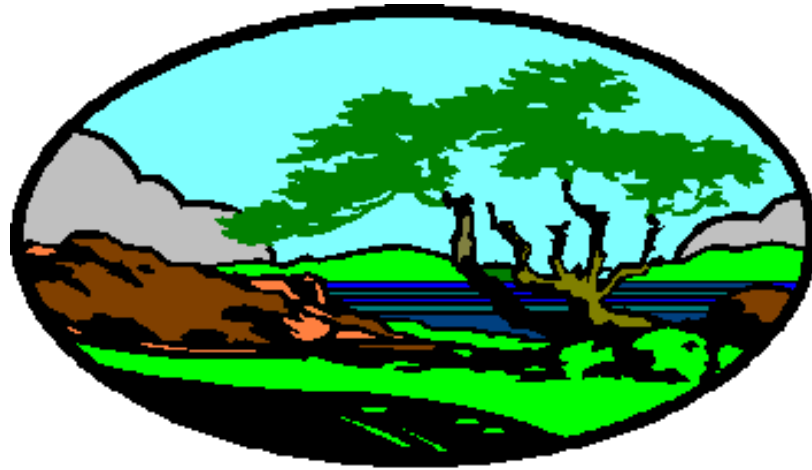


Tritium Deposition



Jim Key

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The Problem



- Tritium Released to the Atmosphere
 - “Recaptured” and seen in surface runoff.
 - See REMP Reports Statements Like:
 - “Tritium recapture (scavenging, deposition, etc.) by precipitation is a known phenomenon.”
 - Only Anecdotal Evidence
- Can the high levels of H-3 in surface runoff be *truly attributed* to recapture of H-3 in the air?

Atmospheric Removal Mechanisms

- Wet Deposition (Precipitation Scavenging)
 - Rain Washout
 - Snow Scavenging
- Dry Deposition
- Condensation onto Surfaces
 - Contaminated Concrete

Rain Washout

- Vapor Exchange Process
 - Occurs both ways.
 - Rate of change not equal.
 - Lower Temps Favor Vapor To Liquid.
 - Think Condensation
 - Higher Temps Favor Liquid To Vapor
 - Think Evaporation

Modeling Rain Washout

- Short Term Precipitation Events Require
 - Detailed Meteorology
 - Cloud Base
 - Rain Spectra
 - Rain Rose
 - Etc.

Modeling Rain Washout

- Long Term Precipitation Washout Can Be Modeled
 - Assumes Along Period of Time
 - Seasonal or Annual
 - Uses Washout Factor
 - W_M – Mass Washout Factor ($\text{kg}_{\text{air}} / \text{kg}_{\text{rain}}$)
 - W_V – Volumetric Washout Factor ($\text{m}^3_{\text{air}} / \text{m}^3_{\text{rain}}$)
 - See NUREG/CR-3332

Rainwater Concentration

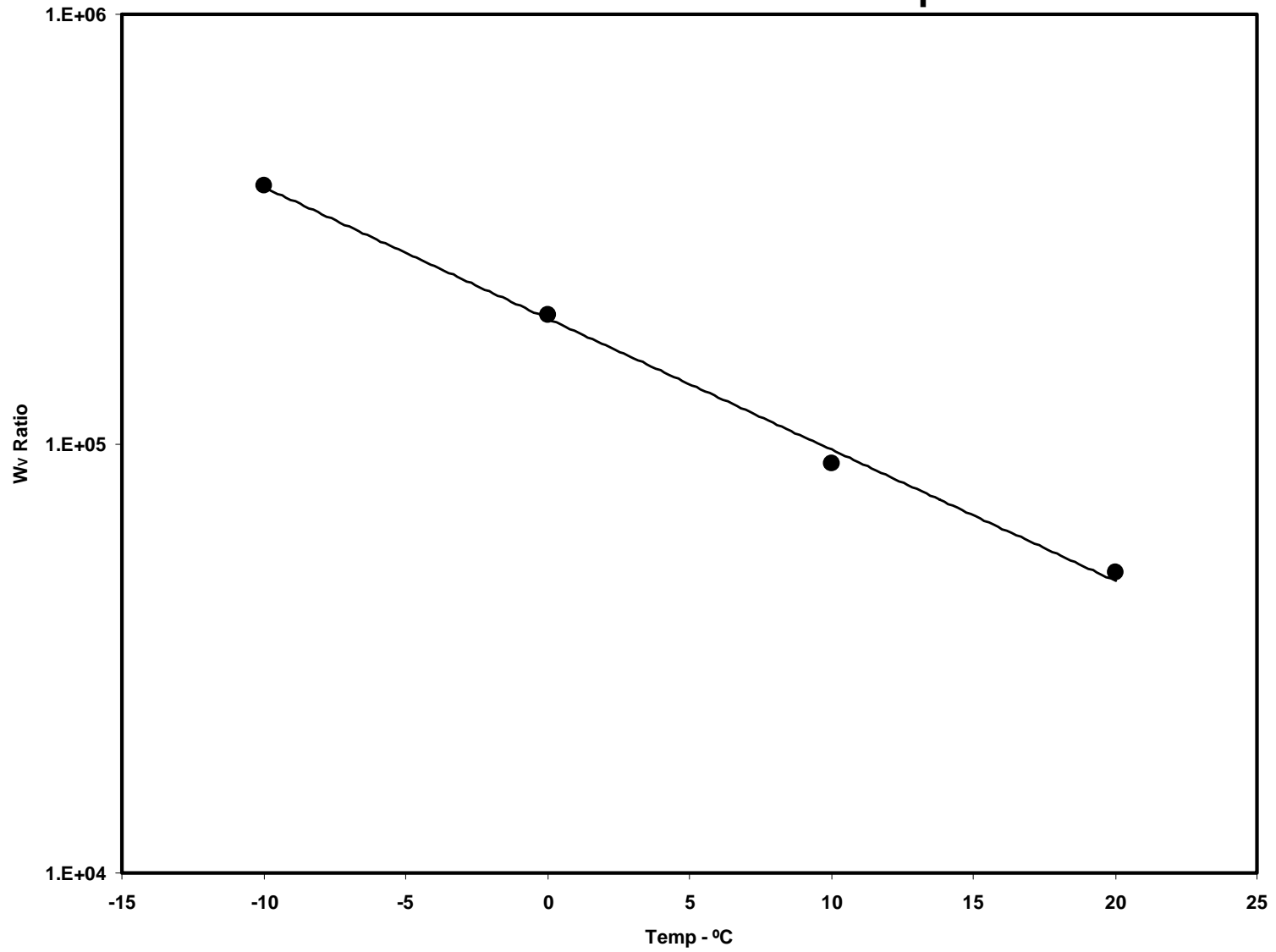
$$C_{\text{RAIN}} = 10^3 \overline{\chi/Q} \dot{Q} W_V$$

- C_{RAIN} Is the estimated concentration in rain water in units of pCi/liter.
- 10^3 Conversion constant to convert from uCi to pCi and from m^3 to liters.
- $\overline{\chi/Q}$ Atmospheric dispersion in units of sec/m^3 .
- W_V Volumetric Atmospheric Washout Factor

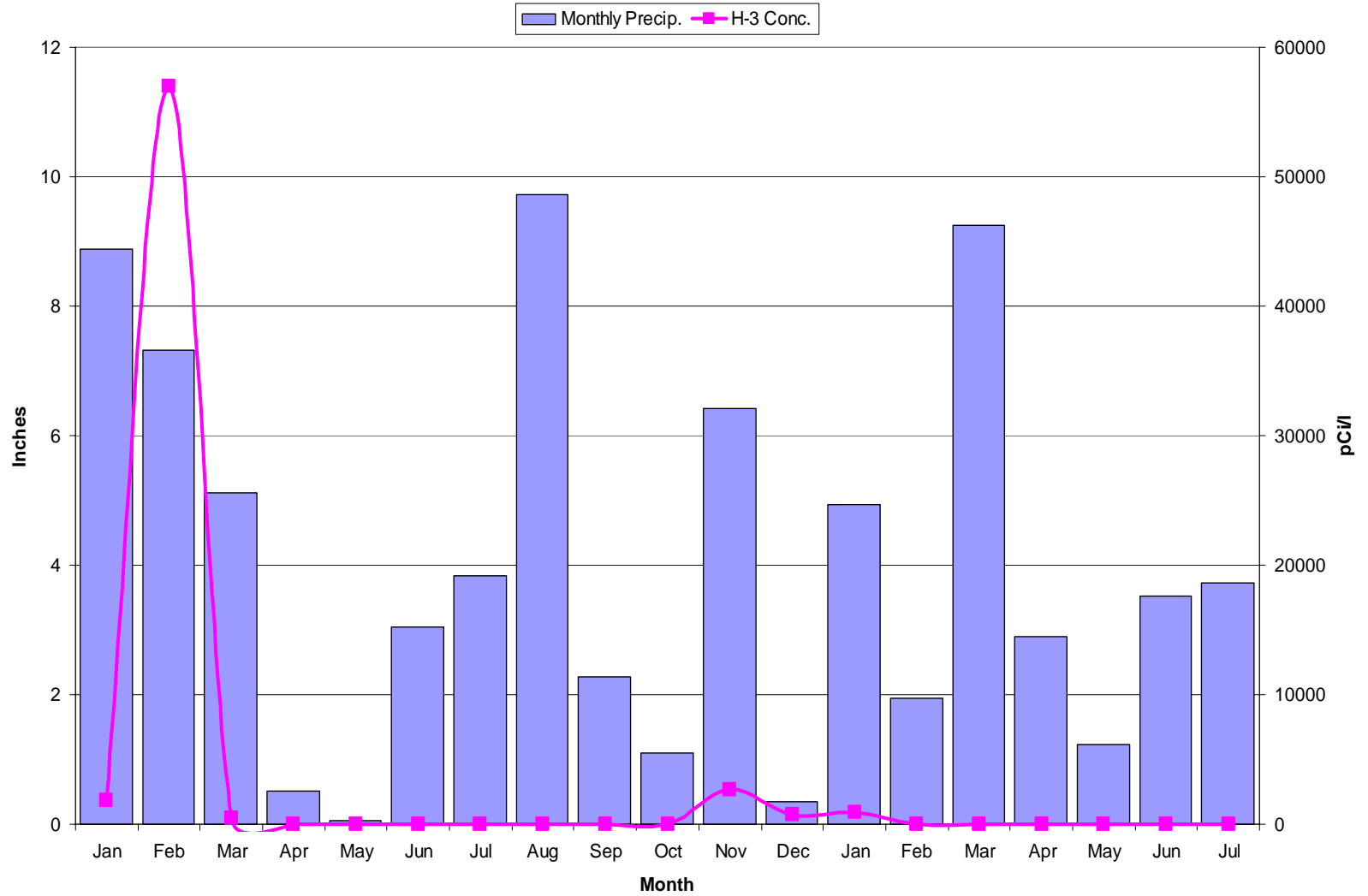
Tritium Washout Factors for Water Vapor

Atmospheric Temperature	Mass Washout Factor W_M (kg _{air} / kg _{rain})	Volumetric Washout Factor, W_V (m ³ _{air} / m ³ _{rain})
0°C	240	200,000
10°C	110	90,000
20°C	60	40,000

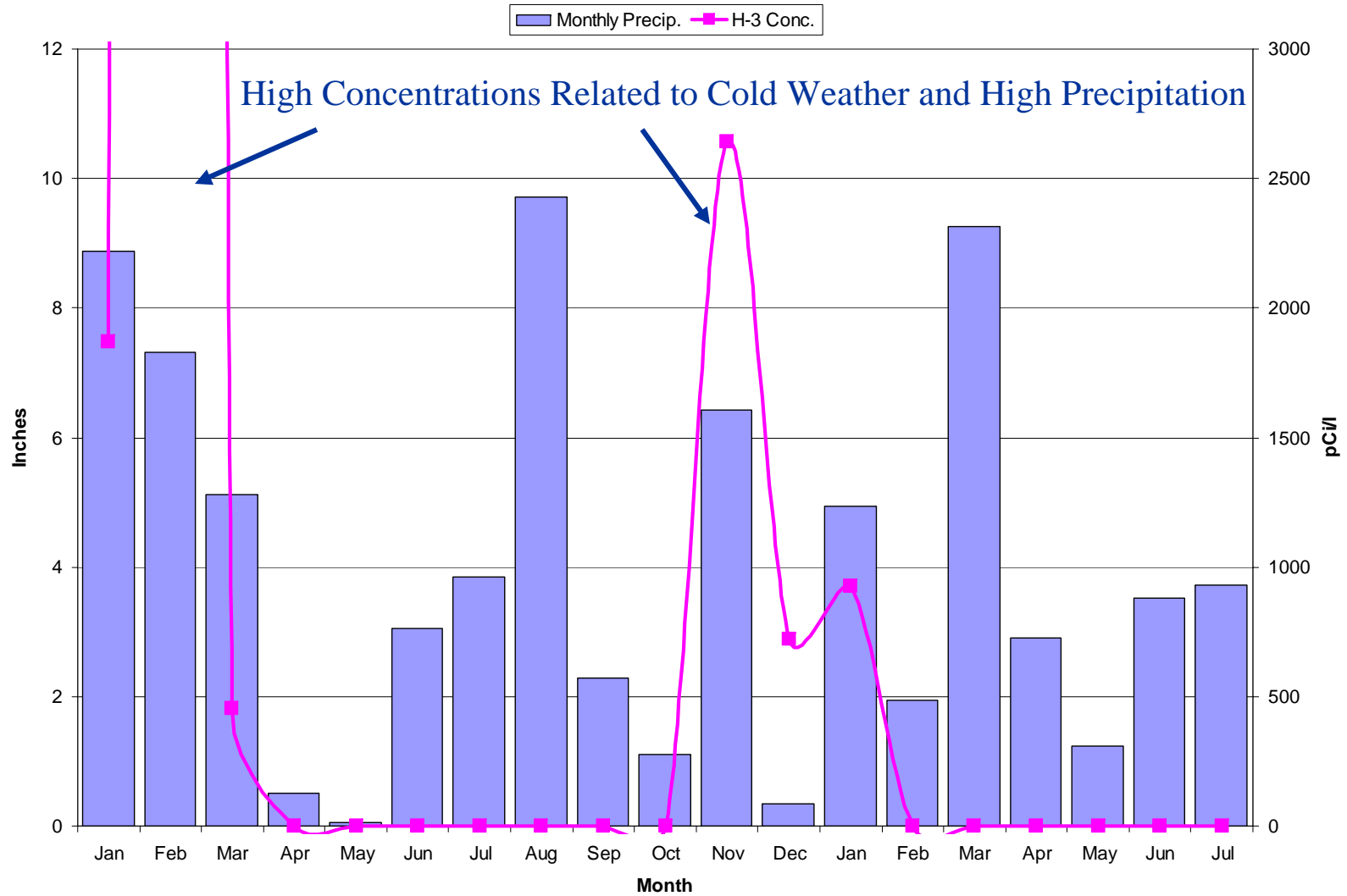
Volumetric Washout Ratio vs Temperature



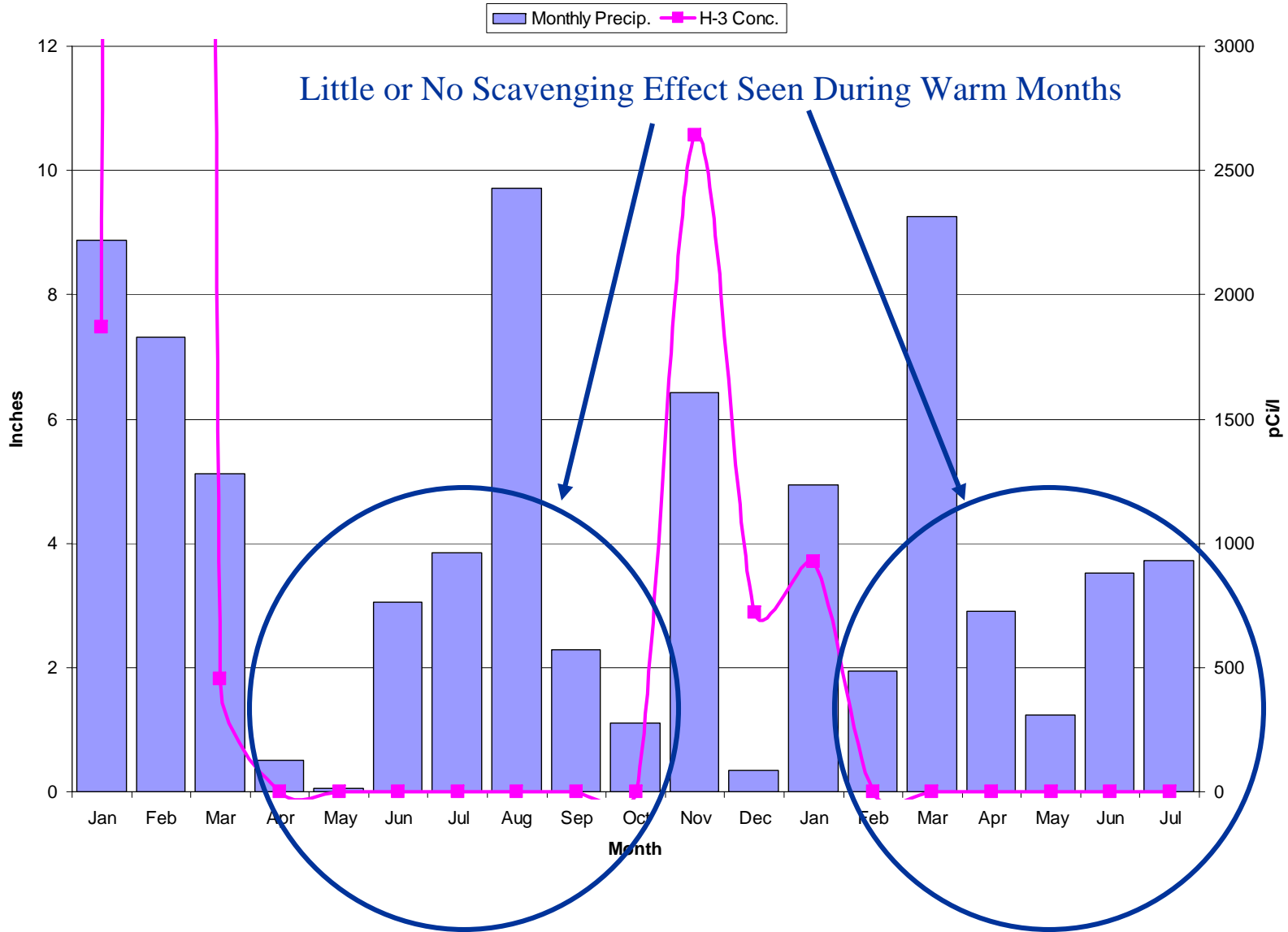
Monthly Precipitation and Tritium Concentrations (Rescaled H-3 Conc.)



Monthly Precipitation and Tritium Concentrations (Rescaled H-3 Conc.)



Monthly Precipitation and Tritium Concentrations (Rescaled H-3 Conc.)



Snow Scavenging

- Dependant Upon Type of Snow
 - Power vs. Heavy Wet Snow
- Two Removal Processes by Snow
 - Scavenging by Snow as it Falls Through the Atmosphere (Snow Fall Scavenging)
 - Scavenging by the Snow Pack After Accumulating on the Ground (Snow Pack Scavenging = Dry Deposition)

Snow Fall Scavenging

- Wet Deposition Process
 - Same Process as Scavenging by Rain Fall
 - Modeled in Similar Way
- Studies Show Efficiency of Snow Fall Scavenging to be One or Two Orders of Magnitude Below Rain Scavenging

High Snow Pack Concentrations

- Hinchcliffe Master's Thesis (ISU)
 - DC Cook Study
 - Measured Only “Fresh Snow” Tritium Concentrations
 - Did See Much Higher Concentrations in “Old Snow” (Snow Pack)
 - High “Old Snow” Concentrations Remained Unexplained

Snow Pack Scavenging

- Dry Deposition Process
 - Direct Vapor Exchange Between Air and Snow Pack [CNSC, IAEA]
 - Long Exposure Time
 - Hours to Days
 - Equilibrium Reached Quickly ~ Hours
 - Results in Substantially Higher Concentrations Than be Explained by Snow Fall Scavenging (Wet Deposition)

Snow Pack Scavenging

- This is an Integrating Process
- H-3 Concentrations in Snow Can Remain High Long After Atmospheric H-3 Levels Have Dropped
- Specific Activity of Water in Snow Pack Can Be a Substantial Fraction of the Specific Activity of Atmospheric Water

Tritium Dry Deposition Onto Snow Pack

$$C_s = 10^9 \frac{X/Q \dot{Q} \rho_w}{H}$$

Where:

C_s Tritium concentration in snow (water) in units of pCi/l.

10^9 Conversion constant to convert from uCi to pCi and from ml to liters.

X/Q Short term atmospheric dispersion in units of sec/m^3 .

\dot{Q} Release rate in units of uCi/sec.

ρ_w Density of water in units of gm/cc.

H Absolute atmospheric humidity in units of gm/m^3 .

Snow Scavenging Information

- Hinchcliffe, William, “Investigation of Tritium Recapture at Cook Nuclear Power Plant form Airborne Effluent Releases,” thesis draft, August 2010, Idaho State University, Pocatello (Dr. Jason Harris’s student).
- IAEA, “Modelling the environmental transport of tritium in the vicinity of long term atmospheric and sub-surface sources”
- Galerio, D., Melintescu, A., Attanasov, D., Guetata, P., and Patyl, L. “Review of HTO Washout,” Power Point presentation, Report to IAEA EMRAS Program Working Group, 2010.
- Orgam, G. L., “Precipitation Scavenging of Tritiated Water Vapour (HTO),” October 28, 1995. Ontario Hydro Research Division report number 85-233-K, Ontario Hydro
- Slinn, W. George N., “Precipitation Scavenging,” from *Atmospheric Science and Power Production*, Darryl Randerson, editor; DOE/TIC-27601, DOE 1984.

Tritium Retention and Release From Concrete

- “Hey it rained on my concrete and now I see tritium.”
 - Containment
 - Other Site Structures
 - Security Barricades
- Concrete Can Act As a Sink and a Source of HTO.

Lot's Going On

- Primary Mechanisms
 - Absorption
 - Adsorption
 - Ion Exchange
- Deep Diffusion
 - Slow Process
 - Small Impact on Short Term De-sorption from Runoff

Lots of Water in Concrete

- Cement Is Major Component in Concrete
- Water is Substantial Component of Cement
 - 67% Pore and Capillary Water
 - 20% Water of Crystallization
 - 12% Constituent Water
 - Hydrogenated Calcium
 - Calcium Silicate

Concrete as a Sink

- Very Fast Shallow Permeation into
 - Concrete Pores
 - Concrete Capillaries
 - Capillary Action
 - Distributes Water Across the Surface of Concrete
 - And Into the Concrete to a Depth of a Few Millimeters
- Deeper Penetration Proceeds Very Slowly

Concrete as a Source

- Water Stored in Shallow Layer of Pores and Capillaries
 - Highly Mobile
 - Exchanges Quickly and Freely With External Water
- Tritiated Easily Water Leaches Out
 - Contaminates Water That Comes in Contact With Concrete Surface
 - Precipitation
 - Condensate Runoff

Concrete as a Source

- Water In Pores and Capillaries
 - Shallow Layer
 - Subject to Removal By Weathering
 - Short Term Storage Mechanism

Cement Chemistry

- Cement is Self Desiccating
 - 20-25% of Cement In Initial Pour Can Remain Un-Hydrogenated
 - Over Time Un-Hydrogenated Cement Will Continue to Absorb Water
 - This is Why Concrete Gets Stronger as it Get Older
 - This Water is Incorporated Into Cement as Water of Crystallization

Water of Crystallization

- Not Available for Direct Exchange
- Is Available for Ion Exchange
- This is a LONG Term Storage Mechanism
 - Retains and Releases H-3 Well After Initial Exposure to HTO

Two Storage Compartments

- Short Term Storage
 - Concrete Pores and Capillaries
- Long Term Storage
 - Cement Continues to Incorporate Moisture As Water of Crystallization

(Note that concrete with a higher aggregate content has less water storage capacity.)

Short Term Storage

- Fast Shallow Penetration into Concrete Pores and Capillaries
- Easily Removed from Pores and Capillaries
- Can Result in Very High HTO Concentration in Surface Runoff Water

Long Term Storage

- Water Continues to be Incorporated into Cement
 - Water of Crystallization
 - Relatively Slow Process
- Ion Exchange Takes Place Slowly
 - Tritium Can Show Up Long After Initial Contact

Cross Over Pathways

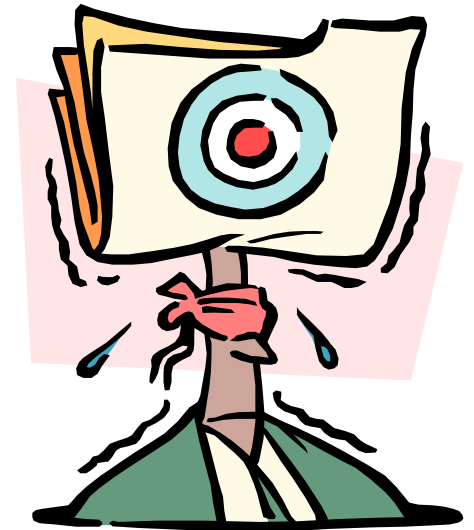
- Atmospheric Release Results in Dose Via Liquid Exposure Pathways
 - H-3 Deposition
- Liquid Release Results in Dose Via Gaseous Exposure Pathways
 - Pond Evaporation

What App I Limits Are Applicable?

- Dose Resulting From Tritium Recapture or Deposition...
 - App I Limits For Liquids?
 - 1.5 mrem/qtr and 3.0 mrem/yr Total Body
 - 5.0 mrem/qtr and 10.0 mrem/yr Any Organ
 - App I Limits for Airborne H-3?
 - 7.5 mrem/qtr and 15 mrem/yr Total Body and Organ

Who Do You Blame If You Challenge The Limits?

- Gaseous Radwaste Guy Say:
 - “It showed up in liquids? I don’t release liquids. It’s not my problem.”
- Liquid Radwaste Guy Say:
 - “Hey the Gas Radwaste guys had it last and they were the ones that let it go.”



Purpose of App I Dose Values

- Design Requirements for Radwaste Treatment Systems
 - Liquid Radwaste
 - Gaseous Radwaste
 - Iodine, Tritium and Particulates Released to the Atmosphere

Why Three Separate Design Requirements?

- Physical Properties of the Effluent!!
 - Liquids
 - Noble Gases
 - Halogens, Tritium and Particulates
- Requires Three Distinctly Different Approaches to Effluent Processing System Design

10 CFR 50.34a

- Design objectives for equipment to control releases of radioactive material in effluents for nuclear power plants
 - (a) “The guides set out in appendix I to this part provide numerical guidance on design objectives...”
 - Radwaste Systems had to be designed to meet Appendix I

Appendix I

- Section II - Specifies Design Objectives for Specific Radwaste Treatment Systems
 - Liquid Effluents (Sec II.A)
 - Gaseous Effluents (Section II.B)
 - Radioiodines, Particulates and Tritium (Section II.C)

Appendix I - Section II.A

- ...dose or dose commitment from liquid effluents for any individual in an unrestricted area from all pathways of exposure...
 - From Liquid Effluents
 - From All Pathways of Exposure
 - Dose Exposure Pathways (See Section III.A.1)
 - Not Discharge Pathways

Appendix I - Section II.B

- ...not result in an estimated annual air dose from gaseous effluents....
 - From Gaseous Effluents (Noble Gases)
 - Air Dose in mrad (B.1)
 - Total Body and Skin Dose in mrem (B.2)

Appendix I - Section II.C

- ...all radioactive iodine and radioactive material ... released ... in effluents to the atmosphere will not result in an estimated annual dose ... from all pathways of exposure...
 - Iodine, Tritium and Particulates Released In Effluents to the Atmosphere
 - From All Pathways of Exposure

Applicable Limits

- If dose is a result of the release of liquid effluents:
 - Regardless of Dose Exposure Pathway(s)
 - 3 mrem Total Body
 - 10 mrem Organ
- If there is no liquid effluent release then no dose is generated.

Applicable Limits

- If dose is a result of the release airborne release:
 - Regardless of Dose Exposure Pathway(s)
 - 10/20 mrad gamma/beta for Noble Gases
 - 15 mrem organ/total body for iodines, tritium and particulates
- If there is no airborne effluent release then no dose is generated.

Questions?

