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Radioactivity in the marine environment: understanding the basics of radioecology

Claudia Benitez-Nelson, Sabine Charmasson, Ken Buesseler, Minhan Dai, Michio Aoyama, Núria Casacuberta, José Marcus Godoy, Andy Johnson, Vladimir Maderich, Pere Masqué, et al.

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LIMNOLOGY AND OCEANOGRAPHY

ASLO

e-Lectures



Radioactivity in the Marine Environment

Understanding the Basics of Radioecology

Claudia R. Benitez-Nelson

Sabine Charmasson, Ken Buesseler,
Minhan Dai, Michio Aoyama, Núria Casacuberta,
José Marcus Godoy, Andy Johnson,
Vladimir Maderich, Pere Masqué, Marc Metian,
Willard Moore, Paul J. Morris, John N. Smith

ASLO *e-Lectures*

What is Marine Radioecology?

Marine radioecology examines how **radioactive substances** interact with the marine environment and the various mechanisms and processes that influence **radionuclide migration** in the marine ecosystem.

This field of study includes aspects of field sampling, design of field and laboratory radiotracer* experiments, the development of predictive simulation models, and dose assessments to humans and biota.

** Includes radionuclides and radiolabeled compounds*

Courtesy of Scott Fowler

Goals of this Lecture

1. Overview of how marine biota uptake radionuclides from marine systems
2. Biogeochemical transfer and transport pathways of radionuclides in the marine environment
3. Radiation dose and exposure
4. Environmental protection: Understanding ecological risk assessment and management

Three main sources of radionuclides to the Marine Environment:

1. **U-Th series radionuclides** – of primordial origin, occur naturally on land and in ocean, and produce a series of “daughter” radionuclides via radioactive decay.

Examples: ^{238}U , ^{234}Th , ^{210}Pb , $^{223, 224, 226, 228}\text{Ra}$ and ^{222}Rn .

2. **Cosmogenic Radionuclides** – continuously being created by cosmogenic rays that interact with materials in the atmosphere and on Earth.

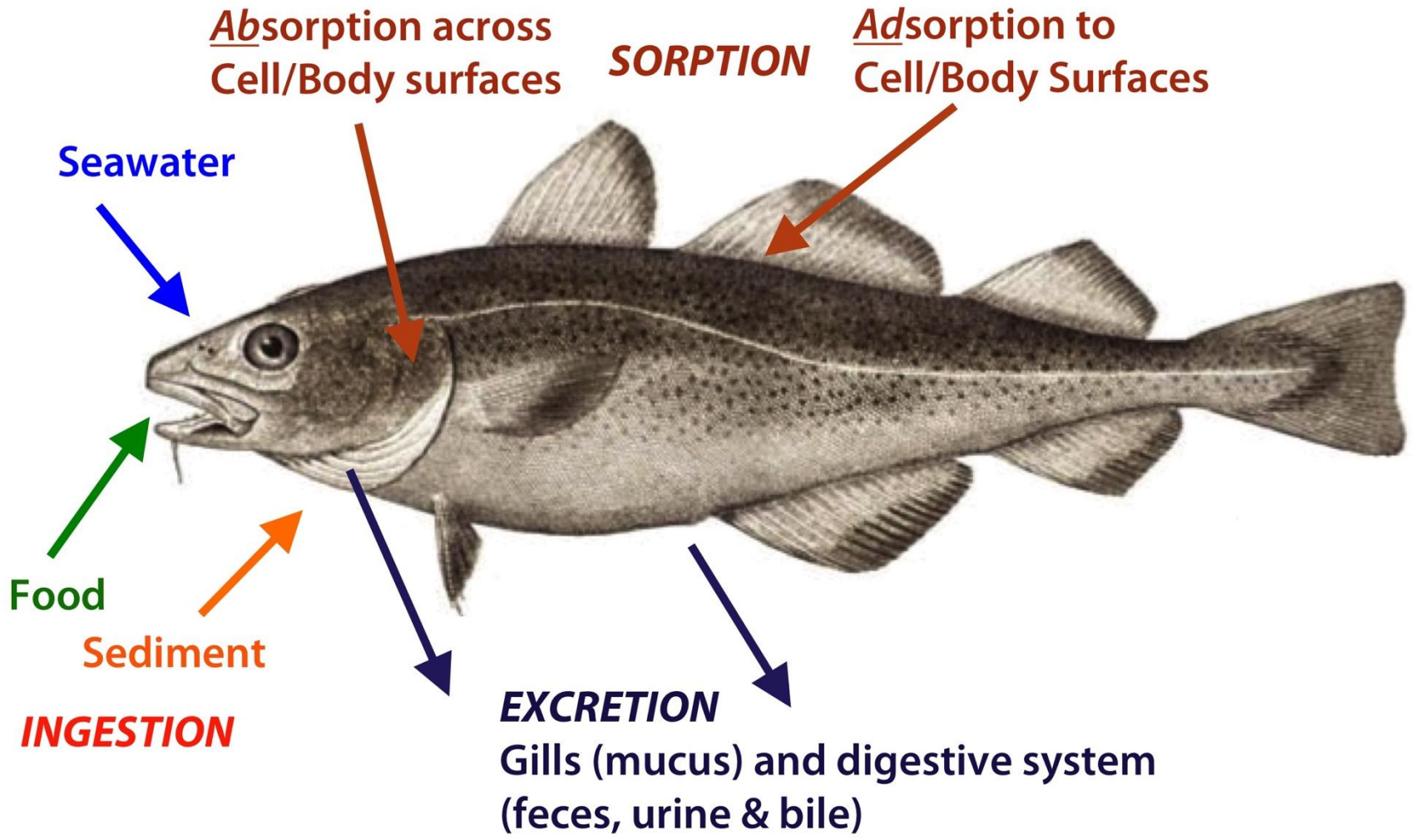
Examples: ^3H , ^{14}C , ^7Be

3. **Anthropogenic radionuclides** – continuously being produced by humans.

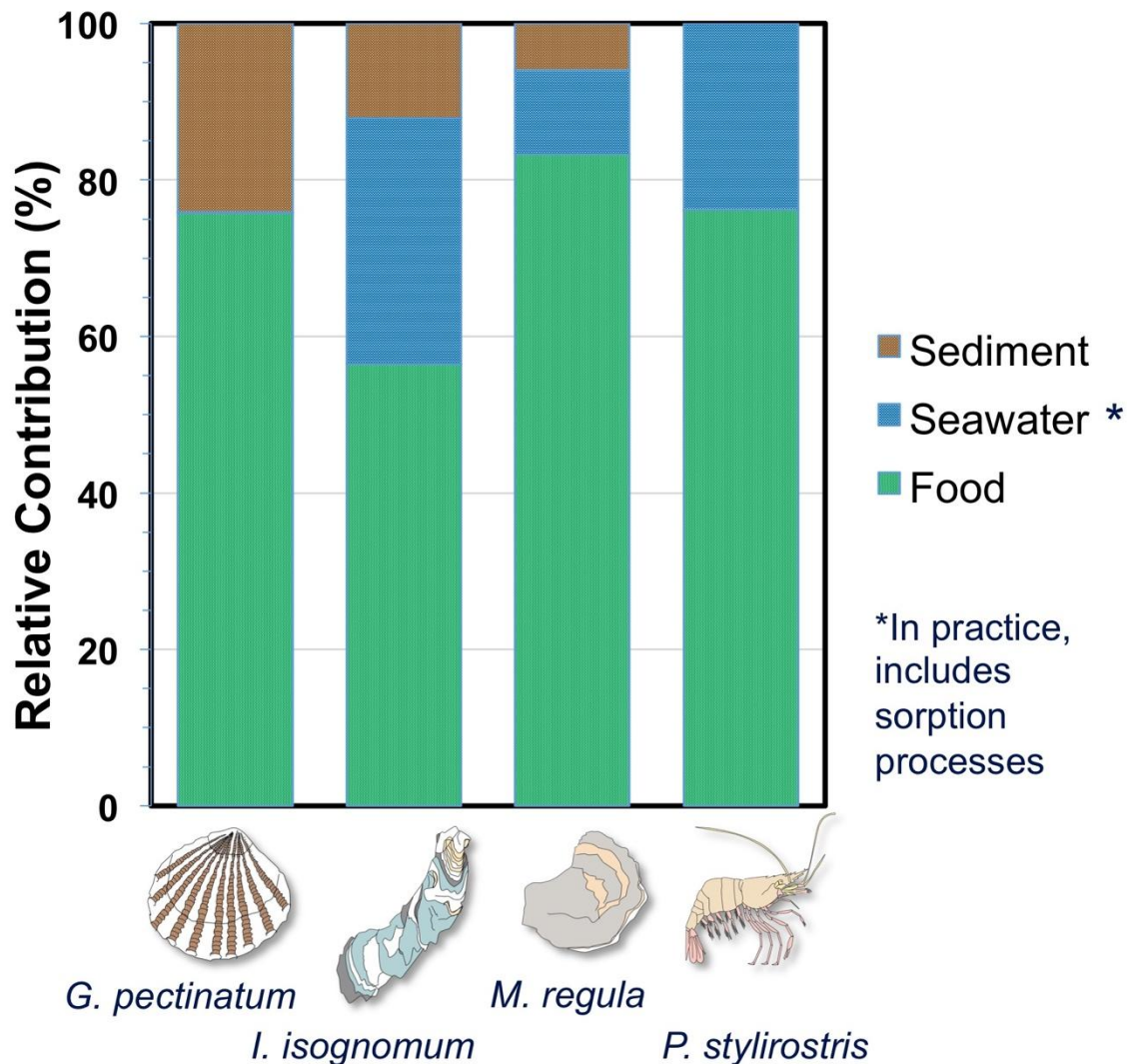
Examples: ^3H , ^{14}C , ^{90}Sr , ^{137}Cs , ^{129}I , $^{238, 239, 240}\text{Pu}$

Note: *Some radionuclides have both cosmogenic and anthropogenic sources (e.g., ^3H and ^{14}C).*

1. Radionuclide **bioaccumulation** in marine organisms?



1. Radionuclide **bioaccumulation** in marine organisms?



Metian et al. (2016)

Ingestion: Relative contributions (%) of three uptake pathways (sediment, seawater, and food) to the total bioaccumulation of ^{134}Cs in marine organisms.

Terminology

Concentration Factor (CF)*: Ratio of the radionuclide activity in a specific organism relative to the radionuclide concentration *in ambient sea water*.

$$\text{CF} = \frac{\text{Bq g}^{-1} \text{ wet weight of organism}}{\text{Bq g}^{-1} \text{ sea water}}$$

Transfer Factor (TF)*: Ratio of radionuclide activity in a specific organism relative to the radionuclide concentration *in sediment or food*.

$$\text{TF} = \frac{\text{Bq g}^{-1} \text{ wet weight of organism}}{\text{Bq g}^{-1} \text{ sediment or food}}$$

Uses:

- Compare relative bioavailability of different radionuclides to a given organism
- Compare ability of different organisms to accumulate a given radionuclide
- Allow the development of models that can predict the resulting radionuclide activity in an organism from known seawater activities.
- To identify potential “sentinel organisms” of radionuclide contamination.

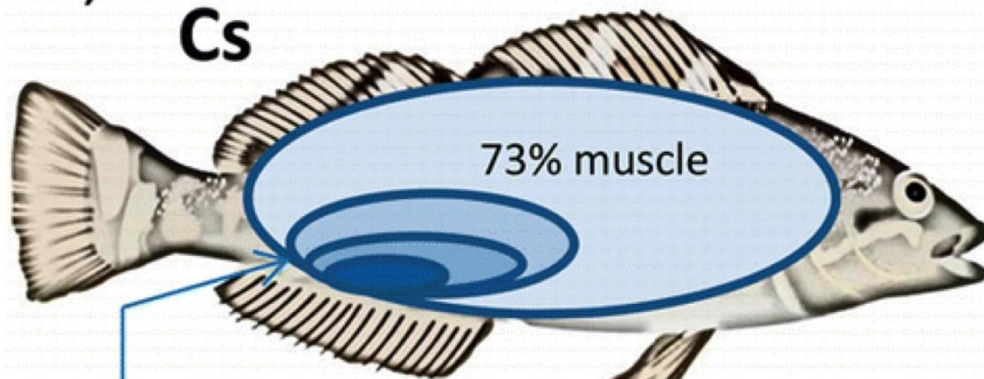
*Assumes input = output and that the radionuclide of interest exists in soluble forms in seawater

Carvalho et al. (2018)

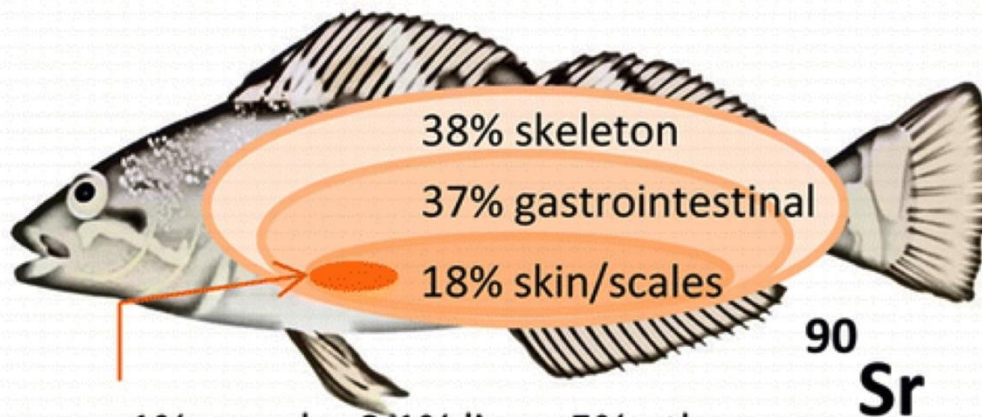
Distribution of $^{134,137}\text{Cs}$ and ^{90}Sr in fish tissue*

$^{134,137}\text{Cs}$

Cs



11% gastrointestinal, 9% skin/scales,
3% skeleton, 2% gills, 1% liver, 1% testes/ovary.



1% muscle, 0.1% liver, 5% other.

^{90}Sr

Johansen et al. (2015)

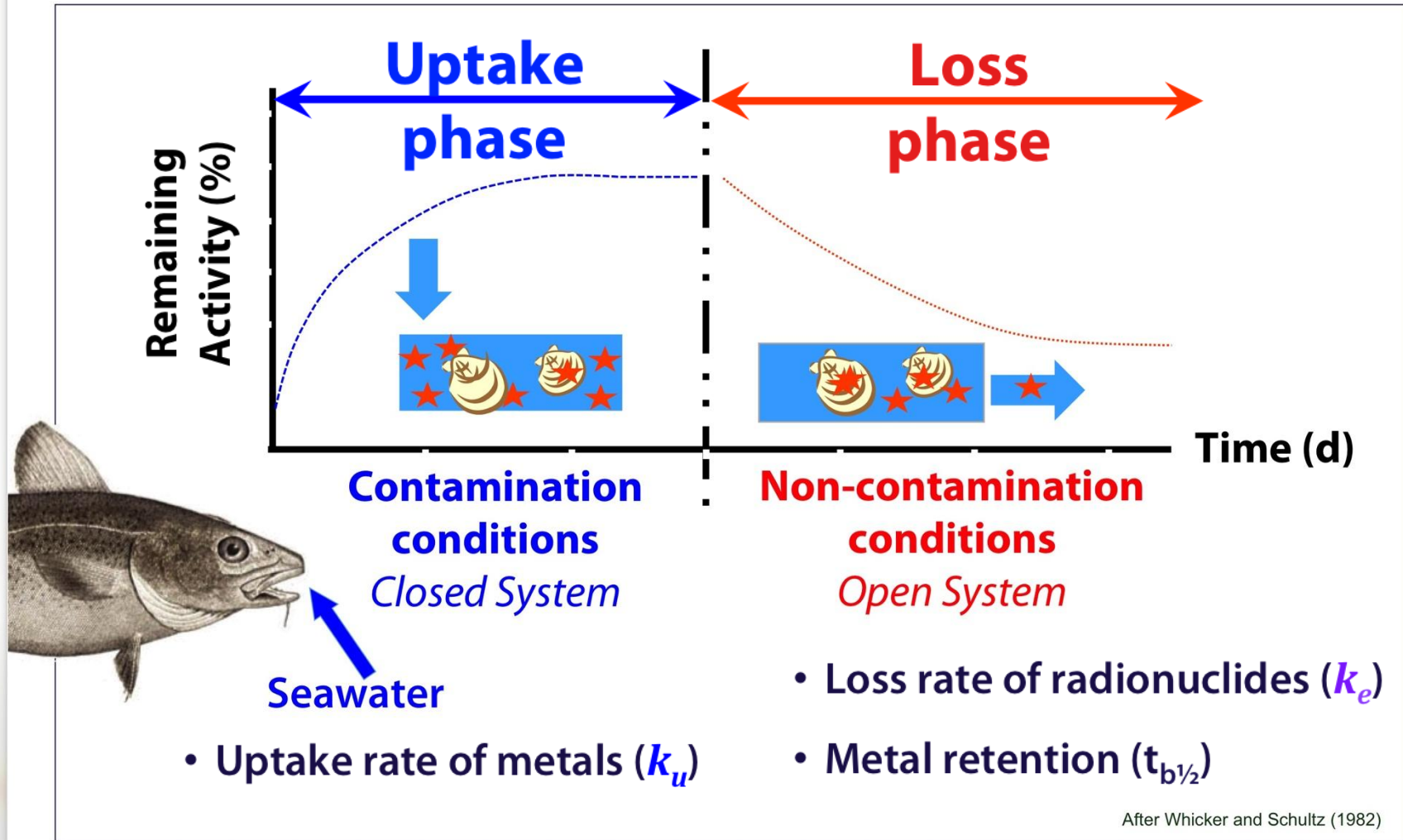
^{137}Cs is analog to ^{40}K

^{90}Sr is analog to Ca

* 2 years after the
Fukushima accident

How to determine the CF: (Lab experiments)

Model development of radionuclide contamination via seawater



How to determine the CF

Uptake phase: Depends on time (kinetic parameters)

$$CF_t = k_u t \quad (\text{During uptake phase, } k_u \text{ dominates})$$

$$CF_t = CF_{ss} (1 - e^{-k_e t}), \text{ where at steady state } CF_{ss} = \frac{k_u}{k_e}$$

Where:

CF_t - the concentration factor at time t (d)

CF_{ss} - the concentration factor at steady state

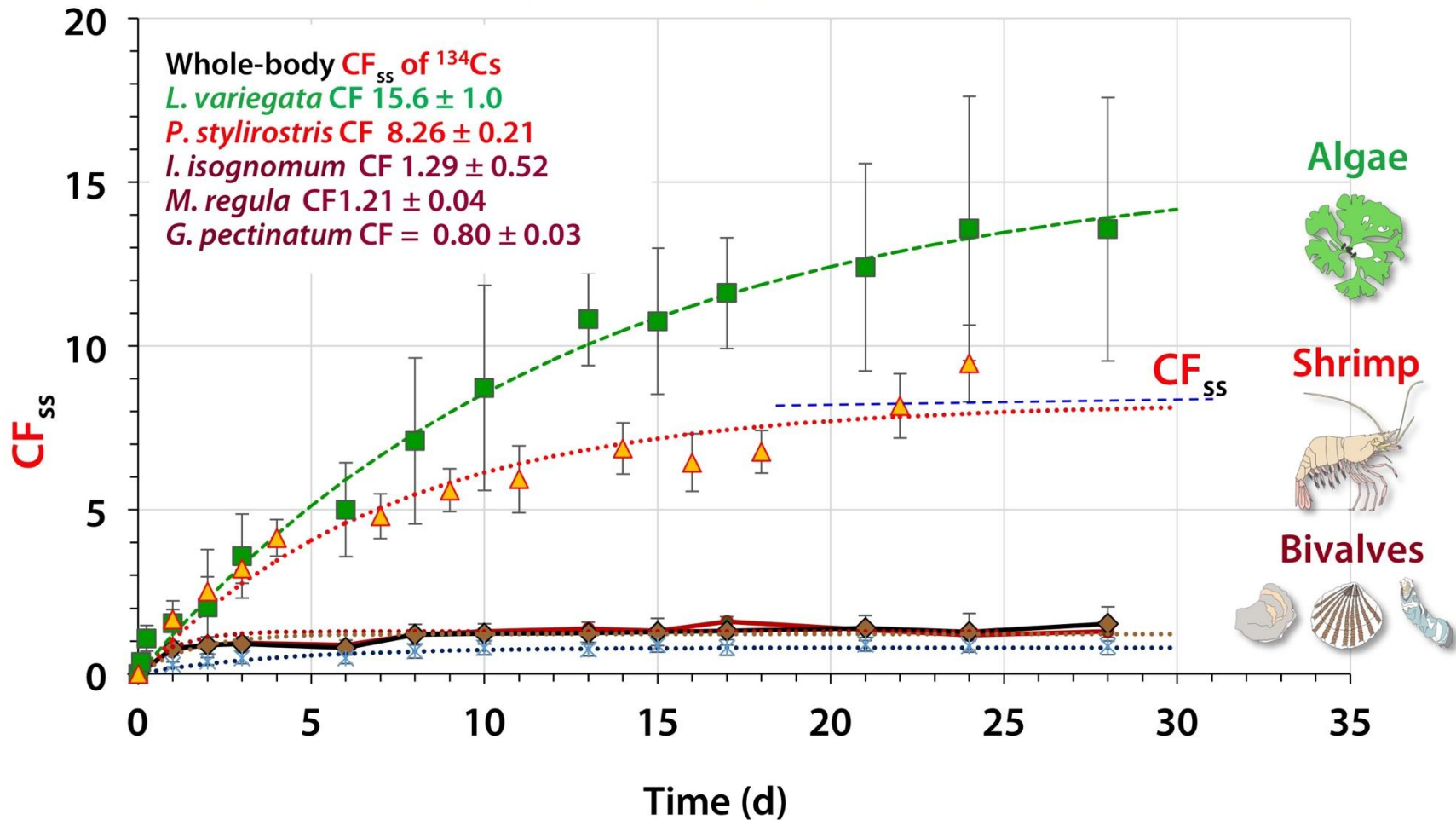
k_u - uptake rate constant (d⁻¹)

k_e - loss rate constant (d⁻¹)

After Whicker and Schultz (1982)

How to determine the CF

Example: Uptake kinetics (k_u) of dissolved ^{134}Cs ($t_{1/2} = 2.06$ y) during 24 - 28 days of exposure



Metian et al. (2016)

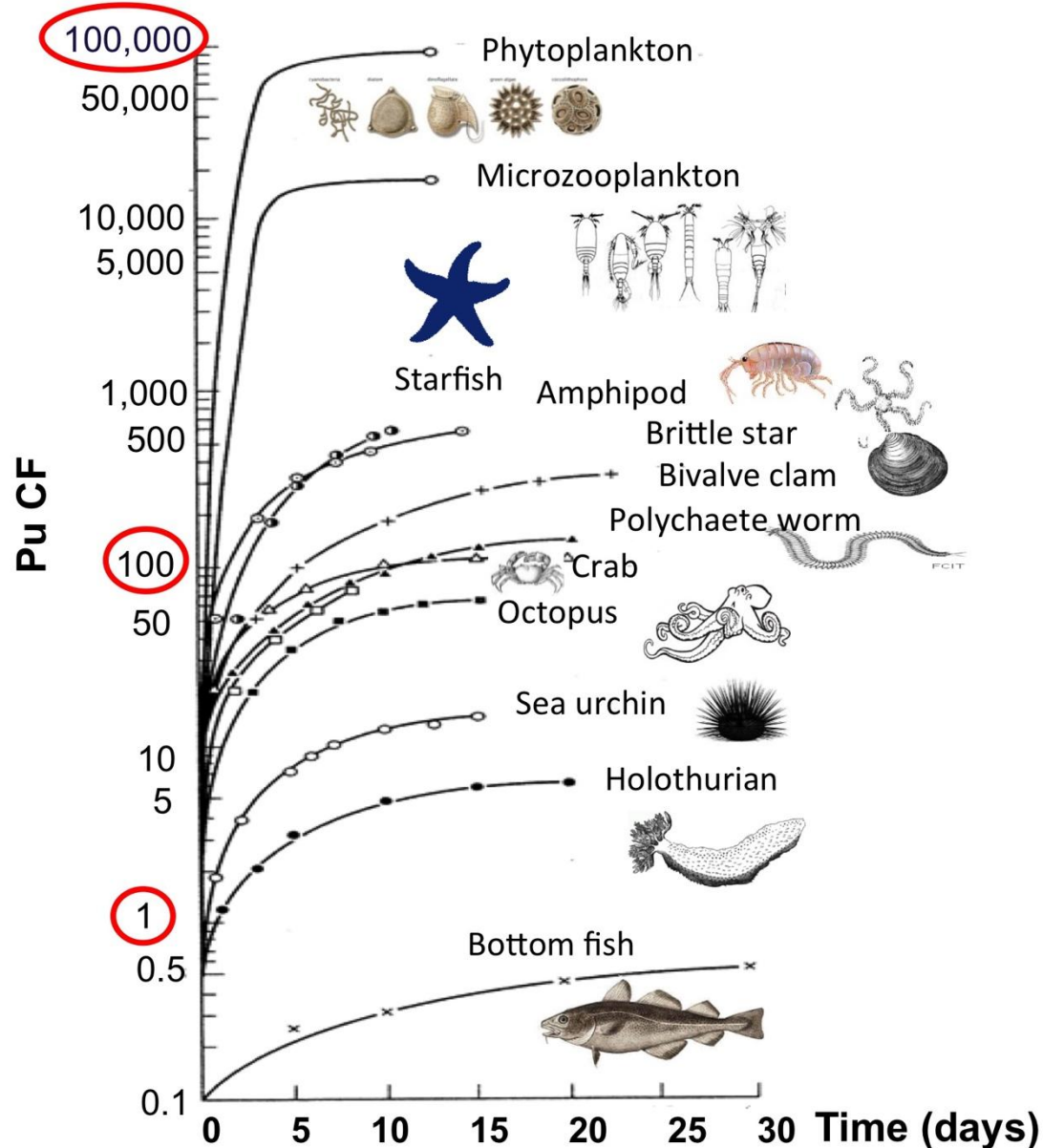


Figure courtesy of Dr. Scott Fowler modified from Fowler et al. (1975)

Species dependent uptake kinetics of **plutonium** from seawater by various marine organisms

Note the variability in k_u

Effect of **Temperature** on the **CF** (specifically k_u) of ^{60}Co , ^{241}Am and ^{134}Cs from water by brown macroalgae

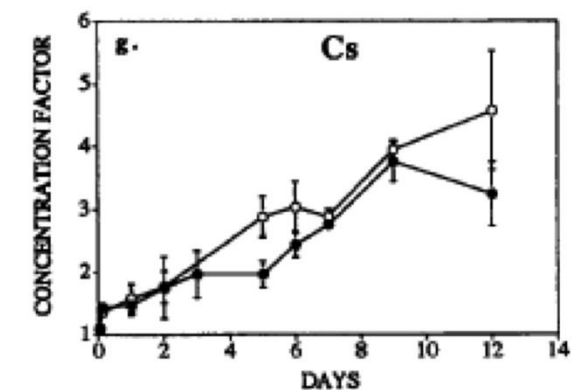
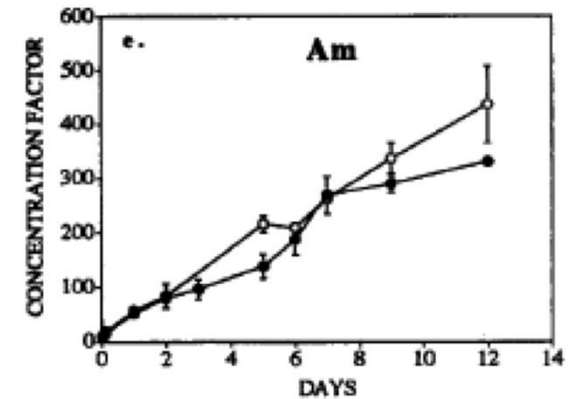
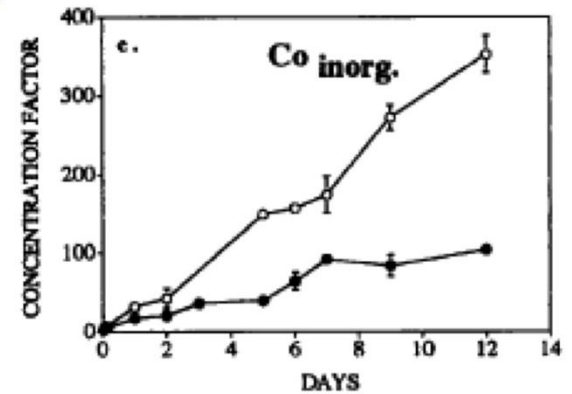


Fucus vesiculosus

Image: http://www.seaweed.ie/descriptions/fucus_vesiculosus.php

12°C (○)
2°C (●)

(Boisson et al, 1997)



Loss (depuration) phase: kinetic parameters (k_e)

Loss kinetics are used to determine the **biological half-life** ($t_{b1/2}$) and can be modeled using a *one component* exponential model:

$$A_t = A_0 e^{-k_e t}$$

Where:

A_t - Remaining activities at time t (%)

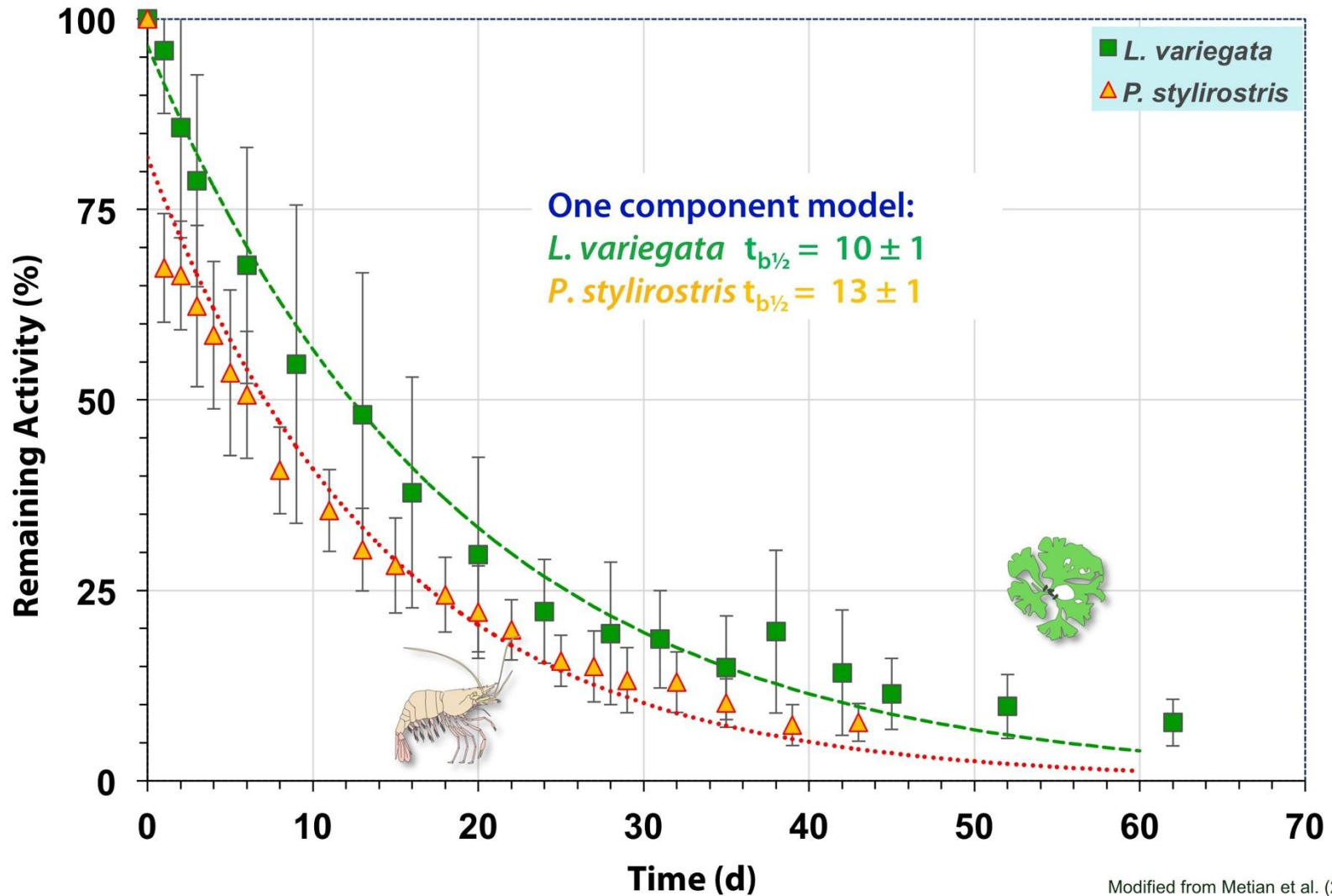
A_0 - Remaining activities at time 0 (%)

k_e - Loss rate constant (d^{-1})

$$T_{b1/2} = \ln(2)/k_e$$

After Whicker and Schultz (1982)

Example: ^{134}Cs ($t_{1/2} = 2.06$ y) depuration kinetics when maintained for 43 – 62 d in clean seawater



Modified from Metian et al. (2016)

Loss (depuration) phase: kinetic parameters (k_e)

In some cases, the loss phase should be described by a *two component model* that may reflect differences in specific tissues where the radionuclide has accumulated.

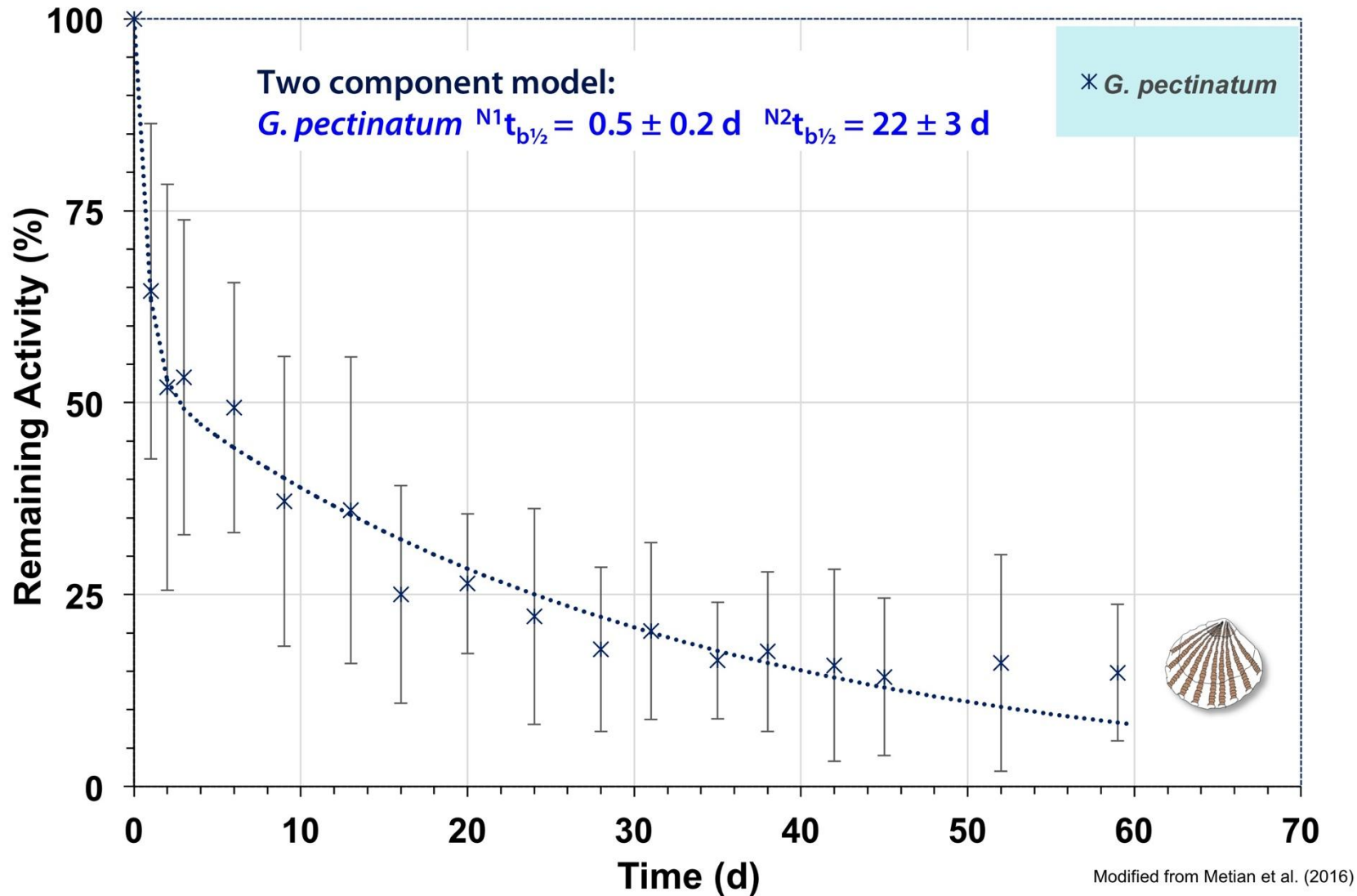
$$A_t = A_0(N_1 * e^{-k_{e1}*t} + N_2 * e^{-k_{e2}*t})$$

- Where:**
- A_t - Remaining activities at time t (%)
 - A_0 - Remaining activities at time 0 (%)
 - k_e - Loss rate constant (d^{-1}) for a specific component N_1 and N_2
 - N_1 and N_2 - Denote differences in the loss rates with respect to specific components (e.g., tissues vs. shell) within an organism, where $N_1 + N_2 = 1$

Biological half-lives (T_{b1} & T_{b2}) can be calculated from the corresponding depuration rate constant (k_{e1} & k_{e2}) according to the relation $T_b = \ln(2)/k_e$

After Whicker and Schultz (1982)

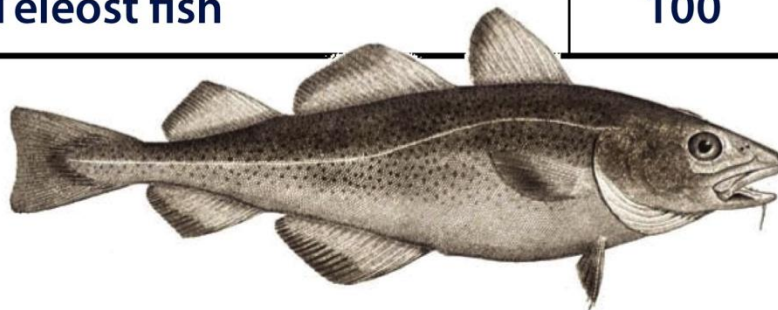
Example: ^{134}Cs ($t_{1/2} = 2.06$ y) depuration kinetics when maintained for 43 – 62 d in clean seawater



CF_{SS} of selected radionuclides within different taxonomic groups

Organism	¹³⁷ Cs	²³⁹⁺²⁴⁰ Pu	²⁴¹ Am	²¹⁰ Po
Macroalgae	50	4,000	8,000	1,000
Phytoplankton	20	200,000	200,000	70,000
Zooplankton	40	4,000	4,000	30,000
Decapod crustaceans	50	200	400	20,000
Bivalve Molluscs	60	300	1 000	20,000
Cephalopods	9	50	100	20,000
Teleost fish	100	100	100	2,000

IAEA (2004)

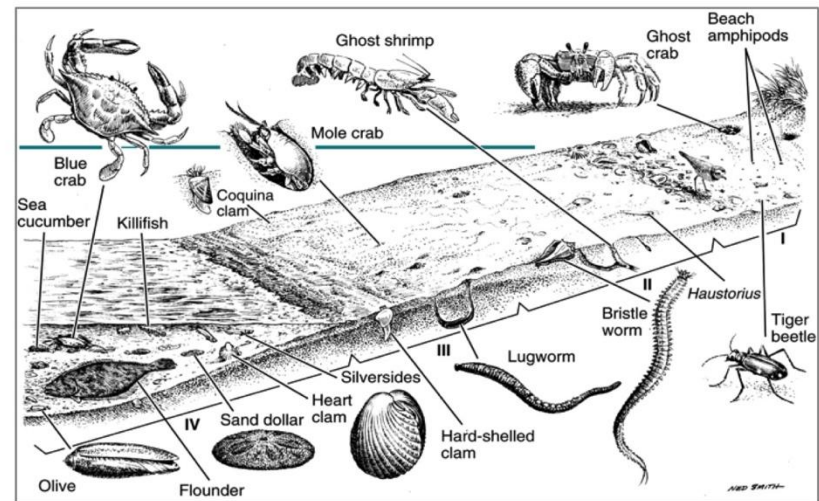


← Seawater

TF of radionuclides accumulated from contaminated **sediments**

Organism	Uptake Time (days)	$^{239+240}\text{Pu}$	^{241}Am	^{137}Cs	^{60}Co
Worms	11 - 50	0.0016 - 0.002	0.0009 - 0.003	0.2	0.06
Clams	40 - 50	0.006 - 0.01	0.004 - 0.02	--	--
Isopod	40 - 50	--	0.006-0.032		
Amphipod	14	0.10	0.11		

Fowler et al. (1997), IAEA (1985)



Assimilation Efficiencies* of selected radionuclides within different taxonomic groups

*AE = The fraction of *ingested radionuclide via food* that is absorbed and retained in body tissues

Organism Food type	^{137}Cs	$^{239+240}\text{Pu}$	^{241}Am	^{210}Po
Zooplankton feeding on Phytoplankton	-	0.8 – 1	0.9 – 10	20 – 55
Decapod crustaceans feeding on artemia	-	10 – 60	8 – 58	5
Bivalve Molluscs feeding on phytoplankton	3 – 4	0.9	0.6 – 38	17
Cephalopods feeding on crabs	23 – 29	-	51 – 60	-
Teleost fish feeding on shrimp	42 – 95	0.1 – 1	0.7 – 6	5

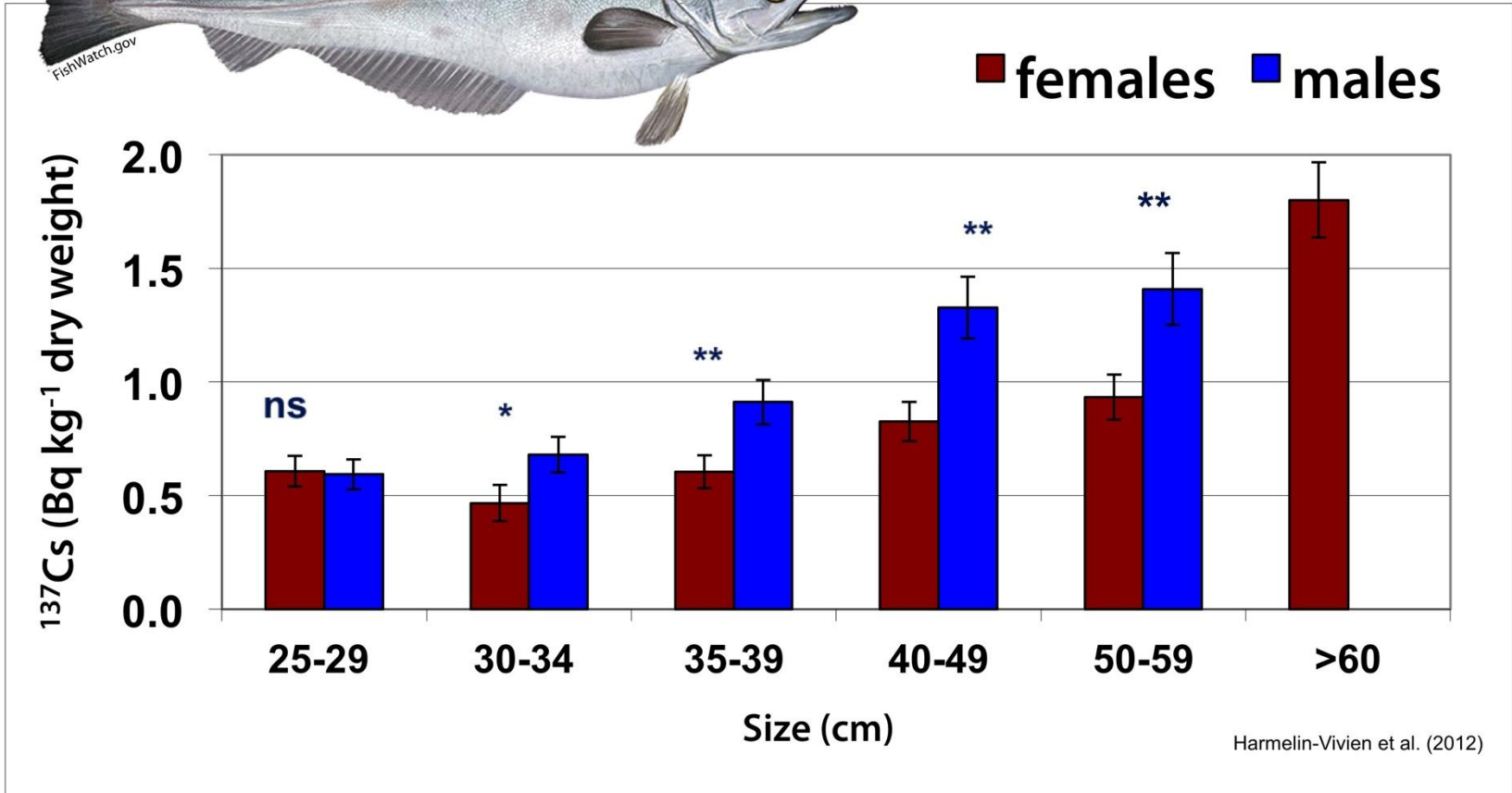
Stewart et al. (2005); Fowler et al. (1975; 2005)



← Food

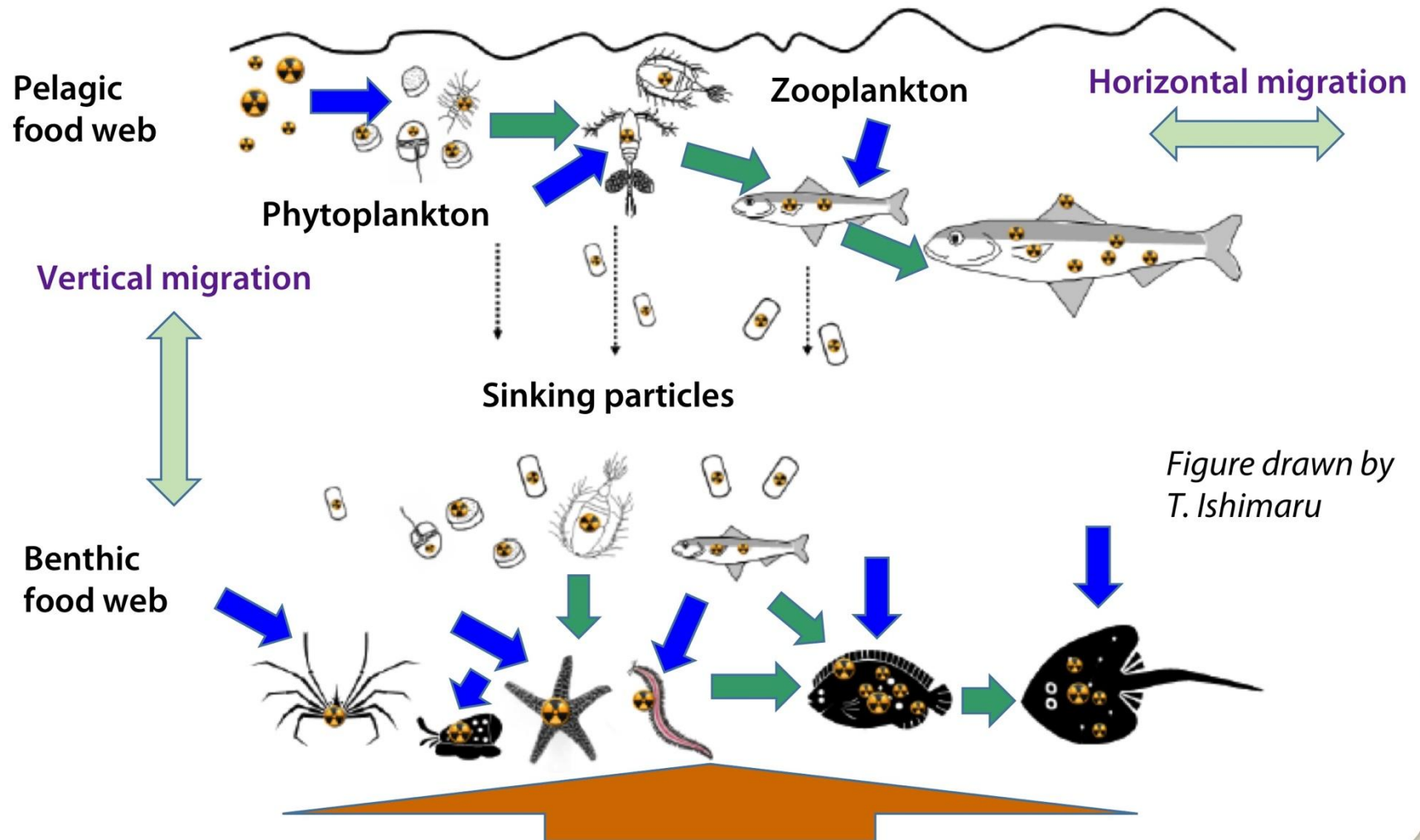
Combined ingestion and excretion pathways

Bulk ^{137}Cs activities in different *size classes* of *male and female* European Hake



Significance of male-female difference: ns = not significant, * $p < 0.05$, ** $p < 0.01$

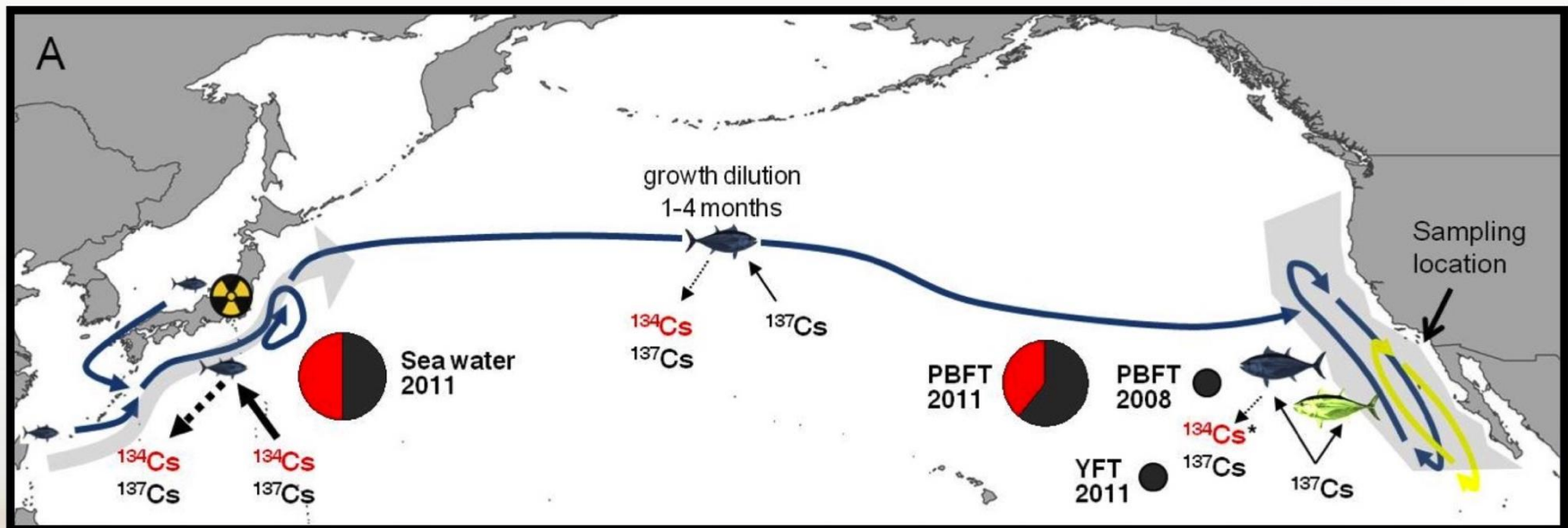
2. Biogeochemical transfer and transport pathways of radionuclides in the marine environment



Horizontal Transport

Here, Pacific Bluefin Tuna (PBFT) transported ^{134}Cs during their migration across the Pacific Ocean after having been contaminated near Japan by the releases of the Fukushima Dai-ichi nuclear power plant.

Note that Yellowfin Tuna (YFT) contained none of the short lived ^{134}Cs , confirming that transport was not by water movement alone, but by **active** horizontal transport by the PBFT.



Madigan et al., 2012

Vertical Transport

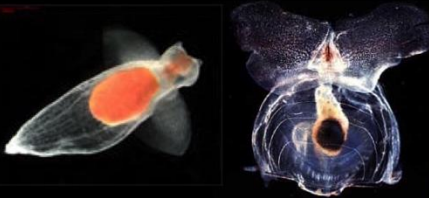
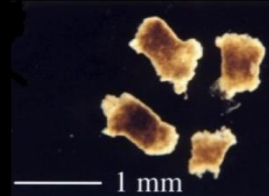
Production of fecal pellets is an efficient way for transporting contaminants to the seabed

Zooplankton

Zooplankton Fecal Pellets



Salps



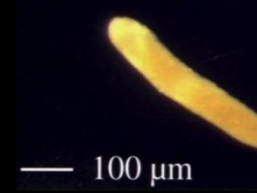
Pteropods
(Gymnosomata
& Thecosomata)



Euphausiids



Copepods



Vertical Transport

Concentrations of selected radionuclides (Bq kg⁻¹ dry) in plankton and sinking particulate matter in the water column

Organism	²³⁹⁺²⁴⁰ Pu	²⁴¹ Am	²¹⁰ Po	¹³⁷ Cs	¹³⁴ Cs
Phytoplankton	0.35 – 1.0	0.052 - 0.22	21 – 61	-	-
Copepods	0.05 – 0.48	0.022 - 0.12	126	*34 ± 7	*22 ± 6
Fecal pellets	1.2 - 3.4	1.0 - 2.7	617	*6300 ± 1000	*3400 ± 600
Euphausiids	0.015	-	40.7	-	-
Fecal Pellets	3.6	-	648 – 1000	-	-
Molts	0.18 (90%)	-	13.3 (2.5%)	-	-
Salps	-	-	260	-	-
Fecal pellets	8.9	5.0	583 - 750	-	-
† Japanese Copepods (Pre – Fukushima)	-	-	-	0.42 ± 0.15	-
** Japanese Copepods (Post – Fukushima)	-	-	-	0 - 56	0 - 46

Courtesy Scott Fowler

* Samples are Chernobyl fallout-enriched (Fowler et al., 1987)

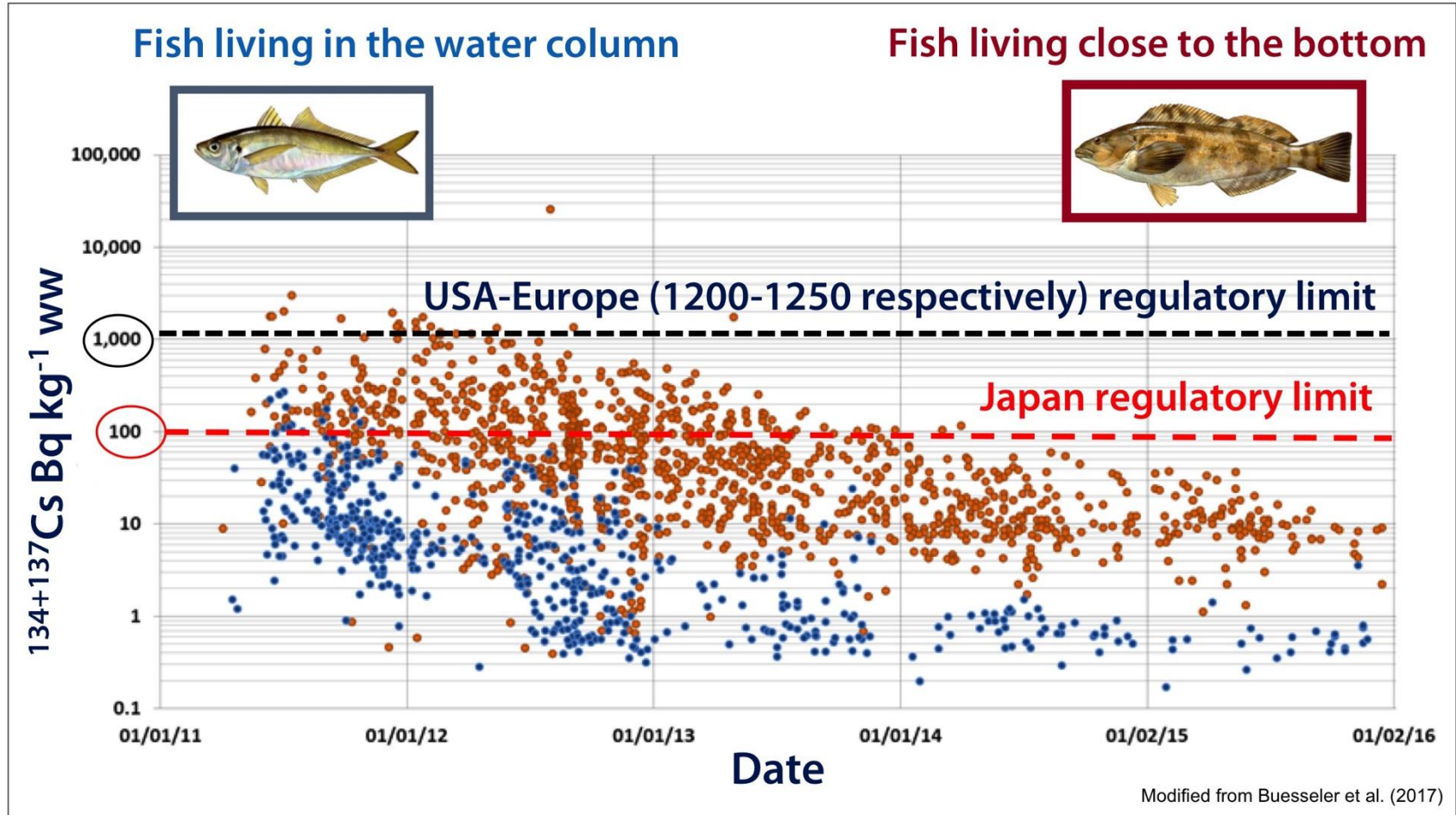
† from Tateda (1998)

** from Buessler et al. (2012)

() = Percent of euphausiid whole body radionuclide concentration contained in the molt

Vertical Transport

Following accidental release from Fukushima Dai-ichi Nuclear Power Plant
Example: radiocesium in fish (Bq kg⁻¹ ww)

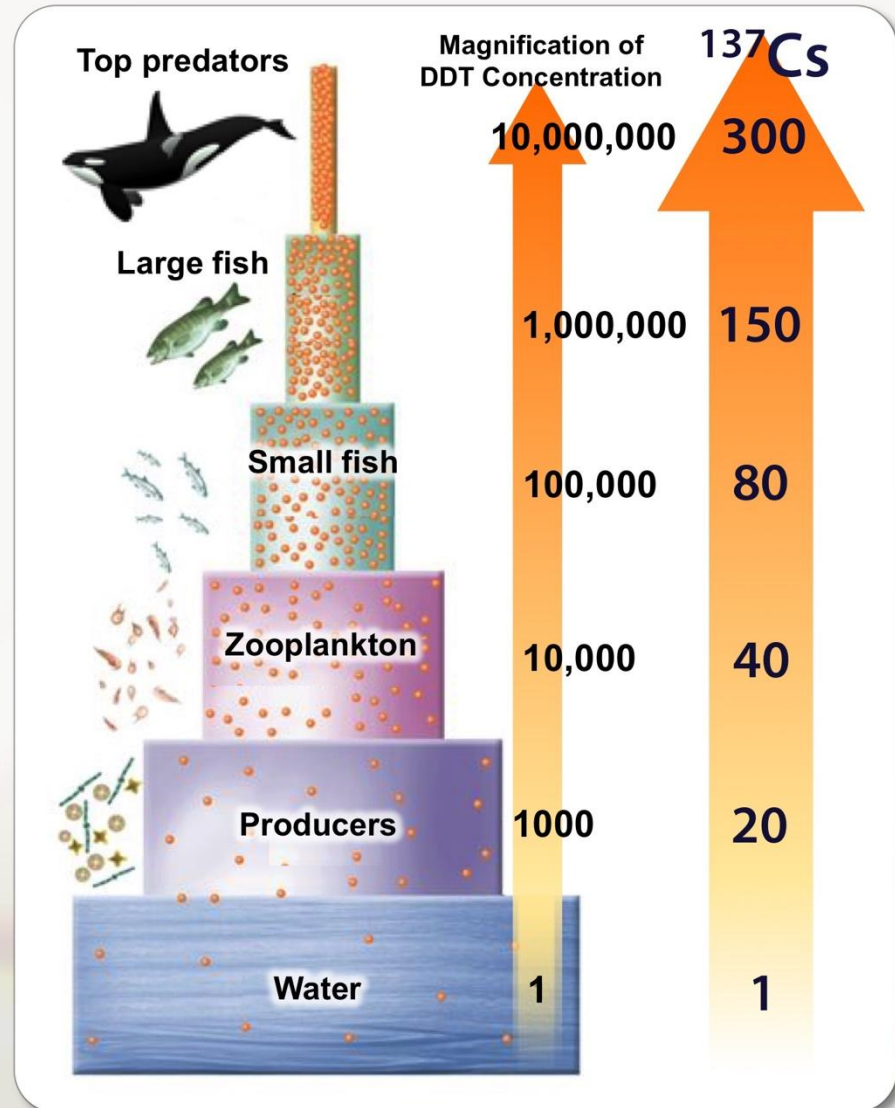


Transport *within* food chains: Do radionuclides biomagnify in marine food chains?

Biomagnification = increasing concentrations in organisms at successively higher levels in the food chain.

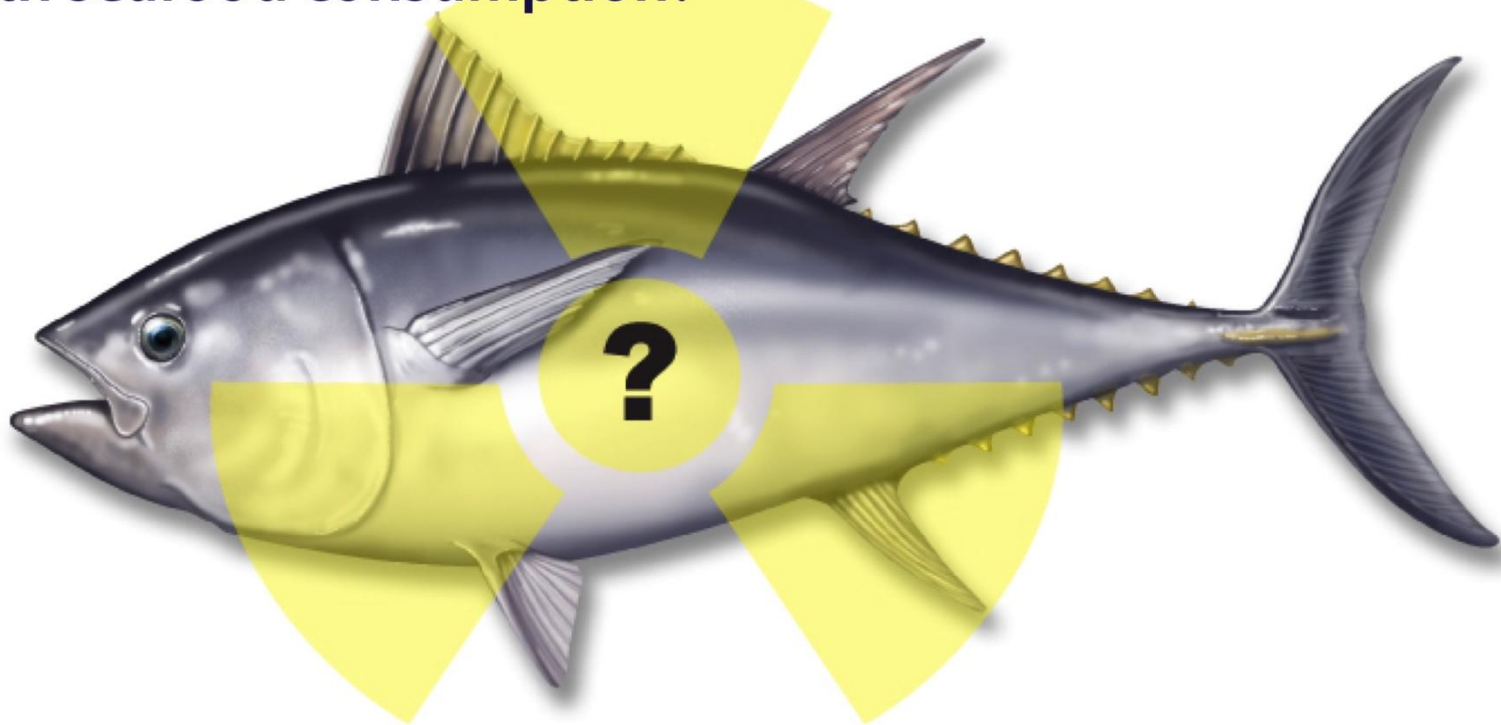
A limited number of substances magnify in marine food chains. For example: **mercury, PCBs, and DDT.**

Cesium and **Polonium** are radionuclides that have *limited biomagnification.*



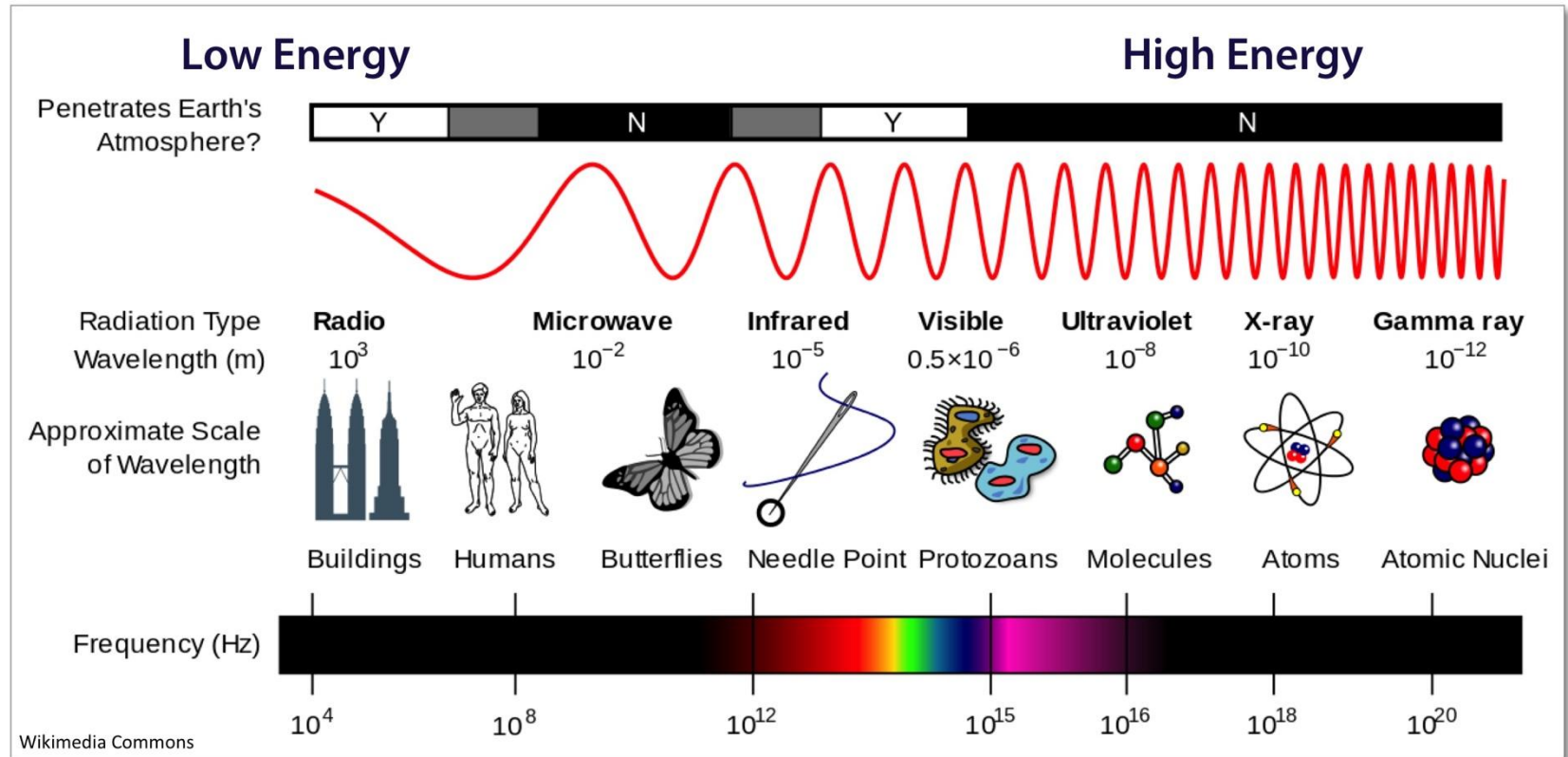
Toxic radionuclides in marine food chains

Once radionuclides are released into the environment and incorporated into marine organisms, how do we assess *potential impacts* on the environment and *risks* associated with seafood consumption?



3. What is Radiation?

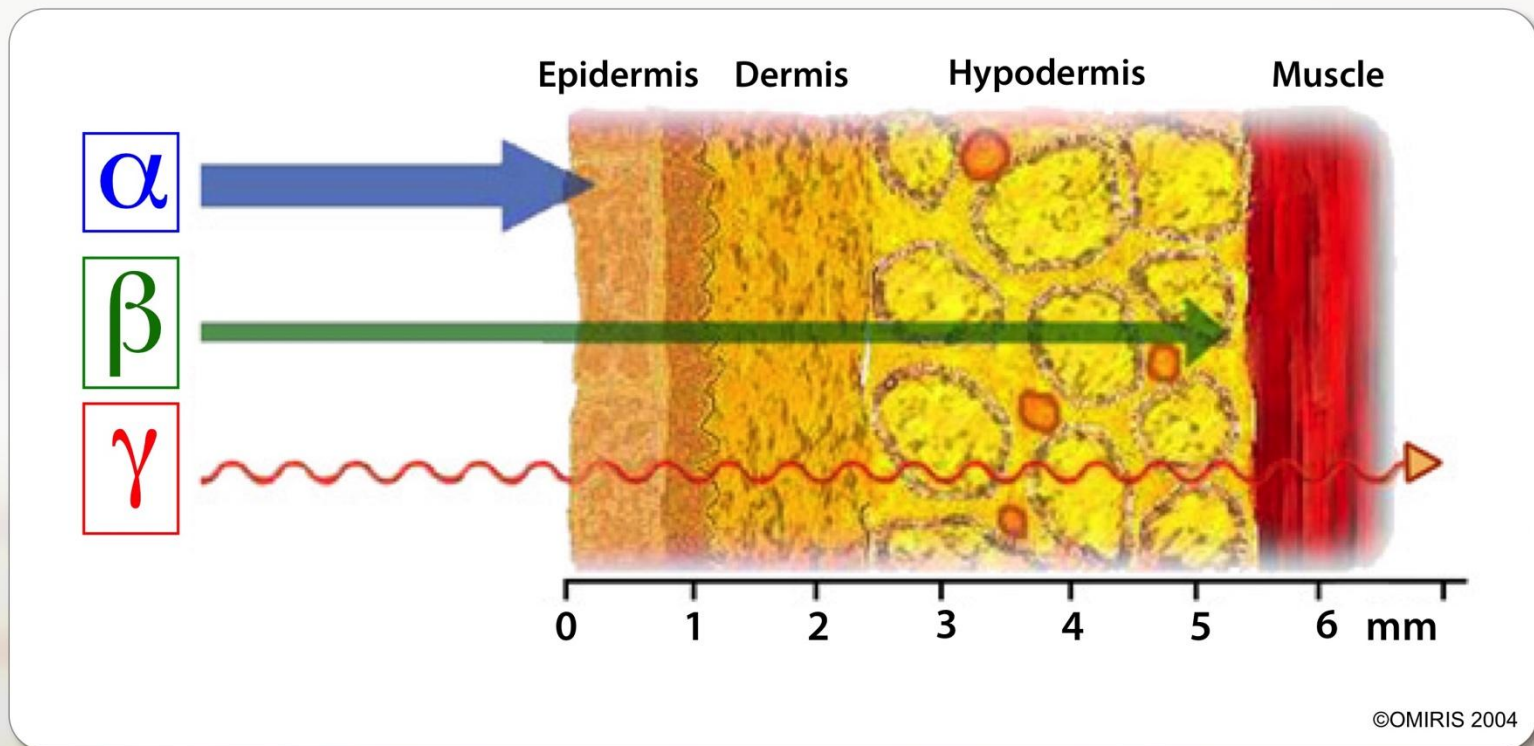
Radiation is energy in the form of high-speed particles (or electromagnetic waves or photons). It can be ionizing or non-ionizing.



Non-ionizing radiation lacks the energy to alter atoms (e.g., visible light and microwaves). **Ionizing radiation has enough energy to change normal cellular functioning.** Ionizing radiation may cause cells to die or transform into a cancerous cell. **It is these particles that are emitted from a radioactive nuclide.**

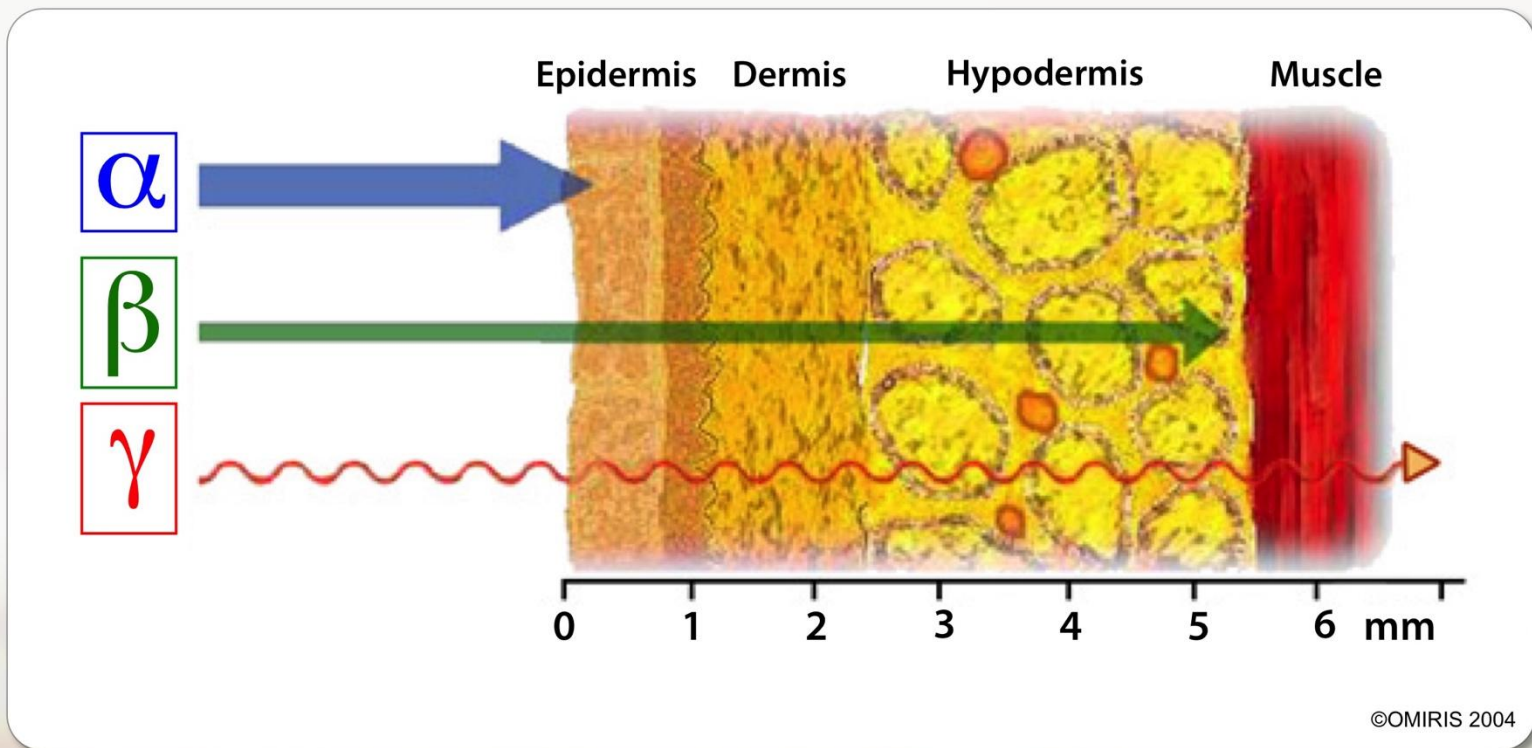
Radiation types we primarily focus on:

Alpha (α) particles: Most densely ionizing, but least penetrating. This means that cells can be protected or shielded from damage by alpha particles by clothing. Even the dead outer layer of your skin will protect you from damage from alpha particles. However, if alpha emitters are **inhaled or ingested or get into a cut on the skin, they can cause damage to cells. As alpha particles are emitted inside the body, the surrounding cells are damaged.**



Radiation types we primarily focus on:

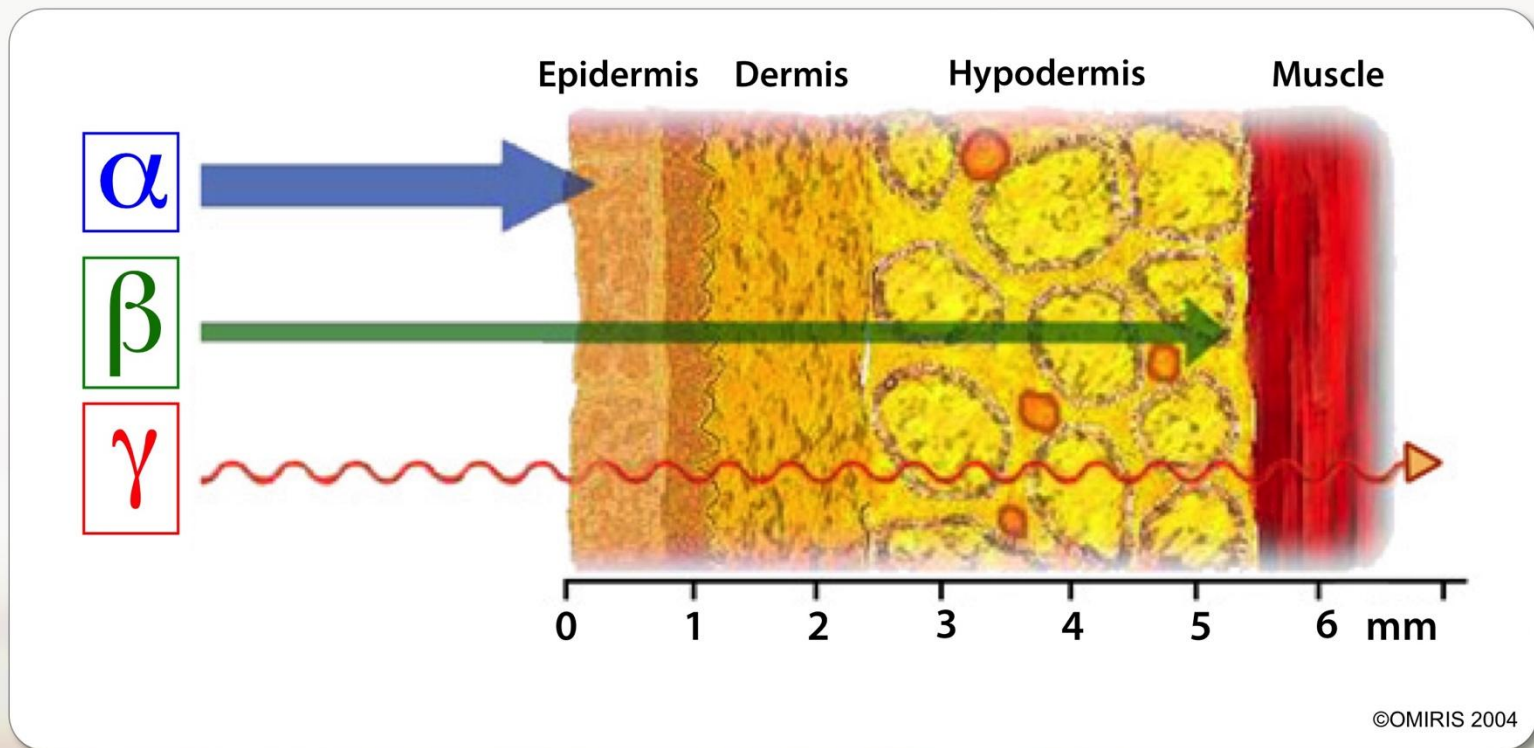
Beta (β) particles: More energetic. Can travel several feet through air. Are stopped with denser materials such as wood, glass, or aluminum foil.



Radiation types we primarily focus on:

Gamma (γ) rays:

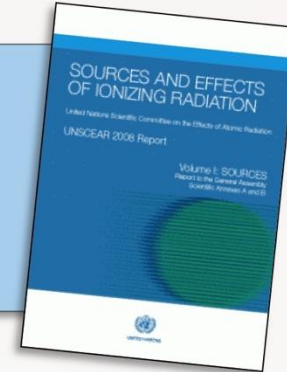
High-energy electromagnetic energy waves and the most penetrating type of radiation. Cells must be shielded from gamma rays with concrete, lead or steel. Not all may do cellular damage, but they must interact with the material to do so.



Development of International Standards

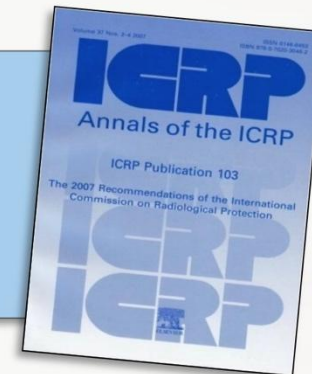
SCIENCE
Doses and effects

UNSCEAR
www.unscear.org



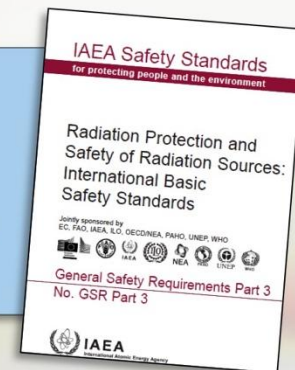
PRINCIPLES
Philosophy and policy

ICRP
www.icrp.org



STANDARDS
Nuclear Safety and Security

IAEA
www.iaea.org



Radiation Dose Concepts

- Adsorbed dose (**Gray, Gy**) is the absorption of radiation per unit mass of tissue. $1 \text{ Gy} = 1 \text{ Joule per kg}$

For humans only:

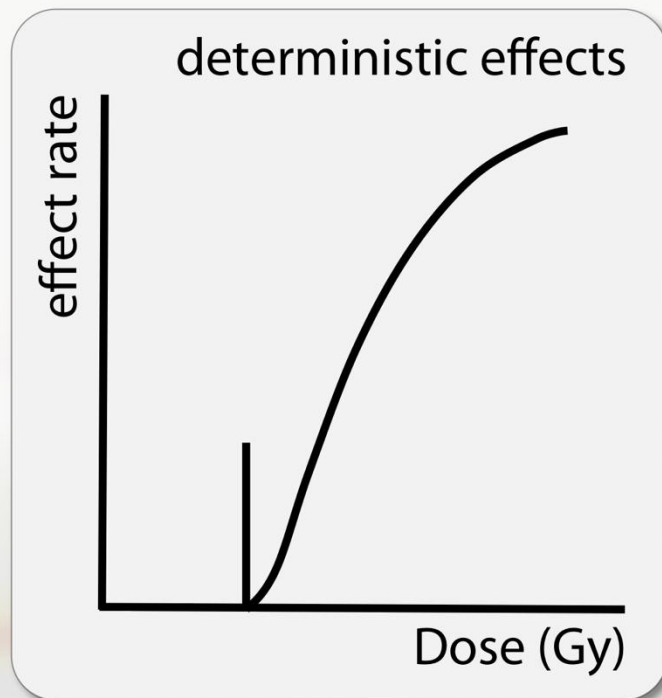
- Equivalent dose (**Sievert, Sv**) adjusts for biological damage by different types of ionizing radiation (α, β, γ) using a weighting factor (W_R)

$$W_R = 1 (\beta, \gamma, X) \text{ and } 20 (\alpha)$$

- Effective dose (**Sievert, Sv**) is the tissue-weighted sum of the equivalent doses in all specified tissues and organs of the human body.

Different types of Effects: **Deterministic** versus **Stochastic**

Deterministic effects: Severity increases with the dose. Radiation dose levels, or “thresholds”, are determined, below which, effects from radiation exposure are absent. Generally they are short term effects and specific to the *individual*.



Dose Threshold for Deterministic Effects*

Tissue	Total Acute Dose Threshold (Gy)	Time to Develop Effect
<u>Lens of eye</u>		
Detectable opacities	0.5–2	>1 year
Cataract formation	5	>1 year
<u>Skin</u>		
Skin reddening	3–6	1–4 weeks
Temporary hair loss	4	2–3 weeks
Skin death and scarring	5–10	1–4 weeks
<u>Testes</u>		
Temporary sterility	0.15	3–9 weeks
Permanent sterility	3.5–6	3 weeks
<u>Ovaries</u>		
Permanent sterility	2.5–6	< 1 week
<u>Gastrointestinal</u>		
Mucosa lining loss	6	6–9 days
<u>Bone marrow</u>		
Reduction of blood cell production	0.5	1–2 months

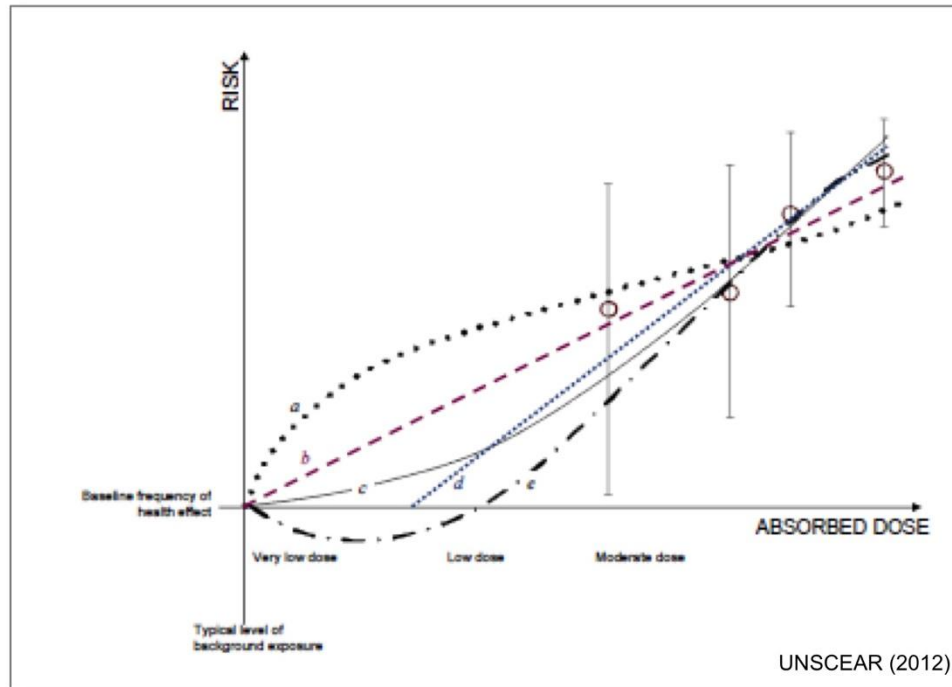
* 1% incidence level based on ICRP publication 103 (2007)

From Peck and Samei (2017)

Different types of Effects: **Deterministic** versus **Stochastic**

Stochastic effects:

Severity of stochastic effects is independent of the dose.



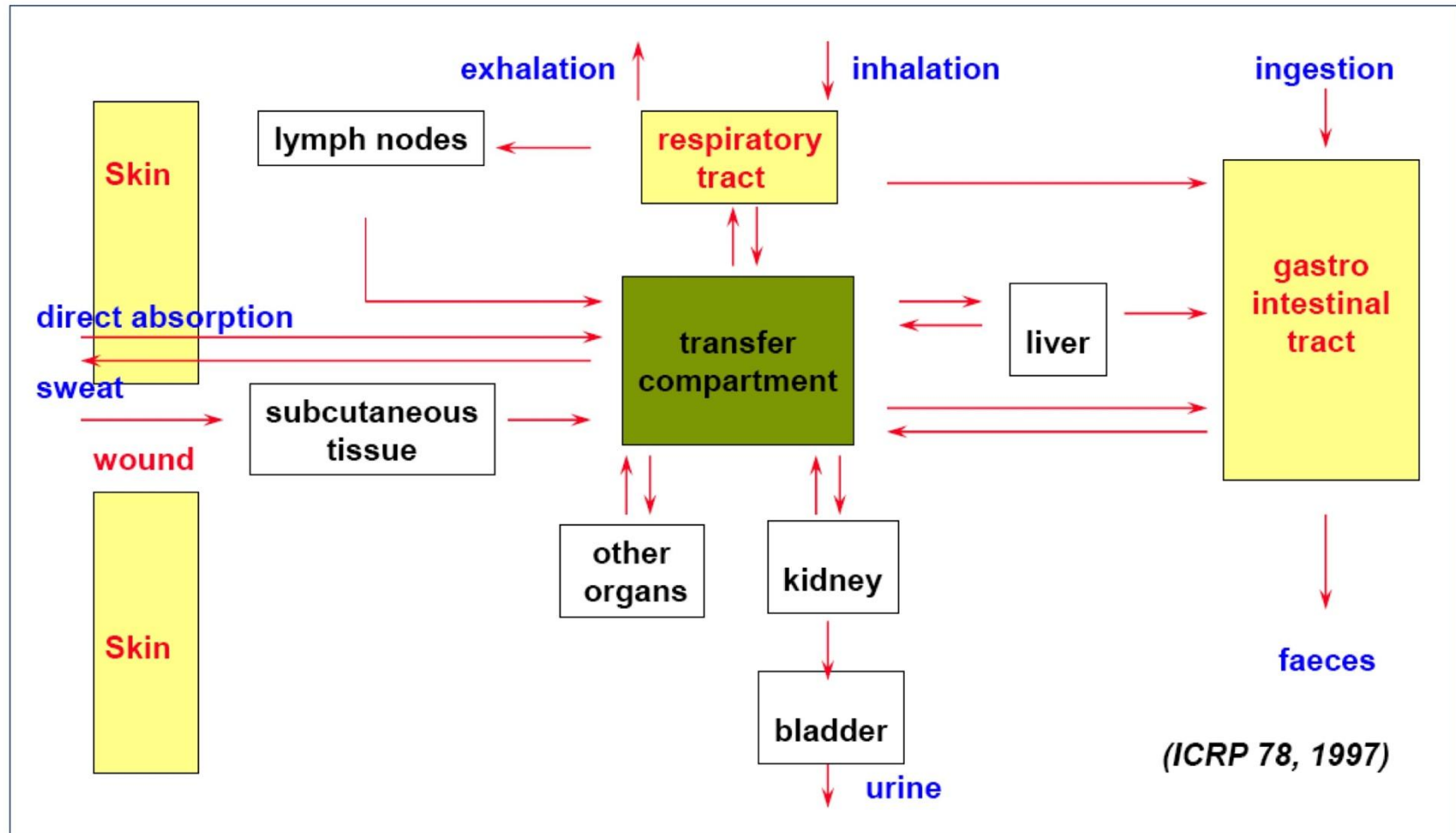
Possible Effects:

- 1) Cancer (e.g., long-term results from smoking)
- 2) Hereditary defects (e.g., Down Syndrome)

There is no threshold, and the **probability** of having effects is **proportional** to the dose adsorbed at the **population level**. In other words, depending on the conditions of exposure, effects may or may not occur.

How do you determine the **Effective Dose** to a human?

Generic Biokinetic Model



How do you determine the **Effective Dose** to humans?

$$E = \sum_T W_T H_T = \sum_T W_T \sum_R W_R D_{T,R}$$

Equivalent dose

Mean absorbed dose imparted to tissue (Gy)

$$E = \sum_T W_T H_T = \sum_T W_T \sum_R W_R D_{T,R} \quad (\text{Sv})$$

Effective dose

Tissue radiosensitivity

Radiation quality

ICRP, 2007

Tissue Weighting Factor: (W_T)

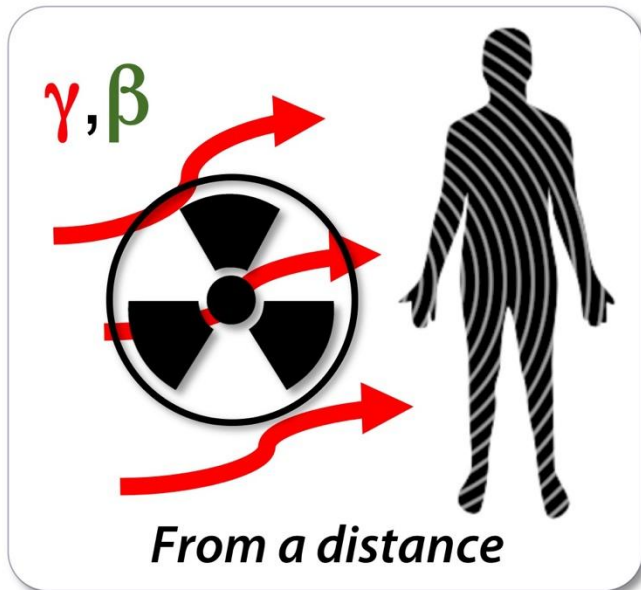
Tissue	W_T	$\sum W_T$
Bone-marrow (red), colon, lung, stomach, breast, remainder tissues	0.12	0.72
Gonads	0.08	0.08
Bladder, oesophagus, liver, thyroid	0.04	0.16
Bone surface, brain, salivary glands, skin	0.01	0.04

Reminder: $w_R = 1$ (β, γ, X) and 20 (α)

Note $\sum W_T$ across whole body = 1

ICRP, 2007

Irradiation versus contamination



Irradiation

Body is exposed to external radiation (i.e., via water, soil/sediment and air)



Contamination

The radioactive substance is directly on the skin (external) or within the body (internal)

Let us examine one pathway as an example and major concern for Marine Systems: **Seafood ingestion**

$$D_{eff-ing} = \sum_i \sum_j A_{i,j} Q_j DC_i$$

Where:

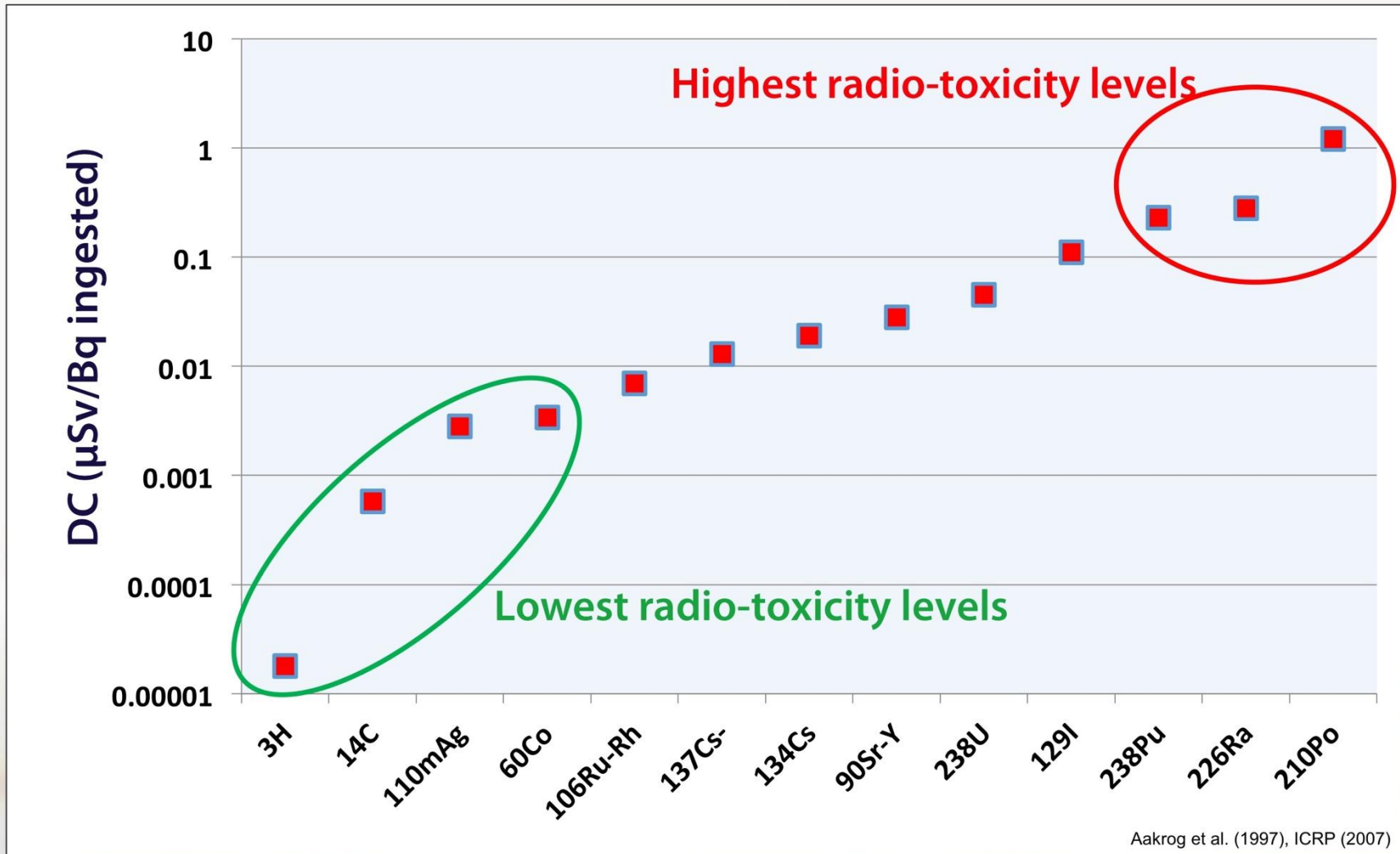
$D_{eff-ing}$	Effective dose by ingestion	Sv y^{-1}
$A_{i,j}$	Radionuclide i massic activity in foodstuff j	Bq kg^{-1}
Q_j	Consumption rate of foodstuff j	kg y^{-1}
DC_i	Dose coefficient for radionuclide i	Sv Bq^{-1} ingested

Specific Cases:

The potassium concentration is kept constant by humans. The proportion of ^{40}K to total K (specific activity: Bq kg^{-1} of potassium) is also constant. So the ^{40}K whole body activity is constant and leads to an effective dose of $\sim 170 \mu\text{Sv y}^{-1}$ for an adult ($185 \mu\text{Sv y}^{-1}$ for a child).

For similar reasons, ^{14}C activity is constant in the human body. This leads to an annual effective dose of $\sim 12 \mu\text{Sv/y}$.

Dose coefficients (DC) for some naturally occurring and artificial radionuclides (ingestion pathway)



After ingestion, how long do these radionuclides exist in the body (Effective $t_{1/2}$)

The **effective half life ($t_{1/2}$)** represents the combination of the physical and biological half-lives.

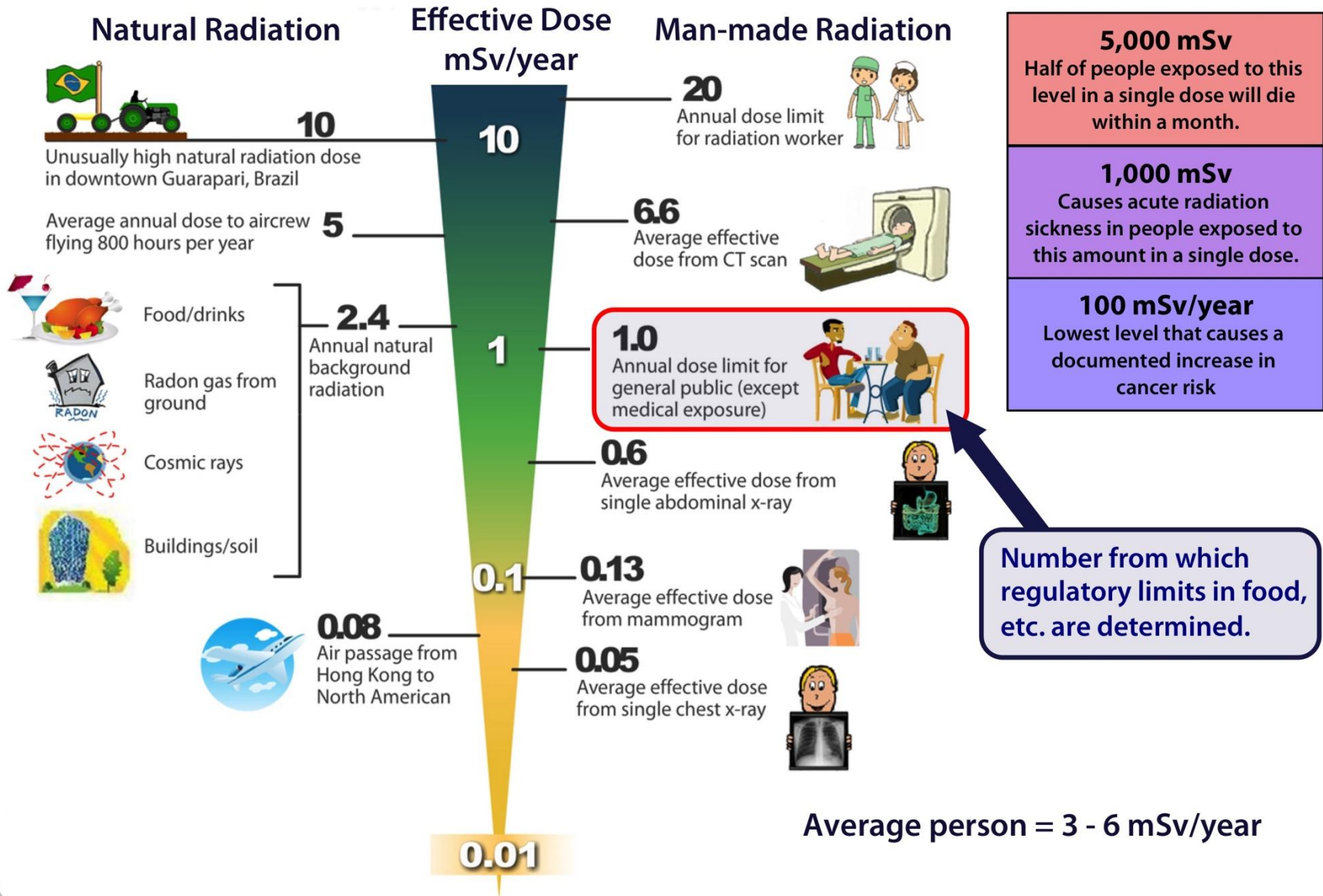
Radionuclide	$t_{1/2}$ (phys)	Part of the body considered	$t_{1/2}$ (eff)
^3H	12.32 y	Body Tissue	12 d
^{14}C	5730 y	Fat	12 d
^{32}P	14.3 d	Gastrointestinal Tract	14 d
^{35}S	87.5 d	Testis	76 d
^{60}Co	5.27 y	Gastrointestinal Tract	0.75 d
^{129}I	1.57×10^3 y	Thyroid	140 d
^{131}I	8.02 d	Thyroid	7.6 d
^{137}Cs	30.17 y	Total	70 d
^{210}Po	138.4 d	Spleen	42 d
^{222}Rn	3.8 d	Lung	3.8 d
^{226}Ra	1600 y	Bone	44 y
^{238}U	4.47×10^9 y	Lung, Kidney	15 d
^{238}Pu	2.41×10^4 y	Bone	64 y
^{241}Am	432.2 y	Kidney	64 y

Radiotoxicity	Radionuclides and radioelements
Group I: Very High	^{90}Sr , ^{226}Ra , ^{238}Pu
Group II: High	^{55}Fe , ^{210}Bi , ^{210}Po
Group III: Medium	^3H , ^{14}C , ^{32}P , ^{35}S , ^{137}Cs
Group IV: Low	^{42}K , ^{64}Cu , ^{85}Kr

Depends on: Radiation emitted, mode of intake, amount, chemical properties/metabolic affinity, and the **effective $t_{1/2}$**

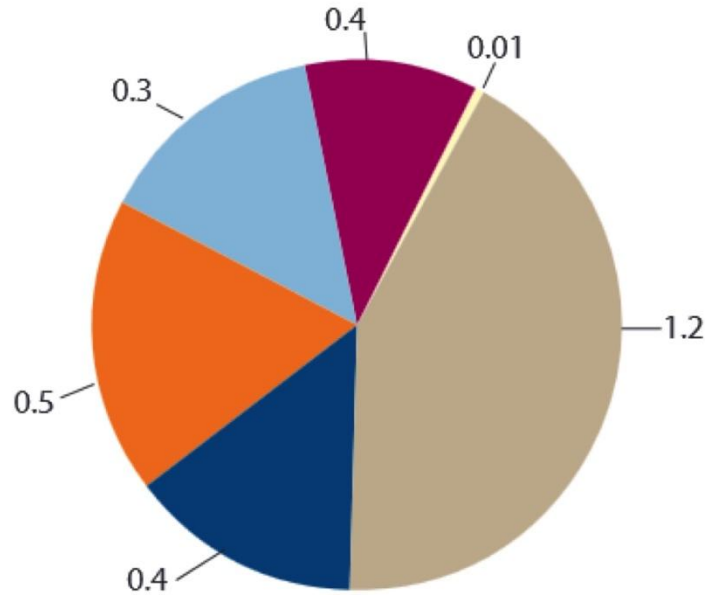
Selected data from Leiser (2008) as adapted from ICRP (1993)

Radiation in daily life



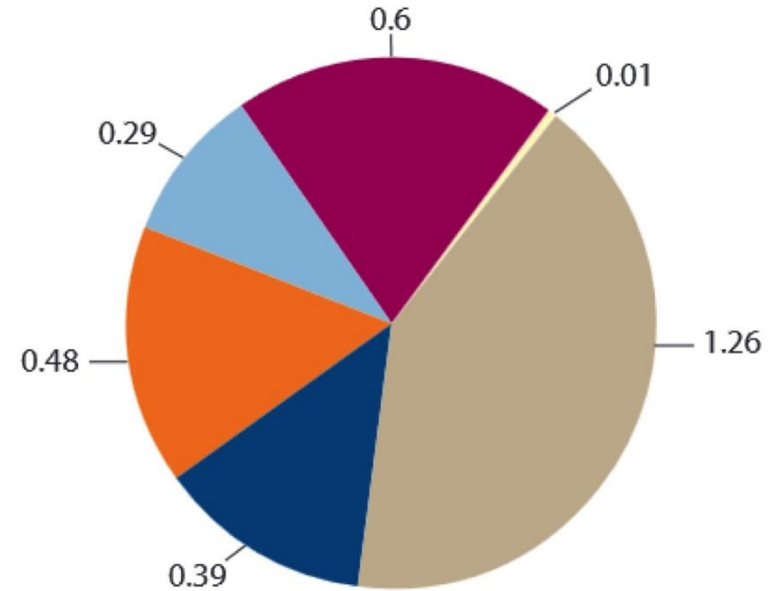
Worldwide average radiation exposure from various sources

GLOBAL (UNSCEAR 2000)



2000: Global average dose
 $\approx 2.81 \text{ mSv y}^{-1}$

GLOBAL (UNSCEAR 2008)



2008: Global average
 $\approx 3.03 \text{ mSv y}^{-1}$



Exposure to ionizing radiation varies among countries and within countries.
 It greatly depends on geographic location and way of life.

Environmental Protection

Ecological risk assessment and management



4. Radiological protection and the environment: Changing perspectives from **anthropocentric** to **ecocentric**

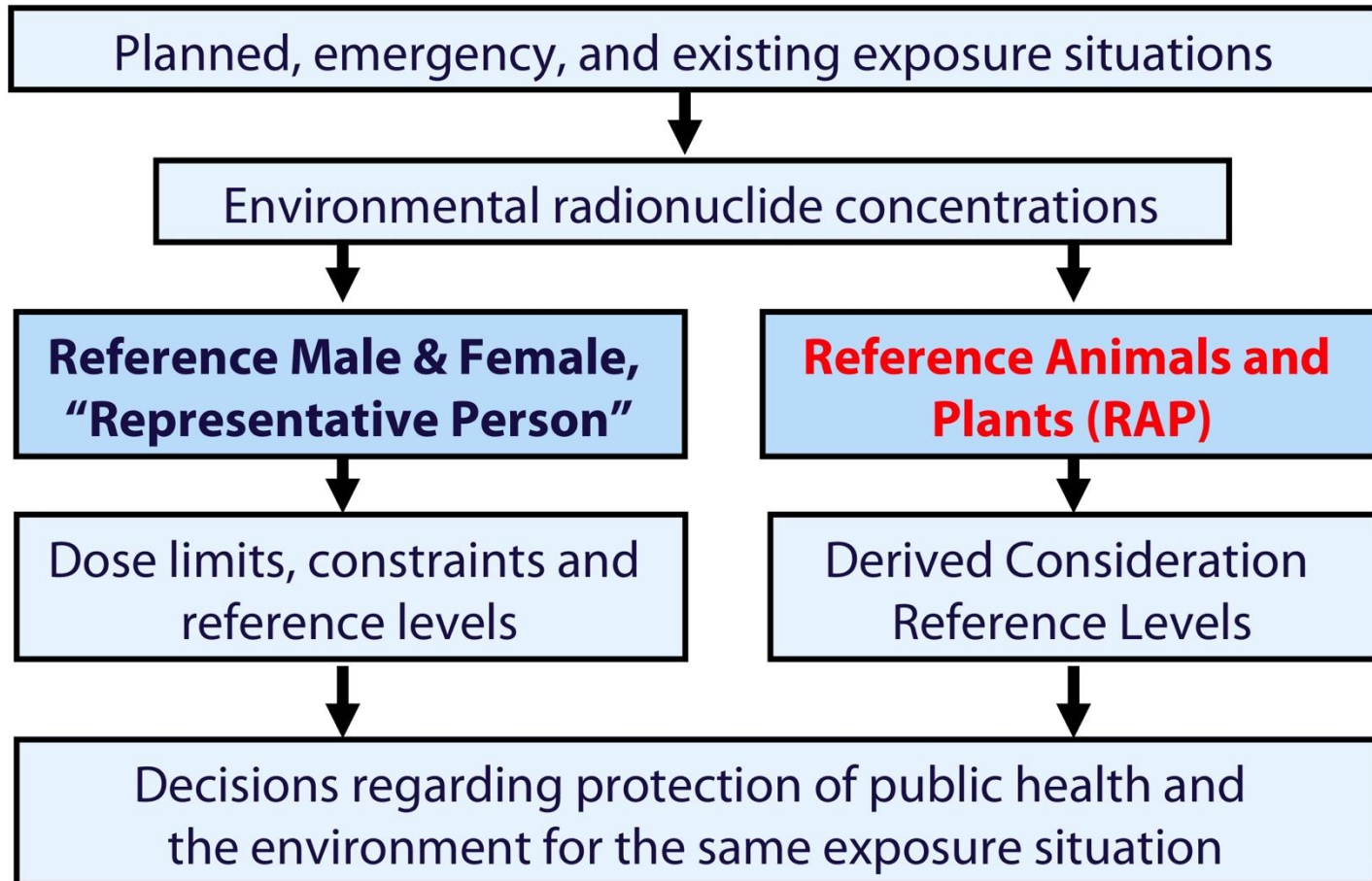
Initial focus was on humankind: It was considered that “if man is adequately protected, *then other living things* are also likely to be sufficiently protected.” (ICRP, 1977)

Beginning in the late 1980s this assumption has been questioned. Due to increasing regulations regarding the protection of the environment, specific criteria for species other than man has reached a consensus by the International Community.

ICRP (2008)

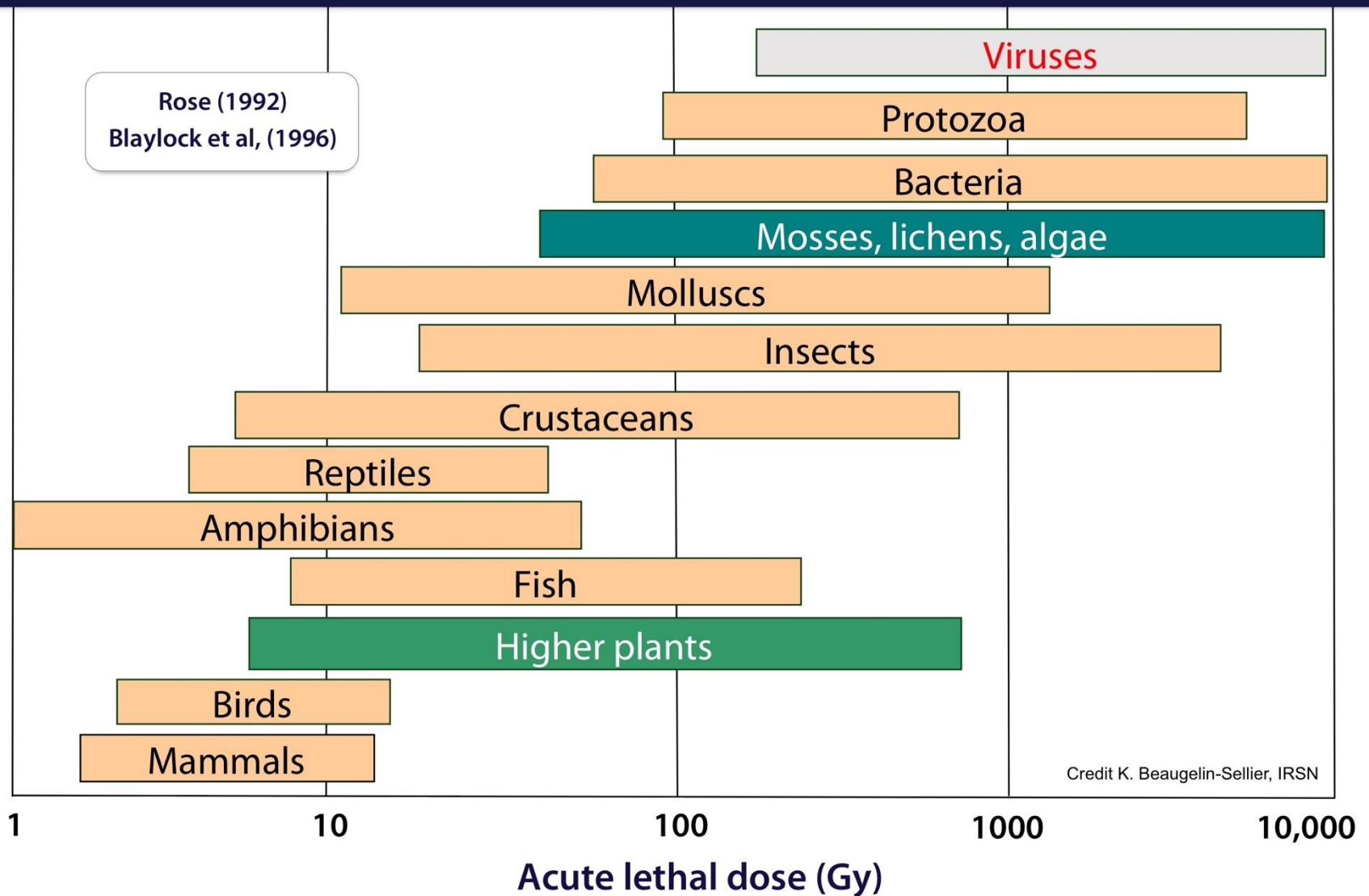
Similarity in approaches: **Humans versus the Environment**

Schematic approach to the protection of both the public and the environment in relation to any exposure situation.



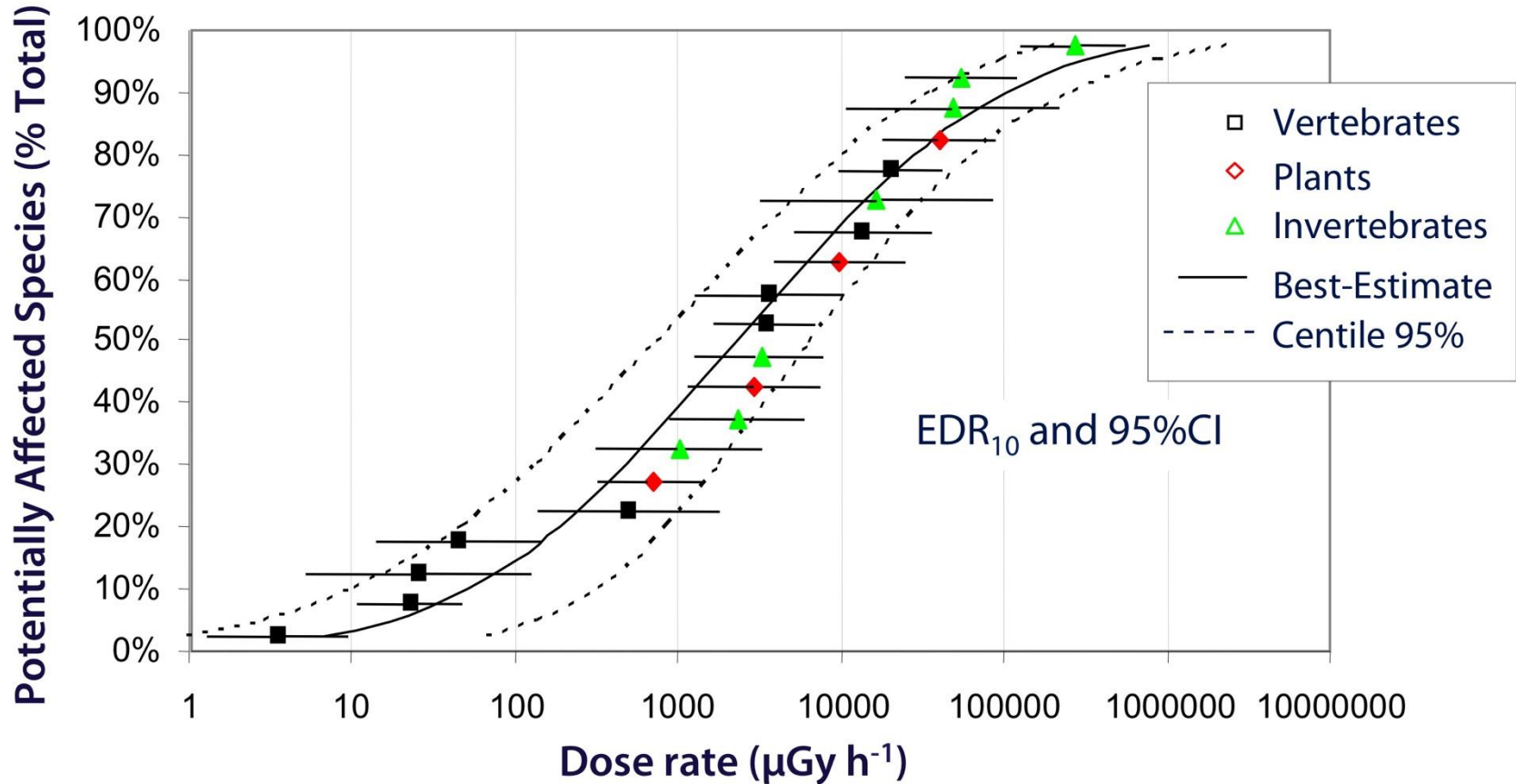
ICRP (2008, 2009); Pentreath (2009)

What we know regarding the effects on organisms: Acute ionizing radiation exposure



Chronic Exposure

Species Sensitivity Distribution for a **generic ecosystem** and assuming chronic gamma (γ) ray exposure



Garnier-Laplace et al. (2006, 2010)

Radiological protection and the environment

- UNSCEAR (2008): Analyses of available data concludes **that chronic irradiation at dose rates up to 400 $\mu\text{Gy h}^{-1}$ to a small proportion of individuals in an aquatic population results in no detrimental effects at the population level**
- ICRP (2008): Defines DCRLs (i.e., a range of dose rates within which it is likely to observe **some deleterious effects**) for a given **Reference Animal or Plant (RAP)**.

Marine RAPs : Flatfish and seaweed: 42-420 $\mu\text{Gy h}^{-1}$
Crabs: 420-4200 $\mu\text{Gy h}^{-1}$

Various European Union programs have agreed on a **conservative Predicted No Effect Dose-Rate** of **10 $\mu\text{Gy h}^{-1}$** , which is ultimately derived from the Species Sensitivity Distributions (see previous slide).

The ERICA tool

The 1st European answer towards demonstration of environmental protection

Environmental Risk *from* Ionising Contaminants: Assessment *and* Management

- The outcome of a suite of European research programs
- 2004-2007 (first release)

ERICA: Suggested screening benchmark: **10 $\mu\text{Gy h}^{-1}$ (87.6 mGy y^{-1})** to protect the structure and function of a generic ecosystem

Development of the ERICA tool:

- A *free software* approach to assessing the radiological risk to terrestrial, freshwater and marine *biota*
- Based on a reference organism approach based *on biological effects on individual marine biota*
- Updated in 2008, 2009, 2011, 2012, 2014 and 2016
- Website: <https://wiki.ceh.ac.uk/display/rpemain/ERICA+Tool>

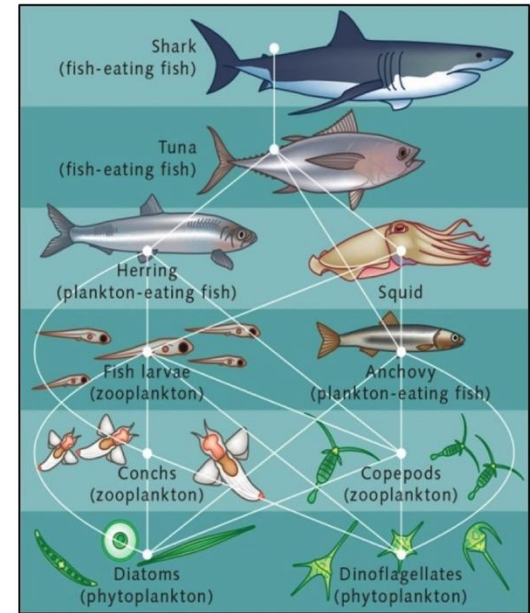
Summary

Radionuclides from a variety of sources are bioaccumulated by marine organisms

Bioaccumulation depends on:

- The marine organism (Species, trophic level...)
- **The radionuclide (physical-chemical forms ...)**
- **Environmental parameters (temperature, salinity...)**

Marine biota transport radionuclides horizontally (migration) and vertically (migration, feces). Some radionuclides may accumulate in higher trophic levels via grazing.



Future Directions:

- There is very little information on radionuclide uptake by plankton communities or in top predators, including mammals.
- Most of the data is derived from temperate climates. Larger geographical studies are needed.
- Comprehensive studies on radionuclide transfer in a food web are uncommon.

Summary

Radionuclides from a variety of sources are bioaccumulated by marine organisms

Doses to humans and biota *mainly* arise from natural sources.

Since 2000, the emphasis has been on the development of specific criteria for environmental protection.

Future Directions:

- More data is needed regarding the radiological chronic exposure effects on marine biota
- Refinement of dose-response relationships under natural versus laboratory conditions
- Environmental Risk Assessment needs to move from individual species to ecosystem (e.g., the impact on biodiversity index, trophic network structure, etc.)

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This work would not have been possible without the generous contributions and thoughtful comments of Drs. Robert Anderson, Kirk Cochran, Peter Santschi and Alan Shiller. We wish to thank two anonymous reviewers who provided constructive comments that improved the presentation. Lectures would not have been possible without the outstanding assistance of graphic designer Jason Emmett.

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**Claudia Benitez-Nelson, Sabine Charmasson, Ken Buesseler,
Minhan Dai, Michio Aoyama, Núria Casacuberta,
José Marcus Godoy, Andy Johnson, Vladimir Maderich,
Pere Masqué, Marc Metian, Willard Moore,
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