In vivo measurement of pre-operational spallation source workers: baseline body burden levels and detection limits of relevant gamma emitters using high-resolution gamma spectrometry

Christopher L Rääf1,5, Anja Almén1,2, Lena Johansson3 and Kristina Eriksson Stenström4

1 Medical Radiation Physics, Department of Translational Medicine (ITM), Lund University, SE-205 02, Malmö, Sweden
2 Swedish Radiation Safety Authority, SE-171 16, Stockholm, Sweden
3 European Spallation Source ERIC, P.O Box 176, SE-221 00, Lund, Sweden
4 Department of Physics, Division of Nuclear Physics, P.O Box 118, Lund University, SE-221 00, Malmö, Sweden

E-mail: christopher.raaf@med.lu.se

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Abstract

As a measure to prepare for long-term internal dose monitoring of workers at the European Spallation Source (ESS) in Lund, Sweden, operated by the European Research Infrastructure Consortium (ERIC), as well as to enhance emergency preparedness against accidental releases, a series of in vivo measurements were conducted using a high-resolution HPGe detector with a 123% relative efficiency (1.332 MeV). This study describes the whole-body counting set-up, calibration procedure, and subsequent validation measurements using conventional NaI(Tl)-scanning-bed geometry on a selection of workers from the ESS. Detection limits for the relevant gamma emitters $^7$Be, $^{172}$Hf, and $^{182}$Ta were determined to be 65 Bq, 130 Bq, and 22 Bq, respectively, using a 2400 s acquisition time. The baseline measurements suggest that care must be taken to ensure that the fluctuations in the presence of radon daughters $^{214}$Bi and $^{214}$Pb are minimised by, for example, ensuring a minimum air exchange

5 Author to whom any correspondence should be addressed.

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between the measuring room and the ambient air, and by demanding that the measured subjects change clothes and shower before measurement. Furthermore, in a monitoring program for internal doses to spallation source workers, the presence of radionuclides originating from non-work-related sources (such as $^{226}$Ra from private water wells or $^{137}$Cs from intakes of Chernobyl contaminated foodstuffs), or radionuclides from previous work history (such as $^{60}$Co within the nuclear power industry), must be considered.

Keywords: high-resolution whole-body counting, European research infrastructure consortium ERIC, European spallation source, detection limit, occupational exposure

(Some figures may appear in colour only in the online journal)

1. Introduction

In connection with the upcoming operations of the European Spallation Source (ESS) in the city of Lund, Sweden, operated by the European Research Infrastructure Consortium (ERIC), background radiation has been measured in ESS workers in order to establish a baseline for future whole-body measurements. This will assess the internal dose of radiation for some exposed workers at the ESS. The baseline measurement will also be useful as a reference in connection with an accidental release or other radiological emergency at the site, involving both workers and the general public in the vicinity of the facility. A number of studies have been conducted to calculate the potential radiological consequences during normal operation (Norlinder and Huutoniemi 2013) and in case of accidental releases (Lundqvist 2013, Malusek and Pettersson 2016, Swedish Radiation Safety Authority (SSM) 2018). A variety of radionuclides generated from nuclear reaction processes, connected to proton interactions with target and surrounding shielding materials, must be considered in the dose estimates for exposed workers in controlled radiation areas at the ESS. This includes the potential radiological impact of pure beta emitters such as $^{32}$P, $^{25}$S and $^{14}$C as well as gamma emitters such as $^{5}$Be (principal gamma line $E_\gamma = 477.6$ keV; branching ratio $n_\gamma = 0.105$), $^{20}$Al ($E_\gamma = 1273.4$ keV; $n_\gamma = 0.906$), and $^{39}$Cl ($E_\gamma = 1273.4$ keV; $n_\gamma = 0.906$) (Jacobsson 2017). With regards to the general public, the radionuclides $^{13}$N, $^{11}$C, $^{41}$Ar, and $^{125}$I will be relevant in terms of a (minor) contribution to the annual dose (Ene et al 2018). However, these radionuclides will be of little relevance for whole-body monitoring because they are either short-lived positron emitters or are in noble gas form, which renders measurable uptakes to systemic tissues unlikely. However, in accidental releases that disperse activation products from the spallation target to the vicinity of the site, a number of radionuclides will be relevant for the radiological assay of members of the public. According to Malusek and Pettersson (2016) and the Swedish Radiation Safety Authority (SSM) (2018), the most relevant radionuclides in an accidental release from the spallation source, in terms of absorbed dose to the public, will be the alpha emitter $^{148}$Gd ($E_\alpha = 3182.8$ keV) and gamma-emitting radionuclides such as $^{172}$Hf ($E_\gamma = 125.8$ keV; $n_\gamma = 0.113$), $^{187}$W ($E_\gamma = 685.8$ keV; $n_\gamma = 0.273$) and $^{182}$Ta ($E_\gamma = 1221.4$ keV; $n_\gamma = 0.270$). Therefore, an emergency preparedness program for whole-body monitoring of affected subjects must consider the latter gamma emitters.

As a measure to prepare for internal dose monitoring during operations of the accelerator at the ESS, a whole-body counting monitoring program will be designed to fulfill general requirements for the detection of occupationally incurred internal doses. In Sweden, the
suggested requirement is set at a limit of 0.25 mSv per year for workers, taking all occupationally related radionuclides into consideration (Swedish Radiation Safety Authority SSM 2017). Therefore, knowledge of the critical limit (CL) and minimum detectable body content (MDC) (Bq) of radionuclides is indispensable for feasible types of measurements such as in vivo whole-body counting. For various time patterns of intake (chronic or acute), assuming that the main pathway for exposure is through inhalation, generic minimum detectable committed effective doses, MDD (μSv), from each radionuclide can be derived from the CL and MDC values for the specific whole-body counting set-up. Conventional whole-body counting techniques may rely on low-resolution NaI(Tl) detectors or high-resolution gamma spectrometers (such as single or multiple HPGe-crystals with typical relative efficiency of 50% at 1.332 keV; del Risco Norrlid et al 2011). However, these whole-body counting systems are designed for operational and accidental releases from nuclear power production, and are not optimised to detect the radionuclide levels and range potentially present at ESS. Therefore, a study using a high-efficiency, high-resolution gamma spectrometer as a whole-body counting set-up was performed at the Medical Radiation Physics group in Malmö, with the aim of calibrating a measurement geometry that can be used to determine the radionuclide contents in subjects of a range of body size, and to determine the MDC (Bq) and the corresponding MDD for radionuclides related to the operation of the ESS accelerator and neutron beams.

In addition to calibrating the set-up to detect the presence of the aforementioned radionuclides, a series of measurements were conducted to assess baseline (background) levels of internal contamination of gamma emitters among workers at the ESS during the pre-operational stage, as well as the internal contamination of subjects from other practices unrelated to but similar to the ESS. This study will be an important contribution to possible future internal dose assays and personnel monitoring programs.

2. Material and methods

The low background counting (LBC) room at the Skåne University Hospital in Malmö consists of 15 cm thick iron walls and is lined with 3 mm of lead. An air venting system equipped with absolute filters is located above the LBC room to maintain an air pressure that is higher than the normal atmospheric level in order to counteract the influence of variations in the ambient air concentration of 222Rn. In the LBC, an existing whole-body counting system, consisting of two 12.7 cm(Ø) × 10.2 cm NaI(Tl) detectors positioned in a scanning-bed geometry at 30 cm above and 10 cm below the patients’ couch (described in Holstein et al 2015) has been in operation since the 1970s. It is currently used, for example, for clinical tests of 75Se contents and monitoring of hospital personnel working with open radioactive solutions such as 99mTc and 131I. The typical detection limits in terms of MDC are 20 Bq for 60Co and 40 Bq for 137Cs (Hansson and Rääf 2011). However, given the wide variety of gamma emitters associated with operational and accidental releases from a spallation source (Bungau et al 2014, Malusek and Pettersson 2016), it may be advantageous to use a high-resolution gamma spectrometry system instead. Therefore, in the present project, a new measurement set-up was established by defining a sitting geometry inside the compartment of the LBC room using a high-resolution gamma spectrometer instead of the conventional NaI(Tl)-system.

Several nuclear power plants have been using electrically cooled high-resolution gamma spectrometry for routine monitoring of internal contamination of gamma emitters. However, the crystal size of these detectors typically corresponds to a relative efficiency of 50% to 65%
at 1.332 MeV (compared with a 7.6 cm(Ø) × 7.6 cm NaI(Tl) crystal) (del Risco Norrlid et al 2011), which means that their counting efficiency is substantially lower than that of the large NaI(Tl)-crystals used in conventional whole-body counters. As our department has access to a nitrogen-cooled, high-efficiency p-type HPGe detector (ORTEC; Model GEM 100-S; Serial #: 46-P41629A; 123% relative efficiency at 1.332 MeV), it was selected for the proposed new high-resolution whole-body counting set-up. The detector was connected to a multi-channel analyzer (Digidart™, ORTEC) using 2048 channels. The recorded spectra were visualised by the Maestro™ (ORTEC) software.

The channel width of the region-of-interest (ROI) used for evaluation of the background and net count rates of the various gamma lines considered, was primarily defined by the algorithm of the software. In cases where there was evident presence of nearby full energy peaks, the ROI was manually narrowed to minimise interference in the evaluated count rate. For the nuclide $^{152}$Hf the ROI was broadened so that it encompassed all three gamma lines at $E_{\gamma} = 122.9$ keV ($n_{\gamma} = 0.0114$), $E_{\gamma} = 125.8$ keV ($n_{\gamma} = 0.113$) and $E_{\gamma} = 127.9$ ($n_{\gamma} = 0.0146$). For $^{182}$Ta, the corresponding ROI was set at 1217.5 to 1225.5 keV around the 1221.0 keV line, and for $^{187}$W the ROI was set at 682.7 to 689.0 keV. A list of the selected gamma lines and photon energy, also exponentially dependent on body weight. Hence, the efficiency appeared to follow an exponential decay as a function of photon energy. Similarly, it was apparent that the counting efficiency was also exponentially dependent on body weight. Hence, the efficiency, $\varepsilon$, as a function of photon energy, $E_{\gamma}$, and three different phantom sizes (which each represented the attenuation properties corresponding to human body weights of 50, 70 and 90 kg), $w$, was fitted by means
Table 1. Minimum detectable whole-body activity content, $MDC$ (Bq), for a 70 kg person, and the corresponding committed effective dose from an inhalation or oral intake, $MDD_{inh}$ and $MDD_{ing}$, just prior to measurement ($\mu$Sv), using the 123% HPGe detector in a Palmer geometry with a pulse acquisition live-time of 2400 s. Also given are $MDC$ and corresponding $MDD_{inh}$ for time delays of one d and ten d after an acute inhalation intake.

| Gamma emitter (gamma energy) | ROI keV | $T_{1/2}$ (d) | $MDC_{1}$ d delay (Bq) | $MDC_{10}$ d delay (Bq) | $MDD_{inh}$ 1 d delay (\(\mu\)Sv) | $MDD_{inh}$ 10 d delay (\(\mu\)Sv) | $MDD_{ing}$ (\(\mu\)Sv) |
|-----------------------------|--------|---------------|-------------------------|--------------------------|--------------------------------|---------------------------------|-----------------|
| $^{137}$Cs (661.6)          |        | 10 960        | 8                       | 12                       | 14                             | 0.05                           | 0.08            |
|                             |        |               |                         |                          |                                |                                 | 0.09            |
| $^{60}$Co (1332)            |        | 1925          | 10                      | 16                       | 180                            | 0.29                           | 0.48            |
|                             |        |               |                         |                          |                                |                                 | 5.2             |
| $^{40}$K (1460.8)           | >10$^{10}$ | 97           | 150                     | 180                       | 0.29                           | 0.45                           | 0.53            |
|                             |        |               |                         |                          |                                |                                 | 0.60            |
| $^{7}$Be (477.0)            | 53.3   | 65            | 110                     | 425                       | 0.003                          | 0.006                          | 0.022           |
|                             |        |               |                         |                          |                                |                                 | 0.36            |
| $^{226}$Ra (186)            | 180.1  | 357.2         | 605                     | 1010                      | 13400                          | 1940                           | 3200            |
|                             |        |               |                         |                          |                                |                                 | $\geq 10^3$ 847 |
| $^{26}$Al (1273)            | 0.0046 | 5             | 1.1 $\cdot 10^{12}$     | —                        | N/A                            |                                 |                 |
|                             |        |               |                         |                          |                                |                                 |                 |
| $^{39}$Cl (1267)            | 0.039  | 9             | 0.6 $\cdot 10^{9}$      | —                        | 0.4 $\cdot 10^{-3}$            | $\geq 25 \cdot 10^{3}$         | $\geq 25 \cdot 10^{3}$ |
|                             |        |               |                         |                          |                                |                                 | 0.8 $\cdot 10^{-3}$  |
| $^{172}$Hf (125.8)          | 683    | 130           | 260                     | 720                       | 4.8                            | 9.6                            | 27              |
|                             |        |               |                         |                          |                                |                                 | 2010            |
| $^{182}$Ta (1221)           | 114.7  | 22            | 40                      | 520                       | 5.2                            | 9.6                            | 27              |
|                             |        |               |                         |                          |                                |                                 | N/A             |
| $^{187}$W (685.7)           | 0.996  | 25            | 65 000                  | 8.6 $\cdot 10^{5}$       | 0.01                           | 21.5                           | 290             |

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of STATISTICA 6.0\textsuperscript{TM} to the following expression (± 1 standard deviation of the estimate), with an explained variance of $R^2 = 0.967$ (equation (1)):

$$\ln(\varepsilon(E_g, w)) = a + b \cdot E_g + c \cdot w,$$

(1)

with $a = -2.01 \pm 0.067$, $b = -0.383 \pm 0.034$ and $c = -0.0247 \pm 0.0009$. A plot of the experimental values of $\ln(\varepsilon)$ for the three different body weights (50, 70 and 90 kg) and for the various photon energies between 0.12–1.41 MeV is given in figure 2. The efficiency calibration enables the quantification of the whole-body content, $WBC$ (Bq), of a gamma-emitting radionuclide based on the net counts of a full energy peak at $E_g$ (MeV), and with a branching ratio, $n_g$, according to equation (2):

$$WBC(w) = \frac{n_{\text{peak}} \cdot \varepsilon(E_g, w) \cdot k_{\text{Decay}}}{n_g \cdot \varepsilon(E_g, w) \cdot k_{\text{Decay}}}.$$

(2)

The net counts in the full energy peak $E_g$ obtained by the gamma spectrometry software (Maestro\textsuperscript{TM}, ORTEC) are defined as: $n_{\text{peak}}$ (cps); $n_{\text{bkg}}$ is the corresponding value in the measurement with an empty phantom; $t_{\text{acq}}$ is the acquisition time (s) of the whole-body count; $t_{\text{bkg}}$ is the corresponding live-time (s) for the empty phantom of 70 kg ($=179,483$ s); and $k_{\text{Decay}}$ is the factor accounting for the physical decay of the radionuclide since a given reference date. Given the counting efficiency and background count rate with an empty phantom (size 50 to 90 kg), the $MDC$ (Bq) for a number of gamma emitters can be derived (equation (3)):

$$MDC(E_g, w, t_{\text{acq}}, t_{\text{bkg}}) \approx 3.29 \cdot \frac{\Delta B_{\text{bkg}} \cdot \sqrt{t_{\text{acq}}}}{t_{\text{acq}} \cdot \varepsilon(E_g, w) \cdot n_g} \cdot \frac{\sqrt{n_{\text{bkg}}}}{h_{\text{bkg}}},$$

(3)

where $\Delta B_{\text{bkg}}$ (counts) is the reported uncertainty by the software of the net peak at a given location of a potential full energy peak, $E_g$ (MeV), in the spectra acquired from the empty
phantoms during time $t_{bg}$ (s). The expression above is based on Currie (1968) and holds approximately if comparing the count rates of the measured subject against the peak-background count rates, $n_{bkg}$, which were obtained from more than 100,000 s long measurements with an empty phantom. N.B. that for $^{172}$Hf, using an ROI covering, all three nearby gamma lines around 125 keV ($E_{g} = 122.9, 125.8$ and 127.9 keV, respectively), the sum of the branching ratios ($\sum_{g} = 0.0114 + 0.113 + 0.0146 = 0.139$) were employed when evaluating the MDC.

Once the MDC of a gamma emitter is established, the corresponding minimum measurable committed effective dose, $MDD$ ($\mu$Sv), from an inhalation intake just prior to the measurement, can be defined using the inhalation dose coefficients for adult workers taken from ICRP (ICRP 2012). N.B. that the committed effective dose refers to the time integrated effective dose over 50 years to an adult incurred by an intake (ingestion or inhalation) of a given radionuclide (ICRP 2007). Of the listed types of airborne particles (classified as Type F, M, S and V; ICRP 2012) available for a given radionuclide, the one type yielding the highest committed effective dose-coefficient, $\epsilon_{inh,max}$ (Sv/Bq), was selected to obtain a conservative estimate of $MDD_{inh}$ (equation (4)):

Figure 2. Calibration curve of counting efficiency, $\varepsilon$ [in mcps/(s $^{-1}$ kg)], of a 123% HPGe detector in Palmer geometry as a function of gamma energy for a body size of 70 kg (using the IRINA phantom P4).
For comparison, the corresponding minimum committed effective dose, $e_{\text{ing}}$ (Sv), from a recent acute ingestion yielding a whole-body content of $MDD$ has also been calculated with $e_{\text{ing}}$ values taken from ICRP 2012 and combined with nuclide-specific biokinetic information provided in ICRP 1989, ICRP 2016a, 2016b and ICRP 2017. When present for a whole-body count the investigated individual may already have excreted the entire fraction of the ingested radionuclide activity that was not initially incorporated through gastrointestinal tract. To account for this missing fraction of a recent ingestion, the $MDC$ has been scaled up by a factor of $(1/f_1)$, where $f_1$ is the gastrointestinal uptake fraction of the radionuclide (equation (5)):

$$MDD_{\text{ing}} = \frac{MDC \cdot e_{\text{ing}}}{f_1}. \quad (5)$$

The $f_1$ values have been taken from ICRP 119 (ICRP 2012). Once the calibration set-up was completed, individuals working at the ESS site in Lund were invited to participate in testing the newly calibrated set-up. Nine adults (two women and seven men; age range 31 to 52 y (mean age ±1 standard deviation of the mean (SDM) 44.2 ± 6.3 y); weight range 60–135 kg (mean weight 83 ± 8 kg)) provided consent to participate and were subsequently measured from June to July 2018. Each measurement was conducted using the Palmer geometry and the acquisition times (2400 s) described previously. One subject was measured on two separate occasions (July and November 2018). In addition, seven more adult volunteers (three women and five men; age range 31 to 53 y (mean age 43.5 ± 3.2 y); weight range 61–115 kg (mean weight 79 ± 6 kg)) working or studying at Lund University were measured in the same set-up, and the result was used as a reference for the baseline measurement of the ESS workers. In all cases, the subjects were allowed to wear their normal clothing, except for special footwear protection; however, wristwatches and jewelry were omitted upon measurement. The measurement of the two groups was also used to validate the new whole-body counting set-up by also measuring the subjects in vivo (see section 3.2). The validation was then based on the assessment of $WBC$ of the naturally occurring $^{40}$K, which is expected to be present in all subjects.

3. Results and discussion

3.1. Calibration and detection limit of gamma emitters

Based on extensive background measurement with an empty 70 kg phantom, it was possible to obtain the estimated $MDC$ (Bq) and the corresponding estimated $MDD$ ($\mu$Sv) for an inhaled intake immediately prior to measurement. One observation is that small amounts of $^{40}$K, $^{137}$Cs, and $^{222}$Rn daughters $^{214}$Bi and $^{214}$Pb are present in the LBC background. Another observation is that the count rates in the $^{214}$Bi and $^{214}$Pb peaks fluctuate significantly between different measurement occasions (up to a factor 5 over four background measurement occasions in 2018), which renders the gamma lines of these radionuclides unsuitable for the calculation of minimum detectable counts above the background level. Nevertheless, these fluctuations were still trivial compared with the variations found when measuring the subjects in vivo (see section 3.2). If using the ROI
width proposed by the software for evaluation of net counts, as was done here, the presence of $^{214}$Bi can introduce another perturbation because of its many gamma lines in the energy interval 608 to 2200 keV. This especially may be the case if the system is used for the monitoring of personnel working in a medical accelerator or in nuclear reactor environments, where $^{214}$Bi lines lie close to full energy peaks of neutron activation; products such as $^{54}$Mn (855 keV), $^{58}$Co (810 keV) and $^{65}$Zn (1115 keV) may thus affect the MDC of these nuclides. Nevertheless, a number of photo peaks in both the background and the calibration measurements could be identified that were considered relatively free from nearby interferences from other peaks. Even though inhalation pathways of the omnipresent alkali metal radionuclides $^{29}$K and $^{137}$Cs are minor compared with ordinary ingestion exposures, they are nevertheless listed for comparison with the other ESS-related radionuclides.

MDC values range from a few becquerels ($^{29}$Al and $^{137}$Cs) to about 600 Bq for $^{226}$Ra. This translates into relatively low inhalation MDD values, where all radionuclides (except $^{226}$Ra) have MDD values far below the aforementioned recommended annual value of 0.25 mSv y$^{-1}$. Detailed dosimetric data is lacking for inhalation exposures to the radionuclides $^{29}$Al and $^{182}$Ta, and must be calculated using retention functions for both stable elements, as well as the absorbed gamma fractions of source and target organs when incorporated into systemic tissues. To consider the practical limitations associated with monitoring potentially exposed individuals, the corresponding MDD levels were calculated for situations with a time lapse of 1 d and 10 d, respectively, between an acute inhalation intake of the radionuclide and in vivo measurements. It is then evident that the short-lived $^{29}$Al cannot be detected by this type of off-site set-up, because the MDC with only a 4 h delay between exposure and measurement, will result in an MDC of 0.2 TBq. To the best of our knowledge, no biokinetic model for $^{29}$Al is readily available at this time, and future calculations should use a biokinetic model for elemental Al combined with suggested specific absorbed photon fraction provided by the ICRP (ICRP 2016a, 2016b). For $^{39}$Cl, it is estimated that the MDD, for a 24 h delay between an acute exposure and whole-body count, will greatly exceed the Swedish Radiation Safety Authority-recommended limit of 0.25 mSv (Swedish Radiation Safety Authority SSM 2017). The elevated MDDs of these two short-lived gamma emitters must be addressed in more detail in connection with the design of a monitoring program for internal dose assessment of workers at a facility such as the ESS. Moreover, further tests may be needed to verify how the MDD values will be affected in cases where workers have been subject to simultaneous uptakes of other gamma emitters than those listed in table 1, especially in cases where one of the gamma emitters predominates the others.

For the quantitative assessments of almost all radionuclides, except for $^{226}$Ra, a straightforward uncertainty assessment was applied combining the uncertainty in the background counts, the uncertainty in the primary measurement counts (during which the phantom was filled with $^{60}$Co, $^{137}$Cs, and $^{152}$Eu), and the uncertainty of the calibration fit. For evaluation of $^{226}$Ra. The potential counts emanating from $^{235}$U were subtracted from the 186 keV peak according to equation (6) to obtain the full energy counts from $^{226}$Ra in the peak. The corresponding stochastic uncertainty estimate of the WBC of $^{226}$Ra, using the gamma line $E_g = 186$ keV, with $n_{g,Ra226} = 0.0369$, is then given by equation (7).

$$WBC(^{226}\text{Ra}) = \frac{\left(\frac{n_{\text{peak}}}{n_{\text{aq}}} - \frac{n_{\text{bg}}}{n_{\text{aq}}}\right) - WBC_{\text{avg}}(^{235}\text{U}) \cdot n_{g,U235}}{(n_{g,Ra226} \cdot \varepsilon(E_g, w))}.$$
\Delta \text{WBC}^{(226}\text{Ra}) = \left( \varepsilon(E_g = 186 \text{ keV}, w) \cdot n_{Eg,\text{Ra}226} \right)^2 \left( \frac{1}{T_{\text{acq}}^2} \cdot n_{\text{peak}} + \frac{1}{T_{\text{bkg}}^2} \cdot n_{\text{bkg}} \right)
\left( \frac{n_{Eg,\text{U235}}(E_g = 186 \text{ keV})}{2 \cdot n_{Eg,\text{U235}}(E_g = 143 \text{ keV})} \cdot WBC_{Eg=143 \text{ keV}}^{(235}\text{U}) \right)^2
\left( \frac{n_{Eg,\text{U235}}(E_g = 186 \text{ keV})}{2 \cdot n_{Eg,\text{U235}}(E_g = 163 \text{ keV})} \cdot WBC_{Eg=163 \text{ keV}}^{(235}\text{U}) \right)^2 \cdot 0.5
\right).

(7)

In equations (6) and (7), \( n_{\text{peak}} \) is the net count in the aggregated 186 keV peak; \( T_{\text{acq}} \), \( T_{\text{bkg}} \), and \( n_{\text{bkg}} \) are the same as described in equation (2), and \( n_{Eg,\text{U235}} = 0.572 \) for \( E_g = 186 \) keV. \( WBC_{\text{avg}}^{(235}\text{U}) \) is the average whole-body content of \( \text{U}^{235} \) calculated from the net peaks of \( E_g = 143.8 \) and 163.4 keV. The branching ratio for the gamma line \( E_g = 185.7 \) keV of \( \text{U}^{235} \) is 0.572, and 0.0508 is the branching ratio for the \( E_g \)-line 186.2 keV of \( \text{Ra}^{226} \). Finally, \( \varepsilon(E_g, w) \) is the weight-dependent counting efficiency calibration fit obtained according to equation (1).

The result of the validation of the efficiency calibrations is given in figure 3, with a plot of the \( ^{40}\text{K} \) concentrations assessed using the HPGe detector versus the detector for the conventional NaI(Tl) system. Note that for practical reasons, two volunteers in the ESS group and one volunteer in the reference group were unable to participate in the validation measurement; as a result, there were a total of 14 validation measurements. Assessments from the HPGe detector yielded \( ^{40}\text{K} \) WBC values that were, on average (±1 SDM), 95% (±3%) of the corresponding value for the customary whole-body counter. In some cases, the discrepancy between the two systems could probably be explained by the body size of the subjects being outside the range of the initial calibration of the HPGe detector (50–90 kg).

An additional validation of the obtained calibration coefficients for the HPGe Palmer-like geometry was done by setting up a virtual measurement geometry by means of the Visual Monte Carlo (VMC) calculation tool (a downloadable software package from Visual Monte Carlo 2018, described in Hunt et al 2004). The calibration coefficient obtained from the calibration of the HPGe detector (123% relative efficiency at 1.332 MeV) for a uniform distribution of \( ^{133}\text{Cs} \) is 1012 Bq cps\(^{-1} \) for a 70 kg person, compared with a Monte Carlo simulated (one million simulations) value of 1140 Bq cps\(^{-1} \) using a geometry in which the reference man is positioned standing close to the detector crystal with the front surface against the stomach. The relative discrepancy (<15%) is therefore likely to be explained by the Palmer geometry leading to a larger part being closer to the detector than in a standing geometry. Given the significant uncertainties associated with the biokinetic modelling (e.g. Li et al 2015), the level of accuracy of \( \varepsilon(E_g, w) \) is considered sufficient for use in future WBC measurements for occupational internal dose monitoring, and participation in future whole-body counter intercomparisons can be used to make further fine-tuning of \( \varepsilon(E_g, w) \).

3.2. Baseline in vivo whole-body counting of ESS workers and a reference group

The resulting individual WBC estimates of all nine ESS subjects and the combined results of seven reference subjects are provided in table 2. A detectable amount of the neutron activation product \( ^{60}\text{Co} \) and an excess of the naturally occurring \( ^{226}\text{Ra} \) were observed in some
individuals. The presence of $^{60}$Co in subject M1 is most likely attributed to a previous professional background within the nuclear power industry. Subject M5 had been living in a dwelling with a private well for a number of years prior to the measurement, which might explain the small (but non-significant) amount of excess $^{226}$Ra ($WBC=0.78\pm1.0\text{ kBq}$). The majority of subjects exhibited detectable whole-body $^{137}$Cs, which is a remnant from previous large-scale fallout from nuclear weapons tests and from the Chernobyl accident in 1986. The average levels are on a par with what is expected based on previous surveys of the $^{137}$Cs concentration among subjects from southern Sweden (Rääf et al 2001 and Rääf et al 2006). As an illustration of the presence of gamma emitters in the ESS group, a plot of the summed spectra of ten measurements of nine subjects is provided in figure 4.

A major perturbation was the fluctuating net excess $^{214}$Bi and $^{214}$Pb contents observed among study subjects. The count rates of these peaks were up to a factor four to five times higher than the recorded peak-background count rates, showing that the largest part of the radon daughter associated fluctuation in the LBC measurements is mainly attributed to the attachment of the radon daughter to clothing through static electricity, and will depend significantly on the type of clothing and ambient conditions outside the LBC room. If the monitoring of workers is not time sensitive, it is advised that the subjects take a shower and change their clothes before entering the LBC room. Furthermore, electrically cooled high-resolution systems may partly circumvent the fluctuation background of radon daughters, since no venting of exhausted nitrogen fumes is required, thus allowing a more efficient radon suppression by the filtered air supply of the LBC room.

![Figure 3](image-url)  
*Figure 3. Measured $^{40}$K whole-body content by means of a double NaI(Tl) scanning-bed geometry versus a 123% HPGe detector in a Palmer geometry, for 14 of the 16 subjects in the weight range from 60 to 135 kg.*
Table 2. Whole-body content (±1 SD) (Bq) of gamma emitters among nine workers at the ESS site based on 2400 s live acquisition times of the 123% HPGe detector in a Palmer geometry. Measurements were taken in the low background room at Skåne University hospital in Malmö, June–July 2018.

| ID  | $^{137}$Cs | $^{40}$K  | $^{60}$Co | $^{214}$Bi | $^{182}$Ta | $^{172}$Hf | $^{7}$Be  | $^{226}$Ra | $^{29}$Al | $^{36}$Cl |
|-----|-----------|-----------|-----------|------------|------------|------------|----------|-----------|----------|----------|
| F1  | 21 ± 5    | 3220 ± 420| <MDC      | 51 ± 11    |            |            |          |           |          |          |
| F2  | 35 ± 7    | 3670 ± 480| 82 ± 15   |            |            |            |          |           |          |          |
| M1  | <MDC      | 4020 ± 530| 16 ± 4    | 140 ± 24   | <MDC       | <MDC       |          |           |          |          |
| M2  | <MDC      | 4480 ± 580| <MDC      | 60 ± 12    |            |            |          |           |          |          |
| M3  | 27 ± 6    | 3280 ± 430| 82 ± 15   | <MDC       | <MDC       |            |          |           |          |          |
| M4  | <MDC      | 6830 ± 1030| 340 ± 58  |            | <MDC       | <MDC       |          |           |          |          |
| M5  | 20 ± 10   | 4510 ± 630| 72 ± 22   |            |            |            |          |           |          |          |
| M6  | 100 ± 15  | 4450 ± 580| 555 ± 77  | 142 ± 81   | 720 ± 1020 |            |          |           |          |          |
| M7  | 16 ± 4.5  | 4140 ± 540| 170 ± 26  |            |            |            |          |           |          |          |
| Mean of all$^a$ | 23 ± 2 | 4090 ± 70 | −1.2 ± 0.8 | 139 ± 5 | 5.7 ± 2.6 | −10 ± 6.0 | 6.9 ± 15 | −93 ± 37 | −0.8 ± 1.2 | 3.6 ± 2.0 |
| Mean of all$^b$ | 27 ± 2 | 4070 ± 70 | −0.2 ± 0.8 | 67 ± 6 | 4.6 ± 2.6 | −5.7 ± 6.0 | 4.5 ± 15 | −190 ± 280 | −0.9 ± 1.2 | 3.9 ± 2.0 |
| Reference mean (7 subjects)$^b$ | 24 ± 2 | 3980 ± 75 | −1.1 ± 0.9 | 6.3 ± 6 | 0.4 ± 3 | −3.2 ± 7 | −2.0 ± 15 | −190 ± 290 | 1.8 ± 1.7 | −1.3 ± 1.5 |

SD, standard deviation.

$^a$ Based on the sum of all spectra and the mean body mass of all measured subjects, subtracted for the background measurement with an empty calibration phantom weighing 70 kg. Uncertainty values refer to 1 SD of the summed photo peak information.

$^b$ Based on the sum of all spectra and the mean body mass of all measured subjects, subtracted for the background measurement of three repeated background counts with no phantom present.
4. Conclusions

A Palmer geometry using a high-resolution gamma spectrometer (an ORTEC™ p-type HPGe detector with relative efficiency of 123% at 1.332 MeV) was calibrated using a polyethylene phantom representative of body sizes between 50 and 90 kg. It was found that an analytical expression of the counting efficiency, $\varepsilon(E_g)$, could be obtained by a multiple regression of the $\ln(\varepsilon(E_g))$ versus body weight and gamma energy ($>100$ keV), as proven by a validation against the customary whole-body counting system (two 12.7 cm($\varnothing$) × 10.2 cm NaI(Tl)-crystals in scanning-bed geometry), and by Monte Carlo simulation. Calculations of the $MDC$ of relevant radionuclides in connection with the operation of the ESS facility were as low as 5 Bq for $^{29}$Al. For gamma emitters associated with accidental releases, $MDC$ values ranged from about 25 Bq (for $^{182}$Ta and $^{187}$W) to 130 Bq for $^{172}$Hf. For the short-lived radionuclides $^{29}$Al and $^{39}$Cl, associated with potential occupational internal exposures, the practical detection limit will depend completely on the time delay between inhalation and measurement. For a 24 h delay between exposure and whole-body counting, the $MDC$ values for the two radionuclides fall in the order of 1 GBq to 1 TBq, corresponding to the minimum detectable inhalation doses, far exceeding the suggested requirement of 0.25 mSv by the Swedish Radiation Safety Authority (Swedish Radiation Safety Authority SSM 2017).

A series of measurements using this new set-up on workers from the European spallation source site, as well as from workers at Lund University, showed that the most prominent radionuclides to be found, before the commissioning of the ESS accelerator, are $^{39}$K, $^{137}$Cs, and radon daughters $^{214}$Bi and $^{214}$Pb, the latter radionuclides fluctuate highly, probably with respect to the clothing of the subject. However, some individuals exhibited presence of the neutron activation product $^{60}$Co and the naturally occurring $^{226}$Ra. The presence of man-made and naturally occurring radionuclides unrelated to the ESS activities must be considered in a future monitoring program for ESS workers. Based on these initial baseline tests, there appears to be little or no difference between the whole-body contents of ESS workers and subjects from other workplaces in the nearby region.
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ORCID iDs

Christopher L. Rääf https://orcid.org/0000-0002-9495-7166

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