The accident that occurred at the Fukushima Daiichi plant on 11 March and the days that followed led to the release of radioactive substances into the environment:

- into the atmosphere, in the form of radioactive gases or very fine radioactive particles dispersed into the air (aerosols), of which a portion fell on the surfaces of the soil in Japan, forming residual radioactive deposits;
- into the marine environment, directly in the form of liquid releases into the sea and indirectly due to fallout on the sea’s surface from radioactive aerosols dispersed over the ocean.

1. RADIOACTIVE RELEASES INTO THE AIR

IRSN’s current estimates, which should still be considered provisional, are the following:

- releases of radioactive noble gases: 6,550 PBq (petabecquerels =10^{15} Bq) (the same order of magnitude as the Chernobyl accident), composed mainly of xenon-133 ($^{133}$Xe, half-life of 5.3 days);
- releases of radioactive iodine: 408 PBq (about ten times less than the Chernobyl accident), including 197 PBq of iodine-131 ($^{131}$I, half-life of 8 days) and 168 PBq of iodine-132 ($^{132}$I, half-life of 2.3 hours);
- releases of radioactive tellurium: 145 PBq, including 108 PBq of tellurium-132 ($^{132}$Te, half-life of 3.2 days) with its decay product iodine-132, and 12 PBq of tellurium-129m ($^{129m}$Te, half-life of 33.6 days) with its decay product tellurium-129 ($^{129}$Te, half-life of 1.2 hours; initial release estimated at 8 PBq);
- releases of radioactive caesium: 58 PBq (about three times less than the Chernobyl accident), including 21 PBq of caesium-137 ($^{137}$Cs, half-life of 30 years), 28 PBq of caesium-134 ($^{134}$Cs, half-life of 2.1 years) and 9.8 PBq of caesium-136 ($^{136}$Cs, half-life of 13.2 days).

The other radionuclides released (38) were estimated to represent a total activity of 29 PBq, less than 0.5% of all radioactive substances released. Only some of these radionuclides have actually been detected, in a low quantity, in the Japanese environment. In particular, plutonium released during the accident (attested by its isotopic composition) was measured in the deposits formed in the northwest of the Fukushima Daiichi plant, but at very low levels, difficult to distinguish from the plutonium from fallout in the atmosphere produced by nuclear weapons testing.

The main releases were produced from 12 to 25 March 2011, in about fifteen events, with the most important releases taking place before 17 March. IRSN has reconstructed these release events, attributing them to one or another of the 3 damaged reactors (see figure below). These estimates still contain uncertainty, for the releases that were dispersed over the Pacific in particular, where no measurement was taken. Furthermore, considering the degraded condition of these damaged facilities, and the significant contamination of the nuclear site, atmospheric releases must have continued over the following months, but at a much lower level, which is more difficult to detect in the environment.
2. ATMOSPHERIC DISPERSION OF RELEASES

In 2011, after the first evaluations performed during the emergency, IRSN developed a more precise model of the atmospheric dispersion of the releases caused by the accident, using the updated release estimates and the interpretation of more complete meteorological data and results from ambient dose rate measurements in Japan. This model provides for the identification of different air contamination phases on a regional and local scale:

- the first releases that took place between 12 and 14 March mainly dispersed to the north, along the eastern coast of the island of Honshu, then northeast and east, over the Pacific;

These releases have probably had little impact on Japan, except for to the immediate north of the damaged plant (increase in dose rate detected 12 March in Minamisoma).
Model on a local scale of the ambient dose rate resulting from the atmospheric dispersion of releases from the Fukushima accident during the day of 12 March (excluding the contribution from radioactive deposition - IRSN pX model).

- on 15 and 16 March, releases from reactor 2 dispersed over Japanese land, while meteorological conditions were changing rapidly. The releases occurring in the morning are believed to have moved in a southern direction, along the coast, whereas those occurring during the nights of the 15 and 16 March moved to the northwest, crossing an intense precipitation front moving in the opposite direction;

Model on a local scale of the ambient dose rate resulting from the atmospheric dispersion of releases from the Fukushima accident during the day of 15 March (excluding the contribution from radioactive deposition - IRSN pX model).
- starting 16 March and over the course of the days that followed, the releases were dispersed to the east, over the Pacific, sparing the majority of Japanese land;

- between the afternoon of 20 March and 23 March, the releases were once again dispersed over Japanese land. After 23 March, the masses of contaminated air moved towards the Pacific, and the releases thereafter were too small to cause a significant increase in the radioactivity of the terrestrial environment.

The different air contamination phases described by the model created by IRSN are consistent with the series of dose rate measurements obtained by the sensors installed on Japanese territory, as illustrated in the graph below. This graph also shows that after each episode of radioactive air contamination the dose rate remains higher than before, due to the radiation emitted by the radioactive deposit formed when the air was contaminated.
Time series of the ambient dose rate measurement in Ibaraki, showing the overlap of the Fukushima accident release phase and deposition phase. ●: ambient dose rate before the accident; ○: first atmospheric contamination event; ■: dose rate caused by residual radioactive deposits formed during the first atmospheric contamination event; □: second atmospheric contamination event; ▲: dose rate caused by residual radioactive deposits formed during the successive atmospheric contamination events.

3. CONTAMINATION OF LAND FROM RADIOACTIVE DEPOSITION

Over the course of the air contamination events, a portion of the radionuclides dispersed into the air in the form of very fine particles (aerosols) or soluble gases (a portion of the radioactive iodine) settled onto the ground surfaces, forming radioactive surface deposits. The source of these deposits is two complementary processes, illustrated below.

Dry deposits are formed on all surfaces, regardless of their nature and orientation (horizontal, vertical or inverted), that come into contact with the contaminated air, due to the effect of air turbulence. The size of these dry deposits depends on, in particular, the concentration of radionuclides in the air at the ground level and the length of time of the air contamination. Dry deposits could have formed inside of the buildings when the internal air was contaminated.

Wet deposits formed on land areas where precipitation (rain or snow) was produced during the dispersion of the radioactive plume. They resulted from the transportation of radioactive particles or soluble gases (the case of iodine) by water droplets travelling through the air. They have not affected the interior of the buildings. The distribution of contamination on ground surfaces with wet deposits is inevitably variable over a short distance, due to the flow of water from rain (or melting snow) on the surface or its infiltration into the soil.
These deposits led to a contamination of Japanese land that has remained after the dissipation of the air contamination caused by the accidental releases. These deposits have had two main consequences:

- a permanent increase in the ambient dose rate due to gamma radiation emitted by the radionuclides contained in the deposits, which progressively decreases over time as a function of the radioactive decay of the radionuclides making up the initial deposit;

- contamination of agricultural products, more or less immediate and more or less long-lasting.

The initial composition of the radioactive deposits in Japan reflected that of the air contamination but also depended on the physical-chemical properties of the radionuclides in the air (e.g., noble gases are chemically inert and remain in the gaseous state). Based on the analysis of a soil sample taken from Iitate Maeta, located approximately thirty kilometres northwest of the Fukushima Daiichi plant (samples provided by ACRO), IRSN was able to reconstruct the isotopic composition of the deposition in this area, for its presumed date of formation (15 March 2011). Changes in the contribution of the different radionuclides to the total activity of the deposits, in reference to the initial activity of the deposit, are presented in the figure below. Similar changes were observed for the dose rate measured in the same area.

From 5 April on, the total activity of the residual deposits was less than 20% of the initial activity on 15 March. This rapid change provides a good illustration of the fact that the radiation protection challenges in Japan were concentrated in the first month after the accident, due to the doses potentially received by external exposure to the deposits and the risk of contamination of plant (leafy vegetables) and animal (milk) food products produced in the zones where the radioactive deposition took place.

Two months after the accident, with the disappearance of the radionuclides having short half-lives ($^{132}$Te/$^{132}$I and $^{131}$I), which accounted for the great majority of the initial deposition, the majority of the residual deposits were made up of caesium-134 and caesium-137, whereas these two only accounted for 9% of initial activity. Thus, from 20 May 2011 on, they were contributing more than 80% of the activity of the residual deposits in Japan, which explains why the maps of deposits published in Japan only addressed these two radionuclides.
Map of cumulative deposits of caesium-134 and caesium-137 (a) and the caesium-137 (b) published by MEXT following the various aerial measurements taken since the end of June 2011.

These maps also call for the following comments.

**Inside the 20-km zone (zone of emergency evacuated at the time of the accident),** surface activities in caesium-137 measured in the soil samples vary between less than 30 kBq/m² and 15,000 kBq/m², a difference of a factor of 500 between the extreme values. Out of some one hundred sampling points in this zone, 11 have a measured surface activity exceeding 3,000 kBq/m² in caesium-137. The highest activities are observed directly to the west of the plant, with 14,000 kBq/m² in caesium-134 and 15,000 kBq/m² in caesium-137.

**In the planned evacuation zone implemented starting 22 April, more than 20 km northwest of the plant** (the communities of Iitate, Katsurao and some of Namie, Kawamata and Minamisoma), the surface activities in caesium-134 and caesium-137 vary between less than 60 kBq/m² and 8,000 kBq/m². Out of some one hundred sampling points in this zone, three have a measured surface activity exceeding 3,000 kBq/m² in caesium-137.

**Beyond these two zones,** cumulative surface activities in caesium-134 and caesium-137 do not exceed 600 kBq/m², except for in a limited section between 20 and 30 km to the southwest.

The graph below presents IRSN estimates of the surface area of the contaminated land in Japan, as a function of the level of contamination in caesium-137. **Contamination levels equal, the surface areas involved in Japan are clearly smaller than those of the contaminated land around Chernobyl,** which can be explained by the fact that a large portion of the releases from the Fukushima accident dispersed over the Pacific. It is estimated that there are approximately 600 km² of land with a caesium-137 deposit greater than 600,000 Bq/m² in Japan (including the part within the 20-km zone), compared to 13,000 km² around Chernobyl, representing 20 times less surface area. However, in addition to the 80,000 people emergency evacuated from the 20-km zone, the population living in these areas is estimated at approximately 70,000, which together represents more than half of the population in the most contaminated areas around Chernobyl (270,000 people in the areas with caesium-137 deposits of greater than 555,000 Bq/m²).
Proportion of surface area affected by the caesium-137 deposits as a function of the amount of these deposits (with the assumption that the deposits in the 9-km zone are the highest)

Outside of the main deposition zone (with surface activity in caesium greater than 300,000 Bq/m²), located less than 80 km from the Fukushima Daiichi plant, significant, but lower-intensity, deposits also formed in the extension of the main deposition zone, and beyond that, in the form of isolated spots (distribution in “leopard spots”). There the surface activity in caesium-134 and caesium-137 is generally lower than 100,000 Bq/m², except for in a few areas, where it may be up to 300,000 Bq/m². These levels are comparable to those observed in various European countries, in Scandinavia or Austria for example, after the Chernobyl accident.

Finally, the initial isotopic composition of the radioactive deposits was not the same in all areas. According to measurements taken in Japanese soil, it appears that the area located to the south of the Fukushima Daiichi plant received far higher deposits in iodine-131 than the area to the northwest (report on initial \(^{131}I/^{137}Cs\) activities (in reference to 15 March 2011), near 50 in the south, compared to 8 to 12 in the northwest). A similar trend was observed for \(^{129m}Te\). In this southern sector, where radioactive deposits were lower than those in the northwest, the higher proportion of radioactive iodine could be explained by:

- different deposit formation conditions (predominance of dry deposits);
- differences in physical-chemical forms of iodine (gas or aerosol) in the air at the time of deposition;
- differences in the isotopic composition of the successive releases emitted by the damaged reactors.

This also means that although the deposits remaining to the south of the Fukushima Daiichi plant are relatively lower, it is possible that concentrations in radioactive iodine in the air may have been just as high as in the northwest during the hours that followed the releases.

There may be great disparity in the distribution of the deposits on a local scale:

- where plant cover is dense and very leafy (in the forest for example), a dry deposit has a tendency to be greater and become concentrated in plants;
- a wet deposit (contaminated rain) has a tendency to drain from plant cover and move into the soil to infiltrate the soil or become surface runoff, depending on the imperviousness of the soil and slope of the ground. As a result, wet radioactive deposits are redistributed locally, leading to higher residual deposits (hot spots), for example around trees (contaminated rainwater dripping onto leaves), under roofs, in rainwater drains or in natural accumulation zones.

All of these phenomena may lead to high heterogeneity in residual deposits, which may vary by a factor of 10 or even more, and which are not illustrated in the maps obtained by the aerial
measurement campaigns, which lack sufficient accuracy. Only ground prospecting with portable spectrometers or radiation meters, or soil samples analysed in the laboratory, provide for the detection of “hot spots”. The different measurements published by the Japanese authorities thus revealed these deposition disparities, in the urban environment in particular.

4. CONTAMINATION OF FOOD PRODUCTS IN JAPAN

The deposits of radioactive substances that formed during the dispersion of the radioactive plume led to the contamination of the aboveground parts of the plants, and consequently, plant products intended for consumption by humans and cattle.

- **Plant products**

Leaf vegetables (lettuce, spinach, leeks, etc.) were the most immediately affected by this contamination. The magnitude of this contamination depends on the intensity of the deposits, as well as the dry or wet form of these deposits (with equal deposits, the dry deposit is more effective in contaminating leaves than a wet deposit). The contamination of leaves was maximal immediately after the formation of the deposits; it then decreased rapidly (see figure below), since the new leaves formed by plant growth had not received radioactive fallout (typically, lettuce harvested 50 days after an accident are one hundred times less contaminated than those that are fully grown immediately after the accident).

Of all data transmitted by the MLHW on edible goods, with all categories mixed together, leafy vegetables demonstrated the most earliest and highest-level contamination:

- 54,100 Bq/kg of iodine-131 in a sample of spinach taken on 18 March in Hitashi (Ibaraki district), approximately 120 kilometres south of the plant;
- 41,000 Bq/kg of caesium-134 and 41,000 Bq/kg of caesium-137 in a kukitachina sample (a local leafy vegetable) taken on 21 March in Motomiya (Fukushima district), approximately 70 kilometres west of the plant.

Over the course of the month of March, several vegetable samples from the Fukushima, Ibaraki, Chiba and Tochigi districts showed contamination in caesium and/or iodine-131 exceeding sales or consumption standards. In April and May, isolated above-standard measurements (only for caesium; iodine-131 was no longer detected after mid-May) were still observed. Until the end of June, caesium activities that were detectable but below sales and consumption standard limits were still found.

No findings have been published in Japan about leafy vegetables from the area to the northwest of the Fukushima Daiichi plant, where deposits were the highest, probably due to the lack of
agricultural production in this area or the early growth stages of the products in the weeks following
the accident (end of winter, snow present). In theory, the levels of contamination in leafy
vegetables could have been very high in this area (exceeding one million Bq/kg wet), as supported
by the results obtained on plants (“weeds”) from Iitate.

Later in the year (end of spring, summer and autumn), other categories of plant-origin
foodstuffs exhibited significant levels of contamination with caesium-134 and caesium-137, but
more moderate than for leafy vegetables (see figure below). The most prominent were:

- **tree fruits**, such as *Japanese apricots* (ume; up to hundreds of Bq/kg in the Fukushima
district), the “*yusu*” (up to 2,400 Bq/kg wet on 26 August in Minamisoma or, later, 860
Bq/kg wet on 11 October in Date (Fukushima district), *kiwis* harvested in autumn (with a
specific activity in caesium reaching 1,100 Bq/kg wet on 14 November in Minamisoma),
*figs*, *pomegranates*, *chestnuts*, etc.

- **bamboo shoots**, harvested starting in the beginning of May, with specific activities in
caesium-134 and caesium-137 of up to several thousands of Bq/kg wet;

- **tea leaves**, harvested starting in May, which may have had contamination of several
hundreds of Bq/kg of radioactive caesium, even at a great distance (the Shizuoka district,
more than 300 km from the damaged plant).

![Changes in caesium-134 and -137 contamination in Japanese apricots, tea leaves and bamboo shoots harvested in different Japanese districts.](image)

For these foodstuffs, the mode of contamination was different from those of the leafy
vegetables: these products, which did not exist at the time of the accident, were not directly
contaminated by the radioactive fallout but rather by transfer of sap (translocation phenomenon)
from contamination deposited on the plant surfaces exposed to the fallout (remaining leaves of tea
trees and bamboo plants, tree trunks and flowers). These plants were thus able to store the
radionuclide intercepted in the reservoir organs or tissues (wood, roots, tubercles, etc.); these
radionuclides were then continuously remobilised by the sap, which carries nutrients to growing
plant parts (new leaves, fruits). For this reason, the contamination in these products persisted in
the same range of values for the whole harvest period of 2011, the variations observed in the results
surely due to the different origins of the tested products. It is probable that a similar
contamination, but on an increasingly lower level, will be observed in the next harvests.

Over the long term, it is the transfer of caesium from the soil to the roots (root transfer) that
will constitute the main cause of contamination in agricultural products. However, for a deposit
of the same size, contamination in crops and grass from root transfer is always far lower (100
times less) than the initial contamination caused by deposition on leaves. Furthermore, given the
migration time of radionuclides in soil, root transfer is only significant for radionuclides having a
sufficiently long half-life, such as caesium-134 and -137.
In 2011, this pathway was secondary in relation to the other sources of food product contamination. However, it was probably the source of the low-level caesium contamination observed in certain rice harvests in the Fukushima district (up to several hundreds of Bq/kg). Of the nearly 3,900 analyses on raw or refined rice carried out between August 2011 and January 2012, less than 10% revealed detectable caesium contamination (samples mainly from the Fukushima district); 3 analyses revealed concentrations higher than those authorised by standards (500 Bq/kg). For these cases, the source of the contamination is likely the combination of root transfer and direct transfer into the plant of caesium dissolved in the water in which the crops were growing.

In 2011, in order to limit the impact of root transfer on agricultural products, Japanese authorities prohibited the cultivation of plots where soil contamination exceeds 5,000 Bq/kg of radioactive caesium per kilogramme of soil. For a root transfer factor of 0.1, a conservative value, the caesium contamination in crops growing in soil with 5,000 Bq/kg could reach 500 Bq per kilogramme of wet product, corresponding to the maximum permitted level in Japan for food products.

Certain mushrooms harvested in the Fukushima district also rapidly exhibited relatively high levels of contamination. This concerns species of which mycelium (an underground, perennial fungal colony) was the closest to the surface (at a depth of a few centimetres) and which, for this reason is particularly sensitive to contamination from rainwater infiltrating into the soil. It is for this reason that activities in iodine-131 and caesium-134 and -137 ranging from a few dozen to thousands of Bq/kg were measured starting in the month of April in mushrooms from the Fukushima district, demonstrating this rapid transfer. The majority of mushrooms with high contamination were dried shiitake mushrooms (it should be noted that drying can concentrate the caesium up to tenfold), but high-level activities were also measured in fresh mushrooms. The highest caesium ($^{134}\text{Cs}+^{137}\text{Cs}$) activity identified among the published results was 28,000 Bq/kg; it was measured in a “weeping milk cap” ($\text{Lactarius volemus}$) mushroom, picked on 1 September 2011 in Tanagura-machi (Fukushima district). Such mushroom contamination should recur over the years to come, justifying routine monitoring and precaution with regard to consumption.

- **Animal products**

Contamination of animals and livestock products (meats, milk, eggs, etc.) is mainly a result of the ingestion of contaminated food products: fodder and, to a lesser extent, water.

In general, based on the results published in Japan, the activities measured in milk seem to have been relatively moderate in comparison to the very high levels of deposits and the contamination of grass to the northwest of the damaged power plant. Thus, the higher activities measured in milk were 10 to 100 Bq/L, with a maximum measurement of 210 Bq/L for each of the caesiums (in Iitate, 19 March 2011) and 100 to 1,000 Bq/L, with a maximum measured value of 5,300 Bq/L for iodine-131 (in Kawamata, 20 March). Between March 2011 and the beginning of February 2012, out of approximately 2,000 analyses of milk and milk product samples, only 23 exhibited iodine or caesium activities that exceeded permitted sales standards.

It is likely that the date of the accident, occurring at the end of winter, limited animal contamination (and thus milk contamination), as animals must be fed with either local fodder harvested during the previous season or imported fodder (the importation of animal feed seems common in Japan). For reference, the graph below shows the contamination levels that could have been reached in cow’s milk in the community of Iitate, approximately 40 km northwest of the Fukushima Daiichi plant, if the cattle had consumed locally grown grass, based on the results published in Japan on samples of “weeds” from this area. The concentrations measured in milk from this area were 100 to 1,000 times lower than these theoretical predictions.

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1 For reference, if it is assumed that the caesium is distributed homogeneously in the first 10 centimetres of soil having a density of approximately 1500 kg/m$^2$, a contamination of 5,000 Bq/kg in the soil corresponds approximately to a caesium surface deposit of 750,000 Bq/m$^2$, a level which, in Japan, is practically only observed in the main contamination zone located to the northwest of the Fukushima Daiichi power plant as well as less than some thirty kilometres southwest of the site.
Activity concentrations of caesium-137 in milk samples having the highest levels of contamination and from the Iitate-Kawamata-Date region, as well as Motomiya (red symbols). Specific activities of caesium-137 measured in samples of “weeds” from Iitate (blue diamonds) and model using the ASTRAL software for caesium-137 activities in these plants (blue line) and in milk (red line) produced by cows that may have consumed them. The model was created assuming an initial caesium-137 deposit of 2 MBq/m² at 80% for a wet deposit.

Like milk, and probably for the same reasons, the meat contamination seems to have remained very moderate in relation to the radioactive deposits in the most affected areas. Iodine-131 was almost never found in the meat: the activities remained below detection limits, even if these are relatively high (usually 20 to 50 Bq/kg). Of the some 65,000 meat sample analysis results (between March 2011 and February 2012), about 200 had caesium activities exceeding sales limits (500 Bq/kg), including 67 game samples (mainly wild boar meat) and 141 samples of beef. For the sake of comparison, if the animals had consumed grass having the same activity level of the weeds measured in Iitate, the caesium-137 contamination in the meat could have reached 200,000 Bq/kg wet in mid-April 2011.

More than the location and size of these deposits, it is food production practices that are the determining factors. Thus, two batches of meat samples were analysed on 8 and 9 July in Minamisoma-shi, north of the Fukushima Daiichi plant, on a farm located just outside the 20-km exclusion zone. The results of the two batches are drastically different: 1,500 to 4,200 Bq/kg for the first batch and 4 to 10 Bq/kg for the second. As pertains to the 11 bovines involved in the first batch with the highest activities, and based on information published on 12 July in the Mainichi Daily News, since the beginning of April, the livestock farmer had been feeding his livestock rice straw harvested the previous autumn that had been stored outside and was consequently contaminated by atmospheric radioactive fallout in March.

In the coming years, activities in caesium-134 and caesium-137 in animal products, milk and beef in particular, will continue to depend mainly on the practices used to feed animals. Therefore, a livestock practice that strictly respects the fodder contamination limit of 300 Bq/kg for the production of beef and milk (MAFF decisions, 14 April 2011), could lead to activities in milk that would be near the sales standard for milk (200 Bq/L) and above-limit activities for meat.

Nearly 350 analyses of game meat were performed in Japan between April 2011 and January 2012, mainly since autumn 2011. They cover wild boar meat (155 analyses), the Asian black bear (85 analyses), pheasant (42 analyses) and cervids (65 analyses). Unlike in livestock products (beef), the caesium activities (\(_{134}^{134}\text{Cs} + _{137}^{137}\text{Cs}\)) were most often above detection limits. The highest levels were observed in wild boar and bear meat. The figure below presents the specific activities measured in wild boar meat. They are extremely variable: from a few Bq/kg wet to a few thousand Bq/kg wet (maximum value recorded of 14,600 Bq/kg wet), the highest being those from the Fukushima district.
5. CONTAMINATION OF THE MARINE ENVIRONMENT

High-level radioactive pollution was observed in the marine environment close to the Fukushima Daiichi nuclear power plant starting on 21 March and in the days that followed, as illustrated in the figure below. This pollution resulted from releases of water used to cool the damaged reactors, which had been in contact with materials heavily contaminated by the atmospheric releases, and some of which flowed to the sea, as well as water that escaped the containments of reactors 2 and 3. In particular, a crack in the reactor pit adjacent to the turbine hall of reactor 2 caused highly contaminated water to be released directly into the sea. On April 6, at approximately 6 AM local time, TEPCO successfully stopped the leak by injecting sodium silicate.

Iodine-131 (\(^{131}\text{I}\)) and caesium-137 (\(^{137}\text{Cs}\)) are the main radionuclides that were monitored in the marine environment in 2011. Although it was predominant at the time of the accident, the amount of iodine-131 fell sharply in the weeks following the accident, due to its rapid radioactive decay. After the end of May, it was no longer detectable. Other artificial radionuclides, most with a short half-life, have also been detected occasionally, at lower concentrations.

The maps below represent the spatial distribution of average caesium-137 concentrations for successive periods between 11 April and 11 July.
Changes in the spatial distribution of $^{137}$Cs concentrations in seawater between 11 April and 11 July 2011.

These maps show a similar distribution of the pollution from the vicinity of the nuclear power plant towards the open sea. The concentrations fell sharply over time, and the area of the coloured zones, corresponding to measurements above the detection limit (around 5 Bq/L), decreased accordingly. After mid-July, the concentrations measured at sea were mostly below the detection limits of the measurement methods used for monitoring. From that time onwards, it was no longer possible to perform a representative inventory of the residual pollution at sea.

Based on this data, IRSN estimates that after 11 April 2011, the quantities of caesium-137 present in seawater inside the calculation zone halved every 6.9 days. This decrease was due to the dilution of the polluted seawater by sea currents that brought a regular supply of uncontaminated water into the zone concerned. This is an especially fast renewal rate. It is due to the strength of the Kuroshio and Oyashio currents, which meet in this area, and their general orientation towards the open sea.

This active dilution effect tended to reduce the impact of the accident on coastal waters. Contaminated waters were carried swiftly eastwards, towards the centre of the Pacific, where they continued to be diluted due to the dispersion of ocean waters.

The maps below illustrate this dispersion of caesium-137 and were created by IFREMER upon the request of IRSN, using a marine dispersion model (MARS 3D).
By interpreting the results of measurements published in Japan, IRSN was able to determine the overall quantity of caesium-137 released into seawater until mid-July. The value thus obtained is estimated at $27 \times 10^{15}$ Bq. Most of the release took place before 8 April, and the estimated releases after that date accounted for only 18% of the total release. This is the largest one-off injection of artificial radionuclides into the marine environment ever observed. However, the estimation of these releases still lacks precision, the different sources published in 2011 offering results that vary by a factor of 1 to 27.

For reference, this total contribution of $27 \times 10^{15}$ Bq of caesium-137, diluted at a depth of 0 to 100 metres throughout Pacific Ocean, would lead to an added concentration of 0.002 Bq/L, which would be double the residual background in the seawater due to the fallout from atmospheric nuclear tests (0.002 Bq/L). Although measurable with current techniques, these concentrations would represent only one $3000^{10}$ of the natural potassium-40 concentrations in seawater (12 Bq/L).

Furthermore, atmospheric caesium-137 fallout on marine surfaces was evaluated by IRSN to be $76 \times 10^{12}$ Bq (76,000 billion Becquerels) in an 80-km radius. This supply of contamination into the sea represents only 0.3% of the overall activity from caesium-137 released directly into the sea by the Fukushima Daiichi power plant, as estimated by IRSN.

The Fukushima Daiichi accident also caused pollution in marine sediment along coastal areas. The results of measurements taken in Japan, obtained for sediment samples taken from the Japanese coasts (up to 186 km) and offshore (up to 70 km) have shown the presence of radioactive caesiums, iodine-131 (until 9 July 2011) and other radionuclides in lower quantities. These results also show that except for in the immediate proximity of the Fukushima Daiichi plant, where concentrations in caesium-137 were higher (100,000 and 150,000 Bq/kg measured near the marine outfall), concentrations in the sediment tended to vary from 1 to 10,000 Bq/kg, with an average tendency toward increase over time. Based on these results (excluding the high values obtained near the outfall), IRSN created a map of the average caesium-137 concentrations in the sediment, presented below.
Map of $^{137}\text{Cs}$ concentrations in sediments (mean of values measured until 14 October).

The concentrations reached were lower than expected, given the high concentrations measured in the seawater and the equilibrium distribution coefficient of caesium between the seawater and the sediments, which is usually greater than 1000. Thus, with concentrations greater than 100 Bq/L measured in coastal seawater, one could have expected to find concentrations of 100,000 Bq/kg in the sediments. The transient pollution of the seawater by caesium-137 probably did not allow equilibrium to be reached with the sedimentary stock sampled.

In the years to come, excluding any possible new releases into the sea from the Fukushima Daiichi power plant, the main source of radioactive pollution in the marine environment will be releases resulting from rain leaching of caesium in deposits on the ground and its transport via waterways. For the prediction of these transfers to the sea and changes in them, there is currently no model that is sufficiently robust and operational to provide reliable forecasts of these future releases and their impact in the marine environment.

Radioactive pollution in the sea caused by the Fukushima Daiichi has also had an impact on marine species, as illustrated in the figure below (concentrations in caesium-134 and -137).
Change over time in $^{137}$Cs + $^{134}$Cs (Bq/kg) concentrations in some sea products. The pink marks indicate diadromous species, mainly fished in rivers or lakes.

Of the marine products sampled, those with the highest contamination levels, detected at the beginning of fishing product monitoring, were the Japanese sand lances (sand eels), fished at the juvenile stage until the end of April. Caesium-137 and -134 have been detected in all samples of this species taken from the Fukushima and Ibaraki districts; the maximum concentrations reached 12,500 Bq/kg wet ($^{134}$Cs+ $^{137}$Cs) off the shore of Iwaki on 13 April.

In addition to exclusively marine animals, three diadromous species (species that migrate between freshwater and saltwater) that were sampled starting in the month of May in lakes or rivers (the ayu, masu salmon and Japanese smelt) also exhibited relatively high contamination levels.

As pertains to the filter-feeding species, such as mussels, oysters and clams, caesium contamination tends to decrease where there are no significant new releases. To the contrary, changes in the contamination levels of certain types of fish do not follow the same decay pattern observed in seawater. In fact, for fish, variations in concentration could very well be due to their movement, for species that are not strictly confined to the area where they were fished, but they also reflect the complex trophic regimes of these organisms, which differ from one species to the next.

Generally speaking, fish will be the best medium- and long-term indicators of caesium contamination in the marine domain. Caesium presents higher concentration factors in fish, and tends to increase in species that are higher up in the trophic chain. Consequently, although in the short term the highest concentrations tend to be found in species positioned at the beginning of the food chain, in the longer term, once the transfer to the different links of the trophic networks has taken effect, the predators at the top of the food chain should present higher levels. These levels should be even higher for species whose way of life involves close contact with sediments and whose habitat is close to the contaminated zone.

Thus, even if the caesium contamination in the sea water has fallen sharply in the vicinity of the Fukushima Daiichi power plant, there is justification for continuing to monitor the marine species fished from the coastal waters of northeast Japan.