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Article

Vertical migration of ^{137}Cs in the South Caspian soil

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ABSTRACT **In order to study the vertical migration of anthropogenic ^{137}Cs, soil inventories of this radionuclide were measured in two regions selected on the basis of a previous comprehensive survey in the northern Iranian province of Guilan located in the South Caspian region. Ten sampling stations were randomly chosen in these regions and split-level sampling was carried out to a depth of 30 cm. Sample analysis was performed using a HPGe detector system. *In situ* gamma measurements in both regions were also carried out with the aid of a portable germanium spectrometer. The experimental data were then compared with the solution of the convection-dispersion equation (CDE) with the proper initial and boundary conditions to evaluate initial deposition as well as transport parameter values. The solution, including the effects of both considered sources, *i.e.* global fallout and Chernobyl releases, fits the measured data well. The effective convection velocity and dispersion coefficients of ^{137}Cs lie in the range of 0-0.25 cm·y^{-1} and 0.32-0.75 cm^2·y^{-1}, respectively, indicating a very slow migration rate in the area. Most of the deposited ^{137}Cs still remained in the top 10-cm layer. Moreover, the fitted depth profiles were then employed to correct the surface activities of ^{137}Cs estimated by *in situ* measurements.**

Keywords: **^{137}Cs / soil migration / spectrometry, gamma / transport parameters**

Introduction

Knowledge of radionuclide migration in soil is of great importance both for the assessment of radiological sensitivity of a given ecosystem and their long-time behavior in the environment (Aarkrog, 1979; Howard *et al.*, 2002; Wicker *et al.*, 1999). There exist two major release sources of anthropogenic radionuclides into the atmosphere. The first one came from Nuclear Weapon Tests (NWF) that occurred between 1945 and 1980 with a peak around 1965, and the second, from the accident at Chernobyl nuclear power plant (Ch) on 26 April 1986 (UNSCEAR, 2000). Wet deposition was the main scavenging process of released radionuclides from the atmosphere. Most of the NWF atmospheric deposition took place in the mid-latitude band, compared with the Chernobyl disaster, which had only deposited whenever the passage of radioactive cloud was accompanied by a

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precipitation (Almgren *et al*., 2009). Over decades, the soil became a reservoir of ground-deposited man-made radionuclides. Among the released radionuclides, ^{137}Cs attracted much interest from the international community of scientists due to its special features, including a relatively high abundance, long half-life (~30 y), bioactivity and convenient gamma energy for measurement (662 keV). The ^{137}Cs migration in soil is a complicated process and depends on many factors, *e.g*. the soil type and texture, its pH, physicochemical interactions with soil and sorption mechanism (Arapis *et al*., 1997; Chibowski and Zygmunt, 2002; NCRP, 2006). Its vertical migration in soils has a direct effect on the level of external radiation exposure to people living in contaminated areas. The amount and distribution of radioactivity in the plant rooting zone affects the transfer of radioactivity to the food chain, and the ^{137}Cs migration rate can also influence its removal by surface water or penetration into the water column (Ehlken and Kirchner, 2002). Several models have been developed to describe ^{137}Cs migration in soils and to explain its vertical distribution. Among them, many analytical solutions of the convection-dispersion equation (CDE) which are different in initial and/or boundary conditions and physicochemical processes have been proposed (Ivanov *et al*., 1997; Konshin, 1992; Likar *et al*., 2001; Smith and Elder, 1999; Schuller *et al*., 1997; Szerbin *et al*., 1999).

Applicability of portable *in situ* gamma spectrometry to acquire ^{137}Cs inventory measurement in the field has gained increasing interest and offers a number of significant advantages. These include relatively rapid field measurement, which could either replace the collection of soil samples for subsequent laboratory counting or could be of use in a survey as a guide for a detail sampling strategy. It could also provide more representative results due to the greater area covered by this method. Because of the larger sample masses associated with direct field measurement, the time duration required by *in situ* ^{137}Cs measurements is often significantly shorter than that required by laboratory analysis (Beck, 1972; Tyler *et al*., 1996). So, it provides a quick, relatively accurate and robust strategy for determination of gamma-emitting radionuclides located in soil. *In situ* measurement of ^{137}Cs activity using portable detectors will be influenced by *a priori* assumptions about the distribution of radionuclides in the soil, which is considered to be an important source of uncertainties in activity concentration estimates and dose rate evaluations (Beck, 1972; Miller and Helfer, 1985). It was proposed by Fink (1992) that in field gamma spectrometry, one can first measure the surface activity and then calculate its corrected value for the real distribution by a correction factor. This was applied successfully by Almgren and Isaksson (2006).

In the present study, with the aim of improving the knowledge on vertical migration of NWF fallout as well as Chernobyl ^{137}Cs in South Caspian soil,

10 sampling sites were studied in two regions selected from more than 50 sites, which showed higher ^{137}Cs inventories in a previous survey of cesium levels in the northern Iranian province of Guilan. *In situ* gamma spectrometry was also carried out in both regions. The transport parameters as well as initial fallout contribution were then estimated through comparison of the CDE solution and field data. Moreover, the applicability of ^{137}Cs activity estimation using *in situ* measurement corrected by the actual distribution was investigated.

1. Transport parameters

For one-dimensional CDE transport of a solute, one might solve the following equation:

$$\frac{\partial C(z,t)}{\partial t} = \frac{\partial^2}{\partial t^2}\left(D\,C(z,t)\right) - \frac{\partial}{\partial t}\left(V\,C(z,t)\right) - \lambda\,C(z,t) \qquad (1)$$

where $C(z,t)$ is the concentration in soil solution, z is the distance from the surface and D and V are the dispersion coefficient and average pore water velocity, respectively. For constant values of D and V and local equilibrium conditions, total ^{137}Cs concentration (sorbed and mobile) could be derived from the solution of the following integral (Konshin, 1992; Schuller *et al*., 1997; Likar *et al*., 2001; Bossew and Kirchner, 2004):

$$C(z,t) = \int_0^{\infty} \left(\frac{1}{\sqrt{\pi D_{eff}\,\tau}}\, e^{-\frac{\left(z - V_{eff}\,\tau\right)^2}{4 D_{eff}\,\tau}} - \frac{V_{eff}}{2D_{eff}}\, e^{\frac{z V_{eff}}{D_{eff}}} \left(1 - erf\left(\frac{z + V_{eff}\,\tau}{2\sqrt{D_{eff}\,\tau}}\right)\right)\right) f(0,t')\, \mathrm{e}^{-\lambda\tau}\, dt' \qquad (2)$$

where $f(0,t')$ represents the fallout function, $\tau = t - t'$, λ is the decay constant of ^{137}Cs in years and $erf(z,t)$ is the error function. The new coefficients of D_{eff} and V_{eff} are called effective convection velocity and dispersion coefficients, respectively. To evaluate the relevant transport parameters from the field measurements of ^{137}Cs activities at each site, equation (2) should be solved for a given fallout profile. For both release sources, $f(0,t')$ could be approximated by the Dirac-Delta function centered at a peak fallout time quite accurately. Substitution of this condition in equation (2) leads to the final expression:

$$C(z,t) = J_0\, \mathrm{e}^{-\lambda\tau} \left(\frac{1}{\sqrt{\pi D_{eff}\,\tau}}\, e^{-\frac{\left(z - V_{eff}\,\tau\right)^2}{4 D_{eff}\,\tau}} - \frac{V_{eff}}{2D_{eff}}\, e^{\frac{z V_{eff}}{D_{eff}}} \left(1 - erf\left(\frac{z + V_{eff}\,\tau}{2\sqrt{D_{eff}\,\tau}}\right)\right) \right) \qquad (3)$$

where J_0 stands for initial deposition in Bq.m^{-2}. Time origins are taken as the 1st of January 1965 for nuclear atmospheric tests, and 5th of May 1986, when the Chernobyl plume passed over our study area. According to recently published

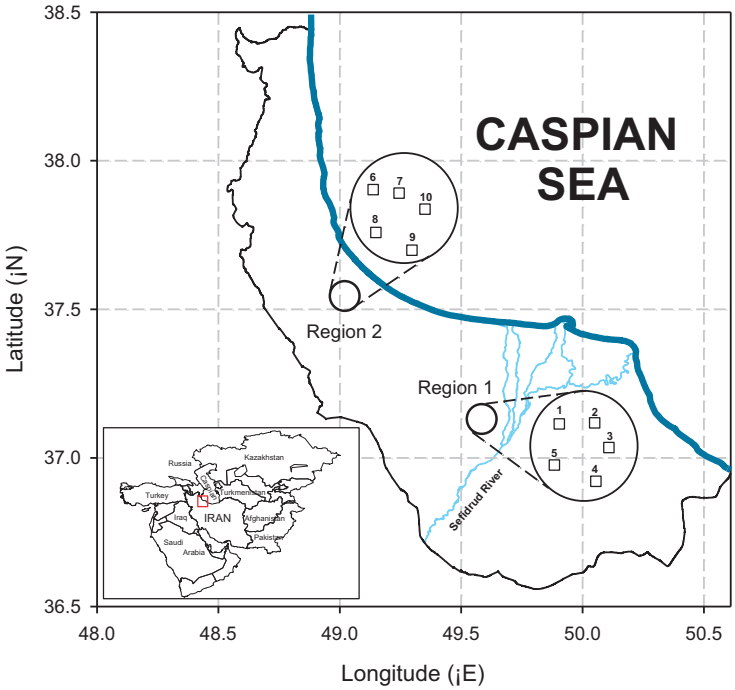

Figure 1 – ***Location of selected regions and sampling sites in Guilan.***

simulation data, a weak front of the Chernobyl plume arrived in the South-West Caspian region from North and North-West directions around 5 May 1986 (Brandt *et al*., 2000; Quelo *et al*., 2007), and local provincial weather stations recorded scattered precipitations in the same period (IRIMO, 1986). For general solution we add the contribution of both sources:

$$C(z,t) = C\left(z,t\right)_{NWF} + C\left(z,t\right)_{Ch}. \tag{4}$$

2. Experimental methods

2.1. The study area

The study area is the northern Iranian province of Guilan, which is located along the South Caspian seashore between 36.6° to 38.5° North and 48.5° to 50.6° East (Fig. 1). The province is divided into plain and highland areas that are mostly covered by native species of natural deciduous forests and is characterized by a warm, temperate and humid climate. The long-term average rainfall on the coast

(−20 m a.s.l.) is approximately 1 000 mm, while at the base of the high mountain range, several km from the sea, it is approximately 1 200 mm and increases to around 2 000 mm at the summit (>2 000 m a.s.l.). The soil type in the coastal plain is humic gley, half-bog soil and alluvial, while is composed of brown, mostly acidic forest soil in the highlands (Hakimian, 1977).

2.2. Soil sampling and analysis

During spring of 2009, soil samples were collected for gamma analysis. Sampling sites are located in selected regions of central and northern Guilan. According to previous studies, these two regions are associated with higher soil inventories of ^{137}Cs. Split-level sampling was carried out in 5 randomly chosen locations at each region using a stainless steel corer (85 mm in diameter) to a depth of 30 cm. Each core was divided into 0-2, 2-4, 4-6, 6-9, 9-12, 12-15, 15-20, 20-25 and 25-30 cm depth intervals.

Soil samples were air-dried at room temperature for several days, ground following removal of stones larger than 2 mm, and finally dried at 85 °C for 24 h. Sub-samples were placed in cylindrical containers with the same geometry as the matrices used for efficiency calibration. The specific activities of ^{137}Cs in dried soil samples were determined by non-destructive γ-spectrometry through its 662 keV gamma using a high-resolution HPGe detector. The detector had a relative efficiency of 80% and a resolution of 1.2 keV at 1.33 MeV. Counting time per sample ranged from 60 000 to 80 000 seconds.

2.3. In situ gamma spectrometry

In situ γ-spectrometry was performed in both regions, along with soil sampling, at 1 m above the ground using a Canberra Eurisys portable HPGe system. The p-type germanium detector has a relative efficiency of 25% with an energy resolution of 1.95 keV at 1.33 MeV. System calibration had been carried out according to standard procedures (Fattahi, 2005). Following the method described by Fink (1992), ^{137}Cs activities in Bq.cm^{-2} for the surface plane source could be calculated using the ^{137}Cs net count rate and detector calibration coefficients, then corrected for the actual distribution of ^{137}Cs found from the soil sampling practice. The Correction Factor (CF) is derived through the ratios of plane source to the actual vertical distribution of primary photon fluence rates:

$$A(\mathrm{Bq.cm^{-2}})_{Actual\ \ Dist.} = A(\mathrm{Bq.cm^{-2}})_{Plane\ \ source}\ CF\ ; \qquad CF = \frac{\varphi_{\mathrm{Plane\ \ source}}}{\varphi_{Actual\ \ \mathrm{Dist.}}}\cdot \qquad (5)$$

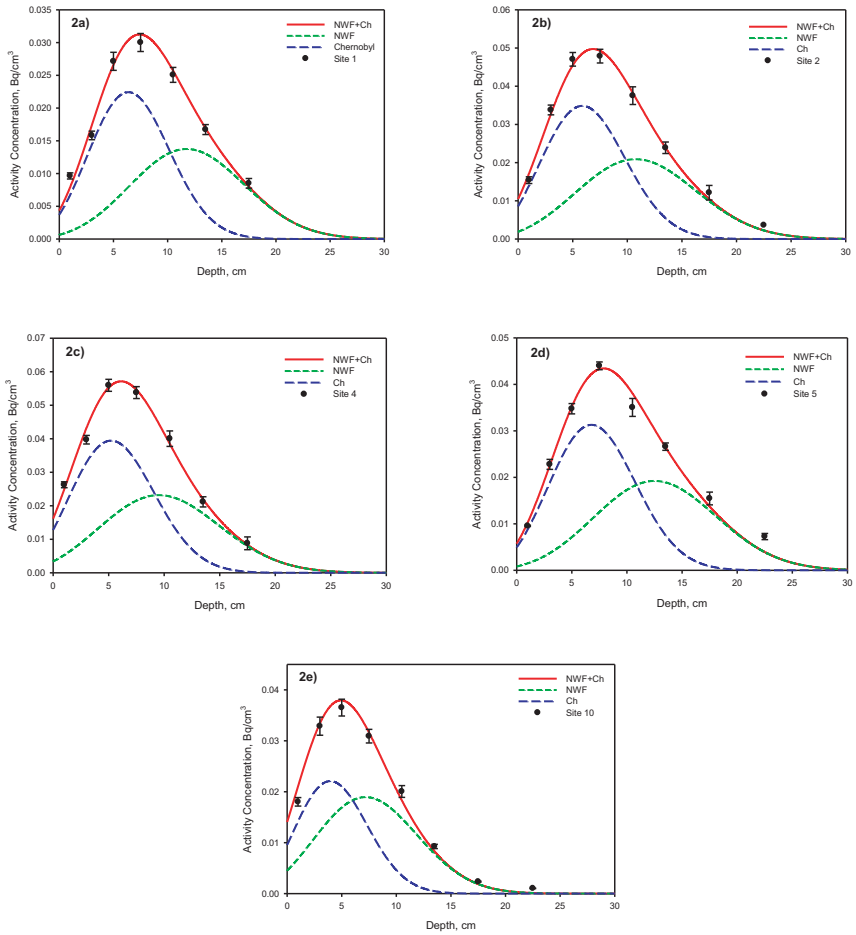

Figure 2 – ***Depth distribution of measured ^{137}Cs activity at sites with subsurface peak profiles. Total and individual contributions of each source in fitted curves are shown as well.***

3. Results and discussion

To evaluate the transport parameters and initial fallout values, the measured specific activities in Bq.kg^{-1} were converted to activity concentration in Bq.cm^{-3}. The results of the fitting process of activity concentration *versus* linear depth, using equation (4) as a CDE solution including both sources, are shown in Figures 2 and 3. The results for site number 3 showed some evidence of soil disturbance, so this site was discarded. As demonstrated from both figures, when

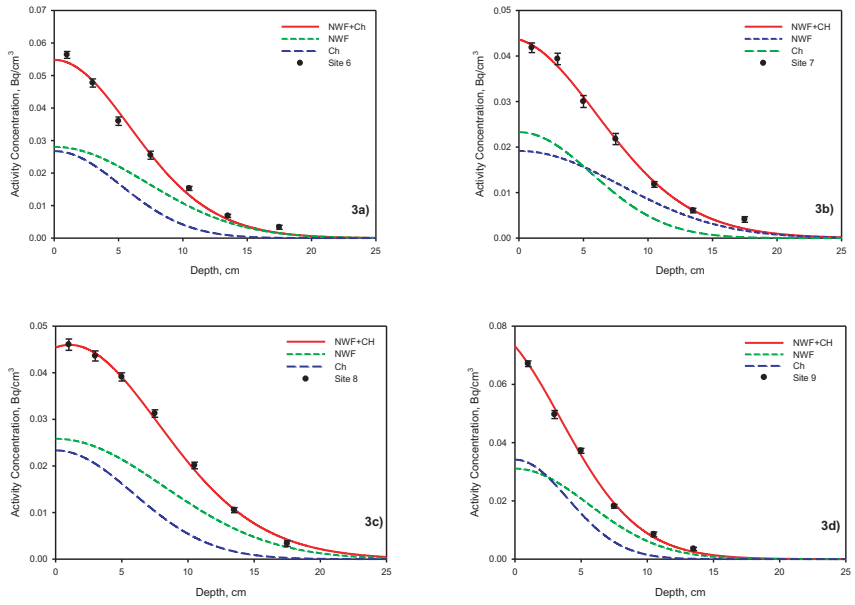

Figure 3 – ***Depth distribution of measured ^{137}Cs activity at sites with monotonically decreasing profiles. Total and individual contributions of each source in fitted curves are shown as well.***

the contribution of NWF and Chernobyl sources is considered, the resulting curves fit the measured data well and generally enhance the quality of the whole fitting procedure, especially at deeper layers. The individual contribution of each source is also shown in these figures by dashed lines. All ^{137}Cs vertical profiles exhibit a tailing effect, *i.e*. the activity extension into deeper layers. At most sites, activity of the one or two last layers were small and stood below the LLD level. Actually, the depth interval of 20-25 cm was the last measurable layer.

To derive transport parameters, equation (4) was compared with the measured data with four free parameters of initial depositions: $J_{0,NWF}$, $J_{0,Ch}$ and the effective convection velocity and dispersion coefficients V_{eff} and D_{eff}. The corresponding best fitted parameters of $J_{0,NWF}$, $J_{0,Ch}$, D_{eff} and V_{eff} which were obtained from the nonlinear fit model using the Levenberg-Marquardt algorithm and a 95% confidence interval are shown in Table I. The initial deposition value of the $J_{0,NWF}$ source ranges from 0.47-0.82 Bq.cm^{-2} with an average value of 0.66±0.12 Bq.cm^{-2}. The latter value is close to, but a bit higher than the calculated deposition of ^{137}Cs for mid-latitudes of the Northern hemisphere (UNSCEAR, 2000), which might be ascribed to the precipitation characteristics of the region as well as occult deposition.

TABLE I

Deduced transport parameters along with differentiated maximum depth inventory for each source.

Site #	$J_{0,NWF}$ (Bq.cm^{-2})	$J_{0,Ch}$ (Bq.cm^{-2})	D_{eff} (cm^2.y^{-1})	V_{eff} (cm.y^{-1})	$Z_{Max,NWF}$ (cm)	$Z_{Max,Ch}$ (cm)	R^2
1	0.47(9)	0.34(9)	0.39(6)	0.24(5)	11.9	6.5	0.97
2	0.76(12)	0.53(11)	0.38(9)	0.22(3)	10.8	5.9	0.99
4	0.82(17)	0.58(12)	0.32(8)	0.18(7)	9.5	5.2	0.98
5	0.72(16)	0.49(10)	0.36(8)	0.25(7)	12.5	6.8	0.94
6	0.68(9)	0.28(7)	0.59(9)	0.00(0)	–	–	0.98
7	0.50(9)	0.30(6)	0.59(6)	0.08(2)	–	–	0.97
8	0.73(8)	0.29(7)	0.75(5)	0.03(1)	–	–	0.98
9	0.60(9)	0.29(7)	0.35(6)	0.02(1)	–	–	0.99
10	0.62(10)	0.24(9)	0.32(6)	0.14(3)	7.2	4.0	0.99

The corresponding value for $J_{0,Ch}$ stands between 0.24 and 0.58 Bq.cm^{-2} with an average value of 0.37±0.13 Bq.cm^{-2}. The major studies of Chernobyl ^{137}Cs deposition have been carried out inside the European territory and there is no published data for the lower Caucasus region (UNSCEAR, 2000). However, recent numerical simulations of Chernobyl releases show the passage of the plume over the South Caspian region in the period of 5 to 7 May 1986 (Brandt *et al*., 2000; Quelo *et al*., 2007). Local weather station data reveal that this plume passage had been accompanied by scattered precipitation in the study area (IRIMO, 1986). Therefore, it is not unlikely to find a Chernobyl contribution in ^{137}Cs inventories of the regional soil. The estimated $J_{0,Ch}$ value is in good agreement with the range of the available nearest deposition density, *i.e*. 0.2-1 Bq.cm^{-2}, above the northern geographical borders of Iran (UNSCEAR, 2000).

The effective convective velocity range is 0-0.25 cm.y^{-1} with an average of 0.12±0.09 cm.y^{-1}, and the dispersion coefficient range is 0.32-0.75 cm^2.y^{-1} with an average of 0.45±0.15 cm^2.y^{-1}. The results of estimated parameters for the sites represented in Figure 3 revealed no advective term and it can be regarded as a pure diffusive process. This might be due to the relatively low clay and higher organic matter content of the soil. On the other hand, the sites represented in Figure 2 contain more clay and a higher retention capacity. It seems gradual increase in clay content with depth has prevented the diffusion of radiocesium to the deepest layer.

TABLE II

Comparison of estimated transport parameters and initial fallout with respect to assumed coefficients.

	Site 2		Site 5	
	fixed D_{eff} & V_{eff}	variant D_{eff} &V_{eff}	fixed D_{eff} & V_{eff}	variant D_{eff} &V_{eff}
$J_{0,NWF}$ (Bq.cm^{-2})	0.76	0.93	0.72	0.89
$J_{0,Ch}$ (Bq.cm^{-2})	0.53	0.44	0.49	0.41
$D_{eff,NWF}$ (cm^2.y^{-1})	0.38	0.40	0.36	0.42
$D_{eff,Ch}$ (cm^2.y^{-1})		0.29		0.27
$V_{eff,NWF}$ (cm.y^{-1})	0.22	0.21	0.25	0.26
$V_{eff,Ch}$ (cm.y^{-1})		0.21		0.24
$Z_{Max,GF}$ (cm)	10.8	10.7	12.5	12.8
$Z_{Max,Ch}$ (cm)	5.9	5.5	6.8	6.2
R^2	0.99	1.00	0.94	0.97

The maximum depth inventory of NWF and Ch sources is at 10.4±2.1 cm and 5.7±1.1 cm, respectively (Tab. I). This shows that over decades, considerable amounts of deposited ^{137}Cs are still found in the top 10 cm of regional soil.

One of the limitations of equation (3) is the assumption of constant transport parameters over space and time. It is believed that at the early stages the initial fast infiltration due to rainwater occurs and the migration is rapid. This may lead to different migration mechanisms. The prolonged tail in activity is observed in many studies (Almgren and Isaksson, 2006; Krstic *et al*., 2004; Szerbin *et al*., 1999). Assumption of fixed values for D_{eff} and V_{eff} coefficients in equation (4) leads to an underestimation of experimental data at depths below 10 cm. This effect could also be ascribed to spatial variation of the hydraulics, sorption properties and biological characteristic of soil (Bunzl, 2001; Krichner *et al*., 2009). The estimated transport parameters are merely the mean corresponding values through depth intervals. If we consider a depth variation of these parameters, taking into account a gap of around two decades between NWF and Chernobyl depositions we might assume different values of D_{eff} and V_{eff} for these two sources. Of course, decreasing degrees of freedom resulting from the two extra parameters require many more data points to reach a satisfactory conclusion. However, we examined such a scenario for our limited available data and headed for determination of different transport coefficients for two sources. In such a case, both time duration and transport parameters affect the outcome of equation (3). The results are presented in Table II and Figure 4, which shows an improvement in the fitted curves. NWF and Chernobyl components are shown as well.

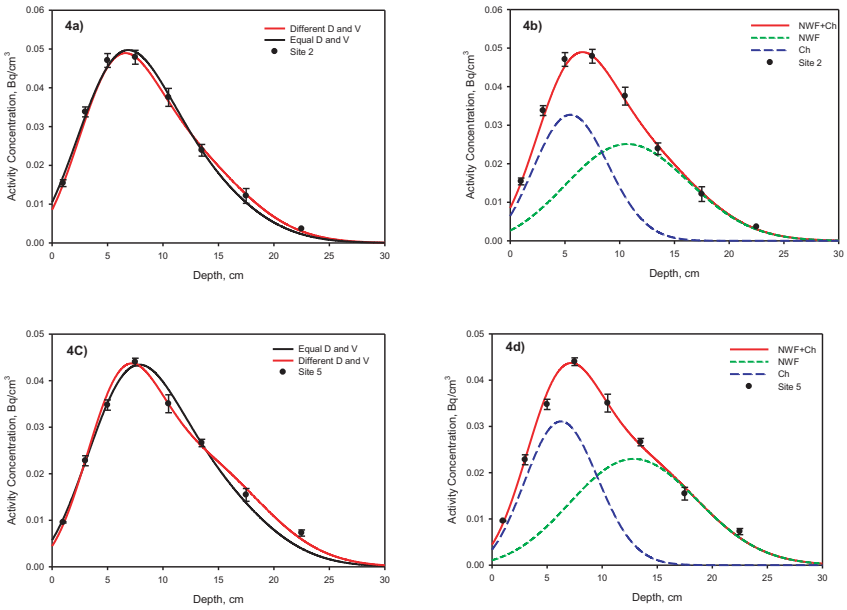

Figure 4 – ***The comparison of experimental fit between fixed and variant transport parameters*****.**

Table II clearly shows that the diffusion coefficient is more affected by following such a procedure. Application of the variant transport coefficient to other sites generally leads to slight improvement in curve fitting quality. The effectiveness of this assumption strongly depends on the availability of field data for numerous layers.

Previous results for transport parameter calculation on the basis of the same CDE solution are presented in Table III. The variation is mainly due to the different types of soil studied.

A comparison between the measured soil inventories of ^{137}Cs and corrected *in situ* measurements for the representative site of each region is presented in Table IV. In this regard, equation (5) was employed to estimate the corrected *in situ* soil inventory. As the R^2 coefficients in Table I indicate, equation (4) with four estimated parameters fits the experimental data well. The corresponding values for corrected activity derived by *in situ* measurements indicate a deviation of approximately 18% relative to measured soil activities. This indicates that the appropriate correction factor could be deduced from such model calculations to be applied to *in situ* measurements, which is of great value for wide-range ^{137}Cs monitoring.

TABLE III

The reported values of transport coefficients using equation (3).

Reports	D_{eff} (cm^2.y^{-1})	V_{eff} (cm.y^{-1})
Krstic *et al*. (2004)	0.34-1.47	0-0.26
Almgren and Isaksson (2006)	0.06-2.63	0-0.35
Bossew and Kirchner (2004)	0.05-0.50	0-0.50
Ivanov *et al*. (1997)	0.06-1.42	0.03-0.90
Butkus and Konstantinova (2008)	0.10-0.13	0.22-0.29
Current study	0.32-0.75	0-0.25

TABLE IV

Comparisons between *in situ* and soil inventory measurements of ^{137}Cs.

Site	Measured soil activity (Bq.cm^{-2})	Corrected *in situ* results (Bq.cm^{-2})
Region 1	0.62	0.73
Region 2	0.42	0.39

4. Conclusions

Considering the importance of soil diffusion study of deposited anthropogenic radionuclides, a series of soil inventory measurements for the fission-product radionuclide, ^{137}Cs, was carried out in selected regions of the South Caspian coast along with gamma *in situ* monitoring. Sampling locations were selected on the basis of a previous comprehensive survey in the area. Cesium-137 activities were determined using a HPGe gamma-spectrometry system. The experimental data were satisfactorily compared with the solution of the convection-dispersion equation considering both global fallout and Chernobyl sources. The resulting convection velocity and dispersion coefficients in the range of 0-0.25 cm·y^{-1} and 0.32-0.75 cm^2·y^{-1}, respectively, reveal a very slow migration rate in the area. Most of the deposited ^{137}Cs still remained in the top 10-cm layer. The fitted depth profiles could be employed as a measure to correct the surface activities of ^{137}Cs estimated by *in situ* measurements. Such a procedure is of great importance in evaluation and control of public exposure in case of emergencies and would provide a basis for identifying any potential changes in the radioactivity levels in soil due to human activities. It could also be particularly valuable in quantifying aerosol and wet deposition processes at sites where conventional methods are not applicable.

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