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A Study of ^{226}Ra Concentration Ratios for Leafy Vegetables

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The aim of this work is to study the factors that influence the ^{226}Ra uptake by two types of leafy vegetables collected from different farms in Al Sharqiya region in Saudi Arabia with their corresponding soils and irrigation waters. Quality assurance and validation of the radioanalyses for the different samples matrices were carried out. It was a reasonable approach to determine the ^{226}Ra concentration ratios on the basis of both the exchangeable and total ^{226}Ra content in the soils. It was found that the activity concentration of ^{226}Ra in soils is independent of its corresponding activities in irrigation water but correlates well with the soil content of clay and silt. No significant dependency of the ^{226}Ra concentration ratios for the studied vegetables on the radium total activities in soils. Good negative correlations were found between the organic matter content of soils and the concentration ratios of ^{226}Ra in cabbage and lettuce. Some variations of the ^{226}Ra uptake were found through the two leafy vegetables which may be explained by other factors related to the metabolic characteristics of the plant species. The ^{226}Ra concentration ratios based on the exchangeable radium leached from soil for cabbage and lettuce were found to be higher than those determined on the basis of its total activity in soil. The exchangeable ^{226}Ra may contribute for a better understanding of its uptake by leafy vegetables in these farms.

Keywords: Concentration ratio, Agricultural farms, leafy vegetables, ^{226}Ra; exchangeable radium, Gamma spectroscopy, Saudi Arabia

Introduction

The agricultural soils consist of radionuclides, other minerals and their compounds; organic matter and water, all of which provide the system for the plants nutritive requirements [1]. The daily intake of vegetables is one of the pathways of radium to our bodies. The transfer factor of radium from soil to vegetables is influenced by many factors. This includes soil type, texture and components, concentration and chemical form of radium in soil, plant species and many other factors [2, 3]. Therefore, in order to evaluate the transfer of radium in soil-plant system, some factors should be considered and the dissolved fraction of radium that is soluble in the soil solution (exchangeable radium) should also be considered [4]. Few studies of the soil to plant transfer factor in Saudi Arabia have been reported in literature [5, 6]. This study was carried out as a part of a project initiated by the National Agriculture and Animal Resources Research Center, Riyadh, KSA. The main goal of this study was to investigate the ^{226}Ra uptake by leafy vegetables collected from some farms in Al Sharqiya region in Saudi Arabia. Two types of leafy vegetables (cabbage and lettuce) were utilized in this study due to their high consumption by the Saudi population and the reported conclusions that leafy vegetables appear to concentrate radionuclides to a higher degree than other types of plants [7, 8].

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Materials and Methods

Sampling and treatment

The sampling of soils, crops and irrigation water was carried out at seven neighboring farms in Al Sharqiya region, Saudi Arabia. Cabbage and lettuce samples were collected from the agricultural area and packed in plastic bags. In the laboratory, they were washed with distilled water, dried in an oven at 100° C for 8 hours. The dried samples were kept in a furnace at 400°C, then grinded and mixed thoroughly. The radium was leached out from the matrices by total acids digestion [9]. The final sample solution was transferred to a defined volume of the gamma counting container. The corresponding soils were collected to a depth of 15 cm, separated from stones and plant roots, packed in plastic bags and transferred to laboratory. The soil samples were oven-dried at 100°c to a constant weight, homogenized, sieved and packed into standard counting containers. Five liters of the irrigation water samples were collected from the wells in each farm using a pump, filtered through 0.45µ membrane filter, acidified and transferred to polyethylene bottles. All gamma counting containers were carefully sealed using adhesive epoxy and tape to prevent radon loss and kept for one month to acquire secular equilibrium between ^{226}Ra and its decay products.

Radioactivity measurements

The activity concentrations of ^{226}Ra in all types of samples were measured using a Canberra high purity coaxial germanium (HPGe) detector with 70% relative efficiency and the counting time was 90000 s. For soil, the absolute efficiency calibration was carried out using IAEA certified reference materials (IAEA-375, IAEA-313 and IAEA-RGU-1). The photopeak absolute efficiencies at each gamma energy line of ^{222}Rn decay products (^{214}Pb and ^{214}Bi) were determined. For water samples, ^{226}Ra and the spiked ^{133}Ba were extracted from four liters volume using a strong cation exchange resin as described by A. El-Sharkawy et al. [10]. For vegetation samples, the ^{226}Ra in the final sample solution was determined by comparison with acidified deionized water spiked with known amount of ^{226}Ra. The exchangeable ^{226}Ra fraction was leached from soils using 1M ammonium acetate [11, 12] and measured by gamma and alpha spectrometry.

Quality assurance

For quality assurance and procedures validation, replicate measurements of the ^{226}Ra standard in the defined volume of the counting container were carried out. Replicate samples of the IAEA-TEL-2012-03-sample 04 hay and IAEA-372 grass samples were analyzed following the same radiochemical procedure. Accuracy, bias and precision were determined for the associated replicate measurements. For the estimation of uncertainty, errors were propagated due to photopeak counts, ^{226}Ra tracer activity, sample weight and volume. The minimum detectable activities were estimated based on the equation presented by Currie [13]. The exchangeable radium was measured by alpha spectrometry following the method described by S. Nour et al. [14] and compared with the gamma measurements as shown in Figure(1).

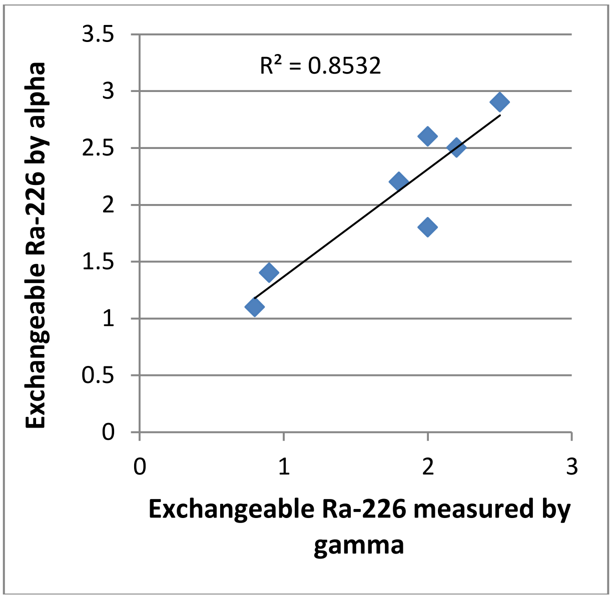

Figure (1): Correlation between the activities of the exchangeable ^{226}Ra measured by gamma and alpha spectrometry

Results and Discussion

^{226}Ra in irrigation water and soil

The activity concentrations of ^{226}Ra were determined in the irrigation water and soils of the selected farms in Bq/L and Bq/kg dry weight respectively as shown in Table (1).

Table (1): Specific activities of ^{226}Ra in water and soil samples ± 1σ uncertainties

Farm	Ra-226 in Water		Ra-226 in Soil	
Code	**Bq/L**	**±**	**Bq/kg**	**±**
F1	1.5	0.07	21.8	1.3
F2	2.8	0.15	61.6	3.9
F3	0.8	0.05	75.3	4.1
F4	2.1	0.14	27.1	1.4
F5	2.4	0.12	33.2	2
F6	1.8	0.1	48.7	3
F7	1.6	0.1	17	0.9

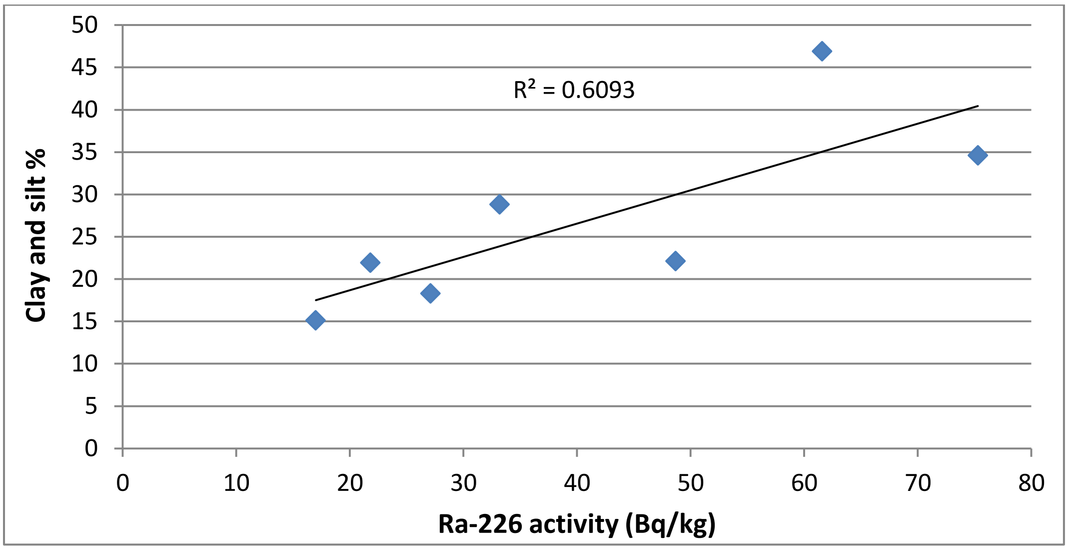

Figure (2): Correlation between soil texture and ^{226}Ra content in soil

The maximum values of ^{226}Ra activities in soil observed in farms F2 and F3 may be explained in terms of the variation in the soil texture [15] and the amounts of fertilizers added. The influence of soil properties on radionuclides content in soil was investigated by correlating the ^{226}Ra activity in soils with their combined clay and silt component as presented in Figure(2).

As shown in Figure(2), a good linear positive correlation was found between the soil clay and silt percent and the radium content, as was also confirmed by other authors in literature [16].

^{226}Ra in Vegetables and the associated Concentration Ratios

The activity concentrations of ^{226}Ra in cabbage and lettuce with the corresponding concentration ratios were determined and presented in Table (2). The observed difference in ^{226}Ra uptake for cabbage when compared with lettuce in some farms (F1, F5 and F7) may be attributed to differences in the uptake and translocation of radium among different plant species, the difference in metabolic rate and also the amount of exchangeable calcium in the soil [17, 18].

Table (2): specific activities of ^{226}Ra in vegetables ± 1σ uncertainties and the associated concentration ratios

Farm	Ra-226 in Cabbage		CR	Ra-226 in lettuce		CR
	Bq/kg dry weight	±	Cabbage	Bq/kg dry weight	±	Lettuce
F1	0.5	0.05	0.02	3.5	0.20	0.16
F2	1.8	0.10	0.03	0.7	0.05	0.01
F3	2.5	0.14	0.03	2.2	0.15	0.03
F4	3.9	0.2	0.14	2.5	0.15	0.09
F5	3.3	0.18	0.10	1.0	0.08	0.03
F6	4.1	0.20	0.08	4.4	0.25	0.09
F7	0.9	0.05	0.05	3.2	0.20	0.19

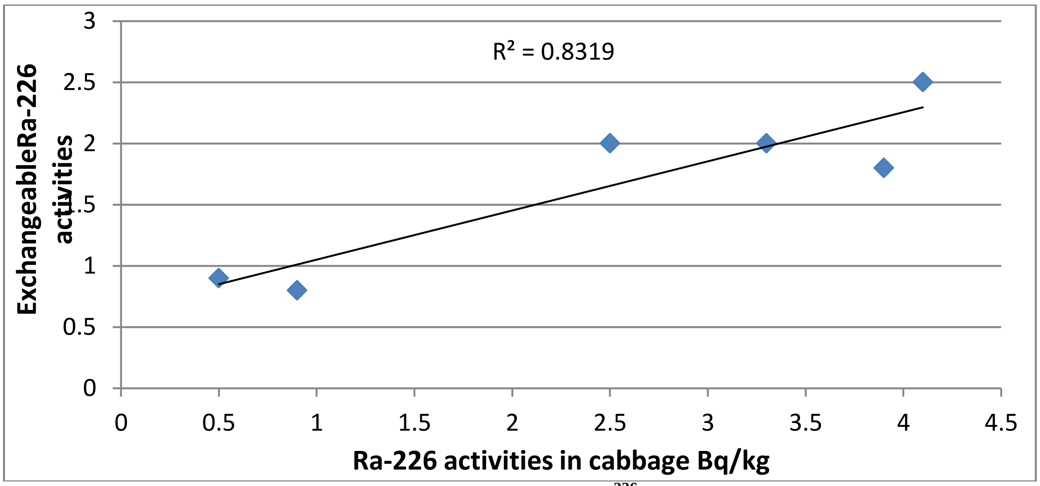

Figure (3): Correlation between the exchangeable ^{226}Ra and its content in cabbage

The results showed no correlations between the ^{226}Ra activities in the analyzed vegetables and its activities in irrigation waters and soils. Previous studies demonstrated that there is no general relationship between the soil and plant radionuclide activity concentrations, as reported by [19]. In contrast, the ^{226}Ra activities in cabbage from the different farms had a strong positive correlation with its activities in the extracted (leached) fraction of ^{226}Ra from soil as presented in Figure(3). In Figure(3), excluding two outliers leads to higher significant linear correlation with R^2 = 0.8 between the exchangeable ^{226}Ra and its activities in cabbage. This finding was in agreement with the reported conclusion that the bioavailable ^{226}Ra fraction in soils was found to be a better predictor of ^{226}Ra activity concentrations in fruits than the total soil activity concentration [20]. The mean concentration ratios for cabbage and lettuce were 0.07 and 0.09 respectively. The IAEA has reported that the soil-to-plant transfer factor of radium for leafy vegetables with a mean value of 0.09, ranged from 0.0018 to 130 [21]. The determined concentration ratios of ^{226}Ra in this work fall within the range of the corresponding values reported in literature in Qassim, Saudi Arabia for alfalfa which ranged from 0.09 to 0.19 [6], and in the Northwest of Saudi Arabia for eggplants which exhibited the highest uptake of ^{226}Ra transfer factor of 0.11 [5]. In a study of the influence of soils components on the concentration ratios of ^{226}Ra in the studied vegetables, a good negative correlation was found between the organic matter content and the concentration ratios of ^{226}Ra in cabbage and lettuce described by R^2 values of 0.80 and 0.88 respectively (excluding one outlier) as presented in Figure(4) and Figure(5). The presence of organic matter reduces radium accumulation by absorbing radium and thus making it less available to the plant. The ^{226}Ra concentration ratios for cabbage and lettuce determined with respect to the exchangeable ^{226}Ra (CRexch) are shown in Table (3).

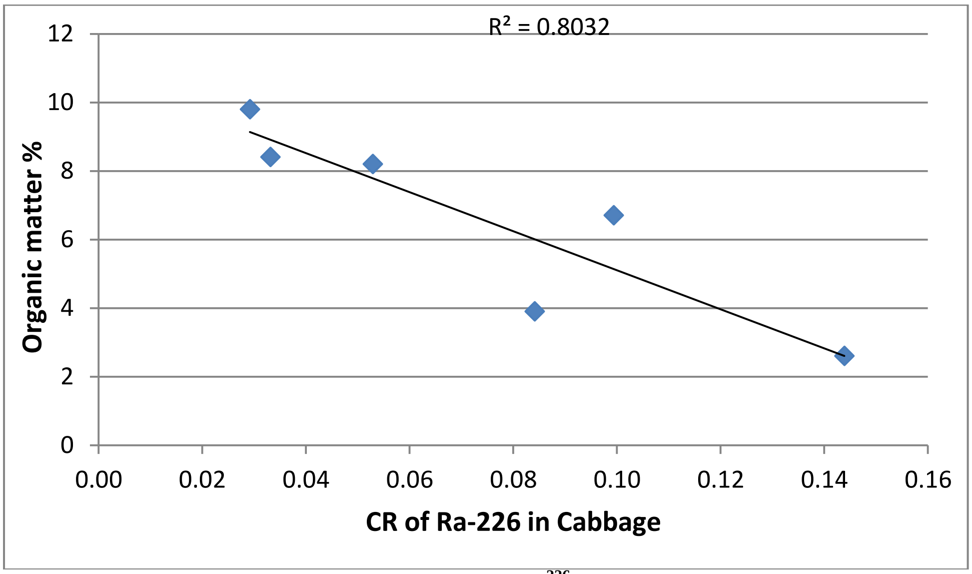

Figure (4): Correlation between concentration ratio of ^{226}Ra in cabbage and organic matter in soil

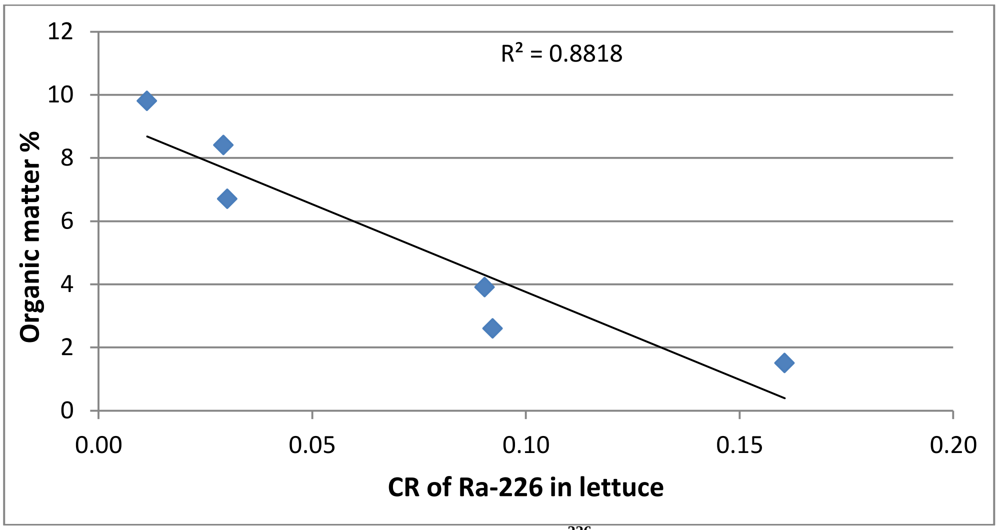

Figure (5): Correlation between concentration ratio of ^{226}Ra in lettuce and organic matter in soil

Table (3): concentration ratios of ^{226}Ra (CRexch) for cabbage and lettuce determined on the basis of the exchangeable radium ± 1σ uncertainties

Farm	CRexch Cabbage		CRexch lettuce	
	Bq/kg	±	Bq/kg	±
F1	0.56	0.05	3.89	0.25
F2	0.82	0.07	0.32	0.04
F3	1.25	0.08	1.10	0.1
F4	2.17	0.2	1.39	0.1
F5	1.65	0.15	0.50	0.05
F6	1.64	0.1	1.76	0.14
F7	1.13	0.08	4.00	0.3

Table (3) shows that higher ^{226}Ra concentration ratios are obtained with the exchangeable radium compared to its activities in soil, with average values of 1.32 and 1.85 respectively. Similar results showed that ^{226}Ra concentration ratios for edible vegetables calculated on the basis of exchangeable radium are one order of magnitude higher than those based on total radium [22].

Conclusion

In the frame of investigating some factors that influence the ^{226}Ra uptake by leafy vegetables, two types of vegetables (cabbage and lettuce) and their corresponding soils and irrigation water were collected from seven neighboring farms in Al Sharqiya region, Saudi Arabia. All samples were measured by gamma spectrometry using s high purity germanium detector. A good positive correlation between the combined clay and silt and the ^{226}Ra content in soils was observed. A highly significant linear correlation was found between the exchangeable ^{226}Ra in soil solution and its activities in cabbage. In addition, good negative correlations were found between the organic matter content and the concentration ratios of ^{226}Ra in cabbage and lettuce. The ^{226}Ra concentration ratios based on the exchangeable radium leached from soil for cabbage and lettuce were found to be higher than those determined on the basis of its total activity in soil. On the basis of the obtained results, it was concluded that the ^{226}Ra activity in soils is independent of its corresponding activities in irrigation water and seems to have no influence on its concentration ratios in vegetation, and correlates with the soil texture. The ^{226}Ra concentration ratios in leafy vegetables are correlated with the organic matter content in soil rather than the ^{226}Ra activities in irrigation water and soil. The ^{226}Ra uptake by leafy vegetables from the soil solution has to be considered, and its variation through different leafy vegetables may be explained by other factors related to the metabolic characteristics of the plant species.

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