



Radionuclide concentrations analysis of Duhok air atmosphere by gamma spectrometry

Análisis de las concentraciones de radionúclidos de la atmósfera del aire de Duhok por espectrometría gamma

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ABSTRACT

Introduction: Atmospheric air is directly related to human health and irreplaceable to human life and thus an influential parameter of environmental science. Radioactive materials in the air may result in exposure of man every day of our lives by inhalation or ingestion of particulate matter suspended or deposited on vegetation or products derived from animals, which has been ubiquitous on earth since its creation. The main goal of this study is to measure the radioactivity concentration of the natural and artificial radionuclides of the Duhok air. **Materials and Methods:** Twenty samples of air filters were collected from different locations of Duhok City and its surroundings by low volume air samplers pump in the summer/autumn and winter/spring seasons. Air filter samples were prepared and analyzed by a well-type thallium-activated sodium iodide NaI(-TI) detector. **Results and Discussion:** The average activity concentrations of ^{214}Pb , ^{214}Bi , ^{212}Pb , ^{228}Ac , ^{40}K and ^{137}Cs are 4.98 ± 1.20 , 4.54 ± 1.27 , 9.76 ± 1.17 , 10.72 ± 2.35 , 211.91 ± 62.19 and 1.78 ± 0.61 mBq/m³ respectively of the summer/autumn seasons. Whereas, in the winter/spring seasons are 3.89 ± 1.23 , 4.21 ± 1.43 , 5.05 ± 1.08 , ND, 147.49 ± 46.38 and 1.78 ± 0.69 mBq/m³ respectively. **Conclusions:** The activity concentrations results confirmed seasonal variations for all study locations. Activities of all radionuclides, except an anthropogenic radionuclide ^{137}Cs which remains about the same in both seasons, record higher values in summer/autumn season. Moreover, all activities are less than the acceptable lower level. It is clear that the prominent radioelements that affected by atmospheric condition is ^{40}K even though it is less than the reported lower level of the EPA

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INTRODUCTION

Today, the issue of environmental pollution has become an important topic to be discussed at international levels, and to organize international conferences to find solutions for the issue. The natural radioactivity distribution as well as the manmade pollution in environment beats the alarm of the hazardous of our life that have become a big problem in this century⁽¹⁾.

Environmental pollution has serious public health implications with changes on chemical, physical and biological characters of main life sources, air, soil and water. This pollution affect the human survival and other living organs or at least limit their activities permanence⁽²⁾. Pollution has considerable potential since ionizing radiation damaging biological material specifically from those of very long half-lives.

Radioactive material has been released to the environment from various sources as a result of actions committed either on purpose or accidentally through negligence or simply ignorance⁽³⁾. Regarding the global radiological effective dose to the public, the major contributors to environmental contamination were the nuclear weapons tests and the accidents of the nuclear reactors or fuel cycle. According to UNSCEAR-2000 report, radionuclides rapidly appear in ground level and air. Air samples can give the first indication of the nature of the contamination⁽⁴⁾. In consequence of radioactive fallout after the Chernobyl nuclear reactor accident in 1986, such as ¹³⁷Cs radionuclide (T_{1/2}=30.07yr). The 2011 WHO report, these radionuclides could present a risk to human health. Low doses caused by ingestion of these radionuclides in drinking water can increase the radiological risk of longer term effects⁽⁴⁾.

The determination of the air particles radioactivity concentration is a major aspect in establishing reference data, allowing the supervision and control of possible future changes due to future radiological contamination⁽⁴⁾. The primary objective of the study is to monitor the concentration of different radionuclides in air particles. The data obtained from the study will be beneficial to predict amount of radia-

tion exposed by the city resident.

MATERIALS AND METHOD

Study Area

Duhok governorate lies in the far north-west of Iraq and forms the western governorate in Iraqi Kurdistan Region. It has a strategic location since it is considered to be a point of joint among three parts of Kurdistan (Syria-Turkey-Iraq). The governorate lies in the portable area between the wavy area and high mountainous area. It lies between the two lines 36.7°N, 42.3°E and 37.4°N, 44.2°E⁽⁵⁾—especially in Syria and Iraq. This study seeks to assess the spatial and temporal characteristics of the drought in the Duhok Governorate in Northern Iraq, focusing on meteorological, agricultural and socio-economic drought at province and village level. Satellite based precipitation data, validated by station data, were used in a meteorological drought assessment. To estimate the decreased precipitation's effects on vegetation, an agricultural drought assessment was performed using Enhanced Vegetation Index (EVI). Duhok-district: with area (1092) km² locates in the center of governorate. Generally, the climate of Kurdistan Region of Iraq (KRI) is of semi-arid type characterized by ranges of temperature about 40 0C in Sumer to freezing in winter, low relative humidity. Precipitation is concentrated in winter, specifically from November to April with about 400 mm annual rain. The northwest wind from Turkey is the dominant throughout the year⁽⁶⁾.

Field Work Preparation

Four different location are chosen depending on population, location usage in addition to the geographical properties related to the study area , a topographic map of the Duhok governorate prepared by JHIC, United Nations Joint Humanitarian Information Centre, have been used for determination of appropriate locations for sampling of air. Identifying and coding the studied samples has been done accordingly. The four locations and number of samples are:

Location	Sample code
1. Traffic light	A01, A02, A03, A04, A05
2. Street	A06, A07, A08, A09, A10, A11
3. Resident area	A12, A13, A14, A15, A16
4. Industrial area	A17, A18, A19, A20

Samples Collection and Preparation

A low volume air pump (Gilian BDXII Abatement Air Sampler, made in USA by SENSIDYNE) with Whatman 934-AH Glass Microfiber filter (25 mm) are used to collection the air samples.

Set of air sampler pump with filters was designed and placed outdoor at 1.50 m above the ground. The weather-protected equipment is housed in a small metal portable cabinet consisting of a pump, flow rate meter, flow adjust controller, and other equipment and/or electronics as necessary. The unit's power may be a hard-wired electrical outlet, batteries. The equipment has been successfully applied to collect atmospheric samples from different locations in Duhok city and around.

The weight of the filter is measured by a sensitive balance with 0.01mg scale or lower. Record the weight into information sheet, place it into cassette, and

assemble the cassette filter holder for the sampling media. A shrink tape around the cassette is used to cover joints and prevent leakage. Each cassette was marked to identify the sampling code, site, location and date which is fixed to a sample head on the exterior of the cabinet, cartridges can also be fixed to an exterior sample head, as shown in Fig. 1. Before and after each day of sampling the BDXII air pump has been calibrated to within $\pm 5\%$ of the true reading. Air pump calibration performed using a precision rotameter that has been calibrated.

A total of 40 air samples were collected from different locations throughout Duhok city and around. The first 20 air samples had been collected for all areas through 2 months of a dry summer/autumn season from 12/9/2017 to 12/11/2017. The other 20 air samples are from the same locations through 2 months of the rainy winter/spring seasons from 1/3/2018 to 29/4/2018. A period of ten to eleven hours for each air sample collection has been adopted with air flow of 2.5 L/m. After collection the weight of the filter is recorded again to know the weight of the contaminants on the filter and stored it inside a sealed plastic bag for measurements, see Figure 1(c).

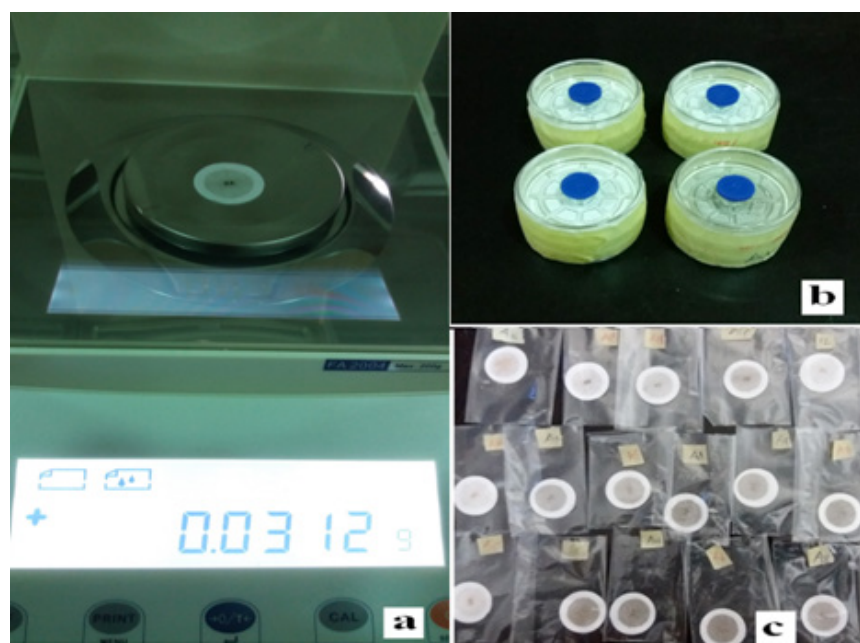


Figure 1. Preparation collection media (a) balance (b) cassette (c) sealed plastic bag and information sheet.

Sample Counting

Gamma spectroscopy is used to determine the concentrations of radionuclides in air samples. Filter of air sampling are placed at a midpoint point of the well detector at 2.5 cm from the bottom inside the active shielded volume and measured for a counting

time of 60000 sec.

The naturally occurring radionuclide considered in the present analysis of the measured γ -ray spectra are: ^{212}Pb , ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{40}K , and ^{137}Cs . Figure. 2 shows a typical spectrum of a sample A01 after subtracted from the background, via the software A65-BW MAESTRO.

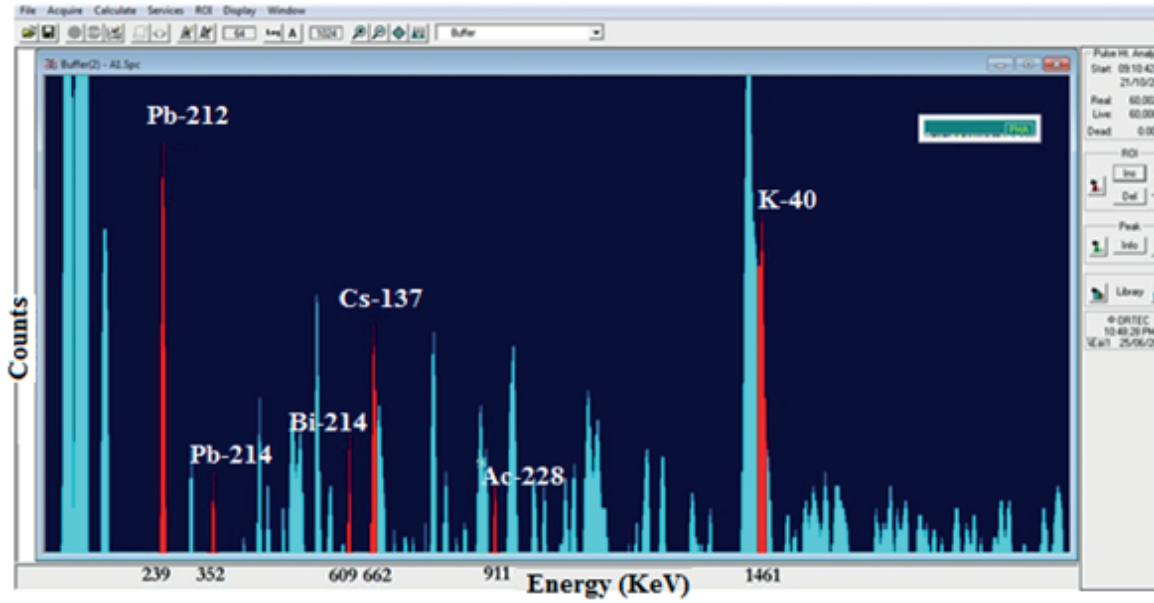


Figure 2. Gamma ray spectrum for air sample (A01) after subtraction of the background spectrum (energy scale zoomed).

Calculation of the Radionuclides Concentrations in Air

Following the spectrum analysis, count rates of each detected photo-peak and the activity for each of the detected nuclides are calculated. The activity A_{Ei} (Bq), of a nuclide i for a peak at energy E_i is given by (7,8).

$$A_{Ei} (Bq) = \frac{N_{Ei}}{\varepsilon_E * t * f_\gamma} \quad 1$$

Where N_{Ei} is the net area under photopeak E_i , t is a detection life time, f_γ is the probability of gamma disintegration of the radionuclide.

Here, $\varepsilon_E = 43.046e^{-0.002E}$, to the resulting points with a correlation coefficient of $R^2 = 0.9828$, is the spectrometric detection efficiency at energy E had been calculated at a midpoint of the NaI detector well by

(9). The average specific activity or activity concentration (activity per unit volume) is determined by the following equation (10):

$$C_e (Bq/m^3) = \frac{A(Bq)}{Q(m^3 \text{ min}^{-1}) * T(\text{min}) * \Sigma} \quad 2$$

Where Q is the average sample flow rate in cubic meters per minute,

T_s is air pumping time of each sample in minutes,

Σ is the collection efficiency of the media (filter) expressed as a dimensionless fraction of unity (typically 0.9 or greater) in our case 0.98, and

A is the activity on (or within) the filter media, given by Eq. 1

The product of two terms in the denominator of Eq.2 (i.e., $Q*TS$) represents the total volume of air sampled and the product of all three terms there ($Q*TS*\Sigma$) represents the total volume of air actually filtered.

RESULTS

The conversion of the measured counts for a time 60000 sec to specific activity or activity concentration (mBq/m^3) is done by Eq. 1 and 2. The average results of each location for natural radionuclides concentration of ^{214}Pb , ^{214}Bi , ^{212}Pb , ^{228}Ac and ^{40}K and artificial radionuclide ^{137}Cs at 1.5 meter above the ground level for 40 samples of air filter during two seasons (summer/autumn 12/9/2017 to 12/11/2017) and (winter/spring 1/3/2018 to 29/4/2018).

From Table 1 (column 3), the average activity concentrations for ^{214}Pb are $4.98\pm 1.20 \text{ mBq}/\text{m}^3$ and $3.89\pm 1.23 \text{ mBq}/\text{m}^3$ and for ^{214}Bi (column 4) are $4.54\pm 1.27 \text{ mBq}/\text{m}^3$ and $4.21\pm 1.43 \text{ mBq}/\text{m}^3$ in sum-

mer/autumn and winter/spring seasons respectively.

From Table 1 (columns 5), the average activity concentrations for ^{212}Pb are $9.76\pm 1.17 \text{ mBq}/\text{m}^3$ and $5.05\pm 1.08 \text{ mBq}/\text{m}^3$ and for ^{228}Ac (column 6) are $10.72\pm 2.35 \text{ mBq}/\text{m}^3$ and ND in summer/autumn and winter/spring seasons respectively.

The activity concentration of ^{40}K (column7) in air filters also revealed some seasonal structure. In the summer/autumn seasons, the arithmetic average was equal to $211.91\pm 62.19 \text{ mBq}/\text{m}^3$ while in winter/spring seasons it recorded $147.49\pm 46.38 \text{ mBq}/\text{m}^3$ at the traffic, street, resident and industrial locations. The activity concentration of ^{137}Cs (column8) are 1.78 ± 0.61 and $1.78\pm 0.69 \text{ mBq}/\text{m}^3$ for summer/autumn and winter/spring seasons respectively.

Figure. 3 summarizes the average activity concentration of the radionuclides of air samples in different locations for comparisons of each radionuclide in both seasons.

Table 1. The activity concentration of the radionuclides of air samples in (mBq/m^3).

Season	Location	^{214}Pb	^{214}Bi	^{212}Pb	^{228}Ac	^{40}K	^{137}Cs
Summer/autumn	traffic	5.58 ± 1.39	3.70 ± 1.04	16.21 ± 1.46	14.88 ± 2.86	189.47 ± 53.81	1.63 ± 0.50
	street	5.49 ± 1.44	5.27 ± 1.40	9.37 ± 1.23	8.92 ± 2.22	224.36 ± 73.42	1.44 ± 0.59
	resident	4.59 ± 1.10	4.68 ± 1.27	8.70 ± 1.15	7.09 ± 1.78	165.35 ± 46.98	1.88 ± 0.62
	industrial	4.26 ± 0.86	4.52 ± 1.38	4.75 ± 0.83	12.00 ± 2.53	268.44 ± 74.56	2.18 ± 0.73
	Average	4.98 ± 1.20	4.54 ± 1.27	9.76 ± 1.17	10.72 ± 2.35	211.91 ± 62.19	1.78 ± 0.61
Winter/spring	traffic	4.30 ± 1.24	3.25 ± 1.17	5.50 ± 0.67	ND	125.12 ± 48.59	1.24 ± 0.53
	street	3.88 ± 1.10	4.43 ± 1.60	4.17 ± 1.04	ND	119.13 ± 38.52	1.76 ± 0.73
	resident	3.82 ± 1.39	4.35 ± 1.48	5.35 ± 1.31	ND	150.67 ± 44.21	2.10 ± 0.73
	industrial	3.55 ± 1.18	4.80 ± 1.45	5.16 ± 1.28	ND	195.05 ± 54.21	2.02 ± 0.79
	Average	3.89 ± 1.23	4.21 ± 1.43	5.05 ± 1.08	ND	147.49 ± 46.38	1.78 ± 0.69
Derived Air Concentrations (DAC)		1×10^7	1×10^7	5×10^5	1×10^5	6×10^6	2×10^9

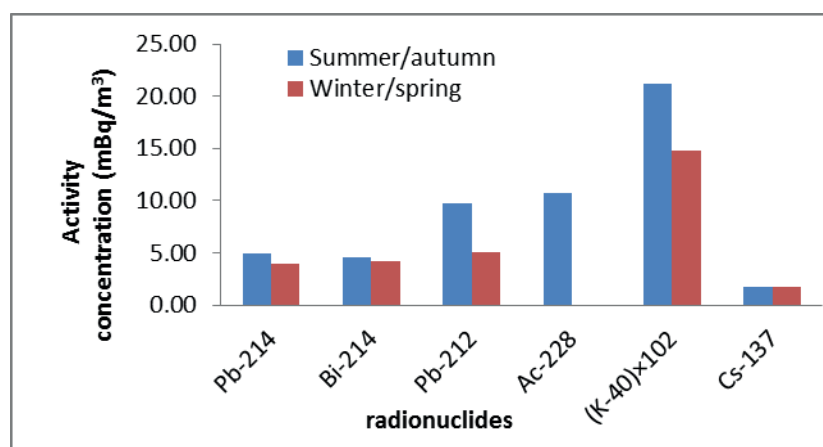


Figure 3. The average activity concentration of the radionuclides of air samples in for different location.

DISCUSSION

These two isotopes of ^{238}U series and come after the inert gas ^{222}Rn of half-life 3.85 day which allow to escape from the soil and ascend into the air. Accordingly their attachment to dust and other particles may be the most probable reason for getting them in air sample. At traffic light locations we get higher activity concentration of ^{214}Pb and ^{214}Bi in air sample compared to the other locations in all seasons. It may be attributed to the rate of sudden increase of vehicles in Duhok, since ^{214}Pb and ^{214}Bi are part of the petroleum fuel used (1).

The average radioactivity value of both ^{212}Pb and ^{228}Ac in the four locations in summer/autumn seasons are higher than that in winter/spring seasons. Since both radionuclides come from ^{232}Th , then they can escape from the soil and ascend into the air by the effect of wind. The presence of ^{212}Pb and ^{228}Ac in atmospheric dust particles and virtually all other types of geological materials and the smoke of the vehicles are the most probable reason of finding activity in air of the traffic light locations (11).

We find it abundantly at Duhok city there are lots of Building that used cement materials for construction. This is the most probable reason for finding ^{40}K (2). The lowest values of ^{40}K were observed in winter/spring season in all locations. This might be due to the big differences of the atmosphere between summer/autumn and winter/ spring seasons and the dry

grass emission-burning. The activity concentration of ^{137}Cs shows no differences in both dry and rainy seasons. This might be due to its original sources and its transportation all around the world by the wind as it is a volatile element (8). The obtained results for activity concentrations of all radionuclides are much less than the permissible limits or Derived Air Concentrations (DAC) for Occupational Exposure (12) as shown in the last row of Table 1 .

As shown in Table 2, the air radionuclides' activity concentrations were comparable to other studies conducted in Iraq. In Baghdad determined a higher activity concentration of ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{40}K and ^{137}Cs compared to this study. While the averaged concentration of ^{40}K attained in this study is close the reported average from Al-Najaf in Iraq.

In table 2, the present study concentrations in air samples of ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{40}K and ^{137}Cs were compared with randomly selected countries evaluations in air samples around the world. The averaged ^{214}Pb and ^{214}Bi concentration attained in this study is lower than all listed countries. The concentration of ^{228}Ac in this study is below the reported average from Bangladesh. The ^{40}K concentration is greater than all listed countries. The concentration of ^{137}Cs is higher than all listed countries except Switzerland. The differences from present values are logical since natural environmental radioactivity depend primarily on the geological and geographical conditions.

Table 2. The average activity concentration of radionuclides in air samples reported from some other locations of the Iraq and world.

Country		Activity concentration (mBq/m^3)						reference
		^{214}Pb	^{214}Bi	^{212}Pb	^{228}Ac	^{40}K	^{137}Cs	
Iraq	Summer	4.98	4.54	9.76	10.72	211.91	1.78	Present work
	Spring	3.89	4.21	5.05	ND	147.49	1.78	
Iraq/Baghdad		31.26	31.26	-	38.51	517.01	29.01	(13)
Iraq/Al-Najaf		-	-	-	-	192.61	-	(14)
Bangladesh		-	-	-	-	97	-	(11)
Bangladesh		402	543	143	57.33	92.9	-	(7)
Egypt		3860	-	-	-	-	-	(15)
USA		-	-	-	-	-	0.42	(16)
Spain		-	-	0.84	-	4.2	-	(17)
Poland		-	-	-	-	0.0147	0.017	(18)
Poland		250-1920	160-1620	9-27.5	-	-	-	(19)
Switzerland		5590	-	145	-	0.95	500	(20)
France		-	-	-	-	0.0077	0.000029	(21)
Italy		-	-	-	-	-	0.018	(22)



CONCLUSIONS

Even though numbers of samples per location are not adequate to come up with highly confirmed determinations and decisions, the results of activity concentrations show that the regional or local variation in the distribution and availability of natural radionuclides are related to the atmospheric condition, and human interventions. While an anthropogenic radionuclide ^{137}Cs distribution remains almost the same in all seasons.

At traffic light locations a higher activity concentration of ^{214}Pb and ^{212}Pb are recorded for air samples compared to the other locations in all seasons.

The main source of ^{40}K is a re-suspension of soil particles and it is nonvolatile element presented with sodium. This might explain why at winter and spring times, with high moist vegetation, ^{40}K levels are somehow lower. While since the atmosphere in summer and autumn is dry and hot, a lot of soil particles flies through the air by the act of wind. Also the dry grass emission-burning, might be a possible reason for higher level in summer/autumn seasons.

Cs is a volatile element, in gas state. Due to continuity of variety nuclear activities, tests and accidents,

the long lived radioactive isotope ^{137}Cs distribution on the earth surface all over the world became a part of Naturally Occurring Radioactive Materials (NORM). Accordingly any study of NORM should include this hazardous radionuclide.

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