Estimation and evaluation of doses of radon exposure and related health hazards incurred in the phosphate industry and through the use of gypsum products in the building industry.

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Introduction

Radon and radon progeny are now believed to be the main sources of public exposure to radiation. Therefore many studies on indoor air are performed. Not much attention is paid to radon exposure in workplaces, although enhanced levels can be found there too. The European Union has therefore established Basic Safety Standards. These must be included in national legislation before May 2000. It is necessary to provide some preliminary case studies to evaluate the impact of this implementation.

The aim of the proposal is to evaluate for different groups of workers the possible health risks caused by natural radioactivity present in some widely produced and used materials. In particular, attention is paid to the acquired dose caused by radon exhalation during production of different phosphate end products from the raw materials.

The health effects of some phosphate-industry end products used in the building industry are studied in detail. Special attention is paid to the use of phosphogypsum as raw material for plaster production.

Materials and methods

The measuring campaign in the phosphate industry consists of 2 parts. At first, the (natural) radioactivity contents of all raw materials used is analysed by gamma spectroscopy, using a HPGe-detector. Attention is also paid to waste products and end products. Next, an inventory of radon exposures in the different working areas is made by means of passive integrating dosimeters. Special attention is paid to workplaces with high occupancy. Long-term measurements have been made, with an exposure time of 3 to 6 months. First we used the diffusion chambers normally used in dwelling studies, but as these are metal chambers, they are not convenient for use in the chemical industry (they become rust-eaten). Afterwards we used ‘classic’ plastic Karlsruhe diffusion chambers. These are better for the extreme conditions of the factory.

To start our measurements in the building industry, we contacted plaster-workers by means of the WTCB (Scientific and Technical Centre for the Building Trade). To prepare these measurements, samples of the different plaster products were investigated. Samples were taken of the plasters most used in Belgium. Gamma-spectroscopy of these samples was carried out to measure the natural radioactivity content. In addition, radon measurements were performed in 2 of the 4 gypsum factories.

As the occupancy of the different workplaces is not always exactly known, it is sometimes better to use personal dosimeters. A selection of different dosimeters was tested to select one that can measure 100 Bq/m³ in one working month with an accuracy of 20 %. As existing techniques are not sensitive enough, a new kind of detector was developed, combining 2 techniques: activated charcoal and a track-etch detector. The activated charcoal adsorbs the radon from the air and the track-etch detector detects the alpha radiation emitted by the radon and its decay products. Use of this combination enhances the sensitivity 10-fold. This makes it possible to measure relatively low radon concentrations in short exposure times.

Results

Measurements were performed in the 3 major phosphate factories in Belgium. From radon measurements in the phosphate industry we can conclude that the radon concentration depends strongly the phosphate ore and production process used. Sedimentary ore contains more radium than volcanic ore, so the radon concentration is higher when sedimentary ore is used. The increase in radon concentration is also dependent on the production process. When the radioactivity of the raw material is concentrated in the waste stream, the radon concentration is enhanced there.

Measurements in Firm 1 indicate that both the activity concentration in the phosphate ore and phosphogypsum and the radon concentration in the different workplaces are very low. Only at the phosphogypsum storage site is a somewhat enhanced value found (35 Bq/m³). The gamma dose rate is also very low, between 0.09 and 0.45 µSv/h. The latter value is found at the phosphogypsum storage site where occupancy is very low. We can thus conclude that the annual dose of these workers is less than 1 mSv.

Firm 2 uses a combination of sedimentary and volcanic ore. Due to the use of sedimentary ore we found a somewhat higher radon concentration. Values up to 200 Bq/m³ were found in the filter room, where the phosphogypsum and phosphoric acid are separated. Enhanced values were also found at the phosphogypsum storage site. Workers of Firm 2 may receive an annual dose somewhat higher than 1 mSv.

Firm 3 uses a different method to process phosphate ore, namely acidulation with hydrochloric acid. Besides this, sedimentary ore is used. This results in a higher radon concentration. Values exceeding 1000 Bq/m³ are found. As the radium present in the phosphate ore is concentrated in the waste products, an enhanced gamma dose is also present (up to 4.4 µSv/h). Taking into account the different occupancies of the different workplaces, this results in an annual dose of 5 mSv. Measurements with active devices are performed to indicate the main radon source. Enhancing the ventilation is proposed as a solution to the radon problem. Further control measurements will be performed.

Measurements of the gamma activity of the different plasters give results comparable to those published in the literature. One firm uses sulphogypsum as raw material. This gives very low values of radium and thorium activity, near the detection limit of the HPGe-detector. Natural gypsum, used in two other firms, also has a low natural radioactivity with a radium activity of about 10 Bq/kg. When phosphogypsum is used as raw material, the radium activity is about 170 Bq/kg and the thorium activity is 140 Bq/kg. When this us used in the plaster, enhanced radon concentrations may occur.
In 2 of the 4 firms radon measurements were carried out. One firm uses sulphogypsum as raw material and the other uses phosphogypsum. For neither firm did these measurements yield particularly high values. The highest value found was 50 Bq/m³ near the oven of Firm 7. This is a rather closed space and great amounts of gypsum pass through it. As the average radon concentration in Belgium is 53 Bq/m³, we can hardly speak of an enhanced value. All other values are around 20 Bq/m³, normal for Belgium.

When the occupancy in the different workplaces is not exactly known, it is better to use personal dosimeters. The dosimeter developed in our lab combines activated charcoal with a track-etch detector. First, a selection of different charcoal types was tested. On the basis of its adsorption capacity and adsorption and desorption times for both radon and water, Carboxen was deemed the best charcoal for use in the personal dosimeter. As track-etch detector, Makrofol is used, because this equipment is well known and provides a low and stable background. We also investigated the influence of the thickness of the charcoal layer on the number of tracks. For layers between 1.5 and 3 mm, no difference was found. A thickness of 2.5 mm is now used. The influence of the relative humidity was also investigated, as the charcoal also adsorbs water, which substitutes for the radon. Attention is paid to the regeneration and storage of the detector overnight, when the workers are not present. With these tests, it is possible to measure 100 Bq/m^2 in one working month with an accuracy of 20 %.

Conclusions

From measurements in the phosphate industry we can conclude that the radon concentration depends strongly on the production process and (natural) radioactivity of the raw material. When volcanic ore is used, there is no radon problem at all and the annual dose stays below 1 mSv, the limit for public exposure. In this case no further action is necessary. When sedimentary ore is used, enhanced values can be obtained and attention has to be paid to the problem. Remedial action can be considered in some cases.

Measurements in the gypsum industry show no enhanced values. All values are close to the average radon concentration in Belgium. The contribution of plasters based on phosphogypsum has to be further investigated. By combining activated charcoal with a track-etch detector it is possible to measure a radon exposure of 100 Bq/m^3 in one working month. This opens prospects for personal dosimetry at workplaces where enhanced concentrations are found and the occupancy is not exactly known.