



# Modelling Exposures and Effects in the Marine Environment after the Fukushima Accident

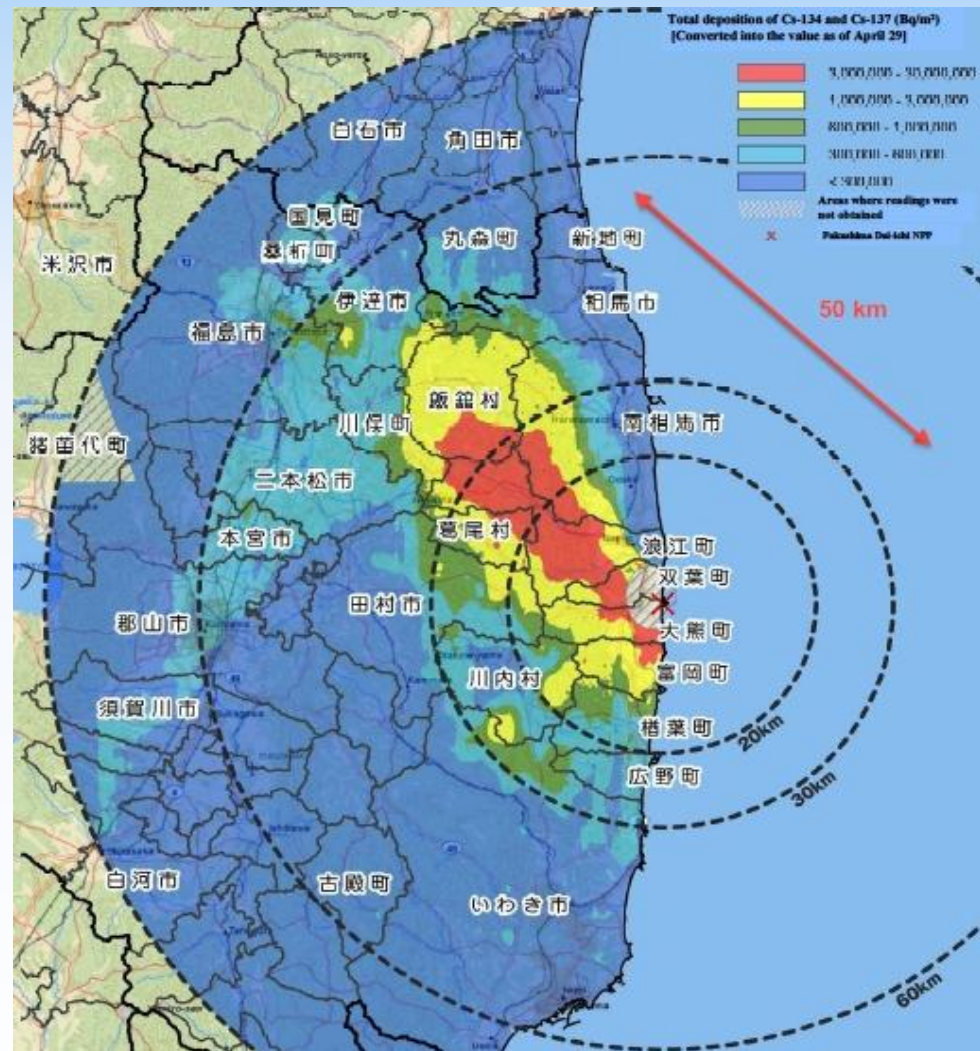
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# Introduction

# The March 2011 earthquake and tsunami

- Levels of radioisotopes 30 kilometres offshore from Fukushima > 10 x those in the Baltic and Black Seas during Chernobyl
- "When it comes to the ocean, Fukushima exceeds Chernobyl"



Pullen et al. (2013)

<http://journals.ametsoc.org/doi/pdf/10.1175/BAMS-D-11-00158.1>

# Introduction – setting the scene

- Largest ‘single’ accidental release of manmade radioactivity to the marine environment from civilian NPPs
- Main radionuclides :  $^{131}\text{I}$  and  $^{134,137}\text{Cs}$ 
  - smaller contribution of  $^{129,129\text{m},132}\text{Te}$ ,  $^{136}\text{Cs}$  and  $^{133}\text{I}$
- $\sim 10^{16}$  Bq of  $^{137}\text{Cs}$  discharged to sea,  $\sim 80\%$  between 11 March - 8 April 2011.
  - Reduction with distance by a factor of  $\sim 1000$  over 30-km
  - Short-lived isotopes disappeared by end of May 2011
  - Further land effluents till July 2011
- Contamination dispersing into the Pacific (winds, currents):
  - Still some delayed inputs from the coast
  - A fraction sinking to sediments e.g. attached to dead plankton
  - Cs and I retained by algae, fish, crustaceans, molluscs & plankton

# Initial estimates of impact on non-human biota

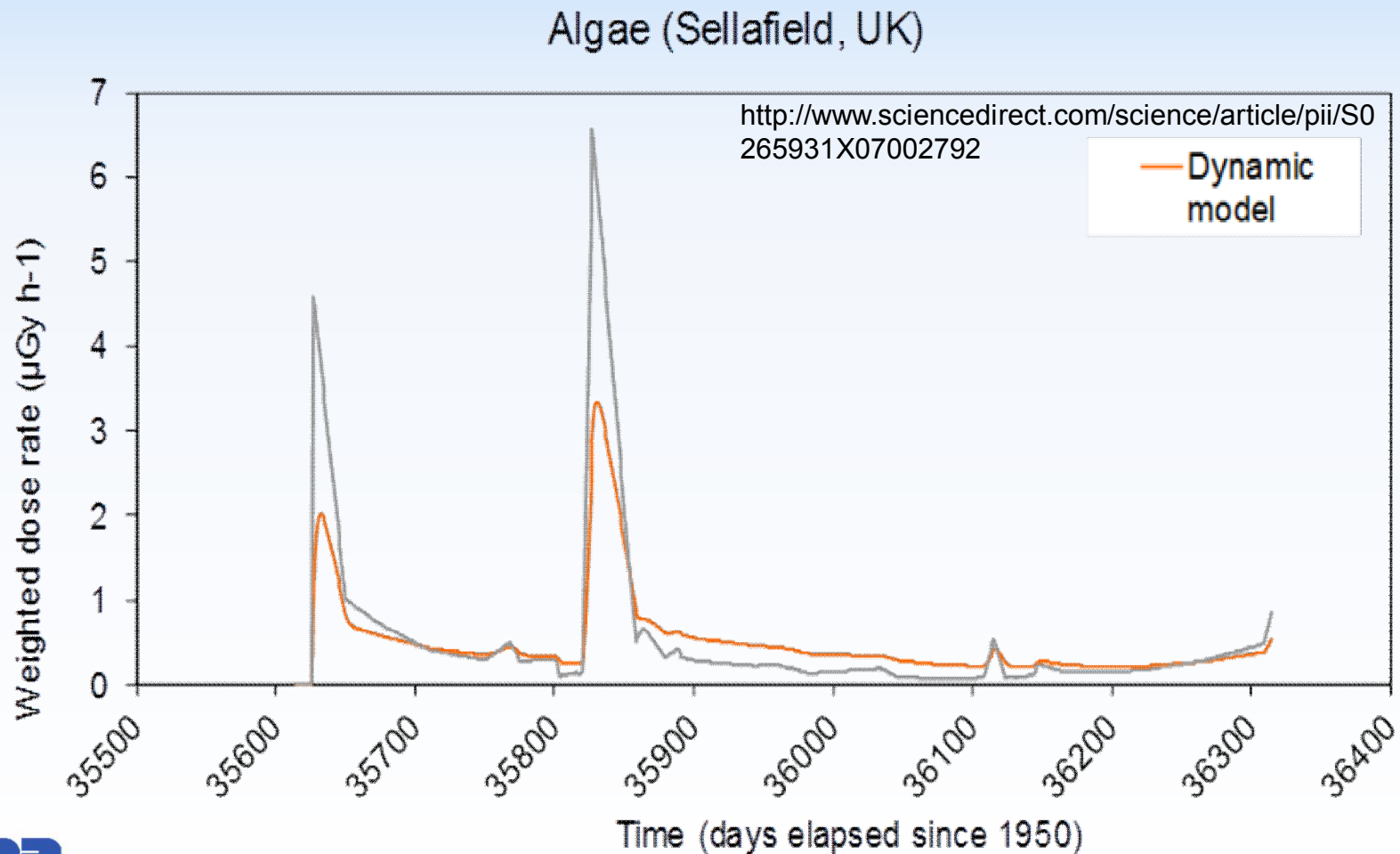
- Initial studies - maximum dose rates of 0.2 to 5 Gy d<sup>-1</sup> (first 3 weeks)
  - Assumes high levels remained constant over the period
- Such dose rates would exceed ERICA screening 'no effects' dose.
  - Possible mutagenic and reproduction effects in fish.
- Exposures reported were based on equilibrium
  - Activity in biota = activity in water x CF
  - Radioactivity was released as a pulse and equilibrium cannot be assumed
- Hypothesis for the early period after the accident:

Radioactivity levels in marine biota were below the maximum concentrations assumed by equilibrium models, because the turnover time of radionuclides is comparable to the discharge fluctuations. As a result, the doses received by the biota may have been overestimated

Possibly reverse trend for longer time periods after the accident

# Marine biota as radionuclide accumulators

- Early study, Sellafield pulsed  $^{99}\text{Tc}$  releases (late 1990's)



# Early accident phase

# Study with initially available data

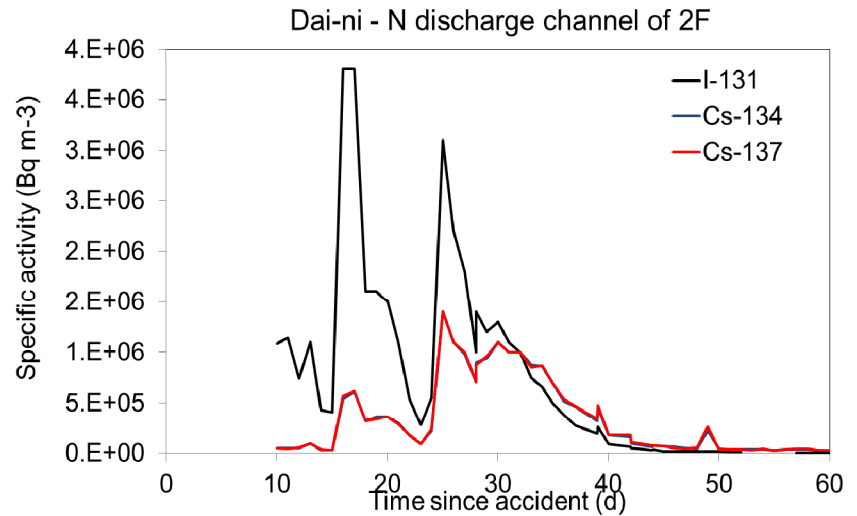
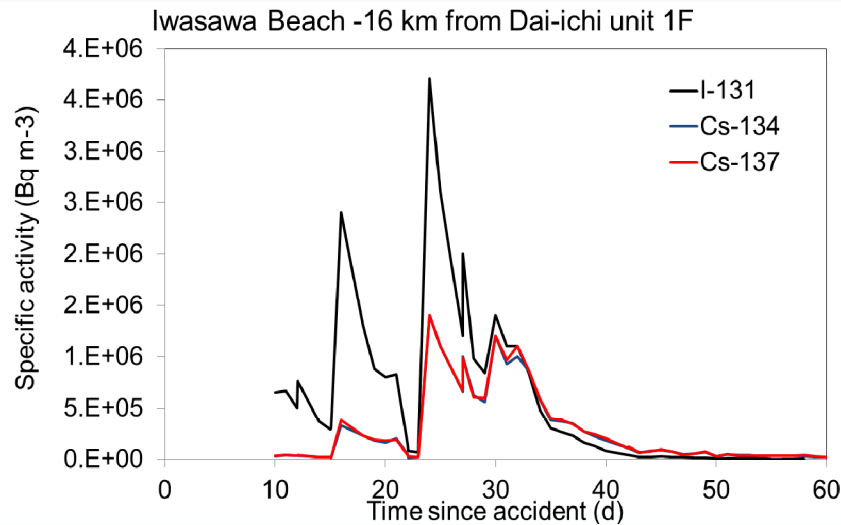
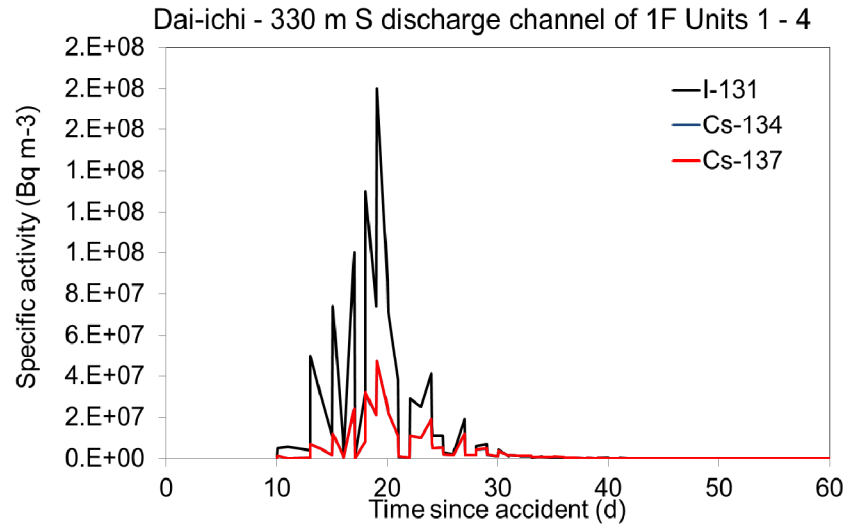
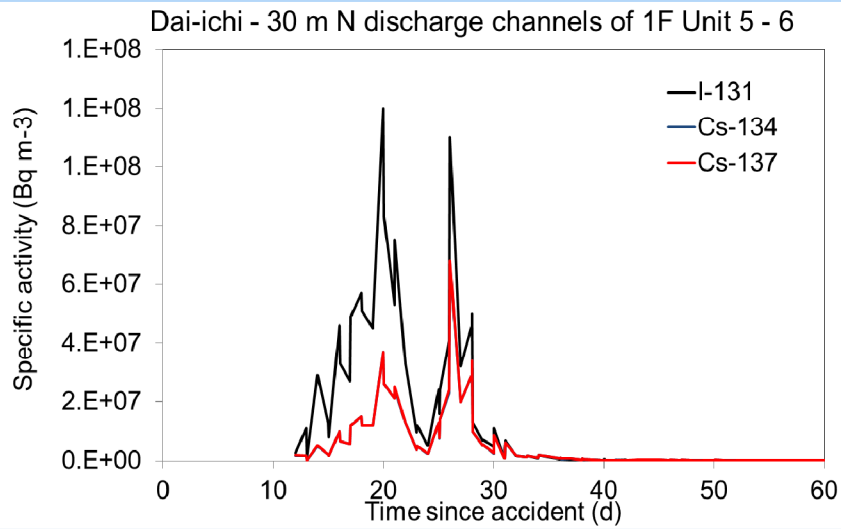
- Assessing radiation dose to biota at 4 coastal stations near FDNPS
- Use of publicly available data
- Concentrations of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in seawater, March – July 2011 (TEPCO)
  - Daiichi N and S channels
  - Iwasawa, 16 km S of Dai-ichi
  - Vicinity of Dai-ni discharge point
- Activity concentrations in sediment, April – July 2011 (MEXT)
- Activity concentrations in coastal fish, algae, molluscs - May & June 2011 (Greenpeace, analysed SCK•CEN)
- Comparison with dynamic transfer modelling (kinetic uptake)



*Greenpeace Fukushima Radiation Survey May 2011 (Lab 1). Accessed 12 September 2013.  
<http://www.greenpeace.org/international/Global/international/publications/nuclear/2011/Report%20SCK%20CEN.pdf>*



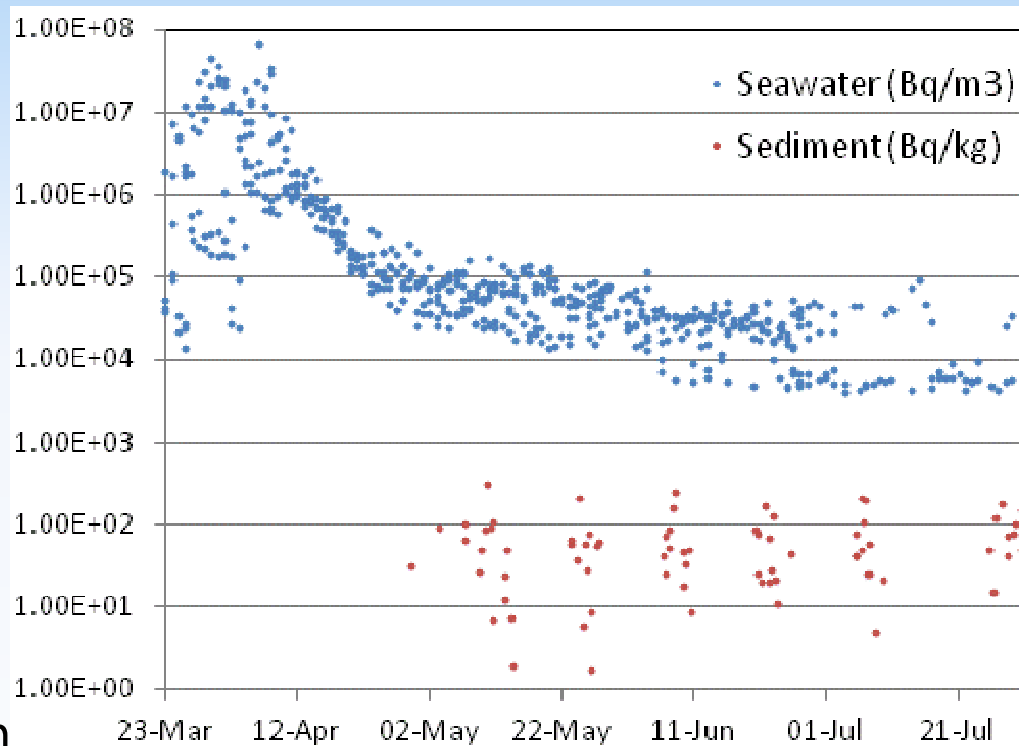
# A look at the data



Data from Buessler, K., M. Aoyama, and M. Fukasawa, *Impacts of the Fukushima Nuclear Power Plants on Marine Radioactivity*. Environmental Science & Technology, 2011. **45**: p. 9931–9935

# Monitoring data

- Seawater activity concentrations peaked at about 20 days
- Concentrations diminished 2 orders of magnitude in 60 days
- Not the same trend for sediment
- Indicates resilience of radionuclides in sediments - biogenic deposition, sorption
- $^{131}\text{I}$  in May 2011 samples – up to  $10^5 \text{ Bq kg}^{-1}$  in seaweed
  - This is due to the higher CF for  $^{131}\text{I}$
- $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in identical proportions
  - Mean 25% reduction between May and June



# Three methods of dose calculation

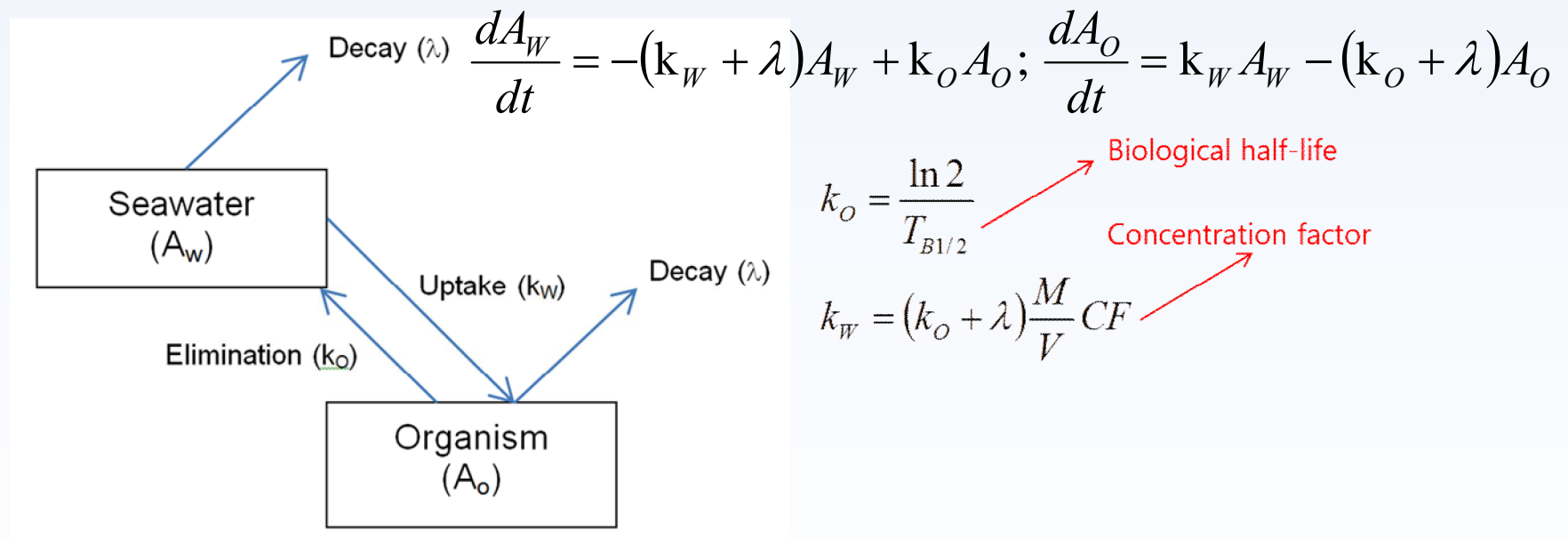
- From actual measurements data in organism, water and sediment

$$C_{\text{org}} \times \text{DPUC}_{\text{int}} + (f_{\text{wat}} \times C_{\text{wat}} \times \text{DPUC}_{\text{wat}} + f_{\text{sed}} \times C_{\text{sed}} \times \text{DPUC}_{\text{sed}})$$

- Using equilibrium model to calculate  $C_{\text{org}}$  from  $C_{\text{wat}}$

$$C_{\text{org}} = C_{\text{wat}} \times \text{CF} \text{ (the "concentration factor")} \rightarrow \text{valid only if } C_{\text{wat}} = \text{const.}$$

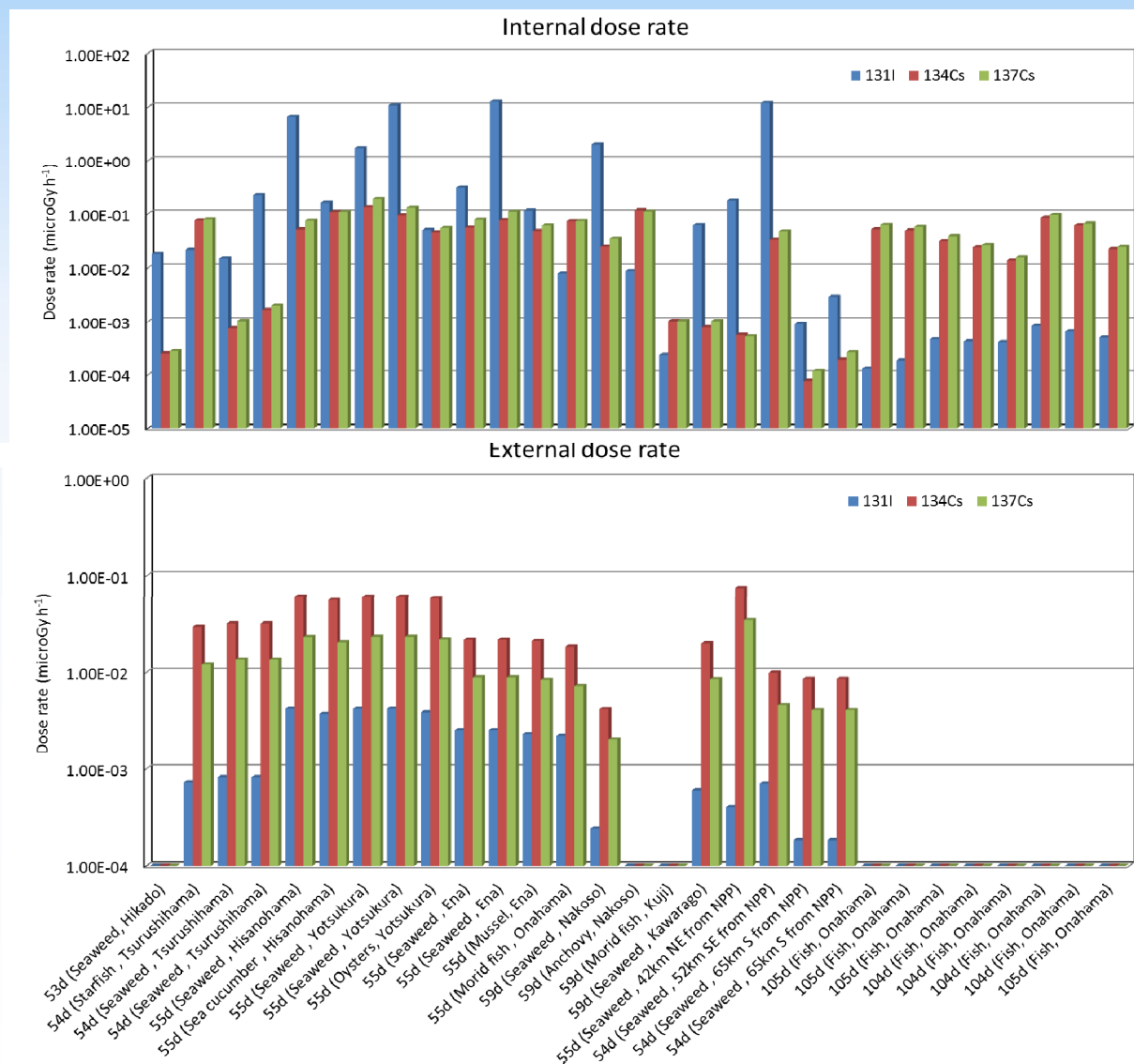
- Solve dynamic model  $\rightarrow$  best when  $C_{\text{wat}}$  is variable



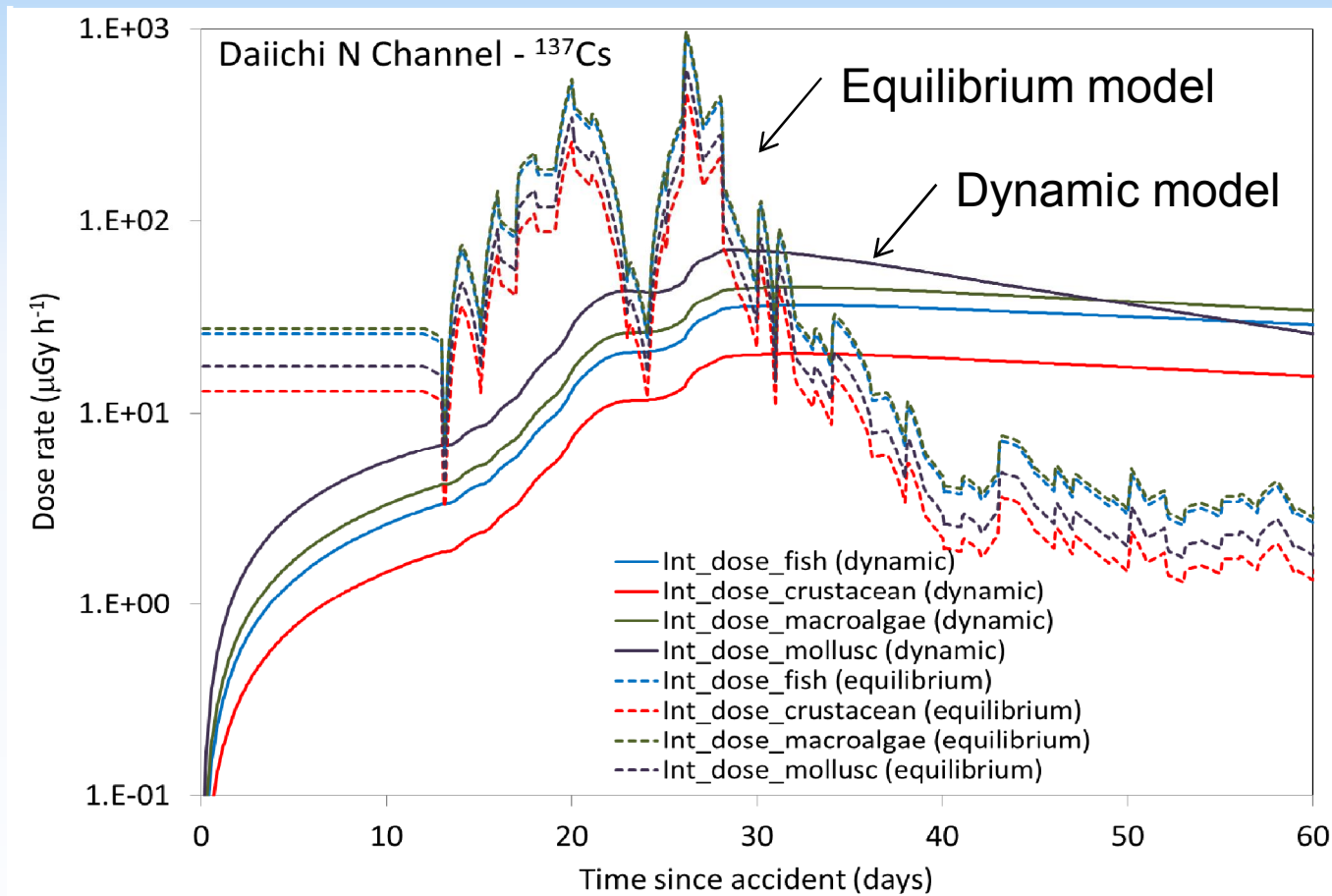
- Compare with  $10 \mu\text{Gy h}^{-1}$  screening value for NHB

# Doses from monitored biota

- Exposure in seaweed is dominant, followed by molluscs and fish
- Internal dose rates < 13  $\mu\text{Gy h}^{-1}$  of  $^{131}\text{I}$  and < 0.19  $\mu\text{Gy h}^{-1}$  of Cs (highest in seaweed, fish)
- External dose rates < 0.004  $\mu\text{Gy h}^{-1}$  of  $^{131}\text{I}$  and < 0.075  $\mu\text{Gy h}^{-1}$  of Cs (highest in seaweed, molluscs)
- Internal exposure higher than external (factor of 4 - 17,000 For  $^{131}\text{I}$  and 0.1 - 17 for Cs)
- Doses much lower than assuming equilibrium



# Doses from dynamic modelling study



- Importance of the dynamic modelling study: it gives results for the early phase of the accident, when biota monitoring data were not available

# Dynamic dose modelling results

- $^{131}\text{I}$  internal doses at FDNPS = 20 - 25 mGy h<sup>-1</sup> in macroalgae and 15 – 60 μGy h<sup>-1</sup> in other species (30 – 40 × lower in outer stations)
- $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  internal doses at FDNPS = 10 – 70 μGy h<sup>-1</sup> for all species (20 × lower in outer stations)
- Most exposed: macroalgae receiving  $^{131}\text{I}$  near the Daiichi outlets.
  - Highest 20 - 30 d post-accident, falling rapidly in weeks
- Organisms outside the FDNPS: < 4 μGy h<sup>-1</sup> (radiocaesium), 750 μGy h<sup>-1</sup> (radioiodine in algae)
- Internal dose rate dominates over external (2 – 3 orders of mag.)
- $^{131}\text{I}$  cumulative dose at FDNPS: 6.5 Gy for macroalgae (60 days)
  - 2 – 3 orders of magnitude lower for fish, crustaceans, molluscs
- $^{137}\text{I}$  cumulative dose at FDNPS: 20 - 50 mGy (maximum for molluscs)
- Cumul. doses out of FDNPS < 3 mGy ( $^{134,137}\text{Cs}$ ), 460 mGy ( $^{131}\text{I}$ )

# Comparison dynamic vs. equilibrium modelling

- Where concentrations in the water increase (30-40 d), dynamic model doses are lower than equilibrium (build-up phase)
- The trend reverses over the subsequent period (delayed retention phase)
- Differences most pronounced for the biota with elimination half-time of >10 d (fish and molluscs) - 2 – 3 orders of magnitude

Location	Mean total dose rate ( $\mu\text{Gy h}^{-1}$ )		Model prediction / measurement	
	Macroalgae	Mollusc	Macroalgae	Mollusc
<u>I-131 dose rates</u>				
Mean Iwasawa / Daini (equilibrium model)	5.15E+04	1.76E+02	8000	1600
Mean Iwasawa / Daini (dynamic model)	9.24E+00	2.25E-01	1.4	2.0
Mean monitoring data	6.38E+00	1.12E-01	-	-
<u>Cs-134 dose rates</u>				
Mean Iwasawa / Daini (equilibrium)	3.02E+03	1.66E+03	24000	15000
Mean Iwasawa / Daini (dynamic model)	1.66E+00	1.58E+00	13	14
Mean monitoring data	1.27E-01	1.12E-01	-	-
<u>Cs-137 dose rates</u>				
Mean Iwasawa / Daini (equilibrium model)	3.78E+03	2.08E+03	28000	23000
Mean Iwasawa / Daini (dynamic model)	2.23E+00	2.03E+00	17	22
Mean monitoring data	1.33E-01	9.12E-02	-	-

# The UNSCEAR assessment



# UNSCEAR – purpose and role

- UNSCEAR – United Nations Safety Committee on the Effects of Atom ic Radiation
  - UN Scientific Committee reporting to General Assembly. Assesses global levels and effects of ionizing radiation and provides scientific basis for radiation protection
- 60 international experts from 18 countries assessing for the United Nations the radiation exposures and health effects due to the accident at Fukushima.
  - Four expert groups: A (measurements), B (source term and dispersion), C (assessment of doses) and D (risk analysis)
  - Japan provided an extensive dataset to the Committee
- Interim report to the General Assembly (A67/46) issued in Sept. 2012
- Report of the Committee to the 68<sup>th</sup> session of the General Assembly will be issued on Friday
- More detailed report with full scientific annexes is due in ~ 2 months

# Key findings

- The highest exposures of wildlife appear to be associated with the marine environment
- Assessment performed with an extensive dataset of over 500 sediment, 6000 seawater and 5000 biota data points
- The report will confirm the main findings of our initial study
  - In general, the exposures to marine biota in other areas are too low for observable acute effects
  - Most exposed organisms are the macroalgae exposed initially to  $^{131}\text{I}$  at the FDNPS discharge zone
  - Any effects would be likely transient, given the short duration of the initial acute phase
  - This is based on current dose effects benchmarks

# The current situation

# The issue hits the news (again!)

Environment > Fukushima

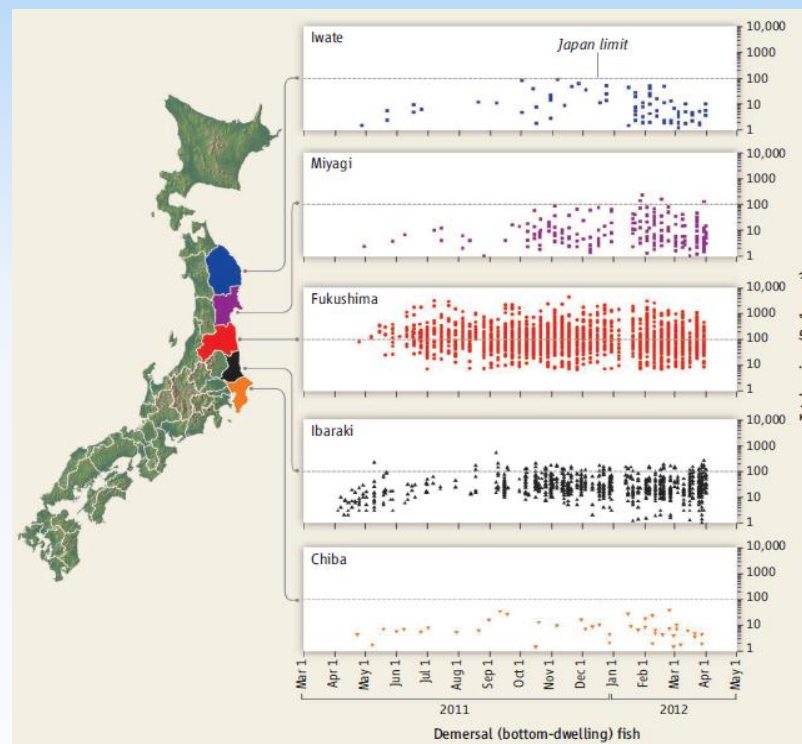
## Fukushima fish 'may be inedible for a decade'

Marine scientist finds levels of radioactivity in fish near stricken Fukushima nuclear plant are higher than expected

Fiona Harvey, environment correspondent  
The Guardian, Thursday 25 October 2012 19.00 BST  
Jump to comments (174)



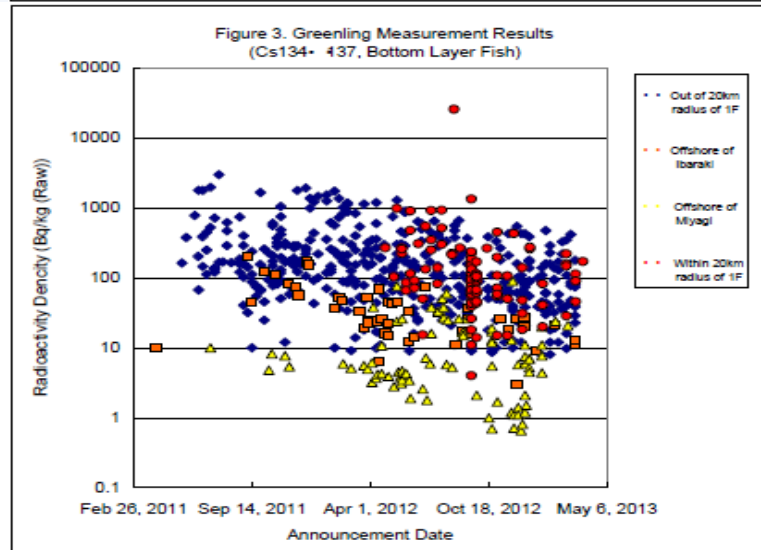
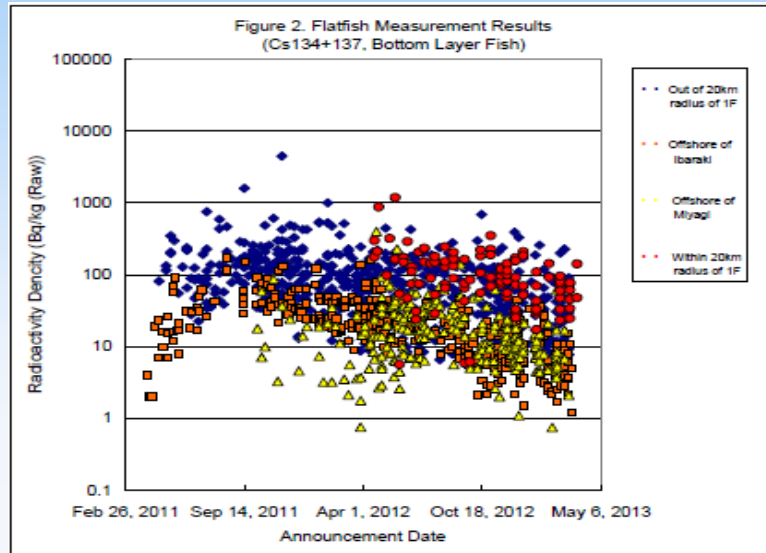
<http://www.who.edu/news-release/fukushima-fish>



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- Radionuclide levels in fish off Fukushima are highly variable but remain elevated, indicating delayed sources of radiation
- Levels up to  $10000 \text{ Bq kg}^{-1}$  Cs exceed  $100 \text{ Bq kg}^{-1}$  Japanese food limits
- For radioecology, the problem is very important, demanding serious study

# Current situation (biota)



- Report from TEPCO (2013) analysed for evidence of changes in biota activity concentration



<http://www.tepco.co.jp/en/nu/fukushima-np/f1/smp/2013/index03-e.html>

# Current situation (seawater and sediment)

- TEPCO report '*Result of Radioactive Nuclide Analysis around Fukushima Daiichi Nuclear Power Station*'
- Seawater contamination at unit 1F has decreased from  $10^5$  Bq L<sup>-1</sup> in March 2011 to ~ Bq L<sup>-1</sup> late 2012 (<sup>134</sup>Cs ~<sup>137</sup>Cs)
- Around 600 Bq kg<sup>-1</sup> total Cs in sediment (offshore Ukedo, Namie town, 1 km offshore – nearest to FDNPS)
- Sediment contamination has not changed significantly and remains a few hundred Bq/kg

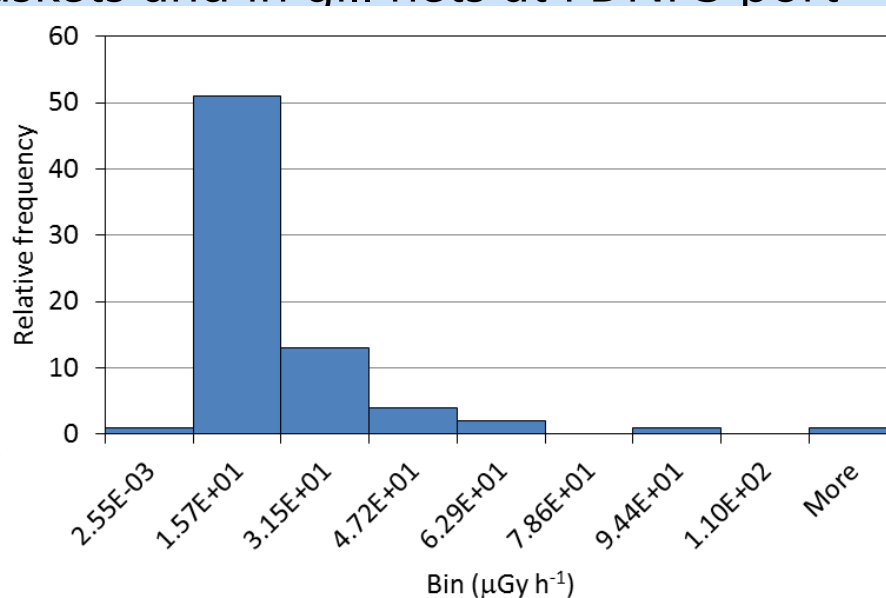
TEPCO, *Result of Radioactive Nuclide Analysis around Fukushima Daiichi Nuclear Power Station*. Tokyo Electric Power Company. Available from: <http://www.tepco.co.jp/en/nu/fukushima-np/f1/smp/2013/index03-e.html> [Accessed 12 September 2013]. 2013.

Station offshore Ukedo (town of Namie)			
Radiocaesium (Bq/kg dry sediment)			
Dist. From shore	1 km	2 km	3 km
Water depth	7 m	10 m	20 m
Apr-12	1,280	175	250
May-12	370	222	290
Jun-12	330	330	310
Jul-12	90	65	48
Aug-12	1,370	49	67
Sep-12	2,600	149	154
Oct-12	39	37	52
Nov-12	31	25	40
Dec-12	2,200	2,370	2,320
Jan-13	630	76	37

TEPCO fish/shellfish measurement stations 2013

# Updating the dose assessment

- Some of the highest levels of radiocaesium, as high as  $5 \times 10^5$  Bq kg<sup>-1</sup> f.w., in greenling from fishing baskets and in gill nets at FDNPS port entrance (late February – early March 2013)
- Highest estimates of internal dose rate (17/02/13, 21/02/13, 04/03/13):
  - 17 - 44  $\mu\text{Gy h}^{-1}$  for <sup>134</sup>Cs
  - 32 – 82  $\mu\text{Gy h}^{-1}$  for <sup>137</sup>Cs
  - However most doses below this maximum
- The data overall are log-normally distributed ( $\sigma = 0.59$ ,  $\mu = 0.32$ )
- The calculated median is  $e^\mu = 1.38$   $\mu\text{Gy h}^{-1}$
- 95% of results below 50  $\mu\text{Gy h}^{-1}$
- External dose rate estimate with limited data available: 0.12  $\mu\text{Gy h}^{-1}$  for <sup>134</sup>Cs and 0.05  $\mu\text{Gy h}^{-1}$  for <sup>137</sup>Cs  $\Rightarrow$  minor component of total dose



# Updating the dose assessment

- The highest doses seem to be still below the  $400 \mu\text{Gy h}^{-1}$  UNSCEAR benchmark for the most exposed individuals of an aquatic population below which population detriment is not expected
- From our histogram analysis, it is very unlikely that the elevated concentrations in individual specimens sporadically found close to the FDNPS signal prolonged exposures to whole fish populations
- However, the highest estimates are of the same order of magnitude than some  $\text{EDR}_{10}$  in some species and endpoints
- They are also exceeding the ERICA screening dose rate of  $10 \mu\text{Gy h}^{-1}$  indicating the need for continued assessment
- The biota are not under threat at the population level
- We are using a limited dataset based on TEPCO's report – need to do this calculation more comprehensively



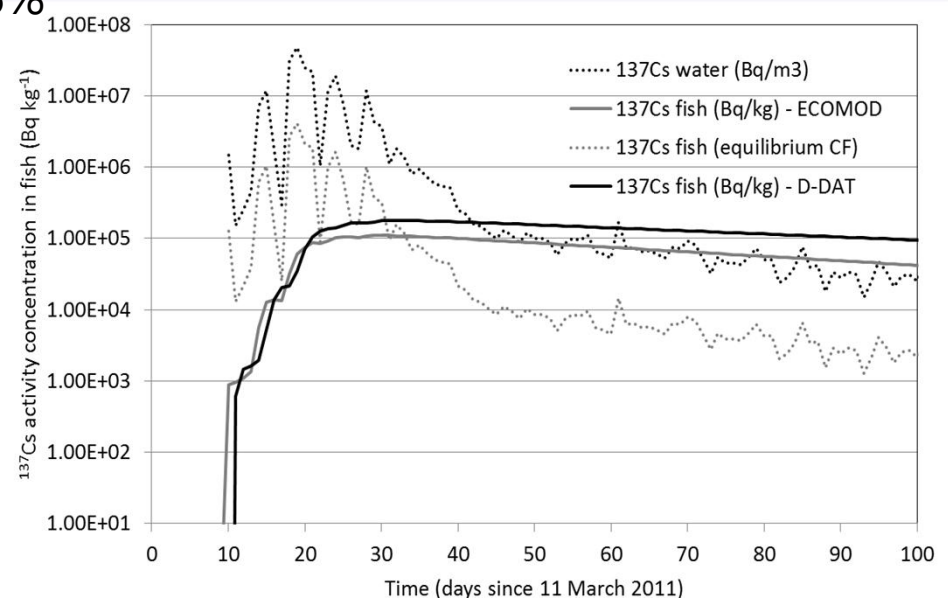
# Discussion

# Main findings

- The doses calculated in the various studies performed are generally below the amounts necessary to cause a measurable effect on populations
- The only exception is iodine in macroalgae close to the discharge point, limited to the earlier part of the accident
- Exposures for marine biota during the late phase fall below thresholds for which population effects are deemed likely
- Further away from the FDNPS, the potential for effects on biota will inevitably be even lower
- Elevated concentrations in individual specimens sporadically found close to the FDNPS in 2013 do not indicate prolonged exposures to whole fish populations (data lognormally distributed)
- However, these exposures are not “insignificant” or “negligible” because they reflect significant activity concentrations in the environment
- More systematic follow-up studies required to generate confidence

# Assessment uncertainties

- Based on previous intercomparisons of non-human dosimetry models, additional uncertainty relating to the dose conversion factors can be estimated as  $\pm 25\%$  for internal and  $\pm 120\%$  for external exposure
- To this one must add the effect on external dose rates, of estimating the activity in seawater and sediment from nearby points.
  - Assuming a 50% dispersion error in data within this radius of influence, the likely effect as not exceeding  $\pm 35\%$
- Consequently, the overall uncertainty of the assessment is likely  $\pm 60\%$  for internal and  $\pm 150\%$  for external dose
- The modelling-based assessment has an additional factor of  $\sim 2$  (based on intercomparison performed within UNSCEAR)



# Key study limitations

- It was not possible to account for some radionuclides present in the initial post-accident period and conspicuously absent from the monitoring data:
  - $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{129}\text{Te}$ ,  $^{129\text{m}}\text{Te}$ ,  $^{136}\text{Cs}$  or the actinides
- Likewise, it was not possible to include exposures from sediment when modelling external doses
  - However, sediments are slow accumulators of radioactivity so the effect on the acute accident phase would be limited
- The lack of match between seawater, sediment and sampling stations precludes an exact matching of internal and external dose
- The amount of data available from the scattered reports and papers publicly available is limited
- However, the broader UNSCEAR study is comprehensive, and will confirm the main findings of this presentation
- A publicly available, quality-assured, comprehensive dataset for free use by scientists worldwide would be a major benefit

# Limited effects data

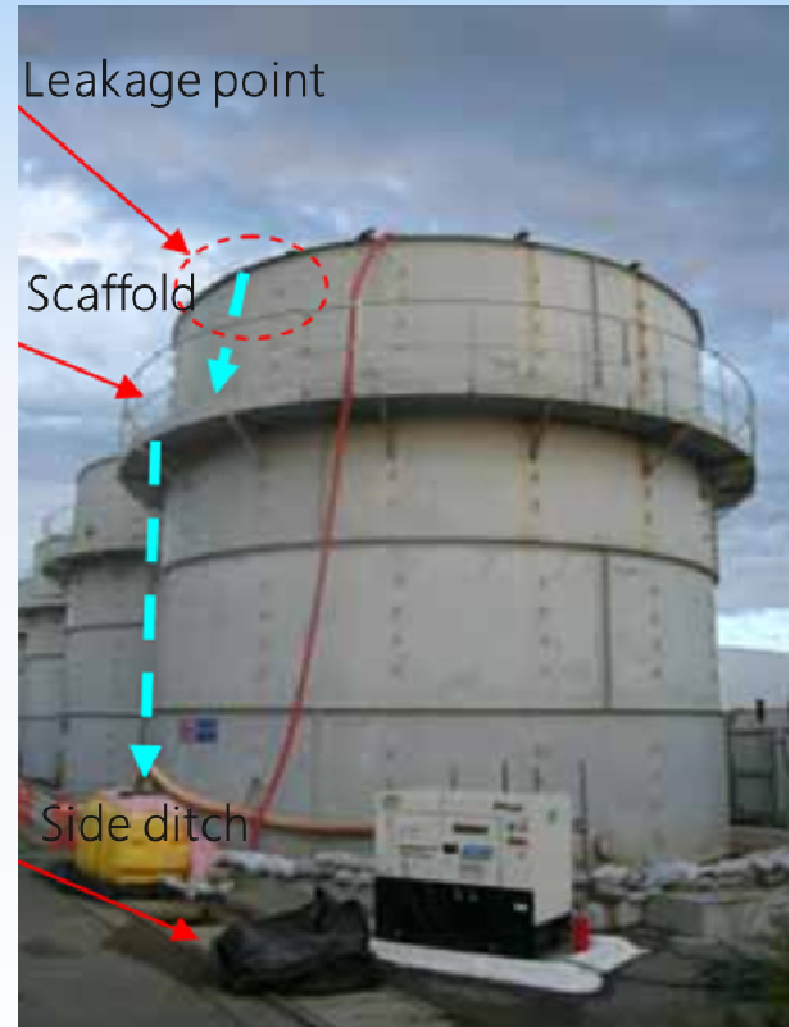
- Application to *average* exposed organisms in an accidental situation is novel and, in the case of the UNSCEAR 400  $\mu\text{Gy h}^{-1}$  benchmark, it is potentially open to scientific questioning (c.f. most exposed organisms)
- Although alterations to population integrity are deemed unlikely, more subtle effects at the individual level cannot be totally ruled out
- Long-term effects over several generations, for instance on reproduction, cannot yet be assessed
- For fish, limited data available on mortality effects indicate that dose rates  $< 4000 \mu\text{Gy h}^{-1}$  at any life stage are unlikely to affect survival
- Most importantly, the lowest value of the chronic dose rate giving 10% effect in reproductive endpoints is equal to 47  $\mu\text{Gy/h}$  for a marine species *Pleuronectes platessa* [Garnier-Laplace et al., JRP 2010]
- For marine invertebrates, the lowest value of EDR10 is found at 36  $\mu\text{Gy/h}$  for annelids [paper Knowles and Greenwood, 1994]
- For marine plants, there are no chronic effect data in the literature to our knowledge

# Limitations of effects predictions

- There is a need to characterise the local hotspot locations and understand the resilience of radioactivity in most exposed biota
  - Attempt to observe any potential effects at these locations
- Our understanding of the biological impacts of radiation on chronically exposed plants and animals is at present based largely upon limited high-exposure data collated under controlled laboratory conditions
- Biota have wide range of inter-species radiosensitivity
- They may react according to a complex dynamic of interactions between absorbed doses (or dose rates) and radiotoxic responses, expressed at different levels of biological and ecological organization
- So far there are no reported observations of such effects in the Fukushima marine environment
- That does not prove that effects on biota did (or will) not happen

# Future issues

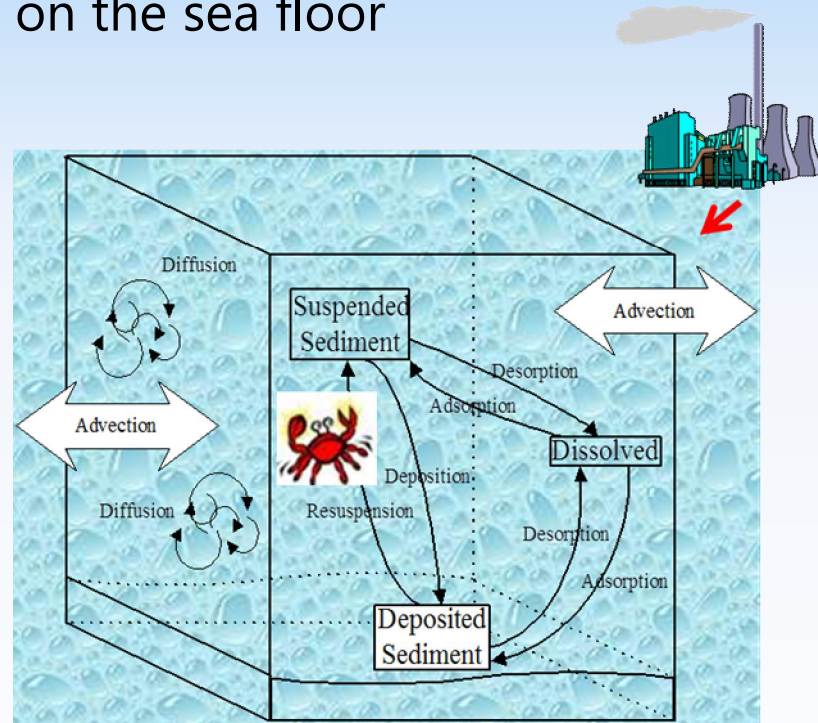
- Recent news of release from storage tank (accident level raised 1 to 3)
- Expect to see leakage to sea via groundwater (weeks to months)
  - Need to study the local hydrology and model groundwater flow
- Expect local spots of higher concentration to persist for significant time
- Biota in some sites will be more exposed than in others, following unexpected patterns
- More fish specimens occasionally found with higher levels than the average
- What about the actinides?



[http://www.tepco.co.jp/en/nu/fukushima-np/handouts/2013/images/handouts\\_131003\\_01-e.pdf](http://www.tepco.co.jp/en/nu/fukushima-np/handouts/2013/images/handouts_131003_01-e.pdf)

# Future issues

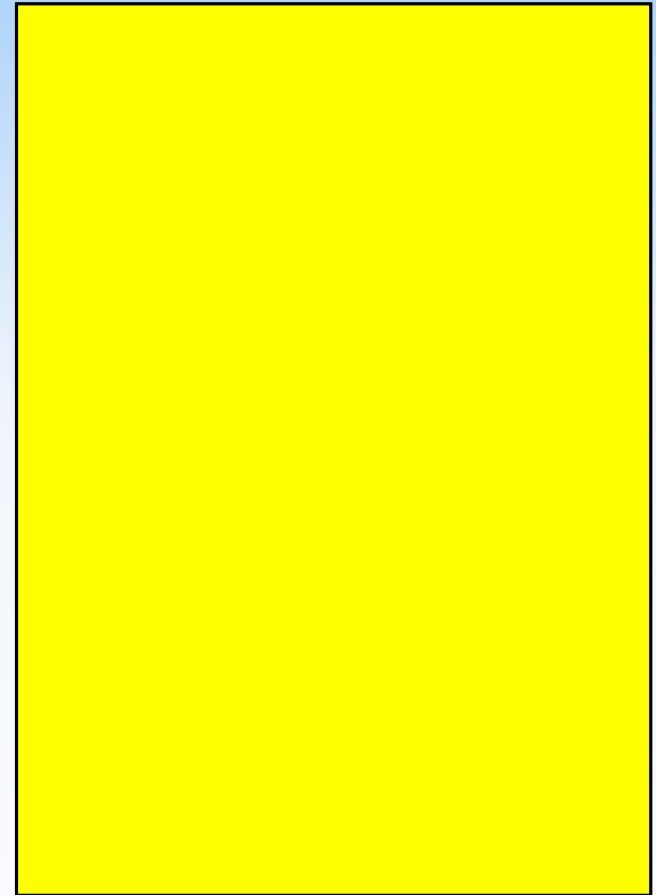
- Meanwhile, there will be continued dispersion through water
- Continued settling of radionuclides on the sea floor
  - Scavenging by biogenic particles
  - Sorption
- Investigations must continue
  - Need to conduct detailed fieldwork (research cruises, observatory sites)
  - Need reliable model predictions
  - Need to conduct long-term effects studies
  - Revisit current assessments in future
  - Assess effects at population dynamics level
- Fukushima is and will remain the main problem for marine Radioecology in the next 20 years





# The abiotic environment

- The impact to the overall environment (including the abiotic component) was beyond the scope of these studies
- We acknowledge that a dose-based assessment implies a biocentric paradigm
- Radiation dose cannot capture impacts to the actual value that mankind assigns to the environment
- "Small" doses  $\sim 2 \mu\text{Gy h}^{-1} \Leftrightarrow$  large  $^{137}\text{Cs}$  activity  $10^4 \text{ Bq kg}^{-1}$  in fish  $\Leftrightarrow 10^5 \text{ Bq m}^{-3}$  in water and  $5 \times 10^5 \text{ Bq kg}^{-1}$  in sediment (w. ERICA CF and  $K_d$ s)
- By comparison,  $3 \times 10^3 \text{ Bq m}^{-3}$  off Sellafield at peak period (late 1970's)
- The environment has a value publicly perceived as being affected
- Hence, qualifiers such as "negligible" and "insignificant" are not useful
- Applicability of RP framework to the abiotic environment under debate



Thank you very much for  
your attention

شكرا لحسن استماعكم

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