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Body burden and excretion of ^{137}Cs and ^{40}K in subjects from the South of Sweden

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Abstract

The equivalent biological half-times, T_e, of ^{137}Cs and ^{40}K in a South-Swedish urban population have been determined through whole-body measurements and urinary excretion analysis. The T_e - values for ^{137}Cs found in males were on average, significantly lower than what is given in the literature. The relatively low average whole-body content of ^{40}K, Q_K, in the males could explain the discrepancy, taking into consideration that a positive correlation between the T_e and Q_K has been suggested in an earlier work. Furthermore, the potassium-normalized caesium urinary excretion was determined for the subjects in the study, and values were found to be in accordance with earlier results. A literature study of previous experimental data on the potassium-normalized caesium excretion however raises some questions about its applicability as a method for estimating the whole-body burden of ^{137}Cs through urine analysis. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: ^{137}Cs; Radiocaesium; Excretion; Whole-body monitoring; Humans

1. Introduction

The formation of the Lund reference group dates back to the beginning of the sixties, when Sweden was subjected to the global fall-out caused by frequently performed nuclear weapons tests in the atmosphere. The group consisted of individuals of different age and gender, living and working in the vicinities of Lund in the south of Sweden. The aim of the group at that time was to monitor the time development of the internal contamination levels of fall-out caesium in a population

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representative for the particular geographical area (Bengtsson, 1967). Regular whole-body measurements were performed on the Lund reference group until 1980, when the average whole-body activity levels of fallout ^{137}Cs had decreased to less than 1 Bq per kg body weight, which was an order of magnitude lower than the maximum value obtained in the middle of the sixties (Lidén & Gustafsson, 1967; Bengtsson, 1967; Mattsson et al., 1989).

In 1986, the Chernobyl accident led to a new dispersion of radiocaesium (^{137}Cs, $T_{1/2} = 30.0$ a and ^{134}Cs, $T_{1/2} = 2.06$ a) into the atmosphere. The deposition of ^{137}Cs over the Lund area was moderate (about 2 kBq/m^2) in comparison with areas in the northern part of Sweden, but was still sufficiently high to motivate a repeated measurement series of the whole-body burden of radiocaesium in the Lund reference group. The Lund reference group was reassembled in spring 1987 and repeated whole-body measurements were performed until spring 1994. This time, the study also included collection of 24-h urine samples in order to relate the urinary excretion rate to the whole-body content by means of the equivalent biological half-time, T_e, as defined in the literature (ICRP, 1989).

The primary aim of this work was also to calculate the equivalent biological half-time of ^{137}Cs as well as the so-called potassium-normalized caesium excretion and investigate if any correlations could be found with respect to age, sex and body composition (measured through whole-body content of potassium and/or 24-h creatinine excretion) in the Lund reference group during the second study period (1987–1994). Results from the Lund reference group were compared with a number of similar previous studies on different population groups through out the world; e.g. Clemente, Mariani and Santaroni (1971), Falk, Eklund, Giertz and Östergren (1991), Heinrichs, Paretzke, Voight and Berg (1989), Johansson and Ågren (1994), Johansson, Wickman Ågren, Eriksson, Jonsson and Travelin (1995), Lebedev and Yakovlev (1994), Lessard, Miltenberger and Greenhouse (1980), Lessard, Miltenberger, Cohn, Musolino and Conard (1984), Lloyd, Mays, Church, Pendleton and Mays (1970), and Uchyiama, Iinuma and Saiki (1969), where correlations between whole-body content of ^{137}Cs and urinary excretion data have been investigated. Both the equivalent biological half-time and the potassium-normalized caesium excretion are parameters which are useful when performing independent estimations of whole-body burdens of ^{137}Cs by gamma spectrometric analysis of urinary samples collected from subjects of a certain population group of interest.

2. Material and methods

2.1. Studied subjects

Between 1987 and 1994, about 5–10 repeated whole-body measurements were carried out on each individual of the Lund reference group. For 16 adult subjects (seven females and nine males) of that group 24-h urine samples had been collected in connection to the whole-body measurements. It should be noted that only those individuals who have been subjected to urinary sampling have been included in this work.

The participants' age during the study period ranged from 28 to 77 years. The median age for the female members of the group during the survey was 50 years and the corresponding figure for the male members was 65 years.

Each of the 16 investigated subjects was interviewed at the time of the whole-body measurement in order to be certain that none of them was under heavy medication which could affect the measurements. Extraordinary health conditions were also recorded.

2.2. Equivalent biological half-time

The ICRP model (ICRP, 1979, 1989) for the whole-body kinetics of caesium is assumed to be representative for an adult worker and the expression for the whole-body retention of caesium, $R(t)$, as a function of time after a single intake, is based on experimental data from both sexes (see Eq. (1)). It contains two components, of which the more rapid one (half-time $T_1 = 2$ d, 10% fractional retention) is related to the accumulation of caesium in the kidneys from the blood pool within hours of intake. The caesium is then directly excreted from the kidneys via the urine. The second and long-term component (half-time $T_2 = 110$ d, 90% fractional retention) describes the retention of caesium by mainly the skeletal muscle tissue from which it is excreted mostly through urine. The total whole-body uptake as a fraction of a single intake is given by f_1 and for ingestion of caesium (fraction transferred caesium from GI tract to blood) this fraction is estimated by ICRP (1989) to be unity, that is $f_1 = 1$.

$$R(t) = f_1(0.1\mathrm{e}^{-\ln 2t/T_1} + 0.9\mathrm{e}^{-\ln 2t/T_2}). \tag{1}$$

Note that this relation refers to stable caesium but can within reasonable accuracy also be applied on the long-lived isotope ^{137}Cs, since its physical half-life by far exceeds T_1 and T_2. If the intake pattern of radiocaesium is continuous, or consists of repeated intakes at a relatively constant rate over a time extending well beyond the longer half-life T_2, a quasiequilibrium is obtained between the whole-body content and caesium excretion (Johansson & Ågren, 1994). Since the intake rate as a function of time for the Lund reference group is unknown, the so-called equivalent biological half-time, T_e, as defined by e.g. ICRP (1989), is used instead as a quantity describing the caesium metabolism. The equivalent biological half-time for caesium, T_e (days), is defined as the ratio between the observed whole-body content, Q (Bq), of a given caesium isotope and the observed total excretion rate, E_{tot} (Bq per day), with all excretion pathways included; urine, feces and transpiration, multiplied by $e_{\log} 2$ (see Eq. (2)).

$$T_e = \ln 2 \frac{Q}{E_{tot}}, \tag{2}$$

T_e (unit in days) has the advantage of being defined at any point of time, regardless of previous intake pattern, and can be experimentally obtainable provided that the fractions of the total excretion through sweat, urine and feces are known. In the case of caesium, the main excretion pathway is via the urine (ICRP, 1988) and it is thus

possible to obtain T_e values for the subjects in the Lund reference group through whole-body measurement in combination with a 24-h urine sampling (Eq. (3)).

$$T_{e,\mathrm{Cs}} = \ln 2 f_u \left(\frac{Q}{E_u} \right). \tag{3}$$

Here f_u denotes the fraction of the total caesium excretion rate, E_{tot}, which is passed via the urine. In this work the value 0.8 for f_u given in ICRP (1988) has been used, although it is not specified in the reference which of the two caesium components this value refers to.

A further illustration of the physical meaning of $T_{e,\mathrm{Cs}}$ is to insert the values of the half-times T_1 and T_2 given above into Eq. (1), and to use the total excretion data for the reference man (ICRP, 1975). In case of an equilibrium between intake and excretion of caesium, $T_{e,\mathrm{Cs}}$ can thus also be looked upon as an average of the component half-times given by the retention function in Eq. (1), where each component is weighted by their fractional retention (10 and 90%, respectively) of the total uptake (see Eq. (4)).

$$\begin{aligned} T_{e,\mathrm{Cs}} &= \ln 2 \left(\frac{Q_{\mathrm{Cs}}}{E_{tot}} \right) = \ln 2 \left(\frac{Q_{\mathrm{Cs}}}{(E_u/f_u)} \right) = \ln 2 f_u \left(\frac{Q_{\mathrm{Cs}}}{E_u} \right) \\ &\approx \ln 2 f_u \left(\frac{f_1}{f_u} \left(\frac{0.1}{(\ln 2/T_1)} + \frac{0.9}{(\ln 2/T_2)} \right) \right) = f_1 (0.1 T_1 + 0.9 T_2). \end{aligned} \tag{4}$$

Based on Eq. (4) we can then theoretically predict a T_e for caesium of about 100 d (as a rough average over both sexes), which is consistent with previous observations (e.g. ICRP, 1989). Unless any members of the reference group suffer from severe metabolic disorders, or are under medication which can influence the excretion rate, the individual variations are expected to be within the range 30–110 d for females and 45–150 d for males (ICRP, 1989).

Although T_e can be defined independently of dietary intake pattern, it is meaningful only if an equilibrium between intake and excretion prevails (Johansson & Ågren, 1994). An approximate equilibrium can be reached in case of a protracted intake of dietary radiocaesium even though a gradual decrease of the dietary caesium intake can be excepted after a single fall-out of caesium, such as the Chernobyl fall-out. Large single intakes of caesium may however disturb this equilibrium, but calculations by Lloyd, Zundel Mays, Wagner and Pendleton (1966) have shown that single major changes in the average intake rate of caesium only have minor effects on the determination of T_e for ^{137}Cs.

2.3. Potassium normalized urinary caesium excretion

When only partial day samples of urine from individuals are available and the collection times are unknown, one must scale the caesium content in the urine samples, $e_{u,\mathrm{Cs}}$ (Bq), to a corresponding 24-h level, in order to use Eq. (3) for whole-body content estimates. It is assumed that the variation of the urinary concentration of caesium during a 24-h interval follow that of potassium due to the similarity of their

chemical properties. The potassium content in the urine sample, $e_{u,\mathrm{K}-40}$ (Bq), could therefore be used as an index of the 24-h urinary excretion rate of caesium, $E_{u,\mathrm{Cs}}$ (Bq), if a standard value of the 24-h urinary excretion of potassium in adults, $E_{u,\mathrm{K}-40,st}$ (Bq), is available that is representative for healthy adults. Since the main fraction of the body potassium inventory is incorporated into the body cells, the 24-h urinary potassium excretion is indirectly a measure of the body cell mass (mainly skeletal muscle tissue) and should therefore vary little with time. ICRP (1975) estimates a urinary excretion level of 2.7 g per day of a total excretion rate of 3.2 g per day for the reference male. The ratio $(E_{u,\mathrm{K}-40,st}/e_{u,\mathrm{K}-40})$ can then be used to scale $e_{u,\mathrm{Cs}}$ to a 24-h level, $E_{u,\mathrm{Cs}-137}$(Bq d^{-1}) (see Eq. (5)). This method is useful only if the total amount of excreted potassium in urine per day, $E_{u,\mathrm{K}-40}$, in the studied subject does not vary too much from the standard value, $E_{u,\mathrm{K}-40,st}$ (Johansson et al., 1995).

$$E_{u,\mathrm{Cs}} = e_{u,\mathrm{Cs}}\left(\frac{E_{u,\mathrm{K}-40,st}}{e_{u,\mathrm{K}-40}}\right). \tag{5}$$

In earlier work (Falk et al., 1991; Rundo & Taylor, 1964; Wallström et al., 1995) the daily caesium excretion rate, $E_{u,\mathrm{Cs}}$, has been normalized also to the whole-body burdens of ^{137}Cs,Q_{Cs}, and ^{40}K,Q_{K}. All four quantities were combined into a factor A, which in this work is denoted as the potassium normalized caesium excretion:

$$A = \left(\frac{Q_{\mathrm{Cs}}}{Q_{\mathrm{K}}}\right)\Big/\left(\frac{E_{u,\mathrm{Cs}-137}}{E_{u,\mathrm{K}-40}}\right). \tag{6}$$

By combining Eqs. (2) and (3) with (6) we may also express A as the ratio between the equivalent biological half-times of caesium and potassium. That is

$$A = \left(\frac{(1/f_{u,\mathrm{Cs}})T_{e,\mathrm{Cs}}}{(1/f_{u,\mathrm{K}})T_{e,\mathrm{K}}}\right). \tag{7}$$

Inserting the values given for T_e ($\sim$ 100 d for ^{137}Cs; $\sim$ ln 2*(140 g/3.2 g) = 30 d for ^{40}K) and f_u (= 0.8 for ^{137}Cs and 0.84 for ^{40}K) for the reference man (based on data from both sexes) in ICRP (1975) and ICRP (1989) we obtain a value of 3.46, which is in reasonable agreement with a large study conducted in Sweden after the Chernobyl accident (Falk et al., 1991). In that study, it is suggested that A is dependent on sex, but varies with age only up to about 20 years of age. For adults, the potassium normalized caesium excretion is assumed to remain relatively constant, since both potassium and caesium have similar biological behavior in humans, and any metabolic change with age, affecting the T_e-values for ^{137}Cs and ^{40}K, respectively, should be canceled out when using the ratio in Eq. (7).

2.4. Creatinine normalized urinary caesium excretion

The urinary excretion of caesium may also be normalized to the excretion of creatinine in urine, since the daily creatinine excretion rate reflects the mass of the skeletal muscle tissue in the body (ICRP, 1975; Forbes, 1987) and therefore should remain relatively stable with time (in analogy with potassium). The creatinine content

in the urinary samples can be scaled to a mean daily creatinine excretion level, and a subsequent scaling of the caesium urinary content on a 24-h level can therefore be done by using the following expression;

$$E_{u,\mathrm{Cs}-137} = e_{u,\mathrm{Cs}-137}\left(\frac{\mathrm{CRE}_{24h}}{\mathrm{cre}_{sample}}\right), \tag{8}$$

where $e_{u,\mathrm{Cs}-137}$ is the ^{137}Cs content (Bq) in a fractional sample of the 24-h urine, cre_{sample} is the creatinine content (mmol) and CRE_{24h} is a standard daily excretion level (mmol d^{-1}) for an adult, taken from the literature or from a specific study.

Although excellent correlation between whole-body cell mass and the 24-h creatinine excretion has been found, the creatinine excretion can vary significantly during the day (Forbes, 1987) and for detailed studies of an individual it is therefore recommended that urinary creatinine excretion data for a single subject be collected over at least 24 h to cancel out any temporal variations. However, when Eq. (8) is applied on a number of samples and averaged out over the studied group, the individual variations in the daily creatinine excretion rates should be less important than if Eq. (8) is applied only to one single sample.

Data on average daily creatinine excretion, CRE_{24h}, already exist (e.g. Jackson (1966), a study that later was quoted in ICRP (1975), Wennmalm, Benthin, Granström, Persson and Winell (1992), Veterans Administration Cooperative Study Group on Antihypertensive Agents VACSGAA (1987) and Geigy Scientific Tables (1981)) but the Lund reference group survey contains data on the 24-h excretion levels of both potassium and creatinine, from a common sample of individuals.

2.5. Experimental studies

The whole-body measurements on the Lund reference group were, with a few exceptions, performed at the Malmö University Hospital. The whole-body counter set-up consisted of two 12.7 cm $\varnothing \times 10.2$ cm NaI(Tl) detectors, mounted in a scanning bed geometry in a room shielded from the background radiation by 15 cm iron and 3 mm lead walls. The two detectors were positioned 30 cm above and 10 cm beneath the patient couch. The scanning length was 150 cm.

The subjects were measured in the whole-body counter for 2000 s, after which their whole-body contents of ^{137}Cs and ^{40}K, $Q_{\mathrm{Cs}-137}$ and $Q_{\mathrm{K}-40}$, were obtained. In connection to each measurement, the daily urine samples from the subjects were collected. A small amount of the 24-h urine was sampled for determination of creatinine concentration, which was carried out by colorimetric analysis at the Department of Clinical Chemistry, Malmö University Hospital. The rest of the urine was then prepared with chloride acid and Cs-carrier and diluted with distillated water to a standard volume of 2 l in plastic bottles (Kautex™). The contents of ^{137}Cs and ^{40}K in the urine samples were then measured by a lead shielded HPGe-detector (Schlumberger Enertec, 5.1% relative efficiency at 1.33 MeV), which was connected to a multi-channel analyzer with a total of 2048 channels.

2.6. Evaluation of the ^{137}Cs and ^{40}K content in urinary samples

A significant fraction of the collected 24-h urine samples had ^{137}Cs concentrations close to the detection limit of the detector set-up at acquisition live-times of typically between 48 and 64 h. We therefore used a manual evaluation method for the acquired gamma spectra in this study, in order to avoid any risk of artifacts from the gamma spectrometric evaluation program that otherwise was used for activity determination. The manual evaluation was carried out by defining three adjacent regions of interest (ROI's) of equal channel width, of which the central one was positioned at the ^{137}Cs full energy peak. The channel widths would roughly correspond to 2 FWHM of the peak. The two other ROI's were separated by three channels from the central one. The number of net counts in the full energy peak, n, was obtained by taking the average number of counts of the two lateral ROI (l and r, respectively) and subtracting this from the total number of counts, c, in the central ROI.

The detection limit, or Minimum Detectable Activity Concentration (MDA), in Bq/l of ^{137}Cs for the set-up was defined as the activity level at which the corresponding signal was equal to three standard deviations of the background signal. For a typical acquisition time of 48 h, the MDA-level was about 0.5 Bq/l.

The daily urinary excretion of ^{137}Cs, $E_{u,\mathrm{Cs-137}}$, and ^{40}K, $E_{u,\mathrm{K-40}}$, in (Bq d^{-1}) was then obtained by

$$E_u = \left(\frac{n}{t}\right)\varepsilon\,\frac{U_{Coll}}{U_{Bottle}}, \tag{9}$$

ε is the sensitivity of the source detector set-up (Bq/s^{-1}) at the photo peak energies of ^{137}Cs and ^{40}K, respectively, t is the live-time (s) of the detector set-up during pulse acquisition, U_{Coll} is the amount of urine collected in 24-h from the subject (l d^{-1}), and U_{Bottle} is the amount of that urine present in the 2 litre Kautex bottle. This ratio was unity in most cases except when the daily urinary excretion of a subject exceeded 2 l.

An estimation of the uncertainty in the value of E_u for ^{137}Cs and ^{40}K yields

$$\Delta E_u \approx E_u\sqrt{\left(\left(\frac{\Delta\dot{n}}{\dot{n}}\right)^2+\left(\frac{\Delta\varepsilon}{\varepsilon}\right)^2+\left(\frac{\Delta U_{Coll}}{U_{Coll}}\right)^2\right)}, \tag{10}$$

where Δn is governed by the Poisson distributed counting statistics and $\Delta\varepsilon$ is the estimated uncertainty of ε due mainly to the uncertainty of the activity in the calibration source (stated as being 5% of ε). Δt and ΔU_{Bottle} are negligible here and have therefore been omitted in Eq. (10).

In general, we have assumed that ΔU_{Coll} is 10% of the estimated value of U_{Coll} to account for daily variations and incompleteness in the emptying of the bladder. However, when estimating U_{Coll} one must rely on the investigated subject collecting their urine correctly, which means that U_{Coll} may sometimes be subjected to errors of unknown magnitude. In a few cases known errors have been corrected for, typically in a case where the subject has excreted a fraction of the daily urine, which for some reason was not included in the collection of the daily sample.

Finally, we obtain the experimental value of the biological half-time of ^{137}Cs by using the expression (3) above:

$$T_{e,\mathrm{Cs}-137} = \ln 2\left(\frac{Q_{\mathrm{Cs}}}{(E_{u,\mathrm{Cs}-137}/0.80)}\right), \tag{11}$$

T_e for potassium is obtained by analogy with Eq. (11), but with the factor $f_{u,\mathrm{K}-40} = 0.84$ (calculated from data given in ICRP, 1975) in place of 0.80.

The relative uncertainty of $T_{e,\mathrm{Cs}-137}$ depends on the relative uncertainty of $E_{u,\mathrm{Cs}-137}$, as well as the relative uncertainty in the determination of the whole-body content of ^{137}Cs, Q_{Cs}. In this work, we have estimated that the maximum relative error of the whole-body measurement has been kept within 20%. This large uncertainty stems from the stripping methods used to separate the different radionuclides in the NaI(Tl)-spectra and the uncertainty in the subject geometry with respect to the calibration phantom, rather than from the pulse statistics.

2.7. Statistical treatment of data

The urinary samples with ^{137}Cs concentration levels close to the detection limit yielded data points which inherently had larger relative uncertainties, $(\Delta T_{e,\mathrm{Cs}-137}/T_{e\mathrm{Cs}-137)_i}$, due to the large contribution from the statistical uncertainty. The index i refers to the data-point coming from the ith measurement (24-h urine + whole-body counting) for a certain individual (typically between 5 to 10 measurements). In order to limit the influence of these data points, each data-point for an individual, during the study period, was weighted by its relative statistical uncertainty obtained from Eq. (10) above. Then a weighted average value, $\langle T_{e,\mathrm{Cs}-137}\rangle_{individual}$, could be obtained for a single individual. Thus,

$$\langle T_{e,\mathrm{Cs}-137}\rangle_{individual} = \frac{\sum_i (1/(\Delta T_{e,\mathrm{Cs}-137}/T_{e,\mathrm{Cs}-137})_i)^2 T_{e,\mathrm{Cs}-137,i}}{\sum_i (1/(\Delta T_{e,\mathrm{Cs}-137}/T_{e,\mathrm{Cs}-137})_i)^2} \tag{12}$$

The error of the weighted average value, $\langle \Delta T_{e,\mathrm{Cs}-137}\rangle_{indiviudal}$, was then obtained by

$$\langle \Delta T_{u,\mathrm{Cs}-137}\rangle_{individual} = \frac{\langle T_{e,\mathrm{Cs}-137}\rangle_{individual}}{\sqrt{\sum_i (1/(\Delta T_{e,\mathrm{Cs}-137}/T_{e,\mathrm{Cs}-137})_i)^2}}. \tag{13}$$

In turn, the same type of weighting was performed when gathering individual data to an average for the whole group, males and females, respectively. The uncertainty levels of $T_{e,\mathrm{Cs}}$ for the male and female members of the Lund reference group presented later will refer to the error of the weighted average value (equivalent to the standard error of the mean, SE, for unweighted average values) obtained from Eq. (13).

The error weighted average value of $T_{e,\mathrm{K}-40}$, $\langle T_{e,\mathrm{K}-40}\rangle$, was obtained in analogy with $\langle T_{e,\mathrm{Cs}-137}\rangle$, where all values were pooled to the individual before the total average was evaluated. The only difference was that virtually all urinary samples had ^{40}K concentration levels well above the MDA of the detector set-up and therefore the error weighting may not have the same effect as it has on $\langle T_{e,\mathrm{Cs}-137}\rangle$.

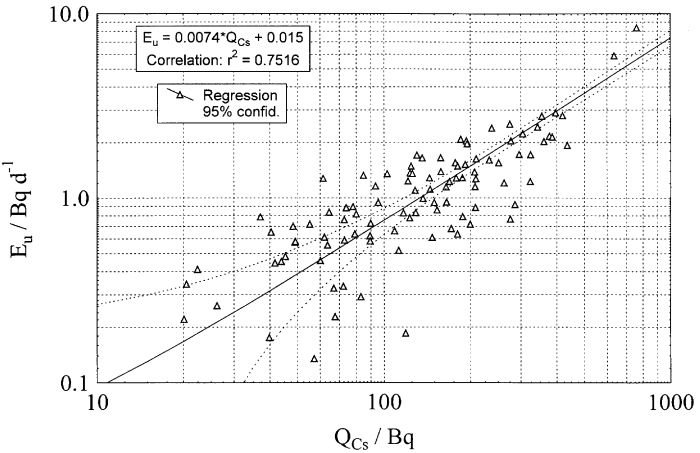

Fig. 1. Daily urinary excretion of ^{137}Cs as a function of the whole-body content:Total outcome from the Lund reference group (non-pooled individual data).

3. Results

3.1. *Whole-body content of* ^{137}Cs *and* ^{40}K

Typical values for the total ^{137}Cs content in the subjects of the Lund reference group ranged from 20 to 400 Bq, which is evident from Fig. 1, where the daily excretion level of ^{137}Cs is plotted vs. the whole-body burden of ^{137}Cs (non-pooled individual data). The whole-body burden levels were more than one order of magnitude lower than was measured in the 1960s by Lidén and Gustafsson (1966).

The average potassium whole-body content ($\pm$ 1 SD) in females was 2 870 $\pm$ 230 and 3 850 $\pm$ 470 Bq in males. The average potassium content per kg body weight ($\pm$ 1 SD) for a subject during the survey was 43 $\pm$ 6 Bq kg^{-1} for the females and 52 $\pm$ 7 Bq kg^{-1} for male members of the Lund reference group. As a comparison, ICRP (1975) predicts a typical potassium concentration of 63 Bq kg^{-1} in a 30 year old male (the reference man) and a 20% lower value for women of the same age. However, for 65 years old males (corresponding to the median age of the male members of the Lund reference group) ICRP (1975) predicts a ^{40}K concentration of about 53 Bq kg^{-1} and for females of 50 years of age ICRP 23 predicts a level of about 45 Bq kg^{-1}.

3.2. *Equivalent biological half-time for* ^{137}Cs *and* ^{40}K

The error-weighted average value of the equivalent biological half-time for ^{137}Cs, $T_{e,\mathrm{Cs}-137}$, of the Lund reference group was found to be 66 $\pm$ 4 d (range, 46 – 79 d) for females and 80 $\pm$ 4 d (range, 62 – 97 d) for the males (reported uncertainty values refer to the error of the weighted average as given by Eq. (13) in the previous section). The distribution of the experimentally obtained $T_{e,\mathrm{Cs}-137}$ values (normalized to the mean

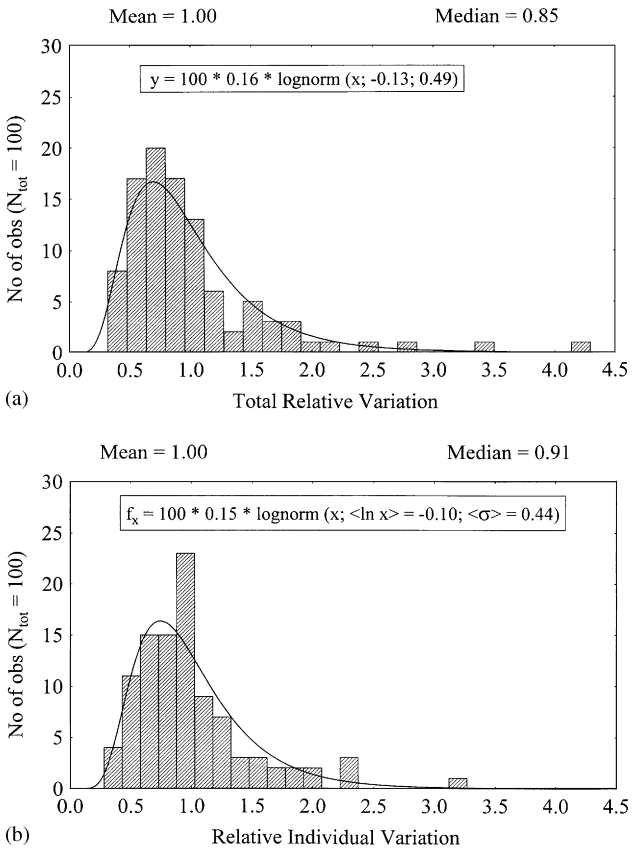

Fig. 2. (a) Variation of all observed equivalent biological half-times, $T_{e,\mathrm{Cs}-137}$, in the Lund reference group. Normalized to total non-weighted average $T_{e,\mathrm{Cs}-137}$. (b) Variation of equivalent biological half-time, $T_{e,\mathrm{Cs}-137}$, for the same individual in the Lund reference group. Superposition of 15 subjects normalized to their individual non-weighted average $T_{e,\mathrm{Cs}-137}$.

value of all observed $T_{e,\mathrm{Cs}-137}$ values in the study) was found to be log-normal (see Fig. 2a). The total variation in $T_{e,\mathrm{Cs}-137}$, expressed as the standard deviation, σ_{tot}, of the distribution, was found to be $= 0.49$, or roughly 50% of the mean value. This variation includes experimental errors, individual variations and the biological variation between the individuals. In Fig. 2b the individual variation in the observed biological half-time is plotted (sum of observed $T_{e,\mathrm{Cs}-137}$ values normalized to the mean value of each individual) and the spread (also including experimental errors) is somewhat less, $\sigma_{ind} = 0.44$.

The error-weighted average value for ^{40}K, $T_{e,\mathrm{K}-40}$, was found to be 21.5 ± 0.8 d (range, 20–26 d) for females and 28 ± 1 d (range, 21 – 39 d) for males.

A positive linear correlation between $T_{e,\mathrm{Cs}-137}$ and the whole-body potassium content, Q_{K}, was found among the male members of the Lund reference group (see

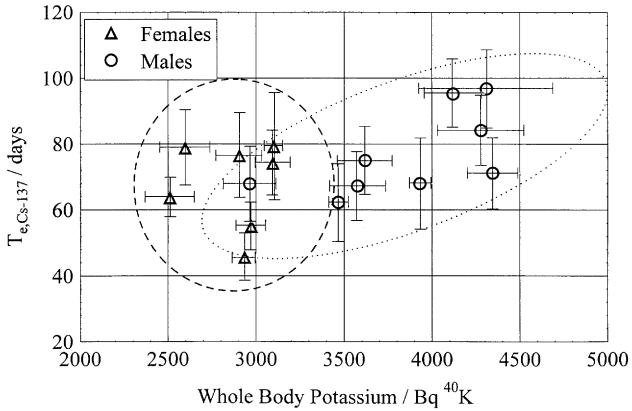

Fig. 3. Equivalent biological half-time, T_e, of ^{137}Cs in the Lund reference group as a function of whole-body content of ^{40}K. Dashed and dotted closed curves indicate 95% confidence area ellipse for females and males, respectively. Errors bars indicate 1 standard error of weighted individual mean.

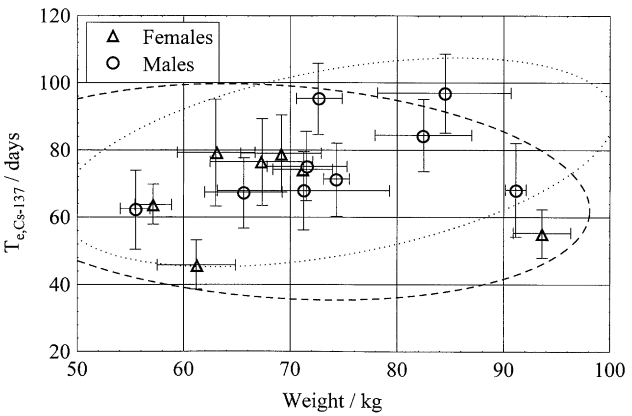

Fig. 4. Equivalent biological half-time, T_e, for ^{137}Cs in the Lund reference group as a function of body-weight. Dashed and dotted closed curves indicate 95% confidence area ellipse for females and males, respectively. Errors bars indicate 1 standard error of weighted individual mean.

Fig. 3; $T_{e,\mathrm{Cs}-137} = 0.017 \times Q_{\mathrm{K}} + 11$, $r^2 = 0.46$) in agreement with earlier work (Leggett, 1986). No such correlation was however found for the women in our study ($r^2 = 0.02$). Poor correlation was also obtained between $T_{e,Cs-137}$ and body-weight ($r^2 = 0.029$ for females and 0.193 for males, see Fig. 4) in the Lund reference group. Omitting one single outlier among the male individuals, the correlation was however significantly improved ($r^2 = 0.567$). No dependency was found between $T_{e,\mathrm{Cs}-137}$ and the subject's median age during the study ($r^2 = 0.070$ for females and 0.054 for males, see Fig. 5).

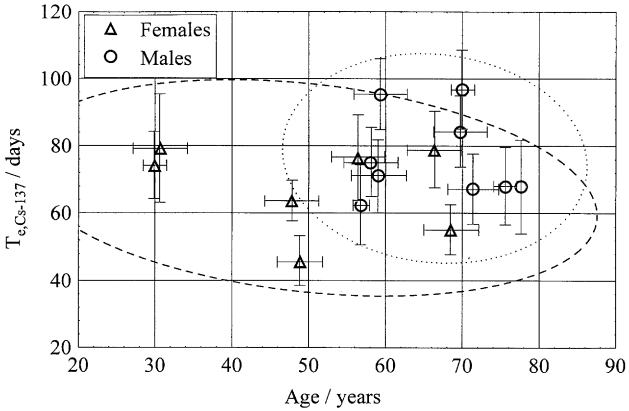

Fig. 5. Equivalent biological half-time, T_e, for ^{137}Cs in the Lund reference group as a function of median age during survey. Dashed and dotted closed curves indicate 95% confidence area ellipse for females and males, respectively. Errors bars indicate 1 standard error of weighted individual mean.

Table 1
Average value of $T_{e,Cs-137}$ for the Lund reference group as obtained by different averaging methods

Method	$T_{e,Cs-137}$ days	
	Females	Males
$(1/\Delta^2)$ weighted average	66 ± 3	81 ± 4
AVG 0 MDA	77 ± 26	85 ± 16
AVG 1 MDA	67 ± 17	77 ± 17
AVG 1.5 MDA	71 ± 14	84 ± 32
AVG 2 MDA	54 ± 9	71 ± 9
ICRP 56 – values	65 (range, 30 – 141 d)	96 (range, 47 – 152 d)

For uncertainty values refer to Eq. (13) for the error-weighted average values and to one standard deviation (1σ) of the arithmetic average for the MDA discriminated values.

3.3. Use of error-weighted average

The average value of $T_{e,Cs-137}$ obtained by weighting each data point with its individual relative uncertainty, was compared with the arithmetic mean value of the data points (pooled to the individual) obtained after omitting values below the levels 1, 1.5 and 2 times the detection limit, MDA. This procedure was done as a quality check of the data material due to the large fraction of urinary excretion data points lying close to the detection limit of $E_{u,Cs}$ (~ 0.5 Bq l^{-1}, see Fig. 1). In Table 1 we see that almost identical values were obtained when comparing the weighted average value and the arithmetic mean value of all points above 1 MDA.

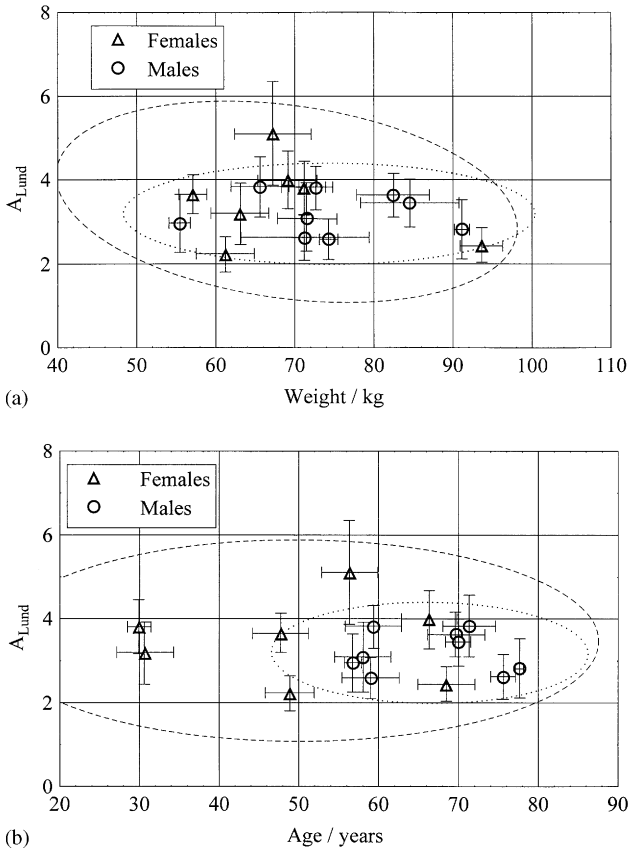

Fig. 6. (a)Potassium normalized caesium excretion, A_{Lund}, in the Lund reference group as a function of average body-weight. Dashed and dotted closed curves indicate 95% confidence area ellipse for females and males, respectively, Errors bars indicate 1 standard error of weighted individual mean. (b) Potassium-normalized caesium excretion, A_{Lund}, in the Lund reference group as a function of median age during survey. Dashed and dotted closed curves indicate 95% confidence area ellipse for females and males, respectively. Errors bars indicate 1 standard error of weighted individual mean.

3.4. Potassium-normalized caesium excretion, A, and 24-h creatinine excretion in urine

The error-weighted average individual value for the factor A defined in Eqs. (6) and (7) for the Lund reference group was 3.4 (range, 2.2 – 5.1) $\pm$ 0.3 (1 WSE) for females and 3.4 (range, 2.6 – 4.2) $\pm$ 0.2 (1 WSE) for males. A previous study by Falk et al. in 1991 gave values of 2.8 (mean) $\pm$ 1.0 (1 SD) for females and 3.4 (mean) $\pm$ 1.4 (1 SD) for males. Any correlation between A and age (A vs. median age during study; $r^2 < 0.001$ for females and < 0.001 for males) or on body-weight (A vs. body-weight; $r^2 = 0.068$ for females and < 0.001 for males) remained undetected in the Lund reference group data (see Fig. 6a and Fig. 6b).

The average 24-h creatinine excretion in the Lund reference group was 9.8 (range, 8.5 – 10.9) $\pm$ 0.9 (1 SD) mmol d^{-1} for females and 12.3 (range, 9.8 – 15.5) $\pm$ 1.2 (1 SD) mmol d^{-1} for males. In the literature (ICRP, 1975) the daily excretion of creatinine is estimated to be 8.8 mmol d^{-1} for females and 15.0 mmol d^{-1} for males.

4. Discussion and conclusions

4.1. Equivalent biological half-time, $T_{e,Cs-137}$ and $T_{e,K-40}$

There is a well-established relation between the potassium content and the muscle tissue content of the body and it is therefore expected that males with a lower muscle content also will exhibit a lower uptake of caesium in the long-lived pool (which is consistent with earlier observations mentioned previously). All humans are expected to be subject to some degree of decrease in body muscle weight with age. The males in the Lund reference group display, on average, both a lower potassium whole-body content (12% less than the reference man) as well as a lower daily creatinine excretion (more than 15% less than the ICRP 23 value), altogether indicating that their muscle content in general is lower than in the reference man (ICRP, 1975). It is therefore not surprising that the error weighted average of $T_{e,Cs-137}$ ($= 81 \pm 4$ (1 WSE) d) is lower than the 96 d stated in ICRP (1989) since the caesium uptake in the body is dependent on its muscle mass (ICRP, 1989; Leggett, 1986). Another Scandinavian study (Häsänen & Rahola, 1971), however, indicates that adult healthy males (average age $= 29$ years) can have significantly lower $T_{e,Cs-137}$ values than the ICRP 56 value without having lower whole-body contents of potassium than the reference man.

The females in the Lund reference group have, on average, a $T_{e,Cs-137}$ value ($= 66 \pm 4$ (1 WSE) d) that more agrees with the ICRP (1989) — value than the corresponding value for the males. However, the whole-body contents of potassium in the Lund females are also in agreement with what is generally expected to be found in adult women (reference woman: about 1.65 g $kg^{-1} \times 31.2$ Bq $g^{-1} \times 58$ kg $= 3000$ Bq ^{40}K (ICRP, 1975)). The main reason for this has been attributed to the lower median age of the Lund females (50 years), having whole-body potassium levels and skeletal muscle contents that are more representative of adult women in previous studies (ICRP, 1975) than are the Lund males of men in previous studies (ICRP, 1975).

The lower average whole-body content of potassium, Q_{K-40}, found in males in the Lund reference group did not correspond to a lower error-weighted average biological half-time of potassium, $T_{e,K-40}$ ($= 28 \pm 1$ (1 WSE) d), which was in accordance with the values given in ICRP 23 (ln $\sim 2 \times (140/3.3) = 29$ days).

Noteworthy is an observation that illustrates how potassium metabolism can be altered during severe health conditions. An extremely low potassium excretion rate, (< 0.8 g d^{-1} compared to the reference value 2.7 g d^{-1}), was recorded on two occasions for a male person suffering from bone metastasizing bladder carcinoma during the later time of the study period effect. These data points were not excluded though, since they did not have a significant effect on $\langle T_{e,K-40} \rangle$ for the whole group of males.

The potassium-normalized caesium excretion levels obtained from female members of the reference group was 35% higher than found in earlier work for a Swedish population (Falk et al., 1991). It is evident from Eq. (7) that the low equivalent biological half-time of potassium for the females in the Lund reference group yields a higher value of A.

No age dependence of T_e could be traced in our material, neither for $T_{e,\mathrm{Cs-137}}$, nor for $T_{e,\mathrm{K-40}}$. The number of members in the study group was too small to suggest that any such dependency could be resolved with statistical significance.

4.2. Application of A_{Lund} as a biological constant

The use of the potassium normalized caesium excretion, A, is an attempt to obtain a quantity that could be generally applicable on any population group. A survey of earlier studies (see Table 2) showed that, apart from a Swedish study (Falk et al., 1991), no clear evidence on a sex dependence of A for adults has been established. Neither did earlier studies indicate an age dependence of A in the caesium metabolism in adults, which nevertheless was consistent with earlier predictions of the ratio A in Eq. (7). The non-existence of an age dependency of A, found in this study (see Fig. 6b), was thus in agreement with earlier assumptions.

The weighted average values of A obtained in this study were in reasonable agreement with the large study conducted by Falk et al. in the late 1980s. However, as is evident from the summary in Table 2, a Japanese study in the mid-sixties (Uchyiama et al., 1969) yielded an A — value significantly lower than obtained from both American and European studies. The low value of A for the studied Japanese subjects was mainly explained by the average potassium excretion rate being at least a factor 1.5 – 2 lower than that observed in the Western studies. Since the average whole-body potassium content of these Japanese subjects was about the same as those in Western study groups (125 vs. 140 g for the reference man), and the fact that there is no strong evidence of certain ethnic groups having different ratios of $Q_{\mathrm{Cs}}/E_{u,\mathrm{Cs-137}}$ (see Table 3),

Table 2
Observed values for the potassium-normalized caesium excretion, A, from different studies

Study	Nationality (ethnicity)	Number of subjects (Females and Males)	Females Ages		Males	
			0–20	> 20	0–20	> 20
This work	Swedish	7 F and 9 M		3.4		3.4
Falk et al. (1991)	Swedish	70 F and 239 M		2.8		3.4
Johansson et al. (1995)	Swedish incl. Saami	30 F + M	2.4		2.1	
ICRP 23, 30 and 56	N/A			2.3		3.5
Clemente et al. (1971)	Italian	19 F & 23 M		2.2		2.1
Heinrichs et al. (1989)	German	5F & 5 M		4.9		4.1
Lloyd et al. (1970)	US	5 M				3.0
Uchyiama et al. (1969)	Japanese	5 – 18 M				1.3

Table 3
Observed values for the ratio $Q_{cs}/E_{u,Cs-137}$ from different studies

Study	Nationality (ethnicity)	Number of subjects (Females and Males)	Adults (> 20 years)	
			Females	Males
This work	Swedish	7 F & 9 M	119	145
Falk et al. (1991)[a]	Swedish	70 F & 239 M	112	175
Lessard et al. (1980)	Bikini Islanders	12 F & 14 M	149	151
Schwartz & Dunning (1982)	Mixed data	29 F + 116 M	110	163
Clemente et al. (1971)	Italian	19 F & 23 M	122	148
Heinrichs et al., 1989	German	5 F & 5 M	184	176
Lloyd et al. (1970)	US	5 M		166
Uchyiama et al. (1969)	Japanese	5 – 18 M		124
Average (± 1 S.D)	N/A	142 F + 411 M	133 ± 29	156 ± 18
Sample sized weighted average	N/A	142 F + 411 M	119	167

[a] Values derived from the median values of the effective biological half-time obtained in the study through urinary to feces branching ratio taken from ICRP 56.

we thus have an indication that the potassium ratio, $Q_K/E_{u,K-40}$, may be significantly dependent on the daily intake of potassium. Therefore, the potassium normalized caesium excretion, A, may also be dependent on the diet, which was also suggested already in the beginning of the 1960s (Rundo & Taylor, 1964) when this quantity was introduced.

A typical Western diet contains about 3 g of potassium per day for an adult (based on excretion data given by ICRP, 1975), which is about 1.5 times more than what was observed in another Japanese study from the sixties by Fujita, Iwamoto, Kondo and Yabe (1969). Evidently, a smaller potassium intake is required for a Japanese to maintain about the same whole-body content as in a typical Western adult. This would also mean that the whole-body burden to 24-h excretion ratio is different, due to more or less excessive dietary intakes of potassium with respect to the body potassium pool. According to Eq. (6) the value of A would then be dependent on the daily intake of potassium and would therefore be unsuitable as a biokinetic constant, if the average daily potassium intake varies greatly between different population groups.

The question for the environmental radiologist is whether the use of the potassium-normalized caesium excretion is justified in many cases, or even inadvisable, if the quantity A, obtained from a reference population is to be applied on population groups with widely different dietary habits.

4.3. Future work

The potassium-normalized caesium excretion and the 24-h creatinine excretion in urine are the measurable parameters which can be used to normalize the caesium content in fractional urinary samples to a 24-h excretion level. In a future study, these two biological parameters will be applied on partial-day urinary samples collected from rural inhabitants in the Bryansk area, Russia, in order to obtain estimates on

their whole-body content of ^{137}Cs. The standard values of A and 24-h creatinine will be taken from the results of the Lund reference group, as well as from a variety of sources in the literature, to enable a more direct comparison to be made between these two normalization methods.

4.4. Use of error-weighted average

The error-weighted averages of $T_{e,\mathrm{Cs}-137}$ for the group of individuals (males and females, respectively) seemed to account well for the large uncertainties inherent in the data points close to the detection limit. Discarding all data points originating from urinary content measurements below the detection limit of the detector set-up and then using a conventional average of each subject to obtain $T_{e,\mathrm{Cs}-137}$, seemed to agree well with the method of error-weighted average. Discriminating at higher levels (1.5 and 2 times MDA) resulted in a decrease in the number of data-points which probably counteracted any gains in statistical accuracy of the result. This especially applied to our study, since the Lund reference group was too small to accommodate any larger reductions in subject material.

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