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Environmental Marine Radioactivity after Fukushima

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The Great East Japan earthquake of magnitude 9.0, the world's fourth largest since 1900 and the largest in Japan since modern instrumental recordings began 130 years ago (USGS, 2011; Koketsu *et al*., 2011), generated the 11 March 2011 Tohoku tsunami. Unexpectedly high waves of 15 m offshore Fukushima, about twice higher than expected, hit the Fukushima Daiichi Nuclear Power Plant (NPP) resulting in total damage of the electrical network (TEPCO, 2011a). Four generators lost their ability to produce electrical power to cool the nuclear reactors 1, 2, and 3 that were in operation, as well as the fuel rod storage pool for reactor number 4. The uncooled fuel rods overheated and subsequently produced hydrogen gas from the water-metal reaction. This resulted in hydrogen explosions of the reactor buildings 1, 3 and 4 that heavily damaged the Fukushima Daiichi Nuclear Power Station, which included a subsequent meltdown of fuel in the pressure vessels of reactors 1, 2 and 3. Radioactivity was released to air and water and resulted in a major evacuation of the surrounding area. Furthermore, liquid discharges to the Pacific Ocean resulted in continuous contamination of the marine environment and the prevailing ocean currents transported radionuclides into the Pacific basin. Recently, contaminated groundwater around the Fukushima Daiichi NPP has become a major concern because this leads to a large uncontrolled and chronic submarine source of radionuclides to coastal waters.

Radionuclide releases during the Fukushima Daiichi accident

The hydrogen explosions and heavy destruction of the reactors resulted in an initial large releases of radionuclides to the atmosphere (e.g. about 150 PBq of ^{131}I, and 15 PBq of each ^{134}Cs and ^{137}Cs)[1] (TEPCO, 2011b). The hot nuclear reactors required permanent cooling by water and partly by seawater resulting in highly contaminated water, which during the first months was also directly released to coastal waters (e.g. about 4 PBq for each ^{134}Cs and ^{137}Cs) (Estournel *et al*., 2012; Aoyama *et al*., 2013; Povinec *et al*., 2013a). The necessity for continuous cooling of the reactors causes high contamination of groundwater, which cannot be completely stored on the site and consequently some finds its way to the shore by groundwater discharges. The operator TEPCO installed procedures to remove radio-cesium from the cooling water but the system failed partly during operations.

The deposition from the atmosphere to the North-western Pacific Ocean shortly after the accident was estimated to be 12-15 PBq for each ^{134}Cs and ^{137}Cs (Aoyama *et al*., 2013). The second important long-lived radionuclide after ^{137}Cs released directly to the ocean was ^{90}Sr, which had an estimated total activity of about 1 PBq (Povinec *et al*., 2013a). The ^{134}Cs has a half-life of 2.05 years, ^{137}Cs of 30 years and ^{90}Sr of 29 years. ^{131}I has only a short half-life of 8 days and was no longer detectable after several months.

[1] PBq: petabecquerel = 10^{15} Bq

However, ^{131}I is responsible for the major part of the dose impact to the population due to its enrichment in the thyroid gland.

The Fukushima accident has been declared by the Japanese Government to be of class 7 on the INES scale[2], the same and highest grade as the Chernobyl accident, although the radionuclide releases during the Chernobyl accident were by about a factor of 10 higher (e.g. 1760 PBq of ^{131}I and 85 PBq of ^{137}Cs) (IAEA, 2006). The largest radionuclide releases to the atmosphere have until now been due to atmospheric nuclear weapons tests (also called global fallout), carried out mainly during fifties and early sixties (e.g. 950 PBq of ^{137}Cs and 600 PBq of ^{90}Sr) (Povinec *et al*., 2013a).

Radionuclides in the atmosphere outside Japan

The radionuclide levels observed in the atmosphere at Fukushima varied considerably. Particulate ^{131}I varied between 1 and 10,000 Bq/m^3, and both ^{137}Cs and ^{134}Cs in aerosols ranged from 0.1 to 200 Bq/m^3. Surface dose rates varied between 0.04 µSv/h (initially) to a maximum value of 10 µSv/h (mid-March 2011), and then decreased to 0.3 µSv/h by the end of April 2011 (Povinec *et al*., 2013a).

The prevailing westerly winds, initially transported atmospheric radioactive contamination from Fukushima over the Pacific Ocean, then over North America, the North Atlantic Ocean to Europe, and then back to Asia (figure 1). Local deposition was studied by numerous authors and Japanese authorities (e.g. Yoshida & Kanda, 2012). The first radionuclide signals outside Japan were registered by the CTBTO (Comprehensive Test-Ban Treaty Organization) stations in the Hawaiian Islands. Before the cloud was also detected in Europe, the radioactive cloud was registered in Iceland. Over Europe, particulate ^{113}I varied between 0.01 and 6 mBq/m^3 (up to 11 mBq/m^3 for the gaseous form), and ^{134}Cs and ^{137}Cs varied equally between 0.001 mBq/m^3 at the end of May 2011 to about 1.5 mBq/m^3 during a peak in the middle of March 2011 (Povinec *et al*., 2013a). An atmospheric signal was also detected in air above Monaco (Masson *et al*., 2011; Pham *et al*., 2012). The extremely low levels of atmospheric ^{137}Cs concentrations that were still measurable from nuclear weapons tests and the Chernobyl accident increased over Europe by about 1000 times; however, these recently elevated levels from Fukushima do not pose any risk to man or animals.

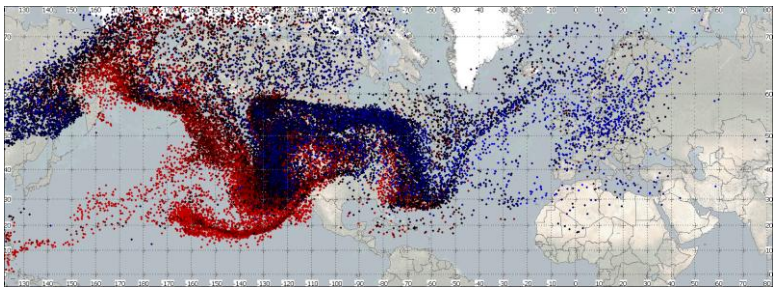

Figure 1. Lagrangian particle modeling of the distribution of radioactive particles in the atmosphere after the Fukushima accident. Particles spacing from 13 to 26 March, 2011, simulated using the Lagrangian dispersion model (time of the particles release was on 12 March 2011). Shades of red indicate particles in the bottom layer (up to 3 km height); black to dark blue indicate the middle layer (up to 6 km); and light blue indicates the layer above 6 km (status on 21/3/2011 at 06:00 UTC; modified after Povinec *et al*., 2013c).

[2] The International Nuclear and Radiological Event Scale (INES) was introduced in 1990 by the International Atomic Energy Agency (IAEA) in order to enable prompt communication of safety-significant information in case of nuclear accidents.

Radionuclides in seawater and biota

The radionuclide levels in seawater offshore from Fukushima varied equally for ^{137}Cs and ^{134}Cs between about 1 kBq/m^3 to about 90 MBq/m^3. The average ^{137}Cs (and similarly also ^{134}Cs) activity concentration off Fukushima during most of 2011 was about 10 kBq/m^3. The pre-Fukushima ^{137}Cs concentration in surface waters of the North-West Pacific Ocean was about 1 Bq/m^3, thus the post-Fukushima levels were by about 10,000 times higher, figure 2 (Buesseler *et al*., 2012; Povinec *et al*., 2013b).

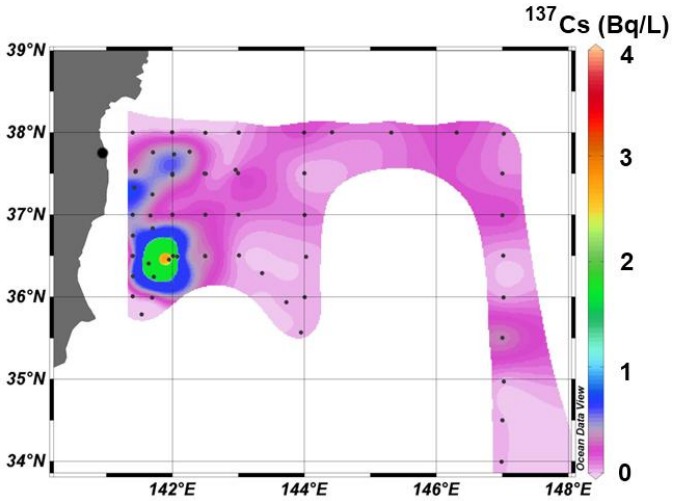

Figure 2. Distribution of ^{137}Cs in surface waters of the Pacific Ocean in June 2011 (data from Buesseler *et al*., 2012; Povinec *et al*., 2013b).

Global ocean circulation modeling has shown that the radioactive plume in the Pacific Ocean is predominantly driven by the fast moving Kuroshio Current that sweeps past the east coast of Japan. Maximum concentrations of ^{137}Cs in the North-western Pacific waters reached about 25 Bq/m^3, figure 3. After 4 to 6 years the plume may reach the west coast of North America with ^{137}Cs concentrations of 3-9 Bq/m^3 (depending on the atmospheric input of ^{137}Cs to the ocean), which is about a factor of 3-9 higher than the present background from testing of nuclear weapons in the Pacific Ocean, and demonstrates the enormous dilution capacity of the oceans for pollutants (Buesseler *et al*., 2012; Masumoto *et al*., 2012; Nakano & Povinec, 2012; Rypina *et al.*, 2013). After 10 years, all the Pacific Ocean surface and medium depth waters (< 1000 m depth) will be labeled with the Fukushima-derived ^{137}Cs with concentrations below 3 Bq/m^3 (Nakano & Povinec, 2012; Povinec *et al*., 2013a).

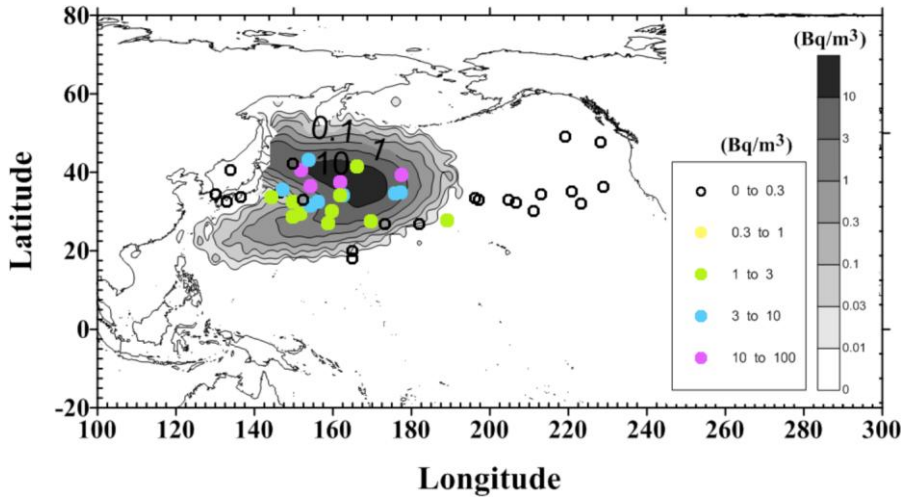

Figure 3. A comparison of predicted (isolines, calculated using the LAMER code, Nakano and Povinec, 2012) and measured ^{137}Cs activity concentrations (dots) in surface water of the NW Pacific Ocean (1 Bq/m^3 from the global fallout background was deducted from the measured ^{137}Cs concentration obtained by Aoyama *et al.* (2013); modified from Povinec *et al*., 2013b).

The ^{137}Cs concentrations in fish caught offshore from Fukushima varied with several orders of magnitude, from about 2 Bq/kg ww (wet weight) to about 5000 Bq/kg ww, with the majority of ^{137}Cs concentrations are in the range of 10-100 Bq/kg ww. 2.5 years after the accident the amount of fish above the limit of 100 Bq/kg is decreasing in fish caught offshore of the Fukushima Prefecture (MAFF-Japan, 2013) and only about 10% showed levels above this limit. Levels away from the coast of Fukushima are significantly lower. There were reports that some species caught in the vicinity of the discharge area showed significantly higher levels, but they are not used as food. After the Fukushima accident, the Japanese Government tightened and applied very strict regulations for radionuclide content in seafood. The Japanese limit for ^{134}Cs and ^{137}Cs in food including seafood after the Fukushima accident was decreased from 500 to 100 Bq/kg ww, by this they became by about a factor of four to ten lower than for other Asian countries. The *Codex Alimentarius*[3] value of 1000 Bq/kg ww, which has been adopted by most of the world countries, assures the maximum effective dose limit to population is well below the ICRP (International Committee on Radiological Protection) and the IAEA (International Atomic Energy Agency) recommended maximum value for the citizens of 1 mSv/year.

Radiation doses

The estimated radiation doses to the Japanese population from inhalation, the external exposure from radioactive clouds and deposited radionuclides, and ingestion of terrestrial food were estimated by the World Health Organization (WHO, 2013) to be generally significantly below 25 mSv per year. The impact of the Fukushima accident should be therefore below the 50 mSv/y limit for the statistical risk of cancer, or the 500 mSv deterministic effect limit when serious health problems could be expected (ICRP, 2012).

Adopting a conservative approach, a conservative upper radiation dose can be estimated for a typical Japanese seafood consumer. Anticipating that the average ^{137}Cs (and similarly also ^{134}Cs) activity

[3] The Codex Alimentarius Commission, established by FAO and WHO in 1963 develops harmonized international food standards, guidelines and codes of practice to protect the health of the consumers and ensure fair practices in the food trade.
http://www.codexalimentarius.org/ further explanation e.g.: http://en.wikipedia.org/wiki/Codex_Alimentarius

concentration in surface seawater off Fukushima during most of 2011 was about 10 kBq/m^3, then the dose commitment would be 0.7 mSv/y from ingestion of radionuclides in fish, shellfish and seaweed (for 4 PBq of $^{134+137}$Cs released to coastal waters). Although this dose is by about four orders of magnitude higher than the pre-Fukushima dose of 0.03 µSv/y, estimated using the pre-Fukushima seawater ^{137}Cs content of 1 Bq/m^3, it is comparable to doses from other natural sources. The individual dose commitment from the consumption of ^{137}Cs and ^{134}Cs in marine biota caught in the open North-West Pacific Ocean in 2012 was estimated to be about 2 µSv/y, which is about 30-times greater than the global fallout background. However, this dose is 350-times lower than the dose from the consumption of natural ^{210}Po in fish and shellfish (0.7 mSv/y). Overall, it is expected that the real doses from consumption of marine food from the Fukushima region are significantly lower because the concentrations in seawater proved to be significantly lower and were usually below 1 Bq/l about 2.5 years after the accident and outside the harbor of the Fukushima Daiichi NPP, and thus should consequently cause no negative radiation effects (NRA, 2014).

The radiation doses to the world population from inhalation and ingestion of terrestrial and marine foods were several orders of magnitude lower than the recommended maximum limit to the public from external sources (1 mSv/y), or the world average dose from natural sources (2.4 mSv/y).

The Fukushima radiation doses to the general public could be compared with radiation doses from other sources, e.g. 0.1 mSv from one simple X-ray test, 0.4-0.6 mSv from a mammogram, and 10-30 mSv from a CT scan, or similar nuclear medicine diagnostics.

Conclusions

The discharges of radioactive substances from the Fukushima Daiichi NPPs into the Pacific Ocean have created a lot of concern in relation to pollution of marine food and wide-range contamination of the marine environment not only for the Japanese population but also in countries around the Pacific Ocean, and even in other parts of the world. However, radionuclide monitoring in seawater and biota offshore Fukushima and in the Pacific Ocean, as well as estimation of radiation doses to the Japanese and world population have indicated that no harmful effects are expected from ingestion of seafood contaminated by Fukushima-derived radionuclides.

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