



RESEARCH ARTICLE

²³⁸U AND ²³²Th CONCENTRATIONS MEASURED IN DIFFERENT MEDICAL DRUGS BY USING SOLID STATE NUCLEAR TRACK DETECTORS AND RESULTING RADIATION DOSES TO THE SKIN OF PATIENTS

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ABSTRACT

Urban populations in Morocco receive free medical drugs as prescribed by doctors in district health centres. To explore the exposure pathway of ²³⁸U, ²³²Th and their decay products to the skin of patients, these radionuclides were measured in various medical drugs by using solid state nuclear track detectors (SSNTDs). The measured concentrations range of ²³⁸U and ²³²Th in the medical drug samples of interest vary from (4.3±0.3) mBq.l⁻¹ to (11.1±0.7) mBq.l⁻¹ and (0.49±0.03) mBq.l⁻¹ to (1.3±0.1) mBq.l⁻¹, respectively. A new dosimetric model, based on the concept of specific alpha-dose and alpha-particle residual energy, was developed for evaluating radiation doses to skin following the application of different medical drugs by patients. The maximum total equivalent effective dose to skin due to the ²³⁸U and ²³²Th series from cutaneous application of different medical drugs by patients was found to be 2.8 mSv y⁻¹cm⁻².

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INTRODUCTION

The skin is the largest organ of the human body, with a total area of about 20 square feet. It protects individuals from microbes, helps regulate body temperature, and limits the sensation of touch, heat and cold. The skin has three layers: (i) The epidermis, the outermost layer of skin which provides a waterproof barrier and creates the skin tone; (ii) the dermis, beneath the epidermis which contains tough connective tissue, hair follicles, and sweat glands, and (iii) the deeper subcutaneous tissue (hypodermis) which is made of fat and connective tissue. The critical cells in the skin are in the basal layer of the epidermis. There are considerable variations in the thickness of human epidermis with respect to body site (International Commission on Radiological Protection, 1990). On the face and trunk the median thickness of the epidermis was 20-40 µm. In general, on the arms and legs it was 40-60 µm, although there were some considerably thicker areas on

the hands and feet (International Commission on Radiological Protection, 1990). A more detailed evaluation of the hands showed fingertips to have the greatest thickness, greater than 160 µm (International Commission on Radiological Protection, 1990). The degree of undulation of the basal layer was found to increase with increasing epidermal thickness. Naturally occurring radionuclides of terrestrial origin are present in various degrees in all media of the environment and contribute significantly to external and internal doses to the population (United Nations Scientific Committee on the Effects of Atomic Radiation, 2000). Among them, important radionuclides of interest belong to the ²³⁸U and ²³²Th series. These radionuclides emit alpha and beta-particles as well as gamma rays. The different forms of emitted radiation have different energies and penetrating power and thus have different effects on living beings. Once the radionuclides of the ²³⁸U and ²³²Th series are placed on the skin, they emit alpha-particles with a range of several tens of microns (between 20 µm and 100 µm). This is comparable with the depth of the basal layer of the epidermis. Due to their presence in soil and phosphate fertilizers, primordial radionuclides and their progeny are transferred via water from soil to plant flowers and medicinal plants and to

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medical drugs. Thus it is necessary to measure the radionuclide contents of medical drugs to assess the potential radiation doses, and if necessary, to take action to reduce the exposure of patients to radiation. ²³⁸U and ²³²Th concentrations have been measured in various medicinal plants by using solid state nuclear track detectors (Misdaq *et al.*, 2001). ²³⁸U and ²³²Th have also been analyzed in aerial parts and roots of *Peperomia pellucida* medicinal plant using alpha spectrometry after radiochemical separation by ionic exchange resins and measurement with a silicon surface-barrier detector (Sussa *et al.*, 2013). However, this technique is both destructive (chemical agents are added to the material sample) and expensive. ²³⁸U and ²³²Th have been analyzed in different food samples using inductively coupled plasma mass spectrometry (ICP-MS), which is also destructive (Shiraishi *et al.*, 2000). Committed effective doses due to the ²³⁸U and ²³²Th radioisotopes following the ingestion of various foodstuffs by individuals have been determined (Misdaq and Bourzik, 2002). In previous works, we evaluated committed effective doses to skin due to only three alpha-emitting nuclei (²³⁸U, ²³²Th and ²²²Rn) from the application of Moroccan black soap (Misdaq and Outeqablit, 2010) and olive oil (Misdaq and Touti, 2012) samples without taking into account the residual energies of the emitted alpha-particles.

In the present work, CR-39 and LR-115 type II solid state nuclear track detectors (SSNTDs) were used for measuring ²³⁸U and ²³²Th alpha-activities per unit volume in different medical drugs. During the full course of medical drugs application to different age groups of patients, the committed effective doses to the skin were evaluated due to alpha-particles emitted by the radio-nuclei of ²³⁸U and ²³²Th series.

MATERIALS AND METHODS

A. Description of the medical drugs studied

Medical drugs are cutaneously prescribed by doctors for patients in dermatology, cardiology, gastro-enterology, anesthesia-resuscitation, gynecology, pneumology, and rheumatology. The properties and dosages of the considered medical drugs are shown in Table 1.

B. Determination of ²³⁸U and ²³²Th alpha-activities per unit volume in medical drugs

The alpha-activities of ²³⁸U and ²³²Th were measured by using the following types of solid state nuclear track detectors (SSNTDs):

- CR-39 discs, 2 cm in radius and 500 μm thick, manufactured by Pershore Mouldings Ltd, United Kingdom;
- LR-115 type II discs, 2 cm in radius, comprising 12 μm of cellulose nitrate on a 100μm thick polyester base manufactured by Kodak Pathé, France, and marketed by Dosirad, France.

The detectors were separately placed in close contact with different medical drugs in hermetically sealed (using glue and a cellophane tape) HDPE (high density polyethylene) cylindrical plastic containers for 30 days (Fig.1). During this period of

time alpha-particles emitted by the nuclei of ²³⁸U, ²³²Th and their daughters inside the medical drug samples exposed the SSNTD films. After the irradiation, the exposed SSNTDs were etched in two NaOH solutions: one was of 2.5 mol l⁻¹ at 60°C during 2 hours for the LR-115 II films and the other of 6.25 mol l⁻¹ at 70°C for 7 hours for the CR-39 detectors (Misdaq *et al.*, 2000). After chemical treatment, the track densities registered on the CR-39 and LR-115 II SSNTDs were determined by means of an ordinary microscope. Backgrounds on the CR-39 and LR-115 II SSNTDs were evaluated by placing these films in sealed plastic containers, containing ambient air, identical to those used for analyzing the medical drug samples for 30 days and counting the resulting track densities. This operation was repeated ten times, and it was found that the track densities registered on the CR-39 and LR-115 II detectors were identical within the statistical uncertainties. The reproducibility of the method was checked by analyzing a set of ten samples of the same medical drug. Track density production rates registered on the CR-39 and LR-115 II detectors were evaluated for the P13 medical drug sample. Data obtained, for instance, for the P13 medical drug sample are: $\rho_G^{CR} = (2.41 \pm 0.01) \cdot 10^{-5}$ tracks cm⁻² s⁻¹ and $\rho_G^{LR} = (9.25 \pm 0.05) \cdot 10^{-5}$ tracks cm⁻² s⁻¹, respectively. The relative uncertainty of the average track density rate determination is smaller than 1%. Fig.1. Arrangement of a solid state nuclear track detector (SSNTD) on a medical drug material sample in a well-sealed plastic container of radius q=2 cm, depth D=1 cm and thickness t=0.5 cm. Glue is put between the plastic cover and plastic container and both are covered by a cellophane tape of 0.2 cm thickness.

There are three main factors which disturb the radioactive secular equilibrium between ²³⁸U and its progeny and between ²³²Th and its daughters: (a) the addition of any chemical compounds to the medical drug sample, (b) any escape of radon and thoron gases and (c) the exposure time if it is smaller than 25 days. As the detection system used was well-sealed (i.e., there was no escape of radon and thoron) and the exposure time was 30 days, a radioactive secular equilibrium is established between ²³⁸U and each of its decay products and between ²³²Th and each of its daughters. For the experimental etching conditions, the residual thickness of the LR-115 type II detectors measured by means of a mechanical comparator is 5 μm. This thickness defines the lower (E_{min}= 1.6 MeV) and upper (E_{max} = 4.7 MeV) energy limits for registration of tracks of alpha-particles

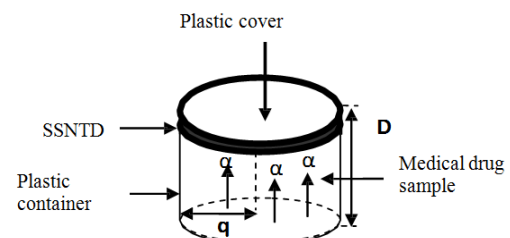


Fig.1. Arrangement of a solid state nuclear track detector (SSNTD) on a medical drug material sample in a well-sealed plastic container of radius q=2 cm, depth D=1 cm and thickness t=0.5 cm. Glue is put between the plastic cover and plastic container and both are covered by a cellophane tape of 0.2 cm thickness

in LR-115 type II films (10). All alpha- particles emitted by the ²³⁸U and ²³²Th series that reach the LR-115 II detector at an angle smaller than its critical angle of etching, θ'_c , with a residual energy between 1.6 MeV and 4.7MeV are registered as bright track-holes. The CR-39 detector is sensitive to all alpha-particles reaching its surface at an angle smaller than its critical angle of etching, θ_c . The critical angles of etching θ'_c and θ_c were calculated by using the method described in detail by Misdaq *et al.* (1999). The global track density rates (tracks $\text{cm}^{-2} \text{s}^{-1}$), due to alpha-particles emitted by the ²³⁸U and ²³²Th series inside a material sample, registered on the CR-39 (ρ_G^{CR}) and LR-115 II (ρ_G^{LR}) detectors, after subtracting the corresponding backgrounds, are respectively given by (Misdaq *et al.*, 2000):

$$\rho_G^{CR} = \frac{\pi q^2}{2S_d} A_c(^{238}\text{U}) \left[\sum_{j=1}^8 k_j \varepsilon_j^{CR} R_j + \frac{A_c(^{232}\text{Th})}{A_c(^{238}\text{U})} \sum_{j=1}^7 k'_j \varepsilon_j'^{CR} R'_j \right] \dots(1)$$

and

$$\rho_G^{LR} = \frac{\pi q^2}{2S'_d} A_c(^{238}\text{U}) \left[\sum_{j=1}^8 k_j \varepsilon_j^{LR} R_j + \frac{A_c(^{232}\text{Th})}{A_c(^{238}\text{U})} \sum_{j=1}^7 k'_j \varepsilon_j'^{LR} R'_j \right] \dots(2)$$

where: $A_c(^{238}\text{U})$, expressed in Bq cm^{-3} , is the activity per unit volume of ²³⁸U inside a medical drug sample. $A_c(^{232}\text{Th})$, expressed in Bq cm^{-3} . S_d and S'_d are respectively the surface areas of the CR-39 and LR-115 II films. R_j and R'_j are the ranges, in the medical drug sample, of an alpha-particle of index j and initial energy $E_{\alpha j}$ emitted by the nuclei of the ²³⁸U and ²³²Th series, respectively. k_j and k'_j are respectively the branching ratios corresponding to the disintegration of the nuclei of the ²³⁸U and ²³²Th series. $\varepsilon_j^{CR}, \varepsilon_j'^{CR}, \varepsilon_j^{LR}$ and $\varepsilon_j'^{LR}$ are respectively the detection efficiencies of the CR-39 and LR-115 II detectors for the emitted alpha-particles (Misdaq *et al.*, 2000).

Combining Eqs. 1 and 2, we obtain the following relationship between track density rates and ²³²Th to ²³⁸U ratios:

$$\frac{A_c(^{232}\text{Th})}{A_c(^{238}\text{U})} = \frac{\frac{S'_d}{S_d} \sum_{j=1}^8 k_j \varepsilon_j^{CR} R_j - \frac{\rho_G^{CR}}{\rho_G^{LR}} \sum_{j=1}^8 k_j \varepsilon_j^{LR} R_j}{\frac{\rho_G^{CR}}{\rho_G^{LR}} \sum_{j=1}^7 k'_j \varepsilon_j'^{CR} R'_j - \frac{S'_d}{S_d} \sum_{j=1}^7 k'_j \varepsilon_j'^{LR} R'_j} \dots\dots(3)$$

The ²³⁸U alpha-activity per unit volume of a medical drug sample is given by (Eq.2):

$$A_c(^{238}\text{U}) = \frac{2S'_d \rho_G^{LR}}{\pi q^2 \left[\sum_{j=1}^8 k_j \varepsilon_j^{LR} R_j + \frac{A_c(^{232}\text{Th})}{A_c(^{238}\text{U})} \sum_{j=1}^7 k'_j \varepsilon_j'^{LR} R'_j \right]} \dots(4)$$

By measuring ρ_G^{CR} and ρ_G^{LR} track density rates and calculating $\varepsilon_j^{CR}, \varepsilon_j'^{CR}, \varepsilon_j^{LR}$ and $\varepsilon_j'^{LR}$ detection efficiencies (Misdaq *et al.*, 2000) we evaluate the $\frac{A_c(^{232}\text{Th})}{A_c(^{238}\text{U})}$ ratio (Eq.3) and consequently the ²³⁸U and ²³²Th alpha-activities per unit volume in a given medical drug sample (Eq. 4). The ranges of the emitted alpha-particles in medical drugs and SSNTDs were calculated by using the TRIM

(Transport of Ions in Materials) programme (Biersack and Ziegler, 1998).

C. A new dosimetric model for evaluating annual committed equivalent doses to skin due to alpha-particles emitted by the nuclei of the ²³⁸U and ²³²Th series from cutaneous application of medical drugs

The epidermis of the human skin is divided into several clearly defined zones (International Commission on Radiological Protection, 2007). Indeed, when a medical drug layer is placed on the skin of a patient, the nuclei of the ²³⁸U and ²³²Th series emit alpha-particles with a range of several tens of microns (20 to 100 μm) (Table 2). This is comparable with the depth of the basal layer of the epidermis which is more sensitive (50 to 100 μm) (International Commission on Radiological Protection, 2010). An alpha-particle of index j and initial energy $E_{\alpha j}$ emitted from a nucleus localized on the point M inside the medical drug layer (Fig.2) has a range:

$$\overline{MF} = x_j + R_j^{Skin} \dots\dots\dots(5)$$

where $x_j(x_j \leq R_j)$, x_j is the range of the alpha-particle inside the medical drug layer) is the distance between the emission point and the skin surface (Fig.2) and R_j^{Skin} is the range of the alpha-particle in skin.

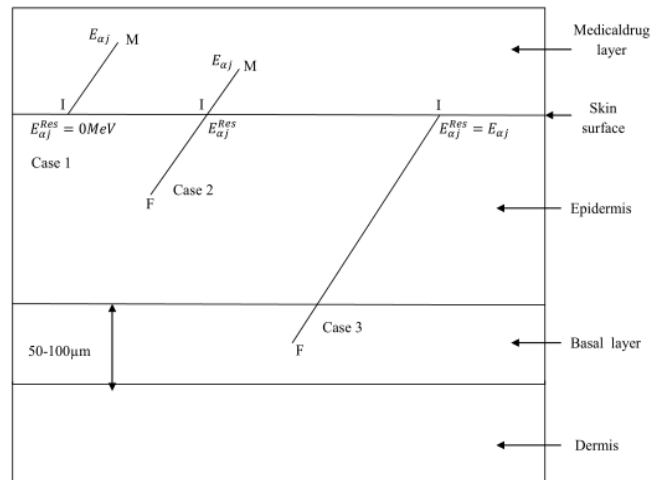


Fig. 2. Ranges of an alpha-particle inside the medical drug layer ($\overline{MI} = x_j$) and epidermis ($\overline{IF} = R_j^{Skin}$). $E_{\alpha j}$ is the initial alpha-particle energy and $E_{\alpha j}^{Res}$ its residual energy on the point I. The medical drug layer has a depth of about 500 μm .

The alpha-particle residual energy $E_{\alpha j}^{Res}$ which corresponds to the $(R_j - x_j)$ determined by using the energy-range relation in medical drug (Fig.3 (a)). By using the energy-range relation in skin one can determine the range of the alpha-particle in skin R_j^{Skin} . For $x_j = R_j, E_{\alpha j}^{Res} = 0$ Mev; there is no energy loss of alpha-particles in skin (case 1 of Fig.2). For $x_j = 0\mu\text{m}, E_{\alpha j}^{Res} = E_{\alpha j}$; the energy loss of alpha-particles in skin is maximum (R_j^{Skin} maximum) (case 3 of Fig.2). For $x_j < R_j, E_{\alpha j}^{Res} < E_{\alpha j}$; the range of alpha-particle in skin are lower than those corresponding to $x_j = 0\mu\text{m}$ (case 2 of Fig.2).

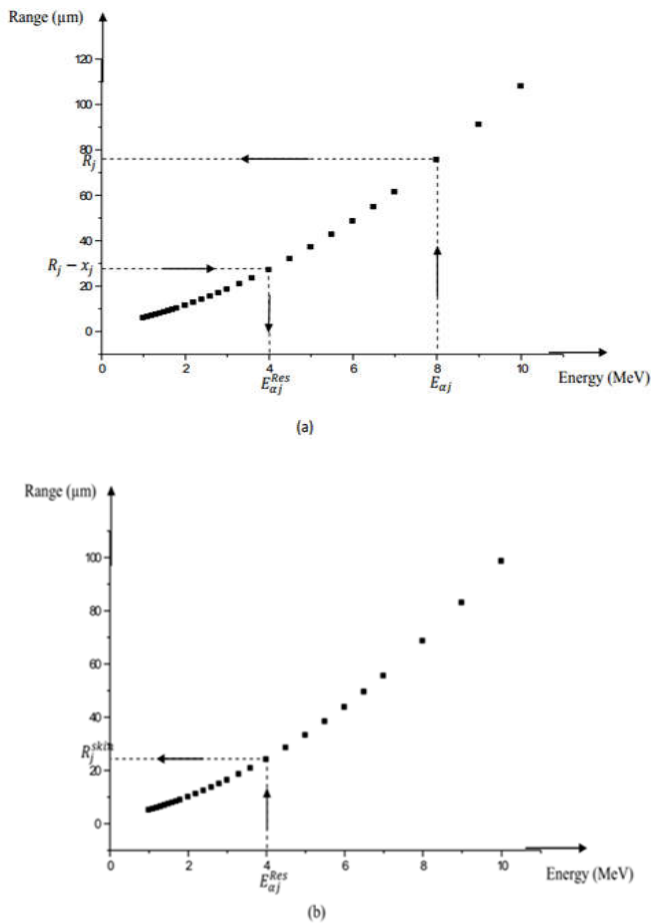


Fig. 3. Alpha particle range-energy relation for a medical drug material sample (a) and skin (b)

Fig. 3. Alpha particle range-energy relation for a medical drug material sample (a) and skin (b). Alpha-equivalent dose rates (Svs⁻¹) to the human skin due to a radionuclide of index j belonging to the ²³⁸U series and a radionuclide of index j' belonging to the ²³²Th series from the application of medical drugs by patients are respectively given by:

$$\dot{H}_{skin}(j)(t) = A_C^{skin}(j)(t) D_{sp}^{skin}(j) W_R \tag{6}$$

$$\dot{H}_{skin}(j')(t) = A_C^{skin}(j')(t) D_{sp}^{skin}(j') W_R \tag{7}$$

Where: $A_C^{skin}(j)(t)$ (Bq) is the alpha-activity, at time t, in skin due to a radionuclide of index j belonging to the ²³⁸U series. $A_C^{skin}(j')(t)$ (Bq) is the alpha-activity, at time t, in skin due to a radionuclide of index j' belonging to the ²³²Th series. $D_{sp}^{skin}(j)$ is the specific alpha-dose (Gy) deposited by 1Bq of a radionuclide of index j belonging to the ²³⁸U series in skin. $D_{sp}^{skin}(j')$ is the specific alpha-dose (Gy) deposited by 1Bq of a radionuclide of index j' belonging to the ²³²Th series in skin. W_R is the radiation weighting factor which is equal to 20 for alpha-particles (International Commission on Radiological Protection, 2007).

The $A_C^{skin}(j)(t)$ and $A_C^{skin}(j')(t)$ alpha-activities are respectively given by:

$$A_C^{skin}(j)(t) = \frac{1}{2} A_C^{Sample} (^{238}U) e^{-\lambda_j t} \times 1cm^3 \tag{8}$$

and

$$A_C^{skin}(j')(t) = \frac{1}{2} A_C^{Sample} (^{232}Th) e^{-\lambda_{j'} t} \times 1cm^3 \tag{9}$$

where $A_C^{Sample}(^{238}U)$ (Bq cm⁻³) is the alpha-activity due to ²³⁸U inside a medical drug sample. $A_C^{Sample}(^{232}Th)$ (Bq cm⁻³) is the alpha-activity due to ²³²Th inside a medical drug sample. λ_j is the radioactive decay constant of a radionuclide of index j belonging to the ²³⁸U series and $\lambda_{j'}$ is the radioactive decay constant of a radionuclide of index j' belonging to the ²³²Th series. The term 1/2 means that only half of the emitted alpha-particles inside a medical drug sample may lose their energies inside the skin.

The $D_{sp}^{skin}(j)$ and $D_{sp}^{skin}(j')$ specific alpha-doses are respectively given by:

$$D_{sp}^{skin}(j) = k \frac{k_j}{d_{skin} S_{skin}} \frac{E_{\alpha j}^{Res}}{R_j^{Skin}} \tag{10}$$

and

$$D_{sp}^{skin}(j') = k \frac{k_{j'}}{d_{skin} S_{skin}} \frac{E_{\alpha j'}^{Res}}{R_{j'}^{Skin}} \tag{11}$$

Where: d_s is the density of skin (g.cm⁻³). S_{skin} is the surface skin (cm²). $k = 1.6.10^{-13}$ (J MeV⁻¹) is a conversion factor. R_j^{skin} is the range, in skin, of the alpha-particle of index j and residual energy $E_{\alpha j}^{Res}$ belonging to the ²³⁸U series. $R_{j'}^{Skin}$ is the range, in skin, of an alpha-particle of index j' and residual energy $E_{\alpha j'}^{Res}$ belonging to the ²³²Th series (Fig.3).

By integrating Eqs.6 and 7, committed equivalent doses (Sv) to skin due to an alpha-particle of residual energy $E_{\alpha j}^{Res}$ emitted by a radionuclide of index j belonging to the ²³⁸U series and an alpha-particle of residual energy $E_{\alpha j'}^{Res}$ emitted by a radionuclide of index j' belonging to the ²³²Th series from the application of a medical drug sample are respectively given by:

$$H_{skin}(j) = \frac{D_{sp}^{skin}(j) W_R}{2\lambda_j} A_C^{Sample} (^{238}U) (1 - e^{-\lambda_j t_a}) \tag{12}$$

$$H_{skin}(j') = \frac{D_{sp}^{skin}(j') W_R}{2\lambda_{j'}} A_C^{Sample} (^{232}Th) (1 - e^{-\lambda_{j'} t_a}) \tag{13}$$

where t_a is the application time.

Committed equivalent doses to the epidermis (EP) of skin (Sv) due all residual energies of an alpha-particle of index j and initial energy $E_{\alpha j}$ belonging to the ²³⁸U series and an alpha-particle of index j' and initial energy $E_{\alpha j'}$ belonging to the ²³²Th series are respectively given by:

$$H(j)(EP) = \frac{k k_j A_c^{Sample} ({}^{238}U)(1 - e^{-\lambda_j t a})}{2 \lambda_j S_{Skin} d_{Skin} E_{\alpha_j}^{Res}} \int_0^{E_{\alpha_j}} \frac{E_{\alpha_j}^{Res}}{R_j^{Skin}(E_{\alpha_j}^{Res})} dE_{\alpha_j}^{Res} \dots (14)$$

and

$$H(j')(EP) = \frac{k k_j A_c^{Sample} ({}^{232}Th)(1 - e^{-\lambda_j t a})}{2 \lambda_j S_{Skin} d_{Skin} E_{\alpha_j}^{Res}} \int_0^{E_{\alpha_j'}} \frac{E_{\alpha_j'}^{Res}}{R_j^{Skin}(E_{\alpha_j'}^{Res})} dE_{\alpha_j'}^{Res} \dots (15)$$

Where $E_{\alpha_j}^{Res}$ and $E_{\alpha_j'}^{Res}$ are the chosen steps.

Committed equivalent doses ($Sv y^{-1} cm^{-2}$) to a surface skin of $1cm^2$ of the epidermis during an exposure time equals to 1 year due to alpha-particles emitted by the ${}^{238}U$ (eight alpha-emitting nuclei) and ${}^{232}Th$ concentrations (seven alpha-emitting nuclei) series from the application of a medical drug sample by patients are respectively given by:

$$H(U)(EP) = \sum_{j=1}^8 H(j)(Tot) \dots (16)$$

and

$$H(T)(EP) = \sum_{j=1}^7 H(j')(Tot) \dots (17)$$

RESULTS AND DISCUSSION

A. ${}^{238}U$ and ${}^{232}Th$ alpha-activities per unit volume in medical drugs

The ${}^{238}U$ ($A_c({}^{238}U)$) and ${}^{232}Th$ ($A_c({}^{232}Th)$) alpha-activities per unit volume were measured in various medical drugs prescribed by doctors for different age groups of patients. Data obtained are shown in Table 3. Since the track detectors utilized were etched in two NaOH solutions at optimal conditions of etching, ensuring good sensitivities of the SSNTDs and a good reproducibility of the registered track density rates determined by means of the same

optical microscope with magnification 40x, only the statistical uncertainty on track counting is predominant. From the statistical uncertainty on track counting the uncertainty on track density production per unit time was determined, and then the uncertainty of the measured ${}^{238}U$ and ${}^{232}Th$ concentrations was determined, which gave values of about 8%. Natural uranium is formed by ${}^{238}U$, ${}^{235}U$ and ${}^{234}U$ radioisotopes with isotopic abundances equal to 99.27%, 0.72% and 0.0055%, respectively. In order to validate this method, 10 medical drugs were analyzed by using Isotope Dilution Mass Spectrometry (IDMS). Isotope Dilution Mass Spectrometry is based on addition of a known amount of enriched isotope (called the spike) to a medical drug sample. After equilibrium of the spike with the natural isotope of the element in the sample, mass spectrometry is used to measure the altered isotopic ratio(s). Data obtained by the two methods, for the ${}^{238}U$ and ${}^{232}Th$ contents, are in good agreement with each other (Table 3)".

So, the contribution of alpha-particles emitted by the ${}^{235}U$ series to the global track densities registered on the SSNTDs utilized is negligible because they induce a relative uncertainty smaller than 1% which is included in the uncertainty on the ${}^{238}U$ and ${}^{232}Th$ concentration determination (8%). The data shown in Table 3 demonstrate that all medical drug samples studied contain more ${}^{238}U$ than ${}^{232}Th$. This is probably due to the fact that raw materials used for the preparation of these medical drugs contain more ${}^{238}U$ than ${}^{232}Th$. It is to be noted that the ${}^{238}U$ contents of the P1, P3, P5, P9, P10, P12, P13, P17, P18 and P20 medical drugs are clearly higher than those of the P2, P4, P6, P7, P11, P14 and P21 medical drug samples (Table 3). We also noted that the ${}^{232}Th$ contents of the P2, P6, P7, P11, P14 and P21 medical drug samples are clearly higher than those of the P1, P3, P9, P13, P17 and P18 samples (Table 3). The minimum detection activities (MDA) for ${}^{238}U$ and ${}^{232}Th$ were found equal to $(0.81 \pm 0.05) mBq l^{-1}$ and $(0.11 \pm 0.01) mBq l^{-1}$, respectively.

Table 1. Description of the studied medical drugs

Medical drugs	Properties	Dosage	Medical specialty
P1	Used for surface anesthesia (skin and mucosa)	1g maximum per 10cm2 during 20 to 30 minutes	Anesthesia
P2	Vascular protective and veinotonic	2 applications per day during 1 month	Cardiology
P3	Dermocorticoid	1 to 2 applications per day during 15 days	Dermatology
P4	Antibacterial agent	1 application per day during 7 to 15 days	Dermatology
P5	Antifungal agent	2 applications per day during 1 month	Dermatology
P6	Antiparasitic agent	2 applications per day during 8 days	Dermatology
P7	Used for antiseptic skin	1 application per day during 7 days	Dermatology
P8	Antiherpetic agent	5 applications per day during 5 to 10 days	Dermatology
P9	Used for antiacne treatment	1 to 2 applications per day during 3 months	Dermatology
P10	Used for antipruritic treatment	2 to 3 applications per day during 3 to 5 days	Dermatology
P11	Used for local treatment of skin ulcers	1 to 2 applications per day during 7 days	Dermatology
P12	Used for the treatment of hemorrhoids	2 to 3 applications per day during 7 days	Gastroenterology
P13	Estrogen agent	1 application per day 24 to 28 days per month during 5 years	Gynecology
P14	Progestin agent	1 application per day during 1 month	Gynecology
P15	Used in adjunctive therapy and as decongestant in respiratory diseases	2 applications per day during 3 days	Pneumology
P16	Non steroidal anti inflammatory	3 to 4 applications per day during 2 weeks	Rheumatology
P17	Used for the treatment of psoriasis	1 to 2 applications per day during 2 months	Dermatology
P18	Keratolytic agent	1 application per day during 3 month	Dermatology
P19	Used for the treatment of hyperpigmented lesions	2 applications per day during 3 month	Dermatology
P20	Used for rosacea treatment	2 applications per day during 3 to 4 months	Dermatology
P21	Used for local treatment of painful muscle contractures	2 applications per day during 2 weeks	Rheumatology

Table 2. Ranges of alpha-particles emitted by the ^{238}U and ^{232}Th series inside skin

Nuclide	$E_{\alpha i}$ (Mev)	K_i	R_i (μm)
(a) Uranium family			
^{238}U	4,19	1	25,64
^{230}Th	4,62	1	29,52
^{234}U	4,77	1	30,94
^{226}Ra	4,78	1	31,03
^{210}Po	5,30	1	36,16
^{222}Rn	5,49	1	38,13
^{218}Po	6,00	1	43,64
^{214}Po	7,68	1	64,20
Nuclide	$E_{\alpha j}$ (Mev)	K_j	R_j (μm)
(b) Thorium family			
^{232}Th	4,01	1	24,09
^{228}Th	5,42	1	37,40
^{224}Ra	5,71	1	40,46
^{212}Bi	6,05	0,36	44,20
^{220}Rn	6,29	1	46,93
^{216}Po	6,78	1	52,73
^{221}Po	8,78	0,64	79,62

Table 3. Data obtained for the ^{238}U and ^{232}Th contents in different medical drug samples

Medical drug samples	ρ_G^{LR} ($10^{-5} \cdot \text{tr} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	ρ_G^{CR} ($10^{-5} \cdot \text{tr} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$)	This method				IDMS	
			C (^{238}U) (ppm)	C (^{232}Th) (ppm)	A_G (^{238}U) (mBq/l)	A_G (^{232}Th) (mBq/l)	C (^{238}U) (ppm)	C (^{232}Th) (ppm)
P1	2.29±0.16	8.74±0.61	0.74±0.05	0.28±0.02	9.10±0.64	1.15±0.08		
P2	1.08±0.09	4.13±0.33	0.35±0.03	0.15±0.01	4.31±0.34	0.62±0.05	0.36±0.01	0.14±0.01
P3	2.33±0.20	8.89±0.8	0.76±0.05	0.28±0.02	9.35±0.71	1.16±0.09		
P4	1.85±0.13	7.05±0.49	0.61±0.04	0.21±0.01	7.50±0.53	0.86±0.06	0.60±0.03	0.20±0.01
P5	2.48±0.2	9.36±0.75	0.81±0.06	0.23±0.01	9.96±0.8	0.95±0.07	0.82±0.04	0.24±0.01
P6	1.29±0.10	4.93±0.37	0.42±0.03	0.17±0.01	5.17±0.39	0.70±0.05		
P7	1.41±0.11	5.39±0.44	0.45±0.03	0.19±0.01	5.54±0.48	0.78±0.06		
P8	2.02±0.14	7.69±0.54	0.67±0.05	0.21±0.01	8.24±0.57	0.86±0.07	0.66±0.04	0.20±0.01
P9	2.24±0.18	8.54±0.71	0.73±0.05	0.26±0.02	8.98±0.76	1.07±0.08	0.74±0.04	0.25±0.01
P10	2.50±0.19	9.51±0.78	0.83±0.06	0.24±0.02	10.21±0.83	0.98±0.08	0.82±0.04	0.25±0.01
P11	1.69±0.12	6.42±0.45	0.56±0.04	0.15±0.01	6.89±0.48	0.62±0.04		
P12	2.17±0.17	8.27±0.66	0.71±0.05	0.24±0.02	8.73±0.70	0.98±0.08		
P13	2.42±0.21	9.24±0.76	0.78±0.06	0.30±0.02	9.60±0.86	1.23±0.10	0.79±0.05	0.32±0.01
P14	1.68±0.12	6.37±0.45	0.57±0.04	0.12±0.01	7.01±0.50	0.49±0.03	0.58±0.03	0.125±0.004
P15	2.07±0.15	7.88±0.63	0.68±0.05	0.21±0.01	8.36±0.67	0.86±0.07	0.67±0.04	0.20±0.01
P16	1.95±0.13	7.42±0.61	0.64±0.04	0.19±0.01	7.87±0.64	0.78±0.06		
P17	2.53±0.18	9.64±0.67	0.84±0.06	0.28±0.02	10.22±0.71	1.15±0.08		
P18	2.58±0.23	9.85±0.89	0.85±0.08	0.32±0.03	10.33±0.87	1.31±0.12		
P19	2.05±0.14	7.80±0.55	0.68±0.05	0.20±0.01	8.36±0.59	0.82±0.06		
P20	2.71±0.18	10.30±0.52	0.90±0.07	0.24±0.02	11.07±0.93	0.98±0.07	0.92±0.04	0.26±0.01
P21	1.22±0.10	4.66±0.37	0.39±0.03	0.16±0.01	4.80±0.38	0.66±0.05		

Table 4 a. Committed equivalent doses to the epidermis of skin ($\text{Sv y}^{-1}\text{cm}^{-2}$) due all residual energies of an alpha-particle of index j and initial energy E_{aj} belonging to the ^{238}U series from cutaneous application of different medical drugs by adult female

Medical drug samples	$\text{H}(^{238}\text{U})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{230}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{234}\text{U})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{226}\text{Ra})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{210}\text{Po})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{222}\text{Rn})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{218}\text{Po})$ ($10^{-9}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{214}\text{Po})$ ($10^{-12}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(\text{U})(\text{EP})$ ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)
P1	1.97	2.2	2.79	2.23	2.43	2.49	3.96	4.20	0.14±0.01
P2	1347	1471	1513	1516	1567	314	1.88	2	77±6
P3	1461	1596	1641	1644	1747	638	4.07	4.32	76±6
P4	1172	1280	1317	1319	1402	512	3.3	3.5	70±6
P5	3113	3400	3497	3503	3620	724	4.34	4.59	179±16
P6	431	471	484	485	522	289	2.25	2.4	27±2
P7	404	441	453	454	490	290	2.41	2.56	25±1
P8	858	937	964	966	1036	503	3.59	3.80	57±4
P9	8421	9196	9459	9476	8782	656	3.91	4.14	460±41
P10	532	581	598	599	648	444	4.45	4.71	34±3
P11	502	549	564	565	610	361	3	3.2	32±3
P12	637	695	715	717	773	458	3.8	4	40±3
P13	33608	36702	37751	37813	23428	701	4.2	4.4	1700±153
P14	2191	2393	2461	2466	2548	510	3	3.23	126±11
P15	2610	2850	2930	2940	3200	652	3.64	3.85	170±15
P16	1230	1343	1381	1384	1471	537	3.43	3.63	73±6
P17	6389	6977	7177	7189	7032	747	4.45	4.72	355±31
P18	9687	10578	10881	10900	10102	757	4.50	4.77	529±47
P19	7839	8561	8806	8821	8175	611	3.64	3.86	428±38
P20	13840	15115	15547	15574	13690	809	4.82	5.11	895±80
P21	750	819	842	844	897	327	2.1	2.2	45±4

Table 4 b. Committed equivalent doses to the epidermis of skin ($\text{Sv y}^{-1}\text{cm}^{-2}$) due all residual energies of an alpha-particle of index j and initial energy E_{aj} belonging to the ^{238}U series from cutaneous application of different medical drugs by adult male

Medical drug samples	$\text{H}(^{238}\text{U})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{230}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{234}\text{U})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{226}\text{Ra})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{210}\text{Po})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{222}\text{Rn})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{218}\text{Po})$ ($10^{-9}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{214}\text{Po})$ ($10^{-15}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(\text{U})(\text{EP})$ ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)
P1	1.72	1.92	2.44	1.94	2.12	2.18	3.46	3.67	0.12±0.01
P2	1177	1285	1322	1324	1368	274	1.64	1.74	68±6
P3	1461	1394	1434	1437	1527	557	3.56	3.70	76±6
P4	1024	1119	1150	1152	1225	447	2.82	3.02	61±5
P5	2720	2970	3055	3061	3163	633	3.79	4.017	156±14
P6	377	411	423	424	465	252	1.96	2.08	23±2
P7	353	385	396	397	428	254	2.11	2.23	22±2
P8	750	819	842	844	905	440	3.14	3.32	46±4
P9	7357	8034	8264	8279	8672	573	3.42	3.62	402±36
P10	465	507	522	523	566	388	3.88	4.12	30±2
P11	439	479	493	494	533	316	2.62	2.78	28±2
P12	556	607	624	626	675	400	3.32	3.52	35±2
P13	29363	32066	32982	33036	20468	613	3.66	3.87	1485±133
P14	1914	2091	2150	2154	2226	455	2.67	2.82	110±9
P15	2280	2490	2560	2570	2790	540	3.18	3.37	150±9
P16	1074	1174	1207	1209	1285	469	3	3.17	64±5
P17	5582	6096	6270	6281	6143	652	3.89	4.12	31±2
P18	8463	9242	9506	9523	8826	659	3.93	4.16	462±41
P19	6849	7480	7693	7707	7143	534	3.18	3.37	374±33
P20	12092	13206	13583	13607	11961	707	4.21	4.46	789±71
P21	655	716	736	738	784	286	1.83	1.94	39±3

Table 5 a. Committed equivalent doses to the epidermis of skin ($\text{Sv y}^{-1}\text{cm}^{-2}$) due all residual energies of an alpha-particle of index j' and initial energy $E_{\alpha j'}$ belonging to the ^{232}Th series from cutaneous application of different medical drugs by adult female

Medical drug samples	$\text{H}(^{232}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{228}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{224}\text{Ra})$ ($10^{-8}\text{Svy}^{-1}\text{cm}^{-2}$)	$\text{H}(^{212}\text{Bi})$ (10^{-9}Sv $\text{y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{220}\text{Rn})$ ($10^{-9}\text{Svy}^{-1}\text{cm}^{-2}$)	$\text{H}(^{216}\text{Po})$ (10^{-12}Sv y^{-1} cm^{-2})	$\text{H}(^{212}\text{Po})$ (10^{-18}Sv $\text{y}^{-1}\text{cm}^{-2}$)	$\text{H}(\text{Th})(\text{EP})$ ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)
P1	1.89	2.47	2.58	8.26	1.24	3.77	6.85	0.080±0.006
P2	1294	1664	1755	13.5	0.6	1.78	3.25	47±4
P3	1403	1818	1908	29.3	1.27	3.9	7.04	45±4
P4	1125	1458	1531	23.5	1.02	3.11	5.65	41±3
P5	2989	3845	4056	31.2	1.35	4.13	7.50	109±9
P6	414	538	563	16.2	0.7	2.14	3.89	15±1
P7	388	505	528	17.4	0.8	2.3	4.17	14±1
P8	824	1071	1122	25.8	1.12	3.4	6.2	30±2
P9	8086	10099	10847	28.1	1.22	3.72	6.76	291±26
P10	511	665	696	32	1.4	4.2	7.69	19±1
P11	482	628	657	21.6	0.9	2.85	5.19	18±1
P12	611	795	832	27.4	1.2	3.6	6.57	22±2
P13	32273	35850	41847	30	1.3	4	7.23	1100±9
P14	2104	2706	2855	22	0.9	2.9	5.28	77±6
P15	251	3270	3420	26	1.1	3.5	6.30	9.0±0.8
P16	1181	1530	1606	24.7	1.07	3.26	5.92	43±3
P17	6135	7775	8288	32	1.4	4.2	7.70	22±1
P18	9302	11617	12509	32	1.4	4.3	7.78	334±30
P19	7528	9402	10123	26.2	1.14	3.5	6.30	271±24
P20	13291	16359	17794	34.7	1.5	4.6	8.34	474±42
P21	720	933	980	15	0.7	2	3.61	26±2

Table 5 b. Committed equivalent doses to the epidermis of skin ($\text{Sv y}^{-1}\text{cm}^{-2}$) due all residual energies of an alpha-particle of index j' and initial energy $E_{\alpha j'}$ belonging to the ^{232}Th series from cutaneous application of different medical drugs by adult male

Medical drug samples	$\text{H}(^{232}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{228}\text{Th})$ ($10^{-8}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{224}\text{Ra})$ ($10^{-8}\text{Svy}^{-1}\text{cm}^{-2}$)	$\text{H}(^{212}\text{Bi})$ ($10^{-9}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{220}\text{Rn})$ ($10^{-9}\text{Sv y}^{-1}\text{cm}^{-2}$)	$\text{H}(^{216}\text{Po})$ (10^{-12}Sv y^{-1} cm^{-2})	$\text{H}(^{212}\text{Po})$ (10^{-18}Sv y^{-1} cm^{-2})	$\text{H}(\text{Th})(\text{EP})$ ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)
P1	1.66	2.16	2.26	7.22	1.08	3.3	5.99	0.070±0.006
P2	1130	1453	1534	11.8	0.5	1.56	2.84	41±3
P3	1226	1588	1667	25.6	1.11	3.39	6.15	45±3
P4	983	1274	1337	20.5	0.89	2.72	4.96	36±3
P5	2612	3359	3544	27.3	1.18	3.61	6.55	95±8
P6	361	470	492	14.2	0.6	1.87	3.40	13±1
P7	339	441	461	15.2	0.66	2	4.17	12±1
P8	720	935	980	22.5	1	2.98	5.42	26±2
P9	7065	8823	9500	24.6	1.07	3.25	5.91	254±22
P10	446	581	608	27.9	1.21	3.7	6.72	16±1
P11	421	548	574	18.9	0.82	2.5	4.53	15±1
P12	534	695	727	23.9	1.04	3.2	5.74	20±1
P13	28196	31321	36561	26.3	1.14	3.5	6.32	961±84
P14	1838	2364	2494	19.2	0.8	2.5	4.61	67±6
P15	2190	2860	2980	25.1	1	3	5.50	81±7
P16	1032	1337	1403	21.5	0.9	2.8	5.18	38±3
P17	5360	6793	7241	28	1.2	3.7	6.67	194±17
P18	8127	10149	10929	28.3	1.22	3.74	6.80	292±26
P19	6577	8214	8845	23	1	3	5.50	236±21
P20	11612	14292	15546	30.3	1.31	4.01	7.28	414±37
P21	629	815	856	13.1	0.6	1.7	3.16	23±2

Table 6. Data obtained for the annual committed equivalent doses to the epidermis of skin due to the ^{238}U (H(U)(EP)) and ^{232}Th (H(Th)(EP)) series from the application of different medical drugs by 15 years children

Medical drug samples	15 years (Female)		15 years (Male)	
	Annual committed equivalent doses ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)			
	H(U)(EP)	H(Th)(EP)	H(U)(EP)	H(Th)(EP)
P1	0.16±0.01	0.0085±0.0007	0.152±0.01	0.0081±0.0007
P4	76±6	44±3	73±6	42±3
P5	191±17	117±10	183±16	112±10
P6	29±2	16±1	28±2	16±1
P9	493±44	311±27	471±42	230±20
P10	37±3	20±1	35±3	19±1
P11	34±3	19±1	33±2	18±1

Table 7. Data obtained for the annual committed equivalent doses to the epidermis of skin due to the ^{238}U (H(U)(EP)) and ^{232}Th (H(Th)(EP)) series from the application of different medical drugs by 10 years and 5 years children

Medical drug samples	10 years		5 years	
	Annual committed equivalent doses ($\mu\text{Sv y}^{-1}\text{cm}^{-2}$)			
	H(U)(EP)	H(Th)(EP)	H(U)(EP)	H(Th)(EP)
P1	0.22±0.01	0.012±0.001	0.32±0.02	0.017±0.001
P4	105±9	61±5	151±13	88±7
P5	270±24	161±14	380±34	232±20
P6	40±3	23±2	58±5	32±3
P10	51±4	28±2	73±6	40±3
P11	47±4	26±2	68±6	38±3

B. Committed equivalent doses to skin due to the radionuclides of the ^{238}U and ^{232}Th series from the application of medical drugs by patients

Committed equivalent doses to the epidermis of skin due to the alpha-emitting nuclei of the ^{238}U (H(U)(EP)) and ^{232}Th (H(Th)(EP)) series from the application of medical drugs by different age groups of patients have been evaluated by means of Eqs.16 and 17, and the results are shown in Tables 4-7. The statistical relative uncertainty of the committed dose determination is 9%. It should be noted that H(U)(EP) and H(Th)(EP) increase with the application time of medical drugs by adults (Tables 1, 4 and 5). It is to be noted from data shown in Tables 4-7 that H(U)(EP) and H(Th)(EP) due to cutaneous application of medical drug P1 are negligible compared to those due to the other medical drugs for adults and children. This is because the application time for medical drug P1, used for surface anesthesia, is shorter than those for the other medical drugs (Table 1). It is to be noted from results shown in Table 4 that committed equivalent doses to the epidermis of skin due to alpha-particles emitted by ^{214}Po (H(^{214}Po)) and ^{218}Po (H(^{218}Po)) are negligible compared to those corresponding to the other alpha-emitters of the ^{238}U series. This is because they have smaller half-lives, $1.6 \cdot 10^{-4}$ s and 3.05 min, respectively than the other radionuclides. Also, one can note that committed equivalent doses to the epidermis due to ^{212}Po (H(^{212}Po)) and ^{216}Po (H(^{216}Po)) are negligible compared to those corresponding to the other alpha-emitters of the ^{232}Th series (Table 5). This is due to the fact that these radionuclides possess smaller half-lives, $3.7 \cdot 10^{-7}$ s and 0.158 s, respectively than the other alpha-emitters of the ^{232}Th series. It is to be noted that total committed equivalent doses due to ^{238}U and ^{232}Th series from cutaneous application of P1, P4, P5, P6, P10 and P11 medical drugs are higher for 5 years children than for the other age groups of patients (Tables 4-7). This is because 5 years children possess smaller surface skin than the other age

groups of patients (International Commission on Radiological Protection, 1990). The maximum total committed equivalent dose to skin due to the ^{238}U and ^{232}Th series was found equal to $2.8 \text{ mSv y}^{-1} \text{ cm}^{-2}$, obtained for women applying the P13 medical drug (Tables 4(a) and 5(a)), which is significantly smaller than the dose limit for the members of the public to the skin which is of $50 \text{ mSv y}^{-1} \text{ cm}^{-2}$ (International Commission on Radiological Protection, 1990).

Conclusion

In this study, it has been shown that the use of CR-39 and LR-115 type II solid state nuclear track detectors (SSNTDs) allows evaluation of ^{238}U and ^{232}Th alpha-activities per unit volume in various medical drug samples. A new dosimetric model was developed for evaluating radiation doses to skin due to the alpha-emitting nuclei of the ^{238}U and ^{232}Th series from the application of medical drugs by patients. The committed equivalent doses to the epidermis of skin due to the alpha-emitting nuclei of the ^{238}U and ^{232}Th series increase with the application time of medical drugs. It has been shown that only nine alpha-emitting nuclei belonging to the ^{238}U and ^{232}Th series significantly contribute to the global radiation dose to the epidermis of skin from the application of medical drugs by patients. It has also been shown that committed effective dose due to the ^{238}U and ^{232}Th series increases when the surface skin of patients decreases. Thus there is no radiation risk to the epidermis from cutaneous application of the studied medical drugs by patients.

The SSNTD's method used has the advantage of being inexpensive, accurate, and sensitive and does not need the use of standard sources for its calibration. It is a useful tool for measuring ^{238}U and ^{232}Th concentrations in medical drugs as well as essential oils extracted from aromatic and medicinal plants.

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