

PLUTONIUM EMISSION FROM THE FUKUSHIMA ACCIDENT

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ABSTRACT

A strong earthquake and subsequent tsunami on 11th March 2011 initiated a severe accident in units 1 to 4 of Fukushima Dai-ichi nuclear power plant, resulting in substantial releases of radionuclides. While much has since been published on environmental contamination and exposure to radio-iodine and radio-caesium, little is known about releases of plutonium and other non-volatile elements. Although the total activities of released ¹³¹I, ¹³⁴Cs and ¹³⁷Cs are of the same order of magnitude as of the Chernobyl accident in 1986, the contribution of little volatile elements, including Pu, is much smaller in Fukushima fallout. The reason is the different physical nature of the accident sequence which led to a release of some 10⁻⁵% of the core inventories only (to be compared with 3.5% from Chernobyl).

In this contribution the available data on Pu in Fukushima fallout will be reviewed. Data sources are mainly reports and press releases by Japanese authorities and a few scientific articles. The mean ratio ²³⁹⁺²⁴⁰Pu : ¹³⁷Cs in the near field around the NPP (mainly part of Fukushima prefecture and districts of adjacent prefectures) can be assumed about 3×10⁻⁷, to be compared to nearly 0.01 in the vicinity of Chernobyl, down to about 3×10⁻⁶ in Central Europe. Isotopic ratios ²³⁸Pu : ²³⁹⁺²⁴⁰Pu are about 2.2 (0.46 and 0.035 in Chernobyl and global fallout, respectively). Activity concentrations of Fukushima-²³⁹⁺²⁴⁰Pu in surface soil were found up to above 0.1 Bq/kg d.m. in the immediate vicinity of the Fukushima NPP and about one order of magnitude less in Fukushima city, about 60 km away. The ²³⁹⁺²⁴⁰Pu activity released into the atmosphere is roughly estimated some 10⁹ Bq (Chernobyl: almost 10¹⁴ Bq).

1. INTRODUCTION

The accident at the Fukushima Dai-ichi nuclear power plant was *physically* caused by a strong earthquake and subsequent tsunami. While the earthquake itself seems not to have caused fatal damage, the tsunami did, as it damaged technical components which are needed to guarantee removal of residual heat from the fuel which is substantial even if a reactor is shut-down. This was the case since the earthquake properly triggered automatic shut-down of three working units, as it should do. (The fourth unit which was also involved in the accident, to minor degree, was shut down for service at the moment. Its fuel was relocated to the storage pool which also requires cooling.) The components fatally affected by the tsunami were seawater intake pumps, back-up diesel generators and their diesel tanks, partly structurally damaged, submerged and washed away by the tsunami. Therefore the ultimate heat sink was lost together with power, as connection to the external grid had been interrupted by the earthquake. Detailed technical information about the accident can be found in several reports, [1,2,3,4]. Also the Wikipedia article [5] is a good introductory source providing many links to literature.

The *political* “root cause” of the accident was neglect of safety provisions and partly missing emergency procedures, in turn caused by insufficient administrative structures in terms of corruption (a Japanese variety called amakudari [6]), independence of the regulatory and supervising authority from the nuclear industry and its lack of power to enforce safety upgrading, even if weaknesses were known. This aspect has been discussed e.g. in [7, 1].

The inability to cool the fuel led to melting of parts of the reactor cores (which parts exactly, is not yet well known), release of radionuclides first into the containment which has been designed for that purpose; but ruptures and leaks whose causes are not entirely clarified allowed hydrogen gas which is generated in a reaction of zirconium (constituent of the fuel hulls) with water to escape, to form oxyhydrogen by mixing with ambient air. This ignited and the explosion seems to have produced further structural damage in the containments, at the one hand, and on the other hand released large amounts of radionuclides into the environment. Pictures of the damage can be seen in [8].

Since emergency response was insufficient for several days release into the atmosphere continued for more than a week. At the same time, measures taken for emergency cooling, using concrete pumps and fire fighter equipment and the like, together possibly with structural leaks in the containment base, resulted in large quantities of partly highly contaminated water being set free. (To a minor degree this continues until today.)

During parts of the atmospheric release period winds blew towards the Pacific Ocean, but substantial fallout occurred over inhabited and agriculturally used land. A significant contamination “trace” extends from the NPP towards Northwest, right to Fukushima City (population ca. 300,000), about 60 km away. In parts of that trace ground contamination amounts to several MBq/m² of ¹³⁷Cs (the radiologically most important radionuclide in longer term perspective) which made evacuation of the population necessary. But also other parts of Central-Northern Japan were subjected to fallout, to different degree. Contamination maps can be found in various reports, e.g. [10,21].

Release of contaminated water into the ocean had different consequences. At the one hand the immense dilution capacity quickly and very greatly reduced concentrations in sea water. On the other hand, accumulation in sediments and above all, in certain marine food-chains can be very efficient. In fact until today sometimes excessively high radio-caesium concentration is encountered in certain species of sea life. For discussion of oceanic dispersion and impact on the marine environment, see e.g. [9,10].

The physical conditions during core melting are not yet known in detail, and they were probably different between the three cores affected within the reactors, and certainly these different from the one relocated in the cooling pool. However, as can be concluded from the composition of the released matter, temperatures were not high enough to set free significant amounts of elements with high evaporation temperatures. Thus, luckily, the contribution of strontium and plutonium isotopes, to name only these two radiologically very unpleasant ones, was very low. Also apparently no explosive fuel fragmentation occurred, so that little, if any of the release happened in the form of fuel fragments. On the other hand large fractions of the inventory of radioactive noble gases, radio-iodine and radio-caesium were released the latter (mainly ¹³¹I and ^{137,134}Cs) being of radiological significance.

Much literature about environmental and foodstuff contamination has been published in the two years since the accident and will not be referenced here in detail. However, little has been published about Pu contamination, understandably given its small contribution, fortunately, which results in that its detailed investigation and discussion is of lower priority. This article is devoted to Pu and comparison of the findings with the situation after the Chernobyl accident, 1986.

Shortly recall the Chernobyl accident. An ill-conducted experiment (ironically aimed at improving safety) together with organisational sloppiness, missing independent control and a heavy-handed administration, and lack of safety culture altogether, lead to the explosion of the unit 4 reactor of that plant. The reactor is of entirely different type, called RBMK (acronym from Russian), than the one of Fukushima, and of most common reactors currently in use all over the world; most are boiling or pressurized water reactors (BWR or PWR, Fukushima of the first type) in which light water is used as coolant and moderator. The RBMK's moderator is graphite instead of water, while the cooling agent is water like in common types. It has no pressure vessel but instead pressure tubes within which the fuel rods are located, and imbedded in a large graphite body (therefore also called LWGMR, light-water graphite moderated reactor). While reactivity decreases in BWR and PWR for physical reasons, if temperature increases, the opposite is true for RBMK, in certain operational regime. Although forbidden, the Chernobyl reactor was brought into such regime by bypassing safety provisions, which led to its explosion. Parts of fuel were ejected into the environment, parts melted. The graphite ignited and the fire led to a powerful convective stream which released large amounts of radionuclides partly high up into the atmosphere during about one week. The event led to contamination of large parts of Europe, easily measurable until today.

Due to the different physical characteristic of the accident the composition of the fallout was different. Violent fracturation caused large amounts of fuel being ejected in larger or smaller particles, or being carried by convection caused by the fire. Small "hot" particles could travel over more than 1000 km. Also temperature was higher, allowing also less volatile elements being released more easily. As a consequence Sr and Pu isotopes (together with other little volatile, but radiologically less important ones such as ^{125}Sb , ^{144}Ce or $^{154,155}\text{Eu}$) contribute more in Chernobyl, than in Fukushima fallout. The large releases of $^{103,106}\text{Ru}$ from Chernobyl are specific: Ru is actually little volatile, but through a reaction with air which occurred in Chernobyl but not in Fukushima, highly volatile compounds are formed. - Comparative data will be given in later sections.

Good introductory overviews on the Chernobyl accident and the RBMK reactor, and links to more detailed sources can also be found in Wikipedia, [11].

2. DATA AND METHODS

2.1. Data Sources

First data about Pu concentrations in the environment in the aftermath of the Fukushima accident were published in English from about late March 2011, i.e. about two to three weeks after the event. Environmental surveillance as well as many nuclear regulatory competences

(the exact structure of responsibilities is very complicated) belongs to the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) which organized and coordinated sampling. A large amount of data, also from other authorities, e.g. the Ministry for Fishery, can be found on the MEXT website [12]. Further important data on Pu are reported on TEPCO's (the owner of Fukushima NPP) web site, [13] (contamination data only until end-2011).

From the beginning also scientific institutions were involved in monitoring programs. These also started investigations in a more scientific scope, and by now a few articles have been published which also address Pu.

As far as I could find in the data sources (and as far as accessible to somebody who does not read Japanese), media systematically assayed for Pu (and other radionuclides) from the beginning are seawater and sea sediments off the coast of Fukushima and adjacent prefectures, soil in the region affected by fallout, air samples (data reported by MEXT, [12]) and soil from the NPP ground as well as liquid effluents (by TEPCO, [13]). Only scattered data are available from the farther surroundings. It can be assumed that continuous and frequent monitoring of environmental media for Pu from locations more distant than a few km was deemed unnecessary by the authorities since it had soon turned out that Pu contributed very little in fallout.

2.2. Attribution of Pu to Emission Sources

Pu which is detected in the environment can stem from different sources. The most important is global fallout, resulting from atmospheric nuclear bomb explosions, for ^{239}Pu and ^{240}Pu and the disintegration of a US satellite in the atmosphere, for ^{238}Pu . A further source is the Chernobyl accident (all Pu isotopes). The Fukushima fallout comes on top of these. Possible further anthropogenic sources are thinkable, such as unaccounted spills or nuclear terrorism. Natural Pu can be excluded from considerations because it exists in minute traces only. In Japan, Chernobyl Pu and for that part of Japan, also Pu from unaccounted sources can be excluded as relevant to my knowledge. Hence global fallout and Fukushima remain as sources.

The contribution of sources can be separated by exploiting the different isotopic ratios, or "isotopic signature" of the different sources. Conventional alpha spectrometry which is the mostly used radiometric method usually reports $^{239+240}\text{Pu}$ and ^{238}Pu , since the alpha lines of ^{239}Pu and ^{240}Pu cannot be separated this way. Isotopes 239 and 240 can be separated by mass spectrometry-type of analytics; this technique is more expensive but increasingly available due to its great use for other purposes involving isotopic analysis.

Given two different sources (global and Fukushima fallout) with different, but known $^{238}\text{Pu} : ^{239+240}\text{Pu}$ ratios, the contributions of the both in a sample which is a mixture of both can be calculated, see e.g. [14]. For three possible sources, results for three isotopes would be necessary, e.g. 238, 239+240 and 241, the latter usually being assayed by LSC. Alternatively, separate analysis of 238, 239 and 240 can do it as well.

The Pu signature of global fallout has been quite well known for years. For Fukushima fallout the ratio can be inferred from samples taken from very close to the NPP which show relatively high Pu concentrations whose origin is likely the NPP accident (unaccounted earlier routine emissions are in principle possible, but unlikely in such amount). These data also underlie the discussion of radionuclide ratios, including the ones of Pu isotopes, in [15] and in [14].

3. RESULTS

3.1. Pu ratio in Fukushima emission

^{238}Pu and $^{239+240}\text{Pu}$ has been measured in soil samples on the ground of the NPP by TEPCO, [13], about weekly in 2011 (no data communicated afterwards). The results are shown as scatter plot of ^{238}Pu vs. $^{239+240}\text{Pu}$ in fig. 1. A clear relation is visible, the clearer, the higher the concentration, because then the contribution of the background – global fallout as can be assumed, if possible, past unaccounted emissions are not considered – becomes less relevant.

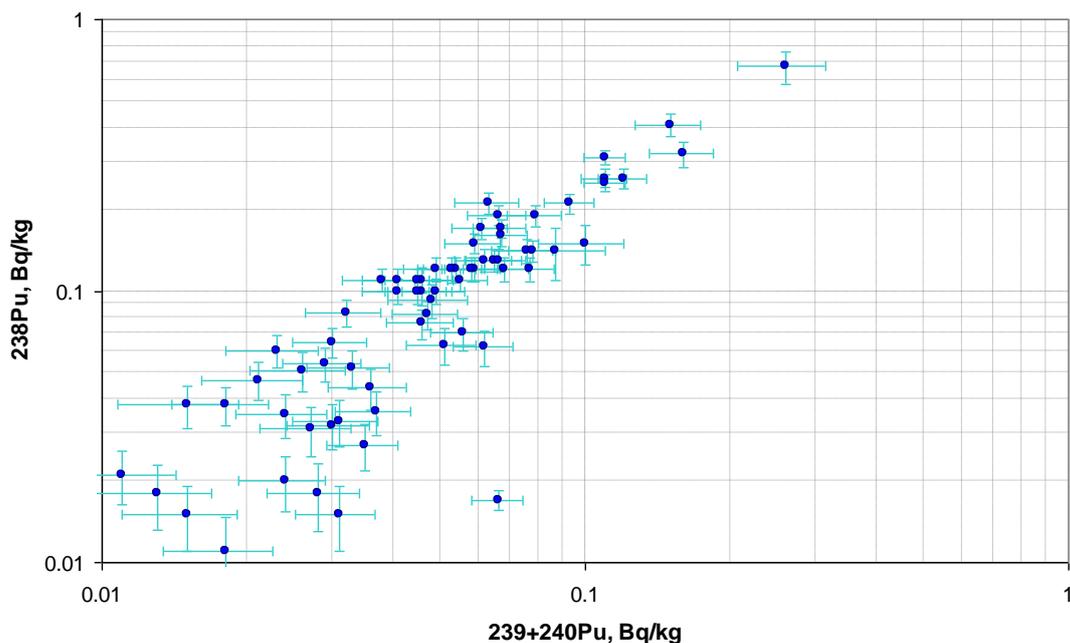


Figure 1: Pu in soil samples from two locations on the grounds of the NPP. Uncertainty bars: from uncertainties given in the data sources.

This becomes more evident if the ratio is plotted against the concentration, fig. 2 From samples taken soon after the accident, we estimated a median 2.28 (95% conf. interval 1.98 – 2.58), [15] and 2.19 ± 0.48 (1σ), [14], for Fukushima emissions.

The background Pu ratio in global fallout has been reported 0.035 ± 0.008 [16,17]. In a MEXT document [18] 0.031 is quoted as Japanese average, based on samples taken before Fukushima. In [19] a ratio 0.0261 is suggested as slope of a scatter plot of ^{238}Pu vs. $^{239+240}\text{Pu}$ in pre-Fukushima samples. - In Chernobyl fallout, the ratio equals 0.46 ± 0.03 [17], for comparison.

In soil samples taken farther from the NPP the Pu ratio is lower since the Fukushima contribution, if any, is “diluted” by global fallout Pu which has much lower Pu ratio.

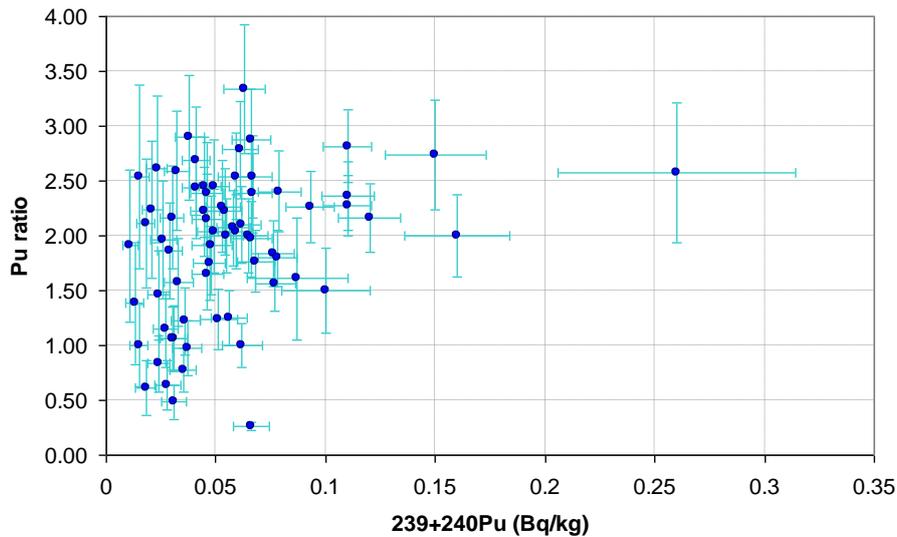


Figure 2: Pu ratio in dependence of $^{239+240}\text{Pu}$ concentration.

3.2. Geographical distribution of the Pu ratio

Data taken from MEXT [18,20] and Yamamoto et al. [24] were used to generate a map of the $^{238}\text{Pu} : ^{239+240}\text{Pu}$ ratio in the region around the NPP (mainly Fukushima prefecture), fig. 3. The map was generated by ordinary block kriging based on a spherical variogram model with nugget estimated from the data. Cell size is 2 km x 2 km, projection Lambert azimuthal equal-area, spherical earth, zero-point 140.5° E / 37.5° N. The region has been blanked deliberately to exclude areas with no samples where high uncertainty must be expected.

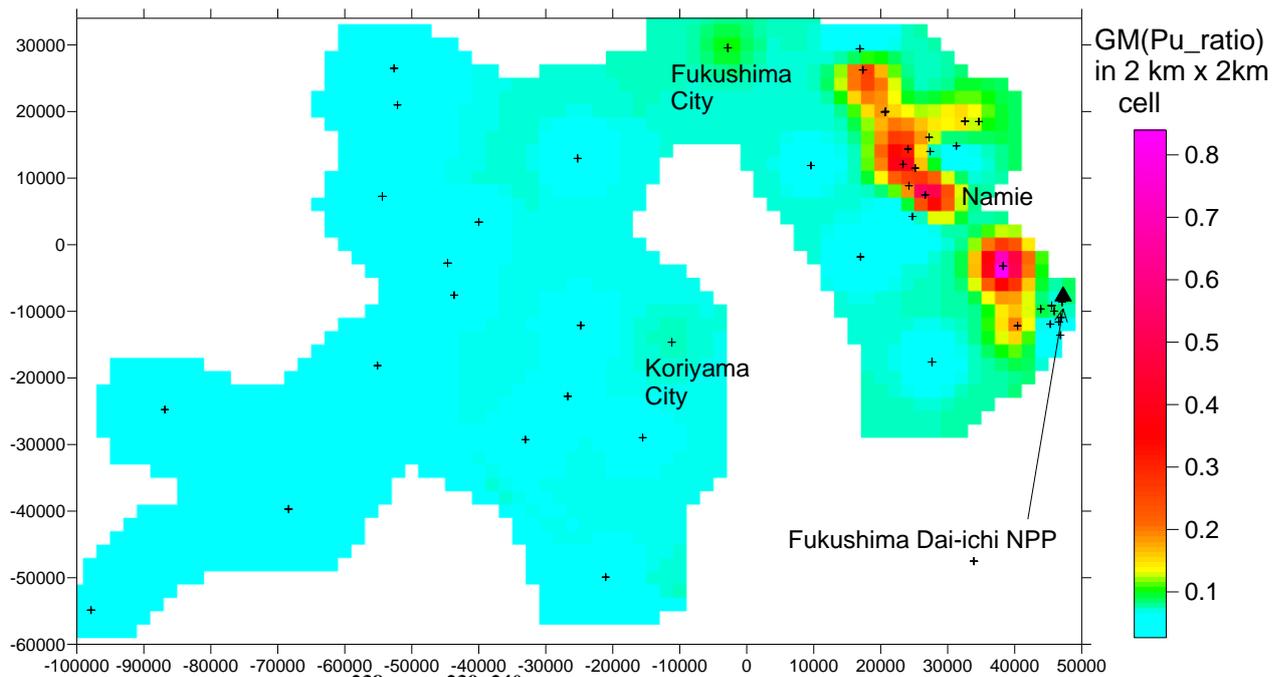


Figure 3: Ratio $^{238}\text{Pu} / ^{239+240}\text{Pu}$ in soil samples from the region around the Fukushima NPP. Coordinates: m. Crosses: locations of samples. Samples from the NPP grounds not included.

The “trace” towards NW from the NPP, in which the Pu ratio deviates strongly from the background (ca. 0.035; the blue area) coincides very well with the contamination trace delineated by areogamma and soil surveys [21,19]. This proves that indeed Pu has been emitted by the accident, if only in small quantities. Given a relatively small number of samples in which both ^{238}Pu and $^{239+240}\text{Pu}$ could be detected, in most of the region, resolution of the map is low.

3.3 Ratio Pu : Cs

The ratio of activity concentrations in Fukushima fallout between Pu isotopes and ^{137}Cs have been estimated by a few authors. The results are summarized in table 1. For this article, soil samples given by MEXT [20,22] were evaluated using the method demonstrated in [14]. (For only three samples results for Pu and ^{137}Cs could be found for coinciding sampling locations. For the samples from the Pu survey no gamma results are available.) As Pu ratio in global fallout 0.035 was used; choosing 0.031 instead does not lead to very different results.

Zheng and colleagues [23] based a similar analysis on the $^{239}\text{Pu} / ^{240}\text{Pu}$ atomic ratio which is well known for global fallout and can be inferred for Fukushima fallout from highly contaminated samples as about 0.30 – 0.33; calculated reactor inventories [15] would yield about 0.39. Yamamoto and colleagues [24] seem to have used a similar method as in this article (no details given).

$^{239+240}\text{Pu}$ activity concentrations attributable to Fukushima fallout are up to over 0.2 Bq/kg d.m. in soil samples from the NPP grounds; up to over 0.01 Bq/kg in soil from Fukushima

City (compared to global-²³⁹⁺²⁴⁰Pu up to 0.4). In terms of deposition density, up to almost 4 Bq/m² Fukushima-²³⁹⁺²⁴⁰Pu are encountered in Namie town (one of the most affected places) which is about four times the global contribution. For ²³⁸Pu, the Fukushima contribution is much higher than the global one in many places (as detectable at all) because the Pu ratio is much higher in Fukushima (~2.19) than in global fallout (~0.035).

Table 1. Activity concentration ratio ²³⁹⁺²⁴⁰Pu : ¹³⁷Cs in Fukushima fallout

author	location	medium	²³⁹⁺²⁴⁰ Pu / ¹³⁷ Cs ratio (Fukushima) (10 ⁻⁷)
Imanaka et al. 2012 [25]	Fukushima zone	soil	< 10
Zheng et al. 2011 [23] *	20 km S NPP	soil	3.6 ± 1.1
Yamamoto et al. [24]**	Itate village, near NPP	soil	1.6 ± 1.2
Bossew 2013 [14]	Koriyama, Fukushima City	soil, moss	2.8 ± 2.0
this article; data from [20,22]	Fukushima region	soil	2.8 (0.35...8.8)***

(* see also remarks in [14] (table 9); (** AM and SD calculated from values given in table 6 of [29] ; (***) AM over 3 samples and 95%-confidence interval, pooled from 500 simulations for each sample.

We can thus assume an activity concentration ratio ²³⁹⁺²⁴⁰Pu : ¹³⁷Cs in Fukushima fallout in the order 3×10^{-7} . For comparison, for Chernobyl fallout, the ratio was about 0.0088 in the surroundings of the Chernobyl NPP and about 3×10^{-6} in Austria (1000 km away) (references in [14]). The ratio decreases with distance from Chernobyl because Pu was ejected mainly with fuel particles which did not travel as far, in general, as releases in gaseous state or as small aerosols. In any case it is evident that the Pu:Cs ratio is significantly smaller in Fukushima, than in Chernobyl fallout.

3.4. Pu activity emitted by the Fukushima accident

A summary of estimates of ¹³⁷Cs emission into the *atmosphere* is given in an IRSN report [10; tab. 6-1, p. 46] together with references. They range from 10 to 50 PBq. Keeping with 15 PBq given by NISA [3; page VI-1], and assuming a ²³⁹⁺²⁴⁰Pu / ¹³⁷Cs ratio equal 2.8×10^{-7} , we find an atmospheric emission of ²³⁹⁺²⁴⁰Pu equal 4.2 GBq. Using the upper estimate of released ¹³⁷Cs, 50 PBq [26], a release of 14 GBq is found.

NISA gives a total release of 6.4 GBq ²³⁹⁺²⁴⁰Pu [3, attachment 4-2, table 5, p.7] corresponding a Pu/Cs ratio 4.3×10^{-7} , which coincides well with our ratio estimated from samples.

Table 2. Activity of $^{239+240}\text{Pu}$ released into the atmosphere by the Fukushima fallout

source	released $^{239+240}\text{Pu}$ activity (GBq)	fraction of inventory
NISA	6.4	-
Zheng et al. 2011 [23]	1.0 ... 2.4	$(1.2 \dots 2.9) \times 10^{-7}$
this article	4.2 ... 14	$(7.0 \dots 23) \times 10^{-7}$ *
Chernobyl, after [23] (refs. there)	87000	0.035

* assuming an inventory of 250 t fuel $\times 2.4 \cdot 10^{13}$ Bq/t [15] = 6×10^{15} Bq

A summary of releases and fractions of reactor inventory is given in table 2. The differences between Fukushima and Chernobyl releases are evident from the figures, and due to the very different physical release conditions.

Actually the fraction of Pu released into the environment can be expected to be higher, as table 2 deals with atmospheric releases only. Certainly some Pu has been released with liquid effluents and discharged into the ocean. The IRSN [9; p. 107] estimated the ^{137}Cs activity directly released into the sea as high as 27 PBq; “directly” means, not counting atmospheric fallout over the sea which is assumed comparatively irrelevant. The liquid discharges certainly also contained Pu. However, data from surveys of the marine environment have been reported by the authorities but the data available to me does not allow calculations analogous to ones above. Also more comprehensive analyses [27,28,29] confirms this.

3. CONCLUSIONS

There is sufficient evidence that Fukushima fallout also contains Pu. The released fraction of Pu of radio-caesium is much smaller than in Chernobyl fallout. The reason is probably found in the very different physical conditions of the accident sequence.

It should be stressed that the evidence of Pu from Fukushima does not pose any radiological concern due to its very low concentrations. Its investigation is however interesting for scientific reasons and for proper documentation of the environmental consequences. As a note, the estimated release of 4.2 GBq $^{239+240}\text{Pu}$ (in the isotopic composition estimated for the reactors) correspond to a mass of about 0.71 grams.

REFERENCES

1. The National Diet of Japan: Report of the Fukushima Nuclear Accident Independent Investigation Commission (NAIIC). <http://warp.da.ndl.go.jp/info:ndljp/pid/3856371/naiic.go.jp/en/report/index.html> (acc. 28.1.2013)

2. TEPCO: Investigation Committee on the Accident at the Fukushima Nuclear Power Stations of Tokyo Electric Power Company; Final report, 23.7.2012.
<http://icanps.go.jp/eng/final-report.html> (acc. 28.1.2013)
3. METI (Ministry of Trade, Economy and Industry), Nuclear Emergency Response Headquarters / Government of Japan: June 2011: Report of Japanese Government to the IAEA Ministerial Conference on Nuclear Safety - The Accident at TEPCO's Fukushima Nuclear Power Stations;
http://www.kantei.go.jp/foreign/kan/topics/201106/iaea_houkokusho_e.html (acc. 28.1.2013)
4. METI (Ministry of Trade, Economy and Industry), Nuclear Emergency Response Headquarters / Government of Japan: Additional Report of the Japanese Government to the IAEA – The Accident at TEPCO's Fukushima Nuclear Power Stations (Second Report), Sept. 2011.
http://www.meti.go.jp/english/earthquake/nuclear/iaea/iaea_110911.html (acc. 28.1.2013)
5. Wikipedia about the Fukushima accident:
http://en.wikipedia.org/wiki/Radiation_effects_from_Fukushima_Daiichi_nuclear_disaster
6. <http://en.wikipedia.org/wiki/Amakudari> (acc. 26.1.2013)
7. Stošić Z.V. (2012): Nuclear Fundamentals Remain. *Thermal Science* **16** suppl.1, 35 -49; doi: 10.2298/TSCI120305059S; download: <http://thermalscience.vinca.rs/pdfs/papers-2012/TSCI120305059S.pdf> (acc. 26.1.2013)
8. Cryptome: <http://cryptome.org/eyeball/daiichi-npp/daiichi-photos.htm> (acc. 28.1.2013)
9. IRSN (2012): Synthèse actualisée des connaissances relatives à l'impact sur le milieu marin des rejets radioactifs du site nucléaire accidenté de Fukushima Dai-ichi 13 juillet 2012;
http://www.irsn.fr/FR/base_de_connaissances/Installations_nucleaires/La_surete_Nucleaire/Les-accidents-nucleaires/accident-fukushima-2011/impact-japon/Pages/3-consequence-environnement-japon.aspx (acc. 28.1.2013)
10. IRSN (2012): Fukushima, un an après; Premières analyses de l'accident et de ses conséquences. Rapport IRSN/DG/2012-001 du 12 mars 2012.
www.irsn.fr/FR/expertise/rapports_expertise/surete/Pages/Rapport-Fukushima-1-an-apres_032012.aspx (acc. 28.1.2013)
11. Wikipedia about the Chernobyl accident:
http://en.wikipedia.org/wiki/Chernobyl_disaster; RBMK reactor:
<http://en.wikipedia.org/wiki/RBMK> (acc. 24.1.2013)
12. MEXT, Monitoring information of environmental radioactivity level:
<http://radioactivity.mext.go.jp/en/> (acc. 24.1.2013)
13. TEPCO press releases: www.tepco.co.jp/en/press/corp-com/release/index-e.html (acc. 25.1.2013)
14. Bossew P. (2013): Anthropogenic radionuclides in environmental samples from Fukushima prefecture. To be published in *Radiation Emergency Medicine* Vol. **2**, No. 1, 2013.
15. Kirchner G., Bossew P. and De Cort M. (2012): Radioactivity from Fukushima Dai-ichi in air over Europe, part 2: What can it tell us about the accident? *J. Environmental Radioactivity* **114**, 35 - 40.
16. Perkins P. W. and Thomas C. W. (1980): Worldwide fallout. In Hanson W. C. (Ed.): *Transuranic Elements in the Environment*. U.S. Department of Energy. ISBN: 978-0870791192.

17. Irlweck K. and Wicke J. (1998): Isotopic composition of plutonium immissions in Austria after the Chernobyl accident. *J. Radioanalytical Nuclear Chemistry* **227** (1-2), 133 – 136.
18. MEXT: Results of the Nuclide Analysis of Plutonium 238, 239+240 and 241 (Second Survey) by MEXT. Document from 21.8.2012; <338_0821_e-pdf>. Retrieved from the MEXT web site, [11].
19. MEXT (no date): Summarized Version of the “Results of the Research on Distribution of Radioactive Substances Discharged by the Accident at TEPCO’s Fukushima Dai-ichi NPP”; <PressR04_0802s.pdf>. Retrieved from the MEXT web site, [11].
20. MEXT: Results of the Radiation Monitoring of Soil in Fukushima Prefecture. Document from 6.4.2012; <232_e_0409.pdf>. Retrieved from the MEXT web site, [11].
21. MEXT: Results of the (i) Fifth Airborne Monitoring Survey and (ii) Airborne Monitoring Survey Outside 80km from the Fukushima Dai-ichi NPP. Document from 28.9.2012; <203_0928_14e.pdf>. Retrieved from the MEXT web site, [11].
22. MEXT: Results of nuclide analysis of gamma-emitting nuclides (Cs-134, Cs-137 and Ag-110m) (second survey) (As of March 1, 2012). Document <xx-0912018_e.pdf> (xx: Japanese characters). Retrieved from the MEXT web site, [11].
23. Zheng J. et al. (2011): Isotopic evidence of plutonium release into the environment from the Fukushima DNPP accident. *Sci. Rep.* **2** / 304; doi: 10.1038/srep00304.
24. Yamamoto M., Takada T., Nagao S., Koike T., Shimada K., Hoshi M., Zhumadilov K., Shima T., Fukuoka M., Imanaka T., Endo S., Sakaguchi A. and Kimura S. (2012): An early survey of the radioactive contamination of soil due to the Fukushima Dai-ichi Nuclear Power Plant accident, with emphasis on plutonium analysis. *Geochemical Journal* **46**, 341 – 352.
25. Imanaka T., et al. (2012): Early radiation survey of Itate village which was heavily contaminated by the Fukushima accident, conducted on 28 and 29 March 2011. *Health Physics* **102** (6), 680 - 686.
26. Stohl A., Seibert P., Wotawa G., Arnold D., Burkhardt J.F., Eckhardt S., Tapia C., Vargas A. and Yasunari T. J. (2011): Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition. *Atmos. Chem. Phys. Discuss.*, **11**, 28319–28394; www.atmos-chem-phys-discuss.net/11/28319/2011/ (acc. 29.1.2013);doi:10.5194/acpd-11-28319-2011.
27. Bu W. T., Zheng J., Aono T., Tagami K., Uchida S., Zhang J., Honda M. C., Guo Q. J. and Yamada M. (2012): Investigating plutonium contamination in marine sediments off Fukushima coast following the Fukushima Dai-ichi Nuclear Power Plant accident. Proceedings of the International Symposium on Environmental monitoring and dose estimation of residents after accident of TEPCO's Fukushima Daiichi Nuclear Power Stations Shiran Hall, Kyoto, Japan, December 14, 2012. http://www.rri.kyoto-u.ac.jp/anzen_kiban/outcome/index.html (acc. 29.1.2013)
28. Bu W. T., Zheng J., Aono T., Tagami K., Uchida S., Zhang J., Honda M. C., Guo Q. J. and Yamada M. (2013): Determination of plutonium isotopes in marine sediments off the Fukushima coast following the Fukushima Dai-ichi Nuclear Power Plant accident. *Biogeosciences Discuss.*, **10**, 643–680, 2013; doi:10.5194/bgd-10-643-2013; www.biogeosciences-discuss.net/10/643/2013/ (acc. 29.1.2013)
29. Zheng J., Aono T., Uchida S., Zhang J. and Honda M. C. (2012): Distribution of Pu isotopes in marine sediments in the Pacific 30 km off Fukushima after the Fukushima Daiichi nuclear power plant accident. *Geochemical Journal* **46**, 361 – 369.