

# Comparison of analysis techniques for the determination of Carbon-14 in solid materials

## Introduction

- Carbon-14 ( $^{14}\text{C}$ ) is a pure beta emitter with a energy of 156.5keV and half life of 5730 years.
- Present in the environment naturally and as a result of weapons fallout.
- Analysis is generally performed using either wet oxidation or combustion furnace.
- Final activity determination using liquid scintillation counting.

## Aim

- Determine whether there was any statistical difference between techniques.
- Assess levels of precision, sensitivity and limits of detection.
- Identify the scientific and practical advantages and disadvantages.
- Assess suitability of IAEA reference materials for low level  $^{14}\text{C}$  analysis.

## Exercise outline

5g of the following materials were sent to 7 radiochemistry laboratories in the UK with 8 returning results.

- IAEA C2 (Travertine) Expected activity =  $11.2 \text{ Bq kg}^{-1}$
- IAEA C6 (Sucrose) Expected activity =  $143 \text{ Bq kg}^{-1}$
- IAEA C7 (Oxalic acid) Expected activity =  $21.3 \text{ Bq kg}^{-1}$
- Cefas QA material (Biot) Expected activity =  $377 \text{ Bq kg}^{-1}$

Sample homogeneity was assessed for IAEA C2 and C6 by measuring the  $^{13}\text{C}$  activity. The results were within 5% of the expected value with a precision of 2%.

## Methodology outline

Two main methods were used:

### Combustion furnace

Sample is burnt in an oxygen rich atmosphere in the presence of a catalyst. The carbon species is converted to  $\text{CO}_2$  and selectively trapped in a series of gas-bubblers containing trapping media which are measured by liquid scintillation counting.



### Wet Oxidation

The analytical procedure consists essentially of conversion of all carbon in the sample to  $\text{CO}_2$  by wet oxidation in a sulphuric/chromic acid mixture, collection of the  $\text{CO}_2$  in sodium hydroxide solution, precipitation and gravimetric determination of carbon as  $\text{CaCO}_3$ , gel scintillation counting for  $^{14}\text{C}$  determination.



## Results

Table 1: Lab method details summary.

Lab	Technique	Catalyst	Counter	Uncertainty	LoD ( $\text{Bq kg}^{-1}$ )
1	Combustion-Instron single	Pt	Packard 2900TR	total	72-135
3*	Combustion-Instron single	Pt	Packard Tricarb 3170 TR/SL	counting	N/A
4	Wet oxidation	N/A	Wallac LKB 1217 Rackbeta	total	33
5	Combustion-Instron single	Pt	Wallac 1415	total	22
6	Radtec Pyrolyser-6 Trio	Pt	Quantulus	total	20
7	Combustion-Instron single & 6 burner	Pt	Quantulus	total	20-25

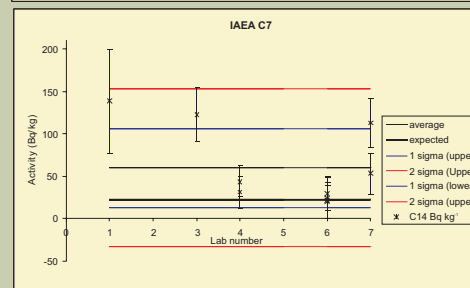
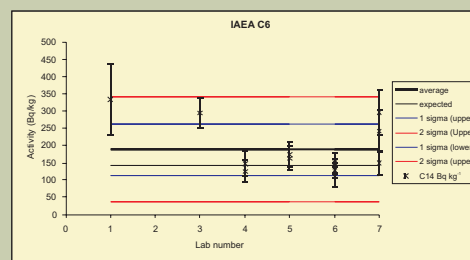
\*The method at lab 3 is currently under development

### IAEA C2 ( $11.2 \text{ Bq kg}^{-1}$ )

- One positive result: Lab 3 ( $180 \pm 40 \text{ Bq kg}^{-1}$ ).
- Remaining labs using combustion =  $< 20 \text{ Bq kg}^{-1}$  except Lab 1 ( $< 70 \text{ Bq kg}^{-1}$  -instrumental issue).
- Wet oxidation technique =  $< 33 \text{ Bq kg}^{-1}$

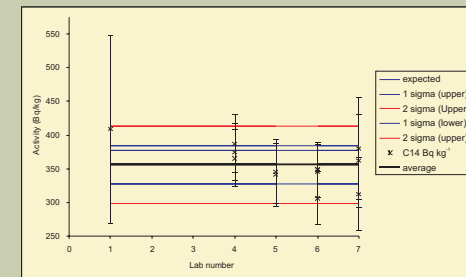
### IAEA C6 ( $143 \text{ Bq kg}^{-1}$ ) & IAEA C7 ( $21.3 \text{ Bq kg}^{-1}$ )

- High level of variation in results.
- Two sets of data for each material i.e. C6 one set at  $\sim 150 \text{ Bq kg}^{-1}$  and another at  $\sim 300 \text{ Bq kg}^{-1}$  and C7  $\sim 30 \text{ Bq kg}^{-1}$  and  $\sim 120 \text{ Bq kg}^{-1}$
- Variation was not technique dependant as seen when measured at the same lab (lab 7) using 2 different furnace types.
- C7 activity is close to the methods LoD and therefore four results were less than LoD and the rest had a high uncertainty.



### Cefas QA Material ( $377 \text{ Bq kg}^{-1}$ )

- Results in good agreement and not technique or furnace type dependant.
- 20% variation in the result obtained across all results.
- If lab 3's result is classed as an outlier the variation decreases to  $\sim 8\%$ .



## Combustion versus wet oxidation?

- Combustion furnaces can take samples up to 20g whereas wet oxidation is dependant on the carbon content of the sample.
- For both techniques, at low activity levels, the blank is critical - an increase of 2 cpm can lead to an activity difference of  $10\text{-}20 \text{ Bq kg}^{-1}$
- Wet oxidative sample chemistry is a much quicker method.
- Both techniques have the ability to analysis OBT and  $^{14}\text{C}$  however the combustion technique can analyse simultaneously.

## Summary

- LoD by combustion for  $\sim 1.5\text{g}$  sample is typically  $20\text{-}25 \text{ Bq kg}^{-1}$
- LoD by wet oxidation for  $\sim 1.5\text{g}$  sample is typically  $33 \text{ Bq kg}^{-1}$
- LoD's are highly dependant on the LS counter used.
- Wet oxidation is more sample efficient i.e.  $\sim 100\%$  is used for final measurement compared to 30-50% for combustion.
- High variation in results for C6 and C7 for the combustion methods.
- Two techniques, wet oxidation and combustion, produced similar activity results.
- Excellent replication in results for the Cefas QA material.
- Lack of suitable certified reference materials at environmental levels. Therefore Cefas biota sample was the only viable QA material.
- Both techniques, from the results of this study, appear to be capable of successfully analysing  $^{14}\text{C}$  in environmental samples.

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