



## Residence Half-times of $^{137}\text{Cs}$ in Undisturbed Surface Soil in Nigeria based on Measured Soil Concentration Profiles

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### Abstract

The transport of fallout  $^{137}\text{Cs}$  within the Nigerian soil has been studied in this paper by a compartment model. The measured concentrations of the radionuclide in the soil layers were examined by the model in order to estimate the transport parameters of residence half-time and the migration velocity of the radionuclide in the soils. The residence half-times were observed to be short and decreased with soil depth with a mean ranging from 0.3 years at 0 – 2cm soil depth in Akure site to 1.4 years at 10-15cm depth in Igbeti site. The migration velocity  $v$ , calculated from the residence times with the consideration of the different thicknesses of the layers were observed to decrease slowly with depth at all the sampling sites with a range from  $3.62 \text{ cm y}^{-1}$  at Igbeti site to  $8.21 \text{ cm.y}^{-1}$  at Ikogosi site. The migration velocities obtained are quite higher than those reported in literature for global fallout.

**Keywords:** Fallout  $^{137}\text{Cs}$ , Undisturbed soil, Vertical migration, Residence time, Compartment model, Migration velocity, Concentration

### 1. Introduction

The concentration and vertical distribution of  $^{137}\text{Cs}$  in an undisturbed soil in southwestern Nigeria had been experimentally studied (Ajayi et al 2007). The study had provided further knowledge on  $^{137}\text{Cs}$  transport and migration behaviour in soil in addition to the several studies that had previously been done by many researchers in the subject (Coughtrey and Thorne, 1983, Lee and Lee 1997, Schuller et al 1997, Kanunakara et al 2001, Grzegorz et al 2003 and Kristic et al 2004.). The experimental study generally showed that more than 45 years after the first nuclear weapon tests carried out by France at Reganne in the Sahara region in northern Nigeria between 1960 and 1961,  $^{137}\text{Cs}$  still remains within the 25cm upper layer of the soils in southwestern Nigeria with varying concentrations along vertical profiles and with a mean effective dose commitment of  $10.77\mu\text{Sv}$

As a follow-up to this study, the measured  $^{137}\text{Cs}$  concentrations in the different soil layers were examined in this study by means of a five-compartment mathematical model to estimate the residence time of the radionuclide in a given layer of soil and its downward migration velocity in the layer. The residence time and the migration velocity of a radionuclide are the basic transport parameters of the radionuclide within the soil. (Muller and Bleher, 1997, Monte et al 2003, Bossew et al 2004, Putyrskaya and Clemt, 2007).

The presence of fallout  $^{137}\text{Cs}$  in the soil results in a long-term radiation dose to man depending essentially on the vertical migration of the radionuclide in the surface-near soil layers. The external gamma dose decreases with time due to an increasing shielding of the gamma radiation by the overlaying soil if the radionuclides are transported deeper into the soil. The internal exposure from radionuclide incorporated by ingestion of contaminated food is also affected by their vertical migration because it determines the residence time of the radionuclide in soil layers relevant for the uptake by plant roots. Therefore, radionuclide migration in the soil is considered very crucial in evaluating the radiation exposure of the population (Muller and Bleher, 1997). For long-term predictions, the activity–depth profiles of the radionuclide can be calculated with the migration parameters obtainable by the transport models.

Many research have particularly focused on  $^{137}\text{Cs}$  and other artificial radionuclides in soils because of their pollution and contamination of the environment. Their fixation to minerals and organic components retards their migration and losses to deep soil and groundwater (Cheshire and Shand 1991, Herman et al.1992.).

The compartment model is a model that has been widely used to study radionuclide migration and behavior in the soil (Frissel and Pennders 1983, Bunzl et al 1989, Bunzl et al 1994 and Kirchner 1998, , etc). The model justifies the use of migration rates which are different for each soil layer because it is assumed that soil properties are uniform for each horizon. A single migration rate is therefore associated with each soil horizon.

The migration of  $^{137}\text{Cs}$  is generally believed to be a complicated process affected by a number of factors like the type of soil, its chemical properties, organic matter content , biological activity of microorganism in soil, etc (Grzegorz et al 2003) and therefore its behaviour in a natural ecosystem is yet difficult to unravel. Some authors (Frissel and Pennders 1983, Denk and Felsmann 1989.) have therefore suggested the use of physical parameters such as residence half-times and migration rates to design dynamic models for the quantitative description of the transport processes in soil.

This study aimed at evaluating the transport parameters of residence half-times and migration velocity of  $^{137}\text{Cs}$  in different soil layers using a simple five compartment mathematical model. Experimental data obtained from the Cs vertical distribution studies earlier carried out in the study area were used for validation purposes.

## 2. Materials and Methods

### 2.1 Site

The experimental study site covered three states in southwestern Nigeria and because there is no nuclear installation in the country yet, the soil can only be expected to contain  $^{137}\text{Cs}$  deposited as a result of global fallout due to nuclear weapons tests. The general level of the region lies between 200 and 270m above sea level with a mean annual precipitation of about 2000mm and a dense forest vegetation. The sampling locations were selected on flat soil surfaces not prone to precipitation run-off.

### 2.2 Sampling and sample preparation

Samples were collected from nine sampling locations in the region. The samples were collected in January 2005 with a frame (10cm x 10cm) at nine plots of the sampling area. Soil samples were collected in depth increments of 0-2, 2-4, 4-6, 6-8, and 8-10cm at the sampling sites by a coring tool which was thoroughly clean and dried before each sampling. Some samples were also collected at depth increments of 0-5, 5-10, 10-15, 15-20, 20-25 and 25-30cm as variation. Before radioanalysis of the samples, the soils were well mixed, weighed and then dried in an oven at  $110^{\circ}\text{C}$ . The samples were then sieved by a 2mm mesh screen and about 200g each of the sieved soil samples were then subjected to gamma spectroscopy for activity determination. Soil samples from each layer of the nine vertical profiles were analysed separately in order to determine the spatial variability of the residence time of the radionuclide in the soil.

### 2.3 Activity determination

$^{137}\text{Cs}$  was determined by direct gamma spectrometry measurements using the coaxial-type HpGe detectors of 50% relative efficiency with a resolution of 2.4keV at 1.33MeV. The detector which was properly shielded in lead castles was calibrated using certified reference standards for various radionuclides. The samples were counted for at least 24hours each to achieve low counting error. Spectra analysis were performed with the Genie2K spectrometry software version 2.1(Canberra industries). The characteristic gamma peak of 662keV of the  $^{137}\text{Cs}$  was used for its identification among the library of radionuclides in the software. The specific activities of  $^{137}\text{Cs}$  in the soil were expressed in  $\text{Bq/m}^2$  of dry mass of soil and corrected for the time elapsed since the sample collection from the sampling site. The results are as depicted in figures 1-3. The total  $^{137}\text{Cs}$  deposition in the sampling sites are as shown in table 1.0

### 2.4 Evaluation of the residence half-times

As mentioned in the introduction, the residence half time of  $^{137}\text{Cs}$  in the various soil layers were evaluated with a five compartmental model (Bunzl et al 1994, Frissel et al 1981, Boone et al 1985). The model can be applied with ease because it does not require detailed information on the actual transport processes of the radionuclide in the soil such as the sorption properties, water infiltration etc. It however has the disadvantage that only the residence times and migration velocity of the radionuclide with the soil layers can be obtained.

## 3. Theory of the compartment model

The multicompartment model (Kirchner 1998, Likar et al 2001) assumes that the soil depth is split into a series of N horizontal layers which are connected by a downward transport rates of the particular radionuclide being investigated. In this case, the transfer of activity  $A_i(\text{Bqm}^{-2})$  of a particular radionuclide in the compartment  $i$  in a small time interval  $dt$  can be mathematically expressed as:

$$\frac{dA_i}{dt} = K_{i-1}A_{i-1} - K_iA_i - \lambda A_i \dots\dots\dots (1.0)$$

Where  $K_i$  is the fractional rate of transfer from one compartment  $i$  and  $\lambda$  is the disintegration constant of the radionuclide.

For the first compartment equation (1) becomes

$$\frac{dA_1}{dt} = -(\lambda + K_1) \times A_1 \dots\dots\dots (2.0)$$

The above differential equation has a familiar solution given by:

$$A_1(t) = A_0 \times e^{-\left(\lambda + k_1\right)*t} \dots\dots\dots (3.0)$$

Where  $A_0$  is equal the rate of deposition in  $Bqcm^{-1}y^{-1}$

$A_0$  is known as a function of time obtained from deposition data.

For the second and deeper compartments the change in activity becomes

$$\frac{dA_n}{dt} = K_{n-1} \times A_{n-1} - \lambda \times A_n \dots\dots\dots (4.0)$$

Integration of the above equation yields

$$A_n(t) = A_{n-1} * \left[ \frac{K_{n-1}}{\lambda + K_n - (\lambda + K_{n-1})} + \frac{\lambda + K_{n-1}}{\lambda + K_n} * e^{-(\lambda + K_n - \lambda - K_{n-1}) * t} \right] \dots\dots\dots (5.0)$$

Which is further simplified as

$$A_n(t) = A_{n-1} * \left[ \frac{K_{n-1}}{K_n - K_{n-1}} + \frac{\lambda + K_{n-1}}{\lambda + K_n} * e^{-(K_n - K_{n-1}) * t} \right] \dots\dots\dots (6.0)$$

For global fallout  $A_{i0}$  at  $t = 0$  has been determined, and for the initial depth profile  $A_{i0}$ , we assume that the activity of  $^{137}Cs$  initially remains completely in the first layer of the soil ( Frissel et al 1981 and Bunzl 2001).

The  $^{137}Cs$  concentration profile determined in the sampling area in September 2006, a period of forty six years after the  $^{137}Cs$  deposition from the global fallout as a result of nuclear weapons tests in the Sahara desert around 1960 was used as the initial depth profile at  $t = 0$  in the model .

Since the  $A_i$  in equation (1) were determined by experimental measurements,  $K_i$  is the only unknown parameter for each layer  $i$ . The  $K_i$  were calculated for by an iterative estimation using MATLAB 3.0. to a good degree of accuracy.

The residence half-time of the radionuclide in layer  $i$  was calculated by the equation below (Boone et al 1985)

$$\tau_i = \frac{0.693}{K_i}$$

In order to compare the values of  $\tau$  obtained for soil horizons of different thicknesses and depths, the ratio of  $\tau/L$  (Frissel et al 1981) was also calculated. The reciprocal of this ratio can be construed as a velocity and therefore the rate of migration of the radionuclide in a given soil layer  $i$  can be expressed as

$$v_i = \frac{L_i}{\tau_i}$$

**4. Results and Discussion**

The total deposition for  $^{137}Cs$  in  $Bqm^{-2}$  as obtained experimentally by summation of the activities of the nuclide in the individual soil layers within the depth are as given in the table 1 below.

*4.1 Depth Profiles*

Figures 1-3 show the typical depth profiles of  $^{137}Cs$  in the nine soil profiles. The results showed that the activity decreases with increasing soil depth and it was below detection limit in the 25-30cm and in the subsequent profiles. This showed that the  $^{137}Cs$  deposited on surface soil through fallout was able to move down to a maximum depth of

25cm in the region in a straight line migration pathway where consideration was not given to the uptake of the nuclide by plants.  $^{137}\text{Cs}$  total deposition at 10cm soil depth was found to be greatest at Ikogosi site 1 with a value of  $90.30\text{Bqm}^{-1}$  and least in Ogbomoso with a value of  $15.60\text{Bqm}^{-2}$  as already shown in table 1.

#### 4.2 Residence half-times and rates of migration

The depth profiles shown above have only illustrated the experimental results obtained from the study of the vertical migration of  $^{137}\text{Cs}$  at the sampling sites. The quantitative description of the vertical mobility can only be explained by the parameters of residence half-times  $\tau$  or migration velocity  $\nu$ . These parameters are more valid. The residence times and the migration velocity as obtained for each soil layer with the compartment model are as summarized in table 2 below.

The variability of the values of  $\tau$  and  $\nu$  between the nine depth profiles as evaluated by the model is as shown in figures 5-8.  $\tau$  increased very slowly with depth while  $\nu$  decreased very slowly with depth. The small variations observed may be due to small scale variability in soil properties of the area. The soil properties were not investigated in this paper for lack of facilities for the purpose.

The residence half-time of the radionuclide was generally observed to increase with soil depth at all the sites with the highest value in of 1.4 years in the 10-15cm depth in Igbeti area site. The lowest half-time of 0.30 years was observed in the 0-2cm depth at Akure sites. The short residence times generally indicate that the radionuclide does not dwell for long times in the soil layers before migrating downward. It is also observed that the with the increase in the residence time with soil depth the radionuclide is likely to dwell for longer times in the deeper layers of soil and therefore be available in the rooting zone for plants that are deep feeders. The migration rates  $\nu$ , calculated from the residence times with the consideration of the different thicknesses of the layers are also shown in the figures. The migration rate of  $^{137}\text{Cs}$  was observed to decrease slowly with depth at all the sampling sites. The decrease in the migration velocity with depth in all the sites may be explained as a result of the presence of K or Na ions or clay minerals with strong sorption properties for  $^{137}\text{Cs}$  in the deeper soil layers. It has been reported that clay minerals are strong enough for strong fixation of cesium. The adsorption of cesium is decreased by the presence of competing ions of K or Na ions in the soil.(Coleman et al 1963, Davis 1963).

The mobility of  $^{137}\text{Cs}$  can be said to be generally slow in the upper layers of the soil with an average velocities of 6.64, 5.67, 7.68, and 3.75  $\text{cm y}^{-1}$  at Akure, Ado-Ekiti, Ikogosi and Igbeti sites respectively using this model. The peak appearance of the radionuclide in depth 6-8 cm in Akure and Ado-Ekiti sites may be explained by the convective transport of Cs-137 in the soils. It can also be explained as the result of the time course of the deposition of the global fallout which was not constant but extended over some time (Krstic et al 2004). The study specifically referred to the nuclear weapon tests of about 46 years ago on the Sahara desert in northern Africa. Several other weapon tests had been carried out after that with the possibility of radiocaesium deposition on the study area. Therefore the results of this study cannot be said to be accurate but rather have provided a preliminary information that can be further investigated or studied in the future. It must however be restated that  $^{137}\text{Cs}$  migration is a generally complicated process which is affected by a number of factors like the type of soil, its chemical properties and organic matter content. Cesium mobility in soil also depends on climatic conditions such as rainfall, temperature or humidity and biological activity of microorganisms in soil (Grzegorz et al 2003).

#### 5. Conclusion

$^{137}\text{Cs}$ , a gamma emitter with energy of 661keV with half-life of 30.1years which is a biologically important fission fragment attributed to atmospheric nuclear weapon testing is still within surface soil in Nigeria. The residence times increased very slowly from one soil layer to another suggesting that the radionuclide does not dwell for long times in the soil layers before migrating downward and implying that the radionuclide is likely to dwell for longer times in the deeper layers of soil and therefore be available in the rooting zone for plants that are deep feeders. The migration velocity was observed to decrease with soil depth at all the sites suggesting the presence of K or Na or clay minerals with strong sorption properties for  $^{137}\text{Cs}$  in the deeper soil layers.

It should however be noted that the mobility of Cs-137 from global fallout in the soil will not remain constant but will rather continue to decrease with time.

#### 6. Acknowledgements

The first author is profoundly grateful to the Abdus Salam ICTP for his appointment as a regular associate of the centre. This paper was written during one of his visits to the centre.

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Table 1. Total deposition of  $^{137}\text{Cs}$  in soil as determined in 2006 at various sampling sites

Sampling Sites	Total $^{137}\text{Cs}$ deposition in Bq m <sup>2</sup>
Akure site 1	27.30
Akure site 2	63.45
Ado-Ekiti site 1	58.35
Ado-Ekiti site 2	30.60
Ikogosi site 1	90.30
Ikogosi site 2	82.80
Igbeti site	29.40
Ogbomoso site	15.60
Eruwa site	24.90

Table 2. Residence half-times and migration velocities in soil layers

Soil Depth (cm)	Residence half-time $\tau$ (years)	Mean migration rate $\nu$ (cm.y <sup>-1</sup> )
Akure sites:		
0-2	0.30	7.10
2-4	0.31	6.84
4-6	0.32	6.60
6-8	0.33	6.38
8-10	0.34	6.29
Ado-Ekiti sites:		
0-2	0.34	5.94
2-4	0.35	5.79
4-6	0.36	5.72
6-8	0.37	5.53
8-10	0.38	5.41
Ikogosi sites:		
0-5	0.61	8.21
5-10	0.63	7.95
10-15	0.64	7.77
15-20	0.66	7.53
20-25	0.72	6.95
Igbeti and other sites:		
0-5	1.31	3.87
5-10	1.36	3.76
10-15	1.41	3.62

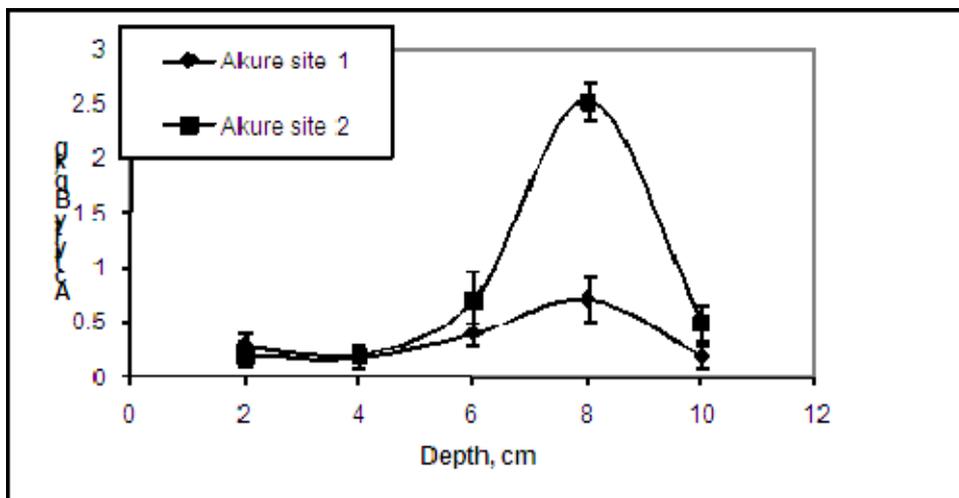


Figure 1. Showing depth distribution of <sup>137</sup>Cs in soil profile at Akure sites

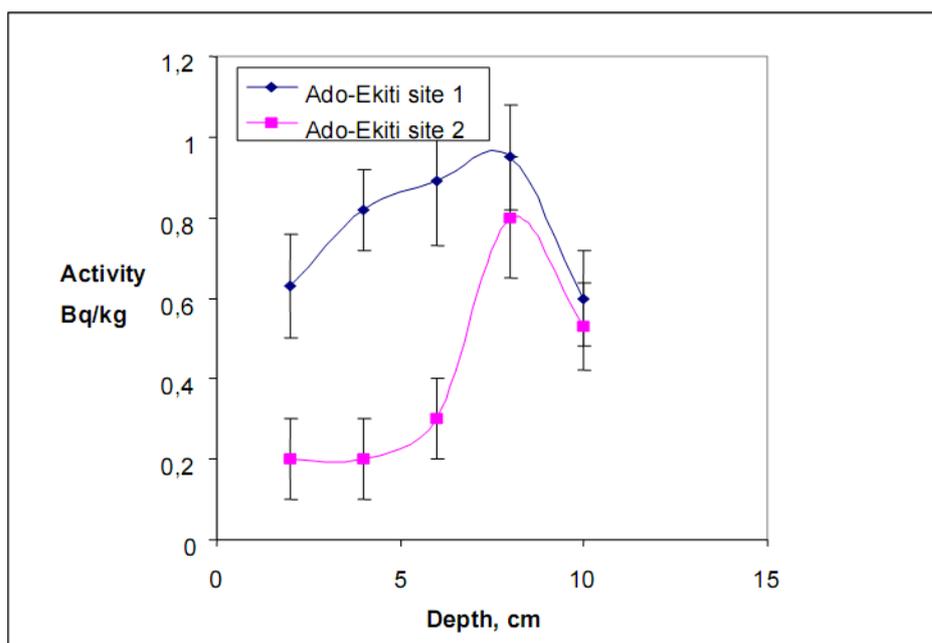


Figure 2. Showing depth distribution of <sup>137</sup>Cs in soil profile at Ado-Ekiti sites

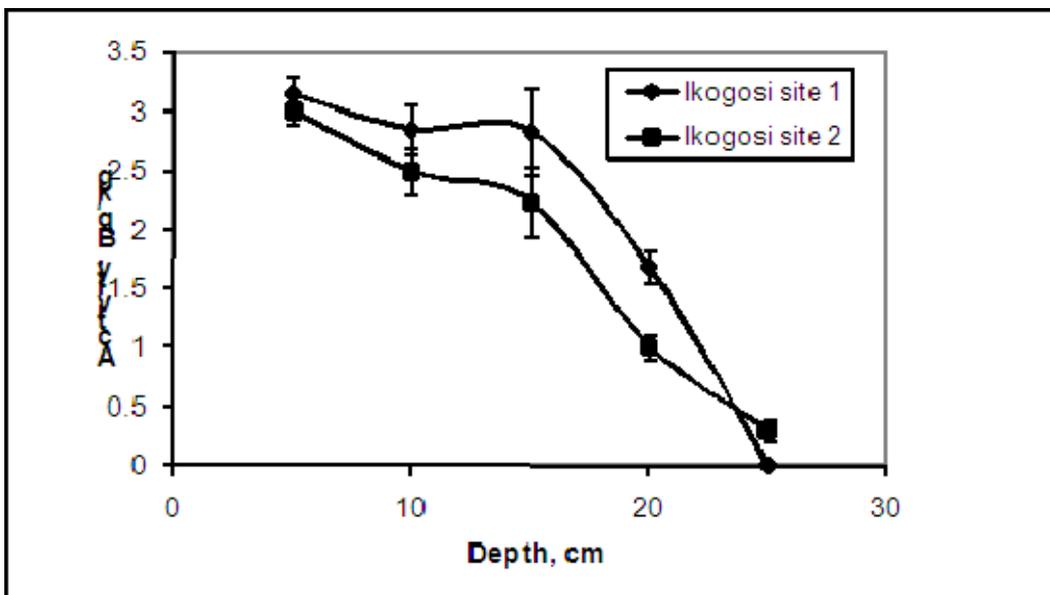


Figure 3. Showing depth distribution of <sup>137</sup>Cs in soil profile at Ikogosi sites

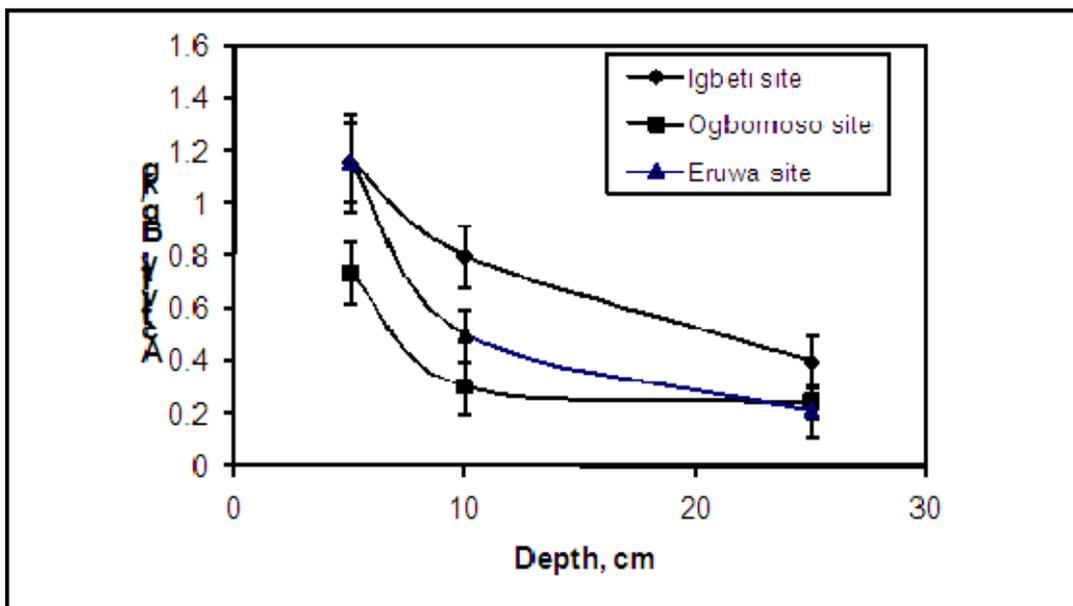


Figure 4. Showing depth distribution of <sup>137</sup>Cs in soil profile at Igbeti, Ogbomoso and Eruwa sites

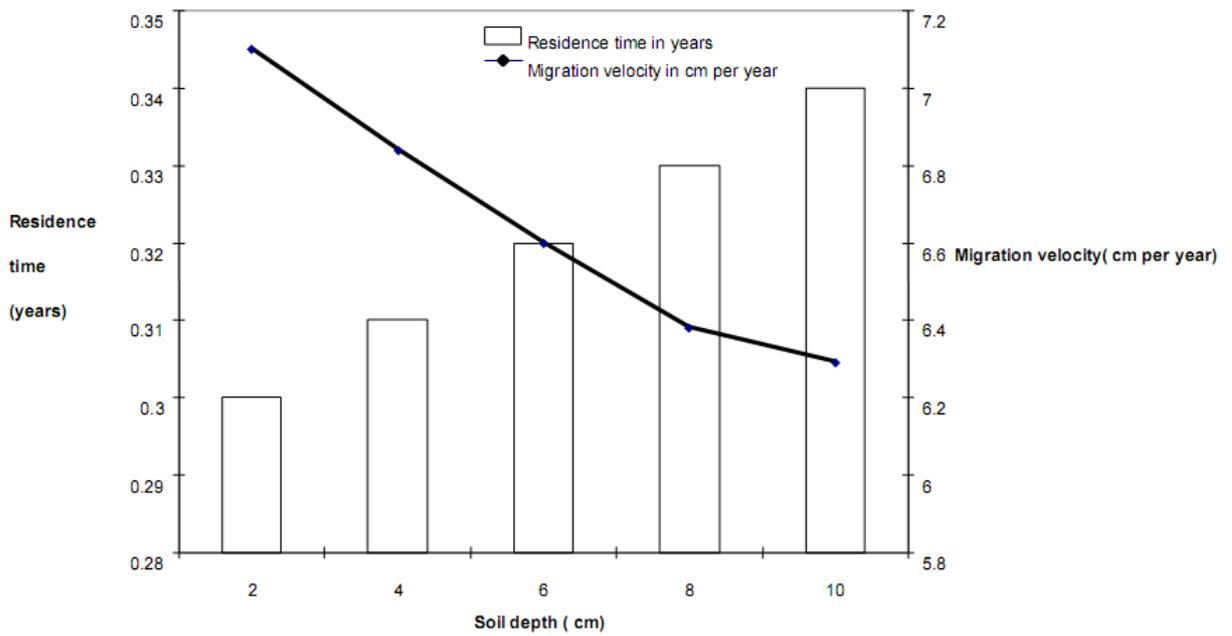


Figure 5. Variation of residence time and migration velocity of Cs-137 with soil depth at Akure

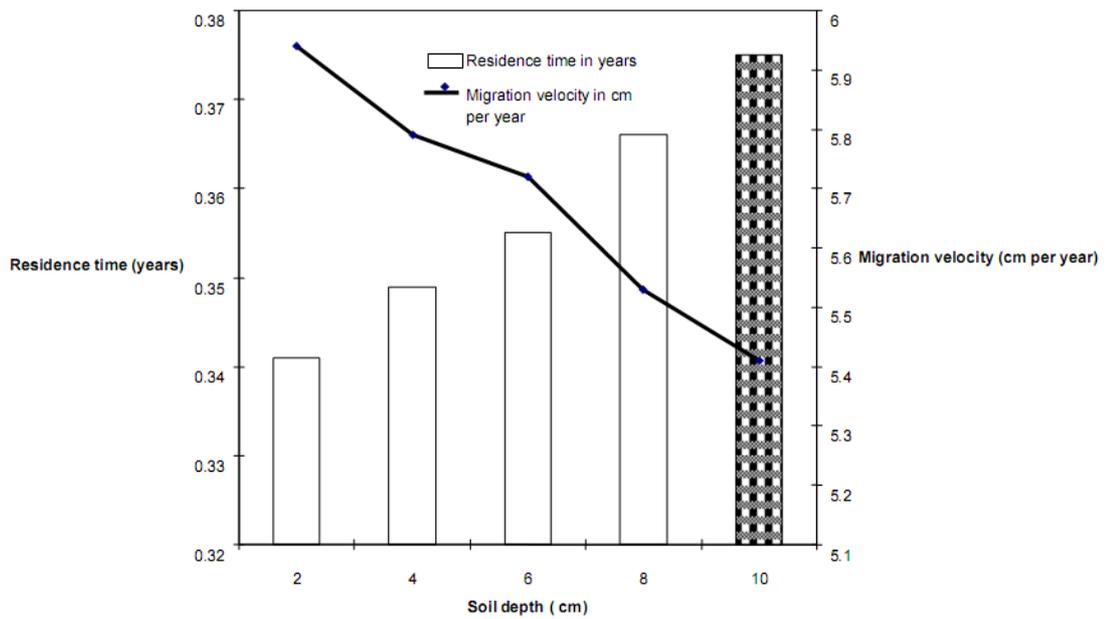


Figure 6. Variation of residence time and migration velocity of Cs-137 with soil depth at Ado-Ekiti

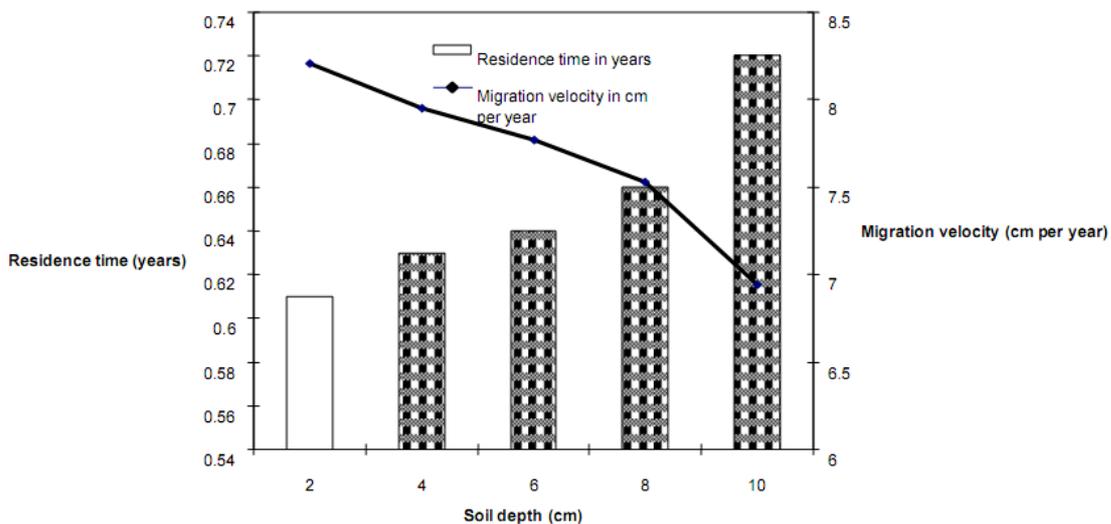


Figure 7. Variation of residence time and migration velocity of Cs-137 with soil depth at Ikogosi sites

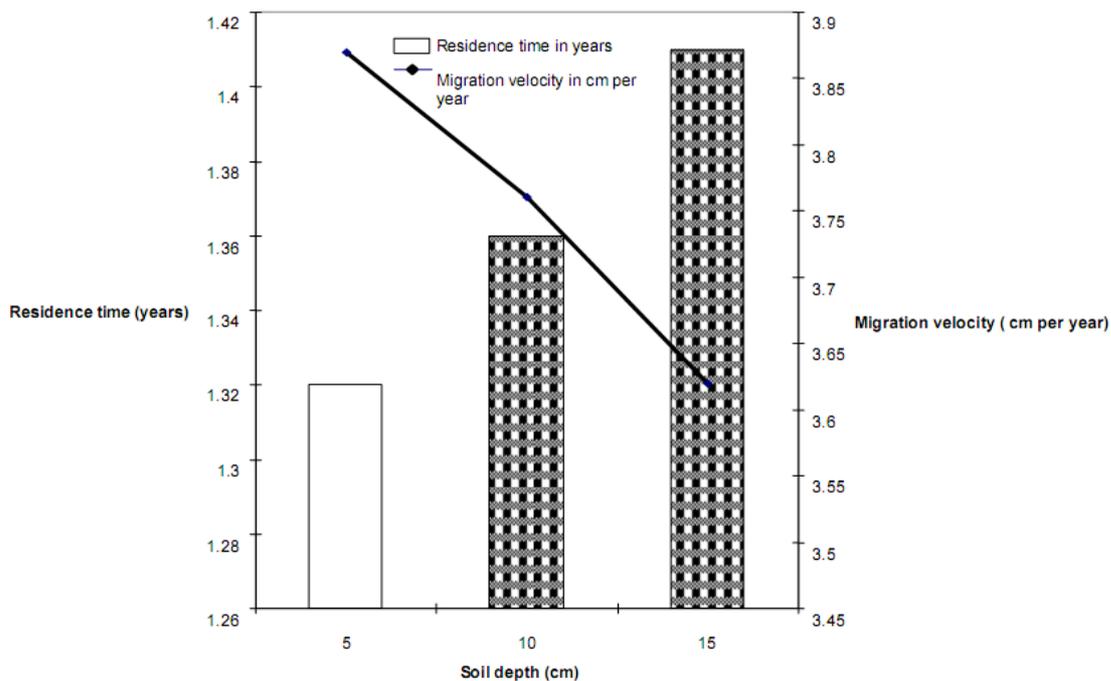


Figure 8. Variation of residence time and migration velocity of Cs-137 with soil depth at Igbeti sites