NORM in Italian tiles and refractories industries

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Abstract. In recent years, the Regional Agency for the Environmental Protection of Veneto (ARPAV) has carried out surveys in refractory and tile industries, investigating, in particular, the natural radioactivity levels of residues and dusts. Elevated activity concentrations have been recorded for ^{210}Pb and ^{210}Po (21 000 and 35 000 Bq/kg, respectively) in dust generated in a refractory fusion furnace. In addition, high values of ^{210}Po (up to 46 000 Bq/kg) have occurred in the hydrated lime used for trapping dust in the tile firing process. Doses to the public as a result of airborne dust emissions from these two processes are estimated using a comprehensive prediction model (PC Cream) for refractories and a simplified model for tiles. In both cases, individual effective doses appear to be less than 1 μSv/a while collective doses are slightly more than 1 man·mSv/a. The PC Cream simulation data were obtained in the course of collaboration between ARPAV and the Catholic University of Brescia.

1. Introduction

Zircon sand is known to exhibit non-negligible levels of natural radioactivity. Zircon sand and zircon flour (a product of sand milling) are used in the manufacture of refractory materials and in the tile industry, both sectors being of considerable importance for the Italian economy. Depending on the process, radionuclides of natural origin occur in products, by-products, residues and wastes [1].

2. Tile industry

2.1. General manufacturing aspects

Italy is one of the world's main tile producers, supplying 13% of the total world market. Several hundred factories form this sector, 80% of which are located in the ceramic tile industry zone in the provinces of Modena and Reggio Emilia [2]. Zircon flour and, on occasion, zircon sand are used in the glaze applied to the tiles and sometimes in the tile mixture itself (the 'biscuit'), as is also the case for white porcelain stoneware. The standard production process involves firing at 1000–1200°C for both unglazed tiles (using a single or double firing process) and glazed tiles. To precipitate dusts released during the firing process, lime is commonly used — this becomes hydrated lime after the dust extraction process. Radionuclides of natural origin are contained in non-negligible amounts in zircon sand and flour and, apart from ^{210}Po, are found diluted in the finished products. The ^{210}Po volatilizes in the firing process and concentrates in dusts, some of which escape to the atmosphere and some of which are precipitated in the hydrated lime.

2.2. Survey data and estimated dose

In 2005, ARPAV carried out a survey in some relevant tile manufacturing factories to assess the activity concentrations in residues and dusts for the complete process [3]. The highest activity concentrations found in hydrated lime were used as input data for estimating the collective effective dose[1] received by the population living near the plant and attributable to the emission of dusts from the firing process stack. The estimation was based on the simplified model given in Annex C (para. 1B) of the 1982 UNSCEAR Report [4]. The model considers two stages of exposure: (1) direct inhalation during the passage of the cloud and (2) intakes (ingestion of contaminated food and inhalation of resuspended material) and external radiation after deposition. The activity released from the stack was calculated by multiplying the dust activity concentrations (assumed to be the same as those in the

[1] All doses mentioned in this paper are committed doses.

hydrated lime) by the dust emitted per year through the stack (261 kg/a)[2]. The dust activity concentrations in the hydrated lime and the estimated radionuclide activities released from the stack are shown in Table 1.

TABLE 1. STACK EMISSIONS FROM A TILE MANUFACTURING PLANT

Radionuclide	Maximum activity concentration in the hydrated lime(Bq/kg)		Estimated activity released from the stack (Bq/a)
	Value	Uncertainty[a]	
^{238}U	9	21%	2349
^{226}Ra	10	7%	2610
^{210}Pb	425	4%	110 925
^{210}Po	46 100	6%	12 032 100
^{235}U	—	—	235
^{232}Th	3	9%	783
^{40}K	369	5%	96 309

[a] 68% confidence interval.

In the first stage of exposure considered in the model, the inhalation dose for the population living in the plant area was calculated from the time integrated activity concentrations in that area arising from the whole radionuclide release. For ^{222}Rn, the dose added by the plant release was calculated by comparison with the inhalation dose due to natural emanation from soil: all radon contained in the processed zircon sand and flour (2976 t/a) was assumed to be released into the air. Some parameters were updated with respect to the 1982 UNSCEAR Report — the age-weighted inhalation effective dose coefficients and breathing rates were taken from Annex B (Table 17 and para.153) of the 2000 UNSCEAR Report [5].

In the second stage of exposure considered in the model, the dose arising from the deposited activity was calculated by comparison with doses resulting from the natural presence of radionuclides in soil. Apart from some fixed parameters [4], the key data used in the calculation were taken from Ref. [5] as follows:

– Natural soil concentrations of 35, 30 and 400 Bq/kg for radionuclides in the ^{238}U and ^{232}Th decay chains and for ^{40}K, respectively;
– Age-weighted effective doses for inhalation and ingestion of various naturally occurring radionuclides as specified in Annex B (Tables 17, 18) of Ref. [5]:
– Age-weighted external radiation dose rates due to the natural levels in soil of the radionuclides in the ^{238}U and ^{232}Th decay chains and of ^{40}K as specified in Annex B (Table 6) of Ref. [5];
– An individual effective dose of 470 μSv/a for inhalation of natural levels of ^{222}Rn.

The results of the model estimates are shown in Table 2 and Fig. 1. The total collective effective dose arising from the plant emissions was about 1.4 man·mSv in a year. Taking the ratio of the collective dose to the population living in the local municipality gave an effective dose per caput of less than 0.1 μSv/a. This is negligible compared with the action level of 300 μSv/a indicated by Italian law (Legislative Decree No. 241/00). The most significant exposure pathway was the ingestion of contaminated food after deposition. The radionuclides that contributed most were ^{210}Po followed by ^{222}Rn (see Fig. 1). When site and plant technical data become available, a more accurate prediction model will be used, as has been described later in the paper for the refractory industry.

[2] The use of hydrated lime data instead of dust specific data could lead to an underestimate of the dust activity concentration, and consequently of the dose, due to the addition of the lime matrix.

TABLE 2. COLLECTIVE DOSE ARISING FROM EMISSIONS FROM A TILE MANUFACTURING PLANT

Radionuclide	Collective effective dose in a year (man·Sv)				
	Passage of the cloud	Deposition			Total
		Ingestion	Inhalation	External radiation	
U-238	2.75E-08	5.70E-10	4.79E-11	2.33E-07	8.65E-08
U-234	3.31E-08	6.39E-10	5.93E-11		9.22E-08
Th-230	1.24E-07	1.32E-09	1.10E-10		1.84E-07
Ra-226	3.70E-08	2.03E-08	6.59E-11		1.16E-07
Rn-222	2.19E-04		1.19E-06		2.20E-04
Pb-210	4.92E-07	3.02E-06	4.31E-07		3.94E-06
Po-210	1.61E-04	9.94E-04	1.40E-05		1.17E-03
Th-232	7.17E-08	3.19E-10	7.45E-11	1.02E-07	1.06E-07
Ra-228	9.02E-09	1.86E-08	1.86E-11		6.17E-08
Th-228	1.26E-07	2.22E-10	2.57E-10		1.61E-07
Rn-220			3.11E-08		3.11E-08
U-235	2.95E-09	5.46E-11	4.96E-12		3.01E-09
K-40				8.90E-07	8.90E-07
Total	3.81E-04	9.97E-04	1.57E-05	1.23E-06	1.39E-03

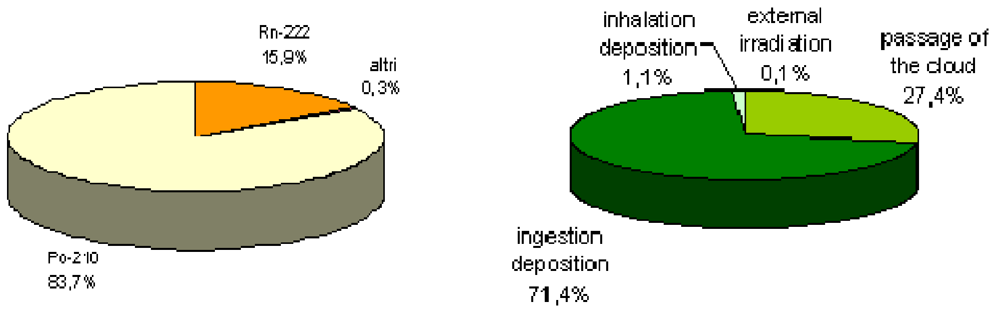

FIG. 1. Contributions of radionuclides and exposure pathways to the collective dose

3. Refractory industry

3.1. General manufacturing aspects

The Italian refractory industry comprises about 40 companies, several of which use zircon sand or other zircon-based components as raw materials [2]. In the standard process, this material is melted in an electric furnace with alumina and silica, after preparation, and the resulting moulded components are sent for finishing or, if not to standard, recycled to the manufacturing process. Radionuclides of natural origin are contained in non-negligible amounts in the raw materials and, apart from ^{210}Pb and ^{210}Po, are found diluted in the finished products. The ^{210}Pb and ^{210}Po volatilize in the fusion process and concentrate in the furnace dust.

3.2. Survey data and estimated dose

A few years ago, ARPAV carried out a radiological survey at the premises of a company in northern Italy (one of the main users of zircon sand). The activity concentrations measured in dust from the fusion stack (Stack 1) and the scraps grinding stack (Stack 2), as reported in Ref. [6], were used as input data for a dose modelling assessment to determine the impact of dust emission into the atmosphere.

To estimate the doses delivered to the population as a consequence of the release of radioactivity from the stacks into air, the simulation code PC Cream developed by the UK Health Protection Agency was used. The code is based on the methodology described in Ref. [7]. The exposure pathways considered were as follows:

– Inhalation of radionuclides in the plume;

– External radiation from radionuclides in the plume;

– External radiation from radionuclides deposited on the ground

– Inhalation of radionuclides resuspended from ground deposits;

– Ingestion of food grown on contaminated land.

The model assumes continuous release conditions and a Gaussian dispersion of the plume. The activity concentrations measured in the two stacks were used as input data, along with the total amount of emitted dust per year, namely 191 and 198 kg for Stacks 1 and 2, respectively. It was assumed that all the ^{222}Rn in the processed zircon sand was released into the air. The dust activity concentrations and estimated activity releases are shown in Table 3. Taking into account the continued vertical rise of the plume due to initial buoyancy and momentum of the flue gases, the effective stack release height was taken to be 70 m. Local meteorological data, averaged over a year, were used as inputs to the model. Individual effective doses were calculated for the critical group living close to the factory in the prevailing wind direction. Individuals in the critical group were assumed to belong to three different age groups (infants, children and adults), to stay indoors 80% of the time and to eat only food of local origin according to the standard diet of north-eastern Italy [8]. The collective effective dose was calculated for the whole EU population.

TABLE 3. STACK EMISSIONS FROM A REFRACTORY PLANT

Radionuclide	Dust activity concentration (Bq/kg)				Estimated activity released from the stack (Bq/a)	
	Stack 1		Stack 2			
	Value	Uncertainty[a]	Value	Uncertainty[a]	Stack 1	Stack 2
^{238}U	358.5	18%	1191	22%	6.86E+04	2.36E+05
^{226}Ra	147	6%	1046	6%	2.81E+04	2.07E+05
^{210}Pb	21 050	19%	1033	4%	4.02E+06	2.05E+05
^{210}Po	35 000	5.7%	1239	12%	6.69E+06	2.56E+05
^{235}U	20	10%	59	9%	3.82E+03	1.17E+05
^{232}Th	27	6%	178	6%	5.16E+03	3.52E+04
^{40}K	10	17%	255	6%	1.91E+03	5.05E+04

[a] 68% confidence interval.

The individual effective doses calculated for the worst-case exposure scenario (a factory at the most critical site operating for a period of 50 y) are shown in Table 4. The doses are less than 0.1 μSv/a — far below the action level of 300 μSv/a indicated by Italian law (Legislative Decree No. 241/00). For all three age groups, the radionuclides contributing most to the dose were ^{210}Po, ^{210}Pb and ^{222}Rn, and the most significant exposure pathway was the ingestion of contaminated food. Details for the adult age group are shown Fig. 3.

The collective effective dose was determined to be 3 man·mSv in a year. This value is close to the estimate of 1.2 man·mSv reported in Ref. [6], which was determined using the simplified model reported by UNSCEAR [4, 5] as used for the tile industry dose assessment described earlier in the paper. Again, the most significant radionuclides were ^{210}Po, ^{210}Pb and ^{222}Rn, while the main exposure pathway was the direct inhalation of radionuclides in the plume.

TABLE 6. DOSES TO INDIVIDUALS IN THE CRITICAL GROUP, ARISING FROM EMISSIONS FROM A REFRACTORY PLANT

Age group	Annual effective dose (μSv)
Infant	7.8E-02
Child	4.5E-02
Adult	2.4E-02

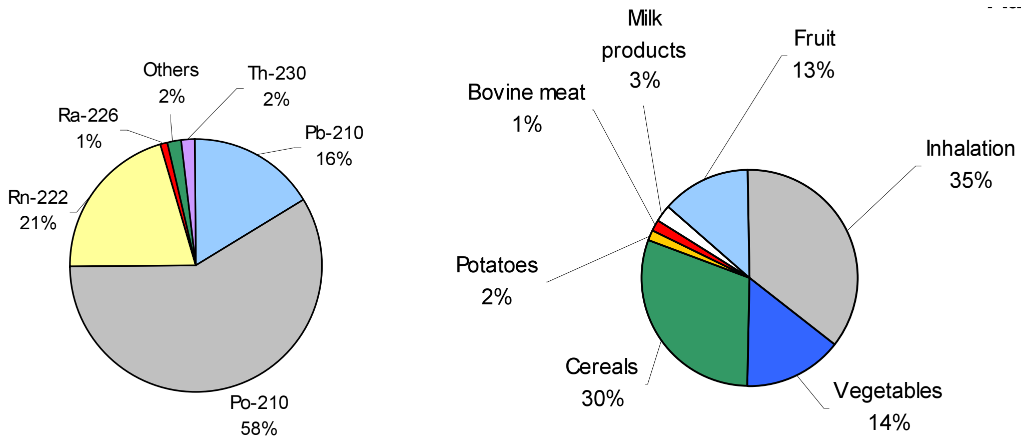

FIG. 2. Contributions of radionuclides and exposure pathways to the dose received by adults

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