Relationship between Radon Gas and Heavy Metals Concentrations in Soil of Selected Farms in Kirkuk City/NE Iraq

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Abstract - In this study, ten farms within Kirkuk city were selected to take soil samples from them at depth 20 cm to measure radon gas and heavy metals concentrations in order to find therelationship between them using SPSSprogram. The passive CR-39and active **RAD-7** techniques connected with irradiationcylinders were used to measure radon concentrations, and the ICP-MStechnique was used to measure heavy metals concentrations. Average radonconcentrations measured by CR-39 and RAD-7in airspace inside the irradiation cylinders were32.616 and 35.17 Bq/m³. Average radon concentrations measured by CR-39 in the samples was 3398.65 Bq/m³. Average uranium (U) concentrations was0.2653ppm and the other heavy metals average concentrations were 5.27As, 53.1Zn, 20.18 Cu, 10.16 Pb, 14.68 Co, 68.7 Cr, 112.28 Ni, and 0.22 Cdppm.It was found that there is no strong correlation between the concentrations of radon and heavy metals in the studied soil samples. Radon concentrations varied from one location to another independently on concentrations of heavy metals except foruranium metal which had a strong correlation to radon concentrations.

Keywords - Radon gas, Heavy metals, RAD-7, Kirkuk city.

I. INTRODUCTION

The rapid increase in human activities today in many sectors without proper planning and control lead to pollute the air, water, and soil by many substances which have big impacts on the entire environment. Pollution of the natural environment by heavy metals and radioactive substances is a global issue because they have toxic effects on the living organisms in different concentrations [1]. Heavy metals are inorganic pollutants that cannot degrade like organic pollutants and turn to harmless products but persist and accumulate in the soil [2]. Concentrations of heavy metals are not stable and vary in the environment according to many factors such as climate change, urbanization, industrial production etc. [3]. Exposure to these metals at high concentrations leads to accumulating them in the body causing serious health risks [4].

Human beings are exposed to radon every day as a result of the natural radioactive decay of uranium in the rocks and soil that exists everywhere, in outdoor air and buildings in varying concentrations [5]. Radonis a natural radioactive gas has a half-life of 3.85 daysthat are long enough to allow it to move through the soil and enter the atmosphere [6]. Exposure to high concentrations of radon and its daughters through inhalation for a long time lead to pathological effects such as respiratory function charges and lung cancer [7].

Radon is a part of uranium²³⁸U decay chain [8],which is considered a heavy metal and this chain finished with stable lead ²⁰⁶pb, which is also considered a heavy metal. This leads to know about the relationship between radiation and heavy metals. A high concentration of radon in a studied area is a sign of high concentration of uranium, lead and other heavy metals.

II. EXPERIMENTAL PROCEDURE

A) Study Area

The study area is located in Kirkuk city on both sides of the Khasa River northeast of Iraq as shown in fig. 1. Samples of soil were collected in ten selected farms at depth 20 cm in order to measure radon and heavy metals concentrations and find the relationship between them. The information about the selected farms is giving in Table I.

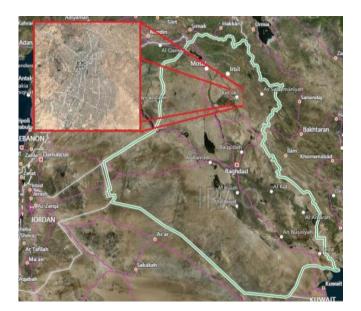


Fig.1. Location of the study area.

B) Preparation of Samples to Measure Heavy Metals Concentrations

Soil samples were dried at room temperature for three days, after that it was sieved by a sieve (180 microns) to obtain a homogeneous powder, then 0.5 g of each sample weighed by a sensitive balance and placed in small bags to be sent to Acme lab in Canada in order to measure heavy metals concentrations by using ICP-MS technique, which is a powerful technique for trace analysis of metals and it is preferred for ultratrace levels due to its higher sensitivity, selectivity, and detection limits is much lower than other multi-metal techniques[9].

C) Preparation of Samples to Measure Radon Concentrations

Radon concentrations were measured in soil samples at depth 20 cm by CR-39 as a passive techniquewhich is an organic nuclear detector, and RAD-7 as an active technique which is a true, real-time continuous radon monitor [10,11]. For this purpose irradiation cylinders (long cylinders) were designed, made by plasticwith dimensions (30 cm high and 7 cm diameter) with two connected valves, filled with soil samples to 3 cm height and the CR-39 detector was placed 25 cm away from the surface of the samples and closed tightly for two months to reach equilibrium radiation between radium and radon during this time period. After that they connected to RAD-7 by the valves to measure radon concentrations in the airspace inside the cylinders using grab protocol as shown in Fig. 2, after the measurements were finished the CR-39 detectors were etched in 6.25N (NaOH) at 70 °C for 3 h and studied under optical microscope at 40X magnification in order to see the effects of radon that's formed on the surface of the CR-39 detectors.



Fig. 2. Measuring radon concentration of soil samples in the Lab by RAD-7.

D) Radon and Uranium Concentrations Measurement

Radon concentrations in airspace inside the irradiation cylinders that contain soil samples can be calculated from equation below [12].

$$C_a = \rho / \eta T \tag{1}$$

Where ρ is the track density on the exposed detector CR-39 (Tr/cm²),T is the exposure time of the samples (days), η is the detection efficiency of the irradiation cylinder (0.66 Tr.cm⁻².d⁻¹/ Bq.m⁻³) [10].

To calculate radon concentrations in the soil samples, equation (2) can be used [13]

$$C_s = \lambda_{Rn} C_a H T/L$$

(2)

Where $C_{\rm s}$ is the radon concentration in the samples (Bq/m³), $C_{\rm a}$ is the radon concentration in airspace inside the irradiation cylinders (Bq/m³), $\lambda_{\rm Rn}$ is the decay constant of radon (0.1814 day⁻¹), H is the height of airspace in the irradiation cylinders (25 cm), L is the thickness of the sample in the irradiation cylinders (3 cm), T is the time of irradiation (days).

To calculate uranium concentrations in the soil samples in (ppm) unit the number of radon atoms must be calculated using equation below [14]

 $A_{Rn} = \lambda_{Rn} N_{Rn}$

From N_{Rn} one can determine the number of uranium atoms in the samples, using the equation of ideal radiation equilibrium [15]

(3)

 $\lambda_{\rm u} N_{\rm u} = \lambda_{\rm Rn} N_{\rm Rn} \tag{4}$

Where, λ_u is the decay constant of uranium (4.883x10⁻¹⁸ d⁻¹).

Uranium weight in the samples in (g) unit can be calculated from equation (5)

 $W_U = N_U M_U / N_A \tag{5}$

Where M_u is the mass number of uranium, N_A is the Avogadro number $(6.02 \times 10^{23} \text{ mol}^{-1})$.

Uranium concentrations in(ppm)unit can be calculated from equation (6)

 $C_U = W_U / M_S \tag{6}$

Where, M_s is the mass of the studied soil samples in (g) unit [14].

III. RESULTS AND DISCUSSIONS

A) Radon and Uranium Concentrations

Radon concentrations values in airspace inside the irradiation cylinders obtained from passive CR-39 and active RAD-7 techniques were found in a good agreement and fig. 3 shows the linear relation between CR-39 and RAD-7. Radon average concentrations in the airspace of irradiation cylinders measured by RAD-7 and CR-39 were 35.17 and 32.616 Bq/m³. Radon concentrations in the airspace of irradiation cylinders measured by RAD-7 were ranged from 25.1Bq/m³ in S6, S7, and S10samples to 58.6Bq/m³ in S1sample. While Radon concentrations in the airspace of irradiation cylinders measured by CR-39 were ranged from 23.86Bq/m³ in S7sample to 46.98Bq/m³ in S1sample. Radon concentrations in the airspace of irradiation cylinders and samples measured by CR-39 depended on the track density on the exposed surface of the CR-39 detector, and the exposure time of the irradiation cylinder samples (days) and the dimensions.

Radon concentrations in the samples were ranged from 2425.9431 Bq/m³ inS7 sample to 4796.4319Bq/m³ in S5sample.Uranium concentrations were ranged from 0.192 ppm in S7sample to 0.367 ppm in S5 sample. There was variation in the results of the studied soil samples due to the difference in soil type and the geological structure of the area. Fig. 4,5, 6 and Table II show radon concentrations in airspace inside the irradiation cylinders ($C_a^{222}Rn$) measured by RAD-7 and CR-39, and concentrations of radon ($C_s^{222}Rn$) and uranium (C_s U)in the soil samples.

B) Heavy Metals Concentrations

The analysis results of the heavy metals are ranged in Table III and shown in Fig.7. Nickel (Ni) concentrations ranged from 91.3 ppm in S1 sample to ppm in S7 sample. Chromium (Cr) 124.9 concentrations ranged from 58 ppm in S1sample to76 ppm in S8 sample. Zinc (Zn) concentrations ranged from 44 ppm in S4 sample to 63 ppm in S10 sample. Copper (Cu) concentrations ranged from 16.4 ppm in S1 and S2 samples to 23.1 ppm in S10 sample. Cobalt (Co) concentrations ranged from 12.2 ppm in S1 sample to 16.7 ppm in S7 sample. Lead (Pb) concentrations ranged from 7.5 ppm S1 and S3 samples to 13.1 ppm in S5 sample. Arsenic (As) concentrations ranged from 4.9 ppm in S2 sample to 6.3 ppm in S3 sample. Cadmium (Cd) concentrations ranged from 0.2 ppm to 0.3 ppm where the highest values were in S1 and S3 samples and the other samples recorded lower values.

Concentrations of heavy metals in the soil depend mainly on the geological structure of the studied area, sources of pollution and the type of technique used for measurement. Measuring a metalsconcentrations usingan ICP-MS technique shows a different result than that measured using the AAS or X-RAY techniques or other techniques. Also the concentrations of heavy metals depend on the depth at which soil samples were taken from, the concentration of a metal at a depth of 20 cm is not the same at depth of 10 cm or 30 cm.

C) Relationship Between Radon and Heavy Metals Concentrations

Statistical package for the social sciences (SPSS) program has been used to find the correlation factor between radon concentrations with heavy metals concentrations. However, it was observed that there is

a difference in the correlation factor between the concentrations of radon and heavy metals from one technique to another, where the correlation factor for radon concentrations measured by RAD-7 and heavy metals was not as that measured by CR-39 because of the accuracy difference between the techniques.

Radon concentrations measured by RAD-7 has apositivecorrelation to Cd and U metals and negative correlation to As,Cu,Co,Cr,Pb,Zn and Ni metals, the highest correlation was 0.850 for U and the lowest was -0.672 for Co. Radonconcentrations measured by CR-39 detectors have a positive correlation to Pb,Cd and U metals and negative correlation to As,Cu,Co,Cr,Zn and Ni metals, the highest correlation was 0.989 for U and the lowest was -0.426 for Co.

The highest concentrations of uranium, lead, and radon in the soil samples were found in S5sample. This is due to the fact that radon and lead both are one of the results or products of the uranium (²³⁸U) decay chain which ended by stable lead (²⁰⁶Pd) after chain of decays of the daughter radon (222Rn). On the other side, the lead concentrations in the other soil samples showed random variation and less dependent on the radon and uranium concentrations in that samples. Also, radon concentrations varied from one location to another independently on concentrations of heavy metals concentrations except for uranium which was the only metal among the measured metals has strong correlation to the concentrations of radon. Table IV shows the results of correlation factor between concentrations of radon and heavy metals and Fig. 8 shows radon (Bq/m^3) and heavy metals(ppm) concentrations in soil samples.

It was difficult to find aevident relationship between radon and heavy metals concentrations in the studied soil samples because the soil samples during preparing them for the measurement process influenced by the externalconditions. Also, the soil samples were taken from farms irrigated with Khasa river water that mixed with untreated wastewater which is contaminated with high concentrations of heavy metals, and these concentrations change rapidly and continuously and effected on the soil coality. There were no previous studies in this field to know what they found in their studies and compare their results with the results obtained in the current study. Therefore, this field requires further studies for different sites and samples and to include more metalsand all radiations types and find their impacts on the environment and public health.

IV. CONCLUSION

In the present study radon and heavy metals concentrations measured for soil samples taken from selected farms in Kirkuk city on both sides of khasa river northeast Iraq. The highest radon concentration in the soil samples was found in sample S5 which was 4796.4319 Bq/m³ and the lowest was in sampleS3 which was 2777.3858 Bq/m³. Average concentrations value of the heavy metals in unit ppm have an order (112.28Ni>68.7Cr>53.1Zn>20.18Cu>14.68Co>10.16 Pb >5.27As >0.22Cd). Statistical package for the social sciences (SPSS) program was used to find the relationship between radon and heavy metals concentrations. It was found that there is no strong correlation between the concentrations of radon and heavy metals in the studied soil samples except with uranium which had a strong correlation to radon concentrations.

V. REFERENCES

- M. J.Issa ,A. SH. Qanber, "Assessment of Heavy Metal Contamination in Euphrates River Sediments from Al-Hindiya Barrage to Al-Nasiria City, South Iraq," Iraqi J. Sci., vol.57, nol 3, pp.184-193, 2016.
- [2] R. A. A. Hanan, A. G. Khaled, and M. G. Hassan, "Heavy Metals in Soils and Plants from Various Metal- Contaminated Sites in Egypt," Terr. Aquat. Environ. Toxicol., vol. 1, no. 1, pp. 7–12, 2007.
- [3] L. N. M. Tawfiq, K. A. Jasim, and E. O. Abdulhmeed, "Pollution of Soils by Heavy Metals in East Baghdad in Iraq," Int. J. Innov. Sci. Eng. Technol., vol. 2, no. 6, pp. 181–187, 2015.
- [4] H. R. Habib, S. M. Awadh, and M. Z. Muslim, "Toxic Heavy Metals in Soil and Some Plants in Baghdad, Iraq," J. Al-Nahrain Univ., vol. 15, no. 2, pp. 1–16, 2012.
- [5] Y. M. Z. Al-bakhat, N. H. K. Al-ani, B. F. Mohammed, N. H. Ameen, Z. A. Jabr, and S. H. Hammid, "Measurement of Radon Activity in Soil Gas and the Geogenic Radon Potential Mapping

Using RAD7 at Al-Tuwaitha Nuclear Site and the Surrounding Areas," Radiat. Sci. Technol., vol. 3, no. 3, pp. 29–34, 2017.

- [6] A. K. Hashim and E. J. Mohammed, "Measurement of Radon Concentration and the Effective Dose Rate in the Soil of the City of Karbala, Iraq," J. Radiat. Nucl. Appl., vol. 23, no. 1, pp. 17–23, 2016.
- [7] P. State, "Note Determination of 222Rn Level in Ground Water Using a RAD7 Detector in the Bathinda District of Punjab," Radiat. Prot. Dosimetry, vol. 156, no. 2, pp. 239–245, 2013.
- [8] N. F. Tawfiq, H. L. Mansour, and M. S. Karim, "Measurement of Radon Gas Concentrations in Tap Water Samples for Anbar Governorate by Using Nuclear Track Detector (CR-39)," Int. J. Recent Res. Rev., vol. VIII, no. 4, pp. 7–15, 2015.
- [9] M. Batsala et al, "inductively coupled plasma mass spectrometry (ICP-MS)," Int. J. Res. Pharm. Chem., vol. 2, no. 3, pp. 671–680, 2012.
- [10] W. J. Speelman, "Modelling and Measurement of Radon Diffusion Through Soil for Application on Mine Tailings Dams," Master's Thesis, University of the Western Cape, South Africa, Dec, 2004.
- [11] H. H. Aziz, "A Study of Radon and Thoron Gases Release from Iraqi-Kurdistan Building Materials Using Passive and Active Methods," Master's Thesis, University of Salahaddin, Iraq, Apr, 2010.

- [12] S. K. Ezzulddin and H. H. Mansour, "Assessment of Radon Exposure in Erbil Drinking Water Resources," ZANCO J. Pure Appl. Sci., vol. 29, no. 4, pp. 184–195, 2017.
- [13] A. H. Ahmed, H. H. Mansour, S. K. Ezzulddin, and A. I. Samad, "Radon (²²²Rn) Content in Bottled Water of Kurdistan Region using Active and Passive Methods," Kirkuk Univ. J. /Scientific Stud., vol. 29, no. 3, pp. 152–159, 2017.
- [14] K. H. Abass, A. O. Muhsen, A. Mayyali, B. Y. Mohammed, and J. M. Hussien, "Measurement the Concentration of Radon Gas Emitted From Infant Powdered Milk Consumed in Iraq Using Nuclear Track Detector CR-39," J. Chem. Pharm. Sci., vol. 10, no. 2, pp. 906–909, 2017.
- [15] A. Panas et al., "Assessment of Rn and U Concentrations in the Soil of Qadafery, Kalar and Zarayan Located in Sulaimani Governorate of Kurdistan Region- Iraq," Am. J. Environ. Prot., vol. 4, no. 1, pp. 40–44, 2015.

No	Site Name		Sample code	Longitude	Latitude
1	Wahid athar	F1	S 1	44°22'04	35°23' 51
2	Domiz	F2	S2	44°22'22	35°24'06
3	In the frontasrawlmafqodinmosque	F3	S3	44°22'42	35°24'20
4	Hay adan	F4	S4	44°22'50	35°24'38
5	In front ofrihab near khabbat bridge	F5	S5	44°23'36	35°28'29
6	Barot khana	F6	S 6	44°24'57	35°29'42
7	Imam Qasim in front of Qazi Muhammad high school	F7	S7	44°23'41	35°28'39
8	In front of Alsabor mosque	F8	S 8	44°23'21	35°27'30
9	In front of Kirkuk Electricity Maintenance Division	F9	S9	44°23'24	35°27'38
10	Qasab khana near ALwilada bridge	F10	S10	44°23'21	35°27'06

Table I Names, codes, and coordinates of sampling sites in the study area

Table II

Radon concentrations in airspace inside the irradiation cylinders C_a^{222} Rn (Bq/m ³) measured by RAD-7 and CR-39,
and concentrations of radon C_s^{222} Rn (Bq/m ³) and uranium C_s U(ppm) in the soil samples

Farm code	Ca ²²² Rn Measured by RAD-7(Bq/m ³)	Ca ²²² Rn Measured by CR-39 (Bq/m ³)	C _s ²²² Rn (Bq/m ³)	C _s U (ppm)
F1	58.6	46.98	4544.9939	0.364
F2	25.2	24.56	2524.5224	0.197
F3	33.5	27.02	2777.3858	0.195
F4	41.9	39.15	3787.371	0.290
F5	50.2	41.21	4796.4319	0.367
F6	25.1	26.03	3029.9876	0.236
F7	25.1	23.86	2425.9431	0.192
F8	33.5	34.71	4039.9834	0.333
F9	33.5	33.93	3282.3882	0.264
F10	25.1	28.71	2777.496	0.215
Av	35.17	32.616	3398.65	0.2653

Table III Heavy metals concentrations (ppm) in soil samples

Farm code	As	Zn	Cu	Pb	Co	Cr	Ni	Cd
F1	5.0	46	16.4	7.5	12.2	58	91.3	0.3
F2	4.9	46	16.4	9.9	13.6	61	98.6	0.2
F3	6.3	48	20.4	7.5	14.4	73	106.8	0.3
F4	5.2	44	17.6	8.2	12.5	62	96.8	0.2
F5	5.3	59	21.5	13.1	14.7	66	113.1	0.2
F6	5.0	61	21.6	9.1	15.6	73	124.5	0.2
F7	5.1	55	21.4	11.0	16.7	73	124.9	0.2
F8	5.4	55	22.1	11.0	15.9	76	124.6	0.2
F9	5.2	54	21.3	11.7	15.2	70	117.7	0.2
F10	5.3	63	23.1	12.6	16.0	75	124.5	0.2
Av	5.27	53.1	20.18	10.16	14.68	68.7	112.28	0.22

Table IV

Metals	As	Cu	Pb	Zn	Co	Cr	Ni	Cd	U	Rn*	Rn**
As	1										
Cu	0.291	1									
Pb	-0.204	0.652^{*}	1								
Zn	-078	877**	721*	1							
Co	0.094	0.888^*	0.637^{*}	0.799**	1						
Cr	0.423	0.908**	0.394	0.694*	0.895**	1					
Ni	0.050	0.934*	0.636*	0.866**	0.972**	0.906**	1				
Cd	0.508	-0.383	-0.686*	-0.475	-0.481	-0.261	-0.534	1			
U	-0.181	-0.138	0.74	-0.53	-0.383	-0.380	-0.259	0.107	1		
Rn*	-0.008	-0.426	-0.235	-0.368	-0.672*	-0.633*	-0.611	0.490	0.850**	1	
Rn**	-0.093	-0.146	0.040	-0.074	-0.426	-0.400	-0.305	0.162	0.989**	0.887^*	1
Rn*. Rac	lon concen	trations m	easured by	RAD-7.							
Rn**. Ra	adon conce	entrations n	neasured b	y CR-39.							
*. Correl	ation is sig	nificant at	the 0.05 le	evel.							
**. Corre	elation is si	ignificant a	t the 0.01	level.							

The correlation factor between radon concentrations (Bq/m³) with heavy metals concentrations (ppm)

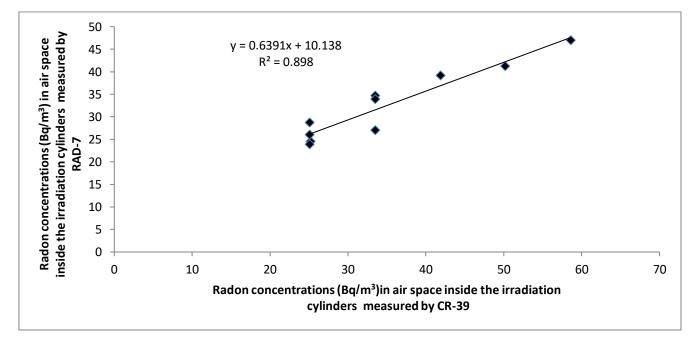


Fig. 3. The linear relation between CR-39 and RAD-7.

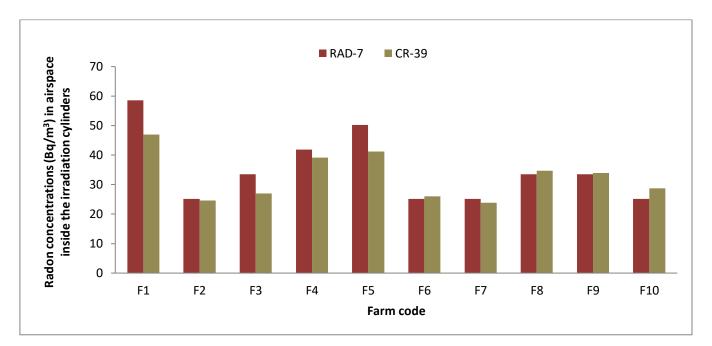


Fig. 4 .Radon concentrations (Bq/m^3) in airspace inside the irradiation cylinders measured by CR-39 and RAD7 in the soil samples

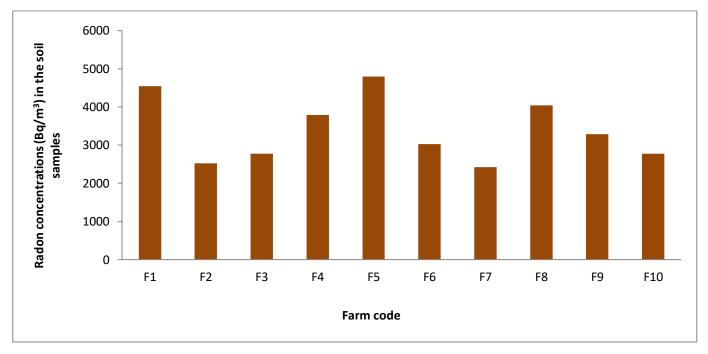


Fig. 5. Radon concentrations (Bq/m³) in the soil samples

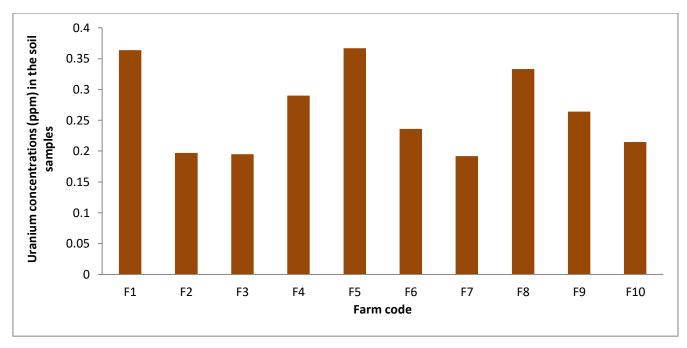


Fig. 6. Uranium concentrations (ppm) in the soil samples

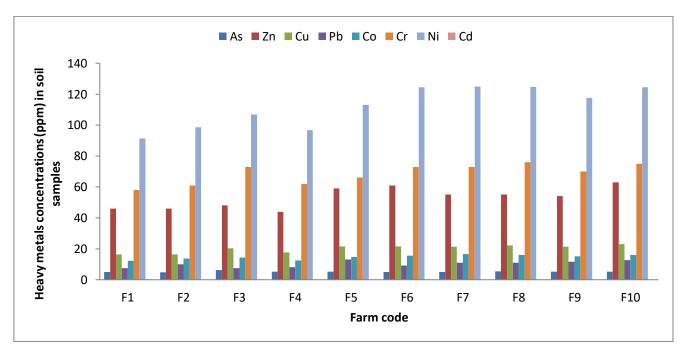


Fig. 7.Heavy metals concentrations (ppm) in the soil samples

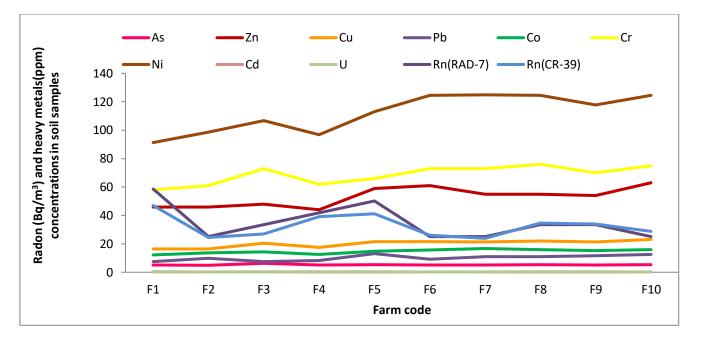


Fig. 8. Radon (Bq/m³)and heavy metals (ppm) concentrations in the soil samples