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EURADOS INTERCOMPARISON ON MEASUREMENTS AND MONTE CARLO MODELLING FOR THE ASSESSMENT OF AMERICIUM IN A USTUR LEG PHANTOM

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A collaboration of the EURADOS working group on 'Internal Dosimetry' and the United States Transuranium and Uranium Registries (USTUR) has taken place to carry out an intercomparison on measurements and Monte Carlo modelling determining americium deposited in the bone of a USTUR leg phantom. Preliminary results and conclusions of this intercomparison exercise are presented here.

INTRODUCTION

United States Transuranium and Uranium Registries (USTUR) Case 0102 was the first whole-body donation to the USTUR (1979), of a worker affected by a substantial accidental ^{241}Am intake(1). Half of this man's skeleton, encased in tissue-equivalent plastic, provides a unique human 'phantom' for calibrating *in vivo* counting systems. In this case, the ^{241}Am skeletal activity was measured 25 y after the intake. Approximately 82 % of the ^{241}Am remaining in the body was found in the bones and teeth. The ^{241}Am activity concentration throughout the skeleton (in all types of bone) was fairly uniform(2).

A protocol has been proposed by a group of *in vivo* laboratories from Europe [CIEMAT-Spain, IRSN-France and Helmholtz Zentrum München (HMGU)-Germany] and Canada (HML) participating in this EURADOS/USTUR intercomparison. The focus areas for the study included: (1) the efficiency pattern along the leg phantom using Germanium detectors (experimental and computational), (2) the comparison of Monte Carlo (MC) results with experimental values in counting efficiency data and (3) the influence of americium distribution in the bone material (volume or surface). The best counting geometry for measurement of activity is discussed below.

THE USTUR LEG PHANTOM

At the *in vivo* facilities participating in this EURADOS intercomparison, 59.5 keV photons from the ^{241}Am activity in the real-contaminated bone of the USTUR leg phantom were detected and evaluated with germanium detectors and gamma spectrometry methods.

Two leg voxel phantoms were generated for MC calculations. The IRSN voxel phantom was obtained from IRSN-produced computed tomography (CT) scan images of the physical USTUR phantom; homogeneous distribution of the ^{241}Am activity in the bone tissue was assumed. The HML Voxel phantom was generated from USTUR website data (CT DICOM image files available at http://www.ustur.wsu.edu/voxel/DICOM0102.html); in this case the ^{241}Am activity was distributed in the individual bones of the leg sections (hip, femur, patella, tibia and fibula) according to USTUR radiochemical data(3).

Computation at reference points was carried out for the purpose of the comparison of MC simulated results among participants as well for comparison of calculations with experimental data, for the vertical position of the germanium detector over the leg phantom.

Preliminary results obtained by IRSN recommended the revision of the total activity value provided by the USDOE certificate for the USTUR ^{241}Am bone phantom (1026 ± 11 Bq ^{241}Am, on 1 February 1980: available at http://www.pnl.gov/phantom/bone/). A re-evaluation of the ^{241}Am content in the real-contaminated bone was carried out by USTUR using the available radiochemical analysis data for the right leg, and the relative weights of the bones from the right and left skeleton at autopsy. To estimate the ^{241}Am activity in the leg phantom, it was assumed that the concentration in the left-side bones (incorporated in the phantom) was equal to that in the corresponding bones from the right side (analysed). The resulting activity-scaling factors were: 1.099 (pelvis), 1.009 (femur), 1.081 (tibia) and 1.078 (fibula). As no relative weight information was available for the patella and foot, the average of the four scaling values above (1.067) was assumed to apply to these bones. Thus, the estimated total ^{241}Am activity of the (left) leg phantom was[3] 1243 ± 11 Bq (on 1 February 1980).

RESULTS OF THE INTERCOMPARISON

CIEMAT WBC (Madrid, Spain)

(1) CIEMAT Calibration with Spitz knee phantom

A technique was developed at CIEMAT Whole Body Counting (WBC) Laboratory for the assessment of ^{241}Am in the knee[4]. The Spitz calibration phantom and the LE Ge system (for lung monitoring) inside a shielded room were used for knee calibration purposes.

The Spitz anthropometric knee phantom was fabricated by University of Cincinnati with 11 % of the total skeleton mass and 34 kBq of ^{241}Am homogeneously distributed in the bone-equivalent material (removable femur, patella, tibia and fibula).

A counting efficiency study was carried out applying MC calculations for optimisation. A maximum photon fluence value of 59.5 keV was obtained in the low segment of the knee, at an angle of 45° towards its inner side. This study resulted in a counting geometry that consists of the four LE Ge detectors covering the lower part of the knee, wrapping maximum photon emissions (Figure 1).

In vivo measurements were performed inside the shielding room of 13-cm steel walls lined with Pb, Cd and Cu. A lung system of four LE Ge (Canberra) detectors, of 2-ACTII configuration, was used. Each LE Ge detector was 7 cm in diameter, 25 cm thick and surrounded by anti-Compton shield.

(1) CIEMAT experimental results of USTUR leg.

The following two counting geometries for the efficiency pattern study were applied:

(1) Vertical counting geometry[5]: one LE GE detector in a vertical position; d=2 cm to the leg phantom.
(2) Wrapping counting geometry[4]: four LE Ge detectors in a wrapping position; d=2 cm to the leg phantom (Figure 2).

An analysis of the efficiency pattern shows a maximum efficiency value for the centre-knee position. This conclusion has a different approach as the result of the CIEMAT study for the optimisation counting efficiency with Spitz calibration phantom, where a maximum photon fluence value of 59.5 keV was obtained in the low segment of the knee.

(1) A comparison study of experimental efficiency versus MC calculations was performed by IRSN for the CIEMAT vertical position of one LE Ge detector. The USTUR leg voxel phantom was generated by IRSN from CT scan images obtained of the USTUR physical phantom. The

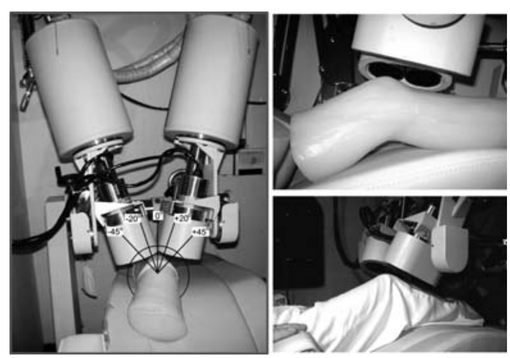

Figure 1. *In vivo* monitoring of ^{241}Am in the knee at CIEMAT.

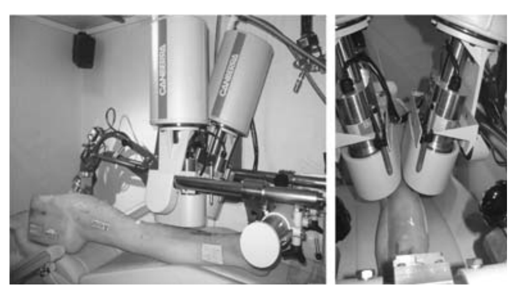

Figure 2. *In vivo* monitoring of the USTUR leg at CIEMAT. Vertical position and wrapping counting geometry.

modelling of LE Ge detector was carried out according to the description of the CONRAD MC intercomparison[3]. A general good agreement was found except at the centre-knee point, where discrepancies are due to the great sensitivity of calculated efficiencies with reproducing detector positioning (Figure 3).

HMGU (Munich, Germany)

Measurements of the USTUR leg phantom were also carried out at the Partial Body Counter of HMGU, where four different high-purity Ge detectors are available. Detectors #2 and #3 are identical, with a diameter of 5 cm and a thickness of 1.2 cm. Detector #4 includes a Germanium crystal with a diameter of 8 cm and a thickness of 2 cm, whereas detector #6 has a diameter of 7 cm and a thickness of 3 cm. The detectors are located inside a massive shielding chamber made of concrete, steel and copper, the chamber being located ~8 m below the surface of the earth to provide additional shielding against cosmic radiation.

Figure 4 shows the detection efficiencies obtained for the detectors #2, #3 and #4 when each of them was moved separately along the central axis of the USTUR leg phantom; positive distance means towards the foot, whereas negative distance means towards the hip. A counting time between 5000 and 10 000 s was chosen depending on detector and detector position.

Clearly all detectors show a maximum of the efficiency at distance zero, at the centre-knee position of the USTUR leg phantom, whereas the efficiency decreases towards the foot and the hip due to the specific distribution of bone material and, correspondingly, of the ^{241}Am within the phantom. As expected, detector #4 shows the highest efficiency due to its large crystal diameter.

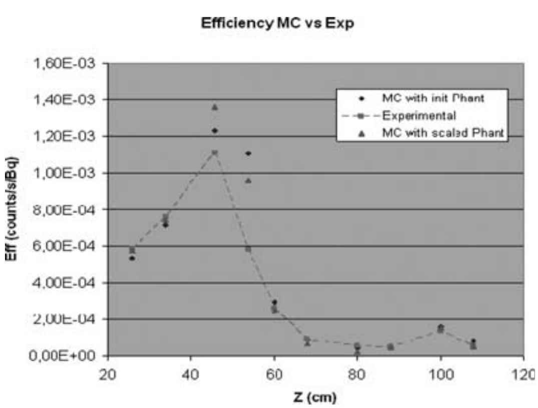

Figure 3. CIEMAT: counting versus MC efficiencies.

When three (Figure 4) or four detectors were used at the same time, it turned out that efficiency of a single detector may change significantly depending on the person who positioned the detectors. However, the total efficiency obtained summing up the spectra from the group of detectors turned out to be much less sensitive to the exact position of the detectors (Figure 4).

HML (Health Canada)

The phantom was measured (1) physically (*in vivo* counting facility) and (2) by MC simulations.

(1) The shielded room of the HML whole-body counter consists of iron walls lined with Pb/Sn/Pu. One Ge detector was placed in different positions so as to perform a scan over the leg. The Ge detector is 8.5 cm in diameter and 3 cm thick. The detector was moved incrementally over the leg; 15 counts were performed. Each count was 60 000 s except for position 6 (300 000 s). Cross-talk was evaluated by measuring the USTUR leg and having an adjacent detector active over a polyurethane cylinder (Figure 5).

(2) MC simulation (MCNPX)—efficiency pattern. Two leg voxel phantoms were used:

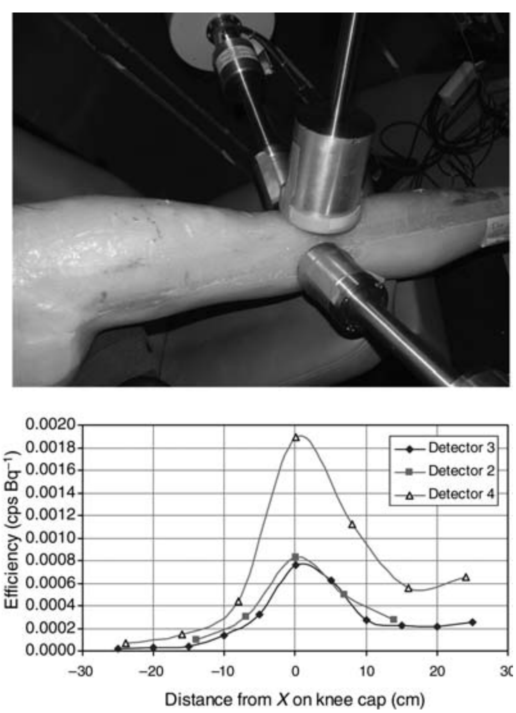

Figure 4. Efficiency pattern at HMGU (central axis, top of the leg), counting geometry: three-detectors configuration.

(a) IRSN Voxel phantom obtained from a CT scan; homogeneous distribution of activity in the skeleton
(b) HML Voxel phantom generated from USTUR website (CT images). Source distributed in the bone of the leg sections (hip, femur, patella, tibia and fibula) according to USTUR radiochemical data.

The HML study of MC simulation versus experimental values for the counting efficiency pattern shows an excellent agreement when using the HML Voxel Phantom (Figure 6).

COMPARISON STUDY

The analysis of the experimental efficiency patterns found at the *in vivo* facilities of the participants shows an agreement on the maximum efficiency value for the centre-knee position (Figure 7). Great difficulties were found in comparing data generated by participants due to the sensitivity of the results to reproducing the positioning of detectors.

CIEMAT missed the maximum of the efficiency pattern due to the expectation of the highest photon fluence at the low segment of the knee (obtained with the Spitz calibration phantom constructed with bone-simulated material).

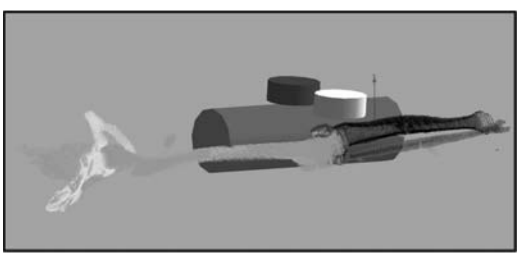

Figure 5. HML Ge detector was placed in the virtual model in the same position as the experimental counts.

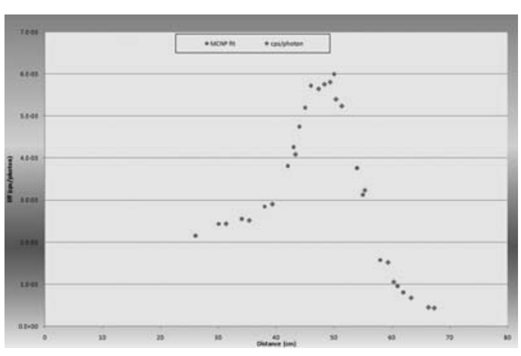

Figure 6. HML counting efficiency of the ^{241}Am bone phantom at 59.5 keV. Experimental versus MC values.

CONCLUSIONS

Each participant proceeded with measurements (Ge detectors) and/or MC simulations. CIEMAT and HMGU provided experimental results; IRSN performed an MC study generating an USTUR voxel phantom, and HML carried out an experimental plus computational approach. Great difficulties were found in comparing data generated by participants due to the sensitivity of the results to reproducing the positioning of detectors.

IRSN recommended the revision of the activity value provided by the USDOE certificate for the USTUR leg phantom; a re-evaluation of the ^{241}Am content was carried out using the available data from the right leg (radiochemistry and relative bone weights). The final value of the activity showed a discrepancy of 15 % with reference of the original certificate value.

Analysis of the efficiency patterns measured at the *in vivo* facilities involved showed an agreement on the maximum efficiency value for the centre-knee position. This conclusion has a different approach when comparing with the CIEMAT study for optimisation counting efficiency using the Spitz calibration phantom, where a maximum photon fluence value of 59.5 keV was measured in the low segment of the knee. Thus, the USTUR leg phantom and the Spitz knee calibration phantom are different. Future work is required to compare both phantoms in more detail.

Excellent agreement was found in the computational efficiency data when compared with the counting (experimental) calibration factors, using the two USTUR voxel phantoms (generated by HML and IRSN, respectively) in the efficiency pattern of HML (Health Canada) data.

This work confirms that the application of voxel phantoms and MC techniques to *in vivo* assessment of internal radionuclide body burdens is a valid alternative for calibration purposes.

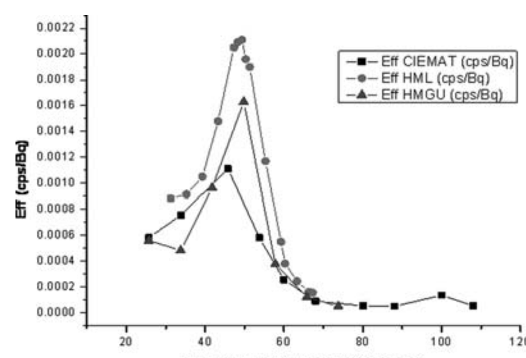

Figure 7. Comparison of counting efficiencies—one Ge detector in the vertical position.

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REFERENCES

1. Breitenstein, B. D. *et al*. *The U.S. Transuranium Registry report on the ^{241}Am count of a whole body.* Health Phys. **49**, 559–648 (1985).
2. McInroy, J. F., Boyd, H. A., Eutsler, B. C. and Romero, D. *The U.S. Transuranium Registry report of the ^{241}Am content of a whole body. Part IV: preparation and analysis of the tissues and bones.* Health Phys. **49**, 587–621 (1985).
3. *USTUR 0102: University of California—1952–4 Wound, Chronic Inhalation—^{241}AmO2: Radiochemistry.* United States Transuranium and Uranium Registries. Washington State University, College of Pharmacy (2010) Available on http://www.ustur.wsu.edu/Case_Studies/Radiochemistry/xls/0102_RadChem_Rev.1.xls(accessed on 23 April 2010).
4. Navarro, J. F., Lopez, M. A. and Navarro, T. *Assessment of the internal dose of ^{241}Am in bone by in-vivo measurements of activity deposited in knee.* Radiat. Prot. Dosim. **127**(1–4), 531–534 (2007).
5. Gomez-Ros, J. M., de Carlan, L., Franckm, D., Gualdrinim, G., Lis, M., Lopez, M. A., Moraleda, M. and Zankl, M. *Monte Carlo modeling for in vivo measurements of americium in a knee voxel phantom: general criteria for an international comparison*. Radiat. Prot. Dosim. **127**(1–4), 245–248 (2007).