Radiation Protection Dosimetry (2014), Vol. 160, No. 4, pp. 293–296
Advance Access publication 20 November 2013
doi:10.1093/rpd/nct302

MEASUREMENTS AND MODELLING OF ^{137}Cs DISTRIBUTION ON GROUND DUE TO THE CHERNOBYL ACCIDENT: A 27-Y FOLLOW-UP STUDY IN NORTHERN GREECE

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Following the Chernobyl accident, an area of ∼1000 m^2 in the University farm of the Aristotle University of Thessaloniki was considered as a test ground for radioecological measurements. The radiocesium deposition in this area, due to the Chernobyl accident, was 20 kBq m^{-2}. The profile of ^{137}Cs in the soil of this area was measured systematically from 1987 to 2012. The form of the profile has changed over the years. During the 1987–2000 period the ^{137}Cs distribution was reproducible by a sum of two exponentials. However, at least since 2005 the ^{137}Cs distribution can be successfully fitted by a single exponential function. The long-time (∼27 y) evolution study of the ^{137}Cs distribution in soil permit one to extract with the use of a simple compartment model, the mean vertical migration velocity of ^{137}Cs. Vertical migration of ^{137}Cs in soil is a very slow process. The mean vertical migration velocity is estimated to be 0.14 cm y^{-1}.The relative good comparison between the time dependence of the ^{137}Cs distribution in soil and the model predictions indicate that the simple model used is realistic.

INTRODUCTION

The main contribution on the long-term external dose from a nuclear accident (like Chernobyl or Fukushima) is due to the ^{137}Cs deposited in the environment. Knowledge of the distribution of ^{137}Cs deposited on soil is essential for a reliable assessment of the external dose[1, 2], as well as for root uptake by plants. Knowledge of its vertical migration is important in this regard and has been subject to research for a long time[3]. The experience gained from the radioecological studies due to the Chernobyl accident will facilitate the planning and interpretation of the radioecological studies in Japan due to the Fukushima accident.

Following the Chernobyl accident, an area of ∼1000 m^2 in the University farm of the Aristotle University of Thessaloniki was considered as a test ground for radioecological measurements. The radiocesium deposition in this area, due to the Chernobyl accident, was 20 kBq m^{-2}. The profile of ^{137}Cs in the soil of this area was measured systematically from 1987 to 2012. In the present study, the long-time (∼27 y) evolution study of the ^{137}Cs distribution in soil permit one to extract (with the use of a simple compartment model) the mean vertical migration velocity of ^{137}Cs.

MATERIALS AND METHODS

Samples of undisturbed soil have been periodically collected from 1987 to 2012 from an area of ∼1000 m^2 located at the University farm of the Aristotle University of Thessaloniki in Northern Greece. The main soil characteristics (e.g. pH 8, clay 22 %, silt 37 %, sand 41 %, organic matter 1.28 %, $CaCO_3$ 1.8 %) have been presented previously[4]. Slices (layers) of thickness of 5 cm were collected, down to a depth of 30 cm. It was ensured during the collection process that there was no cross contamination between the samples of successive depths. In case it happened, the collection process was repeated. The soil samples were stored in plastic bags, air dried in the greenhouse, passed through a 2-mm sieve and used for ^{137}Cs counting. The ^{137}Cs activity of each soil layer was measured by standard gamma spectroscopy, using high-purity germanium detectors of 20 % up to 50 % relative efficiency. About 30 such samplings and measurements were carried out from 1987 to 2012.

RESULTS AND DISCUSSION

Let $T(z)$ be the total activity of soil per unit area and per unit depth, at depth z. The measured activity $A(z)$ of the layer between z-5 cm and z cm is the integral:

$$A(z) = \int_{z-5}^{z} T(z)\mathrm{d}z \qquad (1)$$

The results presented here are expressed as the fractional contribution $R(z)$ of each layer to the total

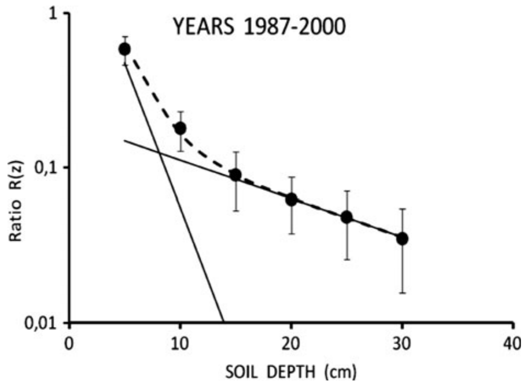

Figure 1. Average distribution of ^{137}Cs in soil, over the years 1987–2000. The bars show the standard deviation for each soil layer. The dashed line is the sum of the two exponentials presented by the two continuous lines.

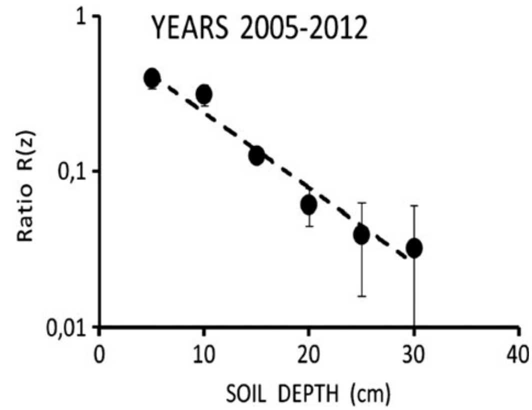

Figure 2. Average distribution of ^{137}Cs in soil, over the years 2005–12. The bars show the standard deviation for each soil layer. The mean ^{137}Cs distribution is well ($R^2=0.963$) fitted by a single exponential decrease.

deposition, i.e.

$$R(z) = \frac{\int_{z-5}^{z} T(z)\mathrm{d}z}{\int_{0}^{30} T(z)\mathrm{d}z} \qquad (2)$$

The ratio of the activities of ^{137}Cs and ^{134}Cs (back-dated to May 1986) when the latter was measurable[4], was found to be ~2 in all layers. Taking into account, that the same ratio 2 was recorded in air filters immediately after the arrival in Greece of the radioactive plume and that ^{134}Cs in soil, is only due to the Chernobyl accident, the authors can suppose that practically all ^{137}Cs in soil is due to the Chernobyl accident, i.e. nuclear weapon tests fallout is negligible. This assumption is also supported by the fact that in the first years after the Chernobyl accident, when ^{134}Cs was measurable, the $R(z)$ distributions of ^{137}Cs and ^{134}Cs were identical[5].

Figures 1 and 2 present the mean measured the distribution of fraction $R(z)$ of ^{137}Cs for the years 1987–2000 and 2005–12, respectively. Unfortunately, the authors did not perform measurements during the years 2001–04. As $R(z)$ is a fraction, there is no need for correction due to radioactive decay of ^{137}Cs. The circles in Figure 1 represent the average of 17 ^{137}Cs distribution measurements performed during the years 1987–2000 and the bars represent the standard deviation for each soil layer. The mean ^{137}Cs distribution can be reproduced (dashed line) by the sum of two exponentials presented in this semi-logarithmic scale by the two continuous lines. Since practically all ^{137}Cs in the soil is due to deposition from the Chernobyl accident, the bending of the profile cannot be attributed to ^{137}Cs from weapons fallout. In order to explain the form of the profile, pure diffusion and diffusion-advection models of ^{137}Cs migration were investigated[4]. It was found[4] that the pure diffusion model could not reproduce the double-exponential slope, while the diffusion advection model could. An interesting point is that the form of the profile has changed at least since 2005[5]. The circles in Figure 2 represent the average of 13 ^{137}Cs distribution measurements performed during the years 2005–12 and the bars represent the standard deviation for each soil layer. The mean ^{137}Cs distribution is well fitted ($R^2=0.963$) by a single exponential presented in this semi-logarithmic scale by a dashed line. The fact that the ^{137}Cs distribution can be fitted by a single exponential function does not mean necessary that the diffusion-advection model is not anymore valid. It has been shown previously[4], that for certain values of the parameters in the diffusion advection equation, the diffusion advection model gives a single slope for the cesium profile, practically that of the pure diffusion model.

The long-time (~27 y) evolution study of the ^{137}Cs distribution in soil permit one to study the time dependence of the $R(z=5)$ and $R(z=10)$ ratios. In Figure 3 the time evolution of the $R(z=5)$ ratio is presented. Despite the scattering of the values, one can clearly observe a small decrease over the years of the fractional contribution of the 0–5-cm layer.

From the decrease of the $R(z=5)$ values over time the authors can make a rough estimation of the mean vertical migration velocity with the use of a simple compartment model (Figure 4). In the compartment model, layers of soil are represented by compartments. In the simple model of this study only the three first layers are considered.

The differential equations describing the flow of ^{137}Cs through the compartments are:

$$\frac{\mathrm{d}R_5}{\mathrm{d}t} = -kR_5 \qquad (3)$$

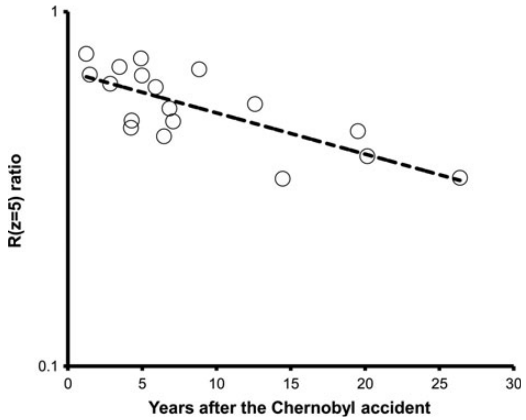

Figure 3. Time dependence of the $R(z=5)$ ratio. The dashed line is the best-fit exponential function passing through the data.

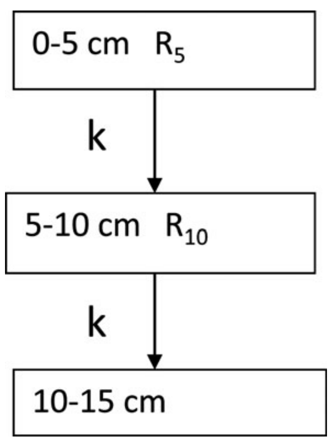

Figure 4. The schematic representation of the compartment model. R_5 and R_{10} are the $R(z=5)$ and $R(z=10)$ ratios, respectively.

$$\frac{dR_{10}}{dt} = kR_5 - kR_{10} \quad (4)$$

R_5 and R_{10} are the $R(z=5)$ and $R(z=10)$ ratios, respectively. k is the net transfer of ^{137}Cs (in y^{-1}) between the compartments. The authors remind that as R_5 and R_{10} are ratios (Equation 2), there is no need to take into account the decrease of ^{137}Cs 7 due to radioactive decay. The year of the Chernobyl accident (1986), which is the time $t=0$, the initial conditions of R_5 $(t=0)=0.71$ and R_{10} $(t=0)=0.1$ were deduced from the first ^{137}Cs distributions performed in the area. For deposition by heavy rainfall, as it was the case in Greece during the Chernobyl accident, radionuclides percolate with rainwater and vertical

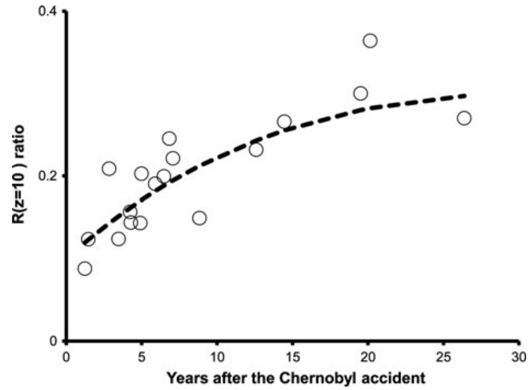

Figure 5. Time dependence of the $R(z=10)$ ratio. In dashed line are the results of Equation (6).

migration can be very fast during the early stage[3]. This is the reason that only a year after the Chernobyl accident ^{137}Cs could be found at even the depth of 30 cm. The solutions of the differential equations are:

$$R_5(t) = R_5(t=0)e^{-kt} \quad (5)$$

$$R_{10}(t) = \{(k \quad R_5(t=0)\,t) + R_{10}(t=0)\}e^{-kt} \quad (6)$$

In Equations (5) and (6) the only free parameter is the factor k. The authors are aware that the k factor is not constant between consecutive soil layers and increases with the soil depth[6]. However, if one use many free parameters it is easy to fit almost everything. The scope of this work is to investigate whether the use of one free parameter can permit to describe the time evolution of R_5 (t) and R_{10} (t).

From the decrease of the $R(z=5)$ values over time the authors can make a rough estimation of the mean vertical migration velocity. From the exponential function (Equation 5) passing through the data (Figure 3) the authors can deduce the k factor ($k=0.027$ y^{-1}), the mean resident time of ^{137}Cs in the layer 0–5 cm ($t=37$ y), and from that the authors can estimate the mean vertical migration velocity ($v=0.14$ cm y^{-1}). These values seems reasonable and are in the same range as values found in other studies (S. Almgen, M. Isaksson[3], and references therein).

In Figure 5 the time evolution of the $R(z=10)$ ratio is presented. As expected, an increase of R_{10} with time is observed. The interesting point is that the dashed line does not represent a fit to the experimental values but corresponds to model predictions (Equation 6). The relative good comparison between the experimental and calculated R_{10} (t) values indicates that the simple compartment model used in the present work is realistic.

CONCLUSIONS

The main conclusions are as follows:

- The form of ^{137}Cs profile in soil has changed over the years. During the 1987–2000 period the ^{137}Cs distribution in soil was reproducible by a sum of two exponentials. However, at least since 2005 the ^{137}Cs distribution can be successfully fitted by a single exponential function. Unfortunately, the authors had no measurements during the years 2001–04.
- The long-time (~27 y) evolution study of the ^{137}Cs distribution in soil permit one to extract (with the use of a simple compartment model) the mean vertical migration velocity of ^{137}Cs.
- Vertical migration of ^{137}Cs in soil is a very slow process. The mean vertical migration velocity is estimated to be 0.14 cm y^{-1}.
- The relative good comparison between the experimental and calculated $R_{10}(t)$ values indicates that the simple compartment model used in the present work is realistic.

REFERENCES

1. Anderson, K. G. and Roed, J. *The behavior of Chernobyl ^{137}Cs, ^{134}Cs and ^{106}Ru in undisturbed soil: implications for external radiation*. J. Environ. Radioact. **22**, 183–196 (1994).
2. Kocher, D. C. and Sjoreen, A. L. *Dose-rate conversion factors for external exposure to photon emitters in soil.* Health Phys. **48**, 192–208 (1985).
3. Almgen, S. and Isaksson, M. *Vertical migration studies of ^{137}Cs from nuclear weapons fallout and the Chernobyl accident*. J. Environ. Radioact. **91**, 90–102 (2006).
4. Antonopoulos-Domis, M., Clouvas, A., Hiladakis, A. and Kadi, S. *Radiocesium distribution in undisturbed soil: measurements and diffusion-advection model*. Health Phys. **69**, 949–953 (1995).
5. Clouvas, A., Xanthos, S., Takoudis, G., Antonopoulos-Domis, M., Ginoviadis, G., Vidmar, T. and Likar, A. *Twenty-year follow-up study of radiocesium migration in soil*. Radiat. Prot. Dosim. **124**, 372–377 (2007).
6. Antonopoulos-Domis, M., Clouvas, A. and Marseguerra, M. *On the compartmental modeling of caesium migration in soils.* Nucl. Sci. Eng. **121**, 461 (1995).