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Radiological Assessment of Plutonium

Recommendation of the German Commission on Radiological Protection

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Radiologische Bewertung des Plutoniums

Empfehlung der Strahlenschutzkommission

In the event of any doubts about the meaning, the German original as published shall prevail.

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1 Dose factors and biokinetic behaviour of radionuclides

The radiological assessment of all radionuclides in their different chemical compounds starts out from the physical properties of these radionuclides and the behaviour of their chemical compounds in the biosphere and in the human organism. The most important aspects for this assessment are:

1. The kinds of radiation emitted by the radionuclide and their energies.
2. The physical half life.
3. The kind of the chemical compounds.
4. Distribution and transport in the biosphere.
5. Biokinetic behaviour in the human body (intake, change of distribution over time in the organs and excretions).

As far the radiological assessment with a view to man is concerned, external and internal radiation impacts have to be taken into consideration. External radiation impact is understood as the irradiation caused by a radiation source outside the organism, and internal radiation impact as that caused by an incorporated radioactive material.

The dose caused by an external radiation impact on man can be determined by direct measurements or from the distribution of the radionuclides in man's immediate environment. When determining the dose caused by an internal radiation impact, the complex processes of the intake of the radioactive material and its biokinetic behaviour have to be taken into consideration. On the basis of observations of humans and the results of animal experiments, biokinetic and dosimetric models have been developed for the calculation of dose factors which permit a uniform procedure in the assessment of the various radionuclides.

These dose factors were calculated for different radionuclides and their chemical compounds and for different conditions of exposures (inhalation and ingestion). With the aid of the dose factors and if the activity intake is known, it is possible to determine the radiation doses of the persons exposed for the corresponding conditions of exposure. It is thus possible to find out whether or not the dose limits have been adhered to in occupationally exposed persons and in the population. The secondary limits (annual limits of intake) have been derived with the aid of the dose factors.

The dose factors are constantly being reviewed on the basis of the latest state of the art. The experience of the past few years has shown that recent investigations have led in particular to a better confirmation of the existing dose factors. Major changes have only been necessary with respect to a few radionuclides.

2 The dose factors of plutonium and their assessment

Most of the isotopes of plutonium are alpha emitters, and some of them emit low-energy gamma and röntgen rays, although the probability of emission is small. Moreover, neutrons make a contribution to the radiation exposure.

The determination of the dose caused by an external radiation impact is possible with the aid of the usual methods of local and individual dosimetry.

For the assessment of the radiation impact by plutonium, the internal radiation impact is very important, as is the case with other alpha emitters as well, e.g. thorium and radium. What these natural radionuclides have in common with the plutonium isotopes which are of importance for radiological protection is that their physical half life and their residence time in the human organism are very long. Plutonium that is incorporated and mainly deposited in the human liver and bones leads to a long-standing irradiation of these organs. This long time of irradiation and also the high biological efficiency of the alpha rays have been taken into consideration in the dose factors so that the resulting radiation dose can be calculated if the incorporation is known.

The biokinetic data on which the new calculations of the dose factors are based have been determined not only in animal experiments but also in carefully evaluated cases of incorporation by humans. Although the data directly referring to humans show a biological spread as was expected, they do in the main confirm the biokinetic data on which the dose factors had previously been based.

Considering the dose factors for plutonium in detail they are found to be of the same order of magnitude as those of thorium and uranium in the case of inhalation. As far as ingestion is concerned, the dose factors for these elements are also of the same order of magnitude, although they are lower than those for inhalation by a factor of 100. The reason for the factor of 100 is found in the low intake of these radionuclides from the gastrointestinal tract into the blood. These findings consider recent results of the intake of plutonium from the gastro-intestinal tract and its distribution in the organism. The dose factor for inhalation which is relevant for occupational radiation exposure has not seen any greater change as a result of recent investigations.

3 Possible measurements for plutonium

The alpha rays emitted by the plutonium isotopes have a range of a few centimeters in the air and a range of some hundredths of a millimeter in the human organs. The direct measurement of the activity of plutonium is based on the detection of the characteristic röntgen radiation in the range of 13 to 17 keV which accompanies the alpha decay. Because of the low energy and the small probability of emission, this method is not sensitive enough for direct measurements such as they are required for regular monitoring. It can only be used for a plutonium activity of 300 Bq or more and is thus mainly suited for incidents. However, plutonium can be detected in the air inhaled, in contaminations at the workplace, in the working clothes, on the skin and in the correspondingly prepared human excretions. Thus, the situation with respect to the monitoring of possible incorporations is similar to that when handling chemicals the incorporation of which may involve a health hazard for the personnel.

The monitoring methods applied in the plants processing plutonium consider the particularities of plutonium in terms of measuring. For a regular check of the mean activity concentration in the air, air dust collectors are permanently installed at representative locations of the plants and are used for the daily registration of increases of activity in the air. In addition, monitors permitting direct reading and provided with alarms are installed at workplaces involving an increased risk of release so that increases in atmospheric activity can

be detected quickly. When leaving the controlled access area, all persons have to undergo a check by means of a hand-footclothes monitor. If the measurements of the air or the contamination measurements in the facial area reveal values above the intervention thresholds, further measurements of nasopharyngeal swabs and urine and stool samples are carried out on the following three days. In special cases, where an addition of gamma-emitting radionuclides to the plutonium-containing material being processed permits an indirect demonstration of the plutonium, the measurements are done with a whole body or lung counter.

Considering the biokinetic data, these incorporation and excretion measurements can be used to determine the intake of plutonium in the specially indicated cases. During the normal course of operations, the determination of the plutonium incorporation is effected on the basis of the measured values of air contamination. As a further confirmation of the incorporation monitoring, operational monitoring of the persons handling plutonium are carried out by means of regular urine and incorporation measurements. Thus, despite the particularities of plutonium in terms of measuring methods, there are sufficient possibilities to safeguard adherence to the limits.

For occupationally exposed persons in the Federal Republic of Germany, reference should be made to the assessments of the plutonium analyses of the past 11 years which have been carried out by the Karlsruhe Nuclear Research Center for its own personnel and for that of the Karlsruhe reprocessing plant (WAK) and of ALKEM. On the average, 900 persons were subjected to such monitoring. During this period, 11 cases of an exceeding of the annual limits of intake have been reported. In 98.7% of these cases, the annual intake was less than 5% of the annual limit of intake, and in 92.5% of the cases the activity intake was below the threshold of detection.

4 Radiation exposure of the population to plutonium

Measurements of Pu 238, Pu 239 + 240 and Pu 241 activities in tissue samples of persons of the normal population are available. The isotope relations measured clearly indicate that these plutonium activities originate from the fallout of nuclear weapons tests in the sixties and from the SNAP-9A satellite which burned out in 1964. For adults born before the beginning of the fallout period, these direct measurements of the plutonium activity in body tissues resulted in the following means of dose equivalents accumulated over 50 years (dose equivalent commitments) from plutonium 239 + 240:

Bone surface	1.7 mSv
Liver	0.38 mSv
Lungs	0.64 mSv
Red bone marrow	0.14 mSv
Other tissues	< 0.1 mSv

The additional dose contribution from Pu 238 and Pu 241 in the fallout is markedly lower. All in all, the resulting effective dose equivalent is about 0.2 mSv, accumulated over 50 years. This corresponds to approximately 1/500th of the mean background radiation to which the population is exposed over the same period of time.

As compared with this plutonium from fallout, the plutonium activity in man which is caused by effluents of nuclear facilities in the Federal Republic of Germany is considerably lower and cannot be detected by measuring methods. On the basis of the available measurements of plutonium effluents from nuclear power plants, the resulting activity and dose near these plants are expected to be lower than the contribution of plutonium from fallout by a factor of at least 10.

5 Results of epidemiological studies

In the United States and in the United Kingdom, investigations of plutonium-exposed collectives have been carried out. These collectives also include groups of employees where accidents have led to increased exposures to plutonium. As a result of the accumulations of plutonium in different organs, the incidences of the following tumors were of interest:

- lung tumors
- osteosarcomas
- liver tumors
- leukemias/lymphomas.

The results available so far indicate the following:

As compared with the mortality rates of the normal population, including those for tumor diseases, the mortality rates of the personnel in the nuclear industry are lower in almost all instances. This applies also to employees who have either incorporated higher activities of plutonium or are monitored for an incorporation of plutonium and other radionuclides. The reduced mortality with respect to the various causes corresponds to the frequently observed “healthy worker effect”. However, at several nuclear facilities the healthy worker effect is markedly less pronounced for the workers monitored for an incorporation of radionuclides than for the totality of all employees, i.e. on an internal basis, the relative mortality rates are higher for those who were subject to possible radionuclide incorporations. A causal relation to the incorporation of radionuclides cannot be ruled out, but does not follow either from these investigations, since the persons monitored for incorporation could also have been exposed to chemical carcinogens and may also differ from the rest of the employees in other aspects which have so far not been investigated sufficiently (e.g. smoking habits).

6 Conclusions

The above considerations show that the methods for the assessment of the radiation exposure to plutonium and of the radiation risk involved do not differ from those used with respect to other radionuclides.

For occupationally exposed persons, a careful planning and implementation of monitoring measures can ensure that the dose limits are not exceeded in the case of plutonium.

The long-term observations at nuclear power plants and other nuclear facilities in the Federal Republic of Germany show that the low plutonium emissions lead to a radiation exposure of the population in the environment of nuclear power plants which is far lower than that caused

by other radionuclides. The total dose as a result of all radionuclides from nuclear power plants and other nuclear facilities is small as compared with the range of variation of natural radiation.

The system of the limitation of doses such as it is contained in the present Radiological Protection Ordinance and in the 2nd Ordinance for the Amendment of the Radiological Protection Ordinance, applies to all nuclear facilities and thus also to reprocessing plants. This ensures that the radiation doses to which the population is exposed and which result from the normal operation of nuclear facilities are generally within the range of variation of natural radiation exposure.