NUKLEONIKA 2010;55(4):573–577

ORIGINAL PAPER

Radon in outdoor air in the Mt. Etna area, Italy

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Abstract. Radon (^{222}Rn) activity concentration in outdoor air was measured, using AlphaGuard radon monitors, at 25 points in the Mt. Etna area, at different distances and in different directions from the summit. Values of up to 93 Bq·m^{-3} were found at three points close to the summit, at altitudes of about 3000 m above the sea level, and in the range of 3.0–19.6 Bq·m^{-3} elsewhere. An average radon concentration of 7.9 Bq·m^{-3} was obtained in the northern region, 4.8 Bq·m^{-3} in the eastern (oriented towards the sea), 9.6 Bq·m^{-3} in the western (the furthest inland), and 6.5 Bq·m^{-3} in the southern.

Key words: radon • outdoor air • Mt. Etna • AlphaGuard monitors

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Received: 10 September 2009
Accepted: 30 December 2009

Introduction

Radon (^{222}Rn) is a radioactive (half-life of 3.85 d) noble gas, originating from radioactive transformation of ^{226}Ra in the ^{238}U decay chain in the earth's crust. Only a fraction of radon atoms thus created are close enough to the surface of solid grains to be able to leave the solid (emanate) and enter the void space, from where they migrate by diffusion, and, to longer distances, by advection or transport by carrier fluids or gases. Radon accumulates in underground caves, caverns and mines, and eventually exhales into the atmosphere [10]. Radon does not undergo chemical interactions with the medium it traverses, but is transformed into its short-lived decay products, i.e., ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po. Its activity concentration in outdoor air may reach several tens of Bq·m^{-3}, and in indoor air of buildings, several hundreds. The transport by carrier fluids and gases is influenced by geophysical parameters, among them seismic and volcanic activities. Eruption of the Karymsky volcano in Kamchatka in 1970 was one of the first documented to be preceded by a distinct increase in radon activity [11]. Since then, the influence of volcanic activity on radon levels has been reported for several other volcanoes worldwide, e.g., Cerro Negro in Nicaragua [7], Galeras in Colombia [13], Popocateptl in Mexico [28], Merapi in Indonesia [31], and Vesuvius [6], Etna [15] and Stromboli [25] in Italy.

At Etna, radon has been monitored in soil gas, indoor air and in groundwater. Variations in radon concentration in soil gas in boreholes at the flanks of Mt. Etna have been proven to be related to changes of geodynamic activity of the volcano [15–17, 22, 23, 26]. Moreover, radon in soil gas has been observed as an indicator of hidden active faults [4]. Radon, together with pH, temperature and CO_2, was measured in three types of groundwater, in wells, springs and drainage galleries, with mean concentrations of 13.5, 10.9 and 7.8 kBq·m^{-3} [9]. The highest radon concentrations were found in the geodynamically most active eastern part. In contrast, on the southern part, although with high seismicity and intense magmatic degassing, values were low, possibly because of huge magmatic flux, resulting in CO_2 oversaturation and thus separating a free gas phase scavenging radon [8]. In water samples from 7 wells, 3 springs and 3 galleries from the Mt. Etna aquifers, in addition to ^{234}U, ^{238}U, ^{226}Ra and ^{228}Ra, also ^{222}Rn was analysed. The activity concentrations of ^{226}Ra, except in one of galleries, were below 10 Bq·m^{-3}, and those of ^{222}Rn varied from 1.4 to 12.7 kBq·m^{-3} [21]. A negative correlation for ^{222}Rn with the total dissolved solids was observed, while it was positive for CO_2 [9]. Indoor air was surveyed in an area on the south-eastern flanks [2]. Higher concentrations have been found in buildings located near or on geological structures, such as faults and fractures. This survey is continuing.

In this paper, results of the first radon measurements in outdoor air in the Mt. Etna area, at different distances and directions from the summit, are reported and discussed.

Geology

Mt. Etna, located in eastern Sicily, is the largest stratovolcano in Europe (more than 3300 m high, 40 km in diameter, with an area of about 1200 km^2) and one of the most active in the world. Its location is close to the collision boundary of the African and Eurasian Plates [5]. The area is covered by several tectonic zones (faults and fissures in different directions) which are pathways for magma [18] and natural gases, including radon. The composition of Etna's lava is hawaiitic alcalic basalts over a thick (> 15 km) sedimentary substratum composed mostly of clays (east and south) and clay, marl and quartz-arenite units (east and west) [24].

Experimental

Radon activity concentration was measured in outdoor air at 22 points at the Mt. Etna volcano. Our survey area extended from the coast on the east to Linguaglossa and Randazzo on the north, Bronte and Adrano on the west, and Paternò and Acireale on the south (Fig. 1). Two AlphaGuard radon monitors (Genitron, Germany) were used, one kept as a reference in permanent operation at Stazzo, a village at the coast several kilometres from Acireale to the north, and the other, transported

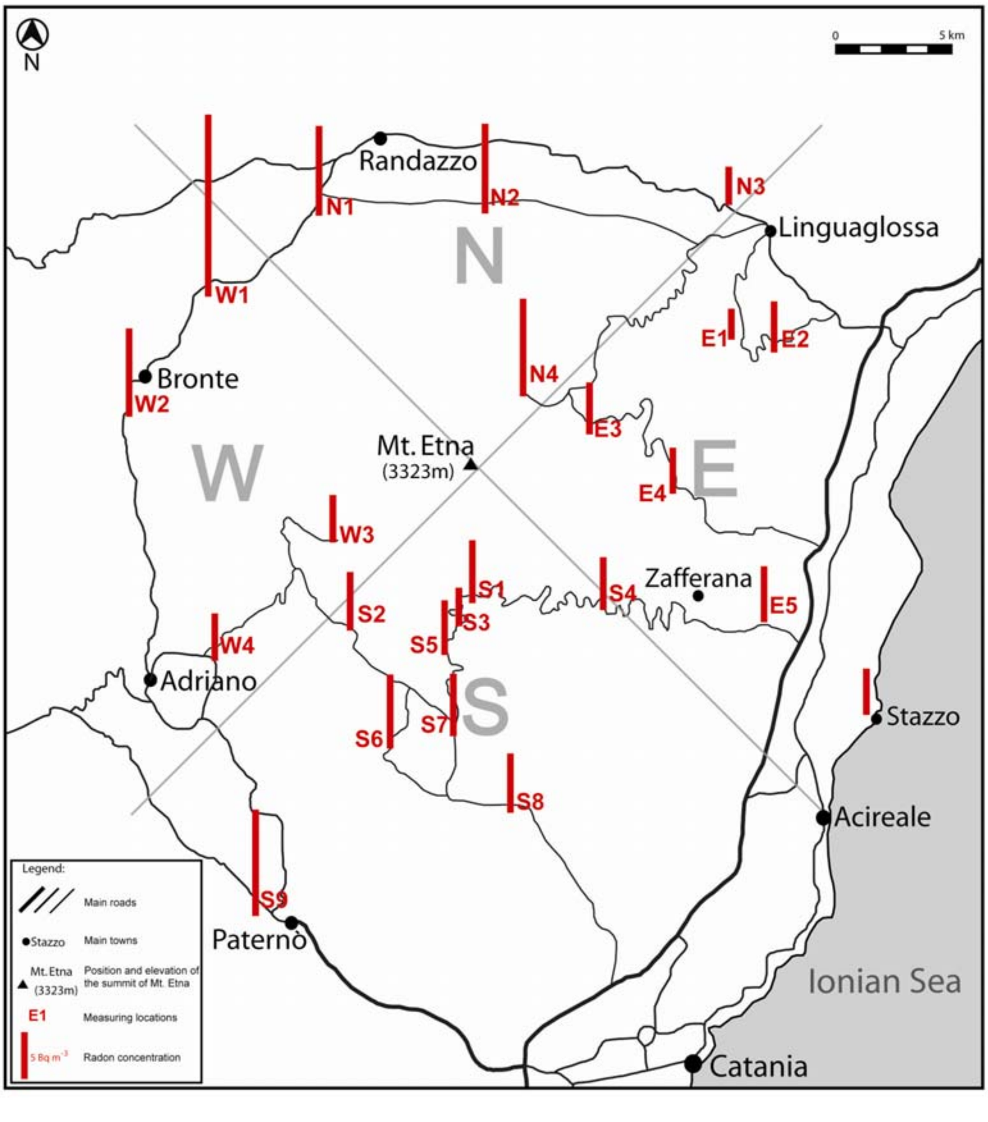

Fig. 1. Map of north-east Sicily with the area around Mt. Etna where radon was surveyed in outdoor air; the bars at measurement points represent values of radon concentration, grouped into north (N), west (W), east (E) and south (S) sections.

from one measurement point to another, during the daily periods between 10 a.m. and 6 p.m. At each point, 4–6 measurements of 20 min each were performed. The instrument inlets were 20 cm above the ground. All measurements were carried out under relatively stable weather conditions in the period September 30 – October 7, 2008, except measurements at Zafferana and at three points close to the summit, and above sea water near Stazzo, in October 2006 and June 2007, respectively. The lower limit of detection was 1–2 Bq·m^{-3} for 20 min measurements and experimental error 1–2 Bq·m^{-3} for levels below 4 Bq·m^{-3}, and about 1 Bq·m^{-3} for levels above that.

Both instruments were recently calibrated in the Radon Chamber of the Laboratory of Radiometric Expertise at the Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, Kraków, Poland [20]. Overnight measurements with both instruments at Stazzo showed a difference of less than 1–2 Bq·m^{-3} between two readings.

Results and discussion

Results are grouped in northern (N), western (W), eastern (E) and southern (S) sections of the area and are shown both in Table 1 and Fig. 1. The average radon concentration for the eastern section is the lowest (4.8 Bq·m^{-3}) and that for the western, the highest (9.6 Bq·m^{-3}), while those for the northern section and southern are between 7.9 Bq·m^{-3} and 6.5 Bq·m^{-3}, reflecting the concentration gradient from the continent towards the Ionian Sea coast. In the western section also the scattering of values was the highest. Measurements at three points close to the summit in October 2006 had shown values of up to 93 Bq·m^{-3}. As expected, the dilution of outdoor air over the flanks of Mt. Etna, with possibly enhanced radon levels, by the air masses from above the sea, with low radon levels (average of 3.3 Bq·m^{-3} during daily period, measured in 2007), appears to be minimal at the western points, the most distant from the coast. This is in contrast to the radon levels found in groundwater, which are the highest in the eastern part [9]. The number of measurement points in this study was too low to enable us to determine whether the higher radon concentrations in outdoor air coincide with the higher values in indoor air observed in proximity to the faults [2].

Bearing in mind the average radon concentrations of 15 Bq·m^{-3} in winter and 5 Bq·m^{-3} in summer in Milan, Italy, obtained during a long-term radon monitoring [29], the volcanic activity of Mt. Etna does not appear to enhance radon levels in outdoor air over a wide area, and its influence is localized to its summit. Values are even lower than those obtained in Slovenia (in the range 3.2–47.2 Bq·m^{-3} [30]), in Munich, Germany (average radon equilibrium equivalent concentration (EEC) of 6.9 Bq·m^{-3} [14]), in Cyprus (average radon concentration of 11 Bq·m^{-3} [27]), and Bucharest, Romania (radon progeny concentration in the range of 5–22 Bq·m^{-3} [1]). They are, however, similar to the levels observed at 13 places in the Czech Republic, with radon progeny concentrations in the range of 1–3 Bq·m^{-3} [3].

Figures 2a and 2b show time series of radon concentration, air temperature and barometric pressure monitored permanently at Stazzo (5 m a.s.l.) in this survey and at Zafferana (756 m a.s.l.) in a previous survey. Radon peak on June 12 in Fig. 2b refers to the points close to the summit. A typical diurnal variation of radon concentration is obvious, although more pronounced at Stazzo than at Zafferana, with minima during hot day

Table 1. Radon concentrations in outdoor air (C_{Rn}) and their average values in the Mt. Etna area, grouped in northern (N), eastern (E), western (W), and southern (S) section (see Fig. 1)

Place code	C_{Rn} (Bq·m^{-3})
N-1	8.9 ± 1.5
N-2	8.9 ± 1.4
N-3	3.8 ± 1.1
N-4	10.0 ± 3.1
average	7.9 ± 2.8
E-1	3.0 ± 1.1
E-2	5.2 ± 2.1
E-3	5.3 ± 1.8
E-4	4.6 ± 0.7
E-5	5.7 ± 2.1
average	4.8 ± 1.1
W-1	19.6 ± 3.7
W-2	9.3 ± 1.5
W-3	4.8 ± 1.6
W-4	4.8 ± 1.4
average	9.6 ± 7.0
S-1	6.5 ± 0.6
S-2	6.0 ± 1.9
S-3	3.8 ± 1.4
S-4	5.4 ± 2.1
S-5	5.6 ± 1.3
S-6	7.7 ± 2.7
S-7	6.5 ± 1.8
S-8	6.1 ± 1.7
S-9	11.3 ± 1.3
average	6.5 ± 2.1

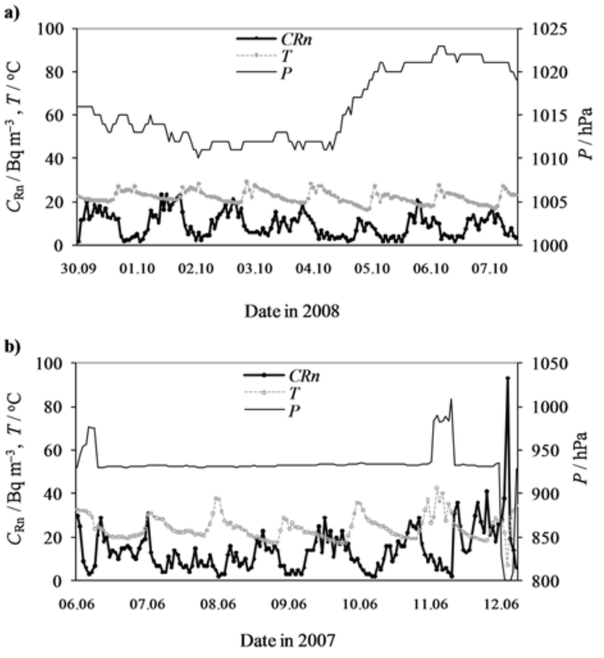

Fig. 2. Time series of radon concentration, air temperature and barometric pressure, obtained using AlphaGuard monitors, at (a) Stazzo and (b) Zafferana.

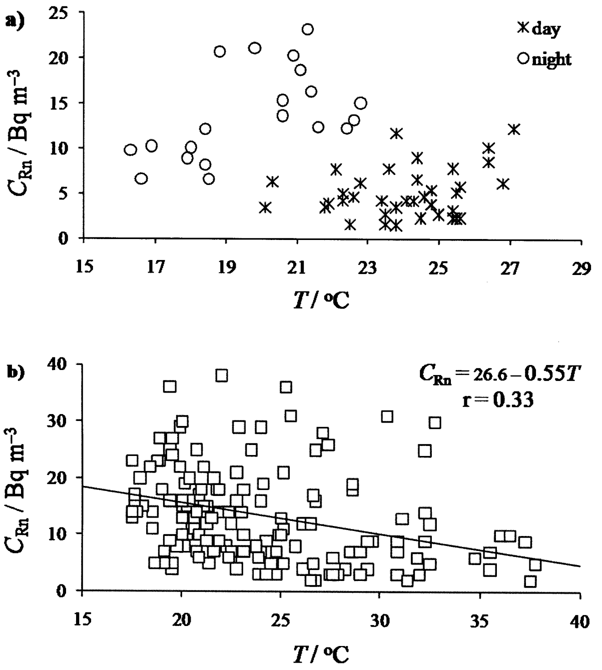

Fig. 3. Relationship between radon concentration in outdoor air and air temperature, obtained using AlphaGuard monitors, at (a) Stazzo and (b) Zafferana.

time and maxima during fresh nights, which is reflected in a weak correlation between concentration and temperature at both places (Fig. 3). At Stazzo (Fig. 3a), two clusters with centroid values of 14.1±3.5 Bq·m^{-3} during night and 5.2±2.8 Bq·m^{-3} during day time, as observed using the Simple *k*-means clustering algorithm [12], are observed rather than a straight line. The average radon concentration at Stazzo during the entire period of measurement was 9.3±2.3 Bq·m^{-3}, and 4.3±1.0 Bq·m^{-3} during the daily periods from 10 a.m. to 6 p.m. (time of measurements at flanks). The latter average may not be simply compared to those obtained at the flanks and is, therefore, not included in the calculation of the average concentration for E-section, because the AlphaGuard monitor at Stazzo was operated under different conditions: it was placed on a desk in a house terrace with a tiled floor, beneath a balcony, open towards the house interior. Neither can averages for the whole period at Stazzo of 9.3±2.3 Bq·m^{-3} and at Zafferana of 13.2±2.9 Bq·m^{-3} be compared, because of different meteorological conditions during the two periods of measurement.

The dependence of radon concentration on barometric pressure was checked by comparing their time gradients, which are expected to be of opposite signs: during a pressure decrease, gas is sucked out of the ground and radon concentration in outdoor air is enhanced [19]. This may not be the case during increased seismic activity when the transport of radon is perturbed, and periods of equal signs of both gradients may be considered as radon anomaly related to earthquakes [32]. Nonetheless, in about 9% of periods without earthquakes, gradients of the same sign were observed, possibly because in these periods the influence of pressure on radon transport was overcome by the action of other environmental parameters [32]. In Fig. 4, the time series of $\Delta C_{Rn}/\Delta t$ and $\Delta P/\Delta t$ are shown

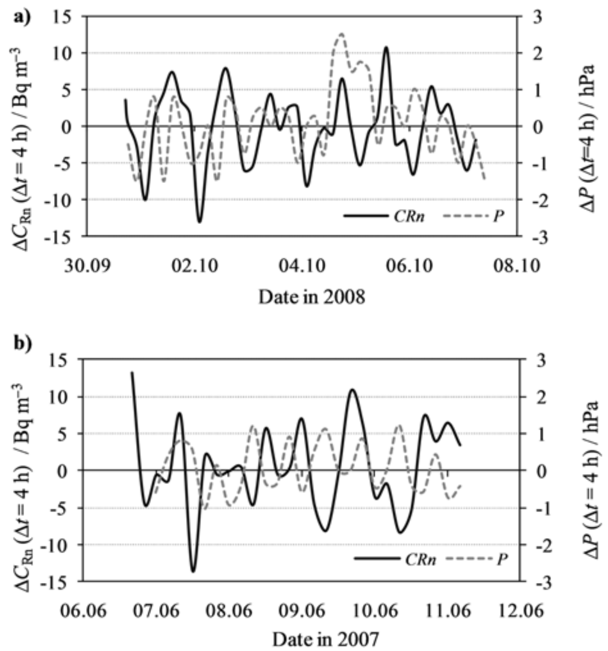

Fig. 4. Time series of time gradients of radon concentration (ΔC_{Rn}) and barometric pressure (ΔP), with Δt = 4 h, obtained using AlphaGuard monitors, at (a) Stazzo and (b) Zafferana.

for Stazzo and Zafferana, with Δt = 4 h. Periods when both gradients have the same sign amounted to about 35% of the total time at both Stazzo and Zafferana. One may speculate that this high percentage is a result of volcanic activity and the presence of active faults in the area.

Conclusion

Radon activity concentrations in outdoor air at 22 points in the Mt. Etna area were in the range of 3.0–19.6 Bq·m^{-3}. The highest average value (9.6 Bq·m^{-3}) was obtained for the western section in the inland, and lowest (4.8 Bq·m^{-3}) in the eastern section, oriented towards the Ionian Sea coast. At three points close to the summit, at an altitude of about 3000 m above the sea level, values of up to 93 Bq·m^{-3} were found. Based on these preliminary results, it has not been possible to confirm elevated values in proximity to tectonic faults. Nonetheless, based on high percentage of periods when time gradients of both radon concentration and barometric pressure have the same sign, one may speculate that in this area radon transport is affected by volcanic and tectonic activities. A continuation of the study is planned, with a targeted network of measurement points.

Acknowledgment. The study was financed by the Slovenian Research Agency and the Ministry of Foreign Affairs of Italy within the bilateral collaboration no. BI-IT/05-08-027.

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