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Earthquake forecasting: a review of radon as seismic precursor

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ABSTRACT The first measurements of radon as a seismic precursor are dated back to 1927, but the first recording, that is reported in many publications, and that encouraged research on seismic precursors, was detected before the Tashkent earthquake of 1966. This paper is a review of the radon measurements performed all over the world, trying to distinguish between discrete and continuous measurements, and between measurements in soil, water or air. The role that the “precursor radon” had in the forecast of strong earthquakes in the past has been examined. The currently monitored sites in Italy are listed, and some of the results obtained are reported.

Key words: earthquake forecasting, radon anomalies.

1. Introduction

Natural disasters either on a global or local scale, with a great influence on both national economy and social development as well, have become conspicuous problems.

The precursors and prediction words, inevitably, bring the mind to distant times, when people tried to relate various previous observable phenomena to an earthquake. In particular, since the life was preponderantly in the countryside, the observations were about everything related to water and land: for example, changes of the water level in wells, different springs flow, burning and cracking of the land. Today, living in towns, it may be difficult to daily catch these “observable” phenomena; however, today, they may be quantified, and also other parameters, which cannot be detected without instruments, are monitored (Riggio and Santulin, 2012).

Other authors have produced thematic papers on radon as a precursor in the past (Friedmann, 2012; Immè and Morelli, 2012); in this paper we will emphasize the importance of the protocols established for the forecast and the role of radon in the formulation of the forecast.

All kinds of seismic precursors are a unified reaction of the earthquake preparation process, although the observed physical quantity of precursor is different from each other. The process of occurrence and preparation of large earthquakes influences the characteristics of the anomaly, when they are systematically studied, including the trend of the observed anomalies lasting one year or several years in the phase of long-medium term before a strong earthquake occurs, and of the abrupt anomalies observed in the short term and impending phase (Zhang *et al.*, 1996).

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In some works of Chinese researchers of the 1990s, it is already reported the influence of the type of mechanism of the earthquake on the characteristics of the anomaly. They identified two classes of events: one class of earthquakes includes stick-slip earthquakes, occurring on large scale strike-slip faults; the other class includes earthquakes that occur within less fractured structural blocks, which are called rupture earthquakes. They discovered that precursor anomaly of the rupture earthquake is more frequent than that of stick-slip earthquake. It shows that the disparity of destructive types of earthquakes results in the difference of precursor anomalies. The precursor phenomena can be observed also in some stations far away from the epicentre; these anomalies are called far-field precursors and reflect changes of regional stress field. Related to far-field precursor, in the study of earthquake cases, it was discovered that some observational stations in special structural locations could detect precursors more effectively than other stations, and even had distinct precursor response to some earthquakes far away. These observatories are called sensitive spots of precursor observation (Zhang *et al.*, 1996). The relationship between the tectonic structures, the crustal deformation and the physical-chemical characteristics of the circulating fluid is, in many cases, supported by studies and measurements which allow correlations between the causes and the quantitative modification of these parameters. In particular, changes in the fluids chemical composition, which occur as transient phenomena, can indeed provide information about the status of crustal deformation. Moreover, in order to evaluate properly precursory phenomena and to be able to use them confidently for predictive purposes, it is necessary to understand the physical processes that give rise to them. Physical models of precursory phenomena are classified in two broad categories: those based on fault constitutive relations, which predict fault slip behaviour but not the change in the properties of the material surrounding the fault, and those based on bulk rock constitutive relations, which predict physical property changes in a volume surrounding the fault. Nucleation and lithospheric loading models are the most prominent of the first type and the dilatancy model is related to the second type (Immè and Morelli, 2012).

In the 1970s, in China, one forecast led to the evacuation of the area identified as a possible epicentre saving many people. The research on seismic precursors, with the enthusiasm of this success, spread all over the world. Many cases in which there was a relationship between changes in physical and chemical parameters and state of stress in the area under study were recorded *a posteriori* but to formulate a deterministic forecast (location, time and magnitude) of the event that will happen, and ensure the authority to enact an ordinance of evacuation, is much more complex.

The forecasts, in the 1960s and 1970s, were based on abnormalities of many seismic precursors. In order to make an assessment of individual precursors observable according to their relationship with the occurrence of strong earthquakes, it is necessary to determine what was the role of the observable analysed in the context of “predictions” with success or failure and check what were the observables considered in the protocols adopted by the nations where there was a forecast plan and subsequent, if necessary, evacuation.

What happens underground some minutes or several months before an earthquake is unknown exactly and it is difficult to make deterministic predictions. Up to now, there are many difficulties in understanding the physics of earthquakes. There is a combination of factors that does not lead to an accurate deterministic prediction: the deformation is not always followed by a rupture, or, in the observation sites, phenomena linked to the deformation are not recorded.

Many precursors and many physical models to explain the existence of the precursor have been reported in previous reviews. In many cases, there are multiple, competing models to explain the existence of an earthquake precursor, even if there is no current scientific agreement about which model is the best (Cicerone *et al.*, 2009). Most the reported models can be associated with the mechanisms proposed by Thomas (1988). Gas emission models are: physical and/or chemical release by ultrasonic vibration (UV model); chemical release due to pressure sensitive solubility (PSS model); physical release by pore collapse (PC model); chemical release by increased loss or reaction with freshly created rock surfaces (IRSA model); physical mixing due to aquifer breaching and /or fluid mixing [AB/FM model: Cicerone *et al.* (2009)]. None of these models explains, exhaustively, the origin of geochemical precursors. The more corroborated one is IRSA that goes back to the theory of dilatancy. Laboratory tests agree with the field tests and have shown a correlation between the concentration of radon in the ground water and the regional variations of stress and strain (Cicerone *et al.*, 2009).

Other authors have reached the same conclusions. Roeloffs (1988) in its review reported anomalies of flow or pressure of the ground water, fluids or gases that have been interpreted as seismic precursors. These anomalies were detected at a distance of several hundreds of kilometres from the epicentre of the earthquake, with time precursors ranging from less than one day to more than one year. First, techniques for the definition of the anomalies are described. The physical models proposed are, the presence of a volume fractured near the hypocentre, the passage of a propagating deformation front and aseismic slipping of part of the fault plane. Many of the anomalies that have been identified could be explained by at least one of these mechanisms. Roeloffs (2006) analysed the latter model in his most recent work. The crustal deformation is always present, in a constant quantity, and varies before earthquakes. The variations of deformation before earthquakes are, therefore, analysed both in subduction zones and tectonic environments. The variations of deformation, however, vary from place to place and from time scale (one hundred seconds to more than a decade).

Experimental data (about 150 claimed gas precursors proposed in the literature have been reviewed) analysed by Toutain and Baubron (1999) showed that the anomalies occur not only in the vicinity of the epicentral area, but also at distances much larger than the typical lengths, as already noted by various authors (Fleischer, 1981; Hauksson, 1981; King, 1986). To accomplish this, it is necessary to assume that changes in stress or strain are propagated from the forthcoming rupture zone to the radon station (Hauksson and Goddard, 1981).

An analysis of variations in the level of the water before 31 earthquakes of magnitude between 4.5 and 8.0 was investigated by Kissin and Grinevsky (1990). The determination of the reported anomalies is based on a qualitative assessment, but nevertheless further analysis gives a good contribution to the development of related physical models. Changes in the stress-strain state before large earthquakes can occur, not only near the future source, but also far from it. Such changes must be related to the occurrence of unstable situations in a block system (Sadovsky, 1986), and to the passage of stress waves that trigger large faults (Rice, 1980) as well as to some other geodynamic processes.

A further contribution to understand the relationship between the emission of radon and crustal deformation was given by Roeloffs (1999) who analysed the data from a natural laboratory consisting of the reservoir of Roselend (Trique *et al.*, 1999). This large-scale laboratory has highlighted the link between radon emission and crustal deformation and the importance that could have radon as a seismic precursor.

A new Earth-realistic model of upward fluid migration from below the brittle crust in response to strain and the link with the fracturing of the seismogenic crust on long and short time scale was descripted by Stefansson *et al.* (2011). It is a very interesting work that relates the migration of fluids with microseismicity and asperities. They also explained the relationship between volcanic eruptions and earthquakes, and concluded that the movement of plates induces the occurrence of both. This is one of the few large-scale interpretations of seismogenic processes. The authors conclude that the radon measurements are an example of surface processes observed in the crust, which should be complemented by geochemical measurements of magmatic fluids.

The objective of this work is to collect all the information about the role of the measurement of radon in earthquake prediction, distinguishing between the various methods of acquisition. The criteria used to collect them are: the source-site distance and the length of the period analysed.

2. Considerations on radon anomalies

Anomalies have been detected in the signals obtained by passive track detectors, by passive detectors recording in continuous and by active detectors, used for measures both in soil and in water. When radon concentrations are measured in continuous mode for a long time and with a time resolution of at least one hour, it is possible to classify the observed radon anomalies according to different trends. Mainly they show two different shapes. The first type, called type A (Friedmann, 2012), shows a rather slow change of the radon concentration and can continue even over years. The other type (type B), involves anomalies which appear much faster and can be followed by a slow increase or a rather constant radon concentration, or be characterised as a short peak (duration: hours to days) in the radon concentration. These peaks can be either positive or negative and are often followed by an earthquake within about ten days.

The problems related to the identification of anomalies are: a) the definition of the anomaly; b) the identification of the maximum distance between the epicentre of an earthquake and the site where the anomaly of radon is observed; c) the identification of the time between the radon anomaly and the occurrence of an earthquake of a given magnitude (time precursor); d) the importance of the tectonic structure.

The first definition of anomaly was done in a subjective manner, based exclusively on the percentage increase from the background value. The method of the 2 sigma was later introduced: any radon variation that can be considered "significant anomaly" must differ from the mean +/-2 standard deviations, according to Igarashi and Wakita (1990). A correlation between radon emission and barometric pressure should be analyzed before the identification of possible radon anomalies. Other methods are the machine-learning methods, applied to exclude the anomalies generated by meteorological parameters. Particularly, the applications of artificial neural network of regressions and of tree models have proven to be useful means of extracting radon anomalies caused by seismic events (Gregoric *et al.*, 2012).

In a summary of 1999, Toutain and Baubron (1999) analysed 15 cases of geochemical precursors reported in the scientific literature. Anomalies appear at distances sometimes much greater than typical source dimensions, and occur in the field of strain higher than 10^{-9}, most of them being in the field of strain higher than 10^{-8}. Taking into account the very high heterogeneity of such a set of data, they suggest that amplitudes of gas anomalies are independent from both magnitudes and epicentral distances of related earthquakes, suggesting local conditions to control amplitudes. On the contrary, precursor time and duration of anomalies seem to increase both with magnitudes and epicentral distances. Similar conclusions were obtained by analysing data recorded continuously at the Friuli site of Cazzaso (Riggio and Santulin, 2012).

Several groups investigated the maximum distances between the epicentre of an earthquake and the site of the observed radon anomaly. Many empirical relationships or relationships based on theoretical considerations on the diffusion of radon, were obtained. Widely used is that of Hauksson and Goddard (1981):

$$M \geq 2.4 \log_{10} D - 0.43 \qquad (1)$$

where M is the minimum magnitude required to obtain a radon anomaly at distance D (km).

The application of this relation, allows making a selection of the used catalogue events, and can give information on the area affected by the deformation process that precedes an earthquake of a given magnitude, defining the distance at which we can detect an anomaly attributable to a given earthquake.

For the determination of the time precursor, one of the first empirical relationships, was that of Rikitake (1976):

$$\log t = 0.76\, M - 1.83 \qquad (2)$$

where t is the time precursor, and M the magnitude of the impending earthquake.

Higher fluid flows are expected in reverse-fault area than in normal faults during interseismic regimes (Muir-Wood and King, 1993). Accordingly, soil gas prospecting might be more effective in detecting fractures in the compressional regime.

3. Radon as precursor

Radon is a natural gas, produced in soil, by the radioactive decay of the radium element, produced in turn by uranium. Radioactive decay is a natural, spontaneous process in which an atom of one element decays or breaks down to form another element by losing atomic particles. Radon itself is radioactive because it also decays losing an alpha particle and forming the element polonium. The half-life of radon is 3.8 days.

Because radon is a gas, it has much greater mobility than uranium and radium, which are fixed in the solid matter of rocks and soils. Radon can leave the rocks and soils more easily by escaping into fractures and openings in rocks and into the pore spaces between grains of soil. It can travel a great distance before it decays and gathers, in high concentrations, also inside a building. Radon travels by diffusion (but in this case it moves slowly) or by convection through gas carrier (as methane, carbon dioxide and nitrogen). Radon is formed in the rocks as a result of the decay of radium-226; high concentrations of radon gas in the soil and subsoil are found, but only where this item can be expelled from the crystal lattice of minerals that contain it (see e.g., Petrini *et al.*, 2012). In particular, in the decay of radium-226, an alpha-particle is emitted and the newly formed radon atom recoils in the opposite direction. The position of the atom of radon in the granule and the direction of recoil atom itself can determine whether or not the leakage of radon from the crystal lattice of the mineral that generated it. Under these conditions, three different situations may occur: the radon atom remains in the granule, the atom of radon enters a adjacent granule, the radon atom is ejected from the crystal lattice and is subsequently removed from the gases from soil or water. Only in the third case, the radon is actually free to move through the soil, to reach the surface, and finally spreading into the atmosphere; its mobility will be linked to the permeability of the soil and to the degree of fracturing of rocks.

Radon moves more rapidly through permeable soils, such as coarse sand and gravel, than through impermeable soils, such as clays. Radon is moderately soluble in water. Its solubility depends on the temperature of the water: colder the water, greater is the solubility of radon. A measure of the solubility of gas in water is given by the solubility coefficient, defined as the ratio between the concentration of radon in water and in air. At 20°C the coefficient of solubility is about 0.25, which means that the radon is preferentially distributed in the air rather than in water (Riggio and Sancin, 2005; Riggio and Santulin, 2012 and references therein).

The connection between the anomalies of chemical and physical parameters and seismic events has been explained, in the past, by the dilatancy model (Scholz *et al.*, 1973): opening of cracks before an earthquake, increases the diffusion of pore fluid and, together with the modified strength and pore pressure, causes variations in the chemical-physical characteristics of the rocks. The increase of the radon concentration, particularly in compact rocks, happens when the cracks start to form in the rocks of the involved area in the impendent earthquake. During the last stage of the dilatancy model, the radon emission can be stable or decrease before the earthquake. The width of the zone involved by the stress loading is proportional to the magnitude and to the depth of the impending earthquakes. The pressure variations, caused by the stress loading, lead changes of the rocks characteristics constituting the "precursor phenomena". The pattern dilatancy does not seem to justify the observation of precursory phenomena even at great distances from the epicentral area of the earthquake that will occur. Actually, the problem lies in the definition of the area to investigate. The preparation of a strong earthquake, or simply a substantial crustal deformation, involves, in general, a very wide area (even hundreds of kilometres). In the monitoring sites of the precursors, even very far from the future epicentral area, some local conditions can be alterated, described by the theory of dilatancy, which allow the occurrence of precursory phenomena. The first objection to the use of radon as an earthquake precursor was that the radon decay time did not allow radon to travel great distances within the Earth. But, in reality, the stress propagation, moving in the soil, creates a lot of "local" radon anomalies, even a hundred miles away.

4. Radon detectors and acquisition methods

Instruments for the measurement of radon and its decay products are based mostly on the detection of alpha particles either emitted by radon itself or by its decay products.

Two techniques are available for measuring radon activity: passive and active mode. In the first, radon enters in the detection system by natural diffusion, in the second, radon is pumped into the device. The radon measurements can be performed in an integrating, discrete (grab sampling) or continuous mode, regarding the measurement duration. The best technique to use depends on the application and on the location of the sounding tube (in soil, in air and in water; Table 1).

Table 1 - Specific data of radon measurements devices (modified from Papastefanou, 2007).

Method	Type	Field area	Volume	Sensitivity	Time period
Alpha track-etch detectors	Integrated	Gas	456 ml	0.03-0.09 tracks cm^{-2}/kBq $^{m-3}$ h	1-2 weeks
Electret ion chambers	Integrated	Gas	50 ml-960 ml	3 Bq m^{-3} h-1.05 kBq m^{-3} h	2-40 days
Alpha scintillation detectors	Instantaneous	Gas/water	0.1 l-3.0 l	0.8-16 cph/Bq m^{-3}	1-5 min
Continuous radon monitors	Continuous	Gas/water	590 ml	0.02 pulses h^{-1}/Bq m^{-3}	1-15 min-48 h

Some types of the most used passive detectors are the following:

- alpha track detectors are integrative, passive radon sampling devices that do not require Ac power. They contain a thin piece of plastic or film mounted in the detector. They are used for in soil and indoor radon measurements. In the Philippines, for example, long-term soil gas radon measurements were carried out in selected sites along the extent of the Valley Fault System using solid state nuclear track detector (SSNTD) employing LR-115 type 2 Kodak film (Ramos *et al.*, 2012). The plastic detector is buried in the ground for at least 30 days after which it is retrieved and replaced with a new plastic detector. This period of time allows the continuous integration of signal produced by the emanating radon and reduces large fluctuations caused by changing meteorological conditions. Radon flows into the detector through a filtered opening. As the radon inside the detector decays, the emitted alpha particles hit the film forming tracks on the film. After the film processing, the tracks are counted to determine radon concentration. The number of tracks recorded corresponds to the integrated radon concentrations and reported in number of tracks per square millimetre per day. The results are, then, analysed statistically to determine the radon anomaly. These detectors are not very effective in measuring high concentrations of radon because these detectors are also sensitive to gamma radiation. Alpha track detectors usually are preferred in situations where confirmation of measurements made with short-term integrating devices is needed;
- other types of devices using SSNTDs are the Electret Ion Chambers (EIC - Rad Elec Inc.) as passive environmental radon monitors for the measurement of radon flux from the soil (Kotrappa *et al.*, 1992). These are integrating ionization chambers wherein the electret (permanently charged Teflon disk) serves both as a source of electrostatic field and as a sensor. It consists of an electret mounted inside a small chamber made out of conducting plastic. The ions produced inside the chamber are collected onto the electret causing a

reduction of the surface charge on the electret. The reduction in charge is a measure of the ionization integrated over a period of exposure to alpha particles emitted by the decay process of radon gas and its decay products. The volume of the ionisation chamber ranges from 50 to 960 ml. The exposure periods range from 2 days to 40 days (Papastefanou, 2007);

- activated charcoal devices are passive sensors that do not require Ac power. They consist of a canister that holds granular-activated carbon (i.e., activated charcoal). The charcoal absorbs radon that enters the canister via a screened opening. After a determined exposure period, the canister is sealed and the charcoal is analysed using an HPGe gamma ray detector. Activated charcoal devices are preferable for short-term measurements (i.e., 1 to 7 days) and are commonly used to determine whether houses exceed a radon reference level. Moreover, they are not practical for locations with high humidity because the charcoal can become saturated.

In the last 35 years, in the United States, long-term alpha track detectors are used to confirm the performance of a mitigation system on an annual basis or for yearly determination of outdoor radon. In Europe, Canada, Asia, and other countries, long-term radon measurements lasting from 90-365 days are emphasized over short-term measurements. The active devices use, prevalently, scintillation or ionization chambers.

The scintillation cell method is one of the oldest and most widely used for grab sampling of radon and its decay products in the field. It was ideal for making measurements in radon exhalation, radon soil-gas, indoor, outdoor, in mining environments, in-water storage radon measurements and in the research. Scintillation cells (e.g., Lucas cells) range in size from 0.1 to 3.0 l and they are made by using metal, glass or plastic containers coated internally with silver activated, ZnS(Ag) powder. The principle of detection is the counting of light photons resulting from the interaction of alpha particles from radon and its decay products decaying in the cell, with the ZnS(Ag) phosphor. For counting the light photons, the scintillation cell is coupled to a photomultiplier tube assembly system. The advantage of using a scintillation cell is that it eliminates the need for sample transfer before counting, and, when properly maintained, it can be reused for years after a very good cleaning with nitrogen gas (inert gas). In the case of soil gas, the gas is pulled off at opened holes of 70 to 100 cm deep. For underground waters, a water-degassing unit (kit) is used.

Some of the new commercial continuous radon monitors are able to work with both the two techniques: passive, where air diffuses in the sensitive volume (ionisation chamber) passively, and active, where the radon is brought to sensitive volume of the instrument by an air pump. They are electronic computerized instruments. They are also used to monitor radon test chambers in which other type of instruments are tested, inter-compared, evaluated and calibrated and they have additional benefits, as simultaneous measurement of air temperature, air pressure and air humidity. Recent instruments are portable, battery powered and used for different measurements of radon. Measurement intervals generally range from a minimum of 1 to 15 min up to 48 h. The primary advantage of continuous radon monitors is that they provide real-time radon concentration data, also handling them remotely, using a modem or satellite modules. The same instruments are able to measure radon in water or other liquids. Usually they need a set of accessories for measuring directly radon gas of liquid samples.

5. Radon anomaly and earthquake prediction

Several radon investigations have been carried out all over the world and many of these have been listed by various authors. Measurements of this gas both in soil and in groundwater have shown that spatial and temporal variations can provide information about geodynamical events.

The problem of the definition of anomaly and its parameters (amplitude, duration, epicentral distance and precursor time) was to be dealt with by different authors (Kissin and Grinevsky, 1990; Toutain and Baubron, 1999; Hartmann and Levy, 2005). The majority of the analysed data put in evidence that the precursor time and duration grow with the magnitude and have a strong correlation between them. The precursor time decreases with the increase of epicentral distance and duration decreases with low distances. The amplitude appears to be independent of the magnitude and distance.

Only after the earthquake of L'Aquila, radon has reached the pinnacle of his fame. Indeed, the first measurements of radon in groundwater were made by Shiratoi (1927) and Imamura (1947) in Japan, and the first measurements of radon in soil were made in 1953, always in Japan, along an active fault zone for two years (Hatuda, 1953). Radon concentration in soil gas was measured and anomalous radon concentrations were reported before the strong earthquake (*M*=8.0) of Tonankai (Japan, December 1944). In 1956, Okabe also showed that the air, close to the soil surface, shows a significant increase in the concentration of radon when an earthquake is approaching (Okabe, 1956). These were the first studies on radon, when the instruments were not very sophisticated and measurements were made on the surface of the soil. The literature of the following years, is full of works about the subject.

One has to distinguish between the results obtained about radon as a precursor and the alarm resulting in the evacuation order.

Some pioneering works on radon in water were made in former Soviet Union during the period 1966-1971. In 1968, Ulomov and Mavashev (1968, 1971) observed anomalies in radon concentration in hot mineral water from an aquifer (1300-2400 m deep) in a Tashkent artesian basin (former Soviet Union), before the Tashkent earthquake of *M*=5.3 in 1966 and some other shocks of *M*=3.0-4.0. The data analysis was made after the earthquake: several years before the earthquake the radon content reached his highest value until the earthquake, and then it returned to its normal value. The same pattern was reported before its larger aftershocks, on a shorter time scale (Fig. 1). The anomalies-epicentre distance was within 5 km. These results gave hope to research on seismic precursors and many studies were performed about radon anomalies and earthquakes. Always in former Soviet Union, since 1974, extensive works have been done regarding radon concentration in groundwater or springs by Sultankhodzhayev *et al.* (1980). Prominent precursor signals were observed before the Markansu (1974), Gasli (1976), Alma-Ata (1978) earthquakes, although some other quakes did not have any distinct precursor signal. The magnitude and the epicentral distance were respectively *M*=7.3 and 530 km; *M*=7.3 and 470 km; *M*=7.1 and 65 km. The duration of radon anomaly was 100, 4 and 50 days respectively. The focal mechanism was respectively of strike slip, normal and transtensional. Many years after the studies of Ulomov and Mavashev, Pulinets *et al.* (1997) compared radon concentration in the well near Tashkent, where an earthquake, with medium magnitude, occurred on December 13, 1980, with peak electron density of the ionosphere in the vicinity of the earthquake epicentre; they resulted in phase opposition on a period of 18 hours. Radon

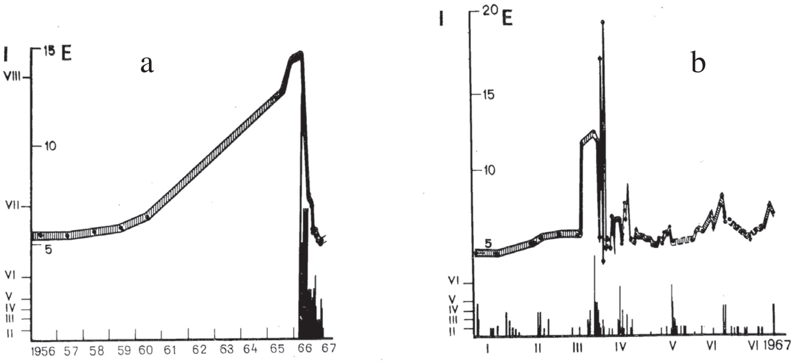

Fig. 1 - The first evidence of radon in groundwater as precursor of earthquakes (from Ulomov and Mavashev, 1971). Radon concentration, in water, before the 1966 Tashkent earthquake of *M*=5.3 (a) and its aftershocks (b).

concentration increased with a sharp maximum few days before the quake, followed by a sharp fall on the day of the shock.

Since 1970, China was the first state organized for earthquake prediction. In 1975 there was the first prediction that resulted in an evacuation, saving the lives of thousands of people. The 7.3 Haicheng earthquake on February 4, 1975 was the first major shock that has been accurately predicted anywhere in the world. Before reaching the forecast, several previsions were made for previous dates without any provisions for evacuation. At the Liaoning provincial Earthquake Office, the Jinxian levelling data and other observations, such as the radon and tilt anomalies discussed by Raleigh *et al.* (1977) were useful in sending alerting signals months and weeks before the earthquake, although there is no evidence that these data played any role in the provincial imminent prediction. Radon anomalies were observed in the water of the wells, distant also 200 km from the epicentre of the earthquake and contributed to the forecast in the medium and short term. The intensification of foreshocks in the epicentral area started the alarm (Wang *et al.*, 2006). A comprehensive prediction was made by analysing all pieces of information acquired both by professional stations and by the many monitoring points operated by amateurs (Fengming and Ge, 1975). One year and half after the earthquake in Haicheng, another strong earthquake struck China on July 28, in Tangshan, with a magnitude of 7.8. This time, however, order to evacuate was no issued even if abnormal signals were mentioned for Beijing, Tianjin, Tangshan, Bohai and Zhangjiakou regions. Changes of the radon content of 27 wells (springs) in north China (300 km far away) before and after the Tangshan earthquake were observed as other precursors, but there was no unified criterion for judging these anomalies. The earthquake was not preceded by foreshocks. The anomalies might be related to both the Haicheng (1975, *M*=7.3) and Tangshan earthquake, owing to their proximity in time (1.5 years) and space (350 km) (Toutain and Baubron, 1999). In one county the alarm was given, but this was not enough to mitigate the disaster.

Even in Japan, the first program for the prediction of earthquakes was defined in the1960s, but the radon measurements were not included in the government program. Nevertheless, many researchers carried out studies and measurements on radon. Hirotaka and his group observed radon in soil anomaly before the Nagano Prefecture earthquake of *M*=6.8 on September 1984 (Hirotaka *et al.*, 1988) and the measuring site was about 65 km away from its epicentre at the Atotsugawa fault. They observed a gradual increase in radon count 2 weeks before the

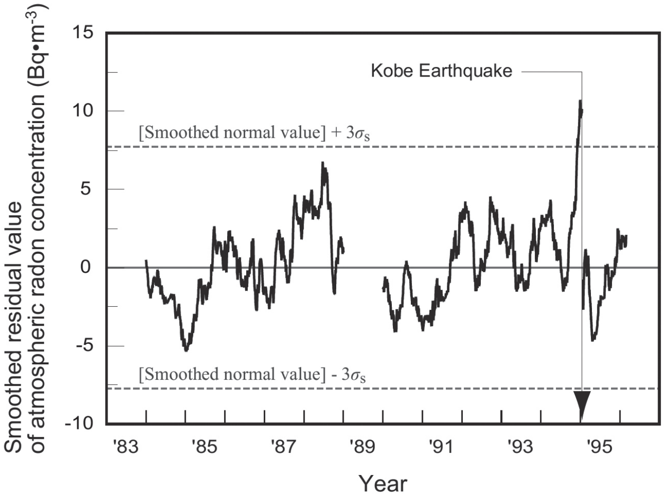

Fig. 2 - Air radon concentration vs. time (by Kobe Pharmaceutical University) before the M=7.2 Kobe earthquake of January 17, 1995 (Kawada *et al.*, 2007).

shock. A spike-like radon in water anomaly was recorded 5 days before the Izu-Oshima-kinkai earthquake [M=7.0, January 14, 1978: Wakita *et al.* (1980)], in a 350-m deep artesian well, 25 km away from the epicentre. The Kobe earthquake of January 17, 1995, M=7.2, which is often taken as an example of the failure of the deterministic forecasting, had been preceded by marked anomalies of radon in water, but, since there was no foreshocks activity, the signal was not taken into account. The observation well was 17m deep and 30 km away from the epicentre. The radon concentration was stable at 20 kBq/m^3 at the end of 1993 and started to increase gradually from October 1994 reaching the value 60 kBq/m^3. Then there was a sudden increase on January 7, 1995, 10 days before the earthquake, and again a sudden decrease on January, 10 1995, 7 days before the earthquake. The sudden increase might be due to the formation of microcracks in the aquifer system and the decrease might be due to sealing of the cracks. The associated fault is a high-angle east-dipping reverse fault that juxtaposes Early Pleistocene sediments with granitic bedrock (Okumura, 1995). In 2007, Omori and his group published a study on the concentration of radon in the atmosphere (Omori *et al.*, 2007). The site investigated is located in Kobe and the measurements of atmospheric radon concentration had been running since 1984, the measurements were carried out every hour, continuously and automatically. They found an anomalous increase during winter 1994-1995, just before the Kobe earthquake (Fig. 2).

A radon monitoring system was installed in the Yugano hot spring, Izu peninsula, in May 1995 (Nishizawa *et al.*, 1998). Ondoh (2009) carried out an extensive study on investigation of radon anomaly in groundwater and investigated the nature of radon anomaly occurred during three earthquakes of M>6.5. No deterministic prediction was made for the largest earthquake that occurred in Japan on March 11, 2011, because there was no longer a national project for the observation of precursory phenomena. *A posteriori*, recordings from satellites of the atmosphere heat were analysed (Ouzounov *et al.*, 2011). The atmosphere above the epicentre of the earthquake, had unusual changes in the days before the disaster, according to preliminary data in line with the “Lithosphere-atmosphere-ionosphere coupling mechanism” theory for which just before an earthquake, the land protrudes a considerable amount of odourless, colourless gas: radon. The primary process is the ionization of the air produced by an increased emanation of radon from the crust (Omori et al., 2007). The increased radon emanation launches the chain of physical processes, which leads to changes in the conductivity and temperature of the air.

In the U.S., radon measurements in soil were made at two sites of the Calaveras Fault, California (King and Slater, 1978), and above the area of the San Andreas Fault. Some anomalies occurred during earthquakes of medium magnitude (coseismic) (Ghosh *et al.*, 2009). High radon concentration during spring 1978 was not associated with any earthquake occurred soon after, but on August 13, however, there was the earthquake in Santa Barbara, which had a moderate magnitude (5.1-5.7) but caused a lot of damage. Vice versa, no radon anomaly was observed during the June 1977 earthquakes of M=4.0 and 4.6.

The best results were obtained from radon measurements in water. Teng *et al.* (1981) measured radon concentration in 14 water wells and spring water samples along the San Andreas Fault. They observed anomalies several hundred percent greater than background before earthquake. Precursor signals were recorded 1 day to few weeks before the Big Bear earthquake of M=4.8 on June, 1979 at both hot and cold springs, within a 60-km distance. Other measures were made, previously, in 16 thermal springs and wells during 1974.

Although geochemical data recorded at properly located stations have shown anomalous changes before earthquakes, the mechanism for these changes was not understood. Radon, hydrogen and carbon dioxide gas, and temperature in groundwater measurements were maintained in wells near Parkfield. Water samples were collected periodically from these and several other wells for chemical analysis. In addition, continuous soil-gas radon and hydrogen monitors were installed near some of the groundwater monitoring sites. However, only two monitoring sites of radon in water were included in the network estimates. The acquisition, however, was not continuous throughout the period.

Radon measurements in soil and water were made also in Turkey. Radon (^{222}Rn) concentration has been continuously measured since 1983 in groundwater at a spring and in subsurface soil gas at five sites along a 200-km segment of the North Anatolian Fault Zone near Bolu, Turkey. The groundwater radon concentration showed a significant increase before the Biga earthquake of magnitude 5.7 on July 5, 1983, at an epicentral distance of 350 km, and a long-term increase between March 1983 and April 1985 (Friedmann *et al.*, 1988). Other studies were made more recently, but not always a correlation was found between the anomalies and seismic activity.

The list of countries that have carried out studies on radon as a precursor is very long.

Just a hint about Iceland because the first studies were made by Hauksson and Goddard (1981), who, based on their results, derived the formula that relates the maximum distance where you can observe an anomaly, to the magnitude of the event that will occur. Radon concentration in geothermal water was measured in 9 wells. The sampling frequency was once or twice per week. The probability of observing radon anomalies before earthquakes with magnitudes between 2.0 and 4.3 (the largest event observed) was found to be approximately 65%. Since 1999, there was an active program in thermal waters, but the results highlighted only co-seismic or post-seismic changes.

In Austria, Friedmann (1985) started radon monitoring in groundwater of 5 springs using ionization chamber in 1977: they recorded an anomalous increase of a factor of 3 for 8 months, 3 months before the Montenegro earthquake, of M=6.9 in April 1979, about 650 km away from the site. The radon concentration in a thermal spring, at Warmbad Villach, is currently measured by an ionisation chamber (Santulin *et al.*, 2005).

In Slovenia, radon measurements are performed both in water and in soil (Zmazek *et al.*, 2005; Gregoric *et al.*, 2008; Vaupotic *et al.*, 2010). The researchers observed several anomalies in radon concentration that might have been caused by seismic events. They developed advanced statistical methods to highlight anomalies caused by tectonic stress from those caused by meteorological and hydrological effects. With these procedures they were able to correctly predict 10 seismic events out of 13 within the 2-year period (Torkar *et al.*, 2010).

In India, measurements of radon in soil gas and groundwater were carried out at Palampur since 1989 by active instruments with a scintillation cell. Radon anomalies were recorded simultaneously in both soil gas and groundwater. Weekly integrated data also showed abnormal radon behaviour during the first week of October 1991 at different recording stations. These recorded anomalies were correlated to an earthquake of magnitude 6.5 occurred in Uttarakashi area in October 1991. Also the Chamdi earthquake of March 1999 with M_b=6.8 was preceded two days earlier by variations of radon in water and soil (Virk and Walia, 2001).

Numerous studies on radon were carried out in Italy, mostly by individual researchers, by institutions or by amateur groups. There was not a coordination of activities at national level. This was due to the fact that it was thought that this field of seismology had no immediate results useful for a deterministic forecast.

The first measures, which were analysed as seismic precursors (Fig. 3), are those made by Allegri and his team from 1979 to 1980 (Allegri *et al.*, 1983; Pulinets and Boyarchuk, 2004; Pulinets *et al.*, 2007). These researches of the Institute of Physics and Institute of Geology of the University of Rome carried out measurements of tilt and groundwater radon content in sites located in central Italy. Beginning from June to November 1980 anomalous variations in the radon content were detected at the Rome and Rieti sites. Water samples for discrete measurements of radon content were drawn periodically from underground strata. The amplitude of the anomalies was 25% and 170% compared with the background level. On November 23, 1980 a strong (M=6.5) earthquake took place in Irpinia (southern Italy) at about 250 km from Rome and Rieti.

Positive anomalies of ^{222}Rn were recorded in the liquid phase of some mud volcanoes of several earthquakes occurred in 1986 and 1987 in the northern Apennines. The data analysis showed that a correlation might exist between large radon anomalies and local earthquakes with M>2.5. (Martinelli, 1987; Martinelli *et al.*, 1995).

Since October 1994, the Department of Earth Sciences of the Trieste University installed a station for the integrated monitoring of the horizontal deformation, tilt variations and radon emanation from soil operating in a natural cave located in a high seismicity area of north-eastern Italy (Garavaglia *et al.*, 1998, 2000). A device built at the I.F.G.A. (Institute of Applied General Physics of Milan) was used to measure the radon exhalation; this is an alpha-meter (a scintillation counter of particles) coupled with a measuring box. Air was pumped from the ground to the measuring box. The best correlation coefficients were found between the radon and the N-S tilt measurements. This direction coincides with the Alpine compressive tectonic stress present in the area. This suggested that the radon increasing is correlated with micro-compressive episodes also without seismicity.

In October 1992, the first site for the continuous measurement of radon in water was created at the geochemistry station of the ING (now INGV) of Pozzo Barozze in Rocca di Papa (Rome), still working (Calcara and Quattrocchi, 1993). Since 2005, also by radon in soil is monitored by cell ionization instrumentation (AlphaGuard), the Geodynamic Observatory of Rocca di Papa (Pagliuca *et al.*, 2007).

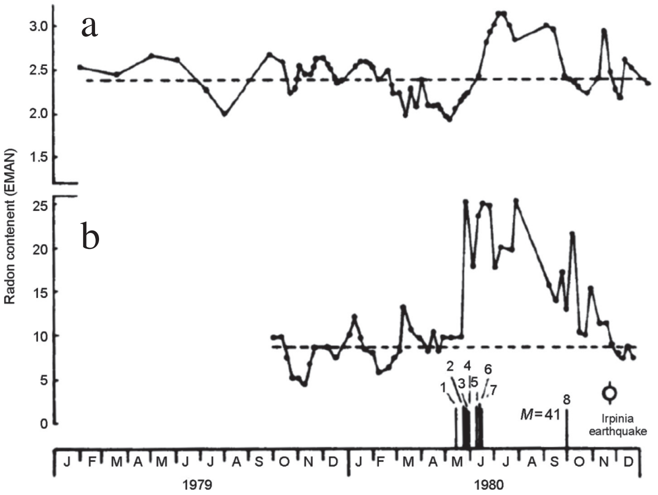

Fig. 3 - The 1979-1980 series of the groundwater radon content at: a) Rieti station, b) Rome station. The vertical bars in the bottom panel show the seismic shocks. The length of the bars is proportional to the magnitude (modified from Pulinets and Boyarchuk, 2004).

In 1996, a radon survey station was set up at Cazzaso, in the Friuli area (NE Italy), by OGS. The survey site is equipped with a Lucas scintillation cell type (Prassi-Silena), settled with continuous radon recording. The accuracy is of 4 Bq/m^3. The air is inhaled by pumping, from a 40.5-m deep well with a 9-cm diameter, at a 7-m depth. Sampling intervals are of 3 hours (Riggio *et al.*, 1999). Unfortunately, the time series is discontinuous because of interruptions due to malfunction and, from 2000 to November 2002, to the closing due to lack of funds. The influence of meteorological parameters was verified by different correlations. The variations due to meteorological causes were lower than those attributable to geodynamic causes (Riggio and Sancin, 2005). Changes in the concentration of radon that exceed the limit of +/- 2 sigma from the mean value, were considered "anomalies", according to Igarashi and Wakita (1990). Both short-term (days, weeks) and long-term (months, years) anomalies were analysed before the occurrence of earthquake. Fig. 4, as an example, shows the concentration of radon in soil, where one can see both of them. In the first case, the seismicity is analysed with individual earthquakes selected according to Hauksson and Goddard (1981) and shown in Fig. 4. The best results have been obtained for local seismic series with main event of magnitude greater than, or equal to, 4.0, but anomalous values have been found in correspondence of all local earthquakes with magnitude greater than, or equal to, 3.0 (Riggio *et al.*, 2003). In the second case, if the value is over the 2-sigma line for a long period, with short breaks of 1-2 days, it is considered a long period single anomaly. This type of anomaly has been observed in 2003-2004 and in 2006-2009 during the periods preceding earthquakes with magnitude greater than 5.0.

In 2000, a collaboration began between OGS and the Jozef Stefan Institute of Ljubljana, formalized by bilateral projects. In the frame of these projects, the Friuli data were analyzed in conjunction with those recorded in the thermal waters of Slovenia with passive Barasol probe. In 2004, a Barasol Algade MC 450 probe was installed, in soil, at a distance of 100 m from the Prassi instrument. A clearly visible and large anomaly was observed three weeks prior to the M_L=5.1 earthquake on November 24, 2004, near the Lake Garda (Fig. 5): Vaupotic *et al.*, 2010.

Since 2006, in collaboration with the INGV of Palermo and the DiGe Department of the Trieste University, the chemical analysis and the radon measurements are performed monthly in the water well and in other water springs in Friuli and Slovenia. The physical characteristics

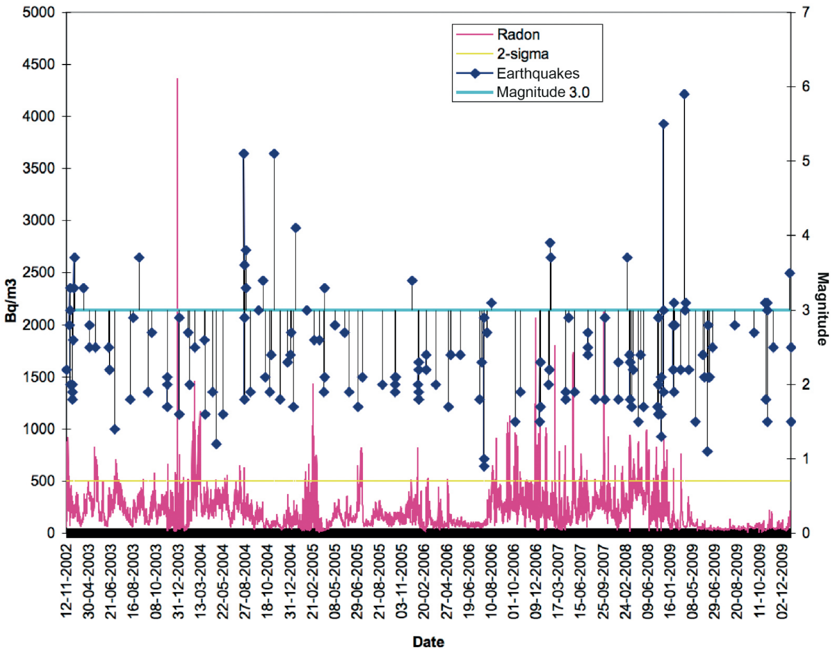

Fig. 4 - Radon in soil at the Cazzaso well (from Martinelli *et al*., 2013).

(temperature, pH, conductivity, and redox potential) are measured in the field. Radon activity was measured, in laboratory, by both a scintillation cell and an ionization chamber (Silena Prassi and Alpha Guard, respectively, with an accuracy of 4 Bq/m^3 and 1 Bq/l respectively). The springs have been chosen following the available scientific literature (Riggio and Santulin, 2012) and according to their location in proximity of faults and to their chemical characteristics (temperature, pH, conductibility).

Within the European project MICRAT, which started as a result of the Umbria earthquake in 1997, INGV installed two sites for the monitoring of radon in water, from 2000 to 2001, with a sensor equipped with scintillation cell, built at the institute. At the same time, monitoring of the chemical characteristics of the water was performed. Important correlations were not highlighted.

In Piemonte, the emission of radon along the fault of Cremosina was controlled by the Geophysical Observatory of Novara from 2002 using the alpha tracks (or “etching method”). Currently, there are 5 sites operating for the continuous monitoring of radon in soil.

In the frame of the INGV-DPC S3 project (Albarello, 2013) a database containing all radon data collected in soil, water, air was created and a large amount of data concerning indoor radon which cannot be easily correlated with seismicity, on the national territory, and whose owners have agreed to include in the data base. The spatial distribution of the data collected is shown in Fig. 6.

6. Conclusions

The results seem to indicate that the radon is a good indicator of crustal activity such as earthquakes, but there are still many tests to be done to achieve a deterministic forecast. Nevertheless, the prediction of catastrophic earthquake is a scientific objective.

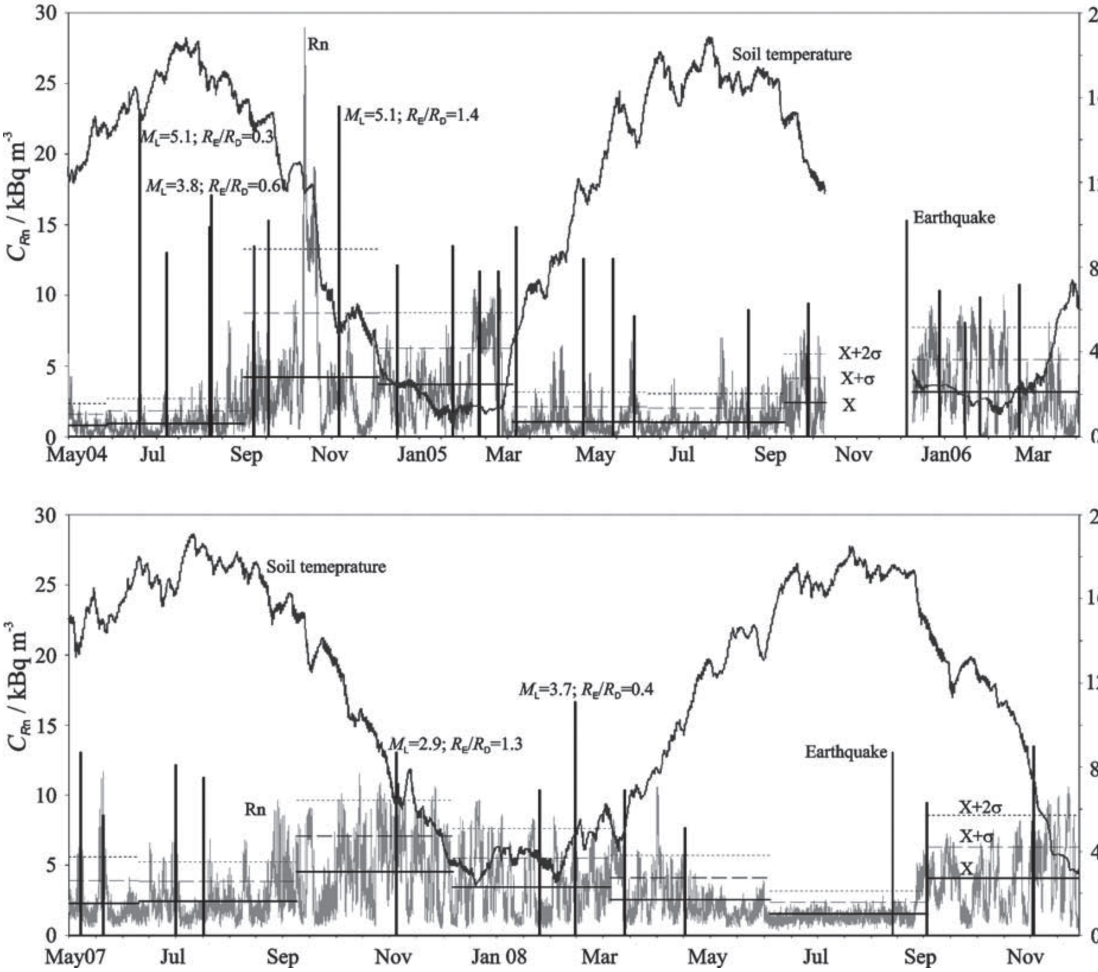

Fig. 5 - Time series of hourly radon concentration in soil gas at Cazzaso (Friuli) recorded by Barasol, and soil temperature for 2 periods: upper graph from May 2004 to April 2006, and lower graph from May 2007 to December 2008. Also earthquakes are drawn as bars. Solid lines indicate average seasonal radon concentration, dashed lines +1-σ deviation, and dotted lines +2-σ deviation from the seasonal average value (modified fromVaupotic *et al.*, 2010).

The amplitude of the deformation zone and of the precursor time for a given magnitude must be well evaluated.

The expectation that an anomaly should be strongest shortly before of the earthquake and nearer to the site of the future earthquake is based on a temporally and spatially smooth mechanical model and is probably too simplistic.

The time series must be long enough to allow the false positives and false negatives identification.

To obtain accurate results, the acquisition must be continuous, teletransmitted and in more observation points.

The possibility of applying the machine-learning methods to identify radon anomalies should be evaluated, but without underestimating the human interpretation because each area has different characteristics.

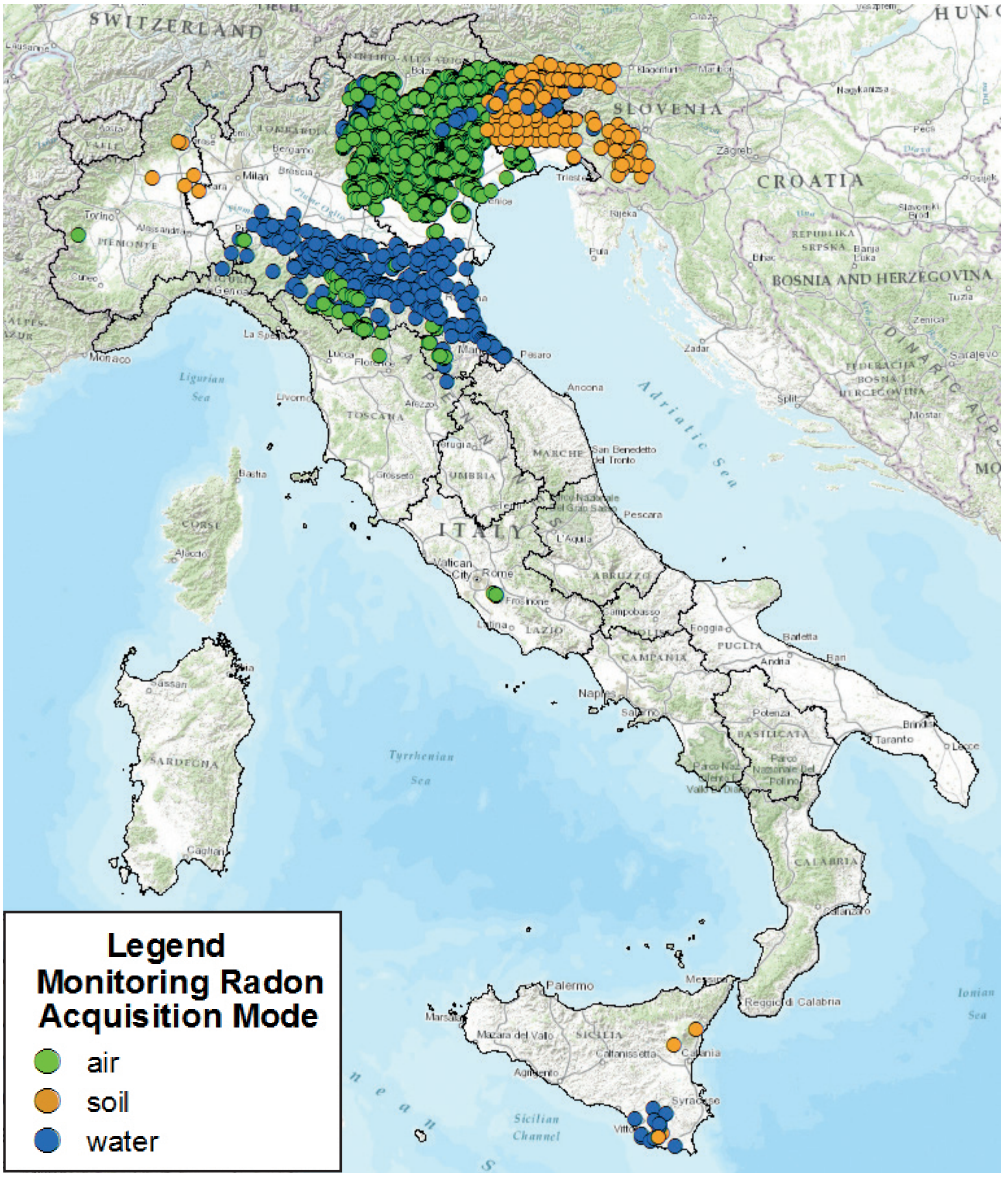

Fig. 6 - Map of the spatial distribution of sites with radon measurements.

Integrated measures of radon in soil and water should be carried out in conjunction with the acquisition of other parameters, in particular, the measurement of soil temperature and radio broadcasts.

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