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Influences of meteorological parameters on indoor radon concentrations (^{222}Rn) excluding the effects of forced ventilation and radon exhalation from soil and building materials

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ABSTRACT

Elevated indoor radon concentrations (^{222}Rn) in dwellings pose generally a potential health risk to the inhabitants. During the last decades a considerable number of studies discussed both the different sources of indoor radon and the drivers for diurnal and multi day variations of its concentration. While the potential sources are undisputed, controversial opinions exist regarding their individual relevance and regarding the driving influences that control varying radon indoor concentrations. These drivers include (i) cyclic forced ventilation of dwellings, (ii) the temporal variance of the radon exhalation from soil and building materials due to e.g. a varying moisture content and (iii) diurnal and multi day temperature and pressure patterns. The presented study discusses the influences of last-mentioned temporal meteorological parameters by effectively excluding the influences of forced ventilation and undefined radon exhalation. The results reveal the continuous variation of the indoor/outdoor pressure gradient as key driver for a constant "breathing" of any interior space, which affects the indoor radon concentration with both diurnal and multi day patterns. The diurnally recurring variation of the pressure gradient is predominantly triggered by the day/night cycle of the indoor temperature that is associated with an expansion/contraction of the indoor air volume. Multi day patterns, on the other hand, are mainly due to periods of negative air pressure indoors that is triggered by periods of elevated wind speeds as a result of Bernoulli's principle.

1. Introduction

Elevated concentrations of the naturally occurring radioisotope ^{222}Rn (hereafter referred to as "radon") in residential indoor air have been increasingly recognized as potential health risk during the last decades (e.g. WHO, 2009). As a consequence a considerable number of large-scale indoor radon surveys have been conducted in several European countries. Major purpose of these surveys was to establish national reference levels and/or threshold values that can serve as basis for the setup of action plans that aim at limiting the indoor radon exposure to humans (e.g. European Council, 2014; Tollefsen et al., 2014; EEA, 2013).

The studies revealed that the indoor radon concentration in residential homes is governed by ventilation habits, meteorological parameters, individual building characteristics, and by the geological and physical conditions of the soil where radon is constantly being produced. Since the latter is (besides forced ventilation) a particularly influential factor, the necessity of the local enforcement of radon related legislation is mainly determined by the geological and geographical setting of the area in question. The related influential parameters of the soil are often summarized as its "radon potential" (e.g. Chen and Ford, 2017; Schmid and Wiegand, 1998). Its large-scale spatial mapping is a key tool for radon related risk assessments of residential areas and allows furthermore the optimization of small-scale radon surveys that aim at preventing or mitigating radon exposure to humans (Ciotoli et al., 2017).

However, radon accumulation in dwellings is not only controlled by radon exhalation from the subsoil or building materials and by forced ventilation. Strongly influential are also the local time-variant meteorological conditions (e.g. Cinelli et al., 2011; Yarmoshenko et al., 2016). Related time-variant influential parameters include soil moisture, soil and air temperature incl. the associated gradients, wind speed, and air pressure (e.g. Schubert and Schulz, 2002). Studies that focused on diurnal variations of indoor radon concentrations have shown that they are generally higher in the early morning hours and lower in the early afternoon (e.g. Murty et al., 2010; Karunakara et al., 2005). While some authors associate this temporal pattern primarily to the increased forced ventilation of the rooms during the daytime (e.g.

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Vaupotic et al., 2012), others suggest the generally observed diurnal variations of wind speed, air pressure and/or temperature as interrelated key drivers (Gogolak and Beck, 1980; Porstendörfer et al., 1991; Porstendörfer et al., 1994; Schubert and Schulz, 2002).

Most recent studies that focus on diurnal changes in indoor radon concentration discuss the combined impact of (i) individual ventilation habits, (ii) building characteristics, (iii) radon exhalation from soil and building material and (iv) meteorological influences. However, approaches that allow disentangling the individual contributions of these impacts are scarce. In the presented study we used an experimental setup that allows focusing exclusively on the meteorological influences by effectively eliminating the influence of both forced ventilation and undefined radon exhalation.

Continuous time series of radon concentration, temperature, air pressure and wind speed were recorded over a two month period inside and outside a closed (but not completely air-tight) container that was exposed to meteorological influences. The container was equipped with a defined radon point source. No other potential radon source (subsoil, building material) was present within the container. Hence, the approach resulted in indoor radon time series that were unaffected by both soil or building characteristics and ventilation habits. The results allowed thus the individual evaluation of the impact of varying indoor/outdoor gradients of meteorological parameters.

2. Materials and methods

2.1. Determination of radon exhalation rate

A piece of high grade uranium ore (pitchblende) from the vein-type deposit Niederschlema-Alberoda, Germany, was used as radon point source. For determination of its radon exhalation rate measurements were carried out under laboratory conditions applying a mobile radon-in-air monitor (AlphaGuard, Saphymo). For the measurements the source was placed in a radon-tight stainless steel box (volume 0.2 m^3) that was equipped with an inlet and an outlet port. After placing the source in the box the box was closed and sealed. Subsequently the enclosed air volume was pumped in a closed loop through box and radon monitor by means of a radon-tight gas pump (AlphaPump, Saphymo). Both detector and pump were placed on the lid of the steel box, which allowed keeping the (also radon-tight) connecting tubing (Tygon, Saint-Gobain) as short as possible (in total ca. 50 cm). The radon inventory of the air volume (I_{Rn}; [Bq]) was recorded continuously as time series in 10 min counting intervals. The total volume of circulating air was 0.2005 m^3. The pump rate was kept constant at 1000 cm^3/min. The experiment was carried out twice in order to improve the statistical reliability of the result. Each measurement started with a radon background concentration of 25 Bq/m^3, which equals a radon background inventory of the closed system of $I_{Rn} = 5$ Bq.

Continuous radon exhalation from the radon point source resulted in a gradual increase of I_{Rn} within the closed system. The slope of that increase started with a virtually linear rise that flattened out with time and gradually approached a steady state plateau. The calculation of the radon exhalation rate of the radon point source was made in two independent ways, (i) based on the virtually linear slope of the I_{Rn} increase that was recorded during the first 3 h and (ii) based on the final steady state inventory of the circulating air volume.

2.2. Container measurements

For investigation of the influences of varying meteorological parameters on the indoor radon concentration excluding the effects of forced ventilation and radon exhalation from soil and building materials a 33.2 m^3 (5.90 × 2.36 × 2.38 m) steel container was placed outdoors where it was exposed to wind, temperature and air pressure. Within the container the radon point source was placed in front of a desk ventilator, which kept the container indoor air continuously in slight motion. Radon concentrations, air temperatures and air pressures were measured both inside and outside the container as continuous time series over a period of two months by means of two mobile AlphaGuard radon monitors. Both monitors were run simultaneously set to a 10 min counting cycle. Additionally the pressure gradient between inside and outside of the container was recorded by means of a low-level pressure difference monitor (Multi Sensor Unit, Genitron). Furthermore the outside wind speed was recorded using a mobile weather station (Kestrel® 4500). Missing wind speed values that resulted from short-term power outages of the weather station were filled with equivalent data from an external nearby station. A bias-correction was applied to these external data using a linear relation between external smoothed measurements as predictor and local smoothed measurements as response ($R^2 = 0.48$).

For radon measurements both indoor and outside air were pumped into the respective AlphaGuard detection chambers at a rate of 0.3 L/min. The outside radon monitor was placed on the roof of the container sheltered by a tarpaulin. The indoor radon monitor was placed in the center of the container. The doors of the container were kept shut all the time. Still, a small ventilation hole in the container wall (2 above the container floor) was kept open to allow limited air exchange with the outside. The hole was ca. 3 cm in diameter and covered by a screen plate that sheltered it from the direct impact of wind gusts. The container was completely purged with outside air before the experiment was started. For the evaluation of the final indoor and outdoor Rn time-series, smoothing by running mean and correlation analysis (Pearson's correlation) was used.

3. Results and discussion

3.1. Determination of radon exhalation rate

Two completely independent datasets were used for calibration of the radon point source (each with n = 2): the initial I_{Rn} slope that developed over the first 3 h of the measurement and the final I_{Rn} equilibrium that was reached after about three weeks and was confirmed

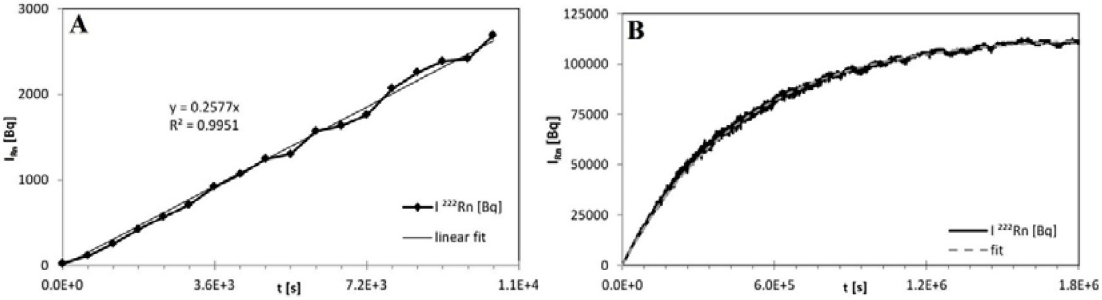

Fig. 1. Source calibration datasets "radon inventory vs. time" revealing an exhalation rate of $F_{slope} = 0.26$ Bq/s based on the initial slope (1A) and an exhalation rate of $F_{eq} = 0.24$ Bq/s based on the steady state inventory ($I_{Rn} = 114$ kBq; 1B).