LSUF 2004

Update on Organically Bound Tritium Standard project

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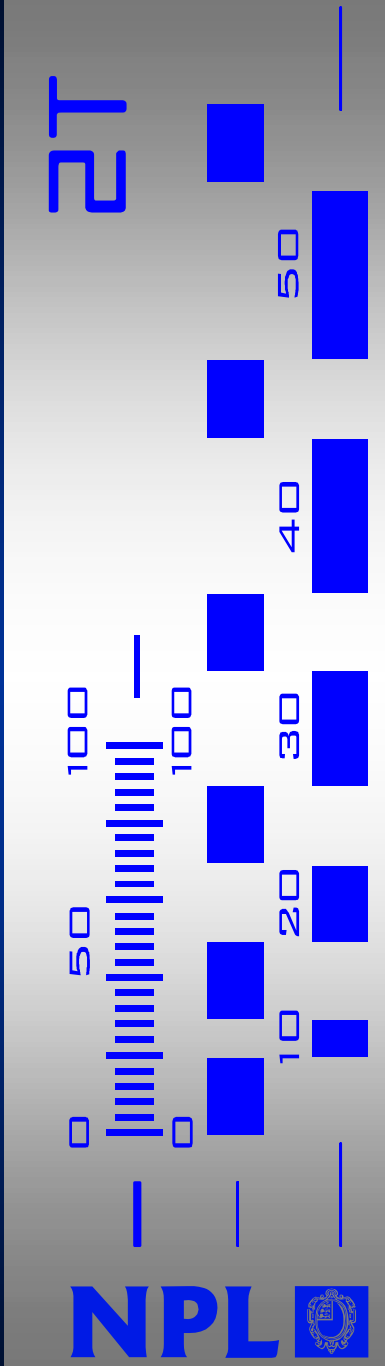
5 May 2004



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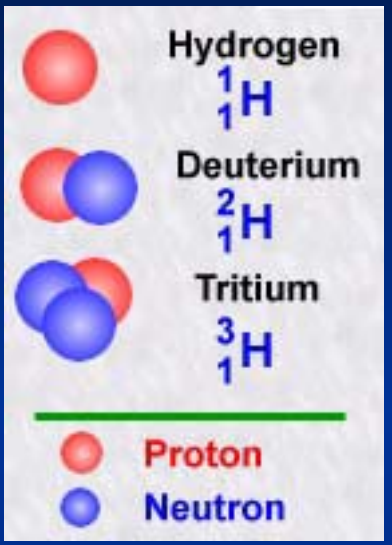
What is tritium?

Tritium is a hydrogen isotope: ^{3}H or T

Half-life: 12.33 years

Emits ß⁻ particles with a maximum energy of 18.57 keV



Produced by
i) Nuclear reactions caused by cosmic rays
ii) Nuclear bomb tests
iii) Nuclear facilities

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How much tritium in the environment?

Global “natural” tritium inventory: about 1.3 x 10^{18} Bq (i.e., 3.6 kg)

Release of nuclear power stations: 2 x 10^{16} Bq y^{-1} (about a third of the “natural” production)

Nuclear bomb tests release (1952 to 1962): ~0.3 x 10^{20} Bq

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What is organically bound tritium (OBT)?

Chemical forms of tritium in the environment: HTO (water), HT (hydrogen gas) and organically bound tritium (OBT)

OBT is tritium bound to carbon

C-H bond is (in general) more inert than H-H, O-H, N-H and S-H bonds

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Tritium in the environment

Most tritium in the environment is HTO

OBT has a much higher accumulation factor (i.e., 20,000) in marine species than HTO (i.e., 1)

Tritium in Bristol Channel:	[Bq kg^{-1}]
Seawater	~10
Fish (dry weight)	up to 200,000

Dose coefficients (in Sv Bq^{-1}) for OBT are twice that of HTO

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Why is it important?

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Why do we need an OBT standard?

General procedure for OBT in environmental samples

(I) HTO fraction: distillation or freeze-drying of sample
(II) Total tritium: combustion of sample at 750 ºC with catalyst

Both fractions are measured separately

OBT is taken to be the difference between (I) and (II)

An OBT standard is needed to validate the combustion procedure (chemical yield may be less than 100%)

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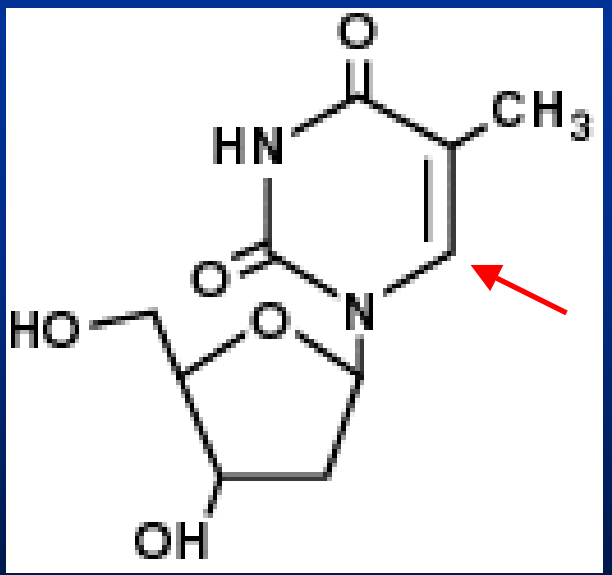
Which OBT standard?

[6-^{3}H]-Thymidine

-Relevant (DNA building block)

-Rather difficult to distill (mp 186 ^{o}C) or combust

-Readily available in tritiated form



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OBT standardisation

OBT standardisation involves

I. Standardisation of activity [Bq g^{-1}]

and

II. Standardisation of a specific chemical form

The OBT standard was standardised with liquid scintillation counting (LSC)

Standard addition method

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Standard addition method (I)

i) Measurement of OBT samples with LSC
ii) Addition of known amounts of standardised HTO
iii) Remeasurement

i) $C_1 = E_1 A$
iii) $C_2 = E_2 (A + B)$, resulting in $A = C_1 B / (C_2 – C_1)$

It is assumed that $E_1 = E_2$

a) No additional quenching occurs due to the addition of HTO
b) Counting efficiency for HTO is identical to that of tritiated thymidine

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Standard addition method (II)

$$A_T = \frac{GDF\ m_H A_H}{m_T} \frac{C_T - C_{BG}}{C_{ST} - C_T}$$

A_T	tritium activity concentration in stock solution [Bq g^{-1}]
GDF	gravimetric dilution factor between stock and working solution
m_H	mass of the HTO spike [g]
A_H	tritium activity concentration in HTO spike [Bq g^{-1}]
m_T	mass of the sample of working solution [g]
C_T	count rate of the sample of working solution [cps]
C_{BG}	count rate of the background [cps]
C_{ST}	count rate of the spiked sample [cps]

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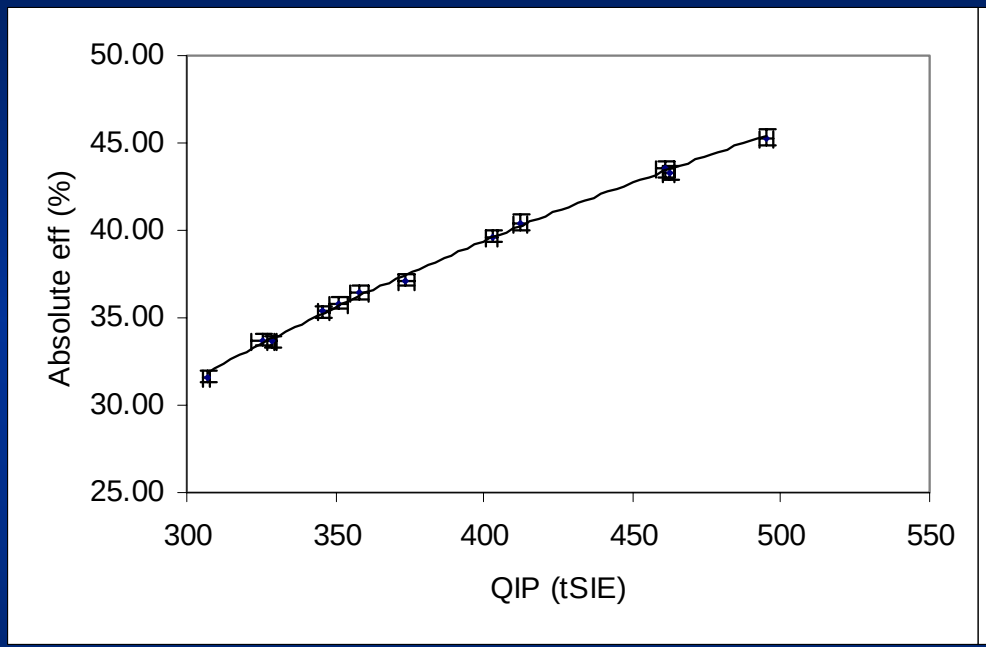
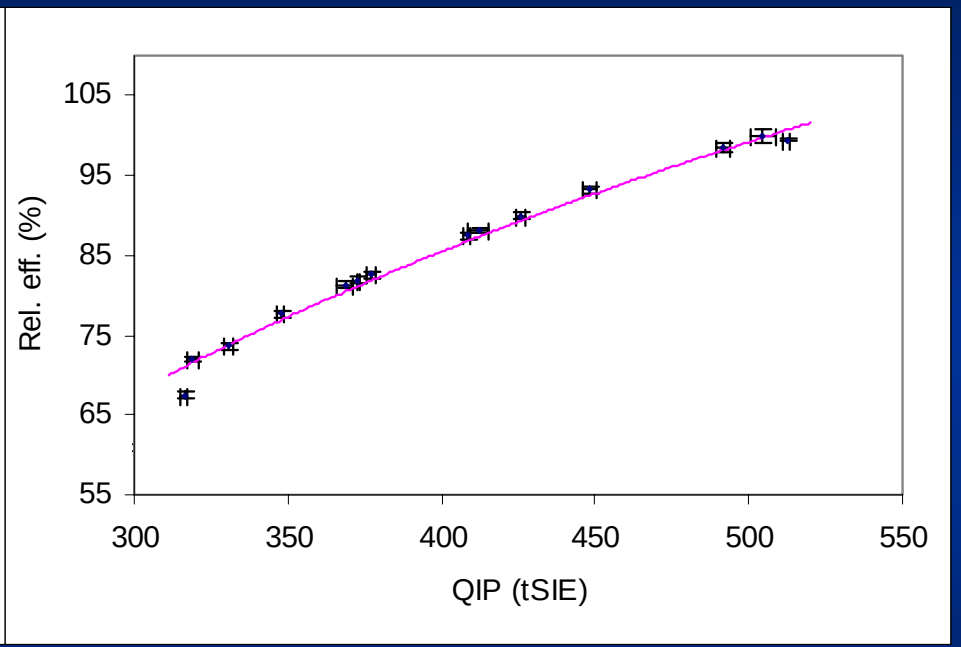
Counting efficiency as a function of quenching



HTO



Tritiated thymidine

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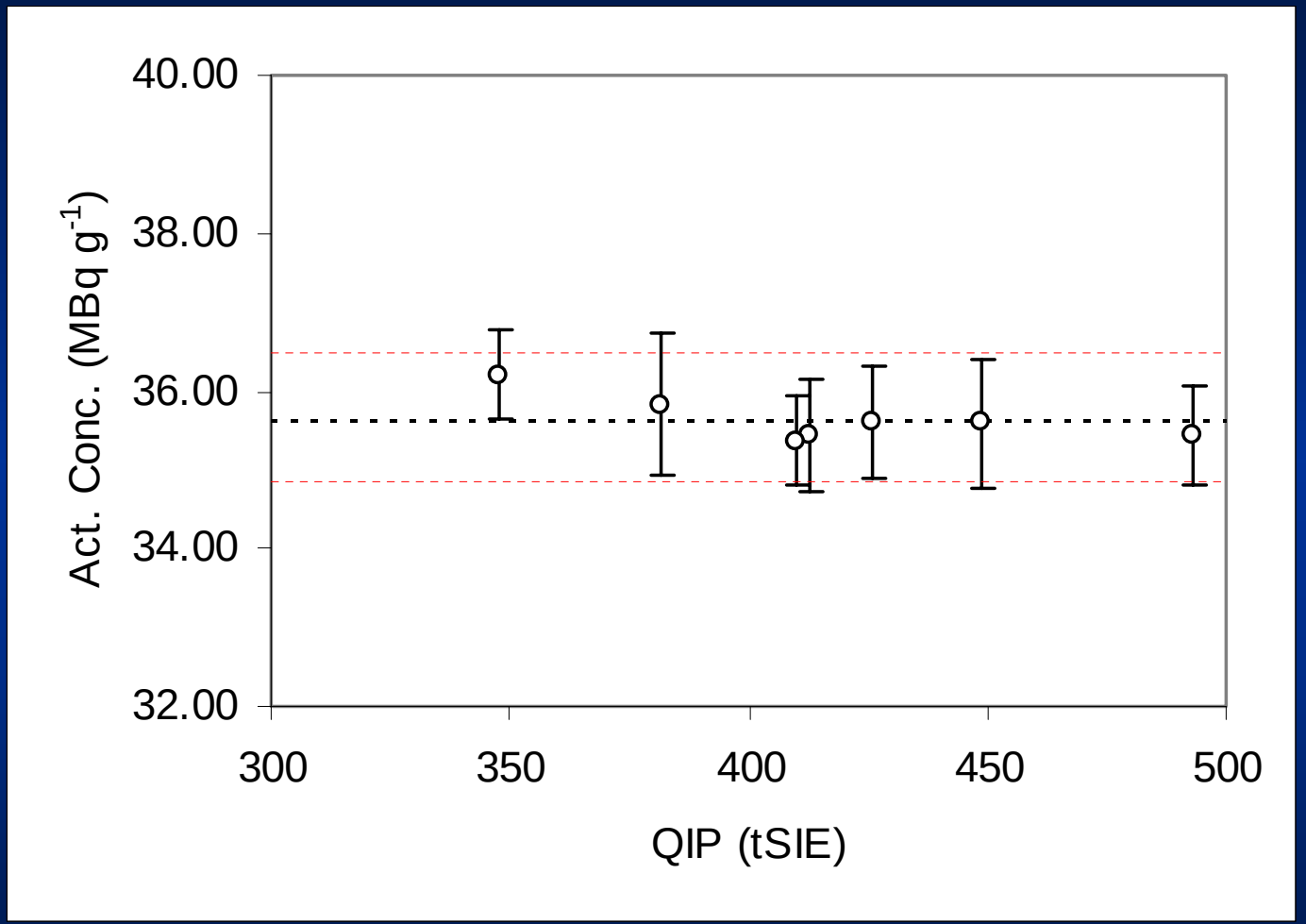
Tritium in tritiated thymidine stock solution



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Stability of OBT

OBT is unstable:

- Nuclear decay
- Direct interaction with emitted beta particles
- Free (hydroxyl) radicals created by radiation
- Chemical or microbiological effects

Formation of OBT fragments or HTO

Self-decomposition rates vary between <0.5% per year and 10% per month (all under recommended storage conditions)

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Separation of OBT and HTO

Separation of thymidine and HTO

-Distillation

-Chromatography

Tritiated thymidine solution was dried in desiccator

Desiccant: dry silica with colour indicator

Solid residue after drying was dissolved and measured; desiccant washed with water and this rinse was measured

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Results

95(1)% (k=1) of tritium in the thymidine fraction

4(1)% (k=1) of tritium in the water fraction

The activity concentration of tritiated thymidine in the tritiated thymidine stock solution was:

33.9 ± 1.1 MBq/g (reference date 1 July 2003; k=2)

Self-decomposition rate: ~0.1% per month of storage at 4 ^{o}C

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Uncertainty components

Component	rel. uncertainty (%)	
-Uncertainty of the weighted mean of seven measurements	0.59	[A]
-Tritium activity concentration in the HTO spike	0.96	[B]
-Counting efficiency	0.2	[B]
-Decay of tritium	0.05	[B]
-Chemical form	1.0	[B]
Combined standard uncertainty	**1.6**	

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Summary

OBT has a much higher accumulation factor in biota than HTO

An OBT standard is needed to validate the analytical procedure

OBT is standardised with LSC using standard addition

OBT standards are unstable (self-decomposition); limited shelf-life

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