RADON DIAGNOSTICS AND TRACER GAS MEASUREMENTS

K. Jílek[1], M. Brabec[2]

1 National Radiation Protection Institute, Department of Radon program, Prague, Czech Republic

2 National Institute of Public Health, Department of Biostatistics, Prague, Czech Republic

Abstract

The notion of radon diagnostics among others comprises several activities focused on the detection of radon sources and quantification of their total entry rates into the house. Most often, the diagnostics is used in the house before the intended radon remedial action and afterwards to estimate its effectiveness. Radon measurements alone do not allow for a reliable estimation of radon entry rate, therefore radon measurements have to be augmented by a tracer gas measurements. For more than four years we have been using a tracer gas technique with good practical results applying carbon monoxide as a tracer gas. In this paper, we will explain the basics and theoretical background of the tracer gas technique, which is used for continuous measurements of air ventilation rate (generally time-varying) and for simultaneous estimation of air ventilation rate and radon entry rate. We will discuss some of its limitations. After introducing the tracer gas technique formally, we will demonstrate its performance for calculation of air ventilation rate on real data from routine measurements. We will also discuss current possibilities for air ventilation rate estimation based on radon measurements only. Further, we will also describe a practical application of the tracer gas technique for simultaneous estimation of air ventilation rate and radon entry rate in a real house where the effectiveness of radon remedy was tested. For all estimations of air ventilation rate and radon entry rate, we will assume calculations in a single zone implicitly.

Key Words: Tracer gas, CO gas, Radon diagnostics, Ventilation rate,

Introduction

Tracer gases are used for a wide range of diagnostic techniques including leak detection [1], atmospheric tracing [2] and in last decade for measurement of air flow in buildings [3]. The main general advantage of tracer gas techniques compared to non- tracer measurement techniques of ventilation rate (i.e.modeling) consists in the possibility of direct measurement in buildings under actual living conditions.

Currently a several dozen of radon diagnostics per year are carried out during remedial actions in houses in the Czech Republic. For correct estimation of their effectiveness separate measurements of radon entry rate and ventilation rate is needed. Since radon measurements alone do not allow for a reliable estimation of these two parameters radon measurement should be augmented by a tracer gas measurement.

The aim of this paper is demonstrate a practical applicability of CO tracer gas technique in the framework of radon diagnostics based on our four years experience with continuous monitor of CO gas.

2. Theoretical background

2.1. Determination of ventilation rate

The fundamental tracer mass balance equation for all tracer gas measurements is:

$$\frac{dc(t)}{dt} = \frac{F(t)}{V} + \lambda(t) \; (c_e(t) - c(t)) \qquad (1)$$

where V is the effective volume of enclosure (in m^3), F(t) represents the entry rate of tracer gas through the enclosure (in kg/s), $c_e(t)$ and c(t) are external respectively internal concentration of tracer gas at time t (in kg/m^3) and $\lambda(t)$ is the effective ventilation rate through the enclosure at time t (in 1/s).
Equation (1) is also valid for radon gas and then $\lambda(t)$ additionally involves decay constant of radon.
Generally, we can assume neglecting the tracer gas concentration in the external air. Then equation (1) can be rewritten for calculation of ventilation rate as follows:

$$\lambda(t) = \frac{F/V - dc(t)/dt}{c(t)} \qquad (2)$$

There are tree basic methods [4] that use equation (2) to determine ventilation rate through the enclosure. In all our experiments we use two of them i.e. the concentration decay method (CDM) which assumes F(t) is zero and the constant tracer injection method (CTIM), which assumes F(t) is well defined and constant during tracer injection, thus F(t) = F.
Further, in principle there are three different approaches [5] for determination ventilation rate from measured tracer gas concentrations. We adopted two of them i.e. regression techniques (RT) and averaging techniques (AT). Regression techniques assume constant ventilation rate over the regression period and the parameters λ and F/V are mostly treated as unknown in the regression.
Averaging techniques are those which yield unbiased estimates of the average ventilation rate over the measuring period and enable continuous determination of ventilation rate. General expressions used in routine in our calculations of ventilation rate based on combination above mentioned techniques and methods are summarized in Table 1.

***2.2.** Simultaneous determination of radon entry rate and ventilation rate based on simultaneous measurements of radon concentration and CO concentration*

The general tracer mass balance equation (1) for measured concentrations both radon gas and CO gas in the measured enclosure at time t can be rewritten as follows:

$$\frac{da(t)}{dt} = \frac{G(t)}{V} + \lambda r(t) \; (a_e(t) - a(t)) \qquad (3)$$

$$\frac{dc(t)}{dt} = \frac{Q(t)}{V} + \lambda(t) \; (c_e(t) - c(t)) \qquad (4)$$

where a(t), c(t) are measured concentrations of radon gas and CO gas, respectively, G(t), Q(t) are radon gas and CO gas, respectively entry rates into the enclosure with volume V, $a_e(t)$,$c_e(t)$ are external concentrations of radon gas and CO gas, respectively, λr (t) is effective ventilation rate through the enclosure for radon tracer gas and $\lambda(t)$ is effective ventilation rate through the enclosure for CO gas.
Now we assume:

- neglecting external concentrations for both radon gas and CO gas
- radon decay constant (0.00755 1/h) is neglected in comparison to actual value of ventilation rate
- $\lambda r\,(t) = \lambda(t)$
- CO gas entry rate to the enclosure is known and constant over all measuring period of time .

Then equation (3) can be rewritten with the use of equation (4) in terms of radon entry rate as follows:

$$\frac{G(t)}{V} = \frac{da(t)}{dt} + \lambda(t)\; a(t) \qquad (5)$$

where $$\lambda(t) = \left(\frac{Q/V - dc(t)/\,dt}{c(t)} \right) \qquad (6)$$

There are three following basic approaches how to estimate radon entry rate according to general equation (5).

A) *Extreme points technique*
On condition that both time variation of radon concentration and CO concentration reach on local or absolute minimum or maximum at the same time or both concentrations are in a steady state at the same time t, equation (5) can be to rewrite as follows:

$$\frac{G(t)}{V} = \left(\frac{Q(t)/V}{c(t)} \right)\; a(t)\;) \qquad (7)$$

We can calculate radon entry rate at time t from the known value of CO entry rate and from measured concentrations of the both tracers at time t.

B) *Averaging technique*
We consider a small time interval Δ measured through the continuous monitor of radon (for used radon monitor Radim3 this time interval Δ represents minimum of its sampling time and equals 30 min). Over this period Δ we can expect that measured radon concentration will be is in a steady state and in equation (5) the term $\frac{da(t)}{dt} \cong 0$ [6]. Then equation (5) can be rewritten in terms of average values:

$$< \frac{G(t)}{V} > \; = \; <\lambda(t)> \;\; <a(t)> \qquad (8)$$

where $<\lambda(t)>$ is average value of ventilation rate over the measured period of time Δ at time t calculated by the averaging method from measured CO concentrations (see Tab.1 in the following text).
$<a(t)>$ is the average value of radon concentration measured over the same period of time Δ at the same time t.

C) *Regression technique*

Regression technique assumes that ventilation rate is known from CO data and constant over the regression period. The regression try to find the best set of parameters that fit time variation of radon concentration for the known special cases as general equation (5) has analytically known form of solution over the regression period (see expressions No.1, No,4, No.5, No.6 in Tab.1). The radon entry rate is always treated as unknown in the regression.

Table 1. *The general review of used methods in the framework of radon diagnostic. C(t) is generally the concentration of radon gas or CO gas (accordingly) measured at time t. By analogy, C(0) is the initial concentration of radon gas or CO gas in the beginning of the regression, F is radon entry rate or CO entry rate generally variable in time t , Ci(t), Cf(t) are initial and final, respectively measured concentrations of CO gas over the sampling period Δ at time t, λ is ventilation rate calculated for radon gas or CO gas at time t, generally variable in time t, both (CDM) and (CTIM) methods have been already mentioned previously, expressions in brackets < > mean their average value over a sampling period Δ .*

No.	*Method*	*Approach*	*Expression*	*Regress. Parameter*	*Measured Quantity*	*Comments*
1.	CDM	RT	$C(t) = C(0)\, e^{-\lambda t}$	Λ	C(t), C(0)	Radon gas, CO gas
2.	CDM	AT	$<\lambda> = \frac{1}{\Delta} \ln \left\| \frac{Cf(t)}{Ci(t)} \right\|$		Cf(t),Ci(t)	CO gas
3.	CTIM	AT	$<\lambda> = < \frac{F}{V.C(t)} > - \frac{1}{\Delta} \ln \left\| \frac{Cf(t)}{Ci(t)} \right\|$		C(t), Cf, Ci	CO gas
4.	CTIM	RT	$C(t) = C(0)\, e^{-\lambda t} + (\frac{F}{\lambda V})\ (1 - e^{-\lambda t})$	Λ, F	C(t), C(0)	Radon gas, CO gas
5.	CTIM	RT	$C(t) = (\frac{F}{\lambda V})\ (1 - e^{-\lambda t})$	Λ, F	C(t)	Radon gas, CO gas
6.	CTIM	RT	$C(t) = (\frac{F}{\lambda V})$	Λ	C(t)	Radon gas, CO gas

3. Materials and methods

3.1. Measurement system

<u>Carbon monoxide</u>
The CO concentration was measured with a portable continuous monitor especially designed for in - situ measurements. The monitor alone is consisted of data logger with connected sensors. The operation memory size of this data logger and utilized 32 bit command SW enable to save on hard disc output data from eight independent sensors at the same time. During all experiments performed in last four years we utilized two electrochemical sensors type Vario-Sense made by

DRAEGER (Germany). The sensors have the reacting time of 50s and adjustable useful dynamic range 0 - 300 ppm or 0-1000 ppm. The reacting time is a period when sensor reaches 90 % of actual value of CO concentration in ambient air. The producer declares for these sensors uncertainty type B = 3 ppm in range 0 – 300 ppm and uncertainty type B = 5 ppm in range 0-1000 ppm .
All indoor measurements with CO gas were naturally performed without presence of people. Used continuous monitor of CO provided average values of measured concentrations of CO over 5 min. sampling interval. Defined stable in time and pre-programmable values of CO gas entry rate into the measured enclosure was realized via programmable digital mass flow rate controller DFC made by Aalborg (U.S.A.). The DFC controller alone was absolutely calibrated against the portable bubble flow rate meter calibrator AMETEK (U.S.A.). The calibrator alone was certified to be accurate within a maximum error of ± 1 % of reading at NIST. The overall uncertainty of CO entry rate was estimated to be less than 3 %. As a source of CO gas was used a portable compressed CO gas tank with known volumetric concentration of CO gas in nitrogen certified by producer.

Radon gas
Radon concentration was measured by both continuous radon monitor Alphaguard made by Genitron Inc. (Germany) and continuous radon monitor Radim 3 made by SMM company (CZ). Monitor Alphaguard measured radon concentration with adjusted 60 min. sampling interval and monitors Radim 3 with adjusted 30 min. sampling interval. To produce stable and defined radon entry rate during our experiments we utilized an artificial dry flow pass through source of radium and efficient and stable pump. The source production of radon gas was certified by the producer EUROTRADE (CZ) and did 3.6 Bq/s with uncertainty type B less than 1.5 %. The output airflow rate from the source was absolutely calibrated against the above mentioned air flow rate meter calibrator AMETEK before each experiment. The overall uncertainty of artificial radon entry rate was estimated to be less than 5 %.

3.2. Experiments

Before utilizing CO as the tracer gas in radon diagnostics it was necessary to verify following two basic aspects:

- validation of the main assumption in equation (5) i.e. verifying if both radon gas method and CO gas method provide the same results of determination of ventilation rate,
- estimation of the inaccuracy in determination of radon entry rate according to equations (7) and (8).

Ventilation rate
To accomplish mentioned validation we have performed several sets of experiments with the aim to compare results of determination of ventilation rate for the following combinations of the basic tracer injection methods:

- the decay method for CO gas versus the decay method and the constant injection method for radon gas,
- the constant injection method for CO gas versus the constant injection method for radon gas

The ventilation rate was calculated according to above mentioned methods No.1-7 (see Tab.1 in par.2) in relevant time intervals. For all the calculations we used the whole width of measured time variations both radon concentration and CO concentration. All experiments were performed with the above mentioned measurement devices in our experimental Lab. They were performed in framework of Institutional Research of NRPI [7]. The Lab was a single room with one window and one door which could be packed very well against the tracer leakage. Its effective volume was estimated $40m^3 \pm 1m^3$. Typical values of entry rates for both the tracers reached values about 300 Bq/m^3 /h and 20 ppm/h respectively. Radon concentration indoors varied from 300 Bq/m^3 to 2000 Bq/m^3 and CO concentration varied from 30 ppm to 300 ppm in dependence on actual ventilation rate through the enclosure. The ventilation rate through the Lab was influenced by the weather only and in addition it can be driven via an adjustable small air gap of slightly opened window. According to our calculations ventilation rate varied from 0.06 1/h to 0.6 1/h. All these experiments took typically from one day to one week. We used with benefit the fact that applied entry rates for both the tracers were constant during all measuring periods and good defined. We were using fan for better mixing of the tracers. Overall uncertainties in determination of ventilation rates for both the tracers was estimated to be less than 20 %.

Radon entry rate

The main idea of all performed experiments consisted in the possibility to compare average values of entry rates calculated according to expressions (7) and (8) to the well define reference value of radon entry rate. During these experiments carried out in our Lab both radon gas and CO gas were injected into Lab with well defined and constant entry rate (see par.3.1.). To get the sufficient number of required data the experiments took typically from three days to one week.

3.3. *Application of CO tracer gas techniques in field to assess the effectiveness of radon remedy*

*T*o assess the effectiveness of radon remedy in the house means to quantify separately both radon entry rate and ventilation rate into the enclosure (i.e. measuring room) generally for two principally similar following situations:

- before remedial action and afterwards (when one-off remedial intervention is applied).
- over the periods of time as used active remedial ventilation system operated neither in mode fan ON or in mode fan OFF.

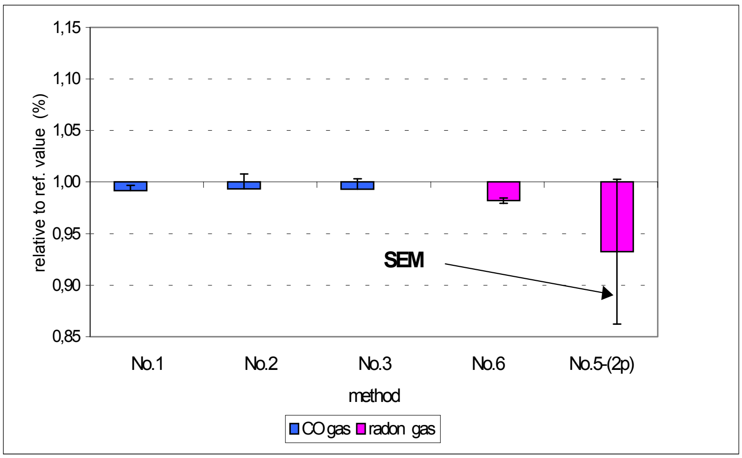
The main commonly used idea in application of the CO gas techniques during the diagnostic measurements consists in possibility to calculate ventilation rate in any time through the mentioned averaging method (see Table1 expression No.3). Then we calculate the radon entry rate according to equation (8) during the two mentioned and investigated situations. Provided that ventilation rate estimated by the averaging method is close to constant over the investigated period, we can estimate the average value of radon entry rate over the measuring period by the proper regression method (see Tab.1). In the regression radon entry rate is treated as single unknown parameter. According to “the air time” from the homeowner point of view we can inject CO tracer gas into the measured space either one-off way (i.e. the CDM method, see Table 1) or invariably (i.e. the CITM method). Typical measuring time for each of investigated situations is approximately 24-48 hours.

4. Results and discussion

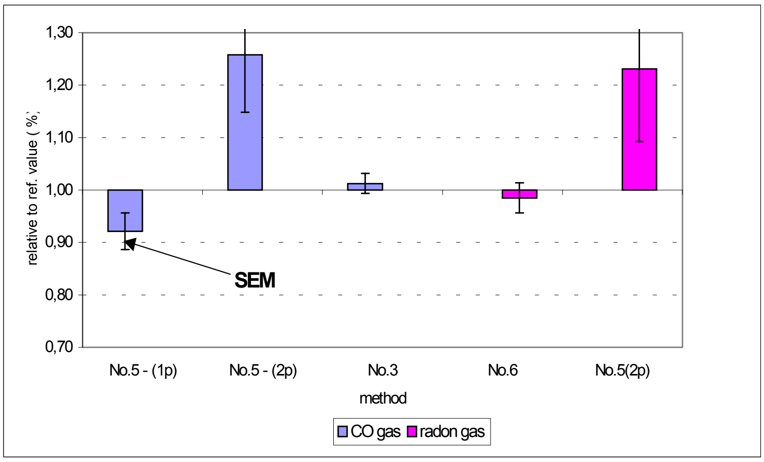
4.1 Ventilation rate

Results of the comparative measurements performed in our Lab with the aim to confirm agreement in determination of ventilation rate through both the radon methods and the CO tracer gas methods (see Tab.1) are shown in Fig.1 and Fig.2. These results are expressed in form of the average relative deviations of each tested approach calculated from all performed experiments from weighted mean calculated from all compared approaches. Considering too outlying results from two parameter non- linear regression (method No.(2p)), its results were not included into the weighted mean. The weighted mean was adopted as a reference value because we had no other possibility to measure ventilation rate through the Lab absolutely.

Fig.1: *Average relative deviations of each separate tested method for determination of ventilation rate defined in Tab.1 from weighted mean calculated from all methods. (situation: one-off injection of CO gas - constant radon gas injection ,expression in bracket "2p" means two parametric non-linear regression).*



Fig,2 : *Average relative deviations of each separate tested method for determination of ventilation rate defined in Tab.1 from weighted mean calculated from all methods. (situation: constant injection both radon and CO gas at the same time),expressions in bracket "1p","2p" means one- or two- parametric regressions.*



From Fig.1 and Fig.2 we can see an acceptable good agreement for all tested methods to determination of ventilation rate for both radon gas and CO gas. On the other hand results of two parametric non-linear regression (see method No.5 in Tab.1) for both the tracers showed statistically significant approximately 20% average relative deviation from the others with relative wide approximately 15 % average standard error of the mean. These results were confirmed at our further work [8] where this method used for radon gas provided average approximately 60 % underestimation against the routine used the averaging technique for CO gas with wide approximately 30 % average standard error of the mean.
Considering that last mentioned two parametric non-linear regression utilizing only radon gas as the tracer, currently makes the basis of radon diagnostics in the Czech Republic, our results would have open a wider discussion about the possible impacts of above mentioned fact at least.

4.2 Radon entry rate

Fiq. 3: *Relative comparison of average values of radon entry rate estimated from our routinely used methods to reference value, method A - radon entry rate is calculated according to equation (7), method B- radon entry rate is calculated according to equation (8), see par. 2.2., abbreviations in bracket behind experiments No. denotes the type of used radon meter.*

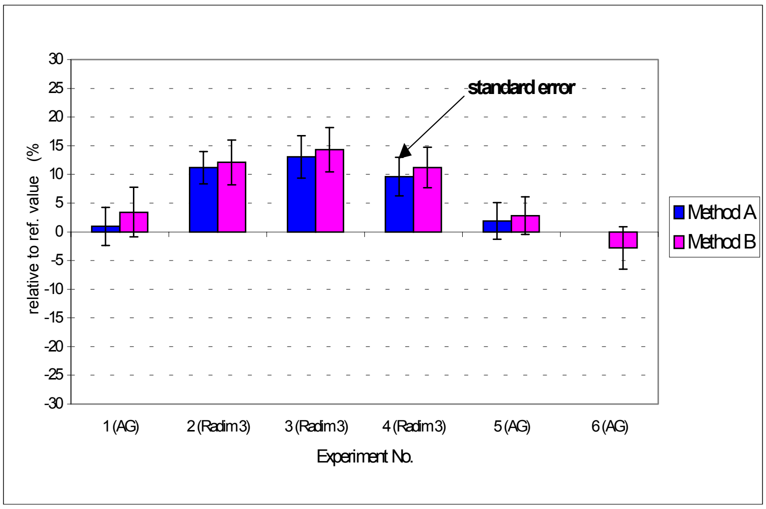


Fig. 3 indicates a very good mutual agreement between tested methods and acceptable less than approx. 15 % average difference for each tested method from the reference value. Further, systematic approximately 10% average overestimation of results from experiments carried out with monitor Radim3 above experiments carried out with monitor Alphaguard was due to approx.10 % difference in absolute calibration between monitors.

4.3. Application of CO tracer gas techniques in field to assess the effectiveness of radon remedy

The typical situation during which one definite remedial measurement, where the CO tracer gas techniques are used, is demonstrated in the following two pictures. To reduce radon entry rate into the house an active remedial ventilation system with fan is used.

Fig.4*: Time variations of radon concentration and CO concentration during both operation modes of the used ventilation remedial system, the constant CO tracer gas injection method was used in both*

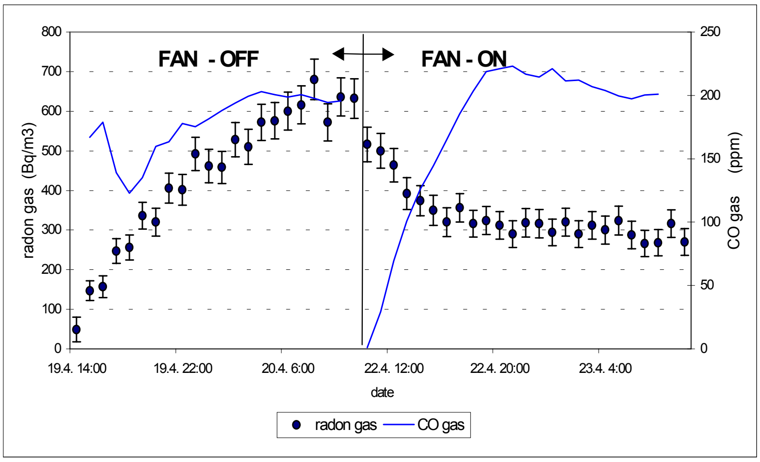
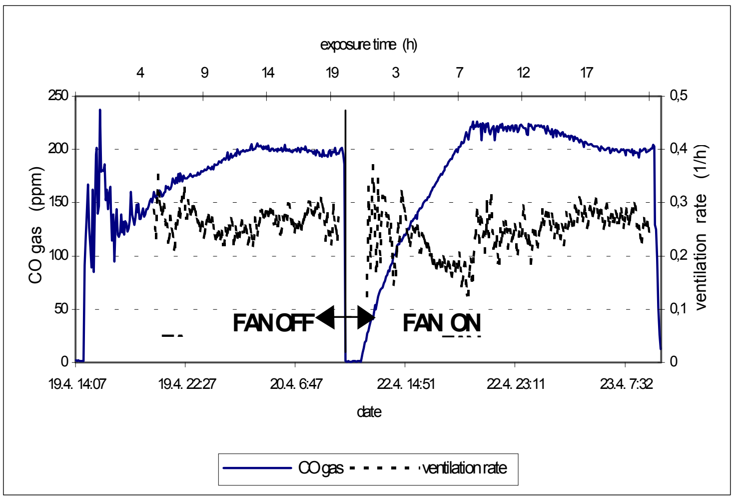


Fig.4 indicates an unusual build- up of CO gas concentration during the situation fan OFF up to several hours after the initial injection caused by the fact that we tried to use no mixing air in the measured space (two connected rooms with estimated effective volume 100 m^3). During the situation fan ON a portable fan for mixing indoor air has been already used.

Fig. 5: *Time variation of ventilation rate calculated through the averaging technique over the whole measured period*



From the Fig.5 we can deduce the following facts:

- The used remedial ventilation system does not have any influence to ventilation rate through the measuring space. The unexpected considerable decreasing of ventilation rate during measurement in situation Fan ON was demonstrably caused only by the weather changes.
- The relatively wide range of estimated ventilation rates approximately up to three hours after initial constant injection of CO gas was known and expected.
- Almost constant values of calculated ventilation rate during both operation modes of used ventilation system allow to use mentioned regression techniques to estimate average values of radon entry for both operating modes of used ventilation system. In these regressions radon entry rate was treated as a unknown parameter. For estimating an average value of radon entry rate during situation fan OFF we can use the regression method No.5 (see Table 1) and during situation fan ON we can use the regression method No.4.

In addition, we calculated time variation of radon entry rate during all the whole remedial measurement according to equation (8).
The results of the above mentioned approaches for the estimation of average values of radon entry rate for both investigated situations are shown in the following Fig. 6.

Fig. 6 *The comparison of used approaches for calculation of average value of radon entry rate, R-method means used regression methods, T- method means method based on calculation of radon entry rate according to equation (8).*

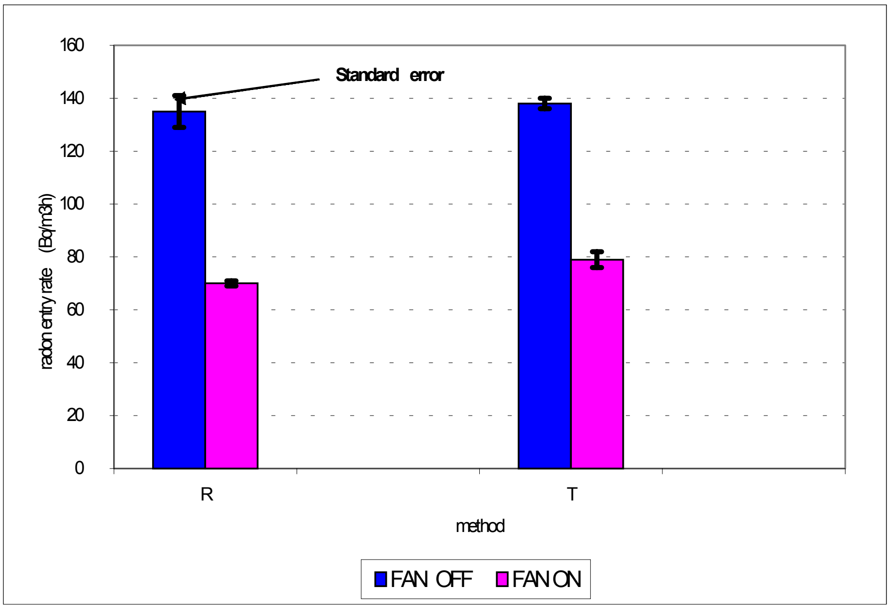


Fig. 6 indicates an expected very good agreement both of the tested methods and also very good effectiveness of accomplished remedy.

Conclusions

We demonstrated very good practical results of applicability of CO tracer gas methods under the real situation of remedial measurement.

We stress the following main advantages of used CO tracer gas techniques:

- The outlined averaging method used for CO data enables through the good continuous monitor of CO gas continuous determination of ventilation rate with good accuracy up to 20 %. Another more hands-on approach to determination of ventilation rate through the CO data from continuous monitor based on the Kalman filtering is demonstrated in [9]. The key property of this statistical model is flexibility and its practical software implementation.
- The newly presented and verified method based on simultaneous measurements of radon concentration and CO gas concentration enables separate continuous measurements of radon entry rate and ventilation rate.

The results of comparative measurements performed with the aim to estimate the inaccuracy in determination of radon entry rate showed acceptable and good agreement up to approximately 10 % .

The results of comparative measurements performed with the aim to estimate the mutual commensuration of used method to determination of ventilation rate confirmed the expected unreliability the two parametric non-linear regression method, most often used in framework of radon diagnostic in the Czech Republic.

Recently our continuous monitor of CO gas has been refilled by a pair of N_2O IR sensors. Due to this fact we can measure indoor ventilation rate in the presence of people and we are able to utilize the existing two zones ventilation model.

References

[1] R. McCullough., Leak testing, ASTM Standardization news, 10 (11), pp. 32-3 (1982)

[2] G.J. Feber,. et.al., Demonstrations of a long range atmospheric tracer system using PTF, BNL – 33846, Brookhaven National Laboratory (1983)

[3] P. Lagus, A.K. Persily., A review of tracer gas techniques for measuring airflows in buildings, ASHRAE Transaction 91 (IIB) p.1075 – 1087 (1985)

[4] M.H. Sherman, D.T. Grimsrud, P.E. Condon, B.V. Smith., Air infiltration measurement techniques, Proceeding 1st AIC Conference Air Infiltration Instrumentation and measuring techniques, Air Infiltration and Ventilation Centre, Coventry UK, (1980)

[5] M.H. Sharmen., Air infiltration measurement techniques, LBL- 10705, Brookhaven National Laboratory (1989)

[6] J. Thomas., Personal letter, National Radiation Protection Institute, Prague, CZ, 2004

[7] K. Jilek,. Continuous monitor of CO gas for determination of ventilation rate, Report of Institutional Research , National Radiation Protection Institute , Prague, CZ, 2001-2

[8] K. Jilek,. Continuous monitor of CO gas for determination of ventilation rate, Report of Institutional Research , National Radiation Protection Institute , Prague, CZ, 2003

[9] M.Brabec., K. Jilek, Improved air ventilation rate estimation based on a statistical model, Proceeding 4 th European Conference on Protection against radon at home and at work, Prague, CZ, 2004