



*Analytical Results of Field  
Measurements for  $^{14}\text{C}$  in Gaseous  
Effluents From Nuclear Power Plants*

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# $^{14}\text{C}$ Chemical Forms

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- Organic carbon
  - Methane
  - Ethane
  - Ethylene
  - Other short chain hydrocarbons
- Inorganic Carbon
  - Carbon dioxide
  - Carbon monoxide
  - Bicarbonate, Carbonate
  - Particulate (?)



# Significance – Principal Radionuclide

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- Regulatory Guide 1.21, Rev 2
  - a radionuclide is considered a principal radionuclide if it contributes either
    - (1) greater than 1 percent of the 10 CFR Part 50, Appendix I, design **objective dose** for all radionuclides in the type of effluent being considered, or
    - (2) **greater than 1 percent of the activity** of all radionuclides in the type of effluent being considered.
  - Once principal radionuclides are identified...monitored IAW the sensitivity levels in the ODCM



# Significance – Principal Radionuclide (cont'd)

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- From RG 1.21 for  $^{14}\text{C}$ 
  - Because the production of C-14 is expected to be relatively constant at a particular site,
    - if sampling is performed for C-14 (instead of estimating C-14 ...), the sampling frequency may be adjusted to that interval that allows adequate measurement and reporting of effluents.
    - If estimating C-14 based on scaling factors and fission rates, a precise and detailed evaluation of C-14 is not necessary. It is not necessary to calculate uncertainties for C-14 or to include C-14 uncertainty in any subsequent calculation of overall uncertainty.
  - Lack of requirement for uncertainty estimate for calculated  $^{14}\text{C}$  *contradicts guidance* from MARLAP as cited in the RG...



# Requirements for Sampling Effluents- Liquids

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- Sample preservation is a necessity
- Liquid samples
  - Inorganic
    - Alkali– Inorganic converted to  $\text{HCO}_3^-$  and  $\text{CO}_3^{2-}$
    - Reduce temperature
  - Organic carbon requires investigation for proper preservation in liquids



# Requirements for Sampling Effluents - Solids

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- Solid Samples
  - Carbon chemical form is not certain
  - May be combination of organic, inorganic and particulate
    - Ship immediately after sampling
    - Refrigerate (minimize loss of volatiles)
    - Analyze immediately upon receipt



# Requirements for Sampling Effluents- Gases

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- Already discussed in first presentation by Jim Holtzclaw.



# Analytical Capabilities

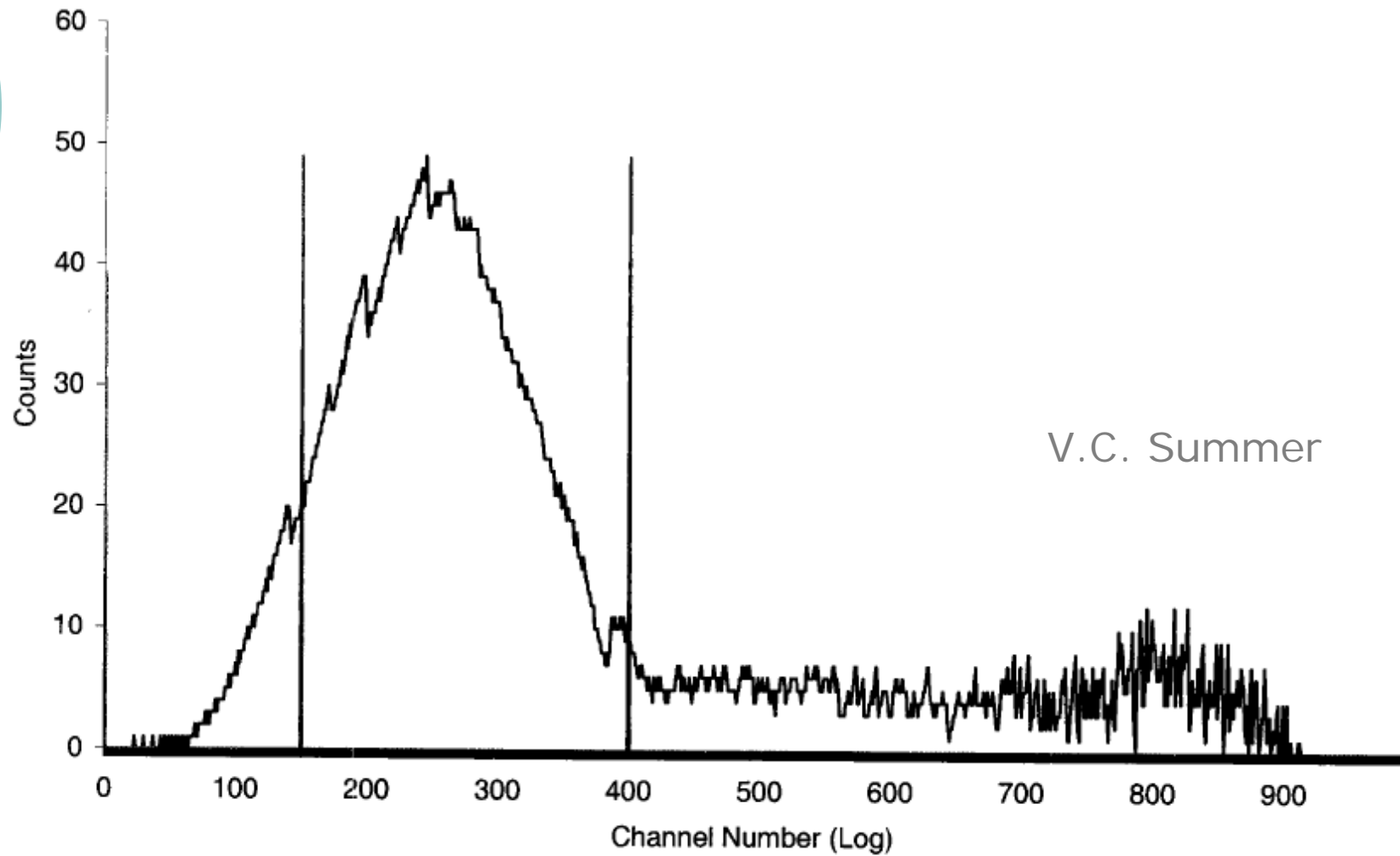
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- Analysis for  $^{14}\text{C}$  is done by Liquid Scintillation Counting
  - Sample Preparation – removes radionuclide impurities
  - $E_{\text{max}} = 156 \text{ keV}$
  - Detection Efficiency =  $\sim 95 \%$
- Analysis can also be done by GPC by precipitating  $\text{BaCO}_3$ .
  - Not as effective at contamination removal
  - Limited ability to assess contamination potential

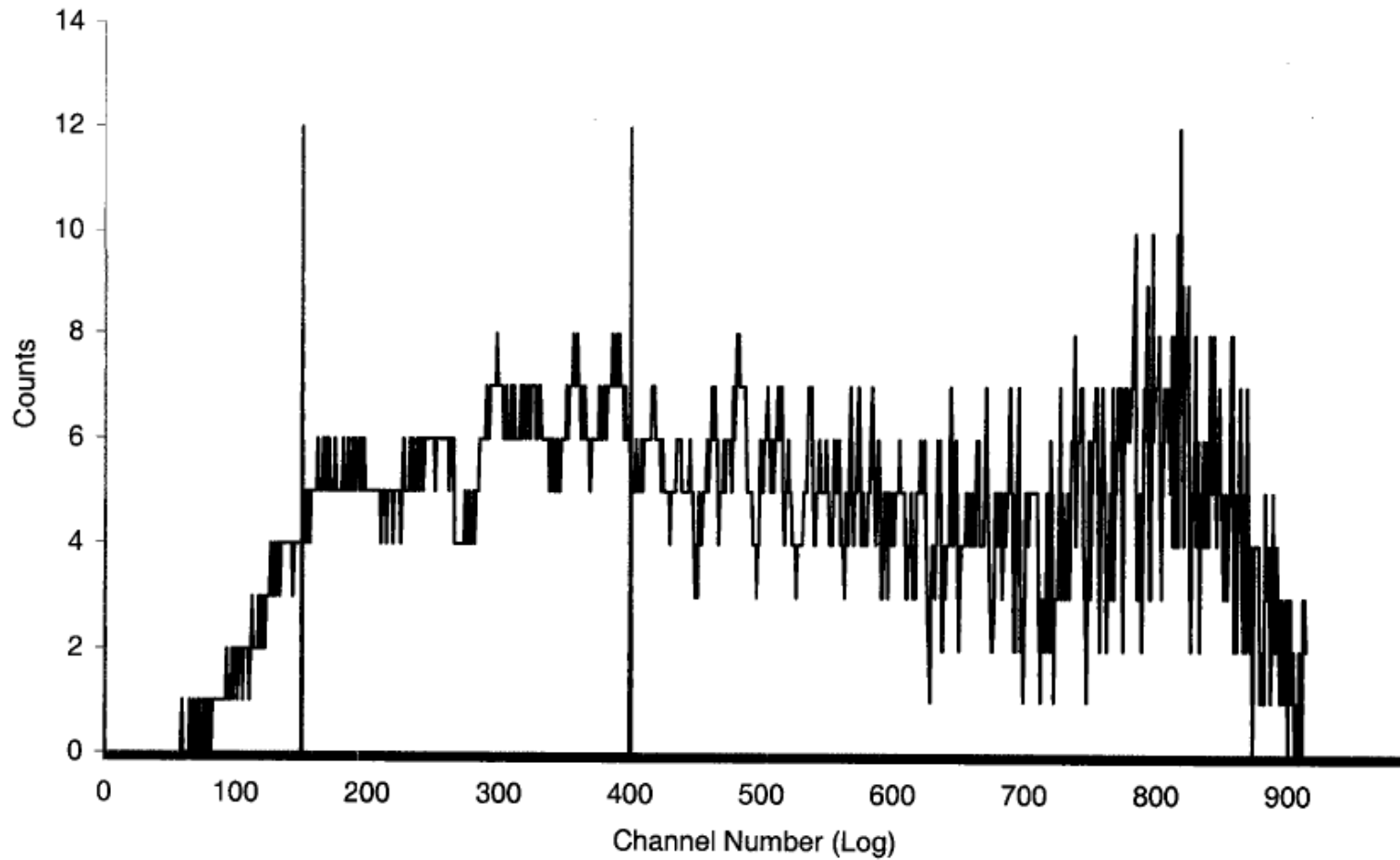


# $^{14}\text{C}$ Sample Spectrum Plant Vent

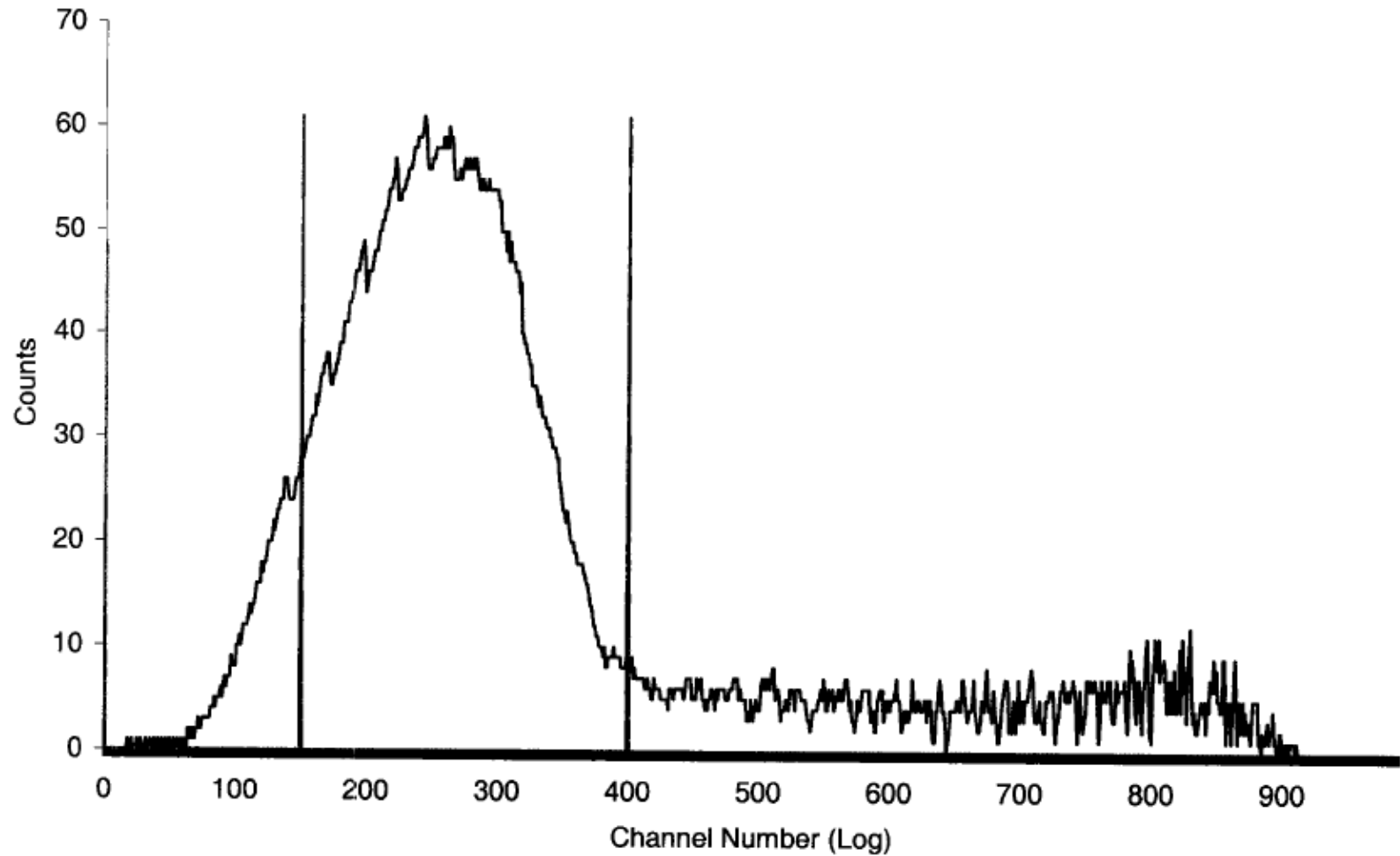
(1.39 pCi/L, 36 L sample, 120 min count time)



# $^{14}\text{C}$ Method Blank Spectrum



# LCS – Ascarite Spiked with $^{14}\text{C}$ Standard



# Analytical QC Data

<sup>14</sup>C in sucrose

Method Blank =  $-1.33 \pm 4.0 \times 10^{-11}$   $\mu\text{Ci/mL}$

QC	Filter blank, $\mu\text{Ci/mL}$	Ascarite Blank (Total) $\mu\text{Ci/mL}$	Ascarite Blank (Inorganic) $\mu\text{Ci/mL}$	LCS, $\mu\text{Ci/mL}$	LCS-Dup, $\mu\text{Ci/mL}$
Lab	$-3 \pm 4.1 \times 10^{-11}$	$-2.9 \pm 7.8 \times 10^{-11}$	$3.0 \pm 8.8 \times 10^{-11}$	$1.11 \pm 0.08 \times 10^{-9}$	$1.11 \pm 0.07 \times 10^{-9}$
Plant	---	$1.1 \pm 11 \times 10^{-11}$	$-7.2 \pm 15 \times 10^{-11}$	$1.83 \pm 0.52 \times 10^{-9}$	$1.87 \pm 0.50 \times 10^{-9}$

# Analytical Results

V.C. Summer

Location	Total, $\mu\text{Ci}/\text{mL}$ [ $\mu\text{Ci}/\text{ft}^3$ ]	% Organic	% Inorganic	Annual flow, $\text{ft}^3$
Plant Vent	$(1.40 \pm 0.18) \times 10^{-9}$ [ $(3.98 \pm 0.51) \times 10^{-5}$ ]	99.3	0.7	$1.1 \times 10^{11}$
Waste Gas #1	$(4.86 \pm 0.84) \times 10^{-3}$ [ $(1.38 \pm 0.24) \times 10^2$ ]	91.9	8.1	775 to 3192 for both
Waste Gas #3	$(5.62 \pm 0.14) \times 10^{-3}$ [ $(1.60 \pm 0.04) \times 10^2$ ]	93.1	6.9	↓

Uncertainty at 2 sigma

# Analytical Results


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Location	Total, $\mu\text{Ci/mL}$ [ $\mu\text{Ci/ft}^3$ ]	% Organic	% Inorganic	Annual flow, $\text{ft}^3$
Plant Vent	$(-6.34 \pm 7.11) \times 10^{-11}$ [ $(-1.8 \pm 2.0) \times 10^{-9}$ ]	---	---	
Waste Gas B	$(1.02 \pm 0.06) \times 10^{-2}$ [ $(2.88 \pm 0.17) \times 10^2$ ]	84.6	15.8	

Uncertainty at 2 sigma

# Analytical Results

## Diablo



Sample Point	Total, $\mu\text{Ci/mL}$	$\text{CO}_2$ , $\mu\text{Ci/mL}$	%	Organic, $\mu\text{Ci/mL}$	%
U1-CTMT	$5.2 \times 10^{-7}$	$3.96 \times 10^{-8}$	7.6	$4.8 \times 10^{-7}$	92.4
U2-CTMT	$1.62 \times 10^{-6}$	$4.12 \times 10^{-8}$	2.5	$1.58 \times 10^{-6}$	97.5
U1- PV	$2.46 \times 10^{-9}$	$1.75 \times 10^{-10}$	7	$2.29 \times 10^{-9}$	93
U2 – PV	$3.35 \times 10^{-9}$	$1.31 \times 10^{-9}$	39	$2.05 \times 10^{-9}$	61
U2-WG-Dis	$4.45 \times 10^{-4}$	$4.7 \times 10^{-6}$	1	$4.40 \times 10^{-4}$	99
WGDT	$1.88 \times 10^{-3}$	$2.06 \times 10^{-5}$	1	$1.86 \times 10^{-3}$	99

Uncertainty at 2 sigma



# Sample Locations

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- Plant Vent
- Containment
- Waste Gas Decay Tanks
- Waste gas header during discharge
- FSB Vent header
- Auxiliary Building vent header
- RCS
- Waste Liquid





# What Happens to $^{14}\text{C}$ Concentration if a Fuel Defect Develops?

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- $^{14}\text{C}$  formed by activation of oxygen in the  $\text{UO}_2$  (3.5 Ci per year - 1,000 MWe)
- Fuel also has some nitride (15 Ci per year - 1,000 MWe)
- Nitrogen in Fuel clad (30 Ci per year - 1,000 MWe)
  - Release to RCS is based on number and type of defects, escape capacity from cladding
- SFP?

Reference: "CARBON-14 PRODUCTION IN NUCLEAR REACTORS", W. Davis, ORNL/NUREG/TM-12 (1977)



# Use of Marker Gamma Emitters

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- Can Gamma ray emitters be used as a marker for estimating  $^{14}\text{C}$  activity?
  - Radionuclide would need to:
    - Be directly related to neutron flux
    - have relatively constant concentration over the fuel cycle
    - be measured with sufficient accuracy to identify changes based on the ratio
    - Measured during normal and fuel defect cycles



# Use of Marker Gamma Emitters

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- Candidates
  - $^{16}\text{N}$
  - $^{133}\text{Xe}$  (RCS)
  - $^{41}\text{Ar}$  (containment)
  - Others?