

# **New Strategies for Assessment of Achieving the Release Criteria for Building Structures of NPPs with in-situ Gamma Spectrometry**

**L. Hummel, B. Sitte**

**TÜV Süddeutschland, Westendstr. 199, Munich, Germany**

This paper focuses on the decommissioning of Nuclear Power Plants (NPP). The release of building structures is emphasised. The measuring strategies dealing with these parts of a nuclear facility are of larger interest due to the fact that very high masses have to be handled and the measuring techniques differ significantly from those in a operating facility.

## **1 INTRODUCTION**

At the end of every decommissioning project cleared building structures and outdoor areas remain on the site ("green field option"). The rising number of nuclear facilities under decommissioning in Germany requires the development of concepts and methods for measuring procedures for the clearance of building structures under decommissioning.

In Germany several criteria of assessment concerning the decommissioning of nuclear facilities (especially NPP's) are existing. Examples therefore are the Recommendation of the German Commission on Radiological Protection concerning the „*Clearance of Materials, Buildings and Sites with Negligible Radioactivity from Practices subject to Reporting and Authorisation*“ (1) based on the IAEA „10 µSv-concept“ (safety series #89, (2)) and the „*Guide to the Decommissioning of Facilities as Defined in §7 of the Atomic Energy Act*“ given by the Federal Ministry for Environmental Protection (3). Several German standards like DIN 25457 („*Activity Measurement Methods for the Release of Radioactive Waste Materials and Nuclear Facility Components*“, (4)) deal with technical subjects of clearance measurements. The content of the basic formal and technical regulations and standards was described in several publications in the past. Therefore this point is neglected here.

Actually in Germany 14 large nuclear facilities are under decommissioning, nine of them are former nuclear power plants. Furthermore two fuel fabrication facilities and two nuclear power plants are already released from the German Atomic Energy Act.

The measuring strategies dealing with building structures and sites of a nuclear facility are of larger interest due to the fact that very high masses have to be handled and the measuring techniques differ significantly from those used in an operating facility. Especially in the past, it was common practice that building structures were released by analysing samples randomly taken from the surface or measuring the surface activity by large proportional counters. Recently in-situ gamma spectrometry became a major part in release measurements.

The substantial part of the work presented in this paper was performed within the scope of a R&D-project funded by the Federal Ministry for Education, Science, Research, and Technology and the State Ministry for Regional Development and Environmental Protection of Bavaria (5).

The „Hot-Steam Reactor Großwelzheim“ (HDR), which is -after the Nuclear Power Plant Niederaichbach (KKN)- the second Bavarian NPP released from the German Atomic Energy Act in 1998 leaving a „green field“ on the site.

It is practice in Germany, to split up the licensing process of the decommissioning of a facility into several steps. With respect to the release of building structures and site the last licenses are of importance. Subject of the final license for HDR was the release of buildings from the Atomic Energy Act after performing release measurements and subsequent dismantling by conventional techniques. The gained experiences are discussed from the point of view of a Technical Support Organisation (TÜV) with the particular task to perform independent measurements to assess the radiological state of the facility.

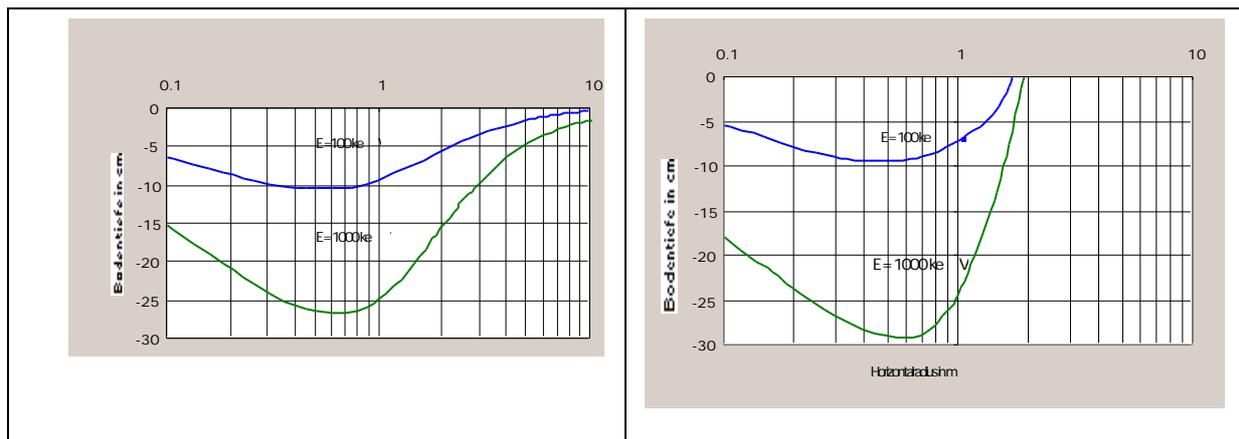
Before two concepts for the use of in-situ spectrometry in NPPs will be described, the measuring device itself is presented.

## 2 IN-SITU GAMMA SPECTROMETER FOR THE USE IN NUCLEAR FACILITIES UNDER DECOMMISSIONING

The “traditional” methods for the clearance measurements of building structures of nuclear facilities under decommissioning are direct measurements with a contamination monitor combined with taking samples and smear tests with subsequent laboratory analysis. Because of their very low sensitivity to  $\gamma$ -radiation, contamination monitors are not able to detect migrated activity suitable. On the other hand the detailed information gained by the analysis in the laboratory is only valid for a very small fraction of the structure under investigation. So the results of the contamination monitor measurements performed in a large scale were recalibrated with the results from laboratory.

The predominant part of measurements in laboratory is gamma spectrometry, therefore it is a more effective strategy to bring the detector to the source than taking a sample of unknown representativity and analysing it in laboratory with high effort. Besides that, the use of in-situ spectrometry for clearance measurements has more advantages:

- The area covered by the measurement is much higher than following a sampling strategy or contamination monitor measurements in a grid.
- The activity ratios of the main gamma emitting nuclides can be verified permanently.
- The artificial part of the activity can be separated easily from the natural background.
- The measuring results due to gamma radiation are nearly independent from the migration of the nuclides into the concrete, compared to beta-sensitive contamination monitors.



**Figure 1: Calculated three-dimensional “field of view” of the uncollimated (left) and the collimated (right) spectrometer as described below one meter above the surface, if a homogeneous distribution of activity is assumed and 90% of the total photon flux is taken into account. The x-axis is the distance from the centre of the “field of view” in meter, the y-axis is the depth relative to the surface in centimetres.**

Due to these advantages in situ gamma spectrometry became a major part in release measurements in the last years. Already in 1992 the IAEA stated (9) that, especially for large areas outside the buildings in situ gamma spectrometry may be the only method of achieving validation of the release criteria.

The leading systematical error is the unknown distribution of localised activity, if this must be assumed<sup>1</sup>. In earlier times in situ gamma spectrometry has been applied in the field without any collimation to determine e.g. the surface contamination in fall-out areas. For measurements inside buildings we generated strategies for clearance measurements with uncollimated in situ spectrometry. For the identification of localised contamination the detector’s “field of view” has to be reduced with a collimator. The impacts of a collimator can be visualised by plots like Figure 1. The data were calculated by (10).

<sup>1</sup> A strong inhomogeneity in the distribution of the contamination leads to large systematical errors in every measuring strategy.

The measuring device consists of a 44% p-type “Extended Range” HP-Germanium detector which is connected to a portable Multi-Channel Analyzer and a notebook to control the detector-electronic system. For collimated in situ measurements the detector is built in a special measuring device, which is shown in figure 2. Due to the rotatable detector the device enables measurements of walls and floors inside building structures. The height of the detector from the floor is adjustable in four positions.

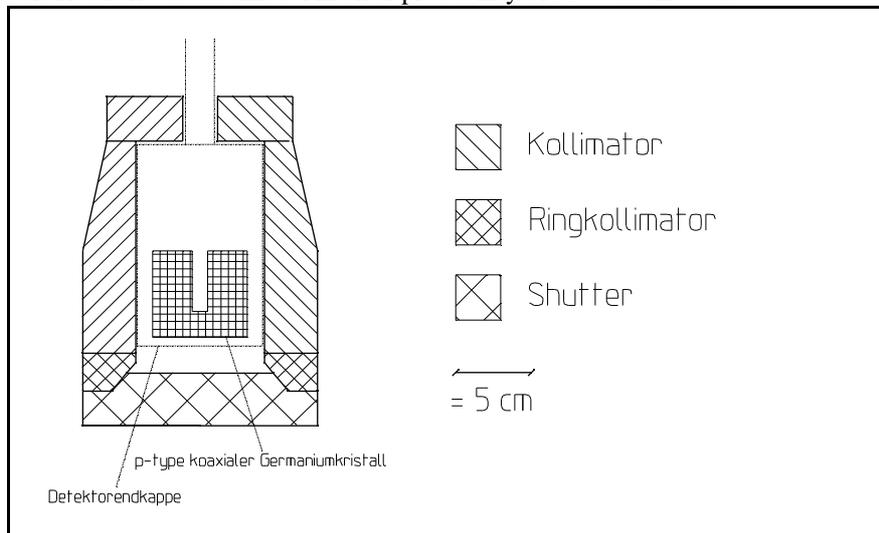
The collimator is made of sintered tungsten to obtain an optimum shielding-mass ratio. Figure 3 shows a drawing of the cross section of the collimator. The construction of the collimator allows two different field of views (“Kollimator” and “Ringkollimator”). Combined with the different heights of the detector the system provides averaging areas between 1 and 8 m<sup>2</sup>. In case of strong irradiation from outside the field of view a plug (“shutter”) can be added to perform a differential measurement.

In addition figure 3 shows one of the detector crystals we use. For uncollimated in-situ measurements, it is profitable to use crystals without any dependence on direction, leading to the need of nearly equal values for length and diameter of crystal.

The system satisfies the demands for release measurements referring to practical and technical aspects and was calibrated according to the standard method (6), which was qualified within the scope of our former research work (7) for collimated systems. Table 1 shows the minimum detectable activity (MDA) for the collimated in-situ gamma spectrometer for different activity distributions. The values were calculated from measurements in buildings with low contamination and with a normal background dose rate. The geometry of collimation was the “Ringkollimator” (see figure 3), a measuring time of 15 minutes and a horizontal homogeneous distribution was assumed. An error probability of 5% was allowed.



**Figure 2: Collimated in situ gamma spectrometer developed in the framework of (5).**



**Figure 3: Drawing of the cross section of the in-situ gamma spectrometer showing the dimensions of the crystal, the cap and different parts of the collimator.**

**Vertical distribution of activity in the source**

Nuclide	Exponential =1 g/cm <sup>3</sup> [Bq/cm <sup>2</sup> ]	Homogeneous [Bg/g]
Co-60	0.013	0.0015
Cs-137	0.022	0.0025
U-235	0.024	0.0037

**Table 1: Brief survey of attainable MDA's of the collimated in-situ spectrometer. For the assumed conditions refer to text.**

### 3 CONCEPTS FOR THE APPLICATION OF IN-SITU GAMMA SPECTROMETRY

#### 3.1 INTENSITY OF MEASUREMENTS DEPENDING ON THE RADIOLOGICAL STATE OF THE STRUCTURE (DECOMMISSIONING OF HDR)

##### 3.1.1 Basics

The HDR plant showed contaminations in parts of the building structures and the site which have been removed before release measurement took place. The total inventory of the plant before decontamination was estimated to be in the order  $10^{10}$  Bq. The decontamination procedures reduced the inventory significantly. In total 28.000 Mg of concrete, 31.000 m<sup>2</sup> of steel and other surfaces and more than 6.000 m<sup>2</sup> outside areas had to be released.

The licensing authority contracted TÜV as Technical Support Organisation as assistance of the decommissioning process. The duties were e.g. the assessment of the licensees documentation and to carry out independent control measurements. The range of these measurements covered the biological shield as well as the meadows along the perimeter of the site. For the first time in-situ gamma spectrometry was implemented in the complete measuring strategy as a diverse measuring technique.

##### 3.1.2 Clearance Levels

The evaluation of the clearance levels for the unrestricted release of materials and the site based on the „10 µSv-concept“ by the IAEA. Based on this concept the clearance levels are depending on the nuclide vector. Therefore the nuclide vectors were determined for the different materials/areas of the plant from representative samples before licensing. After the evaluation of the nuclide vector the clearance levels were related to mass- or surface-specific activities of the dominating nuclide in each case and used as activity limits during the decommissioning process (Table 2).

0,47 Bq/cm <sup>2</sup>	-activity	concrete; reactor building
0,038 Bq/g	Eu 152	concrete; biological shield
0,5 Bq/cm <sup>2</sup>	-activity	steel (not-activated) and sealed outdoor grounds
0,03 Bq/g	Cs 137	unsealed outdoor grounds; max. 0,01 Bq/g Co 60; 0,02 Bq/g Cs 137 may be subtracted (Chernobyl)

**Table 2: Summarised clearance levels (abridged version):**

##### 3.1.3 Measuring Techniques and Strategies

The performed routine activity measurements will be explained below. Table 3 shows the measuring techniques used.

	Decommissioner	TÜV # Building / # Site
Samples, Gamma Spectrometry	✓	✓ 350 / 140
Contamination Monitor	✓	✓ 11.000 / 120
Smear Tests	✓	✓ 600 / 0
in-situ Gamma Spectrometry		✓ 850 / 720

**Table 3: Measuring techniques used by the decommissioner and as control measurements. The numbers indicate the rounded sum of performed measurements.**

The focus of the following chapter lies on the in-situ technique due to the fact that these measurements are not common yet. The other routine measurements were used without special modifications.

For the first time in-situ gamma spectrometry was used during the entire measuring campaigns accompanying the decommissioning process. In the past in-situ gamma spectrometry was used in several, short-term (max. some weeks) measuring campaigns in the framework of the surveillance of nuclear facilities (especially decommissioning projects). The use of the collimated in-situ spectrometry in decommissioning projects was investigated in a project founded by the EC (7). Actually a R&D-project (5) covering the whole subject of the use of different release measurement techniques in the buildings of nuclear facilities is in a late stage.

In-situ gamma spectrometry allows nuclide-specific measurements in the proper place. Figure 3 shows the cross section of one of the two collimated in-situ spectrometers used during the measurements in HDR. Without a collimator the total photon flux from the surrounding building structure is detected. A special calibration technique was necessary to estimate the average surface-specific activity. If the depth distribution of activity in the structure is known, the spectrometer can be calibrated to measure surface- resp. mass-specific activities. Concerning the depth distributions used inside the reactor building of HDR it was shown by sampling in fine layers that the assumed distribution was conservative.

For determining the measuring strategy of the control measurement it was assumed that all relevant contaminations has been removed during the decontamination process. Therefore localised areas with high contamination levels or large areas with raised contamination level have not been expected during the measurements.

**The first stage measurements** were one or a set of uncollimated in-situ measurements depending on the extent and geometry of the considered building structure inside the plant.

As a result of these measurements two criteria had to be met:

- ❶ Keeping of the clearance level averaged over the entire structure
- ❷ Keeping of a „hot-spot-criterion“ depending on the nuclide vector

The second criterion was in the range of several 10 kBq, evaluated from radiological reasons. Under normal circumstances (room geometry) this criterion was infringed prior to the first. In practice the first criterion was kept in every case. By keeping both criteria it was stated that there are no indications concerning activities infringing the clearance levels.

Only if the second criterion was infringed, more detailed measurements were performed as a **second stage**. The following points show several possibilities to choose from in the actual situation:

- Performing several uncollimated measurements to prove „homogeneous activity distribution“.
- Collimated measurements to estimate the mid-scale activity (eff. area: some m<sub>2</sub>).
- Measurements with contamination monitors to localise remaining small area contaminations.
- Sampling to verify the assumed depth distribution.

Besides the discussion above, the use of in-situ spectrometry for release and control measurements has more advantages:

- The area covered by the measurement is much higher than following a sampling strategy or contamination monitor measurements in a grid.
- The activity ratios of the main gamma emitting nuclides can be verified permanently.
- The artificial part of the activity can be separated easily from the natural background.
- The measuring results due to gamma radiation are nearly independent from the migration of the nuclides into the concrete, compared to beta-sensitive contamination monitors.

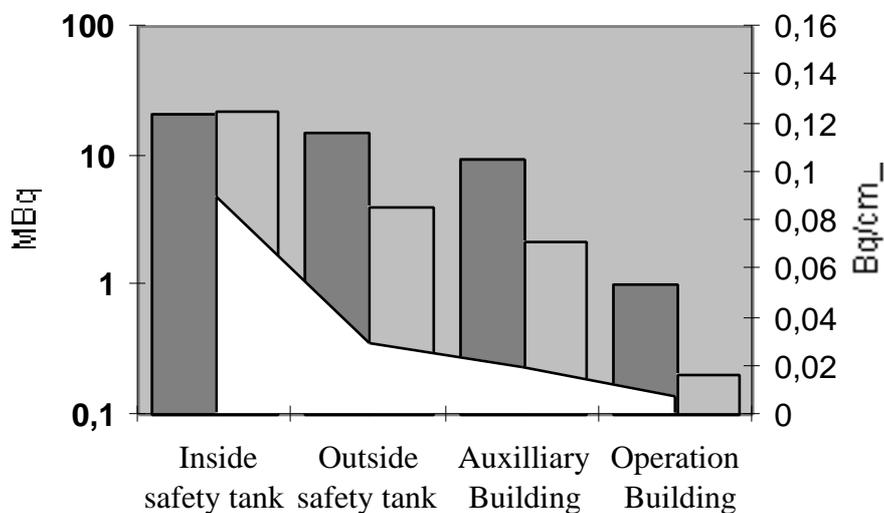
In the PC-based documentation accompanying this project the total masses, surfaces and activity levels gained with the different measuring techniques were recorded. Figure 4 shows a compressed summary of the total artificial inventory of the building and the average contamination level. The decreasing averaged activity level with increasing distance to the former reactor vessel as plotted in Figure 4 is self-explanatory.

The discussion of the remaining total activity and especially the differences between the measuring techniques is a little more complicated. To understand the result three points must be kept in mind:

- Systematically occurring errors which have no tendency are vanishing comparing a data set of many single results. Therefore only systematic errors with a tendency to over- or underestimate the true value appear in this graphic representation.
- The detection limits of the contamination monitors are higher than those of the in-situ gamma spectrometer. If no activity was detected, the detection limits were summed up.
- As long as contaminations are located on the surface only, the activity will be overestimated due to the conservative calibration procedure of the contamination monitor. On the other hand concrete with activity migrated into the structures (e.g. mechanical decontaminated structures) will tend to be underestimated with contamination monitors due to the short ranged beta radiation.

So it can be concluded that there is a break even point where both measuring techniques are leading to the same results. In this context, the good agreement of results for the reactor building inside the safety tank is accidental.

In areas with remaining activity of a higher level (0.2 Bq/cm<sub>2</sub> or higher) the in-situ results were above the contamination monitor values generally. In contradiction to that the summing of detection limits with contamination monitors led to an overestimation of the remaining activity in the most cases.



**Figure 4: Total artificial inventory [MBq] of the building structures (bars) measured by the decommissioner with contamination monitors (left) and during the control measurements with in-situ gamma spectrometry (right). The area plot shows the averaged contamination level [Bq/cm<sub>2</sub>] remaining in the structure decreasing with the „distance“ from the former reactor vessel.**

The procedure of performing a two-staged measuring concept has one great advantage. The expenditure depends on the existing radiological state of the structure. The number of necessary measurements is decreasing with the difference between measured activity value and clearance level. The control measurements upon the building structures we performed for the relevant authority during the decommissioning of HDR-power-plant were basing on this concept and led to a efficient measuring strategy. For a more detailed discussion see (11).

### 3.2 FREE RELEASE BY MEASURING THE DOSE RATE

A complete new strategy for achieving a free release of building structures is mainly based on uncollimated in-situ measurements inside the building. In connection with this concept, it must be kept in mind, that this strategy has the stage of a “contribution for discussion” and was never applied during the release procedure of a building structure in Germany.

The first step in the release procedure normally defines clearance levels referring to activity values (e.g. Bq/cm<sub>2</sub>). The fundamental principle generally is, a criterion depending on the tolerable effective dose like in (2). For nuclear power plants and their typical nuclide vectors, it can be shown that the later use for residential purposes *would* lead to the lowest clearance levels and the exposition *would* be clearly dominated by direct irradiation. Why don't we measure directly the relevant quantity dose-rate, as the dominant contribution to the effective dose?

The calculated dose rate for this unlikely, but most restrictive scenario is extremely low (~1 nSv/h). But this value can be measured nuclide-specific with specially calibrated, portable germanium detectors with tolerable effort (see below).

This strategy will now be compared with the most probable proceeding for release measurements for buildings in Germany in the future (release criteria according to (8) after implementation into the German RPO in May 2000). Therefore a realistic nuclide vector from a German decommissioning project is chosen, reduced to the radiological relevant nuclides and calculated for the clearance levels, which have to be kept (table 4).

<b>Fe-55</b>	34.395	I	II	III	C											
<b>Co-60</b>	<b>16.995</b>	<i>0,36</i>	<i>2,9</i>	<i>0,089</i>	92,6%											
<b>Ni-63</b>	40.191															
<b>Sr-90</b>	0.022	<i>34</i>	<i>34</i>	<i>1,5</i>												
<b>Y-90</b>	0.022															
<b>Cs-137</b>	<b>4.504</b>	<i>1,5</i>	<i>12</i>	<i>0,40</i>	7,4%											
<b>Eu-154</b>	0.054															
<b>U-234</b>	0.004	<i>1,4</i>	<i>11</i>	<i>0,36</i>		<table border="1"> <tbody> <tr> <td></td> <td><i>[Bq/cm_]</i></td> </tr> <tr> <td><b>I</b></td> <td><i>0,44</i></td> </tr> <tr> <td><b>II</b></td> <td><i>3,6</i></td> </tr> <tr> <td><b>III</b></td> <td><i>0,112</i></td> </tr> <tr> <td></td> <td><i>[Bq/g]</i></td> </tr> </tbody> </table>		<i>[Bq/cm_]</i>	<b>I</b>	<i>0,44</i>	<b>II</b>	<i>3,6</i>	<b>III</b>	<i>0,112</i>		<i>[Bq/g]</i>
	<i>[Bq/cm_]</i>															
<b>I</b>	<i>0,44</i>															
<b>II</b>	<i>3,6</i>															
<b>III</b>	<i>0,112</i>															
	<i>[Bq/g]</i>															
<b>U-238</b>	0.005															
<b>Pu-238</b>	0.176	<i>0,3</i>	<i>2,4</i>	<i>0,077</i>												
<b>Pu-239</b>	0.21															
<b>Pu-241</b>	2.919	<i>11</i>	<i>92</i>	<i>3,0</i>												
<b>Am-241</b>	0.485	<i>0,34</i>	<i>2,8</i>	<i>0,091</i>												

**Table 4: Typical nuclide vector for a NPP under decommissioning and the distribution of activity in % (2<sup>nd</sup> column). Surface-specific<sup>2</sup> (Column I&II, [Bq/cm<sub>2</sub>]) and mass-specific (Column III, [Bq/g]) clearance levels according to (8). Column I will be used, if the building may be reused in the future, column II may be used, if the building will be demolished and column III is for building rubble arising during the decommissioning process. Column C gives the weighted contribution to the sum formula<sup>3</sup>, if Column II is used as data-set.**

It was a result of a radiological investigation (5), that the maximum averaging areas for release measurements have a strong dependence on the nuclide vector and the path of release. For the vector used here an averaging area of 10 m<sub>2</sub> can be used. The tolerable total activities (italics in table 5) of the leading nuclides were calculated by multiplying these areas with the nuclide-specific clearance level. These can be compared with the achievable detection limits of the uncollimated<sup>4</sup> and collimated in-situ gamma spectrometry in chapter 2.

[Bq/cm <sub>2</sub> ] <i>[kBq]</i>	Co-60
I	<i>0.34</i> <i>&lt;30</i>
II	<i>2.8</i> <i>&lt;250</i>

**Table 5: Surface-specific and total activity (in italics, calc. with the averaging area mentioned above) for the leading nuclide as a function of the path of release (I: reuse, II: demolition).**

<sup>2</sup> The surface-specific activity must be interpreted as the projection of the total migrated activity upon the surface divided by the unit area.

<sup>3</sup> Sum formula:  $\frac{A_i}{CL_i} \cdot 1$ , where  $A_i$  is the nuclide specific distribution of activity and  $CL_i$  the corresponding clearance level. All nuclides which contribution to the sum formula is less than 10% may be neglected. Therefore only two nuclides remain in Column C.

<sup>4</sup> According to the much larger solid angle radiation is detected the detection limits are lower than for collimated measurements.

According to Table 1, the collimated in-situ spectrometer we use is able to detect 15 mBq/cm<sub>2</sub> Co-60 in 15 minutes or less in the uncollimated case. This is equivalent with a monoenergetic (unscattered) photon flux in the order of 0.005 /cm<sup>2</sup>s or an additional dose-rate of approx. 0.15 nSv/h. This photon flux is also created from a localised source with an activity of 30 kBq Co-60 (table 5) in a distance of several meters.

So, there is a perspective for a simple monitoring programme with small systematical errors leading to a free release of the building structure. For practical applications two disadvantages must be considered.

- On the one hand the remaining contamination level of the building structures *must* be low for using this strategy effectively  
*(consider as a simple case a cubic room with the detector in the centre with a homogeneous surface contamination, then this contamination must have a value of ~4 mBq/cm<sub>2</sub> (1.2% of the clearance level!), to give the same photon flux),*
- on the other hand, large-scale measuring techniques like this one, focus on the problem of defining a radiologically tolerable quantity of potential existing, localised activity, which must be detected during the monitoring programme  
*(see discussion above).*

In most cases especially the latter point is not taken into account, when release measurement strategies are defined. The work started in (5) is a starting point to solve this point, but additional research work is necessary before this strategy may be established as a qualified release procedure. Especially the conservative value for the integral activity of 30 kBq Co-60 may be enlarged by an detailed radiological investigation.

## REFERENCES

- 1 German Commission on Radiological Protection:  
*Clearance of Materials, Buildings and Sites with Negligible Radioactivity from Practices subject to Reporting and Authorisation*  
Submitted Feb. 12<sup>th</sup>, 1998
- 2 International Atomic Energy Agency:  
*Principles for the Exemption of Radiation Sources and Practices from Regulatory Control*  
Safety Series 89, Vienna, 1988
- 3 Federal Ministry for Environmental Protection:  
*Guide to the Decommissioning of Facilities as Defined in §7 of the Atomic Energy Act*  
Submitted June, 14<sup>th</sup>, 1996
- 4 DIN 25457:  
*Activity Measurement Methods for the Release of Radioactive Waste Materials and Nuclear Facility Components*  
Beuth-Verlag, Berlin, 1999
- 5 *Weiterentwicklung der in situ Gammasppektrometrie* (final report of a R&D-project, in preparation)  
Funded by the *Federal Ministry for Education, Science, Research, and Technology* and the *State Ministry for Regional Development and Environmental Protection of Bavaria*.
- 6 Beck, H.L. et al  
*In Situ Ge(Li) and Na(Tl) Gamma-Ray Spectrometry*  
Report HASL-258 (U.S.D.o.E., Environmental Measurements Laboratory, New York)
- 7 European Commission:  
*High Resolution in-situ gamma spectrometer for use on contaminated building structures and outdoor grounds under decommissioning*  
Report EUR 18349, 1998
- 8 European Commission:  
*Recommended radiological protection criteria for the clearance of buildings and building rubble arising from the dismantling of nuclear installations*  
Draft Proposal; Version: 6, March 1999
- 9 International Atomic Energy Agency:  
*Monitoring Programmes for Unrestricted Release related to Decommissioning of Nuclear Facilities*  
Tech. Rep. Series No. 334, Vienna 1992
- 10 C. Brummer  
*Personal Message*  
BfS, Neuherberg, Germany, 1999
- 11 Buß K., Hummel L.  
*HDR: Freigabe von Gebäudeteilen und Geländeflächen*  
atw, Internationale Zeitschrift für Kernenergie, November 1998