HIROSAKI UNIVERSITY

Radiation Emergency Medicine 2013 Vol. 2, No. 1 69-75

REM
Radiation Emergency Medicine

Anthropogenic Radionuclides in Environmental Samples from Fukushima Prefecture

Peter Bossew

German Federal Office for Radiation Protection, Köpenicker Allee 120 - 130, 10318 Berlin, Germany

Received 13 July 2012; revised 26 October 2012; accepted 9 November 2012

Four environmental samples from Fukushima prefecture, taken in March 2012, were analysed with gamma and alpha spectrometry. The aim was determining radionuclide ratios. The estimated ratio of ^{134}Cs / ^{137}Cs equals 1.062, with standard deviation (indicating variability between samples) 0.024. The ratio seems to be somewhat higher than what is mostly reported in the literature, namely between <0.9 and about 1. The only other long-lived gamma radionuclide which could be identified is ^{110m}Ag, whose ratio to ^{137}Cs equals $(2.3 \pm 0.2)\ 10^{-3}$. The ratios of $^{239+240}$Pu and ^{137}Cs in Fukushima fallout are below 10^{-6}, calculated under the assumption that the only Pu sources are global and Fukushima fallout. More samples would be needed to improve the statistical reliability of the results, and to investigate the reason for the found slight discrepancy of Cs ratios with those reported in literature.

Key words: Fukushima nuclear accident, environmental samples, radionuclide ratios

1. Introduction

The accident at the Fukushima Dai-ichi nuclear power plant (NPP) in March 2011 has resulted in substantial radioactive contamination of parts of Fukushima and adjacent prefectures. Many measurement results of dose rate and activity concentrations in environmental media have since been reported. Very understandably the main focus of monitoring is radioprotection. Fewer results, in comparison, have so far been published about accurate determination of radionuclide concentration ratios, in particular for minor long-lived radionuclides.

Radionuclide ratios can contribute to understanding the physics of the accident and the history of its development. An example of this approach has been given in Kirchner et al. (2012)[1], based on the evaluations of air filters sampled in Europe during the passage of the Fukushima cloud.

At the occasion of a monitoring excursion by Hirosaki University I had the opportunity to take a few environmental samples with the purpose of determining radionuclide ratios. In this paper the results of gamma and alpha spectrometric measurements, as well as concentration ratios to ^{137}Cs are presented. A method to separate plutonium values according to their anticipated sources is shown and a Bayesian refinement is suggested to account for high uncertainties and resulting physical implausibility, which can occur when applying the method to low plutonium concentrations.

Peter Bossew: German Federal Office for Radiation Protection,
Köpenicker Allee 120 - 130, 10318 Berlin, Germany
E-mail: pbossew@bfs.de; peter.bossew@reflex.at

Table 1. Types and proveniences of the samples

	Type	location
sample 1	sand, drainage canal of parking area	Koriyama city
sample 2	topsoil, playground, run-off zone at foot of a hill slope	Fukushima city
sample 3	moss 1	same location as sample 2
sample 4	moss 2	same location as sample 2

Fig. 1. Sampling locations:
Left: sample 1; right: samples 2 to 4.

Table 2. Partition of samples and analyses performed

sample	dry mass (g)	AIAE	BfS	UniSalzburg
sample 1, fraction 1	54.78	γ		γ, α
sample 1, fraction 2	7.70		γ	
sample 2, fraction 1	35.57	γ		γ, α
sample 2, fraction 2	6.28		γ	
sample 3	29.7	γ		γ, α
sample 4	1.46	γ		γ

Table 3. Details on gamma spectrometry. Careful efficiency calibration of ^{134}Cs is essential because of coincidence summation

lab	detector (HPGe)	counting time (LT)	efficiency cal. 134Cs
AIAE	18% p-type,Canberra	0.3 - 2.3 ks	numerical (MC)
BfS	40 % n-type, Ortec	210 / 240 ks	^{134}Cs standard
Uni Salzburg	40 % p-type, Ortec	56 - 256 ks	^{134}Cs standard

2. Methods

2.1. Sampling

The samples were collected during an excursion in the Fukushima zone and surroundings, together with colleagues of Hirosaki University in March 2012. For this exercise samples were chosen which, based on experience, could be expected to be rather highly contaminated, supported by measurements of the in situ gamma dose rate. This means that the samples are not representative for the region. Types and sampling locations are summarized in Table 1. Pictures of the sampling locations are shown in Figure 1. Dose rates close to the sources were over 1 μSv/h. The samples were dried and homogenized by sieving (samples 1 and 2; aggregates > ca. 1 mm removed) or manual mixing (samples 3 and 4) and taken to Europe for analysis.

2.2. Gamma spectrometry

Three laboratories performed gamma spectrometry of the samples: the gamma lab of the Austrian Institute of Applied Ecology (AIAE), the lab of the German Office for Radioprotection, Berlin (BfS), and the lab of the division of physics and biophysics of the University of Salzburg. Methodology is routine and not further discussed here in detail, as all labs are experienced in the field. All labs performed summation correction for ^{134}Cs. Counting times were between a few 100 s and a few 100 ks. Results given here are decay corrected to 20 March 2011. Details about

the partition of samples and basic technical details of gamma spectrometry are given in Table 2 and Table 3.

It should be stressed that the exercise was not intended to compare labs in a kind of mini-intercomparison. Instead, labs were interested in measuring Fukushima samples and took the advantage of some being available. The main purpose of this article is indeed presenting results on radionuclide ratios and of plutonium components in particular.

2.3. Alpha spectrometry

Plutonium was investigated by the University of Salzburg only. Three samples were measured twice, since the low activity concentrations render the analysis quite uncertain. Sample 4 was too small for a meaningful analysis. For further evaluation the means of the two measurements were used for each sample. Standard uncertainty was chosen the larger of the standard deviation between replicates and the mean of individual counting plus process uncertainties (around 15%). Radiochemical procedure and radiometry:

The method is a sequence of ashing, acid digestion, separation by ion exchange and co-precipitation with NdF_3. The procedure follows the one developed by La Rosa et al., 1992[2] and Vajda et al., 1992[3], and has been the routine method at Salzburg University for years. For more details see e.g. Bossew et al. 2007[4] and Tieber et al. 2009[5]. Alpha spectrometry was done with a Canberra PIPS type detector.

2.4. Evaluation of plutonium activity concentrations and Pu/Cs ratios

2.4.1. Separation of plutonium contributions

The plutonium isotopes in the samples are assumed to have two sources: global fallout and possible Fukushima fallout. Chernobyl fallout is unlikely to have contributed in Japan and local sources can probably be excluded. If we denote P_9 and P_8 the $^{239+240}$Pu and ^{238}Pu concentrations, respectively, as measured in the samples, and add upper indices 'G' and 'F' to denote global and Fukushima, we can put:

$P_9 = P_9^G + P_9^F$

$P_8 = P_8^G + P_8^F = \alpha^G P^G + \alpha^F P_9^F$,

if α^G and α^F denote the ratios,

α^G, α^F := ^{238}Pu / $^{239+240}$Pu in global and Fukushima fallout, respectively.

The above system can then be written $\mathbf{P} = \mathbf{A}\,\mathbf{P_9}$,

with $\mathbf{P} := (P_9, P_8)^T$, $\mathbf{P_9} := (P_9^G, P_9^F)^T$, and the matrix

$$A := \begin{pmatrix} 1 & 1 \\ \alpha^G & \alpha^F \end{pmatrix}.$$

The wanted components P_9^G, P_9^F are easily found by solving the system (matrix inversion), $\mathbf{P_9} = \mathbf{A^{-1}}\,\mathbf{P}$, and P_8^G and P_8^F by multiplying with the respective α. Standard uncertainties of the results can be calculated by Gaussian propagation. The method has been also used by other authors, e.g. Mietelski and Was (1995)[6] or Tieber et al. (2009)[5] (this paper has a typing error in the respective formula).

2.4.2. Bayesian approach

The uncertainties of the measured values are relatively high, due to small Pu concentrations and low sample sizes. The resulting $P_{8,9}^{G,F}$ can therefore be numerically be below zero, although this is not possible physically. This can be corrected with Bayesian reasoning, stipulating priors for all quantities which force them to be positive definite. The priors are chosen the simplest possible, namely step functions Θ(x).

In practice this is implemented through simulation. We generate random realizations of all quantities (P_9, P_8, α^G and α^F, and also the ^{137}Cs concentration, since we also want to estimate the mean Pu/Cs ratios) as follows. To each reported measured value an "error" ~N(0,unc) is added (chosen normally distributed lacking better information), where 'unc' denotes the reported standard uncertainty. Only values greater than zero are retained. Then the wanted $P_{8,9}^{G,F}$ are calculated and again only those retained which are positive. This is repeated many times and the statistics over realizations are computed (done with home-made software).

3. Results

3.1. Gamma and alpha spectrometry

The activity concentrations of the samples are given in Table 4. The values are means over labs for ^{134}Cs and ^{137}Cs, otherwise the values by Salzburg University which was the only one to evaluate these radionuclides. For the Cs, the uncertainties given in the table are ("external") standard deviations between the results reported by different labs, generally higher than the "internal" ones reported for the individual measurements. (This may point to systematic differences in detector calibration.) For the other radionuclides the uncertainty is the one reported by Salzburg University.

For Pu see section 2.3.

The ratios of the activity concentrations to ^{137}Cs are shown in Table 5. For comparison, ratios for Chernobyl fallout are given, as determined a few km South of the Chernobyl NPP (Bossew et al. 2004[7]) and in Austria (Bossew et al. 2001[8]), about 1000 km away. These values are decay corrected to 1 May 1986. The ratios of nuclides other than ^{134}Cs to ^{137}Cs were calculated from the values of Uni Salzburg only. For the ^{110m}Ag / ^{137}Cs ratio sample 4 was removed since the value appears an outlier. The resulting ratio, 0.0023, is in good agreement with the value 0.0019 reported in Hirose (2011)[9]. As estimate of the mean ^{134}Cs/^{137}Cs ratio we find 1.062 with standard uncertainty u = 0.024. The standard uncertainty of the mean, or standard error, is u/√(number of samples = 4) = 0.012. A more in-depth statistical analysis (not given here) shows essential consistency between laboratories in

Table 4. Measurement results. Units: kBq/kg dry for ^{134}Cs and ^{137}Cs, others Bq/kg dry. Decay corrected to 20 March 2011 (half lives from the LUND database [32]). '<': LLD, α = β=0.05; unc: standard uncertainty

nuclide	sample 1	unc	sample 2	unc	sample 3	unc	sample 4	unc
^{137}Cs	72900	9%	25000	8%	42400	10%	34200	28%
^{134}Cs	79200	16%	26900	8%	43800	13%	36000	31%
^{110m}Ag	186	10%	56	10%	114	10%	388	16%
^{155}Eu	< 13		< 12		< 14		< 44	
^{154}Eu	< 10		< 9		< 10		< 25	
^{144}Ce	< 80		< 70		< 80		< 200	
^{125}Sb	< 48		< 46		< 52		< 130	
^{106}Ru	< 165		< 150		< 175		< 420	
^{95}Zr	< 600		< 500		< 600		< 1400	
^{60}Co	< 3		< 2		< 3		< 11	
$^{239+240}$Pu	0.19	36%	0.27	21%	0.41	15%		
^{238}Pu	0.015	52%	0.038	20%	0.038	20%		

Table 5. Mean ratios of activity concentrations to ^{137}Cs. Decay corrected to 20 March 2011. Pu ratios taken from Table 7, last column. unc: standard uncertainty. Chernobyl data for comparison (corr. 1 May 1986), references see text

nuclide	**ratio, this study**	unc	Chernobyl: Chernobyl zone	Chernobyl: Austria
^{137}Cs	**1**	-	-	-
^{134}Cs	**1.062**	0.024	0.537	0.567
^{110m}Ag	**0.0023**	0.0002		0.034
^{155}Eu	**<1.7 E-04**		0.020	
^{154}Eu	**<1.3 E-04**		0.016	3.8 E-4
^{144}Ce	**<1.0 E-03**			0.067
^{125}Sb	**<6.3 E-04**		0.068	0.023
^{106}Ru	**<2.2 E-03**			0.46
^{95}Zr	**<7.7 E-03**			
^{60}Co	**<4.5 E-05**		0.0022	2.5 E-4
$^{239+240}$Pu (Fukushima)	**2.8 E-7**	2.0 E-7	0.0088	3 E-6
^{238}Pu (Fukushima)	**5.9 E-7**	4.5 E-7	0.0040	1.6 E-6

Table 6. Plutonium components and ratios to ^{137}Cs in three environmental samples from Fukushima prefecture. Activity concentrations in Bq/kg (dry). Calculated with conventional statistics. unc: standard uncertainty

	sample 1	sample 2	sample 3	all
$^{239+240}$Pu (Fukushima)	**0.0040**	**0.013**	**0.011**	
Unc	0.0039	0.004	0.004	
$^{239+240}$Pu (global)	**0.19**	**0.26**	**0.40**	
Unc	0.07	0.06	0.06	
^{238}Pu (Fukushima)	**0.0087**	**0.029**	**0.025**	
Unc	0.0086	0.008	0.009	
^{238}Pu (global)	**0.0066**	**0.0091**	**0.014**	
Unc	0.0029	0.0029	0.004	
$^{239+240}$Pu / ^{137}Cs (Fukushima)	**5.2E-08**	**4.9E-07**	**2.5E-07**	**2.6E-07**
Unc	5.2E-08	1.4E-07	8.8E-08	2.2E-07
^{238}Pu / ^{137}Cs (Fukushima)	**1.1E-07**	**1.1E-06**	**5.4E-07**	**5.7E-07**
Unc	1.1E-07	3.1E-07	1.9E-07	4.8E-07

determination of the ratio.

3.2. Evaluation of plutonium components

The following values of α were chosen: $\alpha^G = 0.035 \pm 0.008$ (Perkins and Thomas 1980[10]; Irlweck and Wicke 1998[11]) $\alpha^F = 2.19 \pm 0.48$ The latter value is taken from results reported by Japanese authorities for soil samples in the close vicinity of the Fukushima NPP (also discussed in Kirchner et al. 2012[1]), where other statistics of this ratio are given. α^F as used here has been recalculated from the original Japanese data. Also references can be found in this paper.)

The results are given in Table 6 for conventional and in Table 7 for Bayes statistic. Major differences appear only for sample 1. This could be expected because the measured values were particularly uncertain for this sample and therefore the chance is relatively high that a realization of the component Pu concentrations would be below zero in conventional statistics.

Table 7. Plutonium components and ratios to ^{137}Cs in three environmental samples from Fukushima prefecture. Activity concentrations in Bq/kg (dry). Bayes correction applied. unc: standard uncertainty

	sample 1	sample 2	sample 3	all
$^{239+240}$Pu (Fukushima)	**0.0056**	**0.013**	**0.013**	
Unc	0.0036	0.004	0.006	
$^{239+240}$Pu (global)	**0.179**	**0.27**	**0.40**	
Unc	0.074	0.05	0.06	
^{238}Pu (Fukushima)	**0.011**	**0.028**	**0.025**	
Unc	0.007	0.009	0.009	
^{238}Pu (global)	**0.0062**	**0.0094**	**0.014**	
Unc	0.0029	0.0034	0.003	
$^{239+240}$Pu / ^{137}Cs (Fukushima)	**7.6E-8**	**4.7E-7**	**2.8E-7**	**2.8E-7**
Unc	5.0E-8	1.7E-7	1.5E-7	2.0E-7
^{238}Pu / ^{137}Cs (Fukushima)	**1.5E-7**	**1.1E-6**	**5.5E-7**	**5.9E-7**
Unc	9.5E-8	3.3E-7	2.0E-7	4.5E-7

As reference ^{137}Cs values the ones measured by the same lab (Salzburg University) were used. The mean Pu : Cs ratios in Fukushima fallout are very small in the investigated samples, with $^{239+240}$Pu : ^{137}Cs = (2.8 ± 2.0) E-7 and ^{238}Pu : ^{137}Cs = (5.9 ± 4.5) E-7. It should be stressed again that these results are valid only under the conditions, (1) the α values are correct for the locations and (2) no other Pu components are present. The presence of a Chernobyl component (α^{Ch} = 0.46 ± 0.03; Tieber et al. 2009[5] after Irlweck and Wicke 1998[11]) would modify the finding; however a third Pu isotope (^{241}Pu) would be needed to evaluate a system of three Pu components. The mathematics would essentially be the same. For a thorough analysis of Pu isotopic ratios in Fukushima fallout, including ^{241}Pu, see Zheng et al. 2011[12].

The ratios to ^{137}Cs include a small downwards bias, since ^{137}Cs has a component of global fallout. Given the high ^{137}Cs concentrations which are certainly traceable to Fukushima fallout, a global contribution can be assumed small, as can be the bias, as a consequence. For the samples analyzed here the bias is estimated below 0.1%.

The result found in this study is consistent with a Pu/Cs ratio reported by Imanaka et al. (2012)[21] for a highly contaminated place in the Fukushima zone as below 1 E-6. The authors have apparently used the same decomposition method for Pu as done here. Zheng et al. 2011[12] found $^{239+240}$Pu/^{137}Cs in soil, close to the NPP, as (3.6 ± 1.1) E-7 (only samples with ^{241}Pu>0 considered, and Fukushima contribution 87% to the sample "J-village, surface soil", as suggested by the authors), which is in good agreement with the results of this study.

4. Further discussion and conclusions

Of longer lived gamma radionuclides, Fukushima fallout appears to contain almost only ^{134}Cs and ^{137}Cs. ^{110m}Ag appears to be present, but in very low concentrations. Even much lower is the contribution of plutonium. ^{90}Sr (pure β) will be evaluated later for some of the samples. This radionuclide can also have (at least) two origins, which can however not be separated because no other isotope, like for Pu, is available for evaluation of contributions. The finding of essential absence long-lived gamma emitters other than ^{134,137}Cs confirms the results other studies (some of the ones quoted in Table 8 also deal with those). This makes a difference to Chernobyl fallout; the list of radionuclides given in Table 5 is motivated by Chernobyl experiences.

4.1. Cs ratio

Table 8 summarizes literature values of the ^{134}Cs/^{137}Cs ratio. References to European articles are not given individually, since most data are included in the evaluation in Kirchner et al. 2012[1]. The table also indicates whether summation correction for ^{134}Cs was performed; neglecting it can cause systematic errors of over -20% for measurement geometries close to the detector. In many cases it is however not known whether the labs considered the effect. For the values evaluated in Kirchner et al. 2012, it can be assumed that most do, since European radiometric labs are usually QA certified.

It appears that Cs ratios tend to be somewhat higher in Japan, particularly to the North of the NPP, than in the North America, in Europe, in Siberia and Southeast Asia. Differences in isotopic composition can be caused by different sources of release, i.e. different reactors or different parts of the cores. The Cs ratio reflects fuel burn-up which was not identical in all reactors and may even be different in different sections of one core. Since atmospheric conditions during release episodes were different in general, so were directions of the plumes and of fallout paths. There is no reason for assuming isotopic fractionation of Cs during transport or in the environment. Some discrepancies may be caused if no summation correction was applied for ^{134}Cs.

Further elucidation could be achieved by linking the data to trajectories of atmospheric dispersion which can in some cases be connected to certain release episodes. Whatever the physical reason, the Cs ratio found from the few samples of this study is higher than what has been found in Europe and North America, and slightly higher than most Japanese

Table 8. ^{134}Cs / ^{137}Cs ratios in Fukushima fallout reported in literature. All values decay corrected to 20 March 2011. Last column: indication whether summation correction for ^{134}Cs has been performed

author	Location	medium	^{134}Cs / ^{137}Cs ratio	sum. corr for ^{134}Cs ?
Amano et al. 2012[13]	Chiba (near Tokyo)	deposition	1.00	?
Bolsunovsky et al. 2011[14]	Siberia	rain, snow, air, pine	0.88 ± 0.23 (6)	?
Cleveland et al. 2011[15]	Canada	rain	0.76 ± 0.04 (6)	?
Diaz-Leon et al. 2011[16]	USA	air filters	0.77 ± 0.13 (6)	?
Endo et al. 2011[17]	Fukushima zone	soil	0.85 - 0.96	?
Fujiwara et al. 2012[18]	Fukushima zone	soil	1.005 ± 0.048 (1)	Y
Higaki et al. 2012[19]	Fukushima pref. & near Tokyo	bamboo leaves	1.17 ± 0.18	?
Hirose 2001[19]	around Japan	deposition	1.03 (1.0 - 1.1)	probably Y
ibd.[9]	near Tokyo	deposition	0.84 - 0.9	? (2)
Honda et al. 2012[20]	Pac. ocean, off Japanese coast	particles & plankton in sea water	1.08 ± 0.13 (6)	?
Imanaka et al. 2012[21]	Fukushima zone	soil	0.873 ± 0.071	?
Kato et al. 2011[22]	Fukushima pref.	top soil (3)	1.008	?
KEK (see[30])	Tsukuba (Ibaraki prefecture)	air filters	1.02 ± 0.24 (8)	?
Kim et al. 2011[23]	S Korea	air filters, rain water	0.939 ± 0.057 (6)	?
Kirchner et al. 2012[21]	Europe	air filters	0.874 (conf. int. 5% - 95%: 0.840 - 0.907)	probably Y
Long et al. 2012[24]	Vietnam	air filters	0.87 ± 0.16 (6)	?
MacMullin et al. 2012[25]	USA	air filter (7)	0.88 ± 0.14	?
Melgunov et al. 2012[26]	SW Siberia	rain, snow	0.90 ± 0.17 (6)	Y
Nomoshima et al. 2011[27]	Fukuoka (SW Japan)	air filters	1.02 ± 0.27 (4)	?
Tagami et al. 2011[28]	20 km S NPP	soil	0.89	?
ibd.[28]	Chiba (near Tokyo)	soil	0.87 ± 0.04	?
Taira et al. 2012[29]	Fukushima prefecture	soil	1.002 ± 0.022 (5)	?
Thakur et al. 2012[30]	USA	air filters	0.85 ± 0.12	Y
this study	**Koriyama, Fukushima city**	**soil, moss**	**1.062 ± 0.024**	**Y**

(1) calculated from the data given in the annex of the article.
(2) "gamma spectrometry problems" suspected as reason for lower values.
(3) calculated from the values of the two topmost soil layers, as given in the article.
(4) calculated from the data given in the article. One outlier omitted (5 May).
(5) calculated from the data of topmost soil layers, as given in the article.
(6) calculated from the data given in the article.
(7) Air filter from 20 March only.
(8) quoted from Thakur et al. 2012[30]; the original values to be found in http://legacy.kek.jp/quake/radmonitor/index-e.html .

Table 9. $^{239+240}Pu$ / ^{137}Cs ratios reported in literature

author	location	medium	$^{239+240}Pu$ / ^{137}Cs ratio (Fukushima) (10^{-7})
Imanaka et al. 2012 [21]	Fukushima zone	soil	< 10
Zheng et al. 2011 [12]	20 km S NPP	soil	3.6 ± 1.1 (1)
this study	Koriyama,Fukushima city	soil, moss	2.8 ± 2.0

(1) calculated from data in the article; samples "S2-Litter", "S3-Litter" and "J-village, surface soil" included. For the "J-village" sample, the authors consider 87% of the Pu as due to Fukushima.

results.

4.2. Pu to Cs ratio

To my knowledge no results on the Pu/Cs ratio have been published for the Fukushima prefecture, except some from the immediate vicinity of the damaged NPP. Results found in literature are summarized in Table 9. From Europe, only one result giving evidence of Fukushima plutonium exists to my knowledge. Lujanienė et al. (2012)[31] report higher Pu isotopic ratio α in air filters sampled during the passage of the Fukushima cloud, than could be expected for global and Chernobyl Pu (which are always present in minute concentrations in European air filters due to resuspension of soil particles). However the Pu/Cs (Fukushima) ratio, estimated about 2×10^{-4}, appears much too high, compared to results from Japanese samples, Table 9.

The results presented here are based on only 4 samples (3 for Pu), which are also not representative for the environment. Even if such detailed analyses contribute little to dose assessment which is of course the primary purpose of sampling in the region, they can still help to better understanding of the nature of the accident and its radio-ecological consequences. It is therefore suggested that similar detailed investigations be performed for a

larger number of samples from different locations in the region affected by the accident. And not least, they present training material for radiometric labs and for evaluation skills.

Acknowledgements

The colleagues of Hirosaki University, M. Hosada and A. Sorimachi, are gratefully acknowledged for providing the opportunity to visit the Fukushima zone and collecting the samples. The gamma-spectrometric analyses were performed by J.Witzani (Vienna), U.K. Schkade (Berlin) and A. Hubmer (Salzburg), who also carried out alpha-spectrometry.

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