Dynamic modelling of the radiological impact of the Fukushima accident on marine biota

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The March 11, 2011 earthquake/tsunami

- Levels of radioisotopes 30 kilometres offshore from Fukushima > 10 times those measured in the Baltic and Black Seas during Chernobyl
- "When it comes to the oceans, says Ken Buesseler, a top chemical oceanographer at the Woods Hole Oceanographic Institution, "the impact of Fukushima exceeds Chernobyl"
INTRODUCTION
Introduction – setting the scene

- Large accidental release of manmade radioactivity to the marine environment.
- Main radionuclides: $^{131}$I and $^{134,137}$Cs
  - smaller contribution of $^{129,129m,132}$Te, $^{136}$Cs and $^{133}$I
- $\sim 10^{16}$ Bq of $^{137}$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 gradually dispersing into the Pacific (winds, currents):
  - Still some delayed inputs from the coast
  - A fraction sinking to sediments attached to dead plankton
  - Cs and I retained by macroalgae, fish, crustaceans, molluscs & plankton
Introduction – impact on non-human biota

- Initial studies - maximum dose rates of 0.2 to 5 Gy d⁻¹ (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.
DESCRIPTION OF APPROACH
Data sources

- Activity concentrations of $^{131}$I, $^{134}$Cs and $^{137}$Cs in seawater, 23 March – 30 July 2011 (TEPCO)
  - Daiichi N and S channels
  - Iwasawa Beach 16 km S of Daiichi discharge
  - Vicinity of Daini discharge point
- Activity concentrations in sediment -29 April – 31 July 2011 (MEXT)
- Additionally, activity concentrations in fish, algae, molluscs - early May and late June 2011 at coastal stations (Greenpeace, analysed at SCK•CEN)
A look at the data
Monitoring data

- Seawater activity peaked at ~20 days and rapidly diminished - 2 orders of magnitude in 60 days
- However, there is no obvious trend for sediment activity, with concentrations sustained over time!
- Indicates the resilience of radioactivity in sediments - biogenic deposition, sorption
- The isotopic ratio $^{134}\text{Cs} / ^{137}\text{Cs}$ is fairly constant at $0.97 \pm 0.14$ for seawater and $0.81 \pm 0.05$ for sediment – dominance of fallout $^{137}\text{Cs}$ at lower concentrations?
- $^{131}\text{I}$ in May samples - up to $10^5$ Bq kg$^{-1}$ in seaweed due to the higher CF for $^{131}\text{I}$
- $^{134}\text{Cs}$ and $^{137}\text{Cs}$ are also present in almost identical proportions, with a mean 25% reduction between May and June

http://www.whoi.edu/website/fukushima-symposium/presentation-files
Dose calculations from actual monitoring data

- Based on the ERICA non-human biota dosimetry approach but our model is fully dynamic (time-dependent dose rate calculations)
- Assigned an ERICA marine reference organism to each biota
- Multiply the activity concentrations in biota were multiplied by a dose per unit concentration (DPUC) for internal exposure - template ERICA run
- Locate the nearest seawater and sediment sampling points to a given biota sampling point using an algorithm
- Average the seawater/sediment data within a set distance (20 km) and time (2 days for seawater, 10 for sediment) from each individual biota
- Multiply the activity concentrations in water/sediment by relevant DPUC for external exposure - template ERICA run
- Add internal and external dose
- Compare with ERICA benchmark value of 10 $\mu$Gy h$^{-1}$ screening value for NHB
Solve model to obtain activity concentrations over time, based on $T_{B1/2}$

- Convert concentrations to time-dependent dose rates using ERICA dose conversion factors, occupancy and radiation weighting factors
- Integrate dose rate curve to give cumulative dose over a 60-day period
- This approach is therefore valid for acute or time-variable exposures
RESULTS AND DISCUSSION
- Exposure in seaweed is dominant, followed by molluscs and fish
- Internal dose rates < $13 \mu\text{Gy h}^{-1}$ of $^{131}\text{I}$ and < $0.12 \mu\text{Gy h}^{-1}$ of Cs (highest in fish)
- Cs doses < $10 \mu\text{Gy h}^{-1}$, $^{131}\text{I}$ marginally above – no likely harm to species
- Internal exposure dominates (factor of 4 - 17,000 for $^{131}$I and 0.1 – 17 for Cs)
- Clearly, the equilibrium assumption does not reflect reality!
Doses from dynamic modelling study (example)
Dynamic dose modelling results

- For $^{131}$I, maximum modelled dose rates at Daiichi channels = 20 - 25 mGy h$^{-1}$ in macroalgae and 14 – 60 μGy h$^{-1}$ in other species (30 – 40 x lower in outer stations)
- For $^{134,137}$Cs, 20 – 60 μGy h$^{-1}$ for all species (20 x lower in outer stations).
- Most exposed organisms: macroalgae receiving $^{131}$I near the Daiichi outlets.
  - Highest doses 20 - 30 d post-accident, but falling rapidly in subsequent weeks
- Accumulated $^{131}$I dose for macroalgae over the first two months is 0.27 Gy.
- For other species, cumulative $^{131}$I doses 2 – 3 orders of magnitude lower
- Away from the Daiini drainage channels, a further order of magnitude below
- For most organisms outside the most immediate area dose rates < 20 μGy h$^{-1}$, with cumulative doses of < 0.1 mGy.
- Internal dose dominates over external from water: 2 – 3 orders of magnitude
Comparison dynamic vs. equilibrium

- Where concentrations in the water increased (30-40 d), dynamic model doses are lower than for an equilibrium model - delayed build-up.
- The trend reverses over the subsequent period – delayed retention.
- Differences most pronounced for the biota with $T_{B1/2}$'s of >10 d (e.g. fish and molluscs for $^{131}$I and all species for $^{137}$Cs) - 2 – 3 orders of magnitude.
- The dynamic model gives drastically reduced doses at peak discharges
- Comparison with actual doses from nearest biota stations (< 50 km):

<table>
<thead>
<tr>
<th>Location</th>
<th>Mean total dose rate (μGy h$^{-1}$)</th>
<th>Model prediction / measurement</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Macroalgae</td>
<td>Mollusc</td>
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<td>I-131 dose rates</td>
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<td>Mean Iwasawa / Daini (equilibrium model)</td>
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<td>Mean monitoring data</td>
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<td>Cs-134 dose rates</td>
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<td>Cs-137 dose rates</td>
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<td>Mean monitoring data</td>
<td>1.33E-01</td>
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CONCLUSIONS
Conclusions

- <3% of the discharged Cs remains in coastal waters - all $^{131}$I decayed
- There is persistent radioactivity adsorbed onto the sediment
- Radioactivity entering the marine biota
- Exposure in seaweed predominates; then molluscs and fish
- Biota doses in the first weeks post-accident initially overestimated due to assuming equilibrium between the water and the organisms
- Initial estimates reduced by 3 orders of magnitude, illustrating the importance of using dynamic modelling
- But for the earlier part of the accident, doses are all below the ERICA 10 $\mu$Gy h$^{-1}$ screening level (except perhaps at a few hot spots)
- The only significant exposures would have been to $^{131}$I in seaweed in the earlier part of the accident, confirmed by our dynamic modelling study
Radionuclide levels in fish off Fukushima are highly variable but remain elevated, indicating a delayed source of radiation => demands further investigation

We are in the period where doses will be approach equilibrium from above

Levels up to 10000 Bq kg\(^{-1}\) Cs give < 20% of 10 \(\mu\)Gy h\(^{-1}\)

Environment not compromised, but long-term follow-up is scientifically advisable