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## Significance of the correction for accidental coincidences in liquid scintillation counting measurements

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## 22        **Introduction**

23    The aim of this paper is to present experimental results that highlight the significance of  
24    the contribution of the accidental coincidences in different types of liquid scintillation  
25    counting measurements. Liquid scintillation counting (LSC) is a technique used for the  
26    measurement of the activity of radionuclides both in the case of low activities (e.g.,  $^{14}\text{C}$   
27    dating, environmental survey, marine studies etc.) and for high activities (measurement of  
28    medical radionuclides, etc.) [1]. LSC is generally performed with instruments equipped  
29    with two or more photomultiplier tubes (PMTs) connected to coincidence counting  
30    circuits. Whenever a coincidence circuit is employed there exists the possibility for two  
31    or more unrelated events to occur within the same coincidence window, thus resulting in  
32    an accidental coincidence. The theoretical basis for the evaluation of the corrections for  
33    accidental coincidences in a LSC system with three PMTs was recently shown in [2]. In  
34    this paper the corrections are expanded also to two-detector and four-detector systems.  
35    The effects of the accidental coincidences on the measurement of half-lives of  
36    radioisotopes, in the case of low-level liquid scintillation counting measurements and in  
37    Compton Triple-to-Double Coincidences Ratio (C-TDCR) measurements are shown.

38    To our knowledge there is are no established procedures for corrections for accidental  
39    coincidences in LSC measurements with commercial devices, two-PMT systems in  
40    general and C-TDCR measurements.

## 41        **Theory**

42    An accidental coincidence occurs when two or more unrelated events are recorded within  
43    the same coincidence resolving time (also called ‘coincidence window’). By event, it is  
44    meant all detected signals that may come from scintillation photons caused by radioactive  
45    decay in the sample, from photons caused by interactions of ionizing radiation coming  
46    outside the sample (background), from sporadic thermal noise of the photocathode of the  
47    PMT, and from electronic noise.

48 The theoretical basis of the evaluation of the corrections for accidental coincidences is  
 49 given in [2]. There, analytical equations were proposed to evaluate the counting rate of  
 50 accidental coincidences in a three PMT liquid scintillation counter. The proposed  
 51 equations can be easily applied also to a two PMT system by fixing the counting rate of  
 52 the third PMT in the equations to zero. Thus, for a two PMT system with PMTs A and B  
 53 the accidental coincidences counting rate  $a_{AB}$  can be evaluated as:

$$\begin{aligned}
 p_A &= n_A - n_{AB} \\
 p_B &= n_B - n_{AB} \\
 a_{AB} &= [2p_A p_B + (p_A + p_B)p_{AB}]\tau,
 \end{aligned}
 \tag{1}$$

55 where  $\tau$  is the coincidence resolving time,  $n_A$ ,  $n_B$  and  $n_{AB}$  are the counting rates in the  
 56 single and coincident channels as reported by the detector. The counting rates  $p_A$  and  
 57  $p_B$  are the ‘pure’ single counting rates, meaning the counting rates for events that are  
 58 single events and are not part of a coincidence.

59 Examining equation (1) there are two terms that constitute the accidental coincidences in  
 60 a two-PMT system: coincidence between unrelated events in the two PMTs or a single  
 61 event in any PMT followed by a true coincidence within the same coincidence window.  
 62 Both terms in the equation depend on the single event counting rates  $p_A$  and  $p_B$ . There are  
 63 two sources of single events that can be distinguished. One are single events that result  
 64 from low-energy interactions in the scintillation cocktail and the other is single events  
 65 that come from the PMT dark noise. For measurements where the activity of the source is  
 66 low, the predominant source of single events would be the dark noise of the PMT.  
 67 Therefore, in low-activity measurements, the contribution of the accidental coincidences  
 68 will be determined from the characteristics of the PMTs. This will also be the case for  
 69 measurements of blank samples in low-background environments.

70 After the calculation of the counting rate of the accidental coincidences  $a_{AB}$ , the  
 71 corrected counting rate  $m_{AB}$  is calculated as  $m_{AB} = n_{AB} - a_{AB}$ .

## 72 **Experimental**

73 Accidental coincidences in the measurement of half-lives of  
74 radionuclides

75 The half-life of  $^{18}\text{F}$  was studied in this experiment by following the evolution of the  
76 activity with time. This measurement was performed with a miniature portable TDCR  
77 counter. A sample was prepared with 1 mL of radioactive solution ready for injection to a  
78 patient (FDG) and dissolved in 10 mL of Ultima Gold LS-cocktail in a glass vial. During  
79 this measurement, the counting rates evolves from about  $430\,000\text{ s}^{-1}$  to about  $160\text{ s}^{-1}$ . The  
80 main objective of this study is not the precise determination of the half-life which could  
81 be used in decay data evaluation, but to illustrate the effect that accidental coincidences  
82 could have.

83 Accidental coincidences in low-level TDCR measurements

84 Low-level LSC measurements are commonly used for environmental survey purpose and  
85 for mandatory monitoring around nuclear activities. These low-level measurements are  
86 performed with LSC systems that need to be calibrated with standards of known  
87 activities. The activity of the standard sample can be determined using the TDCR method  
88 but generally, the activity of the standard is higher than the targeted measurement. Thus,  
89 the non-linearity of the detectors may have an impact on the results and, in some case,  
90 close to environmental level standards are required for detector calibration. Another case  
91 where low-level TDCR measurements can be necessary is for the standardization of some  
92 rare nuclides for which it is difficult to have sufficient amount of activity. For low-level  
93 measurements, the background is significant compared to the signal from the sample and  
94 the contribution of the accidental coincidences to the background counting rates can  
95 propagate to the final results through the background correction.

96 In this experiment we have performed measurements of the pure electron capture (EC)  
97 nuclide,  $^{53}\text{Mn}$  and of the alpha nuclide,  $^{209}\text{Po}$  (99.55% alpha emissions and 0.45% EC).  
98 The measurements of the  $^{53}\text{Mn}$  sample were performed on a miniature portable micro-  
99 TDCR counter inside a low background shielding (10 cm of low-level lead, 2 mm of  
100 cadmium, 2 mm of tin). The counting rate in the logical sum of double coincidences (D)

101 of the blank sample was about  $2.15 \text{ s}^{-1}$  compared to the usually observed  $10 \text{ s}^{-1}$  without  
102 shielding. The  $^{209}\text{Po}$  solution is dissolved in 10 ml of Ultima Gold AB cocktail in a  
103 polyethylene PTFE coated vial. This sample was measured on the same micro-TDCR  
104 counter, but without the low-level shielding.

## 105 Correction for accidental coincidences in C-TDCR measurements

106 In LSC, the primary method that is used for standardization is the TDCR method [3]. The  
107 method relies on the correct description of the non-linearity of the light output of the  
108 scintillator. Instead of using a traditional semi-empirical relation (the Birks equation [4]),  
109 this non-linearity can be directly measured by coupling the TDCR detector to a Compton  
110 spectrometer (C-TDCR). A virtual source is created internally in the LS cocktail from an  
111 external monoenergetic photon source directed at the vial containing the sample [5, 6].  
112 Some of the photons would experience Compton scattering and some of those scattered  
113 photons would be detected by the Compton spectrometer. By gating on a certain energy  
114 of the Compton scattered photons one can select a certain energy of the Compton  
115 electrons in the cocktail. Then, the light output of the cocktail can be calculated from the  
116 LS detector using the TDCR method and the energy of the electron causing the  
117 scintillation burst can be deduced from the Compton spectrometer and the energy balance  
118 of the Compton effect.

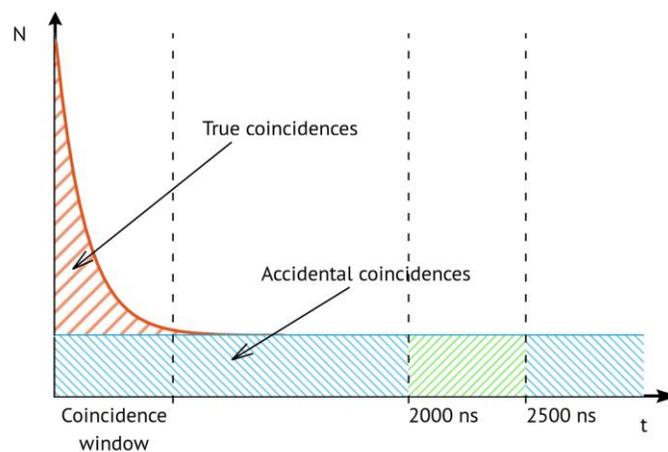
119 One drawback of the C-TDCR methodology is the relatively low detection efficiency of  
120 the gamma channel compared to the TDCR channel. This is caused mainly by the  
121 difficulty in obtaining high geometrical efficiency of the gamma channel as it requires the  
122 use of multiple detectors to cover significant portion of the  $4\pi$  space. In general, the result  
123 of this drawback is that a high counting rate is needed in the PMT channels in order to  
124 obtain a reasonable counting rate in the gamma channel. The low detection efficiency in  
125 combination with high single counting rates can lead to significant contribution of  
126 accidental coincidences.

127 The theoretical calculation of the accidental coincidences in a three PMTs counter has  
128 been derived in [1]. In principle, the same derivation methodology should be applicable

129 to a four detectors system as the C-TDCR. In the four detectors system however, there  
 130 are four single channels, six double coincidence channels, four triple coincidence  
 131 channels and one quadruple coincidence channel. An analytical equation that accounts for  
 132 all possible sources of accidental coincidences would be unwieldy and difficult to use in  
 133 practice. Here we consider another simpler approach that is based on the experimental  
 134 evaluation of the accidental coincidences again proposed in [1]. The technique relies on  
 135 the possibility to perform measurements with large coincidence windows ( $> 2000$  ns).

136 The method consists of performing simultaneous measurements using three coincidence  
 137 window widths: 1. the coincidence window of interest (e.g., 40 ns or 200 ns), 2000 ns and  
 138 2500 ns. Under the assumption of a uniform time distribution of the accidental  
 139 coincidences in the time interval of interest it is possible to estimate them from the  
 140 difference in the counting rates in the 2500 ns and 2000 ns coincidence windows.  
 141 Another assumption that is made is that at 2000 ns coincidence window width all true  
 142 coincidences are registered, thus all additional coincidences in a given coincidence  
 143 channel (ABC, ABG, BCG, etc.) in the 2500 ns are accidental coincidences. The method  
 144 is schematically illustrated in Fig. 1.

145



146 **Figure 1** Schematic illustration of the method for correction of accidental coincidences in  
 147 a four detectors system. The portion of the time interval distribution between 2000 ns and  
 148 2500 ns is used to estimate the plateau of the accidental coincidences.

149 The assumption for the uniform distribution of the accidental coincidences should be  
150 applicable to common measurement scenarios. If it can be assumed that the uncorrelated  
151 events that constitute the accidental coincidences are a Poisson process, then the time  
152 interval between them is exponentially distributed [1]:

$$a(t) = a_0 e^{-\lambda t},$$

153 where  $a(t)$  is the distribution of accidental coincidences,  $a_0$  is the rate of the accidental  
154 coincidences at time  $t = 0$  and  $\lambda$  is the rate of detected events in the coincidence channel.  
155 In the case of C-TDCR measurements the coincidence counting rates are less than  $10^3 \text{ s}^{-1}$   
156 and the coincidence window is less than 2500 ns. Thus, the argument of the exponent will  
157 be less than  $2.5 \times 10^{-3}$ . If the exponential distribution is approximated with the linear  
158 terms in the Taylor series, the result is:

$$a(t) = a_0(1 - \lambda t).$$

159 Therefore, the deviation from the uniform distribution  $a(t) = a_0$  is negligible and this  
160 assumption should be valid.

161 The second assumption of the method is that there are no true coincidences that can be  
162 observed for which the separation between the single events is larger than 2000 ns. That  
163 is, the scintillation photons that result from one ionizing particle in the sample are  
164 grouped in time such that no photon can be emitted after more than 2000 ns after the  
165 excitation from the ionizing particle. As organic scintillators and PMTs are fast detector  
166 systems this is generally true. It has been shown that 1000 – 1500 ns are enough to  
167 capture even photons coming from the delayed fluorescence of the scintillator [7, 8].

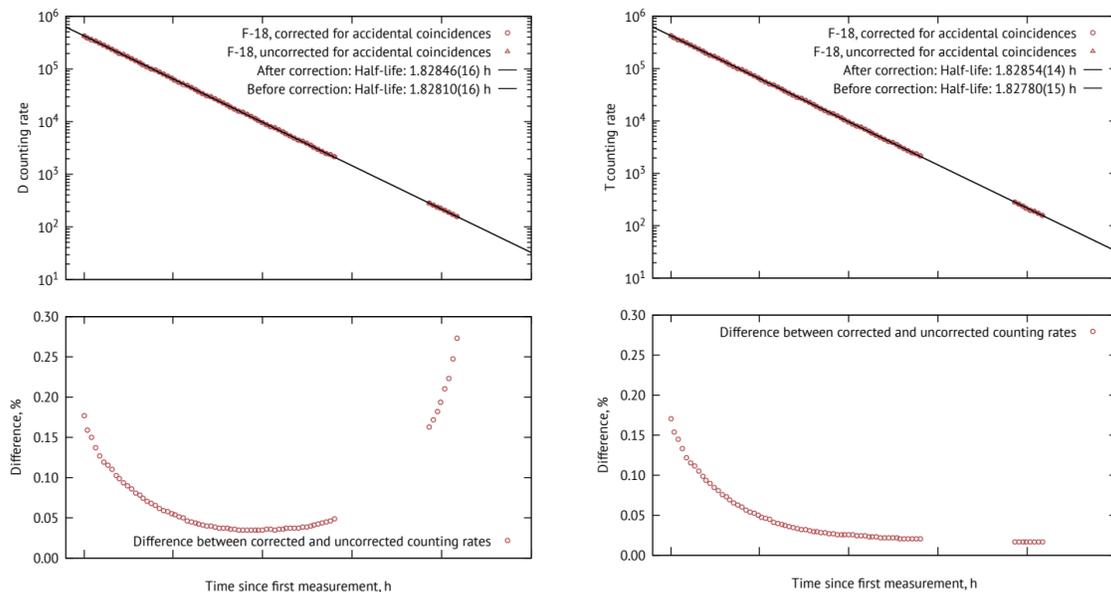
## 168 **Results and discussion**

### 169 **Accidental coincidences and studies of half-lives**

170 In total 72 consecutive 100 s long measurements were performed on the  $^{18}\text{F}$  sample on  
171 the miniature TDCR detector. The detection efficiency for this nuclide is relatively high

172 and is around 99.7%, therefore a large contribution of the accidental coincidences is not  
 173 expected. A blank sample containing no radioactivity was also measured 64 times for  
 174 1000 s. The coincidence window for both sample and blank measurements is 40 ns and  
 175 the base dead-time duration is 10  $\mu$ s. For the studies of the decay curves with corrected  
 176 counting rates, both the blank and source measurements were corrected for accidental  
 177 coincidences.

178 The use of a three-PMT counter allows the study of the decay curve of the logical sum of  
 179 the double coincidences (D) and also on the triple coincidences (T). The results of the  
 180 study are shown in Figure 2. The left figure presents the decay curve following the D  
 181 coincidences and the right figure the decay curve following the T coincidences. The  
 182 difference between the corrected and uncorrected counting rates are shown on the bottom  
 183 plots in percent.



184 **Figure 2** Study of the half-life of  $^{18}\text{F}$ . The decay curve in the left figure is taken from the  
 185 D counting rate in the TDCR counter and from the T counting rate in the right figure. The  
 186 bottom subplots show the difference between the corrected and uncorrected counting  
 187 rates. The values in brackets are the fit uncertainties.

188 There are some interesting observations that can be made from the presented figures.  
 189 Firstly, the accidental coincidences in the D and T channels behave differently. For large

190 counting rates they are both dominated by accidental coincidences between single and  
191 double events caused by  $^{18}\text{F}$  decays and their contribution to the respective channels are  
192 roughly the same. At low counting rates, however, the accidental coincidences are  
193 dominated by single events coming from thermal noise of the PMTs. The probability for  
194 triple accidental coincidences in a scenario with mostly single events is negligible and the  
195 contribution of the accidentals in that case is close to zero. For the D channel the  
196 contribution of the accidental coincidences increases significantly and is even larger than  
197 in the case of  $430\,000\text{ s}^{-1}$  counting rate. A second observation is that, for both the T and  
198 D decay curves, the difference between the obtained half-life for corrected and  
199 uncorrected counting rates is larger than the fit uncertainties. This shows that even for a  
200 very high detection efficiency sample such as  $^{18}\text{F}$  it is necessary to perform corrections  
201 for accidental coincidences when studying the half-life.

202 A more comprehensive evaluation of the half-life of  $^{18}\text{F}$  is outside the scope of this paper  
203 and will be given elsewhere.

204 This study serves to illustrate that in some cases the accidental coincidences can be more  
205 significant at low counting rates than at extremely high counting rates.

## 206 Accidental coincidences in low-level TDCR measurements

207 In this experiment we have performed TDCR measurements of samples containing  $^{53}\text{Mn}$   
208 and  $^{209}\text{Po}$ . The results from the experiment are presented in Table 1. The coincidence  
209 counting rates in the measurements of the radioactive samples are those after subtraction  
210 of the counting rates of the blank sample. The  $^{53}\text{Mn}$  source was measured with 200 ns  
211 coincidence window width. The  $^{209}\text{Po}$  source was measured simultaneously with 40 ns  
212 and 400 ns coincidence windows using the nanoTDCR device [9].

Source	Coinc. window	Counting rate			
			AB	D	ABC
Mn-53 (EC)	200	No correction	40.590(20)	82.514(27)	18.900(15)
		After correction	40.553(20)	82.423(27)	18.889(15)
		Difference	<b>-0.09%</b>	<b>-0.11%</b>	<b>-0.06%</b>
Blank	200	No correction	1.646(4)	2.158(5)	1.415(4)
		After correction	1.640(4)	2.136(5)	1.415(4)
		Difference	<b>-0.38%</b>	<b>-1.01%</b>	<b>0.00%</b>
Po-209 (alpha)	40	No correction	9.388(25)	9.785(27)	9.216(25)
		After correction	9.369(25)	9.726(26)	9.217(25)
		Difference	<b>-0.20%</b>	<b>-0.60%</b>	<b>0.01%</b>
Blank	40	No correction	9.506(8)	10.466(9)	8.958(8)
		After correction	9.420(8)	10.305(9)	8.958(8)
		Difference	<b>-0.90%</b>	<b>-1.53%</b>	<b>0.00%</b>
Po-209 (alpha)	400	No correction	9.160(40)	9.206(62)	9.163(24)
		After correction	9.350(25)	9.797(27)	9.157(24)
		Difference	<b>2.08%</b>	<b>6.42%</b>	<b>-0.06%</b>
Blank	400	No correction	10.463(13)	12.075(23)	9.152(8)
		After correction	9.606(7)	10.474(9)	9.152(8)
		Difference	<b>-8.20%</b>	<b>-13.26%</b>	<b>0.00%</b>

213

214 **Table 1** Low-level measurements of  $^{53}\text{Mn}$  and  $^{209}\text{Po}$  before and after corrections for  
215 accidental coincidences. The BC and AC counting rates are omitted as the results are  
216 similar to the AB counting rate. The values in brackets are the statistical standard  
217 uncertainties calculated as the standard deviation of the mean from ten consecutive  
218 measurements of the same sample in the same measurement conditions.

219 A few observations can be made from the results in Table 1. In all measured blank  
220 samples, the contribution of the accidental coincidences is significant in the double and  
221 logical sum of double coincidences counting rates. In all cases the accidental  
222 coincidences in the ABC channel are negligible. This is most probably caused by the low  
223 double coincidences counting rates leading to low probabilities of accidental  
224 coincidences. Note here that the accidental coincidences in the ABC channel depend on  
225 the double coincidences counting rates because of possible detection of unrelated events  
226 in the same window such as: for example, unrelated AB and C events leading to a false  
227 ABC but also unrelated AB and BC events leading to a false ABC and other such  
228 combinations.

229 Thus the higher the double counting rates, the larger the probability for ABC accidental  
230 coincidences.

231 It should also be noted that for  $^{209}\text{Po}$  measurements the counting rates in the double  
232 coincidences channels decrease after subtracting the accidental coincidences in both the  
233 blank sample and the radioactive sample. This is due to the high relative contribution of  
234 the accidental coincidences in the blank sample that leads to overestimated blank  
235 counting rates before the correction. Comparing the net (background corrected) logical  
236 sum of double coincidences (D) counting rates of the two  $^{209}\text{Po}$  measurements it can be  
237 seen that there is a 5.9% difference between the uncorrected results at 40 ns (9.785 cps)  
238 and 400 ns (9.206 cps) coincidence window width. After applying corrections for  
239 accidental coincidences the D counting rates become  $9.797\text{ s}^{-1}$  at 400 ns coincidence  
240 window and  $9.726\text{ s}^{-1}$  at 40 ns coincidence window. Thus the difference between the two  
241 measurements is reduced to 0.73% after the accidental coincidences correction. Note that  
242 this difference could be due to the general increase in the coincidence counting rates that  
243 is observed in LSC measurements with increase in the coincidence window width in the  
244 case of low-energy emissions [8]. Low-energy emissions that can be detected due to the  
245 decay of  $^{209}\text{Po}$  are the X radiations after the EC (probability for EC 0.454%) and the  
246 emissions from the 2.239 keV level of  $^{205}\text{Pb}$  that is populated after the alpha decay of  
247  $^{209}\text{Po}$  [10]. The latter can be observed independently of the alpha decay due to the 24.2  $\mu\text{s}$   
248 half-life of the excited state.

249 These experiments highlight the importance of applying the corrections for accidental  
250 coincidences not only for TDCR measurements of high-activity sources, but also in low-  
251 level measurements.

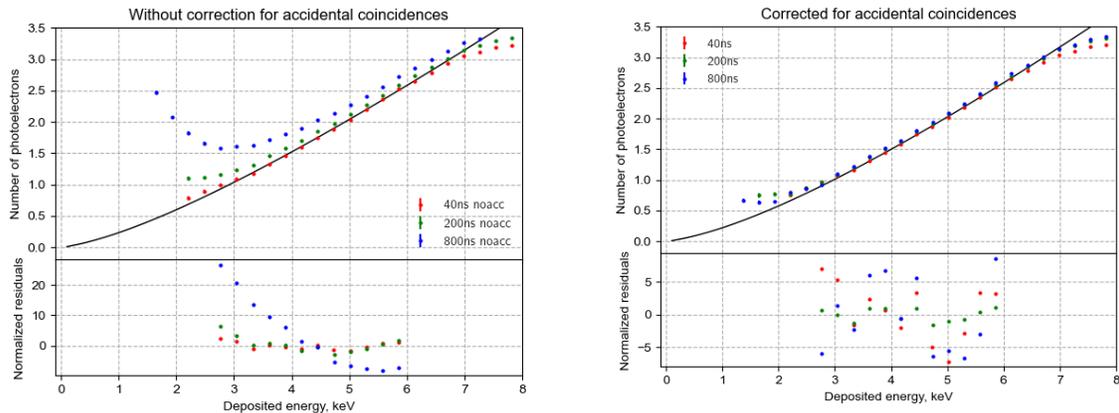
## 252 Correction for accidental coincidences in C-TDCR measurements

253 In order to illustrate the importance of the correction for accidental coincidences in  
254 C-TDCR measurements we have performed measurements of the relative light output of  
255 an Ultima Gold scintillation cocktail. The measurements were performed with a  
256 miniature portable TDCR detector system coupled to a CdTe X-Ray detector. The

257 external source is 74 MBq  $^{241}\text{Am}$  in a steel casing, which provides filtering of the low-  
258 energy emissions. Thus, practically all emissions from the source are from the 59.54 keV  
259 line of excited level of  $^{237}\text{Np}$ . The three PMTs of the TDCR counter and the CdTe are  
260 connected directly to a CAEN 5751 Desktop Digitizer which digitizes all incoming  
261 signals and writes them into list-mode files. These files are later analyzed by a computer  
262 software that applies the common dead-time logic [11, 12]. Thus, with this setup it is  
263 possible to analyze the same measurement with an arbitrary coincidence window width.

264 Using this experimental setup, a single 72-hour measurement of the Ultima Gold sample  
265 was performed. The list-mode data from the measurement was analysed with 40 ns,  
266 200 ns, 800 ns, 2000 ns and 2500 ns coincidence window width. The latter two are used  
267 to estimate the accidental coincidences. The accidental coincidences per nanosecond of  
268 resolving time  $A_0^{(X)}$  in a given coincidence channel  $X$  are calculated as the difference  
269 between coincidence counting rates  $n$  in the channel at 2500 ns and 2000 ns coincidence  
270 window. The accidental coincidences counting rate  $a^{(x)}$  for the coincidence window of  
271 interest  $\tau$  are then obtained as  $a^{(x)} = A_0^{(x)}\tau$ , where  $\tau = 40, 200$  or  $800$ .

272 The results of the measurement without and with corrections for accidental coincidences  
273 are shown in Fig. 3. Several observations can be made from the presented figures. First,  
274 besides the dependence on the coincidence window width, the contribution of the  
275 accidental coincidences also largely depends on the deposited energy in the cocktail. For  
276 lower energies, the contribution increases and the increase is non-linear. This is caused  
277 by the significant reduction in detection efficiency with lower deposited energies which  
278 leads to an increase in the ratio of the single events to the detected coincidences. Second,  
279 the three measurements agree well with each other when the corrections for accidental  
280 coincidences are applied. Some small difference between the long coincidence windows  
281 200 ns and 800 ns and the short 40 ns coincidence window remains even after the  
282 corrections. This effect is expected as for the 40 ns coincidence window a loss of  
283 coincidences is already observed [7, 12, 13].



284 **Figure 3** Average number of photoelectrons produced at the photocathodes of the PMTs  
 285 as a function of the energy deposited in the scintillation cocktail. The black line shows a  
 286 fit of the data from the 40 ns coincidence window with Birks' formula for the non-  
 287 linearity of organic scintillators.

288 Overall, the accidental coincidences are quite significant for the C-TDCR measurements,  
 289 and especially so for low detection efficiencies. For example, the correction is 3.4% at  
 290 3 keV deposited energy at 200 ns coincidence window. It is 0.4% at 40 ns coincidence  
 291 window and 12.4% at 800 ns coincidence window. For higher energies (higher detection  
 292 efficiency) the contribution drops to 0.2% at 7 keV (40 ns) and 0.8% (200 ns).

### 293 Discussion about measurements with commercial liquid scintillation 294 counters

295 The corrections for the accidental coincidences that were proposed in [2] can be used on  
 296 all detector systems that provide information about the coincidence window width as well  
 297 as the single and coincident counting rates. Unfortunately, this is not true for the most  
 298 commonly used commercial liquid scintillation systems, e.g., Quantulus<sup>TM</sup> by  
 299 PerkinElmer<sup>TM</sup>. These systems are commonly used in scenarios with low counting rates  
 300 and low detection efficiencies. Both are a prerequisite for having a significant  
 301 contribution of the accidental coincidences.

302 Despite the impossibility to check the exact value of the contribution of the accidental  
 303 coincidences in commercial counters it is nevertheless of interest to give estimates in

304 some possible real-world cases. Here we have calculated the contribution of the accidental  
 305 coincidences for a hypothetical system with 50 ns coincidence window (CW) width that  
 306 measures samples with coincidence counting rates between  $0.01 \text{ s}^{-1}$  and  $1000 \text{ s}^{-1}$ . It is  
 307 assumed that the pure single counting rate,  $S_p$ , coming from the sample is 50% from the  
 308 measured double coincidence counting rate  $D_m$  – such a case is typical for  $^3\text{H}$   
 309 measurements. Therefore, the measured single counting rate  $S_m$  is:  $S_m = S_0 + S_p = S_0 +$   
 310  $0.5D_m$ , where  $S_0$  is the intrinsic dark noise of the PMT. Three cases are considered:  
 311 commonly observed dark noise of  $300 \text{ s}^{-1}$ , reduced dark noise of  $100 \text{ s}^{-1}$  and greatly  
 312 reduced dark noise of  $10 \text{ s}^{-1}$ . The latter could possibly be achieved with high signal  
 313 discrimination thresholds or cooling of the PMTs. Note that the measured single counting  
 314 rates of the two PMTs are assumed to be the same, i. e., the PMTs are assumed to be  
 315 identical. The double coincidences counting rate after the correction for accidental  
 316 coincidences  $D_c$  is calculated as:  $D_c = D_m - a$ , where  $a$  is calculated according to  
 317 equation (1) substituting  $n_A = n_B = S_0$  and  $n_{AB} = D_m$ .

CW, ns	Intrinsic dark noise $S_0, \text{ s}^{-1}$	Measured singles rate $S_m, \text{ s}^{-1}$	Measured doubles rate $D_m, \text{ s}^{-1}$	Doubles rate corrected for accidentals $D_c, \text{ s}^{-1}$	$(D_m - D_c)/D_m$
50	300	300.03	0.02	0.011	45.01%
50	300	300.15	0.1	0.091	9.01%
50	300	301.5	1	0.991	0.91%
50	300	315	10	9.990	0.10%
50	300	450	100	99.984	0.02%
50	300	1800	1000	999.856	0.01%
50	100	100.015	0.01	0.009	10.00%
50	100	100.15	0.1	0.099	1.00%
50	100	101.5	1	0.999	0.10%
50	100	115	10	9.999	0.01%
50	100	250	100	99.996	0.00%
50	100	1600	1000	999.904	0.01%
50	10	10.015	0.01	0.010	0.10%
50	10	10.15	0.1	0.100	0.01%
50	10	11.5	1	1.000	0.00%
50	10	25	10	10.000	0.00%
50	10	160	100	99.999	0.00%
50	10	1510	1000	999.923	0.01%

318 **Table 2** Calculations of the contribution of accidental coincidences in a hypothetical two  
 319 PMT LSC system. Note that the counting rates are given in reciprocal seconds and

320 commercial counters usually report counting rates in reciprocal minutes (counts per  
321 minute or ‘cpm’).

322 The results of this hypothetical experiment are presented in Table 2. Note that for  
323 extremely low counting rates ( $< 0.1 \text{ s}^{-1}$ ) the contribution of the accidental coincidences is  
324 significant in the discussed cases. When the PMT dark noise is reduced to  $10 \text{ s}^{-1}$  the  
325 contribution of the accidental coincidences is negligible for counting rates between  $0.1 \text{ s}^{-1}$   
326 and  $1000 \text{ s}^{-1}$ . For more commonly observed cases of the dark noise of the PMTs the  
327 contribution of the accidental coincidences can be seen as significant up to  $1 \text{ s}^{-1}$  or  $10 \text{ s}^{-1}$   
328 (60 cpm and 600 cpm respectively).

329 The information about the counting rates in the individual channels and the width of the  
330 coincidence window is important and should be provided to the user even in the case of  
331 routine LSC measurements. For example, one commercially available device that  
332 provides this data is the nanoTDCR analyzer.

### 333 Low-level measurements with a custom two-PMT liquid scintillation 334 counting system

335 The effect of the accidental coincidences on low-level measurements can be illustrated  
336 when using a custom-made two-PMT system with known measurement parameters. This  
337 system was used to perform measurements of two samples of  $^{209}\text{Po}$  with approximately  
338 1 Bq and 60 Bq activity. The two-PMT system consists of two XP2020Q  
339 photomultiplier tubes with quartz windows positioned inside an optical chamber at  $180^\circ$   
340 to each other. The shape of the optical chamber was designed and made at LNE-LNHB  
341 and is covered with reflective foil (ESR by 3M<sup>TM</sup>). The two PMTs are supplied with  
342 +2100 V high voltage. Their output signals are processed using labZY nanoTDCR. The  
343 nanoTDCR device is used here, instead of a commercial counter, because its signal  
344 processing logic and parameters are known and user-selectable, which is not usually the  
345 case for commercial counters. The device also allows user-selectable thresholds for both  
346 signal inputs. The thresholds for both PMTs are set so that the electronic noise is  
347 suppressed as much as possible without sacrificing single-photon sensitivity. The

348 nanoTDCR also allows simultaneous measurements with two different coincidence  
 349 windows and two different dead-time base durations. In this experiment the two  
 350 coincidences windows were set to 40 ns and 200 ns and the dead-time base duration to  
 351 40  $\mu$ s. The nanoTDCR outputs the single counting rate of each PMT and double  
 352 coincidences counting rate between PMTs with a known coincidence window and known  
 353 dead-time base duration [9].

354 In order to reduce the background and perform low-activity measurements the two-PMT  
 355 system was placed inside a shielded chamber. The shielding consists of 10 cm of low-  
 356 level lead, 2 mm of cadmium and 2 mm of tin. The chamber was flushed with nitrogen  
 357 before the measurements to remove radon gas present in the chamber with an average  
 358 activity concentration of 25 Bq·m<sup>-3</sup> in the laboratory.

359 The system was used to perform measurements on two samples of <sup>209</sup>Po. The results are  
 360 shown in Table 3. The value  $n_{AB}$  refers to the double coincidences counting rate before  
 361 any corrections are done. The corrected counting rate  $m_{AB}$  is the net counting rate after  
 362 subtracting the accidental coincidences in the case of the blank samples. In the case of the  
 363 polonium samples it is calculated as:  $m_{AB} = \left( n_{AB}^{(Po)} - a_{AB}^{(Po)} \right) - \left( n_{AB}^{(blank)} - \right.$   
 364  $\left. a_{AB}^{(blank)} \right)$ . The values in brackets are the standard uncertainties calculated as the standard  
 365 deviation of the mean taken from 10 consecutive measurements of the same sample in the  
 366 same conditions.

Sample	CW, ns	PMT A single counting rate, s <sup>-1</sup>	PMT B single counting rate, s <sup>-1</sup>	Before correction $n_{AB}$ , s <sup>-1</sup>	Rate of accidental coinc. $a_{AB}$ , s <sup>-1</sup>	Corrected counting rate $m_{AB}$ , s <sup>-1</sup>	$(m_{ab} - n_{ab})/n_{ab}$
<sup>209</sup> Po (~1 Bq)	40	187.5(17)	645.0(91)	1.0635(47)	0.0101	1.0534(46)	-0.95%
<sup>209</sup> Po (~1 Bq)	200	187.5(17)	645.0(91)	1.1064(53)	0.0508	1.0556(47)	-4.59%
<sup>209</sup> Po (~60 Bq)	40	304.7(96)	960(43)	63.680(98)	0.06	63.686(98)	0.01%
<sup>209</sup> Po (~60 Bq)	200	304.7(96)	960(43)	63.834(98)	0.028	63.862(98)	0.04%
Blank	40	200.9(94)	660.4(111)	0.5336(16)	0.0131	0.5205(16)	-2.46%
Blank	200	200.9(94)	660.4(111)	0.6201(21)	0.0657	0.5544(15)	-10.60%

367 **Table 3** Calculations of the contribution of accidental coincidences in the two PMT LSC  
 368 system for the two samples of <sup>209</sup>Po and the blank sample used for background  
 369 correction. Note that the <sup>209</sup>Po counting rates are background corrected and ‘before

370 correction' and 'corrected' refers to the correction for accidental coincidences. The same  
371 blank measurement was used for background correction for both  $^{209}\text{Po}$  samples.

372 The measurements in Table 3 show that both PMTs are not identical and PMT B has  
373 higher single counting rate probably due to higher intrinsic thermal noise. In the case of  
374 the measurement of the blank sample and the 1 Bq  $^{209}\text{Po}$  sample the major contributing  
375 factor to the accidental coincidences would be the thermal noise of the PMTs. While the  
376 counting rate of the accidental coincidences caused by the thermal noise is very low  
377 ( $0.0101\text{ s}^{-1}$  at 40 ns coincidence window and  $0.0508\text{ s}^{-1}$  at 200 ns) it is significant  
378 compared to the low true counting rates of the source and blank samples. The  
379 coincidence counting rate of the 1 Bq  $^{209}\text{Po}$  sample is 0.95% lower after correction for  
380 accidental coincidences for the 40 ns coincidence window measurement and 2.46% lower  
381 for the 200 ns coincidence window measurement. It should also be noted that the  
382 corrected coincidence counting rates obtained with the longer window are significantly  
383 higher than those with the short window. This shows that some coincidences are lost if  
384 the short 40 ns coincidence window is used. Therefore, the effect of the accidental  
385 coincidences cannot be reduced by simply using a shorter coincidence window as this  
386 would lead to loss of true coincidences also. In the case of the higher activity  $^{209}\text{Po}$   
387 source, the contribution of the accidental coincidences to the background corrected  
388 coincidence counting rate is negligible.

389 These experiments serve to show the importance of applying corrections for accidental  
390 coincidences in the case of low-activity measurements. To do that, the use of a signal  
391 processing unit that gives information about the coincidence window and the single  
392 counting rates is mandatory.

### 393 **Conclusions**

394 These experiments highlight the importance of applying the corrections for accidental  
395 coincidences not only for measurements of high-activity sources, but also in low-level  
396 LSC measurements. The performed C-TDCR measurements show that the accidental  
397 coincidences must be considered in all cases and applying corrections is mandatory if

398 correct results are to be obtained. The hypothetical measurements with commercial LSC  
399 systems indicate that the accidental coincidences can be significant in some cases.  
400 Unfortunately, most of the commercial LSC counters are black boxes and do not allow  
401 the estimation of the accidental coincidences due to the lack of important data such as the  
402 coincidence window width that is used or the single PMT counting rates. LSC systems  
403 that provide information of the single counting rates and used coincidence window width  
404 and dead-time logic should be preferred in cases where accurate measurements are  
405 needed.

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