II/2

^{226}Ra AND ^{228}Ra CONCENTRATIONS IN GYPSUM PLASTERS AND MORTARS USED IN THE NETHERLANDS

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Abstract
A survey has been performed on the natural radioactivity concentrations in gypsum plasters and mortars available in the Netherlands, employing gamma-ray spectrometry. The study encompassed a total of 24 materials from several manufacturers, including products for the non-professional sector. Of some of the materials a new batch was collected and analysed about 12 months after the first analysis, to investigate possible changes in material composition over time. The differences between both series of data were less than the statistical uncertainties and no variations over time could be demonstrated.
The results on specific activity are expressed as ^{226}Ra en ^{228}Ra activities per unit of mass. Three classes of material can be distinguished: class I materials of low specific activity, e.g. natural gypsum; class II products with intermediate levels (100-150 Bq/kg of both ^{226}Ra and ^{228}Ra); and class III products with levels up to 200-250 Bq/kg per radionuclide. The results will be discussed in view of public (inhabitants) and occupational (stucco-workers) exposure.

Introduction
The most important primordial radionuclides are ^{40}K, ^{238}U and ^{232}Th, all having half-lives in excess of several billions of years. ^{238}U and ^{232}Th head natural decay series of 14 and 11 significant radionuclides, respectively, also contributing to natural radiation exposure. The occurrence of these nuclides in building material will enhance the indoor ^{222}Rn and ^{220}Rn concentrations and thus the internal radiation dose due to inhalation of their short-lived decay products. In addition, the primordial radionuclides contribute to the external radiation dose of inhabitants.
The activity concentration of ^{226}Ra and ^{232}Th per unit of mass in building materials varies considerably, depending on both the nature and the origin of the compounds. In the Netherlands, average ^{226}Ra and ^{232}Th specific activity levels in construction materials such as sand-lime brick, concrete and brick are about 10, 20 and 40 Bq/kg, respectively (Ackers et al., 1985; Vaas et al., 1993). The specific activities of natural gypsum are about 10 Bq/kg ^{226}Ra and ^{232}Th or less (Ingersoll, 1983; Ackers et al., 1985; Keller and Schmier, 1988). However, in building products based on phosphogypsum, a by-product of the processing of phosphate fertilisers, levels equal to or in excess of 400 Bq ^{226}Ra per kg are reported (Van Dijk and De Jong, 1991; Othman and Mahrouka, 1994; O' Brien et al., 1995; Rutherford et al., 1995). Thus far only limited attention has been paid to the natural radionuclides in gypsum plasters and mortars (Malanca et al., 1993; Van Deynse et al., 1997). In the present study, the specific activities of ^{226}Ra and ^{228}Ra has been determined for almost all ready mixed plasters and mortars available on the Dutch market. The consequences for doses to both inhabitants and plaster-workers are discussed.

Materials and methods
The professional plasters and flooring mortars were obtained from several suppliers of building materials from different parts of the country. The non-professional products were purchased at local shops. All samples were obtained in their original packing. The sampled materials cover the total market for ready mixed plaster in the Netherlands almost completely. For comparison purposes, some plaster materials were also ordered in Germany.
About 600-1000 grams of each material was weighed into polyethylene bottles and encapsulated. After a waiting time of at least three weeks to establish radioactive equilibrium within the decay series, the samples were analysed by gamma-ray spectrometry for 50,000 s using a Ge detector (EG&G). The recorded spectra were analysed using the Genie-PC software (Canberra). By an extension to this program a correction was made for the self-absorption within the sample material. The ^{226}Ra content was calculated from the ^{214}Pb and ^{214}Bi concentrations. The ^{232}Th

activity was based on the measured activities of the nuclides ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl. The standard deviation, based on counting statistics, is less than 5% for specific activities in excess of 100 Bq/kg. For specific activities at the 10 Bq/kg level or less, the standard deviation ranges from 10 to 40%.

Results

The results of the professional products are shown in Table 1. The alphabetical codes A to E represent the five manufacturers; the successive numbers indicate the different products. Some of these products were sampled again 12 months later, to investigate possible changes in composition over time. These results are also included in Table 1. In general, the differences between both series of data are less than 10% and no significant differences could be demonstrated.
In addition to gypsum for stucco-work, some calcium sulphate mortars, which are used as a screed, were incorporated in this study. The results for these latter samples (codes F1 and G1) indicate low radium levels, similar to those in natural gypsum.
Three products (viz. A1, A3 and A9) were also obtained from Germany. The results on specific activity are presented in Table 2. The samples with the codes A1(G) and A9(G) show low radium levels, contrary to the 'same' products available in the Netherlands (Table 1). The A3-coded samples show similar (low) levels. Some products are also available in retail packages, meant for skilful do-it-yourself enthusiasts. The results of the gamma-ray analysis of these latter samples are shown in Table 3. If these are compared with the similar products for professional use (Table 1), then striking differences are observed for the samples coded A4, A8 and A9.

Table 1: **Specific activities of professional gypsum plasters, obtained in the Netherlands**

Sample code	Description	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)
		1st sampling		2nd sampling	
A1	Single-coat gypsum plaster	240	240	240	230
A2	Plaster for floated surface	120	110	130	120
A3	Plaster for mechanical application	6	< 7	7	1
A4	Gypsum plaster	240	210	260	220
A5	Bonding and finish plaster	230	180		
A6	Plaster for floated surface	120	100	150	130
A7	Joint filler	< 3	< 3		
A8	Bond plaster for floated surface	130	130	130	110
A9	Gypsum bonding plaster	220	210	250	200
A10	Joint filler	7	2		
B1	Gypsum plaster	13	4	2	1
B2	Moulding gypsum	10	2		
B3	Bonding and finish plaster	5	< 3		
B4	Plaster for mechanical application	11	4		
C1	Moulding gypsum	< 3	< 3	3	3
D1	Bonding plaster	220	240		
D2	Plaster for floated surface	120	110		
D3	Plaster for mechanical application	5	< 4		
E1	Plaster for mechanical application	3	< 3		
E2	Plaster for mechanical application	2	3		
E3	Plaster for mechanical application	< 3	3		
F1	Calcium sulphate screed	5	2		
G1	Calcium sulphate screed	5	< 3		

Table 2: **Specific activities of professional gypsum plasters obtained in Germany**

Sample code	Description	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)
A1 (G)	Single coat gypsum plaster	3	< 4
A3 (G)	Plaster for mechanical application	5	< 3
A9 (G)	Gypsum bonding plaster	10	< 4

Discussion

Based upon the measurement results, the gypsum products can be subdivided into three categories:

- Materials of low specific activity;
- Materials having intermediate levels (100-150 Bq/kg of both ^{226}Ra and ^{228}Ra);
- Materials with high activity mass concentrations of the parent radionuclides ranging from 200 up to 250 Bq/kg.

Also, in earlier studies the rather broad range in specific activities have been mentioned (Ackers et al., 1985; Van Deynse et al., 1997). An activity mass concentration around 10 Bq/kg or less corresponds to those found in natural gypsum. The mass concentration found in the other categories might indicate the use of phosphogypsum. Some of the materials contain almost pure gypsum, while others are mixed with large amounts of sand to create a certain surface structure. This might explain the levels in the intermediate category. The higher specific activities may have consequences for two distinct classes of individuals, namely inhabitants and stucco-workers. These will be discussed separately.

Table 3: **Specific activities of non-professional gypsum plasters obtained in the Netherlands**

Sample code	Description	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)
A4 (S)	Gypsum plaster	7	2
A8 (S)	Bond plaster for floated surface	9	3
A9 (S)	Gypsum bonding plaster	5	3
A10 (S)	Joint filler	4	< 3
A11 (S)	Repair product (for holes and cracks)	11	2

The indoor ^{222}Rn concentration in the Netherlands is rather low, with arithmetic and geometric means of 23 and 18 Bq/m^3, respectively. In a recent survey the building materials are indicated as the main source of indoor ^{222}Rn (Stoop et al., 1998). Application of plasters with a 20-25 higher activity mass concentration would therefore increase the ^{222}Rn-source term in homes. However, in view of the thin layers of plasters in practice (i.e. 1-10 mm) compared to the total building mass of a house, it is unlikely that this material will seriously affect either the indoor ^{222}Rn concentration or the gamma dose rate (O'Brien et al., 1995; O'Brien, 1997).

Until now, only limited attention has been paid to the radon isotope from the thorium series, ^{220}Rn. Because of its short half-life of 55 s, it was thought that only building materials and infiltration of outside air accounted for most of the indoor ^{220}Rn. However, recent studies have shown that in certain houses, soil can be the predominant source (Li et al., 1992). Since in the Netherlands most ^{222}Rn originates from the building materials, it might be thought that this applies also to ^{220}Rn. As the diffusion length of ^{220}Rn in building material is in the order of 10 mm, only the surface layer of these materials is involved, which for most inner walls and ceilings mean the gypsum plaster decoration. Therefore, application of plaster layers high in ^{228}Ra may play a prominent role with regard to indoor ^{220}Rn. The dose consequences are, however, hard to evaluate, due to a lack of measurement data on ^{220}Rn and its short-lived decay products in the Netherlands. On the assumption that the base level of the equilibrium equivalent concentration (EEC) is 0.3 Bq/m^3 and the dose coefficient is 32 nSv per Bq.h/m^3 (UNSCEAR, 1993), it is calculated that an elevation of 5-10 times in the ^{220}Rn concentration results in an annual effective dose comparable to that of

^{222}Rn and progeny. Whether plasters high in ^{226}Ra can cause such an effect, taking into account the retarding action of coatings and wallpaper, has to be verified in practice.

For flooring and stucco-workers the main source of exposure is the inhalation of dust particles during mixing and processing. Besides the radionuclide contents of the material, the individual working practice plays a prominent part in the resulting radiation exposure. To investigate whether radiological precautions may be required, some dose assessments have been made. The assumptions used in the assessment were continuous exposure conditions according to ICRP (1994), no respiratory protection, activity concentrations of 250 Bq/kg of the parents and daughter radionuclides, 5 μm AMAD and the EC dose coefficients (Euratom, 1996). Per time-weighed average airborne dust concentration of 1 mg/m^3, an effective dose of about 20 μSv in a year can be derived, suggesting that the radiation exposure remains in all cases well below the prevailing dose limit of 1 mSv/y (Euratom, 1996). Presently, we are engaged in an investigation concerning the annual body burden of airborne particles to stucco-workers using personal air sampling equipment, from which data the radiation exposure level can be determined more accurately.

Acknowledgement—The authors acknowledge the financial support of the Dutch Marketing Board for the Stucco, Terrazzo and Flooring Workers (BSTS).

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