

ASSOCIATION European Atomic Energy Community - EURATOM Commissariat à l'Energie Atomique - C.E.A.

# INDIRECT RADIOACTIVE CONTAMINATION OF THE FOOD CHAIN DETERMINATION OF THE FACTORS OF TRANSFER SOIL/AGRICULTURAL PRODUCE AND SOIL/MILK IN THE EUROPEAN COMMUNITY

by

J. LEHR

1972



Work performed at the CEA Centre d'Etudes Nucléaires de Fontenay-aux-Roses, France Health Protection Department

Association No 003-63-10 PSAF

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#### EUR 4901 e

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In cases where several samples of fodder crops (grass and lucerne) were harvested on the same field in the course of the growing period seasonal variations occurred, to the effect that after an initial high contamination in spring followed a pronounced drop in summer, whereas in autumn the tendency was rather to increase again. It is assumed that the effect depends mainly on fluctuations of the degree of humidity of the soil.

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## ABSTRACT

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Special attention was paid to the variation of the soil factors concerned as a function of the properties of the soil, i.e. in the case of 90 Sr of the contents of the soil of exchangeable calcium and of calcium carbonate and in the case of 137 Cs of the contents of the soil of clay, organic matter and potassium.

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### **KEYWORDS**

**STRONTIUM 90** CESIUM 137 SOILS FOOD CHAIN RADIONUCLIDE MIGRATION FALLOUT QUANTITATIVE CHEMICAL ANALYSIS QUALITATIVE CHEMICAL ANALYSIS CALCIUM COMPOUNDS POTASSIUM COMPOUNDS ORGANIC COMPOUNDS CLAYS SEASONAL VARIATIONS CONTAMINATION HUMIDITY FORAGE

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### I. INTRODUCTION

The problem of the radioactive contamination of the food chain which at first owed its interest to the contamination of the environment by the nuclear test series (1954-1964) still remains topical in view of the necessity to establish security levels for the contamination by the gazeous and liquid effluents of nuclear installations, the number of which is constantly increasing,

Among the vectors of contamination the soil plays an important role, if radionuclides of long half-lives such as 90 Sr and 137 Cs are concerned.

The greater part of the information available on factors of transfer concerns specific geographic areas and mostly does not take into account differences in ecological conditions. There are, otherwise, the results of laboratory trials set up in particular to study the factors of transfer in dependence of the soil characteristics, but which are difficult. to be applied as such in practice.

To study the radioactive contamination of the food chain by way of the soil under the conditions of the European Economic Community while taking into account the differences in ecological con ditions an investigation has been carried out, in which the cumulative deposit in the soil resulting from the radioactive fallout in the years following 1954 has been used as source of radioactive contamination.

To this end a programm of sampling and of analyses has been set up in the years 1966 and 1967 in 4 out of 6 countries of the European Economic Community, comprising crops of importance for human and animal nutrition and the soils having served for their production.

a). Principle of the investigation :

Adopting in principle the formula :

$$C = p_d F_d + p_r F_r \quad (1)$$

in which C represents the concentration of radionuclide in the product;

 $F_d$  and  $F_r$  are the cumulative radioactive deposit in the soil per unit of area and the recent deposit per unit of area either per year or per month (determining the direct **contamination**), resp.;

 $\mathbf{p}_{d}$  and  $\mathbf{p}_{r}$  are the soil and rate factors , resp.

The term  $p_d F_d$  represents that part of the contamination deriving from the soil and which in years of minor fallout would approach the total contamination C (BRUCE<sup>(2)</sup>). Since in the years 1966 and 1967 fallout was not completely negligeable, C, as determined in the products of the sampling program. still needed to be corrected for direct contamination by a term  $p_r F_r = c'$ .

The soil factor thus takes the shape  $\frac{C - c'}{F_d}$ .

b). Institutes having cooperated in the investigation :

Table 1 contains an enumeration of the institutes having cooperated either in the sampling or in the analytical program or in both.

c). Sampling sites :

The map (Figure 1) represents the geographical repartition of the sampling sites corresponding to the following stations :

<u>In Italy (1966, 1967)</u>:

- 1. UDINE, clayey or loamy soils, in part calcareous.
- 2. SANT'ANGELO LODIGEANO (Po-plain), acid sandy soils somewhat loamy.
- 3. SANTA MARIA DI GALERIA (near ROME) : heavy clay soils, neutral or slightly acid.
- 4. CALTAGIRONE in SICILY : soils rich in CaCO<sub>3</sub> and clay

5. ISPRA : mainly acid sandy soils , somewhat loamy.

In Germany - Federal Republic (1967) :

- 6. HAMELN region of the Lüneburgerheide acid humic sandy soils
- 7. HUETSCHENHAUSEN (Pirmasens) : acid loamy soils
- 8. OBERSUELZEN (Alzey) : loamy clay soils, calcareous
- 9. NONNENHOF (Worms) : different soils types : sandy loamy and rich in organic matter, and loamy clay, all soils being calcareous

10. ERDING (Bavaria) : peaty soils

11. SPEYER (on Rhine) : acid sandy soils

<u>In France (1967):</u>

- 12. BENESSE MAREMME (Landes) : sandy region
- 13. BARBENTANE (Provence) : loamy calcareous soils

<u>In Belgium (1967)</u>:

14. MOL (Kempen) : region of acid humic sandy soils.

Practically all farms involved were of the mixed type, combining the production of farm crops with stock breeding. In Italy, however, cattle is mostly being kept in the stable throughout the year and the fodderconsists of products emanating from leys and arable soils. In the other countries of the European Economic Community, the cattle is generally grazing in summer, the pastures being predominantly of the permanent type. In as far as horticultural crops have been included they have been produced at farms combining agriculture with theproduction of market gardening crops. d). Sampling programm :

The crops selected for sampling were :

cereals : wheat, maize, rye (whole grains) vegetables: heading lettuce and cabbage (with removal of outer leaves) potatoes (cleanage by washing) tomatoes (whole fruits)

fruits : apples (whole fruits)

fodder crops: grass from permanent and temporary grassland lucerne (alfalfa) græn maize

other products: hay and concentrates.

rain : in as far as local information on fallout was missing.

milk : has been included, in correspondance to the sampling program of fodder crops, crops and supplements.

Four cases regarding the way of feeding can be distinguished:

- 1. By grass, the animals being for the greater part in the pastures,
- 2. By lucerne in the stable,
- 3. By a mixture of lucerne and grass in the stable,
- 4. By rations consisting of green fodders supplemented by hay and/or concentrates.

In all these cases the direct estimation of factors of transfer soils: milk has been aimed at.

## e). Analyses :

The programme provided for the radiochemical determination in all samples of 9Sr and 137Cs, and for a number of chemical analyses,

necessary either to estimate the factors of transfer, or to allow a better judgment of the results :

In vegetable produce and milk : K and Ca ;

In soils : exchangeable K and Ca , organic matter, pH , CaCO<sub>3</sub>, exchange capacity and mechanical analysis.

For the radionuclides in the soil a total determination was aimed at, for the chemical analyses conventional methods were used, allowing to obtain comparable and reproducible results, suitable to define the factors of transfer.

A number of verifications were made to make sure that all data regarding the soils were sufficiently consistent.

f). Fallout in the years 1966 and 1967

Basing ourselves on all information available the deposit in the years 1966 and 1967 for the stations involved was as follows :

	$mCi \frac{90}{Sr/km^2}$	mCi $137$ Cs/km <sup>2</sup>
1966 : range estimated aver	1,8 <b>- 4</b> ,6 age	2,6 - 6,5
for the C.E.E.	2,7	3,5
1967 : range estimated aver	0,8 - 3,3 rage	1,0 - 4,5
for the C.E.E.	1,3	1,8

g). Cumulative deposit

Table 2 states levels of contamination of the soil in 1966 and 1967, according to results of the present investigation, averaged per station.

### II. RESULTS CONCERNING STRONTIUM - 90

## a). Corrections for direct contamination

Though the years 1966 and 1967 were marked by low and decreasing values of fallout, for a number of crops allowance had to be made for the contribution to the contamination by direct transfer from the environment . When expressing the activity per g Ca the variation of Ca-content of the crops may give rise to important differences in ratio between  $p_r$  and  $p_d$  ("rate" and "soil factor"). The correction for direct contamination may be practically negligeable in the case of a crop like lucerne ( with up to 2 per cent of Ca on dry matter) whereas

it can be quite appreciable in the case of cereals (average Ca content = 0,045 per cent).

The following treatment has been adopted :

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Wheat : Corrections have been made by applying a rate factor of 240 (corresponding to a deposit per month), but by comparing

the results of calculations, either based on the deposit in the month of June, or on the average deposit in the two months before harvest,

- rye: corrections were made taking into account the deposit in the month of May,
- maize and oats : corrections were not deemed necessary in regard to the less exposed position of the grains,

potatoes and root crops : no corrections were deemed necessary,

- heading lettuce and cabbage : the outer leaves having been eliminated at sampling, the contamination has been considered to derive predominantly from the soil,
- herbage : we applied a rate factor of 410 pCi for a deposit of 1 mCi/km<sup>2</sup> per month , this in combination with the deposit in the month preceding harvest.

In the case of grass and lucerne several series of samples were available, each series originating from one and the same field and cut in successive periods of the season. An attempt has been made to use the pertinent observations for a direct estimate of  $p_r$ , the variation of  $F_r$  being known and assuming that  $F_d$  was constant throughout the season. It appears, however, that the fluctuations of C - the total activity - cannot be explained from variations in fallout during the growing season and obviously represent a seasonal effect ( see paragraph III d ).

b). Expression of results :

Two ways of expression have been used for the results, i.e. the Observed ratio (O.R.) and the soil factor  $p_d$ 

The Observed Ratio =  $\frac{pCi^{90}Sr/gCa - agricultural produce}{pCi^{90}Sr/g Ca - substrate}$ 

is considered to be of particular value for physiological investigations since the transfer from environment to vegetable and animal produce, is largely determined by the calcium content of the substrate or of the nutrition. In the present case the exchangeable calcium content of the soil is concerned;

The soil factor 
$$p_d = \frac{pCi^{90}Sr/gCa - agricultural produce}{mCi^{90}Sr/km^2}$$

represents a practical value, conceived for the purpose of the survey to relate the contamination of food stuffs to the radioactive deposit per unit of area. As such it is an empirical average, applicable to a given geographical unit, but without consideration of the varying properties of the soil. If applied to pedological units  $p_d$  has the character of a variable coefficient, related in particular to the Ca content of the soil.

## c). Results

Table 3 presents the values of O.R., computed for the different products, stated for the C.E.E. as a whole and also as average values per country or station (Table 3). (In this and the following tables the indication 0, 0 for activities of soils or agricultural products means that no certainty exists as to the second decimal).

It should be remarked that the application of different ways of correction in the case of wheat and of herbage did not result in important differences in outcome.

The available data suggest a somewhat higher O.R. for rye than for other cereals. It is at any rate important to note thatrye may be heavier contaminated than cereals like wheat and barley owing to the fact that the former crop is mainly cultivated on rather acid soils, for which at equal deposits the 90Sr/Ca ratio is automatically higher than on soils well provided with Ca.

In comparison with potatoes, carrots and beet show on average lower values of O.R.

On the whole, when only judging from average values of O.R. for different products, there is little variation between species.

Values mostly oscillate around O.R. = 0,5 with as exceptions the green vegetables. It should be reminded that the latter have been grown in the same conditions as agricultural crops and that the results are not necessarily applicable to horticultural crops in market gardening centres.

Column III of table 3 states average values for O.R. for cereals, leafy crops and fruit in general, based on literature data from pot experiments and laboratory investigations with soils. Though the general level of the O.R. is the same in both cases there is no close correspondence between the values of columns I and II on the one hand and those of column III on the other, suggesting that under practical conditions still other factors are at work than those that can be studied under laboratory conditions.

d). Soil factors : relation between p<sub>d</sub> and the exchangeable calcium content of the soil : position of the calcareous soils.

A graphical treatment of the available data for all soils concerned (acid, neutral, calcareous) by plotting p against the exchangeable calcium content of the soil, mainly shows two different tendencies, dependent on the type of soil.

In the case of acid and neutral soils (including those containing small amounts of  $CaCO_3$ ) curves of a regressive tendency are obtained, as shown by figures 2 (for grass) and 3 (green maize).

In fact, if the Observed Ratio for a given species or product can be considered to be constant, the soil factor should increase in inverse ratio to the exchangeable calcium content of the soil.

in which x is the exchangeable Ca-content of the soil in g/kg. The latter term can be reduced to the same dimensions as the soil factor. If the weight of the arable layer is taken to be 300.000.000kg/km<sup>2</sup>, the conversion factor <u>pd 3,3</u> and taking into account all units

$$p_{d} = \frac{3,3}{x} \cdot R.O.$$

In the case of grassland we adopt  $P_d = \frac{8}{x} \cdot R O$ .

When looking for a possible relation between  $p_d$  and the CaCO<sub>3</sub> content of calcareous soils (cases of wheat and of the grass) it appears that the tendency for  $p_d$  was rather to increase with the CaCO<sub>3</sub> content, which of course is against expectation (see fig. 4 for grass); since the above relation between  $p_d$  and R.O. seems at any rate 'o loose its validity in calcareous soils an explanation should be looked for in other circumstances, which will be referred to further in the discussion.

e). Transfer of 90Sr from foddercrops to milk and from soil to milk

Table 4 presents the factors of transfer from fodder to milk, from soil to fodder and from soil to milk, considered as a straight transfer. The transfer has been expressed by Observed Ratios, which showed to be less variable than factors expressed in another way.

The O.R. for the mixture of lucerne and grass have been estimated on a 50 per cent/ 50 per cent basis; the composition of the variation of the rations was known in all cases.

No account was made for possible corrections for direct contamination which on average can be set at 10 per cent. The O.R. milk obviously fodder

is not in need of being corrected.

Mean errors have been added, not  $t_{\odot}$  suggest some hypothetical accuracy, but as an indication of the variation of O.R.

A comparison of the 4 groups of alimentation is strictly speaking impossible. It is at any rate of interest that with the exception of the case of lucerne/grass the average O.R. milk/soil varies very little. For purposes of survey it is an important conclusion that the O.R. milk/soil is on the whole somewhat less variable than the O.R. milk/fodder, permitting to define the contamination of the milk straightly as a function of  $F_d$ . As an average value of O.R. milk/ soil we propose 0,07.

## f). Discussion

Irrespective<sup>t</sup> the way in which the passage of the <sup>90</sup>Sr from soil to plant is expressed, the results are always more or less variable, a fact which on ly in part can be explained from the chemical properties of the substrate, i.e. the soil. The impression is thus that apart from these rather invariable characteristics of the soil, circumstantial influences play a role which may in part mask the effects of the soil type under consideration (see IV).

A regular difference which should be pointed out here concerns the behaviour of calcareous and noncalcareous soils. Even the Observed Ratio , which according to experiments in the laboratory represents a rather characteristic value, is on the whole two times higher in calcareous than in noncalcareous soils. The phenomenon suggests a relatively great availability of <sup>9</sup>Sr in regard to Ca in calcareous soils, but its explanation does not lie ready to hand. It could be looked for in a reduction of the soil volume accessible to plant roots, either by bad structure, or the presence of lumps of CaCO<sub>3</sub>, the idea being that <sup>9</sup>Sr is for an important part concentrated on the surface of the soil aggregates whereas Ca is more evenly distributed in the soil. The absence of a permeable sub-soil may furthermore give rise to an increased <sup>9</sup>Sr/Ca ratio. Both conditions may present themselves in soils of the rendzine type, situated on lime stone.

A similar phenomenon may in the case of non calcareous soils be responsible for variable results, according to the structure of the soil and the possible occurrence of gravel or lumps of soil; it may also offer an explanation for a possible disagreement with factors of transfer determined in laboratory trials.

The difference stated above between calcareous and non calcareous soils should be also accounted for when establishing soil factors, for the purposes of survey. Thus table 5 presents an exemple of the possible variation of  $p_d$  in calcareous soils under influence of a reduced soil volume.

In the case of acid and neutral soils including soils containing some percents of calcium carbonate the average O.R. for all crops is 0,6, excepting leafy vegetables. From this a soil factor can be derived by taking into account the thickness of the arable layer and the exchangeable Ca content. Table 6 thus presents an estimate of  $p_d$  for arable crops (including leys), by taking the plow depth at 20 cm, the weight of the arable layer at 300.000 kg and calcium contents of the soil varying between 0,5 and 5 to 6 g/kg.

Values of  $p_d$ -milk have also been calculated, but only concerning the case of cattle grazing on permanent grassland.

### **RESULTS CONCERNING CESIUM - 137**

## a). Corrections for direct contamination

The calculation of the corrections in the case of cesium-137 sets greater problems than for strontium-90 in view of the fact that the direct contamination may by far surpass the contamination by the soil, the information on factors of transfer being also scarce.

A certain contribution of direct contamination originating from other plant argans by dislocation in the plant is also possible, but had to be disregarded in the present investigation.

# <u>Wheat</u>: The case of cereals is especially precarious since direct contamination may roughly account for 80 °/<sub>o</sub> of the total

<sup>137</sup>Cs-activity (case of 1966 and 1967). Plausible results were obtained by applying a rate factor of 115 pCi per kg of wheat for a deposit of 1 mCi/km<sup>2</sup>/month, monthly average of 2 months preceding harvest.

<u>Rye</u>: The same treatment as in the case of wheat

Maize and oats : No corrections

Potatoes : No corrections

<u>Grass</u>: The corrections were based on a value of  $p_r = 288 \text{ pCi/kg}$ dry matter per mCi/km<sup>2</sup>/month.

<u>Lucerne</u>: About one third of the contamination is due to direct contamination but application of corrections is empeded by seasonal effects (see III d). <u>Milk</u>: Corrections have been applied in the case of grass, lucerne and the mixtures of grass and lucerne, but were not possible in the case of rations of more complicated composition. In the latter case the contribution of direct contamination is estimated at 50 °/., which should be taken into account when judging the results of this way of alimentation.

## b). Expression of results .

In the case of cesium - 137 an expression of the type of Observed Ratio seems to be of little value (See par. III g discussion) and the expression most indicated appears to be the soil factor, either

In the following a summary will be given for each product of the estimated values of  $p_d$ , their range and average, as also the range of averages per country.

There after it will be examined in how far the soil factor for  $^{137}Cs$  is correlated with certain properties of the soil, namely with its contents of clay, of organic matter and of potassium.

For cereals an average soil factor of 0,2 appears to be acceptable; it compares well with the estimate of BRUCE (2) for wheat floor,  $p_d = 0,068$ ; which for total grains would correspond to  $p_d$  about 0,17 The higher value of rye is certainly of significance in relation to the type of soil generally used for its cultivation.

The average  $p_d$  of potatoes is not very different from that of carrots (0,4) and that of sugar beet (0,3); in the case of fodderbeet a higher value has been found (1,2), which might be related to differences in depth of rooting and the fact that the fodder-beet root is in part exposed to direct contamination.

In the case of <sup>137</sup>Cs green vegetables appear to be less contaminated by the soil than grass and lucerne. Just the reverse has been found for the contamination by <sup>90</sup>Sr.

## d). Seasonal effects .

An examination of the dependence of the transfer on the soil type is not well possible without a preliminary verification of the influence on the results of the season.

It has already been remarked that great fluctuations of contamination occur in samples of lucerne and grass, taken in the course of the season, always on the same field, and which cannot be attributed to the variation in fallout.

The general trend of the phenomenon, as shown by figure 5 for the 90 sr activity of lucerne is for an initially high activity in spring, a very substantial decline in summer and a tendency to increase again in autumn. For grass the tendencies are the same, but, somewhat less clear.

The drop of the activity in summer has also been noted by SQUIRE and MIDDLETON (3) and SQUIRE (4) in the case of grassland, where the  $^{90}$ Sr/Ca ratio was always high in the beginning of the season and showed a considerable drop in the months of June and July. It has also been found that in a dry year the contamination of grass

is inferior to that in a year with abundant rainfall (5).

According to SQUIRE the drop in activity in summer can be explained by the fact that under dry conditions of the soil the grass is rooting in deeper and less contaminated layers. This explanation seems, however, to be less satisfactory in the case of lucerne. Especially in the case of 137Cs we would rather think of a difference in availability of the deposit in dependence of the water content of the soil. Other factors may interfere such as differences in speed of growing, or a gradual exhaustion of the available potassium in the soil in the course of the season.

The size of the effect can obviously be variable according to local conditions; but for general use it might be reckoned on differences in the order of 3:1 in spring and summer.

e). Relations between  $p_d = \frac{137}{Cs}$  and type of soil.

As individual results show too great a dispersion to be treated graphically the tendencies have been studied by dividing the available points into classes of increasing values of either exchange capacity, clay content, organic matter content or potassium content.

It appears then that in those cases where a clear trend of values is discernable (see tables 8 up to 11 inclusive) this trend mainly concerns the upper limits of the range and the average values for a given class. The lower limits generally do not show a substantial variation;.

el). Exchange capacity and/or clay content.

Potatoes showed a correlation of  $p_d$  which was most pronounced for the German samples - (table 8)

When expressing the soil factor in <u>pCi</u>  $\frac{137}{\text{Cs/g K}}$  the regression mCi  $\frac{137}{\text{Cs/km}^2}$  is also clear, even if eliminating several samples of very low and non measurable activity from the class with high clay contents. (Table 9).

Also in the case of wheat p<sub>d</sub> shows a tendency to decrease with increasing values of the exchange capacity or of the clay content (Table 10)

No clear trend of  $p_d$  as a function of the clay content could be demonstrated in the case of grass, which may be attributable to different circumstances :

- different stage of development of the samples

- overlapping of tendencies by seasonal effet

- different conditions of water economy in the soils con cerned.

It should also be considered that owing to the reduced thickness of the mat horizon its water content is more subject to fluctuations than in the arable layer.

e2). Potassium content

In the case of wheat and potatoes no effect of potassium on  $\ensuremath{\textbf{p}}_d$  has been discernable.

Only in the case of grass is there a certain evolution of  $p_d$  with the potassium content of the soil (Table 11).

e3). Organic matter

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In no case has it been possible to establish a relation between  $p_d$  and the organic matter content of the soil. In view of the restricted number of samples available this result is certainly not conclusive.

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f). Transfer of <sup>137</sup> Cs from foddercrops to milk and from soil to milk

Results have been summarized in table 12, in which the factors of transfer have been presented in two ways, namely as,

$$f_{d} = \frac{pCi^{137}Cs/litre \text{ or } kg}{pCi^{137}Cs/kg}$$
(milk, fodder)  
(fodder, soil )

and as O.R. =  $\frac{pCi^{137}Cs/gK}{pCi^{137}Cs/gK}$  (milk or fodder) (fodder or soil) The passage from soil to milk is considered as a straight transfer (Table 12).

The dispersion of results is much greater than in the case of  ${}^{90}$ Sr, but depending on the way of expression, O.R. being less variable for the transfer from fodder to milk and f for the transfer either from soil to fodder or from soil to milk.

As an average discrimination factor  $\frac{pCi/l \text{ milk}}{pCi/kg \text{ soil}}$  one could

adopt  $f_d = 0,05$  (cases of grassland, lucerne and rations), but when taking into account the differences in the thickness of the mat horizon and of the arable layer it leads to different values of  $p_d$ , namely for grass  $p_d = 0,34$  and for lucerne 0,13.

g). Discussion

The case of  $^{137}$ Cs presents itself in a more complicated way than that of  $^{90}$ Sr for two different reasons :

- whereas strontium in soils remains largely available cesium is fixed for the greater part, the fraction of the deposit which is available for absorption by plants being at most some percents. As such it is variable in dependence of the type of soil, according to its contents of clay, organic matter and of potassium,

- even for the absorption from culture solutions an expression of the type of the observed ratio  $\frac{137 \text{Cs/K} - \text{plant}}{127 \text{Cs/K} - \text{plant}}$  presents no

<sup>137</sup>Cs /K-solution advantages as a factor of transfer, since it varies with the K concentration of the solution. It can, however, be used to define an upper limit of contamination, as in the presence of sufficient potassium its value is very near to unity, whereas in the case of a shortness of potassium this element is absorbed preferentially.

The main objection for its use for practical purpose is, meanwhile, that it cannot be based on the total  $C^s$  content of the soil, unless in combination with a factor for its availability.

An additional investigation having as an objective to relate the absorbable <sup>137</sup>Cs to the characteristics of the soil will be reported on shortly, reason for which we will not enter here into a discussion on the variability of the soil factor in dependence on the type of soil.

It is at any rate important to remark that in view of the subordinate role of the indirect contamination an exact determination of factors of transfer from soil to plant would seem of secondary importance, provided that no critical conditions for the contamination by way of the soil present themselves.

From the literature four factors are known which together may determine a critical condition :

- 1. A low content of clay (sandy soils)
- 2. An appreciable content of organic matter (humic sandy soils, peaty soils),
- 3. A deficiency of potassium,
- 4. An ample supply of water, possibly determined by the climate, the season or a high water level in the soil.

In laboratory experiments the effect of the organic matter has been well established and if in the present investigation no influence on the transfer has been found probably not all conditions for its coming into existence had been fulfilled. In this connection it should be thought in particular of combined effects of organic matter and of water in the soil, to the effect that a sufficiently wet condition of the soil may be a condition for the organic matter to exert its influence .

For the contamination of the food chain the order of the different products is of interest. From table 13 can be seen that according to approximate average values of p the green vegetables (legumes) and apples belong to the heaviest contaminated products, but if expressing the transfer per g potassium per unity deposit (mCi/km<sup>2</sup>) cereals, milk and apples bring in more Cs. For a final

judgment of risks the composition of the daily rations should of course be

taken into account.

# IV. INTERPRETATION OF RESULTS IN REGARD TO DEFINING CRITICAL LEVELS

The present investigation has been carried out with the aim to determine under practical conditions factors of transfer and to use them in the same time for the establishment of critical levels.

As in all cases the determinations were based on single observations, circumstantial conditions have had rather great influence on the results, which could not always be accounted for and have sometimes masked the effects of the type of soil.

Obviously predictions of levels of contamination of crops can only be approximative as there are a number of factors which cannot be controlled. Thus it must be admitted that climatic and seasonal effects may cause factors of transfer to vary by a factor of 3, that merely by different plow depth variations from 1:2 in  $p_d$  are possible, that furthermore differences in tillage and in fertilization or in ground water level and the nature of the subsoil are all of influence, influence in itself variable from year to year.

Though factors of transfer can best be defined on the basis of average conditions, an ample marge of security should be observed for the definition of critical levels (maximum permissible levels) for the deposit in the soil, so as to cope with the most unfavorable conditions.

In the case of <sup>90</sup>Sr principally the light sandy soils (mostly acid and of low adsorptive capacity) can be considered to present critical conditions for the contamination; as far as calcareous soils are also apt to become critical this in a sense will depend more on circumstancial conditions than on the very soil characteristics.

In adopting the values of tables 5 and 6 for the establishment of critical levels for regional use, it would seem appropriate to apply a factor of security of 2 if soils of good cultural conditions are concerned. For soils where the conditions are less favourable (bad structure, shallow plow layer, impermeable under-ground, presence of gravel, high water level, a.o.) the factor should be chosen higher, at least 5.

In the case of 137Cs additional information is wanted more in particular regarding the influence of soil humidity on the availability of the 137Cs.

Investigations into this question are also in progress.

## LIST OF CAPTIONS

- Fig. 1 Map showing the geographical repartition in the European Economic community of sampling sites for the study of the radioactive contamination of agricultural products by way of the soil.
  - 1; UDINE
  - 2. ST ANGELO
  - 3. CNEN CASACCIA
  - CALTAGIRONE
  - 5. ISPRA
  - 6. HAMELN
  - HUETSCHENHAUSEN
  - 8. OBERSUELZEN
  - 9. WORMS
  - 10. KREIS ERDING
  - 11. SPEYER
  - 12. BENESSE MAREMME
  - 13. BARBENTANE
  - 14. CEN MOL
- Fig. 2 Relation between the soil factor for grass (<sup>90</sup>Sr) and the exchangeable calcium content of the soil, acid and neutral soils.
- Fig. 3 Relation between the soil factor for green maize (<sup>90</sup>Sr) and the exchangeable calcium content of the soil.
- Fig. 4 Relation between the soil factor for grass (<sup>90</sup>Sr) and the calcium carbonate content of calcareous soils.
- Fig. 5 Seasonal effects on contamination of lucerne by <sup>90</sup> Sr.











Country or Station	Organism in charge of the sampling	Organism in charge of the analyses
ITALY 1966	Istituto d'Igiene PAVIA Prof. CHECCACCI Prof. LANZOLA	Istituto d'Igiene PAVIA
ITALY 1967	Istituto d'Igiene PAVIA	Istituto d'Igiene
ISPRA 1967	Servizio di biologia Euratom Establishment Dr. BOURDEAU -ISPRA	Servizio di fisica Sanitaria Euratom establishement ISPRA - Prof. MALVICINI
FR <sup>AN</sup> CE 1967	Syndicat pour l'amélio- ration des sols GARGENVILLE Dr. REDLICH	IRAMIR à Marseille Prof. ROUX S.A.S. GARGENVILLE
GERMANY 1967	Landwirtschaftliche Versuchs - und Forschungsanstalt SPEYER - Prof.SIEGEL	Idem SPEYER
MOL 1967	Dpt de Radiobiologie CEN-MOL Dr. KIRCHMANN	Dpt Mesures et Controles des Radiations CEN-MOL Dr. BOULANGER

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Range of value of the average cumulative deposit per station as determined in the present investigation.

	mCi per km <sup>2</sup>		
	90 <sub>Sr</sub>	<sup>137</sup> Cs	
Italy (1966) without ISPRA and CALTAGIRONE	55 <b>-</b> 1 <b>02</b>	113 - 180	
Italy (1967) idem	43 - 62	87 - 104	
Caltagirone	17	37	
Ispra	139	225	
German Federal Republic	35 <b>-</b> 83	6 <b>0 -</b> 141	
France	45 - 60	56 <b>-</b> 125	
Mol (Belgium)	46	97	

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90 Summary of results on Sr (Observed Ratios) for the European Community as a whole and of averages per country.

Product N s	Number of samples	range of single values	average of all values I	range of averages per country or station	mean values per country or station II	values based on data from the litterature III
CEDEALS						
Wheat	27	0.0-3.15	<b></b> 0 56	0 0 1 02	0.37	0.4
Wheat Bro	5	0,0=3,15 0.20=1,33	x 0,50	$0, 0^{-1}, 0^{-1}$	0,57	0,4
Barley	7	0,2,7-1,55	0,70	-	0,12	
Dariey	6	0,0-0,70	0,30	0 23-0 44	0.33	
Maize	14	0,01-1,84	0,46	0,01-0,54	0,37	
FODDER CI	ROPS					0,7à0,8
' Herbage	47	0,0-2,15	0,54	0,15-0,72	0,45	
Lucerne	<b>9</b> 3	0,02-1,39	0,46	0,02-0,64	0,38	
Green Maize	11	0,13-2,89	0,86	0,13-2,89	1,06	
LECUMES						0,7à0,8
	8	0 18-2 86	1 17			
Cabbage	8	0,02-5,84	1,5			
FRIITS						0.4
Tomatoes	5	0.09-1.46	0.53			
Apples	5	0,07-0,72	0,37			
TUBER ANI	I D ROOT CF	ROPS				
Potatoes	22	0,0-2,68	. 0,69	0,14-0,72	0,46	
Carrots	3	0,02-0,57	1 0,23			
Fodder- beet	3	0,06-0,43	0,19			
Sugar beet	6	0,16-0,72	0,44			

\* An extremevalue (ISPRA 7,7) has been discarded from the average

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# Transfer of <sup>90</sup>Sr from fodder to milk, from soil to fodder and from soil to milk, expressed as Observed Ratios.

Alimentation .	Number of	p <u>Ci<sup>90</sup>Sr/gCa-milk</u>	<u>pCi/gCa-fodder</u>	<u>pCi/g Ca-milk</u>
	observations	pCi/g Ca-fodder	°pCi /gCa -soil	`pCi/g Ca-soil
Grass	16	0,21 <u>+</u> 0,15	0,35 <u>+</u> 0,17	0,060 <u>+</u> O,041
	*	71 °/。	49 °/。	68 °/。
Lucerne	4	0,13 <u>+</u> 0,07	0,61 <u>+</u> 0,25	0,069 <u>+</u> 0,024
	*	53 °/°	41 °/。	35°/。
Lucerne/	4	0,30 <u>+</u> 0,21	0,75 <u>+</u> 0,46	0,164 <u>+</u> 0,035
Grass	*	70°/。	61 °/。	21 °/。
Rations	15	0,16 <u>+</u> 0,09	0,61 <u>+</u> 0,30	0,081 <u>+</u> 0,048
-	*	58°/。	49°/。	59°/。

\* Factors of variation

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Approximation of  $p_d = {}^{90}$ Sr in calcareous soils, based on the content of CaCO<sub>3</sub> and the exchange capacity of the soil, if O.R. = 1 and the thickness of the arable layer = 15 cm.

°/。CaCO <sub>3</sub>	EXCHANGE CAPACITY m.e. / 100 g					
	10	20	30			
10	2,4	1,2	0,8			
20	2,6	1,3	0,85			
30	2,8	1,4	0,9			
40	3,0	1,5	1,0			
50	3,3	1,7	1,1			
Neutral non calcareous soil : O.R. = 1 ; arable layer of 20 cm						
0	1,65	0,8	0,55			

Estimate of  $p_d = \frac{90}{3}$ Sr for acid, neutral and slighty calcareous soils, on average for agricultural produce and milk in dependence on the exch<sup>a</sup>ngeable Ca of the soil.

	Sandy soils		Loamy and clavey soils		
	C	Content of exchangeable Ca of the soil			
	0,5g/kg lowest value	lg/kg on average	2,5 g/kg on average	5 to 6 g/kg highest value	
Agri. products	4	2	0,8	0,4	
milk	1	0,6	0,25	0,1	

Summary of results on Cs for the C.E.E. as a whole and of averages per country.

Product	Number of samples	Range of single values	Average of all values	Range of averages per country or station Mean	value to be adopted <b>as</b> a mean
Wheat	27	0,0-0,93	0,19	0,0-0,29 0,18	0,2
Rye	5	0,30-0,45	0,39	0,38-0,39 0,39	0,4
Maize	14	0,0-1,24	0,21	0,14-0,33 0,21	0,2
Potatoes	22	0,0-3,83	0,60	0,0-1,47 0,46	0,5
Grass	47	0,0-14,0	2,58	1,81-4,90 2,8	2,5
Lucerne	33			1,8-2,2 2,0	2,0
Lettuce	8	0,0-2,46	0,74		
Cabbage	8	0,0-5,23	0,96		
Tomatoes	5	0,0 <b>-</b> 0,55	0,22		
Apples	5	0,0-1,87	0,78		

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Dependence on the exchange capacity and the clay content of the soil factor for  $^{137}$ Cs of potatoes.

Exchange capacity	Clay content	Number of samples	Range	Average
I. 4-7 m.eq./100g	0-7°/。	. 5	0,21-1,58	1,00
II. 10-30	11-19°/。	4	0,37-0,85	0,50

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Dependence on clay content of the soil factor for potatoes expressed as <u>pCi 137-Cs/g K</u> mCi 137-Cs/g Km<sup>2</sup>

Clay content	Number	German soils Range Average		All soils with measu: Number Range		able activity Average
I. 0-7°/。	5	0,030-0,125	0,078	9	0,005 <b>-</b> 0,125	0,050
II. 11-20 °/。	4	0,017-0,044	0,026	6	0,017-0,044	0,026

Dependence on exchange capacity and clay content of the soil factor for  $^{137}$  Cs of wheat.

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	Exchange capacity m.e./100g	Number of Samples	Range	p d Average	Clay content	Number of samples	Range	p <sub>d</sub>   Average
I	4-9	8	0,0-0,93	0,34	0-9°/。	5	0,0-0,93	0,29
п	10 - 14	4	0,03-0,26	0,15	10 <b>-</b> 19°/。	12	0,0-0,81	0,19
III	15-19	4	0,0-0,33	0,10	20-40°/。	5	0,0-0,06	0,01
IV	20-30	6	0,0-0,06	0;01				

Dependence on the K content of the soil of the soil factor of grass, expressed either in pCi/kg per mCi/km<sup>2</sup> or in pCi/gK per mCi/km<sup>2</sup>

K content of soil mg/100g	Number of samples	P <sub>d</sub>		pCi/gK mCi /km <sup>2</sup>	
		Italige	Average	Kange	Average
I 25-100	20	0,0-14,2	2,7	0,0-0,42	0,21
II 101-200	8	0,0-4,76	1,9	0,03-0,28	0,09
III 201-400	14	0,0-3,62	2,4	0,0 - 0,16	0,09
IV 401-700	5	0,52-2,98	2,0	0,02-0,12	0,07

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Transfer of <sup>137</sup>Cs from fodder to milk, from soil to fodder and from soil to milk.

Alimentation	Number of Samples	a. $\frac{pCi^{137}Cs / l \text{ or } kg}{pCi^{137}Cs / kg}$		
		milk/fodder	foddær/soil	milk/soil
Grass	14/12	0,03 - 4,3 average 0,11	0,037 - 1,025 0,34 / 0,36	0,002 - 0,920 0,124 / 0,049
Lucerne	4	0,034 - 0,680 0,36	0,03 - 1,24 0,77	0,020 - 0,056 0,042
Grass/Luc.	4	0,06 - 0,28 0,18	0,45 - 0,89 0,61	0,055 - 0,146 0,098
Rations	11/12	0,024 - 0,540 0,16	0,21 - 2,15 0,63	0,014 - 0,125 0,068

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Alimentation	Number of Sam <b>p</b> les	b. <u>pCi <sup>137</sup>Cs /g K</u> pCi <sup>137</sup> Cs / g K		
•		milk/fodder	fodder/soil	milk/soil
Grass	14/12	0,51 - 9,6 2,35/1,81	0,0009-0,0187 /0,0019	0,001 - 0,158 0,016/0,004
Lucerne	4	0,70 - 1,23 1,13	0,0001-0,0153 0,0063	O,0002 - 0,005 0,0065
Grass/luc.	4	0,23 - 3,40 1,28	0,010 - 0,141 0,050	0,0003 - 0,053 0,026
Rations	11/12	0,52 - 3,26 1,49	0,0005 - 0,0320 0,0092	0,0010 - 0,06 0,0184

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Order of contamination by  $^{137}$ Cs of different food stuffs according to average values of  $p_d$  or of  $p_d/K$  content in g per kg

Order	Products	Approximate average value of p d	Average content of potassium °/° on dry matter	p <sub>d</sub> /gK per kg
1.	Milk	0,04-0,10	l,5 g/f	0,03-0,07
2.	Wheat	0,2	0,45	0,04
2.	Maize	0,2	0,4	0,05
3.	Carrots	0,4	2 - 3	0,02
3.	Tomatoes	0,4	4	0,01
4.	Potatoes	0,5	<b>2 -</b> 3	0,02
5.	Lettuce	0,7	4 - 6	0,01
6.	Apples	0,8	0,7	0,12
7.	Cabbage	1,0	4 - 5	0,02

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