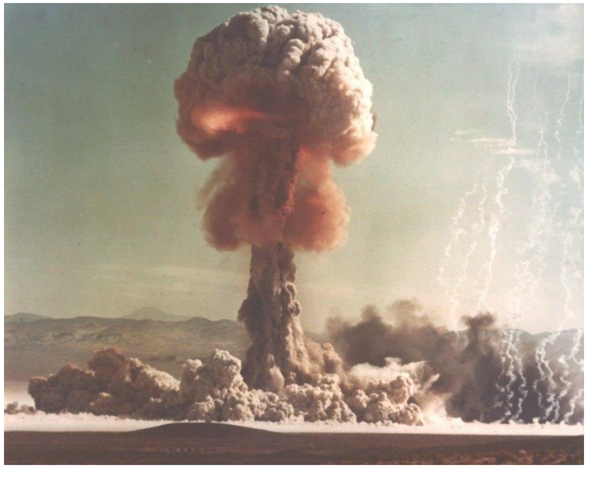
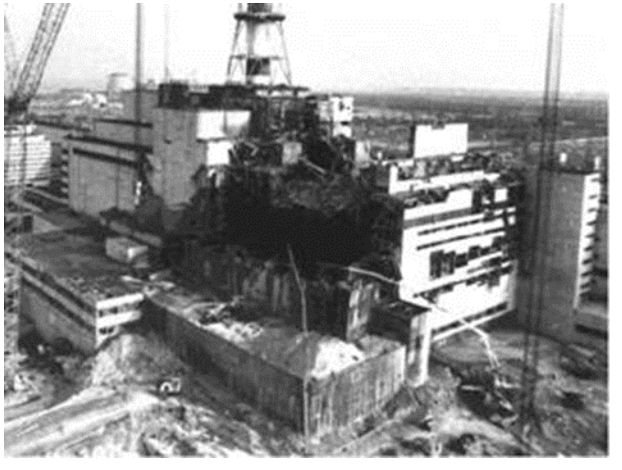
Fate of ^{137}Cs and other radionuclides in rivers and lakes

Prof. Jim Smith

University of Portsmouth

…. and many collaborators !





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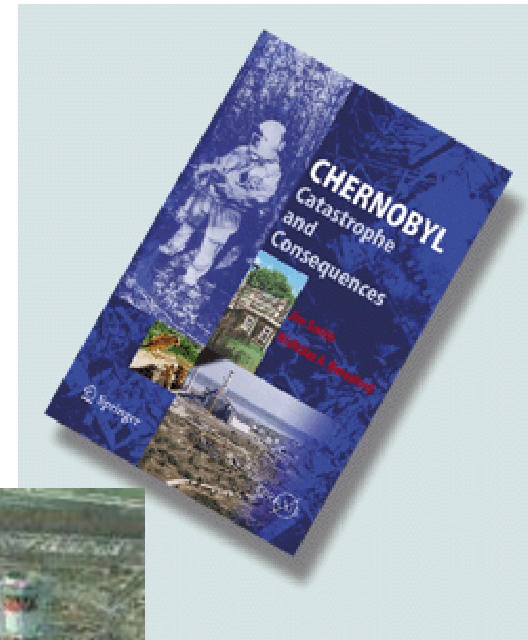
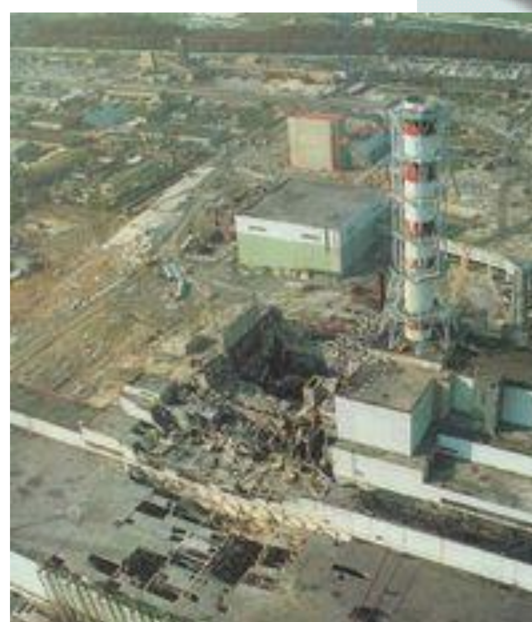
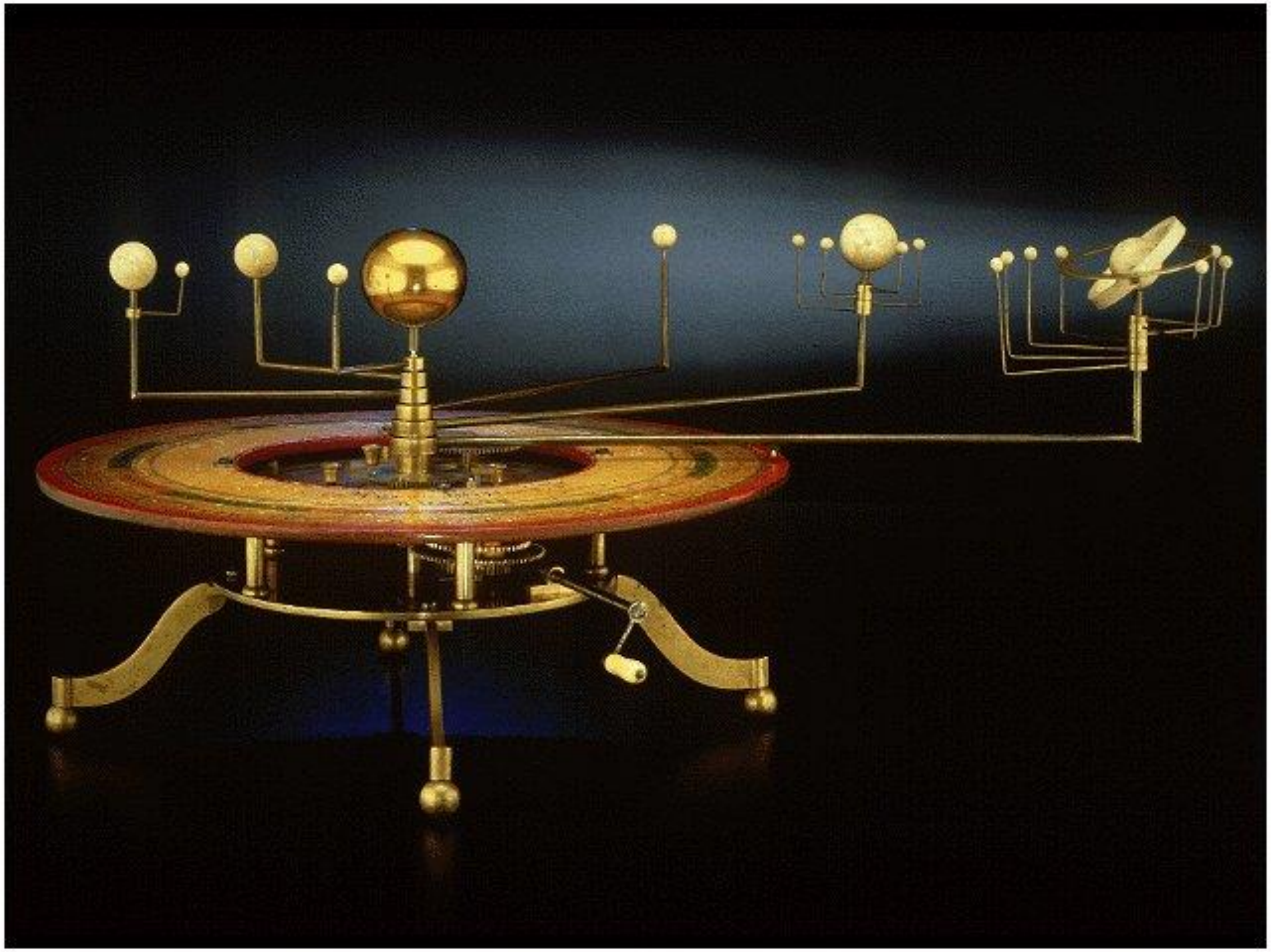
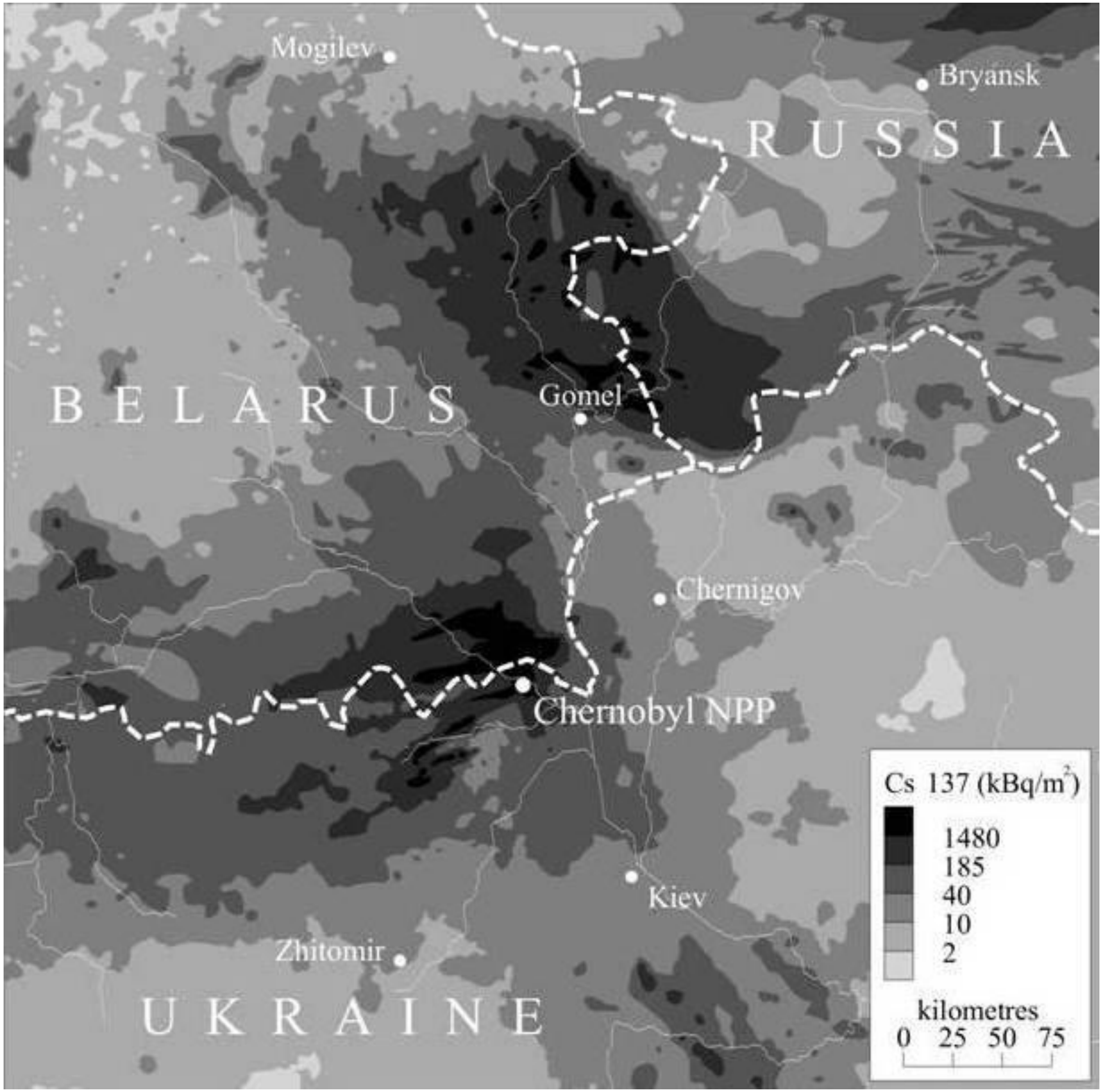
From Astrophysics to Ecology

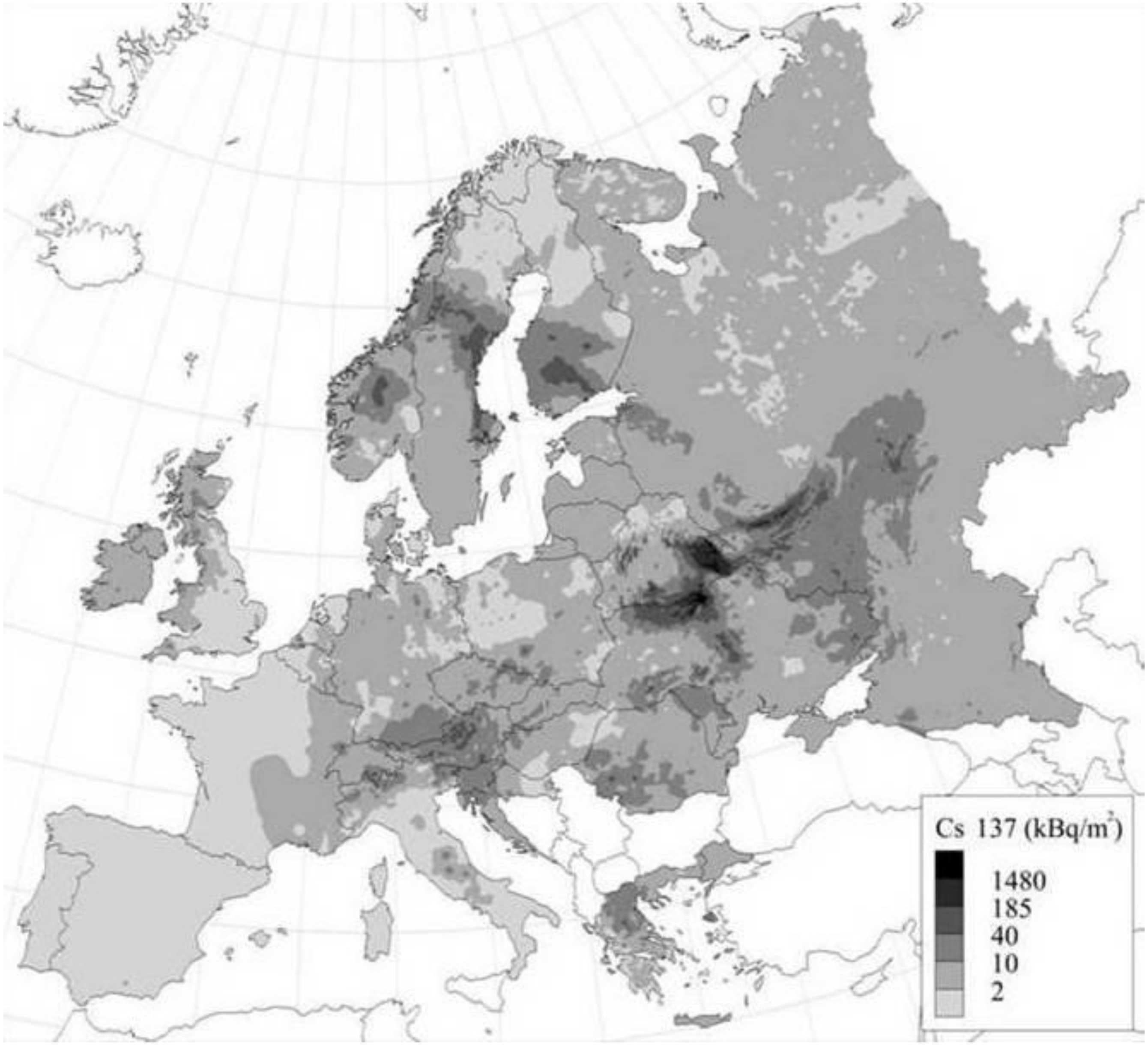






Mechanistic model of the Solar System

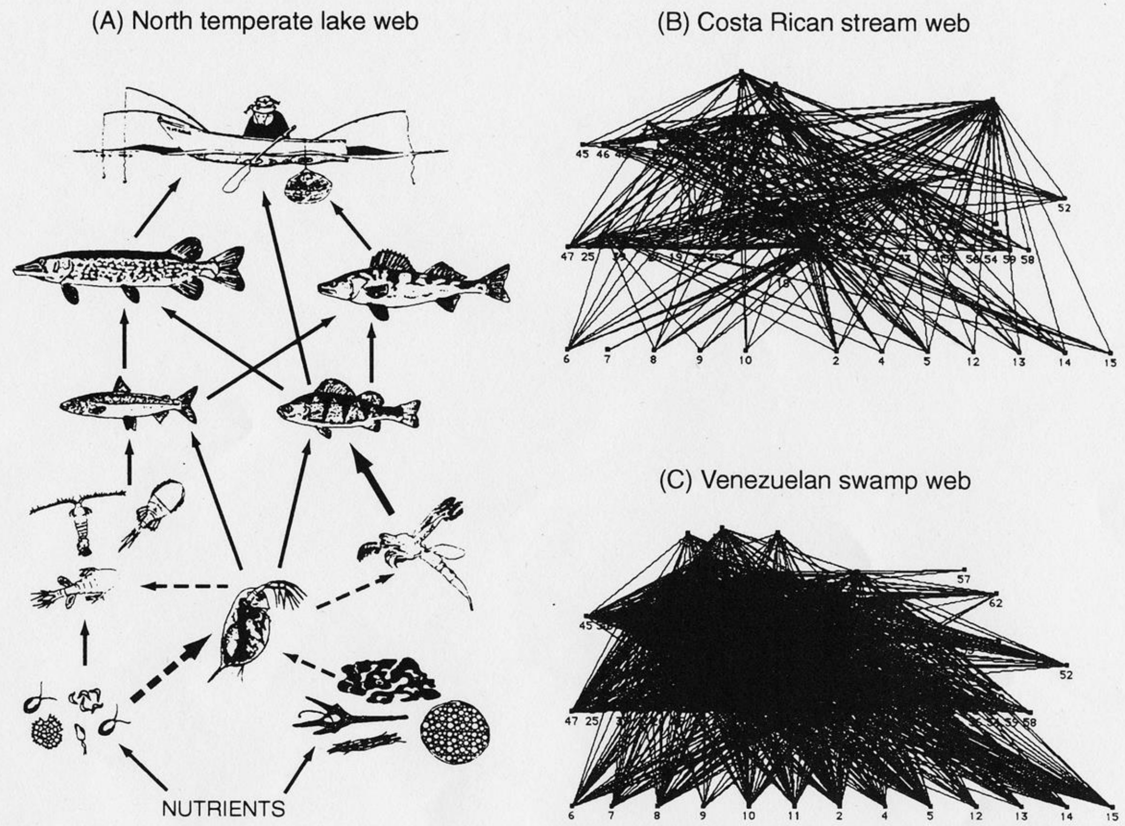

Mogilev
Bryansk
RUSSIA
BELARUS
Gomel
Chernigov
Chernobyl NPP
Kiev
Zhitomir
UKRAINE
Cs 137 (kBq/m^2)
1480
185
40
10
2
kilometres
0 25 50 75


Cs 137 (kBq/m²)
1480
185
40
10
2

Radioactive pollution of lakes

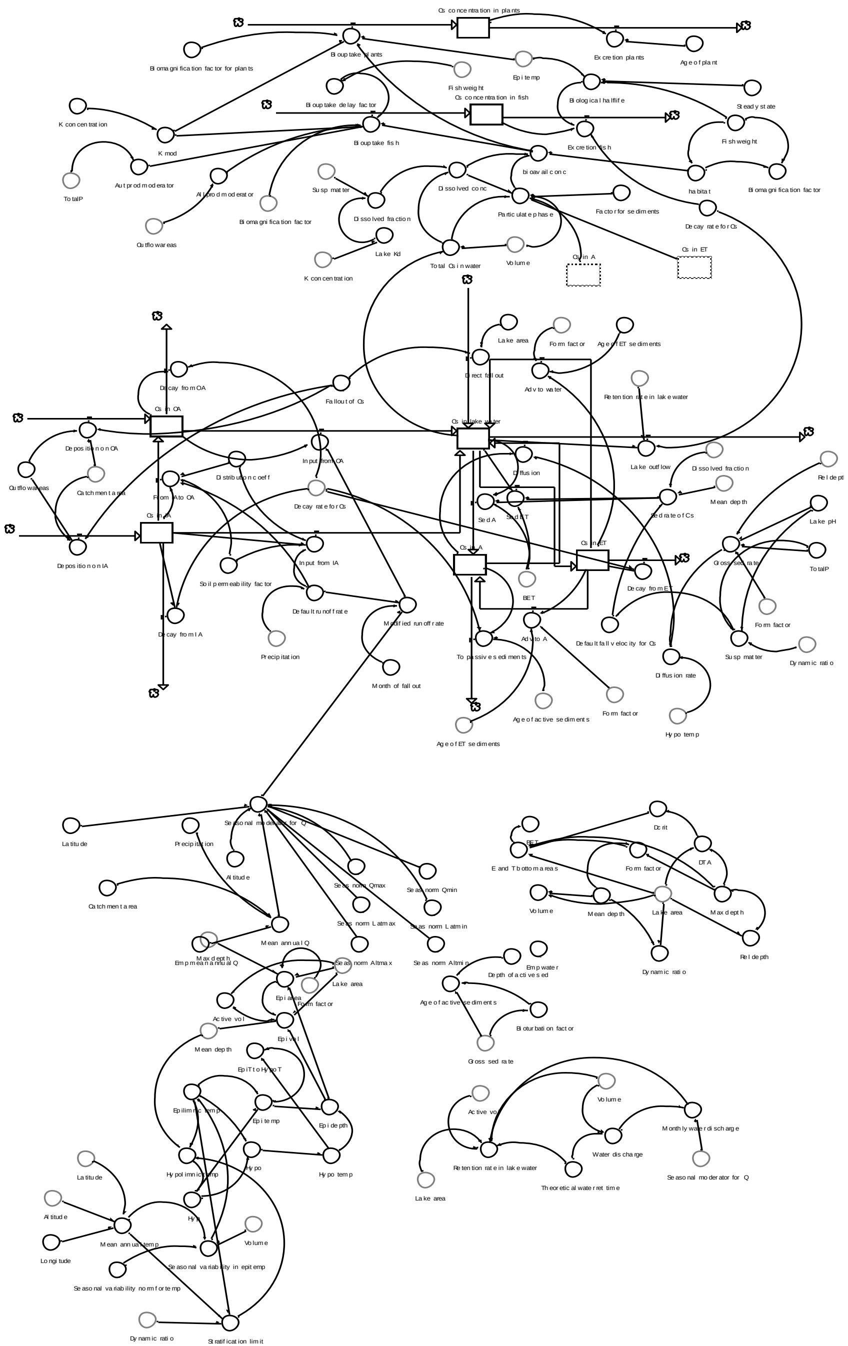
-

Aquatic food webs



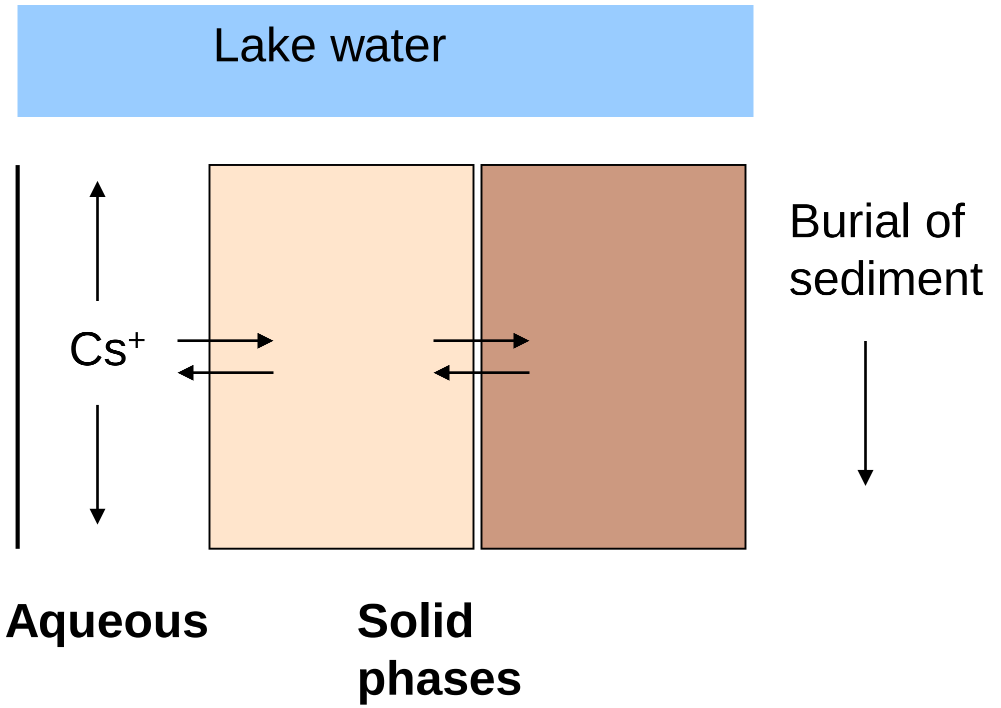
(A) After Rudstam et al. 1992. (B)–(C) From Winemiller 1990; used with permission.

From: *The Diversity of Fishes*, G.S. Helfman et al., Blackwell, Oxford 1997.



Lake model;
Lars Hakansson,
University of Uppsala

Interaction of Cs-137 with lake sediments

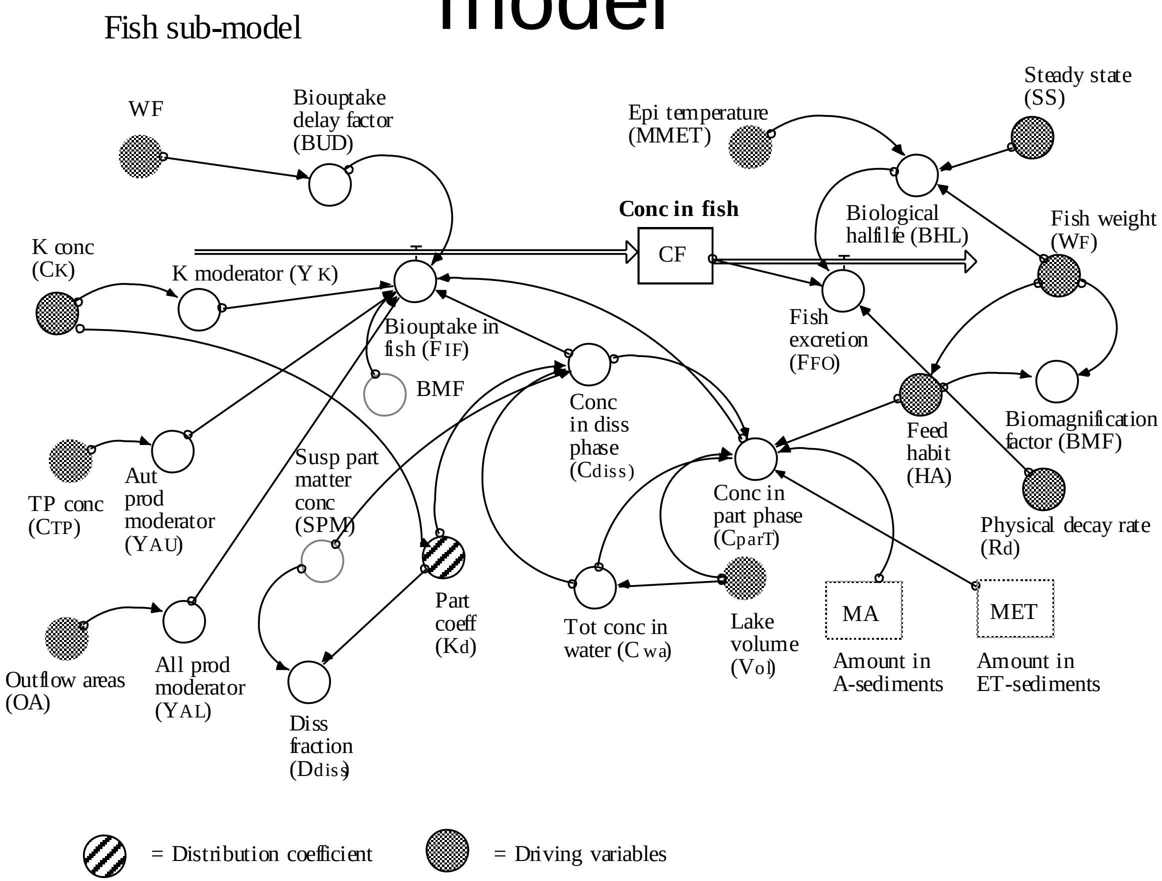


$$\frac{\partial C_e}{\partial t} = \frac{\partial}{\partial x}\left[\phi \psi D_o \frac{\partial}{\partial x}\left(\frac{C_e}{\phi + sK_d^e(x)}\right) - rC_e\right] - k_f C_e + k_b s C_i - \lambda C_e$$

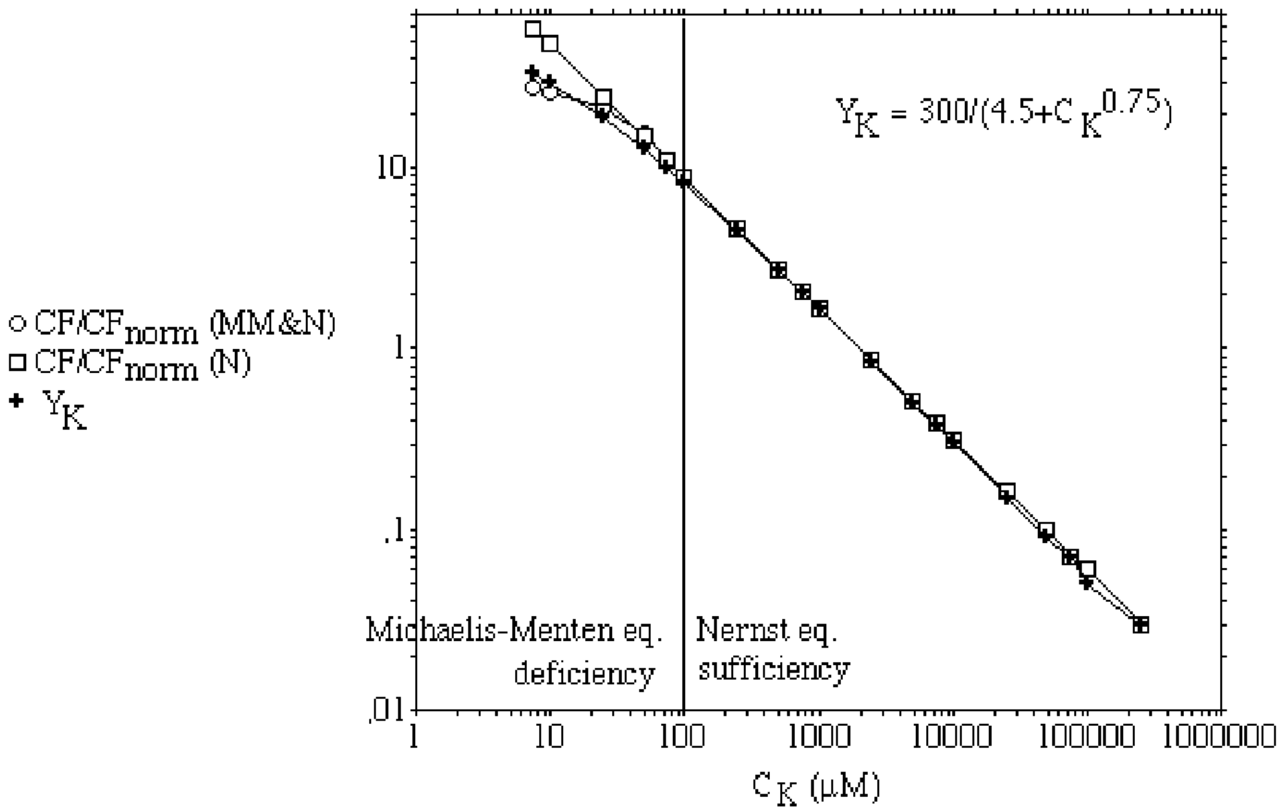
$$s\frac{\partial C_i}{\partial t} = -\frac{\partial (rsC_i)}{\partial x} - k_b s C_i + k_f C_e - \lambda s C_i$$

(Smith & Comans, *Geochim. Cosmochim. Acta*,1996)

Fish sub-model

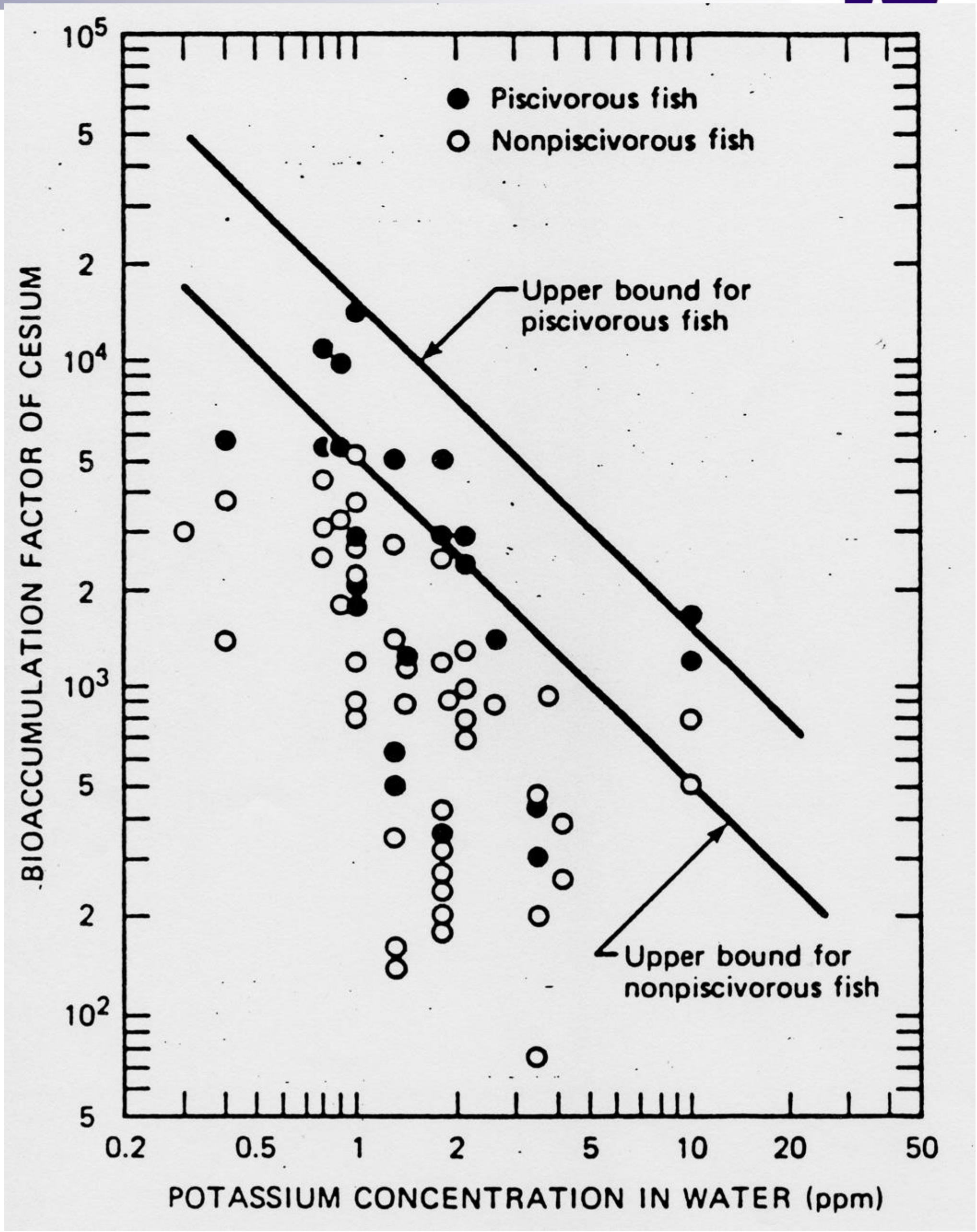


Caesium-potassium model



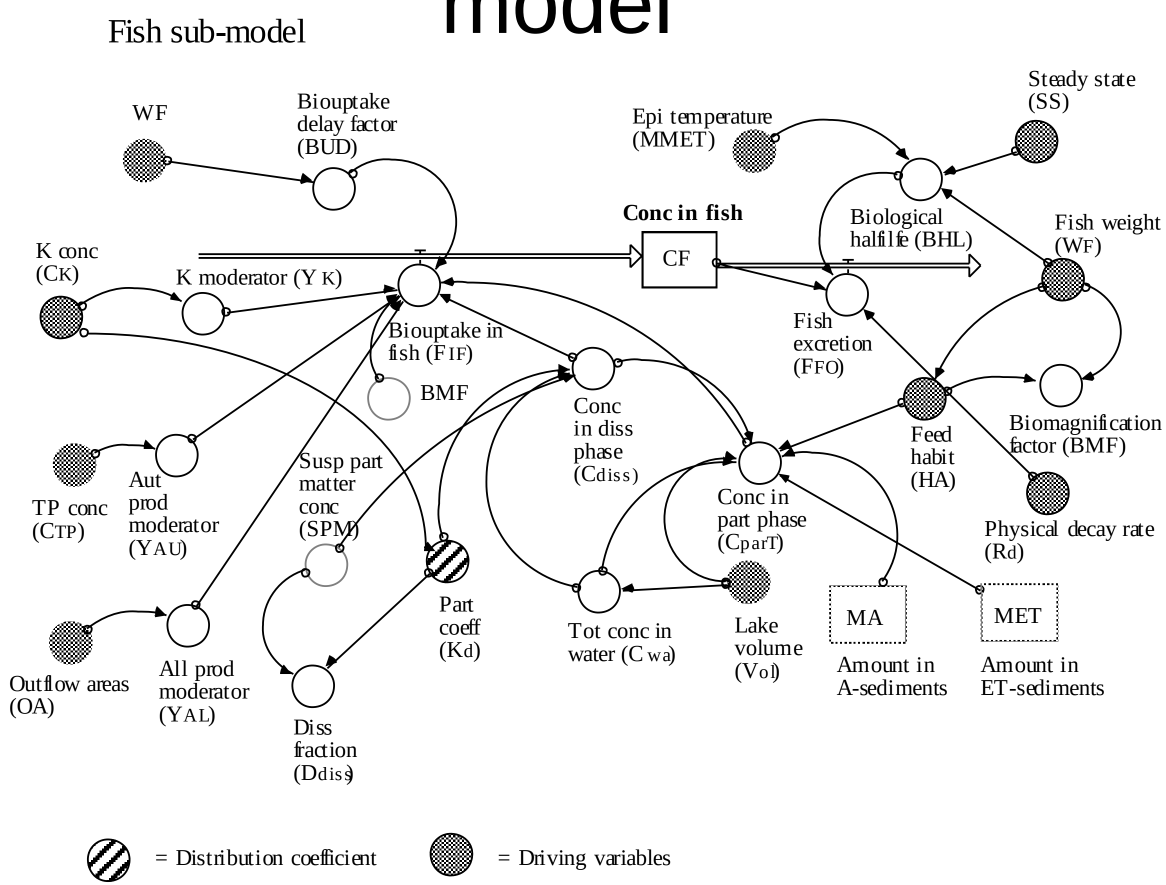
Caesium-potassium model from measurements in lakes pre-Chernobyl

Vanderploeg et al. 1975

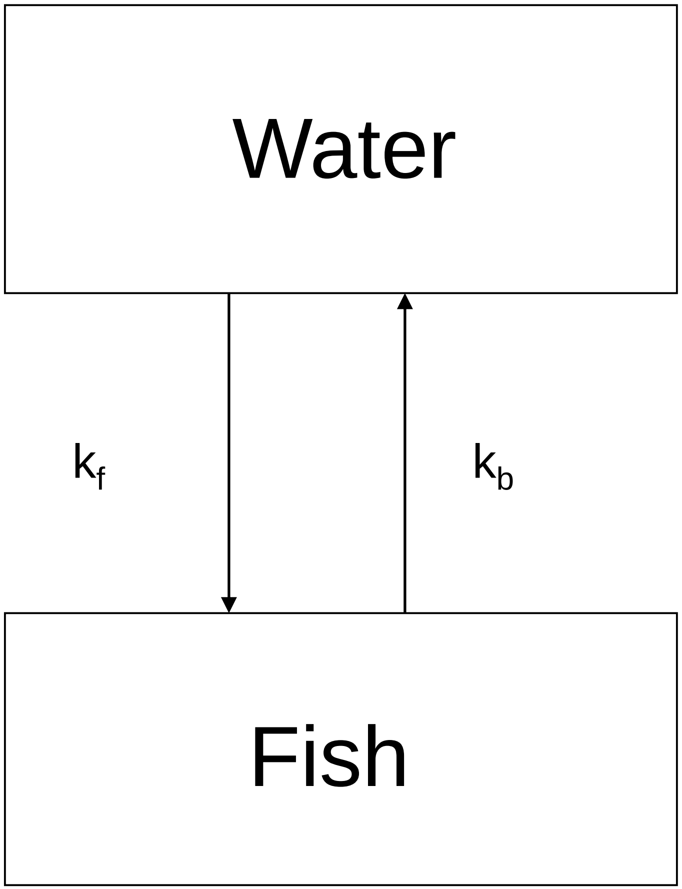


Lesson 1: The old literature and empirical data from the nuclear weapons test period are very important

Fish sub-model



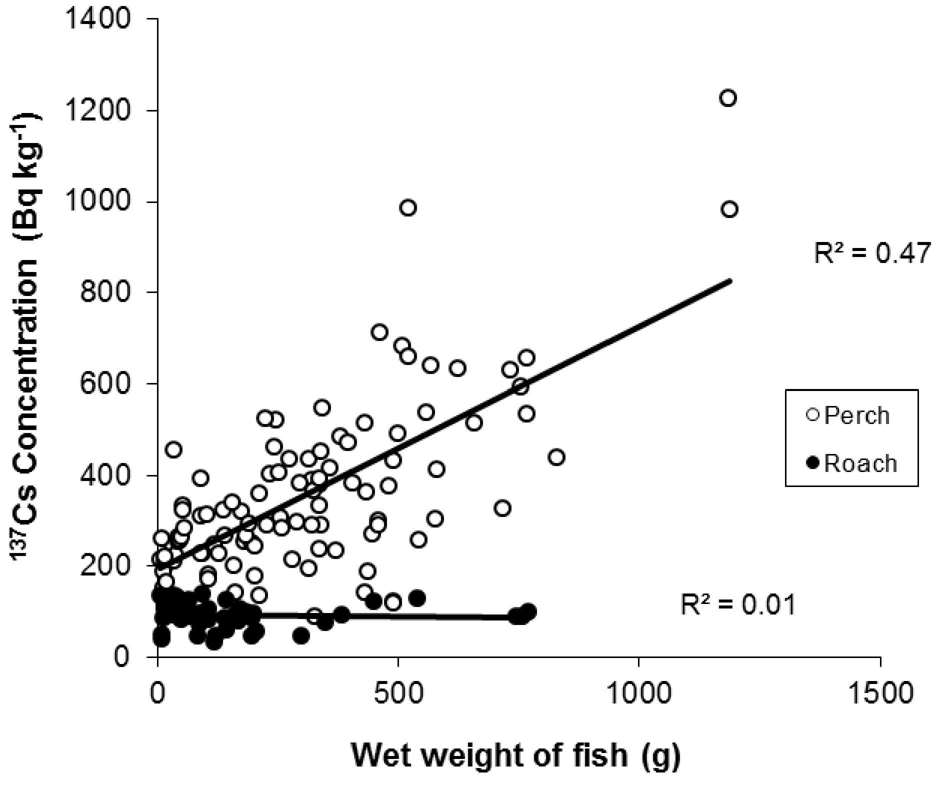
New Model:



Lesson 2: Simple models often make better predictions than complex ones.

Smith, J.T. “Nice work, but is it Science ?” *Nature*, 408 (2000); p. 293.

Models should be as simple as possible…. …. But no simpler ! (Albert Einstein)



Size/trophic level/age effect

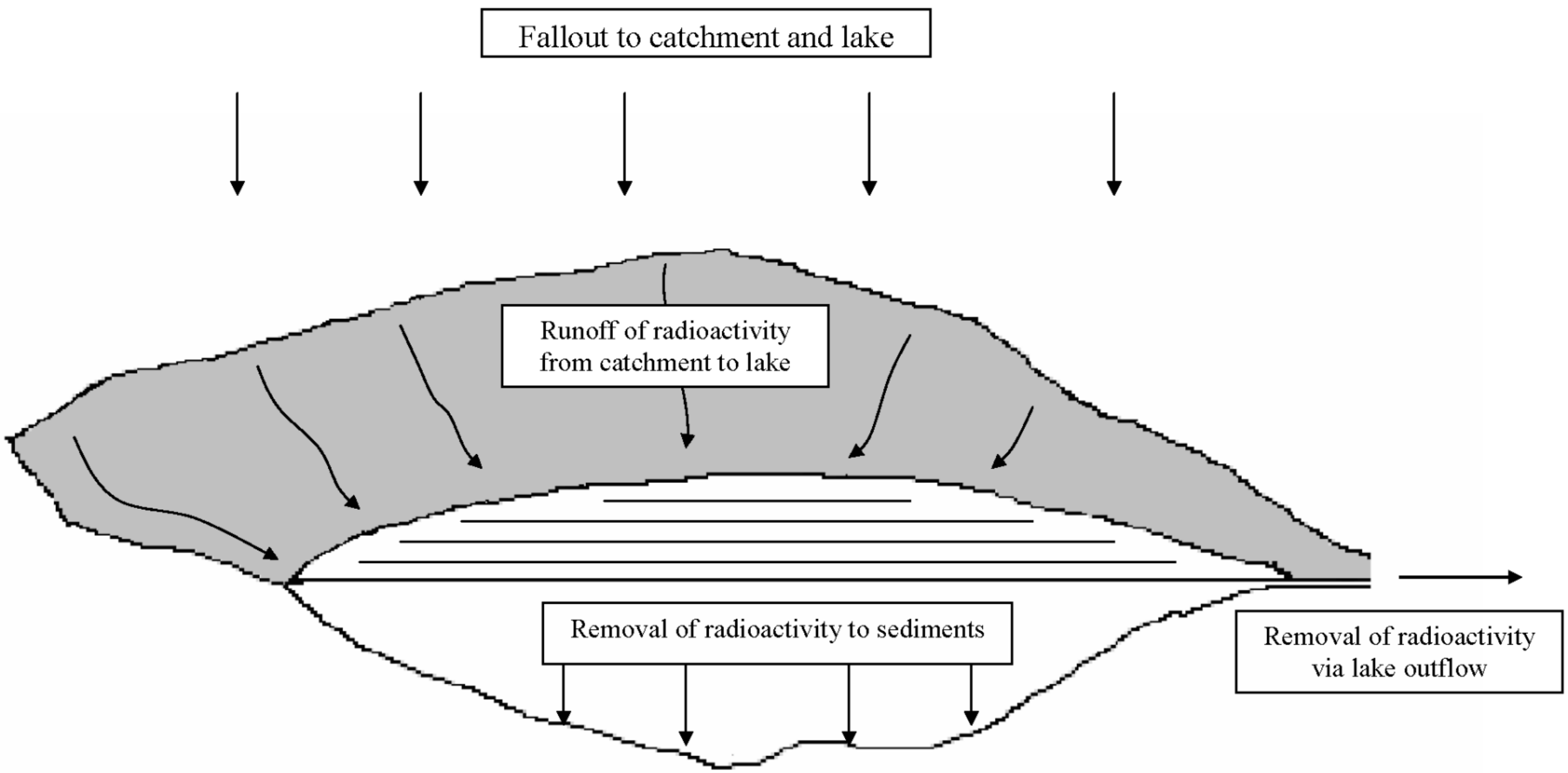
Radiocaesium in fish in the Kiev Reservoir (Sansone and Voitsekhovitch, 1996)

Developing a model for European lakes and rivers

- There are more than 500,000 lakes in Europe
- Even more rivers and streams…

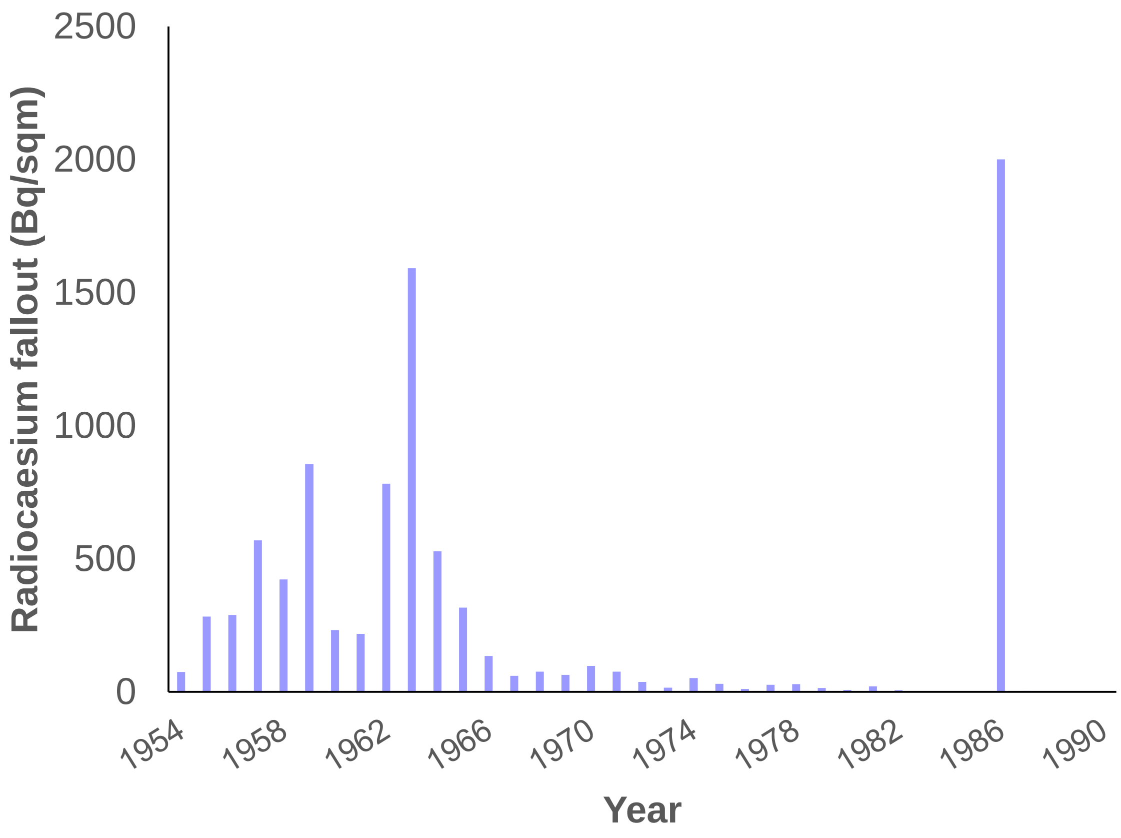
Lesson 3: Need to find general parameters which we can use to make **predictions**

…. AQUASCOPE model

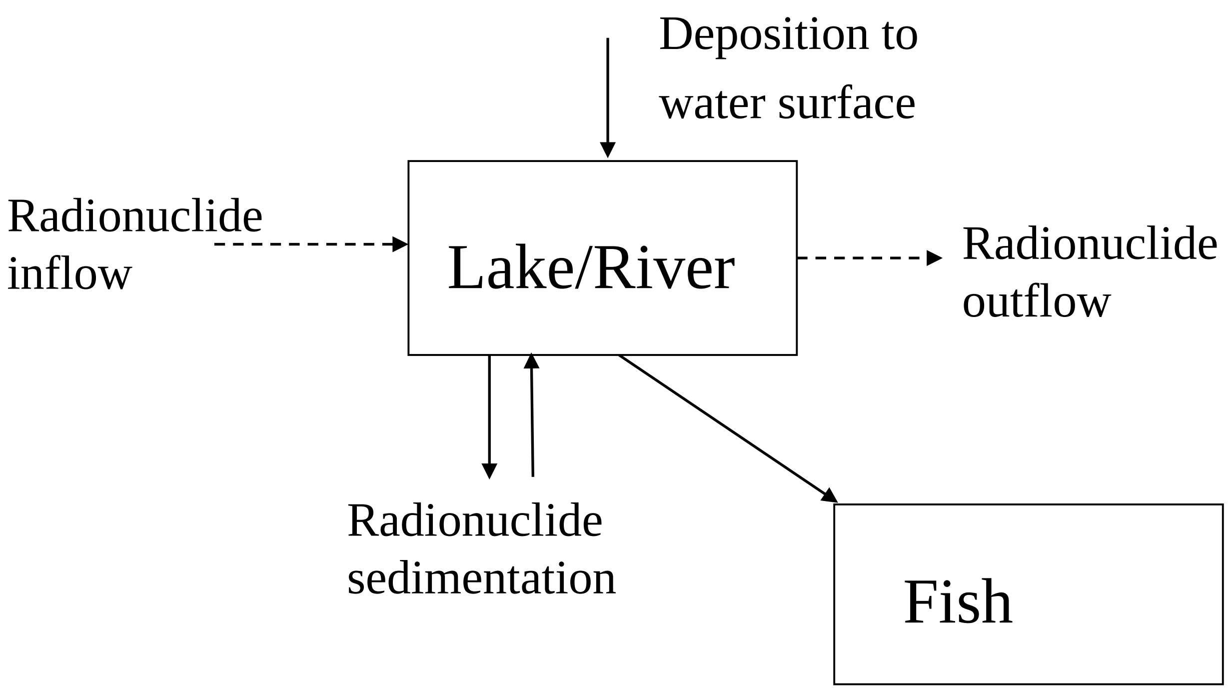


Fallout to catchment and lake
Runoff of radioactivity from catchment to lake
Removal of radioactivity to sediments
Removal of radioactivity via lake outflow

Analysis of data from nuclear weapons testing and Chernobyl

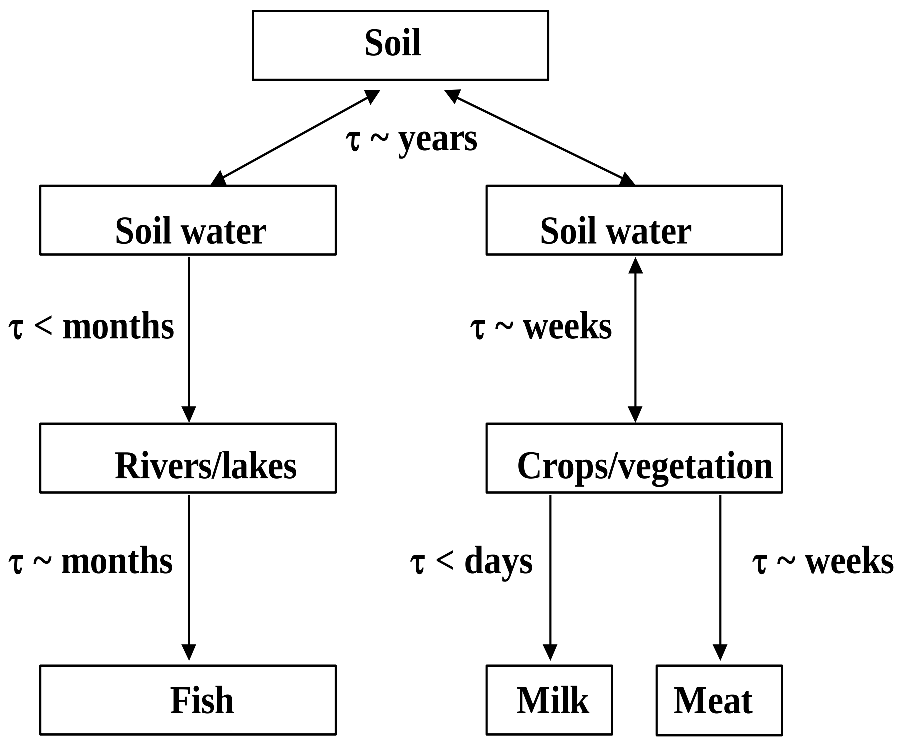


AQUASCOPE Model

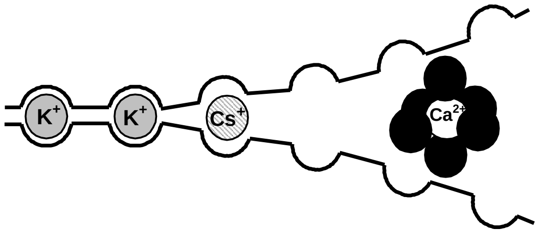


"Blind" predictions of radionuclides in rivers and lakes:

Conceptual model of time changes in radiocaesium



Centre for Ecology & Hydrology



Sorption of ^{137}Cs to soils/sediments

- After a radioactive fallout, sorption of ^{137}Cs to soils increases over time, a process known as "fixation":

Aqueous $\rightleftarrows$ Exchangeable $\rightleftarrows$ "Fixed"

Rapid kinetics

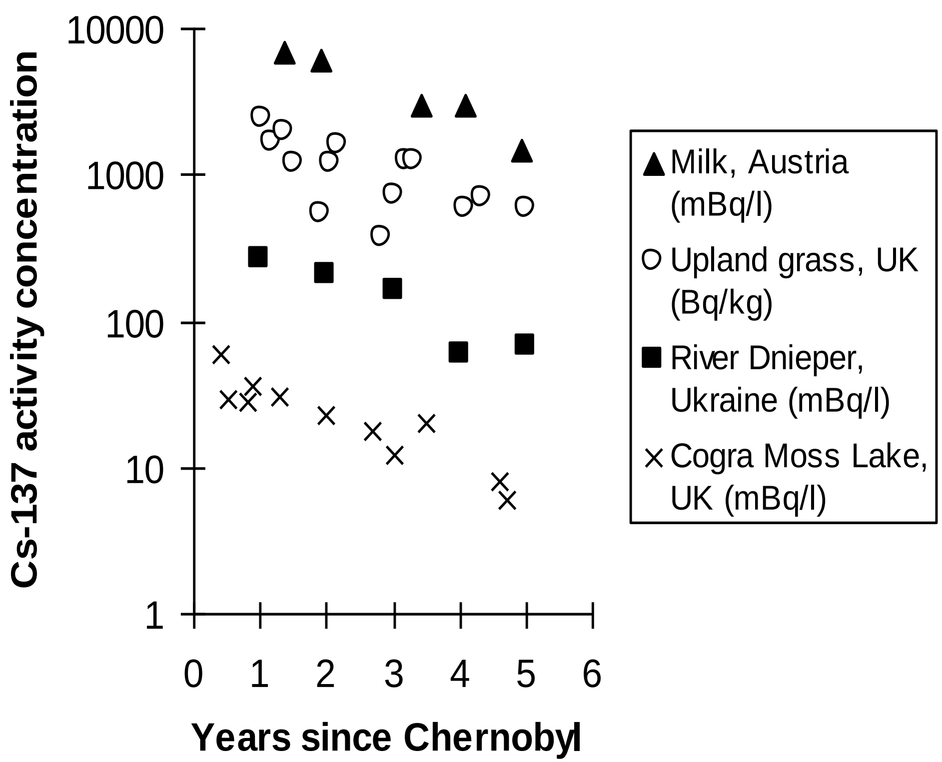
Slow forward and reverse reactions.

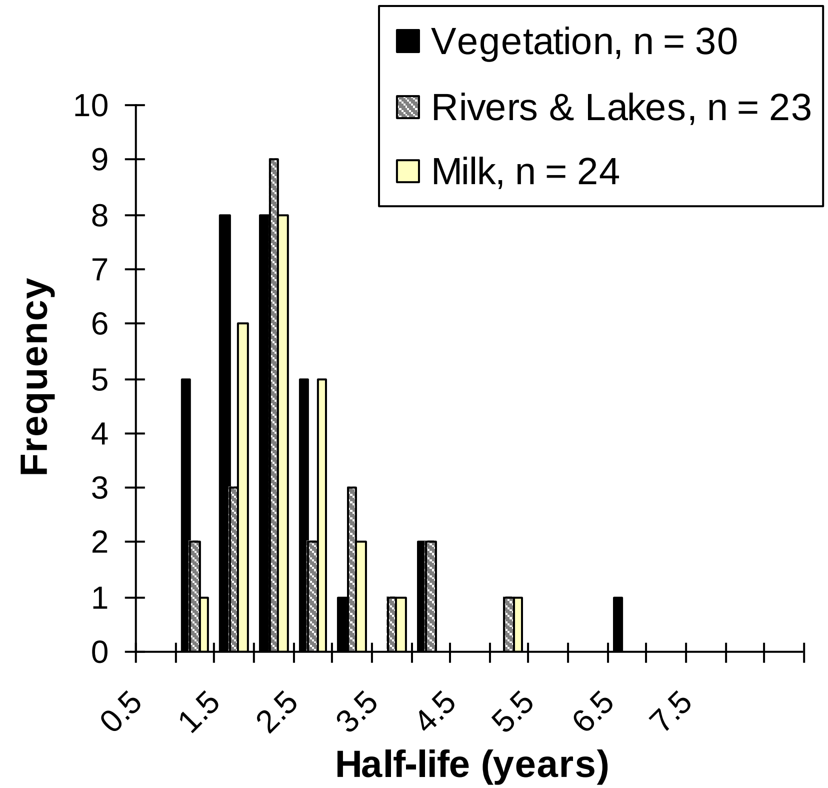
- Reduction of ^{137}Cs concentration in soil water reduces transfers to rivers and lakes and uptake in vegetation.

Cremers et al (1988); Comans et al. (1989); Smith & Comans (1996)

"Fixation" of radiocaesium to soil

University of Portsmouth



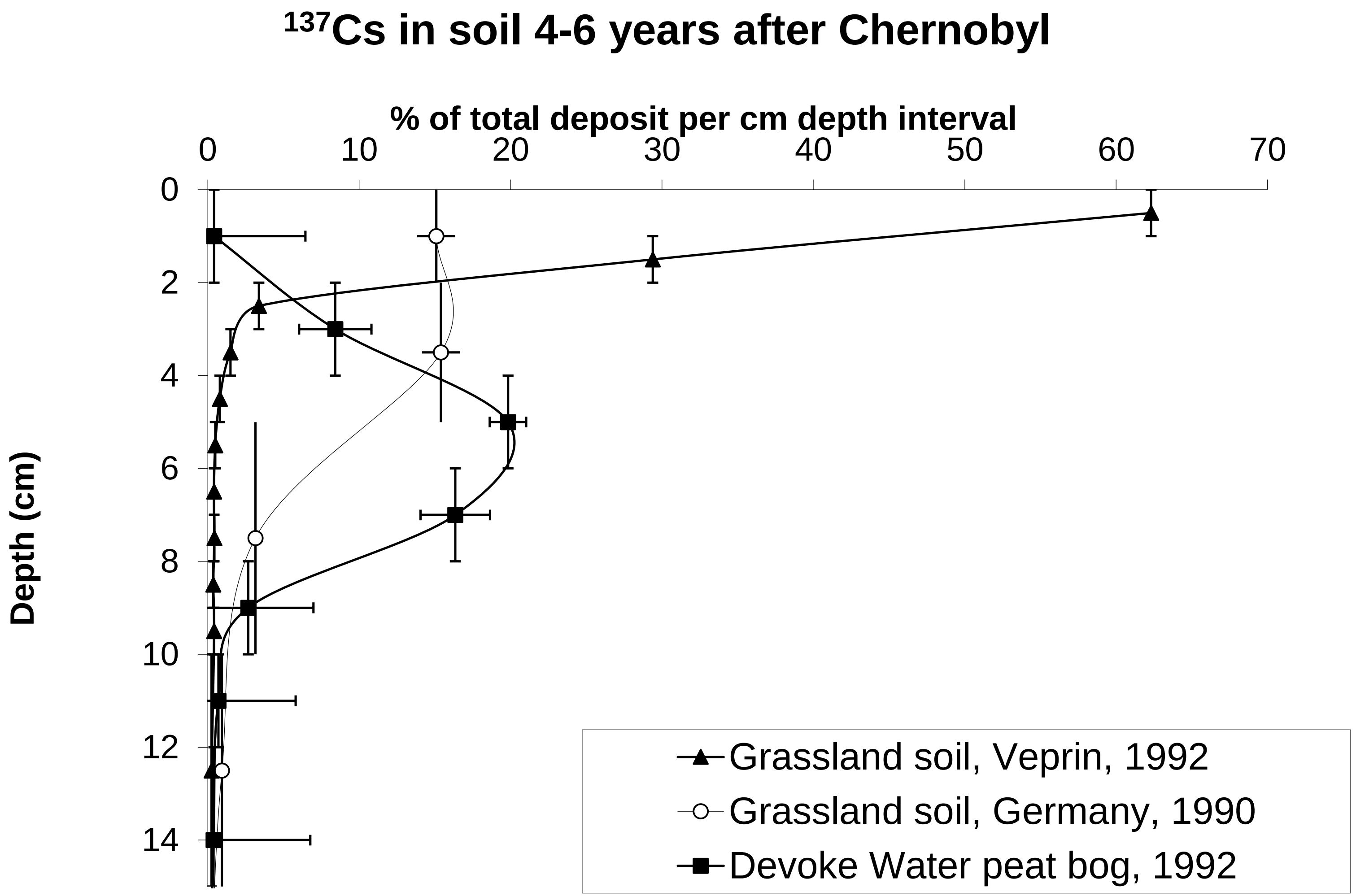


Smith, et al. (1999) *Environmental Science & Technology*, 33, 49-54.

Other processes leading to declines….

- As well as fixation and decay:
  - Slow migration to deeper soil layers by diffusion and bioturbation;
  - Slow removal by runoff in dissolved and particulate phase of river water.

Slow vertical migration in unmixed soils

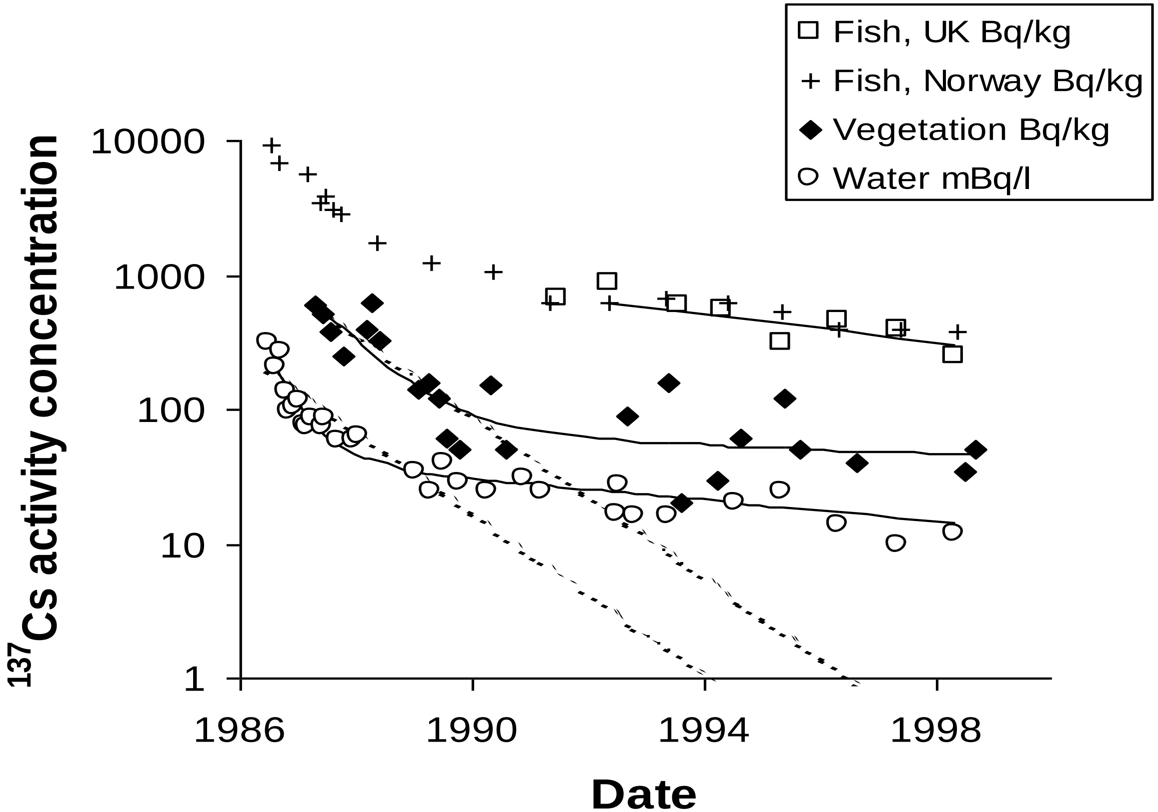


Changes over a longer timescale..

First few weeks
Fast processes of sorption, vertical migration, biological uptake

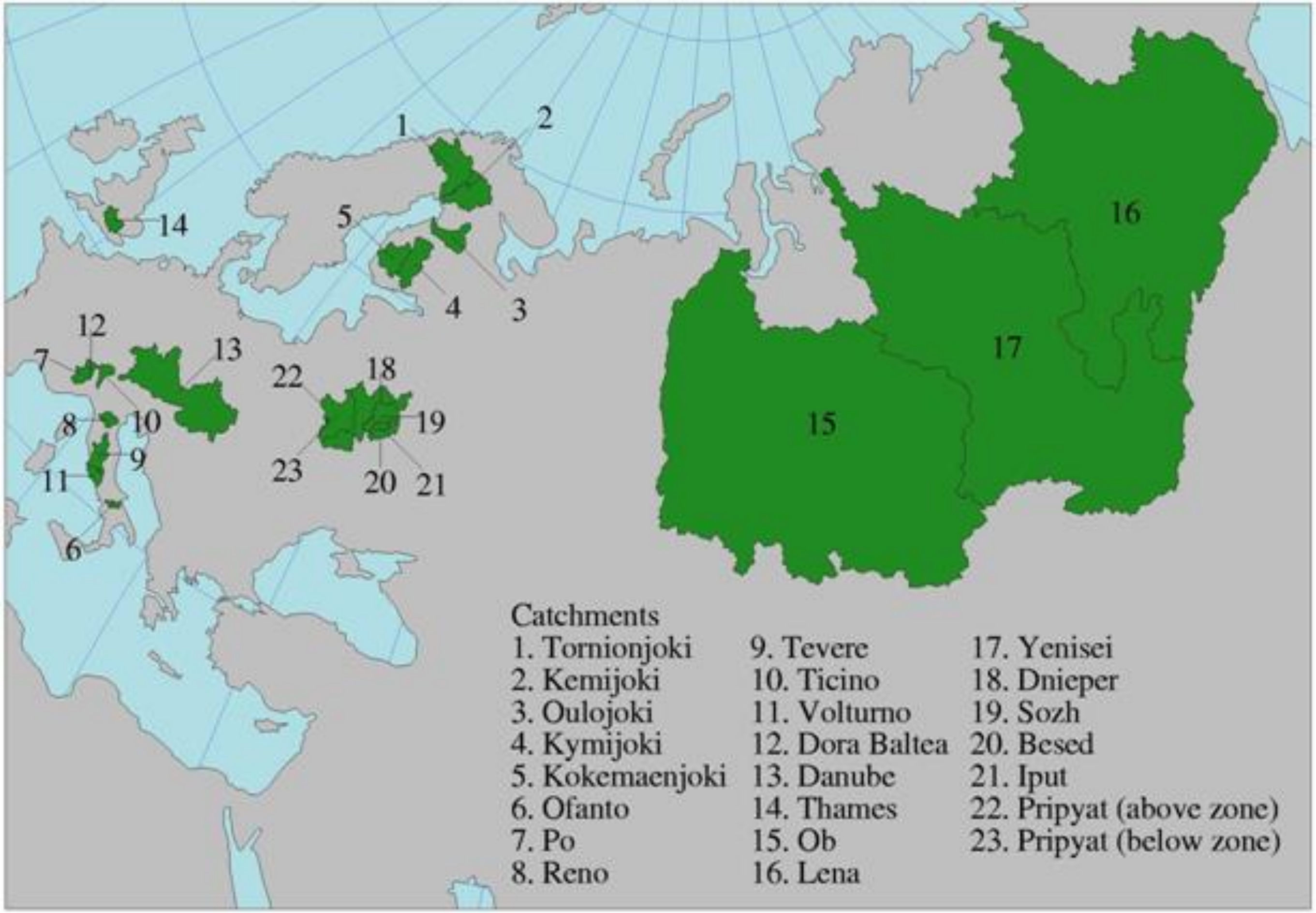
Up to 4-7 years
"Fixation"

More than 4-7 years
Slow desorption, vertical migration, erosion



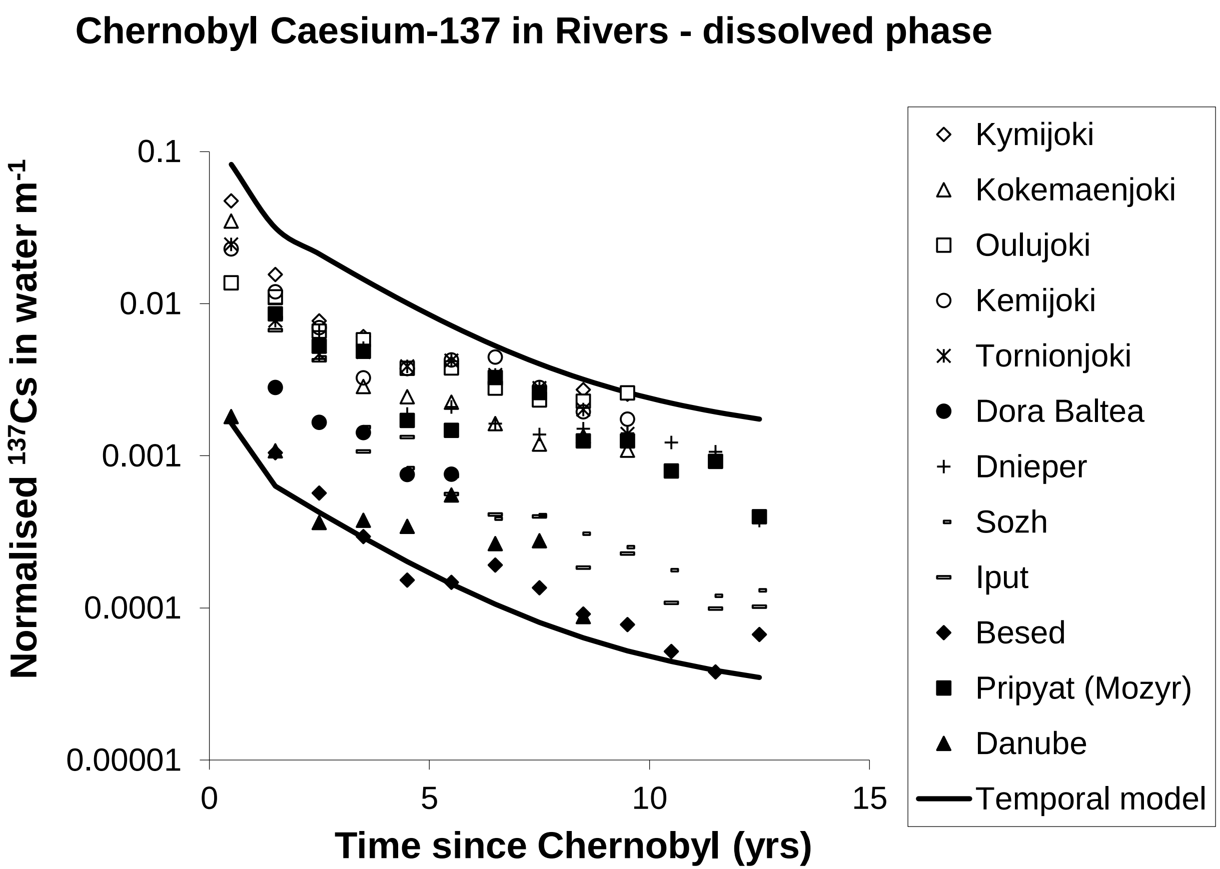
Smith, J.T. et al. (2000) Chernobyl's legacy in food and water. *Nature*, 405.

Runoff from catchments



Cs-137 in rivers contaminated by Chernobyl

$$Runoff\ coefficient = \frac{Concentration\ in\ water\ (Bq/m^3)}{Fallout\ density\ (Bq/m^2)}$$



Analyses for 23 catchments

University of Portsmouth

Radionuclide runoff:

- Percentage Transfer (%)
- Runoff Coefficient (R)
- Effective Ecological Half-Life (T_{eff})

Physical Characteristics:

- Size
- Mean Slope
- Annual Precipitation
- Land Cover Classification
- Soil Texture
- Compound Topographic Index
- Carbon Content

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NATURAL ENVIRONMENT RESEARCH COUNCIL

Results

Summary of the correlations.

*: >99%

*: >95%

*: >90%

: <90%

		Mean Slope (°)	Precipitation (mm y^{-1})	% of Inland Water	Average Soil Texture	% of Clay soils	% of Sand Soil	Mean Soil Wetness (Cti)	Soil Wetness: % >13 (Cti)	Mean Carbon Content	Carbon % > 17
^{90}Sr, NWT	% Transfer	*		*				*			
	Effective Half-Life										
	RunOff Coefficient	*	*	*				*	*		
^{137}Cs, Chernobyl	% Transfer			*			*			*	*
	Effective Half-Life										
	RunOff Coefficient			*			*			*	*

- High dissolved phase ^{137}Cs runoff is associated with high organic matter in catchment soils;

- So-called “Closed” lakes (shallow, slow water inflow/outflow) have high long term radiocaesium;

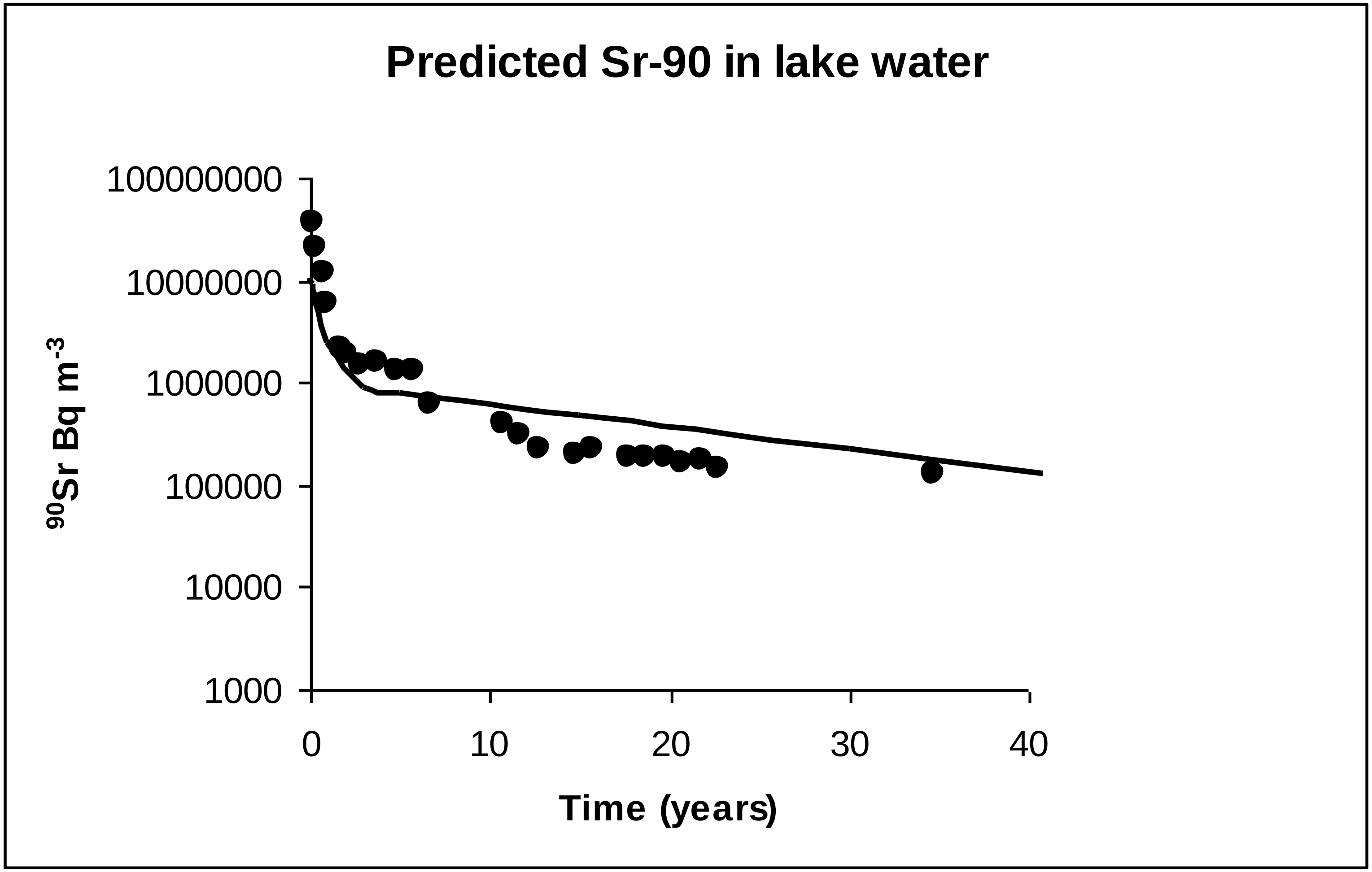
- High uptake to fish in systems with low potassium in the water.

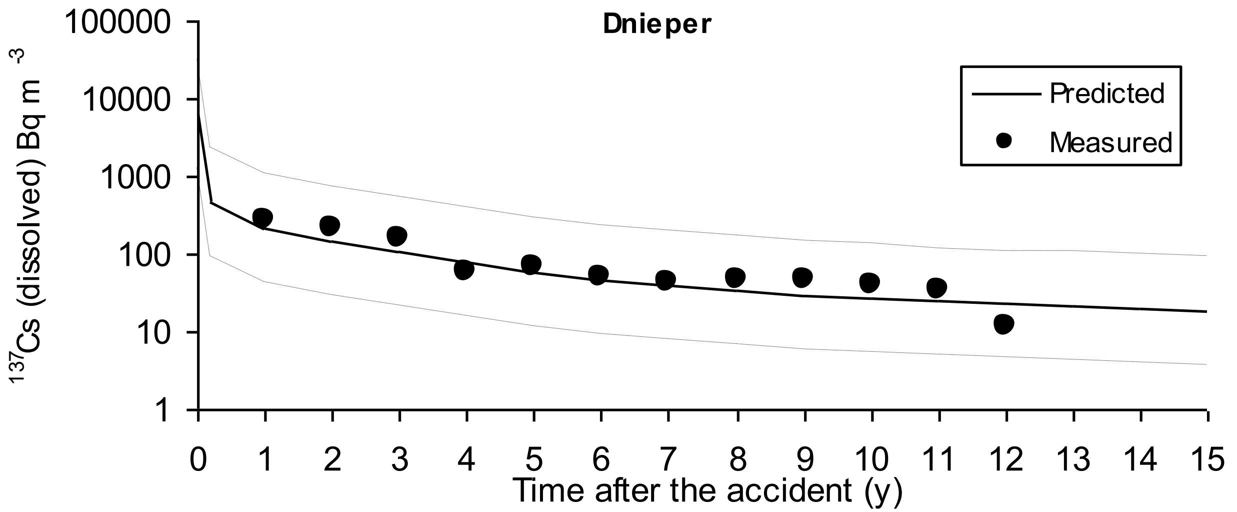
AQUASCOPE Model
for rivers, open lakes and closed lakes

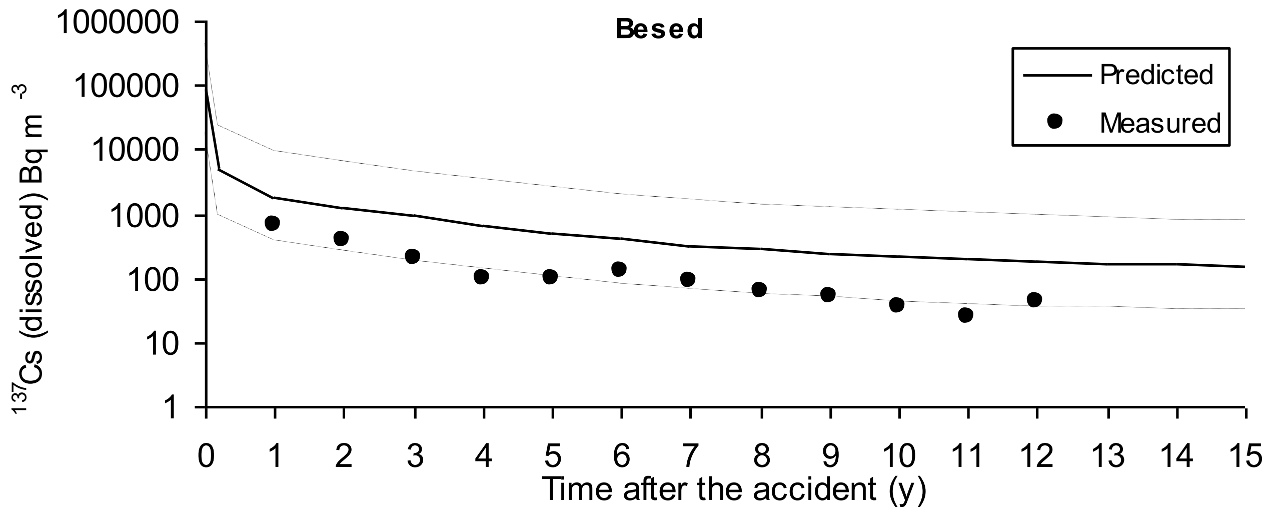
Blind Predictions

Smith et al. (2005) *Health Physics* 89, 628-644.

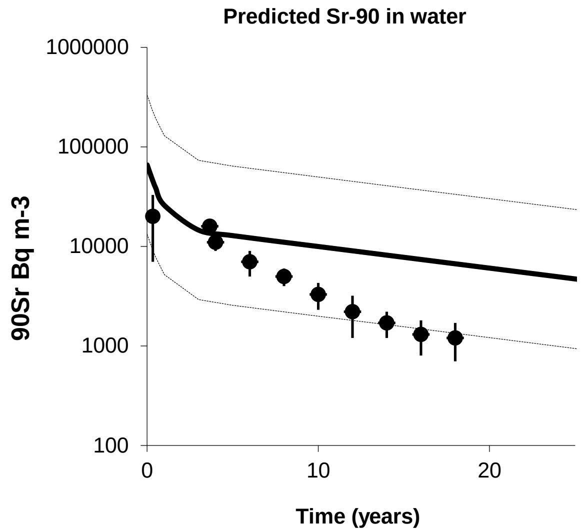
Sr-90 in Lake Uruskul, Siberia

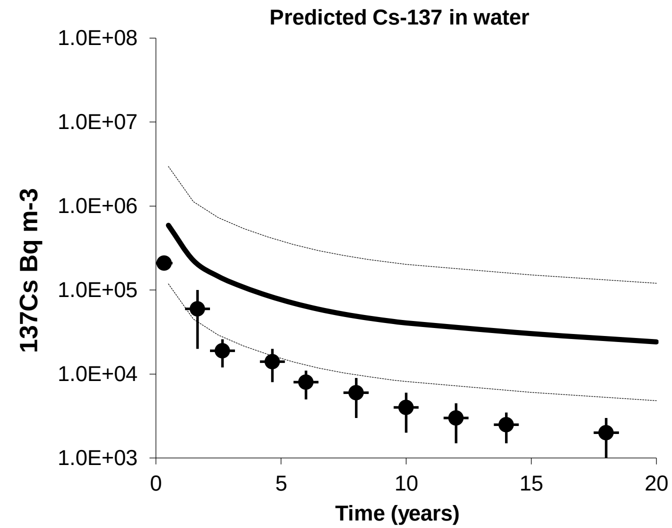


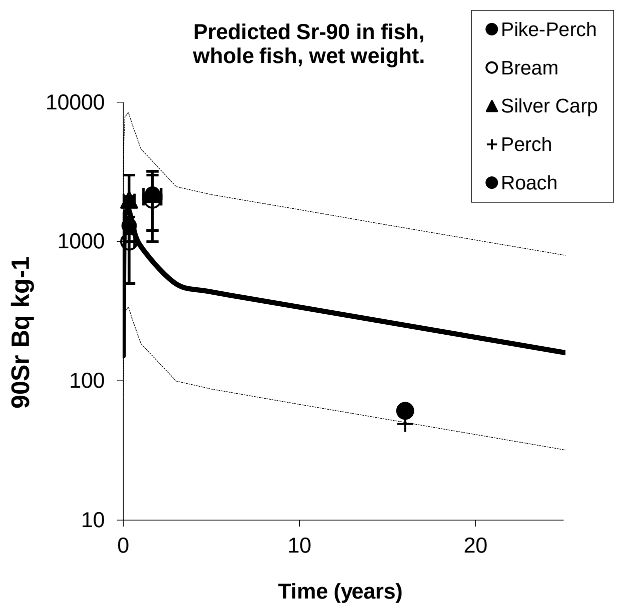

Dnieper
Predicted
Measured
^{137}Cs (dissolved) Bq m^{-3}
100000
10000
1000
100
10
1
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
Time after the accident (y)

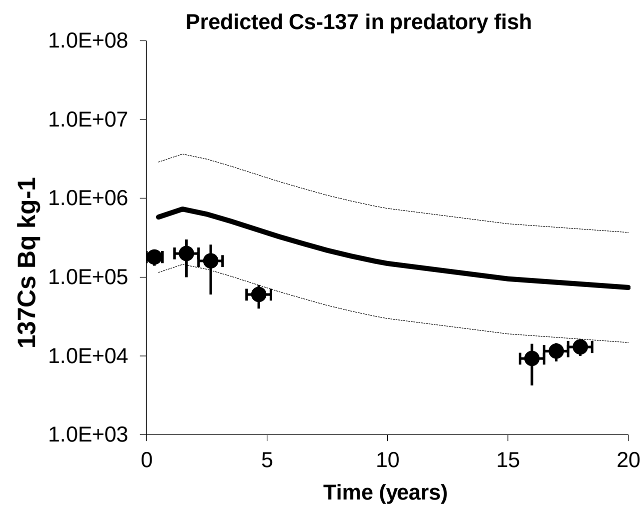

Besed
Predicted
Measured
^{137}Cs (dissolved) Bq m^{-3}
1000000
100000
10000
1000
100
10
1
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
Time after the accident (y)

Chernobyl Cooling Pond Predictions

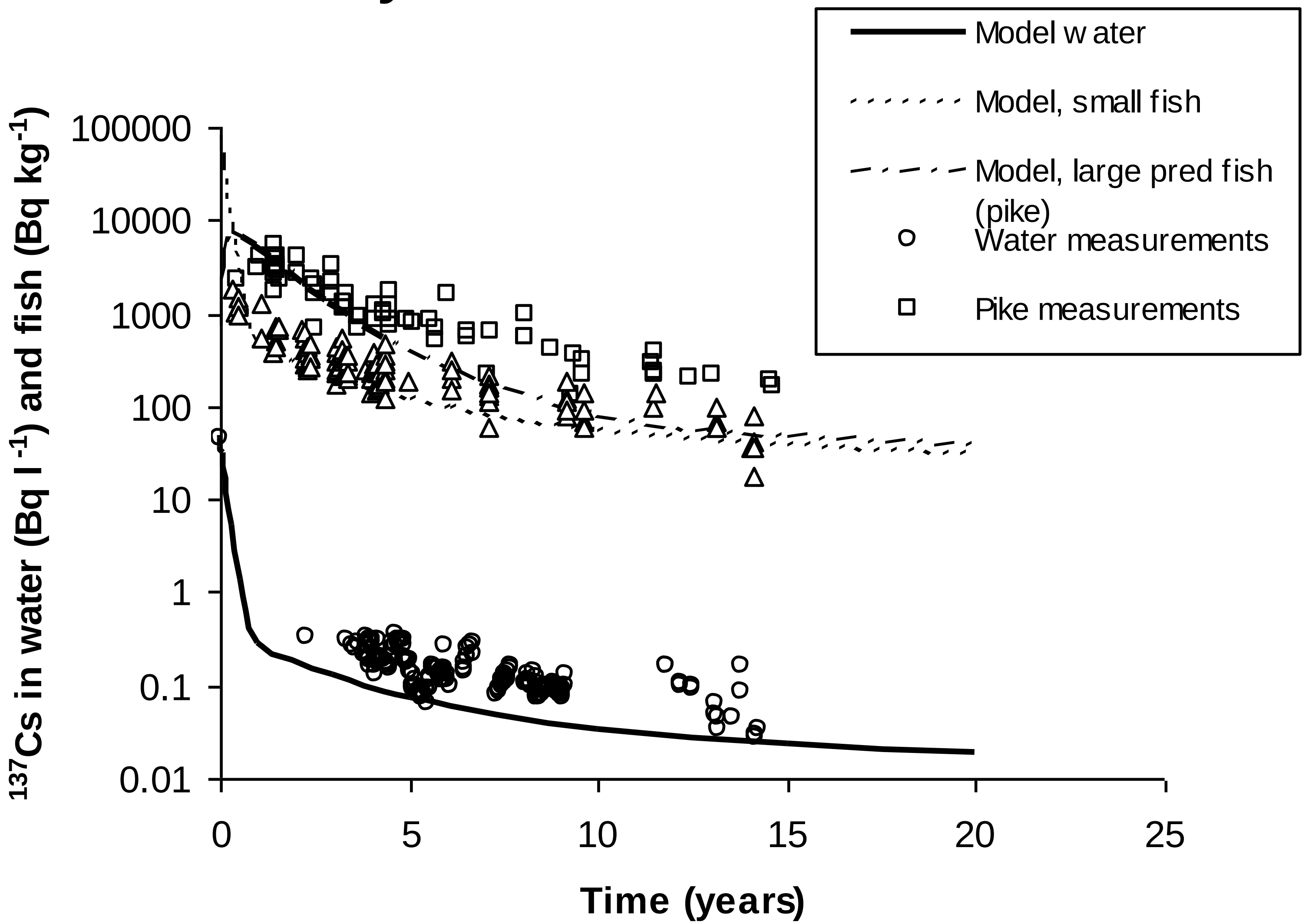






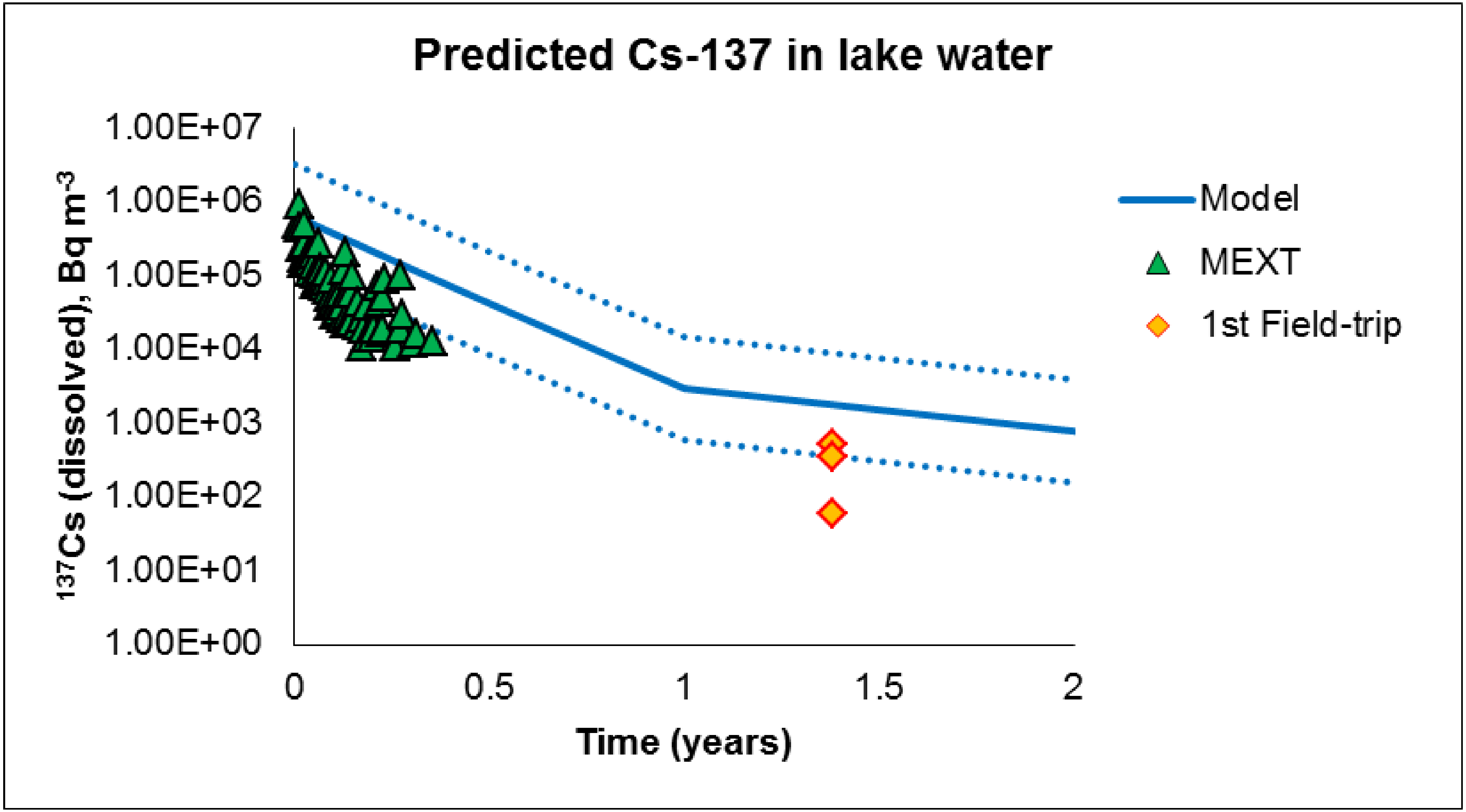


Cs-137 in Lake Vorsee, Germany after Chernobyl



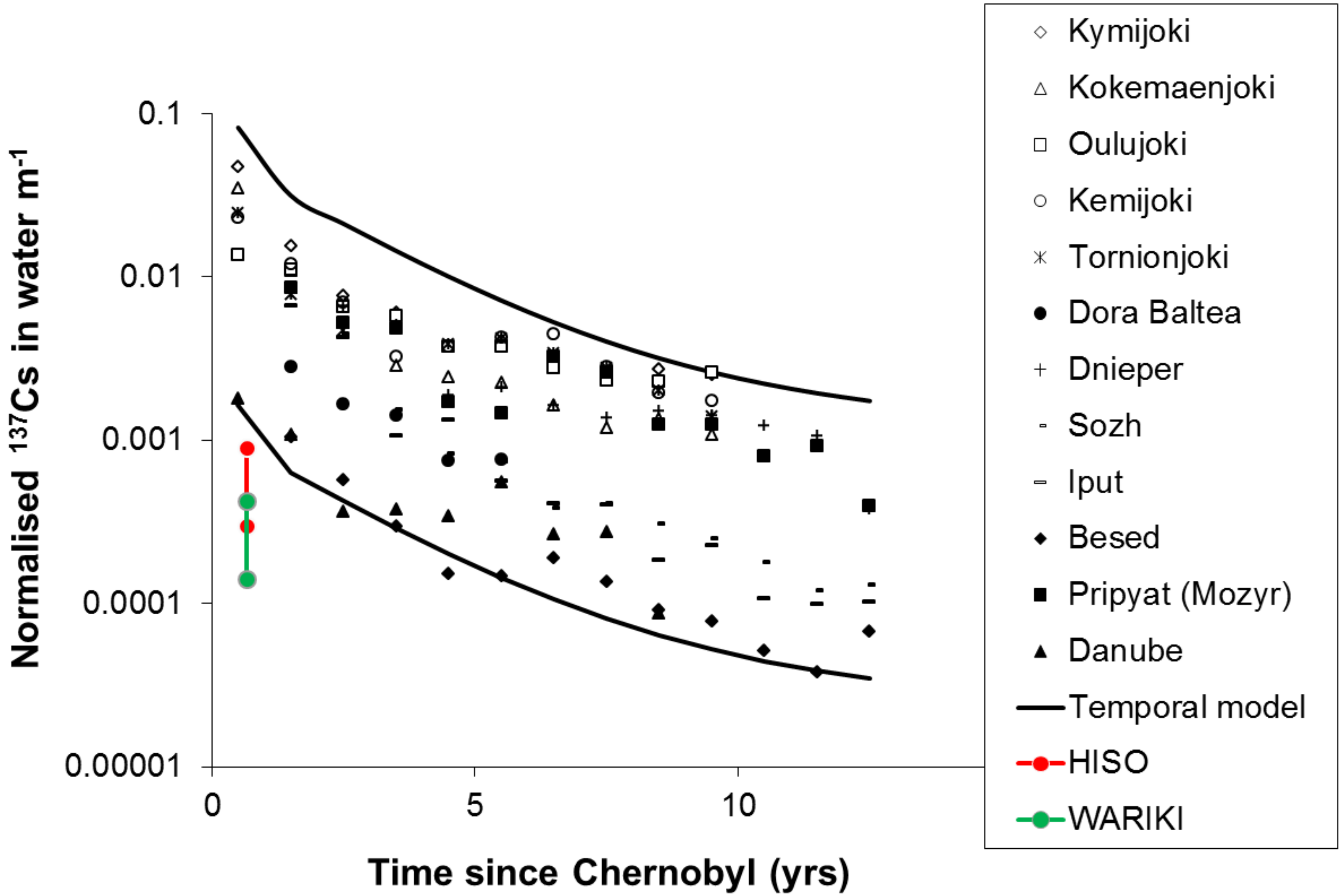
Data from Gregor Zibold, Eckehart Klemt (Fachhochscule Weingarten)

Iitate lake water

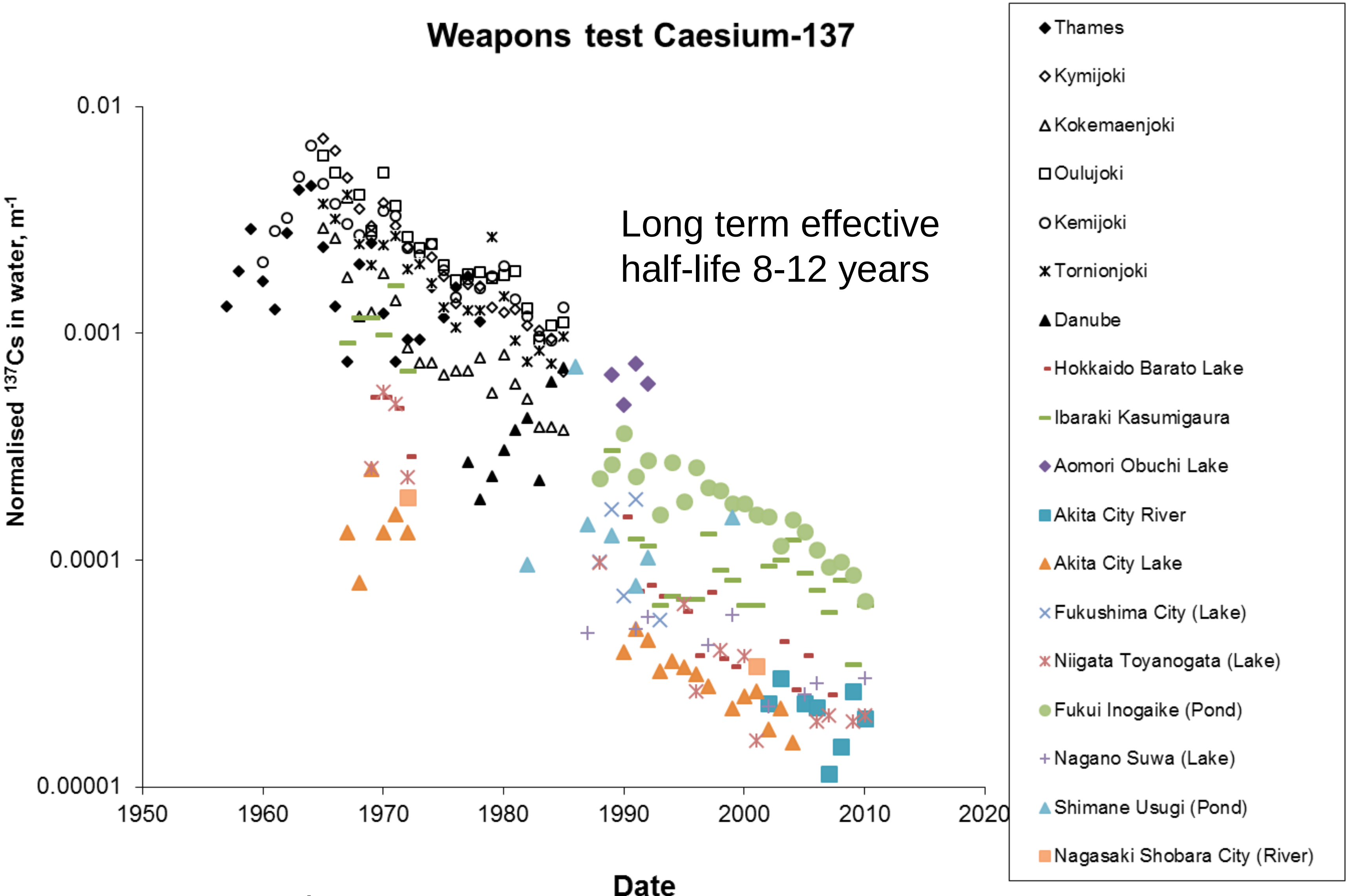


Blind prediction of AQUASCOPE model – Iitate Lake
Chaisan, K., Kameda, Y. (Chiba Inst Technol.), Smith J.T., unpubl. res.

Cs-137 in European Rivers compared to Japanese



Smith J.T., et al. (2004) *Env. Sci. and Technology*, 38, 850-857.
Ueda et al. (2013) *J. Env. Radioactivity,*118; 96-104

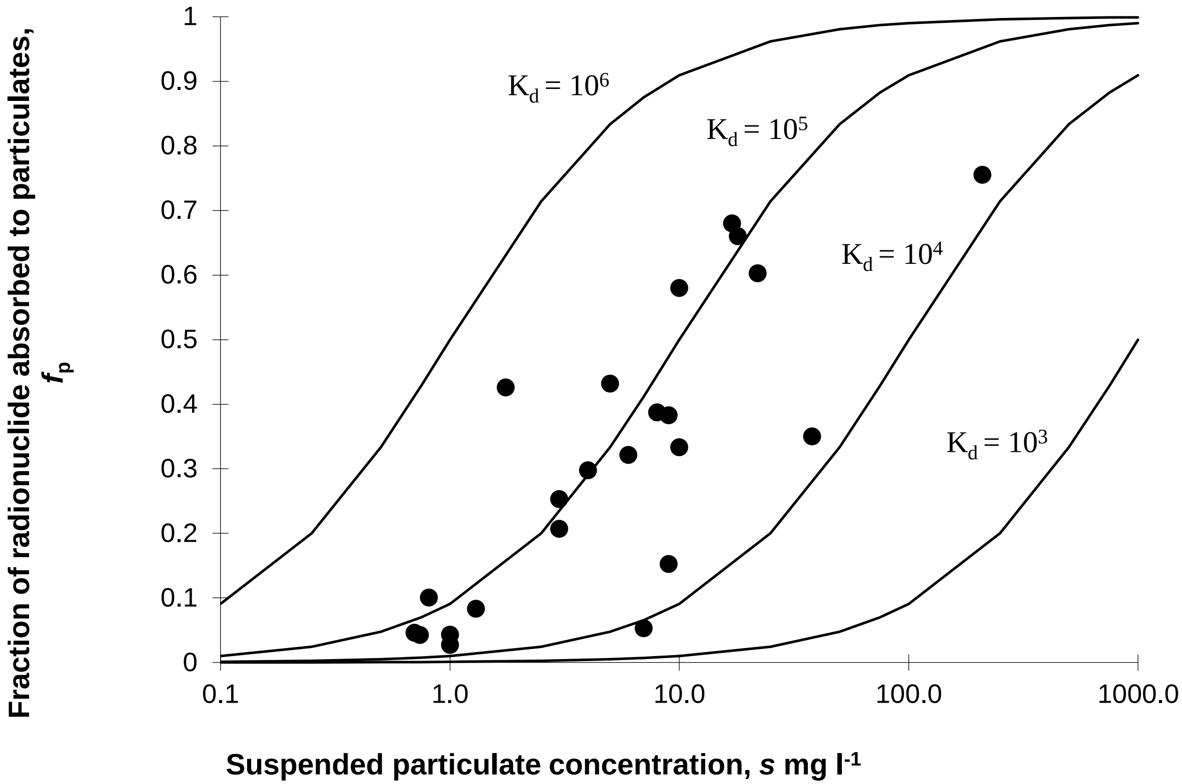


Note: Measurements in Europe are dissolved phase, in Japan total (dissolved+solid)

Jim Smith (Unpublished data) with Keiko Tagami, Shigeo Uchida, NIRS, Japan.

K_d:

$$K_d\ \left({}^{l}/_{kg}\right) = \frac{Concentration\ in\ solid\ phase\ (Bq/kg)}{Concentration\ in\ dissolved\ phase\ (Bq/l)}$$



Data for European lakes from Smith and Beresford (2005)

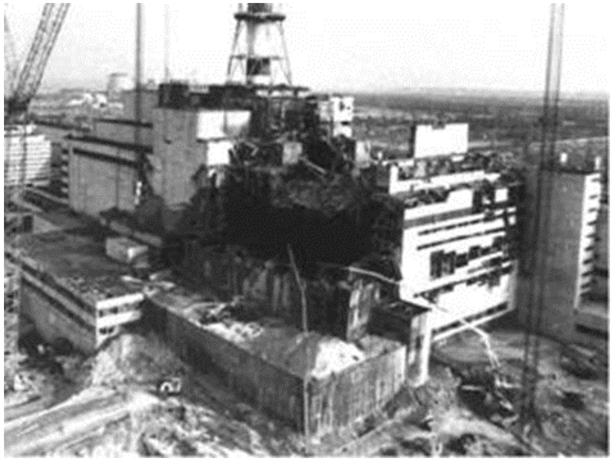
Very high K_d in many soils and sediments in Japan !

(Konoplev, A., et al. *J. Environmental Radioactivity* 151 (2016): 568-578.)

Summary

- Modelling the extensive data from the nuclear weapons test period and Chernobyl can help predict future contamination from the Fukushima accident
- Activity concentrations in freshwater fish can remain high for decades, particularly in “closed” lakes with low potassium concentration of water.
- Soils/sediments in Japan seem to have a high binding capacity for Cs (high Kd)

Aquatic countermeasures



Freshwater dose pathways

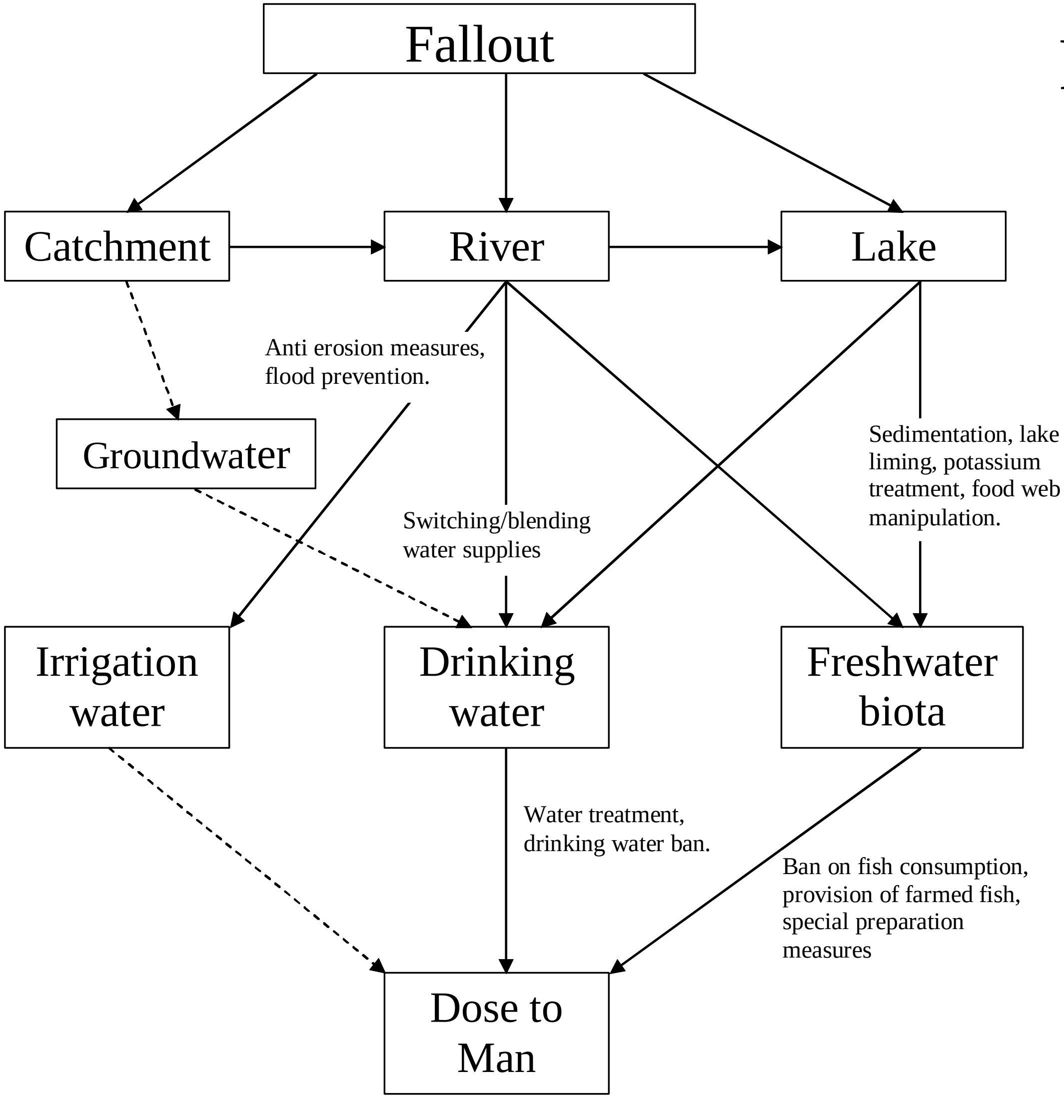


Figure 1. Freshwater dose pathway indicating potential intervention measures. Dashed lines indicate pathways of lower potential importance.

Smith, J.T., Voitsekhovitch, O. et al. A critical review of aquatic countermeasures *J. Env Rad.* **56, 11-32.**

Fear of contamination of the Pripyat-Dnieper reservoir system

Radionuclide levels (dissolved phase) in the River Pripyat at Chernobyl

(Vakulovsky & Voitsekhovitch, 1990; Voitsekhovitch et al. 1991; Vakulovsky et al. 1994; Kryshev, 1995).

RN	Half- life	Radionuclide concentration in water, Bq l^{-1}							
		01/05/86	02/05/86	06/05/86	03/06/86	16/07/86	9/8/86	1987 (mean)	Committed effective dose during 1st year (mSv)
^{137}Cs	30.2 y	250	555	1591	22.2	7.4		1.8	0.57 (a)
^{134}Cs	2.1 y	130	289[1]	827[1]	11.5[1]	3.8[1]		0.94[1]	0.43 (a)
^{131}I	8.1 d	2100	4440	814	33.3	< 0.82 [2]			4.2 (i)
^{90}Sr	28 y	30			1.9			1.5	0.049 (c)
^{140}Ba	12.8 d	1400							
^{99}Mo	3 d	670							
^{103}Ru	40 d	550	814	170	26	15			0.053 (i)
^{106}Ru	365 d	183 [3]	271 [3]	57 [3]	8.7 [3]	5 [3]			0.29 (i)
^{144}Ce	284 d	380				37			
^{141}Ce	33 d	400		89		14.8			
^{95}Zr	65 d	400	1554	167	11	37			
^{95}Nb	35 d	420							
^{241}Pu	13 y	33 [4]					0.6 [4]		0.072 (a)
$^{239+240}$Pu	2.4×10^{4} y 6.6×10^{3} y	0.4					0.0074		0.0046 (a)

1. From 137Cs measurement and 134Cs/137Cs ratio ~ 0.52.
2. Assuming decline from 03/06/86 value by radioactive decay only.
3. From Ru-103 measurement and assuming Ru-103/Ru-106 ratio (~ 3) for Chernobyl fallout.
4. From Pu-239,240 measurement and Pu-241/Pu-239,240 ratio (~ 82) for Chernobyl fallout.

Freshwater internal doses - minor compared to terrestrial

- Drinking water: dose very minor except in the first few weeks;
- Freshwater fish/foodstuffs – activity concentrations/doses can be very high, but doses generally low (low consumption rates) except to critical groups (e.g. fishermen and their families);

Dam which cut contaminated bay from Pripyat River near ChNPP

Floating alternative water supply system near Kiev from Desna River

Filtration dyke on Illya River

Most of these actions were found to be inefficient because a significant part of the radionuclide activity was in dissolved forms and washed out from the catchment areas beyond of ChNPP zone. Most efficient were accepted - short term restriction on water use from contaminated sources

Drinking water pathway – short term

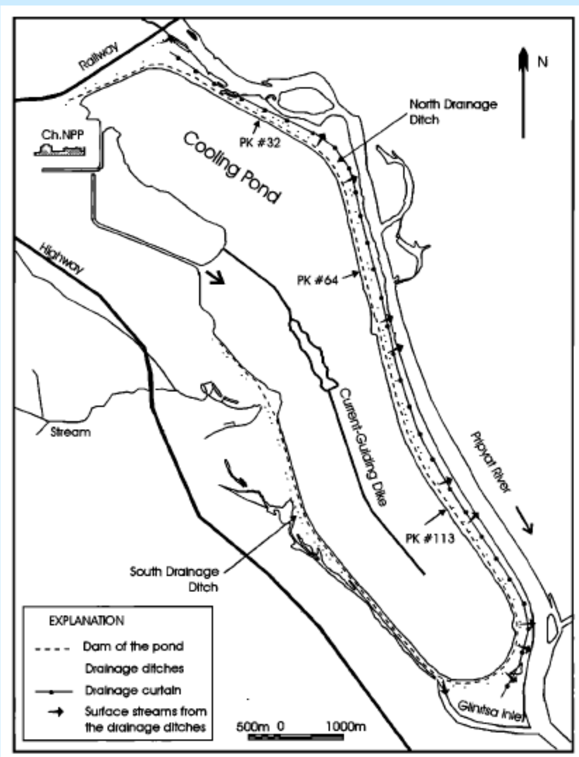
- Switching/blending supplies is effective, but requires rapid response and an integrated network.
  - Groundwaters typically clean.

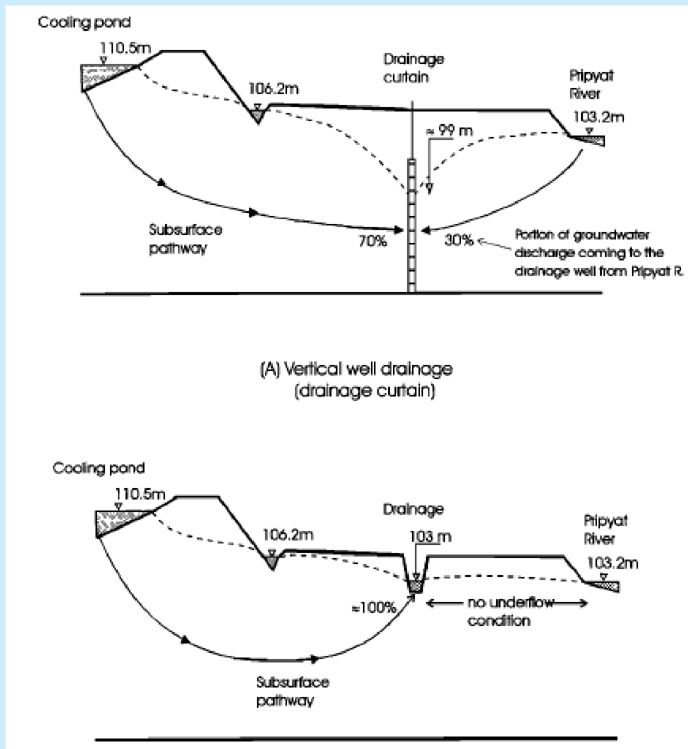
- Radioactivity removed at water treatment:
  - Removal efficiencies
    - Cs 56%; Ru 73%; Co 61%; I 17%.
  - Activated charcoal found to be effective

Smith, J. T., et al. "A critical review of measures to reduce radioactive doses from drinking water and consumption of freshwater foodstuffs." *Journal of environmental radioactivity* 56.1 (2001): 11-32.

Countermeasure to mitigate secondary contamination of Pripyat River with the groundwater

Measures were taken to protect seepage groundwater from radionuclides from the shelter and from radioactive waste sites in the CEZ. These measures focused mainly on the construction of engineering and geochemical barriers around the local hot spots to reduce groundwater fluxes to the river network. Actions to stop precipitation from entering the shelter, and drainage of rainwater collected in the bottom rooms of the shelter, have also to be considered as preventive measures to reduce groundwater contamination around the Chernobyl nuclear power plant industrial site.



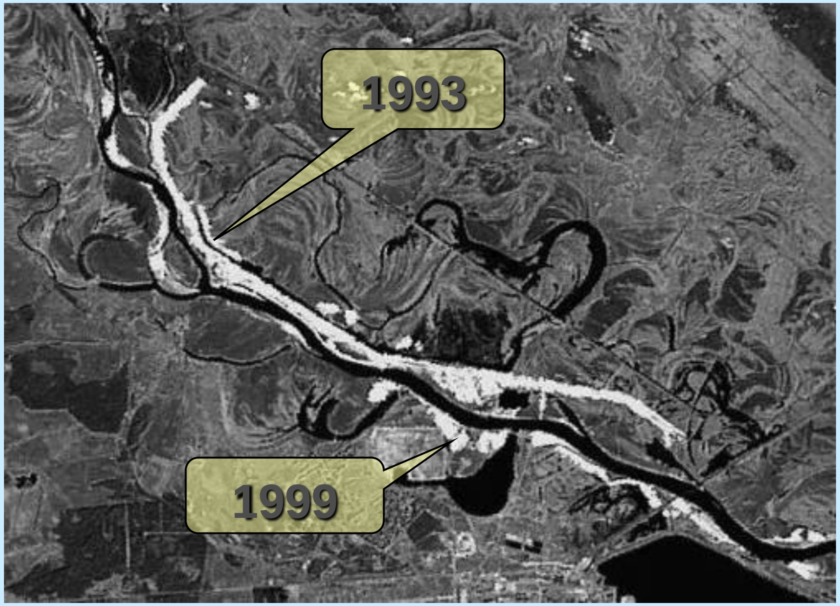


Bugai, DA., et al. "The cooling pond of the Chernobyl nuclear power plant: a groundwater remediation case history." Water resources research 33.4 (1997): 677-688.

Groundwater residence times are long enough and shorter lived radionuclides such as I-131 will be decayed long before they could reach drinking water. Only very small amounts of Cs-137 percolate from surface soils to groundwater

However, contamination of groundwater from buried radioactive waste could lead to significant doses in some circumstances

General View



Flood protective dam has been constructed

Drinking water pathway – long term

- Sediment trapping dams were ineffective;
- Temporary increases in Sr-90 (from ~ 1Bq/l to 10 Bq/l during spring flooding of the Pripyat.
- Dam around the Pripyat floodplain was effective, but only implemented some years after the accident and only reduced relatively small peaks in ^{90}Sr concentration during flooding – effectiveness in dose reduction (or not) is controversial.

Drinking water pathway

- Levels of radioactivity in Kiev Reservoir water fell within one year to around 1 Bq/l or less.
- This compares with
  – up to 6 Bq/l in some mineral waters;
  – 10-13 Bq/l in sea water.
  – Natural potassium-40 in some foodstuffs of 100-200 Bq/kg

Drinking water pathway

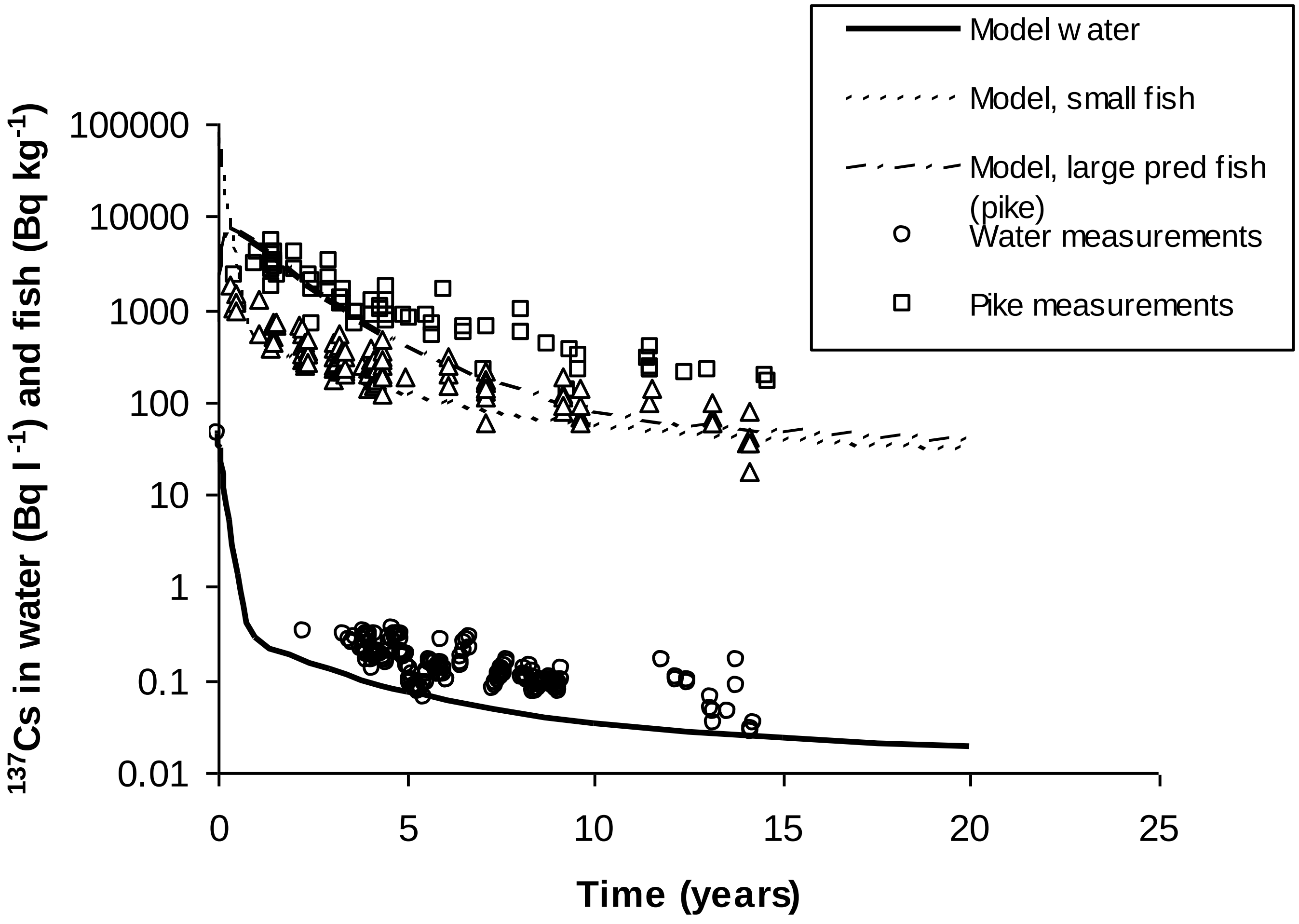
- Public concern over drinking water supply was much greater than the real risk.

Fish consumption pathway

Fish consumption pathway

- I-131 in fish from the Kiev Reservoir
  - 6000 Bq/kg (May 1986)
  - 50 Bq/kg (end June 1986)
- Cs-137 in fish:
  - Devoke Water, UK ~ 1000 Bq/kg in 1987
  - Lake Vorsee, Germany < 5000 Bq/kg, 1987
  - Lake Svyatoe, Belarus up to 100,000 Bq/kg, 1997
  - Kiev Reservoir, Ukraine ~ 1000 Bq/kg (1987), 500 Bq/kg (1993)
- Sr-90 in fish
  - Much less significant than Cs-137

Cs-137 in Lake Vorsee, Germany



Data from Gregor Zibold, Eckehart Klemt (Fachhochscule Weingarten)

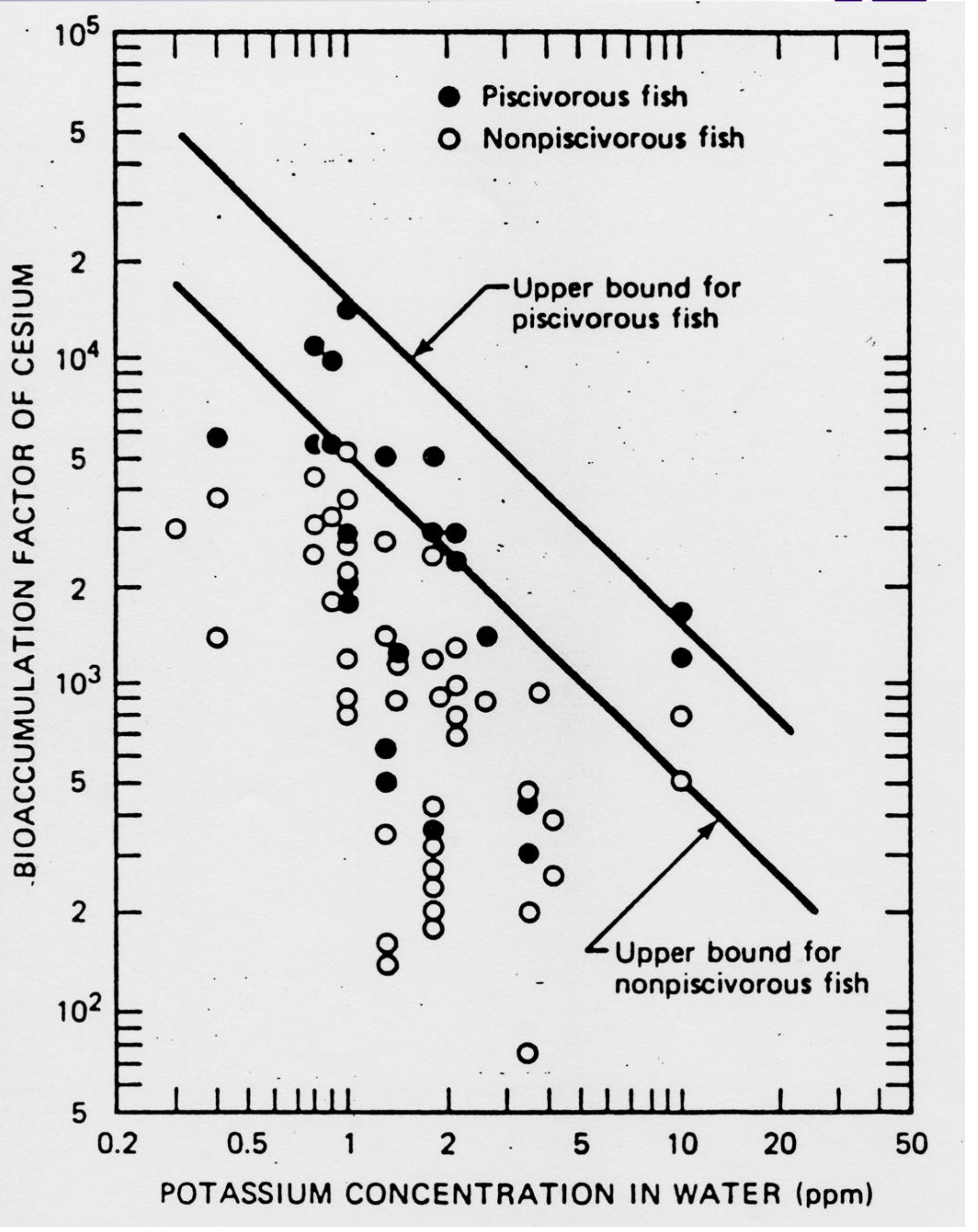
Fish consumption pathway

- Bans on sale/consumption of freshwater fish in parts of fSU, Scandinavia and Germany;
- Guidelines on maximum recommended consumption rates may be appropriate;
- Bans/recommendations may not be adhered to.

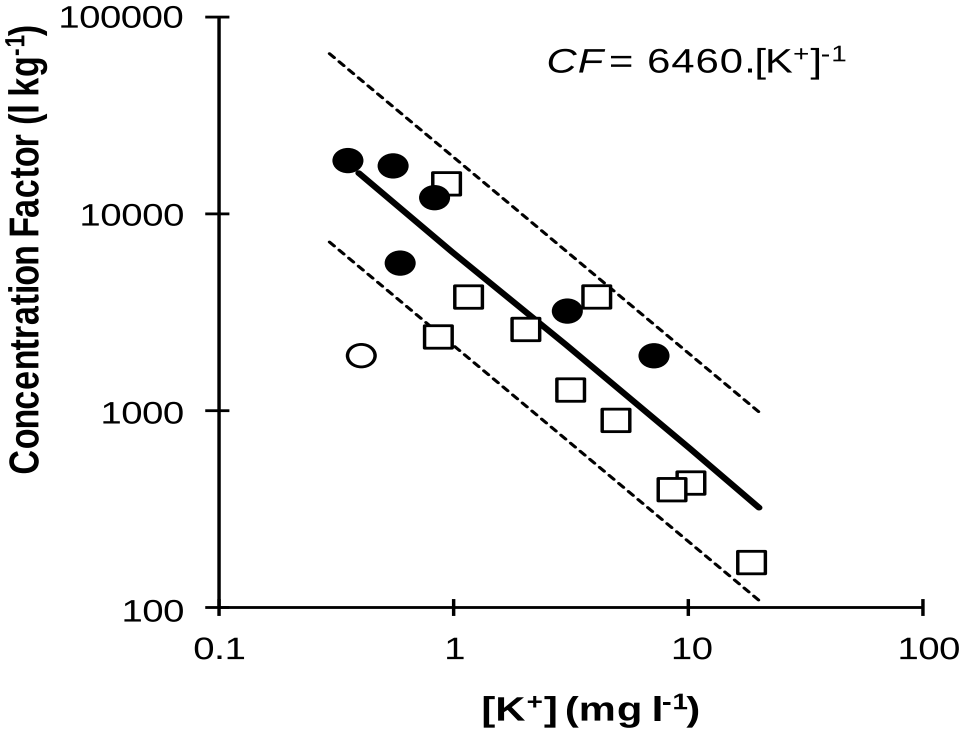
Testing a countermeasure

Caesium-potassium model from measurements in lakes pre-Chernobyl

Vanderploeg et al. 1975



Cs-137 in Perch

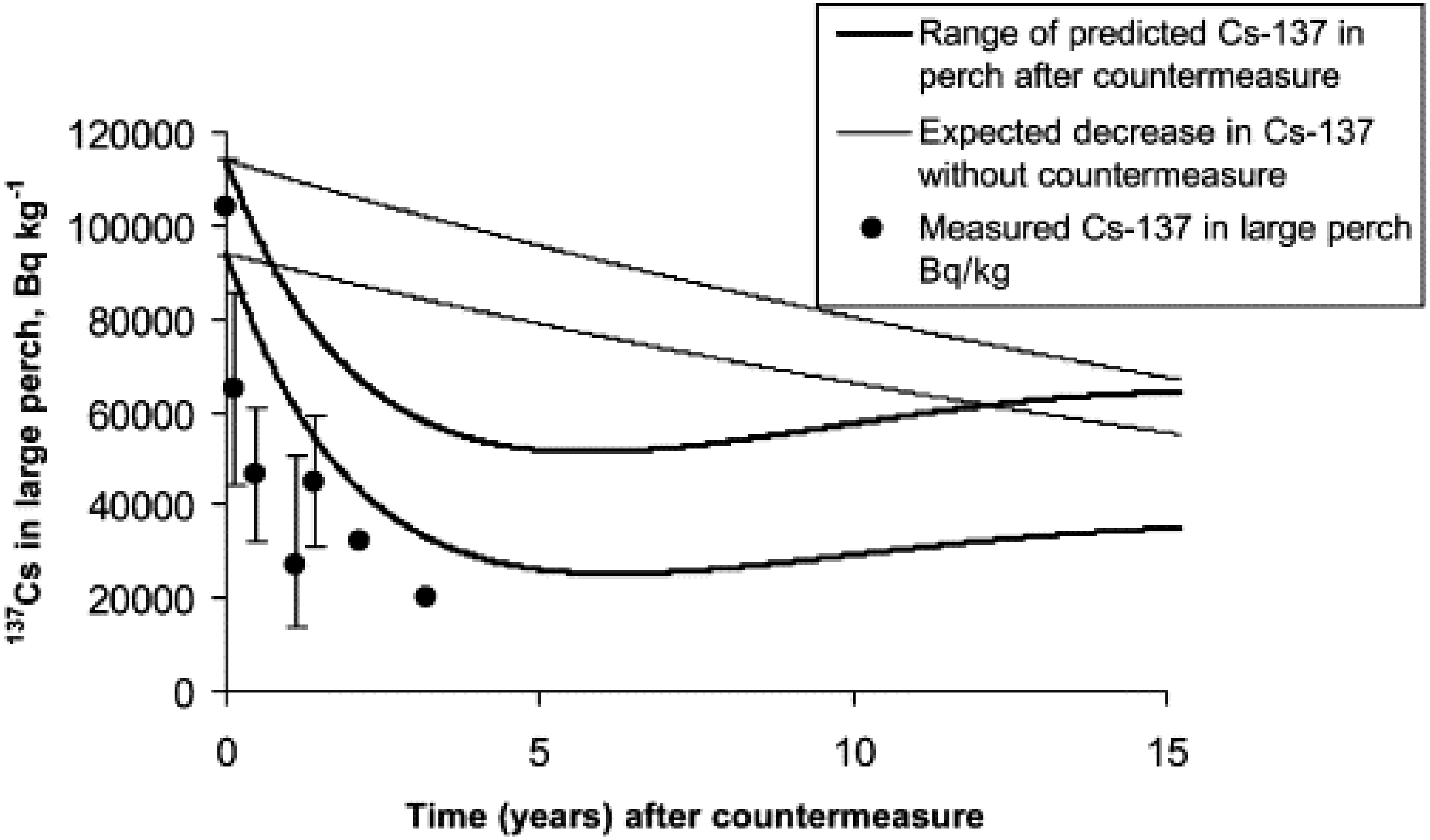


Cs-137 in fish goes down as potassium goes up

Can we use this to reduce Cs-137 in fish ?

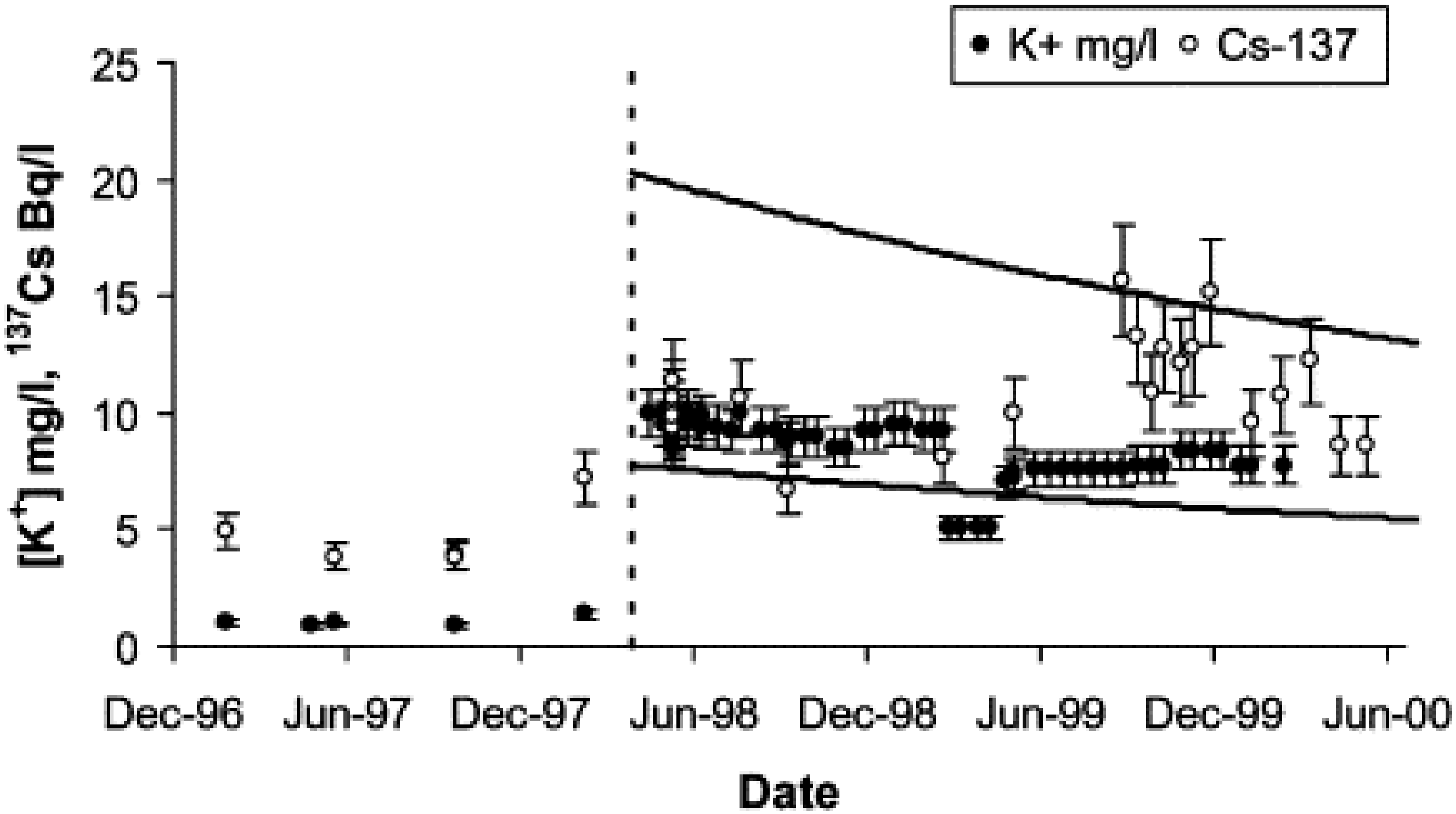
“AQUACURE” project with Belarus Academy of Sciences

Result of 10 x increase in potassium in L. Svyatoe, Belarus



Smith, James T., et al. *Science of the Total Environment* 305.1 (2003): 217-227.

Changes in water chemistry



Countermeasure for Sr-90 ?

- A similar inverse relationship between ^{90}Sr in fish and calcium concentration in water has been observed.

- Ca additions could work in the case of ^{90}Sr contamination, but this has not been tested.

Fish consumption pathway

- Disadvantages of chemical additions:
  - Can be expensive;
  - Can influence the ecology of the water body;
  - Problems maintaining high K or Ca levels;
  - Countermeasure can increase radioactivity in the water.

Fish consumption pathway

- Food preparation measures can be effective:
  - For ^{90}Sr contamination, eat only muscle tissue (~90% of ^{90}Sr is in the bones and skin);
  - Salting fish may significantly reduce ^{137}Cs concentrations, but affects taste, nutritional value of the fish;
  - May not be publically acceptable

Fish consumption pathway

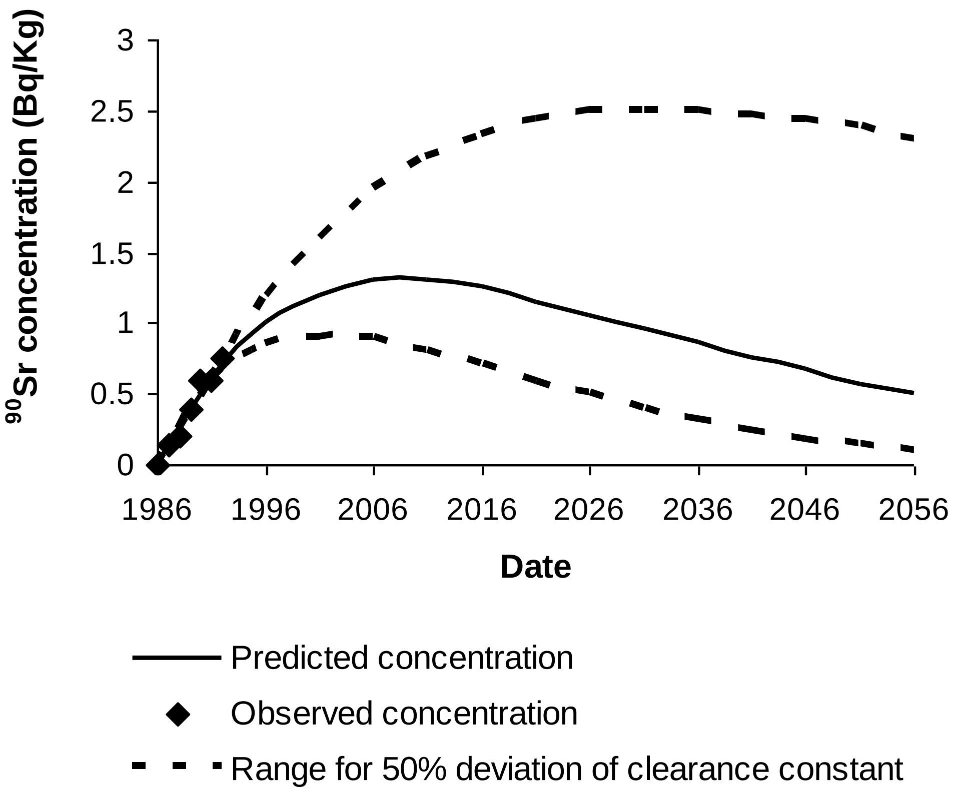
- Fish farms:
  - ^{137}Cs is absorbed by the fish mainly via ingestion: farmed fish (eating clean food) relatively uncontaminated.
  - ^{90}Sr may also be absorbed via the gills.
  - Chemical additions could work well for fish farms but may not be necessary.

Irrigation water pathway

Irrigation water pathway

- For irrigated land contaminated by atmospheric fallout, irrigation will make little difference to concentrations in crops.
- Irrigation can affect “clean” land in the lower parts of a catchment.

Irrigation water pathway



Irrigated rice,

Dnieper Basin

(Perepelyatnikov & Prister, 1992)

Irrigation water pathway

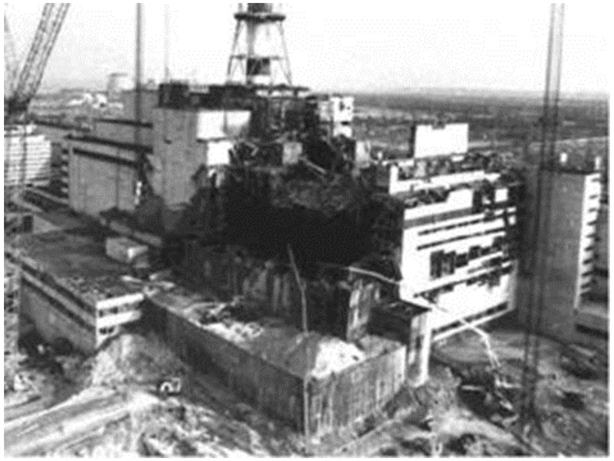
- Surface irrigation likely to be less affected than spray since soils will immobilise a significant proportion of the radioactivity.

Disadvantages of countermeasures

- Often very expensive – is the cost per Person-Sv justifiable ? E.g. if 10-20 psnSv = one averted fatal cancer, and a life is valued at \$X Million, cost per psnSv $\leq$ \$X M ÷10-20
- Dose to clean-up workers; waste generation
- Potential unintended consequences to humans (e.g. salting fish can reduce Cs, but excessive salt intake may outweigh health benefit). Also potential ecosystem damage from large scale measures.

Lesson 4: Sometimes doing nothing ("Monitored Natural Attenuation") can be the best option.

Effects on aquatic organisms



- Immediate effects:
  - “Red” forest died & was cut down.
  - Some cattle died (2nd generation “normal”)

Some people still find big effects..

Anders Moller
University of Paris
Tim Mousseau,
University of South Carolina

“Poisoned Wilderness”

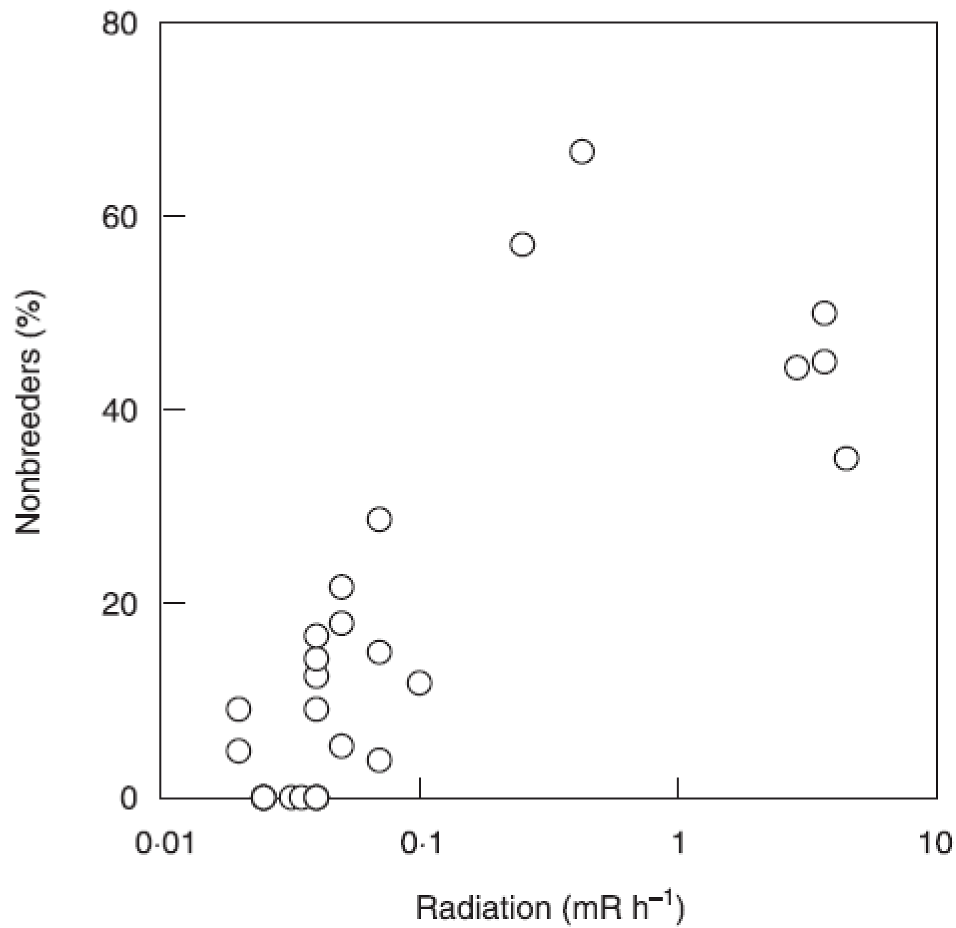
BBC Nature website
27 July 2011

Barn Swallows at Chernobyl

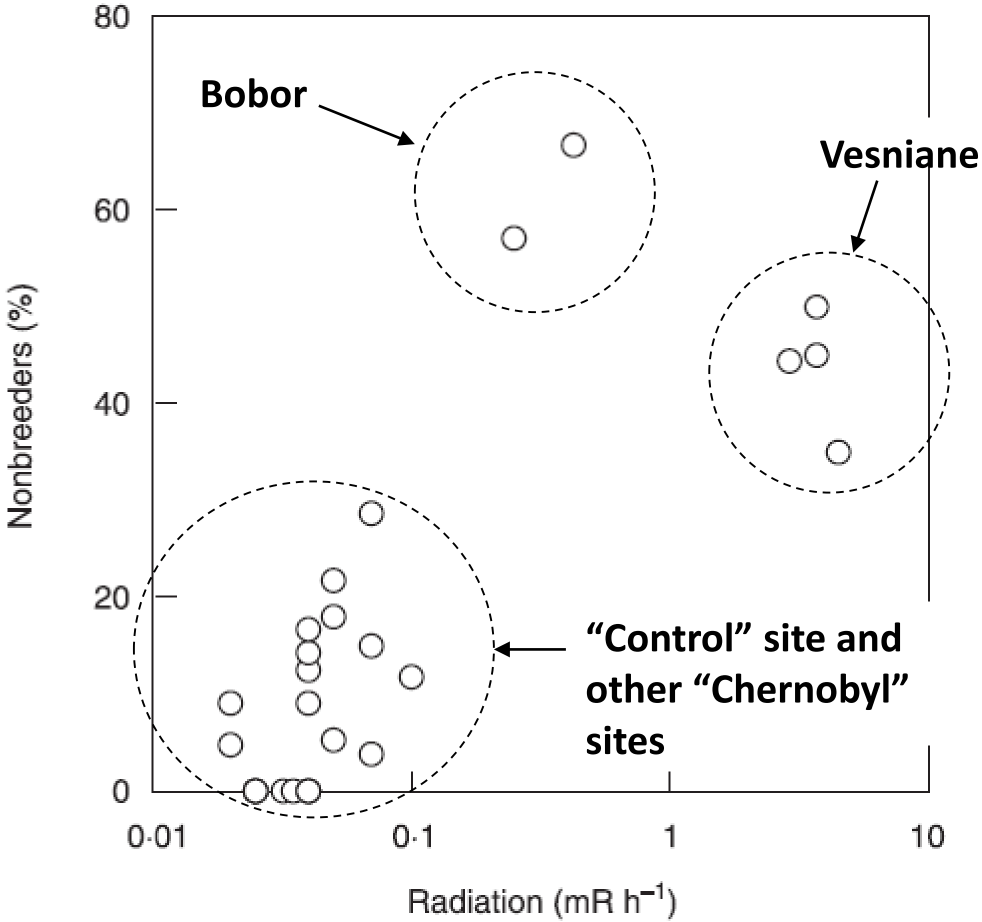
University of Portsmouth

Barn Swallow, *Hirundo rustica*. Photo by sannse, North Devon, 14 May 2004

Moller et al. J. Animal Ecol. (2005)



Barn Swallows at Chernobyl



"we drove along all public roads to visit villages and collective farms in areas with high levels of radiation ***outside the exclusion zone*** near Chernobyl"

"While visits to areas with high levels of radiation is non-random, we see no reason why such a selection should be associated with farming practice, soil quality or abundance of insects, which are the main food for barn swallows".

Moller et al. *J. Animal Ecology,* (2005)

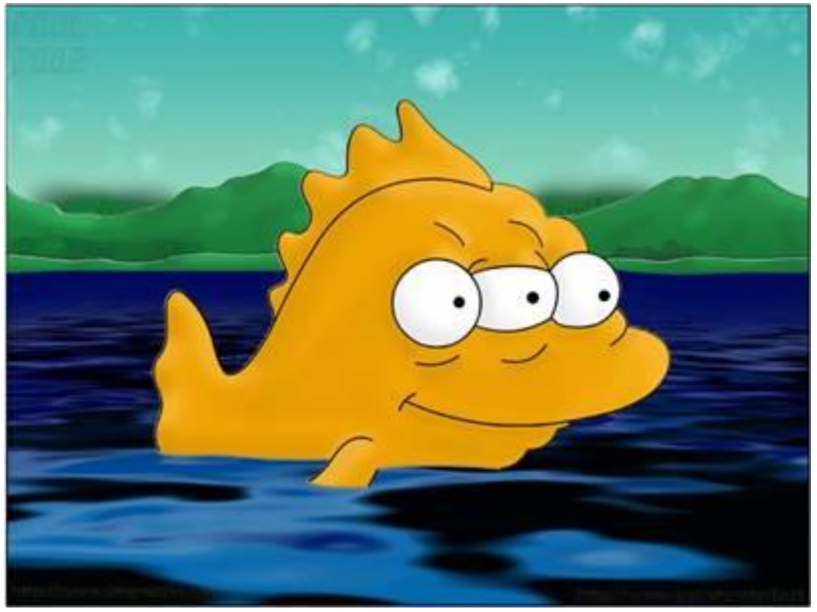
Bobor Village, 2007

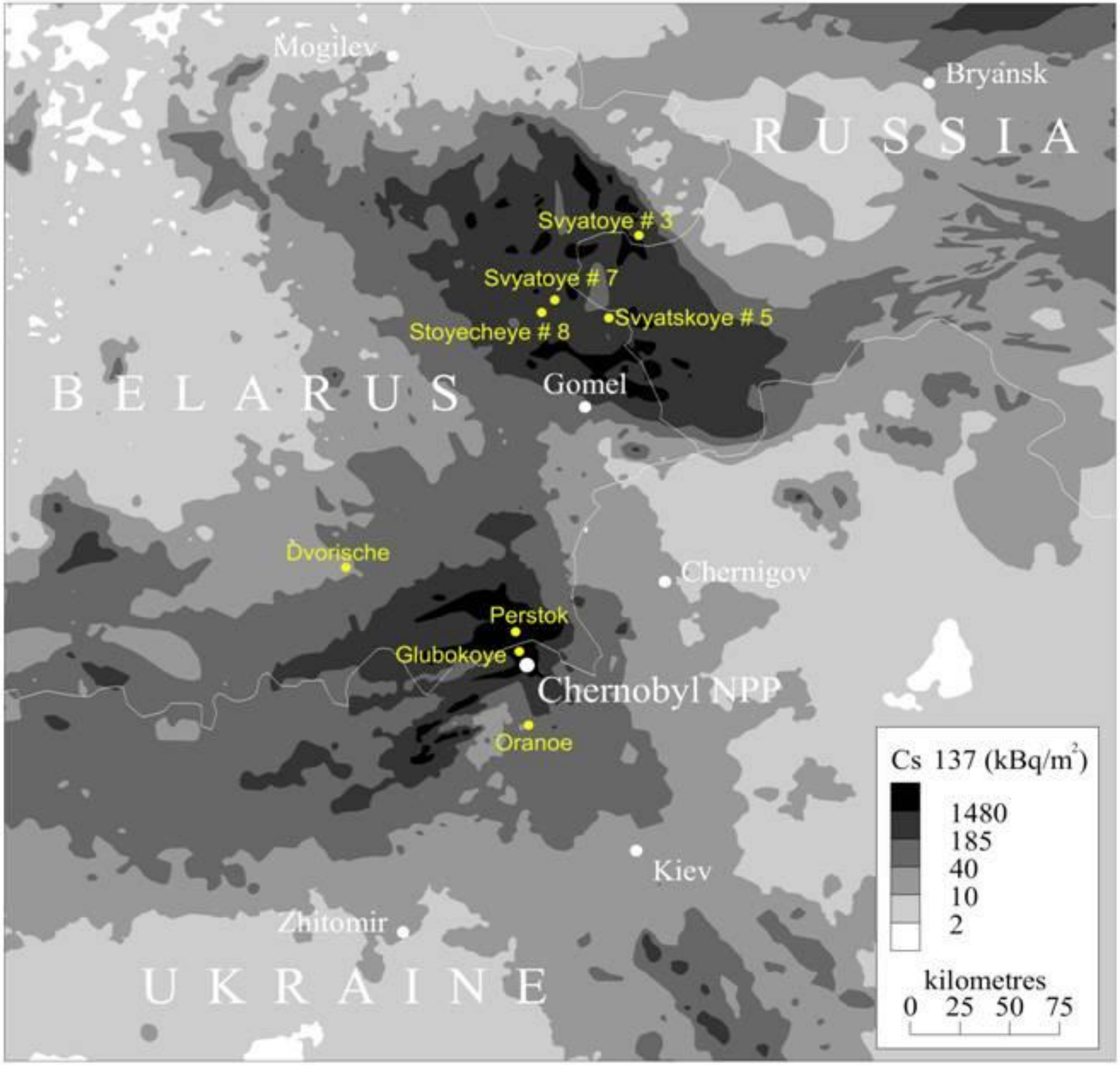
Lesson 5: We have to look at the detail of studies before we accept their conclusions.

Effects on aquatic systems

or..

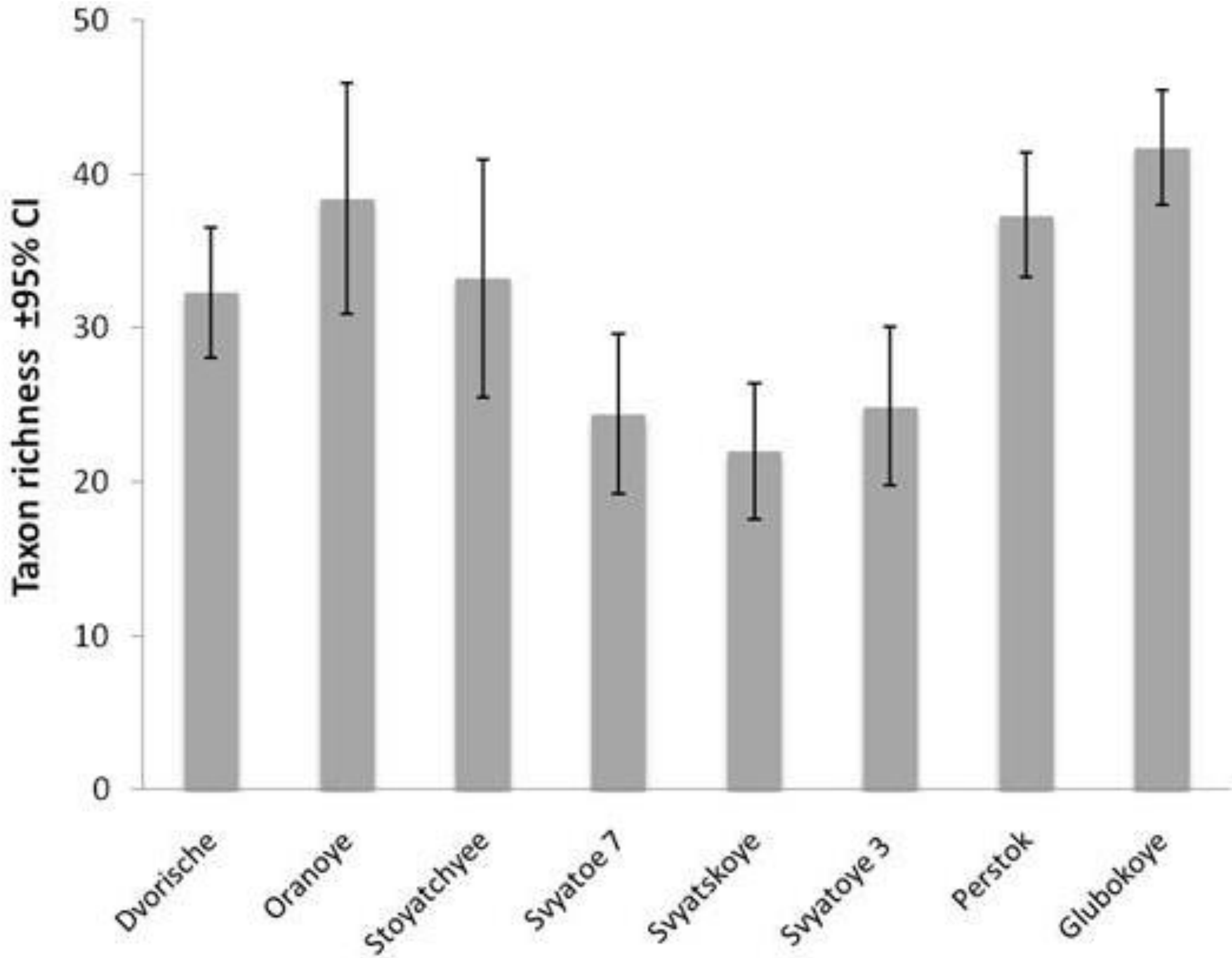
Does “Blinky” from *The Simpsons* live near Chernobyl?




Mogilev
Bryansk
RUSSIA
Svyatoye # 3
Svyatoye # 7
Svyatskoye # 5
Stoyecheye # 8
BELARUS
Gomel
Dvorische
Chernigov
Perstok
Glubokoye
Chernobyl NPP
Oranoe
Cs 137 (kBq/m^2)
1480
185
40
10
2
Kiev
Zhitomir
UKRAINE
kilometres
0 25 50 75

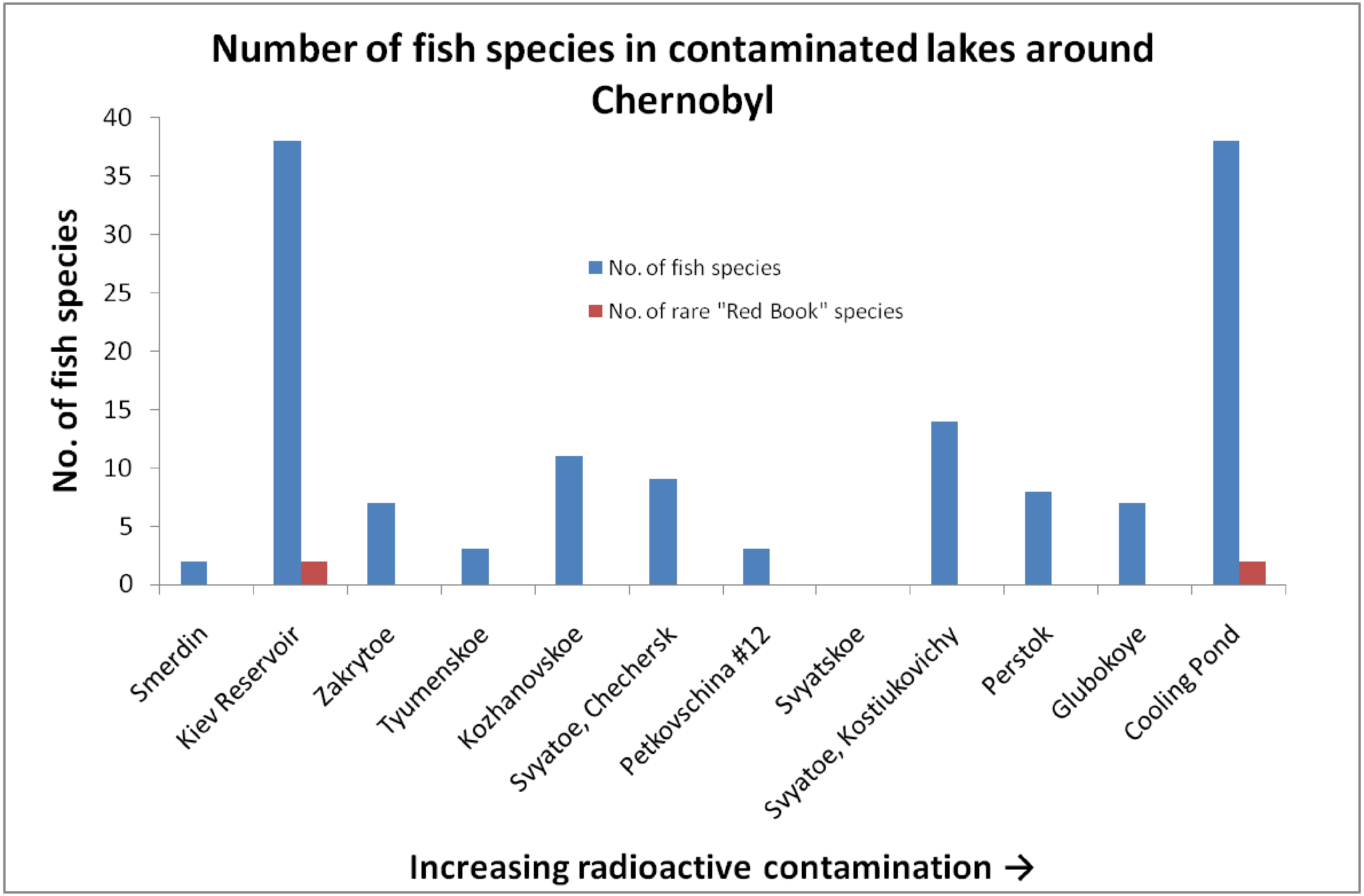
Biodiversity of insects in 8 lakes

- Lake area
- Lake depth
- Conductivity
- pH
- Total hardness
- Phosphate
- ^{137}Cs load

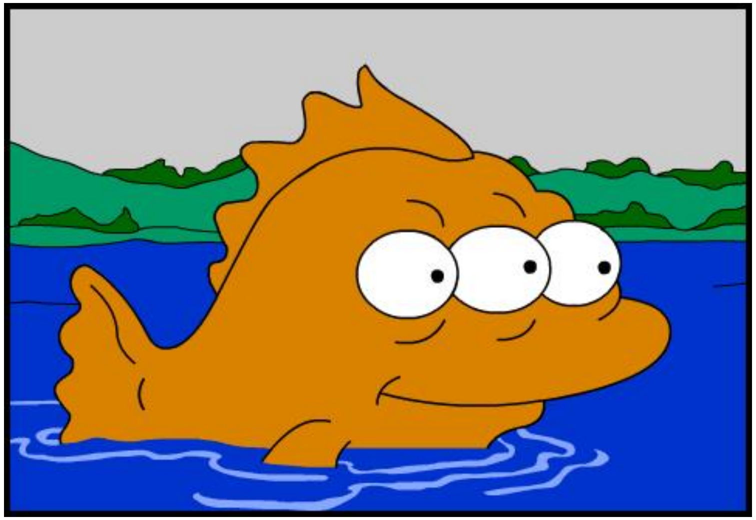


Increasing contamination →

Diversity of fish species



Catfish from the Chernobyl Cooling Pond



“Blinky” from Springfield Nuclear Power Plant Cooling Pond

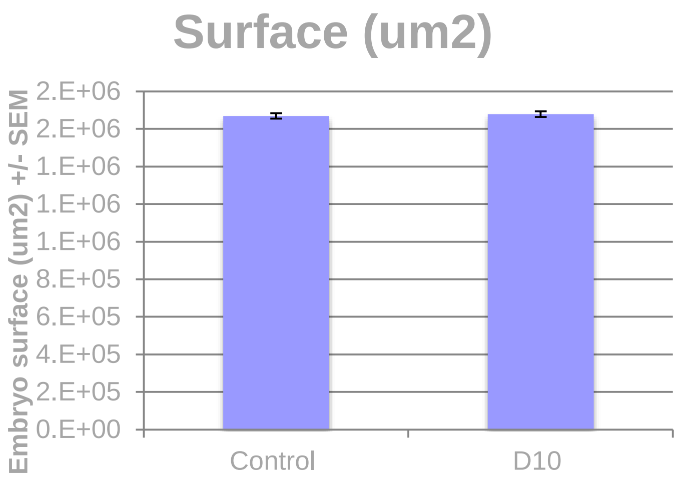
NERC Radioactivity and the Environment Programme

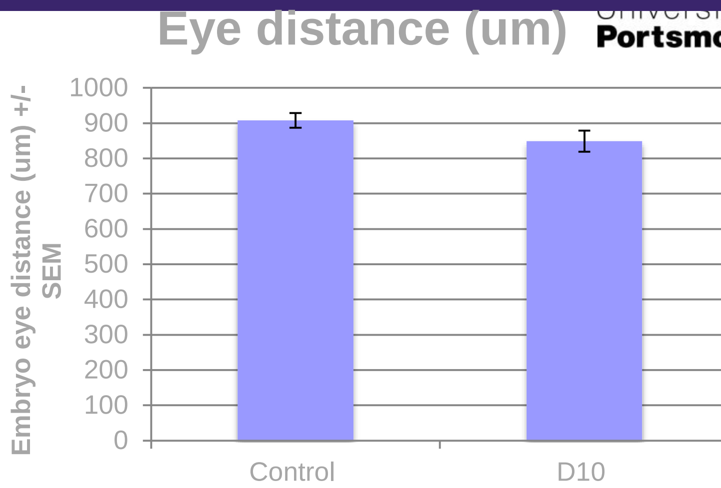
Transfer – Exposure – Effects: Integrating the science to underpin radioactivity assessments for humans and wildlife

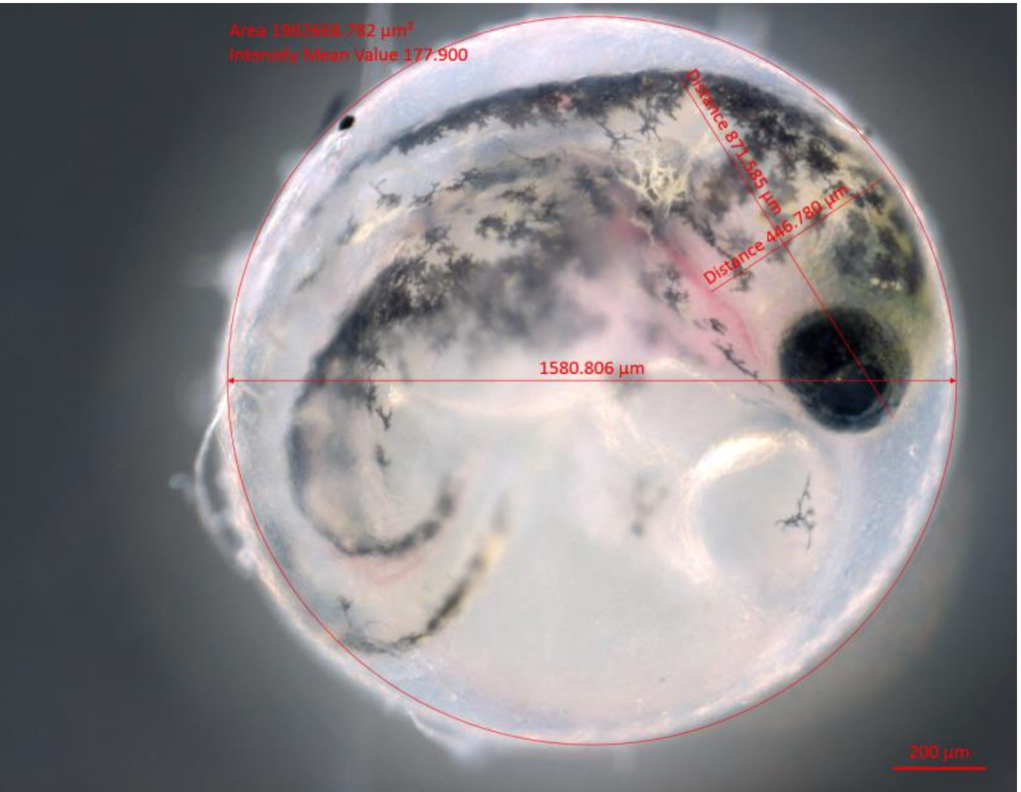
Field experiments

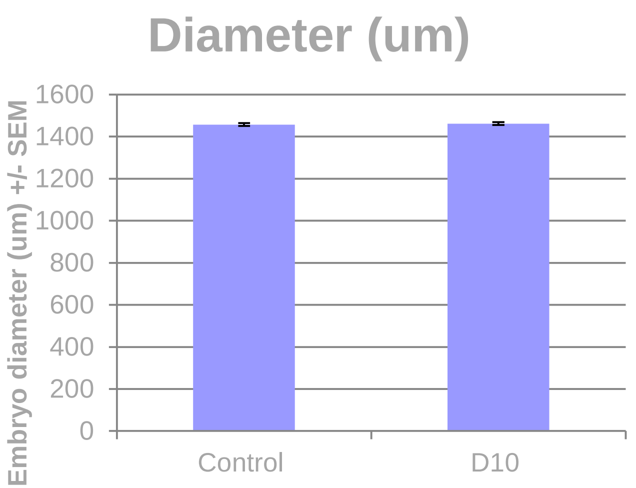
Laboratory experiments

University of Portsmouth





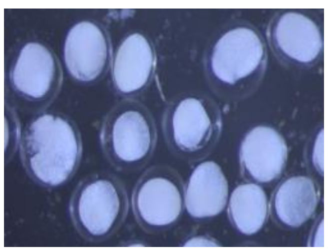




The Biological Effects of Ionising Radiation on Crustaceans: Combining Lab and Field Studies

- This project aims to investigate the effects of ionising radiation on aquatic invertebrates across **phenotypic and genetic endpoints**

- Both laboratory and field studies (**Chernobyl)** are in progress to determine the **effects of long term, environmentally relevant radiation doses on aquatic invertebrates**



e.g. **A comparison of fecundity** (number and weight of broods) **in *Asellus aquaticus* from lakes affected by the Chernobyl incident**

Risk and perception of risk

University of Portsmouth

WORLD VIEW

A personal take on events

UNIV. PORTSMOUTH

A long shadow over Fukushima

One impact of Japan's nuclear crisis is a dim but definite echo of Chernobyl, says **Jim Smith***: decades of caesium-137.*

Three weeks after the Fukushima accident, a clearer picture is beginning to emerge of possible long-term environmental consequences. The US Department of Energy (DOE) aerial survey of radiation doses was a crucial development. A clear trace reaching

quantities of contaminated waste. Consumers may refuse products grown in contaminated areas even when they meet regulations. Chernobyl has taught us that the social and psychological responses to radiation are of great, perhaps paramount, importance.

Nature, April 7th, 2011

- Long term evacuation and contamination of foodstuffs
- Potential social and psychological impacts

Social and psychological impacts of Chernobyl

“*The mental health impact of Chernobyl is the largest public health problem unleashed by the accident to date*”

UN/WHO/IAEA Chernobyl Forum Report, 2006

Lesson 6:

- Dose is not the most important health factor !
- Most important health factor is the sense of control and empowerment in people
- Effort in education and communication needs to equal clean-up effort.

Grigory Mamonin,
Forester