Tritium Deposition

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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

\[ C_{\text{RAIN}} = 10^3 \frac{\chi}{Q} \dot{Q} W_V \]

- **\( C_{\text{RAIN}} \)**: 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.
- **\( \frac{\chi}{Q} \)**: Atmospheric dispersion in units of sec/m\(^3\).
- **\( W_V \)**: Volumetric Atmospheric Washout Factor
# Tritium Washout Factors for Water Vapor

<table>
<thead>
<tr>
<th>Atmospheric Temperature</th>
<th>Mass Washout Factor, $W_M$ (kg\textsubscript{air} / kg\textsubscript{rain})</th>
<th>Volumetric Washout Factor, $W_V$ (m\textsuperscript{3}\textsubscript{air} / m\textsuperscript{3}\textsubscript{rain})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0°C</td>
<td>240</td>
<td>200,000</td>
</tr>
<tr>
<td>10°C</td>
<td>110</td>
<td>90,000</td>
</tr>
<tr>
<td>20°C</td>
<td>60</td>
<td>40,000</td>
</tr>
</tbody>
</table>
Volumetric Washout Ratio vs Temperature

![Graph showing the relationship between Volumetric Washout Ratio and Temperature. The graph illustrates a linear decrease in Volumetric Washout Ratio as Temperature increases.](image-url)
Monthly Precipitation and Tritium Concentrations
(Rescaled H-3 Conc.)

- Monthly Precip.
- H-3 Conc.

- Months: Jan, Feb, Mar, Apr, May, Jun, Jul, Aug, Sep, Oct, Nov, Dec
- Precipitation in Inches: 0, 2, 4, 6, 8, 10, 12
- Tritium Concentration in pCi/l: 0, 10000, 20000, 30000, 40000, 50000, 60000

- Graph shows the distribution of monthly precipitation and Tritium concentrations across the year.
High Concentrations Related to Cold Weather and High Precipitation
Little or No Scavenging Effect Seen During Warm Months
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

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

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.

\( \rho_w \)  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”
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 Desorption 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
• …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?