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The Enhanced Naturally Occurring Radioactivity of Negative Ion Clothing and Attendant Risk

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Abstract: The study investigates commercially available negative ion clothing, and evaluations are made using gamma-ray spectroscopy and Geant4 Monte Carlo simulations. Observed to contain naturally occurring radioactive material (NORM), evaluations are made of the radiological risk arising from the use of these as items of everyday wear, undergarments in particular. Organ doses from these were simulated using the MIRD5 mathematical female phantom, with the incorporation of dose conversion factors (DCFs). At 175 ± 26, 1732 ± 247, and 207 ± 38 Bq, for ^{238}U, ^{232}Th, and ^{40}K respectively, item code S05 was found to possess the greatest activity, while item code S07 was shown to have the least activity, at 2 ± 0.5 and 15 ± 2 Bq, and again for ^{238}U and ^{232}Th, respectively. Sample code S11 recorded least activity, at 29 ± 5 Bq, for ^{40}K. Among the clothing items, sample item code S05 offered the greatest concentrations of Th, U and Zr, with percentage means of 1.23 ± 0.1, 0.045 ± 0.001, and 1.29 ± 0.1, respectively, giving rise to an annual effective dose of 1.57 mSv/y assuming a nominal wearing period of 24 h per day. Accordingly, the annual public dose limit of 1 mSv can be exceeded by their use.

Keywords: consumer products; undergarment; Geant4 Monte Carlo simulation; regulations

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1. Introduction

Humans are continually exposed to ionizing radiation, most pointedly from the natural radionuclides within terrestrial media. The ubiquitous nature of naturally occurring radioactive material (NORM) is well understood, with a presence in soil, water, and air. Less well appreciated is that NORM also appears in consumer products, including in building materials [1]. While exposure to ionizing radiation emitted by ^{238}U, ^{232}Th, and ^{40}K can constitute a risk to the living environment [2,3], the low doses from most natural environments do not typically give cause for concern. However, certain anthropogenic activities can increase radioactivity concentration levels, with elevated doses sometime well beyond those of the natural background [4]. For individuals who are exposed at these levels, NORM contributions may cause cancer [5]. Accordingly, from the viewpoint of radiation protection, the monitoring of natural radioactivity in human surroundings is of primary importance.

In Malaysia, the country in which present study was conducted, a recent health fad concerns online undergarment products markets, and the present study addresses the NORM content of these so-called negative ion garments. Negative ion technology has been suggested to offer alternative medical benefits [6]. At the outset, let it be noted that the term 'negative ion' finds no conventional scientific definition, and is considered by the present authors to be meaningless jargon. The manufacturers of these undergarments state them to be composed of tourmaline or minerals; the purveyors and manufacturers linking the supposed health benefits with negative ion technology, using further jargon such as "negative ion waist shaper". The claimed health benefits include enhanced magnetic therapy, improved cardiovascular function, blood circulation, immune system function, and also the burning of excess fats and calories, together with a boosting of metabolism, the removal of bodily toxins, and assistance in material detoxification [5,6]. Additional claims include that they are a way to maintain a balance in energy and improved emotional well-being, as well as protecting against electromagnetic fields [7].

Despite NORM-added consumer products being readily available in the market, little is known about their effects in terms of dose exposure and any potential hazard to health, and they are fully unmonitored. In the only other related study that we have been able to find in the literature, it was reported that an inhalation risk can exist in the case of misuse of NORM-added toiletry products, available in the Japanese and Korean markets [5,8–10]. Given that, over time, risks have been identified, NORM has become increasingly subject to monitoring and regulation [9]. In particular, while the IAEA safety guide "Radiation Safety for Consumer Products" [11] recommends that the use of such items first be justified, negative ion garments are being sold on the open market with no supportable scientific evidence being proffered in terms of benefits to the general public from the added radioactive component [11,12]. Among notable particular actions are that of the United States Environmental Protection Agency (EPA) via its guidelines for regulation of NORM [13]. Despite such isolated actions, globally harmonized regulations remain to be established, regulating and controlling radioactive content consumer products [14]. A further isolated action in seeking to limit radiation exposure from NORM-added consumer products is that of the Malaysian Atomic Energy Licensing Board (AELB), seeking control of such items via its technical document LEM/TEK/69 [15]. The intent of present study is to investigate negative ion undergarments, the activity of the long-lived radionuclides ^{238}U, ^{232}Th and ^{40}K being measured along with potential dose to wearers, the latter evaluated via use of Geant4 Monte Carlo (GMC) simulation. Accordingly, exposure determination was obtained via the use of the MIRD5 mathematical female phantoms, with evaluation made of dose conversion factors (DCFs) and organ dose, focusing on assessing external exposure to wearers.

2. Materials and Methods

2.1. Sampling and HPGe γ-Spectrometry

The undergarments are said to contain tourmaline, zirconium, anions and monazite, monazite and zirconium being known to contain uranium and thorium [9]. The manufacturers claim that the negative ion underwear products contain more than 2000 negative ions, detailed meaning of this being unspecified. For the current study, a total of 13 different forms of undergarment were purchased online; upon delivery, there was no evidence of information on the radioactivity contained within. The products were made by seven different brands (Figure 1).

Figure 1. Undergarment samples.

Analysis was made through use of a Pb-shielded high purity Ge (HPGe) spectrometer. For this, parts of each sampled undergarment were cut into 2 mm square pieces and placed in a standard 500-mL Marinelli beaker. The volume and shape of the sampled media were fixed to allow for solid angle correction. The duration of counting of radioactivity was 86,400 s (24 h). Given that the undergarments were composed of radioactive and non-radioactive portions, the former were separated out and only these parts were used in gamma spectroscopy. To allow for secular equilibrium, ^{238}U and ^{232}Th were measured subsequent to 30 days of sealed storage. Measurement of gamma emissions were made directly using an ORTEC GEM Series P-type coaxial HPGe spectrometer (GEM20-76-LB-C-SMPCFG-SV-LB-76; 30% relative efficiency; 1.8 keV FWHM at 1332 keV), with spectra were collected over 16,380 channels. The software Gamma Vision 8.1 was used for the gamma-ray spectrum acquisition, analysis, and reporting of the sample. Calibrations were made using a 500-mL standard multi-nuclides source composed of ^{210}Pb, ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co, all distributed in a soil matrix. Offering a wide range of photon energies, all of the emissions used were greater than 300 keV so as to be very minimally affected by any possible differences in densities between the soil standard samples and the samples under analysis.

2.2. Elemental Composition

For elemental analysis, use of a Cartesian Geometry Energy Dispersive X-Ray Fluorescence (ED-XRF) spectrometer (model; NEX CG – CG1240), XRF offering non-destructive investigation of materials composition, including consumer products, was made. The system, supported by new qualitative and quantitative analytical software (RPF-SQX) featuring Rigaku Profile Fitting (RPF) technology, allowed semi-quantitative analysis of almost all sample types without standards, and quantitative analysis with standards [6,16]. For lower atomic numbers, element composition was generally analyzed using characteristic K_α lines, while elements with higher atomic numbers (Cs to U) were typically measured using the respective L-line emissions [17]. The samples were prepared in a homogenized powder form. XRF analysis of four such garments was carried out, with a duration of 12 min per sample.

2.3. Geant4 Monte Carlo (GMC) Simulation and Evaluation of Annual Effective Dose (AED)

Effective dose cannot be directly measured [18]. To estimate the equivalent organ doses and annual effective dose (AED) in use the of the garments, simulations were undertaken using the Geant4 Monte Carlo radiation transport code, version 10.06 patch 3, Physics list:

Geant4 electromagnetic (EM) physics (G4EmStandardPhysics_option4), also involving use of Medical Internal Radiation Dose Pamphlet 5 (MIRD5) mathematical female phantoms, with the female version shown in Figure 2 [9,19,20]. Each phantom consisted of three significant parts: (i) trunk and arms, represented by an elliptical cylinder; (ii) legs and feet represented by two truncated circular cones, and; (iii) the neck and head represented by an elliptical cylinder capped by a half ellipsoid sitting on a circular cylinder. For female breasts half-ellipsoids were attached to the trunk. In calculations of the effective dose for NORM-added consumer products, Yoo et al. [21], Lee et al. [9], and Yoo et al. [22] suggested the use of the point source method instead of using full modeling of the consumer products using GMC simulation methods. Gamma photons are simulated: from ^{232}Th; ^{212}Pb (238 keV), ^{228}Ac (911 keV), ^{228}Ac (969 keV), ^{208}Tl (583 keV), ^{208}Tl (2614 keV), and for ^{238}U series; ^{226}Ra (186 keV) and ^{238}U series. The single emission of ^{40}K was also simulated.

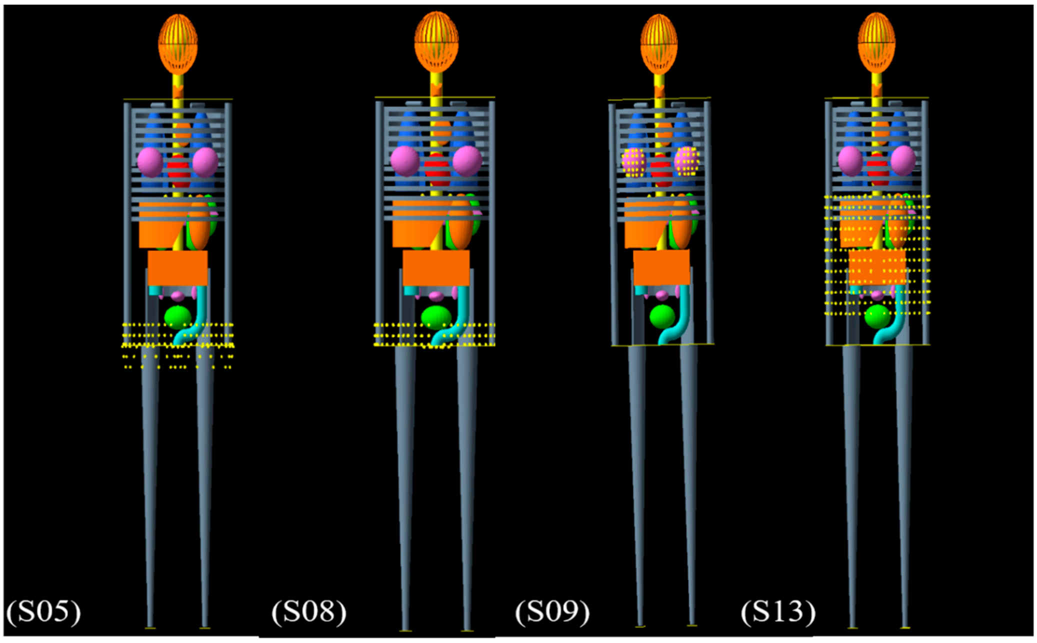

Figure 2. MIRD human phantoms for undergarments, as used in the Geant4 MC simulation; yellow dots represent the undergarment-covered area.

The female phantom measured 174 cm in height and 70 kg in weight. With the exception of the skeleton and lungs, all organs were soft tissue, the MIRD soft tissue density was 0.9869 g/cm^3, while the MIRD skeleton density was 1.4862 g/cm^3, and the MIRD lung material density was 0.2958 g/cm^3. The undergarment products were simulated to be 1 mm in distance from the covered skin surface; the yellow dots of Figure 2 representing the area covered. Dose evaluations were carried out with the undergarments located at four different areas of the female phantom, as indicated.

3. Effective Dose Estimate

3.1. Estimation of Equivalent Organ Dose and Annual Effective Dose in Wearing an Undergarment

In estimating the radiological impact the garments may have on a population of users, assessments were made of the organ equivalent and average effective doses by measuring the activity of ^{238}U, ^{232}Th, and ^{40}K within the negative ion underwear samples. Additionally, dose conversion factors were calculated using Geant4 Monte Carlo simulations and use of the tissue weighing factors. Denoted by H_T, the radiological protection quantity equivalent dose in an organ or tissue is given by:

$$H_T = \Sigma\, w_R\, D_{T,R}$$

where H_T (Sv) is the equivalent dose of an organ or tissue, w_R denotes the radiation weighting factor for gamma radiation, taken as unity (1), and $D_{T,R}$ (Gy) is the mean absorbed dose from radiation (R) in an organ or tissue (T) [23,24]. The effective dose (E_{eff}), defined by the IAEA [25] and the ICRP [23], is formed as a weighted sum of tissue equivalent dose, as:

$$E_{eff} = \Sigma\, w_T\, H_T$$

with w_T being the tissue weighting factor, and the summed w_T is unity [23,26–28]. $D_{T,R}$ is given by the expression:

$$D_{T,R} = A \times t \times DCFs$$

with A being the measured activity of ^{238}U, ^{232}Th and ^{40}K in Bq/product, t is the exposure time due to wearing the undergarment and $DCFs$ represents the dose conversion factors for these radionuclides [29,30].

3.2. Dose Rate Using HPC Software

The dose rate in air, bone, and muscle, calculated from the wearer, was obtained through use of Syberad's Health Physicist's Companion program [31]. This provides for the calculation of the dose rate for any radionuclide, for a given specific activity, and at any distance, with provisions for shielding [28]. Dose rates were calculated for radionuclides ^{238}U, ^{232}Th and ^{40}K in the undergarment, at an close-up distance of 1 mm from the user, as previously mentioned [30].

3.3. Measuring Dose Equivalent Using a Survey Meter

The dose equivalent from the undergarment was also measured, with a calibrated IdentiFinder 2 ULK-NGH being used, and an FLIR Survey Meter, for recording exposure dose rates (in µSv/h) three times for each product, quoted together with the standard deviation, also identifying the radionuclides contained in the undergarments.

4. Results and Discussion

4.1. Measurements of Activity

Spectroscopy measurements were carried out under secular equilibrium conditions, considering the lines of the short-living decay products. Using the coaxial HPGe detector, gamma spectrometry measurements were made for each of the 13 undergarment products (Table 1). For ^{238}U, results were estimated from the average of gamma emissions from ^{214}Pb (295 keV and 351.8 keV) and ^{214}Bi (609 keV and 1764 keV), while for ^{232}Th these were estimated from the average of the gamma emissions of ^{212}Pb (238 keV), ^{228}Ac (338 keV and 911 keV), ^{212}Bi (727 keV), and ^{208}Tl (583 keV and 2614 keV).

Undergarment S05 showed the greatest activity at 175 ± 26, 1732 ± 247, and 207 ± 38 Bq, for ^{238}U, ^{232}Th, and ^{40}K, respectively. Across the same category of undergarments, considerable differences in radioactivity content could be observed, an example being the variation of ^{238}U series of between 2 ± 0.5 and 175 ± 26, while for the ^{232}Th series it was between 15 ± 2 and 1732 ± 247; for ^{40}K it was between 29 ± 5 and 207 ± 38. For elemental content, four undergarments were analyzed via the use of an ED-XRF spectrometer (Table 2). Table 2 shows the elemental compositions of the negative-ion products. The four analyzed samples were made principally of Cl and Ag. NORM (U, Th, and K) and rare earth elements (REEs) are found in all four analyzed samples, S05 recording the greatest concentration percentages of Th, U, and Zr, at 1.23 ± 0.1, 0.045 ± 0.001, and 1.29 ± 0.1, respectively. The X-ray energy line for Th, U and Zr was at L_α; 12.96 keV, L_α; 13.61 keV and L_α; 2.044 keV, respectively. The concentrations (and activities) of Th and U for sample code S05 were 12.31 parts per thousand (1746 Bq) and 450 ppm (194 Bq), respectively. The results for sample S05, as shown in Tables 1 and 2, are comparable.

Table 1. Activity (Bq/product) of ^{238}U, ^{232}Th, and ^{40}K for undergarments.

Sample	Description	Weight (g)	Activity ± 1 Sigma (Bq/Product)		
			U-238	**Th-232**	**K-40**
S01	Briefs underwear	40 ± 0.5	61 ± 9	657 ± 127	135 ± 18
S02	Waist anion slimming hip abdomen pants	89 ± 1	4 ± 0.5	57 ± 3	77 ± 14
S03	Breathing underwear	29 ± 0.3	3 ± 0.5	26 ± 2	75 ± 11
S04	Seamless shapewear negative ion	119 ± 1.4	82 ± 11	487 ± 88	126 ± 23
S05	Shorty brief underwear	35 ± 0.4	175 ± 26	1732 ± 247	207 ± 38
S06	Slimming underwear	109 ± 1.3	11 ± 2	27 ± 4	78 ± 14
S07	Bra	34 ± 0.3	2 ± 0.5	15 ± 2	76 ± 13
S08	Full brief underwear	61 ± 0.7	110 ± 17	1225 ± 184	193 ± 36
S09	Bra-VIP	18 ± 0.2	31 ± 4	180 ± 31	87 ± 16
S10	Set-underwear	69 ± 0.7	3 ± 0.5	33 ± 5	79 ± 14
S11	Breathing underwear	30 ± 0.3	5 ± 1	50 ± 7	29 ± 5
S12	Seamless shapewear negative ion	115 ± 1.4	46 ± 7	328 ± 35	87 ± 15
S13	Ionizer waist shaper	55 ± 0.6	158 ± 24	1648 ± 221	183 ± 31
Minimum			2 ± 0.5	15 ± 2	29 ± 5
Maximum			175 ± 26	1732 ± 247	207 ± 38

Table 2. Elemental composition of the undergarment.

Element	Samples (% Concentration)			
	S05	**S08**	**S09**	**S13**
Cl	38.7 ± 3.2	4.22 ± 0.4	30.5 ± 3.4	3.54 ± 0.11
Ag	24.9 ± 2.1	38.9 ± 2.9	0.92 ± 0.2	2.72 ± 0.13
Al	1.12 ± 0.1	2.57 ± 0.2	0.76 ± 0.1	0.11 ± 0.01
Si	0.17 ± 0.01	7.06 ± 0.3	ND*	0.23 ± 0.01
KCa	0.15 ± 0.004 2.21 ± 0.4	0.03 ± 0.001 1.19 ± 0.1	0.21 ± 0.01 54.1 ± 0.03	0.02 ± 0.001 0.59 ± 0.03
Ti	9.44 ± 0.6	14.1 ± 0.8	3.58 ± 0.3	45.6 ± 3.8
Cr	9.93 ± 0.7	3.63 ± 0.3	ND	33.1 ± 2.5
S	0.46 ± 0.04	ND	0.44 ± 0.03	0.41 ± 0.02
Co	ND	0.43 ± 0.03	0.29 ± 0.02	ND
Sr	0.30 ± 0.03	0.18 ± 0.01	0.20 ± 0.02	0.02 ± 0.001
Zr	1.29 ± 0.1	0.91 ± 0.04	0.41 ± 0.04	0.59 ± 0.03
U	0.045 ± 0.001	0.017 ± 0.001	0.019 ± 0.001	0.028 ± 0.001
Pb	0.04 ± 0.006	0.093 ± 0.001	0.023 ± 0.001	0.02 ± 0.001
La	2.85 ± 0.2	4.14 ± 0.3	1.03 ± 0.1	ND
Ce	ND	0.97 ± 0.1	0.72 ± 0.05	0.64 ± 0.05
Th	1.23 ± 0.1	0.44 ± 0.03	0.28 ± 0.01	0.81 ± 0.06
Eu	0.11 ± 0.01	ND	0.06 ± 0.002	0.08 ± 0.001
Ba	5.13 ± 0.3	10.6 ± 0.9	1.28 ± 0.12	2.07 ± 0.3
Po	0.15 ± 0.01	0.37 ± 0.02	0.37 ± 0.02	0.61 ± 0.06
Ru	0.23 ± 0.01	ND	1.12 ± 0.1	0.51 ± 0.03
Gd	0.28 ± 0.02	0.37 ± 0.03	0.92 ± 0.1	0.19 ± 0.01

ND*: not detected.

4.2. Evaluation of Organ Equivalent Annual Effective Dose

Regarding the negative-ion products S05, S08, S09, and S13, Figure 3 shows the results for a total of 28 organs—the equivalent doses for each organ were calculated using the MIRD5 mathematical female phantom. Of these, the uterus, urinary bladder, ovaries, small and large intestine, breast and heart are among those most greatly exposed, according to their close distance to the source. For S05, the greatest equivalent dose was found to be for the leg bone, at 2.61 mSv y^{-1} for the female phantom due to the area of exposure; a nominal wearing period of 24 h per day, 365 days per year being assumed [5]. Additionally, the equivalent dose to the pelvis and large intestine were also been found to be greater than that of other organs a result of the area of exposure and close distance from the negative-ion product. For S08, the greatest equivalent dose was that of the pelvis, at 2.35 mSv y^{-1}; the urinary bladder recording the next greatest equivalent dose at 2.34 mSv y^{-1}. For S09, the breast, thymus and heart equivalent doses were found to be relatively large, at 1.11, 0.93, and 0.96 mSv y^{-1}, respectively, again due to the area exposed and the close distance to the source. In the case of S13, the greatest equivalent dose was for the uterus and trunk, at 2.10 and 2.09 mSv y^{-1}, respectively. Furthermore, the pancreas and rib cage organs recorded the highest organ equivalent dose after the uterus and trunk, at 2.06 and 1.85 mSv y^{-1}, respectively, due again to the large area of exposure.

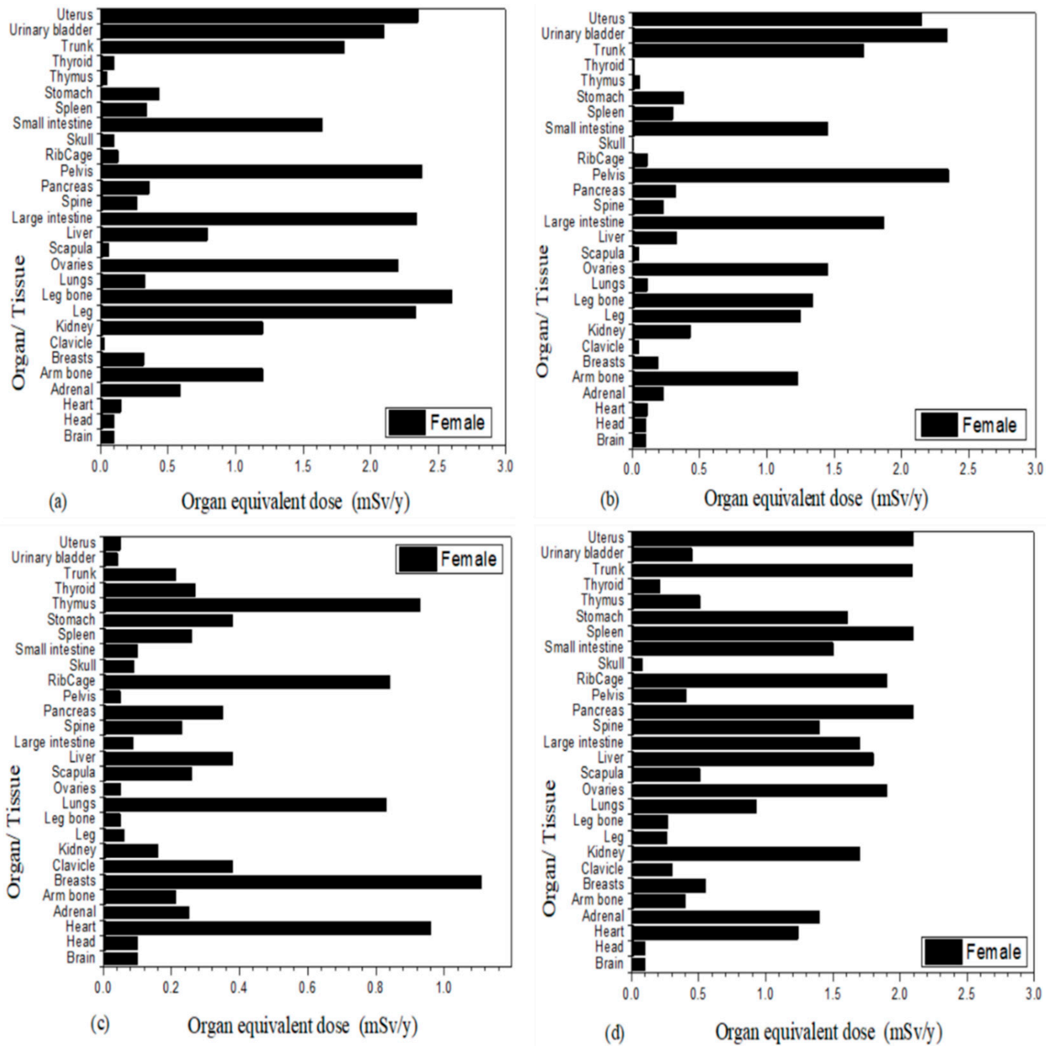

Figure 3. Organ equivalent dose (mSv/y) for 28 organs using the MIRD5 mathematical female phantoms, for garment items (**a**) S05; (**b**) S08; (**c**) S09; and (**d**) S13.

Table 3 shows the equivalent doses (in mSv/y), using Syberad's Health Physicist's Companion program; sample S05 recorded the highest annual doses, at 1.39, 1.61 and 2.04 mSv y^{-1} in air, bone, and muscle, respectively. The HPC program calculated doses based on the wearer being exposed at a distance 1 mm for a nominal period of 24 h per day and 365 days per year. For S08, the equivalent dose in leg bone and leg were 1.34 and 1.25 mSv y^{-1}, respectively, comparable with the HPC results for bone and muscle of 1.24 and 1.43 mSv y^{-1} (Table 3), in support of results obtained using the HPC program.

Table 3. Annual dose (in mSv/y) due to ^{238}U, ^{232}Th and ^{40}K, obtained using the HPC program.

Sample	Air	Bone	Muscle
S05	1.39	1.61	2.04
S08	0.92	1.24	1.43
S09	0.29	0.34	0.36
S13	1.24	1.42	1.75

The effective annual dose for the 13 undergarments are presented in Figure 4; S03 recorded the lowest annual effective dose, at 81 µSv y^{-1}, while undergarment S05 indicated the highest annual effective dose, at 1.57 mSv y^{-1}. As shown in Figure 4, wearing the negative ion undergarment can infer annual effective doses in excess of the annual dose limit of 1 mSv/y for members of the public [11,12]. In Figure 4, use of garments S05, S08 and S13, were seen to exceed the annual public dose limit 1 mSv/y. The beta emissions from the undergarment will be mostly absorbed within the epidermal layers of the skin, while the more deeply penetrating gamma radiation involves large areas of tissue and organs [32].

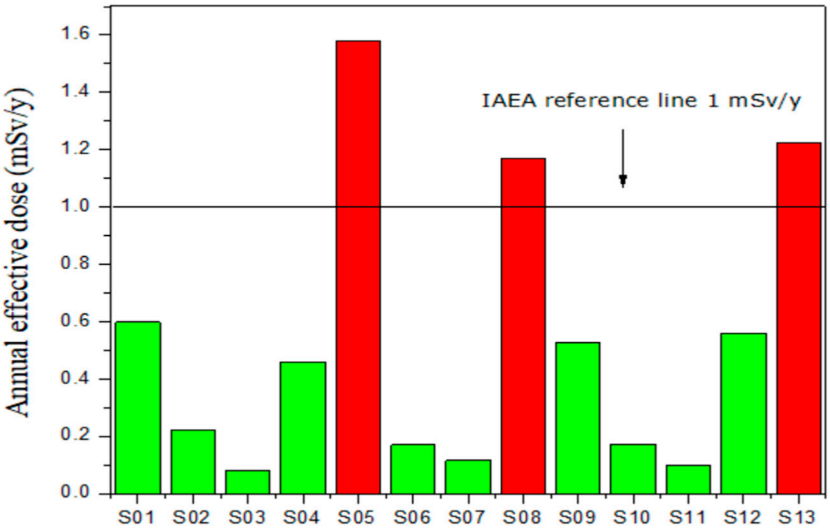

Figure 4. Annual effective dose for 13 undergarment products demonstrating measurable gamma activity.

The dose equivalent was also measured for the thirteen undergarments, capable of giving rise to annual effective doses in excess of 1 mSv/y. For this, use was made of a calibrated IdentiFinder 2 FLIR Survey Meter, which recorded dose rates in µSv/h and identifying the radionuclides contained within the undergarments (Table 4). The highest dose equivalent, at 0.24 ± 0.03 µSv/h, was from S05.

Regarding the wearing period, we suggested 24 h per day, 365 days a year to be likely for those who purchase such items [9]. For this, the annual dose from the use of items S05 and S13 could achieve values of 2.1 and 2 mSv y^{-1}, respectively, values that are comparable with the Geant4 simulation results. Note here that in its LEM-TEK-69 document, per NORM-added consumer product basis, the Malaysian Atomic Energy Licensing Board AELB requires the effective dose for individual members of the public to be less than 10 μSv/y. Conversely, for artificial radionuclides the European Commission advocates 300 μSv/y per product. Prior to ICRP 103, the exemption level was 10 μSv/y per product. The European Commission also prohibits the addition of radioactive material to consumer products [5]. Moreover, the EURATOM Council Directive prohibits the sale of clothes, personal jewelry, toys, foodstuffs, and cosmetics within which NORM have been added.

Table 4. Dose rate (μSv/h) of radioactivity in the undergarments, using a Survey Meter (IdentiFinder 2) portable detector.

No.	Sample	Dose Rate (μSv/h)
1	S01	0.11 ± 0.01
2	S02	0.09 ± 0.01
3	S03	0.12 ± 0.02
4	S04	0.14 ± 0.02
5	S05	0.24 ± 0.03
6	S06	0.11 ± 0.02
7	S07	0.09 ± 0.01
8	S08	0.22 ± 0.03
9	S09	0.17 ± 0.02
10	S10	0.11 ± 0.01
11	S11	0.09 ± 0.01
12	S12	0.19 ± 0.02
13	S13	0.23 ± 0.03

It is apparent that international harmonization of regulation protections is necessary. The idea of justification, 'ALARA' (as low as reasonably achievable) and dose limits, as promulgated by the ICRP, are highly important with respect to public safety. Unjustified exposures, as reported in ICRP-103, can be applied to a range of NORM added consumer products. If consumer products to which NORM have been added cannot be shown to offer benefits that far outweigh the risks, then prohibition would seem to be required [5,7]. This would certainly seem to be the case for the present products. Accordingly, a strong recommendation is made for worldwide prohibition of negative ion products.

5. Conclusions

Exposure to thirteen NORM-added negative-ion products has been investigated in terms of organ equivalent and annual effective dose. Products S05 and S13 recorded the greatest level of radioactivity, ^{238}U, ^{232}Th, and ^{40}K being found to be greater in sample S05. Estimation of organ dose equivalent and annual effective dose was obtained via Geant4 Monte Carlo simulations, leading to the conclusion that close contact with these products can infer annual effective doses of up to 2 mSv per year in excess of the dose constraint of 1 mSv y^{-1} for members of the public. The products are advertised to contain tourmaline, monazite and zircon, all known to contain uranium and thorium. Although absent of any sound scientific support for the inclusion of radioactive components; nevertheless, these products are available for purchase in a number of countries, Malaysia included, and in the absence of any indication of the items containing radioactivity. It is suggested that the lead should be taken from EURATOM, and that such products should likewise be prohibited from being sold. We strongly recommend that consideration be given to the prohibition of the importation and sale of these undergarment products.

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