

# ***Modelling of resuspension, seasonality and losses during food processing***

## ***First report of the VAMP Terrestrial Working Group***

*Part of the IAEA/CEC Co-ordinated Research Programme on the  
Validation of Environmental Model Predictions (VAMP)*



INTERNATIONAL ATOMIC ENERGY AGENCY

**IAEA**

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## FOREWORD

Following the Chernobyl accident and on the recommendation of the International Nuclear Safety Advisory Group (INSAG) in its Summary Report on the Post-Accident Review Meeting on the Chernobyl Accident (Safety Series No. 75-INSAG-1, IAEA, Vienna, 1986), the Agency established a Co-ordinated Research Programme on "The Validation of Models for the Transfer of Radionuclides in Terrestrial, Urban and Aquatic Environments and the Acquisition of Data for that purpose". The programme seeks to use the information on the environmental behaviour of radionuclides which became available as a result of the measurement programmes instituted in the Soviet Union and in many European countries after April 1986 for the purpose of testing the reliability of assessment models. Such models find application in assessing the radiological impact of all parts of the nuclear fuel cycle. They are used at the planning and design stage to predict the radiological impact of planned nuclear facilities, in assessing the possible consequences of accidents involving releases of radioactive material to the environment and in establishing criteria for the implementation of countermeasures. In the operational phase they are used together with the results of environmental monitoring to demonstrate compliance with regulatory requirements regarding release limitation.

The programme, which has the short title "Validation of Environmental Model Predictions" (VAMP), was started in 1988; it is jointly sponsored by the Division of Nuclear Fuel Cycle and Waste Management and the Division of Nuclear Safety and is also supported by the Commission of the European Communities. There are four working groups within the programme; they are the Terrestrial Working Group, the Urban Working Group, the Aquatic Working Group and the Multiple Pathways Working Group. This is the first report of the Terrestrial Working Group.

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## Chapter 1

### INTRODUCTION

The Terrestrial Working Group of VAMP aims to examine, by means of expert review, the state-of-the-art in modelling the transfer of radionuclides from the terrestrial environment to man. An aim of the studies is to evaluate the lessons learned and to document the improvements in modelling capability as a result of post-Chernobyl experience. The topics for review were decided upon following the first Working Group Meeting. In making this choice, consideration was given to the relative importance of a given process as a potential contributor to radiation dose to man, the degree of uncertainty which exists in the area because of lack of knowledge and the possibilities presented by Chernobyl for improvement of understanding of the process.

Reviews of three of the identified topics are presented in this report; they are:

Resuspension - the process in which surface deposits of particulate matter are re-entrained into the air by mechanisms such as wind disturbance and man-made activities.

It has been demonstrated that in dry environments, particles can be resuspended for many years after their initial deposition. In the case of radionuclides this could represent a long term pathway of radiation exposure, especially for those long lived radionuclides which present most hazard via the inhalation route. The validity of the resuspension models developed on the basis of the evidence from dry environments in other more vegetated and damper environments had never been adequately tested. The fall-out from the Chernobyl accident affected environments which are mostly in temperate regions and an opportunity for testing resuspension models was presented.

Food processing losses - the removal of radionuclides from the food-chain to man as a result of food processing and culinary preparation.

The significance of the reductions in radionuclide contamination of foodstuffs which result from food processing and culinary preparation has



been recognised for many years. However, the information available on the reduction factors achieved by the various processes and as a function of radionuclide was scarce and poorly documented. The need for realistic dose assessments after the Chernobyl release made modelling deficiencies of this type apparent and stimulated new work in the area.

Seasonality - the varying response to radioactive contamination of food crops and other environmental systems to the time of year when the contaminating event occurs.

The variation of the contamination of environmental materials with season has been most clearly demonstrated in grain crops. Early experiments showed that depending upon the stage of growth of a crop when contaminated, the level of radionuclide concentration in the final grain product can vary by more than a factor of 100. Because of the season of the year when the Chernobyl accident occurred and because the fall-out affected countries in different geographic latitudes, a significant variation of the apparent degree of radionuclide transfer to the same crop type with latitude was seen. This was due to seasonality effects. Pre-existing food-chain models did not represent the seasonality effect very well, if at all. The Chernobyl experience has emphasised the need for it to be taken into account in predictive models.

The following reviews were prepared by the named authors but have had the benefit of a review process which involved the members of the Terrestrial Working Group of VAMP. The versions presented in this document have been revised to take into account the comments received from the Working Group. Publication was finally decided upon at a meeting of the full VAMP Research Co-ordination Meeting in March 1991, and after the approval of VAMP members (by correspondence) in Autumn 1991.

## Chapter 2

### RESUSPENSION FOLLOWING CHERNOBYL

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#### 2.1. Introduction

Resuspension comprises the processes which may raise deposited material from the ground. It is thus related to, but in principle distinct from, suspension, where no previous airborne condition is implied, and entrainment, which is suspension caused by the wind. However, the resuspension mechanisms usually include the entrainment of particulate material in winds passing over surfaces, and this can occur over any natural or artificial surface from which particles or droplets may be detached by the wind or other disturbance. Therefore, the distinction between resuspension and suspension is often ignored. The material initially deposited may be changed in its physico-chemical characteristics during its contact with the surface. For example, it might become attached to other particles. In conditions of continuous emission from the primary source, it can be difficult to make quantitative measurements of resuspended material, because of its similarity to its source material before deposition and the relative sizes of the concentrations of the material before deposition and after resuspension.

Resuspension has the potential to cause persistent air concentrations following a brief release, and to redistribute material deposited on the ground. Inhalation hazards, recontamination of previously cleaned surfaces and contamination of crops are possible consequences. Plans for coping with the aftermath of accidents that result in spills or airborne releases, and consequence assessment models, must make allowances for resuspension processes.

Despite many measurements, the prediction of resuspension remains uncertain. Many factors influence resuspension, including various facets of the weather, the surface structure and the nature of the contaminant

TABLE I

## SOME FACTORS THAT MAY INFLUENCE RESUSPENSION

|   |
|---|
| Time since deposition   |
| Wind speed  |
| Nature of surface: vegetation, building etc.                              |
| Surface moisture  |
| Soil chemistry and texture  |
| Size distribution of contaminant particles                                |
| Chemical properties of contaminant  |
| The deposition process: wet or dry, wind speed, precipitation amount etc. |
| Mechanical disturbance by traffic, agriculture etc.                       |
| Depth and method of cultivation   |
| Intensity and frequency of rain   |
| Snow cover or freezing of the surface                                     |

(see Table I). Consequently, field measurements show a wide range of behaviour and it is often difficult to associate changes with specific causes.

Data available from observations at American nuclear sites and weapon test sites have been summarised by Sehmel [1] and the wide range of information available on the topic has been described by Sehmel [2] and Nicholson [3]. Generally, arid or semi-desert areas predominate in the results, and a strong dependence on wind speed is apparent. Additional studies have used wind tunnels or controlled applications of tracers to outdoor surfaces to investigate individual resuspension mechanisms (e.g. Garland, [4, 5]. Resuspension by traffic, agriculture and some other types of mechanical disturbance has been demonstrated, as summarised by Nicholson [3]. Experiments designed to investigate such factors have also demonstrated an important reduction in resuspension with time after deposition. Several models have been proposed to describe this effect (see Ref. [6]).

Results from wind tunnel experiments, mechanistic field investigations, and even monitoring results from contaminated areas within nuclear establishments or weapon test sites, may be inappropriate for application in some areas. They may represent too great or too small an area of contaminated surface or the climate, surface roughness, or the density of traffic or other source of disturbance may differ. In principle the effects of these parameters have been investigated and a correction could be attempted for recognised differences, but knowledge is incomplete and uncertainties would be great. This is particularly true for the humid climate of Europe where there have been few investigations of resuspension.

The Chernobyl accident of 26 April 1986 provided a unique opportunity to observe resuspension from a brief event which caused widespread contamination in Europe. This paper attempts to draw together data from several laboratories that have reported measurements following Chernobyl, and to compare the results with expectation based on previous experience.

## 2.2. Definitions

Three parameters have been used to describe resuspension [3]. The first is the resuspension factor:

$$K \text{ (m}^{-1}\text{)} = \frac{\text{Concentration in air, } C_a \text{ (Bq m}^{-3}\text{)}}{\text{Surface deposit, } d \text{ (Bq m}^{-2}\text{)}} \quad (1)$$

which has the advantage that both numerator and denominator are directly measurable. However, measurement of  $K$  may be complicated since the concentration in air may include important contributions from processes other than resuspension. In addition  $K$  is likely to be strongly dependent on such details of measurement technique as the height of air sampling above the surface. In evaluating  $d$  a decision must be made regarding the depth of surface to be included, and it is not straightforward to decide what depth of soil may contribute to airborne radioactivity. In practice, for prediction of resuspension from an accidental dispersion, it is convenient to use the total amount initially deposited, without allowance for vertical mixing into the soil or for removal by weathering.

$K$  has the drawback that it must depend on the variation of  $d$  with distance from the point of measurement. At locations with relatively low

deposit, transport by the wind from areas of high deposit may dominate air concentrations. Although there has been some theoretical investigation relevant to this problem, [7, 8], an appropriate averaging scale for  $d$  has not been established.

In principle, the difficulty of spatial variations in deposit is circumvented by the use of the resuspension rate:

$$\Lambda \text{ (s}^{-1}\text{)} = \frac{\text{Resuspension flux, } R \text{ (Bq m}^{-2} \text{ s}^{-1}\text{)}}{d \text{ (Bq m}^{-2}\text{)}} \quad (2)$$

Thus,  $\Lambda$  is the fraction removed per second by resuspension. This quantity can often be derived directly in wind tunnel measurements and laboratory studies. However, it can only be readily deduced from field observations in exceptional circumstances. In principle,  $\Lambda$  may be included in atmospheric transport models to describe the movement of contamination from one area to another by resuspension and deposition, as well as to predict air concentrations (eg. Ref. [8]). Such models, with a knowledge of  $\Lambda$ , appear essential for predicting air concentrations at locations downwind of a contaminated area.

The use of the equivalent soil concentration

$$S_E \text{ (kg m}^{-3}\text{)} = \frac{C_a \text{ (Bq m}^{-3}\text{)}}{\text{Concentration in surface soil, } C_s \text{ (Bq kg}^{-1}\text{)}} \quad (3)$$

avoids the need to decide what depth of soil to include in the calculation of  $K$ . It allows surrogate soil tracers to be used to estimate air concentrations from a radioactive contaminant in soil, but it has not been much used.

The definitions of  $K$ ,  $S_E$  and  $\Lambda$  make no allowance for the effects of the many environmental variables that may influence resuspension rates. Large variations must therefore be expected with wind speed, time after deposition and with the other parameters listed in Table I.

### 2.3. Available Estimates of Resuspension

The resuspension factor,  $K$ , is the parameter that has been used most frequently to describe the results of previous studies, and for predicting the consequences of the contamination of surfaces. Field measurements

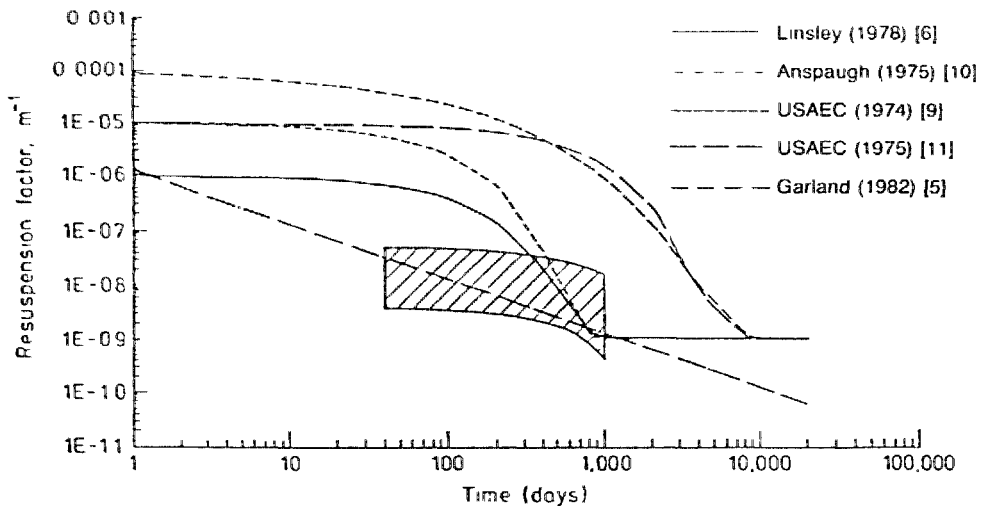


FIG 1 Resuspension factors ( $K$ ) measured in Europe (hatched area) compared with models. The model relationships are: Linsley  $K = 10^{-6} \exp(0.01 t) + 10^{-9}$ ; Anspaugh  $K = 10^{-4} \exp(-0.15 t^{1/2}) + 10^{-9}$ ; USAEC (1974)  $K = 10^{-5} \exp(-0.0139 t) + 10^{-9}$ ; USAEC (1975)  $K = 10^{-5} \exp(-0.00185 t) + 10^{-9}$ ; Garland  $K = 1.2 \times 10^{-6}/t$ ; where  $t$  is time after deposition in days and  $K$  is in  $m^{-1}$ .

(e.g. Ref. [1]) suggest that soon after deposition  $K$  is of the order of  $10^{-6}$  to  $10^{-4} m^{-1}$ , but over a period of years,  $K$  declines to approximately  $10^{-9} m^{-1}$ . A number of empirical models represent this behaviour (see Fig. 1). One model [9] allows  $K$  to vary with particle size, ascribing a value to each of several size intervals and setting  $K$  inversely proportional to deposition velocity.

The last model listed in Fig. 1 is based on wind-tunnel studies [4, 5]. These studies were in agreement with field experience, in that  $K$  increased strongly with wind speed and decreased with time following deposition.  $K$  also increased with particle diameter in the range from about 1 to 5  $\mu m$ . The decrease with time was represented by an inverse-time relationship.

Information on  $K$  long after deposition can be gleaned from measurements of nuclear weapon test debris. Air concentrations at ground level have always been subject to some input from the stratospheric reservoir, so that only upper limits on  $K$  can be deduced. Data for  $^{137}Cs$  in air over the United Kingdom show that, 13 years after peak deposition,  $K$  was less than  $2 \times 10^{-9} m^{-1}$  and, after 22 years, less than  $5 \times 10^{-10} m^{-1}$ . Consideration of radionuclide ratios and trace element concentrations in air and soil has been used to refine limits

further, and Garland [4] and Cambray et al. [12] argue that  $K$  cannot have been greater than  $7 \times 10^{-11}$  or  $2 \times 10^{-10} \text{ m}^{-1}$ , 15 or 18 years after deposition, respectively.

The effect of countermeasures on resuspension is indicated by observations at Palomares. In 1966 two nuclear weapons were destroyed by fire following an accident in the air. The bombs burned on impact with the ground causing plutonium contamination over an area of a few  $\text{km}^2$ . Subsequently, continuous measurements of plutonium in air were made at four sampling stations in the contaminated zone [13]. Using estimates of the initial level of deposition and the air concentration data, the mean values of  $K$  for the first year range from  $0.2$  to  $2 \times 10^{-9} \text{ m}^{-1}$ . Soon after the accident top soil was removed from the worst affected areas, and in other areas surface concentrations were alleviated by deep ploughing. Presumably these actions account for the low values of  $K$ . During the following four years, air concentrations declined rather erratically at all stations about five-fold. At two stations where measurements continued, the air concentrations remained roughly constant at the reduced level for a further seven years, but rose again in 1977 when cultivation began at a previously unused, contaminated area. These data apply to rather arid conditions in southeast Spain.

#### 2.4. Opportunities from Chernobyl

In the following, the observations after Chernobyl will be examined in terms of  $K$ . Generally, only long term mean concentrations are available, and the response of  $K$  to many of the parameters in Table I are hidden in values which are appropriate to the climate, rather than the short term weather.

The Chernobyl accident yielded a rare opportunity for the study of resuspension, since it provided what could be considered as a large single pulse of radionuclides of limited duration. This pulse was fairly rapidly dispersed over a large part of Europe. Deposition continued over a few weeks, and was essentially complete by the end of June. However, data given by Cambray et al. [14] shows that over 90 per cent occurred by mid May.

The main contaminated air mass arrived at locations experiencing considerably different climatic regimes, where, for example, in some cases

spring was well advanced and in others was in the early stages. Continental, maritime and mediterranean situations all received a signal from Chernobyl. This brings the possibility of observing the transfer of radionuclides through the environment, including the resuspension process, under a variety of conditions. The comparison of such observations may enable the relative significance of the various conditions to be established.

Data obtained following Chernobyl permit direct estimation of resuspension in many areas and extend the information available on resuspension to climates not previously represented. However, these observations have limitations. They give information on only one radioelement - caesium. The deposition was very variable in some areas, and even where numerous measurements of the amount deposited have been made their representativeness may be in doubt. There is some question as to whether local resuspension is the only source of material measured in the air: contributions due to transport by the wind of resuspended material from more heavily contaminated areas may be significant.

$^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were the most widely distributed long lived radionuclides from Chernobyl. The ratio between these radionuclides varied within a narrow range about  $^{137}\text{Cs}/^{134}\text{Cs} = 1.6$  [14] and this ratio has been used as evidence that the persistent radiocaesium in the atmosphere did derive from Chernobyl. However, the shorter half-life and smaller quantity of  $^{134}\text{Cs}$  released in the accident make  $^{134}\text{Cs}$  of minor interest in assessing resuspension.

Information on other radioelements would be of significant interest. Resuspension may be influenced by the physico-chemical behaviour of radionuclides in soil and other environmental compartments. For example, Garland [4, 5] observed an increase in resuspension with particle size. Many other radionuclides were observed in Europe soon after the accident. However, they were mostly short lived or were present in small quantities and gave little opportunity for observing resuspension.

Garger et al. [15] observed the resuspension rates of  $^{144}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{95}\text{Zr/Nb}$  within the 30 km exclusion zone around Chernobyl. Some differences in resuspension rates were observed but there was not a consistent pattern. On some occasions the resuspension rate for  $^{137}\text{Cs}$  exceeded that for  $^{144}\text{Cs}$  and for  $^{95}\text{Zr/Nb}$ , while in other cases the reverse was true.



The limited data suggest that any differences between radionuclides are much smaller than the two or three orders of magnitude variability between measurements on different occasions. However, these results apply near the release point, where large fuel particles probably dominated deposition for the radionuclides measured, and a different behaviour might apply in other circumstances.

## 2.5. Data Following Chernobyl

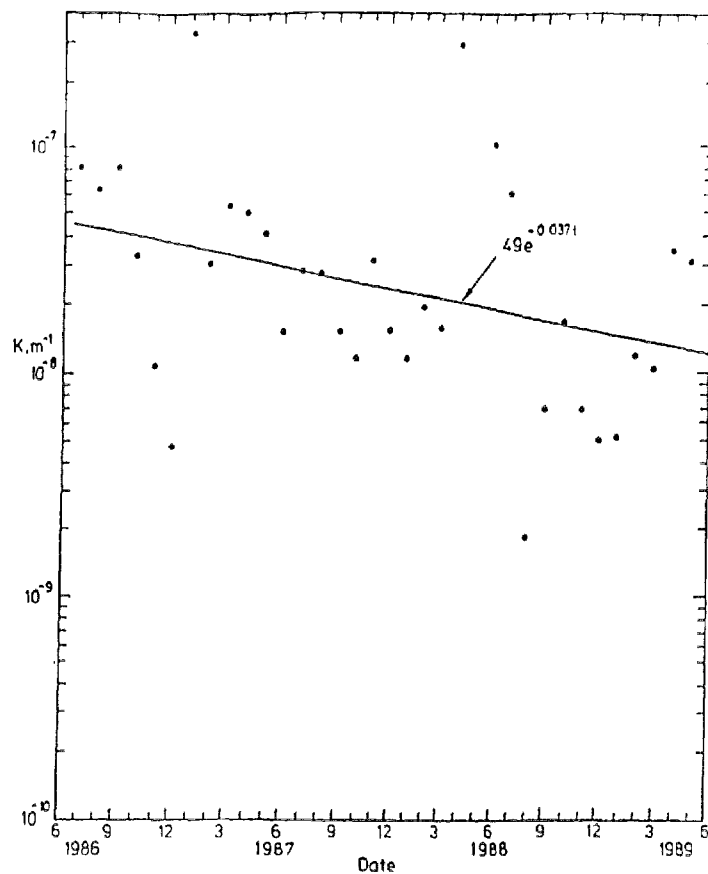
### 2.5.1. Time series resuspension data

Experimental results for monthly average  $^{137}\text{Cs}$  concentrations in near-ground air have been obtained for 14 stations across Europe. Typically, the measurements used high volume air samplers, operating at sampling rates of 1 to 10 m<sup>3</sup> per minute and using high efficiency filters.

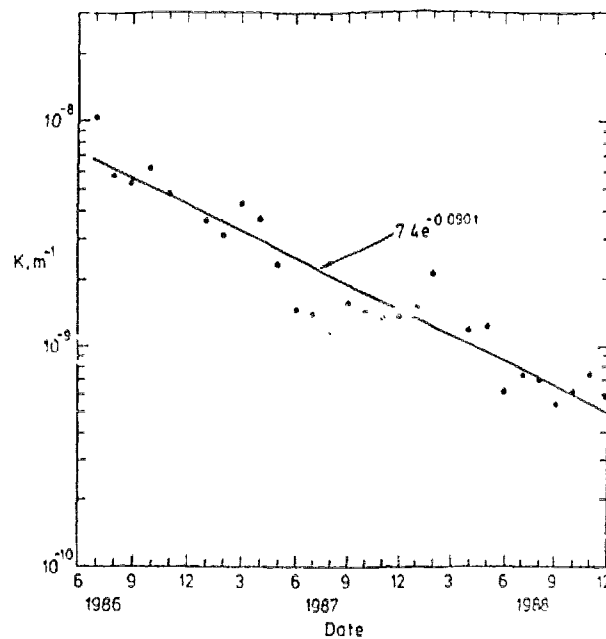
In all cases, measurements of the direct deposit from the Chernobyl accident were available, by either direct deposition (wet and dry) or soil sampling, where the Chernobyl contribution was estimated from the  $^{134}\text{Cs}$  concentration. This has enabled the air concentrations to be converted to resuspension factors. In this review, the Chernobyl direct deposit has been assumed to be the deposit measured between the date of the accident and 30 June 1986. After this time the deposit has been assumed to be due to indirect mechanisms, such as resuspension. Thus, the resuspension factor at time T was calculated by dividing the measured air concentration at time T by the amount deposited soon after the accident, with no correction for gains due to deposition after 30 June 1986, or removal by resuspension or by dispersal or transport within the soil.

Examples of the variation of resuspension factors with time are shown in Fig. 2. Each data set has been fitted by an exponential function which is also shown in the figures. A summary of the parameters is given in Table II.

For the 14 sites, the initial deposit varied from 52 to 43700 Bq/m<sup>2</sup>. In all cases the resuspension factor, K, decreased with time over the observation period, and the exponential function  $A \exp(-Bt)$  gave a reasonable fit to the data. Values of the parameter A, corresponding to K in mid-June, 1986, were in the range  $(3.6 - 49) \times 10^{-9} \text{ m}^{-1}$ , and values of the parameter B, which specified the exponential decline in resuspension, were in the range 0.026 - 0.124 per month.



CHILTON  $^{137}\text{Cs}$  RESUSPENSION FACTOR ( $\text{m}^{-1}$ )



NEUHERBERG  $^{137}\text{Cs}$  RESUSPENSION FACTOR ( $\text{m}^{-1}$ )

Fig. 2. Examples of data for resuspension of  $^{137}\text{Cs}$  from Chernobyl.

TABLE II

 $^{137}\text{Cs}$  RESUSPENSION FACTOR (K)

$$K(t) \text{ (m}^{-1}\text{)} = \frac{\text{Air concentration (t) (Bq m}^{-3}\text{)}}{\text{Surface deposit (Bq m}^{-2}\text{)}}$$

Time dependence fitted by exponential function:

$K = A \exp(-Bt)$  (t = time in months after mid-June 1986)

| Location              | Initial Deposit (Bq m <sup>-2</sup> ) | Period of Observation | A (m <sup>-1</sup> X10 <sup>-9</sup> ) | B (month <sup>-1</sup> ) | Reference   |
|-----------------------|---------------------------------------|-----------------------|--|--------------------------|-------------|
| Klagenfurt, Austria   | 43700                                 | 1/87-12/89            | 3.6                                    | 0.049                    | 16,17,18,19 |
| Neuherberg, Germany   | 19700                                 | 7/86-12/88            | 7.4                                    | 0.090                    | 20,21       |
| Bregenz, Austria      | 13200                                 | 1/87- 8/89            | 5.3                                    | 0.061                    | 16,17,18,19 |
| Ispra, Italy          | 11900                                 | 7/86- 6/88            | 5.9                                    | 0.026                    | 22,23,24,25 |
| Nurmijärvi, Finland   | 7100                                  | 7/86-12/89            | 10.0                                   | 0.063                    | 26,27,28,29 |
| Vienna, Austria       | 4000                                  | 1/87- 8/89            | 12.8                                   | 0.053                    | 16,17,18,19 |
| Warsaw, Poland        | 3200                                  | 7/86-10/89            | 15.0                                   | 0.062                    | 30, 31      |
| Braunschweig, Germany | 2700                                  | 7/86-12/88            | 28                                     | 0.124                    | 32, 33      |
| Eskmeals, UK          | 2500                                  | 7/86- 6/89            | 16.0                                   | 0.082                    | 34, 35      |
| Berlin, Germany       | 2300                                  | 7/86-12/88            | 35                                     | 0.099                    | 32, 33      |
| Risø, Denmark         | 800                                   | 7/86-12/88            | 31                                     | 0.106                    | 36, 37, 38  |
| Bornholm, Denmark     | 620                                   | 7/86-12/88            | 31                                     | 0.082                    | 36, 37, 38  |
| Skibotn, Norway       | 195                                   | 7/86-12/88            | 35                                     | 0.060                    | 32, 33      |
| Chilton, UK           | 52                                    | 7/86- 6/89            | 49                                     | 0.037                    | 39,40,41    |

There is a negative correlation between the initial deposit and A. The Spearman rank correlation coefficient is -0.96 which is significant at the >99.9% level, (see Fig. 3). However, no systematic variation of B with initial deposit is apparent.

The variation of K with deposit has been confirmed in a larger data set in Table III, which includes some stations for which the data were unsuitable for the time trend analysis of Table II. Table III also shows that the dependence of K on deposit has persisted over two years. Moreover, Bjurman et al. [42] observed resuspension at four sites in

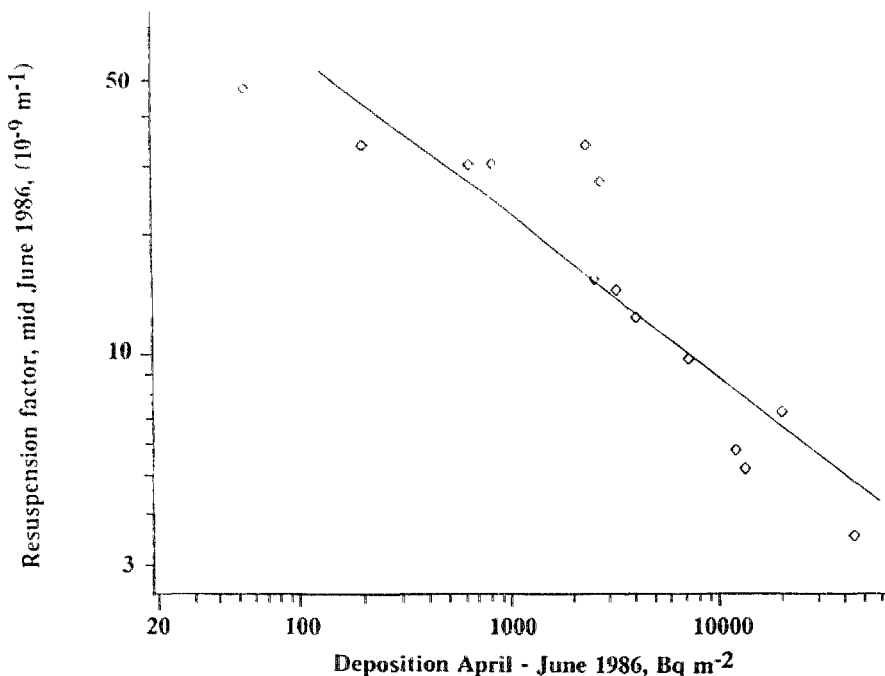


Fig. 3. Variation of the resuspension factor in mid-June 1986 with the amount deposited during April-June 1986.

Sweden. At Gavle, the deposit was 30 to 50 times larger than at the other Swedish sites, and K averaged over July to December 1986 was up to 10 times smaller.

The correlation between A and the initial deposit shows that, although the concentration of <sup>137</sup>Cs in air soon after deposition increases with the amount initially deposited, the rate of increase is significantly less than proportional to the deposit. Two possible explanations for this difference have been considered. Generally, different deposition mechanisms operated for large and small deposits, the large deposits being caused by wet deposition in heavy rain while the small deposits were due mainly to dry deposition. A real difference in resuspension factors, consistent with the observations, might be due to a greater availability for resuspension of material deposited dry than for wet deposited material. However, a 'memory' of a year or more of the circumstances of deposition would be required, and this seems improbable. The second explanation involves long range transport of resuspended material. This would increase the observed concentration in areas of low deposit, but reduce it in areas of high deposit, and produce an apparent variation in

TABLE III

 $^{137}\text{Cs}$  AIR CONCENTRATIONS AND MEAN RESUSPENSION FACTORS

| Site                  | Initial<br>Deposit<br>(Bq m <sup>-2</sup> ) | Mean air<br>concentration<br>( $\mu\text{Bq m}^{-3}$ ) |           | Resuspension<br>factor<br>(10 <sup>-9</sup> m <sup>-1</sup> ) |           |
|-----------------------|---|--|-----------|---|-----------|
|                       |   | 7/86-6/87  | 7/87-6/88 | 7/86-6/87   | 7/87-6/88 |
| Neuherberg, Germany   | 19700                                       | 96   | 27        | 4.9   | 1.4       |
| Ispira, Italy         | 11900                                       | 69   | 54        | 5.8   | 4.5       |
| Nurmijärvi, Finland   | 7100  | 68   | 23        | 9.6   | 3.2       |
| Lerwick, UK           | 4600  | 2.7  | 1.4       | 0.6   | 0.3       |
| Warsaw, Poland        | 3200  | 39   | 14.5      | 12  | 4.5       |
| Braunschweig, Germany | 2700  | 46   | 9.9       | 17  | 3.7       |
| Eskmeals, UK          | 2500  | 37   | 7.8       | 15  | 3.1       |
| Berlin, Germany       | 2300  | 57   | 19.6      | 25  | 8.5       |
| Conlig, UK            | 2200  | 6.7  | 2.6       | 3.0   | 1.2       |
| Eskdalemuir, UK       | 820   | 9.5  | 3.3       | 12  | 4.0       |
| Risø, Denmark         | 800   | 18.5   | 3.6       | 23  | 4.5       |
| Bornholm, Denmark     | 620   | 14.4   | 4.4       | 23  | 7.1       |
| Skibotn, Norway       | 195   | 6.3*   | 3.3       | 32  | 17        |
| Tromsø, Norway        | 175   | 2.6  | 1.7       | 15  | 9.7       |
| Orfordness, UK        | 110   | 10.3   | 4.0       | 94  | 36        |
| Compton, UK           | 105   | 29   | 3.0       | 280   | 29        |
| Chilton, UK           | 52  | 3.1  | 2.7       | 60  | 52        |
| Harwell FC, UK        | 52  | 3.8  | 1.8       | 73  | 35        |
| Harwell CM, UK        | 52  | 69   | 6.2       | 1300  | 120       |
| Milford Haven, UK     | 18  | 1.5  | 0.7       | 80  | 40        |

\* 11 months data

Data sources as for Table II and Garland and Cambray [43] for additional UK stations.

resuspension factor. Very simple modelling of the transport process suggests that such an explanation is plausible.

In the case of the Chilton data set, corresponding to one of the smallest deposits, there is some evidence, albeit inconclusive, of a spring peak in the air concentration. A spring peak is characteristically

observed in the effects of debris from past nuclear weapon tests, and is explained by the return of material from a stratospheric reservoir. Thus there is a suggestion that a small fraction of the Chernobyl release was injected into the stratosphere. (Aoyama, [44], reported similar observations in Japan.) The Chernobyl material returned from the stratosphere would be difficult to identify at locations which had received large deposits because of the relatively large contribution from local resuspension. Hence, there would be more chance of observing a stratospheric effect at Chilton than at most other sites. Stratospheric return would also reduce the differences in air concentration between sampling sites, and might contribute to the lack of proportionality between air concentration and deposit.

#### 2.5.2. Traffic

Measurements of  $^{137}\text{Cs}$  in air at Risø (Aarkrog, et al. [36]) show a pronounced weekly cycle during July and August 1986. Concentrations were high during weekdays and low at weekends. Later this pattern disappeared. The weekly periodicity was attributed to resuspension by traffic of Chernobyl deposit on nearby roads, which were busy on working days but quiet at weekends. After August, the Chernobyl deposit had presumably been removed from roads or fixed to the road surface and was no longer available for resuspension.

A comparison between two sites at Harwell Laboratory also suggested that traffic is important. One sampling site (Harwell CM, in Table III) is located in a car park and the second (Harwell FC), a kilometre away, in a field with no roadway within a few hundred metres. A 20-fold difference in air concentration was observed during the year July 1986 - June 1987, and the difference persisted, though somewhat diminished, in the following twelve month sampling period. Although the car park and the neighbouring roads are busy for only one or two hours each day, the highest post-Chernobyl resuspension factors were observed at this locality.

#### 2.5.3. Scale length for resuspension

Large differences between neighbouring sites show that variations in air concentrations occur on a scale not exceeding some tens of kilometres. Indeed, where local variations in traffic occur, concentrations may vary substantially in a kilometre or so. Further indications of the length

scale for resuspension are found in the data for selected sites. Weekly measurements for  $^{137}\text{Cs}$  in air at the island of Bornholm and Risø were compared (Aarkrog et al. [37]) with the frequency of wind speed and direction. At both sites, there was a positive correlation of air concentration with winds from the east, and negative correlations with westerly winds. Both sites are coastal with land to the east and water to the west, and the correlations support the view that the source of resuspension is dominantly nearby land areas, within a range of about 20 km (the distance across Bornholm). No correlations with wind speed were revealed.

Low resuspension factors were found at Lerwick (Table III) which is near the east coast of a strip of land only 5 km wide on the island of Shetland.

#### 2.5.4. Comparison with models

The range of the exponential function fitted to the values of  $K$  is shown in Fig. 1, along with models previously used to provide a description of resuspension. The models show a wide divergence over the time interval occupied by the data. To some extent this is determined by the different circumstances and uses intended for the models. The Chernobyl data tend to support the models predicting lower rather than higher concentrations, but the limited time range of the Chernobyl data currently available must be kept in mind. In addition, most of the data sets reflect conditions where disturbance by man and vehicles is slight. People working out of doors are liable to generate dust locally and to be exposed to higher concentrations in consequence, and Linsley [6] suggested that an increase of an order of magnitude in predicted concentrations may be appropriate.

#### 2.5.5. Deposition of resuspended $^{137}\text{Cs}$

Garland and Cambray [43, 45] noted two unexpected features of deposition measurements following Chernobyl. Total apparent deposition velocities (i.e. the flux to a deposition collector funnel divided by the air concentration) ranged up to about  $3 \text{ m s}^{-1}$ , and the amount of  $^{137}\text{Cs}$  collected during July 1986 to June 1987 at some sites was comparable to that deposited in May–June 1986. In comparison, Aarkrog [37] noted that at Risø and Bornholm the amount deposited in 1987 was just 2.1 per cent of the initial deposit.

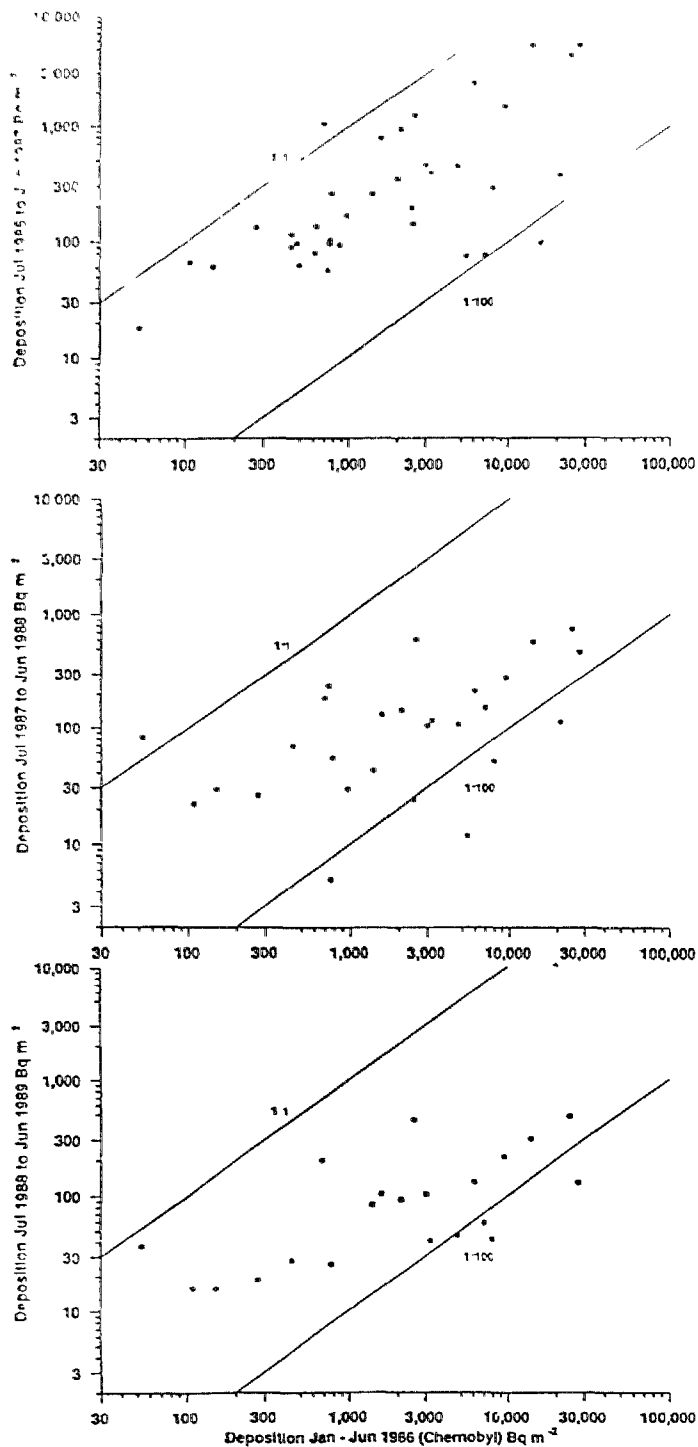


Fig. 4. The amount of resuspended material deposited during (i) July 1986–June 1987;  
(ii) July 1987–June 1988;  
(iii) July 1988–June 1989.



The deposition of resuspended  $^{137}\text{Cs}$  over a period of three years, at several sites in Europe, is displayed in Fig. 4. A decrease with time is apparent, as is also the wide variability in the ratio to the amount originally deposited. There is no apparent trend in this ratio with the amount initially deposited, but the wide scatter in the results may have obscured such a trend.

It seems probable that the observed deposition involves material resuspended very locally, as a result of rain splash, wind or mechanical disturbance. Particles of at least several tens of microns diameter would explain the high apparent deposition velocities. Air samplers are prone to undersample such large particles and their sedimentation velocity is of order tens of centimetres per second. The explanation of apparent deposition velocities of order  $1 \text{ m s}^{-1}$  probably involves an exaggeration of the true deposition velocity as a result of biased measurement of the air concentration. The very variable deposition velocities and fractions re-deposited suggest that local characteristics of surface condition and vegetation cover have a marked effect on the suspension rate, or that the amount collected may be strongly influenced by the size and height and the exposure of the deposition collector.

## 2.6. Significance of Resuspension

### 2.6.1. Air concentration due to resuspended particles

Models which describe resuspension in terms of the resuspension factor,  $K$ , may readily be used to compare the predicted inhalation dose due to resuspended material with the dose caused by the passage of the plume resulting directly from a release of airborne material. This section sets out the results of such a comparison for the resuspension models introduced earlier (Fig. 1) and also considers the corresponding comparison derived from field measurements following Chernobyl.

It is convenient to make these comparisons by calculating the ratio  $A_R/A_I$ , where

- $A_R$  is the air dose (that is, the integral of concentration in air over time) due to resuspended material and
- $A_I$  is the air dose due to the initial passage of the cloud of airborne material.

If  $V_T$  is the total deposition velocity due to dry and wet deposition processes, the quantity deposited during the initial passage of the cloud is given as  $D_I V_T$  and the subsequent concentration in air due to resuspension will be  $D_I V_T K(t)$  where  $K(t)$  is the resuspension factor at time  $t$  following the initial passage of the cloud.

Thus, for each resuspension factor model, the ratio

$$\frac{A_R}{A_I}(T_1, T_2) = V_T \int_{T_1}^{T_2} K(t) dt \quad (4)$$

gives a measure of the relative importance of resuspension during the period  $T_1$  to  $T_2$  in comparison with the initial passage of the cloud. The ratio may be calculated for any value of  $T_1$  and  $T_2$  within the range of validity of the models listed in Fig. 1, while the measured air concentrations following Chernobyl can be used to calculate  $A_R/A_I$  directly for times,  $T$ , exceeding about 2 months. The appropriate value of  $V_T$  for this comparison requires some consideration. Following Chernobyl,  $V_T$  ranged from  $\sim 0.1 \text{ mm s}^{-1}$  in areas where dry deposition occurred to  $\sim 0.1 \text{ m s}^{-1}$  in areas where heavy rain coincided with the arrival of the plume. For the calculations, a value of  $0.01 \text{ m s}^{-1}$  has been used as a representative value. It should be remembered that larger values of  $V_T$  may occur both where heavy rain enhances deposition, or where very large particles are present, and that  $A_R/A_I$  is proportional to  $V_T$ . Table IV shows the values of  $A_R/A_I$  calculated for the resuspension models for times up to 50 years, probably an upper limit of the time for which the model can be considered valid. The table also includes corresponding ratios determined from measurements of  $^{137}\text{Cs}$  in air for a number of locations where sufficient data is available to determine  $A_R$  and  $A_I$ . Some caution is advised in applying these ratios for radiological assessment purposes, since the size distributions and behaviour on inhalation of the initial and resuspended aerosols, may differ substantially.

The values shown in the table emphasise the large differences between the various models. Some predict resuspension doses similar to or greater than the dose due to the original cloud, while others predict resuspension doses which are quite insignificant in this comparison. The measurements generally indicate that the latter resuspension models are closer to the truth. Low values of the ratio were observed even where rain resulted in high deposition.

TABLE IV

THE RATIO OF RESUSPENDED AIR DOSE  $A_R$  TO DIRECT AIR DOSE  $A_I$  FOR  
FIVE K-MODELS AND DETERMINED DIRECTLY FROM  
MEASUREMENTS FOLLOWING CHERNOBYL

| Model or<br>Location | $A_R/A_I$ for time intervals |           |             |              |
|----------------------|------------------------------|-----------|-------------|--------------|
|                      | 0 to 365 d                   | 0 to 50 a | 60 to 425 d | 425 to 790 d |
| Anspaugh             | 5.9                          | 7.6       | 3.8         | 0.84         |
| USAEC 1974           | 0.61                         | 0.64      | 0.27        | 0.002        |
| USAEC 1975           | 2.3                          | 4.7       | 2.1         | 1.0          |
| Linsley              | 0.083                        | 0.10      | 0.045       | 0.0025       |
| Garland              | 0.006                        | 0.010     | 0.0021      | 0.00064      |
| Observations:        |                              |           |             |              |
| Lerwick, UK          |                              |           | 0.1         | 0.005        |
| Milford Haven, UK    |                              |           | 0.01        | 0.005        |
| Eskdalemuir, UK      |                              |           | 0.002       | 0.0007       |
| Compton, UK          |                              |           | 0.026       | 0.003        |
| Orfordness, UK       |                              |           | 0.002       | 0.0008       |
| Tromso, Norway       |                              |           | 0.002       | 0.001        |
| Chilton, UK          |                              |           | 0.003       | 0.003        |
| Harwell CM, UK       |                              |           | 0.06        | 0.005        |

#### 2.6.2. Deposition on crops

It is well established that a fraction of material deposited from the atmosphere is retained on the leaves and other aerial parts of plants (Chamberlain and Garland, [46]): thus direct contamination of pasture grass and leaf vegetables results whenever toxic or radioactive substances are deposited. Here commonly used models are applied to estimate the possible effect of resuspended material in contaminating crops.

The fraction,  $f$ , of material intercepted by the crop canopy can be approximated by

$$f = 1 - \exp(-\mu B), \quad (5)$$

(Chamberlain, [47]), where  $B$  is the dry weight of the above-ground portion of the crop per unit area of ground and  $\mu$  is an empirical parameter, which may be a function of the physico-chemical properties of the material deposited, the mode of deposition, the structure of the crop canopy and the weather.

Observed values of  $\mu$  range from 0.2 to 4 m<sup>2</sup> kg<sup>-1</sup> (Chamberlain and Garland, [46]). There is some evidence that  $\mu$  decreases with increasing particle size. If the inference is correct that the deposition of resuspended particles is dominated by large particles, then values of  $\mu$  in the range 0.2 to 1.0 m<sup>2</sup> kg<sup>-1</sup> are probably applicable.

It is convenient to note that, for  $\mu B < 0.3$ ,  $\exp(-\mu B) \approx (1 - \mu B)$  so that (5) may be simplified to

$$f \approx \mu B. \quad (6)$$

Then the fraction intercepted per unit dry weight of crop is

$$f/B \approx \mu. \quad (7)$$

Material retained on leaves may be lost by processes such as wash-off by rain, erosion of leaf cuticle, blow-off and die back of older leaves. Several observations have shown that the consequence is an exponential decrease of contaminant from crop surfaces:

$$M/M_0 = \exp(-t/\tau), \quad (8)$$

where  $M_0$  and  $M$  are the quantities retained on the crop initially and after time,  $t$ , and  $\tau$  is an empirical time constant. During the growing season,  $\tau \sim 15$  days.

It is possible to use the simple models represented in equations (7) and (8) to estimate the ratio,  $d_R/d_I$ , of the ingestion dose due to resuspension to the ingestion dose due to the initial contamination event. The timing of the contamination event in relation to the development and harvest of the crop is crucial. Hence we consider two examples.

First, consider a hypothetical crop which is harvested and consumed at a constant rate over an extended growing season for T days after the initial deposition. Then the dose ratio will be approximately:

$$\frac{d_R}{d_I} = \frac{\mu_R \tau_R RT}{\mu_I \tau_I} \quad (9)$$

assuming that T is several times  $\tau_R$ .

The parameters  $\mu$  and  $\tau$  are as defined in equations (5) and (8) and the suffixes R and I refer to the resuspended material and initially deposited material, respectively. Because of the particle size considerations mentioned above,  $\mu_R$  may be similar to or less than  $\mu_I$ ; we consider  $\mu_R = \mu_I$  and  $\mu_R = 0.1 \mu_I$ .

R is the fraction of deposited material that is resuspended and deposited again each day. During the year from July 1986 to June 1987 the fraction of  $^{137}\text{Cs}$  from Chernobyl that was re-deposited was in the range 0.03 to ~1.0. Then R may be of order  $10^{-4}$  to  $3 \times 10^{-3} \text{ d}^{-1}$ , perhaps with a most probable value of about  $3 \times 10^{-4} \text{ d}^{-1}$ .

Using the values indicated above for the parameters in equation (9) the ratio  $d_R/d_I$  for growing season of 100 days is expected to be about  $10^{-2}$  with a possible range of about  $10^{-3}$  to  $10^{-1}$ .

This is the dose ratio that would result during the first growing season after deposition, assuming that the growing season continued for 100 days after initial deposition. Deposition, and the dose ratio, in the second growing season might be several times smaller (see Fig. 4) and would add a small fraction to  $d_R/d_I$ .

In an alternative scenario, initial deposition occurs  $T_c$  days before harvest of a seasonal crop. Various loss processes deplete the fraction of the initial deposit that remains on the crop at harvest to  $\mu_I \exp(-T_c/\tau_I)$ , while the accumulation and loss of resuspended material throughout the growing season results in a contribution  $\mu_R R \tau \{1 - \exp(-T/\tau_R)\}$ .

In this case

$$\frac{d_R}{d_I} = \frac{\mu_R \tau_R R \{1 - \exp(-T/\tau_R)\}}{\mu_I \exp(-T_c/\tau_I)} \quad (10)$$

For short  $T_c$  this ratio approximates  $\mu_R R T_c / \mu_I$  and for  $T_c > \tau_R$ ,  $d_R/d_I$  approaches  $\mu_R \tau_R R / \mu_I \exp(-T_c/\tau_I)$ . If  $T_c$  is as long as 100 days this ratio may be substantially greater than unity, as a result of the attenuation of the initial deposit.

These sample calculations suggest that resuspension may cause a modest increase in the total ingestion dose through crop contamination. Resuspension may sustain concentrations in a crop which would have been cleansed by field loss processes and may necessitate countermeasures over extended periods of time after the initial contamination event.

It is important to appreciate that the calculations have used models beyond their usual range of application, and the results must be regarded as tentative.

## 2.7. Conclusions

The measurements reviewed here have demonstrated that resuspension may result in measurable concentrations in air for a period of at least three years following deposition in climates common in Europe. The resuspension factors (K) derived from measurements in different regions of Europe all show a reduction with time after the initial deposit. The deposits varied by a factor of 800. However, with the exception of one or two sites where traffic or agriculture cause substantial disturbance shortly after deposit, K varied by a factor of 14, viz.  $(3.6 - 49) \cdot 10^{-9} \text{ m}^{-1}$ . There is a strong negative correlation between the direct Chernobyl deposit and K. A possible explanation relates to the transport of resuspended airborne activity from areas of high deposit to those of lower deposit. The reduction with time of K can be fitted with an exponential function, with the decay time constant within the range 0.03 - 0.12 per month. With few exceptions K was similar at sites with similar deposits in contrasting climatic regions.

There is evidence that the source of the resuspended material is mainly the area around the sampling site. In the first months after the accident, mechanical disturbance of the deposit in this area, e.g. by road traffic, could provide a large additional contribution to the resuspension. At a site which received a low direct deposit, there is inconclusive evidence of a small contribution to the air concentration from material returned from the stratosphere.

The air dose due to resuspension has been compared to the air dose due to the initial contaminating event at a number of measuring sites. In all cases the air dose due to resuspension during the first two years after the accident was less than 10 percent (and at most sites no more than about 1 percent) of the initial air dose.

The deposition of resuspended material has also been considered. The fraction of the deposited material that was resuspended and deposited again during the first year after the initial contaminating event ranged from 0.01 to 1.0. The potential for contaminating crops and uncontaminated surfaces may be significant.

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## Chapter 3

# THE INFLUENCE OF FOOD PROCESSING AND CULINARY PREPARATION ON THE RADIONUCLIDE CONTENT OF FOODSTUFFS: A REVIEW OF AVAILABLE DATA

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### 3.1. Introduction

#### 3.1.1. Background

Radioactive contamination of the environment may result in the increased radiation exposure of human beings due to their ingestion of radionuclides in food. In the chain of transfer processes which leads from the deposition of radionuclides on to soils and plant surfaces to their presence in the diet, food processing and culinary preparation are the last processes which can effect the radionuclide content of foodstuffs. In the years before the Chernobyl accident the effects of food processing and culinary preparation on the radionuclide content of foodstuffs received comparatively little attention although for some food products the reductions can be substantial. Studies and investigations in this field have instead focussed mainly on the processes of transfer of radionuclides in the components of the human food chain.

Knowledge of the effects of food and processing and culinary preparation is needed when assessing the radiation dose to man from the ingestion of contaminated foodstuffs. Appropriate allowances have to be made for the reductions produced by food processing\* in order that doses are not systematically overestimated. A knowledge of the effects of special food processing or decontamination techniques is needed in order to estimate their effectiveness as possible countermeasures for reducing the

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\*In the remainder of this paper 'food processing' is used as a general term to include industrial processing and domestic food preparation.

dose to man following the contamination of the food chain with radionuclides in the aftermath of an accident.

Measurements of the transfer of radionuclides during food processing are comparatively scarce. Most measurements date back to the sixties and only a few attempts have been made to review this literature [1-6]. These reviews are often limited to selected food classes. Also, the comparability of data is hindered by the variety of definitions used to define changes in radionuclide content of the food. Since the Chernobyl accident in 1986 new data have become available especially on the behaviour of radiocaesium.

### 3.1.2. Industrial and domestic scale processing

Food processing methods vary considerably from foodstuff to foodstuff and also between the industrial and the domestic scale (Fig. 1). A food product may be processed quite differently when grown locally in a garden and consumed by the grower and his family as compared with the large scale commercial production of the same food product followed by factory processing. The resulting losses in the two cases may be significantly different. In the case of milk, modern processing techniques provide several alternative products each of which may contain different concentrations of the contaminating radionuclide (Fig. 2). This may be contrasted with the small scale consumption of whole milk.

### 3.1.3. Data needed for radiological assessments

Depending upon the nature of the radiological assessment, different types of information are needed on food processing losses. For assessments of dose to identified groups of individuals living at particular locations, the ideal approach would be to obtain local information on food processing losses specific to the foods consumed by the group. This approach would minimise the predictive error associated with the food processing component of the dose calculation. However, this is rarely feasible and national or regional compilations of the type contained in this report are usually used. Nevertheless, the potential error in the use of tabulated processing loss factors can be reduced by checking that the sources of information on which the loss factor is based are relevant to the circumstances of the dose assessment, for example, whether the food is locally grown and prepared or is subject to industrial processing.

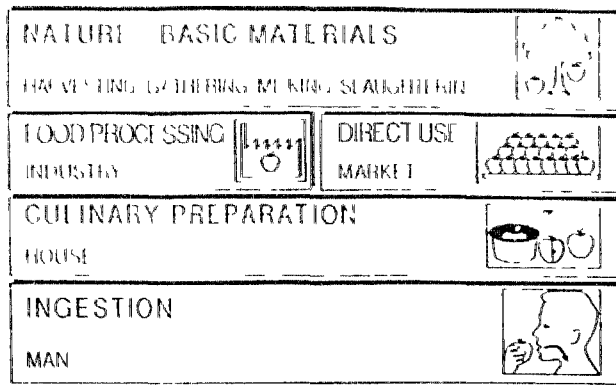
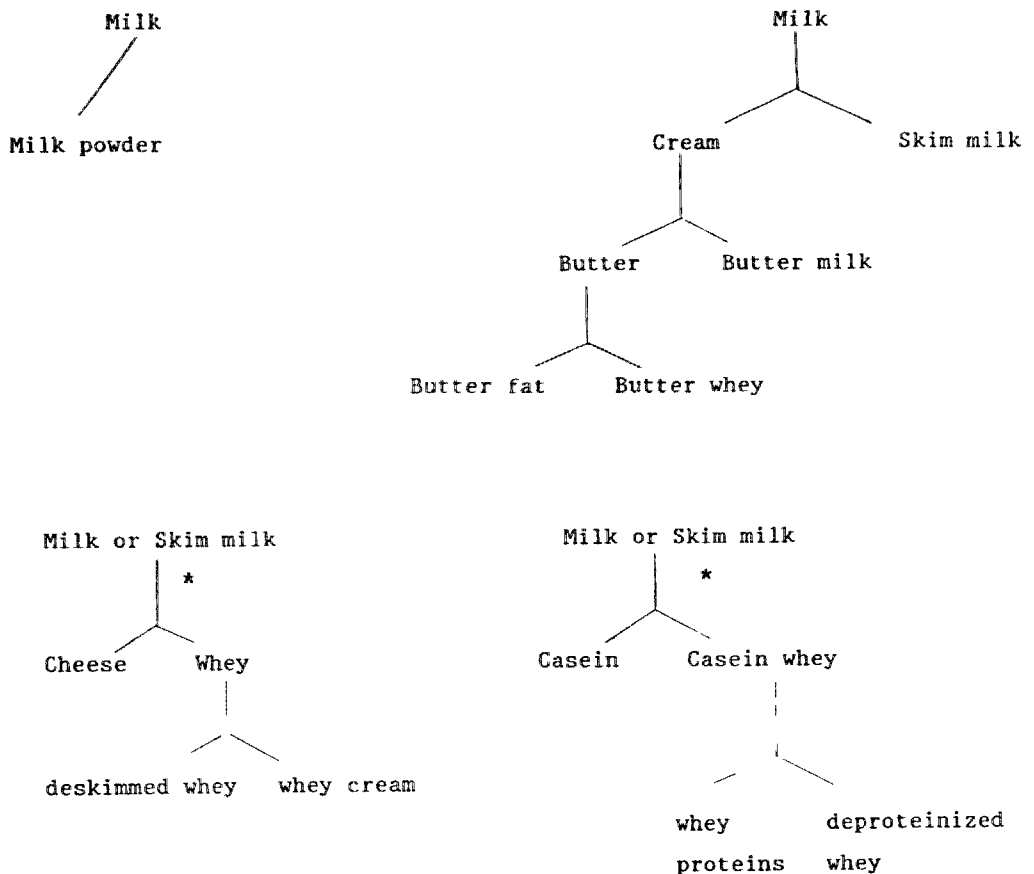


Fig. 1. Different types of food processing.



\* rennet or acid may be used to start the coagulation procedure.

Fig. 2. The fractionation of milk into products and by-products in the dairy industry.

In more generic dose assessments such as those used in planning studies where doses to hypothetical critical groups are being predicted, or in the evaluation of collective dose, regional or nationally averaged values of the loss factor may be appropriate. In one approach to collective dose evaluation it may not be necessary to identify the exposed population group but merely to assume that all food produced in a particular area is consumed. In this case an overall food processing loss factor could, in principle, be used. This is an averaged factor representing the total loss between the activity in the raw food product and that in the consumed material. However, in practice, the available information tends to be at the process level and is in the form of several separate loss factors between the raw product and consumption by man.

#### 3.1.4. Objectives and scope

It is the aim of this paper to present a survey of the information available on radionuclide behaviour during food processing. It expands on an earlier study supported by the Commission of the European Communities (CEC) [1] and takes into account new data. Most of these were presented at the Seminar on "Radioactivity Transfer during Food Processing and Culinary Preparation" organised by the Commissariat à l'Energie Atomique (CEA) and the CEC at Cadarache, France, in 1989. In addition, the paper takes into account the comments of the VAMP Terrestrial Working Group. The data presented are reported in terms of a single definition of the change in the radionuclide content of food as a result of processing. A distinction is made between normal food processing procedures and special decontamination procedures which might be of importance in emergency situations.

If not stated otherwise, all data presented in this review are based on experiments which reflect contamination in food products at a time after an accident, when contamination has been to some extent relocated and is no longer predominantly external. Thus, the data on plant contamination used in this report are generally derived from root uptake experiments, while those on food from animal origin are derived from in vivo studies. Shortly after an accident the contamination will be predominantly on the exterior parts of the product. For vegetables and fruit, separate values are available for contamination which is predominantly on external parts. These data have rather large ranges. Data which refer to the situation shortly after an accident are not available for other classes of food. However, the use of data, which refer to internal contamination, will lead,

in the case of fresh contamination, to underestimation of the decontaminating effect of food processing since newly deposited external contamination will be easier to remove than aged and relocated contamination.

### 3.2. Definitions

Several definitions are used in the literature to define changes in the radionuclide content of food due to processing. Most definitions are equal to or closely related to one of the three definitions presented here.

- (A) The change in radionuclide content due to food processing can be defined by the food processing retention factor,  $F_r$ , which is the total amount of a radionuclide in processed food divided by the total amount of this radionuclide in the original raw food (Bq processed per Bq raw). Thus,  $F_r$  is the fraction of the quantity of the radionuclide which remains in the food after processing. All data presented in this review are based on this definition.
- (B) The effect of food processing on the radionuclide content of food may also be quantified by the ratio of radionuclide concentrations, measured in fresh raw and fresh processed food. (Bq/kg fresh processed per Bq/kg fresh raw).
- (C) The third type of definition is based on a ratio of radionuclide concentrations, measured in dried raw and dried processed food. (Bq/kg processed dried per Bq/kg raw dried).

The  $F_r$  value is considered to be the best way to define changes in radionuclide content of food in this review because (a) a considerable amount of literature data is already defined in this way, and (b) conversion to  $F_r$  values appears to be possible for nearly all collected literature data which is not the case for the other definitions.

In order to avoid misinterpretations, the definition (A) is illustrated with reference to Cs and Sr. The  $F_r$  value of 0.4 for Cs boiled meat indicates that only 40% of the Cs present in raw meat is retained after boiling, 60% is removed in the boiling liquid (Table I). In the case of dairy products (Table II) the yield of each product is important. In the

TABLE I

FOOD PROCESSING RETENTION FACTOR  $F_r$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR MEAT

Underlined data denote best estimates

| Raw Material   | Method of processing                 | Sr         | Cs         | I          | Ru      | Pe   |     |         |
|--|--------------------------------------|------------|------------|------------|---------|------|-----|---------|
| Meat of mammals<br>(cow, pig<br>sheep, deer<br>rabbit) | Boiling meat                         | <u>0.5</u> | 0.4-0.9    | <u>0.4</u> | 0.2-0.7 | 0.6  | 0.3 | 0.7     |
|  | Boiling bone                         | 0.999      |            |            | 0.2-0.3 | 0.98 | 0.7 | 1.0     |
|  | Frying, roasting<br>or grilling meat | 0.8        |            |            | 0.5-0.8 |      |     | 0.4-0.7 |
|  | Mincing meat                         |            |            |            | 0.4     |      |     |         |
|  | Microwave baking                     |            |            |            | 0.4-0.5 |      |     | 0.4-0.7 |
|  | Pickling wet                         |            |            |            | 0.1-0.7 |      |     | 0.9     |
|  | dry                                  |            |            |            | 0.8     |      |     |         |
|  | Marinating                           |            |            |            | 0.1-0.6 |      |     |         |
|  | Sausage production                   |            |            |            | 0.4-1.0 |      |     |         |
| Birds  | Boiling meat                         | 0.5        |            |            |         |      |     |         |
|  | Baking meat                          |            |            |            | 0.7-0.8 |      |     |         |
| Fish   | Boiling meat                         | 0.9        | <u>0.7</u> | 0.2-0.9    |         |      |     | 0.5-0.9 |
|  | Frying meat                          |            |            | 0.8-0.9    |         |      |     | 0.7-0.8 |

References: [3, 6, 29, 42-58]

tables, the yield is presented under the "processing efficiency"  $P_e$  (kg prepared product/kg raw material). For example, an  $F_r$  value of 0.61 for Sr in goat cheese indicates that 39% of the Sr is removed by the conversion of goat milk to cheese, but, due to the 12% yield of cheese, the concentration of Sr in goat cheese is  $0.61/0.12 = 5$  times the concentration of Sr in goat milk.

It must be stressed that all  $F_r$  values referring to extraction procedures such as cooking, frying, etc., are only valid when the extraction liquid is removed and not used for other culinary purposes. The same applies to fractionation processes which take place in eg. the dairy industry or during the milling of cereals. As most fractions are used for human consumption, the use of  $F_r$  values should be supported with a thorough knowledge of the destination of the different fractions. For example, the by-product of cheese-making, whey, was formerly regarded as waste or used to feed animals. Nowadays, a large part of the produced whey is used as an additive for food for humans. Similarly, all fractions of the milling process are usually used to produce food. In these cases, an  $F_r$  value of 1.0 should be used.



TABLE II

FOOD PROCESSING RETENTION FACTOR  $F_p$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR DAIRY PRODUCTS

Underlined data denote best estimates

| Product               | Sr                        | Cs                     | I                     | $P_e$                 |
|-----------------------|---------------------------|------------------------|-----------------------|-----------------------|
| Cream                 | <u>0.07</u> 0.04-0.25     | <u>0.05</u> 0.03-0.16  | 0.06-0.19             | <u>0.08</u> 0.03-0.24 |
| Skimmilk              | <u>0.93</u> 0.75-0.96     | <u>0.95</u> 0.85-0.99  | 0.81-0.94             | <u>0.92</u> 0.76-0.97 |
| Butter                | <u>0.006</u> 0.0025-0.012 | <u>0.01</u> 0.003-0.02 | 0.035-0.01            | <u>0.04</u> 0.03-0.05 |
| Buttermilk            | <u>0.06</u> 0.03-0.07     | <u>0.05</u> 0.02-0.13  | 0.05-0.13             | <u>0.04</u> 0.03-0.14 |
| Butterfat             | 0.001-0.002               | <u>0.00</u> 0.00-0.00  | 0.02                  | <u>0.04</u> 0.04-0.04 |
| Milk powder           | 1.00                      | 1.00                   | 1.00                  | 0.12                  |
| Cheese *              |                           |                        |                       |                       |
| goat                  | 0.61                      | 0.07-0.15              | 0.08-0.14             | <u>0.12</u> 0.08-0.17 |
| cow rennet            | 0.025-0.80                | <u>0.07</u> 0.05-0.23  | <u>0.20</u> 0.11-0.53 | <u>0.12</u> 0.08-0.18 |
| cow acid              | 0.04-0.08                 | 0.11-0.12              | 0.22-0.27             | <u>0.10</u> 0.08-0.12 |
| Cottage cheese rennet | 0.07-0.17                 | 0.01-0.05              |                       |                       |
| Cottage cheese acid   | 0.22                      |                        |                       |                       |
| Yogurt                |                           | 0.34                   |                       |                       |
| Whey *                |                           |                        |                       |                       |
| rennet                | 0.20-0.80                 | 0.73-0.96              | 0.47-0.89             | <u>0.90</u> 0.70-0.94 |
| acid                  | 0.70-0.90                 | 0.75-0.90              | 0.60-0.73             | 0.82                  |
| Casein *              |                           |                        |                       |                       |
| rennet                | 0.10-0.85                 | 0.01-0.08              | 0.02-0.12             | 0.03-0.06             |
| acid                  | 0.05-0.08                 | 0.01-0.04              | 0.03-0.04             | 0.01-0.06             |
| Casein whey*          |                           |                        |                       |                       |
| rennet                | 0.08-0.16                 | 0.77-0.83              | 0.69-0.82             | <u>0.76</u> 0.73-0.79 |
| acid                  | 0.67-0.86                 | 0.83-0.84              | 0.78-0.80             | <u>0.78</u> 0.75-0.79 |
| Milk **               |                           |                        |                       |                       |
| ion exchange          | <u>0.1</u>                | <u>0.01</u>            | <u>0.1</u>            |                       |

## COMMENTS

\* Separate values are given for the rennet and acid coagulation procedures

\*\*Decontamination of milk by ion exchange on commercial scale

References: [3-5, 7-41]

### 3.3. Effects of Normal Food Processing

#### 3.3.1. Data analysis

The quality and reliability of all data on radionuclide behaviour during food processing was checked to the extent possible before they were incorporated in the survey. Firstly, it is required that experimental or test procedures should produce an artificial contamination of the food products which is comparable to the contamination which would occur following an accidental release of radionuclides. Special attention was given to speciation and to the spatial distribution of the radionuclides in food. Also, simulated processing procedures need to be a realistic reflection of the procedures normally applied to food before it is consumed. Information on modified procedures may, however, be relevant in order to know the effectiveness of a planned decontamination method. Special decontamination procedures are included in the survey only when (a) they result in a lower contamination of the processed food than in the case of normal food processing and (b) when the food retains its nutritional value and is not unacceptably changed in taste and appearance.

The data was converted to  $F_r$  values and ordered on the basis of the type of food, the method of processing and the type of radionuclide studied. The large amount of information from more than a hundred articles and reports required a grouping of the different processing modes in order to present the data in a useful format for subsequent applications. The tables present ranges of  $F_r$  values for different foods and processing techniques. Where possible, processing efficiencies  $P_e$  are also included.

#### 3.3.2. Data variation

In general, the data within each separate combination of product, procedure and radionuclide show a large variation. This variation may have different causes. Variation may be caused by differences in the yield,  $P_e$ . For example, the first row of data in Table I show the range in  $F_r$  values of Sr for cream. This range in  $F_r$  values is nearly identical to the range in  $P_e$ . Observed concentrations of Sr in cream are close to those in raw milk, with much less variation in them than the  $F_r$  values in Table I suggest. For dairy products and cereals the differences in yield are an important cause of variation. Variation in the  $F_r$  values of the other products, however, seem to be mainly caused by differences in the application of particular food preparation methods. In fact, the way in

which people prepare their food is a very important and specific part of their cultural background. Some of the most important aspects of the preparation are the temperature, the additives used, the time spent on each step in the procedure and the mechanical treatments such as removal of inedible parts, peeling, slicing, etc. Also industrial processes may vary from country to country, leading to more or less refined products. For external contamination, the time between contamination and harvest will cause variation in  $F_r$  values. In this case, climatological conditions or agricultural techniques, such as the addition of water by spraying rather than by irrigation, may also influence the  $F_r$  values.

Best estimate values of  $F_r$  and  $P_e$  are given whenever the data are adequate to permit these judgements. The most important features are that many data from different regions are available, that the range in the vast majority of the data is much smaller than the total range observed and, that extremes are not typical for certain regions. Regarding the large variation in  $P_e$  and  $F_r$ , the use of  $P_e$  values to convert  $F_r$  to concentration ratios is not recommended for extreme values. However, for best estimates or mean values the use of  $P_e$  will give a good indication of expected concentration ratios.

### 3.3.3. Dairy products

A considerable amount of data are available on the behaviour of the nuclides Sr, Cs and I during milk processing (Table II) (Figure 2). In this case  $F_r$  represents the fraction of the radionuclides which remains in the processed food. However, as under normal circumstances nearly all by-products are used for other food products - even the whey of cheese production is used nowadays for beverages and other foodstuffs - only a small amount of radioactivity will be removed from the diet by milk processing. Following an accident, however, milk processing may be modified deliberately so as to reduce intakes.

The distribution of the radionuclides over the various products is influenced by the yield of each product. For example, a higher yield of cream will lead to a proportionally higher  $F_r$  value for cream if the increase in the yield does not affect the concentration of the radionuclide in the cream.

The coagulation process appears to have a considerable effect on the transfer of Sr to cheese.  $F_r$  values for Sr are quite unpredictable in

the case of rennet coagulation, but often a large transfer to cheese is observed. Coagulation by an acidifying procedure appears to lead to a much lower Sr transfer to cheese. For all nuclides, the variation in  $F_r$  values for cheese production by acid coagulation is smaller than by rennet coagulation.

TABLE III

FOOD PROCESSING RETENTION FACTOR  $F_r$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR LOWER SEA ORGANISMS

| Organism | Method of processing            | $F_r$ values                          | $P_e$ |
|----------|---------------------------------|---------------------------------------|-------|
| crevette | cooking                         | Ra:0.04-0.5 Pb:0.0-0.4<br>Po:0.04-0.8 | 0.35  |
| mussels  | Washing and removal<br>of flesh | Ra:0.01 Pb:0.5 Po:0.02                | 0.25  |
| algues   | alginate production             | Tc:0.02 Ru, Rh 0.07 Sr:0.6            | 0.04  |
|          | satiagum production             | Ru, Rh:0.04 Co:0.04                   | 0.08  |

References: [59-61]

#### 3.3.4. Meat

Tables I and III give data on meat, fish and other marine derived food sources. Data on meat processing do not appear to be influenced by animal type. Radionuclides in fish might behave somewhat differently when compared to mammals, although too few data are available on fish processing to draw any firm conclusions.

#### 3.3.5. Vegetables and fruit

$F_r$  values for vegetables and fruit appear to depend highly on the type of product and the method of processing (Table IV). The variability

TABLE IV

FOOD PROCESSING RETENTION FACTOR  $F_p$  AND THE PROCESSING EFFICIENCY  $P_e$   
FOR VEGETABLES AND FRUIT

Data are based on total contamination of the plant

| Plant        | Method of processing           | Sr      | Cs      | Other Nuclides | $P_e$   |
|--------------|--------------------------------|---------|---------|----------------|---------|
| Spinach      | Washing                        | 0.4-1.0 | 0.6     |                | 1.0     |
|              | Washing & blanching            | 0.4-1.0 | 0.5-0.6 |                | 0.8     |
|              | Cooking & rinsing              | 0.9     | 1.0     | Co 0.9         | 0.7     |
|              | Canning                        | 0.5     | 0.2     |                | 0.7     |
|              | Freezing                       | 1.0     |         |                | 0.7     |
| Lettuce      | Removing inedible parts        |         | 0.5     |                | 0.7     |
|              | Blanching                      | 0.3-0.9 | 0.1-0.6 |                | 0.5     |
| Cabbage      | Marinating                     |         | 0.9     | Ru 0.5         | 0.9     |
|              | Washing                        | 0.3     | 0.9     |                | 1.0     |
|              | Washing & blanching            | 0.4-1.0 | 0.1-1.0 |                | 1.0     |
|              | Cooking & rinsing              | 0.8     |         |                | 0.7     |
|              | Freezing                       | 0.2-0.9 | 0.7     |                | 0.7     |
|              | Canning                        | 0.4     | 0.2     |                | 0.7     |
| Cauliflower  | Peeling                        |         | 0.5     |                | 0.7     |
| Beans        | Washing                        | 0.1     |         |                | 1.0     |
|              | Blanching                      | 0.3-1.0 | 0.6-0.9 |                | 0.9     |
|              | Canning                        | 0.3-0.8 | 0.4-1.0 |                |         |
|              | Froth flotation                | 0.4-0.6 | 1.0     |                |         |
|              | Brine grading                  | 0.6     | 1.0     |                |         |
| Tomatoes     | Washing                        | 0.7     |         |                | 1.0     |
|              | Peeling & slicing              | 0.7     |         |                | 0.9     |
|              | Canning                        | 0.8     |         |                |         |
|              | Frying                         | 0.5     |         |                | 0.5     |
| Onions       | Peeling + washing + boiling    | 0.5     |         |                |         |
| Mushrooms    | Boiling                        | 0.7-0.9 | 0.2-0.5 |                | 0.6     |
|              | Boiling in 2% NaCl             | 0.2     | 0.2     |                |         |
|              | Canning                        | 0.5     | 0.5     |                |         |
|              | Parboiling                     |         | 0.1-0.4 |                |         |
|              | Soaking of dried mushrooms     |         | 0.1-0.2 |                |         |
|              | Parboiling + salting + soaking |         | 0.00    |                |         |
| Cucumbers    | Pickling                       |         | 0.15    |                |         |
|              | Canning                        | 0.35    | 0.06    |                |         |
| Peaches      | Peeling                        | 0.5     |         |                | 0.9     |
|              | Canning                        | 0.5     |         |                |         |
|              | Lye peeling                    | 0.09    | 0.03    |                |         |
| Strawberries | Rinsing                        | 0.7     | 0.6     |                | 1.0     |
| Berries      | Making Puree                   |         | 0.6-0.8 |                | 0.6-0.8 |
|              | Rinsing                        |         | 0.8     |                |         |

References [4, 29, 61-86]

TABLE V

FOOD PROCESSING RETENTION FACTOR  $F_p$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR VEGETABLES AND FRUIT

Data are based on external contamination only

| Plant       | Method of Processing    | Sr      | Cs       | I        | Other Nuclides                     | $P_e$   |
|-------------|-------------------------|---------|----------|----------|------------------------------------|---------|
| Spinach     | Washing                 | 0.2     | 0.2-0.9  | 0.07-0.8 | Ru:0.4-0.8                         | 1.0     |
|             | Washing + blanching     | 0.4-0.7 | 0.2-0.9  | 0.6-0.7  | Ru:0.5-0.8                         | 0.8     |
|             | Cooking + rinsing       |         |          | 0.4      |                                    | 0.7     |
| Lettuce     | Washing                 |         | 0.2-1.0  | 0.1-0.5  | Ru:0.2                             | 1.0     |
|             | Removing inedible parts |         | 0.1-0.4  | 0.1-0.4  | Ru:0.01-0.3                        | 0.7     |
| Cabbage     | Removing inedible parts |         | 0.9      | 0.5      | Ru:0.7-1.0                         | 0.8     |
|             | Washing                 | 0.07    | 0.09     | 0.4      |                                    | 1.0     |
|             | Washing + blanching     | 0.3     | 0.2-0.7  |          |                                    | 0.7     |
|             | Cooking + rinsing       |         |          | 0.2-0.5  |                                    | 0.7     |
| Cauliflower | Peeling                 |         | 0.05-0.2 | 0.03     | Ru:0.02                            | 0.7     |
| Beans       | Washing                 |         |          | 0.7      |                                    | 1.0     |
|             | Blanching               | 0.3     | 0.3      | 0.2      |                                    | 0.9     |
|             | Froth flotation         | 0.4     | 0.4      |          |                                    |         |
|             | Brine grading           | 0.4     | 0.4      |          |                                    |         |
| Tomatoes    | Washing                 |         |          | 0.5      |                                    | 1.0     |
|             | Boiling                 |         |          | 0.2      |                                    | 0.7     |
| Onions      | Removing inedible parts |         | 0.2      | 0.2      | Ru:0.2                             | 0.9     |
|             | Washing                 |         | 0.3      | 0.2      |                                    | 1.0     |
| Mushrooms   | Boiling in 2% NaCl      |         | 0.3      |          |                                    |         |
| Berries     | Rinsing                 |         | 0.8-0.9  |          | Ru:0.8-1.0                         |         |
|             | Making purée            |         |          |          | Ru:0.7                             | 0.6-0.8 |
|             | Boiling                 |         | 0.3-0.5  | 0.2      | Te:0.3-0.7<br>Ba:0.6-0.9<br>Zr:1.0 |         |

References: [29, 61-86]

of the data may be a result of differences in processing time and temperature, in other specific aspects of the processing, and in the rigidity of the cellular structure of the vegetable product, etc. For example, a much more efficient removal was observed on several products for steam blanching when compared to water blanching [78]. The high Sr removal from beans by washing might be due to large amounts of soil attached to the product, but no explanation was given in the relevant report [76].

Data on externally contaminated plants are presented separately (Table V). The scatter in these data is even higher when compared to internally contaminated plants.

TABLE VI

FOOD PROCESSING RETENTION FACTOR  $F_r$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR ROOT CROPS

| Raw material | Method of processing         | Sr      | Cs       | Other radionuclides       | $P_e$   |
|--------------|------------------------------|---------|----------|---------------------------|---------|
| Potato       | Boiling with peel            | 0.9-1.0 | 0.8-0.9  | Po:0.4-0.7                | 0.9     |
|              | Peeling                      | 0.5-0.9 | 0.6-0.8  | Po:0.3-0.5 Pu, Am:0.1-1.0 | 0.8     |
|              | Peeling & boiling            | 0.7-0.8 | 0.6      |                           | 0.8     |
|              | Frying                       | 0.6     |          |                           | 0.6     |
|              | Microwave boiling unpeeled   |         | 0.8      |                           | 0.8     |
|              | Microwave boiling peeled     |         | 1.0      |                           | 0.6     |
|              | Canning                      | 0.7     | 1.0      |                           | 0.6     |
|              | Decontamination              | 0.5     | 0.05-0.2 | Ru:0.5                    |         |
| Carrot       | Scraping + washing + boiling | 0.8     |          |                           | 0.8     |
|              | Peeling                      | 0.7     | 0.5      | Pu, Am:0.4                | 0.8     |
|              | Cooking unpeeled             |         | 0.5-0.8  |                           | 0.8-0.9 |
|              | Microwave cooking unpeeled   |         | 0.7      |                           | 0.8     |
|              | Microwave cooking peeled     |         | 0.5      |                           | 0.7     |
| Beetroot     | Peeling                      | 0.8     | 0.4-0.7  | Pu, Am:0.45               | 0.8     |
|              | Cooking unpeeled             |         | 0.3-0.7  |                           | 0.9     |
|              | Cooking + peeling            |         | 0.3      |                           | 0.8     |
|              | Microwave cooking unpeeled   |         | 0.4      |                           | 0.75    |
|              | Microwave cooking peeled     |         | 0.3      |                           | 0.7     |
| Parsnip      | Peeling                      | 0.7     | 0.6      | Pu:0.3                    |         |
| Swede        | Peeling                      | 0.65    | 0.6      | Pu:0.7                    |         |

References: [29, 62, 63, 69, 72, 73, 76, 77, 82, 83, 84, 86, 87, 88]

### 3.3.6. Root crops

The effects of food processing are generally small for root crops with the exception of beetroot (Table VI). Peeling may be very efficient in removing actinides, probably due to the removal of soil particles attached to the peel combined with the extremely low uptake of actinides from the soil by plants.

### 3.3.7. Cereals

$F_r$  values for milled cereals are generally lower than 0.5 (Table VII). The values depend on the yield of the product, as observed with milk products. The conclusion which has sometimes been drawn that transfer of Cs and Sr to 'dark' wheat flour is lower than transfer to 'white flour' is not justified. Concentrations of Cs and Sr in 'dark' flour are higher than concentrations in 'white' flour, but the yield  $P_e$  of 'dark' - or second quality - flour is much lower in these experiments. Removal of actinides by milling is also high.

### 3.3.8. Drinks

The limited data in Table VIII indicate that normal treatment and processing of beverages has only modest effects on reducing the

TABLE VII  
FOOD PROCESSING RETENTION FACTOR  $F_r$  AND THE PROCESSING EFFICIENCY  $P_e$  FOR CEREALS

| Raw Material      | Method of Processing         | Sr       | Cs       | Pu, Am  | $P_e$    |
|-------------------|------------------------------|----------|----------|---------|----------|
| Wheat grain       | Milling to white flour       | 0.09-0.5 | 0.2-0.6  | 0.1-0.2 | 0.7      |
|                   | Milling to dark flour        | 0.1-0.2  | 0.05-0.1 |         | 0.05-0.1 |
|                   | Milling to semolina          |          | 0.15-0.5 |         | 0.2-0.3  |
|                   | Milling to bran              | 0.6-0.9  | 0.5-0.6  |         | 0.1-0.2  |
|                   | Cooking wheat sprouts        |          | 0.9      |         | 1.8      |
|                   | Shredding or puffing wheat   |          | 0.1-0.15 |         | 0.9-0.95 |
| Durum Wheat grain | Milling to flour             |          | 0.1-0.6  |         | 0.08-0.8 |
|                   | Milling to groat + groatdust |          | 0.3-0.4  |         | 0.6-0.7  |
|                   | Milling to bran              |          | 0.4-0.5  |         | 0.2      |
| Rye grain         | Milling to white flour       | 0.6      | 0.3-0.6  | 0.2     | 0.6-0.8  |
|                   | Milling to dark flour        |          | 0.2      |         | 0.1      |
|                   | Milling to bran              |          | 0.35-0.7 |         | 0.15-0.4 |
|                   | Cooking rye sprouts          |          | 0.8-0.9  |         | 1.9-2.4  |
| Barley grain      | Milling to white flour       | 0.5      | 0.2-0.6  | 0.1-0.2 | 0.6-0.8  |
|                   | Milling to semolina          |          | 0.35     |         | 0.1      |
|                   | Milling to bran              |          | 0.4      |         | 0.4      |
| Oats grain        | Milling to white flour       | 0.3      | 0.4      | 0.4     | 0.4      |
| Pasta             | Cooking                      |          | 0.8-0.9  |         | 2.2      |

References: [16, 29, 62, 67, 86, 89-99]



TABLE VIII

FOOD PROCESSING RETENTION FACTOR  $F_r$  AND THE PROCESSING EFFICIENCY  $P_e$   
FOR DRINKS

| Raw material  | Method of processing                   | Sr  | Cs                 | Other radionuclides       | $P_e$              |
|---------------|--|-----|--------------------|---------------------------|--------------------|
| Surface water | Conventional treatment<br>to tap water | 1.0 | 0.7                | Ru:0.3<br>Co:0.4<br>I:0.8 |                    |
| Tea           | Brewing 2 minutes<br>Brewing 8 minutes |     | 0.4<br>0.6         |                           |                    |
| Herb tea      | Brewing                                |     | 0.04-0.6           |                           |                    |
| Berries       | Pressing to juice<br>Steaming to juice |     | 0.8-0.9<br>0.2-0.6 |                           | 0.8-0.9<br>0.3-0.7 |

References: [29, 61, 68 100-104]

radionuclide content, although it is recognized that special drinking water treatments can be very effective for some elements.

### 3.4. Decontamination

#### 3.4.1. Introduction

In cases of extensive contamination of the environment with radionuclides, processes for the decontamination of foodstuffs may be considered as a countermeasure in order to reduce doses to the population. Although food processing takes place in the household and in industry, changes in household food preparation methods should only be recommended in

very extreme situations because of possible public reactions, social consequences and the lack of the possibility to control the implications of such advice. However, changes in industrial food processing methods may be feasible and lead to a lower transfer of radionuclides to the population.

In general some decrease in the transfer of radionuclides to the population may be obtained by applying selective processing to certain types of products. In some cases, optimised decontamination procedures may decrease food contamination considerably. The feasibility of changes in food processing methods will depend strongly on the product and food technology.

The success of efforts to decontaminate food is, in general, related to the rigidity of the cellular structure. Consequently, decontamination is more efficient for foods of animal origin than for vegetable products, and the results for liquids such as milk are rather successful.

#### 3.4.2. Milk

For the decontamination of milk without losses in nutritional value and deterioration of the taste, ion exchange appears to be the most promising method [14]. The  $F_r$  values presented here refer to milk decontamination on a commercial scale (Table II). Better results have been reported from research on a laboratory scale especially for caesium. Other methods, based on coprecipitation or electrodialysis led to a serious deterioration of the taste of milk. Recently, a method based on complexation of caesium by modified Prussian Blue was developed to decontaminate whey on a commercial scale [104]. It is not yet known whether this method will have adverse effects on taste, appearance or nutritional value of the milk. In the dairy industry it is also possible to shift production towards products which are contaminated to a lesser extent by radionuclides. However, consideration has to be given to the economic value of the by-products. Butter production, sometimes suggested, would lead to contaminated buttermilk, which has an important economic value. A better strategy would be the production of certain types of cheese or refined products such as caseine. The remaining whey is of less economic value and can be regarded as waste.

#### 3.4.3. Meat

Several decontamination procedures have been applied to meat, often resulting in poor decontamination combined with quite unfavourable

consequences for the taste and appearance of the meat. Certain ways of pickling or marinating may, however, result in contamination levels of Cs which are reduced by about an order of magnitude, while the taste remains good [50, 56].

#### 3.4.4. Root crops

In the case of root crops decontamination procedures may lead to an  $F_r$  value of about 0.1 or even lower for Cs [63] but the effects of decontamination are limited for  $S_r$ .

#### 3.4.5. Vegetables

For vegetables, a number of decontamination procedures have been applied, most often based on immersion in a decontamination liquid. The results are, however, not promising and consequences for taste and appearance not favourable. Unexpected effects were observed, such as a decreasing permeability of the cellular structure at higher temperature, leading to lower decontamination [88]. However, some efforts were successful; mushrooms can be decontaminated effectively by a combination of salting and cooking [75].

No important results are known on decontamination of cereals, other drinks than wine or of sea organisms.

### 3.5. Conclusions

The effects of processing on the behaviour of radionuclides depend on the radionuclide, on the type of product and on the method of processing. In general, food processing may halve the amount of radionuclides present in the food. Processing effects are rather small for root crops whereas milling cereal grains to flour will often remove about 70% of the radioactivity. Although attempts to apply decontamination procedures were often not successful, several procedures have been developed which may remove about 90% or more of the radioactivity for certain combinations of radionuclides and products. In the case of the processing of milk, meat and potatoes, available data allow a rather detailed insight into the effects of processing. The effects of processing on vegetables and fruits are rather unpredictable, especially when the radionuclides are adsorbed on the surface of the plants.

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## Chapter 4

### SEASONALITY

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#### 4.1. Introduction

##### 4.1.1. Seasonality

The impact of environmental releases of radionuclides on the human food chain depends, among other things, on the time of the year when the release occurs. The term seasonality, as used in this report, means the varying response to radioactive contamination of environmental samples according to the time of the year, when the contamination occurs. Seasonality effects may be masked, enhanced or confused by other seasonal changes and to assist in clarification a seasonality factor is introduced. The seasonality factor,  $S$ , allows seasonality to be quantified; it is defined in Annex 1. When  $S > 0$  seasonality is present and when  $S = 0$ , no seasonality is observed. Annex 1 also contains an example of the application of  $S$  to seasonality in agricultural crops.

#### Agricultural ecosystems

The most obvious effects of seasonality are seen in relation to direct deposition on the surface of crops. When the contamination has reached the ground, and indirect contamination (root uptake) becomes the dominant pathway, seasonality is usually no longer relevant. Seasonality is of special importance in connection with the contamination of cereals. In general it may be said that a deposition event during winter, when the fields lie fallow and the domestic animals are stabled, will result in a significantly lower radiological impact than if a similar contamination were to take place in the summer before harvest. Seasonality is generally of greater importance in temperate regions than in the subtropics where soils may be cultivated throughout the year.

#### Seminatural ecosystems

Natural and seminatural ecosystems e.g. forests may also show seasonality. Deciduous trees contaminated during the winter will retain

less activity than in case of a summer contamination, when the leaves will retain more of the activity from the air. Hence the forest will be most contaminated if the contamination occurs in the summer. However the effect on the forest floor will be delayed until leaf fall, which is not the case for a contamination during the winter. Coniferous trees will show less seasonality than deciduous trees. But a delay of the contamination of the forest floor will also be seen here.

#### Aquatic ecosystems

In general, seasonality has not been considered in the context of aquatic pathways. However, in the case of freshwater fish living in a frozen lake when the contamination occurs, there will be a delayed and lesser response to the contamination compared with an ice-free lake. There might also be some influence due to plankton. If it is assumed that the contamination occurs during a plankton bloom, then much of the activity may be captured and enter the food-chain of the fish. However, if the radioactive debris is not captured by plankton, it will reach the bottom sediments becoming more or less unavailable to the biota in the lake and the infinite time integral of activity in the fish will be lower than in the case where there is a plankton bloom. Contamination of drinking water derived from groundwater will show no seasonality. However, drinking water coming from surface water may (as discussed above for fish) show a seasonality, due either to plankton bloom or to ice. Radioactivity in drinking water may also be influenced by snow melting. In this case, when the snow melts, contamination of a snow-covered area may affect drinking water derived from a river receiving the meltwater. Contamination with long lived radionuclides during the summer would probably have fewer implications because the activity then would more easily be retained by the soil.

#### 4.1.2. Seasonal variability

Seasonality should not be confused with "seasonal variability". Foodproducts produced throughout the year, e.g.: milk, may show seasonal variability which occurs even though the radioactive deposition is evenly distributed throughout the year. Thus, <sup>137</sup>Cs concentrations may be seen to be significantly higher in summer milk than winter milk, if the cows are grazing in summer, but fed with beets during winter (Fig. 1). Permanent pastures may also show a seasonal variation in their radionuclide

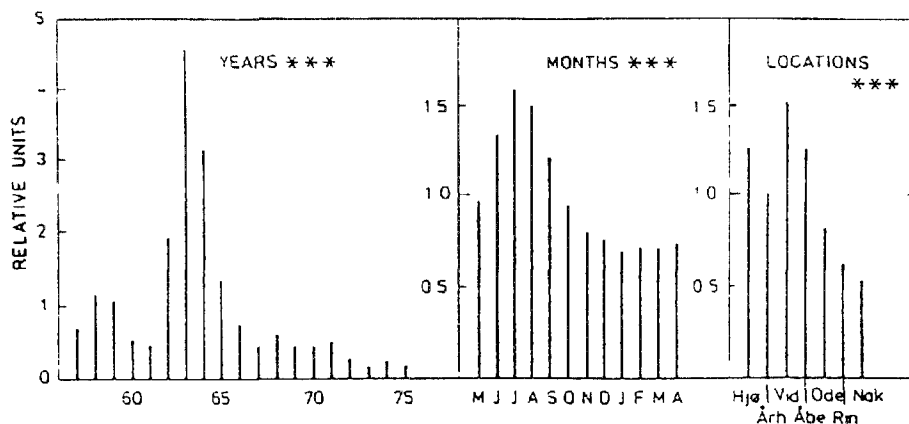


Fig. 1. The variation of pCi Cs-137 (g K)<sup>-1</sup> in Danish dried milk collected monthly at 7 dried milk factories from May 1959 to April 1976. The bars indicate the levels relative to the grand mean 18.5 pCi (g K)<sup>-1</sup> (= 1 at the relative scales) [41].

concentrations due to variations in the uptake and loss of radionuclides throughout the growing season. In the case where only seasonal variability is present with no seasonality, the seasonality factor, S is zero.

#### Agricultural practice

Agricultural practice may have seasonal aspects. Ploughing, harrowing or harvesting giving rise to resuspension may, for example, contaminate neighbouring fields. In case of an evenly distributed fallout rate throughout the year this resuspension will enhance the contamination of crops in neighbouring fields. Variations in crop contamination from this phenomenon belongs to seasonal variability. If the deposition of the activity is unevenly distributed in time and if the resuspension from treatment of neighbouring fields occurs prior to the contamination only a minor additional effect from resuspension will be seen. If however the treatment of the fields takes place after the contamination event a more pronounced additional contamination is expected. The seasonality effect is then either diminished (if contamination took place in winter season) or enhanced (if the contamination occurs in summer). This illustrates how seasonal variability may influence seasonality.

#### Seminatural ecosystems

Karlén et al.[1] have shown seasonal variability in the <sup>137</sup>Cs content of roe-deer in Sweden. A peak is observed in August–September due to a high consumption of mushrooms in these months by the roe-deer.

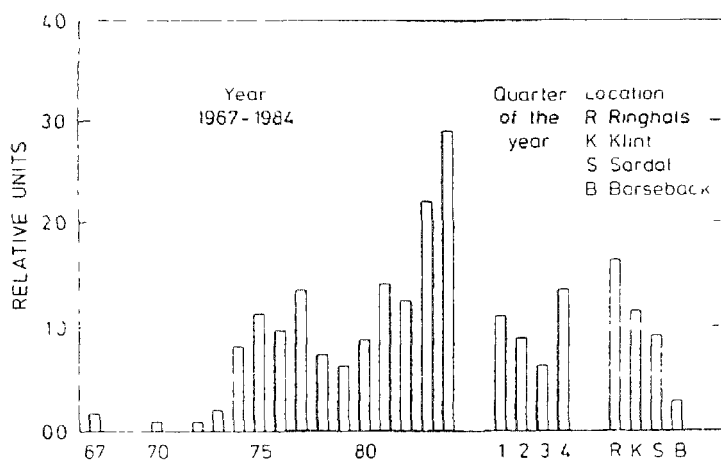


Fig. 2. Time variation of Tc-99 in Fucus in the Danish Straits 1967-1984 [42].

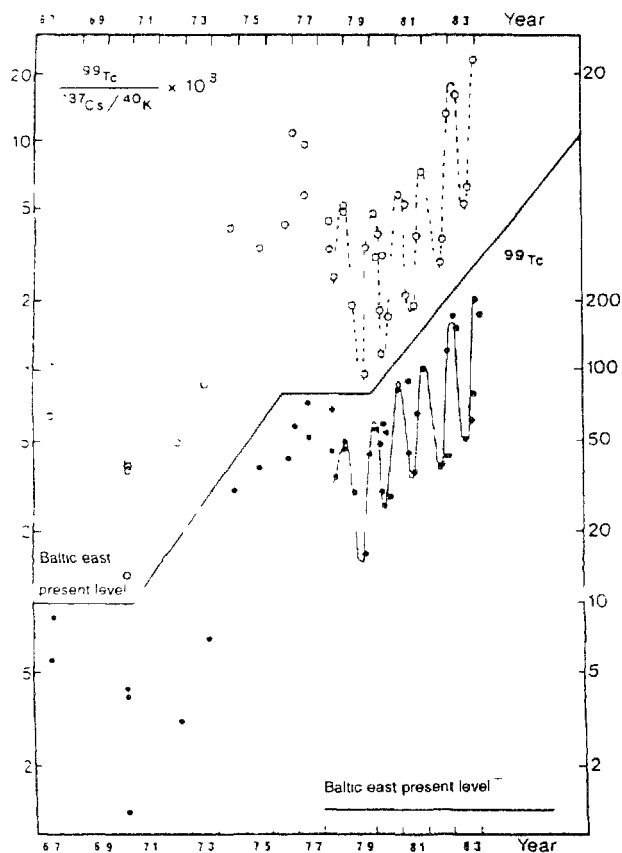


Fig. 3. Activity concentration of Tc-99 in Fucus at the west coast of Sweden (56.76°N, 12.63°E) during 1967-84. In the upper part of the figure the Tc-99/(Cs-137/K-40) ratio is given [43].

## Marine environment

Seasonal variability is also observed in the marine environment, where e.g.  $^{99}\text{Tc}$  (Fig. 2) and  $^{137}\text{Cs}$  (Fig. 3) in brown algae (*Fucus vesiculosus*) from the Baltic area show marked seasonal variations. The winter values for  $^{99}\text{Tc}$  are 2-3 times higher than in the summer, however, for  $^{137}\text{Cs}$ , the summer values are 1.5 times higher than those in the winter. These seasonal variations are primarily influenced by physiological factors, but seasonal variations in the oceanographic conditions may also play a role.

## Global fallout

Environmental contamination due to global fallout from nuclear weapons testing in the atmosphere has shown a pronounced seasonal variation with relatively low fallout rates during autumn and winter and a fallout peak in spring (Fig. 4). This seasonal pattern influences the levels in environmental samples and thus gives rise to seasonality. However, natural as well as anthropogenic factors may, as it has been shown above, superimpose on this seasonality.

## 4.2. Pre-Chernobyl Data and Models

### 4.2.1. Experimental studies

#### Middletons experiments

The first studies on seasonality were carried out by Middleton [2] in the late fifties. He studied the direct contamination of wheat, potato, cabbage and sugar beet after the application of  $^{89}\text{Sr}$  and  $^{137}\text{Cs}$  in a fine spray at different stages in the growth of the plants. He found (Tables I-III) that the extent to which  $^{137}\text{Cs}$  was concentrated in the edible tissues greatly exceeded that of  $^{89}\text{Sr}$ . Usually the concentration of  $^{89}\text{Sr}$  or  $^{137}\text{Cs}$  in each crop varied by less than an order of magnitude irrespective of the state of growth of the crop at the time of contamination. The  $^{89}\text{Sr}$  content of wheat grain, however, was considerably lower when the plants were contaminated before the ears emerged. In later experiments, Middleton and Squire [3] observed that the maximum concentrations of  $^{89}\text{Sr}$  and  $^{137}\text{Cs}$  in the cabbage hearts and  $^{137}\text{Cs}$  in potato tubers were observed after contamination occurred in the middle of the season.



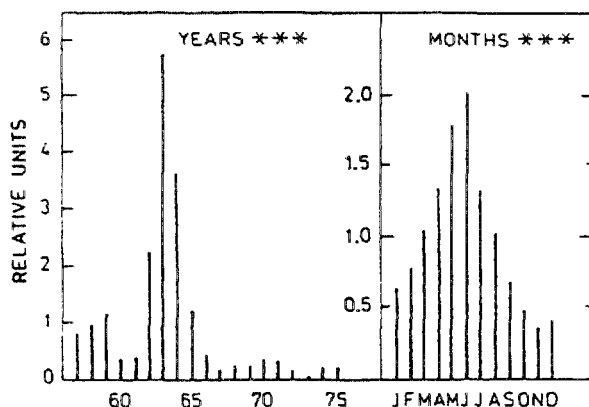


Fig. 4. The annual and monthly variation of Sr-90 in ground-level air collected at Risø 1957-1975. The bars indicate the air concentrations relative to the grand mean:  $5.31 \text{ fCi m}^{-3}$  ( $= 1$  at the relative scales) [41].

TABLE I. FOLIAR CONTAMINATION OF WHEAT WITH  $^{89}\text{Sr}$  AND  $^{137}\text{Cs}$  [2]

| Experimental data                         |  |                                 | Extrapolation to field conditions |                           |                                   |       |
|---|--|---------------------------------|-----------------------------------|---------------------------|-----------------------------------|-------|
| Days from<br>planting to<br>contamination | Content of grain<br>as percentage<br>initial retention<br>by plant | Initial retention on<br>foliage |                                   | Final content in<br>grain |                                   |       |
|   |  | $\mu\text{C}/\text{m}^2$        | $\mu\text{C}/\text{kg}$<br>dry wt | $\mu\text{C}/\text{m}^2$  | $\mu\text{C}/\text{kg}$<br>dry wt |       |
| (a) $^{89}\text{Sr}$                      |  |                                 |                                   |                           |                                   |       |
| 1957                                      | 100  | 0.04<br>1.62                    | 330                               | 1044                      | 0.14                              | 0.6   |
|   | 105  | 1.07<br>2.06                    | 259                               | 563                       | 2.88                              | 11.2  |
|   | 149  | 0.74<br>2.82                    | 273                               | 339                       | 2.00                              | 7.8   |
|   | Sig. diff.   | 0.53                            |                                   |                           |                                   |       |
| 1958                                      | 86   | 0.12<br>0.90                    | $400 \pm 64$                      | 883                       | 0.48                              | 1.5   |
|   | 107  | 1.26<br>2.08                    | $470 \pm 68$                      | 627                       | 5.92                              | 18.7  |
|   | 107†   | 0.15<br>1.15                    | —                                 | —                         | —                                 | —     |
|   | 136  | 1.79<br>2.27                    | $540 \pm 68$                      | 461                       | 9.68                              | 30.6  |
|   | Sig. diff.   | 0.27                            |                                   |                           |                                   |       |
| (b) $^{137}\text{Cs}$                     |  |                                 |                                   |                           |                                   |       |
| 1957                                      | 100  | 8.8                             | 204                               | 499                       | 17.9                              | 58.4  |
|   | 105  | 8.2                             | 301                               | 576                       | 24.6                              | 80.3  |
|   | 149  | 3.2                             | 289                               | 525                       | 9.2                               | 30.1  |
|   | Sig. diff.   | 3.3                             |                                   |                           |                                   |       |
| 1958                                      | 86   | 5.2                             | $446 \pm 84$                      | 986                       | 23.2                              | 58.8  |
|   | 107  | 10.2                            | $523 \pm 41$                      | 559                       | 53.3                              | 135.3 |
|   | 107†   | 5.4                             | —                                 | —                         | —                                 | —     |
|   | 136  | 6.8                             | $585 \pm 52$                      | 432                       | 39.7                              | 100.7 |
|   | Sig. diff.   | 2.3                             |                                   |                           |                                   |       |

† Ears protected from spray.

TABLE II. FOLIAR CONTAMINATION OF CABBAGE WITH  $^{89}\text{Sr}$  AND  $^{137}\text{Cs}$  [2]

| Experimental data                   |            |   | Extrapolation to field conditions |                                 |                          |                                 |
|-------------------------------------|------------|---|-----------------------------------|---------------------------------|--------------------------|---------------------------------|
| Days from planting to contamination |            | Content of heart as percentage initial retention by plant | Initial retention on foliage      |                                 | Final content in heart   |                                 |
|                                     |            |   | $\mu\text{C}/\text{m}^2$          | $\mu\text{C}/\text{kg}$ dry wt. | $\mu\text{C}/\text{m}^2$ | $\mu\text{C}/\text{kg}$ dry wt. |
| (a) $^{89}\text{Sr}$                |            |   |                                   |                                 |                          |                                 |
| 1957                                | 32         | 0.33  | 81                                | 3,000                           | 0.27                     | 0.84                            |
|                                     | 88         | 1.04  | 86                                | 688                             | 0.89                     | 2.82                            |
|                                     | Sig. diff. | N.S.  |                                   |                                 |                          |                                 |
| 1958                                | 58         | 0.18  | $85 \pm 9$                        | 2,180                           | 0.15                     | 0.27                            |
|                                     | 72         | 0.34  | $292 \pm 26$                      | 3,073                           | 0.99                     | 1.83                            |
|                                     | 87         | 0.16  | $309 \pm 40$                      | 1,119                           | 0.50                     | 0.91                            |
|                                     | 99         | 0.11  | $231 \pm 16$                      | 1,227                           | 0.31                     | 0.56                            |
|                                     | Sig. diff. | 0.15  |                                   |                                 |                          |                                 |
| (b) $^{137}\text{Cs}$               |            |   |                                   |                                 |                          |                                 |
| 1957                                | 32         | 0.91  | 42                                | 1,720                           | 0.4                      | 1.3                             |
|                                     | 88         | 1.51  | 37                                | 617                             | 1.3                      | 4.3                             |
|                                     | Sig. diff. | N.S.  |                                   |                                 |                          |                                 |
| 1958                                | 38         | 1.15  | $141 \pm 26$                      | 2,830                           | 1.6                      | 3.6                             |
|                                     |            | 1.02  |                                   |                                 |                          |                                 |
|                                     | 72         | 6.93  | $281 \pm 20$                      | 2,754                           | 19.5                     | 43.4                            |
|                                     |            | 1.80  |                                   |                                 |                          |                                 |
|                                     | 37         | 4.92  | $309 \pm 36$                      | 1,240                           | 15.2                     | 33.7                            |
|                                     |            | 1.62  |                                   |                                 |                          |                                 |
|                                     | 99         | 3.92  | $280 \pm 44$                      | 1,143                           | 10.7                     | 23.8                            |
|                                     | 1.67       |   |                                   |                                 |                          |                                 |
|                                     | Sig. diff. | 0.28  |                                   |                                 |                          |                                 |

The final concentration in the heart as a percentage of the amount initially retained by the plant was experimentally determined; the values for initial retention and final content of the heart under field conditions after a deposition of 1 millicurie per square metre have been calculated.

Final sampling 120 and 113 days after planting in 1957 and 1958 respectively.

Where a logarithmic transformation was necessary for statistical analysis the transformed values are in italics. Significant differences are shown at the 5 per cent level.

### Aarkrog's grain studies

In the sixties and early seventies, Aarkrog [4-7] studied the direct contamination of cereal grain (rye, barley, wheat and oats) applying radioisotopes of Sr, Cs, Mn, Ce, Cr, Fe, Co, Zn, Hg, Pb, Sb, Ru, Be, Na, Cd and Ba as a fine spray during different stages in the growth of the crops. The results of these studies with respect to seasonality were summarized in a number of heuristic models [8] (Figs. 5 and 6).

It appears that two important factors influence the contamination of grain: initial retention and translocation from the vegetative parts of the plants to the seeds. Initial retention is fairly independent of which radionuclide is involved, whereas translocation depends strongly on the radioelement.

TABLE III. FOLIAR CONTAMINATION OF POTATO WITH  $^{89}\text{Sr}$  AND  $^{137}\text{Cs}$  [2]

| Experimental data                         |   |                              | Extrapolation to field conditions |                            |                                   |       |
|---|---|------------------------------|-----------------------------------|----------------------------|-----------------------------------|-------|
| Days from<br>planting to<br>contamination | Content of tuber as<br>percentage initial<br>retention on plant | Initial retention<br>foliage |                                   | Final content in<br>tubers |                                   |       |
|   |   | $\mu\text{C}/\text{m}^2$     | $\mu\text{C}/\text{kg}$<br>dry wt | $\mu\text{C}/\text{m}^2$   | $\mu\text{C}/\text{kg}$<br>dry wt |       |
| (a) $^{89}\text{Sr}$                      |   |                              |                                   |                            |                                   |       |
| 1957                                      | 55  | 0.046                        | 480                               | 9,600                      | 0.22                              | 0.094 |
|   | 69  | 0.038                        | 650                               | 8,666                      | 0.25                              | 0.11  |
|   | Sig. diff.  | N.S.                         |                                   |                            |                                   |       |
| 1958                                      | 47  | 0.14                         | 270                               | 2,570                      | 0.38                              | 0.13  |
|   | 74  | 0.03                         | 320                               | 2,602                      | 0.096                             | 0.034 |
|   | 97  | 0.02                         | 580                               | 4,461                      | 0.12                              | 0.041 |
|   | Sig. diff.  | N.S.                         |                                   |                            |                                   |       |
| (b) $^{137}\text{Cs}$                     |   |                              |                                   |                            |                                   |       |
| 1957                                      | 55  | 25                           | 480                               | 6,760                      | 120                               | 49    |
|   | 69  | 29                           | 520                               | 6,753                      | 151                               | 62    |
|   | Sig. diff.  | N.S.                         |                                   |                            |                                   |       |
| 1958                                      | 47  | 45                           | 240                               | 2,400                      | 108                               | 32    |
|   |   | 1.05                         |                                   |                            |                                   |       |
|   | 74  | 14                           | 430                               | 3,435                      | 63                                | 19    |
|   |   | 1.09                         |                                   |                            |                                   |       |
|   | 97  | 21                           | 390                               | 4,193                      | 32                                | 24    |
|   |   | 1.22                         |                                   |                            |                                   |       |
|   | Sig. diff.  | 0.28                         |                                   |                            |                                   |       |

The final concentration in the tuber as a percentage of the amount initially retained by the plant was experimentally determined. The values for initial retention and final content of the tuber under field conditions after a deposition of 1 microcurie per square metre have been calculated.

Final sampling 125 and 129 days after planting in 1957 and 1958 respectively.

Where a logarithmic transformation was necessary for statistical analysis the transformed values are shown in *italics*. Significant differences are shown at the 5 per cent level.

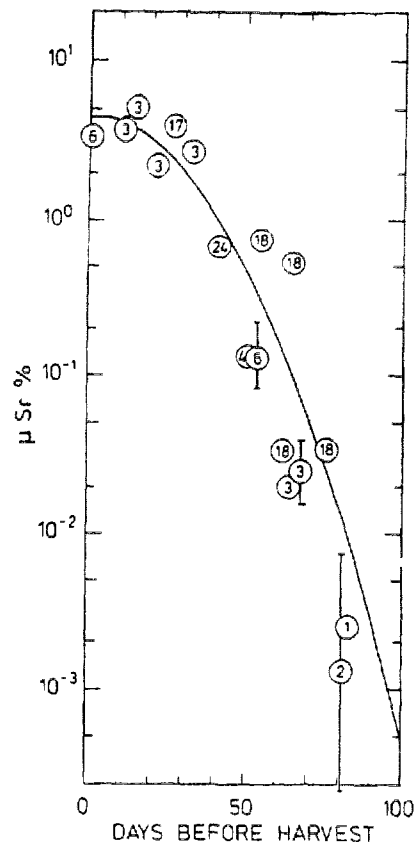


Fig. 5. Percentage of radiostrontium, applied at various dates to 1 m<sup>2</sup> barley field, recovered per kg of mature grain at harvest assuming no radioactive decay. Number of determinations and 1 S.E. are shown (unless S.E. is less than the radius of the circle). Curve calculated from  $\mu(t) = 4.5 \times 10^{-2} e^{-0.00095(t-2)^2}$  [8].

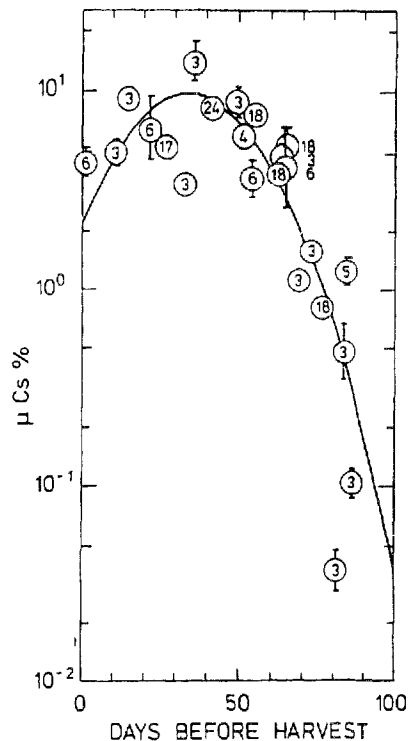


Fig. 6. Percentage of radiocaesium, applied at various dates to 1 m<sup>2</sup> barley field, recovered per kg of mature grain at harvest assuming no radioactive decay. (Number of determinations and 1 S.E. are shown.) Curve calculated from  $\mu(t) = 9.8 \times 10^{-2} e^{-0.0013(t-34)^2}$  [8].

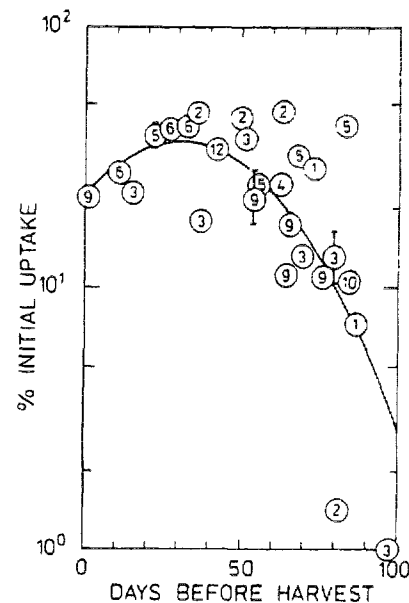


Fig. 7. Percentage of initial uptake in whole crops (above-ground parts) when a barley field at various dates is contaminated by radionuclides. The total crop yield at harvests in 0.8 kg dry matter m<sup>-2</sup>; data are based upon measurements with radioisotopes of Sr, Cs, Co, Mn, Sb, Zn and Fe; number of determinations and 1 S.E. are indicated. Curve calculated from % initial uptake =  $36 e^{-0.00052(t-30)^2}$  [8].

The percentage of initial uptake 3 months before harvest is about 5% of the activity deposited over a field with barley crops, and 1 month before harvest it reaches a maximum of 36% (Fig. 7). If the field is contaminated with, for example  $^{137}\text{Cs}$ , 3 months before harvest (1 kBq  $\text{m}^{-2}$ ), the mature grain contains 2 Bq  $^{137}\text{Cs kg}^{-1}$ ; if the contamination occurs 1 month before harvest, 100 Bq  $^{137}\text{Cs kg}^{-1}$  is found in the mature grain (Fig. 6). In the case of  $^{90}\text{Sr}$ , the corresponding concentrations would have been 0 Bq  $\text{kg}^{-1}$  and 20 Bq  $\text{kg}^{-1}$ , respectively, demonstrating that  $^{137}\text{Cs}$  is translocated to a much greater extent than  $^{90}\text{Sr}$  (Fig. 5).

The highest levels in grain are expected when the contamination takes place in the final month before harvest. The lowest levels will be seen if the fields are contaminated before sprouting.

#### Delmass' fruit studies

Delmas et al. [9] have studied experimentally the contamination of orange fruits with  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . For the same initial direct contamination with the two radionuclides during the flowering of the orange trees, the  $^{137}\text{Cs}$  contamination of the fruit flesh was an order of magnitude higher than that of  $^{90}\text{Sr}$ . The difference was due to the higher translocation of  $^{137}\text{Cs}$  than of  $^{90}\text{Sr}$  to the flesh. If the contamination were to have occurred just prior to the orange harvest, the difference would have been far less.

#### 4.2.2. Models

At a CEC seminar in Dublin in 1983 on the transfer of radioactive materials in the terrestrial environment subsequent to an accidental release to the atmosphere, a number of papers dealt with seasonality viz [10-14]. The consequences of seasonality were illustrated by model calculations.

#### Simmonds' model

The paper by Simmonds [13] concluded that for green vegetables and meat from sheep, season of the year has little effect, but for grain (Table IV) and milk (Fig. 8) or meat from cattle there can be large variations in concentration. Such variations are greater for short-lived radionuclides such as  $^{131}\text{I}$  than for the longer-lived  $^{137}\text{Cs}$  (Fig. 9) and  $^{90}\text{Sr}$ . The variations in the total activity in the diet following

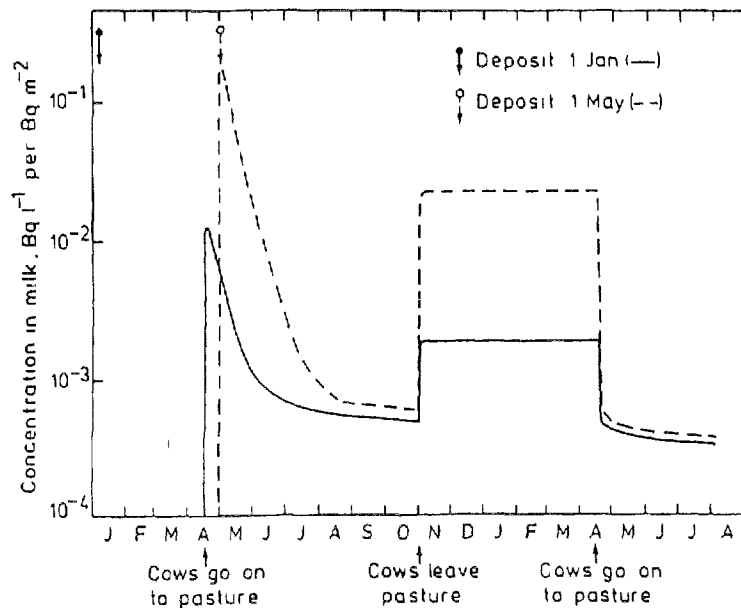


Fig. 8. The concentration of Cs-137 in milk as a function of time following an accidental release at two different times of year [13].

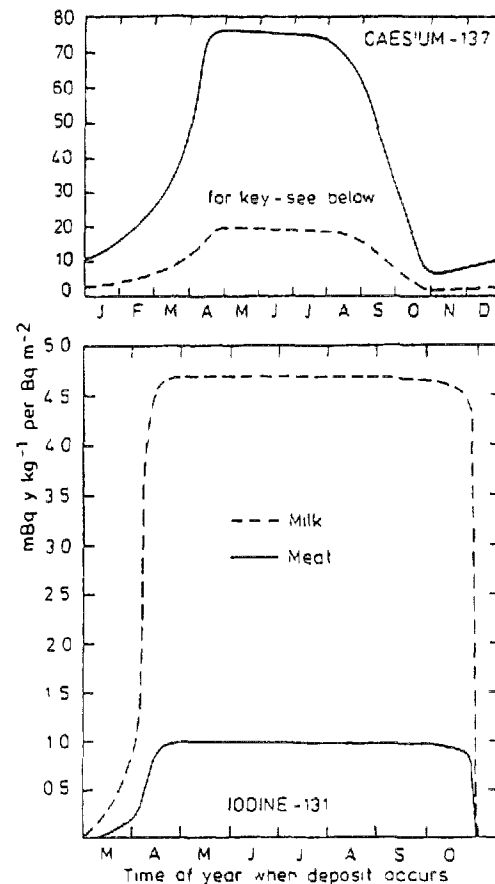


Fig. 9. Integrated concentrations in milk and meat following a single deposit of Cs-137 and I-131 at various times of the year [13].

TABLE IV. TIME INTEGRAL OF CONCENTRATION IN GRAIN PRODUCTS FOLLOWING A SINGLE DEPOSIT AT VARIOUS TIMES BEFORE HARVEST [13]

| Time of deposit before harvest<br>Nuclide | Time integral of concentration <sup>(a)</sup><br>(Bq y kg <sup>-1</sup> per Bq m <sup>-2</sup> ) |                     |                     |                     |                       |
|---|--|---------------------|---------------------|---------------------|-----------------------|
|   | 7d   | 30d                 | 60d                 | 90d                 | winter <sup>(b)</sup> |
| Sr-90                                     | $1.7 \cdot 10^{-2}$  | $1.6 \cdot 10^{-2}$ | $4.5 \cdot 10^{-3}$ | $2.0 \cdot 10^{-3}$ | $1.4 \cdot 10^{-3}$   |
| Ru-106                                    | $1.7 \cdot 10^{-3}$  | $6.8 \cdot 10^{-4}$ | $3.0 \cdot 10^{-4}$ | $2.1 \cdot 10^{-4}$ | $1.2 \cdot 10^{-4}$   |
| I-131                                     | $4.8 \cdot 10^{-4}$  | $2.5 \cdot 10^{-4}$ | $1.1 \cdot 10^{-5}$ | $2.8 \cdot 10^{-7}$ | 0.0                   |
| Cs-134                                    | $2.3 \cdot 10^{-2}$  | $8.5 \cdot 10^{-2}$ | $4.8 \cdot 10^{-2}$ | $1.6 \cdot 10^{-2}$ | $3.1 \cdot 10^{-5}$   |
| Cs-137                                    | $2.8 \cdot 10^{-2}$  | $1.0 \cdot 10^{-1}$ | $5.9 \cdot 10^{-2}$ | $2.0 \cdot 10^{-2}$ | $4.4 \cdot 10^{-4}$   |

Notes

- The integral of concentration in processed grain, integrated over all time.
- Deposition occurs when the fields are fallow before ploughing and seeding.

TABLE V. TOTAL INTAKES VIA FOOD FOLLOWING A SINGLE DEPOSIT AT VARIOUS TIMES OF THE YEAR [13]

| Time of deposit<br>Nuclide | Total intake via food <sup>(a)</sup> (Bq per Bq m <sup>-2</sup> ) |       |        |         |
|----------------------------|---|-------|--------|---------|
|                            | January   | April | August | October |
| I-131                      | 0.17  | 0.42  | 1.7    | 1.6     |
| Cs-137                     | 4.8   | 9.9   | 27     | 7.9     |
| Sr-90                      | 4.9   | 5.3   | 7.5    | 5.0     |

Note

- The total intake of activity from all food at a critical group intake rate integrated over all time

deposits at different times of the year are generally smaller than for individual foods, but are again greater for <sup>131</sup>I than for longer-lived radionuclides (Table V).

### Müller's model

Müller et al. [10] shows in a model calculation (Fig. 10) that a contamination with <sup>137</sup>Cs in May (in Germany) results in an integrated intake from food which is about 60% of that seen if a similar contamination were to take place in June, but 5 times higher than if the contamination were to occur in winter (November-March).

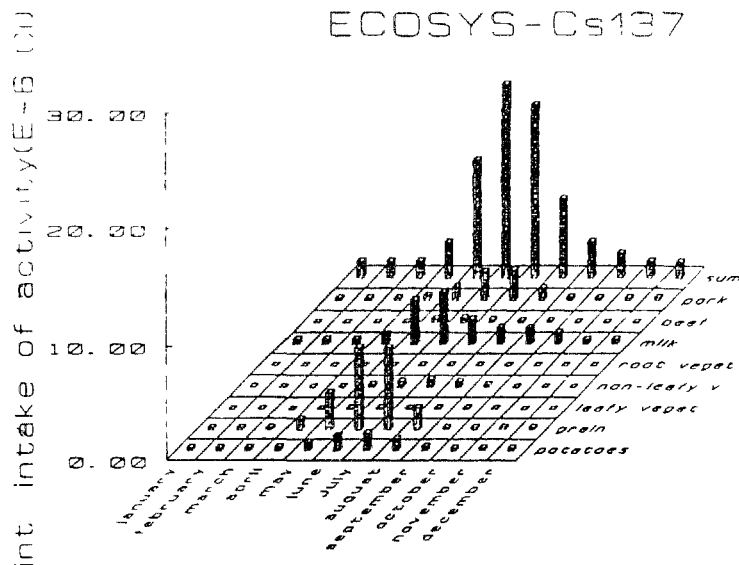


Fig. 10. Integral intake of activity by man over 50 years after deposition of  $1 \text{ Ci/km}^2$  of Cs-137 as function of month of release. The total intake ("sum") is split up into the different foodstuffs [10].

#### AGRID

In a Finnish model study [15] it was found that the expected values of collective 30-year doses in the case of dormant season and growing season seem to differ by a factor on the order of 10. In the first-year doses, the difference would be larger, a factor of about 100. The Finnish AGRID model includes seasonal effects by considering growing season and dormant season separately. But the model makes no seasonal distinctions within the growing season. Table VI shows the variation in the doses according to the AGRID model.

#### PATHWAY

The PATHWAY model [16-18] was developed to estimate radionuclide ingestion by humans exposed to fallout originating at the Nevada Test Site (NTS) during the 1950s. As NTS is an arid region PATHWAY does not consider wet deposition. The model considers seasonal changes in the biomass of vegetation and animal diets, as well as specific plowing and crop harvest dates; thus the integrated radionuclide intakes by humans are dependent on the seasonal timing of deposition. The model does not consider translocation. Table VII shows the PATHWAY model calculations for selected



TABLE VI CONDITIONAL EXPECTED VALUES OF THE  
COLLECTIVE EFFECTIVE DOSES (man Sv) DUE TO  
THE FK-2 RELEASE CATEGORY [15]

|                    | Release during        |                       | On an average<br>during the<br>whole year |
|--------------------|-----------------------|-----------------------|---|
|                    | The growing<br>season | The dormant<br>season |   |
| Plume $\gamma$     |                       |                       | $1.7 \cdot 10^2$                          |
| Fallout $\gamma$   |                       |                       | $1.8 \cdot 10^1$                          |
| Inhalation         |                       |                       | $2.1 \cdot 10^2$                          |
| Milk               | $9.8 \cdot 10^3$      | $8.0 \cdot 10^2$      | $3.3 \cdot 10^3$                          |
| Meat               | $2.6 \cdot 10^3$      | $4.7 \cdot 10^2$      | $1.1 \cdot 10^3$                          |
| Vegetables         | $1.0 \cdot 10^3$      | $5.8 \cdot 10^1$      | $2.1 \cdot 10^2$                          |
| Grain              | $4.0 \cdot 10^3$      | $1.8 \cdot 10^2$      | $8.1 \cdot 10^2$                          |
| Roots              | $5.0 \cdot 10^1$      | $4.1 \cdot 10^1$      | $4.3 \cdot 10^1$                          |
| Nutrition<br>total | $1.7 \cdot 10^4$      | $1.5 \cdot 10^3$      | $[5.4 \cdot 10^3]$                        |
| Total              |                       |                       | $7.5 \cdot 10^3$                          |

TABLE VII INTEGRATED INTAKE BY ADULT MALES<sup>a</sup> OF VARIOUS RADIONUCLIDES FROM  
ALL FOODS PER UNIT FALLOUT DEPOSITION VERSUS SELECTED FALLOUT DATES [17]  
(units are Bq per Bq m<sup>-2</sup>)

| Radio-<br>nuclide | 1 Mar<br>(Tesla)     | 17 Mar<br>(Annie)    | 25 Apr<br>(Simon)    | 19 May<br>(Harry)    | 24 Jun<br>(Priscilla) | 24 Jul<br>(Kepler)   | 31 Aug<br>(Smoky)    | 7 Oct<br>(Morgan)    |
|-------------------|----------------------|----------------------|----------------------|----------------------|-----------------------|----------------------|----------------------|----------------------|
| <sup>140</sup> Ba | $1.6 \times 10^{-2}$ | $1.6 \times 10^{-2}$ | $2.2 \times 10^{-2}$ | $2.6 \times 10^{-2}$ | $4.5 \times 10^{-2}$  | $2.7 \times 10^{-2}$ | $1.5 \times 10^{-2}$ | $1.0 \times 10^{-2}$ |
| <sup>143</sup> Ce | $1.1 \times 10^{-3}$ | $4.2 \times 10^{-4}$ | $1.0 \times 10^{-3}$ | $1.5 \times 10^{-3}$ | $2.4 \times 10^{-3}$  | $2.4 \times 10^{-3}$ | $1.2 \times 10^{-3}$ | $6.2 \times 10^{-4}$ |
| <sup>144</sup> Ce | $3.7 \times 10^{-2}$ | $5.5 \times 10^{-2}$ | $1.5 \times 10^{-1}$ | $2.0 \times 10^{-1}$ | $5.3 \times 10^{-1}$  | $5.5 \times 10^{-2}$ | $4.2 \times 10^{-2}$ | $4.3 \times 10^{-2}$ |
| <sup>136</sup> Cs | $1.0 \times 10^{-1}$ | $1.1 \times 10^{-1}$ | $1.2 \times 10^{-1}$ | $1.4 \times 10^{-1}$ | $1.9 \times 10^{-1}$  | $1.6 \times 10^{-1}$ | $1.1 \times 10^{-1}$ | $1.2 \times 10^{-1}$ |
| <sup>137</sup> Cs | 2.0                  | 2.0                  | 3.0                  | 2.6                  | 3.3                   | 1.4                  | 1.6                  | 2.4                  |
| <sup>131</sup> I  | $2.1 \times 10^{-2}$ | $2.0 \times 10^{-2}$ | $3.3 \times 10^{-2}$ | $4.9 \times 10^{-2}$ | $9.1 \times 10^{-2}$  | $1.0 \times 10^{-1}$ | $5.5 \times 10^{-2}$ | $3.7 \times 10^{-2}$ |
| <sup>133</sup> I  | $4.2 \times 10^{-4}$ | $1.5 \times 10^{-4}$ | $3.7 \times 10^{-4}$ | $1.1 \times 10^{-3}$ | $2.0 \times 10^{-3}$  | $2.9 \times 10^{-3}$ | $1.5 \times 10^{-3}$ | $7.5 \times 10^{-4}$ |
| <sup>135</sup> I  | $7.8 \times 10^{-6}$ | $2.6 \times 10^{-6}$ | $6.9 \times 10^{-6}$ | $1.5 \times 10^{-5}$ | $2.6 \times 10^{-5}$  | $3.4 \times 10^{-5}$ | $1.8 \times 10^{-5}$ | $8.7 \times 10^{-6}$ |
| <sup>99</sup> Mo  | $3.7 \times 10^{-3}$ | $1.9 \times 10^{-3}$ | $3.9 \times 10^{-3}$ | $5.5 \times 10^{-3}$ | $9.2 \times 10^{-3}$  | $1.0 \times 10^{-2}$ | $5.3 \times 10^{-3}$ | $3.0 \times 10^{-3}$ |
| <sup>147</sup> Nd | $1.3 \times 10^{-2}$ | $1.3 \times 10^{-2}$ | $1.8 \times 10^{-2}$ | $2.0 \times 10^{-2}$ | $3.4 \times 10^{-2}$  | $2.0 \times 10^{-2}$ | $1.2 \times 10^{-2}$ | $7.3 \times 10^{-3}$ |
| <sup>239</sup> Np | $2.7 \times 10^{-3}$ | $1.1 \times 10^{-3}$ | $2.6 \times 10^{-3}$ | $3.4 \times 10^{-3}$ | $5.4 \times 10^{-3}$  | $5.2 \times 10^{-3}$ | $2.7 \times 10^{-2}$ | $1.4 \times 10^{-3}$ |
| <sup>239</sup> Pu | $4.0 \times 10^{-2}$ | $6.0 \times 10^{-2}$ | $2.1 \times 10^{-1}$ | $2.9 \times 10^{-1}$ | $8.1 \times 10^{-1}$  | $6.0 \times 10^{-2}$ | $4.7 \times 10^{-2}$ | $4.9 \times 10^{-2}$ |
| <sup>105</sup> Rh | $1.3 \times 10^{-3}$ | $4.8 \times 10^{-4}$ | $1.2 \times 10^{-3}$ | $1.7 \times 10^{-3}$ | $2.7 \times 10^{-3}$  | $2.8 \times 10^{-3}$ | $1.4 \times 10^{-3}$ | $7.2 \times 10^{-4}$ |
| <sup>103</sup> Ru | $6.1 \times 10^{-2}$ | $7.4 \times 10^{-2}$ | $8.2 \times 10^{-2}$ | $9.0 \times 10^{-2}$ | $1.6 \times 10^{-1}$  | $5.9 \times 10^{-2}$ | $5.2 \times 10^{-2}$ | $6.9 \times 10^{-2}$ |
| <sup>106</sup> Ru | $1.5 \times 10^{-1}$ | $1.8 \times 10^{-1}$ | $2.6 \times 10^{-1}$ | $3.1 \times 10^{-1}$ | $6.8 \times 10^{-1}$  | $1.4 \times 10^{-1}$ | $1.6 \times 10^{-1}$ | $2.3 \times 10^{-1}$ |
| <sup>89</sup> Sr  | $5.8 \times 10^{-2}$ | $6.6 \times 10^{-2}$ | $1.1 \times 10^{-1}$ | $1.3 \times 10^{-1}$ | $2.4 \times 10^{-1}$  | $1.0 \times 10^{-1}$ | $6.5 \times 10^{-2}$ | $6.1 \times 10^{-2}$ |
| <sup>90</sup> Sr  | $2.5 \times 10^{-1}$ | $2.4 \times 10^{-1}$ | $6.0 \times 10^{-1}$ | $6.0 \times 10^{-1}$ | 1.1                   | $2.0 \times 10^{-1}$ | $1.6 \times 10^{-1}$ | $1.8 \times 10^{-1}$ |
| <sup>91</sup> Sr  | $4.5 \times 10^{-5}$ | $1.6 \times 10^{-5}$ | $4.0 \times 10^{-5}$ | $6.9 \times 10^{-5}$ | $1.1 \times 10^{-4}$  | $1.2 \times 10^{-4}$ | $6.0 \times 10^{-5}$ | $3.0 \times 10^{-5}$ |
| <sup>132</sup> Te | $4.5 \times 10^{-3}$ | $2.4 \times 10^{-3}$ | $4.6 \times 10^{-3}$ | $5.7 \times 10^{-3}$ | $9.0 \times 10^{-3}$  | $8.4 \times 10^{-3}$ | $4.5 \times 10^{-3}$ | $2.6 \times 10^{-3}$ |
| <sup>97</sup> Zr  | $2.6 \times 10^{-4}$ | $9.1 \times 10^{-5}$ | $2.3 \times 10^{-4}$ | $3.7 \times 10^{-4}$ | $5.8 \times 10^{-4}$  | $6.0 \times 10^{-4}$ | $3.0 \times 10^{-4}$ | $1.5 \times 10^{-4}$ |

<sup>a</sup> Values for adult females average 35% less than males

fallout dates. The  $^{137}\text{Cs}$  intake varies e.g. by a factor of 2.3 in the period March–October, while  $^{90}\text{Sr}$  varies by a factor of nearly 7.

#### 4.3. Post-Chernobyl Data and Models

##### 4.3.1. Observations

In general, the Chernobyl accident confirmed qualitatively the assumptions on seasonality made prior to the accident.

European  $^{131}\text{I}$  and  $^{137}\text{Cs}$  data

In the case of milk (Fig. 11), it was thus seen, as expected, that the radioiodine concentrations were low in northern Europe where the grazing season was just at its beginning, while they were significantly higher in southern Europe where the cows had been grazing for months. This latitudinal effect, which is due to seasonality, was also seen for  $^{137}\text{Cs}$  as shown by UNSCEAR [19]. The effect was most evident for grain (Fig. 12) as was anticipated.

In Denmark (Fig. 13) the relative  $^{137}\text{Cs}$  concentrations of the grain species (rye, barley, wheat, oats) were compared with those observed in the years prior to the Chernobyl accident. It appeared that the rye levels were an order of magnitude higher than those seen in the other species after the Chernobyl accident. This observation reflects

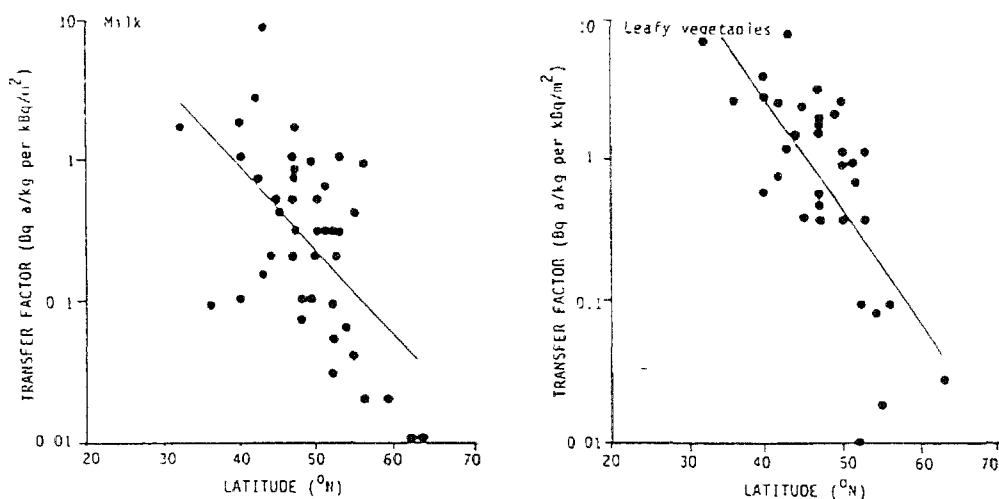


Fig. 11. Integrated concentrations of I-131 in milk and leafy vegetables per unit I-131 deposition density [19].

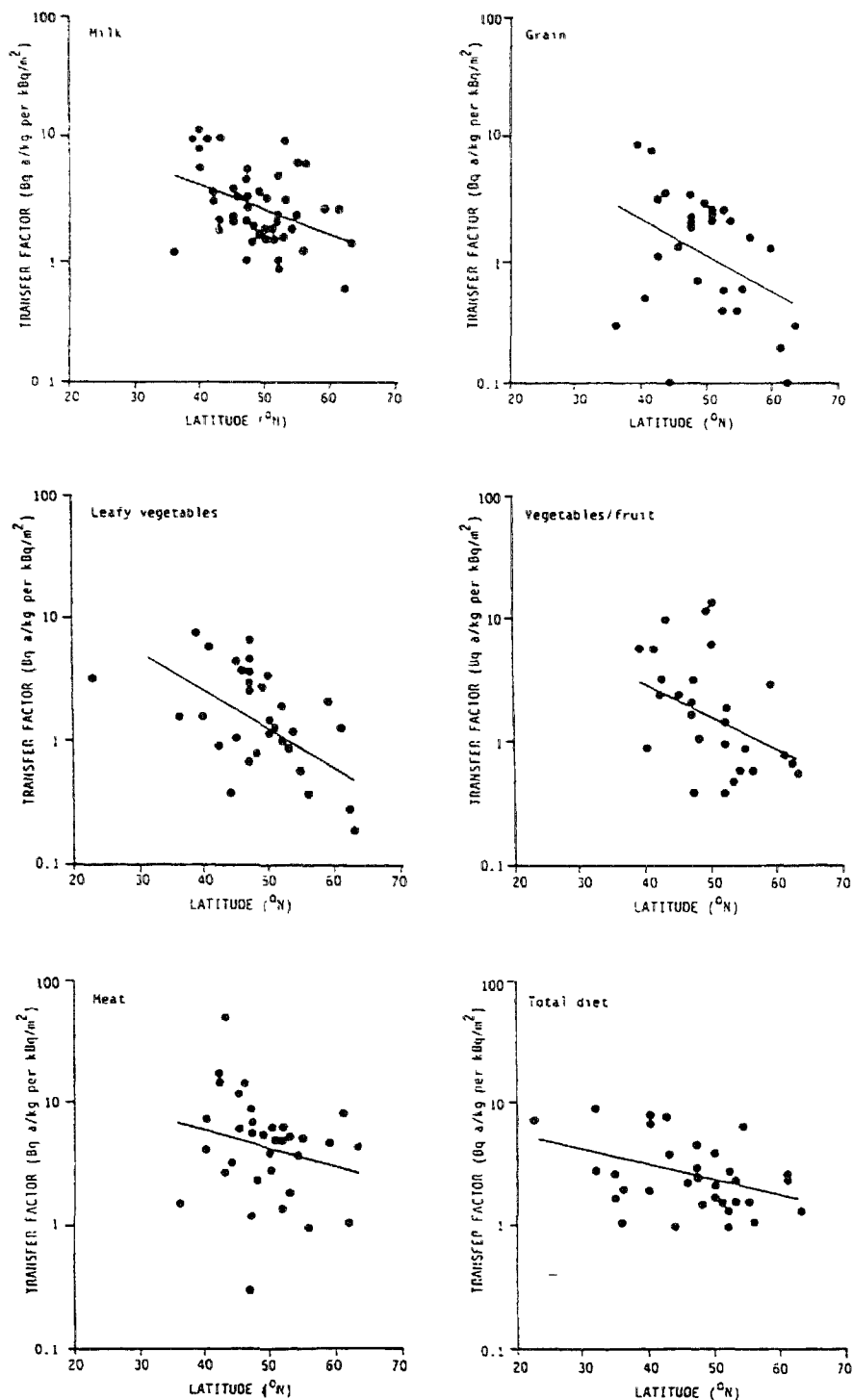


Fig. 12. Integrated concentrations of Cs-137 in foods and total diet in the first year after the accident per unit Cs-137 deposition density [19].

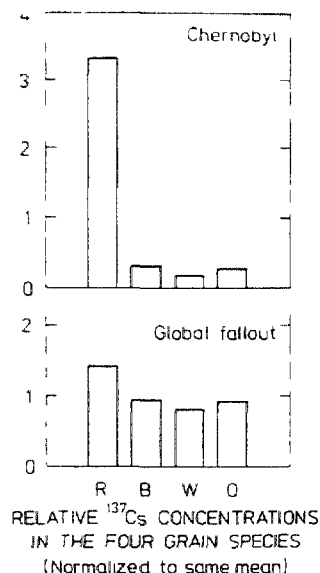


Fig. 13. Relative Cs-137 concentrations in Danish grain, normalized to a mean of 1. The global fallout samples are based on observations in the period 1962-1974. The mean concentration in Danish grain in 1962-1974 was  $7 \times 1 \text{ Bq Cs-137 kg}^{-1}$ . In 1986 the mean level was  $3 \times 3 \text{ Bq Cs-137 kg}^{-1}$ . R = rye, B = barley, W = wheat and O = oats [44].

the precocity of rye compared with the other species, resulting in a higher retention of  $^{137}\text{Cs}$  by the more developed rye crops when the fallout from Chernobyl was deposited. UNSCEAR [19] calculated the transfer factors ( $\text{Bq } ^{137}\text{Cs a kg}^{-1} \text{ per kBq } ^{137}\text{Cs m}^{-2}$ ) in various diets after the Chernobyl accident. It appeared that the factors were usually lower than those observed previously for global fallout  $^{137}\text{Cs}$ , especially for northern Europe. One reason for this was seasonality, which in particular influences the levels for grain and for which the transfer factors post-Chernobyl were 1-2 orders of magnitude lower than those observed for global fallout. Another reason could be a lower availability of the Chernobyl  $^{137}\text{Cs}$  than of that from global fallout with respect to contamination of crops. But seasonality seems to be the most important factor.

#### Fruit seasonality

Berries and nuts showed relatively high radiocesium concentrations after the Chernobyl accident. It has been shown experimentally that radiocesium deposited on the leaves of fruit bushes is translocated to the

berries and the content of radiocaesium in the berries generally increases with time. In an experiment carried out by Kopp et al. [20], it was found that in red currants 0.6% of the deposited  $^{134}\text{Cs}$  activity was transported to the unripe fruit and 3.4% to the ripe berries; only minute amounts of  $^{85}\text{Sr}$  had been transferred to the edible parts, however. In bilberries the corresponding figures for  $^{134}\text{Cs}$  were 0.7% (unripe) and 1.8% (ripe) and for gooseberries 4.4% (unripe) and 3.7% (ripe).

#### 4.3.2. Models

The principal terrestrial foodchain models known to consider seasonality are ECOSYS and FARMLAND.

##### ECOSYS

ECOSYS (Fig. 14) [21] was developed for the rapid prediction of radiation exposure after single releases of radionuclides to the atmosphere. In order to accomplish this for vegetation, the model takes morphological and physiological changes of the plants during their growing period into consideration.

For the modelling of seasonality of plant contamination, the processes deposition, interception, translocation, growth dilution and weathering are considered. Dry and wet deposition are modelled separately. The amount of activity retained by the plant is characterized by the leaf area index (Fig. 15). This index is a suitable parameter for modelling because the leaf area is the boundary layer for dry as well as for wet

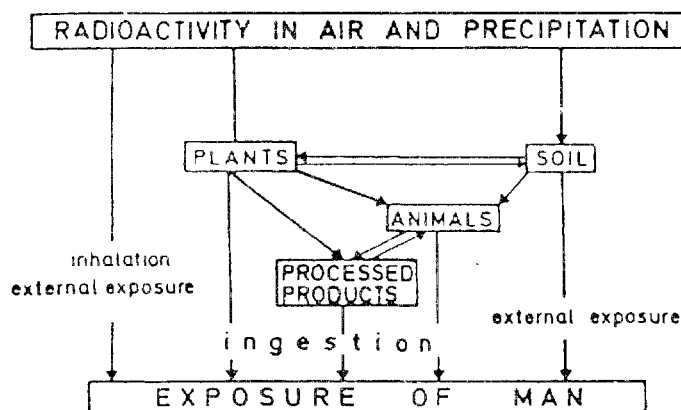


Fig. 14. Structure of the dynamic model ECOSYS [21].

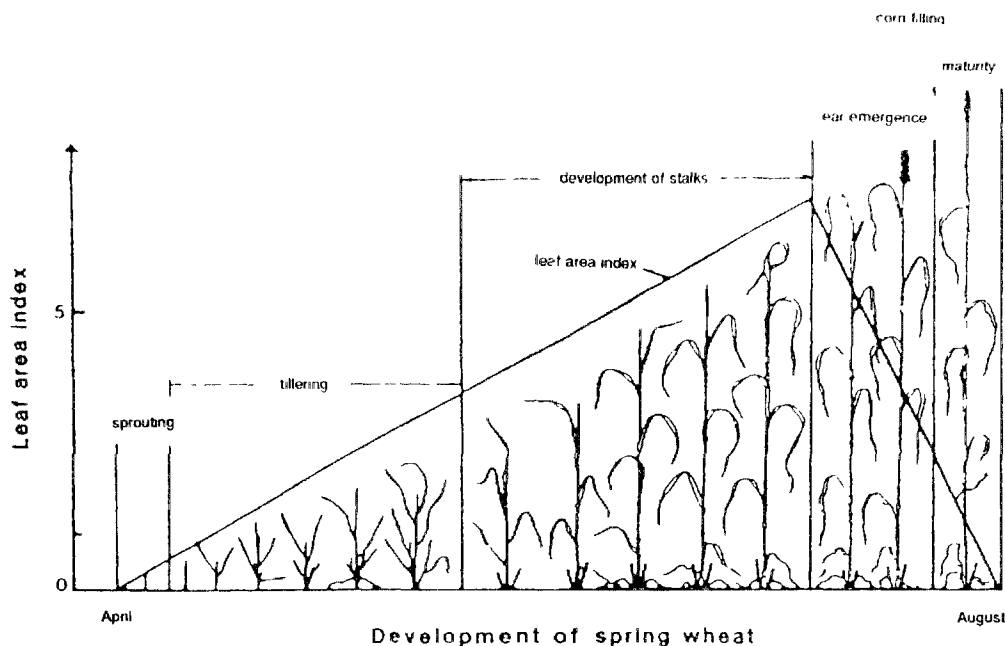


Fig. 15. Leaf area index of spring wheat during the growing period and its stages of development [21].

deposited radionuclides from the atmosphere. Data on the leaf area index are available from investigations on the growth characteristics of agricultural crops [22-26]. For the interception of wet deposition the amount of rainfall is another very important parameter [21,27] (Fig. 21).

The modelling of translocation is based on data from Middleton [2,3], Aarkrog [4 -7], Voigt et al. [28] and Ludwig [29] (Fig. 16).

For pasture grass growth dilution is modelled as a season-dependent process. The overall field loss of activity due to growth dilution and weathering varies by not more than a factor of 1.5 between May and October.

The version of ECOSYS which was available immediately after the Chernobyl accident was able to predict the contamination of foodstuffs quite well starting from the initial retention on plants. There were some discrepancies concerning the activity in different varieties of grain (winter and spring wheat, rye, winter and spring barley, oats) because no differentiation between species was taken into account. The differences in the morphological and physiological status of winter and spring varieties are very pronounced at beginning of May. Whereas winter varieties are

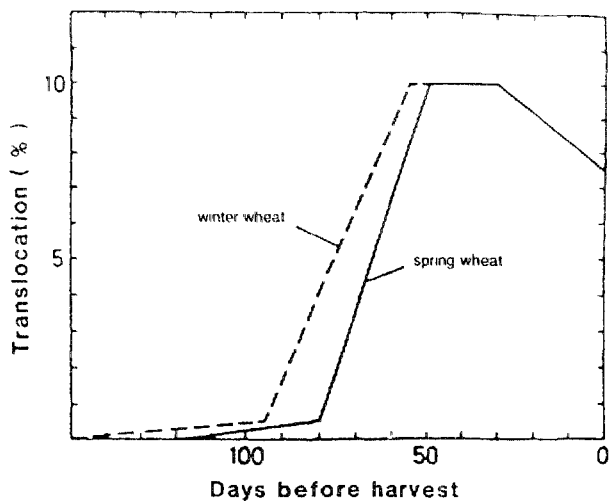


Fig. 16. Translocation of caesium from foliage to grain and its dependence on the period between deposition and harvest [21].

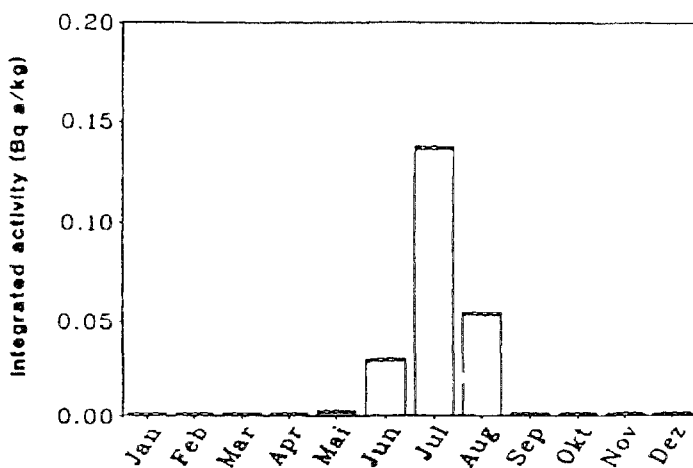


Fig. 17. Integrated Cs-137 activity concentration of spring wheat and its dependence on the month of deposition (dashed part: contribution of root uptake) [21].

normally well developed at this time, spring varieties have only recently emerged and the plants are still relatively small at the beginning of May. This leads to substantial differences in the intercepted activity and to different fractions being translocated from the foliage to the grain.

At later times of deposition (e.g. 1st June or 1st July) these differences would not have been as pronounced as they are at the beginning of May.

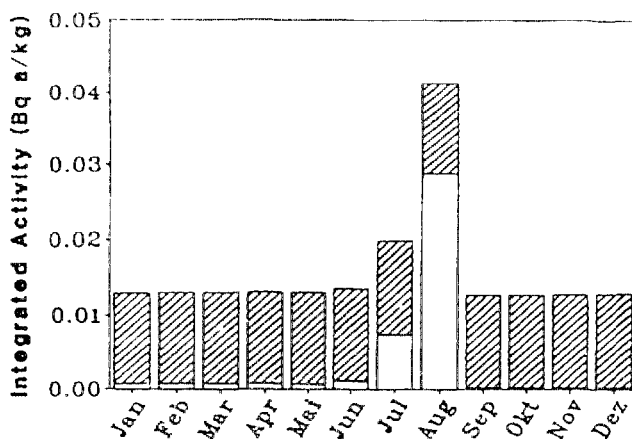
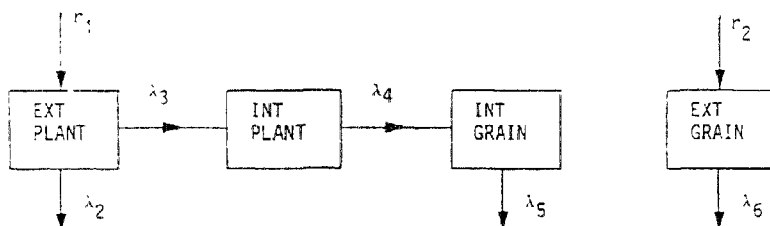


Fig. 18. Integrated Sr-90 activity concentration of spring wheat and its dependence on the month of deposition [27].



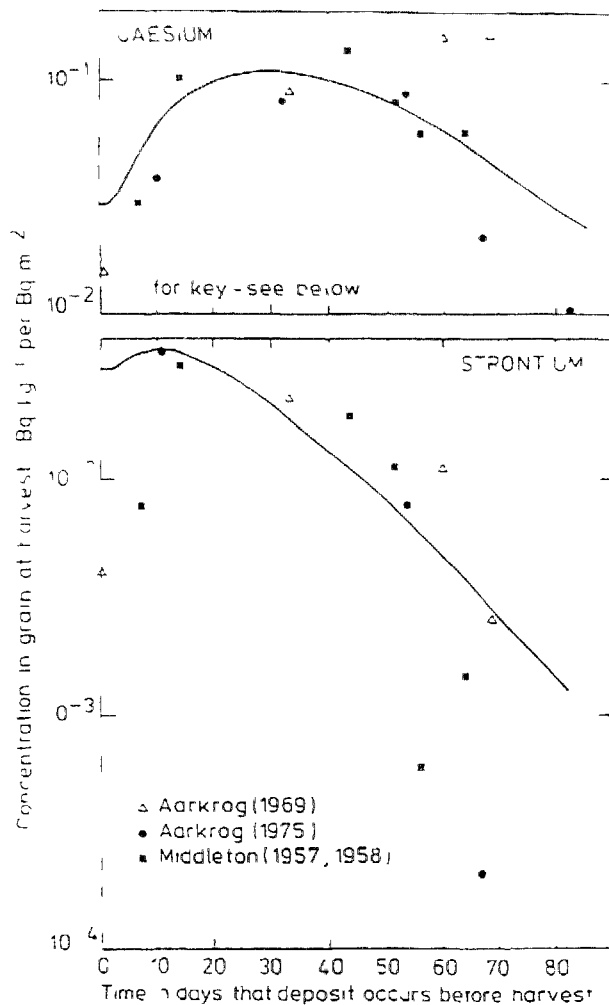
Notes

- The concentration in grain is obtained by summing the contributions from external and internal grain compartments.
- $r_1$  and  $r_2$  represent interception of the deposit by the whole plant and by the grain respectively. They have values of 0.3 and 0.012 respectively for all elements.
- $\lambda_2$  and  $\lambda_6$  represent losses due to weathering from the whole plant and the grain respectively.  $\lambda_2 = 4.95 \cdot 10^{-2} \text{ d}^{-1}$  and  $\lambda_6 = 4.82 \cdot 10^{-2} \text{ d}^{-1}$  for all elements.
- The values of  $\lambda_3$ ,  $\lambda_4$  and  $\lambda_5$  are obtained by fitting to experimental data and are element dependent. The values for strontium and caesium are:-

| Transfer Coefficient | Strontium $\text{d}^{-1}$ | Caesium $\text{d}^{-1}$ |
|----------------------|---------------------------|-------------------------|
| $\lambda_3$          | $3.7 \cdot 10^{-2}$       | $3.4 \cdot 10^{-2}$     |
| $\lambda_4$          | $6.9 \cdot 10^{-2}$       | $6.4 \cdot 10^{-2}$     |
| $\lambda_5$          | $4.5 \cdot 10^{-1}$       | $5.2 \cdot 10^{-2}$     |

Fig. 19. Revised model for the deposition of radionuclides onto grain plants [13].





**Fig. 20. Comparison of model and experimental results for single deposits of caesium and strontium on grain plants at various times before harvest [13].**

## FARMLAND

The National Radiological Protection Board's (NRPB) foodchain model FARMLAND [30-32] has been developed over a number of years and it includes seasonality (Fig. 19). The model and its parameter values were derived from global fallout observations as well as from experimental measurements, notably Middleton [2], Middleton and Squire [3] and Aarkrog [4-8] (Fig. 20). Furthermore compilations of data, notably those of Ng [33] and International Union of Radioecologists (IUR) [34] have been applied.

From a seasonal viewpoint FARMLAND predicted the transfer of radionuclides to food following Chernobyl reasonably well. In particular,

FARMLAND's predictions of a decline in cow's milk concentrations following the deposit followed by an increase when the animals were fed on stored feed was observed following Chernobyl. However, the release occurred at a single time of year and data on the stage of growth of crops from locations other than that on which the model is based are difficult to obtain. Therefore, the testing of the seasonal aspects of the model has, so far, not been as rigorous as is desirable.

In testing the validity of FARMLAND with Chernobyl data some discrepancies were noted with results for particular farms in the United Kingdom. This was due mainly to differences between the general assumptions regarding agricultural practices adopted in FARMLAND and the actual practices at the individual farms. Also there were differences in some cases when the deposition occurred in rainfall as FARMLAND does not take account of the rainfall rate, neither does it distinguish between wet and dry deposition.

#### Risø-models

The curves shown in Fig. 20 are to some extent similar to those in Figs. 5 and 6. These models were made from least squares fitting of the normal distribution curve to the experimental data [8]. The model for Cs (Fig. 6) was applied to the Chernobyl observations in 1986 in Denmark by Aarkrog [35]. The model predicted  $4.2 \text{ Bq } ^{137}\text{Cs kg}^{-1}$  grain but the observed level was  $0.3 \text{ Bq } ^{137}\text{Cs kg}^{-1}$ . One reason for this discrepancy was a more rapid field loss of the Chernobyl  $^{137}\text{Cs}$  than that found during the experiments because intense rain in Denmark removed a good deal of the initially deposited Chernobyl debris. This effect has been shown by Müller and Pröhl [27] (see Fig. 21) who found that the interception factor can vary from nearly one hundred down to only a few percent.

A simpler empirical model for the contamination of grain in Denmark has been based on global fallout data collected since 1959 (Table VIII). This model assumes that the  $^{137}\text{Cs}$  deposition in May-August is decisive for the contamination of the grain at harvest time in August-September. The model gave good predictions prior to the Chernobyl accident; but the predictions in 1986 for Danish grain came out 1-2 orders of magnitude higher than those actually observed. The reason for this was that the empirical models in Table VIII assume that the fallout is evenly distributed over the period May-August, but the majority of the Chernobyl

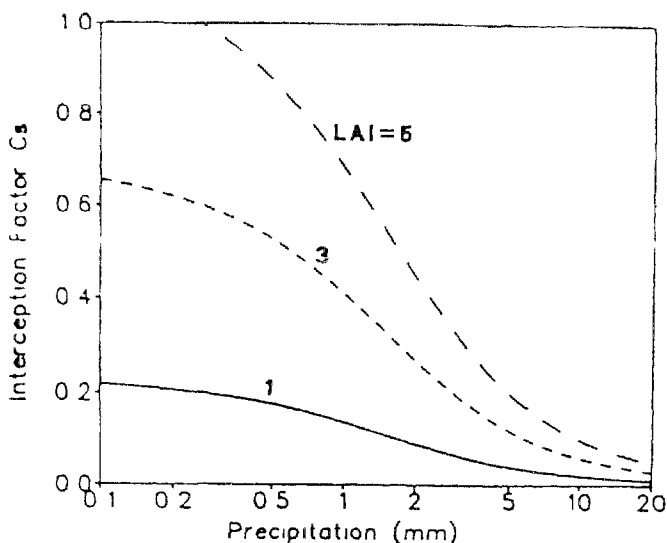


Fig. 21. Interception factor for wet deposition of caesium on grass with different leaf area indices (LAI) as used in ECOSYS [27].

TABLE VIII PREDICTION MODELS FOR  $^{137}\text{Cs}$  IN CEREAL GRAIN [41]  
(unit for radioecological sensitivity  $\text{pCi } ^{137}\text{Cs kg}^{-1}\text{a}$  per  $\text{mCi } ^{137}\text{Cs km}^{-2}$ )

| No | Sample | Area    | Period  | $^{137}\text{Cs}$ -activity, $\text{pCi kg}^{-1}_{(1)}$ | Sensitivity/ | r         |
|----|--------|---------|---------|---|--------------|-----------|
| 1  | Rye    | Denmark | 1962-76 | $79d_{1-1}^{1-1}(\text{May-Aug}) + 5.0d_{1-1}^{1-1}$    | 47           | 0.9991*** |
| 2  | - ' -  | - " -   | - " -   | $85d_{1-1}^{1-1}(\text{May-Aug})$                       | 46           | 0.9957*** |
| 3  | Barley | - " -   | - " -   | $59d_{1-1}^{1-1}(\text{May-Aug})$                       | 32           | 0.9957*** |
| 4  | Wheat  | - " -   | - " -   | $52d_{1-1}^{1-1}(\text{May-Aug})$                       | 28           | 0.9857*** |
| 5  | Oats   | - " -   | - " -   | $50d_{1-1}^{1-1}(\text{May-Aug})$                       | 27           | 0.9952*** |

debris was deposited in May, where the initial retention by the sparsely developed crops was low. Hence, although these empirical grain models have considered seasonality they were not sophisticated enough for dealing with the Chernobyl input.

#### OSCAAR-CHRONIC

Matsuzuru [36] has also proposed models that take seasonality into account. A foodchain model is incorporated in the computer code system OSCAAR-CHRONIC which evaluates chronic exposure due to radionuclides remaining in a Japanese environment after a reactor accident. The model is an extension of the methodology used in the U.S. CRAC code [37]. Four kinds of important Japanese crops are selected: leafy vegetables, cereals,

|                        |               |   |               |   |               |           |   |   |           |               |    |    |
|------------------------|---------------|---|---------------|---|---------------|-----------|---|---|-----------|---------------|----|----|
|                        | ( month )     |   |               |   |               |           |   |   |           |               |    |    |
|                        | 1             | 2 | 3             | 4 | 5             | 6         | 7 | 8 | 9         | 10            | 11 | 12 |
| Pasture grass          | under growing |   |               |   | grazed by cow |           |   |   |           | under growing |    |    |
| Cereals                | dormant       |   |               |   | under growing |           |   |   | harvested | dormant       |    |    |
| Root crop              | har           | d | under growing |   |               | harvested |   |   |           |               |    |    |
| Leafy vegetables       | h             | d | under growing |   |               | harvested |   |   |           |               |    |    |
| Non-leafy veg & Fruits | under growing |   | harvested     |   |               |           |   |   |           | dormant u     |    |    |

Fig. 22. The Model of Agricultural Practice [36].

rootcrops and fruits (including non-leafy vegetables). It is assumed that the year can be divided into 3 and 4 periods for the various crops as shown in Fig. 22 and time dependent functions are postulated for the intake of radionuclide for the various periods. Translocation is not included in the model, hence the model seems to underpredict  $^{137}\text{Cs}$  intake from cereals (Table IX). The parameter values employed in the model are Japanese as far as productivity density of grass/crops, growing periods and consumption rate of foodstuffs are concerned. Foreign data are applied for the other parameters such as interception factors, transfer rates, time delay to consumption and so on. Most of the values are those used in the CRAC code and others are taken from NRPB data sets.

Figures 23 and 24 show the results of the model calculations for milk and cereals, respectively, for Japanese conditions. The models were

TABLE IX. RATIOS OF PREDICTED-TO-OBSERVED CONCENTRATIONS OF  $^{137}\text{Cs}$  IN WINTER GRAIN (w) AND SPRING GRAIN (s) BASED ON GEOMETRIC MEANS OF ALL MEASUREMENTS [38]  
(GM: geometric mean, GSD: geometric standard deviation)

| MODEL    | T<br>ROSKILDE<br>(w) | T<br>ROSKILDE<br>(s) | S<br>GEEL<br>(s) | C<br>TOKAI<br>(w) | H<br>TRANVIK<br>(w) | N<br>MUNICH<br>(w) | L<br>LOVISA<br>(w) | L<br>LOVISA<br>(s) | GM    | GSD  |
|----------|----------------------|----------------------|------------------|-------------------|---------------------|--------------------|--------------------|--------------------|-------|------|
| DETRA    |                      | 8.0                  | 7.5              |                   |                     |                    |                    | 7.5                | 7.7   | 1.0  |
| SIRATEC  | 0.26                 | 0.52                 | 0.2              | 0.029             | 0.14                | 0.43               | 0.13               | 0.18               | 0.18  | 2.4  |
| FARMLAND | 3.3                  | 21                   | 3.8              |                   |                     |                    | 1.6                | 0.65               | 3.1   | 3.6  |
| ECOSYS*  | 1.3                  | 0.13                 | 0.087            | 0.055             | 0.63                | 0.96               |                    | 0.03               | 0.20  | 4.0  |
| SPADE 2  | 3.7                  | 6.6                  |                  | 0.13              | 8.7                 | 4.9                | 9.7                | 0.014              | 1.5   | 12.7 |
| TERFOC   | 0.029                | 0.027                |                  | 0.49              | 0.0052              | 0.17               | 0.019              | 0.013              | 0.036 | 4.7  |
| CHRONIC  | 0.029                | 0.026                |                  | 0.49              | 0.0059              | 0.17               | 0.019              | 0.015              | 0.036 | 4.6  |
| RAGTIME  | 7.1                  |                      |                  | 0.91              | 2.2                 | 6.7                | 1.8                |                    | 2.8   | 2.4  |
| CHERPAC  | 0.34                 | 0.85                 |                  | 4.1               | 7.3E-6              | 7.96               | 0.028              | 0.00033            | 0.049 | 176  |

\* Calculations for ECOSYS are grain specific: listed are P/O ratios for wheat.

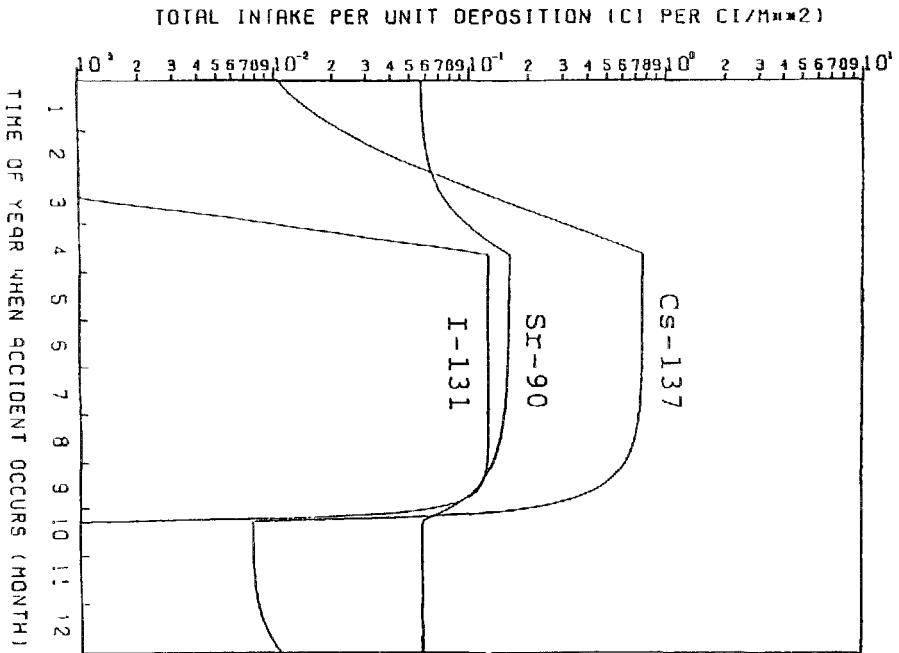


Fig. 23. Total intake from milk [36].

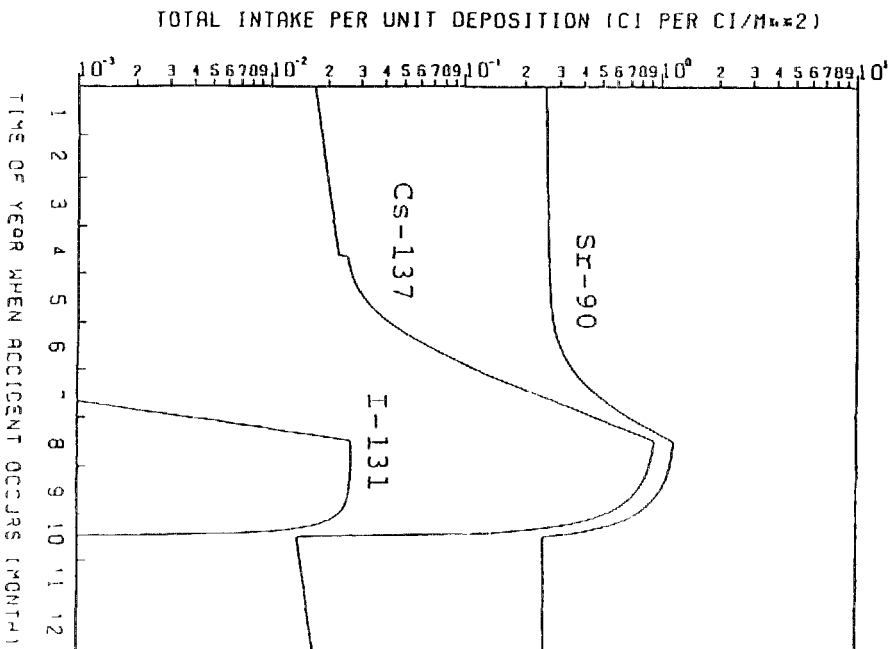


Fig. 24. Total intake from cereals [36].

developed before the Chernobyl accident. In the BIOMOVs study the model was applied to the Chernobyl scenario for  $^{131}\text{I}$  and  $^{137}\text{Cs}$ .

#### BIOMOVs international model validation study

The BIOMOVs group [38] compared a number of model predictions of  $^{137}\text{Cs}$  in grain with the actual concentrations found after the Chernobyl accident in 1986 (Table IX). Three of the models have been described above (ECOSYS, FARMLAND and CHRONIC). None of the model predictions were close to observed values for all locations tested and there was considerable variability in the model predictions. For this scenario, the BIOMOVs group did not attempt to analyze the reasons for predictive variations and discrepancies because of the generally poor quality of the information available from the sites tested.

The BIOMOVs study concludes that it is difficult to find reasons for the discrepancies between the model predictions and the observations. However, it concluded that they may be associated with a strong seasonal factor and the absence of site specific information on the state of maturation of the plants at the time of the accident.

#### 4.4. Need for Model Improvement

##### 4.4.1. ECOSYS

After the Chernobyl accident the ECOSYS model was improved in various ways. Dry and wet deposition is now modelled separately, the leaf area index during the growing period of each plant species is considered and a differentiation between all six major cereal species cultivated in Europe is made.

Furthermore, realistic consideration of animal diets especially for beef and dairy cattle, has been introduced [39]. The radionuclide content of animal feed is, as mentioned earlier, subject to seasonal variability (Section 4.1.2.) due to the differing growth intensities of pasture grass during the growing season. The supply of fresh pasture grass is highest at the start of the growing period and decreases towards the end of the growing period. This means that increasing amounts of fresh pasture have to be substituted by other feedstuffs such as maize, beet, beet leaves etc. Also for a reliable prediction of the activity in milk and meat in the winter following the deposition, the real site-specific feeding regimes

have to be taken into account. It is also necessary, for a realistic prediction of the activity in winter fodder, to have information about the period when hay and silage are prepared, which may vary for climatic reasons.

#### 4.4.2. FARMLAND

With regard to FARMLAND, this model has, according to NRPB, not been modified or "fine-tuned" in any way to take account of the Chernobyl data. However, some particular parameter values may be altered in the light of data obtained after Chernobyl, although care is required not to place too much reliance on a single source of data. The comparison of FARMLAND predictions with post-Chernobyl measurements generally strengthens NRPB's belief in the validity of the model for use in generic radiological assessments, the use for which it was intended. For this reason some of the detailed site specific modifications, such as can be incorporated into the ECOSYS model, are not included. However, it is recognised that further development of FARMLAND is desirable to enable the effect of deposition in varying wet and dry conditions to be taken into account for more detailed applications.

#### 4.4.3. Summary

The Chernobyl accident, which was a major short time input of radionuclides into the biosphere, demonstrated that no existing models (at the time of the accident) were able adequately to predict the effect of seasonality. A number of models have considered the importance of the leaf area index and translocations (as described by Pröhl et al. [21]), but as shown by Müller and Pröhl [27] the meteorological conditions (mm precipitation) also had a significant impact on the interception factor. Furthermore, the meteorological conditions in the time after the deposition had an effect on the field loss, i.e. rate of decrease of the activity in the plants. Field loss is influenced by weathering processes, e.g. washoff, and also by plant growth dilution. All these elements have to be included in models if they are to yield reliable model predictions for single event radioactive contamination. In order to describe the seasonality process adequately, models have to be site specific.

At any particular time of the year the stage of plant development in different regions of the world varies widely. The time of harvest is also variable. (Müller and Pröhl [27] (Fig. 25)). Both of these factors can

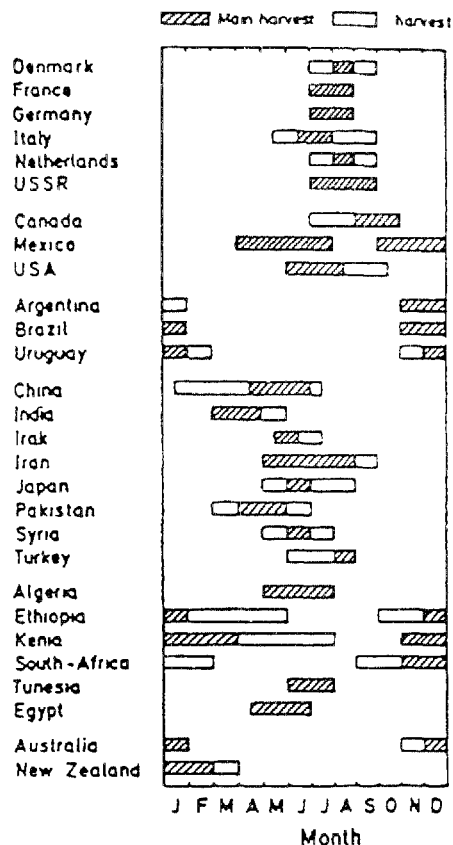


Fig. 25. Times of wheat harvest in some countries [45].

strongly influence the radionuclide content of crops; other factors, such as harvesting methods may also have an influence.

The BIOMOVs study [38] has shown that the more complex dynamic models do not necessarily give more reliable predictions than simple models. However in the case of a short time input of radionuclides into the environment, simple steady-state models for predicting the contamination of cereals will not suffice and dynamic models are required. If the contamination is "chronic" (e.g. as has been the case for global fallout) a simple model will probably be just as good as the complex one. The Riso model shown in Table 8 has thus proved to produce very reliable predictions throughout the years of global fallout  $^{137}\text{Cs}$  in Danish cereals.



#### 4.5. Conclusion and Recommendations

Seasonality may have a strong influence on the radiological impact of environmental radioactive contamination. Seasonality is influenced by a number of environmental factors. In the terrestrial soil-to-plant-animal system the ratio between direct and indirect (root uptake) contamination of the plants is of major importance. Seasonality is pronounced if direct contamination is the dominating pathway. The degree of translocation of radionuclides to the fruit bodies of plants also influences seasonality. High translocation normally reduces seasonality. Contamination of cereal grains with  $^{137}\text{Cs}$  and  $^{131}\text{I}$  shows a pronounced seasonality, whereas  $^{90}\text{Sr}$  in root crops shows little or no seasonality.

In aquatic ecosystems ice cover, plankton bloom and runoff during snow melting may influence seasonality. Natural and seminatural ecosystems show generally lower seasonality than agricultural systems. Temperate zones show higher seasonality than subtropical and tropical regions.

There is a need for a better understanding of the processes that influence seasonality, especially those connected to meteorology, e.g., washoff. The physicochemical behaviour of contaminants with respect to initial uptake and translocation in crops needs further study. Information on seasonality in connection with fruit and vegetable contamination is particularly sparse. Seasonality effects also require further investigation in connection with aquatic pathways (e.g., fish, drinking water). Finally, natural and seminatural ecosystems should be further examined with respect to seasonality.

In order to obtain reliable predictions of the radiological consequences of short term contamination of crops, the model applied should be site specific and be capable of taking account of changes in interception characteristics with plant growth, the effects of translocation, of rain intensity and of the physicochemical characteristics of the contaminant.

## Annex 1

### Definition of the Seasonality Factor, S

The seasonality factor S is the coefficient of variation (CV) of the monthly infinite time integral concentrations of a given radionuclide in a given sample observed throughout twelve months of a year.

A monthly deposition of  $1 \text{ Bq m}^{-2}$  in month (i) of radionuclide (p) results in an infinite time concentration integral in sample item (a) of  $I_{iap} \text{ Bq Kg}^{-1} \text{ yr}$ .

The variation of  $I_{iap}$  between the 12 months of the year is a measure of the degree of seasonality. This variation is quantified as the coefficient of variation CV, i.e. the relative standard deviation of the twelve  $I_{iap}$  values, which have an annual mean of  $I_m$ . Hence

$$S \equiv CV = \frac{\sqrt{\frac{\sum_{i=1}^{12} (I_m - I_{iap})^2}{(12 - 1)}}}{I_m}$$

If  $CV > 0$  seasonality is present. If  $CV \approx 0$  no seasonality is observed.

### Application of S

Pröhl [40] has calculated a variety of  $I_{iap}$  values (see e.g. Figs. 17 and 18) based on a time dependent simulation model SINK, which is closely related to the ECOSYS model (Pröhl et al. [21]). These  $I_{iap}$  values are site specific for the average conditions in southern Germany and should thus be considered only as examples.

Below are summarized some S values calculated from Pröhl [40]

|                 | Sr-90 | I-131 | Cs-137 |
|-----------------|-------|-------|--------|
| MILK            | 0.4   | 1.1   | 0.8    |
| WHEAT           | 0.5   | 2.6   | 1.9    |
| ROOT VEGETABLES | 0     | -     | 1.2    |
| LEAF VEGETABLES | 0.4   | 0.2   | 0.5    |

It is evident (Figs. 17 and 18) that the degree of seasonality depends strongly on the ratio between root uptake and direct contamination of the crops. If this ratio is low the seasonality is high and vice versa. However, other factors, e.g. translocation of certain radionuclides play an important role, in particular in the case of fruits and cereals.

It should further be noticed that for certain sample types, e.g. cereal grain, there may be even monthly seasonality variation, but for practical reasons the definition of the seasonality factor has been based on monthly data.

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