INTERNATIONAL INTERCOMPARISON MEASUREMENT OF SOIL-GAS RADON CONCENTRATION, OF RADON EXHALATION RATE FROM BUILDING MATERIALS AND OF RADON EXHALATION RATE FROM THE GROUND

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Martin Neznal & Matěj Neznal

RADON, v.o.s. corp., Novákových 6, 180 00 Praha 8, Czech Republic

ABSTRACT

The International Intercomparison Measurement was held in Pribram, Czech Republic in September 2002. It was attended by participants representing 10 different institutions from 8 countries. A test site for the intercomparison of soil-gas radon concentration and of radon exhalation from the ground was located in the field, at an area characterized by a high soil permeability and by relatively high values of soil-gas radon concentration. A concrete block made from a slag with a high content of radium was used for the intercomparison of radon exhalation rate from building materials. As for the soil-gas radon concentration and the radon exhalation from the concrete block, a relatively good agreement was observed. The intercomparison differences expressed as a ratio of the standard deviation to the arithmetic mean were about 19 percent in case of radon exhalation from the concrete block, and 28 percent in case of soil-gas radon concentration, respectively. Larger differences were observed during measurements of radon exhalation rate from the ground surface. The value of the ratio SD/mean was 0.43.

INTRODUCTION

This report describes results of the International Intercomparison Measurement of Soil-gas Radon Concentration, of Radon Exhalation Rate from Building Materials and of Radon Exhalation Rate from the Ground Surface, which was held in Pribram, Czech Republic in September 2002 and organized by RADON v.o.s. corp. The original idea to organize the intercomparison appeared at the ERRICCA 2 kick-off meeting in London in February 2002.

The soil-gas radon (^{222}Rn) concentration **c** ($kBq.m^{-3}$) is defined as an average radon concentration in the air-filled part of soil-pores in a given volume of soil-gas. Radon exhalation rate **J** ($mBq.m^{-2}. s^{-1}$) is defined as an average activity of radon emitted from a given surface per unit area and per unit time. First of all, both parameters are used for characterizing the radon potential of soils, but a large range of other applications is known: uranium prospecting, earthquake prediction, risk assessment of waste materials, etc.

Conclusions resulting from several previous intercomparison measurements were available: the International Intercomparison of Measurement of Radon and Radon Decay Products in Badgastein, Austria, 1991 (Cliff et al. 1994), the Sixth International Radon Metrology Programme Intercomparison Test and Workshop, New York, U.S.A., 1995 (Hutter and Knutson 1996, 1998), the International Intercomparison Measurement of Soil-gas Radon Concentration and of Radon Exhalation Rate from the Ground, Praha, Czech Rep., 1996 (Neznal et al. 1996, 1997). As many of these conclusions are still valid, it seems useful to quote several sentences from the last of the above mentionned reports:

„It is obvious that from metrological point of view there are many serious problems connected with organizing any field intercomparison measurement of these parameters. The natural geological environment is almost never homogeneous. The soil-gas radon concentration as well as the radon exhalation rate from the ground may vary, often very greatly, over a small distance, the variations of soil-gas radon with depth are different under changing geological conditions (Hinton 1985, Robé et al. 1992, Washington and Rose 1990, Neznal et al. 1996b). Field intercomparison measurements thus are not intended to be used as an intercalibration of methods and instruments (Hutter and Knutson 1996). They are designed as an intercomparison of results obtained using different instruments and methods employed in the field in order to assess the ability to interrelate diverse measurements. Under these circumstances, values are not reported against a standard or reference measurement.

Participants results are simply compared to each other, in order to obtain an indication of the collective precision of various measurements.“

Geological conditions in a depth of soil-gas sampling as well as conditions on the soil surface should be as homogeneous as possible at the test site. If these requirements are not fulfilled, a large variability of measurement results can be expected. On the other hand, any preliminary measurements should be limited at a chosen test site to avoid a situation that the upper soil layers will remind of a Swiss cheese.

PARTICIPANTS AND METHODS

The intercomparison exercise was attended by participants representing 10 different institutions from 8 countries - France, Germany, Hungary, Poland, Romania, Spain, United Kingdom and Czech Republic. As only some participants compared all three parameters, the number of groups who measured the same parameter ranged from 6 to 8.

The information on measuring methods is given in Table 1.

TEST SITE AND TEST BUILDING MATERIAL

The intercomparison exercise of soil-gas radon concentration and of radon exhalation rate was organized in the field at a reference area near the village Buk, less than 10 km from Pribram. The reference site lies 2 km NNE from Milin, 550 m a.s.l., several meters from the road Buk - Radetice. Geological basement is formed by a medium grained biotitic and amphibol-biotitic granodiorite (Milin type) of the Central Bohemian pluton of Paleozoic age. The eluvial granitic material forms the cover (Matolin 2002). Expected values of soil gas radon concentration are relatively high, soil is highly permeable at the test site. A meadow is on the surface. There is a slight downward gradient to the road Buk - Radetice. At the test site, 12 reference points were marked in a 5 x 5 m grid.

A uniform sampling depth of 0.8 m below the ground surface was recommended for soil-gas radon concentration measurements. The participants were also asked to make several measurements of both parameters in different measuring points and to define the exact locations of their measuring points in the protocol. This approach should enable to decrease the influence of a spatial variability, because not single values, but sets of data would be compared. Last but not least: It was recommended to place the devices for radon exhalation measurements before starting of soil-gas sampling in order to avoid a possibility that an accumulator would be placed on an open hole remaining after soil-gas sampling.

As some participants (A, Q) required a longer exposure period for radon exhalation measurements, the first measurement devices were located at the test site on September, 18, in the evening, about 44 hours before the beginning of a common field exercise. To get an information on temporal variability of radon exhalation, the organizers (participant N) made two sets of radon exhalation measurements - the first one on September 19, in the morning, the second one together with other participants on September, 20, in the afternoon. The same approach was used also for soil-gas radon concentration measurements: the organizers repeated the measurements twice. They have also collected soil-gas samples from a depth of 0.5 m below the surface in order to be able to estimate variations of soil-gas radon concentration with depth.

Table 1. Methods.

Participant’s code	Methods
A	*Radon (radon + thoron) exhalation rate from the ground surface* A couple of solid state nuclear track detectors - KODALPHA detectors under a simple accumulator (the same type as participant N); the first detector exposed inside a calibrated goblet („closed detection“) to get radon exhalation rate, the second detector exposed as a free detector („open detection“) to get radon + thoron exhalation; exposure time about 47 hours; detectors and calibrated goblets were sent by mail; exposure of detectors at the test site was arranged by the organizers (participant N); ventilation factor 0.4 h^{-1} was used for calculation.

Ap	*Soil-gas radon concentration* Depth of measurement ranging from 0.55 to 0.6 m (the measurement depth is the range between the final depth of the outer casing and the point of the inner rod of the soil-gas sampling probe); EDA RD200 portable radon detector; 3 consecutive one minute counts.
B	*Radon exhalation rate* A single accumulator method; air samples collected using a syringe and transfered into previously evacuated Lucas cells; 6 samples collected in regular 15 minutes intervals.
C	*Soil-gas radon concentration* Two different methods - (a) activated charcoal + special pumping, (b) Lucas cell; (a) 64 g of activated charcoal with adsorption constant k = 1.14 (Bq/kg)/(Bq/m^3) put into a flexible tube (40 cm); soil-gas (2 l) extracted under controlled pressure by water empting of a plastic bottle; radon activity adsorbed in the charcoal measured by gamma spectrometry (NaI (Tl) detector) in the laboratory 4 days after sample collection; calibration factor for the whole spectrum (E>60 keV) 3.01 Bq:count/s; detection limit 2 kBq/m^3; two consecutive samples were collected; (b) LUK-3A device (produced by Jiri Plch - SMM Prague) in the „Radon-fast“ mode; sampling depth 0.8 m; soil-gas sampling using a 145 ml syringe; several samples were collected consecutively. *Radon exhalation rate* Two different methods - (a) charcoal adsorption, (b) Lucas cells; (a) 64 g of activated charcoal closed in a plastic canister (1.5 l, measured area 0.02 m^2, exposure time 3.5 hours - for building material), or in a metallic canister (0.8 l, measured area 0.066 m^2, exposure time 2.66 hours - for ground surface); radon activity adsorbed in the charcoal measured by gamma spectrometry (NaI (Tl) detector) in the laboratory 4 days after exposure; detection limit 4 mBq/m^2.s; a leakage correction factor of 15% (building material), or of 10% (ground surface) was applied to the calculated values; (b) accumulator placed on a measured surface (3 l, measured area 0.032 m^2, exposure time 3.5 hours - for building material; 8 l, measured area 0.091 m^2, exposure time 2.94 hours - for ground surface); samples collected using a 145 ml syringe; two samples collected consecutively and measured in three consecutive 100 s intervals using a LUK-3A device in the „CNT“ mode; detection limit 6 mBq/m^2.s; a leakage correction factor of 30% was applied to the calculated values.

Table 1.

Participant's code	Methods
F	*Soil-gas radon concentration* Two different sampling systems: (a) packer probe; (b) simple small-diameter probe with a free sharpened lower end (the same as participant N); sampling depth 0.8 m; RTM 2010 active radon monitor; integration time 1 minute; reported radon concentration = concentration at saturation.
Ch	*Soil-gas radon concentration* Sampling depth 0.8 m; Lucas cells; scintillometer GF-10 + RGR; counting about 3 hours after sample collection. *Radon exhalation rate* A single accumulator; air samples transferred into Lucas cells after about 3 hours of exposure; measurement about 3 hours after sampling.
K	*Soil-gas radon concentration* Sampling probe: stainless cylinder (lenght 1 m, inner diameter 22 mm); in the wall of the probe there are 6 holes situated in a spiral on a 20 cm long section; the section is situated 10 cm from the end of the probe; sampling depth 70±5 and 52±4 cm, respectively; Lucas cell; pumping and measurement using AB-5 apparatus; pumping time 30 minutes; counting starts 3 hours after the end of pumping.

M	*Soil-gas radon concentration* Sampling depth 0.8 m; device Markus 10. *Radon exhalation rate* Two different methods - (a) electret dosemeters; (b) AlphaQuard.
Q	*Soil-gas radon concentration* Two different methods - (a) RTN 2010 active radon monitor; (b) soil-gas samples collected using Lucas cells (1 l). *Radon exhalation rate* Two different methods - (a) accumulator method; air samples transferred into Lucas cells after 1 hour and 2 hours of exposure; measurements 3 hours after sampling; (b) activated charcoal (24 hours of exposure) measured by gamma spectrometry.
N	*Soil-gas radon concentration* The equipment for soil-gas sample collection consists of a small-diameter hollow steel probe with a free, sharpened lower end; sampling depth 0.8 m (samples were collected also from a depth of 0.5 to estimate variations of soil-gas radon concentration with depth); soil-gas samples collected using a syringe and introduced into previously evacuated Lucas cells (0.125 l); counting in the laboratory more than 3 hours after sample collection (one 400 second count; device LUK). *Radon exhalation rate* A single accumulator method; accumulator (cylindrical canister 0.2 m high; measured area 0.08 m^2) placed on an undisturbed soil surface or on a surface of measured building material and sealed; air samples collected using a syringe and transferred into previously evacuated Lucas cells; 4 samples collected in regular time intervals (usually ranging from 30 to 40 minutes); counting in the laboratory more than 3 hours after sample collection.

The weather during the intercomparison exercise (September 20, in the afternoon) was variable. It was mostly overcast in the beginning, with some drizzle, and the soil surface was wet. But after one or two hours the sun also appeared. The temperature of air was about 15°C.

The intercomparison exercise of the radon exhalation rate from a concrete block was organized in a dwelling of National Institute of Nuclear, Chemical and Biological Protection near the mine No. 9, Pribram. A concrete block made from a slag with a high content of radium was used (2820 $Bq.kg^{-1}$). As the exhalation rate from the block is not quite homogeneous, 8 different measuring positions were defined (see Fig. 1). Each position was a circle with a diameter of about 30 cm. Radon exhalation rate from each position had been determined using the same method (RADON, v.o.s.; simple accumulator method) before the intercomparison, on June, 12. The results are given in Table 2. These values were then used as a base for the intercomparison, which was held on September, 20, in the morning.

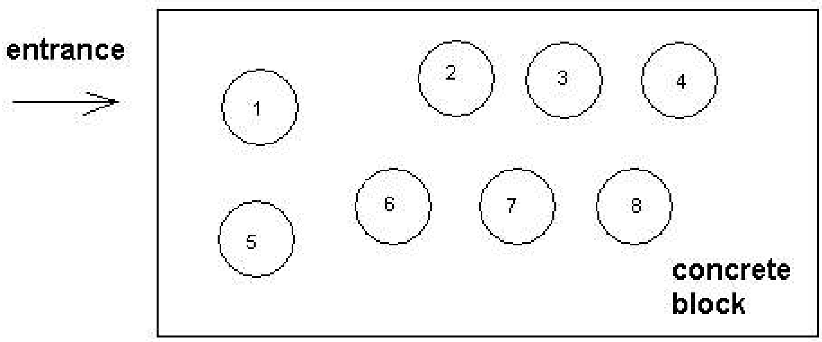

Figure 1: Measuring positions on a surface of the concrete block.

Table 2. Preliminary measurements of radon exhalation rate from a concrete block (June 12, 2002).

Measuring position	Exposure period	Radon exhalation rate ($mBq.m^{-2}.s^{-1}$)	Correction factor eliminating spatial variations
1	10:25 -12:25	32	1,57
2	10:30 -12:30	40	1,26
3	10:38 -12:38	34	1,48
4	10:44 -12:44	47	1,07
5	10:50 -12:50	46	1,09
6	10:56 -12:56	70	0,72
7	11:06 -13:06	63	0,80
8	11:12 -13:12	70	0,72

RESULTS AND DISCUSSION

Soil-gas radon concentration

The minimal number of measurements made by each participant was 2, four participants realized 5 measurements or more. Real sampling depths ranged from 0.5 to 0.8 m. Results of repeated measurements were also reported (participant N).

The intercomparison of all participating institutions is given in Table 3. Results of repeated measurements and results of measurements made at a depth of 0.5 m (participant N) were not taken into account, i.e. the whole set of data consisted of 42 values of soil-gas radon concentration. The spread of all data characterized by the ratio of an arithmetic mean and of a standard deviation was 0.33. Mean values reported by different participants ranged from 77.5 to 213 $kBq.m^{-3}$, ratios SD/mean from 0.08 to 0.49. If only data sets containing at least 4 values are considered, then the mean values reported by different participants range from 125 to 213 $kBq.m^{-3}$ and the ratios SD/mean from 0.08 to 0.30.

Table 3. Intercomparison of soil-gas radon concentration data reported by different participants.

Participant's code	Number of meas.	Sampling depths (m)	Soil-gas radon concentration (kBq.m^{-3})					SD/mean
			min.	max.	median	mean	SD	
Ap	5	0.55 - 0.6	114	141	124	125	9.9	0.08
C	6	0.8	109	238	143	155	45.8	0.30
F	2	0.8	87.0	176	(132)	132	62.9	0.49
Ch	9	0.8	129	274	232	213	54.7	0.26
K	2	0.52 - 0.7	70.0	85.0	(77.5)	77.5	10.6	0.14
M	4	0.8	150	201	191	183	22.8	0.12
Q	2	0.8	120	190	(155)	155	49.5	0.32
N	12	0.8	76.0	207	136	139	34.9	0.25
Total	42	0.55 - 0.8	70.0	274	145	157	51.9	0.33

A more detailed analysis of the data enables to check a possible influence of temporal and spatial variations and the influence of changes of soil-gas radon concentration with depth. The influence of temporal changes and changes with depth can be estimated using measurements realized by the organizers (participant N). At first, two sets of measurements were made from the depths of 0.5 and of 0.8 m at the same time, i.e. from 15:26 to 16:34. Sampling from the depth of 0.8 m was then repeated from 17:27 to 17:45. As can be seen in Table 4, the influence of temporal changes of soil-gas radon concentration to the intercomparison results was low and it can be probably neglected. On the other hand, the soil-gas radon concentrations measured at 0.5 m below the sufrace were significantly lower than radon concentration at the depth of 0.8 m. Soil-gas radon concentrations at the depth of 0.5 m represented only about 70% of the values that were measured at the depth of 0.8 m. It is evident that a correction of this type would improve the agreement between all participating laboratories.

Table 4. Results of repeated measurements of soil-gas radon concentration made by participant N.

Time period	Number of meas.	Sampling depths (m)	Soil-gas radon concentration (kBq.m^{-3})					SD/mean
			min.	max.	median	mean	SD	
15:26 - 16:29	12	0.5	64.5	132	93.3	94.9	20.0	0.21
15:28 - 16:34	12	0.8	76.0	207	136	139	34.9	0.25
17:27 - 17:45	12	0.8	57.3	171	139	131	29.4	0.22

Other interesting findings were obtained when the variability of the data was studied in a horizontal level. The comparison of mean soil-gas radon concentrations that were measured by different participants near to each of 12 measuring points is given in Table 5.

Table 5. Spatial variability of soil-gas radon concentration - analysis of measurements that were made by different participants near to each of 12 reference points.

Reference point	Number of meas.	Soil-gas radon concentration (kBq.m^{-3})					SD/mean
		min.	max.	median	mean	SD	
1	6	109	195	141	151	32.4	0.21
2	6	85.0	266	140	152	61.0	0.40
3	9	70.0	274	138	146	61.2	0.42
4	5	57.0	131	76.0	90.8	32.6	0.36
5	6	101	251	131	145	55.5	0.38
6	9	85.0	238	125	148	52.5	0.36
7	7	70.0	181	124	119	36.0	0.30
8	6	70.0	190	133	132	40.8	0.31
9	6	111	207	123	145	40.8	0.28
10	5	121	243	131	150	52.0	0.35
11	6	114	171	134	138	20.4	0.15
12	6	87.0	207	174	158	46.7	0.30

Note: Some of measurement results were used several times. For example the soil-gas radon concentration measured by participant Ap in measuring point A (=centre of the square 7, 8, 11, 12) was used four times, at reference points 7, 8, 11 and 12. Results of repeated measurements of participant N at the depth of 0.8 m were included, results of measurements of participant N at the depth of 0.5 m were not included.

Values of soil-gas radon concentration measured in the surroundings of the reference point No. 4 seem to be lower than soil-gas radon concentrations at the rest of the reference area.
This local decrease might also affect the spread of data.

As the soil permeability at the reference area was relatively high, it was possible to expect only a rare occurence of sampling errors during the intercomparison exercise, i.e. only a low number of cases when the sealing of a sampling system failed and the soil-gas radon concentration was underestimated. In our opinion, there were three main sources of the spread of intercomparison data: (a) spatial variability of soil-gas radon concentration at the reference area, (b) changes of soil-gas radon concentration with depth and the fact that several participants collected their samples from other depths than 0.8 m, (c) differences connected with primary calibration of instruments. If geological conditions are homogeneous in the sphere of interest, intercomparison differences of 20 percent can be achieved (Neznal et al. 1996). This requirement was fulfilled only partly. In order to decrease the influence of factors (a) and (b) it is possible to exclude measurements that were made in the surroundings of point No. 4. After these corrections the value of SD/mean for the total set of data (see Table 3) has changed from 0.33 to 0.28.

The differences connected with primary calibration are usually lower than 10 percent. For example the 27th EML Radon Gas Intercomparison (Fisenne 1995) resulted in the value of SD/mean equal 0.08, when mean values of radon concentration in air reported by 30 participants were compared.

If not the results of all single soil-gas radon concentration measurements, but only mean values reported by each participant (Table 3) are used for the intercomparison, then SD/mean = 0.27 and after corrections described above SD/mean = 0.26. At the Prague intercomparison in 1996, the spread of mean soil-gas radon concentrations reported by different participants was characterised by SD/mean = 0.14 (Neznal et al. 1996). Some problems with primary calibration thus cannot be excluded.

Radon exhalation rate from the ground surface

The intercomparison of participating laboratories is given in Table 6. Only three participants realized 4 measurements or more. Results of preliminary measurements (participant N; 19.9.) were not considered. The whole set of data contained 21 values of radon exhalation rate from the ground. The spread of all data was large, the ratio of an arithmetic mean and of a standard deviation was 0.43. Mean values reported by different participants ranged from 37 to 159 kBq.m^{-3}, ratios SD/mean from 0.14 to 0.37.

The influence of temporal changes were checked again using measurements realized by the organizers (participant N). Two sets of measurements were made, the first one from 7:20 to 9:47, September 19, the second one from 15:03 to 17:53, September 20. Results are summarized in Table 7. As can be seen, the influence of temporal changes of radon exhalation from the ground surface to the intercomparison results was low.

Spatial variations of radon exhalation rate from the ground surface were comparable with spatial variations of soil-gas radon concentration (see last columns of Tables 3 and 6).

In our opinion, the observed differences among participating laboratories are mainly systematic, but available descriptions of measuring methods do not allow any more detailed analysis. There are two important points (Neznal and Neznal 2002): (1) When a simple accumulator is used for the determination of radon exhalation rate, the radon concentration in the accumulator increases due to the radon entry and it decreases due to the radioactive decay and due to radon losses caused by ventilation of the accumulator. The radon losses during measurement strongly influence measurement results. A constant describing radon losses should be known and included into calculations. (2) Measured values of the radon exhalation rate from the ground surface are substantially affected by conditions on the soil surface. For example, reported values are usually lower when the accumulator is placed on an undisturbed soil surface, than in case when the upper soil layer is removed before measurement and the accumulator is placed several cm below the ground surface.

Note: If our notes are complete, only participants A and N measured the radon exhalation rate from an undisturbed soil surface. The above mentionned expectation (2) was thus not confirmed during the intercomparison exercise.

Table 6. Intercomparison of radon exhalation rate from the ground - data reported by different participants.

Participant's code	Number of meas.	Radon exhalation from the ground ($mBq.m^{-2}.s^{-1}$)					SD/mean
		min.	max.	median	mean	SD	
A	5	44	85	66	66	16	0.25
B	1				159		
C	2	33	41	(37)	37	5.7	0.15
Ch	4	27	77	70	61	23	0.37
M	2	84	122	(103)	103	27	0.26
Q	2	62	82	(72)	72	14	0.20
N	5	51	73	62	61	8.7	0.14
Total	21	27	159	66	68	29	0.43

Table 7. Results of repeated measurements of radon exhalation rate from the ground surface made by participant N.

Time period	Number of meas.	Radon exhalation from the ground ($mBq.m^{-2}.s^{-1}$)					SD/mean
		min.	max.	median	mean	SD	
19.9. 7:20 - 9:47	5	45	75	52	57	13	0.23
20.9. 15:03 - 17:53	5	51	73	62	61	8.7	0.14

Radon exhalation rate from building material

Results of measurements of radon exhalation rate are presented in Figure 2. Five institutions participated in the intercomparison, three of them realized 2 measurements. The correction factors given in Table 2 were used to minimize the influence of spatial variations of radon exhalation rate.

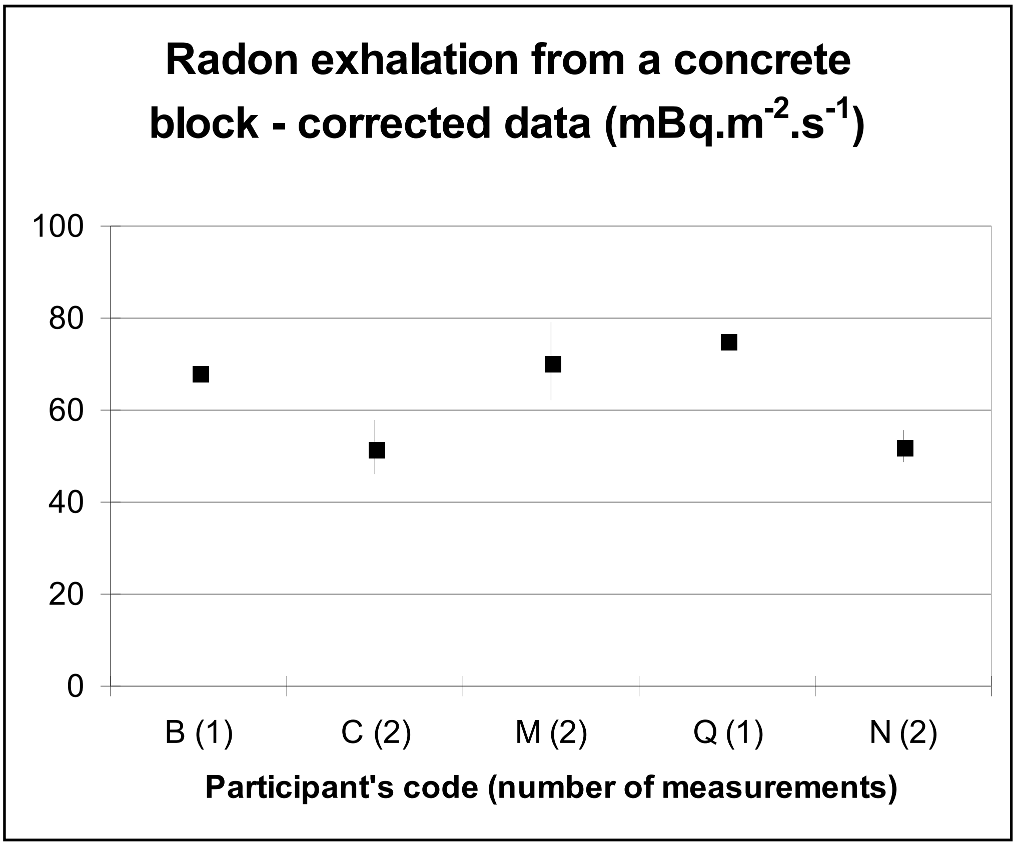

Figure 2. Intercomparison of radon exhalation rate from the surface of a concrete block - corrected data.

Measured and corrected values of radon exhalation rate from the surface of a concrete block are summarized in Table 8. Measured values ranged from 31 to 110 mBq.m^{-2}.s^{-1}, the spread of original data expressed as a ratio SD/mean was 0.39. Corrected data ranged from 46 to 79 mBq.m^{-2}.s^{-1}, the value of SD/mean was 0.19. This agreement among participants is good.

During the E.M.L. intercomparison exercise in 1995 (Hutter and Knutson 1996, 1998), the radon exhalation rate was measured on the surface of an artificially prepared concrete slab with a high content of radium. Identically sized grains and a radium solution were used to get a homogeneous material. Reported values ranged from 81.5 to 490 mBq.m^{-2}.s^{-1}, the value of SD/mean was 0.37 (17 measurements; 8 participating laboratories).

Table 8. Measurement of radon exhalation rate from the surface of a concrete block - statistical evaluation of measured and corrected data.

	Number of meas.	Radon exhalation rate (mBq.m^{-2}.s^{-1})					SD/mean
		min.	max.	median	mean	SD	
measured values	8	31	110	65	69	27	0.39
corrected values	8	46	79	60	62	12	0.19

CONCLUSIONS

As for the soil-gas radon concentration measurements, the intercomparison differences expressed as a ratio SD/mean were 33 percent. A more detailed analysis, including the assessment of spatial variations and of changes of soil-gas radon concentration with depth, resulted in the conclusion that a more exact estimate of intercomparison differences was 28 percent. A realistic target for the intercomparison measurements of soil-gas radon concentration and of radon exhalation rate is about 20 percent. The intercomparison results are thus relatively good, but some differences in primary calibration cannot be excluded.

Larger intercomparison differences were observed during measurements of radon exhalation rate from the ground surface. The value of the ratio SD/mean was 0.43. The observed differences among participating laboratories were probably mainly systematic, but available descriptions of measuring methods did not allow any more detailed analysis.

The third parameter, that was compared, was the radon exhalation rate from the surface of a concrete block. The spread of original data expressed as a ratio SD/mean was 0.39. Correction factors, which had resulted from preliminary measurements, were then used to minimize the influence of existing spatial variations. Corrected data ranged from 46 to 79 mBq.m^{-2}.s^{-1}, the value of SD/mean was 0.19, i.e. the agreement among participants was acceptable.

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