Plutonium release from Fukushima Daiichi fosters the need for more detailed investigations

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The contamination of Japan after the Fukushima accident has been investigated mainly for volatile fission products, but only sparsely for actinides such as plutonium. Only small releases of actinides were estimated in Fukushima. Plutonium is still omnipresent in the environment from previous atmospheric nuclear weapon tests. We investigated soil and plants sampled at different hot spots in Japan, searching for reactor-borne plutonium using its isotopic ratio $^{240}\text{Pu}/^{239}\text{Pu}$. By using accelerator mass spectrometry, we clearly demonstrated the release of Pu from the Fukushima Daiichi power plant: While most samples contained only the radionuclide signature of fallout plutonium, there is at least one vegetation sample whose isotope ratio ($0.381 \pm 0.046$) evidences that the Pu originates from a nuclear reactor ($^{239}\text{Pu}/^{240}\text{Pu}$ activity concentration 0.49 Bq/kg). Plutonium content and isotope ratios differ considerably even for very close sampling locations, e.g. the soil and the plants growing on it. This strong localization indicates a particulate Pu release, which is of high radiological risk if incorporated.

The accident at the Fukushima Daiichi nuclear power plant (FDNPP) in 2011 caused massive contaminations of the Japanese land surface and the Pacific Ocean with mainly volatile fission products. In the course of the venting operations large amounts of radionuclides escaped from the reactor pressure vessels. The release was enhanced by three massive hydrogen explosions that occurred in Units 1, 3 and 4. In the course of the accident, large areas in Japan were contaminated with $^{131}\text{I}$, $^{132}\text{Te}$, $^{134}\text{Cs}$, $^{137}\text{Cs}$ and other volatile radionuclides. Numerous studies revealed these airborne radionuclides in the entire northern hemisphere. Radioisotopes of refractory elements such as strontium, barium, and lanthanides were monitored only in rare occasions. Actinides, however, proved to be the most severely understudied group of elements, despite the fact that many of them are long-lived α-emitters: most importantly $^{239}\text{Pu}$ (half-life 24,110 y) and $^{240}\text{Pu}$ (half-life 6,561 y). Schwantes et al. estimated releases of only 0.002% ($\pm 0.003\%$) of the overall plutonium inventory ($>5.6$ kg in units 1 and 3) into the environment. Recent analyses by Zheng et al. indicated even lower plutonium releases of only 0.00002% of the core inventories. Only few studies targeted the monitoring of actinides (most importantly plutonium) in environmental media (including ocean and river water). Most profound evidence for atmospheric releases of plutonium from Fukushima was presented in the study by Zheng et al.

Since the atmospheric nuclear weapon tests of the 20th century, plutonium has become a ubiquitous element in the environment. However, using the atomic ratio $^{240}\text{Pu}/^{239}\text{Pu}$ as an isotopic signature, it is possible to distinguish between the weapons’ fallout ($^{240}\text{Pu}/^{239}\text{Pu}$ ca. 0.18) and plutonium releases from a nuclear reactor ($^{240}\text{Pu}/^{239}\text{Pu}$ ca. 0.4–0.6). Due to the nuclides’ very similar α-particle energies, radioanalytical methods can usually not be used to distinguish the isotopic ratios between $^{239}\text{Pu}$ and $^{240}\text{Pu}$. Instead, low-ionization MS techniques or accelerator mass spectrometry (AMS) have established as powerful alternatives. AMS is currently regarded as the most sensitive method for the determination of the isotopic composition of environmental plutonium.

Previous indications for the release of actinides from Fukushima, in particular environmental abundance of $^{239}\text{Np}$ (the parent nuclide of $^{239}\text{Pu}$), were presented by Shozugawa et al. In that study, $^{239}\text{Np}$ was detected using gamma spectrometry. Spectral overlap of the most intense peak of $^{239}\text{Np}$ with the fission product $^{129}\text{Te}$, however, made it necessary to investigate both soil and vegetation samples from selected hot spots in the present study using AMS and to search for the isotopic plutonium-signatures at these spots.
Results
Soil and vegetation samples taken from several Japanese hot spots in 2011 (Fig. 1 and Table 1) were investigated with special focus on anthropogenic actinides, in particular plutonium.

The α-spectrometric and AMS results with respect to the atom ratios $^{240}\text{Pu}/^{239}\text{Pu}$ as well as detectable activity concentrations of $^{240}\text{Pu}$ and $^{240}\text{Pu}$ of soil and plant samples are listed in Table 2. Just in a few samples, plutonium could be detected using α-spectrometry. Also, for most samples, AMS revealed no detectable amounts of plutonium. For signals below the decision threshold (according to ISO 11929) only an upper limit for the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio is given. If $^{239}\text{Pu}$ was above the decision threshold, but $^{240}\text{Pu}$ was not, an upper limit for the atomic ratio is given. If neither of the isotopes could be detected by AMS the respective table entry reads “n.d” (not detected). The small amounts available did not allow for splitting of the samples and hence a $^{242}\text{Pu}$ spike concentration was added forming a compromise between high alpha-activity required by alpha spectroscopy and low atom-concentration desirable for AMS measurements. Hence, AMS results of the present work suffer from a very high background count rate.

Soil sample H-S2 revealed a $^{240}\text{Pu}/^{239}\text{Pu}$ ratio and AMS signal strength characteristic for global fallout; hence no quantification was undertaken for this sample. However, vegetation sample, A-V revealed a significantly higher $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio (0.38 ± 0.046). With a much higher analytical uncertainty, sample G-V also may carry an isotopic signature (0.64 ± 0.37) that is higher than the omnipresent fallout from the nuclear weapons tests. Such high ratios are characteristic for nuclear reactor plutonium and hence evidence the release of small but detectable amounts of plutonium from the damaged reactors of FDNPP.

Discussion
Of 20 samples investigated in this study, 17 did not exceed the detection limit for plutonium. One soil sample was contaminated only by global fallout plutonium with its characteristic isotopic ratio of $^{240}\text{Pu}/^{239}\text{Pu} < 0.2$. However, at least one (A-V) or two (G-V; higher uncertainty) of the vegetation samples showed detectable amounts of reactor derived plutonium (isotopic ratio $^{240}\text{Pu}/^{239}\text{Pu} > 0.2$). One can assume that the ubiquitous fallout plutonium background masked the minute contribution of Fukushima-derived plutonium in soil, as illustrated by Zheng et al.11 However, given the low mobility and bioavailability of plutonium, one can expect that plant uptake of fallout plutonium will be negligible. Consequently, dry or wet deposition of airborne plutonium on the surface of the plants will be highly visible for sensitive analytical techniques such as AMS. This probably explains why plants proved to be such suitable bioindicators for airborne plutonium from Fukushima in the present study.

It is remarkable to note that distance alone is no sufficient factor to estimate the findings of refractory elements such as plutonium. Although the vegetation sample taken closest to the reactors (~0.9 km away) exhibited detectable amounts of reactor-plutonium, no other sample in close vicinity of the reactors (1.5, 1.9, … km away) did so. However, a plant sample as far as 16 km away in northwestern direction (G-V) is suspected to contain plutonium from Fukushima. If this observation was confirmed, it would indicate a very nonuniform distribution of plutonium, most probably in particular form. This may also have health physical implications because the inhalation of such plutonium-rich particles may result in high local dose delivery to the lung tissue.

There is common agreement among the scientific community that only minute amounts of actinides have been released in the course of the Fukushima nuclear accident. Schwantes et al.7 calculated the average activity inventories of Units 1 and 3, from which a $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio of 0.441 can be derived. A similar value can be derived from the activity inventory data published by Kirchner et al.20, in particular 0.393. Sakaguchi et al.21 estimated the ratio to more than 0.4. All these predicted, calculated or estimated values are in good agreement with the ratio observed in sample A-V (0.381 ± 0.046) and also within the analytical uncertainty observed in G-V (0.64 ± 0.37). Zheng et al. observed somewhat lower values for soil and litter11 (between 0.30 and 0.33) and concluded that mixing with global fallout plutonium shifted the ratio to lower numbers. However, the actual composition of a particle also can vary from average activity inventory of an entire reactor, depending on its nuclear “history”22. Also some deviations between uranium-operated Unit 1 and MOX-operated Unit 3 can be expected. Sakaguchi et al. found an increased $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio (0.308 ± 0.176) in water from Abukuma River, which fostered the need for further analysis due to relatively large analytical uncertainties.

In any case, the sector field ICP-MS used by Zheng et al. proved to produce results for the isotopic ratio that are quite comparable with AMS of this study. The absolute activity concentrations for $^{239}\text{Pu}/^{240}\text{Pu}$ are also comparable to previously published data11,13, only slightly higher. These values, however, are still within the range of plutonium concentrations found before the Fukushima nuclear accident (0.15–4.31 mBq/g)11,22. The sum activity of $^{240}\text{Pu}/^{240}\text{Pu}$ alone, therefore, provides no sufficient evidence for plutonium releases from FDNPP.

In this context, it is interesting to note that only vegetation samples but none of the soil samples exhibited detectable traces of reactor plutonium. Since the Fukushima nuclear accident seemingly did not significantly increase the environmental plutonium inventory, one can hypothesize that the global plutonium background in soil dilutes the isotopic ratio and blurs the characteristic isotopic fingerprint of much smaller amounts of freshly deposited plutonium particles. Given the low environmental mobility and bioavailability of plutonium, vegetation collects airborne particles on its surface with a negligible background due to “old” plutonium uptake. Future findings of plutonium particles on plant material are increasingly unlikely, because not only atmospheric releases of actinides have ceased since 2011, but also rainfall will have washed down particles from the plant surface or vegetation cycles will probably have renewed the exposed leaf surface. In any case, the plutonium activities found in the vegetation samples (grass, leaves) sampled in late 2011 were (already?) so low that, assuming they were agricultural plants, the plutonium activity concentrations would not have exceeded any of the early regulatory limits for α-emitting radionuclides (actinides) in food13.

The results of this study confirm the very low release of refractory elements from the Fukushima reactors. The plutonium concentrations found herein and reported by Zheng et al.13 are partly more than three orders of magnitude lower than the values obtained in

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Figure 1 | Locations of Fukushima Daiichi nuclear power plant (FDNPP) and samples of this study using the codes of Table 1.
environmental samples around the Chernobyl site after 1986. This is also true for the semi-volatile radionuclide 90Sr that has been monitored only occasionally after the Fukushima accident and revealed relatively low activity concentrations in environmental samples in Japan. As described in a previous study, a vegetation sample from spot G (a spot that is also suspected to be contaminated with reactor plutonium according to Table 2) also carried a comparatively high 90Sr contamination, but a rather low radiocesium activity concentration (activity ratio 90Sr/137Cs approximately 0.1). This 90Sr over 137Cs activity ratio was found to be much smaller with all the other samples investigated in the previous study. Spot G is located outside the main "contamination strip" that goes from the reactor in northwestern direction, which makes the presence of reactor plutonium even more unexpected. If the findings of reactor plutonium at spot G are confirmed, one may speculate what the reason for this unusual radionuclide pattern is (high concentrations of refractory radionuclides, but relatively low in volatile radionuclides). One possible explanation could be that this spot was contaminated with fuel particles that have experienced temperatures high enough to volatilize most of their radiocesium content, before or while they were emitted from the reactors. However, this hypothesis needs further investigations. In any case, it seems that there is not necessarily a correlation between the levels of radiocesium (and other volatile radionuclides) and the radiocesium content, before or while they were emitted from the reactors. However, this hypothesis needs further investigations. In any case, it seems that there is not necessarily a correlation between the levels of radiocesium (and other volatile radionuclides) and the presence of reactor plutonium in the environment. This observation makes it likely that the release of plutonium was a more singular event, whereas the volatile radionuclides were released from the pressure vessels over several days in the course of the early venting operations.

In summary, our study evidenced the release of plutonium from the damaged FDNPP via its isotopic fingerprint. Two vegetation samples exhibited 240Pu/239Pu isotopic ratios of 0.81 ± 0.046 and 0.64 ± 0.37, respectively, both of which are higher than the global fallout background. The 239,240Pu activity concentrations, however, were relatively low (0.49 and 0.17 Bq·kg⁻¹, respectively), confirming early predictions of a low plutonium release from Fukushima. The fact that reactor plutonium has not been found in more than two samples (one of which remains more in question) indicates that plutonium releases and fallout from FDNPP occurred in the form of particulates causing nonuniform plutonium contaminations. Future investigations will aim at a comprehensive screening for plutonium containing fuel particles in larger samples and if applicable detailed investigation of particles by single particle analytical techniques. Detection limits and decision thresholds will be lowered by use of high purity spikes. Furthermore, a 240Pu spike can be used if no alpha measurements are performed on the same sample. This can further improve the performance of AMS on such environmental samples.

If confirmed, a release of plutonium-rich hot particles is of potential health concern upon inhalation or incorporation. Our findings demonstrate the need for more detailed investigations on plutonium distribution and speciation in order to assess potential radiological consequences for the public. In any case, our study supports previous findings that indicated that the environmental plutonium inventory in Japan has not significantly increased after the Fukushima nuclear accident.

**Methods**

**Sampling.** Samples were taken between October and December 2011 from several sites on the eastern Japanese coast (Fig. 1). For the sampling in restricted areas permission was obtained from the mayor of Okuma town. Some of the spots have already been investigated previously with respect to short-lived c-emitting radionuclides19 and 90Sr6. On most spots two types of samples were taken: top soil and vegetation (leaves, grass, conifer needles etc.). The sampling sites included highly contaminated spots such as the gate of the FDNPP (Fukushima I), locations in the close vicinity of the NPP including Fukushima Daini NPP (Fukushima II), but also more remote areas such as Yokohama or Kashio (some 200 km south of FDNPP). Exact locations and sampling dates are given in Table 1.

**Sample preparation and measurement.** All samples were dried at 105°C. All given specific-activities and masses are referring to the dry mass of the respective sample. Soil was available in relatively low quantities, ranging typically between 1–2 g, with the exception of the samples H-S1 and H-S2, which were available in greater amounts. Hence, we refrained from separating the soil skeleton to avoid additional sample losses. The samples were ashed with a low heating-up rate over 103 h up to 450°C. The ash was spiked with about 30 mBq 239Pu and digested using concentrated nitric acid and concentrated hydrofluoric acid. The dry residue was then taken up in a solution of 1 M aluminium nitrate and 3 M nitric acid and filtered. The chemical separation of plutonium was performed by extraction chromatography using Eichrom® TEVA resin15. After the separation the sample was transferred to a stainless steel plate via electrodeposition3,14. The electrodeposition took 2 h at a constant current of 950 mA. After alpha spectrometry had been conducted, the electrodeposited material was re-dissolved in hot nitric acid (3 M) from the alpha-plate for AMS measurement. To optimize the separation from uranium, a second separation step was applied. Plutonium was subsequently co-precipitated by ferric hydroxide and converted into ferric oxide by heating up to 800°C. Finally, the sample was mixed with aluminum powder in a ratio of 1:1 and pressed into an AMS target holder.

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**Table 1 | Sample descriptions and locations**

| Type       | Sample code | Location                  | Coordinates     | Distance from FDNPP in km | Sampling date |
|------------|-------------|---------------------------|-----------------|---------------------------|---------------|
| Soil       | AS          | FDNPP Main gate           | 37°25′02″N 141°01′29″E | 0.88                     | 2011-12-21    |
|            | BS          | 1.5 km from FDNPP         | 37°25′04″N 141°01′01″E | 1.5                      | 2011-12-21    |
|            | CS          | 1.9 km from FDNPP         | 37°25′03″N 141°00′44″E | 1.89                     | 2011-12-21    |
|            | DS          | 4.3 km from FDNPP         | 37°23′19″N 141°00′30″E | 4.25                     | 2011-12-21    |
|            | ES          | Chimyo-ji temple          | 37°29′45″N 141°00′05″E | 8.7                      | 2011-12-21    |
|            | FS          | Fukushima Daini NPP       | 37°20′31″N 141°00′51″E | 11.96                    | 2011-12-21    |
|            | GS1         | Odaka, Minamisoma         | 37°33′57″N 141°59′31″E | 16.42                    | 2011-12-21    |
|            | GS2         | Odaka, Minamisoma         | 37°33′57″N 141°59′31″E | 16.42                    | 2011-12-21    |
|            | HS1         | Minamisoma City           | 37°31′40″N 140°57″E | 30                      | 2011-12-21    |
|            | HS2         | Minamisoma City           | 37°38′14″N 140°57″E | 17.09                    | 2011-10-31    |
|            | IS          | Kashiwa                   | 35°52′48″N 139°59′09″E | 195                      | 2011-10-26    |
|            | JS1         | Yokohama                  | 35°32′07″N 139°38′03″E | 244                      | 2011-07-25    |
|            | JS2         | Yokohama                  | 35°32′07″N 139°38′03″E | 244                      | 2011-07-25    |
| Vegetation | AV          | FDNPP Main gate           | 37°25′02″N 141°01′29″E | 0.88                     | 2011-12-21    |
|            | BV          | 1.5 km from FDNPP         | 37°25′04″N 141°01′01″E | 1.5                      | 2011-12-21    |
|            | CV          | 1.9 km from FDNPP         | 37°25′03″N 141°00′44″E | 1.89                     | 2011-12-21    |
|            | DV          | 4.3 km from FDNPP         | 37°23′19″N 141°00′30″E | 4.25                     | 2011-12-21    |
|            | EV          | Chimyo-ji temple          | 37°29′45″N 141°00′05″E | 8.7                      | 2011-12-21    |
|            | FV          | Fukushima Daini NPP       | 37°20′31″N 141°00′51″E | 11.96                    | 2011-12-21    |
|            | GV          | Odaka, Minamisoma         | 37°33′57″N 141°59′31″E | 16.42                    | 2011-12-21    |
Basically the same procedure was applied for plant samples with some slight changes. The ashing procedure was performed with a heating program over 20 h up to 600 °C. We could not perform alpha-spectrometric measurements, due to the very low sample quantities of 26–230 mg of the sample ash (this fact is referred to as “N/A” (not applicable) in Table 2). For the same reason reduced amounts of only 5 mBq 240Pu were spiked. Furthermore, the digestion procedure was changed to microwave-pressure-digestion using concentrated nitric (8 mL), hydrochloric (5 mL), and hydrofluoric acid (2 mL) at 210 °C. For the AMS-target, the aluminum powder was replaced by niobium because recent tests have shown that uranium and plutonium samples show higher counting rates when dispersed in a niobium matrix.

Accelerator mass spectrometry (AMS). The AMS measurement was performed at the Laboratory of Ion Beam Physics of ETH Zurich, Switzerland, using the compact, low energy 0.6 MV AMS facility “Tandy”25. Over the past few years, the Tandy system was upgraded with an additional magnet on the high energy side and equipped with an improved ion source26. Since 2011, helium is used as a stripper gas for all measurements on the Tandy29. With a transmission of up to 40% for uranium and thorium2, an abundance sensitivity of up to 10^4, the compact (lab-sized) ETH Zurich AMS system Tandy is well suited to detect ultra-trace amounts of actinides.

The setup for plutonium measurements is summarized in ref. 30. Briefly, negative ions of Pu are selected by the magnetic and electrostatic filters1. Alpha spectrometry

### Table 2 | Results of the α-spectrometry and AMS. For plant samples, no quantification of 240Pu and 239Pu was undertaken. 240Pu/239Pu is calculated from the AMS ratios 240Pu/242Pu and 239Pu/242Pu. 240Pu is added as a spike. The values marked with an asterisk (*) are upper limits (decision thresholds according to ISO 11929, see text)

| Sample | ETH Label | Bq/kg | 240Pu/239Pu | AMS at/at ratios |
|--------|-----------|-------|-------------|-----------------|
|        |           |       | 240Pu/239Pu | 240Pu/242Pu     |
|        |           |       | at/at       | at/at           |
|        |           |       | 239Pu/242Pu | 239Pu/242Pu     |
| A-S    | TP0178    | < 2.11| n.d.        | < 0.000005*     |
| B-S    | TP0179    | < 0.47*| < 0.300     | < 0.000005*     |
| C-S    | TP0180    | < 0.66*| < 0.172     | < 0.000005*     |
| D-S    | TP0181    | < 0.50*| < 0.285     | < 0.000005*     |
| E-S    | TP0182    | < 0.29*| < 0.220     | < 0.000005*     |
| F-S    | TP0183    | < 0.39*| < 0.200     | < 0.000005*     |
| G-S1   | TP0184    | < 0.370| n.d.        | < 0.000005*     |
| G-S2   | TP0185    | < 0.152| < 0.246     | < 0.000005*     |
| H-S1   | TP0172    | < 0.155| < 0.173     | < 0.000312      |
| H-S2   | TP0171    | < 0.161| 0.205 ± 0.039| 0.000367        |
| I-S    | TP0188    | < 0.532| < 0.615     | 0.000150        |
| J-S1   | TP0186    | < 0.405*| < 0.103     | 0.000055*       |
| J-S2   | TP0187    | < 0.426*| < 0.322     | 0.000144        |
| A-V    | TP0272    | N/A   | n.d.        | 0.00146         |
| B-V    | TP0273    | N/A   | < 0.000111*| < 0.000184*     |
| C-V    | TP0274    | N/A   | n.d.        | < 0.000111*     |
| D-V    | TP0275    | N/A   | < 0.242     | < 0.000011*     |
| E-V    | TP0276    | N/A   | < 0.973     | < 0.000062*     |
| F-V    | TP0277    | N/A   | < 0.000111*| < 0.000184*     |
| G-V    | TP0278    | N/A   | < 0.000055*| 0.000867        |

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