Modeling of radon exhalation from soil influenced by environmental parameters

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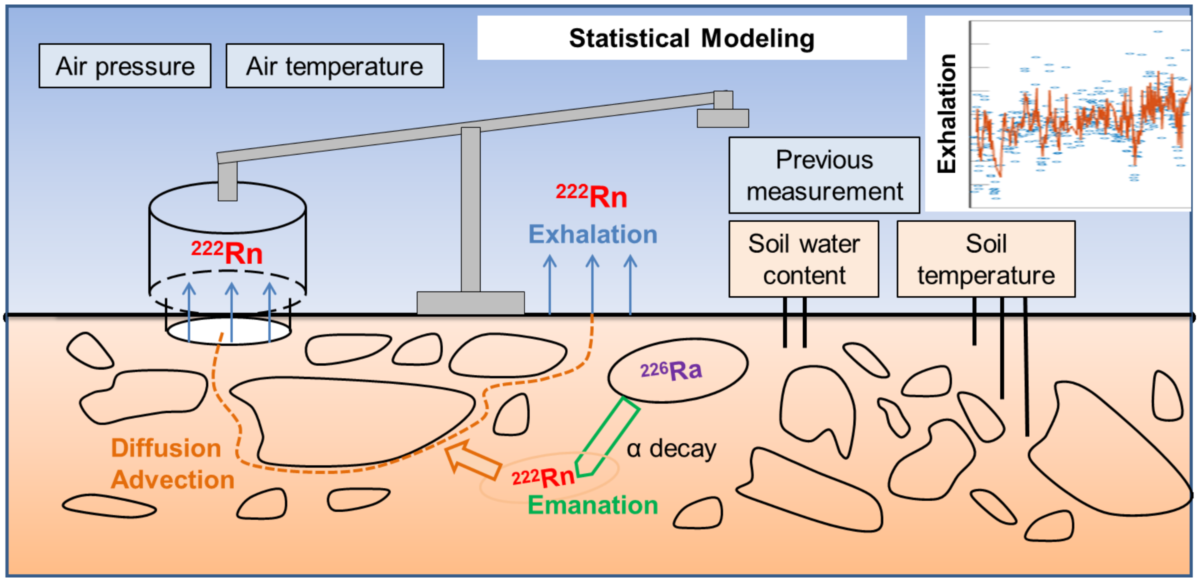
ABSTRACT

Atmospheric radioactive noble gas radon (Rn-222) originates from soil gas exhaled in the atmospheric surface layer. Radon exhalation rates from soil as well as corresponding meteorological and soil parameters were recorded for two subsequent years. Based on long-term field data, a statistical regression model for the radon exhalation and the most important influencing parameters soil water content, temperature of soil and air, air pressure and autocorrelation of the exhalation rate was established. The fitting result showed that the multivariate model can explain up to 61% of the variation of the exhalation rate. First, the exhalation rate increases up to 80 Bq m^{-2} h^{-1} with increasing soil water content. Later, at water content >10%, increasing soil wetness suppressed the exhalation rate: at values higher than 24% to approximately one third. The air temperature had a distinct positive effect while the soil temperature had a strong negative effect on the exhalation rate, indicating their different influencing-mechanisms on the exhalation. The air pressure was negligible. The lagged values of radon exhalation had to be included in the model, as the variable shows strong autocorrelation.

GRAPHICAL ABSTRACT



Keywords: Radon, Exhalation rate, Statistical model, Soil water content, Autocorrelation

Highlights:

- A two years continuous radon exhalation rate measurement was carried out.
- Soil and atmospheric parameters were recorded.
- The compound effects of soil water content on radon exhalation were corroborated.
- A statistical multivariate regression model using environmental parameters and autocorrelation was established.

1. INTRODUCTION

Radon (Rn-222) is one of the most important naturally occurring radioactive elements and contributes to more than half of the ionizing radiation exposure of humans (UNSCEAR, 2000). Its adverse health effect is well approved (WHO, 2009). Radon originates from mineral grains, which contain the parent nuclide radium (Ra-226) by recoil and diffusion mechanisms. A part of radon, which is produced mainly on the surface layer of the minerals can eject out of the grains and may emanate into the interstitial space between them. These radon atoms exist in gaseous form and spread into the pore space driven by diffusion and advection. They are dominantly transported by carrier fluids, whereas the radon migration depends upon the fluid flow characteristics of the soil. Some radon gas will eventually migrate upwards to the soil/air interface and exhales out into the atmosphere. The exhalation rate of soil radon gives the source strength of radon into the atmosphere. The characterization of this transfer process is crucial for the understanding of the following fate of the radioactive rare gas: either as trace substance in atmospheric dispersion or as accumulating contaminant in the indoor environment. This importance made it to an intense subject of investigations.

Former studies have revealed that the radon exhalation process is influenced by various environmental parameters. An overview is given e.g. by Nazaroff (1992) or recently by Sakoda and Ishimori (2017). Many investigators have observed that low soil water content promotes exhalation while abundant wetness depresses the exhalation (Schery et al., 1989; Stranden et al., 1984; Zhuo et al., 2006). Seasonal variations exist with higher exhalation rates in dry summers and autumns and lower ones in rainy winters and springs (King and Minissale, 1994). Stranden et al. (1984) have reported a weak positive effect of soil temperature on the exhalation rate and Iskandar et al. (2004) presented a formula indicating a positive linear correlation between radon emanation power and soil temperature. This is plausible since the increase of soil temperature can reduce the portion of radon adsorbed on soil grains. Therefore, the emanation is enhanced. Moreover, an increasing temperature can promote the radon diffusion process. However, most of the former studies have investigated only either the soil temperature or the air temperature. Note that under stable laboratory experimental conditions, the air and the soil temperature are nearly the same. It is unclear whether the influencing-mechanism of air and soil temperature are similar and in the same direction. But in the natural environment, the air temperature fluctuates more fiercely and may thus be rather different from the soil temperature. Therefore, models implementing both, air and soil temperature, would be necessary to fill the gap. In addition, the exhalation might be expected to increase when the air pressure decreases. The pressure difference is an important driving force of the gas transport in the indoor environment, where significant differences between in- and out-door air pressure occur..

Besides, soil properties such as the grain size as well as the radium content and its distribution in the grains, which is responsible for different emanation patterns (Chau et al., 2005; Chitra et al., 2018; De Martino et al., 1998), precipitation (Ferry et al., 2001; Müllerova et al., 2018) and air pressure (Koarashi et al., 2000) also play an important role in the exhalation process. It is well known that water in the soil space affects both, radon emanation and diffusion (Hassan et al., 2009; Sakoda et al., 2011). On one side, when the radon atoms eject out of the soil grain, water can help retain them in the pore space. On the other side, the radon diffusion coefficient for water is fairly smaller than that for air (Tanner, 1980) and the redundant soil water content would block the diffusion path and suppresses the exhalation. Overall, the promotion effect is dominant when the soil is relative dry; when the soil gets moist, the depression effect becomes prominent.

Most of the former studies were based on laboratory techniques and focused on single-variable effects. However, the laboratory experimental conditions, e.g. by means of disturbed sample material or controlled ambient conditions, could be far different from the field ambiance and would give rise to quite different results (Papachristodoulou et al., 2007). Moreover, the numerous influencing factors affect the exhalation not only directly, but also indirectly by modulating other relevant factors. Therefore, field measurements are needed to investigate how pertinent the impact of environmental parameters on the exhalation process is. Another advantage of *in situ* measurements is the possibility of performing long-term monitoring studies of the exhalation rate and the influence of seasonal variations on meteorological conditions and soil properties. In our study, a self-developed automatic measurement system called exhalometer (Yang et al., 2017), which is similar to a system used by Mazur and Kozak (2014), was applied for the continuous measurement of the Rn-222 exhalation rate from soils for two years, 2015 and 2016.

The aim of the investigation was to test the impact of the environmental parameters on the radon exhalation process. For the first time a two years time record of the exhalation rate is available for statistical analysis. In addition, environmental parameters as soil characteristics, soil water content and meteorological parameters were recorded over the same period. In a first step, the response of the exhalation rate to soil water content was modeled by a parsimonious 2-parameter Rayleigh function and the corresponding shape and scale parameters were estimated. In a second step, together with the remaining environmental parameters, the Rayleigh transformed soil humidity is then included in a linear multivariate autoregressive model of the radon exhalation. By this, the importance of single parameters to explain the radon exhalation variability as well as the forecasting performance of the multivariate model was elaborated.

2. MATERIALS AND METHODS

2.1 Study site, soil characterization and measurement of environmental parameters

Measurements were carried out on an open grassy field at the campus of Helmholtz Zentrum München, about 10 km north of the city of Munich (493 m a.s.l., 48°13'N, 11°36'E). The location can be described as a typical semi-rural area in southern Bavaria. The prevalent wind is from western direction and the amount of mean annual precipitation is 834 mm (1981-2010). The site is located on the Munich gravel plain, an up to 100 m thick Pleistocene glacial outwash plain that developed during the last three ice ages and covers 1500 km² of the Bavarian alpine forelands. Mainly calcareous gravels (<1-2% crystalline rocks) were transported by glaciers from the central and northern Alps and accumulated due to subsequent melt water transport on fluvial terraces in the alpine forelands. On site, in 8-10 m well-rounded gravels, shallow pararendzinas developed, mainly consisting of 10 cm humic topsoils and transitional horizons to the underlying unconsolidated bedrock material of maximum the same thickness. At the same site radon soil gas measurements were performed previously (Bunzl et al., 1998).

For a sedimentological characterization of the site and the analysis of the specific activity of Ra-226 in the sediments, four soil samples of 15-20 cm depth were collected arbitrarily around the measurement field (Table 1, sample no. 1-4). In addition, two depth-integrated samples were taken from a shallow soil profile separately for the humic topsoil (0-10 cm, no. 5) and the weathered bedrock horizon below (10-22 cm, no. 6). The sampling depths of the five subsoil samples correspond to the depths of the soil water content measurements. Bulk soil densities were determined by taking the weight of three volume-related samples, grain densities by applying the pycnometer method on the same samples (Flint and Flint, 2002). Porosities were calculated from the ratio of both densities. After drying the samples at 105°C in an oven, the fraction <2 mm was separated by sieving for the determination of the $CaCO_3$ content (DIN 18129, 2011) and specific activity of Ra-226 by gamma-spectrometry for all samples. Analyses for the grain size distribution according to ISO 11277 (2009) were performed on another two samples from 0-10 cm and 10-22 cm depth in the soil profile, referred to as no. 7 and 8 in Table 1.

The samples were transferred to plastic cups and left for three weeks before gamma spectrometric measurement to achieve a secular equilibrium of Ra-226 and its progeny. The Ra-226 activity was determined by integrating the areas of the full energy peaks at 186.2 keV and at the energies of the progeny Pb-214 (295.2 keV and 351.9 keV) and Bi-214 (609.3 keV and 1764.5 keV). Measurement times for the samples no. 1-6 were between 24 h and 15.5 d in order to diminish the analytical uncertainty to less than 8%. For the samples no. 7-8 uncertainties for the size fractions <2 mm were slightly higher, because the grain size analytical method provided only small amounts of sample material (2-5 g) for gamma-spectrometric measurements. Corrections for the self-attenuation according to Cutshall et al. (1983) and for the overlap of the energy peak of Ra-226 at 186.2 keV with that of U-235 at 185.7 keV were carried out.

Parallel to the continuous measurement of the radon exhalation rate (see chapter 2.2), the environmental parameters air temperature, soil temperature in 20 cm depth, air pressure, the amount of precipitation, humidity, wind intensity and direction were recorded at the measurement site. Soil water content was determined in 10-20 cm depth with an ECH_2O EC-5 sensor (Decagon Devices). It logged the volumetric water content by the dielectric constant of the media using capacitance/frequency (70 MHz) domain technology (e.g., Kizito et al., 2008; Kodešová et al., 2011) every minute. Hourly means were calculated for further evaluation. The accuracy was improved to 1-2% uncertainty by carrying out a calibration of the sensor with soil material from the study site at defined soil water content in the laboratory.

2.2 Exhalometer for the measurement of the radon exhalation rate

An automatic measurement system called exhalometer was developed for the long-term radon exhalation rate measurement during the years 2015 and 2016. It is based on the accumulation method. A bottom opened cylinder hood with diameter 40 cm and height 35 cm is adopted as the accumulation chamber. During the sampling time of 1 h, the accumulation chamber seals onto a collar which is inserted into the soil. The gas sample in the hood is transported into six Lucas scintillation cells successively. With the build-up curve of the radon concentration in the accumulation chamber, the exhalation rate E (Bq m^{-2} h^{-1}) can be determined according equation (1) through the subsequent measurement of the increasing radon concentration with time:

$$C(t) = C_0 + \frac{EA}{V}t \qquad (1)$$

where C (Bq m^{-3}) is the time-dependent radon concentration in the chamber, C_0 (Bq m^{-3}) is the initial radon concentration, A (m^2) is the area of the chamber bottom, V (m^3) is the volume of the chamber and t is the radon accumulation time (Yang et al., 2017). Due to the short half-life of thoron (56s), a low quasi-stable thoron concentration in the accumulation chamber will establish soon. As a contribution to C_0 it will not influence the radon exhalation results.

After sampling, the hood is lifted up and moves off the sampling area for three hours in order to keep the soil surface consistent with ambiance. Details about the experiment setup, measurement cycles and the calculation of the exhalation rates are given in Yang et al. (2017).

2.3 Data and statistical modeling

The radon exhalation is affected by a variety of environmental parameters and the amount and distribution of the radon's parent nuclide Ra-226 in the soil. In our study, the soil water content, soil temperature, air humidity, air temperature, air pressure, precipitation, wind speed and wind directions were recorded. Preliminary correlation and regression analyses indicated that the air temperature, the soil temperature in 20 cm depth, the soil water content and the air pressure most dominantly affect the radon exhalation rate compared to the other measured variables. Therefore, the applied statistical model is based on these four parameters and can be expressed as

$$E = \alpha_0 + \alpha_1 w + \alpha_2 T_s + \alpha_3 T_a + \alpha_4 P + \sum \beta_k E_k \qquad (2)$$

with E = exhalation rate, w = volumetric soil water content, T_s = soil temperature in 20 cm depth, T_a = air temperature, P = air pressure, α_i (i = 1, 2, 3, 4) = regression coefficients, E_k (k = 1, 2 …) = exhalation rate of the k^{th} previous measurement cycle and β_k = regression coefficients of lagged variables.

In the regression model, it is assumed that all variables have a linear relationship with the radon exhalation rate. Due to nonlinear dual influences of the soil water content, a linear fit is inappropriate in the regression model. In this case, the linear function is substituted by a Rayleigh type function. The Rayleigh function *R(w)* provides an elegant and parsimonious parametrization and is determined (up to an intercept) by the parameters *b* and *c*: the inflection point *c* is the location of the global maximum:

$$R(w) = E(w) = \frac{bwe^{\frac{1}{2}\left(1-\frac{w^2}{c^2}\right)}}{c} \quad (3)$$

where *E(w)* is the exhalation rate and *w* is the volumetric soil water content.

Except from the dependence on environmental parameters, the radon exhalation rate has a distinct autocorrelation. The measurement cycle of the radon exhalation rate in this study took four hours. Generally, all environmental parameters that might affect the exhalation process would not change too much within that period. Consequently, it is assumed that also the exhalation rate would not change drastically either. It is supposed, that the previous measurement can deliver some predictive value for the subsequent one. This implies that several previous data points can be used to estimate the next data point, which was applied for lag1, lag3, and lag5 (first, third and fifth measurement value before) in the autocorrelation analysis.

The data sets generated contain 1625 recorded values from January 2nd to December 31st, 2015 and 941 measurements from February 19th to October 17th, 2016. It sums up to four to five exhalation rate readings per day in 2015 and approximately four readings per day in 2016. For the data processing, statistical analyses, and results display, Microsoft Excel 2013, R 3.2.1, Origin 8 (OriginLab Corporation), Wolfram MATHEMATICA 10.4, and SAS/STAT software 9.4 (SAS Institute Inc., 2014) was used.

3. RESULTS AND DISCUSSION

3.1 Soil characterization

As the radon exhalation rate mainly depends on the amount and distribution of its parent nuclide Ra-226 in the sediments, soils at the measurement site are characterized by the analysis of typical sedimentological parameters, which confirms the fluvio-glacial origin of the accumulated sediments. The grain size analysis (Table 1) proves that the subsoil sediments (10-22 cm depth) are dominated by 68% gravel and 26% sand; they are very poor in silt and clay (6%). The content of clay is only about 1.5%. The separate analysis of the total fraction of fine material (<2 mm) defines the sediment as a slightly loamy sand. However, the under-representation of the fraction <2 mm results in a comparably high porosity and high water permeability. It also leads to short-term full water saturation of the soil immediately after strong precipitation events. Calcium carbonate contents are 33% in the topsoil and 37% in the layer 10-22 cm. Soil water contents in these two samples were 7.0% and 9.0%, respectively, on the day of sampling. The mean bulk and grain density was 1.39 g cm^{-3} and 2.45 g cm^{-3}, respectively. The densities are in the range of typical values for well-permeable sandy soils. The porosity is calculated with a high mean value of 0.43 and therefore is in the upper ranges for typical unconsolidated sediments.

The specific activities of Ra-226, being the parent nuclide of Rn-222, are shown in Table 1. The specific activities of the bulk soil samples <2 mm range between 50-131 Bq kg^{-1} indicating the heterogeneity of the Ra-226 distribution over the measurement site. The analysis of grain-size-fractionated samples no. 7 and 8 proves an increase of Ra-226 contents with decreasing grain sizes and increasing specific surface areas of each grain. Higher radon emanation factors for sediments with grain sizes <0.2 mm were already discussed by Chitra et al. (2018). It is worth noting that the measurements of medium and coarse gravels have higher gamma-spectrometric measurement uncertainties than those given in Table 1. The listed values for the specific activities depend on the mineralogical composition of single gravels that were selected for measurement due to limited space in the 250 mL calibrated measurement cups.

Differences in the specific Ra-226 activities between the samples of the two different depths in the soil profile are low. Since grain size distributions in both layers are already comparable, also a similar distribution of Ra-226 activities over depth can be expected.

3.2 Modeling results

The radon exhalation rate is relatively low for dry and wet soils and is relatively high for intermediate soil water content. In a first step, therefore, we modeled this behavior by a parsimonious 2-parameter Rayleigh function and estimated the corresponding shape and scale parameters b and c, respectively (see equation (3)). The non-linear Rayleigh-transformed soil humidity entails a much better overall model fit than just the direct (linear) soil humidity alone when comparing the Akaike Information Criterion (AIC). In a second step together with the remaining environmental parameters, the Rayleigh transformed soil humidity is then included in a linear multivariate autoregressive model of the radon exhalation. The approach is based on a parsimonious so to speak 'hybrid' partially nonlinear autoregressive model. More sophisticated and less parsimonious models yielded not much better results. Moreover, including lagged co-variable measurement values did not improve the model significantly.

3.2.1 Dependence of the radon exhalation rate on soil water content– univariate modeling

The measured radon exhalation rates in dependence of the volumetric soil water content are shown in Figures 1-3 for the years 2015, 2016, and for both years combined, respectively. The exhalation rates range up to 80 Bq m^{-2} h^{-1} with a mean of 25.3 Bq m^{-2} h^{-1} and therefore in the range of typical values that can be found in literature for short term measurements (NCRP, 1988, Porstendörfer, 1994, UNSCEAR, 2000). Using Equation (3) to describe the exhalation rate from the soil water content, the best estimates of the model parameters are shown in Table 2. The value of the soil water content inflection point *c* varies between 8.4% (2015) and 13% (2016) with an average value of 10% for both years 2015 and 2016 combined. Therefore, the soil water content inflection point is consistent with findings from other studies (Bossew, 2003; Hosoda et al., 2007; Schery et al., 1989; Zhuo et al., 2006). However, the R-square obtained by univariate modeling is rather low in the range of only 6-18%. The data are roughly divided into two stages. When the soil is relatively dry, the exhalation tends to increase along with the water saturation. After the inflection point at 8% in 2015 and 13% in 2016, the exhalation rate decreased to approximately one third with an increase in the soil water content to 24% (Figure 3). Water works to promote radon exhalation up to a certain water content and retains the Rn-222 afterwards. The effect of water content on the radon exhalation may be dominated by two processes. (a) Diffusion occurs in air-filled pores at low water contents and radon is distributed between air and water at equilibrium. The partition between gas and liquid phase depends on the relative volume of water in the pore space causing higher concentrations in the gas phase when the water content increases. In addition, emanating radon molecules from the soil grains have a greater probability to stay in the pore space if the density is higher with increasing water content. The radon concentration in the air-filled soil pore space is higher due to partitioning and increased emanation at higher water content.. (b) Diffusion occurs dominantly in water-filled pores at high water contents and air/water equilibrium only exists near interfaces. Under these conditions, the radon concentration in soil air can be low (Faheem and Matiullah, 2008).

3.2.2 Multivariate modeling

It is interesting to consider whether the quality of the fit function of the univariate model can be improved by including further environmental parameters as well as significant variables accounting for the strong autocorrelation in the exhalation rates (Figure 4). Therefore, the soil temperature in 20 cm (T_s), the air temperature (T_a), the air pressure (P), as well as lag1 (n-1), lag3 (n-3), and lag5 (n-5) (previous first, third and fifth) of the exhalation measurements were added to the regression model. Lag2 and lag4 both explained less variability and conveyed larger p-values than lag3 and lag5 in conjunction with the combined lag0 (= original exhalation) and lag1. Therefore only lag1, lag3 and lag5 were introduced. The resulting parameters and confidence limits for both years combined are compiled in Table 3. The R-square increases to 61%. Figure 5 presents pertinent regression diagnostics, which shows an overall reasonable fit quality together with approximately normally distributed residuals. In general the normal quantile-quantile plot demonstrates if the residuals are normal distributed. If this is the case, all residuals follow the diagonal line. In our case this is not given exactly for the lower and higher quantiles, but the middle region fits very well. Also the histogram for the residual distribution shows that it fits quite well to the theoretical normal distribution. Therefore, the fitting function can be improved as:

$$E(n) = 3.00 + 0.18 * R(w) - 0.62\,T_s + 0.52\,T_a - 0.0002\,P + 0.49\,E(n-1) + 0.14\,E(n-3) + 0.18\,E(n-5) \quad (4)$$

wherein *R(w)* is the Rayleigh transformed volumetric soil water content according to equation (3) and according the parameters in Table 2 for the combined years 2015 and 2016.

In Table 3, the insignificant level of the air pressure parameter indicates that the pressure is not important in the present data and the modeling context. This finding is supported by the investigation of Mazur and Kozak (2014) although their study was shorter (only one year). The reason for the insignificance of the air pressure might be the relatively slow change over a large area. Maybe the pressure difference between atmosphere and soil at a certain depth might be a better parameter. The closed accumulation chamber during sampling might reduce the influence of the external air pressure as well.

A rise of temperature has been thought to linearly increase the radon emanation (Iskandar et al., 2004). This may be due to a reduction in physical adsorption of radon onto grains that occurs during the diffusion through the porous material (Sakoda et al., 2011; Stranden et al., 1984). The fitting results show distinct positive and negative effects of air and soil temperature, respectively. This is unexpected and it may imply a different influencing-mechanism of air and soil temperature. The negative effect of soil temperature contradicts the experimental results of some other studies (Iskandar et al., 2004; Schary et al., 1989; Stranden et al., 1984). Nevertheless, the theoretical calculation model by Sakoda and Ishimori (2014) has obtained a similar negative effect. The reason for the negative effect of the soil temperature on the exhalation rate might be that the influence of soil temperature in the natural environment has a time delay. It takes some time for the radon gas to migrate to the soil surface. This delay may mask the real effect of soil temperature. Further research and data analyses are needed to explain the unexpected, however highly significant negative effect of the soil temperature.

The simulation results are compared to measured exhalation rates in Figure 6. It is obvious that after considering the autocorrelation lag1, lag3, and lag5, the forecasting performance of the model has been improved considerably. The R-square increased from 28% (Figure 6, upper graph) to 61% (Figure 6, lower graph). The simulated data is consistent with the measurement data, but the conformity weakens at very high and low exhalation rates >50 Bq m^{-2} h^{-1} and <10 Bq m^{-2} h^{-1}. The autocorrelation functions show distinct autocorrelation for all involved variables (Figure 7). For the independent variables the autocorrelation vanishes after a lag of approximate 100 measurements, which corresponds to approximately three weeks. In contrast, the independent exhalation variable vanishes only after 500 measurement cycles, which corresponds to approximately four months.

The predictive power of the model was tested as well with our model analogous to the 'hold-one-out' method. In the combined data from January 2015 to October 2016, a chosen calendar month is excluded (i.e. one or two months are discarded). Next, the model parameters are estimated from each of the resulting 12 reduced data sets and the correlations of the observed and predicted radon exhalation values for the excluded month(s) are computed. The results are presented in Table 4. Compared to the overall correlation of 0.78 between the observed and predicted values based on all data, the 'hold-one-calendar-month-out' method yields somewhat higher correlations for February to April, and partly considerably lower correlations for the other months. The correlations are especially low for January, November, and December since for these three months data were available for the year 2015 only.

4. CONCLUSIONS

Based on the field measurement data of 2015 and 2016, a statistical multivariate regression model involving soil water content, soil and air temperature and air pressure was established to fit the radon exhalation from soils. This model can explain about 61% of the exhalation variation. As the radon exhalation showed a strong autocorrelation, its implementation improved the model tremendously.

The fitting model corroborates the compound effects of soil water content. The radon exhalation rate increases until the soil water content exceeds about 10%. At higher soil wetness, the exhalation decreases gradually. The model also revealed opposite effects of air and soil temperature on the exhalation rate, which implies their possible different influencing-mechanisms. The negative effect of soil temperature is contrary to some former studies and suggests that a time delay effect might exist, which is not visible in laboratory studies where air and soil temperature are similar. Further experiments and time series analyses testing the influencing mechanisms are needed.

Figure 5 and Figure 6 show that lower or higher extreme values of the radon exhalation are less well represented by our model compared to the intermediate radon exhalation rate measurement data. This might be due to some nonlinear influences of the independent variables, which aspect may be a topic for further research. The correlations in the last column of Table 4 suggest that the model and its predictive power may possibly be significantly improved if data is available for more extended periods of time.

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Figures and Tables

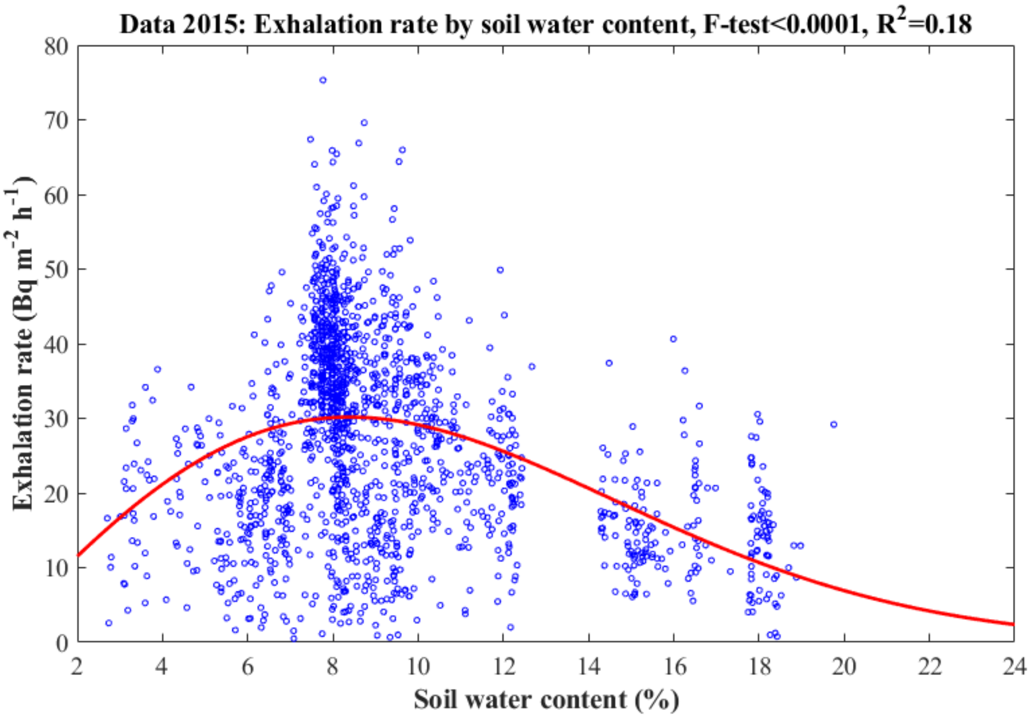


Figure 1. Variation of the radon exhalation rate with soil water content, data of 2015.

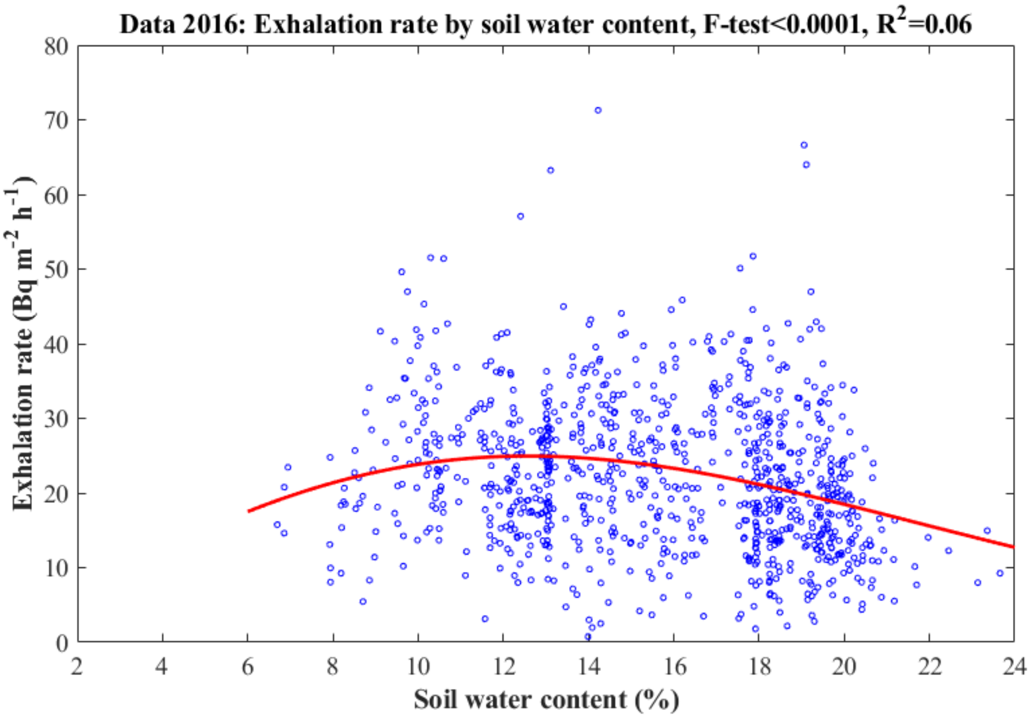


Figure 2. Variation of the radon exhalation rate with soil water content, data of 2016.

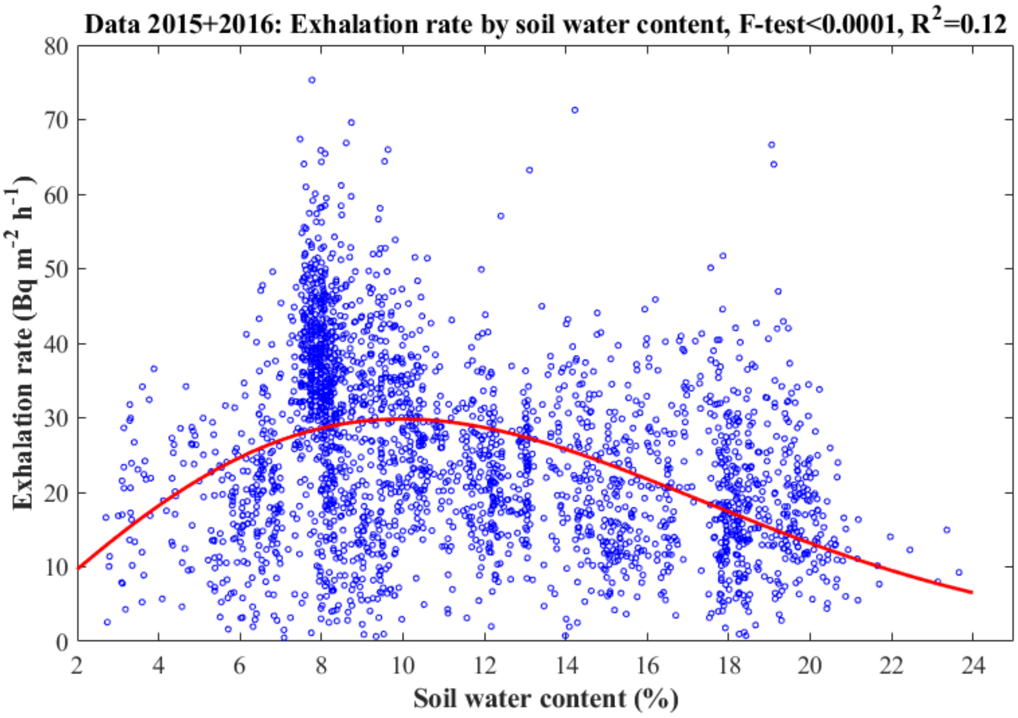


Figure 3. Variation of the radon exhalation rate with soil water content, data of 2015 and 2016.

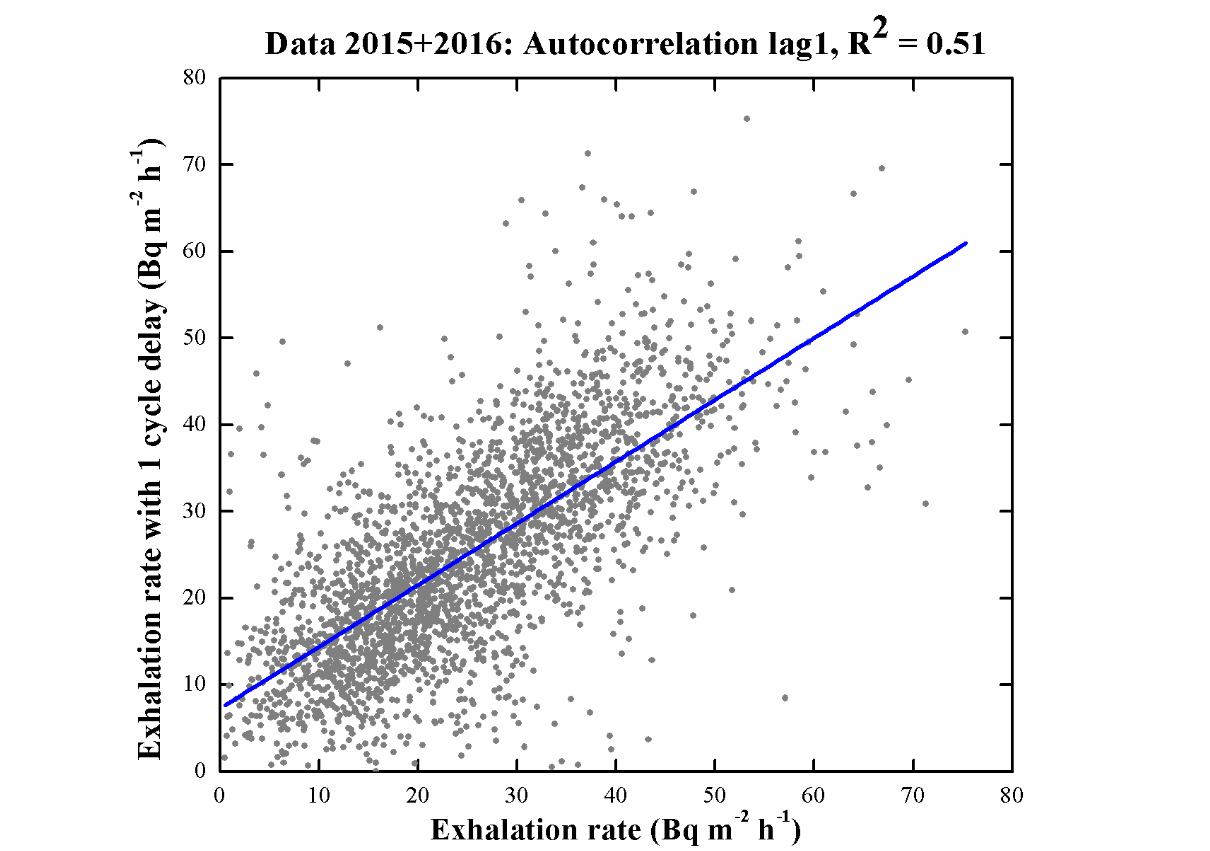


Figure 4. Autocorrelation scatter plot of the exhalation rate measurements versus the exhalation rate with lag1 (data of 2015 and 2016).

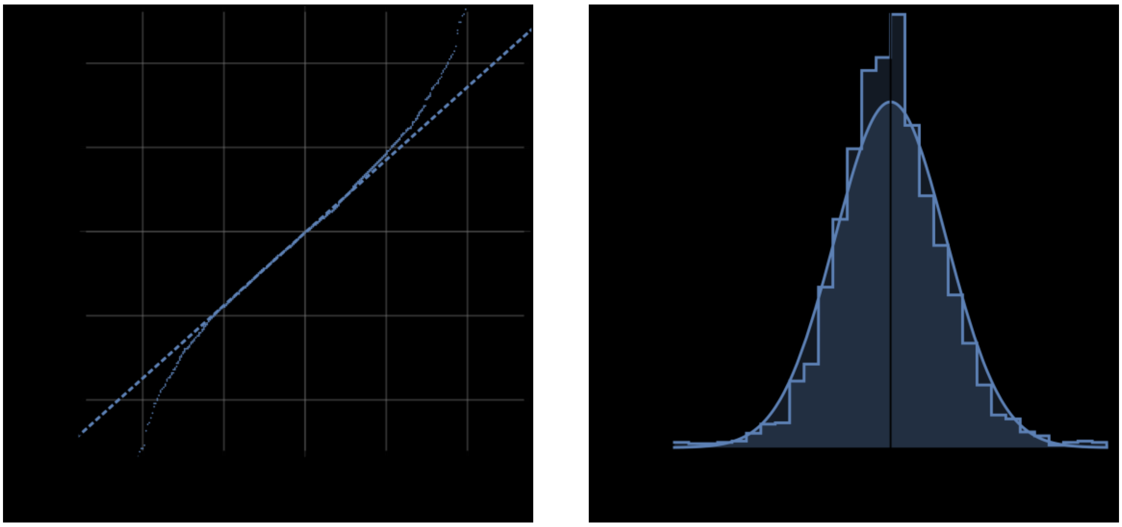
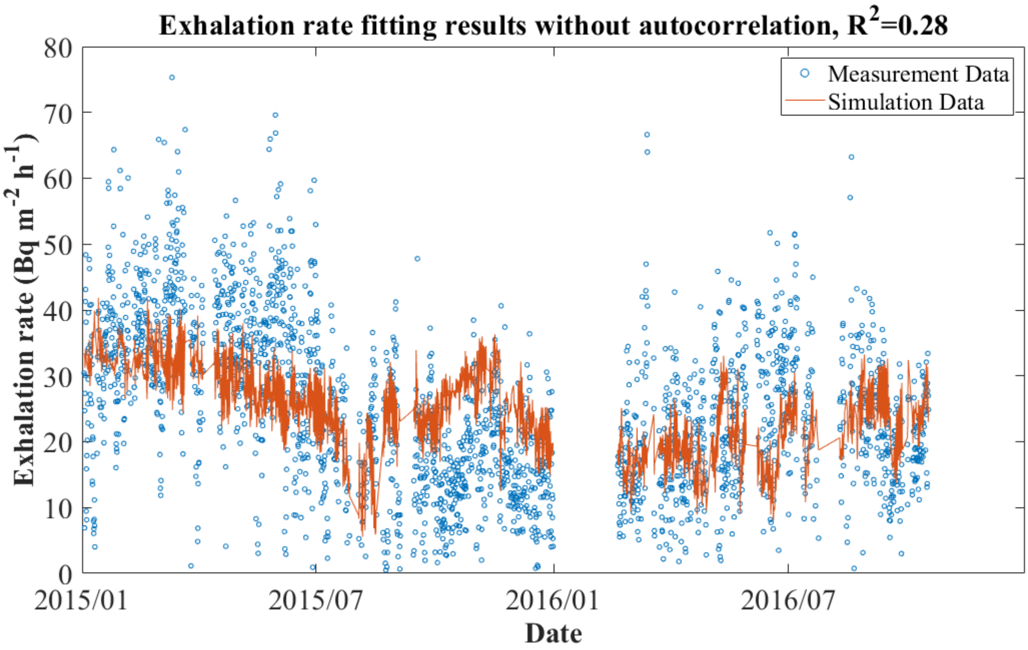


Figure 5. Fit diagnostics for the multivariate time-lagged environmental exhalation model with parameters listed in Table 3; left: quantile-quantile plot of the residuals; right: histogram distribution of the residuals compared to the fitted theoretical normal distribution.



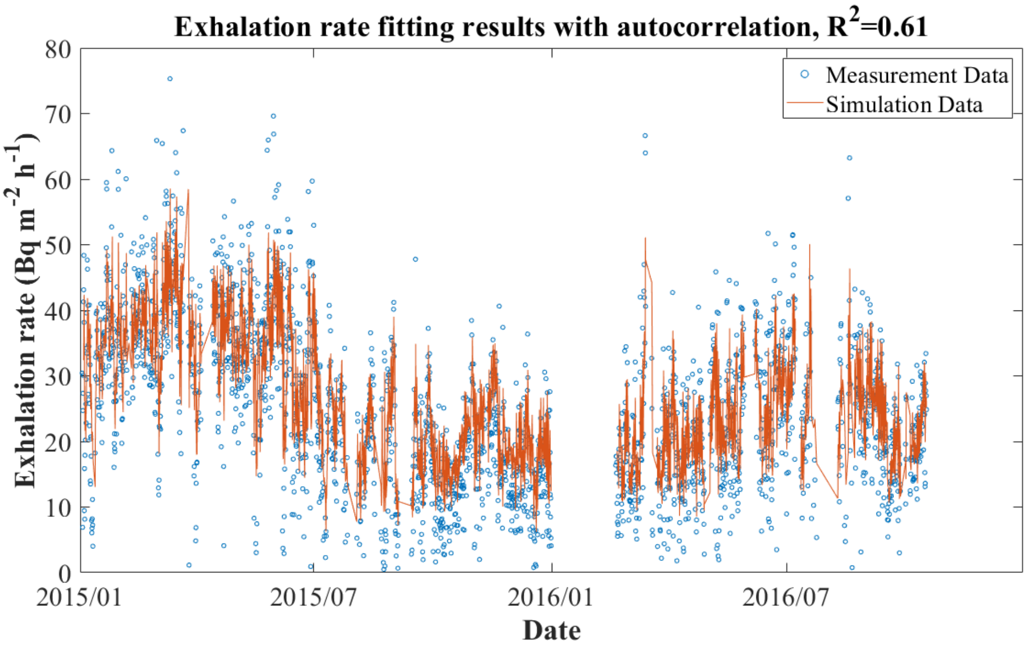


Figure 6. Simulation result of the multivariate time-lagged environmental exhalation model in comparison to the experimental data. In the upper graph the simulation is shown without considering the autocorrelation lag1, lag3 and lag5. In the lower graph the autocorrelation was integrated in the model: the simulated data agree quite well with the measured data (except for the extreme values) and R-square increased to 61%.

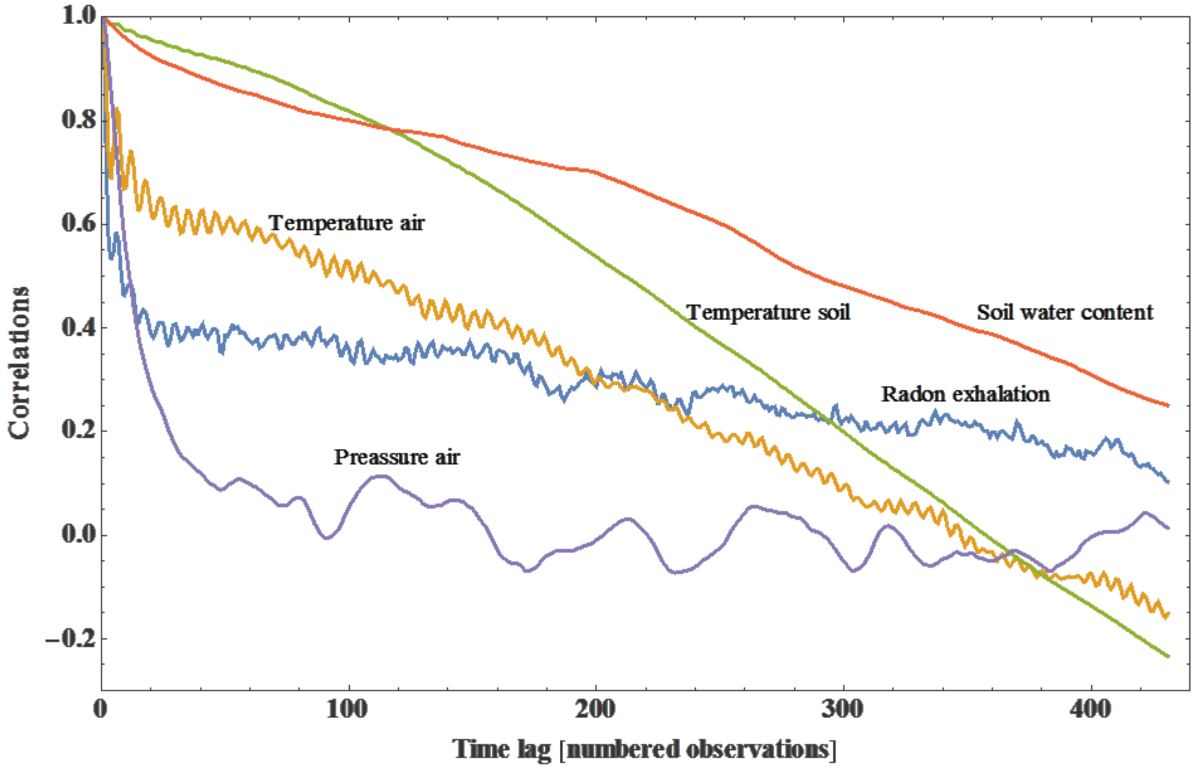


Figure 7. Autocorrelation functions for the radon exhalation and the environmental parameters studied.

Table 1. Grain size analysis and measured specific Ra-226 activities in the bulk fine material < 2 mm (no. 1-6) and the size-fractionated soil samples (no. 7 and 8).

Soil Sample No. (Depth)	Grain Size (mm)	Total Soil Mass (%)	Ra-226 Specific Activity (Bq/kg)	Ra-226 Uncertainty (Bq/kg)
1 (15-20 cm)	<2		91.7	3.0
2 (15-20 cm)	<2		130.8	10.3
3 (15-20 cm)	<2		59.2	2.1
4 (15-20 cm)	<2		92.2	4.3
5 (0-10 cm)	<2		49.7	1.9
6 (10-22 cm)	<2		51.9	1.7
7 (0-10 cm) 8 (10-22 cm)	coarse gravel (20-63)	42.1 22.6	15.7 16.2	0.7 0.7
7 (0-10 cm) 8 (10-22 cm)	medium gravel (6.3-20)	25.7 27.7	23.1 23.8	1.0 1.1
7 (0-10 cm) 8 (10-22 cm)	fine gravel (2-6.3)	13.1 17.8	30.1 35.6	1.3 1.6
7 (0-10 cm) 8 (10-22 cm)	coarse sand (0.63-2)	2.9 5.1	24.5 27.3	2.4 4.1
7 (0-10 cm) 8 (10-22 cm)	medium sand (0.2-0.63)	7.1 12.7	21.4 25.4	2.6 2.0
7 (0-10 cm) 8 (10-22 cm)	fine sand (0.063-0.2)	4.2 8.2	31.3 37.2	4.4 6.0
7 (0-10 cm) 8 (10-22 cm)	silt and clay (<0.063)	5.0 5.8	40.2 67.7	6.4 10.3

Table 2. Univariate modeling of radon exhalation by soil water content using Equation (3) with the parameters b and c for the years 2015, 2016 and the combined years 2015 and 2016.

Year	Parameter	Estimate	Uncertainty	95% confidence limits	
2015	b	30.2	0.3	29.5	30.8
	c	0.0839	0.0015	0.0810	0.0868
2016	b	24.9	0.4	24.1	25.8
	c	0.126	0.003	0.120	0.131
2015+2016	b	29.8	0.3	29.3	30.4
	c	0.0997	0.0010	0.0977	0.1018

Table 3. Multivariate modelling of radon exhalation with the best estimates of the model parameters.

Parameter	Estimate	Uncertainty	95% confidence limits	
Intercept	3.01	19.56	-35.33	41.34
Soil water content	0.179	0.032	0.117	0.242
Temperature soil (20 cm)	-0.617	0.045	-0.705	-0.529
Temperature air	0.520	0.035	0.451	0.589
Pressure air	-0.002	0.020	-0.041	0.038
Lag1	0.492	0.016	0.460	0.523
Lag3	0.138	0.017	0.106	0.170
Lag5	0.177	0.016	0.144	0.209

Table 4. Assessment of the forecasting performance analogous to the 'hold-one-out' method. Parameter estimates of the combined non-linear and auto-regressive linear regressions based on all data excluding any one chosen of all possible 12 calendar months and correlations between the observed and predicted radon exhalation values in the month(s) excluded.

Month(s) in 2015 and 2016 excluded for model parameter estimation	**Rayleigh parameters**		**Parameter estimates of the autoregressive linear regression**								**Correlation between the observed and predicted radon exhaltion values in the month(s) excluded**
	b	**c**	**Intercept**	**Soil water content**	**Temperature soil**	**Temperature air**	**Pressure air**	**Lag1**	**Lag3**	**Lag5**	
None/Reference	**29.833**	**9.975**	**3.005**	**0.179**	**-0.617**	**0.520**	**-0.002**	**0.492**	**0.138**	**0.177**	**0.78**
January	29.520	10.051	-14.226	0.185	-0.641	0.547	0.016	0.473	0.144	0.187	0.68
February	29.061	10.111	1.302	0.172	-0.611	0.539	0.000	0.487	0.138	0.179	0.82
March	28.846	10.016	35.671	0.182	-0.598	0.511	-0.035	0.494	0.138	0.171	0.80
April	29.275	9.996	3.938	0.177	-0.585	0.496	-0.003	0.494	0.140	0.181	0.81
May	29.267	9.960	12.374	0.182	-0.627	0.525	-0.011	0.495	0.122	0.188	0.66
June	29.356	9.818	4.348	0.179	-0.627	0.502	-0.003	0.485	0.147	0.178	0.64
July	30.057	9.895	4.570	0.180	-0.614	0.524	-0.003	0.491	0.140	0.174	0.71
August	30.080	9.939	1.284	0.180	-0.628	0.532	0.000	0.497	0.146	0.165	0.56
September	30.843	9.847	9.621	0.175	-0.574	0.491	-0.009	0.499	0.131	0.176	0.73
October	31.461	9.776	4.153	0.186	-0.608	0.504	-0.003	0.488	0.127	0.173	0.70
November	30.295	10.089	-6.336	0.211	-0.660	0.550	0.008	0.497	0.133	0.165	0.45
December	30.030	10.233	-30.933	0.167	-0.650	0.530	0.034	0.495	0.134	0.168	0.52