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COMPARTMENTAL MODEL FOR UPTAKE OF ^{137}CS BY PINE IN FOREST SOIL

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ABSTRACT

A compartmental model of soil to pine tree transfer of ^{137}Cs following the Chernobyl nuclear accident is presented. The model was validated using data collected in 1996 at five sites in Northern Ukraine. The transfer constants of ^{137}Cs between model compartments are estimated using a semi-empirical method. The concentration ratios obtained show similar observed and predicted results at Ditiatki and Pripiat 1 & 2, while predicted values were not close to observed values at Kopachi and Pripiat 3 sites. The division of the trunk and the bark into three parts is shown to be useful and the branches, needles and bark are demonstrated to be reservoir compartments.

KEYWORDS: forest, ^{137}Cs transfer, compartmental model, pine.

INTRODUCTION

Since the Chernobyl nuclear accident in 1986, long-term radioactive pollution of the environment, particularly of the soil and vegetation, has been a significant problem. In Belarus for example, about 1.5x10^{6} ha of forested lands were contaminated with 40-190 kBq m^{-2} and 2.5x10^{4} ha received more than 1,480 kBq m^{-2} of ^{137}Cs and other long-lived radionuclides such as ^{90}Sr and ^{239,240}Pu (Schell et al., 1996). Many measurements have been made and various models proposed by researchers with the aim of understanding long-term processes of redistribution and recirculation of radionuclides in the environment (Muramatsu et al., 1989; Antonopoulos-Domis et al., 1990; Rybacek et al., 1992; Korun et al., 1994; Mamikhin, 1995; Rauret et al., 1995; Carini, 1999 & 2001; Mitchell, 2001). Modelling is a fundamental tool in assessing and understanding these processes and is essential in predicting doses received by human and non-human organisms (Garnier-Laplace et al., 1997).

Models focussed on soil-to-plant transfer of radiocaesium can often give undue weight to root uptake as the primary mechanism of contamination of perennial plants (UNSCEAR, 1977). Antonopoulos-Domis et al. (1990) challenged this assertion: their model, based on experimental observations, proposed that root uptake might be neglected for the first few years following deposition as well as for long-term contamination. In this case, contamination levels may be described by a single exponential term that describes foliar absorption. Experimental results also suggest that recirculation of the radiocaesium inventory within the body of perennial plants is a significant mechanism involved in accumulation and storage of radiocaesium discharged to the atmosphere through nuclear weapon testing and nuclear accidents (Yoshida et al., 2003). It is important to capture mechanisms such as foliar absorption and biological recirculation within models of long-term radiocaesium behaviour in the environment if those models are to be useful as tools for dose assessment and environmental management. We present here a dynamic model for the prediction of ^{137}Cs concentrations in soils and pine trees at highly contaminated sites in the vicinity of the Chernobyl Nuclear Power Plant. Miscellaneous compartmental models developed since the Chernobyl accident has provided the background for the development of our model (Whicker and Kirchner, 1987; Müller and Pröhl, 1993; Rosen et al., 1995; Mamikhin, 1995; Mayall, 1995; Perianez and Martinez-Anguirre, 1997; Fesenko et al., 1997; Kirchner, 1998; Whicker et al., 1999; Toal et al., 2001; Bulgakov and Konoplev, 2002). Specifically, we describe the structure, calibration and validation of a dynamic model of radiocaesium transfers in pine trees, which have considerable ecological, economic and social importance in Northern and Eastern European countries. Pine trees are used for the manufacture of furniture and paper and can also be used as firewood. The use of wood, in general, can contribute to human radiation exposures in a variety of ways (IAEA, 2004).

Methodology

This section describes the model structure, assumptions made and an evaluation of the model parameters used.

Model description

Our model consists of 10 compartments, namely soil, root, trunk bottom, trunk middle, trunk top, branches, needles, bark bottom, bark middle and bark top with respective numbers $i = 0, 1, 2, \ldots, 9$. These compartments represent the major ecological storage 'units' within a typical mature pine forest, with four structural components of pine trees (roots, trunk, branches and needles) being explicitly represented. Each of these components is easily sampled and analysed for radionuclide contents. Such measurements indicate that activity concentrations of ^{137}Cs in pine trees older than 25 years (generally >9m in height) vary with height along the trunk (Fesenko et al., 2001). Thus, in our model we have divided the trunk vertically into three separate compartments. Physiologically, this is defensible since the lower part of the trunk bottom can be considered to provide the source of upwardly moving xylem sap, the trunk top provides the source of phloem sap and the middle region of the trunk provides a zone of transit for sap moving in both xylem and phloem (Heller, 1981).

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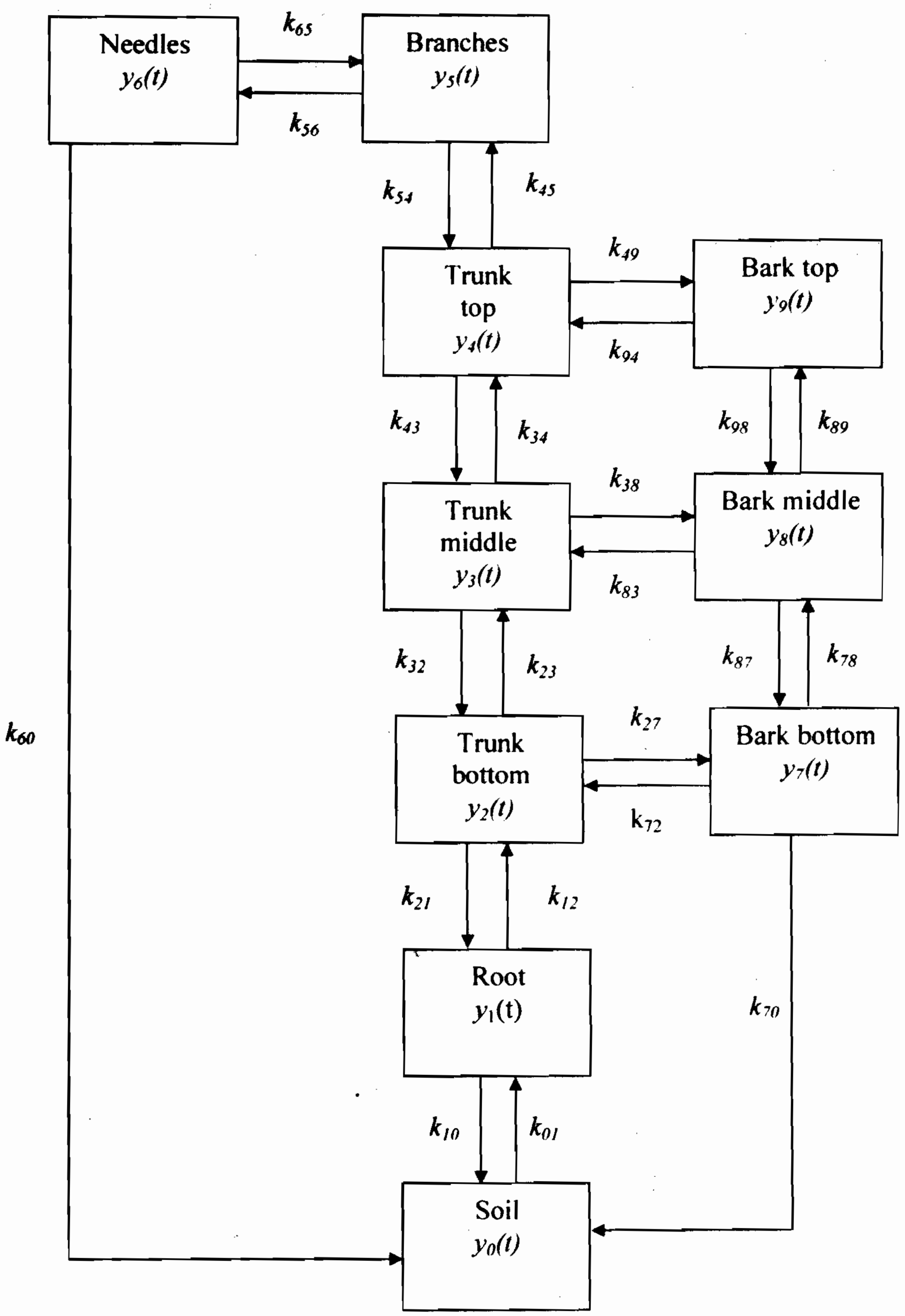

Fig.1: Compartmental model of the soil-pine system. The blocks represent the ^{137}Cs content of the compartments. The k_{ij} are transfer constants of ^{137}Cs between compartments and solid lines are transfer pathways.

The model structure is illustrated in Figure 1 in which 'boxes' represent the ^{137}Cs content of compartments and 'arrows' are transfer pathways. The model describes the radiocaesium transfer dynamics through a set of 10 coupled differential equations:

$$\begin{cases} \frac{dy_0}{dt} = -(k_{01} + \lambda)y_0 + k_{10}y_1 + k_{60}y_6 + k_{70}y_7 \\ \frac{dy_1}{dt} = k_{01}y_0 - (k_{10} + k_{12} + \lambda)y_1 + k_{21}y_2 \\ \frac{dy_2}{dt} = k_{12}y_1 - (k_{21} + k_{23} + k_{27} + \lambda)y_2 + k_{72}y_7 + k_{32}y_3 \\ \frac{dy_3}{dt} = k_{23}y_2 - (k_{32} + k_{34} + k_{38} + \lambda)y_3 + k_{43}y_4 + k_{83}y_8 \\ \frac{dy_4}{dt} = k_{34}y_3 - (k_{43} + k_{45} + k_{49} + \lambda)y_4 + k_{54}y_5 + k_{94}y_9 \\ \frac{dy_5}{dt} = k_{45}y_4 - (k_{54} + k_{56} + \lambda)y_5 + k_{65}y_6 \\ \frac{dy_6}{dt} = k_{56}y_5 - (k_{65} + k_{60} + \lambda)y_6 \\ \frac{dy_7}{dt} = k_{27}y_2 - (k_{70} + k_{72} + k_{78} + \lambda)y_7 + k_{87}y_8 \\ \frac{dy_8}{dt} = k_{38}y_3 + k_{78}y_7 - (k_{83} + k_{87} + k_{89} + \lambda)y_8 + k_{98}y_9 \\ \frac{dy_9}{dt} = k_{49}y_4 + k_{89}y_8 - (k_{94} + k_{98} + \lambda)y_9 \end{cases} \qquad (1)$$

Where:

k_{ij} (d^{-1}) is a transfer coefficient (TC) of radiocaesium activity from compartment i to compartment j;
y_i (Bq) is the activity of radiocaesium in compartment i;
t (d) is the time;
λ (d^{-1}) is the decay constant of ^{137}Cs.

The activity concentration c_i (Bq/kg) of radiocaesium in compartment i is calculated using the relation:

$$c_i = \frac{y_i}{M_i}, \qquad (2)$$

where M_i (kg) is the mass of compartment i. The TC $k'_{i,j}$ (d^{-1}) of the radionuclide concentration from compartment i to compartment j is derived from TC of radionuclide activity through the equation:

$$k'_{i,j} = \frac{M_i}{M_j} k_{i,j}. \qquad (3)$$

This ratio allows us to transform the system (1) that describes the variation of total activities within compartments to an equivalent system which accounts for the activity concentration (c_i) of radiocaesium in compartments.

Environmental sampling

55 year old pine trees were sampled at field sites within the Chernobyl 30 km exclusion zone, each site with different soil compositions (Table 1).

Table 1: Soil composition of the sites

	Ditiatki	Kopachi	Pripiat
Type of soil	soft podsolic and sandy	Sandy and soft humus	clay sandy and soft podsolic

The diameters of the trees were 20-40 cm and the heights were in the range 17-27 m. The understorey consisted largely of a carpet of moss species, with 95-98 % of the ground surface being covered by *Pleurozium schreberi* and *Dicranum polysetum*

One pine tree was cut down at each site and the trunk of the tree separated into three parts. Three 'slices' of trunk, each of 3-4 cm thickness, were taken at locations corresponding to 1 m, 10 m and 20 m heights and the external bark separated from the wood. Branches were lopped off and subsequently divided into thin (<5 mm) and thick (>5 mm) categories according to their diameters. Composite samples of needles and fresh shoots were collected. Samples of one-year and two-year needles were combined into single bulked samples. The fresh mass of each sampled component from the trees was determined in fresh state. The wet weight of samples is ranged from 0.700 kg to 0.070 kg. All samples were air-dried for 72 hours at 100°C and then homogenized. Samples of needles, shoots and thin branches were crushed in several stages to a fine powder. The wet weight of samples is ranged from 0.420 kg to 0.040 kg. ^{137}Cs in the homogenized samples was determined by direct gamma spectrometry. Given the location of the trees sampled, the ^{137}Cs contamination measured can be identified as coming exclusively from aerial deposition following the Chernobyl accident.
The forest litter at each sampling site was sampled layer by layer from an area of 50 x 50 cm. The leaf litter horizon (fresh litterfall), fermentative horizon (semi-decomposed litterfall) and humus horizon (decomposed litterfall) were taken separately.

Estimation of the model parameters

The next step in developing our model involved specifying the values of model parameters and comparing modelled results with experimental measurements. The data used in this work were obtained from the measurements described in the previous section.
The TC k_{ij} of the soil-plant system were evaluated in a semi-empirical way. In the case of transfer from soil to roots i=0 and j=1. For a given soil of the forest, k_{01} is the inverse of ecological half-life (T_{ecol}) of ^{137}Cs

$$k_{0,1} = \frac{1}{T_{ecol}}. \qquad (4)$$

Taking into account that the speed of the xylem sap in the tree ranges between 0.5 and 43.6 m/h (Mazliak, 1981), the values of TC of the ^{137}Cs in the pine tree are obtained by assuming that the length of a compartment is from 4 to 8 m; thus $k'_{i,j}$ are deduced from eq (3). The best values of $k'_{i,j}$ are obtained after successive iterations and matching of the theoretical and experimental values of radionuclide concentrations in various compartments of the pine tree. Table 2 presents the model TC of ^{137}Cs between compartments of the soil-plant system.

Table 2: Transfer constants of ^{137}Cs between soil-pine compartments

Rate constant from compartment i to j	Value (d^{-1})
Soil to root	8.64×10^{-5}
Root to soil	9.994×10^{-1}
Root to trunk bottom	9.48×10^{-1}
Trunk bottom to root	4.54×10^{-1}
Trunk bottom to trunk middle	8.55×10^{-1}
Trunk middle to trunk bottom	8.54×10^{-1}
Trunk middle to trunk top	9.99×10^{-1}
Trunk top to trunk middle	9.62×10^{-2}
Trunk top to branches	9.99×10^{-1}
Branches to trunk top	7.15×10^{-2}
Branches to needles	8.25×10^{-2}
Needles to branches	7.42×10^{-2}
Needles to soil	2.0×10^{-2}
Trunk bottom to bark bottom	9.59×10^{-1}
Bark bottom to trunk bottom	1.012×10^{-4}
Bark bottom to soil	0.00
Trunk middle to bark middle	5.21×10^{-2}
Bark middle to trunk middle	6.9×10^{-1}
Trunk top to bark top	5.98×10^{-1}
Bark top to trunk top	1.048×10^{-4}
Bark bottom to bark middle	2.048×10^{-1}
Bark middle to bark bottom	9.98×10^{-1}
Bark middle to bark top	9.98×10^{-1}
Bark top to bark middle	5.08×10^{-1}

To compare the model results with the experimental one, it is necessary to know the initial concentration of ^{137}Cs in the contaminant soil. Presently, these values are not available and consequently, the comparison is made using the concentration ratio CR_i defined as follows:

$$CR_i = \frac{c_i}{\sum_{k=2}^{9} T_{ecol}}. \qquad (5)$$

The concentration of roots compartment (c_1) is being excluded in this analysis because the experimental value is not available. However in the calculation c_1 is small and does not modify the result of the model. Additionally, this consideration has the advantage to involve dimensionless equations. The relative uncertainty on concentration ratio is calculated by comparison of predicted and observed values at the considered site as follows:

$$\Delta CR_i = \frac{|CR_i - CRP_i|}{CRO_i}. \qquad (6)$$

where CRO_i and CRP_i are respectively observed and predicted concentration ratio at the site.

RESULTS AND DISCUSSION

Table 3 presents the results of concentration measurements realised in five sites in the North of Ukraine.

Table 3: Observed concentration of ^{137}Cs in 1996 in pine compartments

Compartment	Concentration (Bq/kg)				
	Ditiatki	**Pripiat 1**	**Pripiat 2**	**Pripiat 3**	**Kopachi**
Trunk Bottom	218,2	1530,6	1141,0	3005,9	287,2
Trunk Middle	218,2	2603,0	1421,2	2480,0	239,2
Trunk Top	218,2	2652,3	2711,7	2034,2	426,5
Branches	3079,8	41641,4	22818,2	55093,2	1384,4
Needles	3219,5	29845,5	19326,0	28970,8	1283,8
Bark Bottom	3326,0	23791,0	26482,8	19719,6	15644,0
Bark Middle	937,7	2605,2	3093,2	4312,7	6020,6
Bark Top	1358,6	15618,8	-	8921,4	2355,6

The concentration was not measured in the trunk top at Pripiat 2. The concentrations at Ditiaki and Kopachi are the same order of magnitude in the trunk bottom and middle. The concentrations at Pripiat 1, 2 & 3 are more than 5 times higher than those at Ditiatki and Kopachi in the trunk compartments. Moreover, the concentrations at Ditiatki and Kopachi are generally smaller. The pine compartments, except the bark top show a positive correlation of concentration variations at Pripiat 1 & 2

Numerical results were obtained by solving differential equations of ^{137}Cs concentrations. To perform the model calculations, a computer program using Runge-Kutta method of fourth-order has been written in MATHCAD environment. These results are normalised as given in formula (5). The annual predicted and observed concentration ratios of ^{137}Cs in soil in 1996 are presented in the form of histogram in Fig. 2 and 3. These figures show the model results as compared to the results of field experiments.

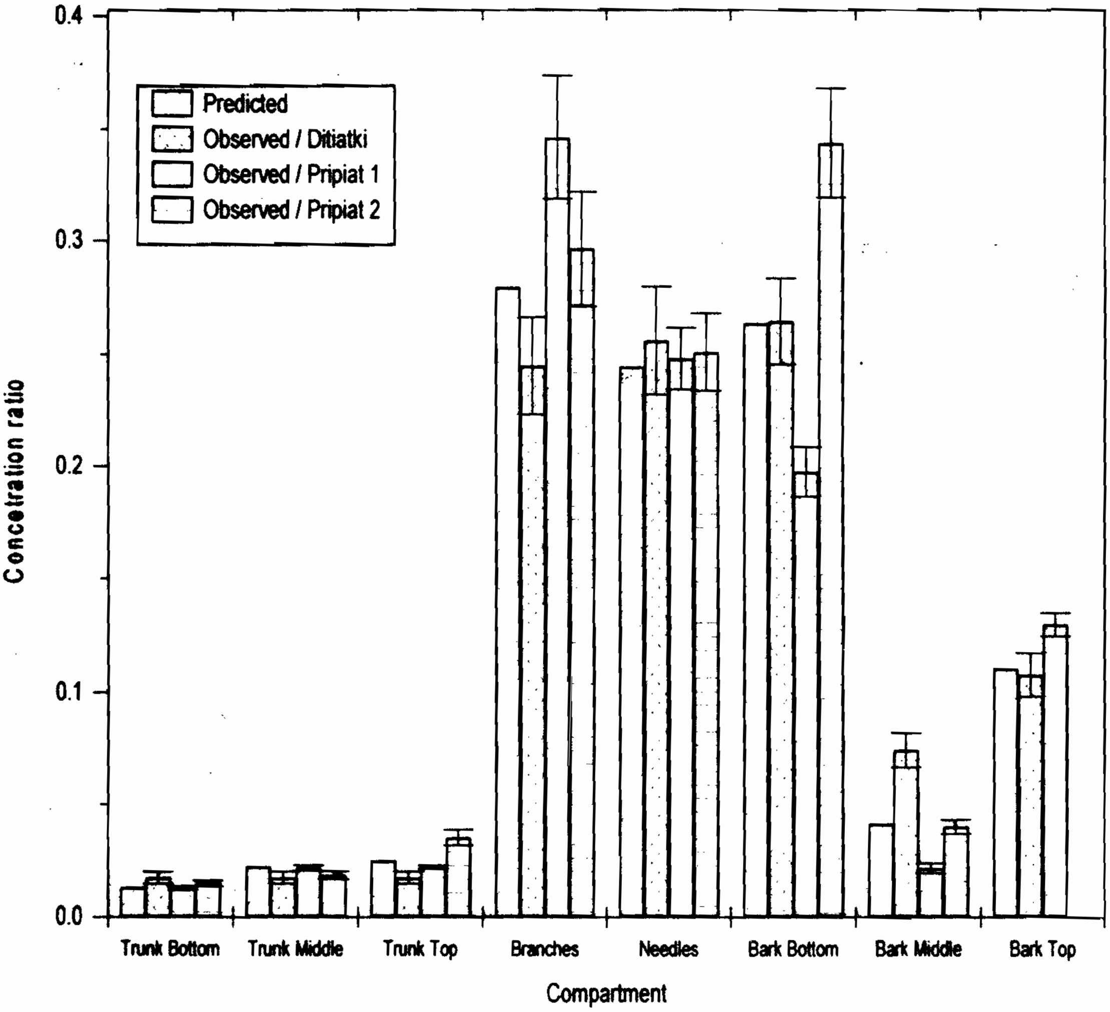

Fig. 2. Observed and predicted concentration ratios of ^{137}Cs in 1996 at Ditiatki, Pripiat 1 and 2.

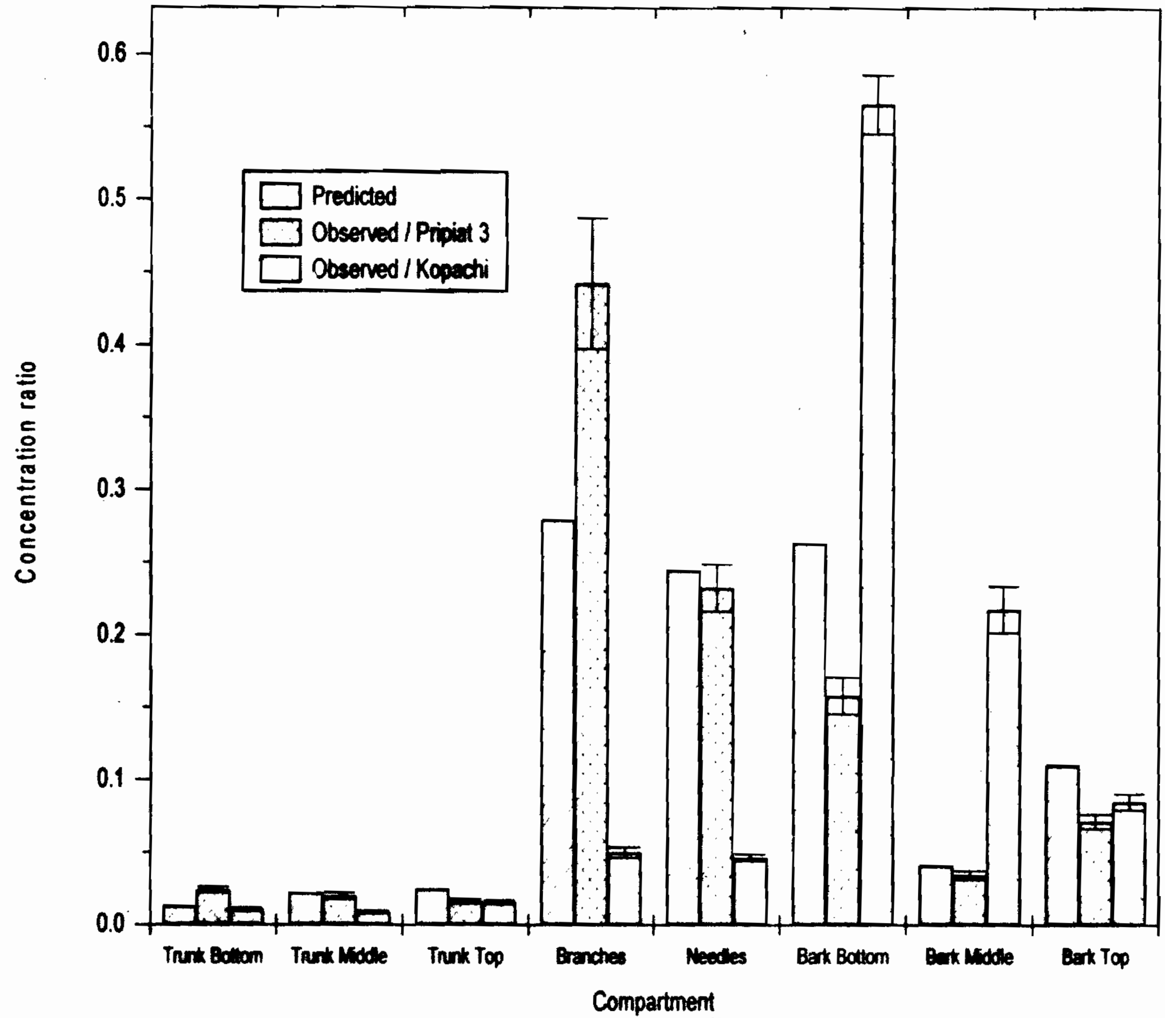

Fig. 3: Observed and predicted concentration ratios of ^{137}Cs in 1996 at Kopachi, Pripiat

The histogram of figure 2 represents model and experimental CR in eight compartments of the pine tree in the sites of Ditiatki, Pripiat 1 and 2. The variations between experimental and model CR correlate except for Pripiat 1 in the bark bottom. Moreover at Ditiatki, experimental CR is 1.78 times that of the model in the bark middle. The difference between experimental and model CR (DEM) does not exceed 32% in the other compartments.

At Pripiat 1, the greatest DEM is observed in the bark middle where the experimental value is 0.52 times that of the model. In the other compartments, the DEM are less than 33.25%. At Pripiat 2, the DEM is less than 30% in all the compartments.

The histograms of figure 3 represent experimental and model CR at Kopachi and Pripat 3. A great difference is observed between model and experimental CR in all the compartments except in bark top and trunk compartments:

- At Kopachi, experimental CR is 2.15 times that of the model in the bark bottom, 5.25 times in the bark middle, 0.39 times in the trunk middle, 0.19 times in the needles and 0.18 times in the branches.
- At Pripiat 3, except for the needles, trunk middle, and bark middle compartments, the DEM is higher than 36.86%.

The increase in experimental measurements corresponds to an increase in the results calculated from the model. This correlation is an indication that the model can qualitatively describe the evolution of the ^{137}Cs distribution in the compartments of the pine tree. It is possible therefore, to determine the most contaminated parts of the pine tree.

Moreover, quantitatively, experimental measurements are different in the trunk compartments in all the sites, except at Ditiatki, and the model data present a similar pattern. The same effects have been observed in the bark compartments in all the sites. The normalised experimental and model data are similar. This could justify the division of the trunk and the bark of the pine tree. Therefore the model could be used to describe the ^{137}Cs transfer.

The parameterisation of the model was made using the experimental results of the site of Pripiat 2. The model results are close to the experimental one at Ditiatki, Pripiat 1 and 2 (Fig. 2). However, there exists a big gap between the model and the experimental results at Kopachi and Pripiat 3 (Fig. 3). The model is therefore, not efficient to predict quantitatively the distribution of the ^{137}Cs in the compartments of the pine trees at Kopachi and Pripiat 3. One may consider that this model is applicable in three of the five sites.

When the parameterisation of the model is made from the measurements obtained at Kopachi, it is noted that the ecological half-life of the ^{137}Cs increases. One can suppose that the ^{137}Cs is mainly in a nonabsorbable form directly from the pine. The model results, in this case, are not close to those of any other sites. When the parameterisation is made from the results obtained at Pripiat 3, it is noted that the ecological half-life of the ^{137}Cs decreases. One can suppose that the

^{137}Cs is mainly available for absorption by the pine trees. Once more, the model results in this case are not close to those of any other site.

The concentration in the needles, the branches and the bark is higher than in the other compartments. This suggests that the radiocaesium is accumulated in these compartments and may constitute the principal internal source of new contamination of the pine trees. For this reason, one can consider the needles, branches and bark compartment as reservoirs of ^{137}Cs in the pine trees.

Taking into account the soil composition (Table 2) and the fact that the concentrations at Pripiat 1, 2 and 3 are overall higher than those of the other sites, one can suggest that the clay content in the soil could increase the ^{137}Cs absorption by the pine by lowering it diffusion. In fact when the soil is sandy, the order of magnitude of the ^{137}Cs concentrations is lower (Table 3). This would mean that the uptake of the ^{137}Cs is very weak due to the high diffusion.

All these sites are located in a zone very strongly contaminated; however, the different distances of each of them from the power station, suggest that the type of ^{137}Cs particles deposited would influence the absorption of the ^{137}Cs by the pine trees and consequently its distribution inside the plant. This is in harmony with the fact that the type of deposit depends on the distance from the power station.

Validation of the model

Bulgakov and Konoplev (2002) considered the trunk of the pine trees only, as one compartment while the experimental results obtained in five sites of Ukraine by using 55 years old pine trees showed that the trunk and the bark should be composed of three compartments each.

The results obtained in the model after parameterisation using data of Pripiat 2 show that the model is efficient for three sites out of five. The division of the trunk in three compartments could be justified by the downward and upward movements of the saps (Heller, 1981). According to Fesenko et al. (2001), this effect can be explained by the fact that the trunk plays the role of a filter for the radionuclides taken by the roots. When the radionuclides are transferred by the sap from the root system to the needles, a part is stored in the plant. This leads to a reduction of the ^{137}Cs activity in the sap. This effect is more pronounced in the trunk top. For Ditiatki, Pripiat 1 & 2, the forecast of the model corresponds to the experimental results. The difference in the observed CR at Kopachi and Pripiat 3 could be due to the kind of soil and the type of deposit. In fact, according to Fesenko et al. (1997), the physicochemical composition of the deposit influences its transfer towards the plant.

When the soil is sandy, the absorption of the ^{137}Cs by the pine is very weak and is independent of the type of deposit, as a consequence of the diffusion. The clay content soil leads to an increase in the absorption. The percentage of fuel particles in the deposit is related to the bioavailability of the ^{137}Cs in the soil (Fesenko et al., 1997). Therefore, the bioavailability of the deposit and the type of soil should be involved in the determination of the model parameters.

CONCLUSION

This work allows us to develop a compartmental model of pine tree with a division of trunk and bark in three compartments each. The iterative method, using the successive comparison of the experimental and numerical values obtained from the model has been used to determine the best values of the TC between compartments. The model enabled us to know the most contaminated parts of the pine trees. In most of the sites, the behaviour of the experimental and theoretical results is similar. The needles, the branches and the bark compartments play the role of reservoir of ^{137}Cs in the pine trees.

The process of the soil-pine transfer of the ^{137}Cs is very complex. A future research could focus on the effect of the diffusion and bioavailability of the radionuclides in the soil.

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