What can you say when there is almost nothing? Decision thresholds associated with multiple measurements and their application to environmental monitoring results

Alain Vivier[1], Guillaume Manificat[2]

[1] CEA, Institut National des Sciences et Techniques nucléaires, Centre de Saclay 91191 Gif sur Yvette Cedex France. Tel 33(0)1.69.08.26.56, alain.vivier@cea
[2] IRSN, Service d'étude et de surveillance de la radioactivité dans l'environnement, BP 40035 - 78116 Le Vésinet Cedex France guillaume.manificat@irsn.fr

When sample activity is measured several times for various reasons, then with each measurement can be associated an individual decision threshold and limit of detection. Each measurement can be analyzed through its own decision threshold. The whole measurements can sometimes present contradictory results, certain measurements being lower than the decision threshold and other higher. The problem then arises to build a decision threshold and a detection limit taking into account all the individual results and to decide if the radioactivity is finally detected or not. It is interesting to note that the global decision threshold, taking account all individual results, could enable the analyst to decide that the radioactivity is present whereas each individual results is negative in terms of individual decision threshold. We are able to show how these thresholds and these coherent limits cumulated can be determined in way according to the experimental conditions. In a general way a rigorous method of cumulating makes it possible to systematically decrease the decision threshold and limit of detection in terms of activity. This approach has interesting applications in gamma spectrometry with multi-emitters, radioactive surveys or periodical environmental measurements. On the basis of measurements realized by the IRSN within the framework of the national monitoring of the environment, we will see the potential impact of these methods on the final assessments.

Key words: decision threshold, detection limits, multiple measurements

1 INTRODUCTION

The experimental situations where it is possible to cumulate multiple measurements are varied but can be distributed according to three possible general outlines:

- **Case n°1 (repeatability):** Iteration of measurements on the same sample under identical measurements conditions, except possibly the duration of counting.
- **Case °2 (simple reproducibility):** reproduction of measurements on the same sample under conditions of different measurements: efficiencies, systems of measurement …
- **Case n°3 (widened reproducibility):** multiple measurements of different samples but obtained starting from a single measurand to be defined.

In all cases, the objective is to consider situations where the accumulation of the obtained values has a sense, wether this accumulation corresponds to the sum of individual measurements, or their algebraic or weighted average, or any other statistics. Let us consider for example the current situation of a measurement by gamma spectrometry of a sample likely to contain a multi-emitter gamma radionuclide. Taking into account all the n regions of interest (*ROI*) of this radionuclide on the experimental spectrum falls under the case n°2. Indeed, each ROI can be considered as a specific nuclear counter of a specific photons energy emitted by this radionuclide. Each one of these "counters" can be used separately to estimate the activity of this radionuclide. But the energy photons, the intensities of emission and the efficiencies are different from one ROI to another, thus answering the criterion of reproducibility. Thus is obtained n estimations A_n of the same true activity $\hat{A}$ under reproducibility conditions (case n° 2). These estimations have vocation to be of the same order of magnitude, but not their uncertainties nor their decisions thresholds.

One can show that in such a situation the best estimation of the true value consists in taking the average of the n measured activities weighted by precision[1]. This corresponds to taking into account all the available information in

[1] *Precision can be defined like the inverse of the square of uncertainty. Thus to a small uncertainty corresponds a high degree of precision. It is also the quantity of information in the sense of Fischer.*

the spectrum, step called here in a generic way "accumulation", and not only the best individual estimation, usually the one having the smallest uncertainty, equivalent to the "fuller" peak. One can then show that the estimation by accumulation is always better than the best individual estimations. This result is intuitively understood as each measurement accumulated brings additional information, which can only improve final information. It is obviously in the best interest of the expert to use all the information available whenever possible.

In gamma spectrometry the use of the weighted average with all peaks of the same radionuclide does not pose any problem if they "are sufficiently filled" and interference-free. The calculation of the weighted average does not create any problem when all estimations are higher than their decision thresholds, and thus considered to be significant of a proven presence of the radionuclide. Problems appear when some of the n measurements are lower than their respective decision thresholds. When this occurs, one decides that the effect is not detected and declare "< LD". The true value, in case of a radioactive sample, is then at least lower than the detection limit associated to the considered ROI.

Thus when *m* measurements out of *n* are declared as "< LD", several questions appear:

- Question 1: is it necessary to integrate these m nonsignificant measurements into the accumulation?
- Question 2: if "yes", how can one take into account information like "<LD"?
- Question 3: if "not", can neglecting such result lead to the risk of biasing the final result?
- Question 4: if "not" what can be done if all measurements are lower than their respective decision threshold?
- Question 5: what are the decision threshold and the detection limit corresponding to the final weighted average?
- Question 6: can one, in a way or another, use the n individual decision thresholds to determine the decision threshold on the result obtained by accumulation?

Various methods exist in the literature to try to exploit nonsignificant measurements [1] [2]. These methods can be rather heavy to implement and vary according to the proportion *m/n*. We will not approach these methods here but they are only stopgap methods. The aim of this article is to answer these various questions, and to show the interest of cumulating multiple measurements correctly whenever possible. We will see that this interest increases withthe number of available measurements. In some experimental situations, like the periodic or continuous follow-up of the radioactivity monitoring in the environment, where measurements are often numerous and close to the decision thresholds, the impact of these methods on the end result can often be spectacular.
One of the most important parts of the answer is that one does not need to know, in the aim of analysing a cumulated result, if individual measurements are lower or not than their own decision threshold. In fact, taking care of only these individual information to make a final decision on cumulated results is not only useless but raises insurmountable difficulties.

2 DETERMINATION OF THE DECISION THRESHOLD CUMULATED IN THE CASE OF COUNTING REPETITION

2.1 Description of the repeatability process and analyze of individual results

On the basis of a simple situation, corresponding to the case n°1, it is possible to lay down without difficulties the principal rules which one will find in all possible cases. Moreover this simple situation of accumulation is already implicitly at work when one takes a single measurement of activity starting from a single background counting and a single sample raw counting. A ten seconds single measurement is nothing else than ten one second measurements accumulation.

One considers here a series of ten measurements of the same sample under repeatability conditions: same measurement device, same distance source-detector, same background condition, same counting duration. For each individual measurement "i" one can distinguish several phases in the operation from measurement to final analysis:

Phase 1: construction of the decision-making aid tool:

- Background counting for a duration T_0 identical to the duration of sample counting noted T_S: the result is noted BkG_i
- Determination of the decision threshold in terms of net counting value following the formula $Lc_{Net\,i} = k_{1-\alpha}\sqrt{2(BkG_i+1)}$ (1)

- Determination of the detection limit in terms of net counting value following the formula $LD_{Net\,i} = k_{1-\beta}^2 + 2Lc_{Net\,i}$ (2), corresponding to the case of alpha and beta identical risks.

In Eq. (1), 1 extra count appears naturally in a Bayesian determination of decision threshold, preventing us from getting zero in case of a zero background [3].

Phase 2: sample measurement:

- Gross count sample: the result is noted $Gross_i$
- Determination of net counting value $Net_i = Gross_i - BkG_i$

Phase 3: analysis of the result:

- Decision threshold Test : $Net_i > Lc_{Net\,i}$?
- If "yes" the effect is considered detected. Estimation of true average net counting value and associated uncertainty $\hat{\mu}_{Net} = Net_i \pm k\sqrt{Net_i + 2BkG_i}$ (typically k=1, 2, or 3)
- If "not" the effect is regarded as not detected. Final expression of the result:

$$"< LD_{Net}" \quad \Leftrightarrow \quad 0 < \mu_{Net} < LD$$

Numerical example:

	Phase 1			*Phase 2*		*Phase3*			
	BkG_i	$Lc_{Net\,i}$	**1.** $LD_{Net\,i}$	$Gross_i$	Net_i	*Net> L*	*Resu*	*uncertainty (*	*Relative uncertain*
meas.	100	28	59	143	43	yes	43	31	73%
meas.	96	27	58	148	52	yes	52	31	60%
meas.	113	29	63	130	17	***no***	< LD	/	/
meas.	117	30	64	158	41	yes	41	33	81%
meas.	142	33	70	134	-8	***no***	< LD	/	/
meas.	110	29	62	154	44	yes	44	32	74%
meas.	126	31	66	142	16	***no***	< LD	/	/
meas.	138	33	69	138	0	***no***	< LD	/	/
meas.	103	28	60	152	49	yes	49	32	65%
meas.	104	28	60	123	19	***no***	< LD	/	/

Table 1: individual analyzes of 10 counting values of the same sample (alpha risk error at 2,5%)

2.2 Accumulation of the results by summation of individual countings

The objective of the accumulation is to synthesize all the information in only one line. Again appears the problem to integrate into this accumulation information of the type "<LD". However in this precise case the starting point of this synthesis is commonplace. Indeed, and for purely physical reasons, the two series of raw counts and background noise can be perceived like intermediate results partial of two single countings: a single background counting value during *10*xT_s and a raw count single of the same duration. This simple report will make it possible to carry out the accumulation without being concerned with knowing wether each result is higher or lower than its threshold of decision.

Thus a fundamental rule appears in this type of problem:

✓ **Rule n° 1**: information of the phase of analysis of individual measurements (phase 3) should not be taken into account in the development of the accumulation and it is necessary to preserve, in the objective to carry out an accumulation, all information of phases 1 and 2 of all individual measurements, whatever they are, including when the net amounts are negative:

Phase 1			*Phase 2*	
BkG_i	$Lc_{Net\,i}$	$LD_{Net\,i}$	$Gross_i$	Net_i

meas.	100	28	59	143	43
.					
.	.	.	.	.	.
meas.	104	28	60	123	19

Table 2: information to be preserved before acccumulation

The operation of accumulation consists in this particular case to consider a single measurement with background value $BkG_\Sigma = \sum_{i=1}^{n} BkG_i$ (3) and a gross measurement value $Gross_\Sigma = \sum_{i=1}^{n} Gross_i$. Insofar as the sum preserves the Poisson character of the counting values, these two cumulated counting values make it possible to apply the same previous procedure:

Phase 1: the construction of the decision-making aid tool of on the cumulated result:

- Background counting value equal to BkG_Σ .
- Determination of the decision threshold "accumulation ": $Lc_{\Sigma Net} = k_{1-\alpha}\sqrt{2BkG_\Sigma}$ (4)
- Determination of the detection limit "accumulation " $LD_{\Sigma Net} = k_{1-\beta}^2 + 2Lc_{\Sigma Net}$ (5),

In equation (4), 1 extra count was not added to the background variance for simplicity.

Phase 2: cumulated sample measurement:

- Gross sample count: the result obtained is noted $Gross_\Sigma$
- Determination of net counting value $Net_\Sigma = Gross_\Sigma - BkG_\Sigma$

Phase 3: analysis of the cumulated result:

- Decision threshold test $Net_\Sigma > Lc_{\Sigma Net}$?
- If "yes" the effect is considered detected. Estimation of true average counting and associated uncertainty $\mu_{Net_\Sigma} = Net_\Sigma \pm k\sqrt{Net_\Sigma + 2BkG_\Sigma}$

  If "not" the effect is regarded as not detected. Final expression of the result: $"< LD" \quad \Leftrightarrow \quad 0 < \mu_{Net} < LD$

It should be noted that the measurand of interest Net_Σ is also equal to $Net_\Sigma = \sum_{i=1}^{n} Net_i$.

Applied to the previous example the following cumulated result is obtained:

	Phase 1			*Phase 2*		*Phase3*			
	BkG_Σ	$Lc_{\Sigma Net}$	$LD_{\Sigma Net}$	$Gross_\Sigma$	*Net*	*Net> l*	*Resu*	*Uncertainty (*	*Relative uncertain*
Accumul. by summ	1149	94	192	1422	273	yes	273	101	37%

Table 3: accumulation by summation

The obvious result showing and justifying the general interest of the accumulation is here the final relative uncertainty equal to 37%, whereas for the whole of the individual results, the average of the relative uncertainties is much higher (see Table 1 or 2 above). One second less obvious indication but of interest to be notified is the fact that the end result in terms of net counting value (273) is now largely higher not only than the decision threshold but also than the detection limit (192). That confirms that the presence of radioactivity is confirmed with a very great probability[2], whereas five individual measurements on the ten did not confirm this presence. Concluding from this last remark that the probability for the sample to be nonradioactive could have been be equal to 50 % would have been strong pessimist (or extremely optimist if it had been preferred that it is not so).

[2] *Rigorous calculation shows that with such values the residual probability that the' sample is nonradioactive is equal to* 3.10^{-8} .

2.3 Determination of the decision threshold on the accumulation starting from the individual thresholds of decision

The accumulation method used here did not require knowing preliminary n individual decision thresholds and detection limits, which seems to contradict the rule n°1 which recommends keeping them. In fact we will show now that these values can be employed to directly find the decision threshold suitable for the cumulated value.

If one considers relations 1, 2 and 4, one can write:

$$\begin{cases} (1)\ \ Lc_{\Sigma Net} = k_{1-\alpha}\sqrt{2BkG_{\Sigma}} \\ (2)\ BkG_{\Sigma} = \sum_{i=1}^{n} BkG_i \\ (4)\ Lc_{Net\,i} = k_{1-\alpha}\sqrt{2BkG_i} \end{cases} \Rightarrow \begin{cases} Lc_{\Sigma Net} = k_{1-\alpha}\sqrt{2\sum_{i=1}^{n} BkG_i} \\ BkG_i = \dfrac{Lc_{Net\,i}^2}{2k_{1-\alpha}^2} \end{cases} \Rightarrow Lc_{\Sigma Net} = k_{1-\alpha}\sqrt{2\sum_{i=1}^{n}\dfrac{Lc_{Net\,i}^2}{2k_{1-\alpha}^2}} = \sqrt{\sum_{i=1}^{n} Lc_{Net\,i}^2}$$

One thus obtains a simple relation between the decision threshold on the cumulated net value and the partial decision thresholds. It is important to note that this relation is similar to the relation between uncertainty on the cumulated result and the partial results. Indeed with u_{Σ} uncertainty associated to Net_{Σ} and u_i uncertainties associated to the partial results Net_i, it is well known that the relation $Net_{\Sigma} = \sum_{i=1}^{n} Net_i$ allows expressing associated uncertainty according to the relation $u_{\Sigma} = \sqrt{\sum_{i=1}^{n} u_i^2}$. This analogy is not worth demonstration, but becomes more relevant if one remembers that basically $Lc = k_{1-\alpha}\sigma_{H0}$ [3], with σ_{H0} the standard deviation of the distribution of the measurements under H_0 hypothesis (nonradioactive sample). Being homogeneous with a standard deviation, the decision thresholds "are propagated" in the model of the cumulated result following the same algebra as uncertainties[3]. This report makes it possible to state the following rule:

✓ **Rule n° 2:** the decision thresholds follow an algebra of standard deviation and are composed in operations of accumulation according to the same formula as uncertainties.

This powerful rule will enable us to carry out an accumulation of ten measurements lots by carrying out a "natural" sum but by considering the averages of the background and gross counting values.

Concerning the limit of detection, the rigorous resolution is more delicate (except in this example, cf. eq. 5) as the limit is not perfectly homogeneous with a standard deviation because of the term $k_{1-\beta}^2$ in the expression (5). However, by keeping the approximate relation $LD \approx 2Lc$ (beta and alpha risk errors are assumed to be the same), one can estimate the cumulated detection limit while applying:

- Either the rule n°2 on the individual detection limits. Following this rule one obtains in our example $LD_{\Sigma Net} \approx \sqrt{\sum_{i=1}^{n} LD_{Net\,i}^2} = 200$
- Or directly the expression $LD \approx k_{1-\beta}^2 + 2Lc_{\Sigma Net}$ giving here $LD_{\Sigma Net} \approx 192$

These approximations are admissible as the thresholds and limits calculated starting from the experimental values are, like all experimental statistics, sullied with uncertainty. A calculation show here that the uncertainty-type (k=1)

---

[3] *In the approach of the standard ISO 11929 one finds this property, insofar as the decision threshold is written* $Lc = k_{1-\alpha}\tilde{u}(0)$. *Although conceptually debatable,* $\tilde{u}(0)$ *is also a standard deviation.*

on the exact value of the detection limit is equal to $u_{LD_\Sigma} \approx 5$ [3], the expanded uncertainty (k=2) $U_{LD_\Sigma} \approx 10$, which makes it possible to accept these approximations.

2.4 Accumulation of the results by average of the individual results

It is quite intuitive to synthesize these ten measurements by carrying out the average of the counting values over one duration T_s. One can note that this average has a sense insofar as countings are carried out under conditions of repeatability for identical durations. Following rule n° 1, i.e. without being concerned with knowing if the individual results are higher or not than the individual decision thresholds, one can write the final result cumulated on the statistics of interest:

$$\overline{Net}_n = \frac{\sum_{i=1}^{n} Net_i}{n} = 27{,}3\,.$$

To determine if this mean value is significant or not, we have to calculate a decision threshold Lc coherent with these statistics. The uncertainty $u_{\overline{Net_n}}$ associated to $\overline{Net}_n$ is equal to : $u_{\overline{Net_n}} = \frac{1}{n}\sqrt{\sum_{i=1}^{n} u_i^2}$.

Rule n° 2 then tells us that the "cumulated" decision threshold is equal to: $Lc_{\overline{Net_n}} = \frac{1}{n}\sqrt{\sum_{i=1}^{n} Lc_{Net\,i}^2}$ (6)

That is to say here $Lc_{\overline{Net_{10}}} = 9{,}4$. The accumulation by the averaging operation makes it possible to obtain the following information:

	Phase 1			Phase 2		Phase3			
	$\overline{BkG}_n$	$Lc_{\overline{Net_n}}$	$LD_{\overline{Net_n}}$	$\overline{Gross}_n$	$\overline{Net}_n$	Net> L	Resu	Expande uncertainty (	Relative uncertain
Avera	114,9	9,4	20.0	142.	27.3	yes	27.3	10	37%

Table 4: accumulation by average

In this particular case of repeatability, the individual decision thresholds are all of the same order of magnitude (see table 1). There is then the approximate relation $\sum_{i=1}^{n} Lc_{Net\,i}^2 \approx n.\overline{Lc}_{Net}^2$ and the relation (6) can be approximated according to the relation: $Lc_{\overline{Net_n}} \approx \frac{1}{n}\sqrt{n.\overline{Lc}_{net}^2} = .\frac{\overline{Lc_{Net}}}{\sqrt{n}}$ (7).

This shows that the decision threshold on the mean value decrease like the inverse of the square root of the number of individual measurements and show the interest of such an accumulation. It will be noted that uncertainty associated with the average decreases in the same proportions. A*n error which one should avoid is to determine here a decision threshold according to the same algebra, leading to* $Lc_{\overline{Net_n}} = \overline{Lc_{Net}}$.

If one had eliminated from the final assessment the five values corresponding to the declaration" < LD", one would have obtained a net mean value $\overline{Net}'_5 = 46$ instead of $\overline{Net}_{10} = 27$. The values presented here come from a data-processing simulation and the true net average is equal to μ_{Net}=23. This simple example shows that not taking into account measurements of the type "< LD" in the accumulation can give biased positive final values.

For this reason, it is important not to erase the experimental values of counting operation in the tables of individual results, if these results are likely to be one day cumulated in a way or of another (rule n°1). This is particularly true in environmental or discharge measurements, usually carried out over long periods. It then happens too often that the recorded results exclude the rough experimental data to store only information of the type "<LD", information, not usable in the accumulation.

To illustrate a completely no-intuitive result, it is possible that all the partial results (table 5) are found nonsignificant (Net<Lc), whereas the accumulation (table 6) highlights a proven radioactivity (with a risk error still lower than the risk β)

	Phase 1			*Phase 2*		*Phase3*			
	BkG_i	$Lc_{Net\,i}$	$LD_{Net\,i}$	$Gross_i$	Net_i	*Net> L*	*Resu*	*Expande uncertainty (*	*Relative uncertain*
meas.	119	30	64	122	3	***no***	< LD	/	/
meas.	119	30	64	136	17	***no***	< LD	/	/
meas.	126	31	66	137	11	***no***	< LD	/	/
meas.	144	33	70	139	-5	***no***	< LD	/	/
meas.	131	32	67	151	20	***no***	< LD	/	/
meas.	119	30	64	145	26	***no***	< LD	/	/
meas.	127	31	66	130	3	***no***	< LD	/	/
meas.	113	29	63	125	12	***no***	< LD	/	/
meas.	127	31	66	145	18	***no***	< LD	/	/
meas.	111	29	62	140	29	***no***	< LD	/	/

Table 5: example with 10 negative individual results (<Lc)

	Phase 1			*Phase 2*		*Phase3*			
	$\overline{BkG}_n$	$Lc_{\overline{Net}_n}$	$LD_{\overline{Net}_n}$	$\overline{Gross}_n$	$\overline{Net}_n$	*Net>*	*Resu*	*Expande uncertainty (*	*Relative uncertain*
Accumulation average	123.	9.7	20.7	137	13.	yes	13.	10	76%

Table 6: accumulation by average. Positive end result (> Lc)

These results were obtained by simulation with a true value $\mu_{Net} = 18$. It shows that 10 negative results are not a final proof of an absence of radioactivity. The "average" of ten "non" isn't "none".

Note 1: A common mistake would be to declare that ten nonsignificant measurements can lead only to one nonsignificant accumulation. If in addition this detection limit is wrongly calculated like the average of the individual limits, the results of table 6 would lead to a final assessment $"\mu_{Net} < 65"$ *instead of* $\hat{\mu}_{Net} = 13 \pm 10$.

Note 2: an indication showing simply that the results of table 5 give an hint that there is "something" is that when one repeats measurements of a nonradioactive sample, then the net values are on average negative 50% of time. However, here nine results out of ten are positive, which allows us to think that the probability that the sample could be free of radioactivity is weak.

The detection limit associated to $Lc_{\overline{Net}_n}$ is calculated according to a similar formula:

$$LD_{\overline{Net}_n} = \frac{1}{n}\sqrt{\sum_{i=1}^{n} LD^2_{Net\,i}}$$

It's easy to see that the detection limit of the mean value decrease approximately as the inverse of the square root of the number of individual measurements, just like the decision threshold ($Lc_{\overline{Net}_n}$).

2.5 Ideal situations of application of the accumulation of decision threshold and opposite case

The rules stated above are applicable as soon as the mean of measurements values have a physical or statistical sense. On the other hand, obtaining such spectacular results occurs only when various measurements can be regarded as achievements of random variables of homogeneous parameters.

This condition is observed in the case of the repetition of measurements of the same sample (case n°1 and 2). If the samples are different (case n°3), picked up in different point or different dates, the interest of the accumulation appears when these samples remain homogeneous and representative of similar average conditions. Thus we see that the preceding example obtained by simulation corresponds if the results obey the same real mean value μ_{Net}=18.

When this condition is not observed, the averaging process can make a very significant individual measurement disappear. Such a situation can appear when the searched activity is present only in one sample, which can be the case in the search for a point source (“hot spot”) or in the case of periodic samplings, with an accidental and isolated contamination in time. In such a scenario, it is hardly beneficial to carry out an accumulation of measurements: the contaminated sample can then be measured to a significant degree positively, whereas the average result, extended to the whole of the other uncontaminated samples, can appear overall negative. The “accidental” (outlier) value disappears in the averaging process.

It is good to keep that in mind when using statistical tools. They are often very relevant with values resulting from homogeneous population, but often lose their relevance when dealing with very heterogeneous populations.

3 ACCUMULATION ON ENVIRONMENTAL MONITORING SAMPLES (CASE N°3)

If one considers n samples of rainwater collected periodically in one geographical place, it is possible to cumulate this information in order to get “the average content of a radionuclide given in a geographical area and over one specified period”. Each sample can be regarded as part of a unit made up of the sum of all the samples. The measurand of interest is then either the sum of the activities of all samples, or their average value. The first quantity is relevant when one is interested in assessments of releases, the second is more relevant if one is interested in the mean radioactivity of the environment.

It is important to note an important difference with the preceding cases. Here each sample is likely to contain a partial true activity $\tilde{A}_i$ significantly different from one sample to another, whereas in the two preceding cases this true value was the same one for all measurements.This will impact the calculation of the detection limit, the cumulated decision threshold being insensitive to this potential dispersion of the true values. We exclude here the case of an accidental and massive punctual contamination, as discussed in chapter 2.5.

Numerical application (*table 9*) with sixteen periodic samples for tritium measurement in rainwater. Counting time of measurements is 200 min.

	Decision threshold an detection limi calculation					Samples measurements v			Testing values and expressed individual r			
	Bk	**Lc N**	**ε (%**	**Lc (B**	**LD (B**	**Gr**	**N**	**Measur activity (B**	**Net >**	**Declarat (Bq/L**	**Uncertai (Bq/L)**	**Rel. un**
Samp. 1	10	8	21%	3.4	7.0	10	4	0.16	*NO*	*< LD*	/	/
Samp. 2	85	81	30%	2.2	4.5	89	3	0.90	*NO*	*< LD*	/	/
Samp. 3	82	80	31%	2.1	4.4	98	1	4.38	*YE*	**4.4 Bq/**	***+/- 2.3 Bq***	51%
Samp. 4	81	79	26%	2.4	5.1	79	-1	-0.60	*NO*	*< LD*	/	/
Samp. 5	90	83	31%	2.2	4.6	96	6	1.73	*NO*	*< LD*	/	/
Samp. 6	72	75	26%	2.3	4.8	77	5	1.69	*NO*	*< LD*	/	/
Samp. 7	78	78	26%	2.4	5.0	95	1	5.32	*YE*	**5.3 Bq/**	***+/- 2.6 Bq***	49%
Samp. 8	10	90	30%	2.4	5.0	11	5	1.62	*NO*	*< LD*	/	/
Samp. 9	73	75	26%	2.3	4.8	88	1	4.63	*YE*	**4.6 Bq/**	***+/- 2.5 Bq***	54%
Samp.10	10	88	31%	2.3	4.8	10	-	-0.05	*NO*	*< LD*	/	/
Samp.11	89	83	30%	2.3	4.7	98	9	2.50	*YE*	**2.5 Bq/**	***+/- 2.4 Bq***	95%
Samp.12	80	79	26%	2.4	5.1	85	5	1.65	*NO*	*< LD*	/	/
Samp.13	77	77	26%	2.5	5.1	79	2	0.81	*NO*	*< LD*	/	/
Samp.14	93	85	28%	2.5	5.2	10	6	1.90	*NO*	*< LD*	/	/

Samp.15	82	79	309	2.2	4.5	92	10	2.92	***YE***	**2.9 Bq/**	**+/- *2.3 Bq***	79%
Samp.16	81	79	279	2.4	5.0	90	8	2.72	***YE***	**2.7 Bq/**	**+/- *2.5 Bq***	92%
Accumulation by weighted ave				**0.6**	**1.2**			**2.07**	***YE***	**2.07 Bq**	**+/-0.6 Bq**	**30%**

Table 7: periodic measurements of tritium in rainwater

The accumulation is done here by weighted average. One notes a relative variability $\sigma_{rel.\varepsilon}$ of efficiencies about 10% around an average efficiency equal to 28%. The calculation of the weighted average activity takes into account (rule n°1) the 16 results, including “negative activities”. These should not be rejected although a true activity can be only positive. Indeed a negative single measurement cannot be regarded as an estimator of the true value, but has still real informational contents. In practices it is important to record the values of measurements wether positive or negative, and not to erase them by replacing them purely and simply by the indication “< LD”.

This is a recommendation made by several scientific authorities like Royal Society off Chemistry [5] or the Federal agency of Environmental protection of the United States [6] We can quote the MARLAP here: “ ***The laboratory should report each measurement result and its uncertainty as obtained (as recommended in Chapter 19) even if the result is less than zero”***”. This does not mean that it is necessary to have all the of measurement data (values of raw count, background noise, net counting value, etc). In fact only the values of the measured activity and possibly the dispersion of the efficiencies are necessary. It is perfectly conceivable to restore this result by specifying that it is lower than the detection limit but by also giving the value measured like its uncertainty.

As comparison, the fact of omitting the nonsignificant results leads to an average of 3,73. Thus compared to the result calculated by taking account of all the values one introduces here a skew of almost 100%. This is not a simple calculative abstraction but corresponds to the physical case where an operator would mix these 16 samples collected throughout the year and would wish to compare the result of measurement of the mixture with those of measurements of each samples.

It is thus possible to rigorously determine the annual mean value of the activity measured in these samples. If one admits the validity of each individual measurement, this operation of accumulation keeps any validity in so far as it has a sense. It is obvious that an accumulation of measurements of samples taken in various points at various times would not have any sense if the samples were not representative of a single measurand defined without ambiguity.

Decision threshold on the weighted average activity:
As in the case of the gamma spectrometry the decision threshold associated with the weighted average activity is calculated like :

$$Lc\left(\hat{A}_{weigh.}\right) = \sqrt{\frac{1}{\sum_{i=1}^{n}\frac{1}{Lc_i^2}}}\ .$$

One obtains a decision threshold on the average activity equal to $0{,}60\ Bq.l^{-1}$, whereas the individual decision thresholds are about $2{,}45\ Bq.l^{-1}$. The gain there still is about $\sqrt{16}$.

Note: if the measurand of interest is the sum of the activities $\hat{A}_{\Sigma} = \sum_{i=1}^{n}\hat{A}_{i.}$, instance in the annual statement of discharge calculated starting from the monthly discharges, then the application of the rule n° 2 is commonplace here and makes it possible to write:

- Associated decision threshold: $Lc\left(\sum_{i=1}^{n}\hat{A}_{i.}\right)=\sqrt{\sum_{i=1}^{n}Lc_{A\,i}^{2}}$
- Limit of associated detection: $LD\left(\sum_{i=1}^{n}\hat{A}_{i.}\right)=\sqrt{\sum_{i=1}^{n}LD_{A\,i}^{2}}$

Detection limit associated to the estimation by weighted average:

The dispersion of the efficiencies characterized by the relative standard deviation of the efficiencies discussed in Section 3 ($\sigma_{rel.\varepsilon}$) introduce a corrective term tending to increase the limit of detection. The latter is calculated according to the formula:

$$LD\left(\hat{A}_{pond.}\right)=\frac{1}{1-k_{1-\beta}^{2}.\sigma_{rel.\varepsilon}^{2}}.\sqrt{\frac{1}{\sum_{i=1}^{n}\frac{1}{LD_{i}^{2}}}}.$$

This expression is similar to the expression of the detection limit in the standard ISO 11929 [7]. The corrective term $1/\left[1-k_{1-\beta}^{2}.\sigma_{rel.\varepsilon}^{2}\right]$ is easily negligible when the efficiencies are little dispersed. For a beta risk equal to 2,5% the corrective term remains lower than 1,1 when the relative variability on the efficiencies remains lower than 15%. It is necessary to have a relative variability of 36% so that this term of correction is equal to 2.

4 CONCLUSION

The implementation of these methods, through the rules n°1 and n°2, can make it possible to lower considerably the decision thresholds and the detection limits in many situations: assessment of rejections, environmental measurements, waste etc. Generally one can expect to decrease the decision thresholds and the detection limits by a factor of $\sqrt{n}$, when the number of cumulatable measurements is equal to n. These methods also make it possible to be freed from heavy statistical treatments or strongly skewed by taking into account rigorously the whole of information given by measurements. This rigor of treatment also gives the possibility of determining if the whole of the results give significant information on the global presence or not of an element. It is important to understand the measurement system and the data collected so that the appropriate cumulative method is applied to the whole measurements. If you improve your decision threshold that means that you have first understood the way your measurement works, and secondly you have improved it.

The application of these new methods by the Service of study and monitoring of the radioactivity in the environment of IRSN, which manages annually about twenty thousand samples on the national territory, which give approximately about thirty thousand analyzes, should bring notable improvements in field of environment survey.

References:

[1] Fiévet B., Della Vedova C. , (2010) Dealing with non-detect values in time-series measurements of radionuclide concentration in the marine environment, *J. Environ. Radioact.* **101(1):**1-7.
[2] Helsel D.R., 2005. Nondetects and Data Analysis: Statistics for Censored Environmental Data. John Wiley and Sons, New York, p. 250.
[3] Vivier A., Aupiais J.,(2007) *J. Radiochim. Acta* **95**, 477-492
[4] Vivier A., Fottorino R., Rousse B., (2010) Seuil de décision et limite de détection : estimation interprétation et optimisation, 1ère partie : principes de bases, *Radioprotection* **45**, 321-343
[5] Multi-Agency radiological laboratory Analytical protocols manual (MARLAP), vol III EPA 402-B-04-001C, chapter 20, US Environmental Protection Agency
[6] AMC. *What should be done with results below the detection limit? Mentioning the unmentionable.*
Analytical methods committee technical brief no. 5, April 2001. Royal Society of Chemistry, London, 2001, Two unnumbered pages.
[7] ISO 11929 (2009) Determination of the characteristic limits (decision threshold, detection limit and limit of the confidence interval) for measurements of ionizing radiations