ON THE VARIATION OF THE LEVELS OF ^{90}Sr AND OTHER FALLOUT NUCLIDES IN THE GRAIN OF RYE, BARLEY, WHEAT AND OATS

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Abstract—The predominant influence of cereal grains as a ^{90}Sr donor (and a ^{137}Cs and ^{54}Mn donor as well, in certain years) in total Danish diet has stimulated the present investigation. A country wide collection of grain samples has been carried out in the years 1959–64 at a number of selected state experimental farms. Along with the grain sampling, data on ^{90}Sr in air, soil and precipitation have been collected.

Analysis of variance shows that rye contained more ^{90}Sr, ^{137}Cs and ^{54}Mn than barley or wheat or oats. It is demonstrated that awns and, to some degree, grain size play an important role in the direct contamination of grain. The variation between locations, as regards ^{90}Sr levels in grain, depended upon the amounts of precipitation and, to some degree, on the calcium level and the stable strontium to calcium ratio of the soil. The variation between years followed the ^{90}Sr activity in air in August and the amount of precipitation in this month. Winter and spring varieties of rye and wheat did not differ significantly in ^{90}Sr and ^{137}Cs contents. The uptake of ^{54}Mn in cereals related to the fallout rate rather than to the accumulated fallout.

By multiple regression analysis it is shown that the pCi ^{90}Sr/g Ca level for the years 1959–64 in the different species of Danish grain considered separately can be described by equations of the type:

$$\text{pCi } ^{90}\text{Sr/g Ca} = k_0 \cdot A^{k_1} \cdot B^{k_2} \cdot C^{k_3}$$

where A, B and C respectively represent mm precipitation in August, pCi $^{90}Sr/m^3$ air in August and accumulated mCi $^{90}Sr/km^2$ in soil by September; k_1, k_2 and k_3 are the respective partial regression coefficients.

INTRODUCTION

In most countries in Europe, North America and Oceania dairy produce is the predominant source of radiostrontium in the human diet.[1] However, in countries where the consumption of wholemeal bread is common, as, for example, in Poland and Denmark, the contribution from grain products to the total ^{90}Sr intake with the diet might be greater than that from dairy products.[2] In the years 1959–65 grain products have on the average contributed approx. half of the daily ^{90}Sr intake in Denmark.

As regards the other important long-lived fission product from worldwide fallout ^{137}Cs, it appears that cereals in the period 1962–5 contributed approx. one third of the total ^{137}Cs intake from Danish diet.

In December 1963 and June 1964 the daily mean intakes of ^{54}Mn with total Danish diet were 134 and 83 pCi respectively. More than 80% of this radiomanganese originated from bread.[3]

The purpose of the present investigation is to elucidate the variations existing in the Danish environment between the contents of various fallout nuclides in grains of rye, barley, wheat and oats, and to demonstrate the relations between ^{90}Sr in cereal grain and the levels of nuclear debris found in air, precipitation and soil.

MATERIAL AND METHODS

The samples used have been obtained from 10 Danish state experimental farms. As indicated in Fig. 1, locations Tl, S, and J have sandy soils, the other farms predominantly clayey soils. Figure 2 shows the relative annual mean precipitation for the years 1953–64 (the period of global fallout). Location S shows a maximum (814 mm per year) and farms Ts a minimum (524 mm/year). The overall mean for the 10 stations was 658 mm/year, i.e. nearly equal to the area weighted country mean: 682 mm/year (in 1953–64).

Samples of rye, barley, wheat and oats have, as far as possible, been collected from each farm since 1959. In 1960 the samples from locations Tl and Ab were lost, and location Ø was first included in the programme in 1961. Wheat has never been obtained from location Tl and seldom from S. Since 1962 samples of spring varieties of wheat and rye have been obtained in several cases along with the more common winter varieties of these species.

The cereal grain in Denmark is harvested in August–September. The mean harvest date

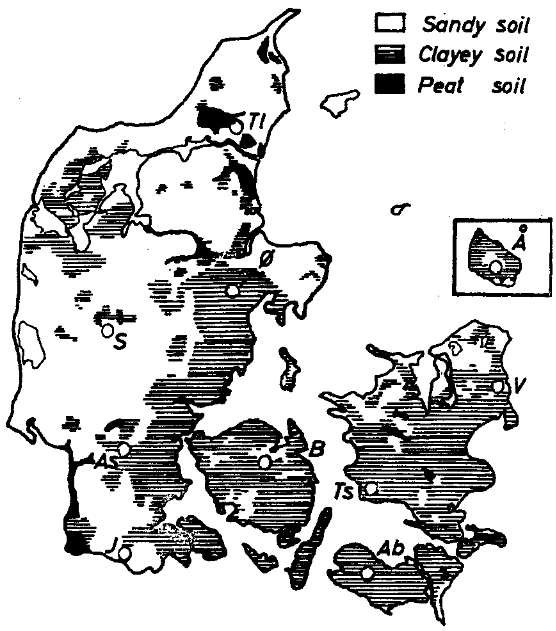

FIG. 1. State experimental farms in Denmark and soil types. Scale: 1:4,400,000.

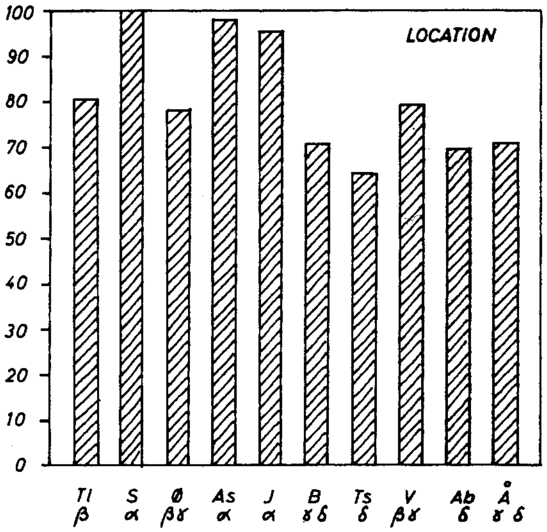

FIG. 2. Relative mean amounts of annual precipitation at the state experimental farms in the period 1953–64.

for the years 1962–4 in this study was August 27 (interval August 4–September 25). Barley is on the average harvested 10 days before the other species. The samples were received for analyses 2–3 months after harvest. Each sample consisted of 1–2 kg grain. Approximately 1 kg of grain was counted in a 1.6 l. can fitting over a 3 × 3 in. NaI (Tl) crystal (hat geometry). The pulses were analysed in a 256 channel TMC pulse height analyser. From this measurement the ^{137}Cs and ^{54}Mn contents were determined. After γ-counting, the grain was ashed at 500–550°C in silica dishes and radiostrontium analyses were carried out according to the classical method using fuming nitric acid.[4] Finally the levels of calcium and stable strontium were determined according to Webb *et al.*[5] Since 1961 most of the strontium analyses of grain have been carried out as double determinations. The estimated overall relative standard deviation (S.D.) of the ^{90}Sr analyses was 10–15%, somewhat higher in the first years than in the last due to higher levels since harvest 1962, and to increasing analytical experience throughout the years. The relative S.D.'s of the γ-measurements were approx. 15%.

The data on ^{90}Sr in air, precipitation and soil used in the multiple regression analysis were obtained from ref. 3. Missing soil figures for the years 1959–60 were estimated from U.K. data.[6] The amounts of precipitation at the different stations were obtained from the Meteorological Institute in Copenhagen.[7]

The statistical methods used in the treatment of the results are given in the statistical literature.[8, 9] Multiple regression analysis and analysis of variance have been calculated on a Gier computer.[10, 11] The programme used for analysis of variance[11] has been especially useful because this programme makes it possible to accomplish the analysis, even when several figures are missing, which is inevitable in a field-study such as this.

In the significance tests the following symbols will be used:

x: probably significant	(confidence level: 0.05)
xx: significant	(confidence level: 0.01)
xxx: highly significant	(confidence level: 0.001)

Columns with identical greek letters in the diagrams do not differ significantly by the *t*-test, but they might, in some cases, be probably significantly different.

RESULTS

Tables 1–5 show the results of the measurements of pCi ^{90}Sr/g Ca (S.U.), g Ca/kg, mg stable Sr/g Ca, ^{137}Cs and ^{54}Mn respectively in Danish cereal grain collected in the period 1959–64. It appears from Table 6 that the variation between species of the levels of S.U., pCi ^{137}Cs/kg and pCi ^{54}Mn/kg were all significant.

^{90}*Sr*

Figure 3 shows that rye displayed a significantly higher pCi ^{90}Sr/g Ca ratio than barley and oats. As regards the pCi ^{90}Sr/kg level (Fig. 4), it was found that the activity in rye was higher than in the other three species, which, as a whole, did not differ significantly. The low S.U. level in oats is a result of the relatively high Ca level in this cereal (Fig. 5).

As regards locations (Fig. 3) the highest levels were found at S, J and Tl, which showed pCi ^{90}Sr/g Ca ratios nearly a factor of two higher than those found at Ab, B and Ts.

If the S.U. mean levels for the period 1959–64 are considered (Fig. 3), it appears that 1963 showed a pronounced peak, which was ten times higher than the minima measured in 1960 and 1961.

^{137}*Cs*

The relative mean contents of ^{137}Cs in grain collected in 1962–4 appear in Fig. 6. Rye contained significantly more ^{137}Cs than the other cereals. The differences between locations were less pronounced than for ^{90}Sr. The ^{137}Cs level in 1963 was approximately twice as high as in 1962 and 1964.

^{54}*Mn*

^{54}Mn in cereal grain (Fig. 7), which was determined only in 1963 and 1964 (the 1962 levels were not measurable), showed a picture similar to that of ^{90}Sr and ^{137}Cs, i.e. rye displayed a higher ^{54}Mn level than the other species. The *t*-test could, however, only prove that the activity in the rye was higher than for wheat.

When comparing the years, it should be noticed that the ^{54}Mn 1964 level was corrected for decay ($t^{1/2} = 310$ days) back to the 1963 harvest dates. The corrected 1964 level was approx. 45% of the 1963 level, i.e. nearly equal to the percentage found for ^{90}Sr (43%), but lower than that for ^{137}Cs (55%).

Table 1. *pCi* ^{90}*Sr/g Ca in Danish Grain,* 1959–64

Location		Tl	S	Ø	As	J	B	Ts	V	Ab	Å
Year	Sort										
1959	Rye	108	135	—	173	188	—	125	129	93	184
	Barley	116	88	—	67	44	63	81	68	65	130
	Wheat	—	—	—	89	—	88	81	61	68	213
	Oats	13	66	—	23	47	57	36	38	24	111
1960	Rye	—	118	—	105	166	77	90	79	—	103
	Barley	—	112	—	66	72	63	56	58	—	60
	Wheat	—	96	—	76	251	82	58	81	—	115
	Oats	—	78	—	71	60	31	35	39	—	45
1961	Rye	116	51	36	123	103	114	98	80	—	92
	Barley	81	106	67	84	208	45	54	87	57	80
	Wheat	—	—	93	76	—	60	61	86	49	118
	Oats	64	74	29	31	64	32	20	45	41	44
1962	Rye	470	418	470	425	627	193	319	297	—	371
	Barley	336	374	231	483	479	171	92	237	229	328
	Wheat	—	—	433	347	548	334	297	—	139	283
	Oats	147	243	175	179	300	93	162	—	109	215
1963	Rye	1781	1168	1500	1870	1077	1286	994	830	—	703
	Barley	1125	1353	559	1293	669	662	439	680	484	443
	Wheat	—	—	1300	903	931	910	878	—	538	831
	Oats	472	466	439	674	290	380	318	453	147	362
1964	Rye	760	733	508	565	535	261	253	425	—	325
	Barley	631	868	398	544	471	107	238	302	209	219
	Wheat	—	—	438	691	—	295	290	413	150	224
	Oats	242	352	189	190	251	106	102	174	91	76

Table 2. g Ca/kg Danish Grain, 1959–64

Location		Tl	S	Ø	As	J	B	Ts	V	Ab	Å
Year	Sort										
1959	Rye	0.35	0.38	—	0.38	0.40	—	0.35	0.45	0.48	0.35
	Barley	0.27	0.44	—	0.39	1.02	0.32	0.37	0.54	0.41	0.48
	Wheat	—	—	—	0.36	—	0.30	0.40	0.52	0.28	0.42
	Oats	0.69	0.98	—	0.85	0.81	0.71	0.79	0.83	0.76	0.55
1960	Rye	—	0.41	—	0.32	0.32	0.51	0.46	0.36	—	0.35
	Barley	—	0.43	—	0.41	0.37	0.40	0.37	0.41	—	0.62
	Wheat	—	0.45	—	0.35	0.30	0.33	0.38	0.32	—	0.35
	Oats	—	0.96	—	0.68	1.02	0.96	0.78	1.20	—	0.85
1961	Rye	0.42	0.30	0.42	0.38	0.22	0.28	0.30	0.26	—	0.65
	Barley	0.47	0.49	0.49	0.41	0.35	0.45	0.43	0.49	0.46	0.49
	Wheat	—	—	0.32	0.43	—	0.42	0.28	0.35	0.47	0.30
	Oats	0.62	0.51	1.48	1.10	0.89	0.87	0.65	0.74	0.89	0.74
1962	Rye	0.33	0.55	0.46	0.42	0.37	0.51	0.50	0.56	—	0.43
	Barley	0.44	0.41	0.65	0.55	0.43	0.55	1.02	0.54	0.50	0.43
	Wheat	—	—	0.43	0.30	0.43	0.39	0.32	—	0.62	0.36
	Oats	0.78	0.71	0.85	0.74	0.68	0.87	0.74	—	1.29	0.69
1963	Rye	0.37	0.41	0.32	0.51	0.40	0.54	0.62	0.50	—	0.44
	Barley	0.66	0.43	0.59	0.62	0.39	0.63	0.38	0.58	0.42	0.60
	Wheat	—	—	0.32	0.59	0.43	0.38	0.42	—	0.46	0.32
	Oats	0.93	0.63	0.79	0.81	0.85	0.83	0.85	0.98	0.98	0.87
1964	Rye	0.65	0.55	0.44	0.41	0.42	0.40	0.39	0.50	—	0.35
	Barley	0.48	0.36	0.54	0.59	0.43	0.98	0.47	0.46	0.48	0.35
	Wheat	—	—	0.39	0.36	—	0.40	0.36	0.35	0.45	0.41
	Oats	0.81	0.56	0.76	0.81	0.85	0.65	0.83	1.12	0.98	0.74

Table 3. mg stable Sr/g Ca in Danish Grain, 1959, 1962–4

Location		Tl	S	Ø	As	J	B	Ts	V	Ab	Å
Year	Sort										
1959	**Rye**	2.8	3.2	—	1.1	2.2	—	1.6	1.8	3.4	1.9
	Barley	1.4	3.5	—	2.2	1.4	2.0	1.6	3.2	2.3	2.6
	Wheat	—	—	—	2.2	—	3.0	2.0	0.9	2.7	1.9
	Oats	2.8	2.4	—	0.7	2.1	1.3	1.7	1.9	2.3	1.6
1962	**Rye**	1.9	2.4	3.6	3.7	2.1	1.5	2.3	1.5	—	2.6
	Barley	2.4	6.9	2.8	3.7	3.0	1.7	2.3	2.5	2.1	3.2
	Wheat	—	—	5.4	8.9	2.4	2.5	5.1	—	2.3	3.7
	Oats	2.3	3.2	2.6	2.8	2.8	1.9	1.9	1.6	1.0	1.3
1963	**Rye**	3.5	2.0	2.6	5.3	1.9	1.4	1.8	2.8	—	1.8
	Barley	3.0	3.9	1.9	4.5	3.2	1.0	1.7	2.1	3.5	1.6
	Wheat	—	—	4.5	4.3	3.2	2.3	1.7	—	2.6	2.1
	Oats	1.1	3.0	1.9	1.7	1.5	1.7	1.7	2.1	1.4	1.8
1964	**Rye**	1.3	2.7	1.7	1.2	2.7	2.0	1.1	1.7	—	1.7
	Barley	2.2	4.9	4.1	1.2	1.4	2.3	2.8	4.2	4.2	2.1
	Wheat	—	—	5.1	3.2	—	2.4	3.3	2.4	3.7	2.8
	Oats	1.3	2.7	1.9	0.6	1.7	1.3	0.8	1.2	1.2	0.9

Table 4. *^{137}Cs in Danish Grain,* 1962–4

Location		Tl	S	Ø	As	J	B	Ts	V	Ab	Å
Year	Sort	pCi ^{137}Cs/g K									
1962	Rye	159	186	218	110	—	80	116	110	—	145
	Barley	148	148	88	123	135	79	81	90	84	122
	Wheat	—	—	124	100	252	108	62	—	70	71
	Oats	92	143	74	—	232	70	117	—	—	42
1963	Rye	264	371	202	537	299	274	325	243	—	183
	Barley	179	264	200	372	214	448	218	148	308	139
	Wheat	—	—	245	608	206	202	152	—	222	226
	Oats	296	183	206	380	168	169	258	174	248	191
1964	Rye	232	248	181	274	252	133	125	125	—	140
	Barley	111	159	92	170	163	63	92	74	71	86
	Wheat	—	—	132	153	—	79	96	92	69	84
	Oats	140	161	101	126	123	70	91	120	59	88
		pCi ^{137}Cs/kg grain									
1962	Rye	570	553	734	446	—	374	486	442	—	610
	Barley	326	385	480	480	500	393	318	354	390	534
	Wheat	—	—	445	415	690	278	238	—	278	342
	Oats	164	166	420	—	805	224	366	—	—	248
1963	Rye	985	1130	775	2300	1200	1060	1910	1225	—	715
	Barley	930	1080	905	1220	898	2120	785	620	1430	530
	Wheat	—	—	812	1780	725	715	533	—	670	685
	Oats	1265	552	760	1520	562	466	1068	758	955	582
1964	Rye	1080	1040	777	1070	944	579	575	405	—	662
	Barley	485	643	434	715	872	408	475	282	360	415
	Wheat	—	—	475	654	—	360	334	344	268	344
	Oats	497	700	309	614	491	297	410	394	294	384

Table 5. *pCi* ^{54}*Mn/kg Danish Grain,* 1963–4

Location		Tl	S	Ø	As	J	B	Ts	V	Ab	Å
Year	Sort										
1963	Rye	1785	3290	1495	3640	2880	1630	1810	1330	—	955
	Barley	1290	3130	1775	2320	1080	2545	1320	2000	1210	945
	Wheat	—	—	1885	1080	1260	1350	1085	—	1100	1040
	Oats	2760	1530	1780	1425	1850	1310	1130	1425	1215	1020
1964	Rye	500	753	340	482	189	294	334	242	—	219
	Barley	458	671	435	682	268	249	364	204	373	246
	Wheat	—	—	253	256	—	192	173	172	198	190
	Oats	424	426	339	288	496	133	286	268	305	226

Table 6. *Summary of the Significance Test from the Analysis of Variance derived from the Logarithm of the Figures in Tables* 1–5

Effect	Source	pCi ^{90}Sr/g Ca (cf. Table 1)	g Ca/kg (cf. Table 2)	mg Sr/g Ca (cf. Table 3)	pCi ^{137}Cs/kg (cf. Table 4)	pCi ^{54}Mn/kg (cf. Table 5)
Main	Sort	xxx	xxx	xxx	xxx	xx
	Location	xxx	—	xxx	xxx	xxx
	Year	xxx	x	x	xxx	xxx
2-factor interaction	Sort · Loc.	xxx	—	—	—	—
	Loc. · Year	xxx	—	xxx	xxx	—
	Year · Sort	xx	—	x	—	—
Coefficient of variation (3-factor interaction and residual error)		0.25	0.22	0.30	0.26*	0.26
Degrees of freedom		202	103	63	41	22

*The errors for ^{137}Cs were not homogeneous for the three years considered; this does, however, not invalidate the significance of the main effects.

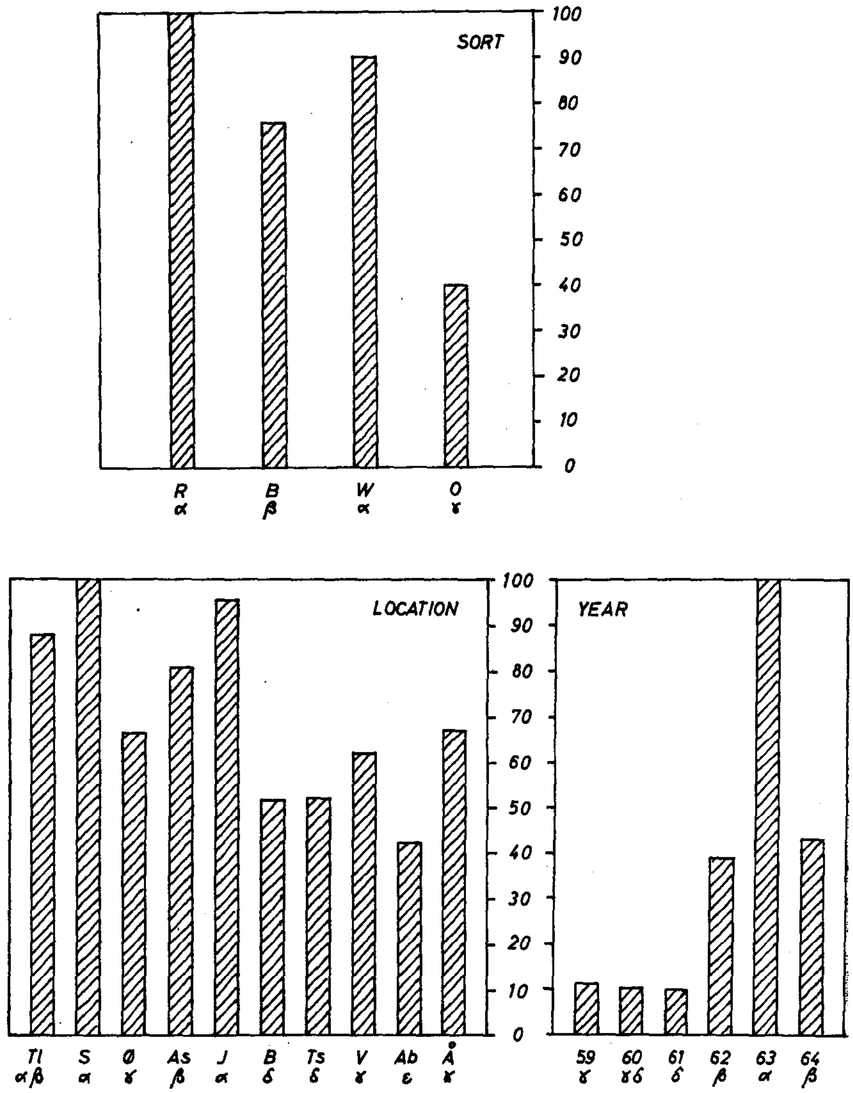

FIG. 3. Relative pCi ^{90}Sr/g Ca ratios in Danish cereal grain, 1959–64.

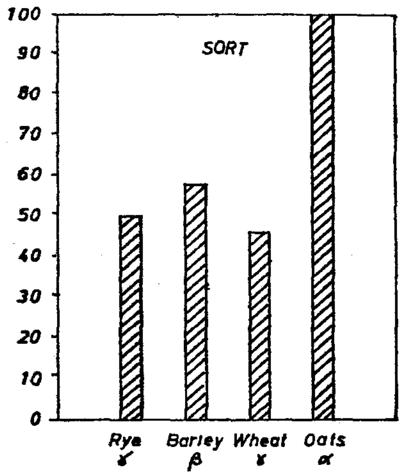

FIG. 4. Relative pCi ^{90}Sr/kg grain, 1959–64.

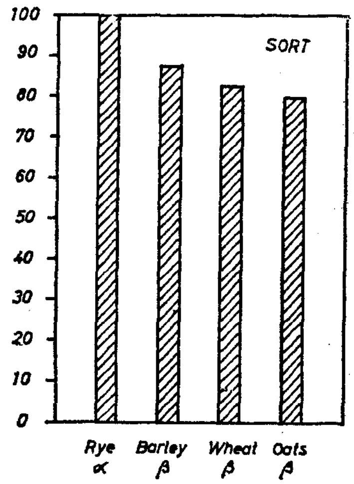

FIG. 5. Relative g Ca/kg grain, 1959–64.

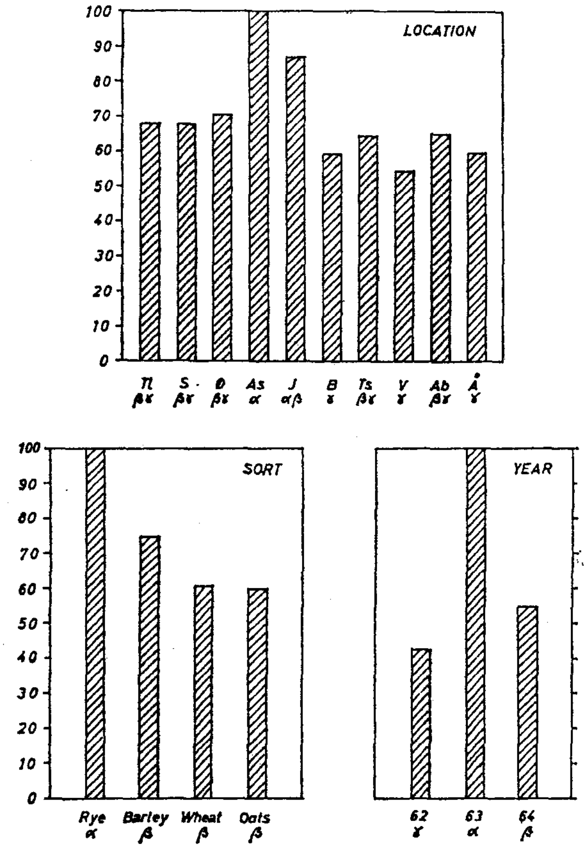

FIG. 6. Relative pCi ^{137}Cs/kg grain, 1962–4.

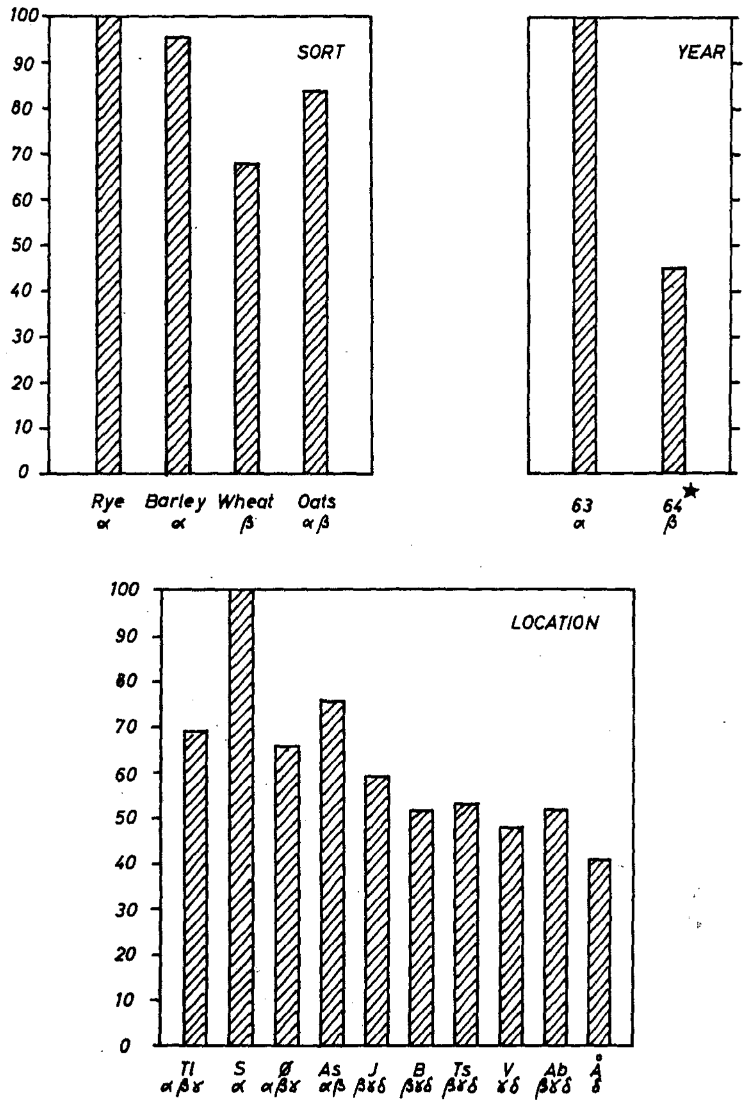

Fig. 7. Relative pCi ^{54}Mn/kg grain, 1963–4. *Corrected for decay back to the 1963 harvest.

DISCUSSION

Variation between species

Experiments with artificial application of activity carried out by Milbourn *et al.*[12] and Middleton *et al.*[13, 14] have shown that the radiostrontium from nuclear weapons tests has entered cereal grain mainly through direct absorption of the debris after the ears have emerged (floral absorption) and only to a minor degree through uptake from the soil. The direct contamination of cereal grain with ^{137}Cs in addition occurs to a significant degree before the emergence of the ears, because ^{137}Cs is easily translocated in the plants.[13, 14] As regards root uptake, this is normally even less for ^{137}Cs than for ^{90}Sr.[15]

An attempt can obviously be made to explain the higher levels of nuclear debris in rye than in wheat by the particular morphological and physiological conditions which make rye more sensitive than the other species of grain to floral contamination. The ears of rye are furnished with awns, and it seems reasonable to imagine that these awns would display some adsorptive effect on the direct fallout.

To test this hypothesis, ^{137}Cs was measured in samples of wheat with awns and without awns,* grown in the same field in 1965. The ^{137}Cs content in the wheat with awns was 50% higher than in the wheat without awns.

Another possible explanation for the high rye levels could be that rye has a relatively small grain size compared with the other species. According to Pedersen[16] the 1000 grain weight of rye is 25 g, whereas the corresponding weights of wheat, barley and oats are 40, 40 and 35 g respectively. Rye thus displays a greater surface-to-weight ratio than the other cereals and might consequently be more susceptible to direct contamination (Andersen[17]).

To examine this theory, fractions (separated according to grain size) of rye and wheat grown in the same area in 1964 were analysed. The S.U. level in small rye grains (15 g/1000 grains) was 18% higher than in large rye grains (37 g/1000 grains). Wheat showed a 26% higher S.U. level in a (25 g/1000) fraction than in a (42 g/1000) fraction. If, however, rye and wheat of similar grain size were compared, the S.U. level in rye was 20% higher than in wheat.

The conclusion of these experiments is that both awns and small grain size will increase the floral absorption of fallout nuclides in cereals, the presence of awns probably playing a more significant role than grain size.

The awns of barley are longer and thicker than those found in rye, but the activity levels in barley are lower than in rye. The difference in grain size between rye and barley (cf. above) would undoubtedly explain some of this difference in activity levels. It should, however, also be recalled that the period from the emergence of the ears to harvest is two or three weeks shorter for barley than for the other Danish grain species.[16] Barley is thus exposed to floral contamination for a shorter period than the other cereals. Hence it seems reasonable that barley shows lower levels than rye for this reason. That the awns on barley on the other hand play an important role for the direct contamination is indicated by the fact that the activity levels in barley in most cases exceeded those found in wheat and oats (Figs. 4, 6 and 7).

While the method of direct contamination of cereal grain with ^{90}Sr and ^{137}Cs is fairly well known,[13, 14] no experimental data has yet been available regarding ^{54}Mn. Recently Sutton and Kelly[18] have found in field surveys that the ratio of pCi ^{54}Mn to mg stable manganese is fairly constant in U.S. wheat grown during 1963, and that this ratio was nearly constant in the different milling fractions of a Kansas wheat sample. They concluded that uptake of ^{54}Mn in wheat had taken place through the root of the plants rather than as a result of the physical adhesion of inert radiomanganese.

From measurements of ^{90}Sr in precipitation and air and of ^{54}Mn in air,[3] it was estimated that the accumulated ^{54}Mn levels by May–August 1963 and 1964 were nearly equal in Denmark, whereas the ^{54}Mn fallout rates in these periods were approx. 10 times greater in 1963 than in 1964. The concentrations of ^{54}Mn in grain (Table 5) were on the average 5 times higher in 1963 than in 1964. To estimate the uptake of fallout nuclides deposited in the soil, barley was grown in 1964 in a greenhouse placed on a field contaminated during 1963 with ^{54}Mn fallout. It was not possible to detect

* Danish wheat normally has no awns.

any ^{54}Mn in the barley grain from the greenhouse, whereas barley grown outside the house, and thus exposed to the 1964 ^{54}Mn fallout, contained 390 pCi ^{54}Mn/kg grain. When these observations are compared with Sutton and Kelly one reaches the conclusion that if ^{54}Mn is taken up through the root system, it must in Denmark have been during a rather short time after the deposition of the ^{54}Mn, that is, probably before the radiomanganese had been fixed to the soil and thus been made unavailable to the cereal plants. Hence the contamination of Danish grain products with ^{54}Mn from nuclear debris depends upon the fallout rate rather than the accumulated fallout.

Variation between locations

A comparison of Fig. 2 with Fig. 3 shows that there exists a correlation between S.U. in grain and the yearly mean amounts of precipitation at the different locations. The locations S, As, J, and Tl show higher levels than Ts, Ab, B, and Å, both as regards S.U. and mm precipitation. However, the variation between pCi ^{90}Sr/g Ca seems to be greater than the variation between amounts of precipitation. The greater difference between S.U. values might be a result of the fact that the soil in the western part of the country is more sandy than the soil in East Denmark (Fig. 1), and that the sandy soil contains less Ca and probably shows a greater mg Sr/g Ca ratio than the clayey soil, and thus favours the root uptake of strontium.

To examine this phenomenon two sets of soil samples were collected, one in Jutland at location S, and one in Funen at B. The samples were collected as double samples and taken in five 25 cm long cores down to a depth of 125 cm. Calcium and stable strontium were determined in the 20 cores by HCl extraction.

The total Ca mean content of the B soil (down to 125 cm depth) was 33 g/kg and in the S soil the level was 0.45 g Ca/kg. The top soil (0–25 cm) at B contained 6 g Ca/kg, and at S 1.3 g Ca/kg soil. As regards mg Sr/g Ca it was also possible to prove significant variation between locations. The soil from S contained 4.4 mg Sr/g Ca and the soil from B 53% of this value, i.e. 2.3 mg Sr/g Ca.

In Fig. 8 the relative amounts of mg Sr/g Ca

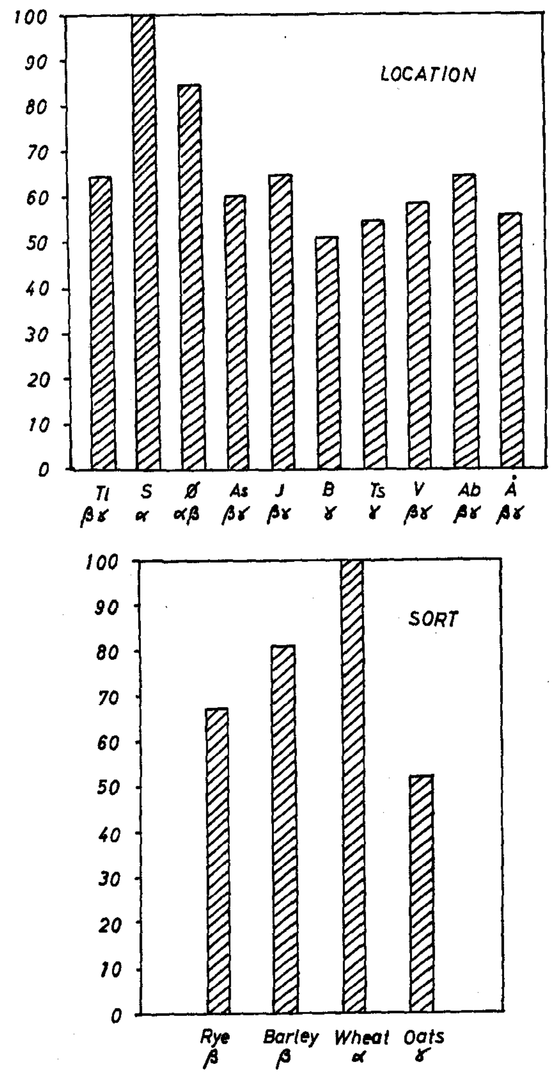

FIG. 8. Relative mg stable Sr/g Ca in Danish grain, 1959, 1962–4.

in grain are plotted. At location B the ratio is 51% of the ratio found at S, i.e. essentially the same as found for the soil samples. Hence it seems reasonable to expect that the S.U. level in grain, at least to some degree, depends upon the stable strontium-to-calcium ratio in the soil.

Figure 8 shows further that the mg Sr/g Ca ratio in the four species of grain differs significantly. Wheat appears to have the highest ratio, oats the lowest. Rye and barley were only probably significantly different.

Variation between years

A comparison of Fig. 9 and Fig. 3 shows that there is a pronounced correlation between air activity in August and the S.U. level in cereals.

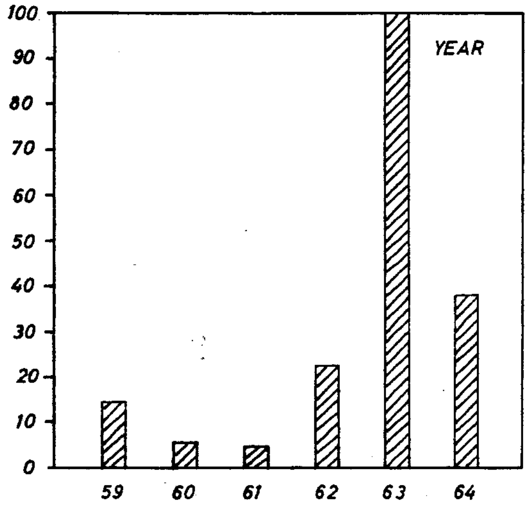

FIG. 9. Relative pCi $^{90}Sr/m^3$ air in August at Risö, 1959–64.

The grain activity, however, is to some degree also dependent on the amount of precipitation in August. This explains why the grain levels in 1959, as compared with 1960 and 1961, were relatively low, and why the 1962 S.U. level was nearly equal to the 1964 level (the amount of precipitation in August 1959, as well as in August 1964, were rather low).

Variation between spring and winter varieties

Wheat and rye grown in Denmark are normally winter varieties, i.e. they are sown in September and harvested in August–September the following year. To a certain extent, however, spring varieties of rye, as well as of wheat, are also grown. These varieties are sown in the early spring, at the same time as barley and oats, and harvested a little later than the winter varieties. It was thus of interest to investigate whether the different growing periods of the two varieties were detectable through their respective activity levels.

Table 7 shows the levels of ^{90}Sr and ^{137}Cs in spring varieties of wheat and rye collected in 1962–4. If these figures are compared by a *t*-test with the corresponding figures in Tables 1 and 4 for winter varieties, it is not possible to

Table 7. *^{90}Sr and ^{137}Cs in Spring Varieties of Rye and Wheat,* 1962–4

Year	Sort	Location	pCi ^{90}Sr/g Ca	pCi ^{137}Cs/g K
1962	Wheat	Ts	251	—
	,,	Ab	215	—
1963	Rye	As	745	485
	Wheat	As	839	386
	,,	Ts	870	200
	,,	Ab	459	236
1964	Rye	S	789	407
	,,	As	831	242
	,,	J	685	312
	Wheat	Ø	419	92
	,,	As	549	195
	,,	Ts	293	84
	,,	V	251	115
	,,	Ab	158	67
	,,	Å	169	72

show a significant difference between winter and spring varieties.

Several authors [19, 20] have, in greenhouse experiments, found that significant differences exist in the root uptake of radiostrontium in grain among a wide range of genotypes in wheat, barley and oats. A field survey [21] carried out on six varieties of hard red winter wheat showed significant differences between the varieties, the upper levels being approx. 25% higher than the lower.

In the present study no attempts have yet been made to investigate the variation in activity content in grain with genotype. If, however, the variance of the ratios: S.U. spring var./S.U. winter var. from the different locations and years are compared by a F-test with the variance of the double determination of the samples, the two sets of variances were not found to be significantly different. Considering that the ratio: S.U. spring/S.U. winter not only includes the analytical error, but also the error from sampling, and differences arising from displaced growing periods and from dissimilarity in genotype, it does not seem likely that the latter are important for the activity contents in the present study. This is, however, probably valid only as long as the direct contamination of the crops is several times the contamination from root uptake, as was the case in 1962–4.

MULTIPLE REGRESSION ANALYSES OF THE S.U. FIGURES

Rivera [22] has calculated a formula for S.U. in the U.S.A. wheat grain collected in the period 1958–62:

$$V = 0.49\,S + 246\,R$$

where V is the production-weighted S.U. ratio in wheat grown in the United States during a given year, S is the estimated ^{90}Sr content of the soil on which the wheat was grown in mCi/km^2 for the middle of the crop year, and R is the fallout rate in mCi/km^2 deposited in June of the crop year. Another formula has been proposed by UNSCEAR [1] mainly based on wheat:

$$C = 0.5\,F_d + 20\,F_r$$

where C is pCi ^{90}Sr/g in the grain, F_d is accumulated deposition of ^{90}Sr (mCi/km^2) and F_r is the current rate of deposition (mCi/^{90}Sr/km^2/year).

The two equations are nearly identical, provided the fallout throughout the year is assumed to be twelve times that measured in June. This is clearly an overestimate of the annual fallout, and consequently it would be expected that the latter equation yields lower results than the former. It is thus not surprising that the UNSCEAR formula estimated the Danish S.U. levels in wheat to be approximately a factor of two lower on the average than those actually measured in 1962–5.

In the present study it has been found that the S.U. levels in grain seem to be lognormally rather than normally distributed. Further it has been found feasible to use air activity (pCi ^{90}Sr/m^3) and mm precipitation instead of directly measuring the value mCi ^{90}Sr/km^2, because the two first-mentioned variables gave a better fit to the grain data than the latter, when tested for the period 1962–4. As the variation in monthly mean air activity has been shown to be rather constant within an area as small as Denmark (44,000 km^2) (cf. e.g. U.K. air measurements [6]), it is only necessary to measure the air activity at one location. To determine the accumulated fallout, the mean value for the ten state experimental farms from the annual sampling in September has been used instead of individual values. This is because the contribution from the soil uptake in the period considered has been rather low, and because factors other than the accumulated fallout in the soil influence the root uptake (e.g. Ca-content of the soil and mg Sr/g Ca ratio), factors which probably conceal the effect of varying ^{90}Sr levels in the soil.

An equation of the following form was used:

$$\text{S.U.} = k_0\, A^{k_1} \cdot B^{k_2} \cdot C^{k_3}$$

or

$$\log \text{S.U.} = k_1 \cdot \log A + k_2 \cdot \log B + k_3 \cdot \log C + \log k_0$$

where S.U. is pCi ^{90}Sr/g Ca in the cereal grain, A is the mm precipitation in certain months (Table 8) near the harvest, B is pCi

$^{90}Sr/m^3$ in air during the same months, measured at one location (Risö), C is mCi $^{90}Sr/km^2$ accumulated in the soil by September of the crop year, calculated as the mean of the accumulated fallout at the ten state experimental farms. k_0 is a constant, and k_1, k_2 and k_3 are the partial regression coefficients.

Table 8 shows the partial regression coefficients, k_0, and the multiple correlation coefficients r, for the four grain species calculated for three different periods by multiple regression analysis using the data in Table 1 and ref. 3 and 7.

It is to be expected that Danish cereals would be exposed to direct contamination with ^{90}Sr predominantly during the period June–August (cf. Middleton[13, 14] and Pedersen[16]). In 1962, where fresh fallout began to appear in Denmark in August[23] the increase in the $^{89}Sr/^{90}Sr$ ratio took place during the Danish harvest period. From a greenhouse experiment it was found that approx. 10% of the ^{90}Sr in the grain of that year's harvest was due to root uptake. Hence it was possible from measurements of the $^{89}Sr/^{90}Sr$ ratio in the grain samples to estimate the age of the florally absorbed ^{90}Sr in the grain, and it appeared that the following equation fitted the data:

$$y = 100\ e^{-t}$$

where y was the accumulated percentage of total ^{90}Sr at harvest time in the grain from direct contamination at a time t-fortnights before the date of harvest. The equation indicates that the contribution of ^{90}Sr from fallout prior to one month (two fortnights) before harvest was less than 15% of the total direct contamination with ^{90}Sr. As the harvest for most Danish grain species normally occurs in the period August 15–September 15, the three contamination periods shown in Table 8 were selected for examination. It appears from the table that it is reasonable to regard August as the only month of significance for the direct contamination of Danish grain with radiostrontium in the period 1959–64. In this connection it should be emphasized that the regression equations indicated throughout Table 8 are valid only for the period and area considered.

The partial regression coefficients (k_3) in Table 8 were all significant, indicating that the

Table 8. Values and Significance (by t-test) of the Partial Regression Coefficients and the Multiple Correlation Coefficients for the Equation:

$$\log Y = \log k_0 + k_1 \log A + k_2 \log B + k_3 \log C$$

Period of direct contamination	Sort	Partial regression coefficients			$\log k_0$	Multiple correlation coefficient
		k_1	k_2	k_3		
July–Aug.	Rye	0.3937	0.6677xxx	0.3720	2.5182	0.94xxx
	Barley	0.4764x	0.4152x	0.9150xx	0.9289	0.91xxx
	Wheat	0.5204	0.5375xx	0.5582	1.6994	0.94xxx
	Oats	0.5931x	0.4202xxx	0.7373	0.6889	0.90xxx
Aug.	Rye	0.3710	0.6560xxx	0.4402	2.4768	0.95xxx
	Barley	0.4628	0.4048xx	0.9636xx	0.8865	0.92xxx
	Wheat	0.5028	0.5241xx	0.6211	1.6447	0.95xxx
	Oats	0.5809x	0.4019xx	0.8121xx	0.6042	0.91xxx
Aug.–Sept.	Rye	0.3323	0.8431xxx	−0.0306	3.6185	0.94xxx
	Barley	0.4876	0.6229xxx	0.3805	2.0825	0.92xxx
	Wheat	0.5337	0.7620xxx	−0.0167	2.9507	0.95xxx
	Oats	0.5930	0.6706xxx	0.0991	2.0986	0.91xxx

Table 9. *S.U. in Danish Grain in* 1965.

	Rye	Barley	Wheat	Oats
Calculated	258 ± 13	278 ± 18	256 ± 18	120 ± 10
Measured	238 ± 32	166 ± 26	227 ± 31	102 ± 12

The error term is the S.E. of the mean of the 10 locations.

air activity, in the period around harvest-time, plays an important role for the ^{90}Sr content of the grain. Accumulated fallout (k_3) seemed only to be of significance for the S.U. levels in barley and oats. It is, however, to be expected that the S.U. levels in grain in the coming years, provided the test-ban continues, will for all species depend increasingly on the accumulated fallout.

Although the equations are valid only for the period 1959–64, the 1965 S.U. levels were estimated from the August regression coefficients in Table 8. Table 9 shows the calculated results compared with those actually measured.

All calculated figures except barley agreed fairly well with the measured levels.

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