


# Multi-parametric approach to the analysis of soil radon gas for its validation as geindicator in two sites of the Phlegrean Fields caldera (Italy)

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**Abstract** Radon is considered one of the short- and immediate-term earthquake precursors by International Association of Seismology and Physics of the Earth's Interior. For this reason, the accurate and reliable measurement of its specific activity in soil gas is mandatory. Since its presence is influenced by many environmental factors, in order to eliminate this influence on the radon activity measurement a multi-parametric and multi-site analysis is necessary. Following this approach, a continuous radon monitoring was carried out in two sites of the Phlegrean Fields caldera (Campania, Italy) using the RaMonA system, which also allows for the measurement of temperature, relative humidity and pressure. In particular, the dependence of measured radon specific activity on the meteorological conditions was studied using the multiple linear regression method. The analysis was implemented also by testing data averages on different timescales. In this paper, the results of a preliminary analysis performed on data collected throughout 2012 are presented; the outcomes obtained allow to emphasize site-specific behavior and to discriminate apparent radon "anomalies" of climatic origin from those tied to the phenomena occurring in the earth's crust. The method

employed showed its usefulness by removing the effects of meteorological conditions and thus to better identify the possible radon anomalies caused by seismo-volcanic activity. The peculiarities of the different sites and some correlations with earthquakes are discussed.

**Keywords** Radon continuous monitoring · Multi-parametric analysis · Radon as geindicator · Phlegrean Fields caldera

## Introduction

Radon and thoron are naturally occurring radioactive gases generated by the decay of  $^{226}\text{Rn}$  ( $^{238}\text{U}$  series) and  $^{224}\text{Ra}$  ( $^{232}\text{Th}$  series), respectively, present in varied quantities in the earth's crust.

The property of this noble gas and its radioactive behavior make  $^{222}\text{Rn}$  easy to be monitored, and short-term changes in its specific activity in soil can be followed with high time resolution. The  $^{220}\text{Rn}$  observation, despite its shorter half-life, also produces useful information on the radon main isotope.

Consequently, an important aspect of radon and thoron monitoring could be their implication in seismo-volcanic activity, since their presence in the soil gas is due to the status of the rock stress, which surely increases when seismic and/or volcanic episodes occur.

Some models explain changes in soil radon concentration as a consequence of strain changes inside the earth's crust before earthquake occurrence (Ghosh et al. 2009; Namvaran and Negarestani 2013; Petraki et al. 2015; Zoran et al. 2012).

If it is right to assess that the soil radon monitoring is surely a very important research field from a geological

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point of view, it is likewise true that one of the conditions necessary to carry out a successful study is to provide a long-term record of data so that the radon signal can be effectively evaluated and all noise sources are accurately eliminated.

Moreover, it is well known that environmental conditions act as a further source of complexity on radon transport and emission since changes in meteorological conditions affect radon exhalation from soil. Iskandar et al. (2004) through an experimental study obtained a relation between radon emanation power and soil temperature; they found that radon emanation increases as the soil temperature increases both for dry and wet soils. Walia et al. (2005) carried out a qualitative evaluation of the influence of the main meteorological properties on radon concentration establishing that temperature, rainfall and relative humidity have positive correlation with radon exhalation coefficients. Planinić et al. (2001) thanks to in situ measurement found a negative correlation between soil radon concentration and atmospheric pressure.

Therefore, it is mandatory to differentiate the changes that are due to phenomena occurring in the earth's crust from those depending on temperature, pressure, humidity and rainfall.

The ultimate goal of this work is the development of a program to validate a reliable methodology in which the radon signal can be used correctly as a suitable "geoinicator."

In order to reach this result, various steps and different approaches are to be considered; hence, starting from the available data, a multi-parametric analysis was chosen and carried out which aimed to highlight site-specific behavior of the different sites mainly with respect to meteorological features.

Then, once the changes depending on temperature, pressure, humidity and rainfall were highlighted and eliminated from those due to phenomena occurring in the earth's crust, an attempt was made toward the assessment of correlations between possible radon anomalies and earthquakes.

In this contest, a collaboration was begun in 2010 between the Naples sections of INFN (National Institute of Nuclear Physics) and OV-INGV (Vesuvius Observatory—National Institute of Geophysics and Volcanology) (De Cicco et al. 2010).

The general plan of this program consists of:

The real-time monitoring of radon specific activity and of the climatic characterization of many sites of the selected area;

The radiometric characterization of the soil in the seismo-volcanic sites;

The comparison of the radiometric data with those by other geochemical and geophysical measurements.

The instrument adopted for the real-time radon monitoring is the RaMonA system (Roca et al. 2004). Up to now, just two sites have been equipped with RaMonA named MSA and OLB; the last site belongs to the monitoring network of INGV located inside the Phlegrean Fields caldera.

In this work, the results of a preliminary analysis of the temporal sequences of radon data collected during 2012 at the two sites are discussed.

## Materials and methods

### Sites description and measurement setup

The Phlegrean Fields caldera is a complex geological structure; it is located on a NE–SW-trending structure in the Campanian Plain that includes the Phlegrean Fields, the city of Naples, the volcanic islands of Procida and Ischia, and the northwestern Gulf of Naples.

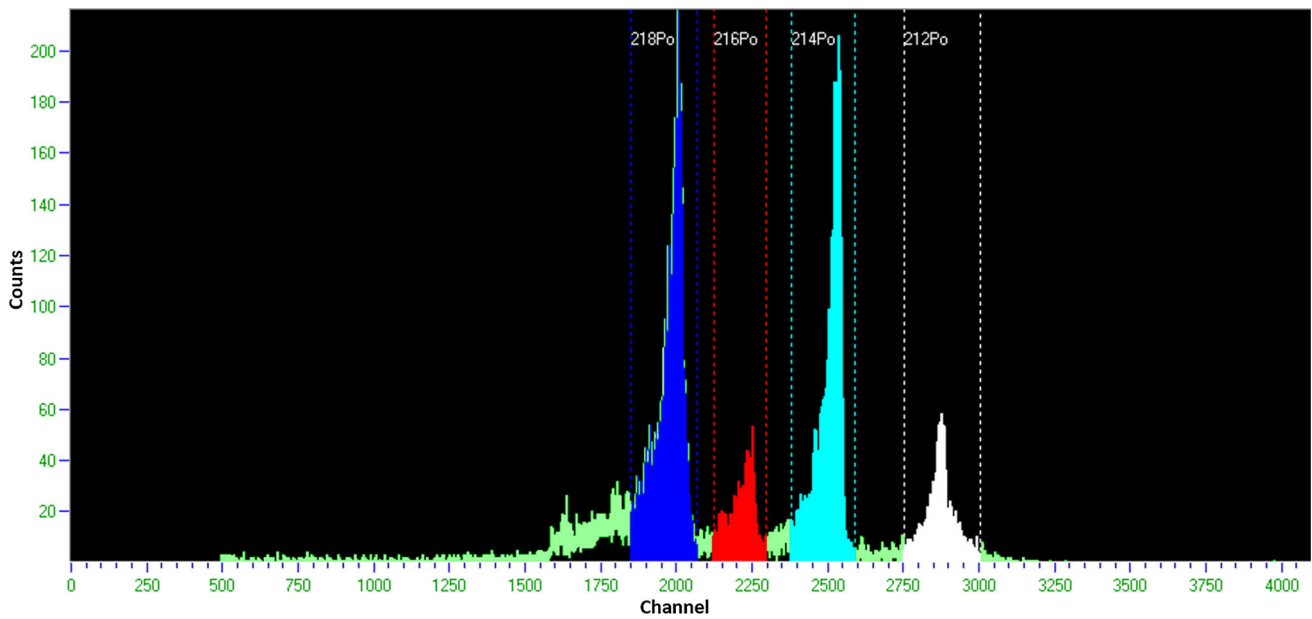
The structural setting of the Phlegrean Fields activity is due to the interaction between tectonics and volcano-tectonic activity. Tectonic activity is carried out through fault systems trending NE–SW and NW–SE that influence the distribution of volcanic activity throughout the Phlegrean district (Piochi et al. 2005). The volcano-tectonic structure is dominated by the presence of a caldera created by a collapse due to high-magnitude eruptions. According to surface and subsurface geological surveys and to archaeological investigations, it has been found that the ground deformation does not occur evenly, but most likely through the vertical displacement of a discrete number of blocks (Orsi et al. 1996).

The volcanic and strain history of the Phlegrean Fields caldera was dominated by two large explosive eruptions:

1. The Ignimbrite Campana eruption about 37,000 years B.C. This is the largest pyroclastic deposit in the Campania region (Barberi et al. 1978); it covers an area of approximately 30,000 km<sup>2</sup> with an estimated volume of erupted magma of about 150 km<sup>3</sup> (Civetta et al. 1997);
2. The Neapolitan Yellow Tuff eruption was the last major eruption occurred about 12,000 years B.C. (Di Vito et al. 1999), forming a smaller caldera inside the main one, centered on the town of Pozzuoli. This event produced the Neapolitan Yellow Tuff, referring to the characteristic yellow rocks present there.

Such eruptions are related to two episodes of collapse that, overlapping, generated the complex caldera that is the most prominent feature of the Phlegrean volcanic district.

A third phase exists dated between 8000 and 500 years ago, marked by an initial activity in the southwest area of



**Fig. 1** Typical alpha spectrum by RaMona

**Table 1** Statistical parameters of analysis carried out on MSA and OLB stations data series

	MSA station statistical parameters				OLB station statistical parameters			
	Mean value	Min	Max	Range	Mean value	Min	Max	Range
C(Rn) [kBq/m <sup>3</sup> ]	(13.0 ± 0.3)	7.3	39.6	32.2	(46.1 ± 1.2)	7.8	134.2	126.5
T (°C)	(27.2 ± 0.3)	16.1	38.4	22.3	(31.2 ± 0.1)	28.7	33.6	4.9
RH (%)	(63.4 ± 0.2)	54.6	68.7	14.1	(81.3 ± 0.1)	79.2	83.1	3.9
P (hPa)	(1043.63 ± 0.3)	1020.5	1059.4	38.9	(1019.0 ± 0.3)	992.0	1033.6	41.6

Bacoli and Baia (10,000–8000 years ago), an intermediate activity in an area concentrated between Pozzuoli, the Montagna Spaccata and Agnano (8000–3900 years ago) and a relatively newer activity which culminated in 1538 with the eruption which brought the formation of Monte Nuovo cone, from which the name (Di Vito et al. 1987; D’Oriano et al. 2005; Parascandola 1947). This phase was characterized by white “pozzolana,” the material that forms most of the volcanoes of the Phlegrean system.

A recent deformative activity with seismic swarms has been observed since the early 1970: It is the well-known phenomenon of bradyseism that is a slow movement of soil.

Other swarm events of more modest energy and number were recorded in 1989 (7.3 cm of uplift), 1994 (1.2 cm of uplift), March–August 2000 (5.2 cm of uplift), from November 2004 to December 2006 (5.1 cm of uplift) and from September 2008 to July 2009 (2.2 cm of uplift).

Therefore, the rich geodynamical history that characterizes the Phlegrean area makes it an ideal stage for experimenting radon monitoring techniques and data analysis methods.

So far, measurements have been taken just in two sites, codified as MSA and OLB.

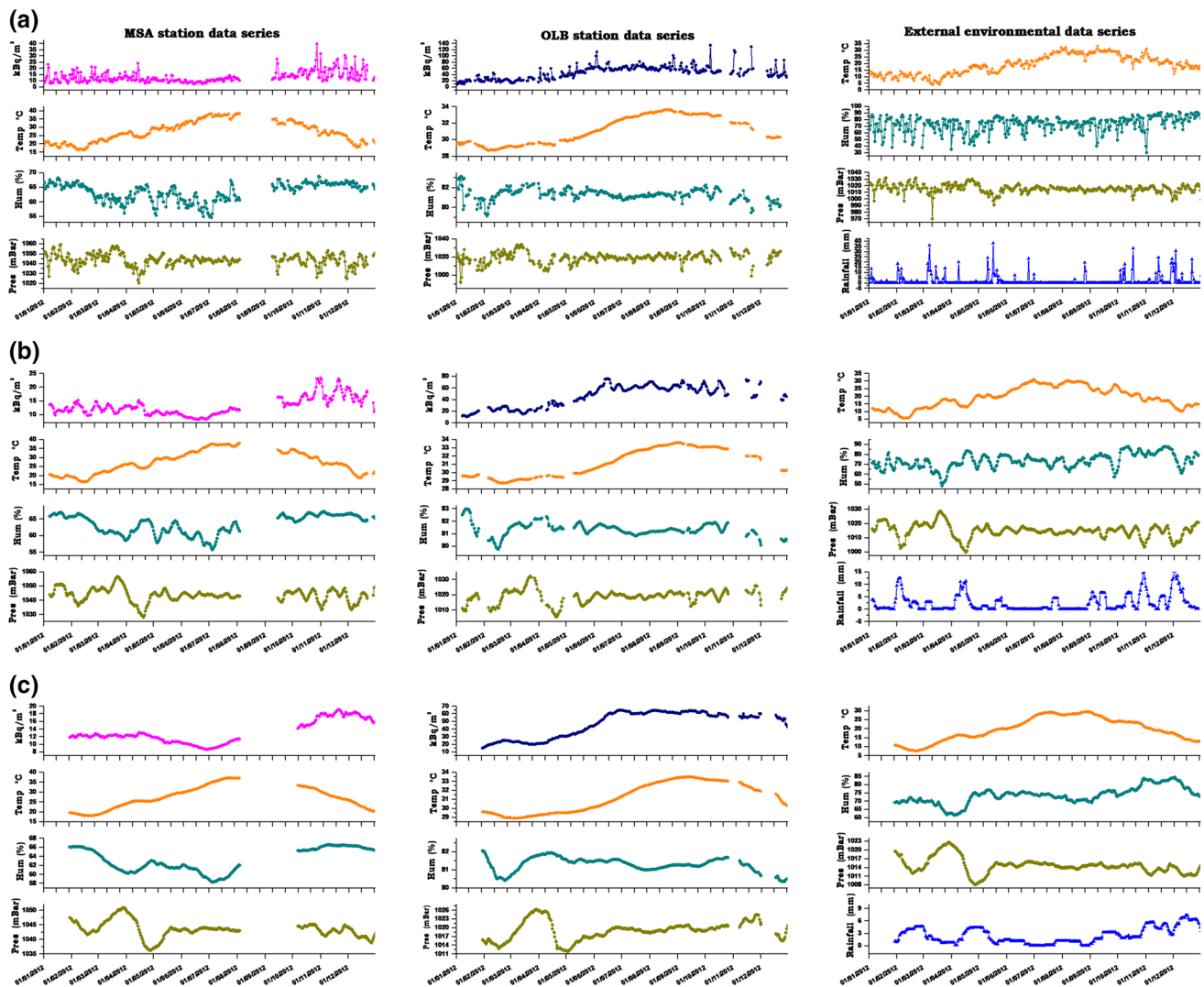
The MSA site is inside the area of the University of Naples “Federico II” in the neighborhoods of the Radioactivity Laboratory; it lies outside the area of interest and can be considered a benchmark against which observations are carried out.

The OLB code identifies the Mount Olibano site whose ground consists mainly of “pozzolana”; it is located under the Mount Olibano inside an old dismissed gallery.

The study consists of the continuous measurement, using RaMonA system, of radon specific activity in soil gas collecting 3-h alpha particle spectra, together with the corresponding mean values of the climatic records.

Since the weather conditions affect radon exhalation from soil superficial layers (Gregorič et al. 2011), the continuous monitoring is performed by pumping the soil gas to a depth of 0.80 m at 1 Lmin<sup>-1</sup> and conveying it to the inlet of the radon monitor.

For radon continuous monitoring, a non-commercial device named RaMonA was employed; it is a device designed and assembled at the Radioactivity Laboratory of the University of Naples “Federico II” (Venoso et al. 2009).



**Fig. 2** MSA, OLB and external data series arranged over daily (a), weekly (b) and monthly (c) timescale

The detector through  $\alpha$  spectrometry of radon and thoron daughters allows the continuous monitoring of the two gases and of temperature, relative humidity and pressure. A typical spectrum from Ramona is shown in Fig. 1.

### Data analysis

The first step in data analysis is the conversion of the counting rate corresponding to radon's descendants ( $^{218}\text{Po}^+ / ^{214}\text{Po}^+$ ) in radon activity concentration; in particular,  $^{218}\text{Po}$  is used to determine radon specific activity. The calibration factor was obtained by means of the Radon Chamber of the Radioactivity Laboratory and resulted to be  $(4.84 \times 10^{-2} \pm 9.32 \times 10^{-4})$  cps/kBqm $^{-3}$  (De Cicco et al. 2010).

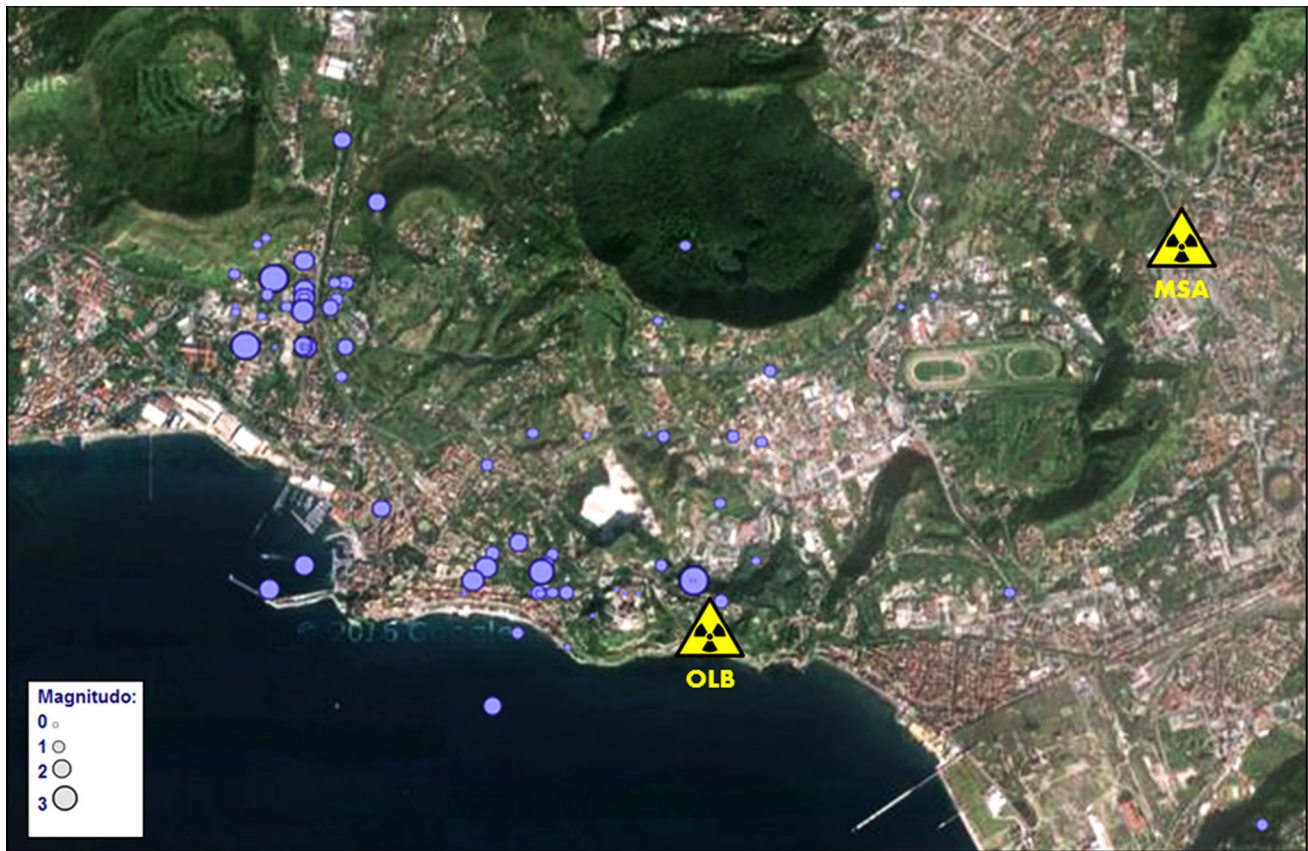
The second step in data analysis consists in investigating the possible dependences of the radon present in soil gas on the environmental data. This is made possible thanks to

RaMonA specifics, which also allow the measurement of climatic values outside the detection chamber. In Table 1, the main statistical parameters resulting from analysis carried out on data provided from stations located in the two sites under study are reported.

Other kind of meteorological data are taken from the historical archive of the Epon center (<http://www.ilmeteo.it/portale/archivio-meteo>), except for rainfall data that are provided from the site of Campania Region Civil Protection where historical rainfall data are stored (<http://centrofunzionale.regione.campania.it/index.php/dati-storici/valori-pluviometrici>).

To best highlight the possible links between environmental data series and radon signal, analysis was performed on three different timescales (Fig. 2):

- Daily values;
- Seven-day moving averages of daily data;



**Fig. 3** Localization of seismic events (*circles*) at Phlegrean Fields from January to December 2012

**Table 2** Seismic events selected according to Dobrovolsky’s empirical formula

DATE	Latitude (DD)	Longitude (DD)	Deepness (km)	Magnitude	OLB distance (km)	MSA distance (km)
26/01/2012	40.889	14.548	5.0	1.9– $M_L$	35	31
14/03/2012	41.041	14.380	5.0	1.8– $M_L$	32	28
20/03/2012	41.033	14.544	5.2	1.3– $M_L$	41	37
24/03/2012	41.106	14.125	11.1	1.2– $M_L$	32	30
27/09/2012	41.176	14.921	10.3	4.2– $M_W$	75	71
27/09/2012	41.161	14.923	10.0	3.5– $M_L$	75	71
27/09/2012	41.160	14.933	11.3	3.8– $M_L$	74	70
16/10/2012	39.670	15.742	275.5	4.6– $M_L$	190	190
25/10/2012	39.875	16.016	9.7	5.2– $M_W$	192	192
05/12/2012	41.161	14.886	21.8	3.2– $M_L$	72	68

c. Twenty-nine-day moving averages of daily data.

Due to the failure of the data network, in some periods the data by Ramona are missing.

Statistical analysis was implemented using multiple linear regression (MLR) method and applied to data relative to both sites considering soil radon specific activity [C(Rn)] as dependent variable and temperature (T), relative humidity (RH), pressure (P) and rainfall (RF) as independent or predictor variables.

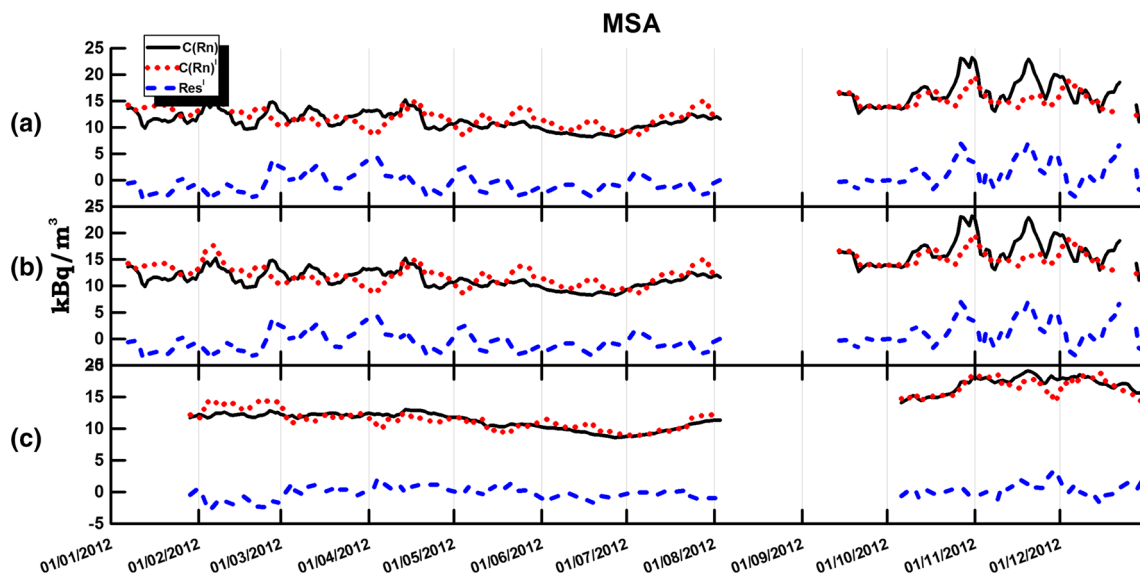
Having once determined the regression equation, it is possible to calculate the predicted values [C(Rn)<sup>I</sup>] of radon concentration, the residuals (Res<sup>I</sup>) and the standardized residuals (ZRes<sup>I</sup>); the last are used to identify “possible” radon anomalies.

To find correlations between earthquakes and radon anomalies, some models have to be taken into account in order to select the events to correlate with radon anomalies.

As previously argued, the Phlegrean Fields are often characterized by the occurrence of earthquake swarms of

**Table 3** Correlation parameters of the MSA data

MSA	Day				Week				Month			
ADJ $R^2$	0.32				0.52				0.88			
	$B$	SE	$\beta$	$p$ value	$B$	SE	$\beta$	$p$ value	$B$	SE	$\beta$	$p$ value
Cost.	177.07	42.96		$4.81 \times 10^{-05}$	-213.07	42.02		$6.85 \times 10^{-07}$	-598.98	33.81		$3.95 \times 10^{-47}$
RF (mm)	0.19	0.04	0.27	$1.30 \times 10^{-06}$	0.51	0.06	0.55	$2.08 \times 10^{-15}$	1.51	0.07	1.01	$1.50 \times 10^{-56}$
T (°C)	0.04	0.04	0.06	$2.60 \times 10^{-01}$	0.12	0.02	0.22	$1.76 \times 10^{-06}$	0.24	0.01	0.45	$1.39 \times 10^{-43}$
RH (%)	0.45	0.08	0.30	$2.37 \times 10^{-08}$	0.58	0.05	0.52	$5.92 \times 10^{-23}$	0.42	0.04	0.37	$2.74 \times 10^{-20}$
P (mb)	-0.19	0.04	-0.25	$5.11 \times 10^{-06}$	0.18	0.04	0.26	$1.50 \times 10^{-05}$	0.55	0.03	0.53	$3.21 \times 10^{-43}$



**Fig. 4** MSA measured (solid line), expected (dot line) values of radon specific activity and residuals (dash line), over daily (a), weekly (b) and monthly (c) timescale

**Table 4** Correlation parameters of the OLB data

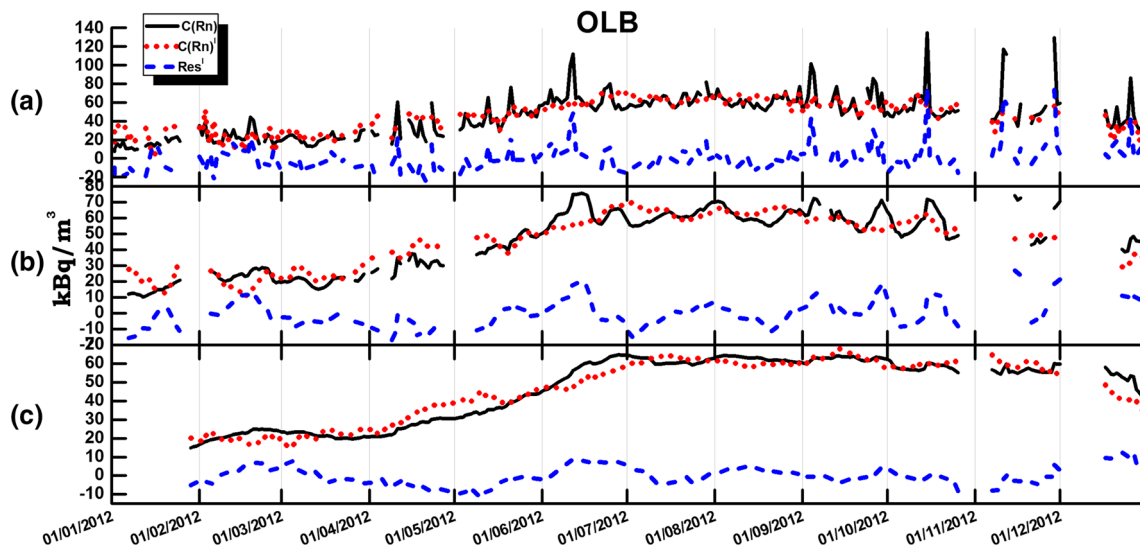
OLB	Day				Week				Month			
ADJ $R^2$	0.61				0.80				0.92			
	$B$	SE	$\beta$	$p$ value	$B$	SE	$\beta$	$p$ value	$B$	SE	$\beta$	$p$ value
Cost.	606.27	142.87		$2.96 \times 10^{-05}$	278.76	163.83		$9.00 \times 10^{-02}$	-1036.20	165.27		$1.23 \times 10^{-09}$
RF (mm)	0.48	0.17	0.11	$5.20 \times 10^{-03}$	0.60	0.31	0.08	$5.20 \times 10^{-02}$	3.28	0.32	0.33	$3.47 \times 10^{-21}$
T (°C)	2.18	0.11	0.72	$5.01 \times 10^{-53}$	2.25	0.08	0.85	$1.44 \times 10^{-78}$	2.52	0.07	0.98	$6.60 \times 10^{-113}$
RH (%)	0.42	0.07	0.23	$5.47 \times 10^{-09}$	0.58	0.08	0.22	$4.74 \times 10^{-13}$	1.14	0.08	0.31	$8.27 \times 10^{-35}$
P (mb)	-0.63	0.14	-0.17	$1.11 \times 10^{-05}$	-0.32	0.16	-0.08	$4.85 \times 10^{-02}$	0.93	0.16	0.15	$1.79 \times 10^{-08}$

extremely low energy and with a maximum depth of about 3–4 km. It was chosen to consider all the earthquakes which occurred in that area throughout 2012.

The geographical localization of seismic events occurred at the Phlegrean Fields from January 2012 to December 2012 was provided by OV-INGV (Fig. 3). There were 101

events:  $M_{Lmax} = 1.8$  and with a depth ranging from 0.49 to 4.65 km; to highlight two swarms: 19–20 April 2012 (14 earthquakes.  $M_{Lmax} = 1.4$ ) and 07 September 2012 (46 earthquakes.  $M_{Lmax} = 1.7$ ).

Since a large event at greater distance might have the same strain impact as a small-magnitude event very near to



**Fig. 5** OLB measured (*solid line*), expected (*dot line*) values of radon specific activity and residuals (*dash line*), over daily (a), weekly (b) and monthly (c) timescale

the stations, a further step was to consider also the earthquakes far from the measuring points whose stress–strain impact might affect the monitoring sites.

A good estimator to select the “region of interest” starting from magnitude is provided by Dobrovolsky’s empirical formula (Dobrovolsky et al. 1979):

$$D = 10^{0.43M} \tag{1}$$

where  $M$  is the magnitude of the earthquake and  $D$  is the effective/strain radius (in km).

This formula was used to calculate earthquakes preparation zone; the selection was carried out from ISIDE (Italian Seismological Instrumental and Parametric DataBase; <http://iside.rm.ingv.it>). The earthquakes that occurred in 2012 fulfilling Dobrovolsky’s empirical formula are summarized in Table 2 where in details are reported:

- Date;
- Latitude and longitude in decimal degrees;
- Event deepness in kilometers;
- Event magnitude in local magnitude ( $M_L$ ) or moment magnitude  $M_W$ ;
- Epicenter distances in kilometers from the sites of MSA and OLB.

## Results

### Multi-parametric analysis

A preliminary study was performed using environmental data both by RaMonA and by Epsom archive;  $ADJ-R^2$  values obtained suggested to use for MSA RaMonA’s T, P

and RH values, while for OLB Epsom’s T, P and RH values. An explanation for this difference can be due to the different setup of the two sites:

- MSA station is located outdoors and therefore directly exposed to atmosphere;
- OLB station is placed under a closed tuff gallery. This material has insulating properties and thermal lag (see Table 1) that can affect radon emanation mechanisms.

In both sites, a numerical analysis using MLR was carried out for data arranged according to daily, weekly and monthly timescales.

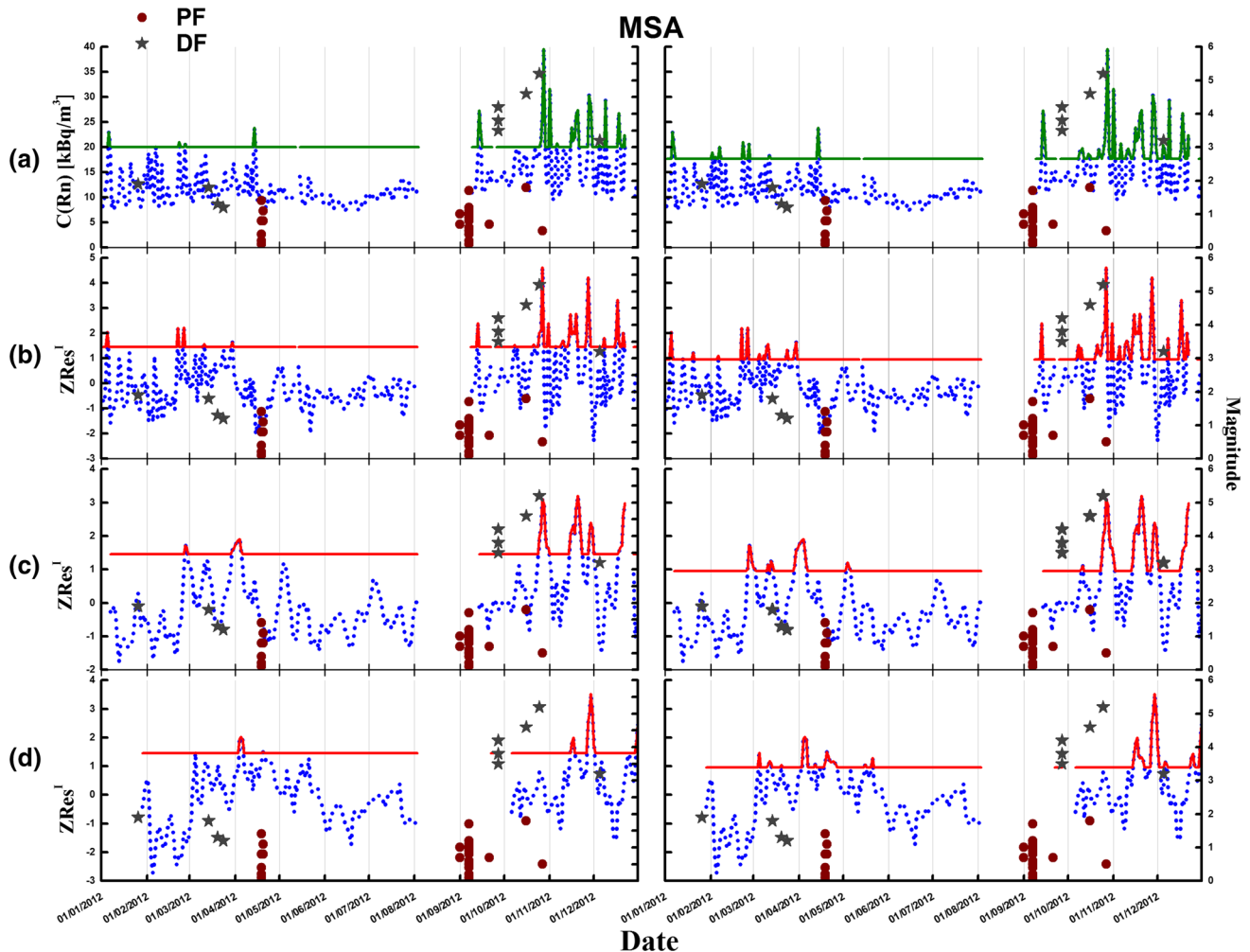
The MLR performed at MSA starting from RaMonA’s values over the three different timescales gave the results shown in Table 3 in terms of:

- Adjusted  $R$ -square ( $ADJ-R^2$ ).
- Non-standardized coefficient ( $B$ );
- Standard error of non-standardized coefficient (SE);
- Standardized coefficient ( $\beta$ );
- $p$  value.

In Fig. 4, measured  $C(Rn)$  and expected  $C(Rn)^I$  temporal trends of radon specific activity are plotted together with the residuals  $Res^I$  resulting from the three different timescale analyses during the observation period (01/01/2012–31/12/2012) at MSA site.

As in case of MSA, the MLR was performed at OLB site with the difference that  $C(Rn)$  was regressed with values T, P and RH taken from Epsom archive; the results are displayed in Table 4.

In Fig. 5,  $C(Rn)$ ,  $C(Rn)^I$  and  $Res^I$  relative to OLB site are plotted over the three different timescales.



**Fig. 6** MSA “possible” anomalies resulting from raw (a), daily (b), weekly (c) and monthly (d) data analysis

### Anomaly analysis

MLR gave information about the dependence of measured radon specific activity on climatic conditions in both sites providing a useful tool in order to better understand the site-specific behavior of radon emanation.

The supplementary purpose of this analysis is to optimize a reliable methodology to validate the use of radon as “geoindicator” after having removed the environmental influence.

To better study radon “possible anomalies” caused mainly by seismo-volcanic activity, the equations of the standardized residuals  $ZRes^1$  were used: Starting from standardized residuals plots, it was proceeded to select the values exceeding a fixed threshold. In order to attempt an estimation of their possible correlation with seismic events, the selected “radon anomalies” were compared with:

- Seismic events selected according to Dobrovolsky’s formula;

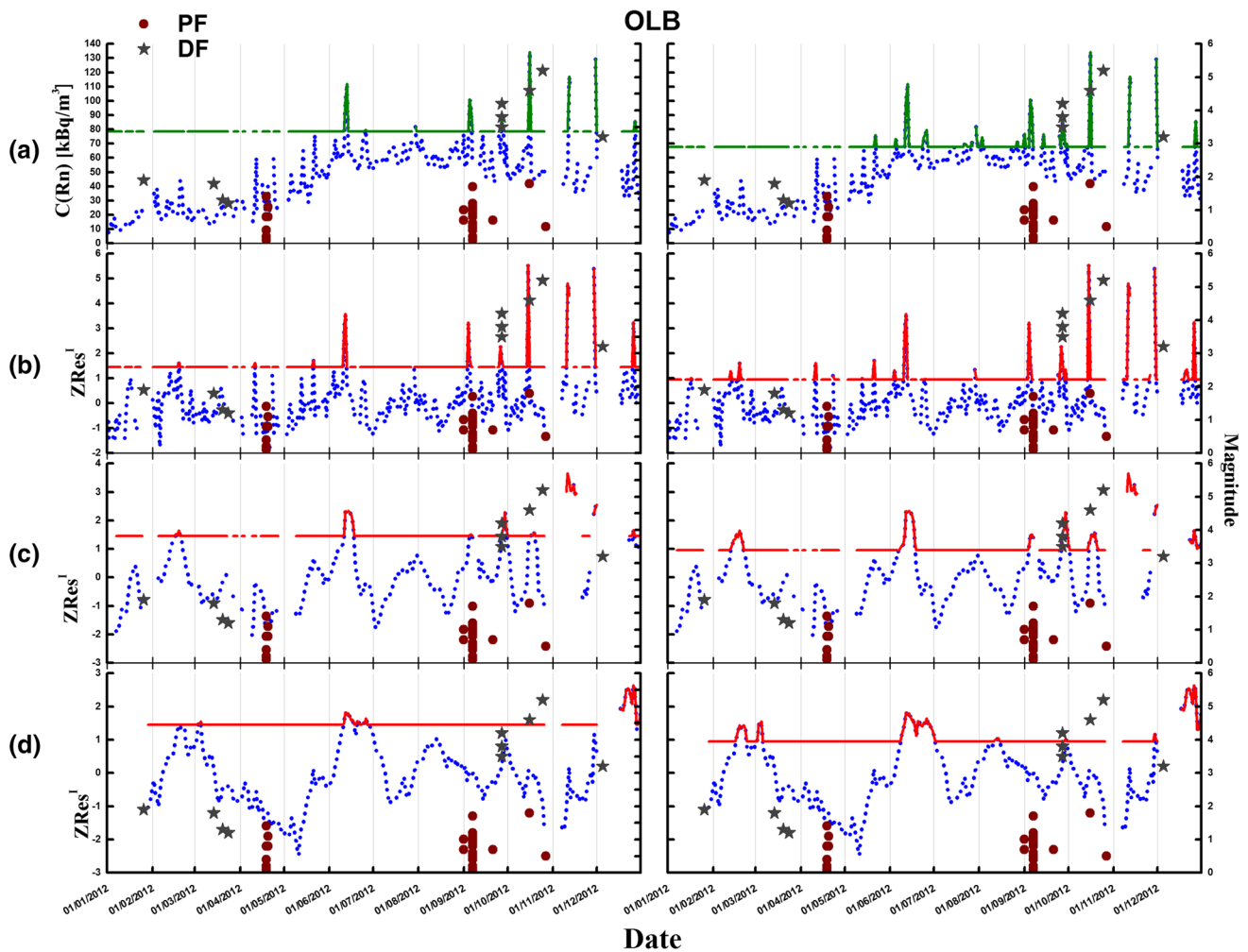
- The two swarms (19–20 April 2012 and 07 September 2012) and all the earthquakes of  $ML > 0.5$  occurred at the Phlegrean Fields.

In order to minimize the number of false anomalies, averages on different timescales were calculated and the analysis was carried out on data selected with different thresholds. The most satisfactory results were obtained in correspondence of threshold values equal to 1.0 and 1.5.

In Fig. 6, the results obtained from analysis carried out on data collected at MSA site are displayed. In detail are plotted:

- Seismic events selected according to Dobrovolsky’s formula (DF—stars);
- Seismic events of small magnitude at Phlegrean Fields (PF—circles);
- Measured values of radon specific activity  $C(Rn)$  (a);
- Standardized residuals  $ZRes$  (b, c and d).





**Fig. 7** OLB “possible” anomalies resulting from raw (1), daily (2), weekly (3) and monthly (4) data analysis

In plot (a), solid line corresponds to the values of measured C(Rn) exceeding  $1.5\sigma$  (left side) and  $1\sigma$  (right side) of the mean value of radon specific activity.

In plots (b), (c) and (d), the solid lines account for the values of  $ZRes^{-1} > 1.5$  (left side) and  $ZRes^{-1} > 1$  (right side).

The results of the analysis carried out on data collected at OLB site are displayed in Fig. 7. The criteria adopted for plots are the same used for MSA.

**Discussion and conclusions**

Results obtained show that surely the method employed is useful to study correlations among radon specific activity and environmental features in order to highlight site-specific behavior.

From the analysis based on daily averages, it is possible to observe that the influence of the climatic conditions on radon specific activity variation in MSA is about the half of

OLB ( $ADJ-R^2$ , respectively, 0.32 and 0.61). This difference is surely due to the lack of significant dependence from T in MSA ( $\beta = 0.06$  and  $p$  value = 0.26). It might be explained by the different setup of RaMonA in the two sites: In MSA, the pipe for sampling soil air is placed outdoors, while in OLB, the whole system is located under a gallery built in the tuff. According to Barbosa et al. (2010), Garavaglia et al. (2000) and Perrier et al. (2004) in an underground tunnel, the thermal gradient between the inside of the gallery and the outside air leads to an accurate correlation between external temperature and soil radon amount.

Apart from this evidence, the other findings seem to be in agreement with most of the results published in the literature (Iskandar et al. 2004; Planinić et al. 2001; Walia et al. 2005); in fact a positive dependence from T, RH and RF and a negative one from P were found for both sites.

Also in weekly and monthly analyses, the two monitoring points show different responses: In MSA, it can be noticed that the dependence on the temperature is retrieved

leading to a progressive increase of  $ADJ-R^2$  and a positive correlation with pressure was recorded as also in OLB monthly analysis.

In summary, in MSA site radon emanation is influenced mainly by rainfall (RF) ( $\beta$  ranging from 0.27 to 1.01) and relative humidity (RH) ( $\beta$  ranging from 0.30 to 0.51), while in OLB site, C(Rn) is ruled firstly by temperature (T) ( $\beta$  ranging from 0.72 to 0.98) and secondly by relative humidity (RH) ( $\beta$  ranging from 0.23 to 0.31).

Likewise, this statistical tool allowing to partially remove the meteorological dependence of radon emanation is useful to better differentiate those radon anomalies caused by seismic events and not by meteorological conditions.

In this context, analysis carried out starting from the 7 and 29 moving averages of daily data is useful; in fact, these smoothed values allow to reduce the presence of high-frequency spikes in radon anomalies.

In particular as can be appreciated in Figs. 6 and 7, there can be some correlations with earthquakes of  $ML > 2$  and swarms for both sites in the results coming from “models” c and d; notice the evident “false” anomaly in July in OLB site.

Better results could be obtained by improving analytical methodologies and also increasing physical and geophysical variables to be investigated.

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