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Activity measurement and effective dose modelling of natural radionuclides in building material

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HIGHLIGHTS

- Dose models for indoor radiation exposure due to natural radionuclides in building materials.
- Strategies and methods in radionuclide metrology, activity measurement and dose modelling.
- Selection of appropriate parameters in radiation protection standards for building materials.
- Scientific-based limitations of indoor exposure due to natural radionuclides in building materials.

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ABSTRACT

In this paper the assessment of natural radionuclides' activity concentration in building materials, calibration requirements and related indoor exposure dose models is presented. Particular attention is turned to specific improvements in low-level gamma-ray spectrometry to determine the activity concentration of necessary natural radionuclides in building materials with adequate measurement uncertainties. Different approaches for the modelling of the effective dose indoor due to external radiation resulted from natural radionuclides in building material and results of actual building material assessments are shown.

1. Introduction

In many countries limits for the excess effective dose due to the content of natural radionuclides in building materials and construction products are recommended or regulated. Generally natural radioactivity in building material originates from natural radionuclides of the decay chains ^{238}U and ^{232}Th as well as from the primordial radionuclide ^{40}K (e.g. EC 122, 1999b; Trevisi et al., 2012). External radiation exposure by gamma radiation and beta particle emissions plus internal exposure due to inhalation of ^{222}Rn and ^{220}Rn progenies cause chronic indoor exposure of the public. Due to increasing indoor habitation of persons – in average about 80% of live-time persons stay indoor – external and internal exposures caused by building materials are of increasing importance. Additionally the production of building materials using industrial by-products and residuals from natural occurring radioactive material (NORM) industry processing necessitate the consideration and regulation of this issue in regard to radiation protection of the public (e.g. SÚJB, 2009; STUK, 2010; EC, 2011a).

In Fig. 1, the average effective dose partitions per year exposed to a person of the Austrian population is shown (Maringer et al., 2012). In Austria, the average effective dose due to all natural and artificial radiation exposures including indoor radon inhalation is about 4.6 mSv yr^{-1} in 2010 (Fig. 1). The regional maximum value of the external radiation exposure in Austria is about 1.3 mSv yr^{-1} due to the crystalline/granitic geology in the northern part of Austria (Bohemian Massif).

According to EC (1999a): p. 36, about 90% and more of external radiation is absorbed indoor by 20 cm concrete walls, floors and ceilings. Allowing (in the sense of radiation protection) this 90%-shielded natural outdoor base-line exposure to be (virtually) filled indoor by the exposure caused by building materials leads to a total exposure limit of 2.2 mSv yr^{-1} caused indoor by building materials in Austria. This 2.2 mSv yr^{-1} effective dose means about 1.2 mSv yr^{-1} natural outdoor external base-line exposure plus additional permitted 1.0 mSv yr^{-1} external indoor exposure from building materials.

In the proposal for a revised European Council Directive (EC, 1996) laying down the basic safety standards for protection

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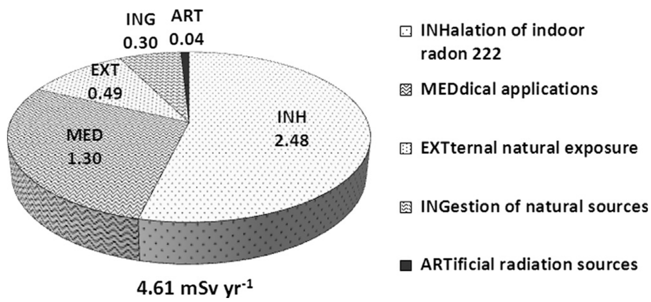

Fig. 1. Contributions to the personal annual mean effective dose in Austrian in 2010 (Maringer et al., 2012).

against the danger arising from exposure to ionising radiation, published in November 2011 (EC, 2011a, 2011b; revised version June 2012: CEU, 2012), the excess effective dose from natural radionuclides in building material to the population is limited to 1 mSv yr^{-1}. This limitation is based on the radiation protection general principles given in ICRP (2007). To comply with this dose limit, annex VII of the directive proposal gives a dose related sum criteria considering the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th. In addition to this proposal, CEN (European Committee for Standardization) technical committee 351, working group 3, prepares radiometrical standard methods for the measurement of natural radionuclides in building materials and dose modelling.

2. Radiometric assessment of natural radionuclides in building materials

Currently, a new standard radiometric method for the determination of the relevant natural radionuclides in building materials is under preparation by task group 3.1 of the CEN/TC 351 working group 3 “Construction products: Assessment of release of dangerous substances – Radiation from construction products”.

To determine the activity concentrations of the relevant key radionuclides ^{226}Ra and ^{232}Th of the natural ^{238}U and ^{232}Th decay chains, the use of the following gamma energy peak scheme is strongly recommended assured by a long-term experimental experience with regard to sufficient discrimination of gamma energy peaks:

- Determination of the activity concentration of ^{226}Ra via gamma energy peaks ^{214}Pb: 351.9 keV and ^{214}Bi: 609.3 keV and 1120.3 keV.
- Calculation of the activity concentration of ^{232}Th via ^{228}Ra and ^{228}Th:

$$a(^{232}\mathrm{Th}) = \frac{a(^{228}\mathrm{Ra}) + a(^{228}\mathrm{Th})}{2} \quad (1)$$

- Determination of the activity concentration of ^{228}Ra via gamma energy peaks ^{228}Ac: 338.4 keV and 911.1 keV.
- Determination of the activity concentration of ^{228}Th via gamma energy peaks ^{208}Tl: 583.1 keV and 727.1 keV.

Activity concentration values $a(\mathrm{RN}_{ij})$ evaluated from individual gamma peaks j of relevant decay chain radionuclides (e.g. ^{214}Pb, ^{214}Bi; ^{208}Tl, and ^{228}Ac) by gamma-ray spectrometry should be combined to the calculated weighted mean of the activity concentration $\overline{a}(\mathrm{RN}_i)$ of the radionuclide i of interest (^{226}Ra, ^{228}Ra, ^{228}Th, and ^{232}Th) using the following:

$$\overline{a}(\mathrm{RN}_i) = \left(\frac{1}{\sum_j (1/u(\mathrm{RN}_{ij}))^2}\right) \sum_j \left(\frac{1}{u(\mathrm{RN}_{ij})}\right)^2 a(\mathrm{RN}_{ij}) \quad (2)$$

where $u(\mathrm{RN}_{ij})$ is the uncertainty of the activity concentration $a(\mathrm{RN}_{ij})$ and $(1/u(\mathrm{RN}_{ij}))^2$ is the weight of the activity concentration of the radionuclide j for the calculation of the mean activity concentration of the radionuclide i.

The evaluation of the measurement uncertainties of the activity concentration values is based on ÖNORM S 5250-1 (2002), ÖNORM S 5250-2 (2005) and ISO 11929 (2010).

An important premise when using activity concentrations values of ^{222}Rn progenies (^{214}Pb, and ^{214}Bi) to determine the activity concentration of ^{226}Ra, is the radon tightness of the building material sample container used in gamma-ray spectrometry.

General pre-condition for the recommended gamma energy peak evaluation protocol is radioactive equilibrium for the analysed building material sample inside both natural decay chains. This assumption is generally true for natural mineralogical building materials of natural origin but could be wrong for NORM industry by-products and residues. For such suspicious materials a more sophisticated radiometric analysis (e.g. alpha spectrometry; ICP-MS; additional analysis after some weeks/months) are appropriate.

In Table 1 an example of a building material sample analysed by gamma-ray spectrometry is given. The additional radionuclides ^{137}Cs, ^{138}La, ^{223}Ra, ^{227}Th and ^{235}U had been analysed because of possible enrichment/contamination of the material at NORM industry processing.

3. Existing and notified regulation in Austria

The limits in the Austrian natural radiation sources ordinance BMLFUW (2008) and the Austrian standard for radioactivity in building materials ÖNORM S 5200 (2009) are based on the average natural radiation exposure outdoor and the mean activity concentrations in building materials in Austria. Due to the increasing consideration of the exposure due to exhalation of ^{222}Rn from building materials, the dose caused by inhalation of ^{222}Rn progenies is strictly considered in the Austrian building material standard (ÖNORM S 5200, 2009). The dose contribution of ^{220}Rn progenies (especially if not in equilibrium with ^{220}Rn) is under discussion and will be probably considered in a future revision of the Austrian building material standard.

The specific application parameters relative ^{220}Rn emanation coefficient (ε) and actual thickness (d) of the building material in the constructive building element are considered in the dose assessment of the Austrian standard.

The radioactivity concentrations requirement for technologically enhanced NORM (TENORM) used in (or as) building materials according to the Austrian NORM Ordinance BMLFUW (2008) is given as follows:

$$I = \frac{a_{\mathrm{Ra-226}} - 40\ \mathrm{Bq\ kg^{-1}}}{40\ \mathrm{Bq\ kg^{-1}}} + \frac{a_{\mathrm{Th-232}} - 25\ \mathrm{Bq\ kg^{-1}}}{240\ \mathrm{Bq\ kg^{-1}}} + \frac{a_{\mathrm{K-40}} - 370\ \mathrm{Bq\ kg^{-1}}}{4000\ \mathrm{Bq\ kg^{-1}}} \quad (3)$$

where $a_{\mathrm{Ra\text{-}226}}$, $a_{\mathrm{Th\text{-}232}}$ and $a_{\mathrm{K\text{-}40}}$ are the activity concentration values in Bq kg^{-1} of the corresponding radionuclides in the building material.

The relevant Austrian regulations which are notified at the European Union are the “Amendments to the building materials list ÖE” of 15. 12. 2002 for the 2nd edition of the “Building materials list ÖE” (OIB, 2004) and the “Amendments to the existing building materials List ÖE” of December, 1st 2004 enacting the third edition of the “Building materials list ÖE” (OIB, 2006). The relevant Austrian standard ÖNORM S 5200 (2009) introduces a more sophisticated formula for the activity concentration index I. This requirement also takes the relative radon emanation factor ε, the building material density ρ and the thickness d for the material