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Radiation measurements as tool for environmental and geophysics studies on volcano-tectonic areas

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Summary. — In the last years there has been an increasing concern about natural-radioactivity measurements both from the point of view of the environmental survey, especially for the human health protection, and of the geophysical-events investigation in volcanic areas and tectonic fault zones. We report on our activity in both these fields, in particular on the measurements of indoor radon concentration in a long-term passive monitoring in dwellings of the eastern region of Sicily. Because this region is characterized by high seismicity, besides the indoor radioactivity survey, in-soil radon measurements in the region (both volcanic and tectonic area) can provide a better insight and a valuable database for the study related to radon anomalies. A synthesis is reported of the results that we obtained, in the last years, in the volcanic and tectonic area of oriental Sicily both from indoor monitoring and from geophysical-events investigation.

PACS `89.60.-k` – Environmental studies.
PACS `93.85.Np` – Radioactivity methods.
PACS `91.40.Zz` – Volcano monitoring; volcanic hazards and risks.

1. – Introduction

In the last few decades a general social concern about the health risk associated with radon has grown worldwide. It is important to underline that the main source of human exposition to radiation derives from natural radioactivity and that among this last one

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the radon is the principal one, in fact ^{222}Rn and its progeny are responsible for about two-thirds of the exposure of the world population to ionizing radiation from natural sources [1]. It is fairly established that radon when inhaled in large quantity causes lung disease. Therefore measurements of radon activity in dwellings are assuming ever-increasing importance. In many countries all around the world mapping of radon risk areas or radon-prone areas are in progress in accordance to the International Committee on Radiological Protection Recommendations [2]. Keeping this in view, in the framework of an INFN project (named Laborad first and Envirad-Splash after), the indoor radon activity levels were carried out in schools and dwellings by involving directly students from schools in oriental Sicily, with the aim to promote physics too. On the other hand, variations of radon concentrations in soil and fluid samples are considered a useful tool for geodynamical monitoring in active fault zones, for surveillance in volcanic areas and for tracing neotectonic faults. Since the 60s [3], and more actively in the last years, in-soil radon measurements were addressed to in geophysical investigations. Actually, even if many other investigations, modeling and *in situ* experiments in different tectonic areas followed [4-8], the origin and the mechanisms of the radon anomalies and their relationship to geodynamical events (seismic or volcanic ones) are yet poorly understood. Several models have been proposed, Singh [9] and Planinic [10] suggested that the radon anomalies are related to mechanical crack growth in the rocks or to change in flow rate of groundwater. This may allow either an opening of new cracks, a widening or closing of old cracks or a redistribution of opened and closed cracks. The diffusion coefficient of radon in the rocks will then be significantly changed, determining a change in the amount of radon which will escape from the rocks. According to an alternative compression mechanism, proposed by King [9], the anomalous radon concentration may be due to an increase in crustal compression, impending an earthquake, that squeezes out the soil gas into the atmosphere at an increased rate. Studies were also carried out in volcanic areas in various parts of the world. The first evidences of a link between radon concentration and volcanic activity were found in the Karimsky (Russia) [11] and in the Kilauea (Hawaii) [12]. A hypothesis on the possible mechanism could be that, during a volcanic event, an increased heat flow or dry steam discharge would push up the available underground radon, or that the increased in-soil radon activity were due to the collapse of pores volume and the following up-flow of deeper radon-rich ground gas. In order to enlighten on transport mechanism of underground radon in both volcanic and tectonic areas, we carried out long-term radon measurements and we determined *in situ* radon diffusion coefficients in Etnean sites and in lab exhalation rates for rock samples from the same sites.

2. – Environmental monitoring

2·1. *Indoor radon measurements.* – Since 2005 (World Year of Physics) an INFN project started with the aim to divulgate physics informing students about natural radioactivity and its effects on health. The project carries out a strategy to specifically orient young people towards physics and in particular towards the natural-radioactivity problem, by involving them in radon measurements; contemporaneously, as a part of an extensive program, indoor radon activity mapping in dwellings and schools was performed in oriental Sicily. The students participating in the survey received introductory instructions explaining the concern about radon and the survey procedures and nuclear particles detection techniques, so that they acquired more confidence with radioactivity topics. Subsequently, they assembled radon detectors, Nuclear Track Detector CR-39 type ($25 \times 25\,\text{mm}^2$ active area) inside a diffusion chamber, that allows only the ^{222}Rn to

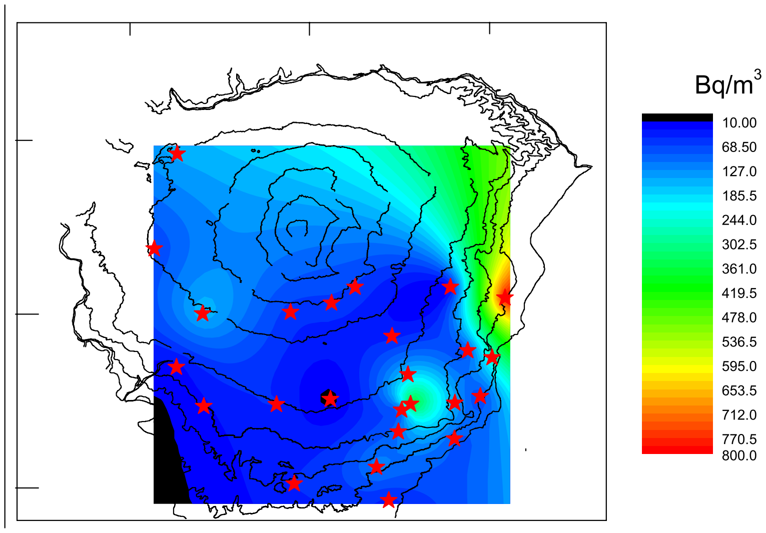

Fig. 1. – Contour plot of indoor radon concentrations in dwellings of the Etnean area.

pass. Subsequently, the students, placed the dosimeters in the classrooms at school and at home. The dwellings were selected randomly so as to cover the whole east and south-east volcanic area of Sicily. After three months of exposure, the CR-39 dosimeters were chemically etched in the laboratory, using a 6.25 M NaOH solution at 98 °C for 1 hour. For the detector reading a semiautomatic system composed of an optical microscope equipped with a CCD camera connected to a PC was realized. A video acquisition software allowed to capture and store the (Field Of View or FOV) images from the microscope. Then the stored images were analyzed by means of ImageJ 1.29× (Image Processing and Analysis in Java) freeware software, in which an opportune routine was edited. This software processes each FOV, discriminating the tracks according to their minor and major axis and their area, and it gives as output both the exposition in Bq h/m^3 and the ^{222}Rn concentration in Bq/m^3, which represents integration over the entire exposition period of the detector. The system was calibrated by exposing the detectors in a radon chamber and an intercalibration with the Radon & Natural Radioactivity Research Laboratory (R & NRRL) University of Dublin was carried out [13]. In the first measurement campaign we collected data from about hundred dwellings in 30 different villages and from twenty schools. The project has allowed the building of a preliminary radon map in the Etnean area (fig. 1). The higher indoor radon concentrations were recorded in zones where faults lie [13].

The distribution of indoor radon concentrations is shown in fig. 2, with an average of 107 Bq/m^3. Only the 8% of measurements of radon concentrations were found (in the Santa Venerina Acireale area) at or above the European recommendation limit [14] of 200 Bq/m^3. The highest indoor concentration of 1325 Bq/m^3 was found in the Guardia Mangano village. Dwellings surveyed for indoor radon can be characterized by referring to their construction materials, construction mode and age of the houses, ventilation conditions, however radon activity has been found prevalently to vary according to the specific geological sites. More extensive measurements are still in progress.

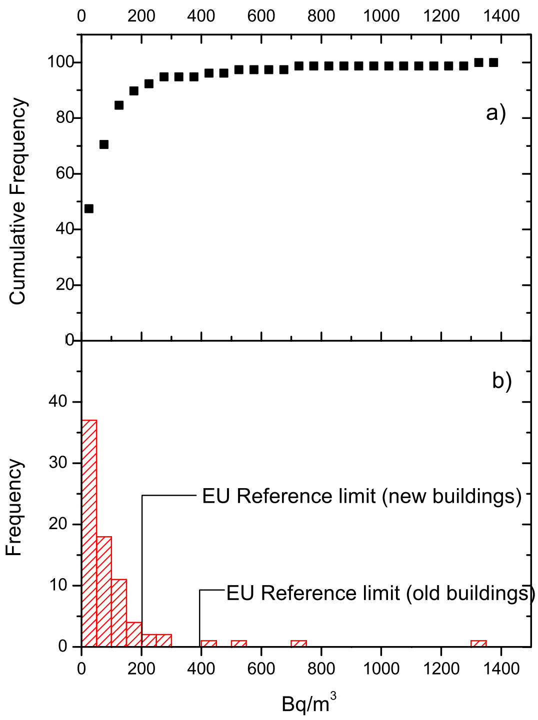

Fig. 2. – a) Cumulative frequency curve and b) frequency hystogram of dwellings with indoor radon concentrations in intervals of Bq/m^3.

2·2. *Radiation measurements in drinking water*. – With the aim of human health protection, measurements of radioactivity in drinking water are widely diffuse, owing to the introduction of radioactivity values as parameters of good quality in the Italian rules (D.Leg.31/2001). We performed measurements in drinking water collected all around the Mt. Etna Volcano [15]. In particular 13 samples were collected on wall, spring and gallery. Measurements of ^{222}Rn and ^{226}Ra were performed by means of liquid scintillation technique after radio-chemical treatment of samples. ^{234}U and ^{238}U measurements were carried out by means of alpha spectrometry on chemical pre-treated samples. Results have shown that: i) all water samples may be classified as low-radon water with activity concentrations up to 12.7 ± 0.6 Bq/l, ii) the activity concentrations of uranium isotopes did not exceed 70 mBq/l, iii) radionuclides are very weakly leached by water from the surrounding reservoir rocks, iv) uranium isotopes constituted the main contribution to the calculated effective doses. Moreover, the obtained values were much below the limit of 100 mSv/year [15]. More recently we have extended water monitoring to tritium amount measurements by means of liquid scintillation technique [16]. Because of the cosmic origin of natural tritium, samples were collected in some superficial drinking water supplies in the Etnean area. All the obtained results show values under the minimum detection limit (3 Bq/l).

3. – Geophysical investigation

In the last years we carried out many investigations on in-soil radioactivity as a tool for geodynamical event studies in Eastern Sicily. We report some of the most significant

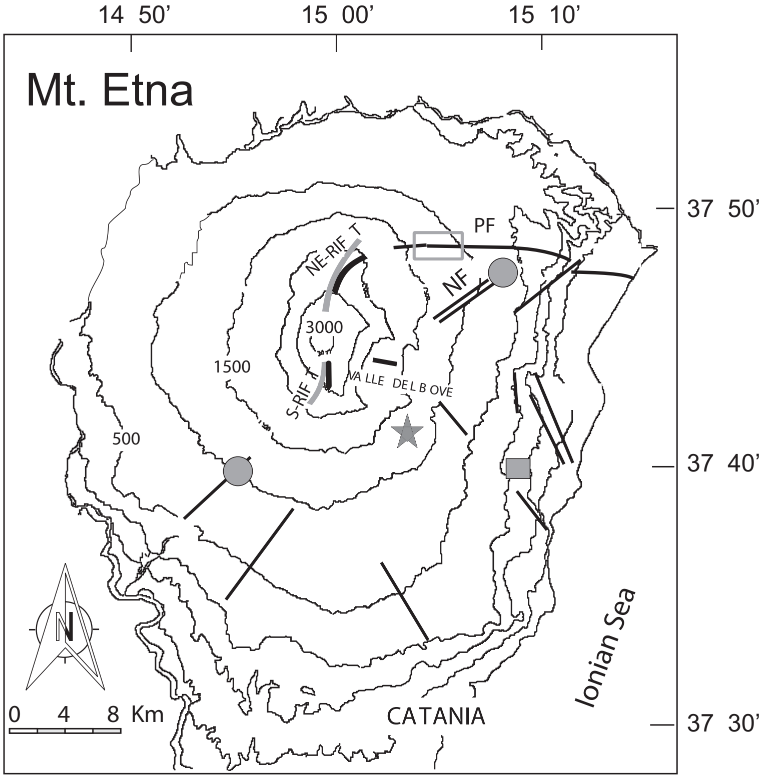

Fig. 3. – Map of Mt. Etna, where the measurement points are evidenced: circles, continuous monitoring in the village of Vena (NE) and Biancavilla (SW); open square, investigation area along the Pernicana faults in which horizontal profiles were determined; star, Cugno di Mezzo Village and square, Santa Venerina Village.

results, in particular those regarding continuous radon measurements in the Etnean area, characterized by volcanic and tectonic events. Different radon measurement techniques were applied.

3˙1. *Continuous in-soil radon measurements.* – Continuous in-soil radon measurements started in 2001 by investigating Etnean area that is a volcanic one characterized by the presence of many tectonic structures too. In particular two sites were chosen near the most active faults, considering that in the Etnean area several fault systems become very active before and sometimes during eruptive events. The most extended among the cropping up structural discontinuities was chosen, and it lies along the NE-SW direction through the volcano. One site (Biancavilla) is in the SW flank, while the other one (Vena) is in the NE flank (circles in fig. 3). In these sites we located two continuous monitoring devices. The in-soil radon measurements were carried out using a portable system that operates with an ionization chamber to detect alpha-particles from radon decay. The system was connected, by means of a pump, to a capillary probe with two filters, one for eliminating the moisture and the other one that allows only ^{222}Rn to pass. The capillary probe is inserted into the soil at one meter depth in order to reduce the influence of the meteorological conditions, that are measured too. Along the entire period of acquisition,

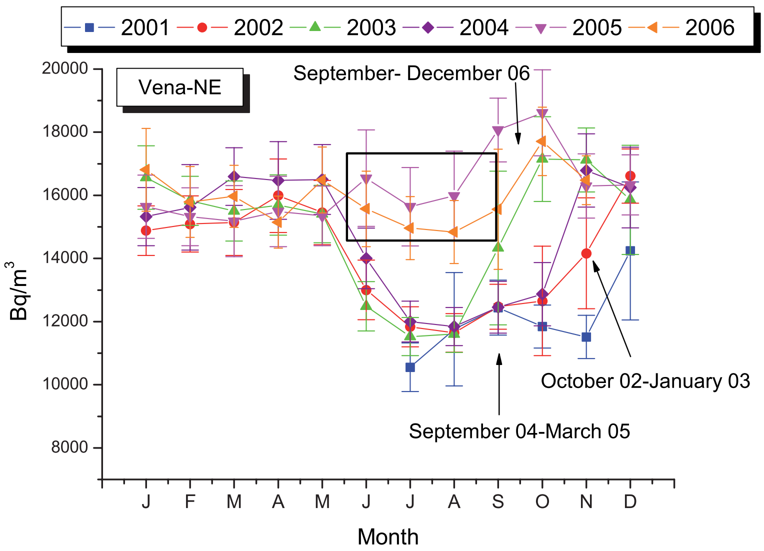

Fig. 4. – Monthly mean values of radon concentration recorded in the Vena site in the 2001 (squares), 2002 (circles), 2003 (up-triangles), 2004 (rhombs), 2005 (down-triangles) and 2006 (left-triangles) years.

the counting time was every ten minutes, with an air flow rate of 0.05 l/min. A seismic station composed by a three directional 1 Hz seismometer connected to a portable digital acquisition data system was also located [17]. The data collected in the two sites, from 2001 till 2004, showed that the two flanks are very different from each other, both for behavior and for concentration absolute values (one order of magnitude higher in the NE flank than in the SW one) [17]. For what concerns a possible correlation with geophysical events, while the SW site shows no significant signals, some evidences were recorded in the NE one [18-22]. A summary of the data continuously recorded at the NE flank, as annual radon concentration trends, is shown in fig. 4, where the main eruptive events are indicated too.

Regarding the 2002 eruption a more detailed analysis [18] has evidenced that the radon concentration values grew up just after the eruption beginning. Moreover, the study of the earthquakes energy release (fig. 5) indicates that the slope of the strain-release curve does not change significantly till 27th October (beginning of the eruption), suggesting that the dynamics of the fractures does not contribute to the radon gas raising. The cumulative curve for the power spectrum, on the contrary, suggests a magma upraising to the upper crustal layers, since July and till the eruption day, preceding a sudden increase of radon concentration. The magma, by raising to the surface, should determine, because of the increasing heat flow, the radon concentration increase that is eased by the fracturations due to a sequence of seismic events that successively occurred. Therefore, we can argue that the radon increase could be linked mainly to the magma uprising [18].

A different behavior was observed during the September 04-March 05 eruption. Higher than expected radon concentration values were recorded at the end of 2003, when no eruption occurred. The analysis of the power spectrum [20] for that period suggested a possible magma rising that did not reach the surface. This magma probably degassed in the shallower layers of the crust and then the eruption of September 2004 followed

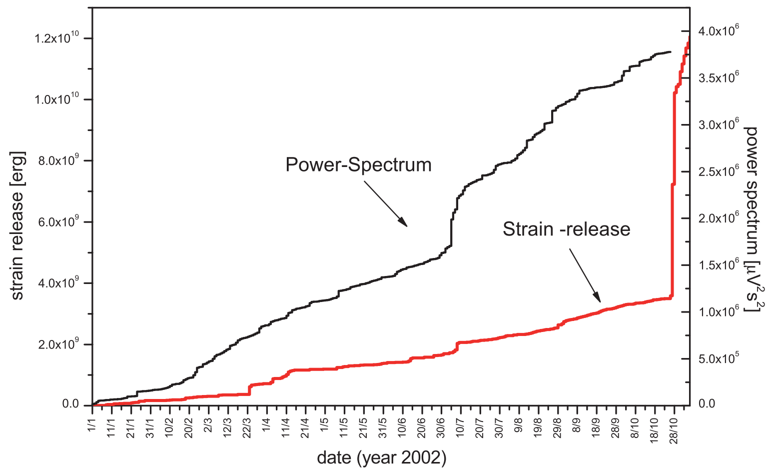

Fig. 5. – Strain-release and power spectrum curves for the 1st January–31st October 2002 period [17].

without signals associated to earthquake swarm and/or increase of volcanic tremor. The last eruption September–December 2006 was preceded by a long period of high radon concentration (squares in fig. 4). The eruption had characteristics similar to those of the 2004 one. Even in this case we can hypothesize that a preliminary high degassing took place before the eruption, that could explain the high in-soil radon concentration.

3·2. *In-soil radon measurements along fault system.* – Since radon data can be suitable to study geogas emission near active faults, measurements were also performed along the very active Pernicana fault (PF in fig. 3). Two different horizontal profiles, orthogonally to the main fault plane, were investigated (open square in fig. 3). The first one was located at 1400 m a.s.l. (AB profile), the second one at 1370 m a.s.l. (CD profile) [23]. Each profile consisted of ten measurement points. Concentrations of ^{222}Rn were obtained by means of three different methodologies: passive, spot and continuous. Passive measurements were performed using nuclear track detectors of CR-39 type, spot and continuous measurements using active portable devices, based on solid-state Si detectors. Continuous ^{222}Rn measurements were carried out very close to some of the AB profile points [23]. Radon data from the two horizontal profiles are reported in fig. 6. The pattern of soil ^{222}Rn values measured in the two profiles is clearly similar: higher values were generally recorded on the up thrown side of the fault and the lowest values occurred generally close to the main fault plane. In the same figure the soil CO_2 efflux values are reported as measured by means of a non-dispersive infrared spectrophotometer. Differently to radon, higher CO_2 emissions were recorded on the fault plane. This behavior can be justified by the in-soil gas transport mechanism. In particular, along the main fault plane, advective transport of deep gases (CO_2, Rn) occurs because of the high ground fracturation and permeability. Near the surface, dilution of radon by CO_2 prevails, thus producing lower radon values.

In fig. 6 data from the different adopted methodologies are compared too. There is a global agreement in the radon measurements, performed along the two profiles, among the data obtained from CR-39 detectors, spot and continuous measurements. This study has evidenced the different performances of the various kinds of detectors for in-soil

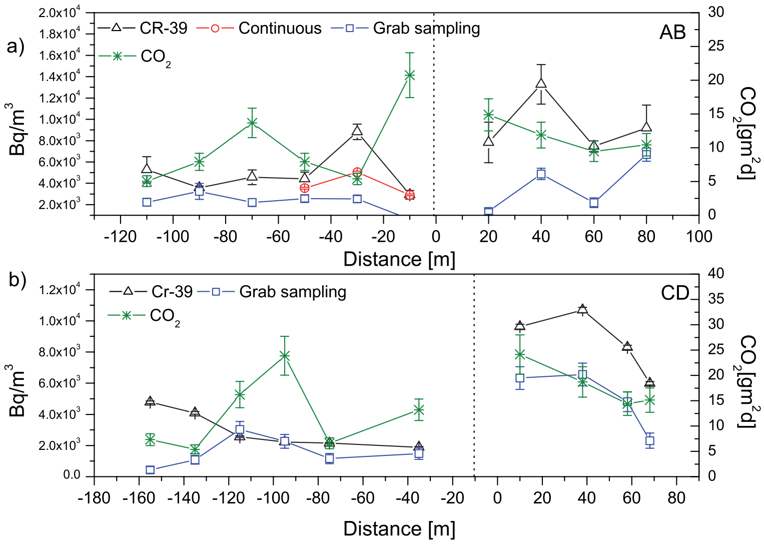

Fig. 6. – In-soil radon horizontal profiles obtained with three techniques: triangles CR-39; squares Grad sampling and circles continuous sampling; and CO_2 profile (stars) for the a) profile AB (1400 m a.s.l.) and b) profile CD (1370 m a.s.l.), along the Pernicana fault.

radioactivity measurements, in particular in the perspective to investigate unknown fault systems. Spot measurements are suitable to identify sites with high radon emission, track detectors, because of their relatively low cost, can be used in a large-scale network of many sites and continuous measurements are suitable for detailed time monitoring.

3˙3. *Measurements of soil physical parameter*. – In order to understand more on the transport mechanism of in-soil radon, some physical parameters were studied in detail, in particular the radon diffusion coefficient and the exhalation rate. Since from continuous measurements the east flank of the volcano seemed more sensible to radon variation, investigation on the soil characteristics were performed in this area. Three sites were chosen on this flank of the volcano, the first one in the same site of the continuous measurements Vena (V), located NE at 825 m a.s.l.; the second one in Cugno di Mezzo (CDM), located E at 1400 m a.s.l. (star in fig. 3); and the last one in the village of Santa Venerina (SV), located SE at 400 m a.s.l. (square in fig. 3). In all of these sites in-soil radon concentration vertical profiles were carried out by means of solid-state nuclear track detectors, CR-39, placed in the soil, at 20 cm intervals, inside a one-meter–depth tube. Vertical profiles of radon concentration are reported in fig. 7 with values of the diffusion coefficient D determined by fitting the acquired data with an exponential trend. The highest diffusion coefficient was found in the site at higher altitude, this could be linked to a major soil faulting, being the submittal area highly involved in the deformation stage during the eruption time.

Moreover measurements of radon exhalation rate E_M were carried out in laboratory using the can technique [24]. Rock samples, 300 g each one, dried at 80 °C, were placed in sealed cylindrical cans (8.6 cm of diameter and 10.5 cm high). A CR-39 detector was fixed on top inside each can and exposed, for three months, to detect alpha-particles from the decay of the exhaled radon from the sample in the remaining volume of the

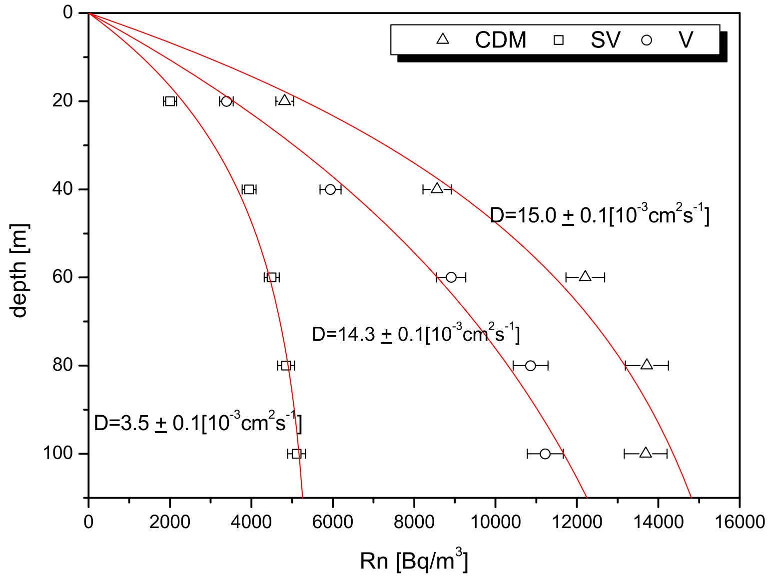

Fig. 7. – In-soil radon vertical profiles in three sites of the Etnean area. In the picture the diffusion coefficients D obtained by fitting the data with an exponential curve are reported.

can. The mass exhalation rate was determined by the following expression [24]:

$$E_M = CVt\lambda/M[t + 1/(\lambda \exp[-\lambda t] - 1)]. \quad (1)$$

Rocks of different types were analyzed, the results are shown in fig. 8. Higher radon exhalation values were recorded for volcanic rocks, according to major uranium amount, as determined by means of a previous gamma-spectroscopy analysis on the same rock samples.

4. – Conclusion

We report a synthesis (for details see refs. [15,17-23]) of our activity carried out in the last few years on the natural radioactivity investigation, both in indoor radon monitoring and in in-soil measurements. The increasing interest on indoor radon, recognized as one of the health hazards, induced us to carry out an extensive indoor radon monitoring by involving students from high schools in an INFN project. The results are reported from a long-term passive monitoring of radon in the indoor environment of the eastern region of Sicily, including volcanic and tectonic zones. For what concerns this analysis, results show in general low indoor radon concentrations, *i.e.* under the European recommendation action limit of 200 Bq/m^3, and only the 8% of the measurements are higher than this limit. The few high concentration spots suggest that geological conditions (faults) in the investigated region, rather than construction materials, may be the determinant factor. Because the monitored region is a zone of high seismicity, the indoor radon measurements of the region can provide insight into and a valuable database for any study related to radon anomalies. Actually we are planning to extend our screening to other villages. We will use nuclear track detectors, that result suitable for large-scale applications, they are

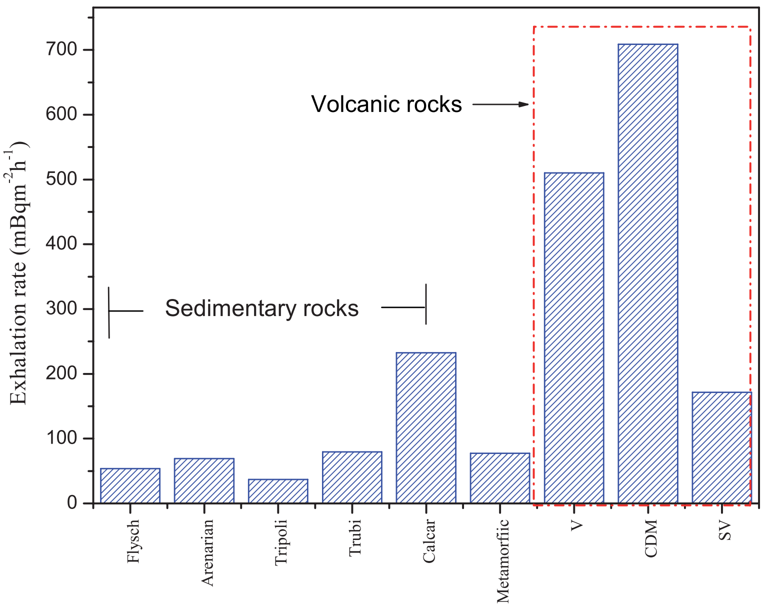

Fig. 8. – Exhalation rates for different rock samples. Higher values were recorded for volcanic rocks collected in the three investigated sites on Mt. Etna.

indeed simple to use, enough accurate, inexpensive and non-invasive. On the other hand, in-soil radon measurements were carried out as a tool to investigate geodynamical events, in particular in the Etnean area characterized by volcanic and tectonic aspects. Previous monitoring of the area evidenced the east flank as the most interesting for this kind of analysis. Detailed temporal data in a site in this flank, ever many years, evidenced some radon anomalies that can be related to magma uprising. Moreover, since the Etnean area is a tectonic area too, active faults are interesting to study. To this purpose in-soil investigations were carried out along the Pernicana fault. Three different methodologies were used to measure in-soil radon, based on both passive and active detection techniques; soil CO_2 efflux was determined too. Along the fault plane in-soil radon concentrations were measured at different distances from the fault plane and were correlated to CO_2 flux values, as a tool to study diffusion process of radon and its role as tracer of geogas. Besides, in order to clarify the radon transport process, we performed *in situ* measurements of concentration vertical profiles to extract diffusion coefficients and in laboratory measurements to determine the exhalation rate for different rock samples, collected in the same sites. In this perspective new methodologies for physical parameter measurements and radon and its progeny detection techniques in volcanic area are in progress at the moment.

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