A Comparison of True Alpha Activities in Air Filter Samples with Values Obtained from Radioactivity-in-air Monitors

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A fundamental problem with measurements of alpha activity on air filters from the nuclear workplace is that the calibration sources used with radioactivity-in-air monitors are not in the same physical form as actual samples. The samples suffer from alpha absorption due to effects such as self-attenuation within the radioactive particle, entrapment of the particle within the filter, and the accumulation of dust layers on the filter. This can lead to underestimation of alpha activity in air and therefore underestimation over time of intake and hence committed internal dose. To obtain a first indication of the magnitude of the problem, NPL obtained a set of nineteen contaminated air filters from three UK nuclear sites and measured them using a number of typical workplace monitors. The same filters were then subjected to radiochemical analysis to measure the activities of Am, Pu and U nuclides. The ratio of true to monitored activities was typically in the range 1 – 5, but in a few cases was of the order of 20. The results are compared with 'correction factors' used at UK nuclear sites and from the literature. The study points to the possibility of significant underestimation of airborne alpha activity in some circumstances and that plant-by-plant determination of ratios may be required to ensure that workplace monitoring of alpha particulate in air is accurate.

Key words: Radioactivity; Air monitoring; Internal dosimetry

1. Introduction

Measurements of radioactivity on air filters are routinely carried out in the nuclear industry both for Radiation Protection purposes and for monitoring of stack discharges. Air is drawn through a filter for a given period of time and the filter is then presented to a suitable detector. Filters are often left sampling air for several weeks, and, as a result, can acquire layers of dust or dirt. In many cases, the detector has been calibrated with standard sources which are physically different from typical samples with the result that the detection efficiency derived from a standard source is not applicable. One example[1] (done by radiochemical analysis of some heavily dust-loaded filter samples) indicated that the true efficiency of measuring filters differed from the efficiency derived from 'standard sources' by a factor of two. A more detailed study of this type (e.g. for several filter and detector types) would provide a better indication of the size of any discrepancies which may be occurring in day-to-day workplace monitoring.

The aim of this project was to acquire some contaminated air filters from nuclear sites, to measure their 'apparent' activities (i.e. from direct monitoring) and their true activities (from radiochemical analysis) and to derive a ratio of 'true to apparent' activity for different filter types and filter conditions. The ratios obtained would be specimen values for indicative purposes only and would NOT be intended to be 'correction factors' for use in practice.

This paper summarises:

- the procurement of the filter samples;
- the non-destructive measurements of the filters;
- the radiochemical analysis of the filters;
- the results and conclusions.

2. Procurement of filter samples

Ideally, the filters would come from several nuclear sites and their physical condition would be variable (i.e. 'clean', dusty or dirty to the naked eye). This would enable data to be acquired for a range of filter types, radionuclides and levels of air quality. Unfortunately, due to limited resources being available, it was possible only to analyse up to 20 filters for this study; however, it was hoped that this would demonstrate the potential magnitude of 'true-to-apparent' activity ratios.

NPL canvassed a range of users, chiefly via presentations at meetings of its Airborne Radioactivity Monitoring Users' Group (ARMUG[2]) and its Ionising Radiation Metrology Forum (IRMF[2]). Users from three sites offered to provide a total of 19 suitable filter samples. These were supplied in confidence and the sites are not identified in this paper. Each filter was assigned an NPL sample code between IM090182 and IM090200 and the type of filter in each case is given in Table 1. Apart from IM090187 (which had considerable surface dirt), none seemed to have significant surface deposits and all were used in this study.

3. Measurements

3.1 Non-destructive measurements of filters

The filters were subjected to non-destructive measurements using:

- High-resolution γ-ray spectrometry;
- Direct monitoring using installed air-sample counters and portable detectors.

3.1.1 High-resolution γ-ray spectrometry

Qualitative high-resolution γ-ray spectrometry was used to check for the presence of any γ-emitting radionuclides which might interfere with the later radiochemical separations of the α-emitting radioelements present. The radionuclides ^{60}Co, ^{134}Cs, ^{137}Cs, ^{152}Eu and ^{154}Eu were each detected on one or more filters, each at levels in the range 0.1 – 200 Bq per filter. The radionuclide 'fingerprint' was different for each filter. None of the radionuclides detected were considered to be a potential problem for the separations.

3.1.2 Direct monitoring using installed or portable detectors

Direct monitoring was carried out using the following instruments:

- Canberra 'iSolo' α and β counting system
- JCS scaler/timer and Harwell 'drawer' counter
- Thermo Mini 900EP15 β and γ probe
- Thermo AP2 α probe

The iSolo (Canberra[3]) is a single-sample counter for counting α and β activity on filters, surface smears or swipes and will accommodate essentially all air filter types. The detector is a solid state PIPS detector. The iSolo discriminates radon (^{222}Rn) and thoron (^{220}Rn) and their daughters from actinides and fission products on the filter. The JCS unit and drawer counter is a similar unit. The Mini 900EP15 (Thermo Scientific[4]) is one of a series of well-known small portable radiation counters. It has an analogue display with a logarithmic scale to facilitate counting at both low and high count-rates. It is fitted with an end-window (15.5 cm^2) halogen-quenched Geiger-Müller tube. The AP2 (also Thermo Scientific) is a hand-held probe containing a ZnS(Ag) scintillator and photomultiplier tube. The detectors were subjected to standard function checks (as appropriate for each detector type) prior to counting the filters.

Each filter was counted on all four detector units. Unlike the iSolo and JCS units, the EP15 and AP2 had no associated sample holders, so for these instruments the detector face was positioned manually in virtual contact with the filter. For measurements using the iSolo and Harwell instruments, an integration time of 600 s per filter was used. For the EP15 measurements, ten readings were taken per filter, each reading consisting of counts observed over a 10 s integration time; the mean of the ten readings was then calculated. The same procedure was adopted for measurements using the AP2, except when the observed count-rate was < 1 s^{-1}, in which case a single integration period of 300 s was used. The results are given in Table 2.

Detection efficiencies of 39 % and 34 % respectively were provided for measurements of ^{241}Am in the iSolo and Harwell counters, and these values have been used to derive a total alpha activity per filter. No efficiencies were applied to either the EP15 or AP2 results, so results from these instruments are expressed in counts per second.

3.2 Radiochemical analysis of filters

The filters were analysed for the presence of any of the following radionuclides:

- ^{241}Am
- ^{238}Pu
- ^{239}Pu/^{240}Pu
- ^{233}U/^{234}U
- ^{235}U/^{236}U
- ^{238}U

Each filter was dissolved using a combination of hydrofluoric acid and nitric acid and microwave treatment. Americium, plutonium and uranium were each separated from the resulting solution by ion-exchange chromatography using Bio-Rad AG1-X8 resin (100-200 mesh). For each radioelement, a set of 3 alpha spectrometry sources was prepared. Each was counted on an ORTEC Octête alpha spectrometer in a manner traceable to national standards of radioactivity (e.g. by using appropriate standardised solutions as tracers). The results are given in Table 3.

The ratio ‘R’ of the true total alpha activity to the value derived from direct monitoring was then calculated for each filter sample and each monitor used. The ratios are given in Table 4 and illustrated in Figures 1 - 4. Note that, for the iSolo and JCS results, R is expressed as Bq (radiochemistry) / Bq (monitored), whereas for the EP15 and AP2 results it is expressed as Bq (radiochemistry) / cps (monitored).

4. Discussion

It is clear from Figure 1 that the ratio 'R' is typically in the range 1 – 5, (which encompasses the value of 2 cited above[1]), but that values of the order of 20 were observed in a few cases. There is no obvious correlation between filter type and R, and the visual appearance of nearly all the filters makes it difficult to correlate R with 'cleanliness'. This is particularly worrying as it could mean that, in practice, significant alpha absorption could be occurring within apparently 'clean' filters.

It is interesting to compare these results with those from a similar study carried out by Barnett et al.[5] at PNNL. They wanted to check the 'correction factor' of 0.85 then used at PNNL for its applicability to Versapor 3000 47mm filters. Note that their factor was effectively the inverse of 'R' as defined in the NPL study. Each filter was counted by gas-flow proportional counting in a planchet. The activity was removed from the filter by acid digestion and the digest then transferred to a clean planchet before being evaporated to dryness. The sample was then counted again by gas-flow proportional counting. For alpha-emitters, a mean ratio of counts before digestion to counts after digestion of 2.1 ± 2.9 (2σ) was obtained, indicating that, on average, *more* activity was detected by direct counting than by counting after acid digestion. The results were considered insufficient to quantitatively verify (or replace) their existing correction factor of 0.85.

Barnett et al. cited other previous studies[6,7] which recommended that alpha detection efficiency losses of up to 40% should be assumed for 'direct' filter measurements, depending on factors such as particle size, face velocity, filter type and dust loading. This implies a value or 'R' greater than 1.

The ratios of 'true to apparent' activity determined in the NPL study appear to be, in some cases, significantly larger than those observed in the above studies, and this may have significant implications for internal dosimetry of airborne alphas.

5. Conclusions

The observed ratios of 'true to apparent' activity from this short study are typically in the range 1 – 5, but factors of the order of 20 were found in a few cases. These are much larger than anecdotal values and values from the literature, which suggest α detection efficiency losses of the order of only 15 – 50%. There is evidence that, in some cases, the factor may even be less than 1 (i.e. more activity is detected by monitoring than by destructive analysis).

Given the wide range of correction factors obtained in the NPL study and elsewhere, and the fact that many data suggest that direct monitoring may underestimate activity and therefore internal dose, it is recommended that this study be taken further via a more rigorous analysis of filters. This might involve collecting samples for a wider range of nuclear facilities, or perhaps trialling different filter materials in several facilities to identify which, if any, give the more consistent value of 'R'. Research of this type could be complemented by the establishment of a UK radioactive aerosol facility to enable the various filter types to be exposed to well-characterised radioactive aerosols. Also, the authors would encourage operators to derive correction factors for their individual sites using methods such as those described above, and to publish the results. A better knowledge of these factors is needed to provide operators with confidence that worker intakes based on the results of air-filter measurements are not being underestimated.

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Table 1 – List of filters and filter type

Filter ID	Type
IM090182	25 mm GFA
IM090183	60 mm GFA
IM090184	25 mm GFA
IM090185	40 mm Bird and Tole paper filter card
IM090186	55 mm 541 Whatman filter paper
IM090187	55 mm 541 Whatman filter paper
IM090188	GFA Harwell card
IM090189	GFA Harwell card
IM090190	55 mm GFA filter paper
IM090191	40 mm Whatman GFA or 25 mm Fluorpore
IM090192	
IM090193	
IM090194	
IM090195	
IM090196	
IM090197	
IM090198	
IM090199	
IM090200	

Table 2 – Alpha activities derived from direct monitoring (uncertainties at $k = 1$)

Filter ID	iSolo (Bq)	Uncertainty (%)	JCS (Bq)	Uncertainty (%)	EP15 (cps)	Uncertainty (%)	AP2 (cps)	Uncertainty (%)
IM090182	0.983	6.6	1.348	5.6	-	-	0.43	8.8
IM090183	4.726	3.0	6.657	2.5	30	1.8	2.04	7.0
IM090184	2.252	4.4	2.833	3.9	18.3	2.3	0.75	6.7
IM090185	43.44	1.0	105.3	0.6	65.8	1.2	20.9	2.2
IM090186	30.51	1.2	63.52	0.8	23.7	2.1	12.9	2.8
IM090188	0.462	9.6	0.785	7.4	0.86	10.8	0.11	17.4
IM090189	114.3	0.6	167.2	0.5	24.7	2.0	29.3	1.8
IM090190	0.43	10.0	0.657	8.1	-	-	-	-
IM090191	0.178	15.5	0.2	14.6	3.06	5.7	0.15	14.9
IM090192	0.317	11.6	0.472	9.5	6.83	3.8	0.2	12.9
IM090193	0.756	7.5	1.118	6.2	10.75	3.0	0.37	9.5
IM090194	0.118	19.0	0.168	15.9	1.11	9.5	0.11	17.4
IM090195	0.178	15.5	0.217	14.0	1.99	7.1	0.133	15.8
IM090196	0.106	20.1	0.16	16.3	1.57	8.0	0.11	17.4
IM090197	0.093	21.4	0.155	16.6	1.18	9.2	0.13	16.0
IM090198	2.256	4.4	2.941	3.8	18.3	2.3	0.96	5.9
IM090199	1.675	5.1	2.255	4.4	10.2	3.1	0.48	8.3
IM090200	6.632	2.5	8.946	2.2	4.62	4.7	2.68	6.1

Table 3 – Alpha activities from radiochemical analysis (individual radionuclide and total) (uncertainties at $k = 1$)

Filter ID	^{241}Am (Bq)	Unc. (Bq	^{238}Pu (Bq)	Unc. (Bq)	^{239}Pu / ^{240}Pu (Bq)	Unc. (Bq)	^{233}U / ^{234}U (Bq)	Unc. (Bq)	^{235}U / ^{236}U (Bq)	Unc. (Bq)	^{238}U (Bq)	Unc. (Bq)	Total alpha (Bq)	Unc. (Bq)	Unc. (%)
IM090182	0.74	0.03	0.1	0.01	0.09	0.01	0.45	0.17	0.69	0.2	0.23	0.11	2.30	0.29	12.46
IM090183	0.67	0.06	1.92	0.04	1.1	0.02	0.53	0.06	0.05	0.03	0.1	0.04	4.37	0.11	2.48
IM090184	1.25	0.08	0.47	0.01	0.33	0.01	0.38	0.07	-	-	-	-	2.43	0.11	4.41
IM090185	0.14	0.03	0.23	0.03	0.06	0.02	155.42	2.66	12.4	0.24	165.1	2.83	333.35	3.89	1.17
IM090186	0.67	0.03	1.43	0.14	0.1	0.04	449.96	9.35	16.87	0.38	55.6	1.18	524.63	9.43	1.80
IM090188	-	-	0.06	0.01	0.01	0.01	0.39	0.02	0.03	0.01	0.41	0.02	0.90	0.03	3.69
IM090189	0.76	0.22	0.01	0.02	-	-	272.55	5.8	11.63	0.31	34.41	0.79	319.36	5.87	1.84
IM090190	0.63	0.1	0.03	0.01	0.01	0.01	0.97	0.05	0.05	0.03	0.22	0.02	1.91	0.12	6.19
IM090191	0.15	0.13	0.06	0.01	0.09	0.01	0.35	0.03	0.06	0.02	0.11	0.02	0.82	0.14	16.72
IM090192	0.6	0.19	0.1	0.01	0.15	0.01	0.05	0.01	0.01	0.02	0.01	0.01	0.92	0.19	20.88
IM090193	1.05	0.1	0.14	0.02	0.25	0.02	0.17	0.03	0.02	0.02	0.04	0.02	1.67	0.11	6.69
IM090194	0.39	0.17	0.21	0.01	0.07	0.01	0.04	0.01	0.02	0.01	0.02	0.01	0.75	0.17	22.86
IM090195	0.6	0.05	0.07	0.01	0.09	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.80	0.05	6.85
IM090196	1.57	0.52	0.14	0.02	0.12	0.01	0.06	0.02	0.03	0.01	0.02	0.01	1.94	0.52	26.86
IM090197	0.21	0.06	0.06	0.01	0.06	0.01	0.03	0.01	0.01	0.02	0.01	0.01	0.38	0.07	17.46
IM090198	1.55	0.84	0.68	0.02	1.15	0.03	0.07	0.01	0.01	0.01	0.01	0.01	3.47	0.84	24.23
IM090199	1.77	0.28	0.29	0.02	0.88	0.02	0.1	0.02	0.09	0.02	-	-	3.13	0.28	9.04
IM090200	6.3	4.74	1.4	0.04	2.81	0.06	0.1	0.01	-	-	0.03	0.01	10.64	4.74	44.55

Table 4 – Ratio ‘R’ of alpha activity (radiochemical analysis) to alpha activity or counts per second (direct monitoring) (uncertainties at $k = 1$)

Filter ID	R, iSolo	Unc. (%)	R, JCS	Unc.(%)	R, EP15	Unc. (%)	R, AP2	Unc. (%)
IM090182	2.3	14.1	1.7	13.7	-	-	2.8	15.3
IM090183	0.9	3.9	0.7	3.5	0.1	3.1	1.1	7.4
IM090184	1.1	6.2	0.9	5.9	0.1	5.0	1.7	8.0
IM090185	7.7	1.5	3.2	1.3	5.1	1.7	8.3	2.5
IM090186	17.2	2.2	8.3	2.0	22.1	2.7	21.1	3.3
IM090188	1.9	10.3	1.1	8.2	1.0	11.4	4.3	17.8
IM090189	2.8	1.9	1.9	1.9	12.9	2.7	5.7	2.6
IM090190	4.4	11.7	2.9	10.2	-	-	-	-
IM090191	4.6	22.8	4.1	22.2	0.3	17.7	2.8	22.4
IM090192	2.9	23.9	1.9	22.9	0.1	21.2	2.4	24.5
IM090193	2.2	10.1	1.5	9.1	0.2	7.4	2.3	11.6
IM090194	6.4	29.7	4.5	27.9	0.7	24.8	3.5	28.7
IM090195	4.5	16.9	3.7	15.6	0.4	9.9	3.1	17.2
IM090196	18.3	33.5	12.1	31.4	1.2	28.0	9.2	32.0
IM090197	4.1	27.6	2.5	24.1	0.3	19.7	1.5	23.7
IM090198	1.5	24.6	1.2	24.5	0.2	24.3	1.9	24.9
IM090199	1.9	10.4	1.4	10.0	0.3	9.6	3.4	12.3
IM090200	1.6	44.6	1.2	44.6	2.3	44.8	2.1	45.0

Figure 1 – Ratio ‘R’ using iSolo monitor

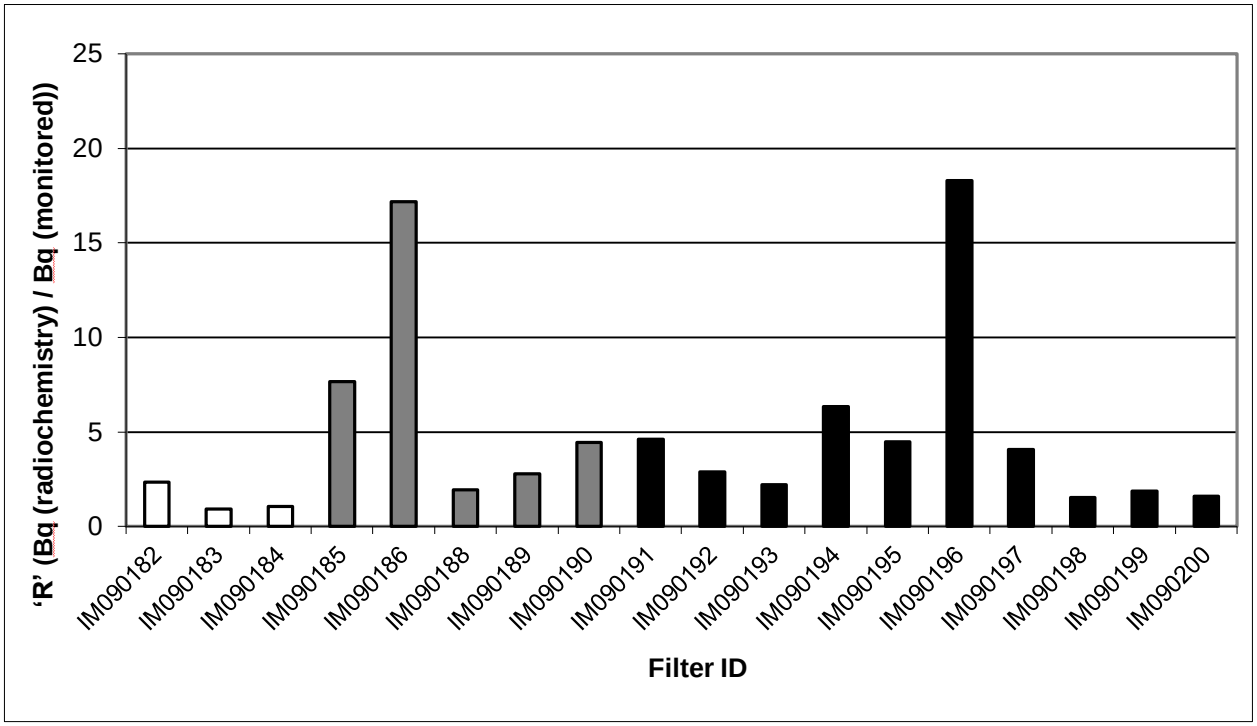

Figure 2 - Ratio ‘R’ using JCS monitor

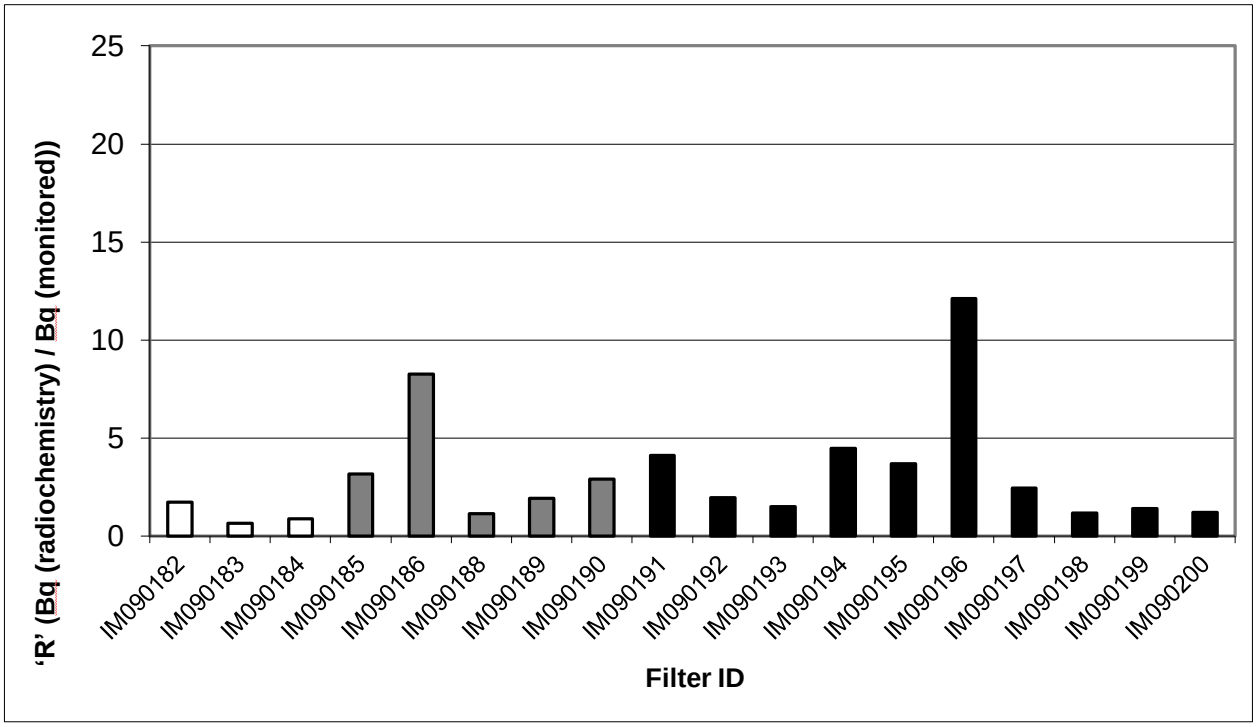

Figure 3 – Ratio ‘R’ using EP15 monitor

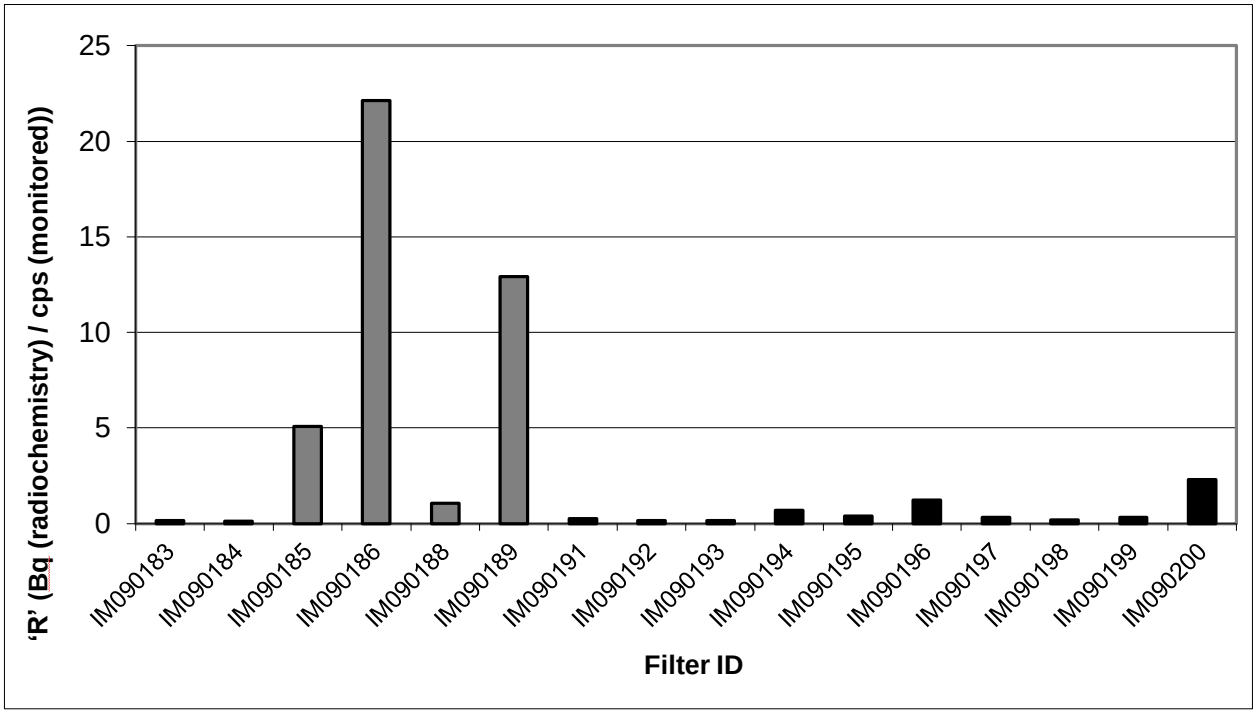

Figure 4 – Ratio ‘R’ using AP2 monitor