Radon and helium as pathfinders of fault systems and groundwater evolution in different Italian areas (*)

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Summary. — Groundwater surveys in some Italian areas with different geological and geodynamical features were performed in order to evaluate dissolved rare gases (^{222}Rn, He and Ar) as potential tracers of fault systems and groundwater evolution (fluid origin, water-rock-gas interaction, fluid migration and mixing phenomena). The obtained results showed that the highest values of Rn and He were found along important fault systems cutting the investigated areas, where the fault-related permeability increases deep-seated fluid circulation. However, dissolved-radon anomalies may be due to: a) a rapid ascent of Rn-bearing fluids, equilibrated with lithologies richer in U than outcropping formation; b) a local enrichment of U due to the action of low enthalpy groundwater or strong carrier gas (CO_2) discharges usually occurring along the fault systems.

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1. – Introduction

Noble-gas geochemistry is currently recognised as a reliable tool to investigate the origin and processes involving fluids during their ascent to the surface. A better knowledge of the nature and evolution of deep fluids establishes a fundamental basis for geological studies, including fault tracing. In fact, fault zones are known to act as preferential pathways, that can readily transmit a variety of fluids, including gases, to the surface (*e.g.*, Banwell & Parizek, 1988; Dall'Aglio *et al.*, 1995). Field data have shown the reliability of dissolved-gas surveys for locating brittle deformations of the

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Earth's surface, even in areas where fault-related surface expressions are difficult to recognise by traditional tools (*e.g.*, Lombardi *et al.*, 1997).

Many parameters affect noble-gas content of sampled fluids. In particular the U content in rocks, their porosity or degree of fracturing, water-rock re-equilibration near surface, degassing and mixing processes constrain the concentration in water of some radiogenic species as Rn, ^{4}He, ^{40}Ar. Moreover some different physico-chemical

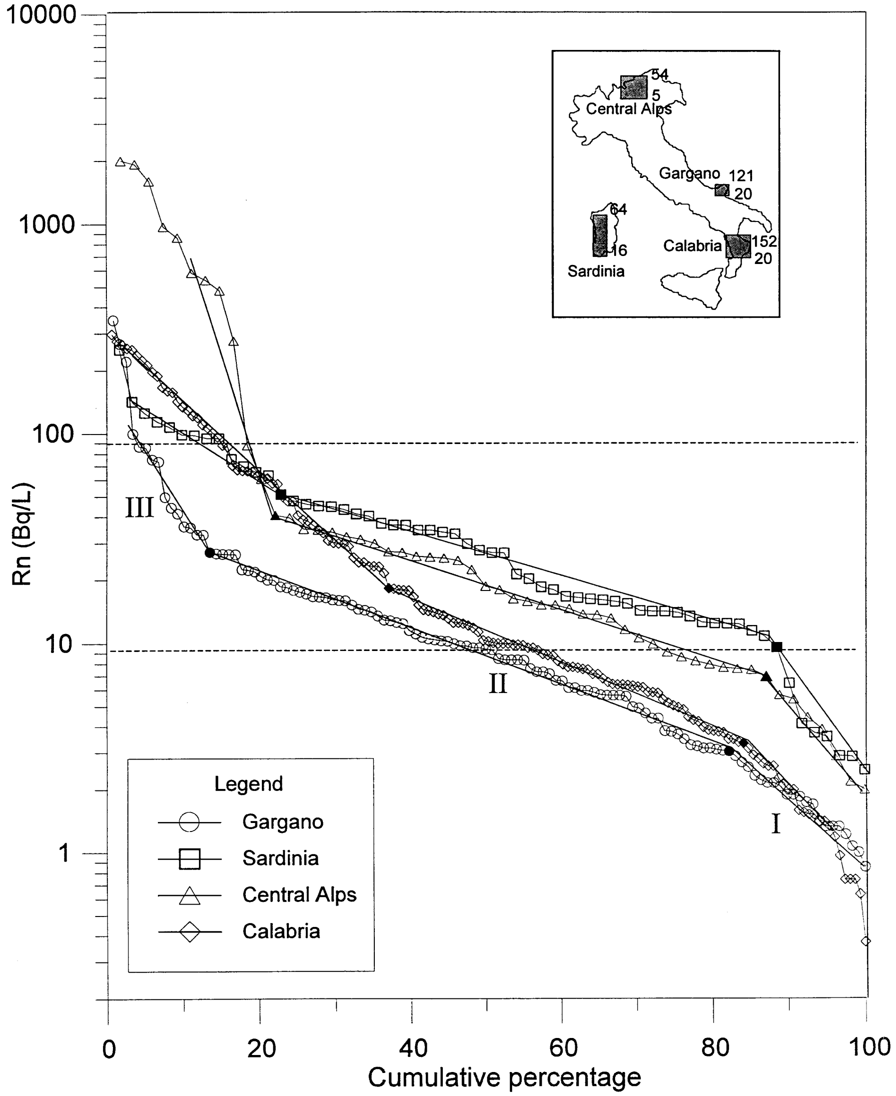

Fig. 1. – Cumulative frequency diagram of radon. Changes of slopes split every plot into three populations. The upper population (III) represents the anomalous values that are related to faulted zones. The intermediate-value population is considered as the background for that area. The dashed lines mark the range of theoretical equilibrium of water with rocks (see text). Aside the location of sampled areas is sketched. For each site the number of samples of Rn (upper) and of other gases (lower) are reported.

characteristics of noble gas such as solubility, stability and fugacity may originate misleading interpretation if a comprehensive examination of a large number of parameters is not properly considered.

This communication summarises the results obtained from an extensive surveying project performed by ING in some Italian areas with different geological (sedimentary and crystalline environments) and geodynamical features (quiescent and high-seismicity areas). Selected areas were (fig. 1): Gargano and Calabria for sedimentary environment, Central Alps and Sardinia for crystalline domains.

2. – Methodology

Sampling was focused as much as possible along the main tectonic discontinuities cutting each area. In addition samples were also collected throughout the areas to provide a *geochemical background*.

Radon concentration in groundwater was determined in the field by α-scintillation method (*EDA Instruments, Toronto, Canada*) after stripping by air bubbling. Dissolved-gas determination was performed in laboratory by coupling an extraction line to a quadrupole gas spectrometer (Lombardi *et al.*, 1997).

3. – Results and discussions

In order to discriminate and evaluate the effect of brittle deformations (faults and fractures) upon dissolved-Rn distribution in different scenarios, a graphical method proposed by Lepeltier (1969) was used. According to this author, slope changes and straight line segments of the cumulative frequency diagram (fig. 1) are helpful for the determination of the background range, the recognition of multiple populations and the identification of anomalous samples. Basically changes of slope split each plot into three straight line segments representing three possible populations. As an example, for data set from Gargano (empty circle) two graphical thresholds can be set at about 2-3 and 25 Bq/L. The population of the lowest Rn values (group I) is thought to result from waters that likely have attained no secular equilibrium with rocks (for ^{222}Rn about 1 month) or that underwent a Rn depletion due to dilution or stripping phenomena. The second group (group II) can be considered as the background population affected by the features (*i.e.*, power emanation, U content) of outcropping and reservoir rocks formations. Finally values greater than 25 Bq/L (group III) would be connected to sectors of enhanced Rn content, mainly linked to the presence of fault or fracture zones and to changes of lithology. Data from Sardinia and Central Alps (empty square and triangle, respectively), that were collected in crystalline domains (intrusive rocks), show a slight higher background ranges than those from sedimentary domains (carbonate in Gargano as well as carbonate and clays in Calabria). For crystalline domains, the upper threshold could be set at approximately 40-50 Bq/L, as a consequence of higher U content of rocks forming the crystalline basement (Carrara *et al.*, 1974; Dall'Aglio *et al.*, 1974).

Graphical thresholds obtained from cumulative frequency diagrams may be compared with theoretical values computed according to the formula (modified after

Wanty *et al.*, 1992), that gives the generation of radon in rocks and transfer to groundwater:

$$Rn_{\mathrm{w}} = 0.0125 \times \varrho \times \left(\frac{1-n}{n} \right) \times E \times [U]_{\mathrm{R}},$$

where Rn_{w} is radon concentration in water (Bq/L); ϱ is the bulk density (kg/m^3); n and E the rock porosity and the emanation power, respectively (dimensionless); $[U]$ is the uranium content, expressed as ppm by mass of rock. Since measurements of ϱ, n, E are not available, their values are assumed on the basis of literature data (Porstendorfer, 1993; Schultz *et al.*, 1992). For $E = 0.2 \pm 0.05$, $\varrho = 2300 \pm 200$, $n = 0.25 \pm 0.05$ and U (ppm) = 2 ± 1, a range of Rn values (9–94 Bq/L) is obtained, consistent with the rocks characterising the investigated areas. This computation states that the threshold between background and anomalous population should occur in the range 9–94 Bq/L (although this is too wide due to lack of data), as verified for all our four surveys.

If the anomalous samples are related to their own geological scenarios, the following consideration can be drawn:

– The anomalous samples from Gargano were collected along the *Candelaro Fault*, an important NW-SE transtensive structure bounding the promontory where relatively high values of the ^{3}He/^{4}He ratio (Hooker *et al.*, 1985; B. Marty in *Geochemical Seismic Zonation* (*GSZ*) Project, unpublished data) point out a slight mantle signature, mainly at the *Mt. Granata* sites, at the crossing point with a NE-SW structure.

– The anomalous samples from Calabria were collected along the most important fault systems of the northern and central Calabrian Arc: the *Crati Graben*, the *Sangineto*, *Cetraro-Rossano* and *Marchesato* lines (this last located within the *Crotone Basin*, see the Rn bubble chart in fig. 2). The Sila massif is characterised by a Rn anomaly too, likely as a consequence of prevailing crystalline lithologies (Calcara and Quattrocchi 1993; Calcara *et al.*, 1996).

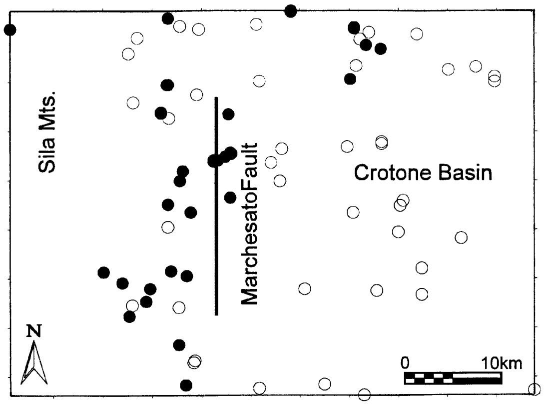

Fig. 2. – Dissolved-radon distribution in the *Crotone Basin*. Rn values exceeding the threshold value (20 Bq/L) obtained from the cumulative frequency distribution (in full) were found along the well-known N-S trending *Marchesato Fault*.

– The anomalous samples from Central-Eastern Alps are related to the *Tonale-Pejo* and *Giudicarie* lines; the greatest values (up to 2000 Bq/L) were found at the intersection of these structures (*Bagni di Mezzo* spring site).

– The anomalous data set from Sardinia is referred to the *Capoterra Fault* zone (fig. 3), located in the southernmost sector of the western *Campidano Graben.* Here brittle deformations cut U-rich granite sandstone (Marcello *et al.*, 1978), the coupling further enhances Rn values.

Additional constraints about the evolution of sampled fluids and their relationship with fault zones are obtained if the relative content of dissolved He, N_2 and Ar is reported in a ternary plot (fig. 4) as proposed by Giggenbach (1991). Data show for each area a common meteoric origin of sampled fluids as suggested by the clustering around the ASW (Air Saturated Water) end-member and in accordance with stable isotope analyses (G. M. Zuppi, personal communication: *GSZ* Project, unpublished data). In addition most of points are aligned along the ASW-He line giving evidence of a progressive water-rock interaction, that supplies He by the decay of radioactive

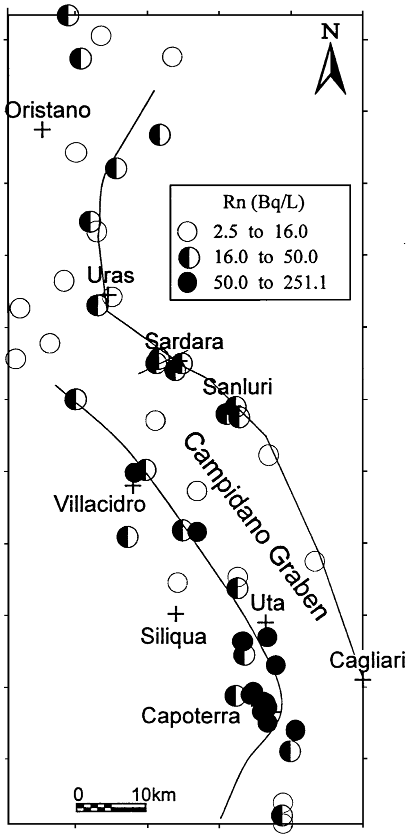

Fig. 3. – Dissolved-radon distribution in the *Campidano Graben.* Also in this case the classes were obtained from the cumulative frequency distribution. The highest values were recorded in the southernmost sector of the western border of the graben (*Capoterra* area). Faults locations (full line) were obtained from Pala *et al.* (1982).

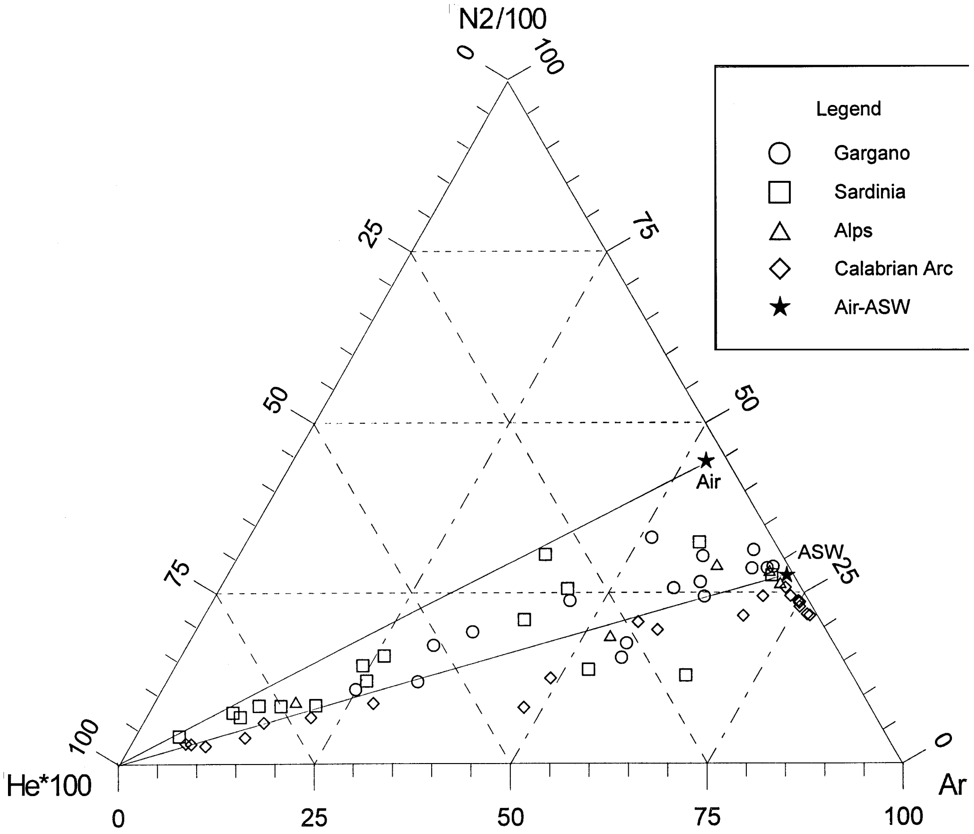

Fig. 4. – Ternary plot of dissolved He, N_2 and Ar. Data show a common meteoric origin of sampled fluids as suggested by the clustering around the ASW (Air Saturated Water) end-member. In addition most of points are aligned along the ASW-He line, giving evidence of a progressive water-rock interaction that supplies He by the decay of radioactive isotopes of the ^{235}U-^{238}U series. This is evidenced for samples collected in areas where crystalline outcrop prevails (Sardinia, Central Alps) or in areas affected by high seismicity that reactivates faults (Calabria).

isotopes of the ^{235}U-^{238}U series. This is evidenced both for samples collected in areas where crystalline outcrop prevails (Sardinia, Central Alps) and in areas affected by active tectonics, recent faulting and high seismicity (Calabria, Gargano), that improves the fault-related permeability (*i.e.*, along the *Candelaro Fault* at the crossing point with a NW-SE line), even if, in Gargano, relative He weighted data do not exceed the 65% of considered gases *vs.* the 90% of the other areas. Deviations from the ASW-He line, *i.e.*, changes of the N_2/Ar ratio, can originate as a consequence of atmospheric contamination due to the excess air (Heaton & Vogel, 1981), or during sampling and analysis procedures as well as during a N_2 production linked to the alteration of organic matter (samples located above the line). For samples with a N_2/Ar ratio lesser than the ASW reference, an overestimation of Ar values at the time of analysis is heavily suspected.

4. – Conclusions

The results obtained in different geologic domains are evocative of the important role played mainly by structural patterns and lithology upon radon distribution in groundwater. Dissolved-radon analysis, based on frequency distribution diagram, has shown for each area the occurrence of three populations representative of the boundary conditions. The population of the lowest Rn values is interpreted as resulting from waters that have attained no secular equilibrium with rocks or that underwent a Rn depletion due to dilution or stripping phenomena. A second threshold divides the background range from the anomalous population connected to areas of enhanced Rn production mainly linked to the presence of fault or fracture systems and to changes of lithology. However, dissolved-radon anomalies may be originated by a rapid ascent of Rn-bearing fluids (*i.e.*, *Candelaro Fault*), equilibrated with lithologies richer in U than outcropping formation; and/or by a local enrichment of U, mostly if coupled with granite sandstone (*i.e.*, *Capoterra* area), enhanced locally by low enthalpy acidic groundwater or by huge flux of carrier gas (CO_2), usually occurring along the fault systems (*i.e.*, *Tonale-Pejo* line). The distribution of radon in groundwater is also affected by the characteristics of the groundwater flow path (*e.g.*, origin of fluids, flow velocity, water-rock interaction, mixing phenomena), that can be recognised and possibly evaluated mainly if considering a large set of variables (*GSZ* unpublished data).

In this research Ar, N_2 and He analysis supported and strengthened the reliability of noble-gas analysis methods for locating faults. This is demonstrated by high values of radiogenic helium (^{4}He) recorded along the faulted areas as a consequence of the deepening of flow path linked to the enhanced fault-related permeability.

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