# Spatial variability of soil <sup>137</sup>Cs in the South Caspian region

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Abstract In a comprehensive program of environmental radioactivity survey in South Caspian region,<sup>137</sup>Cs inventories in soil has been measured at more than 50 sites in the Iranian northern province of Guilan. This has been the first wide-range survey of soil radionuclide inventories in the narrow band sensitive ecosystem of south Caspian shore. Radioactivity measurements were carried out using HPGe gamma-spectrometry system. The activity concentration of <sup>137</sup>Cs in surface soil exhibits a mean value of  $17.6\pm9.4$  Bq kg<sup>-1</sup>, with a range of 2.3–41.7 Bq kg<sup>-1</sup>. In many sites, split-level sampling method has been applied down to a depth of 20 cm. There were found generally two profiles. Most profiles exhibit a negative exponential distribution, while others revealed a clear subsurface peak in 5-10-cm layer. Cesium deposition in the study area has been estimated to be in the range of 0.38-2.9 kBq m<sup>-2</sup> with a mean value of 1.7 kBq  $m^{-2}$ . Distribution patterns of <sup>137</sup>Cs concentration levels and deposition values have been estimated using Kriging interpolation method. Observed hotspots in deposition pattern coincide with areas of higher precipitation.

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Present Address: S. Khoshbinfar School of Physics, Damghan University, Damghan, Iran e-mail: skhoshbinfar@du.ac.ir **Keywords** <sup>137</sup>Cs soil inventories  $\cdot$  South Caspian region  $\cdot$  Guilan Province  $\cdot$  Gamma spectrometry  $\cdot$ Spatial variability  $\cdot$  <sup>137</sup>Cs depth profile

#### Introduction

Substantial amounts of artificial radionuclides have been released to the environment as fallout due to atmospheric nuclear weapons testing conducted during 1950s and early 1960s. They are initially injected into the upper atmosphere and moved slowly to the lower atmosphere and then to the ground. This raised the radioactivity level of most places, mostly in northern hemisphere (UNSCEAR 2000). Moreover, Chernobyl accident in 1986 caused considerable radiological consequences in Ukraine, Belarus, Russia, and Europe which raised the concern on long-term effects on terrestrial environment.

Among the various radionuclides released to the environment, <sup>137</sup>Cs due to its higher fission yield, relatively long half-life of about 30 year, and long-term biological effects of its 662-keV gamma radiation are subjected to many studies worldwide (Ritchie and Ritchie 2005). The population-weighted cumulative deposition density of <sup>137</sup>Cs in northern hemisphere for the period of 1945–2000 is estimated at 81 kBq m<sup>-2</sup>, and the mean fallout per unit area in the northern hemisphere is estimated to be about 2 kBq m<sup>-2</sup> (UNSCEAR 2000, corrected to 2008). None of these values includes the contribution of

<sup>137</sup>Cs from Chernobyl accident. The exposure of the population living in contaminated areas depends directly on the vertical distribution of the gamma emitting radionuclides in soil column. Knowledge of distribution and the level of radionuclides in soil make a great improvement in risk assessment analysis (Nimis 1996). These data might be suitable for constructing local, regional, or even global databases. It is also helpful for study of radiological sensitivity of a given ecosystem and providing a better contamination picture for decision makers during or after occurrence of any future releases (Aarkrog 1979; Howard 2000). In this regard, the potential hazard due to the presence of numerous nuclear installations and test sites around the Caspian Sea should not be ignored (Froehlicha et al. 1999; Kostianoy 2010; Zaidi and Mustafaev 2004).

The study area is located in a sensitive natural ecosystem where no comprehensive study on <sup>137</sup>Cs inventories in soil and its atmospheric deposition pattern have been carried out. Having these points in mind, a wide-range environmental radioactivity survey program conducted in the northern Iranian province of Guilan since 2006. Study of <sup>137</sup>Cs was part of this program which included its soil inventories and vertical distribution in order to construct area distribution maps as well as atmospheric deposition patterns. Such data could be of special value for further atmospheric aerosol deposition studies in the region.

#### **Experimental methods**

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 14,000 km<sup>2</sup> and a population of around 2.5 million. The long-term average rainfall at the coast (-20 m above sea level (a.s.l)) is approximately 1,000 mm, while at the base of high mountain range, only several kilometers from the shore it is approximately 1,200 mm and increases to more than 1,400 mm at the summit (~3,000 m a.s.l), which brought a moderate climate and a unique ecosystem to the region completely different than the central Iranian Plateau (Fig. 1). Soil in the study area exhibits great diversity both in type and extension. The soil type in the coastal plain (mainly paddy fields) is low-humic



Fig. 1 Topography map of study area

gley, humic gley, half-bog soil, and alluvial. At higher altitudes, which are mainly covered by natural forests, lithosols, rendzinas, and podzols are dominant (Hakimian 1977).

In the case of surface soil sampling, top 5-cm soil samples were collected using hand-driven pipe corers and metal surface frames. However, in most cases, split-level sampling technique was applied down to a depth of 20 cm with 5-cm intervals. The surface frame was specially designed (detail of which is presented in Khoshbinfar 2010) to overcome the difficulty of collecting samples of constant thickness due to variable compaction of the highly organic horizons within forest canopies. Soil sampling has been carried out in more than 50 stations throughout the province since October 2006 (Fig. 2).

At each of the sampling sites, three replicate corers were taken to prepare combined samples for each depth interval. The top 5-cm section included the organic matter layers 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 of 300-g weights were placed in cylindrical containers with the same geometry as the





matrices used for efficiency calibration. The activity concentrations of <sup>137</sup>Cs in dried soil samples were determined by non-destructive  $\gamma$ -spectrometry using a high-resolution High-Purity Germanium (HPGe) detector systems with efficiencies of 20% and 40% relative to a  $7.62 \times 7.62$  cm diameter NaI(Tl) detector. Specific activities measurements were carried out using 662-keV photo peak. Efficiency calibration of the gamma spectrometer was performed by experimental method. Calibration sources used in this respect were International Atomic Energy Agency (IAEA) reference materials RGU-1, RGTH-1, RGK-1, and CERCA HM 395. Matrices filled with IAEA-326 and IAEA-327 reference soil materials were also used for the sake of quality assurance. Counting times per sample ranged from 12 to 48 h.

### Geostatistical treatment

In the study of environmental radioactivity, one practically samples in a limited number of points and may desire to get a broader picture of available budget of the quantity looking for its changes across the unsampled sites on the surface of study area. It is possible to create a continuous map using interpolation methods which the values in unknown points are estimated by known values. These predicted values are actually the weighted means of observations in their neighborhood (Almgren 2008; Piegorsch and Bailer 2005).

To build a suitable map representing the distribution pattern of deposition amount of the anthropogenic <sup>137</sup>Cs in soil, the geostatistical consideration should be taken into account. The procedure includes testing the possible existence of spatial autocorrelation in data followed by visualizing and quantifying, then applying a proper model to experimental Variogram and finally incorporating the results into an interpolation process to predict dose rate values (Khoshbinfar 2010). Among different geostatistical interpolation methods, it is suggested that the Kriging is reliable enough for environmental radioactivity interpretations which produces visually appealing maps from irregularly spaced data (Almgren et al. 2006; Mabit and Bernard 2007; Szegvary et al. 2007).

## **Results and discussion**

# Activity concentration of <sup>137</sup>Cs in soil

There have been so many studies of the vertical distribution of radionuclides in soils throughout the world. The split-level sampling is usually applied for such studies. Knowledge of migration parameters based on physical as well as bio-geochemical characteristics, of course, provides a detail understating of the radionuclide transport which in the light of powerful time dependent models, e.g., advection–diffusion equation (ADE) enables us to follow radionuclide fate inside soil. Hydrological and meteorological conditions are other influential parameters in this regard (Bossew and Kirchner 2004).

In Table 1, the variation in mean and range of <sup>137</sup>Csspecific activities collected from 51 sites throughout Guilan is shown. The current activity concentration of top soil layers indicates that over many years since the initial deposition, considerable amount of deposited cesium is still present in the surface layers.

The frequency distribution of measured specific activities is presented in Fig. 3 for surface soil (0-5 cm) and mean value over 20 cm depth.

By fitting mathematical models to our measured activity-depth distributions, we found two types of profiles; one which follows an exponentially decreasing trend (type A), and the other with a subsurface peak at a depth of several centimeters below the ground (type B). We adopted a simple exponential function (Isaksson and Erlandsson 1998):

$$A(z) = a \exp(-bz) \tag{1}$$

in the first case; where, A(z) is the <sup>137</sup>Cs concentration at depth z in cm, a and b are experimentally fitted parameters. Profiles of typical sites which follow this trend are demonstrated in Fig. 4. Though these profiles reveal a more pronounced diffusion vs. advection, however, it might be due to relatively large depth interval which in turn could have affected the precise determination of peak depth.

Gaussian distribution is adopted for the second case:

$$A(z) = a \exp\left[-0.5\left(\frac{z-z_0}{b}\right)^2\right]$$
(2)

where, A(z) is the <sup>137</sup>Cs concentration at depth z in centimeters, a is the specific activity of the top layer of soil, and the coefficient b which show the standard deviation ranges between 3.5 and 6.0 (Fig. 5). The first type profile belongs to sampling locations in the plains, whereas those with subsurface peaks are mostly from high elevated sampling locations. This might be pertained to the soil type of mountainous region which mostly contains higher clay mineral. Based on Guilan province soil-type map, alluvial is dominant in the plain while clay is more abundant at higher altitudes (Hakimian 1977).

To get a broader picture of the deposition distribution, variations across the unsampled sites have to be taken into consideration. Therefore, all measured <sup>137</sup>Cs surface activities were first geo-referred on Guilan grid map using Universal Transverse Mercator (UTM) coordinates. The study area is located in UTM 39N band. Contour map of radiocesium activity was then produced by ordinary Kriging interpolation method. In these calculations, we employed a linear model for Variogram analysis (Kulahcl et al. 2008). All these geostatistical analyses have been carried out using version 9 of SURFER software (SURFER 2009). By applying this procedure, <sup>137</sup>Cs surface (0–5 cm) activity distribution pattern inside the province is estimated, which is plotted in Fig. 6.

This figure indicates that several decades after the termination of atmospheric tests and few decades after Chernobyl accident, a considerable amount of <sup>137</sup>Cs is still found in the surface layer of soil in the region. Cesium-137 soil inventories are relatively higher in high elevated forest areas, which is due to orographic effects including occult deposition and

Table 1Mean and range ofspecific activities of 137Cs(Bq kg <sup>-1</sup> ) measured inGuilan soils		Soil layer, cm			
		0–5	5-10	10-15	15–20
	Mean±SD	17.6±9.4	19.3±9.9	$14.8 \pm 7.8$	7.3±4.1
	Range	2.3-41.7	2.4-45.5	1.6–30.2	1.2-20.1



Fig. 3 The frequency of measured specific activities of  $^{137}Cs$ : **a** for surface soil (0–5 cm) and **b** mean value over 20 cm depth

seeder–feeder mechanism (Fowler et al. 1998). A wide strip including areas of higher surface activities (>30 Bq kg<sup>-1</sup>) corresponds to brown forest soil type with a higher retention capability for deposited  $^{137}$ Cs. From radiological hazards point of view, these areas could be of special concern for the study of future deposition scenarios.

The activity concentration of  $^{137}$ Cs in surface soil exhibits a mean value of 17.6±9.4 Bq kg<sup>-1</sup>, with a range of 2.3–41.7 Bq kg<sup>-1</sup>. This range is compared with measured mid-latitude northern hemisphere soil inventories in Table 2. Such  $^{137}$ Cs surface-activity data would be of great importance as a basis of reference in the case of any future emergency releases in the Caspian Region.

**Fig. 4** Experimental fit of exponential decreasing profile at typical type A sites: 13, 15, 24, and 37. *Error bars* represent counting statistics





**Fig. 5** Experimental fit of Gaussian profile at typical type B sites: 23, 35, 45, and 46. *Error bars* represent counting statistics

Deposition pattern of <sup>137</sup>Cs

The main scavenging mechanism of  $^{137}$ Cs from the atmosphere is wet deposition. In contrast to Chernobyl deposition, global fallout can be assumed equally distributed in northern hemisphere with a general latitude dependency via precipitation in years since release in the atmosphere (UNSCEAR 2000). Studies have shown that the detailed variations in the deposition are closely connected to the precipitation and nature of ground roughness (Pálsson et al. 2006). Cesium-137 deposition in the study area is estimated to be in the range of 0.38–2.85 kBq m<sup>-2</sup> with a mean value of 1.69 kBq m<sup>-2</sup> (Khoshbinfar 2010). Table 3 presents the place of current study results in comparison to global fallout contribution in  $^{137}$ Cs deposition in Northern hemisphere, decay corrected to 2008.

Current estimated mean deposition is also in agreement with a previous measurement in a certain location of the province (Ebrahimi 1999). The predicted value of <sup>137</sup>Cs mean deposition in the region according to UNSCEAR (1982) is about 1082 Bq m<sup>-2</sup> (decay corrected for 2008). The deviation might be due to the exceptional higher precipitation in the study area as well as possible Chernobyl contribution. The estimated mean deposition value of Chernobyl <sup>137</sup>Cs in the northwestern part of the Caspian Sea (state of Dagestan in Russia) is approximately 2–3 kBq m<sup>-2</sup> (De Cort et al. 1998).

Applying the same procedure as in the case of surface-activity distribution, deposition pattern is estimated and plotted in Fig. 7. A comparison between <sup>137</sup>Cs deposition in Fig. 6 and mean annual precipitation maps presented by local meteorological authority (IMO 2009) indicates that hotspots are generally located in the areas of high annual precipitation (>1,000 mm year<sup>-1</sup>).

The long-term mean annual precipitation in the study area is well above the mean value for Iranian Plateau. Since wet deposition is considered to be the main scavenging mechanism from the atmosphere, it would be of interest to look for a correlation between the amount of deposited <sup>137</sup>Cs and annual precipitation in the region. The kind of correlation found for representative sites (due to limited close weather and sampling stations) is illustrated in Fig. 8.



Fig. 6 Cs-137 concentration in the surface soil of Guilan Province (UTM zone 39N)

Table 2	Report	ed values c	of globa	l fallout	contribu	ution	in <sup>11</sup>	<sup>37</sup> Cs
surface-a	ctivity	concentrati	on, dec	ay corre	cted to	year 2	200	8

Country	<sup>137</sup> Cs concentration (Bq kg <sup>-1</sup> )	Reference
France	0.49–24.5	Roussel-Debel et al. (2007)
Japan	33.5(mean value)	Takenaka et al. (1998)
Korea, a	6.21-55.4	Kim et al. (1998)
Korea, b	9.4-43.4	Lee and Lee (1997)
Lithuania	3.84-7.68	Luksien et al. (2006)
Turkey	2.8–76.2	Varinliog Lu and Kose (2005)
Current study	2.3–42.7	

Table 3Cs-137deposition values in various countries, decaycorrected to year 2008

Country	<sup>137</sup> Cs deposition (kBq m <sup>-2</sup> )	Reference
Austria	1.96	Bossew et al. (2001)
Canada	1.86	Blagoeva and Zikovsky (1995)
China	0.86	Tang et al. (2002)
Germany	2.3	Bunzl and Kracke (1988)
Hungary	1.47	Barisic et al. (1999)
Sweden, a	1.35	Isaksson et al. (2001)
Sweden, b	1.10	Isaksson and Erlandsson (1998)
Turkey	0.10-1.44	Karadeniz and Yaprak (2008)
Vietnam	0.96	Quang et al. (2004)
Current study	1.69 (0.38-2.84)	

**Fig. 7** Cs-137 deposition pattern in Guilan Province (UTM zone 39N)



## Conclusion

Fig. 8 The correlation

tion in Guilan Province

between <sup>137</sup>Cs deposition

and mean annual precipita-

The soil concentration and inventory of the anthropogenic  $^{137}$ Cs has been estimated on the basis of their inventory measurements at more than 50 stations in

the Iranian Northern province of Guilan on the southwest Caspian coast. Split-level soil sampling technique was applied and radioactivity analysis has been carried out using HPGe gamma-spectrometry systems. It is shown that the deposition pattern correlates



with the long-term mean annual precipitation in the study area. Soil inventory profiles are also analyzed and the trends are associated with exponential and Gaussian distribution functions fitted to experimental data. Moreover, <sup>137</sup>Cs provincial surface activity and inventory maps are provided using Kriging interpolation method.

The ranges of <sup>137</sup>Cs concentration at successive 5-cm layers are between 2.3–41.7, 2.4–45.5, 1.6–30.2, and 1.2–20.1 Bq kg<sup>-1</sup>, respectively. Their corresponding mean values are  $17.6\pm9.4$ ,  $19.3\pm9.9$ ,  $14.8\pm7.8$  and  $7.3\pm4.1$  Bq kg<sup>-1</sup>. The measured <sup>137</sup>Cs deposition is estimated to be in the range of 0.38–2.85 kBq m<sup>-2</sup> with a mean value of 1.69 kBq m<sup>-2</sup>.

This has been the first comprehensive study of <sup>137</sup>Cs soil inventories in Guilan Province, which is of great importance in evaluation and control of public exposure and provision of a basis for identifying changes in case of any future release in the region.

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