Dose estimates for exposure to radioactivity in gas mantles

DOSE ESTIMATES FOR EXPOSURE TO RADIOACTIVITY IN GAS MANTLES

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ABSTRACT

In this paper dose estimates are given for internal and external exposure that result, due to radioactive thorium, from the use of the incandescent mantles for gas lanterns.

The collective, effective dose equivalent for all users of gas mantles is estimated to be about 100 Sv per annum in the Netherlands. For the population involved (ca. 700,000 persons) this is roughly equivalent to 5% to 10% of the collective dose equivalent associated with exposure to radiation from natural sources. The major contribution to dose estimates comes from inhalation of radium during burning of the mantles. A pessimistic approach results in individual dose estimates for inhalation of up to 0.2 mSv.

1. INTRODUCTION

Thorium nitrate has been used in the production of incandescent mantles for gas lanterns since before the turn of the century. At high temperatures thorium oxide generates a bright light. All isotopes of the element thorium are radioactive. The radionuclide Th-232, which is present in the gas mantles, decays via ten different radioactive daughter nuclides, including Th-228. The application of thorium oxide in gas mantles is based on its physical/chemical properties and is not linked with its radioactivity. The presence of radioactivity in gas mantles must be seen as an undesired side effect, since it may cause radiation exposure to people involved in production, distribution and the application of gas mantles.

Next we must consider the dose estimates for:

- inhalation of radioactive aerosols while the gas mantle is burning
- inhalation of radon (Rn-220) exhalated from the mantle
- inhalation of thorium oxide aerosols in air while manipulating a mantle
- ingestion of thorium oxide
- external irradiation.

The collective dose equivalent is estimated for the Dutch situation on the basis of these results.

In the study consideration is given to the possible radiological consequences of transport and storage of very large quantities of gas mantles. A few remarks are made on the pollutional aspect of the uncontrolled removal to the environment.

2. RADIOACTIVITY IN GAS MANTLES

Thorium nitrate is won from thorium ore. All isotopes of the element thorium are radioactive. Thorium-232 decays via ten different daughter nuclides to the stable Pb-208. One of those daughter nuclides is Th-228. There is secular activity equilibrium in the ore between Th-232 and the radioactive daughter products. After extraction of thorium from the ore, the activity of Th-228 and Th-232 are the same. The activity of all other daughter products are initially zero, immediately after purification. Those activities grow as a result of the radioactive decay of Th-232 and Th-228. About 40 years after the thorium extraction there is again secular activity equilibrium.
Insufficient information is available on the possible abundance of Th-230 occurring in the natural decay chain of uranium (see par. 4.6).

The typical value for Th-232 activity in gas mantles is about 1000 Bq. Figure 1 shows the activity of each radionuclide in a gas mantle as a function of time since the extraction of thorium nitrate from the thorium ore.

The changes in activity of the different nuclides during the use of gas mantles can be summarized as follows.
Thorium activity (both Th-232 and Th-228) remain roughly constant.
About 30% of radium activity (Ra-228 and Ra-224) is mainly emitted during the first 45 minutes of burning.
About 60% to 70% of the original activity for Pb-212 and Bi-212 is distributed to the air. This emission takes place during the first 5 to 10 minutes of use.
Activity of Ac-228 apparently remains unchanged. The decrease in growth as a result of disappearance of the mother nuclide Ra-228 is of no influence. Reduction in Ac-228 activity is only a result of radioactive decay.
No detailed information is held as to the possible emission of Rn-220, Po-216, Po-212 and Tl-208. For all dose estimates stated in this report, the activity for these nuclides is assumed to be equal to the activity of Bi-212 and Pb-212.

It can be seen that the activity content in a gas mantle is significantly different, depending on the age of the thorium nitrate. In the following dose estimates we therefore discriminate between young mantles, with thorium nitrate several years old and old thorium nitrate extracted from the ore more than 40 years ago.
Table 2 shows the estimated radioactivity for the respective nuclides at different stages of use, for three time periods since extraction of the thorium from the ore.

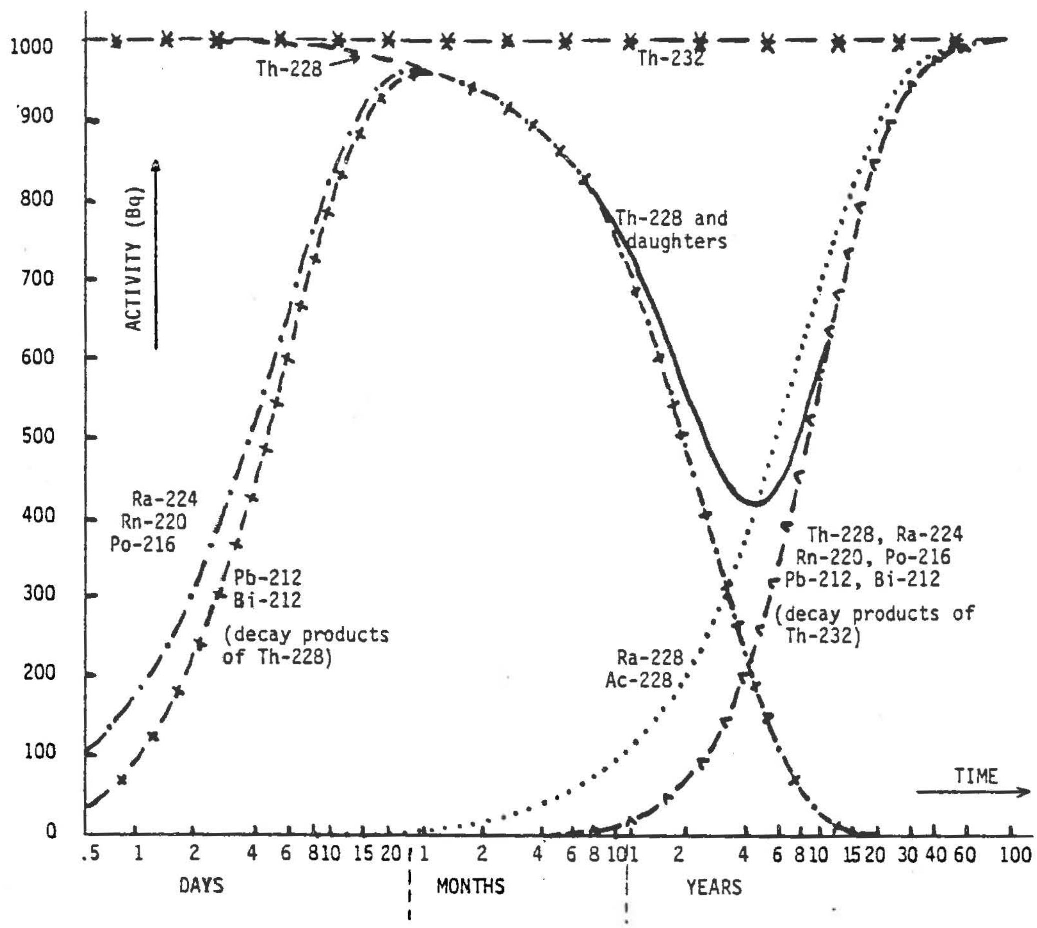

Fig. 1 Activity for Th-232 and decay products as function of "age" of Thorium

TABLE 2

Estimated activity [Bq] in a gas mantle at 3 stages of use for 3 ages of thorium

Stage of use	Unused gas mantle (zero burning hours)			Immediately after two burning hours			22 hours after the first two burning hours		
age [y]	1/12	4	40	1/12	4	40	1/12	4	40
Th-232	1000	1000	1000	1000	1000	1000	1000	1000	1000
Ra-228	10	380	1000	7	270	700	7	270	700
Ac-228	10	380	1000	10	370	980	7	270	700
Th-228	1000	425	1000	1000	425	1000	1000	425	1000
Ra-224	1000	425	1000	700	300	700	700	300	700
Rn-220	1000	425	1000	300	130	300	700	300	700
Po-216	1000	425	1000	300	130	300	700	300	700
Pb-212	1000	425	1000	300	130	300	620	270	620
Bi-212	1000	425	1000	300	130	300	620	270	620
Po-212	640	275	640	190	85	190	400	170	400
Tl-208	360	150	360	110	45	110	220	100	220

3. METHOD FOR DOSE CALCULATIONS

Dose estimates for internal exposure are based on ICRP publication no. 30. The estimated value for the committed effective dose equivalent follows from comparison of the intake with the annual limit on intake as defined by ICRP as follows:

$$H_{50} = \frac{\text{intake}}{\text{ALI}} * 50 \ [\text{mSv}]$$

For radiation protection purposes this approach is acceptable. However, it should be realised that the ICRP-30 model is meant to be used for occupational exposure. Further, it must be realised that results in dose calculations should not be interpreted in individual dose terms. Individual differences in diet, metabolism, etc. can result in significant deviations. N.B. For reasons of readability, effective committed dose equivalent is abbreviated to dose where feasible.

4.1. INHALATION OF AEROSOLS

Burning the gas mantle generates radiaoctive aerosols to the air. As shown in Table 2, the activity of the majority of nuclides decreases significantly in the first 2 hours of use. To some extent new growth takes place, depending on half-life.

For the first 2 hours of use the individual dose is estimated to be 0.01 mSv for a young mantle and 0.025 mSv for an old mantle. As already explained, the difference is caused by the larger amount of radium activity in old thorium nitrate.

Dose estimates are based on assumptions for room volume (15 m^3), ventilation rate (2 per hour) and breath volume rate (1.2 m^3 per hour).

Assuming a use of 2 hours per day for one week, the internal dose from inhalation of radioactive aerosols is calculated as 0.04 and 0.09 mSv per caput. The latter result is based on the assumption that all nuclides except Th-232, Th-228 and Ac-228 are released to the air each time the mantle is burned. Since this a somewhat pessimistic approach, we took the average values of dose calculations and conclude to a typical value of 0.02 a 0.06 mSv per caput and mantle.

It must be noted that calculations for the extreme case that old mantles are burned in a nonventilated room, the calculated dose is 0.2 mSv. Exposure of that kind should (and can) be avoided.

4.2. INHALATION OF RADON

Radon exhalation occurs both when the mantle is burned and when the mantle is not in use. The dose contribution from radon and radon daughters during burning is included in the results in par. 4.1.
The remaining dose contribution from radon and daughters is calculated for the condition that a gas mantle is used for one week, the occupancy is 8 hours per day, with additional assumptions for breath volume rate (0.5 m^3) and ventilation rate (2 per hour).
As an average, Ra-224 activity in the mantle is taken as constant and equal to 500 Bq.
Calculations result in an extra dose contribution of about 0.0015 mSv.
Without any ventilation, this dose contribution would be 0.011 mSv.

4.3. INHALATION OF DUST

By combustion of the tissue the form and robustness of the the mantle changes; what remains is a brittle structure. Any manipulation with used mantles can bring fine dust into the air. The actual spread of dust in air cannot be foreseen, but it must be clear that blowing away the residues of a mantle will be a quite common habit. A tentative dose estimate is based on the assumption that ca. 0.1% of the fine dust is distributed in a volume of 2 m^3 as potentially respirable. The AMAD is chosen as 1 micrometer. An occupancy of 10 minutes in the cloud causes inhalation of a fraction 0.1. The calculated internal dose due to inhalation of thorium oxide results in approximately 0.03 mSv per person. This dose estimate is based on Th-232 and Th-228 only, the lower radiotoxicity of the other nuclides taking into account.

4.4. INGESTION OF THORIUM OXIDE

A fraction of the dust from a used gas mantle could lead to ingestion. For dose calculations it is assumed that ca. 1% of the activity in the form of dust gets on to the hands and that approximately 1% of it will be swallowed. In addition, an intake of 0.01% is assumed, via deposition, on objects and/or foods. New mantles are plastic coated and are therefore assumed not to cause any ingestion.

On these assumptions the intake by ingestion is 0.02% of the total activity. Dose calculations were performed both for mantles in which secular activity equilibrium exists and for mantles in which only Th-232, Th-228 and Ac-228 are present. As an average the dose contribution due to ingestion is estimated to be 0.0002 mSv per caput and gas mantle.

4.5. EXTERNAL EXPOSURE FOR CAMPERS

External radiation exposure can result from gamma rays emitted by the various daughters of Th-232. Under conditions of secular activity equilibrium with Th-232 and neglecting radon exhalation, the dose rate at a distance of 1 metre is calculated to be 3 10^{-7} mSv per hour for a gas mantle with 1000 Bq Th-232. Even the most pessimistic assumptions about effective occupancy at 1 metre distance do not lead to dose contributions from external irradiations that are comparable with the dose contributions from internal exposure discussed in the foregoing.

4.6. THORIUM-230

Since thorium ore often contains uranium it must be noted that the isotope Th-230 could be also present in the thorium nitrate that is used in gas mantles. However, it was not possible to deduce a typical value for the relative abundance of thorium-230 from literature. Based on our own alpha-spectromectric measurements we concluded that Th-230 activity in gas mantles was less than the Th-228 activity. However, it was brought to our attention that high values could occurr for the activity ratio between Th-230 and Th-228 in thorium samples.

Dose calculations on the same model and assumptions for an extra 1000 Bq Th-230 activity in mantles result in an additional dose contribution of 0.007 mSv from inhalation and 2.5 10^{-5} mSv from oral intake of dust. It can be calculated that an additional 1000 Bq thorium-230 would increase the total per caput dose equivalent by an additional 5 to 10%.

4.7. COLLECTIVE DOSE EQUIVALENT FOR USERS OF GAS MANTLES

The calculated individual dose contributions, as already explained, are summarized in Table 3. In addition to average values, the possible ranges are presented.

On the basis of these dose estimates, the yearly collective effective dose equivalent can be calculated for users of gas mantles. It is known that on a yearly basis about 700.000 gas mantles are imported to the Netherlands and sold, 97% of them to campers. It is assumed that a camping unit consists of two persons using two mantles a year.

As summarized in Table 3, the main contribution to the collective dose is from inhalation of aerosols and is estimated to be in the range of 35-80 Sv per year.

The second large contribution results from inhalation of thorium oxide. With no Th-230 present about 17 Sv is estimated. An additional 1-2 Sv per year collective dose equivalent results from inhalation of radon and daughters together with ingestion of radioactive dust. External exposure is estimated as less than 0.02 Sv.

Altogether the collective effective dose equivalent for all users of gas mantles is estimated to be about 100 Sv per annum in the Netherlands. Dose estimates for only young gas mantles result in a range of from 30-76 Sv, while a range of values between 50-150 Sv has been calculated when only old thorium nitrate gas mantles were used. It should be evident that the accuracy of these figures should not be overestimated. The various results of dose calculations rely heavily on the assumptions made as the various parameters.

Of real importance is the relative value of this collective dose. For example, in relation to the collective effective dose equivalent from natural sources which can be taken as about 1200 Sv for the exposed group of 700,000 persons. The collective dose that results from the use of gas mantles is in the order of 5 to 10% of this.

TABLE 3

Dose estimates per caput and collective

	Dose equivalent per caput and gas mantle [in millisievert]			Collective dose [in Sievert]	
age of Th [years]	< 4	40	40	< 4	40
ventilation rate [per hour]	2	2	0	2	2
Inhalation of aerosols	0.025 (0.01-0.04)*	0.06 (0.025-0.09)*	0.2 (0.1-0.3)*	35 (14-56)*	80 (35-126)*
Inhalation of radon	0.001	0.0015	0.011	1.4	2.1
Inhalation of dust	0.024	0.026	0.033	16.8	18.2
Ingestion of thorium oxide	$1.7x10^{-4}$	$2.2x10^{-4}$	$2.2x10^{-4}$	0.12	0.15
External exposure	$< 1.3x10^{-5}$	$1.3x10^{-5}$	$1.3x10^{-5}$	0.02	0.02
Total	0.05 (0.03-0.07)	0.09 (0.05-0.13)	0.2 (0.1-0.3)	53 (30-76)	100 (51-150)

*range due to model assumptions

5. REMARKS

In the complete report of the study dose estimates are also given for transport and storage of large quantities of mantles. Collective dose contribution from external exposure is about 0.1 Sv per year. In abnormal circumstances, especially in the event of fire, relatively high individual doses from internal contamination are possible.

In general, no special measures are taken to regulate the disposal of used gas mantles. Even in a conservative approach it is estimated that local concentrations of thorium in domestic refuse cannot be more than 0.15 Bq Th-232 per kilogram, which is less than 1% of the "natural" concentration in soil (25 Bq per kilogram).

It is concluded that the uncontrolled removal of used gas mantles through the environment (soil) does not result in a significant increase of environmental radiation exposure.

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