Situation of the contamination of the marine environment in 2015, following the Fukushima Daiichi nuclear power plant accident

2015 Summary

The evolution in 2015 of the contamination of the coastal marine environment around the Fukushima power nuclear plant (NPP) is characterised by a relative stability in levels: no detectable reduction in caesium-137 sediment activity and a very slow decline in seawater activity, fuelled by probable releases into the sea by the facilities due to leaks existing on reactors that remain in the cooling phase, permanent input from leaching and soil drainage, as well as the re-suspension of sedimentary particles and the possible desorption of associated[1] radiocaesiums. Over 200 km from the NPP, radiocaesium concentrations have fallen to levels close to those observed prior to the accident. Logically, the organisms living close to coastal sea beds are those showing the highest levels of - and slowest decrease in - activity. However since 2014, outside the Fukushima prefecture, radiocaesium levels measured in marine foodstuffs no longer exceed the marketable limit set by the Japanese health authorities (100 Bq/kg). Within the Fukushima prefecture, this limit is still exceeded on occasions.

1. Estimation of the marine source-term

The estimation of the direct releases ending up in the sea in April 2011 remains a subject of debate between various authors. Numerous evaluations have been carried out concerning ^{137}Cs inputs: 3.6 – 3.7 PBq (Tsumune et al., 2013); 5.1 – 5.5 (Estournel et al., 2012); 12 – 41 PBq (Bailly Du Bois et al., 2012; 2014); 11 - 16 PBq (Charette et al., 2013; Lai et al., 2013; Rypina et al., 2013; Smith et al., 2014). The atmospheric fallout over the sea represented similar quantities (from 0.9 to 5.5 PBq near the coast), but with an extensive distribution over the North Pacific from the first month after the accident (Aoyama et al., 2013). For the marine environment, all of these ^{137}Cs inputs are comparable or superior to those that affected the seas following the atmospheric fallout of the Chernobyl accident (15 - 20 PBq). The difference mainly lies with the direct inputs at sea, which led to particularly significant concentrations close to the outfall.

[1] These radiocaesiums are caesium-134 and caesium-137.

As far as ^{90}Sr is concerned, due to the fact that the number of measurements were far lower than for caesium-137, estimations of the source-term are essentially based on the $^{90}Sr/^{137}Cs$ ratio. As this ratio evolved significantly in 2011, major uncertainties exist. Povinec et al. (2012) estimate the total flow from direct releases at around 1 PBq. Despite major fluctuations between one measurement and the next, the evolution of the $^{90}Sr/^{137}Cs$ ratio measured close to the facility indicates that the residual releases since 2012 of these two radionuclides are, on average, of the same order of magnitude.

2. Contamination of seawater

The contamination of seawater following the Fukushima accident resulted from atmospheric deposits in an initial phase and then from massive direct releases linked to the pouring of seawater then fresh water onto the damaged reactors for cooling purposes. Activity of several tens of thousands of Bq/L reached in the seawater less than 2 km from the site during the first weeks following the accident was quickly diluted by the interaction between the two main currents that circulate off Japan's eastern coast: the Kuroshio, coming from the south, following the coastline, and the lesser Oyashio, coming from the north (figure 1), which led to a rapid reduction in the contamination of the water column. Nevertheless, during this period, part of the radiocaesium was absorbed by suspended matter and sediments, leading to the contamination of the latter.

Since then, within a 30 km radius of the site, water activity has remained globally stable, due to the effect of three input categories that are nonetheless difficult to quantify: input linked to the NPP site (leaks existing on the damaged reactors that remain in the cooling phase (approximately 320 m^3/day of fresh water is injected into the reactor cores, whose outer walls are no longer watertight), and drainage of the highly-contaminated soils at the site), leaching and drainage from the catchment basins whose soils are contaminated following the radioactive deposits[2] (this process is extremely effective, notably during typhoons) as well as the re-suspension of sedimentary particles and the desorption of associated radiocaesiums.

[2] For more information, the reader can consult the following information sheet: "How are radioactive deposits redistributed within catchment basins in post-accident situations? The lessons learned from the Chernobyl and Fukushima accidents".

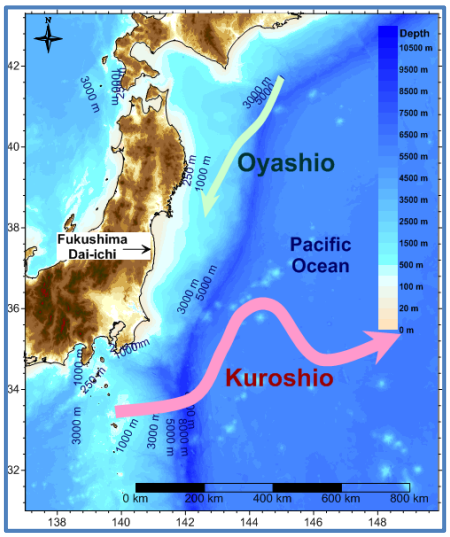

Figure 1: Depth and surface currents facing the Japanese east coasts

Since the summer of 2012, caesium-137 seawater concentrations are therefore, overall, relatively stable within the 30 km radius around the site, with a slight downward trend (figures 2 and 3). Less than 2 km from the NPP (figure 2), concentrations lie within a range of 0.01 to 5 $Bq.L^{-1}$. They are around 10,000 times lower than those detected in April 2011. Between the zone close to the facility and a distance of 30 km along the coast, caesium-137 concentrations lie within a range of 0.007 to 0.2 $Bq.L^{-1}$. With the renewal of marine water being comparable from one year to the next, the stability over the time of these levels can only be explained by a continuation of ^{137}Cs inputs in the seawater in accordance with the sources indicated previously. It is not possible to precisely determine the respective share of these contributions. Nevertheless, the difference in concentrations by approximately factor 10 to 100 between the zone close to the site (less than 2 km) and zones further away (2 to 30 km and 30 to 200 km respectively) (figure 4) indicates that these inputs remain significant.

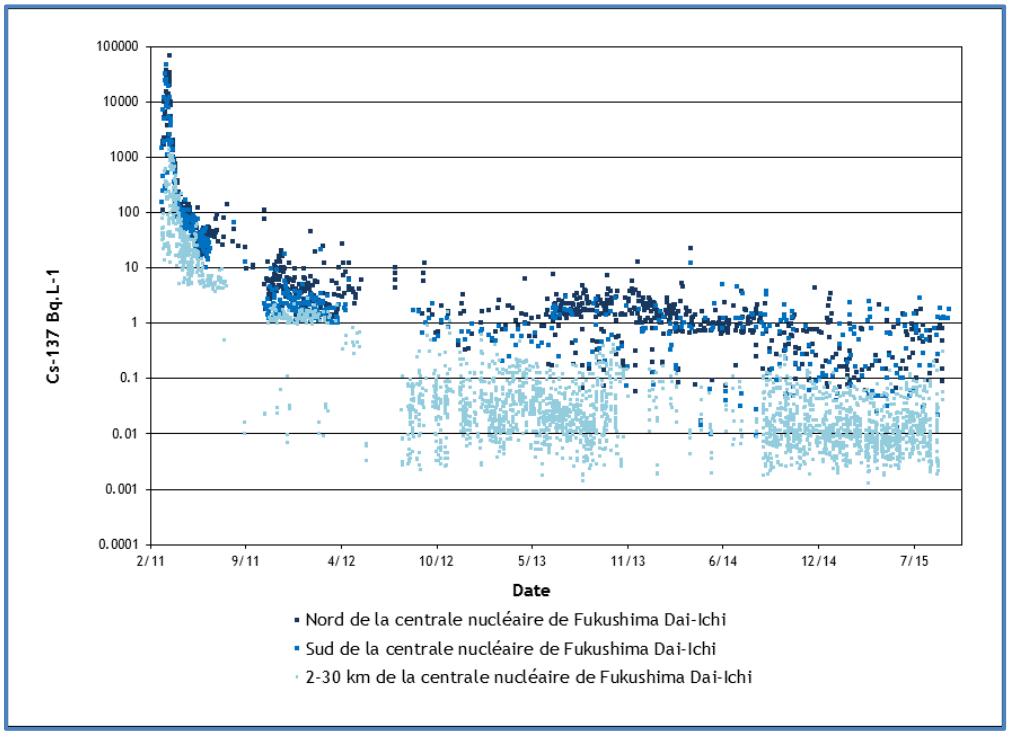

Figure 2: Evolution over time of caesium-137 seawater activity along the coast for distances from the nuclear power plant of: less than 2 km (dark blue, excluding the port zone) and between 2 and 30 km (light blue).

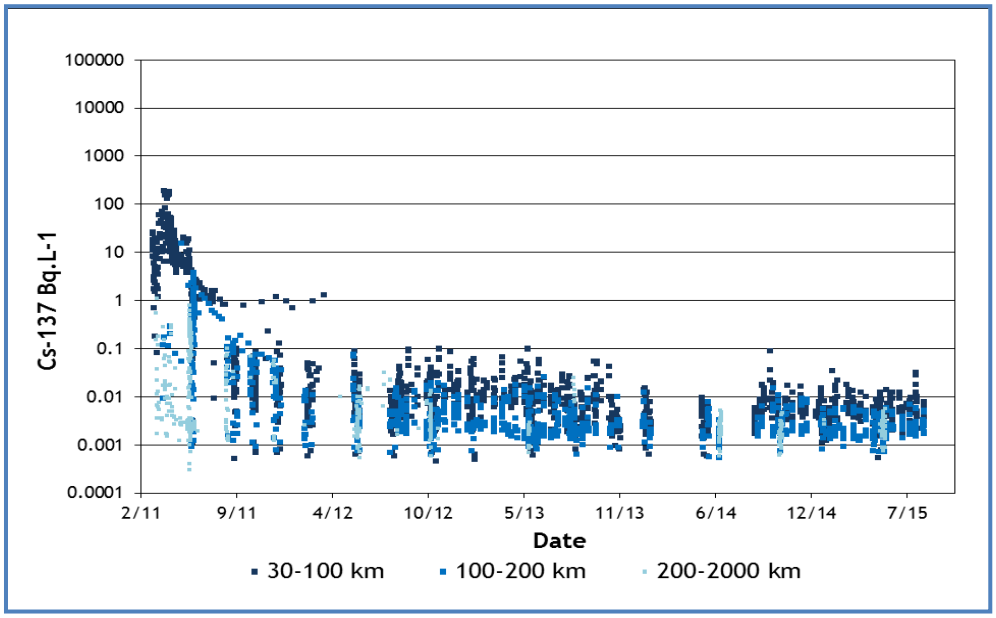

Figure 3: Evolution over time of caesium-137 seawater activity for distances from the damaged nuclear power plant of 30 to 100 km (dark blue), 100 to 200 km (medium blue) and over 200 km (light blue).

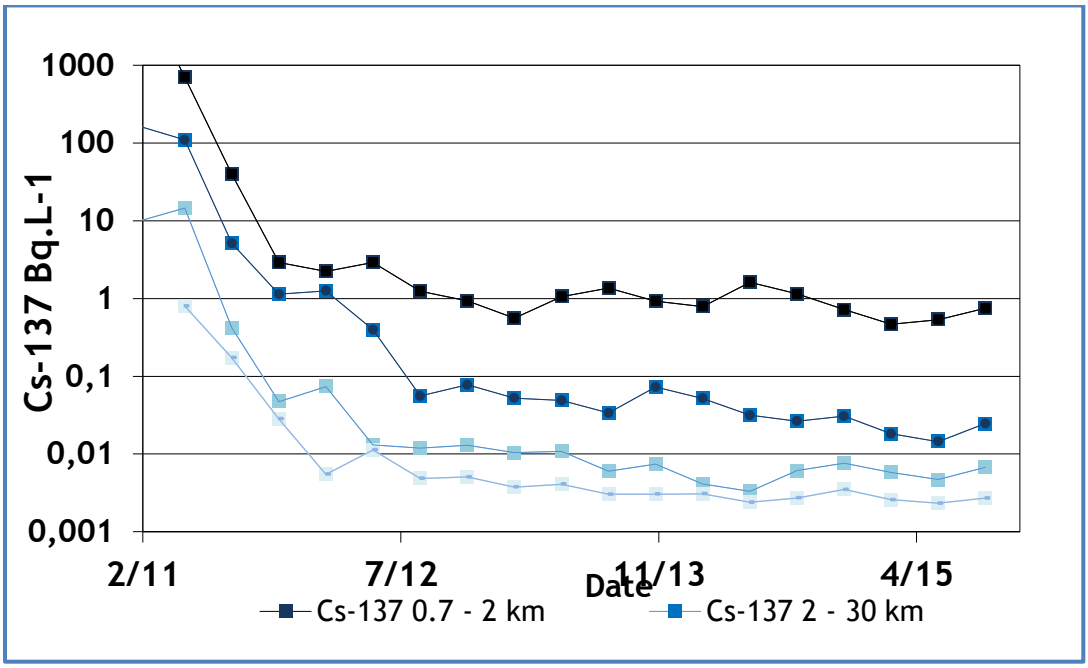

Figure 4: Evolution of average quarterly caesium-137 seawater activity for distances from the damaged nuclear power plant of 0 to 2 km, 30 to 100 km (dark blue) and 100 to 200 km (medium blue) (light blue).

Caesium-137 concentrations measured in 2015 at over 100 km from the nuclear power plant vary between 0.002 and 0.007 $Bq.L^{-1}$. The minimum values correspond to the concentrations measured prior to the accident (0.001-0.002 $Bq.L^{-1}$) that resulted from the atmospheric fallout from aerial nuclear weapons tests. Since 2013, at over 30 km, the maximum values have fallen below those measured in this area of the North Pacific during the 1960s (0.02 $Bq.L^{-1}$ at the peak of this fallout (Povinec *et al.*, 2013)).

The plume marked by the Fukushima releases was notably detected along the American coastline 4,000 km to the east at a rate of 0.001 to 0.005 $Bq.L^{-1}$ according to Smith *et al.* (2014) and in the subtropical zone of the North Pacific, up to 2,000 km to the south of Japan (Kaeriyama *et al.*, 2014; Kumamoto *et al.*, 2015). These plumes were identified as having a depth of between 0 and 500 metres. Works published do not show any marking in the Arctic Ocean, via the Bering Strait.

Radionuclides other than radiocaesiums were measured, associated with the dissolved phase in the water column, but in a far more sporadic manner. These consist mainly of strontium-90 and tritium which, in comparison to radiocaesiums measurements, can reveal different origins. Caesium-137 and 134 were released in similar quantities at the time of the accident, and the results obtained for these two radionuclides are therefore comparable if their respective radioactive decay periods are taken into account (caesium-137: 30 years; caesium-134: 2 years). The concomitant evolution of the Sr-90/Cs-137 ratio close to and further away from the facility indicates a similar origin, coming from the facility area. For the year 2014, tritium marking is comparable to that measured in French rivers (1 - 10 $Bq.L^{-1}$). These results confirm earlier data indicating that tritium releases following the Fukushima accident were very moderate compared to those of caesium-137.

3. Contamination of sediments

With the exception of the port zone to the right of the Fukushima Daiichi site, surface sediment activity shows little evolution between the immediate surroundings of the NPP and up to 80 km away, ranging between around 1 and 1,000 to 5,000 Bq/kg of dry sediment. They also remain constant since 2012 (figure 5) without any significant trend, either upward (which could result from additional inputs) or downward (likely to result from a dispersion of sediments through erosion then lateral transport, or from the burial of the most contaminated particles through bioturbation). The decline linked to the radioactive decay of caesium-137 is not perceptible over the five years given the high variability in concentrations.

Further away, between 80 and 280 km, activity is lower and ranges between approximately 1 and 100 to 500 Bq/kg dry weight.

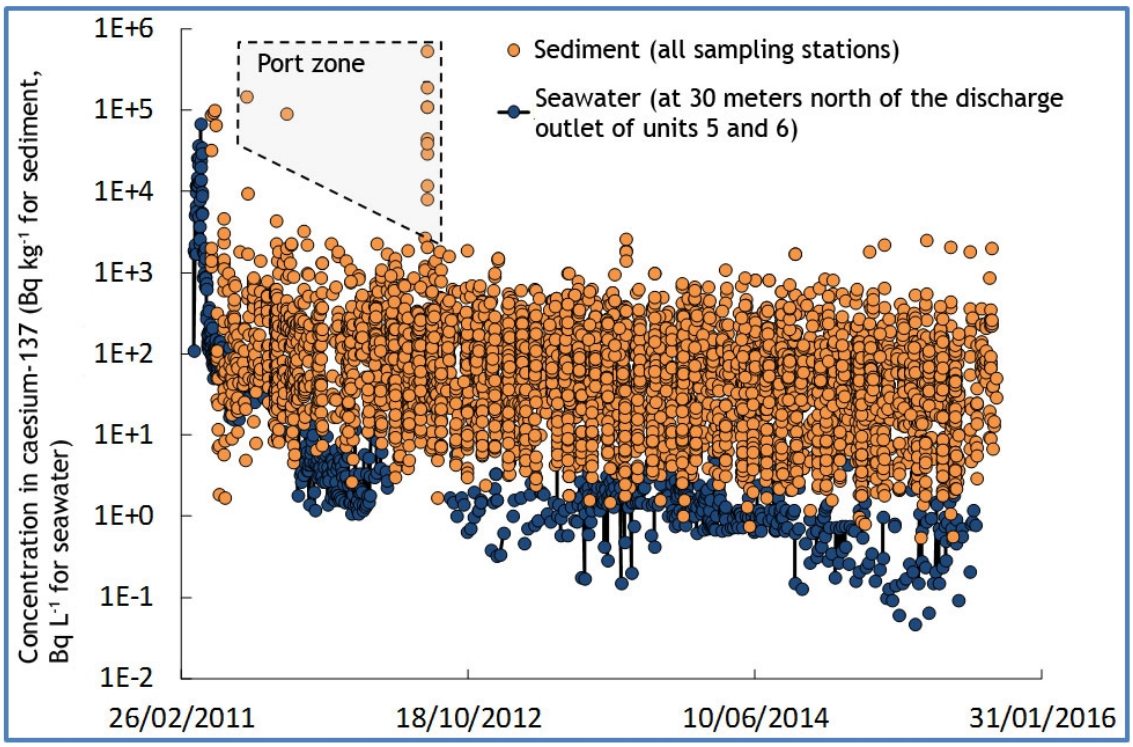

Figure 5: Evolution over time of the caesium-137 concentration in sediments up to 280 km from the coast (samples taken at sea and in the port zone of Fukushima Daiichi) until 27/10/2015; in the absence of information (particularly during the first months following the accident), it is possible that some activity was reported on non-dried matter; data collected by the IRSN Sea Committee (Cellule Mer)

In general, caesium-137 concentrations in sediments are higher to the north of the damaged NPP than to the south. This can be explained by the fact that the contamination of the sediments mainly took place during the first six months following the accident and that, during this period, all of the waters contaminated by caesium-137 would have shifted northwards at the beginning of May 2011, before some returned along the south coast at the end of May 2011.

Although at a regional level (up to 280 km from the coast) the variation range of caesium-137 concentrations is homogeneous, significant local variations in activity were measured by Thornton *et al.* (2013) within a 20 km radius of the NPP using a spectrometer towed over the sediments by a vessel (figure 9). These measurements reveal preferential accumulation zones of contaminated particles related to bathymetry. Zones of activity exceeding average surrounding values by factor 5 to 10 are called "hot spots" (Black and Buesseler, 2014). The concentrations measured in the sediments there reach 5,000 Bq/kg wet weight. The "hot spots" are most often located in areas with a depth of less than 150 m and their characteristic dimensions vary from a metre to around a hundred metres.

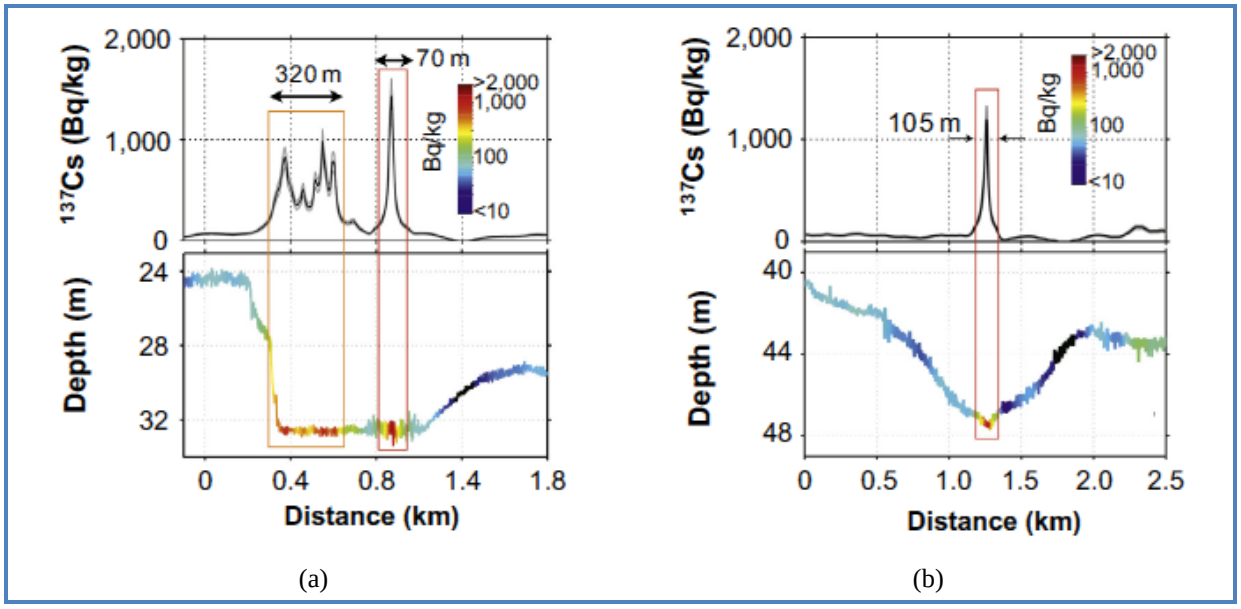

Figure 6: Examples of "hot spots", points of high concentrations of caesium-137 in sediments, corresponding to accumulations in bathymetric "low points", observed within a radius of 20 km around the Fukushima power plant (adapted from Thornton et al., 2013).

Radionuclides other than radiocaesiums are measured on a very occasional basis. Since the accident, strontium-90 concentrations measured vary between 0.045 and 63 Bq/kg dry weight. The average value is 2.3 Bq/kg dry weight.

In the north-western Pacific, the two main sources of plutonium identified to date have been the fallout from aerial nuclear weapons tests. The analysis of a 10 cm long sediment core, sampled in January 2013 at a depth of 300 m and 100 km to the east of the nuclear power plant, suggests the absence of a detectable influence of the Fukushima accident for this radioelement (Bu *et al.*, 2014). Plutonium-239+240 activity measured in surface sediments, of between 0.48 ± 0.01 and 3.53 ± 0.10 Bq/kg dry weight, are close to those measured to the north of Fukushima between April 2008 and June 2011: between 0.37 and 4.1 Bq/kg dry weight. However, due to the high affinity of plutonium for particles and the absence of specific measurements, a marking of sediments located at distances closer to the NPP cannot be excluded.

4. Effects on marine species

Between October 2012 and 30 November 2015, 1,200 samples were taken in the port of the Fukushima nuclear power plant. Thanks to the structures put in place in order to restrict the entry and exit of the port's fish, demersal species (those feeding on the sea bed) were those that mainly remained present in the enclosure. Since the beginning of 2015, radiocaesium concentrations of over 10,000 Bq/kg fresh weight have been measured in three demersal fish species: the jacopever, Japanese black rockfish (or Sebastes cheni) and the brass blotched rockfish. Figure 7 shows the comparative evolutions of the activities of the latter two species sampled either on the coastline or in the port of Fukushima.

Since May 2012, in this coastal zone off the Fukushima prefecture, TEPCO has implemented monitoring of contamination levels in marine organisms at several stations in the coastal zone close to the nuclear power plant. Generally-speaking, stations at sea, over 10 km from the coast, show a decrease in radiocaesium concentrations with, at 28 November 2015, values close to 10 Bq/kg fresh weight (figure 7).

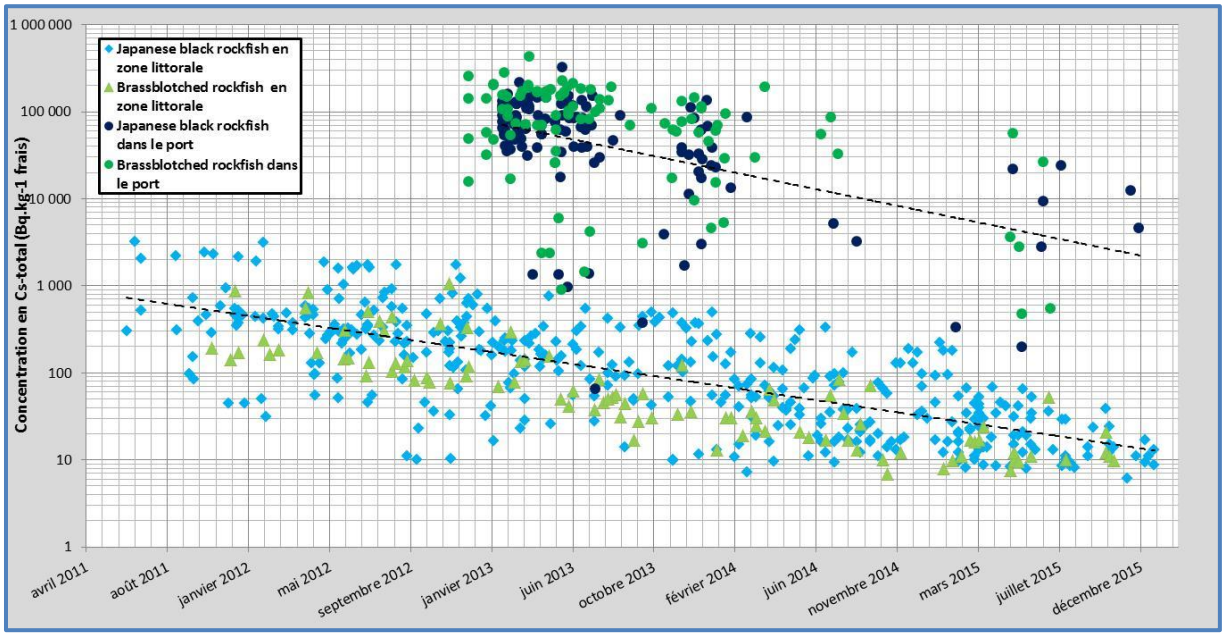

Figure 7: Representation of radiocaesium concentrations in two demersal fish species: Japanese black rockfish (dark blue) and brass blotched rockfish (dark green) inside the port of the nuclear power plant and within a radius of 20 km, in a coastal zone of 20 km (in light blue and light green for each species respectively) (Source TEPCO).

Two coastal stations were sampled to the north of Fukushima: 1 km from the mouth of the Ota river, and 3 km off the coast of Odaka. Despite a steady decline, radiocaesium concentrations on occasions reached values of over 100 Bq/kg fresh weight in 2015 (marketable limit). This content must certainly be linked to inputs from the rivers.

In 2015, radiocaesium concentrations exceeding the 100 Bq/kg fresh weight limit were still observed from time to time. As an example, two of the demersal fish species most sampled by TEPCO, both over time and space, are the Japanese black rockfish (*Sebastes cheni)* and the spotbelly rockfish (or

brass blotched rockfish, *Sebastes pachycephalus*), with 204 and 601 samples respectively. Radiocaesium concentrations in these species (figure 7) enable two trends to be highlighted. Since the start of the sampling carried out by TEPCO (May 2012) in the coastal zone and within a 20 km radius around the NPP, radiocaesium concentration is slowly decreasing in relation to the sediments sampled in this same zone. Contrary to 2012, the variability between measurements at a same date has decreased (figure 7).

The Fukushima accident was originally characterised by a $^{137}Cs/^{134}Cs$ ratio of 1. This ratio increases over time due to the quicker decay of ^{134}Cs than ^{137}Cs; it is currently around 4. This ratio is measured in all samples taken by TEPCO inside or outside the port; it is in line with expectations.

Demersal fish are more marked than pelagic fish.

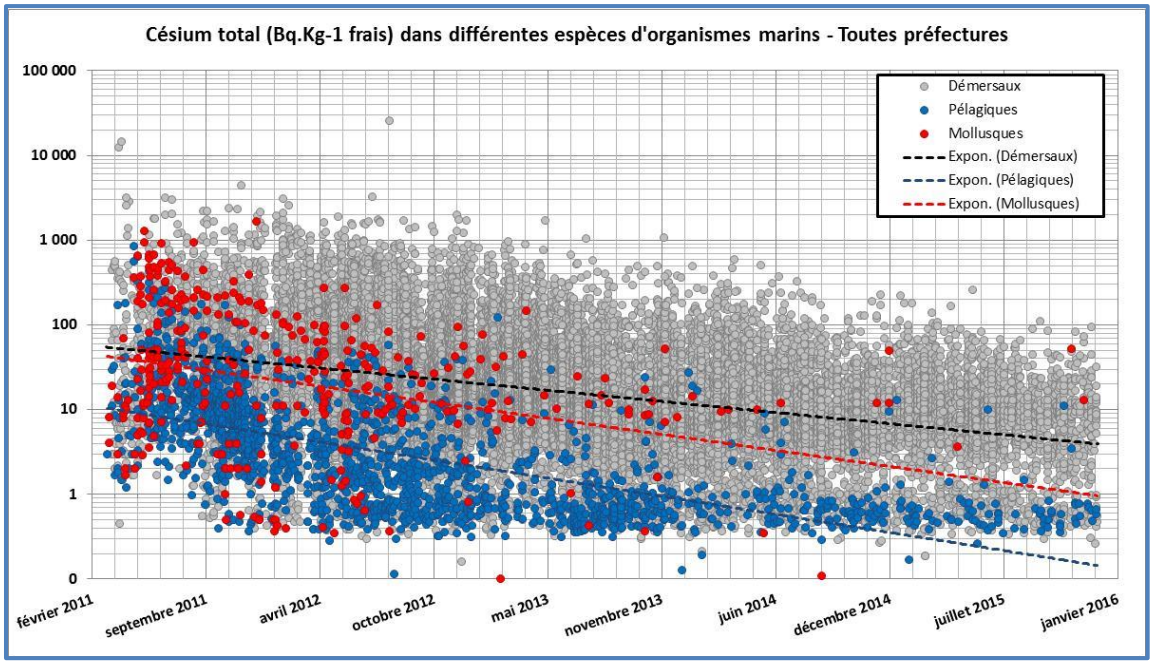

Figure 8: Representation of total caesium concentrations in demersal fish (grey), pelagic fish (blue) and marine invertebrates (red) sampled from the ocean to the north-west of Japan.

The data in figure 8 shows that fish whose lifestyles are linked to sediment (grey) show higher levels than pelagic fish (blue). Since September 2015, radiocaesium concentrations are below the 100 Bq/kg fresh weight marketable limit.

Figure 9 shows that in 2015, off the coast of prefectures other than Fukushima, radiocaesium concentrations in fish arriving to these other prefectures are below the 100 Bq/kg fresh weight marketable limit.

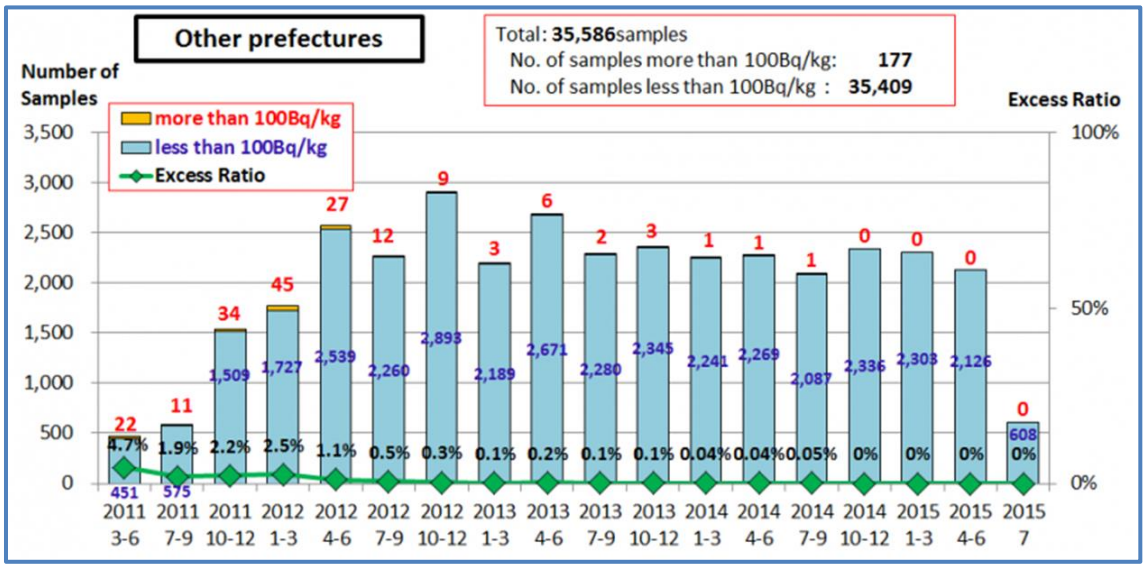

Figure 9: Temporal evolution of the proportion of samples exceeding the radiocaesium marketable limit in fish arriving to prefectures other than Fukushima (http://www.jfa.maff.go.jp/e/inspection/index.html).

Figure 10 shows the evolution of radiocaesium concentrations in demersal fish (those feeding at the sea bed) in six prefectures in East Japan. As expected, the highest levels concern fishing off the coast of Fukushima. More than five years after the accident, demersal species are still close to the permitted limits (100 Bq/kg fresh weight) in the Fukushima prefecture.

In zones far away from the nuclear power plant, the evolution of the activity levels of marine organisms is doubtless linked to the lasting contamination of sediments and inputs via rivers following the leaching of land contaminated by atmospheric fallout.

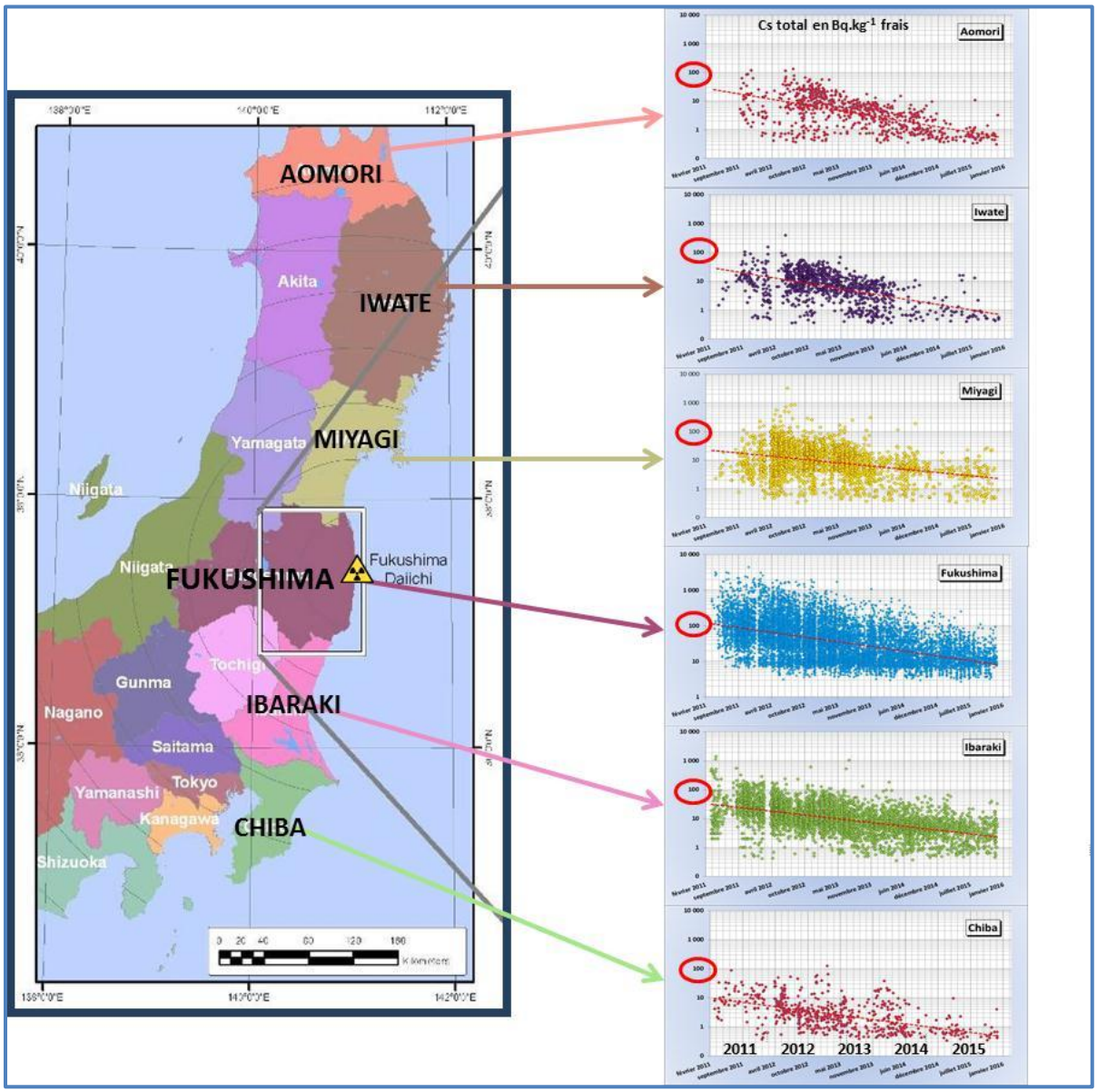

Figure 10: Temporal evolution of radiocaesium concentrations in different species of demersal fish between March 2011 and November 2015, sampled off the coast of the prefectures of Chiba, Ibaraki, Fukushima, Miyagi, Iwate and Aomori.

References

Ambe, D., Kaeriyama, H., Shigenobu, Y., Fujimoto, K., Ono, T., Sawada, H., Saito, H., Miki, S., Setou, T., Morita, T., Watanabe, T. (2014). Five-minute resolved spatial distribution of radiocesium in sea sediment derived from the Fukushima Daiichi Nuclear Power Plant. *Journal of Environmental Radioactivity* 138, 264-275.

Aoyama M., Uematsu M., Tsumune D., Hamajima Y., 2013. Surface pathway of radioactive plume of TEPCO Fukushima NPP1 released 134Cs and 137Cs. Biogeosciences 10, 5 3067-3078. 10.5194/bg-10-3067-2013

Bailly du Bois P., Laguionie P., Boust D., Korsakissok I., Didier D., Fiévet B., 2012. Estimation of marine source-term following Fukushima Daiichi accident. Journal of Environmental Radioactivity 114 2-9. 10.1016/j.jenvrad.2011.11.015

Bailly du Bois P., Garreau P., Laguionie P., Korsakissok I., 2014. Comparison between modelling and measurement of marine dispersion, environmental half-time and 137Cs inventories after the Fukushima Daiichi accident. Ocean Dynamics 64, 3 361-383. 10.1007/s10236-013-0682-5

Black, E.E., Buesseler, K.O. (2014). Spatial variability and the fate of cesium in coastal sediments near Fukushima, Japan. Biogeosciences, 11, 5123-5137.

Bu, W., Zheng, J., Guo, Q., Aono, T., Otosaka, S., Tagami, K., & Uchida, S. (2014). Temporal distribution of plutonium isotopes in marine sediments off Fukushima after the Fukushima Daiichi Nuclear Power Plant accident. J Radioanal Nucl Chem, 1-4.

Charette M.A., Breier C.F., Henderson P.B., Pike S.M., Rypina I.I., Jayne S.R., Buesseler K.O., 2013. Radium-based estimates of cesium isotope transport and total direct ocean discharges from the Fukushima Nuclear Power Plant accident. Biogeosciences 10, 3 2159-2167. 10.5194/bg-10-2159-2013

Estournel C., Bosc E., Bocquet M., Ulses C., Marsaleix P., Winiarek V., Osvath I., Nguyen C., Duhaut T., Lyard F., Michaud H., Auclair F., 2012. Assessment of the amount of cesium-137 released into the Pacific Ocean after the Fukushima accident and analysis of its dispersion in Japanese coastal waters. Journal of Geophysical Research C: Oceans 117, 11. 10.1029/2012JC007933

Kaeriyama, H., Shimizu, Y., Ambe, D., Masujima, M., Shigenobu, Y., Fujimoto, K., Ono, T., Nishiuchi, K., Taneda, T., Kurogi, H., Setou, T., Sugisaki, H., Ichikawa, T., Hidaka, K., Hiroe, Y., Kusaka, A., Kodama, T., Kuriyama, M., Morita, H., Nakata, K., Morinaga, K., Morita, T., Watanabe, T. (2014). Southwest intrusion of 134Cs and 137Cs derived from the Fukushima Daiichi nuclear power plant accident in the western North Pacific. Environmental Science and Technology 48, 3120-3127.

Kumamoto, Y., Aoyama, M., Hamajima, Y., Murata, A., Kawano, T. (2015). Impact of Fukushima-derived radiocesium in the western North Pacific Ocean about ten months after the Fukushima Daiichi nuclear power plant accident. Journal of Environmental Radioactivity 140, 114-122.

Lai Z., Chen C., Beardsley R., Lin H., Ji R., Sasaki J., Lin J., 2013. Initial spread of 137Cs from the Fukushima Daiichi Nuclear Power Plant over the Japan continental shelf: a study using a high-resolution, global-coastal nested ocean model. Biogeosciences 10, 8 5439-5449. 10.5194/bg-10-5439-2013

Povinec P.P., Hirose K., Aoyama M., 2012. Radiostrontium in the western North Pacific: Characteristics, behavior, and the Fukushima impact. Environmental Science and Technology 46, 18 10356-10363. 10.1021/es301997c

Povinec, P.P., Aoyama, M., Biddulph, D., Breier, R., Buesseler, K., Chang, C.C., Golser, R., Hou, X.L., Ješkovský, M., Jull, A.J.T., Kaizer, J., Nakano, M., Nies, H., Palcsu, L., Papp, L., Pham, M.K., Steier, P., Zhang, L.Y. (2013). Cesium, iodine and tritium in NW Pacific waters - a comparison of the Fukushima impact with global fallout. Biogeosciences 10, 5481-5496.

Rypina I.I., Jayne S.R., Yoshida S., Macdonald A.M., Douglass E., Buesseler K., 2013. Short-term dispersal of Fukushima-derived radionuclides off Japan: modeling efforts and model-data intercomparison. Biogeosciences 10, 7 4973-4990. 10.5194/bg-10-4973-2013

Smith, J.N., Brown, R.M., Williams, W.J., Robert, M., Nelson, R., Moran, S.B. (2014). Arrival of the Fukushima radioactivity plume in North American continental waters. Proceedings of the National Academy of Sciences.

Tsumune D., Tsubono T., Aoyama M., Uematsu M., Misumi K., Maeda Y., Yoshida Y., Hayami H., 2013. One-year, regional-scale simulation of 137Cs radioactivity in the ocean following the Fukushima Daiichi Nuclear Power Plant accident. Biogeosciences 10, 8 5601-5617. 10.1007/s10967-012-2033-2

Thornton, B., Ohnishi, S., Ura, T., Odano, N., Sasaki, S., Fujita, T., Watanabe, T., Nakata, K., Ono, T., & Ambe, D. (2013). Distribution of local 137Cs anomalies on the seafloor near the Fukushima Daiichi Nuclear Power Plant. Marine Pollution Bulletin, 74, 344-350.