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ORIGINAL ARTICLE

Observations of surface radon in Central Italy

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Abstract Two years of in situ radon concentration measurements in the atmospheric surface layer have been collected in a central Italy town (L'Aquila), located in the Aterno river valley. These data have been analyzed in order to study the controlling mechanisms of surface radon abundance; observations of coincident meteorological parameters confirmed the role of dynamics on the local removal rate of this tracer. The relatively high negative correlation of hourly data of surface wind speed and radon activity concentration ($R = -0.54$, on annual scale) suggests that dynamical removal of radon is one of the most important controlling processes of the tracer accumulation in the atmospheric surface layer. An attempt is made to quantify the precipitation impact on radon soil fluxes. No anticorrelation of radon and precipitation comes out from the data ($R = -0.15$), as in previous studies. However, since the main physical parameter affecting the ground radon release is expected to be the soil accumulation of water, snow or ice, the emission flux has also been correlated with soil moisture; in this way a much clearer anticorrelation is found ($R = -0.54$).

Keywords Evapotranspiration · Radon · Soil · Precipitation · Radioactivity

Introduction

Radon (^{222}Rn) is a radioactive noble gas emitted mainly by soil. The radioactive decay (mean lifetime of 5.517 days) acts as a net global radon sink. In the atmosphere, radon does not chemically react with other species, it does not attach to aerosols and is not subject to wet and dry deposition (Zahorowski et al. 2004). Besides radioactive decay (which is relevant only on large spatial domains), radon local variations are mainly controlled by soil emissions and transport. The lifetime of radon in the atmospheric surface layer is of the same order of magnitude as for many short-lived atmospheric pollutants (e.g., NO_x, CO, SO_2, aerosols, O_3), making radon a good tracer to study their variability (Gaudry et al. 1990; Ramonet et al. 1996; Vinod Kumar et al. 1999). The advantage of radon is that, being chemically inactive, the balance of its sources and sinks is much simpler than for the above cited pollutants, even though there are still relevant issues of uncertainty (see ahead). The atmospheric radon lifetime is also comparable with convective and synoptic-scale motions, so that radon is a useful tracer for their studies (Jacob and Prather 1990; Allen et al. 1996; Mahowald et al. 1997; Stockwell et al. 1998; Kataoka et al. 1998; Sesana et al. 1998). Calibration and validation of atmospheric transport models is another use of ^{222}Rn observations (Genthon and Armengaud 1995; Li and Chang 1996; Jacob et al. 1997; Stockwell et al. 1998; Dentener et al. 1999). A detailed review of the applications of radon measurements in atmospheric studies is reported by Zahorowski et al. (2004).

Geographic variability of radon emission is not well known due to sparse measurements. The global average emission rate is better understood using Pb-210 deposition measurements. In the absence of knowledge, most investigators conventionally choose a single emission rate to

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apply to all land surfaces that will result in the observed deposition (between 0.1 and 2.5 atoms cm^{-2} s^{-1}, the most used value being 1 atom cm^{-2} s^{-1}) (Zahorowski et al., 2004; Jacob et al. 1997; Lee and Feichter 1996; Considine et al. 2005). Ocean emission of radon is considered between zero and 0.005 atoms cm^{-2} s^{-1}. A reduction of soil radon emission with latitude is applied in global models to take into account the effect of snow cover; Considine et al. (2005) considered a reduction of 40% of the radon emission when air temperature is below 273.15 K, to account for radon flux reduction due to frozen soil.

The reduction of radon emission due to precipitation is something expected and shown in several studies: (Israel and Horbert 1966; Megumi and Mamuro 1973; Ishimori et al. 1998; Galmarini 2006). Nevertheless emission of ^{222}Rn is normally not explicitly correlated with soil moisture in global models, even though the inclusion of a physically-based parameterization could help improve model predictions (Jacob et al. 1997; Dentener et al. 1999; Josse et al. 2004). This lack is mainly due to unavailability of uniform radon observation datasets.

In this paper, 2 years of surface measurements of radon activity concentration are analyzed together with meteorological parameters from a town in central Italy. Diurnal and seasonal radon evolution are analyzed in order to study its control mechanisms. A correlation analysis of radon concentration with surface wind speed proves that dynamical removal of soil emitted radon is one of the most important regulating factors of its abundance. As discussed in Di Carlo et al. (2007), the pronounced anticorrelation of radon with surface ozone is another indirect evidence of the strong link between small-scale convective mixing and the atmospheric surface layer accumulation of this tracer. To point out the role of soil moisture in reducing the radon ground flux, a model has been developed to calculate the actual soil water accumulation, by taking into account evapotranspiration, water runoff and precipitation. The nighttime tendency of radon is calculated during nights when its dynamical removal is absent or negligible (i.e., highly stable nocturnal boundary layer). Under these conditions, the above defined tendency can be regarded as a proxy of the emission flux. A comparison of the nighttime radon tendency with both direct precipitation and accumulated soil moisture is discussed.

This paper is organized as follows: in Sect. “Measurements”, a short description is reported of the site where observations have been taken, followed by a brief description of the instruments used and the diurnal and seasonal variations of radon and meteorological parameters. A brief description of the soil moisture model and discussion of the precipitation impact on radon emission are in Sect. “Soil moisture and radon emission”. The overall discussion of the results and conclusions are summarized in Sect. “Conclusions”.

Measurements

The data reported here are based on 2-year in situ observations in the atmospheric surface layer (2004–2005). The monitored quantities were radon activity concentration, temperature, relative humidity, wind speed/direction, precipitation and sun radiation. A brief description of the site and of the measurement techniques are given here; more details can be found in Di Carlo et al. (2007). The observations have been carried at the University of L’Aquila buildings, located about 3 km Northwest from a town in the central part of Italy (L’Aquila, 42°22′N, 13°21′E). The site is located in a valley at about 700 m above sea level, between the Gran Sasso mountain chain (the highest peak of Apennines, 2,912 m. s.l.m.), and the Sirente mountain chain (2,348 m. s.l.m.); it is mainly affected by continental air and it is far away from strong anthropogenic pollution sources.

Radon activity concentration is measured with a Silena model 5S instrument using a scintillation Lucas cell technique. Radon measurements are made on a 5-min base, from which hourly averages are calculated. The meteorological parameters are measured with a resolution of 5 s, from which hourly averaged data are calculated.

The in situ diurnal cycle of radon is triggered by soil emission and atmospheric dynamics (primarily small-scale vertical mixing), so that when the atmosphere is stable (mostly at night) the radon accumulates at the surface. After sunrise, when the turbulent vertical mixing restarts, the surface concentration rapidly decreases, even though radon is still emitted from the ground. A picture of the radon diurnal variation, calculated averaging the 2 years of data, is reported in Fig. 1a. The radon increase from about 20:00 to 6:00 is mainly due to its local surface accumulation, since the nocturnal stability of the boundary layer

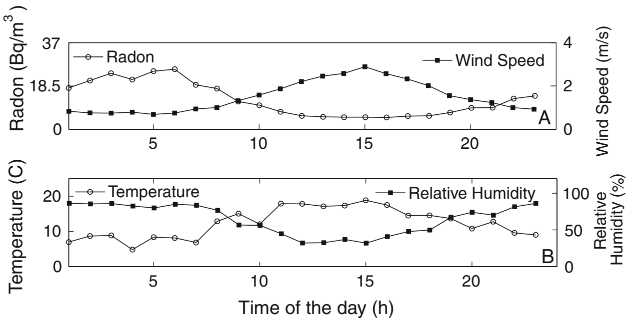

Fig. 1 Diurnal cycles of: surface radon and wind speed (*panel* **a**); temperature and relative humidity (*panel* **b**)

Table 1 Correlation coefficient between radon and meteorological and physical parameters

Radon and …	Year	Winter	Spring	Summer	Fall
Wind speed	−0.54 ± 0.02	−0.50 ± 0.04	−0.61 ± 0.03	−0.57 ± 0.03	−0.49 ± 0.04
Temperature	−0.47 ± 0.02	−0.49 ± 0.04	−0.52 ± 0.03	−0.72 ± 0.02	−0.43 ± 0.04
Relative humidity	0.37 ± 0.02	0.41 ± 0.04	0.25 ± 0.04	0.58 ± 0.03	0.58 ± 0.03
Wind direction	0.17 ± 0.02	0.09 ± 0.05	0.13 ± 0.04	0.34 ± 0.04	0.02 ± 0.05
Ozone	−0.47 ± 0.02	−0.58 ± 0.03	−0.62 ± 0.03	−0.55 ± 0.03	−0.54 ± 0.03

All the data reported in this table are hourly averaged

(very frequent in the L'Aquila site due to typical meteorological conditions, especially during summertime) efficiently inhibits radon removal by vertical mixing (and/or horizontal advection). The nighttime radon activity concentration measured in the L'Aquila site and its time tendency is usually controlled by soil emissions, except of course for (relatively infrequent) unstable conditions of ventilated nocturnal boundary layer. The role of wind speed (Fig. 1a) is evident looking at its diurnal variation that is exactly anticorrelated with radon. During nighttime ($v < 1$ m s^{-1}) the radon activity concentration increases due to a positive net balance between soil emission and dynamical removal, while during daytime the increase of the wind speed (up to 2–3 m s^{-1}) is an indicator of a negative net balance between emission and dynamical removal via dilution in the boundary layer. Temperature and relative humidity are presented in Fig. 1b; both show the expected diurnal variations, that are negatively (T) and positively (RH) correlated with radon.

A quantitative representation of the influence of meteorological parameters on radon variations is given in Table 1, where correlation coefficients of radon with wind speed, ozone, temperature, relative humidity and wind direction. All these data are hourly averaged and divided on annual and seasonal scales. Wind speed on annual scale has the largest correlation coefficient (−0.54 ± 0.02), for the role of ventilation on the surface accumulation of radon (see above). Negative/positive correlations of temperature/relative humidity with radon (−0.47 ± 0.02 and 0.37 ± 0.02, respectively), are expected due to the night/day cycle of both quantities (driven by sun radiation). The clear anticorrelation with ozone (−0.47 ± 0.02 on annual base and −062 ± 0.03 during spring time) is also expected since this gas is efficiently removed at night by dry deposition at surface with little vertical mixing, whereas in daytime it may increase by both mixing with free tropospheric air and in situ photochemical production. Looking at the seasonal correlation coefficients, wind speed is important for radon variations in all seasons, with the highest correlation during spring (0.61 ± 0.03).

Horizontal advection is, in principle, as important as vertical mixing for the dilution of surface atmospheric radon. In particular, being close to the sea border, advection of oceanic radon-free air masses can have a major role in controlling the local radon variability. On the other hand, over an in-land site such as L'Aquila, the direction of horizontal winds is not expected to play a significant role. This has been verified with the data, by checking in the rose diagram for the absence of any preferential wind direction for all measured Rn concentrations, as well as looking at Table 1. On the other hand, the clear anticorrelation of horizontal wind speed with radon (Fig. 2) is, for mass continuity, an indirect indication of radon removal from the surface layer by means of vertical mixing. As an example, four 30-day blocks of data are reported, showing diurnal anticorrelation between radon and wind speed. Besides the pronounced diurnal cycles of anticorrelation, it is interesting to note that, during some periods of time, very low radon activity concentrations were continuously measured together with persistent high speed winds (day numbers: 54–56, 58–59, 103–105, 126–129, 330–333, 343–346, 350–353). During these time intervals, the absence of the typical diurnal cycle of wind speed is an indicator of persistent unstable conditions of the atmospheric surface layer that prevented nighttime radon accumulation (Di Carlo et al. 2007). Looking at the daily averaged data, the negative correlation of radon and wind speed is still clear (Fig. 3), with a correlation coefficient (−0.48) of the same order of magnitude of that using hourly averaged data. This suggests that radon could be used as tracer of atmospheric vertical motions also for daily averaged data.

The radon negative correlation with ozone was discussed in Di Carlo et al. (2007); here it was pointed out that the O_3–Rn highest anticorrelation found during springtime (−0.62 ± 0.03) could be consistent with the ozone background nature of the L'Aquila site. Spring months are the most favorable for coupling direct ozone transport with ozone photochemically produced by precursors that were accumulated in wintertime and then efficiently transported on synoptic scale during springtime (Monks 2000). Since surface ozone is primarily affected by vertical motions in the boundary layer (and by local photochemistry, which is, however, of secondary importance in the L'Aquila site),

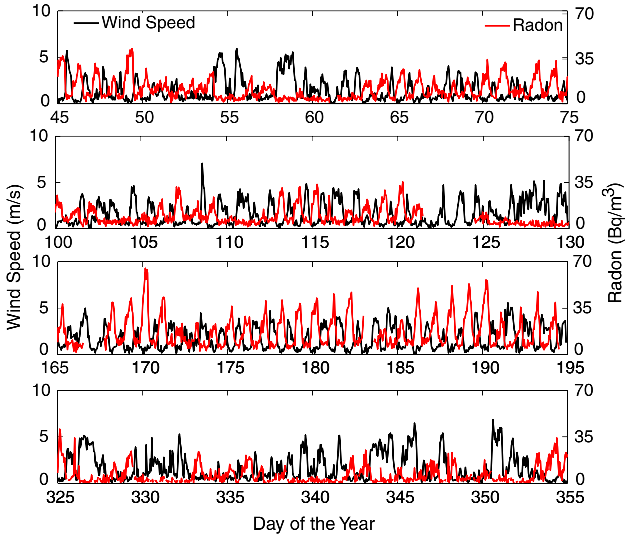

Fig. 2 Four blocks of 30-day radon activity concentration (Bq m^{-3}, *solid red line* right scale) and wind speed data (m s^{-1}, *solid black line* left scale)

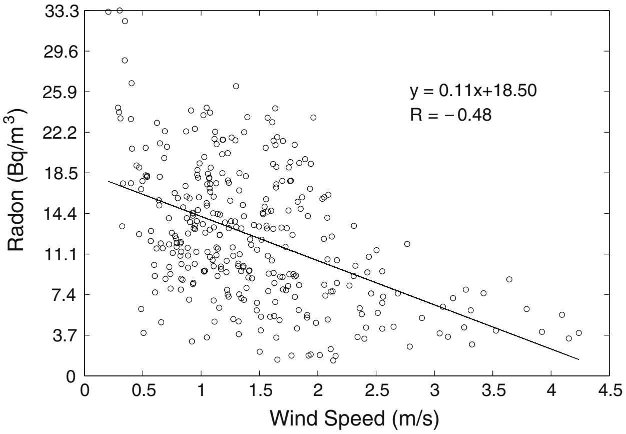

Fig. 3 *Scatter plot* of daily averaged radon activity concentration (Bq m^{-3}) versus horizontal wind speed (m s^{-1})

it is expected the O_3–Rn correlation could give an estimate of the level of stability of the boundary layer as for the Rn–wind correlation shown in Fig. 2. Time intervals without definite diurnal cycles of ozone and with persistent low values of radon are more or less coincident with those in Fig. 2, when horizontal winds are higher than normal at night. These winds, however, are only a first approximation indicators of the vertical motions (via mass continuity).

Soil moisture and radon emission

The evidence of a link between soil radon emission and precipitation events is not evident using direct correlations in the L'Aquila site. This is something observed also elsewhere (Galmarini 2006) and it could be explained with the short duration of intense precipitation events (excess of water runoff). Spectral analysis has helped understand the influence of precipitation on radon emission fluxes (Galmarini 2006). In the present paper, this link is explored using a model to account for the effective soil water content. The actual soil content of water is calculated using the meteorological data measured in the L'Aquila site. The calculation is based on the equation of mass conservation, so that the rate of soil accumulation of water is due to the difference between precipitation, evapotranspiration and runoff:

$$\frac{dW}{dt} = P - E - R \qquad (1)$$

where W is the soil accumulation of water (mm), P is the precipitation rate (mm h^{-1}), E is the evapotranspiration (mm h^{-1}) to take into account the amount of water that evaporates from soil and vegetation and R is the water runoff (mm h^{-1}) to take into account the amount of water removal due to overflow (underground runoff is neglected). The precipitation rate is measured, whereas the evapotranspiration is calculated using the Penman–Monteith method including measured sun radiation, temperature and wind speed (Allen et al. 2006). Water runoff is estimated using an adaptation of the USDA Soil Conservation Service (SCS) Curve Number (CN) technique (US Soil Conservation Service 2004). As an example, Fig. 4a shows the precipitation rate measured in L'Aquila during 2004 and Fig. 4b shows the calculated soil moisture content for the same year (Eq. 1). As expected, Fig. 4 suggests that the time-dependent amount of soil moisture is a combined function of the instantaneous precipitation and the lifetime of the accumulating moisture, which in turn depends on the surface wind speed and radiation budget (the latter being

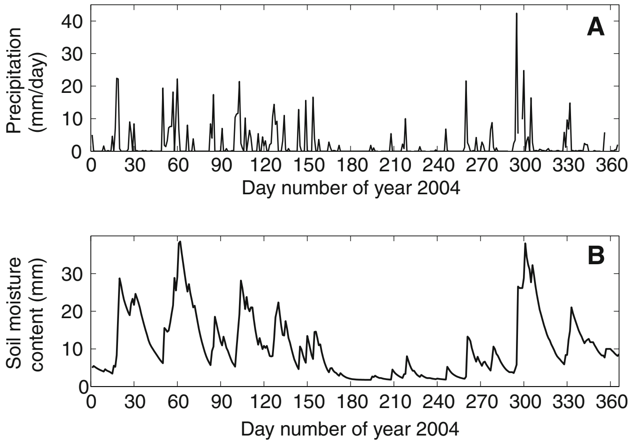

Fig. 4 *Panel* **a** Daily precipitation measured in L'Aquila (Italy) during 2004 (mm per day). *Panel* **b** Soil moisture content (mm), as calculated from Eq. 1 (see text)

much higher in summer months compared to winter). This effect is expected to be potentially relevant for the soil emission flux of radon, which may be affected by the presence of ground water, snow or ice.

The nighttime radon tendency in the atmospheric surface layer (d[Rn]/d*t*) is related to soil emission and atmospheric sinks by means of the following continuity equation:

$$\frac{d[\mathrm{Rn}]}{dt} = \frac{\Phi}{\Delta z} - (L_{\mathrm{mix}} + L_{\mathrm{decay}})[\mathrm{Rn}] \qquad (2)$$

where Φ is the emission flux, L_{mix} is the tracer removal rate from the surface layer due to convective small-scale mixing, Δz is the depth of this layer and L_{decay} is the removal rate from radioactive decay (normally negligible with respect to L_{mix}). Operating in dynamical conditions for which $L_{\mathrm{mix}} \sim 0$, the radon flux is easily obtained from Eq. 2 once the time tendency d[Rn]/d*t* is known. Following Galmarini (2006), it is estimated from nighttime radon activity concentrations, selecting only those nights when local wind speed is less than a fixed threshold (1 m s^{-1} in this case), as an indicator of the stability of the nocturnal atmospheric surface layer ($L_{\mathrm{mix}} \to 0$). During these selected nights, the nighttime radon tendency is calculated as the slope of the radon activity concentration between 8 pm and 6 am and Δz represents the depth of the thermal inversion layer above the surface, where the hypothesis of $L_{\mathrm{mix}} \sim 0$ is verified (about 50 m).

No anticorrelation is found between the nighttime radon tendency and precipitation (−0.15, see Fig. 5a); this result is not surprising, since the main physical parameter affecting the ground release of radon is expected to be the soil accumulation of moisture, which in turn does not instantaneously change with precipitation. Figure 5c clearly show that the anticorrelation of the nighttime radon tendency with the calculated soil water content (see Eq. 1) is much better defined. A rough indirect proof that the lifetime of ground surface water needs to be taken into account is given in Fig. 5b, where the radon tendency in any given “stable” night is correlated with the average precipitation rate during the previous 24 h; the correlation coefficient is larger than in the case of “simultaneous” precipitation (−0.38 vs. −0.15), but still lower than in the case when sinks of surface water (evapotranspiration and runoff) are taken into account together with time accumulation of soil water from precipitation (−0.54).

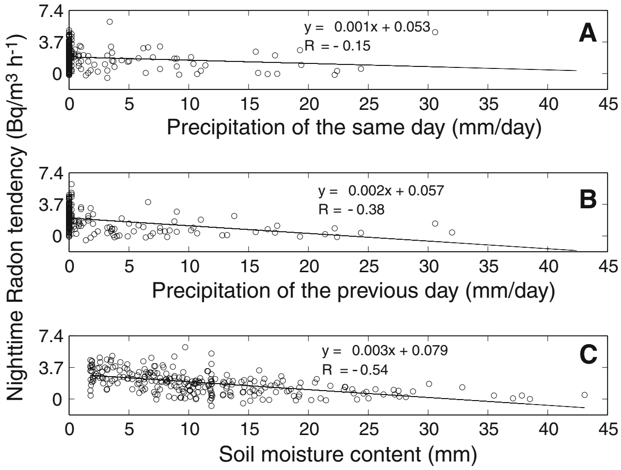

Fig. 5 *Scatter plots* of nighttime radon tendency (Bq m^{-3} h^{-1}) with: same day precipitation (mm per day) (*panel* **a**); previous day precipitation (mm per day) (*panel* **b**); calculated soil moisture content (mm) (*panel* **c**)

Conclusions

In this work, 2-year measurements of surface radon activity concentration have been analyzed along with meteorological parameters, in order to study the impact of meteorological variability on the dynamical mixing and dilution of atmospheric surface radon and on its emission from the ground surface.

It has been found that the radon activity concentration is well anticorrelated with the horizontal wind speed, confirming the hypothesis that radon is a good indicator of the small-scale stability of the atmospheric surface layer.

The instantaneous concentration of radon activity in the atmospheric surface layer is controlled by a coupling of soil emission and convective mixing. If appropriate conditions of negligible vertical mixing are assumed, the time changes of the radon surface concentration may be expected to be largely driven by the magnitude of the emission flux alone. Stable nocturnal conditions are ideal to test this potential direct link.

Precipitation is considered to be able to affect the surface radon evolution; in the L’Aquila site, however, hourly data do not show a significant negative correlation. On the other hand, looking at the radon time tendency in dynamically stable nights, a reasonable correlation with soil water accumulation is evident (Fig. 5c). In addition, Fig. 5 suggests that it may be inappropriate to use instantaneous precipitation as a proxy for the emission flux reduction, at least in this measurements site. A simple calculation of the soil moisture from precipitation, evapotranspiration and water runoff gives a better parameter for an estimate of the radon flux reduction during wet periods. This result could help improve the radon predictions in global models. Measurements in other sites could be valuable to confirm these results.

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