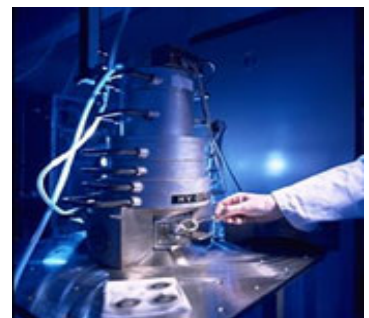
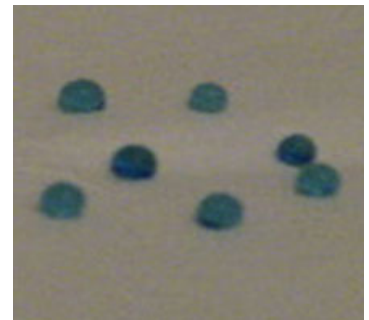


Evaluation of EC measurement comparison on simulated airborne particulates - ¹³⁷Cs in air filters

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SUMMARY

This report describes the full life cycle of the measurement comparison of ^{137}Cs in air filters among 43 European laboratories monitoring radioactivity in the environment. Gravimetrically pipetting droplets of a gravimetrically diluted standardised ^{137}Cs solution onto real air filters, SI-traceable reference values were established for intercomparison filters carrying a large range of activity close to the routine measurement conditions of the corresponding laboratory. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the intercomparison are presented and discussed in detail.

A robust evaluation of the performance of individual laboratories is performed using activity ratios, relative deviations and E_n numbers. Seven out of 48 reported measurement results have relative deviations larger than $\pm 33\%$ from the IRMM reference value (or yield activity ratios outside the range of 0.67 to 1.33). In addition to five results from this group, another seven results (a total of 12) do not fulfil the criteria of the compatibility test based on E_n numbers. These results point at some problems of radioactivity measurement in air filters which need to be addressed by the concerned laboratories. Overall, with 41 out of 48 reported results falling within $\pm 33\%$ of the IRMM reference value, this comparison may be seen as having rendered a rather fair result.

It is observed that many of the reported uncertainty values are not estimated as combined uncertainty of the whole measurement process following the concepts of the "GUM approach" [8]. Some laboratories, however, are able to provide realistic uncertainty estimates consistent with the reported results.

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GLOSSARY

BIPM	Bureau International des Poids et Mesures
CIEMAT	Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
GUM	Guide to the Expression of Uncertainty in Measurement [8]
IRMM	Institute for Reference Materials and Measurements
ISO	International Organization for Standardization
LSC	liquid scintillation counter, liquid scintillation counting
NIST	National Institute of Standards and Technology
PPC	pressurised proportional counter
SI	Système International d'Unités, International System of Units
SIR	Système International de Référence, International Reference System
UTC	Coordinated Universal Time
A	activity measured by participating laboratory
A_0	activity reference value, spiked activity
k	coverage factor according to GUM
E_n	performance statistic E_n number
s	standard deviation, standard uncertainty in counting alone
u_c	combined standard uncertainty according to GUM
U	expanded uncertainty according to GUM
U_{lab}	expanded uncertainty of laboratory result
U_{ref}	expanded uncertainty of reference value
U_{lab}^*	relative expanded uncertainty of laboratory result, U_{lab}/A
U_{ref}^*	relative expanded uncertainty of reference value, U_{ref}/A_0
x	laboratory result
X	reference value

1. Introduction

Under the EURATOM Treaty of 1957, covering responsibilities in the peaceful use of nuclear energy in the European Union, Member States are obliged to monitor radioactivity levels in the environment of their countries (Art. 35) and to regularly report the measured values to the European Commission (Art. 36). Networks for routine and emergency measurement and communication of radioactivity values have been established. The Commission Recommendation 2000/473/Euratom of 8 June 2000 specifies in detail what environmental and food matrices should be monitored for which radionuclides.

In order to obtain more information on the measurement methods and on the quality of the values reported by the Member States, measurement comparison exercises have been conducted regularly by the European Commission through its Joint Research Centre (JRC). Since 2003, the JRC Institute for Reference Materials and Measurements, IRMM, organises these measurement comparisons as support to the Directorate General for Energy and Transport (DG TREN H.4).

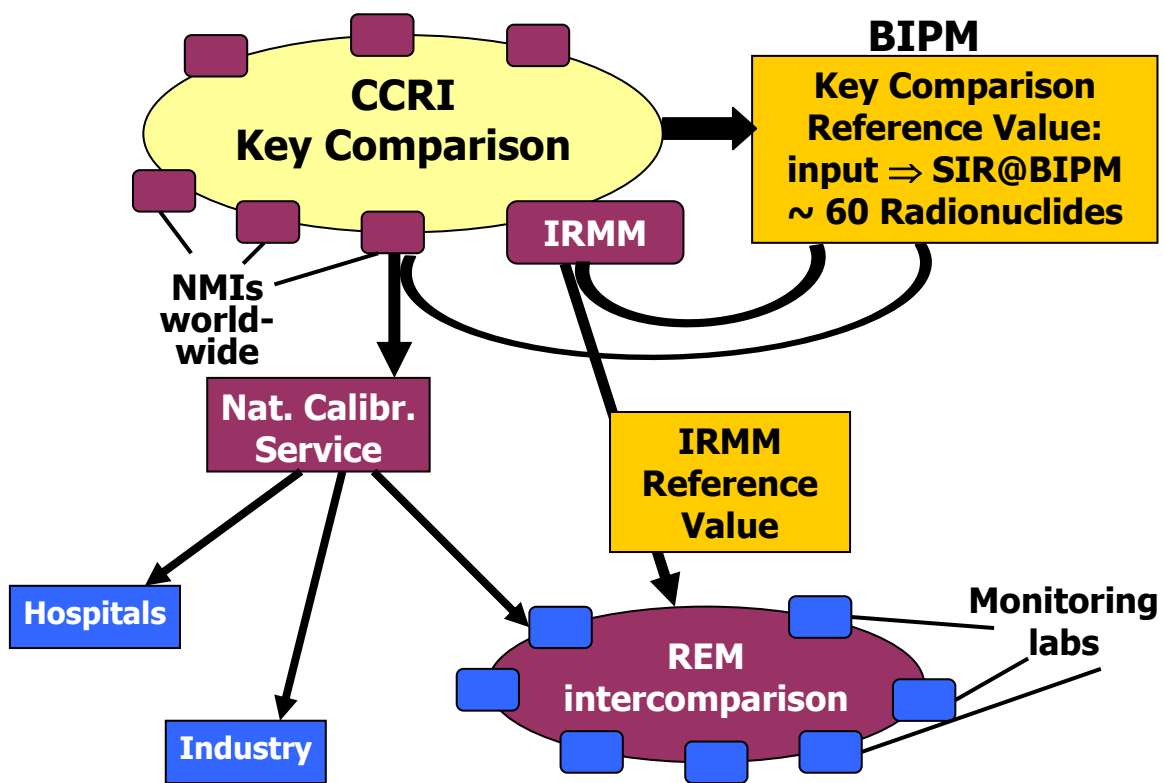


Fig. 1: Key comparison, traceability and intercomparison for monitoring laboratories

The approach of IRMM in organising the comparisons is sketched in Figure 1. As member of the Consultative Committee for Ionising Radiation (CCRI), IRMM is participating in key comparisons among National Metrology Institutes (NMIs), which serve to "realise" the unit of radioactivity, the becquerel. Results of key

comparisons, which are based on primary measurement techniques, i.e. without resorting to other activity standards, are introduced in the International Reference System for Radionuclides (SIR) to determine the SIR calibration factor for that particular nuclide. Since each nuclide has its own decay scheme, all calibration factors are different and need to be experimentally determined for each nuclide. Thus, the SIR system at BIPM, the Bureau International des Poids et Mesures in Sèvres close to Paris, is the world-wide standard for radioactivity, realised as an ionisation chamber.

Due to IRMM's participation in key comparisons and the direct link to the SIR, it can work with standardised solutions which are directly traceable to the SI unit. National metrology institutes, having the same short traceability link to the SIR, usually provide calibration standards through their national calibration services. Such standards can be used for example by air monitoring laboratories to calibrate their measurement equipment. Parallel to this traceability chain, IRMM offers intercomparisons with samples which have their own traceable reference value. Usually such samples are, in physical properties as well as amount of radioactivity, closer to the routine measurement conditions of a monitoring laboratory than the calibration standards. Thus, this kind of intercomparison can serve as an independent and impartial performance check with samples of high credibility.

This report presents the results of the intercomparison exercise organised by IRMM (Action 2111 and Action 3142) together with the JRC Institute for Environment and Sustainability (IES, Action 2114) on ^{137}Cs -spiked air filters. Although radionuclides in air belong to the most often measured environmental radioactivity components (^{137}Cs and gross beta), they have not been subject of a European comparison so far. Therefore, ^{137}Cs in air filters was chosen for the first comparison organised by IRMM.

The principal objective of this exercise was to give the opportunity to individual laboratories for checking their procedures of measuring the major anthropogenic gamma-ray emitter in environmental air filters, and at the same time to provide direct evidence of the comparability of the results from different laboratories to the benefit of both users (EC DG TREN, Member State authorities) and laboratories. In addition, where possible, the extent of variation should be quantified, and possible causes pointed out.

Description of the sample

Nature: blank air filters provided by the participating laboratories and spiked with a standardised ^{137}Cs solution at IRMM

Reference date: 1 October 2003

Recommended half-life of ^{137}Cs : $T_{1/2} = (10981 \pm 11)$ days = (30.065 ± 0.030) years [7], where the numbers following the symbol \pm are the numerical values of an expanded uncertainty U with a coverage factor $k = 1$, corresponding to a level of confidence of about 68 %

Activity levels: approximate activity measured under routine monitoring conditions or slightly higher than the detection limit of the

individual laboratory (^{137}Cs activity ranging from 29 mBq up to 570 mBq)

Shipping: in plastic bags via regular mail

Required reporting of the results

^{137}Cs activity reported as the measured value (Bq/filter) with the associated uncertainty (counting uncertainty alone and expanded uncertainty $U = k \cdot u_c$, with U determined from the combined standard uncertainty u_c and a coverage factor $k = 2$, corresponding to a level of confidence of about 95 %).

Participating laboratories

The exercise was designed to evaluate the performance of the participating laboratories in measuring ^{137}Cs activity on air filters. Participating laboratories were mainly national research institutes and authorities in the EU Member States and the accession countries. The laboratories were nominated by the national representatives* in the expert group according to Euratom Treaty Art. 35 and 36.

In total 43 laboratories (25 from the Member States**, 12 from the Acceding Countries**, 4 from the Candidate Countries**, and 2 from Norway) participated in the intercomparison exercise (the list and addresses of the laboratories are presented in chapter 15). Since anonymity is a requirement in this programme of measurement comparisons, the identity of the laboratories is not shown in the compilation of the results. The order of the listing of participants in chapter 15 is not the same as the laboratory number used throughout the data evaluation and comparison.

Timetable and deliverables

1 Sept. 2003	the participating laboratories are nominated by the national representatives
15 Sept. 2003	the laboratories send back the completed questionnaire on air sampling and radionuclide measurement techniques and provide 2 unused air filters
15 Oct. 2003	standardisation, dilution, quantitative spiking, quality control of air filters at IRMM
17 Oct. 2003	spiked air filters are sent to the participants
20 Nov. 2003	reporting sheets are sent to the laboratories
31 Jan. 2004	laboratories submit their results to IRMM

* They generally represent their national regulatory bodies for radiation protection.

** Status as of end of 2003.

2. Routine sampling of the laboratories participating in the exercise

In the preparative phase of the exercise the participants were asked to fill in a questionnaire that focused basically on technical information on their routine air sampling and radioactivity measurement methodology (Annex 1). Two sets of questions, namely on sampling procedures and on measurement techniques were released. The responses obtained for the second part of the questionnaire regarding the gamma-spectrometric measurements will be briefly summarised in chapter 8 focusing mainly on the measurement of the spiked samples (air filters) distributed in the intercomparison. In the present chapter, the air sampling methodology applied in the participating laboratories is described.

Information about the supplier and type of air sampler, type of size separation (if any is used), cut-off size, air volume flow rate, type of flow control and flow measurement, total volume of air sampled, sampling frequency and sampling period were collected and analysed to have a better understanding of the characteristics of each system. The answers obtained regarding the supplier and type of filter material, the shape and size of filter, the blank weight and the typical aerosol deposit collected helped to plan the spiking of the air filters.

As was expected, air sampling procedures differ widely among (and even within) the countries. A large variety of commercial and home-made air sampler types are being used, with a variety of filter media in sizes ranging from 25 mm diameter to 140 cm x 80 cm. Accordingly, the sampled air volume per filter ranges from 80 m³ to 350 000 m³, and the ¹³⁷Cs activity per measured filter (or stack of filters) varies from < 5 mBq to 4 Bq. In order to approach realistic measurement situations with the intercomparison samples, it was decided to request blank filters from the participating laboratories and to apply droplets of a diluted standardised ¹³⁷Cs solution with an individual total activity close to that routinely measured by the corresponding laboratory – but in any case above the declared detection limit – instead of distributing identical filters to all participants.

In the following, some details extracted from the questionnaire response are presented. Next to specially designed and home-made air samplers, the following commercial air samplers are being used: PTI-ASS 500/1000, Senya Hunter/Snow White, F&J different models, General Metal Works, other commercial types. The air volume flow rate varies from 3 to 2100 m³/h (for large commercial samplers the typical value is approximately 900 m³/h) being influenced by the filter loading and the dimension of filters used. The volume of sampled air varies between 60 and 4 000 m³ for small samplers with circular filters, from 6 000 to 150 000 m³ for medium and large commercial samplers accepting rectangular filter sheets and reaching up to 350 000 m³ (in two laboratories) with special constructions.

In many cases the samplers do not incorporate a flow control system, the flow rate being determined by the loading of the filter or being adjusted manually. In few cases frequency converters controlling the motor revolution of the pump are applied to compensate for the increase of resistance on the loaded filters. Instead, mass, volume and pressure difference flow meters are used to measure the air flow. The reported calibration of these instruments varies from “no calibration” to

“occasional check with a Rotameter” and from “factory calibrated” to “instruments calibrated against each other” and “calibrated by certified institutes every 2 years”.

An important parameter of the whole exercise was the knowledge of filter material, its size and shape determining in fact the spiking methodology. A huge variety of filters are used; the characteristics of these materials are summarised in Table 1. The shapes and sizes of the filters (represented in Table 1 by surface area) together with the measurement geometry and the typical ^{137}Cs activities measured in each laboratory were in fact the critical parameters which determined the spiking conditions. The air filters are commercial types such as Whatman GF/A, F&J, PTI, paper fibre, polypropylene fibre, cellulose and polycarbonate filters with filter sizes ranging from 2.5 to 12 cm in diameter, and from 23 x 18 cm² with a larger group around 44 x 44 cm² to 140 x 80 cm² surface area.

The typical environmental ^{137}Cs activity measured by the participating laboratories varies from < 4.7 mBq up to 4 Bq, corresponding to an activity concentration between 1.5×10^{-7} Bq/m³ and 45×10^{-7} Bq/m³.

Table 1: Type, shape and size of spiked air filters used in the intercomparison exercise

filter type	filter shape and size (cm)	active surface area (cm ²)
Czech Synpore (a kind of Nuclepore)	2.5 Ø	4.9
Russian FPP 15 ¹	4.7 Ø	17.3
Whatman GF/A ²	4.7 Ø	17.3
Schleicher&Schüll 1450 CV/HY	4.9 Ø	18.8
F&J paper fibre	5.1 Ø	20.2
Cellulose filter	5.1 Ø	20.4
Micro 2000, polycarbonate filter	9.0 Ø	63.5
NNC polypropylene filter	9.0 Ø	63.5
Whatman GF membrane filter	9.0 Ø	63.5
F&J polyester fibre	10.2 Ø	81.0
Staplex glass fibre TFAGF41	10.2 Ø	81.0
Macherey Nagel 640 W	10.3 Ø	83.2
paper fibre	11.3 Ø	100.3
Schneider-Poelman Ultra filters Air	11.3 Ø	100.3
Camfil C-577	12.0 Ø	113.0
Glass fibre	23.0 Ø	415
FPM 1515 R	17.5 x 22.5	394
NNC polypropylene filter	23 x 18	414
F&J FP810	20.3 x 25.4	516
Whatman GF/A	20.3 x 25.4	516
PES PC-S organic microfibre	23 x 28	644
Whatman GF	23.0 x 28.5	656
Camfil C-577	44 x 32	1408
Petrianov FPP 15 15-1,5	42 x 42	1764
PTI type G3, PP filter	42 x 42	1764
Petrianov FPP 15 15-1,5	44 x 44	1936
Schleicher&Schüll glass fibre no. 10	4 x 20 x 25	2000
Whatman GF/A	46 x 57	2622
Whatman GF/B	46 x 57	2622
PES PC-S organic microfibre	46 x 57	2622
PTI Petrianov polypropylene filter	54.5 x 54.5	2970
PTI type G3, PP filter	55 x 55	3025
fibre filter FNAE 1267	60 x 53	3180
PTI type G3, PP filter	58 x 58	3364
Camfil CS 5.0	61 x 61	3721
Estonian FPP filter	140 x 80	11200
PTI type G3, PP filter	6 x 56 x 48	16128

¹ PP = polypropylene, ² GF = glass fibre

3. Standardisation of the spike solution

Standardisation of the ^{137}Cs solution used for the spiking of air filters was performed by two different methods: **liquid scintillation counting** and **$4\pi\beta\text{-}\gamma$ coincidence counting**.

The **LSC method** used the technique developed by CIEMAT/NIST [1, 9-10] and was preceded by measurements checking for possible impurities. A small ^{134}Cs impurity was determined in the solution by HPGe gamma-ray spectrometry (see below), but being negligible its value was taken into account only in the uncertainty budget calculations.

All sources were prepared gravimetrically using a Mettler AX26 mass comparator, calibrated using weights traceable to the IRMM reference kilogram, which in turn is directly traceable to the SI unit of mass by comparisons at BIPM. The radioactive solution was dispensed by means of a pycnometer into the 20 mL Packard High Performance LSC glass vials containing the LS cocktail. Sources were prepared in three different cocktails: 15 mL Ultima Gold + 1 mL H_2O , 15 mL Insta-Gel Plus and 15 mL plain Ultima Gold cocktail.

20 samples were prepared in this way, the amount of radioactive solution in each sample ranging from 10 to 52 mg. All samples were measured 9 times (for 10 minutes each) using the Packard 3100 TR/AB LSC and 3 times (for 10 minutes each) using the Wallac Quantulus 1220 LSC over a period of 2 months.

The standardisation methodology including the ^{137}Cs nuclear data, the liquid scintillation parameters, the specification of the counting device and the measurement parameters used for standardisation of the ^{137}Cs solution together with the detailed LSC results can be found in Annex 3 (reproduced from [3]).

Due to the existence of a delayed isomeric state (half-life: 2.552 minutes) of the ^{137}Ba daughter of ^{137}Cs , giving rise to a 661.6 keV gamma-ray transition, ^{137}Cs may not readily be standardised by the **$4\pi\beta\text{-}\gamma$ coincidence counting technique**. It was for this reason that a standardisation of ^{134}Cs solution was performed to facilitate the standardisation of the ^{137}Cs solution by the 'efficiency tracing' technique.

This procedure was performed for a set of mixed sources, and the weighted mean of the results determined to yield the activity concentration of the ^{137}Cs solution.

The standardised solution of ^{134}Cs was quantitatively mixed with the ^{137}Cs solution. The 'mixed' sources were then considered as 'pseudo single-nuclide' sources, and were measured in the $4\pi\beta(\text{PPC})\text{-}\gamma(\text{NaI}(\text{TI}))$ coincidence counting equipment, where the apparent beta efficiency was determined and by extrapolations performed to unit beta efficiency the apparent total activity of the source was determined.

The presence of gamma-ray emitting impurities in both ^{134}Cs and ^{137}Cs solutions was investigated by HPGe gamma-ray spectrometry [2-3]. In the ^{134}Cs solution no impurities, whereas in the ^{137}Cs solution the contaminant ^{134}Cs was found. The ^{134}Cs to ^{137}Cs ratio was 0.013(1) % at reference date: (2003-07-01, 0h UTC). This contribution from the contaminant was taken into consideration in the calculations of both the activity concentration of ^{137}Cs and its associated uncertainty.

All sources were prepared gravimetrically by mass substitutional weighing, using a Mettler AT21 mass comparator, calibrated using weights traceable to the IRMM reference kilogram. The radioactive solutions (carrier 50 $\mu\text{g Cs}^+/\text{mL}$ as CsCl in 0.1M HCl) were dispensed by means of a pycnometer onto VYNS foil substrates ($20 \mu\text{g}\cdot\text{cm}^{-2}$) which were coated on both sides with gold ($13.5 \mu\text{g}\cdot\text{cm}^{-2}$). The evaporation process was assisted by employing the IRMM source drying device, which is using hot jets of dry nitrogen to limit crystal formation during evaporation to small crystal sizes [4].

The equipment setup used for $4\pi\beta\text{-}\gamma$ coincidence counting, the calculations and extrapolations performed and the results obtained as well as the uncertainty components taken into account are detailed in [5], an excerpt of which is reproduced as Annex 4.

The results from the $4\pi\beta(\text{PPC})\text{-}\gamma(\text{NaI}(\text{TI}))$ coincidence counting primary standardisation and the CIEMAT/NIST LSC standardisation are summarised in Table 2, where the numbers in parentheses are the numerical values of the combined standard uncertainties u_c expressed in the unit of the quoted result. Both measurement methods render the same activity value for ^{137}Cs .

Table 2: The results of the standardisation of ^{134}Cs and ^{137}Cs solutions by two different techniques

Nuclide	Activity concentration at reference date* (2003-07-01, 0 h UTC) (kBq/g)	
	$4\pi\beta(\text{PPC})\text{-}\gamma(\text{NaI}(\text{TI}))$ coincidence counting	CIEMAT/NIST LSC technique
^{134}Cs	190.4 (0.8)	189.1 (0.8)
^{137}Cs (mother solution A1)	270.0 (1.7)	270.0 (2.0)

The standardised solutions of both ^{134}Cs and ^{137}Cs were submitted together with the results of the described primary standardisations at IRMM as entry into the International Reference System (SIR) of the Bureau International des Poids et Mesures, BIPM, in order to establish traceability of the activity values in the spiking solutions and subsequently of the reference values of spiked activities on filters. The results of the measurements in the SIR at BIPM, summarised in Annex 5, are confirming traceability (to the key comparison reference value KCRV) within the combined relative measurement uncertainties of 0.8 %.

* It should be noted that the reference date given in Table 2 refers to the standardisation measurements only. For the subsequent intercomparison of air filters – incl. the reference values of ^{137}Cs activity in the filters – the reference date 1 October 2003 was chosen.

4. Dilution technique

In order to approximate the activity level measured in each participating laboratory under routine conditions with the appropriate amount of spiked ^{137}Cs on the filters, three different diluted solutions (C1, C2, C3) were prepared from the standardised mother solution (A1) via an intermediate dilution (B1). Table 3 lists the solutions and dilution factors including the addition of methylene blue, used to visualise the spikes on the filters. The numbers in parentheses are the numerical values of u_c .

Table 3: Dilutions and dilution factors with combined standard uncertainties u_c

Solution	Code	Dilution factor D
Mother	A1	1
Dilution	B1	1160.4 (1.9)
Dilution	C1	1421 (3) $\times 10^2$
Dilution	C2	5017 (9) $\times 10^2$
Dilution	C3	2980 (11) $\times 10^3$

The schematics of the different dilution steps are presented in Figures 2 to 4, where “P” indicates a pycnometer, weighed before and after dispensing the solution to each ampoule or vial.

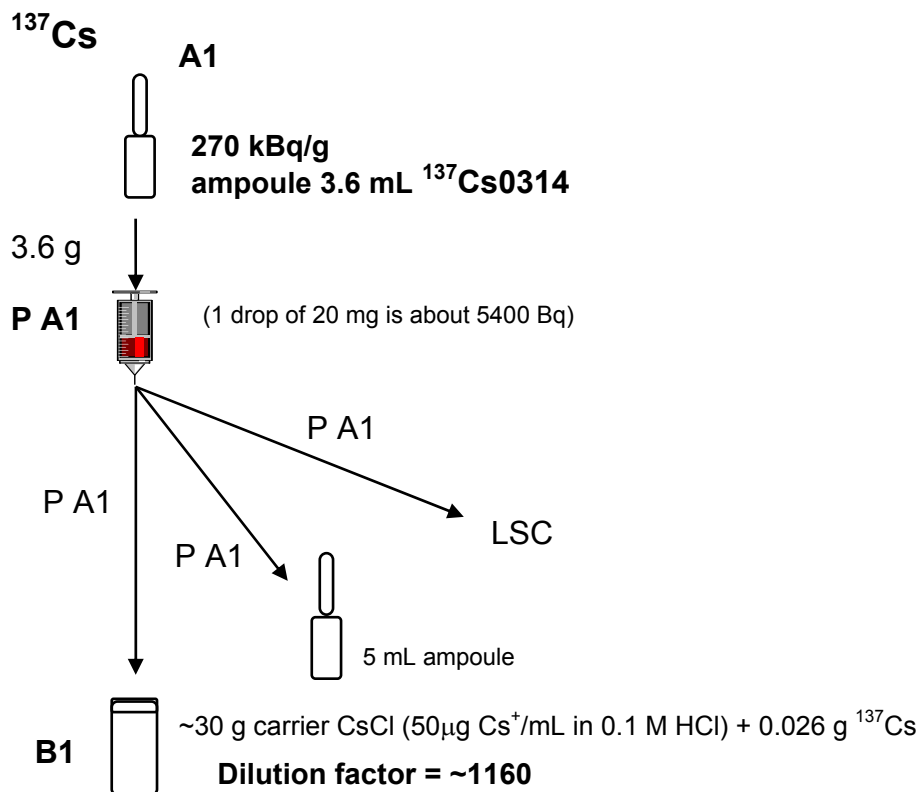


Fig. 2: Preparation of first dilution step B1

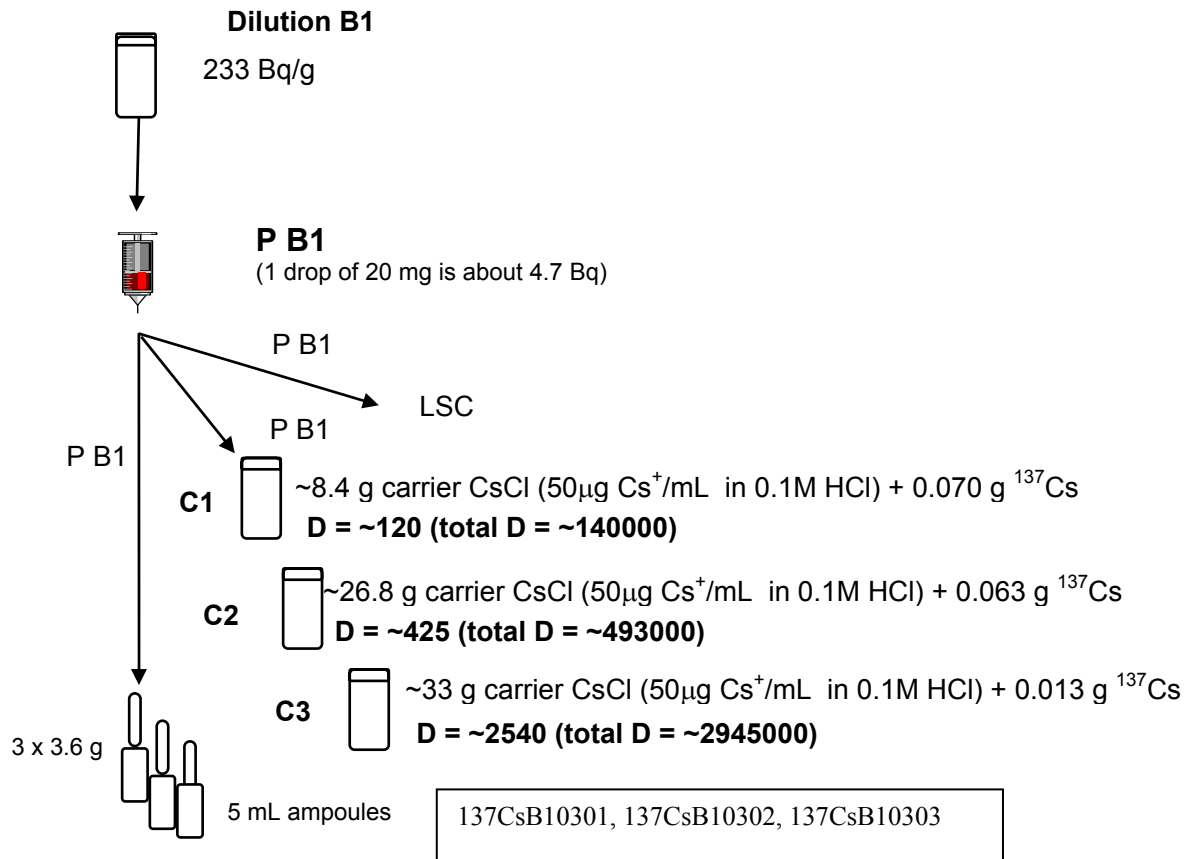


Fig. 3: Preparation of dilutions C1, C2 and C3

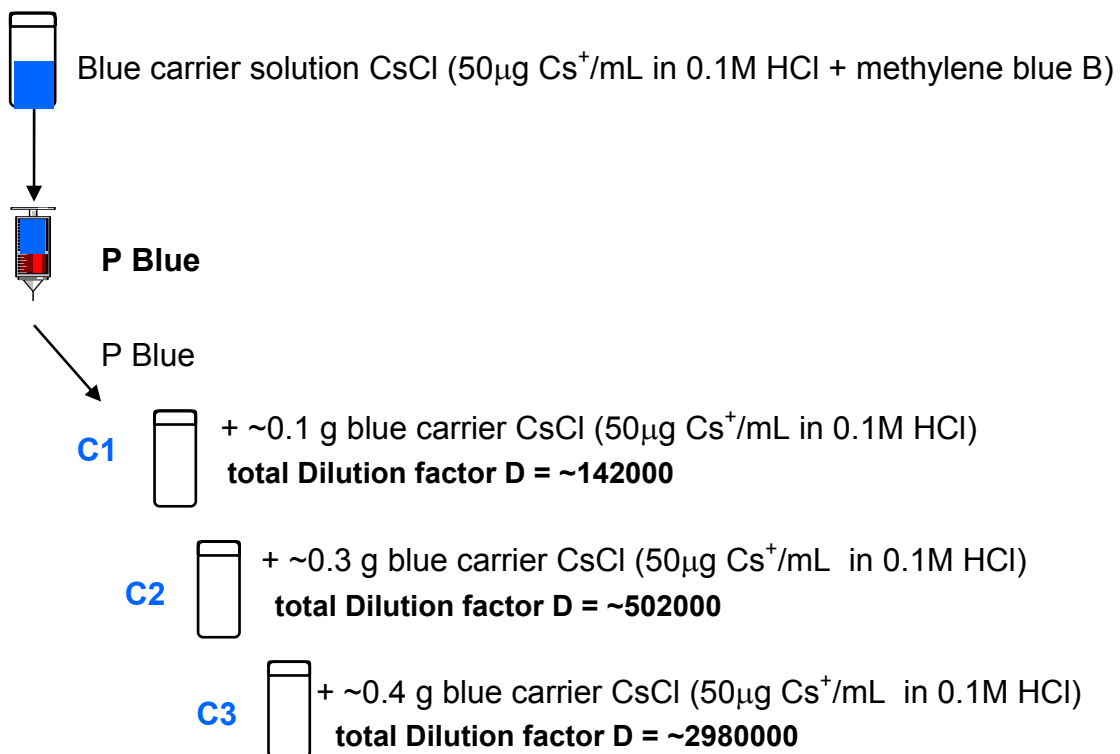


Fig. 4: Adding blue colour to the dilutions C1, C2, C3

It should be noted that all dilutions were prepared gravimetrically with the substitution principle applied for utmost traceability and small uncertainty on the dilution factors. As indicated in the schematics of Figures 2 to 4, samples from all dilutions were also prepared for quality control by liquid scintillation counting (LSC). The results of these measurements are presented in chapter 6 and Annex 6 and confirm – within the comparably large measurement uncertainty – the gravimetric dilution factors.

5. Spiking of the air filters

The IRMM Radionuclide Metrology Sector prepared about 50 air filters (Annex 7) by depositing on each of them gravimetrically an amount of ^{137}Cs from a standardised solution. Each participating laboratory in the exercise had sent a blank filter of the type it is routinely using and after spiking them at IRMM with the ^{137}Cs solution, the filters were distributed back to the participants in order to be measured according to their routine procedure. The amount of ^{137}Cs spiked onto each filter was chosen to resemble the activity routinely measured by the corresponding laboratory in a whole filter (or set of filters if it measures several at the same time). In cases where the laboratory declared to usually measure (sets of) filters with ^{137}Cs activities below detection limit (cf. the questionnaire, Annex 1) an activity higher than the declared detection limit was distributed.

Since a uniform distribution of spikes was not feasible, about 10 to 20 droplets were dispensed from the pycnometer, preferably arranged on circles of about 10 cm diameter in the centre of larger filters. Some laboratories requested a different spike distribution (Table 4), which was, in their opinion, better suited to mimic the situation of a more or less uniform activity distribution on real dust-loaded air filters. In any case, the spiked spots could easily be distinguished due to their methylene blue colouring, which allowed accounting for their discrete distribution when preparing the filters for measurement. The applied spikes ranged between 29 and 560 mBq per filter, having combined standard uncertainties between 0.2 and 4 mBq, respectively (cf. Table 5, discussed in detail below in chapter 9, "Results").

Table 4: Patterns of spike distribution on blank filters of various sizes

filter shape	filter size / mm	number of filters	pattern
Small round	< 120 Ø	15	standard
Large round	230 Ø	1	droplets on circle of 50 mm Ø
Small	< 230 x 290	9	standard
Small	< 230 x 290	1	along diagonal line
Medium	~ 400 x 400	4	standard
Medium	~ 400 x 400	1	along diagonal line
Large	< 600 x 600	6	standard
Large	< 600 x 600	3	along lines parallel with edge
Large	< 600 x 600	2	many droplets distributed over all surface
Large	< 600 x 600	1	mineral ballast added for ashing
Very large	1400 x 800	1	standard

Where the filter was large enough, the filter was folded up in a way that the active part came into the centre of the pack such that any salt “falling off” during transport by regular mail would still be caught in the surrounding filter and thus would not be lost. In the case of small filters this solution was not possible, therefore, the laboratories were asked to also measure the empty plastic bag in which the samples were shipped (or to measure the filter inside the bag) in order to verify that no losses from the filter had occurred. Only five laboratories reported of such measurements of empty shipping bags; none of them found any contamination.

The differences in the spiking technique can be easily distinguished in the photos attached in Annex 8. In many of the filters (e.g. those made of polypropylene or glass fibres) spiking became difficult due to the hydrophobic nature of the filters. The drops of radioactive solution were not absorbed into the filter material, therefore, the spiking had to be followed by several hours of drying at room temperature. One of these examples can be seen in Fig. d) of Annex 8.

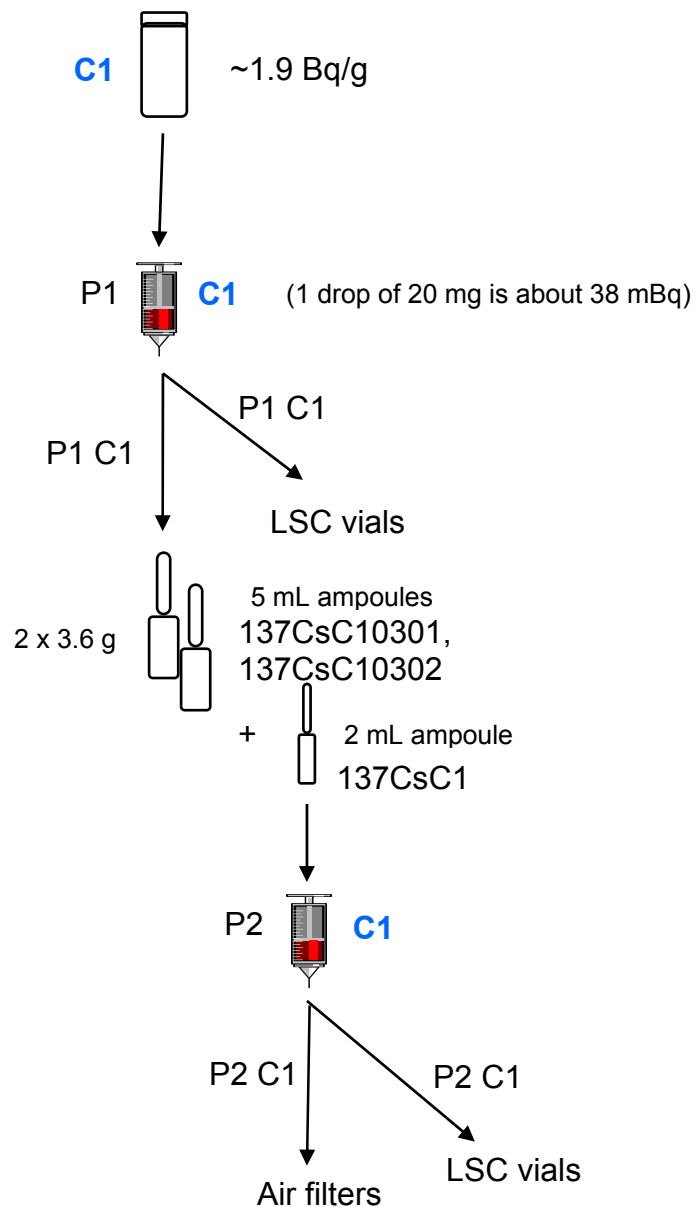


Fig. 5: Preparation of air filters from dilution C1

Details of the air filter spiking from the three dilutions C1, C2 and C3 are presented in Figures 5 to 7, where “P” indicates the pycnometer, weighed before and after dispensing the radioactive solution to each individual air filter, ampoule or vial. A Mettler AX26 mass comparator was used, calibrated with weights traceable to the IRMM reference kilogram and thus to the SI unit of mass.

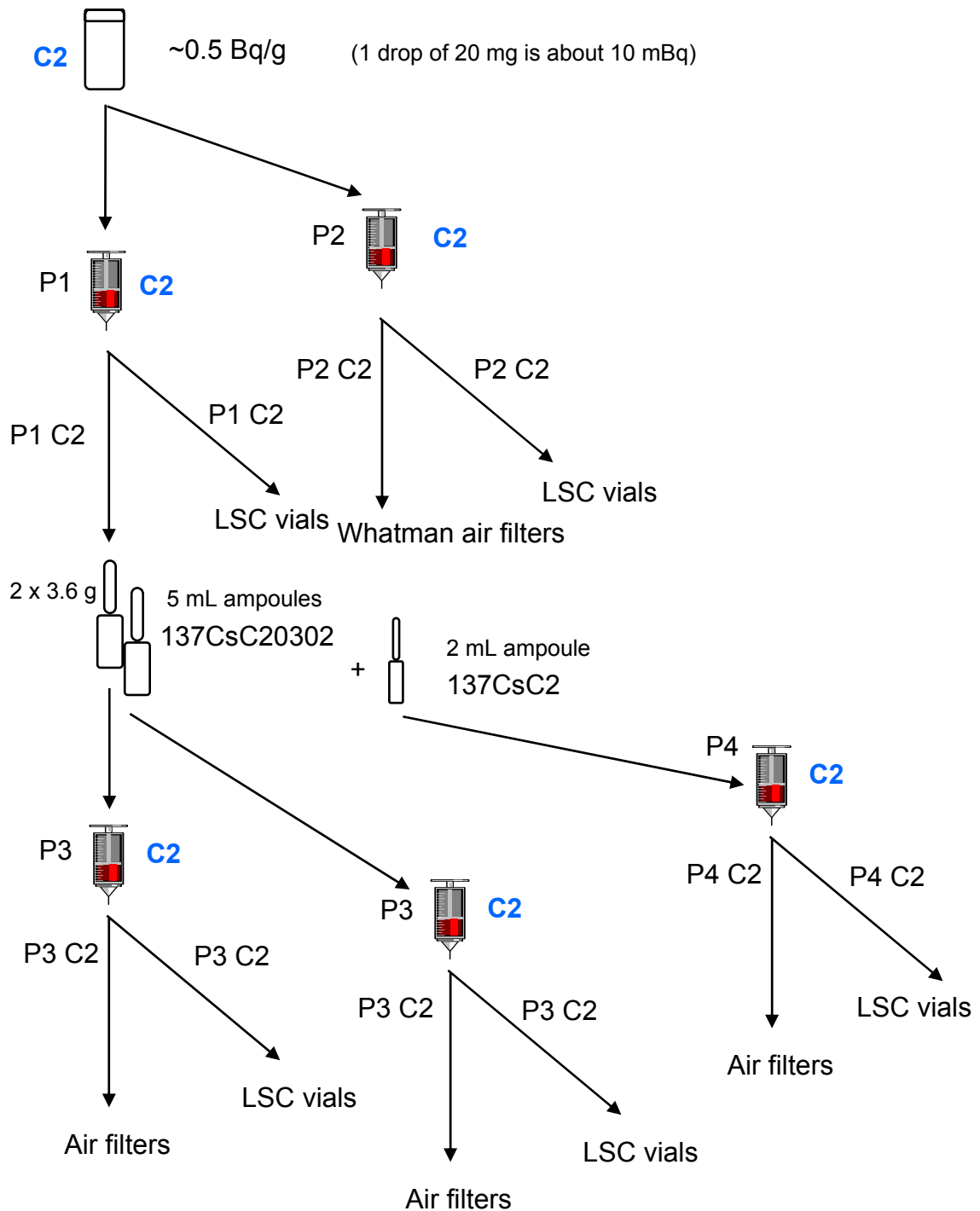


Fig. 6: Preparation of air filters from dilution C2

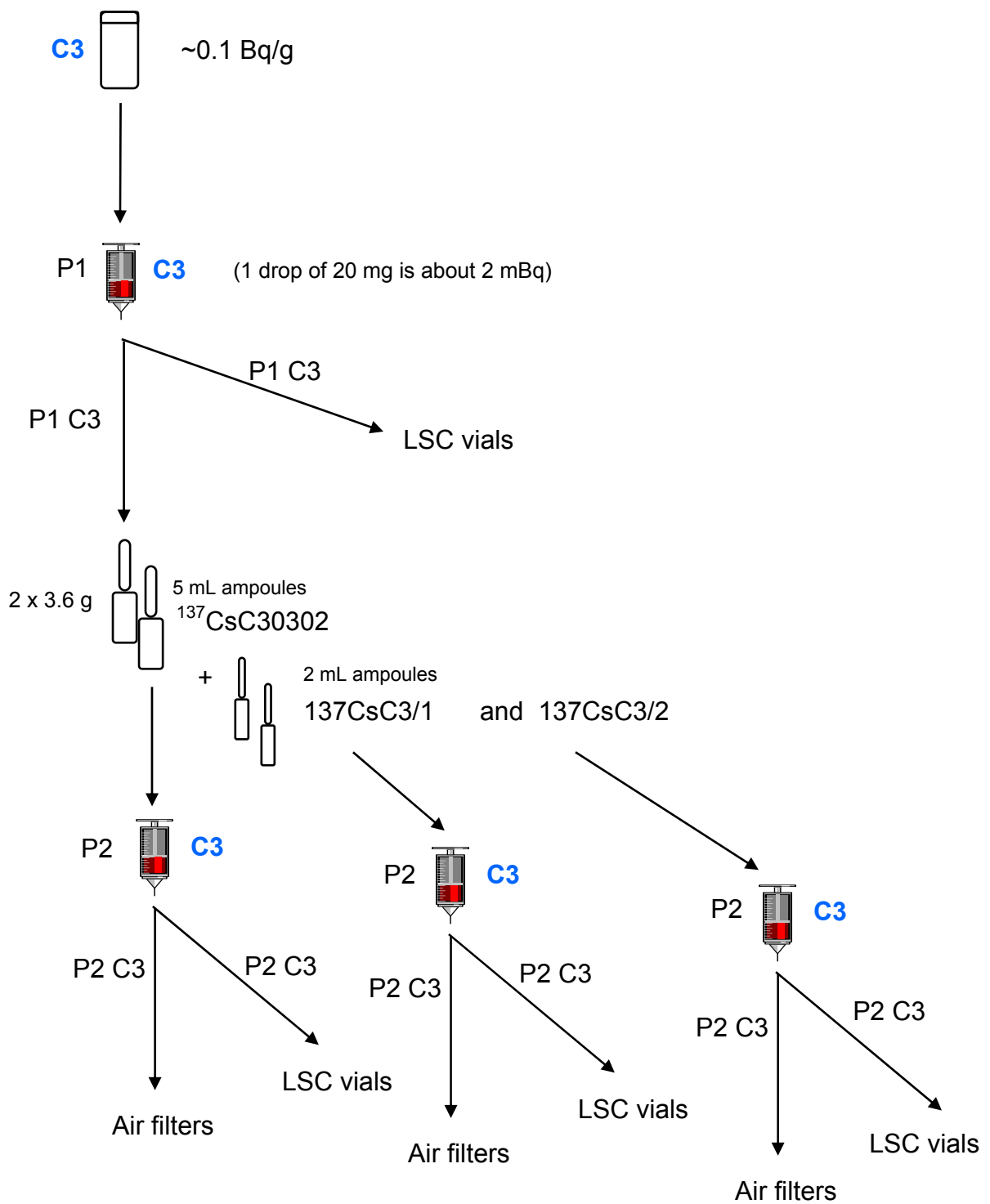


Fig. 7: Preparation of air filters from dilution C3

6. Quality control measurements

As is indicated in the filter spiking schematics of Figures 5 to 7, control samples were taken of all spiking solutions (C1, C2 and C3) in various phases of the procedure. In addition to 37 vials for liquid scintillation counting (LSC), 3 additional filters (Whatman 45 mm Ø) were prepared. The radioactive solutions were dispensed in the same way with a pycnometer into the LSC vials (20 mL Packard High Performance glass vials) containing 15 mL Ultima Gold + 1 mL H₂O cocktail. Annex 7, reproduced here from the quality control report [6], lists all filter samples prepared for the participants as well as all control samples. It shows clearly, how well the quality control samples interleave the rest of the samples prepared.

The 3 control filters were measured by gamma-ray spectrometry at IRMM, using a 36% relative efficiency HPGe detector for counting times of 5.5, 6.5 and 3.2 days, respectively. The LSC samples were measured using the Packard 3100 TR/AB LSC (for 360 minutes each) and the Wallac Quantulus 1220 LSC (vials from A1 for 10 minutes each, B1 for 120 minutes, C1 and C2 for 600 minutes and C3 for 1000 minutes). A relative efficiency calibration of the liquid scintillation counters was used, normalising the measured activity concentration of the samples from dilution B1 to the expected activity concentration in these vials based on the gravimetric dilution.

The results of the quality control measurements [6] for the three control filters and the 37 LSC samples of spiking solutions are given in detail in Annex 6 (all results expressed for the reference date 2003-07-01 0:00 UTC). All samples of the least active spiking solution C3 (QC samples # 30 to 37) were re-measured with prolonged counting times of 3 to 6 4-hour counting periods interleaved with equally long background measurements, resulting in total counting times of 1400 to 2900 minutes (shown in section G of Annex 6). Due to the long time elapsed between LSC sample preparation and measurement of this series G, however, some of the LSC samples can have become instable. In the tables of Annex 6, the measurement uncertainties are given as relative combined standard uncertainties and the major contributions come from the counting statistics and the background variation. In the graphs, the solid line indicates the activity concentration calculated from the gravimetric dilution and the dashed lines indicate the expanded uncertainty ($k = 2$) on the calculated activity concentration. The deviation given in the result tables and graphs is the relative deviation of the measured activity value from the gravimetrically determined one.

In addition, the ampoules used to store the solutions were rinsed and measured in order to detect a possible adsorption of ¹³⁷Cs on the glass walls. The results were negative, no significant adsorption was detected, and, hence, such loss was not taken further into consideration.

In conclusion, the measurement results confirm – within the possibilities of the rather large uncertainty of these LSC measurements – the activity concentrations determined from the gravimetric dilution and spiking procedure. One can safely conclude that the activity spiking procedure was applied successfully.

7. Filter preparation techniques and measurement geometries

Basically six different types of sample preparation were applied in the participating laboratories, namely: “no preparation”, “folding”, “folding and pressing”, “ashing”, “ashing and leaching”, “acid digestion”. These differences are partly due to the large variety in filter type and surface area mentioned earlier in chapter 2. The resulting large differences in sampled air volume and thus radioactivity require different sample preparation methods.

Most of the participants limited the filter preparation to simple mechanical treatments. In case of small circular filters, usually the complete filter area was exposed to the detector for direct measurement. For larger, mainly rectangular air filters, 10 laboratories used folding to confine the measured sample to the dimensions of containers placed directly on the endcap of the detector. 12 laboratories used folding and pressing to obtain the required sample dimensions of the filters, e.g. in the form of pellets. The applied hydraulic or pneumatic presses were set to 10 to 30 kN or the pressing was ended when the prescribed measurement geometry was reached (5 to 6 cm diameter and 2 to 5 mm up to 16 mm thickness).

Two laboratories (numbers 22 and 45) reported that the portion of the filter that had been spiked was cut off and measured directly, thus deviating from their normal measurement routine. Two other laboratories reported special arrangements: In the first arrangement (laboratory 17), filter parts not spiked were cut into small strips and arranged above and below the spiked filter part in order to get the routine counting geometry, while the second arrangement (lab 42) dealt with cutting 3 identical circular shapes (same diameter) from the rectangular air filter (one taken from the spiked area and two others from inactive parts) and pressing them together with the spiked filter in central position in order to obtain a disc shape identical to the reference source used for calibration.

Many participants (15 laboratories) placed the filters or pressed pellets in different plastic containers at prescribed positions or directly on the detector end cap (especially in case of pressed samples). Three laboratories (numbers 5, 6 and 37) measured their small filters in the initially provided plastic bag, 5 used Williams or Marinelli beakers. Larger filters were folded with the active area inwards, or pellets placed directly on the detector with the active side facing the detector or turned upside down. Target positioners were often used to press the sample against the target holder. Some of the participants provided drawings with the measurement geometries.

Only four laboratories preceded the gamma-spectrometric measurement with a chemical sample preparation of the air filters. Three laboratories (9, 11 and 44) used ashing procedures (in one of these - lab 9 - ashing being followed by acid leaching with HNO_3), and one laboratory (number 27) reported acid digestion of the filter material.

Because laboratories tried to process the spiked filters as close as possible to their routine measurement situations, some measured the same filter several times. For example, laboratory 11 measured the air filter first in 1L Marinelli geometry because in emergency situations, the authorities require daily monitoring of gamma-ray emitting radionuclides. However, due to the inhomogeneous activity

distribution of ^{137}Cs on the large air filter, this measurement geometry did not yield a reliable result. Afterwards, as in the routine procedure, the large air filter was ashed. The advantage of ashing is that the measurement geometry can be optimised while the chemical yield of the ashing process can be monitored by weighing the MgO powder which is added and processed in the same way as the sample. Another participant (lab 27) acid digested the glass fiber filter using fluoric acid and dried the residue obtaining a well defined measurement geometry.

Since the measurement system of laboratory 9 is calibrated with standard solutions, this laboratory compressed the filter, followed by ashing and leaching with nitric acid, obtaining at the end of the chemical process a leachate of acid containing the ^{137}Cs , made up to a prescribed mass and volume with deionised water before measurement.

To gain larger confidence in their own results, several laboratories performed internal checks. One of the participants (laboratory 44) used three methods of filter preparation (the sample was measured in a Marinelli beaker, folded into a 200 mL beaker, and finally ashed and the residue inserted into a 2 mL vial in a well detector), while many others measured the same prepared samples on different detectors having different relative efficiencies. Still other laboratories performed measurements of the same sample in different geometries, e.g. varying the distance to the detector, or repeated the measurements with different counting times or varying other parameters. One laboratory (number 43) checked the consistency of its gamma-spectrometric results against results obtained by total beta counting.

8. Measurements

The measurements were performed with commercially available low background gamma-spectrometry systems (Ortec, Canberra, Eurysis etc.) consisting of HPGe, BEGe or Ge(Li) detectors, p- or n-type, coaxial, well or planar types using different measurement geometries and sizes of the samples.

The detector systems were calibrated measuring certified ^{137}Cs solutions or multiple-nuclide standard solutions, or, more often, using blank filters having the same geometry as the measured routine samples spiked with calibrated/certified (multiple) radionuclide solutions. One laboratory performed efficiency calibration using a certified $^{241}\text{Am} + ^{152}\text{Eu}$ source. Some of the laboratories computed the efficiency of their detector system for ^{137}Cs using commercially available software programmes, while other laboratories gave only relative numerical values of the efficiency for ^{137}Cs .

Data evaluation was made using different software - most often Genie (Canberra), Gamma Vision (ORTEC) or a home made version - using default nuclear data of the software. Sometimes extended peak area calculations and background determination equations and functions were attached to the questionnaire, but also simple manipulations (e.g. the background spectrum was subtracted from the obtained spectrum) were used for spectrum analysis with subsequent manual activity calculations. Sample density and geometry corrections (i.e. by Monte-Carlo simulation) were taken into account by several laboratories.

The counting times of the measurements varied between 50 000 and 600 000 seconds; the average time of measurements being 2-3 days. Counting uncertainties at the level of 1s were reported between 2.0 and 32 %, when expressed in relative terms.

9. Results

9.1 Reference values

The spiked activities on the filters (reference values) were calculated using the activity concentration of the mother solution determined by primary standardisation and the gravimetrically determined dilution factors (cf. Table 3) of the spiking solutions C1, C2 and C3. The mass and activity of the dispensed solution per filter was determined once per filter by weighing the pycnometer before and after depositing the total number of drops on the corresponding filter. Table 5 shows the reference values A_0 for the deposited ^{137}Cs activity on each filter and its combined standard uncertainty u_c . The standard uncertainty of activity includes the uncertainty contributions from the primary standardisation, the dilutions and the weighings of the filter spiking aliquots.

The smallest ^{137}Cs activity spiked on an air filter was 28.6 (0.2) mBq (Filter 32), the highest value was 564 (4) mBq (Filter 38).

The reference date for reporting the activity (and likewise of the reference values) is 1 October 2003. The half-life of ^{137}Cs , used in calculating the individual reference values of Table 5 from those of the standardised solution in Table 2 and also recommended to the participants for their calculations, is $T_{1/2} = (10981 \pm 11)$ days = (30.065 ± 0.030) years [7], where the numbers following the symbol \pm are the expanded uncertainties U with a coverage factor $k = 1$.

9.2 Reported results

121 sets of results were returned from the participating laboratories. In case several measurements were performed with the same or different detectors (or any other variation to the measurement scheme) the participants usually reported multiple results. In that case, indicated in Table 6 by the number of measurements n being > 1 , the unweighted laboratory mean of the reported values was calculated by us and taken into account in the following evaluations. For a better overview, Table 6 organises the results in ascending order of the *spiked* ^{137}Cs activities, which at the same time defines the anonymous laboratory number. All reported entities are given in bold face in Table 6 (with the exception of multiple results).

The reported ^{137}Cs activities measured in this intercomparison exercise varied from 19 (4) mBq up to 650 (60) mBq (again, combined standard uncertainties u_c given in parentheses). Laboratories were asked to report uncertainties on a per filter basis (see Annex 2). In case of multiple results, the average of reported uncertainties was estimated by us as the typical single measurement uncertainty. No attempt was made to estimate the (smaller) standard uncertainty of the laboratory mean of the filter activity in such cases.

When looking closely at the reported uncertainties in Table 6, it is evident that contributions other than counting uncertainty to the combined uncertainty - such as calibration and geometry factors - are not identified and taken into account quantitatively by as many laboratories (19) as the number of those which know or at least make efforts to estimate correctly combined standard and expanded uncertainties. Eight laboratories even state expanded uncertainties $U = k \cdot u_c$, with a

Table 5: The reference values of ^{137}Cs activity and their estimated combined standard uncertainties on the air filters prepared for the intercomparison exercise (reference date 1 October 2003)

* this filter was supplied with a too low activity due to a communication problem, replaced by 17a

filter no.	activity A_0 (mBq)	$u_c(A_0) / A_0$ (%)	uncertainty $u_c(A_0)$ (mBq)
Filter_01	109.1	0.66%	0.7
Filter_02	84.2	0.66%	0.6
Filter_03	209.4	0.66%	1.4
Filter_04	339.5	0.66%	2.2
Filter_05	279.0	0.66%	1.8
Filter_06	141.8	0.66%	0.9
Filter_07	99.0	0.73%	0.7
Filter_08	129.0	0.66%	0.8
Filter_09	67.4	0.73%	0.5
Filter_10	347.2	0.66%	2.3
Filter_11	66.0	0.73%	0.5
Filter_12	143.8	0.66%	0.9
Filter_13	125.6	0.66%	0.8
Filter_14	123.4	0.66%	0.8
Filter_15	221.5	0.66%	1.5
Filter_16	75.5	0.66%	0.5
Filter_17	15.2*	0.67%	0.1
Filter_17a	111.5	0.66%	0.7
Filter_18	54.3	0.66%	0.4
Filter_19	71.0	0.66%	0.5
Filter_20	101.4	0.66%	0.7
Filter_21	70.6	0.66%	0.5
Filter_22	135.7	0.66%	0.9
Filter_23	123.8	0.66%	0.8
Filter_24	401.3	0.66%	2.6
Filter_25	125.9	0.73%	0.9
Filter_26	65.3	0.66%	0.4
Filter_27	101.1	0.66%	0.7
Filter_31	67.6	0.66%	0.4
Filter_32	28.6	0.66%	0.2
Filter_33	213.8	0.66%	1.4
Filter_34	108.2	0.66%	0.7
Filter_35	188.6	0.66%	1.2
Filter_36	228.3	0.66%	1.5
Filter_37	368.0	0.66%	2.4
Filter_38	564	0.66%	4
Filter_39	286.0	0.66%	1.9
Filter_40	56.9	0.66%	0.4
Filter_41	59.8	0.66%	0.4
Filter_42	123.3	0.66%	0.8
Filter_43	561	0.66%	4
Filter_44	58.0	0.73%	0.4
Filter_45	37.6	0.73%	0.3
Filter_46	193.0	0.66%	1.3
Filter_47	135.1	0.66%	0.9

Table 6a: Activity values and their estimated uncertainties of the air filters measured in the intercomparison exercise

lab. no.	filter no.	spiked activity A_0 (mBq)	measured activity A (mBq)	n	counting uncertainty s (mBq)	rel. counting uncertainty s/A (%)	expanded uncertainty U (mBq)	rel. expanded uncertainty U/A (%)	measured activity A ————— spiked activity A_0	standard uncertainty u_c (A/A_0)
1	Filter_32	28.6	19	1	4	21	8	42	0.67	0.21
2	Filter_32	28.6	25	1	5	20	10	40	0.88	0.20
3	Filter_32	28.6	33	1	4	12.1	8	24	1.16	0.12
4	Filter_45	37.6	140	1	20	14.3	-	-	3.72	0.14
5	Filter_18	54.3	59.8	5	6.6	11.1	13.2	22	1.10	0.11
6	Filter_40	56.9	76	2	24	32	49	65	1.34	0.32
7	Filter_44	58.0	60.3	2	8.5	14.1	17	28	1.04	0.14
8	Filter_41	59.8	53	1	6	11.3	-	-	0.89	0.11
9	Filter_26	65.3	116	1	23	20	46	40	1.78	0.20
10	Filter_11	66.0	58.8	1	6.8	11.6	8.2	13.9	0.89	0.07
11	Filter_09	67.4	64	1	1.3	2.0	3.7	5.8	0.95	0.03
12	Filter_31	67.6	39.5	1	4	10.1	8	20	0.58	0.10
13	Filter_31	67.6	49.3	1	4.5	9.1	9	19	0.73	0.09
14	Filter_31	67.6	51	1	9	18	18	35	0.75	0.18
15	Filter_21	70.6	84.4	3	22	26	22	26	1.20	0.13
16	Filter_19	71.0	82.3	4	7.7	9.4	16	19	1.16	0.10
17	Filter_16	75.5	77.6	2	13.7	18	29	37	1.03	0.18
18	Filter_02	84.2	80.6	5	9.2	11.4	29	36	0.96	0.18
19	Filter_07	99.0	99	11	6	6.1	16	16	1.00	0.08
20	Filter_27	101.1	173	1	25	14.5	33	19	1.71	0.10
21	Filter_20	101.4	74.6	3	22	29	23	30	0.74	0.15
22	Filter_34	108.2	94	4	9	9.6	18	19	0.87	0.10
23	Filter_01	109.1	95	1	5	5.3	20	21	0.87	0.11
24	Filter_17a	111.5	125	2	13	10.4	29	23	1.12	0.12

Table 6b: Activity values and their estimated uncertainties of the air filters measured in the intercomparison exercise (ctd.)

lab. no.	filter no.	spiked activity A_0 (mBq)	measured activity A (mBq)	n	counting uncertainty s (mBq)	rel. counting uncertainty s/A (%)	expanded uncertainty U (mBq)	rel. expanded uncertainty U/A (%)	measured activity A spiked activity A_0	standard uncertainty $u_c (A/A_0)$
25	Filter_42	123.3	138	7	5.4	3.9	17	12.3	1.12	0.06
26	Filter_14	123.4	118	1	9.4	8.0	17	14.4	0.96	0.07
27	Filter_23	123.8	135	1	26	20	54	40	1.09	0.20
28	Filter_13	125.6	120	1	3.2	2.7	8.2	6.8	0.96	0.03
29	Filter_25	125.9	126	1	9	7.1	18	14.3	1.00	0.07
30	Filter_08	129.0	150	1	40	27	40	27	1.16	0.13
31	Filter_47	135.1	150	1	21	14.0	42	28	1.11	0.14
32	Filter_22	135.7	180	8	20	10.8	15	8.3	1.33	0.04
33	Filter_06	141.8	157	2	8	5.1	16	10.0	1.11	0.05
34	Filter_12	143.8	130	1	4	3.1	18	13.8	0.90	0.07
35	Filter_35	188.6	228	2	34	15	67	30	1.21	0.15
36	Filter_46	193.0	229	1	9.2	4.0	14	6.0	1.19	0.03
37	Filter_03	209.4	180	1	10	5.6	50	28	0.86	0.14
38	Filter_33	213.8	210	4	20	9.5	50	24	0.98	0.12
39	Filter_15	221.5	154	1	8	5.2	20	13.0	0.70	0.07
40	Filter_36	228.3	204	1	15	7.4	30	14.7	0.89	0.07
41	Filter_05	279.0	303	2	12	4.0	30	9.9	1.09	0.05
42	Filter_39	286.0	305	2	10	3.3	20	6.6	1.07	0.03
43	Filter_04	339.5	288	3	9.1	3.2	24	8.2	0.85	0.04
44	Filter_10	347.2	346	6	13.8	4.0	45	12.9	1.00	0.06
45	Filter_37	368.0	392	1	9	2.3	18	4.6	1.07	0.02
46	Filter_24	401.3	472	1	28	5.9	34	7.2	1.18	0.04
47	Filter_43	561	602	8	41	6.8	100	17	1.07	0.08
48	Filter_38	564	650	1	50	7.7	120	18	1.15	0.09

requested coverage factor $k = 2$ (see Annex 2), which are smaller than twice the counting uncertainty alone (cf. Table 6). It is obvious that the concepts of error propagation and in particular of the Guide to the Expression of Uncertainty in Measurement (GUM) [8] are not correctly applied in more than half of the participating laboratories.

In order to evaluate the performance of the laboratories, their measurement results need to be expressed versus the individual spiked activity (IRMM reference value) on each filter. The individual spikes can be normalised to a common reference by calculating the “measured/spiked” activity ratios. The last two columns of Table 6 contain this ratio, A/A_0 , and its combined standard uncertainty u_c . It was determined according to:

$$u_c(A/A_0) = \sqrt{[u_c(A_0)/A_0]^2 + [U(A)/2 \cdot A]^2}$$

based on $U(A) = k \cdot u_c(A)$ with $k = 2$.

Due to the very small uncertainty of the reference values, $u_c(A_0)$, the uncertainty of the activity ratios measured versus spiked, $u_c(A/A_0)$, is dominated by the measurement uncertainty.

Even though it was requested to check for any residual activity on the packing material, only five laboratories reported to have measured separately the plastic bags in which the samples were provided. In three cases “no radioactive contamination” was reported, in two cases “below detection limits”. This small feedback is certainly partly due to the low activity constraints, which would require enormously long counting times. Many laboratories were instead able to measure the filters directly within the packing material.

10. Data evaluation and comparison of data

In order to compare the results, the activity ratios of $A(^{137}\text{Cs})_{\text{measured}}$ and $A_0(^{137}\text{Cs})_{\text{placed by spiking}}$ (second to last column of Table 6) are plotted in Fig. 8b together with the spiked and measured activity, displayed as histogram bars in Fig. 8a. The laboratory numbers were defined in Table 6, sorted by the amount of spiked activity on the filter. In this way, the five laboratories which received two

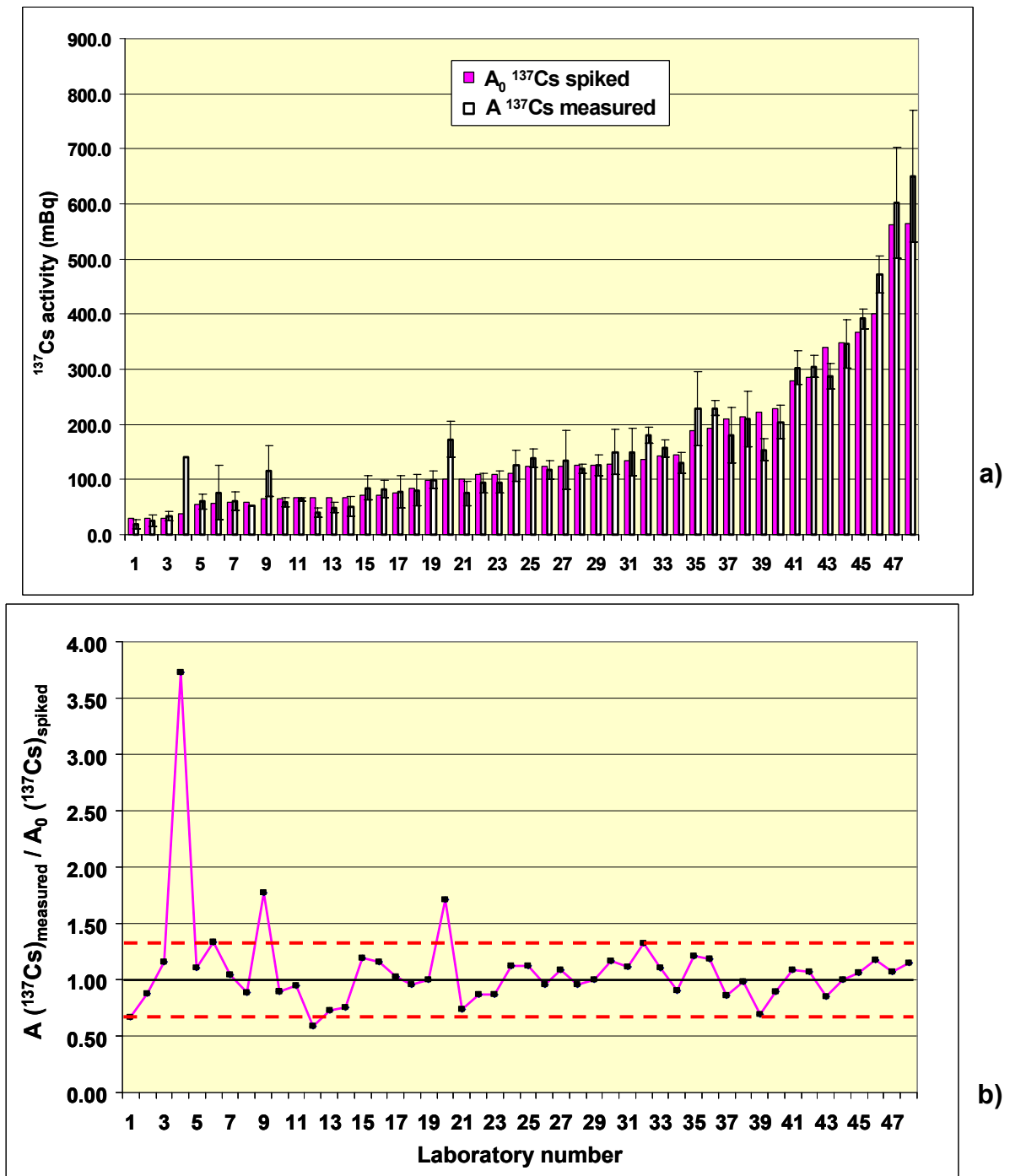


Fig. 8: a) ^{137}Cs activity in the filters, individual reference values A_0 and measurement results A ; b) Calculated ratio $A(^{137}\text{Cs})_{\text{measured}} / A_0(^{137}\text{Cs})_{\text{spiked}}$ for the 48 laboratories, IRMM reference value = 1.00

filters each are assigned two laboratory numbers in Table 6 and Fig. 8. The error bars of Fig. 8a indicate the reported expanded uncertainty of the measurement results. The uncertainty of the reference values is not visible on this scale. The solid line in Fig. 8b represents the IRMM reference value, the dashed lines $\pm 33\%$ deviation from the reference value.

For the sake of clarity, the uncertainty in the activity ratio “measured/spiked” (last column of Table 6) is neither taken into account in Fig. 8 nor in the immediate discussion that follows. We will treat this later in the detailed discussion.

As we can see the results obtained are quite homogeneous with the exception of the four labs No. 4, 9, 12 and 20 that reported results out of distribution with a deviation clearly larger than $\pm 33\%$ from the reference value ($A/A_0 \ll 0.67$ or $A/A_0 \gg 1.33$). Another four laboratories (No. 1, 6, 32 and 39) obtained results just outside or at the lines of 33% deviation.

From Fig. 8 it is apparent that the large deviations from the reference value can not be attributed to the small amounts of ^{137}Cs spikes alone. There is a group of laboratories (No. 2, 5, 7, 8, 10, 11, 17, 18 and 19) which succeeds to measure small amounts of radioactivity (< 100 mBq) within $< 13\%$ relative deviation, covered (with one exception) by the counting uncertainty alone. Neither can a specific sample preparation scheme be the reason. The filter preparation of laboratories 4 and 20 (activity ratios of 3.7 and 1.7, respectively) consisted only of folding, whereas laboratory 9 (activity ratio 1.8) employed a chemical preparation scheme that included pressing, ashing and acid digestion. Laboratory 44, however, using ashing as filter preparation as well, obtained an ideal activity ratio of 1.00. In case of laboratory 20 (Filter_27) no sample preparation but folding was applied, the spiked filter was placed 5 mm from the endcap of the detector and measured directly. The ^{137}Cs activity provided was approximately four times higher than the activities measured routinely by that laboratory, and approximately six times higher than the detection limit reported. Nevertheless, an activity ratio of 1.7 (70% deviation from the reference value) was obtained. Similarly, things went completely wrong in the case of laboratory 4 (Filter_45) which reported a measured activity after simply folding the filter and direct measurement, resulting in a deviation of 270% (or an activity ratio of 3.7) from the reference value.

An alternative way of presentation, albeit yielding no new information versus Fig. 8b, is the deviation chart. Column 6 of Table 7 contains the relative deviation from the reference activity value calculated with the formula

$$\text{rel. deviation} = \frac{A(^{137}\text{Cs})_{\text{measured}} - A_0(^{137}\text{Cs})_{\text{spiked}}}{A_0(^{137}\text{Cs})_{\text{spiked}}} * 100$$

If we represent these values, now sorted in ascending order, in a deviation chart (Fig. 9), where the dashed lines mark again $\pm 33\%$ deviation, the four outlying laboratories 4, 9, 12 and 20 are again clearly visible.

Fig. 9 gives also the strong indication that the results are normally distributed, with the exception of that of laboratory 4 and possibly those of 9, 12 and 20. Moreover, the median of those laboratory results remaining in the distribution coincides with the reference value.

Table 7a: Comparison and compatibility test (E_n numbers) of the results reported by the 48 laboratories

lab. no.	$A_0(^{137}\text{Cs})_{\text{spiked}}$ (mBq)	rel. expanded uncertainty $U_{\text{ref}}^* = k \cdot u_c(A_0)/A_0$ (%)	$A(^{137}\text{Cs})_{\text{measured}}$ (mBq)	rel. expanded uncertainty $U_{\text{lab}}^* = U/A$ (%)	relative deviation (%)	E_n	compatibility
1	28.6	1.33	19	42	-33.5	-0.80	YES
2	28.6	1.33	25	40	-12.5	-0.31	YES
3	28.6	1.33	33	24	15.5	0.64	YES
4	37.6	1.46	140	29	272.5	9.51	NO
5	54.3	1.32	59.8	22	10.2	0.46	YES
6	56.9	1.32	76	65	33.6	0.52	YES
7	58.0	1.46	60.3	28	4.0	0.14	YES
8	59.8	1.32	53	23	-11.3	-0.50	YES
9	65.3	1.32	116	40	77.6	1.96	NO
10	66.0	1.46	58.8	13.9	-10.9	-0.77	YES
11	67.4	1.46	64	5.8	-5.0	-0.84	YES
12	67.6	1.32	39.5	20	-41.6	-2.05	NO
13	67.6	1.32	49.3	19	-27.1	-1.48	NO
14	67.6	1.32	51	35	-24.6	-0.70	YES
15	70.6	1.32	84.4	26	19.5	0.75	YES
16	71.0	1.32	82.3	19	16.0	0.84	YES
17	75.5	1.32	77.6	37	2.8	0.08	YES
18	84.2	1.32	80.6	36	-4.2	-0.12	YES
19	99.0	1.46	99	16	0.0	0.00	YES
20	101.1	1.32	173	19	71.1	3.72	NO
21	101.4	1.32	74.6	30	-26.4	-0.88	YES
22	108.2	1.32	94	19	-13.1	-0.68	YES
23	109.1	1.32	95	21	-12.9	-0.61	YES
24	111.5	1.32	125	23	12.1	0.52	YES

Table 7b: Comparison and compatibility test (E_n numbers) of the results reported by the 48 laboratories (continued)

lab. no.	$A_0(^{137}\text{Cs})_{\text{spiked}}$ (mBq)	rel. expanded uncertainty $U^*_{\text{ref}}=k \cdot u_c(A_0)/A_0$ (%)	$A(^{137}\text{Cs})_{\text{measured}}$ (mBq)	rel. expanded uncertainty $U^*_{\text{lab}} = U/A$ (%)	relative deviation (%)	E_n	compatibility
25	123.3	1.32	138	12.3	11.9	0.96	YES
26	123.4	1.32	118	14.4	-4.4	-0.30	YES
27	123.8	1.32	135	40	9.1	0.23	YES
28	125.6	1.32	120	6.8	-4.5	-0.64	YES
29	125.9	1.46	126	14.3	0.1	0.01	YES
30	129.0	1.32	150	27	16.3	0.61	YES
31	135.1	1.32	150	28	11.0	0.39	YES
32	135.7	1.32	180	8.3	32.6	3.86	NO
33	141.8	1.32	157	10.0	10.7	1.06	NO
34	143.8	1.32	130	13.8	-9.6	-0.69	YES
35	188.6	1.32	228	30	20.9	0.71	YES
36	193.0	1.32	229	6.0	18.6	3.04	NO
37	209.4	1.32	180	28	-14.1	-0.51	YES
38	213.8	1.32	210	24	-1.8	-0.07	YES
39	221.5	1.32	154	13.0	-30.5	-2.33	NO
40	228.3	1.32	204	14.7	-10.7	-0.72	YES
41	279.0	1.32	303	9.9	8.6	0.86	YES
42	286.0	1.32	305	6.6	6.6	0.99	YES
43	339.5	1.32	288	8.2	-15.2	-1.84	NO
44	347.2	1.32	346	12.9	-0.4	-0.03	YES
45	368.0	1.32	392	4.6	6.5	1.36	NO
46	401.3	1.32	472	7.2	17.6	2.41	NO
47	561	1.31	602	17	7.3	0.44	YES
48	564	1.32	650	18	15.2	0.82	YES

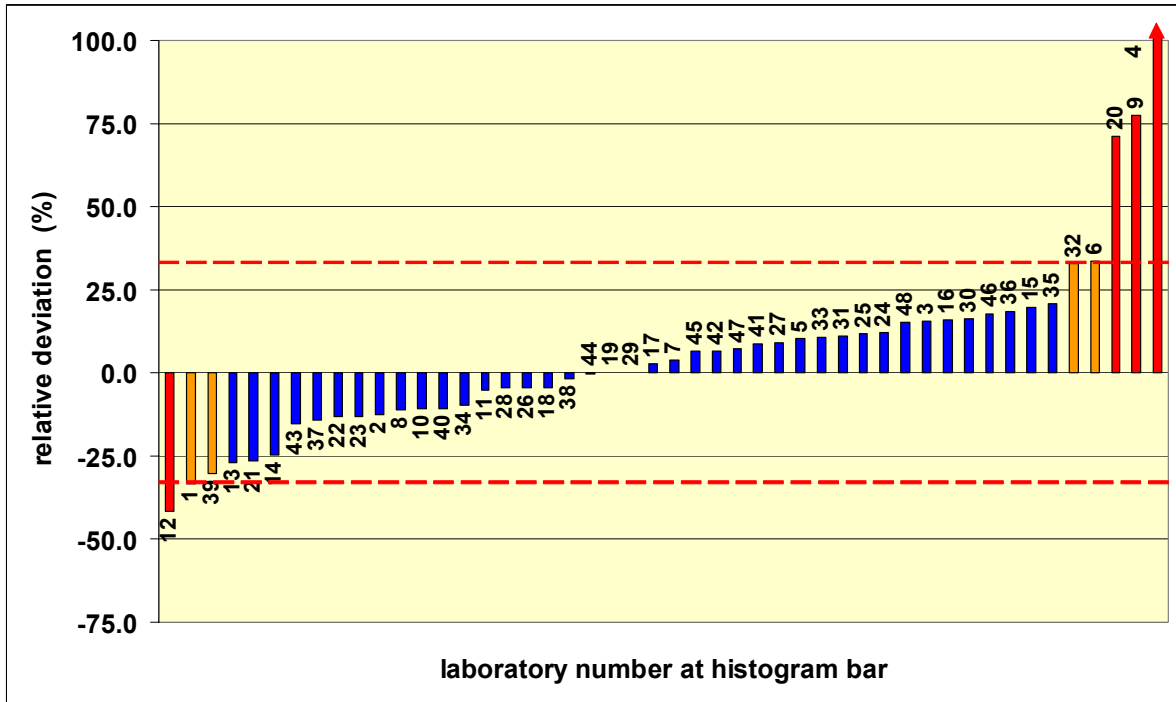


Fig. 9: Relative deviation of the 48 measurement results from the reference value, sorted in ascending order

In order to allow a more detailed analysis, several statistical tests – taking the measurement uncertainty and that of the reference values into account – have been applied. Strictly speaking, tests including measurement uncertainty must be used with caution when the uncertainty estimation is poorly understood. We have already observed, in chapter 9.2, that this holds for more than half of the participating laboratories. Nevertheless, the selected performance test using E_n numbers [11] of the activity ratios A/A_0 proves to be robust enough justifying its use in this evaluation.* It will become clear below that the conclusions from the previous discussion (which ignored uncertainty) will in general be confirmed and some additional insight will be gained.

The performance statistic “ E_n number” is calculated as [11]:

$$E_n = \frac{x - X}{\sqrt{U_{lab}^2 + U_{ref}^2}}$$

where x is the laboratory result, X the reference value, U_{lab} the expanded uncertainty of x and U_{ref} the expanded uncertainty of X . With the varying individual reference values A_0 of this measurement comparison, the activity ratio A/A_0 must be applied to x , and the reference value in the formula becomes $X = 1$. Since both values are ratios, the *relative* expanded uncertainty $U_{lab}^* = U/A$ (Table 6) must be used for U_{lab} . Similarly, U_{ref} is determined as $U_{ref}^* = k \cdot u_c(A_0)/A_0$ (from Table 5) with $k = 2$. In line with what was mentioned earlier, in chapter 9.2, for this measurement comparison the denominator in the formula above is dominated by U_{lab} .

* One should keep in mind that other performance tests usually also have constraints, e.g. a normal distribution of results, which are not always met.

When the estimation of uncertainties is consistent with the Guide to the Expression of Uncertainty in Measurement (GUM) [8], a measurement result with its uncertainty interval giving a level of confidence of 95 % (corresponding to $x \pm U_{lab}$ with an expanded uncertainty $U_{lab} = k \cdot u_c$ with a coverage factor of $k \approx 2$) will overlap with the reference value X (and its expanded uncertainty U_{ref}).

Therefore, E_n numbers are interpreted in the following way:

If $|E_n| \leq 1$, the laboratory values are compatible with the reference value;

If $|E_n| > 1$, “warning signal”, the laboratory values differ significantly from the reference value, sources of deviation should be investigated and corrected (yellow colour in Table 7);

In analogy to the interpretation of z-scores [11], a second level of critical value can be defined:

If $|E_n| > 1.5$, “action signal”, there is urgent need to investigate and find the sources of the large deviation (orange colour in Table 7).

The E_n numbers and test results are given for each laboratory in the last two columns of Table 7. Sorted in ascending order, the E_n numbers are graphically presented in Fig. 10. Whereas 36 results out of 48 (67 %) are compatible with the reference value under the conditions of this test, 12 are not. Among those 12, nine laboratories (No. 4, 9, 12, 20, 32, 36, 39, 43 and 46) report largely incompatible results ($|E_n| > 1.5$). When comparing Figs. 9 and 10 it is clear that results with large deviations from the reference value (red histogram bars in Fig. 9) are scoring bad with E_n numbers as well. That comparison, however, also shows the effect of estimating uncertainty *too small*: Some laboratories with an acceptable small deviation of $< 15\%$ are assigned a critical E_n number, because *not all*

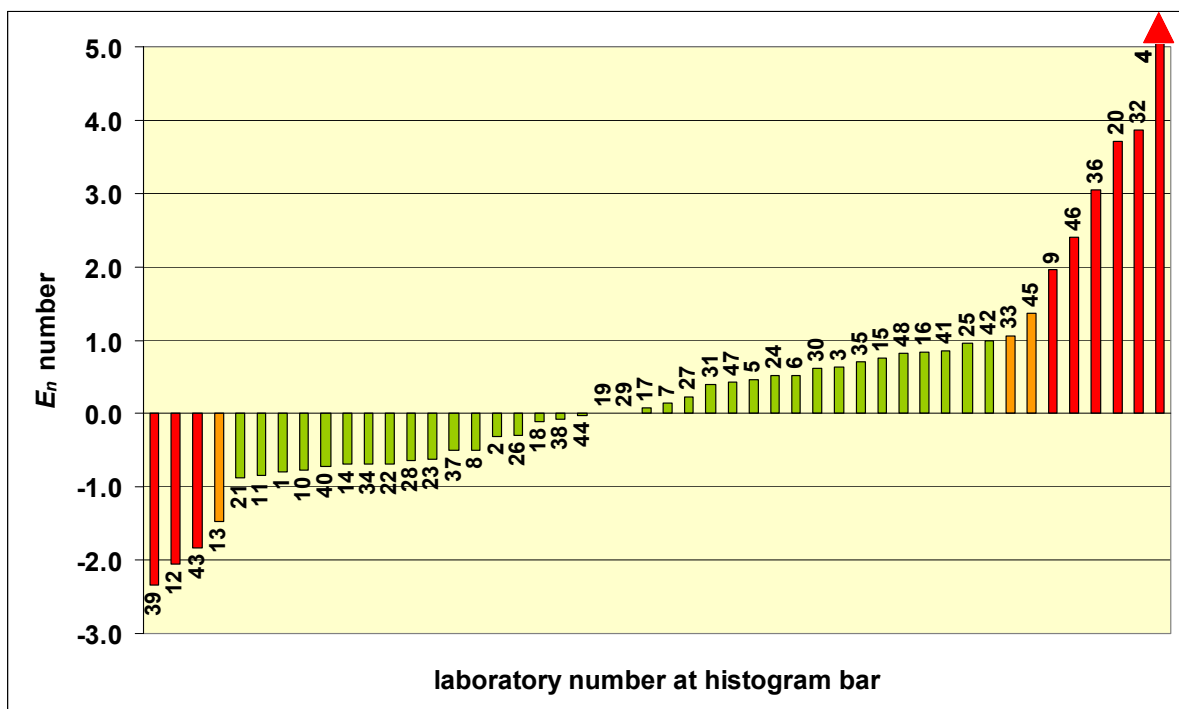


Fig. 10: E_n number of activity ratios obtained by the 48 laboratories, sorted in ascending order

contributions to uncertainty are correctly estimated (laboratories No. 33, 43 and 45). If a laboratory controls the measurement process well with only very small deviations *and* obtains a realistic estimate of uncertainty as well*, the E_n number is close to 0. Table 7 and Fig. 10 depict several laboratories of this arbitrarily chosen category of $|E_n| < 0.1$: No. 17, 19, 29, 38 and 44.

To conclude the discussion of E_n numbers and evaluations where uncertainty is taken into account, Fig. 11 reproduces the activity ratios A/A_0 (Table 6, Fig. 8b), now with the expanded uncertainty of the ratio $U(A/A_0) = k \cdot u_c(A/A_0)$ indicated as error bars in the graph (with $k = 2$). The previous discussion of E_n numbers can be intuitively visualised with this graph as well; note in particular the error bars overlapping or not overlapping, respectively, with the IRMM reference value.

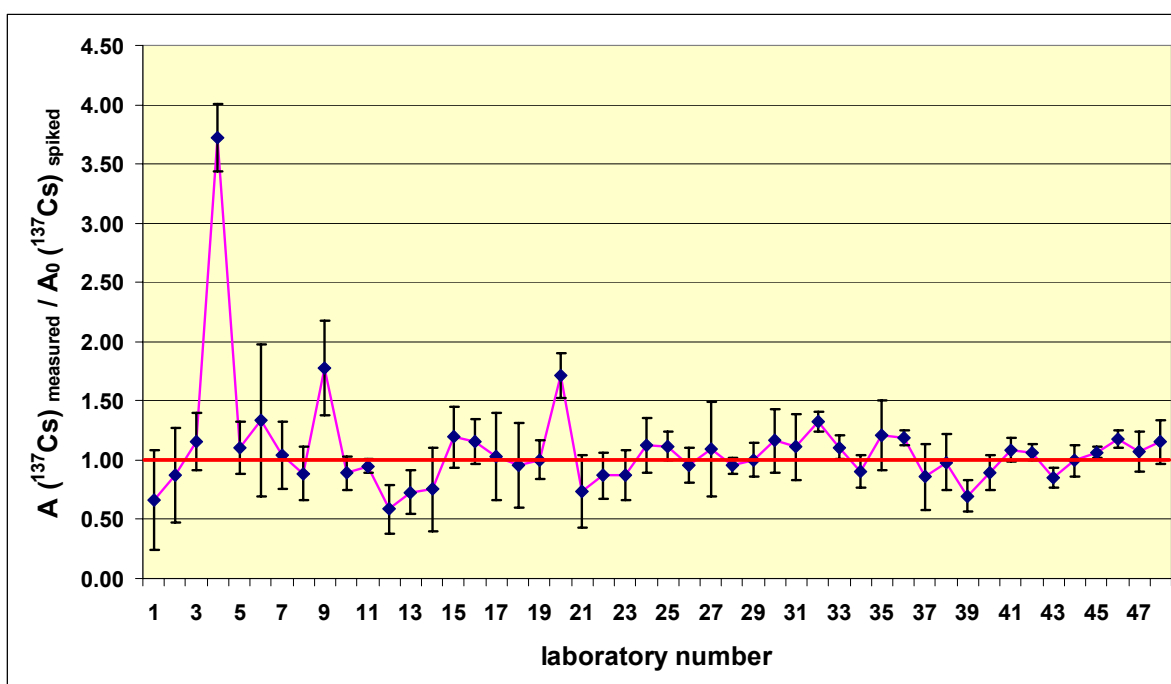


Fig. 11: Activity ratios $A(^{137}\text{Cs})_{\text{measured}} / A_0(^{137}\text{Cs})_{\text{spiked}}$ with their expanded uncertainties $U(A/A_0)$, IRMM reference value = 1.00 (red line)

An important aspect of this measurement comparison was the large difference in the spiked activity the laboratories had to measure. The variation of the ^{137}Cs activity between different filters distributed was due to differences in the values reported to be measured routinely in the monitoring programmes of the various laboratories or due to the reported differences in detection limits of the gamma-ray spectroscopy systems. 18 laboratories received air filters spiked with less than ~ 100 mBq ^{137}Cs , 16 laboratories got ^{137}Cs activities between about 100 and 150 mBq, while 6 laboratories had to measure activities of around 200 mBq, and another 8 laboratories received activities higher than 250 mBq (cf. also Fig. 12).

* Unfortunately, “guessing” a too large uncertainty would reduce the $|E_n|$ number without justification.

Whereas it was evident from the outset that counting uncertainties would dominate the combined measurement uncertainty at the low end of filter activities supplied, it was pointed out in the discussions above that they can not explain the large deviations from the reference value obtained by several laboratories in this comparison.

Since the inhomogeneous distribution of the spiked activity on the filters was often claimed to be another source of large unknown deviations, we made some efforts to estimate its possible influence on measurement results, see the following separate chapter 11.

Some further statistical evaluations serve to characterise this intercomparison in a general way, which should not replace, however, the performance evaluation of individual laboratories given above. Table 8 summarises the basic characteristics of the distribution of measurement results.

Table 8: Statistical analysis of the laboratory results expressed as the activity ratio of $A(^{137}\text{Cs})_{\text{measured}} / A_0(^{137}\text{Cs})_{\text{spiked}}$

Number of results	45*	48
Maximum value	1.34	3.72
Minimum value	0.58	0.58
Mean value	1.00	1.09
Standard deviation of mean	0.17	0.45

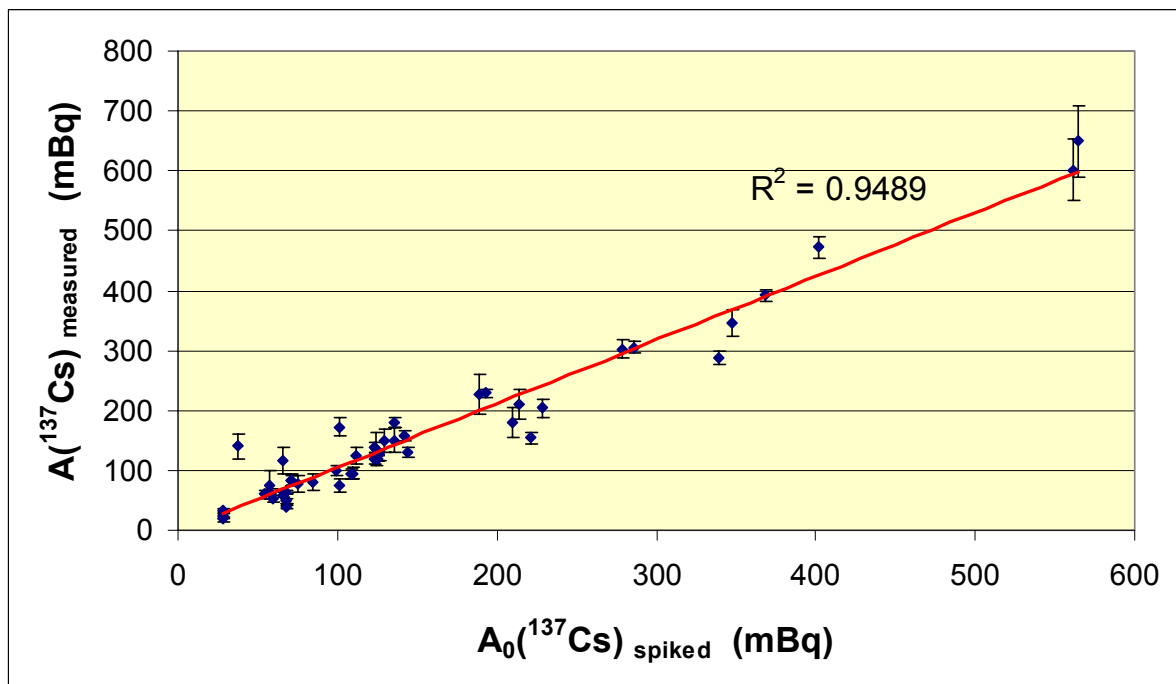


Fig. 12: Scatter chart of measurement results given by the 48 laboratories. Error bars indicate combined standard uncertainties u_c , the regression line is given in red

* excluding the three outlying results No. 4, 9 and 20 according to the Nalimov test

The scatter chart in Fig. 12 shows the deviations of the pairs of data from the linear trend, reflecting the mis-matches between the measured data and reference values. The regression coefficient of 0.9489 indicates that the distribution of data may be approximated by a Gaussian.

The Nalimov test was used to identify purely statistical outliers among the reported results. The result of laboratory No. 4 was flagged in first instance at a level of significance $\alpha = 0.01$, and the results of No. 9 and 20 in the second round of testing at the same level of significance after excluding the first outlier. No further outliers were identified on statistical grounds (at $\alpha = 0.01$) with the Nalimov nor other tests.

After exclusion of the three outlying results identified by the test, the skewness and kurtosis tests indicate indeed the normality of the remaining data at a level of significance of $\alpha = 0.01$. The second column of Table 8 reflects the distribution of results after exclusion of these outliers.

11. Influence of an inhomogeneous spike distribution on the measured activity

The discrete distribution of the spiked activity in the prepared filters, as opposed to the presumably uniform activity distribution on real filters, was studied as a possible cause for the observed deviations in cases where only folding of filters was applied. In order to study this effect a number of Monte Carlo simulations were conducted using EGS4 [12]. Figure 13 depicts the different cases that were modelled. It was assumed that a filter was folded to a height of 11.9 mm and a diameter of 50.0 mm and placed in a container. In order to keep the filter in a fixed position filling material occupied the space inside the container between the filter and the bottom of the lid. The container was placed directly on the endcap of a 20% relative efficiency coaxial HPGe detector with a 1 mm thick aluminium window. The distance from the top of the Ge *crystal* to the bottom of the sample (folded air filter) was 12 mm.

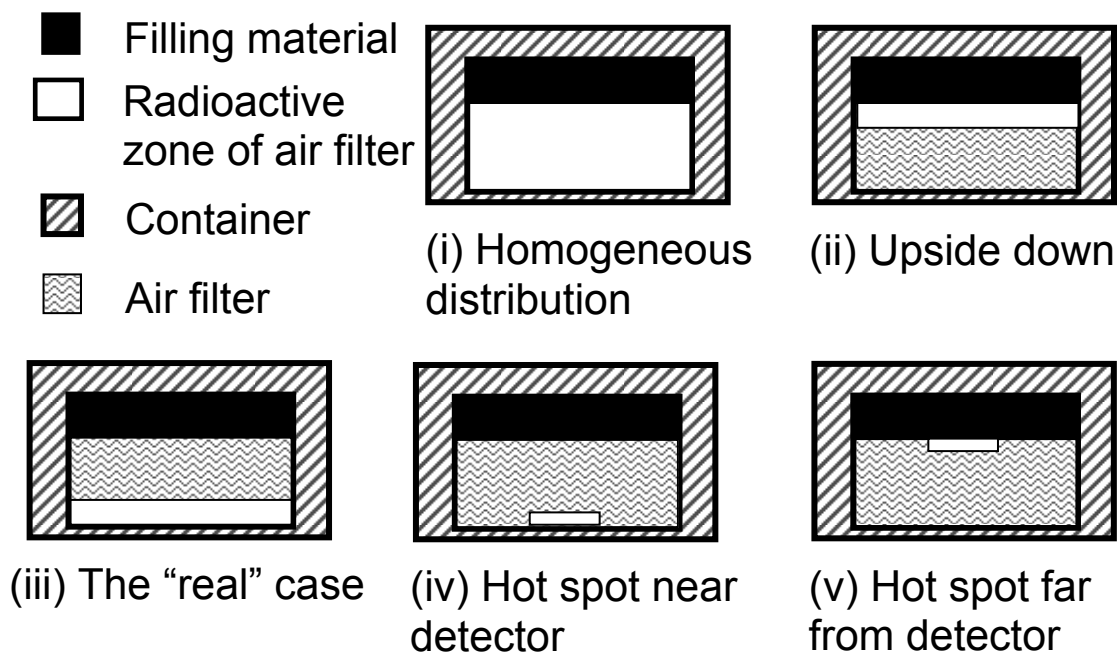


Fig. 13: Different geometries for the activity distribution in the air filter that were modelled using Monte Carlo simulation

Dimensions, matrix composition and density of the folded filter and the container as well as the dimensions of the detector were used as model inputs to the Monte Carlo code. The detector dimensions were determined from a radiograph made at two angles. The dead layer thicknesses (front, side and bottom) were determined experimentally by measuring a number of calibration sources and adjusting the dead layer thicknesses until a good match was obtained. The calibration sources for determining the dead layers for the computer model were ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{51}Cr , ^{137}Cs , ^{54}Mn , ^{113}Sn , ^{85}Sr and ^{65}Zn , traceable to the SIR (International Reference System for Radionuclides) via primary standards at BNM-LNHB*. The

* Bureau National de Métrologie – Laboratoire National Henri Becquerel, Saclay, France

value for the window thickness was taken from manufacturer data. The simulations assumed that the gamma-ray emissions were isotropic and uncorrelated.

The results of the Monte Carlo simulations for the different geometries are given in Table 9. The worst case scenario, in which all activity is assumed to be concentrated in a centred spot of 4.0 mm diameter (hot spot) at the bottom or top of the folded filter (cases iv and v of Fig. 13), resulted in an overestimation of activity by 59 %. "Turning the filter upside down" in its container so that the active spot is located 11.9 mm further away from the end cap, resulted in an underestimation by 12 % (cf. Table 4). In conclusion, keeping in mind the spike patterns applied and their visibility on the filters which allows taking their distribution into account and avoiding "worst cases", the observed measured-to-spiked activity ratios cannot be explained by the discrete distribution of spikes.

Table 9: Full energy peak (FEP) relative efficiency at 662 keV for the different activity distributions depicted in Fig. 13, combined standard uncertainty u_c given in brackets in units of the least significant digit(s) of the corresponding result

¹ Corrected measurement result

² The term "correct distribution" refers to the activity distribution in the folded filter returned from laboratory No. 4

Case	FEP efficiency at 662 keV	Relative difference compared to case (i) (%)	Activity distribution	Measured activity ¹ (mBq)
(i)	0.02462 (2)	0	Homogeneous throughout the whole sample volume	---
(ii)	0.01985 (5)	-19	Correct distribution ² with sample upside down	40.2 (4.5)
(iii)	0.02954 (9)	20	Correct distribution ² with sample at close distance	39.3 (4.0)
(iv)	0.03923 (5)	59	A hot spot near the detector	---
(v)	0.02160 (5)	-12	A hot spot far away from the detector	---

The filter measured by laboratory No. 4 resulting in an activity ratio of 3.72 was returned to IRMM for additional measurements. It was folded (similarly as by laboratory No. 4) into a cylinder with the dimensions given above resulting in the activity being located, in the top or bottom, within 27 % (3.2 mm) of the cylinder height (cases (ii) and (iii) above). The filter was measured on a Ge detector in both orientations (active side up or down); and the results were corrected for geometry effects with the data given in column 3 of Table 9. The corrected measurement results given in the last column (rendering a weighted mean value of 39.7 mBq with a combined standard uncertainty $u_c = 3.2$ mBq) demonstrate that the activity measured at IRMM is within 6 % of the reference value of 37.6 (0.3) mBq, yielding an activity ratio of 1.06 instead of 3.72 obtained by laboratory No. 4.

12. Conclusions

The measurement comparison “ ^{137}Cs in air filters” allowed obtaining a realistic estimate of the accuracy of radioactivity measured on air filters during routine monitoring in EU Member States.

The answers given to the questionnaire revealed a diversity of sampling devices and sampling procedures, leaving much room for variation in the measurement geometry and sample preparation of air filters. If just one type of aerosol filter with the same activity would have been sent to each laboratory, a comparison of results would not have reflected the routine measurement process in the various laboratories. All in all, this measurement process is characterised by large differences in sampling period, volume of sampled air and sample preparation, and differences by three orders of magnitude in the routinely measured activity of ^{137}Cs . Thus, air filters spiked with ^{137}Cs activity close to the various levels routinely measured have been sent to the participants.

With 41 out of 48 reported measurement results lying within $\pm 33\%$ of the IRMM reference value, this comparison renders a rather fair result. However, in addition to five results from the group with larger relative deviations ($> 33\%$), another seven results (12 in total) do not fulfil the criteria of the compatibility test based on E_n numbers. Overall, these results point at some problems of radioactivity measurement in air filters which definitely need improvement. Several laboratories need to investigate possible sources of error in their determination process, which – as was shown in this report – can not merely be explained by an inhomogeneity of the spike distribution, large counting uncertainty due to low activity or insufficient uncertainty estimation.

One difficulty in interpreting the results of the present intercomparison consists in the low ^{137}Cs activities distributed to some of the participants in order to maintain realistic measurement conditions. Many of the laboratories had to measure activities close to their detection limit; therefore the uncertainties associated with the measurement results differ considerably between laboratories.

The largest deviations from the reference value, however, can not be attributed to the small amounts of ^{137}Cs spikes or to a specific sample preparation scheme alone. This is shown, on one hand, by a comparison of the results of laboratories 4, 9 and 20, using a completely different preparation scheme yet obtaining an activity ratio > 1.7 , and laboratories 9 and 44, on the other hand, where laboratory 44 obtains a perfect activity ratio of 1.00 whereas laboratory 9 gets the ratio 1.78 with an almost identical filter preparation scheme.

Apart from the expected variation in measurement uncertainty, several of the reported uncertainty estimates are not acceptable at all. The concept of estimating combined uncertainty of the whole measurement process according to the "GUM approach" [8] is not being applied yet in many of the participating laboratories. Some laboratories, however, are able to provide realistic uncertainty estimates consistent with the reported results.

The large deviations of several results from the reference value demonstrate the continuous need of such comparisons. Ensuring traceability of the IRMM reference values (spiked activity on filters) to the SIR and the possibility to carry out control

measurements have proved beneficial in giving credibility to this comparison scheme.

13. Acknowledgements

This work was possible only with the active participation of 43 laboratories in 25 countries (listed in chapter 15 below), and the support of A. Janssens from the Directorate General for Energy and Transport of the European Commission.

14. References

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16. Annexes

Annex 1: Questionnaire sent to participants at start of project



EUROPEAN COMMISSION
DIRECTORATE GENERAL JRC
JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
IRMM

International Comparison Scheme for Radioactivity Environmental Monitoring:

¹³⁷Cs on air filters

Participants to the intercomparison exercise are requested to return the duly completed questionnaire, together with two new (= unused) air filters. **Both questionnaire and air filters must reach IRMM at the latest by 15 September 2003. They should be sent to the attention of:**

Mr. Uwe Wätjen
European Commission
DG JRC
Institute for Reference Materials and Measurements
Retieseweg
B-2440 Geel
Belgium

In case you would require additional information, please contact Mr. Wätjen at:

Tel: +32 14 571 882
Fax: +32 14 584 273
Email: uwe.waetjen@irmm.jrc.be

Questionnaire for participants

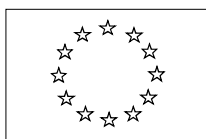
Sampling:

Supplier and type of air sampler	
Type of size separation if any	
Cut-off size (aerodynamic diameter - μm)	
Volume flow rate (m^3/hour)	
Type of flow control if any	
Type of flow measurement	
Calibration of flow measurement	
Total air volume sampled (m^3)	
Sampling frequency (e.g. once a week)	
Sampling period (hours)	
Supplier and type of filter material	
Shape and size of filter	
Blank weight (areal density - mg/cm^2)	
Typical aerosol deposit collected (mg/cm^2)	
Transport of filter between sampling station and measurement laboratory	
Further comments on sampler, filter, sampling etc.	

γ -spectrometric measurement:

Size of sample measured	
Description of sample preparation	
Geometry of measurement (possibly with drawing)	
Type of detector	
Efficiency calibration of detector system for ^{137}Cs in air filters	
Electronics and data acquisition	
Data evaluation (software, peak area determination, nuclear data and equations used)	
Typical ^{137}Cs activity collected (Bq/cm^2 or Bq/filter or Bq/m^3)	
Further comments on γ -spectrometry etc.	

Annex 2: Report form for measurement results



EUROPEAN COMMISSION
DIRECTORATE GENERAL JRC
JOINT RESEARCH CENTRE
Institute for Reference Materials and Measurements
IRMM

Geel, 22 December 2003

International Comparison Scheme for Radioactivity Environmental Monitoring:

^{137}Cs on air filters

Enclosed, please find the long-awaited and long-promised report form for the intercomparison exercise. Please, return the filled-in form **by email and a signed printout by regular mail** to the address given below. **Deadline** for reporting is **31 January 2004**.

The **reference date** for reporting the activity is **1 October 2003**, the recommended half-life of ^{137}Cs is $T_{1/2} = 10981$ (11) days = 30.065 (30) years (D. MacMahon, 2003).

In case several measurements were performed with the same or different detectors (or any other variation to the measurement scheme requires mentioning), please add the necessary columns (or rows) to the report form or use multiples of the form.

With kind regards,
wishing you a Merry Christmas period and a happy and successful New Year,

Uwe Wätjen

Dr. Uwe Wätjen
European Commission
DG JRC
Institute for Reference Materials and Measurements
Retieseweg
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In case you would require additional information, please contact me at:

Tel: +32 14 571 882
Fax: +32 14 584 273
Email: uwe.waetjen@cec.eu.int

Report form ^{137}Cs on air filter

Institution (brief name, place, country): _____

Air filter:

Supplier and type of filter material	
Shape and size of filter	
Description of sample preparation	

γ -spectrometric measurement:

Size of sample measured	
Geometry of measurement (with drawing if useful)	
Supplier and type of detector, nominal relative efficiency	
Efficiency calibration of detector system for ^{137}Cs in air filters	
Data evaluation (method of peak area and background determination, nuclear data and equations used)	
Time of measurement (s)	
^{137}Cs activity measured (Bq/filter) (reference date: 1 October 2003)	
Counting uncertainty (Bq/filter)	
Expanded uncertainty U (Bq/filter, k=2)	
Difficulties observed a.o. comments	

Responsible:

Date:

Signature: _____

Annex 3: Standardisation of the ^{137}Cs solution used for spiking the air filters by Liquid Scintillation Counting (LSC)

Radioactivity measurements of a solution of ^{137}Cs (July 2003)

Name(s) of the person(s) who carried out the measurements:

T. ALTZITZOGLU

Date: August 29, 2003

Remarks: The standardisation was carried out in the frame of the International Comparison Scheme for Radioactivity Environmental Monitoring (ICS-REM).

A. Preliminary measurements

A.1. Adsorption tests

Please take into account the adsorption tests in the evaluation of the final results.

A.1.1. Adsorption tests

Activity remaining in the "empty" original ampoule ----- (on ref. date)

Date of this test -----.

Please explain the measuring procedure used:

No adsorption test was necessary, since the solution was not stored before.

A.2. Impurity checks:

Method of measurement:

HPGe gamma-ray spectrometry

Nuclide:

^{134}Cs

Impurity to ^{137}Cs ratio 0.013%
and its uncertainty 0.001%
at reference date (2003-06-28,
0 h UTC).

The impurity was not taken further into account, considered negligible. It was only included in the uncertainty budget.

B. Source preparation

B.1. Methods used for source preparation :

Possible remarks about drying, precipitation, foils used (gold-coated or not, number, etc.), type of balance used:

All sources were prepared gravimetrically using a Mettler AX26 mass comparator, calibrated using weights traceable to the IRMM kilogram. The radioactive solution was dispensed by means of a pycnometer into the LSC vials (20 mL Packard High Performance (low-potassium) glass vials) containing the LS cocktail. Sources were prepared in 15 mL Ultima Gold + 1 mL H₂O or 15 mL Insta-Gel Plus or 15 mL Ultima Gold cocktail.

12 samples were prepared in 15 mL Ultima Gold + 1 mL H₂O

4 samples were prepared in 15 mL Insta-Gel Plus

4 samples were prepared in 15 mL Ultima Gold

The amount of radioactive solution in each sample ranged from 10 to 52 mg.

All samples were measured 9 times (for 10 minutes each) using the Packard 3100 TR/AB LSC and 3 times (for 10 minutes each) using the Quantulus 1220 LSC over a period of 2 months.

B.2. Solutions, sources

B.2.1. For photon counting and beta counting (if relevant)

Diluent:

<i>dilution number</i>	1	2	3
- <i>dilution factor</i>	-----	-----	-----
- <i>possible remarks</i>	-----	-----	-----
- <i>number of sources prepared</i>	-----	-----	-----
- <i>disposed mass of solution (approx.)</i>	-----	-----	-----

B.2.2. For liquid-scintillation counting

<i>Diluent</i>	None
<i>Dilution factor</i>	1
<i>Scintillator used to prepare the sources</i>	Ultima Gold or Insta-Gel Plus
<i>Volume of scintillator used</i>	15 cm ³
<i>Chemicals used to stabilize the solution</i>	H ₂ O (in some of the sources)
<i>Substances used as quenching agent</i>	CCl ₄
<i>Type of vials used</i>	Packard High Performance Glass Vials (low potassium)

C. Procedures used for the activity measurements

C.1. Method of measurement used

(e.g. CIEMAT-NIST method*, Triple-to-double coincidence ratio method**, or other):

Liquid Scintillation Counting, using the CIEMAT-NIST method.

(For the efficiency calculations, the computer programme CN2001A was used.)

C.2. ¹³⁷Cs nuclear data

(if average values have been used, please explicit the way they were averaged. Indicate also the origin of the data you have used):

$$T_{1/2} = (1.102 \times 10^4 \text{ d}; u = 0.006 \times 10^4 \text{ d})^{***}$$

C.2.1. Beta emission Data as in CN2001A

Emission probability 94.6 %

E_{max} 1173.2 keV

Shape factor allowed

C.2.2. Atomic data

ω_K, ω_L 0.889, 0.1

Relative probability KL, KX 0.812, 0.188

Energy KL, KX 29.7, 33.8 keV

Energy LX 4.3 keV

- Auger electrons

Relative probability KLL, KLX, KXY 0.685, 0.287, 0.041

Energy KLL, KLX, KXY 24.5, 28.6, 32.8 keV

Energy LXY 3.27 keV

* Coursey B. M., Gibson J. A. B., Heitzman M. W. and Leak J. C. Standardisation of technetium-99 by liquid-scintillation counting. *Int. J. Appl. Radiat. Isot.* **35**, 1984, pp.1103-1112

** Pochwalski K., Broda R. and Radoszewski T. Standardisation of pure beta emitters by liquid-scintillation counting. *Int. J. Appl. Radiat. Isot.* **39**, 1988, pp.165-172

*** IAEA-TECDOC-619, X-ray and gamma-ray standards for detector calibration, Evaluation by K. Debertin and M.J. Woods (1991) p. 100.

D.1. Liquid-scintillation equipment

D.1.1. CIEMAT/NIST method

D.1.1.1. Characterisation of the liquid-scintillation counter (LSC)

Type of the counter A	Packard TRI-CARB 3100 TR/AB
Age	May 1992 – upgraded Feb. 1999
Quench parameter	tSIE
Nuclide used as external standard	^{133}Ba
Efficiency obtained with an unquenched standard of ^3H	65.0%
Background (unquenched standard in toluene scintillator, 0 to 2000 or more keV)	0.6 s^{-1}
Options used (e.g. low-level counting)	None

Type of the counter B	Wallac Quantulus 1220
Age	March 2001
Quench parameter	SQP
Nuclide used as external standard	^{152}Eu
Efficiency obtained with an unquenched standard of ^3H	70.4%
Background (unquenched standard in toluene scintillator, 0 to 2000 or more keV)	0.18 s^{-1}
Options used (e.g. low-level counting)	None

D.1.1.2. Characterization of the tracer (e. g. ^3H)

Standard used and its origin	IRMM ^3H standard*
Uncertainty on the standard	0.6%
Date of preparation of the tracer samples	2003-07-03
Chemical composition of the tracer samples	Tritiated water

E.1. Liquid-scintillation parameters

Numerical codes used	CN2001A (E. Günther, PTB, 31-01-2001)
kB value:	0.0075 unit: $\text{cm}\cdot\text{MeV}^{-1}$
Formula used to calculate the ionization quenching correction factor Q(E):	CIEMAT program KB
Are M, N, ... captures taken into account?	No
Are M, N, ... x-ray and Auger electrons taken into account?	No
Model used to evaluate the interaction probability of the photons with the scintillator:	(Monte Carlo Photon Abs)
Values used for cross section of interaction	XCOM

E.1.1. *Calculated data for the liquid-scintillation method*

(for Packard Ultima Gold LSC cocktail 15 mL + 1 mL H₂O)

Efficiency for ¹³⁷ Cs beta emission	94.4% (for 52.27% eff. of ³ H)
Efficiency for ¹³⁷ Cs gamma-ray emission	1.3% (for 52.27% eff. of ³ H)
Total efficiency ¹³⁷ Cs	95.8% (for 52.27% eff. of ³ H)

E.1.2. *Corrections applied*

Data are corrected for:

- decay
- decay during measurement
- background
- dead time (automatically by the counter)

E.2. *Uncertainty components, in % of the activity concentration, due to**

		<i>Remarks</i>
<i>counting statistics</i>	0.1	for individual measurements
<i>weighing</i>	0.2	-----
<i>dead time</i>	0.1	-----
<i>background</i>	0.01	-----
<i>pile-up</i>	-----	-----
<i>resolving time</i>	-----	-----
<i>Gandy effect</i>	-----	-----
<i>counting time</i>	0.05	-----
<i>adsorption</i>		-----
<i>impurities</i>	0.013	-----
<i>tracer</i>	-----	-----
<i>input parameters and statistical model</i>	-----	-----
<i>quenching</i>	-----	-----
<i>interpolation from calibration curve</i>	0.02	-----
<i>decay-scheme parameters</i>	0.63	-----

* The uncertainty components are to be considered as approximations of the corresponding standard deviations (see also *Metrologia*, 1981, **17**, 73 and *Guide to expression of uncertainty in measurement*, ISO, corrected and reprinted 1995).

half life ($T_{1/2} = 1.102 \times 10^4$ d; $u = 0.006 \times 10^4$ d)	0.0006	-----
self absorption	-----	-----
extrapolation of efficiency curve	-----	-----
wall effect	0.05	-----
instrument dependence	0.1	-----
kB	0.03	-----
sample stability	0.1	-----
combined uncertainty	0.7	-----

(as quadratic sum of all uncertainty components)

F. Combination of individual results

(obtained from the individual dilutions, source preparation, etc.)

How have the individual results been used for arriving at the final result (statistical weights)?

All data are first corrected for background, decay and decay during measurement. Then, the weighted mean for each sample is calculated. Finally, the weighted mean of all samples in a given cocktail is calculated.

G. Final result

The radioactivity concentration of the ^{137}Cs solution on the reference date (2003-07-01, 0 h UTC) is

270 kBq g⁻¹,

and the combined uncertainty is

2 kBq g⁻¹, 0.7 %

Remarks

- The result given above represents that obtained from the sources in 15 mL Ultima Gold + 1 mL H₂O.
- The samples in Insta-Gel Plus gave as result (and combined uncertainty) 266 (2) kBq g⁻¹.
- Those in plain Ultima Gold 269 (2) kBq g⁻¹; all within the uncertainty limits.
- The result obtained using Insta-Gel Plus as the LS cocktail is lower than the others probably because of aged cocktail; therefore it was discarded.

Annex 4: Standardisation of the ^{137}Cs solution by $4\pi\beta\text{-}\gamma$ coincidence counting using ^{134}Cs as an efficiency tracer

Excerpts from:



EUROPEAN COMMISSION

JOINT RESEARCH CENTRE

Institute for Reference Materials and Measurements

Geel, Belgium

Radioactivity measurements of a solution of ^{137}Cs by $4\pi\beta\text{-}\gamma$ coincidence counting using ^{134}Cs as an efficiency tracer.

John Keightley

Category 20 Fellow

Contract Duration : 3 years

IRMM Internal Report
GE/R/RN/05/2003/09/02

JRC Reference Laboratory for Radionuclide Metrology

Radionuclide Metrology Unit



1. Introduction

The standardisation of ^{137}Cs was carried out in the framework of the International Comparison Scheme for Radioactivity Environmental Monitoring (ICS-REM).

Accurately weighed aliquots of a diluted solution of the standardised ^{137}Cs will be spiked onto blank aerosol filters, supplied by the laboratories participating in this intercomparison, in order to allow them to benchmark their radioactivity measurements of ^{137}Cs collected from air versus reference filters traceable to the SI.

Due to the existence of a delayed isomeric state (half-life : 2.552 minutes) of the ^{137}Ba daughter of ^{137}Cs , giving rise to a 661.6 keV gamma-ray transition, ^{137}Cs may not readily be standardised by the $4\pi\beta\text{-}\gamma$ coincidence counting technique. It was for this reason that a standardisation of ^{134}Cs was performed to facilitate the standardisation of ^{137}Cs by the 'efficiency tracing' technique.

The standardised solutions of both ^{134}Cs and ^{137}Cs will be submitted to le Système International de Référence (SIR) of the Bureau International des Poids et Mesures, (BIPM).

Throughout this report, quoted uncertainty components may be considered as approximations of the corresponding standard deviations, and are given in parentheses following a given value, referring to the last digits :

i.e. :

270.0 (17) means 270.0 ± 1.7

and

270 (2) means 270 ± 2

.....

4. Procedures used for the activity measurements

An approach similar to that used by the CBNM (Reher *et al*, 1982) for the 1982 international comparison of a solution of ^{137}Cs (Rytz, 1985) was adopted, namely using ^{134}Cs as an efficiency tracer in order to facilitate the standardisation of ^{137}Cs . A list of further references on this technique may be found in the BIPM Report (Rytz, 1983).

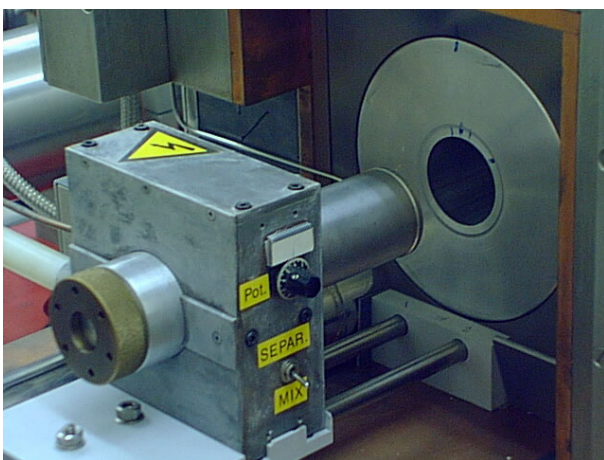


Figure 4 : $4\pi\beta(\text{PPC})\text{-}\gamma(\text{NaI}(\text{TI}))$ coincidence counting detector set-up.

In the present work, the main differences to the technique used by the CBNM in 1982, is that the standardisations were performed using IRMM's $4\pi\beta(\text{PPC})-\gamma(\text{NaI}(\text{TI}))$ coincidence counting facility, where the PPC is inserted into the well of the NaI(TI) detector (Figure 4). Beta efficiency variation performed by repeated adjustment of the lower level threshold in the beta channel.

The general technique used for the standardisation of ^{137}Cs using ^{134}Cs as an efficiency tracer may be briefly summarized as follows:

A standard of the solution of the beta–gamma emitter ^{134}Cs was prepared by $4\pi\beta(\text{PPC})-\gamma(\text{NaI}(\text{TI}))$ coincidence counting. The efficiency of the PPC detector (or 'beta efficiency', ε_β) of the sources (determined as N_c/N_γ where N_c is the corrected count rate of registered coincidences per unit mass of the source, and N_γ is the corrected count rate of registered gamma rays in the NaI(TI) detector per unit mass) was varied by adjustment of the 4π -beta counting threshold. (*Corrections applied include those for background, decay, dead times and 'accidental coincidences' inherent with the use of coincidence resolving times*). (ICRU, 1994), (Smith, 1987).

Plots were made of the corrected beta count rates against the 'beta inefficiency' (1 minus the beta efficiency), and weighted least squares fitting performed, from which extrapolations were performed to unit beta efficiency. The assumption is made that the efficiencies of the various beta branches are functionally related and approach the value one simultaneously. The weighted mean of the extrapolated beta count rates yields the activity concentration ($A_{(134)}$) of the solution.

The ^{137}Cs solution may not be standardised directly by coincidence methods due to the highly delayed nature of the 661.6 keV gamma transitions (Figure 5), stemming from the long lived isomeric state of the ^{137}Ba daughter (2.552 minutes). This facilitates the need for an efficiency tracer nuclide to be used.

The standardised solution of ^{134}Cs was quantitatively mixed with the ^{137}Cs solution (as described in Section 3). These 'mixed' sources may then be considered as 'pseudo single-nuclide' sources, and were measured in the $4\pi\beta(\text{PPC})-\gamma(\text{NaI}(\text{TI}))$ coincidence counting equipment, where the apparent beta efficiency, (N_c/N_γ), was determined as described above, and similar extrapolations performed to unit beta efficiency, yielding the apparent total activity of the source, $A_{(tot)}$.

^{134}Cs is an ideal efficiency tracer for ^{137}Cs , as both nuclides may be prepared in the same chemical format, enabling uniform mixing of the solutions. Furthermore, the principal beta transition from ^{134}Cs has a maximum energy of 657.8 keV (Figure 6), which is similar to the 513.9 keV principal beta transition of ^{137}Cs (Figure 5). The PPC beta efficiency for ^{137}Cs ($\varepsilon_{\beta(137)}$) = N_c/N_γ is readily measured with a mixture source, provided the 661.6 keV gamma transitions following the decay of ^{137}Cs are **not** counted in the gamma channel. This is accomplished by imposing a suitable threshold, (Section 6). The beta efficiency of the PPC for ^{134}Cs does not need to exactly mimic the beta efficiency for ^{137}Cs . The requirement is that a polynomial relationship exists between $\varepsilon_{\beta(137)}$ and the beta inefficiency for the ^{134}Cs tracer ($1 - \varepsilon_{\beta(134)}$) (Rytz, 1983).

i.e. :

$$\varepsilon_{\beta(137)} = f(1 - \varepsilon_{\beta(134)}) = f\left(1 - \frac{N_c}{N_\gamma}\right) = 1 + c_1\left(1 - \frac{N_c}{N_\gamma}\right) + c_2\left(1 - \frac{N_c}{N_\gamma}\right)^2 + K$$

The contribution of the ^{134}Cs tracer activity to the total observed beta count-rate from a mixed source is :

$$N_{\beta(134)} = \frac{m_{(134)} \cdot A_{(134)}}{D_{(134)}} \cdot g(1 - \varepsilon_{\beta(134)})$$

with :

$$g(1 - \varepsilon_{\beta(134)}) = 1 + d_1 \left(1 - \frac{N_c}{N_\gamma}\right) + d_2 \left(1 - \frac{N_c}{N_\gamma}\right)^2 + K$$

where :

$m_{(134)}$ is the mass of ^{134}Cs solution,

$D_{(134)}$ is the decay correction factor, and

d_1, d_2, \dots are obtained from measurements on the pure ^{134}Cs solution, which were used to determine the tracer activity concentration $A_{(134)}$.

Defining $N_{(tot)}$ as the total beta count rate from a mixture source, then :

$$N_{\beta(137)} = N_{(tot)} - \frac{m_{(134)} \cdot A_{(134)}}{D_{(134)}} \cdot g(1 - \varepsilon_{\beta(134)})$$

There is a further complication arising from the decay scheme of ^{137}Cs , namely the high degree of internal conversion associated with the $^{137\text{m}}\text{Ba}$ 661.6 keV gamma transitions. The response of the PPC to the conversion electrons and unconverted photons facilitates the need for a correction to be applied to the extrapolated activity value for ^{137}Cs . Consider the beta count rate $N_{\beta(137)}$ per unit source mass at the reference time for a pure ^{137}Cs source :

$$\frac{N_{\beta(137)}}{m_{(137)}} = A_{(137)} \left(b_1 \varepsilon_{\beta_1} + b_2 \varepsilon_{\beta_2} + \frac{b_1}{1 + \alpha_T} (\alpha_T \varepsilon_{ce} + \varepsilon_{\beta\gamma}) \right)$$

where $m_{(137)}$ is the source mass, b_1 and b_2 are the branching ratios for the β_1 and β_2 branches which have efficiencies ε_{β_1} and ε_{β_2} , α_T is the total internal-conversion coefficient, ε_{ce} is the efficiency of the PPC to conversion electrons and $\varepsilon_{\beta\gamma}$ is the efficiency of the PPC to 661.6 keV photons.

As the extrapolations are performed to unit beta efficiency (for ε_{β_1} and ε_{β_2}), a correction factor must be applied to the extrapolated result, namely

$$A_{(137)} = \frac{N_{\beta(137)}}{m_{(137)} \cdot (1 + C)}$$

where :

$$C = \frac{b_1}{1 + \alpha_T} (\alpha_T \varepsilon_{ce} + \varepsilon_{\beta\gamma})$$

ε_{ce} is considered to be unity for this case. The values for b_1 and α_T are given in Section 5 of this report. The value of $\varepsilon_{\beta\gamma} = 0.001(1)$ was estimated from previous measurements with the PPC system. This yields a value for the correction factor for the term $(1 + C) = 1.0945(27)$.

Thus, to determine the activity concentration of the solution of ^{137}Cs , for each mixed source, the total beta count rate of the mixture $N_{0(mix)}$ (determined by extrapolation to unit beta efficiency) must be reduced by the contribution due to the ^{134}Cs :

$$A_{(137)} = \frac{A_{(tot)} - A_{(134)} \cdot m_{(134)} / D_{(134)}}{(1 + C) m_{(137)} / D_{(137)}}$$

where the terms $D_{(134)}$ and $D_{(137)}$ relate to the appropriate decay correction factors.

This procedure was performed for a set of mixed sources, and the weighted mean of the results determined to yield the activity concentration of the ^{137}Cs solution.

.....

7. ^{134}Cs Results and Uncertainties

7.1 ^{134}Cs results

First order polynomial extrapolations of $N_{\beta(134)}$ versus $(1 - N_c/N_\gamma)$ to unit beta efficiency were performed on the ^{134}Cs source which yielded the highest beta efficiency (134Cs0308). The results of this extrapolation are shown in Figure 10.

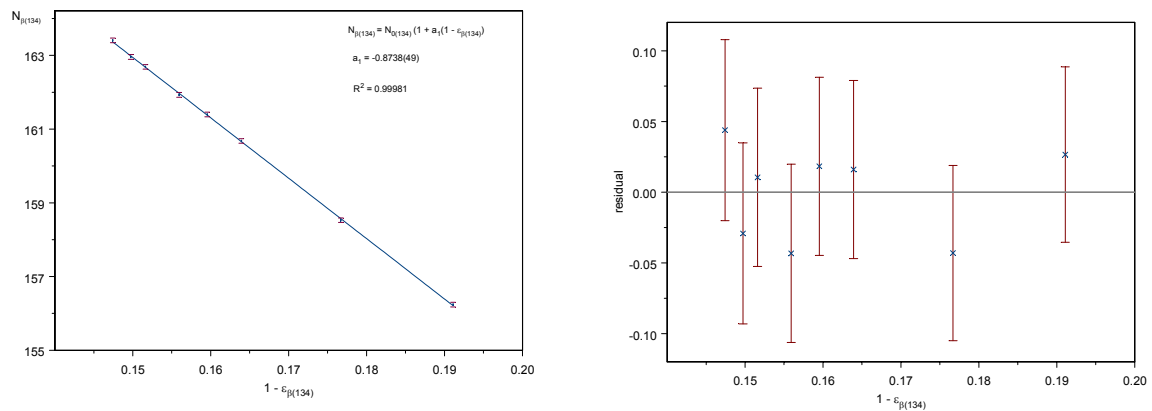


Figure 10 : Efficiency extrapolation for ^{134}Cs source 134Cs0308.

This yielded a mean slope to intercept ratio of $-0.8738(49)$ which was used to correct the data from the remaining ^{134}Cs sources,

i.e.:

$N_{\beta(134)}$ may be expressed as :

$$N_{\beta(134)} = N_{0(134)} [\varepsilon_\beta + (1 - \varepsilon_\beta) \varepsilon_{\beta\gamma}]$$

$$N_{\beta(134)} = N_{0(134)} [1 - (1 - \varepsilon_\beta) \cdot (1 - \varepsilon_{\beta\gamma})]$$

Then the slope is given by $-N_{0(134)} \cdot (1 - \varepsilon_{\beta\gamma})$, the intercept by $N_{0(134)}$ and the slope to intercept ratio is thus $-(1 - \varepsilon_{\beta\gamma}) = a_1$.

The corrected activities for each source of pure ^{134}Cs are given by :

$$N_{0(134)} = \frac{N_{\beta}}{1 + a_1(1 - \varepsilon_{\beta})}$$

The activity concentration for the ^{134}Cs solution, taken as the weighted mean of the results was found to be :

190.40(63) kBq.g⁻¹

at the reference time : (2003-07-01, 0h UTC).

7.2 Uncertainty components of the ^{134}Cs activity concentration

		Remarks
Type A Uncertainty (%)		
	0.3	Standard deviation of mean
Type B Uncertainties (%)		
weighing.	0.12	only 9 sources used
beta dead time.	0.02	propagation of 50 ns error in dead time
gamma dead time.	0.01	propagation of 50 ns error in dead time
background.	0.05	repeated background measurements
pile-up.	0.02	estimated : low count rates
resolving time.	0.02	propagation of 50 ns error in resolving times
Gandy effect.	0.02	propagation of 50 ns error in delay
counting time.	0.05	
slope/intercept ratio.	0.14	prop. of error through calculations
choice of fit.	0.25	half the difference between 1 st order and 2 nd order polynomial results (to account for large extrapolation range)
half life ($T_{1/2} = 754.28(22)$ d)	0.002	prop. of error through calculations
combined uncertainty*	0.43%	

* The uncertainty components are considered as approximations of the corresponding standard deviations, and are combined in quadrature.

8. ^{137}Cs Results and Uncertainties

8.1 ^{137}Cs Results

Only those sources with the common mixing ratio of ^{134}Cs and ^{137}Cs were considered in this standardisation. 2nd order polynomial extrapolations of apparent N_{β} per total source mass versus $(1 - N_{\alpha}/N_{\gamma})$ to unit beta efficiency were performed for the mixture source MixCs0320.

....

This extrapolation data was used to correct the data from the remaining mixture sources, in a manner analogous to that detailed in Section 7.1, only taken to second order in the beta inefficiency.

From the extrapolated “apparent” activities of the mixture sources, the contribution arising from the ^{134}Cs tracer was subtracted as detailed in Section 4.

The mean activity concentration for the ^{137}Cs solution, taken as the weighted mean of the results, was found to be :

270.0(13) kBq.g⁻¹.

at the reference time : (2003-07-01, 0h UTC).

The initial beta efficiencies for all sources were low (around 80%). This coupled with the curvature of the efficiency extrapolation curves for the mixture sources yielded the relatively high uncertainty values for the standardisation of ^{137}Cs . Further work is required to understand the cause of this low efficiency in the PPC.

8.2 Uncertainty components of the ^{137}Cs activity concentration

		Remarks
Type A Uncertainty (%)		
	0.48	Standard deviation of mean
Type B Uncertainties (%)		
weighing	0.12	-----
beta dead time	0.02	propagation of 50 ns error in dead time
gamma dead time	0.01	propagation of 50 ns error in dead time
background	0.05	repeated background measurements
pile-up	0.02	-----
resolving time	0.02	propagation of 50 ns error
Gandy effect	0.02	propagation of 50 ns error in delay
counting time	0.05	-----
^{134}Cs tracer activity. activity concentration.	0.30	propagation of uncertainty in ^{134}Cs
correction factor (1 + C) (including b_1 and $\varepsilon_{\beta\gamma}$ terms)	0.25	prop. of uncertainty (correction factor)
impurities	0.015	from (Altitzoglou, 2003a)
half life of ^{134}Cs tracer ($T_{1/2} = 754.28(22)$ d)	0.002	prop. of error through calculations
half life of ^{137}Cs ($T_{1/2} = 11020(60)$ d)	0.01	prop. of error through calculations
combined uncertainty *	0.63%	

* The uncertainty components are considered as approximations of the corresponding standard deviations, and are combined in quadrature.

.....

9. Final results

9.1 Final result : ^{134}Cs

The activity concentration of the ^{134}Cs solution on the reference date (2003-07-01, 0 h UTC) was found to be **190.4 kBq.g⁻¹**,

with the combined uncertainty **0.8 kBq g⁻¹, (0.43 %)**

9.2 Final result : ^{137}Cs

The activity concentration of the ^{137}Cs solution on the reference date (2003-07-01, 0 h UTC) was found to be **270.0 kBq.g⁻¹**,

with the combined uncertainty **1.7 kBq g⁻¹, (0.7 %)**

....

11 References

Altitzoglou T., (2003a)., Radioactivity measurements of a solution of ^{134}Cs (July 2003). *Private communication*, dated August 29, 2003.

ICRU., (1994). Particle counting in radioactivity measurements. International Commission on Radiation Units and Measurements. ICRU Report 52.

Reher D, Celen E., Vaninbroukx R., Zehner W., (1982). The 1982 International Comparison of Activity measurements of a Solution of ^{137}Cs ., *CBNM Report GE/R/RN/14/82*.

Rytz A., (1983) International comparison of activity measurements of a solution of ^{137}Cs ., *Rapport BIPM-83/8*.

Rytz A. (1985) Activity Concentration of a solution of ^{137}Cs : AN INTERNATIONAL COMPARISON., *Nucl. Instrum. And Meth. In Phys. Res.*, **228**, pp. 506 – 511

Smith D., (1987). Some developments in the Cox-Isham theory of coincidence corrections, including the extension to the computer discrimination method. *Int. J. Appl. Radiat. Isot. A38 813 and corrigenda : Int. J. Appl. Radiat. Isot. A39* (1988) 362

Annex 5: Traceability to BIPM SIR

BIPM, F-92310 SEVRES

International Reference System for the Activity Measurement of Gamma-Ray Emitting Nuclides (SIR)

Radionuclide: ^{137}Cs

Half life adopted: $T_{1/2} = (11\,020.8 \pm 1.2) \text{ d}$

Laboratory	Ampoule number	Method of standardisation	Reference date	Data reported by laboratory			Ionization-chamber measurements carried out at BIPM			
				Activity at ref. date (kBq)	Rel. uncert. Category A B (r ₁ ,%) (r ₂ ,%)	Date	Relative uncertainty (r ₃ , %)	Activity A _e which would produce the same ion current as the Ra source A _e (kBq)	Combined uncert. of A _e A _e (r ₁ ² + r ₂ ² + r ₃ ²) ^{1/2} (kBq)	
IRMM	137Cs0310	4πβ-NaI(Tl)γ and efficiency tracing with ^{134}Cs and LS counting (CIEMAT/NIST method)	2003-07-01	980.8	0.59	0.0	2004-01-20	0.093	27 337	163

The key comparison reference value (KCRV) for ^{137}Cs has been identified as:

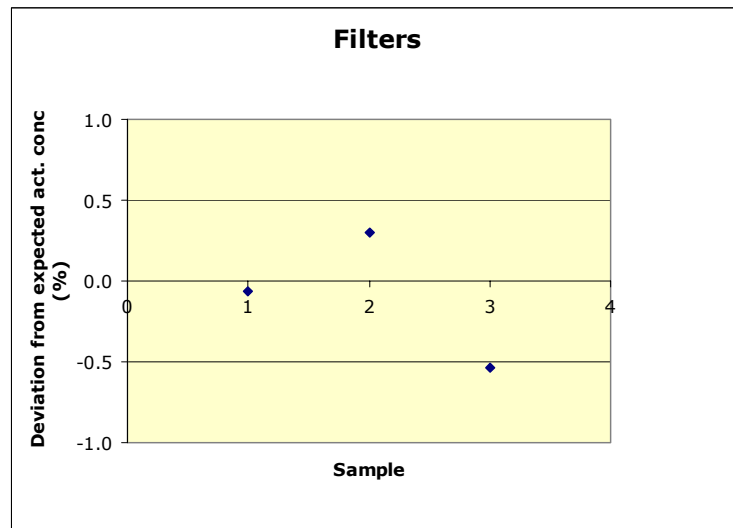
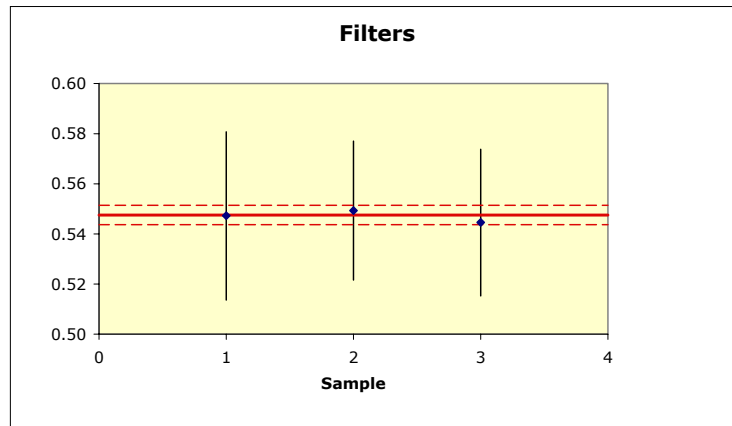
27 549 ± 44 kBq

(BIPM comparison BIPM.RI (II)-K1.Cs-137 of activity measurements of the radionuclide ^{137}Cs and links for the 1982 international comparison CCRI(II)-K2Cs-137, G. Ratel, C. Michotte, BIPM 2003/07/17)

Annex 6: Quality control measurement results of air filter samples and dilution and spiking solutions

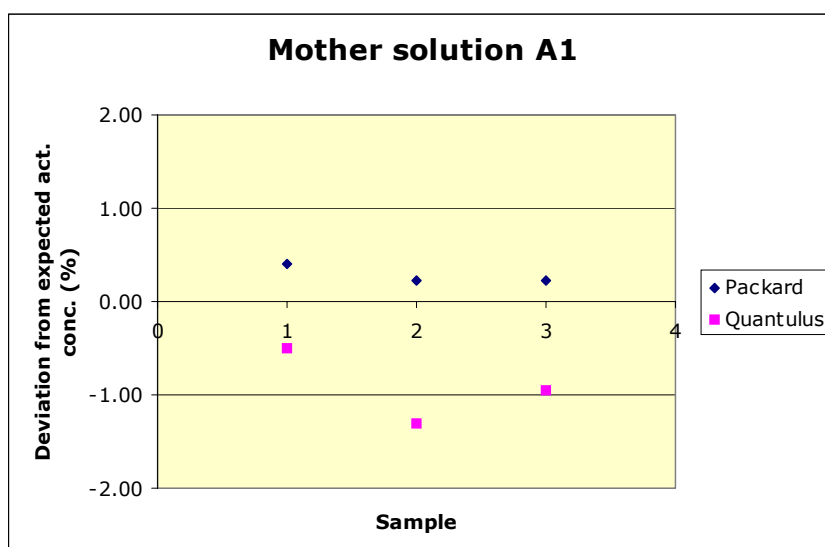
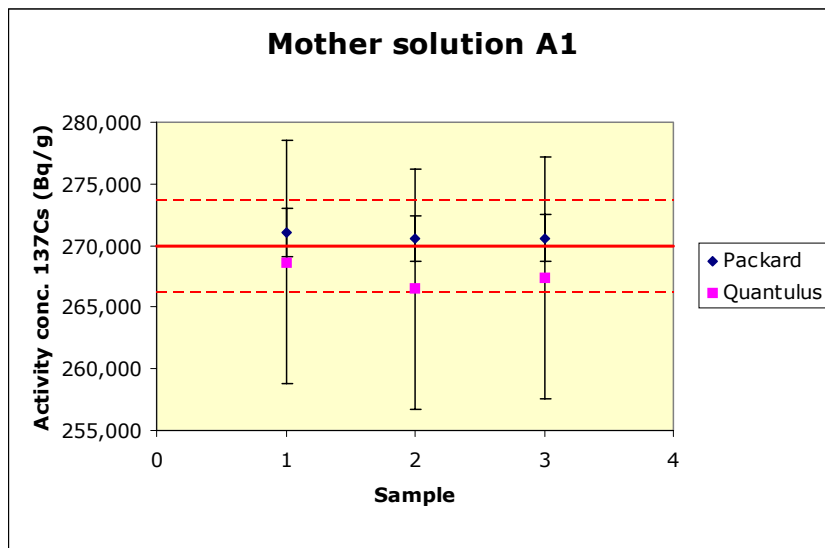
A. Filters

Sample #	Sample Origin and ID	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
F1	bottle C2 Filter_RN_001	0.55	6.1	-0.07
F2	Cs137C20302 Filter_RN_002	0.55	5.0	0.30
F3	Cs137C2/2 Filter_RN_003	0.54	5.4	-0.54



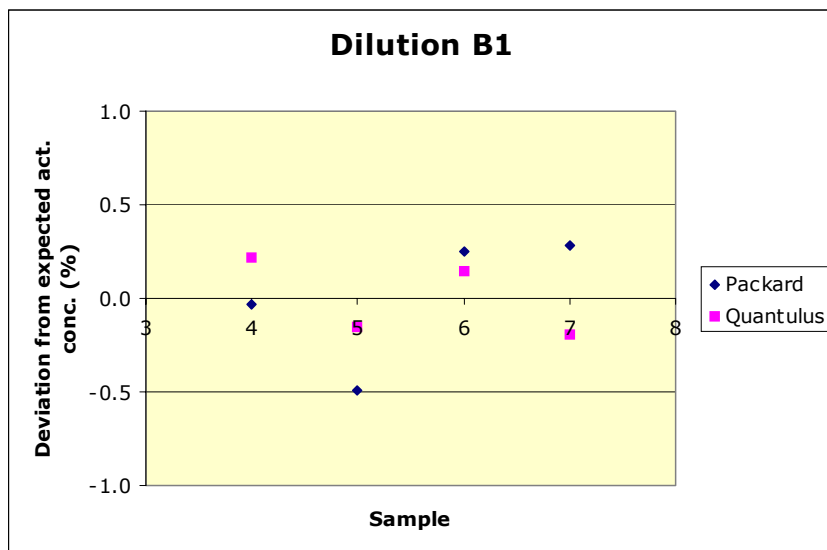
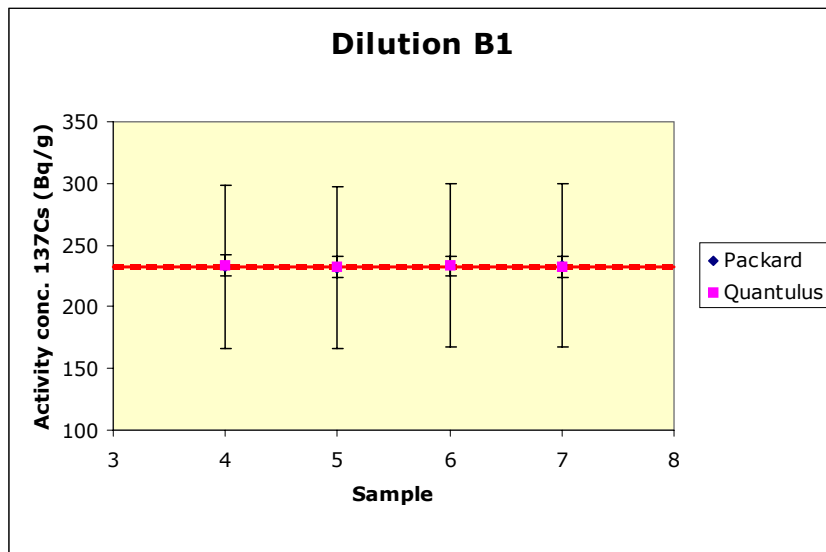
B. Mother solution (A1)

Sample #	Sample Origin and ID	Packard LSC			Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
Mother solution A1							
1	LSC_137Cs_S01	2.71E+05	0.7	0.4	2.69E+05	3.7	-0.5
2	LSC_137Cs_S02	2.71E+05	0.7	0.2	2.66E+05	3.7	-1.3
3	LSC_137Cs_S03	2.71E+05	0.7	0.2	2.67E+05	3.7	-1.0



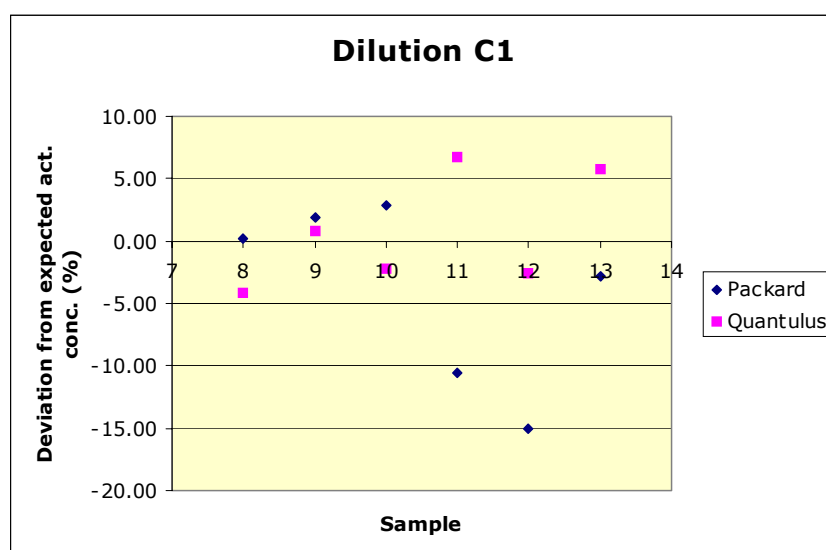
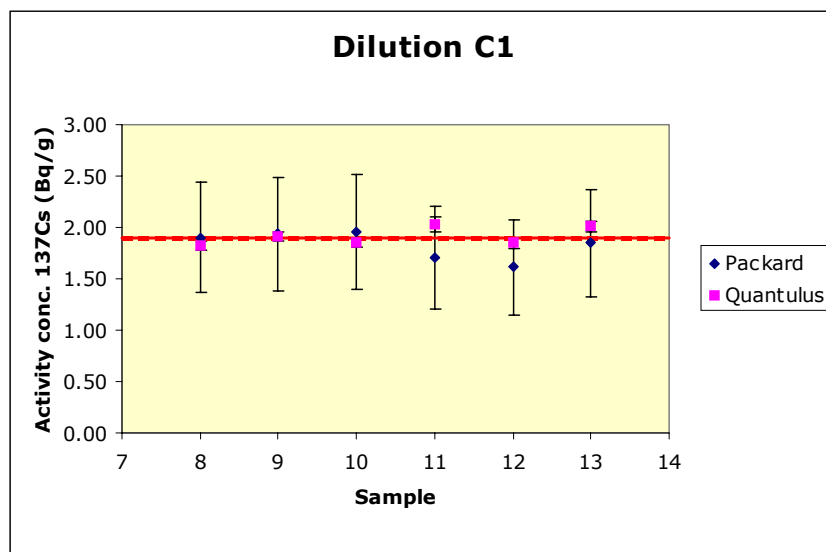
C. Dilution B1

Sample #	Sample Origin and ID	Packard LSC			Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
	bottle B1						
4	LSC_137Cs_S04	233	28	0.0	233	3.7	0.22
5	LSC_137Cs_S05	232	28	-0.5	232	3.7	-0.15
6	LSC_137Cs_S06	233	28	0.3	233	3.7	0.14
7	LSC_137Cs_S07	233	28	0.3	232	3.7	-0.19



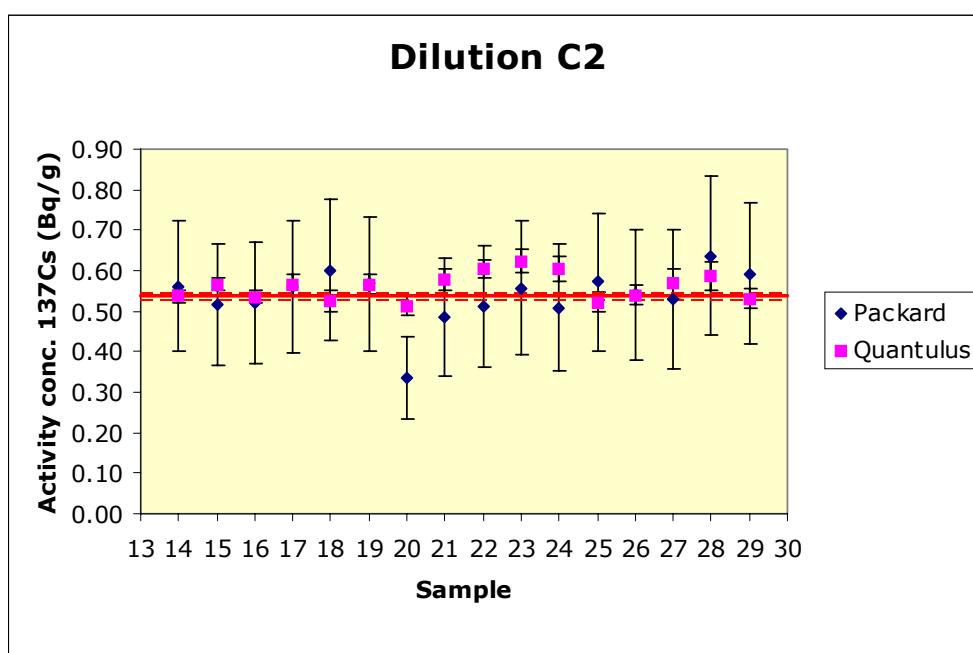
D. Dilution C1

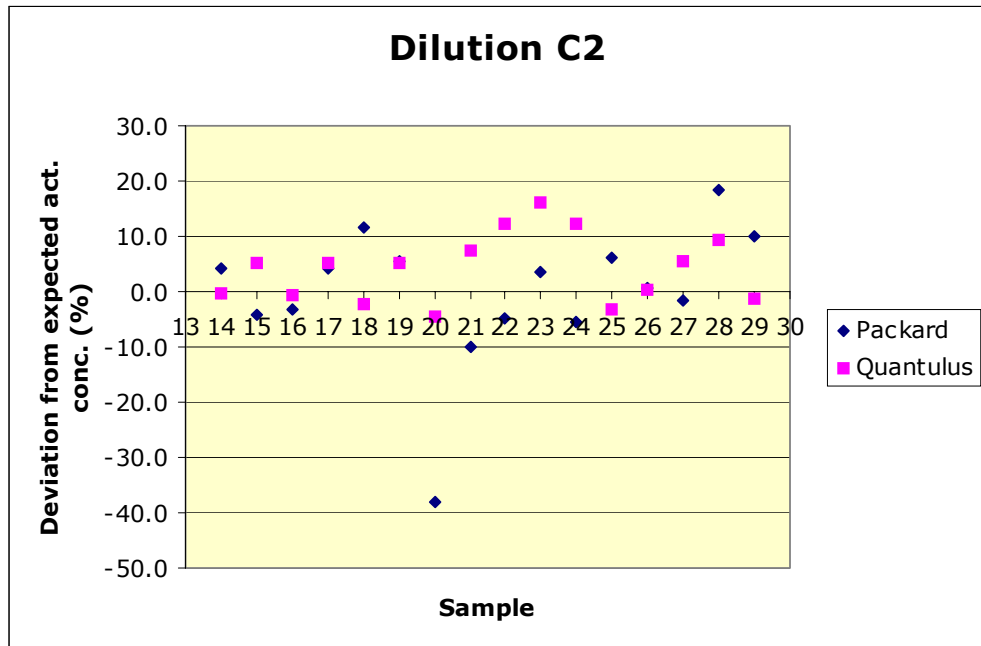
Sample #	Sample Origin and ID	Packard LSC			Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
bottle C1							
8	LSC_137Cs_S08	1.9	29	0.2	1.82	2.4	-4.2
9	LSC_137Cs_S09	1.9	28	1.9	1.91	2.3	0.8
10	LSC_137Cs_S10	2.0	29	2.8	1.86	2.4	-2.2
ampoule Cs137C1							
11	LSC_137Cs_S27	1.7	29	-11	2.03	3.8	6.8
12	LSC_137Cs_S28	1.6	29	-15	1.85	2.9	-2.6
13	LSC_137Cs_S29	1.8	29	-2.8	2.01	2.7	5.8



E. Dilution C2

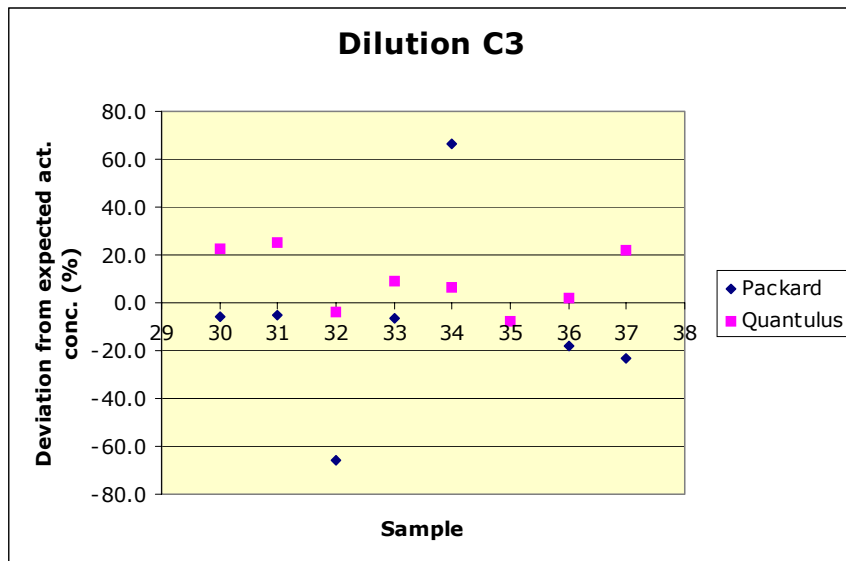
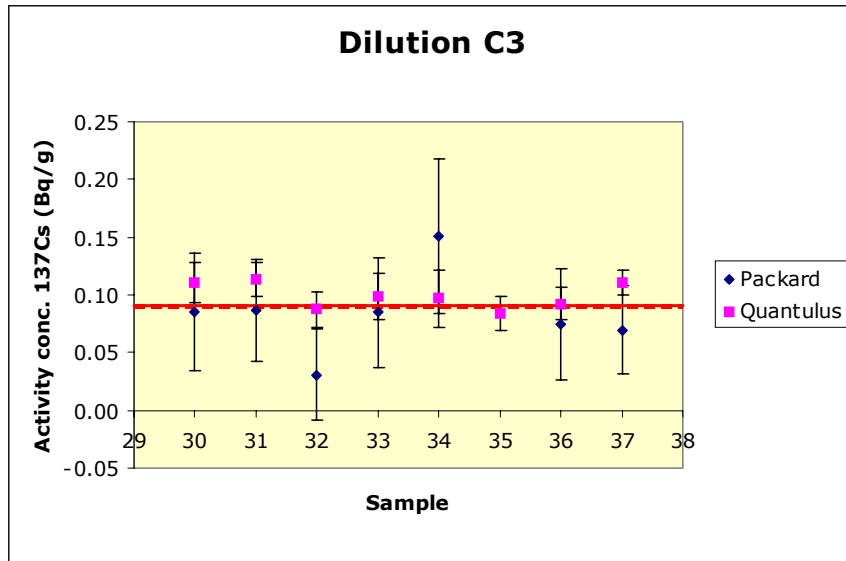
Sample #	Sample Origin and ID	Packard LSC			Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
bottle C2							
14	LSC_137Cs_S11	0.6	29	4.3	0.54	2.9	-0.3
15	LSC_137Cs_S12	0.5	29	-4.1	0.57	2.9	5.1
16	LSC_137Cs_S13	0.5	29	-3.3	0.53	2.9	-0.6
17	LSC_137Cs_S17	0.6	29	4.2	0.57	4.7	5.1
18	LSC_137Cs_S18	0.6	29	11.5	0.53	4.8	-2.3
19	LSC_137Cs_S19	0.6	29	5.6	0.57	4.5	5.3
20	LSC_137Cs_S20	0.3	30	-37.9	0.51	4.7	-4.5
ampoule Cs137C20301							
21	LSC_137Cs_S21	0.5	30	-9.8	0.58	4.6	7.4
22	LSC_137Cs_S22	0.5	30	-5.0	0.60	3.9	12.4
23	LSC_137Cs_S23	0.6	30	3.5	0.62	4.3	16.0
ampoule Cs137C20302							
24	LSC_137Cs_S24	0.5	31	-5.4	0.60	5.1	12.2
25	LSC_137Cs_S25	0.6	30	6.3	0.52	4.5	-3.1
26	LSC_137Cs_S26	0.5	30	0.7	0.54	4.6	0.4
ampoule Cs137C2/1							
27	LSC_137Cs_S35	0.5	32	-1.6	0.57	6.6	5.4
28	LSC_137Cs_S36	0.6	31	18.4	0.59	5.9	9.2
29	LSC_137Cs_S37	0.6	29	10.1	0.53	4.5	-1.4





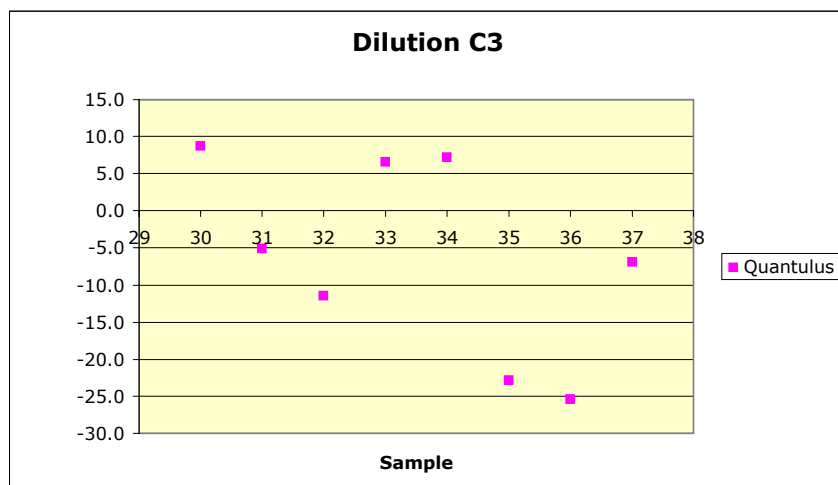
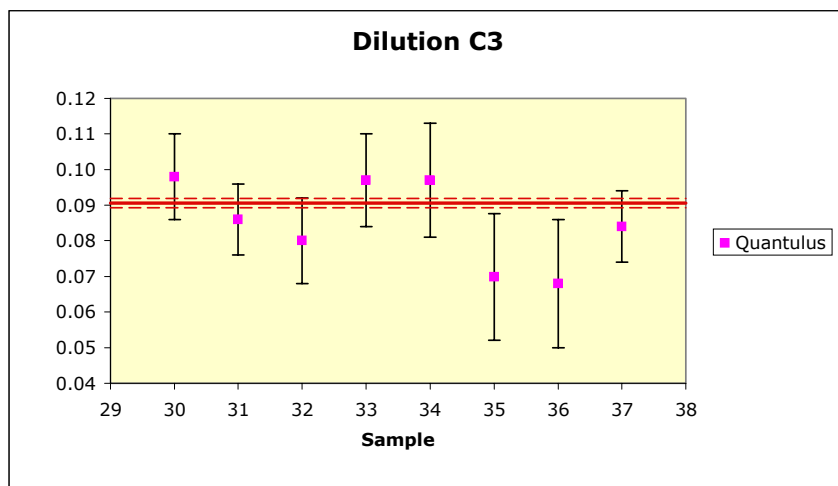
F. Dilution C3

Sample #	Sample Origin and ID	Packard LSC			Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)	Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
bottle C3							
30	LSC_137Cs_S14	0.09	60	-5.9	0.11	15.8	22
31	LSC_137Cs_S15	0.09	51	-4.9	0.11	12.8	25
32	LSC_137Cs_S16	0.03	126	-66	0.09	17.1	-3.6
ampoule Cs137C30302							
33	LSC_137Cs_S30	0.08	56	-6.5	0.10	20.1	9.2
34	LSC_137Cs_S31	0.15	44	66	0.10	25.5	6.8
ampoule Cs137C3/1							
35	LSC_137Cs_S32	<background			0.08	17.8	-7.8
ampoule Cs137C3/2							
36	LSC_137Cs_S33	0.07	65	-18	0.09	15.6	2.0
37	LSC_137Cs_S34	0.07	55	-23	0.11	10.1	22



G. Dilution C3 Re-measured with longer counting times

Sample #	Sample Origin and ID	Quantulus LSC		
		Activity Concentration (Bq/g)	Unc. (%)	Deviation (%)
bottle C3				
30	LSC_137Cs_S14	0.10	12.2	8.7
31	LSC_137Cs_S15	0.09	11.6	-5.1
32	LSC_137Cs_S16	0.08	15.0	-11.4
ampoule Cs137C30302				
33	LSC_137Cs_S30	0.10	13.4	6.6
34	LSC_137Cs_S31	0.10	16.5	7.2
ampoule Cs137C3/1				
35	LSC_137Cs_S32	0.07	25.4	-22.9
ampoule Cs137C3/2				
36	LSC_137Cs_S33	0.07	26.5	-25.4
37	LSC_137Cs_S34	0.08	11.9	-6.9



Annex 7: List of spiking solutions and air filter samples prepared for the REM 2003 exercise

QC Sample #	Sample Origin and ID
	mother solution A1 137Cs0314
	sources from mother solution A1 ampoule 137Cs0314
	LSC vials
1	LSC_137Cs_S01
2	LSC_137Cs_S02
3	LSC_137Cs_S03
	Dilution B1 in bottle B1 CsCl carrier in B1 Cs137 in B1
	sources from dilution B1 bottle B1
	LSC vials
4	LSC_137Cs_S04
5	LSC_137Cs_S05
6	LSC_137Cs_S06
7	LSC_137Cs_S07
	Dilution C1 in bottle C1 CsCl carrier in C1 Cs137 in C1 blue carrier in C1
	Dilution C2 in bottle C2 CsCl carrier in C2 Cs137 in C2 blue carrier in C2
	Dilution C3 in bottle C3 CsCl carrier in C3 Cs137 in C3 blue carrier in C3
	Ampoules 137CsB10301 137CsB10302 137CsB10303
	sources from dilution C1 bottle C1
8	LSC_137Cs_S08 137CsC10301
9	LSC_137Cs_S09 137CsC10302
10	LSC_137Cs_S10
	ampoule Cs137C1
11	LSC_137Cs_S027

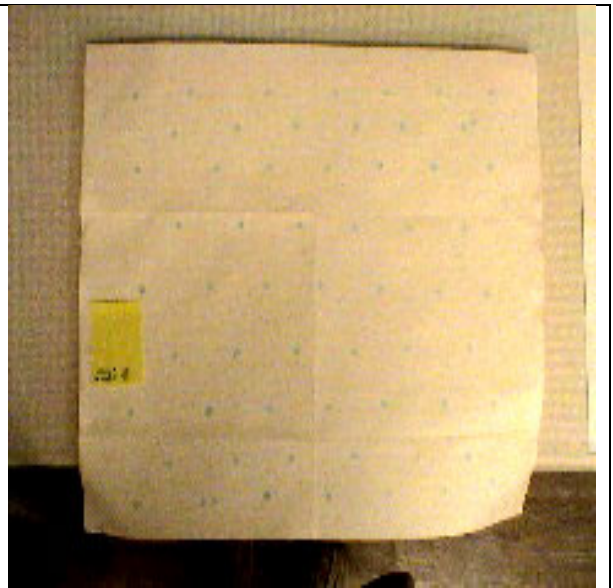
	Filter_005
	Filter_039
	Filter_043
12	LSC_137Cs_S028
13	LSC_137Cs_S029
	sources from dilution C2
	bottle C2
14	LSC_137Cs_S11 137CsC20301
15	LSC_137Cs_S12 137CsC20302
16	LSC_137Cs_S13
	bottle C2
17	LSC_137Cs_S17
18	LSC_137Cs_S18
	Filter_001
	Filter_002
	Filter_012
	Filter_016
	Filter_019
	Filter_035
	Filter_036
	Filter_041
	Filter_017
19	LSC_137Cs_S19
20	LSC_137Cs_S20
	Filter_046
	Filter_047
F1	Filter_RN001
	Filter_038
	ampoule Cs137C20301
21	LSC_137Cs_S021
22	LSC_137Cs_S022
	Filter_003
	Filter_004
	Filter_008
	Filter_013
	Filter_014
	Filter_015
	Filter_031
	Filter_042
	Filter_027
	Filter_020
	Filter_023
23	LSC_137Cs_S023
	ampoule Cs137C20302
24	LSC_137Cs_S024
25	LSC_137Cs_S025
	Filter_006

	Filter_037
	Filter_024
	Filter_022
	Filter_033
26	LSC_137Cs_S026
	Filter_032b
F2	Filter_RN_002
	ampoule Cs137C2/1
27	LSC_137Cs_S035
	Filter_018
	Filter_021
	Filter_026
	Filter_040
	Filter_034
28	LSC_137Cs_S036
29	LSC_137Cs_S037
	ampoule Cs137C2/2
	LSC_137Cs_LS01
	Filter_017a
	Filter_010
	LSC_137Cs_LS02
F3	Filter_RN003
	sources from dilution C3
	bottle C3
30	LSC_137Cs_S14
	137CsC30301
	137CsC30302
31	LSC_137Cs_S15
32	LSC_137Cs_S16
	ampoule Cs137C30302
33	LSC_137Cs_S030
	Filter_007
	Filter_009
	Filter_011
	Filter_045
34	LSC_137Cs_S031
	ampoule Cs137C3/1
35	LSC_137Cs_S032
	Filter_025
	ampoule Cs137C3/2
36	LSC_137Cs_S033
	Filter_044
37	LSC_137Cs_S034

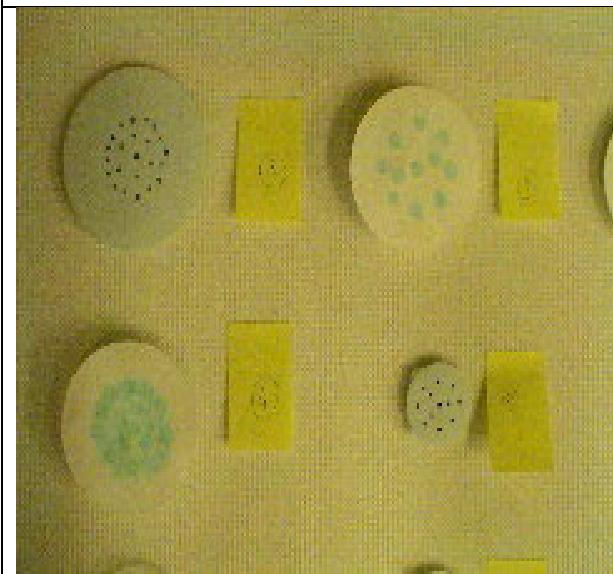
Annex 8: Examples of spiking



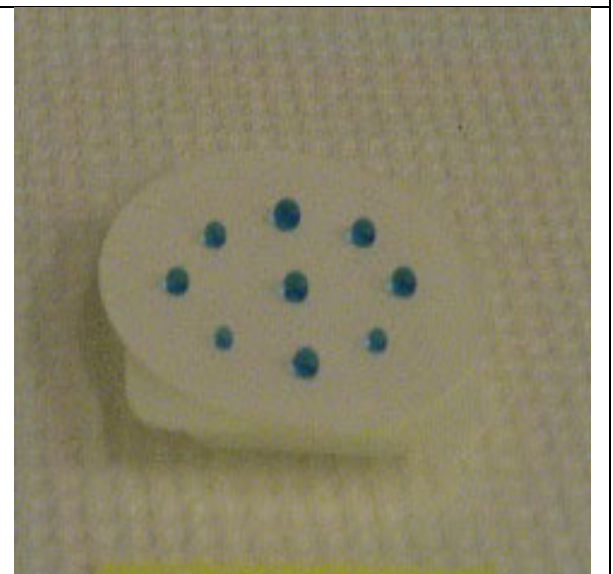
a)



b)



c)



d)

European Commission

EUR 22926 EN – Joint Research Centre – Institute for Reference Materials and Measurements

Title: Evaluation of EC measurement comparison on simulated airborne particulates - ^{137}Cs in air filters

Authors: U. Wätjen, Zs. Szántó, T. Altizoglou, G. Sibbens, J. Keightley, R. Van Ammel, M. Hult and M. De Cort

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Abstract

This report describes the full life cycle of the measurement comparison of ^{137}Cs in air filters among 43 European laboratories monitoring radioactivity in the environment. Gravimetrically pipetting droplets of a gravimetrically diluted standardised ^{137}Cs solution onto real air filters, SI-traceable reference values were established for intercomparison filters carrying a large range of activity close to the routine measurement conditions of the corresponding laboratory. The sample preparation and measurement processes applied in the participating laboratories are described and the results of the intercomparison are presented and discussed in detail. The results point at some problems of radioactivity measurement in air filters which need to be improved by several laboratories. Overall, with 41 out of 48 reported measurement results lying within $\pm 33\%$ of the IRMM reference value, this comparison renders a rather fair result.

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