



Land Remediation Department	Reference:	LRP(07)P028
	Issue:	1
	Date:	25 September 2007
	File No:	D1071.05.06.06

### Tests on the dissolution characteristics of 151 Dounreay particles using dilute hydrochloric acid

#### ABSTRACT

There are four types of radioactive particles known to be present in the local environment at Dounreay. One type, composed primarily of uranium oxide, appears to be rare but is of importance to the assessment of potential health effects, as it is expected to dissolve in the low pH conditions found in the human gut. This would result in different committed effective doses in the event of inadvertent ingestion of one of this type of particle in comparison to the dose from an insoluble particle. In response to recommendation 7.8.7 of the Dounreay Particles Advisory Group (DPAG), UKAEA has carried out laboratory testing of a statistically significant number of randomly-selected particles to determine the proportion which may dissolve when ingested. An experimental protocol was agreed with DPAG and COMARE prior to commencement of the tests.

Based on the gamma spectrometry results and uranium results of sample filtrates, it is concluded that none of the tested particles dissolved in the acidic test solution. This was clear for all particles tested in the range defined by DPAG as 'relevant' and 'significant'. This applied also for most of the 'minor' particles, However there was a small number which were assessed in greater detail. This assessment indicated that these particles were also not soluble, due to the pattern of  $^{137}\text{Cs}$  and uranium concentration levels in the filtrates from the experiment. Therefore the prevalence of particles likely to dissolve on ingestion is confirmed to be very low.

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## 1. INTRODUCTION

As part of a larger study into the potential health effects of Dounreay particles, two UK laboratories were commissioned by SEPA to carry out *in-vivo* and *in-vitro* experiments using several MTR-type and DFR-type particles. The results of this work, published by the National Radiological Protection Board (now Health Protection Agency; Harrison et al, 2005) showed that out of 12 MTR particles and 11 DFR particles studied, one showed an unusually high degree of dissolution in simulated gut fluids, with some 50% of the radionuclides studied passing into solution. This observation necessitated a separate dose calculation to be carried for the inadvertent ingestion exposure pathway, to assess the potential committed effective dose for particles exhibiting this degree of solubility.

Only at the later stages of the preparation of the 3<sup>rd</sup> DPAG report (DPAG, 2006) was it realised that this unusual result could be due to the presence of uranium oxide particles, as this type of experimental fuel element had been used and reprocessed at Dounreay, although much less commonly than the MTR and DFR fuel elements. DPAG recommended that further work should be undertaken to establish a better estimate of the proportion of particles which could be of this more soluble type (recommendation 7.8.7). The only information known about their likely prevalence came from the NRPB work mentioned above (1 observed occurrence in 23) and from results of UKAEA analyses of some particles by energy dispersive X-ray analysis (1 observed occurrence in 186 analyses up to 30<sup>th</sup> June 2006).

UKAEA provided DPAG with a list of all available particles from the offshore and onshore environments (excluding on-site finds). From this list, DPAG provided a random selection of 160 particles, 150 of which were to be subjected to the experimental procedure, allowing for some contingency on suitability of some particles for study. The random selection provided the following distribution of samples by location:

Seabed	– 132
Sandside beach	– 10
Foreshore	- 18

This distribution closely matches that from the particle retrieval campaigns. Nine of the selected particles consisted of two or more fragments, three had <sup>137</sup>Cs activities in excess of 1 E+07 Bq and there were no irradiated stainless steel particles selected (their prevalence is only about 1% of the total).

The terminology developed by DPAG is used in this report:

Minor particles – <sup>137</sup>Cs activity less than 1 E+05 Bq;

Relevant particles – <sup>137</sup>Cs activity between 1 E+05 Bq and 1 E+06 Bq;

Significant particles – <sup>137</sup>Cs activity greater than 1 E+06 Bq;

This report presents and discusses the results of the particle dissolution tests.

## 2. EXPERIMENTAL SETUP

Communications were exchanged in April 2007 between DPAG and UKAEA on the experimental protocol for testing the dissolution characteristics of particles. It was agreed that the conditions would be:

Use of hydrochloric acid solution at pH2 as reagent

Volume of reagent 25 mls

Temperature 37 degrees C

Duration of reaction 2 hours

Include agitation of flasks during reaction

Filtration through 0.45 µm filters

Gamma spectrometry of filtrate and filter

Retention of filtrates for any further analyses e.g <sup>90</sup>Sr would be carried out on filtrates where the particle had shown high % of <sup>137</sup>Cs released.

The method is described in UKAEA method statement MS116/07/01 (Macgregor, 2007) and is covered by dose assessment LRP(07)P015 (Howse, 2007).

Since the measurement of solutions by ICPMS is a relatively simple procedure, UKAEA subjected all sample filtrates to this method to quantify the amounts of Al, U, Nb and Co released into solution.

## 3. RESULTS OF DISSOLUTION TESTS

The results for gamma spectrometric analysis of the original particle, the sample filtrate and the derived proportion of <sup>137</sup>Cs found in the filtrate are presented in Table 1a, 1b and 1c, covering 14 batches of ten samples and one batch of eleven samples. The corresponding results for U, Al, Nb and Co from ICPMS analysis of the filtrates are presented in Tables 2a, 2b and 2c. It is noted that for that first batch the pH was not under the intended temperature-compensated control and the filter pore size was different (coarser) than for all other samples. It is not thought that these differences would have had a large effect on the results compared to the rest of the samples, and they are therefore included.

Figure 1 shows, by increasing particle activity, the percentage amount of <sup>137</sup>Cs which has been released from the 151 particles tested under the experimental conditions, relative to the quantity measured in the particle shortly after retrieval, corrected for radioactive decay to date of the experiment.

Figure 2 compares the total decay-corrected activity of <sup>137</sup>Cs originally in the particle with the sum of the <sup>137</sup>Cs activities found in the filtrate, on the filter and, in some cases, includes residual activity estimated in the original sample vial i.e. an activity balance.

Figure 3 shows the quantity of uranium found in the sample filtrates, in µg, by increasing particle activity. Also shown on this figure are calculated quantities of

uranium which would be present in uranium oxide particles for two different densities of U<sub>3</sub>O<sub>8</sub>, assuming a constant <sup>137</sup>Cs:uranium ratio and scaling to data for a single confirmed uranium oxide particle 983821 (see calculations in section 4).

Method blanks for the various analyses of filtrates were as follows:

Blank #1

<sup>137</sup>Cs 0.55 Bq; Al 1.4 mg, Co 24 µg, U 0.06 µg

Blank #2

<sup>137</sup>Cs 0.62 Bq; Al 1.1 mg, Co 24 µg, U 0.06 µg

#### 4. DISCUSSION

The maximum percentage of <sup>137</sup>Cs which was observed to be released into solution was 75%; this occurred for seabed particle 02/085. This particle had a <sup>137</sup>Cs activity of only 1.3 E+03 Bq – see Figure 1 and Table 1. Particles 982394 (Foreshore, 1.3 E+04 Bq) and 05/097 (seabed, 2.6 E+04 Bq) showed the next two highest <sup>137</sup>Cs losses into the filtrate at 53% and 8.9% respectively. All of these three particles were in the 'minor' category i.e. less than 1 E+05 Bq. All other particles showed <sup>137</sup>Cs losses in the test solution of less than 4%.

In terms of activity balance between the <sup>137</sup>Cs in the original particle (decay-corrected), compared to the activity in the filtrate plus the activity in the filter, Figure 2 shows that the majority of the 151 samples show good or excellent agreement, within a factor of two. One sample 982394 from the Dounreay foreshore shows a large apparent gain of <sup>137</sup>Cs and it is suspected that the original result for this foreshore sample is too low. The original gamma spectrometry sheets were retrieved from archive and showed that the assigned result agreed with the gamma spectrometry printouts. The result from 1998 for this sample is anomalous, with the variance being due possibly to the counting geometry used at that time. Five or six samples show a poor activity balance due to an overall apparent loss of <sup>137</sup>Cs activity. In one case, particle 02/153, a fragment was observed to break from the filter paper, which when retrieved and counted amounted to 4 E+05 Bq, accounting for much of the missing activity. There were also other losses of sub-fragments during particle manipulation, amounting to about 1.6 E05 Bq when combined at the end of the experiments but which could not be assigned to any specific particles. An additional uncertainty in the activity balance comes from counting of the filter paper; it is not always evident precisely where on the filter paper the particle fragment resides. For a point source geometry calibration, the <sup>137</sup>Cs activity result will be good if the particle is at or close to the centre of the filter paper but will be lower if the particle is offset; the result cannot be too high.

The ICPMS results (Table 2 and Figure 3) showed that the maximum amount of uranium released was just under 3 µg. To obtain an idea of the amount of uranium in MTR particles, their weights lie mostly in the range 0.1 to 10 mg and, if the fragments were composed solely of UAl<sub>4</sub> fuel (typically 15% to 20% by weight uranium) without any surrounding aluminium matrix, then there would be between 15 µg and 2000 µg uranium present. Generally only a very small proportion of the tested particles, if they are all MTR, has dissolved. Similarly, to obtain an idea of the amount of uranium likely to be present if the particles were of the uranium oxide type, we can use information available from SEM-EDAX analysis of the only particle so far

confirmed as uranium oxide, sea bed particle 983821. Although this particle was described as too small to wash and weigh, the dimensions of the particle were measured by SEM to be 120  $\mu\text{m}$  at longest point and 80  $\mu\text{m}$  at widest point. Using the SEM images for this particle, the volume of the particle has been estimated as  $6.3 \text{ E-}07 \text{ cm}^3$  and had a  $^{137}\text{Cs}$  activity of  $6.4 \text{ E+}03 \text{ Bq}$  when originally measured. Uranium oxide fuel if present is most likely to exist as  $\text{U}_3\text{O}_8$  in the environment and probably at a density in the range of about  $4 \text{ g cm}^{-3}$  to  $8 \text{ g cm}^{-3}$  (theoretical density is  $8 \text{ g cm}^{-3}$ ). Therefore, it can be calculated that the amount of uranium present in particle 983821 is approximately between 2.2  $\mu\text{g}$  and 4.4  $\mu\text{g}$ , depending on the density assumed. Using a simple but reasonable assumption that the amount of  $^{137}\text{Cs}$  present in irradiated fuel will be approximately linearly proportional to the amount of uranium present, for the same irradiation and cooling time, then the amount of uranium present in particles of  $\text{U}_3\text{O}_8$  can be estimated for any amount of  $^{137}\text{Cs}$ . Table 3 below shows the calculated amounts of uranium which could be present for  $\text{U}_3\text{O}_8$  particles of different  $^{137}\text{Cs}$  activities.

This relationship between particle activity and calculated amount of uranium in the particle if it was of type  $\text{U}_3\text{O}_8$  is given by the two sloping lines in Figure 3. The solid lines indicate the amount of uranium which would be found in the filtrate if  $\text{U}_3\text{O}_8$  particles of different density had totally dissolved. It can be seen from Figure 3 that only 5 particles, all 'minor', show significant amounts of U dissolution relative to the amounts calculated for  $\text{U}_3\text{O}_8$  particles.

It is necessary to undertake a closer examination of the 'minor' particles which showed notable  $^{137}\text{Cs}$  loss during the dissolution tests together with 'minor' particles which showed notable losses of uranium compared to calculated maximum losses possible (for uranium oxide type). The activities and observed behaviour of these minor particles are combined in Table 4.

While the assessments in Table 4 suggest that these small particles are probably not the soluble uranium oxide type for the reasons given, it is clear that in the case of the 'relevant' (between  $1 \text{ E+}05 \text{ Bq}$  and  $1 \text{ E+}06 \text{ Bq}$ ) and 'significant' ( $> 1 \text{ E+}06 \text{ Bq}$ ) particles studied, both the  $^{137}\text{Cs}$  and the U losses to solution are very small (see Figures 1 and 3) and none of these behave like soluble uranium oxide particles. The small number of 'minor' particles exhibit  $^{137}\text{Cs}$  and U contents in the filtrate from the dissolution tests which are rather ambiguous. On the assumption that one should expect both these analytes to show high losses to the dilute acid reagent, it is concluded that there is no persuasive evidence for the presence of uranium oxide particles in the 151 samples studied in this work.

Previous work at UKAEA Dounreay (Fannon, 1994) assessed the amounts of fission and activation products released from three foreshore MTR particles after treatment with 0.01M HCl at 37 degrees C with continuous stirring for 1 hour, similar to the conditions used in the present work. The residual samples were then totally dissolved which allowed calculations of loss via 0.01M HCl treatment. Between 0.4% and 1.8% of the  $^{137}\text{Cs}$ , 0.13 and 0.38% of the  $^{90}\text{Sr}$  and between 0.07% and 0.14% of the U was removed under these conditions. Total dissolution was reported to be achieved readily by 12M nitric acid (Fannon, 1994), in contrast to the protracted treatments for MTR particles reported to be required by NNC (Wharton, 2004).

ICPMS was also used in the present work to assess the quantities of Al, Co and Nb which was lost from the particles to the dilute HCl solutions during the experiments. Unfortunately, a serious interference at mass 93, probably due to a combination of  $^{56}\text{Fe}$  with  $^{37}\text{Cl}$ , precluded measurement of the amount of  $^{93}\text{Nb}$  in these solutions. For cobalt, only three samples showed the presence of Co, although these were at levels

just above the method detection limit. No evidence of  $^{60}\text{Co}$  was found by gamma spectrometry in the filtrates or in the particles. The levels of aluminium in the filtrates ranged from 1 to 47 mg. The levels showed a relationship with the amount of uranium released and with the amount of  $^{137}\text{Cs}$  activity in the particle and is therefore probably linked to physical size.

## 5. CONCLUSIONS

From a random selection of 160 radioactive particles, 151 samples were subjected to test conditions designed to approximate those in the human gut. From the  $^{137}\text{Cs}$  and uranium results of the filtrates, it is concluded that there are no uranium oxide particles in the 'relevant' or 'significant categories of particles i.e. none of the particles greater than  $1 \text{ E}+05 \text{ Bq } ^{137}\text{Cs}$  were soluble. Further, of the rest of the particles ('minor' particles), there were only 7 which showed loss of one or other of these analytes and whose behaviour were then examined further. It was concluded that none of these minor particles showed persuasive evidence for being of the soluble uranium oxide type. The prevalence of these soluble particles therefore remains very low: taken together with two other bodies of work on particles, only two examples of uranium oxide occurrence have been found, one confirmed by UKAEA during SEM-EDAX analysis and one inferred from results of in-vitro experiments by NRPB. From those works and the present work, only 2 out of 360 particles studied have provided results consistent with uranium oxide.

## 6. REFERENCES

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- Wharton, M. (2004) Radiometric analysis of contaminated rat faeces. NNC Analytical Report L2003161.

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	<sup>137</sup> Cs particle activity decay corrected (Bq)	<sup>137</sup> Cs in filtrate (Bq)	% activity in filtrate
Seabed	L80PART/01/134	2.3E+03	1.6E+01	0.71
Seabed	L80PART/01/045	1.0E+05	2.1E+03	2.06
Seabed	L80PART/00/015	1.5E+03	3.3E+01	2.23
Seabed	L80PART/01/102	8.7E+03	1.2E+00	0.01
Seabed	L80PART/00/009	1.0E+06	1.1E+04	1.13
Seabed	L80PART/00/151	3.6E+04	1.0E+01	0.03
Seabed	L80PART/01/021	1.8E+04	1.1E+00	0.01
Seabed	L80PART/00/114	1.3E+06	8.9E+03	0.70
Seabed	L80PART/00/086	3.5E+05	9.5E+02	0.27
Seabed	L80PART/00/030	3.3E+04	9.8E-01	0.00
Seabed	L80PART/00/076	6.8E+05	1.7E+03	0.25
Seabed	L80PART/01/126	1.1E+05	2.2E+01	0.02
Seabed	L80PART/00/024	1.3E+04	1.1E+01	0.08
Seabed	L80PART/00/048	3.0E+04	2.8E+01	0.09
Seabed	L80PART/01/115	1.7E+04	1.5E+02	0.88
Seabed	L80PART/00/023	1.1E+04	4.3E+00	0.04
Seabed	L80PART/00/020	1.4E+06	7.9E+03	0.58
Seabed	L80PART/00/010	4.2E+05	3.3E+03	0.80
Seabed	L80PART/01/071	1.7E+06	5.5E+03	0.31
Seabed	L80PART/01/024	3.4E+05	1.1E+03	0.31
Seabed	L80PART/00/054	5.0E+04	1.1E+01	0.02
Seabed	L80PART/00/073	5.6E+04	5.3E+01	0.10
Seabed	L80PART/00/078	1.1E+04	8.0E+00	0.07
Seabed	L80PART/01/068	4.1E+05	5.9E+03	1.45
Seabed	L80PART/01/092	4.7E+03	5.8E+00	0.12
Seabed	L80PART/01/065	9.6E+04	1.7E+03	1.74
Foreshore	L80PART/00/171	1.2E+06	1.7E+04	1.41
Seabed	L80PART/00/159	4.4E+03	9.0E+00	0.21
Seabed	L80PART/01/047	3.0E+05	8.5E+02	0.28
Seabed	L80PART/00/170	3.3E+05	9.9E+03	2.98
Seabed	L80PART/00/164	4.9E+05	4.4E+03	0.90
Seabed	L80PART/01/082	2.6E+06	2.9E+03	0.11
Seabed	L80PART/01/111	1.2E+05	1.1E+03	1.00
Seabed	L80PART/01/011	2.8E+05	6.0E+03	2.17
Seabed	L80PART/01/051	2.2E+06	1.8E+03	0.08
Seabed	L80PART/01/023	3.0E+03	2.2E+00	0.08
Seabed	L80PART/03/077	1.3E+05	3.4E+02	0.26
Sandside	L80PART/03/008	1.7E+04	1.6E+01	0.10
Seabed	L80PART/03/060	4.5E+03	5.6E+00	0.13
Seabed	L80PART/04/091	4.7E+04	1.1E+03	2.44
Seabed	L80PART/04/053	2.2E+03	3.4E+00	0.15
Seabed	L80PART/04/030	1.7E+03	9.0E+00	0.54
Seabed	L80PART/04/027	1.9E+03	8.8E+00	0.47
Seabed	L80PART/04/032	7.6E+03	2.1E+00	0.03
Foreshore	L80PART/04/108	5.0E+05	8.0E+02	0.16
Seabed	L80PART/04/029	8.6E+03	1.8E+00	0.02
Seabed	L80PART/03/062	1.8E+04	1.0E+01	0.06
Seabed	L80PART/04/035	7.8E+03	1.8E+00	0.02
Seabed	L80PART/03/065	1.9E+05	3.1E+02	0.16
Seabed	L80PART/03/086	3.5E+05	4.7E+02	0.14

**Table 1a - <sup>137</sup>Cs results for batches 1 to 5.**

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	<sup>137</sup> Cs particle activity decay corrected (Bq)	<sup>137</sup> Cs in filtrate (Bq)	% activity in filtrate
Sandside	L80PART/03/003	1.2E+04	2.3E+00	0.02
Foreshore	L80PART/03/001	1.5E+06	2.4E+03	0.16
Foreshore	L80PART/04/136	7.7E+04	6.0E+02	0.77
Seabed	L80PART/03/081	1.2E+04	1.5E+01	0.13
Seabed	L80PART/03/068	3.5E+05	4.4E+03	1.24
Seabed	L80PART/04/048	4.1E+06	4.4E+03	0.11
Seabed	L80PART/04/090	3.4E+04	2.0E+01	0.06
Seabed	L80PART/04/061	3.3E+03	7.4E+00	0.23
Foreshore	L80PART/04/126	3.2E+05	2.4E+01	0.01
Seabed	L80PART/04/067	6.5E+04	2.5E+01	0.04
Seabed	L80PART/04/087	1.0E+06	1.6E+04	1.52
Seabed	L80PART/05/060	2.8E+03	2.4E+01	0.84
Seabed	L80PART/05/098	1.8E+04	1.8E+01	0.10
Seabed	L80PART/04/042	1.5E+05	2.6E+03	1.75
Foreshore	L80PART/05/005	9.1E+05	2.6E+03	0.29
Seabed	L80PART/05/ <sup>137</sup>	2.4E+05	3.6E+02	0.15
Seabed	L80PART/05/136	8.1E+04	1.5E+03	1.86
Seabed	L80PART/05/044	8.5E+04	6.1E+00	0.01
Sandside	L80PART/06/027	4.7E+04	6.2E+01	0.13
Seabed	L80PART/05/090	8.7E+03	4.9E+00	0.06
Foreshore	930417	3.6E+06	1.1E+04	0.31
Foreshore	941510	3.7E+05	2.9E+03	0.78
Foreshore	960300	1.0E+06	3.9E+03	0.38
Foreshore	982394	1.3E+04	6.8E+03	52.57
Seabed	983840	2.5E+06	3.6E+03	0.15
Seabed	990025	7.3E+06	3.0E+04	0.41
Seabed	992444	1.1E+07	1.3E+04	0.12
Foreshore	992647	3.9E+06	4.8E+04	1.24
Seabed	L80PART/05/047	1.4E+05	5.3E+03	3.76
Sandside	L80PART/07/004	3.2E+04	2.6E+01	0.08
Seabed	L80PART/05/123	8.1E+03	2.1E+01	0.25
Seabed	L80PART/05/130	1.9E+05	3.1E+01	0.02
Seabed	L80PART/05/124	4.1E+03	1.6E+01	0.39
Seabed	L80PART/05/057	2.3E+04	2.0E+00	0.01
Seabed	L80PART/05/038	5.4E+04	6.3E+02	1.18
Sandside	L80PART/07/017	1.2E+04	5.1E+00	0.04
Foreshore	L80PART/06/040	3.3E+05	3.2E+01	0.01
Seabed	L80PART/05/052	2.0E+05	1.9E+03	0.98
Sandside	L80PART/07/008	5.6E+04	1.1E+03	1.91
Seabed	L80PART/05/097	2.6E+04	2.3E+03	8.96
Seabed	L80PART/05/134	1.4E+06	3.7E+02	0.03
Sandside	L80PART/06/031	5.9E+04	1.7E+02	0.30
Seabed	L80PART/05/056	3.1E+02	7.5E+00	2.40
Foreshore	L80PART/05/002	1.8E+05	7.9E+01	0.04
Sandside	L80PART/06/029	4.3E+04	9.4E+01	0.22
Seabed	L80PART/02/069	1.4E+04	3.9E+00	0.03
Seabed	L80PART/02/054	1.2E+05	1.8E+01	0.01
Foreshore	L80PART/02/006	8.9E+05	7.4E+03	0.84
Seabed	L80PART/02/023	4.6E+05	1.1E+04	2.31
Seabed	L80PART/02/033	3.6E+05	2.9E+03	0.80

Table 1b - <sup>137</sup>Cs results for batches 6 to 10.

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	<sup>137</sup> Cs particle activity decay corrected (Bq)	<sup>137</sup> Cs in filtrate (Bq)	% activity in filtrate
Seabed	L80PART/02/062	1.9E+04	7.4E+00	0.04
Seabed	L80PART/02/101	1.3E+04	9.3E+00	0.07
Seabed	L80PART/02/140	2.9E+06	7.0E+03	0.24
Seabed	L80PART/02/087	2.5E+06	3.6E+03	0.14
Seabed	L80PART/02/097	8.2E+04	1.0E+01	0.01
Seabed	L80PART/02/085	1.3E+03	1.0E+03	75.01
Seabed	L80PART/02/080	8.0E+04	1.1E+02	0.14
Seabed	L80PART/02/127	3.1E+03	2.8E+00	0.09
Sandside	L80PART/02/005	5.7E+04	3.7E+00	0.01
Seabed	L80PART/02/186	1.2E+07	3.4E+03	0.03
Seabed	L80PART/02/166	3.3E+06	8.5E+03	0.26
Seabed	L80PART/02/159	2.8E+04	4.3E+00	0.02
Seabed	L80PART/02/195	2.9E+06	2.3E+04	0.83
Seabed	L80PART/02/204	1.4E+06	5.5E+04	3.88
Seabed	L80PART/02/203	2.8E+04	9.0E+00	0.03
Seabed	L80PART/02/153	1.2E+06	9.6E+03	0.83
Seabed	L80PART/02/236	2.2E+05	5.7E+02	0.26
Seabed	L80PART/02/183	2.6E+05	6.4E+02	0.25
Seabed	L80PART/02/053	4.1E+04	2.6E+01	0.06
Seabed	L80PART/02/199	3.2E+04	3.2E+01	0.10
Seabed	L80PART/02/331	5.2E+04	2.4E+01	0.05
Seabed	L80PART/02/351	2.9E+06	2.7E+03	0.09
Seabed	L80PART/02/398	7.5E+04	1.1E+01	0.01
Seabed	L80PART/02/267	3.0E+04	4.8E+01	0.16
Sandside	L80PART/02/411	3.5E+04	8.4E+02	2.40
Seabed	L80PART/02/347	3.3E+04	5.1E+00	0.02
Foreshore	L80PART/02/339	7.2E+05	2.3E+04	3.19
Seabed	L80PART/02/293	3.7E+06	3.4E+04	0.93
Seabed	L80PART/02/318	6.5E+06	8.4E+03	0.13
Seabed	L80PART/02/314	2.1E+07	9.6E+03	0.05
Seabed	L80PART/02/253	2.7E+05	4.1E+02	0.15
Seabed	L80PART/02/244	1.3E+06	1.1E+04	0.94
Seabed	L80PART/02/259	1.3E+05	1.2E+03	0.99
Seabed	L80PART/02/260	4.8E+05	1.9E+03	0.39
Seabed	L80PART/02/296	1.7E+05	3.0E+03	1.76
Seabed	L90PART/02/273	2.4E+06	5.7E+04	2.35
Seabed	L80PART/02/262	3.6E+05	1.3E+03	0.38
Seabed	L80PART/02/246	1.6E+06	4.7E+03	0.29
Seabed	L80PART/02/078	6.0E+05	7.7E+03	1.29
Seabed	L80PART/02/301	3.7E+06	8.9E+03	0.24
Seabed	L80PART/02/277	6.2E+06	6.3E+04	1.03
Seabed	L80PART/02/355	1.6E+06	1.2E+03	0.08
Foreshore	L80PART/02/240	6.6E+05	1.3E+03	0.19
Seabed	L80PART/02/126	4.8E+03	1.5E+01	0.31
Seabed	L80PART/02/187	1.3E+06	4.0E+04	2.99
Seabed	L80PART/02/216	4.6E+04	1.3E+01	0.03
Seabed	L80PART/02/237	9.8E+05	2.2E+03	0.22
Seabed	L80PART/02/191	2.9E+04	2.1E+01	0.07
Seabed	L80PART/02/225	2.9E+06	2.8E+04	0.98
Seabed	L80PART/02/210	1.5E+04	1.2E+01	0.08
Seabed	L80PART/02/288	6.7E+06	4.1E+03	0.06

**Table 1c - <sup>137</sup>Cs results for batches 11 to 15.**

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	Al (mg in filtrate)	Co (µg in filtrate)	U (µg in filtrate)
Seabed	L80PART/01/134	4.72	<32	0.4
Seabed	L80PART/01/045	3.68	<32	0.16
Seabed	L80PART/00/015	5.52	<32	<0.08
Seabed	L80PART/01/102	12.8	<32	0.08
Seabed	L80PART/00/009	11.2	<32	0.4
Seabed	L80PART/00/151	1.84	<32	0.08
Seabed	L80PART/01/021	3.92	<32	<0.08
Seabed	L80PART/00/114	24	<32	0.72
Seabed	L80PART/00/086	4.88	<32	0.08
Seabed	L80PART/00/030	7.52	<32	<0.08
Seabed	L80PART/00/076	8.4	<24	0.12
Seabed	L80PART/01/126	2.1	<24	<0.06
Seabed	L80PART/00/024	4.08	<24	0.12
Seabed	L80PART/00/048	2.22	<24	<0.06
Seabed	L80PART/01/115	2.22	<24	<0.06
Seabed	L80PART/00/023	2.16	<24	0.12
Seabed	L80PART/00/020	16.2	<24	0.18
Seabed	L80PART/00/010	9	<24	0.06
Seabed	L80PART/01/071	12.6	<24	0.18
Seabed	L80PART/01/024	3.84	<24	0.06
Seabed	L80PART/00/054	1.5	<24	<0.06
Seabed	L80PART/00/073	12	<24	0.06
Seabed	L80PART/00/078	25.8	<24	<0.06
Seabed	L80PART/01/068	2.04	<24	<0.06
Seabed	L80PART/01/092	7.8	<24	<0.06
Seabed	L80PART/01/065	8.4	<24	0.06
Foreshore	L80PART/00/171	8.4	<24	1.68
Seabed	L80PART/00/159	4.08	<24	<0.06
Seabed	L80PART/01/047	6.6	<24	<0.06
Seabed	L80PART/00/170	4.56	<24	0.06
Seabed	L80PART/00/164	8.4	<24	0.12
Seabed	L80PART/01/082	10.8	<24	0.42
Seabed	L80PART/01/111	9	<24	<0.06
Seabed	L80PART/01/011	12	<24	0.06
Seabed	L80PART/01/051	10.8	<24	0.12
Seabed	L80PART/01/023	9	<24	<0.06
Seabed	L80PART/03/077	26.4	<24	0.42
Sandside	L80PART/03/008	3.24	<24	<0.06
Seabed	L80PART/03/060	5.76	27.6	0.42
Seabed	L80PART/04/091	6	<24	<0.06
Seabed	L80PART/04/053	3.96	<24	0.36
Seabed	L80PART/04/030	6	<24	1.56
Seabed	L80PART/04/027	4.38	<24	0.12
Seabed	L80PART/04/032	3.24	38.4	0.06
Foreshore	L80PART/04/108	4.38	<24	0.18
Seabed	L80PART/04/029	7.2	<24	0.06
Seabed	L80PART/03/062	4.68	33.6	0.06
Seabed	L80PART/04/035	2.64	<24	0.06
Seabed	L80PART/03/065	6.6	<24	0.18
Seabed	L80PART/03/086	7.2	<24	0.12

**Table 2a - ICPMS results for three elements in filtrates, batches 1 to 5.**

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	Al (mg in filtrate)	Co (µg in filtrate)	U (µg in filtrate)
Sandside	L80PART/03/003	2.88	<24	<0.06
Foreshore	L80PART/03/001	15.6	<24	0.3
Foreshore	L80PART/04/136	5.34	<24	<0.06
Seabed	L80PART/03/081	2.94	<24	<0.06
Seabed	L80PART/03/068	6	<24	0.36
Seabed	L80PART/04/048	12.6	<24	0.96
Seabed	L80PART/04/090	1.08	<24	<0.06
Seabed	L80PART/04/061	1.26	<24	<0.06
Foreshore	L80PART/04/126	1.38	<24	<0.06
Seabed	L80PART/04/067	2.28	<24	<0.06
Seabed	L80PART/04/087	15	<24	1.8
Seabed	L80PART/05/060	1.44	<24	<0.06
Seabed	L80PART/05/098	16.8	<24	0.06
Seabed	L80PART/04/042	3.54	<24	<0.06
Foreshore	L80PART/05/005	3.42	<24	0.12
Seabed	L80PART/05/ <sup>137</sup>	2.52	<24	0.12
Seabed	L80PART/05/136	2.28	<24	0.12
Seabed	L80PART/05/044	2.22	<24	<0.06
Sandside	L80PART/06/027	5.46	<24	<0.06
Seabed	L80PART/05/090	1.38	<24	<0.06
Foreshore	930417	7.8	<24	0.18
Foreshore	941510	4.2	<24	0.06
Foreshore	960300	12	<24	0.24
Foreshore	982394	5.58	<24	0.54
Seabed	983840	19.2	<24	0.18
Seabed	990025	66	<24	2.76
Seabed	992444	66	<24	1.86
Foreshore	992647	25.8	<24	2.16
Seabed	L80PART/05/047	4.08	<24	<0.06
Sandside	L80PART/07/004	8.4	<24	<0.06
Seabed	L80PART/05/123	3.96	<24	<0.06
Seabed	L80PART/05/130	2.52	<24	<0.06
Seabed	L80PART/05/124	21.6	<24	<0.06
Seabed	L80PART/05/057	1.74	<24	<0.06
Seabed	L80PART/05/038	12	<24	<0.06
Sandside	L80PART/07/017	7.2	<24	<0.06
Foreshore	L80PART/06/040	10.2	<24	<0.06
Seabed	L80PART/05/052	1.68	<24	<0.06
Sandside	L80PART/07/008	11.4	<24	<0.06
Seabed	L80PART/05/097	1.86	<24	<0.06
Seabed	L80PART/05/134	3.18	<24	<0.06
Sandside	L80PART/06/031	1.26	<24	<0.06
Seabed	L80PART/05/056	2.94	<24	<0.06
Foreshore	L80PART/05/002	2.82	<24	<0.06
Sandside	L80PART/06/029	7.8	<24	<0.06
Seabed	L80PART/02/069	1.74	<24	<0.06
Seabed	L80PART/02/054	6	<24	<0.06
Foreshore	L80PART/02/006	4.92	<24	0.12
Seabed	L80PART/02/023	4.14	<24	0.12
Seabed	L80PART/02/033	4.02	<24	<0.06

**Table 2b - ICPMS results for three elements in filtrates, batches 6 to 10.**

**NOT PROTECTIVELY MARKED**

Location	Lab sample number	Al (mg in filtrate)	Co (µg in filtrate)	U (µg in filtrate)
Seabed	L80PART/02/062	1.98	<24	<0.06
Seabed	L80PART/02/101	7.2	<24	<0.06
Seabed	L80PART/02/140	16.2	<24	0.24
Seabed	L80PART/02/087	4.56	<24	0.12
Seabed	L80PART/02/097	3.78	<24	<0.06
Seabed	L80PART/02/085	0.9	<24	<0.06
Seabed	L80PART/02/080	1.8	<24	<0.06
Seabed	L80PART/02/127	4.32	<24	<0.06
Sandside	L80PART/02/005	1.98	<24	<0.06
Seabed	L80PART/02/186	84	<24	1.8
Seabed	L80PART/02/166	21.6	<24	0.18
Seabed	L80PART/02/159	3	<24	<0.06
Seabed	L80PART/02/195	11.4	<24	0.6
Seabed	L80PART/02/204	17.4	<24	0.42
Seabed	L80PART/02/203	2.7	<24	<0.06
Seabed	L80PART/02/153	9.6	<24	0.18
Seabed	L80PART/02/236	5.58	<24	<0.06
Seabed	L80PART/02/183	1.56	<24	<0.06
Seabed	L80PART/02/053	2.46	<24	<0.06
Seabed	L80PART/02/199	3.72	<24	<0.06
Seabed	L80PART/02/331	3	<24	<0.06
Seabed	L80PART/02/351	9	<24	0.18
Seabed	L80PART/02/398	4.68	<24	<0.06
Seabed	L80PART/02/267	6	<24	<0.06
Sandside	L80PART/02/411	4.92	<24	<0.06
Seabed	L80PART/02/347	7.2	<24	<0.06
Foreshore	L80PART/02/339	17.4	<24	0.48
Seabed	L80PART/02/293	46.8	<24	1.98
Seabed	L80PART/02/318	23.4	<24	1.2
Seabed	L80PART/02/314	44.4	<24	1.5
Seabed	L80PART/02/253	7.2	<24	<0.06
Seabed	L80PART/02/244	11.4	<24	0.24
Seabed	L80PART/02/259	24	<24	<0.06
Seabed	L80PART/02/260	7.2	<24	<0.06
Seabed	L80PART/02/296	20.4	<24	0.18
Seabed	L90PART/02/273	13.2	<24	0.48
Seabed	L80PART/02/262	6.6	<24	<0.06
Seabed	L80PART/02/246	9.6	<24	0.18
Seabed	L80PART/02/078	4.8	<24	0.06
Seabed	L80PART/02/301	7.2	<24	0.24
Seabed	L80PART/02/277	28.2	<24	0.96
Seabed	L80PART/02/355	20.4	<24	0.18
Foreshore	L80PART/02/240	3.42	<24	0.36
Seabed	L80PART/02/126	1.8	<24	<0.06
Seabed	L80PART/02/187	12.6	<24	0.36
Seabed	L80PART/02/216	4.02	<24	<0.06
Seabed	L80PART/02/237	6.6	<24	<0.06
Seabed	L80PART/02/191	1.74	<24	<0.06
Seabed	L80PART/02/225	13.8	<24	0.12
Seabed	L80PART/02/210	5.7	<24	<0.06
Seabed	L80PART/02/288	12	<24	0.36

**Table 2c - ICPMS results for three elements in filtrates, batches 11 to 15.**

Assumed density U <sub>3</sub> O <sub>8</sub> (g cm <sup>-3</sup> )	<sup>137</sup> Cs activity (Bq)	Approx amount U present (µg)	Comments
4	6.4 E+03	2.2	Particle 983821
4	1.0 E+05	33.4	
4	1.0 E+06	334	
4	1.0 E+07	3340	
8	6.4 E+03	4.3	Particle 983821
8	1.0 E+05	67.2	
8	1.0 E+06	672	
8	1.0 E+07	6720	

**Table 3 - Total amount of uranium (µg) calculated to be present in U<sub>3</sub>O<sub>8</sub> particles for different densities and different <sup>137</sup>Cs activities.**

Sample	<sup>137</sup> Cs activity in particle(Bq)	% loss <sup>137</sup> Cs to filtrate	Loss of U to filtrate (µg)	Loss of U to filtrate (% of total if U <sub>3</sub> O <sub>8</sub> )	Comments and conclusions
02/085	1.3 E+03	75	< 0.06	< 13	Similar and low amounts of <sup>137</sup> Cs in filtrate, filter and original particle; no measureable U loss; such very small particles could show artefacts on U or Cs loss due to high surface area: volume ratios or shape; not U oxide particle.
982394	1.3 E+04	53	0.54	6 - 12	SEMEDAX shows it is an MTR particle; original reported particle <sup>137</sup> Cs result much too low so % Cs loss is an artefact of this.
05/097	2.6 E+04	8.96	< 0.06	< 0.6	no measureable U loss, low Cs loss; not U oxide particle.
04/030	1.7 E+03	0.54	1.56	130 - 270	Suggests total dissolution of U but negligible <sup>137</sup> Cs release; such very small particles could show artefacts on U or Cs loss due to high surface area: volume ratios or shape; probably not U oxide particle
03/060	4.5 E+03	0.13	0.42	14 - 28	Low Cs loss, minor U loss; such very small particles could show artefacts on U or Cs loss due to high surface area: volume ratios or shape; probably not U oxide particle
01/134	2.3 E+03	0.71	0.40	25 - 50	Low Cs loss, some U loss; such very small particles could show artefacts on U or Cs loss due to high surface area: volume ratios or shape; probably not U oxide particle
04/053	2.2 E+03	0.15	0.36	24 - 48	Very low Cs loss, significant U loss; such very small particles could show artefacts on U or Cs loss due to high surface area: volume ratios or shape; probably not U oxide particle

**Table 4 - Assessment of combined results for <sup>137</sup>Cs and for U loss from several 'minor' particles**

Proportion of  $^{137}\text{Cs}$  leached by pH2 HCl at 37degC

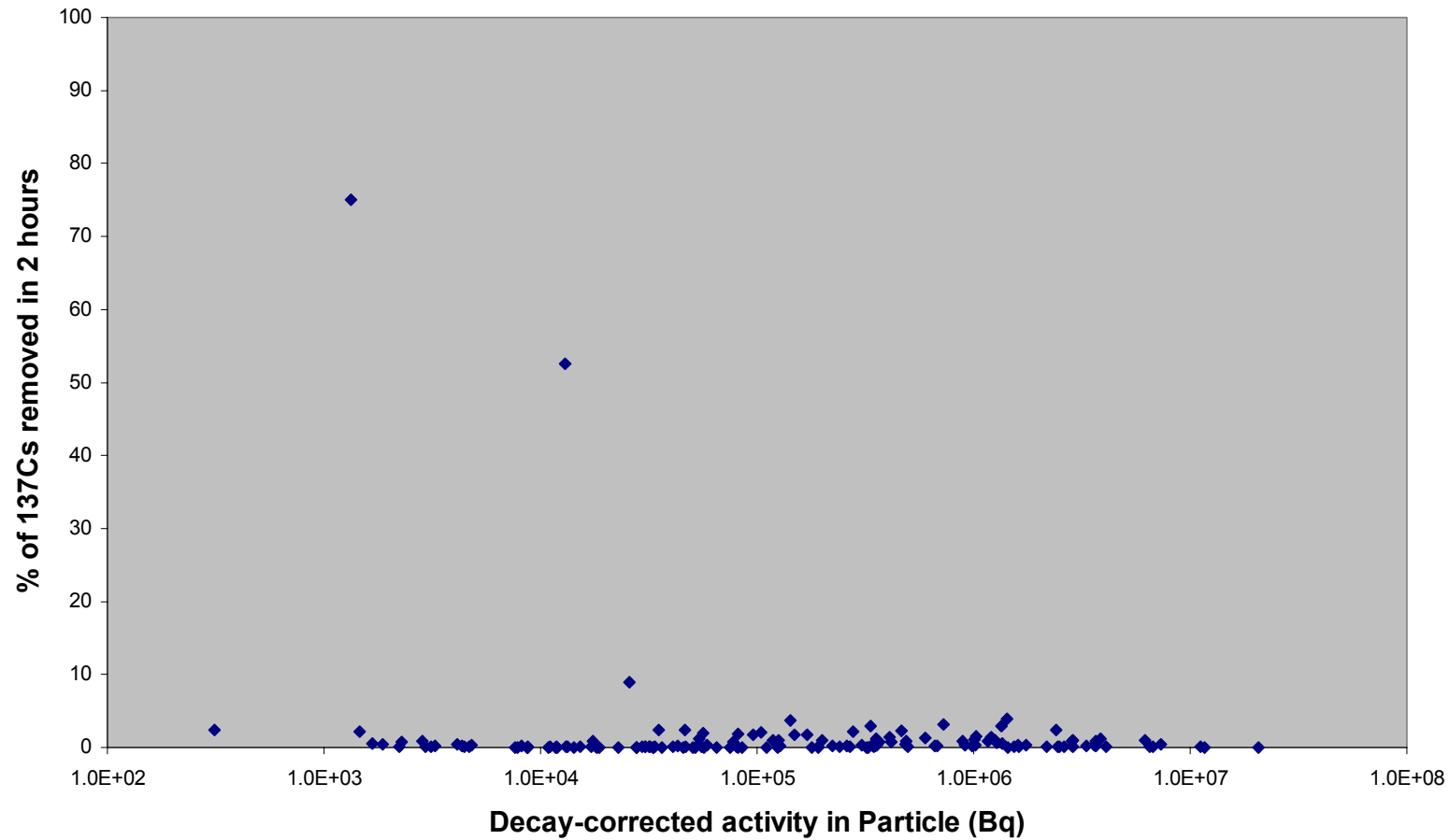


Figure 1 - Proportion of  $^{137}\text{Cs}$  removed from 151 particles under test conditions described.

Decay-corrected  $^{137}\text{Cs}$  activity vs summed activity (filtrate+filter+vial)

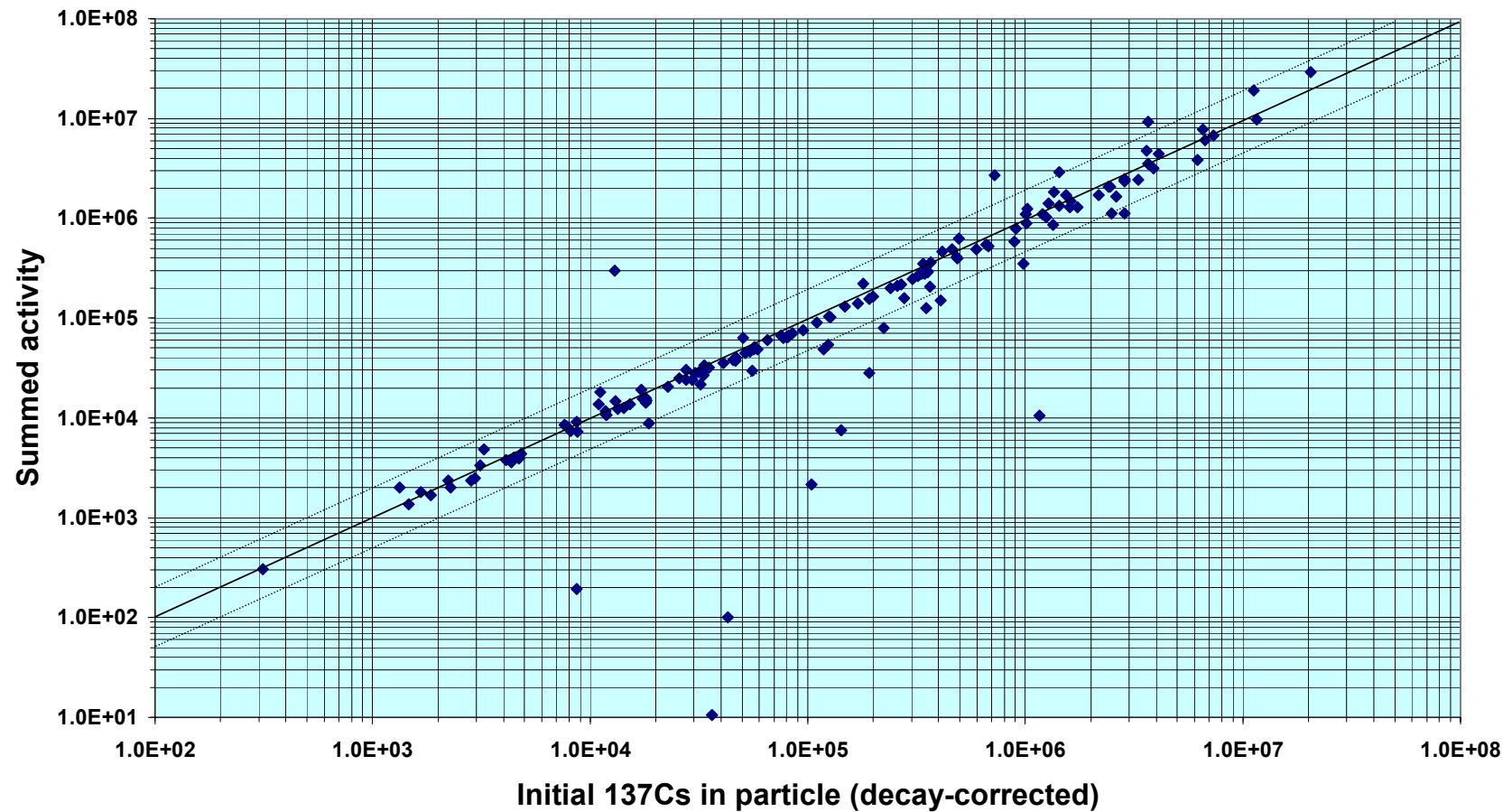
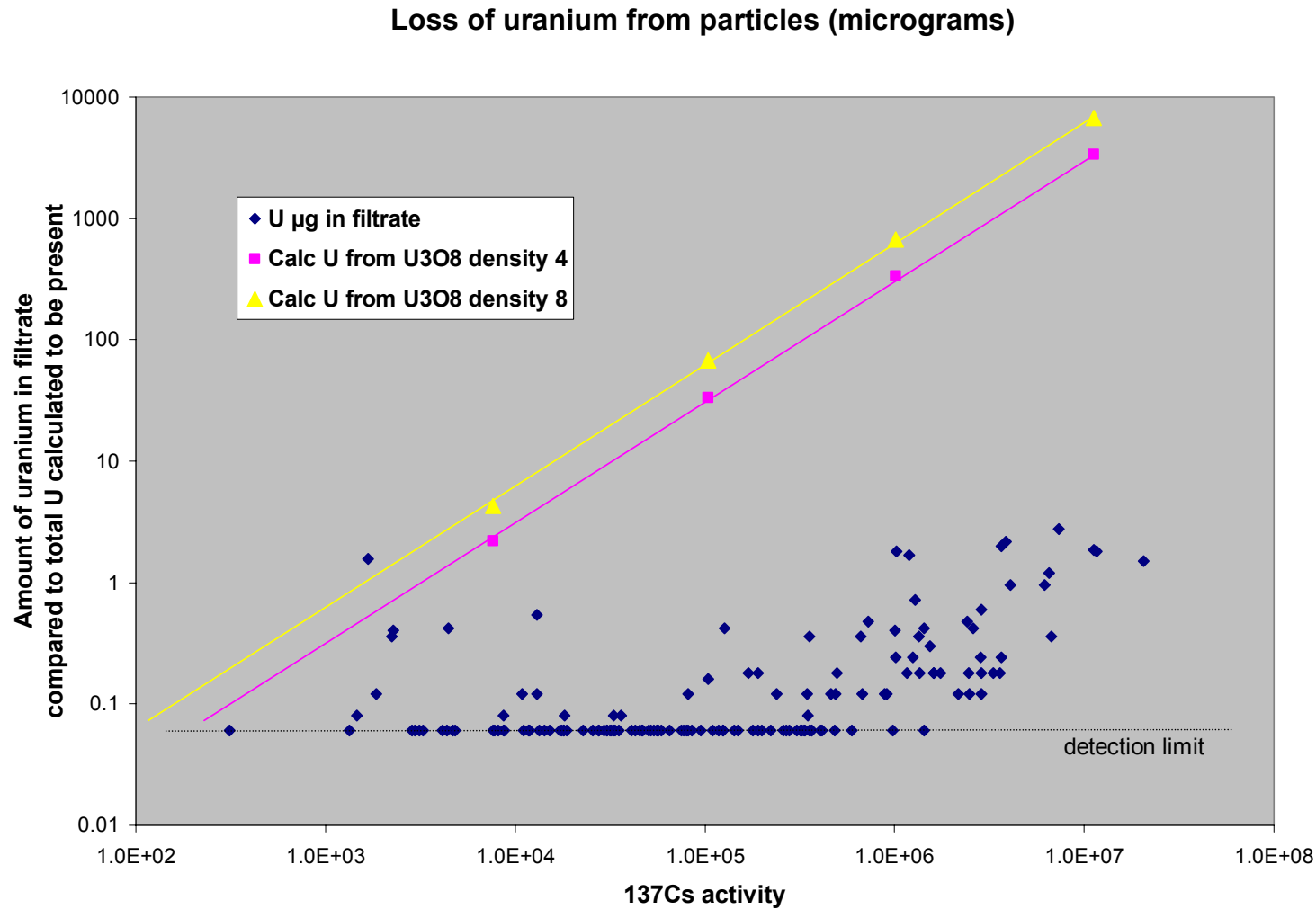


Figure 2 - Activity balance ( $^{137}\text{Cs}$ ) between original activity measured in particle, decay-corrected, and the sum of activities found in the filtrate, on the filter and (in some cases) remaining in the sample vial.



**Figure 3 - Loss of uranium from 151 tested particles, compared to calculated amount of uranium present in uranium oxide particles at two different densities and for a range of  $^{137}\text{Cs}$  activities.**