A Model for Analyzing Components of Uncertainty Encountered in ^{3}H-Standard Efficiency Tracing in $4\pi\beta$ Liquid Scintillation Counting

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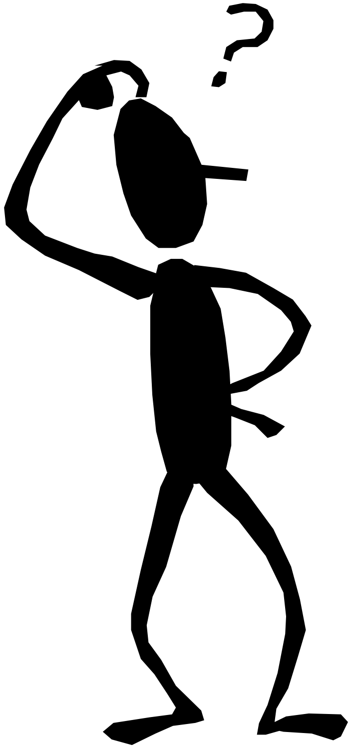
Scope of Presentation

- Goals
  - Introduce ISO philosophy of uncertainty analysis
  - Develop metrological mindset of searching for uncertainty
  - Apply ISO guidelines to development of uncertainty model for LS efficiency tracing techniques
- What is not covered
  - How to analyze all the components, experiment design, etc.
  - What to do with correlations
- Example: Standardization of ^{63}Ni (NIST SRM 4226C

What is a *Measurement Model*?



"Formalization of the relationship between input and output quantities involved in a measurement and how the uncertainty in an input quantity leads to an uncertainty in the output quantity"

ISO Recommendations

- Determine mathematical expression for relationship between measurand Y, from individual input quantities X_i.
  - The input quantities may include variables or uncertainty components that are not explicitly needed to calculate the estimated measurand, y.
  - Example: $R = A\varepsilon d \cdot (xyz\ldots)$
- Determine estimated value of estimated input quantities, x_i, and “standard uncertainties”, u_i.
  - u_i are expressed as estimated standard deviations (or standard deviations of the mean).
  - Uncertainty components assumed to correspond to standard deviations, irrespective of evaluation method. (Type A and B).

ISO Recommendations, Cont’d

- Evaluate covariances.
- Calculate y and “combined standard uncertainty”, u_c (u_i combined in quadrature)
- Form “expanded uncertainty”, $U = k \cdot u_c$, where k is coverage factor. Limits are supposed to correspond to a particular confidence level.
- Report y, u_c *(or U)*, specification of value and assumptions for choice of k, and complete specification of all uncertainty components, u_i.

Simple Case

Real time

Counting interval (livetime)

Total counts

Half-life

"Poisson Counting Error", s_1

Background, s_3

Livetime correction, u_2

Repeatability, s_4

Clock uncertainty, s_7

Uncertainties in corrections, u_5

Half-life uncertainty, s_8

Impurities, u_6

Count rate

$$u_c = \sqrt{\sum_i u_i^2}$$

$$U = k \bullet u_c$$

Component Analysis

- Just because you can think of an effect doesn’t mean that the effect is present
  - Statistical analysis required
  - ANOVA can help
- An effect may be present in one experiment and not in another
- Effects may be embodied in other components

Example: LS Composition Effects

- Prepare two sets of six cocktails, each set having different cocktail composition (aqueous fraction, carrier concentration, etc.)
- Repeat for second radionuclide
- Compare within-measurement and between-measurement variabilities to determine presence of effect.
- ANOVA

Example: Test for Composition Effect

Nuclide X

Trial	C_A,Comp. A	C_A,Comp. B
1	122.6	124.1
2	123.4	124.5
3	123.1	124.6
4	122.8	124.3
5	123.3	124.7
6	123.2	124.2
Mean	123.1	124.4
s	0.3 (0.25 %)	0.2 (0.19 %)

Nuclide Y

Trial	C_A,Comp. A	C_A,Comp. B
1	523.1	523.4
2	520.3	519.8
3	521.6	522.1
4	522.1	523.3
5	522.4	521.2
6	520.5	523.5
Mean	521.7	522.2
s	1.1 (0.21 %)	1.5 (0.28 %)

ANOVA Analysis

Nuclide X

122.6	124.1		Anova: Single Factor						
123.4	124.5								
123.1	124.6		SUMMARY						
122.8	124.3		*Groups*	*Count*	*Sum*	*Average*	*Variance*		
123.3	124.7		Column 1	6	738.4	123.0667	0.094667		
123.2	124.2		Column 2	6	746.4	124.4	0.056		
			ANOVA						
			Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
			Between Groups	5.333333	1	5.333333	70.79646	7.55E-06	4.964591
			Within Groups	0.753333	10	0.075333			
			Total	6.086667	11				

Composition effect present (95 % confidence)

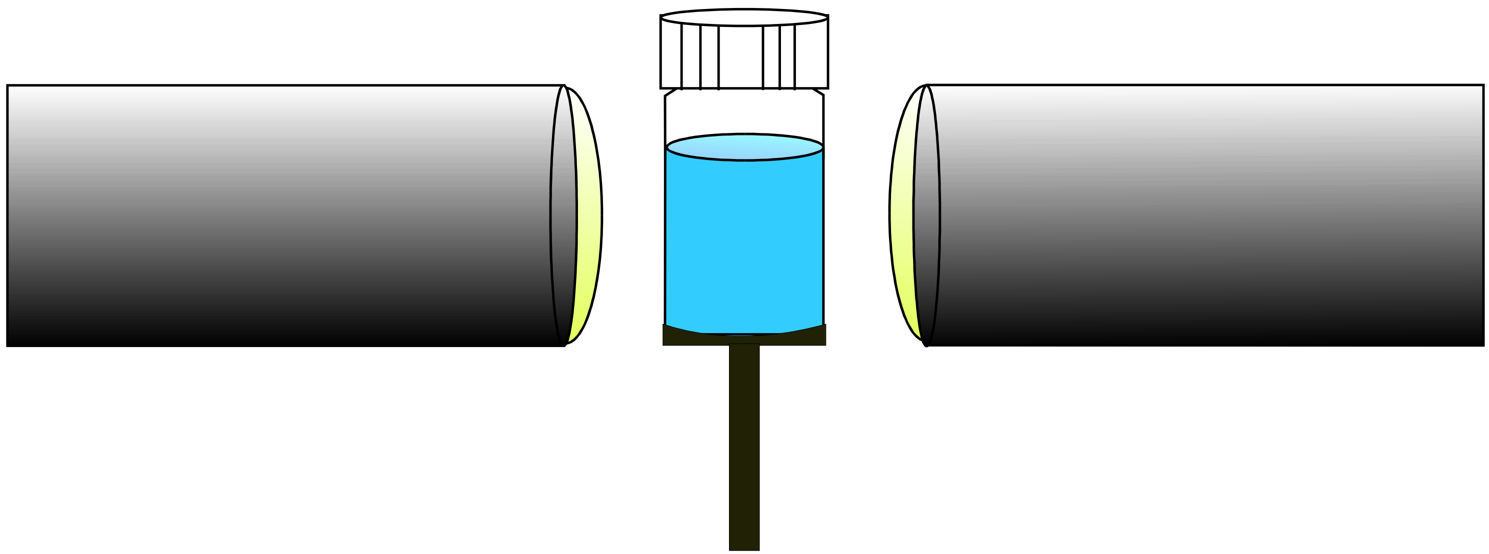
ANOVA Analysis, cont'd

Nuclide Y

523.1	523.4		Anova: Single Factor							
520.3	519.8									
521.6	522.1		SUMMARY							
522.1	523.3		*Groups*	*Count*	*Sum*	*Average*	*Variance*			
522.4	521.2		Column 1	6	3130	521.6667	1.202667			
520.5	523.5		Column 2	6	3133.3	522.2167	2.221667			
			ANOVA							
			Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*	
			Between Groups	0.9075	1	0.9075	0.53003	0.483293	4.964591	
			Within Groups	17.12167	10	1.712167				
			Total	18.02917	11					

No composition effect present (95 % confidence)

Two-Phototube Coincidence Liquid Scintillation Counting with the CIEMAT/NIST Efficiency Tracing Method



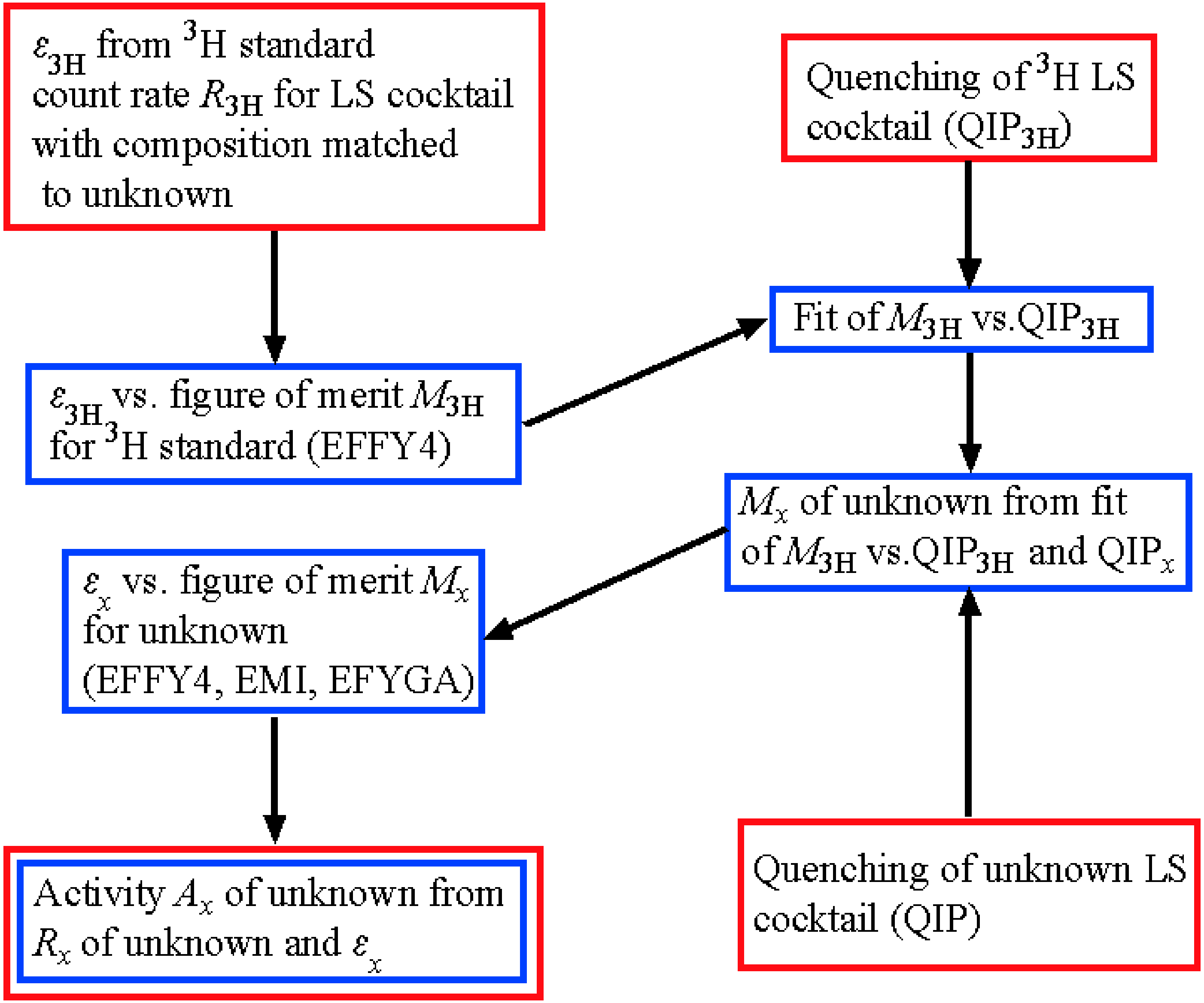
$$\varepsilon = \int_0^{\mathrm{Emax}} \left\{1 - \exp\left[-M^{-1} E \mathrm{Q}(E) \mathrm{W}(E)\right]\right\}^2 \mathrm{P}(Z,E)\,\mathrm{d}E \times \left(\int_0^{\mathrm{Emax}} \mathrm{P}(Z,E)\,\mathrm{d}E \right)^{-1}$$

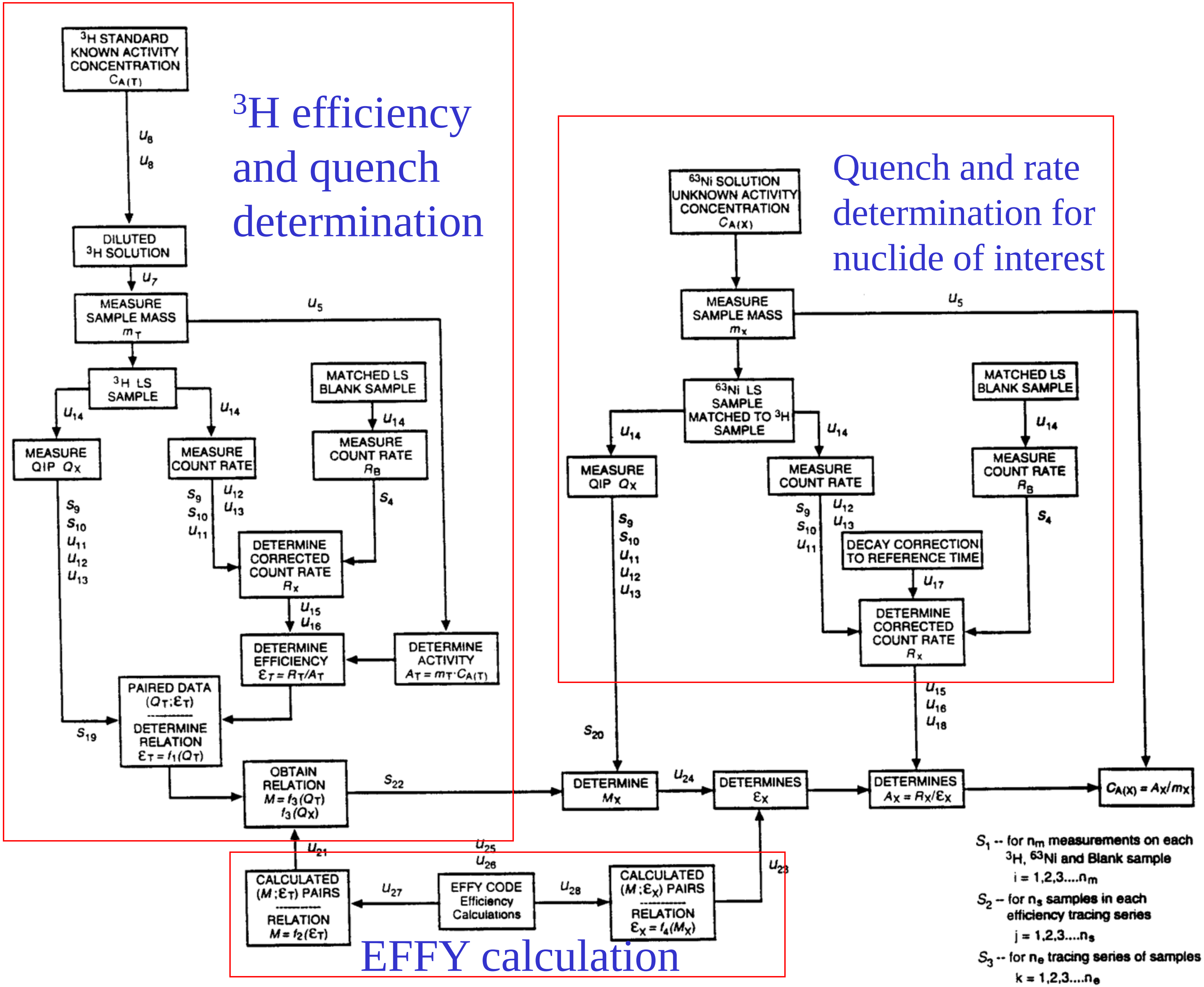
$$\varepsilon = \mathrm{f}(M)$$

CIEMAT/NIST Efficiency Tracing Method

- Prepare chemically-matched LS cocktails of ^{3}H and nuclide of interest over some quenching range.
- Experimental ^{3}H efficiency is used to determine a “Figure of Merit”, *M*, which is used to calculate the efficiency of nuclide of interest.

CIEMAT/NIST Efficiency Tracing Method





^{3}H efficiency and quench determination
Quench and rate determination for nuclide of interest
EFFY calculation
^{3}H STANDARD KNOWN ACTIVITY CONCENTRATION $C_{A(T)}$
u_8
u_8
DILUTED ^{3}H SOLUTION
u_7
MEASURE SAMPLE MASS m_T
u_5
^{3}H LS SAMPLE
u_{14}
u_{14}
MATCHED LS BLANK SAMPLE
u_{14}
MEASURE QIP Q_X
MEASURE COUNT RATE
MEASURE COUNT RATE R_B
S_9 S_{10} u_{11} u_{12} u_{13}
S_9 S_{10} u_{11}
u_{12} u_{13}
S_4
DETERMINE CORRECTED COUNT RATE R_X
u_{15} u_{16}
DETERMINE EFFICIENCY $\varepsilon_T = R_T/A_T$
DETERMINE ACTIVITY $A_T = m_T \cdot C_{A(T)}$
PAIRED DATA $(Q_T;\varepsilon_T)$ DETERMINE RELATION $\varepsilon_T = f_1(Q_T)$
S_{19}
OBTAIN RELATION $M = f_3(Q_T)$ $f_3(Q_X)$
S_{22}
^{63}Ni SOLUTION UNKNOWN ACTIVITY CONCENTRATION $C_{A(X)}$
MEASURE SAMPLE MASS m_X
u_5
^{63}Ni LS SAMPLE MATCHED TO ^{3}H SAMPLE
u_{14}
u_{14}
MATCHED LS BLANK SAMPLE
u_{14}
MEASURE QIP Q_X
MEASURE COUNT RATE
MEASURE COUNT RATE R_B
S_9 S_{10} u_{11} u_{12} u_{13}
S_9 S_{10} u_{11}
u_{12} u_{13}
S_4
DECAY CORRECTION TO REFERENCE TIME
u_{17}
DETERMINE CORRECTED COUNT RATE R_X
u_{15} u_{16} u_{18}
S_{20}
DETERMINE M_X
u_{24}
DETERMINES ε_X
DETERMINES $A_X = R_X/\varepsilon_X$
$C_{A(X)} = A_X/m_X$
u_{21}
u_{25} u_{26}
u_{23}
CALCULATED $(M;\varepsilon_T)$ PAIRS RELATION $M = f_2(\varepsilon_T)$
u_{27}
EFFY CODE Efficiency Calculations
u_{28}
CALCULATED $(M;\varepsilon_X)$ PAIRS RELATION $\varepsilon_X = f_4(M_X)$
S_1 -- for n_m measurements on each ^{3}H, ^{63}Ni and Blank sample i = 1,2,3....n_m
S_2 -- for n_s samples in each efficiency tracing series j = 1,2,3....n_s
S_3 -- for n_e tracing series of samples k = 1,2,3....n_e

Conclusion

- We have developed a model for analyzing uncertainties encountered using the CIEMAT/NIST efficiency tracing method.
- Model is consistent with ISO Guide to Expression of Uncertainty in Measurement.
- Present model neglects correlated effects
- Model can evolve as more data are collected.