Radioactive cesium-bearing particles in various environmental samples

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Radioactive materials released by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident caused extensive radioactive contamination. Adachi et al. (2013) discovered radioactive Cs-bearing particles (Cs-bearing particles) from air filter in Tsukuba (170 km from FDNPP). This finding is an important, since the particle which is likely to be emitted directly from FDNPP may contain various information on the phenomena occurring in FDNPP during the accident. However, because of the difficulty of separating Cs-bearing particle from environmental sample, comprehensive information on physical and chemical properties of the particles as well as distribution of particles in Fukushima contaminated areas is limited. In this study, the distribution and physical and chemical properties of Cs-containing particles were investigated using various types of environmental samples, such as suspended particles in river and surface seawater, tree leaves, and road dust (noted as black substances).
Radioactive cesium-bearing particle in the sample was separated from other particles by the wet-separation method using a NaI scintillation counter. The separated particle was identified by a scanning electron microscope (SEM) equipped with an energy dispersed X-ray spectrometer (EDS). The activities of ^{134}Cs and ^{137}Cs in the identified particle were measured by non-destructive gamma-ray spectrometry.
Spherical particles with diameters of approximately < 5 μm were found in any samples. Particles of this type are similar in terms of chemical composition to those reported so far, and were estimated to be derived from Units 2 or 3 of FDNPP estimated by the ^{134}Cs/^{137}Cs activity ratio. In the dust samples of the northwest direction within 20 km from FDNPP, particles with diameters of several tens to several hundreds of microns were found. This area has been reported to be contaminated with radioactive materials from Unit 1 (Satou et al., 2015). Most of the particles in this region were not uniform in shape. The main components of the particles in this region were Si, Ca, K, and Al derived from Unit 1 from the ^{134}Cs/^{137}Cs activity ratio. In addition, although it is reported that the shape of the particles in this region is not uniform, some spherical particles with a diameter larger than > 20 μm were also found.
From the facts above, it was found that three kinds of radioactive particles are widely present in various environmental samples.

キーワード：福島第一原発、セシウム含有粒子、環境試料、放射能

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Keywords: Fukushima Daiiti Nuclear Power Plant, Cs-bearing particle, Environmental sample, Radioactivity

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ダスト事象の際の放射性セシウム
Atmospheric radioCs in case of the local dust event

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Atmospheric radioCs seems suspended along with surface soil dust when the strong wind creates local dust event. Local surface contamination may have resulted in the enhancement of the radioCs concentration when the local dust event occurred. Although Ishizuka et al. (2017) made the radioCs resuspension computation scheme due to the dust uplift in the air by wind blow, the effect of the local dust event has not been evidenced nor confirmed in detail so far. Furthermore, though Kajino et al. (2016) has simulated yearly resuspension in 2013 due to the dust suspension and those from the forest, event-based case study has not been carried out. In this presentation, some of such events are analyzed in terms of the radioCs resuspension. The dust event occurred in March 2013 was analyzed for meteorology and temporal change in radioCs concentration observed at Meteorological Research Institute in Tsukuba. The simulated results of radioCs concentration in Tsukuba is compared with the observed results. These analysis would suggest qualitatively minor effect of the local dust events on the enhancement of the radioCs concentration.

キーワード：セシウム、風塵現象、濃度
Keywords: radioCs, local dust event, concentration

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福島県の里山に大気沈着した放射性セシウムの長期変動
Long-term Changes in the Distribution of Atmospherically Deposited Radioactive Cs in a Small Forest in Fukushima Prefecture

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2012 年より積雪期を除き 1 ヶ月もしくは 2 ヶ月毎（2015 年以降）に，浪江町南津島の山林でスギと落葉広葉樹の生葉，落葉，表層土壌，底砂の放射性Cs濃度を調査した．福島市ー浪江町間の走行サーベイでは，除染により空間線量率は急速に減衰したが，未除染の山林では物理的減衰と同程度であった．

2014 年以降，落葉広葉樹林では林床（落葉と表層土壌）で放射性Csは物理減衰以上に減少していないが，スギ林では生葉と落葉で減少し，表層土壌に蓄積した．2014 年までスギ落葉中放射性Csは降水による溶脱が顕著であった．2013 年春季には放射性Csはスギ林よりも広葉樹林で表層土壌から深層に移行していたが，2015 年冬季にはスギ林で深層への移行率が上回った．

小川では放射性Csは小粒径の底砂に蓄積しており，一部は浮遊砂として流出するが，表層土壌に対する比は広葉樹林で2013 年：0.54，2015 年：0.29，スギ林で 2013 年：1.4，2016 年：0.31 と下がっており，森林に保持されていることが分かった．しかし，春季にはスギ雄花の輸送による放射性Csの生活圏への流出が懸念された．

キーワード：森林、表層土壌、生葉、落葉、川砂、エッジ効果
Keywords: Forest, Surface soil, Fresh leaves/needles, Litter, Stream sand, Edge effect

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福島第一原子力発電所事故から5年後の樹種別の林床の有機堆積物および土壌の放射性セシウムの分布

Radioactive Cs distribution of litter and soil in forests in each kind tree for 5years after Fukushima Dai-ichi Nuclear Power Plant accident

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The Fukushima Dai-ichi Nuclear Power Plant fall-out contaminated the surrounded areas with high levels of radioactive materials in march 2011. We have been investigated based on soil samples as part of a cooperative pilot survey involving both government and academia since May 2011. The surveys detected widespread radioactive contamination throughout the forested areas in Fukushima prefectures, where approximately 71% of the land area is forested. And, there is an artificial forest more than 35% in Fukushima forest. Usually, artificial forest is made with a specific kind tree. They are processed to plant mushrooms and/or to build houses. Mushrooms are cultured on *Quercus serrata*. It’s a deciduous tree. It means that Quercus serrata had not put leaves in the accident. Another hand, houses are built of *Cryptomeria japonica*. It is an evergreen tree. It means that fall-out was contaminated canopy of *Cryptomeria japonica*. Radionuclides deposited in forested areas by either wet or dry processes encounter the canopy. Most radioactive Cs (>90%) deposited onto the canopy is intercepted and retained by tree leaves and branches and subsequently transferred to the forest floor as a result of weathering by rainwater and wind.
The fall-outed radioactive Cs contaminated any kind tree forest. It is important to know the distribution of radioactive Cs in each kind tree forest for decontamination of forests.
Therefor we studied to know radioactive Cs distribution of litter and soil in the forest floor in each kind tree from 2011 to 2016.
Forests were separated for *Pinus densiflora* forest (PDF), *Pinus densiflora* Mix forest (PD Mix), decicuous tree forest (DTF), *Cryptomeria japonica* forest (CJF) and other coniferous tree forest (OCF). Air dose rate measured 3 times in 2011, 2014 and 2016. The number of samples were each 26, 56, 153, 71 and 52 in 2011, each 7, 28, 88, 52and 33 in 2014, and each 7, 13, 24, 33 and 9 in 2016. The litter and soil radioactive Cs measured in 2011 and 2016.
Each kind tree forests air dose rate was positive correlated an initial deposition radioactive Cs. And, each forests air dose rate decreased with time. 2011-2014 air dose rate decrease was faster than 2014-2016 decrease in PDF, PD Mix, DTF and OCF. Another hand, 2014-2016 air dose rate decrease was faster than 2011-2014 decrease in CJF. It means that the timing of air dose rate decrease was delay in CJF. Each forests air dose rate decrease was faster than the physical decay of radioactive Cs. Another hand, each radioactive Cs inventory of litter and soil decrease was slower than the physical decay of radioactive Cs. CJF’s radioactive Cs inventory of litter and soil increased significantly faster than PDF’s and DTF’s. It means that each forest air dose rate was negative correlated radioactivity of the litter and soil from 2011 to 2016. The litter’s radioactive Cs inventory decreased faster than radioactive Cs physical decay in PDF, PD Mix, DTF and CJF. DTF’s rate of decrease was significantly faster than PD Mix’s and CJF’s. The soil’s radioactive Cs inventory decrease slower than radioactive Cs physical decay in PDF’s and OCF’s. And, the radioactive Cs of soil of PD Mix, DTF and CJF in 2016 were more than the radioactive Cs inventory of soil in 2016. And, CJF’s rate of radioactive Cs inventory of soil increased significantly higher

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than PDF’ s. It means that radioactive Cs distributed from litter to soil in each forest.
Therefore, air dose rate of forest was decreased faster than radioactive Cs physical decay from 2011 to 2016. And, litter’ s radioactive Cs inventory decreased from 2011 to 2016. Soil’ s radioactive Cs inventory increased from 2011 to 2016. It was thought that radioactive Cs distributed from litter to soil in every forest from 2011 to 2016.

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Temporal changes of Cs-137 concentration and its flux (dissolved fraction, suspended sediment, and coarse organic matter) at small Headwater Catchment in Fukushima after Fukushima Dai-ichi Nuclear Power Plant Accident

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Radiocesium migration from headwater forested catchment is important perception as output from the forest which is also input to the subsequent various land use and downstream rivers. In this study, dissolved Cs-137 concentration of stream water, subsurface water, suspended sediment (SS) and coarse organic matter (Org) were measured. Observations were conducted from 2011 at four headwater catchments in Yamakiya district, located 35 km northwest of Fukushima Dai-ichi Nuclear Power Plant (FDNPP).
Stream water discharge was monitored by combination of parshallflume and water level recorder. Stream water was sampled manually at steady state condition in 2-4 month interval. Also intense few hours interval sampling of stream water was conducted during rainfall events using automated water sampler. Suction lysimeters were installed for sampling the soil water. Boreholes were installed for the groundwater sampling. Stream water and groundwater samples were collected for 40 L each. All the water samples were filtered through 0.45 μm pore-size membrane. Water samples with less than few L were concentrated by evaporative concentration. Water samples with more than few L were concentrated using the ammonium molybdophosphate (AMP)/Cs compound method. The SS was sampled using time-integrated SS sampler. Turbidity sensor was set in the streambed. The output of the turbidity (mV) was converted to SS concentration (mg/L). A 15 mm mesh net was placed in the stream channel to trap Org sample carried by the stream flow. The SS and Org samples were dried at 105 degree in an oven for 24 h and then finely crushed. The radioactivity of the samples was measured using gamma-ray spectroscopy.
The fast decline of first phase and secondary comparatively gradual decline were observed in time series of dissolved Cs-137 concentration in stream water and therefore fitted to two-component exponential line. During the rainfall events, the concentration of dissolved Cs-137 in stream water increased temporarily. Also the declining trend of Cs-137 concentration in SS and Org were fitted to exponential line however, those components were started to collect from August 2012 and the fast declining phase were not observed therefor fitted to single-exponential line. After 5 years passed from the accident, the difference of declining trend between catchments are shown.
In conjunction with Cs-137 concentration results, Cs-137 discharge flux were calculated for three components (dissolved fraction, SS, and Org). As a result, it is shown that more than 96% of the Cs-137 discharge was due to the discharge by SS. The total discharge ratio of Cs-137 to its deposition amount at each catchment was ranged 0.002–0.3% per year.

キーワード：セシウム137、福島第一原子力発電所、源流域

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Keywords: Cs-137, FDNPP, headwater catchment

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Dynamic association of radioactive cesium between sediment and suspended sediment in river

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Abstract

After earthquake and tsunami of 11 March 2011,a lot of radionuclides ^{134}Cs and ^{137}Cs were released into environment due to Fukushima Dai-ichi Nuclear Power Plant(FDNPP) accident. For the safety of people and the support of decontamination, the Japanese Ministry of Education,Culture,Sports,Science and Technology(MEXT) has taken a long term monitoring system,in which about river sediment,surrounding environment and water quality,in areas such as Iwate,Miyagi and Fukushima Prefectures since August 2011.Meanwhile,7 sites were newly opened in Nakadori and Hamadori since July 2011,24 sites were opened since October 2012,to monitor the dynamics of water quality and suspended sediment.However due to previous researches,we can see the obviously tendency of declining radiocesium concentration in suspended sediment,which slowed down after one year from the accident.But we cannot conclude the same ideal by analyzing the sediment samples which was taken by MEXT during 2011-2012,owing to the low correlation and widely variation between time and concentration caused by the influences of particle size.So during this survey we reanalyzed the particle size of samples and correction the particle size effect on the concentration of ^{137}Cs.As a result,80% sites' declining rate and the correlationship between time and concentration were improved.On the other hand,we compared the decline rate between suspended sediment and river sediment in the same watershed,we find there are same tendencies and numerical correlations of radiocesium between suspend sediment and river sediment.

キーワード：放射性セシウム、底質、浮遊砂、持続性
Keywords: Radiocesium, Sediment, Suspended Sediment, Persistent

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福島県沖の堆積物と直上水中の放射性セシウム濃度分布
Distributions of radiocaesium activity in sediment and overlying water off the Fukushima

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After the accident at the Fukushima Dai-ichi Nuclear Power Station (FDNPS) happened in March 2011, large amounts of radionuclides including radiocaesium also released from the FDNPS into the terrestrial and marine environments. In marine environment, parts of particulate radiocaesium have transported in seawater and accumulated to seafloor. Then, radiocaesium in sediment have partly re-suspended as particulate form and re-eluted as dissolved form due to several factors such as bottom current and deformation. The characters of seafloor topography are more different in the area off the coast of northern and southern part of Fukushima Prefecture, dividing areas at the Onahama port (Mogi and Iwabuchi, 1961). Because the wave bases in fine and stormy weather are about 20 and 80 m, respectively (Saito et al., 1989), it seems that the area of shallower than 100 m is also affected by erosion and re-sedimentation near seafloor with ocean wave degree. Thus, it is necessary to elucidate interaction for radiocaesium between sediment and seawater close to seafloor with more stations in order to guess radiocaesium activity variation at long times. For example, in the case of collected bottom-layer water with the Conductivity-Temperature-Depth (CTD) system, it is very difficult to collect seawater close to sediment because it is careful not to touch CTD system seafloor. This study was aimed at elucidating the relationship for radioacesium activity concentration between sediment and trapped water on sediment collected using Multiple Corer, which is considered as overlying water.
Sediment samples were collected using a Multiple Corer during UM14-04 cruise in May 2014 at three stations: I01 (37˚14’ N, 141˚07’ E, water depth:60 m), I02 (37˚14’ N, 141˚13’ E, water depth:120 m) and C (36˚55’ N, 141˚20’ E, water depth:190 m).Overlying waters were collected using tube for 2 hours later from collected sediment. In laboratory, collected sediment sample are dried and overlying water samples were filtered through a 0.2-μm pore size filter and was concentrated by the ammonium phosphomolybdate (AMP) method (Aoyama and Hirose, 2008). The radiocaesium activity concentrations in each sediment and overlying water samples were measured by gamma-ray spectrometry using a high-purity Ge-detector and corrected to sampling date.
In overlying water, the dissolved ^{137}Cs activity concentrations (mBq/l) were 3.1-16 and the activity at I01, I02 and C in order from the higher. In the surface-layer sediments (core depth 0-3cm), the activity concentrations (Bq/kg-dry) were 8.4-286 and the high activities at I01 and I02 have characters of relatively high percentage for silt to clay particle compared to those at C. At I02 and C, the activity in overlying water were same value compared those in bottom-layer of seawater, which collected above water depth 10 m from seafloor. On the other hand, the activity in overlying water at I01 was five time higher than those in bottom water. The calculated K_d' (L/kg) of apparent distribution coefficient using ^{137}Cs activity concentrations in surface-layer sediment and overlying water were 8.8×10^2-1.5×10^4 and within rages of recommended K_d value of 2.0×10^3 for caesium by IAEA TRS422.

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キーワード：放射性セシウム、堆積物直上水　、堆積物、福島第一原発事故
Keywords: radiocaesium, overlying water, sediment, Fukushima Dai-ichi Nuclear Power Station accident

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2015年および2016年の北太平洋西部亜熱帯循環域における福島第一原子力発電所事故由来の放射性セシウムの分布
Fukushima-derived radiocesium in the western subtropical gyre of the North Pacific Ocean in 2015/2016

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2011年3月11日に発生した巨大地震とそれに引き続く大津波は、福島第一原子力発電所（FNPP1）の核燃料露出と炉心損傷を引き起こした。その結果、多くの放射性セシウム（^{134}Csと^{137}Cs）がFNPP1より漏えいし北太平洋に放出された。これまでの観測研究によって、日本近海の北太平洋に大気沈着および直接流出した放射性セシウムは北太平洋海流に沿って中緯度表層を東に移行しつつあることが明らかにされた（Kumamoto et al., 2016）。また、黒潮・黒潮続流の南側に大気沈着した放射性セシウムは亜熱帯モード水の亜表層への沈み込みによって、2014年末までに西部亜熱帯域のほぼ南端に相当する北緯15度まで南下したことが確認されている（Kumamoto et al., 2017）。一方、2011年から2015年の約4年余の間、北海道西部、新潟、石川、福井、島根、佐賀、鹿児島、愛媛、静岡県の各原子力発電所の沿岸域では、海水中放射性セシウムの継続な濃度上昇が確認されている（規制庁, 2016）。また、Aoyama et al.(2017)も2015/2016年に同沿岸海域における表面水中濃度の上昇を報告している。放射性セシウム濃度の上昇が観測された海域は黒潮系水の影響が比較的大きい沿岸海域であり、これらの結果はFNPP1事故で西部亜熱帯域全体に拡がった放射性セシウムが、時計回りの亜熱帯循環流によって日本沿岸に回帰していることを暗示している。しかしながら、西部亜熱帯循環域におけるFNPP1事故起源放射性セシウムの時空間変動は明らかではない。我々は2015年および2016年に黒潮・黒潮続流南側の西部亜熱帯域において、表面から深度約800mまでの海水中溶存放射性セシウムの濃度を測定したのでその結果を報告する。海水試料は、新青丸KS15-14(2015年10月)、白鳳丸KH16-03(2016年6月)、および「かいめい」KM16-08(2016年9月)の各観測航海において、バケツ及びニスキン採水器を用いて各10～20リットルを採取された。陸上の実験室（海洋研究開発機構むつ研究所）において硝酸酸性にした後、海水中の放射性セシウムをリンモリブデン酸アンモニウム共沈法によって濃縮し、ゲルマニウム半導体検出器を用いて放射性セシウムの濃度を測定した。濃縮前処理と測定を通じて得られた分析の不確かさは、約8%であった。北緯30-32度/東経144-147度で得られた^{134}Cs濃度（FNPP1事故時に放射壊変補正済）の鉛直分布を、同海域において2014年に得られたそれ（Kumamoto et al., 2017）と比較した。その結果、深度100m程度までの表面混合層においては、2014年には約1 Bq/m^{3}であった^{134}Cs濃度が、2015/2016年には1.5-2.5 Bq/m^{3}に増加したことが分かった。一方、深度300-400mの亜表層極大層におけるその濃度は、2014年から2016年の3回の観測を通じてほとんど変化していなかった（約3-4 Bq/m^3）。この^{134}Cs濃度の亜表層極大層は、亜熱帯モード水の密度層とよく一致していた。一方、同じく黒潮続流南側の北緯34度/東経147-150度における放射壊変補正済^{134}Cs濃度は、2012年から2014年の約3年間に、表面混合層では検出下限値以下（約0.1 Bq/m^3）から約1 Bq/m^{3}に増加し、亜表層の300-400mでは約16 Bq/m^{3}から約3-4 Bq/m^{3}に低下したことが報告されている（Kumamoto et al., 2017）。これらの観測結果は、FNPP1事故から5年以上が経過した2016年までに、亜熱帯モード水によって南方に輸送されたFNPP1事故起源の放射性セシウムが同モード水の時計回りの循環によって、日本南方の西部亜熱帯域北部に回帰してきたことを強く示唆している。その他の起源（例えば陸水）の影響が小さいと仮定できるならば、表面混合層における2012年から2016年の間の^{134}Cs濃度上昇（0.1 Bq/m^{3}以下から1.5-2.5 Bq/m^3）は、亜表層極大の高濃度水がentrainmentによって表面水に取り込まれたためと推測される。講演では、日本沿岸域の2015/2016年の観測結果速報も報告する予定である。この本研究はJSPS科研費24110004の助成を受けた。

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Estimate of spatial and temporal variation of radiocaesium amount derived FNPP1 accident in the North Pacific Ocean

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^{134}Cs and ^{137}Cs (radiocaesium) were released to the North Pacific Ocean by direct discharge and atmospheric deposition released from the TEPCO Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident in 2011. It is recognized that estimation of the total amount of released ^{134}Cs and ^{137}Cs is necessary to assess the radioecological impacts of their release on the environment. It was reported that the inventory of ^{134}Cs or ^{137}Cs on the North Pacific Ocean after the FNPP1 accident was 15.2-18.3 PBq based on the observations (Aoyama et al., 2016a), 15.3±1.6 PBq by OI analysis (Inomata et al., 2016), 16.1±1.64 PBq by global ocean model (Tsubono et al., 2016). These suggest that more than 75 % of the atmospheric-released radiocaesium (15.2-20.4 PBq; Aoyama et al., 2016a) were deposited on the North Pacific Ocean. It was revealed that these radiocaesium existed in the Subtropical Mode Water (STMW, Aoyama et al., 2016b; Kaeriyama et al., 2016) and Central Mode Water (CMW, Aoyama et al., 2016b), suggesting that mode water formation and subduction are efficient pathway for the transport of FNPP1 derived radiocaesium into the ocean interior within 1-year timescale. Kaeriyama et al. (2016) estimated the total amount of FNPP1 derived radiocaesium in the STMW was 4.2 ±1.1 PBq in October–November 2012. However, there is no estimation of the amount of radiocaesium in the CMW. Therefore, it is impossible to discuss about the mass balance of radiocaesium injected into the North Pacific Ocean. In this study, we conducted the optimum interpolation (OI) analysis to estimate the inventory of radiocaesium in the ocean interior as well as surface sweater by using the measured activities. Furthermore, transport speed of radiocaesium in the surface layer in the North Pacific Ocean were also estimated. The data used in this study were derived from all of the available data reported by such as the Tokyo Electric Power Company (TEPCO), the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT), and voluntary cargo ships. The data analysis period was until December 2015 after the FNPP1 accident. It was found that the radiocaesium across the North Pacific Ocean were reached to 180˚ E around 40˚ N latitude at July, 2012 by OI analysis. The transport speed was estimated to 8.5 cm s^{-1}. These were reached to the coastal site of America continent and the activities were increased after the year of 2014. The transport speed across 70˚ W (40˚N latitude) was decreased to 5.2 cm s^{-1}.
We estimated the inventory of radiocaesium in the surface seawater (depth; 0-100m) during the periods from August to December, 2012, based on the OI analysis. Amount of ^{134}Cs inventory was estimated to 4.7 PBq with decay-corrected to 1 October 2012 (7.9 PBq at the time on 11 March 2011). (In the case of ^{137}Cs, the inventory was estimated to 12.5 PBq with decay-corrected to 1 October 2012 and 13 PBq at the time on 11 March 2011 which includes pre-Fukushima ^{137}Cs). These correspond to 43-53% of the injected ^{134}Cs in the North Pacific Ocean. It was reported that the 4.2±1.1 PBq of ^{134}Cs were distributed in the STMW (Kaeriyama et al., 2016). Taking into account these estimation, FNPP1 derived radiocaesium existed in the CMW in the North Pacific Ocean would be about 3-6 PBq.

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