International Journal of the Physical Sciences Vol. 5 (2), pp. 154-157, February, 2010
Available online at http://www.academicjournals.org/IJPS
ISSN 1992 - 1950 © 2010 Academic Journals

Full Length Research Paper

Evaluation of the ^{137}Cs activity-depth profiles by the diffusion-convection model

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Accepted 12 November, 2009

The diffusion-convection model of ^{137}Cs transport has been used to evaluate the activity-depth profiles of soils in southwestern Nigeria in this study. The experimentally determined depth profiles were fitted into the diffusion-convection equation to obtain the diffusion coefficient D and the convective velocity u. Results obtained showed that the range of the diffusion coefficient was 0.72 – 1.02 cm^2 per year while that of the convective velocity was from 0.07 to 0.16 cm per year. It was observed that the diffusion coefficient is higher than the convective velocity in all the sampling points. The low convective velocities obtained in all the locations may imply that convective rates of the radionuclide within the soil layers are insignificant compared to the diffusion rates.

Key words: ^{137}Cs activity, diffusion coefficient, convective velocity, soil depth, radionuclide.

INTRODUCTION

As a result of the nuclear weapons tests carried out by France at Reganne in the Sahara region in northern Africa between 1960 – 1961 and several other global fallout effects a certain amount of ^{137}Cs has been deposited on the Nigerian soil. An experimental study conducted by Ajayi et al., in 2007 (Ajayi et al., 2007) on the concentration and vertical distribution of ^{137}Cs in an undisturbed soil in southwestern Nigeria had shown that more than 45 years after the nuclear weapon tests carried out by France, ^{137}Cs still remains within the 25 cm upper layer of the soils in southwestern Nigeria with varying concentrations along vertical profiles.

The study had also provided further knowledge on ^{137}Cs transport and migration behaviour in soil in addition to the several studies that had previously been done by many researchers in the subject (Kanunakara et al., 2001; Kristic et al., 2004; Lee et al., 1997; Grzegorz et al., 2003; Davis et al., 1963; Bunzl et al., 1994). The effects of the Chernobyl accident of 1986 have not been considered in the region since the large scale contamination from the accident was limited to Europe and some parts of the northern hemisphere. Chernobyl radionuclide contamination has also been shown to be In homogeneous in space and time and dependent on local weather and other conditions (Kristic et al., 2004).

^{137}Cs because of its biological importance as a fission product and a gamma emitter with energy 661.6 KeV and with half-life of 30 years has been extensively investigated in the soil. (UNSCEAR, 1998). The radionuclide is regarded as the most important constituent of the world-wide fallout because sixty percent of the collective effective dose equivalent commitment from external radiation associated with past atmospheric nuclear weapon testing can be attributed to ^{137}Cs (UNSCEAR, 1998). It enhances radiation dose to man directly via external radiation and indirectly by root uptake of plants and transfer into human diet (Shinonaga et al., 2005).

The ^{137}Cs concentration in the surface soil decreases under the influence of various processes like decay, mechanical removing with rain water, vertical migration and diffusion into deeper layers of soil. Two basic processes control the migration of the fallout in the undisturbed soil column. Firstly the particles can percolate downwards with rain water, second, the solvable phase is subject to convection and diffusion with the soil solution and sorption to the soil matter described by the convection-diffusion model equation. CDE (Bossew et al 2004).

The understanding of the mechanisms of sorption and migration of radionuclides in soil and their transfer into

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the food chain is necessary in order to reduce effectively the external and internal radiation exposure to man. Besides, information about anthropogenic radioactive levels and migration of radionuclides in soil is of fundamental importance to evaluate radioecological sensitivity of an ecosystem in terms of risk related to existing and potential radioactive contamination (Aarkrog, 1979; Howard, 2000; Howard et al., 2002 as cited by Schuller et al., 2004; Paulina et al., 1997; Cheshire and Shand, 1991; Herman et al., 1992; Frissel and Pennders, 1983; Schimmack and Schultz, 2006; Beresford and Howard, 1991).

In order to quantitatively study the migration of radionuclides in the soil, several transport models have been developed (Bunzl, 2001; Velasco et al., 1997; Takriti and Othman, 1997; Kirchner, 1998; Isaksson et al., 2001; Szerbin et al., 1999; Likar et al., 2001; Luigi et al., 2003; Victoria, 2007). Szerbin et al. applied the diffusion convection model and a homogenuous Green function as a solution of partial differential equation for infinite medium while Likar et al. (2001) used a proper Green function which satisfies boundary conditions at the soil-air interface and to describe ^{137}Cs vertical profile in soil (Kristic et al., 2004).

In the present study, the vertical distribution of ^{137}Cs as determined by an experimental study is evaluated by a diffusion convection model. The main objective of the work is to determine the diffusion coefficient and the convective velocities in the different sampling location and improve the knowledge on spatial distribution and vertical migration of global weapon's fallout ^{137}Cs in soils in the study area. The work will provide data on radioactive contamination in soils of the area which are very sparse compared to other locations. The experimental results obtained for nine sampling locations were fitted into the diffusion equation.

The theory of ^{137}Cs migration in soil

It is generally accepted that the migration of ^{137}Cs in soil can be modeled by the Fokker-Planck equation governing diffusion and convection activities (Kristic et al., 2004). The equation is stated as

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x^{\partial}} - \lambda C \quad (1)$$

Where; C is Cs-137 concentration in soil, λ is its decay constant, D is effective diffusion coefficient of Cs-137 in soil, υ is convective velocity, *x* is soil depth in respect to the surface and t is time from the deposition to date. The term – λC represents the radioactive decay or the disintegration constant.

The solution of equation 1 is given as below from Szerbin et al. (1999):

$$C\ (x,t) = C_o e^{-\lambda t}\frac{1}{2\sqrt{\pi Dt}}e^{-(x-vt)^2/4Dt} \quad (2)$$

Since C_0 is the initial surface concentration in Bq m^{-2}. C(x,t) is obtained in Bq m^{-3}

The total content C_1 of Cs-137 in soil is the sum of two components of contribution from nuclear weapons tests and the Chernobyl nuclear accident. This is expressed as

$$C_1 = C_{Nw} + C_{Ch} \quad (3)$$

But since we assume there is no significant effect of Chernobyl accident in the study area, equation (3) is written as

$$C_1 = C_{Nw} \quad (4)$$

So the solution is essentially as (2) that is;

$$C_1(x,t) = C_o e^{-\lambda t}\frac{1}{2\sqrt{\pi Dt}}e^{-(x-vt)^2/4Dt} \quad (5)$$

Where; C_0 in this case the initial Cs deposition during the nuclear weapon tests in Bq m^{-2} and t is the time period between deposition and present which in this case is 46 years.

In accounting for the reflection at the air-soil surface, Szerbin et al. (1999) introduced the term C(-x,t). If this term is applied, then final concentration expression will take the form

$$C_{final} = C_1(x,t) + C_1(-x,t) \quad (6)$$

$$C_{final} = C_0 e^{-\lambda t}\frac{1}{2\sqrt{\pi Dt}}e^{-(x-vt)^2/4Dt} + C_o e^{-\lambda t}\frac{1}{2\sqrt{\pi Dt}}e^{-(-x-vt)^2/4Dt} \quad (7)$$

METHODOLOGY

Sample preparation and measurements

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}Cs deposited as a result of global fallout due to nuclear weapons tests. The general level of the region lies between 200 and 270 m above sea level with a mean annual precipitation of about 2000 mm and a dense forest vegetation. The sampling locations were selected on flat soil surfaces not prone to precipitation run-off.

Samples were collected from nine sampling locations in the region. The samples were collected in January 2005 with a frame (10 x 10 cm) at nine plots of the sampling area. Soil samples were collected in depth increments of 0-2, 2-4, 4-6, 6-8, and 8-10 cm 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 25-30 cm as variation. Before radioanalysis of the samples, the soils were well mixed, weighed and then dried in an oven at 110^0C. The samples were then sieved by a 2 mm mesh screen and about 200 g 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.

^{137}Cs was determined by direct gamma spectrometry measurements using the coaxial-type Ge detectors of 50% relative efficiency with a resolution of 2.4 keV at 1.33 MeV. 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 24 h 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}Cs was used for its identification among the library of radionuclides in the software. The specific activities of ^{137}Cs in the soil were expressed in 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}Cs deposition in the sampling sites are shown in Table 1

RESULTS AND DISCUSSION

Depth profile for Akure sites

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

Diffusion coefficient D and the convective velocity u

The decreasing concentration of ^{137}Cs with soil depth can be explained by assuming pure diffusion and that convective migration is weak or never occurred in the soil. The depth profiles above were fitted into the equation 7 above by the gnuplot software to obtain the diffusion coefficient D and the convective velocity u. The results are as shown in Table 1

The range of the diffusion coefficent is 0.002 – 1.002 cm^2 per year. This range is lower than those for Hungarian soil (Szerbin et al., 1999) and in Serbia (Kristic et al., 2004). This implies that the diffusion rate of ^{137}Cs in the soil of the study area is slower compared to those areas mentioned. Some authors have used other kinds of functions to fit experimental data such as the Lorentz function (Hillmann et al., 1996) but could not determine the diffusion coefficient. The convective velocity obtained by this model ranged from 0.007 to 1.00 with most locations having 1.00. It can be observed that the diffusion

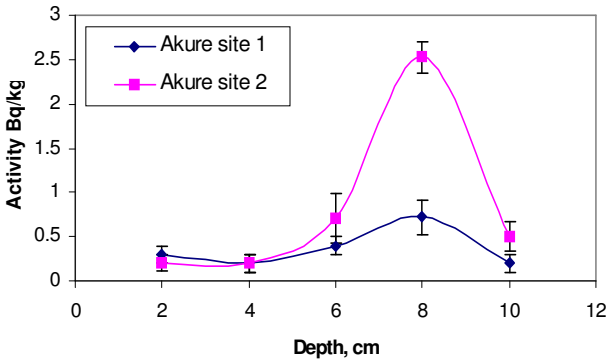

Figure 1. Showing depth distribution of ^{137}Cs in soil profile at Akure sites.

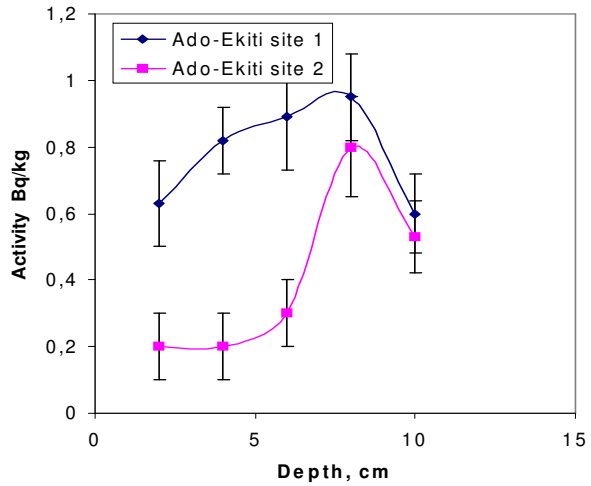

Figure 2. Showing depth distribution of ^{137}Cs in soil profile at Ado-Ekiti sites. Depth Profile For Ado - Ekiti sites.

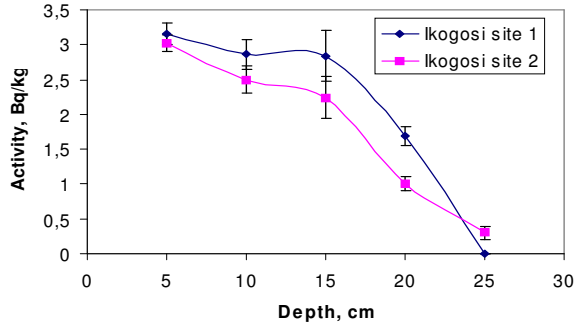

Figure 3. Showing depth distribution of ^{137}Cs in soil profile at Ikogosi sites. Depth Profile for Ikogosi sites.

Table 1. Fitted initial surface Cs-contamination diffusion coefficients and convective Velocities.

Location	Surface deposition (Bqm^{-2})	D(cm^2 $year^{-1}$)	υ (cm $year^{-1}$)
Akure site 1	27.30	0.0002	0.07
Akure site 2	63.45	1.002	0.72
Ado-Ekiti site 1	58.35	1.00	1.00
Ado-Ekiti site 2	30.60	1.00	1.00
Ikogosi site 1	90.30	1.00	1.00
Ikogosi site 2	82.80	1.00	1.00
Igbeti site	29.40	1.00	1.00
Ogbomoso site	15.60	1.00	1.00
Eruwa site	24.90	1.00	1.00

coefficient and the convective velocity are equal for most locations, implying that the diffusion and the convective rates of the radionuclide within the soil layers at those sites are nearly the same. The peak appearance in the depth profiles may be explained by the convective transport of ^{137}Cs in the soils (Kristic et al., 2004). Some authors have recommended that the convective velocity be neglected for cases where the concentration of ^{137}Cs decreases monotonically with depth. (Kirchner, 1998)

Conclusion

The diffusion-convection model of ^{137}Cs transport has been shown to describe the vertical profiles of ^{137}Cs in soil of the study area fairly. The whole experimental data did not actually fit perfectly into the model because some irregularities are normally found in vertical profiles attributed to global fallout. The diffusion and convection processes in ^{137}Cs migration in soil has been established by this study though some other processes different from the two processes might also take place in the transport phenomena. It has been shown that after 46 years the major deposition in the study area ^{137}Cs had diffused into the soil depth of 30 cm and can be expected to still diffuse into deeper layers of soil.

REFERENCES

Ajayi IR, Fischer HW, Burak A, Qwasmeh A, Tabot B (2007). Concentration and vertical distribution of ^{137}Cs in the undisturbed soil of southwestern Nigeria. Health Phys. 92(1): 73-77

Beresford NA, Howard BJ (1991). The importance of soil adhered to vegetation as a source of radionuclides ingested by grazing animals. Sci. Total Environ. 107: 237-254.

Bossew P, Gastberger M, Gohla H, Hofer P, Hubmer A (2004). Vertical distribution of radionuclides in soil of a grassland site in Chernobyl exclusion zone. J. Environ. Radioact. 73: 87-99.

Bunzl K (2001). Migration of fallout-radionuclides in the soil: effects of non-uniformity of the sorption properties on the activity-depth profiles. Radiat. Environ. Biophys. 40: 237-241.

Bunzl K, Forster H, Kracke W, Schimmack W (1994). Residence times of fallout $^{239+240}Pu$, ^{238}Pu, ^{241}Am, and ^{137}Cs in the upper horizons of an undisturbed grassland soil. J. Environ. Radioact. 22: 11-27.

Cheshire MV, Shand C (1991). Translocation and availability of radiocaesium in an organic soil. Plant Soil pp.287-296.

Davis JJ, Shultz V, Klement Jr. AW (1963). Caesium and its relationship to potassium in ecology. In. Radioecology., Reinhold, New York: 539-556.

Frissel MJ, Pennders R (1983). Models for the accumulation and migration of Sr-90, Cs-137, Pu-239,240 and Am-241 in the upper layer of soils in P. J Coughtrey (Ed.) Ecological Aspects of Radionuclide Release. Special Publication Series of the British Ecological Society, no. 3, Blackwell, Oxford. pp. 63-73.

Grzegorz P, Andrzej B, Zbigniew S (2003). Concentration and vertical distribution of ^{137}Cs in Agricultural and undisturbed soils from Chechlo and Czarnocin areas. J. Methods Appl. Absolute Chronol. Geochronometria 22: 67-72.

Herman T, Zech W, Lindner G (1992).The fate of Chernobyl-derived ^{137}Cs in forest soils of south Germany: determination of ^{137}Cs bearing soil components by density fractionation and sieving, in Union Internationale des Radioecologies (Ed), VIIIth Report of the Working Group Soil-to-plant Transfer Factors. Report of the meeting in Madrid, Spain. IUR-Publ. 92(12-02): 30-33.

Hillmann U, Schimmack W, Jacob P, Bunzl K (1996). In situ γ-spectometry several years after deposition of radiocasium part I. Approximation of depth distributions by the Lorentz function. Radiat. Environ. Biophys. 35: 297-303.

Kanunakara N, Somashekarappa HM, Narayana Y, Avadhani DN, Mahesh HM, Siddappa K (2001). 137Cs concentration in the environment of Kaiga of south west coast of India. Health Phys. 81(2): 148-155.

Kirchner G (1998). Applicability of compartmental models for simulating the transport of radionuclides in soil J. Environ. Radioact. 38(3): 339-352.

Kristic D, Nikezic D, Stevanovic N, Jelic M (2004). Vertical profile of 137Cs in soil. Appl. Radiat. Isotopes 61: 1487-1492.

Lee MH, Lee CW (1997). Disribution and characteristics of $^{239,240}Pu$ and ^{137}Cs in the soil of Korea. J. Environ. Radioact. 37(1): 1-16.

Likar A, Omahem G, Lipoglavsek M, Vidmar T (2001). A theoretical description of diffussion and migration of Cs in soil. J. Environ. Radioact. 57: 191-201.

Luigi M, John EB, Lars H, Rudie H, Jim TS, Mark Z (2003). Review and assessment of models used to predict the fate of radionuclides in lakes. J. Environ. Radioact. 69(3): 177-205.

Paulina S, Achim E, Gerald K (1997). Vertical migration of fallout ^{137}Cs in agricultural soils from southern Chile. Sci. Total Environ. 193: 197-205.

Schimmack W, Schultz W (2006). Migration of fallout radiocaesium in a grassland soil from 1986 to 2001 Part I: Activity-depth profiles of 134Cs and 137Cs. J. Environ. Radioact. 368: 853-862.

Schuller P, Bunzl K, Voigt G, Ellies A, Castillo A (2004). Global fallout ^{137}Cs accumulation and vertical migration in selected soils from South Patagonia. J. Environ. Radioact. 71: 43-60.

Szerbin P, Koblinger BE, Koblinger L, Vegvari I, Ugron A (1999). Ceasium-137 migration in Hungarian soils. Sci. Total Environ. 227: 215-227.

Victoria P (2007). Eckehard Modeling Cs-137 migration processes in lake sediments. J. Environ. Radioact. 96(1-3): 54-62.