1999). Moreover, non-tectonic factors related to variations of chemical and physical groundwater parameters may be of importance (Shapiro et al., 1985; Plastino, 2006), requiring proper geological, hydrological and hydrogeological settings (Trique et al., 1999), so only the variations induced by stress–strain processes should be considered in

\* Corresponding author at: Department of Physics, University of Roma Tre, Via della Vasca Navale, 84, I-00146 Rome, Italy. Tel.: +39 06 5733 7277; fax: +39 06 5733 7102.

E-mail address: [plastino@fis.uniroma3.it](mailto:plastino@fis.uniroma3.it) (W. Plastino).

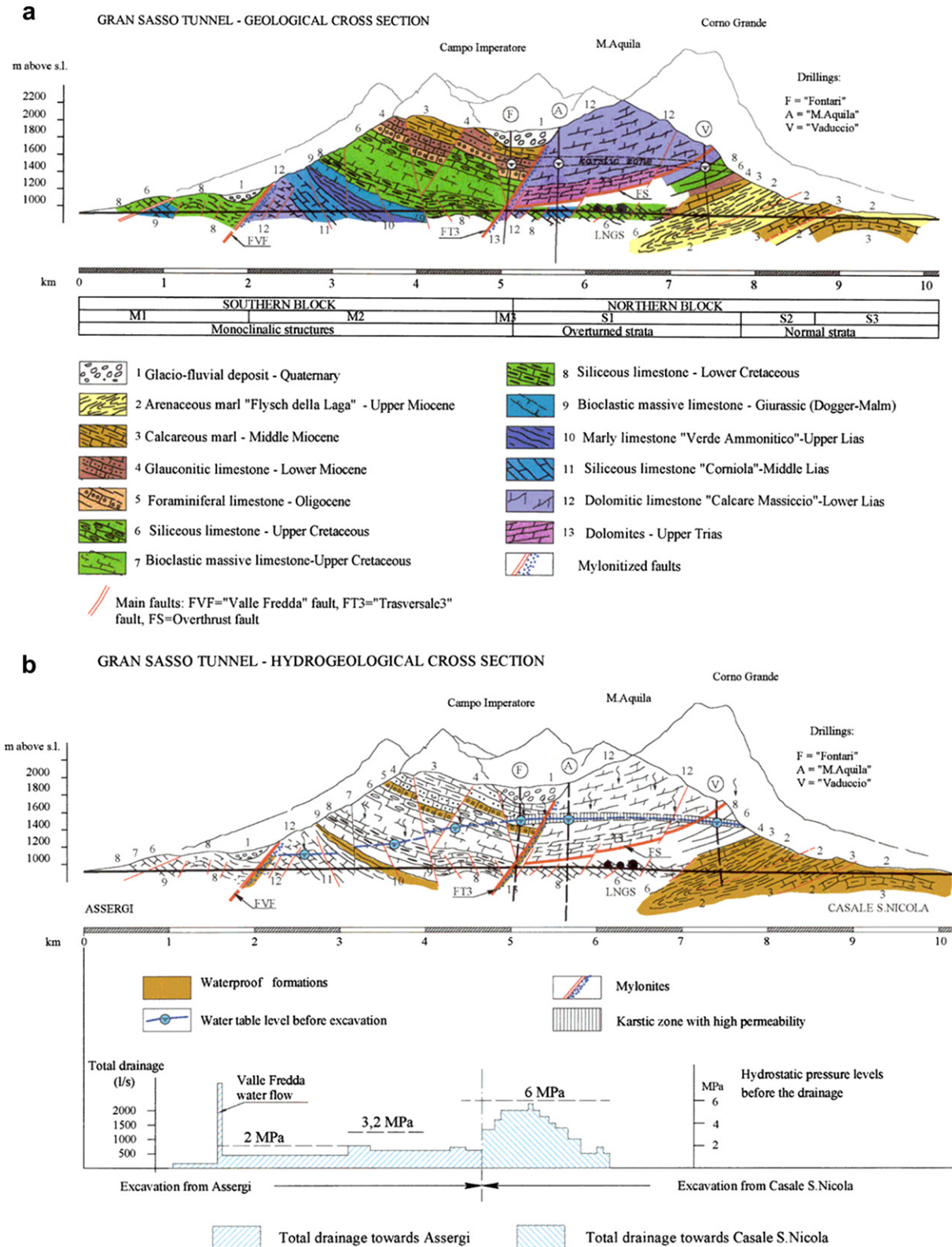


Fig. 1. The Gran Sasso massif: a – geological cross-section. b – hydrogeological cross-section. The location of the LNGS-INFN is also shown.

evaluations. Important issues are the stability and reliability of the monitoring system which should be checked continuously during the measurement time (Plastino and Bella, 2001) or the development of new detectors independent of environmental noise

parameters such as temperature, acid concentrations, humidity, and air pressure (Plastino et al., 2002).

Therefore, uranium (U) groundwater monitoring was planned and performed at Gran Sasso National Laboratory of the National

Institute of Nuclear Physics (LNGS-INFN), Italy, to study the possible pattern for radon sources in groundwater, because anomalies were detected during the Umbria-Marche (Italy) seismic sequence in 1997, located about 80 km from the laboratory (Plastino, 2006). The aim was to test U as a potential strain indicator of geodynamic processes occurring before an earthquake, rather than the consolidated scheme for radon release due to stress–strain processes in the rock.

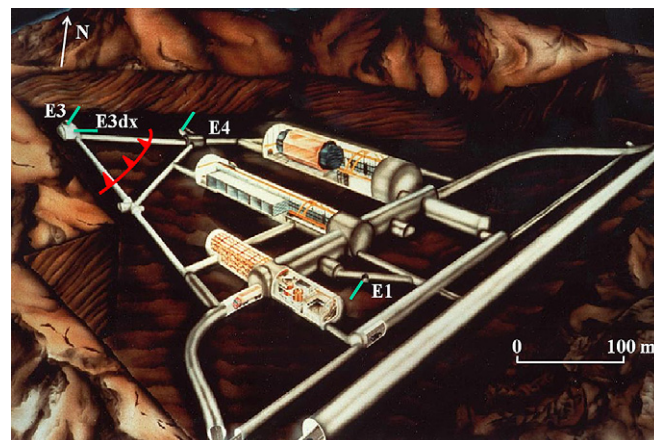
Although, in order to assess the utility of U isotopes as fluid phase earthquake precursors, U concentrations and  $^{234}\text{U}/^{238}\text{U}$  activity ratios have been monitored in thermal waters (Gorbushina et al., 1973; Kuleff et al., 1980; Finkel, 1981), this monitoring at LNGS-INFN was performed in a shallow aquifer with a high dynamic behaviour due to high permeability of the cretaceous limestones that form part of the Gran Sasso massif (Plastino and Bella, 2001). Moreover, these measurements were performed to test a possible contribution to the variation of the neutron flux background (Plastino et al., 2009), because it plays a key-role in several research activities for Neutrino Physics and Dark Matter detection in the underground environment.

The concentration of U in groundwater is usually in the range  $0.1\text{--}50\ \mu\text{g L}^{-1}$  (Giammar, 2001). In groundwaters, the weathering of U-bearing rocks and minerals is the source of dissolved U, which is most concentrated in sedimentary rocks, particularly organic shales, and is also found in significant amounts in metamorphic and igneous rocks, with higher concentrations in granites than in basalts (Gascoyne, 1992). Uranium solubility in aqueous systems is predominantly controlled by three factors: Oxidation-Reduction Potential (ORP), pH, and dissolved carbonate (Langmuir, 1980; Murphy and Shock, 1999). In aqueous solution U can exist in oxidation states of +III, +IV, +V, and +VI; however, under environmental conditions only the tetravalent and hexavalent states are stable. Clay minerals have high specific surface areas and reactive surface groups for binding metals and radionuclides through two different mechanisms: sorption can occur either in the interlayer space (fixed-charge sites) between sheets by an ion-exchange mechanism, or at the edges of the sheets through specific coordination (Zachara and McKinley, 1993; McKinley et al., 1995; Turner et al., 1996). At low ionic strength, significant sorption occurred at fixed-charge interlayer sites even at low pH, but at higher ionic strength U sorption decreases because sodium and calcium ions occupy the fixed-charge sites (Giammar, 2001).

## 2. Experimental

The mountain chain of Gran Sasso is formed by carbonatic rocks of marine origin settled during the long period of time going from the upper Trias and the upper Miocene. A series of tectonic events going from the end of the upper Miocene until the Pleistocene produced the complex structure of the present chain characterized by an anticline-syncline structure with its outmost side upturned. The main structure evolved during the first tectonic phase in a sort of break thrust and successively in an over-thrusted structure. The great overthrust event pushed the dolomitic limestone (the southern block) on top of a marly limestone formation (the northern block), representing the main syncline component (Fig. 1a) (Catalano et al., 1986). Within the massif the circulation of the great water mass permeating from the surface is greatest at the uppermost creek level, along a wide karstic zone situated between 1700 and 1600 m above sea level. The creek waters partially flow in NE direction toward the spilling springs at the contact between the marls and the limestones, and partially in SE direction, parallel to the main tectonic discontinuities oriented NW-SE (Fig. 1b) (Plastino, 2006).

The LNGS-INFN is located in the Gran Sasso massif (central Apennines, Italy) with rock overburden of 1400 m (4000 m of water equivalent), which efficiently shields cosmic rays (Fig. 2). This results in ultra low-levels of background radiation, ideal for Neutrino Physics and Dark Matter experiments. The natural radioactivity of rock and materials used for the internal structures of the LNGS-INFN has been studied in detail (Esposito and Pelliccioni, 1985), and activities of natural radionuclides are well known for the characterization of neutron background (Wulandari et al., 2004). The LNGS-INFN is located inside the largest aquifer of central Italy, within the limestone formation of the upturned syncline, near the main overthrust fault. This separates water masses belonging to two distinct flowpaths (Figs. 1b and 2): the first one, where



**Fig. 2.** Schematic view of the LNGS-INFN. The overthrust fault (red line) and monitoring sites (E1, E3 and E4) are also shown. At site E3 there are two sampling points: E3 which is parallel to the overthrust fault in the North direction; and E3dx which is orthogonal to the fault in the E4 direction into the cataclastic rocks.

the main laboratories are excavated, flows in well drained cretaceous formations, while the latter is within not drained and poorly permeable dolomitic formations (Plastino, 2006).

### 2.1. Sampling and chemical analysis

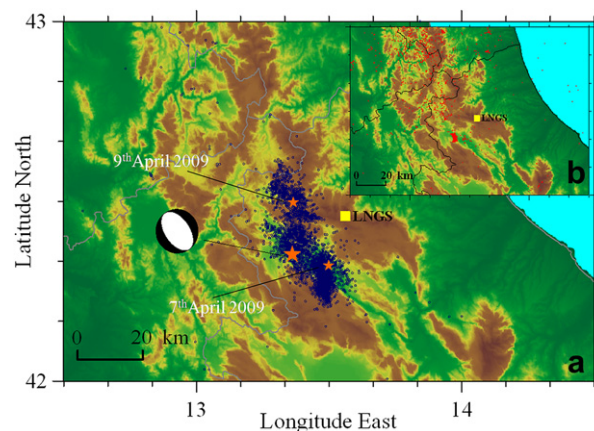
The water samples were collected weekly in three sites located inside the LNGS-INFN underground laboratories (Fig. 2). Each sample was 1 L, and was stored in cleaned and rinsed polyethylene bottles after five minutes of water flushing at maximum flow.

The pH measurements were performed by an electrometric method using a traditional glass electrode. The instrument used was the Accumet<sup>®</sup> pH meter 50 by Fisher Scientific with accuracy of  $\pm 0.1$  pH units. The potentiometric determination of electron activity was carried out with an inert indicator electrode and a reference electrode. The ORP measurements were performed using the Accumet<sup>®</sup> pH meter 50 by Fisher Scientific with precision of  $\pm 22$  mV (one standard deviation) in a closed flow cell. The conductivity of the water samples was measured with a conductivity meter MC 226 from Mettler Toledo<sup>®</sup> with a relative accuracy of 1%.

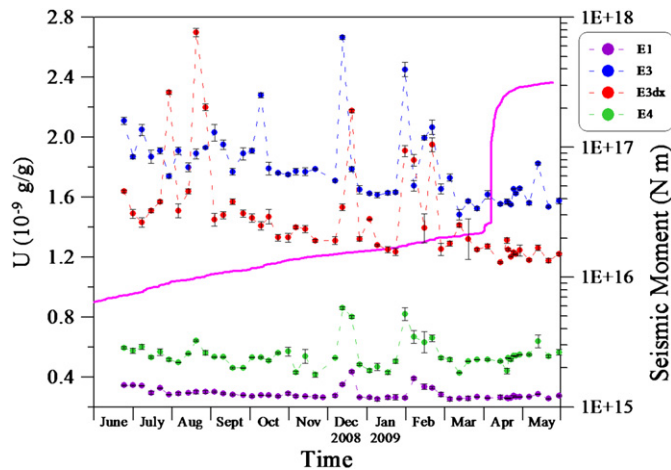
The flow rate measurements of the Traforo spring, located under the tunnel close to the monitoring area, and which collects percolation water from the LNGS-INFN and the highway tunnel (Fig. 1b and 2), were performed by a Venturi flow meter with precision of  $\pm 30$  L/s (one standard deviation). This monitoring is managed by Ruzzo Waterwork Society.

### 2.2. Sample treatment and multi-element determination by ICP-MS

The water samples were diluted 10 times and acidified with 2.5% of nitric acid to stabilize traces in the sample. Reagents of trace analysis grade ( $\text{HNO}_3$  super pure by



**Fig. 3.** a – Seismic activity from the main shock on 6th April, 2009 to 31st May, 2009. The focal mechanism of the main shock and the two strongest aftershocks of  $M_w = 5.6$  (7th April, 2009) and  $M_w = 5.4$  (9th April, 2009) are shown. b – Seismic activity from 1st January 2008 to 5th April 2009. The seismic data are from the Italian Seismic Bulletin, Istituto Nazionale di Geofisica e Vulcanologia.



**Fig. 4.** U concentration of groundwater sampled at E1, E3, E3dx and E4 sites from June 2008 to May 2009, and the cumulative seismic moment from January 2008 to May 2009 in the region of 42.00–42.75 N and 12.75–13.75 E (see Fig. 3a and b). The cumulative seismic moment has been estimated from Italian Seismicity Catalogue (CSI) (Chiarabba et al., 2005; Del Pinto, 2005);  $\ln M_0 = 1.85 M_l + 27.2$ , where  $M_0$  is the seismic moment and  $M_l$  is the local magnitude. The jump-like in the cumulative seismic moment indicates the main shock on 6th April, 2009. The seismic data are from the Italian Seismic Bulletin, Istituto Nazionale di Geofisica e Vulcanologia.

Carlo Erba® Reagenti), ultra-pure water (produced by Millipore MilliQ®-Element), plastic containers and ancillary equipment were used during preparation of water samples because long-lived U radioisotopes at trace levels had to be measured. The ICP-MS measurements were carried out using a quadrupole mass spectrometer from Agilent® Technologies, model 7500a. The tuning of the instrument was optimized in order to reach high sensitivity, stable signal, and low background. A Babbington nebulizer was installed as well. The concentration values were determined in quantitative mode using an external calibration curve, because the matrix effect of the ten times diluted groundwater samples is 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. This solution was added on line using the third line of the peristaltic pump. The calibration curve of U response was corrected using  $^{209}\text{Bi}$ . The accuracy reached in this way generally was better than 5%, which is reasonable for the ICP-MS technique.

### 3. Results and discussion

The area was under investigation from November 2008 to May 2009, a time marked by a seismic swarm with the main shock occurring at 01:33 UT on April 6th, 2009 (moment magnitude,  $M_w = 6.3$ ), located about 18 km far from LNGS-INFN (Fig. 3). In the period 2002–2007, the local seismicity was widespread in the NW sector of the epicentral area of the L'Aquila earthquake with depths ranging from 10 to 20 km and magnitudes from 0.8 to 3.5. The L'Aquila sequence started in November 2008 close to the location of the main event of 6th April, 2009. The depths were between 8 and 12 km and the magnitudes were up to 4.1 (less than 2000 events). After 6th April, 2009, the seismicity continued, migrating to the SE and North sectors with 2 aftershocks of  $M_w = 5.6$  (7th April, 2009) and  $M_w = 5.4$  (9th April, 2009). Until 31st May 2009 more than 6500 events with magnitudes greater than 1.0 with depths between 6 and 20 km were recorded in the region of 42.00–42.75 N and 12.75–13.75 E.

Within the framework of the INFN scientific program ERMES (Environmental Radioactivity Monitoring for Earth Sciences), radon (Plastino and Bella, 2001; Plastino, 2006), radiocarbon (Plastino et al., 2001) and tritium (Plastino et al., 2007) were monitored in groundwater inside the LNGS-INFN, showing different chemical, physical and fluid-dynamic characteristics of groundwater. The U groundwater monitoring was carried out from June 2008 with the aim to better define the radon groundwater transport processes through the overthrust fault (Plastino and Bella, 2001; Plastino, 2006), as well as to check its contribution to the neutron background at the LNGS-INFN (Plastino et al., 2009). The U groundwater results obtained between June 2008 and May 2009 (Fig. 4) showed two different water groups, which were also supported by stable isotope ( $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$ ),  $^{14}\text{C}$  and  $^3\text{H}$  analyses (Rozanski, private communication). While the second group (sites E3 and E3dx, Fig. 2) showed clear U variations (correlated at both sites) with slightly depleted stable isotopes in water (about 0.25‰ in  $\delta^{18}\text{O}$  and 2.1‰ in  $\delta^2\text{H}$ ), the first group (sites E1 and E4, Fig. 2) showed much lower U levels, with a weaker correlation between the sites (Table 1). This indicates that the U groundwater changes strongly depended on the position of monitoring stations and the overthrust fault (Fig. 2).

The highest  $^{14}\text{C}$  concentration was found at site E4 (71.7 pMC (percent modern carbon)) and the lowest at site E3 (57.1 pMC), documenting a longer groundwater–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 transit times of groundwater of the order of some decades.

The observed U variations (Fig. 4) were indicative of a seasonal trend of the water table in the Gran Sasso aquifer due to its recharge modulated by percolation effect related to snow melting during summer period, and some spike-like anomalies not related to hydrological pattern. These U anomalies in groundwater were detected until beginning of March, 2009 about one month before the L'Aquila earthquake of 6th April, 2009. However, during the main shock and aftershocks the variations in the U content were small. Groundwater pH (Fig. 5), ORP (Fig. 6), and electrical conductivity (Fig. 7) did not support the observed U anomalies (Langmuir, 1980; Murphy and Shock, 1999).

The flow rate observed at the Traforo spring located in the tunnel close to the monitoring area (Fig. 8), which collected percolation water from the LNGS-INFN and the highway tunnel, showed a seasonal trend due to snow melting during summer period. This trend was modified, however, from November 2008 with starting of the seismic swarm located about 20 km far from the spring, and drastically during the main shock on 6th April, 2009 with a jump-like anomaly of about 120 L/s. This behaviour emphasized hydrological links between the Gran Sasso aquifer and seismic activities before, during and after the main shock.

The U anomalies observed at site E4 could be explained with the seismic swarm activities, which trigger diffusion processes through the overthrust fault (Plastino, 2006). A correlation between the shear strain in cataclastic rocks and the U content of groundwater was investigated, suggesting a progressive increase in U enrichment with deformation (McCaig, 1989). This behaviour was detected in thermal waters and environments characterized by U

**Table 1**  
Mean values of U, tritium, radiocarbon,  $\delta^{13}\text{C}$ ,  $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$ , pH, ORP, and EC (electrical conductivity) of groundwater sampled at E1, E3, E3dx, and E4 from June 2008 to May 2009.

	U $10^{-9}$ g/g	$^3\text{H}$ TU	$^{14}\text{C}$ pMC	$\delta^{13}\text{C}$ ‰	$\delta^2\text{H}$ ‰	$\delta^{18}\text{O}$ ‰	pH	ORP/mV	EC $\mu\text{S}/\text{cm}$
E1	$0.29 \pm 0.01$	$6.6 \pm 0.4$	$59.5 \pm 1.0$	−9.64	−72.2	−10.93	$8.2 \pm 0.1$	$231 \pm 22$	$255.4 \pm 3.0$
E3	$1.79 \pm 0.02$	$8.8 \pm 0.5$	$57.1 \pm 1.0$	−6.68	−74.6	−11.28	$8.2 \pm 0.1$	$232 \pm 22$	$169.3 \pm 2.0$
E3dx	$1.47 \pm 0.02$	$11.2 \pm 0.6$			−74.4	−11.22	$8.3 \pm 0.1$	$228 \pm 22$	$169.1 \pm 2.0$
E4	$0.54 \pm 0.01$	$10.1 \pm 0.6$	$71.7 \pm 1.0$	−5.74	−72.6	−11.07	$8.2 \pm 0.1$	$240 \pm 22$	$159.1 \pm 2.0$

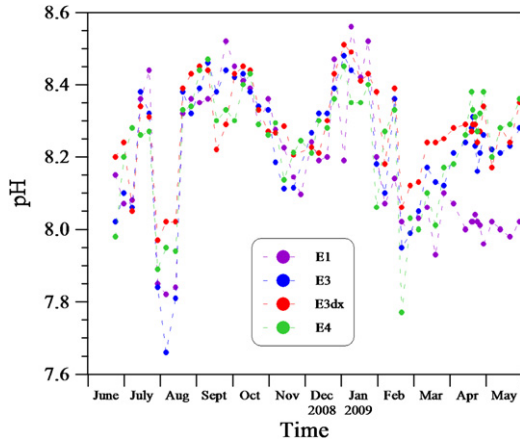


Fig. 5. pH trend of groundwater sampled at E1, E3, E3dx, and E4 sites from June 2008 to May 2009.

concentrations of rocks of several milligram per kilogram. The Gran Sasso karst aquifer, however, does not have similar characteristics, and the U content of the rock is very low, ranging from  $0.42 \pm 0.10$  to  $6.80 \pm 0.67 \times 10^{-6}$  g/g (Esposito and Pelliccioni, 1985; Wulandari et al., 2004).

The fluids are crucial for the mechanics of faults (Sibson, 1992), as they play both passive and active roles during deformations. Variations in the fluid fluxes were observed during seismic sequences, likely associated with variations in the state of stress of the crust. Moreover fluids (CO<sub>2</sub>) can increase pore pressure, and thus trigger a fault rupture along asperities (Miller et al., 2004). The crustal rupture may follow the fault-valve behaviour (Sibson, 1992), with cyclic changes in the transit, accumulation and release of fluids. As fluids decrease the friction on the fault plane, their variation in time could control the episodic activity of faults.

The presence of a locked normal fault in the brittle upper crust does not inhibit an active shearing at depth in the viscous regime, as shown by the steady state strain rate measured by Global Positioning System (GPS) (Kreemer et al., 2002). During the interseismic period, the stick-slip behaviour of the upper crust may correspond to an active stationary shear in the lower

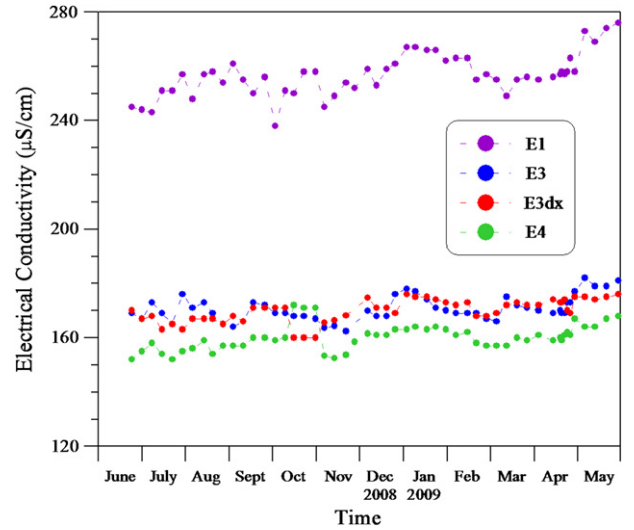


Fig. 7. Electrical conductivity trend of groundwater sampled at E1, E3, E3dx, and E4 sites from June 2008 to May 2009.

ductile crust. The deep strain rate implies an area beneath the locked fault plane where the crust is stretched and the consequent fracturing is expected to be filled by fluids. When the shear stress acting on the locked area of the fault reaches the threshold determining the rupture, the hanging wall of the normal fault starts moving downward. This initial movement could be represented by foreshocks and an increase in fluid pressure. The fluids that filled the cracks are then squeezed out by closure of previously generated vacuum. During the coseismic evolution, the hanging wall may move, squeezing out again the remaining fluids, which had filled the dilated volume during the interseismic period. Long-lasting aftershocks are interpreted as primarily related to the gradual settlement of the hanging wall to the new lower position, and distributed all over the volume within and above the previously dilated rocks. The recent L'Aquila earthquake sequence may be explained using this hypothesis as the area is cross-cut by several NW-SE trending active normal faults (Vezzani and Ghisetti, 1998).

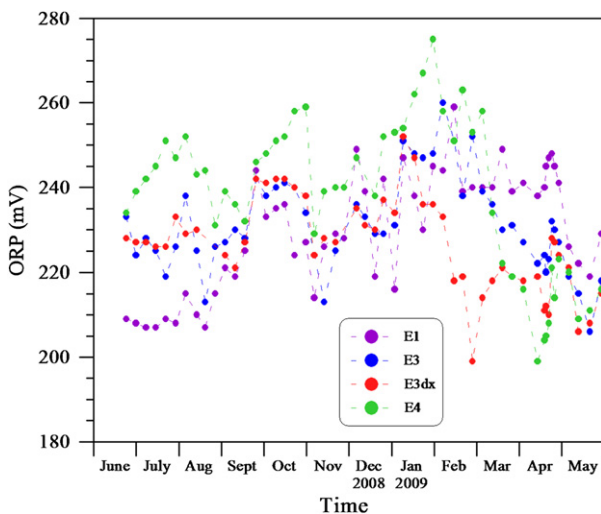


Fig. 6. ORP trend of groundwater sampled at E1, E3, E3dx, and E4 sites from June 2008 to May 2009.

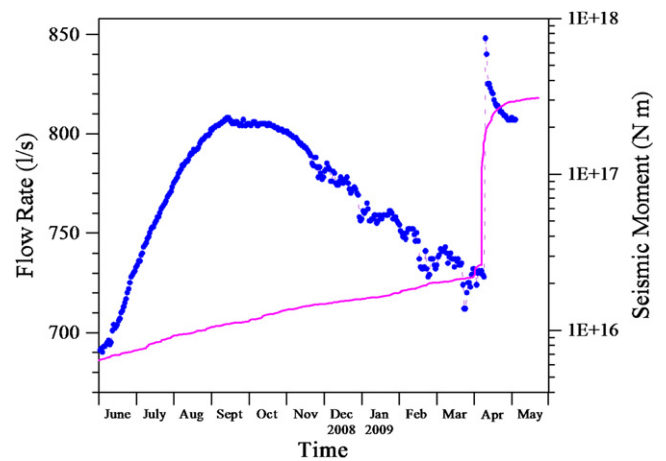


Fig. 8. Flow rate observed at the Traforo spring, and the cumulative seismic moment from January 2008 to May 2009 (see Fig. 3a and b). The jump-like in the cumulative seismic moment indicates the main shock on 6th April, 2009. The Traforo spring is located under the tunnel close to the monitoring area, which collects percolation water from the LNGS-INFN and the highway tunnel. The flow rate data are from the Ruzzo Waterwork Society.

#### 4. Conclusions

The U groundwater anomalies observed before the seismic swarm and the main shock, which occurred on 6th April, 2009 in L'Aquila, were probably associated with geodynamic processes occurring before the earthquake, which triggered diffusion processes through the overthrust fault. This would indicate that more attention should be devoted to the pre-earthquake studies of geodynamic processes, especially on characteristics of fluids filling the fractures before the main shock. Uranium in groundwater can be used therefore as a potential indicator of pre-earthquake processes as it may be associated with geodynamics of preparation phases of earthquakes. Moreover, another possible physical process during the pre- and post-phases of the earthquake could be investigated: the first stage seems to be characterized by U variations in groundwater that can modulate the radon concentration, the second one (after the main shock) do not show any U anomalies, justifying the different radon patterns before and after the main shock.

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#### References

- Catalano, P.G., Cavinato, G., Salvini, F., Tozzi, M., 1986. Analisi strutturale nei Laboratori dell'INFN del Gran Sasso d'Italia. *Memorie Società Geologica Italiana* 35, 647–655.
- Chiarabba, C., Jovane, L., Di Stefano, R., 2005. A new view of Italian seismicity using 20 years of instrumental recordings. *Tectonophysics* 395, 251–268.
- Del Pinto, C., 2005. Ground motion scaling in the Central Apennines. PhD Thesis, Department of Physics, University of L'Aquila, pp. 1–126.
- Esposito, A., Pelliccioni, M., 1985. Study on natural radioactivity in building materials with a view to the construction of the INFN Laboratory of Gran Sasso. *Nuclear Sciences Journal* 22, 291–295.
- Finkel, R.C., 1981. Uranium concentrations and  $^{234}\text{U}/^{238}\text{U}$  activity ratios in fault-associated groundwater as possible earthquake precursors. *Geophysical Research Letters* 8, 453–456.
- Gascoyne, M., 1992. Geochemistry of the actinides and their daughters. In: Ivanovich, M., Harmon, R.S. (Eds.), *Uranium-Series Disequilibrium: Applications to Earth, Marine, and Environmental Science*. Clarendon Press, Oxford, pp. 34–61.
- Geller, R.J., Jackson, D.D., Kagan, Y.Y., Mulargia, F., 1997. Earthquakes cannot be predicted. *Science* 275, 1616–1617.
- Giammar, D., 2001. Geochemistry of uranium at mineral-water interfaces: rates of sorption-desorption and dissolution-precipitation reactions, PhD Thesis, California Institute of Technology, pp. 1–277.
- Gorbushina, L.V., Tyminskiz, V.G., Spiridinov, A.I., 1973. Significance of radiohydrogeological anomalies in a seismically active area for predicting earthquakes. *International Geology Review* 15, 380–384.
- Hauksson, E., 1981. Radon content of groundwater as an earthquake precursor: evaluation of world-wide data and physical basis. *Journal of Geophysical Research* 86, 9397–9410.
- Igarashi, G., Saeki, S., Takahata, N., Sumikawa, K., Tasaka, S., Sasaki, Y., Takahashi, M., Sano, Y., 1995. Ground-water radon anomaly before the Kobe earthquake in Japan. *Science* 269, 60–61.
- Kawada, Y., Nagahama, H., Omori, Y., Yasuoka, Y., Ishikawa, T., Tokonami, S., Shinogi, M., 2007. Time-scale invariant changes in atmospheric radon concentration and crustal strain prior to a large earthquake. *Nonlinear Processes in Geophysics* 14, 123–130.
- Keilis-Borok, V.I., Soloviev, A. (Eds.), 2003. *Nonlinear Dynamics of the Lithosphere and Earthquake Prediction*. Springer-Verlag, Berlin-Heidelberg.
- King, C.Y., Koizumi, N., Kitagawa, Y., 1995. Hydrogeochemical anomalies and the 1995 Kobe earthquake. *Science* 269, 38–39.
- Kreemer, C., Holt, W.E., Haines, A.J., 2002. The global moment rate distribution within plate boundary zones. *Geodynamics Series* 30, 173–189.
- Kuleff, I., Petrov, P., Kostadinov, K., 1980. Uranium content variations in thermal waters from the west Rhodope crystalline massif during earthquakes in the Chepino valley (south Bulgaria). *Journal of Radioanalytical Chemistry* 58, 267–274.
- Langmuir, D., 1980. Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits. *Geochimica et Cosmochimica Acta* 42, 547–569.
- McCaig, A., 1989. Fluid flow through fault zones. *Nature* 340, 600.
- McKinley, J.P., Zachara, J.M., Smith, S.C., Turner, G.D., 1995. The influence of uranyl hydrolysis and multiple site-binding reactions on adsorption of U(VI) to montmorillonite. *Clays and Clay Minerals* 43, 586–598.
- Miller, S., Collettini, C., Chiaraluce, L., Cocco, M., Barchi, M.R., Kohl, T., 2004. After-shock driven by a high pressure CO<sub>2</sub> source at depth. *Nature* 427, 724–727.
- Monnin, M.M., Seidel, J.L., 1992. Radon in soil and in groundwater related to major geophysical events: a survey. *Nuclear Instruments and Methods in Physics Research A* 314, 316–330.
- Murphy, W.M., Shock, E.L., 1999. Environmental aqueous geochemistry of actinides. In: Burns, P.C., Finch, R. (Eds.), *Uranium: Mineralogy, Geochemistry and the Environment*. Mineralogical Society of America, Chantilly, pp. 221–254.
- Nature debates. [http://www.nature.com/nature/debates/earthquake/quake\\_frameset.html](http://www.nature.com/nature/debates/earthquake/quake_frameset.html), 1999.
- Peresan, A., Kossobokov, V., Romashkova, L., Panza, G.F., 2005. Intermediate-term middle-range earthquake predictions in Italy: a review. *Earth-Science Reviews* 69, 97–132.
- Plastino, W., 2006. Monitoring of geochemical and geophysical parameters in the Gran Sasso aquifer. In: Povinec, P.P., Sanchez-Cabeza, J.A. (Eds.), *Radionuclides in the Environment*. Elsevier, Oxford, pp. 335–342.
- Plastino, W., Bella, F., 2001. Radon groundwater monitoring at underground laboratories of Gran Sasso (Italy). *Geophysical Research Letters* 28, 2675–2678.
- Plastino, W., Kaihola, L., Bartolomei, P., Bella, F., 2001. Cosmic background reduction in the radiocarbon measurements by liquid scintillation spectrometry at the underground laboratory of Gran Sasso. *Radiocarbon* 43, 157–161.
- Plastino, W., De Felice, P., De Notaristefani, F., 2002. Radon gamma-ray spectrometry with YAP: Ce scintillator. *Nuclear Instruments and Methods in Physics Research A* 486, 146–149.
- Plastino, W., Chereji, I., Cuna, S., De Felice, P., Kaihola, L., Lupsa, N., Balas, G., Mirel, V., Berdea, P., Baciuc, C., 2007. Tritium in water electrolytic enrichment and liquid scintillation counting. *Radiation Measurements* 42, 68–73.
- Plastino, W., Nisi, S., De Luca, G., Balata, M., Laubenstein, M., Bella, F., 2009. Environmental radioactivity in the ground water at the Gran Sasso National Laboratory (Italy): a possible contribution to the variation of the neutron flux background. *Journal of Radioanalytical and Nuclear Chemistry*. doi:10.1007/s10967-009-0151-2.
- Richon, P., Sabroux, J.C., Halbwegs, M., Vandemeulebrouck, J., Poussielgue, N., Tabbagh, J., Punongbayan, R., 2003. Radon anomaly in the soil of Taal volcano, the Philippines: a likely precursor of the M 7.1 Mindoro earthquake (1994). *Geophysical Research Letters* 30. doi:10.1029/2003GL016902.
- Roeloffs, E., 1999. Radon and rock deformation. *Nature* 399, 104–105.
- Rozanski, K., Private communication.
- Scholz, C.H., Sykes, L.R., Aggarwal, Y.P., 1973. Earthquake prediction: a physical basis. *Science* 181, 803–810.
- Shapiro, M.H., Rice, A., Mendenhall, M.H., Melvin, D., Tombrello, T.A., 1985. Recognition of environmentally cause variations in radon time series. *Pure and Applied Geophysics* 122, 309–326.
- Sibson, R.H., 1992. Implications of fault-valve behaviour for rupture nucleation and recurrence. *Tectonophysics* 211, 283–293.
- Torgersen, T., Beniot, J., Mackie, D., 1990. Controls on groundwater Rn-222 concentrations in fracture rock. *Geophysical Research Letters* 17, 845–848.
- Trique, M., Richon, P., Perrier, F., Avouac, J.P., Sabroux, J.C., 1999. Radon emanation and electric potential variations associated with transient deformation near reservoir lakes. *Nature* 399, 137–141.
- Turner, G.D., Zachara, J.M., McKinley, J.P., Smith, S.C., 1996. Surface-charge properties and UO<sub>2</sub><sup>2+</sup> adsorption of a subsurface smectite. *Geochimica et Cosmochimica Acta* 60, 3399–3414.
- Ulomov, V.I., Mavashev, B.Z., 1967. A precursor of a strong tectonic earthquake. *Doklady Akademii Sciences SSSR, Earth Sciences Sections* 176, 9–11.
- Vezzani, L., Ghisetti, F., 1998. *Carta Geologica dell'Abruzzo, Scala 1:100.000*. Selca, Firenze-Italy.
- Virk, H.S., Singh, B., 1994. Radon recording of Uttarkashi earthquake. *Geophysical Research Letters* 21, 737–740.
- Wakita, H., Nakamura, Y., Notsu, K., Noguchi, M., Asada, T., 1980. Radon anomaly: a possible precursor of the 1978 Izu-Oshima-kinkai earthquake. *Science* 207, 882–883.
- Wakita, H., Igarashi, G., Nakamura, Y., Sano, Y., Notsu, K., 1989. Coseismic radon changes in groundwater. *Geophysical Research Letters* 16, 417–420.
- Wulandari, H., Jochum, J., Rau, W., von Feilitzsch, F., 2004. Neutron flux at the Gran Sasso underground laboratory revisited. *Astroparticle Physics* 22, 313–322.
- Wyss, M., 1997. Cannot earthquakes be predicted? *Science* 278, 487–488.
- Zachara, J.M., McKinley, J.P., 1993. Influence of hydrolysis on the sorption of metal cations by smectites: importance of edge coordination reactions. *Aquatic Sciences* 55, 250–261.