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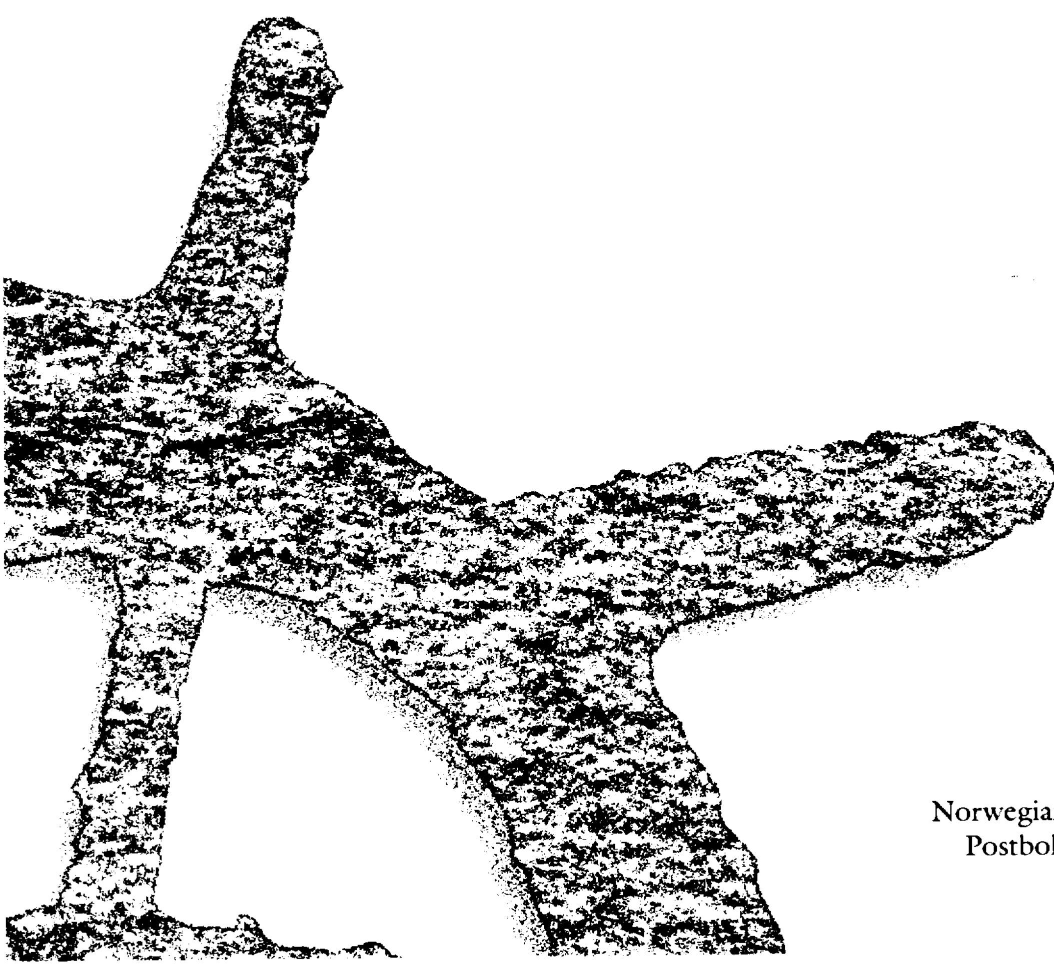

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Mehli H. Radiocaesium in grazing sheep - A statistical analysis of variability, survey methodology and long term behaviour. StrålevernRapport 1996:2. Østerås: Norwegian Radiation Protection Authority, 1996.

Key words:
Caesium. Sheep. Long term behaviour. Fungi. Sampling. Variations.

Abstract:
Since 1987 sheep grazing in the areas of Norway that received Chernobyl-fallout have been monitored before slaughter. These monitoring data formed the basis for development of a model describing the long term behaviour of radiocaesium in unimproved pasture showing that in years with good mushroom abundance 70-80 % of the radiocaesium concentration in sheep is due to fungi consumption. A study of sampling strategy and variability of radiocaesium concentration within flocks was also performed.

Referanse:
Mehli H. Overvåkning av radiocesium i sau - En statistisk analyse av variasjon, utvalgsmetodikk og varighet. StrålevernRapport 1996:2. Østerås: Statens strålevern, 1996. Språk: engelsk.

Emneord:
Cesium. Sau. Varighet. Sopp. Utvalg. Variasjon.

Resymé:
Sau som beiter i områder som mottok Tsjernobyl-nedfall i 1986 har siden 1987 blitt målt før slakting. Disse måledataene ble brukt til å utvikle en modell for å beskrive varigheten av radiocesium forurensningen. Denne viser at i gode soppår er inntak av sopp årsak til 70-80 % av radiocesiumnivået i sau. Utvalgsstrategi og variasjon i radiocesiumkonsentrasjoner innen flokker er også studert.

Head of project: Lavrans Skuterud, NRPA.
Approved:

Erik-Anders Westerlund, director, Environmental Protection Department.

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Preface

This report is a Diploma Thesis in biophysics at the Department of Physics and Mathematics, the Norwegian Institute of Technology (NTH), Trondheim. The project was carried out at the Norwegian Radiation Protection Authority and was finalised in February 1996

Thanks to all who has given input to this project. Special thanks to:

- Prof. Henning Omre, Institute of Mathematics, NTH, who helped with the statistical part
- Johan Teige, District veterinary at Hedmark and Oppland, who provided the relevant data on which this thesis is based
- Lavrans Skuterud, NRPA, who supervised the project in an excellent manner

Østerås, May 1996

Hanne Mehli

Summary

Norway received considerable deposition of radioactive fallout after the Chernobyl accident in 1986. In mountain areas frequently used as pasture for animals, serious long term effects on agriculture have resulted from the persistently high radiocaesium concentrations in animals and animal products. For mutton the acceptable limit for human consumption was set at 600 Bq/kg. In 1987 a method for live monitoring of animals was introduced, which made it possible to determine the radiocaesium level in animals before slaughter, and introduce countermeasures if necessary. The LORAKON project conduct comprehensive surveys of radiocaesium in animals, and since 1987 live monitoring of animals grazing in the most contaminated areas has been carried out routinely. The monitoring is based on *grazing units*, i.e. sheep from several farmers grazing in the same area.

The counties of Hedmark and Oppland were among the most contaminated parts of the country, and animals from grazing units in these areas have therefore been studied thoroughly. In some grazing units live monitoring has taken place every year since 1987, and can therefore be used as the basis for a study of long term behaviour of radiocaesium in sheep. This study should be performed in areas where no countermeasures have been implemented. After a thorough search through the data records the two grazing units of Fåset and Fonnåsfjellet, in approximately the same area, were found to meet the requirements. In both areas live monitoring of sheep has taken place each year since 1987, and no countermeasures had been implemented.

In Vuludalen and Baklia, whole herds of respectively 180 and 212 animals were monitored in 1987. These observations were the basis for a study of sampling strategy and variability of radiocaesium concentration within a herd.

Analysis of variance showed a significant difference in radiocaesium concentration in ewes and lambs, with lambs having approximately 20-40 % higher levels. The approximation to normal distribution was improved when the data sets were split in two groups of ewes and lambs, and the variance in each of the subgroups decreased compared with the variance in the whole herd.

The estimated average radiocaesium concentration in a grazing unit is based on monitoring a sample of 8-10 animals. A study of the variability indicated that more than twice as many animals should be monitored if an estimate of the mean radiocaesium concentration should be obtained with uncertainty less than 10 %.

Observations from the grazing units of Fåset and Fonnåsfjellet in Østerdalen formed the basis for the study of long term behaviour of radiocaesium. The following factors considered for determining the radiocaesium concentration in sheep, RC :

- Contamination level in 1986, C
- Time after the Chernobyl accident t_c
- Effective elimination rate λ_{eff} [y^{-1}], for radiocaesium in the environments, explaining how fast the amount of available radiocaesium decreases. Effective half life T_{eff}, is calculated from λ_{eff} : $T_{eff} = \ln 2/\lambda_{eff}$ [y]
- Amount of fungal fruit bodies available as feed for the animals, F (relative unit)
- Time on pasture with fungi available as feed for the animals t_p [d]
- Biological half life for radiocaesium in sheep, estimated to 0.03 [d^{-1}]

The suggested model built up of these factors was:

$$RC\ (t_c, t_p, F) = C \cdot e^{-\lambda_{eff} t_c} + B \cdot F \cdot (1 - e^{-0.03 \cdot t_p})$$

where B estimated the importance of fungi as radiocaesium source and F was given as the relative unit «low», «medium» and «large» amounts of fruit bodies.

Regression analysis of the observations from Fåset and Fonnåsfjellet gave the following estimated model:

$$RC\,(t_c, t_p, F) = 370 \cdot e^{-0.09 \cdot t_c} + 190 \cdot F \cdot (1 - e^{-0.03 \cdot t_p})$$

with $R^2 = 0.60$, a relative importance between the levels of fungi abundance of 1:4:9 and time for appearance of fruit bodies set to 15.08.

When the influence of ingested fungi is separated, the total radiocaesium concentration in sheep was found to have decreased with an effective half life of 3.9 ± 0.8 years. For ^{137}Cs alone the estimated effective half life for ^{137}Cs was 7.8 ± 2.6 years. This was significantly longer than the estimated effective half life based on July observations from Grøtting, a grazing unit close to Fåset and Fonnåsfjellet, which was 2.4 ± 0.4 years.

The estimated contribution from ingested fungi to the total radiocaesium concentration in sheep in the studied areas was found to be high, up to 75-80 % in 1988 and 1991, during which years fungi were most abundant.

Contents

1. Introduction

During the 1950's and 1960's the atmospheric nuclear weapons tests provided world wide fallout of radioactive materials. In Norway the fallout from the nuclear weapon tests was homogenous distributed, with an average for the country of about 5-8 kBq/m^2 [1]. In April - May 1986 the nuclear power plant accident at Chernobyl in the Ukraine caused large scale releases of radionuclides. The radiocaesium fallout in Norway has been estimated to represent 6 % of the total release of ^{134}Cs and ^{137}Cs from the Chernobyl accident, and the average deposition was estimated to 11 kBq/m^2 [2]. The Chernobyl fallout in Norway was, in contrast to the fallout from nuclear weapons tests, inhomogenously distributed with larger deposition in areas with precipitation at the time the Chernobyl cloud passed.

Experience from the fallout from nuclear weapons tests in the 1960's had shown that ^{137}Cs deposition could be managed on cultivated land, where it can be moved down the soil profile with ploughing and making it less available for root uptake in plants. This was not the case for the environments involving sheep and reindeer herding. On natural pasture in forest and mountain areas the radiocaesium move slower down the soil profile, and is therefore available for uptake in plants for a longer period.

Consequently, the Chernobyl accident gave specially large consequences for agriculture involving animals grazing on unimproved pastures. For mutton, a trade limit for radiocaesium of 600 Bq/kg was set in order to reduce the collective dose to the Norwegian population. After the slaughter of reindeer and sheep in autumn 1986 meat to the value of 100 mill. NOK was condemned because of higher radiocaesium concentrations [3]. Monitoring meat samples was resource consuming, and due to this, and to avoid condemnation of meat, a method for live monitoring of animals was developed. Determining the radiocaesium concentration in live animals made it possible to implement required countermeasures (like feeding the animals uncontaminated feed) to reduce the radiocaesium concentration in animals before slaughter.

The LORAKON system has since 1987 been responsible for annual live monitoring of sheep. Median radiocaesium concentrations in grazing units (sheep from several farmers grazing in the same area) has been estimated from monitoring a selection of animals, and this estimated value determined the requirements for countermeasures in the whole grazing unit.

In 1988 an unexpected high radiocaesium concentration level in sheep was observed, and this coincided with the appearance of large amounts of fungal fruit bodies this year [4]. Different species of fungi has been shown to accumulate high levels of radiocaesium [5], and the observations in 1988 proved that :

1) Intake of fruit bodies affects the radiocaesium concentration in grazing animals
2) The Chernobyl fallout could have more long term consequences than was initially assumed

Estimating the radiocaesium concentration level within a flock of sheep or reindeer from year to year is difficult for several reasons :

1) The animals graze freely over large forest and mountain areas
2) Within these areas there are large local variations in deposition of radiocaesium, soil types and vegetation species, resulting in heterogeneous contamination levels in the pasture.
3) Feed intake is hard to estimate.
4) Intake of fruit bodies affects the radiocaesium concentration in animals, but the time for appearance as well as amount of fruit bodies vary from year to year.

Intake of radiocaesium through fruit bodies will contribute to the total radiocaesium concentration in animals, but the importance of fungi compared to vegetation is not known. Temperature and precipitation are known to be important parameters determining fruit body appearance, but there is currently no model available to study appearance and amounts of fruit bodies. It has therefore been difficult to estimate the importance of ingested fungi for the radiocaesium concentration in animals. In order to study the different factors (with emphasis on fruit bodies) influencing radiocaesium concentration in animals grazing on unimproved pasture, and also the variability within animals in a grazing unit, recorded data from live monitoring of animals grazing in some of the most contaminated areas in Norway were studied.

2. Ionising radiation and radioactivity

2.1 Ionising radiation

Ionising radiation is generally characterised by its ability to excite and ionise atoms of matter with which it interact. To be able to excite or ionise, radiation has to carry kinetic or quantum energy in excess of the energy needed to cause a valence electron to escape an atom, which is in the order 4-25 eV.

There are different kinds of ionising radiation:

Gamma-rays are electromagnetic radiation emitted from a nucleus or in annihilation reactions between matter and antimatter. The frequency ν or wavelength λ gives the energy of the gamma-ray:

$$E_\gamma = h\nu = \frac{hc}{\lambda} \qquad (2.1)$$

where h is Planck's constant ($6.626 \cdot 10^{-34}$ Js) and c is the velocity of light in vacuo.

X-rays are electromagnetic radiation emitted by charged particles in the changing of atomic energy levels (characteristic X-ray, fluorescence) or in slowing down in a Coulomb force field (bremsstrahlung).

Fast electrons are referred to as β-particles if they are emitted from a nucleus. If positive in charge they are called positrons. δ-rays are fast electrons resulting from a charged-particle interaction.

Heavy charged particles include protons, deuterons, tritons, α-particles, pions and other heavy charged particles consisting of the nuclei of heavier atoms, either fully stripped of electrons or having a different number of electrons than necessary to produce a neutral atom.

Neutrons are neutral nuclear particles. Since they cannot themselves be accelerated electrostatically, they are released in nuclear reactions.

2.2 Radioactivity

Radioactivity is the process of spontaneous emission of radiation. Radioactive nuclei, either natural or artificially produced by nuclear reactions, are unstable and tend to seek more stable configurations through expulsion of energetic particles.

2.2.1 Radioactive decay

Radioactivity is a statistical process. For one particular nucleus it is not possible to predict when exactly this nucleus will disintegrate. The probability p for a disintegration in a time interval < 0, t > is however possible to assume dependent of the length of the time interval. For a small Δt, p will be proportional to Δt :

$$p = \lambda \Delta t \tag{2.2}$$

λ is defined as the total radioactive decay constant and has the dimensions reciprocal of time. It is usually expressed in inverse seconds (s^{-1}). λ is assumed to be independent of the age of the atom.

If a nucleus has more than one possible mode of disintegration, the total decay constant can be written as the sum of the partial decay constants:

$$\lambda_{tot} = \lambda_1 + \lambda_2 + ... \tag{2.3}$$

The rate at which a particular decay process occurs in a radioactive sample is proportional to the number of radioactive nuclei present, N:

$$\frac{dN}{dt} = -\lambda N \tag{2.4}$$

By separating variables and integrate from t = 0, when $N = N_0$, we have:

$$\int_{N_0}^{N} \frac{dN}{N} = -\int_0^t \lambda t \tag{2.5}$$

$$\ln \frac{N}{N_0} = -\lambda t \tag{2.6}$$

$$N = N_0 e^{-\lambda t} \tag{2.7}$$

The decay rate λN is often referred to as *the activity* of the sample.

The old unit of activity was the *curie* (Ci), equal to $3.7 \cdot 10^{10}$ s $^{-1}$, which is equal to the activity in 1 g of ^{226}Ra. Today the *becquerel* (Bq) is the SI unit for radioactivity : 1 Bq ≡ 1 s^{-1}.

2.2.2 *Disintegration processes*

Disintegration processes often leave the nuclei in an excited state. In these cases excess energy is often released in form of gamma-rays, or photons, to leave the nuclei in the ground state. The total energy of the photons and other particles released by the disintegration process is equal to the net decrease in the rest mass of the neutral atom. Energy, momentum and electric charge are each conserved in the process.

Alpha disintegration is expulsion of a $^{4}_{2}$He nuclei and occurs mainly in heavy nuclei. An example of α-disintegration is the decay of radium to radon:

$$^{226}_{88}Ra \longrightarrow {}^{222}_{86}Rn + {}^{4}_{2}He + 4.78\,MeV \qquad (2.8)$$

The atomic number decreases by 2 and the atom sheds two atomic electrons from its outermost shell to become a neutral atom. The α-particle will capture two electrons from its surroundings after slowing down and thereby become a neutral $^{4}_{2}$He atom. The rest energy appears as kinetic energy and photons.

Beta disintegration appears in nuclei having an excess of neutrons or protons. Nuclei with excess of neutrons will emit an electron (β^-) and thus be left with one less neutron and one more proton. Nuclei with excess of protons usually emit a positron (β^+), decreases the atomic number Z by 1 while the neutron number increases by 1. β-ray emission leaves many kinds of nuclei in an excited state, and γ-rays are then emitted to reach the ground state. This is the situation for 94.6% of the ^{137}Cs disintegrations:

$$\begin{aligned} ^{137}Cs &\longrightarrow {}^{137m}Ba + e^- \\ ^{137m}Ba &\longrightarrow {}^{137}Ba + 0.662\,keV \end{aligned} \qquad (2.9)$$

Cs-137 disintegrates with a half life of 30.17 years. The Ba-nucleus is left in an excited state and γ-rays are emitted to reach the ground state. The half life for ^{137m}Ba is very short (0.6 μs) compared to ^{137}Cs and is therefore in equilibrium with ^{137}Cs.

Figure 2.1 shows the decay schemes for the isotopes radiocaesium ^{134}Cs and ^{137}Cs. They both decay by β^--disintegration to metastable or stable barium. Metastable barium decay to stable barium by γ-disintegration, and through detection of these photons radiocaesium can be detected with a NaI-detector. ^{137m}Ba emits 662 keV γ-radiation by photoelectric effect while ^{134m}Ba emits packets of γ energy at various wavelengths, including 605 keV, 563 keV and 569 keV.

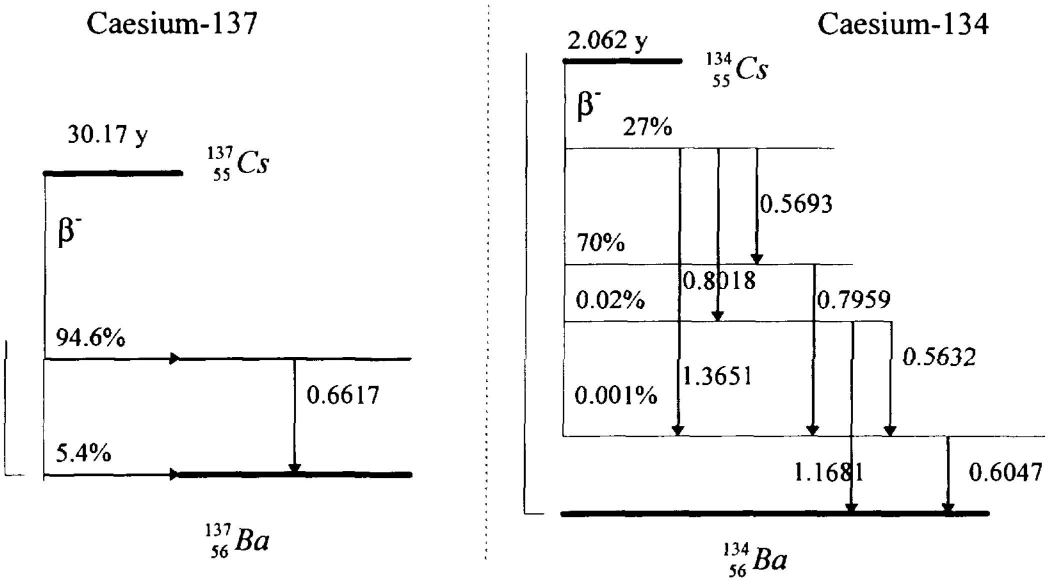

Figure 2.1: Decay schemes for ^{137}Cs and ^{134}Cs [6] . 94.6 % indicates the ratio of the ^{137}Cs disintegrating to a metastable ^{137}Ba, which in turn disintegrates by a 0.6617 MeV photon to stable ^{137}Ba.

Radioactive disintegration through *electron-capture (EC) transitions* are competitive with those by β^+- disintegration. The parent nucleus captures one of its own atomic electrons and emits a monoenergetic neutrino. An electron from a higher orbit will fill the resulting shell vacancies, and a fluorescence x-ray is released.

Internal conversion (IC) is a competitive process to γ-ray emission. Instead of emitting a γ-ray of energy $h\nu$, the excited nucleus can impart the same amount of energy directly to one of its own atomic electrons. This electron escapes the atom with a kinetic energy of $h\nu$ - E_b, where E_b is the binding energy of the electron.

Internal conversion is always possible in place of γ-ray emission by an excited nucleus, but the probability of IC is in many cases very small and can be ignored [7] .

2.2.3 Mean life and half life

The expectation value of the time needed for an initial population of N_0 radioactive nuclei to decay to 1/e of their original number is called the *mean life, T* :

$$\frac{N}{N_0} = \frac{1}{e} = e^{-\lambda T} \Rightarrow T = \frac{1}{\lambda} \tag{2.10}$$

Mean life is the average lifetime of an individual nucleus.

Physical half life, T_{phys}, is the expectation value of the time required for one-half of the initial number to disintegrate and hence for the radioactivity to decrease by half:

$$\frac{\lambda N}{\lambda N_0} = \frac{1}{2} = e^{-\lambda T_{phys}} \Rightarrow T_{phys} = \frac{\ln 2}{\lambda} \tag{2.11}$$

2.2.4 Biological and ecological half life

The factor λ is also used to describe the elimination of a matter from a biological system, and is in these cases termed the elimination rate. Distribution and metabolism of the radionuclide in the animal and the animal's own mechanism for excreting waste products will determine the biological half life. Often the biological elimination is better described using two or more factors, each factor referring to different compartments (ie, organs)in the animal. The physical decay constant and the elimination rate in the biological system set the total elimination rate :

$$\lambda_{tot} = \lambda_{phys} + \lambda_{bio} \Rightarrow \frac{1}{T_{tot}} = \frac{1}{T_{phys}} + \frac{1}{T_{bio}} \tag{2.12}$$

When radioactive matter contaminates an environment it is of interest to know how long it will reside. This is determined by the physical decay constant and all natural physical, chemical and biological processes influencing accumulation and elimination of the matter in the environment. One example is the rate of fixation in soil influencing bioavailability and uptake in vegetation.

For a specific compartment in the environment the long term behaviour of the radioactive contamination is often described by the ecological half life, T_{eco}. T_{eco} is defined as the time required to reduce by half the contamination of for instance an animal or an animal product when the animal continues to live in the same contaminated area. The effective half life T_{eff} is determined by the physical half life of the radionuclide (T_{phys}) and the ecological half life for the radionuclide in the environment (T_{eco}), analogous to Eq. 2.11.

For a short lived radionuclide the effective half life is essentially the same as the physical half life, while the ecological elimination rate will be most important for long lived radionuclides.

2.3 Effects of ionising radiation

2.3.1 Gamma- and X-ray interaction with matter

There are three major types of γ-and x-ray interaction with matter, each dominating at different photon energies. These are illustrated in Figure 2.2.

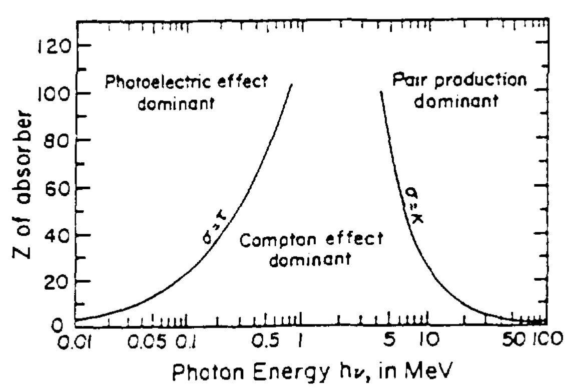

Figure 2.2: Relative importance of the three major types of gamma-ray interactions. The curves show the values of the atomic number Z and energy E for which the probability two types of interactions are equal [7].

When a photon interacts with a «free» electron, i.e. an electron whose binding energy is negligibly small compared with the photon energy, a part of the energy of the photon is given to the electron as kinetic energy (Figure 2.3). This is called *the Compton effect.* The photon with its remaining energy continues deflected from its original path and may take part in further interactions. The net result of Compton interactions is the production of a large number of fast electrons, so-called Compton electrons.

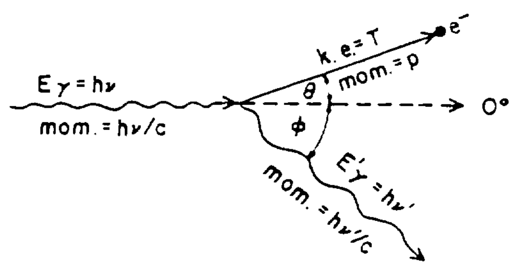

Figure 2.3: Kinematics of the Compton effect. A photon of quantim energy hν incident from the left strikes an unbound stationary electron, scattering it at angle θ relative to the incident photon's direction, with kinetic energy T. The scattered photon hν' departs at angle φ on the opposite side of the original direction, in the same scattering plane. Energy and momentum are each conserved [7].

The photoelectric effect (Figure 2.4) is the most frequent interaction of low energy photons with matter. The incident photon gives up all its energy in an encounter with a tightly bound electron, such as those in the inner shell of an atom. The photoelectric effect cannot take place with a given electron unless the incident photon's energy $h\nu$ is greater than the binding energy E_b for that electron.

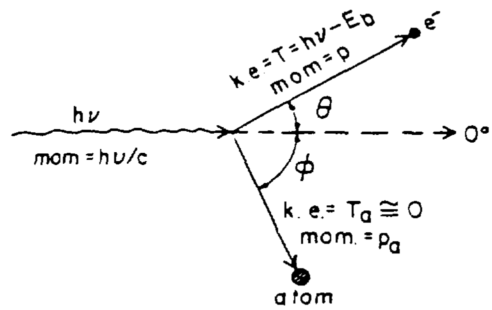

Figure 2.4: Kinematics of the photoelectric effect. A photon of quantum energy hν incident from the left strikes an electron bound to an atom with binding energy E_b. The photon vanishes, giving a kinetic energy $T = h\nu - E_b$ to the electron, which departs at angle θ relative to the incident photon's direction [7].

Pair production (Figure 2.5) is an absorption process in which a photon disappears and gives rise to an electron and a positron . It can only occur in a Coulomb force field, usually that near an atomic nucleus. A minimum energy of $2m_0c^2$ is required, where m_0c^2 is the rest energy of an electron.

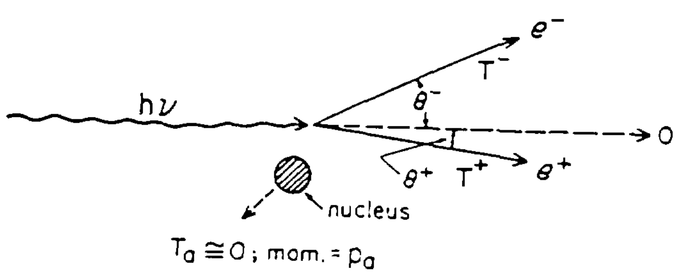

Figure 2.5: Pair production in the Coulomb force field of an atomic nucleus. An incident photon of quantum energy hν vanishes, giving rise to a positron-electron pair [7].

Others, less likely interactions are *Rayleigh scattering* (ie the photon is scattered by the combined action of the whole atom; elastic collision) and *photonuclear interactions* (energetic photon enters and excites a nucleus which then emits a proton or a neutron).

2.3.2 Charged particles interaction with matter

Uncharged radiation like photons and neutrons may pass through a slab of matter with no interactions at all, or they may interact and lose their energy in one or a few events.

Charged particles behave distinctly different from that of uncharged radiation. A charged particle, being surrounded by its Coulomb electrical force field interacts with one or more electrons or with the nucleus of practically every atom it passes. It loses its kinetic energy gradually in a frictionlike process. Charged particles can be roughly characterised by a common pathlength, or *range*. This range is given for particles of a given type and energy in a specific medium.

There are four main interaction processes for charged particles [8] :

- *inelastic collision with «atomic» electrons*, which is the dominating mechanism and leads to excitation or ionisation of one or more atomic electrons
- *inelastic collision with a nucleus* may take place when the charged particle gets close to a nucleus
- *elastic collision with a nucleus* changes the track of the charged particle, but does not involve any radiation or excitation of the nucleus. Electrons have high probability for this interaction
- *elastic collision with atomic electrons* leads to a energy loss in the charged particle that is less than the lowest excitation potential in the atom, and has importance only for lowenergetic electrons (≤ 100 eV).

The *linear energy transfer (LET)* or *collision stopping power* is of great relevance in radiobiology . LET of charged particles is the energy transferred per unit length of the track and has been used as a measure of biological effectiveness. For a given type of charged particle, the higher the energy, the lower the LET, and therefore the lower its biological effectiveness [9].

2.3.3 *Biological effects*

There is strong circumstantial evidence to indicate that DNA is the principal target for the biological effects of radiation [9]. The consequences of DNA strand breaks induced by radiation can be cell death, mutation or carcinogenesis.

There is a connection between biological damage and radiation energy absorbed, or dose. Absorbed dose is defined as the mean energy, E, imparted by ionising radiation to matter of mass m: $D=E/m$. The unit for absorbed dose is the Gray (Gy) and is equal to 1 J/kg. In considering the health or cellular effects of each particle or ray, it is convenient to normalise the various types of radiation. The quality factor Q is a dimensionless variable weighting factor to be applied to the absorbed dose to provide an estimate of the relative human hazard of different types and energies of ionising radiation.

When radiation with high LET (like neutrons and alpha-particles) are considered, *direct action* of radiation is the dominant process. This means that the atoms of the target itself may be ionised or excited, and thus lead to a biological change. The radiation may also interact with other atoms or molecules in the cell (particular water) to produce free radicals that are able to diffuse far enough to reach and damage the critical target. A free radical is a free atom or molecule carrying an unpaired orbital electron in the outer shell, and is therefore associated with a high degree of chemical reactivity. This is called the *indirect action* of radiation. Both direct and indirect action lead to changes from breakage of chemical bonds, which in turn may lead to biological effects.

Two types of damage may occur when a cell is irradiated. *Cell death* is usually seen soon after irradiation and is the early effect of larger radiation doses. The cell may also survive in an altered form called a *transformation*. The transformation can result in a cancer or in genetic damage. The effects of cell transformation may take years to appear and are therefore called the late effects of radiation.

There are several large studies that provide data on the health effects of radiation on people. These include external X-ray and gamma radiation and internal alpha radioactivity. The studies encompass atom bomb survivors, patients irradiated both in therapy and diagnostics, accidents with radiation sources in medicine and nuclear industry and from epidemiological surveys of other exposed groups. The experiences from these studies make the main basis for protection of the population from ionising radiation through the recommendations of International Commission for Radiation Protection (ICRP).

3. Radioactive fallout

3.1 Radioactive fallout in Norway

The main sources of man-made radioactivity are nuclear reactors and nuclear weapons. They are also the main potential sources of radioactive fallout through accidental releases from a reactor or through detonation of a nuclear weapon. The atmospheric nuclear weapons test during the 1950-60's produced world-wide fallout and ^{131}I, ^{137}Cs, ^{14}C and ^{90}Sr were the most important nuclides. The weapons fallout was deposited more or less continuously over a period of years because the detonation in the higher atmosphere caused thorough mixing of nuclides in the atmosphere. In 1986 the deposition in Norway due to nuclear weapons tests was as showed in Figure 3.1.

On April 26 1986 unit 4 at the nuclear power plant in Chernobyl, Ukraine, failure whilst conducting an experimental procedure resulted in an explosion at the power plant. Calculations indicates that ca. 3.5% of the radioactive substances in the core was released [10]. Table 3.1 shows core inventories and total releases from the Chernobyl plant[11] .

During the days 28.04 - 08.05 1986 fallout was deposited in Norway as illustrated in Figure 3.1. In contrast to weapons fallout the Chernobyl fallout was largely deposited over a period of a few days. Iodine-131 and radiocaesium represented the emphasis of the fallout in the Nordic countries [10].

Iodine-131 has a physical half-life of 8 days and is a beta- and gamma-emitter. Iodine-131 can appear as gas or in volatile condition and dominated in concentration in air and fallout during the first days after the accident [10]. Due to its short half life ^{131}I will be a radiological problem only for a few weeks. However, since iodine accumulates in the thyroid, radioiodine can contribute high radiation doses and cause long term effects like thyroid cancer in children [12].

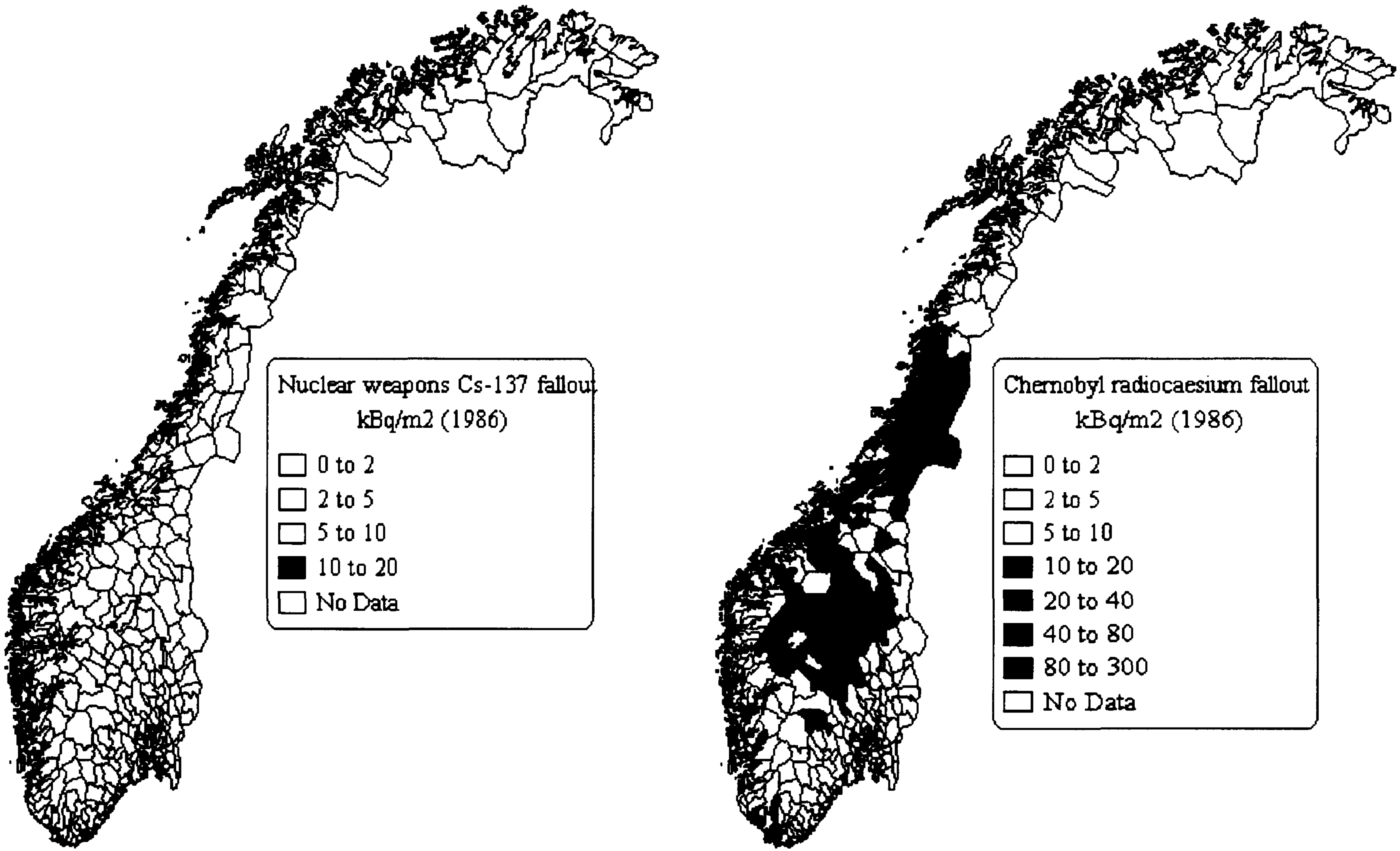

Figure 3.1: Deposition of radioactive fallout from nuclear weapons tests and the Chernobyl accident (NRPA data).

Table 3.1: Core inventories and total releases of radionuclides from the Chernobyl plant [11].

Element	Half-life (d)	Inventory (Bq)	Percentage released
^{85}Kr	3930	3.3×10^{16}	~ 100
^{133}Xe	5.27	1.7×10^{18}	~ 100
^{131}I	8.05	1.3×10^{18}	20
^{132}Te	3.25	3.2×10^{17}	15
^{134}Cs	750	1.9×10^{17}	10
^{137}Cs	1.1×10^4	2.9×10^{17}	13
^{99}Mo	2.8	4.8×10^{18}	2.3
^{95}Zr	65.5	4.4×10^{18}	3.2
^{103}Ru	39.5	4.1×10^{18}	2.9
^{106}Ru	368	2.0×10^{18}	2.9
^{140}Ba	12.8	2.9×10^{18}	5.6
^{141}Ce	32.5	4.4×10^{18}	2.3
^{144}Ce	284	3.2×10^{18}	2.8
^{89}Sr	53	2.0×10^{18}	4.0
^{90}Sr	1.02×10^4	2.0×10^{17}	4.0
^{239}Np	2.35	1.4×10^{17}	3
^{238}Pu	3.15×10^4	1.0×10^{15}	3
^{239}Pu	8.9×10^6	8.5×10^{14}	3
^{240}Pu	2.4×10^6	1.2×10^{15}	3
^{241}Pu	4800	1.7×10^{17}	3
^{242}Pu	164	2.6×10^{16}	3

The caesium isotopes *^{134}Cs* and *^{137}Cs* are beta - and gamma-emitters and have physical half-lives of 2.062 and 30.17 years respectively. They appear partly as gas or in volatile conditions, partly attached to particles. The long half life of especially ^{137}Cs makes radiocaesium a potential long term contamination problem following fallout. Radiocaesium accumulates inside sotf tissues, notably in muscle tissue. Since ^{134}Cs is an activation product, fallout from nuclear weapon tests contained almost no ^{134}Cs. In the Chernobyl fallout there was ^{134}Cs and ^{137}Cs in a ratio about 1:2, making it possible to determine the Chernobyl contribution to the total radiocaesium in the environment.

Strontium-90 is a beta-emitter and has a physical half life of 29 years. Radioactive strontium behaves in an analogous fashion to calcium and can be transferred to people effectively through milk. As strontium accumulates in bone, the radionuclide has a long biological half life.

The fallout from the Chernobyl accident was mainly deposited in mountain areas which are heavily used by grazing animals (see Figure 3.1). These areas are important in animal production, notably those including reindeer, sheep, goat and cattle.

3.2 Transfer of radiocaesium

Caesium is an alkali metal and displays similar electrical, chemical and physical properties to sodium, potassium and rubidium. In sheep and other animals, ^{134}Cs and ^{137}Cs are distributed relatively uniformly throughout most of the soft tissues, and there is little accumulation in bone [13], [14]. Muscle is therefore an important pool for Cs because of its greater relative bulk. In general, they follow the distribution of potassium, and also in soil both isotopes behave similarly and in a manner analogous to potassium.

The transfer of a radionuclide to animals has commonly been characterised using the transfer coefficient *TF*, defined as the ratio of the equilibrium tissue/milk activity concentration to the daily intake of the radionuclide [15]:

$$TF_{plant \to muscle} = \frac{\text{activity in tissue / milk (Bq / kg fresh weight)}}{\text{activity in plant (Bq / kg dry weight)}} \quad (3.1)$$

The transfer coefficient TF_g can be used to measure the transfer from soil to vegetation [15]:

$$TF_g = TF_{soil \to plant} = \frac{\text{activity in vegetation (Bq / kg dry weight)}}{\text{activity deposition per unit area (Bq / m}^2\text{)}} \quad (3.2)$$

The aggregated transfer factor T_{ag} describes the transfer from soil to animals or animal products and integrates the the activity transfer over the areas grazed by the animals [14]:

$$T_{ag} = TF_{soil \rightarrow muscle} = \frac{\text{activity in tissue / milk (Bq / kg fresh weight)}}{\text{activity deposition per unit area (Bq / m}^2\text{)}} \qquad (3.3)$$

3.2.1 Transfer from soil to vegetation

The transfer of radiocaesium from soil to vegetation varies according to soil type and plant species. Soil can be classified according to its ability to immobilise caesium and this immobilisation capacity is a reflection of among others, the clay mineral content and type, organic content, pH, and concentration of competing cations (e.g ammonium, potassium or stable Cs), but the processes are not all understood. A study of radiocaesium transfer from soil to herbage in nordic environments[16] showed transfer factors of 18 - 82 m^2/kg for organic and acidic soils while transfer factors from sandy soils and clay soils were as low as 0.4 - 0.8 m^2/kg.

Mineral soils predominate in the majority of agricultural areas and are characterised by appreciable quantities of clay mineral and moderate to low content of organic matter. Transfer of ^{137}Cs to vegetation is small from these soils due to its fixation in the lattice structure of clay minerals, and high potassium content in soils may reduce transfer further[17] .

Caesium is more available when pastures have developed on organic soil with a distinct root matt and on sandy soils [18] as these soils fix caesium less efficiently and therefore more caesium is available for plant uptake [19]. In relatively cool, humid regions accumulation of dead plant material can exceed the rate of decay and distinct organic layers will develop in the surface horizon under permanent pastures. This explains why transfer of radiocaesium to vegetation remains high several years after its deposition [20].

In undisturbed soils activity of ^{134}Cs and ^{137}Cs will decrease exponentially down the soil profiles [20] . A study of mobility and plant availability of Chernobyl radiocaesium [21] in natural soil-plant systems of varying fertility in subalpine areas in central Norway showed that the ^{137}Cs was fixed in the litter and the upper few centimetres of the soil, and little downward movement in the soil profile was observed. Due to this the variations in transfer levels between different plant species can mainly be explained by the rooting depth. Deep-rooted species have shown lower transfer than species with a superficial rooting system [31].

Radiocaesium is more readily available to plants than stable Cs, since stable Cs is bound in the lattice of soil mineral particles [21] .This may also to some extent explain the different vertical distributions of ^{137}Cs relative to stable Cs in the root zone.

Biomass production on unimproved ecosystems like sheep grazing land is low compared to managed lands. The soils are often acidic and of poor nutrient status, with a content low in potassium and high in organic matter, especially in the surface layers. Such organic soils often have a low content of clay minerals which may explain the continuing high radiocaesium transfer to vegetation and grazing animals. Unimproved ecosystems show a great variety with respect to the number of plant species. The sheep often graze over large areas, and may over the grazing season select a large variety of different plant species. An estimation of vegetation consumption for sheep is 2 kg dry weight daily [22], but intake varies according to the nutrition value and digestibility of the species eaten.

3.2.2 Transfer from feed to animals

The source and chemical form of a radionuclide affects its availability for uptake and transfer into the tissues of grazing animals. Measurement of transfer coefficients have shown differences in the bioavailability of radiocaesium when associated with various sources and apparent differences in transfer between ruminants of different species or physiological status.

Ionic caesium has in experiments shown a significantly higher transfer coefficient than soil-bound caesium, while transfer coefficients for vegetation varied between different species [23]. In an experiment mesuring radiocaesium transfer from different sources to goat milk a transfer factor of 0.12 dL^{-1} was found for $^{134}CsCl$ [24]. For hay the transfer factor varied from 35% (in 1986) to 100% (in 1988) of this. The low transfer for hay from 1986 is due to the lower bioavailability of direct contamination on the plant surface from fallout, compared to when the activity had been incorporated into the grass by root uptake. Bioavailability of radiocaesium associated with fungi was high and approached that of ionic caesium (78 - 87% of transfer from ionic Cs). Transfer from organic soil was only 7% of transfer from ionic caesium.

Physiological factors like pregnancy, lactation and growth rate may influence the radiocaesium uptake by ewes and lambs [25]. Age of animals can also affect the radiocaesium content in animals. For all tissues transfer coefficients are shown to be higher for lambs than for ewes [26]. Experiments measuring transfer of ^{137}Cs from different sources showed a high bioavailability for milk [26], this implies that milk is an important source of radiocaesium for young lambs and may contribute to the higher activities found in lambs compared with ewes. Milk is however the major source of radiocaesium only for young lambs (< 6-7 weeks), while radiocaesium intake from pasture is the major factor determining radiocaesium concentrations in lambs at time of slaughter [27]. A factor like dietary calcium level may also affect the radionuclide uptake, low dietary calcium level might increase the absorption of radiocaesium [28].

Grazing animals will ingest soil adhered onto vegetation. Therefore, as the concentrations of radiocaesium are higher in soil, ingested soil may be an important source of radionuclides to grazing animals. The contribution of adherent soil to the total ^{137}Cs of vegetation samples appears to be gradually increasing with time after the deposition of Chernobyl fallout [29]. Radiocaesium associated with soil, however has lower absorption in the gut compared to radiocaesium incorporated in vegetation [30] and animals grazing pastures with significant amounts of radiocaesium associated with adhered soil will therefore not be as contaminated as radiocaesium concentrations measured in bulk vegetation would suggest. Because of the low transfer factor for soil-bound radiocaesium this factor will have minor influence on radioactive contamination of milk and meat unless the soil concerned exhibits an unusually high bioavailability of radiocaesium.

The biological half life of caesium in muscle is considerably longer than that in other organs and radiocaesium is not equally distributed to different tissues. A study of transfer of radiocaesium to sheep from Chernobyl-contaminated vegetation showed different transfer factors for different tissues: muscle (12.0 d kg^{-1}) > kidney (9.6) > liver (7.1) > milk (5.7, lactating ewes) ≈ lung (5.2) ≈ mammary gland (6.2) [23].

Aggregated transfer coefficients from soil to meat varied from 0.023 m^2kg^{-1} for ewes to 0.031 m^2kg^{-1} for lambs in a Norwegian study [27].

3.2.3 Fungi

Fungi play a key role in nature as they decompose dead organic matter and contribute to the nutritional status in plants. Fungi is also known to be seasonally important in the diets of free ranging animals like sheep, goats, reindeer, cattle and deer.

In 1988 high levels of radiocaesium were observed in meat and milk from animals grazing in the Norwegian mountains. The radioactivity levels in sheep and reindeer increased rapidly during the second half of the summer, an increase that coincided with the early appearance of large quantities of fungal fruit bodies. It has long been known that fungi accumulates caesium more effective than plants, but it was not expected that the large amounts of fruit bodies would be of such importance to radioactivity concentration in animals [4].

Mushroom take up most of their nutrients from the upper organic soil horizon, where most of the radiocaesium from the Chernobyl fallout is still present. Fruit bodies with a very high radiocaesium level are often «acid soil fungi», ie fungi with high capacity for accumulation inorganic ions and heavy metals from environments with a low content of the actual ions [5]. Measuring radiocaesium in fungi and plants collected from the same spots, most of the fungi species contained 10-150 times (dry weight for both fungi and plants) the content found in plants [5]. This has been demonstrated for radiocaesium from nuclear weapon tests as well as from the Chernobyl fallout. There are large differences in accumulation of caesium both among species of fungi, and within the same genus [5]. Transfer factors from soil to fungi were found to vary from 0.004 to 2.8 in a German study [32]. Symbiotic or mycorrhizal fungi, with huge nutrient absorbing network of hyphea showed the highest transfer factors while the lowest transfer factors are associated with saprophytes, a group of fungi that lives on organic substrate. This difference can partly be explained by the depth of the mycelium, as fungi whose mycelium extend to deeper soil layers seem to be less contaminated [22]. Fungal activity may also be important in mobilising deposited radiocaesium and recycling radiocaesium [33].

The factors that influence the appearance of fruit bodies are not all known. It is however suggested that a certain amount of accumulated temperature must be reached, in addition to that a certain amount of soil water must be present during the growth period. Measurements of the soil physical factors and the microbial activity during the summer might possibly give information about factors that influence the mushroom season. Mushroom of different genera seem to develop at different times of the mushroom season, and studies in the Jotunheimen area show that the production of fruit bodies is higher on podzol than on brown earth [34] .

High radiocaesium levels in animals and animal products have been linked directly to the ingestion of fungi on contaminated summer ranges. Since the Chernobyl accident, radiocaesium levels in grazing animals have reflected directly the abundance of fungi [35]. In contrast to vegetation, studies of long term behaviour of radiocaesium in fungi have not detected any significant decrease of the ^{137}Cs activity since the Chernobyl accident [32],[36], and fungal radiocaesium will therefore be of importance for radiocaesium concentrations in animals for many years.

3.3 Countermeasures

During the initial phase after a radioactive fallout situation radionuclides are present in the air, and inhalation is normally the main route of exposure together with external exposure. Later ingestion of radioactivity through food is the main source of radionuclides. There is therefore a need to investigate ways of preventing or reducing the accumulation of radiocaesium in animal products, in order to limit the harmful effect of ionising radiation from radioactive fallout. The use of countermeasures is justified when the cost of their implementation is lower than the expences related to the averted dose (ICRP-60). Another factor influencing the use of countermeasures is the public demand for «clean food».

In animal production there are several ways of reducing the contamination problem:

- reduce uptake from soil to plants by use of fertilisers, ploughing or changing the land use
- reducing transfer from plant to animal by the use of food additives and boli
- reduce intake of contaminated feed before slaughter
- increasing the rate of excretion from the animal

Food production in Norway is in a special situation since sheep, cattle and goats graze freely on unimproved pasture in forest and mountain areas during summer. Under these circumstances soil treatments to reduce uptake from soil to plants are costly and impractical. A more practical way to reduce the radiocaesium level in animal products is to reduce uptake of radiocaesium in the animal by the use of so-called caesium binders, ie different materials known to chemically bind caesium. These may be administered either in the form of intraruminal boli, or incorporated into salt licks which are placed where the animals graze. For meat animals giving the animals uncontaminated feed can also be effective at lower activity levels because of the relatively short biological half life for radiocaesium in animals (about 20 days for sheep [37]).

Stable caesium fed as CsCl will reduce radiocaesium accumulation in muscle tissues [38], blocking the muscle uptake of radiocaesium may lead to higher secretion in milk, which may be disadvantageous.

Potassium loading of rodents has been shown to enhance the excretion of radiocaesium and changes in potassium intake have been suggested as an explanation of the rapid increase in radiocaesium excretion which occurs in reindeer when the diet changes in spring from lichen to green plants with a high potassium content. However, experiments offering KCl licks to reindeer during winter indicated that KCl was not successful as a countermeasure. Similarly, increased excretion in lambs fed supplementary K was not significant [38] .

Clay minerals like bentonite, zeolite and clinoptilite can also reduce the transfer of radiocaesium to ruminants by reducing absorption. They have good ion-exchange capacity, and up to 50 % reduction in the transfer of radiocaesium to milk or meat have been reported. It is not practical to feed clay minerals to free ranging animals and clay minerals are not sufficiently effective on a weight basis to be used in salt licks or sustained release boli [38].

Prussian blue, Iron blue, Berlin blue are synonyms for a range of salts of hydroferrocyanic acids which act as a chelating agent for caesium. AFCF - ammonium-ferric-cyano-ferrate - acts as a specific ion exchange substrate of high capacity and selectivity for radiocaesium. Since the colloidal solubility of CsFCF is low compared with NaFCF and KFCF, AFCF binds caesium so strongly that the unabsorbable Cs-AFCF complex moves through the gastrointestinal tract unchanged and is excreted with the faeces. Small amounts of AFCF will be broken down to free iron and hexacyanoferrat when it passes through the intestine. Hexacyanoferrate will in turn to a very small extent break down to free iron and free cyanide and then be eliminated through the kidneys [39].

Experiments have shown that radiocaesium content in sheep is reduced with 25-75 % by use of Prussian blue in salt licks and 50-87 % by use of sustained release bolus [40], [41] . The radiocaesium transfer to goatmilk was reduced with 80-90 % by the use of Prussian blue bolus[41], [42]. Figure 3.2. shows how the treatment of goats with 1, 2 and 3 Prussian blue boli affected the fraction of administered ^{134}Cs transferred to goat milk.

The first years after the Chernobyl accident concentrates with 5-10% clay minerals were used in Norway and Sweden. Since 1989 Prussian blue has been used as admixture to feed concentrate in Norway, in salt licks and in rumen boli.

Salt licks with 2.5% AFCF have been widely used in mountain pastures in Norway. Because of the low sodium content in inland areas, sheep and cattle are accustomed to visit places where salt is available, and a daily average intake of 1 g salt would be expected to give a 50 %

reduction of meat radiocaesium levels in a 30- 40 kg lamb [38]. The efficiency does however depend upon frequent visits to the licks, and in a grazing herd there will always be some sheep which do not get any AFCF.

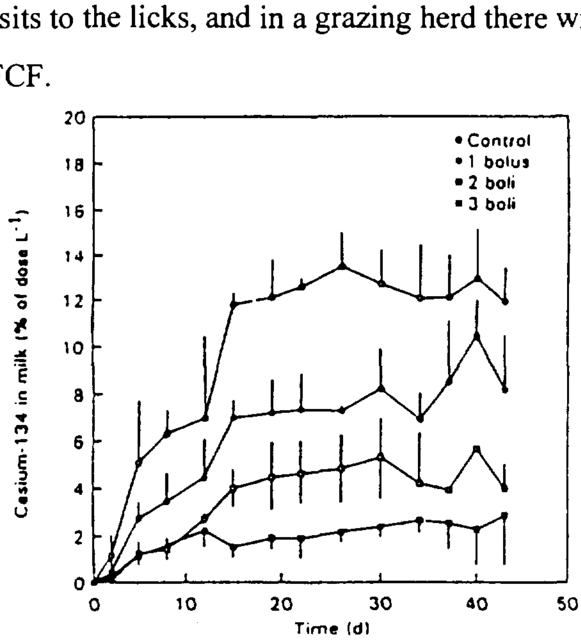

Figure 3.2: Fraction of the daily administered ^{134}Cs amount transferred to milk in control goats and goats treated with 1, 2 or 3 boli (mean and standard deviation for 4 goats) [42] .

The low daily doses of Prussian blue required to keep the radiocaesium level down make these caesium binders well suited for administration by slow release boli, with 15-20 % AFCF, placed in the rumen. Problems arise when treating animals not being in routinely contact with humans (eg sheep and reindeer), since the boli dissolve before the grazing season is over. However, recent improvements have prolonged the life time of the boli by a few weeks [43].

4. Materials and methods

4.1 Monitoring of live animals

A portable, battery operated Canberra series 10 multi-channel analyser was used together with a 3''x 3'' NaI(Tl) -detector to measure the radiocaesium levels in live sheep.

4.1.1 Na-I detector

The thalliumactivated sodium iodide detector scintillate, i.e. emits small quanta of light, when a γ-quanta is absorbed in the crystal. This fluorescence is due to deexcitation of local excited levels in the crystal. A photomultiplicator tube transforms the light quanta to electrical pulses. These pulses are proportional with the energy of the absorbed γ-quanta. The energies of the γ-radiation from radiocaesium are described in Figure 2.1.

4.1.2 Live monitoring

During measurements in the field the counts registered are due both to radiation from the animal and from the surroundings (i.e. background radiation). Therefore the background radiation will contribute to the measured radioactivity and must be subtracted to estimate the true radiocaesium concentration in the animals. The background radiation B_1 must be measured at the same site and at the same height above ground as the animals. For animals of different size, like sheep and lambs, different background measurements need to be taken.

In addition, when the detector is held against an animal, the animal will partly shield against radiation from the ground. To compensate for this the observed background level is corrected with a reduction or shielding factor in calculation of the radiocaesium concentration in the animal. Experiments with «clean» animals (i.e. animals from low contaminated areas) give the estimates for the shielding factor given in Table 4.1:

Table 4.1: Shielding factors for reindeer, sheep and cattle [44].

	reindeer	sheep	cattle
live animals	0.80	0.72	0.71
carcasses	0.88	0.88	0.82

Consequently the background contribution during the live monitoring of sheep is :

$$B_{sheep} = B_l \times 0.72 \tag{4.1}$$

The monitoring instruments are routinely controlled by measuring a phantom with known activity.

When estimating the radiocaesium concentration in the monitored animal from the counts registered in the multi-channel analyser the *geometry factor* is used. The geometry factor G is the ratio between activity in live animals / carcasses and the net counts registered, and is obtained through calibration against measurements of tissue samples from animals which have been slaughtered:

$$G\,[\,\mathrm{Bq}/(\mathrm{kg}\cdot s)] = \frac{\mathrm{A}}{\mathrm{N}} \tag{4.2}$$

where A is radiocaesium concentration in meat sample and N is net counts from the monitoring of the live animal.

The geometry factor will change with γ-energy, geometry and detector. In the LORAKON live monitoring the equipment is set to register pulses both from the 605 keV ^{134}Cs and the 662 keV ^{137}Cs peak; thus G will also change with changing isotope relation (i.e., time). Table 4.2 gives the geometry factors for sheep, reindeer and cattle used in 1993. Growing animals will change in size (i.e. in geometry), and the geometry factor will depend on the time of the year when measurements are performed. It is therefore necessary to estimate different geometrical factors for summer and autumn monitoring of lambs and calves.

Table 4.2: Geometry factor for calculation of radioactivity in meat for reindeer, sheep and cattle [44].

	reindeer			sheep				cattle	
calf :	**17**	**14**	**12**	lamb:	32	29	25		
adult :	**x**			adult:	25			adult:	11

The geometry factor depends on detector efficiency and also on where the detector is placed on the animal. The factors in Table 4.2 are calculated for detectors placed on *os sacrum* (standing animal), between the hind legs (animal lying on its side) and on the back, midway between *os sacrum* and *trochanter major* (standing animal) of sheep, reindeer and cattle respectively. It is a calibration factor defined for a unique method of monitoring and new calibration is necessary if any changes in method, geometry or instrument should find place.

The measured radiocaesium level A depends on radioactivity (counts) intercepted in the detector N, shielding factor S, background radiation B_l and geometry factor G according to the following expression:

$$A = G \cdot (N - S \cdot B_1) \qquad (4.3)$$

4.2 Available survey information

In 1986 radiocaesium concentration in meat were monitored using samples from slaughtered animals from the contaminated parts of Norway. Since 1987 live monitoring of sheep grazing in contaminated mountain areas has been conducted to ensure that only animals with relatively low radiocaesium levels are slaughtered. This is a part of the LORAKON system (Local Radiation Control). Available data from these surveys are:

- person responsible for the measurement
- grazing unit/place for monitoring
- age of animals (ewe or lamb)
- date of monitoring
- use of countermeasures
- applied geometry factor
- measured background radiation
- estimated radiocaesium concentration in animals [Bq/kg]

An example of the live monitoring report sheet is shown in Appendix 1. This information is recorded on basis of *grazing units;* most of the Norwegian sheep farmers are organised in associations based on geographical connections. All animals belonging to members of one associations graze in the same area, a so-called grazing unit. From each grazing unit a total of 8-10 animals (ewes and lambs) are monitored. If the median radiocaesium level in these sheep is above the national limit, i.e. 600 Bq/kg, animals from the whole grazing unit must either be fed uncontaminated feed until the radiocaesium level decreases to below the limit, or, for very high levels, slaughtering of animals from this area is not permitted. The results from the live monitoring are the basis for dividing the country into *zones*; «free zones» where the radiocaesium level is traditionally low and there is no need for further intervention, «observation zones» where further monitoring must be done, and prohibited zones where no production of is allowed.

For this study live monitoring records from the counties Hedmark and Oppland were chosen since these areas received considerable Chernobyl fallout (see deposition map, Figure 3.1). Some of the most contaminated grazing units in these counties have been surveyed all years, giving a complete list of measurements from 1987 to 1995. When using these survey data for studies of long term behaviour of radiocaesium it is important that as much information (i.e., the above list) as possible is available. Unfortunately, many are incomplete regarding monitoring date and information about use of Prussian blue.

As mentioned in Section 3.2.3, fungi is known to have an important effect on radiocaesium concentration in grazing animals. Studies of time for development of fruit bodies are therefore important in order to predict radiocaesium in sheep at the time of slaughter. Independent of the live monitoring of sheep, NRPA has been studying radiocaesium in fungi in mountain areas in Norway where subjects such as radiocaesium transfer from soil to fungi, difference in radiocaesium accumulation between fungi species, appearance of fruit bodies of different genera etc. have been addressed [45]. This makes it possible to get information about the amounts of fungi in some areas from year to year. However, these fungi studies are relatively few and do not cover the same areas as the LORAKON monitoring of sheep.

4.3 Model development

Winter feeds like hay and silage are produced on cultivated land and contain relatively low radiocaesium concentration. Combined with a biological half life of about 3 weeks, the winter feeding makes sheep low in radiocaesium content at the beginning of the grazing season. The

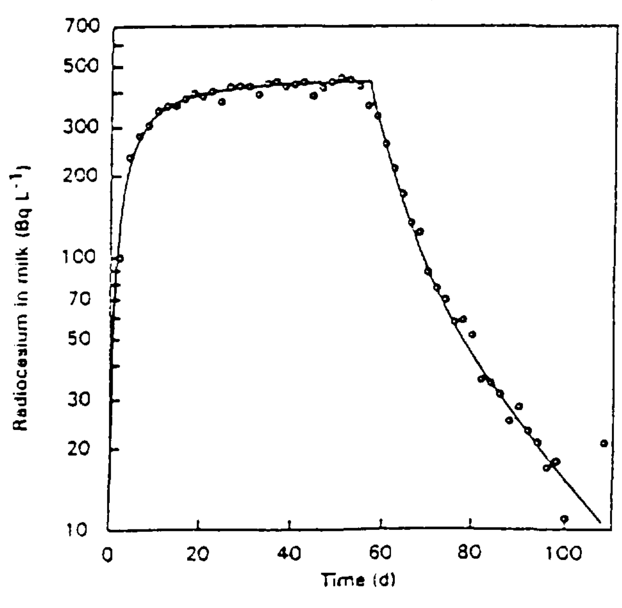

Figure 4.1: Milk ^{134}Cs concentration in goat milk during a 52-d period of feeding 3.4 kBq/d and during a subsequent loss phase [24].

radiocaesium content increases as the animals eat contaminated vegetation. If fed uncontaminated feed, the radiocaesium concentration will decrease again. As an illustration, Figure 4.1 shows the change in goat milk ^{134}Cs concentrations during a period of feeding $^{134}CsCl$ and during a subsequent loss phase.

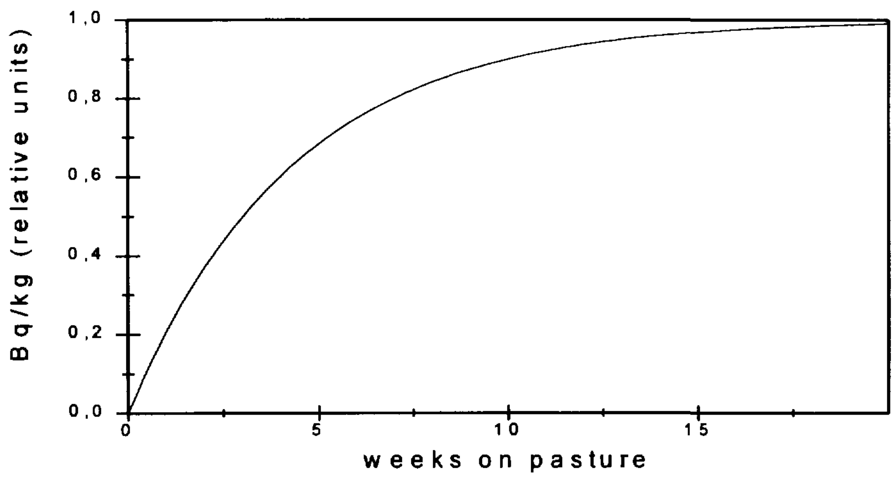

Figure 4.2: Theoretical increase in radiocaesium level in sheep grazing pasture with homogenous contamination in vegetation. An estimated biological half life of 3 weeks is implemented.

Thus, when released on pasture the activity level in meat will theoretically rise from about 0 Bq/kg as illustrated in Figure 4.2. An important assumption is that the animals graze on a pasture with constant radiocaesium concentration in the vegetation. This development can be described using the equation :

$$RC\,(t_p) = B\,(1-e^{-\lambda_{bio} \cdot t_p}) \qquad (4.4)$$

where RC (t_p) is the radiocaesium concentration in animals in Bq/kg, t_p is days on pasture and λ_{bio} is the effective biological elimination rate. The parameter B gives the equilibrium radiocaesium level and will for instance depend on radiocaesium concentration in vegetation, intake of and radiocaesium concentration in fungi, and also on countermeasures in the area.

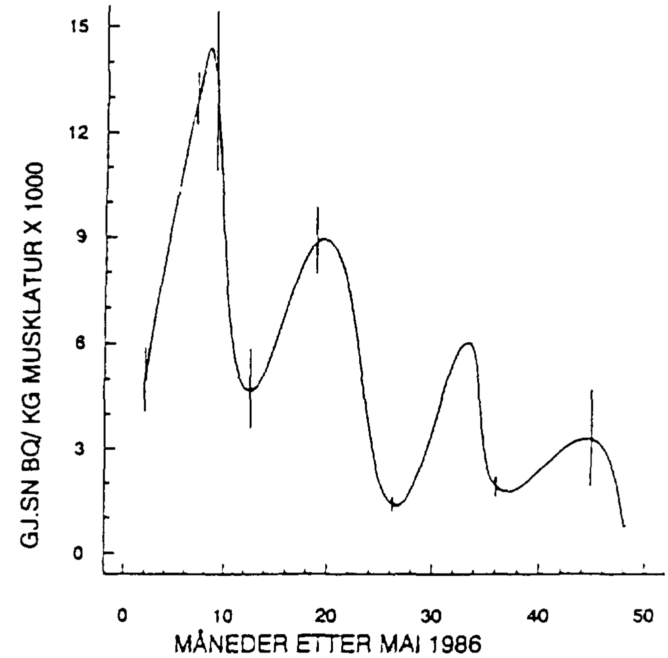

Figure 4.3: Season dependent radiocaesium concentration in reindeer from Knutshø in the period May 1986 to May 1990 as a function of months after May 1986. The vertical lines show the standard error [46].

After a single deposition event like the Chernobyl fallout it is assumed that radiocaesium concentration in an environment will decrease exponentially over years (due to physical decay and ecological processes, as movement down the soil profile), if no measures are taken against the contamination. This exponential decrease is illustrated in Figure 4.3, where also the seasonal change of radiocaesium concentration in reindeer is illustrated.

Another illustration of «ecological decay» is given in Figure 4.4, showing the theoretically decrease in radiocaesium concentration in one compartment of an ecosystem, in this case sheep. The decrease is described by :

$$RC\,(t_c) = C \cdot e^{-\lambda_{eff} \cdot t_c} \qquad (4.5)$$

where RC (t_c) is radiocaesium concentration in sheep in Bq/kg, t_c is time after Chernobyl (in years) and λ_{eff} is the effective elimination rate. *C* is the radioactivity level at time = 0 (1986 in the case of Chernobyl fallout). However, since the fallout in 1986 resulted in direct contamination of above ground plant parts, data from that year can not be included in estimations of ecological half lives, and the value of the constant C is therefore of little interest in this connection. The approximate equilibrium radiocaesium concentration animals on pasture reach during a grazing season will theoretically change from one year to the next according to this exponential decrease. This is illustrated in Figure 4.4 b).

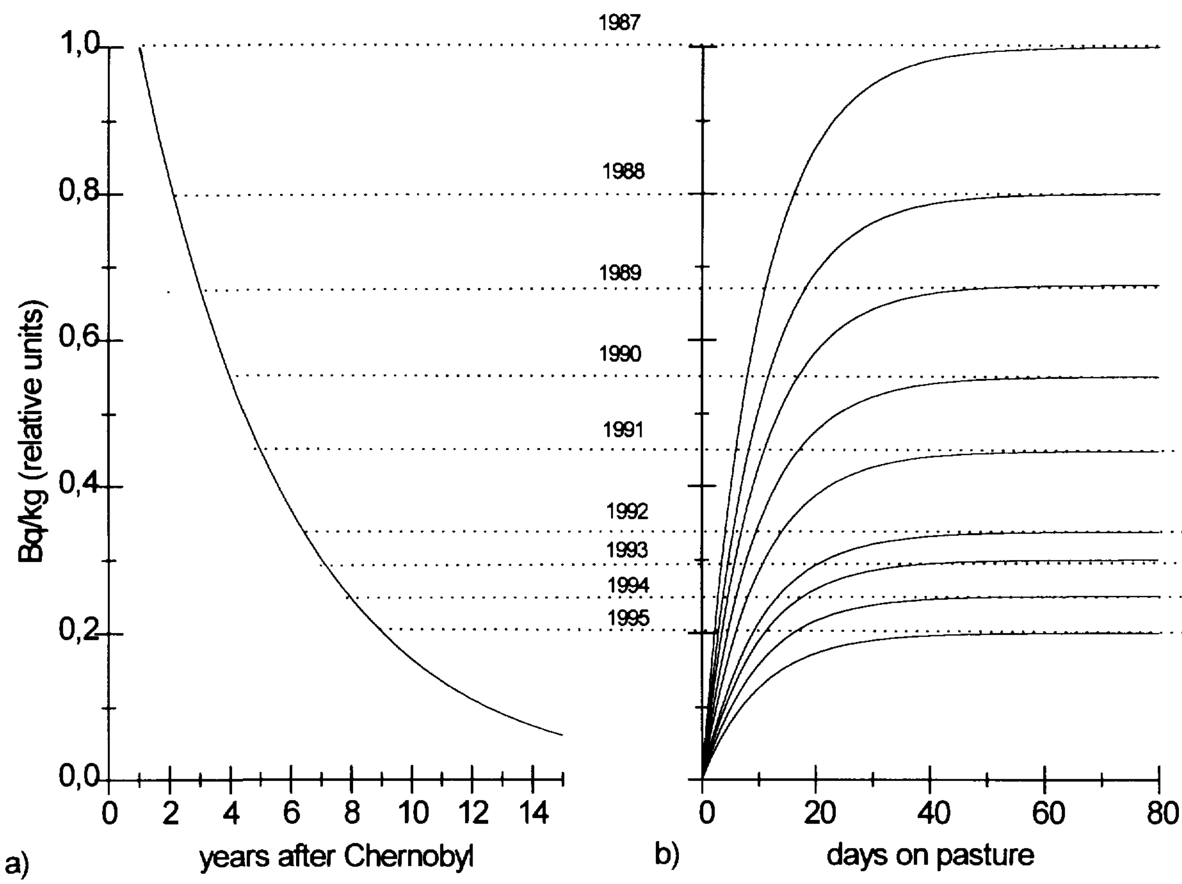

Figure 4.4: Theoretical development in radiocaesium activity concentration (a) illustrates the equilibrium levels for grazing sheep due to intake of contaminated vegetation in different years (b).

The first 4-6 weeks the animals will consume contaminated vegetation and the radiocaesium concentration will increase. Fungi, with high radiocaesium concentration, appear in late summer and will cause higher radiocaesium levels as animals consume them. The increase in radioactivity in animals caused by fungi will depend on which genera that appear, amount of fruit bodies and also on how long the fruit bodies are available as feed for the animals. This is illustrated in Figure 4.5. One reason for unexpected changes in radiocaesium levels in sheep from one year to another (as happened in 1988, Section 3.3.2) is earlier appearance of fruit bodies, appearance of genera with specially high capacity to accumulate radiocaesium, larger amounts of fruit bodies, longer time in pasture etc. If, for simplicity, the radiocaesium

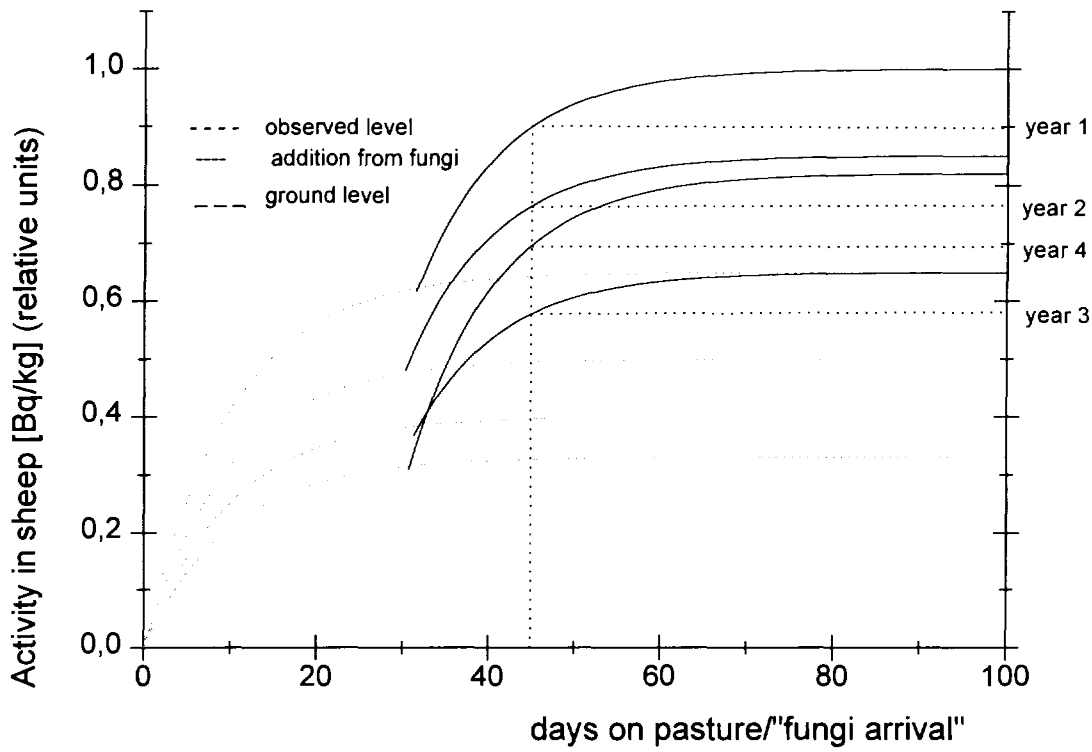

Figure 4.5 : Effect of appearance of fruit bodies on radiocaesium concentration in animals. ...... indicate what would be the observed radiocaesium activity in sheep by live monitoring, - - - illustrates the basic radiocaesium level due to consumption of vegetation and —— illustrates the additional radiocaesium activity due to intake of fungi.

accumulating fruit bodies are assumed to arrive at approximately the same time every year (and the same species arrive in the same amounts), the important factor for radiocaesium concentration in animals will be time on pasture, or more specifically, time on pasture with fungi available as feed for the animals. The radiocaesium level in animals during a grazing season can then be approximated by (see Fig.5.5) :

$$RC\ (t_c, t_p, F) = C \cdot e^{-\lambda_{eff} \cdot t_c} + B \cdot F \cdot e^{-\lambda_{fungi} \cdot t_c} \cdot (1 - e^{-\lambda_{bio} \cdot t_p}) \qquad (4.6)$$

where the first term estimates the activity concentration due to intake of vegetation (Fig. 4.4) and the second term the additional radiocaesium level due to consumption of fungi. The factor F estimates the amount of fungi every year and λ_{fungi} describes the ecological decrease in radiocaesium concentration in fungi. t_p is time on pasture with fungi available as feed. The factors B and C were defined in Eqs. 4.2 and 4.1 respectively. By using different elimination constants for fungi (λ_{fungi}) and vegetation (λ_{eff}) the model allows for the differences in ecology between these two organisms.

Other factors like different areas and ages of animals can be added to the model linearly.

The available information about amount of fungi is recorded simply as relative units «bad», «medium» and «good» year for fungi (and refer to the number of fruit bodies only). Therefore one alternative for setting the values of F is 1, 2 and 3 as estimates of the different amounts. However, the relative importance of the different amounts of fungi for the radiocaesium levels in sheep is not known (i.e. it is impossible to say that a good year is three times more important than a bad year). This relationship can be estimated by dividing B·F in three factors; $D \cdot F_1$, $E \cdot F_2$ and $G \cdot F_3$ where F_1, F_2 and F_3 are indicator variables and

$F_1 = 1$ for a «good year», 0 otherwise,
$F_2 = 1$ for a «medium year», 0 otherwise, and
$F_3 = 1$ for a «bad year», 0 otherwise,

and D, E and G are equivalents to B in Eq. 4.3. Hence, the radiocaesium concentration can be described as a function of t_c, t_p and the indicator factors F_1, F_2, and F_3 :

$$RC\ (t_c, t_p, F_1, F_2, F_3) = C \cdot e^{-\lambda_{eff} \cdot t_c} + (DF_1 + EF_2 + GF_3) \cdot (1 - e^{-\lambda_{bio} \cdot t_p}) \qquad (4.7)$$

Then the ratio D : E : G can then be used as an estimate of the relative importance of the different amounts of fungi.

4.4 Analysis of data and results

Statistical analysis is a common tool for description of data, thus forming a basis for discussion of the data and the results obtained from analysis. The statistical analysis in this project was performed with programware SPSS.

4.4.1 Distributions

Repeated observations that differ because of experimental error often vary about some central value in a roughly symmetric distribution in which small deviations occur more frequently than large ones. The *Gaussian* or *normal distribution* is a continuous distribution that represents this situation.

The normal distribution is characterised by the mean μ and the variance σ^2. The variance σ^2 measures the distance from the mean μ to the point of inflection of the curve.

In biological and ecological systems, where only positive values of a factor is possible, distributions are often skewed and they are often approximated by *lognormal distributions* (i.e. distributions of a random variable whose logarithms are normally distributed). The data are logtransformed before further analysis and calculations that requires normal distributed variables. Radiocaesium concentration in a flock of sheep is an example of data that would be expected to follow a lognormal distribution. A few, high radiocaesium levels will give an important contribution in calculation of the mean, but as negative values are not possible the data would follow an asymmetric distribution with relatively few measurements above mean.

4.4.2 *Finite sample variance*

If a random sample of size n is taken from a population having the mean μ and the variance σ^2, then the mean of the sample $\bar{x}$ is a value of a random variable whose distribution has the mean μ. For samples from a finite population of size N, like the population of sheep in a grazing unit, the variance is:

$$\sigma^2_{sample} = \frac{\sigma^2}{n} \frac{N-n}{N-1} \tag{4.8}$$

When monitoring radiocaesium concentrations in sheep from a grazing unit, 8-10 animals are monitored and the mean is used as an estimate of the average radiocaesium concentration in the whole grazing unit. When a sample mean, $\bar{x}$, is used to estimate the population mean, μ, the chance is virtually non-existent that the estimate will actually equal μ. Hence, it would seem desirable to accompany such a point estimate $\bar{x}$ with some statement as to how close to μ we might expect $\bar{x}$ to be. The error, $\bar{x} - \mu$, is the difference between the estimate and the quantity it is supposed to estimate. If the populations standard deviation σ is known, or if n is large (i.e., $n \geq 30$) it is known that [47] :

$$-z_{\alpha/2} \leq \frac{\bar{x}-\mu}{\sigma_{sample}} \leq z_{\alpha/2} \tag{4.9}$$

where $z_{\alpha/2}$ is the α- quantile for a normal distribution at significance level α. This leads to the following expression for the maximum error of estimate E, where E stands for the maximum value of $|\bar{x} - \mu|$:

$$E = z_{\alpha/2} \cdot \sigma_{sample} \tag{4.10}$$

This expression can also be used to determine the sample size that is needed to attain a desired level of precision in the estimation of μ. If only a maximum prescribed error E is permitted, and this is wanted to be asserted with probability 1 - α, the required sample size taken from a finite population of size N is :

$$n = \frac{N \cdot z_{\alpha/2}^2 \cdot \sigma^2}{z_{\alpha/2}^2 \cdot \sigma^2 + E^2 \cdot (N-1)} \qquad (4.11)$$

If n is small (≤ 30), and it is reasonable to assume that we are sampling from a normal distributed population, calculations may be based on the fact that :

$$t_{\alpha/2,n-1} = \frac{\bar{x} - \mu}{s / \sqrt{n}} \qquad (4.12)$$

is a value of a random variable having the Student t-distribution with n-1 degrees of freedom and s is estimated standard deviation. The required sample size may then be expressed

$$n = \frac{N \cdot t_{\alpha/2,n-1}^2 \cdot s^2}{t_{\alpha/2,n-1}^2 \cdot s^2 + E^2 \cdot (N-1)} \qquad (4.13)$$

In some situations it may be useful to use the coefficient of variation, *CV*, defined as σ/μ .

4.4.3 Regression analysis

The main objective of many statistical investigations is to make predictions, preferably on the basis of mathematical equations. *The method of least squares* or *regression analysis* is the most widely used statistical tool for analysing relationships between variables. The strength of a model can be described by the ratio R^2:

$$R^2 = \frac{regression\ sum\ of\ squares}{total\ sum\ of\ squares} = 1 - \frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{n}(y_i - \bar{y})^2}$$

R^2 gives an indication of how much of the variation in the data set that can be explained by the model. The closer R^2 is to 1, the better does the model fit the data.

5. Variability within herds

The LORAKON monitoring of radiocaesium in sheep from a grazing unit is undertaken by monitoring a sample of 8-10 animals (sum of ewes and lambs) in order to estimate the median radiocaesium concentration for the whole grazing unit. This sample is supposed to be representative for the whole herd, and consequently the sampling method (randomisation, representativeness) is critical. Observation data from a whole herd should be available to check the validity of monitoring a sample of 8-10 animals from a herd of perhaps several thousand sheep. A search through the LORAKON data resulted in two large data sets that could be useful for this purpose. In the grazing units of Baklia (Valdres, Oppland) and Vuludalen (Østerdalen, Hedmark) large groups of animals were monitored in 1987, 212 and 180 animals respectively. These data sets may be implemented in a study of statistical distribution of radiocaesium concentrations in a flock of sheep grazing in the same area.

5.1 The Baklia grazing unit

In the grazing unit of Baklia the radiocaesium concentration in 212 animals were detected in September 1987. Of these animals 81 were ewes (born in 1986 and earlier) and 131 lambs (born in 1987). Description of the monitoring data is given in Table 7.1.

Table 5.1 Description of data from Baklia, Valdres (1987).

	cases	mean ($\overline{x}$)	median	variance (s^2)	coefficient of variance($s/\overline{x}$)
ewe	81	820	781	69900	32 %
lambs	131	1170	1164	108100	28 %
whole herd	212	1030	1001	121800	34 %

Figure 5.1 shows the distribution of the data for the whole herd and the two subgroups, ewes and lambs. There was a relatively clear trend of two subgroups in the data material, where lambs seemed to have generally higher radiocaesium activity than ewes. From the figure it appears that dividing the data material into two subgroups may enhance the approximation to normal distribution. This was examined more closely in Figure 5.2, where data for lambs, ewes and the whole herd were compared with the curve for Gaussian fit (constructed by the plotting software*)).

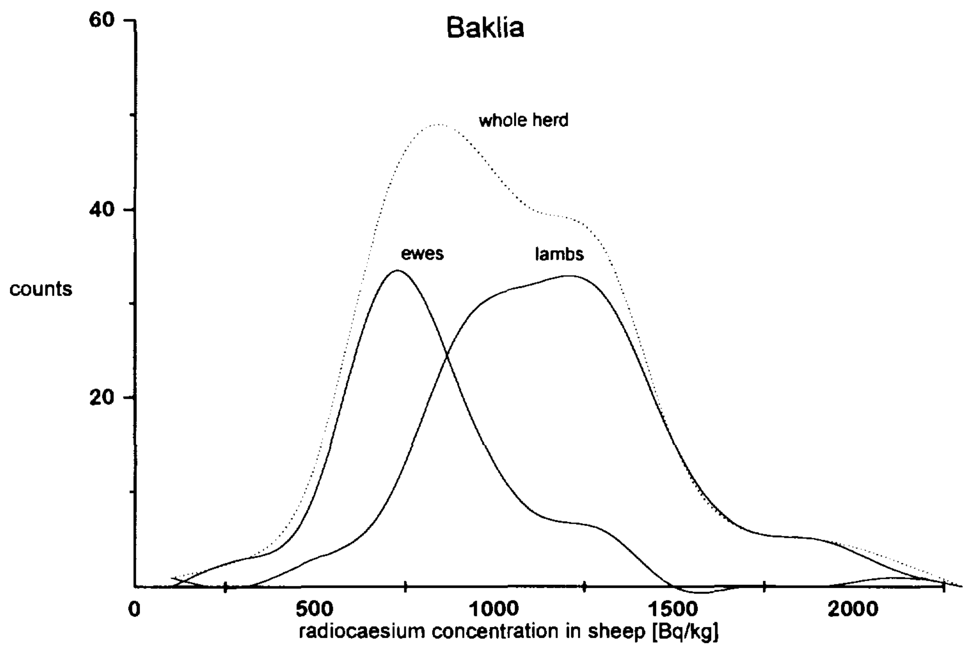

Figure 5.1: Frequency distribution for radiocaesium concentrations in 212 animals monitored in Baklia, Valdres (1987). The figure shows the distribution of ewes (81 animals) and lambs (131 animals) and the total herd. (One part of the histogram for ewes seem to have negative counts; this is caused by the adjustments in the graphics software).

The observations seem to deviate from the normal distribution at larger values, as there was a «tail» of observations showing a clear deviation from the Gaussian fit line. Data from both subgroups seemed to follow this trend.

*) ORIGIN, Technical Graphics and Data Analysis in Windows™, MicroCal Software, Inc.

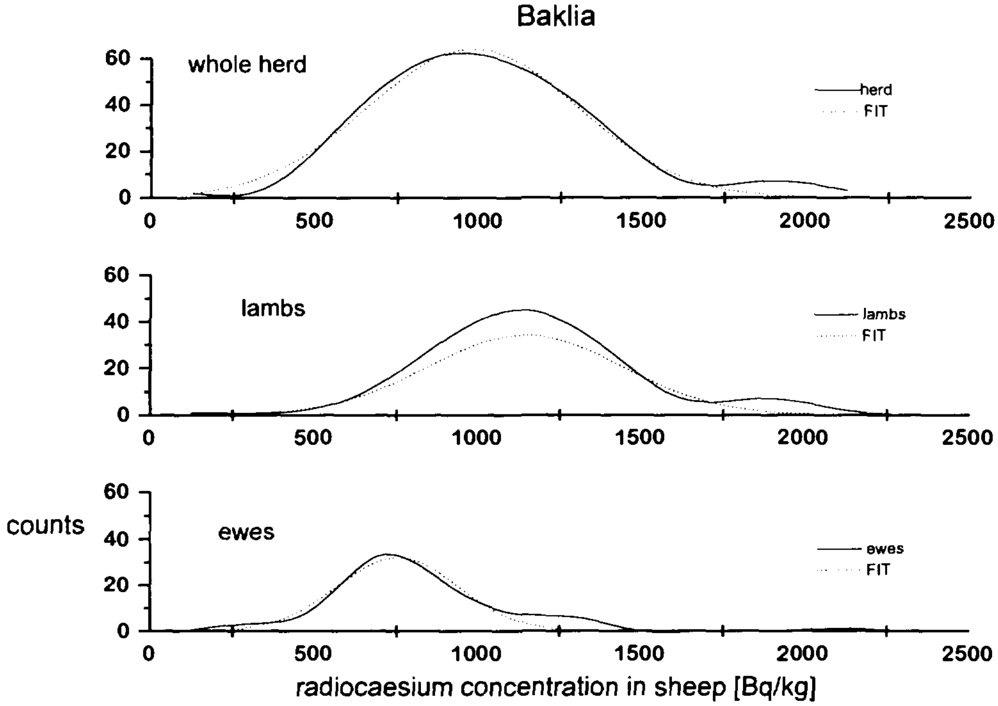

Figure 5.2: Frequency distribution for radiocaesium concentration in 212 animals (81 ewes and 131 lambs) monitored in Baklia (1987). The whole herd and its two subgroups, ewes and lambs are compared with the fitted normal curve.

In ecological and biological systems, where negative values are not possible, skewed (often approximated lognormal) distributions are common. This could also be the case for radiocaesium concentrations in sheep grazing in contaminated areas like Baklia. The data were therefore logtransformed before a new comparison with the curve for Gaussian fit was carried out. Figure 5.3 shows the transformed data and the deviation from a lognormal distribution.

The same trend was seen for the logtransformed data as for the original data. There was a deviation from the theoretical lognormal distribution for higher values, and from these plots the logarithmic transform of data does not seem to be justified. Further studies of the distributions (normal plots and detrended normal plots) are given in Appendix 2.

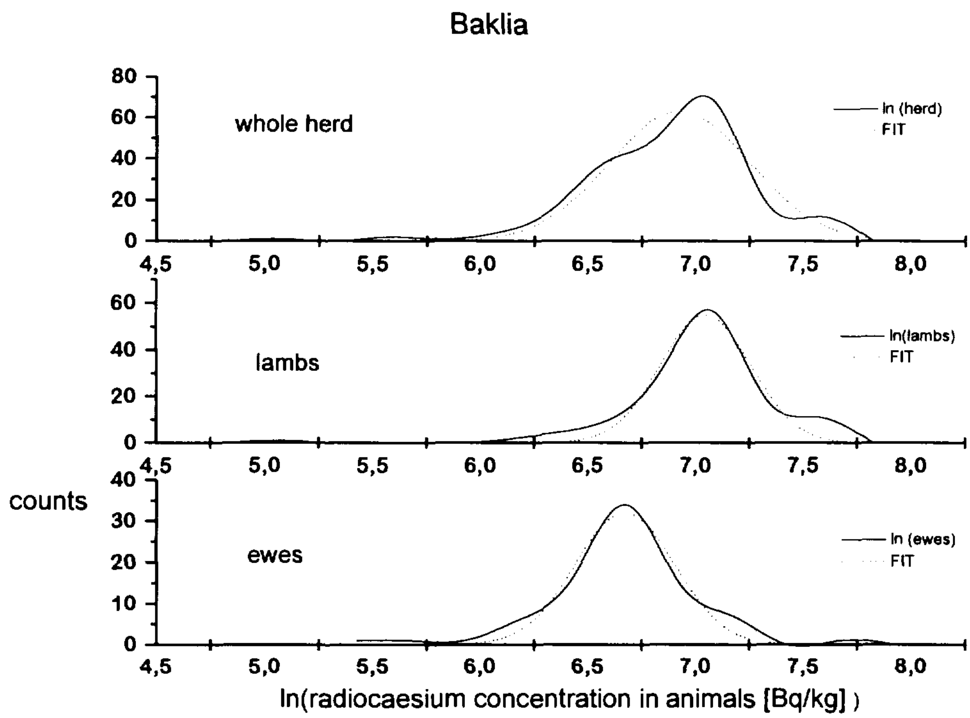

Figure 5.3: Frequency distribution for natural logtransformed experimental data of radiocaesium contents in sheep are shown for the whole herd, ewes and lambs and compared with the curve for Gaussian fit.

In summary, the data did not seem to fit a lognormal distribution any better than a normal distribution. Further analysis has therefore been undertaken with untransformed data even though the deviation from normal distribution will increase the uncertainty in the results. An analysis of variance of the observations gave the results in Table 5.2.

Table 5.2: Data from Baklia (1987)

	cases	mean ($\bar{x}$)	std.error of mean	95% conf.int. for mean	median
ewes	81	820	29	(760 , 880)	780
lambs	131	1170	29	(1110 , 1220)	1160
whole herd	212	1030	24	(990 , 1080)	1000

The analysis of variance showed a significant difference between ewes and lambs ($p < 0.001$). The median was slightly lower than the mean for all three groups, but all three median values were well inside the limits of the respective confidence intervals.

5.2 The Vuludalen grazing unit

In the grazing unit of Vuludalen, Østerdalen, 180 animals, 62 ewes (born 1986 and earlier) and 118 lambs (born 1987) were monitored in September 1987. Table 5.3 shows a description of the observations. These data were treated similarly to the Baklia-data in Section 5.1 in order to check the distribution of the observations. Figure 5.4 shows a frequency distribution of the monitoring data, both the entire herd and the two subgroups of ewes and lambs.

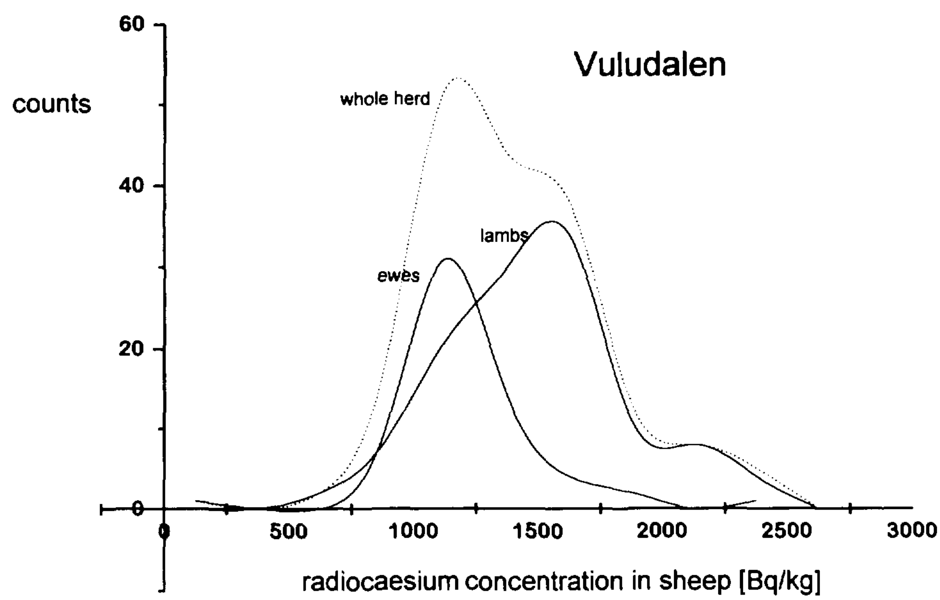

Figure 5.4: Frequency distribution for radiocaesium concentration in 180 animals monitored in Vuludalen, Østerdalen (1987). The figure shows the distribution of the total herd and the distribution for the two subgroups, ewes and lambs.

Table 5.3: Description of data from Vuludalen, Østerdalen (1987).

	cases	mean ($\overline{x}$)	median	variance (s^2)	coefficient of variation (s/μ)
ewes	62	1200	1160	83700	24 %
lambs	118	1490	1470	135500	25 %
whole herd	180	1390	1360	136300	27 %

Although the observations seemed to show more irregular distribution than the observations from Baklia the same trend as in Figure 5.1 is seen. The data seemed to be clearly divided in two subgroups and each of them seemed to be a better approximation to normal distribution

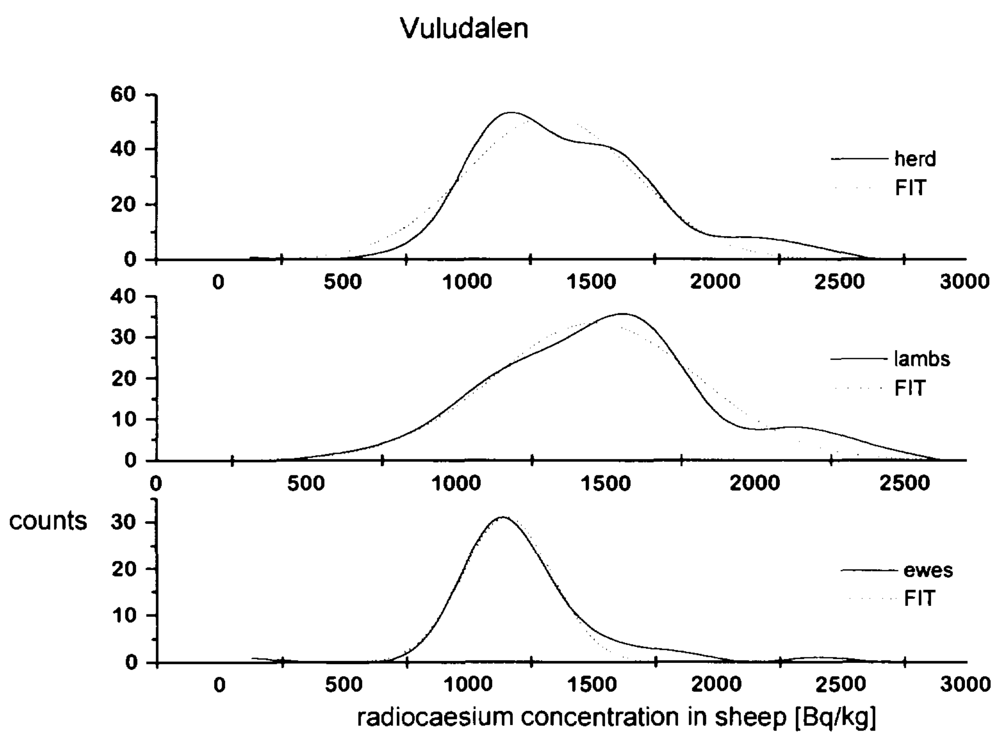

Figure 5.5: Frequency distribution for experimental data from Vuludalen (1987), showing the distribution for the whole herd, lambs and ewes compared with the curve for Gaussian fit.

than the whole herd as one group. The distribution for the three groups was studied more closely in Figure 5.4, where the frequency distribution of the observations for lambs, ewes and the whole herd were compared with the curve for Gaussian fit.

There are deviations from an ideal normal distribution, especially at higher values of radiocaesium concentrations. This may mean that the experimental data would approximate better to a lognormal distribution. The transformed data and resulting frequency distributions are shown in Figure 5.6.

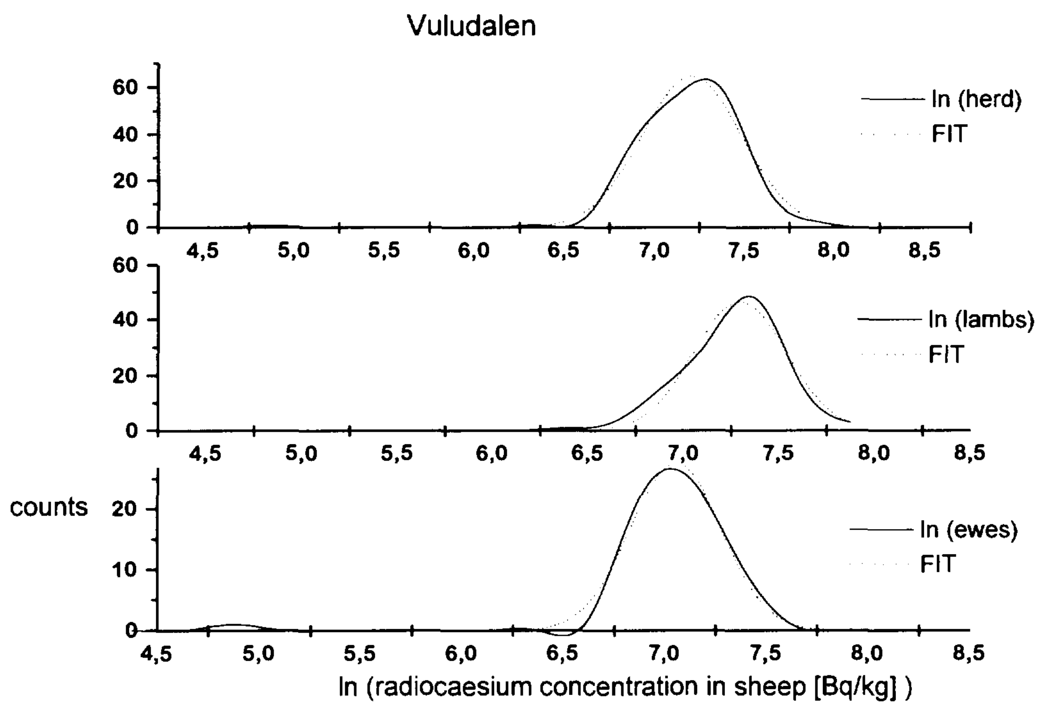

Figure 5.6: Frequency distribution of the natural logtransformed experimental data from Vuludalen (1987). The transformed data for ewes, lambs and whole herd are compared with the curve for Gaussian fit. (Parts of the distribution for ewes seem to have negative counts, this is due to smoothing of the curve in the graphics software).

Transformation of the experimental data seemed to increase the approximation to the Gaussian fit at higher values of radiocaesium concentrations. However, there was still no good approximation to a lognormal distribution at lower radiocaesium levels. The distributions have been examined more thoroughly through normal plots and detrended normal plots in Appendix 2.

Logtransformation of the experimental data did not seem to enhance the approximation to a known distribution, and analysis of variance has therefore been performed on the original observations. The deviation from a normal distribution must be taken into account when discussing the results shown in Table 5.4.

Table 5.4 Variance analysis of experimental data from Vuludalen (1987).

	cases	mean	std.error of mean	95% conf.int. for mean	median
ewes	62	1200	37	(1130, 1280)	1160
lambs	118	1490	34	(1430, 1560)	1470
whole herd	180	1390	28	(1340, 1450)	1360

The confidence intervals for ewes and lambs do not overlap, which means lambs have a significantly higher radiocaesium concentration than ewes. Analysis of variance indicated a clear difference between the groups ($p < 0.001$). The median in each of the three groups was slightly lower than the respective means, but well within the confidence intervals.

5.3 Sampling for the live monitoring

In a grazing unit there may be several thousand sheep. A small sample is monitored from each grazing unit, and the median of these observations set the value for radiocaesium concentration for the whole population. The monitored sheep should be selected randomly from the grazing unit in order to maximise the validity of the results. By the employment of the theory of finite sample variance (Section 4.4.2) the validity of the sampling method may be studied further.

The observations from Baklia and Vuludalen originate from monitoring two whole herds from two farmers. These observations may therefore be looked upon as examples of distribution of radiocaesium concentration in sheep in a grazing unit and be implemented in a study of sampling statistics. For both grazing units the estimated mean $\overline{X}$ and the estimated variance s^2 are known, and it may therefore be interesting to look at the sample sizes needed to attain a certain degree of precision in the estimations of the means of the populations. The theory in Section 4.4.2 is employed on the observed data from the two grazing units. The difference between radiocaesium levels in ewes and lambs is ignored, as this is the case during the live monitoring procedures.

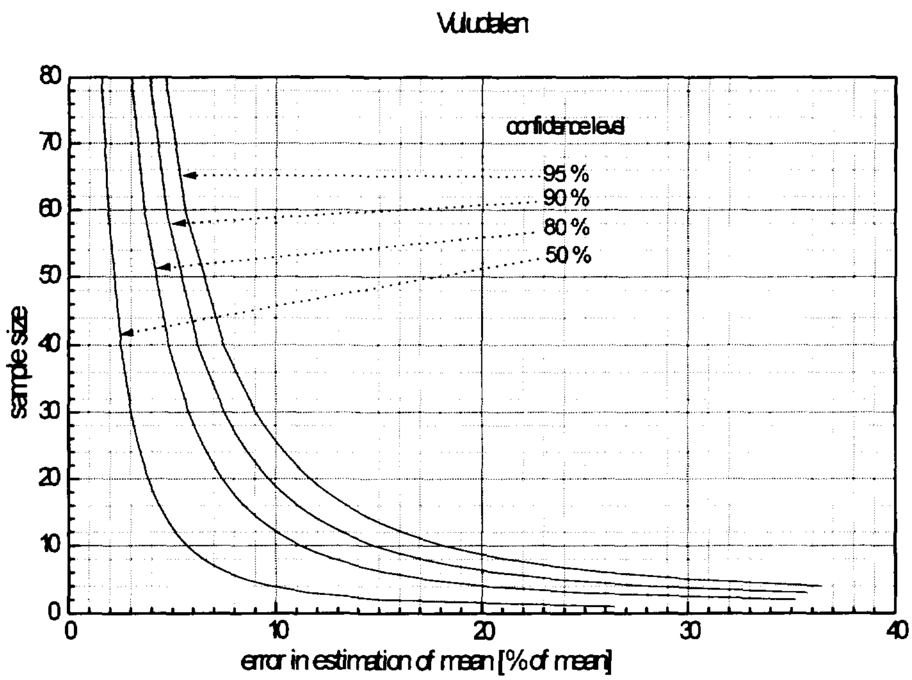

Figure 5.7: Required sample size n as a function of error (in % of mean) in estimating the mean radiocaesium concentration in the population of Vuludalen monitored in 1987.

Figure 5.7 illustrates the required sample size for estimates of mean radiocaesium concentration in the grazing unit of Vuludalen as a function of error in estimate, and for different confidence levels. If the error in the estimate of mean has to be kept below a certain value, the confidence level indicates the probability that the error actually is kept under this value.

The figure shows that the required sample size increases rapidly for small changes in estimate error below 10-15 %. If larger errors are accepted the sample size will be little affected of changes in error of estimate. The figure also shows that with a sample of 10 animals the probability for estimating the mean in this population with an error less than 10 % is about 75 %. If the error must be kept below 10 %, a random sample of 17-18 animals must be monitored to ensure the probability of this being higher than 90 %. The methods used at Vuludalen have also been applied for Baklia with similar results.

The results in Figure 5.7 are only valid for the known population of Vuludalen. A more general sampling strategy may be evolved by implementing the coefficient of variation CV (σ/μ). Studies have indicated that despite the movement in the average activity in a flock, and the wide changes in the activity of individuals, the distribution of the coefficient of variation remained essentially constant, which means that the error is proportional to the value of the results [48]. From the Baklia and Vuludalen data sets a general variation coefficient valid for other sheep herds of 0.3 (30 %) was estimated. An expression for the required sample size n is calculated from Eq. 4.13 :

$$n \geq \frac{t_{\alpha/2,n-1}^2 \cdot CV^2 \cdot N^2}{t_{\alpha/2,n-1}^2 \cdot CV^2 + E^2 \cdot (N-1)} \tag{5.1}$$

According to this expression the sample size will depend on the confidence level, the accepted error in the estimate of mean and the size of the total population. In Figure 5.8 the confidence level is set to 90 %, and the sample size is shown as a function of error in estimation of mean (for different population sizes).

The figure indicates that the population size does not significantly affect the sample size if an error of more than ca. 10 % of mean is accepted. If an error of less than 5 % is desired this requires a large number of measurements, 65-100 animals, depending on the population size. Small decreases in the error require large increases in sample size if an error below 7-8 % is required.

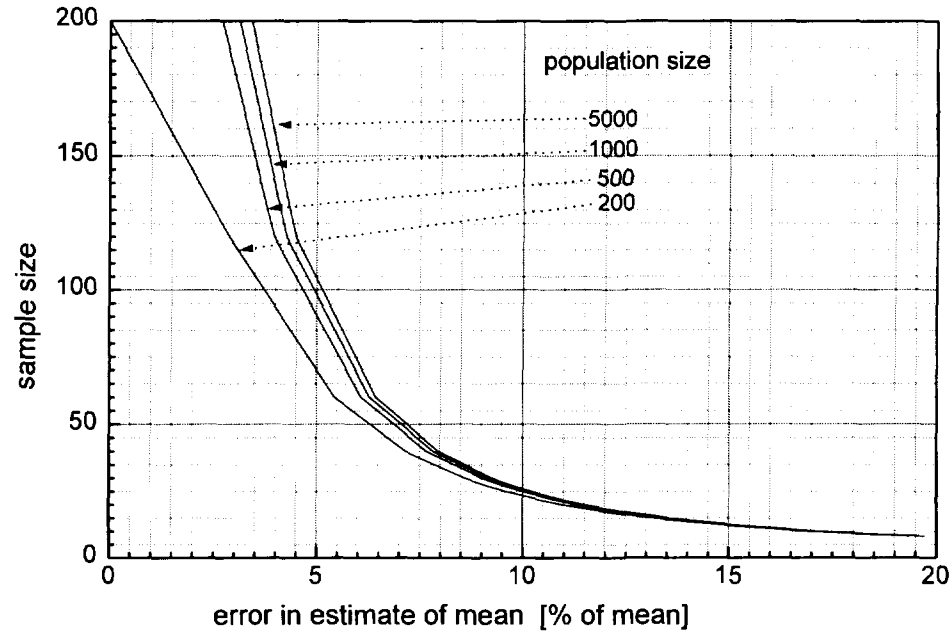

Figure 5.8: Required sample size n as a function of error in estimation of mean (in % of mean) for different population sizes. The confidence level is set to 90% .

This analysis shows that the present live monitoring procedure with selection of 8-10 animals, will at the 95 % confidence level estimate a mean value within approximately 25 % of the true mean (assuming that the median is close to the mean, which is the case for normal distributed data). Thus, when observing a level of 600 Bq/kg this is really a level somewhere within the interval 450-750 Bq/kg. In this study the sample is assumed to be selected randomly from the whole grazing unit. This may not be the fact for real sampling of sheep, as convenience may suggest that animals grazing in specific areas are monitored.

6. Long term behaviour of radiocaesium in sheep

In order to study the long term behaviour of radiocaesium in sheep grazing in mountain areas in Norway the monitoring records from the LORAKON project were examined for suitable data sets. As long term behaviour in environments where countermeasures had not been used was the main interest, data from grazing units where countermeasures had been implemented one or more years could not be used. Information about fungal production from the areas to be studied was also required.

Out of 200 grazing units in Hedmark and Oppland counties only data from two met the requirements as listed in Section 4.2; Fåset (Tynset) and Fonnåsfjellet (Rendalen) in Hedmark. In both areas animals had been monitored every year since 1987, and no countermeasures had been implemented at any time. As a study of fungi is carried out in the nearby grazing unit of Elgvasshø, information about fungal appearance for the full period was available. Figure 6.1 shows the location of the grazing units chosen for data analysis.

For comparison the grazing units Spekdalen and Grøtting are included in the analysis. At Grøtting data from four years of summer monitoring in July were available. These observations may be useful in a study of long term behaviour of radiocaesium when the influence of fungi is minor, since fungi are assumed to have little effect on the radiocaesium concentration in sheep in July. In the grazing unit of Spekdalen countermeasures have been implemented every year since 1990. The use of countermeasures would be expected to increase the variance of radiocaesium in sheep since animals use salt licks to different extents. It may therefore be interesting to compare the observations of radiocaesium concentration in sheep grazing in Spekdalen with the observations from Fåset and Fonnåsfjellet, where no countermeasures have been used.

As Figure 6.1 shows, Grøtting and Spekdalen are located near Fåset and Fonnåsfjellet.

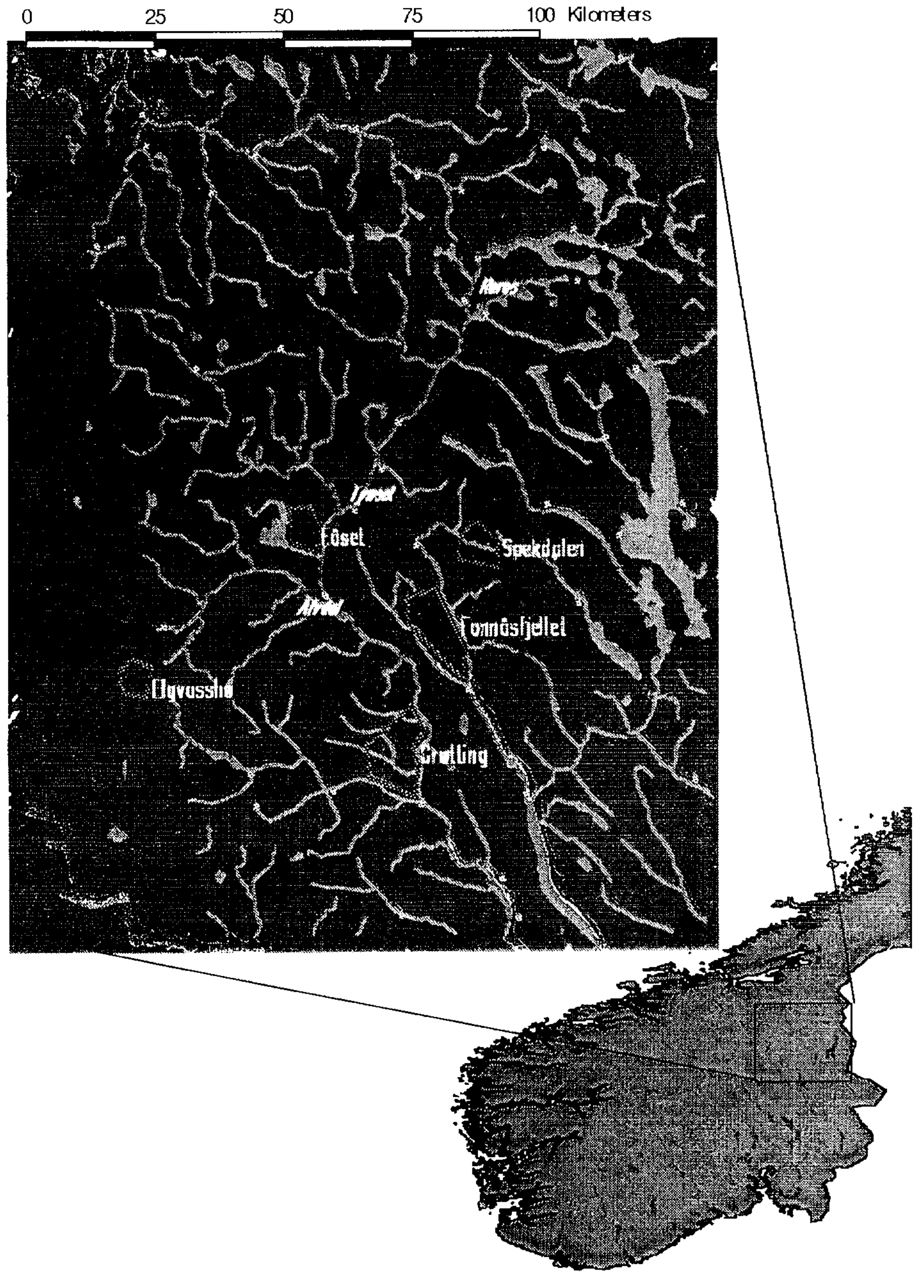

Figure 6.1 : Location of the grazing units studied.

6.1 Data description

The two data sets from Fåset and Fonnåsfjellet consist of altogether 209 measurements from 9 years, 120 lambs and 89 ewes. Monitoring data from 1986 were not included because the radiocaesium concentration in vegetation this year partly was due to direct contamination. Figure 6.2 shows the observations for the whole period.

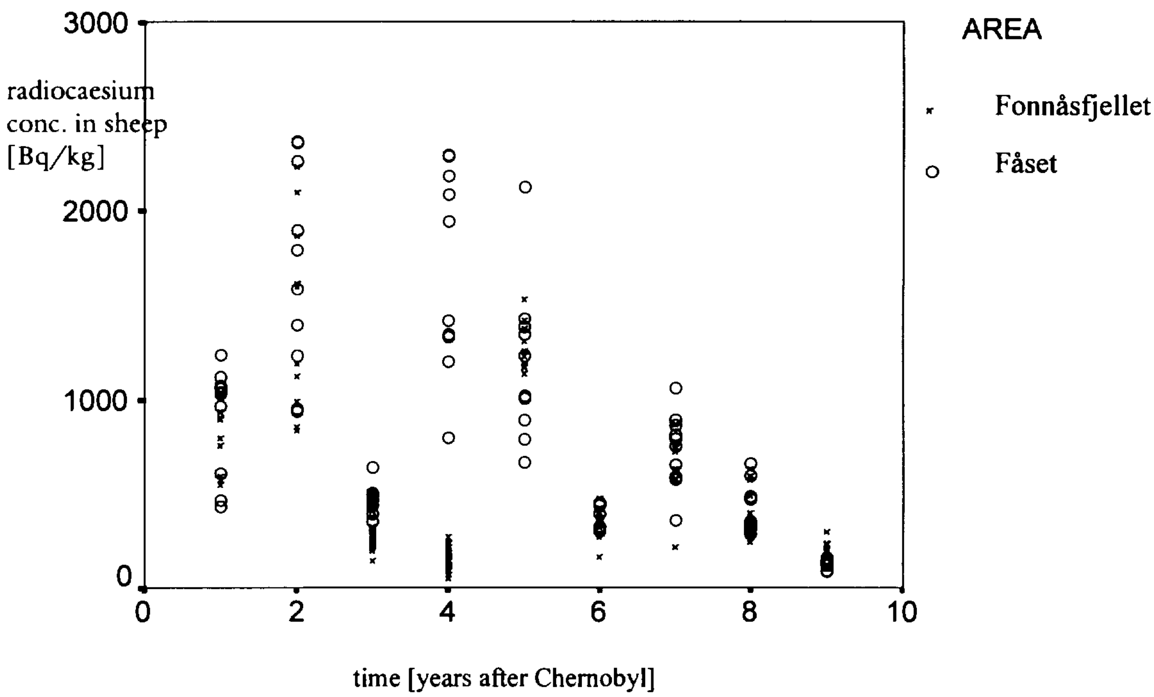

Figure 6.2 : Scatterplot of experimental data from the grazing units of Fåset and Fonnåsfjellet.

The grazing units of Fåset and Fonnåsfjellet are geographically separated, and also separate from the area from where fungal information was derived. A Student t-test showed a significant difference in radiocaesium concentrations in sheep from the two locations ($p<0.001$). However, as other factors were considered (year after Chernobyl, age of animal), variance analysis showed that this difference was no longer significant. When the data were put into the model (4.6) in Section 4.3 the difference between areas was still not significant, neither was the difference between lambs and ewes. These factors were therefore not taken into account during further data analysis. The observations from both areas show the same trend from year to year and the two data sets are therefore analysed as one group.

Alternatively the observed concentration levels could have been normalised to deposition, but deposition information is too scarce to make reliable estimates [2].

The observations are plotted against time on pasture in Figure 6.3.

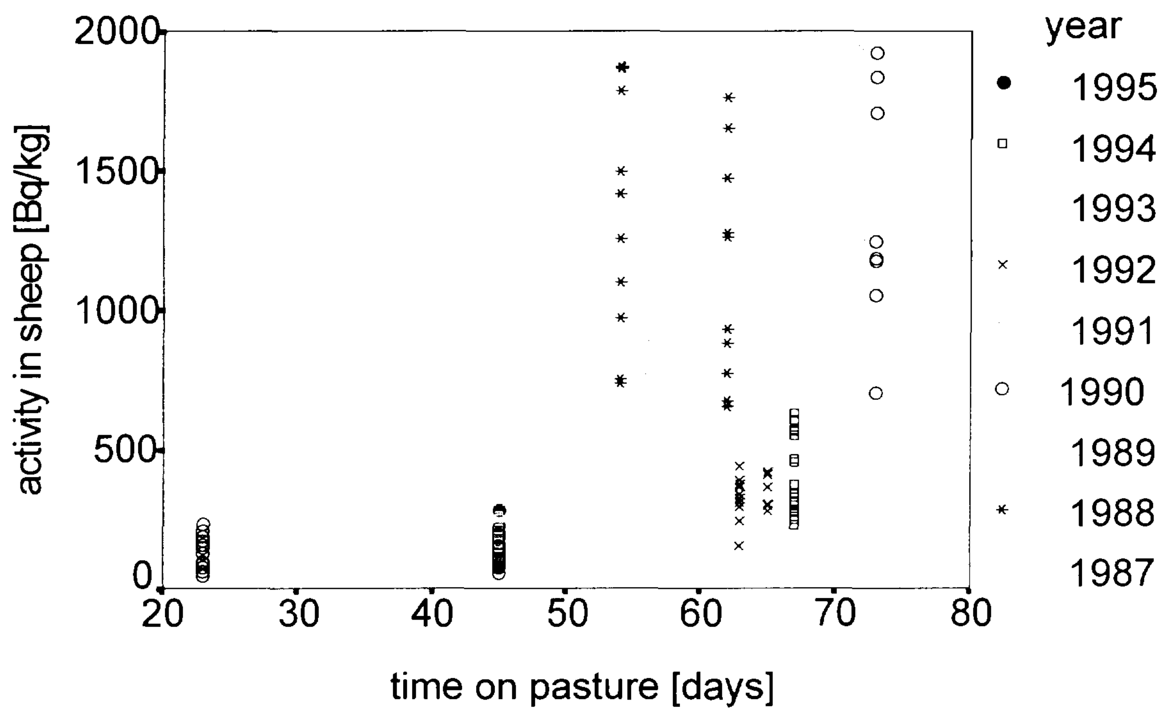

Figure 6.3: Scatterplot of experimental data from Fåset and Fonnåsfjellet as a function of days on pasture (assumed start of grazing season is 01.07.).

6.2 Modelling the long term behaviour

There are several factors not taken into account in model 4.6. The fact that the data are derived from two separate areas increases the uncertainty, and the difference in biological half life for radiocaesium between lambs and ewes is not considered in the model. Furthermore, it is assumed that the fungal fruit bodies arrive at the same time every year. The radiocaesium concentration in vegetation is assumed to follow an exponential decrease over years, while for sheep on pasture it has been assumed that the radiocaesium concentration increases and then stabilise at an equilibrium level determined by the radiocaesium concentration of the vegetation.

The data from the Fåset grazing unit in 1990 have been questioned due to possible use of Prussian blue. Further analysis is therefore performed without these data.

6.2.1 The total sample population

The model in Eq 4.6 allowed for decreasing radiocaesium concentration in fungal fruit bodies through the term $e^{-\lambda_{fungi} \cdot t_c}$, although different studies have found this to be insignificant [36], [49]. The regression analysis showed that λ_{fungi} was non-significant in the present data set. All further analysis is therefore performed without this term, thus reducing the model complexity to:

$$RC\ (t_c, t_p, F) = C \cdot e^{-\lambda_{eff} t_c} + B \cdot F \cdot (1 - e^{-\lambda_{bio} \cdot t_p}) \qquad (6.1)$$

Fungal fruit bodies are to some extent present during the whole summer. August is generally thought to be the main season, and the largest quantities of fruit bodies seemed to be found at the end of August and beginning of September in the nearby study area. This indicates that it is difficult to estimate the exact time for fruit body appearance as input to the model.

Table 6.1 shows the regression results for three different values of «time zero», the time for fruit body appearance. The model is run with 01.08, 10.08 and 15.08 as estimates for the time for fruit body appearance. The table also shows the relative importance between the three levels of abundance of fungi (see Eq.4.7), which must be determined for the different models.

Table 6.1 : Parameters in the regression model for long term behaviour of radiocaesium in sheep.

Fruiting date	levels of fungi	parameter	estimated value	95% conf.int.	R^2	estimated T_{eff} ± std.error
		λ_{eff}	0.10	(0.02, 0.18)		6.9 ± 2.8
01.08	1: 4.3 : 10.3	C	360	(220, 500)	0.65	years
		B	150	(130, 160)		
		λ_{eff}	0.14	(0.07, 0.22)		4.8 ± 1.3
10.08	1: 4 : 9	C	490	(330, 640)	0.63	years
		B	190	(170, 220)		
		λ_{eff}	0.18	(0.10, 0.25)		3.9 ± 0.8
15.08	1: 3.8 : 8	C	620	(450, 790)	0.60	years
		B	240	(210, 270)		

Table 6.2: Parameters in the regression model for long term behaviour of ^{137}Cs in sheep.

Fruiting date	levels of fungi	parameter	estimated value	95% conf.int.	R^2	estimated T_{eff} ± std.error
		λ_{eff}	0.04	(- 0.04, 0.11)		19 ± 19
01.08	1: 4.2: 9.2	C	240	(130, 330)	0.64	years
		B	140	(120, 150)		
		λ_{eff}	0.07	(0.002, 0.14)		9.9 ± 5.0
10.08	1. 4.2: 9	C	300	(200, 400)	0.62	years
		B	170	(150, 190)		
		λ_{eff}	0.10	(0.03, 0.16)		7.1 ± 2.3
15.08	1: 4.4: 9.1	C	380	(270, 490)	0.60	years
		B	180	(160, 210)		

In the Chernobyl fallout the ratio between the isotopes ^{134}Cs and ^{137} Cs was about 1 : 2 [2] . Since ^{134}Cs has a short T_{phys} (2.0 years), the observed decrease in radiocaesium concentration is partly due to the physical decay of this isotope. Table 6.2 shows the same results as Table 6.1 after correction for physical decay of ^{134}Cs. All further analysis was performed using the ^{134}Cs-decay corrected data, and the results thus gives the effective half life for the radiocaesium isotope ^{137}Cs.

Even though the data based on 01.08 gave the best model fit (R^2 = 0.64) the parameter λ_{eff} was not significant. The three levels of fungi abundance were relatively constant in the three cases, and the uncertainty in these estimates of relative amounts of fungi justified the adoption of only one ratio. The results from using the relative ratios 1: 4 : 9 are given in Table 6.3.

Table 6.3: Parameters in the regression model for long term behaviour of ^{137}Cs in sheep (with the adopted fungi ratios 1:4:9 estimating low, medium and high abundance).

fruiting date	parameter	estimated value	95% conf.int.	R^2	estimated $T_{eff} \pm$ std.error
	λ_{eff}	0.03	(-0.04, 0.11)		21 ± 23
01.08	A	220	(130, 320)	0.64	years
	B	140	(130, 160)		
	λ_{eff}	0.06	(-0.003, 0.16)		10.8 ± 5.7
10.08	A	290	(190, 400)	0.62	years
	B	170	(150, 190)		
	λ_{eff}	0.09	(0.03, 0.15)		7.8 ± 2.6
15.08	A	370	(260, 480)	0.60	years
	B	190	(160, 210)		

The estimated effective half lives were slightly longer in the case of one fixed fungi ratio, but the results in Table 6.3 were not significantly different from the results in Table 6.2. The values for R^2 are similar, indicating that the adoption of one fungi ratio did not affect the model fit. However, λ_{eff} was not significant in the case of 01.08 and 10.08 as time for fungi appearance. Due to this 15.08 was selected as the date for arrival of fungi, even though this gives the lowest model fit (R^2 = 0.60). The resulting model for describing the ^{137}Cs concentration in sheep is :

$$RC\,(t_c, t_p, F) = 370 \cdot e^{-0.09 \cdot t_c} + 190 \cdot F\,(1 - e^{-0.03 \cdot t_p}) \qquad (6.2)$$

6.2.2 *Splitting in ewes and lambs*

Age of animals was not a significant factor when added linearly into the model. Radiocaesium has a shorter elimination rate of 0.05 (λ_{bio}) in lambs [37], and the models will therefore differ slightly if the data are divided by ewes and lambs. The results of the regression of these data sets are given in Table 6.4. Figure 6.4 shows the distribution of ^{137}Cs concentration in ewes and lambs.

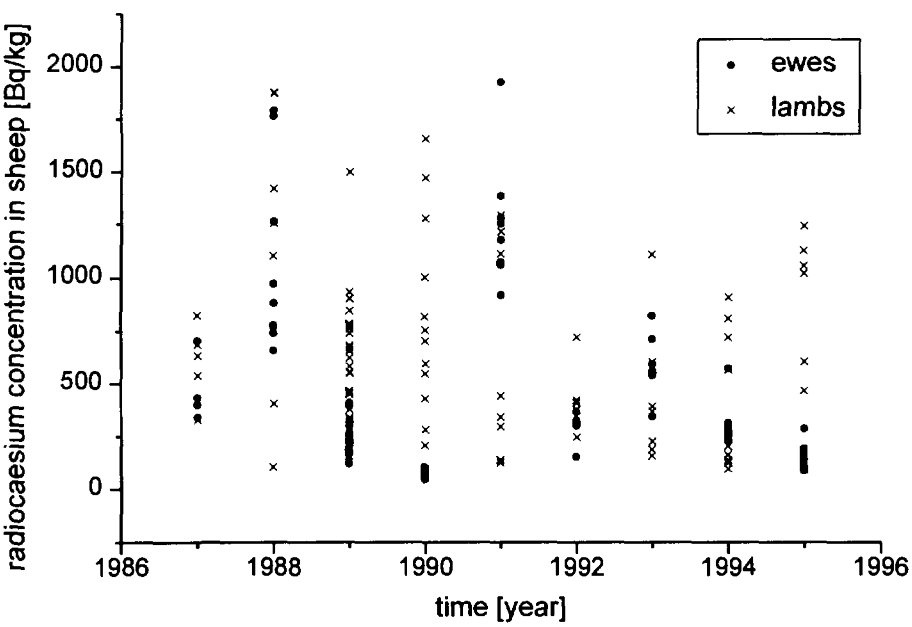

Figure 6.4: Observed ^{137}Cs concentrations in sheep, grouped on basis of age of animals.

From this figure it may appear that there is a trend of lambs having higher ^{137}Cs concentration levels than ewes.

Table 6.4: Parameters in the regression model when data sets are divided into two groups, lambs and ewes. The relative importance of the different fungi levels is set to 1:4:9 as above.

	parameter	estimated value	95% conf.int	R^2-adj.	estimated $T_{eff} \pm$ std.error
lambs	λ_{eff}	0.08	(0.01, 0.14)		8.8 ± 3.6
	C	440	(300, 590)	0.55	years
	B	120	(100, 140)		
ewes	λ_{eff}	0.08	(-0.06, 0.22)		8.4 ± 7.1
	C	240	(90, 400)	0.69	years
	B	210	(180, 240)		

Dividing the data into ewes and lambs improved the model fit to the observed data in the case of ewes while it decreased for lambs. For ewes λ_{eff} was not significant. The confidence intervals for the parameters were larger, reflecting the larger variance in the data (fewer observations compared with the whole data set). The group of ewes consisted of fewer animals (89) compared with the group of lambs (120). The estimated effective half lives were approximately the same for the two groups.

The parameter C in the model reflects the ^{137}Cs level in whilst B estimates the importance of fungi for the ^{137}Cs concentration in animals at time of slaughter. The ratio between B and C can indicate the importance of fungi as radiocaesium source compared with vegetation. The estimated values for C and B for the two groups in Table 6.4 indicate that fungi has more effect on the radiocaesium concentration levels in ewes than in lambs. The relative importance between low, medium and high fungi levels may therefore be different for ewes and lambs.

New ratios between the fungi levels were estimated, and the results from the second regression of the two data sets are found in Table 6.5.

Table 6.5: Parameters in the regression model for long term behaviour of ^{137}Cs in sheep (estimated different importance of fungi in the diet for lambs and ewes).

	levels of fungi	parameter	estimated value	95 % conf.int.	R^2	estimated T_{eff} ± std.err.
		λ_{eff}	0.09	(0.03, 0.16)		7.4 ± 2.6
lambs	1 : 8 : 14	C	470	(320, 620)	0.56	years
		B	75	(60, 90)		
		λ_{eff}	0.03	(-0.09, 0.15)		22 ± 41
ewes	1 : 4 : 12	C	210	(80, 340)	0.70	years
		B	160	(140, 190)		

For ewes, the standard error in λ_{eff} increased to almost 200 % when the new estimate for relative importance for fungi levels was adopted. For lambs there was no significant change in neither the effective half life nor its standard error when the ratio between fungi levels was changed to 1 : 8 : 14. The difference between lambs and ewes could also be influenced by suckling, but as mentioned in Section 3.2.2 this is not assumed significant for lambs older than 6-7 weeks.

6.2.3 Splitting by location

The difference in radiocaesium concentration between the two units of Fåset and Fonnåsfjellet was not significant when added as a factor into the model. The deposition map (Figure 3.1) indicated that the two areas received different amounts of the Chernobyl fallout, and a difference in radiocaesium contamination between the areas could therefore be expected. The data set was divided and the data from each grazing unit was analysed separately. The results are given in Table 6.6.

Table 6.6: Parameters in the regression model when the data set is analysed for each of the two geographically separated grazing units, Fåset and Fonnåsfjellet.

grazing unit	parameter	estimated value	95 % conf.int.	R^2	estimated T_{eff}± std.error
Fåset	λ_{eff}	0.12	(0.04, 0.20)	0.46	5.7 ± 1.7 years
	C	670	(430, 920)		
	B	150	(100, 200)		
Fonnåsfjellet	λ_{eff}	0.04	(-0.03, 0.11)	0.75	18 ± 17 years
	C	240	(150, 320)		
	B	190	(170, 210)		

The effective half life was estimated to be longer in the unit of Fonnåsfjellet (18 years) compared to Fåset (5.7 years). The model fit for Fonnåsfjellet was better (0.75), but the λ_{eff} was not significant (i.e., the decrease in ^{137}Cs concentration in sheep due to decreased concentration in vegetation was not significant).

6.2.4 Importance of ingested fungi

The year 1988 was a special year as regards fungi. Especially large quantities of fruit bodies were observed from early summer and caused high radiocaesium levels in grazing animals [4]. The importance of fungi may therefore be better estimated by introducing a fourth abundance level of fungi accounting for the special situation in 1988. Since the appearance of fungi was important in this case the three estimates of time for fungi appearance from Section 6.2.1 were reintroduced. The results from the regression analysis are shown in Table 6.7, where the ratio for levels of fungi indicate the relative importance of a bad, medium, good, and very good year for fungi.

Table 6.7: Parameters in the regression model when a fourth level of fungi is introduced in order to estimate the special year of 1988.

	levels of fungi	parameter	estimated value	95 % conf.int.	R^2	estimated $T_{eff} \pm$ std.err
		λ_{eff}	0.10	(0.00, 0.19)		7.2 ± 3.6
01.08	1: 3.8 : 5.1 : 7.5	C	220	(120, 330)	0.68	years
		B	220	(200, 240)		
		λ_{eff}	0.05	(- 0.01, 0.11)		13.1 ± 7.3
10.08	1: 4: 7.8 : 14	C	290	(200, 380)	0.68	years
		B	190	(175, 214)		
		λ_{eff}	0.05	(- 0.01, 0.10)		15.1 ± 9.5
15.08	1: 4.8 : 9.3 : 17.6	C	290	(200, 380)	0.68	years
		B	160	(140, 170)		

The parameter λ_{eff} was not significant in the case of four abundance levels of fungi. The model fit was however better than the case with three abundance levels of fungi. When time for fungi appearance is estimated to 01.08 the product $B \cdot F$ for the fourth abundance level of fungi is lower than the estimated time is 15.08. This indicates that delaying the estimate for time for appearance of fungi from 01.08 to 15.08 enhances the importance of the fourth level of fungi on the radiocaesium concentration in sheep. In contrast to the situation with three levels of fungi abundance, the estimated effective half life for ^{137}Cs is longer in the case of 15.08, but λ_{eff} was non-significant in all three cases.

6.3 Analysis based on summer monitoring

In some areas there has been an extended control of radiocaesium in sheep. In an attempt to predict the radiocaesium concentrations later in the season (and the need for countermeasures) an additional monitoring has taken place in some grazing units during July. In several of these so called «observation units» Prussian blue in salt licks has been available for the animals, and the observations are therefore not fully suitable for analysis of long term behaviour (in environments with no external interruption). This is the case for most of the grazing units where July monitoring has taken place regularly since 1987, and it is difficult to find July data from non-countermeasure units that cover a longer time period.

In the grazing unit of Grøtting, Rendalen, summer observations were available for the period 1989 - 1992, and for this period no use of countermeasures has been reported. This area also lies geographically close to Fåset and Tynset (see Figure 6.1).

Since the effect of fungi theoretically should be excluded by monitoring in July the model applied was a simple exponential dependence on time after Chernobyl:

$$A(t_c) = C \cdot e^{-\lambda_{eff} \cdot t_c}$$

The results are shown in Table 6.8 while Figure 6.5 shows the distribution for the July observations from 1989 to 1992.

Table 6.8: Parameters in the regression model for long term behaviour of ^{137}Cs in sheep, regression based on July observations in the grazing unit of Grøtting.

parameter	estimated value	95 % conf. int.	R^2	estimated $T_{eff} \pm$ std.error
λ_{eff}	0.29	(0.21, 0.37)	0.54	2.4 ± 0.4
C	760	(510, 1020)		years

The effective half life was shorter compared to the results obtained for different variants of the model in 6.2, and the uncertainty in the estimated λ_{eff} was lower than earlier models, but the model fit was only 0.54.

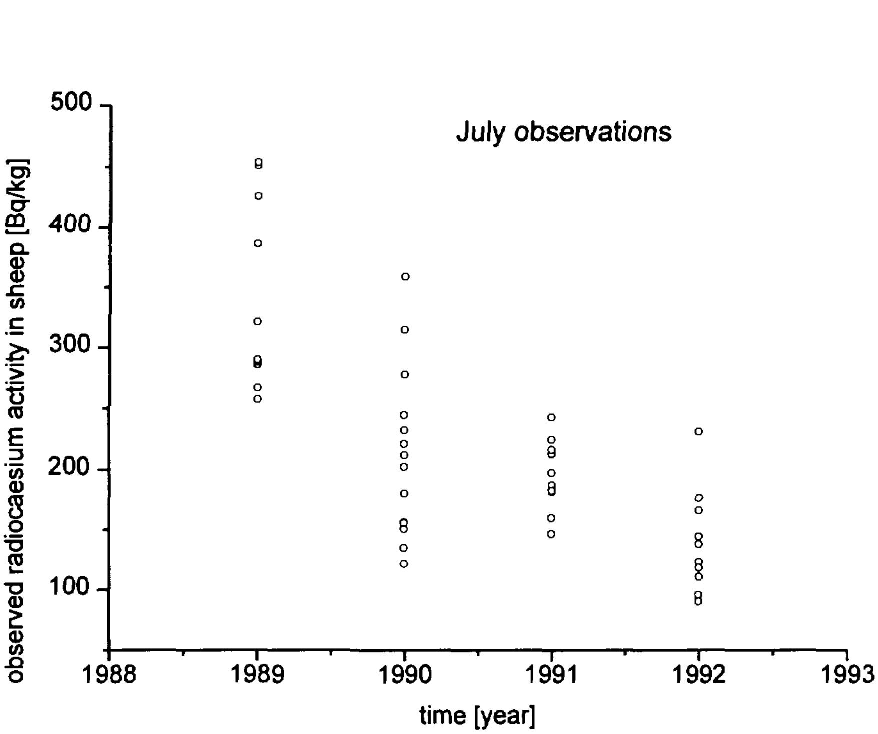

Figure 6.5 : July monitoring : Observed ^{137}Cs activity in sheep from the grazing unit of Grøtting.

6.4 Influence of countermeasures

Prussian blue has been implemented in the most contaminated areas, mainly in the form of AFCF in salt licks. As a demonstration of the effect of use of this countermeasure on the long term behaviour of radiocaesium the model (6.1) was run with the monitoring data from the grazing unit of Spekdalen, Rendalen, which lies in the same mountain area as Fonnåsfjellet (Figure 6.1). Prussian blue as additive to salt licks has been used regularly in this area since 1989. The small geographical distance between Spekdalen and Fonnåsfjellet may justify use of the same information on fungi abundance as in Section 6.2. Figure 6.6 shows the distribution of ^{137}Cs concentration in sheep grazing in Spekdalen for the time period 1987 - 1995.

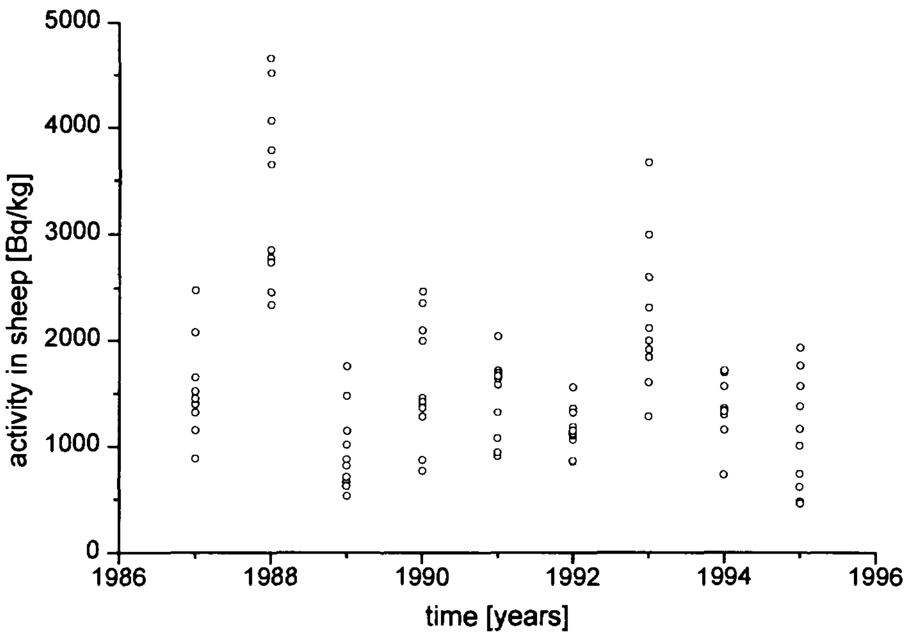

Figure 6.6: Data from Spekdalen where Prussian blue has been in use since 1989.

From Figure 6.6 it is hard to find any decrease in ^{137}Cs concentration in sheep during the observation period. Trying to fit these observation data into the model the analysis returns a significant negative λ_{eff} (which means that ^{137}Cs concentration due to vegetation shows an increase with time), and a lower model fit ($R^2 = 0.08$). At the same time the effect of fungi is negative and the regression returns a high value of C, indicating a high contamination level at the beginning of the period.

The coefficient of variance was estimated for the observations from Spekdalen, Fåset and Fonnåsfjellet for each year in the period 1989-1995 (the time period where countermeasures have been implemented in Spekdalen). In Spekdalen the estimated mean CV for this period was 31 %, which was not significantly higher than the estimated mean CV for Fonnåsfjellet (29 %) and Fåset (23 %) for the same time period.

7. Discussion

7.1 Estimating the radiocaesium level in a grazing unit

The LORAKON live monitoring aims at estimating the average radiocaesium concentration in sheep from a grazing unit. This estimate will have two principal sources of error:

1) Radiocaesium distribution in the grazing unit
2) Error in the monitoring method

These two sources are discussed below.

The variations in the radiocaesium concentration in sheep grazing on unimproved pasture may have several causes. First, wind direction and precipitation the two weeks after the Chernobyl accident together with local variations in the terrain caused large variations in deposition of the Chernobyl fallout, even within a few m^2 [2]. The variation in deposition is reflected in vegetation levels and results in potentially large variation in radiocaesium concentrations in vegetation in different areas. There are also local variations in soil type, which due to differences in fixation potential affects the availability for uptake of radiocaesium in vegetation with consequent variation in radiocaesium concentration in pasture vegetation. In addition there is variation in ability to accumulate radiocaesium between different vegetation types, and between plants, fungi and lichen. Variation in contamination level in vegetation may then be reflected as variation in radiocaesium level in sheep grazing in different areas and with different spatial or dietary preferences feeding, but sheep movement may also integrate and even out local differences in radiocaesium concentrations in soil and vegetation.

The figures in Section 5.1 revealed a «tail» of both lambs and ewes with higher radiocaesium concentrations. This might have been an effect of some of the factors mentioned above; one interpretation is that these were a group of ewes together with their lambs residing in a local deposition or vegetation «hot spot».

Radiocaesium concentration in sheep seemed to follow a normal distribution according to the observations in Baklia and Vuludalen. This assumption was confirmed by dividing the observations into two subgroups based on ewes and lambs, and the standard deviation decreased for both groups compared to the standard deviation for the whole herd. However, these results based on observations from two areas in 1987 cannot be transferred to other areas and later years without reservations. Both Baklia and Vuludalen are situated in areas that received high Chernobyl deposition. Since negative values of radioactivity concentration are not possible, the distribution will be more skewed as the concentration of radiocaesium in animals decreases. This is illustrated in the observations from the grazing units of Fåset and Fonnåsfjellet in Figure 6.2, where a cluster of observations at low levels were seen for years with general low radiocaesium levels. High levels in animals due to for instance intake of vegetation grown on «hot spots» will always cause a tail of observations at higher levels and therefore a skewed distribution.

Live monitoring of sheep have indicated that age of animals does significantly influence the radiocaesium concentration in animals at the time of slaughter[48], and lambs and ewes have therefore been considered as one group in the live animal monitoring. However, the observations from the large data sets from Baklia and Vuludalen show a significant difference in radiocaesium concentration between ewes and lambs in the September monitoring, with generally higher radiocaesium levels in lambs than in ewes. One source of uncertainty may be the geometry factor for lambs, which decreases from 32 to 25 as the lamb grows. The instruments used in the live monitoring are calibrated with one geometry factor for ewes and one for lambs prior to the measurements. During one monitoring session lambs of different size will be monitored, but it is unlikely that the geometry factor will be changed accordingly. The geometry factors implemented in the Baklia and Vuludalen monitoring in 1987 were 30 for lambs and the common 25 for ewes. If this factor was used for all lambs regardless of their size, the radiocaesium concentration in bigger lambs might be overestimated, and therefore contribute to the higher radiocaesium levels found in lambs. The use of the same geometry factor on lambs of different sizes is probably also reflected in the width of the distributions in

Figs 5.1 and 5.4; the lamb distributions are wider than those for ewes (the latter being likely to be more homogenous in size).

Monitoring lambs only may be recommended since lambs have generally higher radiocaesium concentrations which leads to lower uncertainty due to counting statistics in the observations.

Large and variable differences in radiocaesium concentration have been shown by individual sheep[48] within a flock. Individual preferences in grazing behaviour and movement patterns are considered partly responsible for this variability within herds. In addition, as there are no boundaries between the areas used by different grazing units, animals may graze in areas associated with other grazing units. According to the local variation in radiocaesium in vegetation, this may increase the variation in radiocaesium concentration observed in sheep within a herd and within a grazing unit. It is therefore of importance that animals representative for the whole grazing unit are sampled and monitored in order to estimate a mean radiocaesium concentration that reflects the true radiocaesium level in the grazing unit.

The results from the study of sample sizes indicated that in a sample of 8-10 animals selected from the population of Vuludalen in 1987 the probability that the error in the estimated mean is equal to or less than 10 % is about 75 %. For the observations from Baklia (Appendix X) the probability is even lower, since the variance in this data set was somewhat larger compared with the data from Vuludalen. The coefficient of variation (CV) in Vuludalen is ca. 27 % while it is higher, 34 %, in Baklia. In both cases the CV is higher for the whole herd than for each of the subgroups, ewes and lambs.

From the data in Baklia and Vuludalen a general coefficient of variation for radiocaesium concentration within herds was estimated as ca. 30 %. Figure 5.8 showed that when a general CV replaced the real mean and variance for the data from Vuludalen, and if an error in mean of 8-10 % was accepted, the population size had little effect on the required sample size. By implementing the assumed general CV it is possible to study the sample size required to estimate the mean of a general grazing unit with a certain precision.

Figure 7.1 shows the required sample size as a function of error in mean for different confidence levels. The CV is 0.3 and the population size is set to 1000 animals in the calculations. The probability for the error being less than 10 % is less than 75 % when a sample of 10 animals is monitored, similarly to that indicated for Vuludalen. The calculations for general populations are based on a rough estimate of the coefficient of variation, based on the herds in Baklia and Vuludalen. The mean CV for these two units had a standard deviation of ca. 3.5, or 12 %. In a situation where it is desired that as few animals as possible with radiocaesium concentration > 600 Bq/kg are slaughtered, it may seem like a higher degree of precision in the estimation of radiocaesium concentration level within a herd is worth while.

The monitoring data from Baklia and Vuludalen are based on sheep belonging to the same farmer while there are animals from several farmers in a grazing unit. Sheep are residential animals and groups of animals will often tend to reside in specific parts of a grazing area, while other sheep may stick to other areas. It is therefore possible that the data from Baklia and Vuludalen show less variation than would be the case if a sample of the same size was selected from the whole grazing unit, if it is assumed that sheep from different farmers to some extent graze in different parts of the area.

The data from Baklia and Vuludalen are assumed to be normally distributed even though the distributions of the observations showed some deviation from this. Since the sampling analysis is based on normally distributed data this will also contribute to the total uncertainty in the estimation of a population's mean radiocaesium concentration.

An estimate of the total uncertainty in the monitoring method, depending on uncertainty in the detector, the shielding factor and the geometry factor may be calculated by using Eq. 4.3. The uncertainty in the geometry factor is estimated to ca. 10 % (this may be exceeded in sheep with radiocaesium levels of less than 400 Bq/kg) [50]. The uncertainty in the shielding factor can probably be assumed to be similar since similar procedures are involved in the estimation. The uncertainty due to counting statistics is lower and an estimate based on number of counts is ca. 2 % for a radioactivity level of 600 Bq/kg (lower for higher radioactivity levels). According to Eq. 4.3 a rough estimate of the total uncertainty in each monitored level is ca.

15 %. For the mean of 10 animals this will give an error of about 5 %, which is a small contribution to the total error of the estimated average level of radiocaesium in a population.

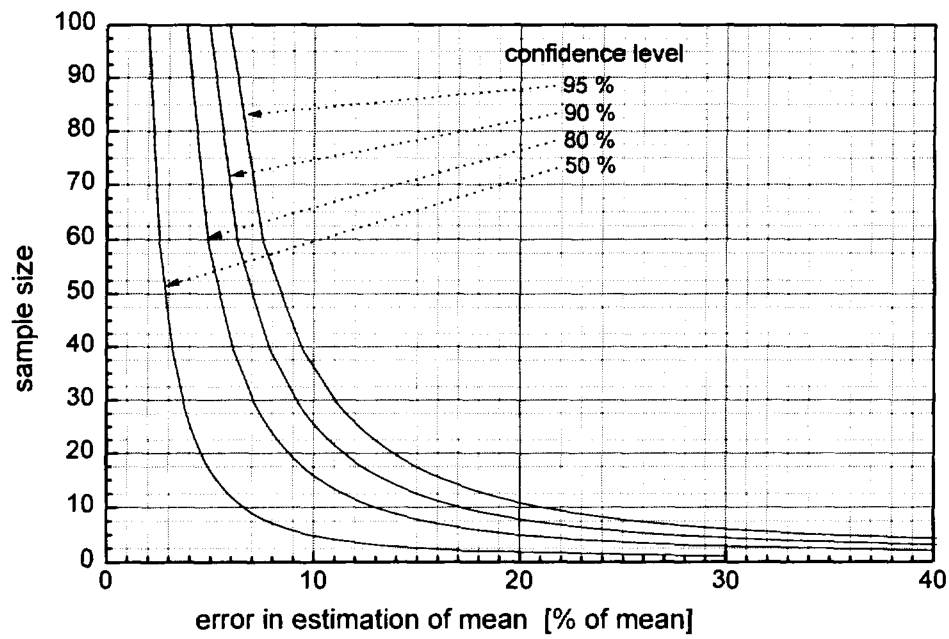

Figure 7.1: Sample size as a function of error in estimation of mean in a general population of 1000 animals.

7.2 Long term behaviour

The distribution of the observations as a function of time on pasture in Figure 6.3 show the same trend as the theoretical model for radiocaesium behaviour in sheep during a grazing season, illustrated in Figure 4.5, hence justifying the use of the second term in model 4.6. However, there are several assumptions in this model that have to be made clear.

1) The radiocaesium concentration in sheep is assumed to follow an exponential increase during the first weeks on pasture, and then stabilise at a level dependent on the radiocaesium concentration in the vegetation in the grazing area. This assumption requires

that the contamination of the vegetation is homogenous in the grazing area throughout the grazing season. However, as deposition, soil type and potential for uptake of radiocaesium by vegetation species, varies across the area. Radiocaesium concentrations in vegetation will also be expected to vary. This may be apparent over different vegetation zones (for instance forested, bogs, highland). Animal movement may to some extent even out these differences, but as indicated by the «tail» in Figs 5.1 and 5.4 there will be differences. Additionally, uptake of radiocaesium in vegetation is not constant throughout the growing season, but seems to be slightly higher in the beginning [51] . Compared to the uncertainty in the other model assumptions this factor is assumed to be of minor importance.

2) The radiocaesium concentration in vegetation is assumed to decrease exponentially with years at a rate determined by the effective ecological elimination rate λ_{eff}. An exponential decrease has been observed for individual vegetation species, but vary between species. The decrease in the average radiocaesium concentration in mixed vegetation may therefore be expected to be better estimated by more than one factor. Besides, the summer data shown in Fig. 4.3 support the assumption.

3) The date of appearance of fungal fruit bodies is assumed to be the same every year. The factor «time on pasture» in the model should ideally have been «time on pasture with fungi available as feed», or time on pasture after the appearance of fruit bodies. The time for fruiting of fungi varies from year to year but the season in the studied area is assumed to start in August. This is supported by records from NRPA's mushroom project [45] and also studies in Griningsdalen [53]. In Griningsdalen (an area in southern Norway) the date for fruiting of fungi has been observed to vary from 25.07 to 05.09. Assuming the same fruiting date every year is therefore one of the more critical factors in the model. Additionally the model assumes that the amount of radiocaesium available in fruit bodies in the pasture is constant from the first day they appear. Similarly as for vegetation, species of fungi differ in the accumulation of radiocaesium, hence the discussion on homogenous concentration in vegetation is also valid for fruit bodies. In contrast with vegetation the composition of fungi species may vary greatly between years, and presence of fruit bodies of different species may therefore be more critical (since some years substantial amounts of certain «high accumulating» species may appear). The model only considers relative amounts of fruit bodies regardless of species. Some of the factors determining fruiting of fungi are partly known (for instance temperature and precipitation), but little is known

about requirements for different species. It is therefore impractical to consider fruit bodies by other means than total amounts.

4) Ewes and lambs are studied as one group, and the difference in biological half lives is not considered in the main regression analysis (see discussion below).

Information about abundance of fungi in the observed grazing areas is adopted from the area of Elgvasshø, which is geographically separated from Fåset and Fonnåsfjellet. However, since the relative, not the absolute amount of fungi is important in this model the fungi information from Elgvasshø is assumed to be valid for the areas of Fåset and Fonnåsfjellet.

Abundance of fruit bodies at Elgvasshø was recorded at approximately the same time every year. This means some uncertainty in determining the amount of fungi since the time for and duration of appearance of fruit bodies changes from year to year. The classification of levels of fungi abundance was subjective and with no clear distinction between levels (for instance, number of fruit bodies per m^2).

Despite many assumptions and sources of uncertainty, the estimated values for radiocaesium concentration in sheep obtained by model 6.2 are in accordance with the observed radiocaesium concentrations. Figure 7.2 shows the observed mean for each year (with error bars showing the standard error of mean) compared to the estimated values. The uncertainty associated with the estimated values is not shown, but is about ± 50 Bq/kg. The underlying exponential decrease in ^{137}Cs concentration in sheep due to decreased contamination level in vegetation is shown as a dot-and-dash line. The difference between the ^{137}Cs level according to vegetation and estimated ^{137}Cs concentration in sheep is an indication of the importance of fungi for the ^{137}Cs concentration in the monitored sheep. The estimated effective half life was 7.1 ± 2.3 years.

The estimated ^{137}Cs concentration for 1987, which was a year with large amounts of fruit bodies (although not as special as 1988), was high compared with the mean of the observations that year. This may be due to the short time after the fallout. Parts of the radiocaesium could still be at the soil surface and therefore not available for uptake and accumulation in vegetation and fungi. This is supported by a German study [52] which found similar ^{137}Cs levels in autumn 1986 compared to autumn 1985 (pre-Chernobyl).

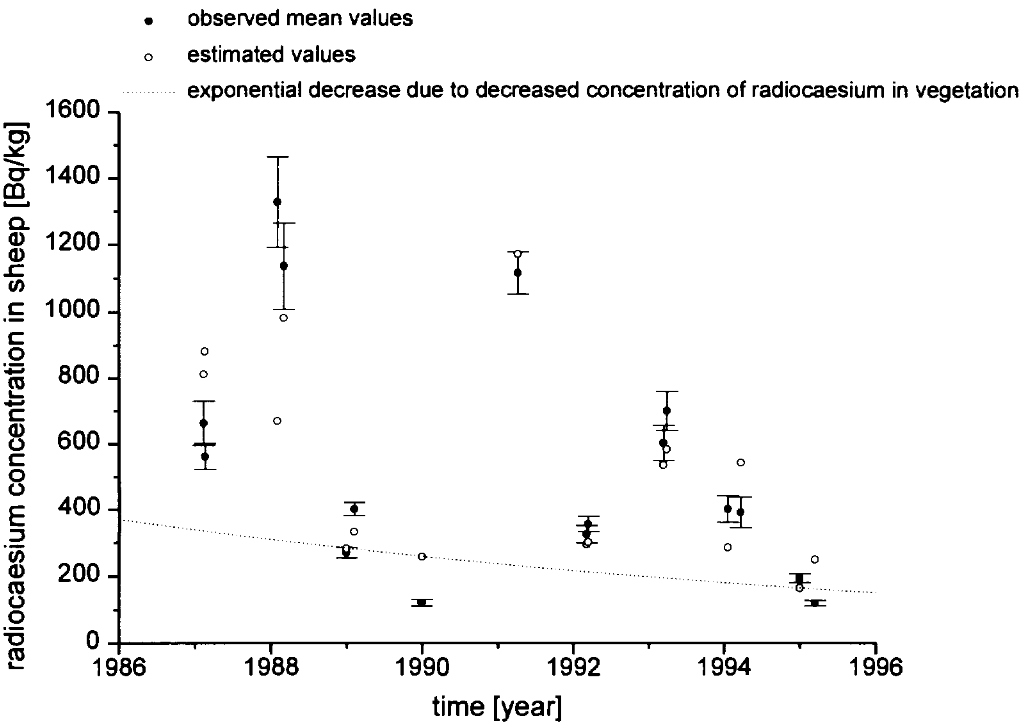

Figure 7.2: Estimated values for ^{137}Cs concentration in sheep are compared with the mean of the observations for each year ± std. error of mean. The dot-and-dash line illustrates the underlying exponential decrease in ^{137}Cs concentration in sheep due to decrease in contamination level in vegetation. Time for appearance of fruit bodies is set to 15.08, and the relative importance between the different levels of fungi abundance is 1:4:9.

Different estimates for time of appearance of fruit bodies were tested, but only 15.08 gave a significant λ_{eff}. However, if the estimated time for fungi appearance is set to 01.08 or 10.08 the regression analysis gives a better model fit, but a non-significant λ_{eff}. The non-significant λ_{eff} indicates that there is no significant decrease in ^{137}Cs available for uptake and accumulation in plants during the observed period. The estimated effect of fungi on ^{137}Cs concentration in sheep increases if fungi are assumed to appear earlier, while the importance of contaminated vegetation decreases. This increases the estimate for effective half life for radiocaesium.

Before the contribution from ^{134}Cs was corrected for, λ_{eff} was significant for all three cases, the model fit was the same, and the estimated ecological half life was shorter, as expected. The significance of λ_{eff} may indicate that the observed decrease in radiocaesium concentrations is mainly due to physical decay of ^{134}Cs, and that the bioavailability of radiocaesium in soil shows no significant decrease during the observed time period.

Splitting the observations into ewes and lambs decreased the number of observations for each group. Some years the mean ^{137}Cs concentration for ewes was calculated on basis of only 2 observations, and the standard deviation in the estimates increased thereafter. This was reflected in the large uncertainty in the estimate of λ_{eff} for ewes, giving a non-significant decrease in ^{137}Cs in vegetation. The ewe-observations fitted the model well, but that may be due to the larger standard deviation in the estimated values of the model parameters. ^{137}Cs concentration in lambs showed a significant decrease during the observation period, and an effective half life of 8.8 ± 3.6 years was estimated. The standard deviation in the parameter estimates was less than for ewes, probably because the regression was based on more observations.

The estimated half life was approximately the same for lambs (8.8 years) and ewes (8.4 years), and the difference was not significant, as expected. This indicates a realistic model, the ecological half life depends on the bioavailability of radiocaesium in the environment, and should not be affected by the age of the animal.

Figure 7.3 shows the estimated ^{137}Cs concentration level for ewes compared with the mean observed radiocaesium concentration. The estimated values show accordance with the observed radiocaesium concentration, but the large standard deviation in the estimated parameters leads to a high degree of uncertainty in the estimation of radiocaesium concentration in ewes.

Fungi may appear to be more important as radiocaesium source for ewes than for lambs. The relative importance $B_{ewes} \cdot F_{ewes}/B_{lambs} \cdot F_{lambs}$ was 1.8, independent of the values for levels of fungi abundance, and indicated that fungi is more important in the diet for ewes than for lambs.

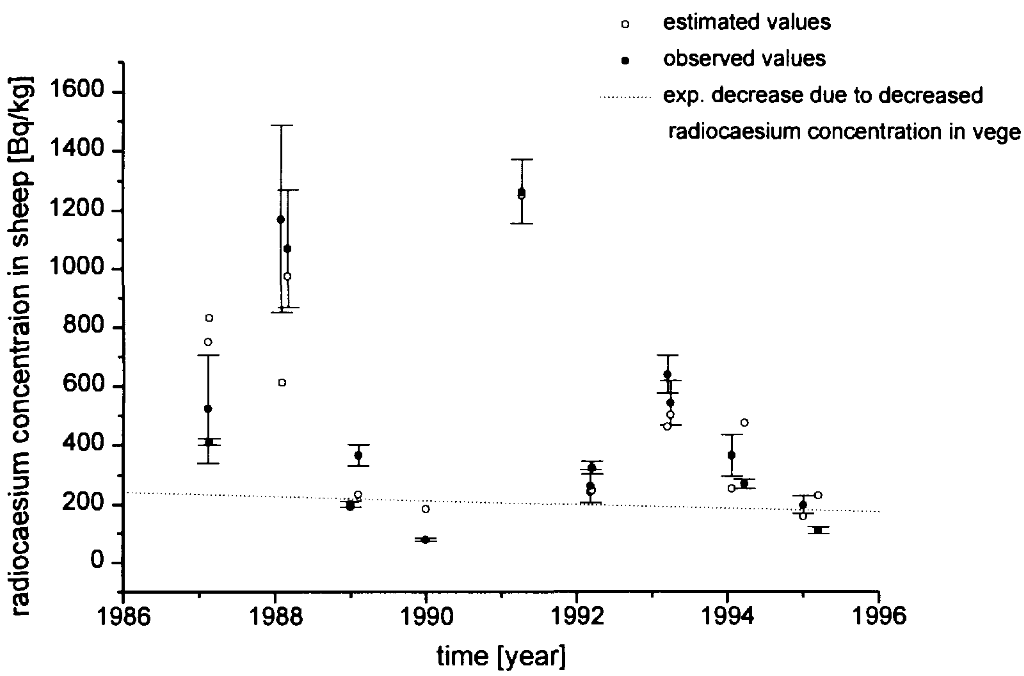

Figure 7.3: Estimated radiocaesium level in ewes compared with the mean of the observed ^{137}Cs concentrations. Error bars show the standard error in estimation of mean.

When the data from the two grazing units were separated regression analysis showed large differences between the two areas. The data from Fonnåsfjellet fitted the model better, but λ_{eff} was not significant. A long effective half life of 18 years was estimated, but the standard deviation in the estimate of λ_{eff} was more than 200 %. Effective half life for the data from Fåset was by comparison estimated as 5.7 years, indicating a significant decrease in ^{137}Cs concentration in vegetation. The difference between effective half lives for the two areas was not significant, in contrast to the estimated radiocaesium level at time 0 (parameter C) which was significantly higher for Fåset.

Figure 7.2 showed that the estimation of radiocaesium concentration level in sheep for 1988 was low compared with the observed ^{137}Cs concentrations. Figure 7.4 shows the estimated ^{137}Cs concentration in sheep when the special year 1988 was given a separate value. The ^{137}Cs level in sheep for 1988 was better estimated using this model, thus supporting the hypothesis that 1988 was a year with especially large quantities of fungi.

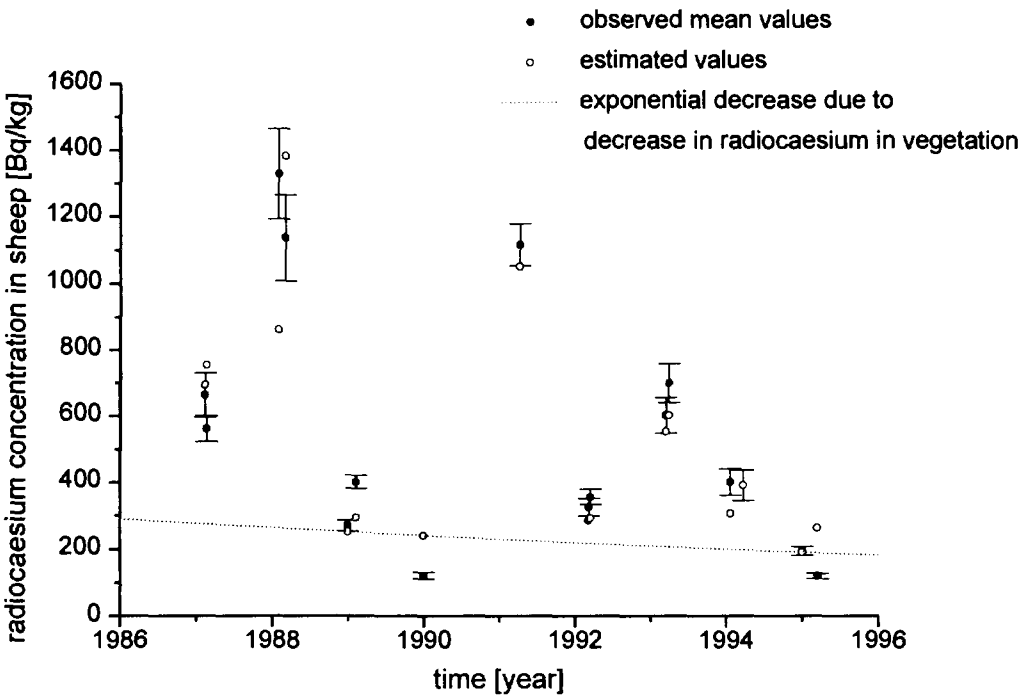

Figure 7.4 : Estimated ^{137}Cs concentration in sheep compared with the mean of the observations for each year, 4 levels of fungi abundance. The dot-and-dash line illustrates the underlying decrease in ^{137}Cs concentration sheep due to decrease in radiocaesium concentration in vegetation.

The model fit was better for 4 levels of fungi abundance, but again λ_{eff} was not significant for any of the estimated dates for fruit body appearance, 01.08, 10.08 and 15.08. The estimated effective half life was shorter when fungi was assumed to appear 01.08 than when this appearance was set to 15.08. This may be due to the enhanced importance of the fourth level of fungi in the last case.

In all variants of the model (ewes/lambs, splitting in areas, 4 levels of fungi abundance), when the uncertainty in T_{eff} increased and the estimated λ_{eff} was non-significant, the model fit increased. This indicates a non-significant decrease in the bioavailability of radiocaesium in soil, and that the changes in radiocaesium concentration in sheep to a large extent depend on the amount of fungal fruit bodies available as feed for the animals.

There are many possible ways of grouping the observations and estimating radiocaesium levels in different groups. However, dividing the data set decreases the number of observations in each group, and increases the uncertainty in the estimates. Regression analysis of the different groups might have given different results if all variations of estimated time for appearance of fungi and introduction of fourth level of fungi abundance had been introduced for each of the different groups analysed (ewes, lambs, Fåset, Fonnåsfjellet), and if relative importance of the fungi abundance had been estimated for each group. However, the relatively few observations were not assumed appropriate for further splitting.

The analysis of the July observations of ^{137}Cs concentration in sheep from the grazing area of Grøtting indicated a short effective half life of only 2.4 ± 0.4 years. The standard deviation in the estimated parameters was low and the data showed a model fit of 54 %. Figure 7.5 shows the observed and estimated ^{137}Cs concentrations in sheep.

The ecological half life estimated for the July observations at Grøtting was low compared with the estimates from Fåset and Fonnåsfjellet. The estimate for Grøtting is based on observations from only 4 years and can therefore only indicate the decrease in radiocaesium concentration during this period (1989-1992). However, this result may also indicate that the models resulting in the shortest T_{eff} in Chapter 6 are better compared with those giving longer T_{eff}, even though the model fit is lower.

Fungi is assumed to have little effect on radiocaesium concentration in animals in July, and the observed radiocaesium activity in sheep should therefore be due mainly to intake of contaminated vegetation. However, certain fungi may fruit as early as the end of June, and when animals are monitored in late July/early August, intake of fungi may have contributed to the total radiocaesium concentration observed in sheep. Another source of uncertainty is the time on pasture before monitoring; For early monitoring sessions, radiocaesium concentrations in sheep may not have reached the equilibrium level.

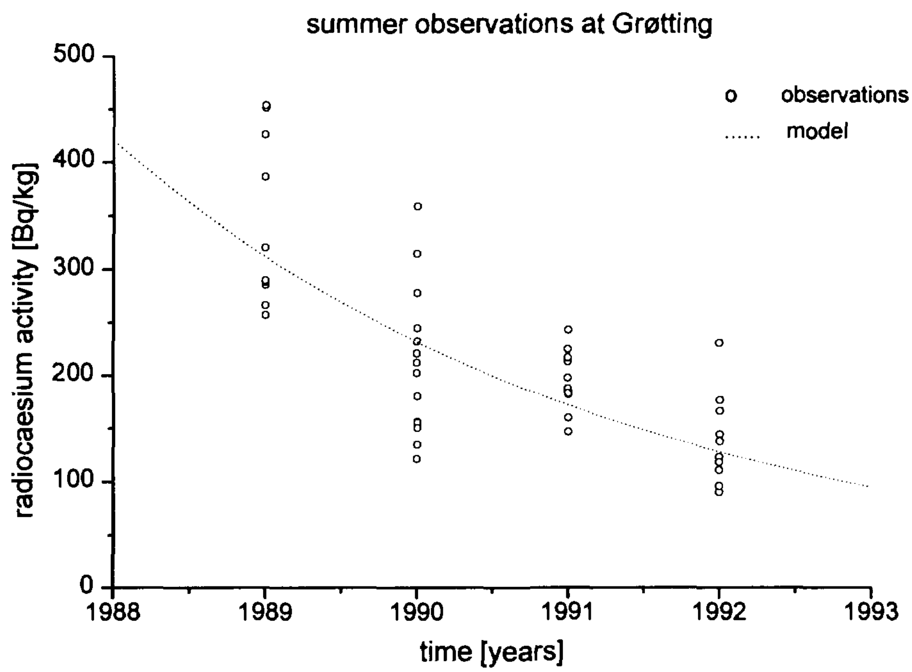

Figure 7.5: Observed ^{137}Cs concentrations in sheep from the grazing unit of Grøtting, monitored in July. The dot-and-dash line shows the ^{137}Cs concentration estimated from model 6.4.

The decreased ^{137}Cs concentration in sheep can be estimated in different ways. This is shown in Figure 7.6, where the observed radiocaesium concentration (not corrected for ^{134}Cs) are compared with the following:

1) Exponential decrease estimated on basis of the observations of radiocaesium concentration in sheep, not corrected for decay of ^{134}Cs. The observations are fit into a simple exponential model without adjustments for changes in fungi abundance from year to year. The expression is $1040 \cdot e^{-0.13 \cdot t_c}$, $R^2 = 0.13$, and the estimated effective half life is 5.3 ± 1.1 years.
2) Exponential decrease in radiocaesium concentration (not corrected for ^{134}Cs decay) when corrected for variations in amount of fungi (relative levels of fungi abundance: 1 : 3.8 : 8)

from year to year. The underlying exponential decrease in radiocaesium concentration in sheep is due to decreased concentration in vegetation, and the estimated model is $620 \cdot e^{-0.18 \cdot t_c} + 240 \cdot F \cdot (1-e^{-0.03 \cdot t_p})$, $R^2 = 0.60$. Estimated effective half life is 3.9 ± 0.8 years.

3) Exponential decrease in ^{137}Cs concentration, based on ^{134}Cs- decay corrected observations. The observations are again fit into an exponential model with adjustments for changes in fungi abundance from year to year. The decrease follows the expression $370 \cdot e^{-0.09 \cdot t_c} + 190 \cdot F \cdot (1- e^{-0.03 \cdot t_p})$, $R^2 = 0.60$ and the estimated effective half life for ^{137}Cs is 7.8 ± 2.6 years.

4) Exponential decrease in radiocaesium concentration in sheep (not corrected for ^{134}Cs decay) based on July monitoring at Grøtting. These observations can not be directly compared with observations from Fåset and Fonnåsfjellet, but the figure gives an illustration of the rapid decrease in radiocaesium concentration in sheep when fungi has little influence. Estimated effective half life is 2.1 ± 0.3 years, $R^2 = 0.54$.

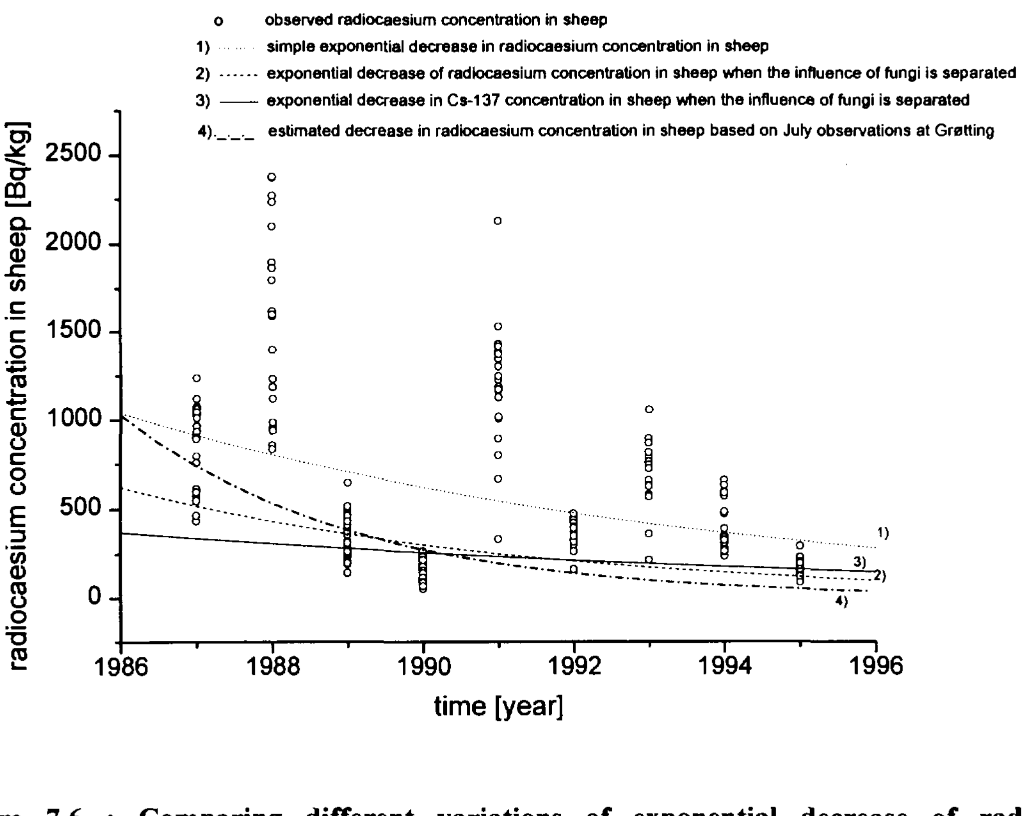

Figure 7.6 : Comparing different variations of exponential decrease of radiocaesium concentrations.

This comparison of different models supports that fungal fruit bodies play a significant role in determining radiocaesium levels in grazing sheep. The simple exponential model has a low model fit compared to the one including influence by fungi. The difference in estimated half lives between the two models also supports that fungi play a role in the long term behaviour of radiocaesium in grazing sheep. Since there is insignificant decrease in ^{137}Cs concentration in fruit bodies they continue to supply radiocaesium to sheep, while the intake through vegetation decreases.

Today only about 2 % of the total activity of radiocaesium is due to ^{134}Cs, and the future decrease in concentrations in sheep will therefore be determined by the effective half life ^{137}Cs. The developed model suggest that this will be 7.1 ± 2.3 years.

The effective half life based on July observations from Grøtting indicate a faster decrease in levels in vegetation. This study has focused on the influence of fruit bodies, but factors influencing vegetation growth (e.g. temperature, precipitation) will also affect radiocaesium concentration in vegetation and hence in animals. Radiocaesium levels monitored in July will probably be more sensitive to these factors than levels monitored in sheep later in the season.

The estimates of effective half life for ^{137}Cs are shorter than the 20 years suggested earlier [54]. However, the rate of decrease in ^{137}Cs levels will differ between environments and areas of the country, and a summary of half lives values in the range 3-20 years [55].

The observations from Spekdalen, a grazing unit where extended use of Prussian blue in salt licks has taken place, turned out to be poorly estimated by the model. The effective half life was significantly negative, the influence of fungi was negative while the estimated contamination level in 1986 was very high, and the model fit was poor (R^2 = 0.08). There is no decrease in radiocaesium concentration in sheep during the period where Prussian blue has been used (1989-1995), and no obvious effect of changes in amount of fungi. The fungi information may be less valid for this area since Spekdalen is located higher above sea level than the other study areas. The variability within the monitored samples is high and illustrates the importance of performing analysis of long term behaviour in areas without countermeasures.

The model with its estimated parameters is only valid for the areas studied and the observed period, and can not without reservations be transferred to other areas or later years. However, it may be interesting to make a prediction of the radiocaesium concentration in sheep grazing in Fåset and Fonnåsfjellet 1996. If 1996 turns out to be a good year for fungi the predicted mean radiocaesium concentration is 770 Bq/kg, while a low abundance of fruit bodies leads to a predicted radiocaesium concentration of 220 Bq/kg, assuming that the sheep pasture until 01.09. This means that if large amounts of fungi appear during August 1996, animals from this area will have a radiocaesium concentration above the 600 Bq/kg limit, and that feeding with uncontaminated feed will be necessary before slaughter.

8. Conclusions

8.1 Variability within grazing unit

The observations from Vuludalen and Baklia in 1987 indicated that the radiocaesium concentrations within the flocks were close to normal distributed. The deviations from the normal distribution within the groups decreased when the data sets were divided into ewes and lambs. However, these results can not be transferred to other areas with lower contamination levels, or later years with lower radiocaesium concentrations in vegetation and animals, because of the skewed distribution that follows when the radiocaesium concentration approaches zero. However, as persistently high radiocaesium levels are observed in many areas these can be assumed to be normally distributed.

The results indicated a significant higher radiocaesium concentration in lambs than in ewes, even at time of slaughter when lambs are assumed to have the same size as adult animals. The variability within lambs and ewes was lower than the variability within the whole herd.

One reason for monitoring live sheep is to check whether the mean radiocaesium concentration in animals from a grazing unit is below the accepted limit of 600 Bq/kg, and thus may be slaughtered without any form of countermeasures. It might therefore seem useful to monitor samples of only lambs, both because of the higher radiocaesium compared to ewes, and because the lower variability within the group decreases the uncertainty in the estimated mean radiocaesium concentration. If the radiocaesium concentration in lambs is below 600 Bq/kg, it is reasonable to assume that the radiocaesium concentration in ewes and the mean radiocaesium concentration in the whole grazing unit, also is below this limit. Better counting statistics and levels well above the detection limit also supports this idea.

The uncertainty in estimated mean radiocaesium concentration in a grazing unit was estimated to about 20 % with the present sampling method of monitoring 8-10 animals. If a probability of 90 % is desired for estimating the mean radiocaesium concentration with an error less than 10 % (uncertainty in the monitoring method not taken into account), the required sample size is ca. 25 animals, or 2.5 times the present sample size.

8.2 Long term behaviour

The suggested model 6.2 gave a good indication of the long term behaviour of radiocaesium in sheep grazing unimproved pasture. Despite many assumptions and uncertainties, the observed and estimated values of radiocaesium concentration in sheep showed good correlation. Most of the variability which remains unexplained by the model seem to result from the relatively low number of animals monitored.

The results support that fungi is an important source of radiocaesium. The contribution from ingested fruit bodies is estimated to 75-80 % of the total radiocaesium concentration in sheep in 1988 and 1991, which both were years with large abundance of fruit bodies in the pasture. If 1996 turns out to be a year with large abundance fruit bodies, radiocaesium concentrations in sheep grazing in the studied area may reach 750-800 Bq/kg. An effective countermeasure seems then to be to reduce the grazing season (but this will influence the weight of slaughter animals).

The estimated effective half life for ^{137}Cs in vegetation in the grazing units of Fåset and Fonnåsfjellet using the general model (3 estimated levels of fungi abundance with relative importance 1:4:9 and no differention between ewes and lambs, or Fåset and Fonnåsfjellet) was 7.8 ± 2.6 years. T_{eff} estimated from different model variants (with significant parameters) was not significantly different from this value. The estimated T_{eff} based on July observations in the grazing unit of Grøtting (2.4 ± 0.4 years) supports the conclusion of an estimated effective ecological half life for ^{137}Cs of 7-8 years, even though the model gave higher R^2-values with longer T_{eff}.

In comparison the estimated T_{eff} for ^{134}Cs + ^{137}Cs (observed T_{eff}) was 5.3 ± 1.1 years, the difference from the above value being due to physical decay of ^{134}Cs. Since little ^{134}Cs is left by 1996 the model suggest that future radiocaesium levels will decrease with a half life of 7-8 years (but will still be highly influenced by appearance of fruit bodies).

9. Recommendations

- The study of sample sizes required in order to determine the mean radiocaesium level with a predetermined degree of certainty did not consider the difference in radiocaesium concentration between ewes and lambs. The same analysis conducted on each of the subgroups ewes and lambs, might turn out differently since the variability within each of the subgroups was lower than within the whole group. Monitoring only lambs might require smaller samples than monitoring both ewes and lambs mixed if a certain level of precision is wanted.
- The abundance of fungi used in this study did not have any clear distinction between «good», «medium», and «bad» year. A more thorough study of the fruit body appearance and more precise determination of the amount of fruit bodies (e.g. number of fruit bodies per m^2) would probably decrease the uncertainty, and increase the validity of the model.
- *Days* on pasture with fungi available as feed is probably a to exact determination of this factor when taking into account all uncertainties and assumptions in the model. A more rough classification, e.g. weeks with fungi, could better be justified, but this requires better knowledge of fruiting date.
- In the area of Griningsdalen a thoroughly study of fungal activity is worked out [53]. In this study the factor influencing the appearance of fruit bodies is examined, and both the exact date for occurrence of mushrooms and the amount of mushrooms is determined. Implementing the suggested model on the Griningsdalen data would be a good test of its validity.

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Appendix 1

Report sheet for live monitoring of sheep

FYLKESVETERINÆREN FOR HEDMARK OG OPPLAND

FYLKESVETERINÆREN FOR HEDMARK OG OPPLAND
13. SEP. 1990
J.NR. ARKIV.

RAPPORTSKJEMA - LEVENDEDYRMÅLINGER -1990

FYLKE..Hedmark.. KOMMUNE..Tynset.....

MÅLINGER UTFØRT AV...[illegible]

STED FOR MÅLINGER..[illegible] i Tynset

BAKGRUNNSSTRÅLING SAU..185... i./s.x F..6.75..

-"- LAM..170... -"- x F..6.75.

-"- ANNET...... -"- x F.........

DATO OG TID FOR MÅLING..11/9.1990 fra 8³⁰-12⁰⁰... Skjemaføring?

MÅLING AV FANTOM .1039..Bq/kg G.fakt.sau.3.47?

FANTOM BEREGNET .1000..Bq/kg G.fakt.lam.4.07?

DIFFERANSE ..39..Bq/kg G.fakt. ......

BAKGRUNNSMÅLINGER
TID Lt ..64..HØYDE.64
I=..................
I=................
I=................
I=................
I-snitt ..111.?..

TID Lt 64..HØYDE.576
I=................
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I=................
I=................
Isnitt...1153?....

OMRÅDE/ BEITELAG	DYREART/ALDER/KJØNN IDENTITET (Nr.)		DYREEIER		BQ/kg
[illegible]	Lam ♀	0007	[illegible]	4	1334
"	Lam ♀	4001	"	5	1465
"	Sau ♀	5054	Tor A. [illegible]	8	2192
"	Lam ♂	0051	[illegible]	1	802
"	Lam ♂	0010	Tor A. [illegible]	10	2301
"	Lam ♂	5125	[illegible]	3	1347
"	Lam ♂	5124	"	6	1443
"	Sau ♀	5033	"	2	1202
"	Lam ♀	0030	Tor A. [illegible]	7	2090
"	Lam ♂	0009	"	9	2299

LAVESTE V. 802	HØYESTE V. 2301	MEDIAN 5+6	1651
KORREKSJON	KORREKSJON	KORREKSJON	0
		MEDIAN KORR	1651

FYLLES UT I 3 EKSEMPLARER. DET ENE EKS. SENDES FYLKESVETERINÆREN UMIDDELBART.
DET ANDRE EKS. BEHOLDES AV DEN SOM UTFØRER MÅLINGENE.

Appendix 2

Test of normal distribution for the data sets from Baklia and Vuludalen.

Baklia

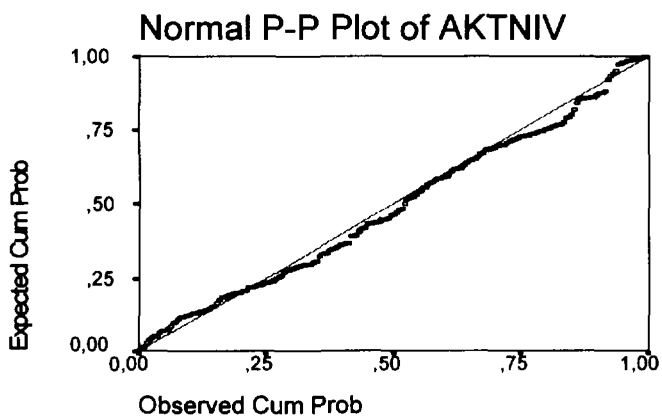

Figure 1: Normal plot of the whole data set from live monitoring if sheep in Baklia 1987 (212 animals).

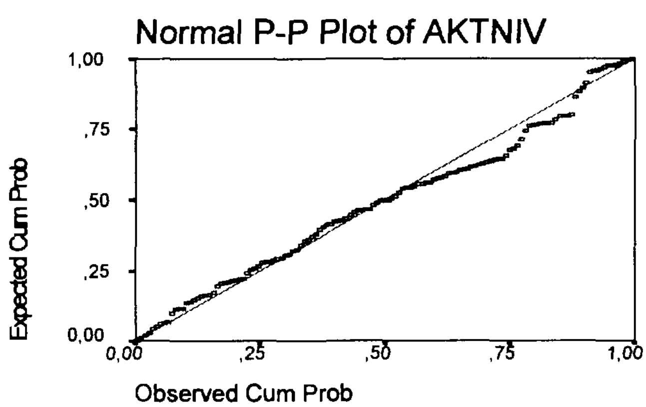

Figure 2: Normal plot of the observed radiocaesium concentration in lambs monitored in Baklia 1987 (131 lambs).

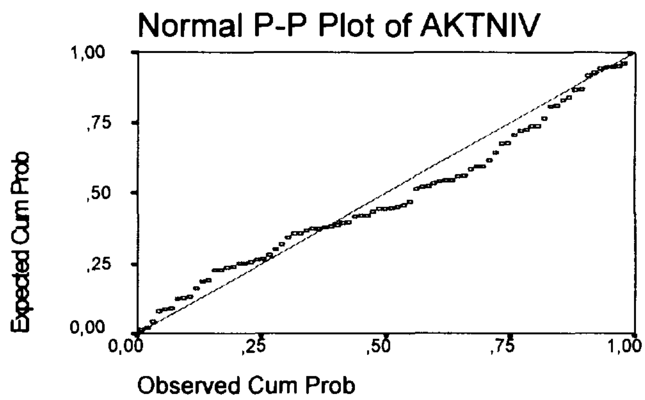

Figure 3: Normal plot of the observed radiocaesium concentration in ewes monitored in Baklia 1987 (81 ewes monitored).

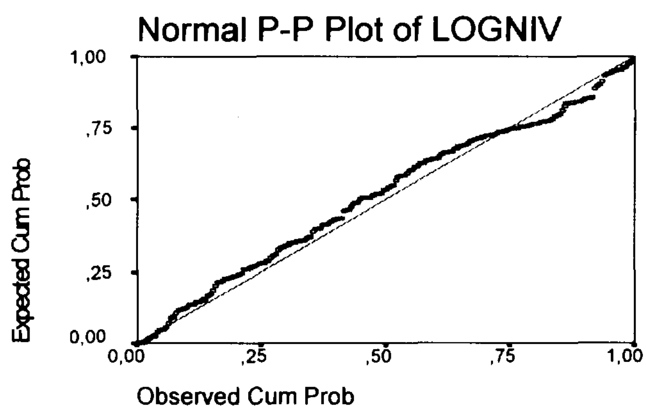

Figure 4: Normal plot of the logtransformed data from the whole herd monitored in Baklia 1987 (212 animals).

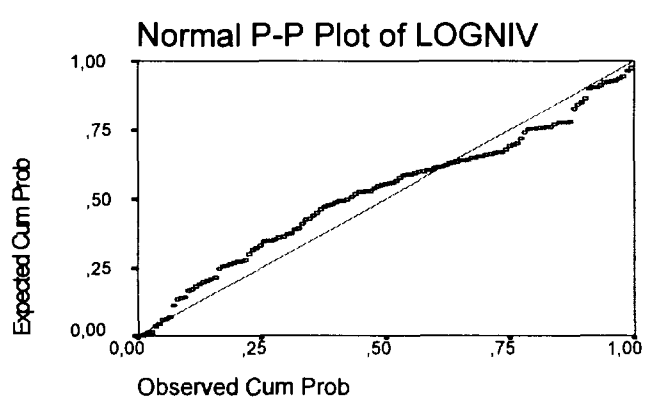

Figure 5: Normal plot of the logtransformed data for lambs from Baklia 1987.

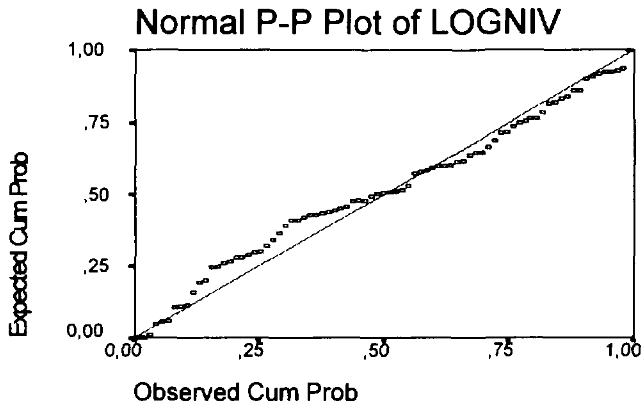

Figure 6: Normal plot of the logtransformed data for ewes monitored in Baklia 1987.

Vuludalen

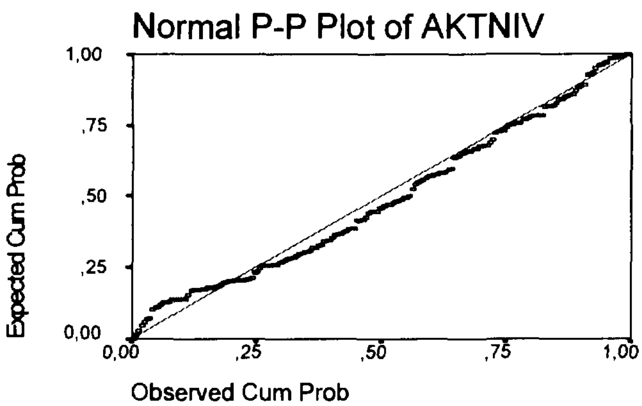

Figure 7: Normal plot of the observed radiocaesium concentration in sheep monitored in Vuludalen 1987 (180 animals monitored).

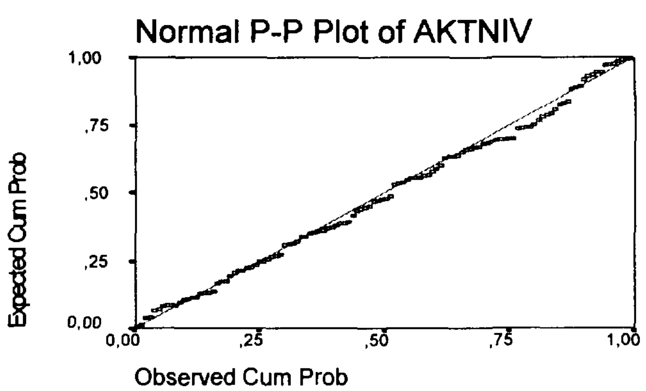

Figure 8: Normal plot of the observed radiocaesium concentration in lambs monitored in Vuludalen 1987 (118 animals monitored).

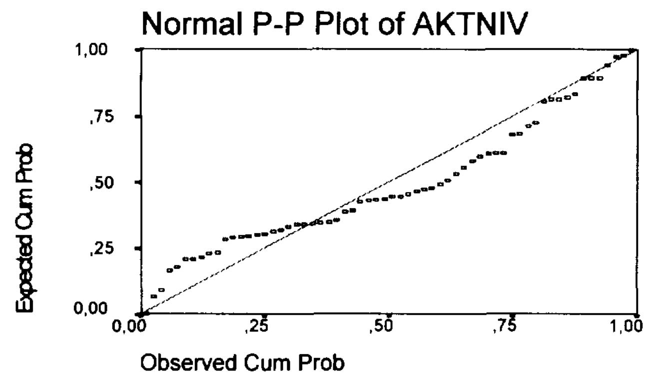

Figure 9: Normal plot of the observed radiocaesium concentration in ewes monitored in Vuludalen 1987 (62 animals monitored).

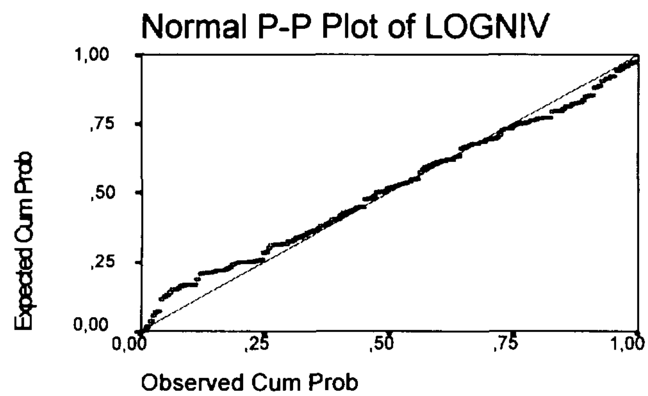

Figure 10: Normal plot of the logtransformed data for sheep monitored in Vuludalen 1987.

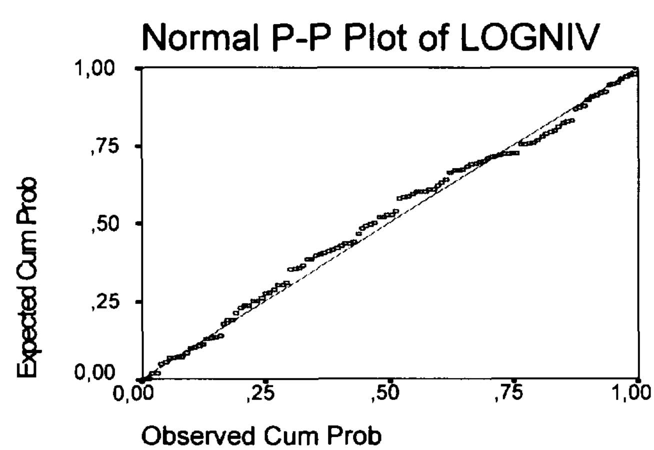

Figure 11: Normal plot of the logtransformed data for lambs monitored in Vuludalen 1987.

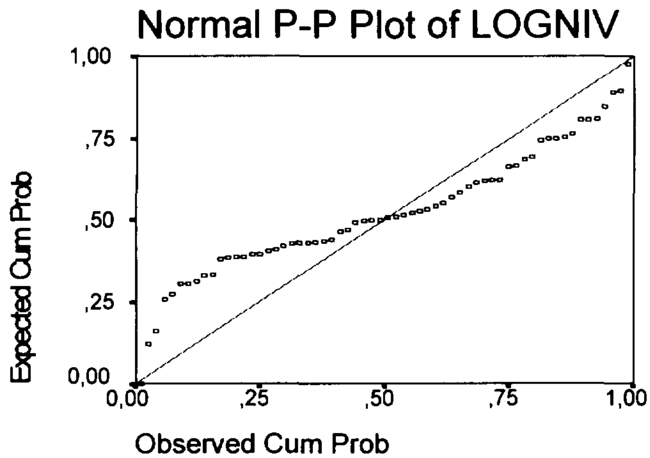

Figure 12: Normal plot of the logtransformed data for ewes monitored in Vuludalen 1987.

Appendix 3 Correction for Cs-134 decay

The Chernobyl fallout consisted of ^{134}Cs and ^{137}Cs , ratio 1:2 [2]. Both isotopes will contribute to the measured activity level, but since the physical half life for ^{134}Cs is short (2 years) compared with ^{137}Cs (30 years) the ratio will change from year to year. The contribution from ^{134}Cs and ^{137}Cs are:

$$^{134}A = {}^{134}A_0\, e^{-\lambda_{134} t} \qquad\qquad {}^{137}A = {}^{137}A_0\, e^{-\lambda_{137} t}$$

where A is activity at time t and A_0 is the activity at t = 0. Since the ratio $^{134}A_0 / {}^{137}A_0 = 1: 2$, the expression for ^{137}Cs activity is :

$$\frac{^{134}A}{^{137}A} = 0.5\, e^{(\lambda_{137}-\lambda_{134})\, t}$$

$$1 = {}^{134}A + {}^{137}A = {}^{137}A\ (1 + 0.5\, e^{(\lambda_{137}-\lambda_{134})\, t})$$

The following expression shows the relative ^{137}Cs content in the radiocaesium from Chernobyl fallout as a function of time after the Chernobyl accident:

$$^{137}A = 1 / (1 + 0.5\, e^{(\lambda_{137}-\lambda_{134})\, t})$$

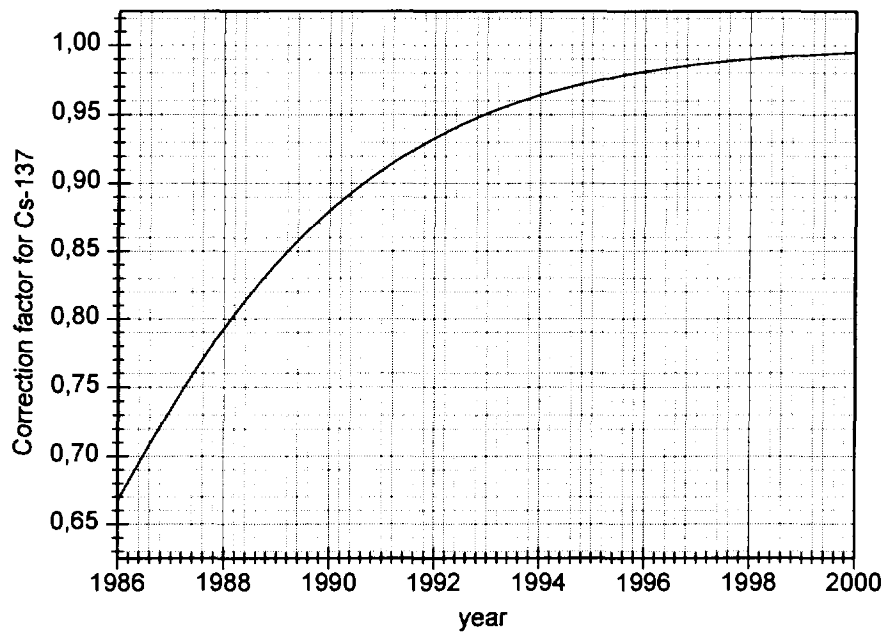

Figure 13: The ^{137}Cs-contribution to the total radiocaesium activity as a function of time after the Chernobyl accident

In order to find the correct ^{137}Cs activity the experimental data is multiplied by the correction factor from Table 1.

Table 1: Correction factor for ^{137}Cs activity, or (^{137}Cs activity)/(total radiocaesium activity) as a function of years after the Chernobyl accident.

time(year)	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**
correction factor	0.73	0.79	0.84	0.88	0.91	0.93	0.95	0.96	0.97