Description and Validation of a Numerical Box Model for Near-Surface Atmospheric Radon

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Abstract: - A numerical box model has been developed to better understand the physical drivers of the diurnal and seasonal variability of Radon-222 in the atmospheric boundary layer and to assess the role of different mechanisms controlling the tracer abundance immediately above the Earth surface. Dynamical removal due to turbulent convective motions is found to be the dominant controlling process. Since the latter is highly dependent on atmospheric meteorological conditions, a realistic representation of the small-scale convective mixing term in the mass continuity equation has been obtained by constraining the box model with meteorological parameters collected in a radon measurement site, located in the town of L'Aquila in central Italy. Model results are finally validated by direct comparison with hourly observations of radon taken in this site: a correlation coefficient of 0.8 is found between measured and modelled radon hourly values over two years of data. It is also shown how the model can be used to infer to the magnitude of the monthly averaged radon soil flux in the L'Aquila measurement site. Radon data collected during March 2009 have also been analysed to find possible signs of perturbation due to the on-going seismic activity that would have reached its peak in the April 6th, 2009 destructive earthquake. No significant radon activity increase was observed in L'Aquila at that time with respect to a previous 'seismically-unperturbed' year, during the same month with similar meteorological conditions, nor any statistically significant increase (or change) of the model derived radon soil flux.

Key-Words: - Box model; Mass continuity equation; Meteorology; Atmospheric boundary layer dynamics and transport; Atmospheric Radon.

1 Introduction

Radon (222Rn) is a radioactive noble gas emitted mainly by soils. The radioactive decay (mean lifetime of 5.517 days) acts as a net global radon sink. Local radon concentrations are mainly controlled by soil emissions and transport, since radioactive decay is relevant only on large spatial domains. 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 [1], so the balance of its sources and sinks is much simpler than for other atmospheric trace species and pollutants. A correlation analysis of radon concentration with surface wind speed shows that dynamical removal of soil emitted radon is one of the most important regulating factors of its abundance [2]. Another indirect evidence of the strong link between small-scale convective mixing and the atmospheric surface layer accumulation of this species is the pronounced anti-correlation of radon with surface ozone [3]. 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, PM, O_3) and is also comparable with timescales of convective motions [4]. This makes radon a good tracer to study the variability of ozone and other trace gases [5] [6] [7]. Observations of 222Rn have also been extensively used for calibration and validation of atmospheric transport models [8] [9].

Atmospheric radon receives attention and need of observations and modelling, due to its well-proved impact on human health. Indoor radon measurements are even more important from this point of view [10] [11]. According to EPA estimates, radon is the second leading cause of lung cancer, due to exposure to ionizing radiation. Radon-222 decades by emission of an α particle into

a series of short lived radioactive progeny, which are solid and normally deposited on fine-mode atmospheric aerosols. Once inhaled, they tend to be deposited on the bronchial epithelium, thus exposing cells to α irradiation [12] [13].
The geographic variability of radon emission is not well known due to sparse measurements [14] [15] [16]; the global average emission rate, on the other hand, is better understood using Pb-210 deposition measurements [17]. Most investigators conventionally apply a constant emission rate to all land surfaces, that will result in the observed deposition range (0.1 ÷ 2.5 Rn-atoms $cm^{-2}s^{-1}$, the most used value being 1 Rn-atoms $cm^{-2}s^{-1}$) [17] [18]. 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. [18] 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 soils. A study on the possible modulation of soil radon emissions due to surface rain accumulation has been made (among others) by Di Carlo et al. [19] and Megumi and Mamuro [20].
As a consequence of these emission and removal processes, the radon concentration shows (on average in a given location) a typical diurnal and seasonal cycle, with maxima at night and during the summer season. In the L'Aquila site, for example, long time series averages show maxima radon activity of about 35 Bq/m^3during summer nights, versus about 15 Bq/m^3 during winter nights, and typical 5 Bq/m^3 values during daytime hours. These diurnal and seasonal cycles are similar to those obtained from much longer and better established time records [6].
In this paper, outdoor radon measurements collected at the Department of Physical and Chemical Sciences of the University of L'Aquila (Italy) are analysed together with meteorological parameters and concentration of chemical tracers. Simultaneous ozone measurements have been used, because the ozone concentration in a site with a low level of anthropogenic emissions (as L'Aquila) is normally well anti-correlated with the concentration of radon [3]. Diurnal and seasonal tendencies of radon are discussed, in order to better understand the control mechanisms of its surface abundance. Besides data collection, the main scientific activity was on the interpretation of the data in the atmospheric environment. This was done by developing a numerical box-model for the release and dispersion of radon, driving the model with simultaneously measured meteorological variables. The purpose of this approach is to discriminate the processes of soil radon emission and its dispersion into the atmosphere, the latter being strongly correlated with higher or lower conditions of dynamical stability. The numerical model has been successfully validated using the radon measurements recorded in L'Aquila from 2004 to 2009. Among other parameters, the numerical model provides an estimate of the radon soil flux in the measurement site. The study of the temporal trend of the gas is essential in order to quantify any significant potential perturbation.
Possible sporadic radon soil emissions associated to seismic activity may also contribute to the variability of the atmospheric radon abundance on small space-time scales. Cicerone et al. [21] have summarized several observations of changes in gas emissions associated with 107 earthquakes. They conclude that there does not appear to be any diagnostic behaviour of either the beginning or the end of a radon anomaly that gives a consistent clue about when an earthquake is going to happen and have demonstrated no significant correlations between radon anomalies and a series of relevant seismic parameters. On the other hand, geologic heterogeneity can lead to strong spatial and temporal variations of the radon emissions, not associated with tectonic processes. Pre-seismic anomalies do not offer sufficiently robust statistical support to the hypothesis that radon could be used as a diagnostic precursor [22].
In the weeks immediately before the April 6, 2009 earthquake of L'Aquila (Italy), several events of magnitude in the range 2.0 – 4.0 were recorded with increasing frequency (30 events from March 1st to 31st), alarming residents, local administrators and the national civil protection. A specific risk-study committee was nominated by the national government, formed by scientists of national and international high-degree reputation in the field of earthquake studies and by representatives of the national civil protection. At the same time, the inhabitants were systematically alarmed by professed scientific forecasts based on hypothetical local measurements of radon, pointing out to a dramatic large increase of the gas, with respect to its "normal" background concentration values in the atmospheric surface layer in the L'Aquila area. This professed evidence was interpreted as a precursor signal of seismic activity and indicated as a warning for a possible destructive event near to come [23]. However, radon data collected at the University of L'Aquila during March 2009 do not show any significant increase with respect to March 2004, as

well as the indirectly calculated radon soil flux value [24].

2 Radon Measurements

Observations have been made outdoor on the flat roof of the University of L'Aquila building, about 4 km Northwest of L'Aquila downtown, in central Italy (42° 22' N, 13° 21' E) from 2004 to 2009. The site is located in the Aterno River valley at about 700 m above sea level (asl), between the Gran Sasso mountain chain (the highest peak of Apennines, 2,912 m asl), and the Sirente mountain chain (2,348 m asl); it is mainly affected by continental air and it is far away from strong anthropogenic pollution sources.

The monitored quantities, beyond radon activity concentration, were temperature, relative humidity, wind speed/direction, precipitation, incoming solar radiation and ozone mixing ratios. Radon activity concentration is measured with a Silena model 5S instrument, using a scintillation Lucas cell technique. Measurements are made on a 5 min base, from which hourly averages are calculated. The meteorological parameters are measured with a time resolution of 5 s, as well as ozone measurements using a UV-absorption analyser; hourly averaged data are finally 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) 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. The schematic cartoon in Fig. 1 summarizes the main physical mechanisms driving time and space variability of atmospheric radon in the planetary boundary layer.

Soil fluxes are of the order of 1 Rn-atom cm^{-1} s^{-1} on land (on average) and close to zero on snow or ice or on the oceans. Besides radioactive decay (lifetime of about 5.5 days), the main radon sink is due to turbulent small-scale convective mixing in the boundary layer, that is, however, highly dependent on atmospheric stability conditions. It is about zero on stable nights, while it is very efficient during daytime or on perturbed and ventilated nights, with a vertical mixing lifetime of the order of few hours.

Fig. 2 shows an example of the radon diurnal cycle and its anti-correlation with ozone and wind speed.

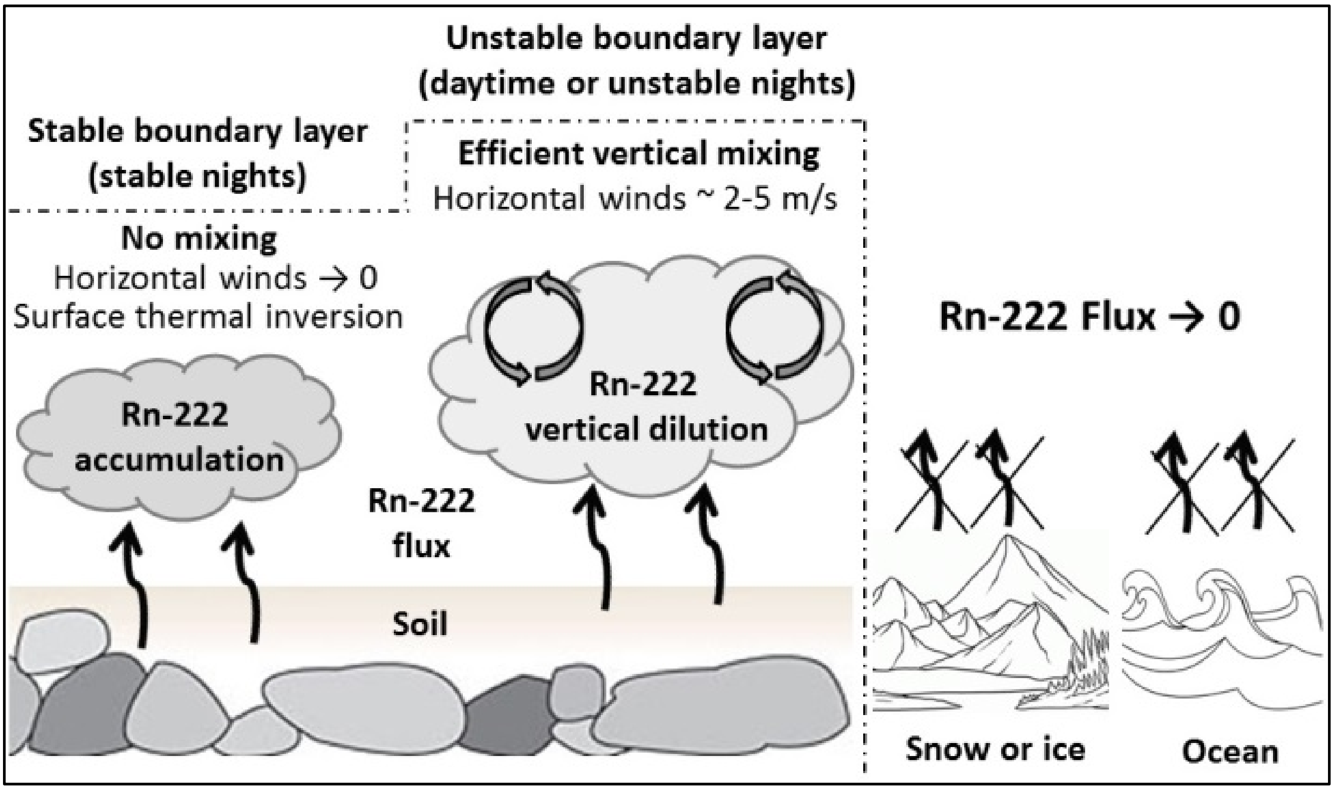

Figure 1. Cartoon of Rn-222 production and sink in the atmospheric boundary layer.

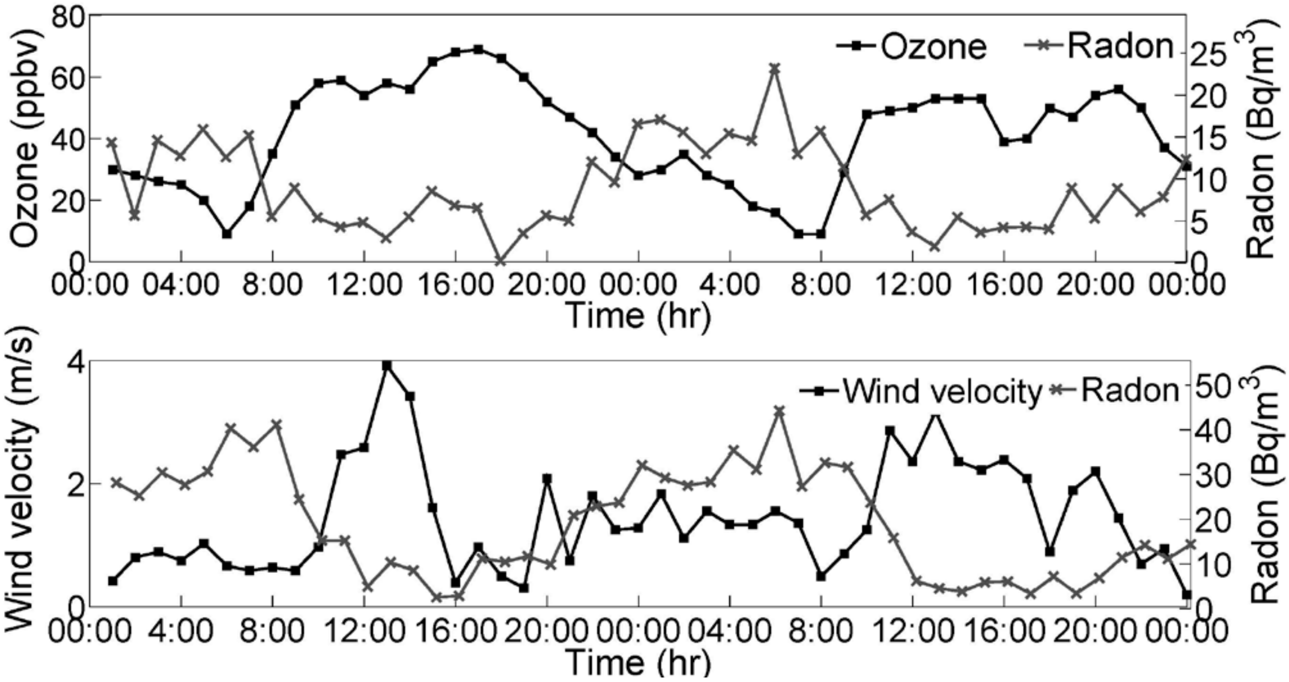

Figure 2. Top panel (a): diurnal cycles of surface radon (Bq/m^3) and ozone (ppbv), as measured on 11th - 12th May 2004. Bottom panel (b): diurnal cycle of surface radon and wind speed (m/s) as measured on 19th – 20th March 2004.

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 (very frequent in the L'Aquila) efficiently inhibits radon removal by vertical mixing (and/or horizontal advection). The night-time 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 ventilated conditions of the nocturnal boundary layer.

During night-time (wind speed less than about 1.0 m s^{-1}) the radon activity concentration increases due to a positive net balance between soil emission and dynamical removal. During daytime, the increase of the wind speed (up to 2–4 m s^{-1}) is an indicator of a negative net balance between emission and dynamical removal, via dilution in the boundary layer by means of vertical mixing (from mass continuity). Sporadically, there may be limited or absent night-time accumulation of radon due to unstable conditions of the atmospheric boundary layer. These are nights featuring absence of surface thermal inversion, that is typical, in turn, during meteorological high pressure conditions or in the absence of a significant cloud coverage associated to synoptic-scale perturbations.

In-situ measurements of ozone mixing ratio are another useful tool to better highlight the dynamical control on the surface radon abundance. Excluding dry deposition, that is rather constant during the day, and taking into account that the L'Aquila site is only marginally affected by in-situ photochemical ozone production (at least not during summer months), vertical mixing is the main parameter that controls the concentration of surface ozone. Therefore this trace species is another proxy to compare to radon levels collected in different years. Radon is highly negatively correlated with ozone (see as an example Fig. 3, for April 2004), with correlation coefficients ranging between -0.56 and -0.70 (see Table 1).

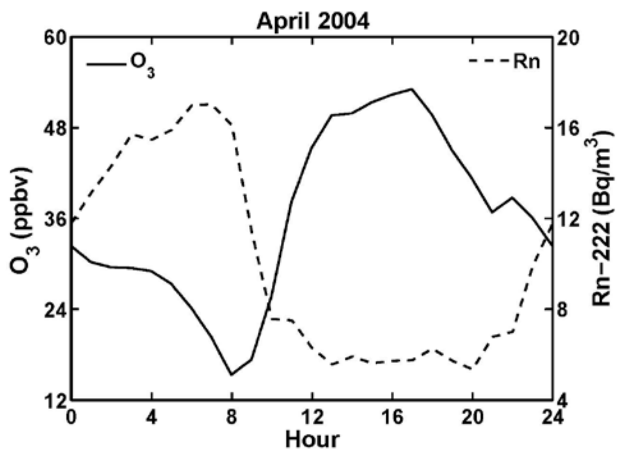

Figure 3. Diurnal cycle of near-surface radon (Bq/m^3, right axis) and ozone (ppbv, left axis), averaged over April 2004.

This confirms the radon ability to trace lower tropospheric vertical motions, which have in turn a big role in the variability of boundary layer ozone accumulation [3]. It should be noted how the anti-correlation is systematically higher during winter months with respect to summer, when the photochemical ozone production is not negligible.
The early morning sudden ozone increase shown in Fig. 2a is closely correlated with the abrupt radon decrease associated to small-scale convective mixing taking place after breaking up of the boundary layer thermal inversion. Background ozone (as the one normally observed in the L'Aquila site, at least in a first approximation) is a perfect tracer to be used as correlation tool with radon, since primary pollutants at the surface are largely controlled by emissions linked to anthropogenic activities (i.e. NO_x, CO, VOCs, PM).

Month	**Rn-χ_{O3}**
January	-0.63
February	-0.70
March	-0.68
April	-0.65
May	-0.67
June	-0.61
July	-0.56
August	-0.62
September	-0.65
October	-0.66
November	-0.62
December	-0.67

Table 1. Average correlation coefficients between hourly values of radon and ozone, per month.

Radon diurnal cycles averaged over July 2005 and January 2006 are reported in Fig. 4a, as an example of the more efficient night-time accumulation during summertime, which is normally dominated by stable high pressure conditions in the Mediterranean region. This effect sums up to the fact that winter months are not only characterized by a higher meteorological variability, but also by rather frequent conditions of frozen or snow-covered soils in the L'Aquila site, preventing radon release. The soil moisture content, much smaller on average from June to early November than during the rest of the year, is another factor capable of controlling the radon soil flux and its seasonal modulation [19].
Results in Fig. 4a are consistent with those reported in Jacob and Prather [6] and relative to a much longer dataset of observations in Chester (NJ, USA).
Radon abundance in an observational site may also be affected by horizontal transport [25] [26], as it is for other short-lived atmospheric tracers [27]. This is particular important for coastal sites when transport pathways from the ocean need to be taken into account, since these air masses tend to bring lower radon concentrations with respect to those from continental sites [28]. For this reason, the above discussed seasonal cycle could be very different on a coastal site, due to the summer impact of the sea breeze circulation, which tends (on a 24 hour average) to decrease the surface radon abundance.

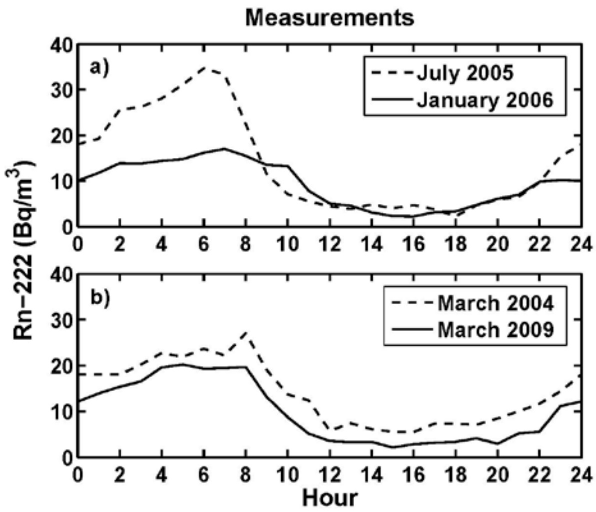

Figure 4. Top panel (a): average diurnal cycles of Radon concentration at L'Aquila, as measured during July 2005 (dashed line) and January 2006 (solid line). Bottom panel (b): as above, but for March 2004 and 2009, respectively

Fig. 4b shows the average diurnal cycles of radon observed in March 2004 and 2009, featuring comparable abundances (on average, 13.8 and 9.7 Bq/m^3 in 2004 and 2009, respectively). It is interesting to note that radon values are lower in March 2009 with respect to March 2004 (30% reduction, on average), even in presence of an already significant seismic activity in 2009. Analyses of the time series of radon, ozone and wind speed during March 2004 and 2009 prove that no significant meteorological differences had affected the boundary layer dynamics during these two months [24]. This is an important evidence that the similarity of radon levels observed during the seismic crisis in March 2009 with those of March 2004 should not be potentially attributed to significant differences in vertical transport and dynamical processes and therefore it is an indirect indication that no significant soil radon release changes were observed during the intense seismic activity in L'Aquila, with respect to an unperturbed background.

3 Box model description

A numerical box model has been used to better evaluate the role of different drivers of the near-surface radon abundance and its daily and seasonal variations. The most important feature of the model is that both production and sink terms are parameterized in terms of meteorological variables that have been measured simultaneously to radon and with instrumentation co-located with the radon analyser. The time tendency of radon (i.e. the time derivative of the Rn-222 near-surface mixing ratio, $d\chi/dt$) is calculated by means of the following continuity equation [24]:

$$d\chi/dt = -1/n_a \, d\Phi/dz - (L_{mix} + L_{decay})\,\chi \qquad \text{Eq. (1)}$$

where $\chi(t)$ is the near-surface radon mixing ratio, Φ is the soil emission flux, n_a the atmospheric number density and z is the altitude above ground level. L_{mix} and L_{decay} are the tracer removal rates from the near-surface box due to convective mixing-dilution in the atmospheric boundary layer and radioactive decay, respectively. Eq. (1) is applied to the box schematically shown in Fig.5: $-1/n_a \, d\Phi/dz$ is the flux divergence, equal to $\Phi/(n_a\Delta z)$ at the box centre ($\Delta z/2$) where the radon measurement instrument is located (20 m in the present case). The top boundary of the box (40 m) corresponds to a height above surface where negligible mixing occurs with the overlying atmosphere during stable nights with meteorological conditions of low wind velocity and surface thermal inversion.

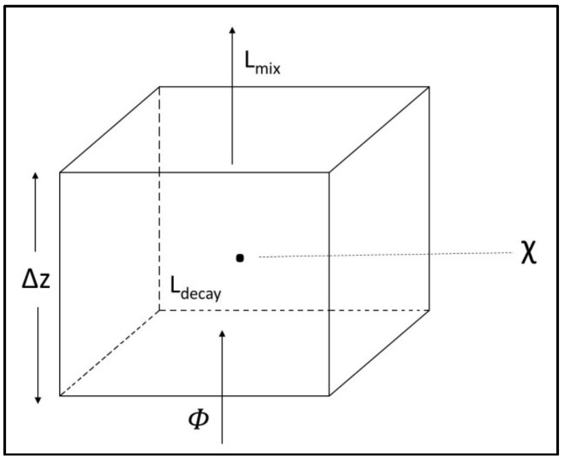

Figure 5. Cartoon of the box model scheme.

The radon observed time series is analysed by selecting only stable nights episodes with clear near-surface radon accumulation (normally 50% to 90% of total nights in the L'Aquila site on monthly basis, depending on seasonal meteorological variability). In these cases L_{mix} can be set to zero in Eq. (1), so that Φ can be easily obtained once the time tendency $d\chi/dt$ is known from the radon measurements [29].

The stability criterion is two-fold: local wind speed must not exceed a fixed threshold (0.6 m s^{-1} in this case), and a surface thermal inversion has to be observed ($dT/dz > 0.02$ K/m). The night-time radon tendency is calculated as the slope of the radon activity concentration for 10 hours before sunrise; Δz represents the depth of the thermal inversion layer above the surface, where the hypothesis of $L_{mix} \sim 0$ is verified (about 40 m). Table 3 summarizes the results of Eq. (1) applied to the radon time series for the selected stable night episodes. Minimum soil fluxes are found in winter, because of the non-negligible impact of frozen or snow-covered soil conditions during these months in the L'Aquila site. Maximum fluxes, on the other hand, are calculated during summer months, because radon exhalation is favoured by drier soil conditions and higher surface temperatures. The 1-σ uncertainty is associated to the different magnitude of the night-time surface temperature inversion (dT/dz), which produces variability in the actual nocturnal mixing depth around the average value of Δz=40 m, as measured by atmospheric radio-sounding profiles available in the L'Aquila site.

As the input for the production term in Eq. (1) (i.e., Φ) is calculated (on monthly basis) from the knowledge of temperature and wind fields, the radon removal by vertical mixing (L_{mix}) is parameterized in the box model as a function of a vertical diffusive flux divergence term. In turn, the diffusion coefficient K_z is modulated by the diurnal cycle (i.e., function of local small-scale convective activity): it is set to zero during stable nights (see above), otherwise is calculated as a linear function of temperature and wind speed. This results in rather large daytime values of L_{mix} (time constant of about 1-2 hours), as expected due to small-scale convection forced by surface warming after sunrise.

Once Φ and L_{mix} are known, using monthly and hourly mean values (respectively) derived from the above described parameterizations in terms of meteorological variables, Eq. (1) is solved in time for each month during years 2004 and 2005. The time derivative is solved with an explicit first-order Runge-Kutta scheme and using as initial condition the radon value measured at midnight of the first day of each month. The atmospheric number density n_a is calculated from the law of ideal gases, using measured hourly values of pressure and temperature.

Results of Eq. (1) are summarized in Fig. 6, using as examples February 2006 (a), May 2004 (b) and July 2005 (c). Here a collection of diurnal cycle sequences of Rn-222 is presented, comparing real observed data with box model results. A realistic behaviour of the box model is evident from Fig. 6 and Table 2, where the monthly averaged correlation coefficients are presented for observed and calculated hourly values of radon. These coefficients range between 0.72 and 0.85, with expected higher values during summer months with respect to winter. The cold season is in fact characterized by more pronounced meteorological variability and conditions of frozen and wet soils that are difficult to predict with the necessary accuracy in numerical models. It should be noted, however, how the model correctly predicts very low radon values for 5 consecutive days during February 2006, due to highly unstable meteorological conditions during those days and consequent absence of nocturnal accumulation (i.e., no surface thermal inversion and significant ventilation, with vertical mixing taking place in a much thicker layer than during stable nights).

Month	Rn_{obs}-Rn_{model}
January	0.75
February	0.77
March	0.72
April	0.75
May	0.85
June	0.81
July	0.83
August	0.84
September	0.84
October	0.80
November	0.84
December	0.75

Table 2. Average correlation coefficients between hourly values of observed and calculated radon activity concentrations, per month.

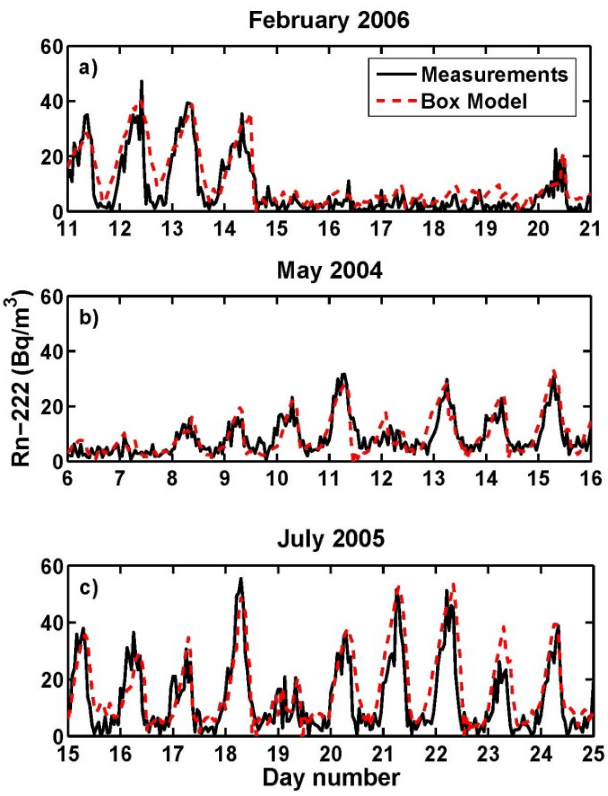

Figure 6. Validation of the numerical box model for the calculation of Radon activity concentration in the atmospheric surface layer at L'Aquila (Italy): time series of hourly data from 11 to 20 of February, 2006 (a); from 6 to 15 of May, 2004 (b); from 15 to 24 of July, 2005 (c). Black solid line is for observations; red dashed line is for box model results.

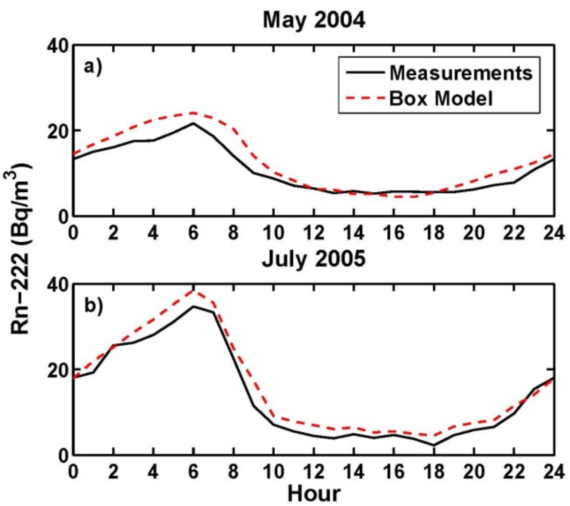

Figure 7. Diurnal cycle of Rn-222 for May 2004 (top panel (a)) and July 2005 (bottom panel (b)), with values from observations (solid line) and box-model calculations (dashed line).

Monthly averaged diurnal cycles are presented in Fig. 7 for May 2004 and July 2005 and compared to the results of the numerical box model, showing again a satisfactory behaviour. A compact picture of the box model performances is presented in two scatter plots of hourly data of measured and calculated radon values, for winter and summer conditions of year 2004 (Fig. 8). The majority of calculated radon values larger than 10 Bq/m^3 deviate by no more than ± 20% with respect to observations.

Pitari et al. [24] have shown that the observed diurnal cycles of Rn-222 for March 2004 and March 2009 (the latter immediately before the L'Aquila earthquake of April 6th, 2009) are similar each other, with the 2009 observed values being smaller by about 30% with respect to a seismic unperturbed period with similar meteorological conditions (March 2004) (see Fig. 4b). In order to completely clean up the radon observations from the effects of even small differences in winds and temperatures, the radon soil flux is calculated from the observed night-time radon activity tendency during March 2009 along with Eq. (1) (Φ=9.3 mBq $m^{-2}s^{-1}$, i.e., 0.44 Rn-atoms $cm^{-2}s^{-1}$) and is compared with the flux inferred from March 2004 data (Φ=11.2 ± 6.6 mBq $m^{-2}s^{-1}$, i.e., 0.53 ± 0.31 Rn-atoms $cm^{-2}s^{-1}$, see Table 3). This potentially 'seismically perturbed' flux is calculated in the uncertainty range of the above adopted flux calculation. This means that no statistically significant increase of the radon soil flux took place in the L'Aquila site during March 2009, before the earthquake.

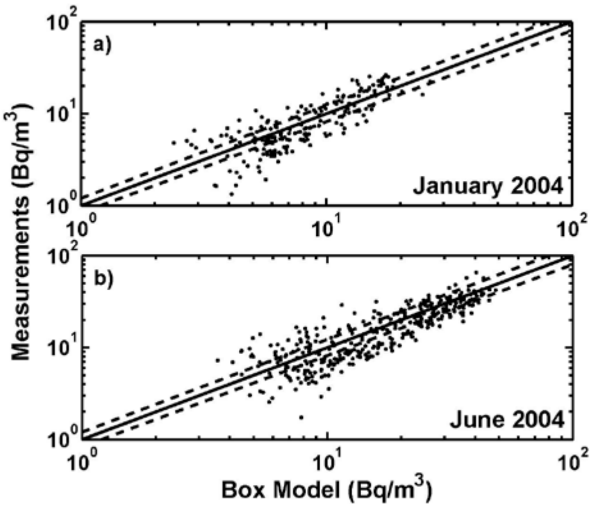

Figure 8. Scatter plot of hourly values of radon from observations (y-axis) and box model calculations (x-axis). Dashed lines highlight ± 20% deviations. Panels (a), (b) are for January and June 2004, respectively.

4 Conclusion

In this work, measurements of surface radon activity concentration made at L'Aquila (Italy) have been analysed together with ozone and meteorological parameters, in order to study the impact of meteorological variability on the dynamical mixing and dilution of atmospheric near-surface radon and on its emission from the ground surface. These observed data have also been used to find possible signs of perturbation due to the on-going seismic activity that would have reached its peak in the April 6th, 2009 destructive earthquake.

Month	Φ	σ
January	9.5	3.5
February	10.8	6.6
March	11.2	6.6
April	13.4	6.1
May	17.9	6.6
June	24.8	5.1
July	28.8	7.4
August	28.9	7.5
September	25.8	8.9
October	18.5	7.0
November	15.2	5.3
December	12.9	8.6

Table 3. Monthly averaged radon soil flux and standard deviation inferred from Eq. (1) applied to night-time accumulation of the tracer (see text). Units are mBq $m^{-2}s^{-1}$.

As expected, radon is well anti-correlated with horizontal wind velocity and ozone concentration 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, time changes of the radon near-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.

A box model constrained with observed meteorological data has been used and validated with the available observations: monthly averaged correlation coefficients between observed and calculated hourly values of radon range between 0.72 and 0.85. As a case study, the box model has been used to produce an indirect estimate of the radon soil flux during the weeks immediately before the April 6th, 2009 destructive earthquake of L'Aquila and demonstrates the non-existence of any evident increase of it. If there had been in place a consistent release of radon associated with the seismic activity of that period, culminated in the large, destructive earthquake that occurred shortly thereafter, these increases would have been detected in the atmospheric surface layer. On the other hand, the study presented in this paper shows that radon was not increasing at that time in L'Aquila with respect to a previous 'seismically-unperturbed' year during the same month with similar meteorological conditions (March 2004). In addition it was observed to experience a 30% decrease. This conclusion is reached from a direct comparison of observed data, but also as a result of the previously validated radon box model constrained by actual meteorological data, where an indirect estimate of a 17% reduction of the radon soil flux is obtained from. This reduction, however, is shown to lay well inside the 1-σ uncertainty range; in other words, no statistically significant changes of the radon soil flux are demonstrated to have taken place in the L'Aquila site before the April 6th, 2009 earthquake.

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