Radiation exposure at production and use of thoriated gas mantles

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Abstract. Even now thoriated gas mantles are quite often used in Germany. The main use takes place in street lighting, lighting of railway points and nautical lighting. During production, maintenance and repair of these gas illumination devices, thorium could be released in inhalable form as dust, leading to internal contamination. The analysis of working places where workers change the gas mantles, clean the lights and do other maintenance work outside show incorporation of radioactivity due to the working conditions. During maintenance of street lighting in workshops there is a risk of workplace contamination, not only in the workshop itself but also in the maintenance van. Results of a measurement campaign are shown in the paper. A few months ago, the production of thoriated gas mantles was closed down in Germany. The radiation exposure during the production process has been investigated very comprehensively. It can be shown that there had been relatively high exposures in the past. The lifetime doses of two workers producing gas mantles may exceed the limit of 400 mSv if the range of errors in estimation is taken into account. The main part of these doses results from incorporation rather than from external exposure, although in some cases external exposure could be fairly high. The paper describes the materials and methods used in assessing the doses and presents the results of workplace-related analyses and internal dose assessments for the production, storage and use of thoriated gas mantles.

1. Introduction

1.1. Reasons for using thorium in gas mantles and frequency of use

Thorium oxide is the main component of gas mantles in order to raise the light emitting efficiency. There have been several investigations to try to replace thorium by other rare earth metals, but until now the results concerning the light emitting efficiency and the mechanical stability are unsatisfactory. The total activity of a single gas mantle can vary from a few Bq up to 17 kBq [1].

The principal use of these gas mantles in Germany is for gas light in some larger cities, for example Dresden, Düsseldorf, Frankfurt and Berlin. There are up to 20 000 gas lanterns with 10 gas mantles each in one single city. In Germany there are approximately 10 000 nautical lighting devices and 100 000 railway points (switches) equipped with gas mantles. Furthermore, thoriated gas mantles are used in camping lights and other private applications. Altogether about 1.5 million gas mantles are sold annually in Germany. These public and railway lighting devices have to be maintained and repaired. It has been estimated that more than 250 people are occupationally involved with thoriated gas mantles in Germany.

1.2. Hazards while using thorium as an unsealed source

Basically two exposure pathways are distinguishable: internal exposure through inhalation of thorium-containing dust and external exposure when higher activities are handled. Normally the dose from external exposure is less than the dose from internal exposure, but nevertheless it cannot be neglected. There are a few workplaces, especially those where gas mantles are produced, where the external exposure could contribute of the order of some millisieverts per year.

During the production and use of thorium-containing gas mantles, internal exposures cannot be totally avoided. Therefore an incorporation monitoring program has to be established to protect the exposed workers. The main procedures are the analysis of 24 h urine samples or the sampling and analysis of individual-related air monitoring samples. Interpretation of the analysis results is difficult because of biased views with respect to naturally occurring radio nuclides.

2. Material and methods

2.1. Reference nuclide

The use of thoriated gas mantles may cause an exposure to ^{232}Th, ^{230}Th and their radioactive progeny. Out of this mixture of radionuclides only the three isotopes ^{232}Th, ^{230}Th and ^{228}Th are radiologically relevant, as a comparison of the dose coefficients shows [2]. None of the other daughter radionuclides make a significant contribution to the overall dose. The radionuclide composition of a gas mantle depends on the time elapsed after chemical separation of the thorium. The activity of ^{228}Th could, at most, be the same as that of ^{232}Th. The activity of ^{230}Th could be of the same order of magnitude as that of ^{232}Th as a worst case, depending on the origin of the raw material. So an upper bound on the estimation will be obtained if — as a first order simplification — the activity of all three thorium nuclides is set to be equal. The actual dose would not be underestimated with this assumption. The advantage of this approach is that one can focus the analysis on the activity of one reference nuclide, namely ^{232}Th.

2.2. Air monitoring

The workers were equipped with personal breathing zone air samplers during their normal work (see Fig. 1). The inhalable dust [3] was collected on membrane filters of the type BIA MF 11301 (pore size 8 µm, 37 mm diameter) by using a sampling unit with an electronically controlled volume airflow of 0.21 m^3 h^{-1}.

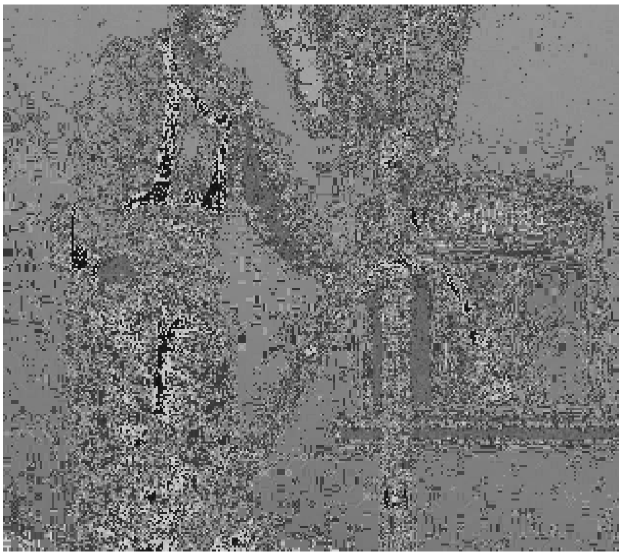

Fig. 1. Personal air monitoring during maintenance of gas lanterns

2.3. Urine samples

In the case of workers who maintain gas lanterns, 24 h urine samples were collected. Second samples were taken two months later. Those workers employed in the production of thoriated gas mantles have been regularly monitored with 24 h urine samples for a long time [4].

2.4. Smear samples

Smear tests were taken with smears of the type Pedi AG, Zürich, Nr. 100829. Several working sites were smeared with a surface area of 100 cm². A removal factor of 10 % was assumed. All smears were analysed for ^{232}Th-acitivity.

2.5. Analysis

For the determination of ^{232}Th it was decided to use a high resolution inductively coupled plasma mass spectrometer (ICP-MS) [5]. Filter-materials and smears were ashed and dissolved with nitric acid. Urine samples were directly measured after addition of nitric acid. The detection limit of this method is 0.001 µg/sample or, for ^{232}Th, 0.01 mBq/sample.

3. Thorium excretion rates due to non-occupational ingestion

Within the framework of a project supported by the German Federal Office for Radiation Protection, individuals who were not occupationally exposed to thorium were chosen as volunteers for a broad investigation of naturally induced thorium excretion rates in urine. The persons lived in different regions of Germany with a wide spectrum of naturally occurring radionuclides in the soil. This is reflected in the results of thorium in urine for these individuals, which span many orders of magnitude. In total, 306 urine samples were measured. The minimum value was lower than the detection limit of 0.001 µg/d and the maximum value was 2.024 µg/d. The median was 0.012 µg/d, equivalent to 0.05 mBq/d ^{232}Th. This median value is a reference value for the evaluation of the excretion rate of ^{232}Th.

4. Exposure estimation for production of gas mantles

For almost a decade, a gas mantle production facility in Germany was investigated regarding the exposure of its workers. Many urine samples were collected and air monitoring measurements were made. The first measurements — ten years ago — showed a high activity concentration in the workplace. Step by step radiation protection measures were installed, for example the enclosure of machinery and installation of air extraction systems. The average air activity concentration of ^{232}Th for the highest exposed procedure (cutting) decreased in the course of these measures from nearly 200 mBq/m³ to 10 mBq/m³.

4.1. Production steps

The manufacture of gas mantles is carried out in several production steps. The basic material is a cotton hose in the form of a coil. This is soaked in thorium nitrate solution and treated with liquid ammonia. The soaked coils are centrifuged, uncoiled, and hung up for drying. The next step involves cutting into smaller pieces of fabric, which are then pressed into the required form and attached to a mounting. The overlapping tissue is cut, following which these intermediate products are burnt. During the later steps, the thorium is in dioxide form. The cotton fabric is totally burnt away, leaving only the thorium dioxide mantles. To stabilize these fragile gas mantles for transport, they are dipped in varnish. The last step is the packaging. Additional operations involving thorium exposure are the testing of selected gas mantles, the cleaning of the working sites and the maintenance and repair of the machinery. During all of these operations, thorium could be inhaled, first in nitrate form and later in dioxide form.

Fig. 2. Personal air monitoring during drying procedure of coils

4.2. Results of the urine samples

More than 200 urine samples from the 10 workers involved were analysed — one urine sample per year up to 1994 and four samples per year since 1995. Most of the results were below the detection limit, which varied over the period of the investigation from 1.5 to 0.2 mBq/d ^{232}Th. However, for nearly every worker there were some results above the detection limit. The maximum value was 2.8 mBq/d. A comparison with the median value of the non-occupationally induced excretion rate of 0.05 mBq/d ^{232}Th (see Section 3) indicated an occupationally induced incorporation among the workers investigated.

The measurements were interpreted in accordance with the ICRP human respiratory tract model [6] and the dose coefficients for intakes of radionuclides by workers [7]. A chronic incorporation and an AMAD of 5 µm were assumed, as well as a mixed chemical form of the inhaled thorium in the ratio of nitrate to dioxide appropriate for the worker concerned. As an example, the maximum rate of thorium excretion corresponded to an annual uptake of about 2600 Bq.

4.3. Results of air monitoring

Between 1995 and 2003, many personal and stationary air monitoring samples were collected. The measurements identified separately the workplaces where thorium nitrate was handled and those where thorium dioxide was used, in order to distinguish between the two inhalation classes and production steps. The results were expressed as uptake per working hour. For every worker, a working matrix was determined, showing the relative periods involved in the different production steps over the course of a year. The total annual uptake was calculated by multiplying the time spent at each workplace by the uptake per working hour at that workplace and then summing over the whole year. The highest annual uptake was about 900 Bq for dioxide and about 90 Bq for nitrate.

4.4. Dose estimation for workers in a gas mantle production facility

The external dose, measured with approved dose meters, had to be additionally taken into account. The annual external doses varied between zero and 6.8 mSv depending on the operations performed by the worker. The highest external doses were found to be associated with working with the barrels in which the thorium nitrate was delivered, mixing of the soak solution and handling of the soaked coils.

For each monitored worker, the assessed values resulting from the urine measurements were compared with those resulting from the air monitoring measurement. The accumulated dose was expressed for each worker as a range of doses over the worker's lifetime to take account of the errors of the estimation. Based on the reference nuclide, all relevant nuclides were taken into account in this assessment. The lowest lifetime dose range for an individual worker was 23–84 mSv and the highest was 134–483 mSv. For two of the workers, the error bars extended beyond the limit on lifetime dose applied in Germany of 400 mSv.

5. Exposure estimation for use of gas mantles

An investigation was performed to estimate the intake of thorium by maintenance workers of a lighting supplier company.

5.1. The working steps

When only cleaning and changing of gas mantles is requested, maintenance of the lantern heads is done at the road location. Workers go to the location in service vans and reach the lantern heads by ladder. The vans are equipped in a way that minor repair work can be done inside. If a lantern needs more extensive repair, the head is removed and brought to a workshop where it is completely disassembled and subsequently reassembled. Leak tests of gas tubes and gas mantles are done in a separate room next to the main workshop. According to the working records, five workers were identified as having mostly done outside maintenance work and were therefore suitable for participation in the project. Of those employees who had done workshop and final checking work, only one could be persuaded to take part in the project. Samples were taken from the participants, as described in Sections 2.2 and 2.3 and smear-test samples were taken from inside the workshops, from inside the vans and from inside the lanterns when they were opened for cleaning and changing of the gas mantles.

5.2. Results of the urine samples

Ten urine samples of five workers who maintained gas lanterns were analysed to determine the excretion rates. All results were above the detection limit of the analytical method. Six of the results fell below the median value of 0.05 mBq/d for natural radioactivity of ^{232}Th as described in Section 3. Four values were in the range 0.1–0.3 mBq/d ^{232}Th. The precise maximum value was 0.32 mBq/d.

5.3. Results of the air monitoring

Three workers were equipped with personal breathing zone air samplers (see Section 2.2) during their normal maintenance work for two days. The exposure time was from 1 h to about 6 h. The results of the filter analysis were in the range 1.28–0.02 mBq per filter. The highest value was associated with an exposure time of 1.5 h.

5.4. Results of the smear samples

Eighteen smear test samples were taken. Only the ^{232}Th activity was determined. This activity was in the range 5–900 mBq per sample. For the ^{232}Th activity alone, a third of the samples exceeded the maximum value of 100 mBq/cm² permitted in Germany. If the other relevant nuclides are taken into account, this value would be exceeded for most of the other workplaces.

5.5. Dose estimation for workers using gas mantles

The excretion rates of ^{232}Th in urine are of the same magnitude as the excretion rates attributable to natural uptake. Therefore, incorporation attributable to occupational exposure is not evident from these measurements. Nevertheless it might be interesting to evaluate the dose of the most highly exposed individuals in this investigation. Under the assumptions described briefly in Section 2.1, a rough and conservative estimation could be made taking into account the dose coefficients for inhalation and the retention values for urine excretion. This results in a dose of 0.16 mSv for air monitoring measurements and 0.27 mSv for urine measurements. This difference can be deemed acceptable when considering the possible sources of errors in the different methods used.

6. Summary

According to the legislation, the maintenance of gas lanterns investigated in this study does not need to be notified to the authorities. The maximum dose of these workers is far below an annual effective dose of 6 mSv and even far below the average dose received by the citizens of Germany from natural sources of about 2.1 mSv per year [8].

The smear measurements require that the stationary workshop and the workshop vans be professionally decontaminated, because most of the surface contamination exceeds the permitted limit of 100 mBq/cm².

The comprehensive investigation conducted at a gas mantle factory has shown that workers could receive relatively high doses. The introduction of radiation protection measures has resulted in a clear reduction in the dose for these workers.

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