MEASUREMENT OF RADON IN NATURAL GAS AND IN PROPANE USING ELECTRET ION CHAMBERS

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Abstract

Electret ion chambers (known by the trade name E-PERM® [2]) have been extensively used for measuring indoor and outdoor radon concentration in air. In view of the recent interest in measuring radon in natural gas, research is initiated to devise arrangement for sampling and analyzing radon in natural gas. Natural gas is chemically very different from air both in terms of density and ionization potentials (energy needed to produce one ion pair) and is expected to have a response different from that of air. Further, electret ion chambers (EICs) use ionization measurements compared to alpha counting used in scintillation cells, the other technique standardized for measuring radon in natural gas. Research results are presented in this paper, intercomparing the two technologies for measurement of radon in natural gas. When radon is measured in natural gas using scintillation cells, the calibration factors derived for air are used. The results need to be divided by a correction factor *f* to arrive at proper results. Kitto determined this factor experimentally to be 1.07 for scintillation cells. The current work determined this factor for EIC to be 1.10, only slightly different from the correction factor for scintillation cells. Large numbers of intercomparison experiments are conducted by collecting the samples from the same source at the same time, both by the EIC system and by scintillation cells. Results indicated excellent agreement confirming the performance of the sampling and analysis system for EIC. The *f* factor was found to be 1.36 for propane when measured with EIC.

Key words: scintillation cell, electrets, electret ion chamber, natural gas, propane, W values

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Introduction

Natural gas is widely used as a domestic fuel for cooking, heating and many other applications. The sources of natural gas can be from natural gas producing wells or from the natural gas produced by hydraulic fracturing of shale located deep in the ground. In all cases the natural gas comes from the ground and is expected to have radon accompanying it. The natural gas in the pipelines closer to the wells may have more radon than the pipelines farther away from the source because of the radioactive decay of radon. There is a possibility of leakage of natural gas containing radon into the ambient air. Radon can be released into a home through the combustion or burning of natural gas.

The measurement of radon in natural gas has been of interest for a long time. Due to their sensitivity and ease of use, alpha scintillation cells are being increasingly used for the measurement of radon in natural gas. Most of the available data on radon in natural gas in existing literature is based on the use of these devices. Usually the calibration constants standardized for measuring radon in air are used to calculate radon concentration in natural gas. Recently, Kitto (2014) and Jenkins (2014) indicated that the calibration constants derived for measuring radon in air are not appropriate for calculating radon in natural gas due to inherent differences in density. After a series of experiments, Kitto (2014) concluded that the correction factor is 1.07 when measured at atmospheric pressure and at room temperature. That means the measured radon concentration in natural gas using calibration constants for air need to be divided by 1.07 to calculate the correct results. This is termed in this presentation as the *f* factor. It is also pointed out by Jenkins (2014) that such factors can be different at various elevations, due to different pressures at the corresponding elevations.

Electret ion chambers (EICs), which are widely used for indoor and outdoor radon measurements, can also be used for measuring radon in natural gas. These have been used by Nenznal (1996) for a large number of radon measurements in natural gas within the concentration range of 132 to 195 pCi/L. He has also made a few measurements using scintillation cells in order to confirm the results from the EICs. The EICs work on a very different principle (ionization) compared to scintillation cells (alpha scintillation counting). The object of the present work is to determine the correction factor *f* for measurements of radon in natural gas using EICs.

The final purpose of the current work is to intercompare the results as measured by scintillation cells and as measured by EICs from a sample taken from an identical source. For this purpose, the same natural gas is sampled both by scintillation cells and by EICs. Scintillation cells are sent to Dr. Kitto for analysis. EICs are analyzed at Rad Elec labs. Results are compared and discussed in light of the technological differences. There is an important difference in analyzing the sample between scintillation cells and EICs. The samples collected by scintillation cells can be analyzed after a delay of 4 hours or more; whereas, the sample collected by an EIC has to be held for some period in the sampling device (1 to 8 days) before proper analysis is possible. A delay correction needs to be applied in order to calculate radon concentration at the time of collection. Delay corrections are also needed if the scintillation cell is analyzed after a known delay.

Commercially available propane is another gas used as fuel for cooking. This is not expected to have radon simply because it is obtained by distillation of crude oil, not from the ground as in the case of natural gas. The only distinction is that the density of propane (1.5 relative to air) is very different from that of natural gas (which is predominantly methane) with much lower density. It is of interest to determine the correction factor *f* for radon in propane gas only as the demonstration of the technique.

The significance of measuring radon in natural gas

Wojcik (1989) has done the calculation of the release of radon into a home atmosphere via burning of natural gas. The following parameters were assumed in Wojcik's experiment: the radon concentration in natural gas is 235 Bq/m^3 (6.4 pCi/L), the kitchen volume is 25 m^3, there are three air changes per hour, daily gas consumption is 1 m^3, and a cooking time of two hours per day. In this scenario, the mean radon concentration of 40 Bq/m^3 (1.1 pCi/L) will be raised by 1.5 Bq/m^3 (0.05 pCi/L) during cooking time and by only 0.13 Bq/m^3 (0.004 pCi/L) on a daily average. These calculations indicate that there is no appreciable contribution to radon in indoor air by the use of natural gas in homes. The situation can be different if the air exchange rate is different than what is assumed or radon concentration in natural gas is different than what is assumed.

Early studies by USGS reported well head concentrations between 0.2 to 1450 pCi/L (Johnson, 1973), Devonian shale level of 151 pCi/L (Gogolak 1980), and Marcellus shale levels of 1 to 79 pCi/L with an average of 37 pCi/L (Rowan 2012). The current studies indicated a measured radon concentration of radon in natural gas at a home in Frederick, MD at approximately 30 pCi/L. This illustrates that the radon in natural gas is not a significant problem in homes. However, there is always a need for technology which allows the measurement of radon concentration in natural gas and in other gases for research and exploration.

Materials and methods

Figure (1) shows the basic equipment used as an accumulator with EIC units used for several applications, including the measurement of radon in water, measuring radon emanation rate from soil and building materials, and for calibrating EICs using NIST radon emanation standards. It consists of a wide-mouth glass jar with a screw cap and a rubber collar that can be tightened to make the unit leak-proof. Two EIC units can be accommodated inside the jar. One important application is to use it for basic calibration of EIC radon monitors (Kotrappa 1994). Knowing the emanation characteristics of NIST emanation standards, it is possible to calculate the expected radon concentration inside the jar after any desired accumulation period and compare this with the EIC measured average radon concentration. This is an air-tight system usable as an accumulator for different applications. The same unit is modified to serve as the system for sampling natural gas as shown in Figure (2). There are two valves which can be opened or closed. The natural gas line is connected through the inlet valve and is allowed to escape via the outlet valve. Once the sample is taken, the valves are closed. EICs measure the average radon concentration inside the jar after any length of retention. When used with NIST emanation standards EICs measure the accumulated average radon concentration. In the present work of determining the response factors for radon in natural gas, there is a need for the radon sources,

which give higher emanations. Two NIST emanation radon standards were available with the following characteristics: Source 1 (SRM 4972), radium strength of 52.04 Bq (NIST-H), emanation coefficient 0.867, and Source 2 (SRM4971-34), radium strength of 5.082 Bq (NIST-L), emanation coefficient 0.891. These are fully described in Kotrappa (1994).

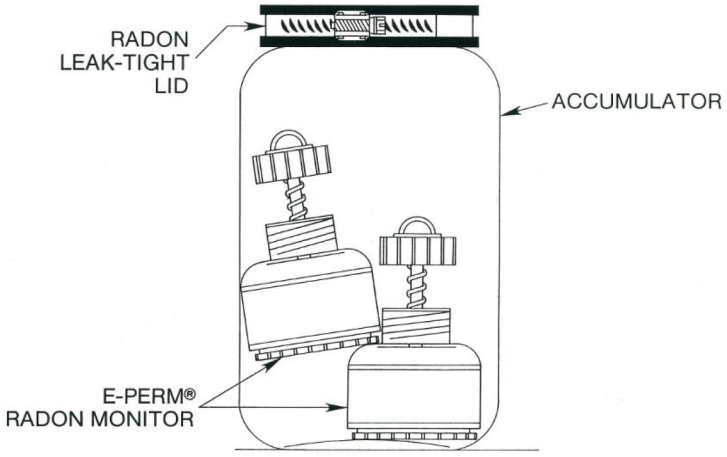

Figure (1): Standard accumulator used for calibrating EIC using NIST sources and other applications

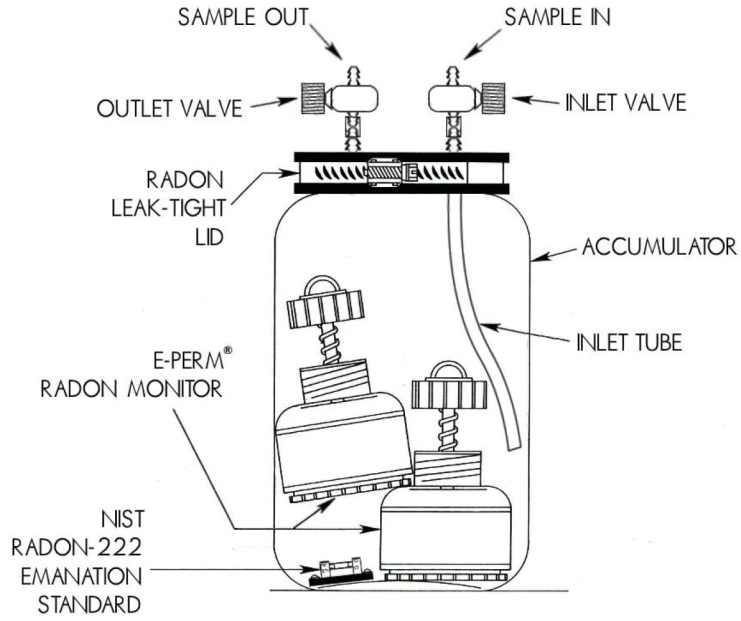

Figure (2): Accumulator system used for sampling natural gas with radon sources (NIST) standards or laboratory sources; for measuring radon in a sample of natural gas the standard source is not used

For some measurements the strength of these sources was not sufficient. Three additional radon sources were built using dry powder of uranium mill tailings. About 30 grams of such powder were loaded into a small pillbox, and the open end of the box was covered with Tyvek® [3] membrane and sealed to the edges of the pillbox. Tyvek® is known to fully transmit the radon emanated from the uranium mill tailings powder located inside the pillbox (Stieff 2012).

Three such sources (designated as 3, 4, and 5) were built. These sources can be used in place of the NIST sources where comparative measurements are needed. Figure (2) shows how the NIST sources are loaded into the sampling jar and can be replaced with the sources built in the laboratory.

Sampling system for measuring radon in natural gas using EICs

Figure (2) gives the schematic of the general sampling system for radon in natural gas using EICs with or without radon sources. NIST and laboratory sources are used for the experimental determination of *f* factors. The system consists of three parts:

1. A flow-through glass jar with two valves which can be sealed or opened.
2. A set of two premeasured SST (or SLT) EICs. Make sure that the EICs are in the "on" position.
3. A radon source when needed.

The procedure is as follows:
Record the initial voltages of electrets in both EICs.
Make sure that the natural gas stream has a flow rate of about 20 LPM (this provides sufficient volume changes to fully displace original air with the sampling gas).
Close the valves.
Connect the inlet valve to the stream of natural gas.
Open the inlet valve.
Open the outlet valve to the atmosphere.
Check for the flow by feeling (and smelling) the flow.
Continue to flow the natural gas for about 2 minutes.
Close the inlet valve.
Leave the outlet valve open for 15 seconds, then close the outlet valve.

Now the natural gas is locked inside the jar at atmospheric pressure. Because normally the stream is under pressure, this procedure eliminates possible higher than the atmospheric pressure in the analysis. The sampling has ended.

After 1 to 3 days unscrew the rubber collar and remove it from the jar. Unscrew the jar top. Take the EICs out and measure the final voltages of both the electrets in EICs.
Use a standard procedure to calculate the average radon concentration in air using initial and final voltages and the analysis time (1 day or any other chosen delay time).

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The results provide duplicate measurements of the average radon concentration in natural gas during the chosen delay period. What is needed is the initial radon concentration at the time of collection. This can be calculated using the procedure given in next section.

Equation for calculating the initial radon concentration (IRC) from the average radon concentration (ARC) as measured by EIC in sealed container for D days

The IRC is higher than the ARC due to the decay of radon over the measurement period. These two are related by equations (1) - (3). The ARC as measured by an EIC in a sealed container for D days is simply the time integrated radon concentration divided by duration, D.

$$ARC = \frac{TIC}{D} = IRC \int_0^D \exp(-\lambda\, t)\, dt \quad (1)$$

where TIC is the time integrated concentration in pCi-days/liter and λ is the decay constant of radon in day $^{-1}$ = 0.1814 day $^{-1}$.

$$ARC = \frac{(IRC)(1-\exp(-\lambda D))}{(\lambda D)} \quad (2)$$

$$IRC = \frac{(ARC \times \lambda\, D)}{1-\exp(-\lambda\, D)} \quad (3)$$

Example: D = 3 days
ARC= 10 pCi/L
IRC = 12.97 pCi/L

Table (1) gives the calculated IRC values for different measurement periods and an ARC value of 10 pCi/L.

Sensitivity of the method

ΔV is the approximate voltage drop when the electret is used in an SST configuration. Sensitivity is defined as the radon concentration that gives a voltage drop of approximately 20 volts for the stated period, which corresponds to an error of roughly 10%. If a measurement period of 3 days is performed, the sensitivity will be around 3.3 pCi/L measurable with an error of 10%. For a measurement period of 1 day, the sensitivity is 10 pCi/L.

Table (1): Calculated IRC from ARC for different analysis periods and sensitivity analysis

Time Period (in Days)	Radon Decay Constant(day $^{-1}$)	ARC (pCi/L)	IRC (pCi/L)	Ratio	ΔV (for 10 pCi/L)	Sensitivity (pCi/L)
1	0.1814	10	10.93	1.093	20	10.0
2	0.1814	10	11.92	1.192	40	5.0
3	0.1814	10	12.97	1.297	60	3.3
4	0.1814	10	14.06	1.406	80	2.5
5	0.1814	10	15.21	1.521	100	2.0
6	0.1814	10	16.41	1.641	120	1.7
7	0.1814	10	17.66	1.766	140	1.4
8	0.1814	10	18.95	1.895	160	1.3
9	0.1814	10	20.29	2.029	180	1.1
10	0.1814	10	21.67	2.167	200	1.0

Experimental verification of leak tightness of the sampling system

The basic assumption made in the design of the sampling system is that the system is leak-tight over an extended period. This would allow the sampling system to analyze the samples at different measurement periods, as needed. Three samples were collected from the same source of natural gas and analyzed after 1, 4, and 8 days. From the measured average radon concentrations, initial radon concentrations were calculated by the method described above. The calculated results are given in Table (2). These results verify that the sampling system is radon leak-tight. Any measurement period between 1 and 8 days is acceptable.

Table (2): Initial radon concentrations from simultaneously sampled jars over time

Time Period (Days)	ARC (pCi/L)	Ratio	IRC (pCi/L)
1	25.5	1.093	27.8
4	21.2	1.406	29.8
8	13.8	1.895	26.2

Technical differences between scintillation cells and EICs

There are a number of differences between scintillation cells and EICs, which should be recognized throughout the study. They are delineated below.

Scintillation cells

The interior surface of the scintillation cell is coated with a layer of zinc sulfide, which serves as the scintillate. The gas to be measured flows through the inlet valve (and escapes via the outlet valve) for about two minutes, ensuring that sufficient air exchanges have occurred and completely displaced the original air in the cell. Immediately afterward, both the inlet and outlet valves are closed and the sampling is complete. After waiting four or more hours (in order to allow the radon decay products to attain equilibrium with the parent radon), an alpha count rate is measured and the resulting radon concentration is calculated. A correction is applied for the delay time between sampling and the beginning of the measurement when calculating the radon concentration. If the density of the measured gas is smaller than that of air, which is the case for

natural gas (as it is rich in methane), more alpha particles strike the walls of the scintillation cell. This obviously increases the response of the cell for radon in natural gas relative to air. Likewise, if the density of the measured gas is larger than that of air (such as with carbon dioxide or propane), less alpha particles strike the walls of the scintillation cell. This decreases the response of the cell for radon in those gases relative to air. Correlating this property with changes in elevation also produces a similar but necessary correction factor. As gas density decreases at higher elevations more alpha particles strike the walls of the scintillation chamber relative to the same gas at sea level, necessitating a correction. This is portrayed below by Table (3). Density appears to be the only significant factor influencing the responses of scintillation cells.

Because the effective densities in natural gas can vary significantly from one source to another (due to its amalgam of various gases), experimentation is the only proper way to arrive at correct radon concentrations. Dr. Kitto has performed repeated measurements with scintillation cells, and has determined a factor of 1.07 as the over-response for scintillation cells for radon in natural gas relative to air, when measured at atmospheric pressure and room temperature.

Table (3): Specific gravities (taking density of air as 1.000) of gases

Name of Gas	Specific Gravity
Air at Sea Level	1.000
Methane	0.554
Natural Gas	0.60 to 0.70
Propane	1.522
Air at 500m Elevation	0.942
Air at 1000m Elevation	0.888
Air at 1500m Elevation	0.835
Air at 2000m Elevation	0.785
Table sourced from www.engineeringtoolbox.com	

Electret Ion Chambers

Electret Ion Chambers are quite different from scintillation cells, as shown in both their composition and methodology when measuring radon concentrations. Electret Ion Chambers measure the ion concentration within the gas; whereas, scintillation cells count the alpha particles reaching the zinc sulfide scintillate. As such, higher densities increase the response of radon in EICs, and lower densities decrease radon's response. This is inversely related to the response for scintillation cells. Also, SST EICs do not show significant effects of density differences (due to elevation) up to 4,000 feet (Kotrappa 1992). As natural gas is an amalgam of several gases as shown in Table (4), which are not in precise ratios to one another, this is only an approximated effect of the *W* value. As a note, it is worth defining the typical composition of natural gas, from Baltimore Gas and Electric Corporation given in Table (4).

Table (4): Composition of natural gas, reported from Baltimore Gas and Electric Corporation

Gas	Composition
Methane	93.32%
Ethane	4.65%
Propane	0.84%
Butane	0.18%
Nitrogen	1.01%

Due to the presence of so many factors, experimentation must be used to determine the actual response of electret ion chambers for natural gas and propane. This has been the goal of the present work. Another factor present in EICs (but absent from scintillation cells) is the ionization potential, which is defined by the *W* value (the energy in electron volts required to produce an ion pair). If the *W* value of the gas is lower than that of air, more ions are produced from the same alpha energy. This leads to an over-response of EICs, relative to the response in air, and is described in detail by Table (5). EICs are expected to give an over-response of approximately 1.15 due to the change in *W* value. Table (6) gives a summary of the distinguishing features between EICs and scintillation cells.

Table (5): *W* values for methane

Theoretical E (MeV)	From Table M/Air	*W* Value (methane relative to air)	Calculated Response in Methane
1.547	0.907	0.905	1.106
1.923	0.901	0.900	1.111
2.453	0.889	0.894	1.119
3.944*	0.879	0.877	1.141
4		0.876	1.141
5		0.865	1.157
6		0.853	1.172
7		0.842	1.188
8		0.830	1.205
Average			**1.150**

*Calculated *W* value is by extrapolation using fitted equation for the first four energies.
Gad Shani, Book, Radiation Dosimetry, Instrumentation and methods, CRC Press, Inc 1991, Boca Raton, Florida 33431. From Table 6 (RDIS) Page 25, Page 194 for correction from TE to air.

Table (6): Summary of differences in response between scintillation cells and EICs

Parameter	Scintillation cells	EICs
Increasing density or pressure	Decreases response	Increases response
Decreasing density or pressure	Increases response	Decreases response
Increasing *W* value	No effect reported	Decreases response
Decreasing *W* value	No effect reported	Increases response

Experimental methods for measurement of *f* for scintillation cells

The *f* factor is defined as the ratio of responses of radon in natural gas (using calibration constants for air) to that in air (using calibration factors in air).

Dr. Kitto's method (2014) of measuring *f* factor for scintillation cells is illustrated below.

Step 1. Radon-free air is bubbled through a NIST radium standard solution at a known flow rate and a sample is taken by scintillation cells and analyzed for radon concentration (designated as RnA).

Step 2. Radon-free natural gas is bubbled through NIST radium standard solution and a sample is taken by scintillation cells and analyzed for radon concentration (designated as RnG).

Step 3. All other conditions being the same, the *f* factor is calculated by taking the ratio between RnG and RnA.

Kitto concluded that the experimentally measured *f* factor is 1.07. This agrees well with the ratio of density of air to that of natural gas at atmospheric pressures and room temperatures.

Experimental method for measurement of *f* for electret ion chambers (EICs)

The principle used is similar to that used by Kitto, except for one noteworthy distinction: a radon source is used inside the sampling jar instead of bubbling the air through a radium solution. The radon source is simply a small pillbox containing about 30 grams of powdered uranium mill tailings. The top opening is covered with a Tyvek® sheet. The Tyvek sheet is sealed to the outside walls of the pillbox. This prevents the powdered uranium tailings from falling out. Radium-226 in the uranium mill tailings releases radon through the Tyvek® sheet, which is transparent to radon gas. This provides a continuous source of radon at a constant rate.

Step 1. Such a source is lowered into the sampling system (Figure 2). The valves are closed. The radon emanated from the source continues to accumulate inside the jar for a known length, such as three days. At the end of the three days the valves are opened. The sampling jar is also opened. The EICs are taken out and measured to calculate the average concentration of radon in the jar (designated as RnA).

Step 2. Leave the sampling system open for a day before starting this step. Repeat Step 1 with no radon source. The results are the background radon concentration in air (designated as RnABG).

Step 3. After venting the sampling system by keeping the jar open for one day, lower the same source and a new set of premeasured EICs. Collect the sample of natural gas using the protocol described in the above section on sampling the natural gas for measuring radon and collect the sample. Close the valves. At the end of 3 days the valves are opened. The sampling jar is also opened. The EICs are taken out and measured to calculate the average concentration of radon in the jar (designated as RnG).

Step 4. Repeat Step 3 with no source. The results are the background radon concentration in natural gas (designated as RnGBG).

With these measured radon concentrations, *f* is calculated using the following equation:

$$f = (\text{RnG-RnGBG}) / (\text{RnA-RnABG}) \quad (4)$$

Note that the natural gas used for EICs had a significant radon concentration, which has to be subtracted. On the other hand, Dr. Kitto used radon-free air and gas, so it is taken as negligible. The results of measuring *f* with EICs are listed in Table (7). The average is about 1.10.

Table (8) gives results of a similar experiment for the *f* value for radon in propane gas. Table (9) gives comparative results for the measurement of radon in natural gas using scintillation cells and using EICs, for samples collected on the same date. Note that calibration constants used are for air in both cases.

Table (7): Summary of 3-day experiments: radon in natural gas and radon in air

Grand Summary of 3-day experiments			Net
Source#	Net Radon in gas (pCi/L)	Net Radon in air (pCi/L)	Gas/air (*f*)
3	155	142	1.092
4	166	154	1.078
5	160	146	1.096
6 (NIST H)	76	69	1.101
6 (NIST H)	75	66	1.136
6 (NIST H)	75	67	1.119
3	155	143	1.084
3	153	141	1.085
		Average	1.099
		STDEV	0.020

Table (8): Summary of 3-day experiments: radon in propane gas and radon in air

Radon Source #	Net Radon concentration in propane gas (pCi/L)	Net Radon concentration in air (pCi/L)	Ratio of radon in propane to radon in air (pCi/L)
NIST-L	11.5	8.5	1.353
NIST-L	11.7	8.5	1.376
NIST-H	92.9	68.9	1.348
		Average	1.359

Table (9): Comparative results from scintillation cells and EIC, samples collected on the same date using calibration constants for air.

Collection Date	Sample Number	Scintillation cell (pCi/L)	EIC (pCi/L)
May 4, 2015	1	28.4 ± 2.2	28.1 ± 2.0
	2	27.4 ± 1.6	27.0 ± 1.9
	3	27.4 ± 1.7	26.9 ± 1.8
	4	n/a	26.5 ± 1.9
	Mean	**27.7**	**27.1**
June 16, 2015	1	27.0 ± 1.4	27.0 ± 1.9
	2	27.9 ± 1.5	26.1 ± 1.9
	3	27.3 ± 1.6	26.3 ± 1.8
	4	26.4 ± 1.8	27.2 ± 1.9
	Mean	**27.2**	**26.7**
June 29, 2015	1	28.8 ± 0.9	28.7 ± 2.0
	2	29.7 ± 1.0	31.9 ± 2.2
	3	28.4 ± 0.9	29.9 ± 2.1
	4	29.3 ± 0.9	28.5 ± 2.0
	Mean	**29.1**	**29.8**
To calculate corrected radon concentration, results of scintillation cells need to be divided by 1.07 for results obtained by scintillation cells and to be divided by 1.10 for results obtained by EICs.			

Results and Discussion

The *f* value for scintillation cells is 1.07 and for EICs is 1.10 as shown in Table (7). These are only slightly different from each other. Accuracies appear to be similar in both cases.

Table (8) gives the *f* value for measuring radon in propane gas. The measured *f* value is 1.38; whereas, it should have been 1.5 if due only to the difference in densities. Such differences can be accounted for the differences in W values and other unknown parameters. Normally propane gas used as a cooking gas does not contain radon because propane is produced by distillation of crude oil. This simply illustrates that the methodology used in this work can be used for arriving at *f* values for measuring radon in other gases, if required.

Table (9) gives comparative results from the measurement of radon in natural gas as measured by scintillation cells and as measured by EICs. Samples are taken at the same location and on the same date and time. Calibration constants used are for air at atmospheric pressure and at room temperature. The results are in good agreement between each other and accuracies are also similar. To be more accurate, results need to be divided by 1.07 for scintillation cell results and results of EICs need to be divided by 1.10.

Recently there was an inquiry whether EIC-based radon flux monitors (used for uranium exploration work) can be used at certain locations where radon is accompanied with natural gas from the ground. Based on the current work the authors can confidently say that the calibration constants for air can continue to be used in such situations, because a small concentration of natural gas in the sample will not significantly affect the measurement.

Scintillation cells have an advantage in that multiple measurements can be done on a single sample; whereas, the analysis can be done only once for each EIC collected sample. Multiple samples need to be collected if more than one measurement is required.

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