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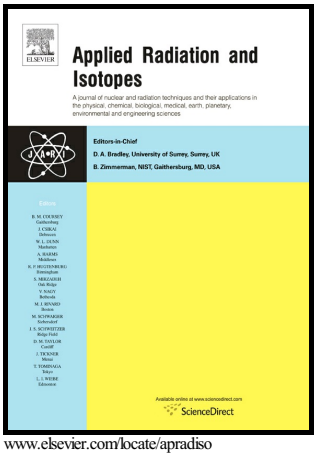

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No Fukushima Dai-ichi derived plutonium signal in marine sediments collected 1.5 - 57 km from the reactors

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Abstract

Based on AMS analysis, it is shown that no Pu signals from the Fukushima accident could be discerned in marine sediments collected 1.5 - 57 km away from the Fukushima Da-ichi power plant (FDNPP), which were clearly influenced by accident-derived radiocesium. The $^{240}Pu/^{239}Pu$ atom ratios (0.21 - 0.28) were significantly higher than terrestrial global fallout (0.182±0.005), but still in agreement with pre-FDNPP accident baseline data for Pu in near coastal seawaters influenced by global fallout and long-range transport of Pu from the Pacific Proving Grounds.

Key words: Nuclear accident, source identification, accelerator mass spectrometry, Fukushima, Pacific Proving Grounds

1 Introduction

Radionuclides released from the damaged Fukushima Dai-ichi Nuclear Power Plant (FDNPP) reactors and spent fuel repositories have caused widespread radioactive contamination in the environment. Volatile radionuclides have been detected at large distances from the accident site (e.g. Masson et al.

(2011)), while more severe radioactive contamination has been limited to a local scale (e.g. Steinhauser et al. (2014)). Plutonium with high $^{240}Pu/^{239}Pu$ atom ratios (0.303-0.330) has been found in debris collected in the terrestrial environment within 30 km of the FDNPP showing clear deviation from global fallout levels, indicating that Pu has been released during the accident (Zheng et al., 2012).

In contrast to the Japanese terrestrial environment, where pre Fukushima $^{240}Pu/^{239}Pu$ ratios were found to be in accordance with global fallout, nuclear weapon detonations at the Pacific Proving Grounds (PPG) have been linked to elevated Pu ratios in samples from off the Japanese coast as well as in most of the North Pacific region. The $^{240}Pu/^{239}Pu$ atom ratios in samples of riverine and marine sediments as well as seawater collected before the FDNPP accident were found to range within 0.17 and 0.32 (Oikawa et al., 2015; Tims et al., 2010; Zheng and Yamada, 2004). Seawater and sediment samples from this area will be influenced by global fallout and long-range transported debris from tests at the PPG and particularly those that took place at the Bikini and Eniwetok atolls during 1946 - 1958. A total of 66 nuclear weapon tests were conducted at the PPG, and in the early 1950s some very high yield detonations were conducted (Björklund and Goliath, 2009; Tims et al., 2010). Enhanced levels of the higher mass Pu isotopes were produced in the early detonations by multiple neutron captures in ^{238}U present in natural U-tamper materials (Diamond et al., 1960). Both contemporary analyses of fallout from these detonations, as well as analysis of Pu in dated coral bands from Guam, down-current of PPG have revealed $^{240}Pu/^{239}Pu$ atom ratios (0.05-0.46, weighted average 0.30±0.08) ranging widely (Lindahl et al., 2011). In addition to direct deposition as weapons fallout, remobilisation from contaminated sediments can act as an additional source of Pu to the North Pacific. This debris is transported westwards by the North Pacific Gyre, eventually leading into the Kuroshio Current passing Japan. Other potential sources of radioactive contamination in this area are global fallout, local and regional fallout from Former Soviet Union (FSU) and Chinese detonations and operational and accidental release from nuclear power plants and reprocessing activities in the region.

In the present study, activity concentrations and atom ratios of ^{239}Pu and ^{240}Pu have been determined in four seawater samples collected in May and September 2013 at distances between 5.7 and 110 km

from the damaged reactor, and in four marine sediment cores collected at distances 1.5 to 57 km away from the FDNPP. To the best of our knowledge, no Pu isotopic data from samples collected closer than 5 km to the FDNPP have so far been published.

Following the accidents, volatile (^{131}I) and less volatile (^{90}Sr) radionuclides released from the reactors and spent fuel repositories, deposited in terrestrial and aquatic ecosystems. In addition, run-off with rivers and from the site transported contaminants to the shore. The ^{240}Pu/^{239}Pu atom ratios observed terrestrially by for instance Yamamoto et al. (2014) and in river catchments by Evrard et al. (2014), indicates an influence from the FDNPP to the marine environment. Thus, the research question focuses on how far into the sea Pu atom signals different from that of pre-FDNPP accident can be observed.

2 Materials and methods

2.1 Sampling

Four seawater samples (May 2013) and four sediment cores (May and September 2013) were collected at varying distances from the Fukushima nuclear power plant during the May 2013 R/V Umitaka Maru and September 2013 Dai-san Kaiyo Maru cruises (Fig. 1, Supplementary Material 1 and 2).

2.2 Seawater

Approximately 25 l seawater samples were acidified by adding 1 ml concentrated HCl per litre of seawater. 35 pg of a ^{242}Pu tracer (National physical laboratory, Teddington Middlesex UK, TW11 0LW, Reference E05080352) was added as a yield monitor to each sample, before 8 g of $FeSO_4$ and 100 g $Na_2S_2O_4$ were added per 100 l of sample in order to change the speciation of Pu to Pu (IV). Thereafter the pH was raised to ~9 by careful addition of NaOH, initiating precipitation of $Fe(OH)_2$ and co-precipitation of the actinides. The final precipitate was transferred to PTFE ultraclave tubes and digested in concentrated HNO_3 in an ultraclave unit (Ultraclave 3, Milestone Ltd). The sample solutions were then diluted to 8M HNO_3 and subjected to extraction chromatography according to the procedure described by Wilcken (2006). U and Pu fractions were evaporated to dryness and the Pu

fraction taken up in 8 M HNO_3 and subjected to anion chromatography. Eluates of U and Pu from chromatography were evaporated and then taken up in concentrated HNO_3 and 2 mg Fe added as $FeSO_4$ and transferred to glass vials prior to evaporation to dryness.

2.3 Sediments

6-8 g of dried sediments were weighed directly into PTFE ultraclave tubes. 35 pg of a ^{242}Pu tracer (National physical laboratory, Teddington Middlesex UK, TW11 0LW, Reference E05080352) was added as a yield monitor to each sample before ~20 ml of HNO_3 (conc) was added, and the samples were left overnight for the acid to completely saturate the matrix. Pu and U were then leached from the samples under high temperature and pressure in an ultraclave unit (Ultraclave 3, Milestone Ltd). After cooling, the sample digests were filtered through glass fibre filters (Whatman GF/C) and the residue discarded. The filtrates were evaporated to dryness, taken up in 3M HNO_3 and subjected to ion exchange chromatography as described in Wilcken (2006).

2.4 Target preparation

Prior to target preparation the samples were then baked at 500 °C for 8 hours in a muffle furnace.

The samples were then scraped off from the internal surfaces of the glass vials, and mixed thoroughly with approximately the same mass of aluminium powder serving as a binder / conductor. The samples were then pressed into Al AMS sample holders.

2.5 Determination of ^{239}Pu and ^{240}Pu

Atom ratios of plutonium were determined by accelerator mass spectrometry (AMS) at the Australian National University (ANU) as described in (Fifield, 2008). Briefly, negative Pu molecular ions (PuO^-) are generated in the ion source; these are then pre-accelerated, mass-analyzed and transported towards the positive terminal of the tandem accelerator. The PuO^- molecules are then dissociated in a low pressure gas stripper, and electrons are removed from the Pu to form positive Pu ions which are further accelerated away from the positive terminal of the tandem accelerator. Pu^{5+} ions are selected by an analysing magnet and passed through a velocity filter (Wien filter) before reaching the gas-ionization detector. The detector identifies and counts the Pu ions. Switching between isotopes is effected by

changing the pre-injection mass-analysing magnet, the terminal voltage of the accelerator, and the Wien filter.

AMS has a high level of suppression against U interference. Any uranium-containing molecular ions (e.g. $^{238}U^{16}OH^-$ or $^{238}U^{17}O^-$) that are injected into the accelerator along with PuO^- are dissociated in the gas stripper in the high voltage terminal. The mass difference between the uranium and plutonium atomic ions then ensures that the uranium ions are rejected by the post-acceleration analyzing magnet.

^{239}Pu, ^{240}Pu and ^{242}Pu (yield monitor) were counted sequentially with counting times of 2, 3 and 1 minutes respectively. The counting sequence (^{242}Pu, ^{240}Pu, ^{239}Pu) was repeated twice with a third ^{242}Pu count at the end; ^{239}Pu and ^{240}Pu count rates were drift corrected based on the variations in ^{242}Pu count rates over the two sequences. ^{239}Pu and ^{240}Pu concentrations were then calculated from the $^{239}Pu/^{242}Pu$ and $^{240}Pu/^{239}Pu$ atom ratios obtained. A certified artificial reference material containing $^{239,240,242}Pu$ with accurately-known ratios close to unity (UKAEA certified reference material No: UK Pu5/92138) was analysed on several occasions for instrumental calibration and method validation purposes. A sample of IAEA 135 material, which is a marine sediment from the Irish Sea, was also processed and measured for validation of the complete method. The measured $^{239+240}Pu$ activity was 242±1.6 mBq/g, which is somewhat higher than the recommended value of 213 mBq, but well within the range of values measured for this material in an international intercomparison. We also routinely measure this material by ICP-MS, and again find an activity of ~240 mBq/g.

The reproducibility for the $^{239}Pu/^{242}Pu$ and $^{240}Pu/^{242}Pu$ atom ratios was 4.6% (relative standard deviation, based on 10 repeated measurements of UKAEA Pu5/92138 with at least 10,000 counts for each isotope per measurement). Analytical blanks spiked with 35 pg ^{242}Pu gave at most 1 count of ^{239}Pu and ^{240}Pu per 2 or 3 minutes counting interval respectively corresponding to a detection limit below 1 fg Pu.

3 Results and discussion

The ^{239}Pu concentrations in seawater are plotted in Fig.2, and the $^{239+240}Pu$ and $^{240}Pu/^{239}Pu$ atom ratio data are presented in Supplementary Material 2. For several of these samples, the number of ^{240}Pu

counts was very low, resulting in large uncertainties on the $^{240}Pu/^{239}Pu$ ratio. The low counts were a consequence of both low ion source output in the early part of the run when the seawater samples were measured, and lower average chemical yield of the seawater samples relative to the sediment samples as evidenced by lower counting rates of the ^{242}Pu spike. This affected all four of the A samples, F-23,24, and NP2-23,24, and the $^{239+240}Pu$ activities of these samples were determined from the ^{239}Pu activities and an assumed $^{240}Pu/^{239}Pu$ ratio of 0.24. For the remaining samples, the measured $^{240}Pu/^{239}Pu$ values were used. For bottom water samples at water depths between 400 and 1100m, Oikawa et al. consistently find $^{239+240}Pu$ concentrations of 10-30 mBq/m^3, and the result we obtain here for the bottom water at site F (F-17,18) of 25.5±1.8 mBq-m^3 is comfortably within this range. This lends support to our result. The large difference between our result and that obtained by Bu et al (J. Chromatography A, 1337 (2014)) of 1.44±0.19 mBq/m^3 is, however, more difficult to understand. As far as we can judge these are the exact same sampling points, and the 1251 m sampling should be the same as the Bu bottom -10 sampling. If, in addition the sampling was done at the same time, or within the same time frame, which is suspected, then there is a discrepancy for which we have no explanation. Nakanishi et al. report depth profiles of $^{239+240}Pu$ concentrations at a range of sites, and find values that range between 10 and 70 mBq/m^3 at depths down to at least 1000m, which again are consistent with the values found at site F in the present work. Our surface sample (F-23,24) at this site has a much lower concentration (1.5±0.8 mBq/m^3) than the deeper samples, which again is consistent with the findings of Oikawa et al., who report lower values for surface waters that range between 2 and 7 mBq/m^3.

Bu et al. (2015) report a value of 4.73±0.45 mBq/m^3 for water from site NP2. The values found here at the same site of 5.5±1.1 and 4.0±0.9 mBq/m^3 are consistent with this.

Results from the determination of Pu in sediment samples are given in Figs 3 and 4. The atom concentrations of ^{240}Pu and ^{239}Pu have been converted to inventories (Bq · m^{-2}) and summed to facilitate comparison with previous work. Sediment layer inventories of $^{239+240}Pu$ ranged within 2.9 and 14.0 Bq·m^{-2} (Fig. Figure 3 and Supplementary Material 1), and were comparable to levels observed in the same area prior to the FDNPP accident, e.g. (Wang and Yamada, 2005). Sediment cores collected south of the plant (12, 13 and 14) showed a high degree of variation in concentration

with depth, and in general, concentrations were increasing with depth. This is in agreement with sediment cores from the Sagami trough (Zheng and Yamada, 2004), and Yangtze river estuary (Tims et al., 2010). This tendency is also described in Lindahl et al. (2011) and Lindahl et al. (2012) who upon analysing dated coral layers found that Pu atom ratios and concentrations had peaked in 1952 and 1954, respectively at Ishigaki Island and 1953 and 1954, respectively at Guam. The cores analysed in the current work are relatively shallow (<20 cm) and it is possible that we have not been able to measure the peak concentration or atom ratio.

The sediment core sampled closest to the FDNPP (core 20), collected within 1.5 km of the damaged reactor at 14 m water depth, showed remarkably low variations in $^{239+240}$Pu concentrations (3.37 - 4.12 Bq · m^{2}). While having the highest surface contamination of ^{134}Cs and ^{137}Cs, this core also has the lowest variation in Cs concentrations through the core, when disregarding the upper 0-1 cm (Black and Buesseler, 2014). It can be assumed that the upper 0-1 cm contains sediment deposited during the first year following the accident, and is therefore likely to contain radionuclides deposited on land in the days and weeks following the accident or contain radionuclides released from the site directly to the sea. The homogeneity of the remaining core could be due to a disturbance in the sediment layer at this site caused by the passage and backwash of the tsunami waves. Investigation of the effects of the 2011 tsunami on the nearby Sendai bay indicated that erosion and re-deposition could affect seafloor topography at relatively large distances from the shoreline (Yoshikawa et al., 2015).

Sources of Pu

The ^{240}Pu/^{239}Pu atom ratio in the sediments (Figure) ranged from 0.21 to 0.28 and were within the range of baseline data for Pu in this region prior to the FDNPP accident, e.g. Zheng and Yamada (2004), Oikawa et al. (2015) and Tims et al. (2010). They also compare well with ^{240}Pu/^{239}Pu atom ratios (0.233 – 0.258) in sediments collected before and after the accident within the 30 km zone around the FDNPP site (Bu et al., 2014; Bu et al., 2015).

Prior to the FDNPP incident plutonium would have would have reached the marine environment of Japan through two routes. Firstly, atmospheric fallout from nuclear weapons detonation in the period 1945-1980 well characterized with ^{240}Pu/^{239}Pu atom ratios close to the 0.18±0.014 for the northern

hemisphere established by Kelley et al. (1999). Secondly, another important source is debris from the PPG where US high yield detonations were carried out in the period 1951-1962. $^{240}Pu/^{239}Pu$ atom ratios at the PPG are significantly higher than global fallout, and ratios as high as 0.46 have been reported following the tests at this site (Lindahl et al., 2011). Debris from the PPG sites is continuously remobilized and transported westwards with sea currents, and $^{240}Pu/^{239}Pu$ atom ratios have been found to be higher than global fallout in areas distant from the test sites (e.g. Lindahl et al. (2012). As a result of this influence, Pu atom ratios off the coast of Japan are relatively higher, and show greater variation reflecting the more complicated two source input, e.g. Zheng and Yamada (2004).

The $^{239+240}Pu/^{137}Cs$ activity ratio is a complicated measure in a situation where a historical source (global fallout and regional PPG-fallout) acts in combination with a more recent source (the FDNPP-accident). Whereas atmospheric nuclear detonations are unconfined high-temperature events leaving a well-mixed debris cloud, severe reactor accidents like the Chernobyl accident and the FDNPP accident give rise to a more heterogeneous release, where volatile radionuclides may have a different release and deposition regime than refractory elements. This was observed after the Chernobyl NPP accident in 1986, where fission products, in particular ^{137}Cs were widely distributed, while refractory elements like Pu isotopes had the strongest impact close to the accident site and were only sporadically observed elsewhere, e.g. Lindahl et al. (2004), Salminen-Paatero et al. (2012). In addition, the behaviour of the different radionuclides post deposition further complicates the situation. Nevertheless, we present the $^{239+240}Pu/^{137}Cs$ activity ratios based on the current work in combination with the results obtained by Black and Buesseler (2014).

The $^{239+240}Pu/^{137}Cs$ activity ratios were found to range within 0.00025 and 0.14 (Figure). The lowest ratio was found in the surface layer of core 20, which was sampled 1.5 km away from the FDNPP. Similarly, in surface sediments (north coastal and south coastal cores) down to 6 cm (Figure), ratios were found to be substantially lower than in pre Fukushima cores collected off the Japanese coast (Sagami Nada and Sagami trough) (Zheng and Yamada, 2004). In addition, all sample segments except the deepest segments of core 12 and 13 display $^{239+240}Pu/^{137}Cs$ ratios significantly lower (>2σ)

than the predicted (^{137}Cs-activity concentrations decay corrected to 2012 assuming peak deposition in 1962) global fallout ratio of 0.036 (Tims et al., 2010; UNSCEAR, 2000).

The $^{239+240}$Pu/^{137}Cs activity ratios were found to be affected most strongly by the concentration of Cs isotopes, (Supplemental Material 3), indicating a high deposition of radiocesium following the Fukushima accident that overwhelmed the historical deposition from nuclear weapons testing.

4 Conclusions

The activity concentrations of ^{239}Pu in seawater collected in May 2013 (distance 5.7 – 110 km from the FDNPP) were comparable to data obtained prior to the FDNPP accident.

The present ^{240}Pu/^{239}Pu ratios determined in sediments collected in May and September 2013 varied between 0.21 and 0.28, suggesting that the majority of plutonium in the investigated area originates from global fallout (^{240}Pu/^{239}Pu atom ratio of 0.182±0.005) and the Pacific Proving Grounds derived Pu (^{240}Pu/^{239}Pu atom ratios up to 0.46). Although ^{134}Cs and ^{134}Cs/^{137}Cs activity ratios reflecting releases from spent fuel have been observed in the present sediment cores, no plutonium FDNPP accident signal could be observed in these sediments collected about 1.5 km from the site. These findings are, however, in agreement with the assumption that the amounts and mobility of Pu released from the damaged fuel should be rather limited.

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Figure captions:

Figure 1. Seawater (NP2, A, N3, F) and sediment (20, 13, 14, 12) sampling stations (red markers) in the present work at distances of 1.5 - 110 km away from FDNPP (indicated with yellow marker).

Figure 2. ^{239}Pu activity concentrations (mBq · m^{-3}) in seawater samples collected off the coast of Japan. Sampling stations were 56 km (station A), 110 km (station F), 100 km (station N3) and 5.7 km (station NP2) away from the FDNPP, respectively.

Figure 3. Inventories of $^{239+240}$Pu (Bq · m^{-2}) in the sediment cores.

Figure 4. ^{240}Pu/^{239}Pu atom ratios in sediment cores from the investigated area outside FDNPP. Error bars are 2σ instrumental uncertainties. The shaded area indicate pre-Fukushima accident variation in sediment cores from the Sagami Bay as reported by Zheng and Yamada (2004).

Figure 5. $^{239+240}$Pu/^{137}Cs activity ratios in sediment cores from the investigated area outside FDNPP. Activity concentrations (reference date April 6th, 2011) of ^{137}Cs are from Black and Buesseler (2014). Crosses: core 12, solid squares: core 13, open squares: core 14, solid triangles: core 20. Uncertainty bars are within the symbols. The shaded areas indicate pre-Fukushima range in sediment cores from the Yangtze river estuary (light grey area in Tims et al. (2010)), and Sagami Bay (dark grey area in Zheng and Yamada (2004)).

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Figures:

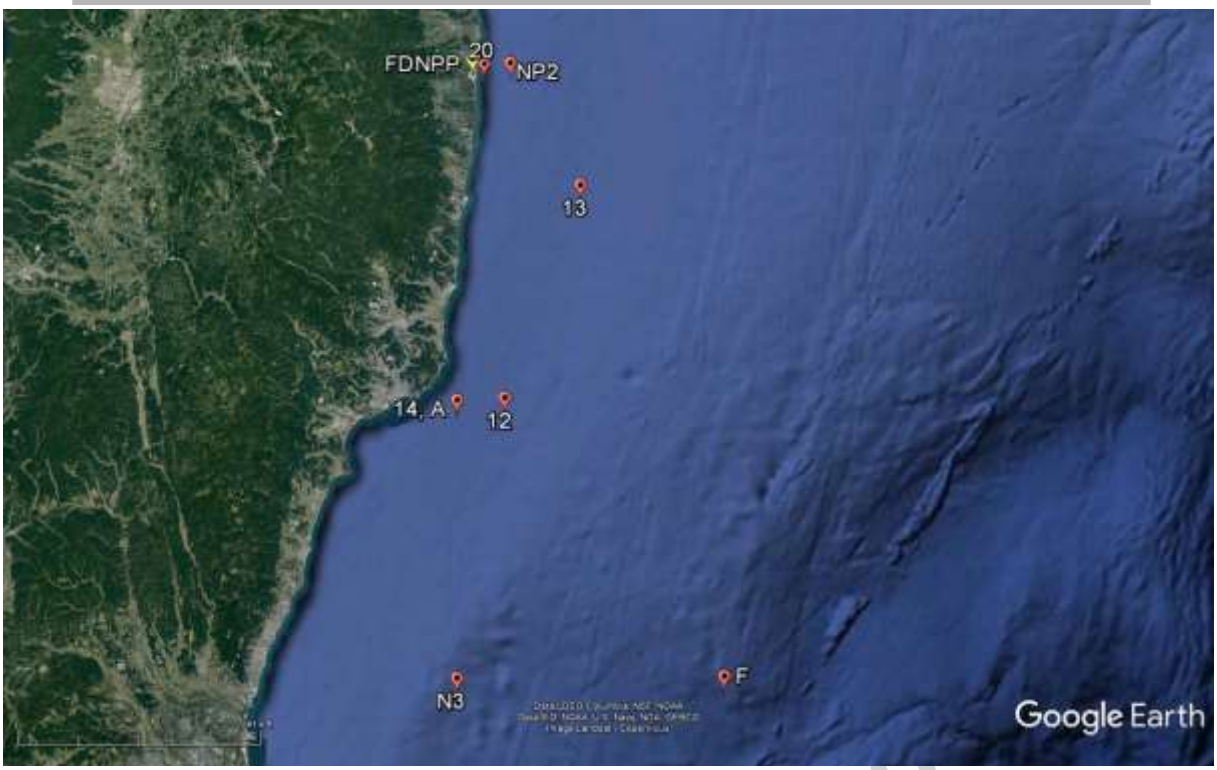


Figure 2. Seawater (NP2, A, N3, F) and sediment (20, 13, 14, 12) sampling stations (red markers) in the present work at distances of 1.5 - 110 km away from FDNPP (indicated with yellow marker).

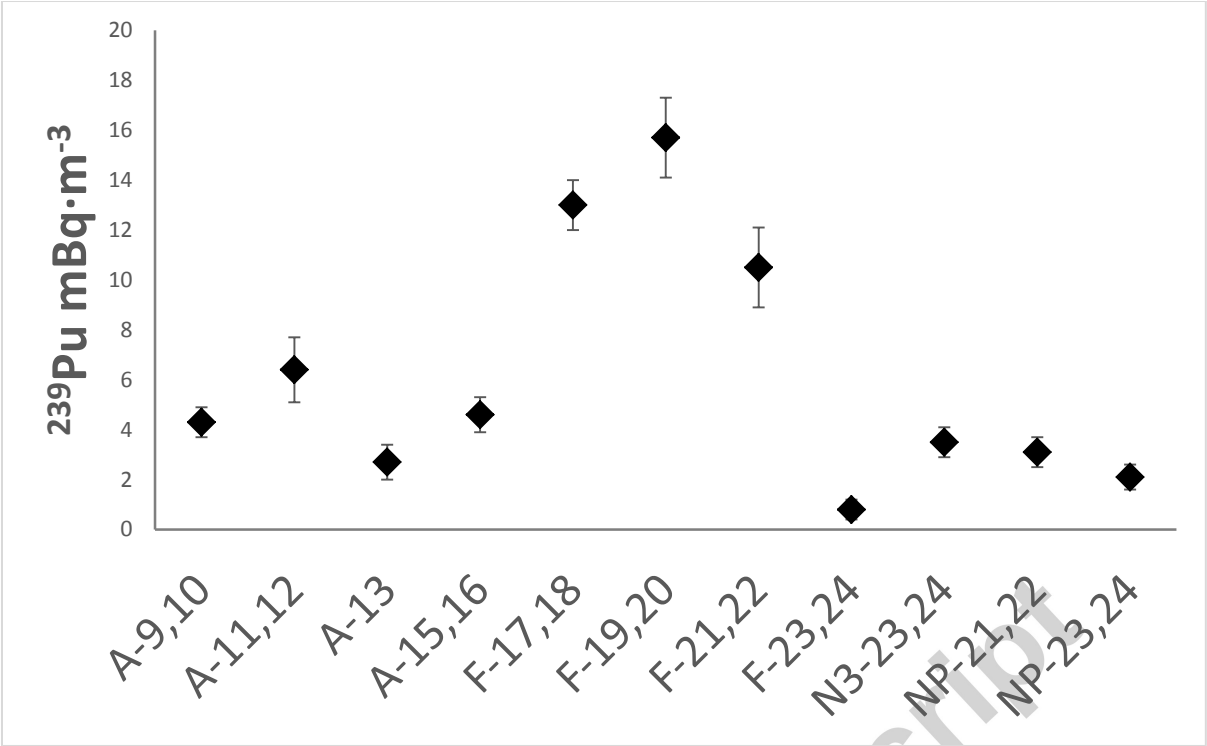


Figure 2. ^{239}Pu activity concentrations (mBq · m^{-3}) in seawater samples collected off the coast of Japan. Sampling stations were 56 km (station A), 110 km (station F), 100 km (station N3) and 5.7 km (station NP2) away from the FDNPP, respectively.

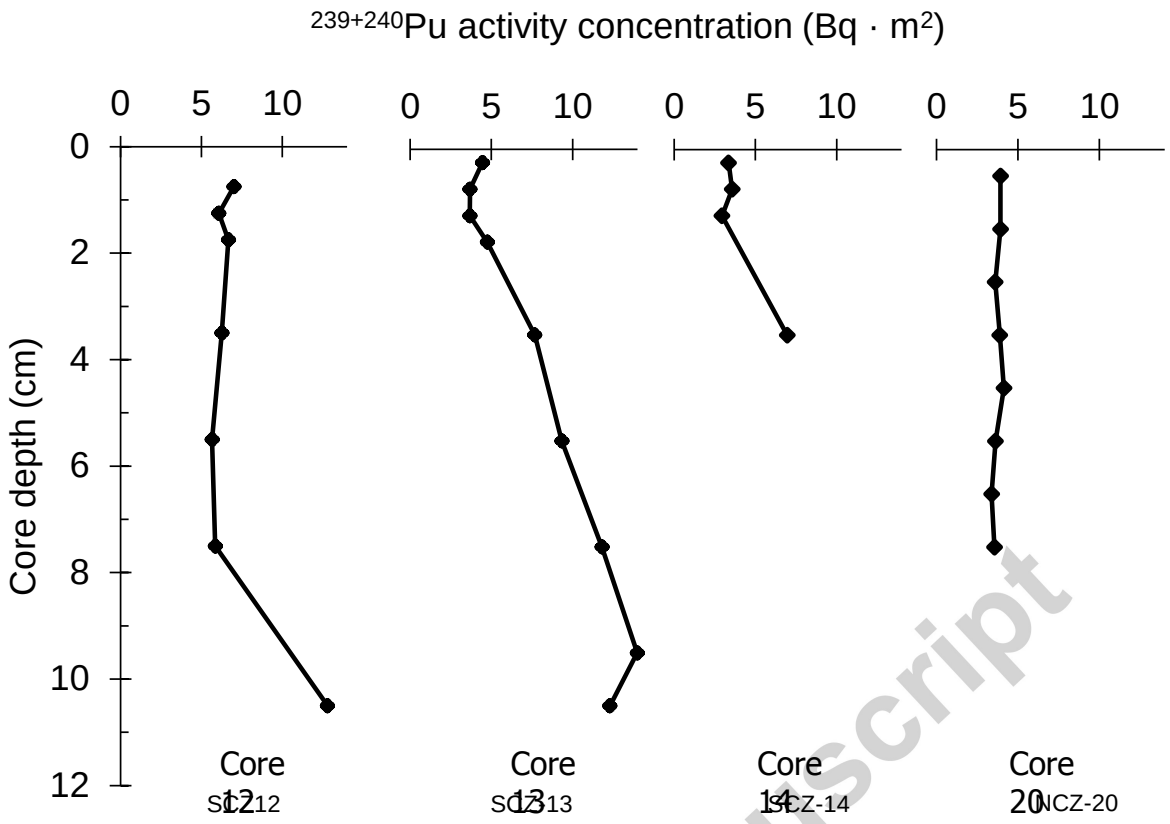


Figure 3. Inventories of $^{239+240}$Pu (Bq · m^{-2}) in the sediment cores.

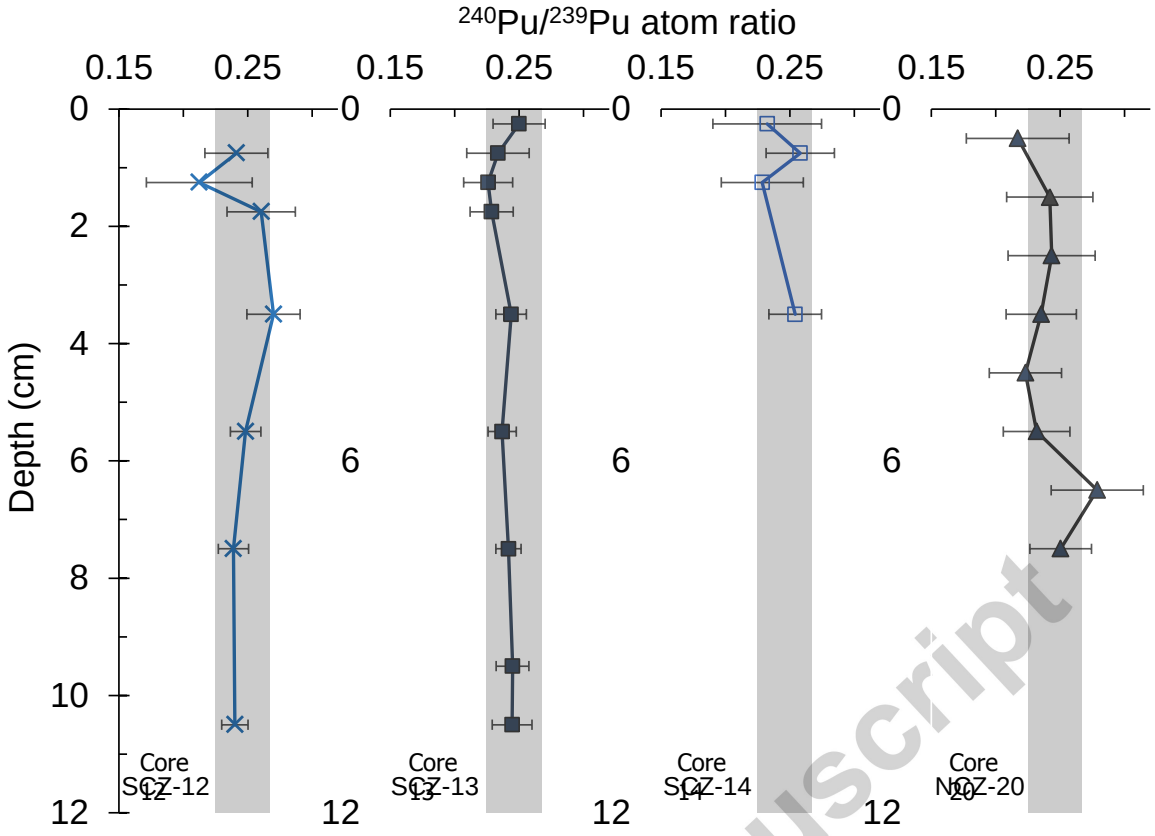


Figure 4. $^{240}Pu/^{239}Pu$ atom ratios in sediment cores from the investigated area outside FDNPP. Error bars are 2σ instrumental uncertainties. The shaded area indicate pre-Fukushima variation in sediment cores from the Sagami Bay as reported by Zheng and Yamada (2004).

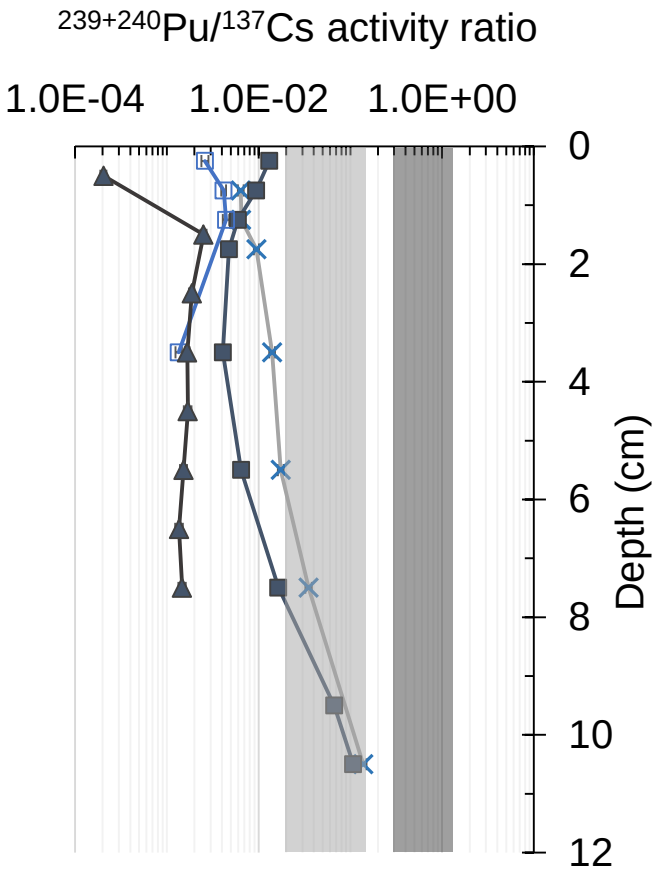
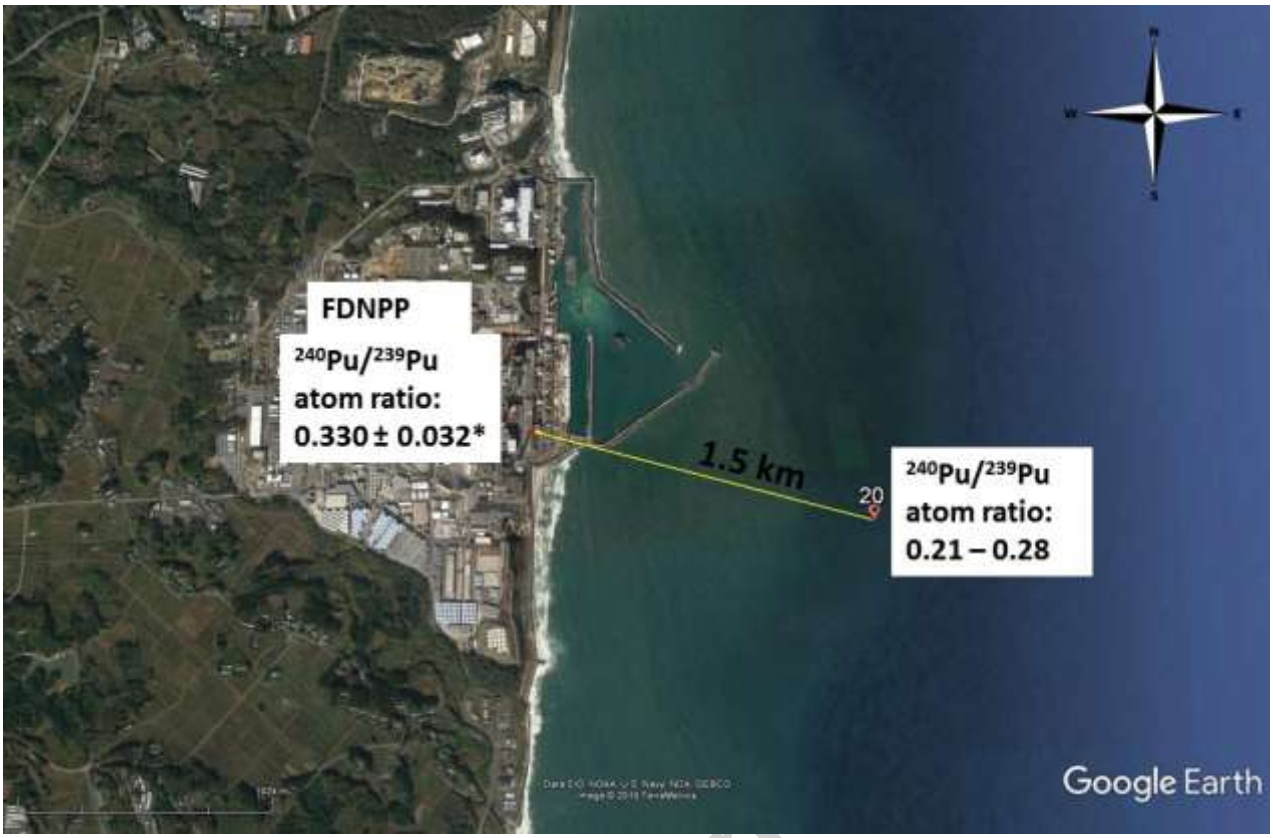


Figure 5. $^{239+240}Pu/^{137}Cs$ activity ratios in sediment cores from the investigated area outside FDNPP. Activity concentrations of ^{137}Cs are from Black and Buesseler (2014). Crosses: core 12, solid squares: core 13, open squares: core 14, solid triangles: core 20. Uncertainty bars are within the symbols. The shaded areas indicate pre-Fukushima range in sediment cores from the Yangtze river estuary (light grey area in Tims et al. (2010)), and Sagami Bay (dark grey area in Zheng and Yamada (2004)).



Graphical abstract

Highlights

- AMS of Pu concentrations and atom ratios in marine samples near Fukushima NPP
- No Pu from Fukushima accident discerned in sediments near (1.5 km) the plant
- Main source of Pu in sediments from global fallout and Pacific Proving Grounds
- Low $^{239+240}Pu/^{137}Cs$ activity ratios in sediments show Fukushima accident influence