



ELSEVIER

Contents lists available at ScienceDirect

## Applied Radiation and Isotopes

journal homepage: [www.elsevier.com/locate/apradiso](http://www.elsevier.com/locate/apradiso)

## $^{60}\text{Co}$ in cast steel matrix: A European interlaboratory comparison for the characterisation of new activity standards for calibration of gamma-ray spectrometers in metallurgy



Faidra Tzika<sup>a,\*</sup>, Oleksiy Burda<sup>b</sup>, Mikael Hult<sup>a</sup>, Dirk Arnold<sup>b</sup>, Belén Caro Marroyo<sup>c</sup>, Pavel Dryák<sup>d</sup>, Aldo Fazio<sup>e</sup>, Laurent Ferreux<sup>f</sup>, Eduardo García-Toraño<sup>c</sup>, Andrej Javornik<sup>g,n</sup>, Seppo Klemola<sup>h</sup>, Aurelian Luca<sup>i</sup>, Hannah Moser<sup>j</sup>, Marijan Nečemer<sup>k</sup>, Virginia Peyrés<sup>c</sup>, Mario Reis<sup>l</sup>, Lidia Silva<sup>l</sup>, Jaroslav Šolc<sup>d</sup>, Anton Svec<sup>g</sup>, Zbigniew Tyminski<sup>m</sup>, Branko Vodenik<sup>k</sup>, Uwe Wätjen<sup>a</sup>

<sup>a</sup> EC-JRC-IRMM, Institute for Reference Materials and Measurements, Retieseweg 111, 2440 Geel, Belgium

<sup>b</sup> PTB, Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany

<sup>c</sup> CIEMAT, Metrología de Radiaciones Ionizantes, Avda. Complutense 40, 28040 Madrid, Spain

<sup>d</sup> CMI, Czech Metrology Institute, Radiová 1a, CZ-102 00 Praha 10, Czech Republic

<sup>e</sup> ENEA-INMIRI, Via Anguillarese, 301 Santa Maria di Galeria, 00123 Roma, Italy

<sup>f</sup> CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), bât 602 PC111, CEA-Saclay, 91191 Gif-sur-Yvette Cedex, France

<sup>g</sup> SMU, Slovenský metrologický ústav, Karloveská 63, 842 55 Bratislava 4, Slovak Republic

<sup>h</sup> STUK, Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881, Helsinki, Finland

<sup>i</sup> IFIN-HH, 30 Reactorului Street, 077125 Magurele, Ilfov, Romania

<sup>j</sup> BEV/PTP, Bundesamt für Eich, und Vermessungswesen, Arltgasse 35, A-1160 Wien, Austria

<sup>k</sup> IJS, Institute Jožef Stefan, Jamova 39, 1000 Ljubljana, Slovenia

<sup>l</sup> IST/CTN, Estrada Nacional 10 (km 139,7), 2695-066 Bobadela LRS, Portugal

<sup>m</sup> NCBJ, Narodowe Centrum Badań Jądrowych, RC POLATOM, ul. Andrzeja Soltana 7, 05-400 Otwock, Świerk, Poland

<sup>n</sup> Strojnícka Fakulta Slovenskej Technickej Univerzity v Bratislave, Nám. Slobody 17, Slovak Republic

### HIGHLIGHTS

- A comparison was run on measurement of  $^{60}\text{Co}$  activity in two batches of cast steel.
- The samples were measured using high resolution gamma-ray spectrometry.
- Different experimental and computational efficiency calibration methods were used.
- Two new standards with  $^{60}\text{Co}$  in steel matrix are available for metallurgy.
- The standards' reference activity contents were determined from comparison results.

### ARTICLE INFO

#### Article history:

Received 27 July 2015

Received in revised form

26 April 2016

Accepted 13 May 2016

Available online 18 May 2016

#### Keywords:

Metrology

Ionising radiation

Intercomparison exercise

Gamma-ray spectrometry

Cast steel, metallurgy

EURAMET

### ABSTRACT

Two series of activity standards of  $^{60}\text{Co}$  in cast steel matrix, developed for the calibration of gamma-ray spectrometry systems in the metallurgical sector, were characterised using a European interlaboratory comparison among twelve National Metrology Institutes and one international organisation. The first standard, consisting of 14 disc shaped samples, was cast from steel contaminated during production ("originally"), and the second, consisting of 15 similar discs, from artificially-contaminated ("spiked") steel. The reference activity concentrations of  $^{60}\text{Co}$  in the cast steel standards were  $(1.077 \pm 0.019) \text{ Bq g}^{-1}$  on 1 January 2013 12h00 UT and  $(1.483 \pm 0.022) \text{ Bq g}^{-1}$  on 1 June 2013 12h00 UT, respectively.

© 2016 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

\* Corresponding author.

E-mail address: [faidra.tzika@ec.europa.eu](mailto:faidra.tzika@ec.europa.eu) (F. Tzika).

## 1. Introduction

The number of the “orphan” radioactive sources in the world, although unknown, is estimated in the thousands (IAEA, 2002). Moreover, increasing quantities of recycled metal originate from dismantled nuclear facilities. Both orphan sources and potentially contaminated metal present a radiation hazard, particularly when unintentionally mixed with metal scrap which undergoes recycling. Despite the fact that metal scrap is normally subject to radioactivity control at the entrance of metal foundries using radiation portal monitors, orphan sources or other radioactive material may successfully pass the control if emitted radiation is shielded by the metal load. As a result, the source may enter the melting process and eventually contaminate the final product and/or its by-products. The vast majority of melting events have occurred in the steel recycling industry (IAEA, 2012) wherein the main radionuclide encountered in the steel product is  $^{60}\text{Co}$  (UN-ECE, 2002).

Many steel foundries perform routine monitoring for radioactivity of gamma emitters in material sampled from the melt. These samples are usually the same as the ones used for routine chemical analysis and typically of 70 g in mass (UN-ECE, 2002). In the framework of “MetroMetal” (Ionising Radiation Metrology for Metallurgical Industry) joint research project (<http://projects.ciemat.es/web/metrometal>), which was conducted under EMRP (European Metrological Research Programme) of Euramet, the existing technical capabilities of metal foundries were studied and further enhanced towards accurate measurements. New radionuclide specific measurement systems and calibration methodologies were proposed together with specially designed and developed traceable activity standards including ones of steel matrix (García-Toraño et al., 2015, Šolc et al., 2015, García-Toraño et al., 2014, Tzika et al., 2015).

The steel standards were designed as “fit for purpose” in metal works and as such consisted of cast steel matrix contaminated with  $^{60}\text{Co}$  and matched routinely using sample geometries. Two types of cast steel of different origin were used. One material was originally contaminated steel and was obtained from the Siempelkamp waste management facility in Germany (<http://www.siempelkamp.com/index.php?id=2003&L=0>). The other material was uncontaminated steel which was spiked with  $^{60}\text{Co}$  at the VUHZ metallurgical company in the Czech Republic (<http://www.vuhz.cz/>).

In order to characterise the activity of the two cast steel standards, an interlaboratory comparison (ILC) was conducted among the project's participants on the measurement of the  $^{60}\text{Co}$  activity concentrations in the two materials. Twelve European National Metrology Institutes (NMIs), and the EC-JRC-IRMM participated in the ILC and the latter coordinated the whole exercise. The parallel objective of this comparison was to apply and test the analytical methodology proposed within the MetroMetal project. Of particular interest was the application of proposed calibration methods and efficiency corrections in the case of these high density materials. In the present work the results of this comparison are presented and the approach to derive the reference activity concentrations in the two cast steel standards is discussed.

## 2. Materials and methods

### 2.1. Samples

Two batches of different origins of cast steel samples were used in the ILC: one series of samples was provided by the PTB and represented originally contaminated material coming from a waste management facility (Siempelkamp), and the other was provided

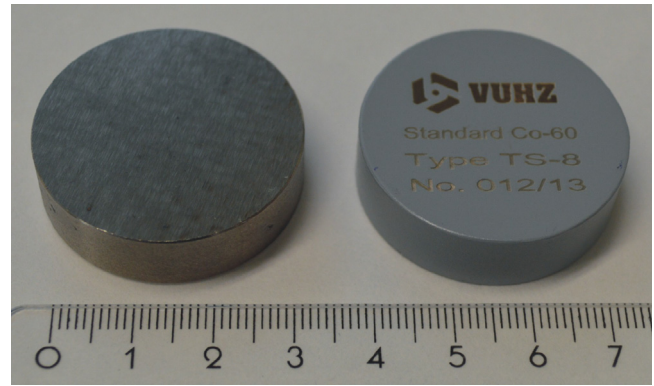


Fig. 1. Cast steel sources containing Co-60. Left: Siempelkamp, Right: VUHZ.

by the SMU and represented spiked material produced by a metallurgical company (VUHZ) by adding  $^{60}\text{Co}$  to cast steel. The raw material in the latter was S355 J0C steel produced according to the EN 10249-1 standard. The ILC was divided into two stages ILC\_A and ILC\_B corresponding to the Siempelkamp and the VUHZ cast steel batches, respectively. The individual samples are shown in Fig. 1. Their masses were  $(70.20 \pm 0.84)$  g for the Siempelkamp and  $(75.12 \pm 0.72)$  g for the VUHZ samples.

The ILC's technical protocol distributed to the participants contained sample information, including elemental composition, instructions and reporting dates. The participants were instructed to analyze the sample in the provided geometry and using gamma-ray spectrometry and to report activity concentrations of  $^{60}\text{Co}$ . The elemental analysis of the raw material of the originally contaminated samples (ILC\_A) was conducted at different partner-laboratories<sup>1</sup> using X-ray fluorescence (XRF) (at the IJS and the CIEMAT) and Proton Induced X-ray Emission (PIXE) (at the IST/CTN) techniques, while carbon content was determined by elemental combustion analysis (at the CIEMAT). The results from different partner-laboratories showed a satisfactory agreement amongst them. CIEMAT's composition data were adopted as most complete. Regarding the spiked samples (ILC\_B) the composition of the raw material was provided from the manufacturer based on analysis using Glow Discharge Optical Emission Spectroscopy (GDOS). The composition data (Table 1) were used as a common basis by the participants conducting Monte Carlo (MC) simulations for relative or absolute efficiency calibration of their gamma-ray spectrometry systems.

### 2.2. Homogeneity testing

The homogeneity testing of each batch of cast steel samples was performed by the PTB and the IRMM, respectively. For each batch, reproducible measurements of all samples were conducted at the same position on the same HPGe (High-Purity Germanium) detector in the respective partner-laboratory. Moreover, the ILC\_B samples were measured in both upright and inverted orientations. The count rates of the samples normalized over the sample mass were compared and the standard deviation of the mean was taken to represent the samples inhomogeneity. The latter was taken as the uncertainty component introduced due to inhomogeneity,  $u_{hom}$ , in the estimation of the overall uncertainty,  $u_{ref}$ , of the reference activity concentration of the samples,  $A_{ref}$ .

The results of homogeneity tests are shown in Fig. 2a and b for ILC\_A and ILC\_B samples, respectively. The adopted  $u_{hom}$  components for the ILC materials are given in Table 2 along with the

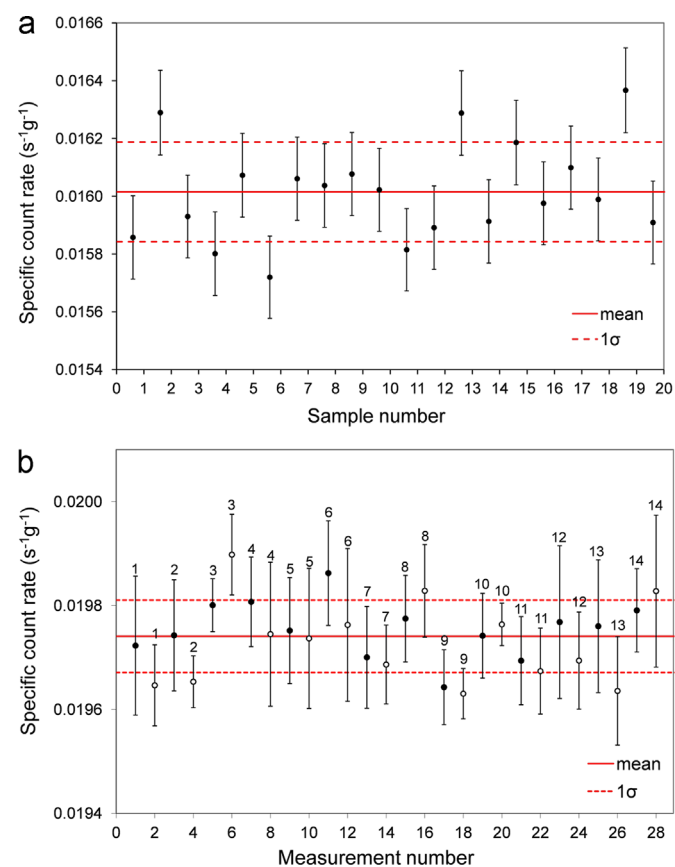
<sup>1</sup> The term “partner-laboratory” is used to distinguish the laboratory's role to provide analytical service from acting as a participant in the comparison exercise.

**Table 1**  
Elemental composition of Siempelkamp and VUHZ raw materials used in the activity standards.

Element	Siempelkamp steel <sup>a</sup> Weight fraction (%)	VUHZ steel <sup>b</sup> Weight fraction (%)
Fe	95.6300(16)	98.623
C	3.1(1)	0.16
Mn	0.3500(5)	0.3
Si	0.3400(5)	0.21
Cr	0.1900(5)	0.2
Ni	0.1100(7)	0.16
Cu	0.1100(4)	0.2
S	0.0575(5)	0.049
Zn	0.0380(5)	
Mo	0.0210(0)	0.03
P	0.0160(5)	0.018
Co	0.0122(1)	0.01
Al	0.0110(14)	0.04
Sn	0.0056(4)	
Pb	0.0048(0)	
V	0.0039(0)	
Nb	0.0019(0)	

<sup>a</sup> Analysis at the CIEMAT by XRF, Carbon content by combustion and iron content by difference.

<sup>b</sup> Composition data as provided by VUHZ based on GDOS. Iron content calculated by difference.



**Fig. 2.** Homogeneity measurements (a) of ILC\_A Siempelkamp and (b) of ILC\_B VUHZ cast steel samples. In (b) the solid and empty data points correspond to upright and inverted orientation of sample during measurement, respectively, and data labels correspond to sample number.

corresponding number of samples,  $N$ , and the number of measurements per sample,  $n$ , performed for each material. In this table the values for  $u_{hom}$  correspond to the standard deviation between  $n \times N$  measurements. It is noted that, for the case of VUHZ cast steel samples, the derived standard deviation between samples

**Table 2**  
Uncertainty contributions from homogeneity study,  $u_{hom}$  (in %), for the ILC materials. ILC.

ILC	Cast steel material	$N$	$n$	$u_{hom}$
ILC_A	Siempelkamp	20	1	1.08
ILC_B	VUHZ	14	2	0.35

from the one way ANOVA on the two groups of data, corresponding to the two measurement orientations, was 0.21%. Uncertainty due to instability was disregarded because it was presumed that the types of the materials do not suffer changes with time or during transport.

### 2.3. Analysis methods

Each of the ILC participants was provided with one sample from each cast steel batch and two technical protocols, one for ILC\_A and one for ILC\_B, containing the relevant information. All participants used gamma-ray spectrometry for measuring the activity, as recommended by the technical protocol, applying different efficiency calibration methods (experimental, Monte Carlo and combined ones). In each laboratory the measurements were performed using one or more HPGe detectors and, in some cases, low level gamma-ray spectrometry (LLGS).

Measurement and calibration approaches for ILC\_A and ILC\_B samples, in any given participant's laboratory, were the same due to the similar geometries and matrix compositions of the two samples. Measurements distances and analysis conditions are summarised in Table 3. The distances from the detector end-cap to the bottom of the sample (SDD) ranged from in-contact to 17 cm.

Table 3 summarizes the methods used by the 13 participants. Twelve participants applied computational efficiency calibration methods (absolute or relative), while one participant used experimental calibration based on a reference source of matching geometry, matrix and nuclide as the sample. Of the ones using computational methods, three participants calculated absolute detector efficiencies, using MC models of their detectors, and nine calculated efficiency correction factors, and applied these factors to the respective experimental efficiencies, obtained using standard sources (relative approach). These correction factors accounted for geometry, matrix and True Coincidence Summing (TCS) effects. Seven of the participants who followed the relative approach used the MC models of their detectors and two applied numerical methods (Korun and Vidmar, 2002; Klemola and Ugletveit, 1997). In total six different MC codes were used, to calculate either detection efficiencies or efficiency and TCS corrections (Salvat et al., 2011; Lépy et al., 2006; Waters, 2002; Sima et al., 2001; Nelson et al., 1985; Jovanović et al., 1997). All MC models used were validated against experimental data obtained using calibration sources. A range of point, filter and voluminous standard sources were used in order to determine the experimental efficiencies where necessary (Table 3).

The densities used by the participants for efficiency and corrections calculations ranged between 7.29 and 7.85 g cm<sup>-3</sup> for Siempelkamp, and between 7.34 and 7.89 g cm<sup>-3</sup> for VUHZ samples. The observed relative standard deviation between the density values in both cases was on the order of 2%, and is explained by the fact that the participants calculated the density of each sample based on their weighed mass and measured dimensions of the sample.

The nuclear decay data used were obtained from Monographie BIPM-5 and the DDEP (Decay Data Evaluation Project) (Bé et al., 2013; Browne et al., 2001).

Each laboratory reported the results of measured activity

**Table 3**  
Analysis methods in participants' laboratories.

Participant	Technique <sup>a</sup>	SDD cm	Calibration method			
			Exp <sup>b</sup>	MC-Abs	MC-Rel	Other-Rel
CIEMAT	GS	10.5, 17.5		PENELOPE/PENNUC		
BEV/PTP	LLGS	0.5		PENELOPE2011		
CEA	LLGS	10.36	VS		ETNA	
CMI	GS	10.6		MCNPX v2.7E		
ENEA	GS	8.2	PS, VS, FS		ETNA V5.5 rev56	
IFIN-HH	GS, LLGS	0.63	VS		GESPECOR v4.2	
IJS	GS		VS			Numerical
IST/CTN	LLGS	0.001, 0.001	VS		GESPECOR	
JRC	LLGS	1.1, 2.1	PS		EGS4	
POLATOM	GS		VS <sup>c</sup>			
PTB	GS	0.7	PS		GESPECOR v4.2	
SMU	GS	16.84	PS		ANGLE	
STUK	LLGS	13.2	VS, FS			DECCA

<sup>a</sup> GS: Gamma spectrometry, LLGS: Low level gamma spectrometry.

<sup>b</sup> PS, VS, FS: point, volume and filter source, respectively.

<sup>c</sup> Calibration source prepared to match sample geometry-matrix-nuclide. No efficiency correction applied.

concentrations,  $A_{lab}$ , and associated combined standard uncertainties,  $u_{lab}$ , in  $\text{Bq g}^{-1}$ , for the reference times specified in the ILC\_A and ILC\_B technical protocols. The participants were asked to provide the uncertainty budgets of their measurement results. Combined standard uncertainties ( $k=1$ ) were reported. The reporting was realised using a distributed Excel reporting template (one each for ILC\_A and ILC\_B) which in addition served as questionnaires accommodating information on employed measurement systems, procedures and methods, and references to data sources. In this way, the evaluation of the results was facilitated and traceability of activity determination was ensured.

#### 2.4. Evaluation criteria/parameters

The reference activity concentration of  $^{60}\text{Co}$  in each batch of samples,  $A_{ref}$ , was taken to be the consensus value of the respective ILC. In each case the consensus value was derived as the power-moderated mean (PMM) (Pommé and Keightley, 2015) of the reported activity concentrations, on the condition that they were not identified as outlier. The standard deviation of the PMM,  $u_{PMM}$ , represented the uncertainty from characterisation,  $u_{char}$ . The uncertainty of the reference activity concentrations,  $u_{ref}$ , incorporated the uncertainty components from characterisation,  $u_{char}$ , and from the inhomogeneity study,  $u_{hom}$ , using the following formula:

$$u_{ref} = \sqrt{u_{PMM}^2 + u_{hom}^2} \quad (1)$$

The individual laboratory performance was expressed in terms of relative deviations  $D_{\%}$  and  $E_n$  numbers (ISO, 2005) of its reported value against the reference value,  $A_{ref}$ , for each batch of samples, using the formulas below:

$$D_{\%} = \frac{A_{lab} - A_{ref}}{A_{ref}} \times 100 \quad (2)$$

where  $A_{lab}$  is the participant's result;

And

$$E_n = \frac{A_{lab} - A_{ref}}{U(d_i)} = \frac{\zeta_i}{2}, \quad (3)$$

where  $U(d_i) = 2u(d_i)$  is the expanded uncertainty of the difference  $d_i = A_{lab} - A_{ref}$  at the 95% level of confidence and  $\zeta_i$  is the zeta-score for the lab  $i$ . Whether  $A_{lab}$  or  $A_{ref}$  are correlated and not correlated,  $u(d_i)$  is calculated using (Eqs. (4) and 5), respectively:

$$u^2(d_i) = (1 - 2w_i)u_{lab,i}^2 + u_{ref}^2, \quad (A_{lab} \text{ included in PMM}) \quad (4)$$

$$u^2(d_i) = u_{lab,i}^2 + u_{ref}^2, \quad (A_{lab} \text{ excluded from PMM}) \quad (5)$$

where  $w_i$  are the normalized weights used in the calculation of the PMM and  $u_{PMM}$ .

$E_n$  numbers are interpreted in the following way: (a)  $|E_n| \leq 1$  indicates a satisfactory performance since the laboratory value is compatible with the reference value; (b)  $1.5 \geq |E_n| > 1$  corresponds to a "warning signal", as the laboratory value differs significantly from the reference value, triggering investigation and correction of sources of deviation; and (c)  $|E_n| > 1.5$ , consists of an "action signal" with a need to investigate and find the sources of the large deviation.

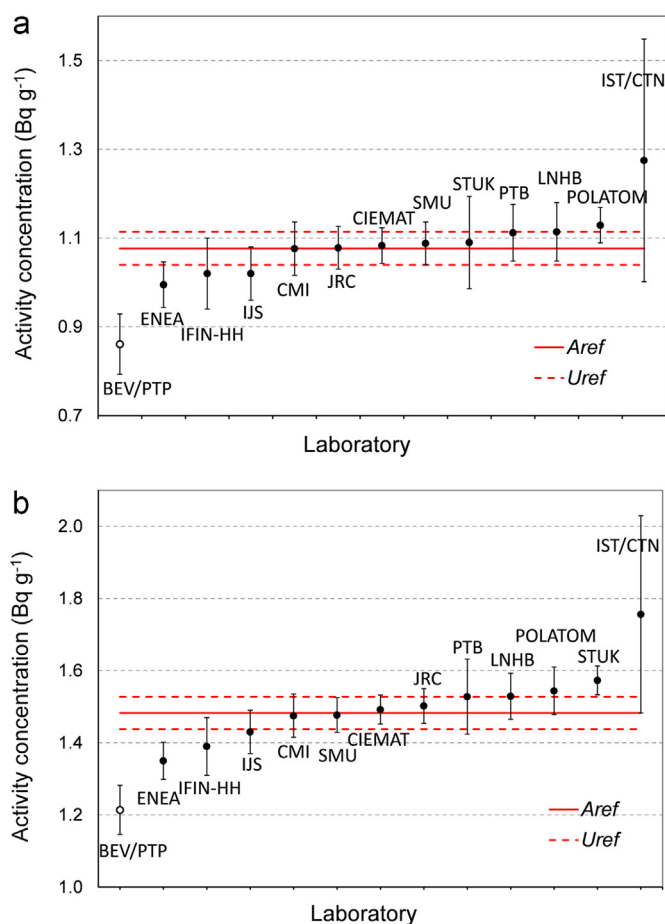
### 3. Results and discussion

The reported activity concentrations of  $^{60}\text{Co}$ ,  $A'_{lab,i}$ , are plotted in ascending order in Fig. 3(a) and (b) for ILC\_A and ILC\_B, respectively. The indicated expanded uncertainties were calculated from the reported combined standard uncertainties, at  $k=2$ . The  $A_{ref}$  shown in these figures was calculated for each ILC (A or B) from all reported activity concentrations except from the result of BEV/PTP. The latter was excluded from the calculation prior to the disclosure of ILCs' results when the participant reported technical problems, namely counting losses, associated with the gamma-ray spectrometer used in their analysis.

The reported uncertainty budgets were analyzed. The minimum and maximum relative uncertainties of partial quantities contributing to the reported combined standard uncertainties, together with the number of reporting participants, are shown in Table 4 for the two ILCs. The highest uncertainty values were reported by the IST/CTN and were on the order of 10% for each ILC. These uncertainties are realistic and, according to the reported uncertainty budgets, their main contribution comes from reproducibility. From Table 4 it can be observed that the combined standard uncertainty values varied by an order of magnitude among the participating laboratories. To further optimize the estimation of uncertainties in gamma-ray spectrometry measurements, dedicated work has been recently published by Cassette et al. (2015) and by Lepy et al. (2015).

The calculated reference activity concentrations, for  $^{60}\text{Co}$  in the Siempelkamp and VUHZ steel samples, and evaluation parameters for both comparisons, are summarised in Table 5. From this table it





**Fig. 3.** Laboratory results for <sup>60</sup>Co activity concentration in (a) Siempelkamp and (b) VUZH cast steel on 1 January 2013 12h00 UT and 1 June 2013 12h00 UT, respectively. Uncertainty bars represent  $U_{lab}$ , calculated from the reported combined standard uncertainties and solid/dashed lines the  $A_{ref} \pm U_{ref}$  ( $k=2$ ). The reference values  $A_{ref}$  are calculated from all reported values except BEV/PTP's (empty data points).

may be observed that 10 out of 13 and 11 out of 13 results of the ILC on Siempelkamp and VUZH cast steel, respectively, showed satisfactory compatibility with the reference activity concentrations for <sup>60</sup>Co. Nevertheless, two results deviated by more than

**Table 4**

Minimum and maximum relative uncertainties (%) of partial quantities contributing to the combined relative uncertainty as reported by participants for the two ILCs.

Uncertainty component	ILC_A (Siempelkamp)			ILC_B (VUZH)		
	Minimum	Maximum	Number of labs	Minimum	Maximum	Number of labs
Counting statistics	0.07	2.50	13	0.02	1.00	13
Weighing	0.0003	0.2	12	0.0000	0.20	12
Background	0.04	50	7	0.04	50	7
Dead/live time	0.0003	0.1	9	0.0001	0.1	10
Pile-up	0.01	0.1	5	0.00	0.1	5
Gamma emission probabilities ( $p_g$ )	0.001	0.3	12	0.00	0.3	12
Half-life	0.01	0.08	12	0.00	0.1	12
Experimental efficiency curve	0.50	2.9	4	0.50	4.3	4
Calibration factor	1	4	7	0.01	4.0	7
Impurities	0.05	0.1	4	0.05	0.1	4
Self-attenuation	0.10	4.6	5	0.1	4.6	6
MC Efficiency simulations	0.20	5	7	0.0001	5.0	7
TCS corrections	0.80	2	5	0.0001	2.0	9
Sample parameters/density	0.40	1.4	4	0.4	1.6	4
Sample position	1.1	1.1	2	1.0	3.0	2
Uncertainty of activity standards	0.8	0.8	1	1.1	1.1	1
Spread of different measurements	0.8	8.6	2	0.9	8.2	2
Combined relative std uncertainty	1.75	10.7	13	1.7	10.3	13

**Table 5**  
Summary of ILCs evaluation parameters.

ILC cast steel	$A_{ref}^a$ Bq g <sup>-1</sup>	No of labs with					
		$ D_{rel} $			$ E_{rel} $		
		≤ 5%	> 5% & ≤ 10%	> 10%	≤ 1	> 1 & ≤ 1.5	> 1.5
ILC_A (Siempelkamp)	1.077(19)	8	3	2	10	2	1
ILC_B (VUZH)	1.483(22)	8	3	2	11	-	2

<sup>a</sup> Reference dates: 1st January 2013 12h00 UT for ILC\_A and 1st June 2013 12h00 UT for ILC\_B samples.

10% from the reference values in each case. One of these two participants was the BEV/PTP, who reported results lower than the reference values (by -20% and -18% for ILC\_A and ILC\_B, respectively) due to technical problems associated with count losses in their detector. These technical issues were reported prior to the disclosure of the ILCs results, and were excluded from calculation of the reference activity concentrations. The reference activity concentrations of <sup>60</sup>Co in the Siempelkamp and VUZH cast steel standards were  $(1.077 \pm 0.019)$  Bq g<sup>-1</sup> on 1 January 2013 12h00 UT and  $(1.483 \pm 0.022)$  Bq g<sup>-1</sup> on 1 June 2013 12h00 UT, respectively.

A comparison between Fig. 3(a) and (b) reveals a similar pattern. For any individual participant, the same efficiency calibration appeared to have been used for the analysis of the two samples as the same radionuclide was determined in samples of similar dimensions and matrix compositions.

Of particular importance for the accurate determination of <sup>60</sup>Co activity concentration in these cast steel samples was the use of MC simulation software which enabled the calculation of the true coincidence-summing corrections of the detection efficiency for the nuclide. These corrections become particularly significant for close measurement geometries such as used in these ILCs.

#### 4. Conclusions

A European comparison on measurement of <sup>60</sup>Co activity in steel matrix was conducted and led to the activity characterisation of two steel reference standards developed in the MetroMetal joint research project.

For each of the a) originally contaminated and b) spiked batch of cast steel samples, used in the two stages of the comparison, a consensus value was derived for the activity concentration of  $^{60}\text{Co}$  in the material. In each case, the consensus value was calculated as the power-moderated mean of the reported results by the 13 participants in accordance with the official CCRI(II) procedure for key comparisons. One result was excluded from the calculation of the consensus value of each ILC on the basis of reported technical problems associated with the detector in the respective laboratory. The consensus values for the activity concentrations of  $^{60}\text{Co}$  in the originally contaminated (Siempelkamp) and in the spiked (VUHZ) cast steel samples were assigned as reference values of the two respective standards. These reference activity concentrations were  $(1.077 \pm 0.019) \text{ Bq g}^{-1}$  on 1 January 2013 12h00 UT and  $(1.483 \pm 0.022) \text{ Bq g}^{-1}$  on 1 June 2013 12h00 UT, respectively.

Ten out of 13 and 11 out of 13 results of the ILC on Siempelkamp and VUHZ cast steel, respectively, agreed within  $2\sigma$  with the ILCs' reference values. The overall comparison between the results of the two ILCs revealed a similar pattern resulting from the determination of the same radionuclide in samples of similar dimensions and matrix compositions.

It should be noted that these comparisons involved measurements of radioactivity in a high-density matrix. High resolution gamma-ray spectrometry in combination with a computational approach for efficiency calibration is the proposed method for such measurements, in particular when standards of the same matrix, shape/size and radionuclide content are not available. In such case, special consideration should be taken of the fact that the method is sensitive to the simulated sample parameters due to the self-attenuation of the gamma rays in the volume of a high density sample. A critical point is to calculate the density of the simulated material from the given mass and measured dimensions of the sample rather than using the nominal density of the material, valid for all sample types.

In this work the ILC results demonstrated the validity of the proposed methods by MetroMetal for the measurement of  $^{60}\text{Co}$  in a cast steel matrix. However, the evaluated approach provides a versatile tool for calibration of systems for a range of sample matrices encountered in metal works (e.g., steel, slag and fume dust).

The two new series of metrologically traceable activity standards of  $^{60}\text{Co}$  in cast steel matrix are available to end users for efficiency calibration of gamma-ray spectrometers in the metallurgical sector for accurate radioactivity monitoring of the steel products.

## Acknowledgements

This work was supported by the EMRP joint research project 'Ionising radiation metrology for the metallurgical industry' (MetroMetal) which has received funding from the European Union on the basis of Decision No 912/2009/EC. The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union. The authors are grateful to all contributors to this work at the participants' laboratories and in particular to Carlos Oliveira, Pierluigi Carconi, Teemu Siiskonen, Franz-Josef Maringer as well as to Maria Sahagia and Thierry Branger who in addition reviewed thoroughly and provided valuable suggestions on the manuscript. The authors gratefully acknowledge the work of Belén Gómez-Mancebo and Fernando Borlaf, from Unidad de

Espectroscopia from CIEMAT, who performed the steel composition analysis. Siempelkamp Nukleartechnik GmbH, Krefeld and VUHZ are thankfully acknowledged for providing the contaminated materials. The IFIN-HH research work was co-funded by the Program Nucleu, project no. PN 09 37 02 05, representing the Romanian national support for the project MetroMetal.

## References

- Bé, M.M., Chisté, V., Dulieu, C., Mougeot, X., Chechev, V.P., Kondev, F.G., Nichols, A.L., Huang, X., Wang, B., France, 2013. Monographie BIPM-5 – Table of Radionuclides (Vol. 7–A= 14 to 245). ISBN-13 978-92-822-2248-5 (<http://www.bipm.org/fr/publications/monographie-ri-5.htm>).
- Browne, E., Bé, M.M., MacMahon, D., Helmer, R.G., 2001. Report on the Activities of the Decay Data Evaluation Project (DDEP). CEA-R-5990(E) Saclay ([http://www.nucleide.org/DDEP\\_WG/DDEPdata.htm](http://www.nucleide.org/DDEP_WG/DDEPdata.htm)).
- Cassette, P., Bochud, F., Keightley, J., 2015. Example of Monte Carlo uncertainty assessment in the field of radionuclide metrology. *Metrologia* 52 (3), S42–S50.
- García-Toraño, E., Peyres, V., Caro, B., Roteta, M., Arnold, D., Burda, O., Ioan, M.-R., De Felice, P., 2015. A novel radionuclide specific detector system for the measurement of radioactivity at steelworks. *J. Radioanal. Nucl. Chem.* 305, 293–298.
- García-Toraño, E., Tzika, F., Burda, O., Peyrés, V., Mejuto, M., Crespo, T., Wätjen, U., Arnold, D., Sochor, V., Svec, A., Carconi, P., De Felice, P., Tecl, J., 2014. Ionising radiation metrology for the metallurgical industry. *Int. J. Metrol. Qual. Eng.* 5 (010), 301. <http://dx.doi.org/10.1051/ijmqe/2014010>. (<http://www.siempelkamp.com/index.php?id=2003&L=0>) (last retrieved July 2015).
- (<http://www.vuhz.cz/>) (last retrieved July 2015).
- IAEA press release 2002/09, 24 June 2002. Inadequate Control of World's Radioactive Sources, Vienna, Available online at (<https://www.iaea.org/newscenter/pressreleases/inadequate-control-worlds-radioactive-sources>).
- IAEA SSS, SSG-17, 2012. Control of Orphan Sources and Other Radioactive Material in the Metal Recycling and Production Industries, Vienna.
- Ionising Radiation Metrology for the Metallurgical Industry. (<http://projects.ciemat.es/web/metrometal>) (last retrieved July 2015).
- ISO, 2005. ISO 13528:2005 – Statistical Methods for Use in Proficiency Testing by Interlaboratory Comparisons, Geneva, Switzerland.
- Jovanović, S., Dlačić, A., Mihaljević, N., Vukotić, P., 1997. ANGLE: a PC-code for semiconductor detector efficiency calculations. *J. Radioanal. Nucl. Chem.* 218 (1), 13–20.
- Klemola, S., Ugleitveit, F., 1997. A method for computer calculation of detector efficiencies: validation and applications. *Trans. Am. Nucl. Soc.* 76, 130–131.
- Korun, M., Vidmar, T., 2002. Calculation of detector responses to photons from extended sources. In: IAEA-TECDOC-1275 on Specialized Software Utilities for Gamma Ray Spectrometry, Vienna.
- Lepy, M.C., Pearce, A., Sima, O., 2015. Uncertainties in gamma-ray spectrometry. *Metrologia* 52 (3), S123–S145.
- Lépy, M.C., Brun, P., Collin, C., Plagnard, J., 2006. Experimental validation of coincidence summing corrections computed by the ETNA software. *Appl. Radiat. Isot.* 64 (10–11), 1340–1345.
- Nelson, W.R., Hirayama, H., Rogers, D.W.O., 1985. EGS4 User Manual. SLAC-265 Appendix 2, Stanford Linear Accelerator Center, Stanford.
- Pommé, S., Keightley, J., 2015. Determination of a reference value and its uncertainty through a power-moderated mean. *Metrologia* 52 (3), S200–S212.
- Salvat, F., Fernández-Varea, J.M., Sempau, J., France, 2011. PENELOPE-2011: A code System for Monte Carlo Simulation of Electron and Photon Transport. OECD/NEA Data Bank, Issy-les-Moulineaux, Available in PDF format from (<http://www.nea.fr/lists/penelope.html>).
- Sima, O., Arnold, D., Dovlete, C., 2001. GESPECOR – a versatile tool in gamma-ray spectrometry. *J. Radioanal. Nucl. Chem.* 248 (2), 359–364.
- Šolc, J., Dryák, P., Moser, H., Branger, T., García-Toraño, E., Peyrés, V., Tzika, F., Lutter, G., Capogni, M., Fazio, A., Luca, A., Vodenik, B., Oliveira, C., Saraiva, A., Szucs, L., Dziel, T., Burda, O., Arnold, D., Martinkovič, J., Siiskonen, T., Mattila, A., 2015. Characterization of a radionuclide specific laboratory detection system for the metallurgical industry. *Radiat. Phys. Chem.* 116, 189–193.
- Tzika, F., Hult, M., Burda, O., Arnold, D., Sibbens, G., Caro Marroyo, B., Gómez-Mancebo, M.B., Peyrés, V., Moser, H., Ferreux, L., Šolc, J., Dryák, P., Fazio, A., Luca, A., Vodenik, B., Reis, M., Tyminski, Z., Klemola, S., 2015. Interlaboratory comparison on  $^{137}\text{Cs}$  activity concentration in fume dust. *Radiat. Phys. Chem.* 116, 106–110.
- UN/ECE/TRADE/278, 2002. Report on the Improvement of the Management of Radiation Protection Aspects in the Recycling of Metal Scrap, ISBN 92-1-116789-2, Geneva.
- Waters, L.S. (Ed.), 2002. MCNPX user's manual, Version 2.3.0. Los Alamos National Laboratory Report LA-UR-02-2607.