

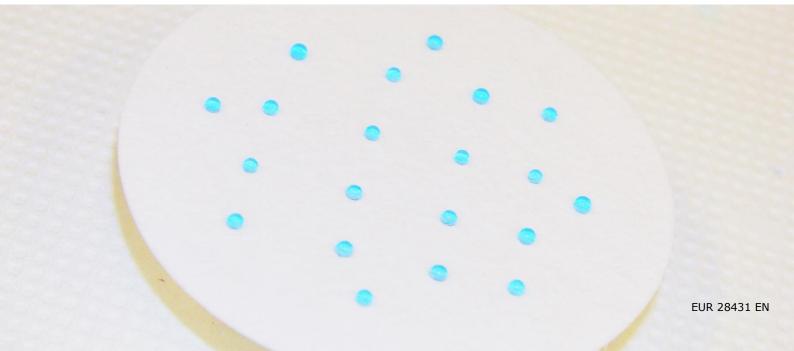
## JRC TECHNICAL REPORTS

Evaluation of the 2016 ENV57/MetroERM measurement comparison on simulated airborne particulates:

<sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I in air filters

Timotheos Altzitzoglou Petya Malo

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## **Abstract**

In 2016, the European Commission (EC) Directorate-General Joint Research Centre (JRC) Geel site (former Institute for Reference Materials and Measurements (IRMM)) organized an interlaboratory comparison (ILC) exercise on the measurement of  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I in air filters. Similar exercises, although EC ILC, were organized in 2014 (Altzitzoglou and Máté, 2016) and in 2003 (Wätjen et al., 2007); in both exercises the measurand was  $^{137}$ Cs only.

The 2016 ILC, which is the subject of this report, was conducted in the frame of the EMRP project with code ENV57 and entitled MetroERM "Metrology for radiological early warning networks in Europe" project with the aim to optimise the metrological foundation of measurements for monitoring airborne radioactivity and promote pan-European harmonisation in data reliability.

The JRC participates in the ENV57 MetroERM project having the responsibility to carry out a number of tasks. Work Package 3 (WP3) addresses the traceability of dose rate and airborne radioactivity measurements and explicitly supports the process of harmonisation of radiological data from early warning networks in Europe by systematic investigations, comparison exercises and the publication of recommendations. Task 3.2 of WP3 entitled "Traceability management for airborne radioactivity" aims in developing traceable reference materials and standard sources in the form of large-area spiked aerosol filters and conducting a laboratory comparison exercise to quantify the performance of the airborne radioactivity measuring field stations.

This report describes the full life cycle of the above mentioned comparison among 67 European laboratories monitoring radioactivity in the environment. JRC provided the comparison samples, which were prepared individually for each laboratory using a gravimetrically diluted solution of  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I. Reference values traceable to the International System of Units (SI) and the International Reference System (SIR) for gamma-ray emitting radionuclides were determined at the JRC. The samples were made by gravimetrically dispensing the appropriate activity amounts, close to those the laboratories routinely measure, on blank air filters provided by the participants. A robust evaluation of the individual performance using three different approaches, percentage difference ( $D_{\%}$ ),  $E_{\text{n}}$  numbers and PomPlots, is presented. Finally, for the laboratories which have participated in the 2003 and 201 exercises, their performance evolution in measuring  $^{137}$ Cs in air filters over the years, is examined.

All 67 participating laboratories reported valid results. The majority of the laboratories reported reliable measurement results for  $^{137}$ Cs and  $^{134}$ Cs; 56 (84%) out of the 67 participants reported values with a percentage difference from the reference value within the  $\pm 20\%$  range. Furthermore, 42 (63%) of the laboratories fulfilled the criterion of the compatibility test based on  $E_n$  numbers for  $^{137}$ Cs and 36 (54%) for  $^{134}$ Cs. As the calculation of the  $E_n$  numbers takes into account the uncertainties on both the measured activity and the reference value, the lower scores for the  $E_n$  numbers reveals that the uncertainty estimation is not adequate in many laboratories and there is a need to improve their application of uncertainty propagation. An observation for the  $^{134}$ Cs results is a negative bias on the reported results compared to the reference values, which might be partly attributed to a non-adequate summing correction applied by some laboratories.

The evaluation of the performance of the laboratories on  $^{131}$ I was complicated by two effects. First, in a number of spiked air filters a fraction of the  $^{131}$ I activity was transferred to the protective plastic bag. Second, the integrity of the  $^{131}$ I activity content of many spiked air filters was compromised to a variable degree. Nevertheless, 20 (30%) laboratories reported results for  $^{131}$ I with a percentage difference from the reference value within the  $\pm 20\%$  range and 9 (13%) with compatible  $E_n$  numbers. This performance is not satisfactory, but the difficulties encountered have to be taken into account. A major output of this project is the

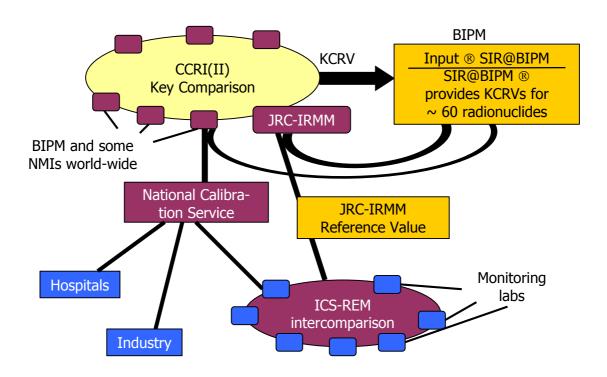
identification of the need of additional studies on how to perform reliable measurements of radioiodine on air filters.

Overall, the majority of laboratories master their gamma-ray spectrometry analytical procedures, including the counting efficiency calibration of the detection systems and the corrections for coincidence summing. More attention has to be paid to the elaboration of the uncertainty to obtain correct and realistic uncertainties on the reported results.

#### 1. Introduction

According to the Articles 35/36 of the Euratom Treaty (Euratom, 2012) and the Commission Recommendation 473/2000 (2000/473/Euratom, 2000) derived from the Euratom Treaty, the Member States (MS) of the European Union (EU) have the legal obligation to inform the European Commission (EC) on a regular basis on the radioactivity levels in their environment (drinking water, soil, air and mixed diet). In order to obtain more information on the MS's measurement methods and on the quality of their reported values for the radioactivity levels determined in their environment, the EC has established the International Comparison Scheme for Radioactivity Environmental Monitoring (ICS-REM) (Wätjen, 2008).

The JRC-Geel (from 1993 to 2016 named "Institute for Reference Materials and Measurements" (IRMM) and earlier CBNM "Central Bureau for Measurements") is one of the seven institutes of the European Commission's Directorate-General Joint Research Centre (JRC). In the frame of ICS-REM, the Directorate for Nuclear Safety and Security and in particular the Radionuclide Metrology Sector of the Standards for Safety, Security and Safeguard Unit of the JRC, organises on request of the Directorate-General for Energy (DG ENER) the EC Interlaboratory Comparisons (EC ILCs) since 2003. The aim of the EC ILCs is not only to evaluate the results submitted by the participants but also to provide help and advice to the participating MS laboratories via workshops and meetings on how to improve the measurements and methods applied by them. During the last decade, the test materials used in EC ILCs have included air filters (2003), soil samples (2010) and foodstuff samples, such as milk powder (2005), bilberry powder (2011), mineral water (2008) drinking water (2012) and <sup>137</sup>Cs in air filters (2014). The approach of JRC-Geel in organising the comparisons is sketched in *Figure 1*.



**Fig. 1**. Key comparisons of CCRI(II) and traceability of the reference values for samples provided by JRC for the interlaboratory comparisons amongst monitoring laboratories (KCRV = Key Comparison Reference Value).

The EU Member States are obliged to report radiological monitoring data of airborne radioactivity to the EC according to the Council Decision 600/87 (87/600/Euratom,

1987) and the Commission Recommendation 473/2000, (2000/473/Euratom, 2000). These measurements are collected by the JRC in the European Union Radiological Data Exchange Platform (EURDEP, 2016), which makes the non-validated radiological monitoring data available in nearly real-time for the countries reporting to the system. For the moment there are about 5000 operational stations to monitor the airborne radioactivity and dose rate within the European early warning network and EURDEP, but the number of stations with sampling equipment for radioactive particulates in air is significantly lower (~240) than those with instrumentation for dose rate measurements. Furthermore, the stations with sampling equipment use a wide variety of different methods, instruments and air filters.

During a radiological emergency with trans-boundary implications in Europe, the European Commission will issue recommendations to EU Member States based on data from national early warning networks. Therefore, metrologically sound monitoring data of ambient dose rate and airborne radionuclide activity concentrations, coordinated with data from international radiological networks, are a prerequisite for adequate environmental radiation monitoring in Europe.

The ENV57 MetroERM "Metrology for radiological early warning networks in Europe" project (MetroERM, 2016) in the frame of the European Metrology Research Program (EMRP) aims at optimising the metrological foundation of measurements (devices and methods) for monitoring airborne radioactivity and promoting pan-European harmonisation in data reliability for area dose rate measurements which are input to the European Radiological Data Exchange Platform (EURDEP) and other monitoring networks. It will provide the unique possibility to comprehensively address the harmonisation of the radiological early warning networks in Europe - the largest and most comprehensive environmental radiation monitoring system worldwide.

The JRC participates in the ENV57 MetroERM project, having the responsibility to carry out a number of tasks. Work Package 3 (WP3) addresses the traceability of dose rate and airborne radioactivity measurements and explicitly supports the process of harmonisation of radiological data from early warning networks in Europe by systematic investigations, comparison exercises and the publication of recommendations.

Task 3.2 of WP3 entitled "Traceability management for airborne radioactivity" aims at developing traceable reference materials and standard sources in the form of large-area spiked aerosol filters and conducting a laboratory comparison exercise to quantify airborne radioactivity field station performance. Deliverables D3.2.1 through D.3.2.8 are directly linked to the present intercomparison exercise, of which the JRC undertook the organization and execution. Similar exercises, although EC ILC, were organized in 2014 (Altzitzoglou and Máté, 2016) and in 2003 (Wätjen et al., 2007).

Initially, a questionnaire was conceived and distributed to all European network operators to collect information relevant to the preparation of the exercise. From the analysis of the answers, it was decided to conduct the ILC using <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I as measurands, a change from the two previous similar exercises where <sup>137</sup>Cs only was present in the distributed samples.

The aim of this ILC (or Proficiency Test (PT)) exercise was to quantify the performance of field stations measuring the airborne radioactivity. The participating laboratories provided blank air filters of the type they routinely use, which the JRC spiked gravimetrically with known amount of radioactivity individually for each laboratory and returned to them on February 22 and 23, 2016. For this exercise, the filters were spiked with  $^{137}\mathrm{Cs}$ ,  $^{134}\mathrm{Cs}$  and  $^{131}\mathrm{I}$  and with activities close to those the laboratories routinely measure, i.e. in the range from 0.2 to 5.0 Bq for  $^{137}\mathrm{Cs}$ , from 0.4 to 6.1 Bq for  $^{134}\mathrm{Cs}$  and from 0.6 to 8.7 Bq for  $^{131}\mathrm{I}$ ,.

The ILC was accompanied by a second questionnaire to collect information about sample collection, measurement and analysis, as routinely carried out by the

participating laboratories. The deadline to report the measurement results and submit the answers to the second questionnaire was 29 March 2016.

The aim of the present ILC was to obtain an overview of the quality of the results reported, of the application of the measurement methods by the participating laboratories and of any changes that have occurred since the last similar exercises in 2014 and 2003.

This report presents in detail all phases of the ILC organized and conducted in 2016, the description of the intercomparison sample preparation at JRC, the analytical methods used by the laboratories, the treatment of the data reported by the participants and, finally, the evaluation and comparison of the results to the reference values. A robust evaluation of the performance of individual laboratories was performed using three different approaches: percentage difference  $(D_{\%})$ ,  $E_{\rm n}$  numbers (ISO, 2005a), and 'PomPlots' (Pommé, 2016).

## 2. The 2016 ENV57 MetroERM measurement comparison

## 2.1 Description of the sample

Nature: blank air filters provided by the participating

laboratories spiked with a known amount of 137Cs,

<sup>134</sup>Cs and <sup>131</sup>I at JRC-Geel

Reference date: 1 March 2016 0:00 UTC

Activity levels: air filters spiked with activity levels similar to those

which the laboratories routinely measure, and in every case, activity levels above the reported detection limit (<sup>137</sup>Cs activity ranging from 0.2 up 5.0 Bq, <sup>134</sup>Cs activity ranging from 0.4 up 6.1 Bq, <sup>131</sup>I

activity ranging from 0.6 to 8.7 Bq)

Shipping: spiked air filters sealed in double plastic bags were

sent via regular mail to the participating laboratories

## 2.2 Participating laboratories

In December 2015, the JRC invited laboratories routinely measuring radioactivity in exposed air filters to participate at the 2016 ENV57/MetroERM interlaboratory comparison.

The laboratories participating in this ILC were 67 European laboratories from 29 countries, of which 61 from 26 EU Member States (MS) and 2 from Norway, 1 from Switzerland and 2 from Turkey. Of these laboratories, 57 had participated in the 2014 EC ILC and 26 in the 2003 EC ILC. Seven of the participating laboratories are also partners in the ENV57 MetroERM project.

All laboratories that registered to the ENV57/MetroERM ILC reported their results for all three radionuclides. The list of all participating laboratories is given in *Annex 1*. Since the anonymity is a requirement in this ILC, the identity of the laboratories is not shown in this compilation of the results. The laboratory numbers used throughout the data evaluation in this report are not related to the order of listing the participants in *Annex 1*, but to the Laboratory Code communicated only to the corresponding laboratory.

## 2.3 Reporting of results

The results for the activity per filter as well as the associated combined uncertainty with coverage factor k had to be reported in the unit Bq.

The reporting of the results together with a questionnaire was performed via an online reporting system, operated by JRC-Geel. Participants were asked to answer all relevant questions regarding the measurement procedure they followed. Information given in this questionnaire was essential for the evaluation of the ILC results. Moreover, it allowed to find possible sources of difficulties and to get an overview of the methods used by the laboratories.

The reference date for all results was 1 March 2016 0:00 UTC.

**Table 1**. Recommended nuclear data for the three radionuclides of interest (standard uncertainties in parenthesis (k=1)) (DDEP, 2016).

Nuclide	Half-life (d)	Energy (keV)	Photons per 100 disintegrations		
<sup>137</sup> Cs	10975 (29)	661.7	84.99 (20)		
		563.2	8.342 (15)		
		569.3	15.368 (21)		
<sup>134</sup> Cs	754.0 (5)	604.7	97.63 (8)		
		795.9	85.47 (9)		
		802.0	8.694 (16)		
		284.3	6.14 (6)		
<sup>131</sup> I	8.0233 (19)	364.5	81.2 (5)		
		637.0	7.12 (7)		

#### 2.4 Timetable of the ILC

26 November 2015: expression of interest

11 December 2015: invitation letter was sent to the interested laboratories

8 January 2016: nominated laboratories sent the blank air filters, together

with information on their routine measurement conditions

February 2016: spiked air filters were prepared

22-23 February 2016: spiked air filters were sent to the participants via express

mail (DHL) together with relevant information on the ILC

29 March 2016: laboratories submitted their results and answered a second

questionnaire with information on the air filter measurements

7-8 April 2016: workshop organized for the participants and the preliminary

results presented

20 September 2016: preliminary results were officially sent to participants

All registered communication with the participants related to this ILC can be found in the *Annexes* of the present report.

#### 3. The Reference value

## 3.1 Standardisation of the spike solution

For the present ILC exercise three radionuclides had been chosen as measurands, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. These three could be considered the major radionuclides to monitor following a nuclear accident, like that in Chernobyl. The determination of <sup>137</sup>Cs is simple and straightforward, as it needs no specific corrections. The <sup>134</sup>Cs had been selected for its significant coincidence summing effect, which calls for appropriate corrections. Finally, <sup>131</sup>I is interesting as it is seldom included in ILCs. It is short lived, easy to measure and it does not present significant coincidence summing effect. Despite its pronounced volatility, it had been chosen to be included in this exercise and it will check for proper application of the decay correction, the correction for decay during measurement and the proper handling of he air filter to avoid loss of the volatile iodine.

An additional difficulty lies in the low to very low activity levels (close to detection limit) and the possible deviation from the routine counting geometry and thus counting efficiency due to the imperfect spiking homogeneity of the filters with the radioactive solution.

The standardisation of the solutions of the three radionuclides used for spiking of the air filters was performed at the JRC-Geel by liquid scintillation counting (LSC). Gamma-ray spectrometry and counting with an ionization chamber were used as secondary methods and as an additional link to older traceable standard solutions.

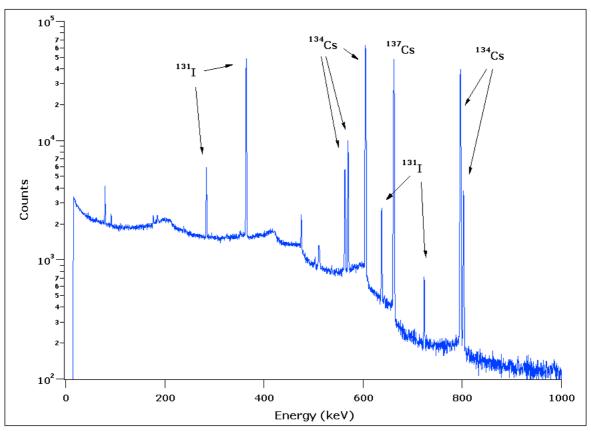
For the LSC, the efficiency tracing method developed by CIEMAT/NIST (Grau Malonda and Garcia-Toraño, 1982) was used. The principle of the CIEMAT/NIST efficiency tracing method is a combination of theoretical calculations of the radionuclide beta particle counting efficiency and an experimental determination of correction factors with the help of a tracer radionuclide, <sup>3</sup>H in this case. For the CIEMAT/NIST efficiency tracer method, the computer code CN2005 (Günther, 2002) was employed to calculate the radionuclide counting efficiencies. As tracer, the IRMM tritiated water standard was used (Spernol and Denecke, 1964; Makepeace et al, 1998).

All sources were prepared gravimetrically using a Mettler AX26 (Mettler-Toledo, Greifensee, Switzerland) mass comparator, calibrated using traceable weights. To prepare the LSC sources, aliquots of the radioactive solution were gravimetrically dispensed using the pycnometer method (Sibbens and Altzitzoglou, 2007; Campion, 1975) into 20-mL low-potassium glass LSC vials containing 15 mL of UltimaGold® (PerkinElmer, Boston, MA, USA) liquid scintillation (LS) cocktail, mixed with 1 mL of deionized water. In a similar way, for the gamma-ray spectrometric measurements, the radioactive solution was dispensed by means of a pycnometer onto a plastic foil supported by a 34-mm thin stainless steel annulus and covered by another plastic foil after drying completely.

The LSC sources were measured using a Packard 3100 Tri-Carb TR/AB (PerkinElmer, Boston, MA, USA) liquid scintillation counter and a Wallac Quantulus 1220 (PerkinElmer, Boston, MA, USA) LSC. For the gamma-ray measurements, two high-purity germanium (HPGe) detector systems were used, one with a 36% relative efficiency co-axial detector (Detector A) and the other with a 92% low-background co-axial detector (Detector B) (Canberra Industries, Inc., Meriden, CT, USA). The first

detector was housed in a 10-cm thick Pb shield of circular cross-section, lined with 1 mm Cd and 1 mm Cu; the inner 2 cm of the Pb shield was made of high radiopurity Pb. The latter was housed in a 5-cm thick Pb shield of square cross-section. Both detectors were connected to commercial analogue electronics.

The gamma-ray spectra analysis was performed using the GammaVision-32 software program (ORTEC, Oak Ridge, TN, USA) and the data analysis for both the gamma-ray analysis and the LSC analysis was done with custom made spreadsheets. *Figure 2* shows a typical gamma-ray spectrum of all three radionuclides of interest.



**Fig. 2**. Typical gamma-ray spectrum of a mixture of the three radionuclides of interest, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. The most abundant peaks are indicated.

#### 3.1.1 137Cs

Seven sources were gravimetrically prepared from the first dilution B1 of the original mother solution (A1) of <sup>137</sup>Cs and 4 from the second dilution (C1). The amount of radioactive solution in each source ranged from 12 to 45 mg for B1 and from 35 to 57 mg for C1. All sources were measured 17 times (for 20 minutes each per run) using the Packard LSC and 6 times (for 15 minutes each per run) using the Quantulus LSC over a period of one and a half month. The search for possible impurities and especially for <sup>134</sup>Cs by high resolution gamma ray spectrometry was negative.

The activity concentration of the mother solution was found to be 3.50 (3) MBq g<sup>-1</sup> on the reference date 1 March 2016 0:00 UTC. As usually, the numbers in parentheses are the numerical values of the combined standard uncertainties  $u_c$  expressed in the unit of the quoted result.

Traceability was guaranteed by using the exact same method as for participation at the ongoing comparison BIPM.RI(II)-K1.Cs-137 (Ratel et al., 2005a), following the standardisation of <sup>137</sup>Cs for the EC ILC organised in 2003; the standardised solution of

 $^{137}$ Cs of the 2003 campaign was submitted together with the results of the described standardisation at JRC-Geel 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 of the spiking solutions and subsequently of the reference values of spiked activities on filters. The results of the measurements in the SIR at BIPM, are confirming traceability. The degree of equivalence for our laboratory with respect to the reference value ( $D_i$ =( $x_i$ - $x_{Ref}$ )) was -0.19 MBq with an uncertainty of 0.32 MBq, which means that the JRC result was 0.73% lower than the reference value with an uncertainty on this difference of 1.16% (k=2).

In addition, 3 sources were prepared with 9 to 36 mg from the dilution B1 and 2 with 11 and 43 mg from the dilution C1 for gamma-ray spectrometry. The sources were measured at two different distances from the detectors, a close one and a farther one, against similar sources prepared during the 2003 EC ILC campaign and the 2014 EC ILC campaign and the discrepancy was determined to be less than 1%.

### 3.1.2 134Cs

Seven sources were gravimetrically prepared from the first dilution B1 of the original mother solution (A1) of <sup>134</sup>Cs and 4 from the second dilution (C1). The amount of radioactive solution in each source ranged from 10 to 45 mg for B1 and from 20 to 54 mg for C1. All sources were measured 31 times (for 20 minutes each per run) using the Packard LSC and 6 times (for 15 minutes each per run) using the Quantulus LSC over a period of one and a half month. No impurities were detected in the solution by high resolution gamma-ray measurements checking for possible impurities and especially for <sup>137</sup>Cs.

The activity concentration of the mother solution was found to be 35.5 (4) kBq g<sup>-1</sup> on the reference date 1 March 2016 0:00 UTC. As usually, the numbers in parentheses are the numerical values of the combined standard uncertainties  $u_c$  expressed in the unit of the quoted result.

Traceability was guaranteed by using the exact same method as for participation at the ongoing comparison BIPM.RI(II)-K1.Cs-134 (Ratel et al., 2005b; Ratel et al., 2007), following the standardisation of  $^{134}\text{Cs}$  in 2003; the standardised solution of  $^{134}\text{Cs}$  of 2003 was submitted together with the results of the described standardisation at JRC-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 of the spiking solutions and subsequently of the reference values of spiked activities on filters. The degree of equivalence for our laboratory with respect to the reference value ( $D_i = (x_i - x_{Ref})$ ) was -69 kBq with an uncertainty of 75 kBq, which means that the JRC result was 0.68% lower than the reference value with an uncertainty on this difference of 0.74% (k = 2).

In addition, 3 sources were prepared with 11 to 39 mg from the dilution B1 and 2 with 32 and 41 mg from the dilution C1 for gamma-ray spectrometry. The sources were measured at two different distances from the detectors, a close one and a farther one, against similar sources prepared during the 2003 EC ILC campaign and the discrepancy was determined to be less than 1%. Additional measurements using an ionisation chamber gave results higher by 0.7% compared to those obtained by LSC and gamma-ray spectrometry.

### 3.1.3 131I

Ten sources were gravimetrically prepared from the original mother solution (A1) of  $^{131}$ I with an amount of radioactive solution in each source ranging from 13 to 55 mg. All sources were measured 16 times (for 20 minutes each per run) using the Packard

LSC and 4 times (for 10 minutes each per run) using the Quantulus LSC over a period of one and a half month.

The activity concentration of the mother solution was found to be 6.12 (6) kBq g<sup>-1</sup> on the reference date 1 March 2016 0:00 UTC. As usually, the numbers in parentheses are the numerical values of the combined standard uncertainties  $u_c$  expressed in the unit of the quoted result.

A further confirmation of the JRC result was obtained by gamma-ray spectrometry measurements. Three point sources were prepared with 23 to 43 mg from the mother solution A1 for gamma-ray spectrometry. The sources were measured at two different distances from the detectors, a close one and a further one. The results agree well with the activity concentration result obtained by LSC within the corresponding uncertainties, as the discrepancy was determined to be less than 1.5%. It is worth noticing that the activity concentration on the certificate by the Czech Metrology Institute (CMI), supplier of the material, was 202.4(2) kBq g<sup>-1</sup> on 20-01-2016, or 4.3% lower than that measured at JRC-Geel.

#### 3.2 Dilutions

In order to approximate the activity level measured at each participating laboratory under routine conditions with the appropriate amount of spiked <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I on the filters, five different diluted solutions (E1, E2A, E3, E4, E5) were prepared following the preparation scheme shown in *Figure 3*. The standardised mother solution

**Table 2**. Dilutions and dilution factors with their combined standard uncertainties  $u_c$  (in parentheses).

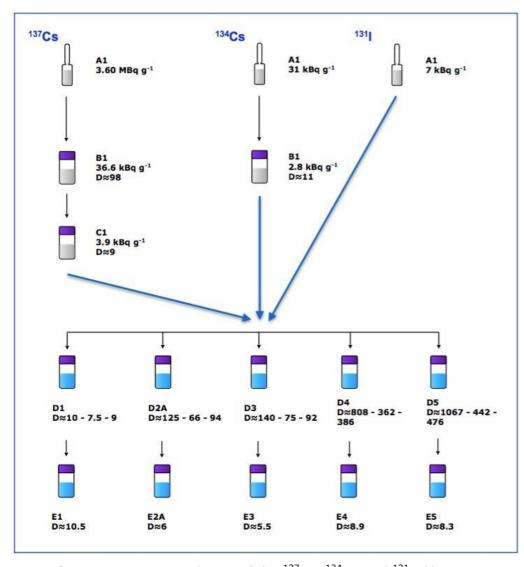
			Dilution factor D	
Solution	Code	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I
Mother solution	A1	1 1		1
Dilution	B1	98.24 (2)	1	1
Dilution	C1	887.7 (2)	10.0218 (9)	1
Dilution	D1	9092 (2)	75.368 (7)	9.1542 (3)
Dilution	D2A	11111 (6) x10 <sup>1</sup>	657.3 (2)	93.91 (3)
Dilution	D3	12500 (7) x10 <sup>1</sup>	751.2 (2)	92.08 (3)
Dilution	D4	7171 (7) x10 <sup>2</sup>	3633 (2)	386.6 (2)
Dilution	D5	947 (1) x10 <sup>3</sup>	4432 (2)	476.4 (3)
Dilution	E1	9571 (2) x10 <sup>1</sup>	793.5 (1)	96.37 (1)
Dilution	E2A	8969 (5) x10 <sup>2</sup>	5306 (2)	758.1 (3)
Dilution	E3	7519 (4) x10 <sup>2</sup>	4519 (1)	553.9 (2)
Dilution	E4	3990 (4) x10 <sup>3</sup>	20215 (8)	2151 (1)
Dilution	E5	842 (1) x10 <sup>4</sup>	3939 (2) x10 <sup>1</sup>	4233 (2)

(A1) of  $^{137}\text{Cs}$  was first diluted via two intermediate dilutions (B1 and C1). The diluent was a solution of 50  $\mu g$  mL $^{-1}$  Cs $^+$  (as CsCl) in 0.1M HCl. The standardised mother solution (A1) of  $^{134}\text{Cs}$  was first diluted via one intermediate dilution (B1). The diluent was equally a solution of 50  $\mu g$  mL $^{-1}$  Cs $^+$  (as CsCl) in 0.1M HCl.

Then, aliquots from the C1 dilution of  $^{137}$ Cs, from the B1 of  $^{134}$ Cs and the mother solution (A1) of  $^{131}$ I were gravimetrically added to 5 vials containing given amounts of a solution of 25 mg g $^{-1}$  KI and 25 mg g $^{-1}$  Na $_2$ S $_2$ O $_3$  to produce the dilutions D1, D2A, D3, D4 and D5. Therefore, the D series dilutions contain all three radionuclides at various levels. Furthemore, methylene blue, used to visualise the spikes on the filters, was gravimetrically added to each of the dilutions.

Finally, the D series dilutions were further diluted by adding amounts of a solution containing 25 mg  $g^{-1}$  KI and 25 mg  $g^{-1}$  Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub> to produce the dilutions E1, E2A, E3, E4 and E5. Those final dilutions were used to spike the air filters.

Table 2 lists the solutions and dilution factors including the addition of methylene blue. The numbers in parentheses are the numerical values of the combined standard uncertainties  $u_c$  expressed in the unit of the quoted result. In *Figure 3* the dilution scheme is shown schematically together with the approximate dilution factors and the activity concentration of each of the solutions prepared.



**Fig. 3**. Preparation scheme of the <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I dilutions.

It should be noted that all dilutions were prepared gravimetrically for utmost traceability and small uncertainty on the dilution factors. In addition, quantitative sources were prepared from all dilutions for quality control by both liquid scintillation counting and gamma-ray spectrometry. The results of these measurements for the last two dilution steps D and E are presented in *Annex 8* and confirm the gravimetrical dilution factors, with the exception of <sup>131</sup>I, which will be discussed later. The scatter of the results is large and it is due to counting statistics; many sources had to be measured in short time. In addition no optimization was done for the counting efficiency for the filter geometry.

## 3.3 Spiking of the air filters

On the basis of the information provided by the participating laboratories in the relevant questionnaire, the <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I activity per filter and the spiking pattern was determined for each filter individually. The amount of the radioactivity 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 activities below detection limit, an activity higher than the declared detection limit was distributed.

The JRC Radionuclide Metrology Sector prepared 67 air filters (*Annex 6*) by depositing on each of them gravimetrically an amount from one of the standard solutions E1, E2A, E3, E4 and E5, containing <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. Each participating laboratory in this ILC exercise had sent two blank filters of the type it is routinely using and after spiking one of them at JRC with the <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I solution, the filters were returned to the participants in order to be measured according to their routine procedure. The second blank filter was kept in reserve. The types, materials, dimensions and shapes of the air filters are given in *Annex 5*.

Since a uniform distribution of spikes was not feasible, depending on the filter size about 10 to 100 droplets were dispensed gravimetrically using a pycnometer, in a more or less symmetric pattern on the air filter. In any case, the spiked spots could easily be distinguished due to their methylene blue color, which allowed accounting for their discrete distribution when preparing the filters for measurement or when calculating counting efficiency corrections.

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 substance falling off the filter 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.

Various spiked filters are shown in *Annex 7*. 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.

#### 3.4 Reference values

The reference activity values, i.e. the spiked activities on the filters, were calculated using the activity concentration of the mother solution determined by primary standardisation and the gravimetrically determined dilution factors (*Table 2*) of the spiking standard solutions E1, E2A, E3, E4 and E5. The mass and activity of the dispensed solution on each filter was determined once per filter by weighing the

pycnometer before and after depositing the total number of drops on the corresponding filter. Tables 5, 10 and 14 show the reference values  $A_0$  for the deposited  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I activity, respectively, 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 spiked aliquots. The lowest activity of  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I spiked on an air filter was 0.2, 0.4 and 0.6 Bq respectively and the highest values were 5.0, 6.1 and 8.7 Bq, respectively. The reference date for reporting the activity (and likewise of the reference values) is 1 March 2016 0:00 UTC. The numbers in parenthesis are the expanded uncertainties U with coverage factor k=1.

## 3.5 Quality control measurements

For the purpose of quality control, sources were prepared at each stage of the preparation of the dilutions and the spiking of the filters. Especially for the filter spiking control, 15 additional filters (Whatman 42 ashless ø70 mm) and 7 point sources were produced. *Table A-6* in *Annex 6* lists in sequence all filter samples prepared for the participating laboratories as well as all control samples. It shows clearly, how well the quality control samples interleave the rest of the samples prepared.

The 15 spiked control filters and the 7 point sources were measured by gamma-ray spectrometry at the JRC-Geel, using the two HPGe detectors mentioned in *Section 3.1*. Some of the sources were measured by both detectors. The point sources were measured at a close distance (2 mm) from the detector window and the counting efficiency calibration was performed with similar standard point sources. The filters were measured in their protective plastic bags and placed directly on the detector window. The acquisition time ranged from 4 to 64 hours. No attempt was made for further geometry corrections, as the results were treated in a relative way, just to confirm the integrity of the dilutions. Due to the large number of quality control sources to measure, the low spiked activity and the limited time, the counting statistics were not optimal. In conclusion, the measurement results confirm the activity concentrations determined from the gravimetric dilution and spiking procedure for <sup>137</sup>Cs and <sup>134</sup>Cs. However, for <sup>131</sup>I are not satisfactory and they will be discussed later.

The measured activity, its ratio to the reference value and its relative deviation from the reference activity, calculated from the mass of the spiked solution and its activity concentration and expressed in percent, are given for the quality control filters in *Tables A-7a* and *A-7b*, while those for the point sources in *Tables A-8a* and *A-8b* of *Annex 8*. The uncertainty is the expanded uncertainty combining both the uncertainty on the measured and reference values. All results are calculated for the reference date of 1 March 2016 0:00 UTC. *Figure A-1 of Annex 8* shows the ratio of the measured to the reference activity for the quality control filters and for all three radionuclides. *Figure A-2* shows the corresponding ratio for the point sources.

## 3.6 Influence of discrete spiked activity distribution on the counting efficiency

The activity distribution collected by air aspiration on air filters is presumably uniform (in the sense of continuous) and assumed mostly homogeneous, although exceptions may exist. The spiked air filters prepared for the present ILC campaign have been prepared by discretely depositing aliquots of a standard radioactive solution on the filters. Therefore, the activity distribution on the spiked filters is discrete and symmetric, but not continuous. When considering a small part of the filter, one can thus claim that the distribution is inhomogeneous. The more drops deposited, the

closer to a homogeneous distribution. This inhomogeneity is expected to affect the counting efficiency, but there are more parameters influencing the counting efficiency, which we will make an attempt to quantify.

A second parameter is the size of the deposited drops. Although the deposited solution mass, and thus activity, per drop is rather constant, depending on the quality and material composition of each filter the drop spreads to a smaller or larger area.

A third parameter for the determination of the spiked activity on the air filters is the counting geometry. For the smaller filters the usual geometry is placing the filter in front of the detector, either on the detector window or at a certain distance from the detector window. For larger filters the usual way is to fold in a suitable manner the filter, or even press it to form a pellet and then place it in front of the detector. Unless the folding brings for some specific reason most of the active spots closer or further from the detector, it is expected that folding the filters distributes uniformly the active spots and consequently improves the homogeneity of the activity distribution.

A fourth parameter could be the size (active diameter) of the counting detector, giving an advantage to larger detectors, as in that case, because of the subtended solid angle, the active area of the filter is better covered by the detector.

In order to study the influence of those parameters on the counting efficiency for the measurement of the spiked air filters, a number of Monte Carlo simulations have been performed using the GEOLEP proprietary computer code (Solé, 1990; Lépy et al., 2010).

The counting efficiency has been calculated for three different HPGe detectors (of different Ge crystal sizes) and for 3 different filter sizes spiked in different patterns and with different number (9 or 16) and size of active spots (0.3, 0.7 and 1.0 mm in diameter). The size of the detectors modelled was  $\emptyset 58.5 \times 53.5$  mm (Detector A),  $\emptyset 77 \times 78$  mm (Detector B) and  $\emptyset 80 \times 30$  mm (Detector C). The dimensions, matrix composition and density of the air filters, as well as the dimensions of the detectors have been used as model inputs to the Monte Carlo code. The simulations assumed that the gamma-ray emissions were isotropic and uncorrelated. The uncertainty of the Monte Carlo simulations was in all cases better than 1%.

The filters in *Figure 4* depict the different cases which have been modelled and *Tables 3a* and *3b* give more details, as well as the results of the Monte Carlo simulations. The last column in the tables gives the percent deviation of the calculated counting efficiency for the spiked air filters from that of an air filter of the same size but with a homogeneous activity distribution.

From *Tables 3a* and 3*b* and the simulated counting efficiency results it is shown that for active filter areas smaller than or equal to the detector active areas, the discrepancies of the counting efficiency between discrete and homogeneous activity distribution is not large and, in the studied cases, below 5%.

However, the worst case scenario was that of Detector C and the larger filter size of 130-mm diameter and a 110-mm diameter active area, which resulted in an overestimation of the efficiency by 35.6% or an underestimation of the activity by the same percentage, most probably because the active spots are placed closer to the center of the detector, whereas on a homogeneous exposed filter the active area on the filter extends further from the center of the detector. If more spots are added towards the circumference (red spots in *Figure 4c*) and the calculations are repeated, the underestimation of the activity is reduced to 10.7%. If we compare the efficiency result to that of a 100-mm diameter active area (more realistic as the spots do not reach the edge of the 100-mm circle) then we will find it only 0.6% lower than that for the homogeneous source.

From these calculations, it can be concluded that the most critical parameter to simulate an exposed air filter, is not the quasi-homogeneous distribution of the active

spots, or the size of the spots, or their number. Instead, it is crucial to cover with active spots the entire area, which is normally loaded with activity in a real exposed filter. Activity deposited more to the center of the filter will increase the apparent counting efficiency and give higher activity results (if the efficiency is not appropriately corrected). Too much activity placed closer to the circumference will lower the apparent counting efficiency and give lower activity results.

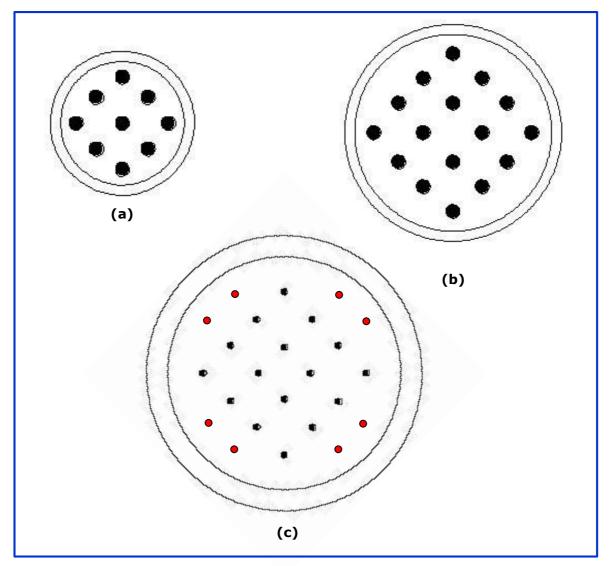


Fig. 4. Different cases of spiked air filters, which have been modelled for Monte Carlo simulations to calculate counting efficiencies with some typical detectors.

a) Ø70-mm filter with 9 active spots
b) Ø110-mm filter with 16 active spots and
c) Ø130-mm filter with 16 active spots and (including the red dots) 24 active spots

**Table 3a**. Deviation of the counting efficiency of a spiked filter from that of a filter with homogeneous distribution of the activity as calculated using the Monte Carlo code GEOLEP (see text). Calculations are performed for two different detectors (A and B), two different sizes of the homogeneous active area (ø50 and 60 mm) and three different diameters of the 9 active spots (ø3, 7 and 10 mm).

Detector and Ge crystal size (dia.xheight in mm)	Measure- ment geometry	Filter diameter (mm)	Homoge- neous active area diameter (mm)	Number of active spots on the spiked filter	Diameter of active spots on the spiked filter (mm)	Efficiency deviation from the homogeneous distribution (%)
					3	-4.27
			50	9	7	-4.50
	on detector	70			10	-5.09
	window	/0			3	6.62
			60	9	7	6.38
Detector A					10	5.71
ø58.5 x 53.5					3	-2.92
	at 10	70	50	9	7	-3.16
					10	-3.63
	mm				3	4.67
			60	9	7	4.42
					10	3.91
					3	-2.54
			50	9	7	-2.67
	on detector	70			10	-2.98
	window	/0			3	4.01
			60	9	7	3.88
Detector B					10	3.54
ø77 x 78					3	-1.96
			50	9	7	-2.13
	at 10	70			10	-2.25
	mm	/0			3	2.86
			60	9	7	2.68
					10	2.56

**Table 3b.** Deviation of the counting efficiency of a spiked filter from that of a filter with homogeneous distribution of the activity as calculated using the Monte Carlo code GEOLEP (see text). Calculations are performed for two different detectors (A and B), two different sizes of the homogeneous active area ( $\emptyset$ 90 and 110 mm) and three different diameters of the 16 active spots ( $\emptyset$ 3, 7 and 10 mm). A third detector (C) was introduced as well and the counting efficiency for filters with homogeneous active area of  $\emptyset$ 110 mm and  $\emptyset$ 100 mm was simulated for 16 and 24 active spots (see *Figure 4c*).

Detector and Ge crystal size (dia.xheight in mm)	Measure- ment geometry	Filter diameter (mm)	Homoge- neous active area diameter (mm)	Number of active spots on the spiked filter	Diameter of active spots on the spiked filter (mm)	Efficiency deviation from the homogeneous distribution (%)
					3	-5.63
			90	16	7	-5.83
	on detector	110			10	-5.82
	window	110			3	4.24
			100	16	7	4.02
Detector A					10	4.03
ø58.5 x 53.5	at 10 mm				3	-4.53
			90	16	7	-4.61
		110			10	-4.52
		110	100	16	3	3.94
					7	3.86
					10	3.96
		110			3	-5.55
			90	16	7	-5.66
	on				10	-5.55
	detector window	110			3	4.94
			100	16	7	4.82
Detector B					10	4.95
ø77 x 78					3	-3.82
			90	16	7	-3.89
	at 10	110			10	-3.82
	mm	110			3	4.08
			100	16	7	4.01
					10	4.08
Detector C	on		110	16	15	35.6
	detector	130	110	24	15	10.7
ø80 x 30	window		100	24	15	-0.6

## 4. Questionnaire on sampling method, air filter used and measurement conditions

In the preparative phase of the exercise the participants were asked to fill in a questionnaire (*Annex 3*) in order to receive air filters spiked with activity levels similar to those which the laboratories routinely measure and to allow them to measure following their usual procedure and measurement geometry. The questionnaire focused on two main fields: (1) general information and (2) measurement details. The questionnaire, besides general information, required information on the sampling method, the air filter used and the measurement conditions. The compilation of the answers provided by the participants to the questionnaire is described in this chapter and in *Annex 4*.

The first part of the questionnaire collected general information about the organisations and laboratories. The questions were grouped into four major topics: 1) contact details, 2) identity of laboratory, 3) accreditation and 4) previous participation in EC ILCs. On the basis of the answers to the question "What is the type of your laboratory?" it could be concluded that almost all (63 out of 67) participating laboratories monitor radioactivity in the environment routinely. Regarding accreditation, two thirds of the laboratories are accredited or certified primarily for gamma-ray spectrometry measurements and one fifth of the laboratories are authorised by the government or responsible body for radioactivity measurements in the environment. Furthermore, most of the participants have already had experience participating in EC ILCs; 57 laboratories had participated in the 2014 EC ILC and 26 in both the 2003 and 2014 EC ILCs.

The second part of the questionnaire contained questions related to the technical part of the measurements of radionuclides in air filter. Information on the air filter type used, the air sampling methods and the radioactivity measurement was gathered. As expected, air filters of various types and sizes are employed by the participating laboratories (*Annex 5*). The most popular air filter materials were glass fibre, nitrocellulose and polypropylene. According to size, they could be grouped in three categories. On the basis of the second part of the questionnaire, it could be concluded that all of the laboratories determine the radionuclides in air filters routinely, but there are no harmonised protocols. No correlation was found between the type and size of air filters, the sampling period, the total volume of air sampled per filter and the sampling frequency, except the fact that if the air filter size is larger, then the total volume of air sampled per filter is usually higher as well. The sampling frequency can be on a daily, weekly, monthly or annual basis or only occasionally.

Finally, in order to decide on the spiking of the individual filters, information on the minimum detectable activity (*MDA*), the typical activity level of <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I measured per filter and any particular wish for a given spiking pattern or packaging were collected in the second part of the questionnaire. The <sup>137</sup>Cs activity per filter (or bunch of filters) measured routinely by the participating laboratories varied from 0.0001 to a few Bq with a mean of 9.62 Bq and a median of 0.10 Bq, but in most cases it is below their detection limit. Similarly, the <sup>134</sup>Cs activity per filter (or bunch of filters) measured routinely by the participating laboratories varied from 0.0001 to a few Bq with a mean of 12.30 Bq and a median of 0.03 Bq. For the <sup>131</sup>I activity, the participants reported activities from 0.004 to a few Bq with a mean of 50 Bq and a median of 0.04 Bq. As many of the values reported were elevated, it can be that mistakenly detection limits were reported instead. Additionally, besides the commonly measured radionuclides, several natural and artificial radionuclides are routinely determined by the laboratories, as indicated in *Table 4*.

**Table 4.** Radionuclides measured by the participating laboratories.

Nuclide	Number of laboratories reported measuring
<sup>7</sup> Be	60
<sup>137</sup> Cs	58
<sup>131</sup> I	49
<sup>40</sup> K	47
<sup>134</sup> Cs	46
<sup>210</sup> Pb	42
<sup>226</sup> Ra	10
<sup>214</sup> Pb, <sup>214</sup> Bi	8
<sup>212</sup> Pb, <sup>228</sup> Ac	7
<sup>90</sup> Sr, <sup>60</sup> Co	5
<sup>212</sup> Bi, <sup>238</sup> U, <sup>228</sup> Ra	4
<sup>106</sup> Ru, <sup>125</sup> Sb, <sup>208</sup> Tl, <sup>243</sup> Am, Gross alpha/beta	2
<sup>22</sup> Na, <sup>75</sup> Se, <sup>95</sup> Nb, <sup>95</sup> Zr, <sup>103</sup> Pu, <sup>141</sup> Ce, <sup>144</sup> Ce, <sup>192</sup> Ir, <sup>232</sup> Th, <sup>234</sup> Th, <sup>238</sup> Pu, <sup>239</sup> Pu, <sup>240</sup> Pu, NORM	1

# 5. Questionnaire on analytical and measurement procedures

The descriptions of the analytical and measurement procedures applied by the participants were collected by means of a questionnaire together with the reporting of the results (*Annex 9*). This questionnaire was divided into three parts: (1) sample treatment, (2) equipment used and (3) measurement and data evaluation including uncertainty budget. The evaluation of the answers provided by the participants to the questionnaire is described in this chapter and in *Annex 10*.

Most of the laboratories (62 or 92.5% of the total) reported that they followed their routine procedures for the determination of activity in the spiked air filter. Three laboratories (Laboratories 19, 22 and 65) mentioned that they usually measure a stack of several (4-6) exposed filters in their routine procedure, one (Laboratory 57) mentioned that folded the spiked filter instead of cutting it and another laboratory (Laboratory 25) mentioned that they used software to correct for the difference in active area from their usual geometry.

It was advised to the participants to measure also the inner protective plastic bag, either together with the spiked air filter or separately, to ensure that no activity from the filter was left in the bag. That proved to be the case for <sup>131</sup>I and it is explained later. Measuring the plastic bag is not straightforward, though, especially those of the large filters. Usually, it is not easy to fit the air filters together with the plastic bag into the routinely used geometry or the geometry of the spiked air filter was not the same as their routine samples. Thirty three laboratories (49%) measured the protective

plastic bag together with the spiked air filter, 26 (39%) measured them separately and 8 (12%) did not measure the plastic bag at all.

The filter preparation for the measurement in the participating laboratories varied from no particular preparation at all, to pressing/compressing the spiked air filters, folding, packing together with blank air filters or the combination of the previous steps.

The majority of the laboratories used high-purity germanium (HPGe) detectors to measure the spiked air filters, of which 79% were using digital signal processing and 21% analogue electronics. Six participants (Laboratories 8, 13, 15, 22, 28 and 54) used self-developed software for the peak area determination and data evaluation, while the rest used commercially available software. For the determination of the counting peak efficiency of the detectors almost half of the laboratories (31 laboratories or 46%) used reference air filters made by spiking single or multiple-nuclide standard solutions onto blank filters. Software was used by 15 laboratories (22%) to determine the counting efficiency by computer simulation. Nineteen laboratires (28%) used a combination of a multigamma standard source and appropriate software. Two laboratories, probably erroneously, reported that no efficiency calibration was performed.

The laboratories measured the sources for a minimum of 1 hour and up to 233 hours (mean 53 h, median 47.9 h) as shown in *Table A-10* of *Annex 10*. Respectively, the background was measured for a period from 15 to 333 hours. For the determination of the *MDA*, the laboratories used commercially available software and/or different calculation methods based on the Currie method, ISO 11929/2010, Risø method, DIN 25482 or other methods.

## 6. Evaluation and comparison of the results

### 6.1 Identification of outliers and normal distribution test

In order to evaluate the performance of the laboratories, their measurement results need to be compared to the individual reference activity value, i.e. the spiked activity on each filter. The individual reported activities were normalised to their assigned reference spiked activity by calculating the measured/reported-to-reference activity ratio.  $u_c$  (k=1) was determined according to the following equation:

$$u_{c}(A/A_{0}) = \sqrt{\left[\frac{u_{c}(A_{0})}{A_{0}}\right]^{2} + \left[\frac{u_{c}(A)}{A}\right]^{2}}$$
(1)

where

A is the activity value reported by the participating laboratory  $u_c(A)$  is the standard uncertainty of a participant's result (k=1) A<sub>0</sub> is the assigned activity reference value  $u_c(A_0)$  is the standard uncertainty of the assigned reference value (k=1)

 $u_c(A/A_0)$  is the standard uncertainty of the ratio  $A/A_0$  (k=1)

Due to the relatively small uncertainty of the reference values, the uncertainty of the activity ratios measured versus spiked is dominated by the uncertainty of the measured values.

The limit of acceptable precision was set to  $\pm 20\%$  deviation from the reference value. A more tolerant  $\pm 33\%$  limit was also included for backwards compatibility with the previous ILCs of 2014 and 2003. The 20% value was chosen as it was assumed that

gamma-ray spectrometry – even with the low activity deposited by spiking on the filters using a technique which cannot provide as homogeneous distribution of the activity on the surface as the normal air sampling – can easily be performed within such uncertainty.

The presence of statistical outliers among the reported results was investigated using the Grubbs' test at a level of significance  $\alpha=1\%$  and  $\alpha=5\%$ , as suggested in ISO/IEC 5725-2 (ISO, 1994). Statistical analysis of the results was carried out for the different working dilutions (E1 to E5) and for the ratio of the measured-to-reference radionuclide activity, separately. The reference value of the working dilutions is based on the standardised reference value of the mother solution and the gravimetrically determined dilution factors. The laboratory values used for the statistical analysis were calculated for each laboratory according to the following formula and grouped on the basis of the working dilution used (E1 to E5):

$$C = \frac{A}{m} \tag{2}$$

where

A is the activity value reported by the participating laboratory (Bq)

m is the mass of the dilution spiked by JRC on the surface of the air filter (g)

Moreover, the distribution of the data was tested using the normal probability plot and the frequency histogram.

The z values are calculated according to the ISO 13528:2015(E), (ISO, 2015) in the following way:

$$z = \frac{(\frac{A}{A_0} - 1)}{\sigma_{nt}} \tag{3}$$

where

A is the activity value reported by the participating laboratory

 $A_0$  is the assigned activity reference value

 $\sigma_{\text{pt}}$   $\phantom{\phi_{\text{pt}}}$  is the standard deviation for proficiency assessment

#### 6.2 Scores and evaluation criteria

#### 6.2.1 Percentage difference

An alternative way of presentation albeit yielding no additional information versus the ratio, is the percentage difference (ISO, 2005a). The percentage difference from the reference activity value is calculated using the formula:

$$D_{\%} = \frac{A - A_0}{A_0} \cdot 100\% \tag{4}$$

where

A is the activity value reported by the participating laboratory

 $A_0$  is the assigned activity reference value

For the environmental radioactivity measurements the criterion of  $\pm 20\%$  difference from the reference value is mostly used.

## 6.2.2 En number

To take the expanded uncertainty of the reported results and that of the reference values into account in the analysis, a performance test using  $E_n$  numbers was applied (ISO, 2005a). The calculation of the  $E_n$  numbers was carried out according to the following formula:

$$E_n = \frac{A - A_0}{\sqrt{U(A)^2 + U(A_0)^2}}$$
(5)

where

A is the activity value reported by the participating laboratory

 $A_0$  is the assigned activity reference value

U(A) is the expanded uncertainty of a participant's result (k=2)

 $U(A_0)$  is the expanded uncertainty of the assigned reference value (k=2)

When uncertainties are estimated according to the Guide to the Expression of Uncertainty Measurement (GUM) (ISO, 2008), a measurement result with its uncertainty interval giving a level of confidence of 95% should overlap with the reference value and its expanded uncertainty. Therefore,  $E_n$  numbers are interpreted as following:

If  $|E_n| \le 1$ , the laboratory values are compatible with the reference value; If  $|E_n| > 1$ , the laboratory values differ significantly from the reference values, the sources of deviation should be investigated and corrected, "warning signal"; If  $|E_n| > 1.5$ , there is an urgent need to investigate and find the sources of the large deviation, "action signal".

### 6.2.3 Compatibility test

The reference values, the reported values, the ratios, the percentage differences and the  $E_n$  numbers are given for each laboratory and for all three radionuclides in the Tables 5, 10, 14.

For results to be compatible with the assigned reference values, they have to satisfy both the percentage difference,  $D_{\%}$  and and  $E_n$  tests. This means that the percentage difference should be withing the  $\pm 20\%$  limit from the reference value and the absolute  $E_n$  should be smaller than or equal to unity. The compatibility for each laboratory and for the three radionuclides is given in the last three columns of *Table 21*.

#### 6.2.4 PomPlot

In order to compare the results, a modern type of graph, the "PomPlot", which underlines the importance of the assigned uncertainties is applied.

The PomPlot, an intuitive graphical method, is used to produce an overview of the results (Pommé, 2016). It displays the relative deviations (D/MAD) of the individual results A from the reference value  $A_0$  on the horizontal axis and relative uncertainties (u/MAD) on the vertical axis (*Figure 5*). For both axes, the variables are expressed as multiples of MAD, which is defined as the median of absolute deviation from the reference value

$$MAD = Median|D_i|, (i = 1,...,n)$$
(1)

where  $D_i$  is the difference between the reported and the reference activity:

$$D_i = \frac{A_i}{A_0} - 1 \tag{2}$$

The median absolute deviation MAD is used because of its robustness.

For every datum point the uncertainty is calculated as an independent sum of the reported combined uncertainties on  $A_i$  and  $A_0$ 

$$u_i^2 = u_c^2(A_i) + u_c^2(A_0)$$
(3)

where  $u_c(A_i) = U(A)_i/k$  and  $u_c(A_0) = U(A_0)/k$  (4)

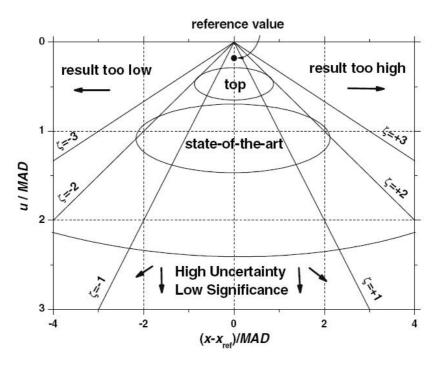


Fig. 5. Interpretation of the PomPlot (Pommé, 2016)

The  $\zeta$ -scores,  $|\zeta| = |D/u| = 1$ , 2 and 3, are represented by diagonal solid lines, creating the aspect of a pyramidal structure. The  $\zeta$ -score is a measure for the deviation between laboratory result and reference value relative to the total uncertainty (ISO, 2005a). The points on the right-hand side of the graph correspond to results that are higher than the reference value whereas lower values are situated on the left. When the uncertainty is small, the corresponding point is situated high in the graph. The most accurate results should be situated close to the top of the pyramid. Points outside of the  $\zeta=\pm 3$  lines are probably inconsistent with the reference value.

#### 7. Results

All 67 participating laboratories registered to this ILC reported valid results for all three radionuclides. Two laboratories reported shortly after the submission was closed and their results were considered valid and were retained.

The Laboratory Code numbers in this report are those communicated to each participant individually and they are confidential. They have no correlation with the laboratory codes used during the exercise.

## 7.1 <sup>137</sup>Cs

## 7.1.1 Reported results

Table 5 presents the reference activities of each individual spiked air filter, the activity results as reported by the participating laboratories, the ratio of the reported to the reference activity, the percentage difference (D%) of the reported to the reference activity and the  $E_n$  number, with their associated standard uncertainties (k=1) for the case of  $^{137}$ Cs. The Laboratory Code numbers are the confidential code numbers communicated to the participants and have no correlation with the laboratory codes used during the exercise.

**Table 5**. <sup>137</sup>Cs reference (spiked) activity, activity result as reported by the participating laboratories, ratio of the reported to the reference activity, percentage difference (D%) of the reported to the reference activity and  $E_n$  number, with their associated standard uncertainties (k=1).

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. $u_c(A)/A$ (%) $k=1$	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$ $k=1$	Percentage Difference D%	Standard uncer- tainty $u_c(A/A_0)$	<b>E</b> <sub>n</sub>
1	0.398	0.003	0.34	0.09	2	0.045	13.2	0.85	0.11	-14.5	1.9	-0.6
2	0.266	0.002	0.247	0.019	1	0.019	7.7	0.93	0.07	-7.2	0.6	-0.5
3	0.450	0.004	0.46	0.05	2	0.025	5.4	1.02	0.06	2.3	0.1	0.2
4	0.364	0.003	0.31	0.031	1	0.031	10.0	0.85	0.09	-14.9	1.5	-0.9
5	0.379	0.003	0.42	0.05	2	0.025	6.0	1.11	0.07	10.9	0.7	0.8
6	0.510	0.004	0.521	0.05	1.65	0.03	5.8	1.02	0.06	2.2	0.1	0.2
7	0.512	0.004	0.447	0.04	1	0.04	8.9	0.87	0.08	-12.6	1.1	-0.8
8	4.287	0.036	5.3	0.6	2	0.3	5.7	1.24	0.07	23.6	1.4	1.7
9	0.365	0.003	0.384	0.048	2	0.024	6.3	1.05	0.07	5.2	0.3	0.4
10	0.197	0.002	0.165	0.017	2	0.0085	5.2	0.84	0.04	-16.4	0.9	-1.9
11	0.215	0.002	0.22	0.04	2	0.02	9.1	1.02	0.09	2.2	0.2	0.1
12	2.121	0.018	1.94	0.14	1	0.14	7.2	0.91	0.07	-8.5	0.6	-0.6
13	0.339	0.003	0.35	0.028	2	0.014	4.0	1.03	0.04	3.1	0.1	0.4
14	0.341	0.003	0.3349	0.0087	1	0.0087	2.6	0.98	0.03	-1.9	0.1	-0.3
15	0.280	0.002	0.35	0.03	1	0.03	8.6	1.25	0.11	24.8	2.1	1.2
16	0.286	0.002	0.354	0.0448	2	0.0224	6.3	1.24	0.08	23.7	1.5	1.51
17	0.252	0.002	0.25	0.02	1	0.02	8.0	0.99	0.08	-0.8	0.1	0.0

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$ $k=1$	Percentage Difference D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	E <sub>n</sub>
18	0.419	0.004	0.3505	0.0297	1	0.0297	8.5	0.84	0.07	-16.3	1.4	-1.1
19	0.493	0.004	0.464	0.027	1	0.027	5.8	0.94	0.06	-5.8	0.3	-0.5
20	0.294	0.003	0.33	0.03	2	0.015	4.5	1.12	0.05	12.1	0.6	1.2
21	0.511	0.004	0.459	0.026	1	0.026	5.7	0.90	0.05	-10.1	0.6	-0.98
22	0.347	0.003	0.372	0.031	1	0.031	8.3	1.07	0.09	7.2	0.6	0.4
23	0.574	0.005	0.6	0.04	2	0.02	3.3	1.05	0.04	4.5	0.2	0.6
24	0.441	0.004	0.42	0.033	2	0.017	3.9	0.95	0.04	-4.9	0.2	-0.6
25	0.210	0.002	0.194	0.04	2	0.02	10.3	0.92	0.10	-7.6	0.8	-0.4
26	0.388	0.003	0.97	0.19	2	0.095	9.8	2.50	0.25	149.7	14.7	3.1
27	4.995	0.042	4.41	0.1	2	0.05	1.1	0.88	0.01	-11.7	0.2	-4.5
28	0.372	0.003	0.534	0.139	2	0.070	13.0	1.43	0.19	43.4	5.7	1.2
29	0.390	0.003	0.3384	0.0291	1.65	0.0176	5.2	0.87	0.05	-13.2	0.7	-1.4
30	0.390	0.003	0.464	0.013	2	0.007	1.4	1.19	0.02	19.1	0.3	5.1
31	0.299	0.003	0.277	0.016	2	0.008	2.9	0.93	0.03	-7.5	0.2	-1.3
32	0.286	0.002	0.3	0.05	2	0.025	8.3	1.05	0.09	4.9	0.4	0.3
33	0.339	0.003	0.37	0.04	2	0.02	5.4	1.09	0.06	9.0	0.5	0.8
34	0.524	0.004	0.492	0.048	1	0.048	9.8	0.94	0.09	-6.2	0.6	-0.3
35	0.485	0.004	0.596	0.244	2	0.122	20.5	1.23	0.25	22.8	4.7	0.5
36	0.499	0.004	0.521	0.038	2	0.019	3.6	1.04	0.04	4.5	0.2	0.6

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$ $k=1$	Percentage Difference D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	E <sub>n</sub>
37	0.332	0.003	0.35	0.03	2	0.015	4.3	1.05	0.05	5.4	0.2	0.6
38	0.379	0.003	0.38	0.03	1	0.03	7.9	1.00	0.08	0.2	0.0	0.0
39	0.368	0.003	0.3	0.03	1	0.03	10.0	0.82	0.08	-18.4	1.8	-1.1
40	0.461	0.004	0.57	0.05	2	0.025	4.4	1.24	0.06	23.6	1.1	2.1
41	0.364	0.003	0.373	0.01	2	0.005	1.3	1.03	0.02	2.5	0.0	0.8
42	0.370	0.003	0.296	0.017	1	0.017	5.7	0.80	0.05	-19.9	1.2	-2.1
43	0.244	0.002	0.26	0.04	2	0.02	7.7	1.07	0.08	6.8	0.5	0.4
44	0.379	0.003	0.3	0.05	2	0.025	8.3	0.79	0.07	-20.8	1.7	-1.6
45	0.670	0.006	0.705	0.026	1	0.026	3.7	1.05	0.04	5.2	0.2	0.7
46	0.343	0.003	0.3	0.05	1	0.05	16.7	0.87	0.15	-12.6	2.1	-0.4
47	0.434	0.004	0.39	0.021	1	0.021	5.4	0.90	0.05	-10.1	0.5	-1.02
48	0.388	0.003	0.43	0.02	1	0.02	4.7	1.11	0.05	10.7	0.5	1.03
49	0.354	0.003	0.4474	0.028	2	0.014	3.1	1.26	0.04	26.4	0.9	3.3
50	0.290	0.002	0.29	0.01	1	0.01	3.4	1.00	0.04	0.1	0.0	0.0
51	0.343	0.003	0.333	0.01	1	0.01	3.0	0.97	0.03	-2.8	0.1	-0.5
52	0.467	0.004	0.431	0.054	2	0.027	6.3	0.92	0.06	-7.6	0.5	-0.7
53	0.531	0.005	0.594	0.055	2	0.028	4.6	1.12	0.05	11.9	0.6	1.1
54	1.024	0.009	0.592	0.015	1	0.015	2.5	0.58	0.02	-42.2	1.1	-12.5
55	0.308	0.003	0.25	0.02	2	0.01	4.0	0.81	0.03	-18.7	0.8	-2.8

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/A (%) k=1	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$ $k=1$	Percentage Difference D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	<b>E</b> <sub>n</sub>
56	0.222	0.002	0.235	0.018	1	0.018	7.7	1.06	0.08	5.7	0.4	0.4
57	0.776	0.007	0.74	0.056	2	0.028	3.8	0.95	0.04	-4.7	0.2	-0.6
58	0.343	0.003	0.34	0.01	1	0.01	2.9	0.99	0.03	-1.0	0.0	-0.2
59	0.414	0.004	0.398	0.022	1	0.022	5.5	0.96	0.05	-3.8	0.2	-0.3
60	0.375	0.003	0.33	0.02	1	0.02	6.1	0.88	0.05	-12.0	0.7	-1.1
61	0.238	0.002	0.2697	0.0394	1	0.0394	14.6	1.13	0.17	13.1	1.9	0.4
62	0.367	0.003	0.342	0.01	1	0.01	2.9	0.93	0.03	-6.7	0.2	-1.2
63	0.394	0.003	0.29988	0.02976	2	0.01488	5.0	0.76	0.04	-24.0	1.2	-3.1
64	0.230	0.002	0.196	0.017	1	0.017	8.7	0.85	0.07	-14.8	1.3	-0.99
65	0.415	0.004	0.3986	0.023	1	0.023	5.8	0.96	0.06	-4.0	0.2	-0.4
66	0.414	0.004	0.375	0.02	1	0.02	5.3	0.90	0.05	-9.5	0.5	-0.97
67	0.476	0.004	0.456	0.023	1	0.023	5.0	0.96	0.05	-4.2	0.2	-0.4

#### 7.1.2 Evaluation

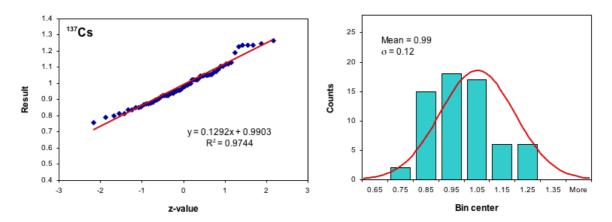
The presence of statistical outliers among the reported results was investigated using the Grubbs' test at a level of significance  $\alpha=1\%$  and  $\alpha=5\%$ , as suggested in ISO/IEC 5725-2 (ISO, 1994). Statistical analysis of the results was carried out for the different working dilutions (E1 to E5) and for the ratio of the measured-to-reference  $^{137}$ Cs activity separately. The reference value of the working dilutions is based on the standardised reference value of the mother solution and the gravimetrically determined dilution factors.

In the case of the ratio values according to the Grubb's test at 1% three results were indicated as outliers: Laboratories 26, 28 and 54. The outlying values were not discarded, but were included in further evaluations, unless it is declared differently.

Table 6 presents the statistical analysis of the results reported by the participating laboratories per dilution used for spiking the air filters. Moreover, the distribution of the data was tested using the normal probability plot and the frequency histogram. According to both of these graphs presented in *Figure 6*, the <sup>137</sup>Cs data are distributed normally and unimodally. The mean values of the reported values for dilutions E1 and E5 are slightly higher than the reference values and slightly lower for the dilutions E2A, E3 and E4. The mean of the ratio is very close to unity.

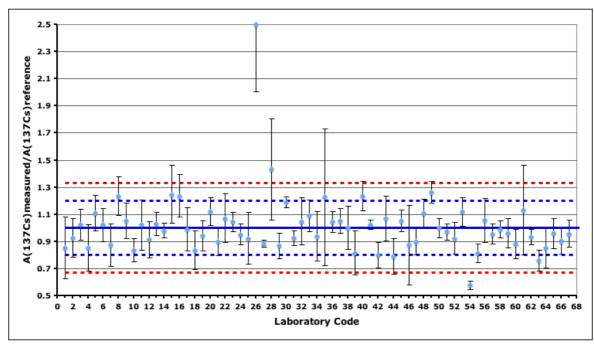
**Table 6**. Statistical analysis of the laboratory reported results for <sup>137</sup>Cs, per dilution. In the last column, the ratios of the measured to the reference values are given.

		E1	E2A	E3	E4	E5	Ratio
	Number of laboratories	2	20	4	9	32	67
	Min (Bq g <sup>-1</sup> )	32.31	2.97	2.69	0.70	0.34	0.58
All	Max (Bq g <sup>-1</sup> )	45.24	4.83	4.47	0.99	1.04	2.50
reported results	Median (Bq g <sup>-1</sup> )	38.78	3.64	4.32	0.81	0.44	0.98
	<b>Mean</b> (Bq g <sup>-1</sup> )	38.78	3.72	3.95	0.81	0.46	1.01
	Standard deviation (Bq g <sup>-1</sup> )	9.15	0.51	0.84	0.09	0.12	0.23
	Number of outliers	0	0	1	0	2	3
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	38.78	3.72	4.37	0.81	0.46	0.99
(5%)	Standard deviation (Bq g <sup>-1</sup> )	9.15	0.51	0.11	0.09	0.12	0.12
	Number of outliers	0	0	1	0	2	3
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	38.78	3.72	4.37	0.81	0.46	0.99
(1%)	Standard deviation (Bq g <sup>-1</sup> )	9.15	0.51	0.11	0.09	0.12	0.12
	Ref value (Bq g <sup>-1</sup> )	36.6	3.91	4.66	0.878	0.416	1
	Expanded unc. (Bq g <sup>-1</sup> )	0.6	0.07	0.08	0.015	0.007	0.01
	Rel. exp. unc. (%)	1.70	1.70	1.70	1.70	1.70	0.01



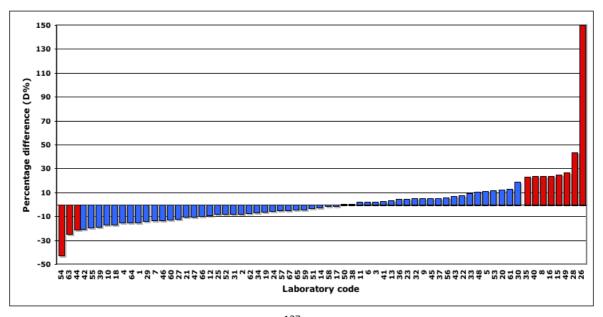
**Fig. 6**. Normal probability plot and frequency histogram of the <sup>137</sup>Cs results after exclusion of the three outliers (see text). The red curve in the frequency histogram is the normal probability distribution.

In Figure 7, the ratio of the  $^{137}$ Cs activity per spiked air filter as measured and reported by the participating laboratory over the individual spiked activity on the filter (JRC reference value) is plotted. The error bars show the expanded uncertainty (k=2). The blue dashed lines indicate the  $\pm 20\%$  limit from the JRC reference value. For backwards compatibility with previous ILCs, the  $\pm 33\%$  limit from the JRC reference value is also indicated by the red dashed lines.



**Fig. 7**. Ratio of  $^{137}$ Cs activity per spiked air filter as measured by the participating laboratory to the individual spiked activity on the filter (JRC reference value). Blue dashed lines indicate the  $\pm 20\%$  limit from the reference value and the red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2).

In Figure 8, the percentage difference, D%, of the results reported by the participants from the reference activity values are plotted in ascending order.

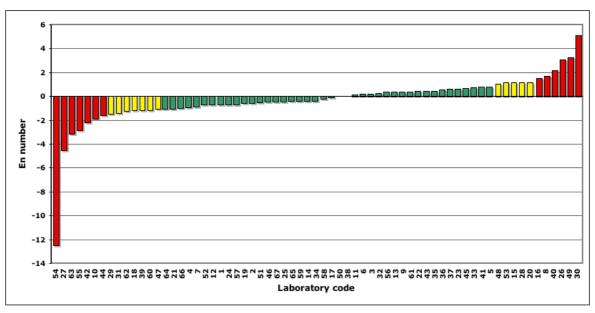


**Fig. 8**. Percentage difference of the  $^{137}$ Cs results reported by the participating laboratories from the reference activity values, plotted in ascending order. Blue colour indicates the results within the  $\pm 20\%$  range from the reference value and red indicates results outside that range.

The majority of the laboratories reported satisfactory results. From *Table 5* and *Figures 7 and 8* it can be seen that 11 laboratories of the 67 reported  $^{137}$ Cs results discrepant by more than 20% from the reference value; this represents 16% of the participating laboratories. Only three (3) laboratories reported results which were outside the more tolerant limit of 33% from the reference value. This is just 6% of the participating laboratories.

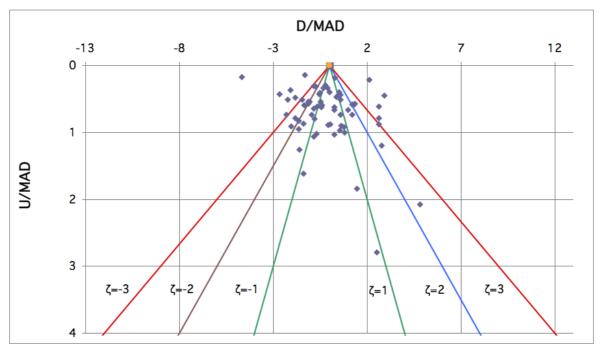
To take the expanded uncertainty of the reported results and that of the reference values into account in the analysis, a performance test using  $E_n$  numbers was applied (ISO, 2005a). The  $E_n$  numbers sorted in ascending order are graphically presented in *Figure 9*.

Under the conditions of this test, 42 (63%) out of the 67 reported results for  $^{137}$ Cs are compatible with the reference value while 25 are not. Among those 25, 12 (18%) laboratories reported results with  $1 < |E_n| < 1.5$  and 13 (19%) laboratories reported incompatible results with  $|E_n| > 1.5$ . Comparing *Figure 8* and *Figure 9*, it is obvious that results with significant deviations from the reference value are scoring badly with  $E_n$  numbers as well. However, that comparison also shows that some laboratories with an acceptably small deviation from the reference value are assigned with an incompatible  $E_n$  number, as they probably underestimated the uncertainty on their reported value.



**Fig. 9**.  $E_n$  numbers for the <sup>137</sup>Cs results, plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

Another graphical way to compare the results, which underlines the importance of the assigned uncertainties, is the "PomPlot". The theoretical description of the PomPlot can be found in *Section 6.2.4*. The PomPlot created on the basis of the reported results for  $^{137}$ Cs is presented in *Figure 10*. The many points outside the  $|\zeta|=3$ , indicate that laboratories underestimated the uncertainties.



**Fig. 10**. PomPlot of the  $^{137}$ Cs data. Green, blue and red solid lines indicate  $\zeta$ -scores=1, 2 and 3, respectively.

## 7.1.3 Parameters possibly influencing the results

Based on the results reported by the participants and the answers provided to the questionnaire,  $Table\ 7$  contains the detailed evaluation of possible influencing parameters to the measurement performance and therefore the  $D_\%$  and  $E_n$  number. There is no obvious correlation between laboratories with and without accreditation or certification and higher ratio of compatible results. However, laboratories measuring a large number (>100) of filters per year showed better performance. No strong correlation can be found between the air filter size, spiked activity level and sample preparation and the deviation of the submitted result from the reference value. In the case of laboratories which did not or could not follow their routine procedure, they all scored very well.

**Table 7.** Possible influencing parameters; statistics and categorisation of the reported results by the participating laboratories.

		Total Nr of Labs	<i>D</i> <sub>%</sub> >20 (Nr of labs)	Percen tage (%)	E <sub>n</sub> >1 (Nr of labs)	Percen tage (%)
All participants		67	11	16.4	25	37.3
Country	EU	62	10	16.1	22	35.5
Country	Non-EU	5	1	20	(Nr of labs)	60
	Accredited (ISO 17025)	30	4	13.3	12	40
	Authorised	13	1	7.7	5	38.5
Accreditation	Certified (ISO 9000)	2	0	0	0	0
	Certified (ISO 9000) and Accredited (ISO 17025)	10	3	30	3	30
<sup>137</sup> Cs	<25	9	2	22.2	5	55.6
measurements per year	25-100	21	3	14.3	9	42.9
per year	>100	37	6	16.2	11	29.7
	Small	29	5	17.2	11	37.9
Air filter size	Medium	11	2	18.2	5	45.5
	Large	27	4	14.8	9	33.3
	0-0.3	15	2	13.3	5	33.3
Cuilead	0.3-0.4	27	5	18.5	13	48.1
Spiked activity level (Bq)	0.4-0.5	13	2	15.4	3	23.1
(P4)	0.5-1	8	0	0	1	12.5
	>1	4	2	50	3	75

		Total Nr of Labs	D <sub>%</sub> >20 (Nr of labs)	Percen tage (%)	E <sub>n</sub> >1 (Nr of labs)	Percen tage (%)
Routine	Yes	62	11	17.7	25	40.3
conditions	No	5	0	0	0	0
	Together with air filter	33	5	15.2	12	36.4
Measurement of plastic bag	Plastic bag measured separately	26	5	19.2	9	34.6
	Plastic bag not measured	8	1	12.5	4	50
	No sample preparation	30	4	13.3	11	36.7
Sample	Pressing/ compressing	26	3	11.5	9	34.6
preparation	Folding	23	4	17.4	8	34.8
	Packing with blank filters	1	0	0	0	0

## 7.1.4 Comparison to the 2003 and the 2014 EC ILC

Cesium-137 was a common measurand for all three ILCs conducted in 2003, 2014 and 2016 by the JRC. This makes it possible to make statistics and draw some conclusions. *Table 8* shows the statistics for the three ILCs involving the measurement of  $^{137}$ Cs in air filters. One can observe a slight improvement over the years, especially for the number of results outside the limits for the ratio and equivalently the percentage difference; the percentage of values outside the  $\pm 20\%$  range from the reference activity dropped from 38% in 2003, to 20% in 2014 and to 16% in 2016. However, the number of laboratories with  $|E_n| > 1$  went from 25% in 2003, to 32% in 2014 and to 37% in 2016. The estimation of the uncertainty is a critical point in the case of reporting results and this is reflected in the  $E_n$  scores.

More information can be found in *Tables 9a* and *9b*. In the first, the performance of laboratories which participated in two of the ILCs, either in 2003 and 2014 or in 2014 and 2016 is shown, In the second, the performance of laboratories which participated in all three ILCs, is presented.

Thirty laboratories participated in both the 2003 and 2014 air filter EC ILCs and, since one laboratory measured 2 air filters in 2003, reported 31 results in 2003 and 30 results in 2014. Nine laboratories improved their performance since they were outliers and/or in the incompatible  $E_n$  number groups in the 2003 exercise and reported compatible values in 2014. Two laboratories, which were in action level regarding  $E_n$  number provided values in warning level, show a slightly improvement. However, 7 laboratories which reported compatible values in 2003 are outliers and/or reported results giving incompatible  $E_n$  numbers in 2014; this could be an indication that the quality of the results provided by these laboratories is not stable over time, or alternatively, are overconfident on the accuracy of their results.

Fifty seven laboratories participated in the 2014 EC ILC and the 2016 ENV57/MetroERM ILC. The number of laboratories reporting results outside the limits is quasi-identical; 5% of the laboratories reported values outside the  $\pm 33\%$  range for the ratio, 16% outside the  $\pm 20\%$  range and 12% with  $|E_n| > 1$ .

**Table 8**. Comparison of the spiked activity levels and the performance of the laboratories in the 2003 and 2014 EC ILC and the 2016 ENV57/MetroERM ILC.

	2003	2014	2016
Number of laboratories	48	76	67
	Spiked <sup>137</sup> Cs acti	ivity levels in Bq	
Minimum	0.015	0.070	0.197
Maximum	0.564	2.34	5.00
Average	0.160	0.359	0.545
Median	0.123	0.195	0.379
Nur	mber of laboratory re	esults outside the lir	nits
Ratio>±33%	6 (13%)	4 (5%)	3 (4.5%)
D <sub>%</sub> >±20%	18 (38%)	15 (20%)	11 (16%)
1< E <sub>n</sub>  <1.5	3 (6%)	11 (14%)	12 (18%)
<i>E</i> <sub>n</sub>  >1.5	9 (19%)	12 (18%)	13 (19%)

**Table 9a**. Comparison of the spiked activity levels and the performance of the laboratories in the 2003 and 2014 EC ILCs and the 2014 EC ILC and the 2016  $\pm 1.00$  ENV57/MetroERM ILC

		participated ILC		s participated 2 ILC		
	2003	2014	2014	2016		
Number of laboratories	3	0	57			
	Spiked <sup>13</sup>	<sup>7</sup> Cs activity leve	ls in Bq			
Minimum	0.029	0.083	0.073	0.197		
Maximum	0.401	2.310	2.310	5.00		
Average	0.162	0.324	0.342	0.516		
Median	0.126	0.194	0.178	0.379		
ı	Number of labor	atory results ou	tside the limits			
Ratio>±33%	2 (7%)	2 (7%)	3 (5%)	3 (5%)		
D <sub>%</sub> >±20%	6 (20%)	4 (13%)	9 (16%)	9 (16%)		
1< E <sub>n</sub>  <1.5	3 (10%)	6 (20%)	7 (12%)	7 (12%)		
$ E_n  > 1.5$	6 (20%)	2 (7%)	7 (12%)	10 (18%)		

**Table 9b**. Comparison of the spiked activity levels and the performance of the laboratories in the 2003 and 2014 EC ILC and the 2016 ENV57/MetroERM ILC.

	Labora	tories participated i	n 3 ILC
	2003	2014	2016
Number of laboratories		26	
	Spiked <sup>137</sup> Cs act	ivity levels in Bq	
Minimum	0.055	0.083	0.84
Maximum	0.404	2.31	2.50
Average	0.174	0.369	1.08
Median	0.136	0.200	1.02
Nu	mber of laboratory r	esults outside the lir	nits
Ratio>±33%	2 (8%)	2 (8%)	2 (8%)
D <sub>%</sub> >±20%	7 (27%)	5 (19%)	5 (19%)
1< E <sub>n</sub>  <1.5	1 (4%)	6 (23%)	4 (15%)
<i>E</i> <sub>n</sub>  >1.5	4 (15%)	2 (8%)	3 (12%)

Finally, 26 laboratories participated in all three ILCs. The overall performance is rather stable over the years; reported results outside the  $\pm 33\%$  range for the ratio was 8% for all ILCs, results outside the  $\pm 20\%$  range was 27% in 2003 and 19% for both 2014 and 2016. The results scoring  $|E_n|>1$  were 19% in 2003, 31% in 2014 and 27% in 2016.

It is worth mentioning that  $^{137}$ Cs is a rather easy to measure radionuclide and the laboratories are scoring consistently well for that nuclide over the years.

## 7.2 <sup>134</sup>Cs

## 7.2.1 Reported results

Table 10 presents the reference activities of each individual spike air filter, the activity results as reported by the participating laboratories, the ratio of the reported to the reference activity, the percentage difference (D%) of the reported to the reference activity and the  $E_n$  number, with their associated standard uncertainties (k=1) for the case of  $^{134}$ Cs. The Laboratory Code numbers are the confidential code numbers communicated to the participants and have no correlation with the laboratory codes used during the exercise.

**Table 10**. <sup>134</sup>Cs reference spiked activity, activity result as reported by the participating laboratories, ratio of the reported to the reference activity, percentage difference (D%) of the reported to the reference activity and  $E_n$  number, with their associated standard uncertainties (k=1).

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed <i>k</i> factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. $u_c(A)/A$ (%) $k=1$	Coinc. Sum- ming Corr.	Ratio (A/A <sub>0</sub> )	Standard uncer- tainty $u_c(A/A_0)$	Percentage Difference <b>D</b> %	Standard uncer- tainty $u_c(A/A_0)$	E <sub>n</sub>
1	0.681	0.007	0.6	0.1	2	0.05	8.3	yes	0.88	0.07	-12.0	1.0	-0.8
2	0.533	0.005	0.525	0.038	1	0.038	7.2	no	0.99	0.07	-1.4	0.1	-0.1
3	0.771	0.008	0.75	0.08	2	0.04	5.3	yes	0.97	0.05	-2.7	0.1	-0.3
4	0.790	0.008	0.75	0.022	1	0.022	2.9	no	0.95	0.03	-5.0	0.2	-0.8
5	0.821	0.008	0.89	0.07	2	0.04	3.9	yes	1.08	0.04	8.5	0.3	0.97
6	1.105	0.011	1.11	0.09	1.65	0.05	4.9	yes	1.00	0.05	0.5	0.0	0.0
7	0.877	0.009	0.73	0.044	1	0.044	6.0	yes	0.83	0.05	-16.7	1.0	-1.6
8	5.243	0.052	5.1	0.6	2	0.3	5.9	no	0.97	0.06	-2.7	0.2	-0.2
9	0.791	0.008	0.814	0.085	2	0.043	5.2	yes	1.03	0.05	2.9	0.2	0.3
10	0.395	0.004	0.344	0.032	2	0.016	4.7	yes	0.87	0.04	-12.9	0.6	-1.55
11	0.431	0.004	0.32	0.05	2	0.025	7.8	yes	0.74	0.06	-25.7	2.0	-2.2
12	3.578	0.036	3.8	0.46	1	0.46	12.1	yes	1.06	0.13	6.2	0.8	0.2
13	0.735	0.007	0.733	0.059	2	0.030	4.0	yes	1.00	0.04	-0.3	0.0	0.0
14	0.739	0.007	0.743	0.021	1	0.021	2.8	yes	1.00	0.03	0.5	0.0	0.1
15	0.608	0.006	0.68	0.05	1	0.05	7.4	no	1.12	0.08	11.9	0.9	0.7
16	0.620	0.006	0.664	0.0582	2	0.0291	4.4	yes	1.07	0.05	7.1	0.3	0.7
17	0.546	0.005	0.55	0.03	1	0.03	5.5	yes	1.01	0.06	0.7	0.0	0.1

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty u <sub>c</sub> (A <sub>0</sub> ) (Bq) k=1	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinc. Sum- ming Corr.	Ratio (A/A₀)	Standard uncer- tainty $u_c(A/A_0)$	Percentage Difference <b>D</b> %	Standard uncer- tainty $u_c(A/A_0)$	E <sub>n</sub>
18	0.718	0.007	0.606	0.0303	1	0.0303	5.0	yes	0.84	0.04	-15.5	0.8	-1.8
19	0.831	0.008	0.754	0.042	1	0.042	5.6	yes	0.91	0.05	-9.3	0.5	-0.9
20	0.638	0.006	0.59	0.05	2	0.025	4.2	yes	0.93	0.04	-7.5	0.3	-0.9
21	0.875	0.009	0.77	0.048	1	0.048	6.2	yes	0.88	0.06	-12.0	0.8	-1.1
22	0.752	0.008	0.755	0.057	1	0.057	7.5	no	1.00	0.08	0.4	0.0	0.0
23	0.983	0.010	1	0.1	2	0.05	5.0	yes	1.02	0.05	1.7	0.1	0.2
24	0.757	0.008	0.647	0.034	2	0.017	2.6	yes	0.86	0.02	-14.5	0.4	-2.9
25	0.420	0.004	0.382	0.078	2	0.039	10.2	yes	0.91	0.09	-9.1	0.9	-0.5
26	0.842	0.008	1.53	0.3	2	0.15	9.8	yes	1.82	0.18	81.8	8.1	2.3
27	6.109	0.061	4.35	0.1	2	0.05	1.1	no	0.71	0.01	-28.8	0.4	-11.1
28	0.807	0.008	0.793	0.143	2	0.072	9.0	no	0.98	0.09	-1.8	0.2	-0.1
29	0.780	0.008	0.6731	0.0501	1.65	0.0304	4.5	yes	0.86	0.04	-13.7	0.6	-1.7
30	0.844	0.008	0.746	0.012	2	0.006	0.8	no	0.88	0.01	-11.7	0.1	-4.7
31	0.599	0.006	0.548	0.027	2	0.014	2.5	yes	0.91	0.02	-8.5	0.2	-1.7
32	0.620	0.006	0.6	0.1	2	0.05	8.3	yes	0.97	0.08	-3.2	0.3	-0.2
33	0.736	0.007	0.79	0.08	2	0.04	5.1	yes	1.07	0.06	7.4	0.4	0.7
34	0.898	0.009	0.74	0.073	1	0.073	9.9	yes	0.82	0.08	-17.6	1.7	-1.1
35	0.832	0.008	0.795	0.478	2	0.239	30.1	no	0.96	0.29	-4.4	1.3	-0.1
36	0.854	0.009	0.626	0.04	2	0.02	3.2	no	0.73	0.02	-26.7	0.9	-5.3

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty u <sub>c</sub> (A <sub>0</sub> ) (Bq) k=1	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinc. Sum- ming Corr.	Ratio (A/A <sub>0</sub> )	Standard uncer- tainty $u_c(A/A_0)$	Percentage Difference <b>D</b> %	Standard uncer- tainty $u_c(A/A_0)$	E <sub>n</sub>
37	0.720	0.007	0.75	0.06	2	0.03	4.0	yes	1.04	0.04	4.2	0.2	0.5
38	0.822	0.008	0.59	0.04	1	0.04	6.8	no	0.72	0.05	-28.2	1.9	-2.8
39	0.630	0.006	0.51	0.02	1	0.02	3.9	yes	0.81	0.03	-19.1	0.8	-2.9
40	0.790	0.008	0.75	0.05	2	0.025	3.3	no	0.95	0.03	-5.1	0.2	-0.8
41	0.788	0.008	0.694	0.015	2	0.008	1.1	no	0.88	0.01	-12.0	0.2	-4.3
42	0.634	0.006	0.405	0.025	1	0.025	6.2	yes	0.64	0.04	-36.1	2.3	-4.4
43	0.528	0.005	0.6	0.07	2	0.04	5.8	yes	1.14	0.07	13.7	0.8	1.02
44	0.758	0.008	0.49	0.06	2	0.03	6.1	yes	0.65	0.04	-35.4	2.2	-4.3
45	1.452	0.015	1.57	0.06	1	0.06	3.8	yes	1.08	0.04	8.1	0.3	0.95
46	0.744	0.007	0.64	0.1	1	0.1	15.6	yes	0.86	0.13	-14.0	2.2	-0.5
47	0.743	0.007	0.605	0.054	1	0.054	8.9	yes	0.81	0.07	-18.6	1.7	-1.3
48	0.841	0.008	0.9	0.03	1	0.03	3.3	yes	1.07	0.04	7.0	0.2	0.9
49	0.767	0.008	0.8311	0.0291	2	0.0146	1.8	no	1.08	0.02	8.4	0.2	1.9
50	0.628	0.006	0.48	0.022	1	0.022	4.6	no	0.76	0.04	-23.6	1.1	-3.2
51	0.742	0.007	0.598	0.033	1	0.033	5.5	no	0.81	0.05	-19.5	1.1	-2.1
52	0.800	0.008	0.787	0.094	2	0.047	6.0	yes	0.98	0.06	-1.6	0.1	-0.1
53	0.910	0.009	0.757	0.064	2	0.032	4.2	no	0.83	0.04	-16.8	0.7	-2.3
54	1.727	0.017	0.838	0.021	1	0.021	2.5	no	0.49	0.01	-51.5	1.4	-16.3
55	0.667	0.007	0.48	0.04	2	0.02	4.2	no	0.72	0.03	-28.0	1.2	-4.4

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinc. Sum- ming Corr.	Ratio (A/A <sub>0</sub> )	Standard uncer- tainty $u_c(A/A_0)$	Percentage Difference <b>D</b> %	Standard uncer- tainty $u_c(A/A_0)$	<b>E</b> <sub>n</sub>
56	0.482	0.005	0.436	0.028	1	0.028	6.4	no	0.90	0.06	-9.5	0.6	-0.8
57	1.330	0.013	1.406	0.341	2	0.171	12.1	no	1.06	0.13	5.7	0.7	0.2
58	0.744	0.007	0.72	0.01	1	0.01	1.4	yes	0.97	0.02	-3.2	0.1	-0.96
59	0.896	0.009	0.83	0.037	1	0.037	4.5	yes	0.93	0.04	-7.4	0.3	-0.9
60	0.813	0.008	0.72	0.02	1	0.02	2.8	yes	0.89	0.03	-11.4	0.3	-2.1
61	0.477	0.005	0.5244	0.05	1	0.05	9.5	no	1.10	0.11	9.9	0.9	0.5
62	0.794	0.008	0.751	0.023	1	0.023	3.1	yes	0.95	0.03	-5.5	0.2	-0.9
63	0.676	0.007	0.49003	0.04499	2	0.022495	4.6	yes	0.72	0.03	-27.5	1.3	-4.0
64	0.460	0.005	0.39	0.03	1	0.03	7.7	yes	0.85	0.07	-15.2	1.2	-1.2
65	0.701	0.007	0.6265	0.034	1	0.034	5.4	yes	0.89	0.05	-10.6	0.6	-1.1
66	0.710	0.007	0.6	0.05	1	0.05	8.3	yes	0.84	0.07	-15.5	1.3	-1.1
67	0.816	0.008	0.738	0.059	1	0.059	8.0	yes	0.90	0.07	-9.5	0.8	-0.7

#### 7.2.2 Evaluation

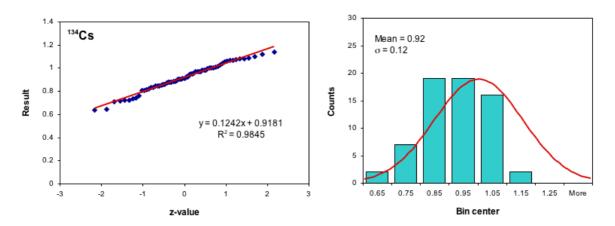
The presence of statistical outliers among the reported results was investigated using the Grubbs' test at a level of significance a=1% and a=5%, as suggested in ISO/IEC 5725-2 (ISO, 1994). Statistical analysis of the results was carried out for the different working dilutions (E1 to E5), as explained in *Section 7.1.2*.

In the case of the ratio values according to the Grubb's test one result was indicated as outlier: Laboratory 26. The outlying value was not discarded, but was included in further evaluations, unless it is declared differently.

Table 11 presents the statistical analysis of the results reported by the participating laboratories per dilution used for spiking the air filters. Moreover, the distribution of the data was tested using the normal probability plot and the frequency histogram. According to both of these graphs presented in *Figure 11*, the  $^{134}$ Cs data are distributed normally and unimodally. The mean values of the reported values for all dilutions are lower than the reference values. The mean value of the ratio is lower than unity by approximately 10%.

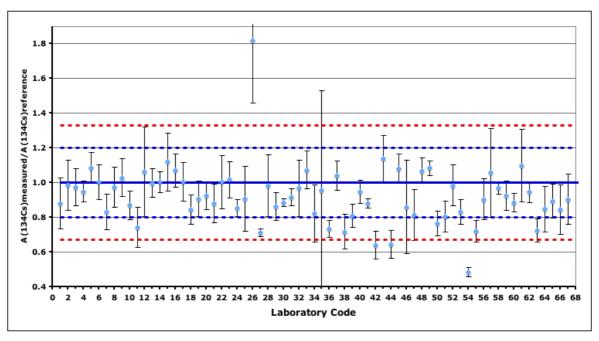
**Table 11**. Statistical analysis of the laboratory reported results for <sup>134</sup>Cs, per dilution. In the last column, the ratios of the measured to the reference values are given.

		E1	E2A	E3	E4	E5	Ratio
	Number of laboratories	2	20	4	9	32	67
	Min (Bq g <sup>-1</sup> )	31.87	4.28	3.81	1.14	0.65	0.49
All	Max (Bq g <sup>-1</sup> )	43.54	7.07	8.35	1.93	1.64	1.82
reported results	Median (Bq g <sup>-1</sup> )	37.70	5.69	7.08	1.53	0.89	0.91
	<b>Mean</b> (Bq g <sup>-1</sup> )	37.70	5.81	6.58	1.54	0.89	0.93
	Standard deviation (Bq g <sup>-1</sup> )	8.25	0.69	1.94	0.23	0.17	0.17
	Number of outliers	0	0	0	0	1	1
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	37.70	5.81	6.58	1.54	0.87	0.91
(5%)	Standard deviation (Bq $g^{-1}$ )	8.25	0.69	1.94	0.23	0.10	0.13
	Number of outliers	0	0	0	0	1	1
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	37.70	5.81	6.58	1.54	0.87	0.91
(1%)	Standard deviation (Bq g <sup>-1</sup> )	8.25	0.69	1.94	0.23	0.10	0.13
	Ref value (Bq g <sup>-1</sup> )	44.8	6.69	7.86	1.757	0.902	1
	Expanded unc. (Bq g <sup>-1</sup> )	0.9	0.13	0.16	0.035	0.018	0.01
	Rel. exp. unc. (%)	2.00	2.00	2.00	2.00	2.00	0.01

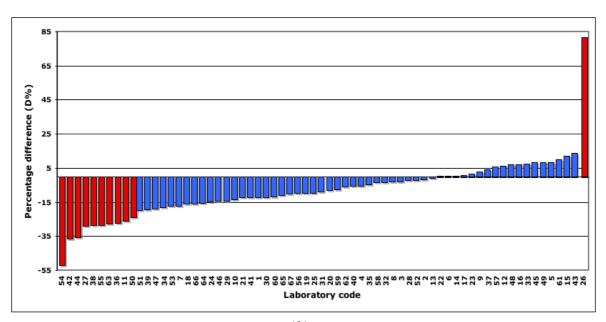


**Fig. 11**. Normal probability plot and frequency histogram of the <sup>134</sup>Cs results after exclusion of the outlier (see text). The red curve in the frequency histogram is the normal probability distribution.

In *Figure 12*, the ratio of the  $^{134}$ Cs activity per spiked air filter as measured and reported by the participating laboratory over the individual spiked activity on the filter (JRC reference value) is plotted. The error bars show the expanded uncertainty (k=2). The blue dashed lines indicate the  $\pm 20\%$  limit from the JRC reference value and the red dashed lines the  $\pm 33\%$  limit from the JRC reference value.

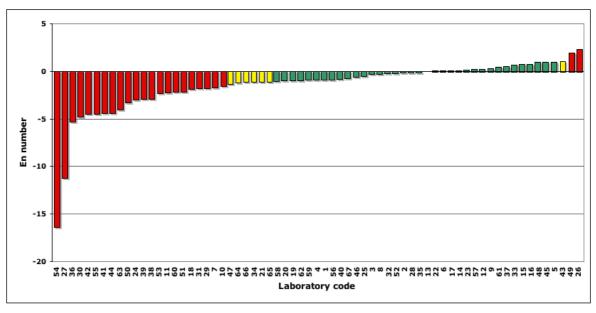


**Fig. 12**. Ratio of  $^{134}$ Cs activity per filter as measured by the participating laboratory to the individual spiked activity on the filter (JRC reference value). Blue dashed lines indicate the  $\pm 20\%$  limit from the JRC reference value and the red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2).



**Fig. 13**. Percentage difference of the  $^{134}$ Cs results reported by the participating laboratories from the reference activity values, plotted in ascending order. Blue color indicates the results within the range  $\pm 20\%$  from the reference value and red indicates results outside that range.

The majority of the laboratories obtained satisfactory results. From *Table 10* and *Figures 12 and 13* it can be seen that 11 laboratories of the 67 (16.4%) reported  $^{134}$ Cs results discrepant by more than  $\pm 20\%$  from the reference value, which is the individual reference activity spiked on each filter. This represents 16% of the participating laboratories. Only 4 laboratories reported results which were outside the more tolerant limit of  $\pm 33\%$  from the reference value. This is just 6.7% of the participating laboratories.

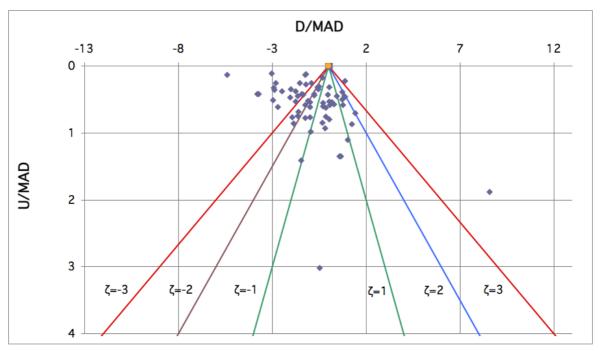


**Fig. 14**.  $E_n$  numbers for the <sup>134</sup>Cs results, plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

To take the expanded uncertainty of the reported results and that of the reference values into account in the analysis, a performance test using  $E_n$  numbers was applied (ISO, 2005a). The  $E_n$  numbers sorted in ascending order are graphically presented in Figure 14.

Under the conditions of this test, 36 out of the 67 (53.7%) reported results for  $^{134}$ Cs are compatible with the reference value, while 31 are not. Among those 31, 24 (35.8%) laboratories reported results with  $1 < |E_n| < 1.5$  and 7 (10.4%) laboratories reported incompatible results with  $|E_n| > 1.5$ .

Another graphical way to compare the results, which underlines the importance of the assigned uncertainties, is the "PomPlot". For the theoretical description of the PomPlot, refer to *Section 6.2.4*. The PomPlot created on the basis of the reported results for <sup>134</sup>Cs is presented in *Figure 15*. The many points outside the  $|\zeta|=3$ , indicate that laboratories underestimated the uncertainties.



**Fig. 15**. PomPlot of the  $^{134}$ Cs data. Green, blue and red solid lines indicate  $\zeta$ -scores=1, 2 and 3, respectively.

# 7.2.3 Parameters possibly influencing the results: Coincidence summing correction

Cesium-134 is relatively easy to measure as it has two relatively abundant gamma-ray lines at relatively high energies at 604.7 and 795.9 keV. However, it suffers from coincidence summing, which occurs for radionuclides emitting two or more photons in sequence within the resolving time of the spectrometer. This results in recording a sum pulse and the consequent loss of the event from the full energy peak of the individual photons. The probability for the summing effect increases with increasing total efficiency, that is with decreasing source-to-detector distance. It is independent of the count rate and thus the activity of the source.

A correction for the coincidence summing can be calculated either using explicit formulae as those given in the literature (Schima and Hoppes, 1983; Debertin and Helmer, 1988) and in *Table 12*, or appropriate computer codes or by consulting available tables (Debertin and Schötzig, 1990). In *Table 12*, the formulae from Schima and Hoppes (1983) for the reciprocal correction factor are given, i.e. the count rates

must be divided by the factor calculated using the formulae to obtain the count rate without coincidence summing. Numbers in parentheses must be replaced by the total efficiency at that energy and numbers in curly brackets must be replaced by the full energy peak efficiency at that energy. As indicative values, those for the two detectors used and described in *Section 3.1* are given as calculated for an active filter of 50 and 90 mm in diameter and measured by placing it on the detector endcap. The correction can be omitted only in the case where the sample is measured relative to a standard of the same nuclide and in the same geometry. The coincidence summing correction factors used and reported by the participants are given in *Table A-11* of *Annex 11*.

**Table 12**. Formulae from Schima and Hoppes (1983) for the reciprocal correction factor and indicative values for Detectors A and B (*Section 3.1*) as calculated for an active filter of 50 and 90 mm in diameter and measured on the detector endcap (see also text).

Energy (keV)	Correction factor (reciprocal)	Detec	ctor A	Detector B		
		36% r	el. eff.	92% r	el. eff.	
	Active area on filter	ø50 mm	ø90 mm	ø50 mm	ø90 mm	
563.2	1-0.007(KX)-0.993(604.7)- 0.997(801.8)	1.370	1.221	1.663	1.439	
569.3	1-0.007(KX)-0.993(604.7)- 0.997(795.9)	1.366	1.218	1.658	1.436	
604.7	1-0.004(KX)-0.074(563.2)- 0.890(795.9)-0.032(1365)- 0.075(801.8)-0.160(569.3)	1.201	1.124	1.342	1.237	
795.9	1-0.006(KX)-0.993(604.7)- 0.179(569.3)	1.213	1.131	1.361	1.248	
801.8	1-0.008(KX)-0.814(604.7)- 0.180(1168)-0.813(563.2)	1.334	1.203	1.573	1.387	

From *Table 13* it can be concluded that the percentage of compatible and non-compatible results is practically independent of the application of the coincidence summing correction. Maybe this is due to the fact that a large number of laboratories used reference spiked filters for the efficiency calibration and possibly spiked with <sup>134</sup>Cs as well, so the correction became unnecessary.

**Table 13**. Distribution of the corrected and non-corrected for coincidence summing results of  $^{134}$ Cs, as compatible and non-compatible according to the  $D_{\%}$  and  $E_n$  criteria.

	Correct coincidence		Non-Corr coincidence		
	45 (6	57%)	22 (33%)		
	Compatible	Non- compatible	Compatible	Non- compatible	
<b>D</b> %	40 (89%)	5 (11%)	16 (73%)	6 (27%)	
E <sub>n</sub>	25 (56%)	20 (44%)	11 (50%)	11 (50%)	

## 7.3 <sup>131</sup>I

## 7.3.1 Reported results

Table 14 presents the reference activities of each individual spiked air filter, the activity results as reported by the participating laboratories, the ratio of the reported to the reference activity, the percentage difference (D%) of the reported to the reference activity and the  $E_n$  number, with their associated standard uncertainties (k=1) for the case of <sup>131</sup>I. The Laboratory Code numbers are the confidential code numbers communicated to the participants and have no correlation with the laboratory codes used during the exercise.

**Table 14**. <sup>131</sup>I reference spiked activity, activity result as reported by the participating laboratories, ratio of the reported to the reference activity, percentage difference (D%) of the reported to the reference activity and  $E_n$  number, with their associated standard uncertainties (k=1).

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinci dence Sum ming Corre ction	Decay during the count- ing interval	Plastic bag measu- red	Ratio (A/A <sub>0</sub> )	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	Percent- age Diffe- rence D%	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	<b>E</b> <sub>n</sub>
1	0.822	0.008	0.7	0.1	2	0.05	7.1	yes	yes	Together	0.85	0.06	-14.9	1.1	-1.2
2	0.863	0.009	0.286	0.023	1	0.023	8.0	no	yes	Together	0.33	0.03	-66.9	5.4	-11.7
3	0.930	0.009	0.53	0.05	2	0.025	4.7	no	no	Together	0.57	0.03	-43.0	2.1	-7.5
4	1.266	0.013	0.0005	0.00033	1	0.00033	66.0	no	no	Together	0.0004	0.00	-100.0	66.0	-49.8
5	1.316	0.013	1.15	0.12	2	0.06	5.2	no	yes	Separately	0.87	0.05	-12.6	0.7	-1.4
6	1.772	0.018	1.51	0.15	1.65	0.09	6.0	yes	no	Not measured	0.85	0.05	-14.8	0.9	-1.4
7	1.058	0.011	0.17	0.012	1	0.012	7.1	yes	yes	Together	0.16	0.01	-83.9	6.0	-27.7
8	7.440	0.075	8	0.9	2	0.45	5.6	no	yes	Separately	1.08	0.06	7.5	0.4	0.6
9	1.269	0.013	1.159	0.122	2	0.061	5.3	yes	yes	Separately	0.91	0.05	-8.6	0.5	-0.9
10	0.640	0.006	0.13	0.02	2	0.01	7.7	yes	yes	Together	0.20	0.02	-79.7	6.2	-21.5
11	0.698	0.007	0.59	0.08	2	0.04	6.8	yes	yes	Together	0.85	0.06	-15.5	1.1	-1.3
12	5.032	0.050	1.462	0.088	1	0.088	6.0	yes	yes	Separately	0.29	0.02	-70.9	4.3	-17.6

Lab Code	Reference activity $A_0$ (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinci dence Sum ming Corre ction	Decay during the count- ing interval	Plastic bag measu- red	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$	Percent- age Diffe- rence D%	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	<b>E</b> <sub>n</sub>
13	1.179	0.012	0.303	0.026	2	0.013	4.3	yes	yes	Separately	0.26	0.01	-74.3	3.3	-24.9
14	1.186	0.012	0.944	0.021	1	0.021	2.2	yes	no	Not measured	0.80	0.02	-20.4	0.5	-5.0
15	0.974	0.010	1.18	0.09	1	0.09	7.6	no	yes	Not measured	1.21	0.09	21.1	1.6	1.1
16	0.994	0.010	0.119	0.0265	2	0.0133	11.1	yes	yes	Separately	0.12	0.01	-88.0	9.8	-26.4
17	0.876	0.009	0.81	0.05	1	0.05	6.2	yes	yes	Separately	0.92	0.06	-7.5	0.5	-0.6
18	0.866	0.009	0.2652	0.02709	1	0.02709	10.2	yes	yes	Together	0.31	0.03	-69.4	7.1	-10.6
19	1.169	0.012	0.229	0.013	1	0.013	5.7	no	yes	Together	0.20	0.01	-80.4	4.6	-26.8
20	1.023	0.010	1.04	0.09	2	0.045	4.3	yes	no	Not measured	1.02	0.05	1.7	0.1	0.2
21	1.056	0.011	0.245	0.017	1	0.017	6.9	yes	yes	Together	0.23	0.02	-76.8	5.4	-20.2
22	1.206	0.012	1.13	0.082	1	0.082	7.3	no	no	Together	0.94	0.07	-6.3	0.5	-0.5
23	1.186	0.012	0.76	0.08	2	0.04	5.3	yes	no	Separately	0.64	0.03	-35.9	1.9	-5.1
24	0.913	0.009	0.057	0.014	2	0.007	12.3	yes	yes	Together	0.06	0.01	-93.8	11.6	-37.1
25	0.681	0.007	0.4	0.081	2	0.041	10.1	yes	yes	Together	0.59	0.06	-41.3	4.2	-3.4

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed <i>k</i> factor	Uncertainty $u_c(A)$ (Bq) $k=1$	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinci dence Sum ming Corre ction	Decay during the count- ing interval	Plastic bag measu- red	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$	Percent- age Diffe- rence D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	E <sub>n</sub>
26	1.350	0.014	0.43	0.077	2	0.039	9.0	yes	yes	Separately	0.32	0.03	-68.1	6.1	-11.3
27	8.669	0.087	0.9	0.02	2	0.01	1.1	no	yes	Together	0.10	0.00	-89.6	1.3	-44.3
28	1.294	0.013	0.556	0.126	2	0.063	11.3	no	yes	Separately	0.43	0.05	-57.0	6.5	-5.8
29	1.264	0.013	0.4588	0.0567	1.65	0.0344	7.5	yes	yes	Separately	0.36	0.03	-63.7	4.8	-11.0
30	1.354	0.014	1.418	0.032	2	0.016	1.1	no	no	Not measured	1.05	0.02	4.7	0.1	1.52
31	0.971	0.010	0.197	0.012	2	0.006	3.0	no	yes	Separately	0.20	0.01	-79.7	2.6	-33.8
32	0.994	0.010	0.92	0.13	2	0.065	7.1	yes	yes	Separately	0.93	0.07	-7.4	0.5	-0.6
33	1.180	0.012	0.78	0.07	2	0.035	4.5	yes	yes	Together	0.66	0.03	-33.9	1.6	-5.4
34	1.084	0.011	0.272	0.028	1	0.028	10.3	no	no	Together	0.25	0.03	-74.9	7.7	-13.5
35	1.003	0.010	1.19	0.48	2	0.24	20.2	no	yes	Together	1.19	0.24	18.6	3.8	0.4
36	1.031	0.010	0.174	0.021	2	0.011	6.0	no	yes	Not measured	0.17	0.01	-83.1	5.1	-29.1
37	1.155	0.012	1.27	0.08	2	0.04	3.1	yes	yes	Separately	1.10	0.04	10.0	0.3	1.4
38	1.318	0.013	0.93	0.09	1	0.09	9.7	no	no	Separately	0.71	0.07	-29.4	2.9	-2.1
39	0.760	0.008	0.08	0.02	1	0.02	25.0	yes	yes	Together	0.11	0.03	-89.5	22.4	-15.9

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed <i>k</i> factor	Uncertainty  u <sub>c</sub> (A) (Bq)  k=1	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinci dence Sum ming Corre ction	Decay during the count- ing interval	Plastic bag measu- red	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$	Percent- age Diffe- rence D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	E <sub>n</sub>
40	0.954	0.010	0.25	0.04	2	0.02	8.0	no	yes	Together	0.26	0.02	-73.8	5.9	-15.9
41	1.264	0.013	0.993	0.015	2	0.008	0.8	no	no	Not measured	0.79	0.01	-21.5	0.3	-9.2
42	0.764	0.008	0.088	0.007	1	0.007	8.0	yes	yes	Together	0.12	0.01	-88.5	7.1	-32.6
43	0.846	0.008	0.26	0.08	2	0.04	15.4	yes	yes	Separately	0.31	0.05	-69.3	10.7	-7.2
44	1.228	0.012	0.23	0.07	2	0.035	15.2	yes	yes	Separately	0.19	0.03	-81.3	12.4	-13.4
45	2.329	0.023	1.87	0.06	1	0.06	3.2	yes	yes	Separately	0.80	0.03	-19.7	0.7	-3.6
46	1.193	0.012	0.96	0.14	1	0.14	14.6	yes	yes	Separately	0.80	0.12	-19.5	2.9	-0.8
47	0.896	0.009	0.104	0.008	1	0.008	7.7	yes	yes	Together	0.12	0.01	-88.4	6.9	-32.9
48	1.349	0.014	1.41	0.05	1	0.05	3.5	yes	yes	Separately	1.04	0.04	4.5	0.2	0.6
49	1.230	0.012	0.6205	0.0323	2	0.0162	2.6	no	no	Together	0.50	0.01	-49.6	1.4	-15.0
50	1.007	0.010	0.81	0.044	1	0.044	5.4	no	no	Separately	0.80	0.04	-19.6	1.1	-2.2
51	1.191	0.012	0.993	0.035	1	0.035	3.5	no	yes	Together	0.83	0.03	-16.6	0.6	-2.7
52	0.965	0.010	0.072	0.013	2	0.007	9.0	yes	yes	Together	0.07	0.01	-92.5	8.4	-38.3
53	1.098	0.011	0.047	0.01	2	0.005	10.6	no	no	Not measured	0.04	0.00	-95.7	10.2	-43.4

Lab Code	Reference activity A <sub>0</sub> (Bq)	Uncertainty $u_c(A_0)$ (Bq) $k=1$	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Report -ed k factor	Uncertainty  u <sub>c</sub> (A) (Bq)  k=1	Relative unc. u <sub>c</sub> (A)/ A (%) k=1	Coinci dence Sum ming Corre ction	Decay during the count- ing interval	Plastic bag measu- red	Ratio (A/A <sub>0</sub> )	Standard uncertainty $u_c(A/A_0)$	Percent- age Diffe- rence D <sub>%</sub>	Standard uncer- tainty u <sub>c</sub> (A/A <sub>0</sub> )	<b>E</b> <sub>n</sub>
54	2.429	0.024	0.344	0.02	1	0.02	5.8	no	yes	Together	0.14	0.01	-85.8	5.1	-33.1
55	1.069	0.011	0.76	0.06	2	0.03	3.9	no	no	Together	0.71	0.03	-28.9	1.2	-4.9
56	0.773	0.008	1.25	0.083	1	0.083	6.6	no	yes	Separately	1.62	0.11	61.8	4.1	2.9
57	1.605	0.016	0.228	0	2	0	0.0	no	no	Together	0.14	0.00	-85.8	0.9	-42.7
58	1.193	0.012	0.93	0.02	1	0.02	2.2	yes	yes	Separately	0.78	0.02	-22.1	0.5	-5.6
59	1.437	0.014	0.295	0.02	1	0.02	6.8	yes	no	Separately	0.21	0.01	-79.5	5.4	-23.2
60	1.303	0.013	1.1	0.03	1	0.03	2.7	yes	no	Together	0.84	0.02	-15.6	0.5	-3.1
61	0.773	0.008	0.6817	0.04	1	0.04	5.9	no	yes	Separately	0.88	0.05	-11.8	0.7	-1.1
62	1.274	0.013	0.391	0.016	1	0.016	4.1	yes	yes	Separately	0.31	0.01	-69.3	2.9	-21.6
63	0.815	0.008	0.088108	0.050394	2	0.02519 7	28.6	yes	no	Together	0.11	0.03	-89.2	25.5	-13.7
64	0.745	0.007	0.14	0.011	1	0.011	7.9	yes	yes	Together	0.19	0.01	-81.2	6.4	-22.8
65	0.985	0.010	0.1707	0.0104	1	0.0104	6.1	yes	yes	Together	0.17	0.01	-82.7	5.1	-28.4
66	0.857	0.009	0.07	0.007	1	0.007	10.0	yes	yes	Together	0.08	0.01	-91.8	9.2	-35.5
67	0.984	0.010	0.246	0.025	1	0.025	10.2	yes	yes	Together	0.25	0.03	-75.0	7.7	-13.7

#### 7.3.2 Evaluation

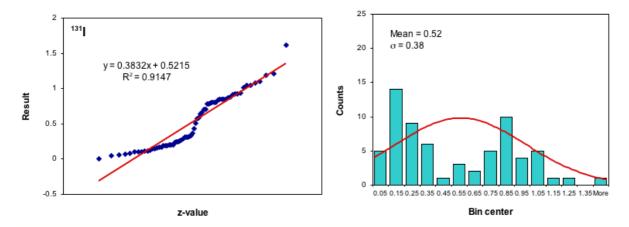
The presence of statistical outliers among the reported results was investigated using the Grubbs' test at a level of significance a=1% and a=5%, as suggested in ISO/IEC 5725-2 (ISO, 1994). Statistical analysis of the results was carried out for the different working dilutions (E1 to E5) as explained in *Section 7.1.2*.

In the case of the ratio values according to the Grubb's test two results were indicated as outliers: Laboratories 1 and 35. The outlying values were not discarded, but were included in further evaluations, unless it is declared differently.

Table 15 presents the statistical analysis of the results reported by the participating laboratories per dilution used for spiking the air filters. Moreover, the distribution of the data was tested using the normal probability plot and the frequency histogram. According to both of these graphs presented in *Figure 16*, the <sup>131</sup>I data are not distributed normally and unimodally. The mean of the reported values for all dilutions is lower than the reference values and the mean of the ratio is only 62 to 66% of the unity.

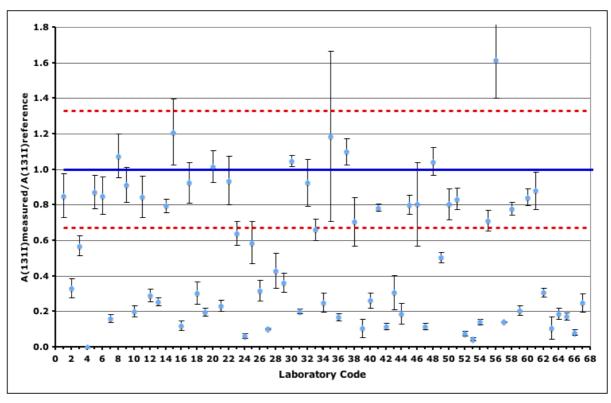
**Table 15**. Statistical analysis of the laboratory reported results for <sup>131</sup>I, per dilution. In the last column, the ratios of the measured to the reference values are given.

		E1	E2A	E3	E4	E5	Ratio
	Number of laboratories	2	20	4	9	32	67
	Min (Bq g <sup>-1</sup> )	6.59	0.35	1.57	0.53	0.00	0.00
AII	Max (Bq g <sup>-1</sup> )	68.29	9.58	3.21	2.51	2.34	2.94
reported results	Median (Bq g <sup>-1</sup> )	37.44	1.33	2.04	0.94	1.16	0.66
	<b>Mean</b> (Bq g <sup>-1</sup> )	37.44	2.31	2.21	1.20	1.06	0.66
	Standard deviation (Bq g <sup>-1</sup> )	43.63	2.42	0.71	0.80	0.50	0.44
	Number of outliers	0	2	0	0	0	2
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	37.44	1.66	2.43	1.20	1.06	0.62
(5%)	Standard deviation (Bq g <sup>-1</sup> )	43.63	1.33	0.69	0.80	0.50	0.32
	Number of outliers	0	4	0	0	0	4
Omitting outliers	<b>Mean</b> (Bq g <sup>-1</sup> )	37.44	1.25	2.43	1.20	1.06	0.62
(1%)	Standard deviation (Bq g <sup>-1</sup> )	43.63	0.66	0.69	0.80	0.50	0.32
	Ref value (Bq g <sup>-1</sup> )	63.5	8.07	11.05	2.846	1.446	1
	Expanded unc. (Bq g <sup>-1</sup> )	0.3	0.03	0.05	0.012	0.006	0.01
	Rel. exp. unc. (%)	0.43	0.43	0.43	0.43	0.43	0.01

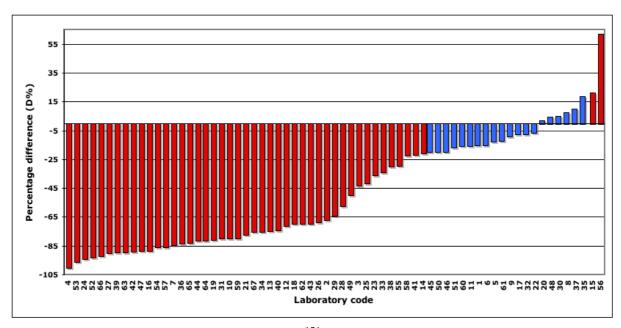


**Fig. 16**. Normal probability plot and frequency histogram of the <sup>131</sup>I results. The red curve in the frequency histogram is the normal probability distribution.

In Figure 17, the ratio of the  $^{131}$ I activity per spiked air filter as measured and reported by the participating laboratory over the individual spiked activity on the filter (JRC reference value) is plotted. The error bars show the expanded uncertainty (k=2). The blue dashed lines indicate the  $\pm 20\%$  limit from the JRC reference value and the red dashed lines the  $\pm 33\%$  limit from the JRC reference value.

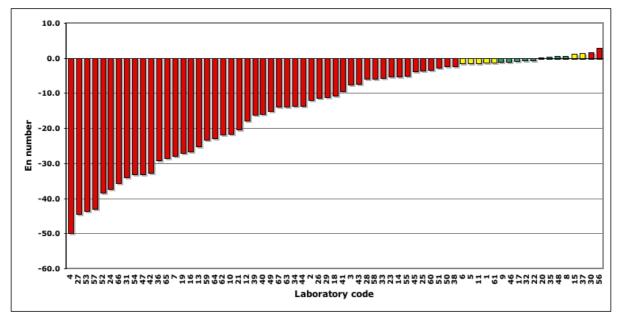


**Fig. 17**. Ratio of  $^{131}$ I activity per spiked air filter as measured by the participating laboratory to the individual spiked activity on the filter (JRC reference value). Blue dashed lines indicate the  $\pm 20\%$  limit from the JRC reference value and the red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2).



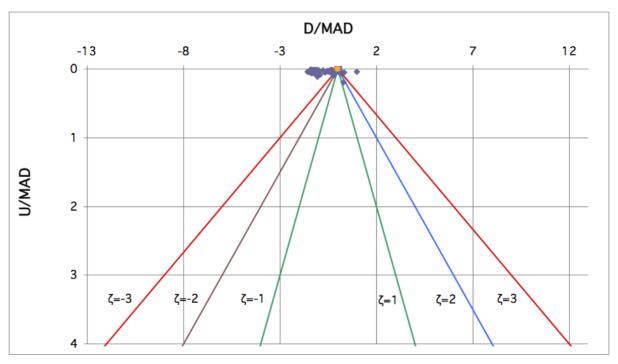
**Fig. 18**. Percentage difference of the  $^{131}$ I results reported by the participating laboratories from the reference activity values, plotted in ascending order. Blue color indicates the results within the range  $\pm 20\%$  from the reference value and red indicates results outside that range.

The majority of the laboratories reported results below the reference activity value. From *Table 14* and *Figures 17* and 18 it can be seen that 47 of the 67 laboratories reported  $^{131}$ I results discrepant by more than  $\pm 20\%$  from the reference value, which is the individual reference activity spiked on each filter. This represents 70% of the participating laboratories. Fourty one laboratories reported results which were outside the more tolerant limit of  $\pm 33\%$  from the reference value. This is just 61% of the participating laboratories.

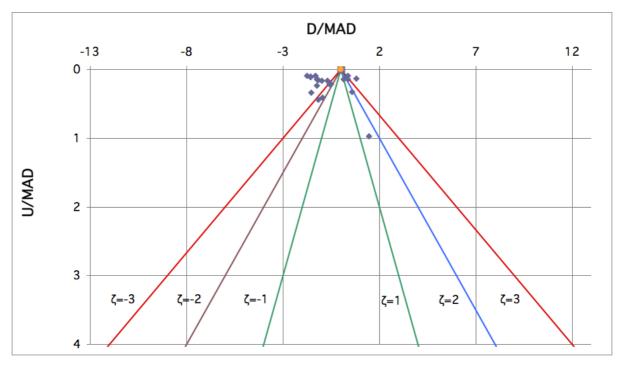


**Fig. 19**.  $E_n$  numbers for the <sup>131</sup>I results plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

To take the expanded uncertainty of the reported results and that of the reference values into account in the analysis, a performance test using  $E_n$  numbers was applied (ISO, 2005a). The  $E_n$  numbers sorted in ascending order are graphically presented in Figure 19.



**Fig. 20a**. PomPlot of the  $^{131}I$  data. Green, blue and red solid lines indicate  $\zeta$ -scores=1, 2 and 3, respectively.



**Fig. 20b**. PomPlot of the  $^{131}$ I data within the  $\pm 20\%$  range from their respective reference values. Green, blue and red solid lines indicate  $\zeta$ -scores=1, 2 and 3, respectively.

Under the conditions of this test, 9 (13%) out of the 67 reported results for  $^{131}I$  are compatible with the reference value, while 58 are not. Among those 58, 7 (10%) laboratories reported incompatible results with  $1 < |E_n| < 1.5$  and 51 (76%) reported incompatible results with  $|E_n| > 1.5$ .

The PomPlot (see theoretical description in *Section 6.2.4*) created on the basis of the reported results for  $^{131}$ I is presented in *Figure 20a*. Because of most reults being far from their respective referencevalues, the MAD value is abnormally high and this results in the peculiar PomPlot of *Figure 20a*. If only results within the  $\pm 20\%$  range from their respective reference values are taken into account, the calculated PomPlot is that depicted in *Figure 20b*. The points outside the  $|\zeta|=3$ , indicate those laboratories that probably underestimated the uncertainties on their reported results for  $^{131}$ I.

## 7.3.3 Parameters possibly influencing the results

Although measuring  $^{131}$ I does not present any problem, as its main gamma-ray lines are emitted at relatively high energies, at 284.3, 364.5 and 637.0 keV with emission probabilities of 6.14, 81.2 and 7.12%, respectively, there are some issues to be brought to the attention of the analyst.

## 7.3.3.1 Decay during measurement

Because of the relatively short half-life of  $^{131}$ I (8.0233 (19) d), besides the usual decay correction applied for reporting the results on the reference date of 1 March 2016 0:00 UTC, a second correction maybe necessary for longer acquisition periods to compensate for the decay during the measurement period (Debertin and Helmer, 1988). As it is shown in *Table 16*, this correction is negligible for an acquisition time of 1 hour, it becomes 1% for a 6-hour acquisition and close to 9% for a 2-day acquisition. An alternative correction is to take in the decay correction calculations as start time of the measurement the mid-point of the measurement time.

Table 1	6. Correction	factors for	or the decay	during m	easurement	of <sup>131</sup> I
Iable T	O. COLLECTION	Tactors re	// LITE UELAV	uui iiiu iii	casal cilicii.	01 1.

Counting interval	Correction factor
1 h	1.002
2 h	1.004
6 h	1.011
12 h	1.022
24 h	1.044
2 d	1.089
3 d	1.135
5 d	1.231
7 d	1.333

Table 17 shows the distribution of the corrected and non-corrected for decay during measurement results of  $^{131}$ I, as reported by the participants, as compatible and non-compatible according to the  $D_{\%}$  and  $E_n$  criteria. Nineteen laboratories did not correct for decay during measurement, but the compatible and non-compatible distribution of the

reported resuts does not differ from the distribution of those reported by laboratories which corrected for decay during measurement.

**Table 17**. Distribution of the corrected and non-corrected for decay during measurement results of  $^{131}$ I, as reported by the participants, as compatible and non-compatible according to the  $D_{\%}$  and  $E_n$  criteria.

	Corrected during mea	-		ted for decay easurement		
	48 (7	72%)	19 (28%)			
	Compatible	Non- compatible	Compatible	Non- compatible		
D <sub>%</sub>	14 (29%)	34 (71%)	6 (32%)	13 (68%)		
E <sub>n</sub>	7 (15%)	41 (85%)	2 (10%) 17 (90%)			

### 7.3.3.2 Coincidence summing correction

The three principal gamma-ray lines of  $^{131}$ I at 284.3, 364.5 and 637.0 keV suffer from coincidence summing (for explanation refer to *Section 7.2.3*). However, the correction factors are close to unity, with the exception of the 284.3 keV line in combination with high efficiency detectors. The formulae from Schima and Hoppes (1983) for the reciprocal correction factor and indicative values for Detectors A and B (*Section 3.1*) as calculated for an active filter of 50 and 90 mm in diameter and measured on the detector endcap are given in *Table 18*.

**Table 18**. Formulae from Schima and Hoppes (1983) for the reciprocal correction factor and indicative values for Detectors A and B (Section 3.1) as calculated for an active filter of 50 and 90 mm in diameter and measured on the detector endcap (see also Section 7.2.3).

Energy (keV)	Correction factor (reciprocal)	Detec	ctor A	Dete	ctor B	
		36% r	el. eff.	92% rel. eff.		
	Active area on filter	ø50 mm	ø90 mm	ø50 mm	ø90 mm	
284.3	1-0.467(KX)-0.389(80.2)	1.044	1.019	1.276	1.169	
364.5	1+0.029{80.2}{284.3}/{364.5}	0.997	0.999	0.989	0.995	
637	1+0.007{364.5}{272.0}/{637.0}	0.999	0.999	0.998	0.999	

### 7.3.3.3 Radioactivity transferred to the protective plastic bag

During the exercise it became obvious that there was transfer of a variable quantity of the  $^{131}$ I activity from the spiked air filter to the containing (inner) protective plastic bag. More affected were the small filters being directly in contact with the plastic bag. The larger filters were folded with the activity inwards and so protected. This activity transfer concerns only  $^{131}$ I; the  $^{137}$ Cs and  $^{134}$ Cs were not affected.

At the JRC we measured originally the quality control spiked air filters in their plastic bags. To check for the activity transfer, we measured two spiked air filters (IRMM-03 and IRMM-06) alone and their corresponding plastic bags separately. The findings are

given in *Table 19*; a fraction of 3 to 6% of the  $^{131}$ I activity had been transferred to the plastic bag.

**Table 19**. Count rate results for the 364 keV line of <sup>131</sup>I from measuring filters IRMM-03 and IRMM-06 and their plastic bags. The percent ratio of the count rate obtained from the plastic bag to that from the filter is given in the last column.

Item	Detector	Count rate (364 keV)	Ratio %
Filter_IRMM-06 in original bag	Α	0.0712	
Filter_IRMM-06 Plastic bag only		0.0023	3.2
Filter_IRMM-06 in new bag		0.0700	
SUM		0.0723	
Filter_IRMM-03 in original bag	В	0.0538	
Filter_IRMM-03 Plastic bag only		0.0046	5.6
Filter_IRMM-03 in new bag		0.0480	
SUM		0.0526	
Filter_IRMM-06_in original bag	В	0.1711	
Filter_IRMM-06 Plastic bag only		0.0058	3.5
Filter_IRMM-06 in new bag		0.1671	
SUM		0.1729	

Although it was originally recommended to the participants to measure the plastic bag as well, they were warned in the course of the exercise (see communication in *Annex 2E*) to measure both the filter and the plastic bag (either together or separately) and add the measured activities. It is evident that the counting efficiency of the plastic bag is different than that of the filter, but even with an estimation of the counting efficiency, adding the transferred activity would be an improvement on the  $^{131}$ I result. *Table 20* gives the distribution of the results for  $^{131}$ I corrected and non-corrected for the transferred activity to the protective plastic bag, as reported by the participants, as compatible and non-compatible according to the  $D_{\%}$  and  $E_n$  criteria. Eight laboratories reported that they did not measure the protective plastic bags, but there is no significant difference in the distribution of the reported results as compatible or non-compatible compared to the laboratories that they carried out the appropriate corrections.

**Table 20**. Distribution of the results for  $^{131}$ I corrected and non-corrected for the transferred activity to the protective plastic bag, as reported by the participants, as compatible and non-compatible according to the  $D_{\%}$  and  $E_{n}$  criteria.

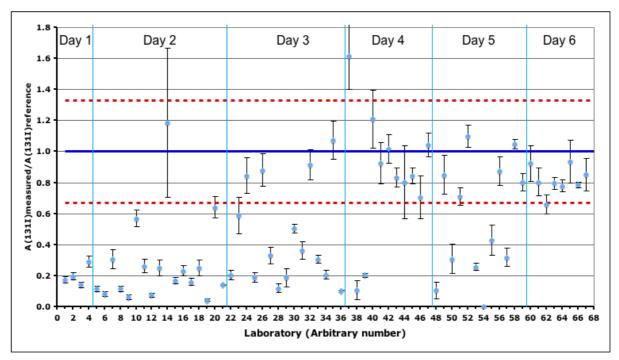
	Plastic bag (together w separa	ith filter or	Plastic bag not measured			
	59 (8	38%)	8 (12%)			
	Compatible	Non- compatible	Compatible	Non- compatible		
<b>D</b> %	17 (29%)	42 (71%)	3 (37.5%)	5 (62.5%)		
E <sub>n</sub>	8 (14%)	51 (86%)	1 (12.5%)	7 (87.5%)		

#### 7.3.4 Conclusions

By measuring the quality control spiked air filters and point sources it became obvious that although the results of the activity were as expected for <sup>137</sup>Cs and <sup>134</sup>Cs, they were systematically low and with significant spread for <sup>131</sup>I. For logistics reasons, the quality control sources were measured at the JRC after dispatching the spiked air filters to the participants. It is worth noticing that both filters and point sources exhibit equally lower than expected activities for <sup>131</sup>I. The first occurrence was observed with point source PS014. Point sources and liquid scintillation sources made before that gave excellent results with deviations from the expected values of less than 1%.

The cause of this decrease of the  $^{131}$ I activity is not clear. All point sources and spiked air filters were prepared gravimetrically in the same way and by the same operator under controlled conditions of temperature, pressure and humidity. The drying of the sources was done overnight and not in a forced way (e.g. heating). Volatility of iodine could be the culprit; a solution of 25 mg g $^{-1}$  KI and 25 mg g $^{-1}$  Na $_2$ S $_2$ O $_3$  was used to keep the iodine in solution, but it might be that the salt concentration was not adequate.

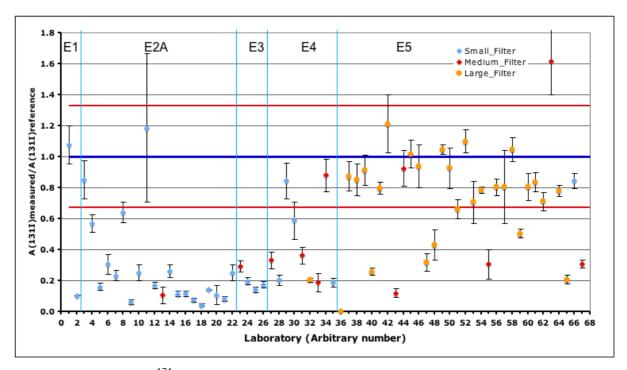
In an attempt to explain the discrepancies, the ratio of the measured to the reference activity values of <sup>131</sup>I was plotted in a number of ways, of which two are shown in *Figures 21* and *22*. In *Figure 21* the ratio was plotted per day of production of the spiked air filters and with ascending of spiked activity (per day). Notice that smaller filters were spiked first and then the larger ones. Dilution E5 shows better performance, but being the most dilute was used for the larger filters. No effect can be observed in terms of activity levels spiked or mass of the spiked solution.



**Fig. 21**. Ratio of  $^{131}$ I activity per spiked air filter as measured by the participating laboratory over the individual spiked activity on the filter (JRC reference value) plotted per day of preparation and with ascending spiked activity (per day). Blue dashed lines indicate the  $\pm 20\%$  limit from the reference value and red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2). The laboratory numbers are arbitrary.

In *Figure 22* the ratio was plotted per dilution used for spiking, with indication of the filter size in color. It can be observed that larger filters behaved better in retaining <sup>131</sup>I spiked on them. This can be due to the material of the filter itself; filters with fiberglass composition behaved better than those from cellulose. This is not absolute though and does not explain why the point sources prepared by depositing aliquots of the radioactive solution onto a plastic foil, exhibit the same behaviour.

Because the mechanism of depositing the activity by spiking is different han that by collecting air particulates, we estimate that such effect should not be observed in air filters loaded by exposing them to an aerosol.

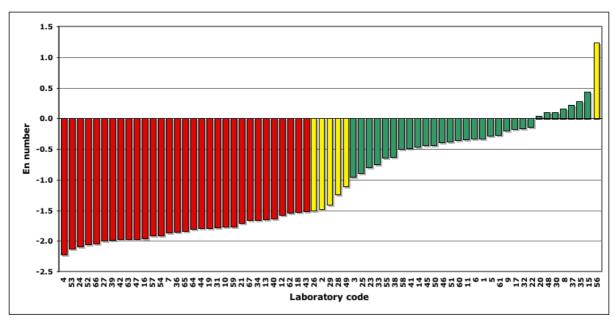


**Fig. 22**. Ratio of  $^{131}$ I activity per spiked air filter as measured by the participating laboratory over the individual spiked activity on the filter (JRC reference value) plotted per dilution used for spiking and with indication (in color) of the filter size. Blue dashed lines indicate the  $\pm 20\%$  limit from the reference value and red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2). The laboratory numbers are arbitrary.

The spiked air filters could also lose some <sup>131</sup>I activity by being exposed to heat. Some participants mentioned in private communication (but not reported in the questionnaire) heating the filters prior to measuring in order to dry them, as routine in their procedure.

One way to compensate for the loss of  $^{131}$ I and reflect the spread of the effective spiking results in the calculation of the  $E_n$  number could be to artificially increase the uncertainty on the reference value to 22.5% (ne half of the mean discrepancy of the quality control sources measured activity from their correspondig expected activity values).

Figure 23 shows the  $E_n$  numbers for the <sup>131</sup>I results plotted in ascending order, when the uncertainty on the reference value is increased to 22.5%. In such case, 30 (45%) laboratories score  $|E_n| \le 1$ , 6 (9%) laboratories score  $1 < |E_n| < 1.5$  and 31 (46%) score  $|E_n| > 1.5$ . The laboratories in the three  $E_n$  categories can be identified from Figure 23.



**Fig. 23**.  $E_n$  numbers for the <sup>131</sup>I reported results plotted in ascending order, when the uncertainty on the reference value is increased to 22.5%. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

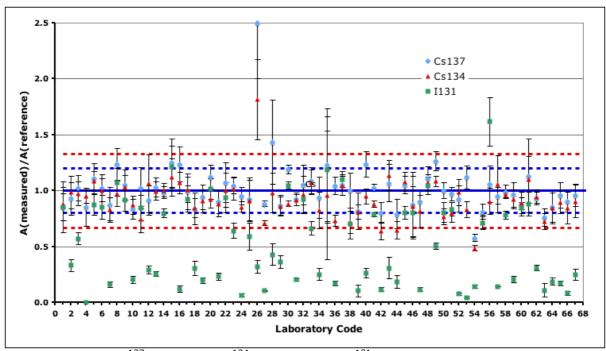
## 8. Conclusions

The 2016 ENV57/MetroERM Interlaboratory Comparison was organized and conducted by the JRC in the frame of the ENV57/MetroERM "Metrology for radiological early warning networks in Europe", a European Metrology Research Program (EMRP). All 67 participating laboratories reported valid activity results for all of the three radionuclides of interest, namely <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. *Figure 24* is a color coded cumulative plot of the ratio of the measured to the reference activity values to judge the performance of the laboratories for all three radionuclides simultaneously.

Table 21 lists the percentage difference,  $D_{\%}$ , and the  $E_n$  number for each of the three radionuclides, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. Values of  $D_{\%}$  outside the ±20% limit from the reference value are in yellow. Absolute values of  $E_n$  between 1 and 1.5 are also in yellow and those above 1.5 are in red.

The compatibility criterion is satisfied when both the percent difference,  $D_{\%}$ , being withing the  $\pm 20\%$  limit from the reference value and the absolute  $E_n$  being smaller than unity. The compatibility for each laboratory and for the three radionuclides is given in the last three columns of *Table 21*.

In *Table 22* the number of results (and the percentage over all results in parentheses) satisfying the  $<\pm20\%$  and  $<\pm33\%$  discrepancy from the reference values is given. The performance of the laboratories is similar for measuring  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  with 84% of the laboratories reporting values within the  $\pm20\%$  limit from the reference values for both nuclides and 63% and 54%, respectively, scoring  $|E_n| \leq 1$ . Equally, 61% and 54% of the laboratories reported compatible results, satisfying both criteria. However, only 40% of the laboratories reported compatible results for both  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  (*Table 23*). This is strange as one would expect that laboratories measuring correctly one of the two nuclides should be able to measure correctly the other as well.



**Fig. 24**. Ratio of  $^{137}$ Cs (in blue),  $^{134}$ Cs (in red) and  $^{131}$ I (in green) activity per spiked air filter as measured by the participating laboratory to the individual spiked activity on the filter (JRC reference value). Blue dashed lines indicate the  $\pm 20\%$  limit from the reference value and the red dashed lines the  $\pm 33\%$  limit. The error bars show the expanded uncertainty (k=2).

For  $^{131}$ I only 30% of the results were within the  $\pm 20\%$  limit from the reference values and 13% obtained  $|E_n| \le 1$ . At least, this poor performance is due partly to the quality of the spiked air filters, in what concerns  $^{131}$ I (refer to Section 7.3.3). It was the first attempt to include  $^{131}$ I in an ILC organised by JRC; it is a radionuclide which could be released in case of a nuclear incident or accident and therefore of interest to the laboratories monitoring the environment. Because of its properties and its rather short half-life it is not very often included as measurand in ILCs or PTs.

Nevertheless, 30% of the participating laboratories reported  $^{131}$ I activities within the  $\pm 20\%$  limit from the reference values and 39% the more tolerent  $\pm 33\%$  range. In those circumstances, it is not surprising that only 13% of the laboratories reported compatible results for  $^{131}$ I, 13% for both  $^{134}$ Cs and  $^{131}$ I and only 7.5%, i.e. 5 laboratories, for  $^{137}$ Cs and  $^{131}$ I or for all three radionuclides simultaneously.

The lower success rate for the  $E_n$  number, compared to the percentage difference,  $D_{\%}$ , reflects the fact that some laboratories underestimated the uncertainties on their results and so they were penalized in the  $E_n$  number test. The latter takes into account the uncertainty and although the results pass the  $D_{\%}$  test, they fail the  $E_n$  test. In Table A-12 of Annex 12 the detailed uncertainty budgets, as reported by the participating laboratories, are given for each of the three radionuclides. Some laboratories did not report the individual components of their uncertainty budgets. In column 13 the combined standard uncertainty as reported by the laboratories is given, in column 15 the calculated from the components combined standard uncertainty is given and in column 16 the combined standard uncertainty as deduced from the reported results. Some discrepencies were observed and the concerned laboratories should improve the way they calculate their uncertainty budgets by following the "GUM approach" and establish a complete evaluation of uncertainty sources. A workshop was organised by JRC on 7 and 8 April, 2016 (Annex 2H) to bring together the participants of this ILC to discuss all aspects of the ILC and provide ideas and suggestions to improve the performance of the laboratories.

**Table 21.** Performance of the laboratories: percentage difference,  $D_{\%}$ , and  $E_n$  number for each of the three radionuclides, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. Values of  $D_{\%}$  outside the ±20% limit from the reference value are in yellow. Absolute values of  $E_n$  between 1 and 1.5 are also in yellow and those above 1.5 are in red.

	<sup>137</sup> Cs		<sup>134</sup> Cs		<sup>131</sup> I		Compatibility		
Lab Code	<b>D</b> %	E <sub>n</sub>	<b>D</b> %	E <sub>n</sub>	<b>D</b> %	E <sub>n</sub>	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I
1	-14.5	-0.6	-12.0	-0.8	-14.9	-1.2	yes	yes	no
2	-7.2	-0.5	-1.4	-0.1	-66.9	-11.7	yes	yes	no
3	2.3	0.2	-2.7	-0.3	-43.0	-7.5	yes	yes	no
4	-14.9	-0.9	-5.0	-0.8	-100.0	-49.8	yes	yes	no
5	10.9	0.8	8.5	0.96	-12.6	-1.4	yes	yes	no
6	2.2	0.2	0.5	0.0	-14.8	-1.4	yes	yes	no
7	-12.6	-0.8	-16.7	-1.6	-83.9	-27.7	yes	no	no
8	23.6	1.7	-2.7	-0.2	7.5	0.6	no	yes	yes
9	5.2	0.4	2.9	0.3	-8.6	-0.9	yes	yes	yes
10	-16.4	-1.9	-12.9	-1.55	-79.7	-21.5	no	no	no
11	2.2	0.1	-25.7	-2.2	-15.5	-1.3	yes	no	no
12	-8.5	-0.6	6.2	0.2	-70.9	-17.6	yes	yes	no
13	3.1	0.4	-0.3	0.0	-74.3	-24.9	yes	yes	no
14	-1.9	-0.3	0.5	0.1	-20.4	-5.0	yes	yes	no
15	24.8	1.2	11.9	0.7	21.1	1.1	no	yes	no
16	23.7	1.51	7.1	0.7	-88.0	-26.4	no	yes	no
17	-0.8	0.0	0.7	0.1	-7.5	-0.6	yes	yes	yes
18	-16.3	-1.1	-15.5	-1.8	-69.4	-10.6	no	no	no
19	-5.8	-0.5	-9.3	-0.9	-80.4	-26.8	yes	yes	no
20	12.1	1.2	-7.5	-0.9	1.7	0.2	no	yes	yes
21	-10.1	-0.98	-12.0	-1.1	-76.8	-20.2	yes	no	no
22	7.2	0.4	0.4	0.0	-6.3	-0.5	yes	yes	yes
23	4.5	0.6	1.7	0.2	-35.9	-5.1	yes	yes	no
24	-4.9	-0.6	-14.5	-2.9	-93.8	-37.1	yes	no	no
25	-7.6	-0.4	-9.1	-0.5	-41.3	-3.4	yes	yes	no
26	149.7	3.1	81.8	2.3	-68.1	-11.3	no	no	no
27	-11.7	-4.5	-28.8	-11.1	-89.6	-44.3	no	no	no
28	43.4	1.2	-1.8	-0.1	-57.0	-5.8	no	yes	no
29	-13.2	-1.4	-13.7	-1.7	-63.7	-11.0	no	no	no
30	19.1	5.1	-11.7	-4.7	4.7	1.52	no	no	no
31	-7.5	-1.3	-8.5	-1.7	-79.7	-33.8	no	no	no
32	4.9	0.3	-3.2	-0.2	-7.4	-0.6	yes	yes	yes
33	9.0	0.8	7.4	0.7	-33.9	-5.4	yes	yes	no

	<sup>137</sup> Cs		<sup>134</sup> Cs		<sup>131</sup> I		Compatibility		
Lab Code	<b>D</b> %	<b>E</b> <sub>n</sub>	<b>D</b> %	<b>E</b> <sub>n</sub>	<b>D</b> %	<b>E</b> <sub>n</sub>	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I
34	-6.2	-0.3	-17.6	-1.1	-74.9	-13.5	yes	no	no
35	22.8	0.5	-4.4	-0.1	18.6	0.4	no	yes	yes
36	4.5	0.6	-26.7	-5.3	-83.1	-29.1	yes	no	no
37	5.4	0.6	4.2	0.5	10.0	1.4	yes	yes	no
38	0.2	0.0	-28.2	-2.8	-29.4	-2.1	yes	no	no
39	-18.4	-1.1	-19.1	-2.9	-89.5	-15.9	no	no	no
40	23.6	2.1	-5.1	-0.8	-73.8	-15.9	no	yes	no
41	2.5	0.8	-12.0	-4.3	-21.5	-9.2	yes	no	no
42	-19.9	-2.1	-36.1	-4.4	-88.5	-32.6	no	no	no
43	6.8	0.4	13.7	1.02	-69.3	-7.2	yes	no	no
44	-20.8	-1.6	-35.4	-4.3	-81.3	-13.4	no	no	no
45	5.2	0.7	8.1	0.95	-19.7	-3.6	yes	yes	no
46	-12.6	-0.4	-14.0	-0.5	-19.5	-0.8	yes	yes	yes
47	-10.1	-1.02	-18.6	-1.3	-88.4	-32.9	no	no	no
48	10.7	1.03	7.0	0.9	4.5	0.6	no	yes	yes
49	26.4	3.3	8.4	1.9	-49.6	-15.0	no	no	no
50	0.1	0.0	-23.6	-3.2	-19.6	-2.2	yes	no	no
51	-2.8	-0.5	-19.5	-2.1	-16.6	-2.7	yes	no	no
52	-7.6	-0.7	-1.6	-0.1	-92.5	-38.3	yes	yes	no
53	11.9	1.1	-16.8	-2.3	-95.7	-43.4	no	no	no
54	-42.2	-12.5	-51.5	-16.3	-85.8	-33.1	no	no	no
55	-18.7	-2.8	-28.0	-4.4	-28.9	-4.9	no	no	no
56	5.7	0.4	-9.5	-0.8	61.8	2.9	yes	yes	no
57	-4.7	-0.6	5.7	0.2	-85.8	-42.7	yes	yes	no
58	-1.0	-0.2	-3.2	-0.96	-22.1	-5.6	yes	yes	no
59	-3.8	-0.3	-7.4	-0.9	-79.5	-23.2	yes	yes	no
60	-12.0	-1.1	-11.4	-2.1	-15.6	-3.1	no	no	no
61	13.1	0.4	9.9	0.5	-11.8	-1.1	yes	yes	no
62	-6.7	-1.2	-5.5	-0.9	-69.3	-21.6	no	yes	no
63	-24.0	-3.1	-27.5	-4.0	-89.2	-13.7	no	no	no
64	-14.8	-0.99	-15.2	-1.2	-81.2	-22.8	yes	no	no
65	-4.0	-0.4	-10.6	-1.1	-82.7	-28.4	yes	no	no
66	-9.5	-0.97	-15.5	-1.1	-91.8	-35.5	yes	no	no
67	-4.2	-0.4	-9.5	-0.7	-75.0	-13.7	yes	yes	no

From the comparison of the statistics of the reported results for  $^{137}$ Cs, it is concluded that the performance of the laboratories analysing this nuclide is rather stable. The results for  $^{134}$ Cs were equally satisfactory, albeit not necessarily from the same laboratories reporting compatible results for  $^{137}$ Cs. The observed negative bias on the reported results compared to the reference values, might be partly attributed to a non-adequate summing correction applied by some laboratories. However, the statistical results show that the coincidence summing correction was taken into account rather successfully, either by correcting or by using suitable sources for the counting efficiency calibration, as there is no significant difference in the distribution of compatible and non-compatible results (*Table 13*).

In the case of <sup>131</sup>I, the transfer of activity to the protective plastic bag was a minor problem and most of the participants measured the plastic bag and added the activity to the activity measured on the spiked air filter.

**Table 22.** Cumulative table showing the performance of the laboratories for the three radionuclides, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I. The numbers give the number of laboratories with the percent fraction in parentheses.

	Number of laboratories						
Nuclides	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I				
<i>D</i> % ≤20	56 (83.6%)	56 (83.6%)	20 (29.9%)				
<i>D</i> % >20	11 (16.4%)	11 (16.4%)	47 (70.1%)				
<i>D</i> % ≤33%	64 (95.5%)	63 (94.0%)	26 (38.8%)				
D% >33%	3 (4.5%)	4 (6.0%)	41 (61.2%)				
<i>E</i> <sub>n</sub>  ≤1	42 (62.7%)	36 (53.7%)	9 (13.4%)				
1< E <sub>n</sub>  <1.5	12 (17.9%)	7 (10.4%)	7 (10.4%)				
<i>E</i> <sub>n</sub>  >1.5	13 (19.4%)	24 (35.8%)	51 (76.1%)				

The integrity of the spiked <sup>131</sup>I activity was a major problem and because of that we cannot arrive at a clear conclusion on the performance of the participating laboratories for this radionuclide. We can however conclude that:

- For laboratories which reported compatible results, it means that they had received an air filter with the integral of the spiked  $^{131}I$  activity and that they had measured  $^{131}I$  correctly.
- For laboratories which did not report compatible results, the reasons can be multiple:
  - Air filter maintaining only part of the spiked <sup>131</sup>I activity;
  - Measurement of <sup>131</sup>I not performed correctly;
  - Both of the above.

This exercise highlights the need for additional studies of the radioiodine behaviour on the air filters.

A number of laboratories (8 or 12%) did not use the recommended values for the halflives and the gamma-ray emission probabilities. However, given the values used and the measurement conditions, we estimate that the discrpencies from using the recommended values would be completely negligible for  $^{134}$ Cs (<0.0001%) and very small (<0.4%) in the case of  $^{131}$ I.

**Table 23.** Cumulative table showing the number of laboratories (percent fraction in parentheses) satisfying both the  $|D\%| \le 20$  and  $|E_n| \le 1$  criteria for the three radionuclides, <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I, for any pair of them and for all three simultaneously.

	Number of laboratories					
Nuclides	<sup>137</sup> Cs	<sup>134</sup> Cs	<sup>131</sup> I			
Single	41 (61.2%)	36 (53.7%)	9 (13.4%)			
Double	27 (40.3%)					
Double		9 (13.4%)				
Double	>		5 (7.5%)			
Triple	5 (7.5%)					

It is worth mentioning that one participant (Laboratory 26) informed us on 11 October 2016 that they had by mistake reported the results in a cyclic permutation. This means that the activity result for  $^{137}$ Cs was reported for  $^{131}$ I, the one for  $^{134}$ Cs as that for  $^{137}$ Cs and the one for  $^{131}$ I as that for  $^{134}$ Cs. *Table 24* lists the correct order of results, the original (calculated with the originally reported results) and the new  $D_{\%}$  and  $E_{n}$  numbers and indeed, it is a set of very good results, compatible for all three nuclides.

**Table 24.** Correct order of results for Laboratory 26 and the original and the new  $D_{\%}$  and  $E_n$  numbers.

Nuclide	Reported activity A (Bq)	Reported uncertainty $u_c(A)$ (Bq)	Reported k factor	Original D <sub>%</sub>	New D <sub>%</sub>	Original <i>E</i> <sub>n</sub>	New <i>E</i> <sub>n</sub>
<sup>137</sup> Cs	0.43	0.077	2	150	10.7	3.1	0.54
<sup>134</sup> Cs	0.97	0.19	2	82	15.2	2.3	0.67
<sup>131</sup> I	1.53	0.30	2	-68	13.3	-1.5	0.26

Unfortunately, we could not accept modification of the results reported after the deadline and the originally reported results were kept for Laboratory 26. An important phase of an analysis campaign is reporting of the results. The most common mistake is to use the wrong units or misplace the decimal point, but other mistakes are not excluded. To err is human and the usefulness of such exercise is to discover and correct any mistake.

We are aware of the complexity of the questionnaire that has to be filled by the participants, but it contains information which can help us to understand how environmental laboratories perform their tasks of air monitoring for radioactivity and to deliver reference samples suitable for such measurements and eventually solve problems that some laboratories could encounter.

The answers given to the accompanying questionnaires 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 air filter with the same activity would have been sent to the participating laboratories, a comparison of results would not have reflected the routine measurement conditions. The radioactivity loaded air filters were prepared on air filters provided by the participants by individually spiking gravimetrically a radioactive solution containing the nuclides of interest.

The fact that some problems are always encountered, which can be detected and corrected confirms that there is a permanent need for such comparisons to reaffirm the performance of the laboratories. The "Metrology for radiological early warning networks in Europe" (ENV57-MetroERM) project, coordinated in the frame of the European Metrology Research Programme and funded by the European Association of National Metrology Institutes (EURAMET) and the EU in 2014, aims as well to the harmonisation of the data provided by the radiological early warning networks in Europe (MetroERM, 2016).

The JRC will conduct more ILCs in the future and has developed to that purpose a special automatic dispenser capable of accurately delivering aliquots of a solution as small as 20  $\mu$ L in any given pattern, based on a X-Y coordinate system. This dispenser can be used in the next ILC to be organized on radioactivity measurement in spiked air filters and it will improve on the homogeneity of the final product.

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#### List of abbreviations and definitions

BIPM Bureau International des Poids et Mesures

BEGe Broad Energy Germanium detector

CIEMAT Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas

CCRI(II) Comité Consultatif des Rayonnements Ionisants, Section 2

DDEP Decay Data Evaluation Program

EC European Commission

EMRP European Metrology Research Programme

EU MS European Union Member States

EURATOM European Atomic Energy Community

EURAMET European Association of National Metrology Institutes

EURDEP European Union Radiological Data Exchange Platform

GUM Guide to the Expression of Uncertainty in Measurement

HPGe High-Purity Germanium detector

ICS-REM International Comparison Scheme for Radioactivity Environmental

Monitoring

EC ILC Interlaboratory comparison organised by JRC-IRMM

ISO International Organization for Standardization

JRC Directorate General Joint Research Centre

JRC-IRMM JRC Institute for Reference Materials and Measurements

JRC-ITU JRC Institute for Transuranium Elements

KCRV Key Comparison Reference Value

LSC liquid scintillation counter, liquid scintillation counting

MetroERM Metrology for radiological early warning networks in Europe, ENV57

NIST National Institute of Standards and Technology

NMI National Metrology Institute

SIR Système International de Référence, International Reference System for

radionuclides

UTC Coordinated Universal Time

A activity measured by participating laboratory

 $A_0$  activity reference value, spiked activity

 $D_{\%}$  percentage difference

 $E_n$  performance statistic  $E_n$  number k coverage factor according to GUM

MAD median absolute deviationMDA Minimum Detectable Activity

stdev standard deviation, standard uncertainty in counting alone

$u_c$	combined standard uncertainty according to GUM
U	expanded uncertainty according to GUM
U(A)	expanded uncertainty of laboratory result $(k=2)$
$U(A_0)$	expanded uncertainty of reference value ( $k=2$ )
u(A)	standard uncertainty of laboratory result $(k=1)$
$u(A_0)$	standard uncertainty of reference value ( $k=1$ )
$u_c(A/A_0)$	combined standard uncertainty of ratio $A/A_0$ ( $k=1$ )

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- **Table A-4**. Information on the technical part of the measurements.
- **Table A-5.** Type, material size and form of air filters used by the participants and as reported by the participants.
- **Table A-6**. Sequence of spiked filters and quality control sources prepared for the 2016 ENV57/MetroERM ILC exercise.
- **Table A-7a**. Gamma-ray spectrometric results of the quality control filters; measured values and the corresponding uncertainties.
- **Table A-7b**. Gamma-ray spectrometric results of the quality control filters; their relative deviation from the reference values and their ratio to the reference values.
- **Table A-8a**. Gamma-ray spectrometric results of the quality control point sources; measured values and the corresponding uncertainties.
- **Table A-8b**. Gamma-ray spectrometric results of the quality control point sources; their relative deviation from the reference values and their ratio to the reference values.

- **Table A-9**. Treatment of spiked air filters before measurement by the participants and equipment used for the measurement of the spiked air filters.
- **Table A-10**. Measurement parameters; acquisition time, count rates, MDA.
- **Table A-11**. Coincidence summing correction factors as applied and reported by the participants.

**Table A-12**. Detailed uncertainty budget table showing the uncertainty components as reported by the participating laboratories, for all three radionuclides. In column 13 the combined relative uncertainty as reported by the participants is given. For comparison, in column 15 the combined relative uncertainty calculated as square root of the quadratic sum of the uncertainty components given in the table is given. Finally, in column 16 the combined relative uncertainty as calculated from the uncertainty accompanying the results is given.

#### **ANNEXES**

### Annex 1: List of participating laboratories per country (in alphabetic order)

#### **AUSTRIA**

Mr. Johannes Klimstein Austrian Agency for Health and Food Safety (AGES) Department for Radon and Radio Wieningerstrasse 8 4020 Linz

Mr. Florian Smecka AGES Vienna Radiation Protection & Radioch Spargelfeldstraße 191 1220 Vienna

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#### **CROATIA**

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#### **CZECH REPUBLIC**

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ROMANIA

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Mr. Branko Vodenik Jozef Stefan Institute F2 Jamova cesta 39 1000 Ljubljana SLOVENIA

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TURKEY

Dr. Nurdan Güngör Turkish Atomic Energy Authority Cekmece Nuclear Research and Training Center Radioactivity Measurement Unit Yarımburgaz Mah. Nukleer Arast Merkezi Yolu No 10 34303 Istanbul TURKEY

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#### **Annex 2: Communication to the participants** 2A. Expression of interest

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You have participated at the 2014 ILC-REM exercise and you might be interested in the new MetrCERM in the new MetrCERM in the new MetrCERM in the new MetrCERM of this e-mail as soon as possible and no later than 4 December 2015. In case of too many interested spossible and no later than 4 December 2015. In case of too many interested of our limited human resources, the selection will be based on a combination of geographical and 'first. come, first-served criteria. Come, first-served criteria. Come, first-served criteria. Complete mail address include in your e-mail for the communication.
                                                                                                                                                                                 The ILC will be conducted according to ISO 17043. The reported results will be treated confidentially and then will be presented without association with the name of the corresponding lab (but they will be available to the concerned lab).
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Institute for Reference Materials and Measurements
Unit O4. Standards for Nuclear Safety, Security & Safeguards
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an official position of the European Commission.
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Best regards,
Timotheos Altzitzoglou
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Project Leader
                                                                                                                                                                                 From: ALTZITZOGLOU Timotheos (JRC-GEEL)

Sest: 23 November 2015 12:55

To: "Claudia landstetter@ags.at!" wolfgang.ringer@ages.at!;
Floworder 20 volve 6 in "mishenkow/abaloo.com";
Isneyers@ack.cken.be' "mishenkow/abaloo.com";
Islandro.get.get.ava.laheime.be'; Vabaloingaloo.com";
Isneyers@ack.chemat.es'; Isneyers@ack.chemat.es';
Isneyers@ack.cenat.es'; Isneyers@ack.chemat.es';
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Ref. Ares(2015)5386863 - 26/11/2015

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confirm your interest in participating at the MetroERM ILC by replying to this earlal as soon as possible and no later than 4 booms possible and hockers of our limited human resources, the selection will be based on a combination of first-served criterion and first-cerved criterion of first-served criterion for the responsible/contact person, complete mail address, complete mail address, the communication.
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PS2 In case you know of other laboratories not in the mailing list that might be interested, please feel free to forward this e-mail to them.
                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     Looking forward to hearing from you.
Best regards,
Timotheos Altzitzoglou
From: ALTZIZOGLOU Timotheos (JRC-GEEL)
Sent: 23 November 2015 13:
To: 'jbh eringer@bfs.de' "mbrugem@sckcen.be';
To: 'jbh eringer@bfs.de' "mbrugem@sckcen.be';
To: 'jbh eringer@bfs.de' "mbrugem@sckcen.be';
To: 'lauvent ferrew@kca.fr' catalina.gasco@ciemat.es'
'lauvent ferrew@kca.fr' catalina.gasco@ciemat.es'
'lauvent ferrew@kca.fr' catalina.gasco@ciemat.es'
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'nanian.buraa@envinet.cz' 'jankubancak@envinet.cz'
'nanian.buraa@envinet.daniel'.zapata@ptb.de' 'parick.kessler@ptb.de';
'artur.o.arga@upc.edu' 'josps sempaudupc.edu' 'parick.kessler@ptb.de';
'artur.o.arga@upc.edu' 'josps sempaudupc.edu' 'parick.kessler@ptb.de';
'artur.o.arga@upc.edu' 'josps sempaudupc.edu' 'parick.kessler@ptb.de';
'artur.o.arga@upc.edu' 'ijosps sempaudupc.edu' 'parick.kessler@ptb.de';
'artur.o.arga@upc.edu' 'ijosps sempaudupc.edu' 'parick.kessler@ptb.de';
'artur.pordy@eneait': 'steven.jude@eneait' 'marick.'
'parinen' odefilc@eneait': 'steven.jude@eneait' 'marick.'
'parinetallompl.co.uk' 'laurence.bricc@mpl.co.uk';
'parinetallompl.co.uk' 'laurence.bricc@mpl.co.uk';
'parinetallompl.co.uk' 'laurence.bricc@mpl.co.uk';
'stratos.valakis@pr.ce.europa.eu' 'Monore.arga';
'valerin.allompl.co.uk' 'laurence.europa.eu' 'Monore.arga';
'valerin.allompl.co.uk' 'laurence.europa.eu' 'Monore.arga';
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'valerin.defilc@pr.de.'parid@pr.de.'parick.'
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The views expressed are purely those of the writer and may not any circumstances be regarded as stating an official position of the European Commission. European Commission
Directorate-General Dint Research Centre
Directorate-General Materials and Measurements
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http://cc.ec.europa.eu/dgs/jr.cr.ec.europa.eu
http://rmm.jr.c.ec.europa.eu Timotheos ALTZITZOGLOU Project Leader

The JRC-IRWM under Task 3.2 'Traceability management of airborne radioactivity' [http://earlywarning-emrp.eu/?page\_id=1106] will organise and conduct an interfaboratory comparison (LLC) on the measurement of radionuclides in air filters, similar to the recent ILC-EMR exercise in 2014. Such exercises contribute to the harmonisation of the radiological early warning networks in Europe.

You have initially expressed your interest in participating at the MetroERM ILC corresponding to the corresponding questionnaire or you are a MetroERM partner. It would be appreciated if you could

The ILC will be conducted according to ISO 17043. The reported results will be treated confidentially and treated confidentially and they will be presented without association with the name of the corresponding they will be well as a be consequently as a large to the concerned lab).

The spiked air filters are expected to be dispatched to the participating backarderies by end of January 12016. The participating laboratories will be requested to send their results to Jacks. The participating laboratories will be requested to send their results to Prekrand Force the end laboratories.

The particularity of the ILC is that the spiked filters will be made to the descriftations of each participant on blank filters supplied by them. This time, the filters will be spiked with 175cs, 13ds and 13LK (if the latter than 13LK of the latter supplied by the latter as and 13LK of the latter of the latte

#### 2B. Invitation e-mail and accompanying letter

Please see the attached letter and follow the instructions.

In order to provide you with air filter spiked with the appropriate acity y level, similar to what you measure tourinely, and allow you to measure following your usual procedure and measurement geometry, we kindly sak you to reply to a usudoprocedure and measurement geometry, we kindly sak you to reply to a week nameng an element and the air filters must reach JRC-IRMM before 8 January 2016.

The questionnaire and the air filters must reach JRC-IRMM before 8 January 2016.

The questionnaire can be found at: https://ec.europa.eu/bearsvey/minerilL/ActorERM2016

The password is, MetroERM2016 (case sensitive)

If you have further questions, please contact us, We will be happy to help you.

Emil: JRC-IRMM.REMA\_COMPARISONS@ec.europarem

Looking forward to your participation in this comparison.

Yours sincerely.

Timothese at TITTOGLOU

Feety Meal

European Commission

European Commission

European Commission

Device Leader

European Commission

Control of the funder safety, Security is Safeguards

European Commission

European Commission

European Commission

European Commission

European Commission

European Commission

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The wholese Actor and European Commission.

The wholese Actor and European Commission."

Bef. Ares(2015)5760735 - 11/12/2015 dabrow@paa.gov.pl; hasan.dikmen@taek.gov.tr; Laurent.ferreux@cea.fr; GEEL); DEHOUCK Pieter (JRC-GEEL); ALTZITZOGLOU Timotheos (JRC-GEEL) Invitation to the MetroERM/ENV57 interlaboratory comparison on Cs-137, jacek.kapala@umb.edu.pl; cvetelina.1980@abv.bg; romain.vidal@irsn.fr; petr.rulik@suro.cz; jerzy.mietelski@ifj.edu.pl; Halim.Taskin@taek.gov.tr; beata.silobritiene@aaa.am.lt; Alasdair.morgan@cavendishnuclear.com; elena.simion@anpm.ro; isajenko@clor.waw.pl; Peter.Jovanovic@zvd.si; arno.achatz@ages.at; jerzy.chytla@wsse.rzeszow.pl; fguillen@unex.es; skalinna@gmail.com; florian.smecka@ages.at; fatih.cinar@taek.gov.tr; Jiri.Rada@sujb.cz; nelida.florea@apmar.anpm.ro; fisico.rc@arpacal.it; aier@ptb.de; Valentin.BLIDEANU@cea.fr; DE CORT Marc (JRC-ISPRA); DE LA CALLE GUNTINAS Maria Beatriz (JRC-GEEL); MALO Petya (JRC-GEEL); HULT Mikael (JRC-(branko.vodenik@ijs.si); pieter.kwakman@rivm.nl; konyi@osski.hu; Tomasz. Pliszczynski@ncbj.gov.pl; madruga@ctn.tecnico.ulisboa.pt; laura.porzio@arpa.piemonte.it; claudia.tabacaru@apmis.anpm.ro; arunas.gudelis@ftmc.lt; Jani Turunen (jani.turunen@stuk.fi); maria.nikolaki@eeae.gr; r.totzeva@ncrrp.org; Branko Vodenik Michal Bonczyk (mbonczyk@gig.eu); isabel.serrano@upc.edu; clouvas@eng.auth.gr; Eia.Jakobson@keskkonnaamet.ee; stefano.pegoretti@provincia.tn.it; johan.kastlander@foi.se; giorgio.evangelisti@arpalazio.it; p.sabatini@arpa.umbria.it; Florin.VLAD@apmmm.anpm.ro; rodolfo.gurriaran@irsn.fr; office@rzi-varna.com; giovanni.ferreri@bag.admin.ch; mbruggem@sckcen.be; nina.cernohlawek@bmlfuw.gv.at; clpuscasu@yahoo.com; denis.cindro@ijs.si; spni@dtu.dk; mauro.magnoni@arpa.piemonte.it; rsogni@arpa.emr.it; m.faureragani@arpa.vda.it; cristofaro.arpam@libero.it; r.rusconi@arpalombardia.it; bus@ire-elit.eu; catalina.gasco@ciemat.es; Petr Kovar (pkovar@cmi.cz); (alexander.mauring@nrpa.no); Bredo.Moller@nrpa.no; faig.mutallimov@gmail.com; milli.monitoring@mail.ru andris.abramenkovs@lvgmc.lv; pasts@lvgmc.lv; marydoris.gambin@gov.mt; public.health.lab@gov.mt; zdenek.borecky@suro.cz; leen.verheyen@sckcen.be; marielle.lecomte@ms.etat.lu; S.Somerville@epa.ie; jbieringer@bfs.de; APO@sis.dk; reszkam@op.pl; lena\_iv.1960@abv.bg; racraiova@yahoo.com; tbituh@imi.hr; acaballero@sgl.moh.gov.cy; Sofia.Eriksson@ssm.se; Alexander Mauring Attachments: Subject: To: ö

Your laboratory has expressed interest in participating in the MetroERM/FNV57 interlaboratory comparison on Cs-137, Cs-134 and I-131 measurement in air filters organised by JRC-IRMM.

Dear colleague,

**EUROPEAN COMMISSION** 

Ref. Ares(2015)5760735 - 11/12/2015

DIRECTORATE.GENERAL
DIOIT RESEARCH CENTRE
Directorate D. Institute for Reference Materials and Measun
Standards for Nuclear Safety, Security and Safeguards

Geel, 11 December 2015 JRC.D.4/TA/PM/ARES

Invitation to the MetroERM/ENV57 interlaboratory comparison on Cs-137, Cs-134 and I-131 measurement in air filters

Subject:

The JRC is partner of the European Metrology Research Programme (EMRP) ENV57 project Metrology for Radiological Early Warning Networks in Europe' [http://earlywarning-emrp.eu/]. The JRC-IRMM under Task 3.2 'Traceability management of airborne radioactivity' [http://deanlywanning-emp-eu/?page\_id=1106] and to fulfil deliverables 30.3.4 to D3.3.7 will organise and conduct an interlaboratory comparison (LC) on the measurement of <sup>13</sup>Cs, <sup>14</sup>Cs and <sup>131</sup> in air filters. Such exercises contribute to the harmonisation of the radiological early warning networks in Europe. Your laboratory has expressed interest in participating in the above mentioned interlaboratory comparison organised by JRC-IRMM. In order to provide you with air filter spiked with the appropriate activity level, similar to what you measure routinely, and allow you to measure following your usual procedure and measurement geometry, we kindly ask you to reply to a questionnaire on sampling method, air filter used and measurement conditions. The questionnaire can be found at:

https://ec.europa.eu/eusurvey/runner/ILCMetroERM2016

The password is: MetroERM2016 (case sensitive)

You are also requested to send us two (2) new (unused, blank) air filters to the attention

Mrs. Petya MALO

Standards for Nuclear Safety, Security & Safeguards European Commission
Directorate-General Joint Research Centre
Institute for Reference Materials and Measurements

B-2440 Geel, Belgium

Please, reply to the questionnaire and send the blank air filters as soon as possible, but no later than 8 January 2016.

Retieseweg, B-2440 Geel - Belglum. Telephone: +32 14 571 211 Fax: +32 14 584 273 Telephone: direct line +32 14 571 286, +32 14 571 266

E-mail: JRC-IRMM-REM-COMPARISONS@ec.europa.eu internet: https://ec.europa.eu/irc/institutes/imm/

Once we receive your answers to the questionnaire and the blank filters, we will give you instructions on how to register online for the interlaboratory comparison. The spiked samples with further instructions will be sent to the address specified in the registration mid February 2016. The participating laboratories will be requested to send their results to JRC IRMM via a web-based questionnaire in early March 2016. The report of the exercise is foreseen to be available in June 2016. form. The spiked filters are expected to be dispatched to the participating laboratories by

The reported results will be treated confidentially without disclosing the name of the corresponding lab. It is worth mentioning that this comparison is <u>not</u> in the series of interlaboratory comparisons organised for REM.

This interlaboratory comparison exercise is free of charge for the participating laboratories.

Should you have any question, please feel free to contact us at:

Email: JRC-IRMM-REM-COMPARISONS@ec.europa.eu

Looking forward to your participation in this comparison.

JACTURE Alizizoglou Yours sincerely,

Project Leader

cc

Messrs. Stefan Neumeier (PTB, DE), Valentin Bildeanu (CEA, FR)
Mr. Marc de Cort (JRC ITU)
Messrs. Willy Mondelaers, Pieter Dehouck, Mikael Hult, Ms. Beatriz de la Calle, Petya
Malo (JRC IRMM)

#### 2C. Registration information (e-mail)

Ref. Ares(2016)699985 - 09/02/2016

### From: To:

tbituh@imi.hr, Branko Vodenik (branko.vodenik@ijs.si); clouvas@eng.auth.gr, spni@dtu.dk; giovanni ferreri@bag.admin.ch; nelida.florea@apmar.anpm.ro; clpuscasu@yahoo.com;

elena.simion@anpm.ro; skalinna@gmail.com; rodolfo.gurriaran@irsn.fr; fisico.rc@arpacal.it; s.somerville@epa.ie; m.faureragani@arpa.vda.it; racraiova@yahoo.com; rsogni@arpa.emr.it; cristofaro.arpam@libero.it; radioactivitate@apmis.anpm.ro; nurdan.gungor@taek.gov.tr;

cvetelina.1980@abv.bg; eia.jakobson@keskkonnaamet.ee; zdenek.borecky@suro.cz; florin.vlad@apmmm.anpm.ro; madruga@ctn.tecnico.ulisboa.pt; lena\_iv.1960@abv.bg; florian.smecka@ages.at; Alexander Mauring

(alexander.mauring@nrpa.no); p.sabatini@arpa.umbria.it; romain.vidal@irsn.fr; helena.mala@suro.cz; l.porzio@arpa.piemonte.it; fatih.cinar@taek.gov.tr; Michal Bonczyk (mbonczyk@gig.eu); ronald.overwater@rivm.nl; strahlenschutz.linz@ages.at;

'guillen@unex.es; leen.verheyen@sckcen.be; isabel.serrano@upc.edu; a.suarez@ciemat.es; g.roselli@arpa.puglia.it; arunas.gudelis@ftmc.lt; johan.kastlander@foi.se; jiri.rada@suro.cz; isajenko@clor.waw.pl; erzy.chytla@wsse.rzeszow.pl; stefano.pegoretti@provincia.tn.it; tony.dieudonne@ire.eu; acaballero@sgl.moh.gov.cy;

giorgio.evangelisti@arpalazio.it; bisi.hristova@mail.bg; jacek.kapala@umb.edu.pl; jerzy.mietelski@ifj.edu.pl; m.magnoni@arpa.piemonte.it; r.totzeva@ncrrp.org;

jakub.osko@ncbj.gov.pl; mary-doris.gambin@gov.mt; Bredo.Moller@nrpa.no; konyi@osski.hu; maria.nikolaki@eeae.gr; Peter.Jovanovic@zvd.si; Sofia.Eriksson@ssm.se; APO@sis.dk; alasdair.morgan@cavendishnuclear.com; Jani Turunen R.RUSCONI@ARPALOMBARDIA.IT; jbieringer@bfs.de; (jani.turunen@stuk.fi); beata.silobritiene@aaa.am.lt;

MONDELAERS Willy (JRC-GEEL); HULT Mikael (JRC-GEEL); DEHOUCK Pieter EMRP MetroERM/ENV57 interlaboratory comparison on Cs-137, Cs-134 and I-131 measurement in air filters - REGISTRATION (JRC-GEEL); DE CORT Marc (JRC-ISPRA); VALAKIS Stratos (JRC-ISPRA); Stefan. Neumaier@ptb.de; Valentin. BLIDEANU@cea.fr

Attachments:

instructions\_for\_registration\_MetroERM\_ILC\_2016.docx

Dear colleagues,

Thank you for sending blank air filters to us. We are now in the process of spiking the filters with  $^{137}$ Cs,  $^{136}$ Cs and  $^{134}$  and we expect to have them ready for dispatching to the participating laboratories by February 22, 2016.

The reporting of laboratory results will be done via Internet. Therefore, we kindly ask you to register your laboratory via the following WEB link and send us the signed registration form: <a href="https://web.irc.ec.europa.eu/ilcRegistrationWeb/registration/registration.do?selComparison=1561">https://web.irc.ec.europa.eu/ilcRegistrationWeb/registration/registration.do?selComparison=1561</a>

# The deadline for the registration is 26 February 2016!

definitive if we receive a signed registration form from you. You can send the signed form by e-mail, by Please read carefully the attached instructions for registration. Note that the registration is only fax or by post.

Should you have any question, please feel free to contact us at: JRC-IRMM-REM-COMPARISONS@ec.europa.eu Email:

Looking forward to hearing from you, we remain,

### Ms Petya MALO

Yours sincerely,

Administrative Assistant

**European Commission** 

Directorate-General Joint Research Centre (JRC) Institute for Reference Materials and Measurements (IRMM)

Unit D.4. Standards for Nuclear Safety, Security and Safeguards B-2440 Geel, Belgium Retieseweg 111

phone: +32 14 571 286

https://ec.europa.eu/jrc/en/institutes/irmm

The views expressed are purely those of the writer and may not in any circumstances be regarded as stating an official position of the European Commission

#### 2D. Spiked air filter dispatch (e-mail) and accompanying letter



Bef. Ares(2016)873248 - 19/02/2016



## **EUROPEAN COMMISSION**

DIRECTORATE-GENERAL
JOINT RESEARCH CENTRE
Directorate D - Institute for Reference Materials and Measurements
Standards for Nuclear Safety, Security and Safeguards

Geel, 22 February 2016 JRC.D.4/TA/pm/ARES(2016)

<Title» «First\_Name» «Surname»</p>

«Street\_Name» «Department»

«Zip\_Code» «City»

«Country»

# Participation in the MetroERM/ENV57 interlaboratory comparison on the 'Measurement of $^{137}{\rm Cs}$ $^{134}{\rm Cs}$ and $^{131}{\rm In}$ air filters'

Dear «Title» «Surname»

Thank you for participating in the EC interlaboratory comparison on <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I measurement in air filters. Enclosed, you will find one of your blank filters spiked with  $^{13}$ Cs,  $^{134}$ Cs and  $^{131}$ Jactivity either at a level you normally measure or higher than the threshold you indicated in the

Please confirm the receipt of the sample by e-mail to <u>IRC-IRMM-REM-COMPARISONS@ec.europa.eu.</u> Please check the spiked air filter and in case of any damage report to the above e-mail address.

This parcel contains:

a) Test item: spiked air filter b) This accompanying letter. Please keep this letter; you will need it to report your results.

On the filter you can distinguish the active spots in blue colour. This should make further preparation of the filter for measuring easier. Large filters had to be folded for shipment; the activity is on the inner side.

For the measurement and analysis, you should follow the procedure you routinely use.

Retieseweg 111, B-2440 Geel - Belgium, Telephone: (32-14)571-211, Office: 010 00/067, Telephone: direct line (32-14)571-266.

E-mail: JRC-IRMM-REM-COMPARISONS@ec.europa.eu

In case you do not measure the filter in the inner plastic bag, it is advisable to measure also the inner bag to make sure that no activity from the filter is remaining in the plastic

# The results must be reported for the Reference Date of 1 March 2016 0:00 UTC.

For your calculations, we recommend to use the data provided by the Decay Data Evaluation Project (DDEP) at <a href="http://www.nucleide.org/DDEP">http://www.nucleide.org/DDEP</a> WG/DDEPdata.htm
For your convenience, the most useful data are listed in the table below.

Photons per 100 disintegrations	84.99 (20)	8.342 (15)	15.368 (21)	97.63 (8)	85.47 (9)	8.694 (16)	6.14 (6)	81.2 (5)	7.12(7)
Energy (keV)	661.7	563.2	569.3	604.7	795.9	802.0	284.3	364.5	637.0
Half-life (days)	10975 (29)	754.0 (5)			8.0233 (19)				
Nuclide	137Cs	134Cs		$I_{1\xi 1}$					

The numbers in parentheses are the uncertainties on the last digit of the corresponding values.

In the case of <sup>134</sup>Cs and <sup>131</sup>I you may consider applying corrections for the true coincidence summing effects, if that is necessary. As usually the measurements are relatively long compared to the half-life of <sup>131</sup>I; a correction for the decay during the counting interval might also be necessary. Finally, we recommend you to measure the spiked air filters as soon as possible after reception, as the activity of the relatively short-lived <sup>131</sup>I will be significantly reduced by time.

Reporting of the results: Some via the login page using the following URL: https://web.jrc.ec.europa.eu/ilcReportingWeb

comparison and your laboratory. Your password key will be communicated to you after the deadline for the registration (26 February 2016). To report your results you need a password key which is unique to this interlaboratory

Save your results but also click on the Submit button. Once you have submitted your results and questionnaire, please remember to send us a signed copy by e-mail to JRC. Please note that only submitted results will be taken into account, therefore, do not only IRMM-REM-COMPARISONS@ec.europa.eu. As you have been informed, the description of your analytical and measurement procedures will be collected via questionnaire using the same URL link as for reporting the results. We kindly ask you to answer all relevant questions regarding the procedures you employed for the measurement of the filter sample. Disregard questions which are

not relevant to the methods used in your laboratory. The uncertainty of the result must be reported in the same units as the activity concentration (i.e. Bq).

Please, notice that during the reporting of your results the Cancel button serves as an exit or return button.

The deadline for reporting results and completing the questionnaire is Tuesday, 29 March 2016.

Should vot have any question, please do not hesitate to contact us.

We wish you success with your measurements.

Kind regards.

Timothees ALIZITZOGLOU

Project Leader

#### 2E. Information to participants during the measurement exercise

The views expressed are purely those of the writer and may not in any circumstances be regarded as stating In case you measure the filter alone, it is strongly advisable to measure also the inner plastic bag and add the results to the value measured for the filter itself. Please, give the individual values in the My measurements showed no indication for Cs-137 and Cs-134. Both Cs-137 and Cs-134 do not leave This makes our recommendation to measure the inner plastic bag more important. f you measure the filter in its plastic bag, you need to do nothing else. Jnit D.4. Standards for Nuclear Safety, Security and Safeguards Institute for Reference Materials and Measurements (IRMM) comments at the end of the questionnaire as well. Directorate-General Joint Research Centre (JRC) an official position of the European Commission Good luck with your measurements. petya.malo@ec.europa.eu Administrative Assistant traces in the plastic bag. phone: +32 14 571 286 European Commission Retieseweg 111 B-2440 Geel, Belgium Ms Petya MALO Best regards,

Ref. Ares(2016)1299683 - 15/03/2016 'michel.bruggeman@sckcen.be'; 'p.badalamenti@arpalombardia.it'; hasan.dikmen@taek.gov.tr'; ALTZITZOGLOU Timotheos (JRC-GEEL); MALO Petya (JRC-GEEL); HULT Mikael (JRC-GEEL); DE CORT Marc (JRC-ISPRA); ja.suarez@ciemat.es'; 'johan.kastlander@foi.se'; 'sofia.eriksson@ssm.se'; 's.somerville@epa.ie'; 'rsogni@arpa.emr.it'; 'cristofaro.arpam@libero.it'; 'fisico.rc@arpacal.it'; 'm.faureragani@arpa.vda.it'; 'leen.verheyen@sckcen.be'; 'tony.dieudonne@ire.eu'; 'skalinna@gmail.com'; 'lena\_iv.1960@abv.bg'; 'cvetelina.1980@abv.bg'; 'helena.mala@suro.cz'; 'jiri.rada@suro.cz'; 'spni@dtu.dk'; 'apo@sis.dk'; eia.jakobson@keskkonnaamet.ee'; 'jani.turunen@stuk.fi'; rodolfo.gurriaran@irsn.fr'; 'romain.vidal@irsn.fr'; 'jbieringer@bfs.de'; peter.jovanovic@zvd.si'; 'fguillen@unex.es'; 'isabel.serrano@upc.edu'; beata.silobritiene@aaa.am.lt'; 'marielle.lecomte@ms.etat.lu'; 'mary-'jerzy.mietelski@ifj.edu.pl'; 'jakub.osko@ncbj.gov.pl'; 'madruga@ctn.tecnico.ulisboa.pt'; 'nelida.florea@apmar.anpm.ro'; nihal.kaya@taek.gov.tr'; 'alasdair.morgan@cavendishnuclear.com' clouvas@eng.auth.gr'; 'maria.nikolaki@eeae.gr'; 'konyi@osski.hu'; giorgio.evangelisti@arpalazio.it'; 'm.magnoni@arpa.piemonte.it'; giovanni.ferreri@bag.admin.ch'; 'nurdan.gungor@taek.gov.tr'; bisi.hristova@mail.bg'; 'r.totzeva@ncrrp.org'; 'tbituh@imi.hr'; stefano.pegoretti@provincia.tn.it'; 'g.roselli@arpa.puglia.it'; 'clpuscasu@yahoo.com'; 'radioactivitate@apmis.anpm.ro'; p.sabatini@arpa.umbria.it'; 'l.porzio@arpa.piemonte.it'; Alexander.Mauring@nrpa.no'; 'bredo.moller@nrpa.no'; acaballero@sgl.moh.gov.cy'; 'zdenek.borecky@suro.cz'; 'florin.vlad@apmmm.anpm.ro'; 'branko.vodenik@ijs.si'; florian.smecka@ages.at'; 'strahlenschutz.linz@ages.at'; r.rusconi@arpalombardia.it'; 'arunas.gudelis@ftmc.lt'; doris.gambin@gov.mt'; 'ronald.overwater@rivm.nl'; 'mbonczyk@gig.eu'; 'jerzy.chytla@wsse.rzeszow.pl'; 'isajenko@clor.waw.pl'; 'jacek.kapala@umb.edu.pl'; racraiova@yahoo.com'; 'elena.simion@anpm.ro'; mportant information for the MetroERM ILC IRC-IRMM-REM-COMPARISONS Dear colleague, Subject: From: Sent: To:

As found by other participants and confirmed by our own measurements, part of the I-131 activity can be found in the inner plastic bag protecting your spiked filter during transport.

In assured some of our control calced filters (a 70 mm) and found that 3.5 % of the activity was left in

You are probably in the course of measuring and evaluating the results of the spiked air filter for the

I measured some of our control spiked filters (ø 70 mm) and found that 3-5 % of the activity was left in the plastic bag. For larger filters the fraction can be even 10% of the spiked activity, as reported by

#### 2F. Reporting results

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Ref. Ares(2016)1168938 - 08/03/2016 ALTZITZGGLOU Timorheos (RC-GEEL)
MALO Petya (IRC-GEE)
MALO Petya (IRC-GEEL)
Filter Set (IRC-GEEL)
MacroERM/LEVST ILC on Cs-137, Cs-134 and I-131 measurement in air
filters – submission of results
MetroERM\_2016\_ILC\_instructions\_submission\_results.pdf - Both the activity results and their uncertainties should be reported in the unit **becquerel (Bq)** (not The online system is now open to submit your results of the measurement of Cs-137, Cs-134 and I-131 in air filters for the ENV57/MetroERM ILC. Finally, the deadline for submitting your results and the questionnaire is Tuesday, 29 March 2016. You can save your results and answers to the questionnaire but they will be ready for submission Should you face any problem with reporting, please feel free to contact us at JRC-IRMM-REM-Once logged in, you can report your results (ignore the column on the technique) and fill in the corresponding questionnaire (see attached file). appropriate link, sign them and send them by e-mail (<u>IRC-IRMM-REM-COMPARISONS@ec.europa.eu</u>), by fax (+32 14 584 273) or by post, to finalise your results After you submit your results and questionnaire, please print them both by clicking at the To access the online system, go to the login page using the URL https://web\_irc.ec\_europa.eu/ilcReportingWeb and fill in your password key. JRC-IRMM-REM-COMPARISONS 07 March 2016 17:26 Petya Malo Logistics Assistant For your laboratory, the password key is XXXXXXXXXXX The reference date is 01 March 2016, 00:00 UTC. Please do not report your results through Excel. only when you save + validate. Timotheos ALTZITZOGLOU We remind you that: Dear colleague, Attachments: Project Leader

#### 2G. Communication of preliminary results to the participants

Ref. Ares(2016)5457470 - 20/09/2016

Geel, 20 September 2016

**Subject:** Preliminary results of the 2016 EMRP ENV57/MetroERM interlaboratory comparison (ILC) and new Laboratory Code

Dear «Title» «Surname»,

Thank you for your participation at the 2016 EMRP ENV57/MetroERM interlaboratory comparison (ILC) for the measurement of  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I in air filters. Currently, we are working on the preparation of the final report. However, for your information we are sending you a preliminary evaluation of the results of this comparison in the attached PDF.

Anonymity being a requirement, each laboratory was assigned a new code number (different than the original one you received with the spiked filter), which is kept **confidential**.

The new code number for your laboratory is «New\_Lab\_Code».

In this preliminary report, the results are presented for all three radionuclides measured, i.e.  $^{137}$ Cs,  $^{134}$ Cs and  $^{131}$ I, in the form of the ratio of the laboratory reported value to the reference value and the E<sub>n</sub> number. More details you can find in the report itself. You can identify your laboratory in the graphs and find out how your reported results compare to the reference values

The final report of this comparison exercise is foreseen to be available later in 2016. If you have any further questions with respect to this comparison, please feel free to contact me at <a href="mailto:JRC-IRMM-REM-COMPARISONS@ec.europa.eu">JRC-IRMM-REM-COMPARISONS@ec.europa.eu</a>.

Best regards,

Timos Altzitzoglou

#### **Timotheos ALTZITZOGLOU**

Project Leader

**Petya Malo** 

Logistics Assistant



#### **European Commission**

Joint Research Centre (JRC) Standards for Nuclear Safety, Security & Safeguards

Retieseweg 111 B-2440 Geel, Belgium +32 14 571266 JRC-IRMM-REM-COMPARISONS@ec.europa.eu

EU Science Hub: https://ec.europa.eu/jrc

Connect with us on:



Ref. Ares(2016)5457470 - 20/09/2016



**EUROPEAN COMMISSION** 

T. Altzitzoglou Contact:

Timotheos.altzitzoglou@ec.europa.eu Tel. +32 14 571266

Geel, September 19, 2016

Subject: Report of preliminary results of the 2016 ENV57/MetroERM Interlaboratory

In December 2015, the European Commission Directorate-General Joint Research Centre, Institute for Reference Materials and Measurements (RC-IRMM) invited laboratories routinely measuring radioactivity in exposed air filters to participate the 2016 ENRP ENVST/MetroERM interlaboratory comparison (ILC). The ILC was organized as part of the JRC contribution to the European Metrology Research Project (ENRP) ENVST/MetroERM "Metrology for radiological early warming networks in Europe". In fact, this work covers Deliverables D3.2.1 to D3.2.7 of Task 3.3. "Traceability management for airborne radioactivity".

The aim of this ILC (or Proficiency Test (PT)) exercise was to quantify airborne radioactivity field station measurement performance. The participating laboratories provided blank air filters of the type routinely use, which the IRC spiked provided blank with known amount of radioactivity individually for each laboratory and returned to them on February 2.2 and 3.3, 2016. For this exercise, the filters were spiked with \$17Cs, \$14Cs and \$131 with activities in the range of several tenths of Bq to a few Bq, close to those the laboratories routinely measure.

about sample collection, measurement and analysis, as they are routinely carried out by the participating laboratories. The deadline to report the measurement results and submit the answers to the second questionnaire was set to March 29, 2016. The ILC was accompanied by two relevant questionnaires to collect information

in cotal, b. laboratories from 29 countries monitoring radioactivity in the monitoring radioactivity in the notation to the presented in this ILC and the preliminary results are presented in this notation to the presented in this ILC.

note in two ways:

As the ratio, R, of the reported by the participating laboratory activity value  $R = \frac{A}{A_0}$ to the reference value, according to the formula

where

is the activity value reported by the participating laboratory is the assigned activity reference value

The ratios are graphically presented in Figure 1 for  $^{137}{\rm Cs}$ , Figure 3 for  $^{134}{\rm Cs}$  and Figure 5 for  $^{131}{\rm L}$ 

Reading the ratio from the provided graphs and since each participant knows his/her reported results, helgshe can easily calculate to a good accuracy the activity reference values,  $A_0$  for his/her particular sample.

1 of 6

As the E, number, according to the formula:

$$E_n = \frac{A - A_0}{\sqrt{U(A)^2 + U(A_0)^2}}$$

where

is the activity value reported by the participating laboratory is the assigned activity reference value is the based uncertainty of a participant's result (k=2) is the expanded uncertainty of the assigned reference value (k=2) is the expanded uncertainty of the assigned reference value (k=2)

The  $E_n$  number takes the expanded uncertainty of the reported results and that of the reference values into account in the analysis. The  $E_n$  numbers are interpreted as follows:

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

If  $|E_n| > 1$ , the laboratory values differ significantly from the reference values, the sources of deviation should be investigated and corrected, "warning

signal";

If  $|E_n| > 1.5$ , there is an urgent need to investigate and find the sources of the large deviation, "action signal" The E<sub>n</sub> numbers sorted in ascending order are graphically presented in Figure 2 for <sup>137</sup>Cs, Figure 4 for <sup>134</sup>Cs and Figure 6 for <sup>131</sup>I.

Laboratory Code number (which you can find in the accompanying e-mail); using the Laboratory Code you can consult the graphs below to obtain your particular scores. To preserve anonymity, each participating laboratory has been given a unique

In the following pages you can find the graphs of the ratios and the  $E_n$  numbers, progether with some comments, for all three radionuclides  $^{13}\text{Ce}_s$ ,  $^{13}\text{Ce}_s$  and  $^{13}\text{I}_s$ , that were the subject of this ILC. A full report is in preparation, including details for the filter spiking and the analysis of the results reported by the participants.

The Reference Date for all results is: 1-Mar-2016 0:00 UTC

Thank you once more for participating at this 2016 EMRP ENV57/MetroERM interlaboratory comparison.

Timos Altritzoglou Yours sincerely,

2 of 6

# 2016 ENV57/MetroERM Interlaboratory Comparison Report of preliminary results

Nuclide: 137Cs

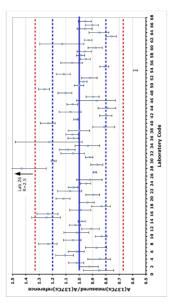
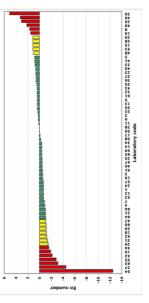


Fig. 1. Ratio of <sup>137</sup>Cs activity per filter as reported by the participating laboratory over the spiked activity on the filter (JRC reference value) sorted in ascending order of the Laboratory Code. Red dashed lines indicate the 33% limit and blue dashed lines the 20% limit from the JRC reference value.



 ${\bf Fig.}$  2.  $E_n$  numbers for  $^{137}{\rm Cs}$  plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

The majority of the laboratories reported reliable measurement results; only 3 out of 67 participants reported values with a percentage difference larger than ±33% of the reference value. Even with a stricter criterion, only 11 laboratories reported values with devlation larger than 20% from the reference value. Furthermore, 2 laboratories passed successfully the criterion of the compatibility test based on Ennumbers, 12 laboratories reported incompatible results with [En]-1 and 13 with [En]-1.5. The performance of the laboratories is very similar compared to the 2014 and 2003 EC interlaboratory comparisons on the measurement of 137Cs in air filters.

Nuclide: <sup>134</sup>Cs

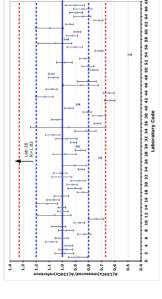
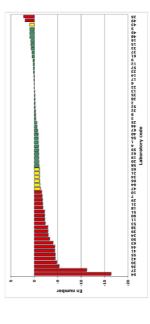


Fig. 3. Ratio of <sup>134</sup>Cs activity per filter as reported by the participating laboratory over the spiked activity on the filter (IAC reference value) sorted in ascending order of the Laboratory Code. Red dashed lines indicate the 33% limit and blue dashed lines the 20% limit from the JRC reference value.



**Fig. 4.**  $E_n$  numbers for  $^{134}$ Cs plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal.

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3 of 6

6 of 6

Most of the laboratories reported reliable measurement results; only 4 out of 67 participants reported values with a percentage difference larger than ±33% of the reference value. Even with a stricter criterion, only 11 laboratories reported values with deviation larger than 20% from the reference value. For the E<sub>n</sub> number compatibility test, 36 laboratories fulfilled the criterion, 7 laboratories reported incompatible results with [E<sub>n</sub>] > 1 and 24 with [E<sub>n</sub>] > 1.5...

## Nuclide: 131

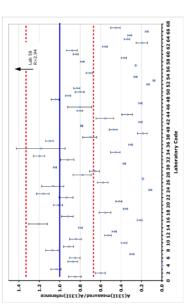
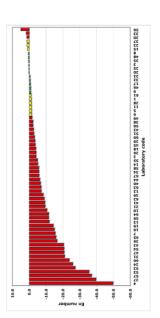


Fig. 5. Ratio of <sup>131</sup> activity per filter as reported by the participating laboratory over the spiked activity on the filter (JRC reference value) sorted in ascending order of the Laboratory Code. Red dashed lines indicate the 33% limit from the JRC reference the Laboratory Code.



**Fig. 6**.  $E_n$  numbers for  $^{13}I$  plotted in ascending order. Green color indicates compatible results, yellow indicates warning signal and red indicates action signal. 5 of 6

In the case of <sup>131</sup>f, 32 of the 67 participating laboratories reported measurement results within the 33% limit from the reference value and 35 reported values with a percentage difference larger than ±33% from the reference value. Furthermore, only 11 laboratories passed successfully the criterion of the compatibility test based on E<sub>n</sub> numbers; 9 laboratories reported incompatible results with |E<sub>n</sub>|>1.1 and 47 with |E<sub>n</sub>|>1.5. The majority of the laboratories reported lower activity values than the nominal reference activity value.

Analyzing the quality control air filters and point sources that were prepared together with the spiked air filters sent to the participating aboratories, it was found that the amount of activity measured on the air filters was lower than expected. As iodine is volatile, and despite the precautions taken, one can think that a variable amount of activity was lost from each spiked air filter by evaporation during the preparation phase. In addition, as initially indicated by some participants, some activity was also transferred to the plastic bag containing the air filter. Although the relative amount of that activity was not high, measuring the air filter without its plastic bag resulted in lower activity values.

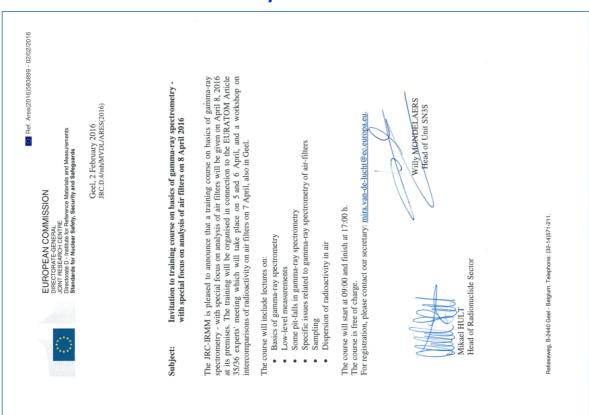
Because of these two observations, the results from the measurement of <sup>131</sup>1 in the air filters are non-conclusive. Indeed, those laboratories that reported correct values for <sup>131</sup>1, it is safe to say that they received air filters with the integral <u>spliked activity</u> and that they <u>performed correctly</u> the measurements. For those laboratories that reported significantly lower activity values for <sup>131</sup>1 than expected, it could mean that

- 1. the laboratory received a spiked air filter with  $\overline{lower}$   $^{131}I$  activity than the nominal, but performed the measurement  $\overline{correctly},$  or
- the laboratory received a spiked air filter with the <u>nominal</u> <sup>131</sup>I activity, but did not <u>perform correctly</u> the measurement (including any eventual sample preparation), or
- the laboratory received a spiked air filter with lower <sup>131</sup>1 activity than nominal, and did not perform correctly the measurement (including eventual sample preparation).

the

We further investigate the case and more details will be given in the final report on the 2016 ENV57/MetroERM Interlaboratory Comparison.

#### 2H. Invitation to the Workshop



Ref. Ares(2016)583899 - 02/02/2016 The JRC-JRMM is pleased to announce that the ICS-REM and MetroERM workshop on the measurement of radionuclides on air filters to be held on 7 April 2016 at its premises. The workshop will be organised in connection to the EURATOM Article 35/36 experts' The most recent laboratory intercomparison (measurement of <sup>137</sup>Cs on air filters) performed under the scope of EURATOM Treaty Article 35 will be presented. Also the intercomparison performed under the EMRP-project (European Metrological Research Programme) MetroERM<sup>1</sup> involving the measurement of <sup>131</sup>L, <sup>134</sup>Cs and <sup>137</sup>Cs on air filters The workshop will include (i) background information, (ii) description of the sample preparation, (iii) presentation of the results, (iv) discussions on specific pit-falls and Willy-MONDELAERS Invitation to ICS-REM and MetroERM air filter workshop on 7 April 2016 Geel, 2 February 2016 JRC.D.4/mb/MVDL/ARES(2016) Head of Unit SN3S The workshop will start at 09:00 and finish at 17:00 h.
The workshop is free of charge.
For registration, please contact our secretary: mira.van-de-lucht@ec.europa.eu. Metrology for early warning radiological networks in Europe; project number ENV57 DIRECTORATE GENERAL

OUNT RESEARCH CENTRE

Directorate D - Institute for Reference Materials and Measurn
Standards for Nuclear Safety, Security and Safeguards meeting which will take place on 5 and 6 April, also in Geel **EUROPEAN COMMISSION** Retieseweg, B-2440 Geel - Belgium. Telephone: (32-14)571-211. (v) presentations by some participants. Timotheos ALTZITZOGLOU will be described Subject:

Bef. Ares(2016)910617 - 22/02/2016

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mbruggem@sckcen.be; hasan.dikmen@taek.gov.tr; ja.suarez@ciemat.es; Contribution to the Workshop on the 2014 EC interlaboratory comparison skalinna@gmail.com; strahlenschutz.linz@ages.at; arno.achatz@ages.at; MONDELAERS Willy (JRC-GEEL); DE CORT Marc (JRC-ISPRA); HULT Mikael and the MetroERM/ENV57 interlaboratory comparison on radioactivity helena.mala@suro.cz; petr.rulik@suro.cz; cvetelina.1980@abv.bg; romain.vidal@irsn.fr; isabel.serrano@upc.edu; clouvas@eng.auth.gr; tbituh@imi.hr; acaballero@sgl.moh.gov.cy; florian.smecka@ages.at; radioactivitate@apmis.anpm.ro; claudia.tabacaru@apmis.anpm.ro; madruga@ctn.tecnico.ulisboa.pt; marielle.lecomte@ms.etat.lu; jiri.rada@suro.cz; fisico.rc@arpacal.it; bisi.hristova@mail.bg; Alexander.Mauring@nrpa.no; florin.vlad@apmmm.anpm.ro; .porzio@arpa.piemonte.it; laura.porzio@arpa.piemonte.it; jacek.kapala@umb.edu.pl; nelida.florea@apmar.anpm.ro; jerzy.chytla@wsse.rzeszow.pl; jerzy.mietelski@ifj.edu.pl; catalina.gasco@ciemat.es; mary-doris.gambin@gov.mt; nurdan.gungor@taek.gov.tr; Halim.Taskin@taek.gov.tr; giovanni.ferreri@bag.admin.ch; office@rzi-varna.com; eia.jakobson@keskkonnaamet.ee; mbonczyk@gig.eu; public.health.lab@gov.mt; g.roselli@arpa.puglia.it jbieringer@bfs.de; fguillen@unex.es; apo@sis.dk; lena\_iv.1960@abv.bg; rodolfo.gurriaran@irsn.fr; 3redo.Moller@nrpa.no; s.somerville@epa.ie; (JRC-GEEL); stefan.neumaier@ptb.de

Subject:

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measurement in air filters ague, aiready been informed about the Workshop on the 2014 EC interlaboratory comp

Dear colleague,

You have already been informed about the Workshop on the 2014 EC interlaboratory comparison and the MetroERM/ENV57 interlaboratory comparison on radioactivity measurement in air filters, to be held at EC-JRC-IRMM in Geel on April 7, 2016.

During the Workshop, we will present you with the details of these two ILCs and we will show and discuss

As we see this Workshop as a forum to discuss the measurement of radioactivity in air monitoring filters and to exchange indeas and experiences we invite von to actively narticinate by presenting

and to exchange ideas and experiences, we invite you to actively participate by presenting the method you use to prepare the filters for measurement. the way you perform the measurements in your laboratory

which geometry you use for the measurements and how you calibrate your detectors how you perform the analysis and the calculations and by sharing your experiences and discussing any problem encountered.

We encourage you to send an e-mail to <u>timotheos altrizoglou@ec europa.eu</u> with a few words on what you would like to present and a tentative title. Depending on the number of presentations, they can be from 5 to 15 minutes each. The sooner we get your expression of interest, the sooner we will be able to

Looking forward to hearing from you, Best regards,

finalise the Workshop agenda.

Trince Alzibrogou

Project Leader

The views expressed are purely those of the writer and may not in any circumstances be regarded as stating an official position of the European Commission.

Dr. THNOS ATZITZOGLOU

Commission and C



#### **OUR MISSION**

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Working in close cooperation with policy Directorates-General, the JRC addresses key societal challenges while stimulating innovation through developing new methods, tools and standards, and sharing its know-how with the Member States, the scientific community and international partners.

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# Measurement on simulated airborne particulates – <sup>137</sup>Cs in air filters

Workshop for the participants of the 2014 EC and the 2016 EVN57 – MetroERM measurement comparisons

Programme

Thursday 7 April 2016

Joint Research Centre Institute for Reference Materials and Measurements

> Joint Research Centre

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### Measurement on simulated airborne particulates - $^{137}$ Cs in air filters JRC-IRMM - 7 April 2016

08:15 Pick up at the hotel, Geel

08:45 Arrival at the JRC-IRMM entrance

09:00 Welcome - Introduction - Overview Timos Altzitzoglou (JRC)

09:15 About EURDEP / REM Marc De Cort (JRC)

09:45 Scope and organization of the measurement comparisons

Borbala Maté & Timos Altzitzoglou

(JRC)

10:05 Radionuclide standardization and filter spiking
Timos Altzitzoglou (JRC)

10:30 Coffee break

11:00 Filter measurement and Quality Control

Timos Altzitzoglou (JRC)

11:30 Results-Performance-Conclusions: REM ILC

Borbala Maté (JRC)

11:50 Results-Performance-Conclusions: MetroERM ILC Timos Altzitzoglou (JRC)

12:15 Discussion

12:45 Lunch

14:00 Radioactivity Air Monitoring Stefan Neumaier (PTB)

14:30 Contribution by participants – Short presentations by:

Sven Poul Nielsen (DTU, DK),

Bredo Møller (NRPA, NO),

Tereza Jezková (SÚRO, CZ),

Asser Nyander Poulsen (SIS, DK),

Paola Sabatini (ARPA Umbria, IT),

Lidia Silva (C2TN, PT),

Giovanni Ferreri (FOPH, CH),

Michèle Pallmer (Division de la Radioprotection, LU)

15:45 Coffee break

16:00 Laboratory visit

17:00 End

17:15 Departure to the hotel, Geel



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# Basics of gamma-ray spectrometry and analysis of air filters

Training - Programme

Friday 8 April 2016

Joint Research Centre Institute for Reference Materials and Measurements

> Joint Research Centre

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#### Basics of gamma-ray spectrometry and analysis of air filters

### JRC-IRMM – 8 April 2016

08:15	Pick up	at the	hotel,	Geel
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08:45 Arrival at the JRC-IRMM

09:00 Welcome and introduction to the SN3S-unit Wim Mondelaers

09:15 The Radionuclide Metrology Sector

09:30 Legislation and BSS: Marc de Cort, JRC

09:45 The basic equation for gamma-ray spectrometry
Mikael Hult, JRC

10:15 Coffee break

10:45 Full Energy Peak Efficiency Sean Collins, NPL

11:15 Coincidence summing Tim Vidmar, SCK-CEN

Mikael Hult, JRC

11:45 Instrumentation and Low-level gamma-

12:15 Lunch

13:45 Gamma-ray spectrometry of air-filters

14:15 Quality System and LIMS Michel Bruggeman, SCK-CEN 14:45 Aerosol physics
Christina Isaxon, Lund University

15:30 Coffee break

16:00 Dispersion of radioactivity in the atmosphere

Johan Camps, SCK-CEN

16:45 Discussion and evaluation Mikael Hult, JRC

17:00 End

17:15 Departure to the hotel, Geel

Annex 3: First questionnaire sent to the participants to collect information on air filters and sampling

*1.2.1 Organisation Please write the full title in English.	1.2.2 Organisation department	* 1.2.3 Organisation address	*1.2.4 Zip code	*1.2.5 City	*1.2.6 Country	*1.2.7 Mailing address (to which the filter will be sent back):  Please note that the courrier company will not deliver to a post box address.  Same as organisation address.  Different than organisation address.	*1.2.8 Please provide the complete mailing address here:		Monitoring of nuclear facilities  Fissile material control or safeguards  Governmental laboratory  University laboratory
Questionnaire for participants in ILC MetroERM 2016	Fields marked with * are mandatory.	1 Organisation and contact details	1.1 Person details 1.1.1 Title  Mr	<ul><li>Mrs</li><li>Dr</li><li>*1.1.2 First name</li></ul>	* 1.1.3 Familiy name	*1.1.4 E-mail address	*1.1.5 Gender  Male  Female	1.1.6 Telephone (with country code) 1.1.7 Fax (with country code)	1.2 Organisation details

	* 2.1 Sumpling period:  * 3.1.5 Please define here:  * 4.1.5 Please define here:	3.2. Air filters: define the following information on the air filter material used  4.3.2.2 Type and material of the air filter:  a.3.2.3 Shape of the air filter:  a.3.2.4 Size of the air filter:  a.3.2.5 Blank weight (areal density):  mg/cm2  4.3.2.7 Typical acriss of deposit collected:  mg/cm2  4.3.2.7 Typical activity level measured for:  mg/cm2  4.3.2.8 Would you like to get the filters spiked with coloured solution in order to optically identify the active spoots?  active spoots?  active spoots?  active spoots?  by yes  no
4.3.2.9 Specify the requirements for the spiked filters (more than one choice is possible):  (1.6. How many measurements does your laboratory perform per year?  (2.5.100)  3 3 4 4		

*3.2.10 Specify any required pattern for spiking the activity on the air filter:	*4.2 Please specify here:
*3.2.11 Specify any required margin to be left blank while spiking the air filter:	4.3 Comments on sampler, filter, sampling, measurements, etc.:
*3.2.12 Specify any other requirements:	4.4 Comments on the questionnaire:
3.3 Sample preparation and measurements	
*3.3.1 What type of detector is used for the measurements?  Ge(L) detector  Hoge detector  BEGe detector  Well type detector  Nal(T) detector  Other	
*3.3.2 Please specify here:	
*3.3.3 Describe briefly the measurement methods, including sample preparation if any:	
*3.3.4 What is the typical detection limit of your routine method for:  (Insert "no" if you do not measure this nuclide)  Cs-137  Cs-134  I-131	
4 Additional information	
4.1 Which radionuclides do you determine routinely in air filters?  Be-7 K40 Cs-134 Cs-137 I I-131 Pb-210 Sr-90  other	
5	9

## Annex 4: Responses of the participants to the questionnaire for collecting information on air filters and sampling

To determine the % ratios in *Table A-1* and the subsequent tables, the 67 participants were taken as 100%.

**Table A-1**. General information on the 2016 MetroERM ILC participating laboratories.

Question	Number of answers	Ratio (%)		
"What is the type of your laboratory (more tha	n one choice is po	ossible)?"		
Research and development	23	34		
Radioactivity in the environment	63	94		
Monitoring of nuclear facilities	15	22		
Fissile material control or safeguards	2	3		
Governmental laboratory	32	48		
University laboratory	6	9		
Other	2	3		
"Is your laboratory certified, accredited or authorised (more than one choice is possible)?"				
Certified (ISO 9000) and Accredited (ISO 17025)	10	15		
Certified (ISO 9000)	2	3		
Accredited according to ISO 17025	30	45		
Authorised	13	19		

**Table A-2**. Air filter size categories.

Size group	Size of air filter	Number of air filters	Ratio (%)
small	Ø 4.7-13 cm	29	44
intermediate	18x20-23.5x28 cm	11	17
large	24.6x41-60x70 cm	26	39

**Table A-3**. Information on the air sampling methods.

Question	Answer				
	Range	Average	Median		
Small size group					
Total volume of air sampled/filter (m³)	0.1-14000	1340	127.5		
Sampling period (h)	3.5-300	75	24		
Sampling frequency	Sampling frequency Mostly on daily basis				
Intermed	Intermediate size group				
Total volume of air sampled/filter (m³)	18-30000	13029	11000		
Sampling period (h)	2-720	206	168		
Sampling frequency	quency Mostly on a weekly basis				
Large	size group				
Total volume of air sampled/filter (m³)	600-300000	105733	100000		
Sampling period (h)	84-720	193	168		
Sampling frequency Mostly on a weekly basis			pasis		

**Table A-4**. Information on the technical part of the measurements.

Question	Number of answers	Ratio (%)			
"How many measurements ( <sup>137</sup> Cs in air filters) does your laboratory perform per year?"					
< 25	9	13			
25-100	21	31			
> 100	37	55			
"Would you like to get the filters spiked with coloured solution in order to optically identify the active spots?"					
Yes	60	90			
No	7	10			

"Specify the requirements for the spiked filters:"						
No special requirement	39	58				
Pattern - requests for homogeneous distribution	15	22				
Margin - active area	15	22				
Other - air filter's side to spike, special folding, sealed bag/holder requirements	3	4.5				
"What type of detector is used routinely for the determination of <sup>137</sup> Cs in air filters?"						
Ge(Li) detector	2	3				
HPGe detector	58	87				
BEGe detector	13	19				
Well type detector	1	1.5				
NaI(TI) detector	0	0				
Other	0	0				
"Do you determine any additional radionuclio air filters?"	"Do you determine any additional radionuclide(s) besides <sup>137</sup> Cs in the air filters?"					
Yes	63	94				
No	4	6				

## Annex 5: Type, material, size and form of the air filters used by the participants

**Table A-5.** Type, material size and form of air filters used by the participants and as reported by the participants.

Lab Code	Type and material of the air filter	Supplier	Filter size (cm)
1	cotton filters (high-quality cotton linters	Whatman	ø 4.7
2	HB5773	Hollingsworth & Vose	11x16
3	The Filters are made of refined pulp and linters with over 98% alphacellulose content	Sartorius	ø 5.5
4	PVC		60x60
5	Polypropylen filter type G-3	PTI Physik-Technik- Innovation	44x44
6	Polypropylen-Filter Typ G-3	PTI Physik-Technik- Innovation	60×60
7	glass fibre	F&J Specialty Products, Inc	ø 4.7
8	Model: No.880052.01-4	micropo	ø 10.2
9	Petrianov Filter FPP-15-1,5	PTI Physik-Technik- Innovation	44x44
10	Glass fiber. Model no: FP102M2	F&J Specialty Products, Inc	ø 10.2
11	Cellulose	Bernard Dumas, France	ø 11.5
12	Glass Microfibre Filters GF/A NO. 1820-866	Whatman	20x25
13	GF/A glass microfibre filter	Whatman	45x56
14	Petrianov filters type FPP-15-1.5	PTI Physik-Technik- Innovation	44x44
15	Petrianov FPP 15-1.5, PVC	Russian made	44x44
16	Glass microfiber filters, Grade GF/A	Whatman	30x23
17	Polymer	VF, a.s., Cerna Hora, Czech Republic	30x23
18	Glass microfibre, type GF/A 55 mm ø	Whatman	ø 5.5
19	glass fibre	F&J Specialty Products, Inc	ø 4.7
20	Petrianova FPP	CLOR	40x40
21	FP-47m , glass fiber filter	F&J Specialty Products, Inc	ø 4.7
22	Polypropylene	PTI Physik-Technik- Innovation	45x56
23	Part no. FP2063-47, Hydrophobic borosilicate glass microfibre with acrylic resin binder	Hi-Q Environmental Products, USA	ø 4.7

Lab Code	Type and material of the air filter	Supplier	Filter size (cm)
24	AP4004705 - glass fiber filters - with a 0.7 µm pore size	Millipore	ø 4.7
25	Polypropylene	3M	ø 12.7
26	Cellulose	PTI Physik-Technik- Innovation	44x44
27	Model NO. FP – 47	F&J Specialty Products, Inc	ø 4.7
28	Whatman GF/A Glassfibre	Whatman	45x56
29	Glass Microfibre Filters GF/A	Whatman	20x25
30	Petrianov fabric FPP-15-1.5 (Postchlorinated Polyvinylchloride PCV Fiber)		44x44
31	Whatman Glass Microfiber Filter GF/A	Whatman	45x56
32	Polypropylene (3M)		40x40
33	Chlorinated vinyl polychloride filter	Sorbent, Russian Federation	44x44
34	Glass fiber paper MODEL NO. FP-47M	F&J Specialty Products, Inc	ø 4.7
35	Filter Paper (Whatman)	Whatman	ø 4.7
36	Glass Fibre (Circular Plastic Holder)		ø 4.7
37	FPM 1545	AS ESFIL TEHNO, Estonia	45x56
38	ΦΠΠ-15-1,5 (FPP-15-1,5), a layer of ultra-thin fibers with an average size of 1.5 microns, deposited on the gauze	JSC Sorbent	60x70
39	Glass Fibre GF/A (Whatman)	Whatman	30x23
40	glass fiber filters (steel impactors)		ø 4.7
41	Fiberglass	TECNASA S.A.	44x44
42	FP-47m glass fiber filter	F&J Specialty Products, Inc	ø 4.7
43	Glass fibre sheets	Munktell	20x25
44	GF/A Cat. No 1820-866	Whatman	20x25
45	FPP-15	Esfil Tehno AS Sillamae, Estonia	87x74
46	Filter, polypropylene; type G3, PTI	PTI Physik-Technik- Innovation	45x56
47	Glass Fibre Filter	GELMAN SCIENCES	ø 4.7
48	Petryanov FPP_15_1.5	PTI Physik-Technik- Innovation	45x56
49	Petryanov's filters FPP-15-1.5		40x40
50	3M polypropylene	3m	45x56
51	Petrianov filter type FPP-15-1.5	(Russia)	44x44

Lab Code	Type and material of the air filter	Supplier	Filter size (cm)
52	GF6 Glass Fiber Filter	Whatman	ø 4.7
53	0.8 microns, nitrocellulose	Millipore	ø 4.7
54	MODEL FP-4.0 M	F&J Specialty Products	ø 10.2
55	PETRIANOV FILTER, FPP-15-1.5	Central Laboratory for Radiological Protection	40x40
56	type FPM 1530	ESFIL TEHNO AS, Estonia	30x23
57	quartz microfibre filters	FILTER -LAB	ø 10.2
58	HB5773, Glasfiber	Hollingworth	45x56
59	Whatman Glass Microfiber Filters GF/A	GE Healthcare Life Sciences	45x56
60	Pads A500 GS, glass fibre	Camfil	ø 7.7
61	EPM 2000 Glass Microfibre Filter	Whatman International Ltd, Maidstone, England.	20x25
62	Flat filter paper, SIO2	TRM Filter d.o.o.	30x30
63	Glass fibre papers	Schleicher & Schuell	ø 4.7
64	F&J model no. FP-4.0M, borosilicate microfiber	F&J Specialty Products, Inc	ø 10.2
65	glas fibre	F&J Specialty Products, Inc	ø 4.7
66	Glass Fiber Filter	GELMAN SCIENCES	ø 4.7
67	Glass Fiber Filter	F&J Specialty Products, Inc	ø 4.7

## Annex 6: Sequence of spiked filters and quality control sources prepared for the 1026 ENV57/MetroERM ILC exercise

**Table A-6**. Sequence of spiked filters and quality control sources prepared for the 2016 ENV57/MetroERM ILC exercise.

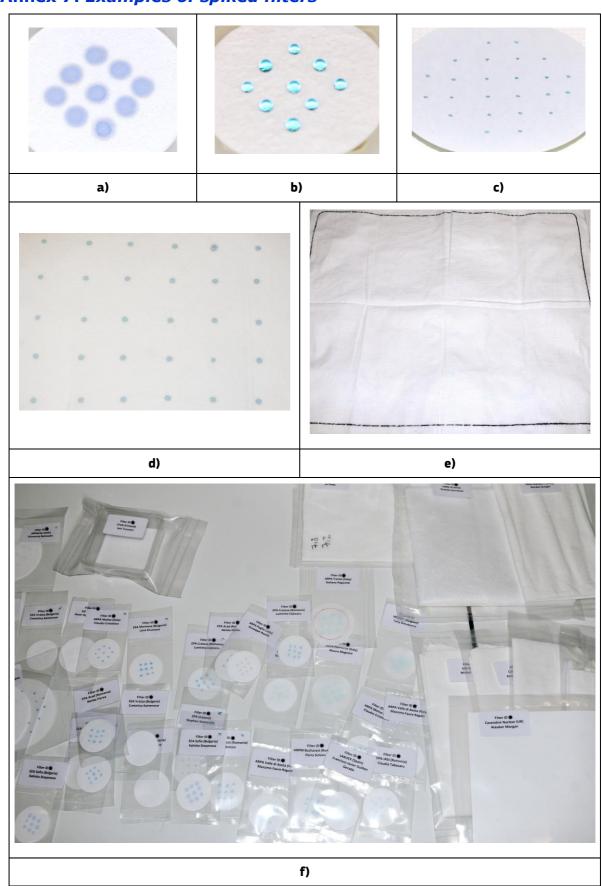
Lab Code	Sample ID	Source Type	Solution
	IRMM-01	Filter	E3
65	Filter_006 Filter		E3
19	Filter_023	Filter	E3
54	Filter_057	Filter	E3
12	Filter_053	Filter	E3
	PS_020	Point Source	E3
	IRMM-02	Filter	E4
	PS_021	Point Source	E4
53	Filter_041	Filter	E2A
52	Filter_019 Filter		E2A
42	Filter_051	ter_051 Filter	
18	Filter_055	Filter	E2A
	IRMM-03	Filter	E2A
3	Filter_047	Filter	E2A
35	Filter_043	Filter	E2A
36	Filter_045	Filter	E2A
47	Filter_013	Filter	E2A
	IRMM-04	Filter	E2A
67	Filter_010	Filter	E2A
24	Filter_007	Filter	E2A
23	Filter_002	Filter	E2A
57	Filter_028	Filter	E2A

Lab Code	Sample ID	Source Type	Solution
66	Filter_011	Filter	E2A
21	Filter_029	Filter_029 Filter	
40	Filter_067	Filter	E2A
7	Filter_026	Filter	E2A
34	Filter_020	Filter	E2A
	IRMM-05	Filter	E2A
	PS_022	Filter	E2A
	IRMM-06	Filter	E1
8	Filter_059	Filter	E1
27	Filter_064	Filter	E1
	PS_023	Point Source	E1
25	Filter_012	Filter	E4
44	Filter_024	Filter	E4
10	Filter_027	Filter	E4
29	Filter_015	Filter	E4
11	Filter_005	Filter	E4
61	Filter_054	Filter	E4
	IRMM-07	Filter	E4
	PS_024	Point Source	E4
2	Filter_058	Filter	E4
64	Filter_065	Filter	E4
49	Filter_004	Filter	E5

Lab Code	Sample ID	Source Type	Solution
62	Filter_066	Filter	E5
16	Filter_034	Filter	E5
	IRMM-08 Filter		E5
59	Filter_032	Filter	E5
9	Filter_008	Filter	E5
	PS_025	Point Source	E5
20	Filter_022	Filter	E5
32	Filter_001	Filter	E5
60	Filter_046	Filter	E5
56	Filter_039	Filter	E5
48	Filter_025	Filter	E5
15	Filter_052	Filter	E5
51	Filter_016	Filter	E5
46	Filter_017	Filter	E5
38	Filter_040	Filter	E5
	IRMM-09	Filter	E5
31	Filter_063	Filter	E4
63	Filter_056	Filter	E2A
37	Filter_033	Filter	E5
13	Filter_037	Filter	E5
55	Filter_049	Filter	E5
28	Filter_061	Filter	E5
	IRMM-10	Filter	E5
5	Filter_042	Filter	E5
43	Filter_031	Filter	E5
4	Filter_060	Filter	E5
	IRMM-11	Filter	E5

Lab Code	Sample ID	Source Type	Solution
30	Filter_048	Filter	E5
26	Filter_062	Filter	E5
45	Filter_021	Filter	E5
	IRMM-12	Filter	E2A
1	Filter_038	Filter	E2A
39	Filter_003	Filter	E2A
50	Filter_018	Filter	E5
22	Filter_036	Filter	E5
58	Filter_014	Filter	E5
	IRMM-14	Filter	E5
	PS-026	Filter	E5
41	Filter_050	Filter	E5
17	Filter_030	Filter	E5
14	Filter_035	Filter	E5
	IRMM-15	Filter	E5
33	Filter_044	Filter	E5
6	Filter_009	Filter	E5

**Annex 7:** Examples of spiked filters



## Annex 8: Quality control filters and sources; measurement results and their deviation from the reference activity values

**Table A-7a**. Gamma-ray spectrometric results of the quality control filters; measured values and the corresponding uncertainties.

Sample_			13	<sup>7</sup> Cs	13	<sup>4</sup> Cs	1:	<sup>31</sup> I
ID (IRMM- ##)	DILUTION	DETECTOR	Measured Activity (Bq)	Uncertainty (Bq) (k=1)	Measured Activity (Bq)	Uncertainty (Bq) (k=1)	Measured Activity (Bq)	Uncertainty (Bq) (k=1)
01	E3	В	0.540	0.032	0.800	0.036	0.626	0.024
02	E4	В	0.089	0.013	0.200	0.026	0.164	0.012
03	E2A	В	0.546	0.038	0.887	0.046	0.442	0.018
04	E2A	В	0.484	0.027	0.733	0.031	0.414	0.015
04	E2A	В	0.482	0.028	0.736	0.031	0.427	0.017
05	E2A	В	0.483	0.034	0.655	0.037	0.369	0.016
06	E1	В	1.935	0.092	2.138	0.080	1.408	0.050
07	E4	Α	0.082	0.011	0.145	0.016	0.103	0.024
08	E5	В	0.101	0.011	0.168	0.018	0.121	0.007
09	E5	В	0.141	0.010	0.264	0.019	0.254	0.011
10	E5	В	0.087	0.007	0.176	0.013	0.210	0.009
11	E5	Α	0.115	0.012	0.223	0.014	0.200	0.009
12	E2A	В	0.380	0.019	0.574	0.022	0.747	0.038
14	E5	В	0.089	0.007	0.185	0.014	0.175	0.011
15	E5	Α	0.081	0.008	0.186	0.012	0.126	0.008

**Table A-7b**. Gamma-ray spectrometric results of the quality control filters; their relative deviation from the reference values and their ratio to the reference values.

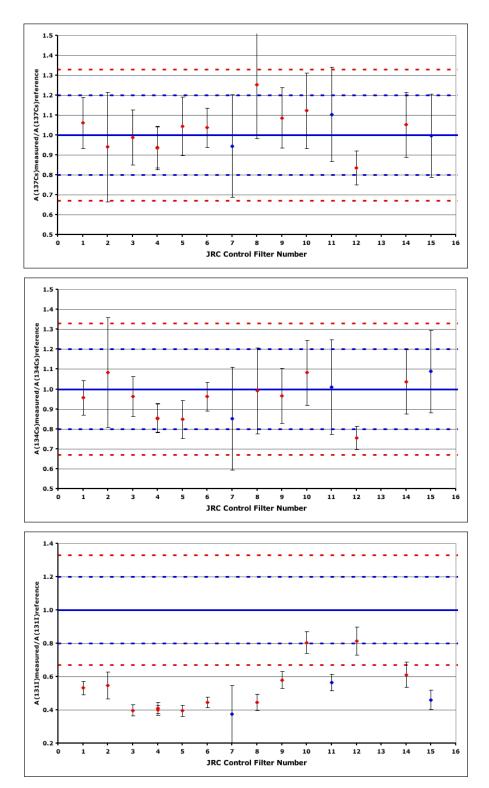
	<sup>137</sup> Cs				<sup>134</sup> Cs		<sup>131</sup> I			
Sample _ID (IRMM- ##)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncer- tainty (k=2)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncer- tainty (k=2)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncertainty (k=2)	
01	6.1	1.06	0.13	-4.4	0.96	0.09	-47	0.53	0.04	
02	-6.0	0.94	0.28	8.4	1.08	0.28	-45	0.55	0.08	
03	-1.1	0.99	0.14	-3.7	0.96	0.10	-60	0.40	0.03	
04	-6.1	0.94	0.10	-14.7	0.85	0.07	-60	0.40	0.03	
04	-6.5	0.94	0.11	-14.4	0.86	0.07	-59	0.41	0.03	
05	4.3	1.04	0.15	-15.2	0.85	0.10	-60	0.40	0.03	
06	3.8	1.04	0.10	-3.7	0.96	0.07	-55	0.45	0.03	
07	-5.5	0.94	0.26	-14.7	0.85	0.19	-62	0.38	0.17	
08	25.2	1.25	0.27	-0.8	0.99	0.22	-55	0.45	0.05	
09	8.6	1.09	0.15	-3.4	0.97	0.14	-42	0.58	0.05	
10	12.2	1.12	0.19	8.3	1.08	0.16	-20	0.80	0.07	
11	10.4	1.10	0.24	1.0	1.01	0.13	-44	0.56	0.05	
12	-16.5	0.84	0.08	-24.5	0.75	0.06	-18	0.82	0.08	
14	5.2	1.05	0.16	3.7	1.04	0.16	-39	0.61	0.07	
15	-0.3	1.00	0.21	8.8	1.09	0.14	-54	0.46	0.06	

**Table A-8a**. Gamma-ray spectrometric results of the quality control point sources; measured values and the corresponding uncertainties.

			13	<sup>7</sup> Cs	13	<sup>4</sup> Cs	<sup>131</sup> I	
Sample_ ID (PS_##)	DILUTION	DETECTOR	Measured Activity (Bq)	Uncertainty (Bq) (k=1)	Measured Activity (Bq)	Uncertainty (Bq) (k=1)	Measured Activity (Bq)	Uncertainty (Bq) (k=1)
15	D1	А	30.9	1.1	37.0	1.3	24.2	0.9
15	D1	В	31.0	1.4	33.5	1.2	24.1	0.9
16	D3	В	1.40	0.07	2.13	0.08	2.30	0.09
17	D4	Α	0.23	0.02	0.45	0.03	0.53	0.03
18	D5	В	0.36	0.03	0.69	0.04	0.94	0.05
19	D2A	Α	1.93	0.08	3.42	0.13	2.23	0.09
20	E3	В	0.64	0.04	0.95	0.04	0.77	0.03
21	E4	Α	0.11	0.01	0.20	0.02	0.19	0.01
22	E2A	Α	0.65	0.03	1.05	0.05	0.59	0.03
23	E1	В	1.41	0.07	1.58	0.06	0.83	0.04
24	E4	В	0.18	0.01	0.14	0.02	0.12	0.01
25	E5	В	0.058	0.006	0.12	0.02	0.12	0.01
26	E5	Α	0.063	0.010	0.13	0.01	0.15	0.01

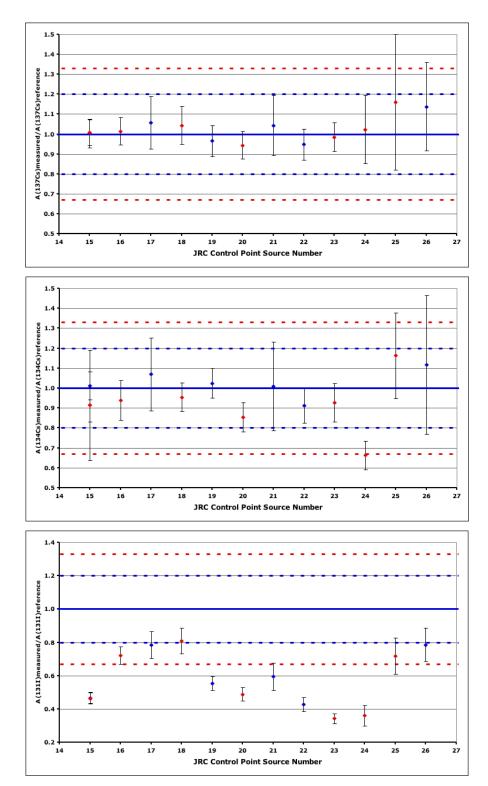
**Table A-8b**. Gamma-ray spectrometric results of the quality control point sources; their relative deviation from the reference values and their ratio to the reference values.

		<sup>137</sup> Cs			<sup>134</sup> Cs		<sup>131</sup> I		
Sample _ID (PS_# #)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncer- tainty (k=2)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncer- tainty (k=2)	Deviation from Ref. Activity (%)	Activity RATIO (Measured to Reference)	Uncer- tainty (k=2)
15	0.33	1.00	0.07	1.1	1.01	0.07	-53	0.47	0.03
15	0.7	1.01	0.09	-8.4	0.92	0.07	-54	0.46	0.03
16	1.45	1.01	0.10	-6.1	0.94	0.07	-28	0.72	0.05
17	5.8	1.06	0.18	6.9	1.07	0.13	-22	0.78	0.08
18	4.4	1.04	0.15	-4.5	0.95	0.10	-19	0.81	0.08
19	-3.5	0.97	0.08	2.4	1.02	0.08	-45	0.55	0.04
20	-5.6	0.94	0.10	-14.6	0.85	0.07	-51	0.49	0.04
21	4.4	1.04	0.22	0.8	1.01	0.15	-41	0.59	0.08
22	-5.25	0.95	0.09	-8.9	0.91	0.08	-57	0.43	0.04
23	-1.5	0.98	0.10	-7.3	0.93	0.07	-66	0.34	0.03
24	2.3	1.02	0.19	-34	0.66	0.17	-64	0.36	0.06
25	16.0	1.16	0.24	16.3	1.16	0.34	-28	0.72	0.11
26	13.8	1.14	0.35	11.6	1.12	0.22	-21	0.79	0.10



**Fig. A-1**. Ratio of measured-to-reference activity and expanded uncertainty (k=2) of the quality control **filters** prepared from the <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I dilutions used for spiking the air filters. The blue diamond symbol is used for results obtained with Detector A and the red diamond symbol for those obtained with Detector B. The solid blue line is the Reference value (unity), the blue dashed lines designate the  $\pm 20\%$  range and the red dashed lines the  $\pm 33\%$  range.

Top: 137Cs, Middle: 134Cs, Bottom: 131I



**Fig. A-2**. Ratio of measured-to-reference activity and expanded uncertainty (k=2) of the quality control **point sources** prepared from the <sup>137</sup>Cs, <sup>134</sup>Cs and <sup>131</sup>I dilutions used for spiking the air filters. The blue diamond symbol is used for results obtained with Detector A and the red diamond symbol for those obtained with Detector B. The solid blue line is the Reference value (unity), the blue dashed lines designate the  $\pm 20\%$  range and the red dashed lines the  $\pm 33\%$  range.

Top: <sup>137</sup>Cs, Middle: <sup>134</sup>Cs, Bottom: <sup>131</sup>I

# Annex 9: Reporting form and second questionnaire for submitting the measurement results

Milc questionnaire	1.3. Did you apply any preconcentration or chemical treatment? *
Comparison for Cs-137, Cs-134 and I-131 measurement in air filters	O a) Yes
As it was previously announced, the reporting of your measurement method and results will be done via an online questionnaire. We kindly ask you to answer all relevant questions regarding the procedures you used for the measurement of the filter sample. Questions marked with an asterisk "of are mandatory to answer. Please note that only upon reception of the signed reporting form your submission is final! Be aware that the deadline for the submission of the results and the questionnaire is Tuesday, 29 March 2016. Thank you for your cooperation!	1.3.1. If yes, please describe the method (chemicals and procedures used, chemical recovery obtained, method of chemical recovery determination, etc.): *
Submission Form	
	1.4. Describe the method used for the sample preparation (more than one choice is possible): *
1. Sample Treatment	a) Packing together with blank air filters b) Folding
1.1. Was the comparison sample treated according to the same analytical procedure as routinely used in your laboratory for the same type of samples? *	c) Pressing/compressing d) No sample preparation
○ a) Yes ○ b) No	e) Other
1.1.1. If not, please specify the differences here: *	1.4.1. If other, please specify here: *
<ul><li>1.2. Did you measure the sample together with the inner plastic foil or did you measure the foil separately?</li></ul>	2. Equipment
a) Together      b) The plastic foil was measured separately      c) The plastic foil was NOT measured	2.1. Which type of detector was used for the measurements (more than one choice is possible)? *    a) Ge(Li) detector   b) HPGe detector
1.2.1. If separately, please provide the measured activity of the plastic foil (in Bq) with its uncertainty: *	C) Brose actection     d) Well type detector     f) Other
- Page 1 of 10 -	- Page 2 of 10 -

i) Other	2.3.1. If other, please specify here: *		See table <b>Acquisition time and measurement cycles</b> at bottom	3.3. Provide the limit of detection (value in Bq): *  See table <b>Detection limit</b> at bottom	ne choice is  *  a) Genie 2000  b) GammaVision  c) Curie method  d) ISO 11929  f) Risø method  f) Risø method  g) Other method and/or software
2.1.1. If other, please specify here: *	2.1.2. Provide the supplier, the model of the detector and its relative efficiency:	2.1.3. Describe how the efficiency calibration of the detector is done (to determine the counting efficiency for the air filter measurement) and provide the source of nuclear data used: *	22. What type of electronics and data acquisition was used? *      a) Analog signal processing (spectroscopy amplifier, etc)      b) Digital signal processing      c) Other	2.2.1. If other, please specify here: *	2.3. Specify the software for peak area determination and data evaluation used (more than one choice is possible): *    a) Genie 2000 family   b) Gamma-Vision   c) InterWinner   d) Gamma-W   e) EMCA+   f) Gamma-track   g) SAMPO   h) HYPERMET

3.7.1. If yes, please insert the correction factor below: *	4. Uncertainty budget * See table Uncertainty budget at bottom	4.1. If you indicated "Other" in the table above, please specify the component(s):	5. Additional information 5.1. Difficulties encountered	5.2. Comments on this interlaboratory comparison excercise:	5.3. Comments on this questionnaire:		- Page 6 of 10 -
3.4.1. If other method and/or software, please specify here: *	3.5. Did you use the recommended half-lives and gamma-ray emission intensities? *      a) Yes      b) No	3.5.1. If not, please indicate the values you used: * See table Half-lives and gamma-ray emission intensities at bottom	3.5.2. Please indicate the reference for the values you used:	3.6. Did you apply corrections for the coincidence summing? *  O a) Yes  O b) No	3.6.1. If yes, please give the correction factors you used in the table below: * See table Coincidence summing correction factors at bottom	<ul> <li>3.7. Did you apply a correction for the decay of I-131 during the counting interval? *</li> <li>() a) Yes</li> <li>() b) No</li> </ul>	- Page 5 of 10 -

Acquisition time and measurement cycles

Questions/Response table	Acquisition time (sec)	Measurement cycles	Count rate for the 605-KeV Cs-134 peak(counts/sec)	Count rate for the 364-KeV I-131 peak (counts/sec)
Sample				
Background/blank				

Coincidence summing correction factors

Questions/Response table	Coincidence summing correction factor
Cs-134 (604.7 KeV)	
Cs-134 (795.9 KeV)	
1-131 (364.5 KeV)	
1-131 (637.0 KeV)	

Detection limit

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Questions/Response table	Detection limit (Bq)
Cs-137	
Cs-134	
1-131	

Half-lives and gamma-ray emission intensities

Questions/Response table	Half-life used (days)	Gamma-ray emission intensity used (in %)					
Cs-137 (661.7 KeV)							
Cs-134 (604.7 KeV)							
Cs-134 (795.9 KeV)							
I-131 (364.5 KeV)							
I-131 (637.0 KeV)							

Measurements period

Questions/Response table	Start date	End date
Measurements period		

- Page 8 of 10 -

Uncertainty	buck	last

The combined relative standard uncertainty (i.e. the quadratic sum of the uncertainty components) should be the same as the standard uncertainty reported with your results.

Questions/Response table	Relative uncertainty component (%) - Cs-137	Relative uncertainty component (%) - Cs-134	Relative uncertainty component (%) - I-131
Counting statistics			
Blank/background measurement			
Efficiency calibration (including the activity of the calibratuion source)			
Chemical yield (if applicable)			
Self-absorption in the source			
Activity of the calibration source			
Decay correction			
Other 1			
Other 2			
Other 3			
Combined relative standard uncertainty (quadratic sum of the components)			

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## Annex 10: Responses to the second questionnaire accompanying the reporting form

To determine the ratios, the 67 participants were taken as 100%.

**Table A-9**. Treatment of spiked air filters before measurement by the participants and equipment used for the measurement of the spiked air filters.

Question	Number of answers	Ratio (%)			
"Were the comparison sample treated according t routinely used in your laboratory for the					
Yes	62	92.5			
No	5	7.5			
"Did you measure the sample together with the pla separately?"	astic foil or did you meas	sure the foil			
Together	33	49			
Separately	26	39			
The plastic foil was not measured	8	12			
"Did you apply any pre-concentration o	or chemical treatment?"				
Yes	0	0			
No	67 100				
"Which type of detector was used for the determ	nination of <sup>137</sup> Cs in the a	ir filter?"			
Ge(Li) detector	2	3			
HPGe detector	58	87			
BEGe detector	13	19			
Well type detector	1	1.5			
NaI(TI) detector	0	0			
Other	0	0			
"What type of electronics and data a	cquisition was used?"				
a) Analog signal processing (spectroscopy amplifier, etc.)	19	21			
b) Digital signal processing	72	79			
c) Other	0	0			
Use of recommended h	alf-lives				
Yes	59	88			
	i	1			

No	8	12					
Coincidence summing correction applied							
Yes	48	72					
No	19	28					
Decay during measurement correction applied (for <sup>131</sup> I)							
Yes	48	72					
No	19	28					

**Table A-10**. Measurement parameters; acquisition time, count rates, MDA.

	Minimum	Maximum	Mean	Median
Measurement time of sample (h)	1	233	53.0	47.9
Count rate for <sup>137</sup> Cs peak in sample (cps)	0.0011	0.153	0.0176	0.0132
Count rate for the 605 keV <sup>134</sup> Cs peak (cps)	0.0015	0.174	0.0267	0.0264
Count rate for the 364 keV <sup>131</sup> I peak (cps)	0	0.314	0.0347	0.0209
Measurement time of background (h)	15	333	80.8	63.0
Count rate for <sup>137</sup> Cs peak in background (cps)	0	0.124	0.0032	-
Count rate for the 605 keV <sup>134</sup> Cs peak in background (cps)	0	0.161	0.0038	-
Count rate for the 364 keV <sup>131</sup> I peak in background (cps)	0	0.039	0.0022	-
MDA for <sup>137</sup> Cs (Bq)	0.003	0.12	0.039	0.029
MDA for <sup>134</sup> Cs (Bq)	0.002	0.53	0.046	0.025
MDA for <sup>131</sup> I (Bq)	0.0002	0.50	0.051	0.025

## Annex 11: Coincidence summing correction factors as applied by the participants

**Table A-11**. Coincidence summing correction factors as applied and reported by the participants.

Lab	134	Cs	13	¹I
Code	604.7 keV	795.9 keV	364.5 keV	637.0 keV
1	n.a software determined	n.a software determined	n.a software determined	n.a software determined
2				
3	1.02	1.018		
4				
5	1.15	1.15		
6	0.867	0.864	1.005	1
7	0.823	0.817	1.01	1
8				
9	0.801	0.799	1	not used
10	0.76532	0.76795	1	not used
11	1.17	1.168	0.997	0.999
12	0.788	0.787	free	free
13	1.22	1.23	1	1
14	0.76497	0.76133	1.0092	1.0017
15				
16	by software	by software	by software	by software
17	1.137	1.138		
18	0.899	0.898	1.004	
19	0.818	0.815		
20	1.25	1.25	1	1
21	0.795	0.789	1.01	free
22				
23	0.87	0.87	1.01	1
24	1.269	1.267	0.995	0.998
25	0.78363	0.78101	1.0097	1
26	0.851	0.85	1.003	free
27				
28				
29	0.939	0.928	1.009	
30				
31	1.118	1.117		
32	1.277		1	
33	0.87175	0.87174	1	
34	0.0825	0.821		
35				
36				
37	1.14	1.14	0.998	0.999
38				
39	0.947	0.946	1.002	

Lab	134	Cs	13	¹I
Code	604.7 keV	795.9 keV	364.5 keV	637.0 keV
40				
41				
42	0.828	0.825	1	free
43	1.07	1.14	0.97	1.08
44	0.79499	0.79405	1.0092	
45	NA	0.93	0.97	NA
46	0.901	0.9	1.003	1
47	1.18	1.2	1	
48	0.87	0.869	1.004	1
49				
50				
51				
52	0.712	0.692	1	
53				
54				
55				
56				
57				
58	1.24	1.24	1	1
59	0.849	0,848	1	
60	1.299	1.303	0.99	1
61				
62	1.279	1.279	0.988	0.995
63	1.35	1.35	1	1
64	1.252	1.249	0.995	
65	0.863	0,859		
66	1.18			
67	1.197	1.199	0.991	

**Note:** The coincidence summing correction factors in the table above are given as reported by the participants. As there is net summing in for the  $^{134}$ Cs peaks and summing out for the  $^{131}$ I, the correction factors should be >1 for the two  $^{134}$ Cs peaks and <1 for the two  $^{131}$ I peaks. It is possible that some participants reported the reciprocal of the correction factor.

### Annex 12: Uncertainty budget as reported by the participating laboratories

**Table A-12**. Detailed uncertainty budget table showing the uncertainty components as reported by the participating laboratories, for all three radionuclides. In column 13 the combined relative uncertainty as reported by the participants is given. For comparison, in column 15 the combined relative uncertainty calculated as square root of the quadratic sum of the uncertainty components given in the table is given. Finally, in column 16 the combined relative uncertainty as calculated from the uncertainty accompanying the results is given.

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
1	Cs-137	26.4	n.d.	1.20	n.d.	n.d.	4	n.d.	2.5	8		28	2	28.0	26.5	O1 (reproducibility, stability, weight of multigamma
1	Cs-134	17.1	n.d.	1.20	n.d.	n.d.	4	n.d.	2.5	8		19.5	2	19.5	16.7	source)+ O2 (other software
1	I-131	16.8	n.d.	1.20	n.d.	n.d.	4	n.d.	2.5	8		19.2	2	19.2	14.3	calculation, not explicit)
2	Cs-137											see 4,1	1	0.0	7.7	We do not, as of yet, report the individual contribution of factors to the unc budget. Instead, we have a generously set value for random uncertainties, which includes
2	Cs-134											see 4,1	1	0.0	7.2	factors such as
2	I-131											see 4,1	1	0.0	8.0	sample weight. This value is 5 %.

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
																The uncertainties relating to the bac
3	Cs-137	5.053											2	5.1	10.9	relating to the bac
3	Cs-134	2.893											2	2.9	10.7	
3	I-131	4.052											2	4.1	9.4	
4	Cs-137	100											1	100.0	10.0	
4	Cs-134	100											1	100.0	2.9	
4	I-131	100											1	100.0	66.0	
5	Cs-137	4		1.1	0.96							5.95	2	4.3	11.9	
5	Cs-134	3.2		1								3.95	2	3.4	7.9	
5	I-131	2.1		4.3								5.2	2	4.8	10.4	
6	Cs-137	7.3		2.35			1.2		3	3.4		9	1.65	9.0	9.6	
6	Cs-134	6.1		2.35			1.2		3	3.4		8	1.65	8.0	8.1	sample
6	I-131	8.5		2.35			1.2		3	3.4		10	1.65	10.0	9.9	preparation
7	Cs-137	2.1	7	5		1		1	0.24	1	0	8.91	1	9.0	8.9	1-inhomogeneity, 2-random error, 3-
7	Cs-134	1.3		5		2		1	0.24	1	2	6.06	1	6.1	6.0	true coincidence
7	I-131	2.7		5		2		2	0.24	2	2	6.95	1	7.0	7.1	correction factor
8	Cs-137												2	0.0	11.3	
8	Cs-134												2	0.0	11.8	
8	I-131												2	0.0	11.3	
9	Cs-137	6	2	5		5	3	1	5	2		11.4	2	11.4	12.5	Sample
9	Cs-134	4	2	5		5	3	1	5	2		10.4	2	10.4	10.4	preparation; peak area determination
9	I-131	4	2	5		5	3	1	5	2		10.1	2	10.4	10.5	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
10	Cs-137	2.5	0	3.18	0	0	0.95	0.00021	0.000 0092	1.27	3	4.9	2	5.3	10.3	(1) Correction factor during counting period,
10	Cs-134	1.7	0	3.24	0	0	0.95	0.0015	0.000 65	1.27	3	4.6	2	5.0	9.3	(2) Repeatability (Equipment) (3)
10	I-131	5.6	0	3.61	0	0	1.68	0.021	0.008	1.27	3	7.3	2	7.6	15.4	Reproducibility
11	Cs-137	7.16	2	3		1	2	0.27	3.7	2	0.2	9.3	2	9.3	18.2	positioning of the sample; summing correction; combination
11	Cs-134	2.1	0	4		1	2	0.024	4.5	2	0.2	7	2	7.0	15.6	(diameter; sample density; sample
11	I-131												2	0.0	13.6	composition)
12	Cs-137	1.87		1.25					0.27	0.02	6.7 0	7.20	1	7.1	7.2	Other1=unc. on decay constant; Other2=unc. on emission yield; Other3=repeatabili
12	Cs-134	1.61		1.94					0.02	0.23	6.7 0	7.20	1	7.2	12.1	ty(+summing correction for Cs-
12	I-131												1	0.0	6.0	134)
13	Cs-137												2	0.0	8.0	
13	Cs-134												2	0.0	8.0	
13	I-131												2	0.0	8.6	
14	Cs-137	2.53	0	0.89	0	0	0	0	0	0	0	2.6	1	2.7	2.6	Other 1: Gamma emission probability. Other 2: Coincidence
14	Cs-134	1.37	0	0.9	0	0	0	0	0.2	1	2.1	2.83	1	2.9	2.8	summing. Other 3:

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
14	I-131	0.88	0	1.43	0	0	0	0	0.25	1	1	2.22	1	2.2	2.2	Type A uncertainty evaluation
15	Cs-137	3.78	19.4										1	19.8	8.6	
15	Cs-134	2.45											1	2.5	7.4	
15	I-131	1.31											1	1.3	7.6	
16	Cs-137	5.17	-	3.65	-	-	-	0.099	-	-	-	6.33	2	6.3	12.7	
16	Cs-134	2.42	-	3.65	-	-	-	0.242	-	-	-	4.39	2	4.4	8.8	
16	I-131	6.88	-	8.75	-	-	-	0.037	-	-	-	11.13	2	11.1	22.3	
17	Cs-137	3.7	0	1.2		0.2			5			6.3	1	6.3	8.0	1 - unhomogenity.
17	Cs-134	1.4	0	1.2		0.2			5	0.3		5.2	1	5.3	5.5	2 - summing correction
17	I-131	1	0	1.2		0.2		2	5			5.6	1	5.6	6.2	
18	Cs-137			7.3					х			8.5	1	7.3	8.5	Uncertainty is combined by
18	Cs-134			7.3					х			5	1	7.3	5.0	Genie2000, budget
18	I-131			9.3					х			10.2	1	9.3	10.2	not specified.
19	Cs-137	2.6		4				1	1	3		5.81	1	5.8	5.8	1-Self absorption in the sample; 2-
19	Cs-134	1.8		4				1.5	1	3		5.61	1	5.6	5.6	Radioactivity distribution on the
19	I-131	2.6		4				2	1	3		6.06	1	6.1	5.7	filter
20	Cs-137	80	5	5	0	5	5					0.02	2	80.6	9.1	
20	Cs-134	80	5	5	0	5	5					0.05	2	80.6	8.5	
20	I-131	70	5	5		5	5	10				0.09	2	71.4	8.7	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
21	Cs-137	2.12	0	5	0	1	0	1	0.24	1	0	5.71	1	5.7	5.7	Other 1=INHOMOGENEIT Y; Other 2=RANDAOM ERROR; Other 3=TRUE
21	Cs-134	1.97	0	5	0	2	0	1	0.24	1	2	6.24	1	6.2	6.2	COINCIDENCE
21	I-131	2.79	0	5	0	2	0	2	0.24	2	2	6.99	1	7.0	6.9	CORRECTION FACTOR
22	Cs-137								7				1	7.0	8.3	Generic 7% uncertainty added to uncertainties
22	Cs-134								7				1	7.0	7.5	from weighted averages across 3
22	I-131								7				1	7.0	7.3	detectors
23	Cs-137												2	0.0	6.7	
23	Cs-134												2	0.0	10.0	
23	I-131												2	0.0	10.5	
24	Cs-137											7.9	2	0.0	7.9	
24	Cs-134											5.2	2	0.0	5.3	
24	I-131											23.8	2	0.0	24.6	
25	Cs-137	5.36	0	20	0	0	3	0	0.5	0	0	20.81	2	20.9	20.6	
25	Cs-134	3.77	0	20	0	0	3	0	2	0	0	20.45	2	20.7	20.4	emission intensity
25	I-131	2.64	0	20	0	0	3	0	1.25	0	0	20.29	2	20.4	20.3	
26	Cs-137	6		5			5		2.89			14	2	9.7	19.6	combined
26	Cs-134	6		5			5		2.89			14	2	9.7	19.6	operational
26	I-131	6		5			5		2.89			14	2	9.7	17.9	uncertainty
27	Cs-137											1 171	2	0.0	2.3	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
27	Cs-134											1 153	2	0.0	2.3	
27	I-131											3 781	2	0.0	2.2	
28	Cs-137	10		5					2				2	11.4	26.0	Correction for density and fill height
28	Cs-134	10		5					2				2	11.4	18.0	
28	I-131	10		5					2				2	11.4	22.6	
29	Cs-137	6.81		2.32			1.2		3	3.4		8.59	1.65	8.5	8.6	
29	Cs-134	5.32		2.32			1.2		3	3.4		7.46	1.65	7.4	7.4	preparation and handling
29	I-131	30.98		2.79			2.2		3	3.4		31.51	1.65	31.4	12.4	nanamig
30	Cs-137	2.8		2.02									2	3.5	2.8	
30	Cs-134	1.61		2.01									2	2.6	1.6	
30	I-131	2.28		3.03									2	3.8	2.3	
31	Cs-137	3	2.5	4			2.5					5.9	2	5.6	5.8	
31	Cs-134	1.9	2.5	4				1.5				5	2	5.1	4.9	
31	I-131	2.5	2.5	4				1.8				6.2	2	5.3	6.1	
32	Cs-137	1.20 %	0	1.5		3			3	3	3		2	6.2	16.7	coincidence calculation / efficiency transfer gespecor /
32	Cs-134	0.8	0	1.4		3			3	3	3		2	6.2	16.7	systematic bias due to
32	I-131	0.5	0	1.4		3			3	3	3		2	6.2	14.1	compression
33	Cs-137	3.65		3.301	0.24				0.13	1.6	0.2	5.2	2	5.2	10.8	Geometry Repeatability,
33	Cs-134	2.19		4.32	0.35				0.13	1.6	0.2	5.1	2	5.1	10.1	Volume& system
33	I-131	2.22		3.53	0.74				0.13	1.6	0.2	4.5	2	4.5	9.0	stability, Tcc error

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
34	Cs-137	4		6			1.2	0.2	5	4		9.72	1	9.6	9.8	the inhomogeneity of the sample and approximating a
34	Cs-134	4.2		6			1.2	0.39	5	4		9.81	1	9.7	9.9	circular shape for
34	I-131	4.4		7			1.2	1	5	4		10.76	1	10.5	10.3	the active spots
35	Cs-137					0			8	19		20	2	20.6	40.9	Other 1: Uncertainty budget without intra- reproducibility;
35	Cs-134					0			6	19		20	2	19.9	60.1	Other 2: Intra-
35	I-131					0			7	19		20	2	20.2	40.3	reproducibility for this geometry
36	Cs-137	8.23	0	3.16	0		3	0.5	5	0	0	10.58	2	10.1	7.3	-
36	Cs-134	4.21	0	3.16	0		3	0.1	5	0	0	7.85	2	7.3	6.4	Systematic errors
36	I-131	21.27	0	3.16	0		3	0.1	5	0	0	22.28	2	22.1	12.1	
37	Cs-137	4.42	0	6					0			7.5	2	7.5	8.6	Coincidence summing
37	Cs-134	3.92	0	6					2			7.4	2	7.4	8.0	correction
37	I-131	1.79	0	6					0			6.3	2	6.3	6.3	uncertainty
38	Cs-137	3.43	6.61	3.54			1.51		5			7.84	1	9.6	7.9	Other 1: the sample homogeneity, Other 2: the
38	Cs-134	2.33	6.14	3.54			1.51		5			6.87	1	9.0	6.8	activity left on a
38	I-131	1.56	4.63	3.54			1.51		5	7		9.59	1	10.5	9.7	groundwork
39	Cs-137								х				1	0.0	10.0	uncertainty budget put in edit and

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
39	Cs-134								×				1	0.0	3.9	calculated by
39	I-131								Х				1	0.0	25.0	Genie analysis software
40	Cs-137								9				2	9.0	8.8	The calculations were performed
40	Cs-134								7				2	7.0	6.7	using Gamma
40	I-131								15				2	15.0	16.0	Vision Software
41	Cs-137					1						2.68	2	1.0	2.7	
41	Cs-134					2						2.16	2	2.0	2.2	
41	I-131					2						1.51	2	2.0	1.5	
42	Cs-137	2.08	0	5	0	-	0	1	0.24	1	0	5.69	1	5.6	5.7	Other 1=INHOMOGENEIT Y; Other 2=RANDAOM ERROR; Other
42	Cs-134	1.54	0	5	0	-	0	1	0.24	1	2	6.12	1	5.8	6.2	3=TRUE COINCIDENCE
42	I-131	5.21	0	5	0	-	0	2	0.24	2	2	8.26	1	8.0	8.0	CORRECTION FACTOR
43	Cs-137	11.45	-	2.08	-	1	1.6	-	5	-	-	12.77	2	12.8	15.4	
43	Cs-134	5.85	-	2.11	-	1	-	-	5	-	-	7.98	2	8.0	11.7	Error of repeatability
43	I-131	32.97	-	1.66	-	1	-	-	5	-	-	33.39	2	33.4	30.8	, ,
44	Cs-137	6.4	113	1.75		NA	1		3.3			7.5	2	113.2	16.7	
44	Cs-134	4.2	69.6	1.75		NA	1		4.1			6.2	2	69.9	12.2	repeatability
44	I-131	12.4	54	1.75		NA	1		6.7			14.2	2	55.8	30.4	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
45	Cs-137	3.29	NA	1.3	NA						1	3.68	1	3.7	3.7	1. coincidence summing; 2. efficiency curve interpolation; 3. sample fitting to
45	Cs-134	2.57	NA	1.5	NA				1.8	1.2	1	3.82	1	3.8	3.8	the standard
45	I-131	1.4	NA	1.5	NA			0.03	1.5	1.2	1	2.99	1	3.0	3.2	geometry
46	Cs-137	4.41		10					10.2			14.8	1	14.9	16.7	Other 1: Folding and positioning of
46	Cs-134	2.53		10					10.2			14.5	1	14.5	15.6	the filter on the
46	I-131	3.88		10					10.2			14.9	1	14.8	14.6	detector
47	Cs-137	1.7		0.5		-			5			5.3	1	5.3	5.4	geometry factor,
47	Cs-134	1.6		0.5		-			5	7.2		8.9	1	8.9	8.9	coincidence correction
47	I-131	5.5		2.1		-			5			7.7	1	7.7	7.7	
48	Cs-137	2.827	-	1.17	-		1.17	1.67E-05	1.71	0.23 5	1.2	3.89	1	3.9	4.7	systematic, yield,
48	Cs-134	1.645	1	2.12	=		1.17	0.00061	1.71	0.08 2	1.2	3.6	1	3.6	3.3	monte carlo corrections
48	I-131	0.983 5	ı	1.6846	ı		1.17	0.0021	1.71	0.61 6	1.2	3.15	1	3.1	3.5	
49	Cs-137	6.02				not applicabl e						6.02	2	6.0	6.3	
49	Cs-134	3.8										3.8	2	3.8	3.5	
49	I-131	5.2										5.2	2	5.2	5.2	
50	Cs-137	2.4	in countin	2.4	not applica	0	includ ed in	included in result				3.4	1	3.4	3.4	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
			g statistic s		ble		efficie ncy									
50	Cs-134		-			0							1	0.0	4.6	
50	I-131					0							1	0.0	5.4	
51	Cs-137	2.103	4.677	1.3264	0		1.43	0.26	0	0	0	5.487	1	5.5	3.0	
51	Cs-134	1.494	0	1.5172	0		1.43	0.07	0	0	0	2.565	1	2.6	5.5	
51	I-131	1.156	0	2.7404	0		2.04	0.02	0	0	0	3.607	1	3.6	3.5	
52	Cs-137	2.35		5.49			2.27		2	0.58		6.33	2	6.7	12.5	Other 1: Additional random uncertainty; Other 1: Additional
52	Cs-134	7.17		5.49			2.27		2	0.58		9.27	2	9.5	11.9	systematic
52	I-131												2	0.0	18.1	uncertainty
53	Cs-137	91.44		8.66									2	91.8	9.3	
53	Cs-134	91.44		8.66									2	91.8	8.5	
53	I-131	91.44		8.66									2	91.8	21.3	
54	Cs-137	4.5	2.3	2.8								5.8	1	5.8	2.5	
54	Cs-134	4.5	2.3	2.8								5.8	1	5.8	2.5	
54	I-131	4.5	2.3	2.8								5.8	1	5.8	5.8	
55	Cs-137	2.9	0.5	3.5		0.1	3.4	0.2				5.7	2	5.7	8.0	
55	Cs-134	1.7	0	3.5		0.1	3.4	0.2				5.2	2	5.2	8.3	
55	I-131	1		4.7		0.5	3.4	0.1				5.9	2	5.9	7.9	
56	Cs-137	2	13.2	2			1.4		0.15			13.6	1	13.6	7.7	Gamma-ray
56	Cs-134	1.2	0	10					0.1			10	1	10.1	6.4	emission intensity
56	I-131	0.94	0	10					1			10	1	10.1	6.6	used

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
57	Cs-137	6.99		3					0.24			7.62	2	7.6	7.6	Gamma photons
57	Cs-134	24.12		3					0.08			24.3	2	24.3	24.3	emission
57	I-131												2	0.0	0.0	probability
58	Cs-137	2.7	0	1.4					0.5				1	3.1	2.9	Relative positioning difference between calibration source and sample, decay correction uncert. added by Genie2000, eff. calib. uncertainty 1.4% due to differences in
58	Cs-134	1.4	0	1.4					0.5				1	2.0	1.4	calibration source
58	I-131	1.2	0	1.4				0	0.5				1	1.9	2.2	and sample, in addition to the uncertainty in the efficiency curve ca
59	Cs-137	4.1		3.7								5.52	1	5.5	5.5	
59	Cs-134	2		3.9								4.4	1	4.4	4.5	
59	I-131	4.3		4.24								6.04	1	6.0	6.8	
60	Cs-137											5.3	1	0.0	6.1	
60	Cs-134											2.6	1	0.0	2.8	
60	I-131											2.7	1	0.0	2.7	
61	Cs-137											14.61	1	0.0	14.6	
61	Cs-134											9.53	1	0.0	9.5	
61	I-131											5.86	1	0.0	5.9	

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
62	Cs-137	2.92	22	5.6			2.5		1			6.4	1	23.0	2.9	Other 1 = Inhomogeneity. Self absorption uncertainty contribution is included in Efficiency calibration uncertainty. Sample was
62	Cs-134	1.36		2.86			2.5		1			4	1	4.2	3.1	measured on
62	I-131	2.4		5.35			2.5		1			6.4	1	6.5	4.1	several spectometers, the uncertainty budget is given for one measurement only.
63	Cs-137	4.174		1.192			3		2.3			9.92	2	5.8	9.9	other 1 is budget
63	Cs-134	3.195		1.192			3		2.3			9.18	2	5.1	9.2	about
63	I-131	26		1.192			3		2.3			28.6	2	26.3	57.2	inhomogeneity
64	Cs-137	6	3.7	4.5								8.4	1	8.4	8.7	
64	Cs-134	5	2	5.5								7.7	1	7.7	7.7	
64	I-131	5.4	3.7	4.5								7.9	1	7.9	7.9	
65	Cs-137	3.2		4				1	2.5			5.78	1	5.8	5.8	distribution of blank filters
65	Cs-134	2.4		4				1.5	2.5			5.5	1	5.5	5.4	spiked, is random
65	I-131	3.4		4				2	2.5			6.14	1	6.1	6.1	, ,
66	Cs-137	1.9	0	0.5			0.8	0.2	4.6			5	1	5.1	5.3	geometry factor, coincidence

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
Lab Code	Nuclide	Counting statistics	Blank/background measurement	Efficiency calibration (including the activity of the calibratuion source)	Chemical yield (if applicable)	Self-absorption in the source	Activity of the calibration source	Decay correction	Other 1	Other 2	Other 3	Combined relative standard uncertainty	Reported k factor	Calculated from components as quadratic sum	Calculated from the reported results	Explanation of "other" components, notes
66	Cs-134	1.2	0	0.6					4.5	6.5		8	1	8.0	8.3	correction
66	I-131	8.9	0	0.7					4.5			10	1	10.0	10.0	
67	Cs-137	1.95		0.5			0.8	0.24	4.5			5	1	5.0	5.0	geometry factor, coincidence
67	Cs-134	1.5		0.7				0.1	4.5	6.4		8	1	8.0	8.0	correction,
67	I-131	6.1		2.7				0.62	4.5		6.3	10	1	10.2	10.2	Homogeneity

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