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Estimation of natural gas contribution in indoor ^{222}Rn concentration level in residential houses

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Abstract

The effect of natural gas use on indoor radon concentrations was studied in the dwelling of two cities in Cyprus using an AlphaGUARD radon detector. The radioactivity concentration of ^{222}Rn capsule natural gas samples were measured, and the contribution of natural gas consumption to generate indoor ^{222}Rn in residential houses was calculated. The average values of the indoor ^{222}Rn concentrations were found between 42.16 and 146.47 Bq m^{-3} in CNG and LPG samples, respectively. The obtained annual effective dose and excess lifetime cancer risk (ELCR) have been compared with the world average values specified by the WHO and UNSCEAR reports.

Keywords Radon in natural gas · AlphaGUARD · Dwelling · Annual effective dose · Lifetime cancer risk (ELCR)

Introduction

Natural radiation can be found in the human environment in a variety of locations. Radon-222 (^{222}Rn) is a radioactive gas produced by the decay of uranium-238 (^{238}U) series and daughter of radium-226 (^{226}Ra) with a short half-life (3.8 days) which is a radioactive colourless inert gas [1]. It has a high solubility in organic compounds, with a solubility value of 12.7 in toluene scintillation at 20 °C [2]. Natural gas is now a common energy source because of its low carbon dioxide and nitrogen oxide emissions [3].

On the other hand, liquid petroleum gas (LPG) and natural gas are significant radon sources [4]. Natural gas is typically supplied from various wells and fields, and as a result, the proportions provided from multiple sources will vary over time [5, 6].

Table 1 shows the radiation dose sources contribution from natural radioactive substances. As seen in Table 1, the main dose contribution is due to Rn gas. Radon and its decay components are the most significant contributors to radiation dose. Radon is a significant contributor to the general population's ionising radiation dosage and lung cancer [7]. Recent research in Europe, North America, and Asia on indoor radon and lung cancer shows that radon can cause many lung cancer cases in the general population [8]. Many variables affect indoor radon concentrations, including construction materials, indoor-outdoor temperatures, relative humidity, air turbulence, air flow and ventilation volume, and geological formations. The other pathway to ^{222}Rn entrance in residential houses is natural gas using for cooking and heating.

International and national organisations such as the USEPA, WHO, ICRP, and HPA set indoor radon concentration limits worldwide. The USEPA suggests radon levels of less than 148 Bq m^{-3} in the home and 400 Bq m^{-3} in the workplace [9].

To limit the health risks associated with indoor radon radiation, the WHO suggests a reference standard of 100 Bq m^{-3}. However, the preferred reference standard does not exceed 300 Bq m^{-3} if this level cannot be met under the current country-specific conditions. The European Union (EU) has agreed to the ICRP-65's proposed action rate of 500–1500 Bq m^{-3} [10]. The Action Level should be 200 Bq m^{-3}, and the Target Level should be 100 Bq m^{-3}, according to the HPA, expressed as the annual average radon concentration in the household. The Health and Safety Executive (HSE) in the United Kingdom has set a radon action standard for workplaces of 400 Bq m^{-3}.

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Table 1 The radiation dose sources contribution [3]

Source	^{222}Rn gas	Artificial	Gamma-ray	Food and drink	Cosmic ray
Dose contribution (%)	50	14.5	14	11.5	10

Some of the studies have also presented the ^{222}Rn concentration and radiological risk in building materials [11–13] and in water [14–16], in soil and rock [17], in the air [18].

This study's objectives were to apply a new design and method to evaluate ^{222}Rn radioactivity concentration in piping and capsule natural gas to residential houses and calculate natural gas contribution to indoor ^{222}Rn concentration. Also, the evaluation of radiological hazrads due to ^{222}Rn in indoor environment was calculated.

Materials and methods

A total of 5 natural gas samples, three compressed natural gas (CNG) and two liquefied petroleum gas (LPG) were selected to perform this experiment in Nicosia and Kyrenia, Cyprus.

To measure ^{222}Rn gas, an AlphaGUARD model PQ 2000 pro (SAPHYM Co) was used. The passage of ^{222}Rn gas from a filter to the ionisation chamber is the basis of this device's operation. This radon monitor is suitable for the continuous monitoring of radon concentrations between 2 and 2,000,000 Bq m^{-3}. It is both suited for short or long-term examination inside (e.g. in buildings) and outdoor and is capable of flow mode in a 1- or 10-min measuring cycle. Radon measurements were taken in a specially designed cubic chamber (70 × 50 × 60 cm) with sealed walls. An AlphaGUARD monitor was placed in the cubic chamber. The cubic chamber was connected to the vacuum pump to create a negative pressure (approximately 0.1 bar) inside the chamber. The natural gas flow was sent to the chamber by controlling pressure and temperature gauge. Due to the detector's sensitivity to hydrocarbon materials, measurements were performed with relatively low gas pressure. In this study, the flow mode with 10 min measuring cycle was used for at least one hour. The volume of natural gas regarding the most common method that standard cubic metre is a quantity of gas, which at the pressure of 1013.25 mbar and temperature of 15 °C has a volume of 1 m^3 [19]. The background of ^{222}Rn concentration value at pressure 0.1 bar was measured and subtracted from all samples. The calibration of AlphaGUARD was carried out using a standard solution of ^{226}Ra, code 71L07 was supplied from Amersham and diluted to activity concentration of 24.1(12) $\times 10^{-2}$ Bq mL^{-1}. The ^{226}Ra solution was in polyethylene and crammed into a leak-proof glass bulb, where ^{222}Rn accumulated over time. In SAPHYM Co Frankfurt radon chamber [20], AlphaGUARD was factory calibrated. The NIST ^{226}Ra SRM-4968/CP-100 principal standard source was used to calibrate a factory reference unit AlphaGUARD in the factory laboratory. This unit was used as an operational transfer standard device. The calibration of the AlphaGUARD used in this work was performed at the factory against the transfer device.

The schematic diagram in Fig. 1 shows the radon concentration measured by the active setup method. The gas samples were measured after one h in (n = 4–5) times to get average results. By Alpha View/Expert software, the final activity of ^{222}Rn gas (A_{Rn}) with decay correction was computed. However, the radon portion releasing from natural gas can be calculated with the following formula.

$$C_{Rn-Gas}\left(\mathrm{Bq\ m^{-3}}\right) = A_{Rn} \times [\alpha R_C + \beta R_H] \tag{1}$$

and

$$C_{Cooking} = \alpha R_C\left(C_T\right) \tag{2}$$

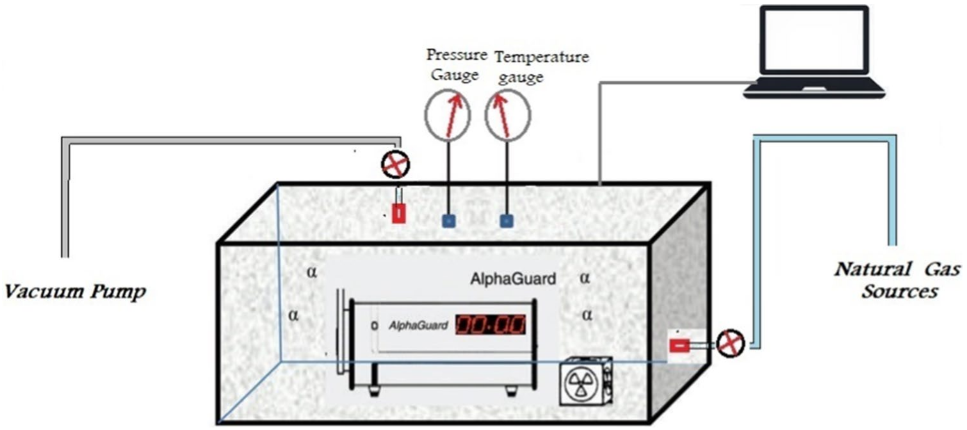

Fig. 1 Schematic diagram showing the radon concentration measurements of natural gas samples by an active setup method

$$C_{Heating} = \beta R_H \left(C_T \right) \tag{3}$$

where C_{Rn-Gas} is radon released from natural gas consumption $\left(\mathrm{Bq\ m^{-3}}\right)$; A_{Rn} is radon activity concentration mixed with natural gas (Bq m^{-3}); $C_{cooking}$ is natural gas consumption for cooking (m^3 year^{-1}); $C_{Heating}$is natural gas consumption for heating (m^{3}year^{-1}); $C_T$is natural gas consumption per year; R_C and R_H are radon releasing factor from cooking and heating, respectively. The parameters α and $\beta$are the natural gas consumption coefficient for cooking and heating, respectively.

Figure 2 shows the value of those parameters for ^{222}Rn concentration calculation from natural gas consumption. These parameter values were selected according to Energy Resource Guide, Turkey. At the same time, these parameters can be defaulted by local conditions and consumption scale.

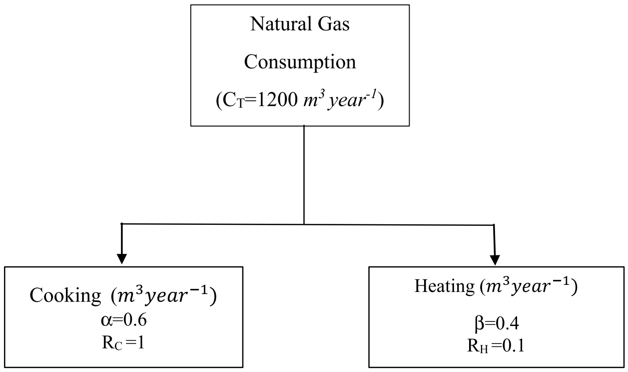

Fig. 2 The flow chart and default parameters to determine ^{222}Rn concentration in natural gas

The average annual effective dose received due to radon gas and its daughters (mSv year^{-1}) is derived from Eq. (4) [10, 21, 22]:

$$D\left(\mathrm{mSv\ year^{-1}}\right) = 5.56 \times 10^{-6} \times C_F \times E_F \times C_{Rn-Gas} \times T \times Q_F \tag{4}$$

where D is annual effective dose (mSv year^{-1}); C_F is dose conversion factor; for members of the public is 1.1 m Sv. (mJ h m^{-3})$^{-1}$ and 1 Bq m^{-3} = 5.56 × 10^{-6} mJ m^{-3}; E_F is equilibrium factor for radon progeny; 0.4; A_{Rn} is radon concentration (Bq m^{-3}); T is time in hours for a year; (365 × 24 = 8760 h); and Q_F is coefficient of occupancy; 0.8.

Excess lifetime cancer risk (ELCR) is the chance of a person contracting cancer if they are subjected to a given dose of radiation during their lifetime. By multiplying the parameters of annual effective dose D $\left(\mathrm{mSv\ year^{-1}}\right)$, the average period of life (DL = 70 years), and risk factor (RF = 0.05 Sv^{-1}), the risk of life- cancer was estimated by the following equation [20]:

$$ELCR = D \times RF \times DL \tag{5}$$

Results

In this research, a new method for collecting and analysing radon gas from a natural gas source was introduced. The ^{222}Rn concentration in natural gas samples with the system's pressure and temperature are presented in Table 2. The error of calculation was considered within one standard deviation. The ^{222}Rn concentrations in CNG gases were found to be in the range of 42.16 ± 3.11 to 94.55 ± 6.24 Bq m^{-3} with an average value of 74.54 ± 5.58 Bq m^{-3}. The ^{222}Rn

Table 2 The ^{222}Rn measured concentration (A_{Rn}), ^{222}Rn partial released to home (C_{Rn-Gas}), annual effective dose rate (D) and excess lifetime cancer excess risk (ELCR) from natural gas samples

Sample code	Pressure (mbar)	Temperature (°C)	^{222}Rn concentration in gas A_{Rn} (Bq m^{-3})**	C_{Rn-Gas} released to home (Bq m^{-3})	D (mSv year^{-1})	ELCR (10^{-4})
CNG-1 (n = 4)* Nicosia	1015	15	42.16 ± 3.11	26.88 ± 2.54	0.46	0.16
CNG-2 (n = 4) Nicosia	1014	17	94.55 ± 6.24	60.51 ± 5.70	1.04	0.36
CNG-3 (n = 5) Kyrenia	1020	16	86.91 ± 6.02	55.62 ± 3.91	0.95	0.33
Avg.			74.54 ± 5.58	47.67 ± 4.06	0.82	0.29
LPG-1 (n = 4) Nicosia	1020	20	146.47 ± 12.48	93.74 ± 8.41	1.61	0.56
LPG-2 (n = 5) Kyrenia	1018	18	108.71 ± 9.27	69.57 ± 6.08	1.19	0.42
Avg.	–	–	127.59 ± 11.51	81.65 ± 7.25	1.40	0.49

*The number of measurements

**Uncertainties are given within 1 standard deviation concentrations in LPG gases were found to be in the range of 108.71 ± 9.27−146.47 ± 12.48 Bq m^{-3} with an overall average value of 127.59 ± 11.51 Bq m^{-3}.

^{222}Rn released to home (C_{Rn-Gas}), average annual effective dose (D), and excess lifetime cancer risk (ELCR) values are presented in Table 2. Minimum C_{Rn-Gas} concentration value 26.88±2.54 Bq m^{-3} was observed in CNG-1 sample, and maximum C_{Rn-Gas} concentration value 93.74 ± 8.41 Bq m^{-3} was observed in LPG-1 sample. The average concentration value of C_{Rn-Gas} (64.26 ± 5.54 Bq m^{-3}) was calculated.

The minimum and maximum annual effective dose (D) value were found to be 0.46 mSv $year^{-1}$ in CNG-1 and 1.61 mSv $year^{-1}$ in LPG-1, respectively. The average annual effective dose value was calculated as 1.11 mSv $year^{-1}$ (Table 2). Minimum excess lifetime cancer risk (ELCR) value (0.000016) was recorded in CNG-1, whilst maximum ELCR value (0.000056) was measured in LPG-1. The average ELCR value was calculated as 0.000039 (Table 2).

Discussion

In view of the different radon concentrations in natural gas, these changes depend on parameters such as the natural gas extraction area. ^{222}Rn is decay product of ^{226}Ra element which generating from ^{238}U elements. ^{238}U is present in soil, rocks and Earth's bedrock. Whereas the natural gas exists in bedrocks and due to high pressure and temperature the radon mobility from rocks to natural gas occurs. Since the concentration of ^{238}U in bedrocks varies from place to place on earth. So, the natural gas used in houses has different content of radon gas.

Natural gas ^{222}Rn concentrations and annual effective dose calculations were carried out in 5 dwelling houses. It is observed that the average of ^{222}Rn concentrations in LPG gas samples were higher than the CNG gas samples.

The results of the ^{222}Rn concentration in the natural gas of the study are compared with the values of other areas reported in literature. Although ^{222}Rn concentrations range of this study was less than the ^{222}Rn concentrations value reported in Texas, Panhandle (370–1924 Bq m^{-3}), in Colorado (407–1665 Bq m^{-3}), USA [23]; Netherlands [24] (150,000–380,000 Bq m^{-3}); and Algeria [25] (1085–5800 Bq m^{-3}). Whereas the results of this study were comparable with ^{222}Rn concentrations in China Beijing (49–88 Bq m^{-3}) [26]. The results of this study were within the range reported from Thailand (16–197 Bq m^{-3}) [27] and Poland (30–1400 Bq m^{-3}) [28].

Figure 3 shows a boxplot of ^{222}Rn concentration with WHO reference level of ^{222}Rn concentration in dwelling houses. As seen in this Figure, the ^{222}Rn concentration in all CNG samples is less than the reference level, while the ^{222}Rn concentration in all LPG samples is more than the reference level.

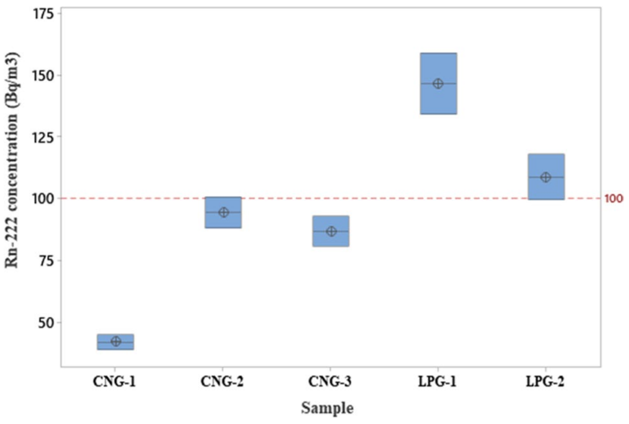

Fig. 3 The boxplot and WHO reference level for indoor ^{222}Rn concentration

The higher values than reference may be attributed to U and Th content in geological media and soil porosity. Also, radon emanation coefficient can be the factor that affected the radon concentrations in LPG gas.

It was observed that the annual effective dose due to natural gas in study area was lower than the ICRP 2010 reference range of 3 to 10 mSv $year^{-1}$ [28] and the WHO suggested reference limit of 10 mSv $year^{-1}$ (WHO 2009) [29] (Fig. 4).

The risk factor of developing cancer increases by 0.004%, 0.04%, 0.4%, and 4% with the increment of ELCR values corresponding to the effective dose of 1, 10, 100, and 1000 mSv $year^{-1}$, respectively [30]. The excess lifetime cancer risk (ELCR) for radon exposure ranged from 1.6×10^{-5} to 5.6×10^{-5} with an average of 3.7×10^{-5} (Fig. 5), whereas those values are lower than the world's average (2.9×10^{-4}) [31].

Conclusions

In this study, the activity concentrations of ^{222}Rn in natural gas samples have been measured by a direct method. ^{222}Rn concentration in LPG gas samples was higher than WHO reference level, whilst ^{222}Rn concentration in CNG gas samples was lower than WHO reference level. Also, the indoor ^{222}Rn contribution from natural gas regarding cooking and heating consumption was calculated. The annual effective dose (D) and the excess lifetime cancer risk (ELCR) for radon exposure from natural gas pathways was lower than the world's average reported by UNSCEAR, 2000 reference range limit. This study shows the importance of using a suitable filter to reduce the amount of ^{222}Rn in the natural gas purification process, and the authors of this study recommend it.

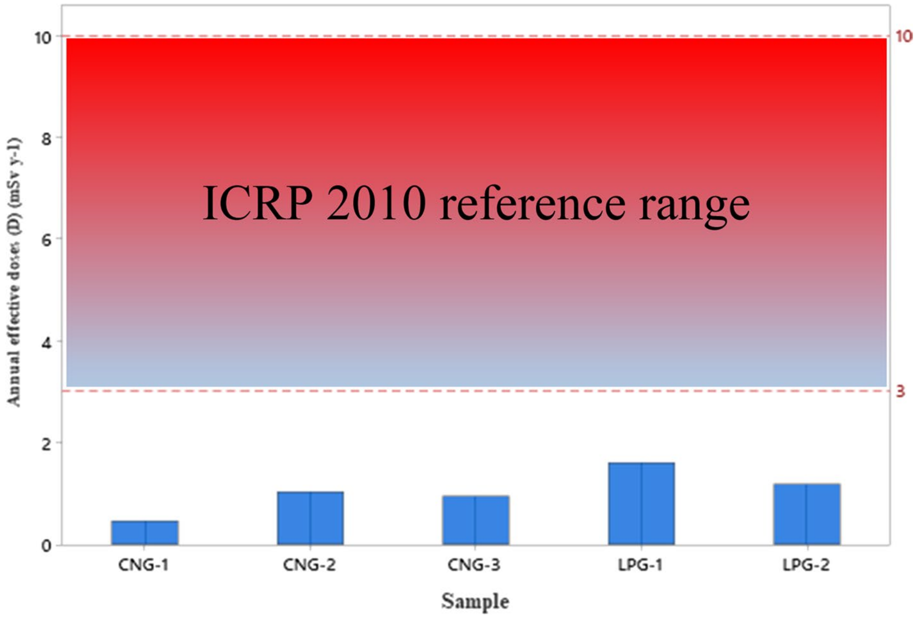

Fig. 4 The chart of annual effective doses (D) with ICRP 2010 reference range limit

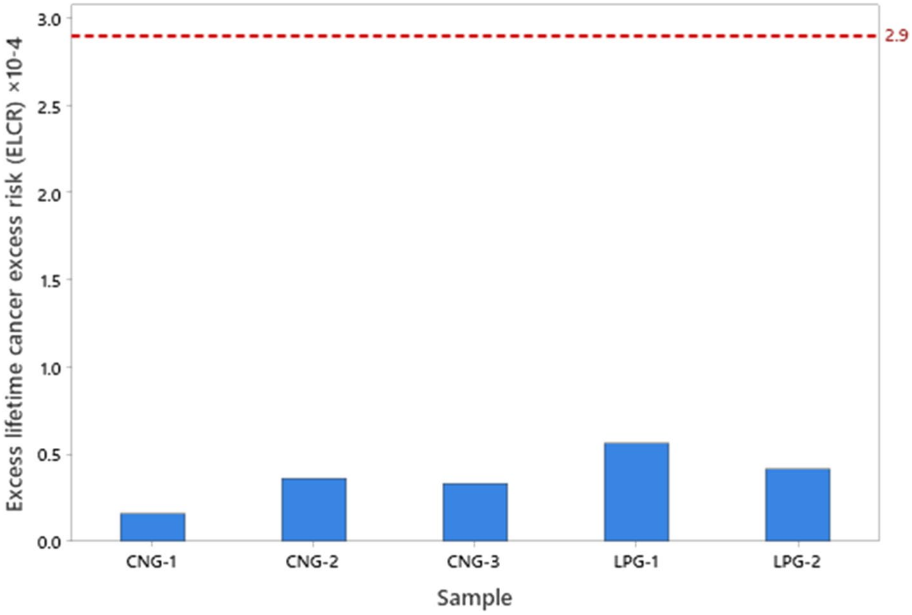

Fig. 5 The chart of Excess lifetime cancer excess risk (ELCR) from natural gas samples with UNSCEAR, 2000 reference range limit

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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