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Temporal reduction of the external gamma dose rate due to ^{137}Cs mobility in sandy beaches

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In the present paper the contribution to the external gamma dose rate due to ^{137}Cs in soil as a function of time is presented. Sampling sites were selected along the Calabria and Basilicata Regions coastal beaches (southern part of Italy) to assess the external gamma dose rate in air, 1 m above the ground level. A convection–dispersion model, with constant parameters was used to approximate the radiocesium soil vertical migration. The model was calibrated using the initial ^{137}Cs activity deposition in this region (Chernobyl fallout) and ^{137}Cs activity concentration down the soil profile, measured 10 years later. The dispersion coefficient and the advection velocity values, were respectively: 2.17 cm^2 y^{-1} and 0.32 cm y^{-1}. The Radionuclide Software Package (RSP), which uses a Monte Carlo simulation code, was used to determine the primary ^{137}Cs gamma dose contribution in air 1 m above the ground surface. The resulting ^{137}Cs external dose rate ranged from 0.42 nGy h^{-1} in 1986, to 0.05 nGy h^{-1} in 2007.

Introduction

After an accidental or intentional release of radioactivity in the environment, radionuclides deposited on the ground surface could significantly contribute to the external gamma radiation exposure of the population and biota. This exposure declines rapidly during the initial time period due to the decay of short-lived radionuclides, and also as a result of the migration of radionuclides down the vertical soil profile. The vertical migration of radionuclides along the soil profile is the principal factor contributing to the decline of external irradiation from long-lived radionuclides, such as ^{137}Cs.[1,2]

The assessment of the external gamma irradiation is extremely complex due to the large number of environmental factors which affect the photon flux in air originating from the ground surface. The radionuclide soil vertical distribution, the mobility of radionuclides, the soil surface roughness, and the soil bulk density and its variations with depth, constitute the main factors to be considered in this assessment.[3]

In a previous study,[4] this methodology was used to assess the contribution to the external gamma dose rate of ^{137}Cs and ^{40}K in soil, and the fraction originated by cosmic radiation at the same sampling site. These contributions were compared with the gamma dose rate measured at each sampling site, and the results showed that they account for almost 70% of the measured gamma dose rate.

This paper assesses how the radiocesium mobility along the vertical soil profile could influence its contribution to the external gamma dose rate. To describe the radiocesium soil vertical transport process, a dispersion–advection model[5] was calibrated using the ^{137}Cs deposited on the ground surface due to the Chernobyl event (1986) and the ^{137}Cs activity concentrations measured along the soil profile 10 years later. The external dose rate contribution attributable to ^{137}Cs measured along the soil profile, as a function of time, was then assessed using the Radioecological Software Package (RSP).[6]

The data used in the present study refer to the environmental radioactivity investigation carried out in 1997 by the Italian Environmental Protection Agency (Istituto Superiore per la Protezione e la Ricerca Ambientale – ISPRA) along the coasts of the Calabria and Basilicata regions (southern part of Italy).[7–8]

Experimental

The sand sampling points were selected along the Italian southeast and southwest coasts. Geographical coordinates of the sampling sites, soil properties, and experimental procedures have been described in a previous paper.[4]

At each sampling point, sub-samples over an area of 900 cm^2 were collected at different depth layers along the vertical profile (0–2 cm, 2–5 cm, 5–10 cm, 10–15 cm, and 15–20 cm) at the corners and center of a 10 meter square. ^{137}Cs activity concentrations were determined by high-resolution gamma spectrometry using HPGe detectors.

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Theoretical approach for radiocesium soil migration

A convection-dispersion model with constant parameters was used to describe the ^{137}Cs soil vertical mobility.[10] In this approach it is assumed that the flow is one-dimensional and the soil moisture content is constant. The concentration of a radionuclide in solution is given as:

$$\frac{\partial R(z,t)}{\partial t}+\frac{\rho}{\varepsilon}\frac{\partial S(z,t)}{\partial t}=D\frac{\partial^2 R(z,t)}{\partial z^2}-\\ -v_w\frac{\partial R(z,t)}{\partial z}-\lambda\left[R(z,t)+\frac{\rho}{\varepsilon}S(z,t)\right] \qquad (1)$$

where $R(z,t)$ (Bq cm^{-3}) is the ^{137}Cs concentration in resident fluid at depth z (cm) and at time t (y), $S(z,t)$ (Bq g^{-1}) is its sorbed concentration in the soil matrix, D (cm^2 y^{-1}) is its dispersion coefficient, v_w (cm y^{-1}) is the mean pore water velocity, ρ (g cm^{-3}) is the bulk density of the soil and ε its porosity, and λ (y^{-1}) is the ^{137}Cs physical decay constant.

Assuming that the sorption processes can be described by a linear isotherm

$$S(z,t)=K_d R(z,t) \qquad (2)$$

where K_d (cm^3 g^{-1}) is the distribution coefficient of ^{137}Cs. Then Eq. (1) is:

$$\frac{\partial C(z,t)}{\partial t}=D_s\frac{\partial^2 C(z,t)}{\partial z^2}-v_s\frac{\partial C(z,t)}{\partial z}-\lambda C(z,t) \qquad (3)$$

where

$$C(z,t)=\left(1+\frac{\rho}{\varepsilon}K_d\right)R(z,t) \qquad (4)$$

represents the total ^{137}Cs concentration (Bq cm^{-3}), including both the mobile and the sorbed radiocesium.

The parameters D_s and v_s are, respectively, the effective dispersion and the effective advection coefficient for the total ^{137}Cs concentration $C(z,t)$ (Bq cm^{-3}). Both parameters are dependent on the soil properties and other environmental conditions. The parameters D_s and v_s are related to D and v_w in this manner:

$$D_s=\frac{D}{1+\frac{\rho}{\varepsilon}K_d} \quad v_s=\frac{v_w}{1+\frac{\rho}{\varepsilon}K_d} \qquad (5)$$

Assuming that:

a) The fallout deposition is approximated by an individual of the pulse-like inputs

$$C_{pulse}=C_0\,\delta(t-t_0) \qquad (6)$$

where C_o (Bq m^{-2}) is the ^{137}Cs deposition activity concentration at the earlier time $t=t_0$, and δ is the Dirac delta function.

b) The soil column is semi-infinite and the initial condition is:

$$C(z,0)=0, \qquad 0\le z\le\infty \qquad (7)$$

c) The boundary conditions, which state that there are no current leakage activities across the soil surface:

$$\left(-D_s\frac{\partial C(z,t)}{\partial z}+v_s C(z,t)\right)=0 \;\; for\; z=0 \;\;\; and\; t>0 \qquad (8)$$

Then, the solution of Eq. (3) is:[11]

$$C(z,t)=C_o\,exp\left[-\lambda(t-t_o)\right]\times\left\{\frac{1}{\sqrt{\pi D_s(t-t_o)}}exp\left[-\frac{(z-v_s(t-t_o))^2}{4D_s(t-t_o)}\right]\right.$$

$$\left.-\frac{v_s}{2D_s}exp\left[\frac{v_s}{D_s}z\right]erfc\left[\frac{(z+v_s(t-t_o))}{\sqrt{2D_s(t-t_o)}}\right]\right\} \qquad (9)$$

In this equation $erfc(\xi)=1\text{-}erf(\xi)$, where $erf(\xi)$ is the error function.

Temporal changes in dose rate assessment

The external dose rate is usually evaluated using the Dose Rate Factor (DRF) by means of the following equation:[6,12]

$$\dot{H}(t) = C(t)\,DRF \qquad (10)$$

where, $\dot{H}$ (Gy h^{-1}) is the external dose rate at time t; C is the soil volumetric activity density (Bq cm^{-3}) and DRF (Gy h^{-1} per Bq cm^{-3}) is the dose rate conversion factor.

DRF values in air at 1 m above the ground surface can be analytically calculated for a wide range of discrete photon energies for sources uniformly distributed in a slab of finite thickness or exponentially distributed with depth.[12] Alternatively algorithms based on a Monte Carlo method have been developed to simulate the photon transport calculation for the soil/air configuration.[6,13]

Eq. (10) can be used to assess the external dose rate due to a soil layer of thickness Δz_i situated between the depths z_{i-1}, z_i, ($\Delta z_i = z_i - z_{i-1}$). In fact, if in this soil layer the mean activity concentration is $\overline{C}_i$, and $DRF(z_i)$ is the dose rate factor corresponding to a soil layer between ground and z_i, then the external dose rate contribution due to this internal soil layer is:

$$\dot{H}_i(t)=\overline{C}_i(t)\left[DRF(z_i)-DRF(z_{i-1})\right] \qquad (11)$$

In order to assess the contribution to the external gamma dose rate due to ^{137}Cs, as a function of time, the following steps were carried out:

1. In considering the initial activity deposition in each site and the soil activity concentration measured almost 11 years later, the coefficients D_s and v_s were obtained by fitting Eq. (9).

2. By spatial integration of the function $C(z,t)$ [Eq. (9)] between the limits z_{i-1} and z_i, the mean activity concentration value ($\overline{C_i}$) was assessed, for each soil layer, as a function of time.

3. Using Eq. (11) and RSP,[6] the contribution to the external dose rate due to each soil layer was calculated as a function of time.

The total external dose rate was assessed by summing the partial contribution of each soil layer.

Results and discussion

Figure 1 shows the theoretical curves of $C(z,t)/C_0$ vs z, for four different times since the initial deposition. In this figure, 132 months (middle of 1997) corresponds to the sampling time. The last time considered, 252 months, was approximately the middle of 2007. The D_s and v_s values obtained, respectively, were: 2.17 cm^2 y^{-1} and 0.32 cm y^{-1}. The D_s value is one order of magnitude higher than the values reported by other authors for this parameter, although the v_s value is similar to those reported in such studies.[14,15] The significant difference in the value of the dispersion coefficient could be attributable to the differences in the soil composition. In fact, in a previous research,[16] using a similar mathematical model, it was shown that radiocesium from the Chernobyl accident deposited in natural ecosystems of other Italian regions, had demonstrated lesser mobility to deeper soil than in the present study.

For the same times considered in Fig. 1, and for the soil layer with depths: 0–1 cm; 1–3 cm; 3–5 cm and 5–10 cm, in Fig. 2 have been represented with vertical bars, the mean areal activity concentration (Bq m^{-2}) obtained by integration of Eq. (9).

Considering the estimated initial activity deposition[4] of 210 Bq m^{-2}, Fig. 3 shows the total external dose rate contribution due to ^{137}Cs content in soil (black bar) and partial contributions due to the same soil layers considered in Fig. 2. In Figure 3, dose rate was evaluated using Eq. (11) for 6 different times, including the times taken into account in Fig. 2.

The attenuation of the estimated external dose rate can be acceptably fitted by an exponential decreasing function (Fig. 3):

$$\dot{H}(t) = \dot{H}_0 exp(-\lambda_{eff} t) \tag{12}$$

Where $\dot{H}_0$ is the external dose rate at deposition time (t=0) and λ_{eff} is the effective characteristic parameter of the exponential function. This parameter can be associated with the effective half life of the dose rate ($t_{1/2eff}$) (time taken for the dose rate to decay to half of its initial value), by the equation:

$$t_{1/2,eff} = ln 2 / \lambda_{eff} \tag{13}$$

Previous researchers have modeled this decreasing behavior of the external dose rate (see for example, Ref. 2). The behavior is attributable to two independent phenomena:

a) The radionuclide physical decay. This natural process is characterized by the parameters λ_{phys}, and $t_{1/2,phys}$.

b) The downward radionuclide migration to deeper soil. The contribution of this effect on the temporal change in the external dose rate, Eq. (12), has been characterized by the parameter: λ_{mig}, and $t_{1/,2mig}$.

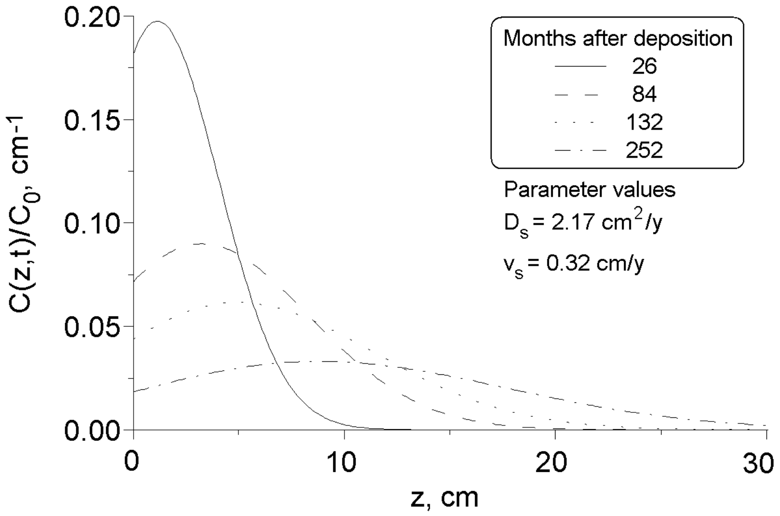

Fig. 1. Theoretical approach for the normalized ^{137}Cs activity concentration as a function of soil depth, for four time periods after deposition. Dispersion and advection coefficients were obtained by fitting experimental data

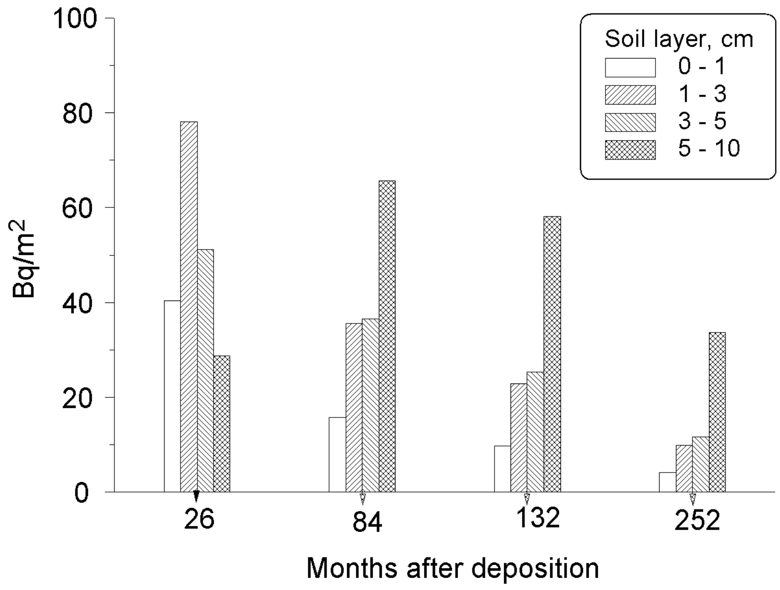

Fig. 2. ^{137}Cs calculated activity concentration in different soil layers, for four time periods after deposition

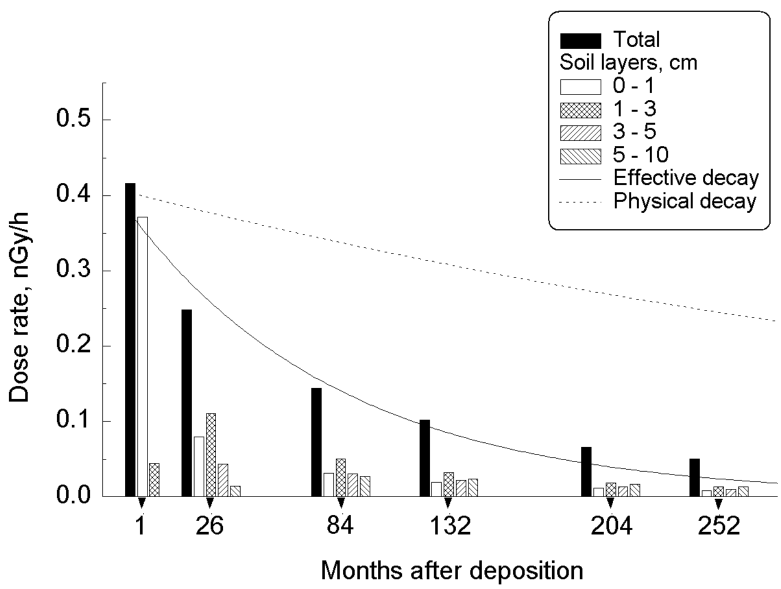

Fig. 3. External dose rate due to ^{137}Cs activity content in soil as a function of time after deposition: Total contribution and partial contribution of different soil layers. Continuous line denotes the theoretical approach of the dose decay as a function of time, dash line shows the physical radionuclide decay

Table 1. Exponential decay constant and half life for the external dose rate: physical, downward migration and effective values

Parameter	Physical	Downward migration	Effective
$\gamma (y^{-1})$	0.023	0.103	0.126
$t_{1/2}$ (y)	30.2	6.73	5.50

These quantities verify the relationship:

$$\lambda_{eff} = \lambda_{phys} + \lambda_{mig} \tag{14}$$

Or, equivalently:

$$\frac{1}{t_{1/2,eff}} = \frac{1}{t_{1/2,phys}} + \frac{1}{t_{1/2,mig}} \tag{15}$$

By fitting the calculated external dose rate using Eq. (12) (Fig. 3, continuous line) we obtain $\dot{H}_0$ = 0.36 nGy h^{-1} and λ_{eff} = 0.126 y^{-1} (R^2 = 0.96).

Table 1 gives the exponential decay constants and half lives associated with each individual process that affects the calculated temporal variation of the external dose rate.

Conclusions

This paper has calculated the variation of dose rate for ^{137}Cs in soil, due to its downward mobility through time. In general, the external contribution to gamma dose due to radionuclides in soil in equilibrium is known. The present study is done by applying two models: the first one takes into account the vertical radionuclide mobility, while the second simulates the dose rate using Monte Carlo. The calibration of the first model with experimental data for ^{137}Cs allows us to determine the dispersion coefficient and the convection velocity for the particular conditions of the sites studied. Through the simulation we know the attenuation in external dose intensity as a function of time due to the source mobility. An exponential decreasing function provides a good fit to the attenuation of the external dose rate and the elapsed time from the initial deposition. For the particular situation investigated, we obtained that the effective half life of the external dose rate was 5.49 years. The results have some relevance from the viewpoint of radioprotection and for possible actions to be taken in contaminated areas.

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