NATURAL RADIOACTIVITY IN BUILDING MATERIALS - CZECH EXPERIENCE AND EUROPEAN LEGISLATION

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Abstract

An overview is presented of regulation and control of the natural radioactivity in building material in the Czech Republic (Atomic act 18/1997) and evolution of attitudes in the past 20 years. The sense is explained of regulation based on activity index and Ra-226 concentration, investigation levels and limit values for different materials and radiation protection optimisation process. The results of measurements are summarised. Czech experiences with several thousand houses contaminated by Ra-226 in the past (highly emanated aerated-concrete houses up 1kBq/kg, slag concrete houses up 3kBq/kg and houses in Joachimstahl up 1MBq/kg) are summarised. The EU recommendation "Radiological Protection Principles concerning the Natural Radioactivity of Building Materials" is discussed, too.

1. Introduction

Historical background:

The Czech Republic belongs to countries with highest indoor radon concentration in the world (mean radon value 140 Bq/m^3). The primary cause is the high radon concentration in the soil. However, during the seventies and eighties there were found also three groups of houses with significantly elevated radium concentration in the building materials:
1) houses in the small town Jáchymov (Joachimstal) contaminated by residues from uranium paints factory and radium factory,
2) houses from highly emanated autoclaved aerated concrete produced from flying ash,
3) family houses from slag concrete. Some of the important details are summarised.

Houses in Jachymov (Joachimstal)

Jachymov (Joachimstal), the mining town known from Middle Age thanks to silver-mines and coinage (minted coins called Joachims-thalers gave their name to the Thaler and the US dollar), later uranium mining industry and radium producing factory. Due to silver and uranium exploration and factory producing uranium paints and radium before the World War II, the town was locally contaminated in the past. The residues from factories were also used as additive into plaster and mortar in a lot of the Joachimstal´s houses, with ^{226}Ra mass activity up to 1 MBq/kg in extreme cases and indoor gamma dose rates in the range of 10-100 μGy/h. The contamination was not uniform. The case was revealed in the seventies, but that time there was no national legislation concerning indoor radon and indoor radioactivity.

The remedy measures carried out in the seventies were drastic - the worst houses were demolished and material taken away and processed in uranium ore mills. Most remaining contaminated houses were mitigated later in the nineties. Remedial measures were based on detailed radon and gamma diagnostic and targeted removal of plasters and mortar, if there was only local contamination.

Family houses from autoclaved aerated concrete

In the 1980 there was found the group of family houses built from autoclaved aerated concrete (^{226}Ra mass activity up 1 kBq/kg). Because of high emanation coefficient of this materials (range 15- 30 %), the indoor radon concentration was up to 1000 Bq/m^3 in extreme cases. The indoor gamma dose rates were in the range of 0.1-0.3 μGy/h, what is the upper level of normal outside dose rate background in Bohemia. Some 20 000 houses from this material were built in the period 1963-1980. Fortunately, the aerated concrete was used in most of them only as the minor part of building material and hence indoor radon concentrations exceeded the intervention level only in about 1-2 % of these houses.

Forced central or local ventilation systems were used for effective mitigation . Different radon barriers (special painting and special wallpapers) were tested but without good results.

Family houses from slag concrete

The last group was discovered in the 1987. There were found some 3000 factory-made family houses from slag concrete panels (^{226}Ra mass activity in the range 1-10 kBq/kg). The source of the activity was slag from a small power plant burning high radioactive local coal from a mine near Prague. The first Producer Company knew the radioactive danger of the slag from the fifties. After changes in the factory ownership the new management took no care of this danger and more than 2000 family houses were built in the period of 1972-1983 distributed all over the country, most of them in Central Bohemia around Prague. All the peripheral and supporting walls were made from this material, while some partition walls were from bricks. Because of small emanation coefficient of the material (only 1-5%) the indoor radon concentration was only in the range of 200-800 Bq/m^3. The indoor gamma dose rates in this group of houses were in the range of 0.5-2 μGy/h; the spatial variation in rooms was characterised by a factor of 2, with highest values in the corner of peripheral walls.

The owners of these houses were aware of the cause of their trouble and applied for remedial measures or the possibility to buy up these houses by the government. The government has agreed after great struggle in 1991. Most of the owners have accepted remedial measures; only 4 % owners have sold their house. After some experiments, it was clear that radon removal by forced ventilation was the only effective and reasonable mitigation measure. The ventilation system with heat recovery, controlled by a central computer, was found to be most effective countermeasure and was used in practice. Radon level was reduced to 30% of the former values on average. Other remedial measures (gamma shielding, removal of building material, wall covering by special radon proof materials, etc.) were tested in some cases but were rejected as noneffective. This case revealed the differences in risk perception. As it turned out:
- despite the effective doses were the same, the fear of gamma exposure was generally much higher than that of radon exposure (radon exposure was supposed to be easily mitigated),
- the human exposure in houses built from man-made “radioactive” building materials (caused by failure of governmental supervision), was perceived worse compare to exposure from natural soil radon,
- while at the beginning nearly all owner called for buy-up of their radioactive houses by the government, only about 4 % really decided to sell it.

2. Development of regulation in the Czech Republic after 1987

It was the mentioned experiences from the past which lead to strict regulation of natural radioactivity. The first regulation was prepared in 1987 for indoor radon a gamma exposure in existing houses and at the same time for natural radioactivity in building materials.

2.1. Interventional levels for existing houses

The first recommendation on limitation of both indoor exposures (gamma and radon) in the houses was prepared in 1987. Recommended value for remediation of indoor radon exposure in existing building was set to 400 Bq/m^3. Recommended value for indoor gamma dose-rate was set to 2 μGy/h. Having in mind both radon and gamma exposures, the special intervention level that summed both exposures was defined by index *S*:

$$S = \frac{D}{2\mu Gy/h} + \frac{C_{Rn}}{400 Bq/m3}$$

where D is the gamma dose rate (μGy/h) and C_{Rn} is the long-term radon concentration (Bq/m^3). This sum rule (used only if $D > 0.5$ μGy/h) and value $S = 1$ were used for decision making on remedial measures with governmental support.

2.2. Regulation concerning the natural radioactivity of building materials

The limit value ^{226}Ra was set to 120 Bq/kg. This value was derived from model room calculation so that the building material will contribute to indoor radon limit value (200 Bq/m3) in new houses not more than 30% (under conservative conditions), having in mind that the underlying soil is the most important source of indoor radon. The other systems of regulation based on limitation of radon exhalation rate or emanation coefficient were discussed but rejected at the end because of sophisticated measurements of exhalation, long term changes and complicated system of limitation (YU 1997, Roelofs 1994, Petropoulos 2001).

3. The European regulation

3.1. Philosophy

The radiological protection principles concerning the natural radioactivity of building materials were developed in the second half of the nineties in the Europe. The European Commission (EU Recommendation no.112, 1999, Mustonen 1997, Markkanen 2001)) formulated philosophy, which could be summarised as follows:

- Building materials cause exposure by direct gamma radiation and by radon Rn released from the materials into indoor air. Their relative importance varies considerably, but both pathways should be considered when establishing radiological criteria for building materials.
- When limits are set for exposure to natural radiation from building materials it must be clearly defined as to what extent the exposure caused by ‘normal’ background is included. The dose criteria used for controls should, therefore, be defined as the excess exposure caused by building materials, i.e. the background dose from natural radionuclides in ‘normal’ Earth’s crust need to be subtracted
- The doses to the members of the public should be kept as low as reasonably achievable. However, since small exposures from building materials are ubiquitous, controls should be based on exposure levels which are above typical levels of exposures and their normal

variations.

- All building materials contain some natural radioactivity. Small, unavoidable exposures need to be exempted from all possible controls to allow free movement of most building materials within the EU. The concentrations of natural radionuclides in building materials vary significantly between and within the Member States. Restricting the use of certain building materials might have significant economical, environmental or social consequences locally and nationally. Such consequences, together with the national levels of radioactivity in building materials, should be assessed and considered when establishing binding regulations.
- The amount of radium in building materials should be restricted at least to a level where it is unlikely that it could be a major cause for exceeding the design level for indoor radon introduced in the Commission Recommendation (200 Bq m^{-3}) or - better - some fraction of it in order to allow some contribution from other sources, especially from the underlying soil, without exceeding the design level.
- Exceptionally high individual doses should be restricted. Within the European Union, gamma doses due to building materials exceeding 1 mSv a-1 are very exceptional and can hardly be disregarded from the radiation protection point of view. When gamma annual doses are limited to levels below 1 mSv, the ^{226}Ra concentrations in the materials are limited, in practice, to levels which are unlikely to cause indoor radon concentrations exceeding the design level of the Commission Recommendation (200 Bq m^{-3}).

Controls on the radioactivity of building materials can be based on the following radiological criteria and principles:

a) Dose criterion for controls

Controls should be based on a dose criterion which is established considering overall national circumstances. Within the European Union, doses exceeding 1 mSv a-1 should be taken into account from the radiation protection point of view. Higher doses should be accepted only in some very exceptional cases were materials are used locally. Controls can be based on a lower dose criterion if it is judged that this is desirable and will not lead to impractical controls. It is therefore recommended that controls should be based on an annual dose in the range 0.3 – 1 mSv. This is the excess gamma dose to that received outdoors.

b) Exemption level

Building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at the most. This is the excess gamma dose to that received outdoors.

Separate limitations for radon or thoron exhaling from building materials should be considered where previous evaluations show that building materials may be a significant source of indoor radon or thoron and restrictions put on this source is found to be an efficient and a cost effective way to limit exposures to indoor radon or thoron.

Investigation levels can be derived for practical monitoring purposes. Because more than one radionuclide contribute to the dose, it is practical to present investigation levels in the form of an activity concentration index. The activity concentration index should also take into account typical ways and amounts in which the material is used in a building.

The activity concentration index (*I*) is derived for identifying whether a dose criterion is met:

$$I = \frac{C_{Ra}}{300\ Bq/kg} + \frac{C_{Th}}{200\ Bq/kg} + \frac{C_K}{3000\ Bq/kg}$$

where C_{Ra}, C_{Th}, C_K are the radium, thorium and potassium activity concentrations (Bq/kg) in the building material. The activity concentration index shall not exceed the following values depending on the dose criterion and the way and the amount the material is used in a building:

Dose criterion	**0.3 mSv (annually)**	**1 mSv (annually)**
Materials used in bulk amounts, e.g. concrete	I < 0.5	I < 1
Superficial and other materials with restricted use: tiles, boards, etc	I < 2	I< 6

The activity concentration index should be used only as a screening tool for identifying materials which might be of concern. Any actual decision on restricting the use of a material should be based on a separate dose assessment. Such assessment should be based on scenarios where the material is used in a typical way for the type of material in question. Scenarios resulting in theoretical, most unlikely maximum doses should be avoided.

For the radon pathway, the evaluation of the excess dose caused by building materials is more complicated and the contribution of building materials must be evaluated by using theoretical model and general assumptions on the parameter values. However, it is very difficult to take into account all parameters, e.g. surface-volume ratio, the effect of surface treating done at the building site, and the ventilation rate. The reasonable approach for considering the radon pathway is to limit the amount of ^{226}Ra in the building material so that the indoor radon concentration cannot rise above some pre-set level even under unfavourable conditions.

3.2. Application

The dose criterion used for national controls should be chosen in a way that the majority of normal building materials on the market fulfil the requirements. Usually measurements of activity concentrations are needed only in case where there is a specific reason to suspect that the dose criterion for controls might be exceeded. The Member States should require, as a minimum, the measurement of types of materials which are generically suspected.

Appropriate dose assessments should be performed if it is discovered that the reference value of the activity concentration index is exceeded. Normally the producer or dealer would be responsible for ensuring and showing that a material put on the market fulfils the radiological requirements set by the Member State. However, other approaches might also be applied according to national circumstances and administrative practices, e.g. the builder or designer of the building could be the responsible party for ensuring that a new building complies with the radiological requirements. The disadvantage of this approach is that appropriate training in radiological modelling should be arranged to all designers and builders.

Materials should be exempted from all controls concerning their radioactivity if it is shown that the dose criterion for exemption is not exceeded. This can be done by comparing results of activity concentration measurements with the activity concentration index, or as appropriate, by means of a material-specific dose assessment. An exempted material should be allowed to enter the market and to be used for building purposes without any restrictions related to its radioactivity. In the case of export within the EU, it is understood that the value of the activity concentration index or a declaration of exemption should be included in the technical specifications of the material.

When industrial by-products are incorporated in building materials and there is reason to suspect that these contain enhanced levels of natural radionuclides, the activity concentrations of these nuclides in the final product should be measured or assessed reliably from the activities of all component materials. Where necessary, also other nuclides than ^{226}Ra, ^{232}Th and ^{40}K shall be considered. The dose criterion should be applied to the final product.

Some traditionally used natural building materials contain natural radionuclides at levels such that the annual dose of 1 mSv might be exceeded. Some of such materials may have been used already for decades or centuries. In these cases, the detriments and costs of giving up the use of such materials should be analysed and should include financial and social costs.

4. CONTEMPORARY REGULATION IN THE CZECH REPUBLIC

Contemporary Czech legislation concerning indoor natural radioactivity and remedial measures is based on the Atomic Act No.18/1997 and Decree No. 307/2002.

The producers and importers of building materials are obliged to ensure systematic measurement of the natural radionuclides in the building materials and to submit results to the State Office for Nuclear Safety. It affords unique opportunity to get nearly complete data set of natural radioactivity in the building material in the Czech market. The central database was prepared at the beginning of 1998 and more than 5000 results of measurements were obtained up to now (Vlcek 2007, SURO 2005). The results of measurements were obtained from 20 laboratories from the Czech Republic. The laboratories are periodically tested and they take part in the comparison organized by National Radiation Protection Institute.

The framework of regulation in Czech Republic is based on two-step system:

1 step: screening (exemption) level for activity concentration index (I), based on gamma dose-rate estimation

$$I = \frac{C_{Ra}}{300\ Bq/kg} + \frac{C_{Th}}{200\ Bq/kg} + \frac{C_K}{3000\ Bq/kg}$$

2 step: limit levels for ^{226}Ra mass activity. The sense - limitations of radon exhaling from building materials under unfavourable conditions

Table 1: screening levels (activity concentration index I)

Index I	Type of building material
0,5	Material used in bulk amount (e.g. brick, concrete, gypsum)
1	Raw material (e.g. sand)
2	Material used in „small“ amount (e.g. tiles)

Table 2: limit levels for ^{226}Ra

type of building material	Limit value ^{226}Ra (Bq/kg) Buildings with	Limit value ^{226}Ra (Bq/kg) Other construction

	stay of person	without stay of person
Material used in bulk amount (e.g. brick, concrete, gypsum)	150	500
Other material used in small amount (e.g. tile..) and raw material (sand, building stone, gravel aggregate, bottom ash..)	300	1000

The activity concentration index is used as a screening tool for identifying materials which might be of concern. The index mean values in Czech Republic market are (Vlček 2007): natural building stones 0.4, clay bricks 0.6, concrete 0.3, aerated concrete 0.53, slag concrete 0.69, coal ash and slag 0.63, gypsum 0.17. It was shown that for concrete, aerated and light-weight concrete, clay bricks, natural building stones exposure above 0,3 mSv is possible almost anywhere bulk amounts of material are used. Exposure above 1 mSv is possible if bulk amounts of the concrete contain slag, fly ash or natural sand or rock rich in natural radionuclides

If activity concentration index *I* is above exemption level, the producer of building material is obliged to perform the cost-benefit analysis (the process of the optimisation of radiation protection). The producers need not carry out intervention if the costs are higher than the benefits of such remedial measures. In other word, the costs related to reduction of radionuclide concentration in building materials (namely by a change of raw materials or their origin, by sorting raw materials, by a change of technology and other suitable intervention), would be demonstrably higher than the risks in health detriment. The benefits of remedial measures is calculated in such a way that a reduction of collective effective dose for a group of individuals being assessed is multiplied by a factor of 0.5 million CZK/Sv for the exposure to natural radionuclides. The aim is to reduce the public doses to level as low as reasonably achievable.

5. Conclusion

There are several challenges in the possible harmonisation of controls on the radioactivity of building materials. It would be desirable that controls would be sufficiently uniform to allow movement of building materials within the EU. The levels of natural radionuclides in building materials vary significantly between countries and areas and the recommendations for harmonising controls should provide certain flexibility for taking into account specific national circumstances. The Czech experience after 20 years of application of regulation shows that it is possible to regulate all important exposures caused by natural radionuclides in building materials. It is obvious that high indoor gamma dose-rate can be hard to reduce post facto in the existing house. The aim of regulation is to restrict exposure in new buildings especially the highest individual doses. One must also have in mind decreasing ventilation rate (even below 0,1 h^{-1}) in new energy-saving building can lead to problems with indoor air quality. The doses to the members of the public should be kept as low as reasonably achievable, but the dose criterion used for controls is chosen in a way that the majority of normal building materials on the market fulfil the requirements. If aim of regulation would be decrease future collective dose however, the low activity materials should be recommended and public awareness on this issue should be fostered.

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