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Uranium time series analysis: A new methodological approach for event screening categorisation

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ABSTRACT

Uranium (U) groundwater anomalies, detected before the L'Aquila earthquake (April 6th, 2009), represent a key geochemical signal of a progressive increase of deep fluids fluxes at middle–lower crustal levels associated with the geodynamics of the earthquake. Although the analyses performed in association with the seismic pattern around Gran Sasso National Laboratory and the geophysical and geochemical patterns of the Gran Sasso aquifer supported this hypothesis, a new approach for time series analysis has been developed for event screening categorisation and to highlight U as possible strain meter in geodynamical processes, particularly those which characterise active normal faulting.

1. Introduction

Radon (Rn) has been investigated as a possible strain meter in geodynamical processes and its anomalies have been widely detected (Ulomov and Mavashev, 1967; Scholz et al., 1973; Wakita et al., 1989; Hauksson, 1981; Monnin and Seidel, 1992; Virk and Singh, 1994; Igarashi et al., 1995; King et al., 1995; Plastino and Bella, 2001; Plastino, 2006; Richon et al., 2003; Kawada et al., 2007), but there is no clear evidence that it is really a good earthquake's precursor. The physical processes associated with radon anomalies are based on changes of radon emanation rates due to strain signal near the earthquake's nucleation point. Particularly, its behaviour before, during and after the main shock is unclear, considering the consolidated scheme for radon release due to stress–strain processes in the rock (Plastino et al., 2010).

Although, in order to assess the utility of U isotopes as fluid phase earthquake precursors, U concentrations and $^{234}U/^{238}U$ activity ratios have been monitored in thermal waters (Gorbushina et al., 1973; Kuleff et al., 1980; Finkel, 1981). The data presented in this paper refer to the monitoring at the Gran Sasso National Laboratory of the Italian National Institute of Nuclear Physics, 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). Therefore, uranium has been investigated as a possible strain meter, particularly in those environments characterised by active normal faulting (Plastino et al., 2009, 2010, 2011).

Since June 2008, in the Gran Sasso National Laboratory of the Italian National Institute of Nuclear Physics, uranium has been frequently analysed in groundwater to study the possible pattern for radon sources in groundwater, its contribution to neutron flux background, and the hydrological patterns in the aquifer (Plastino et al., 2009, 2013).

The Gran Sasso National Laboratory of the Italian National Institute of Nuclear Physics 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 flow paths: the first one, where 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).

The water samples were collected weekly in three sites located inside the underground laboratories. Each sample was 1 L, and was stored in cleaned and rinsed polyethylene bottles after five minutes of water flushing at maximum flow (Plastino et al., 2009). 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 (HNO_3 super pure by 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 Inductively Coupled Plasma – Mass Spectrometry (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,

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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}Bi. The accuracy reached in this way generally was better than 5%, which is reasonable for the ICP-MS technique (Plastino et al., 2009).

The Gran Sasso area was marked by the relatively intense seismic activity, which could be mostly clustered between October 2008 and December 2010 culminating in the event of April 6th, 2009 with the moment magnitude of 6.3, and by relatively quiet seismic periods preceding and following it. U anomalies in groundwater were detected until beginning of March 2009 about one month before this earthquake. Those anomalies have been investigated by seismic analysis (Plastino et al., 2010; Ciarletti et al., 2016) as well as by hydrological approach (Plastino et al., 2007, 2013). Furthermore, U and Th decay chain disequilibria in different water environments have been widely used in investigating of mixing problems among different reservoirs (Bourdon et al., 2003; Smith et al., 2012; Copia et al., 2015, 2017). All those analyses provided a key geochemical signal of a progressive increase of deep CO_2 fluxes at middle-lower crustal levels. Repeated sharp U enrichments in groundwater, that can be directly associated with the geodynamics of the earthquake, represent a much more precise strainmeter than Rn, whose presence could be modulated by U content during the preparation phase of the earthquake, and only successively released by microfracturing during the main shock and aftershocks (Plastino et al., 2011).

2. Methods

In order to assess the U anomalies, the used methodology for the time series analysis is introduced here. First, the trend of the time series is estimated by a linear fit. Then, following Bianchi et al. (2018), the Generalised Lomb-Scargle (GLS) periodogram (Lomb, 1976; Zeichmeister and Kürster, 2009)

$$P(\omega) = \frac{1}{2\sigma^2}\left\{\frac{N\left[\sum_j (X_j - X)\cos(\omega(t_j - \tau))\right]^2}{N\sum_j \cos^2(\omega(t_j - \tau)) - \left[\sum_j \cos(\omega(t_j - \tau))\right]^2} + \frac{N\left[\sum_j (X_j - X)\sin(\omega(t_j - \tau))\right]^2}{N\sum_j \sin^2(\omega(t_j - \tau)) - \left[\sum_j \sin(\omega(t_j - \tau))\right]^2}\right\} \tag{1}$$

can be evaluated in order to extract periodic components and quantify their percentage weight with respect to the total time series where N is the length of the time series, $\omega = 2\pi\nu$, σ^2 the variance of the data, X the mean of the data and τ is given by (Zeichmeister and Kürster, 2009; Press and Rybicki, 1989)

$$\tau = \frac{1}{2\omega}\tan^{-1}\left\{\frac{N\sum_j \sin(2\omega t_j) - 2\sum_j \cos(\omega t_j)\sum_j \sin(\omega t_j)}{N\sum_j \cos(2\omega t_j) - \left[\sum_j \cos(\omega t_j)\right]^2 + \left[\sum_j \sin(\omega t_j)\right]^2}\right\} \tag{2}$$

The GLS periodogram can handle the case of missing data without any interpolation required, and it is also associated with a threshold that allows to discriminate peaks with high probability to be associated to a periodic signal. Once peaks corresponding to periodicities have been removed, residual time series can be investigated. First, it is arranged in a histogram, then normalised to zero mean and unit variance. Those values that exceed a certain number of standard deviations are considered as outliers, i.e. anomalous values.

Finally, persistence of the residual time series is inspected via Detrended Fluctuation Analysis (DFA; Peng et al., 1993). The residual time series is first integrated, obtaining a new time series Y_t, and then divided in time intervals of length n. The data in every interval are fitted with a least squares line Y_t^{fit} and the root mean square fluctuation in each interval of length n is computed. Then, all the root mean square fluctuations are averaged, and the whole procedure is repeated for different sizes n of the time interval, obtaining

$$F(n) = \left[\frac{1}{N}\sum_{t=1}^{N}(Y_t - Y_t^{fit})^2\right]^{1/2} \sim n^H \tag{3}$$

with H being the Hurst exponent. Values of H between 0.0 and 0.5 indicate anticorrelation, $H = 0.5$ corresponds to white noise, $0.5 < H < 1.0$ indicates long range correlations, $H = 1.0$ is the equivalent of pink noise (i.e. power spectrum going as the inverse of the frequency), $1.0 < H < 1.5$ indicates stronger long range correlations and the time series is non-stationary, with $H = 1.5$ corresponding to Brownian noise. Also, a local Hurst exponent H_t can be defined (Ihlen, 2012). Generalising Equation (3) to order q,

$$F_q(n) = \left[\frac{1}{N/n}\sum_{s=1}^{N/n}\left[\frac{1}{n}\sum_{t=1}^{n}(Y_t - Y_t^{fit})^2\right]^{q/2}\right]^{1/q} \sim n^{h(q)} \tag{4}$$

with $s = 1, \ldots, N/n$ being the number of interval with length n and $h(2) = H$ being the Hurst exponent. Given N the length of the time series, $F_q(N)$ is common to all the orders q. Taking a small window that runs over the total length of the time series, root mean square fluctuations can be computed in every window in order to obtain the values of the fluctuations as a function of time. Then, these values and the value of $F_q(N)$ can be connected with a straight line to obtain a local Hurst exponent H_t as the slope of the line, i.e. the Hurst exponent for a single time window.

Finally, the Detrended Cross-Correlation Analysis (DCCA; Podobnik and Stanley, 2008; Zebende, 2011) has been applied to find correlations (in terms of scaling behaviour) between beryllium and meteorological parameters. Provided the two series x and y have the same length N, Equation (4) for $q = 2$ and for two different time series is now given by

$$F_{xy}(n) = \left[\frac{1}{N}\sum_{t=1}^{N}(Y_{xt} - Y_{xt}^{fit})(Y_{yt} - Y_{yt}^{fit})\right]^{1/2} \tag{5}$$

Equation (5) is computed for different n, and a cross-correlation coefficient, ranging between -1 and $+1$, can be defined this way,

$$\rho_{DCCA}(n) = \frac{F_{xy}^2(n)}{F_{xx}(n)F_{yy}(n)} \tag{6}$$

The cross-correlation coefficient depends on the time scale n, thus correlations could not be the same at all time scales.

From June 2008 to July 2014, U concentration, electrical conductivity, pH, and the Oxidation-Reduction Potential (ORP) of groundwater have been measured at the Gran Sasso National Laboratory of the National of the Italian Institute of Nuclear Physics (Ciarletti et al., 2016). The U concentration time series at four different sites (E1, E3, E3dx and E4) are studied, and then correlated with the other parameters in order to find possible correlations.

3. Results and discussion

The method of analysis is principally focused on outliers and noise structure. The four U time series are shown in Fig. 1. E1 and E4 exhibit small values of U concentrations, with respect to the other two sites, E3 being the one with the highest concentration. Significant peaks can be noticed around 2009, in correspondence of the L'Aquila earthquake