Paper

LONG TERM RADIOCESIUM CONTAMINATION OF FRUIT TREES FOLLOWING THE CHERNOBYL ACCIDENT

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Abstract—**Radiocesium contamination from the Chernobyl accident of fruits and leaves from various fruit trees was systematically studied from 1990 to 1995 on two agricultural experimentation farms in Northern Greece. The results are discussed in the framework of a previously published model describing the long-term radiocesium contamination mechanism of deciduous fruit trees after a nuclear accident. The results of the present work qualitatively verify the model predictions.**

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Key words: ^{137}Cs; Chernobyl; contamination; accidents, nuclear

INTRODUCTION

Prediction of long term radiocesium contamination of fruit trees following a major nuclear accident is of obvious importance. Transfer of radiocesium between parts of trees and to the environment was initially studied experimentally by artificial introduction of ^{137}Cs in the body of trees (Olson 1965; Waller and Olson 1967; Hoffman 1972). Following the Chernobyl accident the same studies (Bergman et al. 1991; Bunzl et al. 1989; Desmet et al. 1990) have been performed under real conditions.

Systematic studies of radiocesium contamination of fruit trees in the submediterannean environment of Northern Greece were carried out starting in 1987, i.e., one year after the Chernobyl accident (Antonopoulos-Domis et al. 1990, 1991). Based on early experimental observations a compartment model for long term radiocesium contamination of fruit trees was proposed (Antonopoulos-Domis et al. 1990). In the present work we report on the experimentally observed evolution in time of such contamination and compare it with the model prediction. The measurements support the validity of the model and provide estimates of model parameters. A brief qualitative description of the model is presented in the following chapter; detailed description can bc found in the corresponding publication.

THE MODEL

The model postulates two mechanisms of contamination of new tree products (fruits, leaves, etc.):

1 Translocation from the tree reservoir of cesium tha was formed by absorption of a fraction of the cesiun deposited on above-ground parts of the tree durin May 1986. A fraction of this cesium in the body of thc tree is available for translocation to new tree product while most of the cesium in the tree reservoir is bounc i.e., not available for translocation. The model postulates a net flow from the compartment of availabl cesium to that of unavailable.
2 The other contamination mechanism is root uptak from the soil. Cesium in the soil is described by tw compartments: one free, i.e., available for root uptake and the other bound in soil minerals and not availabl for root uptake, with a net transfer from the free to th bound compartment.

The main important prediction of the model was th radiocesium concentration C_m (Bq kg^{-1}) of fruits o leaves at year $m \geq 1$ after the accident is described by th sum of two exponentials:

$$C_m = A \exp(-\gamma_b \cdot m) + B \exp(-\gamma_s \cdot m), \quad m \geq 1, \quad (1$$

where, A, B, $\gamma_b > 0$ and $\gamma_s > 0$ are constants. Definitio of these constant may be found in Antonopoulos-Domi et al. (1990). The first term of eqn (1) is due t translocation of cesium from the tree reservoir, and th second term is due to root uptake. Experimental evidenc suggested (Antonopoulos-Domis et al. 1990) that roc uptake was initially negligible with respect to translocation from the tree reservoir, i.e., $A \gg B$. The first fou years following the accident the measured time dependence of the contamination of fruits and leaves wa described by a single exponential function and constar "γ_b" was deduced (Antonopoulos-Domis et al. 1991) fc different fruit species.

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EXPERIMENTAL PROCEDURE

Radiocesium contamination of fruits for the years 1987–1990 for a wide variety of fruit trees in Northern Greece have been presented in a previous publication (Antonopoulos-Domis et al. 1991). In the following years it was impossible to measure the radiocesium contamination of fruits (except for sweet cherries for the years 1991–1995) due to radiocesium concentrations smaller than the detection limit (0.1 Bq kg^{-1}) of our experimental set up. We overcame this difficulty by measuring the radiocesium contamination of leaves since it was found (Antonopoulos-Domis et al. 1991) that they have the same time-dependence of radiocesium concentration as the fruits; the same is confirmed for the years 1991 to 1995 by comparison of Figs. 1 and 2.

The experimentation farms used in the present study are located in Northern Greece and were selected on the basis of their different but relatively high levels of radiocesium deposition. The deposition of ^{137}Cs due to the Chernobyl accident on the selected farm lands was indirectly determined (Antonopoulos-Domis et al. 1988) from properly performed soil measurements and was found to be 15 kBq m^{-2} at the Thessaloniki farm and 63 kBq m^{-2} at the Naousa farm. The value of 63 kBq m^{-2} is one of the highest reported (Simopoulos 1989) values of radiocesium deposition in Greece. Three trees of each of three species (sweet cherries, apples, peaches) that received the radiocesium deposition in 1986 were selected in both locations for the present work. In addition, leaves were collected from three trees (of each of three species) planted after the accident; these trees have, of course, not received the direct deposition. Two samples (leaves) were collected at the beginning of November of every year from each fruit tree for the years 1991–1994. In the case of sweet cherries, where it was possible to detect radiocesium contamination of fruits, samples were collected during the years 1991–1995 from the trees which have been also used in the previous samplings (1987–1990). All samples were carefully washed before radioactivity measurements to exclude any external ^{137}Cs contamination of the samples due to resuspension of ^{137}Cs and interception. In a few of the samples, we compared radiocesium concentration before and after washing and found practically no difference. The samples (leaves), after the cleaning procedure, were oven dried, ground, and used for radiocesium counting. The amount of ^{137}Cs in the collected samples was determined by standard γ-spectroscopy using a high-purity Ge detector connected to a multichannel analyzer and microcomputer. The measurement time of each sample, placed inside shielding appropriate for low-level counting, was about 24 h.

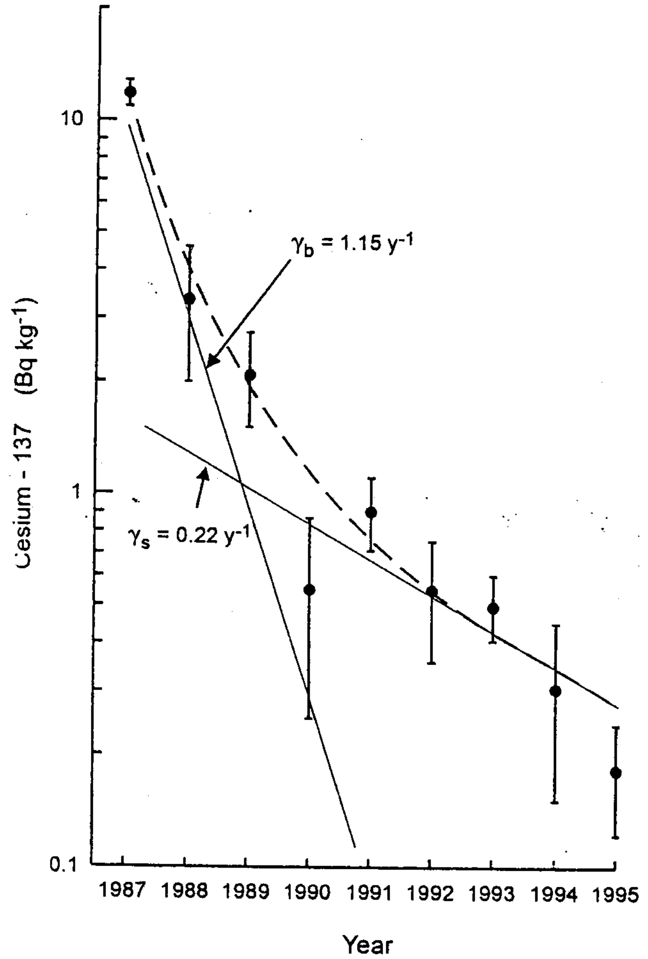

Fig. 1. Radiocesium concentration (Bq kg^{-1} fresh weight) of sweet cherries collected during the years 1987–1995 from the Naousa farm. In dashed line is the sum of two exponentials presented in this semilogarithmic scale by the two continuous lines.

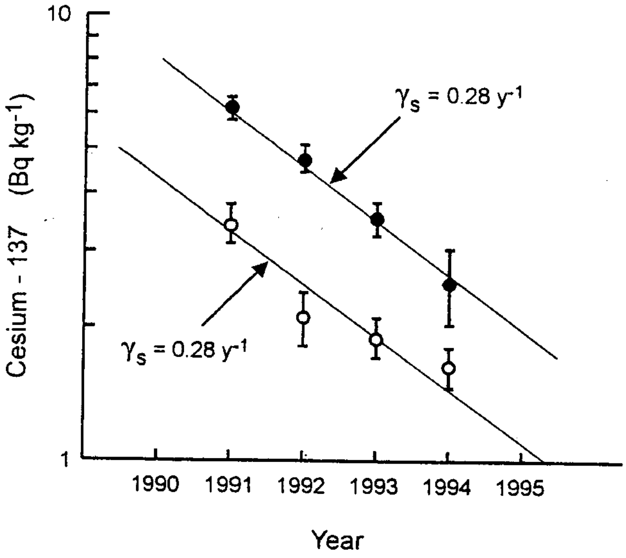

Fig. 2. Radiocesium concentration (Bq kg^{-1} dry weight) of leaves of sweet cherries trees collected during the years 1991–1994 from the Naousa farm. Close and open circles corresponds to leaves collected from trees which have been planted before and after the Chernobyl accident respectively.

RESULTS AND DISCUSSION

Radiocesium concentration (Bq kg^{-1} fresh weight) of sweet cherries collected during the years 1987–1995 from the Naousa farm is presented in Fig. 1. The decay constants γ_b and γ_s presented in this figure were deduced from the experimental results as follows: an exponential term was fitted to the data from 1990 to 1995 by non-linear least squares fit; this gave the value $\gamma_s = 0.22 \pm 0.06$ y^{-1}. This exponential function is presented in Fig. 1 as the second straight line. The values of this function were then subtracted from the experimental data of the years 1987 to 1989; these data were again fitted by non-linear least squares fit by an exponential function; this function is presented in Fig. 1 as the first straight line and gave $\gamma_b = 1.15 \pm 0.1$ y^{-1}. The dashed line in Fig. 1 is the sum of these two exponential functions; it can be seen that this dashed line fits well the experimental data.

The decay constant $\gamma_b = 1.15$ y^{-1} is in very good agreement with the values $\gamma_b = 1.05$ y^{-1} deduced for sweet cherries in Northern Greece from a 4-y study 1987–1990 (Antonopoulos-Domis et al. 1991) and $\gamma_b = 1$ y^{-1} predicted by the compartment model (Antonopoulos-Domis et al. 1990). The second slope in Fig. 1 was predicted by the compartment model which also indicated a decay constant $\gamma_s = 0.7$ y^{-1}, i.e., of the same order of magnitude of the decay constant $\gamma_s = 0.22$ y^{-1} experimentally observed. A second slope was also very recently observed for tea plants (Unlu et al. 1995).

In the framework of the model (Antonopoulos-Domis et al. 1990) the first exponential of Fig. 1 (decay constant $\gamma_b = 1.15$ y^{-1}) describes the contamination of fruits and leaves due to translocation (transfer of radiocesium to leaves and fruits from the radiocesium content in the body of the tree), and the second exponential (decay constant $\gamma_s = 0.22$ y^{-1}) describes the contamination due to root uptake. At this point it is important to note that the physical decay constant of ^{137}Cs ($\lambda = 0.023$ y^{-1}) is one order of magnitude smaller that the "γ_s" decay constant and 50 times smaller than the decay constant "γ_b" and thus could be neglected, but there is no need for that. In the model the physical decay constant λ is properly accounted for in the decay constants γ_b and γ_s (Antonopoulos-Domis et al. 1990), and the measured radiocesium concentrations were corrected for physical decay.

It can be seen in Fig. 1 that for the years 1991–1995 the contribution of the first exponential (with decay constant γ_b) to the radiocesium concentration of fruits is negligible and only the second exponential (with decay constant γ_s) is important. As leaves and fruits have the same time-dependence of radiocesium concentration (M. Antonopoulos-Domis et al. 1991), it is logical to expect that after 1991 the radiocesium concentration of leaves would show a single exponential decrease with time. This is shown in Figs. 2–5. Radiocesium concentration (Bq kg^{-1} dry weight) of leaves of fruit trees of different species collected during the years 1991–1994 from the two selected experimental farms are presented in Figs. 2–5. In all figures closed circles correspond to leaves collected from trees which have been planted before the Chernobyl accident (BC), thus receiving the direct deposition, and open circles correspond to leaves collected from trees that have been planted after the Chernobyl accident (AC) and thus did not received the direct deposition in 1986. It is obvious that radiocesium contamination of leaves from the AC trees is only due to root uptake. In all figures we generally observe an exponential decrease (continuous straight line) of radiocesium concentration of leaves for all fruit trees species and in both locations. The decay constant of each exponential decrease is also presented in each figure.

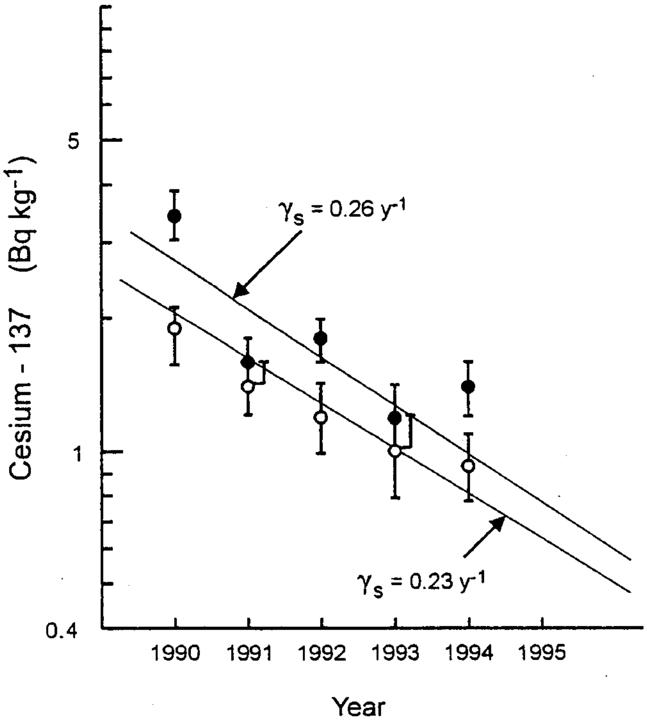

Fig. 3. Radiocesium concentration (Bq kg^{-1} dry weight) of leaves of peach trees collected during the years 1990–1994 from the Naousa farm. Close and open circles corresponds to leaves collected from trees which have been planted before and after the Chernobyl accident respectively.

For all fruit trees species and in both experimental farms it can be seen in Figs. 2–5 that the decay constant is about the same for leaves collected from trees planted before and after the Chernobyl accident. This implies that the radiocesium contamination mechanism of the leaves of the AC and BC trees is the same. Given that the only radiocesium contamination of leaves of AC trees is root uptake, it is concluded that for BC trees the major contamination mechanism after 1991 is root uptake. This radiocesium contamination is, however, extremely low: two orders of magnitude smaller than natural radioactivity due to ^{40}K.

It was found (Antonopoulos-Domis et al. 1991) that contamination of fruits has the same time dependence as the contamination of leaves when root uptake was negligible. It can be seen that the slope of the exponential

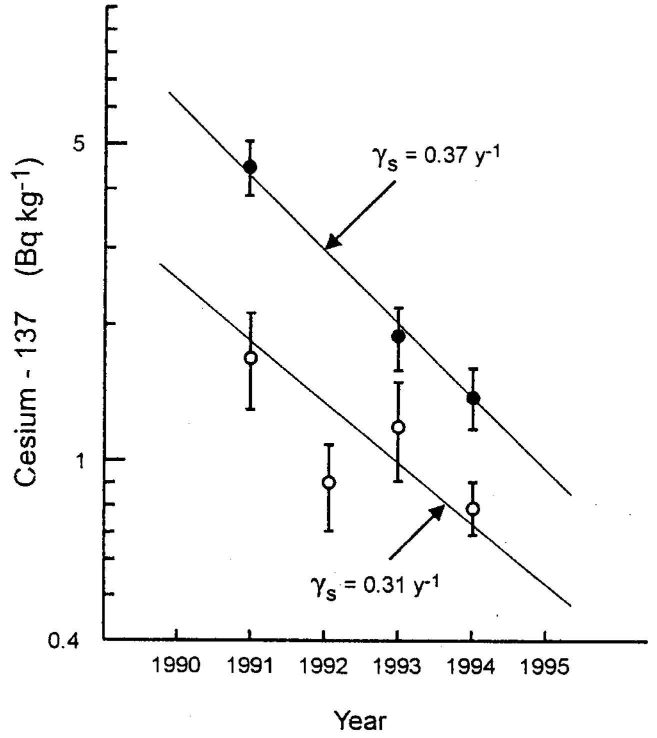

Fig. 4. Radiocesium concentration (Bq kg^{-1} dry weight) of leaves of sweet cherries trees collected during the years 1991–1994 from the Thessaloniki farm. Close and open circles corresponds to leaves collected from trees which have been planted before and after the Chernobyl accident respectively.

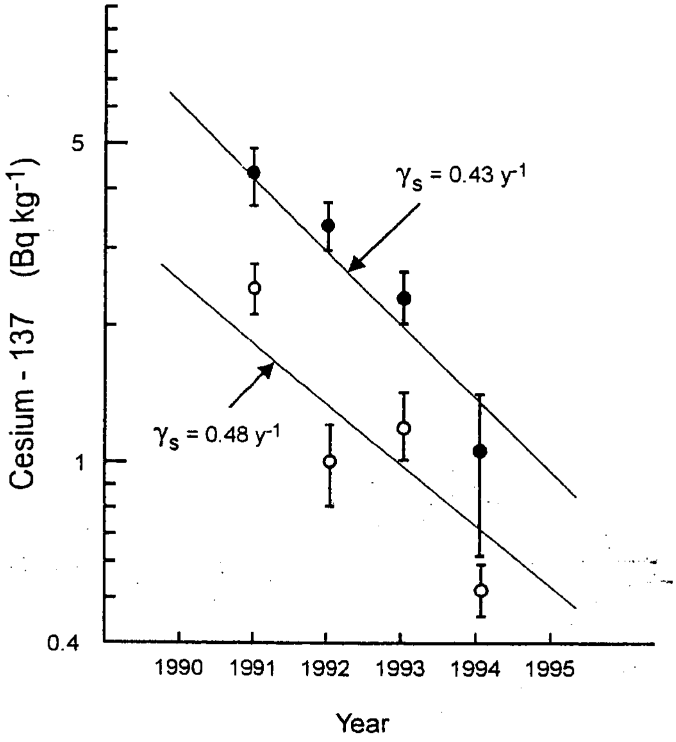

Fig. 5. Radiocesium concentration (Bq kg^{-1} dry weight) of leaves of apple trees collected during the years 1991–1994 from the Thessaloniki farm. Close and open circles corresponds to leaves collected from trees which have been planted before and after the Chernobyl accident respectively.

decrease of the contamination of sweet cherries after 1991 (Fig. 1) is practically the same as that for the leaves of sweet cherries for the same period (Fig. 2). It is therefore concluded that fruits and leaves present the same time dependence of contamination even when root uptake is the dominant contamination mechanism.

CONCLUSIONS

Contamination of fruits and leaves with ^{137}Cs from the Chernobyl accident was systematically studied at two experimental farms in Northern Greece. The main experimental results of the present work are summarized in Table 1 and compared to predictions of a previously presented model (Antonopoulos-Domis et al. 1990). From Table 1 it is clear that the results of the present work verify at least qualitatively the model predictions.

Contamination of fruits was found to have same time dependence as that of leaves, even when root uptake is the dominant contamination mechanism.

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Table 1. Main experimental results of the present work compared to model predictions.

Model predictions	Experimental results
Radiocesium concentration of fruits or leaves of a fruit tree in year *m* after a nuclear accident is given by the sum of two exponentials $C_m = A \exp(-\gamma_b m) + B \exp(-\gamma_s m)$	Verified
$\gamma_b = 1$ y^{-1}	$\gamma_b = 1.15$ y^{-1}
The second exponential (decay constant γ_s) is due to root uptake	Verified
$\gamma_s = 0.7$ y^{-1}	0.22 y^{-1} < γ_s < 0.48 y^{-1}

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