Radiocesium in the Northeastern Part of Italy After the Chernobyl Accident: Vertical Soil Transport and Soil-to-Plant Transfer

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ABSTRACT

Radiocesium deposited in soil following the Chernobyl accident (May 1986) has remained persistently available for plant uptake in some areas of Europe. In this paper, the RABES model was used to explain the vertical migration of $^{137}$Cs in soil, in three different areas in the Northeastern region of Italy. The soil-to-plant transfer factor has been calculated as a function of the elapsed time since the radionuclide deposition. The influence of the soil characteristics on the transfer process was studied. © 1997 Published by Elsevier Science Ltd.

INTRODUCTION

In case of accidental releases, the transfer of radionuclides through food chains is one of the main reasons of radiation exposure to the population. Principally radionuclides of a long half-life are important in the internal exposure of humans due to the ingestion of contaminated foodstuffs. In this process, the radionuclide vertical migration in soil toward the root zone and the subsequent root uptake have a singular importance.
Radiocesium deposited in soil following the Chernobyl accident (May 1986) has remained persistently available for plant uptake in some areas of Europe for many years.

In this paper, we study the $^{137}\text{Cs}$ distribution in soil and the soil-to-plant transfer process in some areas situated in the Northeastern region of Italy. In a previous paper (Velasco et al., 1993), a dynamic model was developed and calibrated to study the migration of $^{137}\text{Cs}$ in undisturbed soil profile. The model, called RABES, is based on the supposition that the radionuclide vertical distribution pattern can be described by a decreasing exponential function, where the characteristic coefficient of the distribution (alpha-factor) is dependent on the elapsed time since the deposition. In this paper, the RABES model has been calibrated with new experimental data. The dynamic process of root uptake has been determined using the soil-to-plant transfer factor. This factor can be expressed in terms of just two parameters. The first one depends on the rate of root uptake, while the other one is a measure of the rate at which radiocesium in soil is unavailable for root uptake.

The model predicts, with reasonable accuracy, the variation of activity levels in soil and vegetation.

**MATERIALS AND METHODS**

Three sampling areas have been selected in the mountainous area of the Friuli-Venezia Giulia region, where the $^{137}\text{Cs}$ deposition following the Chernobyl accident ranged from 20 to 40 kBq m$^{-2}$ (Belli et al., 1990). The radiocesium vertical migration along surface soils and the soil-to-plant transfer have been studied in this region.

Sampling soils was carried out with seasonal frequency from July 1987 to July 1992. The periodic soil samples were taken at distances of at least 1 m apart to avoid interferences.

A soil monolith of surface area 30 cm x 30 cm was isolated by digging a trench. Soil samples were taken to a depth of 20 cm, dividing the monolith in horizontal layer of thicknesses 5 cm. The error associated with this sampling method is about 20% in homogeneous areas.

All soil samples were air-dried and sieved through a 2-mm mesh, and the litter samples were oven-dried at 1000°C and then ground. All samples were analyzed by gamma spectrometry using HPGe detectors.

The following analyses were also performed on the soil samples: texture, cation exchange capacity, content of organic matter and pH. The total measured $^{137}\text{Cs}$ was separated into its weapons fallout and Chernobyl
contributions using the $^{137}\text{Cs}:^{134}\text{Cs}$ ratio of 2, measured in air rainfall samples gathered in May 1986.

Forage crop samples were collected in the three sampling stations, four times a year, during the growth period, from 1988 to 1991. Areas 1 and 2 are natural grassland, while the vegetation type in Area 3 was medicinal herbs. The grasses were cut 2 cm above the soil level over an area of 1 m², and dried at 60°C.

**Model application**

The radionuclide, vertical distribution patterns in soil have been described, in the RABES model, by the following equations:

$$C(x, t) = Q_{T0} \alpha(t) \exp[-\alpha(t)x] \exp(-\lambda t), \quad (1)$$

where $C$ (Bq kg⁻¹) is the radionuclide concentration at depth $x$ (cm) and at depth $x$ (cm) at time $t$ (days after the initial deposition); $Q_{T0}$ (Bq kg⁻¹ cm⁻¹) (initial integrated concentration) is the definite spatial integral of $C(x, 0)$, equivalent to the initial radionuclide deposition $Q_0$ (Bq cm⁻²) divided by the soil density (kg cm⁻³); $\lambda$ (day⁻¹) is the radioactive decay constant, and $\alpha$ (cm⁻¹) is called the alpha-factor of the distribution. The alpha-factor has been assumed to be an exponential function of time as follows:

$$\alpha(t) = \alpha_0 \exp(-\alpha_1 t) + \alpha_2, \quad (2)$$

where $\alpha_0 + \alpha_2$ (cm⁻¹) is the alpha-factor value as $t$ approaches to zero, $\alpha_2$ (cm⁻¹) is the alpha-factor in the equilibrium condition, and $\alpha_1$ (day⁻¹) is the rate of change of the vertical distribution.

According to the RABES model, any horizontal surface in the soil has uniform concentration (horizontal symmetry). This concentration depends only on the depth of this surface. It is possible to deduce a theoretical expression for the vertical velocity of this surface. In fact, if, at depth $x_m$, the radionuclide concentration were $C_m$, eqn (1) leads to

$$x_m = -\frac{1}{\alpha(t)} \frac{d\alpha}{dt} \left[ \log \left( \frac{C_m}{Q_{T0}} \right) - \log[\alpha(t)] + \dot{\lambda} t \right]. \quad (3)$$

Then, the vertical velocity $v_m$ of the horizontal surface with concentration $C_m$ is:

$$v_m = \frac{dx_m}{dt} = \frac{1}{\alpha(t)^2} \frac{d\alpha}{dt} \left[ \log \left( \frac{C_m}{Q_{T0}} \right) - \log[\alpha(t)] + \dot{\lambda} t + 1 \right] - \frac{\dot{\lambda}}{\alpha(t)}. \quad (4)$$
Soil-to-plant transfer

The dynamics process of root uptake has been determined using the well known transfer factor (TF) relating soil concentration in the root zone to plant concentration of contaminants, which are usually defined as (Bergeijk et al., 1992):

\[
TF = \frac{^{137}\text{Cs concentration in dry vegetation (Bq kg}^{-1})}{^{137}\text{Cs concentration in dry soil (Bq kg}^{-1})}.
\] (5)

The TF factor for radiocesium is a continuous time decreasing function because of the fixation process of the radionuclides in the soil. Noordijk et al. (1992) have estimated this decrease to be a factor of 1.5 after 1–2 years, up to a factor of 4 after 7 years. Antonopoulos-Domis et al. (1990) and Müller and Pröhl (1993) suggest that the radionuclide availability for root uptake is an exponential decreasing function of time. In the same way, we propose that TF, in undisturbed soil, can be expressed as follows:

\[
TF(t) = TF_0 \exp(-\gamma t). \tag{6}
\]

In the last equation, \(TF_0\) depends on the radionuclide, the soil characteristics (Bergeijk et al., 1992) and on the weather conditions. The constant \(\gamma\) (day\(^{-1}\)) is the fixation rate of cesium in the soil. It is usually assumed that \(\gamma\) (day\(^{-1}\)) depends on the relative abundance of clay (Noordijk et al., 1992).

If we assume that, in natural grassland, the root depth zone is conformed by the first \(x_r\) (cm) of soil, then, according to the RABES model, the mean soil concentration in this soil layer is:

\[
<C(x_r, t)> = \frac{Q_{T0}\exp(-\lambda t)}{x_r} [1 - \exp(-\alpha(t)x_r)]. \tag{7}
\]

Combining the previous equation, we conclude that the theoretical expression for the radiocesium concentration in dry vegetation, \(C_v\), is:

\[
C_v(t) = TF_0 \exp(-\gamma t) < C(x_r, t) >. \tag{8}
\]

RESULTS AND DISCUSSION

Table 1 shows the main soil properties in each area. The textures of the three soils are similar to each other. The variation range of the principal properties is: clay contents from 14 to 21%; organic matter (O.M.) from 2.3 to 14%; and such variation is accompanied by a change in the cation
<table>
<thead>
<tr>
<th>Area</th>
<th>Soil layers (cm)</th>
<th>Sand (%)</th>
<th>Silt (%)</th>
<th>Clay (%)</th>
<th>pH (potassium chloride)</th>
<th>Cation exchange capacity (meq 10^-2 g^-1)</th>
<th>Organic matter (%)</th>
</tr>
</thead>
<tbody>
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<td>60</td>
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<td>7.2</td>
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<td>9.2</td>
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</table>

*a meq = milliequivalent.*
exchange capacity (C.E.C.) in a range of 0.24–0.55 meq g\(^{-1}\) with respect to the pH values, Areas 2 and 3 are similar, while Area 1 presents a lower value.

Figure 1 graphically shows the \(^{137}\)Ch concentration (Chernobyl contribution) in each area. For each sampling time, and taking into account the total \(^{137}\)Cs found, Fig. 1 shows the percentages corresponding to each of the four soil layers considered (0–5 cm, 5–10 cm, 10–15 cm and 15–20 cm).

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**Fig. 1.** The \(^{137}\)Cs distribution in soil. The vertical bars represent, for each sampling time, the percentual concentration measured in each one of the soil layer considered: 0–5 cm, 5–10 cm, 10–15 cm and 15–20 cm.
In all cases, we assumed that \( t = 0 \) (initial deposition time) corresponds to the Chernobyl accident date, May 1986.

The RABES model (Velasco et al., 1993) was used to describe the vertical migration in soil. The values obtained for the parameters \( \alpha_0 \), \( \alpha_1 \) and \( \alpha_2 \), during the model calibration, are summarized in Table 2. It is important to remark on the value of the sum \( \alpha_0 + \alpha_2 \) for each area. This sum is equivalent to the alpha-factor value when \( t \) approaches zero, a result proportional to the initial integrated concentration in soil. In fact the sum \( \alpha_0 + \alpha_2 \) for Area 1 is 0.39 cm\(^{-1}\) where \( Q_{T0} \) is 3.8E + 03 Bq cm kg\(^{-1}\), for Area 2 the sum \( \alpha_0 + \alpha_2 \) is 0.89 cm\(^{-1}\) and \( Q_{T0} \) is 4.0E + 03 Bq cm kg\(^{-1}\), and for Area 3 the sum \( \alpha_0 + \alpha_2 \) is 0.29 cm\(^{-1}\) and \( Q_{T0} \) is 8.7E + 02 Bq cm kg\(^{-1}\). The value \( \alpha_1 \) represents the time variation of the radionuclide distribution along the soil profile. The value obtained in Area 2 is greater in one order of magnitude than the one obtained for the other areas. However, the relation \( \alpha_1/Q_{T0} \) (radionuclide mobility per unit of initial deposition) that is reported in the right column of Table 2, has the same order in all the cases.

Figure 2 shows the values of alpha-factor, calculated for Areas 1, 2 and 3, respectively, together with the theoretical curves of \( \alpha(i) \).

Figure 3 shows the vertical distribution of \( ^{137}\text{Cs} \) concentration obtained in soil profile in each area. The experimental values and the theoretical curves are reported for three different times (including the first and the last sampling time).

Using equation (4), the vertical migration velocity for a horizontal surface \( C_m \) was evaluated. We consider, as a reference, a value of \( C_m \) (Bq kg\(^{-1}\)) 1000 times lower than that of the initial integrated concentration \( Q_{T0} \) (Bq kg\(^{-1}\) cm). Figure 4 shows the theoretical curves of this velocity for each sampling area. The larger value of this velocity for Area 2 in the first 3 years, is in agreement with the greater values of \( \alpha_1 \) and the initial deposition in this area.

The soil-to-plant transfer factor in each area was performed using equation (5). The mean concentration in the root zone has been evaluated using equation (4) with \( x_r = 10 \) cm, as suggested by Müller and Pröhrl (1993) for natural grassland. The values obtained in each area for the

<table>
<thead>
<tr>
<th>Area</th>
<th>( \alpha_0 ) (cm(^{-1}))</th>
<th>( \alpha_1 ) (day(^{-1}))</th>
<th>( \alpha_2 ) (cm(^{-1}))</th>
<th>( \alpha_1/Q_{T0} ) (kg Bq(^{-1}) cm(^{-1}) day(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Area 1</td>
<td>2.9E-01</td>
<td>6.0E-04</td>
<td>1.0E-01</td>
<td>1.6E-07</td>
</tr>
<tr>
<td>Area 2</td>
<td>6.1E-01</td>
<td>2.5E-03</td>
<td>2.0E-01</td>
<td>6.3E-07</td>
</tr>
<tr>
<td>Area 3</td>
<td>2.4E-01</td>
<td>5.2E-04</td>
<td>7.0E-02</td>
<td>5.9E-07</td>
</tr>
</tbody>
</table>

TABLE 2
Coefficient Values Obtained for Each Area
Fig. 2. Estimated values of the alpha-factor for each area. The theoretical curves are obtained from equation (2).

Parameters $T_{F0}$ and $\gamma \text{(day}^{-1})$ are, respectively, as follows: in Area 1, 9·0E-02 and 7·7E-04; in Area 2, 2·1E-01 and 1·0E-03; in Area 3, 4·5E-01 and 1·7E-01. According to Bergeijk et al. (1992), radiocesium transfer increases with increasing organic matter content. In this manner the difference of the value of $T_{F0}$ for Areas 1 and 2, could be attributed to the greater organic matter content in Area 2. We attribute the sensibly different value of $T_{F0}$ in Area 3, to the vegetation type of this Area (medicinal herbs).
Area 3 presents the maximum value for $\gamma$ (this parameter measures how radiocesium becomes inaccessible for the plant), that could be explained because of the greater content of clay and the lower content of organic matter in this area. The importance of clay abundance in the fixation process of cesium to the soil is generally recognized (Cremers et al., 1988; Noordijk et al., 1992). On the other hand, radiocesium forms a reversible ion exchange complex with the organic matter, and remains available for plant for extended periods of time (Cremers et al., 1990).
The theoretical curves for $^{137}$Cs concentration in vegetation ($C_v(t)$) was obtained introducing the correspondent $T_{F0}$ and $\gamma$ in equation (8). Figure 5 shows these curves and the experimental values corresponding to the $^{137}$Cs concentration measured in vegetation, from 1988 to 1991.

REFERENCES


Fig. 5. $^{137}$Cs concentration in vegetation: experimental values and theoretical curves.

of pH, soil type and soil organic matter content on soil-to-plant transfer of radiocesium and strontium as analyzed by a nonparametric method. *J. Environ. Radioactivity*, 15, 265–76.


