Optimization of liquid scintillation measurements applied to smears and aqueous samples collected in industrial environments

Arnaud Chapon [a,b,*], Gilbert Pigrée [c], Valérie Putmans [d], Gwendal Rogel [b]

[a] LPC Caen, ENSICAEN, Université de Caen, CNRS/IN2P3, Caen, France
[b] CERAP – Conseil et Etude en Radioprotection, Cherbourg-Octeville, France
[c] IMOGERE, Université de Caen, Caen, France
[d] PerkinElmer, Zaventem, Belgium

ABSTRACT

Search for low-energy β contaminations in industrial environments requires using Liquid Scintillation Counting. This indirect measurement method supposes a fine control from sampling to measurement itself. Thus, in this paper, we focus on the definition of a measurement method, as generic as possible, for both smears and aqueous samples' characterization. That includes choice of consumables, sampling methods, optimization of counting parameters and definition of energy windows, using the maximization of a Figure of Merit. Detection limits are then calculated considering these optimized parameters. For this purpose, we used PerkinElmer *Tri-Carb* counters. Nevertheless, except those relative to some parameters specific to PerkinElmer, most of the results presented here can be extended to other counters.

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Introduction

Liquid Scintillation Counting (LSC) is the detection of an ionizing radiation through the scintillation light produced in certain solutions. The number of photons emitted during the process is dependent on the type of primary ionizing particle and its energy. Liquid scintillation has the advantage over solid scintillators of mixing the sample within the sensitive medium. Therefore, this technique enables the detection of low-energy radiations (e.g. the beta emission from tritium) by avoiding self-absorption in the sample or interface issues. On the other hand, factors like the amount of scintillator solute present in the solution, the presence of impurities in the sample (quenching) impact the detection efficiency of the process [1]. The *Tri-Carb* family of computer-controlled bench top liquid scintillation analyzers presents one of the most sensitive detectors on the market for measuring small amounts of beta, gamma and alpha radioactivity. For the purpose of this study, we performed most of the measurements using a *Tri-Carb 2100TR PerkinElmer* counter, while some have been obtained using a *Tri-Carb 2900TR PerkinElmer*.

Measurement requirements

As mentioned in [2], the recommended operating ambient temperature for *Tri-Carb PerkinElmer* counters should go from 15 °C to 35 °C and operating relative humidity should be between 30% and 85% without condensation. These must be kept away from any source of radiation and protected from direct sunlight and any source of fluorescent light.

Sources of background noise

Background generates events that can be mistakenly attributed to radioactivity emanating from the measured sample. Among them:

- cosmic radiation,
- ambient background radiation, particularly the one present in the laboratory, in the shielding around photomultipliers (PMTs), in glass of PMTs (or the glass of the vial if any) or the composition of the cocktail,
- the phenomenon of chemiluminescence, resulting in the emission of photons produced during chemical processes in the cocktail,
- the phenomenon of photoluminescence, due to excitation of the cocktail with UV light,

* Corresponding author at: LPC Caen, ENSICAEN, Université de Caen, CNRS/IN2P3, Caen, France.

- electrostatic discharge, spontaneously emitted from the surface of the vials (if made in plastic), and resulting in showers of bright low-energy photons,
- PMTs' electronic noise, or thermal noise,
- direct lighting transfer between the PMTs.

These noise sources are usually divided into two categories [3]: one, said *quenchable*, due to the presence of interference in the scintillation cocktail (internal or external interactions radioactivity) and the other, said *unquenchable*, from outside of the sample. The two contributions may be estimated separately by measuring vials containing different amounts of scintillation liquid. The *quenchable* background noise is then proportional to the volume of scintillation liquid, unlike the *unquenchable* contribution. The two noise sources having different signal shapes and algorithms based on their fine analysis allow to limit the *unquenchable* background noise. Fig. 1 shows the count rate against the volume of scintillation liquid in the vial.

The *unquenchable* background noise is thus responsible for about 7 counts per minute (CPM) in both the $[0:12]$ keV window (corresponding to the ^{3}H β energy) and the $[12:156]$ keV window (corresponding to the ^{14}C β energy). Then, background noise is dominant at low energy as shown in Fig. 2, where $[0:12]$ keV and $[12:156]$ keV windows are respectively colored in yellow and green. Its origins are various and can be limited as described below.

Reduction of the background noise level

A lead shielding is placed around the PMTs, inside the counter, to limit the background due to cosmic radiation and ambient radioactivity in the laboratory. However, it remains preferable to limit the presence of radioactive sources in the laboratory, close to the counter.

Moreover, the phenomena of photoluminescence and chemiluminescence are characteristic of an excitation of the cocktail, either during preparation or during storage, and decrease according to a double-exponential law. Typically, maintaining samples a few minutes to an hour in the dark, distant from any source of direct white light (Sun or neon), can limit the phenomena of photoluminescence, also essentially present at low energy (<5 keV). Fig. 3 illustrates the result of such an excitation; two vials, each containing 10 mL of UltimaGold scintillation cocktail,[1] were stirred for 10 minutes, one to sunlight and the other in the light of a neon, before being measured. The photoluminescence decay appears. The one induced by the sunlight decreases to 10% of its highest level after two hours. A faster decrease is observed when the photoluminescence is induced by the light of a neon, which is less by a factor of about 1000. Whatever the nature of the excitation, the scintillation cocktail is returned to a stable state after maintaining for three days in the dark. None of the two vials presents any excessive counting rate.

The chemiluminescence problem, in contrast, is specific to the composition of the cocktail and is not affected by light exposure at which the sample is subjected. Only temperature and aging affect this phenomenon. Nevertheless, if an increase in temperature creates chemiluminescence, its decrease is thereby accelerated. Scintillation cocktail must therefore be maintained at a constant temperature (room temperature).

Static electricity is applicable only to polyethylene (PE) vials. Nevertheless, it can be avoided by handling the vials with anti-static gloves[2] and in an atmosphere with sufficient relative humidity (>60%). If these precautions are essential, the *Tri-Carb 2900TR PerkinElmer* counter is also equipped with a static controller, which consists to the generation of an electric field around the sample to ionize the air, and consequently evacuate the latent electrical charges from the faces of the PE vial. In addition to this device, it is strongly recommended to pass the vials under a stream of water and pat them on paper filters before insertion in the counter. That allows to eliminate any trace of labile contamination on the outer of the vial: electrostatic phenomena may especially cause the adsorption of dust or fragments, some of which may be radioactive, and when passing through the crown, they are peeled off and may cause a deposit in the counting chamber and disrupt all subsequent measurements.

In any case, the use of the static controller generates ozone, which can increase the low energy background. Limiting its help by vials cleaning and longer count times, or waiting its dissipation by a delay before count, are therefore recommended.

Optimization of measurement conditions

Setting counting parameters

In addition to the noise reduction algorithms based on the signal-shape analysis, both coincidence time and energy windows should be adjusted depending on the radionuclide of interest. That consists in maximizing the Figure of Merit (*FoM*) as given in Eq. (1) [4].

$$FoM = \frac{\epsilon^2}{b} \tag{1}$$

where *epsilon* is the detection efficiency and *b* background noise level.

Acquisition mode

As mentioned above, the *Tri-Carb 2900TR PerkinElmer* counter provides signal-shape analysis algorithms. This is typically the case of the "High sensitivity" count mode which is designed to search for "after-pulses" in the tail of the signal (slow charge collection). This mode is the one with the best sensitivity for low activity samples counts, as shown by Table 1 where measurements made on the same sample in "Normal" and "High sensitivity" count modes are presented successively. *b* is the background, *n*, the number of hits counted in the sample and ϵ, the resulting efficiency.

Luminescence correction

The *Tri-Carb 2900TR PerkinElmer* counter provides a luminescence correction, designed to quantify and correct unexpected luminescence in liquid scintillation samples. Nevertheless, assuming weakly radioactive samples, which are expected in contamination search, correction of luminescence is not desirable since it appears disproportionate and induces more errors than it corrects, hence the importance of removing all sources of luminescence during preparation and storage of samples.

Coincidence window

When the coincidence time window is tuned, the counting rate should increase rapidly while it is shorter than the sum of the peak width and the gap between the two PMT signals. Once past this threshold, counting rate increases only because of fortuitous coincidences, regardless of the radioactivity present in the sample. The optimum setting of the coincidence window "Coincidence time" is thus the one corresponding to this transition (typically ~20 ns).

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[1] PerkinElmer – Ref: 6013321.
[2] the gloves that produce less load on friction with polyethylene are those made of nitrile then come those in vinyl and those in latex.
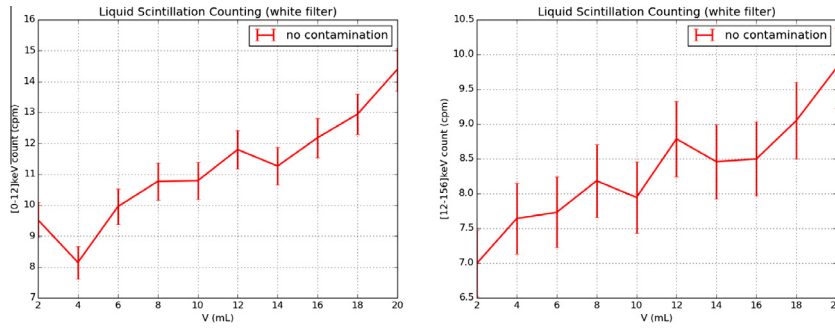
Fig. 1. Measured count rates in the ^{3}H and ^{14}C windows as a function of the volume of scintillation liquid in the vial.
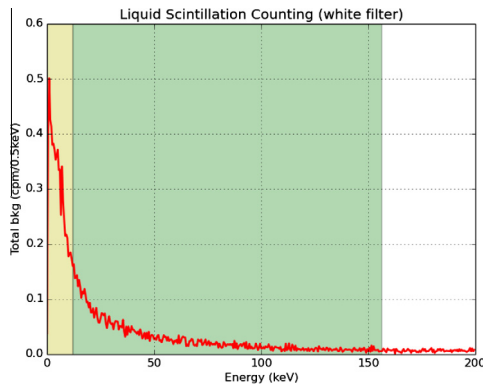

Fig. 2. Typical background energy spectrum of a *Tri-Carb 2900TR PerkinElmer* counter.

Secondly, the "Delay before burst" is the length of time that the detector delays before looking for additional pulses after the prompt pulse of an event. It has an interest for naturally slow scintillation liquids such as the UltimaGold family. A typical value of 200 ns maximizes the *FoM* for measurements of aqueous samples in 10/10 proportions with UltimaGold LLT.

Energy window

Energy window can be set by maximization of the *FoM* by changing lower and upper limits of the selected energy window (Eq. (1)). For example, in one of our experiments, for tritium, energy window that optimizes the *FoM* is the [0 : 8] keV one, as shown in Fig. 4. Furthermore, the figure of merit is deteriorated less quickly when changing the upper bound than when changing the lower one (of course because of the shape of the spectrum, dominant at low energy).

Table 1
Comparison of "Normal" and "High sensitivity" count modes from a tritiated water solution, HTO, with 450 Bq/L activity in 10/10 proportions with UltimaGold LLT; the relative uncertainties associated with these results are 1.5%.

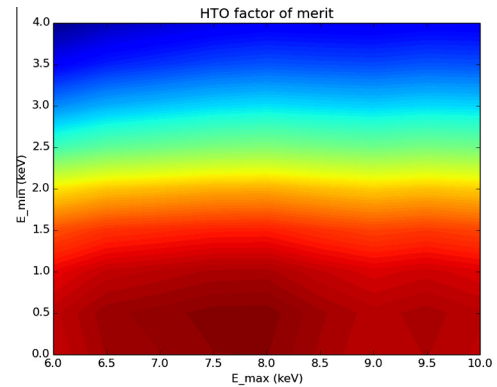| [0 : 10] keV	NORM	HS
b (CPM)	5.56	4.66
n_{HTO} (CPM)	70.36	71.10
ϵ_{HTO}	26.1	26.3
FoM	122.5	148.4

Fig. 4. Optimizing energy window for tritium count.

Of course, the figure of merit is specific to given measurement conditions and adjusting the energy window on that basis depends on the background model. Indeed, it plays a critical role in determining the limits of the counting window that maximizes the figure of merit. For an example, it is possible to select a counting window for the ^{14}C excluding any contribution of ^{3}H, but any search for ^{3}H contamination is impacted by the presence of ^{14}C
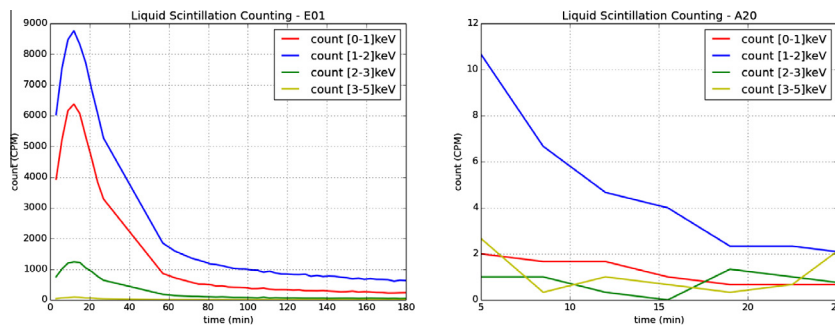

Fig. 3. Luminescence decay induced by the sunlight (left) and the light of a neon (right).

Table 2
Summary of analytical results for consumables selection. *FoM* is calculated on the basis of ^{3}H marking.

Type of vial	Type of smear	Mean *FoM*
Glass	Cotton	176.4
Glass	Folded paper	184.6
Glass	Rolled paper	203.1
Glass	None	231.1
PE	Cotton	356.2
PE	Folded paper	400.4
PE	Rolled paper	510.4
PE	None	758.3

in the cocktail, and more generally by the presence of any radionuclide with higher β energy, changing the background model.

Choice of consumables

To answer the question of the choice of consumables, series of triplets of samples have been prepared in 20 mL vials containing 10 mL of UltimaGold LLT[3] and using several types of vials and smears: glass or polyethylene (PE), with makeup remover cottons or cellulose filters,[4] the last folded or rolled, or no smear. For this purpose, "Coincidence time" and "Delay before burst" are respectively set to 20 ns and 200 ns, "Luminescence correction" is switched off and the energy window is systematically adjusted to maximize the *FoM*. As shown in Table 2, because of its best transparency, samples without smears present the highest *FoM*. Nevertheless, if a smear has to be used, the best configuration is obtained with a rolled cellulose filter in a PE vial.[5]

Vial

Polyethylene vials are the best suited for this kind of measurements. If glass vials have better transparency and prevent accumulation of electrostatic charge, the presence of ^{40}K is however a significant noise source for low activity measurements, as illustrated by lower values of the figure of merit. In addition, economic and security reasons linked to transportation and waste management encourage to use preferentially polyethylene vials.

Smear

Mechanical resistance

As shown in Table 2, only makeup remover cottons and cellulose filters have been compared as support for swipe assays. Glass fiber disks have not been considered due to their low mechanical resistance, which is critical for applications in industrial environments, as illustrated on Fig. 5.

Absorption and retention capability

By capillarity, a filter rolled into a vial containing 1 mL of scintillation cocktail is completely moistened after one hour, without stirring. Also, it does not seem necessary to completely immerse smears to measure the contamination since a light signal is produced once a beta interacts in the cocktail, i.e., once the smear is wet. Nevertheless, the light produced is proportional to the energy deposited and increases with the amount of scintillation cocktail crossed. Radionuclides whose β partially loses its energy in the scintillation cocktail cannot be discriminated from another because of a compression of its energy spectrum at low values.
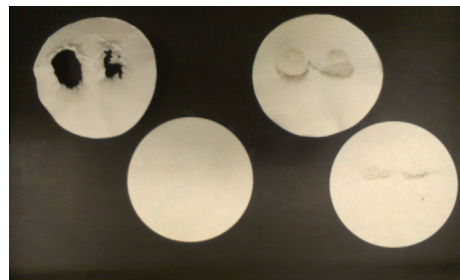
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[3] PerkinElmer – Ref: 6013371.
[4] Whatman Grade 1, paper filter – cat no: 1001-055.
[5] PerkinElmer – Ref: 6001087.

Fig. 5. Aspect of smears after being rubbed on 10 cm of concrete; top: paper filters; down: glass fiber disks; left: wet; right: dry.

That can be seen in Fig. 10, on which integrated number of events remains constant.

Optimal way to measure Contamination is then to use a rolled filter in case of single-marking, but a folded filter is preferable in any other case to maximize energy deposition in the scintillation cocktail.

Therefore, it is preferable to fold the filter enclosing the rubbed area inside, to avoid contaminating the vial during insertion of the filter, then roll it in the diameter, to promote the exchange of the rubbed area with the scintillation cocktail, as shown in Fig. 6.

Contamination collection

PerkinElmer recommends to moisten the smear using ∼100 μL of water or water/alcohol mixture in 30/70 proportions. In that case, the collection performance increases from ∼10% to ∼50% in tritium and to ∼30% in ^{238}Pu. The risk of cross-contamination is however greater due to the moisture which encourages migration of contamination to gloves, through the smear. Nevertheless, the advantage of ensuring that a maximum of potentially contaminated material is collected should take precedence over the risk of cross contamination.

Scintillation cocktail

In the panel of UltimaGold products offered by PerkinElmer [5], cocktail with the best detection efficiency for ^{3}H measurement in aqueous samples is UltimaGold (∼56%). If some classic cocktails have upper counting efficiencies, benefits for both the environment and operators of the UltimaGold[6] family compensate.

The panel of UltimaGold products remains wide and an informed choice of cocktail supposes a detailed comparison of their features: detection efficiency, miscibility of contamination and transparency, maximum load (in the case of liquid samples measurement). In particular, three have been tested in this study to identify a sufficiently generic cocktail for all the measurements taken in industrial environments, both liquid samples and smear samples:

- UltimaGold LLT, offering the best detection limits in aqueous samples, since it accepts the largest volumes of water,
- UltimaGold, whose detection efficiency for ^{3}H is the best, as mentioned above,
- UltimaGold XR recommended by PerkinElmer for search for contamination on smears.

Load

If the maximum load for UltimaGold LLT is 100% in water (10 mL of water for 10 mL of UltimaGold LLT), allowed loads for UltimaGold and UltimaGold XR vary differently, as shown in Fig. 7.

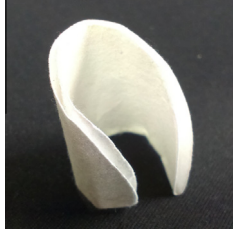[6] Using DIPN (di-isopropylnaphthalene) as solvent.

Fig. 6. Folding/rolling strategy recommended for the sampling of paper smears.

A scintillation liquid with a larger load capacity allows to simultaneously measure a larger amount of sample, making it preferable.

Transparency

The addition of aqueous solution contributes to decrease the transparency of the cocktail, as shown in Fig. 8 where 10 mL of water has been gradually added to 10 mL of scintillation liquid. In particular, 10 mL of water in 10 mL of UltimaGold induces two phases, as shown in Fig. 9. This phenomenon, called "demixing of the cocktail", is not necessarily immediate, and therefore it is recommended to ensure the homogeneity of the cocktail after counting.

If higher transparency increases the measurement efficiency, it also avoids the compression of the spectra and promotes discrimination in cases of multi-marking (like that may be the case of samples we have to consider), making UltimaGold XR disadvantaged because of a lower discrimination capability.

Scintillation liquid quantity

As mentioned above, for low energy β emitters, a very small amount of scintillation liquid is enough to convert their energy into light, because of their short range in matter. For β with a larger energy, however, a larger amount of scintillation liquid is required to convert all their energy. This is shown in Fig. 10 on the ^{137}Cs spectrum.

To ensure the complete immersion of the smear in the liquid scintillation cocktail and promote the distribution of a maximum of light, it is recommended to fold it and add 10 mL of scintillation liquid for measurement. In the case of smears in which only one radionuclide is searched, however, a lesser amount of cocktail can be used and vials of 7 mL may be enough.

For liquid samples, it is entirely appropriate to use the same amount of scintillation liquid, which also allows to standardize practices. Then, the maximum amount of sample that can be introduced depends on the nature of both the sample and the scintillation liquid which are mixed. For aqueous samples, maximum loads to 10 mL of scintillation liquid are:

- 2 mL with UltimaGold,
- 5 mL with UltimaGold XR,
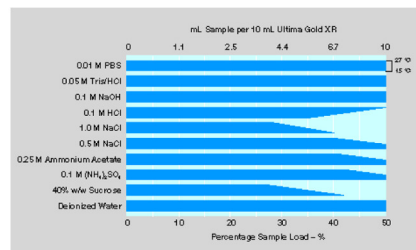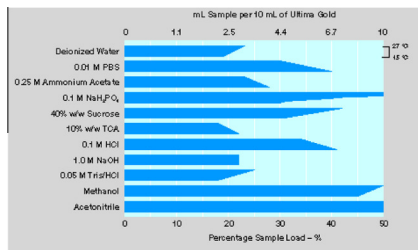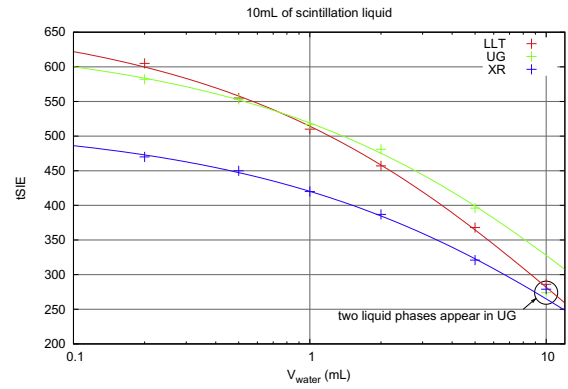- 10 mL with UltimaGold LLT.

Fig. 8. Quenching value (tSIE) as a function of the amount of aqueous solution introduced in 10 mL of scintillation liquid.
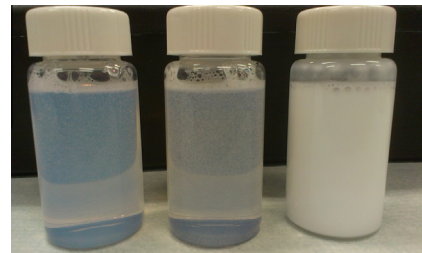

Fig. 9. Photography of vials after adding aqueous 10/10 proportions; left: UltimaGold LLT; center: UltimaGold XR; right: UltimaGold.
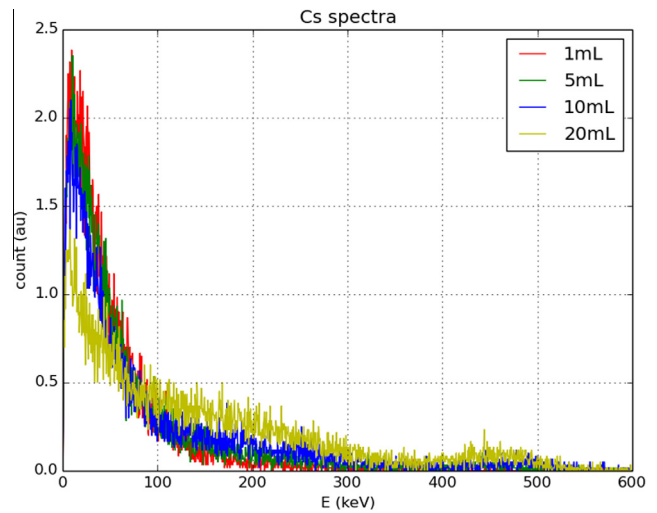

Fig. 10. ^{137}Cs spectra built with different amount of scintillation cocktail, from 1 mL to 20 mL.

Fig. 7. Maximum load for UltimaGold (left) and UltimaGold XR (right), for various liquid samples.

Table 3
Maximum *FoM* results for optimal energy windows using UltimaGold LLT; maximum load is 10 mL of aqueous sample.

Solution volume (mL)	Window ^{3}H (keV)	*FoM* ^{3}H	Window ^{14}C (keV)	*FoM* ^{14}C	Window ^{137}Cs (keV)	*FoM* ^{137}Cs	Window ^{40}K (keV)	*FoM* ^{40}K
0.2	[0.5 : 7.5]	**277.1**	[10.5 : 94.0]	**891.2**	[27.5 : 302.0]	**73.1**	[88.0 : 732.0]	**1192.8**
0.5	[0.5 : 7.0]	**266.3**	[10.5 : 78.0]	**867.9**	[27.5 : 258.5]	**567.8**	[105.0 : 722.5]	**1285.0**
1.0	[0.5 : 6.5]	**252.9**	[11.5 : 73.5]	**866.2**	[28.5 : 235.5]	**904.8**	[104.0 : 705.0]	**1301.3**
2.0	[0.5 : 6.0]	**228.9**	[9.5 : 60.5]	**807.5**	[18.0 : 339.0]	**900.1**	[89.5 : 634.0]	**962.1**
5.0	[0.5 : 5.0]	**189.4**	[9.0 : 50.0]	**759.8**	[16.5 : 305.0]	**823.2**	[89.0 : 520.0]	**891.1**
10.0	[0.5 : 3.5]	**129.2**	[6.0 : 39.5]	**670.9**	[12.5 : 237.0]	**677.8**	[110.0 : 420.5]	**760.4**

Table 4
Maximum *FoM* results for optimal energy windows using UltimaGold; maximum load is 2 mL of aqueous sample.

Solution volume (mL)	Window ^{3}H (keV)	*FoM* ^{3}H	Window ^{14}C (keV)	*FoM* ^{14}C	Window ^{137}Cs (keV)	*FoM* ^{137}Cs	Window ^{40}K (keV)	*FoM* ^{40}K
0.2	[0.5 : 7.0]	**309.4**	[11.5 : 81.5]	**915.1**	[24.5 : 425.5]	**90.5**	[65.5 : 752.0]	**1315.6**
0.5	[0.5 : 7.5]	**280.9**	[12.5 : 73.0]	**891.2**	[29.5 : 466.5]	**606.2**	[66.0 : 728.5]	**1230.5**
1.0	[0.0 : 8.0]	**254.3**	[12.5 : 73.0]	**896.9**	[29.5 : 438.5]	**909.0**	[94.5 : 735.0]	**1274.5**
2.0	[0.5 : 7.0]	**243.0**	[12.5 : 65.0]	**825.9**	[29.5 : 385.0]	**953.1**	[87.0 : 712.5]	**871.1**
5.0	[0.5 : 4.5]	200.6	[7.5 : 56.5]	783.1	[19.5 : 324.5]	890.6	[119.5 : 686.0]	447.9
10.0	[0.5 : 4.0]	53.2	[5.5 : 46.5]	547.8	[14.5 : 256.5]	717.0	[110.5 : 636.0]	333.5

Table 5
Maximum *FoM* results for optimal energy windows using UltimaGold XR; maximum load is 5 mL of aqueous sample.

Solution volume (mL)	Window ^{3}H (keV)	*FoM* ^{3}H	Window ^{14}C (keV)	*FoM* ^{14}C	Window ^{137}Cs (keV)	*FoM* ^{137}Cs	Window ^{40}K (keV)	*FoM* ^{40}K
0.2	[0.5 : 5.5]	**226.6**	[8.5 : 67.5]	**783.0**	[24.5 : 227.5]	**71.9**	[104.5 : 632.0]	**991.0**
0.5	[0.5 : 5.5]	**226.6**	[9.5 : 62.5]	**793.0**	[20.0 : 370.5]	**561.2**	[87.0 : 682.0]	**1163.8**
1.0	[0.0 : 5.5]	**232.4**	[10.0 : 62.5]	**819.7**	[20.0 : 352.5]	**841.7**	[85.5 : 604.5]	**1262.4**
2.0	[0.5 : 5.0]	**209.7**	[9.5 : 52.5]	**763.0**	[20.0 : 316.5]	**902.2**	[63.0 : 579.5]	**944.3**
5.0	[0.5 : 3.5]	**182.9**	[6.5 : 46.0]	**714.7**	[24.0 : 259.0]	**857.8**	[68.0 : 459.5]	**882.3**
10.0	[0.5 : 3.0]	108.3	[3.5 : 33.0]	610.7	[10.0 : 207.5]	659.9	[82.0 : 408.5]	767.9

Counting windows and detection limits

Counting windows and figure of merit

Several measurements have been performed to assess the figure of merit associated with the measurement of increasing amounts of contamination with the three mentioned scintillation liquids. Four radionuclides were used, whose characteristics are detailed here:

- Organically Bound Tritiated (OBT) solution (E_β = 18.6 keV), pH 7, activity of 247 Bq/mL,
- ^{14}C solution (E_β = 157 keV), pH 7, activity of 21.8 kBq/mL,
- ^{137}Cs solution (E_β = 512 keV), pH 1, activity of 18.35 Bq/mL,
- ^{40}K solution (E_β = 1312 keV), pH unknown, activity of 1.5 Bq/mL.

The results of these measurements are summarized in Tables 3–5 in which, besides the figure of merit (in bold), is indicated the counting window that maximizes it. Grayed lines correspond to samples whose volume exceeds the maximum load for the scintillation cocktail used.

For example, Figs. 11–14 present the measured spectra and the result of the optimization algorithm of the figure of merit for 2 mL of contaminated solution mixed with 10 mL of UltimaGold scintillation liquid.

As noted in the preceding paragraph, windows that maximize the figure of merit are narrower for UltimaGold XR than for other proposed scintillation liquids, due to the compression of the spectrum caused by light attenuation.

In particular, Fig. 14 shows that on a fairly wide range, the figure of merit associated with ^{40}K is quite insensitive to the lower limit of the energy window.

Efficiency and detection limits

For each of the configurations described, the detection limit is calculated according to Eq. (2) [6]:

$$LD(Bq/L) = \frac{4}{T_c} \times \left[1 + \sqrt{1 + 2 \times (b \times T_c)}\right] \times \frac{1}{\epsilon \times V} \qquad (2)$$

where T_c is the counting time (in s), b, the expected noise level (in CPS), ϵ, the detection performance for the relevant radionuclide (in %) and V, the volume of the measured sample (in L).

Detection limit, in this form, is expressed at 2σ. Values are reported in Fig. 15 for each radionuclide ^{3}H, ^{14}C, ^{137}Cs and ^{40}K considering ten minutes of counting time.

Based on these results, the UltimaGold seems to offer the best compromise for a standardized sampling procedure for both smears and liquids' measurements. In both cases, the cocktail should contain 10 mL of UltimaGold, enabling further standardization in preparation methods.

Under these conditions, and assuming adding 2 mL of liquid sample (maximum load allowed), the detection limit is ~100 Bq/L for tritium in a liquid sample and ~50 Bq/L for other mentioned
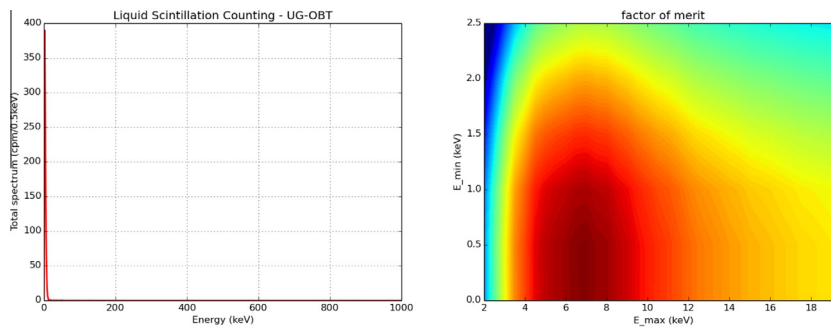
Fig. 11. OBT spectrum and optimization algorithm of the figure of merit for UltimaGold.
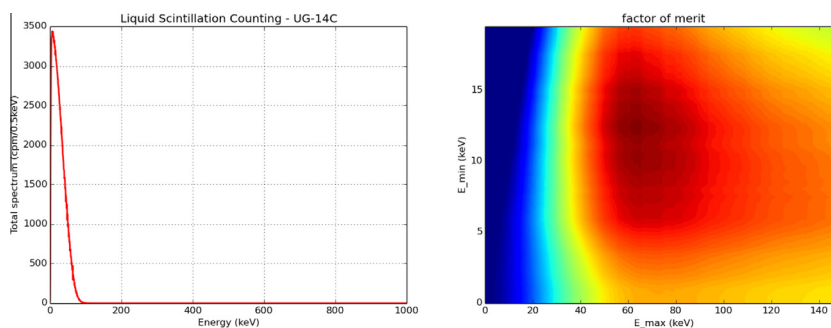

Fig. 12. ^{14}C spectrum and optimization algorithm of the figure of merit for UltimaGold.
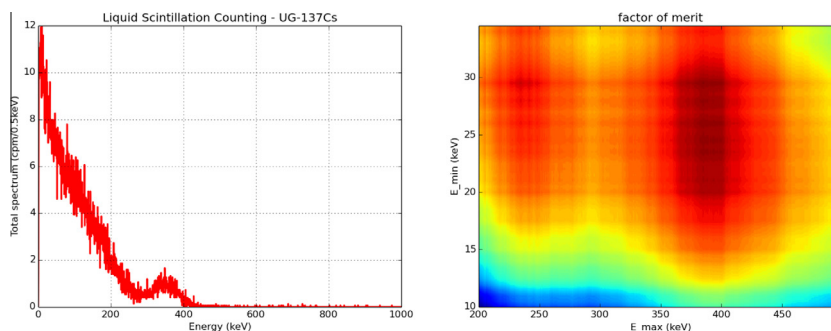

Fig. 13. ^{137}Cs spectrum and optimization algorithm of the figure of merit for UltimaGold.

radionuclides after a reference measurement of ten minutes. For comparison, the detection limits achieved in ten minutes with 10 mL of the same sample mixed to 10 mL of UltimaGold LLT is ~25 Bq/L for tritium and ~10 Bq/L for other radionuclides. Achieving similar sensitivities with UltimaGold requires counting time about twenty times higher. The UltimaGold LLT remains the scintillation liquid most suitable for measurement of aqueous samples of very low activities, but these are not the most common in industrial environments.

Impact of the choice of standards for disintegrations per minute (DPM) measurements

In order to quantify the exact amount of radioactivity in a sample, the identity of the radioactive nuclide as well as a corresponding calibration factor has to be known. In liquid scintillation, this factor is determined by the use of several quenched standards whose activity is well known but with a different quench indicator (e.g. tSIE). The radionuclide used is the one expected in the sample.

This enables to quantify the efficiency of the liquid scintillating measurement as a function of the quench indicator.

In the following tests, different kinds of standards have been used:

- PerkinElmer standards,
- ACRO standards in 10/10 proportions (20 mL),
- two homemade with a solid opacifying agent (dust) with UltimaGold LLT (10 mL – solid).

Finally, we used samples we previously prepared with different scintillation liquids as additional calibration points, given that their activities are known and consequently associated detection efficiency can be calculated.

PerkinElmer standards were counted both in "Normal" and "High sensitivity" count modes. All these data are superimposed on Figs. 16 and 17 for ^{3}H and ^{14}C respectively.

Except the UltimaGold sample with a value of tSIE close to 280 (corresponding to a 10/10 proportion which does not allow the
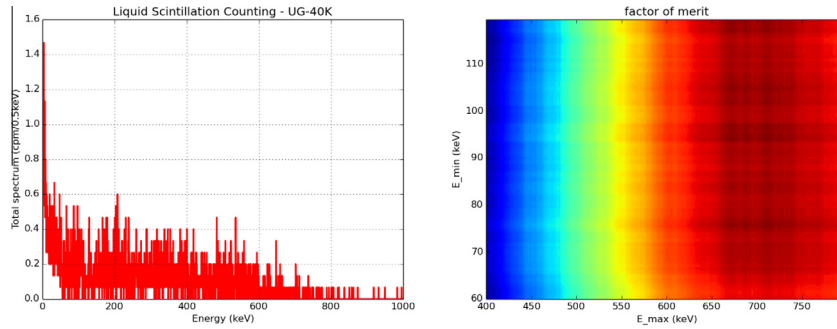
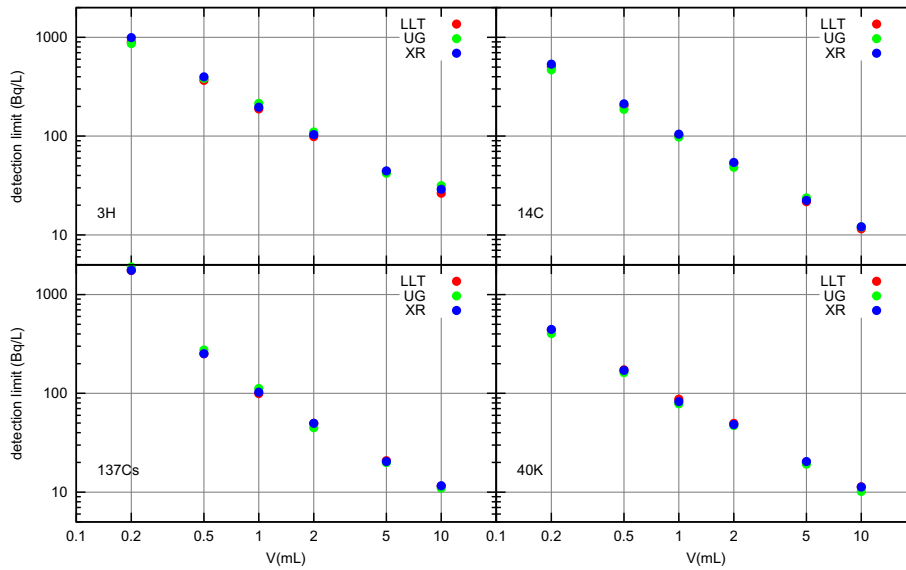**Fig. 14.** ^{40}K spectrum and optimization algorithm of the figure of merit for UltimaGold.

Fig. 15. Detection limits for contaminations searches in ^{3}H, ^{14}C, ^{137}Cs and ^{40}K, defined in windows deemed to promote factor of merit seeing Tables 3–5 for each scintillation liquids considered in this study (in "Normal" count mode).

uniformity of the cocktail (Fig. 8)), detection efficiency in ^{3}H liquid samples is related to the quenching indicator by a polynomial law checked better than 5% whatever the volume and the type of the scintillation cocktail. Regarding samples containing dust, they have consistently about 5% lower activities than measured on other samples. This is due to the inhomogeneity of the cocktail. Indeed,
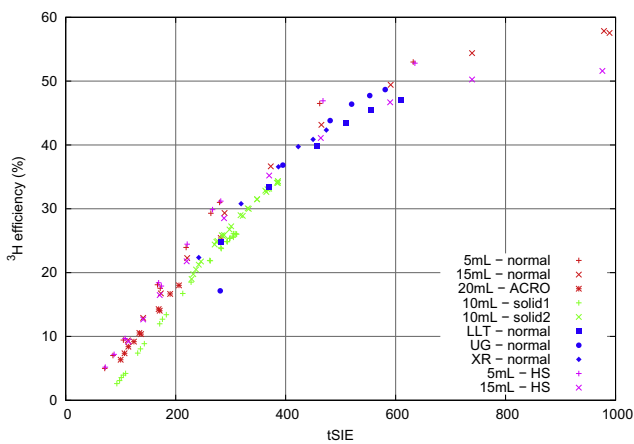

Fig. 16. ^{3}H measurement efficiency as a function of the quenching indicator. Energy window is $[0 : 8]$ keV. In red: PerkinElmer standards in "Normal" count mode; in green: using dust as solid opacifying agent in "Normal" count mode; in pink: PerkinElmer standards in "High sensitivity" count mode; in blue: previous liquid samples in "Normal" count mode. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 17. ^{14}C measurement efficiency as a function of the quenching indicator. Energy window is $[12 : 80]$ keV. In red: PerkinElmer standards in "Normal" count mode; in pink: PerkinElmer standards in "High sensitivity" count mode; in blue: previous liquid samples in "Normal" count mode (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
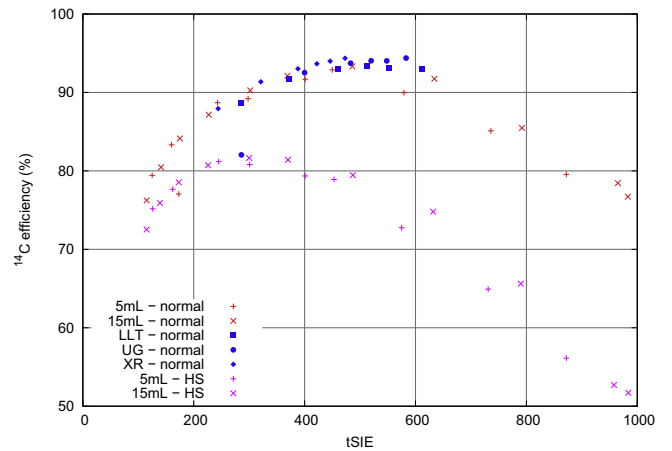
dust preferentially deposited at the bottom of the vial resulting in a measurement bias in the quenching indicator. ^{3}H contamination measured from a smear sample presents then an error higher than 25% if the tSIE is less than 200, as shown in Fig. 16.

These curves also show the impact of the counting mode (i.e. normal or HS) on the resulting efficiency and the additional error that can be made in a DPM measurement. One may conclude following these points that the count mode should be the same as that of the quench curve determination.

Conclusion

Facilities of a laboratory in terms of temperature and lighting have to be implemented to ensure a constant temperature in the environment in which are prepared and measured samples. These are also to be protected from direct light source (including sunlight), to limit risks of photoluminescence.

Furthermore, based on an optimization of the figure of merit, the most suitable consumables are 20 mL polyethylene vials, containing:

- 10 mL of UltimaGold scintillation liquid to measure radioactivity on smears with cellulose filter folded and rolled, placed at the bottom of the vial,
- 10 mL UltimaGold scintillation liquid to measure radioactivity in liquids with 2 mL of liquid sample mixed.

Given the levels of considered activities, settings of the counter should be as follows:

- assay types: CPM,
- static controller: ON,
- high sensitivity count mode: ON,

- luminescence correction: OFF,
- quench indicator: tSIE/AEC,
- coincidence time: 20 ns,
- delay before burst: 200 ns.

Moreover, the passage of vials under a stream of water before measurement is recommended and a waiting time of two hours in the dark prior to the first vial counting is desired.

Under these requirements, best counting windows suited to the desired radionuclides are:

- [0 : 8] keV for ^{3}H (eff>~40%),
- [12 : 80] keV for ^{14}C (eff>~70%),
- [25 : 400] keV for ^{137}Cs (eff>~80%),
- [100 : 750] keV for ^{40}K (eff>~75%).

These windows and associated detection efficiencies are indicative and may be refined according to the measurement conditions, particularly depending on whether the sample type is a liquid or a smear.

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