Soil Gas Radon Concentration and Permeability at “Valle della Caffarella” Test Site (Roma, Italy). Evaluation of Gas Sampling Techniques and Radon Measurements Using Different Approaches

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Abstract

Soil gas radon concentration at “Valle della Caffarella” test site (Roma, Italy) was monitored in December 2009 and June 2010, using three different gas sampling and measurement techniques (alpha spectrometry, scintillation cells and ionization chambers). Gas permeability of soil was associated to radon measurements to evaluate its effects on soil radon results.
During the winter campaign, the three methods provided comparable results at depths of 50 and 80 cm (stations in areas A and B), where the gas permeability is low. Stations in area C apparently do not respond to this simple model. Determinations carried out at depths of 20 and 30 cm are generally 60 % lower using the continuous radon monitor (alpha spectrometry device), because larger volumes of air are extracted using the built-in pump. The proximity to soil/air interface probably drains atmospheric air into the subsoil and radon-poor air is then delivered to the instrument. This effect is enhanced by larger gas permeability. Results from the summer campaign substantially mirror those of the winter fieldwork, even if soil radon concentrations are strongly reduced both at 30 and 80 cm depth because of enhanced radon release to the atmosphere when soil is drier, warmer and more permeable. The recourse to a mixed approach (Radon v.o.s. probe + RAD7 continuous monitor) shows that the volume of extracted air affects soil radon results less than the combined effect of the probe tip size and the poor sealing of the sampling hole.

Introduction

Radon is the second cause of lung cancer in the general population, after smoking. Epidemiological studies have provided convincing evidence of an association between indoor radon exposure and lung cancer, even at the relatively low radon levels commonly found in residential buildings. A number of approaches is used worldwide to assess the potential for elevated indoor radon concentrations across geographic areas of various dimensions. Radon-prone areas can be identified directly by using indoor measurements or indirectly using radon concentration in the soil, provided there is an established correlation with the radon concentrations in homes. The United States of America developed its radon map based on a combination of indoor measurements, geological characteristics, aerial radioactivity, soil permeability and foundation type (USEPA 1993). In Germany, the map is based on radon concentrations in soil gas. In Austria, the classification is based upon the mean radon concentration within a given area (Friedmann 2005). Another approach used in some countries, such as the Czech Republic (Neznal et al. 2004), involves testing individual building sites prior to construction to establish a radon index for the site. The index is then used to define the degree of radon protection needed for building on that site. However, in countries including Finland, Ireland, Norway, Sweden, Switzerland, the United Kingdom, and the United States of America, the most cost-effective approach appears to be the use of radon control options in all new homes (WHO 2007). Sometimes this approach is restricted to radon-prone areas. In Italy local administrations are recommended to introduce in urbanistic instruments specific instructions to adopt simple and cheap building options aimed at reducing radon concentration in the subsoil before construction begins. Similar recommendation should also apply in case of building restoration. Nonetheless, the control of radon concentration in building and subsoil is carried out in many Italian areas characterised by fracturation and faulting.

Here, we compare soil radon measurements carried out using a commercial probe, distributed by Durridge Co. and connected to a continuous radon monitor (RAD 7, Durridge Co.) with determinations making use of grab soil air samples delivered to scintillation cells or ionization chambers. Permeability measurements are associated to radon determinations in order to assess the ease with which soil transmits air. Since soil radon concentration and permeability varies seasonally and are influenced by meteo-climatic factors like temperature and precipitation resulting in different physical properties of the soil (temperature, moisture content, permeability), two campaigns were carried out in December 2009 and June 2010 at "Valle della Caffarella" test site (Roma, Italy).

Geological background

Roma (Figure 1) develops on a volcanic-derived background referred to the activity of Colli Albani and Sabatini districts, located SE and NW of the city, respectively. The outcropping products are mainly Quaternary ignimbrites, with the presence of lava flows in the south-easternmost periphery. Tevere River flows across the municipal territory in NS direction and receives the input of some tributaries, the main of which is Aniene River in the NE quadrant. The floodplains of Tevere and Aniene mainly consist of fine-grained alluvial deposits alternated with sands and peat levels at depth as results from drillings stratigraphy (Campolunghi et al., 2007). The pre-volcanic sedimentary basement consists of Plio-Pleistocene marine to transitional deposits outcropping in the NW quadrant (Bozzano et al., 2006).

"Valle della Caffarella" test site

Valle della Caffarella is located in the SE area of the city (Figure 1) and its geological background is represented by ignimbrites and alluvial deposits. Ignimbrites were erupted from Colli Albani volcano (central Italy) between 460 and 350 ka BP (Soligo & Tuccimei, 2010). They belong to the fourth lithosome of the volcano, the Vulcano Laziale caldera complex, namely to Pozzolane Rosse and Villa Senni (Tufo Lionato + Pozzolanelle ignimbrites) lithostratigraphic units (Giordano et al., 2010). The alluvial deposits of Valle della Caffarella stream (Almone river) are characterised by an average thickness of around 30 metres. The upper portion consists of 3 m thick clayey silts derived from historical floods covering silty clays that represents the main body within the alluvial sediments (Campolunghi et al., 2007).

The bedrock of measuring stations is Pozzolanelle ignimbrite, characterised by a matrix (60-90%) made by coarse-ash shard and abundant crystal fragments of leucite, clinopyroxene and biotite. The framework mainly consists of highly porphyritic scoria and minor xenoliths of lava, skarn and intrusives. The composition is tephritic-phonolothic to phonolithic-tephritic (Freda et al., 1997).

In the last two years, a permanent station to measure soil radon concentration has been working at Caffarella Valley, recording monthly radon fluctuations at 30 and 80 cm depth, as well as climatic data (temperature and precipitation). During the winter, when soil humidity is higher and soil temperature lower, radon gas is forced in a partially sealed subsoil and reach

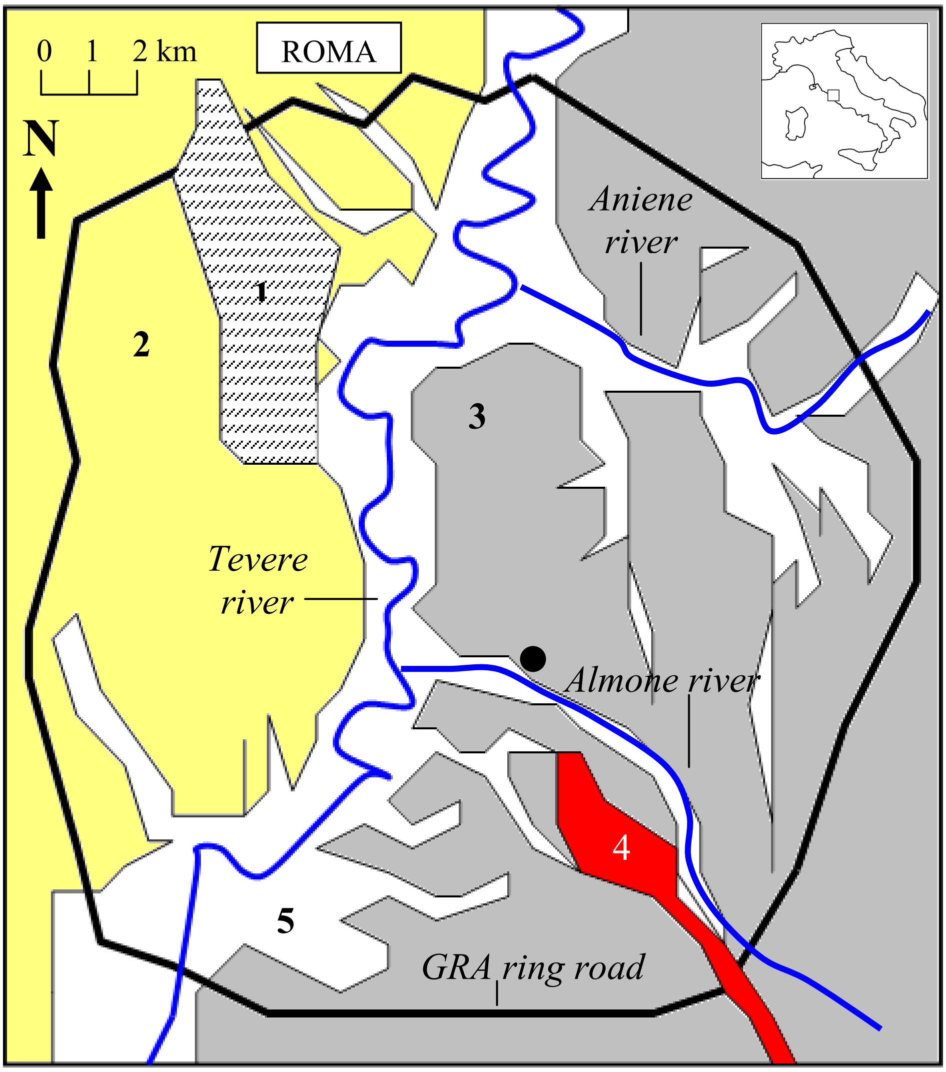

Figure 1. Simplified geological map of Roma (Italy). 1 Plio-Pleistocene marine to transitional deposits, 2 Sabatini district volcanites, 3 Colli Albani district ignimbrites, 4 Colli Albani district lavas, 5 Alluvial sediments of Tevere River and its tributaries. Closed circle stand for soil radon and permeability measuring station in the site of Valle della Caffarella.

maximum values, whereas in summer scarce precipitations and high temperature favour gas release to the atmosphere leading to minimum values of soil radon activities (Figure 2). Soil radon concentrations at Valle della Caffarella test site have been also mapped in different seasons (Figure 3), showing overall seasonal fluctuations similar to those observed at the permanent station. The site is also used for instruction and improvement courses organised by "Roma Tre" University and regional professional associations devoted to geologists working in the field of radon risk. The last was held in June 2010.

Sampling techniques

Three areas were chosen for coupled measurements of soil radon and soil permeability. The first, labelled A, is where the above-mentioned permanent station is located; here the soil gas was collected at the fixed depth of 90 cm. The other two, labelled B and C, are mobile sampling places, at about 50 metre distance from the permanent station; sampling depths were fixed at 30 and 80 cm. In each area, a similar array of sampling stations was followed with a central radon station for soil radon measurements using the Durridge approach and three more

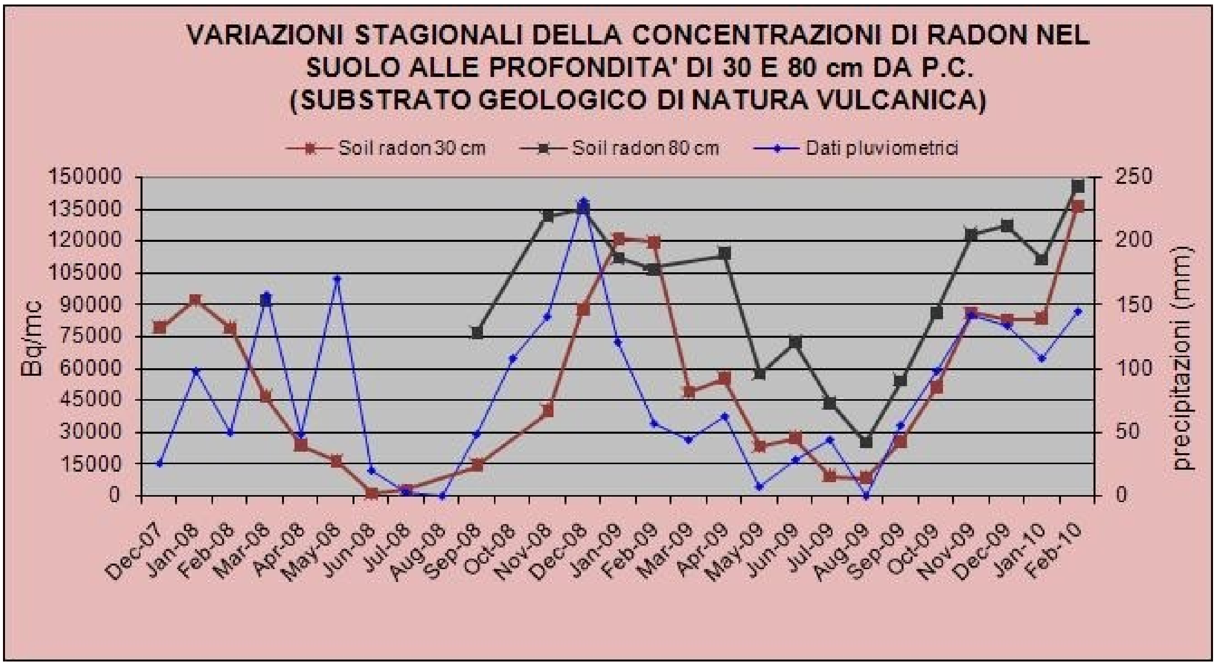

Figure 2. Soil radon fluctuations at "Valle della Caffarella" test site in the period 2008-2010.

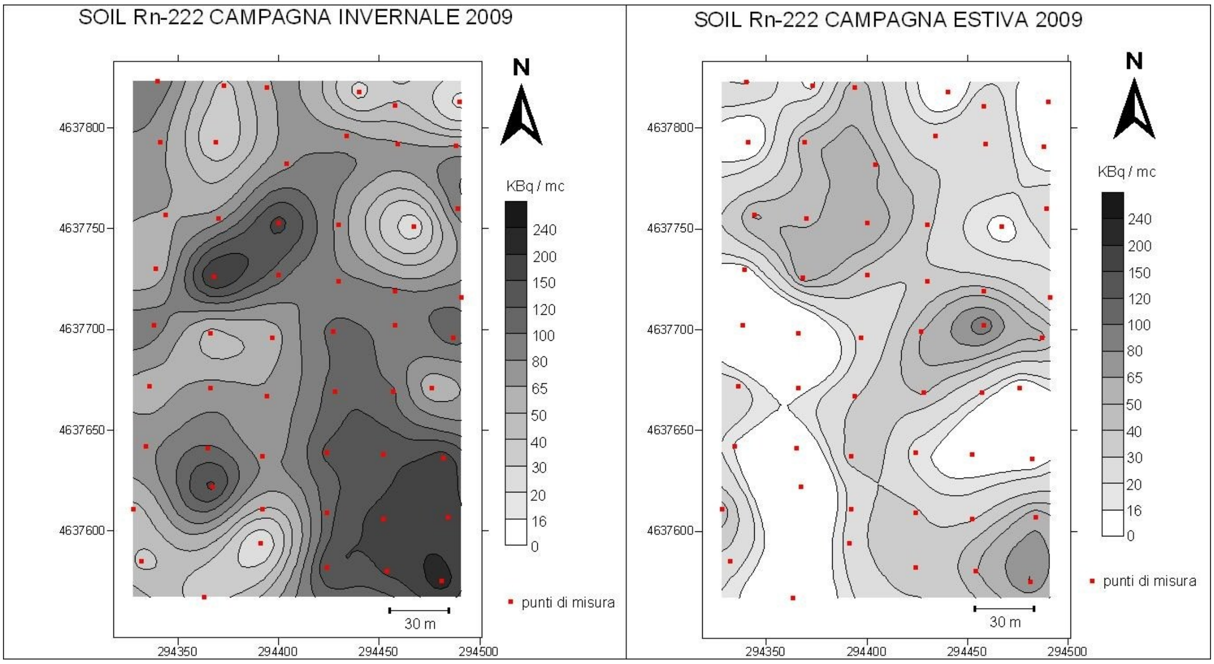

Figure 3. Soil radon mapping at Valle della Caffarella" test site in winter and summer 2009.

locations placed around the inner site at 1.10-metre distance for soil gas sampling and permeability measurements at increasing depths using the Radon v.o.s. approach (Figure 4).

Durridge approach: The soil gas probe (Durridge) hollow steel tube, 1⁄2" OD and 1⁄4" ID, was inserted to 80 cm depth, checking for a reasonable seal between the probe shaft and the surrounding soil, so that the ambient air did not descend around the probe to dilute gas sample. A water trap and a drying unit were connected between the probe head and the continuous radon monitor (RAD 7, Durridge Co.) in order to keep relative humidity below 10 %. The pump current gives good indication of soil porosity, warning when the soil permeability is low or when saturation conditions occur with obvious trouble with gas sampling and measurement.

Radon v.o.s. approach: A probe, 12 (OD) and 8 (ID) mm in diameter and 1.1 m in length is equipped with a “lost” sharp tip, 12 mm in diameter. The probe was pounded to the desired depths, 20, 50 and 80 cm at sampling area A and 30 and 80 cm at areas B and C. Proper distance ring and screw are used to extrude the tip and create an exact volume for soil permeability measurements and then for soil air sampling. The soil-gas is sucked and samples of a controlled volume (100 mL for Lucas Cell and 150 mL for ionization chambers) are collected at each depth using a large-volume syringe (150 mL).

Figure 4. Location of sampling areas (A) and array of sampling stations in each area for replicate soil permeability and soil gas sampling with different approaches (B). Numbers. 1 and 2 stand for permanent and mobile sampling areas, respectively; 3 is the inner station where the Durridge approach was tested and 4 are the sites where radon v.o.s system for soil radon monitoring and soil permeability measurements was applied.

Analytical Methods

Measurements of soil radon concentration using the continuous radon monitor

Dried soil air is delivered to the RAD7 radon monitor (Durridge Company Inc) by pumping. The instrument is equipped with a solid-state alpha detector that allows the measurement of ^{222}Rn using the activity of its daughter ^{218}Po that reaches equilibrium with the father in about 15 minutes. A single measurement has an average duration of 25 – 30 minutes, with partial readings every five minutes (cycles), and ends when the relative difference between the last two cycles (starting from the fourth, when equilibrium conditions are reached) is lower than 15 %. In this case, the final result is the average value of last two readings.

Measurements of soil radon concentration using ionization chambers

Measuring system RM - 2 is based on an ionization chamber operating in a current mode. The measuring system consists of a set of cylindrical ionization chambers IK-250 with a detection volume of 250 ml and an electrometer ERM-3 (voltage reader device). Two measuring modes are followed: (i) measurement starting 15 minutes after the transfer of the sample into the chamber and (ii) standard equilibrium measurement. Time period needed for the measurement of one sample is 120 s.

Measurements of soil radon concentration using Lucas Cells

Glass-type Lucas cells with the active volume of 125 ml in combination with the scintillometer SISIE were used for measurements. The detector response in the equilibrium state between radon and radon progeny, i.e. more than 3 hours after the transfer of the soil-gas sample into the Lucas cell, is about 2 pulses/s per 1 Bq of radon concentration deposited in the cell. Two measurements of each sample in the equilibrium state between radon and radon progeny were made using the counting time 100 s.

Measurements of gas permeability of soil

The principle of the equipment – permeameter RADON-JOK - consists of air withdrawal by means of negative pressure. Air is pumped out from the soil under constant pressure through a specially designed probe with a constant surface of contact between the probe head and the soil. The special rubber sack, with one or two weights, pumps the air from the soil and allows to perform measurements at very low pressures. The gas permeability is calculated using the known air flow through the probe.

Results and discussion

Soil radon concentrations in the winter campaign (December 2009), measured using alpha spectrometry (RAD 7) or a combination of ionization chambers and Lucas cells are reported, respectively, in Tables 1 and 2. Table 3 displays data of soil gas permeability obtained using the RADON-JOK at the same time. Results from the summer campaign (June 2010) are accordingly reported in Tables 4, 5 and 6.

Winter campaign

Measurements at depths of 50 and 80 cm

Soil radon concentrations determined using the three different methods are generally comparable (relative difference < 15 %) for analyses carried out at depth ≥ 50 cm when permeability is low (averagely $8.5 \cdot 10^{-12}$ m^2 at 50 cm and $< 3.5 \cdot 10^{-14}$ m^2 at 80 cm depth, see stations A1, A2, A3, B1, B2 and B3). Detected radon concentration range from 110 to 167 kBq/m^3 at 50 cm depth and from 124 to 203 kBq/m^3 at 80 cm and are compatible with those found at correspondent depths in the month of December in the course of last two years monitoring (Figure 2).

In station C4 does not seem to respond to this simple model because its radon concentration at 80 cm depth (102 kBq/m^3) is lower than those measured at stations C1, C2 and C3 at the same depth (147 – 230 kBq/m^3), but maybe station C is naturally characterised by smaller soil radon contents than other stations and the recorded difference is not dependent on specific sampling and measurement approach. This hypothesis is supported by the extreme spatial variability of soil radon concentration, as results from soil radon mapping in Figure 3. Another possible explanation could be related to unsatisfactory caution to tamp down the soil around the shaft, responsible for driving air down the outside of the probe tube and diluting soil radon concentration. This effect will be better described in the following section.

Measurements at depths of 20 or 30 cm

Determinations carried out at lower depth (20 and 30 cm) are more variable (from 32 to 181 kBq/m^3) than those performed at depths ≥ 50 cm. Specifically, measurements obtained according the Durridge approach are about 60 % systematically lower than others. This could be explained by the shape of the sampling hole and the worker ability to seal it. Basically, the diameter of the Durridge probe tip is wider than the rest of the tubing, so if the shaft is not adequately sealed, pumping large volumes of soil air in proximity of soil/air interface drains atmospheric air into the subsoil and radon-poor air is then delivered to the radon monitor.

Actually, RAD7 pumps a larger volume of gas if compared with that removed with the syringe; this different response to sampling techniques, evidenced close to the ground level, is enhanced when soil gas permeability is higher, as shown by permeability values at –30 cm, (about $2.5 \cdot 10^{-12}$ m^2) that are effectively higher than those found at higher depths (mostly in the range of 10^{-14} m^2).

Table 1 - Soil radon concentration at "Valle della Caffarella" test site (Roma, Italy) in December 2009, using the Durridge approach. For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - sampling depth	Soil-gas ^{222}Rn concentration (kBq/m3)	Soil-gas ^{220}Rn concentration (kBq/m3)
A - 90 cm	133	176
B - 30 cm	83	303
B - 80 cm	152	549
C - 30 cm	32	147
C - 80 cm	102	402

Table 2 - Soil radon concentration at "Valle della Caffarella" test site (Roma, Italy) in December 2009, using the Radon v.o.s approach. For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - sampling depth	**Soil-gas radon concentration (kBq/m^3)**			
	Scintillation method		**Ionization chambers**	
	first meas.	**repeated meas.**	**15 min**	**equlibrium**
A1 - 20 cm	**131**	**129**	**148**	**144**
A1 - 50 cm	**167**	**152**	**153**	**158**
A1 - 80 cm	**171**	**188**	**203**	**193**
A2 - 20 cm	**106**	**104**	**129**	**125**
A2 - 50 cm	**123**	**110**	**139**	**141**
A2 - 80 cm	**123**	**127**	**129**	**132**
A3 - 20 cm	**124**	**134**	**124**	**124**
A3 - 50 cm	**138**	**144**	**150**	**141**
A3 - 80 cm	**147**	**142**	**139**	**145**
B1 - 30 cm	**140**	**134**		
B1 - 80 cm	**175**	**170**		
B2 - 30 cm	**156**	**151**		
B2 - 80 cm	**193**	**191**		
B3 - 30 cm	**132**	**128**		
B3 - 80 cm	**127**	**124**		
C1 - 30 cm	**161**	**163**		
C1 - 80 cm	**188**	**183**		
C2 - 30 cm	**180**	**181**		
C2 - 80 cm	**230**	**225**		
C3 - 30 cm	**147**	**147**		
C3 - 80 cm	**212**	**206**		

Table 3 - Soil gas permeability at “Valle della Caffarella” test site (Roma, Italy) in December 2009, using the Radon JOK. For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - depth	**number of weights**	**time (s)**	**soil permeability (m^2)**
A1 - 20 cm	1	68	**$1.6 . 10^{-12}$**
A1 - 50 cm	1	23	**$4.7 . 10^{-12}$**
A1 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
A2 - 20 cm	2	1575	**$4.0 . 10^{-14}$**
A2 - 50 cm	1	11	**$9.9 . 10^{-12}$**
A2 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
A3 - 20 cm	2	37	**$1.7 . 10^{-12}$**
A3 - 50 cm	1	10	**$1.1 . 10^{-11}$**
A3 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
B1 - 30 cm	1	70	**$1.6 . 10^{-12}$**
B1 - 80 cm	2	72	**$8.7 . 10^{-13}$**
B2 - 30 cm	1	22	**$4.9 . 10^{-12}$**
B2 - 80 cm	2	373	**$1.7 . 10^{-13}$**
B3 - 30 cm	1	89	**$1.2 . 10^{-12}$**
B3 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
C1 - 30 cm	1	60	**$1.8 . 10^{-12}$**
C1 - 80 cm	1	28	**$3.9 . 10^{-12}$**
C2 - 30 cm	1	31	**$3.5 . 10^{-12}$**
C2 - 80 cm	2	1240	**$5.1 . 10^{-14}$**
C3 - 30 cm	1	51	**$2.1 . 10^{-12}$**
C3 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**

Summer campaign

In June 2010, soil radon concentration and soil gas permeability measurements were repeated in the same sampling areas (A, B and C) with the same array; sampling stations were labelled with new progressive numbers (4, 5 and 6). A new sampling and analytical approach was tested in station C4, jointly with the Radon v.o.s. approach. This mixed approach makes use of the Radon v.o.s. probe connected to the RAD7 continuous monitor. The choice of a probe, whose tip is as large as the tube, prevents the flow of atmospheric air down the outside of the shaft. Large volume of air are still extracted by pumping in order to evaluate this effect on soil radon concentration, when no room is left between the probe and the surrounding soil.

Measurements at depths of 80 cm

Soil radon concentrations determined using the three different methods are always comparable (relative difference around 5 %). This good data agreement is much better than in the winter campaign because stronger care was adopted to seal the sampling hole at surface. Detected radon concentrations range from about 40 to 160 kBq/m^3 and are compatible with those found at correspondent depths in the month of June the course of last two years monitoring (Figure 2).

At station C4, the comparison shows substantially the same results between measurements carried out pumping air from the same probe (the Radon v.o.s. probe). Ionization chambers data (112-115 kBq/m^3) are more similar to those obtained with the continuous radon monitor (108 kBq/m^3) while Lucas Cells outcomes are different. In particular, the air sample sucked before the measurement with RAD7 gave higher radon concentration (128 kBq/m^3) than that collected after (88.5 kBq/m^3). This could depend on the order of gas sampling for Lucas Cell

Table 4 – Soil radon concentration at “Valle della Caffarella” test site (Roma, Italy) in June 2010, using Durridge approach. In sampling area C, the central station was not considered, while in C4 sampling site, a modified version of the Durridge approach was tested (see text for explanation). For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - sampling depth	Soil-gas ^{222}Rn concentration (kBq/m^3)	Soil-gas ^{220}Rn concentration (kBq/m^3)
A - 90 cm	60	-
B - 30 cm	18,5	-
B - 80 cm	40	-
C4 - 30 cm	48	-
C4 - 80 cm	108	-

Table 5 - Soil radon concentration at “Valle della Caffarella” test site (Roma, Italy) in June 2010, using the Radon v.o.s approach. For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - sampling depth	Soil-gas radon concentration (kBq/m^3)		
	Scintillation method	Ionization chambers	
		15 min	equlibrium
A4 - 30 cm	37.1	36.4	36.2
A4 - 80 cm	82.9	91.5	86.8
A5 - 30 cm	34.5	31.5	35.0
A5 - 80 cm	73.7	73.9	69.1
A6 - 30 cm	30.6	27.3	27.7
A6 - 80 (70 - 80) cm*	36.1	-	39.9
A - 90 cm	57.1	-	-
B4 - 30 cm	39.3	40.1	36.1
B4 - 80 (65 - 80) cm*	72.6	62.6	65.2
B5 - 30 cm	41.2	40.1	45.3
B5 - 80 (65 - 80) cm*	57.7	80.4	81.7
B6 - 30 cm	45.6	36.5	40.7
B6 - 30 cm (repeated sampling)	42.8	-	-
B6 - 80 cm	171	157	152
C4 - 30 cm	47.8	50.5	43.7
C4 - 80 cm (before)**	128	115	112
C4 - 80 cm (after)**	88.5	115	113
C5 - 30 cm	46.2	43.5	40.0
C5 - 80 cm	92.1	81.2	80.4
C6 - 30 cm	29.9	27.7	29.1
C6 - 80 cm	80.1	69.0	70.6

*) very low permeability of soil - the probe was retracted to the surface to enlarge the cavity at the lower end of the sampling probe before soil-gas sampling

**) before = before measurement of the Italian team; after = after measurement of the Italian team

Table 6 - Soil gas permeability at “Valle della Caffarella” test site (Roma, Italy) in June 2010, using the Radon JOK. For location of sampling areas and configuration of sampling stations, see Figure 4.

Measuring point - depth	**number of weights**	**time (s)**	**soil permeability (m^2)**
A4 - 30 cm	1	15	**$7.2 . 10^{-12}$**
A4 - 80 cm	2	91	**$6.9 . 10^{-13}$**
A5 - 30 cm	1	21	**$5.2 . 10^{-12}$**
A5 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
A6 - 30 cm	1	12	**$9.1 . 10^{-12}$**
A6 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
A - 90 cm	1	7.5	**$1.4 . 10^{-11}$**
B4 - 30 cm	1	23	**$4.7 . 10^{-12}$**
B4 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
B5 - 30 cm	1	12	**$9.1 . 10^{-12}$**
B5 - 80 cm	2	>1800	**$<3.5 . 10^{-14}$**
B6 - 30 cm	1	19	**$5.7 . 10^{-12}$**
B6 - 80 cm	2	1567	**$4.0 . 10^{-14}$**
C4 - 30 cm	1	31	**$3.5 . 10^{-12}$**
C4 - 80 cm	2	503	**$1.3 . 10^{-13}$**
C5 - 30 cm	1	14	**$7.8 . 10^{-12}$**
C5 - 80 cm	1	16	**$6.8 . 10^{-12}$**
C6 - 30 cm	1	31	**$3.5 . 10^{-12}$**
C6 - 80 cm	2	216	**$2.9 . 10^{-13}$**

and ionization chambers analyses. The former is the first to be extracted and could be affected by some disequilibrium or instability in a low permeable soil that disappear later when soil air is pumped for following measurements.

Measurements at depths of 30 cm
Determinations carried out at 30 cm range from 18 to 50 kBq/m^3. Differences among sampling and analytical methods are strongly reduced. Soil gas permeability data show a strong increase of permeability and explain the reduced values of soil radon detected in summer. C4 station data correspond within the error range, using the three different methods and sampling air from the same probe.

Conclusions

During the winter campaign, soil radon concentrations seem to be affected by the sampling technique. Using the Durridge approach, if the ground is not tamped down around the Durridge probe, pumping may drive atmospheric air to flow down along the outside of the shaft, causing a dilution of soil radon concentration. This process is more significant if the sampling depth is shallow and the gas permeability is high.

Furthermore, the three analytical methods (alpha spectrometry, scintillation cells and ionization chambers) do not affect the results of soil radon concentration and can be considered comparable.

Results from the summer campaign substantially mirror those of the winter fieldwork, even if soil radon concentration are strongly reduced both at 30 and 80 cm depth because of enhanced radon release to the atmosphere when soil is drier, warmer and more permeable. The recourse to the mixed approach (Radon v.o.s. probe + RAD7 continuous monitor) shows that the volume of extracted air affects soil radon results less than the combined effect of the probe tip size and the poor sealing of the sampling hole.

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