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Derivation of Soil to Plant Transfer Factors of Radiocesium in Northern Greece after the Chernobyl Accident, and Comparison with Greenhouse Experiments

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ABSTRACT

Radiocesium contamination of cereals, due to the Chernobyl accident, was systematically studied in two selected experimental agricultural farms in Northern Greece for the years 1987, 1988 and 1989. Radiocesium contamination of all annual crops is very low, of the order of 1 Bq kg^{-1}, and appears to be, for the first 3 years after the Chernobyl accident, time independent, the differences lying within the experimental error. Transfer factors, relating radiocesium deposition to contamination of crops, are deduced from the experimental results. Results are also discussed in the framework of UNSCEAR's empirical model, and the corresponding parameters are deduced. In addition, greenhouse experiments show that the obtained Transfer Factors are independent of the initial radiocesium deposition and that radiocesium from the Chernobyl fallout behaves differently from usual experimental sources, such as $^{137}CsCl$.

INTRODUCTION

Radiocesium activity of soils and vegetation in Greece due to the Chernobyl fallout has been observed and reported by several laboratories (Antonopoulos-Domis *et al.*, 1988*a,b*; Papanicolaou & Kritidis 1988; Papastefanou *et al.*, 1988; Sawidis, 1988; Simopoulos, 1989). Three years after the passage of the Chernobyl cloud over Greece, traces of radiocesium contamination can still be detected in almost all annual crops. Despite the

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very low level of crop contamination, Cesium activity may be used for testing existing models of radiocontamination by root uptake of contaminants from the soil.

Radiocesium contamination of annual crops over extended periods following a nuclear accident, may be predicted by:

(a) the well known Transfer Factors (*TF*) relating soil concentration to plant concentration of contaminants which are usually defined as:

$$TF = \frac{\text{Bq kg}^{-1}\text{ (crop)}}{\text{Bq kg}^{-1}\text{ (dry soil)}} \quad (1)$$

and have been obtained mainly from greenhouse experiments (e.g. Saas, 1982). Considerable caution should be exercised in using these results as there are significant differences between the actual behaviour of contaminants in fields and in greenhouse experiments. In addition, Transfer Factors depend on the physiochemical form of the Cesium isotope and it is now known that 'Chernobyl' radiocesium is different from that of 'pre-Chernobyl' radiocesium (Kirton *et al.*, in press) resulting from nuclear-weapon tests and from that of usual radiocesium sources, such as ^{137}CsCl Maubert, *et al.*, in press).

(b) UNSCEAR's empirical model (UN, 1972, 1977; Lassey, 1979) in which concentration Cm of ^{137}Cs, measured in Cesium Units (CU = pCi (Cs) g^{-1} (K)) in a specific foodstuff at a specific locality during the *m*th year after a single deposition F_0 (mCi km^{-2}), can be expressed in terms of just two parameters as:

$$\text{Cm} = pF_0 \exp(-\mu m) \qquad m \geq 1 \quad (2)$$

Parameter p depends on the rate of root uptake of ^{137}Cs and μ is a measure of the rate with which Cesium in the soil is being made unavailable for root uptake, mainly due to fixation in soil minerals and to radioactive decay.

The major source of uncertainty in estimating contamination levels resulting from root uptake is the well known (Russel, 1966) strong dependence of soil to plant transfer parameters (*TF* and p) on soil type and consequently on geographic location. In the present work, these parameters are deduced from the follow-up of Chernobyl radiocesium at two specific areas in Northern Greece for the years 1987, 1988 and 1989. Greenhouse experiments were also carried out in order to investigate the dependence of crop contamination on the amount of fallout.

EXPERIMENTAL METHOD

After the Chernobyl accident (1986), we selected two agricultural experimental farms in Northern Greece (one belonging to the University of

Thessaloniki and the other to the Ministry of Agriculture), as natural radioecological laboratories. Agricultural products (mostly cereals) were collected for the years 1987, 1988 and 1989 from these locations, selected on the basis of their different but high levels of radiocesium deposition. The plants were harvested at maturity, air-dried in the greenhouse and oven-dried at 70°C. Considerable caution was exercised in the preparation of samples in order to remove any external ^{137}Cs contamination of samples due to resuspension of ^{137}Cs and interception. Samples from the tilled soil (0–20 cm depth), were also collected from each selected area. These were dried, passed through a 2-mm sieve and used for ^{137}Cs and ^{40}K counting, as well as for physical and chemical analyses. The deposition of ^{137}Cs on the selected areas, following the Chernobyl accident was indirectly determined from properly performed soil measurements which had also shown that more than 90% of radiocesium deposition was in the first 10 cm of the undisturbed soil (Antonopoulos-Domis *et al.*, 1988*a*). Furthermore, we found an exponential decrease with depth of the radiocesium contamination in undisturbed soils and a uniform distribution in tilled soils.

For the greenhouse experiment, soil (0–20 cm depth) was collected from the University of Thessaloniki experimental farm and air-dried. Five portions, weighing 11 kg each, were contaminated with ^{137}Cs by evenly spraying and continually stirring the soil mass with appropriate solutions of $^{137}CsCl$ in order to obtain five levels of additional contamination, i.e. 0 kBq, 11 kBq, 20 kBq, 38 kBq and 58 kBq. Samples of the soil were then taken and measured to ensure the even distribution of ^{137}Cs. Five plastic 10-liter pots were then filled with the soil at a depth of 40 cm and grass (*Lolium perenne* L.) was grown for one year. One year after the initial contamination of the soils with $^{137}CsCl$, wheat was planted and grown outdoors under field conditions. Plants were harvested in June 1989 and separated into straw and grain.

Cylindrical plastic jars (73 mm diameter, 105 mm height) were filled with the appropriately prepared samples for radioactivity counting. The amount of ^{137}Cs and ^{40}K in the collected samples was then determined by standard γ-spectroscopy, using a high purity Ge detector connected with a multichannel analyser and a microcomputer. The time of measurement of each sample, placed inside a shielding appropriate for low-level counting measurements, was about 24 h and four samples for each agricultural product were measured.

RESULTS AND DISCUSSION

The basic characteristics of soils from the selected areas are presented in Table 1. They indicate that these soils, as almost all soils of Northern Greece

TABLE 1
Basic Characteristics of the Soils from Selected Locations

	Thessaloniki	*Ptolemais*
pH	8	6·9
Clay %	22	24
Silt %	37	42
Sand %	41	34
Organic matter %	1·28	3·78
Exchangeable potassium	0·48 me 100 g^{-1}	2·37 me 100 g^{-1}
Total potassium	800 mg 100 g^{-1}	1 650 mg 100 g^{-1}
CEC	22·28 me 100 g^{-1}	26·08 me 100 g^{-1}
$CaCo_3$ %	1·8	1·9

TABLE 2
Radiocesium Concentration (Bq kg^{-1}) in Annual Crops (Years 1987, 1988 and 1989) from the University of Thessaloniki Experimental Farm with a ^{137}Cs Deposition of 15 kBq m^{-2}. The Corresponding Soil to Plant Parameters *TF* and *p* (see text) are also Presented

Crops	*CS-137 (Bq kg^{-1})*	*K-40 (Bq kg^{-1})*	*TF*	*p CU per mCi km^{-2}*
		(year 1987)		
Wheat (grain)	<0·5	160 ± 10	<0·008	<0·005
Wheat (straw)	3·2 ± 1	339 ± 40	0·05	0·015
Barley (grain)	<0·5	155 ± 32	<0·008	<0·005
Maize (grain)	<0·5	86 ± 18	<0·008	<0·01
Rice	2·8 ± 0·3	95 ± 13	0·043	0·049
		(year 1988)		
Wheat (grain)	0·5 ± 0·1	170 ± 15	0·009	0·006
Wheat (straw)	5·7 ± 1	570 ± 80	0·1	0·02
Barley (grain)	<0·5	160 ± 30	<0·009	<0·006
Barley (straw)	3·2 ± 0·8	457 ± 50	0·06	0·014
		(year 1989)		
Wheat (grain)	0·5 ± 0·1	165 ± 15	0·009	0·006
Wheat (straw)	2 ± 0·8	400 ± 50	0·037	0·01
Barley (grain)	<0·5	160 ± 30	<0·009	<0·006
Barley (straw)	5·1 ± 1	463 ± 30	0·09	0·022
Maize (grain)	<0·5	105 ± 20	<0·009	<0·009

TABLE 3

Radiocesium Concentration (Bq kg^{-1}) in Annual Crops (Years 1987, 1988 and 1989) from the Ptolemais Agricultural Farm with a ^{137}Cs Deposition of 34 kBq m^{-2}. The Corresponding Soil to Plant Parameters *TF* and *p* (see text) are also Presented

Crops	*Cs-137 (Bq kg^{-1})*	*K-40 (Bq kg^{-1})*	*TF*	*p* *CU per mCi km^{-2}*
		(year 1987)		
Wheat (grain)	0·7 ± 0·3	165 ± 4	0·006	0·004
Wheat (straw)	3 ± 0·9	792 ± 91	0·025	0·004
Barley (grain)	<0·5	157 ± 7	<0·004	<0·003
Barley (straw)	5·3 ± 0·8	939 ± 40	0·05	0·005
Maize (grain)	1·1 ± 0·2	107 ± 11	0·009	0·009
Peas (seeds)	3·6 ± 0·4	107 ± 5	0·03	0·029
Peas (stover)	10·5 ± 1	252 ± 27	0·09	0·038
Chickpeas (seeds)	<0·5	171 ± 9	<0·004	<0·003
Chickpeas (stover)	12 ± 2	740 ± 42	0·1	0·015
		(year 1988)		
Wheat (grain)	1·1 ± 0·4	161 ± 10	0·009	0·006
Wheat (straw)	3·1 ± 1	683 ± 80	0·025	0·004
Barley (grain)	0·6 ± 0·2	176 ± 10	0·005	0·003
Barley (straw)	3·2 ± 1	940 ± 50	0·026	0·003
Maize (grain)	0·6 ± 0·2	105 ± 5	0·005	0·005
Peas (seeds)	2·3 ± 0·5	101 ± 10	0·02	0·02
		(year 1989)		
Wheat (grain)	0·8 ± 0·3	141 ± 15	0·006	0·005
Wheat (straw)	3·6 ± 0·9	635 ± 20	0·03	0·005
Barley (grain)	0·5 ± 0·1	117 ± 20	0·004	0·004

(Papanicolaou & Kritidis, 1988), are supplied with a sufficient amount of exchangeable potassium and other cations, and they are mostly medium or heavier in texture.

The activities of ^{137}Cs and ^{40}K, of various agricultural products collected from the two selected farms, for the years 1987, 1988 and 1989 are presented in Tables 2 and 3. It can be seen that radiocesium contamination of grain is very low, almost always less than 1 Bq kg^{-1}. It should be noted that radiocesium contamination of the vegetative parts of the plants is more than 1 Bq kg^{-1}, but almost always less than 10 Bq kg^{-1}. For all crops, the natural radioactivity due to ^{40}K is about two orders of magnitude greater than that due to radiocesium contamination. We may recall that in the same location (Ptolemais) in 1986 we had a radiocesium contamination of wheat grain of about 300 Bq kg^{-1} due to direct deposition. The activity of ^{134}Cs in agricultural products was not detectable. However, the ^{134}Cs in the soil was measured and the ratio $^{134}Cs/^{137}Cs$ referred to the date of the Chernobyl accident was found to be 0·5, which is an indication that pre-Chernobyl radiocesium concentration was too low to be considered.

For the first 3 years after the Chernobyl accident we generally observed that the radiocesium contamination of almost all annual crops appeared to be time independent, the differences lying within the experimental error. This could be easily understood if the non-availability of radiocontaminants in the soil for root uptake was due only to radioactive decay, which in the case of ^{137}Cs ($\lambda = 0{\cdot}023$ year^{-1}) could explain the obtained results. However, it is well known (Russel, 1966) that radiocesium in the soil is made unavailable for root uptake, not only due to radioactive decay, but also due to fixation in soil minerals.

Transfer Factors, relating radiocesium deposition to contamination of crops, are also shown in Tables 2 and 3. For cereals (wheat grain) the TF values were found to be between 0·006 and 0·009, in rather good agreement with the mean value 0·012 obtained (Papanicolaou *et al.*, in press) for 24-soil–plant pairs (wheat grain) in Greece one year after the Chernobyl accident. Our TF values for barley are, however, one order of magnitude smaller than those deduced from field experiments in Germany (Bunzl & Kracke, 1987) and England (Cawse & Baker, 1984). This difference can be partly attributed to different soil characteristics. The properties of our soils (relatively high amounts of exchangeable potassium, soil pH values, percent clay content) favor adsorption and fixation of radiocesium in the soil and adversely affect its availability to growing plants. Considerable caution should be exercised when we are comparing Transfer Factors obtained with 'Chernobyl' radiocesium to those deduced from field experiments obtained with Cesium from nuclear-weapon tests. Results up to now show (Kirton *et al.*, in press) that ^{137}Cs from Chernobyl has a greater availability to growing plants than the pre-Chernobyl ^{137}Cs originating from the fallout of the nuclear tests.

It can be seen in Tables 2 and 3 that radiocesium contamination of almost all products remains practically constant over the 3 years following the initial deposition of Cesium from the Chernobyl cloud. This means that the exponential term in UNSCEAR's model, expression (2) here, is very close to unity even for the third year ($m = 3$), i.e. parameter μ is much smaller than unity and of the same order of magnitude as the radioactive decay constant, $\lambda = 0{\cdot}023$ y^{-1}, of ^{137}Cs.

Potassium content in each crop necessary for the determination of concentration Cm in CU units, was indirectly determined by measuring ^{40}K content in every product by γ-spectroscopy. Taking $\exp(-\mu m) = 1$, p parameter of expression (2) is readily deduced from the experimental results, and is presented in Tables 2 and 3. For almost all annual products studied here, p parameter was found to be in the range between 0·003 and 0·03 CU per mCi km^{-2}. However, one cannot overemphasize the fact that plant uptake of Cesium from soils is extremely variable, so that the values deduced here for soils of Northern Greece are not typical of other regions. As an example, our values of p parameter are at least one order of magnitude smaller than those ($p = 0{\cdot}13$ CU per mCi km^{-2}) reported for cereals in the vicinity of Chicago (Evans & Bennet, 1976) and at least two orders of magnitude smaller than the values ($p = 0{\cdot}83$ CU per mCi km^{-2}) deduced from measurements at Faroe Islands, whose soils are known to transfer an abnormally large proportion of their Cesium to plants (Lassey, 1979).

Parameters TF and p in Tables 2 and 3 are of obvious practical interest in the case of the Chernobyl accident for the long term prediction of dietary contamination by radiocesium. However, the use of these results in the case of a hypothetical nuclear reactor accident is subject to caution since the conditions of the hypothetical accident will, most probably, not be the same as the Chernobyl accident.

In order to check whether the parameters relating radiocesium deposition to contamination of annual crops are independent of initial deposition, at least for a deposition range of practical interest, we have undertaken greenhouse experiments in which wheat was grown in soils contaminated with different levels of ^{137}CsCl. Radiocesium concentration in soils, wheat grain and wheat straw and the Transfer Factors relating soil concentration to plant concentration of radiocesium are presented in Table 4. In pot number 1, no additional ^{137}CsCl was introduced and the value of 54 Bq kg^{-1} of radiocesium contamination in the soil is due to the Chernobyl accident. Knowing that radiocesium deposition due to the Chernobyl accident on the farm of Thessaloniki University was 15 kBq m^{-2}, we can deduce the hypothetical depositions for pots 2–5. They range between 250

TABLE 4
Radiocesium Concentration (Bq kg^{-1}) in Soils, Wheat Grain and Wheat Straw for Different Soils Contaminated with Different Levels of ^{137}CsCl

Pot	*Soil* ^{137}*Cs (Bq kg*$^{-1}$*)*	*Wheat grain* ^{137}*Cs (Bq kg*$^{-1}$*)*	*TF*	*Wheat straw* ^{137}*Cs (Bq kg*$^{-1}$*)*	*TF*
1	54	8	0·15	15	0·28
2	972	64	0·06	103	0·11
3	1 827	81	0·04	196	0·11
4	3 487	229	0·06	316	0·09
5	5 287	268	0·05	481	0·09

In pot number 1, no additional ^{137}CsCl was introduced. The 54 Bq kg^{-1} of radiocesium contamination in the soil is due to the Chernobyl accident. Transfer Factors relating soil concentration to plant concentration of radiocesium are also reported.

and 1500 kBq m^{-2} and they are considerably higher than the maximum reported value 137 kBq m^{-2} of radiocesium deposition due to the Chernobyl accident in Greece. The results presented in Table 4 indicate that:

—Radiocesium contamination of wheat straw and grain, due to the Chernobyl accident, is bigger when wheat is grown in a pot than in the field. This can be easily understood from the fact that in the field part of the wheat root system is located in undisturbed soil (more than 30 cm depth) which has a small radiocesium contamination due to the extremely slow process of Cesium migration in undisturbed soils.

—Transfer Factors are independent of initial soil contamination (pots 2–5). This result has an obvious practical interest. The same result has been found for lower soil contamination in field experiments using the 'pre-Chernobyl' radiocesium originating from the nuclear weapon tests. Indeed in expression (2), based in UNSCEAR's empirical model from analyses of two decades of localized fallout data, the *p* parameter which determines the rate of root uptake of Cs and which is similar to the TF is independent of the initial radiocesium deposition.

—Transfer Factors are roughly three times bigger for Cesium from Chernobyl fallout than for artificial contamination with $^{137}CsCl$. This difference shows the different behavior in soils of the Chernobyl radiocesium compared with that of $^{137}CsCl$. This result has also been reported by Maubert *et al.* (1989). However, they found that Transfer Factors from soil to radishes were three times lower for Cesium from the Chernobyl fallout than for artificial $^{137}CsCl$ contamination. We do not have any simple explanation for this disagreement which leaves the question open for further investigation.

CONCLUSION

In this work, radiocesium contamination of different annual crops (cereals) due to the Chernobyl accident, was systematically studied at two experimental agricultural farms in Northern Greece for the years 1987, 1988 and 1989. Radiocesium contamination of grains is very low, almost always less than 1 Bq kg^{-1}. For all crops, the natural radioactivity due to ^{40}K is about two orders of magnitude larger than that due to radiocesium contamination. Radiocesium contamination of cereals (wheat and barley) seems to be time independent for both locations studied. This unexpected result is of obvious practical interest and calls for further investigation.

Transfer Factors and the parameters in UNSCEAR's empirical model were deduced from the experimental results. Transfer Factors, relating radiocesium deposition to contamination of crops were found to be for wheat and barley (grains) between 0·004 and 0·009, i.e. one order of magnitude smaller than those deduced from field experiments in Northern European countries, mainly due to different soil characteristics. In general, the properties of Greek soils favor adsorption and fixation of the radiocesium in the soil and so adversely affect its availability to growing plants. Greenhouse experiments show that the obtained TF values may be used in an hypothetical reactor accident with a completely different initial radiocesium deposition, taking into account, however, the different behavior of the Cesium isotope in soil due to its chemical form.

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