INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Updated summary of knowledge concerning the impact on the marine environment of radioactive release from the damaged nuclear site of Fukushima Dai-ichi

13 July 2012

This memo presents and comments on the latest information gathered by IRSN since the previous summary published on 26 October 2011 on the same subject and IRSN report entitled "Fukushima, one year later - Initial analyses of the accident and its consequences" published on 12 March 2012.

As a reminder, the Fukushima accident led to a pollution of the marine environment caused by two main sources:

- *Radioactive liquids discharged directly into the ocean initially composed of various radionuclides. These were mainly iodine-131, iodine-132, tellurium-132 and caesium-134, caesium-137 and caesium-136. IRSN estimates that more than 80% of these discharges were produced between 21 March and 8 April 2011, and 27.10^{15} Bq of caesium-137 were discharged up to July 2011;*
- *Polluting atmospheric deposition on the surface of the ocean represents approximately 10% of the total radioactive discharges into the ocean, according to the latest IRSN estimates described in this memo,*

From June 2011 onwards, caesium-134 and caesium-137 were the only remaining detectable radionuclides in seawater, and their concentrations off the coast beside the Fukushima nuclear power plant have been steadily decreasing over the course of 2011 as they disperse in the ocean. These concentrations declined more slowly in the second half of 2011 and the first half 2012, however, indicating that radioactive caesium is still regularly entering the ocean along the coast, particularly in the immediate vicinity of the damaged nuclear plant. This is the result of discharges from the Fukushima plant, which are significantly lower than those in March 2011, and intake from run-off due to leaching of the contaminated land.

In the coastal area, the sediment was initially contaminated through direct contact between surface and polluted seawater; the contamination levels now change as a result of the intake of sediment particles by the rivers, particularly after rainy periods.

Significant concentrations of radioactive caesium are still measured in various marine species (fish, molluscs and sea urchins) caught in the coastal waters of the Fukushima, Miyagi and Ibaraki prefectures, sometimes exceeding the food standards which were tightened on 1 April 2012. These concentrations are gradually decreasing in filter-feeder species such as mussels, oysters and clams or grazing organisms such as sea urchins and abalones, but they are virtually unchanged in sedimentary species (ainames, halibuts, gurnards, rays, rockfish, etc.). As a result, the monitoring of marine species caught in the north-eastern coastal waters of Japan should be continued.

The scientific publications of 2011 and 2012, as well as the analyses conducted by IRSN on a wahoo sample caught in October 2011 to the north of one of the Hawaiian islands (Oahu), show that traces of caesium-134, which can be attributed to the Fukushima accident, can be found in pelagic species (such as some species of tuna) that were in the waters off the Japanese coast at the time of the accident and then migrated over large distances (to the Californian coast, for example). Although the levels found in these migratory species are very low and not dangerous to human, the continuation of such studies would provide a better understanding of the importance of this transport on the oceanic scale.

1. CHANGES IN THE RADIOACTIVE POLLUTION IN SEAWATER

1.1. Main radionuclides observed in seawater since 21 March 2011

On 21 March 2011 and the following days, considerable radioactive pollution was detected in the marine environment near to the Fukushima Dai-ichi nuclear power plant. This pollution was mainly characterised by means of measurements conducted on samples of seawater, sediment and species living in the marine environment. The results have been published by MEXT[1] and TEPCO. IRSN, which could not conduct measurements in Japan, has regularly collected and analysed these results in order to monitor changes in the radioactive pollution in the marine environment. The latest measurements date from June 2012.

The measurement results published in Japan mainly concern the gamma-emitter radionuclides listed in Table 1.

Table 1 – Main radionuclides measured in seawater

Main radionuclides detected	
Radionuclide	*Half-life*
Iodine-131 (131I)	8 days
Caesium-137 (137Cs)	30.15 years
Caesium -134 (134Cs)	2.1 years
Caesium-136 (136Cs)	13.1 days
Tellurium-132 and Iodine-132 (132Te and 132I)	78 hours

Other artificial radionuclides, most of which have a short half-life, have also been detected occasionally, but at lower concentrations. There are fewer measurements concerning pure beta-emitter radionuclides: Nine results for strontium-90 in seawater, at concentrations ranging from 1 to 10 Bq/L, representing between 1 and 20% of the radioactivity of caesium-137 measured in the same samples, i.e. a higher proportion than that observed in the atmospheric fallout from the accident onto land areas of Japan, which is in the region of 0.1%.

Iodine-131 (^{131}I) and caesium-137 (^{137}Cs) are the main radionuclides monitored in 2011 in the marine environment. Although it was predominant at the time of the accident, the amount of iodine-131 declined sharply in the following weeks as a result of its rapid radioactive decay. As a result, it has no longer been detectable since the end of May.

This radioactive pollution has had two main sources:

- **Liquid radioactive discharges from the damaged site:** in interpreting the seawater radioactivity measurement results published in Japan, IRSN has estimated an overall discharge of 27.10^{15} Bq of caesium-137 into the sea between March and July 2011, the majority of which (82%) occurred before 8 April 2011, the date on which a rapid and regular reduction (50% every 6.9 days) was observed in the caesium-137 levels detected in the coastal waters near to the damaged plant up to summer 2011;
- **Atmospheric fallout on the surface of the sea when the radionuclides are discharged into the air:** the resulting diffuse pollution has been difficult to single out in the sea because the surface water receiving the atmospheric fallout rapidly mixes with the rest of

[1] Ministry of Education, Culture, Sports, Science and Technology in Japan.

the seawater as the result of advection and dispersion. Only the concentrations measured in seawater more than 10 km away from the plant before 24 March 2011 (or a little later, further away), when the direct liquid discharges were still relatively low, can be attributed to these atmospheric discharges. IRSN's work in 2011 to estimate the atmospheric discharges caused by the accident and model their atmospheric dispersion has led it to revise its estimate of the cumulative deposition of caesium-137 on the sea within an 80-km radius to 2.6×10^{15} Bq, representing approximately 10% of the overall activity of caesium-137 directly discharged into the sea by the Fukushima Dai-ichi plant.

With regard to the atmospheric fallout onto the sea, it should be emphasised that the air contamination or the depositions onto the marine environment are not measured directly, making any precise estimate of this intake impossible, unlike the Japanese terrestrial environment, in which many environmental measurements are performed. IRSN's study of caesium-137 concentrations in seawater and its modelling of the radionuclide's dispersion in the Japanese coastal waters seem to support its latest estimates of atmospheric fallout, however (see paragraph 1.3, below). In any event, the existing uncertainties regarding the estimated fallout should be relativised, as this fallout forms only a secondary part of the radioactive pollutants entering the marine environment during the course of the accident, which mainly consisted of liquids directly discharged from the Fukushima Dai-ichi plant.

1.2. Changes in seawater contamination on the coast near to the Fukushima Dai-ichi plant

The measurements performed near the power plant revealed considerable contamination of the marine environment from 21 March 2011 onwards as a result of highly-contaminated water flowing from the damaged reactors into the sea.

From June 2011 onwards, caesium-134 and caesium-137 remained the only detectable radionuclides in the seawater, with an activity ratio of approximately 1 relative to the date of the accident. The temporal and spatial changes in the caesium-137 concentrations are representative of those of all of the radionuclides measured in the sea.

Figure 1 shows the changes in the concentrations measured near to the damaged plant between the end of March 2011 and the beginning of May 2012, where the detection limit was revised from 10 Bq/L to 1 Bq/L in October 2011; these concentrations are representative of the flow of radionuclides discharged by the plant. Although there are still some significant values, increasing numbers of the concentrations are below the detection limit. The levels decrease regularly over the course of time, with an apparent decay period of approximately two months starting from June 2011. The same is true of the measurements performed along the coast at a distance from the damaged plant, albeit with a slightly longer apparent decay period of approximately three months. These kinetics are much slower than those identified immediately after the direct discharges. They reflect a continuous yet declining intake of caesium-137 in solution into the seawater. At a distance from the coast, the concentrations are approximately 0.01 Bq/L, five times higher than those measured before the accident.

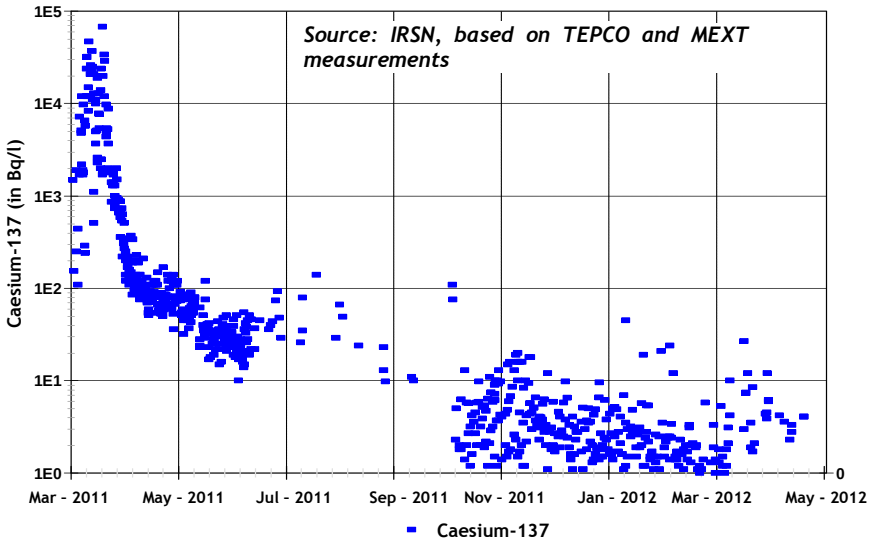

Figure 1 – Changes in ^{137}Cs concentrations in seawater within 500 m of the Fukushima Dai-ichi nuclear power plant

Figure 2 shows the spatial distribution of average caesium-137 concentrations between January and May 2012. This map shows that concentrations have decreased by a factor of approximately 1,000 since the accident. The source of this residual contamination seems to be spread along the coast 50 km to the north and south of the damaged plant. The distribution's similarity with that of the sediment contamination (see map in Figure 5 in Chapter 2) suggests that the caesium comes from a similar source. It may be due to the leaching of soils contaminated by meteoric water, bringing a steady flow of water loaded with caesium to the sea, or to the release of the caesium deposited on the sediment in 2011.

The changes in the radionuclide concentrations in the seawater indicate that radioactive caesium is still regularly entering the sea along the coast, particularly in the immediate vicinity of the plant. This is the result of discharges from the nuclear power plant and run-off due to leaching of the contaminated land.

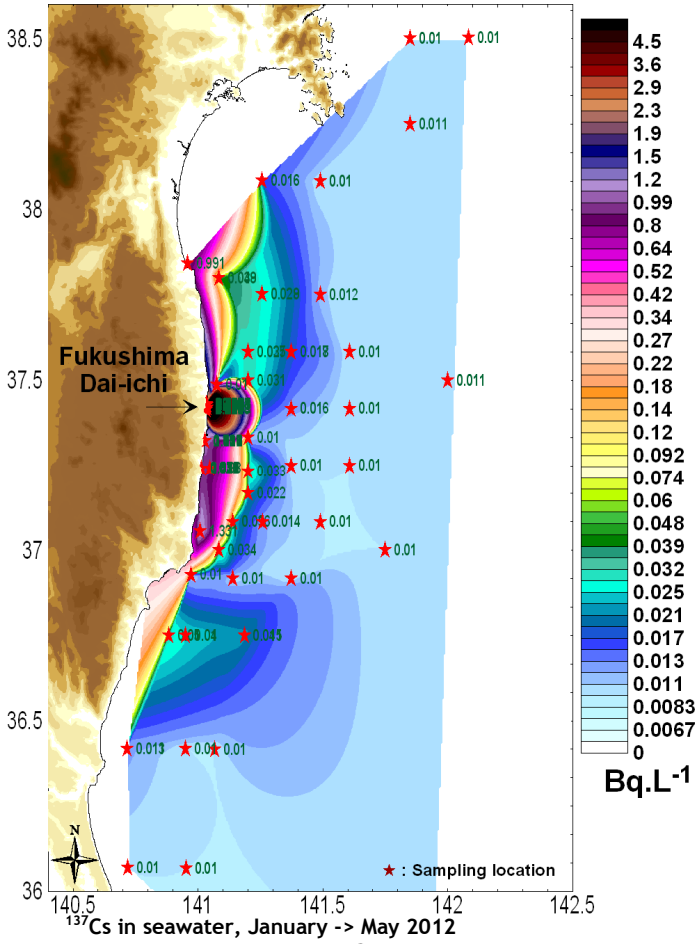

Figure 2 – Spatial distribution of average concentrations in ^{137}Cs in seawater, deduced from all measurements available between January and May 2012

1.3. Simulation of caesium-137 dispersion in seawater off the coast of Japan

IFREMER was approached by IRSN to run simulations of discharge dispersion from Fukushima Dai-ichi. It used the Mars-3D model, Mercator-Ocean hydrodynamic limit conditions and NCEP meteorological forcing.

The caesium-137 direct discharge measurement used in this simulation is taken from the calculations shown in the previous summaries and in IRSN's report "Fukushima, one year later" published on 12 March 2012: 27 PBq.

As stated in paragraph 1.1 above, the caesium atmospheric fallout onto the surface of the sea within 80 km of the Fukushima Dai-ichi nuclear power plant has been revised to 2.6 PBq as a result of IRSN's work in 2011 to estimate the atmospheric discharges caused by the accident and model their dispersion (see IRSN's report "Fukushima, one year later" published on 12 March 2012). These discharges are mainly in the form of wet deposits (during rain), and their spatial distribution estimated by IRSN by means of modelling is shown on the map in Figure 3.

When the dispersion of the caesium 137 discharged into the sea (direct liquid discharges and atmospheric fallout) was modelled, IRSN adopted the hypothesis that the radionuclide was solely in soluble form.

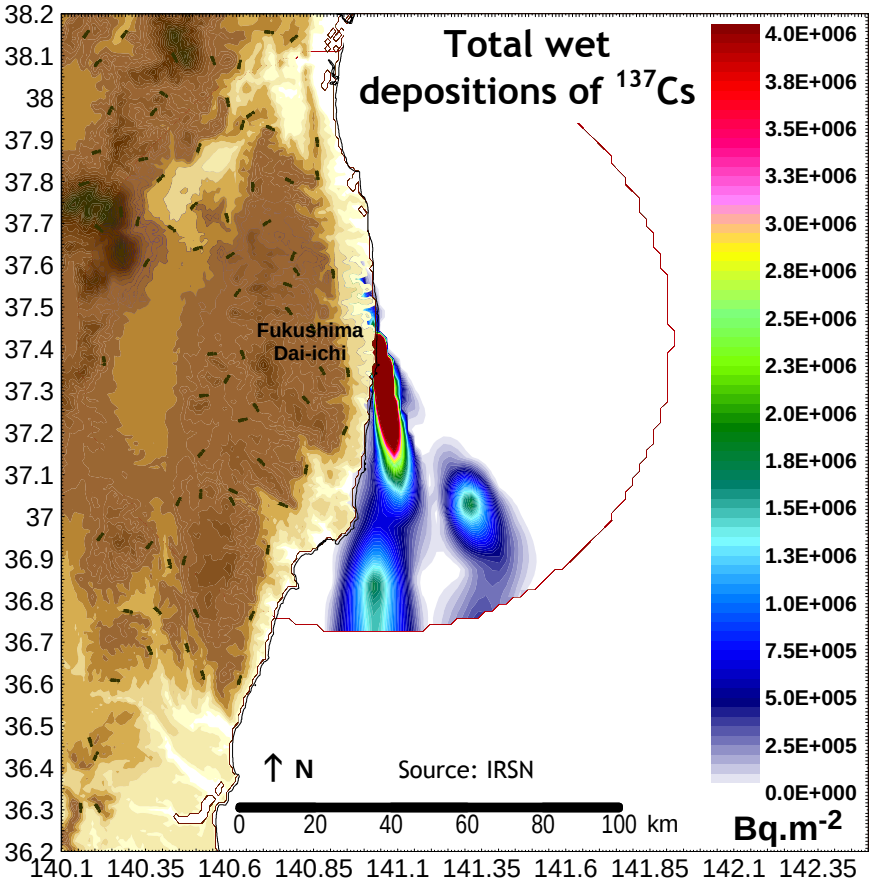

Figure 3 – Distribution of the total atmospheric fallout of ^{137}Cs onto the sea as at 23 March 2011, estimated by IRSN by modelling the atmospheric dispersion of discharges from the Fukushima Dai-ichi accident

The caesium-137 marine dispersion model has been modified in order to reproduce the renewal kinetics of the water masses deduced from the caesium-137 measurements (Figure 4). The model does not tend to underestimate or overestimate the concentrations (see Figure 5); the average difference between the simulated and measured values is 55%. The obtained results, when compared with the measured caesium-137 concentration values in seawater at a distance from the damaged nuclear power plant or even further from the coast, also confirm the estimated caesium-137 quantities resulting from atmospheric fallout.

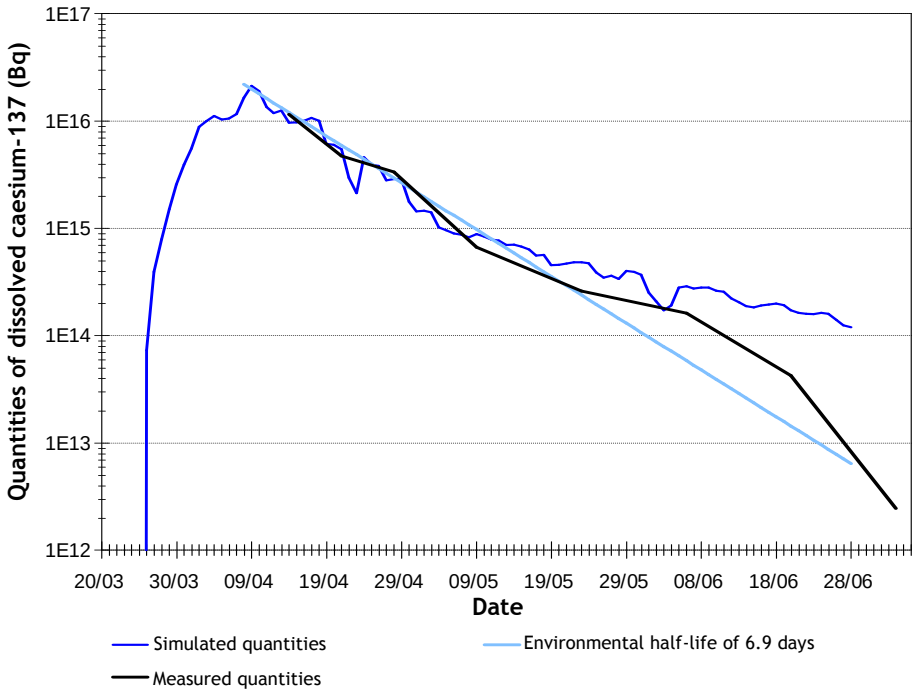

Figure 4 - Quantities of ^{137}Cs in the measured and Mars-3D-simulated seawater between 15 April and 26 July 2011 within a 50-km radius of the plant

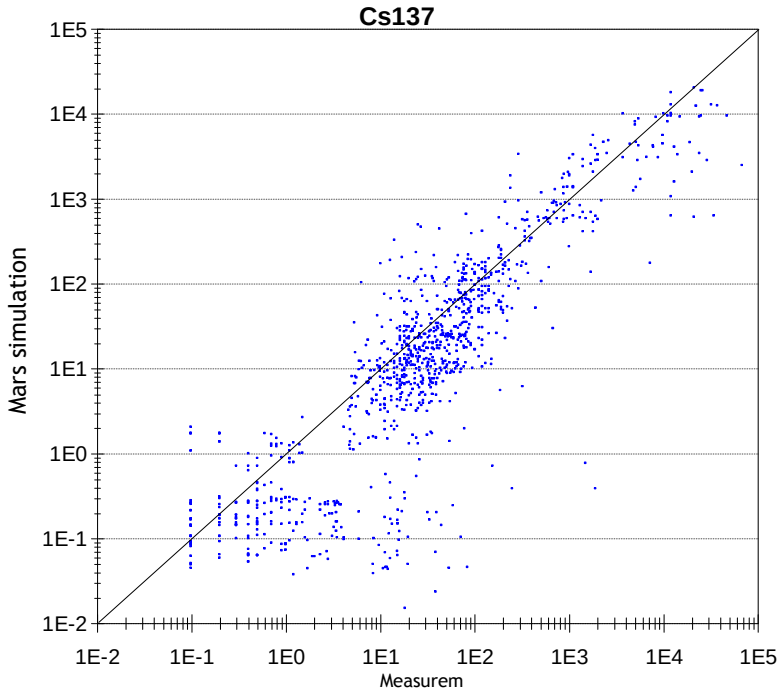

Figure 5 - Comparison of measured and simulated ^{137}Cs concentrations in seawater between March and June 2011

2. RADIONUCLIDES IN SEDIMENT

The materials in suspension in the seawater tend to fix some of the radionuclides dissolved in the water column according to the levels of radioactivity in the surrounding water. These particles end up as sediment on the sea floor, creating a contaminated superficial deposit.

Figure 6 shows a map of caesium-137 concentration distribution in sediment. The actual concentrations are generally less than 1000 Bq/kg. In the coastal area, the sediment was initially contaminated through direct contact between its surface and the polluted seawater; its contamination level now changes as a result of sediment particles carried by the rivers, particularly after rainy periods.

The finer caesium-bearing particles found in the sedimentary mix are likely to carry the adsorbed radionuclides for long distances.

Benthic organisms living in direct contact with sediment, together with filter feeders, are most seriously affected by the residual pollution in sediment.

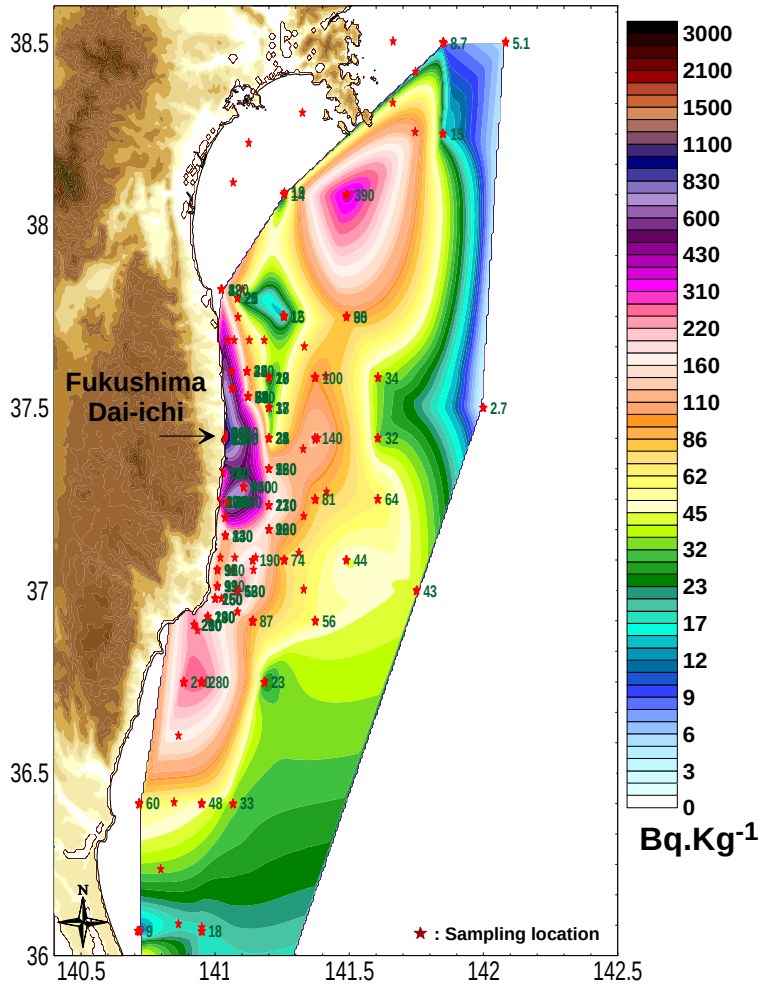

Figure 6 - Map of average ^{137}Cs concentrations in sediment, measured between January and May 2012

3. RADIONUCLIDES IN MARINE SPECIES

3.1. Marine species exceeding the maximum permitted consumption levels for caesium

In the last few months and before 1 April 2012, the date on which the new, more stringent food standards were introduced by the Japanese authorities, the only marine organisms whose caesium concentration exceeded the standard in force at the time (500 Bq/kg wet) were fish, all of which were from the Fukushima prefecture. No organisms caught in the sea outside the area close to the power plant or off the coast exceed this standard.

After this standard was tightened to 100 Bq/kg wet, it was sometimes exceeded not only by various species of fish, but occasionally also by clams and sea urchins. This concerns samples caught along the coast of the Fukushima, Ibaraki and Miyagi prefectures.

3.2. Concentrations observed in marine animals

The graph in Figure 7 shows the measured caesium concentration levels in various marine species between March 2011 and March 2012, which are published on a regular basis.

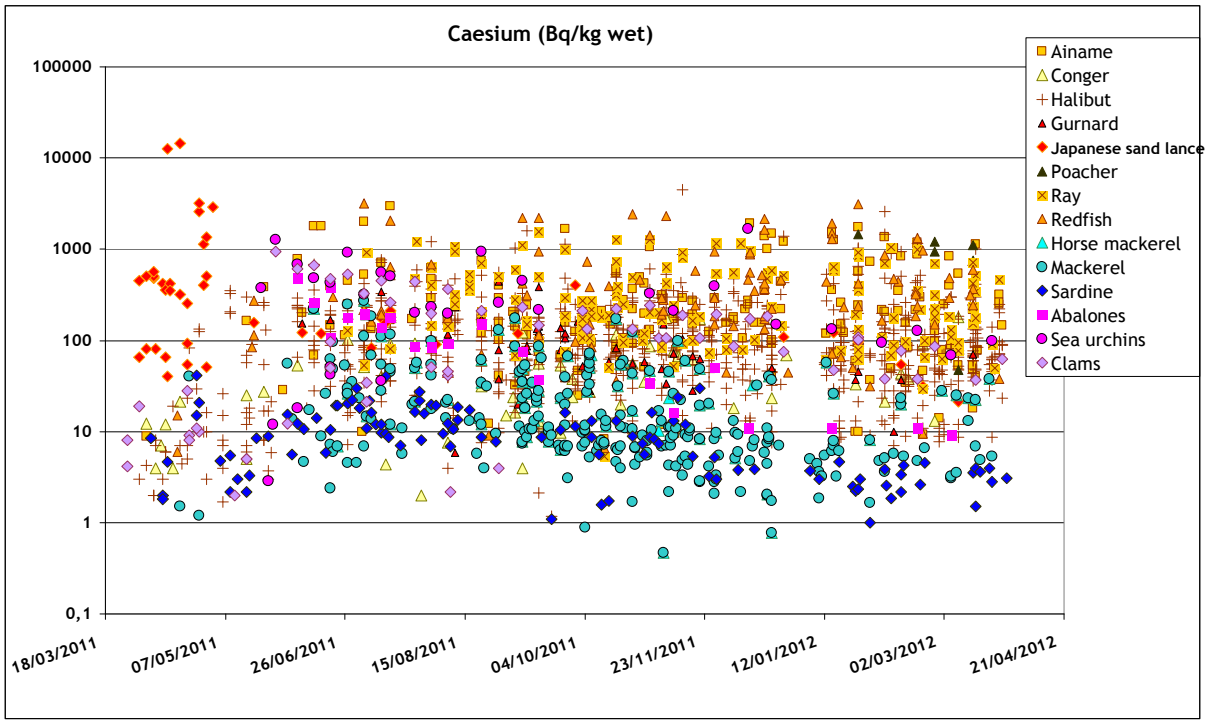

Figure 7 – Changes over time of $^{137}Cs+^{134}Cs$ (Bq/kg wet) concentrations in some marine products, between March 2011 and March 2012

Of all of the sampled marine products, the Japanese sand lances were found to have the highest contamination levels, which were detected early in the fishery product monitoring programme. Caesium-137 and caesium-134 were detected in all Japanese sand lance samples caught in the Fukushima and Ibaraki prefectures; the maximum concentrations were 12,500 Bq/kg wet ($^{134}Cs+^{137}Cs$) off the Iwaki coast on 13 April.

The Japanese catch and eat Japanese sand lances (*Ammodytes personatus*) in their larval and juvenile stages, both of which are pelagic (i.e. live in the water column) in the period from January

to April. The adults, however, live buried in the sediment from May to December and are no longer caught, which explains why the data for this species practically vanish from late April 2011 onwards. Monitoring was resumed at the beginning of 2012, when the pelagic Japanese sand lances (sandeels) could be caught once more. Their levels range from 54 to 122 Bq/kg wet ($^{134}Cs+^{137}Cs$) in samples from Iwaki city (Fukushima prefecture), while the samples caught off the coast of Soma city range from 20 to 30 Bq/kg wet. These levels reflect the considerably lower concentrations in the surrounding environment, as Japanese sand lances mainly feed on zooplankton, which is in strict equilibrium with the environment in which it grows.

In addition to the concentrations found in Japanese sand lances (sandeels), Figure 7 shows the concentration curves for both caesium isotopes in other regularly-sampled species. It is difficult to find a trend in the contamination of fish over the course of time, given the wide range of results obtained.

Overall, sedimentary fish (represented by orange symbols in Figure 7) such as ainames, halibuts, gurnards, rays and rockfish, tend to have higher concentration levels than pelagic species (represented by bluish symbols in Figure 7) such as mackerel, sardines and horse mackerel;

In addition to fishes, it should be pointed out that the sea urchin, abalone, and clam samples caught in the Fukushima prefecture sometimes also contained high levels.

Iodine-131 has not been detected in the organisms since mid-June 2011, reflecting its level in the seawater.

3.3. Expected changes for marine species

The concentrations found in filter feeders such as mussels, oysters and clams, or grazing organisms such as sea urchins and abalones, are decreasing due to the absence of new discharges. The changing levels in these organisms over the course of time reflect the changes in the surrounding environment quite well, and the trends clearly show the decreasing concentrations over time (Figure 8).

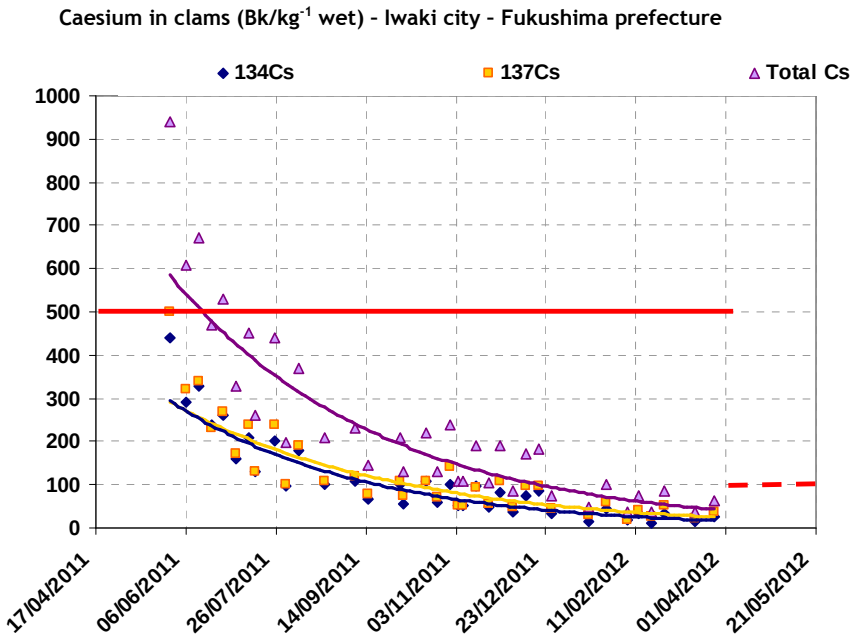

Figure 8 - Changes in caesium concentrations in clams (Pseudocardium sachalinense) caught off the Iwaki coast in the Fukushima prefecture over time, up to March 2012. The red lines represent the maximum permitted levels for food consumption (solid line up to 1 April 2012: 500 Bq/kg wet; dotted line from 1 April 2012 onwards: 100 Bq/kg wet)

As expected, the change in fish contamination levels over time does not reflect the decrease detected in the seawater. Figure 9 takes as an example the changes in concentrations in a sample of a species of skate (*Okamejei kenojei*) caught in the coastal waters of the Fukushima prefecture. A very slight trend towards a decline in activity seems to be emerging. The highest values are to be found on the Iwaki city coast. In fish, concentration variations could very well be partly due to their movements in the case of species that are not strictly confined to the area in which they were caught, but they also reflect their complex trophic regimes. In addition, the biological elimination periods for caesium in fish are longer than in other marine organisms.

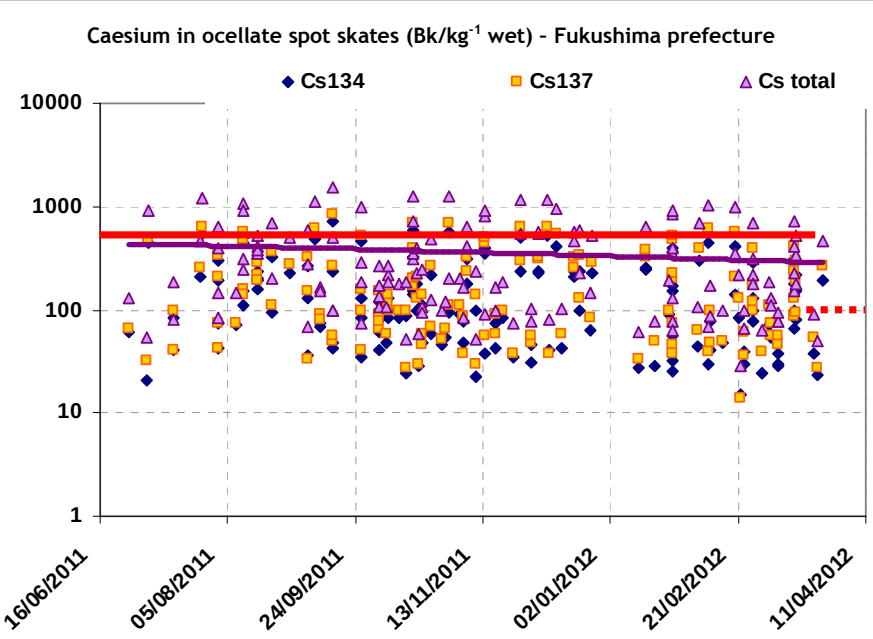

Figure 9 – Changes (and trend) in radioactive caesium concentrations over time, in a species of spot skate (Okamejei kenojei) caught off the Iwaki coast in the Fukushima prefecture. The red lines represent the maximum permitted levels for food consumption (solid line up to 1 April 2012: 500 Bq/kg wet; dotted line from 1 April 2012 onwards: 100 Bq/kg wet)

Generally speaking, fish will be the best medium- and long-term indicators of caesium contamination in the marine domain. Caesium has higher concentration factors in fish, and they tend to be greater in species that are higher in the trophic chain. As a result, although in the short term the highest concentrations tend to be found in species at the beginning of the food chain, the predators at the top of the food chain should exhibit higher levels in the longer term, once the caesium has been transferred through the various links in the trophic networks. These levels should be even higher in the case of species whose way of life involves close contact with sediment and whose habitat is close to the contaminated area.

As a result, the monitoring of marine species caught in the north-eastern coastal waters of Japan should be continued.

3.4. Other data published in the standard literature

Between 4 and 18 June 2011, American researchers organised a sampling campaign in the North Pacific (Buesseler et al., 2012[2]). As well as measuring concentrations in the water, they also provide various results regarding plankton and mesopelagic fish samples. The caesium-134 and caesium-137 concentrations range from the detection limit to 56.4 Bq/kg dry, depending on where the samples were taken and their type. The $^{137}Cs/^{134}Cs$ activity ratios are usually approximately 1.

It should be noted that silver-110m was detected in the plankton in almost all cases, at levels ranging from the detection limit to 23.6 Bq/kg dry. This element was not detected in the fish,

[2] Buessler K.O., Jayne S.R., Fisher N.S., Rypina I.I., Baumann H., Baumann Z., Breier C.F., Douglass E.M., George J., Macdonald A.M., Miyamoto H., Nishikawa J., Pike S.M., Yoshida S., Fukushima-derived radionuclides in the ocean and biota off Japan. Proc Natl Acad Sci USA, 109:5984-5988.

however. The presence of this radionuclide is not mentioned in the marine discharges, but it was detected in the soil samples from around the plant.

Recently, another scientific article (Madigan et al., 2012[3]) presented measurement results obtained in tuna caught off the Californian coast. In this work, samples of two tuna species were measured: firstly, the Pacific bluefin tuna (*Thunnus orientalis*), which is a migratory species and secondly, the yellowfin tuna (*Thunnus albacares*), which is also a migratory species but tends to remain off the coast of California and Mexico. Whereas the bluefin tuna samples revealed the presence of both caesiums (^{137}Cs and ^{134}Cs), the yellowfin tuna samples contained only caesium-137. The levels are low, in the order of a few Bq/kg dry. The caesium-134 found in the bluefin tuna is the result of exposure while passing through the waters contaminated by discharges caused by the Fukushima nuclear power plant accident, off the coast of Japan. The absence of this radionuclide in the yellowfin tuna leads us to conclude that this species of fish caught off the Californian coast was not exposed to discharges caused by the Fukushima accident but contains only the residual caesium-137 resulting from old fallout due to old nuclear weapon tests in the atmosphere and from the Chernobyl accident. Migratory species are therefore a global vector of radionuclide transport across the oceans. Even though these migratory species exhibit low radionuclide levels, the continuation of such studies would provide a better understanding of the importance of this transport and the migration of these species.

During the Tara's campaign in the North Pacific, a sample wahoo was caught for IRSN in October 2011 to the north of Oahu, one of the Hawaiian islands. This sample was analysed by IRSN's Environmental Monitoring Laboratory (LESE) in Tahiti; it revealed traces of caesium-134 (approximately 0.05 Bq/kg dry), confirming that it was contaminated by the discharges caused by the Fukushima accident.

These various results emphasise the usefulness of additionally monitoring migratory species in the South Pacific. To quote only the example of Pacific bluefin tuna, fish have been observed migrating from the North-West Pacific toward the waters of New Zealand.

[3] Madigan D.J., Baumann Z., Fisher N., Pacific bluefin tuna transport Fukushima-derived radionuclides from Japan to California. Proc Natl Acad Sci USA, *doi:10.1073/pnas.1204859109*