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RADIOCHEMICAL ANALYSIS OF PRECIPITATION, TAP WATER AND MILK IN NORWAY 1957 - 1958

METHODS, RESULTS AND CONCLUSIONS

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SUMMARY

Radiochemical investigations of precipitation, tap water and milk conducted at the Norwegian Defence Research Establishment, Division for Chemistry up to the end of 1958, are described.
Data for Sr-90, Sr-89, Cs-137 and I-131 are given and the theories for the distribution of these isotopes in the biosphere are discussed. An account is given of radiochemical experiments carried out and the methods of radiochemical analysis are given in detail in Appendix I and II.

1 INTRODUCTION

The series of nuclear weapon tests in later years has led to a world-wide distribution of radioactive fallout, and up to October 1958 about 3.2 megacuries of Sr-90 have been deposited, Alexander 1959 (1). According to information from USA (2) up to May 1959 some 65 megatons - TNT equivalent - fission energies have been exploded. This corresponds to 50 kg Sr-90, equal to 6 - 7 megacuries. Approximately 50 per cent of this amount is deposited on the earths surface. During the past 10 years extensive studies have been made of this artificially produced radioactivity. From USA, UK, USSR and other nations a great number of papers dealing with the general pattern of fallout have become available, see (3 - 7) and others. The main concern of these papers are the possible hazards of fallout. From this aspect it is of importance to know the conditions which determine the geographical distribution of fallout, the cumulation in water, soil and plants, the uptake in fish and grazing animals, and finally the transfer to the human population. There are obviously numerous ways in which this transfer takes place, and the relative importance of these will vary from one country to another.

Since the problem of radioactive fallout contamination is of a world-wide character and related to a number of widely different fields of science, from atom physics and meteorology to biology and genetics, the situation has called for international exchange of information and cooperation in a number of ways. As a result of this several thousand papers have appeared in the open scientific literature during the past years and the broad outlines of the problem are fairly well understood. A number of details are however, not yet fully understood, and it should be remembered that the particular problems of a selected region and its inhabitants do not necessarily find their solution within the framework of general information alone, but must depend upon investigation of local conditions. This is particularly so because the climatological conditions and also the ways of living are so different for example in this country compared to many others.

The general purpose of the investigations of radioactive fallout which have been carried out at the Division for Chemistry, Norwegian Defence Research Establishment, has been to determine by radiochemical methods the amount and distribution of those radio-active isotopes which are considered to constitute the most serious danger. The particular object of the present work has been to obtain technical information concerning the contamination in Norway from the bomb tests carried out so far. This information constitutes an important part of the basic knowledge needed to evaluate possible hazards from future atomic explosions. Another important aspect has been to establish an analytical laboratory with the necessary equipment and to train the personnel in the radiochemical methods which will be needed in case of an emergency.

In the present report data showing the content of Sr-90, Sr-89, Cs-137 and I-131 in precipitation, tap water and milk are given. Also an account is given of radiochemical experiments and studies of analytical methods, which have been carried out to establish routine methods for further work in this field. The analytical procedures are based upon known methods and apart from the method for determination of I-131 only minor modifications have been introduced by us. The detailed laboratory manuals are given in Appendix I. In Appendix II the methods used for standardization

are described. Most of the analytical data have previously been published without comments in short communications from this laboratory (8). In the present report they are given with our full comments, and some conclusions which only can be drawn from an evaluation of the complete material are given.

2 SAMPLING AND PREPARATION

At the start of this work in 1956 the knowledge of artificial radioactivity in milk and water in Norway was scanty and the few data available indicated very low activities of Sr-90 and Cs-137.

With the available counting equipment, activities as low as 1 cpm could be determined with a statistical accuracy of ± 20 per cent. Each sample had to be sufficiently large to give at least this activity in the final preparation.

Preliminary tests showed that 3 - 5 l of milk, 5 - 10 l of rain water and 50 l of tap water were required for Sr-90 and Cs-137 determinations, whilst I-131 could be determined in half a liter of milk.

It was apparent that the large samples needed concentration treatment before radiochemical analysis could be started. Because of the large number of samples which were planned, the methods for concentration had to be rapid and convenient. A description of the sampling and concentration methods which were tried, is given below.

2.1 Sampling of water and snow

a) Tap water has been sampled from three localities, Bergen, Stavanger and Kjeller in Norway, see map figure 2.1. The first sampling attempted was to collect batches in iron drums or glass flasks. The iron drums were painted inside with laquer and the glass containers were treated with silicone. Provided sufficient amount of carrier was added no activity seemed to be lost in these vessels. For continuous sampling of water, a distillation still has been used, figure 2.2. The apparatus swings freely about the fulcrum a. The Y-tube is a bypass and the cooling water is

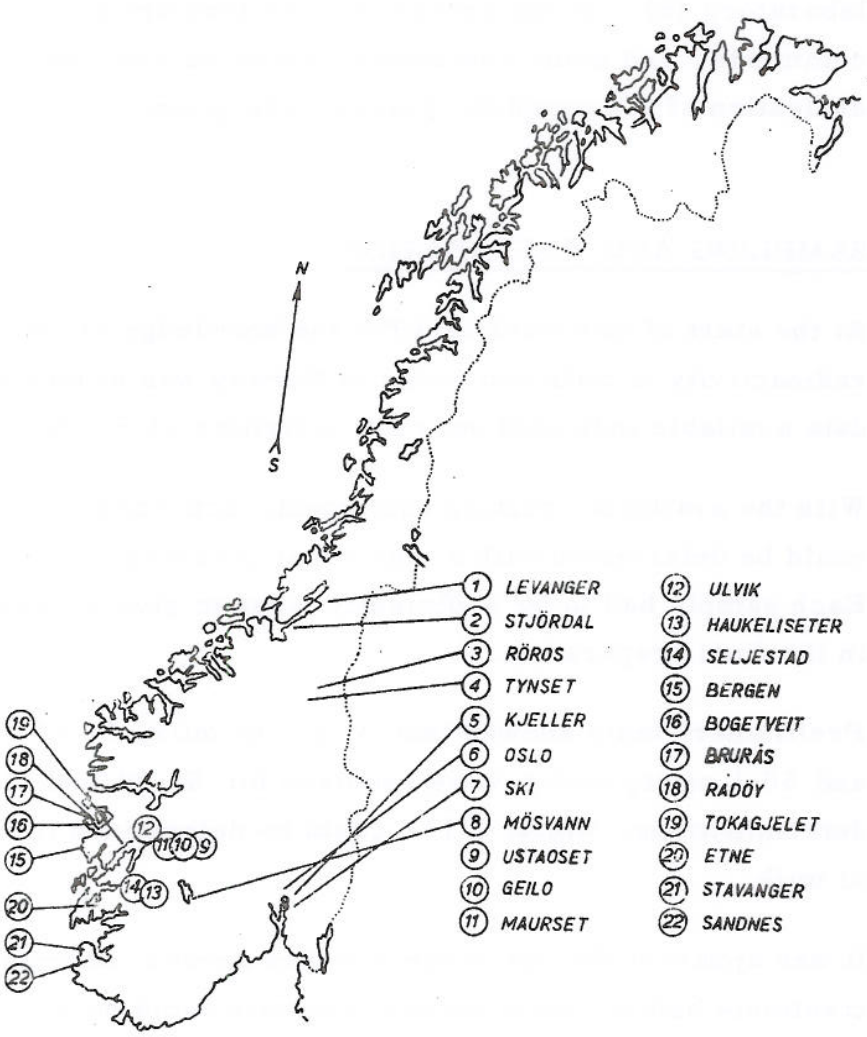

Figure 2.1 Map of Norway with sampling points indicated

adjusted with the screw clip c. The tilt of the apparatus controls the feeding into the distillation flask. The heating coil - 900 watts - is placed in a quartz tube which goes through a metal lid sealed with O-rings. Carrier is added before distillation starts. This arrangement functions without attention and has proved reliable if the water pressure does not vary too much.

b) Rain water has been sampled at Bergen, Møsvann, Kjeller and Ski, see map figure 2.1. Rain is collected through polyvinylchloride funnels into polythene flasks. The collecting area is 1 m^2 for two localities, Ski and Møsvann, at the sampling station in Bergen the area was cut to 0.3 m^2. At Kjeller the

collecting vessel has been a rectangular stainless steel tray of 2 m^2 area. The amount of precipitation is checked by means of an ordinary precipitation collector as used by The Norwegian Meteorological Institute.

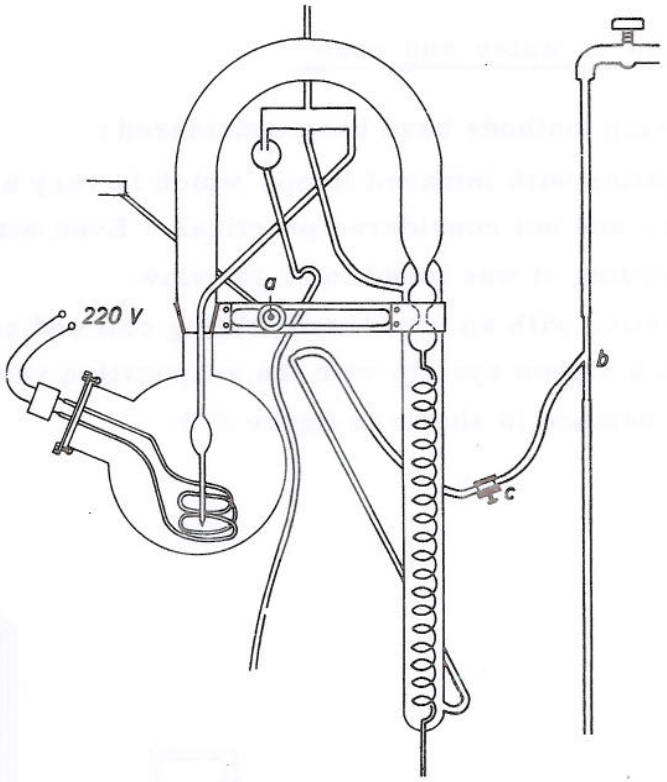

Figure 2.2 Apparatus for continuous distillation of tap water

c) Snow has been sampled at Ski and Mφsvann, see map figure 2.1. This sampling offered serious difficulties, since snow is easily carried about by winds and whirled out of the collecting vessel as well as carried into it from the ground.

The purpose of the meteorological measurement of precipitation is to give a true value of the amount of rain and snow. Our object of sampling precipitation has been to obtain cumulative data for radioactive fallout on the ground. For this purpose snow samples have been collected in fairly flat fields with trees growing round the edges to avoid winds mixing the snow layers. The collecting vessels consist of 1 m^2 wooden frames, 5 cm high, over which

a polythene foil has been fixed so as to form a tray inside the frame. They are placed on the ground before the first snowfall. The polythene foil stops possible drainage due to thawing periods which mays occur in midwinter. The snow of each month was separated with colored perlon thread markers, and for each month during the winter season two frames were collected.

2.2 Preparation of water and snow

The following methods have been considered:

a) Evaporation with infrared lamps which is very useful for small samples was not considered practical. Even with a Mariotte flask feeding it was found to be too slow.

b) Evaporation with an immersed heating coil and continuous feeding through a siphon system with the evaporation vessel placed on a simple balance is shown in figure 2.3.

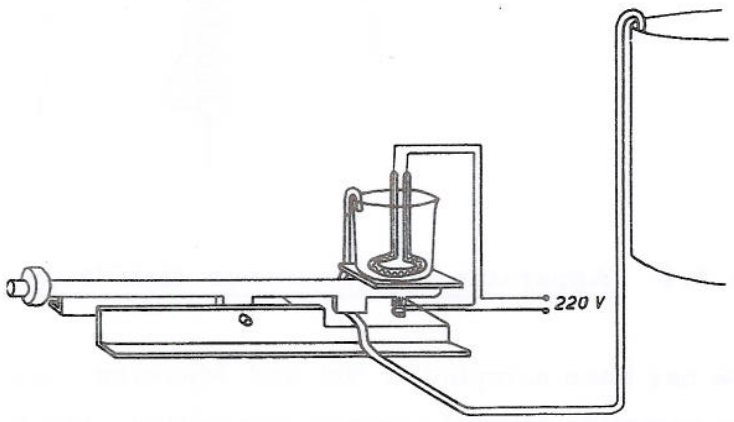

Figure 2.3 Evaporation of large water samples with continuous feeding

The feeding rubber tube is squeesed by a lever when the water rises above a predetermined level. A simple switch turns off the current to prevent the heating coil going dry. With this equipment it was possible to evaporate 50 l in less than two days with very little attention. Loss by sputtering was prevented by a stainless steel gauze placed on top of the beaker.

c) Scavenging with calcium phosphate, this method was suggested by J H Harley of the US AEC Health and Safety Laboratory. The method gives a very good carrier recovery of all isotopes tried including Cs-137. In a test on tap water containing 3.4 pc Sr-90/l ($p = 10^{-12}$) a second precipitation showed no activity. The method is outlined as follows: 2 g of calcium phosphate is added to the water sample, and pH is adjusted to 8 with ammonia. The sample is heated to 70°C and filtered. With large samples the filtration is rather troublesome even with the use of filter aid. Either the filtrate is not clear or the filter clogs up completely and has to be changed frequently. The filtration of 50 l often took 2 - 3 days of continuous attention.

d) Continuous extraction of Sr and Cs from tap water in a cation exchange column was tried. The column was connected to the tap through an overflow arrangement to eliminate variations in water pressure. This method was also attempted with 50 l batch samples using syphon feeding. The flow rate in both cases was about one liter per hour. No attempt was made to add carrier solutions to the tap water, but Sr-, Cs- and Ce-carriers were added to the batch samples. A few experiments showed, however, that quantitative capture of Sr-90 did not occur with this flow rate. When tap water was run through two identical columns in series, it was found that the first column had captured Sr-90 equivalent to 2.6 pc Sr-90/l, and the second 1.0 pc Sr-90/l. The procedure for batch sampling was judged according to carrier recovery. This was poor, in 34 experiments recovery varied from 35 to 55 per cent.

e) A better procedure for preparation of batch samples has since been adopted. Carrier solution containing 20 - 30 mg each of Sr, Cs and Ce is mixed with the sample. Vigorous mechanical stirring is applied and 15 g Dowex-50 cation exchanger in the H-form is added. Batches of up to 10 l require 2 hours stirring, see figure 2.4, while 50 l batches are stirred for 15 - 20 hours. The ion exchanger is easily recovered by decantation and is eluated with three portions of 6 M HNO_3. Ashing of the ion exchanger has been tried, but this offers no advantages. On the contrary, the ash sometimes gives a residue which is not soluble in acid and therefore has to be fused with soda. This complicates the procedure considerably. Several batch samples have, after

ion exchange treatment, been evaporated, but no Sr-90 or Cs-137 has been found in the residue.

f) Continuous distillation as previously described has been tried and gives good results for tap water concentration.

In table 2.1 the experience from testing of different concentration methods for water samples is summed up.

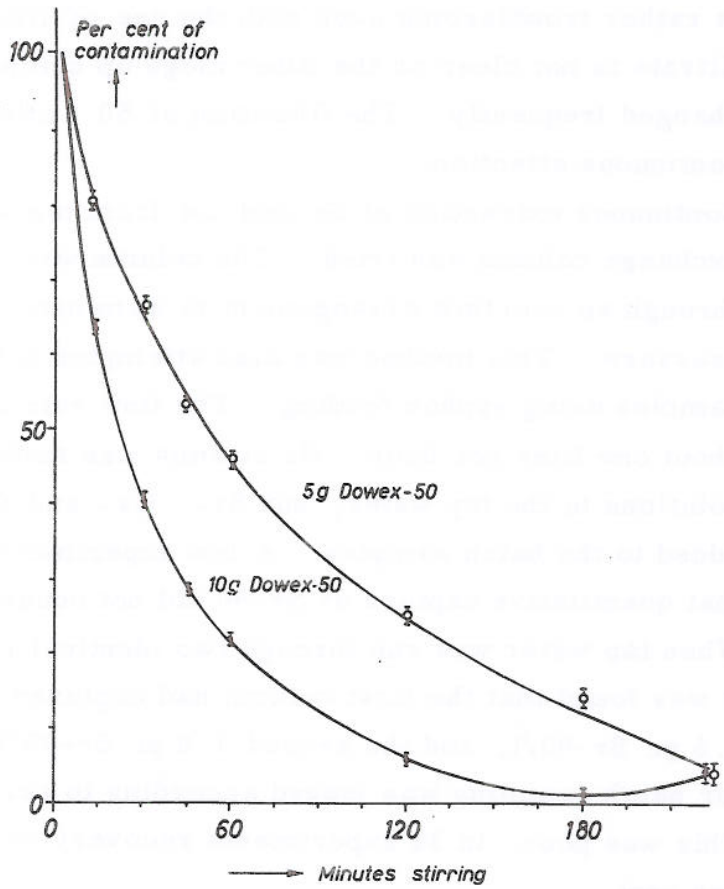

Figure 2.4 Removal of radioactivity from water with Dowex-50, Sr-89 and Sr-carrier added

2.3 Sampling and preparation of milk

Milk samples have been collected from Levanger, Tynset, Røros, Lillestrøm, Oslo, Sandnes and Bergen, see map figure 2.1. Both wet and dried milk have been sampled. Preliminary investigations showed that halfliter samples were sufficient for the determination of I-131. Samples of 3 - 5 l or the equivalent amount of dry milk were required for the determination of Sr-90, Sr-89 and Cs-137.

Sample treatment	Power required for 50 l sample	Attention	Time needed for 50 l sample	Carrier recovered Per cent	Used for
a) Evaporation with infrared lamps	50 kWh	Every 2 - 5 hrs	3 days	60 - 90	Small samples
b) Evaporation with immersed heating coil	50 kWh	Every 4 hrs	2 days	60 - 80	Large batches
c) Coprecipitation with calcium phosphate	Heating to 70°C	Continuous during filtration	3 - 6 days	70 - 99	Large batches Field method
d) Ion exchange column	Evaporation of eluate	15 min	3 - 4 days	35 - 55	Large batches
e) Batch sample stirred with ion exchanger	Stirring	No	15 - 20 hrs	60 - 80	Large batches Small batches
f) Continuous distillation	50 kWh	Twice a day	36 - 48 hrs	70 - 90	Tap water

Table 2.1 Concentration methods for water samples

The following procedures have been applied :

a) Ashing of 0.5 kg dry milk at 450°C. This takes 12 hours, and has been carried out in quartz vessels of one liter volume. A number of simple muffle furnaces were built for this purpose.

b) Wet milk with carrier added was evaporated to dryness in the quartz vessel by means of a hot plate and infrared lamps in the fumehood and then ashed. The evaporation is a time consuming procedure, but does not require much attention. A faster method would, however, be preferable.

c) The time of evaporation may be reduced by precipitation of

calcium and proteins followed by evaporation of the remaining liquid. This method has been tested with added Sr-89 and Cs-137, and some results are given in table 2.2.

Isotope added	Precipitate	Agent	Total activity in precipitate Per cent
Sr-89 + 100 mg Sr	Ca and Sr	Am-oxalate	71
Sr-89 + 100 mg Sr	Proteins	Acetic acid	0
Sr-89 + 100 mg Sr	Ca and Sr Proteins	Am-oxalate Acetic acid	98

Table 2.2 Sr-89 precipitation in milk

Cs-137 is not precipitated by these methods. The disadvantage of the method is that it requires attention and manipulation. If Sr-90 only is to be determined, precipitation with ammonium oxalate may be recommended.

d) Much work has been carried out to adapt electrodialysis for extraction of ions from milk. A modified apparatus based on Løddesøl's model, figure 2.5, has been found to work well with milk volumes up to one liter. The results of some experiments with isotopes added are shown in figure 2.6. Addition of HCl speeds up the extraction. For larger volumes successful experiments have been carried out by Holager (9). These experiments have also been performed with milk from a cow fed with Sr-89. The results are similar to those given in figure 2.6 and prove that all strontium in the milk is removable by electrodialysis, regardless of whether the radiostrontium has passed the metabolic chain or not.

e) The use of ion exchanger for milk is possible, but not very profitable due to the large amounts of Ca and K present.

f) A very convenient method is to use a laboratory spray dryer to produce dry milk from wet samples. Such equipment has recently become available to us, and spray drying is to day our standard

procedure. The dry milk is ashed and divided into two samples. One is used for radiochemical analysis and the other for Cs-137 determination by gamma spectrography.

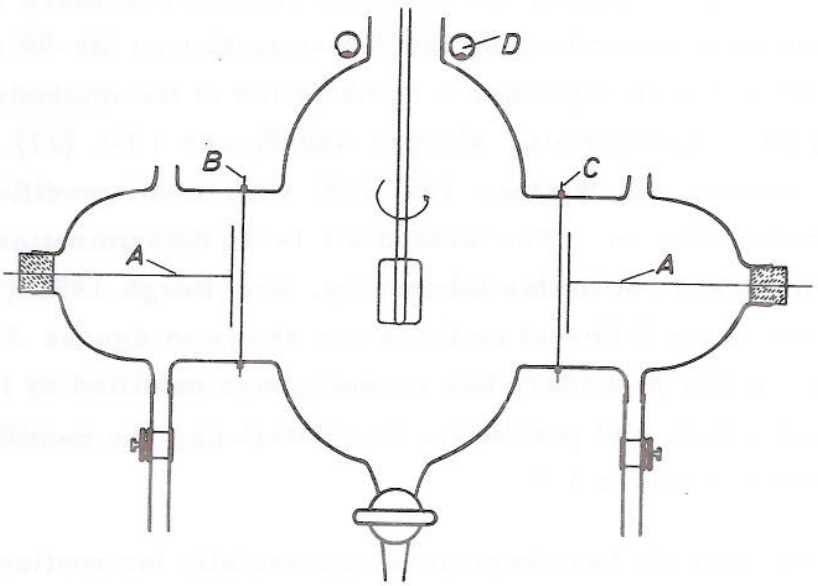

Figure 2.5 Apparatus for electrodialysis of milk samples
A - Pt-electrodes, B and C - membranes,
D - water cooling

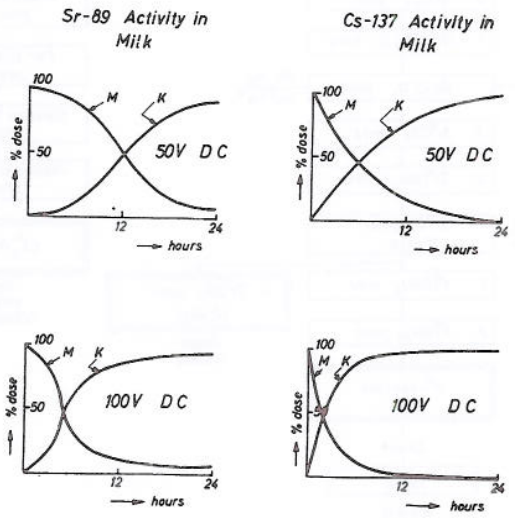

Figure 2.6 Electrodialysis experiments with Sr-89 and Cs-137 in milk
M - activity of milk, K - activity of kathode solution

3 RADIOCHEMICAL AND CHEMICAL PROCEDURES

In Appendix I the radiochemical procedures used for routine analysis of Sr-90, Sr-89, Cs-137 and I-131 in water and milk are given in detail. The methods are based on current literature (10 - 26). The procedures described for the determination of Sr-90 and Cs-137 in water and milk represent a combination of the methods given by Bryant, Chamberlain, Morgan and Spicer 1956 (27) and 1957 (28), and Harley and Whitney 1957 (29) with minor modifications introduced by us. The method for I-131 determination in milk has been worked out in this laboratory, see Bergh 1958 (30). Flow sheets of the different methods are shown in figures 3.1 - 3.4. The Cs-137 procedure has recently been modified by leaving out the cobaltinitrite and perchloric precipitations, the modified procedure is given in figure 3.3.

These methods have been used successfully for routine analysis in our laboratory through 1957 and 1958.

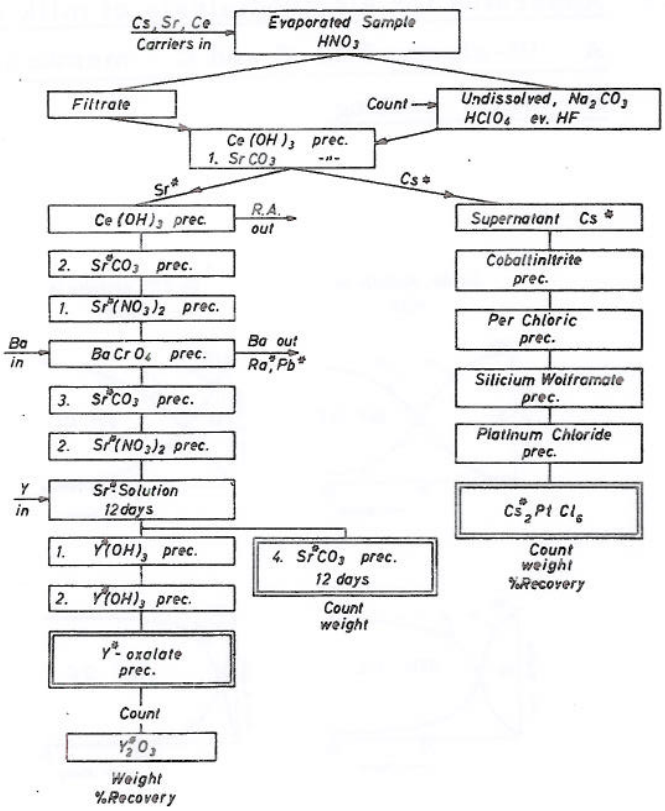

Figure 3.1 Flow sheet for the determination of Sr-89, Sr-90 and Cs-137 in water

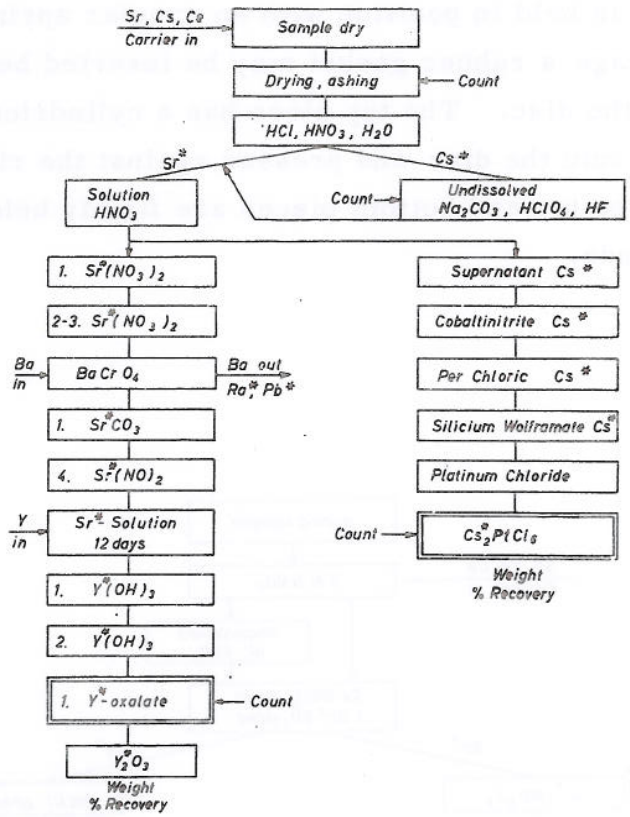

Figure 3.2 Flow sheet for the determination of Sr-89, Sr-90 and Cs-137 in milk

3.1 Filtering equipment

The final step of a radiochemical procedure usually is the filtration of a precipitate, and the transfer of this to a counting disc. To reduce the number of operations and increase the uniformity of the precipitations, a filtering procedure with a special type of filterstick has been used. A filterstick which was used at AERE, UK has been modified so that the filter support fits into the counter to avoid transfer of filter and precipitate from one disc to another. The filter assembly, shown in figure 3.5, consists of two cylindrical parts. They are made of perspex, which is sufficiently resistant to alcohol, and offers advantages over teflon in price and availability. The bottom piece fits into the ground joint of a suction flask. The upper part of it is shaped as an annular seat,

corresponding to the flange of the filter support. The combined filter support and counting disc is made of polythene. The disc is shown in figure 3.6, it has 12 - 14 holes of 1.5 mm diameter per cm^2, and is held in position with an annular spring. In order to avoid leakage a rubber gasket may be inserted between the filter-stick and the disc. The top piece has a cylindrical protrusion which fits into the disc and presses against the rim of the filter paper. The top and bottom pieces are firmly held together with rubber bands.

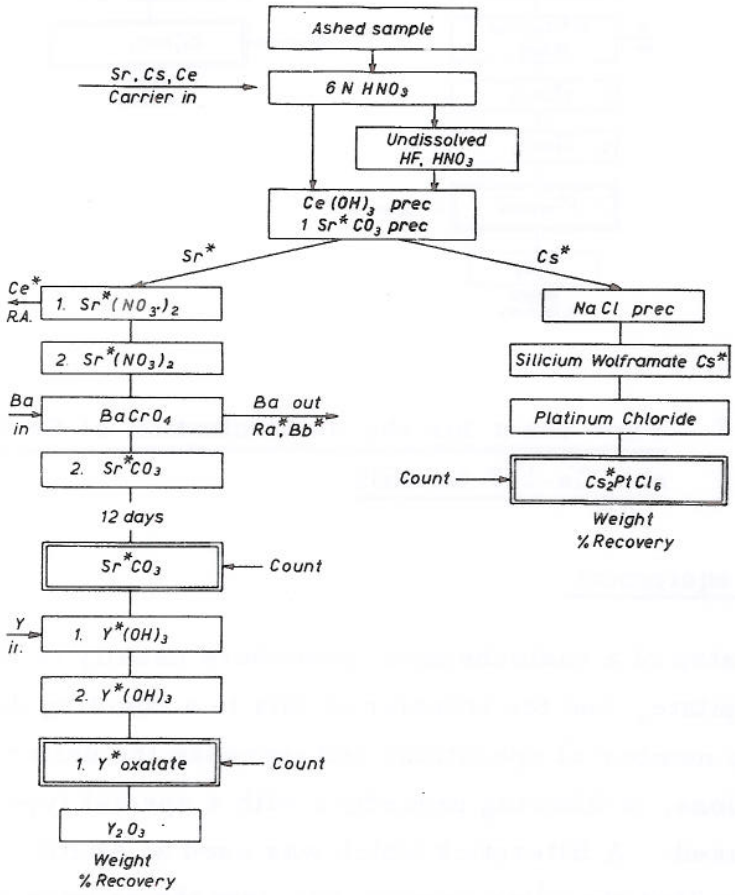

Figure 3.3 Modified method for the determination of Sr-89, Sr-90 and Cs-137 in water, milk, plant and peat soil

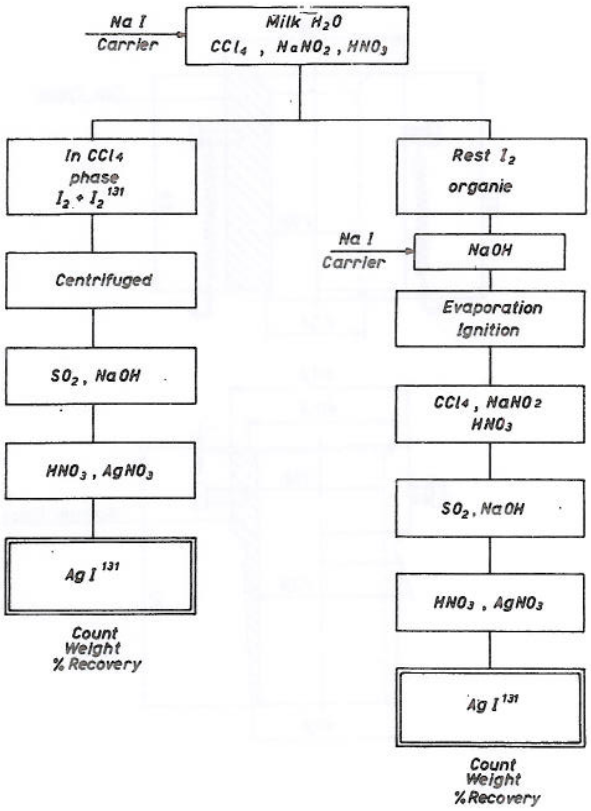

Figure 3.4 Flow sheet for the determination of I-131 in milk

3.2 Testing of methods

The analytical procedure for Sr-90 and Cs-137 determination has been checked using Sr-89 and Cs-137 as tracers. The isotopes were added as carrier-free solutions. A similar investigation has been carried out with I-131 in milk. A total of 1260 net cpm of Sr-89 was added to distilled water and the activity measured in all precipitates and filtrates throughout the entire procedure. The distribution of the Sr-89 tracer is given in table 3.1.

The final $SrCO_3$ precipitate contained 60 per cent of the added Sr-89. Some 28 per cent of the radioactivity was lost in the second $SrCO_3$ and $Sr(NO_3)_2$ precipitates. Corrected for recovery of added Sr-carrier, however, some 93 per cent of the radioactivity was accounted for in the analysis.

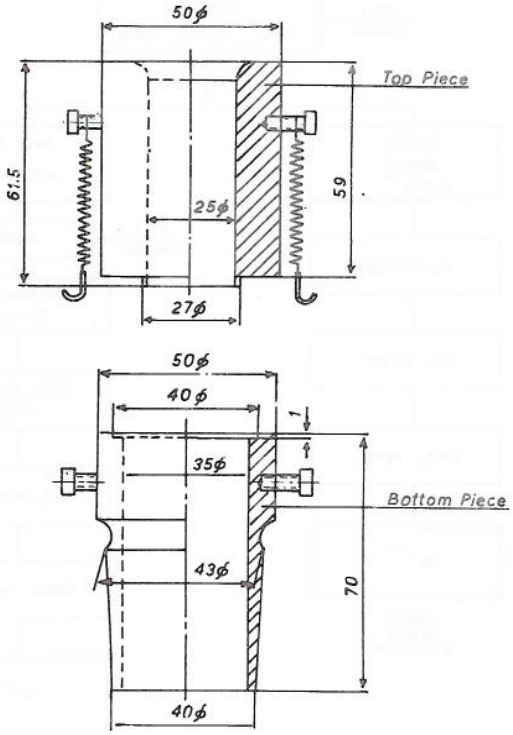

Figure 3.5 Filterstick, all measurements given in millimeters

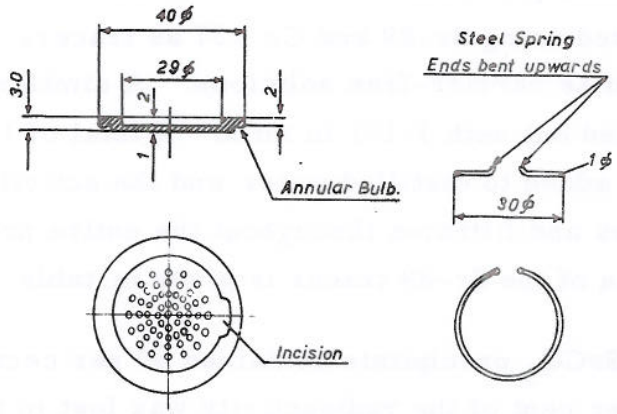

Figure 3.6 Combined filter and counting disc, all measurements given in millimeters

Material		Sample blank	Sample with Sr-89	Sr-89 found per cent
$Ce(OH)_3$	mg	27.1	26.4	
Ce	mg	19.9	19.4	
net cpm		0.1	76.2	6.1
$BaCrO_4$	mg	36.7	36.6	
Ba	mg	19.9	19.9	
net cpm		0.1	71.6	5.7
$Y_2(C_2O_4)_3$	mg	32.5	30.6	
Y	mg	9.6	9.8	
net cpm		0.0	1.4	0.1
Cs_2PtCl_6	mg	11.6	20.3	
Cs	mg	4.6	8.0	
net cpm		2.6	3.6	0.3
Supernate 1 $SrCO_3$	mg	817.5	946.8	
net cpm		0.0	1.1	0.1
Supernate 2 $SrCO_3$	mg	772.5	835.0	
net cpm		2.5	132.0	10.5
Supernate 1 $Sr(NO_3)_2$	mg	19.9	12.1	
net cpm		0.0	1.0	0.1
Supernate 2 $Sr(NO_3)_2$	mg	16.4	9.5	
net cpm		0.0	222.6	17.7
$SrCO_3$	mg	21.2	21.0	
Sr	mg	12.6	12.5	
net cpm		0.0	755.5	60.0
Total net cpm		5.2	1265.0	Sum Sr-89 99.8 per cent
Calculated from recovery Sr-89			1173.1	$SrCO_3$ 93.1 per cent

Table 3.1 Distribution of Sr-89 in the analytical procedure

To test the Cs procedure, a total of 1430 net cpm of Cs-137 was used. The distribution of the Cs-137 tracer is given in the table 3.2.

Material		Sample blank	Sample with Cs-137	Cs-137 found per cent
$Ce(OH)_3$	mg	26.0	25.4	
Ce	mg	19.1	18.6	
net cpm		0.0	0.9	0.0
$BaCrO_4$	mg	41.9	39.1	
Ba	mg	22.7	21.1	
net cpm		0.8	0.8	0.0
$Y_2(C_2O_4)_3$	mg	34.4	33.3	
Y	mg	10.1	9.8	
net cpm		0.0	0.2	0.0
Cs_2PtCl_6	mg	34.7	31.0	
Cs	mg	13.7	12.2	
net cpm		0.1	827.8	56.7
SiW prep.	mg	124.7	121.7	
net cpm		0.0	89.6	6.1
$SrCO_3$	mg	33.8	33.1	
Sr	mg	20.1	19.7	
net cpm		0.5	0.4	0.0
Supernatant from $SrCO_3$	mg	663.1	893.2	
net cpm		0.5	40.0	2.7
Supernatant from Co-prep.	mg	671.9	574.3	
net cpm		0.4	470.0	32.2
Total net cpm		2.3	1429.7	Sum Cs-137 97.7 per cent
Calculated from recovery Cs-137			1357.0	Cs_2PtCl_6 92.9 per cent

Table 3.2 Distribution of Cs-137 in the analytical procedure

The final Cs_2PtCl_6 precipitate contained 57 per cent of the added Cs-137 activity. More than 30 per cent was lost in the supernate from the cobaltinitrite precipitation. Corrected for recovery of added Cs-carrier, however, approximately 93 per cent of the radioactivity was accounted for. Due to the heavy loss of Cs-activity in the cobaltinitrite and to obtain some short cut, the method was modified as shown in figure 3.3.

I-131 determination in milk has been checked by adding known amounts of I-131 to the milk. Recovery data are given in table 3.3.

No.	I-131 pc added	I-131 pc found	Recovery per cent
1	63	61	96.8
2	102	101	99.6
3	275	270	98.5
4	430	410	95.3
5	941	878	93.3
6	2691	2484	92.3
		Average recovery	96.0

Table 3.3 Recovery of added I-131 to milk

To test the recovery also in vivo, I-131 was fed to a cow in sufficient amounts to give milk in which the activity could be measured directly with a dipping counter. This activity measured directly was compared with the results from the analytical procedure. Satisfactory agreement between the two methods was found.

A further check of the Sr-90 determination has been made by exchange of standard samples with US AEC Health and Safety Laboratory. Satisfactory agreement between the determinations of this laboratory and ours has been found.

3.3 Calcium in strontium carbonate

A possible source of error in the strontium carrier recovery is contamination of the final strontium carbonate with calcium. Strontium carbonate precipitates have therefore been analyzed spectrographically on calcium.

The working curve was made up with the line Ca-4289.4 against Sr-4361.7 as internal standard.

About 200 final strontium precipitates have been analyzed, and all showed less than 0.3 per cent calcium, this proves that the high carrier recovery is not caused by co-precipitation of calcium.

3.4 Natural strontium in milk

Since the recovery of strontium carrier in our separations was relatively high, it occured to us that this might be due to the presence of natural strontium in milk. To eliminate this possible source of error, a quantitative spectrographic determination of natural strontium in milk ashes has been performed.

The working curve was made up for the range 0.001 - 1.0 per cent Sr with the line Sr-4607.3 against Ca-4581.4 as internal standard. Calibration standards were prepared by adding known amounts of $SrCO_3$ to a matrix of the following composition:

$Ca_3(PO_4)_2$	K_2CO_3	NaCl	MgO	Fe_2O_3	
51.5	35.5	10.5	2.2	0.3	per cent

The results show, see chapter 7.2, that the content of natural strontium varies from a few tenths of a mg to several mg's per liter of milk. According to this the recovery has been corrected whenever the amount of natural strontium exceeds 0.5 mg Sr/l. In chapter 7 the measured values are given together with a discussion of the correlation between the contents of natural strontium and Sr-90 in milk.

4 INSTRUMENTATION

4.1 Automatic GM counting equipment

The basic instruments used throughout the present work have been two automatic sample changers in connection with Tracerlab scalers and printing timers. A block diagram of the layout is shown in figure 4.1. (Detailed wiring diagram for the automatic relay unit and workshop drawings of the sample changer are available upon request.) In figures 4.2 and 4.3 photographs of the instruments are reproduced.

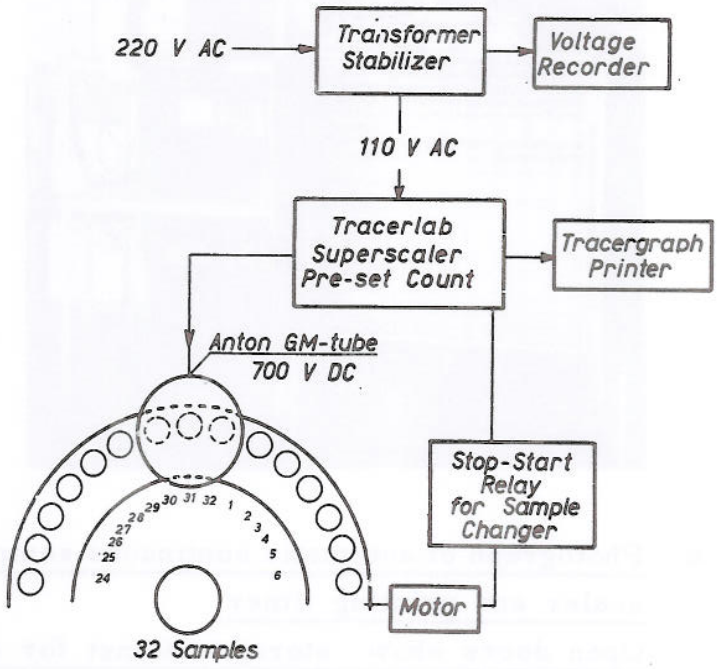

Figure 4.1 Diagram of continuous automatic counting equipment

The instruments have been used with the GM tubes, TGC-2 and Anton 1007 T. Both tubes have window thickness less than 2 mg/cm^2. The distance between specimen disc and endwindow has been 2 mm with a reproducibility of 0.02 mm. This has been achieved by an accurate working of the wheel and use of standardized polythene specimen discs. The distance between specimen and endwindow

is thus only dependent on the thickness of the sample. The sample changers have been used continuously for two years, enabling us to measure radioactivity in 24 hours a day over a 7 day week. The instruments have timed a preset number of counts, customarily set to 1000.

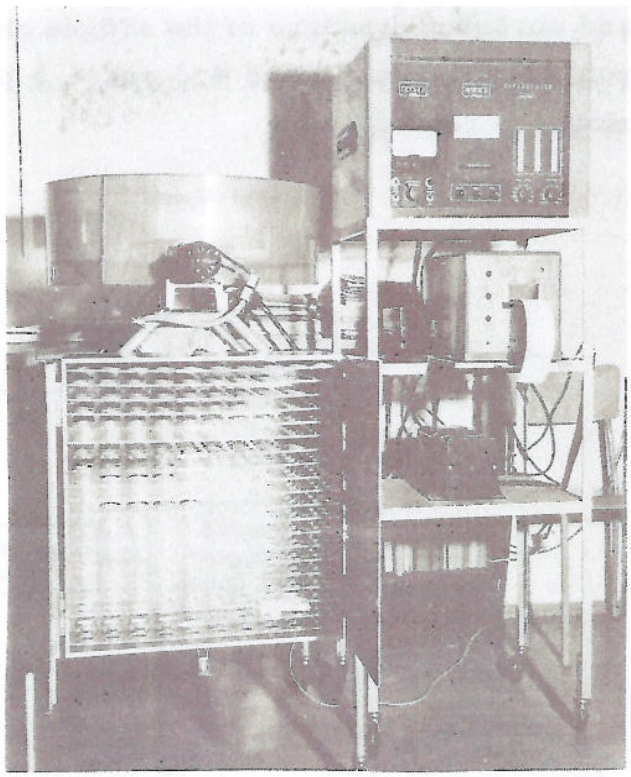

Figure 4.2 Photograph of automatic continuous sample changer, scaler and printing timer
Open doors show storage cabinet for samples

4.2 Special counting equipment

In the study of specimens with very low beta activity it is highly desirable to have GM tubes with anticoincidence screening and specially built shielding. At this Institute a low beta background counter based on Tracerlab's CE-14 equipment has been installed. A modification of the shielding, a layer of 10 cm 20 per cent boron mixed paraffine is supposed to take care of most of the neutrons produced by cosmic rays in the iron shielding. With this shielding the background has been reduced to less than 0.5 cpm.

Several institutes in Norway are already equipped with gamma-spectrometers of different types. By courtesy of The Joint Establishment for Nuclear Energy Research, the Physics Department, University of Oslo, and The Norwegian Radium Hospital, three different gamma-spectrometers have been tried for analysis of milk, plants and peat soils. Special detection outfit is desirable for this type of analysis. At NDRE a 50 channel gamma-spectrometer is now available. The instrument is to be fitted with a 3" x 4" diameter NaI, Tl activated detection unit and will have automatic sample changing.

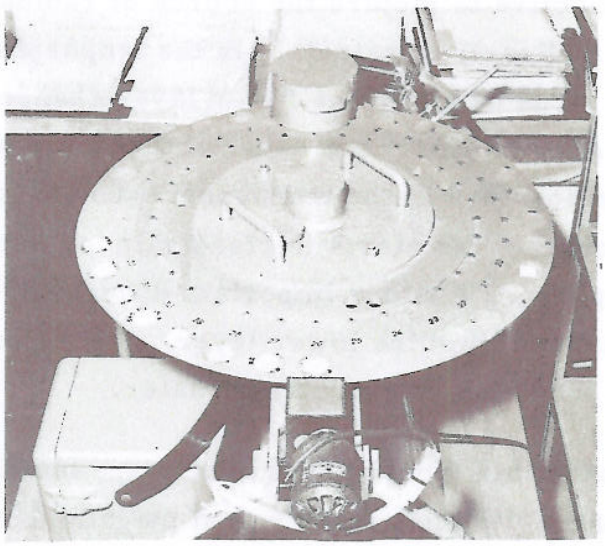

Figure 4.3 Photograph of sample changer with dust cover removed

4.3 Standardization

The standardization has been performed by measuring isotopes of known absolute activity. The ratio between the measured activity, net cpm, and the absolute activity, dpm, is used to convert the activity of the unknown sample into absolute units.
Details concerning the standardization with the isotopes used are given in Appendix II. The standard factors are of course specific for the instruments used. In our case they usually are independent of specimen weight within the range of practical interest. This may in part be due to the fact that as specimen

weight and self absorption increases, the distance between top of specimen and counting tube is slightly reduced.

In Appendix II is also given the method used for calculation of the standard deviation due to counting.

5 FALLOUT IN PRECIPITATION

5.1 Introduction

5.1.1 Fallout deposition and sampling procedures

It may be instructive to picture the source of radioactive fallout as the content of radioactive materials in the troposphere and the stratosphere. The height of the lower layer changes with latitude and season. A main difference between these two layers is that the overall temperature in the troposphere decreases with altitude up to the tropopause and starts to rise again or becomes constant in the stratosphere. Another important difference is that while snow and rain originates from the lower layer, the upper layer, the stratosphere, contains practically no water.

Radioactive materials from kiloton bombs are mainly distributed in the troposphere, while in the case of megaton bombs 90 - 100 per cent of the radioactivity may be brought up into the stratosphere, see Libby (31). According to Greenfield (32, the radioactive particles in the troposphere through their Brownian movements will be picked up by water droplets in clouds and brought down as fallout with rain and other atmospheric precipitation. The lifetime of free particles in a cloud increases with the size of the particle up to a diameter of 0.5 micron. For larger particles it decreases again. The average residence time for radioactive particles in the troposphere has been measured by Stewart, Osmond, Crooks and Fisher (33) to 35 days. Later investigations (34, 35) have shown that the residence time in the troposphere depends upon the altitude. Thus, the residence time of the fallout from the lower part of the troposphere may be only a few days.

The radioactive particles in the stratosphere have to move into the lower layer, the troposphere, before they can be brought down by the action of the moisture. This is a slow process, and the average residence time in the stratosphere has been estimated to years.

In addition to the precipitation process fallout is also brought down as dry dust through turbulent deposition. This is not due to gravitation, but to eddy currents and Brownian movements which bring the particles in contact with surfaces where they may stick. The rate of this deposition becomes proportional to the free surface area, the air concentration and the average velocity of the particles. In ordinary gaseous diffusion this velocity is given by the temperature and the mass of the particle. In the present case, however, it also depends upon the turbulence of the air, which among others is very dependent upon the shape and surroundings of the collecting device. Stewart et al 1958 (33, 36) have determined an effective velocity - the velocity which multiplied by air concentration gives the rate of deposition - over horizontal sheets of gummed paper to 0.07 cm/sec. The same effective velocity is found for their standard polythene collecting funnel. They find that this rate of deposition is small compared to the cumulation caused by atmospheric precipitation.

It should be noted that the effective velocity of other funnels or ground covered with vegetation may be very different from 0.07 cm/sec. Thus Blifford (37) reports that a vertical screen kept normal to wind direction collected 20 times as much as a horisontal gummed paper of the same area. Rosinski 1958 (38) working with an artificial grass mat arrived at similar results. These investigations show clearly that the results obtained are highly dependent on the sampling method used.

The final steps in the fallout mechanism always are atmospheric precipitation and turbulent deposition. The standard procedures in fallout sampling have been the use of sticky films, pots and air filters. Recent investigations have shown that none of these methods are capable of giving a true picture of the total deposition on ground covered with vegetation. Only in the case of oceans,

lakes, deserts etc, true relationships between the mentioned standard sampling procedures and the actual deposition may be found (37).

Investigations on the radioactivity of vegetation by Herbst, Langendorff, Phillipp and Sommermeyer (39) and Aarkrog and Lippert (40) show that the activity of the leaves is roughly proportional to the area of the leaves and the concentration of radioactivity in the air. Ground covered with vegetation thus will take active part in the process of fallout.

It has frequently been noted that the amount of fallout deposition is roughly proportional to the amount of precipitation. This statement obviously does not include the process of turbulent deposition. It has further been stated that more radioactivity is brought down at the beginning of a shower than later on. This has been carefully investigated by Cowan and Steimers (41) for the isotopes Mo-99, I-131 and Sr-90. They find that the rate of fallout during single showers may vary in a most irregular manner. Frequently a higher concentration of fallout is observed in a small shower than in a larger, but a great many cases to the contrary are also observed. The proportionality between gross activity and volume of rainfall is much more strongly indicated. In a number of cases the particulate fraction of gross activity is reduced as a period of rainfall progresses, however, in many cases it stays constant or fluctuates. Their conclusion is that the activity of rain water as function of time depends upon a number of unknown factors.

5.1.2 Stratospheric and tropospheric contributions

According to the picture given, the fallout consists of two parts, tropospheric and stratospheric fallout. The first has not been outside the troposphere and should therefore be relatively fresh in radiochemical composition. The second has spent a considerable time in the stratosphere and its composition may correspond to a few years of age. It is important to learn which fraction of the fallout is of tropospheric or stratospheric origin. The reason

for this is that the main store of fallout materials at present is in the stratosphere. The future amounts of Sr-90 and other long lived fallout isotopes on the earths surface are therefore determined by the rate of stratospheric decontamination.

Libby (31) originally suggested that stratospheric fallout was evenly distributed over the whole of the earths surface, while tropospheric fallout deposited at the latitudes where the bombs were fired in a zone of about 10° width. In the first instance he assumed a sharp limitation in the width of these zones, which lead to a steplike latitudinal distribution of fallout. Libbys theoretical curve for the total sum of Sr-90 fallout up to December 1957 is reproduced in figure 5.1. This curve is calculated for an assumed world average precipitation of 770 mm/year. It is further based upon an estimated half life of 10 ± 5 years for the stratospheric reservoir. This estimate has recently been modified by Libby (35).

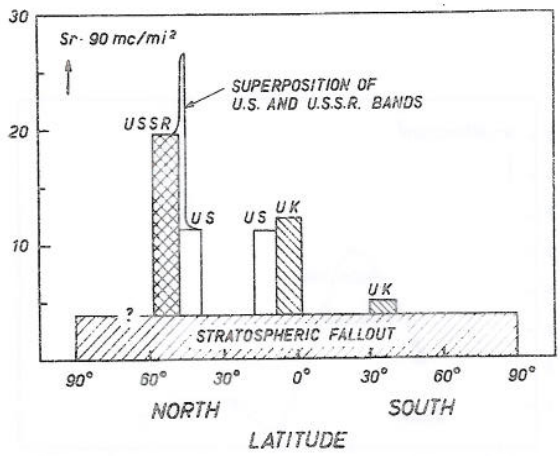

Figure 5.1 Theoretical latitudinal fallout
Libby 1958 (31)

If we assume that the initial fission product distribution is not seriously disturbed by the transport through the atmosphere, and that the average residence time of fallout in the troposphere is much shorter than in the stratosphere, then the Sr-89/Sr-90 ratio may be used to determine the fractions of tropospheric and stratospheric fallout. Due to its short half life - 52 days - all Sr-89 may be assumed to originate from tropospheric fallout and

its average age to be about 35 days. The corresponding average ratio between the two isotopes may be taken to be 100:1, the initial ratio being set to 154:1. The corresponding amount of Sr-90 is then given, and the rest of the measured Sr-90 should be due to stratospheric fallout. This method has been used by Stewart et al (36), and their conclusion is that the main part - some 90 per cent - of the Sr-90 fallout at middle latitudes is of stratospheric origin. They assume that this result is due to a general circulation which has previously been suggested by Dobson (42) and Brewer (43) to explain the ozone and water distribution in the atmosphere. According to this theory tropospheric air rises into the stratosphere at the equator and enters the troposphere again at higher latitudes. The seasonal variation in fallout deposition are due to the formation of a pool of cold air above the winterpole in the late winter months when the pole is in the shadow. The latitudinal distribution of Sr-90 fallout according to this theory is shown in figure 5.2

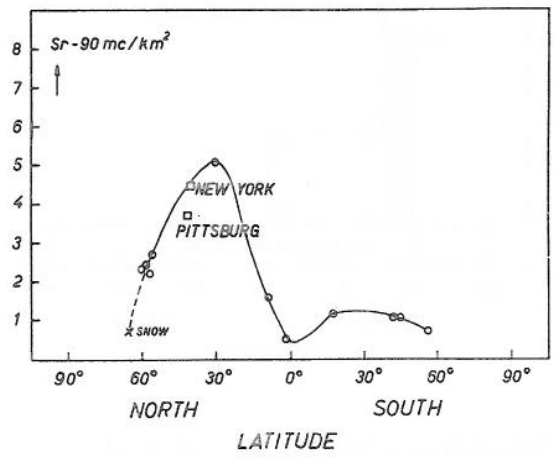

Figure 5.2 Total deposition of Sr-90 in 1956 at various latitudes Stewart et al 1958 (36)

The theory of Libby has been criticized by Machta and List (44). They have reinterpreted the experimental data used by Libby and reached the conclusion that the latitudinal variation of Sr-90 fallout is mainly due to variations in the stratospheric part of the fallout. They also find that the average residence time of Sr-90

in the stratosphere is only half the value estimated by Libby in 1958. Their latitudinal distribution of fallout with stratospheric and tropospheric components is reproduced in figure 5.3.

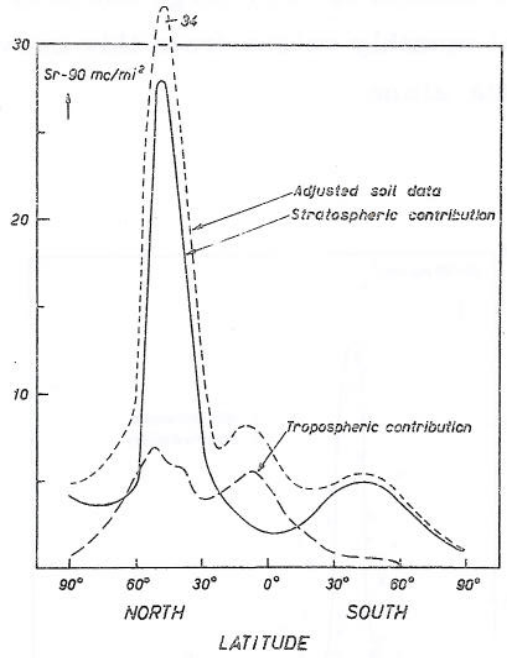

Figure 5.3 Total deposition of Sr-90, soil data Machta and List 1958 (44)

It should be noted that the existence of a marked peak at 0° - 10° latitude north as pointed out by Machta and List, may be rather doubtful. It is not well reflected in the experimental data reproduced in figure 5.4.

Libby (31) has used the ratio between the local amount of rainfall and the world-wide average rainfall of 770 mm to calculate the measured fallout from his basic distribution. Integrated fallout curves for a few locations over a number of years give reasonable agreement with the experimental values. When, however, the differential increase in the basic distribution is used to calculate monthly average fallout values, the precipitation factor dominates and there is actually no significant agreement with the increase in the basic distribution, such as suggested by Libby. By dividing the experimental monthly average values for Sr-90 in 1956 and 1957 given by Libby, with the corresponding amounts of precipitation

and multiplying by the world average of 770 mm, the monthly variation in the basic distribution is obtained. In figure 5.5 these values are given on a logaritmic scale. The corresponding distribution of data from Stewart et al 1958 (36) is also given in figure 5.5. The data in figure 5.5 show that the spread in the average distribution is very large and it is hardly possible to forecast local monthly values from this curve and precipitation data alone.

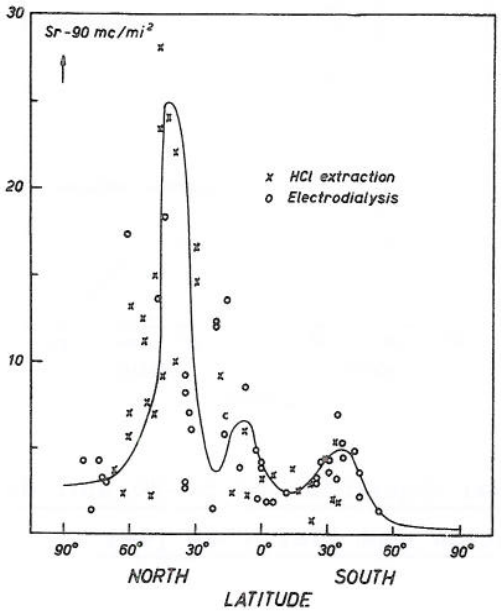

Figure 5.4 Total deposition of Sr-90, soil data
Machta and List 1958 (44)

The theory of Stewart et al has been critically reviewed by Machta (45). According to Machta there are serious meteorological objections to the details of the model used by Stewart et al. Storebϕ (46) has suggested that spring time increase of bomb debris deposition is due to subsidence caused by the break down from above of the circulation around a cold polarnight low pressure center at high altitude.

Libby (31) has thrown some doubt on the accuracy of the radiochemical determination of the Sr-89/Sr-90 ratio used by Stewart and by Machta in their evaluation. Libby has further pointed out that the ratio between Ba-140 and Sr-90 should be

more suited for this purpose, as the half life of Ba-140 is only 14 days. This method has been worked out in detail by Kuroda (47). His investigations largely verify that the main part of the Sr-90 fallout is of stratospheric origin.

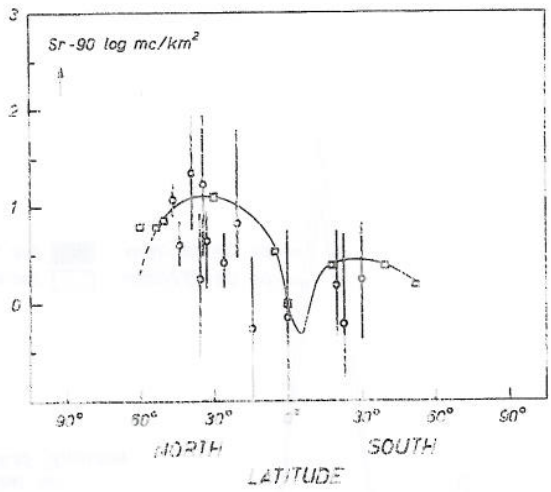

Figure 5.5 Monthly variation in the calculated differential distribution of Sr-90 in precipitation 1956 and 1957
Mean values with maximum and minimum values are given
Data from (31) and (36)

The interpretation of the rate and distribution of the fallout is seriously complicated by the fact that the fission products originate from a number of different bomb tests. Recently during the 1958 USA summer tests in the Pacific, elements which produced W-185, half life 74 days, and Rh-102, half life 220 days, were added (48). By using these elements as tracers some of the previous difficulties are overcome. Results from the W-185 test have recently been published (49), and they show an approximately symmetric distribution of the air activity about the equator.

5.3 Experimental data and discussion

In the present work Sr-90, Sr-89 and Cs-137 have been determined in samples of rain from Bergen, Mǿsvann, Kjeller and Ski in

1957 and 1958. At Møsvann and Ski snow has also been collected and analyzed during the winter 1957/1958 and 1958/1959. The sampling points are indicated on the map in figure 2.1. The data are given in tables 5.1 - 5.4. Some of the data from 1957 have previously been published (8). The Sr-90 and Cs-137 data for 1958 are also given graphically in figures 5.6 - 5.9.

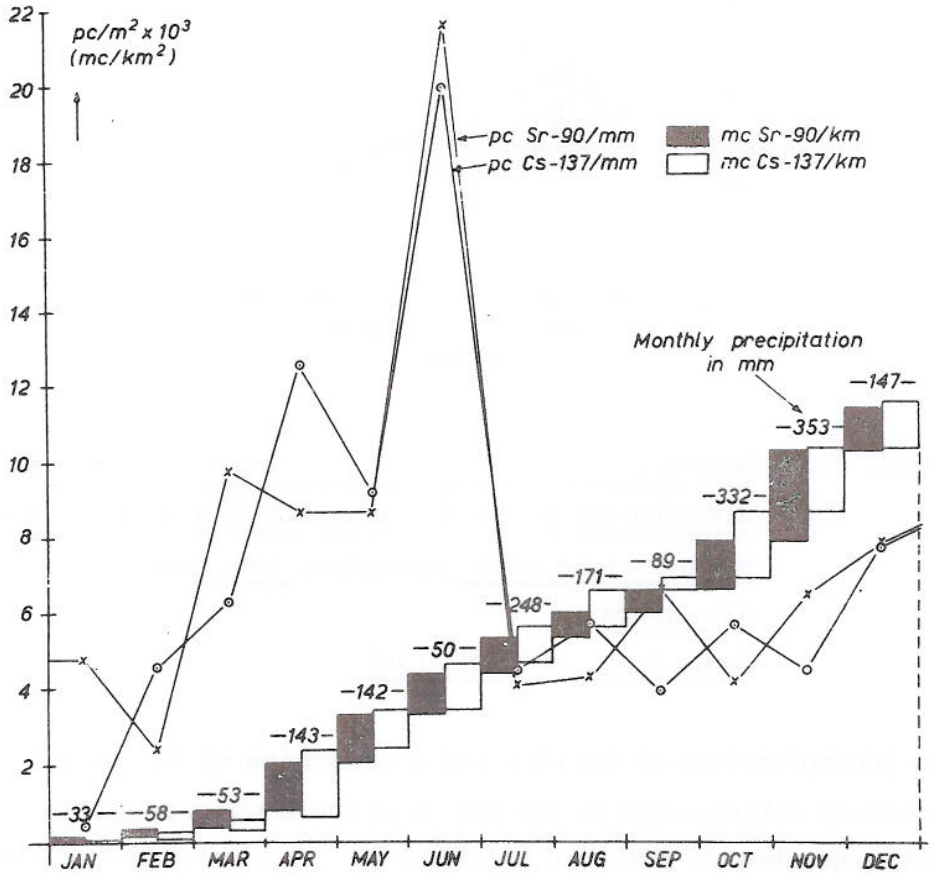

Figure 5.6 Cumulation of Sr-90 and Cs-137 from precipitation at Bergen, Norway 1958

In the following the conformity of these data with previous results will be discussed.

5.2.1 Rate of Sr-90 and Cs-137 cumulation

The monthly cumulation of Sr-90 and Cs-137 in different localities is given in figures 5.6 - 5.9. In the same figures are also given the monthly precipitation in millimeters and the average monthly

Date collected		Precipitation mm	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D	Cs-137 / Sr-90	Sr-89 / Sr-90
1957										
Nov	23 - 24	45.5	2.1	0.5	15.8	2.0	0.7	0.1	0.33	7.5
	24 - 25	100.2	1.3	0.4	11.0	1.6	0.6	0.1	0.46	8.5
	25 - 26	28.2	2.3	0.4	32.6	2.5	1.7	0.4	0.74	14.2
	26 - 27	13.3	1.1	0.3	9.4	2.1	1.0	0.8	0.91	8.5
	27 - 28	20.1	1.4	0.3	35.0	2.3	2.2	0.4	1.57	25.0
Dec	4 - 6	36.9	2.1	0.3	35.1	2.4	1.5	0.5	0.71	16.7
	7 - 17	34.9	5.6	0.5	45.0	3.7	2.2	0.7	0.39	8.0
	24 - 25	22.7	1.8	0.3	26.4	2.0	1.8	0.5	1.00	14.7
	25 - 26	35.3	5.3	0.5	105.0	5.9	4.5	0.6	0.85	19.8
	26 - 27	38.7	2.3	0.3	34.7	2.7	2.5	0.4	1.09	15.1
	27 - 11	32.9	4.8	0.4	63.2	3.1	0.4	0.4	0.08	13.2
Average			2.5				1.7		0.68	
1958										
Feb	12 - 16	16.0	2.7	0.3	8.6	3.2	3.2	0.4	1.19	3.2
	16 - 2	42.0	2.3	0.3	22.6	3.2	5.1	0.5	2.22	9.8
Mar	2 - 4	30.8	8.5	0.7			3.7	0.4	0.44	
	4 - 13	22.2	11.5	0.7	75.0	10.3	9.9	0.8	0.86	6.5
Apr	13 - 15	23.6	9.5	0.6	34.9	5.3	7.4	0.6	0.78	3.7
	15 - 19	16.0	4.5	0.4	30.2	5.0	3.2	0.2	0.71	6.7
	19 - 20	21.0	5.1	0.5	70.4	5.8	8.1	0.7	1.59	13.8
	20 - 21	19.7	9.9	0.7	61.2	9.2	11.0	0.9	1.11	6.2
	21 - 25	39.5					8.7	0.5		
	25 - 29	26.2	15.4	1.3	38.5	8.5	8.7	0.5	0.57	2.5
	29 - 2	36.0	6.5	0.6	138.0	10.0	16.6	1.2	2.55	21.2
May	2 - 6	26.2	3.9	0.4	35.9	1.4	3.6	0.3	0.92	9.2
	6 - 9	10.3	12.6	0.8	122.3	5.3	11.7	0.7	0.93	9.7
	9 - 10	19.1	16.7	0.9	92.8	5.9	13.9	0.7	0.83	5.6
	10 - 11	18.7	11.1	0.7	94.6	4.3	11.1	0.6	1.00	8.5
	20 - 22	20.0	3.8	0.5	33.8	4.1	5.8	0.4	1.53	8.9
	22 - 25	20.4	11.1	0.9			12.3	0.7	1.11	
	25 - 18	31.8	26.2	1.0	141.6	18.7	23.7	1.1	0.91	5.4
Jun	18 - 8	17.8	13.5	0.6	63.9	9.6	13.0	0.6	0.96	4.7
Jul	8 - 9	41.8	4.5	0.3	37.4	3.0	4.0	0.2	0.89	8.3
	9 - 13	38.7	5.6	0.3	45.2	4.9	7.0	0.4	1.25	8.1
	13 - 22	31.5	5.6	0.4	35.2	3.8	7.2	0.5	1.24	6.3
	22 - 25	31.2	3.9	0.3	18.9	2.5	4.7	0.3	1.21	4.8
	25 - 30	40.5	3.7	0.3	24.3	2.6	4.9	0.4	1.32	6.6
	30	35.8	0.8	0.2	6.1	1.4	1.1	0.5	1.38	7.6
	30 - 2	28.0	4.5	0.3	27.4	3.2	2.0	0.3	0.44	6.1
Aug	2 - 5	31.6	2.7	0.3	7.1	1.9				2.6
	5 - 6	32.6	1.1	0.2	14.9	1.7	3.2	0.3	2.91	13.5
	6 - 7	20.7	4.8	0.5	19.7	3.5	5.2	0.5	1.08	4.1
	7 - 13	31.1	5.5	0.3	62.5	4.1	9.1	0.9	1.66	11.4
	13 - 15	19.5					7.7	0.5		
	15 - 29	35.4	7.2	0.5	191.0	8.2	4.1	0.7	0.57	26.5
	29 - 9	27.8	8.9	0.6	190.7	8.2	6.5	0.4	0.73	21.4
Sep	9 - 21	37.0	5.8	0.5	82.8	5.4	1.9	0.4	0.33	14.3
	21 - 30	23.8	5.1	0.4	61.7	4.0	4.2	0.6	0.82	12.1
	30 - 5	28.4	2.9	0.3	66.6	4.6	6.0	0.5	2.07	22.9
Oct	5 - 8	27.6	1.9	0.3	16.4	3.7	8.6	0.6	4.53	8.6
	8 - 10	43.5	3.8	0.4	70.2	7.0	4.8	0.4	1.26	18.5
	10 - 12	22.9	4.0	0.4	17.4	1.4	8.3	0.8	2.08	4.4
	12 - 15	36.0								
	15 - 20	22.6	3.5	0.4	127.6	7.2	3.2	0.3	0.91	36.5
	20 - 23	38.6	3.0	0.3	49.2	4.1	3.7	0.3	1.23	16.4
	23 - 24	40.5	2.8	0.5	36.7	3.7	3.4	0.3	1.21	13.1
	24 - 25	33.6	4.6	0.7	59.5	6.1	5.4	0.7	1.17	12.9
	25 - 30	36.0	7.9	0.4	219.7	11.6	9.9	0.5	1.25	27.8
	30 - 31	38.2	6.1	0.6	187.0	10.5	4.6	0.4	0.75	30.7
	31 - 3	33.4	7.2	0.6	259.2	13.3	2.9	0.3	0.40	36.0
Nov	3 - 6	41.2	6.2	0.5	347.7	17.0	7.7	0.4	1.24	56.1
	6 - 7	29.0	4.1	0.4	162.4	8.3	6.5	0.4	1.59	39.6
	7 - 21	117.0	6.4	0.6	197.2	10.3	4.4	0.4	0.69	30.8
	21 - 25	107.0	7.6	0.6	254.6	13.2	3.0	0.3	0.40	33.5
	25 - 30	25.0	4.0	0.5	128.2	6.7	5.2	0.4	1.30	32.1
	30 - 7	33.0	11.9	0.8	443.2	23.2	11.0	0.6	0.92	37.2
Dec	7 - 22	43.5	4.7	0.5	95.4	6.2	5.3	0.5	1.13	20.3
	22 - 31	70.5								
Average			6.3				6.4		1.02	

Table 5.1 Sr-90, Sr-89 and Cs-137 in precipitation at Bergen, Norway 1957 and 1958

Date collected	Precipitation mm	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D	Cs-137 / Sr-90	Sr-89 / Sr-90
1957									
Nov 15 - Dec 15	22.6	1.1	0.2	5.8	1.1	0.5	0.2	0.45	5.3
" " - " "	22.6	1.6	0.3	4.2	2.1	0.6	0.2	0.38	2.6
Dec 15 - Jan 15	110.0	1.5	0.3	13.1	1.7	1.1	0.2	0.73	8.7
" " - " "	99.5	1.7	0.3	12.3	1.9	1.3	0.2	0.77	7.2
" " - " "	81.4	1.5	0.3			1.4	0.3	0.93	
" " - " "	88.0	1.7	0.3	15.6	2.1	1.6	0.2	0.94	9.2
Average		1.5				1.1		0.70	
1958									
Jan 15 - Feb 15	73.5	1.4	0.1	14.5	1.2	1.3	0.2	0.93	10.4
" " - " "	71.2	1.9	0.2	11.6	1.5	1.9	0.2	1.00	6.1
" " - " "	72.4	2.0	0.2	18.2	1.8	1.1	0.1	0.55	9.1
" " - " "	62.0	3.7	0.3	15.7	2.4	1.4	0.1	0.38	4.2
" " - " "	63.5	4.1	0.4	13.7	2.5	1.5	0.1	0.37	3.3
Feb 15 - Mar 15	4.0					2.8	0.5		
" " - " "	8.0					3.4	0.2		
" " - " "	8.0					2.9	0.3		
" " - " "	9.2	3.5	0 7	17.5	6.3	3.9	0.2	1.11	5.0
" " - " "	7.1	6.2	0.9			3.1		0.50	
Mar 15 - Apr 15	4.0					3.2	0.4		
" " - " "	0.7	2.2	1.9	64.7	21.8	82.3	8.1	37.41	29.4
" " - " "	4.8	8.2	0.7			8.4	0.4	1.02	
" " - " "	4.6	0.4	0.3	6.0	1.7	2.4	0.3	6.00	15.0
May 20 - May 25	12.5								
" 26 - " 27	14.4								
Jun 17 - Jun 18	13.5	18.2	0.8	100.3	12.6	15.7	1.3	0.86	5.5
" 18 - " 23	14.0	11.1	0.5	72.8	9.0	11.5	0.6	1.04	6.6
" 23 - " 27	16.0	7.8	0.4	69.7	7.5	9.8	0.5	1.26	8.9
" 27 - Jul 2	8.6	14.7	0.6	14.3	7.3	21.9	1.1	1.49	1.0
Jul 2 - " 4	16.3	12.7	0.6	122.6	8.4	13.3	0.8	1.05	9.7
" 4 - " 14	22.9					7.0	0.4		
" 14 - " 22	20.3	7.3	0.4	81.2	6.7	8.7	0.9	1.19	11.1
" 22 - " 24	17.2	5.9	0.4	37.1	4.1	7.2	0.2	1.22	6.3
" 24 - " 28	15.7	5.9	0.4	25.2	3.8	6.5	0.4	1.10	4.3
" 28 - " 31	12.4	3.6	0.3	27.6	3.0	4.9	0.3	1.36	7.7
Aug 1 - Aug 6	14.0	5.1	0.4	10.4	4.4	2.9	0.3	0.57	2.0
" 6 - " 13	14.7	3.6	0.3	25.7	3.9	2.6	0.3	0.72	7.1
" 13 - " 24	19.0	4.9	0.4	50.3	6.8	4.5	0.4	0.92	10.3
" 24 - " 26	14.0	2.5	0.3	11.2	3.7	3.6	0.4	1.44	4.5
" 26 - " 31	12.7	6.3	0.5	135.8	9.9	7.1	0.6	1.13	21.6
" 31 - Sep 15	10.2	9.2	0.6	70.3	5.4	3.6	0.3	0.39	7.6
Sep 15 - " 21	29.0	3.1	0.4	20.2	3.5	1.5	0.6	0.48	6.5
" 21 - " 23	20.1	3.6	0.3	35.5	4.4	15.3	0.8	4.25	9.9
" 23 - " 25	10.2	3.0	0.3			3.0	0.5	1.00	
" 25 - Oct 1	17.7	6.4	0.5			4.8	0.4	0.75	
Oct 1 - " 5	27.4	3.7	0.5	35.5	3.1	5.6	1.3	1.51	9.6
" 5 - " 7	23.5	1.0	0.03	22.7	2.1				22.7
" 7 - " 11	20.0	4.3	0.4	48.9	4.7	2.4	0.4	0.56	11.4
" 11 - " 16	19.8	3.4	0.3	36.9	3.8	7.3	2.3	2.15	10.9
" 16 - Nov 3	7.5	12.5	1.1	310.7	15.0	8.6	0.5	0.69	24.9
Nov 3 - " 8	17.9	4.4	0.4	197.6	10.0	5.2	0.4	1.18	44.9
" 8 - " 16	17.5	2.0	0.1	27.8	1.9	1.5	0.2	0.75	13.9
" 15 - Dec 1	15.0	7.3	0.6	472.4	22.9	3.9	0.5	0.53	64.7
" " - " "	17.4	3.7	0.5	75.6	5.3	2.7	0.3	0.73	20.4
" " - " "	16.5	5.4	0.4	89.3	6.0	4.1	0.3	0.76	16.5
" " - " "	15.9	3.5	0.3	99.1	5.6	2.8	0.2	0.80	28.3
Dec 15 - Jan 16	90.1	1.5	0.2	85.5	4.1	2.8	0.2	1.87	57.0
" " - " "	87.2	2.0	0.2	80.3	4.0	2.7	0.2	1.35	40.2
" " - " "	97.7	1.6	0.2	69.7	3.4	2.4	0.2	1.50	43.6
Average		5.0				5.6		1.12	

Table 5.2 Sr-90, Sr-89 and Cs-137 in precipitation at Møsvann, Norway 1957 and 1958

Date collected	Precipitation mm	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D	Cs-137 / Sr-90	Sr-89 / Sr-90
1957									
Jul 5 - 22	14.6	13.5	1.0	172.0	12.1	19.1	1.5	1.41	12.8
24 - 25	2.7	7.0	1.0	108.5	6.7	9.5	3.0	1.36	15.5
25 - 30	16.1	3.7	0.6	58.5	3.1	2.8	0.8	0.76	15.8
30 - 2	12.7	2.4	0.4	34.1	2.4	1.9	0.5	0.79	14.2
Aug 2 - 8	11.5	2.1	0.4	45.5	2.8	2.2	0.6	1.05	21.7
8 - 12	51.2	1.6	0.3	25.5	2.7	3.4	1.0	2.13	15.9
12 - 13	12.9	1.8	0.3	6.1	1.1	0.4	0.2	0.22	3.4
13 - 14	2.8	5.4	0.6			3.3	0.9	0.61	
14 - 27	19.8	5.2	0.6			3.0	0.8	0.58	
27 - 2	6.2	2.1	0.4			2.6	0.5	1.24	
Sep 9 - 11	8.6	0.7	0.3	96.5	4.5	2.8	0.5	4.00	137.9
11 - 13	19.2	0.9	0.2	58.3	2.7	1.1	0.6	1.22	64.8
13 - 14	33.4	0.3	0.2	11.4	1.1				37.7
14 - 16	26.9	1.1	0.2	14.0	1.1	1.2	0.8	1.09	12.7
16 - 3	8.5	2.9	0.4	160.0	9.5	1.0	0.3	0.34	55.2
Oct 3 - 17	12.0	2.1	0.3	88.2	3.4	7.6	1.6	3.62	42.0
17 - 21	22.0	3.3	0.3	53.8	2.4	1.9	0.4	0.58	16.3
21 - 24	12.0	0.7	0.2	16.0	1.0	1.2	0.3	0.17	22.9
24 - 28	7.7	2.0	0.3	40.0	2.6	2.3	0.5	1.15	20.0
28 - 5	36.6	1.8	0.2	42.0	2.9	1.9	0.4	1.06	23.3
Nov 5 - 26	8.0	2.1	0.4	10.0	2.0	0.7	0.3	0.33	4.8
Average		2.5				3.3		1.32	
1958									
May 10 - 13	3.8	11.4	0.7	89.3	8.8	10.6	0.6	0.93	7.8
14 - 16	20.7	13.3	0.6	65.6	9.5	12.8	0.2	0.96	4.9
16 - 28	20.7	27.4	1.4	236.7	20.0	24.3	1.1	0.89	8.6
28 - 10	0.0	(27.1	2.3)	(124.5	19.8)	(27.9	2.4)	(1.03)	(4.6)
Jun 10 - 18	7.7	12.6	0.6	103.0	8.0	14.2	0.7	1.13	8.2
18 - 19	20.4	15.1	0.6	67.0	7.4	17.7	0.3	1.17	4.4
19 - 26	11.5	10.1	0.4	76.4	6.7	13.7	0.8	1.36	7.6
26 - 30	19.4	9.7	0.5	96.1	6.8	15.6	0.8	1.61	9.9
30 - 14	20.9	5.3	0.3	66.9	5.3	6.7	3.1	1.26	12.6
Jul 14 - 15	3.4	5.3	0.4	44.2	4.0	6.4	0.5	1.21	8.3
15 - 25	34.3	8.9	0.4	74.7	6.5	8.8	0.5	0.99	8.4
25 - 28	13.8	5.9	0.4	33.7	4.0	7.8	0.4	1.32	5.7
28 - 30	13.8	6.2	0.4	31.8	3.8	8.8	0.5	1.42	5.1
30 - 1	6.7	5.9	0.3	56.4	4.8	10.5	0.2	1.78	9.6
Aug 1 - 4	14.0	6.9	0.4	57.7	4.9	7.4	0.4	1.07	8.4
4 - 8	7.1	3.0	0.2	16.4	1.5	3.3	0.2	1.10	5.5
8 - 14	8.7	7.1	0.4	111.1	6.0	10.0	0.7	1.40	15.6
14 - 25	23.6	3.9	0.4	60.9	3.7	3.6	0.6	0.92	15.6
Sep 11 - 22	18.0	5.3	0.4	80.5	5.0	1.9	1.9	0.36	15.2
22 - 23	2.3	7.5	0.6	60.3	5.4	5.9	0.6	0.79	8.0
23 - 27	8.0	10.4	0.8	125.4	8.2	2.8	0.5	0.27	12.1
27 - 2	15.1	10.9	0.6	158.8	8.4	6.2	0.4	0.57	14.6
Oct 2 - 3	8.9	9.9	0.6	162.6	8.0	12.2	0.8	1.23	16.4
3 - 6	16.7	6.5	0.5	65.8	6.4	6.1	0.6	0.94	10.1
6 - 11	19.1	3.7	0.6	50.9	5.8	3.6	0.8	0.97	13.8
11 - 16	25.7	1.8	0.2	47.3	3.1	3.5	0.3	1.94	26.3
16 - 8	21.2	5.8	0.5	186.5	10.3	6.1	0.5	1.05	32.2
Nov 8 - 21	19.2	6.1	1.1	50.5	6.2	2.0	0.3	0.33	8.3
Average		8.5				8.7		1.02	

Table 5.3 Sr-90, Sr-89 and Cs-137 in precipitation at Kjeller, Norway 1957 and 1958

Date collected	Precipitation mm	Sr-90 pc/l	S D	Sr-90 pc/l	S D	Cs-137 pc/l	S D	Cs-137 / Sr-90	Sr-89 / Sr-90
1957									
Dec 13 - Jan 14	45.4	2.1	0.3	26.3	1.5	2.4	0.7	1.14	12.5
" " - " "	42.0	1.5	0.3	17.5	1.3	4.7	0.9	3.13	11.7
Average		1.8				3.5		1.92	
1958									
Jan 14 - Feb 13	52.5	2.3	0.4			2.4	0.3	1.04	
" " - " "	77.0					1.5	0.2		
" " - " "	39.0	2.9	0.5	8.1	3.1	1.8	0.2	0.62	2.8
" " - " "	48.8					2.2	0.3		
Feb 13 - Mar 12	22.5	1.2	0.2	9.9	1.5	2.2	0.4	1.83	8.3
" " - " "	26.0	1.2	0.2	12.9	1.8	1.8	0.2	1.50	10.8
" " - " "	27.0	0.7	0.3	6.3	1.9	1.6	0.2	2.29	9.0
" " - " "	17.5	3.2	0.2			2.1	0.2	0.66	
Mar 12 - Apr 11	9.5	2.3	0.3	19.1	4.0	1.1	0.1	0.48	8.3
" " - " "	11.0	1.7	0.2	35.0	2.6	5.5	0.2	3.24	20.6
" " - " "	8.5					1.5	0.2		
" " - " "	7.6	1.4	0.2	21.0	2.9	1.0	0.3	0.71	15.0
" " - " "	11.2	2.8	0.3	19.9	2.6	2.0	0.1	0.71	7.1
" " - " "	8.5	1.9	0.2	21.0	3.2	2.9	0.2	1.53	11.1
Apr 28 - May 3	9.9	3.9	0.3	45.3	4.2	6.0	0.4	1.54	11.6
May 4 - " 12	32.0	7.4	0.4	61.7	6.4	4.5	0.3	0.61	8.3
" 12 - " 23	37.6	10.1	0.5	85.4	8.0	9.2	0.5	0.91	8.5
" 23 - Jun 1	24.1	12.7	0.6			15.0	0.7	1.18	
Jun 2 - " 21	22.5	10.8	0.4	15.6	5.9	13.6	0.7	1.26	1.4
" 21 - Jul 1	28.5					11.6	0.5		
Jul 12 - " 21	54.2	6.1	0.3	40.1	4.3	5.2	0.2	0.85	6.6
" 21 - " 24	43.8	6.4	0.3	38.7	3.8	6.5	0.3	1.02	6.0
" 25 - " 31	64.4	3.6	0.2	25.2	2.4	4.0	0.2	1.11	7.0
Aug 1 - Aug 9	42.1	2.5	0.3			2.5	0.3	1.00	
" 10 - Sep 1	54.6	0.2	0.03						
Sep 1 - " 25	37.7	3.2	0.2	42.3	2.4	4.6	0.2	1.44	13.2
" 26 - Oct 11	77.3	2.7	0.1	49.6	2.7				18.4
Oct 11 - " 21	45.6	0.4	0.05	37.6	17.9	1.2	0.08	3.00	94.0
" 21 - Nov 15	55.2	4.1	0.6	140.3	11.2	4.1	0.05	1.00	34.2
Nov 15 - Dec 9	5.9	6.4	0.5	180.0	10.8				28.1
Dec 10 - " 20	25.5	1.0	0.2	25.3	1.9	4.5	0.3	4.50	25.3
" " - " "	24.4	4.2	0.3	143.7	7.9	4.2	0.3	1.00	34.2
" 19 - Jan 24	114.7	4.2	0.3	75.6	4.2	3.3	0.3	0.79	18.0
" " - " "	114.7	3.1	0.3	85.8	4.6	2.8	0.2	0.90	27.7
" " - " "	114.4	3.3	0.3	92.6	4.8	3.2	0.3	0.97	28.1
Average		3.8				4.6		1.20	

Table 5.4 Sr-90, Sr-89 and Cs-137 in precipitation at Ski, Norway 1957 and 1958

concentration of Sr-90 and Cs-137 in precipitation.
From these data the corresponding total contributions of Sr-90 and Cs-137 from precipitation in 1958, as given in table 5.5, have been calculated. Table 5.5 show that the total amounts of Sr-90 and Cs-137 brought down with rain and snow over a long period - one year - are only roughly proportional to the amount of precipitation, see also figure 5.23.

Sampling Place	Sr-90 mc/km^2	Sr-90 mc/km^2/m	Cs-137 mc/km^2	Cs-137 mc/km^2/m	Precipitation mm	Cs-137 / Sr-90
Bergen	11.47	5.32	11.69	6.44	1816	1.02
Møsvann	3.37	5.04	3.77	5.64	668	1.12
Kjeller 7 months	3.42	8.45	3.50	8.65	405	1.02
Ski	2.97	3.82	3.58	4.61	777	1.21
Average		5.66		6.33		

Table 5.5 Cumulation of Sr-90 and Cs-137 in 1958 at Bergen, Møsvann, Kjeller and Ski

Studying the monthly averages for Sr-90 and Cs-137 per mm precipitation in figures 5.6 - 5.9, large deviations from this rule are observed and the data given in tables 5.1 - 5.4 show that still larger deviations are observed by day to day measurements. This is in general agreement with the observations of Cowan and Steimers 1958 (41) from single showers. Similar observations have been made by Small (50) measuring the total activity of air filters taken at intervals of 3 hours. Remembering that about 90 per cent of the long lived isotopes are considered to be of stratospheric origin, these results may be interpreted in the following way:

The stratospheric fallout is transferred to the troposphere in portions through short term mixing processes. Due to the short residence time in the troposphere these primary variations in time and space persist and result in irregular daily variations. Seasonal variations may be explained as due to seasonal variations in the mixing process, possibly superimposed on an orderly movement. From this picture of the fallout process it will be understood that the amounts of Sr-90

and Cs-137 brought down with snow and rain are correlated with the total amounts of precipitation over large periods only.

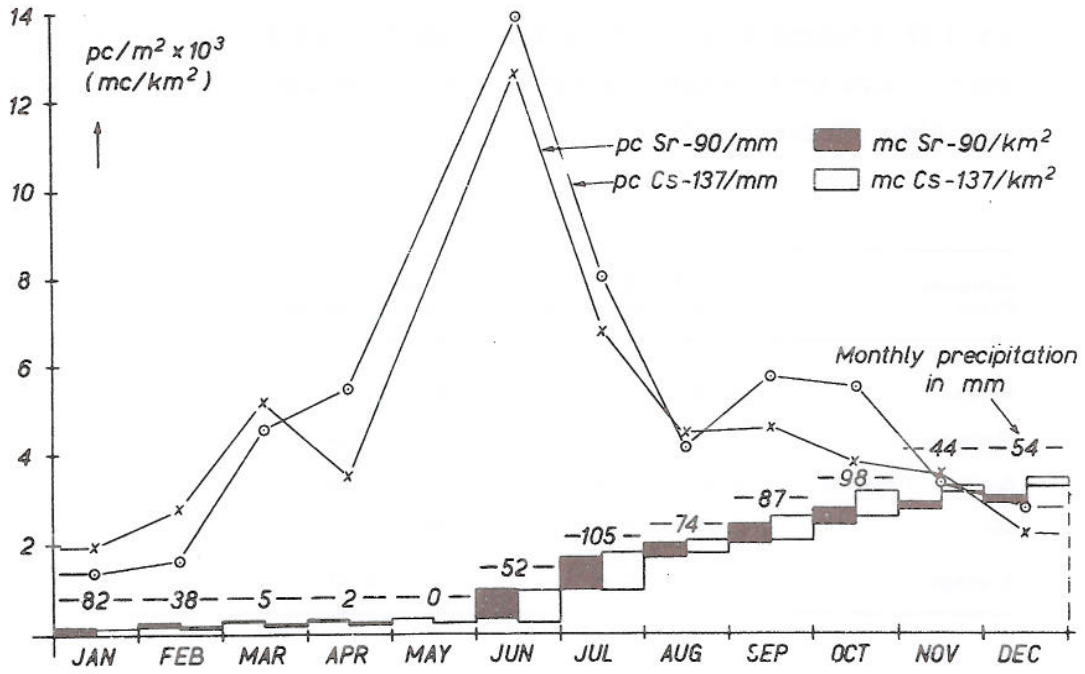

Figure 5.7 Cumulation of Sr-90 and Cs-137 from precipitation at Møsvann, Norway 1958

In figure 5.10 the monthly average concentrations of Sr-90 and Cs-137 in precipitation from the sampling stations are given together with data from other countries. The seasonal variations which have been recorded by Stewart et al (36), seem to appear also in the present observations. Stewart et al have explained these variations as a consequence of their atmospheric circulation model. The details of this model may be questioned, but it may safely be stated that the seasonal variations are due to variations in the rate of transfer of fallout from the stratosphere to the troposphere. An extensive study of these variations on monthly samples from different locations, especially at higher latitudes, would seem profitable.

A striking feature of the curves in figures 5.7 and 5.9 is the small amounts of Sr-90 and Cs-137 brought down with snow. This is due to small amounts of precipitation as well as low concentrations of Sr-90 and Cs-137 in the snow samples. Hvinden (51, 52) has

measured the total beta activity in precipitation from Bergen and Kjeller. The data are reproduced in figures 5.11 and 5.12 and show that small amounts of fallout have been brought down by snow at the beginning of 1958. The total beta activity is, however, not reduced at the end of 1958. This is probably due to bomb tests in the autumn of 1958. The same conclusion is arrived at from a study of the Sr-89/Sr-90 ratios in figure 5.13. The concentrations of Sr-90 and Cs-137 in 1958 precipitation are given in figures 5.14 and 5.15 and do not show a different picture for the autumn. This is as expected, as the main source of Sr-90 and Cs-137 is the stratospheric reservoir.

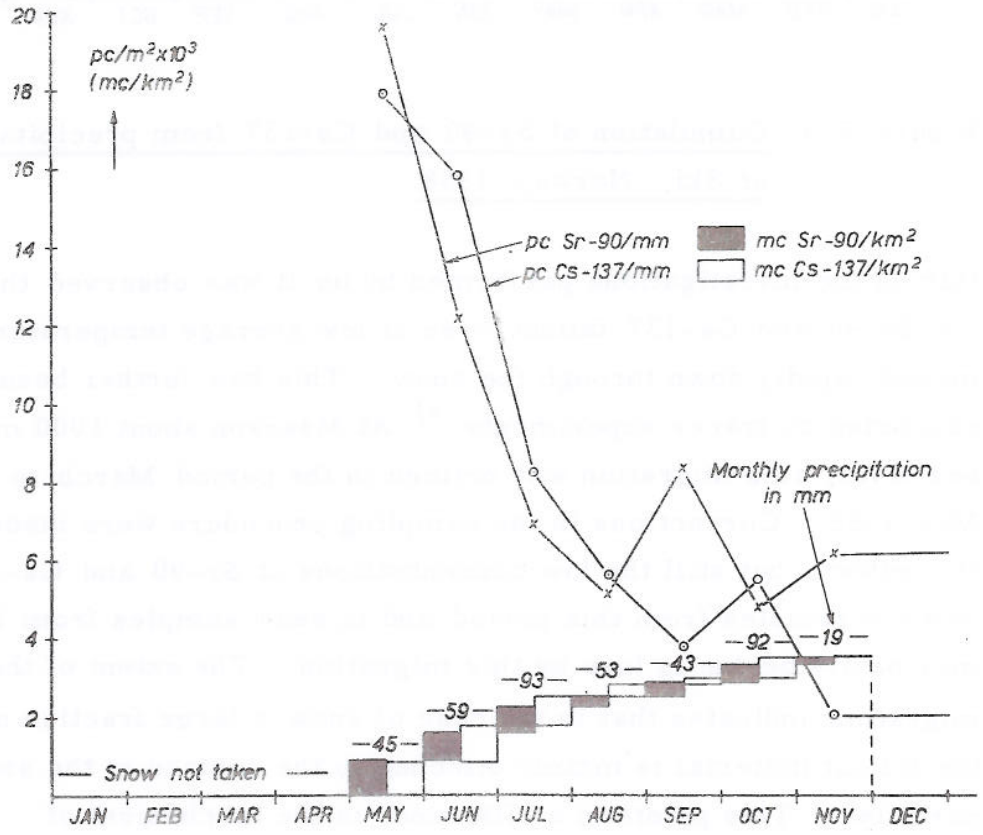

Figure 5.8 Cumulation of Sr-90 and Cs-137 from precipitation at Kjeller, Norway 1958

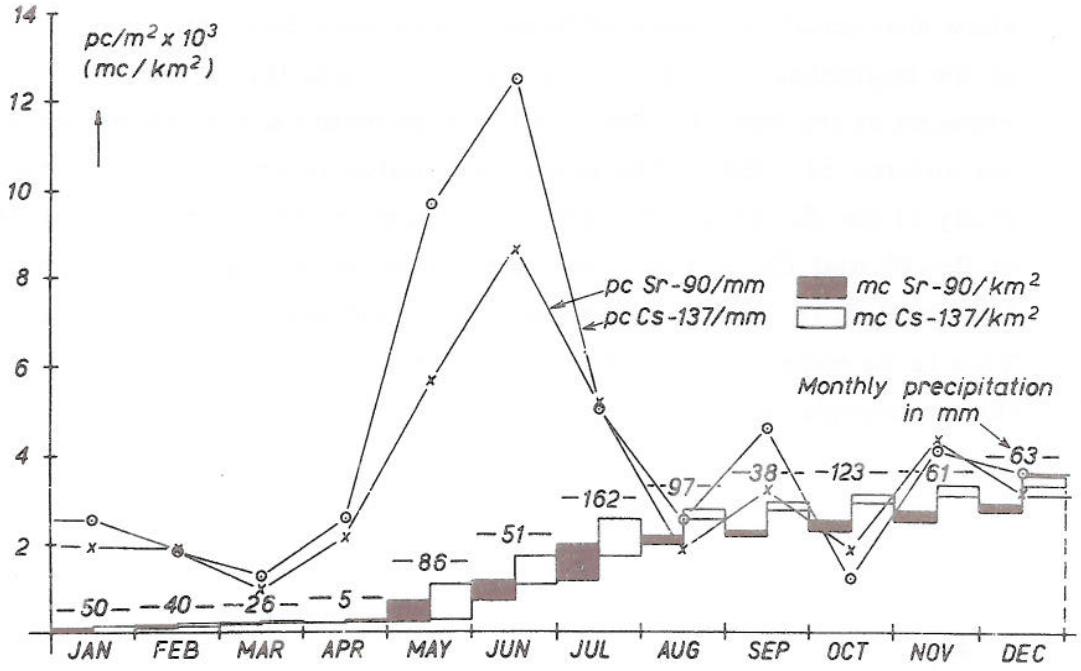

Figure 5.9 Cumulation of Sr-90 and Cs-137 from precipitation at Ski, Norway 1958

During the investigations performed by us it was observed that the Sr-90 and Cs-137 fallout even at low average temperatures moved rapidly down through the snow. This has further been supported by tracer experiments. x) At Møsvann about 1000 m above sea level, this migration was noticed in the period March to May 1958. Corrections in the sampling procedure were made for this effect, but still the low concentrations of Sr-90 and Cs-137 in snow samples from this period and in snow samples from Ski may partly be due to loss by this migration. The extent of the migration indicates that in the case of snow a large fraction of the fallout material is mainly attached to the surface of the snow particles. This points to a difference in the mechanism of fallout capture between snow and rain. Referring to the theory of Greenfield (32) this might be explained by variations in the capture process. Further there is the possibility that snow and rain originates from different levels in the troposphere. If the precipitation is contaminated in proportion to the air concentration, then in general, precipitation from the higher altitude will be more

x) Details to be published

contaminated. As the height at which the snow is formed may be estimated from the crystalline form, a study of these problems might prove rewarding.

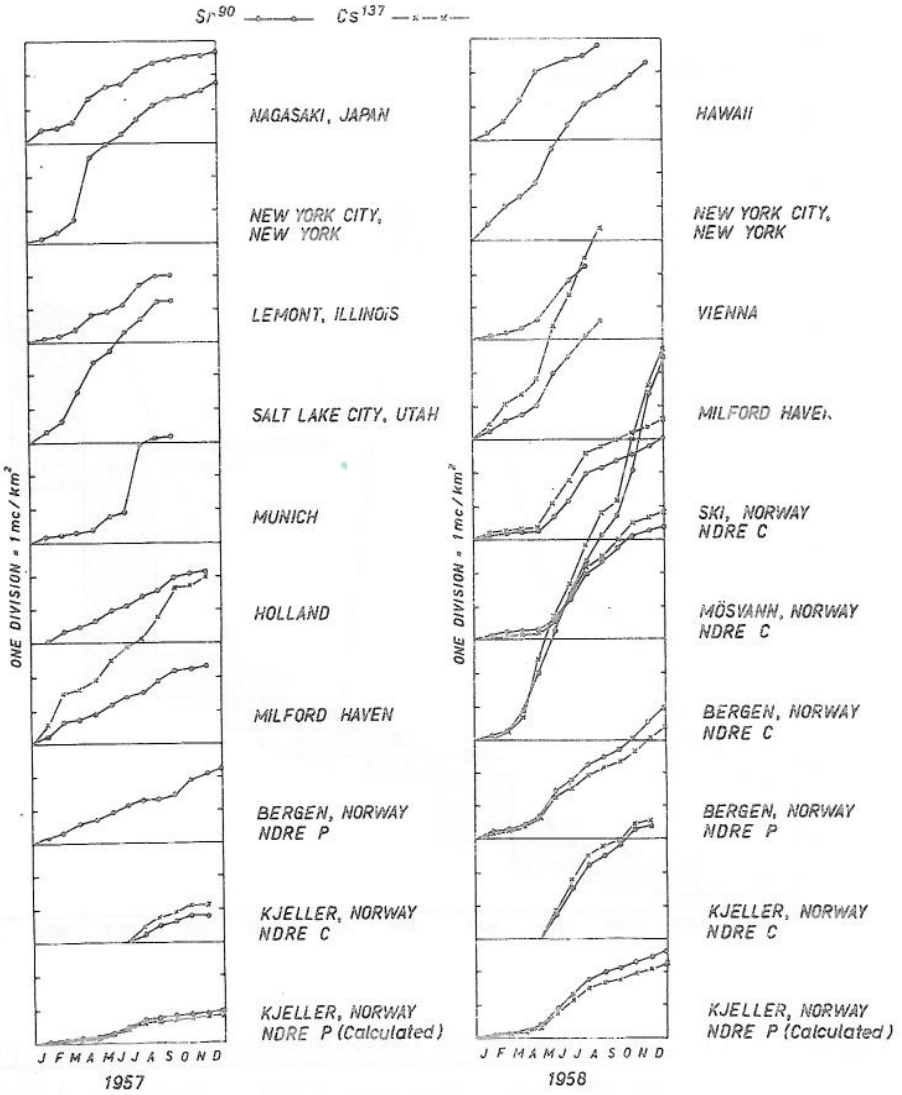

Figure 5.10 The content of Sr-90 and Cs-137 in precipitation at Bergen, Møsvann, Kjeller and Ski 1957 and 1958, compared to data outside Norway, cumulative values NDRE C experimental data from radiochemical investigation, NDRE P calculated data from total beta activity measurements

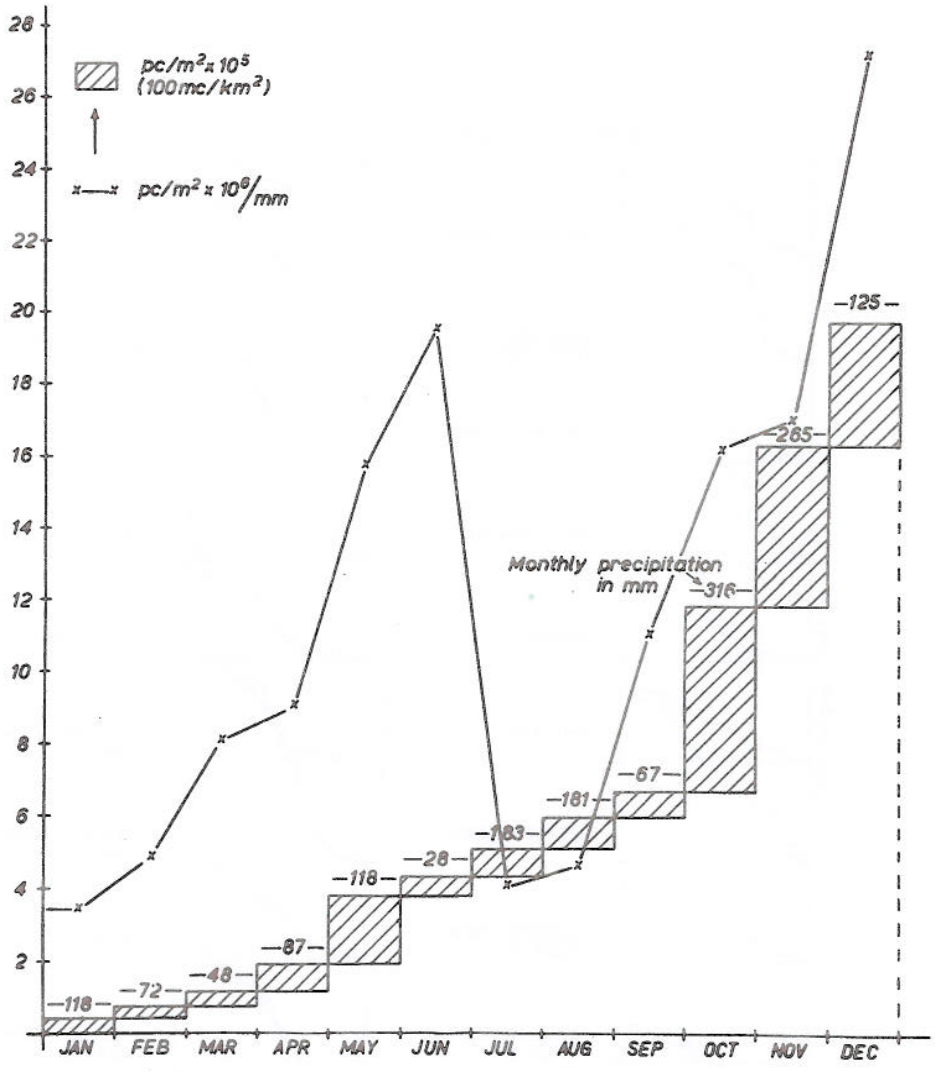

Figure 5.11 Cumulation of total beta activity from precipitation at Bergen, Norway 1958 Hvinden 1959 (52)

The experimental data in figure 5.10 for the two years of observation show that the amounts of Sr-90 and Cs-137 in fallout are nearly the same. There are a number of Sr-90 observations in the literature for collation. For Cs-137, however, there are only data from Milford Haven. The slope of the curve for Sr-90 in rain sampled at Milford Haven is very similar to our observations from Kjeller, Ski and Mφsvann, while the curve obtained for Cs-137 at Milford Haven is much steeper, and can only be compared with our data

from Bergen 1958. The average concentration of Sr-90 given in microcuries per km^2 per mm precipitation from Milford Haven is 5.3 for the first 8 months of 1958, whereas for Cs-137 it is 9.4. This Cs-137 figure seems to be remarkably high.

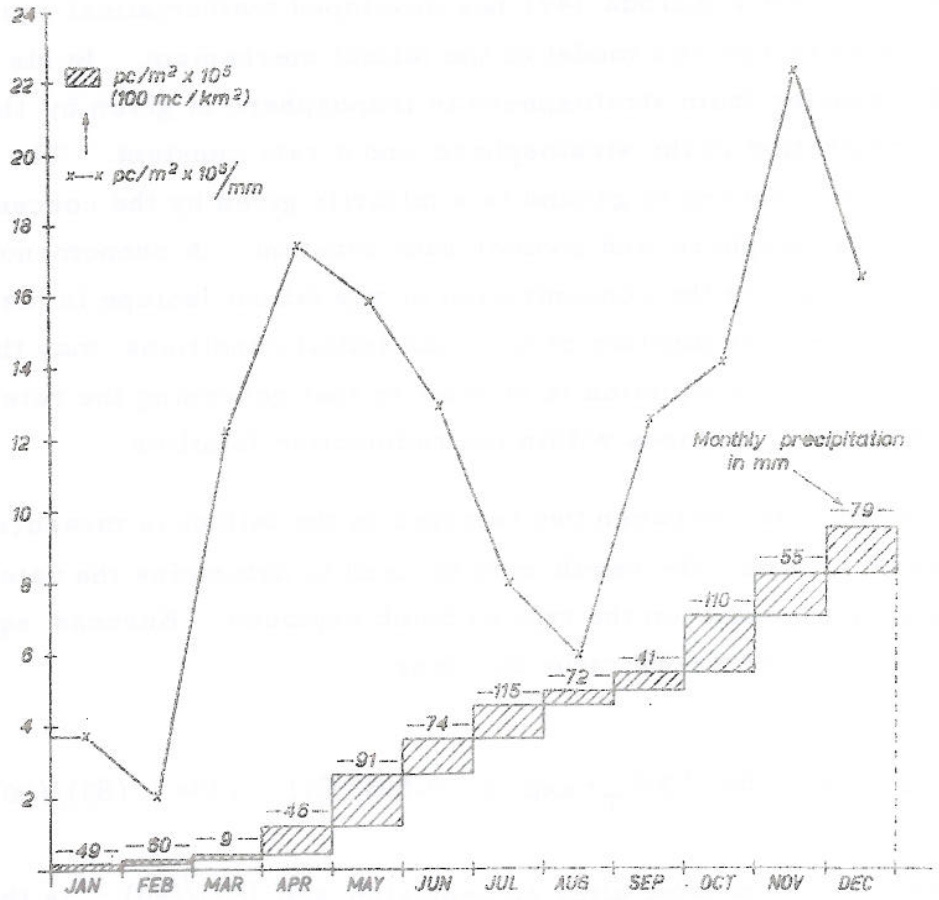

Figure 5.12 Cumulation of total beta activity from precipitation at Kjeller, Norway 1958 Hvinden 1959 (52)

From total beta activity data and decay measurements Hvinden (51, 52) has calculated the content of Sr-90, Cs-137 and a number of other isotopes in the precipitation from 1957 and 1958 in Norway. The values for Sr-90 and Cs-137 from Bergen and Kjeller are reproduced in figure 5.10. As can be seen the calculated values are low by a factor of 2 - 3. This shows that the real content of Sr-90 and Cs-137 in precipitation can not be accurately calculated by assuming the composition of each sample to be given

by decay of the initial products. This is to be expected since the tropospheric and stratospheric parts will be of very different age.

5.2.2 The ratio of Sr-89/Sr-90

As mentioned the ratio of Sr-89/Sr-90 may be used to estimate the tropospheric and stratospheric contributions of fallout in precipitation. More recently Kuroda (47) has developed mathematical equations for a more specific model of the fallout mechanism. In his model the transfer from stratosphere to troposphere is given by the concentration in the stratosphere and a rate constant. The transfer from troposphere to ground is similarily given by the concentration in the troposphere and another rate constant. A phenomenologic equation giving the concentration of any fallout isotope in the troposphere as function of time and initial conditions may thus be obtained. The equation is similar to that governing the rate of growth and decay of isotopes within the radioactive families.

When the ratio between two isotopes in the fallout is measured for a period of time, the result may be used to determine the date of explosions and even the type of bomb exploded. Kurodas equation for the Sr-89/Sr-90 ratio becomes:

$$\alpha = [154 - (89)/(90)_T \cdot \exp(t \cdot 0.693/52)] \cdot [154 - (89)/(90)_{S,o}]$$

where t is the time after an explosion and $(89)/(90)_T$ is the measured ratio between the isotopes in the troposphere at time t. $(89)/(90)_{S,o}$ is the ratio between the isotopes in the stratosphere at the time $t = 0$, just prior to an atomic explosion. α is the fraction of delayed fallout in the total amount of fallout.

The previous estimate of the tropospheric fraction is obtained if the value of $(89)/(90)_{S,o}$ is assumed to be zero and the age of the tropospheric Sr-89 to be 35 days.

In figure 5.13 the ratio of Sr-89/Sr-90 as function of time is given for the present data. The high values indicate fresh fallout from bomb tests. The aim of the present investigation has, however,

been to determine the total amounts of fallout brought down by showers, the sampling periods are therefore too long to be useful for the precise determination of test dates.

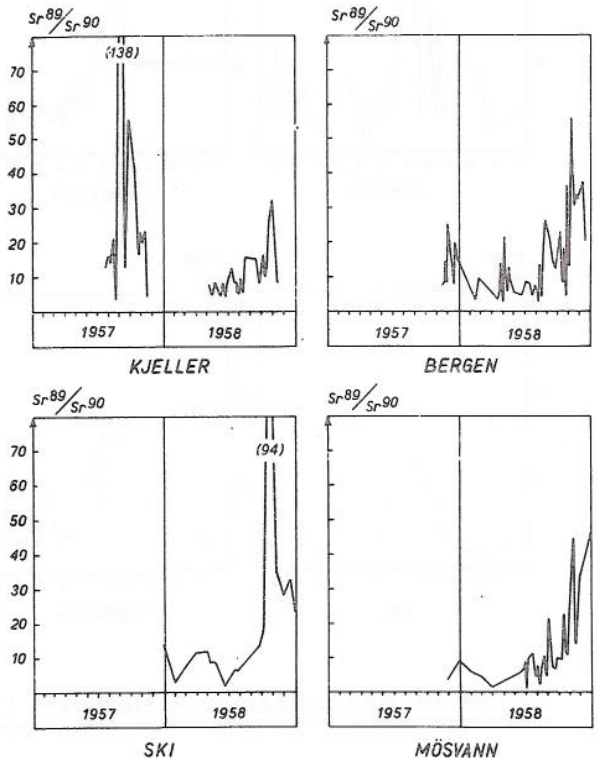

Figure 5.13 Variations of the Sr-89/Sr-90 ratio in precipitation Norway 1957 and 1958

In figure 5.16 the distribution of the precipitation in mm over intervals of the Sr-89/Sr-90 ratio is given for all sampling stations in Norway. The initial ratio of Sr-89/Sr-90 is set to 154 : 1. Values higher than 100 : 1 have only rarely been observed. This is presumably due to the presence of older stratospheric fallout. The most frequent values obtained are in the range between 4 : 1 and 40 : 1. The corresponding average age would be 300 - 100 days. The diagrams further show that the age of all fallout lies within the limits 35 to 400 days. Storebø (46) has recently reviewed the Sr-90 deposition data and concludes that the residence time in the stratosphere should not be much more than one year. The mean transport time for explosions performed in the tropics might be

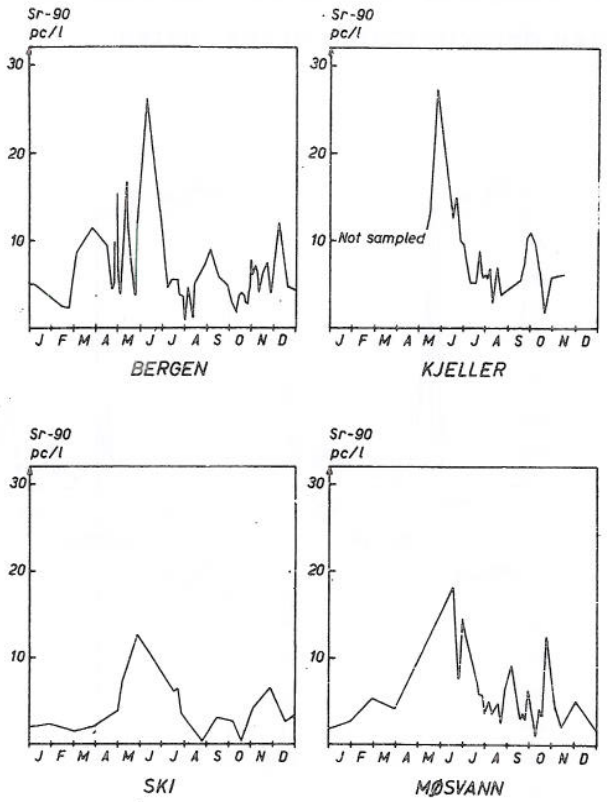

Figure 5.14 Sr-90 in precipitation, Norway 1958

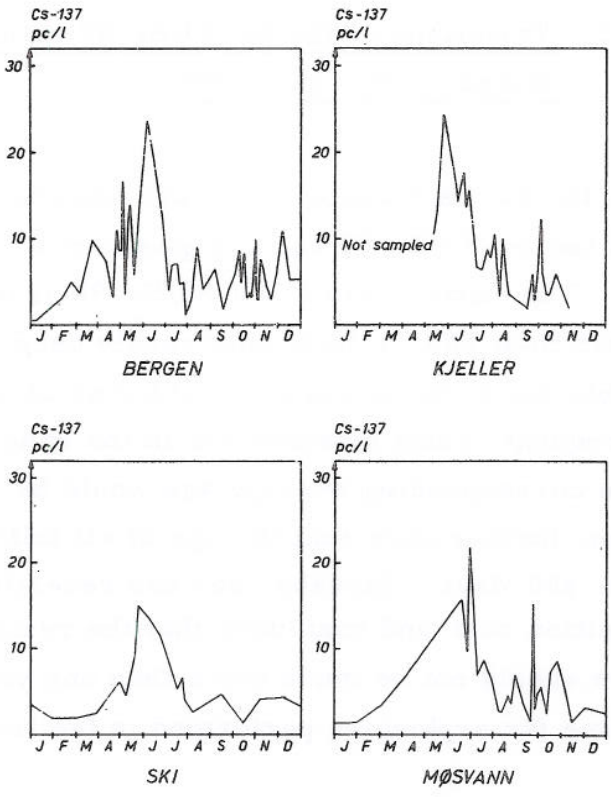

Figure 5.15 Cs-137 in precipitation, Norway 1958

longer than for explosions performed at higher latitudes. Our data given in figure 5.16 fit well into this picture.

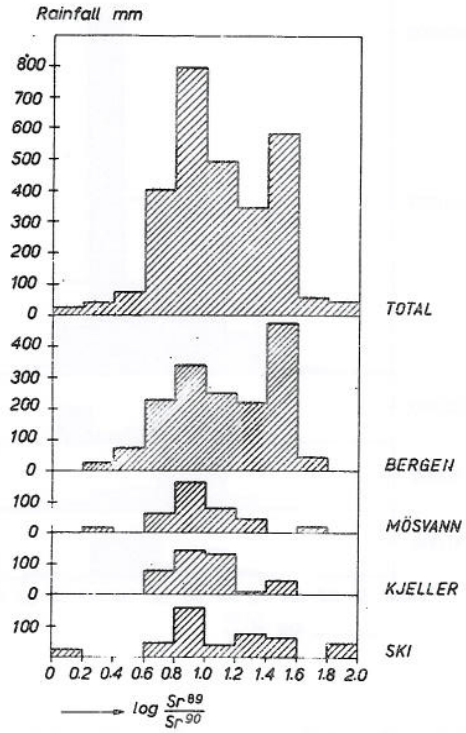

Figure 5.16 The distribution of the precipitation in mm over intervals of the Sr-89/Sr-90 ratio, Norway 1957 and 1958

There are, however, objections against this interpretation. The residence time in the troposphere has been estimated from measurements to be 35 days (33). Kuroda has from a study of the Ba-140/Sr-90 ratio obtained a value of 10 days from single explosions. As the average age calculated above is considerably higher, not only the Sr-90, but also the main part of the Sr-89 would have to be of stratospheric origin. It is, however, known that a number of bomb tests took place in 1958, and the peak between 1.4 and 1.6 for the Sr-89/Sr-90 ratio in figure 5.16 from Bergen must be mainly due to fresh fallout in the autumn. The observed average age of 80 - 100 days is most probably too long. The explanation must be that there is a considerable fraction of delayed fallout present. The average age thus represents a compromise between the residence times of tropospheric and stratospheric fallout. In figure 5.17 the

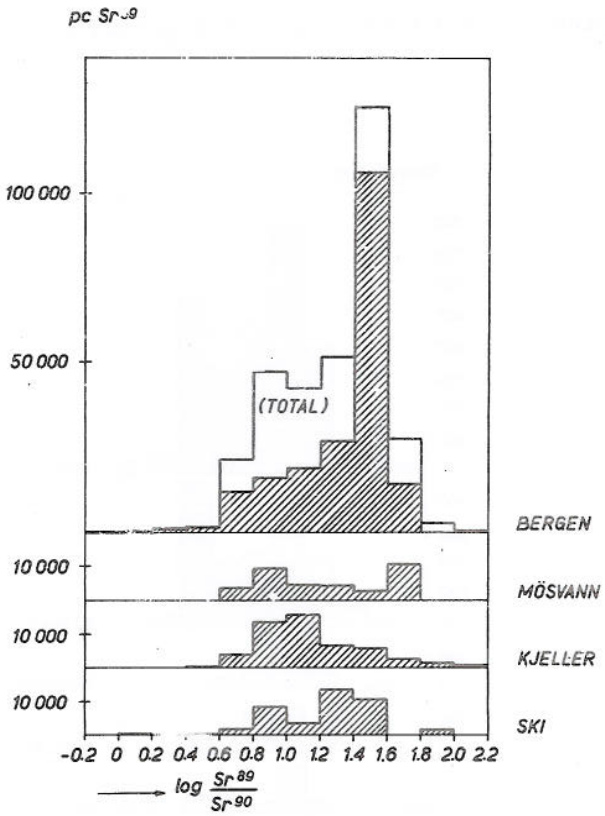

Figure 5.17 The distribution of pc Sr-89 in precipitation over intervals of the Sr-89/Sr-90 ratio, Norway 1957 and 1958

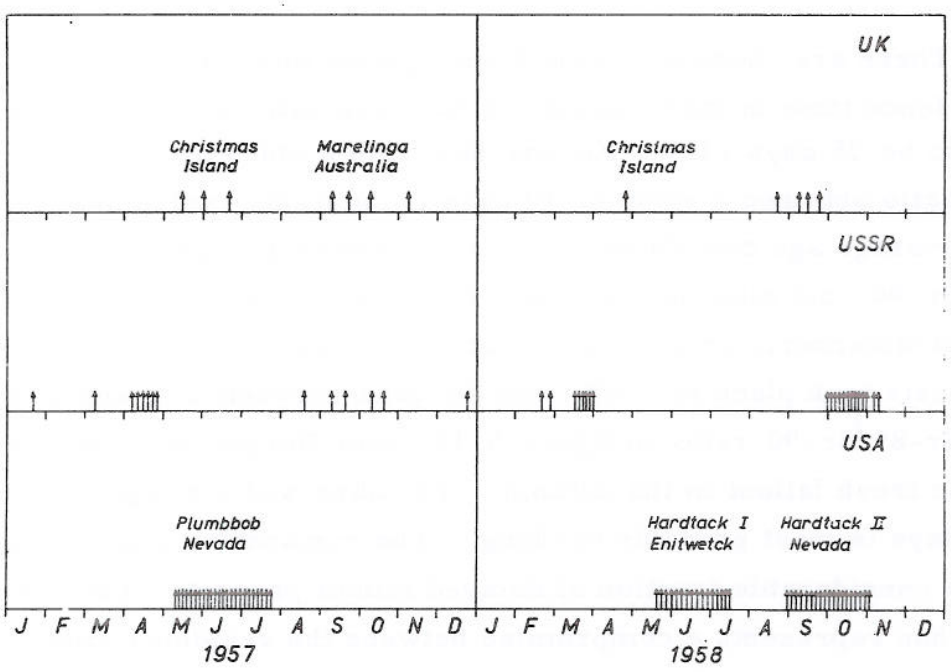

Figure 5.18 Nuclear detonations officially announced by USA, USSR and UK 1957 and 1958

distribution of Sr-89 over intervals of the Sr-89/Sr-90 ratio is given. Especially the peaks in the diagrams from Bergen and Mφsvann show clearly that the high Sr-89/Sr-90 ratios are due to fresh fallout. In a report from 1959, Martell (53) has stressed the importance of the time, place and intensity of an explosion in connection with evaluation of the stratospheric residence time. In figure 5.18 are indicated actual bomb tests known in 1958 according to Hardy and Klein 1959 (48). This figure shows that there are a number of possible sources for fresh fallout in the period of observation.

An exact evaluation of the stratospheric and tropospheric parts of the fallout is difficult. Some simplifications have to be made. Instead of using an average age for each sample, it is believed that a better interpretation of the data is obtained by assuming all Sr-89 to be due to tropospheric fallout of an average age of 35 days. In figure 5.19 the distribution of Sr-90 over intervals of the Sr-89/Sr-90 ratio is given, and the corresponding amounts of tropospheric Sr-90 are shown.

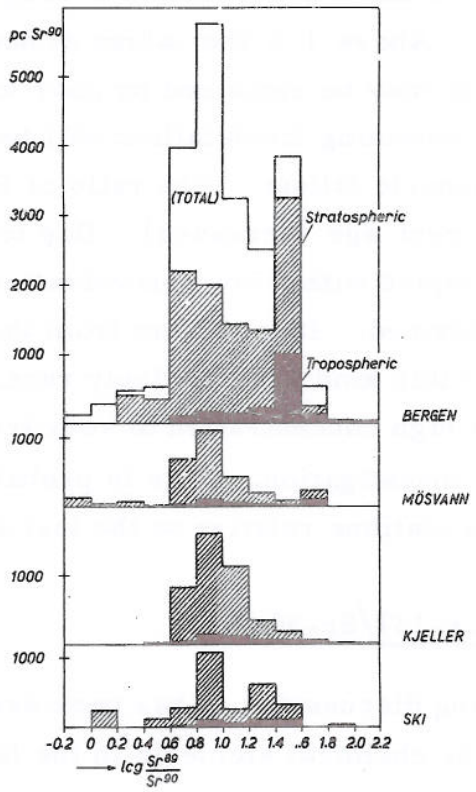

Figure 5.19 The distribution of pc Sr-90 in precipitation over intervals of the Sr-89/Sr-90 ratio, Norway 1957 and 1958

If the tropospheric residence time is assumed to be 100 days, the amount of tropospheric Sr-90 will be approximately doubled. Regardless of these residence times the main part of the Sr-90 fallout will be of stratospheric origin. The conclusion from this discussion therefore must be as pointed out by Stewart et al (33), that the main part, possible more than 90 per cent of the Sr-90 in the fallout is of stratospheric origin.

By comparison of figures 5.16 and 5.19 it is seen that the distribution of Sr-90 largely follows the amounts of precipitation, when observations for a whole year are compared. Minor deviations are observed, but these evidently show no systematic variation with the Sr-89/Sr-90 ratio. In figure 5.20 the distribution of the concentration of Sr-90 in precipitation from all sampling stations in Norway is given for intervals of the Sr-89/Sr-90 ratio. The curve indicates a constant average value corresponding to 5.8 mc/km^2/m. The same average value is obtained from the values in table 5.5. In figure 5.21 the logarithm of the concentration of Sr-89 has been plotted in the same way. In this case a straight line with a 45° slope is obtained for values of log Sr-89/Sr-90 below 1.8. This supports the previous conclusion of a constant average Sr-90 concentration in these samples. Above 1.8 the values of both Sr-90 and Sr-89 decrease. This may be explained by poor mixing in the troposphere. Normally air containing fresh fallout will be mixed with volumes of older, stratospheric fallout. The ratio of Sr-89/Sr-90 is thereby reduced (appearent age increased). Due to incomplete mixing of the air masses samples with a low concentration of fresh fallout are occationally obtained. It is evident from the diagram in figure 5.20 that samples of this kind are relatively rare. It is remarkable that samples with a high concentration of very fresh fallout do not seem to occur in the investigation. This is probably due to the position of the sampling stations relative to the test sites.

5.2.3 The ratio of Cs-137/Sr-90

In the preceeding discussions it has been assumed that no appreciable separation of the chemical elements in the fallout takes place in the atmosphere. This assumption has been discussed by Stewart et al (33) and by Storebø (54). Stewart et al determined the ratio of

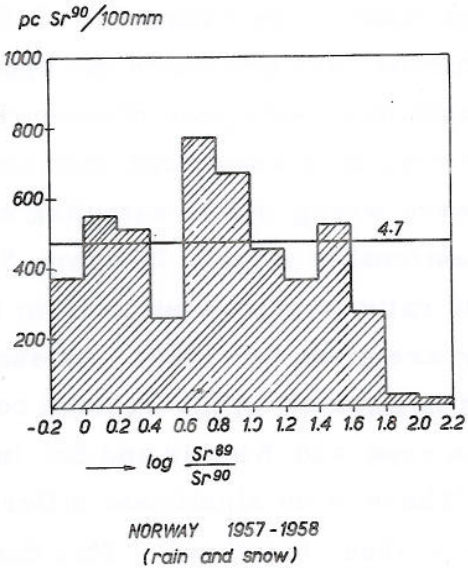

Figure 5.20 Average concentration of Sr-90 in precipitation over intervals of the Sr-89/Sr-90 ratio, Norway 1957 and 1958

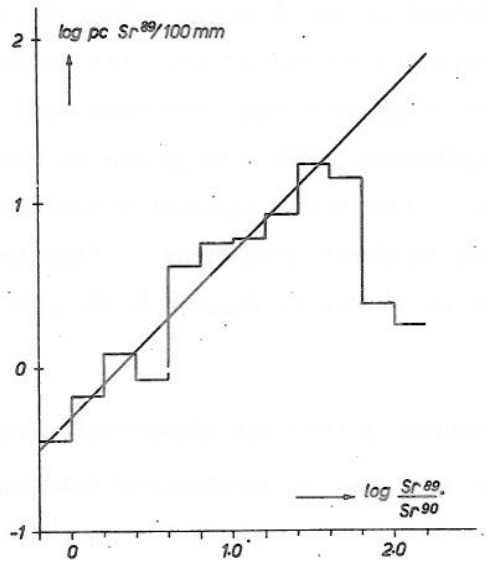

Figure 5.21 The log of the concentration of Sr-89 in precipitation over intervals of the Sr-89/Sr-90 ratio Norway 1957 and 1958

Cs-137/Sr-90 in precipitation at different geographical locations and found that this ratio in many cases deviated significantly from an assumed constant value in the range 1.0 - 1.5. From a study of available data Storebø (54) presented the hypothesis that Sr-90 is more easily brought down with precipitation than Cs-137. Later Stewart et al (36) have announced that some of their data in this connection were wrong due to sampling errors. These data are therefore of questionable value. In figure 5.22 the distribution of the Cs-137/Sr-90 ratio in precipitation from four different locations in Norway are given for 1958. As shown on the map in figure 2.1, Bergen is situated on the western coast, Møsvann in the central mountain area and Kjeller and Ski in eastern part of southern Norway. There is no significant difference in the Cs-137/Sr-90 ratio at these locations. This does, however, not exclude the possibility of a minor effect.

There remains, however, the remarkable fact that the Cs-137/Sr-90 ratio in the present data show considerable variations from an expected constant value. This observation has also been made by Stewart et al (36) and by Kuroda (47). The mean values for the different locations are given in table 5.5 and figure 5.23. The main part of the observed Cs-137/Sr-90 ratios are in the range 0.4 - 2.5. This spread is much larger than the combined errors from the radiochemical determination, see tables 5.1 - 5.4. In figures 5.24 and 5.25 the total amounts and the concentration of Cs-137 in precipitation 1958 are given as function of the Cs-137/Sr-90 ratio. The large spread of the data around the mean value is also evident in these diagrams. The time dependency of the Cs-137/Sr-90 ratio is shown in figure 5.26 and seems to be quite irregular.

The general impression is that the observed variations in the Cs-137/Sr-90 ratio are due to incomplete mixing in the atmosphere. In the first instance the fission products may not be evenly distributed in the primary cloud formed by the explosion. These primary variations may persist during the transport through the atmosphere. The fallout isotopes are partly formed in the atomic explosion, and partly through the disintegration of parent isotopes. As the cloud

cooles down, the isotopes will be picked up by the condensing materials from the bomb and its surroundings. They may also be expected to react with oxygen or water vapour to form oxides and hydroxides. Depending on the conditions, the chemical differences of the particles may give rise to a preferred precipitation of either Sr or Cs.

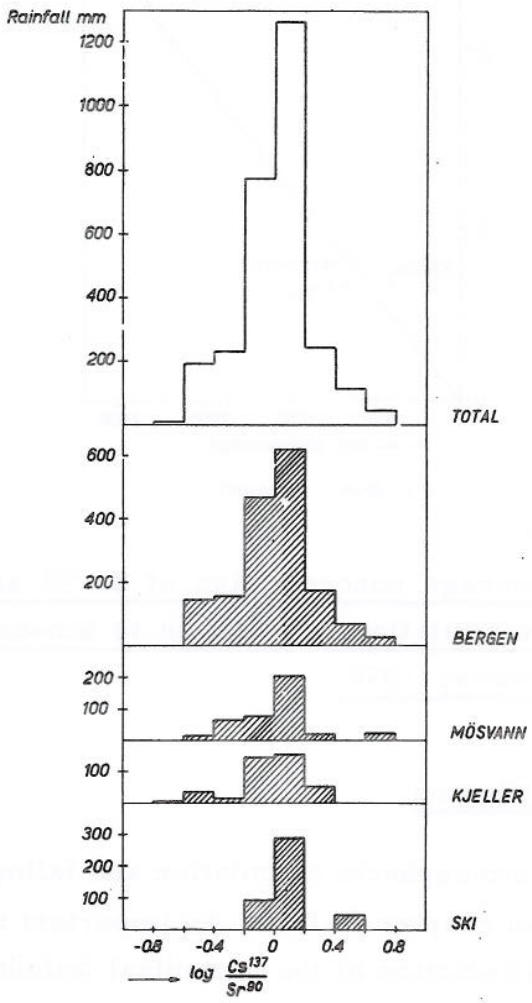

Figure 5.22 The distribution of the precipitation in mm over intervals of the Cs-137/Sr-90 ratio, Norway 1957 and 1958

The present investigation does not throw any light on the details of these possible processes, but since the mean value of the Cs-137/Sr-90 ratio is nearly the same from one location to another, the variations seem to be of a local character in space and time. The general impression is that they may be due to variations in the primary composition of the fallout particles. These variations are not

abolished during the transport through the atmosphere and they may become locally affected by short term chemical influence upon the precipitation process.

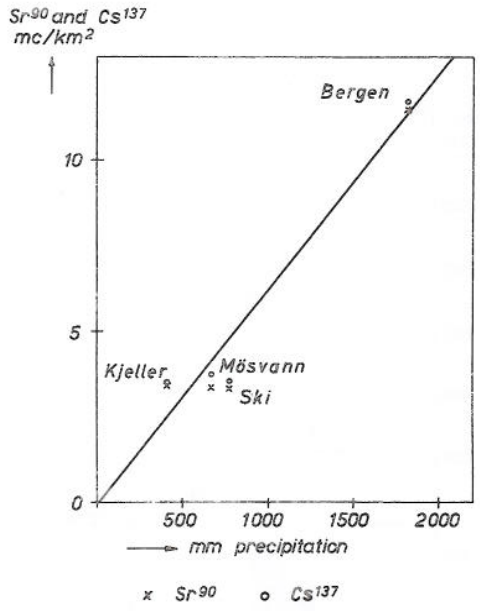

Figure 5.23 Average concentration of Sr-90 and Cs-137 in precipitation in relation to amount of precipitation, Norway 1958

5.2.4 Latitudinal distribution

The theories of atmospheric circulation and fallout deposition have been discussed in chapter 5.1.2. An important aim of these theories is the prediction of the latitudinal fallout distribution. According to both Libby and Stewart et al, the cumulative Sr-90 fallout at 60° northern latitude should, when corrected for rainfall, be low compared with observations at lower latitudes. The present results indicate that this is not the case, and in the following, experimental data and methods will be discussed.

The methods normally used for determination of total fallout are analysis of soil and cumulative precipitation. Sometimes snow layers from glaciers have also been used. In principle these methods look simple, but in practise there are serious difficulties, and only approximate agreement between different methods is obtained.

There is no equipment available which will readily measure the absolute amounts of rain. Standard equipment may, depending on local conditions, deviate by as much as 20 per cent. Difficulties are still larger with snow. This has been the reason for using the frames described in chapter 2.1 c for snow sampling.

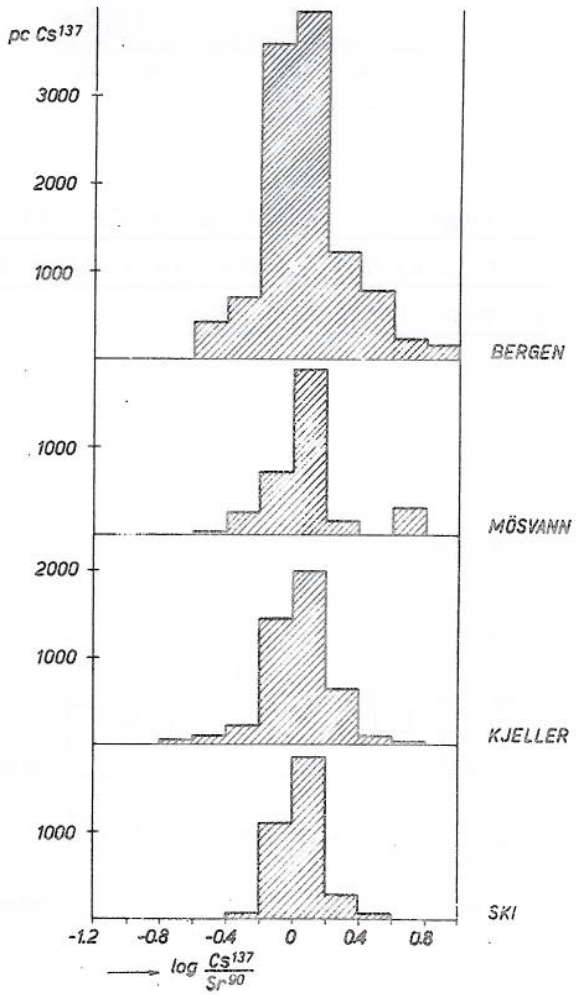

Figure 5.24 The distribution of pc Cs-137 in precipitation over intervals of the Cs-137/Sr-90 ratio, Norway 1957 and 1958

The turbulent deposition is not taken into proper account by ordinary meteorological sampling equipment. Experiments have shown that the double amount of fallout may be collected by replacing the standard pot with an artificial grass mat (38). Turbulent deposition may thus contribute with amounts which are not at all negligible as frequently supposed.

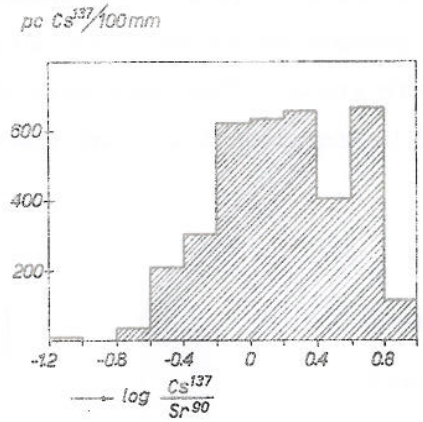

Figure 5.25 Average concentration of Cs-137 in precipitation over intervals of the Cs-137/Sr-90 ratio, Norway 1957 and 1958

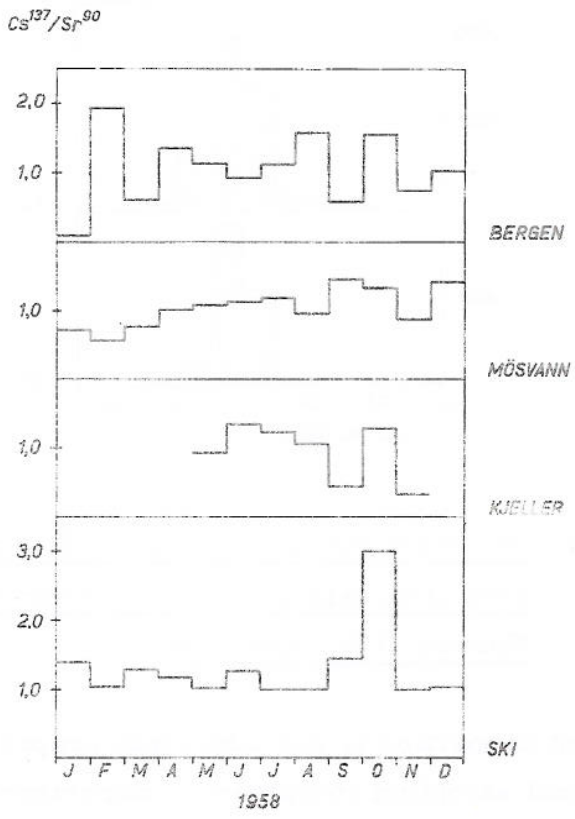

Figure 5.26 Variations in the Cs-137/Sr-90 ratio during 1958

Incomplete extraction of fallout material from samples of mineral soil may be the cause of considerable errors, and this difficulty can only be overcome through very elaborate procedures (1).

The sampling of soil offers a number of problems. One of these is leaching. The main part of the fallout normally is adsorbed in the top layer of 10 - 15 cm of the soil. A certain fraction, however, is leached out by the precipitation. Proof of this is the fallout content in rivers and lakes, the activity of which corresponds to 0.5 $mc/km^2/m$. When soil samples are taken to a depth of 10 - 15 cm some adsorption in deeper layers must be considered, and the amounts actually washed out from the top layers may be much higher. Due to differences in density and composition of an appearently uniform soil the fallout will not be evenly distributed in the surface layer. A large number of samples have to be taken to obtain a significant average.

Using glaciers for fallout cumulation measurements similar difficulties are met with. In our experience such measurements have proved very doubtful. Investigations of the migration of Sr-90 and Cs-137 in snow show that fast downward migration of the isotopes occur. The migration appearently takes place on the surface of snow crystals, probably in a microscopic liquid layer. The extent of this migration shows that the main part of the Sr-90 and Cs-137 is present in this surface layer.

To avoid some of these difficulties in the determination of cumulated amounts of fallout, an investigation of peat soil was started at this laboratory in 1957. Compared to mineral soil, peat soil is more easily analyzed as the low ash content gives a reasonable size of sample, which is easily dissolved in acid and still carries sufficient Sr-90 and Cs-137 activity for accurate radiochemical determination.

It is well established that peat soils act as ion exchangers with a high captive capacity. Our investigations have shown that the peat soil will fix the main part of the basic fallout activity in the upper 10 cm. In the present work deep ombrogenic peat bogs were selected for sampling. In such peat bogs there is no sideways transport of water. Layers of the peat bog down to 50 cm and

samples of the water from the peat have been analyzed. From these investigations, which are to be continued, preliminary data are given in tables 5.7 - 5.9. To these values must be added the amounts of Sr-90 and Cs-137 leached out by precipitation to obtain the true values. The amounts leached out so far may be estimated to a few mc/km^2 in the case of peat bogs.

The first estimates of total Sr-90 and Cs-137 fallout in Norway were made by Hvinden (51). The values were based on a few analysis of soil and snow from 1956. Later estimates have been made by adding amounts calculated from total beta activity measurements in precipitation. As seen from figure 5.10 these calculated values are too low, and the turbulent deposition has not been taken into account. By this approach a value of 10 - 12 mc $Sr-90/km^2$ were estimated for the total Sr-90 fallout at Kjeller up to the end of 1958.

Recently Alexander (1) has published values for the rate of Sr-90 deposition and the cumulation of Sr-90 in soil at different latitudes. His data are reproduced in figures 5.27 and 5.28, and they represent a general survey of the world-wide fallout situation. The period 1956 - 1958 does not show any decrease with increasing latitude in the northern hemisphere up to 80° northern latitude. Otherwise the data are very similar to the data given by Libby and by Stewart et al.

The specific data for the Norwegian samples are reproduced in table 5.6. It will be noticed that the measured data given by Alexander for Oslo and Bergen in 1958 are only a few mc/km^2 higher than the values estimated by Hvinden. According to the present precipitation measurements from Bergen 8 mc $Sr-90/km^2$ were brought down in the period November 1957 - October 1958. Analytical data for the period February 1957 - November 1957 are not available. A good estimate is probably 4 mc $Sr-90/km^2$ the same as for New York during the same period. The total amount brought down by precipitation in the period February 1957 - October 1958 may then be 12 mc $Sr-90/km^2$. In soil from Bergen Alexander finds an increment of 7.7 mc $Sr-90/km^2$. The total content of the soil in Bergen is found by Alexander (1) to be 21.3 mc $Sr-90/km^2$.

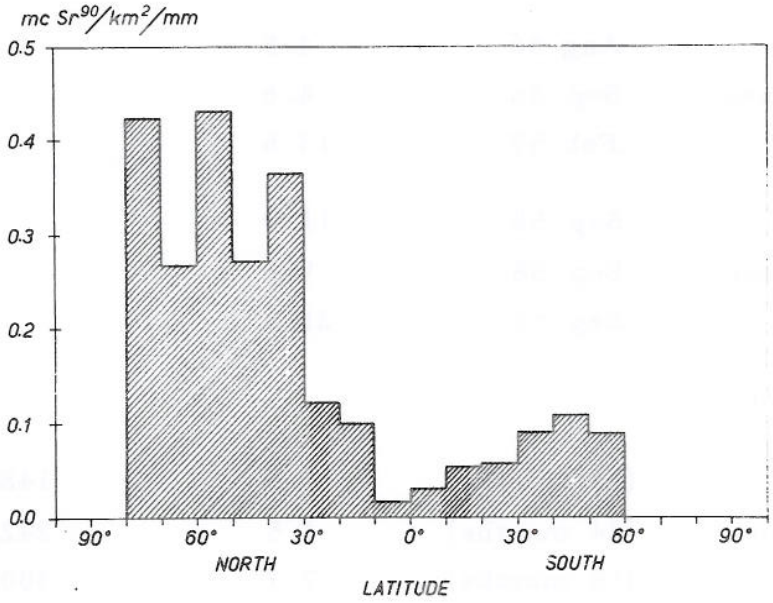

Figure 5.27 Rate of Sr-90 deposition in mc/km^2/mm of precipitation 1956 - 1958, Alexander 1959 (1)

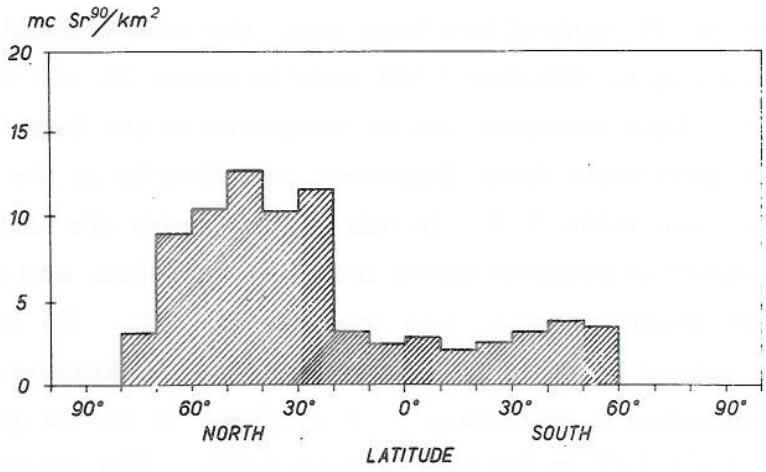

Figure 5.28 Cumulation of Sr-90 in soil 1958, Alexander 1959 (1)

Sampling place	Date collected	Sr-90 mc/km^2	Precipitation mm
Oslo	Aug 56	4.5	
Lake Finse	Sep 56	4.6	
Bergen	Feb 57	13.6	
Oslo	Sep 58	11.0	
Lake Finse	Sep 58	9.1	
Bergen	Sep 58	21.3	
Increment			
Oslo	(24 months)	6.5	1480
Lake Finse	(24 months)	4.5	2420
Bergen	(18 months)	7.7	3000

Table 5.6 Cumulation of Sr-90 in soil, Alexander 1959 (1)

This indicates that considerable leaching has taken place. Only about half the Sr-90 brought down with precipitation has been retained in the upper 15 cm of the soil. If it is assumed that one half of the Sr-90 content has been lost, the total amount of Sr-90 brought down up to October 1958 may be some 32 mc Sr-90/km^2 in Bergen. This estimate can be compared to the data from analysis of peat soils from Bogetveit and Brurås in the neigborhood of Bergen, see table 5.7. In this table results are also given from a number of samples taken from the coastline and eastwards through the mountain area, see map figure 2.1. To the measured values in table 5.7 must be added the amount leached out. This is estimated to minimum 1 - 2 mc/km^2 of Sr-90 and 2 - 4 mc/km^2 of Cs-137 in the case of peat soils. The amount of Sr-90 actually brought down to the ground up to the end of 1958 on the western coast of Norway then sums up to 30 - 40 mc/km^2.

Since the autumn of 1957 the cumulation of Sr-90 and Cs-137 has been studied in two selected areas, one at Ski and one at Mφsvann,

see map figure 2.1. The average data from Møsvann are given in table 5.8 below.

Sampling place	Sr-90 mc/km^2	Cs-137 mc/km^2	Precipitation mm	
Radøy	19.6	17.4	1670	(Herdla)
Brurås	30.0	34.0	1670	(- " -)
Bogetveit	27.1	25.4	1670	(- " -)
Tokagjel	25.0	28.9	2750	(Norheimsund)
Etne	25.0	25.4	1810	
Ulvik	18.5	18.7	1740	(Granvin)
Maurset	12.0	14.5	670	(Ustaoset)
Seljestad	25.7	22.0	1180	(Svandalsflona)
Haukeli	28.8	25.2	1180	(- " -)
Geilo	9.5	12.8	680	

Table 5.7 Data from peat soil analysis sampled in Norway October 1958

Date collected	Sr-90 mc/km^2	Cs-137 mc/km^2
October 1957	26.3	21.5
October 1958	38.9	33.0
Increment	12.6	11.5
Measured in precipitation	3.4	3.8

Table 5.8 Increment of Sr-90 and Cs-137 in peat soil sampled at Møsvann October 1957 and October 1958

It is believed that the difference between the increment in the soil and the amounts measured in precipitation is due to turbulent deposition. The peat bog is situated in an open area at 1000 m above sea level, and there would be small possibilities for the formation of stagnant air volumes over the sampling area.

The results from Ski give a somewhat different picture. Two peat bogs separated by a distance of 2 km have been studied. One (A) is situated in an open area on the top of a hill, the other (B) is closed in by small hills covered by dense wood of spruce. The data are given below in table 5.9.

Date collected	(A) Sr-90 mc/km^2	(A) Cs-137 mc/km^2	(B) Sr-90 mc/km^2	(B) Cs-137 mc/km^2
October 1957	10.7	7.4	7.6	9.0
October 1958	27.7	25.1	13.0	11.7
Increment	17.0	17.7	5.4	2.7
Measured in precipitation	3.0	3.6	3.0	3.6

Table 5.9 Increment of Sr-90 and Cs-137 in peat soil sampled at Ski October 1957 and October 1958

In the shielded area (B) the amounts brought down with precipitation approximately make up for the soil increment. In the unshielded area (A), however, much more Sr-90 has been added to the peat soil than can be ascribed to precipitation alone. Turbulent deposition in this unshielded area seems to be of great importance.

In general the data collected seem to indicate that in open areas covered with vegetation turbulent deposition is an important factor in this country, and the less the precipitation, the greater the contribution from turbulent deposition becomes.

The data given for peat soils are preliminary and the values given may be adjusted when more analytical results are available, but it is expected that only minor modifications will have to be made in the general conclusions.

The total amount of Sr-90 fallout at 60° northern latitude seems to be considerably higher than predicted by the current theories Precise measurements concerning fallout in soil at higher latitudes are not available. Hvinden (52) has measured the total beta activity in precipitation at a number of stations from 58° - 70° northern latitude. Correcting for the amount of precipitation, he finds that a reduction of only 30 per cent with increasing latitude has taken place in this region. If the same samples are measured 6 months later insignificant variations with latitude are observed. This may indicate higher values of both Sr-90 and Cs-137 than are expected from published data elsewhere. An extended investigation of peat soils would be of considerable interest in this case.

Dealing with the latitudinal distribution of fallout it is obvious that many factors are working together to give the final result : The cumulation of fallout in a given area. One of these factors is the amount of precipitation. Another is the circulation between troposphere and stratosphere. These factors have been taken into account in the present theories. Others, like turbulent deposition, the incomplete mixing in the atmosphere, and possible effects due to chemical reactions of the fallout material have been neglected. As mentioned Martell (53) has stressed the importance of time, strength and place of injection of the bomb into the atmosphere.

The reason for this selection of two of the factors in question has been the search for one or a few predominant effects which would explain the main features of fallout deposition. Some of these, for example the amount of precipitation and the general circulation of the atmosphere have been pointed out, but the present investigations show that these are not sufficient to explain all the observed facts.

Instead of selecting a few factors as being of major importance, it may be interesting to see what the result will be if air currents and other factors are not specified, but lumped together in a simple theory of eddy diffusion. If the radioactive material from an atomic explosion

in the northern hemisphere in the first instance is distributed in a zone at the latitude where the explosion took place, one may assume that an eddy diffusion takes place towards the pole and the equator. If the height of the atmospheric layers where this diffusion takes place is h, r is the radius of the earth ($r \gg h$) and α the latitude, then the cross section through which the diffusion takes place is :

$$A = 2\pi r h \cos\alpha \qquad (5.1)$$

and the diffusion equation becomes :

$$\cos\alpha \frac{\partial c}{\partial t} = \frac{D}{r^2} \cdot \frac{\partial}{\partial x} \left(\cos\alpha \frac{\partial c}{\partial x}\right) \qquad (5.2)$$

A remarkable property of this diffusion equation is that the cross sectional area A becomes zero at the pole and has a maximum $2\pi r h$ at the equator. Variations in h are not taken into account. This means that if a bomb is fired at middle latitudes, the air concentration will be much more reduced with time towards equator than towards the pole. The equation has been solved numerically for the limiting values :

$$t = 0, \quad c = c_o, \quad \text{for } \alpha = \alpha_o$$

$$c = 0, \quad \text{for } \alpha \neq \alpha_o$$

$$\text{and} \quad \frac{\partial c}{\partial x} = 0, \quad \text{for } \alpha = \pm \frac{\pi}{2}$$

The resulting types of concentration curves are shown in figure 5.29, and it is observed that they represent the general trend of the observations.

The way in which the experimental latitudinal fallout distribution curves have been drawn by many authors seem to indicate that the curve slopes down to zero at the pole. It may well be that reduced values will be found towards the pole, but the tangent of the curve at the pole must always be horizontal. To obtain a closer relation to the cumulated fallout data, curves of the type given in figure 5.29 should be added up for all explosions over an appropriate period of time.

If data from suitable test series were known, the diffusion coefficient might be determined and used to predict the fallout contribution on a world-wide scale. This is not possible with the data available to us.

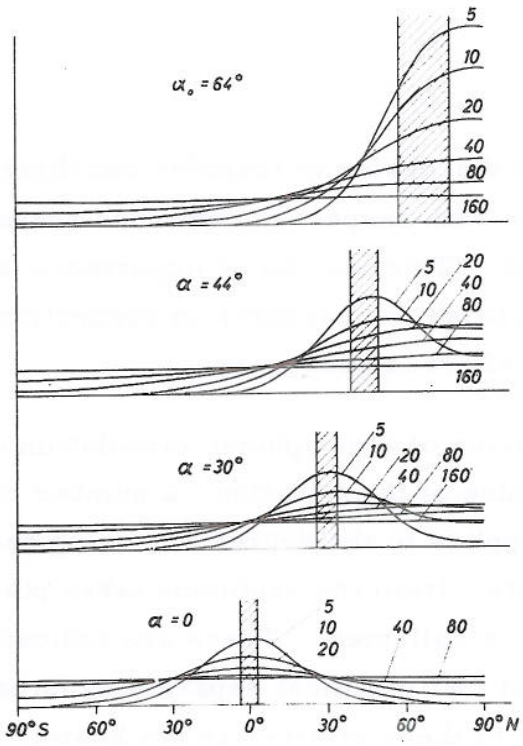

Figure 5.29 Concentration curves obtained by diffusion in a spherical shell, numbers indicate relative time

The diffusion theory may be applicable to both tropospheric and stratospheric fallout, the distribution of the resulting fallout will, however, be very different in the two cases. Since the average residence time in the troposphere is short, one month or less, most of the radioactivity will be deposited before the zone has broadened much. In the case of stratospheric fallout the rate of deposition is much slower, and the distribution curves in figure 5.29 may correspond to the main fraction of the stratospheric fallout. The diffusion theory given here is the simplest possible. It may be worked out more in detail by introducing variable rates of transfer from the stratosphere and by taking into account local variations in precipitation and turbulent deposition.

In the case of short term observations this diffusion theory will of course not be valid. On the other hand, when long term observations are made, the diffusion theory may be expected to reproduce the observations more and more closely. A similar approach has been suggested by Machta (55) in connection with estimates of the future atmospheric contamination from atomic reactors.

5.3 Conclusions

In concluding the discussion of transfer and distribution of fallout, some suggestions with respect to continued research in this field will be mentioned. This may be of importance as there are unlimited possibilities for research in connection with fallout and concentration of effort is necessary.

The present theories of atmospheric circulation only give an approximate picture of the situation, a number of details are lacking. This applies to the degree of mixing and separation of the fallout components, from the explosion takes place, till the time when the sample is collected. There are indications that the mixing is incomplete and that chemical separations do take place. As long as the magnitude of these effects are not known, little confidence can be placed in information given about single isotopes calculated from total activities. It further applies to the rates of transfer from the stratosphere to the troposphere and from the troposphere to the ground. The suggestion by Libby (35) and the work of Kuroda (47) have shown promising results in utilization of the Ba-140/Sr-90 ratio for this study. The seasonal variations in the amount of fallout isotopes as pointed out by Stewart et al, and the short term variations in fallout composition discussed in this paper, may be expected to give still more valuable information if more isotopes could be covered. These and similar investigations call for frequent sampling and detailed analysis.

Total cumulative data of Sr-90 fallout as determined recently by Alexander by soil analysis are of fundamental importance for calculation of the future contamination of the biosphere. As pointed out by him our present knowledge is limited to a general survey of

the world-wide distribution. Large local variations have been noticed, but a detailed understanding which would make local prediction possible is still lacking. Also the sampling methods may be questioned. These problems call for samples from a large number of places once or twice a year, and detailed analysis.

In the future it may therefore be recommended that in general the efforts, when limited, should be worked in two directions. One may be called "fine structure" studies which will cover detailed investigations with frequent sampling in space and/or time. The other type may be called "cumulative" studies. These may be carried out by monthly sampling of precipitation and airborne materials and by yearly sampling of soil from a comprehensive number of stations. It is believed that studies of a more intermediate type will be of little value.

Common to all these investigations should be analysis of a number of different fallout isotopes. Total activity determinations may in some cases be useful as a guide, but they can not be relied upon to give a true picture of the complex of phenomena which constitutes the fallout distribution.

6 FALLOUT IN TAP WATER

6.1 Introduction

Tap water must be considered as a possible source of radioactive fallout for the human population. Its content of fallout is usually very low since the fallout isotopes are adsorbed to soil and other organic and inorganic materials through which the water migrates. Extremely low concentrations of fallout are found in water from deep wells, since this water normally passes a long distance. through the ground. In surface reservoirs the contamination will depend upon the properties of the precipitation area, as for instance the relative size of the reservoir surface, the adsorptive properties of the ground and the amount of soluble materials which can act as carriers. The duration of the contact between water and soil also plays a part. Water from cisterns may become contaminated to the same degree as precipitation, but it has

been found that even collecting vessels are able to retain a considerable fraction of the fallout. This phenomenon is one of the sources of error mentioned in connection with sampling of precipitation, see chapter 2.1.

The retention of fallout in the ground is due to adsorption, to ion exchange and to biological metabolism taking place all the way from precipitation to the tap. The adsorption and ion exchange capacity of soils may be very variable. In figure 6.1 results are given from some laboratory tests performed with Sr-89 and Cs-137. It is shown that clay is a most effecient material for decontamination of water. Humus also has a considerable ion exchange capacity, and the efficiency of different soils to decontaminate water is mainly given by their contents of humus and clay minerals, especially those exhibiting a platestructure, like bentonite and vermiculite. Coarse gravel and sand have little or no ability to retain fallout.

The decontaminating effect of clay on tap water has been studied in great detail by Lacy (56), who found that clay can remove Ce-141 and 144, Pr-144, Zr-95, Nb-95, Ba-140, La-140, Sr-90 and Y-90, while Ru-106 and Rh-106 are only partly removed, and I-131 is not captured at all. Similar results have been obtained by Wright and Monahan (57) in experiments with vermiculite.

6.2 Experimental data and discussion

The present investigations have been made on surface water, as this is the main source of tap water in Norway. The contents of Sr-90, Sr-89 and Cs-137 have been analysed in tap water sampled in 1957 and 1958 at Kjeller in the eastern part of Norway and at Stavanger and Bergen in the western part, see map figure 2.1. The results are given in tables 6.1 - 6.3 and in figures 6.2 - 6.4. A study of the total beta activity in tap water samples from all over Norway has been made by Hvinden (51, 52). His results for 1957 and 1958 are reproduced in figure 6.5.

Generally, the concentration of the isotopes Sr-90 and Cs-137 in tap water in Norway are one tenth or less of the concentration in precipitation. Table 6.4 gives the average contents of Sr-90 and

Cs-137 in tap water for the periods of sampling in 1957 and 1958 at the three locations.

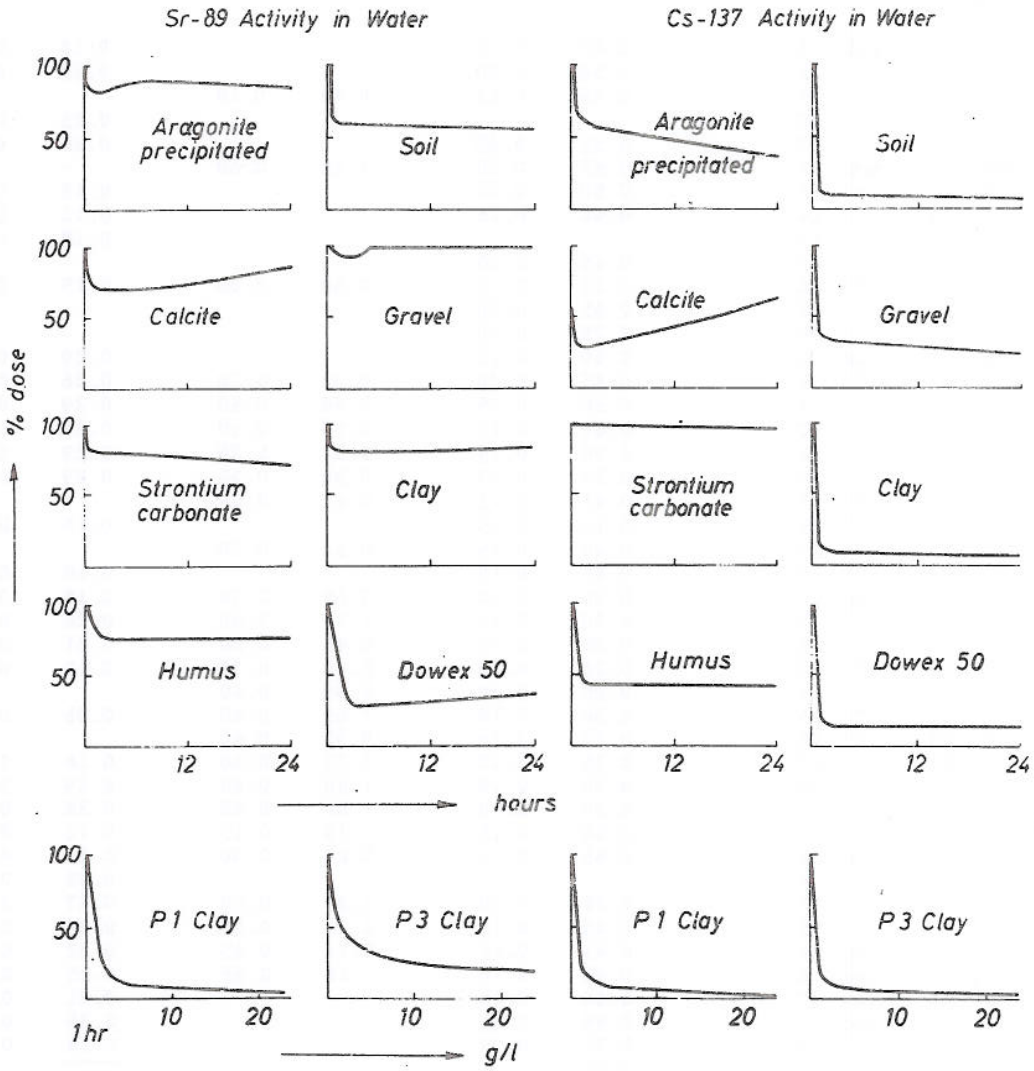

Figure 6.1 Decontamination experiments on tap water

There is a marked increase of the concentration of Sr-90 and Cs-137 in tap water from 1957 to 1958 at Bergen and Kjeller. At Stavanger only 4 observations are available from 1957. These samples have been collected in November and December, and the average values given cannot be considered significant for the year.

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Feb 4 - Feb 5	0.40	0.15			0.18	0.06
" 10 - " 12	0.50	0.20			0.29	0.12
" 14 - " 15	0.35	0.15	0.44	0.18		
" 19 - " 20					0.15	0.06
" 26 - " 27	0.55	0.20			0.24	0.08
Mar 1 - Mar 2	0.45	0.20	1.12	0.60		
" 11 - " 12	0.50	0.20			0.15	0.06
" 13 - " 14	0.40	0.15			0.12	0.06
" 18 - " 19					0.18	0.06
" 20 - " 22	0.45	0.20				
" 25 - " 26	0.35	0.15	0.68	0.60	0.15	0.06
" 27 - " 28	0.45	0.20				
" 29 - " 30	0.75	0.40				
Apr 1 - Apr 3	0.40	0.15			0.29	0.12
" 3 - " 5	0.45	0.20	0.32	0.20	0.26	0.12
" 6 - " 8	0.50	0.15	0.74	0.40	0.29	0.12
" 8 - " 10	0.40	0.15	0.96	0.40	0.21	0.08
" 10 - " 12	0.50	0.15	0.51	0.20	0.29	0.12
" 13 - " 15	0.25	0.10	0.86	0.35	0.29	0.12
" 15 - " 17	0.45	0.15	0.47	0.20		
" 23 - " 25	0.10	0.05			0.15	0.06
" 25 - " 27	0.40	0.15	0.53	0.20		
" 27 - " 29	0.45	0.15			0.18	0.06
" 30 - May 2	0.30	0.10	0.68	0.30	0.18	0.06
May 4 - " 6	0.20	0.10	1.13	0.50	0.26	0.08
" 6 - " 8	0.35	0.15	0.45	0.20	0.21	0.08
" 8 - " 10	0.20	0.10	0.60	0.30	0.15	0.06
" 11 - " 13	0.25	0.10	0.90	0.40		
" 13 - " 15	0.30	0.10	1.65	0.40	0.06	0.06
" 18 - " 20	0.20	0.10	0.95	0.40		
" 21 - " 23	0.25	0.10	1.73	0.40	0.18	0.12
" 23 - " 25	0.30	0.10	1.80	0.40	0.29	0.12
" 27 - " 29	0.30	0.10	1.80	0.40	0.32	0.12
" 29 - " 31	0.50	0.15	1.14	0.15	0.12	0.06
Jun 3 - Jun 5	0.45	0.10	0.80	0.30	0.44	0.06
" 5 - " 7					0.35	0.08
" 11 - " 13	0.35	0.10	1.30	0.50	0.47	0.12
" 13 - " 15	0.45	0.10	1.24	0.50	0.35	0.08
" 23 - Jul 26	0.41	0.15	1.12	0.45	0.32	0.08
Aug 6 - Aug 8	0.45	0.10	1.10	0.45	0.35	0.06
Nov 18 - Nov 27	0.40	0.05			0.21	0.12
" 27 - Dec 13	0.48	0.05			0.35	0.07
Dec 13 - " 29	0.27	0.07			0.16	0.03
Average	0.39				0.24	
1958						
Dec 29 (57) - Jan 24	0.58	0.10			0.27	0.05
Jan 24 - Feb 12	0.71	0.12			0.38	0.04
Feb 12 - Mar 4	0.39	0.05			0.31	0.04
Mar 6 - " 21	1.67	0.10	13.40	2.90	0.43	0.05
" 23 - Apr 23	0.42	0.06			0.49	0.07
May 24 - Jun 14	0.23	0.03	0.60	0.20	0.50	0.05
Average	0.67				0.40	

Table 6.1 Sr-90, Sr-89 and Cs-137 in tap water sampled at Kjeller, Norway 1957 and 1958

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Sep 19 - Sep 22	0.30	0.05			0.76	0.06
" 24 - " 27	0.20	0.05	1.20	0.50	0.82	0.12
Oct 1 - Oct 4	0.40	0.05	1.90	0.75	0.18	0.03
" 7 - " 12	0.50	0.05	2.40	1.00	0.12	0.03
" 14 - " 18	0.45	0.05	1.30	0.50	0.15	0.03
" 21 - " 26	0.40	0.05	2.70	1.10	0.35	0.06
" 28 - Nov 5	0.55	0.05	1.90	0.75	0.41	0.03
Nov 5 - " 27	0.65	0.05	1.10	0.45	0.35	0.03
Average	0.43				0.39	
1958						
Mar 17 - Mar 20	0.47	0.05	1.10	0.30	0.40	0.04
" 24 - " 28	0.68	0.05	1.02	0.40	0.55	0.04
Apr 21 - Apr 24	0.69	0.06			0.51	0.03
" 29 - May 3	0.73	0.07			0.41	0.04
May 5 - " 9					0.77	0.06
" 19 - " 22	0.83	0.06			0.91	0.13
" 27 - " 30	1.22	0.12			0.63	0.20
Jun 2 - Jun 5	1.23	0.12			0.43	0.07
" 9 - " 12	0.50	0.03	1.15	0.32	0.41	0.04
" 16 - " 19	0.78	0.06			0.34	0.03
" 24 - " 27	0.83	0.07			0.43	0.04
Jul 1 - Jul 4	0.79	0.07				
" 6 - " 9	0.87	0.05				
" 14 - " 17	1.70	0.13				
" 20 - " 23	0.93	0.07				
" 28 - " 31	1.28	0.11				
Aug 3 - Aug 6	0.98	0.10			0.47	0.04
" 11 - " 15	1.49	0.08			0.47	0.03
" 17 - " 20	1.61	0.11			0.21	0.02
" 26 - " 29	0.61	0.05	1.33	0.29	0.45	0.05
Sep 8 - Sep 11	1.17	0.10				
" 15 - " 18	0.99	0.08				
" 22 - " 25	0.47	0.04			1.04	0.08
" 30 - Oct 2	0.54	0.05	2.25	0.50	0.51	0.06
Oct 6 - " 9	0.71	0.06	1.66	0.54	0.47	0.05
" 13 - " 16	0.24	0.03	5.51	0.38	0.57	0.05
" 20 - " 23	0.84	0.05	1.16	0.46		
" 27 - " 30	0.41	0.04	3.16	0.36	0.46	0.04
Nov 3 - Nov 6	0.53	0.05	3.45	0.39	0.40	0.04
" 18 - " 21	0.68	0.04	3.69	0.59	0.42	0.05
" 24 - " 27	0.84	0.11			0.44	0.05
Dec 1 - Dec 4	0.99	0.07			0.40	0.04
" 8 - " 11	0.64	0.05	3.64	0.49	0.37	0.03
" 22 - " 25	0.61	0.05	4.56	0.47	0.45	0.04
Average	0.84				0.50	

Table 6.2 Sr-90, Sr-89 and Cs-137 in tap water sampled at Bergen, Norway 1957 and 1953

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Nov 12 - Nov 25	0.15	0.05			0.24	0.03
" 25 - Dec 7	0.52	0.05			0.45	0.03
Dec 7 - " 16	0.48	0.06			1.03	0.06
" 16 - " 24	0.44	0.05			3.18	0.20
Average	0.40				1.23	
1958						
Jan 7 - Jan 15	0.26	0.05				
" 16 - " 23	0.22	0.05			1.05	0.12
" 23 - " 31	0.37	0.05			0.39	0.04
Feb 4 - Feb 14	0.33	0.08	1.24	0.20	0.66	0.11
" 14 - " 24					0.55	0.04
" 24 - Mar 4					0.81	0.07
May 10 - May 15	0.53	0.03			0.76	0.06
" 16 - " 21	0.99	0.07				
" 21 - " 25					0.77	0.06
" 27 - " 30	0.46	0.09				
Jun 3 - Jun 6	0.55	0.03			0.47	0.01
" 11 - " 17	0.51	0.04			0.76	0.06
" 18 - " 23	0.73	0.04			0.65	0.05
Aug 18 - Aug 20	0.60	0.05				
" 22 - " 26	0.55	0.05			0.42	0.03
" 26 - " 29	0.56	0.06				
" 30 - Sep 3	0.94	0.06			0.60	0.06
Sep 4 - " 5	0.70	0.06			0.73	0.05
" 6 - " 9	1.39	0.10			1.40	0.16
" 22 - " 24	0.82	0.06			0.89	0.08
" 24 - " 26	0.94	0.07			0.98	0.09
" 26 - " 30	0.66	0.07			0.81	0.06
" 30 - Oct 2	0.67	0.08			0.62	0.06
Oct 2 - " 4	0.88	0.06			0.63	0.06
" 6 - " 8	0.75	0.06	1.78	0.54	0.78	0.08
" 8 - " 10	1.62	0.10			0.42	0.06
" 10 - " 14	0.42	0.05	2.74	0.53	0.84	0.08
" 14 - " 17	0.72	0.08				
" 17 - " 21	0.46	0.04	2.59	0.37	1.14	0.10
" 21 - " 23	0.22	0.03			1.09	0.08
" 23 - " 25	0.79	0.06			0.80	0.03
" 27 - " 29	0.33	0.04			1.13	0.06
Nov 1 - Nov 4	0.34	0.04			0.65	0.05
" 4 - " 6	1.23	0.20	1.51	0.87	1.14	0.13
" 8 - " 11	1.62	0.25			2.81	0.22
" 11 - " 14	0.72	0.12			1.00	0.07
" 14 - " 18	0.65	0.13			1.09	0.07
" 18 - " 20	1.68	0.19			1.12	0.08
" 20 - " 23	1.20	0.16			1.59	0.07
" 24 - " 26	0.80	0.09			1.13	0.10
" 26 - " 28	1.07	0.10			0.90	0.06
Dec 11 - Dec 16	1.67	0.11			0.88	0.05
" 16 - " 20	1.27	0.11			0.98	0.06
Average	0.78				0.90	

Table 6.3 Sr-90, Sr-89 and Cs-137 in tap water sampled at Stavanger, Norway 1957 and 1958

Sampling place	Sr-90 pc/l	Cs-137 pc/l	Total beta activity pc/l
1957			
Bergen	0.43	0.39	
Stavanger	(0.40)	(1.23)	
Kjeller	0.39	0.24	
Average	0.40	0.62	29
1958			
Bergen	0.84	0.50	
Stavanger	0.78	0.90	
Kjeller	0.67	0.40	
Average	0.76	0.60	41

Table 6.4 Average content of Sr-90 and Cs-137 in tap water sampled in Norway 1957 and 1958
The total beta activity in tap water is also given, Hvinden (51, 52)

The present results may be compared to data given by Hardy and Klein (48) for New York, reproduced in figure 6.6. It will be seen that the New York tap water contains less Sr-90 than has been found in this country, the average for 11 months in 1958 beeing 0.12 pc Sr-90/l.

No average values for Sr-89 are given since analysis has been carried out less regularly. In general the values range from zero to a few pc/l, see tables 6.1 - 6.3.

In a surface reservoir the fallout isotopes are partly precipitated directly into the reservoir and partly leached out of the ground in the precipitation area. It is of interest to estimate what fractions of the fallout finally reach the tap water in these ways. The data from

Bergen can be used for this purpose. The tap water from Bergen is from the lake Svartediket, which is situated between the mountains on the eastern side of the town. The precipitation area has an extension of 12.13 km^2, and the precipitation data discussed in the previous chapter are from this area. This reservoir has been thoroughly investigated by Gaarder and Vindenes 1933 (58) and by Gaarder and Theisen 1953 (59). It is described as a generally steep sloping ground which consists mostly of bare rocks with a scanty covering of plants, mainly lichen, mosses and heather. The ground consists of granite and gneiss granite and the soil is generally acid raw humus. The water which runs into the lake Svartediket is a pronounced surface water with pH about 5.4. The mean composition is :

Ca	Mg	Fe	Cl	SO_4	SiO_2	Humus ($KMnO_4$)
0.45	0.54	0.22	5.14	0.29	1.18	6.64 mg/l

In the area there are two lakes, Svartediket and Isdalsvannet, connected with a tunnel. The combined area is 0.2782 km^2. The outlet is from the lake Svartediket, the volum of which at normal level is $3300 \cdot 10^6$ liter. The water of the reservoir is replaced twice a year.

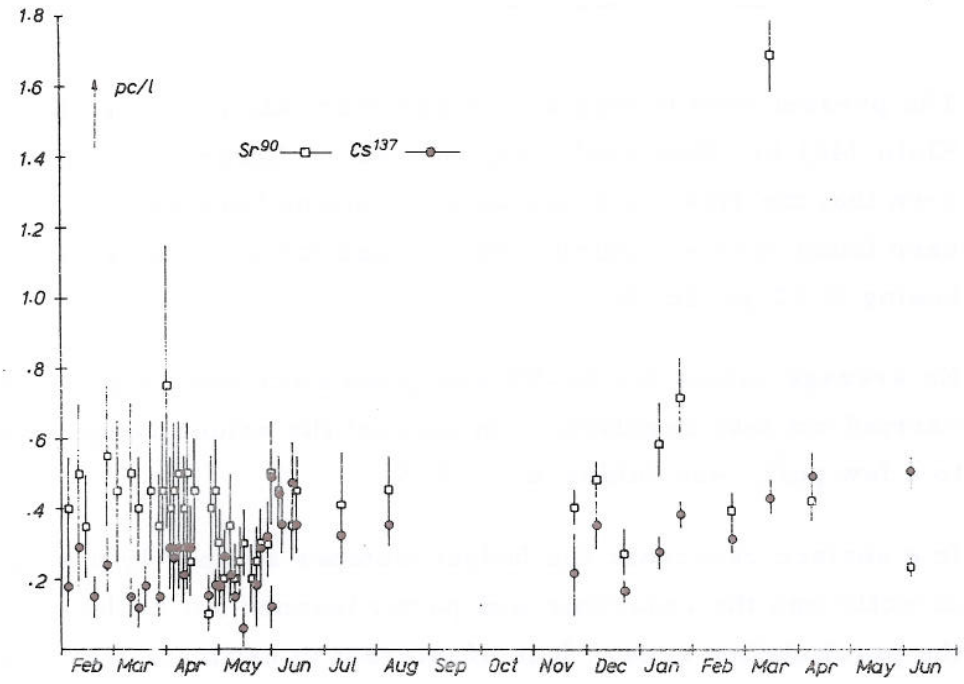

Figure 6.2 Sr-90 and Cs-137 in tap water from Kjeller, Norway 1957 and 1958

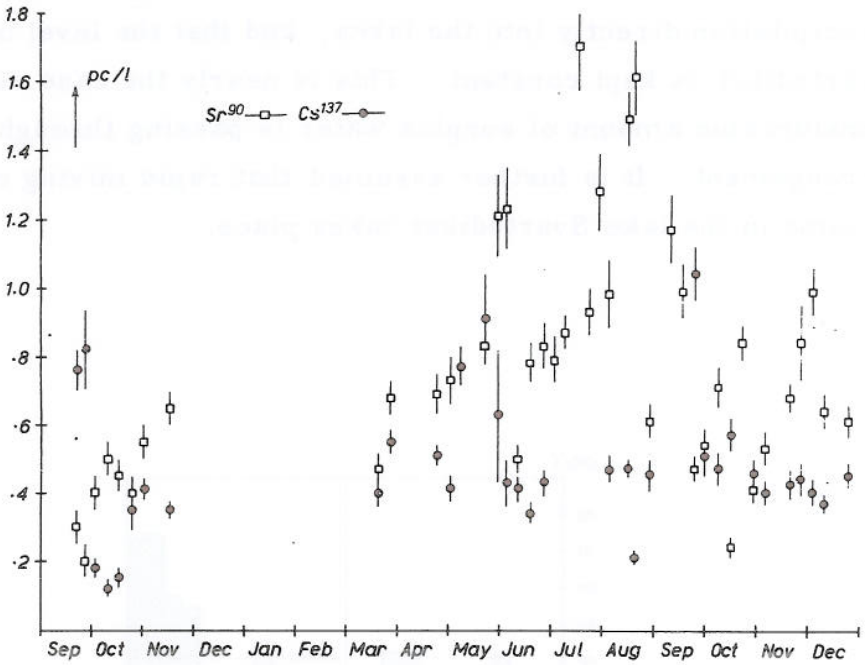

Figure 6.3 Sr-90 and Cs-137 in tap water from Bergen, Norway 1957 and 1958

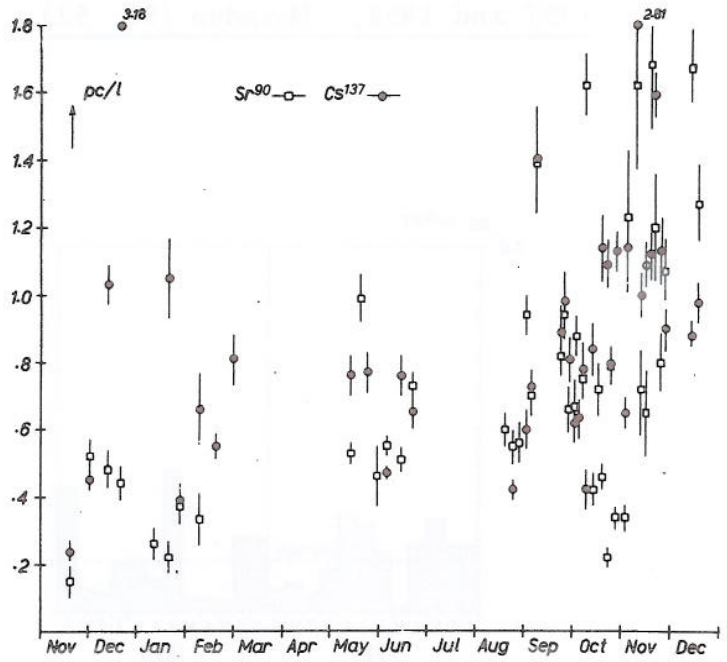

Figure 6.4 Sr-90 and Cs-137 in tap water from Stavanger, Norway 1957 and 1958

In the following calculation it will be assumed that the fallout found in the tap water is due only to the amount brought down by precipitation directly into the lakes, and that the level of the lake Svartediket is kept constant. This is nearly the case as a considerable amount of surplus water is passing through an overflow arrangement. It is further assumed that rapid mixing of the water volume in the lake Svartediket takes place.

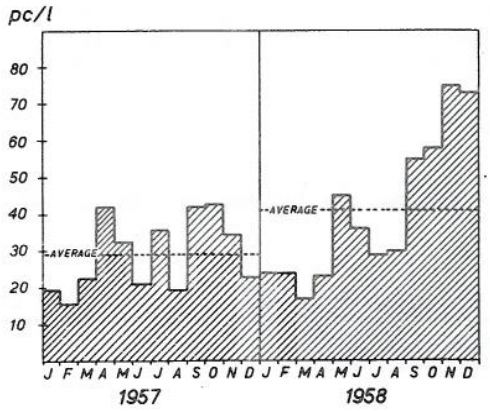

Figure 6.5 The total beta activity in tap water sampled in Norway 1957 and 1958, Hvinden (51, 52)

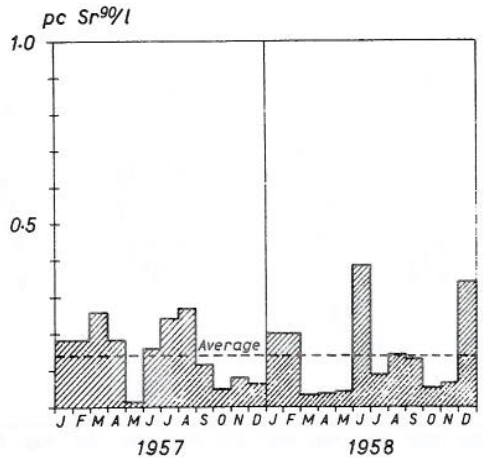

Figure 6.6 Sr-90 in tap water sampled in New York 1957 and 1958 Hardy and Klein 1959 (48)

By using this model the activity change dc in the lake of any fallout isotope caused by a precipitation dx may be calculated. The total precipitation area is called A, and the area of the lake B.
A concentration a of the fallout isotope in the precipitation gives a concentration $b = a \cdot B/A$ of the water running into the lake. The constant volume of the lake is V. The ventilation equation gives:

$$V\,dc = (b - c) \cdot A\,dx \qquad (6.1)$$

The initial condition is $c = c_o$ for $x = 0$. In the present case the monthly mean value will be used for b and the equation integrated in steps for each month. The solution then becomes for b = constant:

$$c = a\,B/A \cdot (1 - \exp(-Ax/V)) + c_o \cdot \exp(-Ax/V) \qquad (6.2)$$

and with numerical values inserted:

$$c = a \cdot 2.29 \cdot 10^{-3} + (c_o - a \cdot 2.29 \cdot 10^{-3}) \exp(-3.68 \cdot 10^{-3} x) \qquad (6.3)$$

This equation has been used to calculate the activity of Sr-89 and Sr-90 in the tap water from the lake Svartediket. In the case of Sr-89 an approximate correction for decay has been made by reducing the value of c_o by a factor of 0.5 for each step of integration of one month. In figures 6.7 and 6.8 the measured activities of Sr-89, Sr-90 and Cs-137 in tap water are given together with the calculated values.

The curves for Sr-89 agree well with respect to order of magnitude, and also the time variation is analogous. The theoretical values of Sr-90, however, are only 10 - 20 per cent of the measured values in the tap water. This shows that the main part of the Sr-90 activity in the tap water, 80 - 90 per cent, is brought into the lake Svartediket by leaching of the ground in the precipitation area. This takes considerable time, and for Sr-89 with a half life of 52 days, leaching from the precipitation area needs not be considered. The curve for Cs-137 in precipitation is very similar to the curve for Sr-90, but the Cs-137 activity in tap water is somewhat lower than the Sr-90 activity and more constant with time in the sampling period.

It may be estimated, as for Sr-90, that the main amounts of Cs-137 in the tap water, 60 - 70 per cent, have been leached out of the ground. In figures 6.9 and 6.10 the monthly average Cs-137/Sr-90 ratios in tap water sampled in Norway 1957 and 1958 are given. The figures show that more Cs-137 than Sr-90 is retained in the ground and the mean value of the Cs-137/Sr-90 ratio is below 1.0.

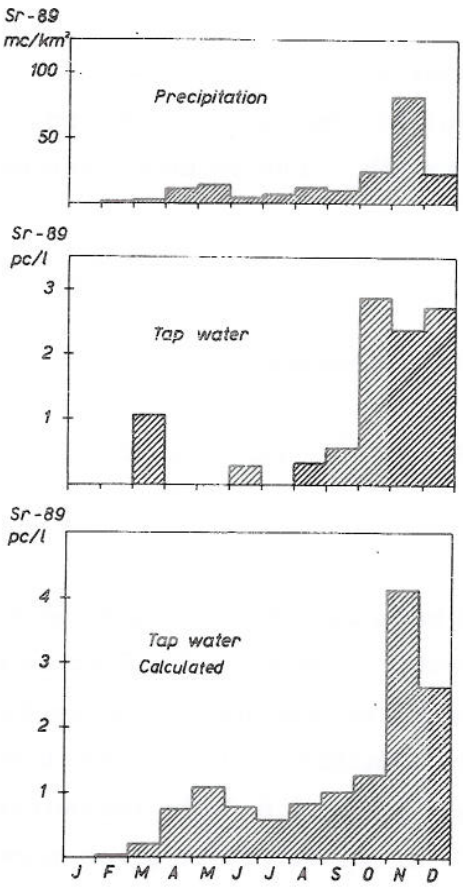

Figure 6.7 Sr-89 content in tap water, experimental and calculated data

6.3 Conclusions

The investigations of tap water in Norway show that the concentration of Sr-90 at present constitutes, according to the health authorities, little or no health hazard. Approximately one tenth of the fallout activity present in precipitation will be found in the tap water. In one area, at Bergen, it has been shown that the main part of the

Cs-137 and Sr-90 activity in the tap water has been leached out of the ground in the precipitation area, while the Sr-89 activity is due to fallout coming directly into the lakes. In Bergen, the natural possibilities of decontamination are unfavourable, the yearly precipitation is very high and there is little soil, no clay, and scanty vegetation in the precipitation area. One should therefore expect the tap water in Bergen to contain higher Sr-90 amounts than the average, this is found to be the case. Clay minerals have been shown to be very efficient decontamination agents, and in areas where clay minerals are a main constituent of the ground, relatively low activities of fallout in the tap water may be expected.

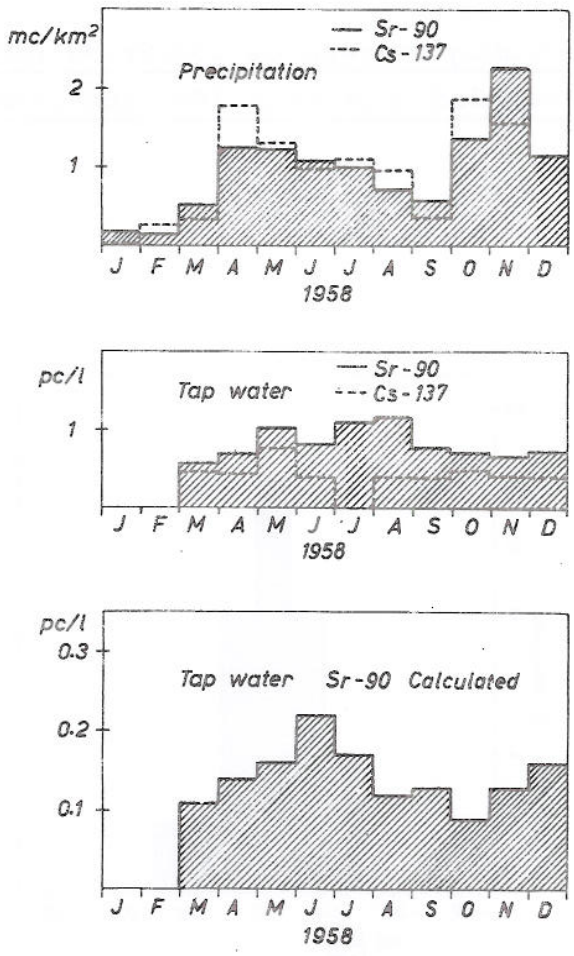

Figure 6.8 Sr-90 and Cs-137 content in tap water, experimental and calculated data

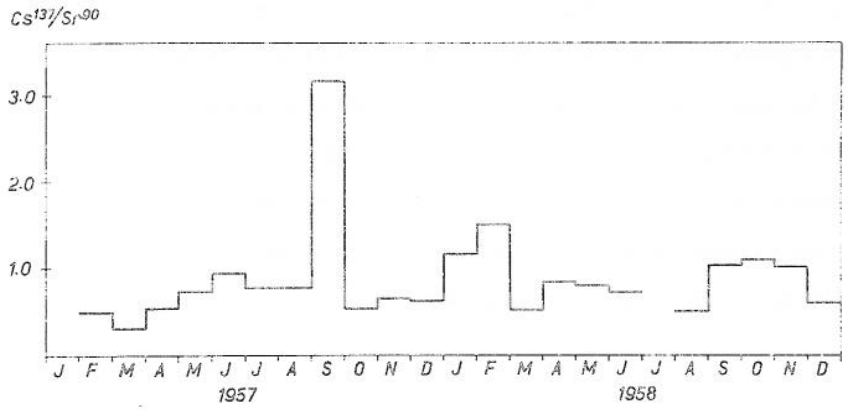

Figure 6.9 Monthly average Cs-137/Sr-90 ratios in tap water sampled in Norway 1957 and 1958

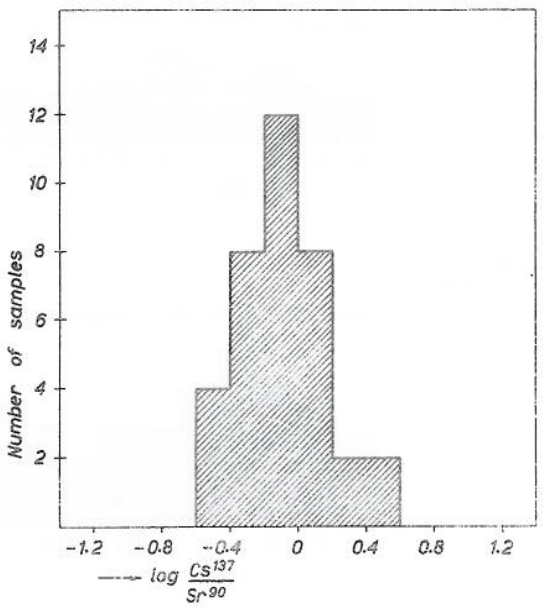

Figure 6.10 Monthly average Cs-137/Sr-90 ratios in tap water to number of samples, Norway 1957 and 1958

7 FALLOUT IN MILK

7.1 Introduction

Milk is the principal food in which Sr-90 and Cs-137 enter diet in Scandinavia, for infants it may be the sole nutrient. It is therefore important to follow closely the content of Sr-90 and Cs-137 in milk. The fallout isotopes are primarily taken up by the cow and then secreted into the milk. So far the main concern has been connected with Sr-90, which largely follows Ca in the metabolic chain, but it is well known that also large fractions of I-131 and Cs-137 are secreted into the milk. The isotope Cs-137 follows the metabolism of K, while I-131 joins the natural I. The amounts of these fallout isotopes taken up by the living organism have been found to depend within limits on the amounts of natural Ca, K and I present in the feed. If the feed is deficient in any of these essential elements, more of the radioactive isotope will be taken up.

There are minute amounts of natural Sr and Cs present in the feed, but as these elements are not known to play any essential role in the metabolic reactions, their presence in men an animals so far have more or less been neglected. For I it is well known that the element is essential and accumulates in the thyroid gland.

The milk samples studied in the following are from dairies and therefore represent an average for a given district. No detailed studies of the connection between the fallout content of feed and milk have been carried out. It is, however, generally recognized that the main source of Sr-90 and Cs-137 in the feed is the grass which in addition to the isotopes taken up from the soil, also will contain large amounts deposited directly on the leaves.

Throughout the year 1957 milk produced in Norway was analyzed regularly for Sr-90, Sr-89, Cs-137 and I-131. In 1958 this milk program had to be curtailed. Dry milk samples from Levanger were, however, stored through the year 1958 and analyzed for Sr-90 and Cs-137 in 1959. In January this year regular sampling and analysis of milk were started again. In (8) preliminary data from the present investigations have been published, in this report the results are given together with some comments.

7.2 Experimental data and discussion

The methods of sampling and analysis of milk have been described in chapters 2.3 and 3. In the following the results for each of the different isotopes will be discussed.

The samples used for analysis of I-131 have been collected from dairies in Røros, Oslo and Sandnes, see map figure 2.1. In tables 7.1 - 7.3 and figures 7.1 - 7.3 the results from these analysis are given. It could be expected that Røros situated in the eastern part of Norway in a mountain area known to be short of natural iodine, would deliver milk with a high concentration of I-131. The following average values were found : Røros 108 pc I-131/l, Sandnes 83 pc I-131/l and Oslo 57 pc I-131/l. The highest value, 1350 pc I-131/l, was found in milk sampled in March at Røros. These figures may be compared to data from USA where the yearly average values for a period ending January 1959 vary between 28 and 83 pc I-131/l in milk from different regions. The maximum value found in USA in this period was 624 pc I-131/l in milk sampled in July 1958 at St Louis (48).

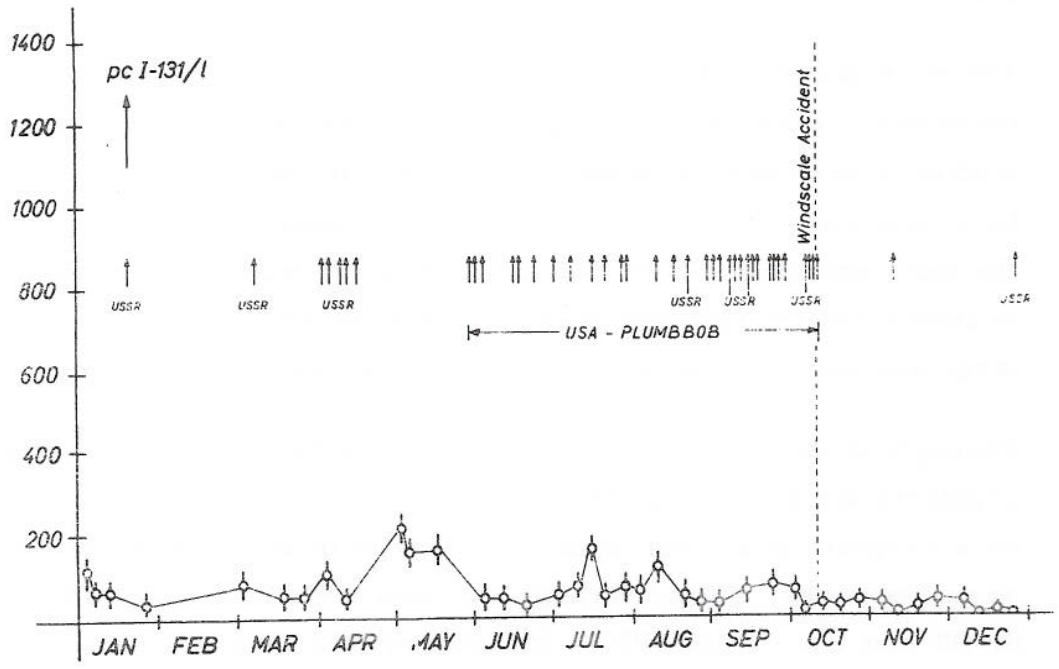

Figure 7.1 I-131 in milk from Oslo, Norway 1957

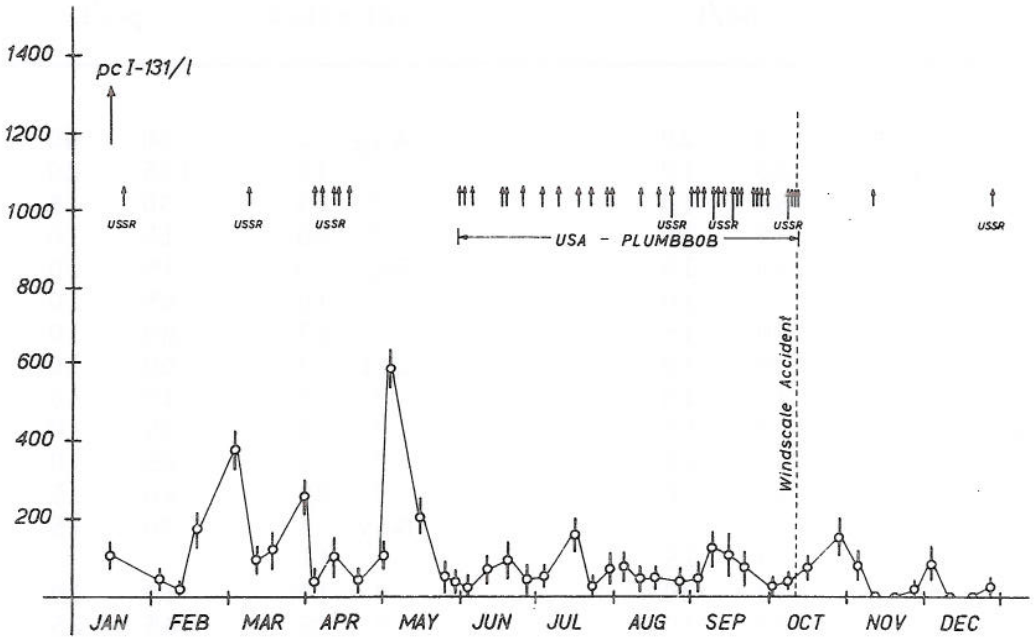

Figure 7.2 I-131 in milk from Sandnes, Norway 1957

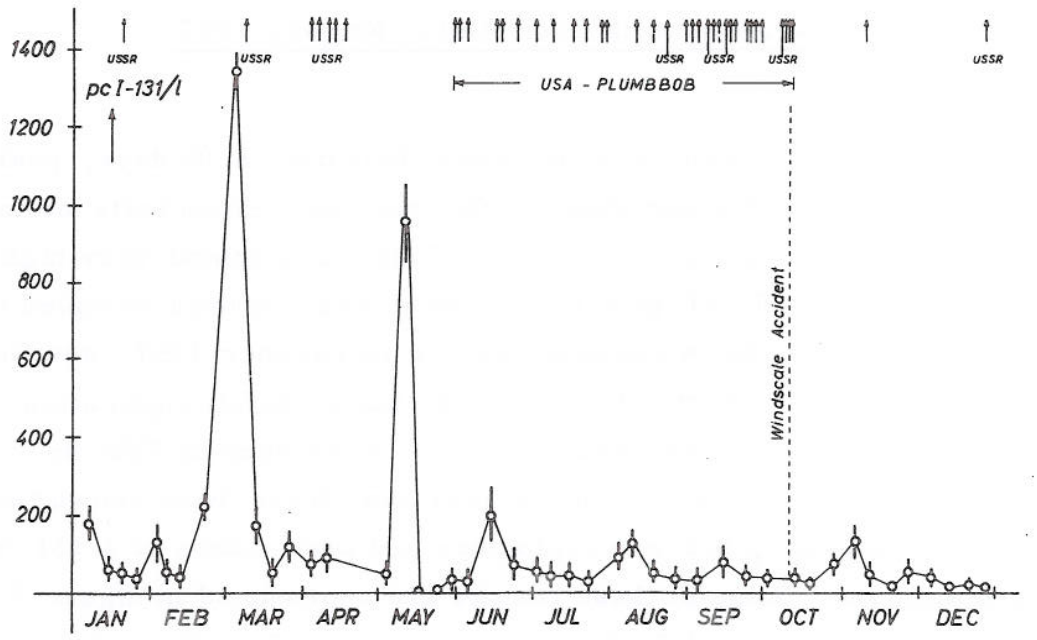

Figure 7.3 I-131 in milk from Røros, Norway 1957

Date collected	I-131 pc/l	S D	Date collected	I-131 pc/l	S D
Jan 2	110	20	Aug 2	60	40
" 4	60	10	" 12	115	20
" 9	60	10	" 19	30	15
" 28	30	10	" 26	15	10
Mar 6	90	10	Sep 9	15	10
" 20	70	10	" 16	65	10
" 27	70	15	" 23	80	10
Apr 3	100	10	Oct 3	60	10
" 9	30	10	" 8	10	10
May 2	210	50	" 14	35	10
" 3	160	100	" 22	25	10
" 18	160	20	" 28	40	15
" 27			Nov 4	30	15
Jun 3	30	15	" 11		
" 8	30	20	" 19	10	10
" 18	20	10	" 25	45	25
Jul 1	50	20	Dec 2	35	15
" 9	85	15	" 10		
" 15	160	20	" 16	5	5
" 22	35	15	" 23		
" 29	65	25			
			Average	56	

Table 7.1 I-131 in milk from Oslo, Norway 1957

Since I-131 is an isotope with a short half life, 8.05 days, peak values are only obtained shortly after nuclear weapon tests or in connection with reactor accidents. Thus, in England very high values, up to 800 000 pc I-131/l were found in milk sampled in the surroundings of the Windscale reactor in October 1957, see Dunster (60). Due to its short half life, I-131 has a usefull application in the control of nuclear weapon tests. Work done in USA by Van Middlesworth (61), Schrodt (62) and others have shown the close relationship bet veen explosions and peak values of I-131 in men and animals. Some of these results are reproduced in figure 7.4. In the figures 7.1 - 7.3 the dates of known nuclear detonations during 1957 are indicated. Some of the USSR test series seem to be well recognized at Rφros and Sandnes approximately 30 days after detonation.

Date collected	I-131 pc/l	S D	Date collected	I-131 pc/l	S D
Jan 16	100	25	Jul 23	10	10
Feb 4	40	20	" 30	30	15
" 12	10	10	Aug 6	65	15
" 25	180	40	" 13	30	15
Mar 6	380	40	" 16	20	20
" 14	90	20	" 26	15	15
" 18	110	50	Sep 2	45	40
" 30	250	40	" 9	110	20
Apr 2	20	10	" 16	105	60
" 12	100	45	" 23	70	40
" 20	20	10	Oct 1	10	10
May 1	100	20	" 7	35	15
" 8	590	50	" 14	80	60
" 16	200	40	" 28	150	50
" 25	45	45	Nov 4	80	60
" 28	30	20	" 11		
Jun 6	10	10	" 18		
" 13	60	40	" 25	10	10
" 20	90	50	Dec 2		
" 27	20	20	" 9	10	10
Jul 3	25	10	" 16	10	10
" 16	175	30	" 23	70	20
			Average	83	

Table 7.2 I-131 in milk from Sandnes, Norway 1957

In tables 7.4 - 7.11 and figures 7.5 - 7.8 results from the analysis of Sr-90, Sr-89 and Cs-137 in milk from Lillestrøm, Sandnes, Tynset, Levanger, Bergen, Røros and Stjørdal are given. In figures 7.9 and 7.10 the variation of Sr-90 and Cs-137 with time in Norway is compared with data from other countries.

The Sr-89 data all show higher values in the summer than in the winter. This may be explained by the fact that the cows in the summer are grazing in the fields, while during the winter they are kept indoors feeding on crops collected in the autumn or on older mixed fodder. Sr-89 has a relatively short half life and therefore disintegrates during the winter.

Date collected	I-131 pc/l	S D	Date collected	I-131 pc/l	S D
Jan 9	90	15	Jul 1	50	20
" 16	40	10	" 8	45	15
" 21	30	15	" 15	40	30
" 29	20	15	" 22	10	10
Feb 4	120	50	Aug 5	95	20
" 12	20	10	" 12	125	25
" 19	20	10	" 19	25	15
" 27	220	50	" 26	30	15
Mar 6	1350	50	Sep 2	20	20
" 12	180	15	" 16	90	30
" 18	50	10	" 23	30	10
" 25	110	15	Oct 1	35	15
Apr 2	70	15	" 14	35	15
" 8	90	15	" 21	15	10
May 2	40	20	" 29	75	25
" 7	960	140	Nov 5	140	50
" 14			" 11	45	30
" 22			" 18		
" 31	20	15	" 25	55	40
Jun 5	10	10	Dec 3	35	10
" 15	200	90	" 10		
" 25	70	30	" 16	10	10
			" 30		
			Average	105	

Table 7.3 I-131 in milk from Røros, Norway 1957

Also in the case of Sr-90, see figure 7.9, higher activities in the milk are indicated during the summer. The variations are, however, much smaller. As to Sr-90, decay is of no importance and the variations must be due to differences in composition of the feed in winter and summer. If the contamination of the grass increases from year to year, one would expect an increased activity in the spring, when the cows start feeding on grass with a higher Sr-90 content. The drop of activity in the autumn may be due to a change in composition of the feed, less contaminated feed beeing added to the fodder.

The curves for Cs-137, see figure 7.10, show great similarity to the Sr-90 curves, only the absolute values are considerably higher.

This is due to a difference in the metabolic system of the cow, the discrimination factor for Cs-137 is considerably lower than for Sr-90.

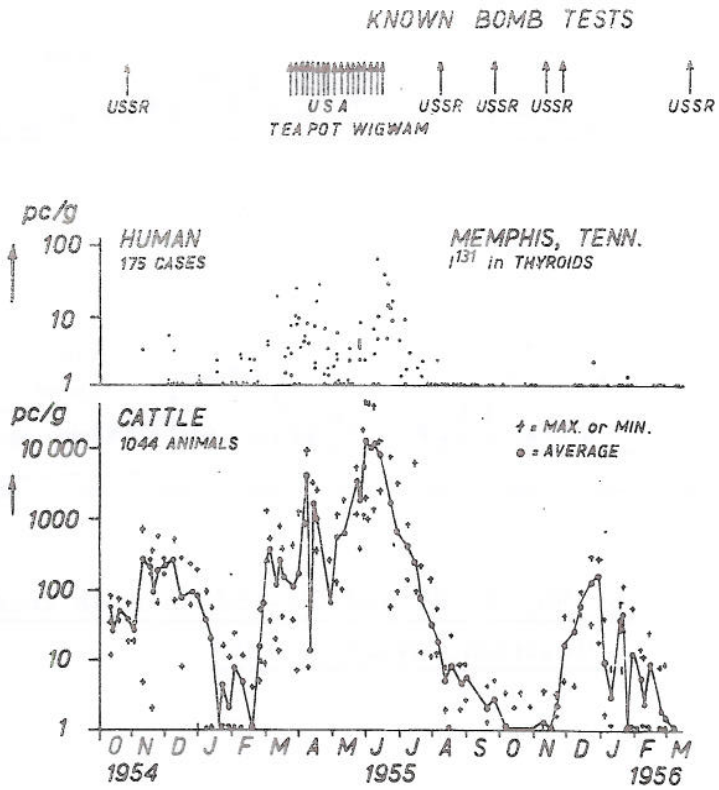

Figure 7.4 Relationship between explosions and peak values of I-131 in men and animals, Van Middlesworth (61)

As mentioned neither Sr nor Cs are known to have any important biological function in men or animals. In plants Sr has been claimed to be a necessary micronutriment, see Wolf and Cesare (63). In larger amounts Sr is known to be toxic to animals if substituted to a considerable degree for Ca, Comar (64). So far it has generally been assumed that natural Sr and Cs are taken up as impurities together with the essential elements Ca and K. It is an open question whether variations in minute amounts of natural Sr or Cs present in the diet do influence the uptake of the corresponding radioactive isotope.

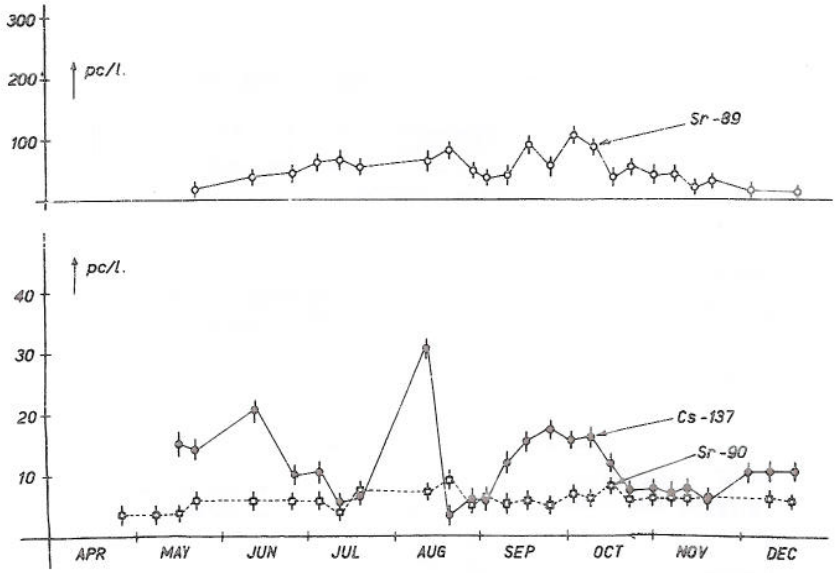

Figure 7.5 Sr-90, Sr-89 and Cs-137 in milk from Lillestrøm, Norway 1957

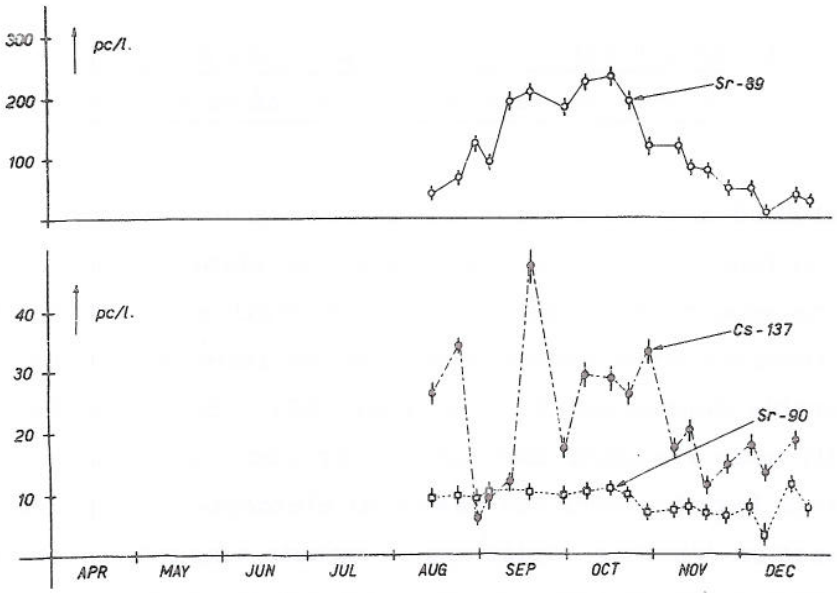

Figure 7.6 Sr-90, Sr-89 and Cs-137 in milk from Sandnes, Norway 1957

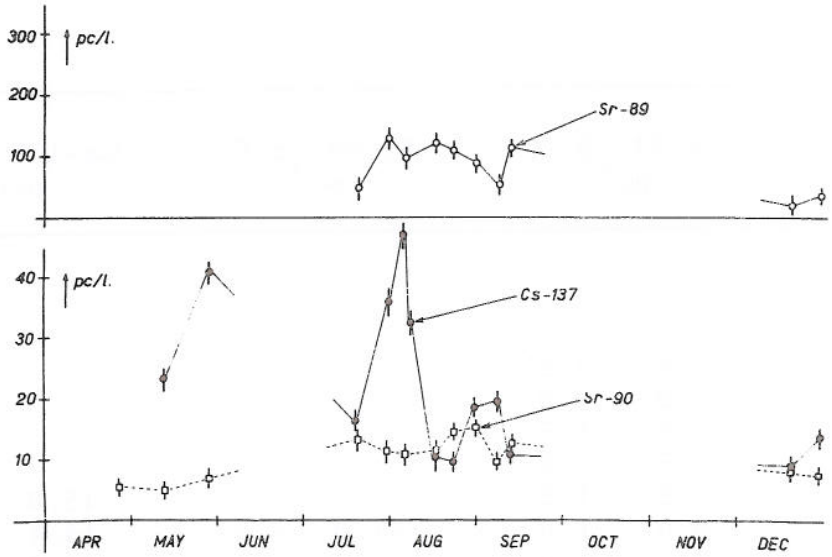

Figure 7.7 Sr-90, Sr-89 and Cs-137 in milk from Tynset, Norway 1957

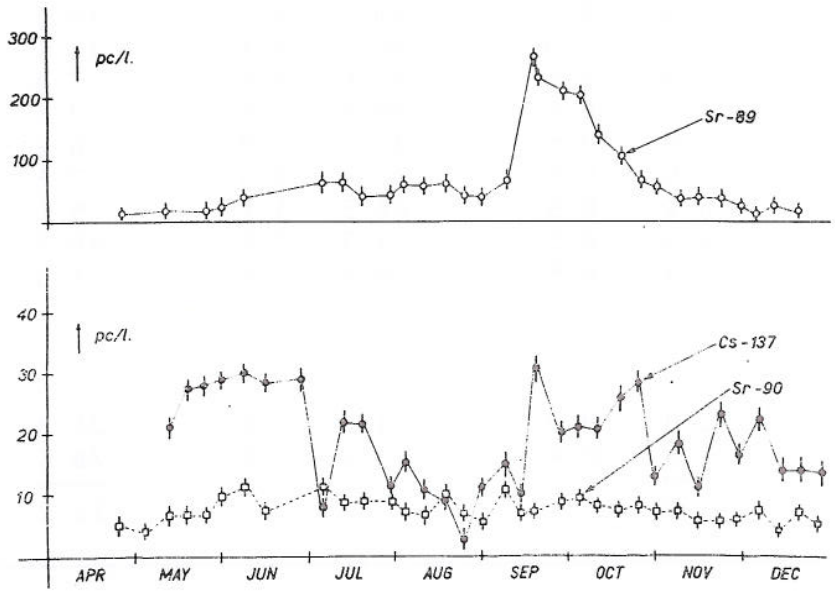

Figure 7.8 Sr-90, Sr-89 and Cs-137 in milk from Levanger, Norway 1957

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Feb 15	6.0	1.5				
Mar 20	8.0	2.0				
Apr 25	4.5	1.5				
May 7	4.5	1.0				
" 15	4.5	1.5			15.0	1.2
" 21	6.0	1.0	11.0	2.2	14.4	1.2
Jun 11	6.0	1.0	33.2	3.0	21.2	1.5
" 25	6.0	1.0	48.5	4.1	10.0	0.6
Jul 3	6.0	1.0	60.5	3.2	10.9	0.6
" 10	4.5	1.0	61.8	4.2	5.6	0.9
" 17	7.5	1.0	53.0	4.0	6.2	0.9
Aug 12	7.0	0.7	67.5	4.5	30.6	1.5
" 19	9.0	0.8	84.0	4.4	3.3	0.5
" 26	5.0	0.6	49.5	3.5	5.9	0.4
Sep 2	6.5	0.4	36.0	1.9	6.2	0.5
" 9	5.0	0.5	45.5	2.8	11.5	0.8
" 16	6.0	0.5	92.0	6.0	15.3	0.9
" 24	5.0	1.2	55.0	4.8	17.1	0.9
Oct 2	7.0	0.5	103.0	5.2	15.6	0.8
" 7	6.5	0.5	90.0	4.9	16.5	1.0
" 15	8.0	2.0	40.5	3.4	12.1	0.6
" 22	5.5	0.5	51.0	3.8	7.4	0.7
" 30	6.0	0.5	38.8	2.0	7.6	0.6
Nov 6	6.0	0.5	40.0	1.9	6.2	0.4
" 11	6.0	0.5	22.4	2.9	7.6	0.9
" 19	6.0	0.5	28.5	3.1	5.6	0.5
Dec 10	6.2	0.5	17.7	2.8	10.3	0.7
" 17	5.3	0.5	16.8	2.5	11.3	0.9
1958						
Jan 11	2.0	0.3	24.3	1.8	21.3	1.0
" 25	4.6	0.4	13.9	1.8	28.5	1.7
Average	5.9				12.2	

Table 7.4 Sr-90, Sr-89 and Cs-137 in milk from Lillestrøm, Norway 1957 and 1958

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Aug 14	9.0	0.6	49.0	4.3	26.2	1.2
" 21	9.5	0.7	65.0	4.0	34.1	0.4
" 27	9.0	0.7	124.0	6.5	5.3	0.3
Sep 2	10.0	0.5	97.5	5.9	8.8	1.0
" 9	11.0	0.6	191.0	10.5	11.2	0.7
" 16					62.9	3.2
" 26	10.0	0.6	203.0	10.5	47.3	2.2
" 30	9.5	0.6	181.0	10.0	17.1	1.0
Oct 7	10.0	0.7	219.0	11.0	29.4	1.5
" 14	10.5	0.5	232.0	12.2	28.8	1.5
" 21	9.5	0.7	191.0	10.0		
" 28			116.5	6.5	25.9	1.2
Nov 4	7.0	0.6	114.0	6.5	32.9	1.5
" 11	7.5	0.6	81.5	5.5	17.1	0.9
" 18	6.5	0.6	74.5	5.0	20.6	0.9
" 25	6.0	0.7	43.5	5.0	10.9	0.9
Dec 2	7.5	0.5	46.5	4.4	14.4	0.9
" 9	3.0	1.6			17.3	1.1
" 16	11.5	0.8	36.4	5.2	13.1	0.8
" 23	8.0	0.7	25.0	4.0	18.2	1.1
1958						
Jan 2	8.7	0.9	67.5	5.7	32.7	1.8
" 6	7.1	0.5	25.0	2.6	15.6	0.9
" 13	8.8	0.6	18.9	2.9	17.3	1.1
" 18	6.3	0.3	15.6	1.6	28.3	1.6
" 20	8.6	0.6	20.0	2.7	18.0	1.3
Average	8.4				23.0	

Table 7.5 Sr-90, Sr-89 and Cs-137 in milk from Sandnes, Norway 1957 and 1958

Tables 7.12 - 7.15 give the content of natural Sr and Sr-90 found in milk in the present investigation. In table 7.16 average values from four different localities in Norway are compared with the value given by Harley et al 1957 (29) for New York. In figure 7.11 corresponding values of Sr-90 and natural Sr in milk are plotted, in the figure the number of samples within intervals are given.

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Mar 15	4.5	1.5				
Apr 26	5.5	1.0				
May 13	5.0	1.0			23.2	1.5
" 28	7.0	1.0			41.2	2.9
Jul 19	15.4	0.7	48.0	5.2	16.5	0.9
" 29	13.9	0.6	126.0	7.8	36.8	2.1
Aug 3					48.8	2.4
" 5	11.0	0.6	93.5	5.7	32.6	1.8
" 16	12.0	0.7	120.5	7.0	10.9	0.5
" 22	14.5	1.1	101.0	6.3	9.7	0.6
" 29	15.0	0.6	92.5	6.8	18.4	1.2
Sep 6	9.1	0.6	55.5	4.4	19.6	1.1
" 10	12.5	0.7	111.0	6.5	10.9	0.8
Dec 19	8.0	0.5	21.0	2.5	8.9	0.8
" 28	7.8	0.7	32.3	3.6	14.2	0.9
1958						
Jan 9	6.0	0.4	18.4	2.4	80.5	3.8
" 15	6.4	0.3	18.1	1.7	43.8	2.8
Average	9.6				27.7	

Table 7.6 Sr-90, Sr-89 and Cs-137 in milk from Tynset, Norway 1957 and 1958

The line represents average Sr-90 activity against natural Sr content. The local average values of table 7.16 are also marked in the same figure. This graph indicates that there is a probability of finding high values of Sr-90 when the natural Sr content is low. The statistical evaluation shows that the data may be represented by:

$$(\text{Sr-90}) \cdot (\text{Sr})^{n} = \text{constant}$$

with a 95 per cent probability of finding $-0.26 < n < -0.02$

Analogous observations have been made by Catsch in 1957 (65) in an investigation of the deposition of Sr-90 in rat bones as a function of injected alkaline earth carriers. Palmer, Thompson and Kornberg (66)

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1957						
Apr 25	5.0	1.0	2.3	1.0		
May 3	4.5	1.0				
" 11	7.0	1.0	5.0	1.0	21.2	1.5
" 18	7.0	1.0			27.9	1.8
" 25	7.0	1.0	5.6	2.1	28.8	2.1
Jun 1	10.0	1.0	11.2	2.6	29.7	1.8
" 8	11.5	1.0	36.0	2.1	30.3	1.8
" 15	7.5	1.0			28.8	1.8
" 29					29.4	1.5
Jul 6	11.5	1.0	61.0	5.7	7.9	0.6
" 12	8.0	1.0	61.5	3.7	22.6	1.2
" 19	9.0	1.0	43.0	3.9	22.3	1.2
" 29	9.5	1.0	46.0	4.0	11.8	0.6
Aug 3	7.5	0.5	56.5	4.0	15.6	0.9
" 10	7.0	0.6	52.5	4.1	11.8	0.6
" 16	10.0	0.7	57.5	2.8	9.7	0.6
" 24	7.0	0.6	42.0	3.6	2.4	0.2
" 31	6.0	0.4	39.6	3.1	11.5	1.6
Sep 7	11.5	0.7	69.0	6.5	15.0	0.9
" 14	7.0	0.5	270.0	12.5	10.9	0.6
" 18	7.0	0.5	246.0	12.0	31.8	1.9
" 28	8.5	0.5	224.0	11.0	20.6	1.1
Oct 3	9.5	0.6	210.0	11.0	21.8	0.9
" 10	8.5	0.6	148.0	8.5	21.5	0.8
" 17	7.0	0.7	109.0	5.3	26.5	1.5
" 24	8.0	0.5	68.0	4.4	28.2	1.5
" 31	7.5	0.5	51.5	3.9	12.9	0.9
Nov 7	7.5	0.6	33.7	3.7	18.8	0.9
" 14	5.5	0.5	36.3	3.3	11.2	0.6
" 21	6.2	0.4	35.5	4.0	23.2	1.2
" 28	6.7	0.5	25.0	3.6	16.2	0.9
Dec 5	7.5	1.8	12.6	6.3	22.5	1.2
" 12	4.0	1.6	28.3	7.0	14.0	0.8
" 19	7.6	0.6	23.0	3.5	14.0	0.8
" 26	5.5	0.5			13.8	0.8
Average	7.7				19.2	

Table 7.7 Sr-90, Sr-89 and Cs-137 in milk from Levanger, Norway 1957

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1958						
Jan 11	5.0	0.7	10.9	2.8	46.2	2.3
" 24	7.8	0.4			27.8	2.1
" 31	8.7	0.5			14.4	0.9
Feb 7	8.1	0.4			5.8	0.4
" 15	10.2	0.5			12.1	0.8
" 21	7.0	0.3			8.1	0.6
" 25	5.8	0.4			20.7	0.8
Mar 7	8.4	0.4			30.5	1.2
" 11	9.5	0.5			27.0	1.1
" 21	6.1	0.4			11.9	0.6
" 28	9.2	0.5			32.7	1.3
Apr 11	7.1	0.4			62.3	2.5
" 29	11.9	0.8			24.2	1.2
May 2	12.5	1.5			27.8	1.9
" 10	7.9	0.5			21.9	1.4
" 17	8.2	0.6			11.9	0.8
" 30	15.9	0.8			30.4	1.5
Jun 6	12.2	0.7			32.4	1.6
" 13	16.1	1.0			32.4	1.6
" 20	18.1	0.3			58.5	4.3
" 27	22.2	1.1			36.7	1.7
Jul 11	21.8	1.1			39.6	1.3
" 18	17.2	0.9			41.8	1.2
Aug 1	20.2	1.9			74.0	3.5
" 8	23.4	1.2			75.2	2.6
" 15	19.8	1.0			71.0	3.4
" 22	18.7	1.2			83.8	3.9
Sep 12					38.9	1.9
" 19	15.2	0.8	29.1	6.5	23.5	1.2
" 26	16.7	1.1	64.9	8.8	66.3	3.1
Oct 10	13.5	0.6	55.5	4.2	20.3	1.0
" 17	11.3	0.6	46.1	3.4	50.8	1.8
" 24	11.3	0.6	32.9	4.6	50.9	1.8
" 31	11.6	0.7	7.6	2.9	24.2	1.3
Nov 7	14.0	0.6			50.4	2.3
" 14	13.6	0.8			50.5	2.4
" 28	13.8	0.8			61.9	2.9
1959						
Jan 2	16.4	1.0			30.1	1.4
" 9	18.2	2.1			34.6	1.6
Feb 6	10.2	0.8	8.3	2.7	34.3	2.6
Average	13.0				37.4	

Table 7.8 Sr-90, Sr-89 and Cs-137 in milk from Levanger, Norway 1958 and 1959

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1959						
Jan 14	28.2	1.1			141.2	6.1
" 21	18.6	1.0			132.7	4.5
" 28	18.9	1.8			218.4	8.1
Feb 4	43.7	2.4			216.6	7.5
" 12	18.1	1.3			61.4	3.1
" 20	22.3	1.4			100.5	3.4
" 27	24.7	1.7			183.7	7.9
Mar 4	18.7	1.3			159.0	6.9
" 11	27.5	1.7			113.2	5.0
Average	24.5				147.4	

Table 7.9 Sr-90, Sr-89 and Cs-137 in milk from Bergen, Norway 1959

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1959						
Jan 15	12.9	0.6			72.7	3.5
" 22	11.2	0.6			97.2	3.4
" 30	10.4	1.1			87.4	3.5
Feb 5	12.7	1.1			99.1	3.9
" 11					100.6	5.0
" 19	14.6	1.2			93.4	3.5
" 26	9.6	1.0			96.0	3.9
Mar 6	10.6	1.1			79.0	3.7
" 12	12.3	1.0			80.2	3.9
" 18	13.5	1.1			57.0	2.7
Average	12.0				86.3	

Table 7.10 Sr-90, Sr-89 and Cs-137 in milk from Røros, Norway 1959

Date collected	Sr-90 pc/l	S D	Sr-89 pc/l	S D	Cs-137 pc/l	S D
1958						
Nov 1	9.8	0.8	15.1	3.2	39.1	2.1
" 3	10.8	0.9	12.7	3.3	40.1	2.1
" 5	9.8	0.9	15.7	3.4	45.1	2.4
" 7	11.3	1.2	11.5	3.7	48.0	2.6
" 8	8.8	0.8	18.3	3.1	47.8	2.8
Average	10.1				44.0	

Table 7.11 Sr-90, Sr-89 and Cs-137 in milk from Stjørdal, Norway 1958

conclude from experiments on rats that the uptake of Sr-90 may be more closely related to natural Sr concentrations than to the Sr-90/Ca ratio. According to Comar, Scott Russell and Wasserman (67) the Sr/Ca ratio is lower in the milk than in the feed by a factor k of 0.10 - 0.14, i e:

$$(Sr/Ca)_{milk} = k \cdot (Sr/Ca)_{feed}$$

In short term feeding the value of k is higher, thus a 4-fold increase of the amount of Ca in the feed over a short period gave only a 35 per cent reduction of the Sr-90 content in the milk. It has been assumed that addition of natural Sr will give much the same effect as Ca, but this has not been verified. The present results indicate that the factor k is increased if the content of natural Sr in milk becomes less than 1 - 2 mg Sr/g Ca. Using the value of k given by Comar et al (67) for the transfer of Sr-90 from contaminated feed to milk, this means that from a preventional point of wiew, it might be worth while to insure that the Sr-level of the feed is above 10 - 20 mg Sr/g Ca. It may be mentioned that the results obtained hardly can be correlated with changing amounts of Ca in the feed.

7.3 Conclusions

In figures 7.9 and 7.10 the variation with time of the content of Sr-90

and Cs-137 in milk from Norway is compared with data from USA, UK, Sweden and Germany. These figures show that the Sr-90 and Cs-137 content of the milk is increasing from year to year and distinct seasonal variations are observed. It is further noticed that the values found in Norway are high compared with other countries. Since milk is known to be the principal source of Sr-90 and Cs-137 in the diet, the estimation of the future contamination is of great interest. Some general considerations may be made in this connection.

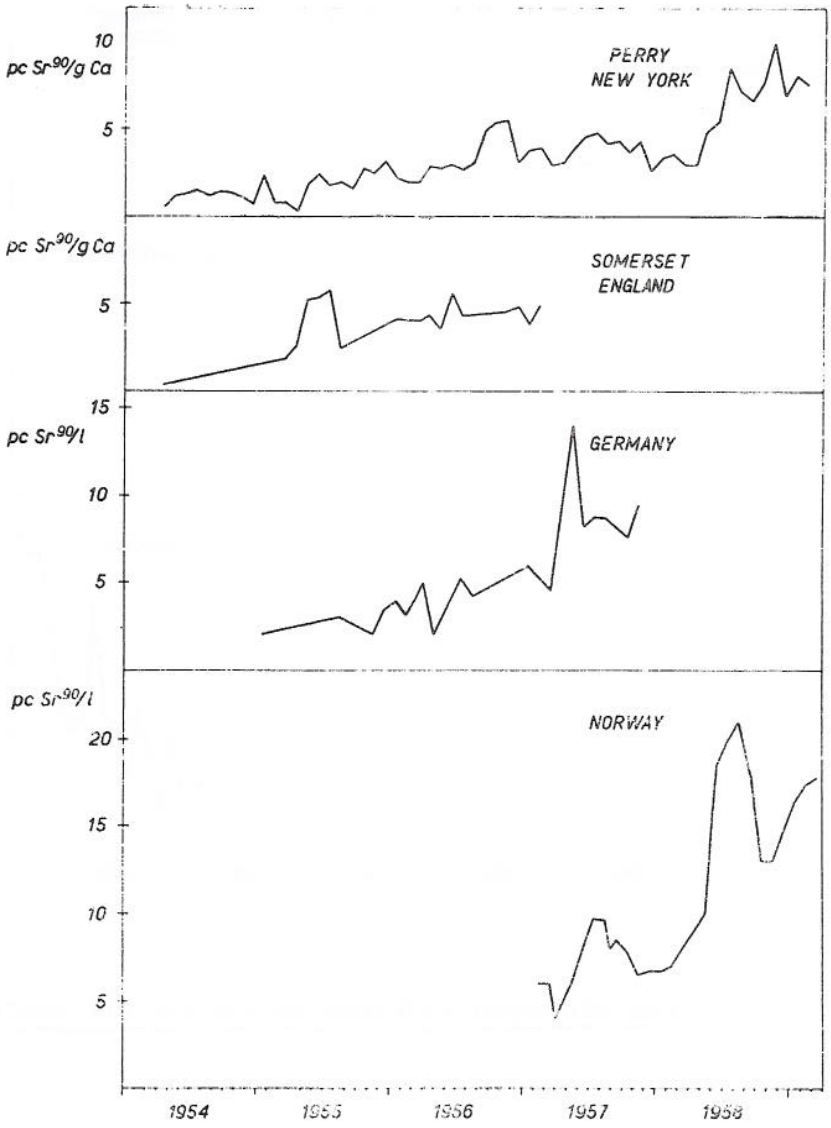

Figure 7.9 The variation with time of the Sr-90 content in milk

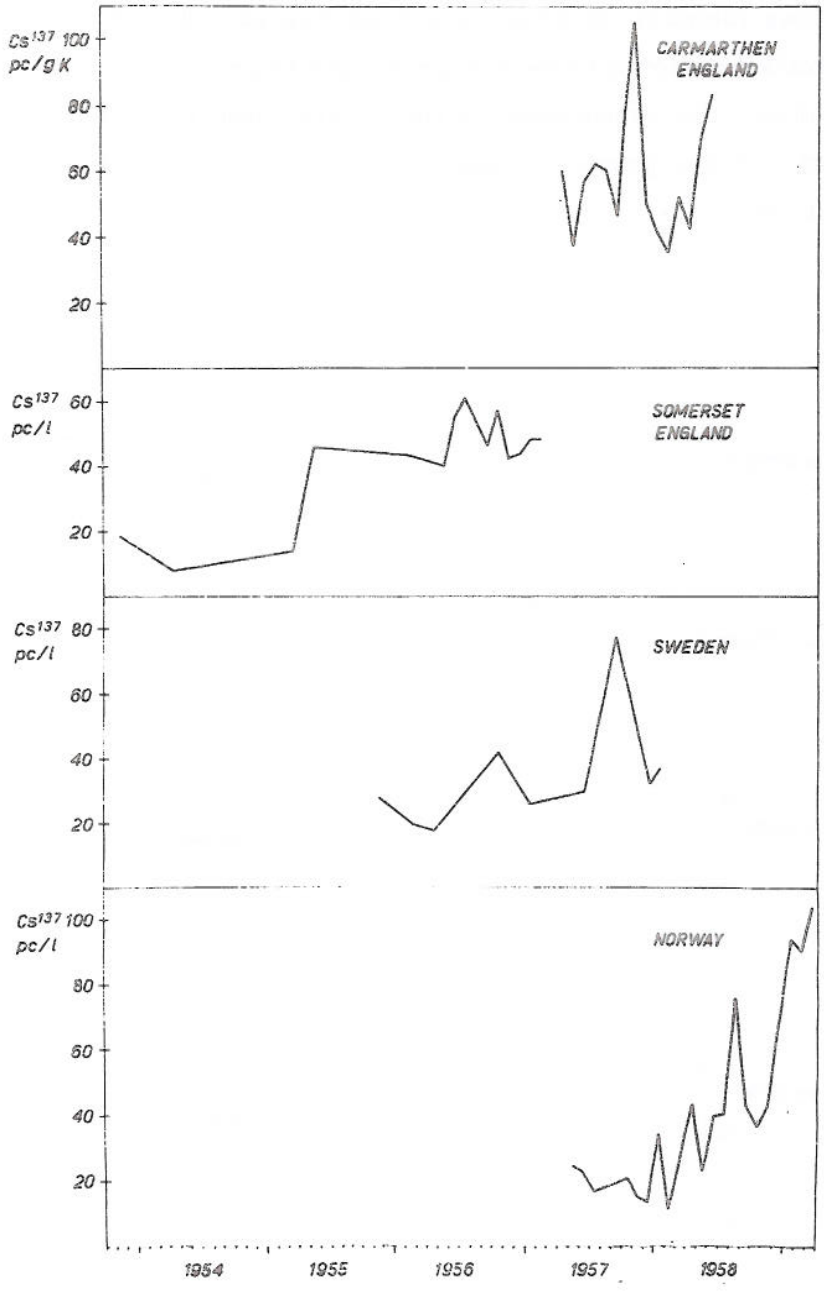

Figure 7.10 The variation with time of the Cs-137 content in milk

Date collected	Sr mg/l	Sr-90 pc/l	Date collected	Sr mg/l	Sr-90 pc/l
1957					
Aug 21	0.3	9.5	Nov 18	4.4	6.3
" 27	0.6	9.2	" 25	0.5	6.0
Sep 2	0.4	9.8	Dec 2	1.1	7.5
" 9	0.3	10.9	" 9	0.3	3.0
" 26	0.7	9.8	" 16	0.4	11.5
" 30	0.3	9.4	" 23	0.5	8.0
Oct 7	0.05	10.0			
" 14	0.2	10.7	1958		
" 21	0.2	9.4			
" 28	0.5	6.5	Jan 2	0.4	8.7
Nov 4	0.3	7.2	" 6	0.4	7.1
" 14	0.4	7.5	" 13	0.3	8.8
			" 20	0.4	8.6
			Average	0.57	8.9

Table 7.12 Natural Sr and Sr-90 in milk from Sandnes, Norway 1957 and 1958

Date collected	Sr mg/l	Sr-90 pc/l	Date collected	Sr mg/l	Sr-90 pc/l
1957					
Jun 25	1.4	6.0	Oct 22	0.4	5.5
Aug 19	0.4	9.0	" 30	0.5	6.0
" 26	0.4	5.2	Nov 6	0.3	6.0
Sep 2	0.5	6.4	" 19	0.6	5.9
" 9	3.0	4.8	Dec 10	0.7	6.2
" 16	0.7	6.2	" 17	0.5	5.3
" 24	3.4	5.2			
Oct 2	0.4	7.0	1958		
" 7	0.4	6.5			
" 15	1.0	8.0	Jan 11	0.5	2.0
			" 25	0.4	4.6
			Average	0.89	6.3

Table 7.13 Natural Sr and Sr-90 in milk from Lillestrøm, Norway 1957 and 1958

Date collected	Sr mg/l	Sr-90 pc/l	Date collected	Sr mg/l	Sr-90 pc/l
1957					
Jul 19	0.6	15.4	Sep 10	0.4	12.5
" 29	0.2	13.9	Dec 19	0.4	8.0
Aug 3	0.1		" 28	0.3	7.8
" 16	0.3	12.0			
" 22	0.3	14.5	1958		
" 27	0.2	15.0			
Sep 7	0.7	9.0	Jan 15	0.4	6.4
			Average	0.36	11.5

Table 7.14 Natural Sr and Sr-90 in dry skimmed milk from Tynset, Norway 1957 and 1958

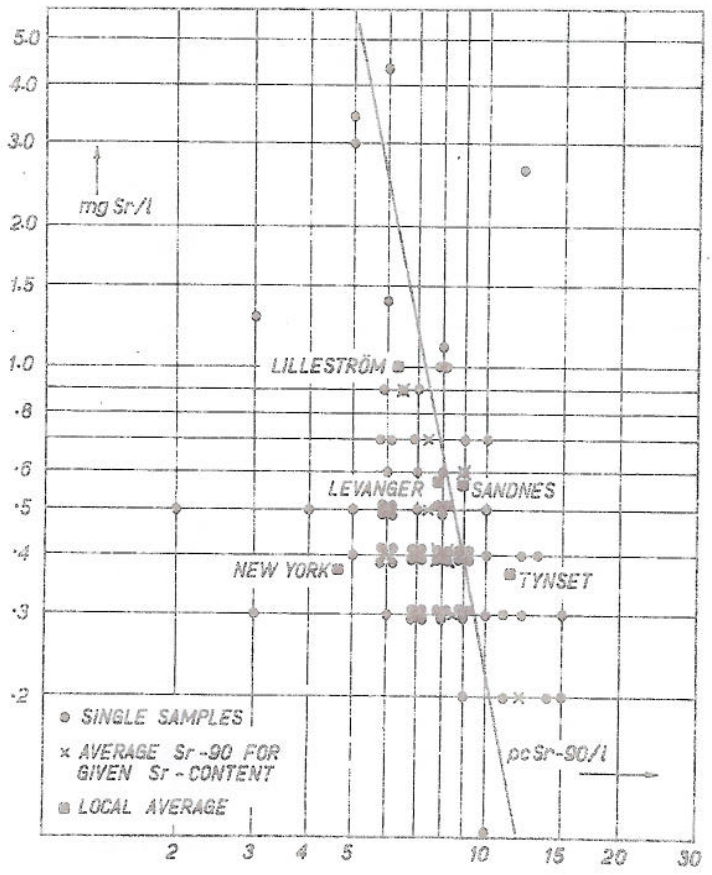

Figure 7.11 Relationship between Sr-90 and natural Sr in milk from Norway 1957

Date collected	Sample	Sr mg/l	Sr-90 pc/l
1957			
Jun 15	Whole milk	0.4	7.5
Jul 19	" "	0.4	9.2
" 29	" "	0.5	9.5
Aug 3	" "	0.3	7.5
" 24	" "	0.3	6.8
" 31	" "	0.4	5.8
Sep 7	" "	2.7	11.5
" 18	Skimmed milk	0.6	7.2
" 26	" "	0.4	8.5
Oct 3	" "	0.3	9.3
" 10	" "	0.4	8.4
" 17	" "	0.3	7.0
" 24	" "	0.3	8.1
" 31	" "	1.0	7.5
Nov 7	" "	0.5	7.5
" 14	" "	0.9	5.7
" 21	" "	0.4	6.2
" 28	" "	0.7	6.7
Dec 5	" "	0.6	7.5
" 12	" "	0.5	4.0
" 19	" "	0.5	7.6
" 26	" "	0.4	5.5
1958			
Jan 18	" "	0.5	6.3
	Average	0.58	7.9

Table 7.15 Natural Sr and Sr-90 in milk from Levanger, Norway 1957 and 1958

Origin	No of samples	Sr mg/l	Sr-90 pc/l
Tynset	11	0.36	11.5
Levanger	23	0.58	8.9
Sandnes	22	0.57	7.9
Lillestrøm	18	0.89	6.3
New York City	14	0.37	4.6

Table 7.16 Average values for natural Sr and Sr-90 in milk sampled in Norway 1957 compared to data from New York (29)

The Sr-90 and Cs-137 content of the milk is within limits proportional to the content of these isotopes in the feed. The Sr-90 and Cs-137 in the feed is partly due to uptake of the isotopes in the plants from the soil, and partly due to activity deposited directly on the leaves. The present investigations of peat soils, see chapter 5.2.4 and other recent results (28, 40) indicate that the last effect may be very important. The soil - plant uptake largely will be given by the integrated amounts of Sr-90 and Cs-137 in the soil, while the transfer air - plant will be dependent upon the rate of deposition of the isotopes from the atmosphere.

If the cumulated amounts in the ground as function of time are called A, then the rate of deposition becomes $dA/dt = A'$. The concentration c of an isotope in the milk may then be given by an expression of the general form:

$$c = k_1 \cdot A + k_2 \cdot A'$$

The values of A and A' will depend on the number, place and types of bomb tests. The values of k_1 and k_2 will depend on the composition of the feed, and will change with season and location.

For given conditions $k_1 \cdot A$ and $k_2 \cdot A$ may be expected to change with time as shown in figure 7.12.

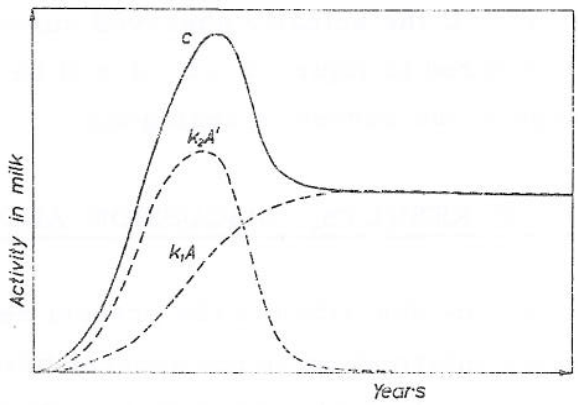

Figure 7.12 Predicted uptake of a fallout isotope in milk

The level at which the curves flatten out will depend on the test program, and this level will not be reached until a few years after the last bomb test, or when decay and test rate some time in the future may be brought to an equilibrium.

If k_2 is chosen to represent conditions under which the contribution proportional to rate of fallout is maximum for milk, in the summer, and k_1 is chosen to represent conditions at which there is minimum contribution from direct deposition, late winter, then the actually observed concentrations of Sr-90 in milk should show yearly oscillations as indicated in figure 7.13.

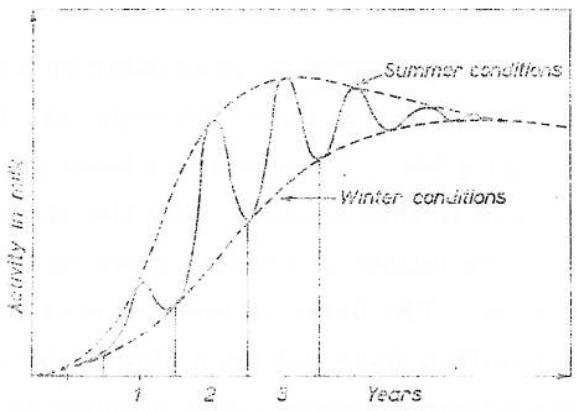

Figure 7.13 Predicted yearly minimum and maximum values of a fallout isotope in milk

The c-curves representing summer - respectively winter - conditions should form envelopes of the observed curve. It is not possible to give the exact shape of these curves, but it is felt that the general idea may be helpful in future estimation of fallout contamination. If the actually observed curves in figures 7.9 and 7.10 are compared to figure 7.13, it will be noticed that the general shape of the curves is analogous.

8 SUMMARY OF RESULTS, DISCUSSION AND CONCLUSIONS

The investigations described in the present report have been performed to obtain some information on the general fallout situation in Norway. Guided by the results published from investigations in other countries and the extensive work on total activity distribution by Division of Physics, NDRE, some of the more important isotopes, Sr-90, Sr-89, Cs-137 and I-131 have been studied in samples where they may be expected to occur in significant quantities. In table 8.1 are given average and maximum values for Sr-90 and Cs-137 in precipitation, tap water and milk, average monthly data are given in figure 8.1. The results of these investigations show that Norway receives more world-wide fallout than most other countries. This is due to the geographical situation of the country between 60° and 70° northern latitude. The high precipitation has also been considered. The present knowledge about atmospheric circulation and fallout deposition reveals that bomb tests at high northern latitudes will throw an increased share of fallout over Norway. For further studies of the fallout distribution representative data collected at higher northern latitudes will be of great value.

The fallout is brought down by precipitation and by turbulent deposition. The contribution from the latter process has frequently been considered negligible. Evidence has been produced that this is not allways the case in this country. On the contrary there are situations where even more fallout is brought down by turbulent deposition than by precipitation. The detailed mechanisms in connection with turbulent deposition have not been studied by us. Most probably the extent of this process depends on a number of local climatological

Sample		Sr-90 pc/l Ave	Sr-90 pc/l Max	Cs-137 pc/l Ave	Cs-137 pc/l Max
Precipitation					
Bergen	1957	2.5	5.6	1.7	4.5
Kjeller	1957	3.0	13.5	5.3	45.3
Ski	1957	2.0	2.7	2.1	3.2
Bergen	1958	6.3	26.2	6.4	23.7
Kjeller	1958	8.5	27.4	8.7	27.9
Ski	1958	3.8	12.7	4.6	15.0
Møsvann	1958	5.0	18.2	5.6	21.9
Tap water					
Bergen	1957	0.43	0.65	0.39	0.82
Kjeller	1957	0.39	0.75	0.24	0.47
Stavanger	1957	(0.40)	0.52	(1.23)	3.18
Bergen	1958	0.84	1.70	0.50	1.04
Kjeller	1958	0.67	1.67	0.40	0.50
Stavanger	1958	0.78	1.68	0.90	2.81
Milk					
Norway	1957	7.7	15.4	17.1	62.9
Norway	1958	11.4	23.4	38.1	80.5
Norway	1959 (6 mon)	17.8	70.2	92.2	276.1

Table 8.1 Average and maximum values for Sr-90 and Cs-137 in precipitation, tap water and milk sampled in Norway 1957, 1958 and 1959

factors, for instance air turbulens, temperature inversion and local fog formation. This field may be an interesting and important field for future studies.

The contamination of tap water has been studied in several areas and in one case been related to the amounts of fallout in precipitation and details of the precipitation area. The decontaminating properties

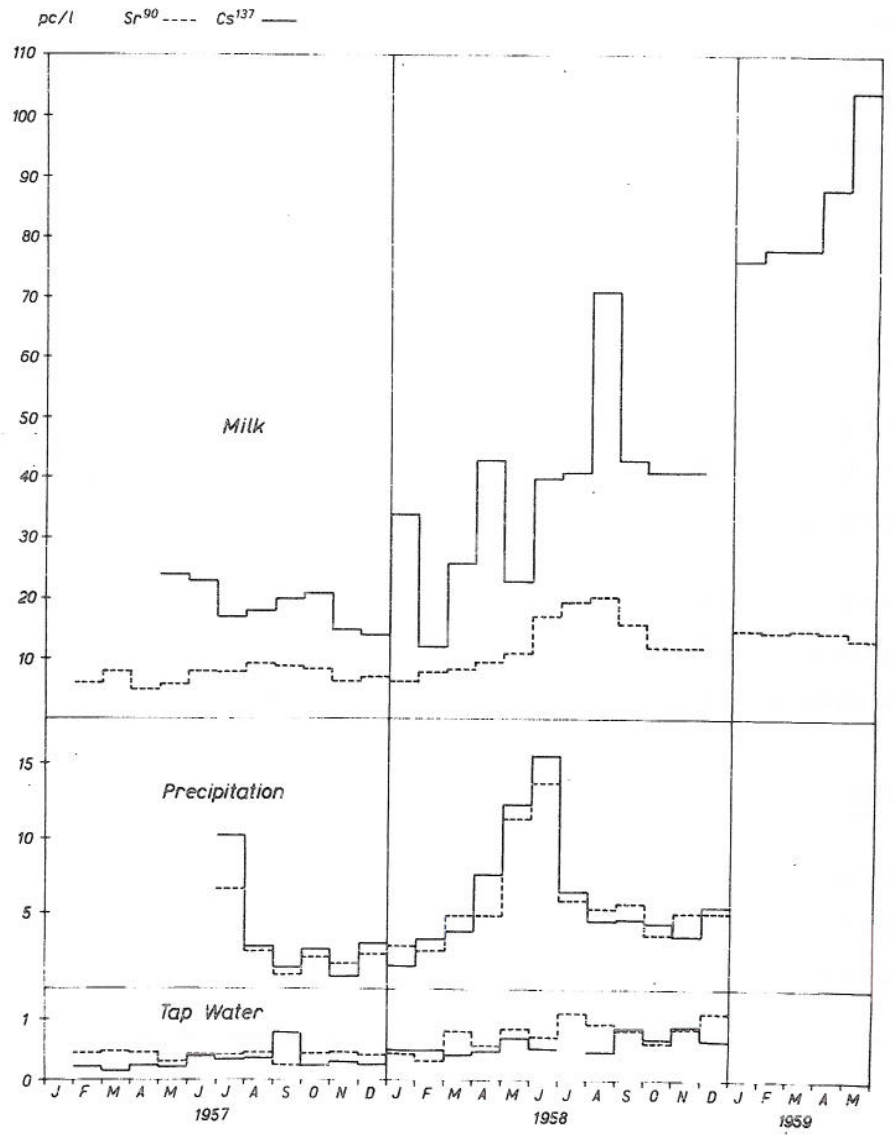

Figure 8.1 Monthly average concentrations of Sr-90 and Cs-137 in tap water, precipitation and milk sampled in Norway 1957, 1958 and 1959

of this precipitation area have been discussed and probably more studies of the factors of importance in this connection might give the water works some basis for calculating the degree of contamination to be expected in an emergency situation.

The principal contribution of Sr-90 and Cs-137 to the norwegian population comes through milk and milk products. The increase and yearly variations of the content of these isotopes in the milk have been discussed. At present the average concentrations of Sr-90 and Cs-137 are well below the limits recommended by the International Commission on Radiological Protection, and in a recent report (68) from health authorities in Scandinavia no security measures have been considered necessary at the present level. The present studies have been performed with milk sampled from central dairies. In future, it may be important to study the dominant factors affecting the uptake of fallout isotopes in milk. More information concerning these factors is imperative if decontamination of milk has to be considered.

At several occations in the present report the possibilities of disclosure and identification of nuclear bomb tests have been discussed. For this purpose it may be recommended that the Ba-140 analysis of air filters and precipitation is taken into routine use at a few stations, also I-131 analysis in precipitation, air and milk seems to be a promising approach. An early and exact identification of bomb tests and other possible sources of radioactive contamination is important for the evaluation of future hazards and vital in case of nuclear disasters.

Independent of the special investigations suggested in this report, it is felt that the "cumulative studies" of fallout in norwegian soil, diet etc should be continued. The sampling for this purpose should be limited to suitable time intervals and arranged to cover the country in a statistical, representative way (69). On the other hand, when and where "fine structure" investigations of the fallout are desired, they should be performed in great detail. If capacity is limited, such investigations should rather be restricted to certain periods and areas, than in number of samples in the period under study.

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APPENDIX I

RADIOCHEMICAL PROCEDURES FOR THE DETERMINATION OF Sr-90, Sr-89, Cs-137 AND I-131 IN WATER, MILK, PLANTS AND SOIL

Contents

1 REAGENTS AND CARRIER SOLUTIONS

The chemicals used must be radiochemically pure, or at least, contain a negligible activity compared with any single activity to be measured in a particular sample. Radioactivity tests should be run on each single reagent and on blanks following the radiochemical procedure in question.

1.1 List of reagents

Acetic acid 6 M
Ammonia 6 M and 1 M, Ammonium acetate 3 M
Ethyl alcohol 96 per cent and 100 per cent
Hydrochloric acid concentrated and 6 M
Hydrogen peroxide 35 per cent
Nitric acid fuming sp gr 1.49, concentrated and 6 M
Oxalic acid 8 per cent
Perchloric acid concentrated
Phenolphtalein 1 per cent in alcohol
Platinum chloride 0.2 M
Silicotungstic acid 400 g/l
Silvernitrate 1 g $AgNO_3$/100 ml
Sodium chromate 1.5 M, Sodium hydroxide 1 M and 6 M
Sulfurdioxide water saturated
Sulfuric acid 3 M

It is important to know the exact concentration of the nitric acid. The specific gravity of fuming nitric acid should therefore be controlled every time a new supply is taken into use. The volumes used to separate strontium from calcium in the radiochemical procedure should be adjusted accordingly.

The ammonia must be carbonate-free to prevent undue precipitation of strontium.

2. PREPARATION OF CARRIER SOLUTIONS

2.1 Strontium carrier solution, 10 mg Sr/ml

24 g of strontium nitrate is dissolved in water, 5 ml conc HNO_3 is added and the solution made up to 1 liter. If the carbonate is used,

17 g is dissolved in nitric acid (about 16 ml conc HNO_3), the carbon dioxide is boiled off, the solution is cooled, filtered if necessary, and diluted to 1 liter after adding 5 ml conc HNO_3.

Two 5 ml portions of the carrier solution are each transferred to a 100 ml beaker. 2 ml 3 M H_2SO_4 are added while shaking, then 7 ml alcohol, and the precipitate is allowed to stand overnight. The following day the $SrSO_4$ solution is filtered through a S-S 589 filter paper, the precipitate is washed with 50 % alcohol containing some drops of H_2SO_4 and then with abs alcohol until the filtrate is acid-free. The filter paper is transferred to a porcelain crucible, ashed at 300°C, ignited at 700°C and weighed.

2.2 Yttrium carrier solution, 10 mg Y/ml

13 g of yttrium oxide is dissolved in conc HNO_3 and water (about 24 ml conc HNO_3). After filtration dilute to 1 liter.

Two 20 ml portions of the carrier solution are each mixed with 20 ml 8 % oxalic acid. After heating on a water bath for 15 min, it is cooled and filtered through a S-S 589 filter paper. The beaker is rinsed with alcohol to secure quantitative transfer to the filter paper, and the precipitate is washed 2 - 3 times with alcohol. The filter is then ashed in a porcelain crucible and ignited to constant weight at 800 - 900°C.

2.3 Cesium carrier solution, 10 mg Cs/ml

15 g of cesium nitrate is dissolved in water. 5 ml conc HNO_3 is added and the solution is made up to 1 liter. If the chloride is used, 13 g is mixed with 25 ml conc HNO_3 and evaporated on a water bath to near dryness. The residue is dissolved in water and diluted to 1 liter.

Two 5 ml portions are each mixed with 5 ml conc. HCl. 4 ml 0.2 M platinum chloride solution is added, stir well and let stand for 10 min at 70°C, cool and filter through a tared porcelain filter crucible. The precipitate is washed 3 times with 5 ml portions of 6 M HCl and 3 times with 5 ml portions of abs alcohol. The crucible

is dried for 20 min at 110°C, cooled in a desiccator and weighed.

2.4 Cerium carrier solution, 10 mg Ce/ml

A radiochemically pure cerium-salt is not easily obtained. Examination pointed out ammonium cerium (IV) sulphate to be the best of the cerium compounds stocked in our laboratory. It was converted to the cerium (III) nitrate. Standardization is not necessary since cerium only acts as a holdback carrier.

45 g of $2\,(NH_4)_2SO_4 \cdot Ce(SO_4)_2 \cdot 2\,H_2O$ is dissolved in 500 ml cold water and the solution is filtered directly into a suitable centrifuge tube. Concentrated ammonia is added while stirring until no more cerium is precipitated. When centrifuged, the supernate is discarded and the precipitate is suspended in 500 ml dilute ammonia (about 1 M). After vigorous stirring it is again centrifuged and the supernate rejected. The washing procedure is repeated 4 times. The hydroxide is then dissolved by addition of 6 M HNO_3 until the solution is slightly acid, it is warmed to 60 - 70°C on a water bath and while stirring vigorously, 35 % H_2O_2 is added slowly until the solution is colorless. A few drops of hydrogen peroxide is added in excess and the solution is boiled for about 5 min. The solution is then cooled and diluted to 1 liter.

To test the presence of sulphate, 2 ml of the cerium solution is mixed with 2 ml of the strontium carrier solution. If a turbidity or precipitate results, the precipitation with ammonia and washings have to be repeated.

If a cerium (III) salt is used, the reduction with H_2O_2 may be omitted.

2.5 Barium carrier solution, 10 mg Ba/ml

Barium is used as a "scavenger" and no standardization is needed. 19 g of $Ba(NO_3)_2$ is dissolved in water and made up to 1 liter.

2.6 Iodine carrier solution, 2 mg I/ml

2.36 g of NaI is dissolved in dist water, 5 ml conc HNO_3 is added and the solution is made up to 1 liter with dist water.

Two 10 ml portions of NaI solution containing 20 mg I are each precipitated with 2.0 ml $AgNO_3$ solution. Yellow AgI is settled by boiling, filtered on a prepared and weighed filterdisc covered with a S-S 589 filter paper. The precipitate is washed with water and aceton, dried under an infrared lamp and weighed.

3 DETERMINATION OF Sr-90 AND Sr-89 IN WATER

3.1 The contents of the sample flask is transferred to a suitable beaker (800 - 1000 ml) and the flask is washed with a little dilute HNO_3 (1 M). The sample is evaporated to 20 - 25 ml and filtered. The filter is washed with a little water, and the filtrate set aside. The filter paper containing the solids is ashed in a platinum crucible and ignited at 500°C. After cooling the residue is thoroughly mixed with 3 times its own volume of Na_2CO_3 and fused. The melt is dissolved in a small amount of 6 M $HClO_4$, heated to fumes of perchloric acid. The contents of the crucible is transferred to a centrifuge tube with a little water and then centrifuged. The supernate is added to the filtrate mentioned above. Residual solids are dried and tested for radioactivity.

All samples examined so far in our laboratory have given residual solids in varying amounts. These solids (probably silicates) are often comparatively active, in some cases they have activities far exceeding those found for Sr-90 and Cs-137 in the same sample. Addition of carriers for strontium and cesium to the residual solids and repeated treatments with Na_2CO_3 and perchloric acid have in all cases failed to give additional strontium and cesium activities. Nor have complete dissolution of the solids by means of hydrofluoric acid given any additional activities.

3.2 The combined filtrates are evaporated in the centrifuge tube to about 25 ml. Cerium is precipitated by addition of the least possible amount of 1 M NaOH, and then 0.25 g solid Na_2CO_3 is added to precipitate strontium. It is centrifuged and the supernate reserved for the determination of radiocesium (4).

3.3 The precipitate in the centrifuge tube is dissolved in a minimum amount of 6 M HNO_3 and CO_2 is boiled off. Cerium is precipitated with carbonate-free NH_3, the solution is warmed to coagulate the

precipitate and then centrifuged. The precipitate is treated again with HNO_3 and cerium precipitated and centrifuged. The supernates are analyzed for radiostrontium. The precipitate is rejected if the rare earths are not to be extracted.

3.4 0.25 g $(NH_4)_2CO_3$ is added to the solution from 3.3. It is warmed and centrifuged. The supernate is rejected.

3.5 To separate strontium from calcium 10 ml water and 28 ml fuming HNO_3 (sp gr 1.49) are added. The acid must be added slowly and carefully under agitation. The tube is cooled in ice for 20 minutes before centrifugation. The supernate is rejected.

3.6 The strontium nitrate is dissolved in 10 - 15 ml of water, 2 ml of barium carrier solution and 1 drop of methyl red indicator are added. Excess acid is neutralized with 6 M ammonia and the solution is buffered with 1 ml of 6 M acetic acid and 2 ml of 3 M ammonium acetate solution. The solution is diluted to about 30 ml, heated on a boiling water bath and barium precipitated with 1 ml 1.5 M Na_2CrO_4 solution. After stirring and heating for about 5 min, it is centrifuged, and the supernate transferred to a clean tube. The barium chromate is rejected.

3.7 Repeat of 3.4.

3.8 Repeat of 3.5.

3.9 The strontium nitrate is dissolved in 10 - 15 ml water. 1 ml of yttrium carrier solution is added and the solution is stored for 12 days to build up the Y-90 activity.

3.10 After 12 days storage the solution is made alkaline with carbonate-free ammonia and heated on a boiling water bath. The time for the separation of yttrium from strontium is reported. After centrifugation the supernate is transferred to another tube, whilst the precipitate is dissolved in 6 M HNO_3, diluted to 10 - 15 ml and precipitated with carbonate-free NH_3. Centrifugation is repeated and the two supernates are combined and treated as in 3.13.

3.11 The precipitate is dissolved in the minimum amount of 6 M HNO_3, added dropwise. 20 ml of 8 % oxalic acid is added. It is warmed on a water bath for 10 - 15 min. After cooling it is filtered on a tared filterdisc in the filterstick (S-S 589 filter paper). The precipitate is washed three times with alcohol. The yttrium oxalate is dried under an infrared lamp to constant weight. The radioactivity of the yttrium sample is measured as soon as possible and the time for measurement reported.

3.12 When the yttrium oxalate has been counted, the filter paper is moistened with alcohol and transferred to a tared porcelain crucible. The paper is ashed and the crucible ignited for 1 hour at 800 - 900°C. The Y-recovery is calculated from the weight of Y_2O_3.

3.13 From the combined supernates (3.10) strontium is precipitated by addition of 0.25 g $(NH_4)_2CO_3$ and heating on a water bath. After cooling it is filtered through a tared disc (S-S 589 filter paper) and washed first with water and then with alcohol. The carbonate is dried in air to constant weight and the Sr-recovery calculated. Immediately afterwards the carbonate is counted. The time is reported.

3.14 The activity of Sr-90 and Sr-89 in the sample is calculated and expressed in pc/l. (1 pc = 1 picocurie = 10^{-12} curie).

$$\text{Activity}_{\text{Sr-90}} = \frac{C_{\text{Y-Ox}} \cdot F_{\text{Y-90}} \cdot 100 \cdot 100 \cdot f_1}{R_{\text{Sr}} \cdot R_{\text{Y}} \cdot V \cdot 2.22} \quad \text{pc/l}$$

$C_{\text{Y-Ox}}$: Net counts per min for the Y-oxalate

$F_{\text{Y-90}}$: Standard factor for Y-90

f_1 : Correction factor for the decay of Y-90

R_{Sr} : Recovery of strontium (per cent)

R_{Y} : Recovery of yttrium (per cent)

V : Amount of sample in liters

$$\text{Activity}_{Sr\text{-}89} = \text{Activity}_{Sr\text{-}90+89} - \text{Activity}_{Sr\text{-}90}$$

$$= \frac{C_{SrCO_3} \cdot F_{SrCO_3} \cdot 100 \cdot f_2}{R_{Sr} \cdot V \cdot 2.22} - \text{Activity}_{Sr\text{-}90} \text{ pc/l}$$

C_{SrCO_3} : Net counts per min for the $SrCO_3$

F_{SrCO_3} : Standard factor for $SrCO_3$

f_2 : Correction factor for grow-in of Y-90

The other symbols represent the same as above.

A period of 12 days for growing-in of Y-90 will give only 95.6 per cent of the Y-90 equilibrium value. In accurate work correction should be made also for this.

4 DETERMINATION OF Cs-137 IN WATER

4.1 The solution from 3.2 containing the cesium is made slightly acid with 6 M acetic acid, carbon dioxide is boiled off and the volume of the solution is reduced to about 25 ml. The solution is cooled in ice and about 0.5 g of sodium cobaltinitrite is added, stirred well and set aside to settle for 10 - 15 min, centrifuged, and the supernate rejected.

4.2 The precipitate is dissolved in 0.5 - 1.0 ml 6 M HNO_3 (warmed if necessary). After cooling in ice, 5 ml conc $HClO_4$ is added followed by 15 ml alcohol. The precipitate is allowed to settle, centrifuged and the supernate rejected.

4.3 The cesium perchlorate is dissolved in 4 ml water. 12 ml conc HCl is added, after which the solution is diluted to 20 ml and 1 ml of the silicotungstic acid is added. After stirring and waiting for 5 minutes, it is centrifuged, and the supernate rejected. The precipitate is washed twice with 10 ml 6 M HCl and the washings rejected.

4.4 The precipitate is dissolved in 5 ml 1 M NaOH (heating) and 5 ml

conc $HClO_4$ is added. The solution is evaporated until white fumes of perchloric acid and the heating is continued for another 20 minutes. It is diluted to 10 ml, centrifuged and the supernate is transferred to a clean tube (supernate 1). The precipitate is dissolved in 5 ml 1 M NaOH, 5 ml conc $HClO_4$ is added and the above procedure repeated. The second supernate is combined with supernate 1, and the residue which should be of a bright yellow color, is discarded.

4.5 The combined supernates are cooled in ice, acidified with 6 M HCl if not already acid, (2 ml in excess). After addition of 15 ml alcohol, cesium is precipitated with 1 ml 10 % platinum chloride. It is stirred and cooled in ice for 10 - 15 min, centrifuged and the supernate rejected.

4.6 5 - 10 ml of ice-cooled alcohol is added to the precipitate, stirred and filtered through a tared disc. Cold alcohol is used to transfer the chloroplatinate quantitatively to the filterdisc. The cesium chloroplatinate is dried in air until constant weight and the carrier yield is calculated. The activity of the sample is measured.

4.7 The beta-activity of Cs-137 in the original sample is calculated and expressed in pc/l.

$$\text{Activity}_{\text{Cs-137}} = \frac{C_{Cs_2PtCl_6} \cdot F_{Cs} \cdot 100}{R_{Cs} \cdot V \cdot 2.22} \quad \text{pc/l}$$

$C_{Cs_2PtCl_6}$: Net counts per min for the cesium chloroplatinate

F_{Cs} : Standard factor for Cs-137

R_{Cs} : Recovery of Cs in per cent

V : Amount of sample in liters

5 DETERMINATION OF Sr-90 AND Sr-89 IN MILK

Milk offers particular analytical problems mainly owing to its high content of calcium and potassium which complicates the separation of strontium and cesium respectively. The procedure for milk differs so much from that of water that a detailed description is offered.

Sr-90 activity in milk and other materials rich in calcium, are often expressed in strontium-units, i e pc of Sr-90 per g of Ca. We have expressed our data in pc per liter of milk. These data, however, will not differ appreciably from the corresponding strontium-units because 1 liter of milk contains about 1 g of calcium.

5.1 The milk ash is dissolved in 30 ml conc HNO_3 and 50 ml water. If the ash originates from dry milk, carriers are added at this stage. Then it is filtered directly into a 500 ml centrifuge tube and the filter washed with 10 - 15 ml water. The filter paper should now be practically free from residual particles, but if a more or less dark film of solids should remain, the filter paper can nevertheless be discarded. No strontium or cesium activities have been detected on these filters. The volume of the filtrate is estimated by comparison with a known volume in an identical tube. It is usually about 100 ml.

5.2 With mechanical stirring a volume of fuming HNO_3 twice that of the filtrate is added. The stirring is continued for 15 min, then it is cooled in ice and centrifuged. The supernate is reserved for radiocesium separation (6) and is conveniently decanted into a 500 ml beaker, while the precipitate is worked up for radiostrontium.

5.3 The precipitate (from 5.2) is dissolved in 100 ml water and precipitated as in 5.2 by addition of 200 ml fuming HNO_3, centrifuged and the supernate discarded.

5.4 The precipitate is dissolved in 60 ml water, and 160 ml fuming HNO_3 is added, centrifuged and the supernate is discarded.

5.5 The strontium nitrate is dissolved in 20 - 25 ml water, 20 mg Ba-carrier and 1 drop of methyl red indicator are added. The solution is neutralized with 6 M NH_3 (carbonate-free), buffered with 1 ml 6 M acetic acid and 2 ml 3 M ammonium acetate solution. It is then diluted to about 50 ml, warmed on a water bath and 1 ml 1.5 M sodium chromate solution is added. After agitation and standing on the water bath for 5 min, it is cooled and centrifuged. The supernate is decanted into a clean tube, and the barium chromate discarded.

5.6 The supernate is made alkaline with 6 M NH_3 and 1 g of solid $(NH_4)_2CO_3$ is added. After agitation to dissolve the carbonate, the solution is placed on a water bath for 15 min. Then it is cooled, centrifuged and the supernate rejected.

5.7 The precipitate is dissolved in 15 ml water and 40 ml fuming HNO_3 is added slowly while stirring vigorously. It is cooled in ice, centrifuged and the supernate rejected.

5.8 The strontium nitrate is dissolved in 25 ml water and 1 ml of yttrium-carrier solution (10 mg Y) is added. The solution is stored for 12 days.

5.9 It is now continued as described in paragraphs 3.10, 3.11, 3.12, 3.13 and 3.14 with the exception that in 3.13 1 g of solid $(NH_4)_2CO_3$ is used for the precipitation of strontium carbonate.

6 DETERMINATION OF Cs-137 IN MILK

The acid supernate from 5.2 is evaporated almost to dryness.

6.1 After cooling a sticky residue will appear. This is dissolved in 250 ml water and transferred to a 500 ml beaker. The solution is made slightly alkaline (pH about 9) with 6 M NaOH, the latter being added slowly while stirring vigorously. A voluminous precipitate occurs, which after cooling is centrifuged and the supernate decanted into a 500 ml beaker. The precipitate is suspended in 200 ml water and centrifuged. The supernate is

combined with the first one in the beaker and the precipitate rejected.

6.2 The liquid in the beaker is evaporated to about 100 ml and then transferred to a 250 ml centrifuge tube with an equal amount of conc HCl. The tube is left in ice water for an hour, centrifuged and the supernate is decanted into a clean tube. 2 ml silicotungstic acid solution is added and the mixture allowed to stand over night.

6.3 After centrifugation the supernate is rejected, the precipitate is washed once by suspension in 20 ml 6 M HCl and recentrifugation. The residue is dissolved in 10 ml 1 M NaOH and transferred to a 50 - 60 ml centrifuge tube with the minimum amount of water. 7.5 ml conc $HClO_4$ is added and the tube is heated until white fumes of perchloric acid. The heating is continued for 20 min, then cool and dilute with 15 ml water, the tube is vigorously shaken and centrifuged. The supernate is decanted into a 250 ml centrifuge tube, the residue is dissolved in 10 ml 1 M NaOH, and 7.5 ml conc $HClO_4$ is added. The procedure described is repeated and the supernates are combined in the 250 ml tube.

6.4 5 ml conc HCl is added to the solution in the tube and sufficient alcohol to make a final concentration of 45 - 50 % (V/V). When cooled in ice water the solution should remain clear. If any turbidity occurs, the solution must be centrifuged. Then 2 ml of platinum chloride solution is added and the tube is allowed to remain in ice water for one hour.

6.5 The precipitate is filtered through a tared filterdisc (S-S 589 filter paper) and is washed 5 - 6 times with 45 - 50 % alcohol. The cesium chloroplatinate is dried under an infrared lamp until constant weight and the chemical yield of Cs is calculated. The activity of the sample is measured.

6.6 The activity of Cs-137 in the original sample is calculated as described in 4.7.

7 DETERMINATION OF I-131 IN MILK

7.1 A 200 ml sample is placed in a separatory funnel of 1500 - 2000 ml capacity. To this is added 10 ml NaI carrier solution, 1 liter dist water, 50 ml ethyl ether, 2 - 3 g of $NaNO_2$ and 200 ml CCl_4.

7.2 After thorough mixing, 5 - 10 ml conc HNO_3 is added carefully. (Release gas pressure.) Free I_2 is extracted with 100 ml CCl_4.

7.3 The CCl_4 layer is allowed to settle and is transferred to another funnel. After 3 - 4 extractions with 50 ml portions the CCl_4 will be colorfree. Remaining milk is used for determination of organic iodine (7.7).

7.4 The combined CCl_4 portions are washed with water and centrifuged. The CCl_4 is decanted and shaken 2 - 3 times with 10 - 20 ml of dist water to which is added 1 ml of saturated sulfurous acid solution.

7.5 The iodine solution from 7.4 is adjusted to pH 8 with NaOH and phenolphtalein. Sulfur dioxide is boiled off. The solution is cooled and acidified with HNO_3 to pH 3 - 4. 2.0 ml $AgNO_3$ solution is added and the solution is heated to boiling.

7.6 The precipitate is collected on a tared filterdisc with a S-S 589 filter paper. The precipitate is washed with water and acetone, dried under an infrared lamp, weighed and the activity measured. Note. The operations 7.5 - 7.6 should be carried out in darkness.

7.7 The milk from 7.3 is made alkaline with NaOH, 10 ml NaI carrier solution is added and the sample is evaporated and ashed. The ash is transferred to a separation funnel with 1.0 - 1.5 liters of water, and treated as in 7.1 - 7.6.

7.8 The weight of AgI is used for recovery calculation. The activity of I-131 in the milk sample is calculated and expressed in pc/l.

$$\text{Activity}_{\text{I-131}} = \frac{C_{AgI} \cdot F_{AgI} \cdot f \cdot 100}{R_I \cdot V \cdot 2.22} \quad \text{pc/l}$$

C_{AgI} : Net counts per min for the AgI

F_{AgI} : Standard factor for I-131

f : Correction factor for decay of I-131

R_I : Recovery of iodine in per cent

V : Sample in liters

Sum of inorganic and organic I-131 gives total I-131 activity in sample.

8 MODIFIED PROCEDURES

8.1 Pretreatment of peat soil and plants

According to radioactivity expected 100 - 500 g of dry ground sample is ashed in quartz vessels at 450°C. The ash is weighed and placed in a 400 ml beaker. Sr-carrier 100 mg, Cs-carrier 50 mg and Ce-carrier 20 mg are added. The ash is dissolved by boiling for 15 min with 25 ml 6 M HNO_3 per gram ash. Undissolved rest is filtered on a S-S 589 filter paper, the filtrate is placed in a 500 ml centrifuge tube, dist water is used for washing. Undissolved rest is ashed in a platinum crucible, 5 ml HF is added and the mixture is evaporated three times with conc HNO_3. The residue is heated with 10 ml 6 M HNO_3 in the platinum crucible and transferred to a 100 ml centrifuge tube and centrifuged. The supernate is added to the filtrate in the 500 ml centrifuge tube. The undissolved rest is shaken with 5 ml dist water and centrifuged, the supernate added to the filtrates. Undissolved rest is filtered, dried, weighed and counted for radioactivity.

8.2 Pretreatment of milk

250 - 300 g of dry milk is ashed in quartz vessels at 450°C. The ash is weighed into a beaker, and carriers are added as for peat soil (8.1). The ash is dissolved by boiling for 15 min in 75 ml 6 M HNO_3. Undissolved is filtered on a S-S 589 filter paper and washed with dist water. This rest will not contain Sr-90, Sr-89 or Cs-137, and is discarded.

8.3 Pretreatment of water

Water samples are received as eluates from the ion exchanger. The solution is evaporated to 25 ml volume and transferred to a 100 ml centrifuge tube.

8.4 Separation of Sr from Cs

Acid filtrates from 8.1 - 8.3 are made alkaline with NaOH to pH 8, stirred, and 300 mg solid Na_2CO_3 is added. Stirring is continued until all sodium carbonate is dissolved. The precipitated hydroxides and carbonates are centrifuged and the supernate is transferred to a 500 ml centrifuge tube. The precipitate is washed with 50 ml hot dist water and centrifuged. The supernates are combined and go to Cs determination, the precipitate to Sr determination.

8.5 Sr separation

The precipitate containing Ca and Sr carbonates is carefully dissolved in the minimum quantity of 6 M HNO_3, and twice the volume of fuming HNO_3 is added with stirring. After 10 min stirring it is cooled in ice, centrifuged and supernate decanted into a glass stoppered flask and stored. The precipitate is dissolved in 80 ml dist water and precipitated again with twice the volume of fuming HNO_3. After cooling in ice the precipitate is centrifuged and supernate decanted and discarded.

The precipitate is dissolved in 25 ml dist water, transferred to a 100 ml centrifuge tube, 20 mg Ba-carrier and 1 drop of methyl red indicator added. The solution is neutralized with 6 M carbonate-free ammonia. 1 ml 6 M HNO_3, 1 ml acetic acid and 2 ml 3 M ammonium acetate are added. Then water to 50 ml, and 1 ml 1.5 M sodium chromate is mixed in. The tube is heated on a water bath for 5 min and centrifuged. The supernate is decanted into another centrifuge tube and the $BaCrO_4$ is stored. The Sr solution is neutralized with 6 M NH_3 and 1 g ammonium carbonate is dissolved by shaking. The suspension is heated for 15 min on a water bath and centrifuged. The $SrCO_3$ is filtered on a filterdisc, carefully dried and weighed. Count after 12 days.

8.6 Y-90 milking

After counting, the $SrCO_3$ is dissolved in a 50 ml beaker in 10 ml 6 M HNO_3. 20 mg Y-carrier is added and CO_2 is boiled off. The solution is transferred to a 100 ml centrifuge tube, and $Y(OH)_3$ is precipitated by adding conc carbonate-free NH_3 dropwise. Time is reported. The precipitate is centrifuged and the supernate is transferred to another 50 ml centrifuge tube. The precipitate is dissolved in a minimum quantity of 6 M HNO_3, diluted to 10 ml with dist water and heated on a water bath for 5 min. $Y(OH)_3$ is precipitated again as above, after centrifugation the supernate is added to the 50 ml centrifuge tube. The $Y(OH)_3$ is dissolved again in 6 M HNO_3 and yttrium precipitated by addition of 20 ml 8 per cent oxalic acid. The precipitate is heated on a water bath for 10 min and filtered through a filterdisc covered with a S-S 589 filter paper. The Y-oxalate is washed 3 times with dist water, a little alcohol, dried, weighed and counted immediately and time is reported. After beeing counted, the Y-oxalate is ignited at 800°C and Y recovery is calculated. To the centrifugate is added 0.25 g ammonium carbonate, the tube is heated for 5 min on a water bath and the precipitate is filtered on a tared disc. The disc is stored for control.

8.7 Cs separation

The supernate from 8.3 is diluted with equal amounts of conc HCl. NaCl precipitates together with some KCl. It is cooled in ice and centrifuged. The supernate is transferred to a 500 ml centrifuge tube and 2 ml silicotungstic acid is added. The mixture is left over night and then centrifuged. The precipitate is washed twice with 20 ml portions of 6 M HCl and centrifuged, then dissolved in 8 ml 2 M NaOH by heating on a water bath. The solution is transferred to a 100 ml centrifuge tube with a little water, 7.5 ml conc $HClO_4$ is added and the solution is evaporated carefully till fumes of $HClO_4$. This takes approximately two hours. After cooling, 15 ml of water is added and the solution centrifuged. The supernate is decanted into another 100 ml centrifuge tube. The residue is again treated with 8 ml 2 M NaOH and 7.5 ml conc $HClO_4$.

The two supernates are combined and the precipitate is discarded. To the solution is added 5 ml conc HCl, alcohol to 20 - 30 per cent (V/V) and 2 ml platinum chloride solution. This is left over night, and the Cs_2PtCl_6 precipitate is filtered on a tared filterdisc and washed 5 - 6 times with 45 - 50 per cent (V/V) alcohol. The precipitate is dried carefully under an infrared lamp to constant weight and counted. Cs-recovery is calculated from the weight of Cs_2PtCl_6.

APPENDIX II

STANDARDIZATION OF COUNTING EQUIPMENT

Contents

1 STANDARDIZATION OF THE COUNTING EQUIPMENT

To convert the recorded radioactivity of a sample into absolute units a standard factor F is used. This factor is the ratio between the known number of disintegrations per minute dpm of a standard, and the observed number of counts per minute cpm of the standard when measured under exactly the same conditions as the sample.

For instance, in the case of Y-90 the standard factor is given by

$$F_{Y\text{-}90} = \frac{A_{Sr\text{-}90} \cdot R_Y}{f \cdot 100 \cdot C_{Y\text{-}Ox}}$$

$A_{Sr\text{-}90}$: dpm of Sr-90 standard = dpm of Y-90 in equilibrium

R_Y : Recovery of yttrium carrier in per cent

f : Correction factor for decay of Y-90 after separation from Sr-90

$C_{Y\text{-}Ox}$: cpm of Y-oxalate

The standard factor is determined as a function of specimen weight. Normally, the standard factor will be nearly constant at low specimen weights. For larger weights it will increase in proportion to the weight of the specimen, as the counting rate becomes independent of sample thickness.

For standardization, the same isotope as the one under consideration, should be used. If an identical standard is not available, one whose energy spectrum approximates that of the desired isotope as closely as possible, may be applied.

In our case standards from JENER Norway (I-131), AERE England (Sr-90, Y-90, Cs-137, I-131) and US Bureau of Standards USA (Sr-90, Y-90) have been used. Besides K_2CO_3 has also been applied with the activity of 1 g K equal to 1776 dpm.

The standard factors used by us are given in table 1.1. All these factors have been determined under identical counting conditions.

Isotope	Compound	m mg	F dpm/cpm
Sr-89	$SrCO_3$	20 - 250	6.5
Sr-90	$SrCO_3$	40 - 250	6.5 - 12.0
Y-90	Y-oxalate	20 - 100	6.5
Cs-137	Cs_2PtCl_6	20 - 50	5.0
I-131	AgI	20 - 40	10.0
K-40	K_2CO_3	20 - 350	7.0 - 9.0

Table 1.1 Standard factors

For the standardization of total activities of fallout, preparations of K_2CO_3 are frequently used.

In the following the procedure for standardization with each isotope is given.

1.1 Standardization with Y-90

Standard solutions from AERE England and from US Bureau of Standards have been used.

Known activities of carrierfree Sr-90 in equilibrium with Y-90, and 10 mg of Y-carrier are mixed, and Y-oxalate precipitated as described in Appendix I, p 6. After counting and correction for decay, the standard sample is ignited and weighed as Y_2O_3 to determine the recovery of the analytical procedure.

The standardization curve shows no appreciable dependency upon the weight of the specimen in the range 20 - 100 mg, and a constant value of F = 6.5 has been used.

1.2 Standardization with Sr-90

In the remaining solution from the Y-90 standardization Sr-90 is

worked up as described in Appendix I, p 4, with varying amounts of Sr-carrier.

The specimen activity is corrected for any growing in of Y-90. The sample is usually counted again after a period of 12 days to check the build up of Y-90. The weight of $SrCO_3$ is used to calculate the recovery.

The standard factor increases approximately linearly with the weight of the specimen in the range 40 - 250 mg $SrCO_3$. The standard factor has been expressed by $F = 5.50 + 6 \cdot m/250$ where m is the weight of the specimen in mg $SrCO_3$.

1.3 Standardization with Sr-89

A standard solution from AERE England has been used.

From 10 - 150 mg Sr-carrier is added to known activities of carrierfree Sr-89, and a $SrCO_3$ sample is worked up as described in Appendix I, p 4. The weight of $SrCO_3$ is used for calculation of recovery.

The standard factor shows no appreciable dependency upon the weight of the specimen in the range 15 - 250 mg $SrCO_3$ and a constant value of $F = 6.5$ has been used.

1.4 Standardization with Cs-137

A standard solution from AERE England has been used.

Known activities of carrierfree Cs-137 with 10 - 40 mg Cs-carrier is worked up as described in Appendix I, p 7, weighed and counted as Cs_2PtCl_6.

The standard factor shows no appreciable dependency upon the weight of the specimen in the range 10 - 100 mg Cs_2PtCl_6, and a constant value of $F = 5.0$ has been used.

1.5 Standardization with K_2CO_3

Powdered anhydrous K_2CO_3 p a is dried at 110°C and suspended in abs alcohol. Varying portions of the suspension are filtered through tared discs, dried at 50°C and weighed. To protect it from atmospheric moisture, the sample is finally moistened with a few drops of a thin cellulose varnish and allowed to dry.

The standard factor shows no appreciable dependency upon specimen weight in the range 20 - 50 mg. Above 50 mg it increases approximately in proportion to the specimen weight.

The standard factor obtained may be expressed by $F \approx 7.15$ in the range 10 - 50 mg K_2CO_3, and by $F \approx 7.15 + 0.0064\,(P - 50)$ in the range 50 - 350 mg K_2CO_3. P is the weight of K_2CO_3 in mg.

1.6 Standardization with I-131

Standard solutions from JENER Norway and AERE England have been used. Known activities of carrierfree I-131 are mixed with 10 - 20 mg I-carrier and worked up as described in Appendix I, p 12. The weight of the precipitated AgI is used for recovery calculation.

Within the range 20 - 40 mg AgI the standard factor shows no appreciable dependency upon the weight of the specimen and a constant value of $F \approx 10$ has been used.

1.7 Calculation of Sr-89 activity in a mixed sample

In practise the unknown Sr sample is usually a mixture of Sr-89, Sr-90 and possibly Y-90. In this case the absolute activity of Sr-89 may be calculated in the following way.

If C_T is the measured activity cpm of the mixed sample, and C_{Sr-90}, C_{Sr-89} and C_{Y-90} are the contributions from the single isotopes present, then

$$C_T = C_{Sr-90} + C_{Sr-89} + C_{Y-90} \qquad (1.1)$$

The equilibrium value of Y-90 will be denoted C°_{Y-90}.

If the standard factor for Sr-90 alone under the same conditions (same weight of carbonate, same geometry etc) is denoted by F_{Sr-90}, then

$$A_{Sr-90} = C_{Sr-90} \cdot F_{Sr-90} \qquad (1.2)$$

where A is the absolute activity dpm of the Sr-90 present. Similarly, for other isotopes

$$A_{Sr-89} = C_{Sr-89} \cdot F_{Sr-89} \qquad (1.3)$$

$$A_{Y-90} = C_{Y-90} \cdot F_{Y-90} \qquad (1.4)$$

The value of A_{Sr-90} is known from the milking of Y-90 in a separate process. The value of A_{Y-90} may be calculated from the age of the $SrCO_3$ sample.

From eqs (1.1 - 1.4) the following equation for A_{Sr-89} is obtained

$$A_{Sr-89} = \left(C_T - \frac{A_{Sr-90}}{F_{Sr-90}} - \frac{A_{Y-90}}{F_{Y-90}}\right) F_{Sr-89} \qquad \ldots\ldots\ldots \quad (1.5)$$

Since F_{Sr-89} and F_{Y-90} are independent of the specimen weight in the range of interest, we may introduce C^o_{Y-90} (the cpm value of Y-90 in the equilibrium mixture), by

$$A_{Sr-90} = A^o_{Y-90} = F_{Y-90} \cdot C^o_{Y-90} \qquad (1.6)$$

and

$$A_{Y-90} = F_{Y-90} \cdot C^o_{Y-90} \cdot f(t) \qquad (1.7)$$

where f(t) denotes the fraction of C^o_{Y-90} which is present in the mixed specimen. The value of f(t) depends upon the time elapsed after the milking.

Combination of eqs (1.5), (1.6) and (1.7) gives

$$A_{Sr-89} = [C_T - C^o_{Y-90} \cdot \frac{F_{Y-90}}{F_{Sr-90}} - C^o_{Y-90} \cdot f(t)] F_{Sr-89} \quad \ldots\ldots\ldots (1.8)$$

or

$$A_{Sr-89} = [C_T - C_{Y-90} (q(m) + f(t))] F_{Sr-89} \quad (1.9)$$

where $q(m) = F_{Y-90}/F_{Sr-90}$ is a function of m, the weight of the mixed specimen. Values of q(m) are given in table 1.2.

m mg	20	40	50	100	150	200	250
q(m)	1.09	1.00	0.97	0.82	0.72	0.63	0.57

Table 1.2 Values of q(m) in relation to weight of $SrCO_3$

In the present work usually the two limiting cases with $f(t) = 0$ or $f(t) = 1$ have been utilized.

2 CALCULATION OF STANDARD DEVIATION

The standard deviations given include statistical errors originating from the counting procedure only, as other errors normally are of less significance.

Determination of the radioactivity of a sample in absolute units is connected with 4 independent measurements of counting rates, 2 of which have a bearing on the standard factor. The errors introduced by this factor may, however, be disregarded.

The two counting rates to be considered are then the activities of sample and background. The latter is assumed to be constant within statistical fluctuations during the measurements.

If N_s denotes the total number of pulses recorded when the sample is counted for a time t_s, and N_b and t_b are the corresponding background values, the following relationship holds,

$$\sigma^2 = \left(\frac{\sqrt{N_s}}{t_s}\right)^2 + \left(\frac{\sqrt{N_b}}{t_b}\right)^2 \qquad (2.1)$$

where σ is the standard deviation in counts per min. Since the counting rate is $C = N/t$, this may be written

$$\sigma = \left(\frac{C_s}{t_s} + \frac{C_b}{t_b}\right)^{1/2} \qquad (2.2)$$

where C_s and C_b are the counting rates for sample plus background, and background.

In the case of Sr-89 the activity is determined as a difference between quantities which are measured and calculated separately,

$$A_{Sr\text{-}89} = A_T - 2A_{Sr\text{-}90} \qquad (2.3)$$

which gives

$$\sigma^2_{Sr\text{-}89} = \sigma^2_T + 2\sigma^2_{Sr\text{-}90} \qquad (2.4)$$