

## Chapter 16

# Evaporation Losses of Organic Samples from Liquid Scintillation Counting Vials

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## INTRODUCTION

There is a wide variety of liquid scintillation counting vials at present in use and their characteristics have been reviewed both in this country and abroad.<sup>1,2</sup> The choice of vial for a particular application is dependent on factors such as background count rate, efficiency and cost. One area which has not been subject to much investigation is the rate of loss of material from the vials during the period for which the sample is counted. In many applications this is unimportant as counting times are limited to a few minutes or perhaps hours. However, in low level applications, counting may extend over much longer periods and, moreover, standards must be kept, semi-permanently, in vials identical with sample vials. This is certainly true of the British Museum Radiocarbon Dating Laboratory where typical samples may be counted intermittently over a period of several weeks and standards may be kept for a year or more. Detailed descriptions of our methods have been given elsewhere.<sup>3,4</sup>

With this in mind we embarked on a series of experiments to evaluate three vial designs (Fig. 1) which have each been used for low level  $^{14}\text{C}$  measurements. Design A, in everyday use at the British Museum Laboratory, is the standard 20 ml, low-potassium borosilicate glass vial as supplied by several manufacturers. Design B, developed by the Low Level Measurements Laboratory, Harwell, uses the standard vial but a different closure described by Otlet and Slade<sup>5</sup> of Harwell in these words: 'A good seal is now achieved with an indium foil washer (0.008 cm) held firmly to the specially ground vial top by a rubber washer (0.165 cm) used in place of the usual cork/aluminium pressure pad. A Teflon washer (0.026 cm) in between prevents damage to the indium when tightening the screw cap.' Finally, design C is a Teflon vial sealed with a Viton 'O' ring held in an aluminium cap and was developed in Australia by Calf and Polach<sup>6</sup> specifically for low activity  $^{14}\text{C}$  samples.

## EXPERIMENTAL WORK

As a pilot experiment, six standard vials (design A) were taken and filled with 5 ml of benzene and 10 ml of toluene. These quantities were chosen to approximate

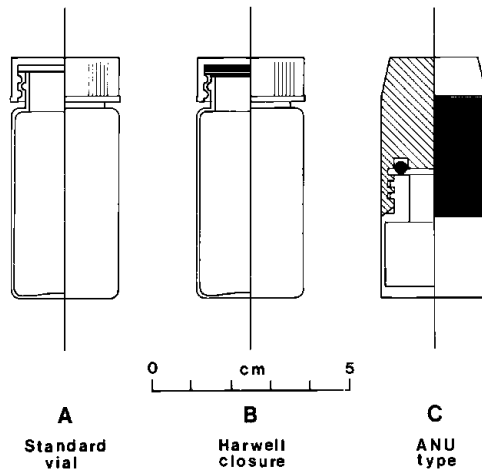


Fig. 1. Counting vials used for low level  $^{14}\text{C}$  measurements. (Copyright: British Museum.)

the counting cocktail in use at the British Museum Laboratory and were adhered to in all subsequent evaporation measurements. Five vials were cooled to  $0^\circ\text{C}$  by placing in the sample changer of the laboratory's Packard 3315 counter and the sixth was left in the open laboratory at about  $20^\circ\text{C}$ . They were then weighed at intervals and the results were as summarised in Tables 1(a) and 1(b). These indicated that solution loss was occurring at a rate of about 0.5 mg per day at  $0^\circ\text{C}$  which if left uncorrected in a sample counted for one month would in a typical case give rise to an extra

Table 1(a). Cap weight increase (mg).

	Days				
	4	12	35	104	769
Mean ( $0^\circ\text{C}$ )	12.1	22.2	45.0	52.9	72.9
S.D.	1.2	2.0	9.1	4.1	6.9
Mean ( $20^\circ\text{C}$ )	2.2	3.2	5.0	16.7	69.0

Table 1(b). Solution loss (mg).

	Days				
	4	12	35	104	769
Mean ( $0^\circ\text{C}$ )	2.9	13.7	24.7	45.4	159.7
S.D.	0.4	6.9	10.6	22.0	33.5
Mean ( $20^\circ\text{C}$ )	9.4	20.1	57.2	147.6	1051.1

Table 2(a). Cap weight increase (mg).

	Days												
	2	3.8	8.7	14.8	20.7	36	39	42	52.8	70	72	77	79
Mean (0°C)	2.5	4.2	9.3	19.2	17.9	24.3	27.4	26.2	29.0	34.3	34.6	34.7	34.6
S.D.	2.0	2.3	2.9	3.5	3.2	3.5	6.8	4.0	3.5	3.3	3.3	3.2	3.2
Mean (20°C)	2.2	5.0	2.8	0.2	-3.1	-0.5	4.9	3.5	2.6	14.7	12.9	10.7	9.3
S.D.	0.7	4.7	0.6	0.4	0.3	1.5	9.7	1.3	0.4	4.1	0.5	0.4	0.3

Table 2 (b). Solution loss (mg).

	Days												
	2	3.8	8.7	14.8	20.7	36	39	42	52.8	70	72	77	79
Mean (0°C)	7.9	9.1	11.4	23.0	28.4	32.6	35.9	37.7	51.2	60.0	61.0	62.1	64.6
S.D.	8.2	8.4	8.3	15.3	19.2	20.3	17.8	16.5	17.7	19.3	18.8	19.0	19.4
Mean (20°C)	10.6	19.2	33.4	41.1	47.8	67.9	78.7	86.8	103.0	126.0	135.0	144.0	147.0
S.D.	16.3	22.8	23.4	25.6	27.5	31.6	25.2	23.3	24.4	33.9	30.9	31.6	31.8

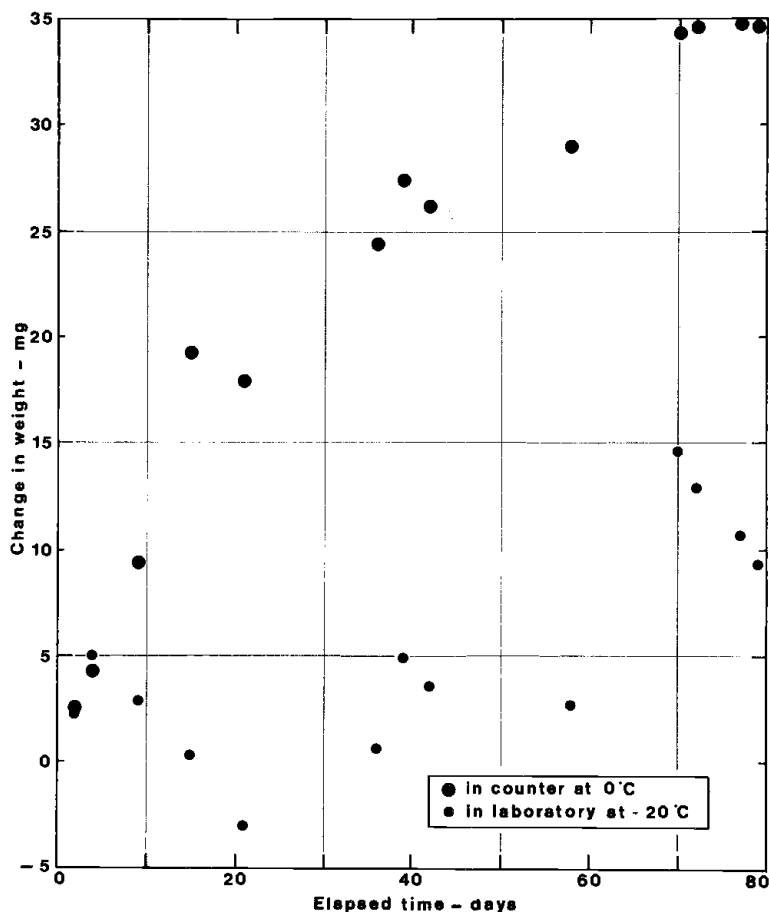


Fig. 2. Change in weight of standard vial caps. (Copyright: British Museum.)

radiocarbon date measurement error of about 10 years. However, two other features were also apparent: (i) the standard deviation of the amount of solution loss increased with time, indicating that some vials consistently lost more material than others; (ii) the vial caps increased in weight during the course of the experiment and it is conceivable that a substantial proportion of the material lost from the solution was being absorbed by the cap, although the rate of increase of the cap weight was not maintained for the whole of the experiment.

To investigate this more thoroughly the experiment was repeated with ten samples, five maintained at 0°C and five at 20°C, and more frequent weighings. The results are presented in Tables 2(a) and 2(b) and Figs. 2 and 3 and it is clear that for the samples at 20°C, solution loss is considerably greater than the cap weight increase. Preliminary results of experiments at present in progress indicate that the increase in cap weight for samples at 0°C is caused by the high humidity in the Packard counter. In the early part of this experiment, several vials lost weight at a rate substantially in excess of the trend before settling down. This appeared to be due to a phenomenon which Otlet<sup>7</sup> has christened 'ratcheting', where the vial cap eases open sometime after it is originally tightened.

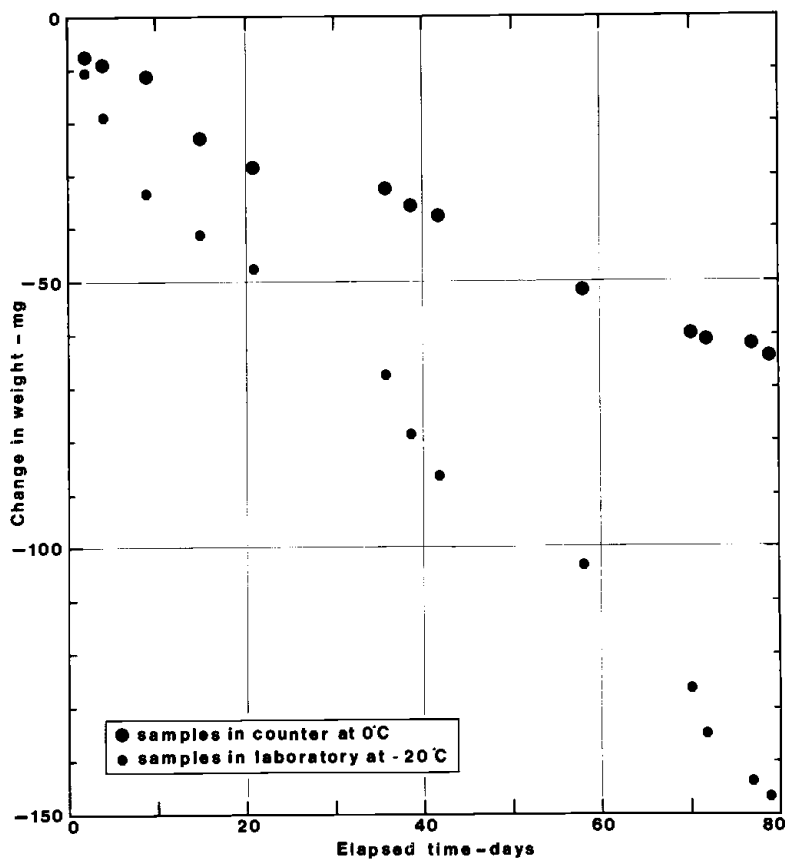


Fig. 3. Sample weight loss from standard vials. (Copyright: British Museum.)

Next the vial caps from the samples maintained at 0°C were removed and allowed to stand in the laboratory for several weeks, their weights being remeasured at intervals. The results (Table 3) showed that the caps lost about 50 mg in 3 weeks and then hovered around their new weight.

Finally, five each of designs A and B, and the single example of design C available to us, were each filled, sealed and left in the counter for 28 days. On reweighing (Table 4) the Harwell-developed design B proved to have performed best with a mean loss of 0.7 mg which was substantially better than either of its rivals. The result for design C was disappointing, particularly in view of the claims of Calf and Polach that

Table 3. Cap weight loss in laboratory (mg).

	Hours								
	22.6	65	142	185	331	472	591	723	840
Mean	13.6	22.4	28.7	34.1	42.7	48.8	47.6	35.9	49.4
S.D.	3.9	3.9	4.8	4.5	4.5	5.0	5.0	7.2	4.9

Table 4. Solution loss (mg).

	Design		
	A	B	C
Mean	11.3	0.7	13.5
S.D.	2.9	0.6	—

the loss rate should be 0.5 mg per week at 18 °C. It was noticeable that the 'O' ring increased in weight by 7 mg during the experiment and the solid aluminium cap by 3 mg. This is consistent with the results for an adapted cap produced in the British Museum Laboratory which was sealed by a Viton 'O' ring held in an aluminium plug. The leakage rate for this design proved to be unacceptable and incidentally the 'O' ring and the aluminium plug were observed to gain weight in a similar manner to design C.

## CONCLUSIONS

The sealing qualities of the standard vials (design A) are not entirely satisfactory over long periods due to variable loss rates and the risk of occasional more severe leakage. The overall weight of a sample sealed in a vial is an unreliable guide to the leakage rate as the cap weight changes with environment. Design B, although a somewhat more cumbersome system, provided a highly effective seal but it is possible that the indium itself contributes to the background count rate.<sup>a</sup> Possibly the best arrangement for low level <sup>14</sup>C counting would be a Teflon vial with its advantages of low background and high counting efficiency sealed in a manner similar to the Harwell design. It is our intention to produce and test such a vial in the near future.

## ACKNOWLEDGEMENTS

We would like to thank Mr. R. L. Otlet of AERE Harwell for useful discussions and for kindly allowing us to use his vial sealing system in our experiments.

## REFERENCES

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5. R. L. Otlet and B. S. Slade, *Radiocarbon* 16(2), 178 (1974).

<sup>a</sup> <sup>115</sup>In, 96% natural abundance, 6 × 10<sup>4</sup> years half-life, emits β-particles with an energy of 0.6 MeV.

6. G. E. Calf and H. A. Polach, in *Liquid Scintillation Counting – Recent Developments* (eds. P. E. Stanley and B. A. Scoggins), Academic Press, London, 1974, p. 223.
7. R. L. Otlet, personal communication.

## DISCUSSION

**C. McEvoy:** At the A.R.C. Institute in which I work, we have completely sealed glass vials in which we wish to keep standards; thus we have no diffusion loss as the entire vial is sealed glass. Would this be a better way of sealing your samples and keeping your standards?

**A. Hewson:** These would not be suitable for our particular application as the vials used are selected for their low background and are re-used.

**D. I. Chapman:** You said that a loss of 50 mg from vials in 100 days was equivalent to an error of 10 years in 5,000 years in your dating procedures. Since you consider it important to reduce this loss, are you therefore claiming an accuracy of 10 years when dating a sample about 5,000 years old?

**A. Hewson:** No, this is a 10 year systematic error in addition to the random measurement error of typically  $\pm 40$  years.