# Development of Rapid Liquid Scintillation Counting Method for Determination of Tritium in Foods


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# Motivations

- To improve routine method used for monitoring discharge of $^{3}H$ from nuclear power plant into atmospheric, aquatic, and terrestrial ecosystems
- To enable rapid screening of $^{3}H$ in foods for prompt decision making in the event of a nuclear or radiological emergency
- To compare performance of different LSC instrument technologies for analyzing $^{3}H$ in foods
- To develop a versatile LSC method for detecting $^{3}H$ in water and a wide variety of agricultural products
- To determine $^{3}H$ baseline radiation by examining $^{3}H$ activity in foods purchased from local market

# Objectives

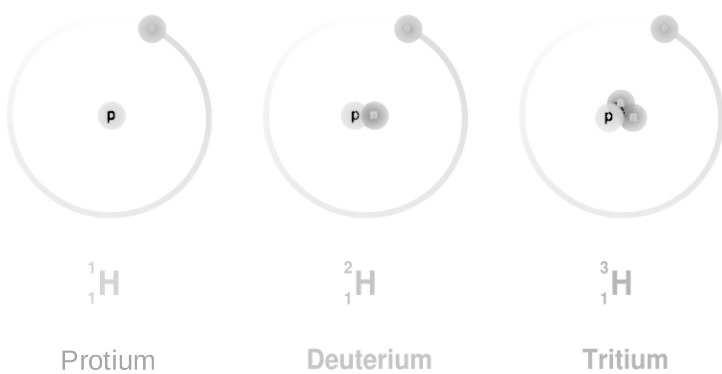
- Evaluate and validate distillation procedure to rapidly extract free-water from different types of foods suitable for analysis of $^{3}H$ by liquid scintillation counting
- Develop a simple method for rapid and accurate determination of water contents in foods to relate the $^{3}H$ activity measured in LSC sample to the $^{3}H$ activity presented in foods
- Determine optimum sample and instrument parameters for high throughput $^{3}H$ analysis complying with the data quality objectives per regulatory guideline
- Establish a rapid and versatile LSC method suitable for analyzing $^{3}H$ in a wide variety of foods and agricultural products

# About Tritium ($^3H$)

$^3H$ decays with a half-life of 12.3 years by emitting a β-particle to form $^3He$.

$$^3H \rightarrow {}^3He + \beta^- + \text{anti-neutrino}$$

*The* $E_{max}$ and $\bar{E}$ of β-ray emitted by $^3H$ are only 18 and 6 keV, which are too low for the radiation to penetrate skin. However, it has the ability to incorporate with the DNA and raise cancer risk when unduly inhaled, ingested, or absorbed over a prolonged period. As a result, its presence in potable water and food products must be monitored per regulatory guidelines.



# Current Methods Used for $^3H$ Analysis

**NH State Lab:**

- ✓ Uses heating-mantle distillation method validated for various types of water samples
- ✓ Based on heating sample to release water then collecting condensed vapor
- ✓ Enables quick water extraction from sample
- ✓ Distills water from sample with an open system
- ✓ Accepts large size sample

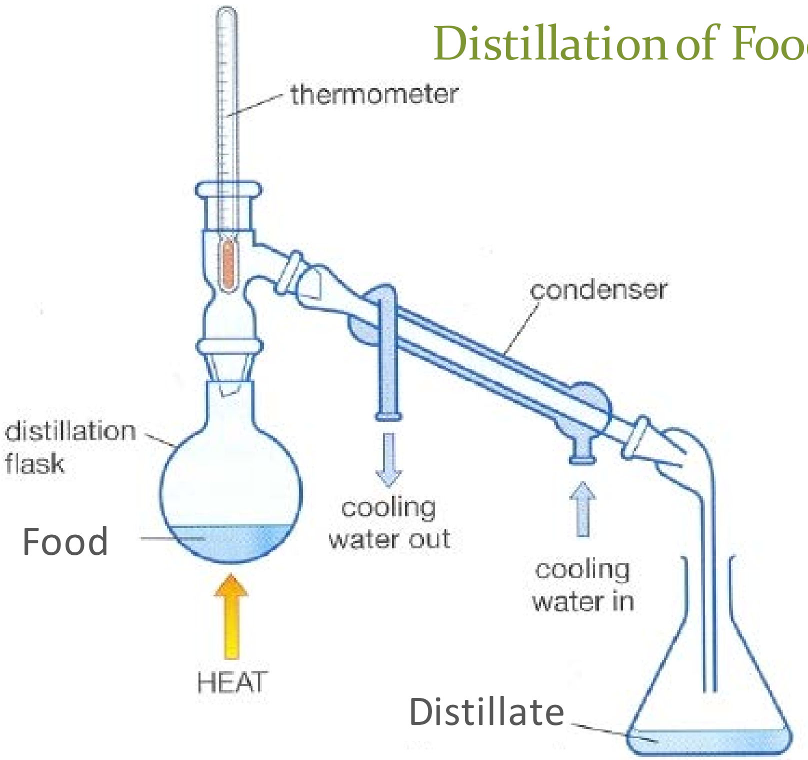
**FDA Rad Lab:**

- ✓ Uses vacuum distillation method validated for water and various foods
- ✓ Based on slowly extracting free water from foods
- ✓ Requires over night extraction
- ✓ Extracts water from sample with a closed system
- ✓ Limits sample size to ~30 grams

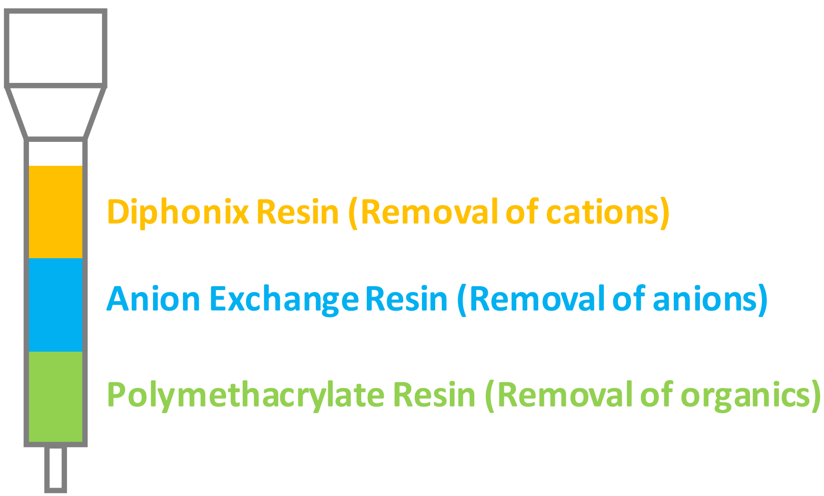
# Extend Heating-Mantle Method for Food Analysis

Free water in foods is readily separable from matrix by heating and reclaiming by condensation. Some color and organic substances may be found in distillate and cause color/chemical quench that biases low measurement results. Treatment of sample distillate with $^3H$ column may be necessary for certain foods.

Distillation of Food




Eichrom Tritium Column



# Measurement Model

Calculation of $^3$H concentration in food:

$$C_T = \frac{R_T \times F_W \times 10}{E_T \times W_{LSC} \times F_Q}$$

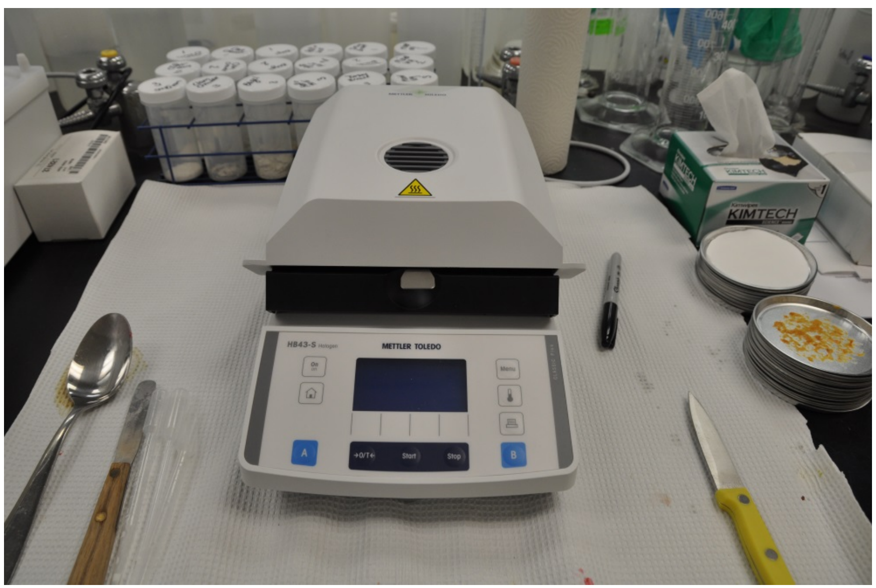
where,

$C_T$ = $^3$H concentration in food at time of sample analysis, Bq/kg
$R_T$ = Net sample tritium count rate, cps
$F_W$ = Food water content, %
$E_T$ = Tritium counting efficiency, cps/Bq
$W_{LSC}$ = Weight of distillate used for LSC counting, g
$F_Q$ = Quench correction factor
10 = Conversion factor

# Determination of Food Water Content

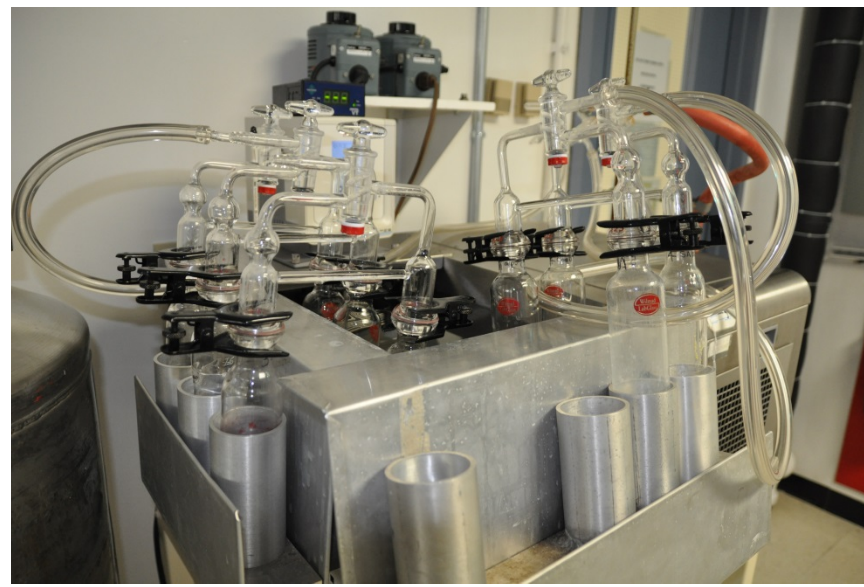
Moisture analyzer was proposed for rapid determination of food water content.  To validate its acceptance, the following two methods were used and compared

Moisture analyzer




Quick (~30 min)
Simple

Vacuum distillation



Slow (over night)
Accurate

# Comparison of Results

Food water contents determined by moisture analyzer and vacuum distillation

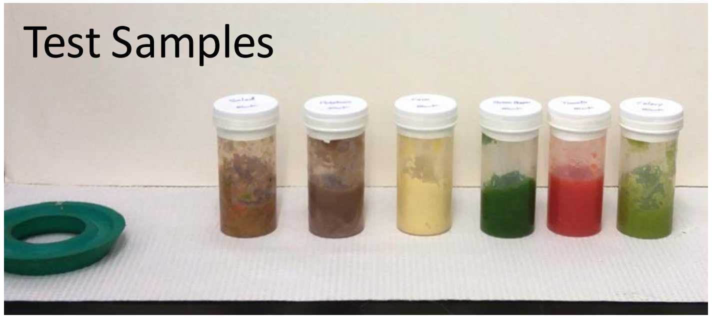
| Food | N | Moisture Analyzer Mean ± 2s | Vacuum Distillation | Diff., % |
|---|---|---|---|---|
| Lettuces | 3 | 96.35 ± 0.40 | 94.95 | -1.5 |
| Mushroom | 3 | 91.08 ± 0.34 | 89.66 | -1.6 |
| Pear | 3 | 83.23 ± 0.52 | 83.33 | 0.1 |
| Cucumber | 3 | 95.66 ± 0.17 | 95.32 | -0.4 |
| Zucchini | 3 | 94.24 ± 0.06 | 93.60 | -0.7 |
| Green Beans | 2 | 90.73 ± 0.18 | - | - |
| Sweet Potato | 2 | 79.00 ± 0.24 | - | - |
| Greek Yogurt | 2 | 76.78 ± 1.78 | - | - |
| Watermelon | 2 | 91.61 ± 0.04 | - | - |
| Whole Milk | 2 | 88.04 ± 0.13 | - | - |
| Mustard Green | 3 | 89.25 ± 0.34 | 87.01 | -2.6 |
| Chard | 3 | 91.13 ± 0.51 | 92.80 | 1.8 |
| Grape | 3 | 82.58 ± 0.52 | 82.06 | -0.6 |
| Orange | 3 | 86.06 ± 0.34 | 84.13 | -2.3 |
| Fish | 3 | 77.64 ± 0.51 | 77.25 | -0.5 |
| Ground Beef | 3 | 60.11 ± 2.02 | 61.46 | 2.2 |
| Ground Pork | 3 | 62.62 ± 1.47 | 64.77 | 3.3 |
| Strawberry | 3 | 89.14 ± 0.29 | 88.78 | -0.4 |
| Corn | 3 | 69.75 ± 0.12 | 69.16 | -0.8 |
| 2% Milk | 3 | 89.15 ± 0.11 | 87.77 | -1.6 |

# Test Materials and Method Procedure

List of matrix blanks and matrix spikes used

| Food | Weight, g | Food | Weight, g |
|---|---|---|---|
| Corn | 65.21 | Strawberry | 65.55 |
| Celery | 65.47 | Blueberry | 65.69 |
| Potatoes | 65.27 | Green Beans | 65.64 |
| Tomato | 65.45 | Sweet Potato | 67.66 |
| Green Pepper | 65.18 | Fish | 60.64 |
| Mixed salad | 65.96 | Apple | 70.40 |
| | | Plain Greek Yogurt | 66.85 |
| | | Watermelon | 66.37 |
| | | Whole Milk | 66.30 |
| | | 2% Milk | 65.26 |
| | | Corn | 69.28 |


# Preparation of sample for $^{3}H$ analysis


Test Samples

Each test sample was used entirely

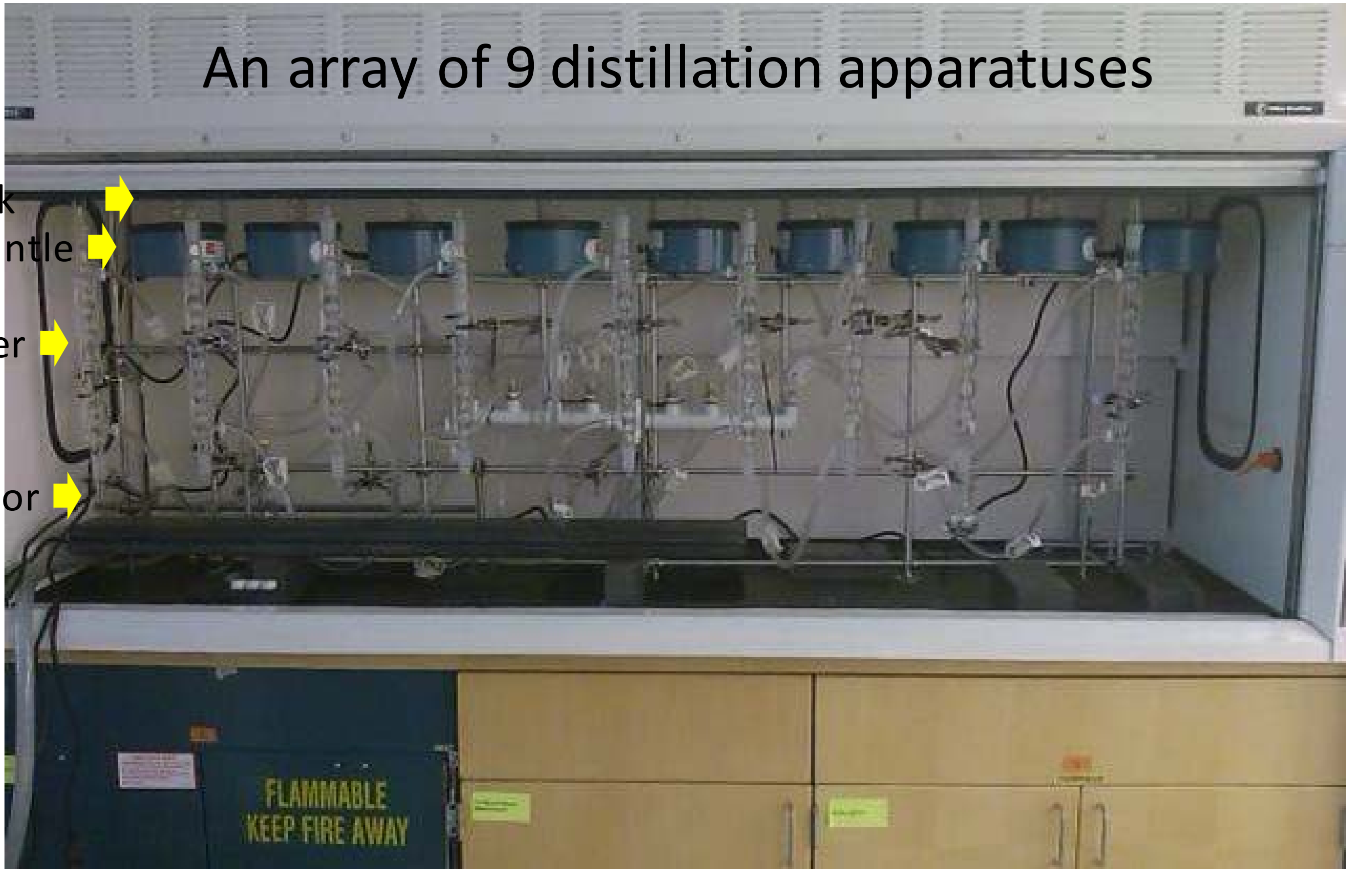

Sample Transfer


Post Distillation

Distillation of foods were found to be mostly straightforward but certain types of foods need additional treatment to obtain clear and colorless distillate before LSC counting.

# Distillation System Setup

- Capable of distilling 9 samples at a time within one hour
- Assembled inside a fume hood to prevent $^3H$ from escaping into the room



# Materials and Method (Cont.)

- Figure of Merit:

$$FOM = \frac{(Efficiency^2).(Volume^2)}{Background}$$

- 8 mL sample + 12 ml of Ultima Gold LLT
- Teflon-coated plastic vial
- Efficiency = 43% (Hidex 300SL)
- Measurements done with and without dark adaptation: the activities for non-dark adapted blank samples were ~10% higher compared to the 2-hour dark adapted ones

# Removal of Sample Color/Cloudiness

- Eichrom's Tritium column was used to treat colored and/or cloudy sample distillate to eliminate the need for repeating the distillation, thus expedite the sample preparation


Before treatment


After treatment

# Comparison of Instruments

- Liquid scintillation counters used:
  - Hidex 300SL
  - Qunatulus 1220
  - TriCarb 3170 TR/SL
- Count time:
  - 100 min – Hidex
  - 60 min – Quantulus 1220
  - 60 min – TriCarb 3170 TR/SL
- Background suppression:
  - TDCR + Digital shielding + Cooling – Hidex
  - Anti – coincidence + Pb Shield – Quantulus 1220
  - Anti – coincidence + BGO Shield – TriCarb 3170 TR/SL

*Hidex 300SL*

*TriCarb 3170 TR/SL*


*Quantulus 1220*

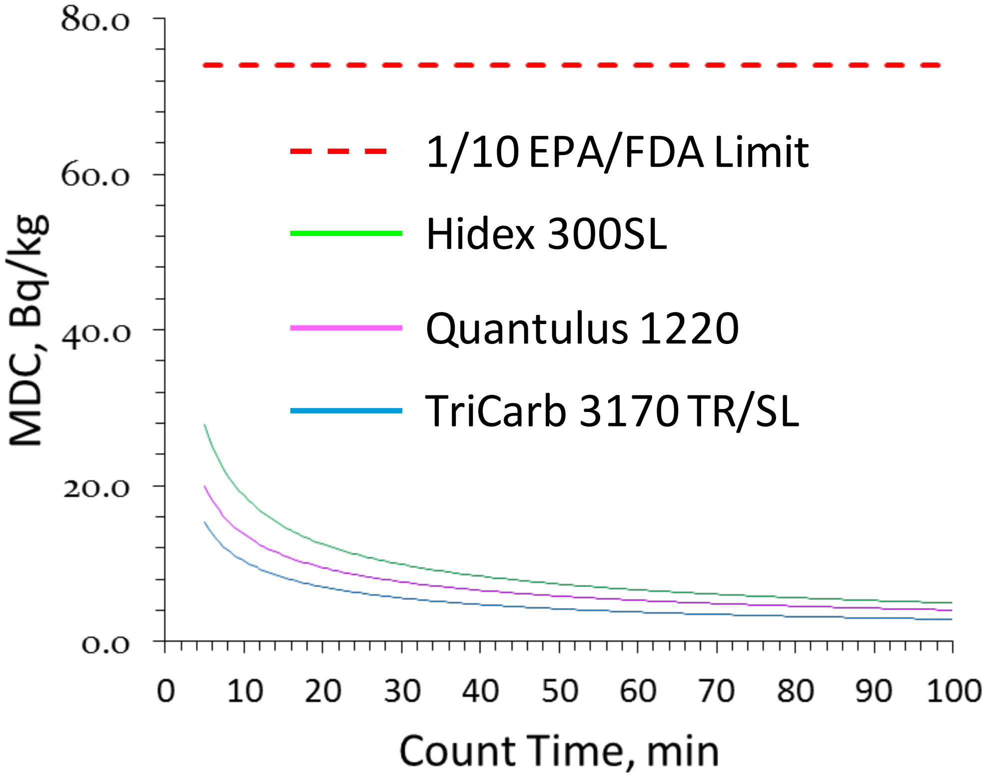
# Comparison of $^3H$ Matrix Blank Count Rates

| Food Matrix | Hidex 300SL, cpm | Quantulus 1220, cpm | TriCarb 3170 TR/SL, cpm |
|---|---|---|---|
| Celery | 6.95 ± 0.68 | 3.20 ± 0.47 | 0.72 ± 0.41 |
| Potatoes | 7.45 ± 0.70 | 3.03 ± 0.45 | 0.15 ± 0.37 |
| Tomato | 7.02 ± 0.68 | 3.20 ± 0.47 | 0.56 ± 0.40 |
| Green Pepper | 7.19 ± 0.69 | 3.30 ± 0.47 | 0.41 ± 0.39 |
| Mixed Salad | 7.07 ± 0.69 | 3.01 ± 0.45 | 0.51 ± 0.39 |
| Watermelon | 6.58 ± 0.66 | 3.33 ± 0.48 | 0.60 ± 0.41 |
| Blueberry | 6.05 ± 0.64 | 3.15 ± 0.46 | 0.52 ± 0.41 |
| Strawberry | 7.05 ± 0.68 | 3.90 ± 0.52 | 0.49 ± 0.41 |
| 2% Milk | 7.73 ± 0.72 | 3.71 ± 0.50 | 0.67 ± 0.42 |
| Whole Milk | 6.80 ± 0.67 | 3.25 ± 0.47 | 0.31 ± 0.39 |
| Corn | 6.89 ± 0.67 | 4.02 ± 0.52 | 0.15 ± 0.39 |
| Fish | 7.33 ± 0.70 | 3.49 ± 0.49 | 0.32 ± 0.38 |
| Sweet Potato | 7.45 ± 0.70 | 3.32 ± 0.48 | 0.25 ± 0.39 |

All uncertainties are given at 95% confidence level

# Estimated Detection Limits vs EPA/FDA Limit

- Sample volume = 8 mL
- Ultima Gold LLT = 12 mL
- Method blank = ~7 cpm Hidex 300SL (Eff = ~43%)
- Method blank = ~2 cpm Quantulus 1220 (Eff = ~29%)
- Method blank = ~0.5 cpm TriCarb 3170 TR/SL (Eff = 21% UltraLow level mode)




# Observed Sample Quench Effect

| Food Matrix | Quantulus 1220 Quench Index, SQP | TriCarb 3170 TR/SL Quench Index, tSIE |
|---|---|---|
| Celery | 715.94 | 349.52 |
| Potatoes | 714.50 | 345.89 |
| Tomato | 722.49 | 368.72 |
| Green Pepper | 718.76 | 360.54 |
| Mixed Salad | 723.60 | 371.24 |
| Watermelon | 724.75 | 375.57 |
| Blueberry | 725.44 | 384.88 |
| Strawberry | 723.17 | 379.12 |
| 2% Milk | 723.77 | 376.89 |
| Whole Milk | 727.39 | 384.67 |
| Corn | **701.13** | **311.20** |
| Fish | 725.45 | 383.03 |
| Sweet Potato | **698.34** | **313.27** |

Abnormal sample quench and instability were observed for foods with high starch contents, which biased the results without polishing the sample distillate with Eichrom tritium column

# Comparison of $^3H$ Results

| Food Matrix | Known Bq/kg | Hidex Bq/kg | Quantulus 1220 Bq/kg | TricCarb 3170 Bq/kg |
|---|---|---|---|---|
| Celery | 636.18 ± 7.84 | 603.07 ± 11.49 | 651.30 ± 17.45 | 604.96 ± 19.9 |
| Potatoes | 638.13 ± 7.87 | 622.20 ± 10.69 | 469.24 ± 13.42 | 468.04 ± 16.0 |
| Tomato | 636.37 ± 7.84 | 651.07 ± 11.86 | 679.39 ± 17.66 | 651.48 ± 20.5 |
| Green Pepper | 639.01 ± 7.88 | 657.06 ± 11.79 | 643.52 ± 16.99 | 634.18 ± 20.0 |
| Mixed Salad | 631.45 ± 7.78 | 629.69 ± 11.79 | 676.57 ± 17.86 | 658.44 ± 20.9 |
| Watermelon | 502.04 ± 6.19 | 457.24 ± 9.49 | 520.18 ± 15.29 | 501.52 ± 17.55 |
| Blueberry | 253.62 ± 3.13 | 274.85 ± 7.29 | 272.22 ± 10.45 | 264.12 ± 12.36 |
| Strawberry | 254.16 ± 3.13 | 260.63 ± 7.20 | 278.80 ± 10.71 | 258.52 ± 12.41 |
| 2% Milk | 638.22 ± 7.87 | 619.17 ± 10.84 | 707.36 ± 17.83 | 632.82 ± 19.49 |
| Whole Milk | 628.21 ± 7.74 | 626.41 ± 10.79 | 681.71 ± 17.18 | 621.47 ± 19.08 |
| Corn | 601.19 ± 7.41 | 563.68 ± 9.73 | 479.82 ± 13.63 | 439.65 ± 15.26 |
| Fish | 549.48 ± 6.77 | 634.32 ± 10.37 | 565.13 ± 14.92 | 527.35 ± 16.77 |
| Sweet Potato | 246.23 ± 3.04 | 212.43 ± 6.49 | 211.96 ± 8.82 | 194.76 ± 10.30 |

All uncertainties are given at 95% confidence level

# Conclusions

- A moisture analyzer is reliable and accurate for determination of food water content needed to relate the $^{3}H$ activity measured in LSC sample to the $^{3}H$ activity presented in foods
- The preliminary results were found to be acceptable per FDA's data quality objective
- Considering the sensitivity, rapidness, and simplicity achieved, the method is suitable for high throughput $^{3}H$ analysis
- The method can eliminate color quench and maintain constant sample quench using Eichrom tritium column when necessary

# Conclusions (Cont.)

- Additional study is needed to fully validate the method with a wider range of food matrices
- Up to 36 food samples can be prepared for LCS counting per day
- Despite that the method presented an alternative approach for analyzing $^{3}H$ in foods, additional studies on the method performance characteristics are still needed before official use

# Acknowledgement

We would like to thank Lablogic Systems, Inc. (Hidex) for assisting instrument optimization and use of application software during the study.

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Thank you!

Any questions?

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