Human metabolism of caesium

Christopher L. Rääf (Editor)
Lund University, Sweden

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Abstract

A study of the human biokinetics of caesium in two forms, i.) incorporated in foodstuff (\(^{137}\)Cs in perch and mushrooms) and ii.) in ionic state (\(^{134}\)Cs in aqueous solution) has been carried out at the department of Radiation Physics in Malmö, starting in 2001. The results of the pilot study were published in 2004, and a continuation of that study has now been carried out by means of NKS funding (NKS-B Cskinetik). The aim is to, i.) investigate whether Scandinavian populations exhibit shorter biological half-time of radiocaesium than other populations; ii.) extend the biokinetic study to additional human subjects from the other Nordic countries.

Results from the continued study further indicate a near complete absorption of radiocaesium in the gastro-intestinal tract, be it in ion state or contained in food matrix. So far, the literature survey of Nordic studies on biokinetics of Cs suggests that the biological half time is somewhat shorter among Scandinavian males (84 days vs. ICRP-value of 110 days), although females do not exhibit any significant difference (64 days vs ICRP value of 65 days).

Key words

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HUMAN METABOLISM OF CAESIUM

Final Report on NKS-B project Cskinetik
September 2005

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Abstract — A study of the human biokinetics of caesium in two forms, i.) incorporated in foodstuff (137Cs in perch and mushrooms) and ii.) in ionic state (134Cs in aqueous solution) has been carried out at the department of Radiation Physics in Malmö, starting in 2001. The results of the pilot study were published in 2004, and a continuation of that study has now been carried out by means of NKS funding (NKS-B Cs kinetik). The aim is to, i.) investigate whether Scandinavian populations exhibit shorter biological half-time of radiocaesium than other populations; ii.) extend the biokinetic study to additional human subjects from the other Nordic countries. Results from the continued study further indicate a near complete absorption of radiocaesium in the gastro-intestinal tract, be it in ion state or contained in food matrix. So far, the literature survey of Nordic studies on biokinetics of Cs suggests that the biological half time is somewhat shorter among Scandinavian males (84 days vs. ICRP-value of 110 days), although females do not exhibit any significant difference (64 days vs ICRP value of 65 days).
Introduction

Huge amounts of data on Cs-kinetics in human adults exist today (e.g.; Leggett, 2003; Schwartz and Dunning, 1982), but there are still uncertainties of the variation in uptake fractions and biological half times with regard to the chemical state of Cs when ingested. It has also been suggested that there are regional variances in the biokinetics (ICRP, 1989). In order to improve the accuracy of predictions of long-term transfer of deposited Cs to humans in the Nordic ecosystems, it is important to study more thoroughly the variance of the biokinetics of Cs. A shorter biological half time in humans than the 110 days presented by the ICRP, may in part explain the discrepancies observed between the measured $^{137}$Cs body burdens and estimates based on detailed food surveys in some Nordic human populations (NKS, 1999). The effect of feed digestibility, and particularly crude fibre, on absorption of radiocaesium in the gastrointestinal tract has been highlighted in a recent review of physiological parameters that affect the uptake of radiocaesium in ruminants (Skuterud et al., 2005). Given that humans have a lower digestibility of fibre compared to most ruminants, this parameter deserves attention also in humans. It was not possible to accomplish this within the framework of the Cskinetik project.

In April 2004 the project CsKinetic was launched within the NKS-B programme with the aim of investigating the human biokinetics of radiocaesium, specifically targeting the results from studies carried out in the Nordic countries. The project is a continuation of a study conducted in Sweden where human adults have ingested known amounts of $^{134}$Cs and $^{137}$Cs and subsequently have been subject to excretion analysis as well as whole body countings (Rääf et al., 2004).

Materials and methods

Collection of samples

The foodstuffs applied in the pilot study in Malmö were perch and mushrooms originating from an area south of Gävle (Figure 1), and contained up to 10 kBq kg$^{-1}$ dry matter. The supply of these samples ran out during experiments in 2003, and there was thus need for additional environmental samples that could be used for the present NKS-study. Concentrations of at least 5 kBq kg$^{-1}$ $^{137}$Cs are required in order to enable detectable levels (within reasonable acquisition times) by gamma spectrometry in excreta the first few days upon ingestion.

Figure 1

Map over the location of the lake Hedsjön.
A brief survey of possible food samples to be used for kinetic the study was done in late spring 2004. Individual reindeer in some areas in Norway continuously contain 6-7 kBq kg\(^{-1}\), but the number of animals is not large and it is not straightforward to obtain meat of such animals. Instead it was therefore decided to return to the area south of the city of Gävle in Sweden, where the food samples of the previous study were collected. On August 11, 2004, eight number of perch, each about 1 kg, were caught in the lake Hedsjön (Figure 1.). Individual concentrations of \(^{137}\)Cs were determined at the SSI in Stockholm (Figure 2) before being sent to Malmö for preparation to edible food portions. With an average concentration of about 7 kBq kg\(^{-1}\) it may require up to 5 hectograms of fish meat in order to have more than 2 kBq \(^{137}\)Cs in edible form (taking into account of cooking losses of up to 30%).

**Figure 2**

![Graph showing \(^{137}\)Cs contents in perch (Sample 1-8) and pike (Sample 9) from lake Hedsjön, Gävle kn, Sweden](image)

Samples harvested in August 10-11, 2004

**Subjects**

Three adults (one females, two males) volunteered to participate in the kinetic study in Cskinetik. The subjects reside in Denmark, and have readily access to gamma spectrometry facilities at the Risö National laboratory. More subjects are desired, but tedious sampling and access to whole-body countings at narrow time intervals after ingestion of food portions, make the recruitment of subjects a challenge. The subjects are preferably selected from personnel and their relatives, working at laboratories equipped with whole-body counters.

**Experiment**

Food preparation of the fish caught in Hedsjön into food portions was carried out in Malmö in September 2004. The fishmeal consisted of boiled perch filets that had been stewed in milk, aiming at minimal loss of juices from the perch. The fishmeal was portioned into 110ml plastic flasks that were used as reference geometry in both Malmö and Risö. In connection with the food processing three aliquots of \(^{134}\)CsCl were prepared for the subjects in the study. The food portions and aliquots were brought to Risö in the beginning of October 2004, where the three volunteers ingested the portions on October 6 and 7. Daily urine samples (from all three subjects) and fractional faecal samples (from two of the subjects) were collected during five days after the ingestion. The urine was analysed by gamma spectrometry at Risö, whereas the faecal samples were sent for further preparation at the Dept of Radiation physics in Malmö before being analysed by gamma spectrometry at. The intention was to use the same sample geometry; a
110 ml plastic flask, when determining the $^{134,137}$Cs content in the food portions, the $^{134}$CsCl-aliquots, the faecal and the urine samples, in order to enhance the accuracy of the determined values.

**Biokinetic model**

It is verified from many studies that the time dependence of the Cs-retention in human adults, $R(t)$ follows a dual exponential function (Eq. 1). ICRP (ICRP, 1989) has adopted the retention function of Eq. 1, using generic values to be applied conservatively to humans. It is assumed that full absorption of Cs occurs when entering the gastro-intestinal tract, that is $f_0=1$ in Eq.1. The caesium in plasma is then assumed to be distributed to viscera and muscle tissues, and then released from the body with a short-term biological half-time of $T_1=2$ days ($a_1=10\%$ of initial retention), and a major component ($a_2=90\%$ of initial uptake), with a biological half-time of $T_2=110$ days. A double exponential curve as in Eq. 1 was thus fitted to the whole-body burden data from one of the subjects (BL), in order to obtain the biological half-times, $T_1$ and $T_2$.

$$R(t) = f_0(a_1 \cdot e^{-\frac{2\ln 2}{T_1}} + a_2 \cdot e^{-\frac{2\ln 2}{T_2}})$$

Eq. 1

For continuous intakes of Cs, spanning over several months or years, a semi-equilibrium is achieved in the intake and excretion. The ratio between the whole-body content and equilibrium excretion rate is defined by ICRP (ICRP, 1989) as the equivalent biological half time, $T_e$, and is approximately related to the retention parameters as in Eq. 2. Many studies conducted on human Cs kinetics in the Nordic countries have been carried out on populations that were subjected to continuous intakes of $^{137}$Cs through contaminated foodstuff. In order to obtain comparable data between previous documented observations and the findings in the present study the approximate relation in Eq. 2 has thus been used.

$$T_e = \ln(2) \frac{\text{observed}_\text{body-content}}{\text{observed}_\text{24h-excretion}} \approx a_1 \cdot T_1 + a_2 \cdot T_2$$

Eq. 2

The gastrointestinal uptake fraction is related by the amount of excretion through faeces per unit-ingested amount of radiocaesium, during the first few days after ingestion as in Eq. 3. It has been experimentally determined by the accumulated faecal excretion of $^{134}$Cs and $^{137}$Cs during 5 days after intake. The fraction of radiocaesium excreted through urine during the same period reflects the size of the rapid uptake component, $a_1$ in Eq. 1.

$$f_0 \leq 1 - \frac{\text{excreted}_\text{faeces}}{\text{total}_\text{ingestion}}$$

Eq. 3

Present-day levels of $^{137}$Cs in humans in urban Nordic populations are very low, typically between 50 and 100 Bq (Rääf et al., 2005), and are not assumed to have interfered with the studies conducted on the subjects at Risö.

**Results and discussion**

**Excretion of radiocaesium**

The urinary excretion in fractions of ingested amount of the three subjects varied from 0.09 to 0.17, due to individual variation (Figure 3). No systematic difference between $^{134}$CsCl and $^{137}$Cs incorporated in perch has been observed. The dispersion in the fractional excretion of faeces was lower than for urine (Figure 3). The results are in accordance with the findings of the Swedish pilot study (Rääf et al., 2004), which indicate no difference in the gastro-intestinal uptake of Cs.
incorporated in certain food matrices and Cs in ionic state (aqueous solution). In Figure 4 are plotted both the new data from Denmark and the results of the previous study in Sweden. The plot illustrates that the difference between the accumulated fractional excretion of $^{137}$Cs and $^{134}$Cs as a function of time elapsed after ingestion, averaged over all participants, is close to zero.

**Figure 3**
Fractional excretion of urine and faeces in three adult subjects upon ingestion of $^{137}$Cs in perch and $^{134}$CsCl in aqueous solution.

**Figure 4**
Difference in accumulated fractional excretion of $^{134}$Cs and $^{137}$Cs in five Nordic subjects. The absolute difference refers to the fraction excreted $^{134}$Cs minus fraction $^{137}$Cs expressed in %-units.

In subjects BL and JSH, the accumulated faecal excretion over 4 days was less than 2% of the ingested amount for both forms of Cs. This suggests an uptake fraction from the gastro-intestinal tract, $f_0$, of about 98%, irrespectively of the two forms of Cs ingested. The result is in accordance with the previous study (Rääf et al., 2004(6)), as well as with other international studies, which all point to a near 100% uptake to plasma immediately after intake. Observed discrepancies between estimated average $^{137}$Cs concentration in human population using the ICRP-model (NKS, 1999(4)) do therefore not appear to originate from erroneous assumption of complete plasma uptake.
Whole-body retention of radiocaesium

The observed biological half times, using the retention model given in Eq. 1, showed no difference between the two forms of radiocaesium in subject BL (Table 1). Curve fits to $^{134}$Cs data for the subject did not exhibit realistic values unless data points shortly and long time after the ingestion were considered separately. For the male subjects (CL, JS), biokinetic values are currently only available for $^{137}$Cs, which is presented in Table 2.

Table 1
Short-term and long-term biological half-time of $^{134}$Cs and $^{137}$Cs observed for subject BL

<table>
<thead>
<tr>
<th>Subject</th>
<th>$T_1$</th>
<th>$T_2$</th>
<th>$a_1$</th>
<th>$a_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{134}$Cs</td>
<td>6.5±N/A</td>
<td>79±2.5</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>2.4±0.4</td>
<td>84±4</td>
<td>0.35</td>
<td>0.65</td>
</tr>
</tbody>
</table>

Literature survey of Nordic studies on Cs kinetics

The results of the literature study in Table 2 are still incomplete (lacking Finnish data), but do so far suggest that the half times of body retention in Scandinavian male adults are shorter than what is observed elsewhere. The weighted average biological half time, $T_2$, is found to be 84 days compared with the 96 days found in males. For the few Scandinavian females studied the observed trend is reversed, with a $T_2$ of 82 days compared with 65 days for females in an extensive survey of international studies (Schwartz and Dunning, 1982). For the studies conducted on continuous intakes, there is however no indication of Scandinavian males having shorter biological half time of $^{137}$Cs. The equivalent biological half time, $T_{eq}$, would according to Eq. 2 translate to a $T_2$ of 104 days, which is agreement with the ICRP value.
Table 2
Overview of the biological half-times of $^{137}$Cs in human adults observed in Nordic studies.

<table>
<thead>
<tr>
<th>Study</th>
<th>N</th>
<th>Comments</th>
<th>$a_1$</th>
<th>$T_1$</th>
<th>$a_2$</th>
<th>$T_2$</th>
<th>$T_e$</th>
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<td><strong>Single intake</strong></td>
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<tr>
<td>This study</td>
<td>1 F</td>
<td></td>
<td>0.35</td>
<td>2.4</td>
<td>0.65</td>
<td>84</td>
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<tr>
<td></td>
<td>2 M</td>
<td></td>
<td>0.33</td>
<td>0.47</td>
<td>0.67</td>
<td>83</td>
<td>67</td>
</tr>
<tr>
<td>Lauridsen and Sogaard-Hansen, 1992&lt;sup&gt;(8)&lt;/sup&gt;</td>
<td>1 F</td>
<td></td>
<td>0.12</td>
<td>0.95</td>
<td>0.88</td>
<td>70.5</td>
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<tr>
<td></td>
<td>2 M</td>
<td></td>
<td>0.23</td>
<td>3.6</td>
<td>0.77</td>
<td>83</td>
<td></td>
</tr>
<tr>
<td>Lauridsen and Sogaard-Hansen, 1988&lt;sup&gt;(9)&lt;/sup&gt;</td>
<td>1 F</td>
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<tr>
<td>Häsänen and Rahola, 1971[]</td>
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<td>Miettinen et al., 1963[]</td>
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<tr>
<td>Madshus et al., 1966&lt;sup&gt;(10)&lt;/sup&gt;</td>
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<td>0.12</td>
<td>0.95</td>
<td>0.88</td>
<td>70.5</td>
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<tr>
<td>Naversten and Lidén, 1964&lt;sup&gt;(11)&lt;/sup&gt;</td>
<td>5 M</td>
<td>1 subject ingested Cs repeatedly 2 females after partus</td>
<td>0.088</td>
<td>1.15</td>
<td>0.91</td>
<td>79.8</td>
<td></td>
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<tr>
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<tr>
<td>Johansson, 1997&lt;sup&gt;(13)&lt;/sup&gt;</td>
<td>1 M</td>
<td></td>
<td>0.2</td>
<td>4.4</td>
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<td>98</td>
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<td>1 F</td>
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<tr>
<td>Rääf et al., 2004&lt;sup&gt;(14)&lt;/sup&gt;</td>
<td>2 M</td>
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<td>0.07</td>
<td>0.9</td>
<td>0.93</td>
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<tr>
<td>Males</td>
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<td></td>
<td>0.135</td>
<td>1.23</td>
<td>0.865</td>
<td>84.4</td>
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<td><strong>Continuous intake of foodstuff</strong></td>
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<tr>
<td>Falk et al., 1991&lt;sup&gt;(14)&lt;/sup&gt;</td>
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<td>97</td>
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<tr>
<td>Rääf et al., 2000&lt;sup&gt;(15)&lt;/sup&gt;</td>
<td>7 F</td>
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<td></td>
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<td>9 M</td>
<td></td>
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<td></td>
<td>80</td>
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<tr>
<td>Johansson et al., 1991&lt;sup&gt;(16)&lt;/sup&gt;</td>
<td>27</td>
<td>(M+F)</td>
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<td>Females</td>
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</table>

Conclusions

The following can be concluded from the study;

- No systematic difference in the initial uptake and excretion of radiocaesium being incorporated into perch muscle and in ionic form was observed in this study. The similarity in absorption and excretion observed in the previous Swedish study is supported by the additional results of this study. No control was made of possible differences in the digestibility of the diet otherwise consumed by the participants of the study.

- Faecal excretion during the first few days appears to account for a minor fraction of maximum 2-3% of the initial gastrointestinal content of caesium. This indicates a near 100% uptake from the gastro-intestinal tract to plasma. This suggests that the ICRP-model from 1989 is realistic with respect to the uptake fraction.

- Nordic subjects do not appear to exhibit significantly shorter biological half times of $^{137}$Cs compared with international observations. Considering only controlled biokinetic studies, using single intake experiments, there appears to be a shorter long-term half time of $^{137}$Cs in males than what is observed internationally.
Future activities

- The participants of the project have compiled a literature study containing more than 50 references on biokinetic studies of radiocaesium, with special focus on studies including some form of excretion sampling. There is more work to be done regarding the literature study of Nordic biokinetic studies on human radiocaesium metabolism, and results of previous kinetic studies that are unpublished need to be considered as well.

- There are still $^{137}$Cs contaminated foodstuffs, such as perch and pike, readily available for human biokinetic experiments. The time schedule of further human experiments depends on various factors, availability of sampling and access to whole-body counters, etc. It is, however, expected that other Nordic countries can contribute to additional human volunteers within a 1-year period.

Acknowledgement

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Title Human metabolism of caesium

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