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^{222}Rn concentration in the atmosphere in Milan and in the plain around

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Summary. — Radon measurements at ground level are a useful tool for the study on atmospheric stability or mixing conditions. Radon concentration in the atmosphere was measured in Milan over a continuous four-year period from 1996 to 1999. In the town area pollutants emitted during late evening and night hours by motor traffic as well as by industrial plants accumulate at low height concurring with radon accumulation in the Nocturnal Stable Layer. In years 1998-1999 radon measurements were taken in Landriano, an area located 30 km south of the town. In Lombard plain there are numerous huge plants, thermoelectric power stations burning by methane and carbon, oil refineries; moreover other plants are being carried out. With the aim to control the contribute of these plants to air pollution, it is important to know atmospheric stability and turbulence conditions. Landriano is only few kilometres from the thermoelectric power station in Tavazzano. In this paper the results of measuring campaigns as well as the comparison of radon concentrations between the two sites surveyed are reported. The general criteria of the measurements, the interpretative models of radon concentration in the atmosphere are discussed.

PACS 92.60.Ek – Convection, turbulence, and diffusion.
PACS 23.60.+e – Alpha decay.
PACS 89.60.Ec – Environmental safety.

1. – Introduction

The study of ^{222}Rn concentration in the atmosphere gives information on the state of turbulence and the stability of the lower atmosphere as well as highlights the transport and origin of air masses.

A natural radioactive tracer, ^{222}Rn, is emitted from the ground at an almost constant rate. Two characteristics aid the study of ^{222}Rn: its half-life (3.82 days); and the noble-

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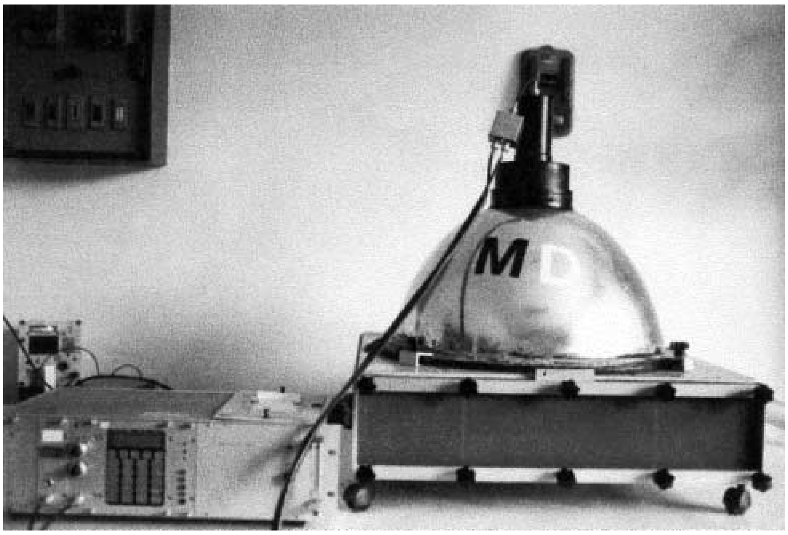

Fig. 1. – Radon measurement detector.

gas nature so that radon does not interact with the chemical and photochemical processes present in the atmosphere [1-3].

Beginning from year 1996 to 2000 the Physics Institute has been carried out numerous outdoor radon measurements campaigns in the town of Milan and in a few sites of the Lombard plain.

Measurements in Milan were taken in the garden of the Physics Institute located in the east side of the town.

In years 1998-1999 measurements in Landriano, an agricultural area located 30 km south of the city, were performed.

Radon stations operated in open spaces away from trees; the air intake was at a height of 3 m from the ground.

The present paper is concerned with a view of the results as well as a comparison of radon concentrations between the two sites surveyed.

The general criteria of the measurements, the interpretative models of radon concentration in the atmosphere are discussed.

2. – Materials and methods

2·1. *Instruments*. – The measurements of ^{222}Rn in the atmosphere were performed by means of high volume scintillation chambers (fig. 1).

A lucite disk with a diameter of 400 mm is covered with a thin activated zinc sulphide layer, capable of scintillation when hit by alpha-particles. The disk is set on a surface aluminium specular paraboloid, the focus of which houses a photomultiplier.

The airflow is set at 5 l/min.

A fibre-glass filter to capture aerosols and with them the decay products of ^{222}Rn and ^{220}Rn present in the atmosphere was placed on the air inlet. The connection between the air intake and the scintillation chamber was a tube of an appropriate diameter and length so that the decay of the ^{220}Rn ($t_{1/2} = 56$ s) takes place before detection. Thus the counts obtained using the scintillation chamber refer only to ^{222}Rn and the products

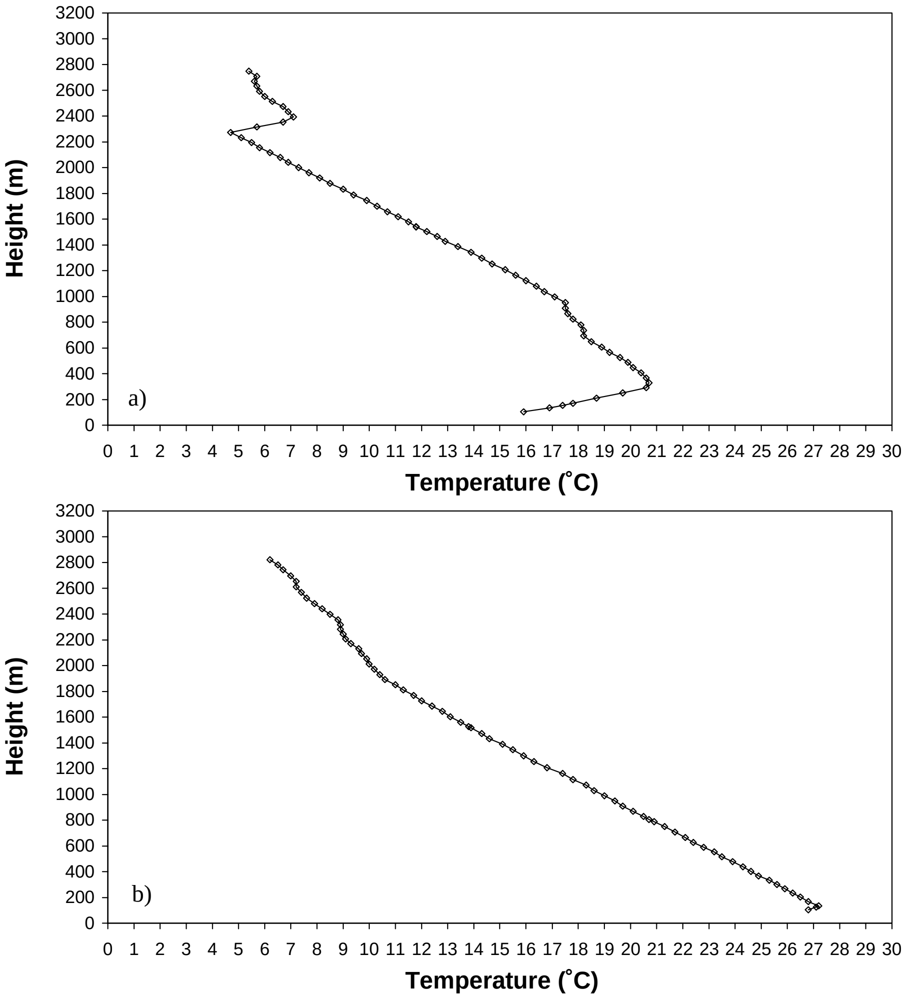

Fig. 2. – Temperature profile at Linate Airport, 11 May 1998: a) 5 a.m. b) 5 p.m.

of its decay, which form in the chamber itself.

The signal given by the photomultiplier is sent via preamplifier and amplifier to the analysing and recording system. The calibration of the counter is carried out using air containing a known concentration of radon; the background count is made with radon-free air. Values of 20–30 counts per hour per Bq/m^3 of ^{222}Rn present in the atmosphere and background values of 150–250 counts per hour, respectively, were obtained for the two instruments.

The apparatus runs continuously: the data of the measured concentrations refer to the beginning of the hour.

2˙2. *Models of analysis*

2˙2.1. Inversion of temperature with base at ground level. In the Milan area and in general in the plain, after days of intense solar radiation, in clear windless nights, an inversion of temperature occurs with its base at ground reaching heights of a few hundred metres. In the Milan area the inversion can be observed directly by means of the thermal soundings that are carried out daily at the nearby Milan Linate Airport, particularly from night and early morning soundings (fig. 2a).

On these nights the Planetary Boundary Layer (PBL) is divided into separated layers, the Nocturnal Stable Layer (NSL) below the inversion of temperature and above this the Residual Layer, in which the remixing remains unaltered up to the PBL [4].

The most interesting aspect of the study of the conditions of radon is the fact that on these nights, radon emitted from soil accumulates in the NSL reaching high concentrations, up to 15–50 Bq/m^3.

The nocturnal accumulation of radon does not occur in conjunction with rain or strong winds.

In the next paragraph drawings of nocturnal accumulation of radon are given.

The nocturnal accumulation of radon begins in the evening and extends through the night reaching maximum values at around 6 a.m. Radon concentration then decreases until it reaches its minimum value in the afternoon.

The concentration of radon indicates the formation of NSL and highlights its dissolution in the early hours of the morning.

The box model originally suggested by the Tolosa group [1] describes the accumulation of radon within the NSL.

This allows for an estimation of the equivalent remixing height h_{e} on the basis of the following hypotheses:

– the flow of radon from the ground is constant over a vast area and over time;

– radon is distributed uniformly within a layer of height h_{e};

– advection is not evident; radon mixing is only vertical.

The formula that describes the accumulation of radon from time t_{i} to time t_{f} is the following:

$$h_{\mathrm{e}}C_{\mathrm{f}} = \frac{\phi\left(1 - e^{-\lambda(t_{\mathrm{f}}-t_{\mathrm{i}})}\right)}{\lambda} + h_{\mathrm{e}}C_{\mathrm{i}}e^{-\lambda(t_{\mathrm{f}}-t_{\mathrm{i}})}\,, \tag{1}$$

where

C_{f} is the concentration of radon at time t_{f} (Bq/m^3);

C_{i} is the concentration of radon at time t_{i} (Bq/m^3);

ϕ is the flow of radon from the ground and one assumes indicatively $\phi = 72$ Bq/m^{2}h; λ is a constant of radioactive decay.

The term $\frac{\phi(1-e^{-\lambda(t_{\mathrm{f}}-t_{\mathrm{i}})})}{\lambda}$ represents the quantity of radon accumulated over time $(t_{\mathrm{f}} - t_{\mathrm{i}})$, keeping account of radioactive decay.

The hour-by-hour application of the formula presents some difficulties and above all a statistical uncertainty of the data and possible sharp variations of the concentration.

If the hourly radon concentration from minimum to maximum raises in linear way, with a constant derivative, it is possible to obtain an average value of h_{e} with the following formula:

$$h_{\mathrm{e}}C_{\max} = \frac{\phi\left(1 - e^{-\lambda(t_{\mathrm{f}}-t_0)}\right)}{\lambda} + h_{\mathrm{e}}C_0e^{-\lambda(t_{\mathrm{f}}-t_0)} \tag{2}$$

where

C_0 is the radon concentration at the initial time t_0;

$C_{\max}$ is the maximum concentration at the time t_f.

Whenever there is evidence for abrupt variations of radon accumulation during the night formulae (1) and (2) cannot be applied and a more detailed analysis is needed. For example this is the case of advection due to breezes.

2˙2.2. General atmospheric mixing and radon dissolving in height. At sunrise, remixing of atmosphere occurs at higher altitudes up to PBL.

Thermal soundings carried in Linate point out the different steps of this process; during the spring and summer, at midday atmospheric mixing rises to a height of 700–1000 m, and in the afternoon it reaches 1500–2000 m (fig. 2b).

Carson describes in his model the altitude of the mixed layer [5]: the heat, transmitted from the sun to the ground, is gradually transferred to the atmosphere. This model, hourly applied to thermal soundings, allows estimation of the raising of mixing level. During atmospheric mixing, radon mixing is directly observable; decreasing radon concentration starts in early morning; it reaches its minimum value in the afternoon.

Applying box model's formula, only to the early hours of the morning, it is possible to plot h_e as a function of time.

It appears more difficult to follow radon's evolution during both midday's hours and the afternoon, when mixed layer reaches higher altitudes.

However an indication of atmospheric diffusive properties during maximum mixing hours can be deducted by the observation of the minimum values of radon concentration in the afternoon. In many sequences of hourly radon concentrations it can be noted that the minimum value remains constant for a succession of many days.

Monthly average has been made on values of radon concentrations reached at 5-6-7 p.m. with the aim to give information about diffusive properties of the atmosphere during the maximum mixing hours.

3. – Results of measurements

3˙1. *Milan*. – Figure 3 is related to radon concentration in Milan during September months of 1996-1997-1998 and 1999. There is a frequent radon accumulation during the night, which is followed by dispersion during morning and afternoon's hours. Maximum values, reached during early morning hours are on the order of 15–25 Bq/m^3, whereas minimum values during afternoon's hours are about 4–6 Bq/m^3.

As discussed in the previous section, radon accumulation corresponds to a ground-based temperature inversion and thus to the formation of a NSL in which radon emitted from soil does accumulate.

Figures 4a, 5 represent patterns of radon concentration in winter months of December 1997 and January 1998. It can be seen how through the whole day radon concentrations are averagely high with peaks of nocturnal accumulation up to 40 Bq/m^3. Generally high values of daytime radon concentrations indicate the presence of a low-height mixing layer.

Note the long period of stability in the time from 1 to 19 January.

In addition it can be observed that nocturnal accumulation of radon does not take place in the presence of a strong wind. Figure 4 shows along with radon concentration the plot of wind velocity.

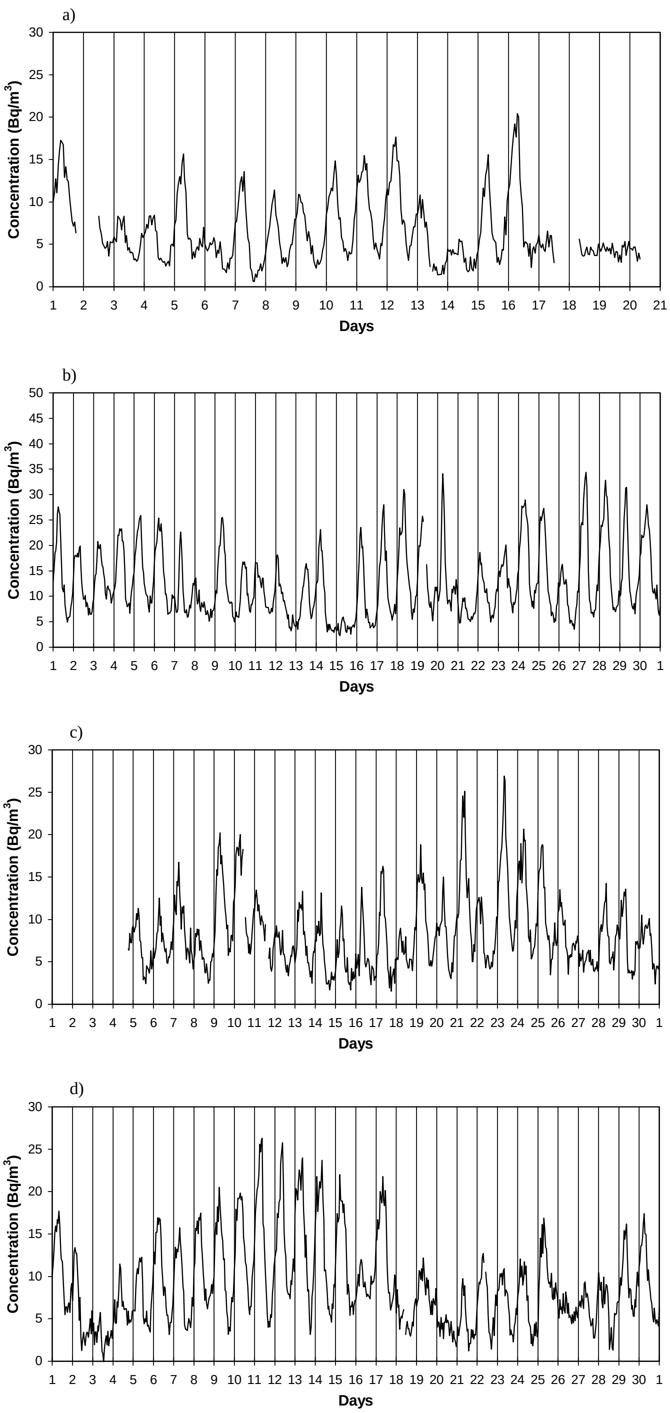

Fig. 3. – Trend of radon concentrations: Milan, September a) 1996, b) 1997, c) 1998, d) 1999.

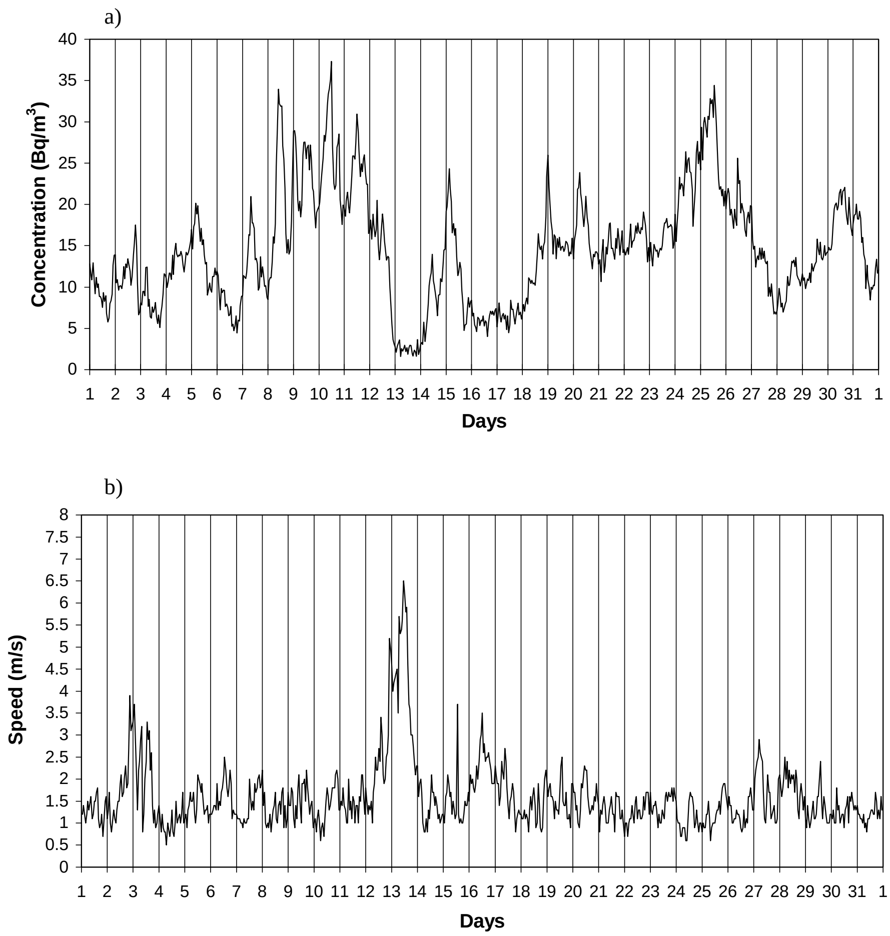

Fig. 4. – Trend of radon concentrations a) and wind speed b): Milan, December 1997.

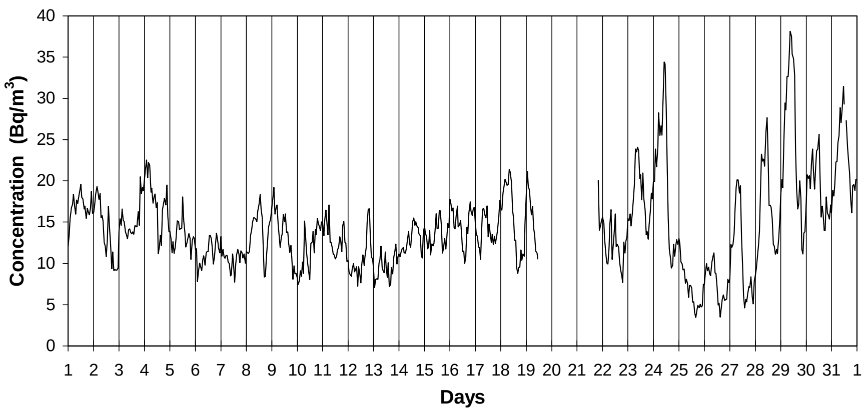

Fig. 5. – Trend of radon concentrations: Milan, January 1998.

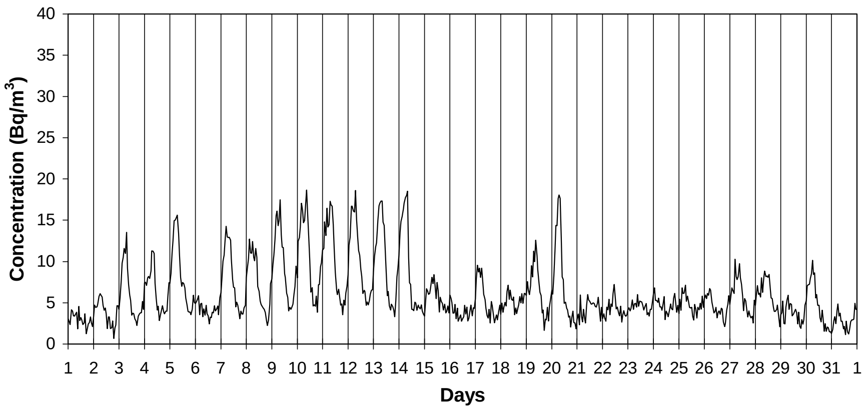

Fig. 6. – Trend of radon concentrations: Milan, May 1998.

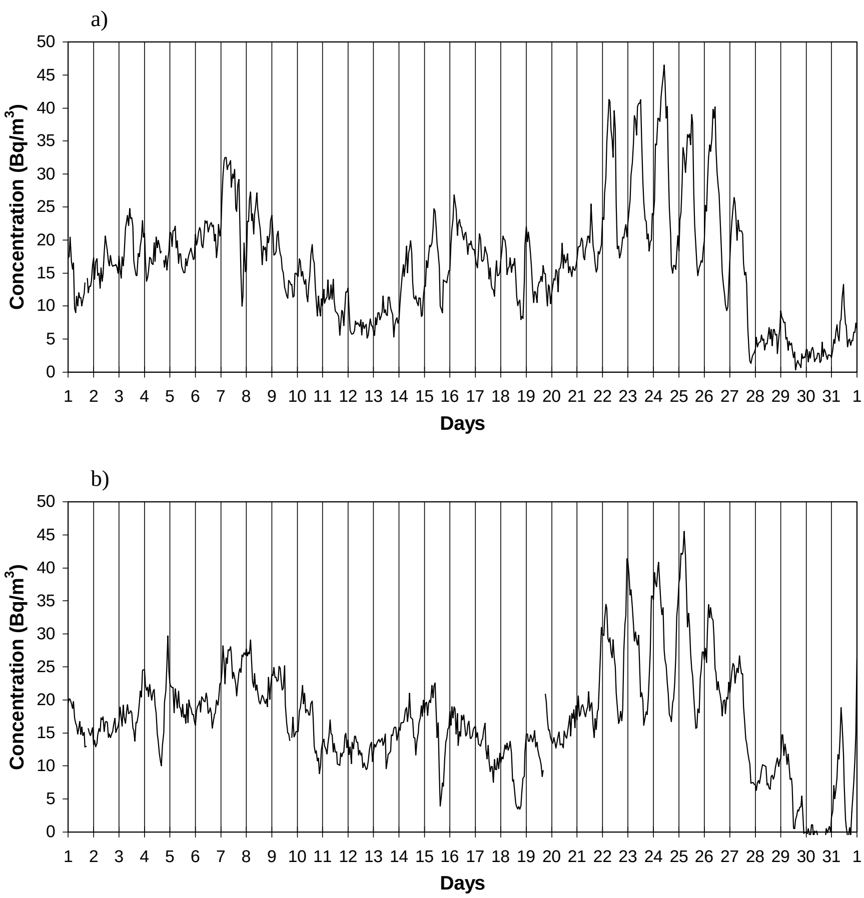

Fig. 7. – Trend of radon concentrations for January 1999: a) Milan, b) Landriano.

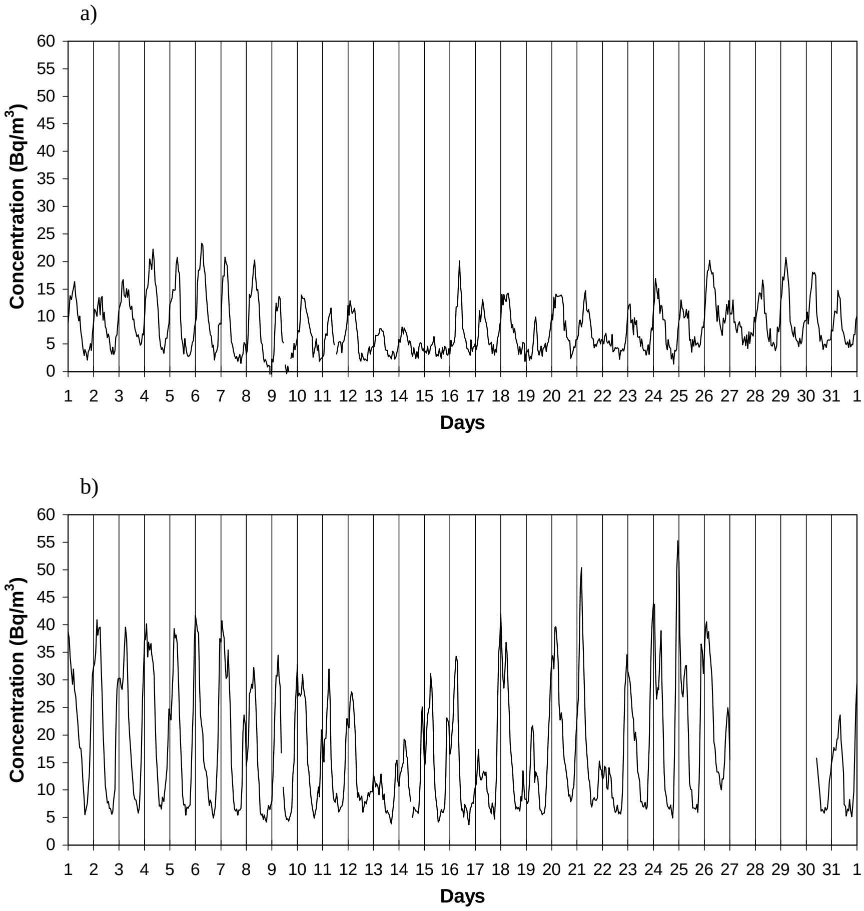

Fig. 8. – Trend of radon concentrations for August 1999: a) Milan, b) Landriano.

In spring and summer, periods of nocturnal accumulation alternate with remixing ones, in which series of days characterized by low radon concentrations are clearly visible. Such sequences corresponds to a strong remixing in height up to PBL.

In May 1998 (fig. 6) days 7 up to 14 show a regular nocturnal accumulation and minimum afternoon values around 4–5 Bq/m^3, whereas in following days radon remixing at high altitude during night and day can be seen generally; in these days wind velocity is elevated.

Figures 7a, 8a are related to January and August 1999, respectively; fig. 9a is concerned to October 1999.

In January peaks values of 50 Bq/m^3 are present. Elevated minimum values and complex pattern are also present.

In spring and summer (see August), nocturnal accumulation is more frequent and there are peak values reaching 20 Bq/m^3.

Note that October is characterized by elevated minimum values of concentration, which remain constant for many days.

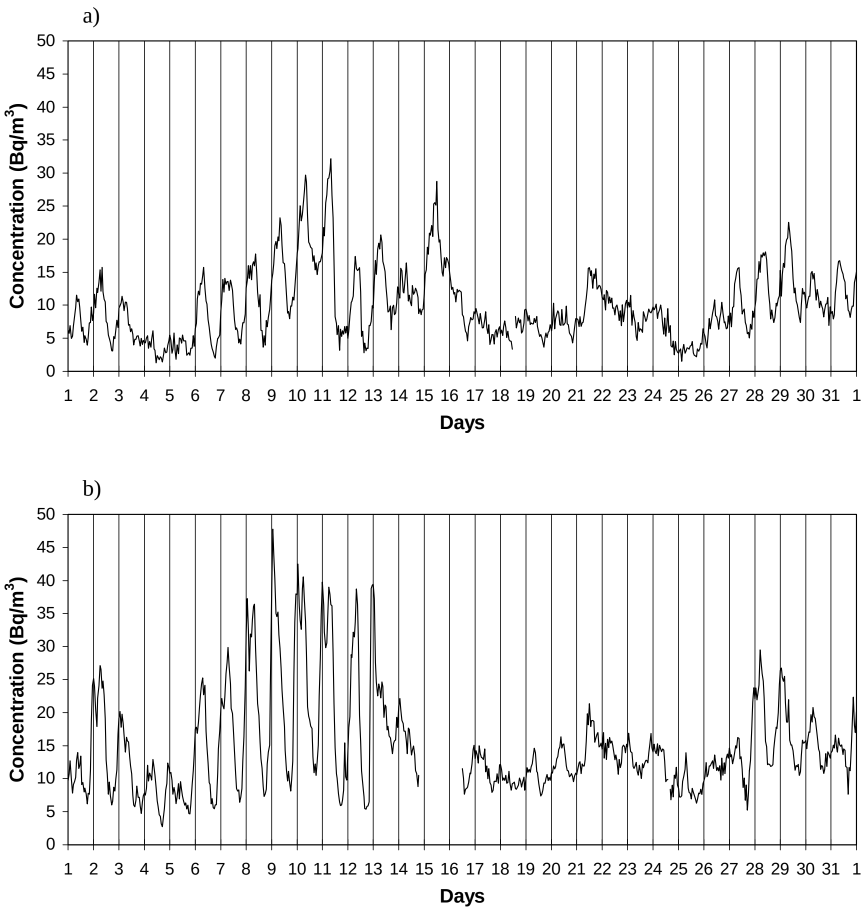

Fig. 9. – Trend of radon concentrations for October 1999: a) Milan, b) Landriano.

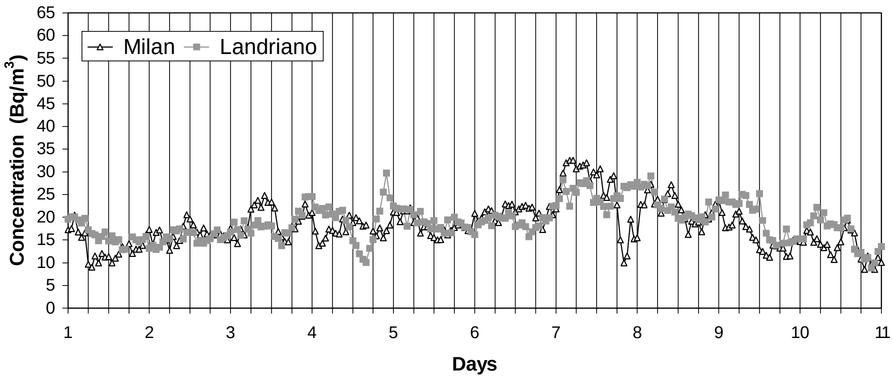

Fig. 10. – Comparison of radon concentrations between Milan and Landriano in January 1999.

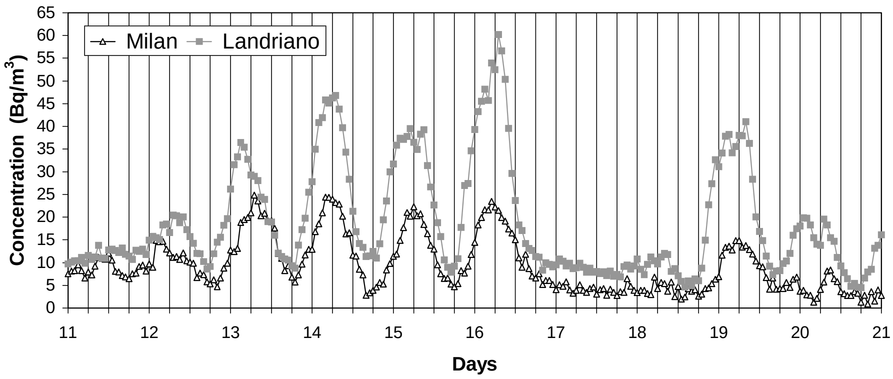

Fig. 11. – Comparison of radon concentrations between Milan and Landriano in March 1999.

3·2. *Landriano*. – In figs. 7-9 concentration values measured in Landriano are directly compared with the corresponding values measured in Milan.

Radon concentration of January 1999 is plotted in figs. 7 and 10.

During winter months radon concentration in agricultural plain is nearly equal to that observed in Milan indicating a high minimum value and nocturnal accumulation up to 50 Bq/m^3 (see 22-26 January).

Figures 8, 9 show the trend of radon concentrations observed in August and October 1999 for Milan and Landriano. The nocturnal accumulation at Landriano can be observed (fig. 8b). Peak values of accumulation result higher by about a factor of 2 in comparison with Milan.

Such comparison is also shown in fig. 11 for March 1999.

4. – Averaged values

In table I the number of days in which nocturnal accumulation in Milan takes place over a month is reported for the four years studied. The average length of the period of accumulation $\overline{\Delta t}$ is also indicated. Nocturnal equivalent remixing height has been calculated for these days: the $\overline{h_e}$ values averaged all over the month are reported.

The $\overline{h_e}$ values referred to Landriano station are also presented.

Nocturnal accumulation occurs with a frequency that is almost the same for the two stations. Taking a look at $\overline{h_e}$ values it can be seen that the lower ones are generally observed in Landriano as corresponding to the higher nocturnal radon concentrations.

The radon concentrations averaged over both measured values at 5-6-7 p.m. and all over the month are listed month by month in table II.

It is interesting to note how in year 1999 average minimum values for the two stations do not differ, so it can be considered that a similar remixing occurs over the whole area.

TABLE I. – *Average monthly values of h_e referred to days of nocturnal accumulation for Milan and Landriano.*

Milan 1996			
Month	Days	$\overline{\Delta t}$ (h)	$\overline{h_e}$ (m)
July	7	10	60
August	8	8	51
September	8	11	62
October	12	12	65
November	5	12	66
December	5	12	56

Milan 1997			
Month	Days	$\overline{\Delta t}$ (h)	$\overline{h_e}$ (m)
January	10	13	54
February	9	12	60
March	9	12	48
April	7	11	52
May	5	12	59
June	5	9	47
July	7	11	52
August	11	12	45
September	17	12	41
October	5	15	38
November	6	13	47
December	3	14	44

Milan 1998			
Month	Days	$\overline{\Delta t}$ (h)	$\overline{h_e}$ (m)
January	4	14	44
February	16	13	45
March	12	11	54
April	3	12	60
May	12	11	63
June	7	8	49
July	13	10	61
August	10	12	48
September	8	12	54
October	10	12	49
November	12	14	48
December	5	13	39

TABLE I. – *Continued.*

Milan 1999			
Month	Days	$\overline{\Delta t}$ (h)	$\overline{h}_e$ (m)
January	5	14	38
February	6	13	48
March	6	11	53
April	3	12	68
May	2	11	33
June	8	9	33
July	7	11	34
August	12	10	51
September	10	12	58
October	7	13	59
November	6	12	51
December	5	13	46

Landriano			
Month	Days	$\overline{\Delta t}$ (h)	$\overline{h}_e$ (m)
December 1998	6	7	19
January 1999	6	7	23
February	10	7	17
March	9	8	22
May	4	7	18
June	10	6	17
July	10	6	17
August	15	6	15
October	5	10	23

TABLE II. – *Average monthly values of radon concentration referred to 5-6-7 p.m. for Milan and Landriano.*

Milan 1996	
	Average monthly minimum values for 5-6-7 p.m. (Bq/m^3)
July	6.5
August	9.1
September	3.6
October	6.6
November	6.8
December	11.4

Milan 1997	
	Average monthly minimum values for 5-6-7 p.m. (Bq/m^3)
January	12.2
February	7.4
March	4.8
April	3.2
May	4.7
June	5.8
July	3.8
August	5.6
September	7.0
October	7.7
November	11.6
December	13.6

TABLE II. – *Continued.*

Milan 1998	
Average monthly minimum values for 5-6-7 p.m. (Bq/m^3)	
January	12.3
February	8.2
March	3.6
April	4.2
May	3.7
June	3.7
July	3.9
August	4.9
September	4.9
October	5.9
November	11.0
December	16.5

Milan 1999	
Average monthly minimum values for 5-6-7 p.m. (Bq/m^3)	
January	12.9
February	5.1
March	4.7
April	4.0
May	5.8
June	5.9
July	6.5
August	4.1
September	4.9
October	7.0
November	10.9
December	11.9

Landriano	
Average monthly minimum values for 5-6-7 p.m. (Bq/m^3)	
December 1998	22.1
January 1999	13.8
February	9.0
March	8.8
May	5.8
June	6.4
July	6.5
August	7.2
October	9.5

5. – Conclusions

Long-period study of atmospheric radon concentration allows characterization of a site with regard to atmospheric stability and turbulence conditions. In particular, it is possible to highlight the presence of NSL by simple measurements.

In fact nocturnal radon accumulation points out the presence and evolution of NSL; and allows an estimation of the equivalent remixing height h_e.

In particular the study of radon accumulation allows identification of nights in which the condition of "reactive atmosphere" is verified.

In these nights, nitrogen oxides, and volatile organic compounds (VOC) mixtures interact with each other: this condition is similar to a chemical reactor in which reactions are favoured by the great density of chemical and photochemical pollutants.

For example, there is a considerable formation of HONO, nitrous acid, which sensibly contributes to the morning increase in O_3 production [6, 7].

In afternoon hours, when radon concentration presents lower values it can be seen in concomitance with vertical and horizontal remixing a dilution at high altitude and all over a wide area (condition of dispersive atmosphere).

By comparison of measurements a similar behaviour in agricultural plain and in the town area can be observed.

In Lombard plain there are numerous huge plants, thermoelectric power stations burning by methane and carbon, oil refineries; moreover other plants are being carried out.

With the aim to control the contribute of these plants to air pollution, it is important to know atmospheric stability and turbulence conditions.

By measurements in Landriano it is clear that emissions by chimneys of the next thermoelectric power station in Tavazzano take place above the remixing heights of NSL; these emissions do not reach ground level but disperse at height.

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