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# TRANSFER OF $^{137}\text{Cs}$ FROM CHERNOBYL DEBRIS AND NUCLEAR WEAPONS FALLOUT TO DIFFERENT SWEDISH POPULATION GROUPS

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**Abstract** — Data from measurements on the body burden of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in various Swedish populations between 1959 and 2001 has been compiled into a national database. The compilation is a co-operation between the Departments of Radiation Physics in Malmö and Göteborg, the National Radiation Protection Authority (SSI) and the Swedish Defense Research Agency (FOI). In a previous study the effective ecological half time and the associated effective dose to various Swedish populations due to internal contamination of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  have been assessed using the database. In this study values of human body burden have been combined with data on the local and regional ground deposition of fallout from nuclear weapons tests (only  $^{137}\text{Cs}$ ) and Chernobyl debris (both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ), which have enabled estimates of the radioecological transfer in the studied populations.

The assessment of the database shows that the transfer of radiocaesium from Chernobyl fallout to humans varies considerably between various populations in Sweden. In terms of committed effective dose over a 70 y period from internal contamination per unit activity deposition, the general (predominantly urban) Swedish population obtains 20-30  $\mu\text{Sv/kBq m}^{-2}$ . Four categories of populations exhibit higher radioecological transfer than the general population; *i.*) reindeer herders ( $\sim 700 \mu\text{Sv/kBq m}^{-2}$ ), *ii.*) hunters in the counties dominated by forest vegetation ( $\sim 100 \mu\text{Sv/kBq m}^{-2}$ ), *iii.*) rural non-farming populations living in sub-arctic areas (40-150  $\mu\text{Sv/kBq m}^{-2}$ ), and *iv.*) farmers ( $\sim 50 \mu\text{Sv/kBq m}^{-2}$ ). Two important factors determine the aggregate transfer from ground deposition to man; *i.*) dietary habits (intakes of foodstuff originating from natural and semi-natural ecosystems), and *ii.*) inclination to follow the recommended food restriction by the authorities. The transfer to the general population is considerably lower ( $\sim$  a factor of 3) for the Chernobyl fallout than during the 1960's and 70's, which is partly explained by a higher awareness of the pathways of radiocaesium to man both by the public and by the regulating authorities, and by the time-pattern of the nuclear weapons fallout during the growth season in Sweden.

Keywords: Radiocaesium; in-vivo body burden; aggregate transfer; effective dose

## 1. Introduction

The era of major global dispersion of anthropogenic radionuclides started in the beginning of the 1950s with the program of nuclear weapons testing in the atmosphere (UNSCEAR 1977; UNSCEAR 1982). The debris consisted of long-lived nuclides such as  $^{90}\text{Sr}$  ( $T_{1/2,\text{phys}}=28$  y),  $^{137}\text{Cs}$  ( $T_{1/2,\text{phys}}=30.2$  y) and short-lived radionuclides such as  $^{131}\text{I}$  ( $T_{1/2,\text{phys}}=8.06$  d). These radionuclides have significant radiological consequences for man due to their elemental properties, giving rise to high specific uptakes in living tissues. In a study from 1961 it was discovered that reindeer herders in Scandinavia had significantly higher body burdens of  $^{137}\text{Cs}$  compared with what was found in urban areas (Lidén, 1961). During the following decades several studies on the body burden of  $^{137}\text{Cs}$  in reindeer herders in the Nordic countries and in other sub-Arctic populations were carried out (Miettinen et al., 1964; Soumela and Rahola, 1994; Hanson et al., 1996; Wright et al., 1997). Certain ecosystems were then demonstrated to be more sensitive to transfer of deposition of fission products to man, especially within the Sub-Arctic ecosystems where populations mainly lived on herding of the caribou or reindeer (Lidén 1961; Lidén and Gustafsson, 1966; UNSCEAR 1977; UNSCEAR 1982; Falk et al., 1991; Johansson and Ågren, 1994; Howard et al., 1995; Ågren, 1998). After the Partial Test Ban Treaty was signed by the superpowers in 1963, the number of atmospheric detonations decreased considerably, leading also to gradually decreasing global fallout.

After the Chernobyl accident in 1986, which resulted in large fallout of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in Sweden, many of the human body burden studies were re-launched. This time the deposition was much more inhomogeneously distributed, where certain areas obtained more than  $100 \text{ kBq m}^{-2}$  of  $^{137}\text{Cs}$ , compared with, on average,  $2\text{--}3 \text{ kBq m}^{-2}$  from the global fallout (Edvarson, 1991; Vintersved et al., 1991; Isaksson and Erlandsson, 1998; Wright et al., 1999). Studies were also conducted on other critical populations in Sweden, such as hunters and farmers living in regions with high deposition of radiocaesium (Ågren, 1998).

The Swedish government has assigned the Swedish Radiation Protection Authority, SSI, the responsibility for obtaining and assuring one of the fifteen national environmental quality objectives, *A Safe Radiation Environment* (Swedish Government, 2001). One of the criteria for a safe radiation environment is that no human receives dose contributions from the sum of all man-made sources higher than  $1 \text{ mSv}$  effective dose per year. It is therefore important to compile existing experimental data on internal contamination levels in man and other organisms, and relate these values to the levels of radioactive contaminants in the environment. During spring 2001 SSI launched a project where data on body burdens of radioactive cesium in various Swedish populations during the past four decades were compiled into a single database for a general assessment. The aim of the database is to study the various exposure pathways and identify which of these pathways could be of most importance in a future exposure scenario. It is also intended to be used as a reference data set in the national environmental monitoring program by the SSI.

In this assessment we aim to investigate the magnitude and dispersion of the transfer of ground deposition of radiocaesium to man in Sweden using two different approaches to compare and define a representative transfer factor. A comparison of the estimated committed effective dose per unit activity deposition on the ground is made between different critical groups that are known to be more sensitive to radioecological transfer. We also intend to compare the radioecological transfer to some Swedish populations from  $^{137}\text{Cs}$  originating from the nuclear weapons fallout versus the Chernobyl fallout.

## 2. Material and methods

Data from body burden studies of various Swedish populations conducted by the Swedish Defense Research Agency (FOI), the National Radiation Protection Authority (SSI), and the Departments of Radiation Physics at the universities in Lund, Malmö, Göteborg and Umeå, have been pooled to one database in a spreadsheet format. It contains data on the age of the investigated subjects, place of residence, time at the whole body counting, occupation, body-weight and the body burdens of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  (after the Chernobyl accident in 1986) and  $^{40}\text{K}$ . The radionuclide  $^{40}\text{K}$  is an important chemical analogue to cesium and the body potassium content is often obtained simultaneously in connection with the whole-body counting (Leggett, 1983). The identities of the participating subjects in the database are coded. The

place of residence of the individuals is recorded in the form of coordinates as defined by the Swedish RT90-system (Lantmäteriet, 2003) as well as by the name of the home municipality (and county) of the residence. In this study, however, the populations are not primarily divided into their geographical habitation, but rather after their occupation. Some occupations are associated with dietary habits that are known from previous studies to be particularly sensitive to the transfer of radioactive cesium (Ågren, 1998). The following categorization of the population has thus been done; *i.*) reindeer herders, *ii.*) hunters, *iii.*) farmers, *iv.*) rural non-farming populations and *v.*) urban populations. In addition to these categories a national survey conducted by the SSI in 1987, 1991 and 1994, has been included in this assessment. The individuals in these studies are heterogeneous and mixed in terms of occupation and dietary habit and can thus not be categorized as the other populations. In Table 1 is given an overview of the populations considered in this study.

## 2.1 Aggregate transfer factor

UNSCEAR (1982) has presented a model for the global transfer of radionuclides from atmospheric fallout to man (Fig. 1). The transfer between compartments,  $P_{ij}$ , describes the rate at which radionuclides are transferred from one compartment to another. The transfer rate from the ground deposition to man,  $P_{24}$  [(Bq kg<sup>-1</sup>)/(kBq m<sup>-2</sup>)], depends on the different types of pathways in a given ecosystem. Some time after the initial ground deposition the radionuclide contents in the foodstuff compartment can be assumed to be in equilibrium with the radionuclide concentration in humans; for <sup>137</sup>Cs a quasi-equilibrium has been found to occur in human populations about one year after the on-set of continuous global fallout (Fredriksson et al., 1963). If all transfer pathways to humans in a given ecosystem are considered the quantity  $P_{24}$  is approximately proportional to the aggregate transfer factor, here denoted as  $T_{ag}$  [(Bq kg<sup>-1</sup>)/(kBq m<sup>-2</sup>)]. The quantity  $T_{ag}$  can be expressed as a ratio between the average activity concentrations in humans,  $q_{Cs}$  [Bq kg<sup>-1</sup>] and the cumulated deposition on ground,  $A_{cum}$  [kBq m<sup>-2</sup>] at a certain point in time,  $t$  after a deposition event. Assuming a mono-exponential biological half-time of caesium in humans, that is, a constant biological excretion rate, ( $\lambda_{biol}$ =constant in time), which to a first approximation is the case for <sup>137</sup>Cs (Räaf, 2000),  $T_{ag}$  will thus represent an instantaneous value of the aggregate transfer rate to humans at time  $t$  (Eq. 1).

$$T_{ag}(t) = \frac{P_{24}(t)}{(1 + \lambda_{phys} + \lambda_{biol})} = q_{Cs,avg}(t) / A_{cum}(t) \quad \text{Eq. 1}$$

where

$q_{Cs,avg}(t)$	= the average body concentration of <sup>137</sup> Cs at time $t$ for a given population [Bq kg <sup>-1</sup> ];
$\lambda_{phys}, \lambda_{biol}$	= physical decay rate and biological decay rate respectively [year <sup>-1</sup> ]. The biological decay rate is inversely proportional to the biological half-time, $T_b$ , which for <sup>137</sup> Cs ranges from 0.2-0.5 years in adults (ICRP 56, 1989);
$A_{cum}(t)$	= the observed total activity deposition on ground averaged over some representative area (see later sections)

In this study the aggregate transfer of radiocesium to the investigated populations has been calculated in two ways in order to achieve *i.*) a quantitative measure of the transfer integrated over time that can be compared between various populations, and *ii.*) a measure of the committed effective dose per unit activity deposition to the populations considered. The two methods are based on a time integration or summation of the mean <sup>137</sup>Cs body concentrations, normalized to the body potassium concentration, (the ratio is here denoted <sup>137</sup>Cs/K), in unit of grams stable potassium, (gK)<sup>-1</sup>, and the average accumulated total deposition on ground,  $A_{cum}$  (kBq m<sup>-2</sup>) in an area that is assumed to supply the main fraction of cesium-containing foodstuff to the population considered.

UNSCEAR presented in 1977, a definition of the time integrated aggregate transfer from ground to man, here denoted as  $T_{ag,int,UN}$  [Bq (gK)<sup>-1</sup> y/kBq m<sup>-2</sup>] in Eq. 2;

$$T_{ag,int,UN} = \frac{\int_{t_0}^{t_1} ({}^{137}\text{Cs} / K)_{avg}(t) dt}{\sum_{t=t_0}^{t=t_1} A_{ann}(t)} \quad \text{or} \quad = \frac{\sum_{t=t_0}^{t_1} ({}^{137}\text{Cs} / K)_{avg}(t) \cdot \Delta t}{\sum_{t=t_0}^{t_1} A_{ann}(t)} \quad \text{Eq. 2}$$

where

$({}^{137}\text{Cs}/K)_{avg}(t)$  = average ratio of body burden of  ${}^{137}\text{Cs}$  and potassium at time  $t$  for a given population [ $\text{Bq (gK)}^{-1}$ ];  
 $A_{ann}(t)$  = annual ground deposition density of  ${}^{137}\text{Cs}$  [ $\text{Bq m}^{-2} \text{ y}^{-1}$ ]; the annual deposits are summed without correction for continuous physical decay.  
 $t_0$  = beginning of the time period considered for deposition cumulation (y);  
 $t_1$  = end of the time period considered (y);  
 $\Delta t$  = time interval between consecutive observations of  $({}^{137}\text{Cs}/K)_{avg}(t)$  when using trapezoid summing

This method is suitable when considering populations living within a well-defined area, such as the administrative divisions of countries into counties and municipalities, over which the fallout is averaged. An advantage of this method is the possibility to convert the time-integrated transfer to humans of  ${}^{137}\text{Cs}$  from a single deposition event, such as the Chernobyl fallout in 1986, into a committed effective dose from internal contamination per unit activity deposition density,  $E_{ag,int}$  [ $\text{mSv/kBq m}^{-2}$ ], using the following relationship (Eq. 3);

$$E_{ag,int} = \frac{\int_{t_0}^{t_1} q_{Cs,avg}(t) \cdot 0.00140 \cdot dt}{\sum_{t=t_0}^{t=t_1} A_{ann}(t)} \quad \text{or} \quad = \frac{\sum_{t=t_0}^{t_1} q_{Cs,avg}(t) \cdot 0.00140 \cdot \Delta t}{\sum_{t=t_0}^{t_1} A_{ann}(t)} \quad \text{Eq. 3}$$

where a dose conversion factor of 0.00140 ( $\text{mSv Bq}^{-1} \text{ y}^{-1}$ ) for homogeneous distribution of  ${}^{137}\text{Cs}$  in an adult male of body size 70 kg, has been adopted from Falk et al., 1991. The transfer factor and the committed effective dose have been integrated over a time interval of 70 years [ $t_0, t_1$ ]. This corresponds to the ICRP recommendation for individual effective dose assessment of members of the public (ICRP, 1991).

Some of the populations investigated here are dispersed over large geographical areas (illustrated in Fig. 2), which makes the definition of the aggregate transfer in Eq. 1 somewhat unsuitable. The reindeer herders and hunters that were investigated after the Chernobyl fallout live at different locations with a highly varying amount of ground deposition density. In order to enable a general comparison between different populations with respect to dietary habits rather than place of residence, an alternative method of estimating the transfer has also been used here (Eqs 4-5). The modified transfer factor used in this work (TW),  $T_{ag,int,TW}$  [ $\text{Bq (gK)}^{-1} \text{ y} / (\text{kBq m}^{-2})$ ], is the population mean of the ratio between the individual  ${}^{137}\text{Cs}/K$ -ratio [ $\text{Bq (gK)}^{-1}$ ] and the average cumulated activity deposition,  $A_{cum}$ , [ $\text{kBq m}^{-2}$ ].  $A_{cum}$  is the deposition that is directly measured when performing soil sampling at time  $t$ , and is related to the annual deposition,  $A_{ann}$ , as in Eq. 5. (See Appendix A.1 for more details on estimating the ground deposition). In this work, the individual value at time  $t$  of the  ${}^{137}\text{Cs}/K$ -ratio is thus divided by  $A_{cum(i)}(t)$  [ $\text{kBq m}^{-2}$ ], which is the average cumulated activity deposition density at the location (municipality or county) which the individual inhabits. This ratio is in turn averaged over all individuals,  $i$ , belonging to the population considered, irrespectively of where they live:

$$T_{ag,int,TW}(t) = \sum_{Avg} \left\{ \left( \frac{{}^{137}Cs / K}{A_{cum(i)}} \right)_i \right\} \quad \text{Eq. 4}$$

where

$$A_{cum(i)}(t) = \int_{t_0}^t A_{ann}(t') \cdot e^{-\lambda \cdot (t-t_0)} dt' = TP(t) \cdot \int_{t_0}^t A_{ann}(t') dt' \quad \text{Eq. 5}$$

and

$A_{cum(i)}(t)$  = cumulated total activity deposition at time  $t$  [kBq m<sup>-2</sup>], averaged over the municipality or county of the subject  $i$ ;  
 $TP(t)$  = function fitted to time-pattern of annual deposition – obtained graphically if  $A_{ann}(t)$  varies irregularly with time (See Table A1-1);

From previous studies it has been shown that between 1965 and 1985, and after 1987 (one year after the Chernobyl deposition), the gradual decrease in the <sup>137</sup>Cs concentration of various Swedish populations follows mono- or dual exponential time-patterns (e.g. Ågren, 1998; Rääf, 2000). If feasible, curve fits of observed values of  $T_{ag}$  as a function of time has been carried out by means of non-linear regression, in order to enable a time-integration and a further extrapolation over a 70-year period. For pre-Chernobyl  $T_{ag}$  values  $t_0$  is thus set to 1965, and for the Chernobyl fallout  $t_0$  is set to 1987.3. These curve fits are, however, not applicable to  $T_{ag}$  values deduced from data prior to 1965, and therefore trapezoidal summing has been used in these cases (See Appendix A.2).

## 2.2 Estimation of cumulated total ground deposition of <sup>137</sup>Cs in Sweden prior to and after the Chernobyl accident

In Sweden a detailed mapping has been carried out of the ground deposition of <sup>137</sup>Cs from the Chernobyl fallout based on air surveys performed after 1986 (Byström, 1990; Byström, 2000). The deposition values have been processed into various data sets, one of which gives the average equivalent surface deposition of <sup>137</sup>Cs in kBq m<sup>-2</sup> for each Swedish municipality (Lindgren et al., 2002). The equivalent surface deposition as defined by Finck (Finck, 1992), represents the surface deposition density a given signal intensity from mobile field measurements will yield if the deposition is assumed to be a true surface source with no penetration into the soil. This definition is used in cases when extensive data on ground deposition are needed in the absence of ground penetration data of the deposited fallout, which was the case during the time period of airborne mapping of the Chernobyl fallout in Sweden. The equivalent surface deposition values have been scaled upwards by a factor 1.7 in order to obtain estimates of the regional and local averages of the total deposition,  $\Sigma A_{ann}(t)$  in eqs 1-2, in May 1986. The factor 1.7 corresponds to the mean ratio between the total and equivalent surface deposition in Sweden as determined in a soil sampling study by Edvarson, 1991. From this data set it is possible to generate various regional and local averages for the deposition values (Table A.1-3).

Considering the fallout prior to 1986, there is little or no local mapping of the deposition in the country. There does, however, exist data on pre-Chernobyl fallout based on reconstructions from metrological records, since the annual fallout was highly correlated to the annual precipitation (e.g. Isaksson *et al.*, 1997; Wright et al., 1999). The time-pattern of the cumulated deposition of the nuclear weapons fallout in Sweden, described by the function  $T(t)$ , has thus been reconstructed by combining Danish data on annual <sup>137</sup>Cs fallout,  $A_{ann}$ , from 1950 and onward (Aarkrog et al., 1995; Fig. 3), with meteorological data on annual precipitation at various locations in Sweden, resolved into a 5\*5 km grid (Swedish Meteorological and Hydrological Institute, 2002). The Danish data has been used instead of similar Swedish time-series due to its extended time range. (There are no Swedish annual fallout data before 1962). The data on annual precipitation in Sweden has been combined with soil sampling data (Edvarson, 1991; Vintersved et al.,



1991) in order to reconstruct the geographical variation of the pre-Chernobyl fallout. The reconstructed values have been averaged over municipalities and counties (Fig. 4).

Algorithms for reconstructing the deposition and obtaining estimates of the aggregate transfer of radiocaesium from ground deposition to an individual at time  $t$ , residing at a location  $x$ , are given in Table A.2-1. The transfer of  $^{137}\text{Cs}$  has been calculated based on *i.*) the average deposition in the municipality of the subject's place of residence, and *ii.*) the average deposition in the county of the subject's residence. This has been done in order to investigate the influence of the size of the geographical area to which the estimation of  $\Sigma A_{\text{ann}}(t)$  and  $A_{\text{cum}}(t)$  refers in Eqs 1-5, and to have comparable geographical areas as in the assessment of the Chernobyl fallout. The average size ( $\pm 1$  STDV of mean) of a municipality in Sweden is  $1,416 \pm 147 \text{ km}^2$ . The average county size in Sweden is  $14,500 \pm 3,330 \text{ km}^2$  (Statistics Sweden, 2002). For urban populations in the major metropolitan areas in Sweden, the local deposition (average deposition in the municipality) is not expected to reflect the transfer to man in the same manner as the regional average (average deposition in the county), which cover the surrounding rural municipalities that supply a large part of the foodstuff consumed in the urban areas.

### 3. Results

#### 3.1 Time-integrated aggregate transfer and committed effective dose per unit activity deposition from Chernobyl fallout using the UNSCEAR-method

The estimated values of the time-integrated aggregate transfer,  $T_{\text{ag,int}}$  using the UNSCEAR definition (Eqs 2-3), combined with the average deposition in the county of residence, are given in Table 2, with the reindeer herders (SAMI\_W1, SAMI\_N1, and SAMI\_N2) exhibiting the highest time-integrated transfer,  $2640 \pm 590$  (1 standard error of mean)  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$  from the pre-Chernobyl fallout, and between 180-300  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$  from the Chernobyl fallout.

Using the UNSCEAR definition of  $T_{\text{ag,int}}$  allows an estimation of the committed effective dose,  $E_{\text{ag,int}}$  (time-span 70 years) from internal contamination of radiocaesium per unit activity deposition from a single-deposition event, such as the Chernobyl fallout in Sweden. The values are presented in Table 2 in units of  $[\mu\text{Sv}/\text{kBq m}^{-2}]$ . The reindeer herders (SAMI\_N1 and SAMI\_N2) are estimated to obtain, on average, 700-800  $\mu\text{Sv}$  from internal contamination of Chernobyl  $^{137}\text{Cs}$  per 1  $\text{kBq m}^{-2}$  deposition in their county of residence, compared with 17-28  $[\mu\text{Sv}/\text{kBq m}^{-2}]$  for the urban reference groups (URB\_S1, URB\_S2, URB\_M1 and URB\_W1) at the lower end of the range.

#### 3.2 Time-integrated aggregate transfer using the modified method

The time-integrated transfer factors for  $^{137}\text{Cs}$  using the modified definition presented in Eqs 4-5 for the various Swedish populations are presented in Table 3. The same pattern as discussed in the previous section is found here. The highest values of the time integrated aggregate transfer is obtained for populations that consume large amounts of locally produced food, such as the Sami populations whose  $T_{\text{ag,int}}$  from Chernobyl is projected to be 200-320  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$ , depending on whether the deposition is considered over the municipality or over the county. The corresponding values for the rural non-farmers living in the North of Sweden, (RUR\_NONFARM\_N1 and RUR\_NONFARM\_N2), are 15-50  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$ , followed by the hunters with a  $T_{\text{ag,int}}$  with about 11-30  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$ . Higher values are obtained for most populations when using county averages of the  $^{137}\text{Cs}$  ground deposition from the Chernobyl fallout. A marked agreement in the time integrated aggregate transfer is achieved when considering the average deposition over the county for the urban populations in the metropolitan areas (URBAN\_M1, URBAN\_W1; URBAN\_S1 and URBAN\_S2), with values around 6-10  $[\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})]$ . The corresponding value for the reindeer herders in Västerbotten County, SAMI\_N1, appears to be at least 20 times higher. The reindeer herders investigated in Norrbotten County, SAMI\_N2, exhibit an even higher transfer of  $^{137}\text{Cs}$ , which is up to 40 times higher than the urban populations.

Furthermore, the time-integrated aggregate transfer appears to have been 3 times higher for the global fallout in the populations investigated both before and after the Chernobyl fallout (URBAN\_M1 and URBAN\_S1 och S2). This tendency is even more pronounced when comparing the  $T_{ag,int}$  values for the reindeer herders (SAMI\_W1), whose time-integrated transfer appears to be a factor of 5 times higher than the Sami populations investigated after the Chernobyl fallout.

### 3.3 Use of the Potassium Normalization

Normalizing the individual body burden of  $^{137}\text{Cs}$  to that of potassium has been known to partially compensate for the biological variance within studied populations, which was the reason it was used in the UNSCEAR-procedure from 1977 to calculate the time-integrated aggregate transfer. For the populations considered here use of potassium normalization of the aggregate transfer of  $^{137}\text{Cs}$  does not decrease the relative variation (1 standard deviation/group average). The relative variation in the aggregate transfer for the adult reindeer herders in Västerbotten county, SAMI\_N1, comparing the use of the body concentration of  $^{137}\text{Cs}$ ,  $q_{Cs}$  [Bq], with the use of  $^{137}\text{Cs}/\text{K}$ -ratio [Bq (gK) $^{-1}$ ] in the expression given in Eq. 3, is given in Table 4. It should be noted that the observed variation also includes experimental uncertainties in the determination of  $q_{Cs}$  and the  $^{137}\text{Cs}/\text{K}$ -ratio. The lack of reduction in the relative variance when using potassium normalized body burdens in adult populations observed here is in accordance with previous results on the relative variance in the body burden of  $^{137}\text{Cs}$  (Räaf et al., 2005).

The national survey conducted by the SSI in 1987, 1988 and 1994 shows that there is a moderate correlation between the average body concentration of  $^{137}\text{Cs}$  in the individuals selected for the survey (REF\_NAT) and the average deposition in the municipality ( $r^2=0.52$  for the 1987 year sample). Using the  $^{137}\text{Cs}/\text{K}$  ratio [Bq (gK) $^{-1}$ ] instead of  $^{137}\text{Cs}$  concentration improves the correlation coefficient insignificantly ( $r^2=0.53$  for the same year). Considering the degree of correlation between body burden and average county deposition, no significant improvement was obtained by using potassium normalization (Figs 6-7), with corresponding values of  $r^2=0.83$  and  $0.82$  respectively. The results indicate, however, an improved correlation between  $^{137}\text{Cs}$  body concentration and  $^{137}\text{Cs}/\text{K}$ -ratio at a given point in time and the activity deposition on the ground, when considering county averages (Fig. 7) rather than municipality averages (Fig. 6).

## 4. Discussion

### 4.1 The aggregate transfer of $^{137}\text{Cs}$ from ground deposition to man in different populations in Sweden

The highest aggregate transfer of Chernobyl  $^{137}\text{Cs}$ , independent of which of the two definitions is used, was found among the reindeer herder populations, after which followed (in order of decreasing transfer) the rural non-farming population in Norrbotten County (RUR\_NONFARM\_N2), hunters in the Swedish counties dominated by forest vegetation, farmers living in the municipality most affected by the Chernobyl fallout in Sweden (Gävle), the rural non-farming population living in the same municipality, RUR\_NONFARM\_M1, and urban populations in the Swedish metropolitan areas (Malmö/Lund and Stockholm). The time integrated aggregate transfer for the latter group is about 10 to 20 times lower than for the reindeer herders in Sweden (Table 3).

The results also indicate the influence of the size of the area for which the activity deposition is considered in the estimates of the time-integrated transfer. A marked accordance between the studied urban populations (URB\_S1, URB\_S2, URB\_M1, and URB\_W1) is observed when using county averages (Table 3). This is also illustrated in Fig. 5, where the  $T_{ag}$  vs. time is plotted for the populations considered using the average deposition over municipalities and counties (left and right panels of Fig. 5 respectively). The urban populations will mostly be subjected to transfer of radiocaesium from dairy products and beef. Dairy milk is, however, still produced, distributed and consumed regionally in Sweden. Since this produce is one of the key transfer pathways of cesium (e.g. Möre et al., 1994), it is logical to assume that the aggregate transfer to the urban population is best described by the ratio between the human  $^{137}\text{Cs}$  concentration and the average deposition level in the county instead of the municipality. The results indicate also homogeneity of the dietary habits of the urban populations in Sweden, which live mostly on

foodstuffs that are distributed by the major food industry chains. These distribution chains tend to dampen local variations in the concentration levels of foodstuffs other than dairy milk. This may also explain why the rural non-farming population in the Gävle municipality (RUR\_NONFARM\_M1) exhibits a similar transfer of radiocaesium as the urban population in the major metropolitan areas.

For some populations (farmers and hunters), using county averages yields lower transfer factor values than using municipality averages. This indicates that the  $^{137}\text{Cs}$  concentration in these populations is more dependent on the contamination levels in food produced locally (e.g. game, berry, mushrooms, domestically grown vegetables, milk and beef from the local farms) rather than in the foodstuffs handled by the major dairy and beef distribution chains in Sweden.

Previous studies in Sweden show that the transfer of radiocaesium to key diet components and to human populations was higher for the nuclear weapons fallout than for the Chernobyl debris (Aarkrog, 1988; Hanson et al., 1966; Rääf, 2000). According to this study the time-integrated aggregate transfer to urban populations appears to be about 2-3 times higher for the nuclear weapons fallout than for the Chernobyl fallout (Tables 2-3). The main pathway for radiocaesium to travel from soil deposition to urban populations is by direct contamination of growing fodder crops and grazing, rather than the less dominant mechanism of root uptake of the element previously deposited on the soil into the edible parts of the plants. Since the continuous global fallout in the 1960s and 70s occurred over the growing seasons, the short-term transfer of radiocaesium resulted in higher contamination levels in the diet and in man compared with the Chernobyl scenario (see also e.g. Aarkrog, 1988). In addition to the time-pattern of fallout, the amounts of countermeasures taken in the form of restricted distribution of foodstuff and recommendations issued by the authorities have influenced the transfer to human populations. In Västerbotten county with an average  $^{137}\text{Cs}$  deposition of  $14 \text{ kBq m}^{-2}$ , the reindeer herders (SAMI\_N1) exhibit significantly lower radioecological transfer of  $^{137}\text{Cs}$  after the Chernobyl accident compared with the 1950s and 60s (Tables 2-3). This is probably to a large part an effect of the greater amount of countermeasures undertaken after Chernobyl in the region (Ågren, 1998). This is further supported by the fact that the reindeer herders in the county of Norrbotten (SAMI\_N2), which was to a much lower extent affected by the Chernobyl fallout ( $2 \text{ kBq m}^{-2}$ ), exhibit transfer values that are higher than for similar populations in the Västerbotten County.

Based on the UNSCEAR definition of radioecological transfer of  $^{137}\text{Cs}$  to man the projected aggregate transfer over a 70 y period after the Chernobyl fallout in Sweden results in a committed effective dose per unit activity deposition of about  $25 \text{ } \mu\text{Sv/kBq m}^{-2}$  in Swedish urban populations (Table 2). From an event similar to the Chernobyl fallout scenario, including similar remedial actions taken in terms of restricted distribution of contaminated foodstuffs, it is predicted that an activity deposition of  $1 \text{ kBq m}^{-2}$  of  $^{137}\text{Cs}$  in the northern counties in Sweden will result in a committed effective dose of 0.7 mSv in the reindeer herders (SAMI\_N1 and SAMI\_N2) and about 0.15 mSv to certain rural non-farming populations (RUR\_NONFARM\_N2).

Which of the two methods applied in this study are appropriate for use in future situations, or in other countries than Sweden, depends on the aim of the assessment. The UNSCEAR-method is recommended if the specific aim is to predict committed effective dose to populations located in certain regions and with assumed similar dietary habits. Time-integration of the average activity concentrations in human populations can be estimated by means of certain reference groups, as well as a time-integration of the average local or regional deposition. If, however, one wants to study differences in transfer between certain populations (defined in terms of, e.g., dietary habits), irrespective of their locations and of the regional variances in the deposition, then the modified method presented here is favorable. As an example, some subgroups or populations may be dispersed over large areas, such as the reindeer herders in Scandinavia, or the Inuit populations in Arctic Canada. Using the definition of  $T_{ag,int,TW}$  presented here enables aggregate evaluations of the population as a whole, not only on a local or regional level. The drawback of this method is that there is no clear relationship between  $T_{ag,int,TW}$  and the committed effective doses of individuals, and the values obtained must be compared relatively between the populations considered.

## 4.2 Use of the Potassium Normalization

Ideally the individuals selected to be part of the reference groups should be a cross-section of the population that is aimed to be represented in terms of  $^{137}\text{Cs}$  body concentration. Since human metabolism of alkali metals, such as Cs and K, is known to vary with age and gender (Leggett et al., 1984), the individuals should be replaced continuously in order to maintain the age- and gender composition of the reference group. In practice renewal of these reference groups during the time-span of the study is not always possible and therefore predictions of the local and regional transfer of  $^{137}\text{Cs}$  to humans must be made from groups consisting of individuals that may have grown older than the average population. In theory the ageing effects, such as the decreasing uptake of  $^{137}\text{Cs}$  to ageing humans, is assumed to be compensated for by normalization to the potassium content, hence the use of the  $^{137}\text{Cs}/\text{K}$ -ratio in the UNSCEAR definition of  $T_{ag,int}$  (UNSCEAR, 1977). There is, however, no evidence in this database that the use of normalizing the radiocaesium body contents in humans with their respective potassium content will result in higher degree of correlation between the regional average of the ground deposition and body burden (in terms of either  $^{137}\text{Cs}$  concentration or  $^{137}\text{Cs}/\text{K}$ -ratio) at a specific point of time after a fallout (Figs 6-7).

## 4.3 Variance of the estimated transfer factors

The total uncertainty in the transfer factor depends mainly on the variances in activity concentrations in humans and the geographical variation in the activity deposition in the region. The individual  $^{137}\text{Cs}$  concentrations are highly variable, with standard deviations typically between 60-100% of the population mean value and are in many cases highly log-normally distributed (Räaf et al., 2005). These variations have, however, less importance when considering the uncertainty in the population mean values of  $T_{ag,int}$  and  $E_{ag,int}$ , which is governed primarily by *i.*) the variance in the curve regressions of  $T_{ag,int}$  and in the trapezoid summing before 1986; and *ii.*) uncertainty in the  $A_{cum}$ -estimates, both for the global fallout in the 1950s and 60s and after 1986 for the Chernobyl fallout. The uncertainty of  $T_{ag,int}$  and  $E_{ag,int}$  is thus estimated from the sums of squares of the non-linear regressions of the  $q_C(t)$  and the  $T_{ag}$  population mean values (Appendix A.1). The time-integration of these quantities will roughly yield a standard deviation of the  $T_{ag,int}$ -estimate that is proportional to the interval used for the time-integration, that is 70 y in our case, as well as the residual sums of squares of the regression curve that is integrated,  $\text{SSR}/N$ ; where  $N$  is the number of points used in the regression (SSR is provided by the statistical software). For the  $T_{ag,int}$  estimates between 1945 and 2015, which are based partly on trapezoid summing, an estimation of the uncertainty can be done by quadratic summing of the standard error of mean of the  $^{137}\text{Cs}$  concentration before 1986 (Appendix A.1). These are in the order of less than 10% for all three populations considered (URB\_S1, URB\_M1 and SAMI\_W1), which is comparable with the uncertainty of the spatial and temporal variation of  $A_{cum}$  (Fig. 3). For the Chernobyl fallout, the uncertainty of  $A_{cum}$  can be assumed to originate from the geographical variation of the  $^{134}\text{Cs}/^{137}\text{Cs}$ -ratio observed initially after the deposition event based on the results from Edvarson, 1991 and Byström, 2000 (Table A.1-3). The relative uncertainty (1 SD/mean value) in the  $^{134}\text{Cs}/^{137}\text{Cs}$ -ratio is 25% (Edvarson, 1991), and is the largest contributor to the overall uncertainty in the  $T_{ag,int}$  estimates, irrespective of the methods used. A further comparison of the Swedish transfer data is given in Table 2 where  $T_{ag,int}$  values from populations in Denmark and Argentina presented in UNSCEAR is given (UNSCEAR, 1977). It is evident that the Swedish urban population exhibits radioecological transfer of  $^{137}\text{Cs}$  similar to what has been observed internationally.

## 5. Conclusions

- The time-integrated aggregate transfer of  $^{137}\text{Cs}$  for the nuclear weapons fallout was 3 times higher than from Chernobyl debris for Swedish urban populations. This difference is attributed mainly to the different time-patterns of the fallout, where the main fraction of nuclear weapons fallout occurred continuously over the spring seasons, giving rise to repeated short term transfer of  $^{137}\text{Cs}$  to humans. Some part of this difference is also likely explained by the increasing awareness of radioecological transfer pathways by the authorities and the public.

- For reindeer herders the time integrated transfer factor of nuclear weapons fallout was almost 10 times higher than for the Chernobyl fallout. A large part of this difference is attributed to the remedial actions undertaken and recommendations by the regulatory authorities regarding slaughter and distribution of contaminated reindeer after the Chernobyl fallout in 1986.
- The time-integrated transfer factor for Chernobyl  $^{137}\text{Cs}$  appears to be more than 25 times higher for reindeer herders in Sweden than for the urban reference groups. Hunters exhibit, on average, a factor of 3 times higher values, and farmers a factor of 2 times higher.
- The projected committed effective dose from internal contamination of Chernobyl  $^{137}\text{Cs}$  per unit activity deposition is observed to be 17–28  $\mu\text{Sv}/\text{kBq m}^{-2}$ . The highest values in Sweden are obtained for reindeer herders with an estimated radioecological transfer of 700  $\mu\text{Sv}/\text{kBq m}^{-2}$ .
- The deposition values,  $A_{cum}(t)$ , averaged over the counties in Sweden appears to reflect the transfer and uptake in urban humans living in the county, rather than the average deposition values in the municipalities.

## APPENDIX A.1

Reconstruction of the geographical and temporal variation of the deposition of  $^{137}\text{Cs}$  in Sweden before and after the Chernobyl accident.

(Table A.1-1, Table A.1-2 and Table A.1-3)

## APPENDIX A.2

Methods of calculating the time-integrated aggregate transfer factor from ground deposition to man of radiocaesium

(Table A.2-1)

## REFERENCES

- Aarkrog A, Bøtter-Jensen L, Chen QJ, Clausen J, Dahlgard H, Hansen H, Holm E, Lauridsen B, Nielsen SP, Strandberg M, Søgaard-Hansen J. Environmental radioactivity in Denmark in 1992 and 1993. Risø-Report R-756(EN), Roskilde, Denmark, 1995.
- Aarkrog A. The radiological impact of the Chernobyl debris compared with that from nuclear weapons fallout. *J. Environ. Radioactivity* 1988;6:151-162.
- Ågren, G. Transfer of radiocaesium to the Swedish Population and subgroups of special interest. Thesis, Swedish University of Agricultural Sciences, Uppsala, Sweden, 1998.
- Byström S. Cesium-137 beräknat ur flygmätningar utförda av SGU/SGAB från och med 1986 tom 2000, på uppdrag av SSI; Statens strålskyddsinstitut. Report to Swedish Radiation Protection Authority, Project: SSI P1075.98, SGU dnr 08-783/98, Swedish Geological Airborne Division, Uppsala, Sweden, 2000 (in Swedish).
- Byström S. Technical Report about the airborne gamma ray survey performed in Sweden after the Chernobyl accident in April 1986. Part One. SGU-report TFRA9005, Swedish Geological Airborne Division, Uppsala, Sweden, 1990.
- Edvarson K. Results from a research programme on environmental radioecology. In: Moberg L, editor. The Chernobyl fallout in Sweden, Results from a research programme on environmental radioecology. The Swedish Radiation Protection Authority, SSI, Stockholm, Sweden, 1991; 47-65.
- Falk R., Eklund G, Giertz H, Östergren I. Cesium in the Swedish population after Chernobyl: Internal radiation, whole-body counting. In: Moberg L, editor. The Chernobyl fallout in Sweden, The Swedish Radiation Protection Authority, SSI, Stockholm, Sweden, 1991; 547-577.
- Finck, R. High resolution field gamma spectrometry and its application to problems in environmental radiology. 1992; Thesis, Depts of Radiation Physics, Malmö and Lund, University of Lund, SE-205 02 Malmö, Sweden.
- Fredriksson L, Garner RJ, Russell S. Radioactivity and the human diet, 1st ed., Pergamon Press Ltd., Scotland, 1966, 317-352 pp.
- Hanson WC, Watson DG, Perkins RW. Concentration and retention of fallout radionuclides in Alaskan arctic ecosystems. In: Åberg B, Hungate FP, editors. Radioecological concentration processes, Proceedings of an International symposium, Pergamon Press, Stockholm, Sweden, 1966; 233-245.
- Howard BJ, Hove K, Strand P, Pronevich V. Aggregated transfer coefficients: A simple approach to modeling transfer of radionuclides to food products from semi-natural ecosystems, International Atomic Energy Agency Report-SM-339/20, IAEA, Vienna, 1995; 247-258.
- ICRP, International Commission on Radiological Protection. Recommendations of the International Commission on Radiological Protection. Oxford: Pergamon Press; ICRP Publication 60, 1991; Ann. ICRP 21:1-3; 1990.
- Isaksson M, Erlandsson B. Investigation of the distribution of <sup>137</sup>Cs from fallout in the soils of the city of Lund and of province of Skåne in Sweden. *J. Env. Radioactivity* 1998; 38:105-131.
- Johansson L, Ågren G. Distribution of radioactive cesium in the population of Northern Sweden 1988 – 1993. *Radiat. Prot. Dosimetry*, 1994; 62:131–142.
- Lantmäteriet – Land and geographic information, Swedish geodetic reference frames, SWEREF 99, RT 90 and RH 70, Website accessed in 2003, <http://www.lm.se/geodesi/refsys/eng/refsys-eng.htm>.
- Leggett R. Metabolic data and retention functions for the intracellular alkali metals. Report Nureg / CR-3245 ORNL/TM-8630, U.S. Nuclear Regulatory Commission, Washington, DC, 1983.
- Leggett R, Eckerman K, Dunning D, Christy M, Crawford-Brown D, Williams L. Dose rates to organs as a function of age following internal exposure to radionuclides. Washington, DC: Division of Facility Operations. Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission; Report Nureg/CR-3245 ORNL/TM-8265; 1984.
- Lidén K, Gustafsson M. Cs-137 levels of different population groups in Sweden. *Acta Radiol. Suppl.* 1966; 254:38-46.
- Lidén K. Cesium-137 burdens in Swedish Laplanders and reindeer. *Acta Radiol.* 56:237-241, (1961).
- Lindgren J, Hubbard L. Microsoft Excel™ calculation spreadsheet produced by the Swedish Radiation Protection Authority, Sweden, 2002.
- Miettinen JK, Jokelainen A, Roine P, Lidén K, Naversten, Y. Cesium-137 and potassium in Finnish Lapps and their diet. *Radiological Health Data* 1964; 5:83-97.



- Möre H, Becker W, Falk R, Brugård-Konde Å, Swedjemark GA. Matkorgsundersökning under hösten 1994. SSI-report 95-22, Swedish Radiation Protection Authority, Stockholm, Sweden, 1994 (in Swedsh).
- Räaf CL, Hubbard L, Falk R, Ågren G, Vesanen R. Ecological half-time and effective dose from Chernobyl debris and nuclear weapons fallout of  $^{137}\text{Cs}$  as measured in different Swedish populations. To be published.
- Räaf CL. Human metabolism and ecological transfer of radioactive caesium - Comparative studies of Chernobyl debris and nuclear weapons fallout in Southern Sweden and in Bryansk, Russia. Thesis, Lund university, Lund, Sweden, 2000.
- SCB, Statistics Sweden. Official website: <http://www.scb.se>. Data obtained from database of SCB <http://www.ssd.scb.se/databaser>. Accessed in September 2001.
- Soumela M, Rahola T. Studies in environmental science 62, Nordic radioecology: The transfer of radionuclides through Nordic ecosystems to man. Elsevier Science Publishing Co., Roskilde, Denmark, 1994, 7-20 pp.
- Swedish Government, The Swedish environmental objectives – Interim targets and action strategies. Swedish Governmental Bill 2000/01:130, Sweden, 2001.
- Swedish Meteorological and Hydrological Institute (SMHI), Microsoft Excel calculation spreadsheet purchased by the SMHI, Norrköping, Sweden, 2002.
- United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR. Sources and effects of ionising radiation, 1977 Report to the General Assembly, Annex C. United Nations Publication, New York, 1977.
- United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR. Sources and effects of ionising radiation, 1982 Report to the General Assembly, Annex E. United Nations Publication, New York, 1982.
- Vintersved I, Arntsing R., Bjurman B. Resuspension of radioactive caesium from the Chernobyl accident. In: Moberg L, editor. The Chernobyl fallout in Sweden. Results from a research programme on environmental radioecology. The Swedish Radiation Protection Authority, SSI, Stockholm, Sweden, 1991; 85-106.
- Wright SM, Howard BJ, Strand P, Nylén T, Sickel MAK. Prediction of  $^{137}\text{Cs}$  deposition from atmospheric nuclear weapons tests within the Artic. *Env. Pollution*, 1999; 104:131-143.
- Wright SM, Strand P, Sickel MA, Howard BJ. Spatial variation in the vulnerability of Norwegian Arctic countries to radiocaesium deposition. *Sci. Total Environ*, 1997; 202:173-184.
- Ågren, G. Transfer of radiocaesium to the Swedish Population and subgroups of special interest. Thesis, Swedish University of Agricultural Sciences, Uppsala, Sweden, 1998.



Table 1

Overview of the populations considered in this study.

Category	Population	Code	Period
Reindeer herders	Reindeer herders in Västerbotten county (most affected county from the Chernobyl fallout in 1986)	SAMI_N1	1986 – 2001
	Reindeer herders in Norrbotten county (moderate or no fallout from Chernobyl)	SAMI_N2	1992, 1997
	Reindeer herders in Jämtland county (relatively high deposition from Chernobyl fallout)	SAMI_N3	1992-2001
	Reindeer herders in the middle of Sweden	SAMI_W1	1965 – 1976
Hunters	Hunters in various municipalities in the counties most affected by the Chernobyl fallout.	HUNTER	1994 – 2001
Farmers	Farmers living in villages in Gävle municipality (own housekeeping).	FARMERS	1986 – 1998
Rural non-farmers	Rural non-farmers living in villages in Gävle municipality.	RUR_NONFARM_M1	1986 – 1998
	Non-Sami populations living in rural municipalities in Västerbotten county in the north of Sweden	RUR_NONFARM_N1	1991 – 1996
	Non-Sami populations living in rural municipalities in Norrbotten county in the north of Sweden	RUR_NONFARM_N2	1991 – 1996
Urbans	Urban population in Stockholm in the middle of Sweden	URB_M1	1959 – 2001
	Urban population in Göteborg in the west of Sweden	URB_W1	1986 – 1989
	Urban population living in the vicinities of city of Lund, in the south of Sweden	URB_S1 <sup>a</sup>	1960 – 1994

	Urban population living in the vicinities of city of Malmö, in the south of Sweden	URB_S2	1986 – 1994, 2002
Mixed population	National survey of subjects (randomly selected subjects in Sweden; stratified for the regional variation in $^{137}\text{Cs}$ -deposition)	REF_NAT	1987, 1988 <sup>b</sup> and 1994

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<sup>a</sup> A complete evaluation of the *in-vivo* body burden of this study group was carried out previously by Rääf in 2000.

Table 2

Projected time integrated aggregate transfer factor of  $^{137}\text{Cs}$  as defined by UNSCEAR from nuclear weapons fallout (1945 to 2015) and the Chernobyl fallout (1986 to 2056) and the internal effective dose commitment between 1986 and 2056 per unit deposition in the county ( $\mu\text{Sv kBq m}^{-2}$ ). Uncertainty estimates refer to  $\pm 1$  standard deviation of the estimated population mean values (See also Table A.2-1).

Population	$T_{\text{ag,int,UN}}$		$E_{\text{ag,int}}$	
	[Bq (gK) $^{-1}\text{y}/(\text{kBq m}^{-2})$ ]		[ $\mu\text{Sv}/\text{kBq m}^{-2}$ ]	
	1945-2015	1986-2056	1945-2015	1986-2056
FARMERS		13.4 $\pm$ 3.7		55 $\pm$ 15
HUNTERS		26 $\pm$ 6.5		106 $\pm$ 26
RUR_NONFARM_M1		6.2 $\pm$ 1.9		24 $\pm$ 7
RUR_NONFARM_N1		16 $\pm$ 4.0		39 $\pm$ 10
RUR_NONFARM_N2		59 $\pm$ 15		150 $\pm$ 38
SAMI_N1		180 $\pm$ 76		670 $\pm$ 280
SAMI_N2		300 $\pm$ 75		780 $\pm$ 190
SAMI_W1	2640 $\pm$ 590		10500 $\pm$ 2300	
URBAN_M1	14 $\pm$ 2.7	7.9 $\pm$ 2	81 $\pm$ 17	28 $\pm$ 7.5
URBAN_S1-2	18.5 $\pm$ 4.0	5.0 $\pm$ 1.7	58 $\pm$ 11	17 $\pm$ 6
URBAN_W1		7.3 $\pm$ 1.8		26 $\pm$ 13
Denmark <sup>b</sup>	11			
Argentina (1966-1974) <sup>b</sup>	11			

<sup>a</sup> Dose values refer to an adult male of 70 kg body size.

<sup>b</sup> Values taken from UNSCEAR (1977)

Table 3

Modified time-integrated aggregate transfer of  $^{137}\text{Cs}$  [ $\text{Bq (gK)}^{-1}\text{y}/(\text{kBq m}^{-2})$ ] normalized to natural potassium for nuclear weapons fallout and Chernobyl debris as defined by Eq. 4. Uncertainty estimates refer to  $\pm 1$  standard deviation of the estimated population mean values (See also Table A.2-1).

Population	Global Fallout (1945-2015)		Chernobyl Fallout (1986-2056)	
	Municipality	County	Municipality	County
	Average		Average	
FARMERS			4.3 $\pm$ 1.1	16 $\pm$ 4.3
HUNTERS			11 $\pm$ 2.7	30.6 $\pm$ 7.6
RUR_NONFARM_M1			1.9 $\pm$ 0.6	7.2 $\pm$ 2.1
RUR_NONFARM_N1			7.1 $\pm$ 2.0	15 $\pm$ 5.4
RUR_NONFARM_N2			49 $\pm$ 12	48 $\pm$ 12
SAMI_N1			115 $\pm$ 74	200 $\pm$ 83
SAMI_N2			240 $\pm$ 60	320 $\pm$ 80
SAMI_W1	3200 $\pm$ 640	3300 $\pm$ 660		
URBAN_M1	33 $\pm$ 6.6	32 $\pm$ 6.5	14 $\pm$ 4	9.6 $\pm$ 2.6
URBAN_S1-2	27 $\pm$ 5.4	27 $\pm$ 5.5	7.2 $\pm$ 2.6	7.0 $\pm$ 2.4
URBAN_W1			7.5 $\pm$ 2.6	5.8 $\pm$ 2.6

Table 4

Relative variance (1 standard deviation/mean value) of the aggregate transfer  $T_{ag}$  (Eq. 3) for Chernobyl fallout  $^{137}\text{Cs}$  in the reindeer herders of Västerbotten county (SAMI\_N1) using  $^{137}\text{Cs}$  concentration and potassium normalization, respectively.

Year of survey	Municipality average deposition		County average deposition	
	$q_{\text{Cs}}$	$^{137}\text{Cs}/\text{K}$	$q_{\text{Cs}}$	$^{137}\text{Cs}/\text{K}$
1994	0.85	0.95	0.96	0.92
1997	0.86	0.79	0.93	0.76
1998	0.89	1.24	1.02	1.31





## FIGURE CAPTIONS

Fig. 1

Conceptual model for the transfer of radionuclides from atmospheric fallout to man based on UNSCEAR 1982.

Fig. 2

Location of the various populations investigated by whole-body counting in Sweden.

Fig. 3

Cumulated  $^{137}\text{Cs}$  deposition,  $A_{\text{cum}}$ , from global fallout corrected for continuous physical decay in Denmark and Sweden ( $\Delta$ =estimate from Wright et al., 1999). Solid line indicates average deposition according to geographical variation and fitted time-function  $TP(t)$  in Sweden. Dashed lines indicate  $\pm 1$  sigma.

Fig. 4

Estimation of ground deposition of  $^{137}\text{Cs}$  from nuclear weapons fallout as per 1 July 1985 (prior to the Chernobyl fallout) and Chernobyl fallout as per 1 May 1986.

Fig. 5

Aggregate transfer of  $^{137}\text{Cs}$ , as defined in Eqs 4-5, from ground deposition of Chernobyl fallout to various Swedish populations after 1986, using municipality averages (left panel) and county averages (right panel).

Fig. 6

Correlation between average deposition in municipality and body burden of  $^{137}\text{Cs}$  and  $^{137}\text{Cs}/\text{K}$ -ratio respectively according to the national survey in 1987.

Fig. 7

Correlation between average deposition in county and body burden of  $^{137}\text{Cs}$  and  $^{137}\text{Cs}/\text{K}$ -ratio respectively according to the national survey in 1987.



Figure 1.

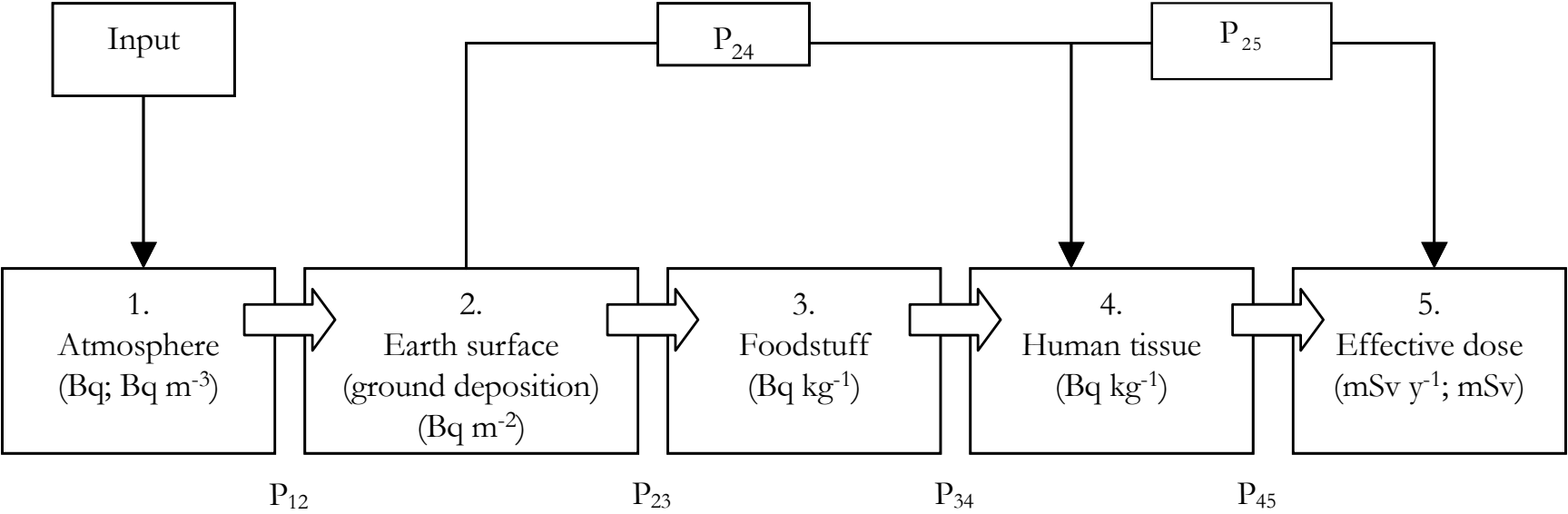


Figure 3.

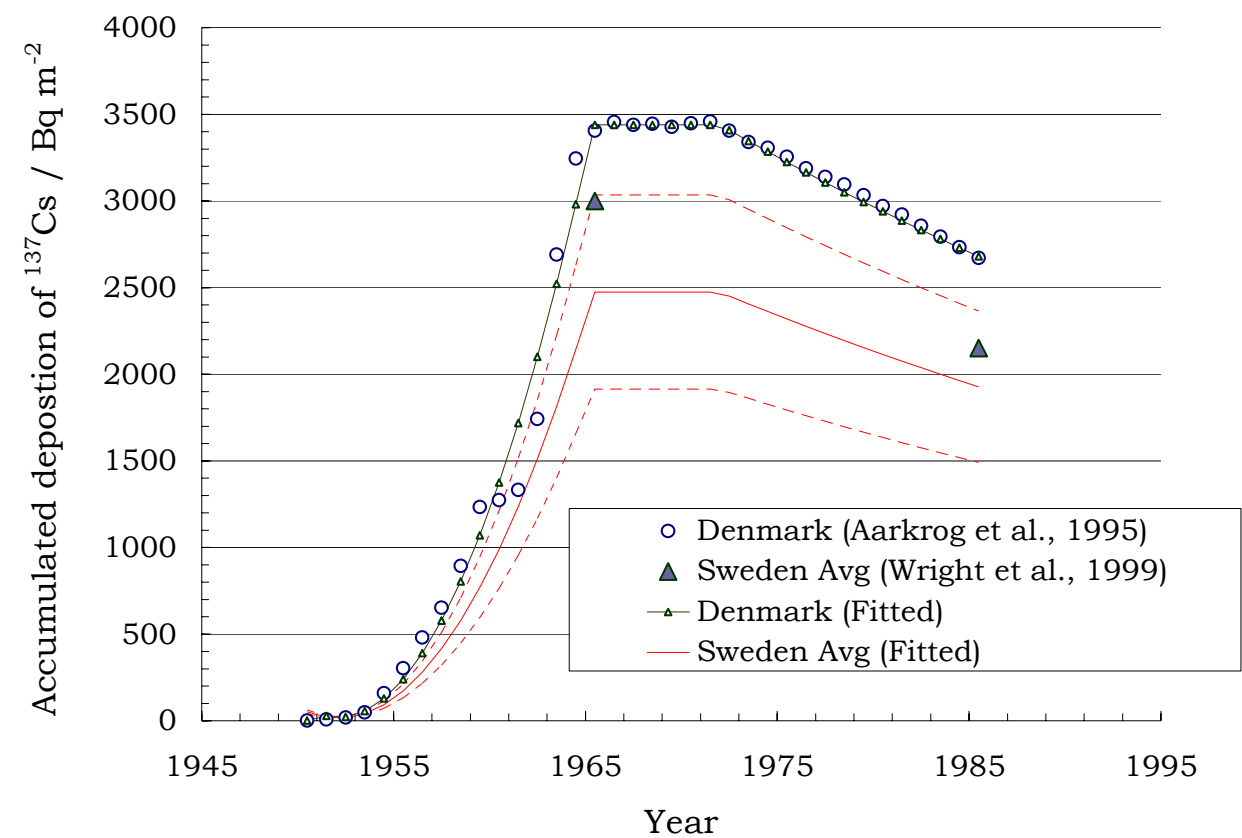






Figure 2.

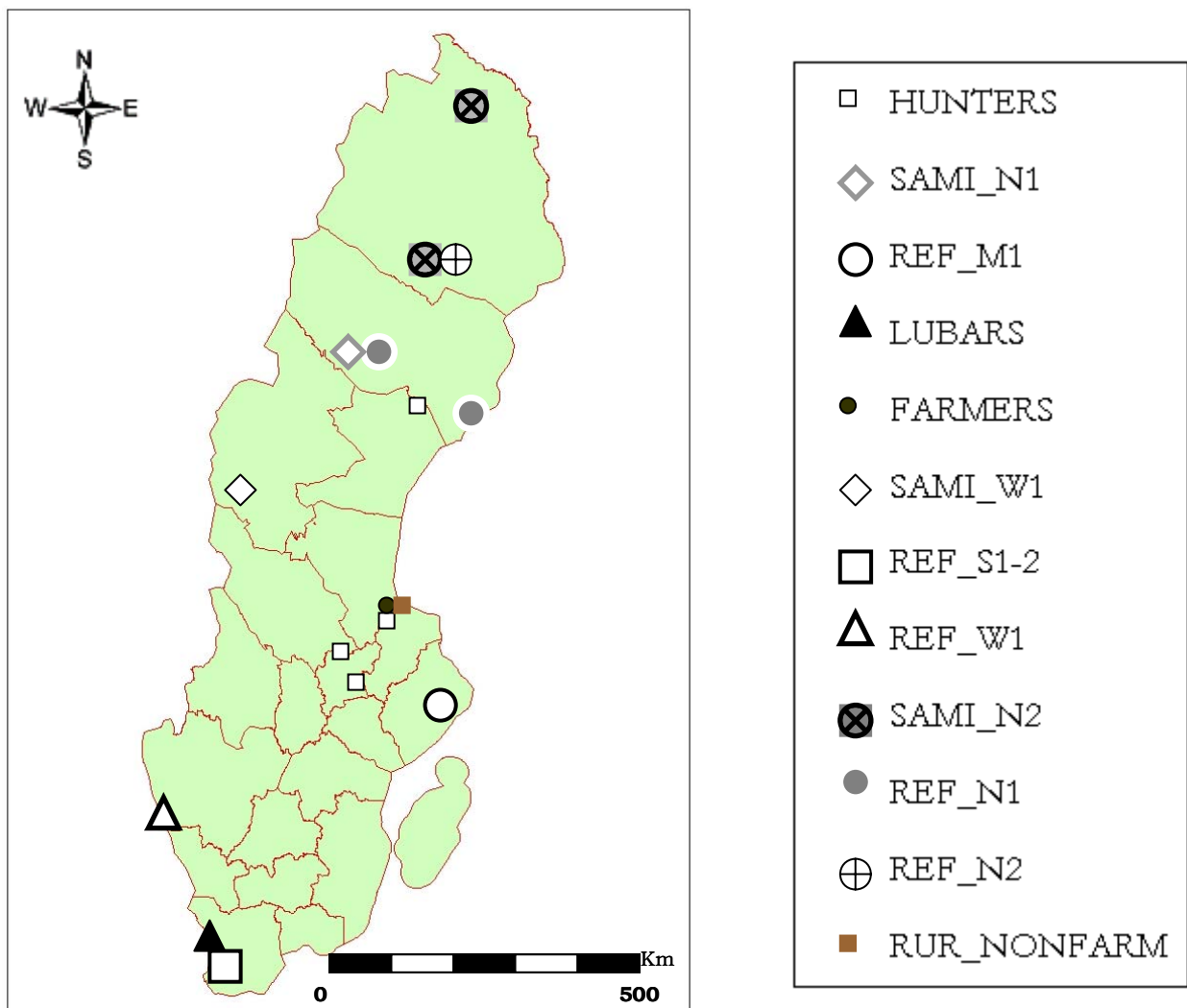


Figure 4.

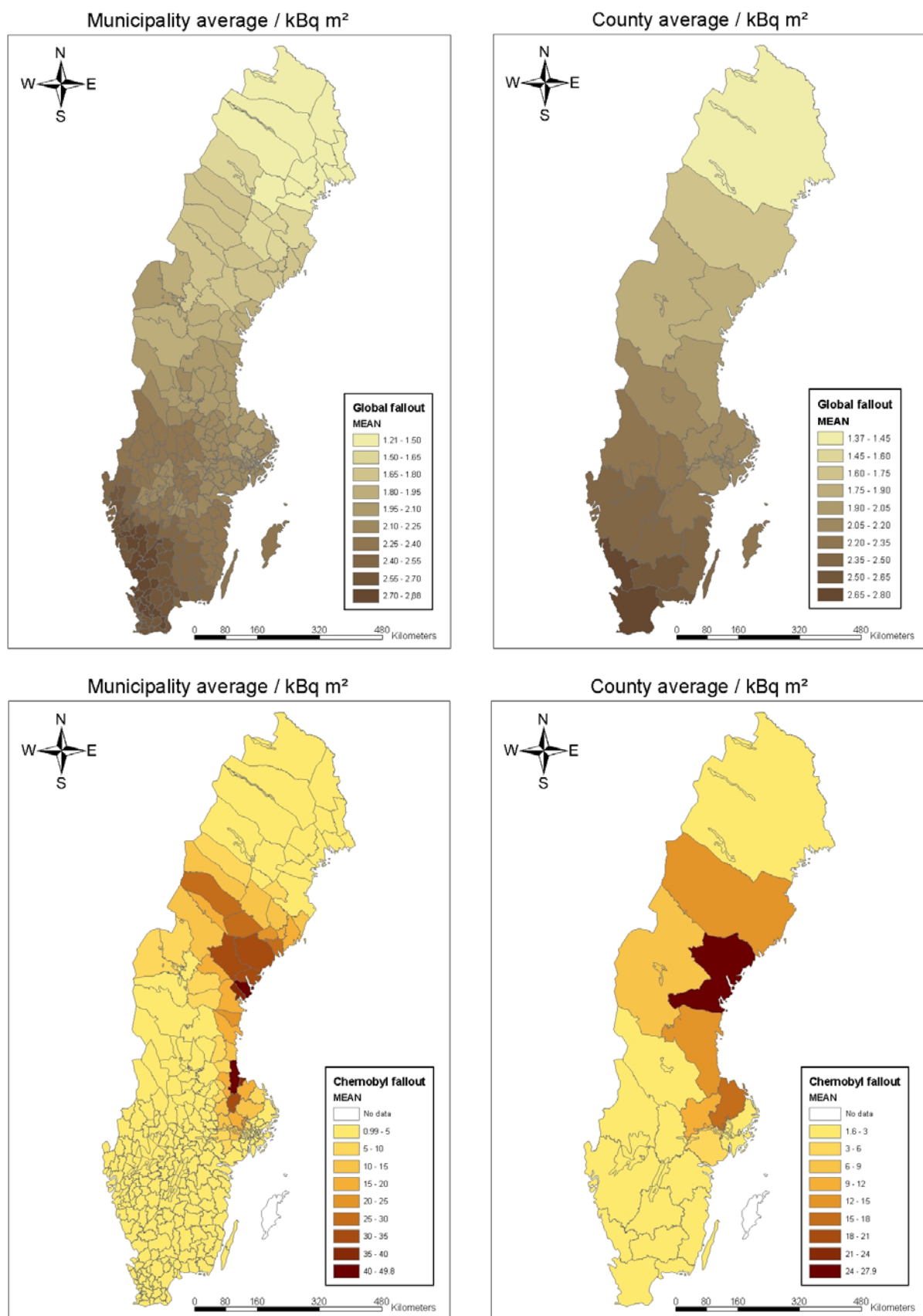






Figure 5.

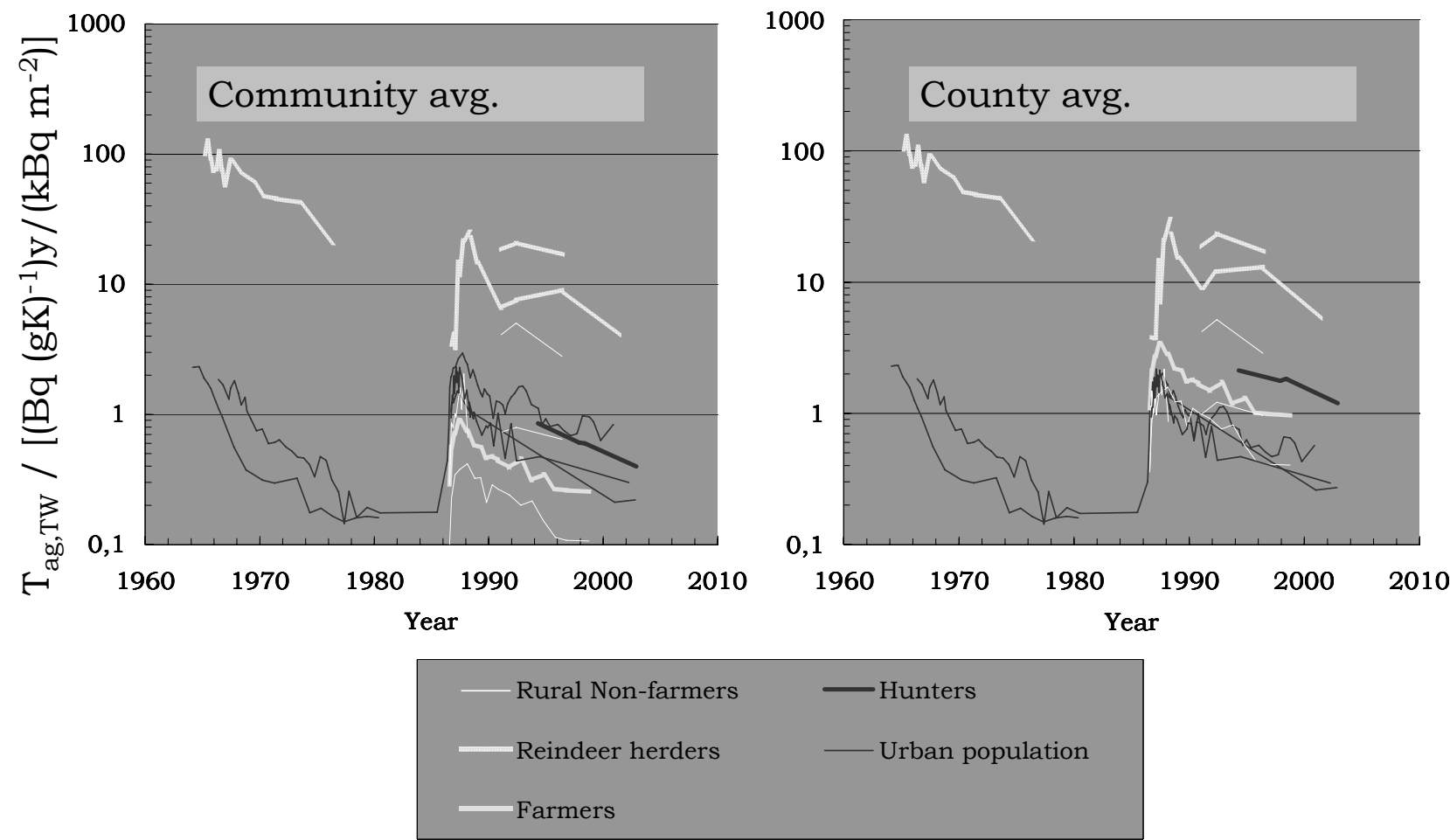


Figure 6.

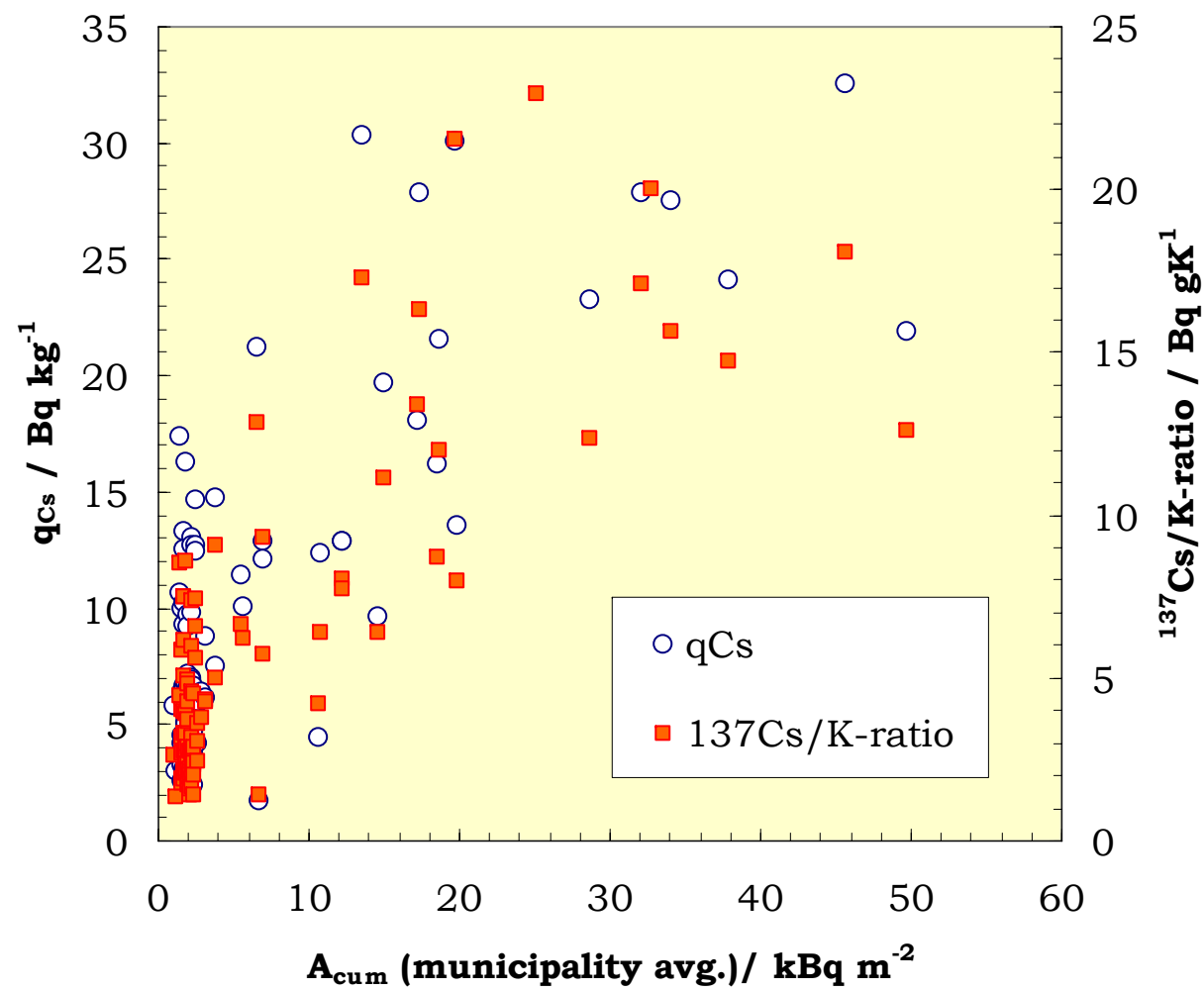


Figure 7

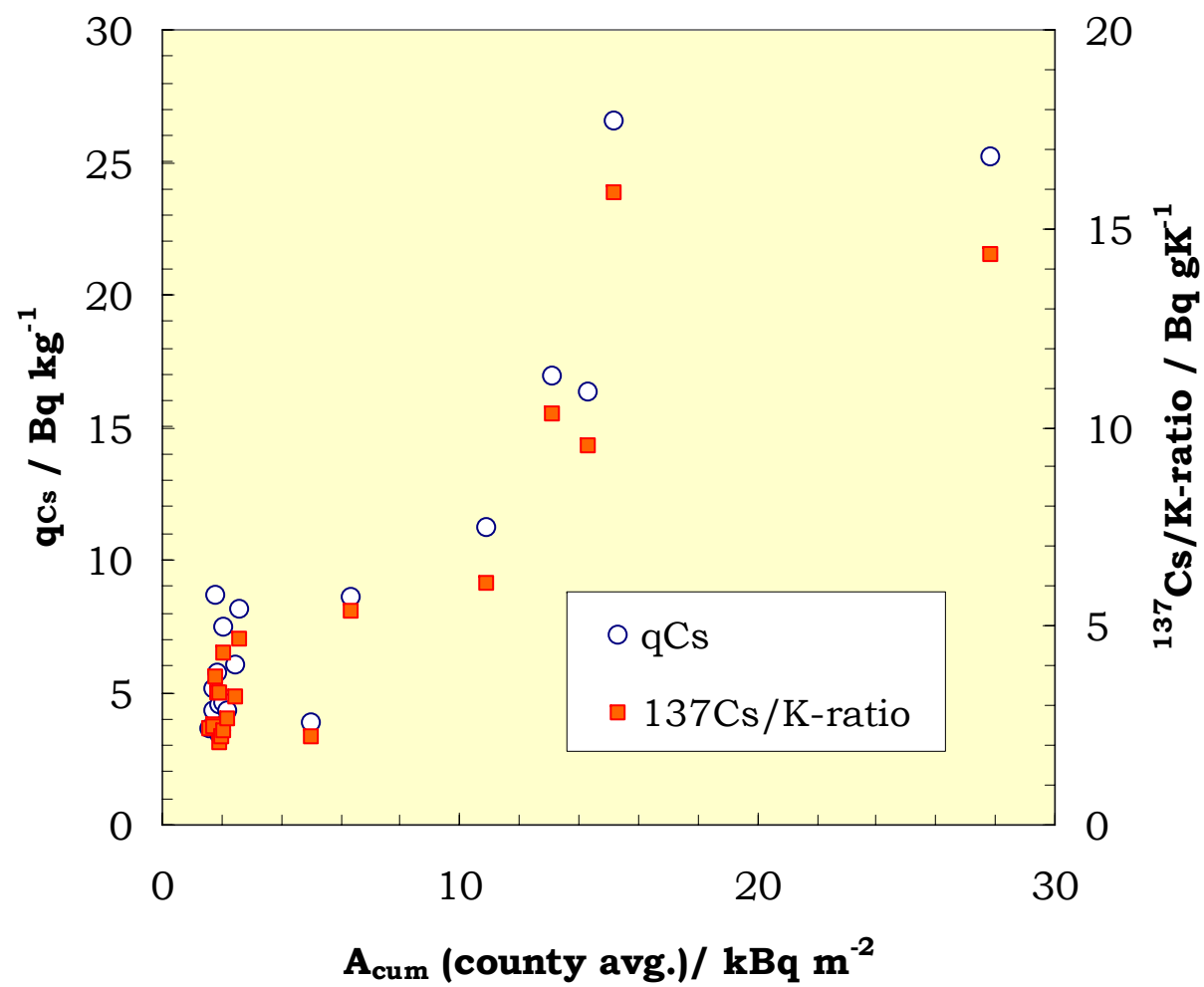


Table A.1-1

Reconstruction of the geographical variation of the **Nuclear Weapons Fallout deposition of  $^{137}\text{Cs}$**  in Sweden at date 30 June, 1985.

Cumulated total deposition,  $A_{cum}(t=1985.5)$  in 1985 of  $^{137}\text{Cs}$  ( $\text{kBq m}^{-2}$ ) based on soil sample data from seven locations in Sweden taken from Vintersved et al., 1991. Data is separately presented in Table A.1-2.



Average annual precipitation in  $\text{mm y}^{-1}$ ,  $Prec(x)$ , during 1960-1990 over a grid of  $5 * 5 \text{ km}^2$  in RT90-coordinates ( $x=RT_{North}, RT_{East}$ ) for the whole of Sweden acquired from the SMHI (SMHI, 2001).



Multiple regression of  $A_{cum}(t=1985.5)$  shown in Table A.1-2:

$$A_{cum}(t=1985.5)=a_0+a_1*RT_{North}+a_2*Prec(RT_{North}, RT_{East}).$$

Regression estimates ( $\pm 1$  Standard asymptotic error) and goodness of fit, Pearsons  $R^2$ :

$$A_{cum}(RT_{North}, RT_{East})=7720(\pm 1380)-0.948(\pm 0.165)\times 10^{-3}\times RT_{North}+1.156(\pm 0.715)\times Prec(RT_{North}, RT_{East}); R^2=0.902$$

Estimated variance of  $A_{cum}(t=1985-07-01) \sim \text{SSR}/N$  of regression = 4%. Model uncertainty is assumed to be significantly higher.



Grid of estimated  $A_{cum}(t=1985.5)$  based on annual precipitation and  $RT_{North}$  coordinate over  $5 * 5 \text{ km}^2$  is processed to yield a map of municipality and county average values in Sweden (Fig. 4).



A time-pattern factor,  $TP$ , is introduced in order to obtain the pre-Chernobyl deposition at an arbitrary time,  $t$ :

$$A_{cum}(t) = A_{cum}(t=1985.5) \cdot TP(t).$$

$TP(t)$  is a time dependent function that is a fit of the time-pattern of the annual fallout between 1950 and 1985 as recorded by Risö laboratory in Denmark

(Aarkrog et al., 1995); See also Fig. 3.

$$= 1.29 \times e^{-(\ln 2 / 37.5) \times (t - 1972)}; t > 1972$$

$$TP(t) = 1.29; 1965 < t < 1972$$

$$= 0.0404 - 0.0309 \times (t - 1950) + 0.00725 \times (t - 1950)^2; t < 1965$$

Variance in  $TP(t)$  due mainly to model uncertainty; assumed to be in the order of 10%.

↓

Nuclear weapons fallout as deposition activity in a specific municipality (or county) at a specific time can be obtained;

$$A_{cum}(t, Municipality/County) = TP(t) \cdot A_{cum}(1985.5, Municipality/County).$$

Table A.1-2

Cumulated deposition of  $^{137}\text{Cs}$  at seven locations in Sweden in 1985 as a function of annual precipitation and northern co-ordinate.

Measurement station	North	Annual precipitation	Deposition	Fitted data from multi-regression
	Coordinate	(SMHI, 2001)	$A_{cum}(t=1985-07-01)$ (Vintersved et al., 1991)	
	RT90	mm y <sup>-1</sup>	Bq m <sup>-2</sup>	Bq m <sup>-2</sup>
Kiruna	7536453	518.2	1250	1178
Umeå	7087575	648.9	1750	1755
Östersund	7009825	538.1	1700	1701
Stockholm (Ursvik)	6586746	485.8	2000	2041
Grindsjön	6553528	637.7	2000	2248
Göteborg	6403768	798.4	2600	2576
Ljungbyhed	6218854	630.8	2750	2558

Table A.1-3

Reconstruction of the geographical variation of the Chernobyl fallout deposition of  $^{137}\text{Cs}$  in Sweden

Chernobyl fallout in terms of equivalent surface deposition,  $A_{esd}$  (refer to Finck, 1992) in 1986-05-01:

Municipality or county averages of  $A_{esd}$  ( $\text{Bq m}^{-2}$ ) taken from Lindgren and Hubbard, SSI.



Total activity deposition in 1986-05-01:

$$A_{cum}(t=1986.33) = 1.7 * A_{esd}(t=1986.33)$$

$A_{esd}$ -values have been obtained through airborne gamma ray surveys with an approximate relative uncertainty of 10-20% (Byström, 1990).

The factor 1.7 is the national average of the ratio between the equivalent surface deposition and soil sample deposition concentrations of  $^{134}\text{Cs}$  in 17 different locations in Sweden taken from Edvarson, 1991.

Variance in  $A_{cum}$  dominated by the variance in the soil penetration factor:  $1.7 \pm 0.4$  (1 SD).



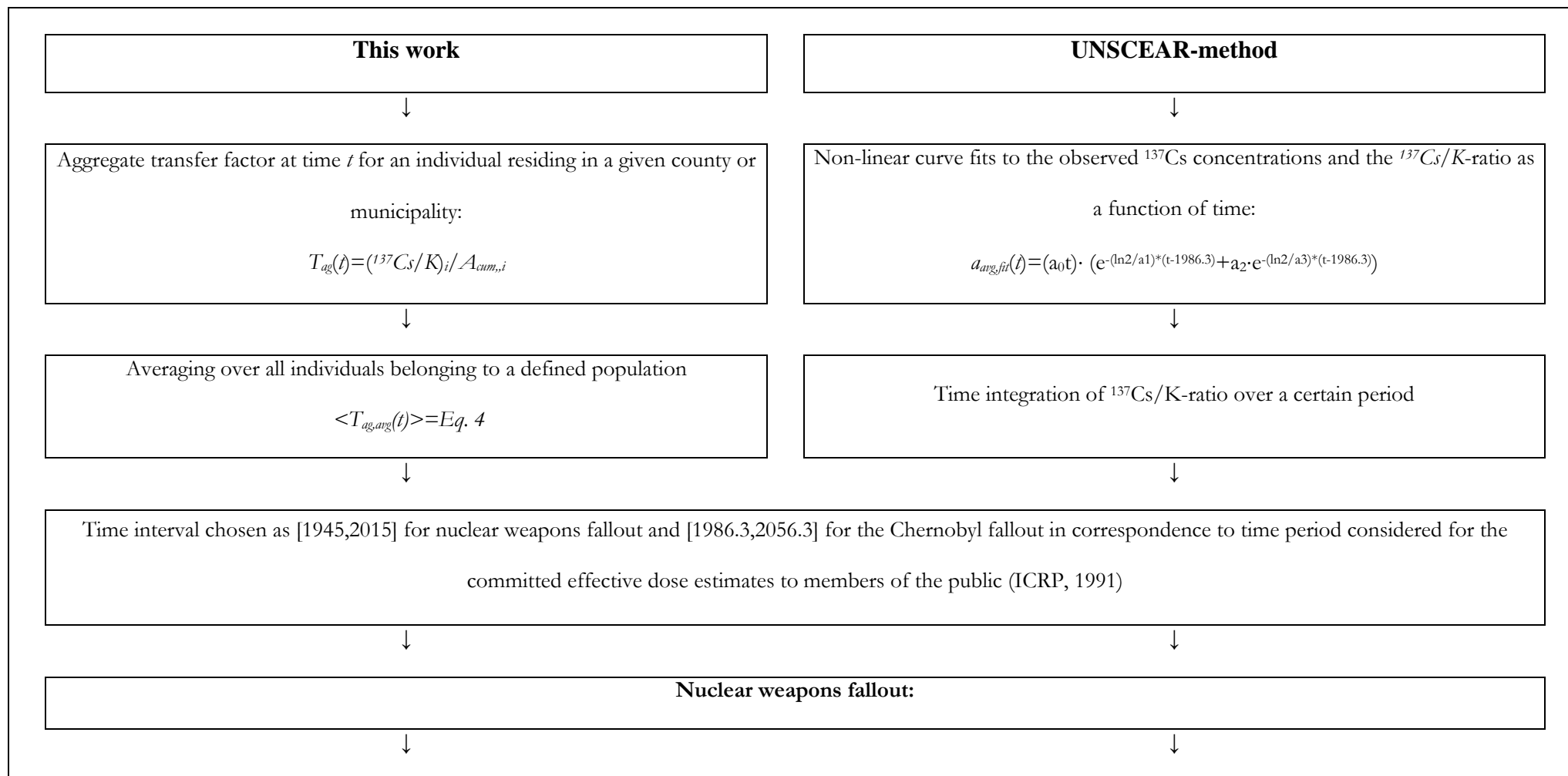
Chernobyl fallout at time  $t$  after 1986-05-01 in a given county or municipality can be estimated:

$$A_{cum}(t,x) = A_{cum}(1986-05-01; \text{Municipality or County}) * e^{-(\ln 2 / 30.2) * (t - 1986.33)}$$



Table A.2-1

Two different methods of assessing the Aggregate transfer of the Nuclear Weapons and Chernobyl Fallout  $^{137}\text{Cs}$



*Contribution 1945-1960(ca):*

Unknown deposition and average  $^{137}\text{Cs}/K$ -ratios in the 1940s and 1950s must be reconstructed using a scaling of  $\Sigma A_{ann}$  between 1960 and 1985 and between 1950 and 1985 using available annual fallout from Denmark (Aarkrog et al., 1995).

Additional fallout assumed negligible.

$$\int_{1945}^{1960(ca)} T_{ag,avg} dt = \frac{\sum_{1960}^{1985} A_{ann}(t)}{\sum_{1950}^{1985} A_{ann}(t)} \cdot \sum_{1960(ca)}^{1985} T_{ag,avg} \cdot \Delta t$$

Estimated variance: Model uncertainty dominates, 1 SD assumed at least 10%.



*Contribution 1945-1960(ca):*

Unknown deposition and average  $^{137}\text{Cs}/K$ -ratios in the 1940s and 1950s must be reconstructed using a scaling of  $\Sigma A_{ann}$  between 1960 and 1985 and between 1950 and 1985 using available annual fallout from Denmark (Aarkrog et al., 1995).

Additional fallout assumed negligible.

$$\int_{1945}^{1960(ca)} (^{137}\text{Cs} / K)_{avg} dt = \frac{\sum_{1960}^{1985} A_{ann}(t)}{\sum_{1950}^{1985} A_{ann}(t)} \cdot \sum_{1960(ca)}^{1985} (^{137}\text{Cs} / K)_{avg} \cdot \Delta t$$

Estimated variance: Model uncertainty dominates, 1 SD assumed at least 10%.



*Contribution 1960(ca)-1986:*

Time integration using trapezoid summing of observed  $T_{ag,avg}$  is performed over the time period of body burden investigations before the event of Chernobyl fallout in 1986.

$$\sum_{t=1960(ca)}^{1986} T_{ag,avg}(t) \cdot \Delta t$$

Estimated variance:  $\sim (\Delta q_C / q_C)^2 + (\Delta A_{cum} / A_{cum})^2 + \text{Var}(TP)$

$(\Delta q_C / q_C)$  = relative standard error of the population mean of  $q_C$ ,  $\sim 10\%$ ;

$$\Delta A_{cum} < 5\% (1 \text{ SD})$$

$\text{Var}(TP)$  is dominated by model uncertainty, assumed  $\sim 10\%$  (1 SD)



*Contribution 1960(ca)-1986:*

Time integration using trapezoid summing of observed  $^{137}\text{Cs}/K\text{-ratio}$  is performed over the time period of body burden investigations before the event of Chernobyl fallout in 1986.

$$\sum_{t=1960(ca)}^{1986} (^{137}\text{Cs} / K)_{avg}(t) \cdot \Delta t$$

Estimated variance:  $\sim \text{Var}((^{137}\text{Cs}/K)_{avg}(t)) < 10\%$  (1 SD)



*Contribution 1986-2015:*

Non-linear curve fit to observed  $T_{ag,avg}$  during previous period is used to project the long-term time-integrated  $^{137}Cs/K$ -ratio using exponential fits with 1965 as the peak year:

$$\int_{1986} a_0 \cdot t \left( e^{-\frac{\ln 2}{a1} \cdot (t-1965)} + a_2 \cdot e^{-\frac{\ln 2}{a3} \cdot (t-1965)} \right) dt$$

Estimated variance:  $\sim \int \sigma^2(T_{ag,int,TW})(t) dt + (\Delta A_{cum}/A_{cum})^2$

$\sigma^2(T_{ag,int,TW})$  approximated as constant over time  $1986.3 < t < 2015$

$\sigma^2(T_{ag,int,TW}) \sim ((SS_{res}/N) \text{ 29 years})$

$N = \#$  of observed group mean values

$(\Delta A_{cum}/A_{cum}) = 5\%$



*Contribution 1986-2015:*

Non-linear curve fit to observed  $^{137}Cs/K$ -ratio during previous period is used to project the long-term time-integrated  $^{137}Cs/K$ -ratio using exponential fits with 1965 as the peak year:

$$\int_{1986} a_0 \cdot t \left( e^{-\frac{\ln 2}{a1} \cdot (t-1965)} + a_2 \cdot e^{-\frac{\ln 2}{a3} \cdot (t-1965)} \right) dt$$

Estimated variance:  $\sim \int \sigma^2(^{137}Cs/K)_{avg}(t) dt$

$\sigma^2(^{137}Cs/K)_{avg}$  approximated as constant over time  $1986.3 < t < 2015$

$\sigma^2(^{137}Cs/K)_{avg} \sim ((SS_{res}/N) \text{ 29 years})$

$N = \#$  of observed group mean values



Aggregate transfer for the period 1945 to 2015 is obtained by summing the three contributions of time-integrated  $T_{ag,int}$ .

$$\sum_{1945}^{1960(ca)} reconstructed + \sum_{t=1960(ca)}^{1986} T_{ag,avg}(t) \cdot \Delta t + \int_{1986}^{2015} T_{ag,avg}(t) dt$$

Overall variance in estimated population mean >20% (1 SD)



Aggregate transfer,  $T_{ag,int}$ , is obtained by summing the three contributions of time-integrated  $^{137}\text{Cs}/\text{K}$ -ratios, divided by the total estimated ground deposition,  $\Sigma A_{ann}$  between 1950 and 1985 in the particular county or municipality of the population.

$$\frac{\sum_{1945}^{1960(ca)} reconstructed + \sum_{t=1960(ca)}^{1986} (^{137}\text{Cs} / \text{K})_{avg}(t) \cdot \Delta t + \int_{1986}^{2015} (^{137}\text{Cs} / \text{K})_{avg}(t) dt}{\sum_{1950}^{1985} A_{ann}(t, \text{Municipality} / \text{County})}$$

Overall variance in estimated population mean >20% (1 SD)



### Chernobyl fallout:



Non-linear curve fit to the observed aggregate transfer factor,  $T_{ag,avg}(t)$ :

$$\int_{1986.3}^{2056.3} (a_0 \cdot t) \cdot (e^{\frac{-\ln 2}{a1} \cdot (t-1986.3)} + a_2 \cdot e^{\frac{-\ln 2}{a3} \cdot (t-1986.3)}) dt$$

Estimated variance:  $\sim \int \sigma^2(T_{ag,avg})(t) dt + Var(A_{cum}(1986-05-01))$

$\sigma^2(T_{ag,avg})$  approximated as constant over time  $1986.3 < t < 2056.3 = 70 \text{ years}$ ;

$$\sigma^2(T_{ag,avg}) \sim (SS_{res}/N) \text{ 70 years};$$

$$(\Delta A_{cum} / A_{cum}) = 25\%$$



Non-linear curve fit to observed  $^{137}\text{Cs}/\text{K}$ -ratio:

$$\int_{1986.3}^{2056.3} a_0 \cdot t (e^{\frac{-\ln(2)}{a1} \cdot (t-1986.3)} + a_2 \cdot e^{\frac{-\ln(2)}{a3} \cdot (t-1986.3)}) dt$$

Estimated variance:  $\sim \int \sigma^2(^{137}\text{Cs}/\text{K})_{avg}(t) dt$

$\sigma^2(^{137}\text{Cs}/\text{K})_{avg}$  approximated as constant over time  $t_0 < t < t_0 + T = 70 \text{ years}$ ;

$$\sigma^2(^{137}\text{Cs}/\text{K})_{avg} \sim (SS_{res}/N) \text{ 70 years};$$

↓

↓

Aggregate transfer,  $T_{ag,int}$ , is obtained by dividing the time integration of the mean population  $^{137}\text{Cs}$  activity (or  $^{137}\text{Cs}/\text{K}$ -ratio) by the total Chernobyl deposition of  $^{137}\text{Cs}$  at the time of fallout in the particular county or municipality of the

population:

$$\frac{\int_{1986.3}^{2056.3} a_0 \cdot t \left( e^{-\frac{\ln 2}{a1} \cdot (t-t0)} + a_2 \cdot e^{-\frac{\ln 2}{a3} \cdot (t-t0)} \right) dt}{A_{cum}(1986-05-01, \text{Municipality} / \text{County})}$$

Estimated variance:  $\sim \int \sigma^2(q_{Cs})(t) dt + \text{Var}(A_{cum}(1986-05-01))$

$\sim \sigma^2 q_{Cs}(t) \int dt + \text{Var}(A_{cum}) \sim (\text{SS}_{\text{res}}/\text{N})$  70 years

$(\Delta A_{cum} / A_{cum}) = 25\%$  (1 SD)

↓

***County and Municipality averages of  $T_{ag,int,TW}$  can be compared***

Committed effective dose during 1986 to 2056 per unit activity deposition for an adult of weight  $w$ :

$$E_{ag,int} = \frac{\int_{t0=1986.33}^{2056.33} a_0 \cdot t \left( e^{-\frac{\ln(2)}{a1} \cdot (t-t0)} + a_2 \cdot e^{-\frac{\ln(2)}{a3} \cdot (t-t0)} \right) dt \cdot 0.00140 \cdot w^{0.111}}{A_{dep,Chernobyl}(t = 1986 - 05 - 01, Municipality / County)}$$

Estimated variance is assumed proportional to the relative variation in  $T_{ag,int,UN}$