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Evaluation of the accidental coincidence counting rates in TDCR counting

Chavdar Dutsov [a,*], Philippe Cassette [b], Benoît Sabot [b], Krasimir Mitev [a]

[a] Sofia University "St. Kliment Ohridski", Faculty of Physics, 5 James Bourchier Blvd, 1164, Bulgaria
[b] CEA, LIST, Laboratoire National Henri Becquerel, LNE-LNHB, 91191, Gif- sur-Yvette Cedex, France

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ABSTRACT

This paper presents analytical and experimental methods to evaluate the accidental coincidence counting rates in a Triple-to-Double Coincidence Ratio (TDCR) Liquid Scintillation (LS) measurement. The experimental method we propose is based on the analysis of the distribution of the time delays between the first detected events in each photomultiplier tube. The underlying assumption is that events separated by several microseconds in time are not correlated, thus the accidental coincidence counting rates could be determined from the time interval distribution of uncorrelated events. The analytical evaluation of the accidental coincidence counting rates is based on the conditional probabilities for the occurrence of uncorrelated events within the same coincidence resolving time. The analytical and experimental evaluations of the rate of accidental coincidences give consistent results for TDCR measurements of ^{3}H, ^{55}Fe and ^{14}C. The two methods were used to evaluate corrections for accidental coincidences for Monte Carlo (MC) generated list-mode files of ^{3}H measurements with increasing activities. The counting rates, corrected for accidental coincidences using the analytical method, are within 0.29% of the MC reference up to 100 kBq and the corrected, using the experimental method, are within 0.21% up to 200 kBq.

1. Introduction

The TDCR method is widely used for primary standardization of α-, β- and some electron-capture radionuclides [1,2]. The application of the TDCR method requires the use of specialized counters with three photomultiplier tubes (PMTs) and electronics that apply extending type dead-time and record coincidence events between three PMTs (triple coincidences) and pairs of PMTs (double coincidences). A common problem of systems of multiple detectors working in coincidence is the possibility of two or more unrelated events to occur within the same coincidence resolving time, thus resulting in an accidental coincidence.

Recent advances in the digital electronics have allowed the use of longer, user-selectable coincidence resolving times by the use of fast digitizers [3] or FPGA-based acquisition systems [4–6]. With the use of long coincidence resolving times, the rate of accidental coincidences increases and an accurate calculation of their contribution is required. Moreover, a recent trend is observed towards the development of miniature portable TDCR counters aiming to perform in-situ metrology of LS-sources used in the nuclear energy and medical fields [7,8] where the measured sources could have very large activities. In such cases accidental coincidences cannot be neglected.

The aim of this paper is to present an experimental method as well as an analytical expression for the evaluation of the accidental coincidences counting rates in TDCR measurements.

2. Materials and methods

Experimental evaluation. Due to the life-time of the excited states in the scintillator, the scintillation events are detected at different times with respect to the moment of radioactive decay. It has been observed that for high-energy emitters, like ^{18}F (E_β max. 633 keV), the first detected events in each PMT (primary events) are grouped within 16 ns, but for lower energy emitters, like ^{3}H (E_β max. 18.6 keV) the spread of events is much larger, with reports up to 250 ns [9] and above 300 ns [10]. Larger time differences between primary events in true coincidences could still be observed, but the probability for their occurrence is decreasing with time and, thus, after a few microseconds, the contribution of such events can be neglected. In this case, it can be assumed that primary events separated by several microseconds in time are produced by uncorrelated events, such as two uncorrelated radioactive decays, electronic noise events or background events.

The experimental method for evaluation of accidental coincidences is based on the analysis of the distribution $\varphi_i(t)$ of the time differences Δt between the first primary event and last primary event in a given coincidence channel ($i = AB, BC, AC, D, T$). Fig. 1 illustrates how the time difference Δt is calculated in different cases for the AB, D and T channels. Assuming a common dead-time detector (as the MAC3 module [11]), the distribution of the rate of events $f_i(t)$ with a given

* Corresponding author.
E-mail address: ch.dutsov@phys.uni-sofia.bg (C. Dutsov).

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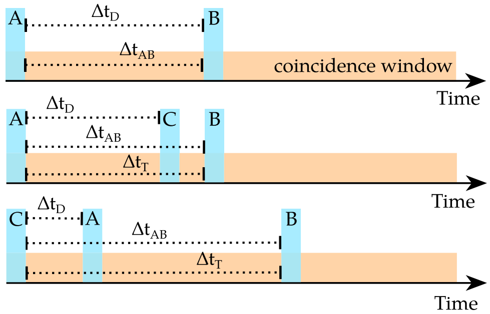

Fig. 1. Time differences between the first and last primary events in the AB, D and T channels. Note that, in the third case, the C event is the first primary event in both the AB and T channels because the T coincidences are a subset of the AB coincidences.

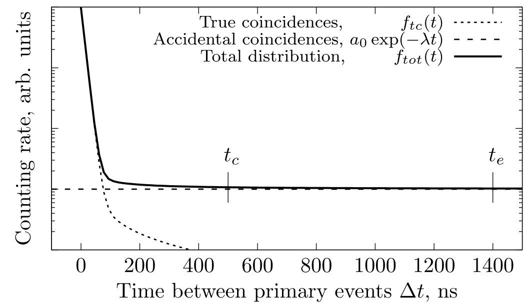

Fig. 2. The measured distribution of the time between first and last primary event in a given coincidence channel is a linear combination of the distribution of the true and accidental coincidences.

time difference t for a given coincidence channel i can be defined as:

$$f_i(t) = \frac{\varphi_i(t)}{L}, \tag{1}$$

where L is the live-time of the detector. The distribution $f_i(t)$, hereafter referred to as time interval distribution, is defined in such a way that:

$$n_i = \int_0^\tau f_i(t)dt, \tag{2}$$

where n_i is the counting rate in channel i that would be recorded by a TDCR detector with coincidence resolving time τ.

The experimentally observed time interval distribution for a given coincidence channel is the sum of the distribution of the true coincidences f_{tc} and the distribution of the accidental coincidences f_{acc} (illustrated in Fig. 2). As the events forming the accidental coincidences are uncorrelated and, if it can be assumed that their detection is a Poisson process, the time interval between them is exponentially distributed. The total coincidence counting rate in a given channel f_{tot} as a function of the time difference between primary events can then be expressed here as:

$$f_{tot}(t) = f_{tc}(t) + a_0 e^{-\lambda t}, \tag{3}$$

where a_0 is the rate of accidental coincidences at $t = 0$ and λ is the rate of detected events in the coincidence channel. For sufficiently large time differences t, the probability of true coincidences will tend towards zero. In that case the total distribution will be determined only by the distribution of the accidental coincidences:

$$f_{tot}(t) = a_0 e^{-\lambda t} \text{ for } t > t_c, \tag{4}$$

where t_c is the cut-off time difference, after which the probability for true coincidences can be considered negligible.

In most practical cases the measured counting rates are lower than the order of 10^4 s^{-1} and the width of the analyzed time interval distribution (t_e) is less than 2×10^{-6} s, thus the argument of the exponent will generally be less than 2×10^{-2}. In such a case it is acceptable to approximate the exponent with only the linear terms in the Taylor series:

$$f_{acc}(t) = a_0(1 - \lambda t). \tag{5}$$

The equation can be simplified further under assumptions for lower counting rates, where the distribution $f_{acc}(t)$ can be considered uniform:

$$f_{acc}(t) = a_0 \text{ for } t < t_e, \tag{6}$$

where t_e should be short enough for λt_e to be considered negligible.

The parameters of the distribution of the accidental coincidences λ and a_0 can be estimated by fitting f_{acc} from Eqs. (4), (5) or (6) to the experimentally obtained time interval distribution in the interval (t_c, t_e). The contribution of the accidental coincidences to the total counting rate then can be calculated as the integral of the distribution of accidental coincidences within the coincidence resolving time:

$$a_i = \int_{t=0}^\tau f_{acc}^{(i)}(t)dt. \tag{7}$$

where $f_{acc}^{(i)}(t)$ is the distribution of accidental coincidences of the ith coincidence channel (AB, BC, AC, D or T).

The main advantage of the experimental evaluation is that the only required assumption is that primary events separated by several microseconds are uncorrelated. The validity of the assumption can be checked when obtaining the time interval distributions for the coincidence channels.

Analytical evaluation. The analytical evaluation of the accidental coincidence counting rates is based on the conditional probabilities for the occurrence of uncorrelated events within the same coincidence resolving time.

In a three PMT detector we can distinguish many channels of scintillation events, defined in [12] as: three single event channels (A, B, C) and their logical sum ($S = A \vee B \vee C$), three double coincidence channels ($AB = A \wedge B, BC = B \wedge C, AC = A \wedge C$), the logical sum of the double coincidences channel ($D = AB \vee BC \vee AC$) and the triple coincidence channel ($T = A \wedge B \wedge C$), where $\wedge$ is the logical "and" operator and $\vee$ is the logical "or" operator. In this notation, a correlation exists between the channels. For example, all events in the T channel are also included in the S, D, AB, BC, AC, A, B and C channels and all AB events are also A, B and S events.

Another set of channels with uncorrelated events can be constructed from this one as: three pure single event channels (PA, PB, PC) that exclude doubles and triples: $PA = A \wedge \neg(B \vee C)$ with similar expressions for the other two channels, where $\neg$ is the logical "not" operator; three pure double coincidence channels (PAB, PBC, PAC) that exclude triples, where $PAB = AB \wedge \neg C$ with similar expressions for the other two channels; the logical sum of the pure single events channel ($PS = PA \vee PB \vee PC$); the logical sum of pure double coincidences channel ($PD = PAB \vee PBC \vee PAC$) and the pure triple coincidence channel ($PT = T$). For example, the pure channels PA and PBC are shown circumscribed by dash lines in Fig. 3.

The counting rates p_i in the uncorrelated set of channels ($PA, \dots, PAB, \dots, PS, PD, PT$) can be estimated as:

$$\begin{aligned}
p_A &= n_A - n_{AC} - n_{AB} + n_T \\
p_B &= n_B - n_{AB} - n_{BC} + n_T \\
p_C &= n_C - n_{AC} - n_{BC} + n_T \\
p_{AB} &= n_{AB} - n_T \\
p_{BC} &= n_{BC} - n_T \\
p_{AC} &= n_{AC} - n_T \\
p_S &= p_A + p_B + p_C = n_S - n_D \\
p_D &= p_{AB} + p_{BC} + p_{AC} = n_D - n_T \\
p_T &= n_T
\end{aligned} \tag{8}$$

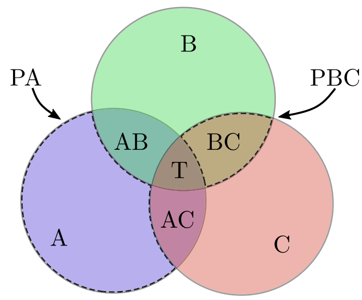

Fig. 3. Illustration of the relationship between the uncorrelated and correlated coincidence channels. The areas representing PA and PBC are surrounded with dashed lines.

where n_i are the counting rates in the single and coincident channels ($A, B, C, S, AB, BC, AC, D$ and T, respectively).

The rate of accidental coincidences N_a between two uncorrelated channels with counting rates N_1 and N_2 is given by [13]:

$$N_a = 2\tau(N_1 N_2), \tag{9}$$

where τ is the coincidence resolving time of the detector.

The accidental coincidences in a given channel can be evaluated using (9) by summing all contributions from coincidences between events occurring within the same resolving time in two different uncorrelated channels.

There are two types of contributions, the first consisting of uncorrelated events that, when detected in the same coincidence resolving time, are registered as an event from another channel. For example, a PAB and a PBC event detected during the same resolving time will be falsely registered as a T event as well. Note here, that the same logic cannot be used for the AB and BC channels, as some events in them are true T coincidences.

The second type of accidental coincidences in a given channel are uncorrelated events of that channel that are registered during the resolving time started by an event in another channel. For example, the case where an event in channel PC starts the coincidence resolving time and an event in channel PAB is registered after. The PAB event will contribute to the accidental coincidences in channel AB as it is registered during the coincidence window started by an uncorrelated channel. As the time of arrival of these events is immutable in the context of accidental coincidences, the factor of 2 must be omitted in (9).

For example, the accidental coincidences in the AB channel can be evaluated by summing all possible contributions from the two types of accidental coincidences. The first type contributions in the case of channel AB are:

1. PA in the same window as PB: $2\tau(p_A p_B)$
2. PA in the same window as PBC: $2\tau(p_A p_{BC})$
3. PB in the same window as PAC: $2\tau(p_B p_{AC})$
4. PAC in the same window as PBC: $2\tau(p_{AC} p_{BC})$

and the second type contributions are:

1. PAB in the window started by a non PAB event: $\tau(p_A + p_B + p_C + p_{BC} + p_{AC})p_{AB}$
2. PT in the window started by a non PAB event: $\tau(p_A + p_B + p_C + p_{BC} + p_{AC})p_T$

The accidental coincidence counting rates a_i in the other channels can be evaluated using the same considerations and are expressed as:

$$\begin{aligned}
a_{AB} =& \left[2\left(p_A p_B + p_A\, p_{BC} + p_B p_{AC} + p_{AC} p_{BC}\right)\right.\\
&\left. + \left(p_S + p_D - p_{AB}\right)\left(p_{AB} + p_T\right)\right]\tau\\
a_{BC} =& \left[2\left(p_B p_C + p_B\, p_{AC} + p_C p_{AB} + p_{AB} p_{AC}\right)\right.\\
&\left. + \left(p_S + p_D - p_{BC}\right)\left(p_{BC} + p_T\right)\right]\tau\\
a_{AC} =& \left[2\left(p_A p_C + p_A\, p_{BC} + p_C p_{AB} + p_{AB} p_{BC}\right)\right.\\
&\left. + \left(p_S + p_D - p_{AC}\right)\left(p_{AC} + p_T\right)\right]\tau\\
a_D =& \left[2\left(p_A\, p_B + p_B\, p_C + p_C\, p_A\right) + p_S(p_D + p_T)\right]\tau\\
a_T =& \left[2\left(p_A\, p_{BC} + p_B\, p_{AC} + p_C\, p_{AB}\right) + \left(p_S + p_D\right) p_T\right.\\
&\left. + 2\left(p_{BC}\, p_{AB} + p_{AC}\, p_{BC} + p_{AC}\, p_{AB}\right)\right]\tau
\end{aligned} \tag{10}$$

The analytical estimation of the accidental coincidences can be applied to all existing TDCR acquisition systems that provide information about the single, double and triple counting rates. In practice, the coincidence counting rates n_{AB}, n_{BC}, n_{AC} and n_T reported by the detector, used in to calculate the pure counting rates, already include the accidental coincidences. Thus, if the contribution of the accidental coincidences to the total measured coincidences is large, this could introduce a bias when equations (10) are used. To reduce this effect it is necessary to perform measurements of the studied LS-source with coincidence resolving time, which is large enough not to lose real coincidences, but not too large to increase the accidental coincidence counting rates. Second and higher order accidental coincidences, for example, uncorrelated PA, PB and PC events arriving within the same coincidence resolving time and producing a T coincidence, have negligible contribution and thus are not considered. These approximations and considerations are not necessary to be taken into account when applying the experimental method for the evaluation of the accidental coincidences, thus the analytical approach should be preferred only when their contribution is not overwhelming. Thus far, no methods for the estimation of the uncertainty of the evaluated accidental coincidence counting rates has been developed and this will be the subject of future studies.

3. Results

In order to validate the two methods, two ^{3}H (UltimaGold in Polyethylene vial), one ^{55}Fe (UltimaGold in Polyethylene vial) and one ^{14}C (UltimaGold in glass vial) LS-sources were measured. Two of the sources, the high-activity ^{3}H and ^{55}Fe, were also measured with a 75% transparent gray filter. The measurements were performed with a portable TDCR counter connected to a CAEN DT5751 Digitizer with 1 GS/s sampling rate, working in list-mode — i.e., recording the timestamp of each detected event in all three PMTs in text files in the memory of a computer. The list-mode files were analyzed off-line by a dedicated software, which can apply the common dead-time logic with user-selectable dead-time and coincidence resolving time. The code can also be used to obtain the time interval distributions in all the coincidence channels.

Application of the experimental method. In order to illustrate the application of the experimental method, the time interval distributions in the D channel for the higher activity 23 kBq ^{3}H source without filter and the 6.2 kBq ^{14}C source are shown in Fig. 4. Due to the time spread of true events, the region where the total time interval distribution is dominated by accidental coincidences is considered in the interval 400 ns to 3000 ns for the ^{14}C measurement and 1300 ns to 3000 ns for the ^{3}H measurement. As the D counting rates for both sources are not very high, the exponential distribution of the accidental coincidences can be approximated well with a uniform distribution in the studied time interval. In such a case, Eq. (6) can be used. The D accidental coincidence counting rate per nanosecond resolving time evaluated using the experimental method is 0.107 s^{-2} for the ^{3}H source

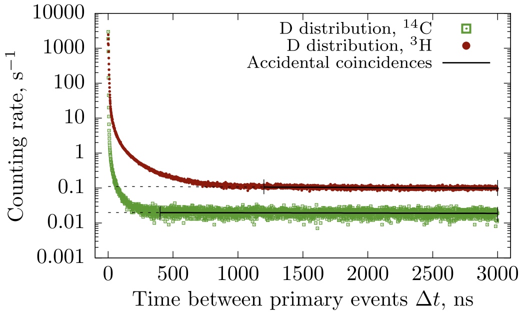

Fig. 4. Time interval distributions of a ^{3}H and ^{14}C LS-sources in the D channel. The solid lines show the average value of the plateau formed by the coincidences between uncorrelated events.

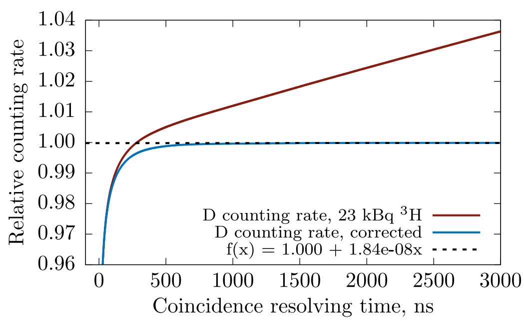

Fig. 5. Counting rate in the D channel as a function of the resolving time for the high-activity ^{3}H source without and with correction for accidental coincidences using the experimental method. The counting rates are normalized by the corrected value at 2 μs. The dashed line shows a linear fit after 1.3 μs.

and 0.018 s^{-2} for the ^{14}C source. The same methodology can be applied to all coincidence channels.

The same list-mode measurement of the 23 kBq ^{3}H LS-source was analyzed multiple times with the dedicated software in order to obtain the coincidence counting rates for a number of resolving times from 40 to 3000 ns. The ratio of the D counting rate at a given resolving time to the D counting rate at 2 μs resolving time is shown in Fig. 5 (solid red line). For coincidence resolving times less than 500 ns a significant loss in the D counting rate can be observed due to the large time spread of scintillation events. For higher coincidence resolving times, a constant increase in the counting rate can be seen, which is caused by the increasing probability for accidental coincidences. The counting rates were corrected by subtracting the accidental coincidences obtained from the experimental method assuming a uniform distribution (solid blue line). After a long enough coincidence resolving time that includes all correlated events, no further increase in the corrected counting rate is observed for larger coincidence resolving times.

Comparison of experimental and analytical methods. In order to compare the experimental and analytical evaluations of the accidental coincidences, the same list-mode files from the measurements of the four sources were processed with a computer code that applies the common extending type dead-time counting logic. The software was set to 40 μs dead-time base duration and the resolving time necessary to include all true coincidences, which was found to be 100 ns for ^{14}C and 800 ns for ^{3}H and ^{55}Fe. The necessary resolving time, which was used for each LS-source, was determined by analysis of the corrected counting rates as a function of the resolving time in each of the coincidence channels. In the example given in Fig. 5, to obtain 99.9% of the D counting rate at 2 μs, the needed resolving time is 550 ns and to obtain 99.95% it is 800 ns. The complete analysis of the influence of the resolving time on the counting rates is outside the scope of this article and will be described elsewhere. The obtained single counting rates and double and triple coincidence counting rates were used with Eqs. to calculate the accidental coincidence counting rates in each coincidence channel, according to the analytical method.

To apply the experimental method, the time interval distributions for each coincidence channel was analyzed for each measurement and the accidental coincidences were evaluated using Eq. (7) under the assumption for uniform distribution in the interval from 2000 ns to 2500 ns. The comparison of the accidental coincidence counting rates obtained with the experimental and analytical methods is presented in Table 1. The corrected counting rate is calculated by subtracting the estimated rate of accidental coincidences from the measured counting rates. The difference between the results obtained with the two methods (Δ) is calculated as:

$$\Delta = N_{\mathrm{calc}}/N_{\mathrm{expt}} - 1, \tag{11}$$

where N_{calc} is accidental coincidence counting rate as estimated by the analytical approach and N_{expt} by the experimental method.

The estimates by the analytical and experimental methods for the rate of accidental coincidences agree within 3.30% for the measurements of the high-activity ^{55}Fe and ^{3}H sources. The differences seem to be larger for the measurements with filter compared to the measurements without a filter. The two methods give identical results for the measured ^{14}C source. The agreement between the methods is satisfactory for the 2.4 kBq ^{3}H source where the accidental coincidences counting rate is low and thus prone to statistical fluctuations.

Despite some differences in the accidental coincidence counting rates which were estimated by the two methods, an excellent agreement between the corrected counting rates can be observed. Here, the relative deviations are below 0.13% in all studied cases and below 0.02% for the higher efficiency measurements without a filter. It is important to note that, considering the ^{3}H sources without a filter, a 10 times increase in ^{3}H activity leads to 50 times increase in T and 20 times increase in D accidental coincidence counting rate. Also, if the measurements of the high-activity ^{3}H source with and without a filter are compared, the relative contribution of the accidental coincidences in the D channel increases from 0.82% to 1.73%. Similar behavior can be observed for the high-activity ^{55}Fe sources, where for the measurement without a filter the contribution of the accidental coincidences is 0.66% and for the measurement with filter it increases to 1.93%.

Dependence of the analytical estimation on the resolving time. As the analytical estimation of the accidental coincidences depends on their contribution, which increases with increase in the coincidence resolving time, it is interesting to study whether this dependence is significant. To do so, the list-mode measurement of the 23 kBq ^{3}H LS-source was analyzed with 40 μs dead-time base duration and resolving times from 200 ns to 3000 ns. The results from the experiment are presented in Table 2. The single and coincident counting rates obtained with different resolving times were used with Eqs. to calculate the accidental coincidences (Method: Calc.) as well as the true counting rates (accidental coincidences subtracted from the measured counting rates). The D and T time interval distributions of the source were analyzed and Eq. (7) was used under the assumption for uniform distribution in the interval between 2000 ns and 2500 ns, to evaluate the accidental coincidences (Method: Expt.). As the experimental evaluation of the accidental coincidences do not depend on the choice of coincidence resolving time, the results from this method are used as a reference. The difference between the two is calculated using Eq. (11). No significant difference in the true D and T counting rates for the resolving times between 200 ns and 3000 ns can be seen for the measured source and the two methods agree within 0.1% for all studied resolving times.

Monte Carlo Simulation. A major disadvantage of the experimental validations of the proposed methods for the evaluation of the accidental coincidence counting rates is that, in all real world measurements, the true coincidence counting rates are unknown. In order to circumvent, this we have used a Monte Carlo (MC) code for generating realistic

Table 1
Comparison of the analytical (Correction method: Calc.) and the experimental (Correction method: Expt.) methods for the calculation of accidental coincidences. The values a_i/n_i show the relative contribution of the accidental coincidences a_i, determined from the experimental method, to the measured (corrected + accidental) counting rates n_i.

Nuclide	TDCR	Correction method	Accidental coincidences, s^{-1}			Corrected counting rate, s^{-1}		
			AB[a]	T	D	AB[a]	T	D
^{3}H 23 kBq[b]	0.3998	Calc.	84.74	85.82	83.49	5986.8	4055.9	10145.2
		Expt.	85.24	86.34	83.59	5986.3	4055.4	10145.1
		Δ	−0.59%	−0.60%	−0.12%	0.01%	0.01%	0.00%
		a_i/n_i				1.40%	2.08%	0.82%
	0.2080 (with filter)	Calc.	54.63	37.43	83.52	2209.8	981.5	4719.5
		Expt.	54.44	37.09	83.29	2210.0	981.8	4719.7
		Δ	0.36%	0.93%	0.27%	−0.01%	−0.04%	0.00%
		a_i/n_i				2.4%	3.64%	1.73%
^{3}H 2.4 kBq	0.4018	Calc.	2.57	1.67	4.39	548.5	370.2	921.4
		Expt.	2.57	1.72	4.63	548.5	370.2	921.2
		Δ	0.00%	−3.27%	−5.15%	0.00%	0.02%	0.03%
		a_i/n_i				0.47%	0.46%	0.50%
^{55}Fe 24 kBq[b]	0.2809	Calc.	40.07	38.98	41.99	3300.1	1806.0	6429.4
		Expt.	40.24	39.04	42.69	3300.0	1806.0	6428.7
		Δ	−0.42%	−0.14%	−1.63%	0.01%	0.00%	0.01%
		a_i/n_i				1.20%	2.12%	0.66%
	0.1248 (with filter)	Calc.	25.65	14.03	48.80	989.1	299.5	2399.6
		Expt.	25.26	13.63	47.24	989.5	299.9	2401.1
		Δ	1.54%	2.93%	3.30%	−0.04%	−0.13%	−0.06%
		a_i/n_i				2.49%	4.35%	1.93%
^{14}C 6 kBq	0.9315	Calc.	1.93	1.82	1.94	5651.3	5513.3	5918.6
		Expt.	1.93	1.82	1.94	5651.3	5513.3	5918.6
		Δ	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%
		a_i/n_i				0.03%	0.03%	0.03%

[a]The results for the *BC* and *AC* channels were found to be similar and were omitted for brevity.

[b]The same source was measured twice, first without a filter (upper row) and a second time with a 75% transparent gray filter (lower row).

Table 2
Comparison between the analytically calculated (Calc.) and experimentally (Expt.) obtained *D* and *T* accidental and true coincidence counting rates at different resolving times.

Resolv. time, ns	Corr. method	D acc., s^{-1}	D true, s^{-1}	T acc., s^{-1}	T true, s^{-1}
200	Calc.	21.3	10056.5	21.3	3994.2
	Expt.	20.9	10056.8	21.6	3993.9
	Δ	1.69%	0.00%	−1.38%	0.01%
800	Calc.	83.5	10145.2	85.8	4055.9
	Expt.	83.6	10145.1	86.3	4055.4
	Δ	−0.12%	0.00%	−0.60%	0.01%
1000	Calc.	104.1	10148.5	107.4	4057.8
	Expt.	104.5	10148.1	107.9	4057.3
	Δ	−0.41%	0.00%	−0.49%	0.01%
2000	Calc.	205.4	10155.0	215.8	4059.3
	Expt.	209.0	10151.5	215.8	4059.2
	Δ	−1.71%	0.04%	0.00%	0.00%
3000	Calc.	304.2	10158.2	325.0	4055.8
	Expt.	313.5	10149.0	323.8	4057.1
	Δ	−2.94%	0.09%	0.39%	−0.03%

time sequence of PMT detection events, as they would occur in a TDCR measurement. The functionality of the code is briefly described in [10] where it was used for the comparison between common and individual dead-time counting logics used in TDCR. The code uses as inputs the activity of the source whose measurement would be simulated, as well as the single and coincidence counting rates. The time to next decay is sampled from exponential distribution taking into account the activity. The time between PMT detection events, produced by a decay, is sampled from a close approximation of a real time interval distribution, in this case the time interval distribution taken from a ^{3}H measurement. The code outputs list-mode data with timestamps and PMT number, similar to a measurement with a digitizer. The generated files can be analyzed with the same software used for the experimental measurements. An important feature of the MC code is that it can output the simulated counting rates in the single and coincidence channels. They can serve as a reference to which the measured and corrected for accidental coincidence counting rates can be compared.

The MC code was used to generate list-mode files for ^{3}H with activities from 2 kBq to 200 kBq. The files were analyzed with the same analysis software used in the previous experiments for the real list-mode data. As the MC code does not generate noise or PMT after-pulses, the chosen dead-time for the analysis was 10 μs. The coincidence resolving time was set to 800 ns, same as for the real ^{3}H measurements. The measured counting rates (Correction method: None) were compared to the MC reference (see Table 3). An increasing discrepancy between the measured and MC reference counting rates can be observed with increase in the activity of the simulated measurement. The discrepancy in the *T* channel is 0.14% for the 2 kBq simulation and increases up to 15.28% for the 200 kBq simulation.

The measured coincidence counting rates were used with Eqs. (8) and (10) in order to estimate the contribution of the accidental coincidences according to the analytical method. The accidental coincidence counting rates were subtracted from the measured counting rates and the true coincidence counting rates were obtained. These were then compared to the MC reference (Correction method: Calc.). For simulated activities up to 60 kBq, the discrepancy between the corrected and MC reference counting rates is less than 0.12%. For 100 kBq and 200 kBq the increase in the discrepancy could be attributed to the large contribution of the accidental coincidences to the measured counting rates, which are used to calculate the correction. Nevertheless, even for activities as high as 100 kBq, the corrected for accidental coincidence counting rates are within 0.29% from the MC reference.

The MC generated list-mode data was analyzed in order to obtain the time distributions in the coincidence channels. For the activities up to 40 kBq the experimental method was used with the assumption for uniform distribution of the accidental coincidences in the interval between 1500 ns and 2000 ns and Eq. (6) was used to calculate the accidental coincidences in each channel. For the higher activities, from 60 kBq to 200 kBq, the assumption is invalid, due to the very high counting rates. In these cases, the accidental coincidence counting rates

Table 3
Comparison of the uncorrected and corrected by the two methods (Calc. and Expt.) counting rates with the MC generated reference counting rates.

Activity	Corr. method	Difference from MC reference, %		
		AB[a]	D	T
2 kBq	None	0.11	0.06	0.14
	Calc.	−0.01	−0.02	−0.02
	Expt.	−0.01	−0.02	−0.02
10 kBq	None	0.59	0.42	0.76
	Calc.	−0.03	0.00	−0.05
	Expt.	−0.01	0.01	−0.04
20 kBq	None	1.23	0.85	1.59
	Calc.	−0.01	0.01	−0.03
	Expt.	−0.02	0.03	−0.06
40 kBq	None	2.43	1.64	3.13
	Calc.	−0.07	−0.01	−0.12
	Expt.	0.01	0.04	−0.02
60 kBq	None	3.69	2.50	4.78
	Calc.	−0.04	0.02	−0.10
	Expt.	−0.03	−0.01	−0.06
100 kBq	None	6.07	4.11	7.90
	Calc.	−0.12	0.08	−0.29
	Expt.	0.10	0.16	0.13
200 kBq	None	11.71	7.91	15.28
	Calc.	−0.61	0.20	−1.31
	Expt.	0.08	0.21	−0.18

[a]The results for the *BC* and *AC* channels were similar and were omitted for brevity.

were obtained by fitting the linear equation (5) to the time interval distribution in the interval from 1500 ns to 2500 ns. The corrected for accidental coincidence counting rates were compared to the MC reference (Correction method: Expt. in Table 3). For activities up to 60 kBq, the corrected using the experimental method coincidence counting rates are within 0.06% of the MC reference. In the case of 100 kBq and 200 kBq the discrepancy is higher, but still within 0.21%.

4. Conclusions

An experimental method to evaluate the accidental coincidence counting rates in TDCR measurements was proposed. The method was used to develop and validate analytical expressions for the counting rate of accidental coincidences.

The analytical and experimental evaluations give consistent results for ^{3}H, ^{55}Fe and ^{14}C measurements. The contribution of the accidental coincidences increases with reduction of the counting efficiency and, for the measured ^{3}H sources, 10 times increase in the activity leads to 50 times increase in T and 20 times increase in D accidental coincidences. The true coincidence counting rates evaluated using the analytical method, which uses the single and coincident counting rates obtained in a typical TDCR measurement, does not seem to depend significantly on the choice of coincidence resolving time for the studied 23 kBq ^{3}H LS-source.

Both methods were used to evaluate the accidental coincidence counting rates for MC generated ^{3}H measurements with activities from 2 kBq to 200 kBq. The coincidence counting rates corrected with the analytical method agree with the MC references to within 0.29% for activities up to 100 kBq. When the experimental method is used it results in less than 0.06% deviation from the reference counting rates for activities up to 60 kBq and less than 0.21% up to 200 kBq.

The analytical evaluation of accidental coincidences can be applied to all existing TDCR acquisition systems that provide single, double coincidence and triple coincidence counting rates. The correction for accidental coincidences gives the opportunity to use long coincidence resolving times that seem to be necessary for the standardization of low-energy radionuclides. Correcting for accidental coincidences could improve the non-linearity of TDCR detection systems when recording the decay of short half-life radionuclides like ^{11}C, ^{15}O or ^{18}F and allow TDCR measurements of high-activity sources, where accidental coincidences cannot be neglected.

CRediT authorship contribution statement

Chavdar Dutsov: Conceptualization, Investigation, Methodology, Software, Writing - original draft. **Philippe Cassette:** Supervision, Methodology, Resources, Funding acquisition. **Benoît Sabot:** Resources, Investigation. **Krasimir Mitev:** Supervision, Methodology, Software, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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