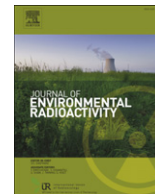


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Estimation of marine source-term following Fukushima Dai-ichi accident

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ABSTRACT

Contamination of the marine environment following the accident in the Fukushima Dai-ichi nuclear power plant represented the most important artificial radioactive release flux into the sea ever known. The radioactive marine pollution came from atmospheric fallout onto the ocean, direct release of contaminated water from the plant and transport of radioactive pollution from leaching through contaminated soil. In the immediate vicinity of the plant (less than 500 m), the seawater concentrations reached 68 000 Bq.L⁻¹ for ¹³⁴Cs and ¹³⁷Cs, and exceeded 100 000 Bq.L⁻¹ for ¹³¹I in early April. Due to the accidental context of the releases, it is difficult to estimate the total amount of radionuclides introduced into seawater from data obtained in the plant. An evaluation is proposed here, based on measurements performed in seawater for monitoring purposes. Quantities of ¹³⁷Cs in seawater in a 50-km area around the plant were calculated from interpolation of seawater measurements. The environmental halftime of seawater in this area is deduced from the time-evolution of these quantities. This halftime appeared constant at about 7 days for ¹³⁷Cs. These data allowed estimation of the amount of principal marine inputs and their evolution in time: a total of 27 PBq (12 PBq–41 PBq) of ¹³⁷Cs was estimated up to July 18. Even though this main release may be followed by residual inputs from the plant, river runoff and leakage from deposited sediments, it represents the principal source-term that must be accounted for future studies of the consequences of the accident on marine systems. The ¹³⁷Cs from Fukushima will remain detectable for several years throughout the North Pacific, and ¹³⁷Cs/¹³⁴Cs ratio will be a tracer for future studies.

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1. Introduction

Contamination of the marine environment following the Fukushima Dai-ichi accident occurred firstly from dry and wet deposition processes from an atmospheric contaminated plume directed mainly towards the sea between March 12 and March 23; secondly from direct releases of highly contaminated waters into the sea; and thirdly from the transport of radioactive pollution by leaching through contaminated soil. This event represents the most important artificial radioactive liquid release flux into the sea ever known on a short time and space scale basis (Aarkrog, 2003; Linsley et al., 2005; Livingston and Povinec, 2000; Table 2; Buesseler et al., 2011). Liquid controlled releases from the Sellafield reprocessing plant could be compared in term of total quantities, but they occur

during several years (1970–1980) instead of weeks and months for Fukushima accident.

Estimation of the quantities introduced in the marine environment is a key issue for assessing their consequences and governs the future studies on the behaviour of contaminated waters and radioactivity transfers to living species and sediments.

This work attempts to estimate the marine source-term, based on measurements performed at sea by the plant operator TEPCO and the Japanese regulatory authority MEXT. This estimation was mainly based on ¹³⁷Cs and ¹³¹I measurements, which are representative of dissolved substances.

Caesium-137 is essentially soluble in seawater; it will be carried over very long distances by marine currents and dissipated throughout the ocean water masses (Aoyama and Hirose, 2003; Livingston and Povinec, 2000; Povinec et al., 2004; Sanchez-Cabeza et al., 2011). A significant part of caesium and other radionuclides will bind to suspended particles and cause sedimentary contamination by deposition on the seafloor, as observed previously for atmospheric fallout from nuclear tests (Lee et al., 2005;

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Moon et al., 2003). Consequences will be significant for months and years in the vicinity of the plant. This will require long-term monitoring in Japanese coastal areas.

Although different origins of the pollution could result in variable ratios to ^{137}Cs for other radionuclides in the source terms, ^{137}Cs fluxes give a good estimate of other radionuclides released at the same time. Activity ratios of other radionuclides versus ^{137}Cs were derived when data were available. To facilitate comparison between different labelling, all radionuclide/ ^{137}Cs ratios were decay corrected to the initial ratio, at the date of the tsunami 2011 March 11, when the power plant was stopped. It is called IR(radionuclide/ ^{137}Cs).

The paper describes briefly the hydrodynamic of the zone. The different radionuclide sources are presented and estimation of ^{137}Cs released quantities is reported. Direct release fluxes are associated with the time series concentrations close to the plant. The discussion compares the results with other main releases of ^{137}Cs in oceans.

2. Hydrodynamics

The Fukushima Dai-ichi power plant is located on the East coast of the island of Honshu, 200 km north-east of Tokyo. The coast runs north-south, facing the Pacific Ocean. The depth increases steadily offshore, reaching some 200 m at 50 km from the coast; it then increases suddenly to 5000 m beyond about 100 km offshore.

In the zone mainly affected by radioactive pollution, the currents are generated by the tide, the wind and the general circulation of the Pacific water masses. On a short term basis, the effect of the tide is predominant; the tide moves the water masses in a back and forth motion, north and south along the coast, at speeds of the order of one meter per second and a periodicity of 12 h. The wind influences the circulation of surface water.

The general large-scale circulation is the result of the interaction between the strong Kuroshio oceanic current which comes from the south and runs along the coasts of Japan, and the Oyashio current which comes from the north. The coastal waters in the vicinity of the Fukushima Dai-ichi plant are located in the zone where these two currents interact, creating variable gyrotory currents. These currents will determine the medium-term dispersion of the radioactive pollution. The long-term migration of the surface waters could be southwards but will not extend beyond the latitude of Tokyo. The Kuroshio current will act as a frontier and carry the plume to the east towards the centre of the Pacific (Jayne et al., 2009).

The radioactive substances mix in a 20–50 m thick surface layer, near the coast. This layer can reach 100 m in thickness further offshore. Dispersion of the soluble fraction of the radionuclides will mainly take place in the surface mixing layer. The fraction of radionuclides associated with solid particles nevertheless could be transported to the bottom of the sea by sedimentation (Lee et al., 2005; McCartney et al., 1994; Moon et al., 2003).

Prior to the accident, concentrations of radionuclides in Pacific surface waters resulted mainly from global fallout following atmospheric nuclear weapon tests. For ^{137}Cs , concentrations were in the range 1–4 Bq.m⁻³ (Nakanishi et al., 2010; Povinec et al., 2004).

3. Source-terms

3.1. Nuclides of concern

Caesium-137 concentration levels in seawater off the eastern Japan coast prior to the accident were in the same order of magnitude as other surface oceanic waters, between 1 and 3 Bq.m⁻³ for ^{137}Cs (Nakanishi et al., 2011). After the accident,

measured concentrations in a 30 km perimeter around the plant exceeded 10 Bq.L⁻¹ (10 000 Bq.m⁻³) and reached 68 000 Bq.L⁻¹ close to the plant (68 E⁶ Bq.m⁻³).

The radionuclides with short radioactive half-life (less than a few tens of days, like ^{131}I) ceased to be detectable after a few months and should not have any large-scale and long-term impact. Others, such as ^{134}Cs and ^{106}Ru will persist in the marine environment for several years. Their persistence in the water column is dependent upon the respective affinity of the radionuclides for the particles in suspension in surface waters which are likely to settle and to carry the radionuclides to the sea bed. The concentrations in ^{131}I and ^{137}Cs are representative of other soluble radionuclides measured in the sea.

All results presented here come from compilation of monitoring data provided on the Internet by TEPCO and MEXT. It includes 948 significant seawater measurements (above the detection limits) for ^{137}Cs alone at the date July 20 2011.

3.2. Atmospheric fallout onto the surface of the sea

Following explosions and pressure venting of the reactor containments in the Fukushima Dai-ichi nuclear power plant, an atmospheric plume of contaminated aerosols was transported mainly to the sea between March 12 and March 23. Atmospheric releases were dispersed over long distances within only a short delay: an homogeneous labelling of North Hemisphere was simulated and measured after about 15 days (Masson et al., 2011). Part of the radionuclides contained in the atmospheric plume fell back to the surface of the sea by dry deposition or wet scavenging of the plume, yielding in diffuse pollution of the surface water up to tens of kilometres from the nuclear power plant. Higher surface concentrations resulted from wet deposition which represented more than 99% of the labelling at a distance of 50 km from the plant, whilst dry deposition on water was quite small.

On the sea, this contamination was difficult to appraise because deposition involved large surfaces and was quickly advected and dispersed. Only seawater measurements performed a few days after the release could give an estimation of the quantities. Moreover, atmospheric models have large uncertainties regarding the deposition parameters onto the sea.

Before March 24 when direct liquid release was still relatively low, the concentrations measured in seawater at more than 10 km from the coast could be attributed to atmospheric deposits. They ranged from 9 to 13 Bq.L⁻¹ for ^{137}Cs with IR($^{131}\text{I}/^{137}\text{Cs}$) of 5–12. At the same time another labelling was observed along the coast at more than 10 km south of the plant in the range 20–100 Bq.L⁻¹ of ^{137}Cs with an activity ratio $^{131}\text{I}/^{137}\text{Cs}$ (IR($^{131}\text{I}/^{137}\text{Cs}$)) of 35–110. This could be attributed as well to atmospheric deposit or to previous direct release from the plant or influence of river after washout of contaminated soil by rain. These labelling results revealed at least two different kinds of radionuclide origins, possibly from different plants (Fukushima Dai-ichi unit n° 1, 2 or 3).

The values measured on March 25 tended to indicate a reduction in surface concentrations at more than 10 km from the coast. This may be the result of either mixing with deeper water (dilution effect) or renewal of surface water by sea currents.

The total amount of ^{137}Cs deposited on the sea surface was derived from atmospheric dispersion calculations using the Gaussian puff model of the IRSN technical crisis centre, named pX (Soulhac and Didier, 2008). The model was forced with 3D meteorological forecasts at a resolution of 0.125° from the ECMWF (European Centre for Medium-range Weather Forecast), provided by Météo France (the French meteorological office). The atmospheric source-term was first estimated by IRSN technical

crisis centre during the crisis (IRSN, 2011) based on reactor parameters and other available information (dates of venting and explosions). It was later refined using the chronology of events (TEPCO, 2011), but also based on atmospheric gamma dose rate measurements both on-site and throughout Japan. This estimation remains highly uncertain, both in terms of released activity and timing. This is especially true for releases that were dispersed towards the sea, since there were no atmospheric measurements available that could be used to adjust the atmospheric source-term. A more detailed description of the simulations parameters and results, and comparisons to measurements in the atmosphere are given in Quélo et al. (2011) and Mathieu et al. (2011). The deposition velocities used over seawater are lower than over lands: a value of $5.10^{-5} \text{ m}^2 \text{ s}^{-1}$ was taken for all particulate radionuclides (Pryor et al., 1999; Maro, D., personal communication). Wet scavenging was modelled similarly over the sea and the land, with a scavenging coefficient of the form $A p_0^B$, with $A = 5.10^{-5} \text{ h mm}^{-1} \text{ s}^{-1}$, $B = 1$, and p_0 the rain intensity in mm h^{-1} (Sportisse, 2007).

According to these simulations, the estimate of atmospheric deposits on the surface of the sea calculated at 80 km from Fukushima Dai-ichi plant is 76 TBq with $\text{IR}^{(131\text{I}/137\text{Cs})} = 14$. Pittauerová et al. (2011) found a $^{131\text{I}}/^{137\text{Cs}}$ ratio of 10 at the end of March in Germany, which corresponds to a ratio higher than 20 on March, 11. It is difficult to conclude whether this ratio could be influenced by different deposition processes for $^{137\text{Cs}}$ and $^{131\text{I}}$ during transport in the northern hemisphere. Our estimation is based on a total atmospheric release of 11.5 PBq of $^{137\text{Cs}}$. The cumulated $^{137\text{Cs}}$ deposit calculated by pX onto the sea is presented in Fig. 1.

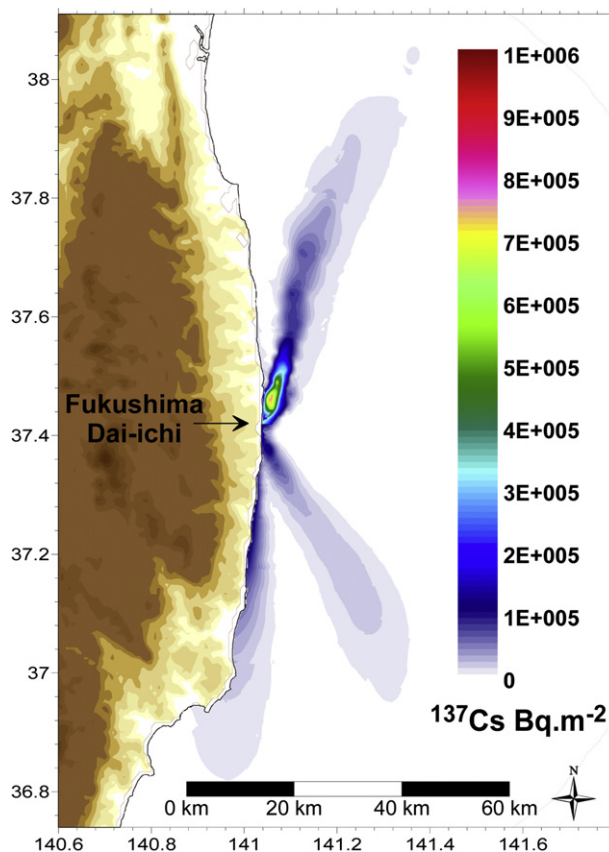


Fig. 1. Calculated cumulate deposit of $^{137\text{Cs}}$, 2011 April 23.

3.3. Rain water washout

The radioactive terrestrial deposits that occurred during the dispersion of the atmospheric releases from the Fukushima Dai-ichi plant could be partially leached by rainwater and then transported to the sea by runoff. The contaminated land areas thus drained can represent several thousands of square kilometres. The measurements currently available did not enable to differentiate these diffuse inputs from the other sources of radioactive pollution. This input will probably be detectable several months after the accident. When seawater concentrations along the coasts have decreased, this contribution will become detectable.

3.4. Direct liquid release from Fukushima Dai-ichi plant

The high concentrations recorded in the seawater in the immediate vicinity of the Fukushima Dai-ichi power plant indicated that there were one or several sources of radioactive liquid effluents escaping directly from the nuclear power station. They consisted of the water used to cool the damaged reactors, part of which has washed over surfaces contaminated by radioactive deposits formed during the atmospheric release. It is equally possible that part of the water present in the damaged reactors (in particular reactor No. 2 of which the bottom part was damaged) has leaked out of the containment building, and subsequently run into the sea.

The influence of these liquid releases was particularly significant from March 26 to April 8 in the vicinity of the nuclear facilities (mean concentration of $15\,700 \text{ Bq.L}^{-1}$ of $^{137\text{Cs}}$, maximum of $68\,000 \text{ Bq.L}^{-1}$, Fig. 2). The $\text{IR}^{(131\text{I}/137\text{Cs})}$ was roughly homogeneous during this period, with a slight trend to decrease from 24 to 18. It revealed a regular process of $^{131\text{I}}$ elimination (in the plant or in seawater) with a half-time of 35 days. The same evolution was observed between 10 and 20 km southward of the plant between March 27 and April 16. This evolution was not perceptible far from the coast. At present, we have no explanation of this behaviour.

The drop in the concentrations measured after April 10 in the vicinity of the facilities showed that they were far smaller direct releases after this date. At this moment large changes in $\text{IR}^{(131\text{I}/137\text{Cs})}$ ratio were measured, probably resulting from processes applied in the plant to reduce the amounts of radioactivity released, with probably different and variable efficiency for caesium and iodine.

At more than 5 km from the coast (Fig. 3), atmospheric radioactive fallout influence was detectable before April 2 with $^{137\text{Cs}}$ concentrations of $10\text{--}30 \text{ Bq.L}^{-1}$ and $\text{IR}^{(131\text{I}/137\text{Cs})}$ lower than close to the plant, as expected for this contribution. After this date the direct release dominated all seawater measurements available at a distance below 50 km.

Regarding other radionuclides detected in seawater with a radioactive decay half-time longer than 100 days, $^{134\text{Cs}}$ shows the same behaviour as $^{137\text{Cs}}$ with a $\text{IR}^{(134\text{Cs}/137\text{Cs})}$ close to 1 ($n = 773$, 95% interval: 0.73–1.46). $\text{IR}^{(90\text{Sr}/137\text{Cs})}$ and $\text{IR}^{(99\text{Tc}/137\text{Cs})}$ were around 0.02 and 0.01 respectively ($n = 8$). These values are only rough estimations resulting of monitoring measurements with a detection limit higher than 1 Bq.L^{-1} .

4. Estimation of $^{137\text{Cs}}$ released quantities

When, for a given period, individual measurements in seawater gave a general coverage in space with a good representation of concentrations variations, it was possible to draw maps of concentrations in seawater by spatial interpolation. This method has been applied before in the English Channel and North Sea to quantify and evaluate the behaviour of industrial releases (Bailly du

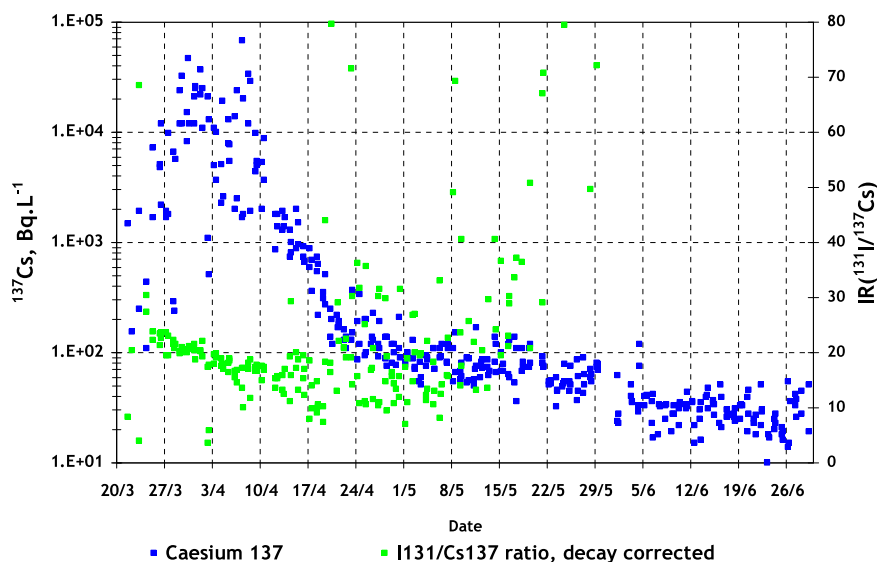


Fig. 2. Evolution of ^{137}Cs concentrations and $^{131}\text{I}/^{137}\text{Cs}$ ratios in seawater at less than 2 km from the Fukushima Dai-ichi power plant.

Bois et al., 1995; 1999). The interpolation method used is the default “Point Kriging” method from “SURFER”[®] software. This method produces nearly the same average values as linear interpolation with triangulation, with more realistic contours. Such maps were drawn with an averaging period of 7 and 14 days between April 11 and June 25 in the area where measurements were the more homogeneous in space and time, i.e.: latitude 36.93° – 37.82° , longitude: 140.9° – 141.45° ($100\text{ km} \times 50\text{ km}$ latitude/longitude width, blue frames in Fig. 4). Measurements before March 21 and April 11 were not accounted for interpolation because the concentration gradients were too steep ($63\,000$ – 5 Bq.L^{-1}) and sampling distributions were not liable to reproduce the plume extension close to Fukushima Dai-ichi.

There were fewer measurements at depth than at the surface (28% of measurements). When they were available, they represented one or two samples below the mixing layer and they had lower concentrations. Such sparse data did not make it possible to interpolate concentrations with depth reliably. To estimate the

volume and quantities of labelled waters, we assumed that surface measurements were representative of concentrations in the surface mixed layer. The thickness of this layer was deduced from bathymetry and hydrographic measurements of temperature and salinity performed by JAMSTEC – MERI (Marine Ecology Research Institute) around the site between March 28 and May 3. A map representing the spatial variation of this thickness was used for calculation of ^{137}Cs inventories; the average depth is 32 meters. This map uses bathymetric data when the mixing layer reach the sea bed (JODC, 2011). The error associated with this quantification is estimated to be about 50%.

Fig. 4 shows the evolution of ^{137}Cs concentrations over time. Estimated total quantities present in the area at different periods are reported in Table 1. It appears clearly that this evolution followed a power law with a constant environmental halftime of 6.9 days (95% interval: 5.7–8.6 days; Fig. 5). The environmental halftime represents the time in which one-half of the radioactivity is removed from the calculation area. This decrease

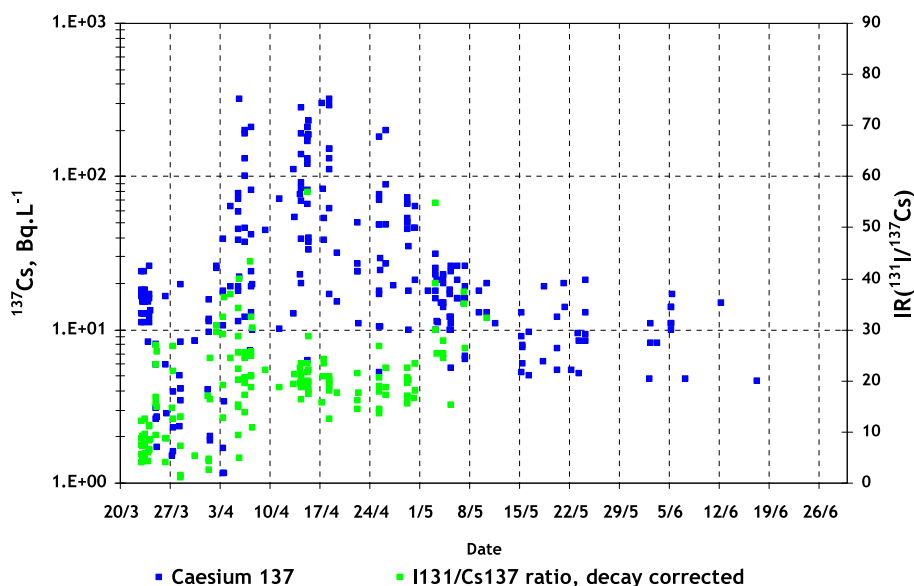


Fig. 3. Evolution of ^{137}Cs concentrations and $^{131}\text{I}/^{137}\text{Cs}$ ratios in seawater at more than 5 km from the coast.

Table 1
¹³⁷Cs quantities from interpolation of individual measurements in seawater in a 100 × 50 km box off Fukushima Dai-ichi plant (see blue frames in Fig. 4).

Measurement period			Number of measurement	¹³⁷ Cs quantity in PBq (±50%)
Begin	End	Middle		
11/04/11	18/04/11	14/04/11	92	11.6
18/04/11	25/04/11	21/04/11	77	4.75
25/04/11	02/05/11	28/04/11	118	3.38
02/05/11	16/05/11	09/05/11	293	0.667
16/05/11	30/05/11	23/05/11	233	0.261
30/05/11	13/06/11	06/06/11	227	0.163
13/06/11	27/06/11	20/06/11	250	0.042
27/06/11	12/07/11	04/07/11	202	0.0024

resulted from dilution by currents and inputs of non-contaminated water into the area considered for calculation. The regularity of this dilution is remarkable, considering the variation of the circulation pattern observed in this area (JHOD observations). It means that, whatever the current direction, water mass fluxes were governed by the general strong currents Kuroshio and Oyashio that were stable at this scale. This efficient dilution diminished the local impact of the accident in coastal waters. Contaminated waters will be transported quickly to the east, into the central Pacific.

Extrapolation of the regression curve at the date of the April 8 allows estimating the total amount of ¹³⁷Cs discharged at the end of

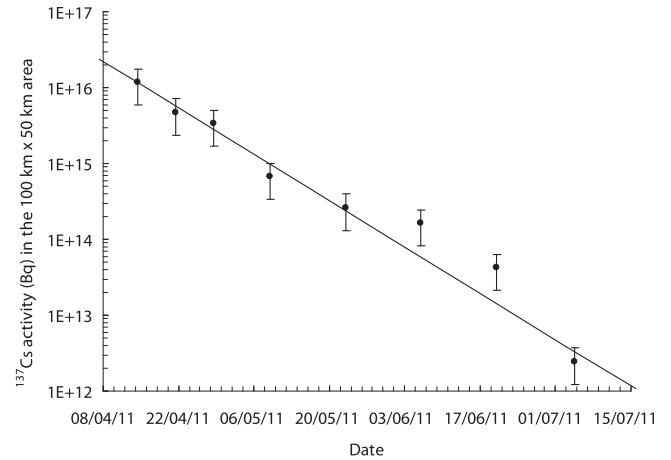


Fig. 5. Evolution of the ¹³⁷Cs quantities present in seawater at more than 5 km from the coast.

the main release period (March 26–April 8). The corresponding quantity is 22.0 PBq. A 95% confidence interval associated to the calculation of the linear regression gives values between 20.8 and 23.1 PBq. The main uncertainty is associated to the estimation of the thickness of the mixing layer (errors bars in Fig. 5). The same calculation with minimum and maximum values of the ¹³⁷Cs

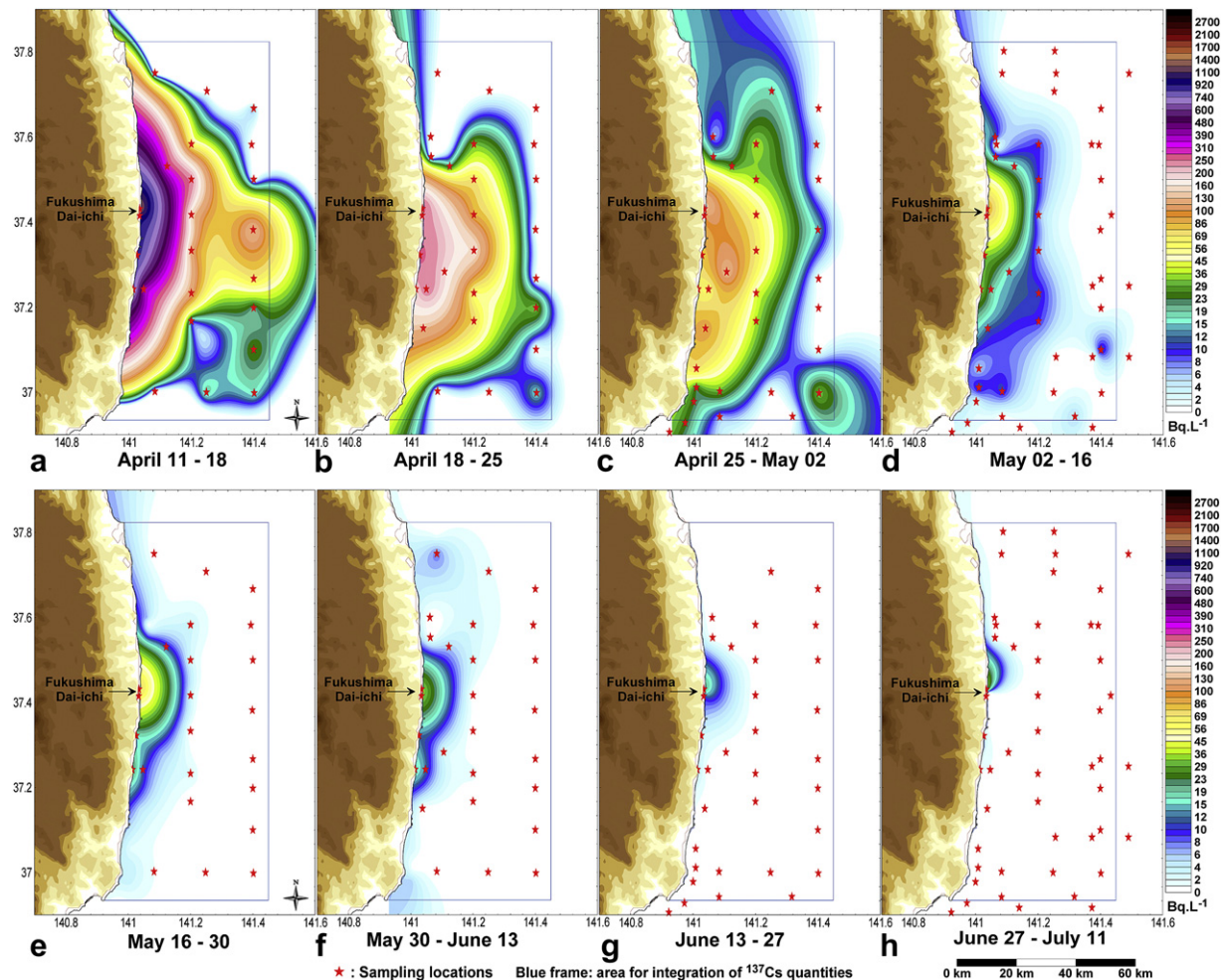


Fig. 4. Concentrations of ¹³⁷Cs in seawater between April 11 and July 11 2011.

quantities in the calculation area gives an interval of 10–34 PBq at the date of the April 8.

5. Flux estimation from ^{137}Cs measurements obtained 30 m north of Fukushima Dai-ichi

Data analysis showed that the sampling location situated 30 m north of Fukushima Dai-ichi discharge channel appeared to be represented by the general outflow from the plant (Fig. 6, left Y axis). It can be considered as a measurement of the direct flux of radionuclides from the facility. This assumes that dilution was stable and homogeneous at this location and that former releases did not influence the latter ones. We consider that the amount of 22 PBq deduced from measurements at the date of April 8 corresponds to the quantity of ^{137}Cs released from March 26 to April 8, when releases were the most important. It was then possible to associate the quantity of 22 PBq with the average concentration measured at this location during this period ($15\,716\text{ Bq.L}^{-1}$, number of values = 28, duration = 13.2 days). Fluxes of ^{137}Cs could be deduced from concentrations by applying the factor Quantity/(Concentration \times Duration). It gives $22\text{E}^{+15}/(15\,716 \times 13.2) = 1.06\text{E}^{+11}\text{ Bq.L}^{-1}/\text{Bq.day}^{-1}$. Fig. 6, right Y axis illustrates this conversion. It includes errors due to uncertainty on the calculation of 22 PBq of ^{137}Cs present on April 8 and does not take into account the release that left the inventory area between March 26 and April 8. Nevertheless, it provides an order of magnitude of the flux and shows the time-variation of releases. More accurate evaluation of the actual flux will be established by model/measurement comparisons. The quantity of ^{137}Cs released between March 25 and July 18 based on this calculation represented a total of 27 PBq. By accounting for the uncertainty on the 22 PBq value, the confidence interval for the total direct release is 12 PBq to 41 PBq. Releases after April 8 represented 18% of the total.

6. Fukushima accident release versus others sources to the ocean

Compared with other major origins of artificial radionuclides supplied to marine environment, Fukushima Dai-ichi direct liquid release represents the most important source-term ever known over such a short period of time and a located place. Table 2 synthesizes the major sources and releases conditions for ^{137}Cs .

The highest levels observed in open seas that could be compared to the concentration measured in the vicinity of Fukushima Dai-ichi are those measured in the Irish Sea in 1974–1976 with ^{137}Cs levels higher than 10 Bq.L^{-1} (Fig. 7). These values were one thousands times lower than the peak value observed close to Fukushima Dai-ichi plant, but they concerned the whole Irish Sea and persisted for years due to continuous controlled releases from Sellafield. Fukushima seawater peak labelling remained much more important during a short time. Nevertheless it appears that, for seawater concentrations, the duration of the major labelling from Fukushima direct releases will be about six-month. With the power law obtained in section 4, it could be calculated that the amount of ^{137}Cs present in the calculation area will be $171\text{E}^{+6}\text{ Bq}$ on October 11 and only 50 000 Bq on December 31. This estimation does not take into account potential residual releases from the plant, inputs from rivers, leaching from contaminated sediments and recirculation of contaminated waters.

Caesium essentially remains dissolved in seawater and it will be detectable for many years. The ^{134}Cs was released in similar quantities as ^{137}Cs . Their different radioactive decay half times, respectively 2 and 30 years, may be used to measure time-dependant labelling of water masses involved by Fukushima releases on the scale of North Pacific surface waters.

To give an order of magnitude of the potential labelling of oceanic waters, assuming that ^{137}Cs released into the sea during the

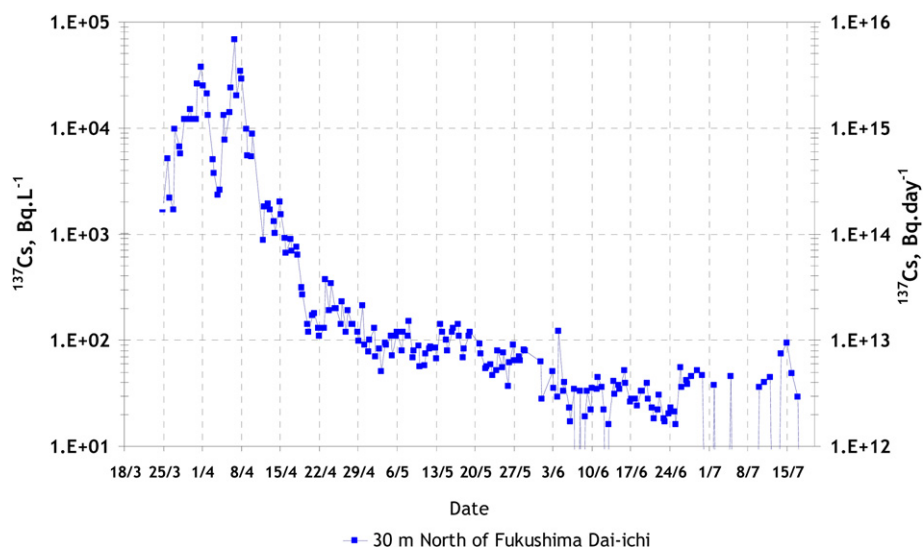


Fig. 6. Evolution of concentrations and corresponding fluxes of ^{137}Cs in seawater.

Table 2

Inventory of principal sources of ^{137}Cs in oceans in PBq (10^{15} Bq).

Nuclear tests atmospheric fallout	Reprocessing plants 1951–2010		Chernobyl accident	Solid waste dumping at sea (beta + gamma emitters)	Fukushima Dai-ichi	
	BNFL Sellafield	Areva-NC La Hague			Direct release	Atmospheric deposit
948 ^a	41.21	1.04	15–20 ^a	78–82 ^{a,b}	27 (12–41)	0.076 (80 km radius)

^a Aarkrog, 2003.

^b Linsley et al., 2005.

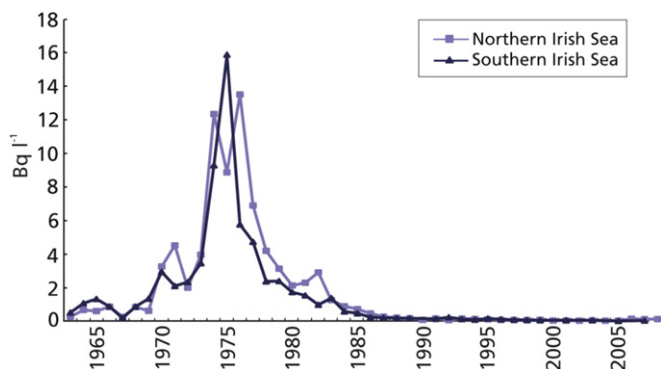


Fig. 7. Concentrations of ^{137}Cs in the Irish sea seawater (from RIFE, 2009).

accident was diluted in the Pacific oceanic seawater surface layer (for example: 100 m thickness, $180\text{E}^{+6}\text{ km}^2$), the expected concentration could rise by about $0.006\text{ Bq}\cdot\text{L}^{-1}$. This is a four-fold increase compared to that observed in the seawater off the Japanese coast before the accident (Nakanishi et al., 2011).

7. Conclusion

Contamination of the marine environment following the accident in the Fukushima Dai-ichi nuclear power plant represents the most important artificial radioactive direct liquid release into the sea ever known on a short time and space scale basis.

Quantities of ^{137}Cs in seawater within a $100\text{ km} \times 50\text{ km}$ area around the plant were calculated from interpolation of individual seawater measurements. The rate of seawater renewal was deduced from the time-evolution of these quantities. It appeared constant with an environmental half-time of 7 days.

These data allowed inferring the amount of direct liquid release after the end of the major releases, on April 8: 22 PBq (10 PBq–34 PBq). The evolution with time of the flux was calculated, based on concentration measurements close to the plant. It corresponded to a total of 27 PBq (12 PBq–41 PBq) of ^{137}Cs essentially from direct releases from March 21 to July 18. By this date, concentrations close the plant were often below the detection limit ($1\text{--}5\text{ Bq}\cdot\text{L}^{-1}$). More sensitive measurement methods should be used to continue to appraise residual inputs from the plant, river runoff and leakage from deposited sediments.

Estimation of the quantities of radioactive releases into the seawater could be improved by a better knowledge of the mixing layer in the area of measurement, model/measurement comparisons and better accuracy of seawater measurements. The main error was related to estimation of the thickness of the mixing layer; the uncertainty was estimated to be about 50% which mainly explain the confidence interval applied.

Caesium-137 and ^{134}Cs from Fukushima will remain detectable for several years around the North Pacific. Their ratio could be used as a tracer for studies on water masses pathways and transit times.

Even if their influence becomes weak, releases from Fukushima Dai-ichi represent a major source-term that must be accounted for in future studies of consequences of accidents in marine systems. Rate of dilution close to the plant means that radioprotection consequences of the accident would become low for pelagic species after six-months. It will be different for benthic species living close to the plant and species concerned by transfer through suspended matter coming from deposited sediments, rivers or desorption in seawater of radionuclides fixed onto sediments. Such processes have been clearly identified in the Irish Sea after Sellafield releases (Jones et al., 2007; Finegan et al., 2009). Coastal bottom sediment

covering was modified by the tsunami, and how this affects the radionuclide transfer is unknown. A steady state situation will take time to set up and a long-term monitoring will be certainly essential to assess the consequences for uses of marine resources.

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References

- Aarkrog, A., 2003. Input of anthropogenic radionuclides into the World Ocean. Deep Sea Research Part II: Topical Studies in Oceanography 50 (17–21), 2597–2606.
- Aoyama, M., Hirose, K., 2003. Temporal variation of ^{137}Cs water column inventory in the North Pacific since the 1960s. Journal of Environmental Radioactivity 69 (1–2), 107–117.
- Bailly du Bois, P., Salomon, J.C., Gandon, R., Guéguéniat, P., 1995. A quantitative estimate of English Channel water fluxes into the North Sea from 1987 to 1992 based on radiotracer distribution. Journal of Marine Systems 6 (5–6), 457–481.
- Bailly du Bois, P., Guéguéniat, P., 1999. Quantitative assessment of dissolved radiotracers in the English Channel: sources, average impact of la Hague reprocessing plant and conservative behaviour (1983, 1986, 1988 and 1994). Continental Shelf Research 19, 1977–2002. FluxManche II dedicated.
- Buesseler, K., Aoyama, M., Fukasawa, M., 2011. Impacts of the Fukushima nuclear power plants on marine radioactivity. Environmental Science & Technology 45 (23), 9931–9935.
- Finegan, P., Vintró, L.L., Mitchell, P.I., Boust, D., Gouzy, A., Kershaw, P.J., Lucey, J.A., 2009. Accumulation, solid partitioning and remobilisation of ^{99}Tc in subtidal and intertidal sediments in the Irish Sea. Continental Shelf Research 29 (16), 1995–2010.
- IRSN, 2011. IRSN Publishes Assessment of Radioactivity Released by the Fukushima Daiichi Nuclear Power Plant (Fukushima I) through 22 March 2011. http://www.irsn.fr/EN/news/Documents/IRSN_fukushima-radioactivity-released-assessment-EN.pdf.
- JAMSTEC – MERI Japan Agency for Marine-earth Science and Technology, Marine Ecology Research Institute, <http://www.godac.jamstec.go.jp/monitoringdata/>
- Jayne, S.R., Hogg, N.G., Waterman, S.N., Rainville, L., Donohue, K.A., Randolph Watts, D., Tracey, K.L., McClean, J.L., Maltrud, M.E., Qiu, B., Chen, S., Hacker, P., 2009. The Kuroshio Extension and its recirculation gyres. Deep Sea Research Part I: Oceanographic Research Papers 56 (12), 2088–2099.
- JHOD, Hydrographic and Oceanographic Department, Japan Coast Guard, Quick Bulletin of Ocean Conditions, http://www1.kaiho.mlit.go.jp/KANKYO/KAIYO/qboc/index_E.html
- JODC, 2011. JODC-Expert Grid data for Geography (J-EGG500), 500 m gridded bathymetric data set of Japan. http://www.jodc.go.jp/data_set/jodc/jegg_intro.html.
- Jones, D.G., Kershaw, P.J., McMahon, C.A., Milodowski, A.E., Murray, M., Hunt, G.J., 2007. Changing patterns of radionuclide distribution in Irish Sea subtidal sediments. Journal of Environmental Radioactivity 96 (Issues 1–3), 63–74.
- Lee, S., Povinec, P.P., Wyse, E., Pham, M.K., Hong, G., Chung, C., 2005. Distribution and inventories of ^{90}Sr , ^{137}Cs , ^{241}Am and Pu isotopes in sediments of the Northwest Pacific ocean. Marine Geology 216 (4), 249–263.
- Linsley, G., Sjoblom, K., Cabianna, T., 2005. Overview of Point Sources of Anthropogenic Radionuclides in the Oceans, vol. 6 Chapter 4, Issues 17–21, Pages 109–138.
- Livingston, H.D., Povinec, P.P., 2000. Anthropogenic marine radioactivity. Ocean & Coastal Management 43 (8–9), 689–712.
- Masson, O., Baeza, A., Bieringer, J., Brudecki, K., Bucci, S., Cappai, M., Carvalho, F.P., Connan, O., Cosma, C., Dalheimer, D., Didier, G., Depuydt, L.E., De Geer, A., De Vismes, L., Gini, F., Groppi, K., Gudnason, R., Gurriaran, D., Hainz, A., Halldorsson, O., Hammond, D., Hanley, O., Holey, K., Homoki, Z., Ioannidou, A., Isajenko, K., Jankovic, M., Katzberger, C., Kettunen, M., Kierepko, R., Kontro, R., Kwakman, P.J.M., Lecomte, M., Leon Vintro, L., Leppanen, A.P., Lind, B., Lujanovic, G., Mc Ginnity, P., Mahon, C.M., Mala, H., Manenti, S., Manolopoulou, R.M.W., Mattila, M., Mauring, A., Mietelski, A., Moller, J.W., Nielsen, B., Nikolic, S.P., Overwater, J., Palsson, S.E., Papastefanou, C., Penev, I.,

- Pham, M.K., Povinec, P.P., Rameback, H., Reis, M.C., Ringer, W., Rodriguez, A., Rulik, P., Saey, P.R.J., Samsonov, V., Schlosser, C., Sgorbati, G., Silobritiene, B.V., Soderstrom, C., Sogni, R., Solier, L., Sonck, M., Steinhäuser, G., Steinkopff, T., Steinmann, P., Stoulos, S., Sykora, I., Todorovic, D., Tooloutalaie, N., Tositti, L., Tschiersch, J., Ugron, A., Vagena, E., Vargas, A., Wershofen, H., Zhukova, O., 2011. Tracking of Airborne radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European Networks. *Environmental Science & Technology* 45 (18), 7670–7677.
- Mathieu, A., Benoit, J.-P., Didier, D., Groëll, J., Isnard, O., Korsakissok, I., Quélo, D., Quentric, E., Saunier O. and Tombette, M., 2011. Radioactive dispersion in the atmosphere. Submitted to *Elements* (special Fukushima issue).
- McCartney, M., Kershaw, P., Woodhead, D., Denoon, D., 1994. Artificial radionuclides in the surface sediments of the Irish Sea, 1968–1988. *Science of the Total Environment* 141 (1–3), 103–138.
- Moon, D., Hong, G., Kim, Y.I., Baskaran, M., Chung, C.S., Kim, S.H., 2003. Accumulation of anthropogenic and natural radionuclides in bottom sediments of the Northwest Pacific Ocean. *Deep Sea Research Part II: Topical Studies in Oceanography* 50 (17–21), 2649–2673.
- MEXT Monitoring Measurements following Fukushima Accident. <http://www.mext.go.jp/english/incident>
- Nakanishi, T., Aono, T., Yamada, M., Kusakabe, M., 2010. Temporal and spatial variations of ^{137}Cs in the waters off a nuclear fuel reprocessing facility in Rokkasho, Aomori, Japan. *Journal of Radioanalytical and Nuclear Chemistry* 283 (3), 831–838.
- Nakanishi, T., Zheng, J., Aono, T., Yamada, M., Kusakabe, M., 2011. Vertical distributions of ^{99}Tc and the $^{99}\text{Tc}/^{137}\text{Cs}$ activity ratio in the coastal water off Aomori, Japan. *Journal of Environmental Radioactivity* 102 (8), 774–779.
- Pittauerová, D., Hettwig, B., Fischer, H.W., 2011. Fukushima fallout in Northwest German environmental media. *Journal of Environmental Radioactivity* 102 (9), 877–880.
- Povinec, P.P., Hirose, K., Honda, T., Ito, T., Scott, E.M., Togawa, O., 2004. Spatial distribution of ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in surface waters of the Pacific and Indian Oceans—GLOMARD database. *Journal of Environmental Radioactivity* 76 (1–2), 113–137.
- Pryor, S.C., Barthelmie, R.J., Geernaert, L.L.S., Ellermann, T., Perry, K.D., 1999. Speciated particle dry deposition to the sea surface: results from ASEPS '97. *Atmospheric Environment* 33 (13), 2045–2058.
- Quélo, D., Groëll, J., Didier, D., Mathieu, A., Korsakissok, I., Tombette, M., Quentric, E., Benoit, J.-P., Isnard, O., 2011. Atmospheric transport modelling and situation assessment of the Fukushima accident. In: 15th Annual George Mason University Conference on Atmospheric Transport and Dispersion Modeling, Virginia, USA.
- RIFE, 2009. RIFE – 14 Radioactivity in Food and the Environment, RIFE 2008. CEFAS – Center for Environment, Fisheries and Aquaculture Science, Environment Agency, Environment and Heritage Service, Food Standards Agency and Scottish Environment Protection Agency, Bristol, Belfast, London and Stirling, ISSN 1365–6414 pp. 248.
- Sanchez-Cabeza, J., Levy, I., Gastaud, J., Eriksson, M., Osvath, I., Aoyama, M., 2011. Transport of North Pacific ^{137}Cs labeled waters to the south-eastern Atlantic Ocean. *Progress in Oceanography* 89 (1–4), 31–37.
- Soulhac, L., Didier, D., 2008. Projet pX, note de principe pX 1.0. Note technique IRSN/DEI/SESUC/08–39. IRSN.
- Sportisse, B., 2007. A review of parameterization for modelling dry deposition and scavenging of radionuclides. *Atmospheric Environment* 41, 2683–2698.
- TEPCO. Monitoring Measurements following Fukushima Accident. www.tepco.co.jp/en/index-e.html
- TEPCO, 2011. The Great East Japan Earthquake and Current Status of Nuclear power Stations. http://www.tepco.co.jp/en/nu/fukushima-np/f1/images/f12np-gaiyou_e.pdf.