

Measurement of radon concentration and assessment of associated cancer risk in some fertilizer warehouses in the Punjab province of Pakistan

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Received: 4 August 2017 / Published online: 10 November 2017
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Abstract Radon concentration was measured by using ³⁹CR track etched detectors in five fertilizer warehouses in the Punjab province of Pakistan. The average concentration of radon was determined to be 20–88 Bq m⁻³. Annual effective dose (AED) and excess lifetime cancer risk (ELCR) have also been assessed using the models of UNSCEAR and ICRP. The corresponding annual dose and ELCR to the workers of warehouses have been estimated to be 0.30–1.02 mSv year⁻¹ and 0.44–1.02%, respectively.

Keywords Fertilizer warehouse · Track etched detector · Radon concentration, Effective dose · Cancer risk

Introduction

The present estimated population of Pakistan is about 0.2 billion, which is growing at the rate of almost 2.4% annually [1]. Punjab is the largest province of Pakistan where around 69% population lives in rural areas and depends mainly on the production of agricultural crop. Agriculture in Pakistan accounts for about 24% of the GDP (growth domestic product), and about 50% of the labour

force and contributes about 60% to export earnings [2]. Average fertilizer consumption in India, Pakistan and Bangladesh is forecasted to grow annually at the rate of 4% over the period, with corresponding increases for nitrogen (3.3%), phosphate (4.9%) and potash (5.9%) compared with average worldwide consumption rate of 2.6, 2.8 and 2.7% respectively [3].

The world phosphate fertilizer demand increased from 41.7 to 42.7 million tonnes in 2013–2014, at a growth rate of 2.4%. It is expected to touch 46.6 million tonnes in 2018 at a growth rate of 2.2% per year. Among the Asian countries, about 3% of the growth in world demand of phosphate is expected in Pakistan up to 2018 [4].

Phosphate rock (PR) is the raw material used worldwide for manufacturing various brands of chemical fertilizers [5]. Phosphorus forms chemical complexes with uranium and the concentration of ²³⁸U and its decay products in phosphate deposits is about 1500 Bq kg⁻¹ [6, 7]. The manufacturing of most commercial phosphate fertilizer (PF) begins with the production of phosphoric acid, which can have dissolved high amounts of natural radioactive elements [8, 9]. TENORM (Technologically Enhanced Naturally Occurring Radioactive Materials) in PR contains radioactivity of uranium and its progeny [10]. Moreover it may be transferred to the PF made from these rocks [11].

The fertilizer samples like MAP (Monoammonium Phosphate) and DAP (Diammonium Phosphate) that were derived directly from phosphoric acid may contain significant amount of radioactivity concentrations. The high concentration of radionuclides in fertilizers may increase the radon concentration in the surrounding air causing great risk to working personnel in the warehouses or fertilizer storage areas [5].

Radon gas emanates from the TENORM in PF and accumulates in PF storage places. The decay of radon gas

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and its progeny is characterized by alpha-radiation and can induce risk of lung cancer engendered by inhalation of radon [12]. The inhalation of radon gas and its progeny may expose the lungs to alpha radiation that may damage the cells in the lining of the lungs and increase a person's risk of lung cancer. The risk of lung cancer is higher in those who have been working for many years in a radon-contaminated warehouses.

As the offtake of PF in the Punjab Province of Pakistan is large as compared with the other provinces [13]; therefore, radon survey has been carried out in various fertilizer warehouses of the Punjab province. Radon concentration in the respective warehouses was measured by passive method using ^{39}CR track etched detectors. The measured concentration was then used to estimate the effective annual dose and lung cancer risk for the exposed workers of these warehouses. All the studied parameters were then compared through Arc GIS mapping.

Materials and methods

Warehouses

Due to seasonality of fertilizer consumption, all the fertilizer industries of Pakistan have established a network of over 120 field warehouses to meet their storage requirements. Owing to time limitations and inaccessibility to most of the warehouses, the current study only limits to five warehouses of FFC (Fauji Fertilizer Corporation) located in the Punjab province. The concentration of radon was measured in those warehouses where different type of PFs were kept for some interim period. The detail of every fertilizer warehouse is given in Table 1, and is shown in Fig. 1 where every warehouse is indicated by an asterisk.

Radon concentration measurement

The passive method was applied for the measurement of radon in the understudy fertilizer warehouses of the Punjab, Pakistan. Track etched detection technique consisting of ^{39}CR solid state nuclear track detectors (SSNTD) was applied. A large sheet of ^{39}CR (polyallyl diglycol carbonate) having thickness of 500 μm (supplied by Page

Mouldings, Ltd, UK) were cut into small pieces of size 20×20 mm. Some of these detectors were kept in the refrigerator to determine background. The other detectors were pasted within the NRPB (National Radiation Protection Board) radon dosimeter holders. This dosimeter assembly is a diffusion cup that is designed by the National Radiological Protection Board now called the Radiation Protection Division of the Health Protection Agency [14]. The dosimeter body is made of polypropylene plastic and consists of two parts, namely, a circular base with a recess for detector and a domed circular upper portion with an internal circular base-retaining strut [15]. Radon gas diffuses into the dosimeter and exposes the ^{39}CR detector. It assures the discrimination of radon progeny (particulate daughters) by allowing only radon 222 gas to enter into the dosimeter [16].

Five sites were selected for the measurement of radon concentration in a warehouse. A total of 100 ^{39}CR -based radon gas dosimeters were mounted at a height of about 1.5 m in the indoor environments of the understudy warehouses for about 1 year. After every three months a batch of 25 detectors were removed from the five warehouses.

The exposed ^{39}CR detectors were etched in 25% NaOH at 80 $^{\circ}\text{C}$ for 16 h and the etched tracks were counted under an optical microscope of magnification 400. After applying the background correction, track densities were converted to radon concentration level using a calibration factor of 2.7 tracks $\text{cm}^{-2} \text{h}^{-1}$ per kBq m^{-3} [16, 17]. The representative concentration of radon in a warehouse was the average of all the concentrations measured at these sites. The measured radon concentration given in Table 2 is the presentation for every batch of the year. The values are compared in Fig. 2.

Radon effective dose

In order to investigate the annual mean effective dose H (mSv year^{-1}) due to radon to the persons working in the understudy warehouses, the concentration of radon was converted to effective dose by using the following relation [18]:

$$H = C \times F \times D \times T \times O \quad (1)$$

Table 1 The understudy warehouses

Location	Capacity (m^2)	Fertilizer storage
Gujranwala	2530	DAP (Diammonium phosphate), Urea, Boric acid
Gujrat	2670	DAP, Urea
Rawalpindi (Rawat)	2030	DAP, SSP (Single superphosphate), Urea
Sheikhupura	2340	DAP, Urea
Sialkot(Daska)	1800	DAP, Urea

Study Area

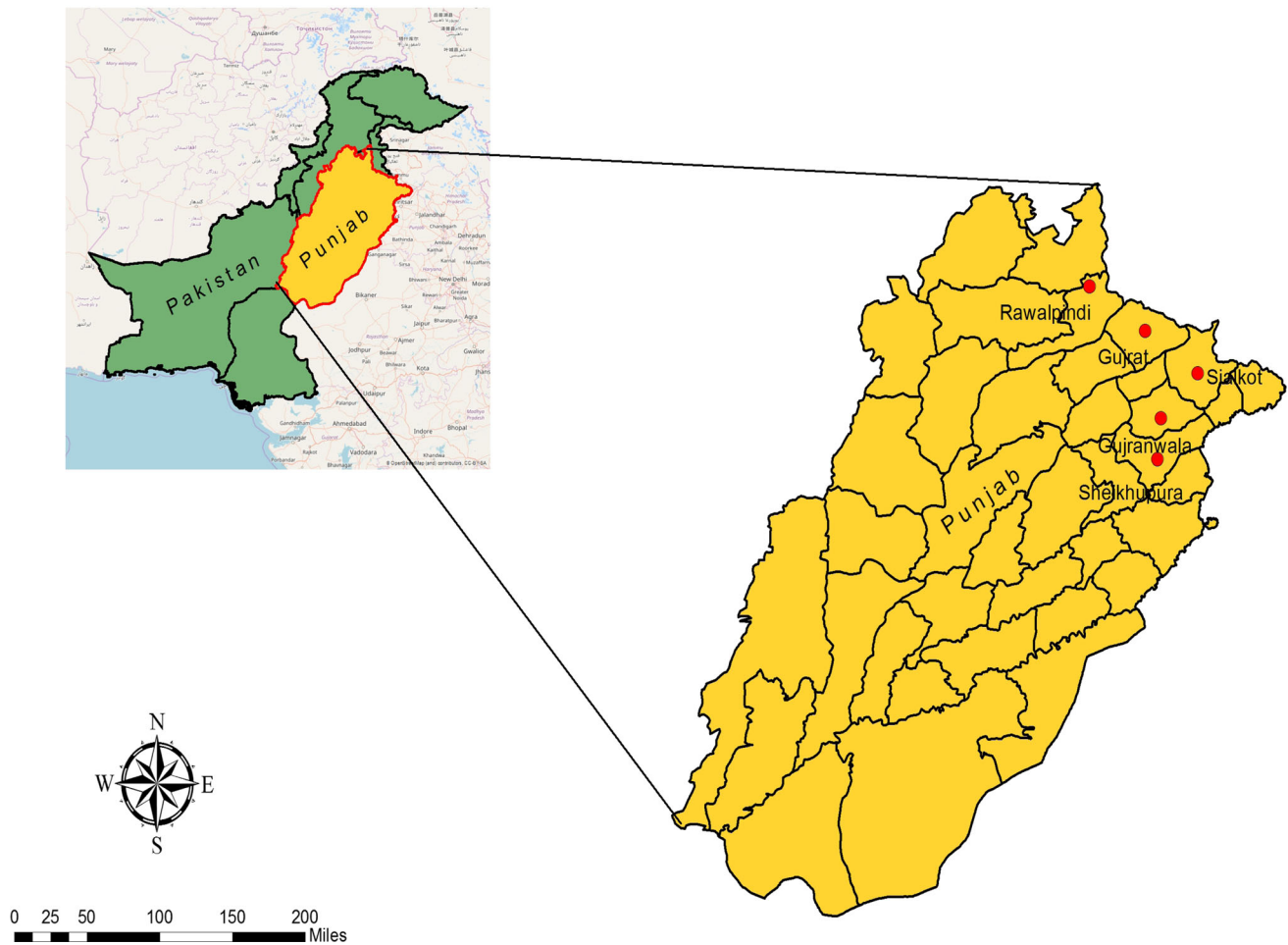


Fig. 1 The location of the understudy warehouses in the Punjab province

where C stands for radon concentration in $Bq\ m^{-3}$, F for equilibrium equivalent concentration (EEC) factor, D for dose conversion factor ($9 \times 10^{-6}\ mSv\ h^{-1}$ per $Bq\ m^{-3}$) based on the most recent recommendation of the International Commission on Radiological Protection [19], T for time ($8760\ h\ year^{-1}$), and O for occupancy factor. The value of F as mentioned in the UNSCEAR report is 0.4 for the indoor environment and O is taken to be 0.35, as the workers spend about 8–9 h per day in a fertilizer warehouse.

Lung cancer risk

The excess lifetime cancer risk (ELCR) due to mean radon concentration in fertilizer warehouses was determined using the following relation [20]:

$$ELCR = C \times O \times F \times T \times C_R \times T_L \tag{2}$$

where, C, O, F and T are the same as in Eq. (1); C_R is the nominal risk coefficient deterrment for radon exposure for the population of all ages is taken as 8×10^{-10} per $Bq\ h\ m^{-3}$ and T_L is the average life-time expectancy approximately equal to 65 year for the population of the Pakistan [21].

Results and discussion

Radon exposure

The concentration of radon measured in all the five understudy warehouses is presented in Table 2. The representative concentration of radon in a warehouse is the average of all the five concentration levels measured at these sites. The concentration of radon in all the studied areas varies slightly from place to place. The concentration measured in a specific period is also different from that

Table 2 Batch-wise radon concentration in phosphate fertilizer warehouses in five districts of Pakistan

Exposure locations	Exposure period	Concentration (Bq m ⁻³)	
		Mean value	Range
1st Batch			
Gujranwala	January–March	88 ± 3	(79–91)
Gujrat	January–March	56 ± 2	(44–61)
Rawalpindi (Rawat)	January–March	33 ± 2	(27–40)
Sheikhupura	January–March	49 ± 2	(38–51)
Sialkot (Daska)	January–March	66 ± 2	(58–72)
2nd Batch			
Gujranwala	January–June	58 ± 2	(50–63)
Gujrat	January–June	64 ± 2	(60–73)
Rawalpindi (Rawat)	January–June	25 ± 1	(21–33)
Sheikhupura	January–June	54 ± 3	(46–60)
Sialkot (Daska)	January–June	37 ± 2	(28–43)
3rd Batch			
Gujranwala	January–September	41 ± 2	(34–48)
Gujrat	January–September	20 ± 1	(16–27)
Rawalpindi (Rawat)	January–September	26 ± 1	(20–30)
Sheikhupura	January–September	35 ± 1	(30–41)
Sialkot (Daska)	January–September	26 ± 1	(21–30)
4th Batch			
Gujranwala	January–December	77 ± 3	(69–83)
Gujrat	January–December	22 ± 1	(15–29)
Rawalpindi (Rawat)	January–December	29 ± 1	(22.56–34)
Sheikhupura	January–December	30 ± 1	(24–34)
Sialkot (Daska)	January–December	38 ± 2	(31–45)

determined during other monitoring periods as clearly depicted in Fig. 2.

The concentration level of radon in the warehouse at Rawat varied throughout the year and is found to be lower than that in other understudy warehouses. This may be due to the fact that the warehouse at Rawat is quite large and properly ventilated as compared to those occurred at other places [22]. This warehouse is mostly filled with non-phosphate fertilizers, the PF occupied only a small fraction of the warehouse. Moreover, radon value increased slightly within January–March due the retention of more stock of DAP fertilizers in the warehouse during that period [5].

The value for radon concentration in the warehouse at Gujrat is relatively higher for period of January to June that is due to the storage of large quantity of DAP in the warehouse and because of poor ventilation. A sudden decrease of radon concentration is observed during July to December when the warehouse was almost empty of DAP. Also the contribution of radon is small from soil of district Gujrat due to its relatively lower radon exhalation rate [23].

The storage capacity of the warehouse at Daska in Sialkot district is relatively lower than that in other understudy warehouses and the ventilation rate is also low.

The concentration of radon varied from minimum in summer to maximum in winter which is in accordance with the facts mentioned in literature [15, 24].

The concentration of radon in the warehouse at Gujranwala is relatively higher than that in the other understudy warehouses. The concentration of radon in the warehouse at Sheikhupura is similar to that in the warehouses at Daska and Gujranwala.

The data on radon concentration in fertilizer warehouses is scarce in literature, which could only be found for Greece and Nigeria. Radon concentration in the other warehouses such as cement and decorative stones has been measured in Iran. The data on radon concentration in warehouses compiled from literature is presented in Table 3. The concentration of radon in fertilizer warehouses of Pakistan is lower than that in other warehouses in comparison, whereas it is exceptionally higher in fertilizer warehouses in Greece.

The acceptable level of radon recommended by US EPA is 148 Bq m⁻³ [29] which have been adopted as radon mitigation level by the OSHA (occupational safety and health association) [30]. The ICRP recommended maximum level of radon for occupational radiological

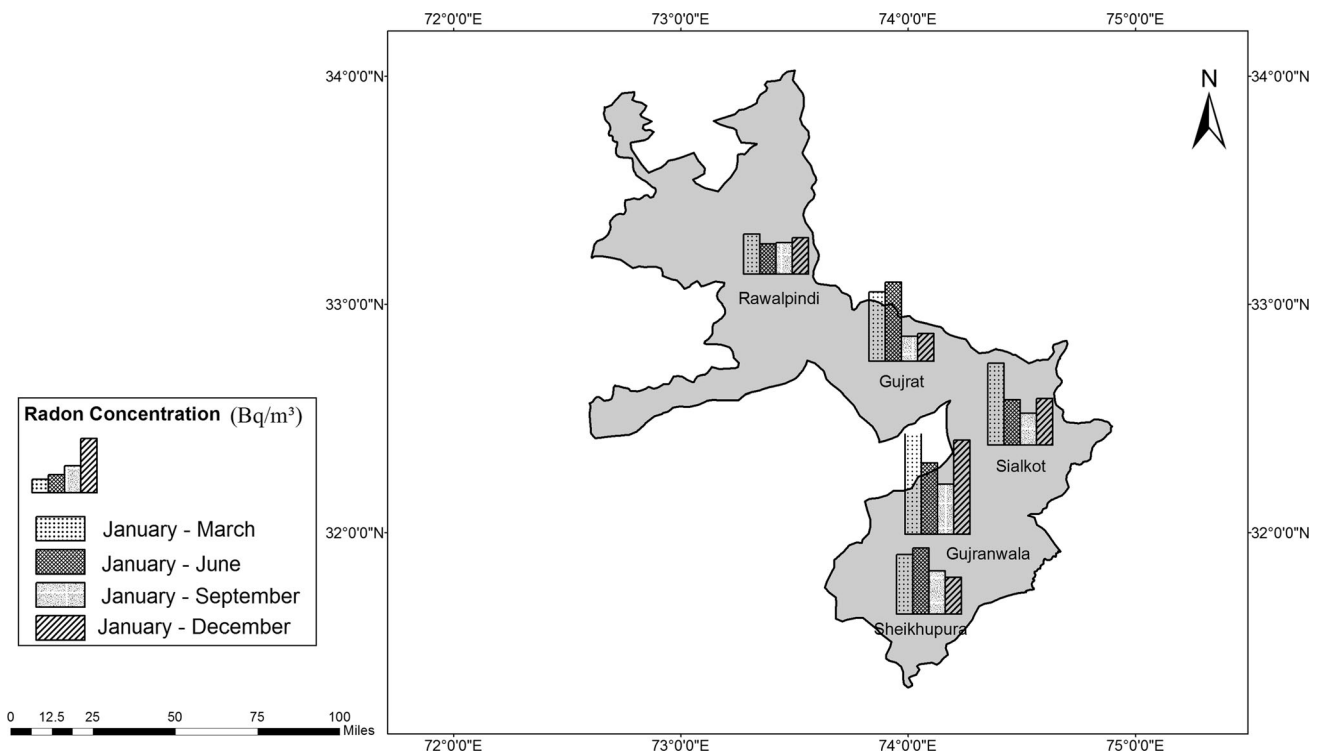


Fig. 2 Radon concentration in phosphate fertilizer warehouses in study area

Table 3 The radon concentration in warehouses of some countries of the world

Location	Type of warehouse	Radon concentration (Bq m ⁻³)	References
Greece	Fertilizer warehouse	(0.54–3.32) × 10 ³	[25]
Iran	Black cement warehouses	154	[26]
Iran	Decorative stones warehouses	73	[26]
Iran	Cement storage warehouses	133.6 ± 33	[27]
Nigeria	Fertilizer warehouse	37–117	[28]
Pakistan	Fertilizer warehouse	20–88	Present study

protection requirements is 1000 Bq m⁻³ [19]. The present measured radon concentration levels 20–88 Bq m⁻³ are lower than the EPA value and far below the ICRP recommended concentrations.

Annual effective dose and Cancer risk

Annual effective doses (AED) and excess life-time cancer risk (ELCR) due to radon inside the understudy warehouses were calculated using Eq. (1) and are presented in Table 4.

The AED values are compared in Fig. 3. The coloured circles in Fig. 3 are related to the AED values bigger is the circle greater are the effective dose associated to it.

The AED and ELCR have been estimated on the basis of annual radon concentration. Radon effective dose in the warehouses varies between 0.25 and 0.85 mSv year⁻¹ with an average value of 0.44 mSv year⁻¹, which is less than that of worldwide environmental radon dose of 1.15 mSv

Table 4 Annual effective dose and cancer risk due to radon in the understudy warehouses

Exposure location	Average radon concentration (Bq m ⁻³)	Effective dose (mSv year ⁻¹)	ELCR (%)
Gujranwala	77	0.85	1.02
Gujrat	22	0.25	0.30
Rawalpindi (Rawat)	33	0.36	0.44
Sheikhpura	30	0.33	0.40
Sialkot (Daska)	38	0.41	0.50

year⁻¹ given in UNSCEAR 2000 report [18]. Annual effective radon dose in the present study is much lower than the reference level of 10 mSv year⁻¹, set by the ICRP [19] in occupational regions.

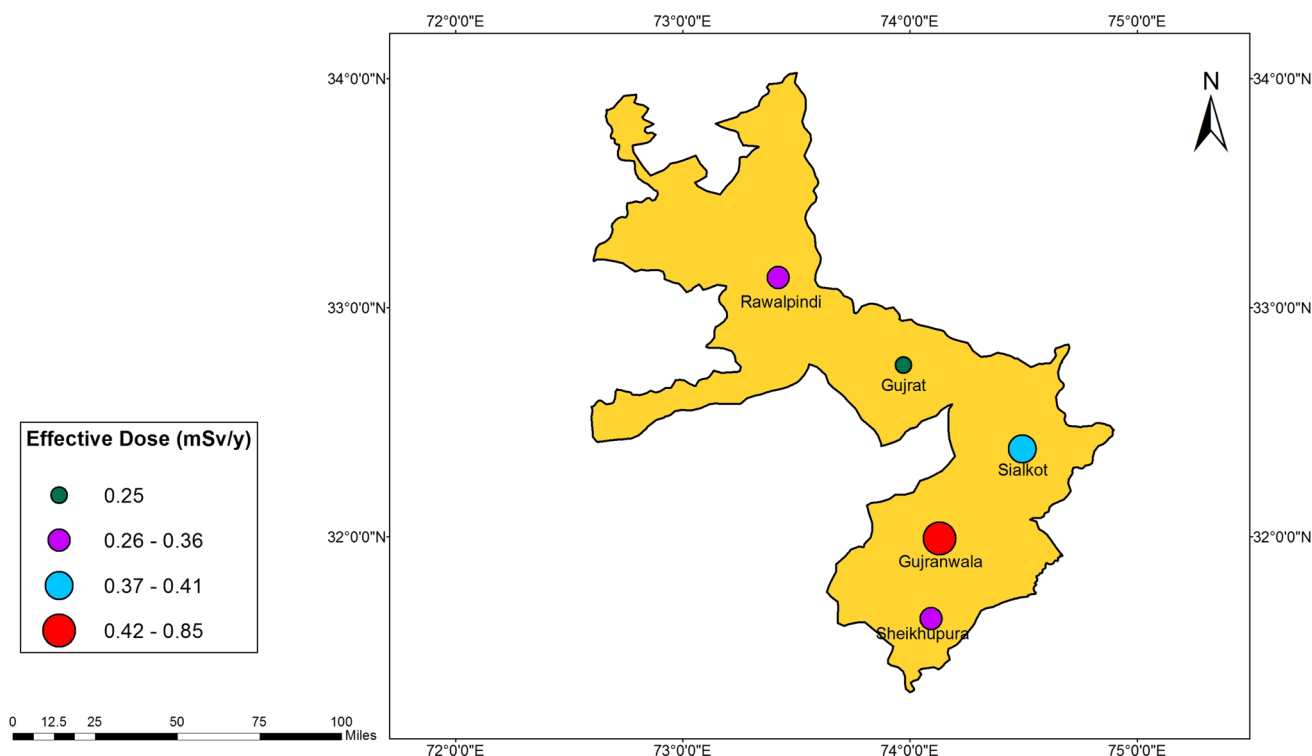


Fig. 3 Annual effective dose due to radon in the understudy warehouses

The corresponding ELCR due to radon inside the warehouse was calculated using Eq. (2) and also given in Table 4. The calculated ELCR 0.44–1.02% is less than the estimated risk of 2.3% from the lifetime exposure at 148 Bq m^{-3} , the action level given by US EPA [29] for entire population and the same approved by occupational safety and health association [30].

Conclusions

The mean radon concentration measured in the fertilizer warehouses was $20\text{--}88 \text{ Bq m}^{-3}$. Radon concentration remained variable in every quarter of the year, showing the seasonal impact. The other factors which influences the radon concentration in the present study may be temperature, ventilation system of the site, radon exhalation rate from the soil and buildings of the warehouses. Moreover, the presence of phosphate fertilizers in the study area may increase the radon concentration. Average annual radon concentration, effective dose and excess lifetime cancer risk in some warehouses of five districts of the Punjab province were found below the references level imposed by the ICRP, US EPA and OSHA. It means that all types of fertilizers stored in the warehouses do not have significant radon sources and negligible health hazard is expected to the workers of

these warehouses due to radon exposure in the existing situation.

Acknowledgements One of the authors, Sabiha-Javied, is grateful to Higher Education Commission (HEC) for its financial support through “Indigenous Scholarship Scheme for PhD studies in Science and Technology”.

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