# FIRST STEPS IN THE DEVELOPMENT OF A POSSIBLE MEASUREMENT METHOD TO ESTIMATE THE RADON CONCENTRATION AS AN INDICATOR OF THE INDOOR AIR QUALITY

by

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The energy conservation regulation provides upper limits for the annual primary energy requirements for new buildings and old building renovation. The actions required could accompany a reduction of the air exchange rate and cause a degradation of the indoor air quality. In addition to climate and building specific aspects, the air exchange rate is essentially affected by the residents. Present methods for the estimation of the indoor air quality can only be effected under test conditions, whereby the influence of the residents cannot be considered and so an estimation under daily routine cannot be ensured. In the context of this contribution first steps of a method are presented, that allows an estimation of the progression of the air exchange rate under favourable conditions by using radon as an indicator. Therefore mathematical connections are established that could be affirmed practically in an experimental set-up. So this method could provide a tool that allows the estimation of the progression of the air exchange rate and in a later step the estimation of a correlating progression of air pollutant concentrations without limitations of using the dwelling.

Key words: radon, air exchange rate, air quality, air pollutant

#### INTRODUCTION

In Central Europe adults remain between 80% and 90% of the day in closed interior [1]. This leads to an exposure to a multitude of air pollutants, which can accumulate in the indoor air. The presence of humans can lead to emissions of CO<sub>2</sub> or exhalations. Due to building products, furniture or further objects of daily handling substances like volatile organic compounds (VOC) are emitted [2]. High indoor air humidity can lead to mould formation, which emits spores and allergens [3]. In this context radon is kept in mind since it can accumulate in the indoor air too and can mean a health risk.

The concentrations of these air pollutants depend on one hand on the particular polluter and on the other hand on the ventilation of the room [4]. Good ventilation and so a high air exchange rate, arrange for the avoidance of the accumulation of air pollutants in the indoor air and for the continuance of the concentra-

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tion on a low level. Yet the energy conservation regulation provides upper limits for the annual primary energy requirements for new established buildings and old building renovations [5]. For the reduction of the energy requirements it is necessary to insulate the casing of the building and to install sealed doors and windows. This can lead to a reduction of the air exchange rate that can result in an increase of the air pollutant level [6, 7]. To avoid this, there are certain requirements for the ventilation of a building which can be achieved by an active ventilation system, or adapted ventilation behaviour (passive ventilation). Normally active ventilation systems are qualified for the avoidance of an increase of the air pollutant level. Yet, for using the passive ventilation there are high addictions to the behaviour of the residents. Deficient passive ventilation can lead to an increase of the air pollutant level.

Thus the knowledge of the air exchange rate is fundamental for the estimation of the indoor air quality. Present methods for the estimation of the air exchange rate can only be effected under test conditions or under the guarantee of a constant air exchange rate

[8, 9]. A consideration of the room use as an important influence on the air exchange rate cannot be effected. Alternatively to the present methods, the first steps of an approach that is using radon as an indicator shall be presented. This could allow the evaluation of the progression of the air exchange rate under daily routine.

The evaluation of progressions of the air exchange rate by measured progressions of the radon concentration is achieved by several steps. At first, an evaluation of the radon source at a preferably low and constant air exchange rate is accomplished. The desired signal gained by a long term measurement is filtered by suitable filter functions. Finally, via a reconstruction method developed for this purpose, the progression of the air exchange rate is evaluated by the filtered progression of the measured signal of the radon concentration. The target of the presented method is guarantee of a preferably evaluation of the radon source and a preferably low-noise reconstruction of the air exchange rate by a noisy, long term measurement signal of the radon concentration.

For a first practical verification of the method, measurements are conducted by using a measurement chamber with a dimension of a real dwelling. The measurement chamber is featured with a programmable ventilation control system, which allows the implementation of selectable progressions of the air exchange rate. Measurements in real dwellings shall be effected in future developments.

#### **MODELLING**

To build a model, which describes the mathematical connection between the air exchange rate k(t) and the radon concentration c(t), a differential equation is determined. In this juncture the radon which degasses out of the ground soil and the building material is considered in terms of the radon source  $Q_{v}$ , normalised on the volume of the dwelling V. For simplification, the radon source and the radon mixing in the air are assumed as constant at first. The radioactive decay arranges for reduction of the radon concentration and is considered in terms of the differential form of the decay law with the decay constant  $\lambda$ . To establish a connection between the air exchange rate and the radon concentration, the air exchange is listed with the delivery air and the discharged air, at which via the delivery air, outdoor air with the radon concentration  $c_a(t)$  is carried into the room and via the discharged air, indoor air with the radon concentration c(t) is carried out of the room

$$\frac{\mathrm{d}c(t)}{\mathrm{d}t} \quad Q_{\mathrm{V}} \quad \lambda c(t) \quad k(t)c_{\mathrm{a}}(t) \quad k(t)c(t) \quad (1)$$

Via the solution of the differential equation (eq. 1) a time discrete equation can be constructed, which allows the iterative evaluation of the radon concentra-

tion c(t) by the air exchange rate k(t). In this juncture  $\Delta t$  describes the time interval of the time discrete data

$$c(n) \quad e^{\left[\lambda - k(n)\right]\Delta t}$$

$$c(n-1) \quad \frac{Q_{V} - c_{a}(n)k(n)}{\lambda - k(n)} \quad \frac{Q_{V} - c_{a}(n)k(n)}{\lambda - k(n)} \quad (2)$$

Because eq. 2 cannot be solved to the air exchange rate, it is necessary to transform the differential equation (eq. 1) into a difference equation. In turn this can be dissolved to the time discrete air exchange rate k(n)

$$k(n) = \frac{\frac{c(n-1) - c(n)}{\Delta t} - Q_{V} - \lambda c(n)}{c_{a}(n) - c(n)}$$
(3)

For c(n) measured data of the radon concentration can be used which are recorded with the measurement interval  $\Delta t$ . Before starting the long term measurement the radon source  $Q_{\rm V}$  is evaluated via a method developed for this purpose. According to the constancy of the radon source the evaluation can be repeated several times. The outdoor radon concentration can either be supposed constant  $c_{\rm a}$ , or can be considered in the equation via an own series of measurements  $c_{\rm a}(n)$ . When measuring a low level indoor radon concentration it will make sense to record the progression of the outdoor concentration separately.

Thus eqs. 2 and 3 deliver the tool to evaluate the progression of the air exchange rate by the radon concentration and the progression of the radon concentration by the air exchange rate.

#### **EVALUATION OF THE RADON SOURCE**

All parameters of eq. 3 are known, i. e., metrologically accessible except the radon source  $Q_{V}$ . The radon source normalised to the volume of the dwelling  $Q_V$  represents the full characterisation of the dwelling. It can be evaluated via recording a saturation curve of the radon concentration by increasing the air exchange rate at first (passive or active ventilation) to lower the radon concentration in the dwelling to a level which is nearly in accordance with the outdoor radon concentration. Afterwards the air exchange rate is lowered (ending the "ventilation situation"). This leads to a new increase of the radon concentration in the dwelling and thus to a saturation curve, which follows with the guarantee of a preferably constant air exchange rate an exponential progression. This saturation curve is recorded with a radon monitor. Via using the Levenberg-Marquardt algorithm the time constant  $\tau$ , the radon concentration at the beginning  $c_0$  and the radon saturation concentration  $c(\infty)$  are fitted to the following equation

$$c(t) \quad e^{-\frac{t}{\tau}} [c_0 \quad c(\infty)] \quad c(\infty) \tag{4}$$

The determined time constant  $\tau$  and the radon saturation concentration  $c(\infty)$  are used for the evaluation of the radon source. Following connection can be derived from the solution of the differential equation (eq. 1)

 $Q_{\rm V} = \frac{c(\infty)}{\tau} c_{\rm a} \frac{1}{\tau} \lambda$  (5)

Using the parameters  $\tau$  and  $c(\infty)$  determined in eq. 4 and the constant parameters  $\lambda$  and  $c_{\rm a}$  the radon source normalised on the volume of the dwelling  $Q_{\rm V}$  can be evaluated via eq. 5, which represents the full characterisation of the dwelling.

### FILTERING SERIES OF MEASUREMENTS OF THE RADON CONCENTRATION

The reconstruction of progression of the air exchange rate is effected by eq. 3. In this connection, there are high requirements on the measured radon concentration signal because of the noise sensitivity accounted by the term c(n+1)-c(n). Because of the statistical character of the measurement of radon concentration, the measured signal is interfered with noise and so it is necessary to smooth the measured signal. This is effected by a theoretical system approach of filtering using window functions (rectangle, Hamming, Hanning, etc...) [10] in four steps. In the first step called "zero-padding" the measured signal is expanded with zeros to achieve the same length of the measured signal and the window function. In the second step with zeros expanded signal is transformed into the frequency domain by using the Fourier transformation. In the third step the frequency spectrum is multiplied by an adequate window function. Finally the resulting filtered spectrum is transformed into the time domain by using the inverse Fourier transformation. In order to realise a preferably efficient filtering without loss of attributes of the desired signal, an adequate choice of the filter function and its width is of essential importance. This choice depends on the frequency spectrum of the measured signal. After all, the progression of the air exchange rate can be evaluated via the reconstruction of the filtered measured data of the radon concentration.

#### **EXPERIMENTAL SET-UP**

The goal for the first practical verification of the constructed measurement model in a measurement chamber was to eliminate disturbance values, create controlled conditions and hence to allow the receipt of repeatable results. The measurement chamber is built up of a cooling chamber (producer: Viessmann) that provides the vantages of thermal insulation and high

tightness. It has a volume of 10 m³ and therefore it is big enough to emulate a real dwelling and to accommodate experimental constructions. It is composed of several segments which have a thermal insulation of 60 mm PUR foam and a stainless steel surface. The thermal insulation offers the advantage of insensibility of the indoor temperature from the outdoor temperature. To achieve a high tightness of the chamber wall, the gaps are sealed with aluminum covered adhesive foil and sealing mass.

To control the ventilation and so to regulate the air exchange rate a ventilator is applied, whose speed can be regulated by a microcontroller. The microcontroller is connected with a PC, which allows the import of any progressions of the air exchange rate. The ventilator is installed at a measurement distance that is placed in the delivery air channel. The air flow rate at this measurement distance is measured by a thermo-anemometer (Ahlborn FVA 935 TH4) with a measurement range of 0.08-2 m/s. With an effective cross-sectional area of 15.1 cm² in the measurement distance pipe, the effectively air exchange rate affecting the measurement chamber can be determined with a range of 0.05-1 per hour. Hence, the measurements can be effected with air exchange rates like they appear upon real dwellings.

Inside and outside the measurement chamber the radon concentration, the air temperature, the air humidity and the air pressure are measured. For this purpose radon-monitors of the type AlphaGUARD (producer: Saphymo) are used.

All measurements are effected by recording time lines with sample times of 10 minutes. An exterior view of the measurement chamber is shown in fig. 1.

#### RESULTS

Measurements on the measurement chamber were performed at which uranium ore was used as the radon source. In the following two measurement examples are presented. The first example was recorded using uranium ore with a high source power  $Q_{\rm V}=670~{\rm Bq/hm^3}$ . As a test signal of the air exchange rate a rectangle progression was adjusted by the ventilation control system. The resulting pro-



Figure 1. Exterior view of the measurement chamber

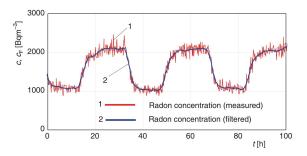


Figure 2. Example 1: measured radon concentration (1) in the measurement chamber and the progression filtered with a rectangle window with a width of 512 samples (2)

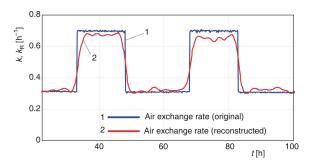


Figure 3. Example 1: comparison of the air exchange rate reconstructed by the progression of the radon concentration (2) and the original air exchange rate (1)

gression of the radon concentration inside the measurement chamber is shown in fig. 2. To filter the measured progression of the radon concentration a Hamming window with a width of 512 samples is used, which eliminates the high frequencies of the spectrum and represses the noise component.

To reconstruct the air exchange rate by the filtered progression of the measured radon concentration, the progression of the air exchange rate is determined iteratively via the implementation of the reconstruction algorithm (eq. 3). The reconstructed progression of the air exchange rate is shown in fig. 3.

Figure 3 shows that the reconstructed progression of the air exchange rate is in good accordance with the original progression. But it can be seen that the reconstructed progression is superposed by frequency parts resulting from not eliminated noise components of the measured signal of the radon concentration. Yet a more intense smoothing of the measurement signal will lead to an elimination of the desired signal too. Thus for every recorded measured progression, the elimination of the desired signal must be compromised with the non-elimination of the noise component.

The first example was recorded with a higher source power than it occurs in real dwellings. To achieve more daily routine like measurement conditions the second example was recorded with a lower radon source of  $Q_V = 11$  Bq/hm<sup>3</sup>. A rectangle progression of the air exchange rate was adjusted by the ventilation control system. The resulting progression of the radon concentration inside the measurement chamber is shown in fig. 4. The measured progression of the radon concentration was filtered with a hamming window with a width of 256 samples.

The reconstruction of the air exchange rate is effected like in example 1 described. The comparison of the original and reconstructed air progression of the air exchange rate is shown in fig. 5.

It can be seen that the reconstruction in example 2 is effected less accurate than the reconstruction in example 1. The reason for this is the lower level of the radon concentration which leads to a higher noise in the measured signal and consequently to a less accurate reconstruction of the air exchange rate. The consequences of these circumstances are a lower time resolution of the reconstructed air exchange rate and limitations of the reconstruction of high air exchange rates at low radon sources.

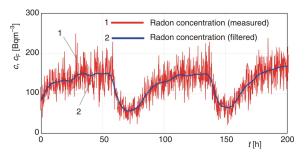


Figure 4. Example 2: measured radon concentration (1) in the measurement chamber and the progression filtered with a hamming window with a width of 256 samples (2)

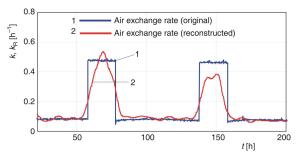


Figure 5. Example 2: comparison of the air exchange rate reconstructed by the progression of the radon concentration (2) and the original air exchange rate (1)

#### DISCUSSIONS

With the aid of the presented method progressions of the radon concentrations and the air exchange rate can be converted into each other under favourable conditions. The principal point of the method is represented by the evaluation of the progression of the air

exchange rate, by a measured progression of the radon concentration. Compared to present methods which allow no time resolution of the air exchange rate, or have to be effected under test conditions, the presented method provides a time resolution of the air exchange rate. The use of the dwelling which has a big influence on the air exchange rate can be considered. Only the evaluation of the radon source has to be effected under test conditions.

For the room that has to be analyzed certain requirements exist, which describe potential limitations for the method. For the evaluation of the radon source, the radon saturation concentration must be high enough for achieving a solvable saturation curve. If there is no solvable saturation curve, an evaluation of the radon source is impossible. There are similar limitations for long term measurements. With availability of only a low radon source and because of the high statistical noise there are difficulties to filter the measurement signal, on one hand and to reconstruct the air exchange rate, on the other hand. The presented measurement example 2 shows first impressions of these difficulties, at low radon sources, in the form of a lower time resolution, especially at high air exchange rates.

The filtering of the progression of the radon concentration leads to a reduction of the noise component, on one hand and to a reduction of several frequency parts of the desired signal, on the other hand. This implicates the circumstance that the original progression of the air exchange rate can exactly be reconstructed in the rarest cases. Thus it is a question of an approach on the original progression of the air exchange rate. The accuracy depends on the discovered level of the radon concentration and the choice of the fitting parameters.

A critical point of the reconstruction of the air exchange rate is the constancy of the radon source. It is known that the radon source can vary under some conditions, for example at air pressure variations [11-13]. Especially in areas with mining industry, variations of the radon source can be measured [14]. But, until now, there is not enough information about the dimension of this variability. Hence the dimension of the variability of the radon source must be checked and the decision must be made about which dimension of the variability can be accepted. If there is a slow variability over several days, for example, the evaluation of the radon source can be repeated every few days or, if there is a small variability, the reconstruction can be effected with a mean radon source. The smaller the variability of the radon source, the better the reconstruction of the air exchange rate works.

#### **OUTLOOK**

The presented method pictures an early development stage that serves for future developments as a basis. Further steps provide the evaluation of the limitations of the method, the analysis of the constancy of the radon

source, the construction of a "multi-room-model" and the referencing of the indoor air quality.

The reconstruction of the air exchange rate at low level radon concentrations (especially less than 100 Bq/m<sup>3</sup>) is subjected to certain difficulties. A low radon concentration can lead to the circumstance that the radon source cannot be evaluated, on one hand and because of the statistical noise of the radon measurement, the air exchange rate cannot be reconstructed, on the other hand. Hence with a decrease of the radon source the reconstruction of high air exchange rates is more difficult. Which values of the air exchange rate can be reconstructed at which values of the radon source, shall be reviewed in future analyses. These analyses shall be effected under several requirements on the accuracy and the time resolution of the reconstruction. At this juncture, the statistical characteristics of the used radon-monitors at low level radon concentrations, shall be consulted.

The inspection of the constancy of radon source represents an important duty. A first approach can be achieved by evaluating the radon source in real dwellings, repeated every few days. All doors inside the dwelling should be open to apply the presented "single-room-model". A variation of the radon source evaluated every few days could deliver information about the long term variability. The short time variability can be analysed by performing reference measurements of the air exchange rate with the tracer gas SF<sub>6</sub>. A comparison of the air exchange rate reconstructed by the measured radon concentration and the air exchange rate reconstructed by the tracer gas SF<sub>6</sub>, allows drawing conclusions from the variability of the radon source. The inspection of the constancy of the radon source has to be effected in real buildings or dwellings to achieve real conditions relating to pressure or other climatic variations. Hereupon a decision should be made which dimension of variations can be accepted and which dimensions of variations are representing limitations for the method.

To transfer the method to real buildings and dwellings a "multi-room-model" shall be build based on the presented "single-room-model". Therefore a system of differential equations should be induced containing one equation for every room of the building or the dwelling that needs to be analyzed. The dissolution of this system could allow the evaluation of individual air exchange rates under favourable conditions.

To establish a relationship to the indoor air quality, mathematical connections between the air exchange rate and several air pollutants shall be established. At this juncture it can be about any air pollutants like volatile organic compounds (VOC), formaldehyde or CO<sub>2</sub>. To build a mathematical model, the model presented in this paper shall be used and adapted according to the air pollutant. If there are adequate models available to estimate several air pollutant concentrations following approach will be applied. At

first the air exchange rate will be estimated by the radon concentration via the method presented in this paper. In the next step air pollutant concentrations will be estimated via the respective mathematical models. Finally a representative choice of the considered air pollutants allows the evaluation of the indoor air quality.

Because of the interaction between several air pollutants and other substances or climatic parameters, the construction of a mathematical model for estimation of an air pollutant concentration by the air exchange rate, requires a high complexity. Thus it will be effected only for a choice of important air pollutants and therefore primarily serve to deliver tools which can be applied to arbitrary air pollutant combinations [15].

#### **AUTHOR CONTRIBUTIONS**

The theory and the method was developed by F. A. Rossler, T. A. Jai, V. Grimm, H. Hingman, and J. Breckow. The experiments were made by F. A. Rossler, T. Orovwighose, and N. Jach. All authors analysed and discussed the results. The manuscript was written and the figures were prepared by F. A. Rossler.

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## ПРВИ КОРАЦИ У РАЗВИЈАЊУ МЕРНЕ МЕТОДЕ ЗА ПРОЦЕНУ КОНЦЕНТРАЦИЈЕ РАДОНА КАО ИНДИКАТОРА КВАЛИТЕТА ВАЗДУХА У ЗАТВОРЕНОМ ПРОСТОРУ

Прописи о очувању енергије дају горње границе за годишње потребе примарне енергије нових зграда и реновираних старих зграда. Потребни захтеви могу довести до смањења брзине измене ваздуха и тиме деградирати квалитет ваздуха у затвореном простору. Поред климатских услова и специфичних аспеката грађевинског објекта, на измену ваздуха суштински утичу и становници зграде. Актуелне методе за процену квалитета ваздуха у затвореним просторима могу се спровести само при условима тестирања, док се утицај становништва не може узети у разматрање и тиме се не може осигурати процена у свакодневном животу. У циљу доприноса овом питању, представљени су први кораци методе која омогућава процену прогресије брзине измене ваздуха под најпогоднијим условима, користећи радон као индикатор. Стога су утврђене математичке релације које се могу потврдити практично кроз експеримент. Отуда ова метода може да буде средство за процену прогресије брзине измене ваздуха и у каснијем кораку процену корелације прогресије концентрације загађивача ваздуха без ограничавања употребе зграде.

Кључне речи: радон, брзина измене ваздуха, квалишеш ваздуха, загађење ваздуха