

LBNF Target Pile Tritium Mitigation – - some details of the transport model

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Japan-US breakout meeting

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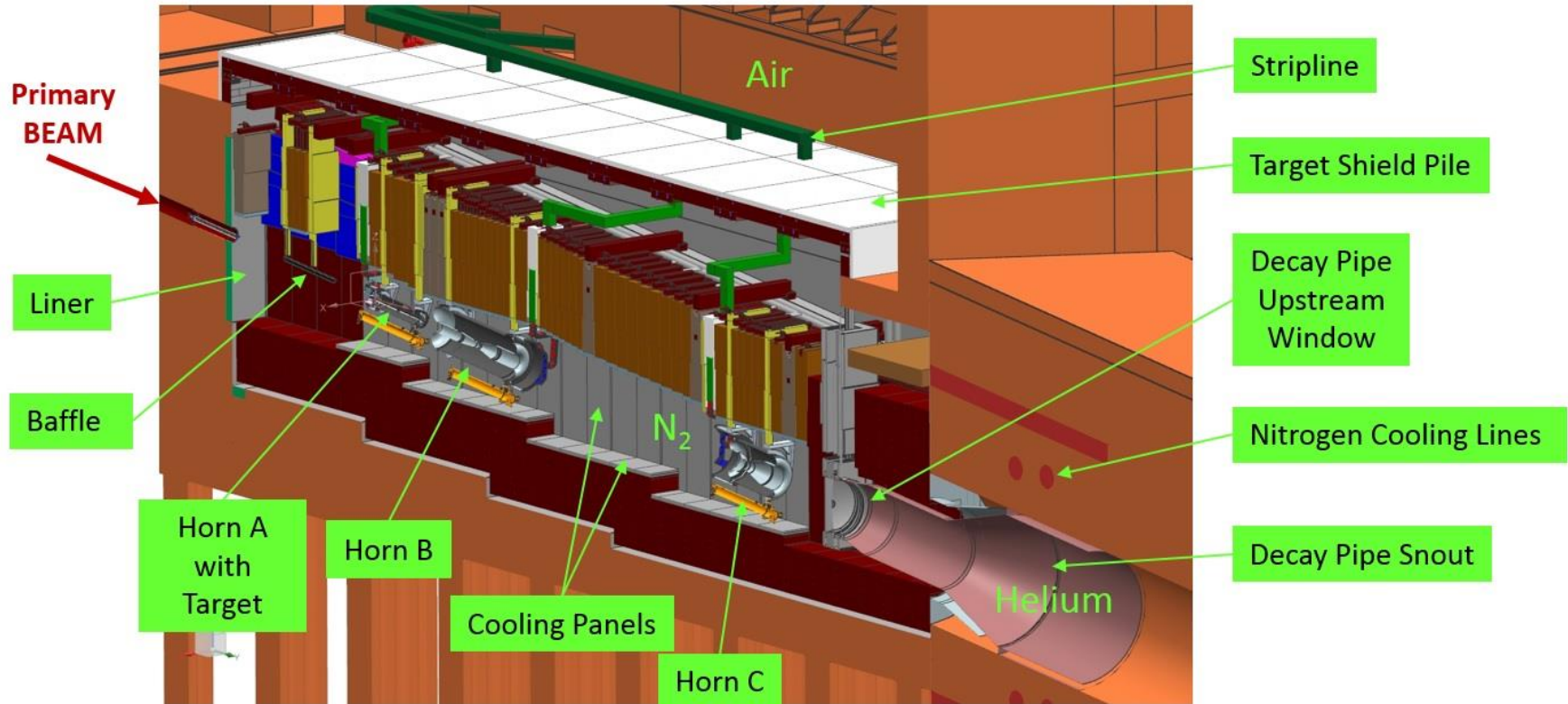
Supplemental material to NBI talk

For breakout

Tritium scrubbing scheme:

- Did 1st pass rough calculations to identify important elements and possible showstoppers, get feeling for transport timescales
 - In this talk, discuss a few transport steps and relevant calculations
- Will proceed to more refined calculations
- Want to identify any prototype tests that need to be done
 - Measure tritium transport through painted versus unpainted steel interface as function of temperature and humidity?
- Looking to get feedback about this scheme

LBNF target shield pile in target hall



- Central chase for components is 2.2 m/2.0 m wide, 34.3 m long, nitrogen-filled
- Bulk steel is inside N_2 vessel, N_2 cooled
- 4" inner layer of shielding water-cooled; replaceable cooling panels

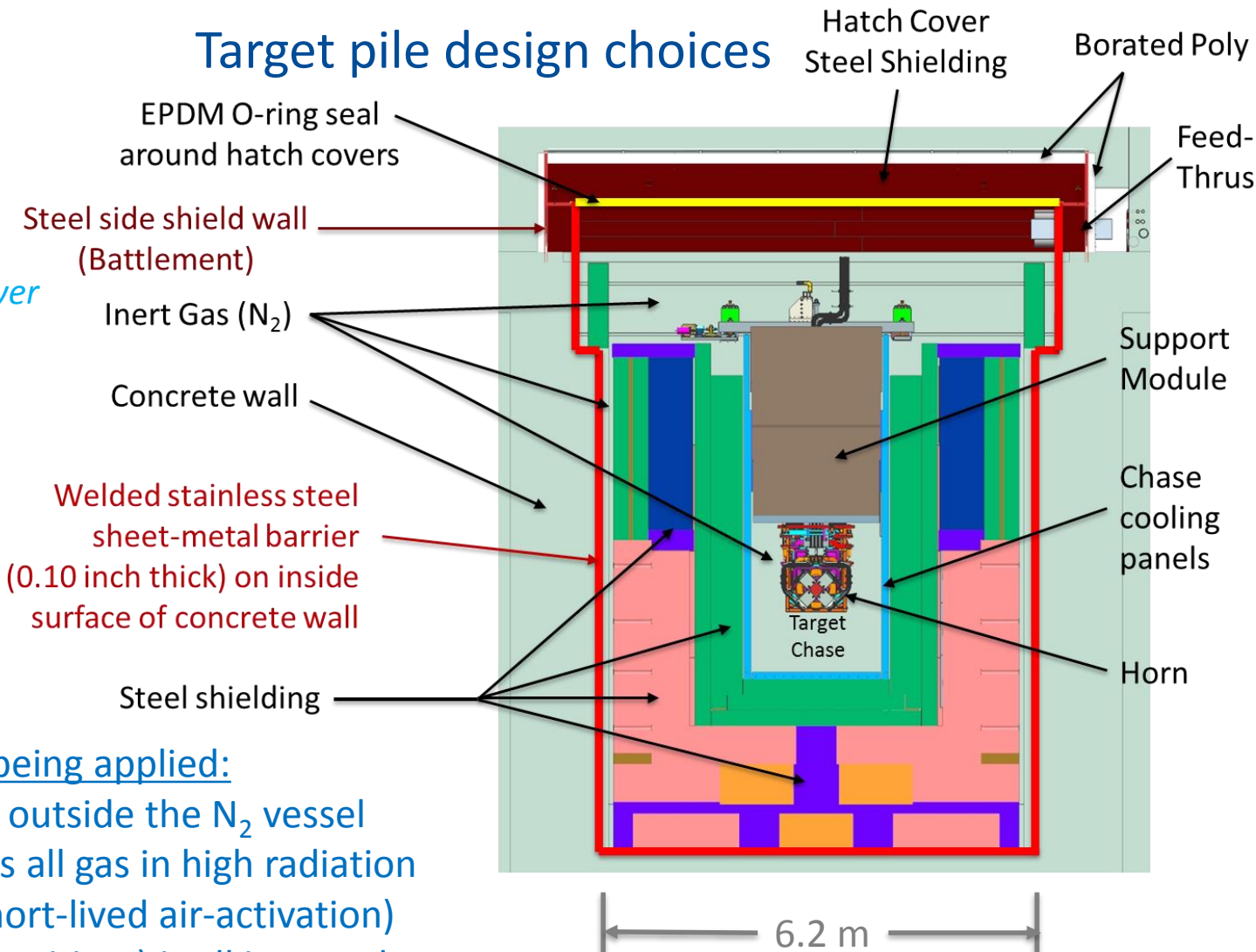
Target pile design choices

Replaceable water cooling panels are used for innermost steel layer

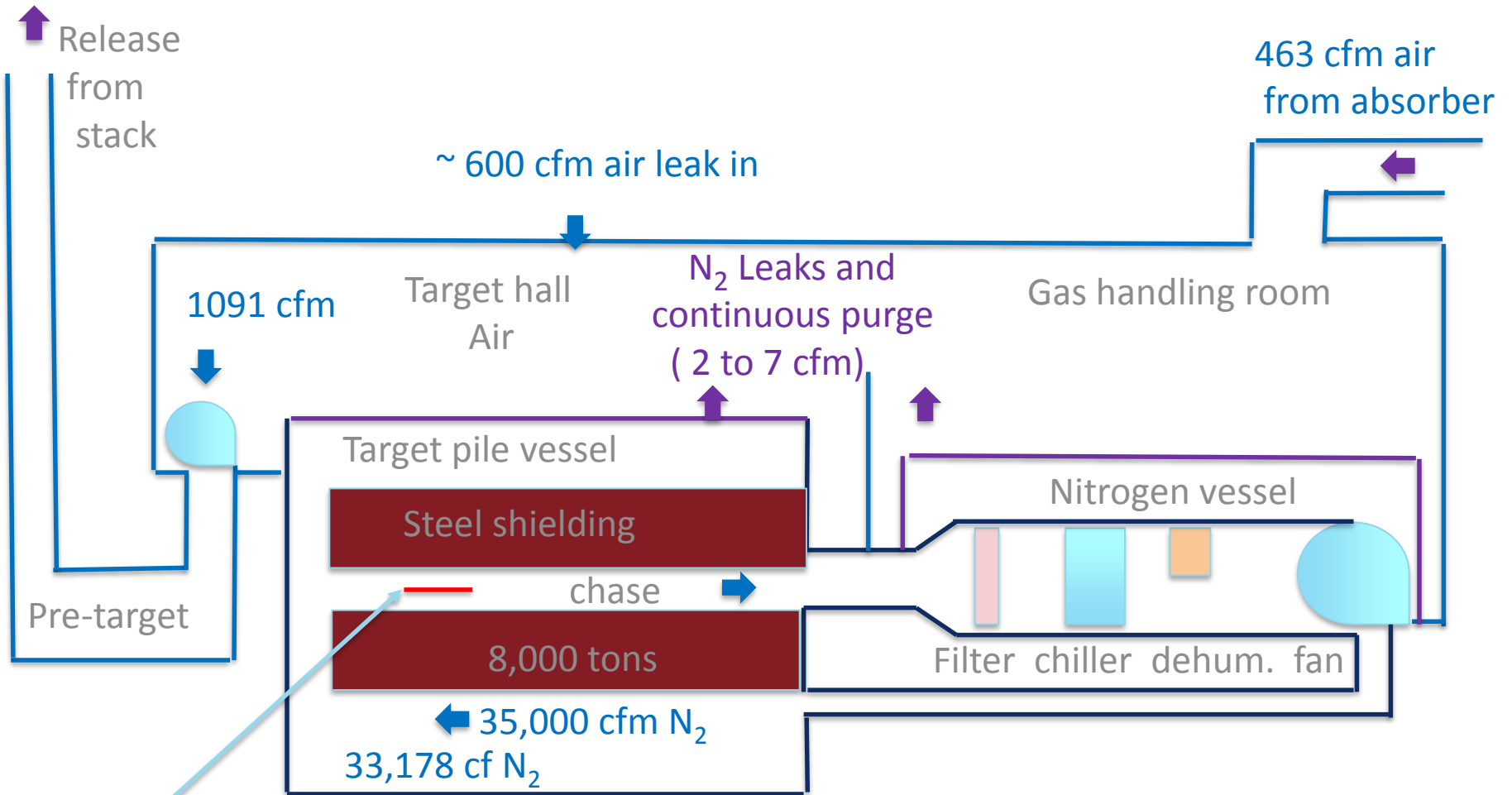
Bulk shielding is cooled by 35,000 ft³/minute flow of N₂ gas

Lessons learned being applied:

- Concrete is all outside the N₂ vessel
- Vessel includes all gas in high radiation (containing short-lived air-activation)
- Steel (emitting tritium) is all in vessel
- Continuously purge tritium by slow N₂ release (1 to 7 cfm)



LBNF Target hall ventilation, during operation



Target, the source of radiation that produces Tritium on inner layers of steel

First model iteration

– assume **equilibrium**, locate possible rate limiting steps

The steel blocks need thin layer of water – (OH layer possible as base)

- Nishikawa et al, Journal of Nuclear Materials 277 (2000) 99-105 “Tritium trapping capacity on metal surface” subdivides surface water as
 - Structural water
 - Chemically bound water
 - Physical water – most easily exchanged with gas
- The formula they provide for physical water is calculated from partial pressure of H_2O , temperature, and a constant that depends on the metal surface
 - In the range of 20 C to 100 C for their materials this is around a few E-4 mole/m²
 - I put 1e-4 mole/m² in the model to check if this essentially covers the surface, or we end up with partially dry surface -> looks fully covered to me
 - Note: used input H_2O pressure 101 Pa (i.e. 1 atm at 1000 ppm H_2O)
 - Note: very generic rough; our blue blocks and water cooling panels may vary
- To get *order of magnitude* estimate of tritium stored in the water on surface of steel, took **WAG** of 5000 m² of steel surface, 3e-4 mole/m² of water: get ~ 1 mCi

Table 1
Equations obtained for water of various type on metal surface

Aluminum	Physical adsorption	$q_{ad,p} = \frac{5.4 \times 10^{-7} P \exp(5.56 \text{ kJ}/RT)}{1 + 1.1 \times 10^{-2} P}$
	Chemical adsorption	$q_{ad,C} = 4.15 \times 10^{-9} P^{1/2} \exp\left(\frac{19.5 \text{ kJ}}{RT}\right)$
	Structural water	$q_{str} = 6.2 \times 10^{-5} + \frac{3.9 \times 10^{-30} \exp(180 \text{ kJ}/RT)}{1 + 8.9 \times 10^{-26} \exp(180 \text{ kJ}/RT)}$
Copper	Physical adsorption	$q_{ad,p} = \frac{4.72 \times 10^{-8} P \exp(9.7 \text{ kJ}/RT)}{1 + 5.0 \times 10^{-3} P}$
	Chemical adsorption	$q_{ad,C} = 0$
	Structural water	$q_{str} = \frac{2.1 \times 10^{-10} \exp(35 \text{ kJ}/RT)}{1 + 8.0 \times 10^{-6} \exp(35 \text{ kJ}/RT)}$
304SS	Physical adsorption	$q_{ad,p} = \frac{2.82 \times 10^{-6} P \exp(2.32 \text{ kJ}/RT)}{1 + 4.5 \times 10^{-3} P} \alpha_f$
	Chemical adsorption	$q_{ad,C} = \frac{1.93 \times 10^{-23} P^{1/2} \exp(121 \text{ kJ}/RT)}{1 + 8.2 \times 10^{-19} \exp(121 \text{ kJ}/RT)} \alpha_f$
	Structural water	$q_{str} = \frac{3.06 \times 10^{-8} \exp(27 \text{ kJ}/RT)}{1 + 3.6 \times 10^{-4} \exp(27 \text{ kJ}/RT)} \alpha_f$

First model iteration

– assume **equilibrium**, locate possible rate limiting steps

Molecular exchange between humidity and surface layer

- I used our NuMI experience with tritium leaching out of the air when it went down the decay pipe passageway to WAG how fast the humidity exchanges with a water layer. The time constant for NuMI was $\ll 1$ hr. I cross-checked this with some literature where people were doping tritium onto samples, and removing tritium from samples. It needs to be documented in a more refined manor, but this exchange rate appears to be fast enough to not be rate-limiting in this version of the calculation. Recall time scale of the diffusion in steel to water layer is days.

First model iteration

- assume **equilibrium**, locate possible rate limiting steps
-

Exchange rate between surface water and steel

- My impression from those same studies of tritium is the transport between water layer and near-surface steel is fairly fast, and should not be rate-limiting, but need better documentation.
- Note there are barriers, like an oxide layer on stainless steel, that can significantly slow down the transport. A cautionary statement in fusion research is “barrier layers tend to break down in radiation environment”.
- The T in the steel can basically trade places with an H in the water layer

- Is the layer of black iron-oxide at T2K on the vessel walls why there seems to be significantly less release of tritium at T2K than NuMI?

First model iteration

- assume **equilibrium**, locate possible rate limiting steps
-

Equilibrium between water layer and steel

- Assume ratio of tritium densities in the surface water to steel is the ratio of Tritium Solubilities in water to steel
- There is a measured solubility for tritium in stainless steel, but I have not located a paper documenting solubility in more normal steel. Using some web information, assuming it is not hugely different between tritium and hydrogen, I used
 - hydrogen solubility estimate of $0.3 \text{ (mole H/m}^3\text{)}/\sqrt{P_{\text{atm}}}$, (*ref Ispat*)
 - which given a water solubility estimate of $4\text{E}4 \text{ (mole H/m}^3\text{)}/\sqrt{P_{\text{atm}}}$ (*very rough extrapolation from Sharpe et al “Tritium migration to the surfaces of stainless-steel, aluminum 6061, and oxygen free high-conductivity copper”*)
 - and an assumption that both solubilities are at 1 atm,
 - gives a **solubility ratio of 1.3E5**
- This is a very rough number now, but use it to do some calculations.
- In humidify/dehumidify scheme, steel builds up concentration of equilibrium value in a few minutes, so this transfer should not be rate limiting

First model iteration

8

– assume **equilibrium**, locate possible rate limiting steps

Steel near target

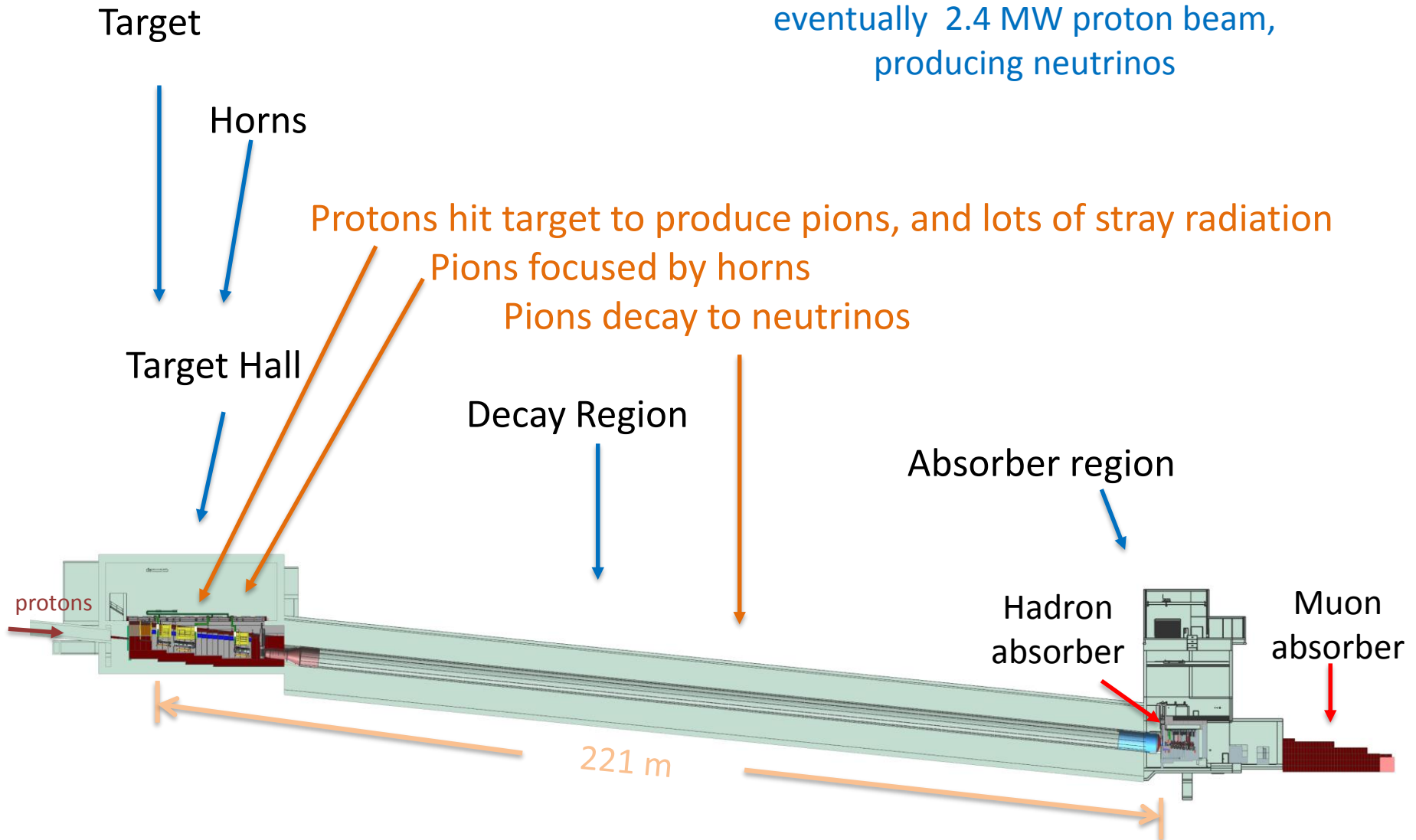
- 18% of produced Tritium is in the volume of the cooling panels nearest the target
 - 2 sides x 7.32m x 2.24 m x 0.1016 m= 3.33 m³ -> 26 metric tons
 - ~ 1 mCi if entire block was at equilibrium
- At that rate, the time constant of production build-up toward “equilibrium” is a few minutes for those panels

Conclusion: for tritium scrubbing with humidity as shown (1 ppt H₂O in N₂), diffusion through steel is the rate limiting effect, and that time scale is days for the highly irradiated cooling panels.

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- BACKUP

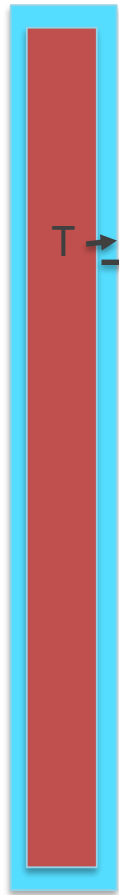
LBNF neutrino beam line

1.2 MW of proton beam power initially,
eventually 2.4 MW proton beam,
producing neutrinos



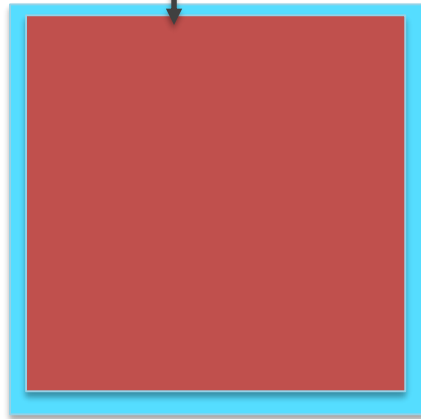
Environment for transport

Tritium **produced** mostly in steel closest to target
Steel has **microscopic water layer**



N2 gas at ~ 1atm, 33,178 cf
with 0.1% water molecules

8,000 tons of bulk steel shielding
also has microscopic water layer



Stainless
Steel
Vessel
wall

2 cfm
leak

Air
flowing
by at
1091 cfm

Tritium Basics

- Tritium (^3H) decays to ^3He by beta emission, beta has average energy 5.69 keV and maximum 18.6 keV.
- Half-life is 12.32 years.
- Highly mobile, has high diffusion coefficient through metals
- If there is water around, Tritium mostly ends up as HTO
- The beta will not make it through your skin
- Harmful if ingested by drinking or breathing HTO, or absorption through skin
- Tritium released as HT has significantly less human uptake than HTO, so trying to shift release in that direction would be consistent with ALARA

- For neutrino beamline, Tritium is nearly all produced by spallation in the shielding

Tritium release limits for Fermilab

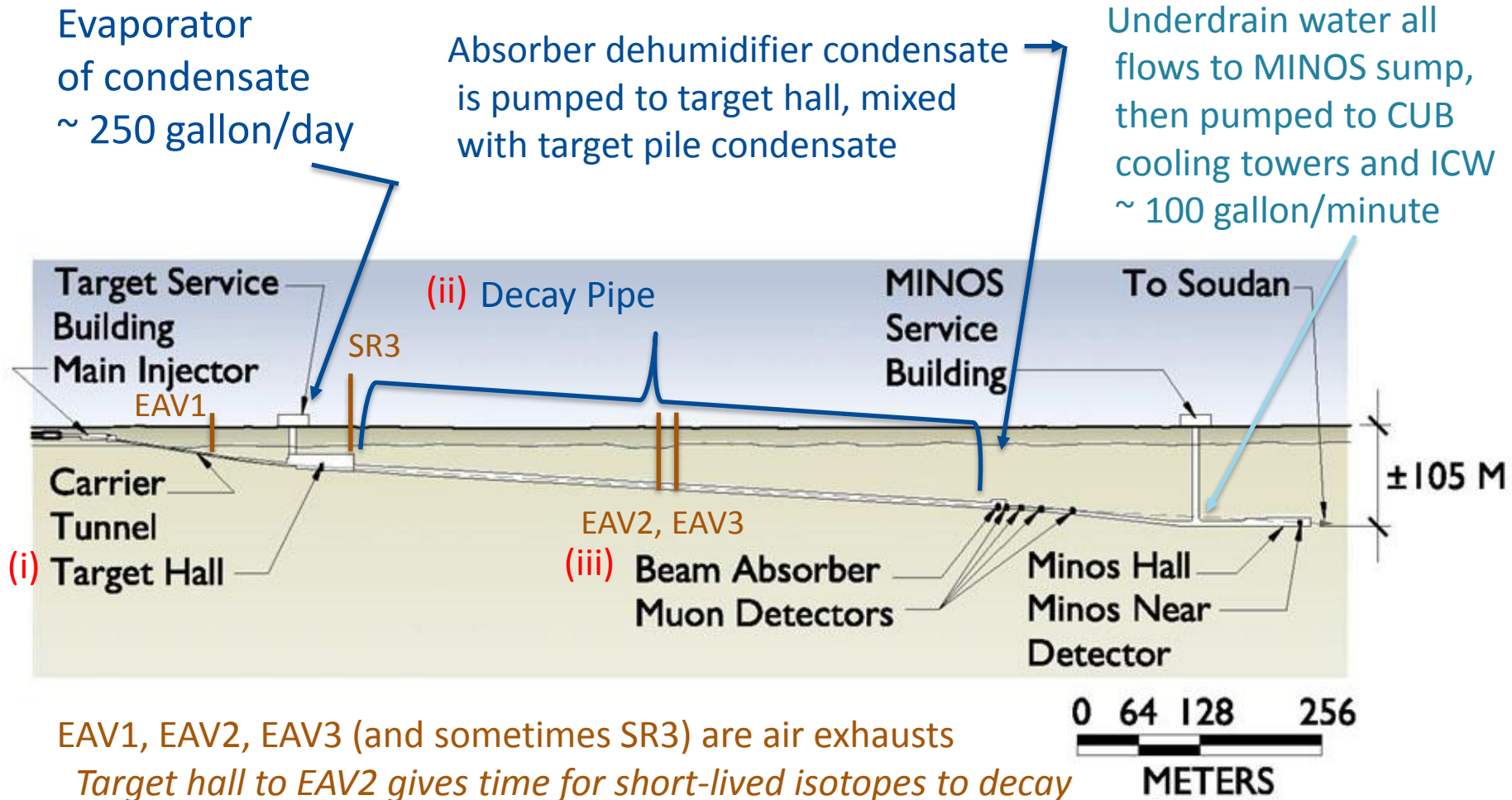
- Sanitary Sewers
 - 5 Ci/yr
 - 9,500 pCi/ml
- Surface water
 - 1,900 pCi/ml
- Class 1 groundwater
 - Non-degradation (taken as “detectable” = 1 pCi/ml)
 - (aside: general drinking water limit is 20 pCi/ml)
- Dose through air to public (Maximally Exposed Offsite Individual)
 - 0.1 mrem in a year (summed with all other FNAL air release radiation)
 - Roughly 3000 Ci of HTO release to air would give 0.1 mrem to MEOI
- Dose through air to workers
 - Defined Air Concentration on next slide

Tritium release experience at NuMI

In recent years, measured release of NuMI tritium:

- 90% evaporated up roof stack, air exhaust
- 7% through underdrains to sump, then used as cooling water, eventually evaporates from cooling ponds
- 2% collected from target cooling water, barreled and disposed off-site
- 1% collected from horn water cooling system, barreled and disposed off-site
- Small amounts collected in Air Handling Units; to sewer for now
- Tiny amounts blow down on ground; it re-evaporates naturally

NuMI, as background for tritium discussion



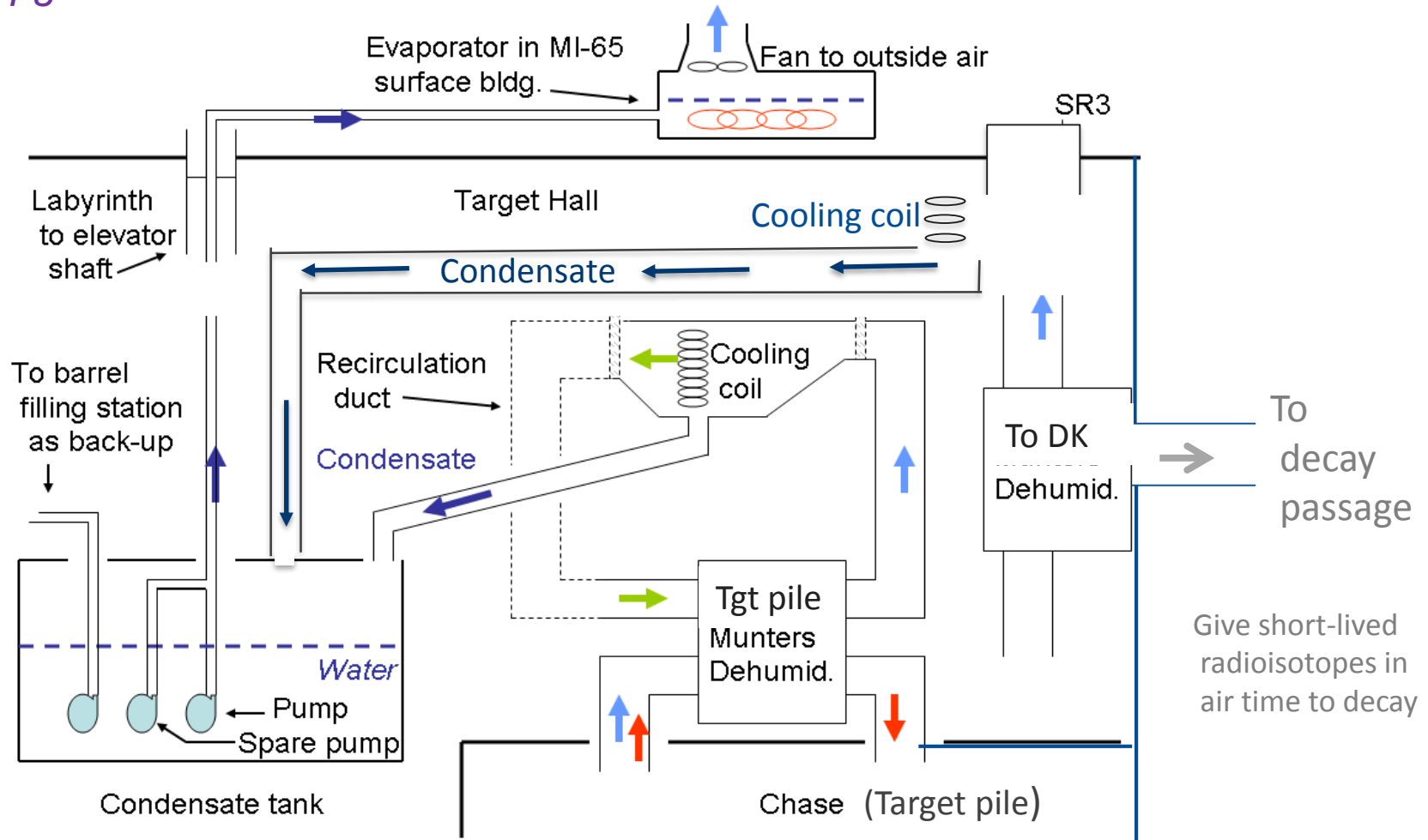
EAV1, EAV2, EAV3 (and sometimes SR3) are air exhausts

Target hall to EAV2 gives time for short-lived isotopes to decay

Tritium producing particle shower power is deposited ~ 1/3 in each of (i) target hall, (ii) decay pipe, and (iii) absorber at end of decay pipe

Schematic of main NuMI Tritium release path

All of this dehumidification and evaporation equipment was added to NuMI as upgrades to deal with Tritium.



NuMI Tritium Production and Release

MARS Ci/10 ²⁰ POT	Monte Carlo Produced in
24	target pile steel
11	decay pipe concrete
2.5	decay pipe steel
0.22	chase air
0.03	decay pipe helium
??	absorber
1.3	horns
1.7 - 4.2	Target
41 - 44	TOTAL

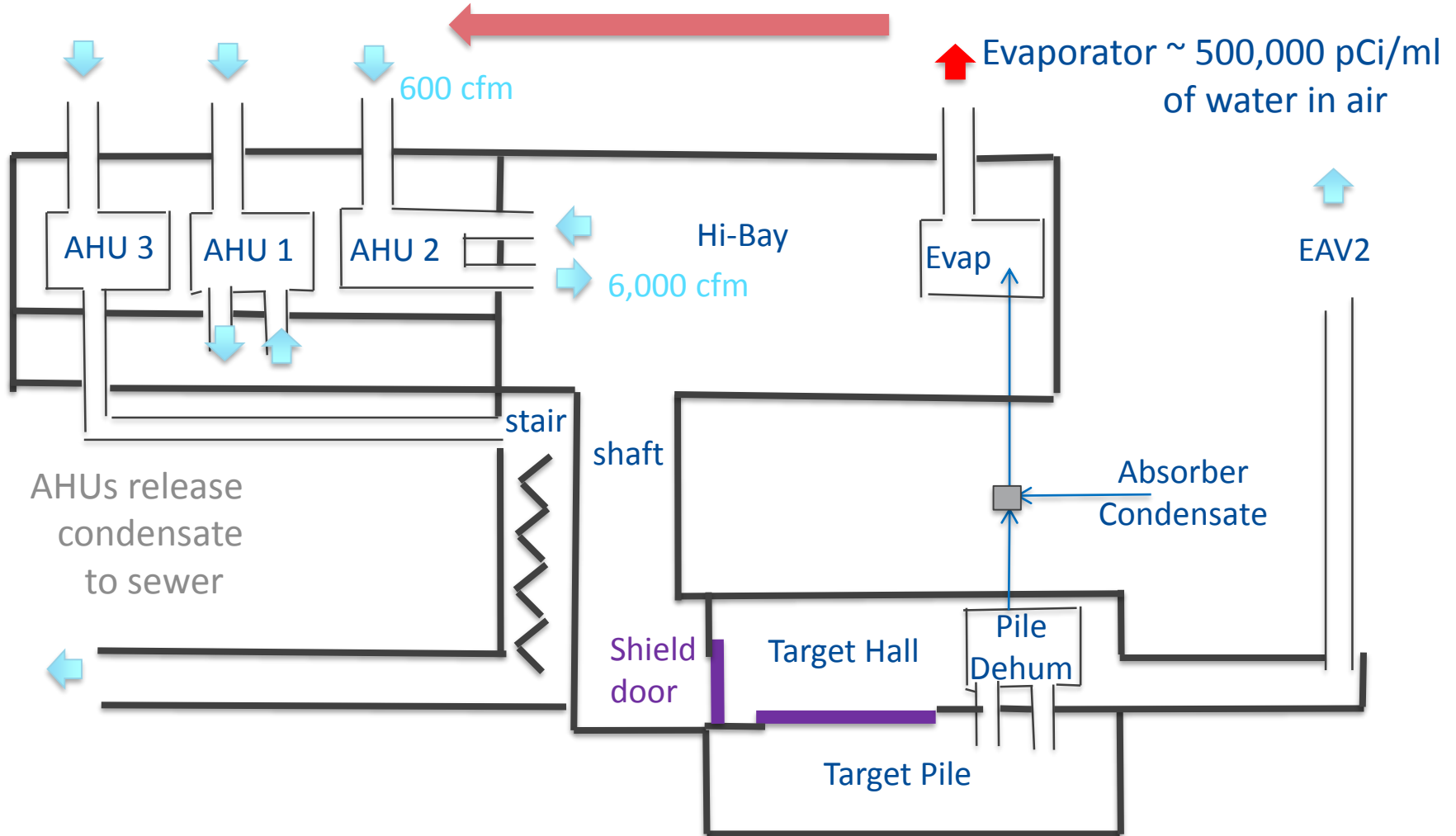
MARS Monte Carlo
of Tritium production
in NuMI

6 Ci/10²⁰ POT Measured Release 2008 – 2011 at 300 kW
 ~ 26 Ci/10²⁰ POT Measured Release 2016 at 520 kW

*Release jumped non-linearly with beam power as steel temperature ~ 100 C,
 and is approximately equal to M.C. production rate in steel components*

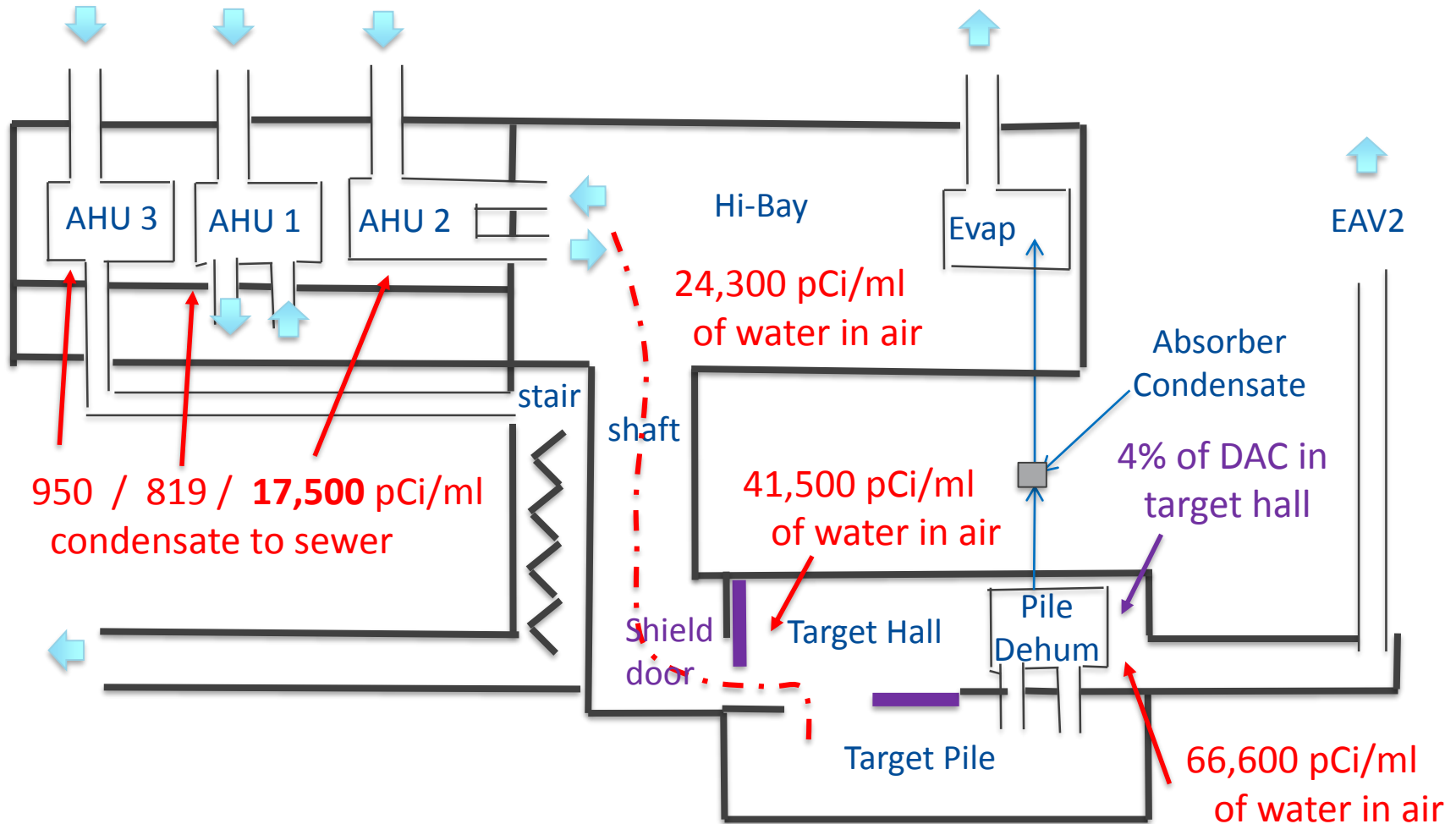
During NuMI operation, MI-65 ventilation

AHU condensate up to 1,720 pCi/ml, AHU1, AHU2, AHU3 fairly similar



9/11/2018 short circuit to AHU during access

Target pile open, target hall shield door open, pile dehumidifier off



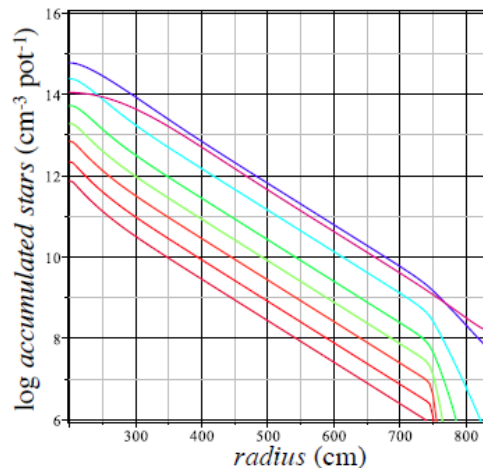
Decay Pipe Tritium Diffusion Study

(taken from B. Lundberg, May 2013, LBNE-doc 7254)

- α (Diff. const.) for tritium in concrete has been estimated from data from nuclear power industry
- Varies from ~ 0.1 to $20 \text{ mm}^2 \text{ d}^{-1}$
- In what follows, use maximum: $\alpha = 20 \text{ mm}^2 \text{ d}^{-1}$

- Assume :

- 700 kW for first 5 y
- 2 MW for next 25 y
- OFF after 30 y



Plot : $\log("C")$ vs radius

@ t =

1, 30, 100, 300, 1000, 3000, 10950
and 18500 days

Purple curve after 20yr
shutdown

- After 30yr operation: $\sim 0.24 \text{ mCi y}^{-1}$ ← Leak out from concrete
- Same analysis gives $\sim 9 \text{ Ci y}^{-1}$ inner conc. ← Leak into decay pipe N_2 cooling annulus, and purged