Assessment of Natural Radioactivity of some Quarries Raw Materials in El-Minya Governorate, Egypt

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ABSTRACT

This study was undertaken to evaluate natural radionuclides activities of ^{238}U, ^{232}Th, and ^{40}K as well as silica and radon dusts as contaminants to assess the background radiation and distribution of these radionuclides in the soil of some quarries in El-Minya governorate, Egypt. Also it was done to establish a baseline data on the radiation profile of this area and to assess the radiological impact of these nuclides, silica and radon dusts on the health of the labors children in these quarries as well as assessing the environment quality of the study area. This work was performed in 10 quarries located in 5 different districts at El-Minya governorate. The radioactivity concentrations of the natural radionuclides (^{238}U, ^{232}Th and ^{40}K) in the collected quarries soil samples have been determined by gamma ray spectrometry. The radioactivity concentration values in the investigated soil samples ranged from (25.0±0.3) to (68.0±0.9) Bq kg^{-1} for ^{238}U, (37.0 ±0.4) to (88.0±2.0) Bq kg^{-1} for ^{232}Th, and (480.0±11.0) to (820.0 ±13.0) Bq kg^{-1} for ^{40}K. The study was performed on 50 working children and 50 non-working controls children. Five working children have been chosen from each quarry under 17 years old. All selected working and controls children were subjected to standard questionnaire, clinical examination with special emphasis on respiratory system, chest X-ray PA view and peak flow meter testing. The total absorbed dose rate values increase with the activity concentration, and consequently enhances the radiological impact on the workers in the quarries.

***Key words; Quarries/Raw Material/Natural radionuclides/*Labors children/El-Minya Governorate**.

INTRODUCTION

The need for protection from environmental pollution led to extensive studies regarding environmental radioactivity, migration and speciation of radionuclides. So, it is necessary to estimate the various natural radioactivity levels in the environment. Egypt has different sites of ornamental stone quarries for extraction the ornamental stones. El-Minya governorate (Fig.1) represents one of these regions.

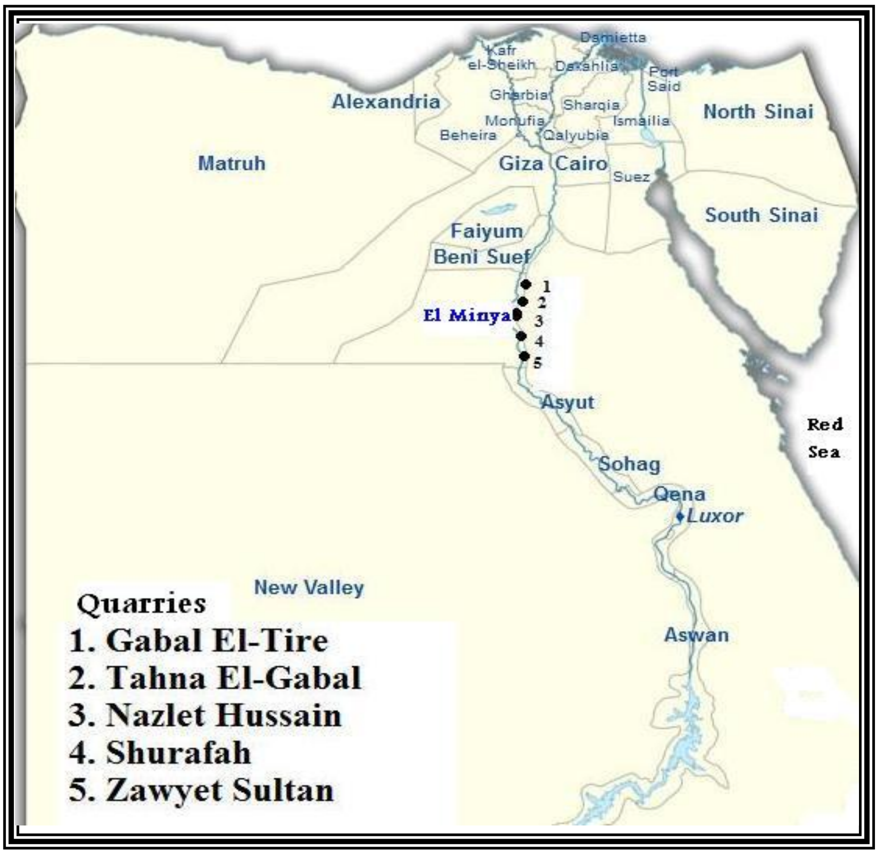

Figure (1) Studied area in El-Minya governorate

Ornamental stone have been used in decorative functions and is extracted by both surface and underground mining methods, depending on the working methods adopted for extraction and the challenges that face the working stability**(1)**. Quarrying operations have hazards effect on workers. Assessing the consequential risks of the extraction cycle has been divided into individual work stages. The basic approach consists in assessing the risk by examining each task in the work stage in relation to material agents with which an individual worker comes into contact in the course of his work**(1).** During stone cutting in quarries the workers exposure to airborne dusts, so the risks of respiratory diseases arise. These airborne dusts (1 μm to 100 μm in diameter) are mainly in the form of particulates, which produced through the mechanical processes such as breaking, grinding and pulverizing, and the primary route of exposure is inhalation**(2).** The small particles present greater hazard as they remain for longer periods and are more likely to gain access to respiratory system and either deposited or enter the circulatory system. There is clear connection between exposure to the dust and disease, where exposure to silica dust during stone cutting in quarries carries the risk of development of silicosis, progressive massive fibrosis, asthma, chronic obstructive pulmonary disease, and airways obstruction in exposed workers(2&3).

Naturally occurring radionuclides contribute significantly to the exposure of human for radiation. Among these radionuclides are the radioactive isotope of potassium ^{40}K and the radionuclides originated from the decay of ^{238}U and ^{232}Th series, both widely spread in soil and rocks of the earth's crust. Naturally occurring radionuclides exposure may be internal or external. External exposure is caused prevalently by gamma radiation from radionuclides in the ^{238}U and ^{232}Th series and from ^{40}K. Higher radiation levels are associated with igneous rocks, such as granite, and lower radiation levels with sedimentary rocks. Internal exposure is linked to radionuclides intake but the main cause is the inhalation of ^{222}Rn and its short-lived decay products. Radon ^{222}Rn is part of the radioactive decay of the ^{238}U series, which is present in building materials. Inhalation of the short lived decay products of ^{222}Rn provide is the main pathway for radiation exposure of the lung. There are number of circumstances in which materials containing natural radionuclides are recovered, processed or brought into position that results in exposure to radiation, i.e. human intervention causes enhanced exposures. In August 2000, the Italian authorities implemented the European Basic Safety Standards Directive (EU BSS) laying down safety standards for the protection of the health of workers and the general public against the dangers from ionizing radiation into national legislative regulation. According to the title VII of EU BSS, the new legislative regulation introduces a special section regarding working activities carried out in quarries where the processing of extracted materials is a source of potential exposure because of natural radioactivity high levels arising from a large amount of primordial radionuclides in the ^{238}U and ^{232}Th series and ^{40}K. Italian regulations are fixed the action level of 1 mSv y^{-1} for workers in these activities. This study was undertaken to evaluate natural radionuclides activities of ^{238}U, ^{232}Th, and ^{40}K as well as radon and silica as contaminants to assess the background radiation and distribution of these radionuclides in the soil of some quarries in El-Minya governorate, Egypt, to establish a baseline data on the radiation profile of this area and to assess the radiological impact of these nuclides on the health of the labors children in these quarries as well as assessing the environment quality of the study area.

MATERIALS AND METHOD

Five representative soil samples used for this study were collected close to the east of the River Nile at El-Minya governorate. The soil sample locations include the following villages: Shurafah, Zawyet Sultan, Nazlet Hussain, Tahna El-Gabal, and Gabal El-Tire. Two quarries of each village selected to collect one representative soil sample for every village. The samples were ear-dried at room temperature for a week. Then milled and sieved through 0.2mm sieves.

Fifty working children were selected for this study. Five working children have been chosen from each quarry under 17 years old. All selected working children were subjected to standard questionnaire, clinical examination with special emphasis on respiratory system, chest X-ray postreoanterior (PA) view, and peak flow meter testing. The exposed children were compared to 50 children as control; with no history of working in quarries. The controls children were matched for the same age groups and they were subjected to the same tests done to the quarries working children.

Monitoring air quarries have been done to detect silica dust, radon and particulate matter less than 10 μm. Determination of silica occurred by dissolving the dust sample in HF, and then measured by using atomic absorption. The activities of the investigated radionuclides in the soil samples were determined by direct gamma spectrometry, using a high purity germanium (HPGe) detector, with an efficiency of 30% relative to a 3″x 3″ NaI(Tl) scintillator and an energy resolution (FWHM) of 1.8 keV for the 1.33 MeV reference transition of ^{60}Co, was utilized for the measurements. The detection of gamma radiation is used to determine the concentration of the ^{40}K, ^{238}U and ^{232}Th. The polyethylene counting vessels (Marinelli beakers) were sealed gas-tight and stored for 4 weeks to allow radioactive equilibrium of the uranium and thorium series depending on the assumption of secular equilibrium, where the rate of decay of the daughters becomes equal to the rate of decay of the parent[4&5]. The prepared Marinelli beakers were placed on the detector endcap. Both the sample and the detector were surrounded by a cylindrical graded-Z shield of 5cm thickness of Lead, 1cm thickness of Iron and 1cm thickness of Aluminum to suppress the background radiation. A spectroscopic amplifier, with an efficient pile-up rejecter, and an 8k ADC (Analog-to-Digital Converter) processed the signal. The MAESTRO-32 multi-channel analyzer emulation software was utilized for data acquisition, storage, display and online analysis of the spectra. Measurements with an empty Marinelli beaker, under identical conditions, were also carried out to determine the ambient background in the laboratory site. The latter was subtracted from the measured spectra to obtain the net radionuclide activities. The gamma lines; 351.9 kev of ^{214}pb, 609.3 kev of ^{214}Bi, 1120.3 kev of ^{214}Bi and 1674.5 kev of ^{214}Bi were used for determining ^{226}Ra (daughter of ^{238}U).The gamma lines 338.34 kev of ^{228}Ac, 583.0 kev of ^{208}Tl, and 911.1 kev of ^{208}Ac were used for determining ^{232}Th series., while ^{40}K was measured by its gamma line 1460.7 kev. The uncertainty in the calculated efficiency was estimated to be 5 %. Statistical analyses were carried out using "Statistical Package for Social Science (SPSS) Inc., Chicago, IL, USA" (version 17).

RESULTS AND DISCUSSION

The results of gamma-ray measurements of ^{238}U, ^{232}Th and ^{40}K activity concentrations of all quarries soil samples are given in Table 1 and Figures 2&3. The activity concentrations vary from site to site, which means a large variation in chemical and mineralogical properties of soil samples[6]. In all samples of quarries, activity concentrations are in order ^{238}U < ^{232}Th < ^{40}K. The radioactivity concentration values in the investigated soil samples ranged from (25.0±0.3) to (68.0±0.9) Bq kg^{-1} for ^{238}U, (37.0 ±0.4) to (88.0±2.0) Bq kg^{-1} for ^{232}Th, and (480.0±11.0) to (820.0 ±13.0) Bq kg^{-1} for ^{40}K. It is clearly evident that ^{40}K always contributed to the most specific activity compared with ^{238}U and ^{232}Th. The sample of Zawyet Sultan presents the lowest activity concentrations of ^{238}U, ^{232}Th and ^{40}K. Table (1) presents also the elemental concentrations of ^{238}U, ^{232}Th and ^{40}K. They were calculated after conversion of ^{238}U, ^{232}Th and ^{40}K in Bq/Kg to concentrations of ^{238}U and ^{232}Th in (ppm) as well as ^{40}K in % using the conversion factors given by Polish Central Laboratory For Radiological Protection:, 1Bq kg^{-1} of ^{238}U = 0.08045 ppm of ^{238}U, 1 Bq kg^{-1} of ^{232}Th = 0.24331 ppm of ^{232}Th and 1 Bq kg^{-1} of ^{40}K = 0.003296 %^{40}K[7&8]. The results of these calculations are also given in Figure (3). The elemental concentration of ^{238}U is ranged from 2.008 ppm to 5.462 ppm. The elemental concentration of ^{232}Th is ranged from 9.139 ppm to 21.736 ppm. It is clear that the elemental concentration of ^{238}U and ^{232}Th are within the international accepted values of 10 mg/kg and 20 mg/kg, respectively, except in quarry of Gabal El-Tire has slightly higher concentration of ^{232}Th[9].

In order to assess the radiological impact of the investigated radionuclides in the soil samples, the γ- radiation doses can be estimated by employing the convenient formula [10].

$$D = (0.462AU+0.621ATh+0.0417AK)\ nGyh^{-1}$$

Where:

D is absorbed gamma dose rates,
AU, ATh, and AK represent the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in $Bqkg^{-1}$, respectively.

The absorbed gamma dose rates in air 1m above the ground surface for the uniform distribution of radionuclides (^{232}Th, ^{238}U, and ^{40}K) were computed on the basis of guidelines provided by European Commission Report on Radiological Protection Principles (1999) [11]. The conversion factors used to compute absorbed gamma dose rates (D) in air per unit activity concentration in ($1Bqkg^{-1}$) samples are 0.621 $nGyh^{-1}$ for ^{232}Th, 0.462 $nGyh^{-1}$ for ^{238}U, and 0.0417 $nGyh^{-1}$ for ^{40}K[10,12&13]. The recommended acceptable total absorbed dose rate by the workers in areas containing γ -radiations from ^{238}U and ^{232}Th series and their respective decay progenies, as well as ^{40}K, must not exceed 55 $nGyh^{-1}$[11]. It is obvious that the calculated total absorbed dose rates for all samples are in the range of the accepted dose levels. It is clear that the absorbed dose rates depend on the activities of γ -emitters (e.g. ^{238}U, ^{232}Th and ^{40}K). Therefore, the total absorbed dose rate values increase with the activity concentration, and consequently enhances the radiological impact on the workers in the quarries**.** Literatures indicate an average outdoor terrestrial gamma dose rate in the world ranging from 10 $nGyh^{-1}$ to 200 $nGyh^{-1}$[14] It is clear that the average gamma dose rates are in the range of the world average. The level of gamma radiation is directly associated with the activity concentrations of radionuclides in the sediment samples.

The Annual effective dose equivalent (Deff):

The annual effective dose equivalent received by a member has been calculated from the absorbed dose rate by applying dose conversion factor of 0.7 $SvGy^{-1}$ for the conversion coefficient from absorbed and the occupancy factor for outdoor as 0.2 using the following equations[15]:

$$\text{Deff (Outdoor) } (mSvy^{-1}) = (\text{Absorbed dose})\ nGy\ h^{-1} \times 8760h \times 0.7\ SvGy^{-1} \times 0.2 \times 10^{-6}$$

The annual effective dose equivalent received values ranged from 0.0729 mSv y^{-1} to 0.1383 mSv y^{-1}.

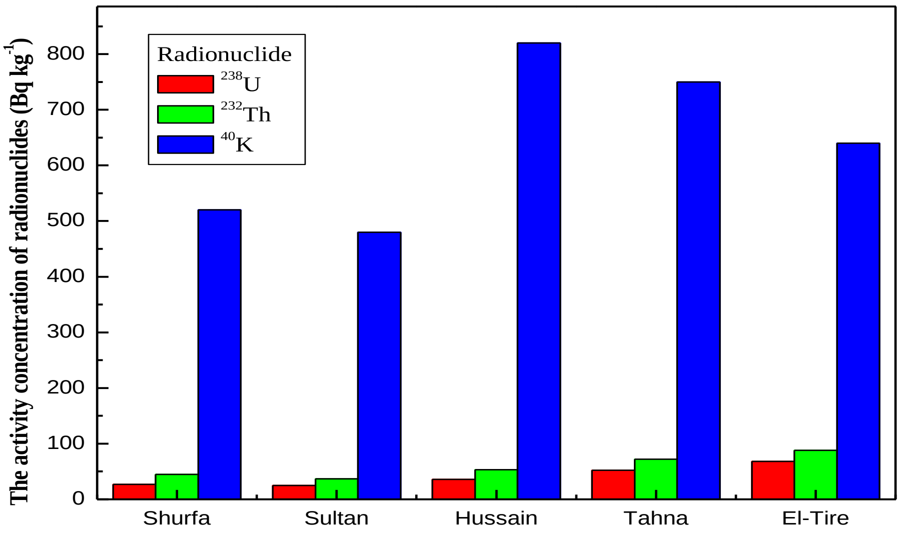

Fig (2): Activity concentrations of ^{238}U, ^{232}Th and ^{40}K ($Bq\ kg^{-1}$) in different quarries at El-Minya governorate

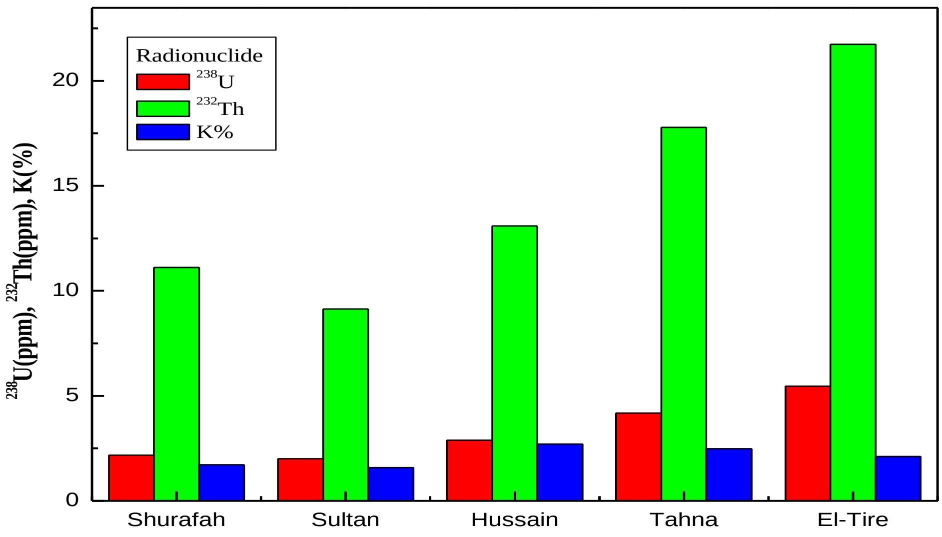

Fig. (3): Concentrations of ^{238}U (ppm), ^{232}Th (ppm) and K (%) in different quarries at El-Minya governorate.

Table 2 shows high concentration for Radon at Tahna El-Gabal, Nazlet Hussain and Zawyet Sultan above the action level which may possess a risk for workers, this is agree with a study done in India which found a higher concentrations of radon and thoron in granite quarries above the permissible levels and suggests radiation health effects on workers and public around the quarries(16).

As shown in table (3), there is a significantly increase in Silica levels in all areas under study. Sufficient exposure to silica can cause silicosis, a typical pneumoconiosis that develops insidiously after years of exposure. Exceptionally high exposure can cause acute or accelerated silicosis within months with significant impairment or death occurring within a few years. Exposure to silica is also associated with an increased risk of tuberculosis, lung cancer and of some autoimmune diseases, including scleroderma, systemic lupus erythematosus and rheumatoid arthritis. Freshly fractured silica dust appears to be more reactive and more hazardous than old or stale dust. This may be a consequence of a relatively higher surface charge on freshly formed particles(3).

As shown in Table (4), it is clear that there is a higher prevalence of Dysnea (76%) among workers and chest pain (32%) while it was 64% for cough symptoms. These results were comparable with results of another study in Nigeria, where the most common problems were occasional chest pain (47.6%), occasional cough (40.7%) the least problem was sputum mixed with blood (0.5%)(17). Also these results were comparable with results of previous studies in Iran and Rio de Janeiro, Brazil. The study in Iran reported irritative cough in 75% of the respondents(18), while that in Rio de Janeiro reported cough in 31.9% with expectoration in 41.7%(19). It therefore, seems reasonable to associate the respiratory problems recorded in the present study with the respirable quarry dust.

Data on the availability of medical care in the quarry industrial site and use of protective device by the quarry workers indicated that there was a lack of medical care in the study area and a near absence of protective device usage by the workers. This may account for the high prevalence of some of the respiratory problems reported in this study. Data from the present study suggest that chronic exposure to dust may increase susceptibility to respiratory problems and impaired lung function with cigarette smoking and increased length of service as additional predisposing risk factors. Suggested mitigating measures include provision of safety measures (*e.g.*, face masks, apron, and hand gloves), discouraging workers from cigarette smoking through public health education, frequent assessment of lung functions and redeployment of workers with severely reduced lung functions to other less hazardous occupations.

Table (1): The activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}) and the γ- radiation absorbed doses (D, nGyh^{-1}) as well as the annual effective dose equivalent (D_{eff}, mSvy^{-1}) in different quarries at El-Minya governorate.

Quarries sites	^{238}U		^{232}Th		^{40}K		D	D_{eff}
	Bq Kg^{-1}	ppm	Bq Kg^{-1}	ppm	Bq Kg^{-1}	%	nGyh^{-1}	mSvy^{-1}
Shurafah	27±0.5	2.169	45±0.8	11.115	520±9	1.714	66.396	0.0814
Zawyet Sultan	25±0.3	2.008	37±0.4	9.139	480±11	1.582	59.511	0.0729
Nazlet Hussain	36±0.6	2.892	53±0.9	13.091	820±13	2.703	83.739	0.1027
Tahna El-Gabal	52±0.7	4.177	72±1	17.784	750±12	2.472	100.011	0.1226
Gabal El-Tire	68±0.9	5.462	88±2	21.736	640±12	2.109	112.752	0.1383

Table (2): Radon levels in different quarries site in El-Minya governorate

Quarries site	Mean $\pm$ SD	P	Significance
Shurafah	4.88 $\pm$ 1.2	0.04618	NS
Zawyet Sultan	8.29 $\pm$ 2.3	0.00637	S
Nazlet Hussain	10.47 $\pm$ 2.7	0.0003	HS
Tahna El-Gabal	6.36 $\pm$ 1.9	0.00131	S
Gabal El-Tire	3.25 $\pm$ 0.8	0.04956	NS

N.B: * Action level for Radon is: (0.02 WL; 4 pCi/L).
S=Significant, NS=Non Significant, HS=Highly Significant.

Table (3): Silica levels in different quarries site in El-Minya governorate

Quarries site	Mean $\pm$ SD	P	Significance
Shurafah	626.2 $\pm$ 33.2	0.00268	S
Zawyet Sultan	725.4±36.3	0.00137	S
Nazlet Hussain	589.7±36.7	0.0381	NS
Tahna El-Gabal	855.8±40.3	0.00126	S
Gabal El-Tire	780.44±45.2	0.00149	S

N.B: * The control value is 15000 µgm / m^3, or 150 mgm / m^3,
or 0.015%. S=Significant, NS=Non Significant.

Table (4): Relationship between sample members and control groups in clinical examination;

Clinical Examination		Total			
		Sample members		Control group	
		Cases	%	Cases	%
Respiratory Symptoms	Cough	34	68.0	10	20.0
	Phlegm	32	64.0	2	4.0
	Dyspnea	38	76.0	6	12
	Wheezing	31	62.0	0	0.0
	Bronchitis	2	4.0	0	0.0
	Bronchial Asthma	25	50.0	5	10
	Chest Pain	16	32.0	2	4.0
	Dizziness	10	20.0	5	10.0
	Palpitation	16	32.0	4	8.0
	Creptation	3	6.0	0	0.0
	Low Breathing sound	3	6.0	1	2.0
	Breathlessness	20	40.0	2	4.0
	Respiratory Tract Infection	20	40.0	3	6.0
Non Respiratory Symptoms	Conjunctivitis	18	36.0	0	0.0
	Skin Cracks	34	68.0	0	0.0
	Skin Diseases	7	14.0	1	2.0

P value = 0.00165 (significant)

CONCLUSIONS

The highest activity concentrations of ^{238}U and ^{232}Th were found in Gabal El-Tire while ^{40}K was found in Nazlet Hussain. The lowest activity concentrations of ^{238}U, ^{232}Th and ^{40}K were recorded in Zawyet Sultan.

From environmental point of view, results indicated that quarries have human impacts. There are some potential risk of such industry has an effect on the environment, which requires attention, mitigations, and management to protect the existing human health.

RECOMMENDATIONS

Based on this study, it is recommended that:

1- Quarries must be properly inspected and maintained to safeguard people's health and safety.

2- Quarries management strategy should be implemented to avoid the negative impacts of dust contamination.

3- All workers in quarry plants should use face masks and possibly thermoluminescent dosemeters (TLDs) to protect themselves from the dangers of radioactive contamination.

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