Establishing one set of scaling factors from a series of measured activity concentrations

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Abstract. For clearance measurements it is possible, under some circumstances, to calculate the activity concentrations of difficult-to-measure radionuclides from the measured activity concentration of a key nuclide that is more straightforward to measure. This is done by multiplying the activity concentration of the key nuclide by scaling factors. In cases with many objects to be measured, it is preferable to have only one set of scaling factors to be used for all the objects.

The paper explains a method for establishing a single set of scaling factors from a series of activity concentration measurements on samples from the former Hot Cell facility at the Risø DTU site in Denmark. This facility has mainly been used to examine spent fuel samples and is contaminated with fission products and isotopes of uranium and transuranic elements.

It is shown that the distribution of the measured activity concentration ratios can be described by normal distributions. It is discussed how to identify and deal with outliers. Conservative values of the activity concentration ratios to be used as scaling factors in the clearance measurements on objects from the Hot Cell are calculated from the normal distributions. Standard uncertainty values are assigned to the scaling factors.

KEYWORDS: clearance, scaling factor, nuclide vector, spent fuel, transuranic elements, Hot Cell, decommissioning.

1. INTRODUCTION

Danish Decommissioning is the state owned company responsible for decommissioning the nuclear facilities which are situated at the Risø-DTU site. At present two research reactors have been decommissioned. It is part of the company decommissioning philosophy to try to clear as much waste as possible for unconditional use. An infrastructure for clearance measurements within Danish Decommissioning has been established under the name of *the Clearance Function* (CF) (Søgaard-Hansen et al. 2008). This function develops the procedures used for the clearance measurements and performs the measurements. Mass concentrations of radionuclides are measured by γ-spectroscopy while surface contamination is measured with surface contamination monitors. In 2007 the CF obtained accreditation from The Danish Accreditation and Metrology Fund (DANAK).

The present accreditation does not allow measurements of non-γ-emitters unless they are calculated from the concentration of γ-emitters using neutron activation calculations or they can be measured with surface contamination monitors. This has not been an obstacle in the clearance of materials from the two decommissioned research reactors. With the former Hot Cell facility, it is necessary to determine bulk concentrations of non-γ-emitters by using scaling factors that have been obtained from a series of concentration ratio measurements.

2. CLEARANCE CRITERIA AND COMPLIANCE

Materials can be cleared if the sum of nuclide activity concentration, C_i, to clearance level, CL_i, ratios is below unity. In practice this criterion must be met using some specified rules of compliance. The CF uses Eqn. 1, where $u(C_i)$ is the standard uncertainty on the concentration value C_i.

$$\sum_i \frac{C_i}{CL_i} + 1.65 \cdot \sqrt{\frac{u(C_i)^2}{CL_i^2}} < 1 \qquad (1)$$

Complying with Eqn. 1 will clear a given item and ensure (assuming normal distributed measured activity concentrations) that the probability that the sum of true activity concentration to clearance level ratios is less than 1, is at least 95%.

For a radionuclide that can be present but shows no measurable activity, the critical value of activity concentration determined by the measurement conditions is used as an activity concentration for this nuclide, and the uncertainty of determining the critical value is used as uncertainty.

If the activity concentration of a nuclide is difficult to measure but can be obtained from the concentration of a more easy-to-measure nuclide by multiplying this concentration by a factor (scaling factor), the resulting concentration is used and the uncertainty in the scaling is included in the uncertainty calculation. When scaling factors are used in clearance measurements, some degree of conservatism must be included in the determination in order not to underestimate activity contents.

3. FROM ACTIVITY CONCENTRATIONS TO SCALING FACTORS

The Hot Cell facility comprises six concrete cells separated by doors and shutters. The cells have been used to examine spent nuclear fuel and handling ^{60}Co sources. The examination of the fuel included cutting cross sections in the fuel which contaminated some of the cells heavily. The operation of the Hot Cell facility stopped in 1989 and almost all equipment outside the cells was dismantled and removed along with equipment from the cells. Some decontamination was made a few years later, but otherwise the facility was sealed. As the contamination of most items is a mix of contamination from more than one cell it would be preferable only to have one set of scaling factors (one scaling factor for each nuclide) valid for all objects. The difficult-to-measure radionuclides are ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{242}Cm, ^{243}Cm and ^{244}Cm. Uranium isotopes will also be present in the contamination but in activity concentrations insignificant compared to the other nuclides. The scaling factors for the nuclides are relative to the easy-to-measure radionuclide ^{137}Cs.

3.1 PRIMARY DATA

19 paint samples were taken from the cells. Samples were taken from all cells. Seven samples (one from each cell, one cell being a double cell) were taken from inner surfaces of plugs in the front walls of the cells. Six samples (one from each separate ventilation system) were taken from the far end of the ventilation pipes inner surfaces. Six samples were taken from the surfaces of the shutters between each cell. The samples were obtained by scraping off the paint with a metal tool. Sample weights were in the range 0.2 - 1 g.

The Radiation Research Programme (NUK) at Risø-DTU did the radiochemical processing and analysis on the samples. NUK performed most analyses on subsamples of the individual samples. PIPS detectors, GM detectors and ICP-MS were used as measuring techniques. The CF measured the samples for the content of ^{137}Cs and ^{241}Am with γ-spectroscopy.

3.2 EXTRACTING SCALING FACTORS

A priori all samples were considered to be equal in terms of information, because they were selected to be so and they were processed equally and had approximately identical relative uncertainty on the activity concentrations. Activity concentration ratios were calculated and their distribution compared to a normal distribution with mean and standard deviations taken as the mean of the ratios (μ) and the experimental standard deviation of the ratios (σ). The comparison was made by visually comparing data in a normal quantile plot. The plot is made in the following way. At first the measured *n* ratios are sorted in increasing order. For the *j*'th ratio, x_j, the P_j quantile is calculated as $\frac{j-0.5}{n}$. For this value the inverse cumulative probability distribution function is calculated (qnorm(P_j,0,1)). In the normal quantile plot the qnorm(P_j,0,1) values are plotted against the observed x_j ratios, and the coordinate pairs should fall approximately on the straight line given by the equation: $y = \frac{x}{\sigma} - \frac{\mu}{\sigma}$. If the underlying distribution is a normal distribution it is used to extract the scaling factor. For the plutonium and curium isotopes the scaling factors was obtained as a product of activity concentration ratios. This was done because of a small discrepancy found in the measured ^{241}Am concentration by NUK and by the CF. The scaling factor for an isotope, *PC*, was calculated as:

$$SF_{PC} = \frac{PC_{\mathrm{NUK}}}{^{241}\mathrm{Am}_{\mathrm{NUK}}} \cdot \frac{^{241}\mathrm{Am}_{\mathrm{NUK}}}{^{241}\mathrm{Am}_{\mathrm{CF}}} \cdot \frac{^{241}\mathrm{Am}_{\mathrm{CF}}}{^{137}\mathrm{Cs}_{\mathrm{CF}}} \qquad (2)$$

The strontium isotope ^{90}Sr

The ratios of nuclide activity concentration to the concentration of ^{137}Cs were calculated for the samples and some simple statistics of the ratios were made. An extreme value = 0.74 (outlier) was identified because the numerical difference from the median value was greater than 1.5 times the difference between the 75 percentile and the 25 percentile. The outlier was not discarded as this would have lowered the scaling factor and therefore not be a conservative act. With the outlier included, the mean value was 0.28 and the experimental standard deviation was 0.16. The standard deviation corresponds to a relative standard deviation of 40%. This is a larger value than expected from a calculated standard deviation based on the assigned relative standard deviations on the activity concentration measurement (10% for both ^{90}Sr and ^{137}Cs) which gives 15%. The difference can be ascribed to the variation in the Hot Cell environment itself. Figure 2 shows the normal quantile plot of the ^{90}Sr to ^{137}Cs ratios. Apart from the outlier all coordinate pairs are approximately on a straight line.

The scaling factor for the ^{90}Sr activity concentration relative to the activity concentration of ^{137}Cs is defined as the ratio value, which will give a value of 0.95 in the cumulative probability distribution of the normal distribution with the mean and standard deviation of the measured ratio values. This definition will ensure that 95% of the ratios will be below the scaling factor value or that the probability that a measured ratio is less than the scaling factor value is 95%. The standard uncertainty on the scaling factor is taken to be the experimental standard deviation on the measured ratios. The scaling factor value is therefore: $SF_{\mathrm{Sr90}} = 0.54$ with a standard uncertainty of 0.16.

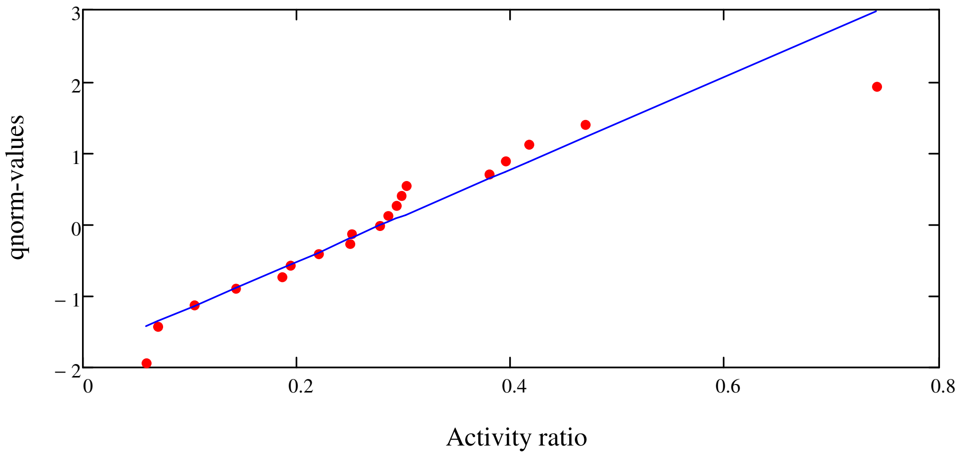

Fig. 2 Normal quantile plot of the ^{90}Sr to ^{137}Cs ratios.

The americium isotope ^{241}Am

The analysis was made in the same way as for ^{90}Sr. No outliers were found. The mean value was 0.042 and the experimental standard deviation was 0.031. The standard deviation corresponds to a relative standard deviation of 74%. This is a larger value than expected from a calculated standard deviation based on the assign relative standard deviations on the activity concentration measurement (10% for both ^{241}Am and ^{137}Cs) which gives 15%. The difference can be ascribed to the variation in the Hot Cell environment itself. The scaling factor is: SF_{Am241} = 0.093 with a standard uncertainty of 0.031. Figure 3 shows the normal quantile plot of the ^{241}Am to ^{137}Cs ratios. All coordinate pairs are approximately on a straight line.

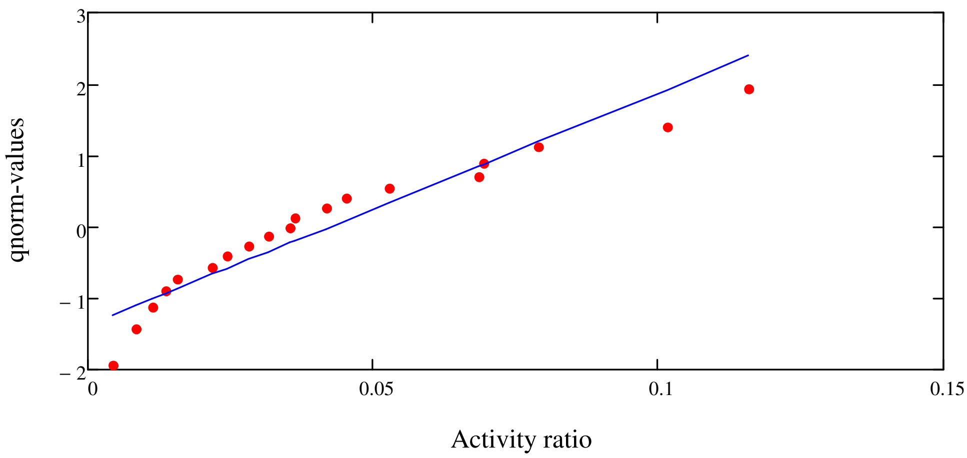

Fig. 3 Normal quantile plot of the ^{241}Am to ^{137}Cs ratios.

Plutonium and curium isotopes

In order to use the products of ratios as explained above, it was investigated if the contents of plutonium and curium isotopes correlated with the content of ^{241}Am in the samples (all values measured by NUK). Figure 4 shows a plot of the plutonium and curium activity concentrations values against the ^{241}Am activity concentration in the samples and good correlations are observed for all isotopes. The isotopes ^{243}Cm

and ^{244}Cm are considered to be one isotope as are the isotopes ^{242}Cm and ^{242m}Am. All coefficients are greater than 0.990 and all but one are greater than 0.999.

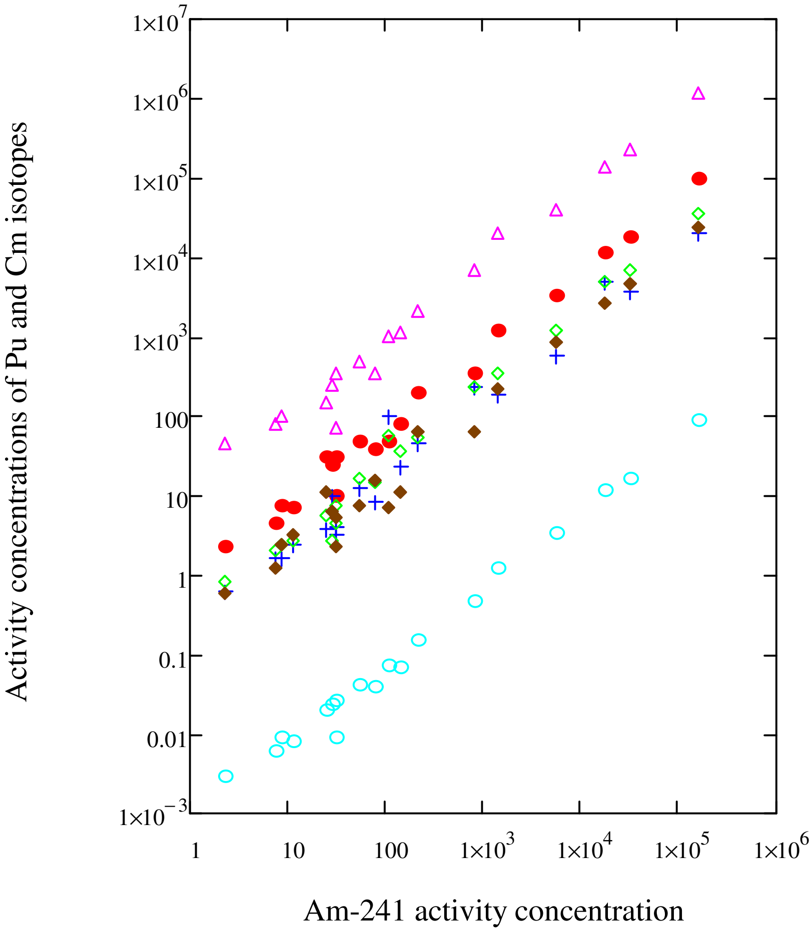

Fig. 4 Activity concentrations of plutonium and curium isotopes in the samples plotted versus the ^{241}Am activity concentrations. All values determined by NUK.

The distribution of the ratios of ^{241}Am concentrations measured by NUK and the CF, respectively, was examined. The distribution of the ratios is described by a normal distribution, with a mean of 0.58 and an experimental standard deviation of 0.33. The standard deviation corresponds to a relative standard deviation of 57%. An outlier value of 1.5 was found, but not discarded as it would decrease the mean of the ratios (decreasing scaling factors) if excluded. The mean is larger than expected from a calculated standard deviation based on the assigned relative standard deviations on the activity concentration measurement (~5% for NUK's measurements and ~10% for the CF measurements), which gives ~11% for the ratios. The difference can be due to the fact that the CF measurements were made on total samples whereas the NUK measurements were made on sub-samples. Figure 5 shows the normal quantile plot of the ^{241}Am ratios. All coordinate pairs except for the outlier lie approximately on a straight line.

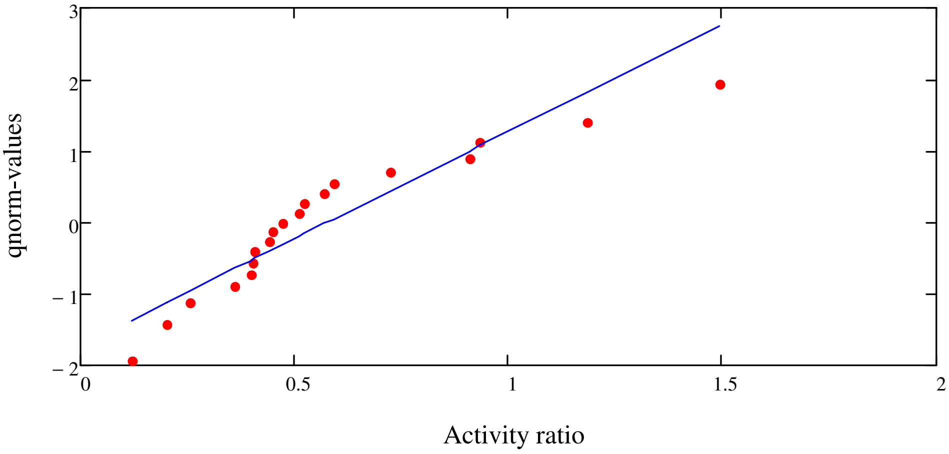

Fig. 5 Normal quantile plot of the ^{241}Am (NUK) to ^{241}Am (CF) ratios.

A ratio to be used in Eqn. 2 is chosen as the ratio, which in 95% of all measurements will be larger than a measured ratio (similar to the above extraction of scaling factors). The ratio is 1.12 with a standard uncertainty of 0.33.

The isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, (^{243}Cm + ^{244}Cm) and (^{242}Cm + ^{242m}Am).

The ratios of ^{238}Pu activity concentration to the concentration of ^{241}Am (measured by NUK) were calculated and analysed. No outliers were found. The mean value for the ratios was 0.71 and the experimental standard deviation was 0.21. The standard deviation corresponds to a relative standard deviation of 30%. This is a larger value than the expected ~8 %. The difference may be due to the variation in the Hot Cell environment itself. Figure 6 shows the normal quantile plot of the ^{238}Pu to ^{241}Am (NUK) ratios. All coordinate pairs are approximately on a straight line.

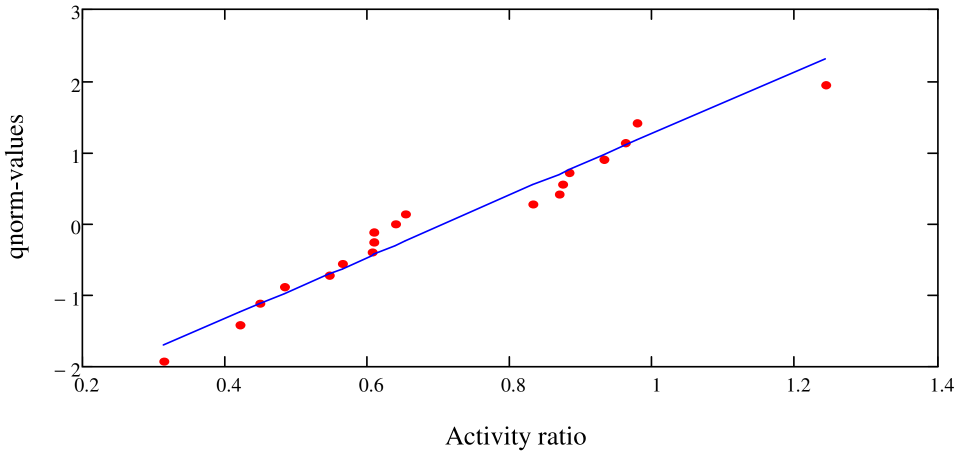

Fig. 6 Normal quantile plot of the ^{238}Pu to ^{241}Am (NUK) ratios.

A ratio which is larger than measured ratios with a probability of 95% is calculated. The ratio is 1.09 with a standard uncertainty = 0.23. The scaling factor relative to the ^{137}Cs content is calculated using Eqn. 2:

$$SF_{\mathrm{Pu238}} = \frac{^{238}\mathrm{Pu}_{\mathrm{NUK}}}{^{241}\mathrm{Am}_{\mathrm{NUK}}} \cdot \frac{^{241}\mathrm{Am}_{\mathrm{NUK}}}{^{241}\mathrm{Am}_{\mathrm{CF}}} \cdot \frac{^{241}\mathrm{Am}_{\mathrm{CF}}}{^{137}\mathrm{Cs}_{\mathrm{CF}}} \Rightarrow \tag{3}$$

$$SF_{\mathrm{Pu238}} = 1.09 \cdot 1.12 \cdot 0.093 = 0.114$$

The standard uncertainty on this scaling factor is 0.056.

For the other radionuclides the same method was used. Figures 7, 8, 9, 10, 11 and 12 show the quantile plots. It is common to all the figures that the coordinate pairs except for outliers approximately lie on a straight line.

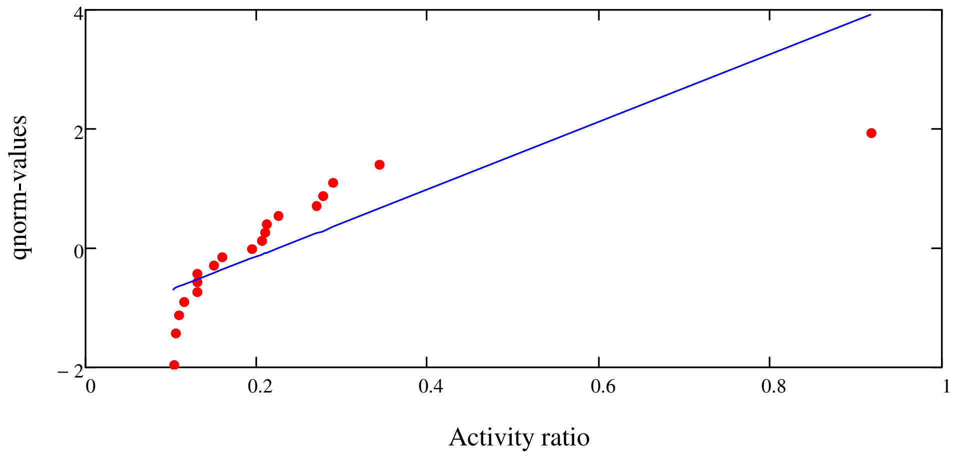

Fig. 7 Normal quantile plot of the ^{239}Pu to ^{241}Am (NUK) ratios.

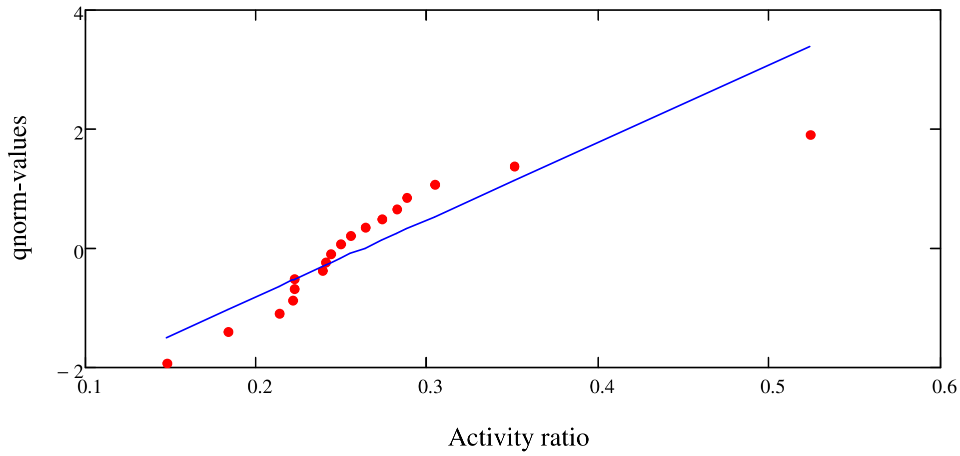

Fig. 8 Normal quantile plot of the ^{240}Pu to ^{241}Am (NUK) ratios.

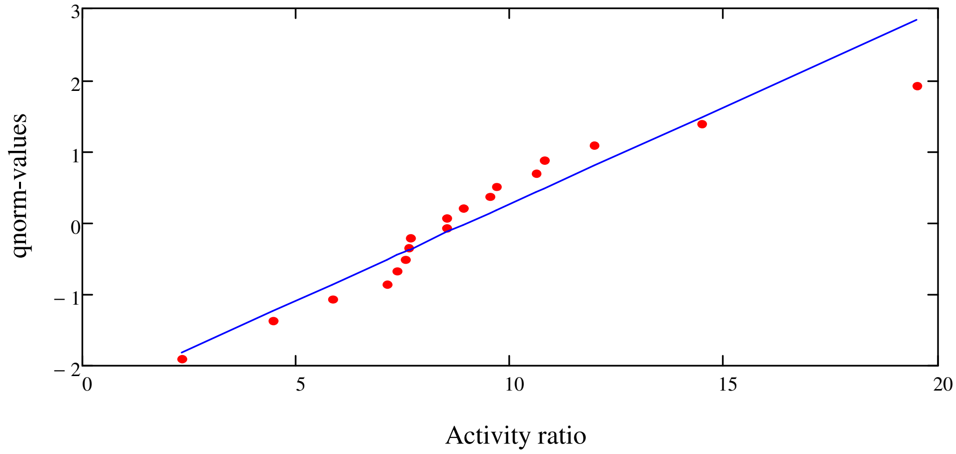

Fig. 9 Normal quantile plot of the ^{241}Pu to ^{241}Am (NUK) ratios.

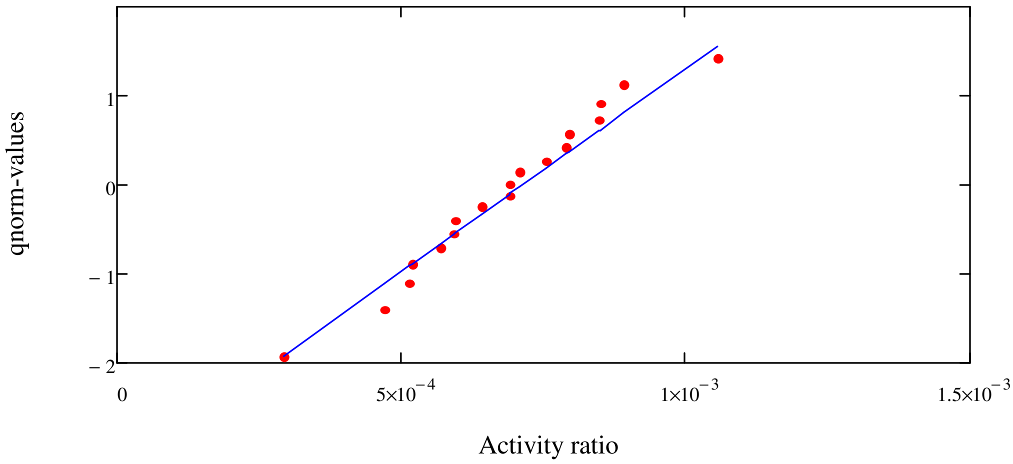

Fig. 10 Normal quantile plot of the ^{242}Pu to ^{241}Am (NUK) ratios.

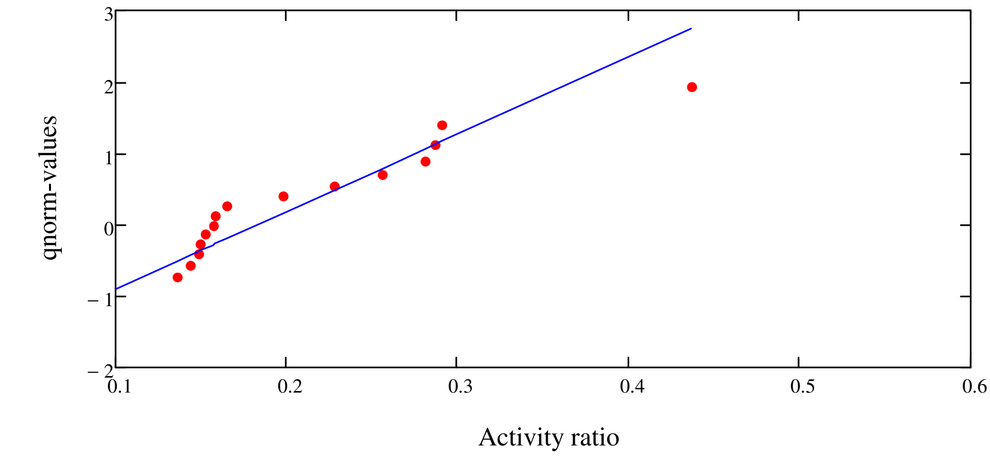

Fig. 11 Normal quantile plot of the (^{243}Cm+^{244}Cm) to ^{241}Am (NUK) ratios.

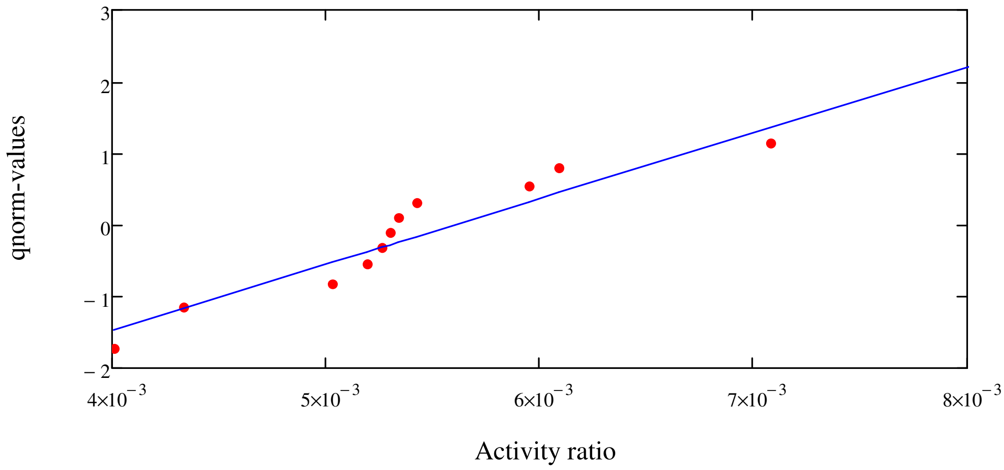

Fig. 12 Normal quantile plot of the (^{242}Cm+^{242m}Am) to ^{241}Am (NUK) ratios.

Table 1 summarizes the results of the data analysis. For completeness the table also contains the result for ^{90}Sr and ^{241}Am. The observed experimental uncertainties are larger than expected from the uncertainties on the activity concentration measurements. These differences can be explained by variations in the Hot Cell environment itself.

Table. 1 Results from the analysis of the nuclide ratio data. For ^{90}Sr and ^{241}Am the mean of ratios are mean of activity concentrations to ^{137}Cs activity concentrations. For the other nuclides the mean of ratios are mean of activity concentrations to ^{241}Am (NUK) concentrations. The 95% ratios are the ratio values, which are larger than a measured ratio with a probability of at least 95%. All scaling factors are conservative nuclide concentrations relative to the concentration of ^{137}Cs.

Nuclide	**Mean of ratios**	**Experimental standard uncertainty**	**Outlier value (i) (included) (e) (excluded)**	**95% ratio (uncertainty)**	**Scaling factor *SF* (uncertainty)**
^{90}**Sr**	0.28	0.16	0.74 (i)	0.54 (0.16)	0.54 (0.16)
^{241}**Am**	0.042	0.031	-	0.093 (0.031)	0.093 (0.031)
^{238}**Pu**	0.71	0.21	-	1.09 (0.23)	0.114 (0.056)
^{239}**Pu**	0.22	0.18	0.92 (i)	0.52 (0.18)	0.054 (0.030)
^{240}**Pu**	0.262	0.077	0.096 (e) 0.52 (i)	0.389 (0.077)	0.041 (0.020)
^{241}**Pu**	9.0	3.7	19 (i)	15.1 (3.7)	1.57 (0.80)
^{242}**Pu**	0.00071	0.00022	0.0013 (i)	0.00108 (0.00022)	0.000112 (0.000055)
^{243}**Cm +** ^{244}**Cm**	0.183	0.092	0.44 (i)	0.334 (0.092)	0.035 (0.018)
^{242}**Cm +** ^{242m}**Am**	0.0056	0.0011	0.0082 (i)	0.0074 (0.0015)	0.00077 (0.00038)

4. CONCLUSION

The measured activity concentration ratios can all be considered to be sampled from normal distributions. Based on these normal distributions, which are characterized only by mean and experimental standard deviation of the ratios measured, scaling factors have been extracted. This extraction is conservative as scaling factors have been chosen as values which have a 95% probability of being larger than a measured value. Most scaling factors have been chosen as a triple product of ratios. For these scaling factors, further conservatism has been introduces as the ratios in the products have been chosen with the 95% criterion. Using the scaling factors in clearance measurements is therefore considered to be conservative (overestimation of activity concentration) on the average. The probability of such a measurement not to be conservative is much less than 5%. The scaling factors for ^{242}Pu and (^{242}Cm+^{242m}Am) can be set to zero as the clearance index value (left side of Eqn. 1) will not be affected as a two digit truncation always is used.

REFERENCES

Søgaard-Hansen, J., Hedemann Jensen, P., Ingemann Larsen, T., Lauridsen, B. (2008). An accredited infrastructure for clearance of decommissioning waste. *IRPA 12.*