NAMP Young Investigators Series: Radioecology

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National Analytical Management Program (NAMP)
U.S. Department of Energy Carlsbad Field Office
TRAINING AND EDUCATION SUBCOMMITTEE

Meet the Presenters…

Dawn Montgomery is a Ph.D. student in Environmental Engineering and Earth Sciences at Clemson University working with Drs. Nicole Martinez and Brian Powell. Her academic and research emphasis is within the environmental health physics and radioecology disciplines. Currently, her research is centered on plant uptake of various radioactive contaminants and the associated role that plants have on the transport of those contaminants in the environment. Additionally, she is interested in dosimetric modeling of non-human biota and has developed several phantoms for the grass species used in her uptake experiments and stylized phantoms for an adult duck and a duckling. Montgomery earned a B.S. in Applied Mathematics from North Carolina State University in 2005. She expects to graduate in December 2019.

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Presentation: **Uptake and dosimetric modeling of ^{99}Tc, ^{133}Cs, ^{237}Np, and U in a native grass**

Uptake and dosimetric modeling of ^{99}Tc, ^{133}Cs, ^{237}Np, and U in a native grass

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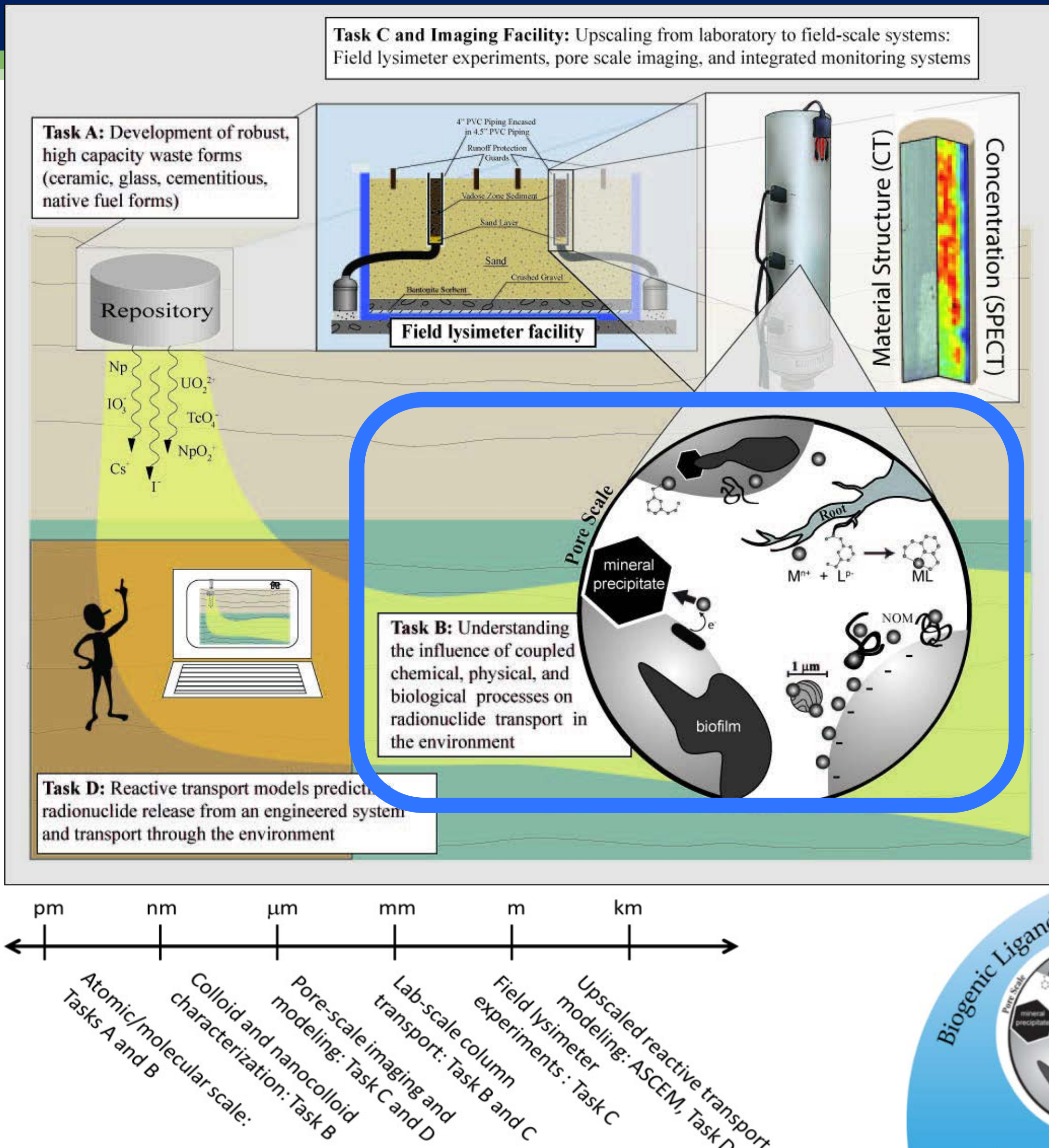
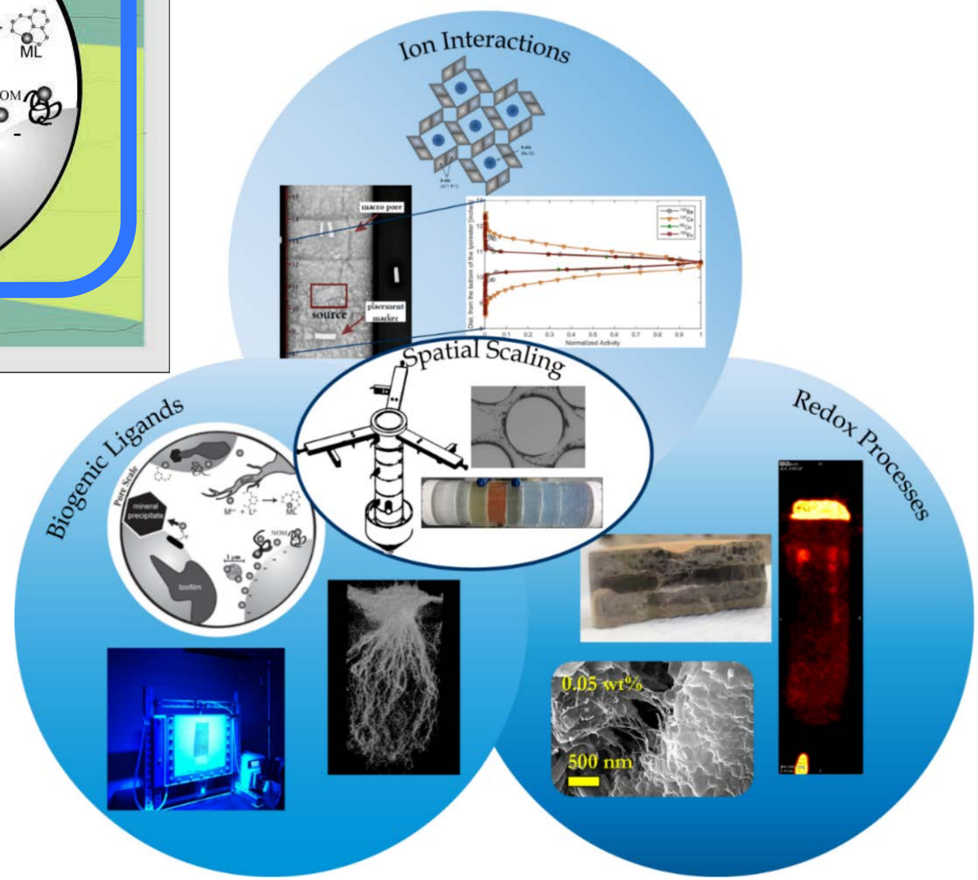
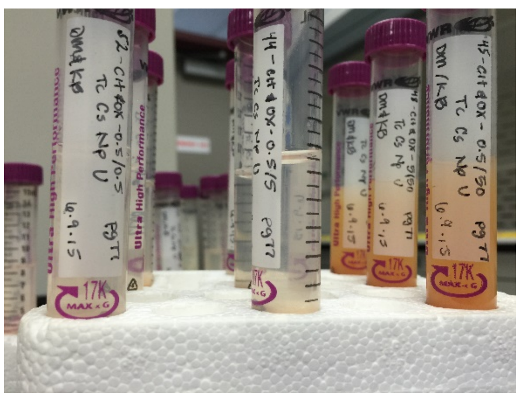
Multiple Scales and Processes





Soil Sorption → Hydroponics → Plant-Soil Columns

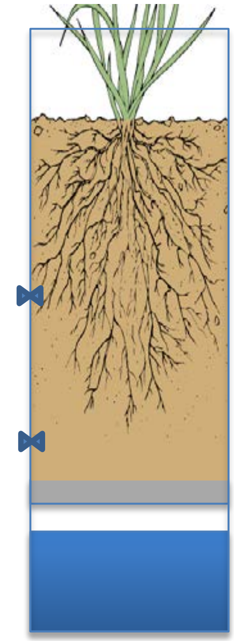


Baseline K_D and effects due to ligands

Montgomery et al 2017

Uptake and dosimetry

Montgomery et al 2018



Column block diagram

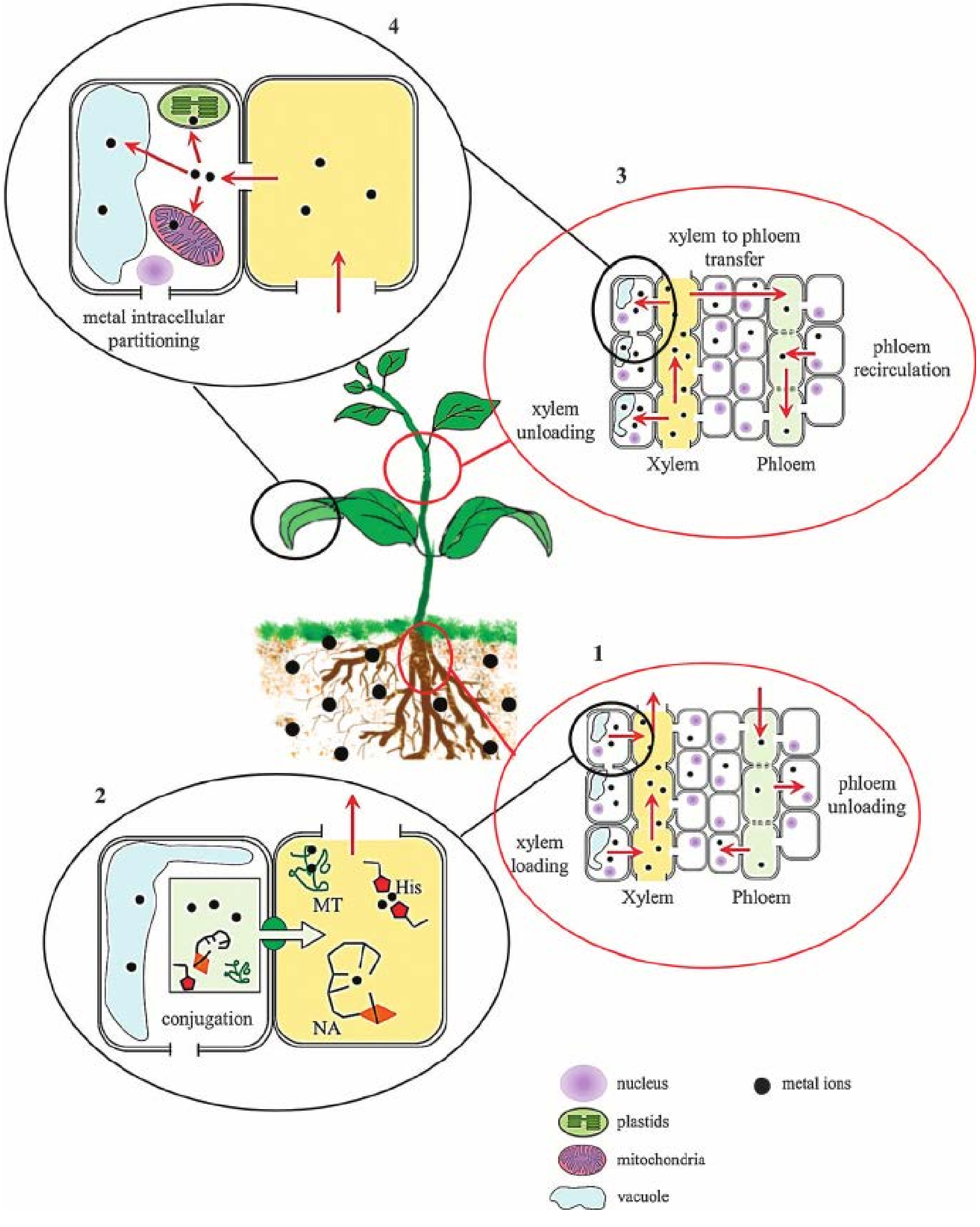
Root structure image credit: http://science.howstuffworks.com/dictionary/plant-terms/root-info.htm

Combining soil sorption and uptake

Nuclides of concern

- ^{99}Tc, ^{237}Np, ^{135}Cs/^{137}Cs and U
  - Long-lived nuclides present in nuclear waste and potential spent fuel recycling streams
  - Expected to be mobile and potentially hazardous to human and environmental health
  - Complex biogeochemical behavior with different sorption mechanisms, redox activity, solubility, overall mobility, bioavailability, and analogous nature to plant nutrients
- Technetium(VII) as pertechnetate, TcO_4^-
  - Oxyanion, very weakly complexing, highly mobile
- Cesium, Cs^+
- Neptunium(V), NpO_2^+
- Uranium(VI), UO_2^{2+}

Plant Uptake Studies: Hydroponics & Columns



Main route followed by metal elements in plants

Metal ions are loaded into the xylem as free ions or conjugates.

Moving with water, ions are delivered to the shoot.

In the shoot, metal ions are subcellularly partitioned or detoxified.

A small portion of ions can cycled back to the root tissue.

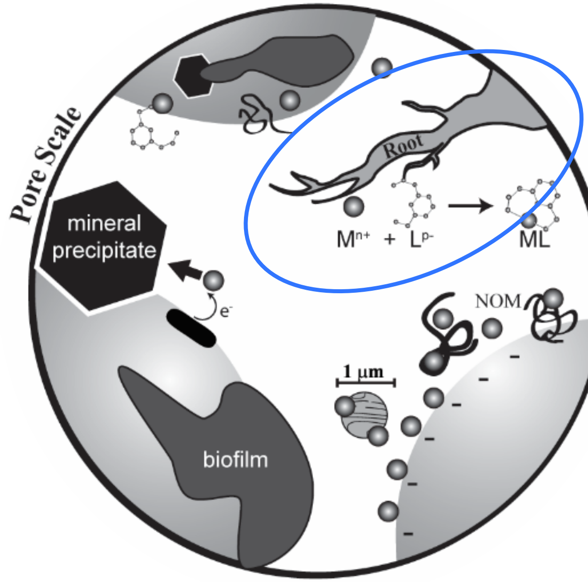
DalCorso et al., 2014

Plant Uptake Motivation & Objectives

- Insight needed into potential plant mediated mechanisms of observed upward migration of radionuclides in soil columns.
  - *A. virginicus* is a common ground covering in the Southeastern US (and at Savannah River Site)
  - The suite of nuclides considered encompass a wide range of biogeochemical behavior

www.perennialfarmmarketplace.com

- Evaluate the propensity of *Andropogon Virginicus* to take up ^{99}Tc, ^{133}Cs (stable analog for ^{137}Cs), ^{237}Np and ^{238}U
  - Interested in the effect of plant root exudates on the uptake and mobilization of radionuclides.
  - Working towards understanding radionuclide mobilization in soils as influenced by plant root foraging activities and microbial associations.



Hydroponic Experimental Set-Up

Acclimation

- 1 week laboratory acclimation
  - 16 plants (4 groups of 4)
  - 12 hour light cycle
  - Hoagland nutrient solution

Spiked Hydroponic Solution

- 3 spiked groups, 1 control (non-rad)
  - ~75 ppb ^{99}Tc
  - ~10 ppb ^{237}Np, ^{238}U, & ^{133}Cs

Harvest

- Harvest at 1, 3, and 5 days
- Roots rinsed and shoots separated
- One plant per group selected for autoradiography

Drying and Digestion

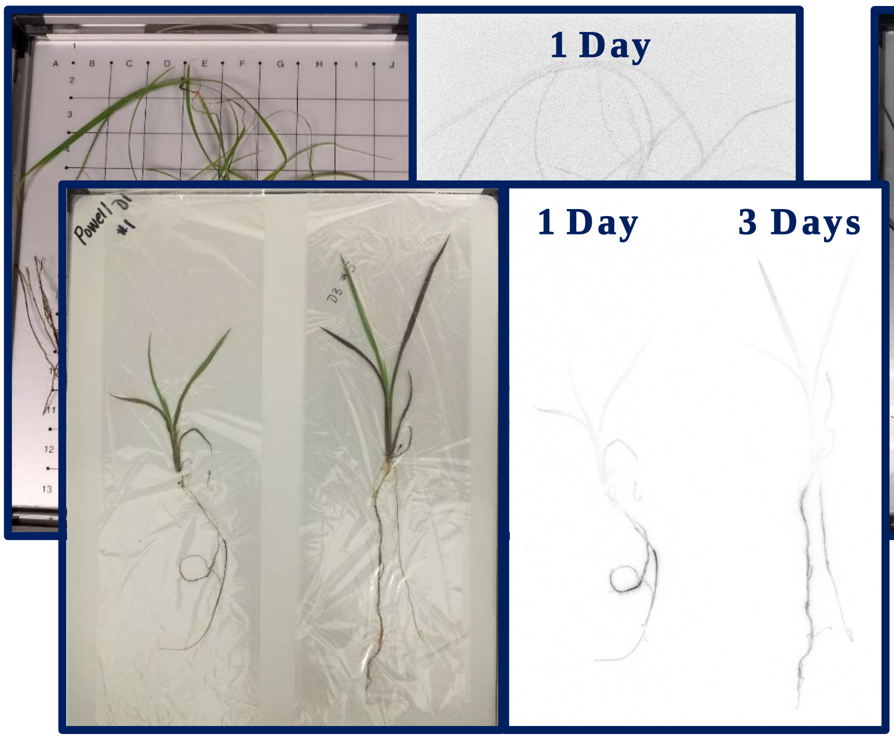
- Dried at 50 °C to a constant mass
- Bench top digestion with HNO_3/H_2O_2 (EPA 3050B)

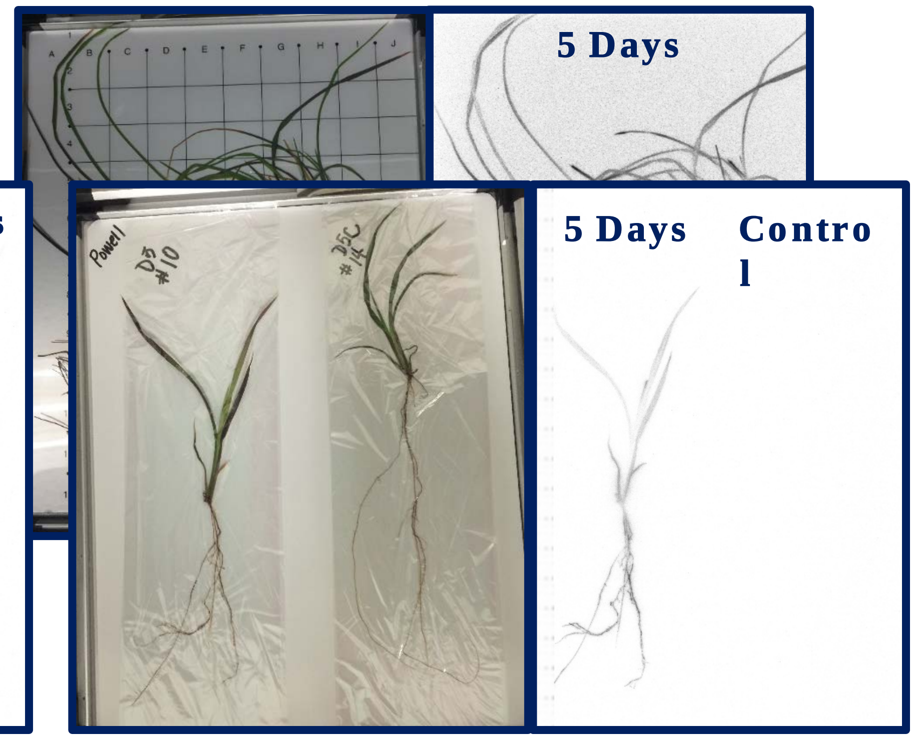
Analysis

- Analysis of hydroponic solution and plant digestate via ICP-MS and LSC

Autoradiography of ^{99}Tc uptake

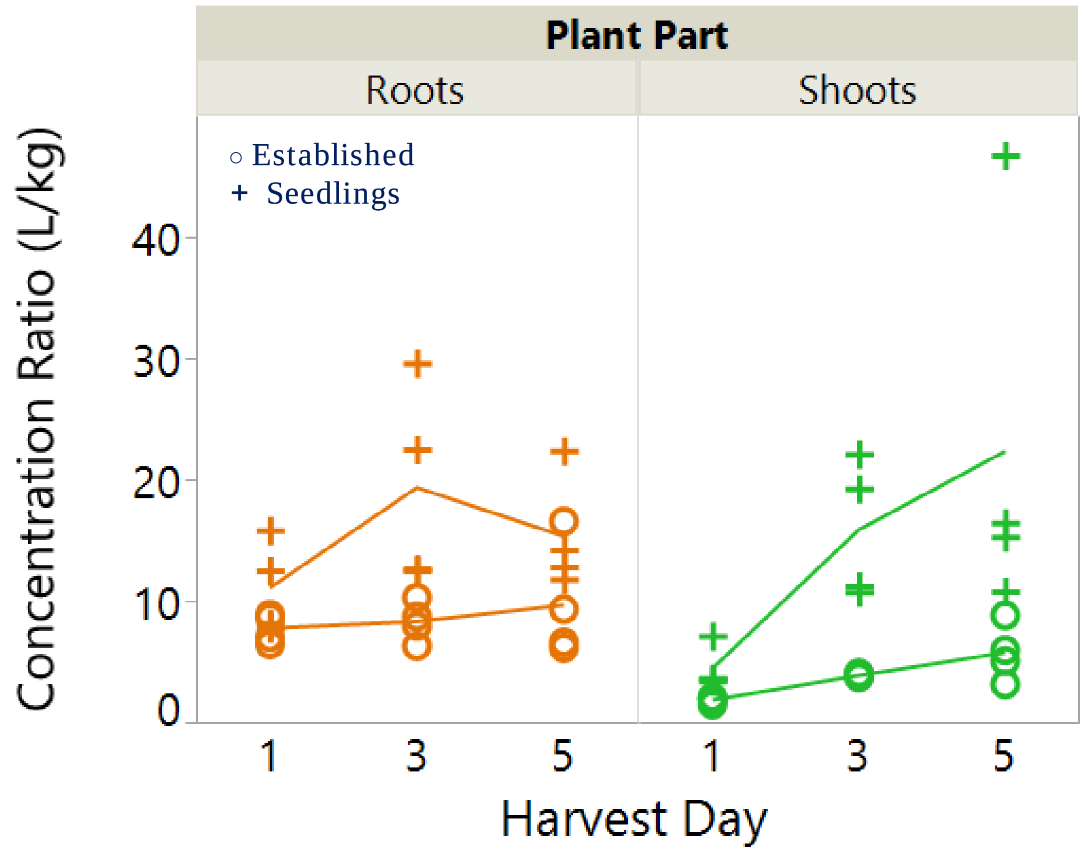
- Intensity positively correlated with growth time
- Root-shoot connection and/or shoot tips appeared to have higher ^{99}Tc content
- Seedlings/smaller plants used to investigate differences in uptake





Uptake – Concentration Ratios, ^{99}Tc

- TcO_4^- proposed to be associated with uptake mechanisms for:
  - SO_4^{-2}, MoO_4^{-2}, SeO_4^{-2}, NO_3^-, Cl^-, PO_4^{-3}
- Plant part is significant for established plants
- Harvest day is significant for seedlings
- Plant age (experiment) is significant



ANOVA analysis P-Values

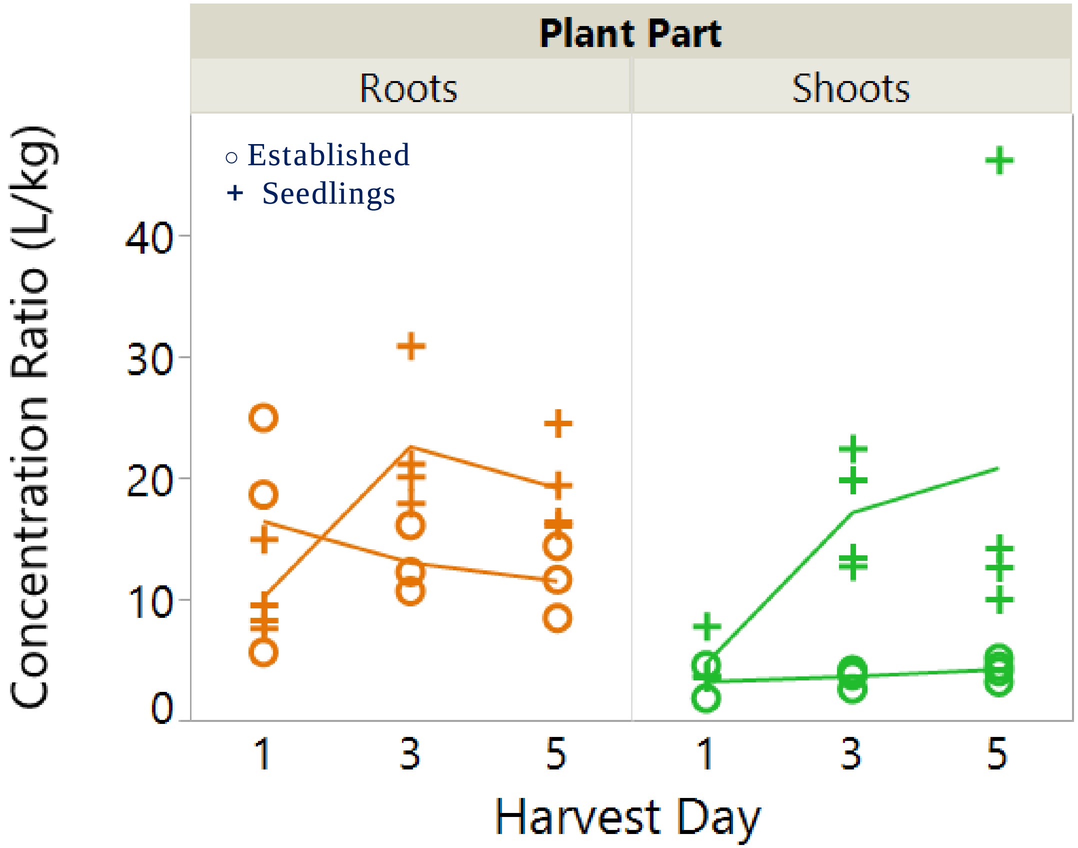
Factor	All Plants	Established	Seedlings
Plant Part	0.108	<0.001	0.763
Harvest Day	0.006	0.071	0.030
Experiment	<0.001	---	---

$$CR = \frac{C_{plant\ part}}{C_{HP\ solution}}$$

Bennett and Willey, 2003; Cataldo et al., 1983; Robertson et al., 2003

Uptake – Concentration Ratios, ^{133}Cs

- Analogous to K^+
- Plant part is significant for established plants
- Harvest day is significant for seedlings
- Plant age (experiment) is significant



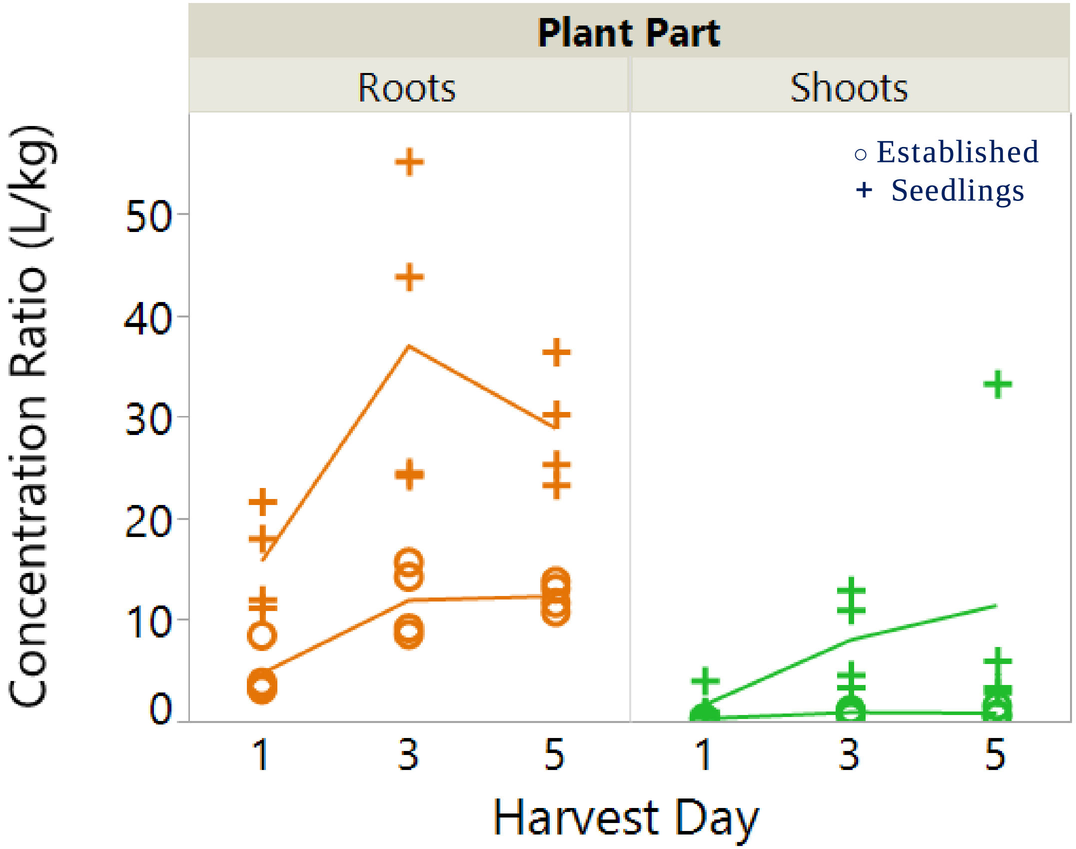
ANOVA analysis P-Values

Factor	All Plants	Established	Seedlings
Plant Part	0.004	<0.001	0.355
Harvest Day	0.075	0.738	0.007
Experiment	0.002	---	---

$$CR = \frac{C_{plant\ part}}{C_{HP\ solution}}$$

Uptake – Concentration Ratios, ^{237}Np

- No nutrient analog
- Plant part and harvest day are significant
- Plant age (experiment) is significant



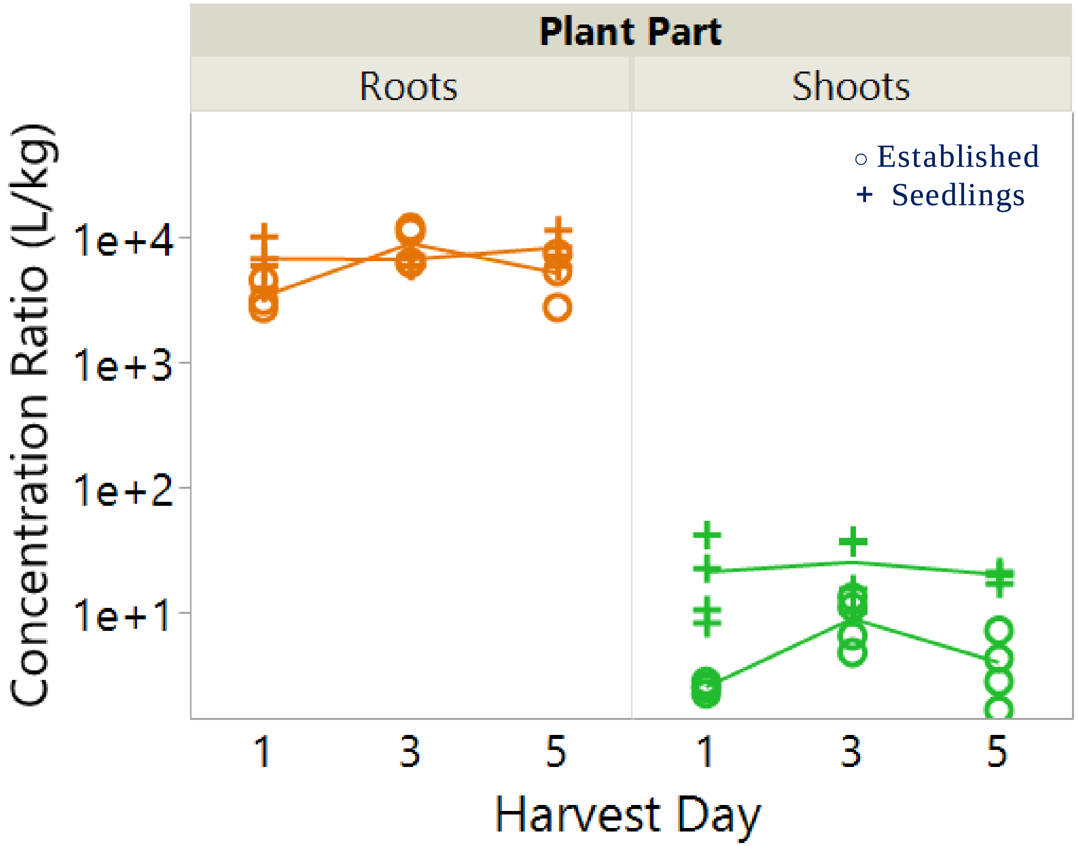
ANOVA analysis P-Values

Factor	All Plants	Established	Seedlings
Plant Part	<0.001	<0.001	<0.001
Harvest Day	0.001	<0.001	0.020
Experiment	<0.001	---	---

$$CR = \frac{C_{plant\ part}}{C_{HP\ solution}}$$

Uptake – Concentration Ratios, ^{238}U

- No nutrient analog
- Plant part is significant for both
- Harvest day is significant for established plants
- Plant age (experiment) is **not** significant



ANOVA analysis P-Values

Factor	All Plants	Established	Seedlings
Plant Part	<0.001	<0.001	<0.001
Harvest Day	0.046	0.006	0.480
Experiment	0.105	---	---

$$CR = \frac{C_{plant\ part}}{C_{HP\ solution}}$$

Major findings: Hydroponic Studies

- Many radionuclides are readily incorporated into plants and the partitioning appears to be related to solubility, complexation affinity and similarity to plant nutrients.
- Concentration ratios vary by radionuclide, time of exposure, and plant part.
- Roots generally have higher concentration ratios, particularly for analytes without nutrient analogs, likely due to radionuclide sorption to the root vice true uptake.
- Plant size/age seems to have an influence on uptake

Soil Column Experiments (In Progress)

Transplant & Acclimation

- 50/50 SRS soil/sand (25 cm)
- *A. virginicus* seedlings transplanted (3-5 leaves)
- Irrigated (2-3 d), effluent collected

Spike Introduction

- Injection via Rhizon CSS© sampler at 7.6 cm depth
- ~1000 µg L^{-1} ^{99}Tc
- ~100 µg L^{-1} ^{133}Cs, ^{237}Np, U

Shoot Harvest

- Shoots harvested at 4 weeks
- Columns covered & held at 5 °C until segmentation

Column Segmentation

- 1 cm thick transverse segments
- Separating roots from soil

Digestion & Analysis

- Plant & soil segments dried
- Digestion/leaching (EPA 3050B)
- Analysis via ICP-MS and LSC (^{99}Tc)

Dosimetric Model Development

Dosimetry Motivation & Objectives

- Establishment of appropriate protection standards requires sufficient knowledge of dose effects
  - Although organ-specific screening levels maybe impractical, considering only “whole body” (or above ground plant part) dose rates may not be adequately protective
- Dose determination
  - Direct measurement or robust dosimetric modeling
  - Lack of appropriate models results in occasional controversy: need refined and consistent dosimetric modelling approaches

Jim Beasley at SREL trapping and placing GPS-dosimeter collars on wild pigs (see Hinton et al 2015)

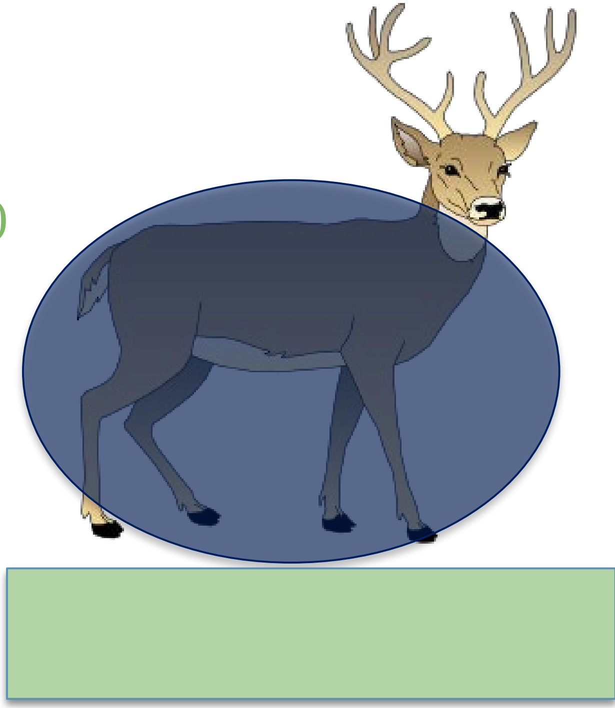
- Describe the development, application, and comparison of dosimetric models utilized in the internal dosimetry of non-human biota
- Ultimate goal of this work is to combine refined dosimetric models with models describing temporal uptake in *A. virginicus* to obtain temporal dose rates

Current methodology

- Certain reference organisms applied to similar species
  - ICRP: 12 “Reference Animals and Plants”
  - ERICA tool: about 40 reference organisms
  - RESRAD-BIOTA: 4 reference organisms

- Dose rates approximated using dose conversion factors (DCFs)
  - Absorbed dose rate per unit activity concentration (μGy d^{-1} per Bq kg^{-1})

- Assumptions:
  - Uniform radionuclide distribution in organism or environmental media (representative conditions)
  - Ellipsoidal body, spherical organs (if included)
  - Decay properties of specific radionuclides
  - Monte Carlo based radiation transport codes



Beresford and Brown 2014, ICRP 108

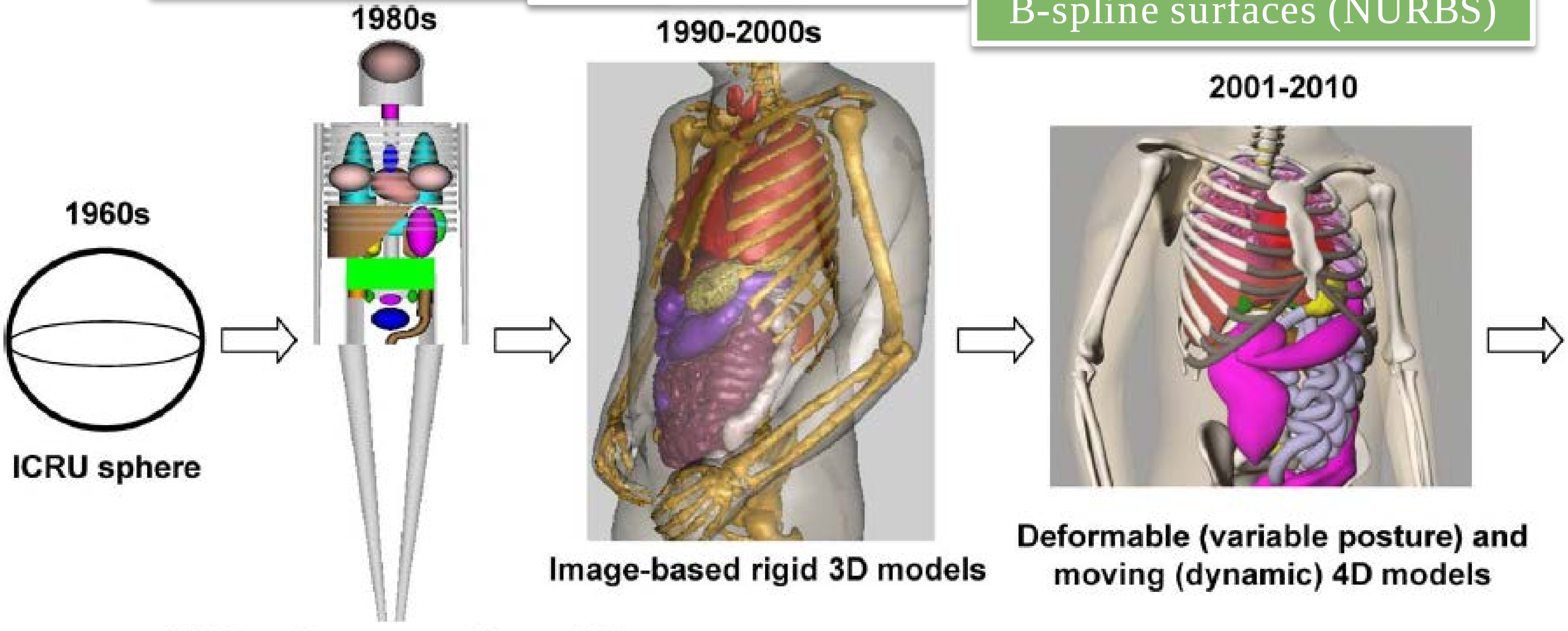
Computational models

- Incorporated into Monte Carlo based radiation transport
  co
  as

Stylized phanto
Combinations of si
equation-based su
for object represen

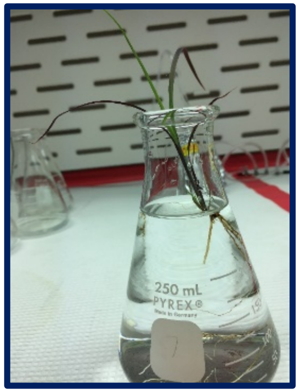
Voxel phantoms
Objects represented by three dimensional voxel matrices

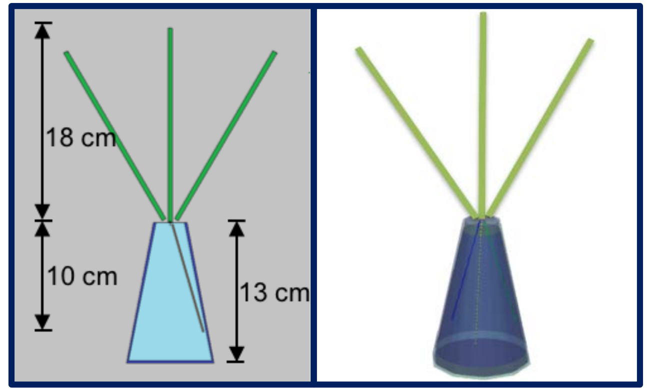
Hybrid phantoms
Combination of stylized and voxel phantoms
Utilize non-uniform rational B-spline surfaces (NURBS)



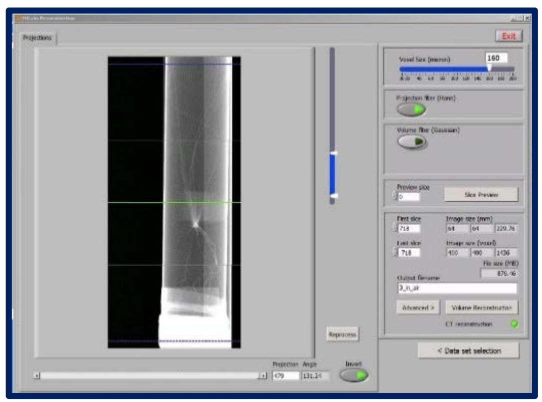
Zaidi and Tsui 2009

Model Development

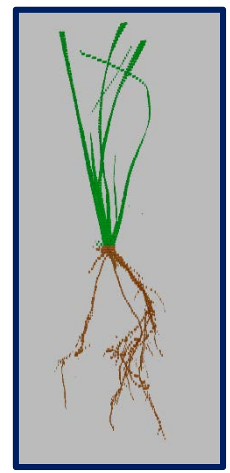




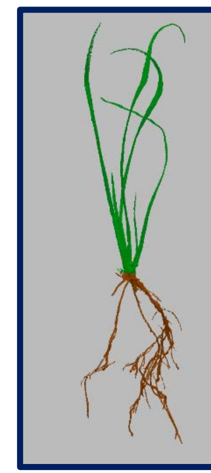
Stylized Phantom



Micro-CT of plant

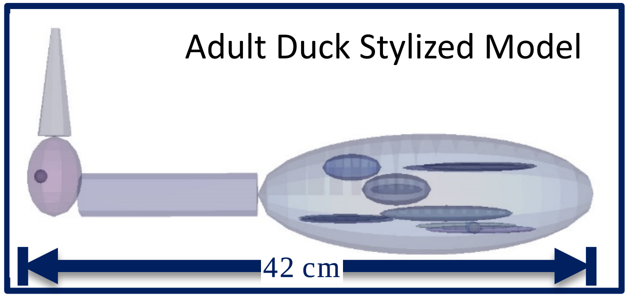


Voxel Phantom: 3D Doctor



Hybrid Phantom: Rhinoceros 3D

Example Non-Human Phantoms

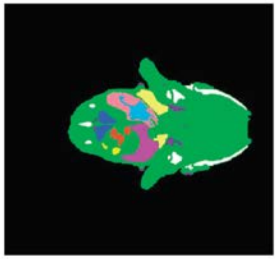


Montgomery et al., 2016
Stylized Duck and Duckling

Some others: Mouse, Rat, Rabbit, Crab, Flat Fish, Honey Bee, Bee Hive, Pine, Arabidopsis thaliana

(a)

Kinase, 2008
Voxel Frog Phantom



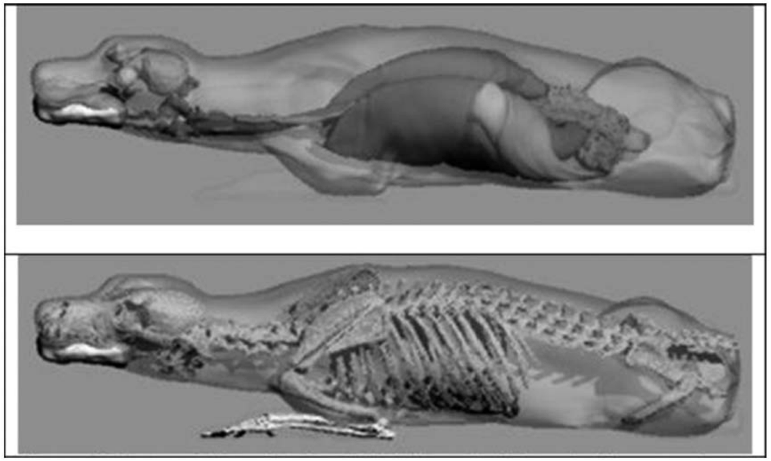
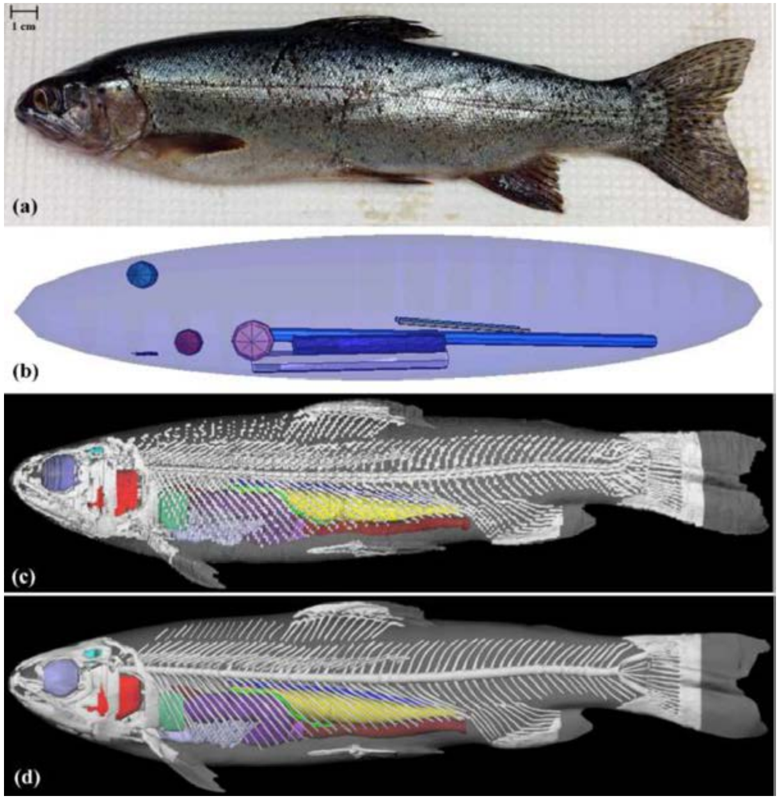


Fig. 1. Image of the male dog NURBS model (Rusty). Above, selected organs; below, selected skeletal segments.

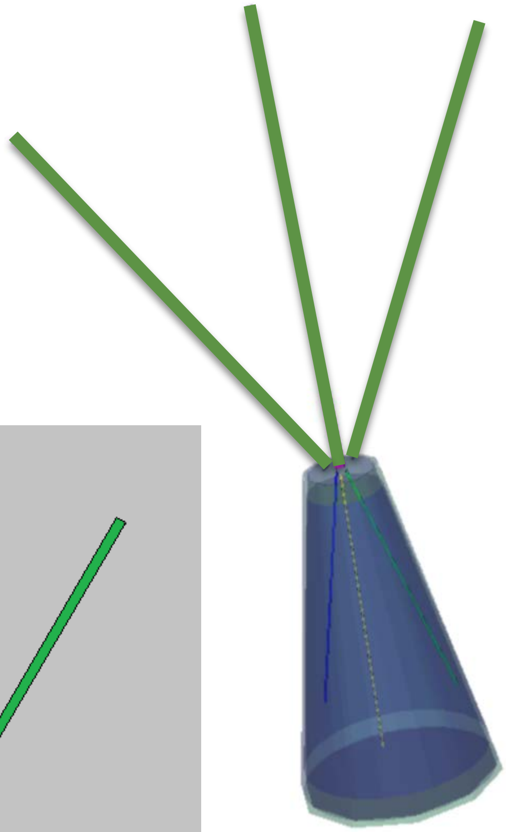
Stabin et al., 2015
Hybrid Beagle Phantoms

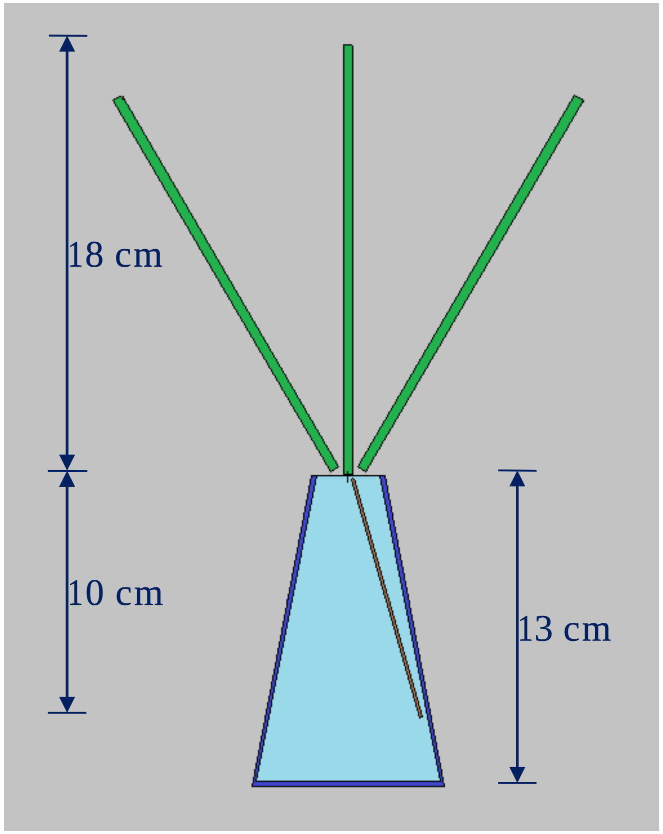


Martinez et al., 2016
Stylized, Voxel, Hybrid Trout

Stylized Phantom

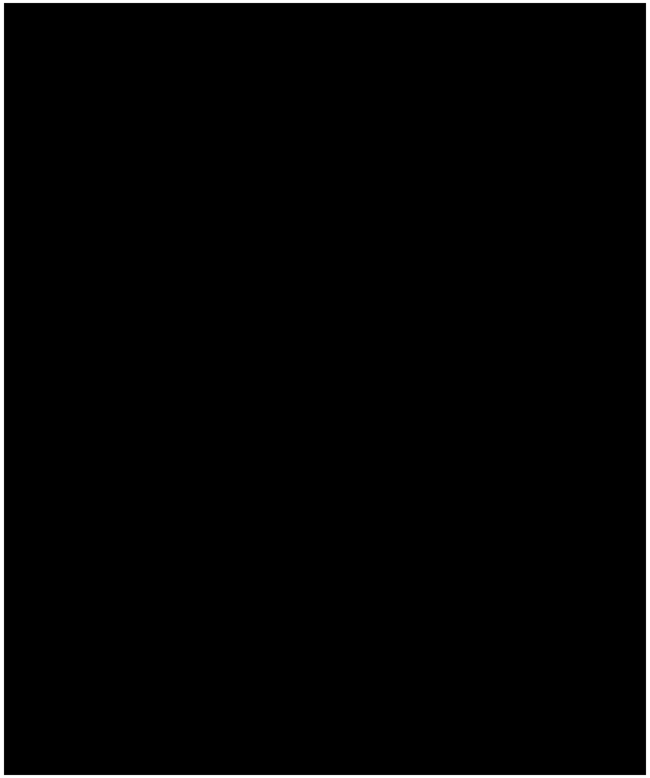
- Roots (3):
  - Cylindrical
  - 10 cm x 0.05 cm
  - immersed in water in a glass flask
- Shoots (3):
  - Elliptical cylinders
  - 18 cm x 0.2 cm x 0.05 cm





Voxel Phantom

Contour slices



Import micro-CT
into 3D Doctor

3D Render

Export to
lattice tool

Verify geometry

Voxel → NURBS (Hybrid) Phantom

Voxel Phantom

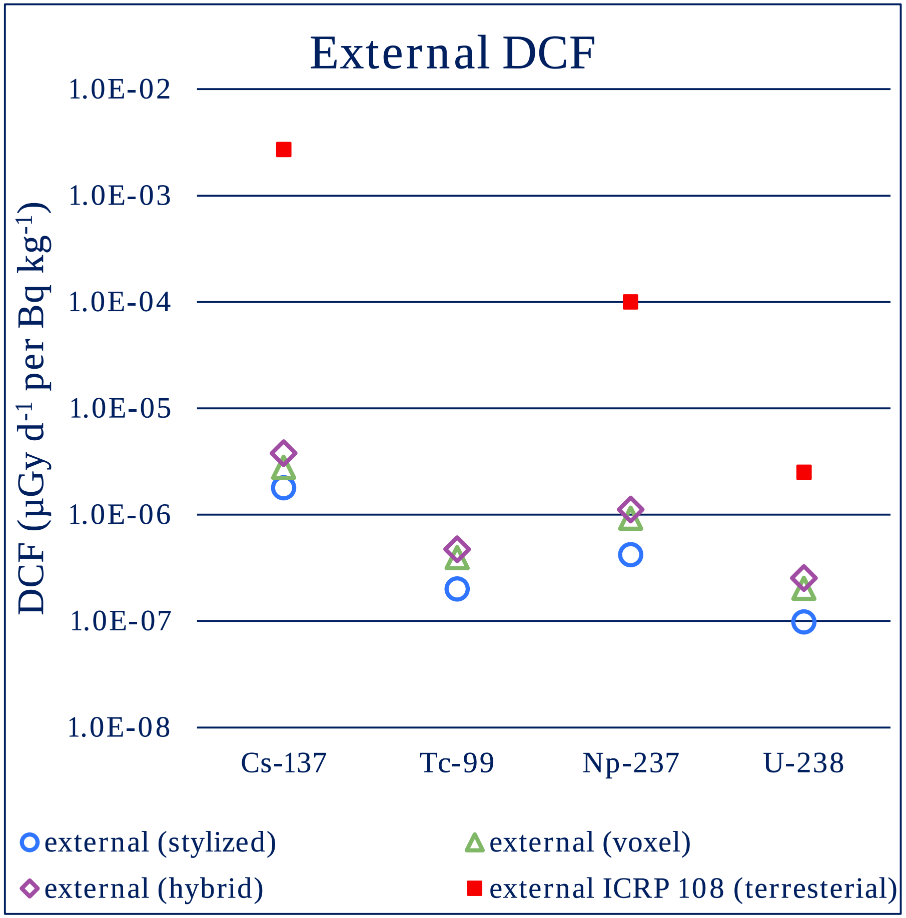
Rhinoceros 3D

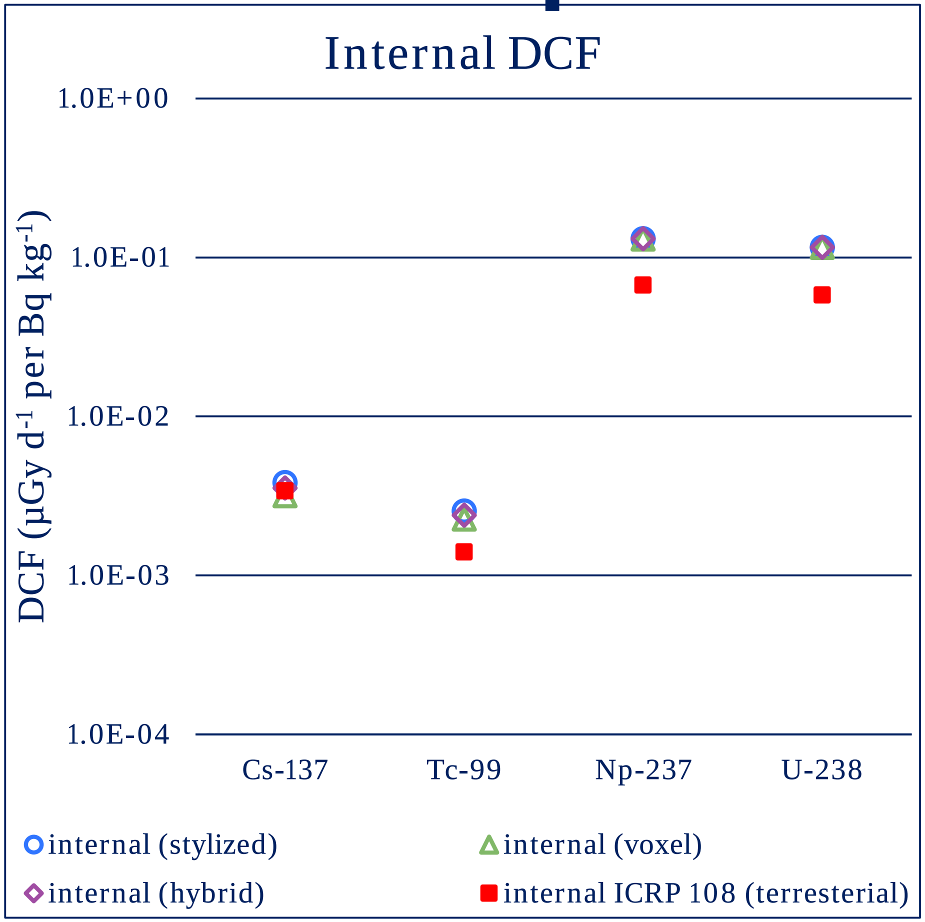
Hybrid Phantom

Radiation Transport Simulation

- Monte Carlo
  - MCNP/MCNPX, EGS4, GATE, GEANT 4
- MCNP
  - Output is energy deposition (MeV) normalized per disintegration (*f8 tally)
  - Tally energy deposition in tissues of interest
  - Dose conversion factor × activity concentration = dose rate
  - Absorbed fractions (most commonly reported)
- Generally need supercomputer (Palmetto Cluster)

Whole Plant DCF ICRP 108 Comparison





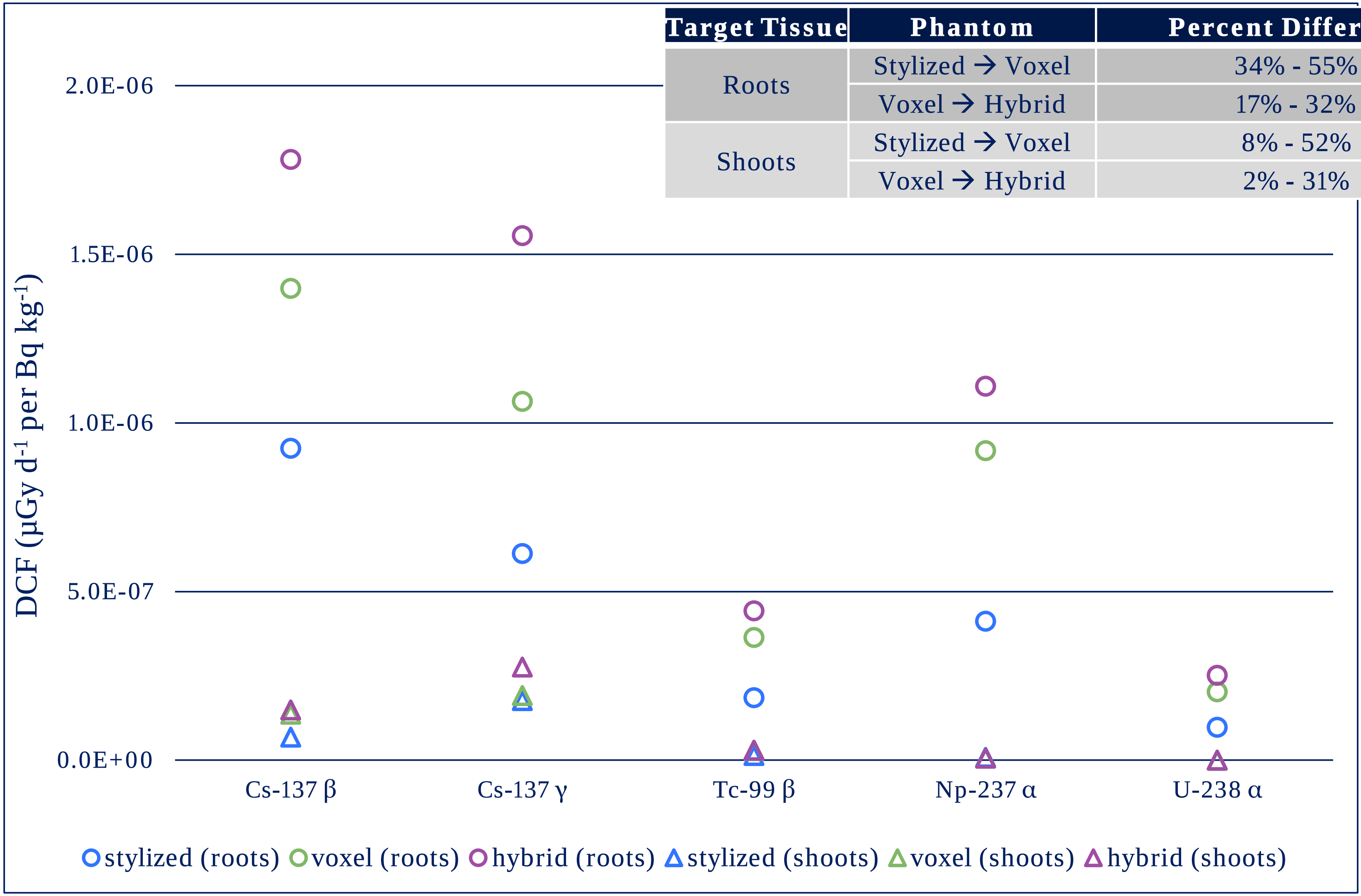
~1 – 3 orders of magnitude difference
geometry effects

<0.3 order of magnitude difference
(~4% – 50% difference)

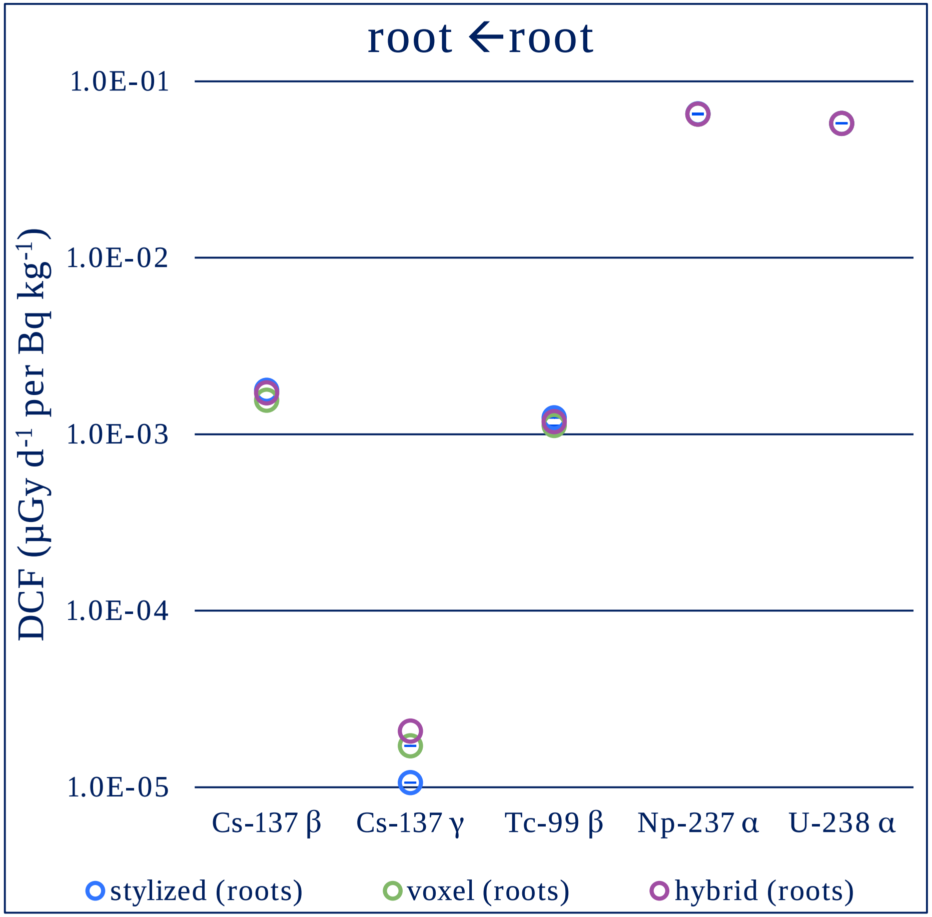
$$DCF\left(\frac{\mu\text{Gy day}^{-1}}{\text{Bq kg}^{-1}}\right) = \frac{\text{MeV}}{\text{disintegration}}\left|\frac{\text{dis/sec}}{\text{Bq}}\right|\frac{1.602\times10^{-13}\,\text{J}}{\text{MeV}}\left|\frac{86400\text{ s}}{\text{day}}\right|\frac{\text{Gy}}{\text{J/kg}}\left|\frac{10^{6}\,\mu\text{Gy}}{\text{Gy}}\right.$$

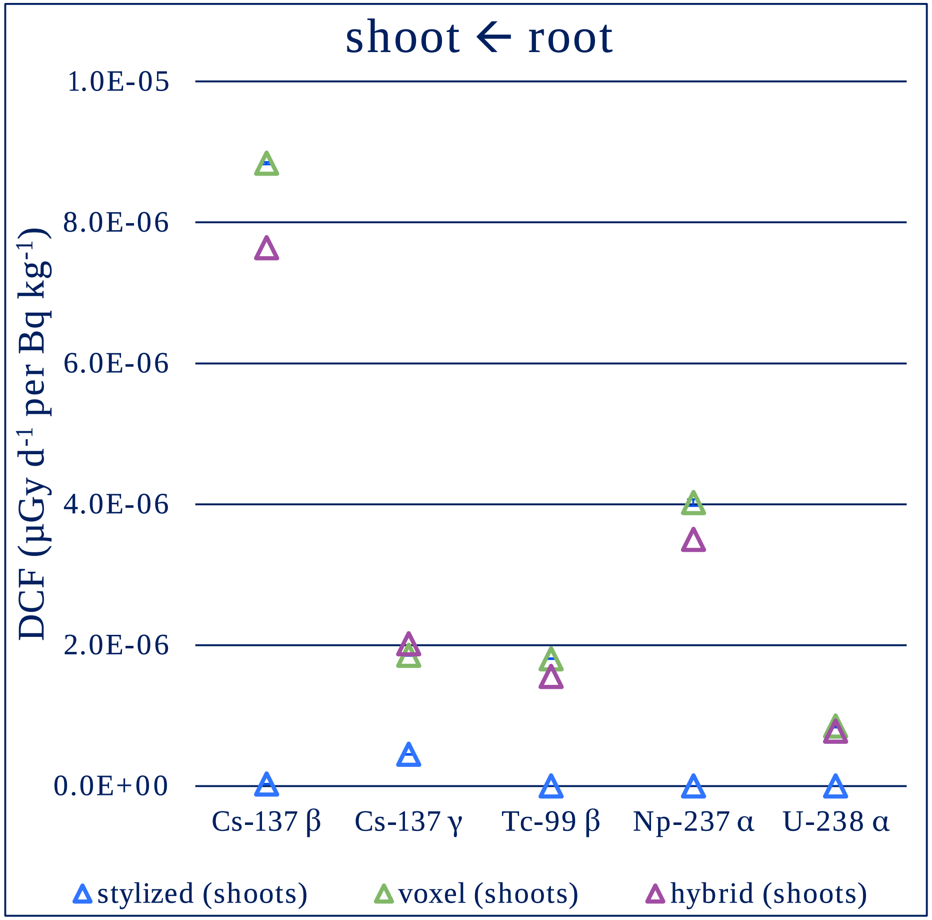
External DCF Phantom Comparison

Target Tissue	Phantom	Percent Difference
Roots	Stylized → Voxel	34% - 55%
	Voxel → Hybrid	17% - 32%
Shoots	Stylized → Voxel	8% - 52%
	Voxel → Hybrid	2% - 31%



Internal DCF Phantom Comparison



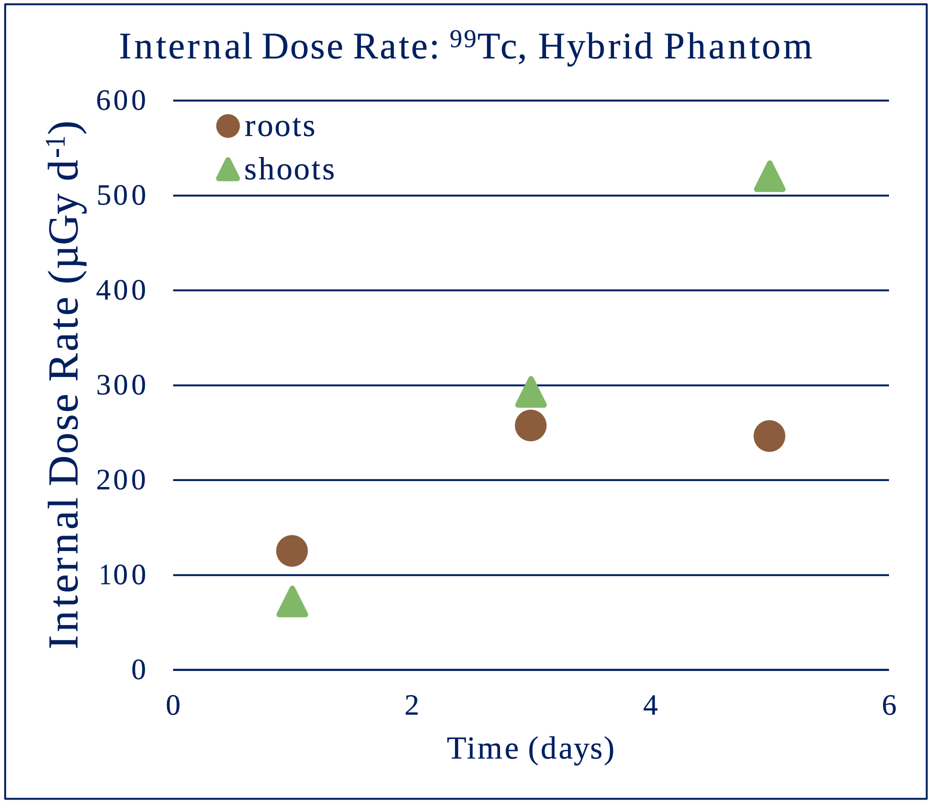


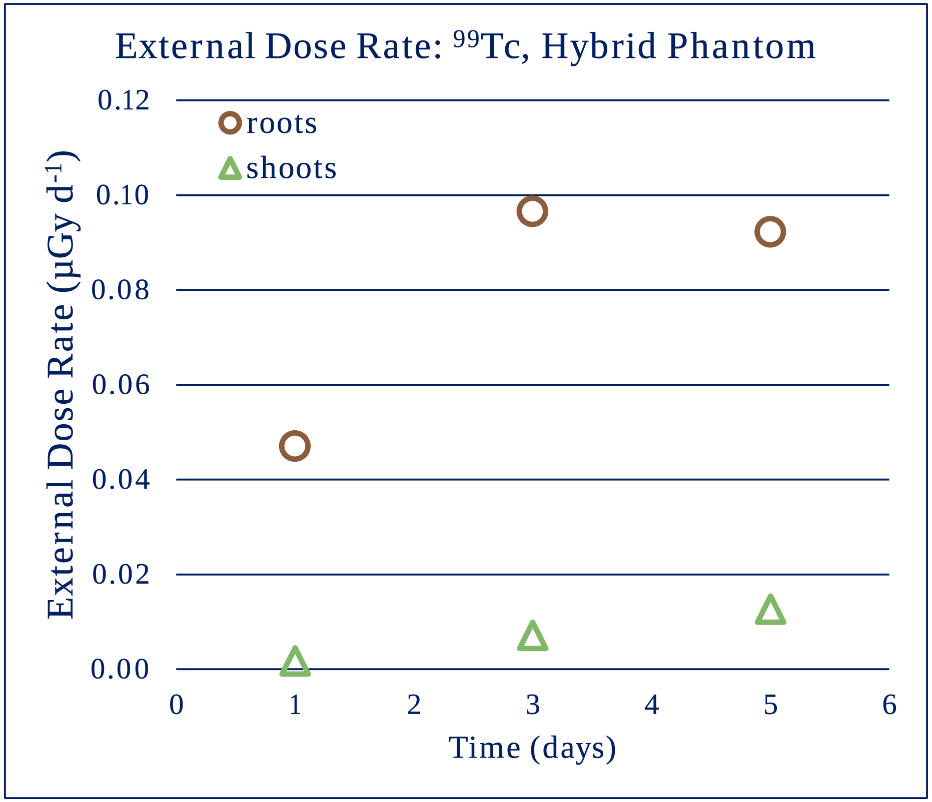
Cs γ: 38% difference (stylized → voxel)
Most others < 10%

0.62 to 4.15 orders of magnitude difference
(stylized – voxel or hybrid)

Greatest relative differences when source ≠ target
due to root-shoot gap in stylized phantom.

Combining uptake with DCF





Dosimetric Modeling Key Points

- Detailed dosimetric models provide higher fidelity and flexibility than traditional ellipsoid models, but some limitations remain
  - Looking for a **balance** between detail and resource requirements
- Normally dose rates from anthropogenic activities are low, although **screening values** have been exceeded
  - If screening values are exceeded, it may be helpful to consider a more detailed or realistic phantom
- Screening criteria are **not consistent** between countries and **no specific approach** exists for performing a detailed environmental impact assessment should a criteria be exceeded
- Recent international consideration has resulted in development of a **multi-part framework** for impact assessment
  - Includes undertaking progressively more refined assessments and improved models

Smith et al 2010, Jackson et al 2014

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Acknowledgements

Advisors: Drs. Nicole Martinez and Brian Powell
Committee Members: Drs. Dan Kaplan and Nishanth Tharayil
Nimisha Edayilam for providing plant specimens and assistance
Powell & Martinez research groups

This material is based upon work supported by the U.S. Department of Energy Office of Science, Office of Basic Energy Sciences and Office of Biological and Environmental Research under Award Number DE-SC-00012530 (uptake and student support) as well as the United States Nuclear Regulatory Commission Nuclear Education Grant #NRC-HQ-13-G-38-0002 (dosimetry and faculty support).

Clemson University is acknowledged for generous allotment of compute time on Palmetto cluster.

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Fukushima's Wildlife

Mammalian Species in and Around the Exclusion Zone

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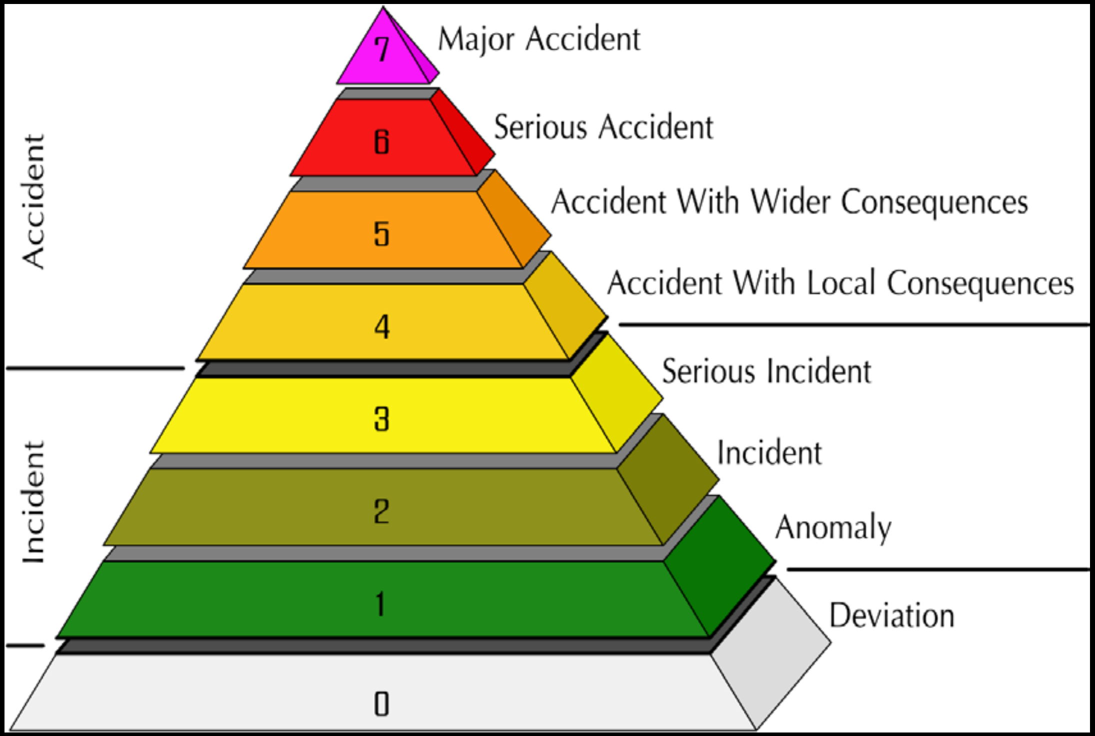
Roadmap

- Background
- Study area
- Methods
- Results

Background: Nuclear Accidents

- Three Mile Island
  - March 28, 1979
  - Level 5
- Chernobyl
  - April 26, 1986
  - Level 7
- Fukushima
  - March 11, 2011
  - Level 7

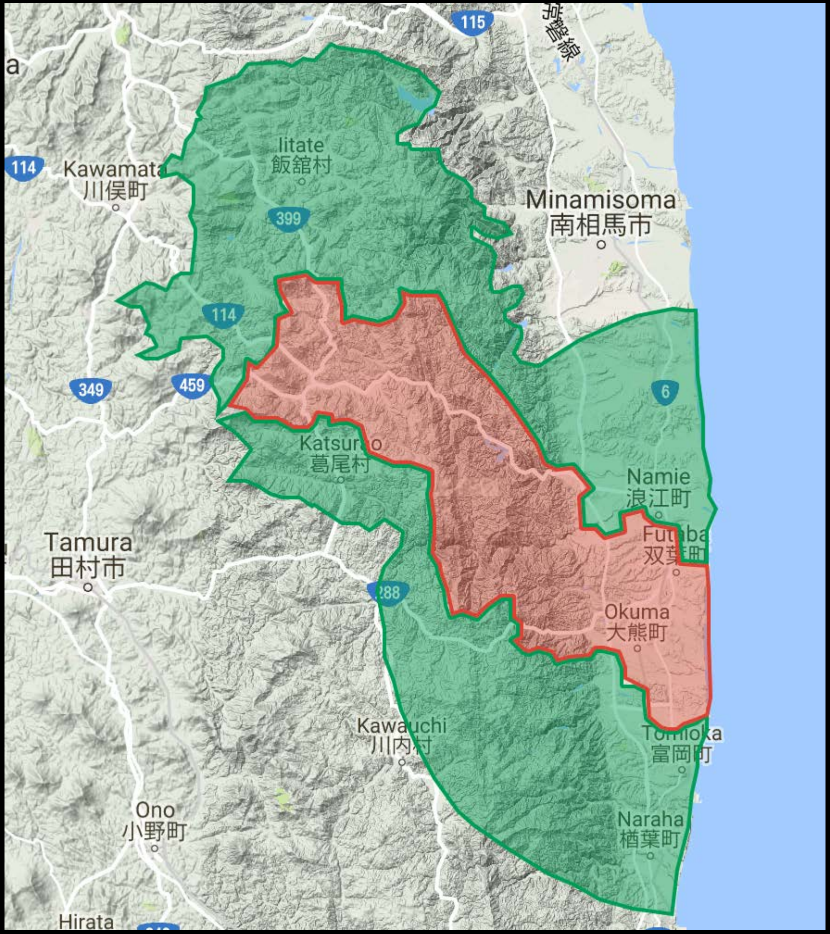


Fukushima Disaster

- Great East Japan Earthquake: magnitude 8.9
- Tsunami waves >40m (133 ft)
- 4 nuclear power plants damaged
- Fukushima Daiichi: 3 reactor meltdowns

Exclusion Zone

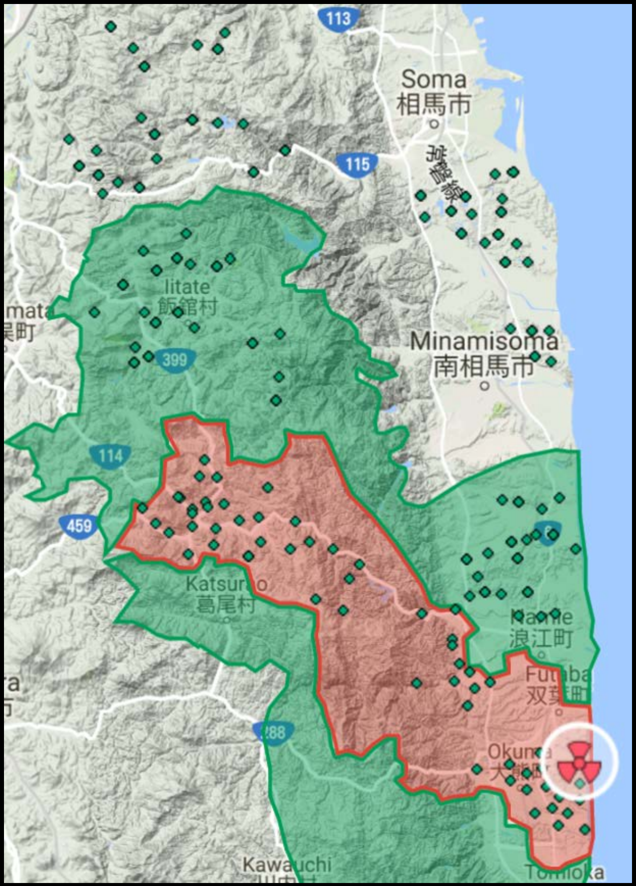
- Created on the day of the earthquake
- Maximum radiation: 91 µSv/hr
- 1150 km^2
- 170-200k people evacuated




千葉県産
岩がき
1ケ ¥1000 円
千葉県産
天然
岩がき
1ケ ¥700
千葉県産
天然
1ケ
生

Study Area

- **120 locations**
  - **40 locations** per area/zone
- **60 cameras** deployed per each two-month iteration
  - **Red zone:** 10 upland, 10 lowland
  - **Green zone**: 10 upland, 10 lowland
  - **Control area:** 10 upland, 10 lowland

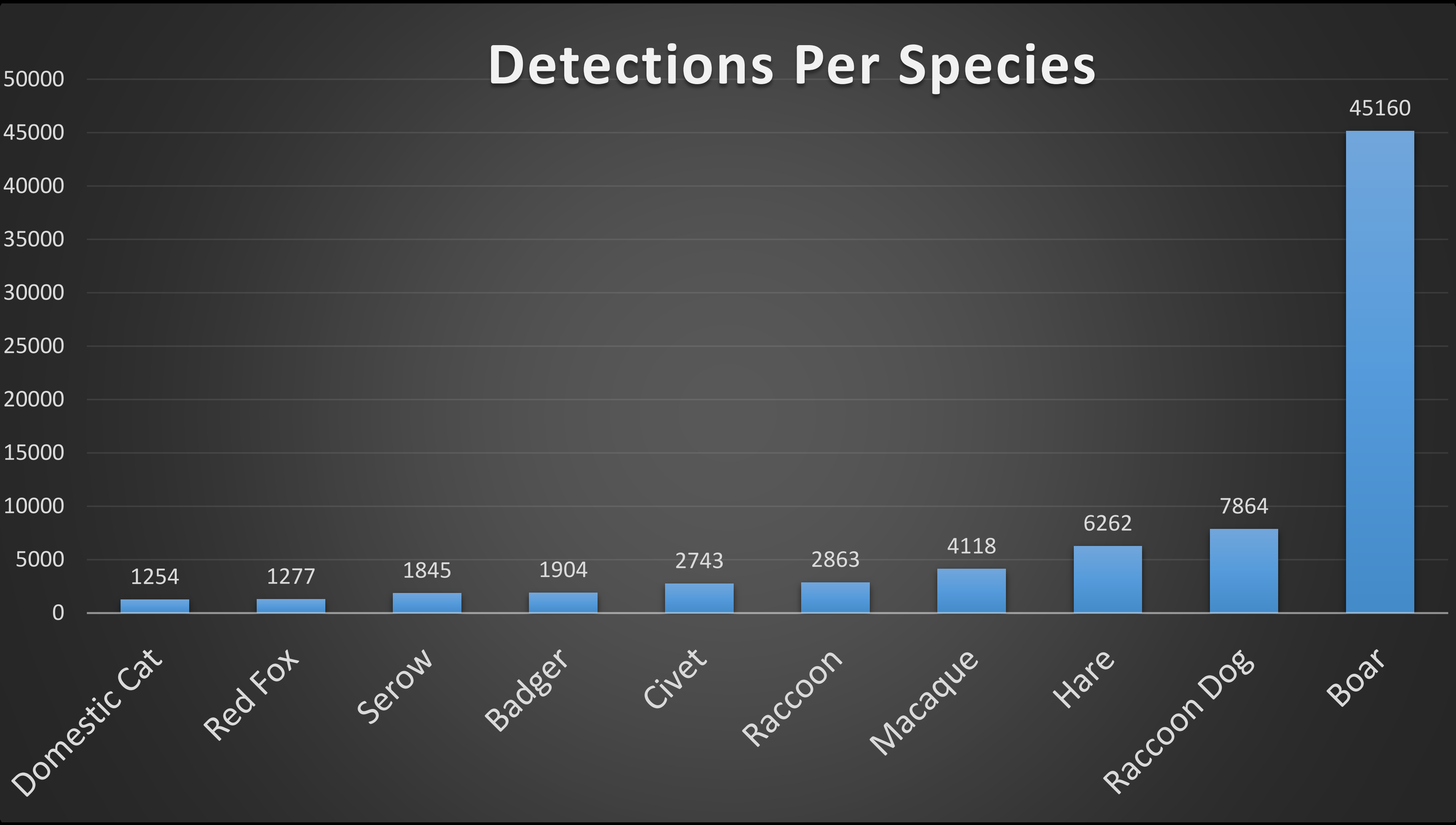


Trap Design

- Initial install dates: May 6-13, 2016
- Cameras placed ~ 60cm from base of tree along well-traveled trails
- Three-shot burst of photos per detection
- No quiet period between detections

Raw Data

- 14,400 camera days
- 268,961 images
- 1120 mean images per camera deployment
- 17 mammalian species



Detections Per Species
50000
45000
40000
35000
30000
25000
20000
15000
10000
5000
0
1254
1277
1845
1904
2743
2863
4118
6262
7864
45160
Domestic Cat
Red Fox
Serow
Badger
Civet
Raccoon
Macaque
Hare
Raccoon Dog
Boar

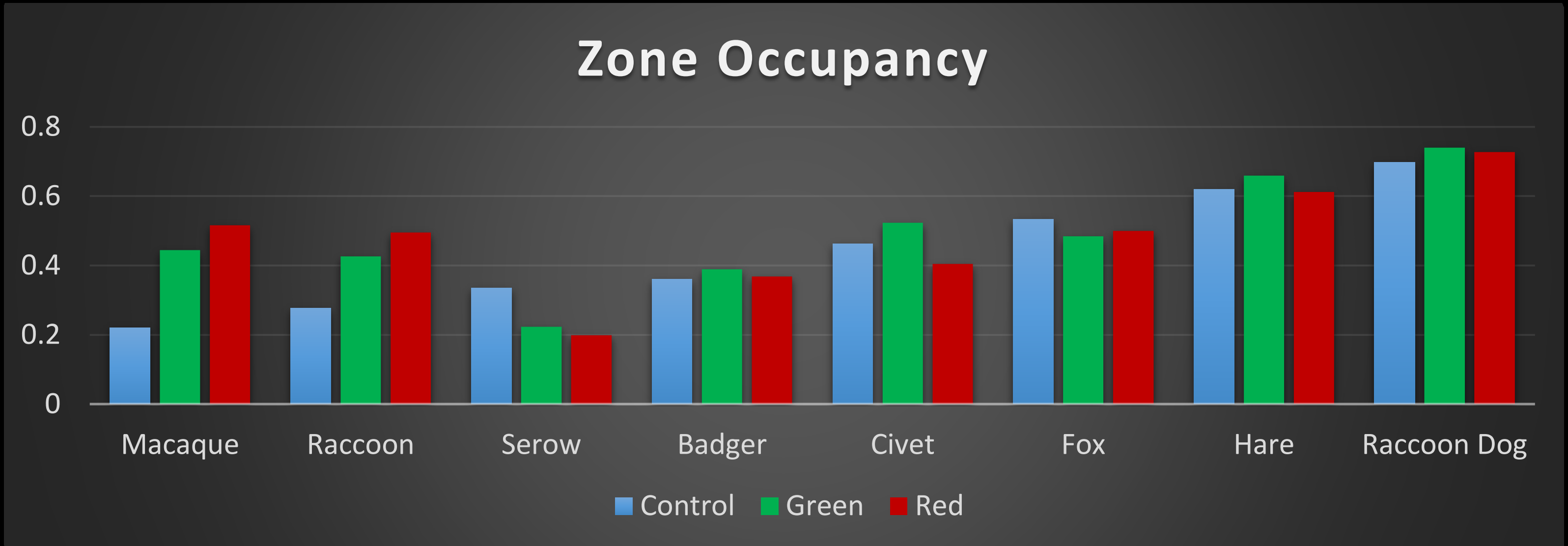

2016-06-11 13:44:39 M 3/3 96°F
UGA
2016-06-15 13:10:41 M 2/3 68°F
UGA
2016-07-30 22:45:39 M 3/3 72°F
UGA
2016-06-08 12:56:56 M 1/3 72°F
UGA
2017-02-09 14:22:26 M 2/3 38°F
UGA
2016-05-12 14:03:07 M 2/3 73°F
UGA

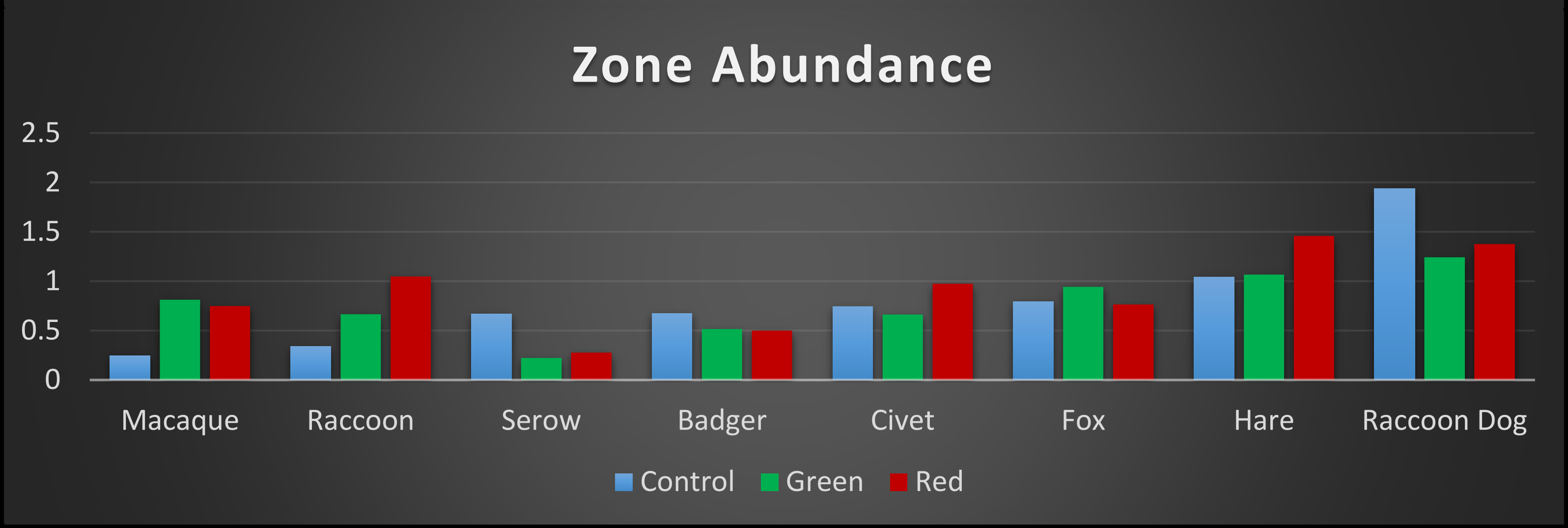
Analysis

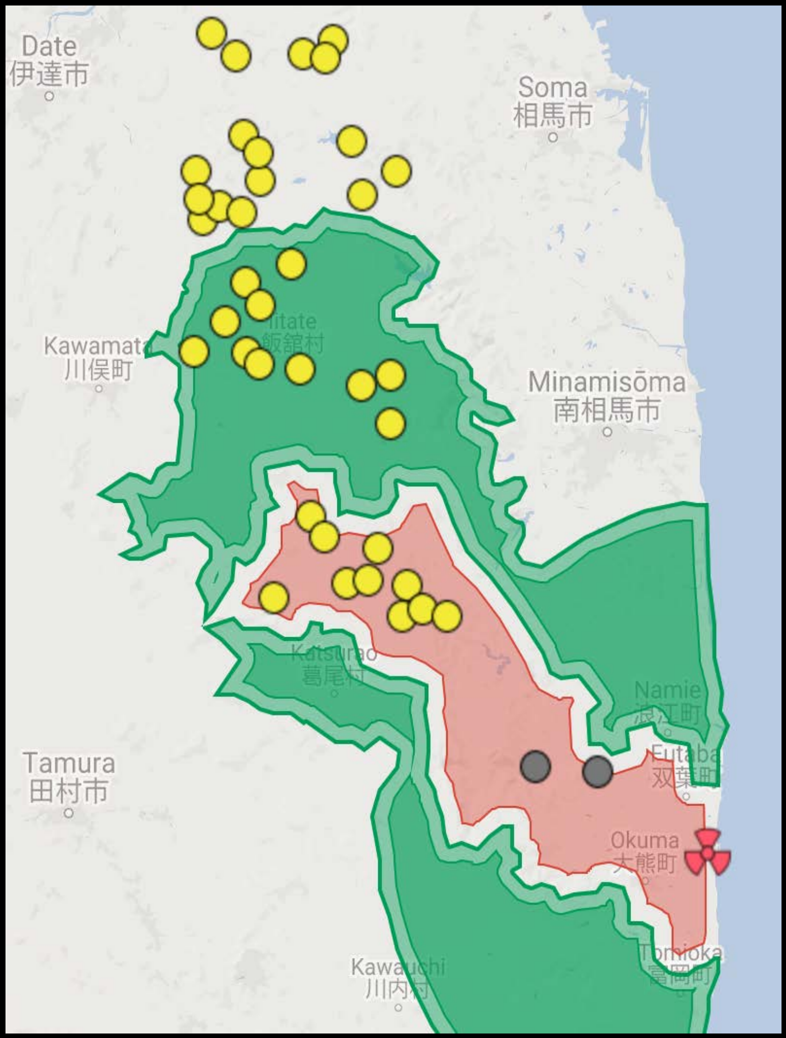
- Daily presence/absence design
- Utilized the R program UNMARKED for both occupancy and abundance
- Occupancy: Mackenzie et al. 2002 methods
- Abundance: Royle and Nichols 2003 methods

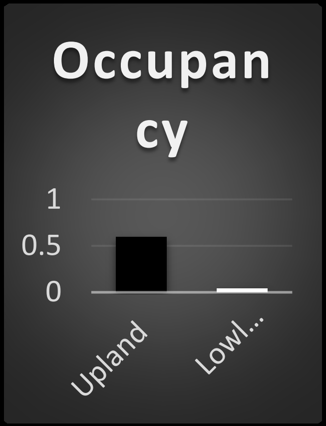
Covariates

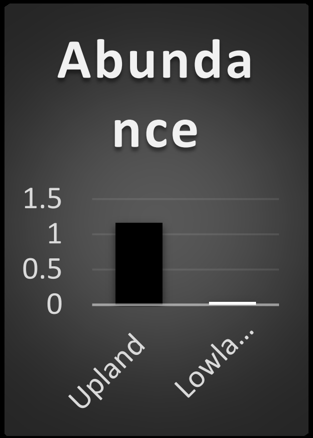
- Vegetation
- Distance to water source
- Site microseiverts per hour
- Distance to nearest road
- Human trail usage
- Zone
- Elevation


Zone Occupancy
0.8
0.6
0.4
0.2
0
Macaque
Raccoon
Serow
Badger
Civet
Fox
Hare
Raccoon Dog
Control
Green
Red


Zone Abundance
2.5
2
1.5
1
0.5
0
Macaque
Raccoon
Serow
Badger
Civet
Fox
Hare
Raccoon Dog
Control
Green
Red





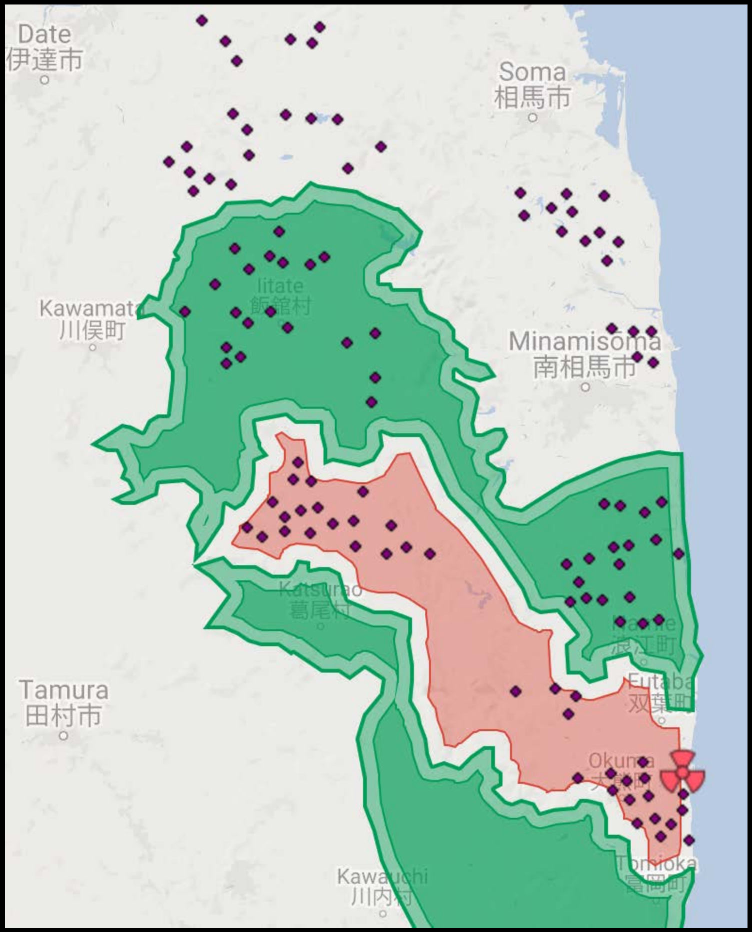


Serow

Capricornis crispus

Wild Boar

Sus scrofa



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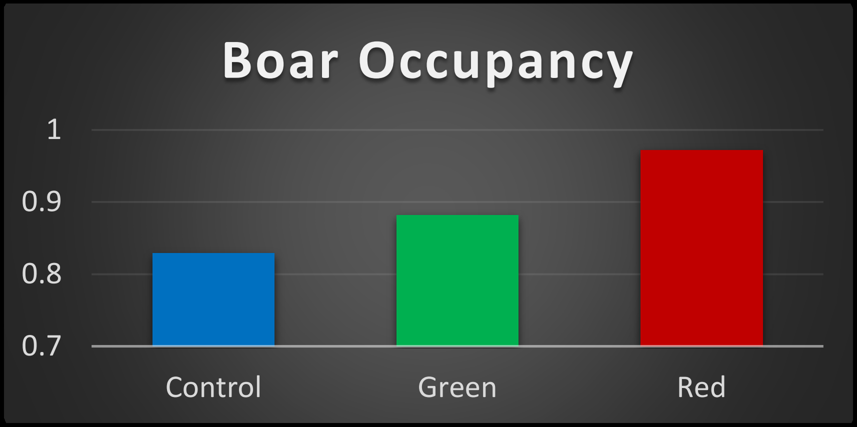
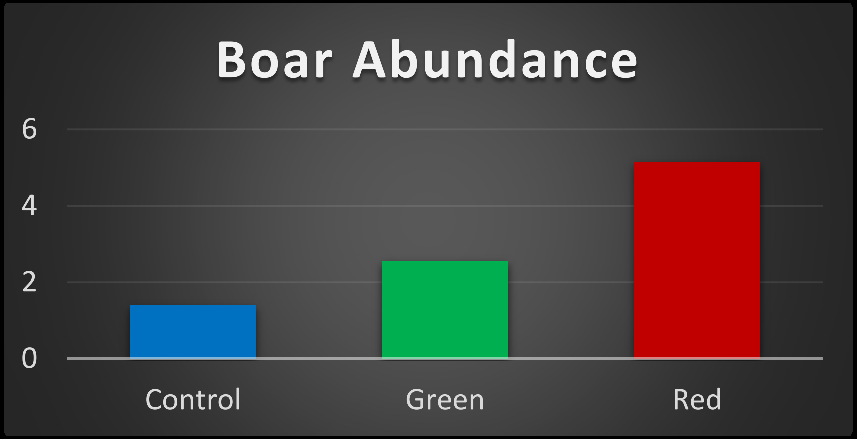
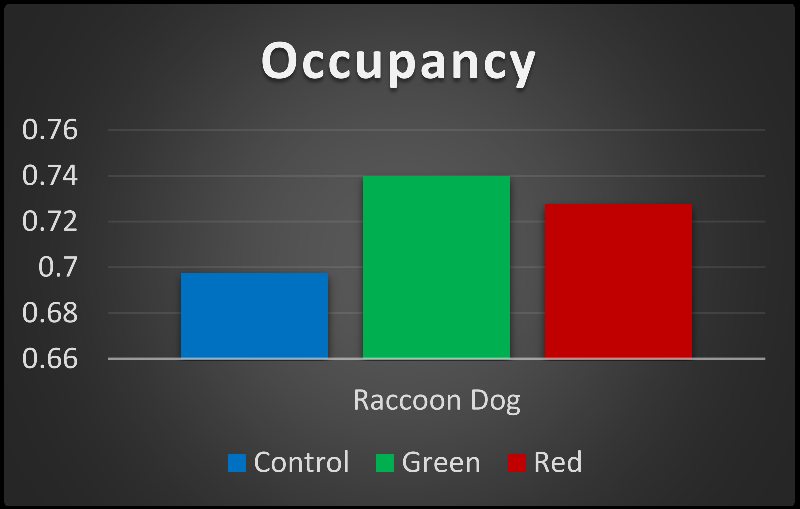
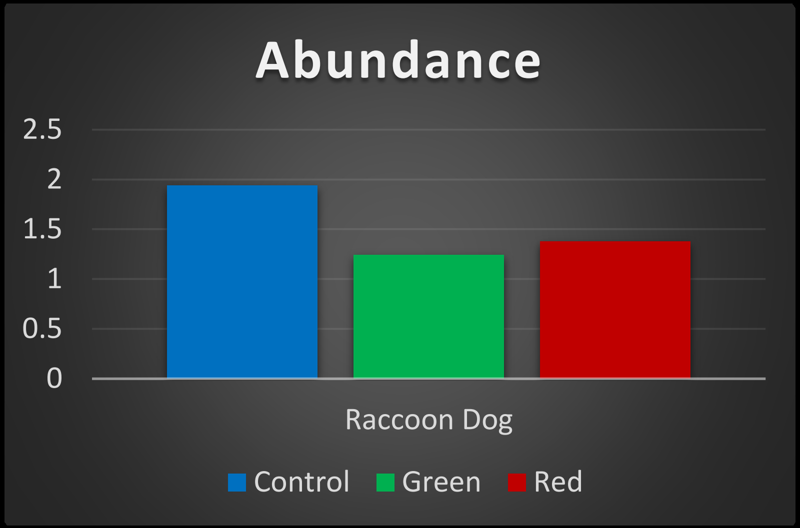




Raccoon Dog

Nyctereutes procionoides





Discussion

- Occupancy higher in exclusion zone for 6 of 9 species
  - Pig, Macaque, Raccoon, Badger, Hare, Raccoon Dog)
- Abundance higher in exclusion zone for 6 of 9 species
  - Pig, Macaque, Raccoon, Civet, Fox, Hare
- Human presence may have the strongest impact on area wildlife occupancy and abundance
- Radiation exposure has minimal impact on area wildlife occupancy and abundance

Acknowledgements

- Matt Hamilton
- Dr. Tom Hinton
- Dr. Kei Okuda
- Hannah Gerke
- Dr. James Beasley

Questions?

Discount Albacore Radioecology: Hunting Cs-137 on a budget

Delvan R. Neville
Oregon State University

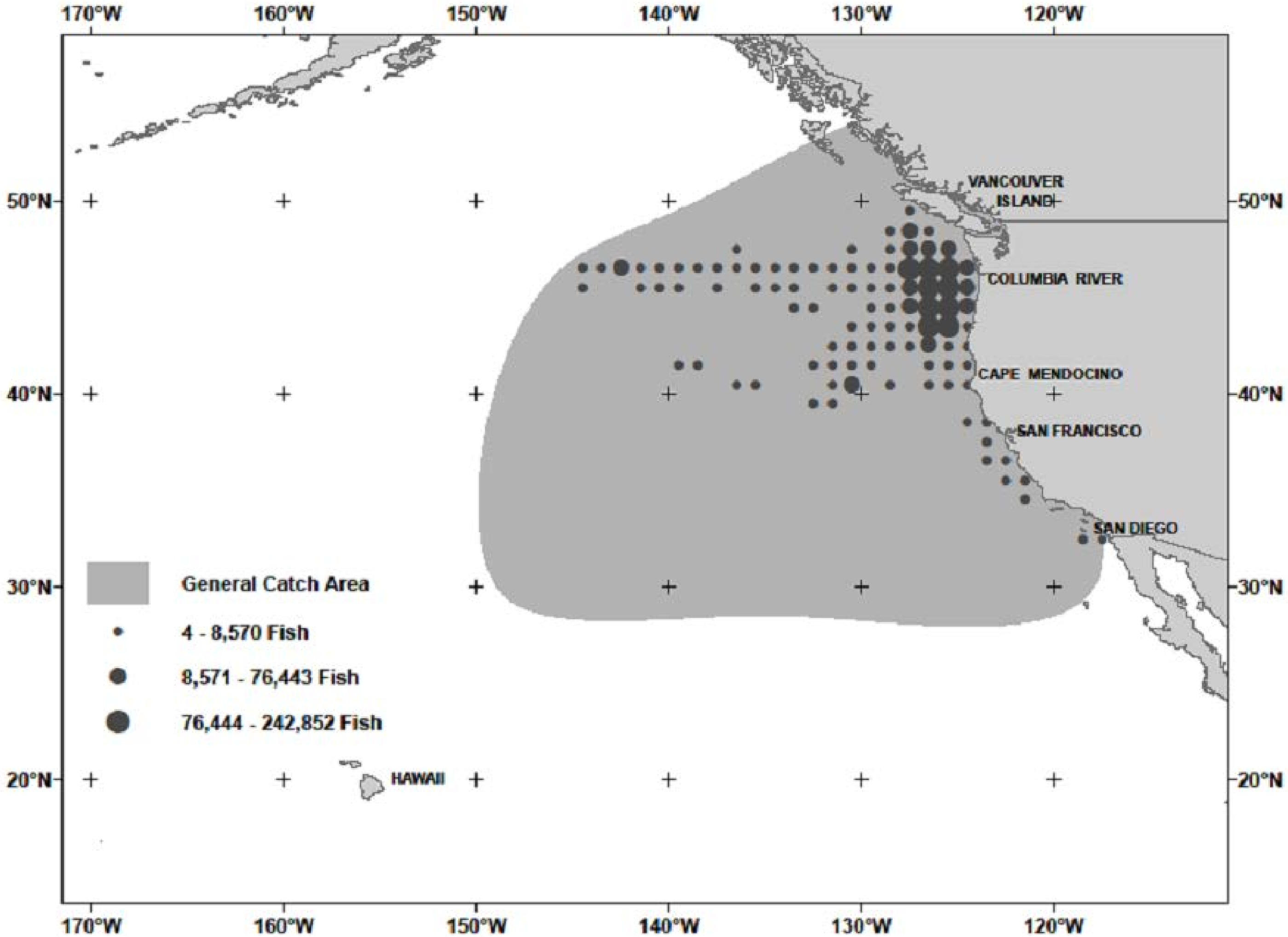
National Analytical Management Program (NAMP)
U.S. Department of Energy Carlsbad Field Office

TRAINING AND EDUCATION SUBCOMMITTEE

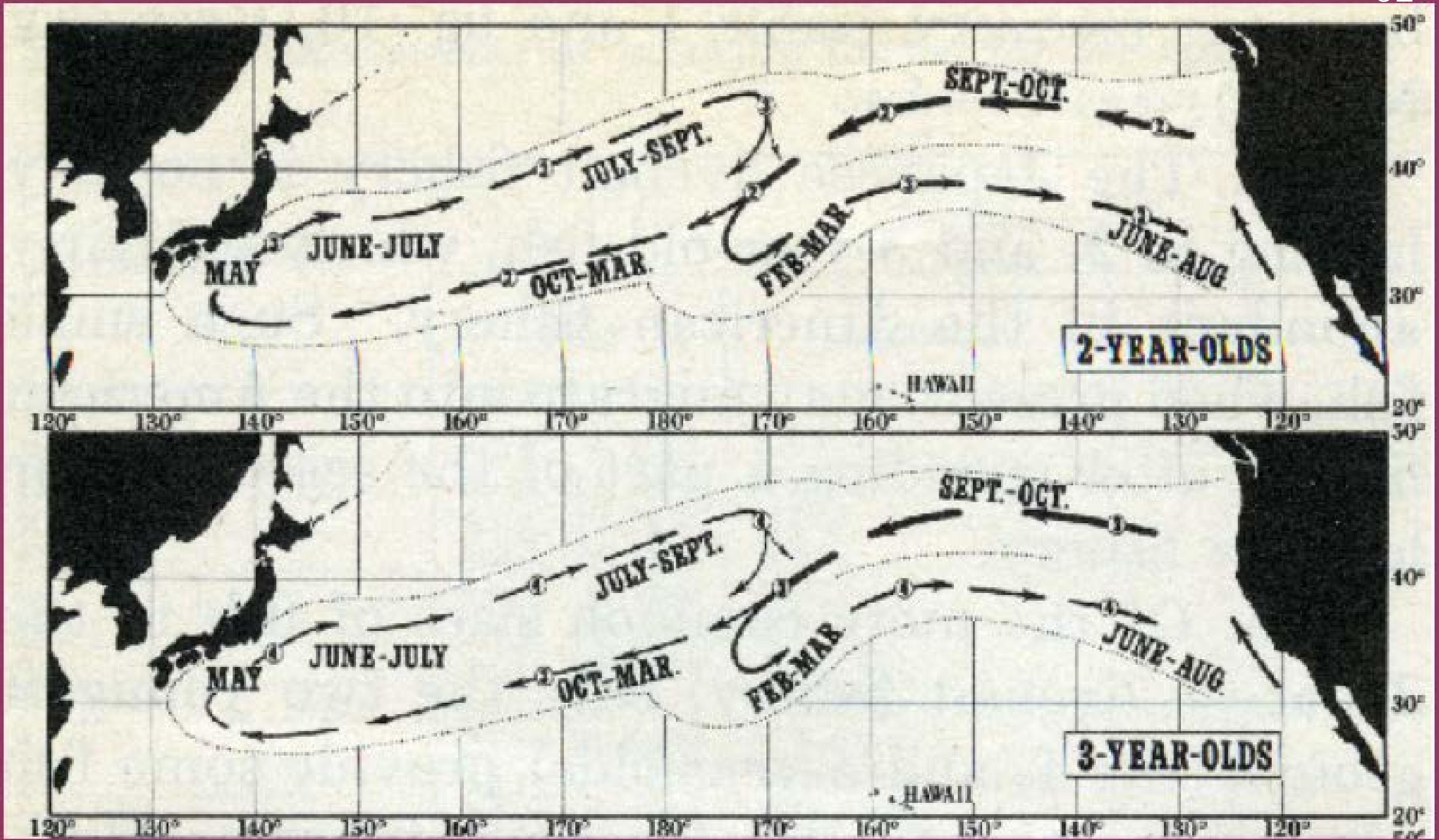
US Pacific albacore

- Primarily long-line trolling
- Fishery took off ca. 1915 @ 20,000,000 lbs/yr
- 3rd largest gross $ in Oregon
  - Behind Dungeness crab & Pink shrimp
- Open fishery – No catch limits

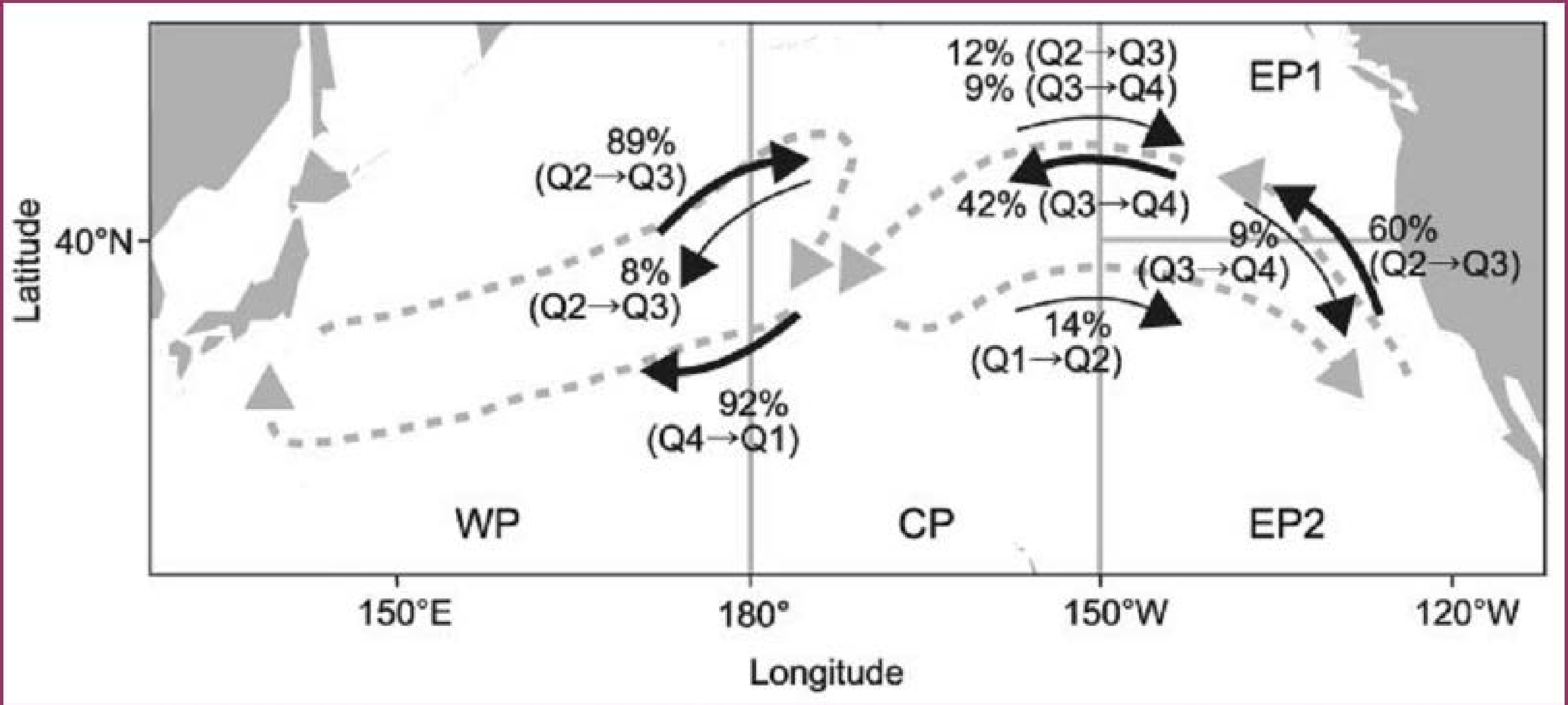
2009 Troll & Pole Catch



Childers & Pease, 2012



Otsu, T. and R.N. Uchida. 1962. Model of the migration of albacore in the North Pacific Ocean. *Fishery Bulletin* 63 (1): 33- 44.



Ichinokawa et al. 2008. Transoceanic migration rates of young North Pacific albacore, Thunnus alalunga, from conventional tagging data. *Can. J. Fish. Aquat. Sci.* 65:1681-1691.

Dried isn’t enough!

No funds! What to dry ash in?

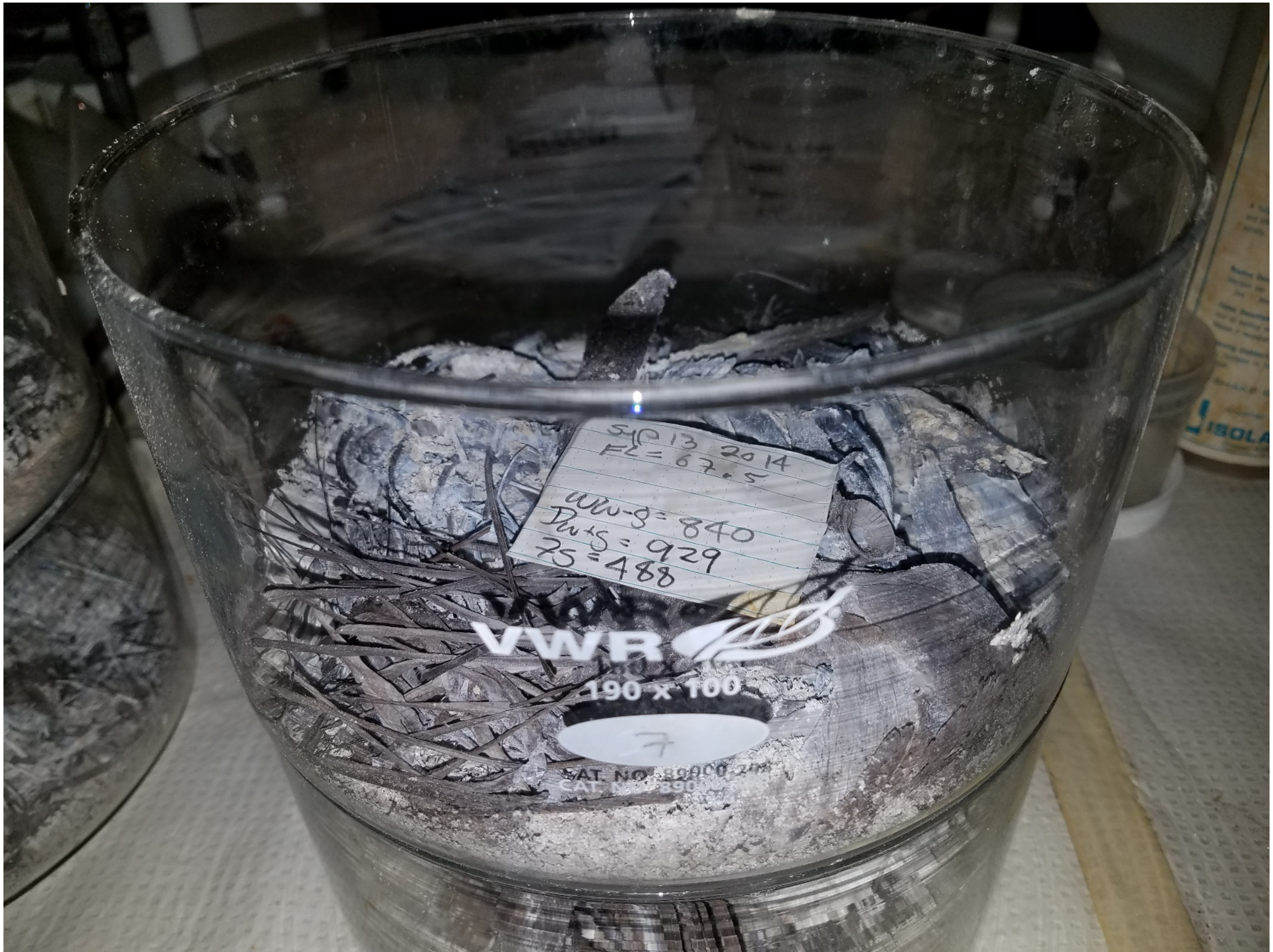
This won’t do

Stainless steel?

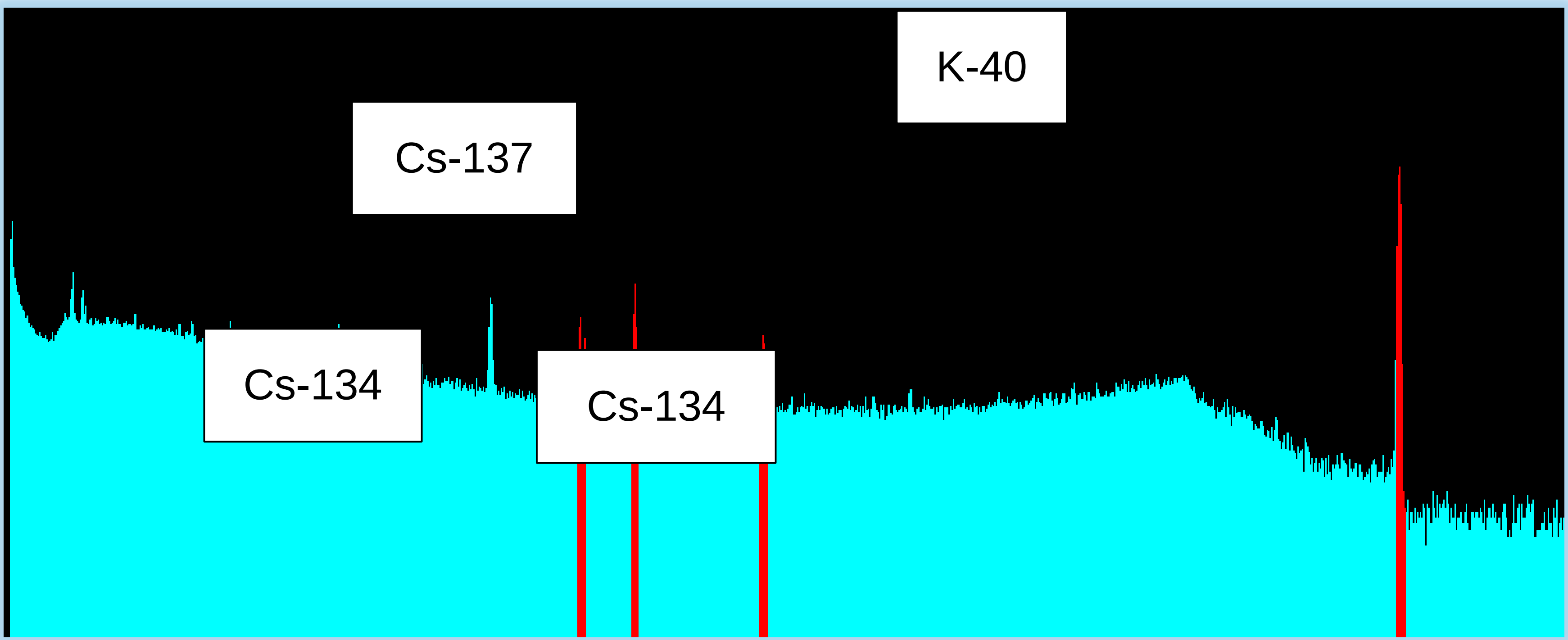
Wear & tear & K-40

EXTREME WORKING TEMPERATURE

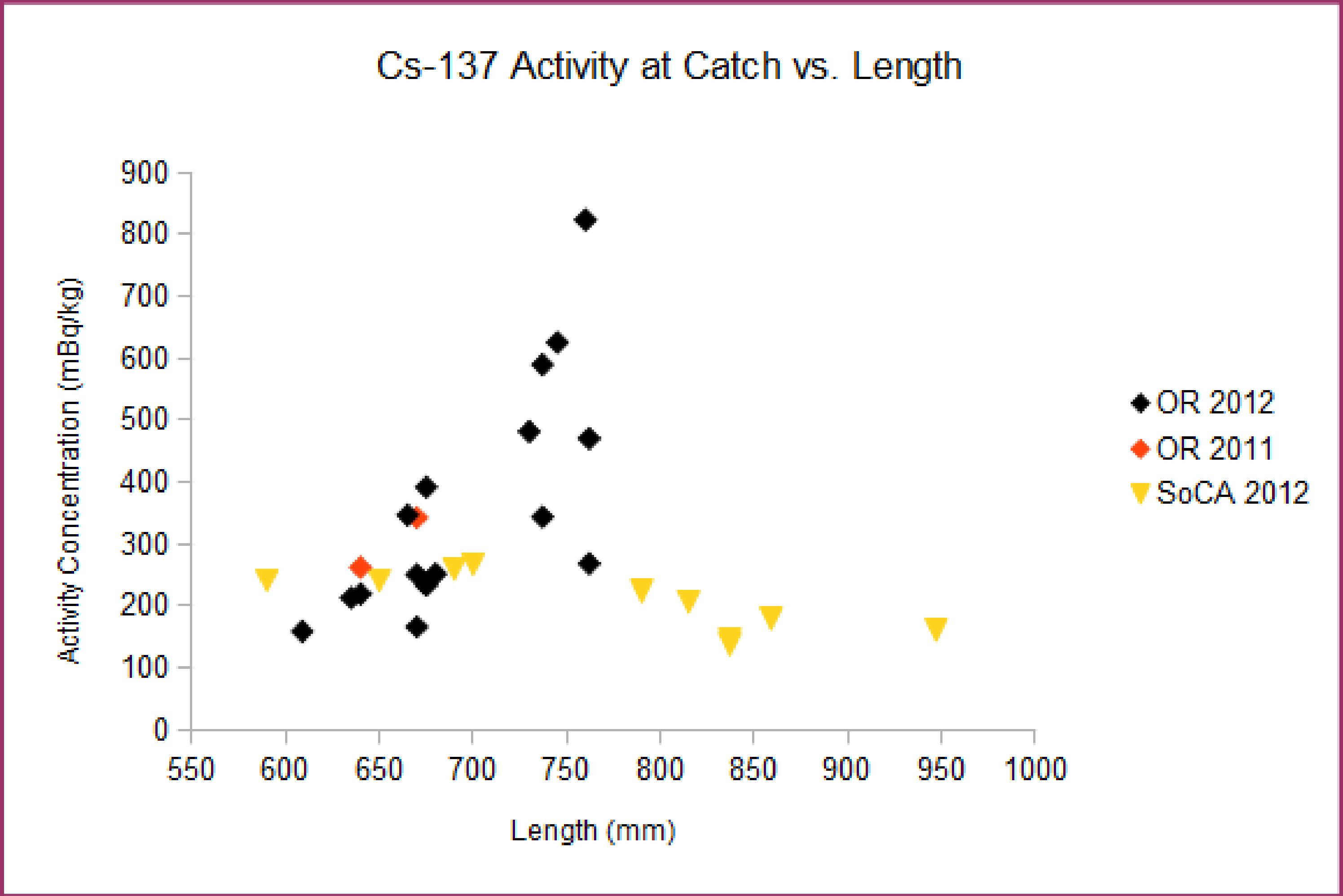
EXTREME WORKING TEMPERATURE



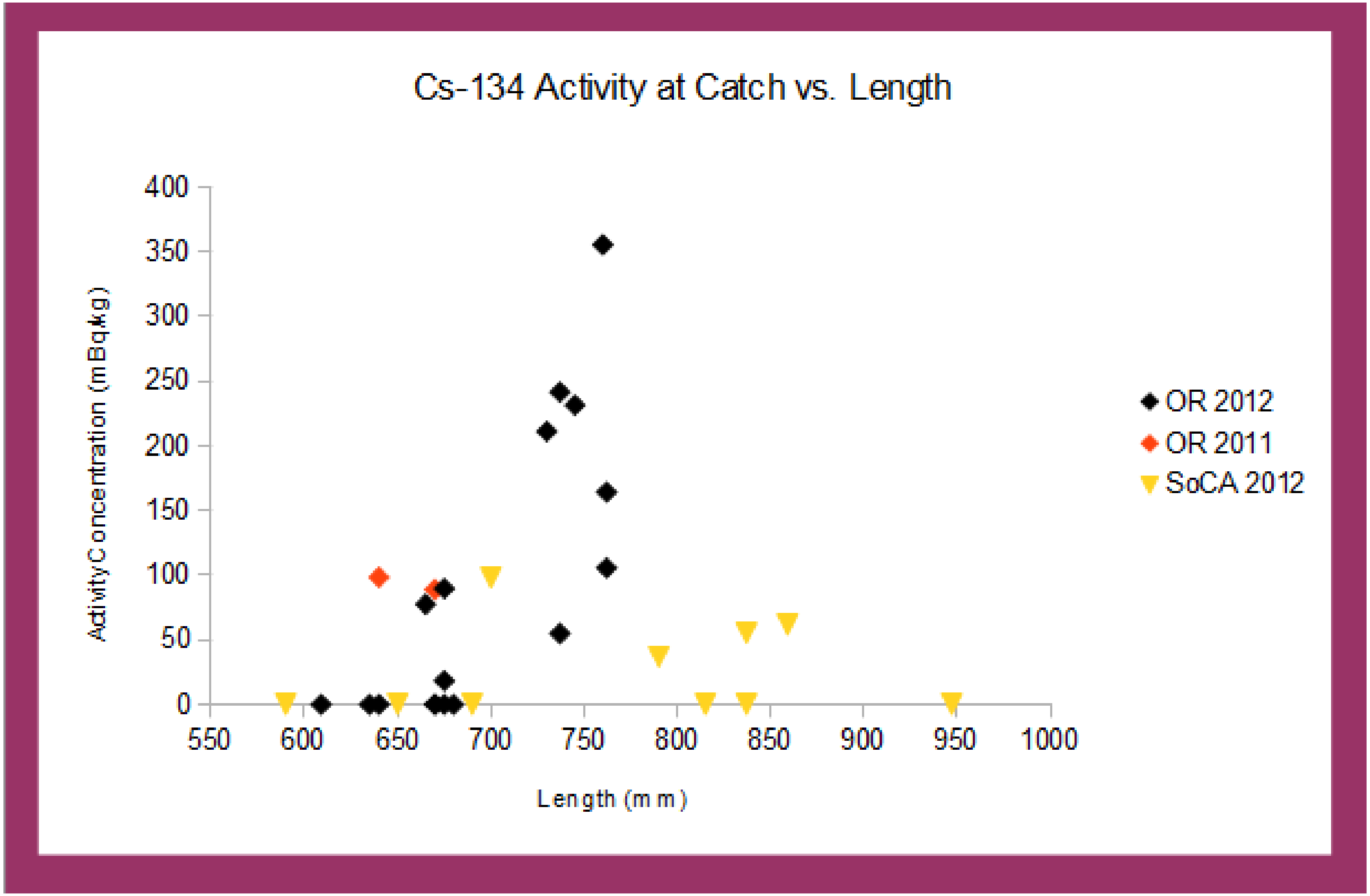
"Hottest" to date: <2 Bq/kg ww

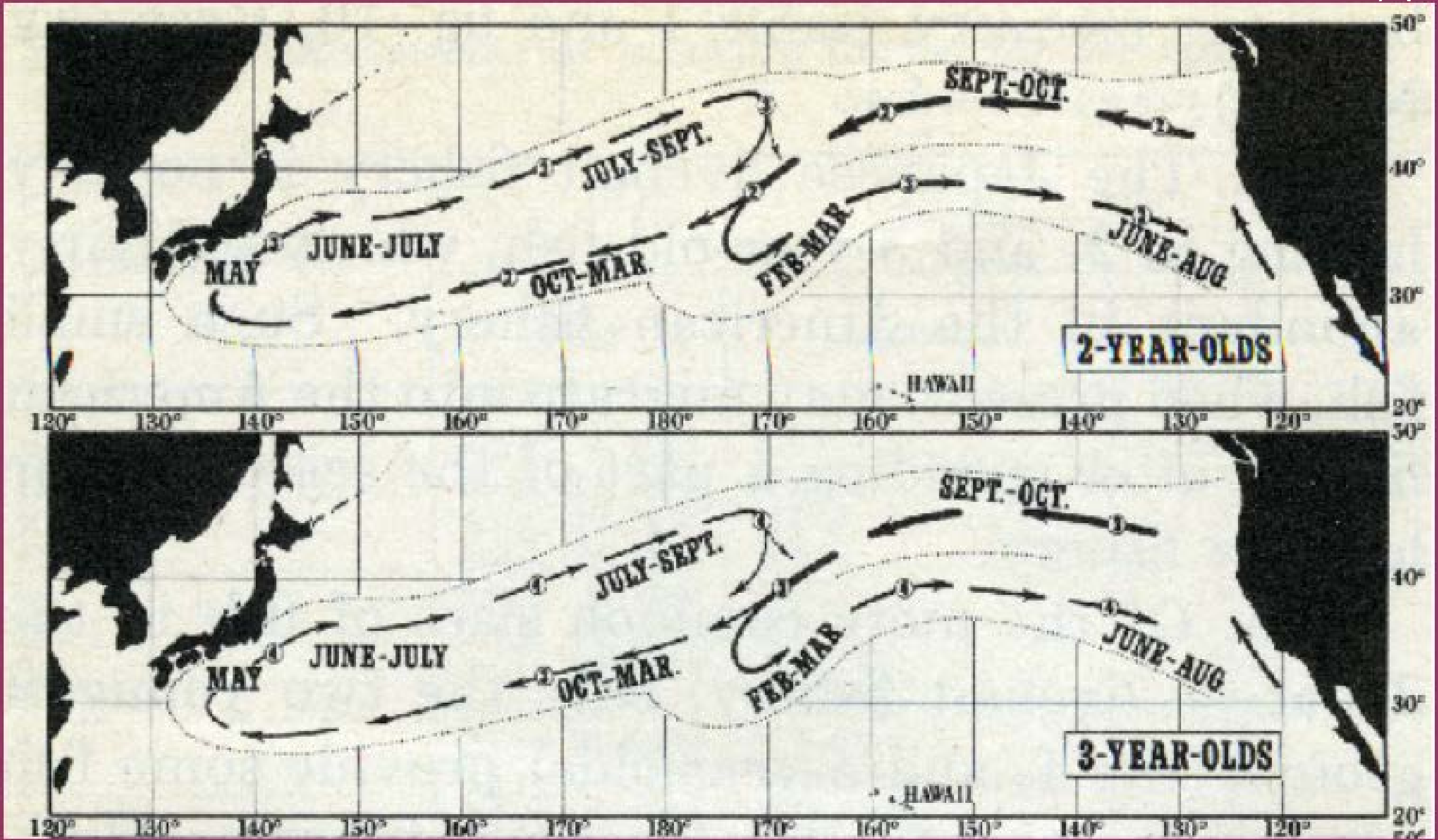


Preliminary Results – Cs-137



Preliminary Results – Cs-137





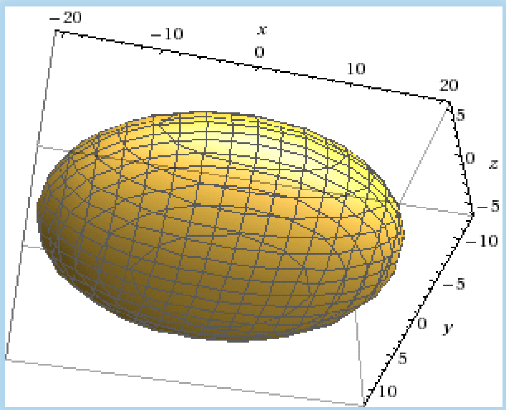
Otsu, T. and R.N. Uchida. 1962. Model of the migration of albacore in the North Pacific Ocean. *Fishery Bulletin* 63 (1): 33- 44.

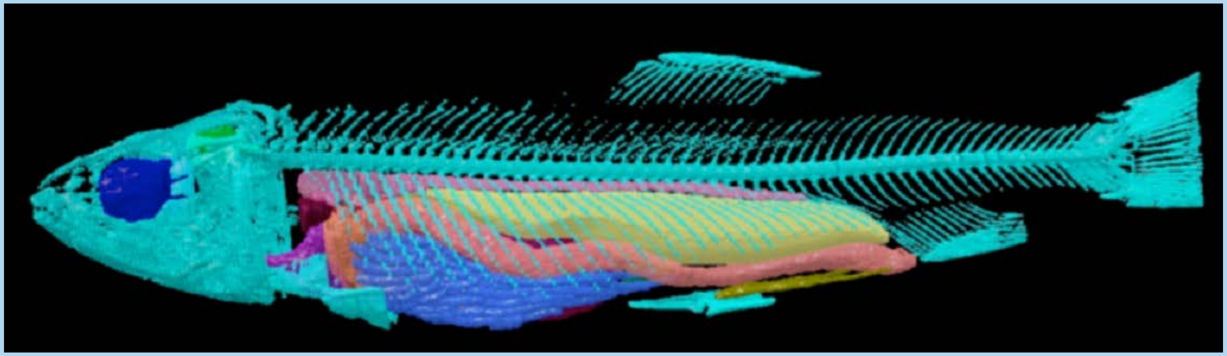
Preliminary Conclusions

- Food safety?
  - No issue
- Different populations?
  - Larger sample size needed -> Future work
  - Adjust for PDO & ENSO
- Dose impact?
  - Likely minimal, but opportunity to plan for larger oceanic release -> Future work

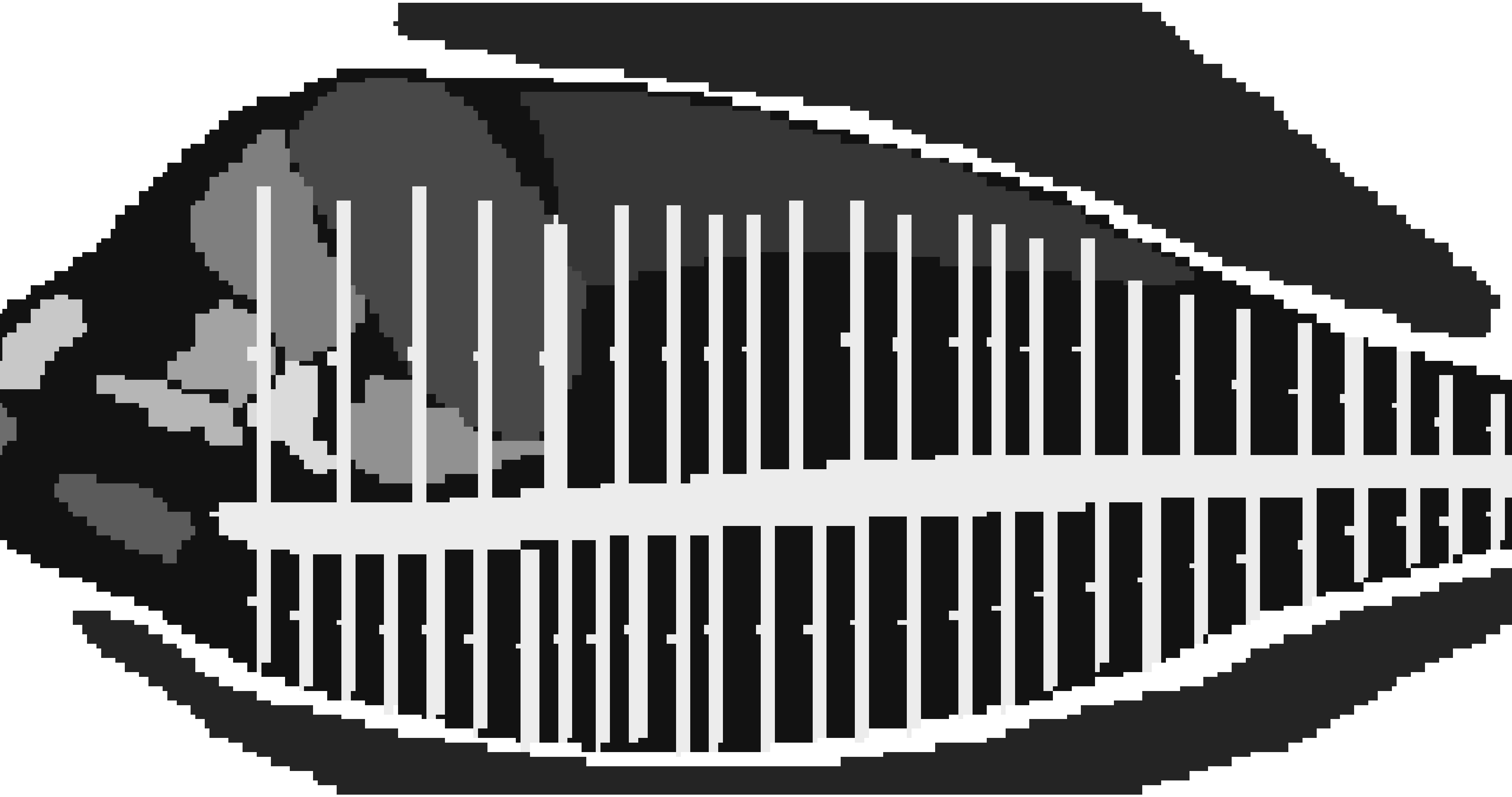
Dose in Pacific albacore

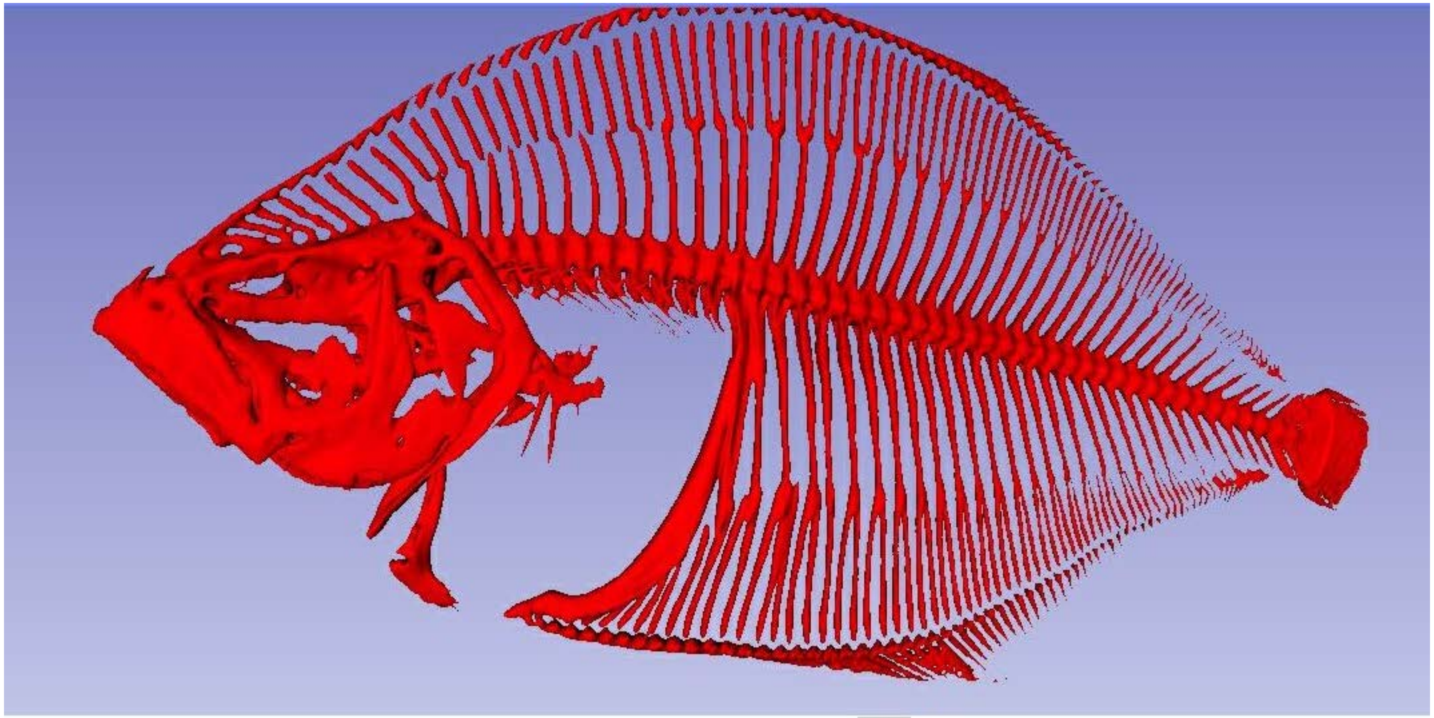
- ICRP Reference trout & flatfish
  - Neither perfect surrogate
  - Officially both ellipsoids
- Voxel models
  - Presumed more accurate
  - Time intensive

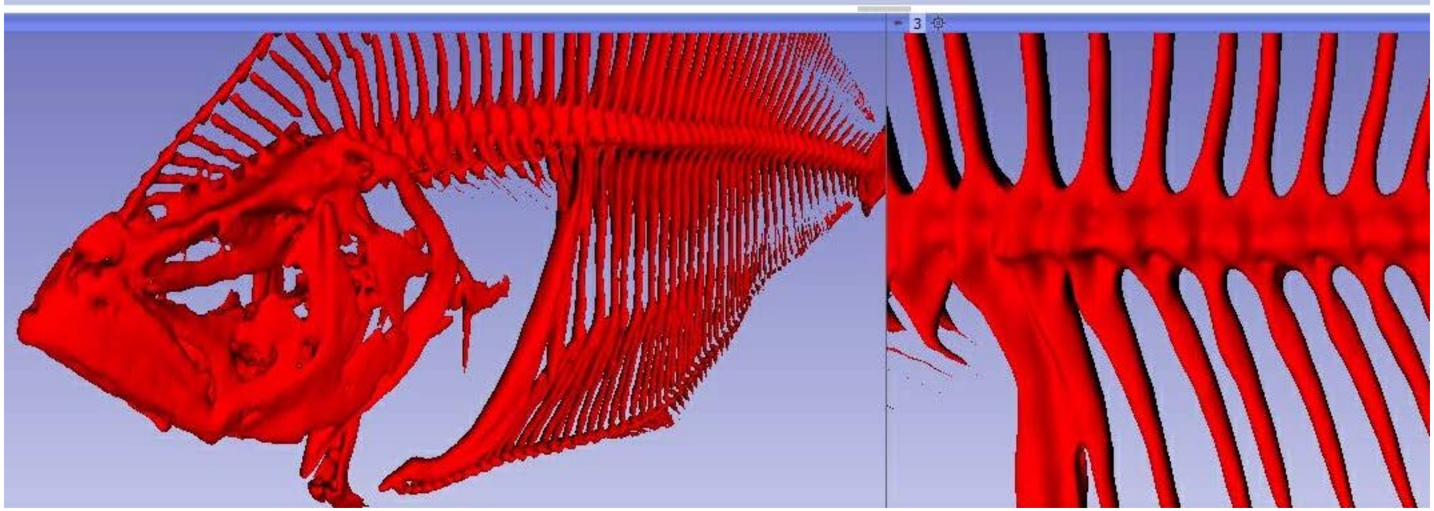




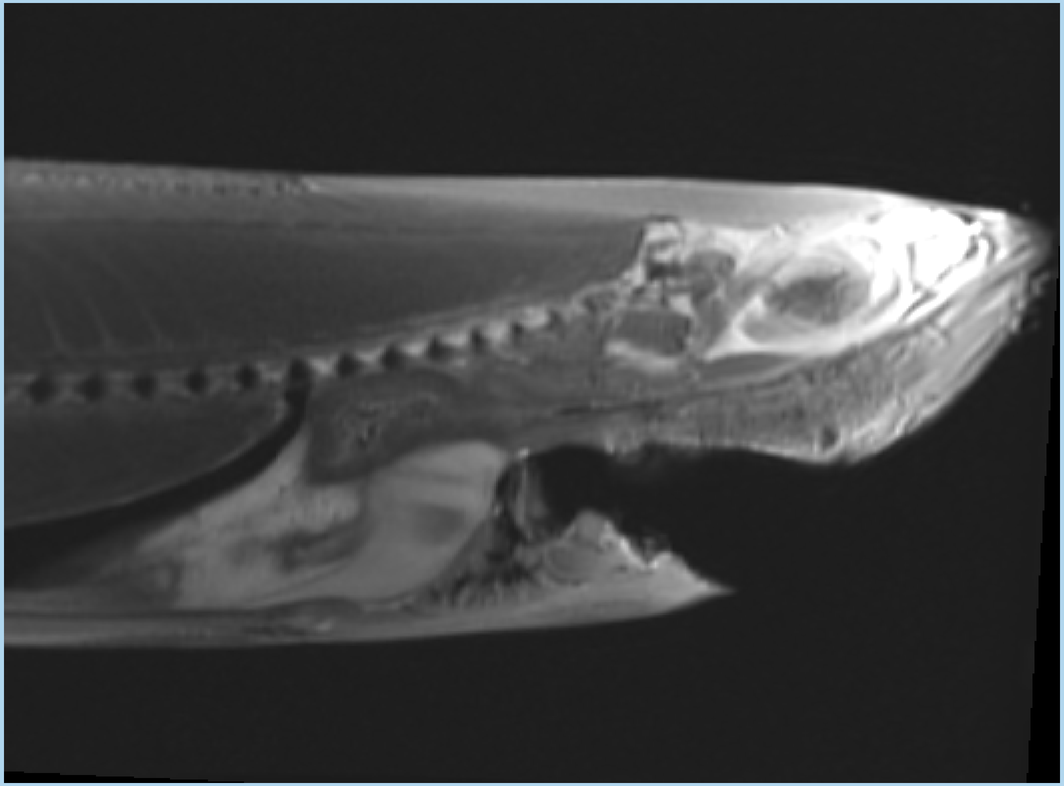
Voxel models – Variable accuracy



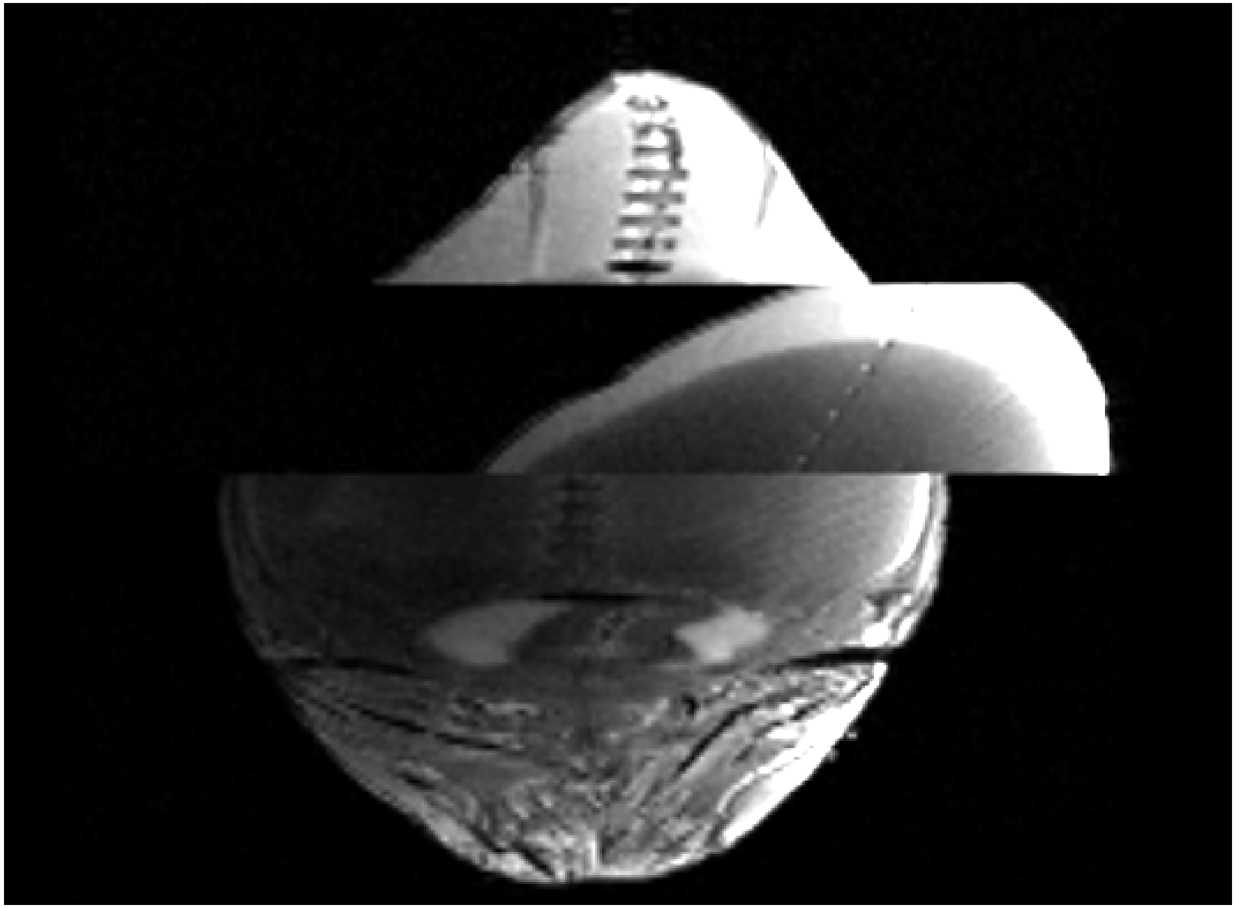




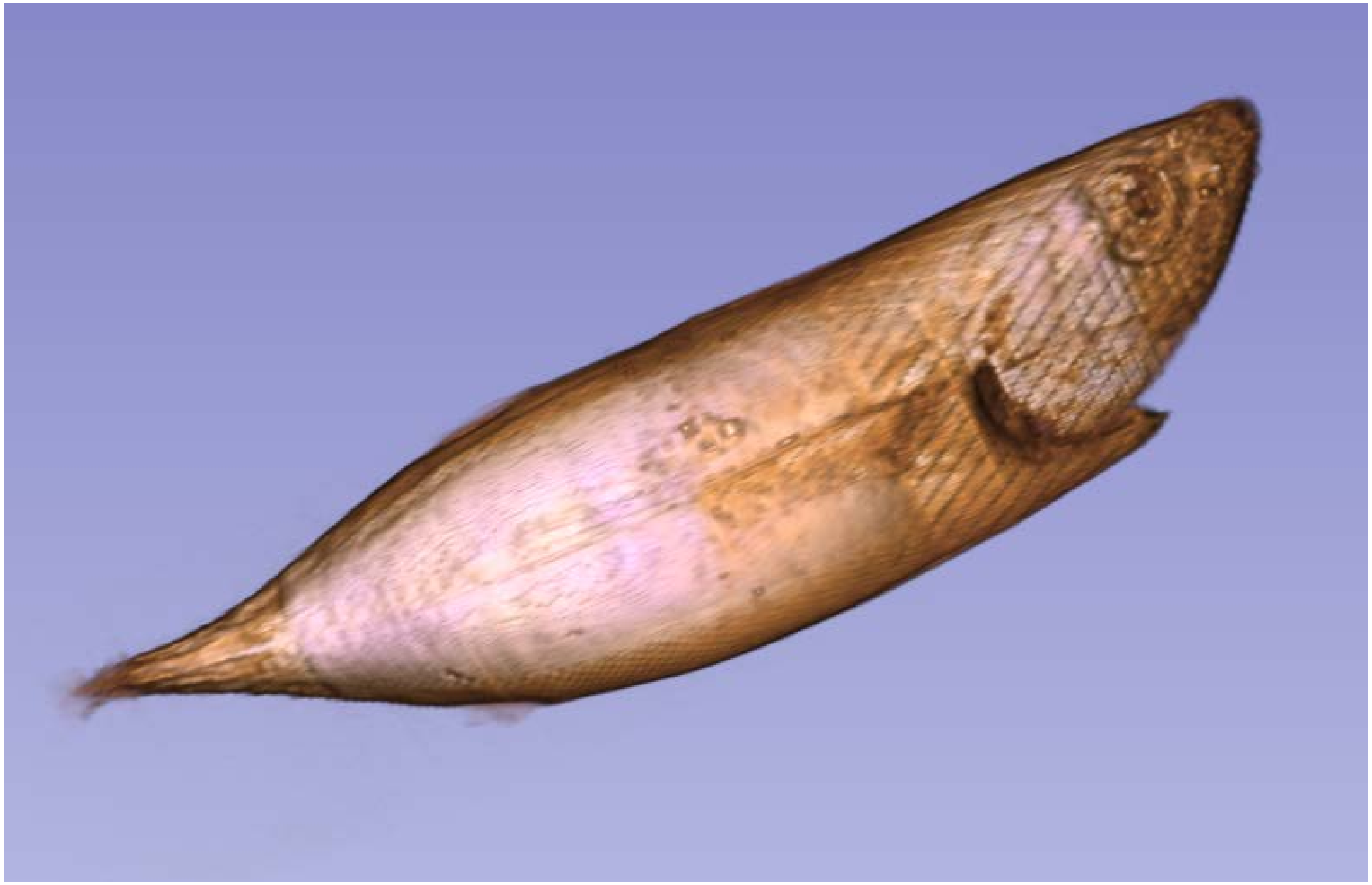
Imaging Albacore – Too big!



Stitch 3D medical images?



Stitch 3D medical images



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- Otsu & Uchida, 1962. Model of the migration of albacore in the North Pacific Ocean. Fishery Bulletin 63(1):33-44
- Ichinokawa et al. 2008. Transoceanic migration rates of young North Pacific albacore, Thunnus alalunga, from conventional tagging data. Canadian Journal of Fisheries and Aquatic Sciences 65: 1681-1691
- Childers, Snyder, Kohin, 2011. Migration and behavior of juvenile North Pacific albacore (Thunnus alalunga). Fisheries Oceanography 20(3): 157-173

Acknowledgements

- Kathryn Higley
- Jason Phillips
- Oregon Sea Grant
- Newport albacore fishing community

Questions for our presenters??

Upcoming Webinars

- Young Investigators Series: Novel Uranium Nanostructures
- Radiopharmaceutical Series: The History of Nuclear Medicine and Radiopharmaceuticals
- Radiopharmaceutical Series: Basics of Radiochemistry for Radiopharmaceuticals/Target, Ligand, Chemistry and Radiochemistry

NAMP website http://www.wipp.energy.gov/namp/