Methods of Measuring Radioactivity in the Environment

Mats Isaksson
Lund 1997

DEPARTMENT OF NUCLEAR PHYSICS
UNIVERSITY OF LUND
SWEDEN
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The map on the front cover depicts the deposition of $^{137}\text{Cs}$ from Chernobyl over Skåne on May 8th 1986.
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Fil mag, Kr.

## Title and subtitle
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## Abstract

In this thesis a variety of sampling methods have been utilised to assess the amount of deposited activity, mainly of $^{137}$Cs, from the Chernobyl accident and from the nuclear weapons tests.

Starting with the Chernobyl accident in 1986 sampling of air and rain was used to determine the composition and amount of radioactive debris from this accident, brought to southern Sweden by the weather systems. The resulting deposition and its removal from urban areas was then studied through measurements on sewage sludge and water.

The main part of the thesis considers methods of determining the amount of radiocaesium in the ground through soil sampling. In connection with soil sampling a method of optimising the sampling procedure has been developed and tested in the areas of Sweden which have a comparatively high amount of $^{137}$Cs from the Chernobyl accident. This method was then used in a survey of the activity in soil in Lund and Skåne, divided between nuclear weapons fallout and fallout from the Chernobyl accident. By comparing the results from this survey with deposition calculated from precipitation measurements it was found possible to predict the deposition pattern over Skåne for both nuclear weapons fallout and fallout from the Chernobyl accident.

In addition, the vertical distribution of $^{137}$Cs has been modelled and the temporal variation of the depth distribution has been described.

## Key words
radioactivity, $^{137}$Cs, soil sampling, depth distribution, nuclear weapons fallout, Chernobyl, deposition, sewage sludge
Preface

This thesis is based on the following six papers, referred to in the text by their Roman numerals:


VI: Calculation of the deposition of Cs-137 from the nuclear bomb tests and from the Chernobyl accident over southern Sweden based on the precipitation. M. Isaksson, B. Erlandsson & M.-L. Linderson. Manuscript.

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1. SOURCES OF RADIOACTIVE ISOTOPES IN THE ENVIRONMENT

1.1 Natural radionuclides
The naturally occurring radionuclides could be divided into two different groups according to their origin: primordial and cosmogenic. The primordial radionuclides have their origin in the earth's crust and could belong to a decay series or be singly occurring. Examples of decay series elements often encountered in gamma-spectroscopic measurements are $^{228}$Ac and $^{214}$Bi belonging to the uranium series; among the singly occurring radionuclides $^{40}$K is the most abundant one in environmental measurements.

The cosmogenic radionuclides, on the other hand, have their source in outer space and in the sun as well as in nuclear reactions between the cosmic radiation and the earth atmosphere. The radionuclides reach the ground-level through precipitation or atmospheric mixing. Examples of this group of radionuclides are $^7$Be and $^{14}$C.

1.2 Anthropogenic (man-made) radionuclides

1.2.1 World wide fallout from nuclear weapons testing
Since the first test detonation of a nuclear weapon, "Trinity" at the Alamogordo desert, USA, on July 16 1945, 423 atmospheric tests have been made by the United States, the Soviet Union, the United Kingdom, France and China together (UNSCEAR, 1982). In 1963 a nuclear weapons test-ban agreement was signed by the United States, the United Kingdom and the Soviet Union who thereby stopped the atmospheric tests, but France and China continued until 1974 and 1980, respectively. Atmospheric testing of nuclear weapons was thus performed during the years 1945 - 1980.

The world-wide deposition caused by the atmospheric nuclear weapons tests consisted initially of a large number of different radionuclides where the most long-lived are still present and contribute to the natural background radiation. The fallout consists of gamma, beta and alpha emitting radionuclides where the largest contribution to the effective dose equivalent
commitment to the world population comes from (in order of importance) \(^{137}\)Cs, \(^{60}\)Zr, \(^{90}\)Sr, \(^{106}\)Ru, \(^{134}\)Ce, \(^{3}H, \(^{131}\)I, \(^{239}\)Pu, \(^{140}\)Ba, \(^{103}\)Ru, \(^{240}\)Pu, \(^{241}\)Pu, \(^{54}\)Fe, \(^{241}\)Am, \(^{89}\)Sr, \(^{144}\)Ce, \(^{238}\)Pu, \(^{136}\)Cs, \(^{64}\)Mn and \(^{85}\)Kr. The total effective dose equivalent commitment from these radionuclides has been calculated to 3800 µSv (UNSCERAR, 1982).

Due to its comparatively long half-life (30 y) and abundance in the fallout, \(^{137}\)Cs is today one of the largest contributors to the external dose. In Sweden the deposition of \(^{137}\)Cs from atmospheric nuclear weapons testing amounts to about 1.1 PBq (Mattsson & Lidén, 1975; DeGeer et al., 1978) with the most abundant fallout in the mid sixties. Table 1 shows the yearly deposition of \(^{137}\)Cs from nuclear weapons fallout in Ljungbyhed in the years 1962 to 1981 (DeGeer et al., 1978; FOA 41).

1.2.2 The Chernobyl accident

The importance of accurate methods in assessing the extent and behaviour of radionuclides released in the environment was once again accentuated in the spring of 1986. The accident at the Chernobyl nuclear power plant on April 26 caused a widespread deposition over large areas of the former USSR and in Europe. In Sweden the first reports of increased radiation levels came from the nuclear power plant at Forsmark in the morning of April 28 but the very first traces of radioactivity were shown to have

---

1 The quantity dose equivalent, \(H\), at a point in a tissue is defined by ICRP (1977) as the product \(H = D 	imes QN\), where \(D\) is the absorbed dose, \(Q\) is a quality factor (equals 1 for photons and electrons) and \(N\) is the product of all other modifying factors (presently assigned the value 1 by ICRP). The quantity dose equivalent commitment is then defined as

\[ H_{c,e} = \int_{0}^{\infty} H(t) dt \]

where \(H\) is the per caput dose-equivalent rate in a given organ for a specified population. Finally, the quantity effective dose equivalent is defined as

\[ H_{e} = \sum_{T} w_{T} H_{T} \]

where \(H_{T}\) is the dose equivalent to tissue \(T\) and \(w_{T}\) is a weighting factor which relates the stochastic risk from irradiation of tissue \(T\) to the total risk resulting from a uniform irradiation of the whole body. With these definitions the effective dose equivalent commitment can be written

\[ H_{c,e} = \int_{0}^{\infty} H_{e}(t) dt \]

The above definitions have been revised and new recommendations (which are not yet in official use) are given in ICRP (1990).

2 DeGeer et al. report an accumulated deposition of 66 mCi/km\(^2\) (2.44 kBq/m\(^2\)) in Stockholm in the years 1953-1970. Mattsson & Lidén found a deposition of 64.2 mCi/km\(^2\) (2.38 kBq/m\(^2\)) for a moss carpet in southern Sweden in the years 1969-1973. Since the area of Sweden is 449,964 km\(^2\) this yields about 141,401 Bq.
reached the Swedish east coast as early as the night between April 26 and 27 (Erlandsson et al., 1987; Kjelle, 1991).

The deposition in Sweden of $^{137}\text{Cs}$ from the Chernobyl accident was, according to Snihs (1996), of the same order of magnitude as the fallout $^{137}\text{Cs}$ from nuclear weapons testing in the atmosphere: 4.25 PBq and 1.25 PBq, respectively. The nuclear weapons fallout amounted to about 3 kBq/m$^2$ and was rather evenly distributed whereas the Chernobyl deposition, however, showed large variations between about zero and 200 kBq/m$^2$ (Edvarson, 1991).

Table 1. Deposition at Ljungbyhed of $^{137}\text{Cs}$ from nuclear weapons fallout 1962–1981

<table>
<thead>
<tr>
<th>Year</th>
<th>Yearly deposition (Bq/m$^2$)</th>
<th>Remaining 1994 (Bq/m$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1962</td>
<td>357</td>
<td>170</td>
</tr>
<tr>
<td>1963</td>
<td>905</td>
<td>442</td>
</tr>
<tr>
<td>1964</td>
<td>433</td>
<td>217</td>
</tr>
<tr>
<td>1965</td>
<td>233</td>
<td>119</td>
</tr>
<tr>
<td>1966</td>
<td>105</td>
<td>55.0</td>
</tr>
<tr>
<td>1967</td>
<td>56.6</td>
<td>30.3</td>
</tr>
<tr>
<td>1968</td>
<td>62.2</td>
<td>34.1</td>
</tr>
<tr>
<td>1969</td>
<td>35.2</td>
<td>19.8</td>
</tr>
<tr>
<td>1970</td>
<td>57.7</td>
<td>33.1</td>
</tr>
<tr>
<td>1971</td>
<td>61.0</td>
<td>35.9</td>
</tr>
<tr>
<td>1972</td>
<td>14.8</td>
<td>8.90</td>
</tr>
<tr>
<td>1973</td>
<td>4.44</td>
<td>2.73</td>
</tr>
<tr>
<td>1974</td>
<td>25.9</td>
<td>16.3</td>
</tr>
<tr>
<td>1975</td>
<td>11.5</td>
<td>7.41</td>
</tr>
<tr>
<td>1976</td>
<td>3.81</td>
<td>2.51</td>
</tr>
<tr>
<td>1977</td>
<td>17.3</td>
<td>11.7</td>
</tr>
<tr>
<td>1978</td>
<td>18.8</td>
<td>13.0</td>
</tr>
<tr>
<td>1979</td>
<td>6.85</td>
<td>4.84</td>
</tr>
<tr>
<td>1980</td>
<td>3.98</td>
<td>2.88</td>
</tr>
<tr>
<td>1981</td>
<td>11.2</td>
<td>8.29</td>
</tr>
<tr>
<td>$\Sigma$ 62-81</td>
<td>2424</td>
<td>1235</td>
</tr>
</tbody>
</table>

In this work the deposition over the southern part of Sweden was studied from the time of arrival and further on with respect to deposition characteristics and the behaviour of the deposited material in the ground. The initial phase of the deposition was studied by air sampling and

$^{31}$ The deviation from the value 1.1 PBq previously calculated is probably due to rounding errors.
deposition measurements in the city of Lund, where the plume arrived on April 28.

The first part of the debris deposited by gravitational settling (dry deposition) and the major part did not deposit until May 7 due to a rainfall. In the debris 21 isotopes of Chernobyl origin were identified in various amounts.
2. SAMPLING METHODS

All samples, regardless of method of sampling and preparation as described below, were analysed with gamma-spectrometric methods using a Ge(Li) detector of efficiency 18% and resolution 1.9 keV (FWHM) at 1332 keV shielded against ambient background radiation with 10-15 cm of lead. The detector system was efficiency calibrated using the method described by (Bjurman et al., 1987) in which a liquid calibration source is mixed with a matrix material of the same density as the environmental samples. The matrix material containing the radionuclides from the calibration source may then be transferred to any desired geometry. With this method it is possible to achieve a calibration for all kinds of samples, without the need for additional density or geometrical corrections.

2.1 Air

Ground level air sampling was made using a centrifugal pump with a capacity of 270 m$^3$/h. The air was drawn through vertically mounted 0.50 × 0.50 m$^2$ glass-fibre filters with a linear flow velocity of 0.3 m/s. The difference in flow rate between the beginning and the end of a sampling period was measured and found to be less than 10%. This was the same type of equipment as was used by the National Defence Research Institute at seven of their sampling sites (Vintersved and DeGeer, 1982; Vintersved et al., 1987). The length of the sampling periods varied between 1 and 4 weeks and the volume of the air drawn through the filter was calculated from the sampling time. At the end of each sampling period the filters were packed into 180 ml plastic jars and analysed with gamma-spectroscopy. The activity of the different radionuclides present was calculated in Bq/m$^3$ of air.

2.2 Precipitation

Rain water samples for activity measurements were collected using a precipitation collector consisting of two 1 × 2 m$^2$ aluminium sheets slightly horizontally angled. A slightly inclined gutter was placed in the angle between the sheets to collect the rain water in a plastic container at the end of the gutter. The rain water was first filtered through 2-4 glass fibre filters to remove coarse particles and then passed through 30 ml Dowex 1-X8
Sampling methods

anion ion-exchange resins and 30 ml Dowex 50 WX8 cation ion-exchange resins at a flow rate of about 0.5 l/h. The ion-exchange resins and the filter papers were then dried at 60° C, packed together into 60 ml plastic jars and analysed with gamma-spectroscopy. The activity of the different radionuclides present was calculated in Bq/dm³ of water. The benefit of this method of concentrating the radioactive elements in ion-exchange resins is obvious in that it is possible to concentrate tens of litres of water into a much smaller volume, e.g. 60-100 ml.

In the initial rains during the passage of the plume from the Chernobyl accident, the activity concentration was high enough to make it possible to measure the activity in the rain water directly in 180 ml plastic jars. The effectiveness compared with measurements on the water directly has been shown to be about 85%.

Rain water was collected during all periods of precipitation in the first three months after the Chernobyl accident and due to the relatively large area of the precipitation collector (4 m²) it was possible to determine the activity in such small precipitation showers as 0.5 mm.

In addition to rain water sampled on the precipitation collector at our department in Lund a large network of sampling stations (Ellesson, 1993) was used to determine the amount of precipitation at different places in Skåne.

2.3 Deposition

2.3.1 Dry deposition
Gravitational settling of particles dispersed in the air is usually denoted as dry deposition. The amount of dry deposited radionuclides was determined by wiping the aluminium sheets of the abovementioned precipitation collector with paper towels moistened with an organic solvent. The paper towels were then packed into 180 ml plastic jars and analysed with gamma-spectroscopy. The activity of the different radionuclides present in the dry deposited material was calculated in Bq/m².

The first wiping of the precipitation collector was performed on April 28 and the subsequent measurement made at 17:30 on the same day revealed the composition of the Chernobyl debris at Lund. The measurement on the same sample was repeated 24 hours later to include the half-lives in the
identification of some radionuclides. The precipitation collector was then wiped daily until May 7, when the first rain began to fall.

From the dry deposited activity, the rate of sampling and the air activity concentration it is possible to calculate the dry deposition velocity, \( v_d \), defined as (Chamberlain and Chadwick, 1953)

\[
v_d = \frac{\text{Dry deposition flux} \left[ \text{Bq/m}^2 \text{s} \right]}{\text{Air activity concentration} \left[ \text{Bq/m}^3 \right]} \tag{2.1}
\]

The deposition velocity, usually given in cm/s, was calculated for various radionuclides from April 28 until May 7.

### 2.3.2 Wet deposition

In contrast to dry deposition, wet deposited material is removed from the air by precipitation scavenging. The amount of wet deposition was determined by measuring the activity concentration in the rain water and since the area of the precipitation collector is well known the activity concentration in rain water can be recalculated as deposited activity per area unit.

The rate of scavenging can be expressed either as the wet deposition velocity, \( v_w \), or the washout ratio, \( w \), defined as

\[
v_w = \frac{\text{Wet deposition flux} \left[ \text{Bq/m}^2 \text{s} \right]}{\text{Air activity concentration} \left[ \text{Bq/m}^3 \right]} \tag{2.2}
\]

and

\[
w = \frac{\text{Precipitation activity concentration} \left[ \text{Bq/m}^3 \right]}{\text{Air activity concentration} \left[ \text{Bq/m}^3 \right]} \tag{2.3}
\]

both the wet deposition velocity, given in cm/s, and the washout ratio were calculated for a number of radionuclides in the initial phase of the deposition.
2.4 Sewage sludge
Measurements of various radionuclides in sewage sludge has been shown to be a sensitive indicator for radionuclides spread in an urban environment. It has been demonstrated that the activity concentration of caesium in the sludge is 3 to 4 orders of magnitude greater than in outgoing water from the plant (Erlandsson et al., 1989). The fact that most municipal sewage systems transport run-off water after precipitation as well as waste water from e.g. households and hospitals makes them a good instrument for monitoring the releases from these sources of radionuclides. An important parameter in determining how long it will take for an urban environment to get rid of deposited radionuclides through the sewage system is the time required for the deposited material to enter the plant from different run-off areas such as trees, streets etc. and the mean residence time in the plant. The mean residence time of the sludge in the plant varies with the type of the plant from 2-3 days up to 3-4 weeks.

Sewage sludge has been analysed once a week since 1983 in Lund and after the Chernobyl accident the rate of sampling was raised to twice a week.

Samples of sewage sludge were dried at 105°C, homogenised, packed in 180 ml tubs and analysed with gamma-spectroscopy.

2.5 Soil

2.5.1 Compilation of soil-sampling methods
Soil can be sampled and analysed for many reasons, e.g. to survey an area for fresh fallout or to determine the moisture content and the vertical distribution of radioactivity in the ground for subsequent use in in situ measurements. Three main methods to take soil samples can be identified:

- The template method
  The usual application of this method is to scrape or shovel off layer after layer of soil within a chosen area, which could be defined by some sort of rigid frame, in some cases pressed down into the soil to a certain depth (Papastefanou et al., 1988; Simopoulos, 1989; HASL-300, 1990; Bonazzola et al., 1993; Antonopoulos-Domis et al., 1995). The size of the area usually varies between a quarter of a square metre up to one square metre.
The advantage of this method is that a reproducible area is sampled but there is also a possibility that the particular area chosen for measurement is not representative for the measurement site. It may contain an unrepresentatively high or low amount of $^{137}\text{Cs}$ compared with other areas in the close vicinity, e.g. a "hot" or a "cold" spot. The existence of these irregularities especially after the Chernobyl accident is well documented (Finck, 1992) and could lead to serious misinterpretations of the results. It is also difficult to ensure that the thickness of the different layers is identical over the entire area. This is perhaps not so crucial when surveying the surface soil to establish the magnitude of deposition but could lead to difficulties when the vertical distribution should be determined.

Soil sampling with this method is also very time consuming and results in a large amount of soil, from which aliquots are usually taken. In addition there is a tendency for the sampled area to decrease with increasing depth (when no frame is used). The method could under certain conditions be suitable for determining the vertical distribution of radionuclides at a well defined location. One such condition is when the presence of rocks in the ground will make the core method (see below) difficult to use. The horizontal distribution, however, over a larger area remains unknown unless several areas are sampled at a sampling site.

- The trench method
Another method to take soil samples from different depth is the trench method (Karlberg, 1987; HASL-300, 1990; Kagan & Kadatsky, 1996) in which a trench is dug to a depth of about one half to one metre. The samples are then taken horizontally from the walls of the trench. The main advantage of this method is the possibility of taking samples at a precisely defined depth and with a standardised corer all samples could be made to represent a well defined area. With a proper technique there is also little risk for cross contamination. The main drawback is the hard and lengthy work or the need for machinery to dig the trench.

One variety of this method is the use of a monolite for sampling (McGee et al., 1995; Velasco et al., 1993). Either the whole monolite or subsamples from its sides are sampled.
• Soil sampling with bore cores

The third method of examining the activity concentration in the soil over an area is to take samples in the form of bore cores arranged in different geometries, or at random, over the area of interest (Arnalds et al., 1989; Miller et al., 1990; Blagoeva & Zikovsky, 1995; Rosén et al., 1996). If the purpose of the sampling is to determine the depth distribution of radionuclides the bore cores could then be divided into horizontal sections and analysed. The bore cores could otherwise be scanned lengthwise with a collimated detector (Finck, 1992). If several samples are taken over a sampling site and analysed separately, this method gives a good estimate of both the horizontal as well as the vertical distribution of the activity concentration at the measuring site.

The analysis of the soil samples could be very time-consuming if the number of bore cores is large and the cores in addition are divided into several different layers. The number of samples could be reduced if the soil at a given depth from different cores is mixed and analysed together as general samples (HASL-300, 1990; III). Although this certainly will shorten the time of analysis, most of the information about the horizontal distribution will be lost. An average vertical distribution can, however, still be determined.

One obvious problem with the core method is the risk for cross contamination and compression of the soil. The cross contamination occurs when soil from upper, more contaminated, layers are pressed down to greater depths by the bore. Compression of the soil could be expected when the soil is pressed out of the bore.

The soil sampling in this work was done entirely by means of the bore core method.

2.5.2 Criteria for the selection of sampling sites

The selection of sites for soil sampling is based on a number of criteria, depending on the expanse of the region to examine and on the purpose of the sampling. On a larger scale the choice of sampling sites could be based on criterias such as:

• Expected distribution of the deposition. This could be estimated from precipitation data in the case of wet deposited radionuclides (VI).
• Occurrence of meteorological stations in the vicinity.
• Availability.
• Occurrence of previously examined reference areas or farms picked out for deposition measurements on crops or milk.
• Previously performed or planned field gamma spectroscopy in the vicinity.

The chosen site should preferably fulfil the following requirements:

• Flat and open terrain without shadowing vegetation or buildings.
• Representative for the actual site with respect to vegetation and soil composition.
• No run-off areas or swampy sites.
• Not too abundant ground vegetation.
• A possibility to take samples to the desired depth.
• Not containing "hot spots".

2.5.3 Sampling equipment
All soil samples collected to be used in this work were taken with an 8 cm diameter metal tube on horizontal ground. The bottom edge of the tube was sharpened to minimise the disturbance of the soil and to make it easy to push down into the ground. The tube was generally pushed into the ground to a depth of 15-20 cm.

After being pushed down to the desired depth the tube is pulled up and the soil core can be pressed out of the tube with the aid of a built-in piston (Figure 1). The cores are then cut into slices of 2-3 cm thickness at the end of the tube and stored in plastic bags.

When the soil is moist it is often possible to press it out in the form of a small cylinder while the tube is in a horizontal position. When the soil is very dry or sandy there is, however, a possibility that most of the bore core remains in the hole, or falls out of the tube when it is pulled up. This can be avoided by digging around the tube and inserting a cover over the bottom of the tube while it is still in the ground. When the tube is removed, the soil can then be pressed out and scraped off when the tube is turned upside down.
Sampling methods

In the first measurements where this equipment was used (III), nine bore cores were evenly spaced in a quadratic grid with 30 cm between the cores on a 1 m² grass area and soil samples were collected down to a depth of 12 cm. The bore cores were then cut into 2 cm thick slices.

In order to minimise the number of bore cores and hence the sampling and analysis time another method of positioning the bore cores was utilised in the following measurements (III). The nine bore cores were collected in a triangular pattern shown in Figure 2 and divided into 2 or 3 cm layers.

The soil samples from the three triangles at each site were all analysed separately at first to determine the horizontal variation of the deposition on different length scales. In the subsequent measurements (IV, V) only one triangle, consisting of three bore cores, was taken at each site. In addition, the three samples from a given depth were mixed together and analysed as one general sample.
2.5.4 Preparation of samples

The soil samples were dried at 60°C for about 24 h, weighed and homogenised in a domestic food processor. All sampled material except larger rocks and pebbles was included in the homogenised samples. The homogenised samples were put into 60 ml or 180 ml plastic tubs and analysed with a Ge(Li) detector (efficiency 18% and resolution 1.9 keV (FWHM) at 1332 keV) for $^{134}$Cs, $^{137}$Cs and $^{40}$K. The detector was surrounded by a 10 cm thick lead shield and was energy calibrated with standard reference sources. The efficiency calibration was performed using the same detector-sample geometry by mixing a calibration source of known strength with the soil (Bjurman et al., 1987).

Due to the rather low activity concentration in the samples a counting time between 10 h and 24 h was necessary to obtain an acceptable statistical accuracy (0.5%-7% for the 662 keV photopeak from the decay of $^{137}$Cs).
3. CHARACTERISATION OF THE FALLOUT

3.1 Horizontal variation

The fallout in Sweden from the Chernobyl accident showed large variations in the deposition pattern (SGAB, 1986), which could be attributed in part to the passage and the composition of the plume but mainly to the differences in precipitation. The deposition caused by the nuclear weapons fallout, on the other hand, shows a much more homogeneous pattern (Snihs, 1996).

The deposition of $^{137}\text{Cs}$ at a particular site is therefore subject to spatial variability which could be divided into (Owens & Walling, 1996a):

- random spatial variability
- systematic spatial variability
- sampling variability
- measurement precision

The first source, the random part of the variability could be attributed to small-scale variations in the composition of the soil and vegetation cover together with the topography on the small scale. The small-scale topography affects for instance the behaviour of the precipitation with the formation of dry spots and puddles. The systematic variability depends on large-scale variations such as varying amounts of precipitation and the distribution of soil and vegetation type. The sampling variability reflects the possibility that the sampled area differ somewhat between samples and also that the amount of soil or the sampling depth is not accurately determined. The measurements precision, finally, is due to the uncertainties involved in the preparation of the samples and subsequent analysis, including counting uncertainties. In comparing several samples at two different sites Owens & Walling (1996a) found that the dominating part of the variability within each site was the random spatial variability. A comparison of several measurements of the $^{137}\text{Cs}$ inventory at a number of sites and soil horizons (McGee et al., 1995; Owens & Walling, 1996a) showed a coefficient of variation between 5.1% and 64%.
3.1.1 Small scale variation

The variability of $^{137}\text{Cs}$ on the small scale is highly affected by local topography and also by the presence of a snow cover at the time of deposition. Even over comparatively small areas such as a field, the variability could be of importance. Finck (1992) found that the variability, expressed as the standard deviation of 18 soil samples, at an airfield amounted to about 80%. This high variability was attributed to a snow cover at the time of deposition which subsequently melted and caused streams of run-off water. In situ measurements at the same site showed a standard deviation of only 10% for five measuring points. This discrepancy is concluded to be due to variations on the metre scale.

In this work (III) the small-scale variability of $^{137}\text{Cs}$ is studied in different ways. Nine soil samples are taken with the metal tube described above and spread out in the triangular geometry according to Figure 2. First, three cores are taken with a distance of 30 to 40 cm from each other. Then, three such triangles of cores are taken at a distance of about one metre apart, also in a triangular pattern. The soil samples are sliced and each slice is analysed separately. Since the surface area of each bore core is accurately known it is possible to calculate the deposition, $S$, in Bq/m$^2$ of each core.

The variability on the metre-scale can be estimated by the standard deviations within each small triangle of samples, $s_{\text{within}}$, based on the deposition values, $S_j$, for the three different bore cores $j = 1, 2, 3$ found within each small triangle as shown in Figure 3. It may also be described by calculating the mean values of the deposition, $S_i$, within each triangle $i = A, B, C$ and study the standard deviation, $s_{\text{between}}$, of these mean values (also shown in Figure 3). In this way we get an estimate of the variability between the small triangles. Finally, the above figures can be compared with the overall standard deviation, including all nine cores at a given site.

To determine the variation on the scale of approximately ten metres, a fourth triangle, consisting of three bore cores, was placed at a distance of about 17 metres from the other nine cores at one of the sampling sites. To include the mean value of the deposition found for these samples gives further information of the variability on the scale of one to ten metres.
A couple of bore core slices were also divided into four parts which were all analysed separately. The parts were the outermost 5 mm of the slice, centreplug with a radius of 19 mm, a quarter of the resulting ring and remainder. This division was done to give the centre and circumference subsample approximately the same mass. The variability on the centimetre scale may now be identified with the standard deviation between the parts of a single bore core slice.

It can be shown (Sokal & Rohlf, 1981) that the number of samples needed to give a "true" difference between mean values of different groups of measurements is given by

\[ n \geq 2 \left( \frac{\sigma}{\delta} \right)^2 \left[ t_{\alpha \left( v \right)} + t_{2\left[1-p\left( v \right)\right]} \right]^2 \]  

(3.1)

where

- \( n \) = number of samples for the mean of each group
- \( \sigma \) = standard deviation of the samples in each group
- \( \delta \) = the smallest difference between means one wishes to detect
\( v \) = degrees of freedom (\( n \) samples in each of \( a \) groups gives 
\( v = a(n-1) \))

\( \alpha \) = significance level

\( p \) = probability that a difference as small as \( \delta \) will be found to be significant

\( t_{\alpha, v} \) and \( t_{2(1-p), v} \) are found from a two-tailed table of \( t \)-distribution with \( v \) degrees of freedom and probabilities of \( \alpha \) and \( 2(1-p) \), respectively.

To solve this equation for \( n \) an iterative calculation is required, but the equation may also be used to find the smallest detectable difference \( \delta \) between means with a given number of samples and a given standard deviation. It was found (III) that the triangular sampling pattern with three bore cores gave a standard deviation between the cores of approximately 20%. This can be used to estimate the minimum difference between the mean of these three samples and another three samples at another site (or within the same sampling site) that could be detected.

If we accept a probability of 80% of finding a difference between the means at the 95% confidence level (i.e. in 5% of the comparisons the difference is due to chance and the two samples taken from distributions with the same mean and hence there is no true difference) distribution we thus have the following parameters:

\[
\begin{align*}
\alpha &= 0.05 \\
\sigma &= 20\% \\
n &= 3 \\
v &= 2(3-1) = 4 \\
t_{0.025(4)} &= 2.776 \quad 4) \\
t_{0.40(4)} &= 1.005 \quad 5)
\end{align*}
\]

which yields \( \delta = 62\% \).

With the sampling method described in (III) it is therefore possible to say that the deposition at two sites is different if the difference between the mean values of each site is greater than 62%. Considering the short time needed to sample and analyse the soil with this method it can still be justified in an emergency situation where the main task is to get a general view of the amount of deposition for radiation protection purposes.

4) Given in one-tailed tables as \( t_{0.025(4)} \)

5) Given in one-tailed tables as \( t_{0.40(4)} \)
3.1.2 Large scale variation

In paper IV the spatial variability on the larger scale (orders of kilometres) was estimated by soil sampling at sixteen sites in the city of Lund and also at sixteen sites in the county of Skåne. At each sampling site three bore cores were sampled in the triangular geometry shown as one of the triangles A, B or C in Figure 2 and analysed as general samples. The variability was then expressed as the standard deviation of the measured total deposition of $^{137}$Cs (Bq/m$^2$) for Lund and Skåne respectively.

Several investigations have been done to correlate the amount of deposited $^{137}$Cs to the amount of precipitation. For the deposition of nuclear weapons fallout in Montana, USA, Arnalds et al. (1989) found the relationship

$$C(\text{Bq/m}^2) = 1480 + 0.37 \cdot P(\text{mm})$$

(3.2)

where $C$ is the total soil inventory of $^{137}$Cs and $P$ is the mean annual precipitation. The correlation coefficient was 0.62.

For the inventory of bomb fallout $^{137}$Cs in superficial soil in Canada Blagoeva & Zikovsky (1995) found that

$$C(\text{Bq/m}^2) = 532 + 1.97 \cdot P(\text{mm})$$

(3.3)

with the correlation coefficient 0.84 and where the term 532 Bq/m$^2$ is identified as the amount of dry deposition.

In Ireland Mitchell et al. (1990) showed that the integrated $^{137}$Cs from nuclear weapons fallout could be related to the mean annual rainfall by

$$C(\text{Bq/m}^2) = (631 \pm 128) + (2.67 \pm 0.11)P(\text{mm})$$

(3.4)

with a correlation coefficient of 0.993.

From deposition and precipitation data at Ljungbyhed (DeGeer et al., 1978) and in Lund (I) it was possible to calculate the activity concentration in the rain water, which also can be expressed as deposition per unit precipitation$^6$, both for the Chernobyl fallout and for the fallout from nuclear weapons tests (VI). The activity concentration in the precipitation

$^6$ 1 Bq dm$^{-1}$ 1 Bq m$^{-2}$ mm$^{-1}$
can be expected to be proportional to the air activity concentration and if the spatial variability of the air activity concentration is low over a certain region\textsuperscript{7} the activity concentration in rain water can be considered homogeneous in this region. Precipitation data from a large number of rain gauge stations was then used to calculate the deposition at the locations of these stations, based on quarterly precipitation, and deposition maps were constructed by interpolation between station values. This calculated deposition was thereafter compared with the deposition measured by soil sampling (IV), \textit{in situ} measurements (Arntsing \textit{et al.}, 1991) and aerial surveys (SGAB, 1986).

\section*{3.2 Vertical variation}

\subsection*{3.2.1 Exponential depth distribution}

According to ICRU (1994) the largest source of uncertainty when performing field gamma spectrometry is the assumptions about the depth distribution of the radionuclides in the soil which have to be made. Therefore it is of great importance to find a proper way to measure and describe this distribution.

The static, time independent, vertical distribution of radionuclides in the ground is often described as an exponential function of depth, which may generally be written as (Mattsson, 1975)

\begin{equation}
    c(z) = \alpha \cdot e^{-\beta z^p}
\end{equation}

Here, \(c(z)\) is the activity concentration (Bq/kg) of the radionuclide at the depth \(z\); \(\alpha\), \(\beta\), and \(p\) are experimentally determined parameters. The parameter \(\alpha\) can be identified as the activity concentration of the radionuclide as \(z\) approaches zero and, when \(p\) equals 1, the reciprocal of parameter \(\beta\) is referred to as the relaxation length (Beck, 1966)\textsuperscript{8}. It is assumed that \(\alpha\), \(\beta\), \(z\) and \(p\) are all positive numbers.

\textsuperscript{7} This was found both in connection with the nuclear weapons tests (Bernström, 1969) and the Chernobyl accident (VI), although the regions with similar air activity concentration can be expected to be smaller in the latter case due to the finite extent of the plume.

\textsuperscript{8} A \(p\)-value of 0.75 has also been used to describe the depth distribution of \(^{137}\text{Cs}\) in both a lichen (Mattsson 1975) and a moss carpet (Mattsson & Liden, 1975).
The main effect of $p$ on the depth distribution is to determine the shape of the depth distribution curve. When $p$ equals 1 the curve is simply a straight line in a semi-logarithmic plot. When $p>1$ the curve becomes convex, thereby indicating a distribution which changes more slowly with depth in the upper part of the soil compared to the deeper part of the soil; when $p<1$, the curve is concave and the main change in the depth distribution occurs in the upper part of the soil, compared to the deeper part (Figure 4). A distribution with $p<1$ can thus be assumed to be caused by a process which is more efficient in distributing activity in the ground.

![Figure 4](image-url)

**Figure 4.** Effect of the parameter $p$ on the shape of the depth distribution.

The effect of the parameter $\beta$ is to raise or lower the curve, hereby altering the shape of the curve and the gradient of the decreasing activity concentration with depth (Figure 5). The main appearance of the curve, i.e. convex or concave, is however not affected by $\beta$. This is easily shown by studying the second derivative of the logarithm of Equation 3.5.

\[
\frac{d^2}{dz^2} \left( \ln c(z) \right) = \frac{d^2}{dz^2} \left( \ln \alpha - |\beta| \cdot z^p \right) = \\
= |\beta| \cdot p \cdot (1 - p) \cdot z^{p-2}
\]
The sign of Equation 3.6, which determines the shape of the curve, only depends on whether $p$ is smaller or greater than unity since $z$, $p$ and $\beta$ are all supposed to be greater than zero.

Figure 5. Effect of the parameter $\beta$ on the shape of the depth distribution for some different values of $p$. 
3.2.2 Different ways to measure depth

The depth may be measured either as linear depth $z$ (often expressed in cm) or as mass depth $z \rho$ (kg/m$^2$), where the concept of mass depth is introduced because of the density dependence of the attenuation of radiation. If the depth is given as mass depth, assuming $p = 1$ for reasons of simplicity, Equation 3.5 can be written in the form (HASL-300, 1990; Finck, 1992)

$$c(z) = \alpha \cdot e^{-\left(\frac{\beta}{\rho}\right) \cdot \rho \cdot z}$$  \hspace{1cm} (3.7)

where $\rho$ is the in situ soil density. Also $\rho / \beta$ is often called relaxation length or mass relaxation length (Miller et al., 1990) and in this case measured in kg/m$^2$. The density is often assumed to be constant down to the maximum sampling depth and the mass depth is found by simply multiplying the linear depth with the soil density.

The soil density is, however, seldom constant with depth. As can be seen from (III, Table 1) the densities show the greatest variability in the first 0-6 cm, due to the composition of the soil material, and then stabilises and remain fairly constant at the deeper parts of the ground. If a soil core includes litter and newly decomposed matter together with mineral soil the density may vary considerably in the core and Equation 3.7 could then be written

$$c(z) = \alpha \cdot \exp \left[ -\frac{\beta}{\rho(z)} \int_0^z \rho(z') \, dz' \right] = \alpha \cdot e^{-\left(\frac{\beta}{\rho(z)}\right) \cdot z}$$  \hspace{1cm} (3.8)

where $\rho(z)$ and $\rho(z')$ is the density at the depth $z$ and $z'$, respectively.$^{91}$ The mass depth $\zeta$ down to depth $z$ in a bore core may be found by slicing the core and approximating $\rho(z)$ with a constant density

$^{91}$ ICRU Report 53 (ICRU, 1994) suggests that the activity distribution with depth, $A_m(\zeta)$, can be written as

$$A_m(z) = A_{m,0} \cdot \exp(-z / \beta)$$

where $\beta$ is called relaxation mass per unit area (earlier denoted by $\rho \alpha$, which in this work is denoted by $\rho / \beta$ because this is the expression used in paper III - V) and $\zeta$ is the mass per unit area down to depth $z$ (in this work referred to as mass depth), defined by $\zeta = \int_0^z \rho(z') \, dz'$.

where $\rho(z)$ is the soil density at depth $z$:  

24
where $\rho_i$ is the soil density in layer $i$, $\Delta z_i$ is the thickness of layer $i$ and $n$ is the number of layers. If the parameter $\beta/\rho(z)$ is assumed to characterise the distribution down to the maximum sampling depth it must have a constant value and cannot depend on the depth $z$. Actually, there are no physical reasons why this parameter should not vary with depth, but the distribution is often found by a fit to experimental points and the main interest is to try to describe the entire distribution down to a given depth.

Because the density varies with depth, the only way to determine a unique parameter $\beta$, and hence relaxation length, for a given distribution is to assume a constant mean density, $\bar{\rho}$, and the depth distribution takes the form

$$c(z) = \alpha \cdot e^{-\beta/\bar{\rho} \cdot z} \quad (3.10)$$

If the relaxation length is expressed as mass relaxation length (kg/m$^2$) the mean density is implicit in the fitting procedure.

The use of a mean density does, however, not account for the variation of density with depth. An alternative expression, which does not explicitly depend on the density is

$$c(z') = \alpha \cdot e^{-\phi \cdot z'} \quad (3.11)$$

where $z'$ is the mass depth, expressed in kg/m$^2$, determined from field samples and obtained by weighing each sample (e.g. each slice from a sectioned soil core). This means that $z'$ can be considered to be the experimentally determined counterpart to $\zeta$ from Equation 3.10. With this method it is not possible to determine a relaxation length (1/$\phi$) in centimetres, but the mass relaxation length, expressed in kg/m$^2$, could be identified with 1/$\beta'$.

---

10) The mass depth $z'$ for layer $k$ could be calculated as

$$z' = \sum_{i=1}^{k-1} \rho_i \Delta z_i + \frac{1}{2} \rho_k \Delta z_k$$
The parameter $\beta^*$ may, however, in some soils with fairly constant density be transformed into $\beta$ by comparing Equations 3.10 and 3.11, which yields $\beta = \beta^* \rho$. It should be noted that $\beta^*$ is a parameter resulting from a fit of Equation 3.11 to experimental points and while $\beta^*$ to some extent takes the density variations into account, $\beta$ however, does not.

If, however, the above expression (Eqn. 3.11) is used with the depth given as linear depth it may be written as

$$c(z) = \alpha \cdot e^{-\beta z}$$ \hspace{1cm} (3.12)

The parameter $\beta$ is now determined from a fit of measured points and the relaxation length $(1/\beta)$ can be found. It was one of the objectives of paper III to see if the method of sampling described gives comparable results regardless of the depth units used.

The changing properties of downward migration with depth, which could be described by a depth dependent $\beta^*$ or $\beta^*$-parameter could also be contained in the parameter $\rho$ as defined in Equation 3.5. In the general case the equations for the downward transport would then read

$$c(z) = \alpha \cdot e^{-\beta z}$$ \hspace{1cm} (3.13)

with $z$ expressed as linear depth and

$$c(z') = \alpha \cdot e^{-\beta z'}$$ \hspace{1cm} (3.14)

with $z$ expressed as mass depth.

The drawback with the use of $\rho$ to characterise the distribution of radionuclides in the ground is that the unit in which $\beta$ and $\beta^*$ are measured are dependent on the value of $\rho$ (e.g. $\rho = 2$ gives $[\beta] = \text{cm}^2$ or $\text{m}^2$ whence $\rho = 0.75$ gives $[\beta] = \text{cm}^{0.75}$ or $\text{m}^{0.75}$). This makes a direct comparison of the relaxation length or the mass relaxation length at different sites impossible if the composition of the soil and hence the transport properties differ greatly, i.e. the parameter $\rho$ differs from site to site.
3.2.3 Determination of total activity

The total accumulated activity, expressed either as Bq or Bq/m$^2$, can be determined from soil sampling in two ways: adding the activity found for each layer in a soil core or integrating the analytical expression found from a fit of the sample. In the addition the activity in each layer for a given bore core is calculated from the activity concentration and the weight of the soil in that layer. It is assumed that the sampling depth is sufficient to collect all of the activity present in the soil. The activity per unit area (Bq/m$^2$) can easily be found when the cross section area of the bore core is known.

The method of integration makes use of the analytical relationship found by the fit to the measured activity concentration. The total activity per unit area (Bq/m$^2$) when the depth is expressed as mass depth, $z'$, is given by

$$S = \int_0^{z_{\text{max}}} \alpha \cdot e^{-\beta z'} \, dz' = \frac{\alpha}{\beta} \left(1 - e^{-\beta z_{\text{max}}} \right) \tag{3.15}$$

where $z'_{\text{max}}$ could be the depth of the bore core or the depth below which no activity is supposed to be contained.\textsuperscript{11}

With the depth expressed as linear depth it is necessary to assume a mean density, $\rho_m$, for the soil core and the expression is

$$S = \rho_m \int_0^{z_{\text{max}}} \alpha \cdot e^{-\beta z} \, dz = \rho_m \frac{\alpha}{\beta} \left(1 - e^{-\beta z_{\text{max}}} \right) \tag{3.16}$$

and $z_{\text{max}}$ is chosen analogous to $z'_{\text{max}}$ in Equation 3.15.\textsuperscript{12} Since the density could be highly variable in the soil, especially in the upper few centimetres compared to the deeper mineral soil, it is advisable to perform the integration in two steps: for example from 0 cm to 2 cm and from 2 cm to $z_{\text{max}}$, using appropriate densities for these two parts of the soil.

3.2.4 Time-dependent exponential depth distribution

It can be of interest to try to give some physical meaning to the above mentioned experimentally determined parameters and to identify them with physical factors which govern the transport in the ground. If that can be

\textsuperscript{11} [S] = Bq/m$^2$ since [$\alpha$] = Bq/kg and [$\beta$] = m$^{-1}$/kg

\textsuperscript{12} [S] = Bq/m$^2$ since [$\alpha$] = Bq/kg, [$\beta$] = m$^{-1}$ and [$\rho_m$] = kg/m$^3$
done the development of the parameters with time could also be studied and the simple static equation (Eqn. 3.5) may be transformed into a time-dependent equation for the depth distribution after a deposition event.

It is assumed, in a first approximation that the transport is purely diffusional. In this case the concentration $c(z,t)$ at a given depth and time can be found by solving the diffusion equation (Fick’s second law of diffusion)

$$\frac{\partial^2 c(z,t)}{\partial t^2} = D \frac{\partial^2 c(z,t)}{\partial z^2}$$

which for one-dimensional diffusion in one direction from a plane source has the solution (Crank, 1975)

$$c(z,t) = \frac{M}{\sqrt{\pi D t}} e^{-z^2/4Dt}$$

Here $M$ is the amount of substance (or the activity) per unit area deposited at time $t = 0$ in the plane $z = 0$ and $D$ is the (constant) diffusion coefficient (often measured in cm$^2$/s or in this context the more appropriate cm$^2$/y). The form of this function is shown in Figure 6.

A comparison between Equations 3.5 and 3.18 shows that under the assumption that the diffusion coefficient, $D$, is constant with depth and time the parameters may be identified according to

$$p = 2$$

$$\alpha = \frac{M}{\sqrt{\pi D t}}$$

$$\beta = \frac{1}{4Dt}$$
Figure 6. The form of Equation 3.18 at different times, \( t \) for \( M = D = 1 \).

The parameters \( \alpha \) and \( \beta \) are seen to decrease with time whereas the parameter \( p \) is unaffected. The time-dependence of the \( \alpha \)- and \( \beta \)-parameter may be summarised as

\[
\alpha \propto t^{1/2} \quad \quad (3.22)
\]

\[
\beta \propto t^1 \quad \quad (3.23)
\]

and the depth distribution may be written

\[
c(z, t) = \frac{\beta}{\sqrt{t}} \cdot \frac{1}{\alpha^p} \cdot e^{-\frac{z^p}{\alpha^p} \cdot t} \quad \quad (3.24)
\]

The diffusion coefficient does not necessarily have to be constant with depth and time but can vary with the concentration of the diffusing substance as well as with the degree of sorption in the material it passes and
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with the soil density. In addition other processes than diffusion could influence the migration.

If the vertical transport is regarded as a diffusion-convection process Equation 3.17 transforms into (Konshin, 1992a)

$$\frac{\partial^2 c(z,t)}{\partial z^2} = D \frac{\partial^2 c(z,t)}{\partial z^2} - V \frac{\partial c(z,t)}{\partial z}$$

(3.25)

where $D$ is the diffusion coefficient (describing the random dispersion process) and $V$ is a measure of the vertical directional transport by mass flow, or convection (cm/y). A solution to this equation when the activity $c_0$ per unit area is distributed at depth $z = 0$ at time $t = 0$ gives

$$\frac{c}{c_0} = \frac{1}{2\sqrt{\piDt}} \exp \left\{ -\frac{(z-Vt)^2}{4Dt} \right\}$$

(3.26)

where $c$ is the activity concentration at depth $z$ at time $t$ (Konshin, 1992a).

To account for the rapidly decaying tail of Equation 3.26 and the subsequent underestimation of the activity concentration at greater depths (Konshin, 1992b) suggests a modification by introducing a lognormal distribution

$$\frac{c}{c_0} = \frac{1}{2\sqrt{\pi Dt}} \exp \left\{ -\frac{\left( \ln z - \ln \frac{1}{Vt} \right)^2}{4 Dt} \right\}$$

(3.27)

Since no sorption parameter is included in Equation 3.25 the solutions (Eqn. 3.26 and Eqn. 3.27) describes the migration of a non-sorbing radionuclide which is not the case for caesium (Hsu & Chang, 1994; Antonopoulos-Domis et al. 1997). According to Konshin (1992b) this could be accounted for by letting the parameters $D$ and $V$ decrease with time so that

$$D = \frac{0.49}{\gamma_D}; V = \frac{0.40}{\gamma_V}$$

(3.28)
A third approach is taken by Antonopoulos-Domis et al. (1995) who proposes a transport equation of the form

$$\frac{\partial}{\partial t} C(z,t) = D \frac{\partial^2 C(z,t)}{\partial z^2} - k C(z,t) - \lambda C(z,t)$$  \hspace{1cm} \text{(3.29)}

where $C(z,t)$ denotes the activity concentration of free caesium, available for migration and the term $kC$ is interpreted as the net transfer from free to bound state. The activity concentration of total caesium, $T(z,t)$, is then given as the sum of free, $C(z,t)$, and bound, $B(z,t)$ caesium. The equation also takes into account explicitly the physical decay of the radionuclide (by the term $\lambda C$). This may, however, also be included in the previous expressions either by calculating a relative activity or by using decay-corrected activity concentration in the calculations.

If the activity at $t = 0$ is present in an infinitesimal layer at the soil surface the solution is again of the form

$$C(z,t) = \frac{C_0}{2\sqrt{\pi Dt}} \exp\left(-\frac{z^2}{4Dt}\right) \exp\left[-(k + \lambda)t\right]$$  \hspace{1cm} \text{(3.30)}

and decreases with $z^2$ as the rest of the models discussed so far. However, Antonopoulos-Domis et al. (1995) have found that if the bound caesium is taken into account the total caesium is found to decrease with $z$ below a certain depth. This is explained by introducing a convective term $(-\partial vC/\partial z)$, where $v$ is the velocity of the water carrying free caesium) in Equation 3.29, describing the transport of caesium with water into the ground. This term was, however, shown to be significant only at an initial period with a large amount of free caesium in relation to bound caesium.

The behaviour of caesium in the ground, discussed above from the physical models point of view, supports the simple exponential model (Eqn. 3.24). The time-correction of the parameters according to this model was used ($\text{V}$) to estimate the present depth distribution from nuclear weapons fallout $^{137}\text{Cs}$ using the depth distributions from the fallout since the sixties.
3.2.5 Modelling the present depth distribution of bomb fallout $^{137}$Cs

The ratio of the $^{134}$Cs to the $^{137}$Cs activity concentration for the deposition from the Chernobyl accident (26th of April 1986) was 0.52-0.58 but the first part of the cloud that reached Sweden showed a ratio of 0.63 (Erlandsson, Asking & Swietlicki, 1987). The variation in this ratio was found to depend on different release and transport conditions (Devell, 1991). For the deposition in Lund a ratio of 0.53-0.56 have been used to calculate the activity concentration of $^{137}$Cs originating from the Chernobyl accident ("new" $^{137}$Cs) by measuring the activity concentration of $^{134}$Cs in the soil samples. Thereafter the contribution from nuclear weapons fallout ("old" $^{137}$Cs) could be calculated.

3.2.5.1 Transformation of depth distribution by an exponential model

Equation 3.24 could be tested by repeated measurements of the depth distribution at the same site (V). As an example, consider Figure 7 which shows the measured activity concentration of new $^{137}$Cs at two sites in Lund, Lund 7 and Lund 11, measured in 1993 together with a fit according to Equation 3.13 with $p = 2$. Measurements have also been made in 1991 at the same sites and the aim is to transform the 1991 depth distribution into the 1993 depth distribution.

By letting $t = 1$ and $p = 2$ and fitting Equation 3.24 to the 1991 data the parameters $\alpha_0$ and $\beta_0$ can be found. These parameters are then transformed to 1993 ($t = 2$ years) according to

$$\alpha = \frac{\alpha_0}{t}$$  \hspace{1cm} (3.31)

and

$$\beta = \frac{\beta_0}{t}$$  \hspace{1cm} (3.32)

This transformed depth distribution is also shown in Figure 7.

The agreement is enhanced if the diffusivity is allowed to decrease with time due to the sorption of radiocaesium in the soil matrix. According to
Equation 3.28 the diffusivity may be assumed to vary as $t^{-0.78}$ which suggests that $\alpha$ has a time-dependence of the form

$$\alpha = \frac{\alpha_0}{t^{0.11}}$$

(3.33)

![Figure 7. Measured $^{137}$Cs of Chernobyl origin at two sites in Lund together with model predictions.](image)
The varying diffusivity would, however, according to Equation 3.21 also affect \( \beta \) which would reduce the good agreement in shape of the depth distribution curves. The effect of the varying diffusivity on \( \beta \) may, however, eventually be neutralised if also convection is taken into account since \( V \) only affects \( \beta \) and not \( \alpha \) according to Equation 3.26. This model can be useful in predicting the migration of radiocaesium in the ground on a time-scale of two years (V).

The time-scale could then be extended by incorporating also nuclear weapons fallout \(^{137}\text{Cs}\) in the model. The deposition of fallout \(^{137}\text{Cs}\) in Lund could be based on the measured deposition at Ljungbyhed, situated about 60 km from Lund (deGeer et al., 1978). If the deposition is correlated with precipitation and expressed as activity per mm precipitation a calculation of the deposition in Lund, based on precipitation (in Lund), can be made (see discussion in section 3.1.2). It was found that from April 1962 to December 1966 the deposition from the nuclear weapons tests amounted to 927 Bq/m\(^2\) (corrected for decay to 1994) which is 85\% of the total deposition measured up to 1977. It is therefore assumed here that this deposition of nuclear weapons fallout \(^{137}\text{Cs}\) could be assigned to a single occasion in 1964. This deposition thus occurred 29 years before the last measurements in 1993.

The parameters for the initial distribution in 1964 can be found under the assumption of an exponential depth distribution (Eqn. 3.24 with \( t = 1 \) and \( \rho = 2 \)) in which case \( \alpha_0 \) is given by the expression

\[
S = \rho_m \int_0^{0.02} \alpha_0 \cdot e^{-\beta_0 \cdot z^2} \, dz + \rho_{m2} \int_{0.02}^{0.2} \alpha_0 \cdot e^{-\beta_0 \cdot z^2} \, dz
\]

(3.34)

where \( S \) is the activity per unit area (927 Bq/m\(^2\)).

The rather large differences in soil density at different depths could to some extent be accounted for by assuming two different densities: \( \rho_{m1} \) and \( \rho_{m2} \) which are the mean densities (kg/m\(^3\)) in the soil layers 0-2 cm and 2-20 cm, respectively.

The parameter \( \beta_0 \), needed to solve Equation 3.34, could be found using the \( \beta \)-parameter from a fit to new \(^{137}\text{Cs}\), measured in 1991 or 1993, which is transformed back to 1986 by

\[
\beta_0 = \beta \cdot 1
\]

(3.35)
This parameter, which reflects the distribution of new Chernobyl $^{137}\text{Cs}$ at the time of deposition (1986), is then assumed to reflect also the initial distribution of the nuclear weapons fallout (1964). It is therefore assumed that the deposition at a particular site has the same initial distribution, regardless of source and year of deposition.

The next step is to find the depth distribution of nuclear weapons caesium today, using the previously determined parameters $\alpha_0$ and $\beta_0$, assumed to be applicable for the fresh fallout in 1964. The parameters are transformed to take into account the redistribution of caesium between 1964 and 1994 (30 years) according to Equations 3.31 and 3.32. The resulting distribution (Eqn. 3.24 with $p = 2$) is shown in Figure 8 which also shows the measured old $^{137}\text{Cs}$ at the two sites, corrected for decay to 1994 for comparison.

Figure 8. Measured $^{137}\text{Cs}$ from nuclear weapons fallout at two sites in Lund together with model predictions.
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It can be seen that the calculated distribution underestimates the measured distribution, although the slope of the distribution is not dissimilar at any of the sites. The correction for varying diffusivity (Eqn. 3.33) does enhance the agreement but not to the same extent as in the case of two years of migration (V).

3.2.5.2 Time dependence of activity in successive layers

The measurements of new $^{137}$Cs at different times, e.g. 1991 and 1993 (V), could be used to calculate the relative amount of activity in different layers 5 and 7 years after the deposition in 1986. Likewise could the old $^{137}$Cs, measured in 1993, give the relative amount of activity in each layer 29 years after the deposition of nuclear weapons fallout under the assumption that the deposition occurred as a single event in 1964. It is further assumed that at the time of the deposition of nuclear weapons fallout (0.5 years used here to avoid singularities at time zero) 100% of the activity is contained in the first layer (here 0-2 cm) and that no activity is present in the deeper part of the soil.

In paper V the calculations are carried out regarding Lund 7 and Lund 11 as one single site and a mean value of the relative amount of activity in the first layer may then be found at times 0.5, 5, 7 and 29 years after a deposition event (utilising both nuclear weapons fallout and Chernobyl $^{137}$Cs). For subsequent layers the relative amount of activity can be found at times 5, 7 and 29 years after deposition (Chernobyl and nuclear weapons fallout, respectively)

The relative amount of activity, $A_r$, as a function of time for each layer in the Lund measurements is shown in Figure 9 and the experimental values fit well to a power function of the form

$$A_r = \xi \cdot t^\delta$$  \hspace{1cm} (3.36)

where $t$ is time since deposition (years); $\xi$ and $\delta$ are parameters. It can be seen from Figure 9 that even the point representing the nuclear weapons fallout $^{137}$Cs, i.e. a deposition that occurred 29 years ago, fits nicely to the curve.
As shown in the figure, the activity in the upper two layers (0-4 cm) decreases with time and the activity in the next layer, 4-6 cm, is practically constant. In the deeper parts of the soil the activity is seen to increase with time.
This change in time-behaviour with depth could be quantified by the depth dependence of the parameters $\xi$ and $\delta$ (Figure 10) given by

$$\xi = 73.4 \cdot e^{0.058 \cdot z^{0.8}}$$

and

$$\delta = 0.14 \cdot z - 0.698$$

where $z$ is the depth in cm.

Figure 10. Depth dependence of the parameters $\xi$ and $\delta$. 
The relative amount of activity in the soil at the Lund site can thus be summarised as

\[
A_r = 73.4 \cdot e^{-0.058 z} \cdot \frac{1^{137}}{t^{0.14}} \cdot z^{-0.698}
\]  

(3.39)

which is shown in Figure 11. The figure could be used to determine the depth of maximum activity \(A_r (z_{max})\) at different times after a deposition. For 30 years of migration it can be seen from Figure 11 that the maximum relative activity is located 5.7 cm below the surface and it is thus possible to assign a mean downward speed of 0.2 cm/y for the nuclear weapons fallout \(^{137}\)Cs. This is also in accordance with results previously reported by Denk & Felsmann (1989) and Owens et al. (1996b).

Figure 11. Relative amount of activity of \(^{137}\)Cs as a function of depth and time.

The depth of maximum \(A_r\) could also be calculated from the equation

\[
\frac{dA_r(z_{max})}{dz} = 0
\]

(3.40)

and the downward migration expressed by the speed with which the peak of the activity concentration is moving, \(dz_{max}/dt\). The solution of Equation 3.40 is generally not given by an analytical expression for all values of \(z\) and \(t\) but
a numerical solution, however, gives real solutions when \( t \) lies within the approximate interval \( 1.63 \leq t \leq 38.5 \text{ y} \). An approximate expression for \( z_{\text{max}} \) is then given by

\[
z_{\text{max}} = \left( \frac{0.14}{0.058 \cdot 1.88} \cdot \ln t \right)^{1/88}
\] (3.41)

and is shown in Figure 12. This figure also shows the migration rate of \( z_{\text{max}} \) given by \( \frac{dz_{\text{max}}}{dt} \). As expected the migration rate rapidly decreases during the first few years (by a factor of four in five years) and thereafter the radiocaesium is relatively fixed in the soil.

![Figure 12. Depth of maximum activity at different times after deposition and corresponding migration rate.](image)

### 3.2.5.3 Compartment modelling

Another approach in describing the time-dependence of activity in different layers is compartment modelling of varying complexity. In a simple compartment model as given in Equation 3.41 the compartments could consist of layers of a given thickness (Velasco \textit{et al}., 1993) or of the different soil horizons (Rühm \textit{et al}., 1996).

If the activity in layer \( i \) is designated as \( A_i \) and no upward movement is assumed the transport is given by
\[ \frac{dA_1}{dt} = -\lambda_1 \cdot A_1 \]
\[ \frac{dA_2}{dt} = \lambda_1 \cdot A_1 - \lambda_2 \cdot A_2 \]
\[ \frac{dA_3}{dt} = \lambda_2 \cdot A_2 - \lambda_3 \cdot A_3 \]
\[ \frac{dA_4}{dt} = \lambda_3 \cdot A_3 - \lambda_4 \cdot A_4 \]
\[ \frac{dA_5}{dt} = \lambda_4 \cdot A_4 - \lambda_5 \cdot A_5 \]  

where \( \lambda_i \) is the time constant describing the rate of transfer from compartment \( i \) to compartment \( i+1 \). The notation in Equation 3.42 assumes decay corrected values of the activity.

In paper V the relative amount of activity in each layer was used in a fit to the solutions to the above equations. Although the activity of old \(^{137}\text{Cs} \) was reduced by a factor of two to account for the smaller amount of deposition from nuclear weapons fallout compared to the amount of deposition from the Chernobyl accident (V) the value does not fit into the curves.
4. **SUMMARY OF PAPERS**

The papers included in this thesis have been chosen so that a general trend of the work can be outlined. The Chernobyl accident in 1986 marks the starting point and the composition and amount of radioactive debris from this accident, brought to Sweden by the weather systems, is the scope of **Paper I**. **Paper II** deals with the resulting deposition in an urban area through measurements on sewage sludge and in **Paper III** the deposition on soil is discussed. In this paper a method of soil sampling was developed which was used to study the vertical and horizontal distribution of $^{137}$Cs in the ground. This method was then used in an extended study of the activity in soil in Lund and Skåne, divided into nuclear weapons fallout and fallout from the Chernobyl accident (**Paper IV**). The vertical distribution was further discussed in **Paper V** which also concerns the temporal variation of the depth distribution. Finally, **Paper VI** connects the deposition with precipitation and presents a method to predict the deposition over larger areas from measurements of precipitation.

4.1 **Paper I:**

*Relation between the air activity and the deposition of Chernobyl debris*

*B. Erlandsson & M. Isaksson*

*Environment International. 14, pp. 165-175, 1988.*

The first measurement at Lund made at 17:30 on April 28 revealed 21 different radioisotopes present in the dry deposition. The activities are in good resemblance with the reported release of radionuclides from the damaged reactor (USSR, 1986) and the discrepancies (mainly $^{132}$Te, $^{131}$I, $^{141}$Ce and $^{237}$Np) could be explained by changes in the composition of the plume during transport to Lund.

For the first few days the same decay pattern are shown for the studied radioisotopes ($^{95}$Zr, $^{95}$Nb, $^{99}$Mo, $^{96}$Re, $^{103}$Ru, $^{132}$Te, $^{131}$I, $^{134}$, $^{136}$, $^{137}$Cs, $^{140}$Ba, $^{144}$Ce and $^{237}$Np). This decrease is interpreted as insufficient wiping of the precipitation collector and is not assumed to reflect the actual time variation. During May 2, 3 and 4, however, a clear increase with a
maximum on May 4 is seen, followed by a decrease for all but the volatile element $^{131}$I. This is in accordance with the reported behaviour of the emission of radionuclides from the reactor (USSR, 1986).

The first rain that fell in the area (0.5 mm between 22:00 and 22:23 on May 7) coincided both with the increased emission from the reactor on May 3, 4 and 5 and a change in weather conditions which brought the plume to southern Sweden on May 7 and 8. This resulted in an 10-50 fold increase in accumulated deposition.

The initial dry deposition velocities (April 28) show large variations, ranging from $0.012 \pm 0.001$ cm/s for $^{134}$Cs to $0.22 \pm 0.1$ cm/s for $^{144}$Ce. According to experiments (Ahmed, 1979) and models (Sehmel, 1980; Slinn et al., 1978) of dry deposition velocity as a function of particle diameter this implies that the Cs particles were smaller in size than Ce particles and is in good agreement with other observations of particles from the first Chernobyl plume that reached Sweden (Erlandsson et al., 1987; Appelgren, 1986), Denmark (Roed, 1987) and Switzerland (Jost et al., 1986).

Furthermore, a relation was found between the air concentration, $A$, of $^{103}$Ru and $^{137}$Cs and the activity concentration in precipitation.

### 4.2 Paper II:

**The urban contamination after the Chernobyl accident studied in sewage water and sludge from Lund and Gothenburg**

*B. Erlandsson, M. Isaksson, S. Mattsson & B. Hemdal*


The fate of radioactive fallout in an urban environment was studied by sampling sewage sludge and outgoing water from the Källby plant in Lund. In addition, the contribution from citizens was determined using data on whole body measurements made at the Department of Radiation Physics in Malmö. A comparison was also made between the Källby plant and the Rya plant in Göteborg in a collaboration with the Department of Radiation Physics in Göteborg.
It was found that the response of the two plants was considerably different; the Rya plant reacted much faster on the initial fallout than the Källby plant, a phenomenon which is due to construction differences between the two plants. In both Lund and Göteborg the dominating radionuclides measured in sewage sludge shortly after the deposition was $^{103}\text{Ru}$ and $^{106}\text{Ru}$, also $^{239}\text{Np}$ was measured at both places. One year after the accident only $^{137}\text{Cs}$, and to some extent $^{106}\text{Ru}$, were found to be of importance.

Using a known ratio between the activity in sludge and the activity in sludge plus water (Erlandsson et al., 1989) it was possible to calculate the total outgoing activity of $^{137}\text{Cs}$ from the Källby plant in Lund. Because the sewage system in Lund is a duplicate system it was also possible to calculate the amount of the activity which came from the tap water and from the citizens of Lund, respectively. It was found that in the winter 86/87 about 10% of the total outgoing activity of $^{137}\text{Cs}$ from the plant was coming from the citizens.

### 4.3 Paper III:

**Experimental determination of the vertical and horizontal distribution of $^{137}\text{Cs}$ in the ground**

M. Isaksson & B. Erlandsson


The aim of this work was to determine the distribution, horizontally as well as vertically, of $^{137}\text{Cs}$ in the ground by the method of bore core sampling. The use of this method is usually connected with three main problems:

(i) cross contamination.

(ii) minimising the number of bore cores necessary to obtain an accurate determination of the activity concentration.

(iii) the need for an experience-based inaccuracy on the measured activities when the number of bore cores are minimised.

Soil sampling with the bore core method was found to be a good tool for accurate determination of the vertical as well as the horizontal variation of radionuclides in the ground. It was shown that the problems usually associated with sampling by bore cores, *i.e.* compression of the soil and
cross contamination, can be avoided and was negligible with the sampling equipment used.

It was also found that it is possible to shorten the overall time required to obtain an accurate determination of the deposition in two ways. Firstly, the sampling time is shortened by taking only three cores at each site. and secondly, the analysis of these samples can be performed by combining them to one general sample for each depth.

The results show that the horizontal variation of the deposition is of the same order of magnitude regardless of the size of the investigated area, e.g. over a single bore core or over a whole field. This variability is generally around 15-20%.

The depth was determined either as linear depth (cm), or mass depth (kg/m²). The mass depth was calculated for each of the different layers in each bore core to account for the density variation with depth. It was found that, with this sampling method, the depth dependence is equally well described with the depth defined as linear depth or mass depth. The results also suggest that the depth distribution of ¹³⁷Cs in the ground is often not well described by an exponential function of depth. Instead, a better fit is achieved if the depth in the exponential is raised to a power of site-specific magnitude.

A comparison between the mass relaxation lengths found in this work and reported values from a large number of locations, summarised in ICRU (1994) shows a good agreement. The values reported by ICRU (1994) ranges from 0.1 to 15 g/cm² for Chernobyl fallout caesium and from 2 to 19 g/cm² for fallout from nuclear weapons tests. The corresponding values in this work are 2-5 g/cm² (Chernobyl) and 3-11 g/cm² (nuclear weapons fallout).

The measured depth distributions also showed a slow transport and a strong retention of ¹³⁷Cs, thus implying a practical lowest depth of the bore core of 12 to 15 cm.
4.4 Paper IV:

*Investigation of the distribution of* $^{137}$Cs *from fallout in the soils of the city of Lund and the province Skåne in Sweden*

*M. Isaksson & B. Erlandsson*

*Journal of Environmental Radioactivity, xx, pp. x-x, 1997.*

The aim of this work was to use the soil sampling method described in III in a survey of the distribution of $^{137}$Cs in the ground in Skåne and Lund.

Soil samples from sixteen different sites in Lund and sixteen locations in Skåne were collected in a grid pattern for the determination of the vertical as well as the horizontal variation in the deposition of $^{137}$Cs in the ground.

The vertical distribution of $^{137}$Cs was described by variations of a general exponential function by fitting the function to the field data consisting of total $^{137}$Cs (Chernobyl and nuclear weapons fallout Cs). In some of the samples the actual distribution was quite well described by an exponential decrease with depth, but in many cases the agreement was poor. The agreement was enhanced when the depth in the exponential was raised to a power specific to the particular site.

The mass relaxation lengths varied between 2.8 and 7.7 g/cm$^2$ in Lund and between 1.7 and 19.2 g/cm$^2$ in Skåne, in agreement with results reported by ICRU (1994). Also the (linear) relaxation lengths, 2.7-6.3 cm in Lund and 1.7-17.5 cm in Skåne, are in agreement with results previously reported in the literature (Korune et al., 1991; Yesin & Çakir, 1989).

By comparing fitting parameters it was also found that, with this sampling method, the depth distribution is equally well described with the depth given as linear depth (cm) or mass depth (kg/m$^2$).

A comparison between accumulated activity (Bq/m$^3$) from measured soil samples and the total activity found by integrating the fitted analytical expressions for the actual depth distribution showed that in many cases the deviation was within 20%. The investigation of the depth distribution also showed a slow transport and a strong retention of $^{137}$Cs, thus confirming the result found in III that a bore core need be no longer than 12 - 15 cm.

The horizontal variability of total $^{137}$Cs (both nuclear weapons fallout and deposition of Chernobyl origin) found in these measurements was about 20% in the city of Lund and 40% in Skåne. This figures agree with previously reported results (III). When the contribution from old nuclear
weapons fallout relative to caesium of Chernobyl origin was analysed the nuclear weapons fallout was found to account for between 0% and 83% of the total $^{137}\text{Cs}$ activity. This spread reflects the difference in deposition mechanisms of the two sources and also to some extent the differences in sampling depth between the sampling sites.

4.5 Paper V:

*A field study of the vertical migration of $^{137}\text{Cs}$ in the ground*

M. Isaksson & B. Erlandsson

*Submitted to Journal of Environmental Radioactivity.*

The aim of this work was to investigate and describe the changes in depth distribution with time for nuclear weapons fallout $^{137}\text{Cs}$ as well as $^{137}\text{Cs}$ of Chernobyl origin. The study was made on soil samples collected at two sites in the town of Lund on two different occasions at each site.

The accumulated total deposition down to 12 cm depth at a given site showed a coefficient of variation of 9% between measurements in 1991 and 1993. This can be considered as quite small since it was earlier shown (III) that the variation in deposition on the metre scale is of the order of 15-20%.

The deposition of $^{137}\text{Cs}$ caused by nuclear weapons tests, corrected for decay to April 26th 1986, is about 1300 Bq/m$^2$ in Lund. This value is based on the actual amount of precipitation at Lund and on the deposited activity of $^{137}\text{Cs}$ measured at Ljungbyhed (deGeer et al., 1978) about 60 km from Lund. The agreement with the accumulated deposition of $^{137}\text{Cs}$ from nuclear weapons fallout calculated from measurements of $^{134}\text{Cs}$ in this study, with mean values of about 1400 Bq/m$^2$, is rather good.

A comparison between the exponential depth distribution and diffusion theory suggests a model for redistribution of caesium with time. This model was applied to the measurements of new $^{137}\text{Cs}$ at Lund to predict the behaviour of $^{137}\text{Cs}$ in two years of migration and shows a good agreement with actual measurements. The agreement is, however, not that good when the model is used to predict the present depth distribution of nuclear weapons fallout $^{137}\text{Cs}$ from the 1960's. It may be concluded that this method
could be useful in predicting the fate of radiocaesium in the ground, at least on the time-scale of two years.

Using measurements of both Chernobyl and nuclear weapons fallout $^{137}$Cs it was shown that the relative activity could be described by

$$ A_r = 73.4 \cdot e^{-0.058 \cdot 1.88} \cdot t^{0.14 \cdot z - 0.698} $$

where $A_r$ is the relative amount of activity, $z$ is the depth in cm and $t$ is the time in years since deposition. For 30 years of migration the depth of maximum relative activity is found 5.7 cm below the surface and it is thus possible to assign a mean downward speed of 0.2 cm/y for the nuclear weapons fallout $^{137}$Cs.

In addition, a simple compartment analysis in which each layer constitutes a compartment was used for the relative activity in different layers and gave a time constant of 0.25 y$^{-1}$ for the first layer (0-2 cm), a result also found by Rühm et al. (1996) for $^{134}$Cs in the L-horizon in a coniferous forest.

4.6 Paper VI:

Calculation of the deposition of Cs-137 from the nuclear bomb tests and from the Chernobyl accident over southern Sweden based on the precipitation

M. Isaksson, B. Erlandsson & M.-L. Linderson
Manuscript.

The aim of this work was to calculate the spatial distribution of the deposition of $^{137}$Cs from the Chernobyl accident and from the nuclear weapons tests. This calculated deposition is then compared with measurements on soil samples, with in situ measurements and with aerial surveys. Due to existing early deposition measurements from the 60ies, detailed measurements of deposition on the first days after the Chernobyl accident and registrations from a dense network of precipitation collectors the province Skåne in the southern part of Sweden was well suited for this kind of investigation.
Measurements of the deposition from nuclear weapons fallout and precipitation measurements at Ljungbyhed in the central part of Skåne (Lindblom, 1966; Bernström, 1969; 1974; DeGeer et al., 1978, FOA 41) was used to calculate the deposition per unit precipitation. Using precipitation measurements from 113 raingauge stations (Ellesson, 1993) it was then possible to calculate the deposition at each of these stations. By interpolating between the stations a deposition map of nuclear weapons fallout could be constructed. Detailed measurements of the wet-deposition and precipitation on the first days after the Chernobyl accident (I) made it possible to construct similar maps for the first day of wet-deposition and for the remainder of 1986.

Comparisons with soil sampling (IV) shows good agreement. The mean of the ratios for the calculated to measured deposition at Skåne is $0.96 \pm 0.20$, $0.96 \pm 0.21$ and $1.07 \pm 0.38$ for the total, pre-, and post-Chernobyl deposition, respectively. The corresponding means in a comparison between calculated deposition and deposition measured in situ (Arntsing et al., 1991) are $2.26 \pm 0.48$, $3.88 \pm 0.93$ and $1.46 \pm 0.49$ which also shows a good agreement considering that the pre-Chernobyl deposition has a vertical distribution in the ground and therefore is underestimated by the in situ measurements. The general trend in the distribution of the deposition as shown in the deposition maps are also found in aerial surveys made in 1986 (SGAB, 1986).

The results show that it is possible to get a good knowledge of the deposition from measurements of the deposition at a few stations and of the precipitation from the network of sampling stations.
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Chapter 6

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