IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

Impact on the marine environment of radioactive releases following the nuclear accident at Fukushima Daiichi

13 May 2011

This information note updates and supplements the previous note of 4th April on the same subject

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Significant radioactive contamination of the marine environment was identified after the accident in the nuclear power plant at Fukushima Daiichi. This radioactive pollution comes from fall-out into the ocean of some of the radionuclides present in the atmospheric plume dispersed after the accident, and from direct spillage of contaminated water from the plant. In the immediate vicinity of the plant (less than 500 m), the concentrations at the end of March and in early April reached up to several tens of thousands of becquerels per litre (Bq/L) for caesium 134 and 137 and even exceeded 100,000 Bq/L for iodine 131. These concentrations began to fall in this area as of 11th April and, at the end of April, had reached a value close to 100 Bq/L for caesium 137. The radionuclides dissolved in the seawater will continue to be transported by ocean currents and will disperse in the oceans over very long distances, with increasingly small concentrations. The radionuclides which bind to the suspended particles in the water or to the sediment on the sea bed will lead to sedimentary contamination, in particular along the coast close to the plant, where contamination of the sediments was measured in late April. Iodine 131, with a short radioactive half-life, is now less present than the longer-lived radionuclides, in particular caesium 134 and 137. These latter will remain detectable for several years around the North Pacific, but in very low concentrations (about 5000 times lower than the concentration of potassium 40, a naturally-occurring radionuclide permanently present in seawater). However, significant pollution of the seawater along the coastline close to the damaged plant could persist because of the continuous input of radioactive substances transported to the sea by surface water run-off from contaminated soils.

The radioactive pollution of the seawater leads to contamination of the plant and animal species exposed to it. For example, certain species of fish (Japanese sand-eels) landed in the ports south of the Fukushima Daiichi plant contained radionuclide concentrations of several thousand becquerels per kilogram, sometimes up to 25 times the level permitted for commercial sale. A theoretical, indicative estimate of the expected concentrations in fish, based on the values measured in the seawater, indicate that this maximum permitted level could be exceeded up to more than a hundred kilometres from the plant.

1. ORIGINS OF CONTAMINATION OF THE MARINE ENVIRONMENT

Since the beginning, on 21st March, significant radioactive pollution has been observed in the marine environment.

This radioactive pollution has three possible origins: liquid radioactive releases from the damaged site, atmospheric fall-out onto the surface of the sea, and transport of radioactive pollution by leaching through contaminated soil.

The main radionuclides regularly measured in the seawater are (T = radioactive half-life): iodine 131 (T = 8 days), caesium 137 (T = 30 years), caesium 134 (T = 2.1 years), caesium 136 (T = 13.1 days), tellurium 132/iodine 132 (T = 78 hours). Others were also occasionally detected, at far lower concentrations: tellurium 129m/tellurium129 (T = 33.6 days), barium 140/lanthanum 140 (T = 12.7 days), ruthenium 105 (T = 4.4 hours), ruthenium 106 (T = 368 days), molybdenum 99/technetium 99m (T = 65.9 hours), cobalt 58 (T = 70.9 days).

Iodine 131 and caesium 137 are representative of the dispersion of all the radionuclides and they are the main ones considered in the rest of this note. Although it was predominant at the time of the accident, iodine 131 is now measured at lower levels than caesium 137 owing to its rapid radioactive decay.

Our understanding of the quantities of radioactive products released into the seawater is still approximate. It could be fine-tuned by continuing to take seawater measurements and interpreting the results by means of dispersion simulations.

1.1. Direct releases into the sea close to the damaged reactors

The high concentrations measured in the seawater in the immediate vicinity of the Fukushima Daiichi plant are the result of liquid radioactive releases which no doubt have several origins. This includes the water used to cool the damaged reactors, which was in contact with materials heavily contaminated by the atmospheric releases, and some of which trickled out to sea. Another part comes from the water which escaped from the containments of reactors 2 and 3. In particular, a crack in the pit adjacent to the reactor 2 turbine hall led to the release of heavily contaminated water directly into the sea. TEPCO stopped this release on 6th April at about 6h00 local time, by plugging the leak with an injection of sodium silicate.

The precise quantities and the duration of the liquid releases into the sea are unknown. Based on an IRSN estimate, using the concentrations measured in the water of the n°2 reactor turbine hall, 2.3x10^{+15} Bq (2.3 million billion becquerels) of caesium 137 could have been released into the sea. In addition, from 4th to 10th April, TEPCO discharged "slightly contaminated"" water into the sea. This was 10,000 tons of liquid effluent stored in tanks. These discharges, estimated by TEPCO at 1.5x10^{+11} Bq, did not significantly increase the concentrations resulting from the previous releases.

If direct releases into the sea took place after 10th April, then the fall in the concentrations measured in the vicinity of the facilities show that they were far smaller than the previous releases.

Figure 1 shows the evolution of the concentrations measured in the seawater less than 500 metres from the Daiichi plant. They rose between 25th and 28th March (up to 12 000 Bq/L of caesium 137, 74 000 Bq/L of iodine 131) peaking between 29th March and 7th April (up to 63 000 Bq/L of caesium 137, 180 000 Bq/L of iodine 131). As of 11th April, the concentrations of iodine131 and caesium 137 began to fall. They stabilised at around 100 Bq/L for caesium 137. Iodine131 will continue to fall owing to its radioactive half-life of 8 days.

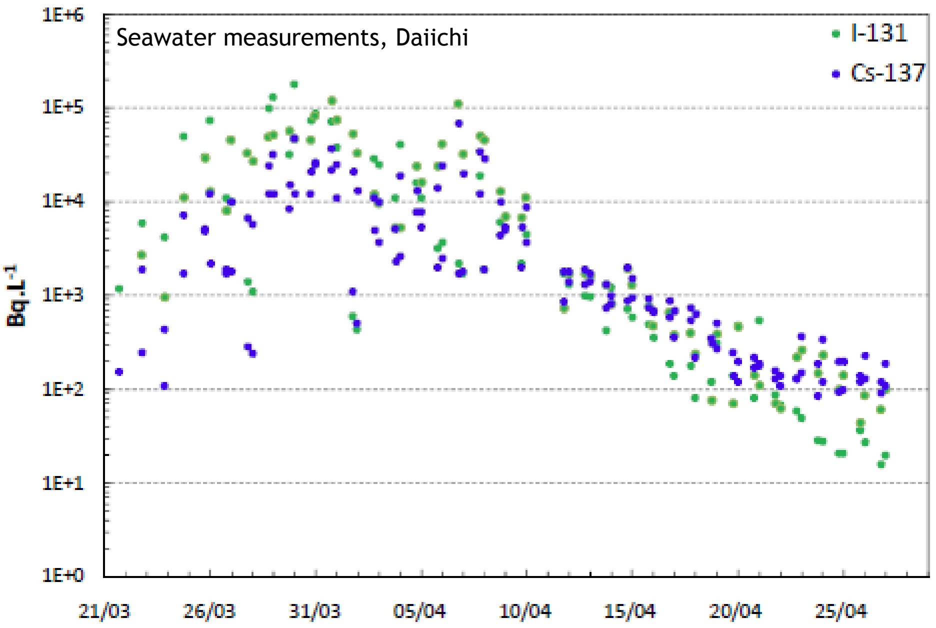


Figure 1 ***Evolution of concentrations of iodine 131 (131I) and caesium 137 (137Cs) in the seawater less than 500 m from the Fukushima Daiichi power plant.***

1.2. Atmospheric fall-out onto the surface of the sea

Between 12th and 23rd March, atmospheric radioactive releases caused by the explosions and pressure venting of the reactor containments in the Fukushima Daiichi nuclear power plant were dispersed long distances. Some of the radionuclides contained in the plume fell back to the surface of the sea, leading to diffuse pollution of the surface water up to tens of kilometres from the nuclear power plant.

The estimate of the atmospheric deposits on the surface of the sea calculated by IRSN is about 10^{+16} Bq of caesium 137 (10 million billion becquerels). This estimate is currently being reviewed on the basis of the available measurements.

Figure 2 shows the evolution of the concentrations in the seawater 30 km out to sea. The concentrations measured before 30th March were primarily due to atmospheric deposits. They vary from 2 to 27 Bq/L for caesium 137 and 3 to 57 Bq/L for iodine131. After falling to a minimum on 30th March, the concentrations in the seawater rose during the first half of April. This was caused either by the arrival of masses of water polluted by large scale atmospheric fall-out, or the dispersion offshore of liquid radioactive releases from the plant.

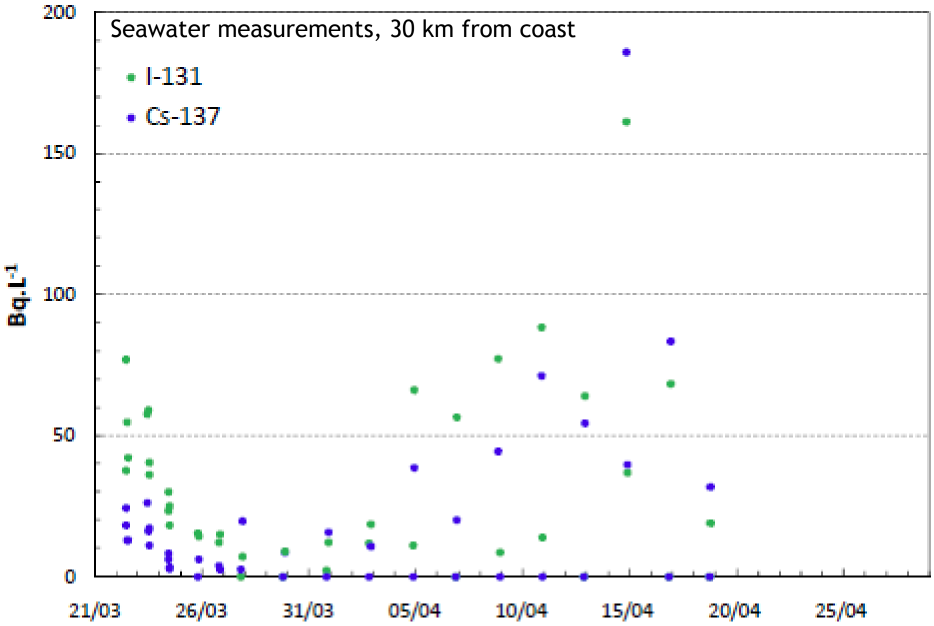


Figure 2 Evolution of concentrations of iodine 131 (131I) and caesium 137 (137Cs) in the seawater 30 km from the coast, off the Fukushima Daiichi power plant.

1.3. Transport of radioactive pollution by leaching of contaminated soil

The radioactive deposits that occurred during the dispersion of the atmospheric releases from the Fukushima Daiichi plant can be partially leached by rainwater and then transported to the sea by run-off. The contaminated land areas thus drained can represent several thousand km2. The measurements currently available do not enable us to differentiate between these diffuse inputs and the other sources of radioactive pollution. This input will probably be detectable several months after the accident, when the concentrations in the coastal waters have decreased.

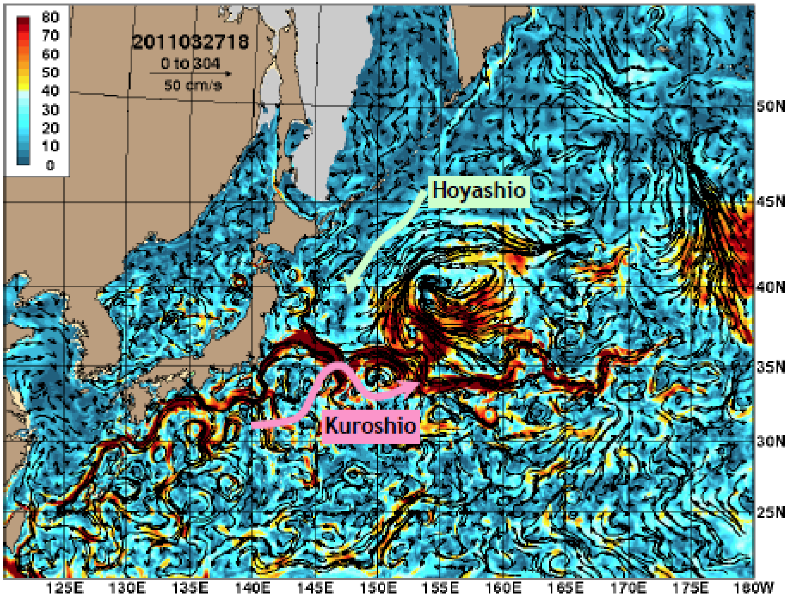
2. DISPERSION AT SEA OF RADIOACTIVE POLLUTANTS

2.1. Ocean currents off the Japanese coasts

The Fukushima power plant is located on the coast of the island of Honshu, more than 200 km north-east of Tokyo. The coast runs north-south, facing the Pacific Ocean. The seabed shelves off gently to a depth of 200m, 50 km from the coast and then drops suddenly to more than 5000 m, about a hundred kilometres offshore.

In the coastal zone, the currents are generated by the tide, wind and the general Pacific circulation. In the short-term, the tidal effect is predominant. It moves the masses of water with a rapid alternating motion along the coast towards the north and towards the south. The wind influences the circulation of surface waters.

The general large-scale circulation is the result of the interaction between the Kuroshio oceanic current, which comes from the south and runs along the coasts of Japan, and the less powerful Hoyashio current, which comes from the north (0). The intensity and scale of the Kuroshio are comparable to those of the Gulf Stream. The coastal waters in the vicinity of the Fukushima Daiichi plant are situated in the zone where these two currents interact, creating variable gyratory currents. It is these currents which will be determining factors in the medium-term dispersion of the radioactive pollution.



Speed/currents layer 1 Mar 27, 2011 00Z 00Z 90.9

Figure 3 Surface currents in the north-west Pacific (http://www.hycom.org/)

The considerable sea depths off the coast and the weak currents lead to stratification of the masses of water. The radioactive substances are mixed in a surface layer 20 to 50 metres thick, near the coast. This layer can reach 100 metres in thickness further offshore. Dispersion of the soluble fraction of the radionuclides will mainly take place in the surface layer. The fraction of radionuclides associated with solid particles could be transported to the bottom of the sea by sedimentation.

2.2. Charts showing the distribution of caesium 137 in the seawater

The concentrations of caesium 137 are representative of the dispersion of all the radionuclides present at sea; the following charts were produced from the results of seawater measurements published by the Japanese authorities.

The spatial distribution and the time breakdown of the caesium 137 concentrations are reproduced on two charts (zone A and zone B). Zone A (figure 4) corresponds to the monitoring stations located close to the Fukushima plant (between latitudes 37.18°N and 37.55°N). Zone B (figure 5) covers a larger marine area (between latitudes 36.9°N and 37.9°N).

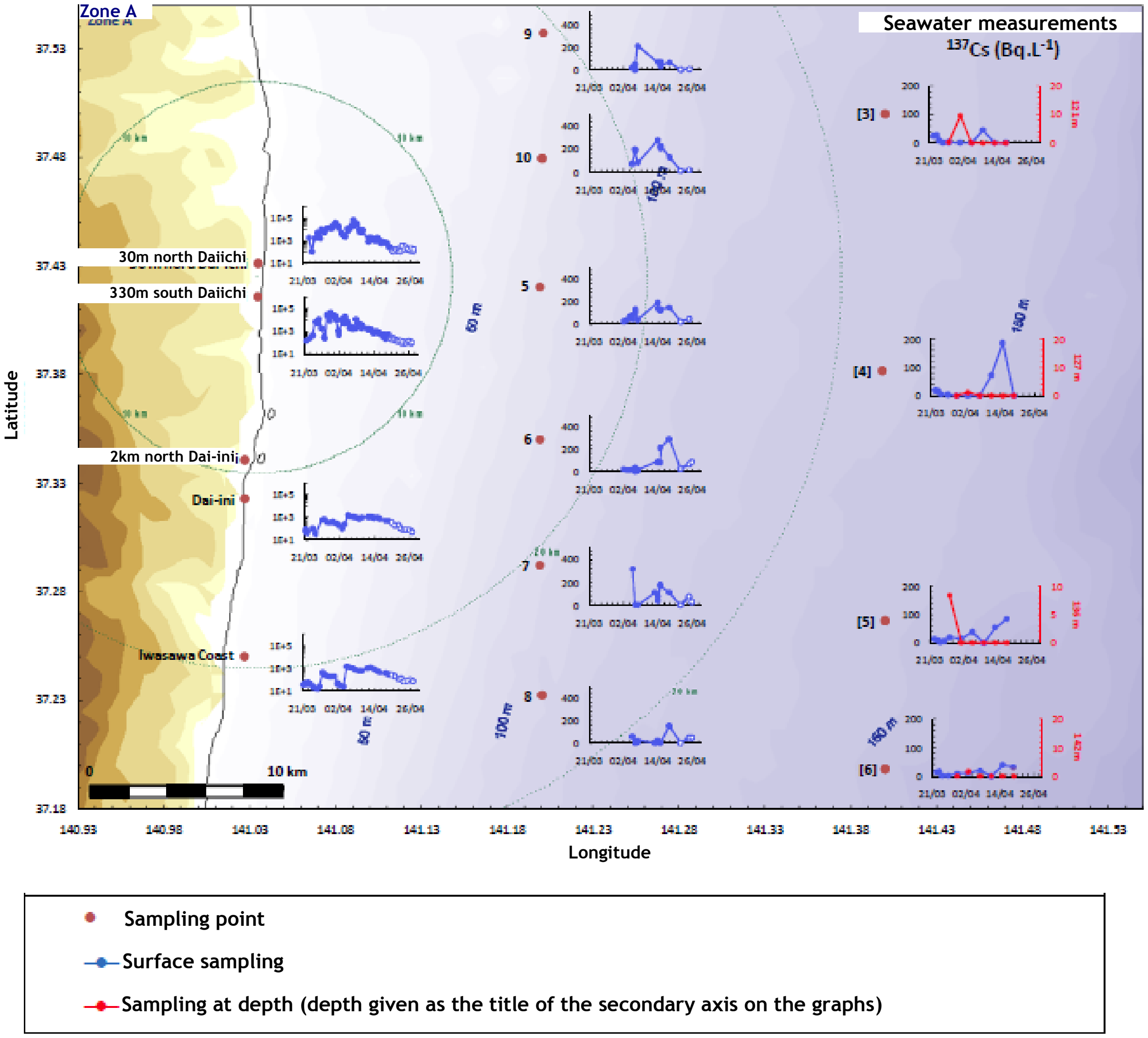


Figure 4 Evolution of concentrations of caesium 137 (137Cs) in the seawater on 14 sampling sites close to the power plant (zone A); all the concentrations are in Bq/L.

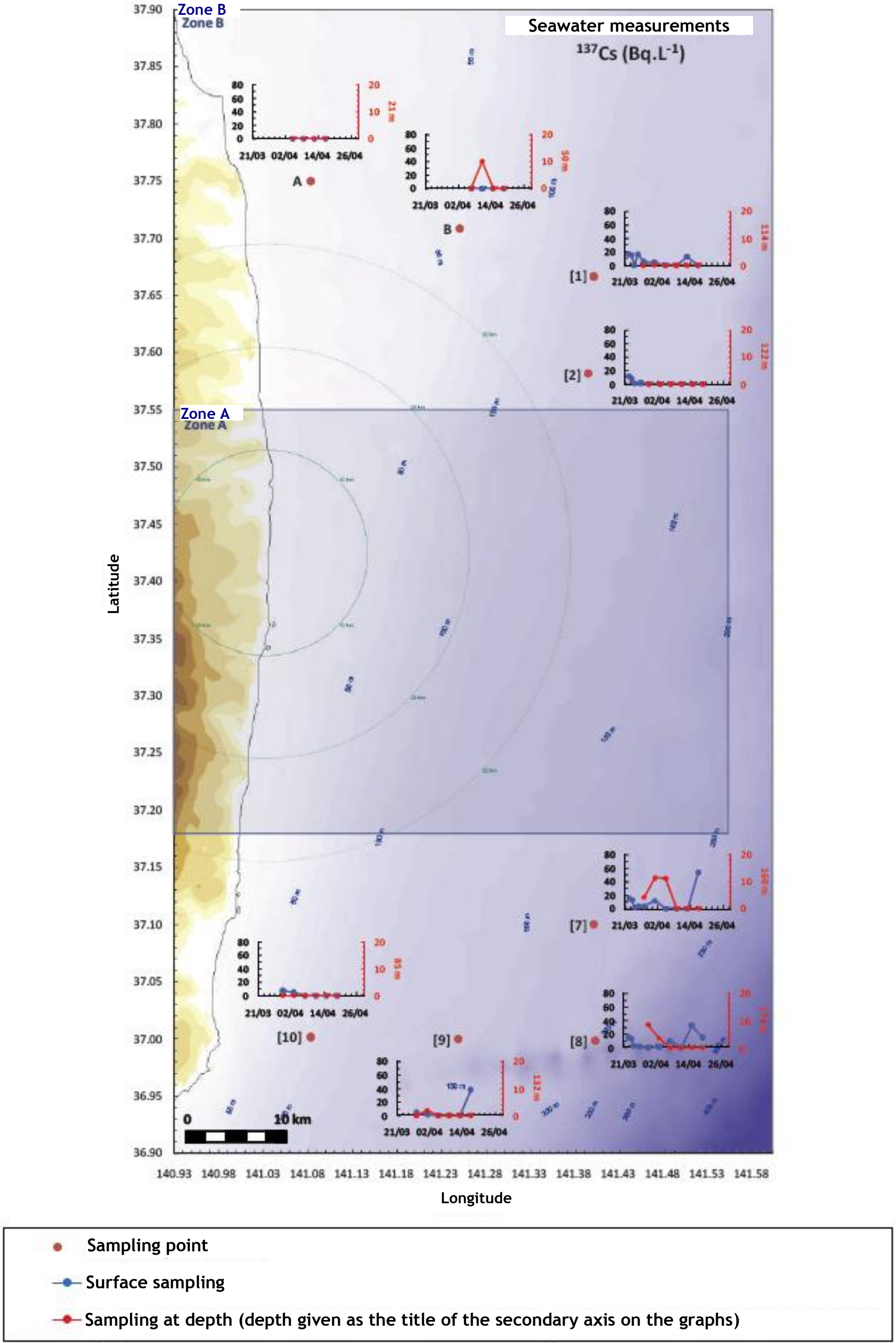


Figure 5 Evolution of concentrations of caesium 137 (137Cs) in the seawater on the 8 sites furthest from the power plant (zone B); all the concentrations are in Bq/L.

In zone A, four zones were identified according to the contamination levels initially measured:

- less than 500 metres from the Fukushima Daiichi power plant;
- coastal zone to the south;
- 15 km off the coast;
- 30 km off the coast.

During the first half of April, the concentrations measured at sea varied considerably between these different zones, by a factor of about ten from one zone to another (more than 50 000 Bq/L at 500 metres from the plant, 10 to 50 Bq/L at 30 km). On 27th April, the concentrations measured along the coast were similar from one location to another, and less than 100 Bq/L of caesium 137, except for the readings close to the plant, which were twice as high.

The concentrations recorded offshore in zone A were lower, but of the same order of magnitude as close to the coast, indicating a homogenisation of the radioactive products.

The chart in figure 6 represents the caesium 137 concentration iso-values obtained from the mean values of the measurement results over the period from 20th to 27th April 2011. It shows persistence of a concentration gradient from the coast to offshore, but one that is lower than that observed in the previous period. Based on these data and the simplifying hypotheses used (uniform concentration for the first 50 metres of depth, uniform gradient from the coast to offshore), the quantities introduced into the sea following the accident can be estimated at 5.7x10+15 Bq for caesium 137 and 3.3x10+15 Bq for iodine131. These results are consistent with the estimates for direct release to sea of caesium 137 produced by the IRSN from the results of the measurements taken on the contaminated waters present on the nuclear site (see chapter 1.2 above).

Eventually, assuming that the caesium 137 released into the sea during the accident is diluted in the seawater surface layer (between 0 and 100 metres depth) and over a surface of about 5000 x 5000 km (corresponding approximately to the surface area of the North Pacific), the concentration could be about 0.002 Bq/L, in other words a level comparable to that observed in the seawater of the Japanese coast before the accident. Although measurable using present techniques, this level of concentration would represent less than one 5000th of the concentrations in seawater of potassium 40 (12 Bq/L), a naturally occurring radionuclide permanently present in water.

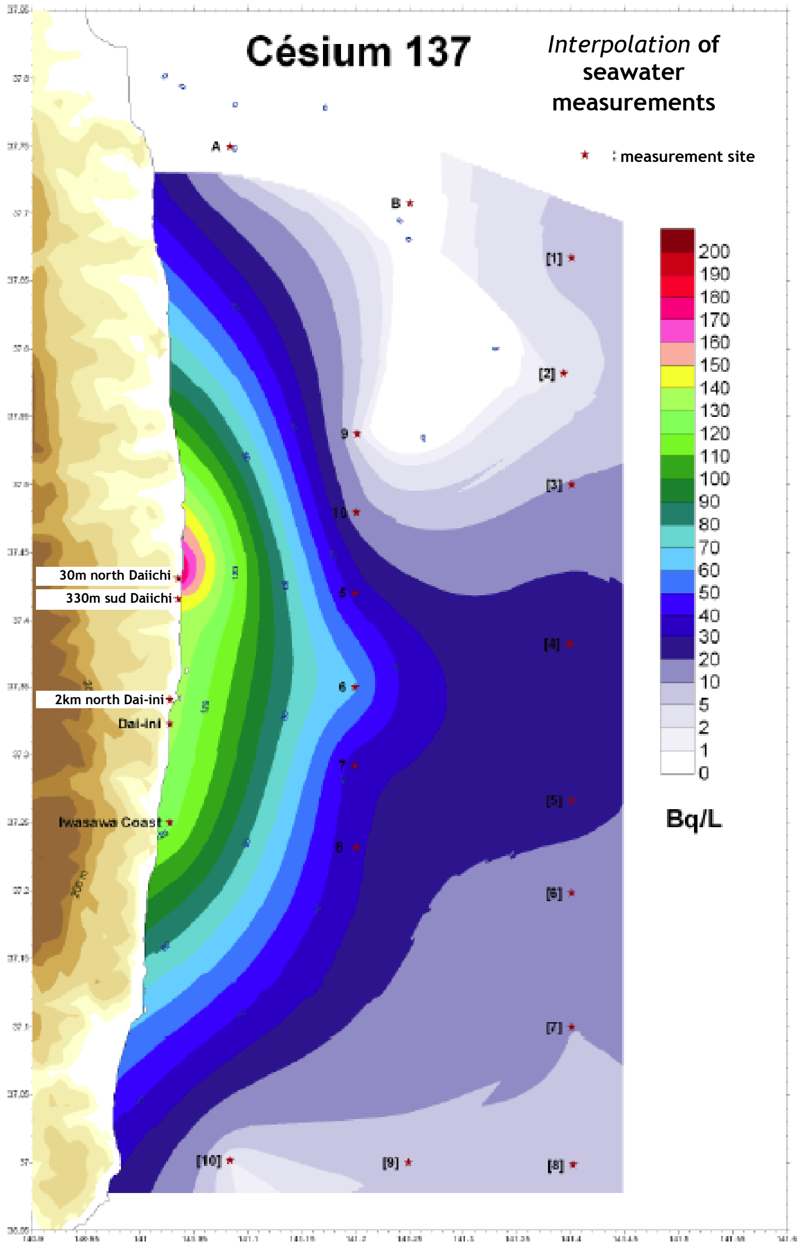


Figure 6 Chart showing the concentration of caesium 137 in the seawater (in Bq/L) produced by interpolating the seawater measurement results (mean values between 20th and 27th April 2011) off the Fukushima Daiichi power plant.

2.3. Simulations of dispersion

The dispersion at sea of this radioactive pollution was simulated using hydrodynamic models by a CNRS laboratory in Toulouse (Sirocco, figure 7), and by a Japanese institute (figure 8). They reflect the complexity of the circulation of masses of water in this region.

The turbulent structures which mix the Kuroshio and Hoyashio currents north of the 36th parallel are complex and unstable. In the coming months, they will no doubt lead to a homogenisation of the concentrations over a width of about 100 km north of the Kuroshio (figure 3) gradually taking the contaminated plume out to the centre of the North Pacific.

Validation of the Sirocco calculations less than 50 km from the plant were carried out using the available measurements.

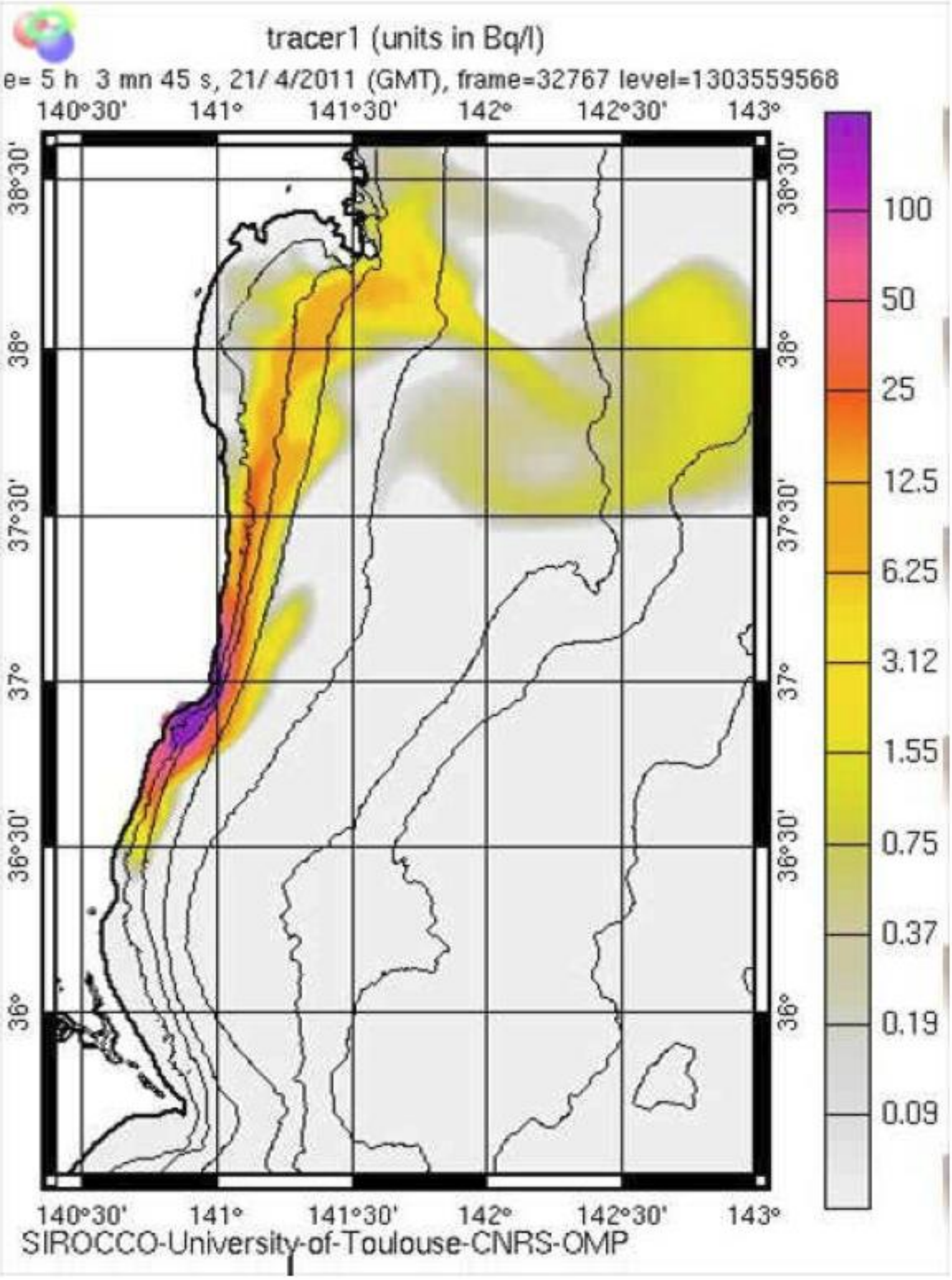


Figure 7 Simulation of the dispersion at sea of the liquid releases of caesium 137 dated 21st April

Source: CNRS and University of Toulouse

[*http://sirocco.omp.obs-mip.fr/outils/Symphonie/Produits/Japan/SymphoniePreviJapan.htm*](http://sirocco.omp.obs-mip.fr/outils/Symphonie/Produits/Japan/SymphoniePreviJapan.htm)

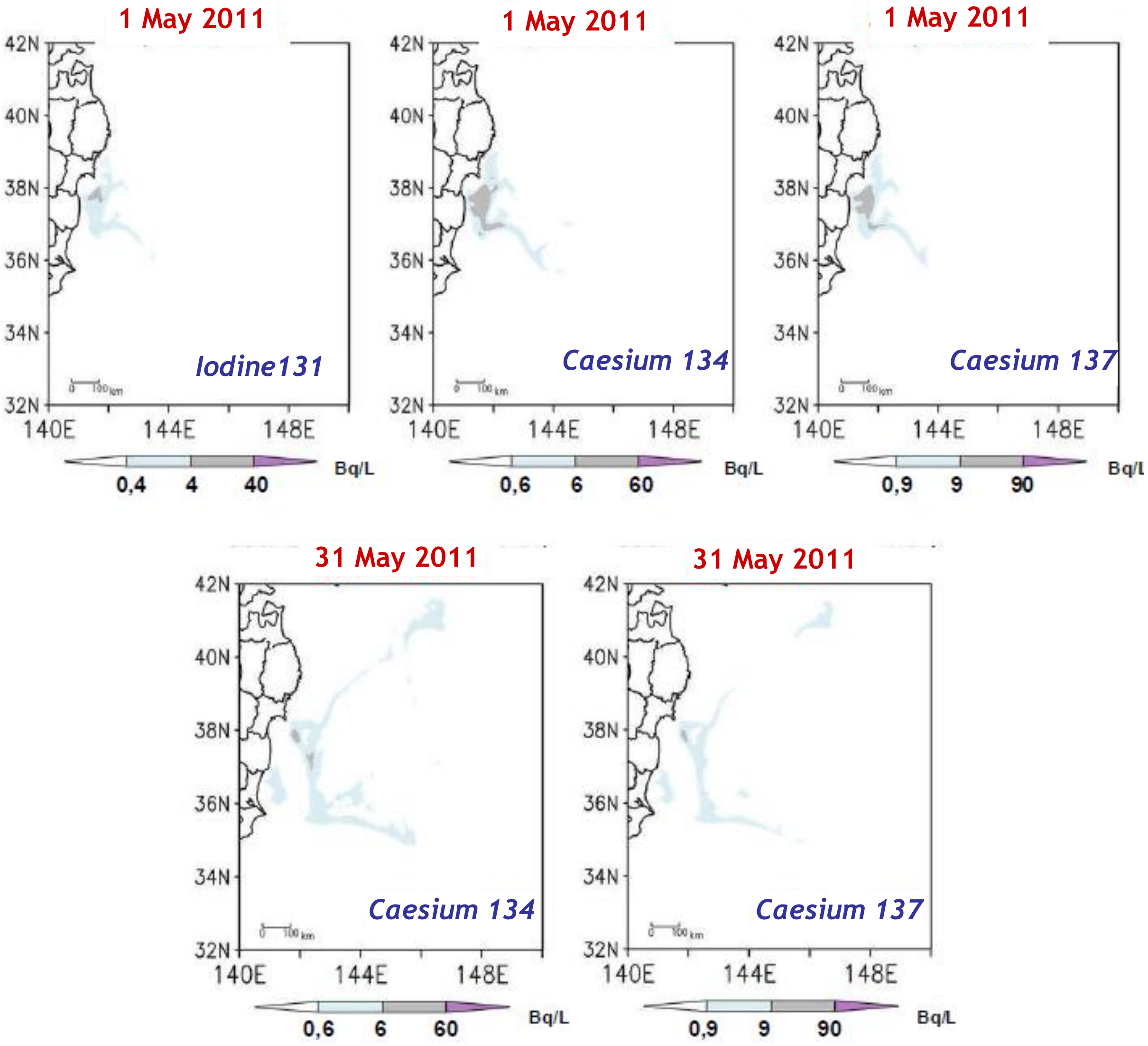


Figure 8 Simulation of the dispersion at sea of iodine131, caesium 134 and caesium 137. For iodine131, the dispersion forecasts as at 31 May 2011 give results less than 0.4 Bq/L. Source: JCOPE2 model developed by the Japan Agency for Marine-Earth Science and Technology (JAMSTEC), http://www.mext.go.jp/english/incident/1305758.htm

3. IMPACT OF RADIOACTIVE POLLUTION ON LIVING SPECIES

3.1. Concentrations observed in fish

The IRSN has collected and analysed the results of measurements on the species of fish with the highest levels and which is also the best represented among the samples taken in Japan, in other words the Japanese sand-eel (*Ammodytes personatus, figure* 9). This fish is caught locally on a seasonal basis (January to April).

Figure 9 Japanese sand-eels (Ammodytes personatus (Girard)). (a) larvae, (b) juveniles.

The analyses were taken on fish landed in 6 ports, all located south of Fukushima (figure 10). In the absence of any precise information, the IRSN considers that the values supplied correspond to the whole fish, not dried.

Figure 10 Location of ports where the Japanese sand-eels were sampled.

The highest levels were reached on 13th April in individuals landed in the port of Iwaki in Fukushima prefecture: 12000 Bq/kg of iodine131, 6200 Bq/kg of caesium 134 and 6300 Bq/kg of caesium 137 (table 1). However, individuals of the same species landed in this fishing port on the same day showed far lower values; 300, 190 and 200 Bq/kg respectively. This type of difference was also observed on the two samples of 18th April from the same port. This variability can be explained by differences in the fishing locations, even if the catch is then landed in the same port. The IRSN is not familiar with these precise fishing locations.

Consistently with the records of the releases, there would seem to be trend towards a fall in the iodine131 activity in the latest samples. This is not the case for caesium owing to its longer radioactive half-life.

Table 1 Activity levels measured in Japanese sand-eels (in bold: the results exceeding the maximum permitted levels[1] of 2000 Bq/kg for iodine and 500 Bq/kg for the sum of caesium 134 plus caesium 137)

[1] MPL: Commission Implementing regulation (EU) n°351/2011 of 11th April 2011

Prefecture	Port of offloading	Date of sample	^{131}I	^{134}C	^{137}Cs	Total Cs
			Bq/kg fresh*			
Fukushima	Port of Iwaki	07/04/2011	1700	280	290	**570**
Fukushima	Port of Iwaki	07/04/2011	1200	240	260	500
Fukushima	Port of Iwaki	07/04/2011	1500	240	240	490
Fukushima	Port of Iwaki	07/04/2011	1100	240	240	480
Fukushima	Port of Iwaki	13/04/2011	**12000**	**6200**	**6300**	**12500**
Fukushima	Port of Iwaki	13/04/2011	300	190	200	390
Fukushima	Port of Iwaki	18/04/2011	**3900**	**7100**	**7300**	**14400**
Fukushima	Port of Iwaki	18/04/2011	120	160	160	320
Ibaraki	Port of Hiragata	01/04/2011	**4080**			**447**
Ibaraki	Hitachinaka -Port of Isozaki	04/04/2011	600			83
Ibaraki	Port of Hitachinaka	08/04/2011	598			81
Ibaraki	Port of Hitachinaka	13/04/2011	397			66
Ibaraki	Port of Hitachinaka	21/04/2011	180			93
Ibaraki	Port of Hitachinaka	29/04/2011	53			51
Ibaraki	Port of Kitaibaraki	04/04/2011	1700			**526**
Ibaraki	Port of Kitaibaraki	11/04/2011	**2300**			420
Ibaraki	Port of Kitaibaraki	11/04/2011	52			170
Ibaraki	Port of Kitaibaraki	13/04/2011	1600			357
Ibaraki	Port of Kitaibaraki	14/04/2011	520			419
Ibaraki	Port of Kitaibaraki	28/04/2011	430			**1129**
Ibaraki	Port of Kitaibaraki	29/04/2011	420			**1374**
Ibaraki	Port of Takahagi	14/04/2011	830			348
Ibaraki	Port of Takahagi	21/04/2011	670			254
Ibaraki	Port of Takahagi	28/04/2011	330			404

**In the absence of any specific details, values considered to be measured on whole, fresh fish*

For certain samples, the available information only concerns the total activity of the two caesium isotopes (134+137); however, the ratio between the activity of these two isotopes is close to 1 in the releases, so one can consider that these two radionuclides have approximately the same activity levels in the environment.

Iodine 131 and caesium 134 and 137 were detected in all the individuals of this species, which should no doubt be attributed to their lifestyle. They are currently at the juvenile stage. Their contamination comes both from direct exposure to contaminated water, but also the consumption of contaminated food consisting of phytoplankton and zooplankton. The geographical location is also a factor to be considered when interpreting these levels, because this species is caught close to the coast.

The other values concern other fishery products (various fish such as flat fish, mackerel, anchovies, sea bass as well as shellfish) and are generally below 200 Bq/kg for iodine 131, and 100 Bq/kg for the two isotopes of caesium.

Information about the precise fishing locations would be crucial for a pertinent interpretation of the measurements obtained from the fish. This information is no doubt not available because the primary objective of these measurements is protection of the populations, and therefore the monitoring of the radioactivity in the catches landed.

Generally speaking, fish will be the best medium and long-term indicators of caesium contamination. Monitoring of other sea products of course remains necessary in the short-term for the other radionuclides released (iodine 131 for example should be monitored in algae, in particular brown algae, until it disappears through radioactive decay). It should be remembered that caesium reaches high levels of concentration in fish (the average caesium concentration of the edible part – the muscles - is 100 times higher than that of the water) and has a tendency to increase with the trophic level occupied by the species. Consequently, in the short-term, the highest concentrations will be found more in the species situated in the first part of the food chain (such as the Japanese sand-eel) which are the most quickly exposed to contamination through food consumption. Then, once transfer to the various parts of the trophic networks is effective, the predators at higher trophic levels (whiting, tuna, etc.) should show the highest levels.

3.2. *Expected concentrations based on the seawater measurements*

The exchange of radionuclides from the seawater to living species and sediments has been the subject of considerable scientific work. This work established coefficients which can in principle, based on the concentration of a given radionuclide in seawater, be used to calculate the concentration of this radionuclide induced in a living species over a lengthy period of exposure (several months). These coefficients, called concentration factors for living species and distribution coefficients for sediments, are known for most of the radionuclides and groups of species. The precision of the calculations made with these coefficients is about factor ten owing to the numerous potential environmental influences. Furthermore, the estimates thus obtained are only valid for prolonged exposure to the same level of concentration in the water (equilibrium time), which is not the case for an accidental release of limited duration, leading to concentrations in the water which are constantly changing.

The caesium concentration factor established for fish (consumable part) is 100 (IAEA Technical Report n°422). This means that seawater with a concentration of radioactive caesium (134+137) exceeding 5 Bq/L could lead to contamination of fish exceeding the maximum permitted level for commercial sale for human consumption (500 Bq/kg – Commission Implementing Regulation (EU) n°351/2011 of 11th April 2011). Assuming that the caesium 137 concentrations given in figure 6 remain constant for several months, it is possible to estimate the zones within which the permanently resident fish would be liable to be contaminated with concentrations exceeding this maximum permitted level (chart in figure 11). This chart is purely indicative and these estimates are no doubt upper bounds because they only take account of the concentrations measured up until 27th April, assuming that they remain constant for several months. In reality, they are liable to change rapidly owing to transport and dilution. Moreover, the fish can travel long distances and pass through this zone in too short a time to reach equilibrium with the concentrations in the seawater.

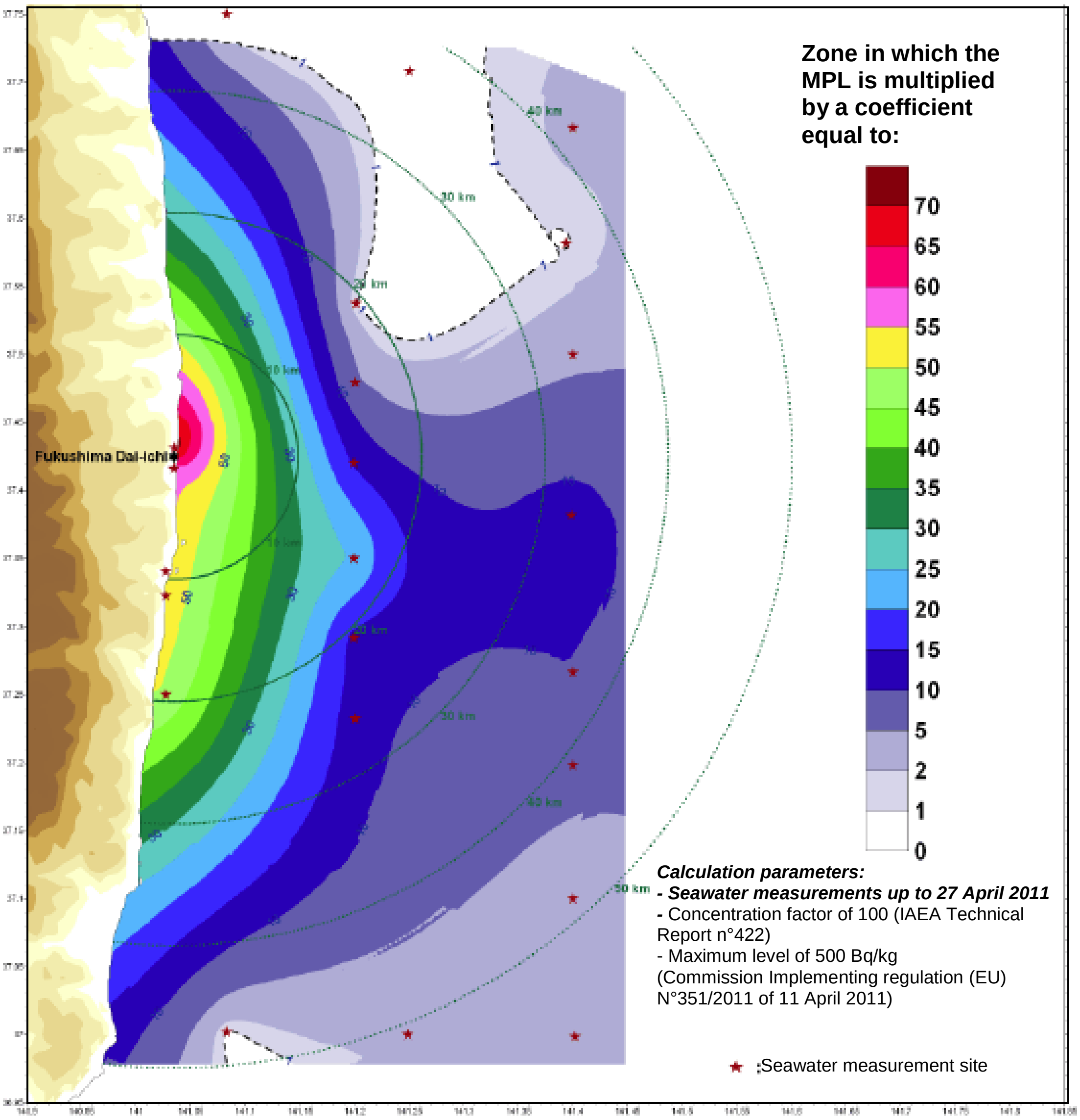


Figure 11 Theoretical estimation of the levels in excess of the maximum permitted concentration (MPL = 500 Bq/kg fresh) for commercial sale of fish, assuming that the caesium 134 and 137 concentrations observed during the last week of April are maintained for several months. The dotted black line marks out the zone in which the MPL would be exceeded and the coloured zones correspond to the various multiples of the MPL.

Assuming that all the radioactive caesium (134 and 137) released into the sea during the accident (estimate for both of these two radionuclides together: 11.4x1015 Bq; see section 2.2 above) continues to disperse and that its concentration becomes homogeneous in the first 100 metres of water and in all directions of the marine environment from the Fukushima Daiichi plant, the distance at which levels exceeding the maximum permitted level could be observed would be about 120 km (figure 12).

This theoretical estimate presupposes that the fish species and their food resources remain permanently in this zone. It can only be indicative and is limited by a lack of precise knowledge of the quantities of caesium actually released and does not take account of the actual dispersion of the masses of water on the surface and at depth, nor the lifestyle of the fish.

Figure 12 Marine zone potentially concerned by levels in excess of the maximum permitted level (caesium 134 and 137) for commercial sale of fish, assuming continued dispersion of caesium in all directions and down to a depth of 100.

4. RADIONUCLIDES IN THE SEDIMENTS

Samples of sediment collected at 15 km and 20 km from the plant show concentrations close to 1400 Bq/kg for caesium 137 and 1300 Bq/kg for caesium 134, while the iodine 131 levels are around 190 Bq/kg (table 2). It appears that these samples were taken at between 20 and 30 m depth and at a distance of 3 km from the coast. Nonetheless, these values are purely for information, because if the volume concentrations currently measured in the seawater (about 100 Bq/L) are maintained, then concentrations of several hundred thousand Bq/kg could be reached in the medium-term in the sediments. These values are heavily dependent on the nature of the sediments sampled: the finer the sediment, the greater the exchange surface with the seawater and the higher the binding capacity. The measurement results also depend on the thickness of sediment samples, as the surface layer of the sediment in direct contact with the seawater is more contaminated than the deeper levels.

Tableau 2 – Activity measured in the sediment samples taken on 29/04/2011, in Bq/kg (TEPCO) the values are assumed to be given per kg of fresh sediment.

Sampling site	3 km off Okada	3 km off Iwasawa
^{131}I	190	98
^{137}Cs	1300	1200
^{134}Cs	1400	1200