



**HAL**  
open science

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

P. Bailly Du Bois, P. Laguionie, D. Boust, I. Korsakissok, D. Didier, B. Fiévet

► **To cite this version:**

P. Bailly Du Bois, P. Laguionie, D. Boust, I. Korsakissok, D. Didier, et al.. Estimation of marine source-term following Fukushima Dai-ichi accident. *Journal of Environmental Radioactivity*, 2012, 114, pp.2-9. 10.1016/j.jenvrad.2011.11.015 . hal-02433222

**HAL Id: hal-02433222**

**<https://normandie-univ.hal.science/hal-02433222>**

Submitted on 14 Jan 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

P. Bailly du Bois<sup>1</sup>, P. Laguionie<sup>1</sup>, D. Boust<sup>1</sup>, I. Korsakissok<sup>2</sup>, D. Didier<sup>2</sup>, Fiévet<sup>1</sup> B.

<sup>1</sup> IRSN/DEI/SECRE/LRC - Institut de Radioprotection et de Sûreté Nucléaire, Direction de l'Environnement et de l'Intervention, Laboratoire de Radioécologie de Cherbourg - Octeville, rue Max Pol Fouchet, B.P. 10, 50130 Cherbourg-Octeville – France.

<sup>2</sup> IRSN/DEI/SESUC/BMTA - Institut de Radioprotection et de Sûreté Nucléaire, Direction de l'Environnement et de l'Intervention, Bureau de Modélisation des Transferts Atmosphériques – avenue de la Division Leclerc, BP 17, 92262 Fontenay-aux-roses, France.

### Correspondence:

P. Bailly du Bois      Surname: Bailly du Bois    First name: Pascal  
Laboratoire de Radioécologie de Cherbourg - Octeville (LRC)  
IRSN-LRC  
rue Max Pol Fouchet  
B.P. 10  
50130 Cherbourg-Octeville      France  
Tel. direct line:      (33) 02 33 01 41 05  
Tel. secretary:      (33) 02 33 01 41 00  
FAX:                    (33) 02 33 01 41 30  
Email:                    pascal.bailly-du-bois@irsn.fr

Received at Editorial Office: 29 Jul 2011

Article accepted with minor revision: 14 Nov 2011

Article revised: 17 Nov 2011

Article accepted for publication: 17 Nov 2011

DOI: 10.1016/j.jenvrad.2011.11.015

---

### 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<sup>-1</sup> for <sup>134</sup>Cs and <sup>137</sup>Cs, and exceeded 100 000 Bq.L<sup>-1</sup> for <sup>131</sup>I 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 <sup>137</sup>Cs in seawater in a 50-km area around the plant were calculated from interpolation of seawater measurements. The environmental half-time of seawater in this area is deduced from the time-evolution of these quantities. This half-time appeared constant with a period of seven days to divide by a factor of two the measured quantities of <sup>137</sup>Cs. 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 <sup>137</sup>Cs 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 <sup>137</sup>Cs from Fukushima will remain detectable for several years throughout the North Pacific, and <sup>137</sup>Cs/<sup>134</sup>Cs ratio will be a tracer for future studies.

---

### Highlights

> Fukushima Dai-ichi accident is the most important artificial radioactive release flux into the sea. > Quantities of <sup>137</sup>Cs in seawater are deduced from individual measurements. > Local concentrations in seawater diminish regularly by a factor of two in seven days. > Total amount of direct releases of <sup>137</sup>Cs estimated is 27 PBq (12 PBq - 41 PBq). > Time-evolution of release fluxes is drawn.

Keywords: Fukushima; source-term; <sup>137</sup>Cs; <sup>134</sup>Cs; <sup>131</sup>I; environmental half time

## 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 toward 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). 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  $^{137}\text{Cs}$  and  $^{131}\text{I}$  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; Livingstone and Povinec, 2000; Povinec et al., 2004; Sanchez 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; 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  $^{137}\text{Cs}$  for other radionuclides in the source-terms,  $^{137}\text{Cs}$  fluxes give a good estimate of other radionuclides released at the same time. Activity ratios of other radionuclides versus  $^{137}\text{Cs}$  were derived when data were available. To facilitate comparison between different labelling, all radionuclide/ $^{137}\text{Cs}$  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/ $^{137}\text{Cs}$ ).

The paper describes briefly the hydrodynamic of the zone. The different radionuclide sources are presented and estimation of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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 hours. 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  $^{137}\text{Cs}$ , concentrations were in the range 1 - 4  $\text{Bq}\cdot\text{m}^{-3}$  (Nakanishi et al., 2011 ; 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  $\text{Bq}\cdot\text{m}^{-3}$  for  $^{137}\text{Cs}$  (Nakanishi, 2011). After

the accident, measured concentrations in a 30 km perimeter around the plant exceeded  $10 \text{ Bq.L}^{-1}$  ( $10\,000 \text{ Bq.m}^{-3}$ ) and reached  $68\,000 \text{ Bq.L}^{-1}$  close to the plant ( $68\text{E}^6 \text{ Bq.m}^{-3}$ ).

The radionuclides with short radioactive half-life (less than a few tens of days, like  $^{131}\text{I}$ ) ceased to be detectable after a few months and should not have any large scale and long-term impact. Others, such as  $^{134}\text{Cs}$  and  $^{106}\text{Ru}$  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 seabed. The concentrations in  $^{131}\text{I}$  and  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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  $13 \text{ Bq.L}^{-1}$  for  $^{137}\text{Cs}$  with  $\text{IR}(^{131}\text{I}/^{137}\text{Cs})$  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  $20 - 100 \text{ Bq.L}^{-1}$  of  $^{137}\text{Cs}$  with an activity ratio  $^{131}\text{I}/^{137}\text{Cs}$  ( $\text{IR}(^{131}\text{I}/^{137}\text{Cs})$ ) 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 wash out 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  $^{137}\text{Cs}$  deposited on the sea surface was derived from atmospheric dispersion calculations using the Gaussian puff model of the IRSN technical crisis centre, named pX (Souhlaç and Didier, 2008). The model was forced with 3D meteorological forecasts at a resolution of  $0.125^\circ$  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 sea water are lower than over lands: a value of  $5.10^{-5} \text{ m}^2 \cdot \text{s}^{-1}$  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  $\text{A}p_0^B$ , with  $\text{A}=5.10^{-5} \text{ hr.mm}^{-1} \cdot \text{s}^{-1}$ ,  $\text{B}=1$ , and  $p_0$  the rain intensity in  $\text{mm.hr}^{-1}$  (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.

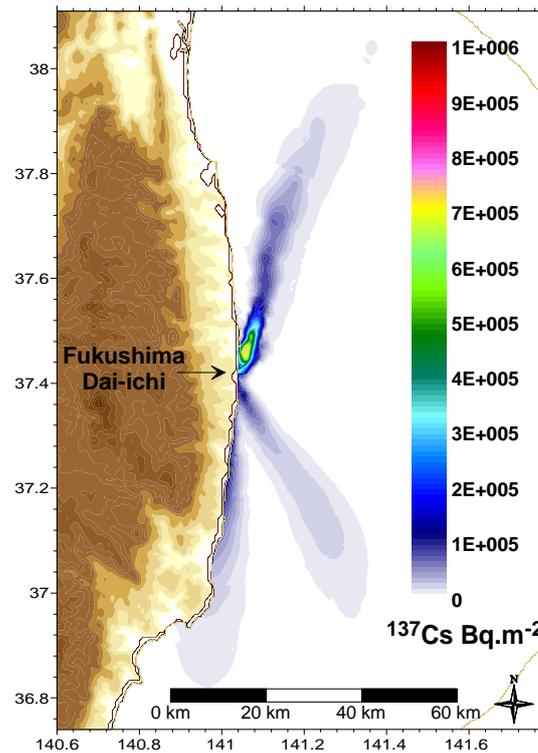


Fig. 1 Calculated cumulate deposit of <sup>137</sup>Cs, 2011 April 23

### 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 run-off. 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 sea water 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 Bq.L<sup>-1</sup> of <sup>137</sup>Cs, maximum of 68 000 Bq.L<sup>-1</sup>, Fig. 2). The IR(<sup>131</sup>I/<sup>137</sup>Cs) was roughly homogeneous during this period, with a slight trend to decrease from 24 to 18. It revealed a regular process of <sup>131</sup>I elimination (in the plant or in seawater) with a halftime 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.

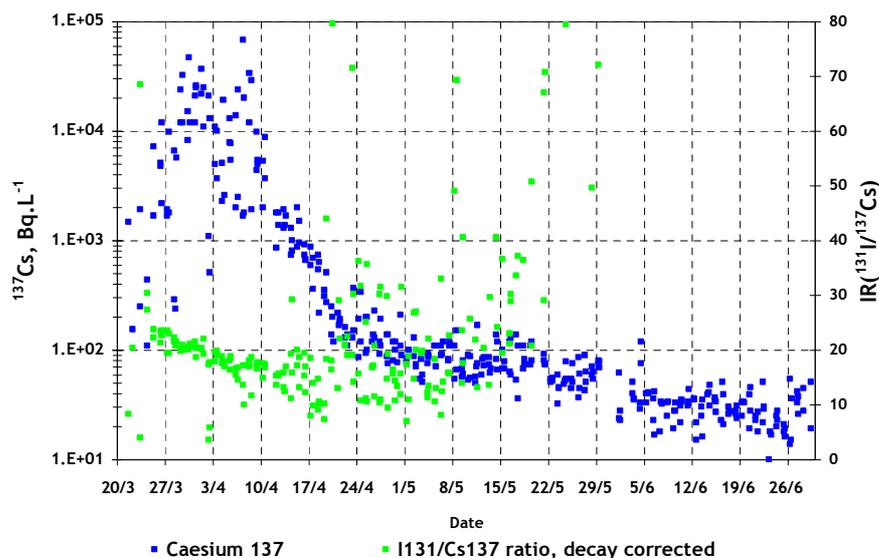


Fig. 2 Evolution of  $^{137}\text{Cs}$  concentrations and  $^{131}\text{I}/^{137}\text{Cs}$  ratios in seawater at less than 2 km from the Fukushima Dai-ichi power plant.

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 – 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.

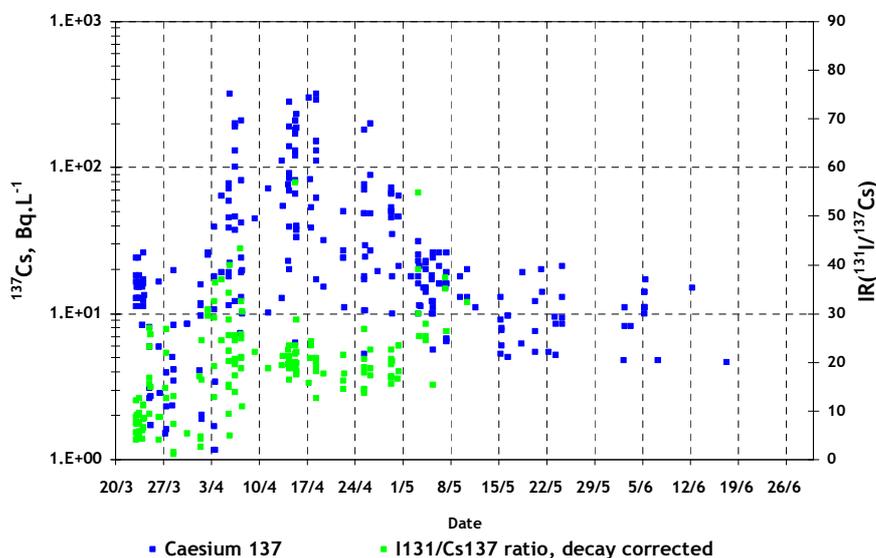


Fig. 3 Evolution of  $^{137}\text{Cs}$  concentrations and  $^{131}\text{I}/^{137}\text{Cs}$  ratios in seawater at more than 5 km from the coast.

Regarding other radionuclides detected in seawater with a radioactive decay halftime 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  $\text{Bq.L}^{-1}$ .

#### 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 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 km x 50 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 ( $63\,000 - 5\text{ Bq}\cdot\text{L}^{-1}$ ) 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  $^{137}\text{Cs}$  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 ca. 50%.

Figure 4 shows the evolution of  $^{137}\text{Cs}$  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 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.

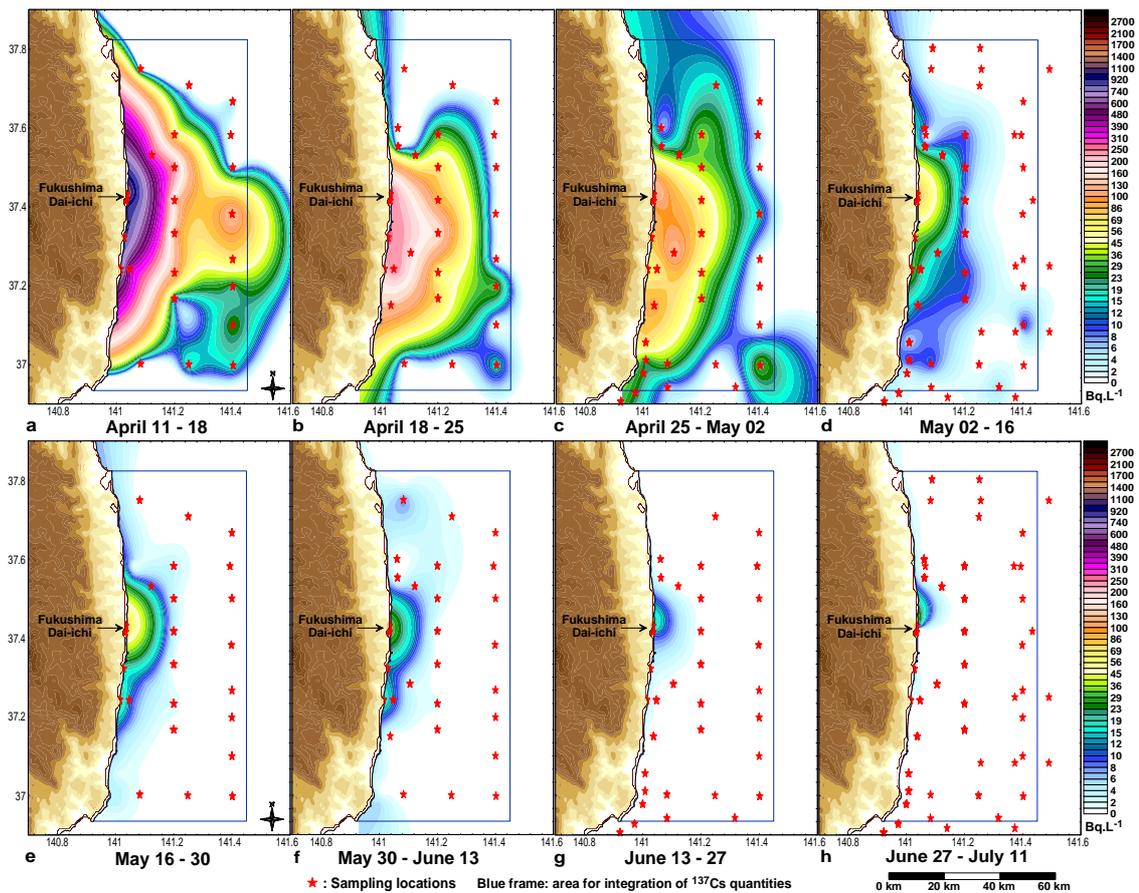


Fig. 4 Concentrations of  $^{137}\text{Cs}$  in seawater between April 11 and July 11 2011.

Measurement period			Number of measurement	<sup>137</sup> 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

Table 1 <sup>137</sup>Cs Quantities from interpolation of individual measurements in seawater in a 100 x 50 km box off Fukushima Dai-ichi plant (see blue frames in Fig. 4)

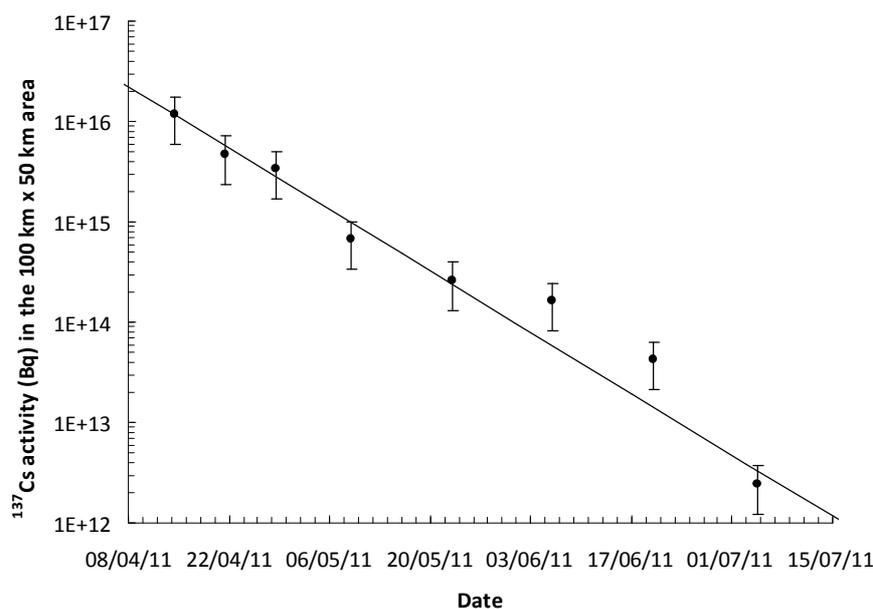


Fig. 5 Evolution of the <sup>137</sup>Cs quantities present in seawater at more than 5km from the coast.

Extrapolation of the regression curve at the date of the April 8 allows estimating the total amount of <sup>137</sup>Cs discharged at the end of 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 figure 5). The same calculation with minimum and maximum values of the <sup>137</sup>Cs quantities in the calculation area gives an interval of 10 to 34 PBq at the date of the April 8.

## 5 Flux estimation from <sup>137</sup>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 <sup>137</sup>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 Bq.L<sup>-1</sup>, number of values = 28, duration = 13.2 days). Fluxes of <sup>137</sup>Cs could be deduced from concentrations by applying the factor Quantity/(Concentration x Duration). It gives 22E+15/(15 716 x 13.2) = 1.06E+11 Bq.L<sup>-1</sup> / Bq.day<sup>-1</sup>. Fig. 6, right Y axis illustrates this conversion. It includes errors due to uncertainty on the calculation of 22 PBq of <sup>137</sup>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 <sup>137</sup>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 22PBq value, the confidence interval for the total direct release is 12 PBq to 41 PBq. Releases after April 8 represented 18% of the total.

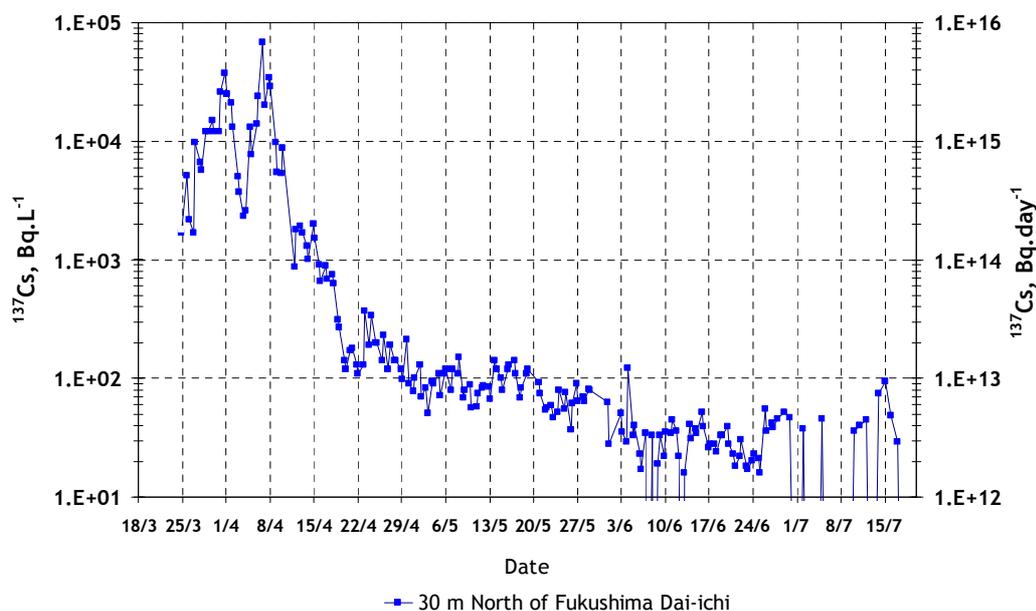


Fig. 6 Evolution of concentrations and corresponding fluxes of  $^{137}\text{Cs}$  in seawater.

## 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  $^{137}\text{Cs}$ .

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 <sup>a</sup>	41.21	1.04	15-20 <sup>a</sup>	78 - 82 <sup>a, b</sup>	27 (12 - 41)	0.076 (80 km radius)

<sup>a</sup>: Aarkrog, 2003; <sup>b</sup>: Linsley et al., 2005

Table 2 Inventory of principal sources of  $^{137}\text{Cs}$  in oceans in PBq ( $10^{15}\text{Bq}$ ).

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  $^{137}\text{Cs}$  levels higher than  $10\text{ Bq.L}^{-1}$  (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  $^{137}\text{Cs}$  present in the calculation area will be  $171\text{E}^{+6}\text{ Bq}$  on October 11 and only  $50\,000\text{ Bq}$  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.

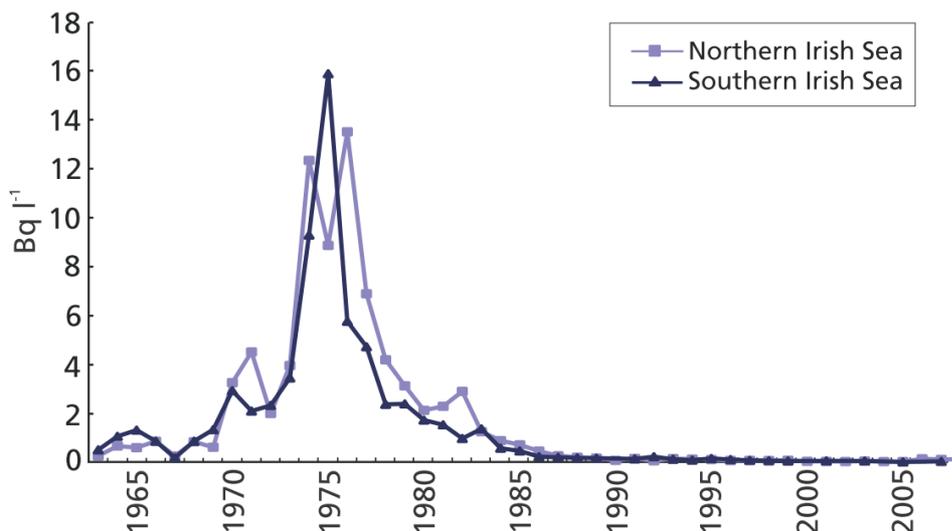


Fig. 7 Concentrations of  $^{137}\text{Cs}$  in the Irish Sea seawater (from Rife, 2009)

Caesium essentially remains dissolved in seawater and it will be detectable for many years. The  $^{134}\text{Cs}$  was released in similar quantities as  $^{137}\text{Cs}$ . 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  $^{137}\text{Cs}$  released into the sea during the accident was diluted in the Pacific oceanic seawater surface layer (for example: 100 metres thickness,  $180\text{E}^{+6}\text{ km}^2$ ), the expected concentration could rise by about  $0.006\text{ Bq.L}^{-1}$ . 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  $^{137}\text{Cs}$  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 halftime of 7 days: time-lapse for a 2-fold decrease of  $^{137}\text{Cs}$  quantities in the inventory area.

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  $^{137}\text{Cs}$  essentially from direct releases from March 21 to July 18. By this date, concentrations close the plant were often below the detection limit ( $1 - 5\text{ Bq.L}^{-1}$ ). 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 ca. 50 % which mainly explain the confidence interval applied.

Caesium-137 and  $^{134}\text{Cs}$  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.

## 8 Acknowledgements

We are very much concerned by the consequences of the tsunami and want to express all our sympathy and support to Japanese people.

We thank MEXT and TEPCO for the Internet publication of the concentration data collected in the environment and that were used in the present study.

We are indebted to the “Centre Technique de Crise” staff of the French Institute of Radioprotection and Nuclear Safety who helped collecting the data on the Internet and thank specially Celine Duffa, Mireille Arnaud, Olivier Connan and Vanessa Parache for compiling and checking thousands of measurements.

We thank Takuya Kobayashi for his advises and connections with Japanese institutes.

## 9 References

- Aarkrog, A., 2003. Input of anthropogenic radionuclides into the World Ocean. *Deep Sea Research Part II: Topical Studies in Oceanography* Volume 50, Issues 17-21, Pages 2597-2606.
- Aoyama, M., Hirose, K., 2003. Temporal variation of <sup>137</sup>Cs water column inventory in the North Pacific since the 1960s. *Journal of Environmental Radioactivity* Volume 69, Issues 1-2, Pages 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 ; FluxManche II dedicated volume* 19, 1977-2002.
- Finegan, P., Vitró, L.L., Mitchell, P.I., Boust, D., Gouzy, A., Kershaw, P.J., Lucey, J.A., 2009. Accumulation, solid partitioning and remobilisation of <sup>99</sup>Tc in subtidal and intertidal sediments in the Irish Sea. *Continental Shelf Research* Volume 29, Issues 16, Pages 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](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* Volume 56, Issues 12, Pages 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](http://www1.kaiho.mlit.go.jp/KANKYO/KAIYO/qboc/index_E.html)
- JODC, 2011, JODC-Expert Grid data for Geography (J-EGG500), 500m gridded bathymetric data set of Japan. [http://www.jodc.go.jp/data\\_set/jodc/jegg\\_intro.html](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* Volume 96, Issues 1-3, Pages 63-74.
- Lee, S., Povinec, P.P., Wyse, E., Pham, M.K., Hong, G., Chung, C., 2005. Distribution and inventories of <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>241</sup>Am and Pu isotopes in sediments of the Northwest Pacific Ocean. *Marine Geology* Volume 216, Issues 4, Pages 249-263.
- Linsley, G., Sjöblom, K., Cabianca, T., 2005. Overview of point sources of anthropogenic radionuclides in the oceans. *Volume Volume 6, Chapter 4, Issues 17-21, Pages 109-138.*
- Livingston, H.D., Povinec, P.P., 2000. Anthropogenic marine radioactivity. *Ocean & Coastal Management* Volume 43, Issues 8-9, Pages 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., Katzlberger, C., Kettunen, M., Kierepko, R., Kontro, R., Kwakman, P.J., M., Lecomte, M., Leon Vintro, L., Leppanen, A.P., Lind, B., Lujaniene, 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., Steinhauser, 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* Volume 45, Issue 18, Pages 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* Volume 141, Issues 1-3, Pages 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* Volume 50, Issues 17-21, Pages 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 <sup>137</sup>Cs in the waters off a nuclear fuel reprocessing facility in Rokkasho, Aomori, Japan. *Journal of Radioanalytical and Nuclear Chemistry* Volume 283, Issues 3, Pages 831-838.
- Nakanishi, T., Zheng, J., Aono, T., Yamada, M., Kusakabe, M., 2011. Vertical distributions of <sup>99</sup>Tc and the <sup>99</sup>Tc/<sup>137</sup>Cs activity ratio in the coastal water off Aomori, Japan. *Journal of Environmental Radioactivity* Volume 102, Issues 8, Pages 774-779.
- Pittauerová, D., Hettwig, B., Fischer, H.W., 2011. Fukushima fallout in Northwest German environmental media. *Journal of Environmental Radioactivity* Volume 102, Issues 9, Pages 877-880.
- Povinec, P.P., Hirose, K., Honda, T., Ito, T., Scott, E.M., Togawa, O., 2004. Spatial distribution of <sup>3</sup>H, <sup>90</sup>Sr, <sup>137</sup>Cs and <sup>239,240</sup>Pu in surface waters of the Pacific and Indian Oceans--GLOMARD database. *Journal of Environmental Radioactivity* Volume 76, Issues 1-2, Pages 113-137.
- Pryor, S. C., Barthelmie, R. J., Geernaert, L. L. S., Ellermann T. and Perry, K. D., 1999. Speciated particle dry deposition to the sea surface: results from ASEPS '97. *Atmospheric Environment* Volume 33, Issue 13, Pages 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. 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 248p.
- Sanchez-Cabeza, J., Levy, I., Gastaud, J., Eriksson, M., Osvath, I., Aoyama, M., 2011. Transport of North Pacific <sup>137</sup>Cs labeled waters to the south-eastern Atlantic Ocean. *Progress In Oceanography* Volume 89, Issues 1-4, Pages 31-37.
- Soulhac, L. and D. Didier, 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*, Volume 41, Pages 2683-2698.
- TEPCO, Monitoring measurements following Fukushima accident. [www.tepco.co.jp/en/index-e.html](http://www.tepco.co.jp/en/index-e.html)
- TEPCO, Monitoring measurements following Fukushima accident. [www.tepco.co.jp/en/index-e.html](http://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](http://www.tepco.co.jp/en/nu/fukushima-np/f1/images/f12np-gaiyou_e.pdf)