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A Comparative Study of Field Gamma-ray Spectrometry by *NaI(Tl)* and *HPGe* Detectors in the South Caspian Region

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ABSTRACT

Natural radionuclides present in soil as well as certain anthropogenic radionuclides released to the environment are the major contributors to terrestrial outdoor exposures. In the assessment of human exposures from environmental radioactivity, besides the conventional method of soil and vegetation sampling combined with laboratory based analyses of environmental media, the other choice would be field spectrometry which is a rapid, efficient and economical means of identification of radionuclides in the environment. Newly developed high resolution solid state gamma-ray detectors provide a state of art means for such a purpose. However, they are relatively expensive, may not provide the highest intrinsic efficiency possible and their use is complicated by the need for cryogenic cooling of the detector. Scintillation detector spectrometry systems are considered to be capable of yielding satisfactory results particularly for natural background measurements at a fraction of cost. This paper describes a comparative study on application of *NaI(Tl)* scintillation and *HPGe* solid state systems for *in-situ* measurements of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs soil inventories at selected regions on the south coast of Caspian Sea, along with the results from laboratory analyses of collected soil samples in the area. Based on *in-situ* measurement results and field experience, it is concluded that *NaI(Tl)* spectrometry system provide satisfactory results which might be even improved by incorporating special spectrum analysis techniques, is relatively less expensive and is operationally easier to carry out than either *HPGe* system or direct laboratory based analyses of soil samples.

Keywords: *Field gamma-ray spectrometry, Gamma in-situ measurements, HPGe detector, NaI(Tl) detector, Natural radionuclides, ^{137}Cs.*

INTRODUCTION

Environmental radioactivity includes natural and anthropogenic sources. Natural sources consist of cosmic radiation, cosmogenic radionuclides and inventories of primordial radionuclides in the earth's crust. The latter category consists of singly occurring primordial radionuclides such as ^{40}K and ^{87}Rb, and decay series of primordial origin two of which identified by the long-lived parents ^{238}U and ^{232}Th contribute appreciably to human exposure to natural radiation. Worldwide studies have been carried out to determine population exposures from natural background. Level of terrestrial environmental radiation depends on geological composition of the bed rocks, type of soil, and geographical conditions, whereas the cosmic radiation contribution varies with elevation and latitude (NCRP, 1987; UNSCEAR, 2000; McLaughlin et al., 2005). The isotopes of interest are ^{226}Ra, ^{232}Th and ^{40}K, among which ^{226}Ra is a radionuclide in the ^{238}U series (Ateba, 2010; Petrinec *et al.* 2010). The ^{226}Ra nuclide is often chosen in the majority of the published papers because the external exposure to the population is mostly by gamma rays emitted from its two main daughters, namely ^{214}Pb (295.2 and 351.9 keV) and ^{214}Bi (609.3 keV) (Beck & Planque, 1968; Saito & Jacob, 1995).

In addition to naturally occurring radionuclides, ^{137}Cs which is an anthropogenic radionuclide might also contribute to human external exposure. Cesium-137 has been released to the environment mainly through atmospheric testing of nuclear arms and accidents in nuclear installations. This relatively high-yield fission product due to its relatively long half-life (~30y), biogeochemical behavior in the environment, and its 662 keV gamma-ray is of great concern among other released radionuclides to the biosphere (Faw & Shultis, 1993; Cooper et al., 2003; NCRP, 2006).

The conventional method of assessing the distribution of environmental radioactivity is by collection of soil and vegetation samples combined with laboratory based analyses of environmental media. This method suffers from the disadvantages associated with the representative nature of the sample or samples, access, coverage, time required for laboratory analyses and delays in obtaining results. Real time *in-situ* measurements, on the other hand, bring immediate benefits to survey for the purpose of prospecting, baseline monitoring and contamination mapping. In addition, a detector based at 1 m above the ground will typically measure 4×10^4 times more soil than a soil core of 10 cm diameter and about 30 cm depth (Beck et al., 1972; Zombori et al., 1995; Tyler, 2008). Field gamma-ray spectrometry provides a convenient method for the determination of contamination levels following an accidental release of radionuclides to the environment.

While field spectrometry can be performed with hyperpure germanium (*HPGe*) spectrometry systems, which provide the highest Peak-to Compton ratio and resolution, their use is complicated by the need for cryogenic cooling of the detector. They must be kept constantly cooled at 77 K with liquid nitrogen or electromechanical coolers. The size of the cryostat largely dictates the portability as well as the longevity of the system between re-fills, whereas power supply issues required by electromechanical systems remain a barrier for their convenient implementation (East *et al.*, 1982; Gilmore, 2008; Tyler, 2008). *NaI(Tl)* scintillation detectors have the advantage of being more robust and thus relatively more portable, and can be manufactured in a range of sizes and consequently able to provide substantially higher relative detection efficiencies (Tyler, 2008). Furthermore, one should consider the cost of *HPGe* systems which are more than 10 times higher than the *NaI(Tl)* spectrometry systems (Gilmore, 2008).

For environmental purposes, there are distinct advantages and value when the results from different techniques are comparable. In our study, *in-situ* measurements by sodium-iodide [*NaI(Tl)*] scintillation-type spectrometry system have been compared with results obtained from the application of a high resolution hyperpure germanium [*HPGe*] semiconductor-type system at exactly the same locations, and the results from laboratory analyses of collected soil samples at selected regions of the Iranian northern province of Guilan on the south coast of Caspian Sea. The aim was to assess the capability of *NaI(Tl)* portable spectrometry system for widespread measurement of soil inventories of certain natural radionuclides and ^{137}Cs as the first stage of a comprehensive monitoring of environmental radioactivity in the South Caspian region based on field gamma-ray spectrometry. The overall objective is to provide a baseline of radioactivity for both natural and anthropogenic radionuclides and improve the essential role of monitoring changes in levels of radiation and their potential impact on the health of human populations. The baseline maps would also enable the rapid impact assessment of nuclear accidents (Cooper et al., 2003).

MATERIALS AND METHODS

Study Area

The study area is the north Iranian province of Guilan, which is located along the south Caspian Sea shore between 36.59° to 38.45° North and 48.54° to 50.61° East, with an area of about 14000 km^2 and a population of around 2.5 million. The long-term average rainfall at the coast (-20 m a.s.l) is approximately 1,000 mm, while at the base of high mountain range, only several km from the sea it is approximately 1,200 mm and increases to around 2,000 mm at the summit (~3000 m a.s.l), which brought a moderate climate and a unique ecosystem to the region completely different than the central Iranian Plateau. Soil in the study area exhibits great diversity both in type and extension. The soil type in the coastal plain (mainly paddy fields) is humic gley, half-bog soil and alluvial, while is composed of brown mostly acidic soil at higher altitudes, which are mainly covered by natural deciduous forests (Hakimian, 1977). Two regions were selected for the comparative study in central and northern Guilan, namely Saravan and Paresar, respectively, as shown in Figure. 1. According to previous studies, these two regions are associated with higher soil inventories of ^{137}Cs.

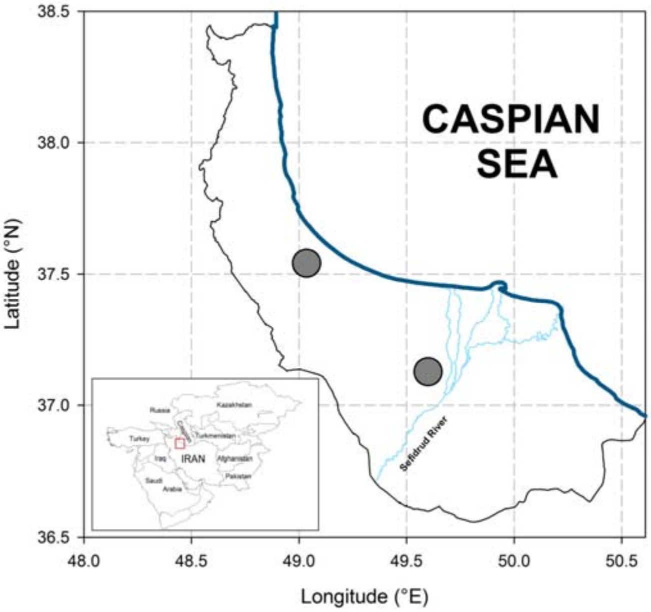

Fig 1. Selected study regions in Guilan.

In-situ gamma spectrometry

In-situ gamma-spectrometry was performed in both regions using *NaI(Tl)* and *HPGe* spectrometry systems. The standard reference height of 1 m above the ground was used in all cases using both tripod-mounted detectors.

The scintillation system used comprises a 76 mm × 76 mm diameter Scionix *NaI(Tl)* scintillation detector in conjunction with a battery powered multichannel analyzer for spectral analysis along with a laptop computer. The detector has a resolution of about 8% at 662 keV of ^{137}Cs.

The suggested window settings by the IAEA (IAEA, 1989) are shown in Table 1.

Table 1. Recommended window settings for a NaI(Tl) gamma-ray spectrometer (IAEA, 1989)

Energy window	Detected nuclide	Recommended window limits (keV)
K	^{40}K	1370-1570
U(^{226}Ra)	^{214}Bi	1660-1860
Th	^{208}Tl	2400-2800

Full-energy peaks of concern are 1.46 MeV for ^{40}K, 1.76 MeV for ^{226}Ra (^{214}Bi), 2.62 MeV for ^{232}Th (^{208}Tl) and 662 keV for ^{137}Cs (Abdi *et al.*, 2009; Isinkaye & Shitta, 2010).

The semiconductor system consists of a Canberra Eurisys portable HPGe detector along with its cooling system, MCA, and laptop computer. The p-type germanium detector has a relative efficiency of 25% with an energy resolution of 1.95 keV at 1.33 MeV of ^{60}Co.

The calibration process comprises the determination of the factors that relate the count rate under a photopeak to soil radioactivity concentration and the dose rate in air which was carried out according to the method developed by Beck *et al.* (1972), Beck (1980), Helfer and Miller (1988), Miller and Shebell (1993) and ICRU (1994) through another project (Fattahi, 2005).

Soil Sampling and Analysis

Soil sampling had been carried out in more than 50 stations throughout the province, shown in Figure 2, in a previous project. Undisturbed-soil sampling sites were chosen in line with population distribution and far from any obstructions.

Split-level sampling was carried out to a depth of 20 cm. Each core was divided into 5 cm intervals and combined samples were collected from three sampling points one meter apart. The top 5 cm section included the organic matter layer for the open field samples and the litter layer for the woodland soils. Coarse twigs were removed from the woodland soils before sampling. Under canopy samples were taken in each case from the interior of forest, under foliage and away from the bases of trees. Soil samples were air-dried at room temperature for several days, grounded following removal of stones larger than 2 mm and finally dried at 85 °C for 24 h. Subsamples were placed in cylindrical containers with the same geometry as the matrices used for efficiency calibration and kept for more than three weeks before measurement for the sake of secular equilibrium in radioactive series.

The activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th, and ^{137}Cs in dried soil samples were determined by non-destructive γ-spectrometry using a high-resolution HPGe detector system with an energy resolution of 2.0 keV at energy of 1332 keV gamma from ^{60}Co and an efficiency of 40% relative to a 7.62 × 7.62 cm diameter *NaI(Tl)* detector.

Specific activities measurements were carried out using the following photo peaks: 1460 keV for ^{40}K, 609 keV of ^{214}Bi and 352 keV of ^{214}Pb for ^{226}Ra, 583 keV of ^{208}Tl and 911 keV of ^{228}Ac for ^{232}Th and 662 keV of ^{137m}Ba for ^{137}Cs (Klement, 1982; Garcia & Madurga, 1990; MARLAP, 2004; EPA, 2006). Efficiency calibration of the gamma spectrometer was performed by experimental method to calculate efficiencies over the corresponding range of gamma-ray energies.

RESULTS AND DISCUSSION

In-situ measurements by both spectrometry systems were carried out in the study regions during summer 2009. Collected spectra have been analyzed in order to determine the special activities for radionuclides of concern. A typical spectrum collected by *NaI(Tl)* scintillation detector is shown in Figure 3. When the source geometry is taken into account, the concentrations or inventories of these radionuclides in the soil can be inferred along with the contribution to the above ground exposure rate by the method described in the previous section.

Specific activities of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs inferred from collected spectra in study regions through calculations based on efficiency calibrations, by *NaI(Tl)* and *HPGe* systems are presented in Tables 2 and 3, respectively. The results from laboratory analyses of collected soil samples throughout Guilan Province along with mean values and ranges of estimated soil specific-activities for concerned radionuclides in Iran and world average values extracted from UNSCEAR (2000) are given in Table 4.

Specific activities of ^{40}K have been determined using *full energy peak* and *energy band* methods (Tyler, 2008) through 1.46 MeV gammas from the decay of ^{40}K to ^{40}Ar.

Specific activities of ^{226}Ra have been determined using the same methods as described for ^{40}K through 1.76 MeV gammas from the decay of ^{214}Bi (a daughter product of ^{226}Ra). The case for ^{232}Th is through 2.61 MeV gammas from the decay of ^{208}Tl (a member of ^{232}Th decay series), and for ^{137}Cs through 662 keV gammas of its daughter product ^{137m}Ba.

While presented results in Tables 2 and 3 show a better precision for *HPGe* results due to its higher energy resolution, the specific activity values inferred from *NaI(Tl)* system are quite compatible and in the range of activities measured through laboratory-based analyses of soil samples. The activity values for Iran and world average values are just presented for the case of a general comparison. It should be noted that the higher inventory values of ^{137}Cs in the study regions and Guilan Province might be due to both the higher precipitation rate in the south Caspian region and possibly Chernobyl depositions.

In the process of *NaI(Tl)* spectrum analysis one should notice the possibility of peak interferences due to low resolution characteristics of the detector, for instance from ^{134}Te and ^{214}Bi peaks in the case of ^{137}Cs. There are, of course, specially developed procedures for quality spectral processing (Miller, 1997; Tyler, 2008), which is planned to be followed during comprehensive survey stage of nvironmental radioactivity in the south Caspian region.

Examining the results of this pilot study along with the practical experience gained in the process of field gamma spectrometry in the selected sites leads us to the conclusion that by carful application of *NaI(Tl)* spectrometry system we can achieve reasonable and satisfactory results as far as certain natural radionuclides and ^{137}Cs are concerned. It provides a measure which is operationally easier to carry out than the *HPGe* system, it is economical, efficient and less time-consuming with respect to soil and vegetation sampling combined with laboratory based analyses of environmental media.

Fig 2. Distribution of soil-sampling sites.

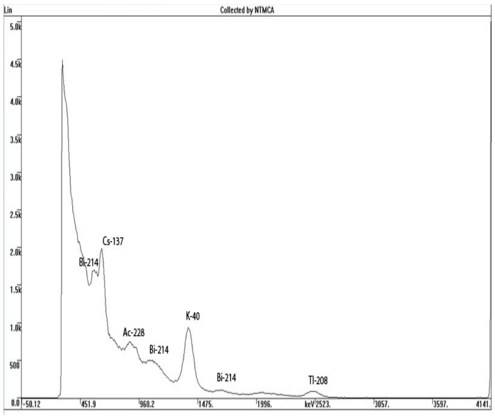

Fig 3. A typical spectrum collected by NaI(Tl) scintillation spectrometer.

Table 2. Soil specific-activities of concerned radionuclides in study regions measured by *NaI(Tl)* spectrometer.

Region	Specific Activity (Bq kg^{-1})			
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Saravan	346 ± 106	25 ± 4	34 ± 5	35 ± 6
Paresar	430 ± 112	20 ± 3	22 ± 8	40 ± 8

Table 3. Soil specific-activities of concerned radionuclides in study regions measured by *HPGe* spectrometer.

Region	Specific Activity (Bq kg^{-1})			
	^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Saravan	295 ± 17	23 ± 3	30 ± 4	30 ± 4
Paresar	328 ± 18	19 ± 3	18 ± 3	33 ± 3

Table 4. Specific-activities of concerned radionuclides measured through laboratory-based analyses of soil samples.

Region		Specific Activity (Bq kg^{-1})			
		^{40}K	^{226}Ra	^{232}Th	^{137}Cs
Guilan Province	Mean	548 ± 138	24 ± 6	26 ± 11	21 ± 10
	Range	285 - 851	7 - 38	5 - 47	3 - 60
Iran*	Mean	640	28	22	-
	Range	250 - 980	8 - 55	5 - 42	-
World*	Mean	420	33	45	-

**Adapted from UNSCEAR (2000)*

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مقایسه بیناب سنجی میدانی پرتوهای گاما توسط آشکارسازهای یدور سدیم و ژرمانیوم فوق خالص در منطقه جنوب خزر

ع. صدرممتاز، م. وهابی مقدم، س. خوشبین فر و ع. مقدسی

چکیده

هسته های پرتوزای طبیعی موجود در خاک و نیز برخی هسته های پرتوزای مصنوعی رها شده به محیط، بخش عمده پرتوگیری زمینی را موجب می شوند. در ارزیابی پرتوگیری بشر از پرتوزایی محیطی، غیر از روش سُنتی نمونه برداری از خاک و پوشش گیاهی و آنالیز آزمایشگاهی متعاقب آن، گزینه ی دیگر انجام بیناب سنجی میدانی (در مکان) است که شیوه ای سریع، مؤثر و اقتصادی جهت تعیین موجودی هسته های پرتوزا در محیط بشمار می رود. آشکارسازهای ژرمانیوم فوق خالص با توان تفکیک زیاد که اخیراً توسعه یافته اند بدین منظور مناسب به نظر می رسند؛ امّا نسبتاً گرانند، معمولاً از بالاترین بازده ذاتی برخوردار نیستند و از نقطه نظر ضرورت تأمین برودت بهنگام بهره برداری مشکلات خاص خود را دارند. برای سنجش زمینه ی پرتوی طبیعی، سیستم های بیناب سنجی سوسوزن (سنتیلاسیون) با امکان دستیابی به نتایج رضایتبخش و قیمت بسیار کمتر مطرحند. این پژوهش به بررسی قیاسی بین دو سیستم سوسوزن NaI(Tl) و نیمه هادی HPGe در کاربرد بیناب سنجی میدانی به منظور تعیین موجودی هسته های پرتوزای ^{40}K، ^{226}Ra، ^{232}Th و ^{137}Cs در خاک مناطق برگزیده در ساحل جنوبی دریای خزر پرداخته است. نتایج آنالیز آزمایشگاهی نمونه های خاک منطقه نیز در این بررسی منظور شده است. تجارب میدانی و نتایج سنجش بعمل آمده حاکی از آنست که سیستم بیناب سنجی سوسوزن NaI(Tl) نتایج رضایتبخشی را بهمراه داشته است و با کاربرد نرم افزارهای ویژه ی آنالیز بیناب نتایج مطلوب تری نیز قابل حصول است. کاربرد میدانی این سیستم با وجود ارزانی نسبی از سهولت بیشتری در مقایسه با سیستم HPGe و آنالیز آزمایشگاهی نمونه های محیطی برخوردار است.