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Review article

An overview of current knowledge concerning the health and environmental consequences of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident

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ABSTRACT

Since 2011, the scientific community has worked to identify the exact transport and deposition patterns of radionuclides released from the accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in Japan. Nevertheless, there still remain many unknowns concerning the health and environmental impacts of these radionuclides. The present paper reviews the current understanding of the FDNPP accident with respect to interactions of the released radionuclides with the environment and impacts on human and non-human biota. Here, we scrutinize existing literature and combine and interpret observations and modeling assessments derived after Fukushima. Finally, we discuss the behavior and applications of radionuclides that might be used as tracers of environmental processes. This review focuses on ^{137}Cs and ^{131}I releases derived from Fukushima. Published estimates suggest total release amounts of 12–36.7 PBq of ^{137}Cs and 150–160 PBq of ^{131}I. Maximum estimated human mortality due to the Fukushima nuclear accident is 10,000 (due to all causes) and the maximum estimates for lifetime cancer mortality and morbidity are 1500 and 1800, respectively. Studies of plants and animals in the forests of Fukushima have recorded a range of physiological, developmental, morphological, and behavioral consequences of exposure to radioactivity. Some of the effects observed in the exposed populations include the following: hematological aberrations in Fukushima monkeys; genetic, developmental and morphological aberrations in a butterfly; declines in abundances of birds, butterflies and cicadas; aberrant growth forms in trees; and morphological abnormalities in aphids. These findings are discussed from the perspective of conservation biology.

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1. Introduction

The accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in Japan on March 11, 2011 resulted in a release of about 73 radionuclides (135 in total including radioactive progeny) (IRSN, 2012). It was a direct consequence of a high-magnitude earthquake (~9.0), which occurred in the Pacific Ocean near Japan's east coast, creating two massive tsunamis that in turn struck Japan about 1 h following the earthquake (Akahane et al., 2012). The main electric power grid was disabled by the earthquake and seawater intrusion to the backup power supply caused power loss and the cooling systems of the four nuclear reactors were disrupted, leading to increased pressure levels and the production of hydrogen gas due to the extreme heating of the cooling water. During the following days, hydrogen explosions released large amounts of radioactivity into the atmosphere. Fearing more severe damage and larger releases, plant managers ordered seawater to be used as a cooling medium (IRSN, 2012). This highly radioactive water was discharged to the Pacific Ocean, with discharges (to both land and sea) ongoing more than three years following the accident (Kumamoto et al., 2015).

Subsequently, several studies were conducted to document global transport and deposition of the most important fallout radionuclides (e.g. Christoudias and Lelieveld, 2013; Evangeliou et al., 2013b; Kristiansen et al., 2012) and the potential impacts to human populations (e.g. Evangeliou et al., 2014a; Ten Hoeve and Jacobson, 2012; WHO, 2013) and to animals and plants (e.g. Aliyu et al., 2015a; Evrard et al., 2012; Garnier-Laplace et al., 2011; Hiyama et al., 2012; Møller et al., 2012; Møller and Mousseau, 2011b). Simultaneously, many research groups initiated experimental monitoring programs to assess potential consequences in Europe, Asia, USA and Japan (e.g. Evrard et al., 2012; Kim et al., 2012; Kinoshita et al., 2011; Kritidis et al., 2012; Truong et al., 2012; MacMullin et al., 2012; Paatero et al., 2012; Pham et al., 2012; Povinec et al., 2012a).

Some of the major radioactive fission products released into the atmosphere and the ocean included ^{131}I ($t_{1/2}$ = 8.02 days), ^{134}Cs ($t_{1/2}$ = 2.06 years), ^{137}Cs ($t_{1/2}$ = 30.07 years) and ^{90}Sr ($t_{1/2}$ = 28.78 years), which were then dispersed globally by the prevailing winds (Masson et al., 2011) resulting in the contamination of both terrestrial and marine ecosystems (Butler, 2011; Chino et al., 2011; NSCJ, 2011).

Many fundamental questions concerning the FDNPP accident still need to be resolved. For example, what was the exact amount of the released radioactivity? How is the environment being affected? What are the predicted consequences for human populations? The present paper aims to review the existing literature related to the FDNPP accident in order to address these questions. Furthermore, we hope to stimulate further discussion among the scientific and regulatory communities concerned with nuclear safety and radiation protection (e.g., CTBTO, IAEA, UNSCEAR, ICRP, etc.).

2. Methods of literature search

The discussion in this paper is based on keyword searches in Google Scholar, Scopus and Web of Science. The keywords 'source term', 'radioactive contamination', 'human health' and 'biota' were combined with the primary keyword 'Fukushima' during literature searches. The inclusion criteria for all publications were as follows: (i) they discuss the release of radionuclides from FDNPP to the air or sea; (ii) they highlight the impacts of the FDNPP accident on either human or non-human biota; (iii) they report the specific activities of fission products at Fukushima prefecture and some parts of Japan pre- and post- FDNPP accident. We conducted more than 200 literature searches to arrive at over 170 publications included in this review. A few articles which considered the radioecological impacts of the Chernobyl nuclear accident and fallout from atomic weapons tests were included for the purpose of comparisons with the manifested impacts of the FDNPP accident.

3. Source term assessment

Following the FDNPP accident, there was much public concern about the release of radioactivity into the atmosphere and ocean, and many independent groups attempted to estimate total release amounts. For the releases to the atmosphere, the Japanese government estimated the source terms for ^{131}I and ^{137}Cs to be 160 PBq and 15 PBq, respectively (RJG, 2011). The Nuclear Safety Commission of Japan (NSCJ) estimated that the total amounts of ^{131}I and ^{137}Cs released into the atmosphere were 150 PBq and 12 PBq, respectively (NSCJ, 2011). Masson et al. (2011) estimated that 153 PBq ^{131}I and 13 PBq ^{137}Cs were released, whereas other researchers reported the total emission of ^{137}Cs to be as high as 13–35.8 PBq (Chino et al., 2011; Stohl et al., 2012). In order to estimate the source term of ^{137}Cs and ^{131}I, Chino et al. (2011) adopted an estimation method using an atmospheric dispersion model. Their results (~13 PBq) were comparable to those of the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT, 2011) and have been validated by subsequent analyses (Katata et al., 2012; Terada et al., 2012). In a more recent study, Katata et al. (2015a) modified the computational methodology to address some of the limitations of previous studies (Katata et al., 2012; Terada et al., 2012). They considered a new scheme of wet, dry, and fog-water parameterization of both gaseous and particulate radionuclides in their simulation. Observations from both land in different parts of Japan and the Pacific Ocean were coupled to this new model, leading to estimated total release amounts of 151 PBq of ^{131}I and 14.5 PBq of ^{137}Cs. The reported total release amount of ^{131}I accords with the estimate of the Nuclear Safety Commission of Japan (NSCJ, 2011) and lies within the range for total release amount of ^{131}I by this review (150–160) PBq (Table 1). However, Chino et al. (2011) Terada et al. (2012) and Katata et al. (2015a) estimated the total release of ^{137}Cs (13–15 PBq), based on an inverse estimation of the source term by coupling Japanese environmental monitoring data with regional atmospheric dispersion simulations. In other words, they used data from Japanese stations only and a regional simulation domain and assumed constant radioactivity ratios for the different radionuclides based on iodine and cesium concentrations in rain, snow and vegetation. Chino et al. (2011) noted that estimates of release amounts using this methodology are associated with an uncertainty of at least a factor of five.

Other research groups and national laboratories have also estimated the total release amounts of ^{131}I and ^{137}Cs from FDNPP, reporting releases that are higher than the Japanese estimates. For example, the French Institute of Radioprotection and Nuclear Safety (IRSN, 2012) reported releases of ^{137}Cs of 20.6 PBq—25% higher than those of Katata et al. (2015a). In addition, Stohl et al. (2012) used an inverse modeling technique employing data from the Comprehensive Nuclear Test-Ban Treaty Organization (CTBTO) measurement network to address emission inventories of ^{137}Cs and ^{133}Xe. They reported ^{137}Cs emissions of 36.7 PBq and ^{133}Xe 15.3 EBq ($\times 10^{18}$ Bq) based on a larger number of observations. The estimates of Stohl et al. (2012) found 2.5 times higher emission rates compared to that of Chino et al. (2011); Terada et al. (2012) and Katata et al. (2015a). However, the authors state that their estimates are associated by an uncertainty of about 50%.

Table 1
Source term of anthropogenic radionuclides in the atmosphere and the ocean (PBq).

Radionuclide	Global fallout		Chernobyl		Fukushima	
	Atmosphere	Ocean	Atmosphere	Ocean	Atmosphere	Ocean
^{131}I	–	–	1760[b]	–	150–160[c]	–
^{132}I					35.8[d]	
^{137}Cs	950[a]	600[a]	85[b]	16[b]	12–20[e]	4–27[h]
					37 ± 18[f]	
^{133}Xe					15,300 ± 7650[f]	
All noble gases					12,134[d]	
^{90}Sr	600[a]	380[a]	1[b]	–	0.01–0.14[g]	0.1–2.2[g]
^{239,240}Pu	10.87[a]	6.6[a]	0.087[b]	–	(1–2.4) × 10^{-6}[g]	–

[a] UNSCEAR (2000).
[b] NEA (2002).
[c] Masson et al. (2011), NSCJ (2011), RJG (2011).
[d] IRSN (2012).
[e] IRSN (2012), Masson et al. (2011), NSCJ (2011), RJG (2011).
[f] Stohl et al. (2012).
[g] Povinec et al. (2012a).
[h] Bailly du Bois et al. (2012), Kawamura et al. (2011), Tsumune et al. (2012).

The Science Council of Japan (SCJ, 2014) compared various model estimates for the total amounts of radioactive materials released from Fukushima. They adopted the source term estimation of Stohl et al. (2012) as a prior emission in order to compare the discrepancy between field observations and computed concentrations. The SCJ's approach documented the total release of ^{137}Cs between March 11 and 19 as 19.4 ± 3.6 PBq. This value lies between the estimates of Terada et al. (2012) and Stohl et al. (2012), but they are closer to those of the IRSN (2012). However, the SCJ noted some limitations in their analysis. For example, only one Eulerian aerosol model (MASINGAR), developed by Tanaka and Chiba (2005), was used and the bias of model transport could affect the source term estimate. In order to avoid such problems, various model simulations with common experimental settings are usually used and the source term estimates are compared. Another issue is the poor horizontal resolution of the global Eulerian aerosol model; this can be eliminated using a regional chemical transport model and collecting hourly observation data.

More recently, Saunier et al. (2013) used gamma dose rate observations to estimate the source term using inverse modeling. They employed a Eulerian model developed by the IRSN and calculated that 105.9 PBq of ^{131}I, 35.8 PBq of ^{132}I, 15.5 PBq of ^{137}Cs and 12,134 PBq of noble gases were emitted. The details of the theoretical formulation of the IRSN model are available in the literature (e.g. Isnard, 2006). The authors compared their data with field observations of activity concentration and surface deposition, and showed that ^{137}Cs and ^{131}I emissions are likely realistic, ^{132}I are probably underestimated, and noble gases are overestimated. Winiarek et al. (2014) also used inverse modeling to reconstruct FDNPP releases. Their technique can handle heterogeneous types of data in the same inversion. They estimated that the released ^{137}Cs ranged between 11.6 and 19.3 PBq, with an estimated standard deviation of 15–20%..

Finally, Chai et al. (2015) used an inverse emission estimation system based on a transfer coefficient matrix (TCM) created using the HYSPLIT Lagrangian dispersion model and a cost function that measures the differences between the model predictions and the actual air concentration measurements. The system was first tested with identical twin experiments, in which pseudo observations are generated with the same model used to estimate the transfer coefficients. With the pseudo observations generated at the same location and time as the actual ^{137}Cs observations to be assimilated later, the system is able to accurately recover the release rates and obtain better release estimates than the use of the singular value decomposition (SVD) method. This inverse estimation approach is found to be robust and not overly sensitive to the first guess of the release. As for Fukushima, a penalty term is added in the cost function to create smooth temporal changes. While the temporal variations of the release rates at 6-h segments cannot be fully retrieved using 24 h observations, the features at larger time scales are well recovered. Given that only one first guess estimation was taken into account (Katata et al., 2015b), the results were close to those of Katata et al. (2014).

Fission products were also released directly into the cooling water of the FDNPP and were transported to the sea along the east coast of Japan. Accurate estimation of the fission products directly discharged into the sea and their respective amounts is the basis for any impact assessment on marine life. Therefore, since 2011, a number of groups have investigated the amounts of Fukushima-derived fission products that were directly discharged into the ocean (Buesseler, 2012; Buesseler et al., 2012; Charette et al., 2013; Kumamoto et al., 2015; Tsumune et al., 2012). The total amounts of ^{137}Cs and ^{90}Sr released into the Pacific Ocean along the East Coast of Japan were estimated to be 1 to ~42 PBq (1 PBq = 10^{15} Bq) (Bailly du Bois et al., 2012; Kawamura et al., 2011; Tsumune et al., 2012) and 0.1–2.2 PBq (Povinec et al., 2012a), respectively. Clearly, these estimates are associated with considerable uncertainty, which stems from the lack of available information from the FDNPP accident (Chino et al., 2011; Ohno and Muramatsu, 2014; Oza et al., 2013; Ten Hoeve and Jacobson, 2012). Using a regional oceanic modeling system, ROMS, developed by Shchepetkin and McWilliams (2005), Tsumune et al. (2012) estimated that the total ^{137}Cs directly released was 3.5 ± 0.7 PBq from 26 March to the end of May, 2011. They noted that ^{137}Cs moved southwards along the coast during the simulation period; then the eastward-flowing Kuroshio current and its extension transported ^{137}Cs during May 2011, reducing ^{137}Cs concentrations to less than 10 Bq L^{-1} by the end of May 2011. Kawamura et al. (2011); Bailly du Bois et al. (2012) and Tsumune et al. (2012) showed that the discharged ^{137}Cs ranged between 1 and 42 PBq. Kawamura et al. (2011) used two models driven by data obtained using the WSPEEDI-II model. A limitation of this study was that it only considered deposited radionuclides from the aerosol, whereas releases via direct discharge of the cooling waters were not taken into account. Thus, it is likely that they underestimated the source term. The authors estimated the total amounts discharged into the ocean to be ~11 PBq for ^{131}I and ~4 PBq for ^{137}Cs for the period from March 21 to April 30, 2011.

Table 1 shows the total amounts of major radionuclides released from the FDNPP accident; these amounts are compared to global fallout from Nuclear Weapons Testing and the Chernobyl accident. Although uncertainty associated with total releases from the

FDNPP accident reaches 50% in certain cases, these data indicate that the Chernobyl accident resulted in the largest (estimated) release of radionuclides, whereas the Fukushima accident resulted in the largest direct release to the ocean. It is important to note that FDNPP accident is argued to have resulted in largest release of radionulclides into the ocean because the amount of radionuclides released by the Dnjepr River into the Black Sea has not been estimated. Kumamoto et al. (2015) reported that the mean value for water column inventories of the Fukushima-derived ^{134}Cs and the atomic-bomb-testing-derived ^{137}Cs were nearly similar (1020 ± 80 and 820 ± 120 Bq m^{-2}, respectively) suggesting that the FDNPP accident and nuclear bomb testing resulted in similar deposition amounts to the Pacific Ocean.

A significant fraction of the iodine released from the FDNPP accident remains as a gas (gaseous/total ratio: 77.2 ± 13.6% 23); hence, it is difficult to accurately measure total iodine. For this reason, the International Atomic Energy Agency (IAEA) has adopted the use of the "iodine equivalent" calculated from measured activities of ^{134}Cs and ^{137}Cs. Details of the methodology are presented in IAEA's International Nuclear Event Scale (INES, 2009). By using the IAEA/INES method, the Nuclear and Industrial Safety Agency (NISA) of Japan estimated the release of "iodine-131 equivalent" due to the FDNPP accident to be 770 PBq (NERHJ, 2011), which is considerably higher than the 150 to 160 PBq reported in Table 1. There are still ongoing efforts to computationally ascertain the source term of FDNPP nuclear accident with a great degree of accuracy. However, current estimates suggest total release amounts of 12–36.7 PBq of ^{137}Cs and 150–160 PBq of ^{131}I. These radioactive fission products have been transported to and deposited at locations around the globe. The two fundamental modes of transportation were the atmosphere and the sea.

4. Atmospheric transport and deposition of Fukushima derived radionuclides

Once the radioactive fallout was injected into the atmosphere, its trajectory was dispersed globally by the prevailing winds (Bolsunovsky and Dementyev, 2011; Huh et al., 2012; Leon et al., 2011; Manolopoulou et al., 2011; Pittauerová et al., 2011). Aerosol species were removed from the atmosphere by wet and dry processes, whereas noble gases were only lost due to radioactive decay. Evangeliou et al. (2013b) used the atmospheric transport model LMDZORINCA to simulate the global dispersion of radiocesium

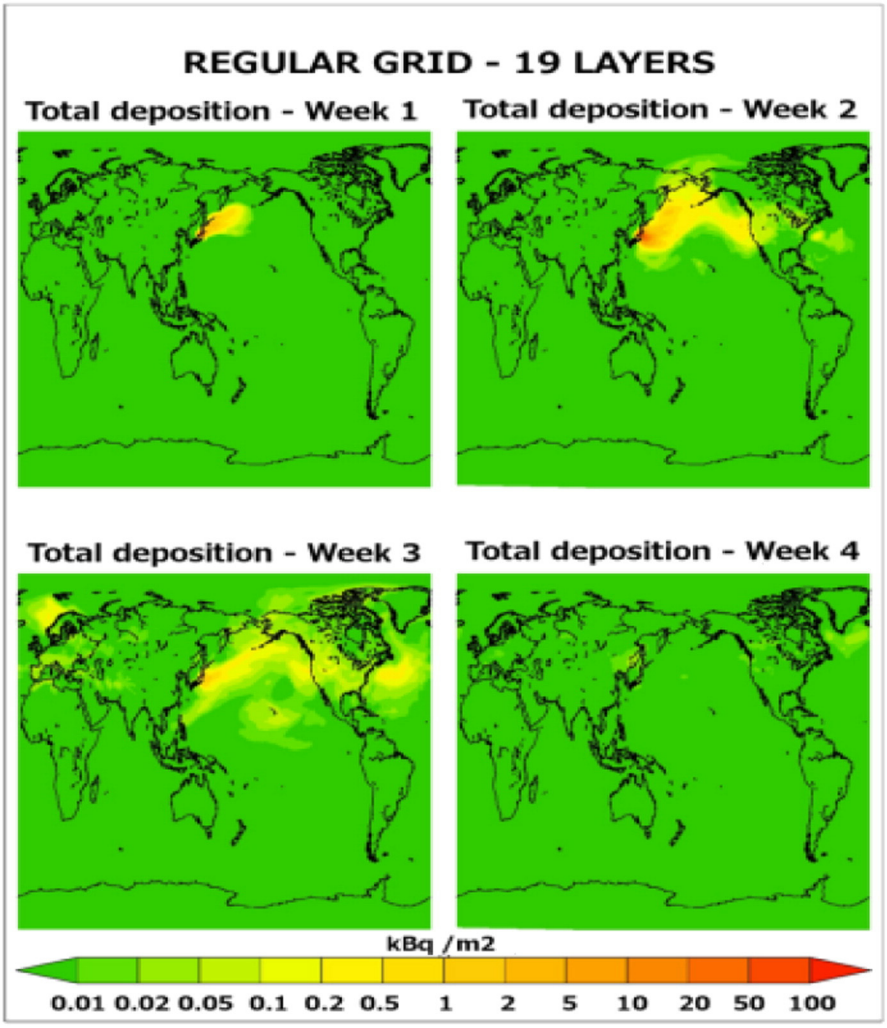

Fig. 1. Total deposition of ^{137}Cs (kBq m^{-2}) during the first 4 months after the FDNPP accident. The deposition is expressed in weekly intervals. The LMDZORINCA model was used in the resolution of 2.50° × 1.27° over 19 sigma-p vertical layers.
Source: Evangeliou et al. (2013b)].

Table 2
Fukushima-derived radionuclides detected in locations all over the world.

Location (reference)	Period (DD/MM/YY)	Maximum recorded activity concentrations in samples
National Institute of Radiological Sciences (NIRS) Chiba, Japan (Ishikawa et al., 2014)	15/03/2011 to 1 year later	Air (Bq m^{-3}): ^{132}Te: 9.9, ^{131}I: 16.2, ^{134}Cs: 10.13, ^{137}Cs: 10.27, ^{129m}Te: 4.0, ^{136}Cs: 123
Regional scale field work at Ibaraki, Fukushima, Chiba, Tochigi, Saitama, and Tokyo Japan (Kinoshita et al., 2011)	03/2011 to 05/2011	Surface (Bq m^{-2}): ^{137}Cs: 9×10^5
Gunsan and Busan, South Korea (Kim et al., 2012)	28/03/2011 to 31/05/2011	Surface (Bq m^{-2}): ^{137}Cs + ^{134}Cs: ~3 Air (Bq m^{-3}): ^{131}I: 3.12×10^{-3}, ^{134}Cs: 1.19×10^{-3}, ^{137}Cs: $.25 \times 10^{-3}$
Uljin and Ansan, South Korea (Hong et al., 2012)	07/4/2011 to 28/04/2011	Surface (Bq m^{-2}): ^{137}Cs: 649, ^{134}Cs: 802
Waste Isolation Pilot Plant, Carlsbad, New Mexico, USA (Thakur et al., 2012)	17/03/2011 to 04/04/2011	Air (Bq m^{-3}): ^{134}Cs: 0.38×10^{-3}, ^{137}Cs: 0.45×10^{-3}, ^{131}I: 3.85×10^{-3}
Krasnoyarsk (Russia) (Bolsunovsky and Dementyev, 2011)	04/04/2011 to 03/05/2011	Melted snow (Bq L^{-1}): ^{131}I: 0.62, ^{137}Cs: 0.075, ^{134}Cs: 0.095
Southwest USA (Wetherbee et al., 2012)	08/03/2011 to 05/04/2011	Surface (Bq m^{-2}): ^{137}Cs: 30–240, ^{134}Cs: 2–46
Alaska, USA (Wetherbee et al., 2012)	08/03/2011 to 05/04/2011	Surface (Bq m^{-2}): ^{137}Cs: 16–27, ^{134}Cs: ~55
USA (Wetherbee et al., 2012)	08/03/2011 to 05/04/2011	Surface (Bq m^{-2}): ^{137}Cs: 1–45, ^{134}Cs: 1–3
La Spezia, Italy (Barsanti et al., 2012)	28/03/2011 to 12/04/2011	Surface (Bq m^{-2}): ^{137}Cs + ^{134}Cs: ~0.3
Athens, Greece (Kritidis et al., 2012)	2011	Surface (Bq m^{-2}): ^{137}Cs: ~10
Thessaloniki, Greece (Bolsunovsky and Dementyev, 2011; Manolopoulou et al., 2011)	24/03/2011 to 09/04/2011	Air (Bq m^{-3}): ^{131}I: 497×10^{-6}, ^{137}Cs: 145×10^{-6}, ^{134}Cs: 126×10^{-6} Rain water (Bq L^{-1}): ^{131}I: 0.7
Nuclear Science Research Institute, Ibaraki, Japan (Saegusa et al., 2013)	01/03/2011 to 01/04/2011	Evaporated water (Bq m^{-2}): ^{137}Cs: 1.4×10^4 Rain water (Bq L^{-1}): ^{137}Cs: 41
Hei Longjiang, Ji Lin, Qin Huangdao, Tian Jin and Beijing area, China (Tuo et al., 2013)	02/04/2011 to 13/04/2011	Air (Bq m^{-3}): ^{131}I: 1720×10^{-6}, ^{134}Cs: 247×10^{-6}, ^{137}Cs: 289×10^{-6}, ^{136}Cs: 23×10^{-6} (Tuo et al., 2013) Rain water and vegetation (Bq kg^{-1}): ^{131}I: 2.68
University of Milano-Bicocca, Milan, Italy (Clemenza et al., 2012)	12/03/2011 to 05/04/2011	Air (Bq m^{-3}): ^{131}I: ~4.2×10^{-4}, ^{137}Cs: ~9.4×10^{-5}, ^{134}Cs: 8×10^{-5}
Ten air sampling sites, Czech Republic (Rulík et al., 2014)	22/03/2011 to 17/05/2011	Air (Bq m^{-3}): ^{131}I: 5.6×10^{-3}, ^{137}Cs: 0.72×10^{-3}, ^{134}Cs: 0.64×10^{-3}
Vilnius, Lithuania (Lujanienė et al., 2013; Lujanienė et al., 2012a)	23/03/2011 to 15/04/2011	Air (Bq m^{-3}): ^{131}I: 3700×10^{-6}, ^{137}Cs: 1040×10^{-6}
Pic du Midi observatory, France (Evrard et al., 2012)	22/03/2011 to 10/04/2011	Surface (Bq m^{-2}): ^{134}Cs: ~2 Air (Bq m^{-3}): ^{134}Cs: 0.2×10^{-3}, ^{137}Cs: 0.7×10^{-3}
Centre d'Etudes Nucléaires de Bordeaux Gradignan, Université de Bordeaux, France (Perrot et al., 2012)	26/03/2011 to 14/05/2011	Air (Bq m^{-3}): ^{131}I: 2.4×10^{-3}, Rain water (Bq L^{-1}): 3.5
Comenius University Mlynská Dolina, Bratislava, Slovakia (Povinec et al., 2012b)	19/03/2011 to 25/05/2011	Surface (Bq m^{-2}): ^{137}Cs: 2–114, ^{134}Cs: <100 Air (Bq m^{-3}): ^{131}I: 0.5×10^{-3}, ^{137}Cs: 0.07×10^{-3}, ^{134}Cs: 0.0197×10^{-3} Rain water (mBq L^{-1}): ^{131}I: 1130, ^{137}Cs: 12, ^{134}Cs: 90
Spanish environmental radiological monitoring network in Barcelona, Caceres and Seville, Spain (Baeza et al., 2012)	16/03/2011 to 17/04/2011	Air (Bq m^{-3}): ^{131}I: 3080×10^{-6}, ^{137}Cs: 690×10^{-6}, ^{134}Cs: 620×10^{-6}, ^{132}Te: 330×10^{-3}
Stations in Novi Sad, Rimski Sancevi, Kisac, Melenci, Krcedin, Subotica, Kumane, Zrenjanin, Bikovo, Nadalj, Serbia (Bikit et al., 2012)	16/03/2011 to 20/04/2011	Air (Bq m^{-3}): ^{131}I: 2.7×10^{-3}, ^{137}Cs: 0.21×10^{-3} Rain water (Bq L^{-1}): 1.8
Hanoi, Dalat, and Ho Chi Minh City, Vietnam (Truong et al., 2012)	27/03/2011 to 22/04/2011	Air (Bq m^{-3}): ^{131}I: 193×10^{-6}, ^{134}Cs: 33×10^{-6}, ^{137}Cs: 37×10^{-6}
Lancaster; Spark Bridge; Stirling, etc. (United Kingdom) (Beresford et al., 2012)	01/04/2011 to 13/04/2011	Grass (Bq kg^{-1}): dry matter for ^{131}I: 55, dry matter for ^{137}Cs: 8
Cluj, Bihor, Bistriţa-Nŭsaŭd, Maramureş, and Arad, NW Romania (Cosma et al., 2012)	28/03/2011 to 11/04/2011	Rain water (Bq L^{-1}): ^{131}I: 1.69, ^{134}Cs: 0.042, ^{137}Cs: 0.067
Mt. Zeppelin global atmosphere watch station, NyÅlesund, Svalbard, Norway (Paatero et al., 2012)	12/03/2011 to 02/05/2011	Air (Bq m^{-3}): ^{131}I: 810×10^{-6}, ^{134}Cs: 659×10^{-6}, ^{137}Cs: 675×10^{-6}
Sacavém, Lisbon, Portugal (Carvalho et al., 2012)	17/03/2011 to 20/04/2011	Surface (Bq m^{-2}): ^{137}Cs + ^{134}Cs: ~1 Air (Bq m^{-3}): ^{131}I: 1.47×10^{-3}, ^{137}Cs: 134×10^{-3} Soil (Bq m^{-2}): ^{131}I: 1.03, ^{134}Cs: 0.65, ^{137}Cs: 0.74
Vancouver Island, Canada (Sinclair et al., 2011)	20/03/2011 to 28/04/2011	Air (Bq m^{-3}): ^{133}Xe: 30–70
Countrywide, Finland (Leppänen et al., 2013)	19/03/2011 to 28/04/2011	Air (mBq m^{-3}): ^{131}I: 10.6, ^{134}Cs: 0.39, ^{137}Cs: 0.41, ^{136}Cs: 28, ^{129}Te: 129, ^{129m}Te: 234, ^{132}Te: 51 Higher concentrations were observed in Southern Finland than in the North
Huelva Iberian Peninsula, Spain (Lozano et al., 2011)	28/03/2011 to 07/04/2011	Air (mBq m^{-3}): ^{131}I: 3.69, ^{132}I: 0.162, ^{134}Cs: 0.88, ^{137}Cs: 0.95
Çnaem, Istanbul, Turkey (Güngör et al., 2014)	07/03/2011 to 12/09/2011	Air (mBq m^{-3}): ^{131}I: 1.03, ^{134}Cs: 0.25, ^{137}Cs: 0.23 Rain water (mBq m^{-3}): ^{134}Cs: 4.1, ^{137}Cs: 4.7
Mount Lulin, Taiwan (Huh et al., 2012)	20/03/2011 to 29/04/2011	Air (mBq m^{-3}): ^{131}I: 0.06, ^{134}Cs: 0.028, ^{137}Cs: 0.030
Vienna Austria, Austria (Steinhauser et al., 2013a)	24/03/2011 to 13/04/2011	Air (Bq m^{-3}): ^{131}I: 1.2×10^{-3} Rain water (Bq L^{-1}): ^{131}I: 5.2
Budapest, Hungary (Homoki et al., 2013)	08/03/2011 to 22/05/2011	Air (μBq m^{-3}): ^{131}I: 96, ^{137}Cs: 10^2, ^{134}Cs: 10^2
Monaco (Pham et al., 2012)	23/03/2011 to 07/05/2011	Surface (Bq m^{-2}): ^{137}Cs + ^{134}Cs: 1–2 Air (μBq m^{-3}): ^{131}I: 354, ^{134}Cs: 30, ^{137}Cs: 37 Deposition rate (mBq m^{-2} d^{-1}): ^{134}Cs: 13.7, ^{137}Cs: 19.1

(continued on next page)

Table 2 (*continued*)

Location (reference)	Period (DD/MM/YY)	Maximum recorded activity concentrations in samples
Bremen, Seefeld and Schifford, Germany (Pittauerová et al., 2011)	12/03/2011 to 14/05/2011	River sediment (Bq kg^{-1}): ^{131}I: 0.113, ^{137}Cs: 2.67, ^{134}Cs: 0.08 Soil (Bq kg^{-1}): ^{131}I: 0.68, ^{137}Cs: 3.00, ^{134}Cs: 0.069 Rain water (Bq kg^{-1}): ^{131}I: 0.31, ^{137}Cs: 0.02, ^{134}Cs: 0.02
Darwin, Townsville, Melbourne, Perth and Cacos Island, Australia (Carpenter and Tinker, 2012)	01/40/2011 to 30/04/2011	Air (Bq m^{-3}): ^{133}Xe: 12×10^{-3} No Fukushima-derived radio iodine and cesium was detected in Australia, ^{133}Xe was only detected in Darwin
Institute of Nuclear Physics Krakow, Poland (Mierelski et al., 2014)	21/03/2011 to 30/10/2011	Air (mBq m^{-3}): ^{131}I: 5.73, ^{134}Cs: 0.461, ^{137}Cs: 0.436

during the first four months after the accident. A weekly intervals of the total deposition of ^{137}Cs is presented in Fig. 1.

Global scale simulations by Christoudias and Lelieveld (2013) and Evangeliou et al. (2014b) showed that the plume generated by the FDNPP was initially affected by northerly and easterly winds, which gradually reached the USA via the North Pacific and Alaska.

Table 2 summarizes observations of Fukushima-derived fission products all over the world, including air concentrations (in Bq m^{-3}), surface deposition densities (in kBq m^{-2}) and concentration in both fresh and marine water (in Bq L^{-1}). The data in Table 2 can be used for worldwide health assessments based on field observations.

Based on observations from the CTBTO network (Christoudias and Lelieveld, 2013), fallout from FDNPP reached Eastern Siberia after four days, Alaska and the West Coast of the USA after 8–10 days (Thakur et al., 2012), the east coast after 11–12 days, the North Atlantic (i.e., Iceland) after about 13 days, Europe after 14–17 days, and Asia after 18–20 days. This is much longer than the ~10-day period it took for the Chernobyl fallout to spread across the globe (Harrison et al., 1993). This is mainly because in Chernobyl there was a tremendous explosion and nuclear fire, which injected radionuclides directly into the troposphere (Evangeliou et al., 2013a) resulting in faster transport, whereas in Fukushima the releases occurred inside the planetary boundary layer (PBL). The radioactive plume from Fukushima was transported over the Pacific Ocean; over the ocean, variation in the boundary layer is low and this may have resulted in the prolonged time for travel of the Fukushima radioactive cloud around the world. The variability in surface temperatures over the ocean are small compared to those over land, and this results in mixing of larger air masses and hence longer transport times for aerosols that are transported over ocean (Aliyu, 2014; Jilani, 2009; Sorbjan, 1989). In other words, the turbulence over the ocean is relatively low compared to terrestrial ground surfaces where the presence of trees, mountains and other rough surfaces increases the exchange of mass and momentum between the ground surface and the atmosphere (Jilani, 2009; Marshall and Plumb, 1965).

Observations in Table 2 are very close to those of the CTBTO and agree with the estimation that the plume arrived in Europe more than 10 days after the FDNPP accident. Outside Japan, fission products were detected in Korea, Vietnam, Taiwan, USA, Canada, and Europe (CTBTO, 2011; Huh et al., 2012; Kim et al., 2012; Leon et al., 2011; Masson et al., 2011) (Table 2). In Europe, the first traces of the radioactive fallout were detected on 19 March 2011 (slightly more than one week after the accident), while the first peak of activity level was observed between 28 and 30 March. Measurements suggest that airborne activity levels are currently of minimal concern for public health in Europe (Masson et al., 2011). The activity concentrations of radionuclides in the plume decreased exponentially with downwind distance (Hsu et al., 2012) due to dry and wet scavenging and subsequent deposition along the path of dispersion (Aliyu et al., 2015b).

In Japan, air concentrations of ^{134}Cs and ^{137}Cs in the city of Fukushima were still elevated (613 μBq m^{-3} and 829 μBq m^{-3}, respectively) 7 months after the accident (Toyoshima et al., 2011). Intensive monitoring was conducted on smaller Japanese Islands, which included 45 measuring stations (Hirose, 2012). These measurements suggested that the FDNPP accident affected areas in the central and eastern parts of Honshu Island, although fission products were deposited all over Japan. Hirose (2012) also used daily deposition of radionuclides in rain water measured by Prefectural Governments in Japan to estimate the atmospheric resident time of Fukushima-derived ^{137}Cs within 300 km from the FDNPP site: ~10 days.

Earlier studies (e.g. Buck, 2011; Ten Hoeve and Jacobson, 2012; Thakur et al., 2012) showed that radionuclides were transported by winds over the Pacific Ocean towards the West Coast of the USA. Fig. 2 depicts the atmospheric dispersion pattern of the fission products at different heights of the PBL during the first 10 days of the FDNPP, using single-particle trajectories employed by the HYSPLIT model (Draxler, 2004; Draxler and Hess, 1997). The start time of release of particles from the FDNPP was 12 March 2011. The lower trajectories [100 and 500 m above ground level (AGL)] circulated along the East Coast of Japan towards the northern part of the country. The particles at higher altitudes (1 and 1.5 km AGL) were transported across the Pacific Ocean to the West Coast of North America. Thakur et al. (2012) showed that 19% of the total fallout was deposited on Japan and only 2% reached other land areas of Asia and North America, while the remainder was deposited over the Pacific Ocean and the Arctic. In a recent study, Evangeliou et al. (2015) estimated that about 23% of the released ^{137}Cs was deposited over Japan and 76% of the radiocesium was deposited over the North Pacific and North Atlantic oceans. From the remainder, 163 TBq was deposited in North America, mostly in the USA (95 TBq), Canada (40 TBq) and Greenland (5 TBq), about 14 TBq in Europe (mostly in European Russia, Sweden and Norway), 47 TBq in Asia (mostly in Asian Russia, Philippines and South Korea), and traces deposited over Africa, Oceania and Antarctica. Considering that the plume followed a northward direction before it arrived in the USA and then in Europe, about 69 TBq was deposited over the Arctic (see Fig. 2).

Deposition of the Fukushima-derived radionuclides over land are directly linked to their atmospheric release. Surface deposition levels are associated with the risk of internal radiation exposure via inhalation, whereas aerosol species are important when considering ingestion pathways. Soil around the FDNPP and neighboring prefectures was contaminated with ^{137}Cs and ^{134}Cs at levels of more than 10^5 and 10^4 Bq m^{-2}, respectively (Burns et al., 2012; Yasunari et al., 2011), and other rare refractory elements might also have been important. Kinoshita et al. (2011) reported that the surface deposition of ^{137}Cs in central and eastern Japan ranged from 0.4 to 900 kBq m^{-2}, which is 0.06–130 times the cumulative deposition (100–1000 Bq m^{-2}) from atmospheric nuclear bomb testing (Kinoshita et al., 2011). It is estimated that a total of more than 5.6 PBq of ^{137}Cs was deposited over Japan (Yasunari et al., 2011). For example, soil concentrations of ^{134}Cs and ^{137}Cs in the Iitate Village area located 25 km northwest of the FDNPP were 62.4 and ~73 kBq kg^{-1}, respectively, and the topsoil concentrations of ^{131}I and ^{137}Cs 23 km south of the Fukushima Reactor Unit 1 were reported to be 140 kBq m^{-2} and 80 kBq m^{-2}, respectively (Garnier-Laplace et al., 2011).

Radiation dose measurements can be used to extrapolate the level of surface contamination due to the FDNPP accident in Japan. For instance,

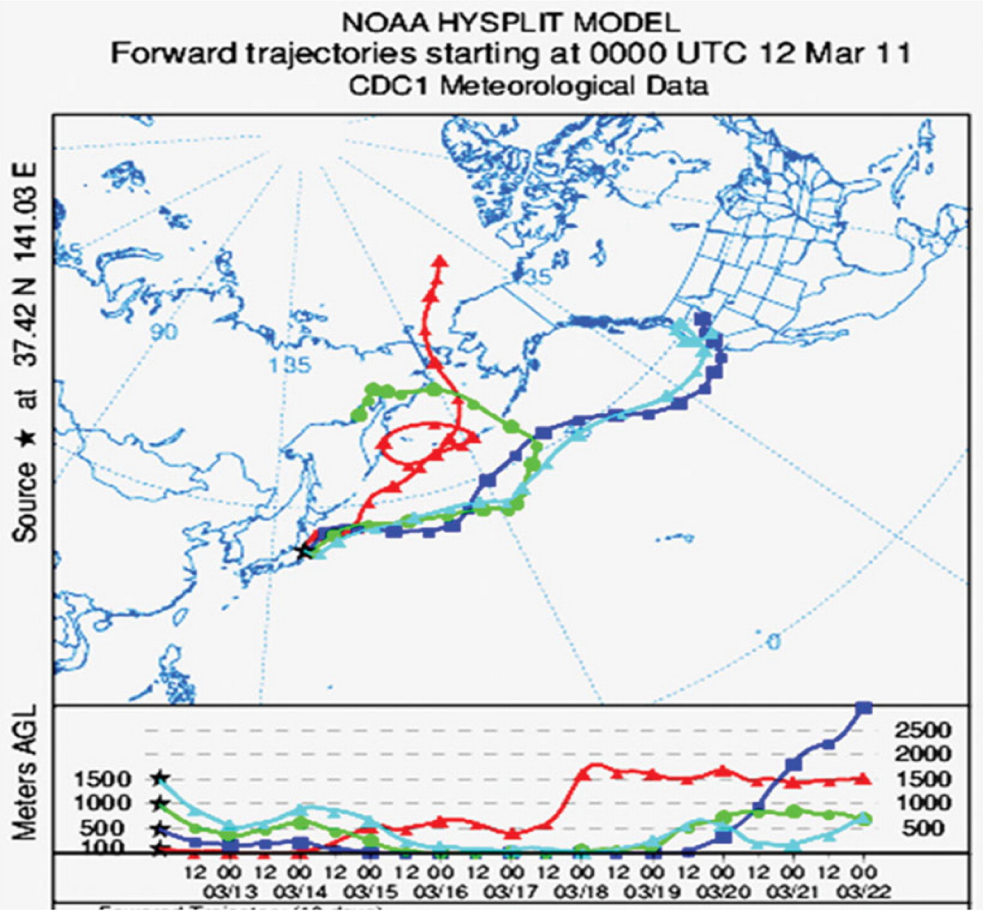

Fig. 2. Calculated trajectories for air masses released at the site of the Fukushima Daiichi reactor (Aliyu et al., 2015a).

the environmental external radiation dose rate before the nuclear accident in the Fukushima region was approximately 0.05 μSv h^{-1} (Kinoshita et al., 2011). The external radiation dose rates in prefectural areas due to the FDNPP accident have been estimated to be 1.14 μSv h^{-1} in Naka-Dori, 4.57 μSv h^{-1} in Iitate, 0.02 μSv h^{-1} in a region between northern Ibaraki and eastern Saitama, and 0.23 μSv h^{-1} in southern Ibaraki and northern Chiba prefectures (Kinoshita et al., 2011). Møller et al. (in pressb) reported that dose rates at 400 locations around the Fukushima region varied from 0.5 to 38 μSv h^{-1}. This indicates that large areas in eastern and northern Japan were strongly contaminated by ^{137}Cs, whereas western regions were largely shielded by mountainous topography. However, similar to Chernobyl, deposition was highly heterogeneous even within the most contaminated areas (Møller et al., in pressb, in pressc).

The data presented in Table 2 can also be used to predict the dose and risk in Europe, North America and Asia using empirical environmental data rather than estimates based on mathematical models that may result in over- or underestimation of the impacts of the accident. The question of how fission products from FDNPP accident were transported and deposited in locations within and outside Japan will continue to be of interest as it is the basis for any radioecological impacts assessment involving Fukushima-derived long-lived radionuclides which could, even at trace quantities, bioaccumulate in plant and animal tissues, thereby increasing the risk of radiation induced effects in the exposed populations.

5. Impacts of the FDNPP accident on humans

Given that the potential health effects of the FDNPP accident will likely be of great social and economic importance for decades to come, modeling of predicted doses can be a useful tool for a preliminary assessment of the health risks. Table 3 illustrates the predicted health effects of the FDNPP accident on local and global populations. In all cases, a Linear Non Threshold (LNT) model of human exposure was used to calculate potential radiological health effects. This model assumes that the disintegration of each radionuclide has the same probability of causing cell transformation, and that each transformed cell has the same probability of developing a cancer tumor. Although the LNT model has been employed extensively in radiation safety (NRC, 2006; UNSCEAR, 2010), several arguments concerning its validity and response at low doses still remain unresolved (Cuttler, 2010; Tubiana et al., 2009). Traditional epidemiological studies have only considered doses above 100 mSv as causing statistically significant increases in stochastic cancer risk, although there are a growing number of studies suggesting possible effects at doses well below 100 mSv (e.g. Brenner and Sachs, 2006; Brenner et al., 2003; Eidemüller et al., 2010; Mathews et al. 2013). On the other hand, proponents of the LNT model claim that the difficulty in detecting and attributing a small number of cancers to low doses does not indicate that there is an absence of risk (Hoffman et al., 2011). Note that the NRC (Nuclear Regulatory Commission) and the WHO (World Health Organization) accept the LNT hypothesis for such assessments.

Ten Hoeve and Jacobson (2012) estimated that there would be an additional 130 (15–1100) and 180 (24–1800) lifetime cancer related mortalities and morbidities related to the Fukushima radiological disaster, respectively, with over 90% of the projected impact occurring in Japan. However, Beyea et al. (2013) argued that the failure by Ten Hoeve and Jacobson (2012) to consider the long-term dose estimate from radiocesium in their studies may lead to an underestimation of the global effects of the accident. By including a more realistic

Table 3
Cancer related morbidities (cases) and mortalities (deaths) due to the FDNPP accident. The table shows number of individuals that are expected to suffer from all solid cancers.

Reference	Morbidities (incidents)	Mortalities (deaths)	Collective dose in Japan man-Sv
IRSN (2012)	70–250	–	–
UNSCEAR (2013)	–	5000	48,000
IPPNW (2013)	–	10,000	95,000
Von Hippel (2011)	–	750–1500	–
Ten Hoeve and Jacobson (2012)	24–1800	15–1100	–
Evangeliou et al. (2014b)	160–880	730–1260	–

population perspective, Beyea et al. (2013) suggested that the estimate for the number of future cancer mortalities due to the FDNPP accident is probably closer to 1000 than 125. Other health estimates for Fukushima-related cancers were made by Von Hippel (2011) who estimated the cancer mortalities to be 770–890 and the highest external doses outside the 20-km evacuation area to be between 20 and 40 mSv.

Evangeliou et al. (2014a) estimated that the highest doses would rise to between 50 and 100 mSv. Using the LNT model, in the northernmost area of the FDNPP (with about 300,000 inhabitants) they estimated the lifetime mortality risks to be 750–1500 individuals. The IRSN (2012) estimated that a population of 21,100 received more than 16 mSv; a population of 3100 received more than 50 mSv, and another group of 2200 individuals received 100–500 mSv near Fukushima. Assuming the LNT hypothesis, an exposure of 100 mSv will increase lifetime cancer risk by 1% (Cuttler, 2010), giving rise to 70–250 excess cancers depending on the precise amount of radiation received by each grouping.

On the other hand, several studies have reported much higher values for projected cancer mortalities in Japan. For example, the most detailed model, used in a report published by IPPNW (2013) in late March 2013, estimated a collective effective dose of 95,000 mSv, whereas UNSCEAR (2013) estimated 48,000 mSv. In terms of fatal cancers (mortalities), the UNSCEAR estimates imply (via the LNT theory) that ~5000 people in Japan will die in the future from Fukushima-related cancers. This figure is obtained by applying a fatal cancer risk of 10% per Sv; the UNSCEAR report, like the previous WHO reports, no longer applies a DREFF (dose and dose-rate effectiveness factor) of 2 to risk estimates. However, the methodologies and assumptions used in the new UNSCEAR report will require scrutiny before a final conclusion can be drawn. It is noteworthy that the UNSCEAR report adds that "The collective effective dose to the population of Japan due to a lifetime exposure following the FDNPS accident is approximately 10–15% of the corresponding value for European populations exposed to radiation following the Chernobyl accident. Correspondingly, the collective absorbed dose to the thyroid was approximately 5% of that due to the Chernobyl accident."

In a recent study, Evangeliou et al. (2014b) considered numerous fission products (^{131}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{129m}Te, ^{132}Te, ^{95}Nb, ^{90}Sr, ^{110m}Ag, ^{99}Mo, ^{241}Am, ^{238}Pu, $^{239-240}$Pu, ^{241}Pu, ^{242}Cm and $^{243-244}$Cm) in their assessment. The authors also took into account several arguments reported after the Ten Hoeve and Jacobson (2012) estimates were issued, pointing out shielding effects and the exponential decrease of the deposition densities of the studied radionuclides as a result of vertical migration in soil. For a discussion of vertical migration of deposited radionuclides from fallouts see, for example, Almgren and Isaksson (2006); Nakanishi et al. (2014), and Takahashi et al. (2015). Evangeliou et al. (2014b) estimated that solid cancer incidences and mortalities from Fukushima would be 160 to 880 and 110 to 640, respectively, which are close to previous estimations. By adding thyroid cancers, the total estimate rises from 230 to 850 for incidents and from 120 to 650 for mortalities (see Figs. 3 and 4).

Estimated fatalities due to workers' exposure and mandatory evacuation have been reported to be around 610, increasing total estimated mortalities to 730–1260 (Evangeliou et al., 2014b). These estimates are 2.8 times higher than those previously reported for radiocesium and ^{131}I, and 16% higher than those reported based on radiocesium only.

Prefectural Governments in Japan have adopted the use of highly sensitive ultrasound systems to scan for radiation-induced thyroid cancer due to the Fukushima accident (Tronko et al., 2014). Tronko et al. (2014) reported that, by the first quarter of 2014, about 80% of the target population (36,000 of the residents aged up to 18 years) were examined. Out of the examined population, 75 cases of suspected malignancy were identified. Thirty-four patients have received surgery; pathological diagnoses include one benign tumor, one suspected to be poorly differentiated thyroid carcinoma, and 32 papillary thyroid carcinomas. This is arguably an alarming case of prevalence considering the examination period of less than 3 years following exposure. However, the study was inadequately controlled and failed to effectively assess thyroid cancers in preschool children in the broader unexposed population, a point that was used by Tronko et al. (2014) to argue that the high prevalence was not due to the FDNPP nuclear accident. Arguably, the highly sensitive equipment that is in use in Fukushima might have resulted in this high prevalence of thyroid anomalies that would not normally have been detected. However, Tronko et al. emphasized that additional analysis will need to address thyroid cancer cases that may appear in the coming years, once the period of latency has passed. They noted that attention should be paid to thyroid dose reconstruction, age at exposure and diagnosis, and whether there will be a "harvesting effect," which is a spike in cases after introduction of intensive screening programs.

Tokonami et al. (2012) and Hosoda et al. (2013) attempted to measure and estimate the thyroid doses in evacuees from the Fukushima nuclear accident site, respectively. Tokonami et al. (2012) conducted screening tests of 62 residents and evacuees using a portable Na(TI) spectrometer to measure ^{131}I activity in the thyroid. These measurements were conducted a month after the FDNPP accident (12 to 16 April 2011). The age of examinees ranged from <1 to 80 years; 17 of them were from Tsushima District of Namie Town, a region which is severely contaminated with radioactive materials released from the crippled FDNPP. Forty-five of the examinees were evacuees from coastal areas, including Minami-soma City located to the north of FDNPP. The median (and maximum) thyroid equivalent doses were estimated as 4.2 (23) mSv and 3.5 (33) mSv for children and adults, respectively, which are much smaller than the mean thyroid dose in the Chernobyl accident (490 mSv in evacuees). Hosoda et al. (2013) used the measurement data for ^{131}I from the earlier study by Tokonami et al. (2012) to estimate the external exposure of thyroid to ^{131}I based on the accumulated ^{134}Cs among evacuees. The maximum values of ^{131}I/^{134}Cs activity ratio corresponding to thyroid uptake factors of 0.3, 0.1 and 0.03 were evaluated to be 0.9, 2.6 and 8.7, respectively. The maximum value of the ^{131}I/^{134}Cs activity ratio was used to obtain the most conservative thyroid equivalent dose estimation. The maximum internal exposure of the thyroid to ^{131}I on the basis of cumulative ^{134}Cs measured by the whole-body counter was estimated to be 18 mSv. This value is higher than the maximum thyroid doses for children and adults reported in the earlier study.

Computer models are currently used to predict the human deaths and health effects of the FDNPP nuclear accident based on the LNT hypothesis. More field research is needed in order to investigate the effects of the nuclear accident in exposed human populations. This project should involve continuous monitoring of the exposed population as radiation effects on humans may take longer before they are expressed especially for cancers which can have latency periods of decades. The study of non-human biota, which often have much shorter lifespans (compared to humans), provides opportunities to investigate the biological consequences of radiological contaminants on a short timespan and this approach has been fruitful for study of the consequences of both the Fukushima and Chernobyl disasters.

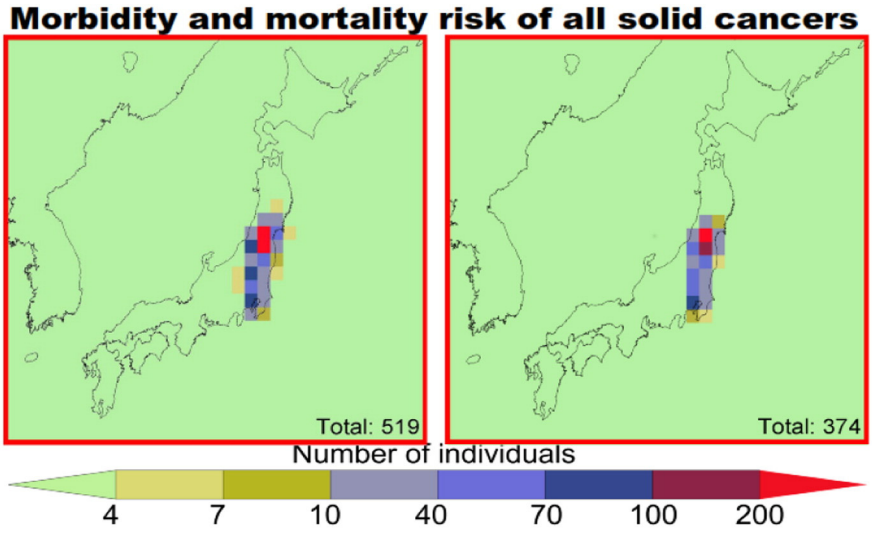

Fig. 3. Excess lifetime morbidity (incidence) and mortality (death) risk of all solid cancers from the uptake of ^{134}Cs, ^{136}Cs, ^{137}Cs ^{133}Xe, ^{129m}Te, ^{132}Te, ^{95}Nb, ^{90}Sr, ^{110m}Ag, ^{99}Mo, ^{241}Am, ^{238}Pu, $^{239–240}$Pu, ^{241}Pu, ^{242}Cm and $^{243–244}$Cm in Japan from all internal and external exposure pathways using the Linear Non Threshold models for a lifetime of 89 years. The calculations were performed using the LMDZORINCA model for a spatial resolution of 0.45°–0.51°.
Source: Evangeliou et al. (2014b).

6. Impacts of FDNPP accident on marine ecosystems

Contamination of the marine environment following the FDNPP accident represents the most important anthropogenic radioactive release into the sea ever recorded, on par with fallout from atomic bomb testing. The radioactive marine pollution came from atmospheric fallout onto the ocean and direct release of contaminated water from the plant.

In the immediate vicinity of the plant, Cs levels of the ocean surface at the point of discharge showed up to 10^3 times higher activities than had been reported by Aoyama and Hirose (2004) before the Fukushima accident (Aoyama and Hirose, 2004), with a peak of 68 Bq m^{-3} on 6 April 6, 2011 (Buesseler et al., 2011). Dilution of the contaminants occurs rapidly due to ocean mixing, particularly in the energetic coastal waters off Japan where the Oyashio waters move south and interact with the rapidly flowing and offshore meandering of the Kuroshio Current. These currents, tidal forces and eddies mix the waters rapidly offshore. As a consequence, one month after the accident the ^{137}Cs level decreased by a factor of 10^3.

Honda et al. (2012) reported that the ^{137}Cs activity in surface seawater ranged from 0.004 to 0.284 Bq kg^{-1} in the western North Pacific from 14 April to 5 May. Tsumune et al. (2012) argued that if no additional releases occur in the future, the effect of the ^{137}Cs may be smaller than that of global fallout in the North Pacific. The timescale of transport to other basins is several decades. Thus, the effect of the ^{137}Cs release on other oceanic basins might be negligible. With respect to dose effects on humans, Buesseler et al. (2011) calculated that the dose due to direct exposure during human indirect contact with the water was 0.1 μSv day^{-1} when the ^{137}Cs level approaches 10^2 kBq m^{-3}, which is lower than the average dose from all sources to the Japanese population of about 0.17 μSv day^{-1}.

Cesium-137 concentrations in suspended solids (SS) from surface and subsurface waters off the western North Pacific have been measured by Honda et al. (2012); it ranged from 5.92 to 32.42 Bq kg^{-1} on a dry weight basis (kg-dw^{-1}). A similar pattern was observed for ^{134}Cs. Thus, it is likely that SS were contaminated with radionuclides from the FDNPP (Honda et al., 2012). The SS-seawater distribution

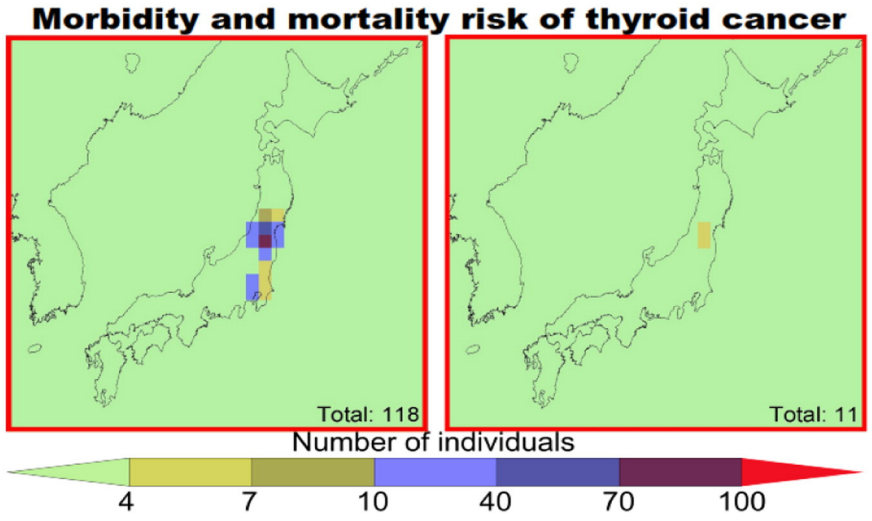

Fig. 4. Excess lifetime morbidity (incidence) and mortality (death) risk of thyroid cancer from the uptake of ^{131}I from all internal and external exposure pathways using the Linear Non Threshold model for a lifetime of 89 years. The calculations are performed using the LMDZORINCA model for a spatial resolution of 0.45°–0.51°.
Source: Evangeliou et al. (2014b).

coefficient (K_ds: Bq kg-dw^{-1} for SS/Bq kg^{-1} for seawater) was further estimated to be 200–4400, with an average of 2100. This average K_ds is comparable to the suggested K_ds for the open ocean: 2000 (IAEA, 2004).

In terms of potential biological impacts, radiation doses in marine organisms are generally dominated by the naturally occurring radionuclides ^{210}Po (an alpha emitter) and ^{40}K, even when organisms are exposed to anthropogenic radioactivity discharged to coastal waters (Aarkrog et al., 1996). Fukushima-derived radiocesium was detected in zooplankton and mesopelagic fish (Buesseler et al., 2012), with the ^{137}Cs concentration in zooplankton off the western North Pacific coast at 13.46–71.46 Bq kg-dw^{-1}, which is significantly higher than that of the southern Baltic Sea and the northern Adriatic Sea one month after the Chernobyl accident: 0.4–5 Bq kg-ww^{-1} (Knapinska-Skiba et al., 2003; Marzano and Triulzi, 1994). In contrast, the ^{137}Cs concentration in zooplankton around Japan was less than 0.1 Bq kg-ww^{-1} during the past decade (Kaeriyama et al., 2008; Kasamatsu and Ishikawa, 1997). Thus, the observed concentrations were two orders of magnitude higher than before the accident. Meanwhile, the concentration factor [CF: i.e. the ratio of the Cs concentration of zooplankton to that of ambient seawater (Bq kg-ww-1/Bq kg^{-1})] was calculated to be 200–840, which is an order of magnitude higher than the 10–100 of previous observations (IAEA, 2004; Kaeriyama et al., 2008; Kasamatsu and Ishikawa, 1997).

Buesseler et al. (2012) suggested that radiation risks of ^{137}Cs derived from the FDNPP to marine organisms and human consumers of seafood were well below those from natural radionuclides. At the same time, the total cesium levels in demersal (bottom-dwelling) fish off Fukushima was investigated (Buesseler, 2012), where the cesium levels in 40% of fish were above the new regulatory limit (100 Bq kg^{-1} wet) set by the Japan Ministry of Agriculture (JMA, 2011). However, this is lower than the new regulatory limit of cesium in fish products in four prefectures (Iwate, Miyagi, Ibaraki and Chiba).

The effect of the FDNNP accident on marine products in Fukushima was also assessed by Wada et al. (2013). The authors concluded that the total activity concentrations of radiocesium in marine products have decreased significantly. However, the authors noted that higher activity concentrations have been observed in shallower waters south of the FDNPP, and long-term monitoring of marine products from around the FDNPP is needed before restarting coastal fisheries (Wada et al., 2013). Chen (2013) reported the radioactivity level in fish products from other prefectures adjacent to Fukushima, based on monitoring data of July 2013. Activity concentrations of ^{134}Cs, and ^{137}Cs varied from 0.1 to 338 Bq kg^{-1} (average: 11 Bq kg^{-1}) and from 0.1 to 699 Bq kg^{-1} (average: 18 Bq kg^{-1}), respectively. Three years later, the dose rate to the most affected fish species near the FDNPP have remained above baseline levels; a theoretical human consumer of 50 kg of fish caught a short distance (3 km) from the FDNPP in 2013 would have received a total committed effective dose of approximately 0.95 mSv year^{-1}, with FDNPP contributing ~14% of total committed effective dose (Johansen et al., 2014).

Numerous studies have assessed the impacts of the Fukushima accidents on the marine and terrestrial environments in both Japan and other countries (Aliyu et al., 2015a; Buck, 2011; Chen et al., 2014; Kim et al., 2012; Periáñez et al., 2013; Sin et al., 2014; Wu et al., 2013). For instance, Aliyu and co-authors concluded that, within Japanese Coastal areas, the accident did heavier damage to marine bionts compared to terrestrial flora and fauna. Even though many of the studies outside Japan have demonstrated that the levels of radiocesium in seafood were not likely to have discernible health effects (Chen et al., 2014), investigations of the marine environment have continued in order to address public concern about the impact of the accident.

Buck (2011) reported that the radiation in the ocean will be diluted along the Japanese coast; however, exposed marine organisms and contaminated tsunami debris would likely reach the USA. Time-series measurements of ^{134}Cs and ^{137}Cs in seawater showed that the first arrival of the Fukushima derived radio-cesium in a location 1500 km west of British Columbia, Canada was in June 2012, which is 1.3 years after the accident (Smith et al., 2015). Wu et al. (2013) showed that the activity concentration of ^{137}Cs in the China Sea ranged from 0.75 ± 0.07 to 1.43 ± 0.08 Bq m^{-3}, which is below the regulatory standard in China. More recent reports have suggested that Fukushima-derived cesium arrived off the coast of British Columbia, Canada in February 2015, and off the coast of Del Mar., CA, USA, in April 2015 (KO Buesseler, 2015 pers. comm., 3 September).

Strontium-90 is of significant health and environmental concern given that it is a calcium analog and, thus, may bioaccumulate in the bones and teeth of many organisms. The activity concentrations of ^{90}Sr have been measured in selected "hotspots" around Japan (Steinhauser et al., 2013b). The authors reported that the ^{90}Sr activity concentration in soil and vegetation samples from the hot spots was lower than the Japanese regulatory limit. The activity concentrations of ^{90}Sr were less than 10% of ^{137}Cs. The contamination of the marine ecosystem by fission products from FDNPP had raised questions about the safety of consuming seafood from the regions of Japan. Continuous monitoring programs were initiated by the Japanese authorities to ensure that the levels of radioactivity in the Prefectural waters remained low. This can only be achieved by ensuring that no further release of contaminated cooling water from the destroyed nuclear facility occurs. Although few studies have explicitly examined the biological consequences of radio-strontium in natural systems, perhaps in part because of the technical challenges and financial costs related to its measurement, its presence in aquatic systems is of great concern because of the possibility of biomagnification in the food web.

7. Impacts of FDNPP accident on terrestrial ecosystems

Although it is perhaps fortunate for the people of Japan that due to prevailing meteorological conditions at the time of the accident, the bulk of radioactive emissions were transported to the ocean (Evangeliou et al., 2014b), there is a growing body of literature that documents a range of physiological, developmental, genetic, morphological, and behavioral consequences of exposure to anthropogenic radioactivity derived from the FDNPP accident on the terrestrial flora and fauna of the Fukushima region of Japan. Hiyama et al. (2012) used the pale blue grass butterfly, *Zizeeria maha*, as indicator species to evaluate the environmental impact of ionizing radiation. Some of the morphological abnormalities detected in some individuals of adult butterflies collected in May 2011 increased in number and with time. Similar results were also observed when the sampled adult butterflies were compared to old museum specimens. Genetic studies indicated that certain abnormalities were inherited in the F2 generation of the grass butterfly. These abnormalities may be explained by random mutation in important genes or by epigenetic mechanisms. By comparing their data with those obtained from control populations, the authors concluded that Fukushima-derived anthropogenic radionuclides likely caused the morphological and genetic damage to the butterfly species. The study by Hiyama et al. (2012) was significantly strengthened by laboratory experiments that used both internal and external radiation sources, and these findings validated observations of the elevated mutation rates and phenotypic effects observed in the field (Møller and Mousseau, 2013). However, a recent article by Copplestone and Beresford (2014) has highlighted some drawbacks in the work of Hiyama and colleagues. Copplestone and Beresford (2014) argued that based on data reported by Hiyama et al. (2012) the lethal dose required to kill 50% of exposed individuals (LD_{50}) was equivalent to the radiation levels that organisms receive from exposure to background natural radiation sources. This disconnect between empirical findings and predictions based on theoretical models highlights the many uncertainties in accurately estimating doses in wild populations (Caffrey et al., 2014); it is likely that wild animals are significantly more sensitive

to the effects of radiation than predicted by conventional approaches (Garnier-Laplace et al., 2013).

Low blood cell counts were reported in wild monkeys from the forests of Fukushima City, and compared to the counts in monkeys from Aomori prefecture, 400 km away (Ochiai et al., 2014). The total radiocesium concentration in the muscle of Fukushima wild monkeys ranged from 78 to 1778 Bq kg^{-1}, while the cesium concentration in the control population was reported to be below detection limits. A negative correlation was observed between activity concentrations of Cs and low blood counts in juvenile Fukushima monkeys. Ochiai et al. (2014) concluded that the observed hematological aberration was due to exposure to Fukushima-derived anthropogenic radionuclides. This study was based on a rather limited ecological design. First, the population sizes of the monkeys (61 from Fukushima and 31 from Shimokita) used in the study were rather small and did not include individuals from the more contaminated areas of the region. A similar result showing hematological variations in the Fukushima monkeys had been presented by the same group of authors in an earlier study (Hayama et al., 2013). The level of radiocesium in the Fukushima wild monkeys was similar to that found in sheep in some parts of the UK following the Chernobyl accident—that is, extremely low in terms of damage to the animals themselves (Stallard, 2014). However, the findings of this study are greatly strengthened by comparisons to children living in contaminated areas of northern Ukraine near Chernobyl where similar hematological effects were observed by Stepanova et al. (2008).

A recent study of bull sperm and testes from the Fukushima region found no evidence of significant histological changes (Yamashiro et al., 2013); however, this study was very preliminary with only two bulls from a relatively uncontaminated part of Fukushima used for the analysis of sperm. However, further investigation of reproductive function in a diversity of organisms is warranted given findings of similar studies in Chernobyl which have found evidence for significant male reproductive impairment stemming from exposure to radionuclides (e.g. Hermosell et al., 2013; Møller et al., 2014).

Conservation biologists are concerned with the effects of environmental perturbation on population abundances and biodiversity as these are indicators of overall ecosystem "health." Initial studies of the Fukushima region in July 2011 surveyed abundance and diversity of many species at 300 locations that varied from in exposures of 0.5 μSv/h to more than 30 μSv/h. Several bird species, as well as butterflies and cicadas, were significantly reduced in numbers in the more radioactive areas, even after correcting for other environmental factors known to influence population sizes (Møller et al., 2012, 2013). An additional three years of biological inventories at 400 locations documented that bird abundance and diversity had continued to fall at contaminated locations (Møller et al., in pressb; Møller et al., in pressc). A more detailed analysis of barn swallow (*Hirundo rustica*) populations found strong evidence of population declines, although preliminary molecular analysis yielded no evidence for significant increases in genetic damage (Bonisoli-Alquati et al., 2015). A recent reanalysis of the Møller et al. (in pressa, in pressb) data uses modern radioecological methods to reconstruct doses to individual birds and provides evidence for dose response effects on bird abundances adding further support to the hypothesis that radioactive contaminants are the likely cause of the observed declines in Fukushima birds (Garnier-Laplace et al., in press). Overall, these investigations of population effects are similar to those found for Chernobyl-affected regions of Ukraine and Belarus, providing some additional support for the hypothesis that radioactive contaminants are the likely causes for the observed declines (Møller and Mousseau, 2009, 2006, 2007a, 2007b, 2011b, 2015; Mousseau and Møller, 2014).

The effects of the FDNPP accident on goshawk (*Accipiter gentilis fujiyamae*) reproduction in North Kanto, Japan were investigated by Murase et al. (2015). This is the only study comparing the situation before and after the accident using the same population. The authors of this recent work compared the historical data concerning the four stages of goshawk reproduction namely occupancy (stage 1), incubating (stage 2), hatching (stage 3), and fledging (stage 4). The study spans more than two decades (22 years) of data collection. The results suggest that radiation had a negative impact on reproductive performance with a progressive decline in the four stages of reproduction over time being directly related to the air dose rate under nests. Another recent paper by Matsui et al. (2015) has demonstrated that radiation exposure in avian populations depends on the quality (or weight) of nesting material. The authors suggest that the effects of elevated levels of radioactivity in nests could be of particular importance to offspring that are hatched in highly contaminated nests or during the incubation stage of reproduction given the known radio-sensitivity of early developmental stages.

A recent study of Japanese fir trees (*Abies firma*) suggests plants have also been affected by radioactive contaminants (Watanabe et al., 2015). This study compared trees at four locations that varied from 0.13 μSv/h to 33.9 μSv/h ambient dose rate and found a highly significant positive relationship between frequencies of morphological abnormalities and ambient radiation levels. Although the mechanism underlying the observed abnormalities is unknown, similar effects have been reported for pine trees in Chernobyl (e.g. Kozubov and Taskaev, 2006; Mousseau et al., 2013) providing support for the suggestion that radionuclides are the cause of the observed developmental abnormalities.

The potential ecological consequences of the Fukushima accident have been assessed using the ERICA assessment tool for different types of biota in two studies (Aliyu et al., 2015a; Garnier-Laplace et al., 2011). The exposure profile to ^{131}I, ^{134}Cs, and ^{137}Cs one month after the FDNNP was used by Garnier-Laplace et al. (2011) for estimated dose rate (mGy day^{-1}) calculations to a variety of organisms. Dose rates to selected reference species were 1.5 mGy day^{-1} for birds, 2.3 mGy day^{-1} for soil invertebrates, and 3.9 mGy day^{-1} for forest trees. Although largely hypothetical, such dose rates could be biologically significant, especially for longer-lived or radiosensitive organisms.

A long-term monitoring program of human health and ecosystem consequences of chronic exposure to radioactive materials in the environment would contribute to a better understanding of effects exposure to low-dose ionizing radiation. In studying the immediate and long-term effects of exposure to low dose radiation one also needs to understand a number of relevant issues such as the possible induction of long-term or multigenerational effects and the capacity of organisms to adapt to stress. There is considerable evidence that many species are able to evolve resistance or tolerance to pollutants through the process of natural selection and a recent study of birds in Chernobyl indicates that adaptation to radiation is possible, at least for some species (Galván et al., 2014). On the other hand, there is considerable evidence for radiation-induced damage to DNA that likely reduces individual fitness and that of offspring when such genetic damage (i.e. mutations) is passed to subsequent generations. Chronically exposed populations are subject to multigenerational mutational accumulation and are more likely to experience local extinction events (Higgins and Lynch, 2001), a subject of great concern to conservation biologists (Møller and Mousseau, 2011a). At this stage, relatively little is known about the genetic (or epigenetic) mechanisms associated with population responses to stressful contaminants. Much greater investment in basic research is thus warranted.

8. Application of isotopes as tracers for environmental processes

Natural and anthropogenic radionuclides can be used as tracers for environmental processes, particularly oceanic transportation and water mixing. In the oceans, the behavior of cesium is thought to be conservative; that is, it is soluble (with only a small fraction adsorbed to marine particulates) and is carried primarily with ocean waters. As such, cesium has been used as a tracer of water mass mixing and transport (Aoyama et al., 2011; Bowen et al., 1980; Buesseler et al., 1991). Following the FDNPP disaster, cesium activity and the $^{134}Cs/^{137}Cs$

activity ratio have been measured to assess its environmental effects and to study oceanographic processes. Studies based on samples of atmosphere, soil, seawater and marine biota off Fukushima have shown that the $^{134}Cs/^{137}Cs$ activity ratio in the Fukushima derived radionuclides was nearly 1.0 (Buesseler et al., 2011; Hirose, 2012). This ratio is higher than the $^{134}Cs/^{137}Cs$ activity ratio of 0.5 measured in the Chernobyl fallout (IAEA, 1986; UNSCEAR, 2000). This makes the tracking of Fukushima derived radionuclides in the ocean quite straightforward since, given its relatively short half-life (2 years), the only source of ^{134}Cs in the North Pacific at this time would be the FDNPP. Hence, in addition to the elevated cesium activities, the presence of ^{134}Cs is a unique isotopic signature for tracking these waters and provides a means to study the rates of vertical and horizontal mixing processes in the Pacific Ocean.

Pu has high particle affinity compared with cesium, and thus provides a tool for studying a variety of processes in the marine environment such as particle fluxes and scavenging, and for validating various biogeochemical ocean models. However, the most useful aspect of Pu as an environmental tracer is its well-defined source terms (Lindahl et al., 2010). This is because the composition of Pu isotopes from different sources in marine environments varies widely due to the characteristic isotopic composition corresponding to the means of production. For example, reactor-grade Pu typically contains more than 35% ^{239}Pu with a $^{240}Pu/^{239}Pu$ atom ratio of 0.2–1.0 after fuel burn-up (Yamana et al., 2001). In contrast, the $^{240}Pu/^{239}Pu$ ratio in weapons-grade Pu is much lower (0.02–0.06) because burn-up is kept low to minimize the production of higher Pu isotopes (Marka, 1993). This review shows that the Fukushima derived $^{240}Pu/^{239}Pu$ ratio is 0.216–0.255, which is higher than that of global fallout (0.18), but lower than that of Chernobyl. A summary of the composition of Cs and Pu isotopes for Fukushima, Chernobyl and the global fallout is presented in Table 4.

9. Conclusions

Despite the large number of studies aimed at assessing the impacts of the FDNPP accident, considerable uncertainty remains concerning all aspects of this disaster. Source term estimates vary from 12 to 36.7 PBq for ^{137}Cs and 150 to 160 PBq for ^{131}I. Predictions of human health impacts range to 10,000 estimated deaths, and 1500 and 1800 maximum cancer mortalities and morbidities. Studies of terrestrial systems, although few, have revealed possible significant impacts on some species, notably birds and butterflies; the paucity of data may reflect the very few funded research projects currently underway. Similarly, impacts on marine systems are largely unknown, with data limited to estimates of body burdens for commercially important species. It is perhaps surprising that there have not been greater efforts to directly assess the health and environmental impacts of this disaster.

Our review of the current literature has shown that it took the radioactive plume resulting from the FDNPP accident a longer time to travel around the world's atmosphere than that of the Chernobyl plume. The explanation is that Fukushima radioactive clouds were released into the PBL, whereas Chernobyl plumes were injected into the troposphere. Of paramount need is a concise summary of field measurement data for FDNPP-derived radionuclides detected in locations around the world. These field measurement data could be used to drive computer models for robust worldwide assessments of human and environmental impacts of the FDNPP accident. There is no substitute for direct measurements when attempting to assess the hazards and risks of any disaster.

Despite the environmental ramifications of fission products released from nuclear accidents and atomic bomb testing, scientists have developed few methods for using radioactive fission products as tracers of environmental processes such as ocean mixing, atmospheric transport and soil processes. In this review, we have provided a few examples for how Fukushima-derived radionuclides have been used to study marine and atmospheric processes. There are undoubtedly many other potential applications of these approaches.

Overall, it is very clear that much remains to be learned concerning the impacts of the FDNPP disaster and we hope that greater investments will be made in the coming years into the basic scientific study of the global health and environmental impacts of this accident.

Table 4
Anthropogenic nuclide isotope ratio characteristics.

Source term	Fukushima	Chernobyl	Global fallout
$^{134}Cs/^{137}Cs$ (activity ratio)	~1[i]	0.5[n]	–
$^{137}Cs/^{239,240}Pu$ (activity ratio)	1.95×10^5–2.53×10^7[j]	–	38.5[r]
$^{238}Pu/^{239,240}Pu$ (activity ratio)	1.2[k]	0.5[p]	0.032[t]
$^{241}Pu/^{239,240}Pu$ (activity ratio)	107.8[m]	–	13–16[u]
$^{240}Pu/^{239}Pu$ (atom ratio)	0.323–0.330[m]	0.408[q]	0.180[v]
$^{241}Pu/^{239}Pu$ (atom ratio)	0.128–0.135[m]	–	0.00194[w]

[i] Buesseler et al. (2011).
[j] Hirose (2012).
[k] Lujanienė et al. (2012a, 2012b).
[m] Zheng et al. (2012).
[n] UNSCEAR (2000).
[p] IAEA (1986).
[q] Muramatsu et al. (2000).
[r] Kelley et al. (1999), Krey et al. (1976).
[t] Krey et al. (1976).
[u] Paatero et al. (2012).
[v] Kelley et al. (1999), Krey et al. (1976).
[w] Holm (1988).

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