TITLE

Measurements and Monte Carlo simulations of 241Am activities in three skull phantoms: EURADOS-USTUR Collaboration

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**Abstract**

An international intercomparison was organized by Working Group 7 on “Internal Dosimetry” of the European Radiation Dosimetry Group (EURADOS e.V) in collaboration with Working Group 6 on “Computational Dosimetry”, for measurement and Monte Carlo (MC) simulation of 241Am in three skull phantoms. The main objectives of this combined exercise were a) comparison of the results of counting efficiency in fixed positions over each head phantom using different germanium detector systems, b) calculation of the activity of Americium in the skulls, c) comparison of MC simulations with measurements (spectrum and counting efficiency) and d) comparison of phantom performance. This initiative collected knowledge on equipment, detector arrangements, calibration procedures and phantoms used around the world for in-vivo monitoring of 241Am in exposed persons, as well as on the MC skills and tools of participants. Three skull phantoms (BfS, USTUR and CSR phantoms) were transported from Europe (10 laboratories) to North America (U.S. and Canada). The BfS skull was fabricated with real human bone artificially labelled with 241Am. The USTUR skull phantom was made from the United States Transuranium and Uranium Registries whole-body donor (Case 0102) who was contaminated due to an occupational intake of 241Am; one half of the skull corresponds to real-contaminated bone, the other half is real human bone from a non-contaminated person. Finally the CSR phantom was fabricated as a simple hemisphere of equivalent bone and tissue material. The three phantoms differ in weight, size and shape which permitted an efficiency study accordingly. Based on their own skull calibration, the participants calculate the activity in the three EURADOS head phantoms. The MC intercomparison was organized in parallel with the measurement exercise using the voxel representations of the three physical phantoms, counting with 16 participants. Three tasks were identified with increasing difficulty, including a) MC simulation of the simple CSR hemisphere and the HMGU HP Ge detector for calculating the counting efficiency for the 59.54keV photons of 241Am in established measurement geometry, b) MC simulation of particular measurement geometries using the BfS and USTUR voxel phantoms and the HMGU HPGe detector, c) application of MC methodology to calculate the calibration factor of each participant for the detector system and counting geometry (single or multi-detector arrangement) to be used for a person monitoring in each in-vivo facility, using complex skull phantoms. The results of both exercises allowed the conclusion that none of the three available head phantoms is appropriate as a reference phantom for the calibration of Ge detection systems for measuring 241Am in exposed persons. Good agreement was found between MC results and measurements which supports MC calibration of body counters as an alternative method when appropriate physical phantoms are not available.

*Key words*: internal dosimetry, in vivo measurements, skull calibration, Monte Carlo, voxel phantoms, americium

# Introduction

EUROPEAN Radiation Dosimetry Group (EURADOS e.V; [www.eurados.org](http://www.eurados.org)) was founded in 1981. Currently it represents a self-sustainable network of more than 70 Voting Members (VM) and 560 scientists (Associate Members). Voting Members are research laboratories, universities, regulatory bodies, companies, and services dealing with the dosimetry of ionizing radiation (www.eurados.org). Since 2012, EURADOS has been involved in developing and updating the Strategic Research Agenda (SRA) for Europe in the field of dosimetry of ionizing radiation (Ruehm et al. 2016).

EURADOS Working Group 7 on internal dosimetry (WG7) includes 35 full members and 70 corresponding members from 40 institutes of 21 countries from Europe, America and Asia (Lopez et al. 2011a). The work program is focused on eight research tasks including (i) implementation and quality control of biokinetic models, (ii) development of the decorporation therapy models for actinides, (iii) activities related to individual monitoring (e.g. for emergency scenarios), (iv) application of Monte Carlo(MC) methods to in-vivo monitoring of radionuclides, (v) uncertainty studies, (vi) education and training, (vii) internal microdosimetry, and (viii) biodosimetry in case of accidental internal exposures (in collaboration with EURADOS WG10 on “retrospective dosimetry”).

In 2009, the EURADOS WG7 and the United States Transuranium and Uranium Registries (USTUR) started successful collaboration that allowed the members of both organizations to work together and share the data, capabilities and expertise in the field of actinide biokinetics and decorporation therapy modelling, as well as in-vivo measurements of 241Am in the body and MC simulation studies using voxel phantoms.

In 2010, a EURADOS WG7 and USTUR initiated the joint project to carry out an intercomparison on measurements and MC modelling of 241Am deposited in the bone of a USTUR leg phantom (Case 0102) (Kramer et al. 2011; Lopez et al. 2011b). Two further exercises were organized within EURADOS as a collaboration of EURADOS WG 7 and WG 6 (“Computational Dosimetry) for (a) MC modelling of germanium detectors for the measurement of 241Am in a knee phantom (Gómez-Ros et al. 2006), and (b) for MC modelling of in-vivo lung monitoring of enriched uranium (Broggio et al 2012). As a result of these exercises, the EURADOS-KIT training course (Breustedt et al. 2016) on MC methods for the calibration of body counters was organized in Karlsruhe (Germany) for the dissemination of knowledge on the application of MC techniques and voxel phantoms for in-vivo monitoring of x-ray and gamma emitting radionuclides incorporated into the human body.

Americium is a highly radiotoxic bone-surface seeker element. Its most important isotope, 241Am, has a long physical half-life of 432.6 years and a biological half-life in the skeleton of 46.6 years (ICRP 1997). In case of internal exposures 241Am deposition in the body is usually evaluated by in-vitro monitoring of excreta samples using alpha spectrometry. In vivo monitoring is used for detection of 59.5 keV photons of 241Am in the lungs and bones (knee or skull) by γ spectrometry with germanium (Ge) detectors. Appropriate anthropomorphic phantoms simulating distribution of 241Am in these parts of the body are required for detector calibration.

An international intercomparison was organized by the EURADOS Working Group 7 on “Internal Dosimetry” in collaboration with the EURADOS Working Group 6 on “Computational Dosimetry” for in vivo measurement and MC simulation of 241Am in three skull phantoms. Both the measurement and the simulation studies allowed the comparison of the detection systems, counting geometries and phantoms used for calibration and measurement of 241Am in skull by different in-vivo monitoring laboratories worldwide. This intercomparison also allowed examining whether these three phantoms met the criteria for calibration sources representing the americium deposition in human skull. Another objective was to evaluate the agreement of MC simulations with measurements (spectrum and counting efficiency) and to verify the appropriate skills and tools of the participants.

The 241Am measurement exercise was coordinated by Helmholz Zentrum München (HMGU, Germany) and included 12 laboratories, 10 from Europe and two from North America (U.S. and Canada) (Nogueira et al. 2015). The main objectives were: (i) to evaluate detector counting efficiency for various fixed-positions over each phantom (Task 1); (ii) to estimate 241Am activity in each phantom (Task 2). A MC simulation exercise, coordinated by Czech Technical University in Prague (CTU-Prague, Czech Republic) was organized in parallel, and used voxel representations of the three physical phantoms (Vrba et al. 2014, 2015). This exercise included 16 participants from Europe, America and Asia.

# Materials and Methods

**EURADOS intercomparison of measurements and MC simulations of 241Am in skull phantoms**

As a continuation of the EURADOS strategy to promote the application of MC to individual monitoring of internal exposures, a new intercomparison for monitoring and MC modelling of 241Am in three skull phantoms was launched for the measurement exercise in 2011 and for the MC simulations in 2012.

**Phantoms used in EURADOS intercomparison**

The three skull phantoms used in EURADOS intercomparison of measurements and MC simulations of 241Am are depicted in Fig. 1.

The BfS skull phantom was fabricated by the New York Medical Center (USA) (Hickman and Cohen 1988) with a real human bone artificially labelled with 5,239.3 ± 226.8 Bq (2σ) of 41Am (activity given for 2012) on its inner and outer surfaces. The interior of the phantom is filled with paraffin wax the outside of the phantom is covered by wax with a thickness of 6 mm.

The USTUR skull phantom (formerly known as BPAM skull phantom) was fabricated from the first whole-body donation to the Registries – Case 0102. This individual was occupationally exposed to 241Am. A detailed description of the Case 0102 and the post-mortem analysis can be found in Breitenstein et al. (1985).The phantom contains the right side of the skull from Case 0102 and the left side of a skull from a non-exposed individual. The skull was filled and covered with tissue equivalent material (Hickman and Cohen 1988). The activity of the phantom is 287.2 ± 7.4 Bq (2σ) of 241Am (decay correction to 2012).

The CSR phantom (Vrba et al. 2013) was fabricated for this EURADOS Intercomparison by NRPI (Czech Rep.) and SZU (Slovakia) as a simple hemisphere of equivalent bone and tissue material representing the top of a human skull with 241Am activity of 981.4 ± 19.6 Bq (2σ) (activity given for 2012). The main purpose of the CSR phantom was to provide the experimental spectrum and efficiency value for the validation of MC simulations considering a simple counting geometry (Task 1).

The transport of the three skull phantoms (Fig. 2) started at HMGU (Germany) in 2011, and then continued to BfS (Germany), KIT (Germany), NRPI (Czech Republic), SCK-CEN (Belgium), IRSN (France), CIEMAT(Spain), PHE (UK), STUK (Finland), Health Canada, MSAR (USA) and the NCNR (Poland).

The voxel representations of the three physical phantoms were developed from their CT scan images and were used for the MC intercomparison, to simulate the spectra and to calculate detector efficiencies according to the protocol developed by the EURADOS task group.

**Exercise 1 - measurements of 241Am in three skull phantoms.**

Two tasks were identified in the protocol of the monitoring exercise. Task 1 included measurement of the counting efficiency for the detection of 241Am using one germanium detector over each of the three skull phantoms, for different (defined) positions at a distance of 1 cm between detector surface and skull phantom surface. The calibration factor (counting efficiency) Eff (cps/Bq) was calculated as the ratio of count rate (cps) for the 59.5 keV peak in the gamma spectrum and the (known) activity (Bq) of 241Am in the respective skull phantom. Only one simple counting geometry was defined for the measurement using the CSR phantom (at vertical position *P*0). Measurements using the BfS and BPAM (USTUR) phantoms were performed at different positions *Pi*, *i*= 1,…*n* defined in the protocol (Nogueira et al. 2015). Recommended detector positions and inclinations for BfS and USTUR phantoms are summarized in Table 1 and Table 2, respectively. Images of counting positions are depicted in Figures 3 and 4.

Task 2 of the exercise included measurement of the 241Am activity in the three skull phantoms. For this, the participants had to use their own detector system and define their own counting geometry. In particular, the participants had to calibrate their counting system based on their own procedure (i.e., not using one of the three skull mentioned above).Only six participants provided results for Task 2. Three of the six participants used calibration factors obtained from MC simulations and voxel phantoms.

**Exercise 2 - Monte Carlo modelling of the measurement of 241Am in skull phantoms.**

The MC intercomparison (Vrba et al. 2014, 2015) included 16 participants, some of them also provided results for exercise 1 (measurements). Three tasks were identified. Task 1 consisted in the MC simulation of the CSR hemisphere and HMGU Ge detector (Fig.5) for calculating the counting efficiency for 59.54keV photons of 241Am in simple measurement geometry.

Task 2 required MC modelling of the detector in the laboratory of the participant using detection geometries defined for the measurement Intercomparison using the voxel representations of the three skull phantoms. Spectra and calibration factors obtained by the MC simulations were then compared with those calculated from the measurements.

Task 3 consisted in the MC simulation of the whole specific detection process of each participant, to calculate calibration factors by modelling the whole detector system (single or multi-detector arrangement, maximum four detectors setup) in the counting geometry routinely used in the respective facility for the monitoring of individuals contaminated with 241Am.

# Results

Regarding Exercise 1, good agreement was found among the results of the 12 participants of Task 1 (Nogueira et al. 2015), with relative deviations of less than 15% for the BfS phantom, and less than 17% for the USTUR phantom when the counting efficiencies in defined positions were compared. Note that the germanium detectors used here by the participating in-vivo laboratories were fabricated either by ORTEC or Canberra with different sizes, detection areas and entrance windows (most of them used windows made of carbon, one used a window made of beryllium, and one used a window made of aluminium).The influence of detector size (detector diameter varying from 50 cm to 85 cm) on the calibration factor was studied and the position of maximum counting efficiency was defined for each phantom (Fig. 6 and Fig. 7). As expected, highest efficiencies were obtained by the biggest detectors. The detector with an aluminium window (P18) showed a significantly lower calibration factor than the detectors with other windows when detectors of similar diameter were compared. Therefore, measurement results for P18 detector were not used in final regression analysis of data.

The 241Am activities calculated by six participants (Task 2) using their own calibration factors showed discrepancies of up to a factor of 3.4, mainly due to the physical differences between the calibration phantom used by each participant and the phantoms used in this EURADOS intercomparison (activity distribution, skull size and filling, etc.). This is an important outcome that requires further discussion. Only half of participants of Task 1 provided results for Task 2, showing the difficulty to get appropriate real calibration phantoms for actinides in bone. Due to this difficulty, three laboratories provided results of activity (Bq) of 241Am in the skull using MC calibration factors generated by modelling their own Ge detectors together with a dedicated voxel skull phantom. The voxel phantoms used by these three participants were based on a 38-year old woman voxel phantom, on the skulls of the International Commission on Radiological Protection (ICRP) reference man and woman phantoms, and the MAX-06 voxel phantom (Kramer et al 2006). All these phantoms are different in size and shape and, accordingly, the calculated 241Am activity in the three skulls of the present exercise (using MC calibration factor) were different from the corresponding reference values. More specifically, the ratios of calculated/true activities were in the range from 0.77 (for USTUR phantom) to 3.44 (for BfS phantom), showing always an overestimation of the 241Am activity, except for one participant that used the UCIN phantom and obtained a ratio of 0.77 for the USTUR phantom.

Regarding the calculated 241Am activity, best results were obtained for the USTUR phantom in defined positions of the germanium detector over the real contaminated half of the skull. The participant using the Cohen head phantom (López et al. 2004) for calibration (similar size and shape as USTUR phantom, but fully labelled with 241Am) obtained the best match (ratio to true activity= 1.04) (Pérez López et al. 2016). The participant using the voxel phantom of a 38-y old woman (Vrba 2007) also obtained an excellent result (ratio to true activity= 1.08). Larger ratios were obtained for the BfS phantom, due to the small size of this phantom. The participant using Cincinnati skull phantom (Kellar 1995) for calibration obtained a ratio of 1.49, similar to the participant using the phantom of a 38-y old woman.

With reference to Exercise 2 dealing with MC simulations and voxel versions of the 3 physical phantoms, the results of 15 participants of Task 1 (simple counting geometry with one Ge detector over the CSR hemisphere phantom marked with americium) showed very good agreement between simulated and experimental efficiency for 241Am (E= 59.5 keV) (Vrba et al. 2014). Regarding the simulated spectra obtained when comparing with physical measurement, the majority of the participants achieved a good agreement in the region of 35-60 keV, but not so good match was found in the range of 28-32 keV.

The results of Task 2 (MC modelling the detector from participant laboratories) also showed good agreement between measured and simulated spectra, for the counting positions over the CSR and USTUR phantoms. Slightly worse results were obtained for the BfS phantom. Simulated and measured efficiency for E= 59.5 keV photons from 241Am were within ±6% and did not exceed ±12%.

The results of Task 3 (modelling real counting geometries for the measurement of contaminated people in the participant laboratories) were a good opportunity to study monitoring methods of in vivo laboratories around the world. Different arrangements (single/multiple detectors) and several detector-head distances were used. Very good agreement in measured vs. simulated efficiency was obtained (discrepancies within ±7%) (Vrba et al. 2015).

# Conclusions

An EURADOS action was initiated to improve and harmonize in vivo counting methods for the determination of incorporated 241Am. Participants were from Europe and North America. Focus was on the comparison of measurement and MC simulation approaches, based on counting efficiency values obtained by the participants using three different 241Amskull phantoms. More specifically, the main objectives of this combined exercise were comparison of detection efficiencies deduced for fixed positions over each skull phantom using different germanium detector systems, calculation of the 241Am activity in the skull phantoms, comparison of the results of MC simulations with those from measurements (spectrum and counting efficiency), and comparison of the phantoms themselves. This initiative required knowledge on equipment, detector arrangements, calibration procedures and phantoms used around the world for in-vivo monitoring of 241Am in exposed persons, as well as on Monte Carlo simulation and tools of the participants. Based on the results of both exercises we conclude that none of the three available physical skull phantoms is appropriate as reference for the calibration of Ge detection systems used to measure 241Am in exposed individuals. Good agreement was found between results from MC simulations and measurements, which supports the use of MC calibration of body counters as an alternative method when appropriate physical phantoms are not available.

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FOOTNOTES

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LIST OF FIGURE CAPTIONS

Fig. 1.Phantoms used in EURADOS intercomparison (left to right): USTUR Case 0102 skull phantom, CSR skull phantom, and BfS head phantom.

Fig. 2. Transport of thephantomsamong participating laboratories.

Fig.3. CIEMAT counting geometry for the BfS phantom positions 1, 3, 4, 7 and 12.

Fig.4. CIEMAT counting geometry for the USTUR phantom positions 0, 1, 2, -2, 3, -3 and 4.

Fig. 5. Monte Carlo Exercise – Task 1: modelling HMGU detector over CSR hemisphere phantom.

Fig. 6. Results of Task 1 – Comparison of counting efficiencies using different Ge detectors and different positions over BfS phantom

Fig.7. Results of Task 1 – Measurement exercise - Comparison of counting efficiencies using different Ge detectors and different positions over USTUR phantom.. Results for P18 participant were excluded from regression analysis.