Current Knowledge Concerning the Impacts of the Fukushima Daiichi Nuclear Power Plant accident on the environment

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Abstract

It is now more than four years since the Fukushima nuclear accident; how it affects the environment will continue to be an issue of great concern to the public and energy policy makers. Thus, scientists around the world have been investigating the impacts of the accident and the question of how the Fukushima nuclear accident has affected man and his environment will remain one of great interest for centuries. Here, we review the current understanding of the Fukushima nuclear accident’s interactions with the environment, which includes studies on the source term assessment, radioactive contamination of the atmosphere, soil and the marine environment. This review addresses some fundamental scientific questions concerning the environmental impacts of Fukushima nuclear accident with the aim of stimulating further discussion on the topic. Finally, the paper presents a concise discussion of how the released radionuclides from the Fukushima nuclear accident could be used as tracers for studying environmental processes.

Keywords: Fukushima nuclear accident, environmental effect, radioactive contamination, 137Cs
1. Introduction

The confluence of a magnitude 9.0 earthquake, a subsequent gigantic tsunami, and design flaws related to the plant site and emergency cooling systems resulted in the Fukushima Dai-ichi nuclear power plant accident (FDNPP) on March 11, 2011. As a result, enormous quantities of radionuclides were released into the atmosphere and the ocean. Major radionuclides released to the atmosphere included $^{131}I$ ($t_{1/2} = 8.02$ d), $^{134}Cs$ ($t_{1/2} = 2.06$ y), $^{137}Cs$ ($t_{1/2} = 30.07$ y) and $^{90}Sr$ ($t_{1/2} = 28.78$ y), which were then distributed globally leading to the contamination of both terrestrial and marine environments. Additionally, large amounts of radionuclides were directly released into the ocean as liquid wastes via discharge from the FDNPP, which widely contaminated seawater in areas offshore of Japan. The total amounts of $^{137}Cs$ and $^{90}Sr$ directly released to the sea have been estimated to be 1~42 PBq (1 PBq = 10$^{15}$ Bq) and 0.1~2.2 PBq, respectively. Obviously, there has been considerable uncertainty concerning the total amounts released from the FDNPP. It is clear that currently there is a poor understanding about some fundamental questions of the FDNPP (for example, the release amounts and rate of radionuclides); hence, we cannot derive reliable conclusions concerning the impacts of FDNPP on the environment. Answering some of these fundamental questions will lead to a better understanding of how the uncontrolled release of radioactive fission products from the FDNPP has and will continue to affect both human and non-human biota.

This article will summarize the current state of understanding about the major radionuclides derived from the FDNPP, including the amounts released and isotopic composition, the effects on environment (seawater, sediment, terrestrial and marine biota), and their application as tracers for environmental processes. This paper also aims to present the results of global field measurements of radionuclides stemming from Fukushima, which were obtained from the literature. This field measurement data may be used in the investigation of environmental process as well as the basis for conducting country-wise comparative impacts assessment of the FDNPP accident.

2. Source term assessment
Following the Fukushima nuclear accident, there was tremendous public concern about the release of radionuclides into the environment (i.e. atmosphere and ocean). Several research institutes and scientists conducted many studies in an attempt to estimate the total release amounts as a measure of the source term. For instance, the Japanese government estimated the source term for $^{131}$I and $^{137}$Cs to be 160 PBq and 15 PBq, respectively as the total atmospheric releases of radioactivity from the FDNPP. The Nuclear Safety Commission of Japan (NSCJ) estimated that the total amounts of $^{131}$I and $^{137}$Cs discharged into the atmosphere were 150 PBq and 12 PBq, respectively. Masson, et al. estimated that $^{131}$I (153 PBq) and $^{137}$Cs (13 PBq) were released to the atmosphere. Other researchers have reported the total emission of $^{137}$Cs as high as 13–35.8 PBq. To estimate the source terms of radioactive cesium ($^{137}$Cs) and iodine ($^{131}$I), Chino, et al. adopted an estimation method to compute the source terms using an atmospheric dispersion model. Their results were comparable to that of the Ministry of Education, Culture, Sports, Science and Technology, Japan. The authors verified the validity of this technique in later studies. In a most recent study, the group improved its computational methodology to account for limitations in their earlier studies. The new study considered a new scheme of parameterizations for wet, dry, and fog-water depositions of both gaseous and particulate radionuclides. Environmental data that are coupled into the new model include those from other parts of Japan and the Pacific Ocean. The new study methodology estimated the total release amount of total $^{131}$I and $^{137}$Cs to be 151.0 and 14.5 PBq, respectively considering environmental data from both land and sea (New-landsea). For the new land data, the total release amount for total $^{131}$I and $^{137}$Cs are ~27% and ~32%, respectively less than the results of the WINSPEEDI-II simulation that considered New-landsea data. The reported total release amount of $^{131}$I is in good agreement with the estimate of the NSCJ and lies within the range estimate for total release amount of $^{131}$I by this review ~ (150 – 160) PBq (See Table 1).

Additionally, the important source of ocean radioactive contamination was
derived from direct discharge into the cooling waters at the NPP. Accurate estimation of radioactive pollutants directly discharged into the sea is the basis for assessment of the impact to the marine habitat. There have been many studies concerning this question. Using a regional ocean model, Tsumune, et al. \(^6\) estimated that the total \(^{137}\)Cs directly released to the ocean was \(3.5 \pm 0.7\) PBq from March 26 to the end of May, 2011. The total amounts of \(^{137}\)Cs directly released into the ocean have been estimated to range from 1 to 42 PBq \(^5\)-\(^7\). We have summarized the total release amounts of major radionuclides derived from the FDNPP and compared them with global fallout and the source term from the Chernobyl accident (Table 1)\(^8\), \(^19\), \(^20\).

Despite the uncertainty associated with the total release of fission products from Fukushima, Table 1 depicts that the Chernobyl accident resulted in the largest discharge of \(^{137}\)Cs into the environment (85 PBq), and Fukushima resulted in the largest accidental source term release to the ocean. Ten months later, in the winter of 2012, the mean value for water column inventories of Fukushima-derived radiocesium (\(^{137}\)Cs) and bomb-derived \(^{134}\)Cs were nearly equal. These values were \(1020 \pm 80\) and \(820 \pm 120\) Bq m\(^{-2}\), respectively \(^21\). This suggests that the accident and nuclear bomb testing had almost the same impacts on the Pacific Ocean during this period.

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

| Radionuclide | Global fallout | | Chernobyl | | Fukushima | |
|--------------|----------------|--|-----------|----------------|-----------|
|              | atmosphere | Ocean | Atmosphere | Ocean | Atmosphere | Ocean |
| \(^{131}\)I   | 1760         | 160   | 153        | 15   | 3.5±0.7   |       |
| \(^{137}\)Cs | 950          | 600   | 85         | 16   |           |       |
| \(^{90}\)Sr  | 600          | 380   | 1          | 0.14 | 0.1       | 0.01   |
| \(^{239,240}\)Pu | 10.87    | 6.6   | 0.087      | (1-2.4)×10\(^{-6}\) |   |
3. **Environmental effect**

3.1 *Atmosphere transportation*

Vast amounts of radioactivity were emitted into the atmosphere by the accident at the FDNPP, and the radioactive plume spread over the globe in the free troposphere by the prevailing westerly winds. Table 2 shows the results of measurements and detection of Fukushima-derived radioactive fission products in receptor locations (sampling points) around the globe. The readings are for air concentrations (in Bq m\(^{-3}\)), soil deposition (in Bq m\(^{-3}\)) and concentration in both fresh water and marine environments (in Bq l\(^{-1}\)). The data in Table 2 may be used in the investigation of environmental processes as well as basis for conducting country wise comparative impacts assessment of the FDNPP accident.
Table 2 Fukushima-derived anthropogenic radioisotopes detected in locations around the world

<table>
<thead>
<tr>
<th>Location, Country</th>
<th>Media (Soil Air or Water): Activity concentrations of radionuclide, (ref)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Japan</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 9.9, $^{134}$Cs: 10.13, $^{137}$Cs: 10.27, $^{129m}$Te: 4.0, $^{136}$Cs: 123 $^{25}$</td>
</tr>
<tr>
<td></td>
<td>Soil (Bq m$^{-2}$): $^{137}$Cs: 900 $^{28,29}$</td>
</tr>
<tr>
<td>South Korea</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 3.12 x 10$^{-3}$, $^{134}$Cs: 1.19 x 10$^{-3}$, $^{137}$Cs: 0.25 x 10$^{-3}$ $^{30}$</td>
</tr>
<tr>
<td></td>
<td>Soil (Bq m$^{-2}$): $^{137}$Cs: 649, $^{134}$Cs: 802 $^{31}$</td>
</tr>
<tr>
<td>United States</td>
<td>Air (Bq m$^{-3}$): $^{134}$Cs: 1.11 x 10$^{-3}$, $^{137}$Cs: 1.18 x 10$^{-3}$, $^{131}$I: 3.85 x 10$^{-3}$ $^{32}$</td>
</tr>
<tr>
<td>Krasnoyarsk (Russia)</td>
<td>Water (Bql$^{-1}$): $^{131}$I: 0.62, $^{137}$Cs: 0.075, $^{134}$Cs: 0.095 $^{23}$</td>
</tr>
<tr>
<td>Thessaloniki (Greece)</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 497 x 10$^{-6}$, $^{137}$Cs: 145 x 10$^{-6}$, $^{134}$Cs: 126 x 10$^{-6}$ $^{24}$</td>
</tr>
<tr>
<td></td>
<td>Water (Bql$^{-1}$): $^{131}$I: 0.7 $^{23,24}$</td>
</tr>
<tr>
<td>Ibaraki (Japan)</td>
<td>Soil (Bq m$^{-2}$): $^{137}$Cs: 1.4 x 10$^{4}$ $^{33}$</td>
</tr>
<tr>
<td>China</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 1720 x 10$^{-6}$, $^{134}$Cs: 247 x 10$^{-6}$, $^{137}$Cs: 289 x 10$^{-6}$, $^{138}$Cs: 23 x 10$^{-3}$ $^{34}$</td>
</tr>
<tr>
<td></td>
<td>Soil (Bq kg$^{-1}$): $^{131}$I: 2.68 $^{34}$</td>
</tr>
<tr>
<td>Milan (Italy)</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: ~4.2 x 10$^{-4}$, $^{137}$Cs: ~9.4 x 10$^{-5}$, $^{134}$Cs: 8 x 10$^{-5}$ $^{35}$</td>
</tr>
<tr>
<td>Czech Republic</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 5.6 x 10$^{-3}$, $^{137}$Cs: 0.72 x 10$^{-3}$, $^{134}$Cs: 0.64 x 10$^{-3}$ $^{36}$</td>
</tr>
<tr>
<td>Lithuania</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 3700 x 10$^{-6}$, $^{137}$Cs: 1040 x 10$^{-6}$ $^{37,38}$</td>
</tr>
<tr>
<td>France</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 2.4 x 10$^{-3}$, $^{134}$Cs: 0.2 x 10$^{-3}$, $^{137}$Cs: 0.7 x 10$^{-3}$ $^{39,40}$</td>
</tr>
<tr>
<td>Bratislava (Slovakia)</td>
<td>Air (Bq m$^{-3}$): $^{131}$I: 0.5 x 10$^{-3}$, $^{137}$Cs: 0.07 x 10$^{-3}$ $^{41}$</td>
</tr>
<tr>
<td>Location</td>
<td>Sample Type &amp; Description</td>
</tr>
<tr>
<td>---------------------------</td>
<td>------------------------------------------------------------------</td>
</tr>
<tr>
<td>Spain</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Serbia</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Vietnam</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>UK</td>
<td>Grass (Bq kg⁻¹): dry matter for ¹³¹I: 55, dry matter for ¹³⁷Cs: 8</td>
</tr>
<tr>
<td>Romania</td>
<td>Water (Bq L⁻¹):</td>
</tr>
<tr>
<td>Svalbard (Norway)</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Lisbon (Portugal)</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td></td>
<td>Soil (Bq m⁻²):</td>
</tr>
<tr>
<td>Vancouver Island (Canada)</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Finland</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Iberian Peninsula</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Istanbul (Turkey)</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Taiwan</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Austria</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td></td>
<td>Rain water (Bq L⁻¹):</td>
</tr>
<tr>
<td>Hungary</td>
<td>Air (Bq m⁻³):</td>
</tr>
<tr>
<td>Monaco</td>
<td>Air (Bq m⁻³):</td>
</tr>
</tbody>
</table>

N.B Only maximum detected values were considered for all sampling points.
<table>
<thead>
<tr>
<th>Location</th>
<th>Air (Bq m$^{-3}$):</th>
<th>Soil (Bq kg$^{-1}$):</th>
</tr>
</thead>
<tbody>
<tr>
<td>Germany</td>
<td>$^{131}$I: 2.1 x 10$^{-3}$</td>
<td>$^{131}$I: 0.093, $^{137}$Cs: 3.07, $^{134}$Cs: 0.69</td>
</tr>
<tr>
<td>Darwin (Australia)</td>
<td>$^{133}$Xe: 12 x 10$^{-3}$</td>
<td>No Fukushima-derived radio iodine and cesium was detected in Australia.</td>
</tr>
<tr>
<td>Krakow (Poland)</td>
<td>$^{131}$I: 6.08 x 10$^{-3}$, $^{134}$Cs: 0.50 x 10$^{-3}$, $^{137}$Cs: 0.474 x 10$^{-3}$</td>
<td></td>
</tr>
</tbody>
</table>

It has been estimated that it took radionuclides from FDNPP ~18 days to reach locations around the globe. This is much longer than the time (~10 days) it took for the radiation clouds emitted from the Chernobyl accident to do the same, and it is longer than the time (~13 days) for Asian dust to go around the world. It is known that the largest portion of the radioactive plume from Fukushima was transported through the atmosphere and deposited over the Pacific Ocean. This might have resulted in the longer time for travel around the world as the space and temporal variation of the boundary layer over the oceans is relatively slow. This is because the surface temperature of the sea varies little during the diurnal cycle owing to large mixing within the top layer of the ocean.

Airborne radioactivity from FDNPP accident was detected at many laboratories around the world and possible health effects from the radioactivity transported by the radioactive plumes from Japan was of concern. In Japan, as of October 21 2011 (7 months after the accident), the air concentrations of $^{134}$Cs and $^{137}$Cs in Fukushima city were still elevated to 613 µBq m$^{-3}$ and 829 µBq m$^{-3}$, respectively. Other prefectural governments also conducted environmental radioactivity monitoring, which included 45 stations covering the Japanese Islands. Outside Japan, as shown in Table 2, radioactive materials were detected in Korea, Vietnam, Taiwan, U.S., Canada, and Europe. In Europe, the first signs of the radioactive cloud were detected on March 19th, 2011 (slightly more than one week after the accident) while the first peak
of activity level was observed between March 28th and 30th. Measurements have shown that airborne activity levels are now of minimal concern for public health in Europe. The activity concentrations of the radionuclides in the plume exponentially decreased with downwind distance due to deposition (dry and wet) and scavenging.

3.2 Soil deposition

Deposition of Fukushima-derived radionuclides on land mostly stemmed from the release to the atmosphere of relatively volatile fission products, most importantly $^{137}$Cs, $^{134}$Cs and $^{131}$I and radioactive noble gases, which present a large risk for internal radiation exposure via inhalation and ingestion of contaminated agricultural crops, stock farming, and thus human life. The soils around the FDNPP and neighboring prefectures have been extensively contaminated with $^{137}$Cs and $^{134}$Cs depositions of more than $10^5$ and $10^4$ Bq m$^{-2}$, respectively. Similarly, Kinoshita, et al. has also reported that the top soil deposition of $^{137}$Cs in the central-east Japan due to the Fukushima accident ranged from 0.4 to 900 kBq m$^{-2}$, which are 0.06-130 times the cumulative deposition from the atmospheric nuclear test (100-1000 Bq m$^{-2}$). It is estimated that a total of more than 5.6 PBq $^{137}$Cs was deposited over the Japanese Islands. High activity concentrations of radionuclides have been reported. For example, soil activities for $^{134}$Cs and $^{137}$Cs in the Iitate Village area (located 25-45 km northwest of the Fukushima site) were 62400 and 72900 Bq kg$^{-1}$, respectively. As of March 2011, the top soil concentration of $^{137}$Cs was measured to be 80 kBq m$^{-2}$. The environmental external radiation dose rate before the accident was approximately 0.05 µSv h$^{-1}$. Additionally, the radiation dose rate of other prefectural areas due to the Fukushima accident have also been estimated, for example, 1.14 µSv h$^{-1}$ in Naka-Dori, 4.57 µSv h$^{-1}$ in Iitate, 0.02 µSv h$^{-1}$ in the region between northern Ibaraki and eastern Saitama, and 0.23 µSv h$^{-1}$ in southern Ibaraki and northern Chiba prefectures. This clearly demonstrates that large areas in eastern and northern Japan were strongly contaminated by $^{137}$Cs derived from Fukushima, while in contrast, the western part of Japan was shielded by its mountainous terrain.
However, as with Chernobyl, deposition was highly heterogeneous even within the most contaminated areas e.g. 70, 71 with ground level readings ranging from 0.5 to >30 µSv h⁻¹ within Namie township. The information on soil contamination which was detected in countries in Asia, Europe and in North America is presented in Table 2. The advantage of the information in Table 2 is that it is based on empirical environmental data and not computer models. The advantages of empirical data stem from the fact that dependence solely on environmental models may result in over or under estimation of the impacts of the accident as it has been observed in the study of Hoeve and Jacobson 11. The authors 11, estimated that there would be an additional 130 (15-1100) and 180 (24-1800) cancer related mortalities and morbidities related to the Fukushima radiological disaster, respectively with over 90% of the projected impacts occurring in Japan. However, Beyea, et al. 72 argued that the failure by Hoeve and Jacobson 11 to consider the long-term dose estimate from radiocesium in their studies may lead to under estimation of the global effects of the accident. By including a more realistic population perspective, Beyea, et al. 72 suggested that the mid-range estimate for the number of future cancer mortalities due to Fukushima accident is probably closer to 1000 than 125. Recently, Evangeliou, et al. 73 developed a pioneering environmental modeling study that considered numerous fission products. This study estimated that cancer fatalities are likely to be fewer than 2000.

3.3 Marine environment

Other than fallout from atomic bomb testing, contamination of the marine environment following the FDNPP accident represents the most important anthropogenic radioactive release 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 the transport of radioactive pollution from leaching through contaminated soil. For instance, the total amount of ¹³⁷Cs directly released to the ocean has been estimated as 1–42 PBq 5-7. In the immediate vicinity of the plant, the Cs levels of surface ocean at the discharge point are up to 10³ times higher activities than it has been (~1-2 Bq m⁻³ ) 74, with a peak of 68 kBq L⁻¹ on
April 6, 2011. Certainly, dilution of the primary contaminant signal occurs quite 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 quite rapidly offshore. As a consequence, one month after the accident the $^{137}$Cs level decreased by a factor of $10^3$.

Honda, et al. reported that the $^{137}$Cs activity in surface seawater ranged from 0.004 to 0.284 Bq kg$^{-1}$ in the western North Pacific on 14 April to 5 May. Tsumune, et al. argued that if no additional releases occur, the effect of the $^{137}$Cs might 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. calculated the dose due to direct exposure during human indirect contact with the water to be 0.1 µSv d$^{-1}$ 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 d$^{-1}$. Thus there is unlikely to be any significant direct external dose effect on humans via this pathway.

Cesium-137 concentration in suspended solids (SS) from surface and subsurface waters off western North Pacific has been measured by Honda, et al.; it ranged from 5.92 to 32.42 Bq kg$^{-1}$ on a dry weight basis (kg-dw$^{-1}$). The same is true for $^{134}$Cs. Thus, it is likely that SS were contaminated with radionuclides from the FDNPP. The SS-seawater distribution 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 recommended $K_{ds}$ for open ocean (2000) reported.

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. Fukushima-derived radioesium was detected in zooplankton and mesopelagic fish, with the $^{137}$Cs concentration in zooplankton off western North
Pacific 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\(^{-1}\)) \cite{79,80}. In contrast, the \(^{137}\)Cs concentration in zooplankton around Japan was less than 0.1 Bq kg-ww\(^{-1}\) during the last decade \cite{81,82}. 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 \cite{19,81,82}.

Buesseler, et al. \cite{78} suggested that radiation risks of \(^{137}\)Cs derived from the FDNPP to marine organisms and human consumers of seafood are well below those from natural radionuclides. Meanwhile, the total cesium levels in demersal (bottom-dwelling) fish off Fukushima were investigated \cite{83}, where the cesium levels in 40% of fish were above the new regulatory limit (100 Bq kg\(^{-1}\) wet) set by Japan Ministry of Agriculture \cite{84}. 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. \cite{85}. 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. Long-term monitoring of marine products around the FDNPP is important before restarting the coastal fisheries \cite{85}. For other prefectures that are adjacent to Fukushima, Chen \cite{86} reported the radioactivity level in the fish products from these prefectures based on monitoring data of July 2013. The 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 \cite{86}. Three years later, the dose rate to the most impacted fish species near the FDNPP have remained above baseline levels and a theoretical human consumer of 50 kg of fish, gathered a short distance (3 km) from the FDNPP in 2013, would have received a
total committed effective dose of approximately 0.95 mSv y\(^{-1}\) with FDNPP contributing ~14\% of total committed effective dose\(^8\)

Numerous studies have assessed the impacts of the Fukushima accidents on the marine and environment in both Japan and other countries\(^{30, 88-93}\). For instance, Aliyu and co-authors concluded that within Japanese Coastal areas, the accident had heavier damage to marine bionts compared to terrestrial flora and fauna. Even though many of the studies outside Japan have demonstrated that the levels radiocesium in seafood were not likely of discernible health effects\(^8\), further investigations of the marine environment have continued in order to address public concern about the impact of the accident.

Buck\(^9\) reported that the radiation in the ocean will be diluted along the Japanese coast, however exposed marine organisms and contaminated tsunami debris will likely reach the U.S. Time series measurement of \(^{134}\)Cs and \(^{137}\)Cs in seawater showed that the first arrival of the Fukushima derived radio-cesium in North America in a location was in June 2012, which is 1.3 y after the accident\(^9\). This was in a location that is 1500 km west of British Columbia, Canada.

An assessment of the impact of the accident on marine environments in China Sea showed that the activity concentration of \(^{137}\)Cs ranged from 0.75 ± 0.07 to 1.43 ± 0.08 Bq m\(^{-3}\), which is below the regulatory standard in China\(^9\).

Strontium-90 is of significant health and environmental concerns 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 hotspot in Japan\(^9\). The author reported that the \(^{90}\)Sr activity concentration in soil and vegetation samples from the hot spots is relatively lower than the Japanese regulatory limit. The activity concentrations of \(^{90}\)Sr were less than 10\% of \(^{137}\)Cs.

4. **Effects of FDNPP accident on some terrestrial species**

Hiyama, et al.\(^9\) used the Pale Blue Grass butterfly, *Zizeeria maha*, as indicator species to evaluate the environmental impact of ionizing radiation. Observed morphological abnormalities were detected on some individuals of adult butterflies
collected in May 2011 increased in number and with time. Genetic studies indicated that some abnormalities were inherited in the F2 generation. These abnormalities may be explained by random mutation in important genes or by epigenetic mechanisms. By comparing their data with that 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. 96 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 97. However, a recent article by Copplestone and Beresford 98 has highlighted some major drawbacks in the work of Hiyama and colleagues. Copplestone and Beresford 98 argued that based on data reported by Hiyama, et al. 96, the lethal dose required to kill 50% of exposed individuals (LD$_{50}$) is 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 associated with accurate estimation of dose in wild populations 99 and the likelihood that wild animals are significantly more sensitive to the effects of radiation than predicted by conventional approaches 100. In a later study 101, the authors clarified some of the questions raised concerning their earlier study and presented some new data on the study.

Low blood cell counts were reported in wild monkeys from the forests of Fukushima City compared with the counts in monkeys from Aomori prefecture which is 400 km away 102. The total radiocesium concentration in the muscle of Fukushima
wild monkeys was measured to range 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.\(^{102}\) concluded that the observed hematological aberration was due to exposure to Fukushima-derived anthropogenic radionuclides. This study is based on ecological design that is rather limited. Firstly, the population size of the monkeys (61 from Fukushima and 31 from Shimokita) considered in the study is rather small and does not include individuals from the more contaminated areas of the region. A similar result which showed hematological variations in the Fukushima monkeys was presented by the same group of authors in an earlier study\(^{103}\). 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, i.e. extremely low in terms of damage to the animals themselves\(^{104}\). The greatest strength of this study comes from comparisons to children living in contaminated areas of northern Ukraine near Chernobyl where similar hematological effects were also observed\(^{105}\).

A recent study of bull sperm and testis from the Fukushima region found no evidence for significant histological changes in the testes or sperm morphology\(^{106}\) although this study was very preliminary with only two bulls from a relatively uncontaminated part of Fukushima represented for the analysis of sperm.

Conservation biologists are concerned with the effects of environmental perturbances 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 at 300 locations that varied from 0.5 usv/h to more than 30 usv/h. It was found that several bird species as well butterflies and cicadas showed significant reductions in numbers in the more radioactive areas even after correcting for other environmental factors known to influence population sizes\(^{107, 108}\). Following an additional three years of biotic inventories at 400 locations, it was found that bird abundance and diversity had continued to fall at contaminated locations\(^{70}\).
A more detailed analysis of barn swallow (*Hirundo rustica*) populations found strong evidence of population declines although preliminary molecular analysis found no evidence for significant increases in genetic damage. Overall, these investigations of population effects bear a striking resemblance to those found for Chernobyl-affected regions of Ukraine and Belarus, providing some support for the hypothesis that radioactive contaminants are the likely cause for the observed declines.

The radiological consequence of the Fukushima accident has been assessed using ERICA assessment tool for different biota in two known studies. The exposure profile to $^{131}$I, $^{134}$Cs, and $^{137}$Cs, one month after Fukushima was used by Garnier-Laplace, et al. for the dose rates (mGy d$^{-1}$) calculations. The dose rate to selected reference species were 1.5 (mGy d$^{-1}$) for birds, 2.3 (mGy d$^{-1}$) for soil invertebrates, and 3.9 (mGy d$^{-1}$) for forest. Such dose rates could be biologically significant especially for longer-lived or radiosensitive organisms.

5. Application of isotopes as tracers for environmental processes

In the oceans, the behavior of cesium is thought to be conservative, i.e. it is soluble (less than 1% adsorbed to marine particulates) and is carried primarily with ocean waters and as such has been used as a tracer of water mass mixing and transport. Following the Fukushima nuclear accident, cesium activity and the $^{134}$Cs/$^{137}$Cs activity ratio have been measured to assess its environment effect and to study oceanographic processes. Studies have shown that the $^{134}$Cs/$^{137}$Cs activity ratio in the Fukushima derived radionuclides was nearly 1.0, which is based on the samples of atmosphere, soil, seawater and marine biota off Fukushima. It is higher than that in the Chernobyl fallout ($^{134}$Cs/$^{137}$Cs activity ratios:0.5). This makes the tracking of Fukushima derived radionuclides in the ocean quite straightforward, since given its relatively short half-life (2 yr), the only source of $^{134}$Cs in the North Pacific at this time would be the Dai-ichi NPPs. Hence in addition to the elevated cesium activities, the presence of $^{134}$Cs is a unique isotopic signature for tracking these waters and...
providing a means to study the rates of vertical and horizontal mixing processes in the Pacific Ocean.

Compared with cesium, Pu has high particle affinity, which thus provides a tool for studying a variety of processes in the marine environment such as particle fluxes and scavenging, the validation of various biogeochemical ocean models. However, the most useful aspect of Pu as an environmental tracer is its well-defined sources terms. This is because the composition of Pu isotopes from different sources in marine environment varies significantly 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. The $^{240}$Pu/$^{239}$Pu atom ratio in weapon-grade Pu is much lower (0.02–0.06) because burn-up is kept low to minimize the production of higher Pu isotopes. The current study shows 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 3.

<table>
<thead>
<tr>
<th>Source</th>
<th>Fukushima</th>
<th>Chernobyl</th>
<th>Global fallout</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{134}$Cs/$^{137}$Cs (activity ratio)</td>
<td>~1</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>$^{137}$Cs/$^{238,240}$Pu (activity ratio)</td>
<td>$1.95 \times 10^7$–$2.53 \times 10^7$</td>
<td>38.5</td>
<td></td>
</tr>
<tr>
<td>$^{239,240}$Pu/$^{239,240}$Pu (activity ratio)</td>
<td>1.2</td>
<td>0.5</td>
<td>0.030</td>
</tr>
<tr>
<td>$^{241}$Pu/$^{239,240}$Pu (activity ratio)</td>
<td>107.8</td>
<td></td>
<td>13–16</td>
</tr>
<tr>
<td>$^{240}$Pu/$^{239}$Pu (atom ratio)</td>
<td>0.216–0.255</td>
<td>0.408</td>
<td>0.180</td>
</tr>
<tr>
<td>$^{241}$Pu/$^{239}$Pu (atom ratio)</td>
<td>0.132</td>
<td>0.00194</td>
<td></td>
</tr>
</tbody>
</table>

In soil, Fukushima-derived $^{137}$Cs has been deposited in Japan and other parts of the globe. Atmospheric and soil measurements of Fukushima-derived $^{137}$Cs activity concentrations, revealed that the detection of $^{137}$Cs in the ambient aerosol ended 2–3 months after the initial emissions. This indicates that topsoil activity concentrations of $^{137}$Cs that are obtained after this period are reliable for
monitoring soil processes as most of the Cs in the air has been deposited on the soil and surface of water bodies. Cesium is absorbed on soil particles, thus restraining its movement by either biological or chemical means. The movement of ${}^{137}\text{Cs}$ in the environment is by physical processes hence it can be used as a tracer for studying the processes of sedimentation and erosion. By determining the spatial distribution patterns of ${}^{137}\text{Cs}$ in vertical and horizontal planes across the landscape, the rates of soil loss or deposition can be measured for different parts of a watershed. As for sedimentation rate, it can be measured by matching the vertical distribution of ${}^{137}\text{Cs}$ in sediments with the temporal deposition of fallout ${}^{137}\text{Cs}$ from the atmosphere to locate sediment horizons. Highlights of some of the state-of-the-art methods for using anthropogenic radionuclides as tracers in soil process are presented in Zapata. The soil retention mechanism of Cs derived from FDNPP was investigated by Mishra, et al. The authors reported that Cs is sorbed on the surface of clay particles; and analysis of the vertical profile of deposited Cs showed that the radionuclide largely retained in the top 5cm of the soil.

**Conclusions**

In this paper we have reviewed the current state of knowledge concerning the environmental impacts of the Fukushima nuclear accident. First, we assessed the source term for the Fukushima nuclear accident and concluded that the FDNPP released a total amount of 10~15 PBq of ${}^{137}\text{Cs}$ to the atmosphere and 1~42 PBq to the ocean. Next, we analyzed the radioactive contamination of atmosphere, soil and marine environments (seawater, suspended solid and biota), whereby global contamination was mediated through atmospheric dispersion. High deposition rates of fission products have been observed in Japan. The radionuclides directly released into the ocean caused activity levels and total inventory to significantly increase. The concentrations of radionuclides in SS and marine biota have also been discussed. We also summarized recent studies concerning the hematological, morphological and genetic impacts of the accident on Fukushima wild monkeys, cows, birds, and
butterflies. Finally, we discussed the application of anthropogenic radionuclides (for example, $^{134}$Cs/$^{137}$Cs activity ratio and $^{240}$Pu/$^{239}$Pu atom ratio) as a tracer to study marine and soil processes and/or identify sources.

Could the atmospheric and oceanic dispersion of anthropogenic radionuclides provide a basis for climate change monitoring? This question should be considered by climate change scientists as data from the CTBTO global monitoring facilities may provide clues concerning mechanisms of global climate change.

Acknowledgements

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