

Radioactivity from Fukushima Dai-ichi in air over Europe; part 2: what can it tell us about the accident?

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ABSTRACT

It is shown which information can be extracted from the monitoring of radionuclides emitted from the Fukushima Dai-ichi nuclear power plant and transported to Europe. In this part the focus will be on the analysis of the concentration ratios. While ^{131}I , ^{134}Cs and ^{137}Cs were reported by most stations, other detected radionuclides, reported by some, are ^{95}Nb , $^{129\text{m}}\text{Te}$, ^{132}Te , ^{132}I , ^{136}Cs and ^{140}La . From their activity ratios a mean burn-up of 26.7 GWd/t of the fuel from which they originated is estimated. Based on these data, inventories of radionuclides present at the time of the accident are calculated. The caesium activity ratios indicate emissions from the core of unit 4 which had been unloaded into the fuel storage pool prior to the accident.

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1. Introduction

Emissions of radionuclides during the Fukushima accident were high enough to be detected all over the northern hemisphere. Due to decay of short-lived isotopes, to dispersion and to wet and dry deposition, however, atmospheric concentrations were effectively diluted and no longer posed a radiological risk to humans when the air masses reached other continents. It is our intention to demonstrate that these measurements are not only limited to illustrate air concentration time series, but might also be used to provide additional information valuable for emergency response. By using ^{131}I as an example, in part 1 of this article we presented a methodology to estimate its geographical distribution from air concentration measurements. This could be a useful tool for predicting spatial distributions of potential human exposures in nuclear emergency situations (Bossew et al., 2012).

The Chernobyl accident showed us that the nuclide composition of the source term, which is emitted during a severe reactor accident and dispersed within the atmosphere, can be used as a 'fingerprint' to derive information on the burn-up and decay time of the fuel prior to the accident (Kirchner and Noack, 1988) and on

the conditions of the fuel during the release phase (Powers et al., 1987). Compared to Chernobyl, the Fukushima Dai-ichi accident presents a much more difficult situation, since various reactor cores and spent fuel pools may have contributed to the emissions. Here, we show the inferences possible on (1) the conditions of the fuel during the release of the radionuclides detected in contaminated air masses in Europe and (2) on the fractions which may have originated from the spent fuel pools. In addition, inventories of radionuclides not detected, but of potential radiological concern such as the plutonium isotopes, are calculated.

2. Data and statistical methods

2.1. Data

The database compiled for the study described in part 1 (Bossew et al., 2012) is used. Many stations reported on radionuclide concentrations other than ^{131}I which will be evaluated here. Since ^{137}Cs airborne concentrations which were measured during the passage of the Fukushima Dai-ichi cloud are up to three orders of magnitude higher than the background level, no correction of the original data was necessary for our evaluation.

As we are not aware of any strontium and plutonium analyses from European air samples, the results given in Table 2 are derived from Japanese sources (MEXT, 2011a; TEPCO, 2011). All values

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Table 1
Fuel element specification and irradiation conditions used for the fuel inventory calculations of the Fukushima Dai-ichi units (NEI, 2004; IAEA, 2011).

Fuel element type	GE-11
• Assembly geometry	9 × 9
• Rods per assembly	72
• Active fuel length	3587.5 mm
• Pellet diameter	9.5 mm
• Clad thickness	0.7 mm
• Rod diameter	11.0 mm
• Rod-to-rod pitch	14.37 mm
Initial ²³⁵ U enrichment	3.8%
Power density	25 MW/t
Final burn-up	45 GWd/t
Irradiation	5 × 360 d
Downtimes	4 × 30 d

published until 9 May (Sr) and 30 May (Pu) were used. For strontium, media are soil, plant and sea water as reported by MEXT (2011a) and soil from the vicinity of the NPP (“Playground”, “Waste disposal site” and “Forest of wild birds”) and ocean bottom soil as given by TEPCO (2011). For plutonium, media are soil from the vicinity of the NPP (“Playground” and “Waste disposal site”) as reported by TEPCO (2011).

2.2. Estimation of radionuclide ratios

Radionuclide ratios were evaluated estimating the median of individual ratios.

Uncertainties and confidence intervals of the ratios were estimated by bootstrapping. The calculation of the median was carried out for a random fraction of data (here 50% is chosen in most cases; 70–80% if only few data are available, viz. the ^{129m}Te: ¹³²Te, ¹⁴⁰La: ¹³⁷Cs and ⁹⁵Nb : ¹³⁷Cs ratios), and repeated many times (here 200 simulations were chosen). The idea behind the approach is to estimate confidence limits in a situation where traditional least squares regression is not appropriate; the chosen method also allows accounting for individual data uncertainty and mitigates the influence of high leverage points and possible outliers. The confidence intervals are estimated from the distribution of the results, as well as the median reported in Table 2.

Data uncertainties were accounted for by adding a random Gaussian noise component to the data, which was independently sampled for each simulation. If available, uncertainty was taken from the reported data. Otherwise a reasonable default value (Bossew et al., 2012) was used. Its choice had little effect on the result. For example, for ¹³¹I (particulate) 21% was used as default

Table 2
Radionuclide ratios in air samples over Europe, referenced to 20 March 2011; the statistics are for 800 simulations of the ratio median (see text); 5% and 95%: estimated confidence limits, ‘med(ratio), raw’: median of the individual sample ratios, p level: Significance with *p* < 0.05 (1-sided) labelled with an asterisk.

	Number of samples	Median	5%	95%	<i>p</i> level	Med (ratio), raw
¹³¹ I (part.): ¹³¹ I (gas)	136	3.71	3.19	4.36	<0.001*	3.94
¹³¹ I (part.): ¹³⁷ Cs	862	28.6	27.3	29.8	<0.001*	28.5
¹³⁴ Cs : ¹³⁷ Cs	393	0.874	0.840	0.907	<0.001*	0.857
¹³⁶ Cs : ¹³⁷ Cs	38	0.132	0.099	0.164	<0.001*	0.120
¹³² Te : ¹³⁷ Cs	121	2.45	2.21	2.75	<0.001*	2.55
^{129m} Te : ¹³² Te	14	0.46	0.30	0.73	0.002*	0.460
⁹⁵ Nb : ¹³⁷ Cs	10	0.9	0.5	2.3	0.37	0.69
¹⁴⁰ La : ¹³⁷ Cs	8	0.2	0.04	0.4	0.91	0.09
⁸⁹ Sr : ⁹⁰ Sr	34	11.2	8.9	12.2	<0.001*	10.1
²³⁸ Pu : ²³⁹⁺²⁴⁰ Pu	20	2.28	1.98	2.58	<0.001*	2.14

standard deviation, which is the 75% percentile (deliberate choice) of the reported uncertainties.

Significance is checked by testing against a null hypothesis of randomness. The idea is that a ratio *y/x*, estimated as *median{y/x}*, only makes sense if the dispersion of the ratio is small compared to the one resulting from randomly distributed data. Technically, we reshuffled the associations between *x* and *y* at random, thus creating an artificial set with the same individual distributions of *x* and *y*, but with different joint distributions. Estimation of the median was repeated 800 times. For the original and each of the randomised data sets *i* the relative quartile deviations, Δ*Q_i*, as measure of dispersion of the median was calculated as

$$\Delta Q_i = \frac{Q_{75}(\frac{y}{x}) - Q_{25}(\frac{y}{x})}{\text{median}(\frac{y}{x})} \tag{1}$$

where *Q₂₅*, *Q₇₅* denote the lower and upper quartile of the ratios. The empirical distribution *F*(Δ*Q*) of the Δ*Q_i* of the randomised data was built. As a test for significant deviation from the null hypothesis of the data points to be randomly distributed in the (*x,y*)-plane, and the median ratio therefore being meaningless, Δ*Q* of the original data was compared to the distribution. To reject the null hypothesis that the concentration ratio for the real dataset is an artefact, its Δ*Q* must be in the critical region of the distribution of the ones derived from the randomised values *F*(Δ*Q*).

2.3. Fuel inventory calculations

Simulations of the build-up and decay during irradiation and storage of the Fukushima Dai-ichi reactor fuel were performed using the ORIGEN-ARP module of the SCALE-5.1 code system (Gauld et al., 2006). ORIGEN-ARP uses pre-processed burn-up dependent cross-section libraries generated for a variety of reactor types, fuel element geometries and irradiation histories. These include a boiling water reactor using GE-11 fuel elements, as the Fukushima Dai-ichi units 1–4. The core characteristics and irradiation conditions assumed for our simulations are summarised in Table 1. They are derived from the information given in the IAEA Power Reactor Information System on the Fukushima Dai-ichi units prior to the accident (IAEA, 2011). No attempt has been made to reconstruct the detailed irradiation histories of the units prior to the accident, since differences compared to the inventories resulting from the generic irradiation histories used (Table 1) are expected to remain low, in particular for activity ratios of short-lived isotopes. One of the cores (unit 3) has been operated with 6% mixed oxide fuel using low enriched plutonium as fissile material. Compared to neutron-induced fission of ²³⁵U there are some differences in fission yields of ²³⁹Pu. These can be disregarded, however, due to both the small fraction of mixed oxide fuel elements and to our interest in isotope ratios, as the ratios of fission yields of various isotopes of a fission product element are almost identical for both fissile elements.

It should be noted that in boiling water reactors neutron energy spectra, and thus radionuclide build-up within a fuel element, show considerable radial and axial variations which are caused by the differing initial ²³⁵U enrichments of the fuel rods and by the axial decrease of the moderator density. Although the cross-section libraries available in ORIGEN-ARP had been generated by a two-dimensional model which takes these radial and axial variations into account, the code calculates mean fuel inventories. This simplification, however, does not pose a limitation for our application, since the radionuclides emitted into the atmosphere are most likely to originate from vaporisation of major parts of the affected cores and thus present a core average.

3. Results and discussion

3.1. Radionuclide ratios

The evaluated data have been sampled between 25 March and the end of April. Decay correction to the reference date of 20 March 2011, 0:00 has been applied. This date is a deliberate choice. Extensive surveys of radiocaesium concentrations in the most affected prefectures around the Fukushima Dai-ichi site (MEXT, 2011b; Tagami et al., 2011) found $^{134}\text{Cs} : ^{137}\text{Cs}$ ratios in Japanese soils which are very close to the values given in Table 2 detected in the atmosphere in Europe in spring 2011.

Scatter plots of the most relevant isotope ratios, including the lines corresponding to the ratio and the confidence limits, are shown in Figs. 1 and 2. The ratios show small to moderate scattering only, with the exception of the values of the ^{95}Nb and ^{140}La to ^{137}Cs ratios. These must be taken with care as the test indicates no significant difference from random.

None of the ratios exhibit a significant temporal trend, although those ratios including ^{137}Cs show a slight increase at the beginning. An example is given in Fig. 3. During the first days that radionuclides from the Fukushima Dai-ichi nuclear accident were observed in Europe, the concentrations were low; with an annual average of about $0.4 \mu\text{Bq m}^{-3}$, the values for ^{137}Cs include a small, but varying background contribution (Bieringer et al., 2009). During the first days, therefore, ^{137}Cs measurements are likely to overestimate the air concentration originating from the Fukushima Dai-ichi site.

3.2. Release conditions

Inferences on the fuel burn-up and the decay time before the release of radionuclides during a nuclear accident require the analysis of ratios of isotopes belonging to the same chemical element, as these show identical release fractions and thus reflect the ratios which were present in the core at the time of the accident. To be useful for such an analysis, either one of the following conditions has to be met: a) if two isotopes show half-lives long enough that during irradiation both have not attained equilibrium between generation and decay and are produced via different pathways, their ratio strongly depends on the burn-up of the fuel, but is quite insensitive to moderate decay times after the chain

reaction ceased. b) If at least one of the two isotopes has a half-life short enough to attain its equilibrium concentration during irradiation, their ratio is strongly affected by decay time after irradiation.

From the list of the isotope ratios calculated for the Fukushima Dai-ichi emissions (Table 2), only $^{134}\text{Cs} : ^{137}\text{Cs}$ can be used for burn-up establishment: their half-lives (^{134}Cs : 2.04 y, ^{137}Cs : 30.1 y) are long enough, and, other than the direct fission product ^{137}Cs , the build-up of ^{134}Cs is mainly by neutron capture of ^{133}Cs generated during reactor operation. The decay time of the emitted radionuclides prior to the accident indicates whether emissions are from the cores in operation at the time of the earthquake or from the fuel stored on-site. It can be deduced from the $^{136}\text{Cs} : ^{137}\text{Cs}$, $^{129\text{m}}\text{Te} : ^{132}\text{Te}$ and $^{89}\text{Sr} : ^{90}\text{Sr}$ activity ratios given in Table 2, since each of these includes at least one short-lived isotope.

As Fig. 4 indicates, the median $^{134}\text{Cs} : ^{137}\text{Cs}$ activity ratio present in the Fukushima Dai-ichi fallout corresponds to a burn-up of 26.7 GWd/t with a 90% confidence interval between 25.4 and 28.0 GWd/t, which has been calculated from the uncertainties quoted in Table 2. Since at the end of each operation cycle approximately 20% of the fuel elements are replaced by fresh fuel, a mean burn-up of 26.7 GWd/t represents a core close to the end of its fuel cycle. One can, therefore, conclude that the released radionuclides originate from any of the cores of either units 1–3 which had been in operation for several months at the time of the accident or from unit 4 which already had been transferred into the storage pool for fuel replacement. The irradiated fuel elements which after reaching their final burn-up had been replaced and stored in the pools of units 1–3 can be excluded as a major source of atmospheric release, since at the final reference burn-up of 45 GWd/t they would show a $^{134}\text{Cs} : ^{137}\text{Cs}$ activity ratio of approximately 1.3 (Fig. 4). Deviations from the generic irradiation history used could affect our result mainly due to decay of ^{134}Cs in case of longer shutdown periods for maintenance and exchange of fuel elements. The estimated burn-up of 26.7 GWd/t will be modified by approximately +1.0 GWd/t per additional month of shutdown.

For the mean core burn-up of 26.7 GWd/t established from the observed $^{134}\text{Cs} : ^{137}\text{Cs}$ activity ratio, activity ratios calculated by ORIGEN-ARP of those elements which include a short-lived radionuclide are given in Fig. 5 for varying decay times after the irradiation ceased. Comparison with the activity ratios observed in Europe (Table 2) indicate decay times (90% confidence level) of 11–16 d ($^{129\text{m}}\text{Te} : ^{132}\text{Te}$) and of 13–23 d ($^{136}\text{Cs} : ^{137}\text{Cs}$), respectively. Taking into account that the ratios given in Table 2 refer to March 20, decay times derived from the Te isotope ratios support the assumption that the emissions originate from units 1–3, which were shut down immediately after the earthquake. Part of the radiocaesium, however, seems to have been emitted by fuel with a longer decay time. This indicates that a significant fraction of the caesium may originate from the core of unit 4 which had been unloaded into the fuel storage pool after reactor shutdown on Nov. 29, 2010 (NISA & JNES, 2011). In this fuel, more than 90% of the inventories of the two tellurium isotopes had been decayed before the initiation of the accident (see also Table 3). Potential emissions of these semi-volatile tellurium isotopes from the unloaded core of unit 4 thus were low and did not significantly shift the activity ratio reflecting their inventories in the cores of units 1–3.

The $^{89}\text{Sr} : ^{90}\text{Sr}$ ratios are more difficult to interpret. The strontium measurements reported by the Japanese Government (Government, 2011) give a median ratio of 8.7 (reference date: March 20, 2011), which corresponds to a decay time of 9 d, albeit with considerable uncertainty (90% confidence interval: 0–35 d). However, taking into account all measurements that we are aware of shifts the ratio to a median of 11.2 at March 20, 2011, which is higher than the ratio calculated at shutdown (Table 3).

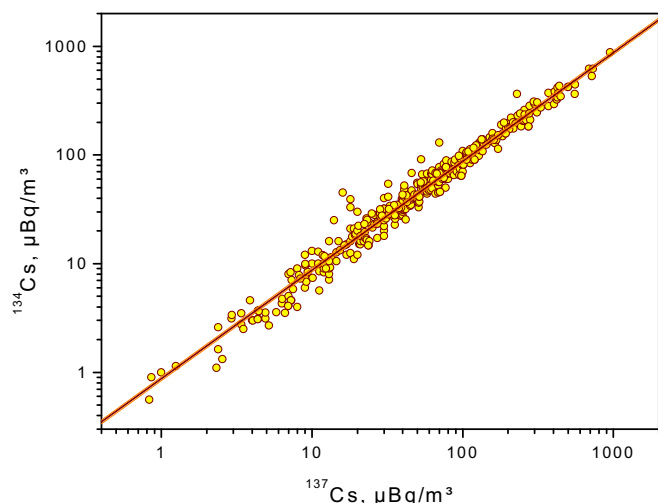


Fig. 1. Scatter plot of activity concentrations of ^{134}Cs and ^{137}Cs and line corresponding to the estimated median ratio; the 95% confidence band is hardly visible, as it is close to the median; reported uncertainties are about 5% (median, range 2–30%) for ^{134}Cs and about 7% (median, range 2–50%) for ^{137}Cs .

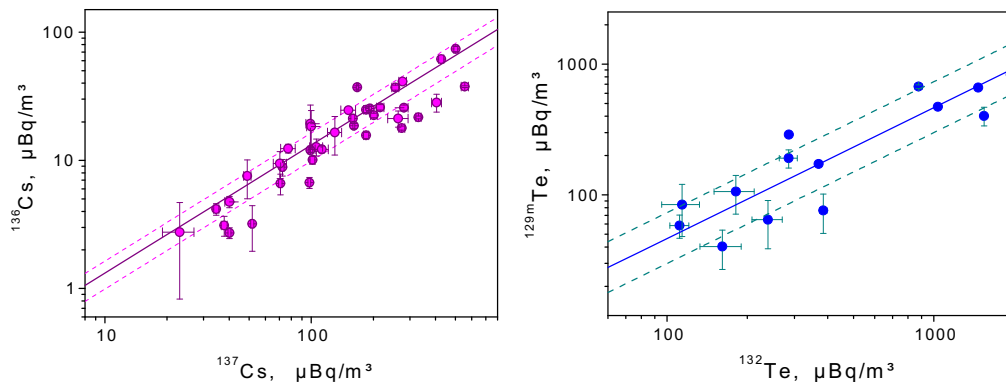


Fig. 2. Scatter plots of activity concentrations of ^{136}Cs and ^{137}Cs (left), and of $^{129\text{m}}\text{Te}$ and ^{132}Te (right), and lines corresponding the estimated median ratio (with 95% confidence bands).

This indicates either an overestimation of ^{90}Sr or an underestimation of ^{89}Sr in fuel at the time of the accident. Since both ^{89}Sr and ^{90}Sr are direct fission products with well known fission yields, their calculated inventories should not show systematic errors.

The apparent discrepancy could be explained by strontium originating from fuel fragments representing the specific burn-up of the affected fuel rather than by the core average given in Table 3 and Fig. 5. A major part of the strontium and plutonium emitted during the Chernobyl was present in fuel particles (Kashparov, 2003) which had been fragmented and dispersed by the initial steam explosion caused by the power excursion. Although such a violent explosion did not occur at the Fukushima Dai-ichi site, it can not be excluded that some fuel particles have been dispersed by the three hydrogen explosions which happened during the first days after the earthquake. Strontium-90 builds up linearly with burn-up. Concentrations of the short-lived ^{89}Sr (50.5 d half-life) become almost saturated after approximately 150 d of reactor operation and do no longer increase with burn-up. Its concentrations at saturation, however, are proportional to local power densities within the core, as these reflect the local neutron flux densities and, as a consequence, the neutron-induced fission rates. Thus, lower than average inventories of ^{90}Sr are present in fuel fragments with burn-up <26.7 GWd/t, higher than average ^{89}Sr will be expected in fuel particles which have been exposed to high local power densities. The possibility that the strontium originates

from fuel fragments exhibiting a lower than core average burn-up can be excluded, since it is at variance with the $^{238}\text{Pu} : ^{239+240}\text{Pu}$ ratio present in these samples which indicate a higher than average burn-up (see below). Higher than average ^{89}Sr concentrations may be explained by the actual fuel loading schemes and operation in the cycle prior to the accident. However, we could not get any information about this. Similarly, the presence of fuel fragments in the area around the Fukushima Dai-ichi site is still an open question.

The activity ratio of the alpha-emitting plutonium isotopes traditionally is used to distinguish weapons' testing fallout plutonium with its $^{238}\text{Pu} : ^{239+240}\text{Pu}$ ratio of approximately 0.03 (Kirchner et al., 2009) from other sources. For the average burn-up of emissions from the Fukushima Dai-ichi site, a $^{238}\text{Pu} : ^{239+240}\text{Pu}$ activity ratio of 1.9 is expected (Table 3). The median ratio of 2.28 (Table 2) calculated from values measured in soil samples close to the Dai-ichi nuclear power plant units corresponds to a burn-up of 30.8 GWd/t (90% confidence interval: 28.4 to 33.0 GWd/t). This deviation from the mean core burn-up indicates that plutonium may be associated with fuel fragments that hypothetically could have been released as a consequence of the hydrogen explosions.

Table 3 summarises the inventories of a number of radionuclides of potential radiological concern for the estimated mean fuel burn-up of 26.7 GWd/t. It includes various isotopes which are not detectable by gamma spectroscopy and thus may assist in evaluating potential radiation exposures due to emission of these nuclides. For the short-lived radionuclides, the inventories given in

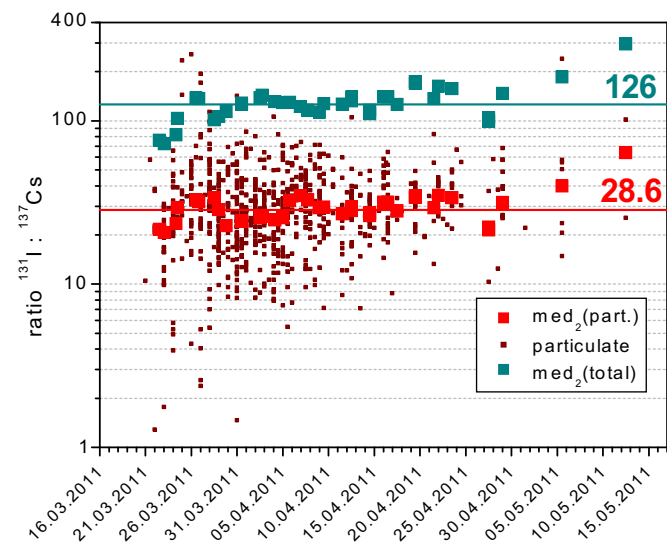


Fig. 3. Temporal variability of the $^{131}\text{I} : ^{137}\text{Cs}$ ratios. Horizontal lines and figures: overall medians. med₂: sliding medians over 2 consecutive days.

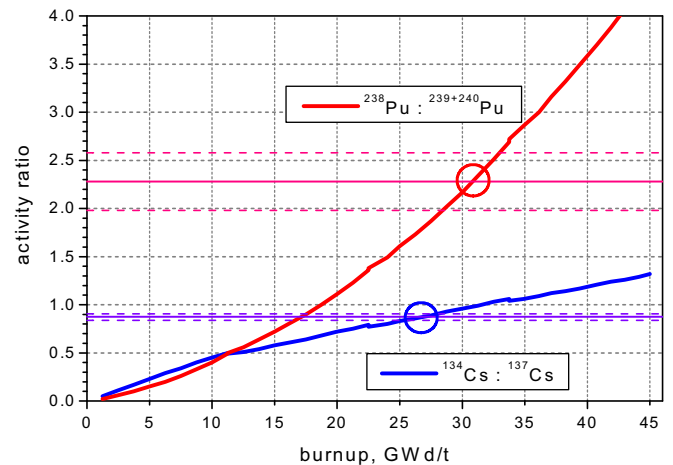


Fig. 4. Burn-up-dependent variation of the $^{134}\text{Cs} : ^{137}\text{Cs}$ and $^{238}\text{Pu} : ^{239+240}\text{Pu}$ activity ratios as calculated for fuel of the Fukushima Dai-ichi units. Horizontal lines: medians and confidence bands according to Table 2.

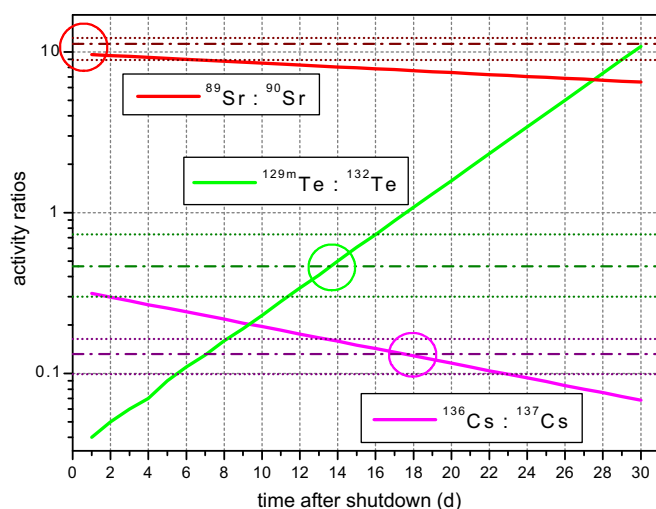


Fig. 5. Variation with time after reactor shutdown of activity ratios including a short lived isotope of the element considered; dot-dashed lines denote the median ratios, dotted lines the 90% confidence intervals calculated from the atmospheric measurements (Table 2).

Table 3 after 105 d decay are specific for the core of unit 4 unloaded into the fuel storage pool.

As discussed by Powers et al. (1987) and Feuerstein (1987), activity ratios of isotopes of different chemical elements can be useful to derive information on the situation of the nuclear fuel and thus on the accident mechanisms at the time of their release. The information available from our analyses is summarised in Table 4. All activity ratios and release fractions are given relative to ^{137}Cs . Obviously, release fractions are high for the volatile elements I, Te (and Cs), but in contrast to the Chernobyl fallout only a few

Table 3

Calculated inventories of major radionuclides in Fukushima Dai-ichi fuel with 26.7 GWd/t burn-up at reactor shutdown and after 105 days decay.

Nuclide	Half-life	Inventory [Bq/t]	
		Shutdown	105 d decay
^3H	12.3 a	1.6×10^{13}	
^{60}Co	5.27 a	4.1×10^{14}	
^{89}Sr	50.5 d	2.5×10^{16}	5.9×10^{15}
^{90}Sr	28.8 a	2.5×10^{15}	
^{95}Zr	64.0 d	4.3×10^{16}	1.4×10^{16}
^{103}Ru	39.3 d	3.6×10^{16}	5.7×10^{15}
^{106}Ru	374 d	1.1×10^{16}	9.3×10^{15}
^{129}I	1.57 E7 a	7.3×10^8	
^{131}I	8.02 d	2.5×10^{16}	3.0×10^{12}
$^{129\text{m}}\text{Te}$	33.6 d	1.2×10^{15}	1.4×10^{14}
^{132}Te	3.2 d	3.5×10^{16}	7.0×10^{16}
^{134}Cs	2.06 a	2.8×10^{15}	2.6×10^{15}
^{136}Cs	13.2 d	1.0×10^{15}	4.1×10^{12}
^{137}Cs	30.1 a	3.2×10^{15}	
^{140}Ba	12.8 d	4.5×10^{16}	1.5×10^{14}
^{140}La	1.68 d	4.8×10^{16}	1.7×10^{14}
^{144}Ce	285 d	3.3×10^{16}	2.6×10^{16}
^{235}U	7.04 E8 a	1.2×10^9	
^{238}U	4.47 E9 a	1.2×10^{10}	
^{239}Np	2.36 d	4.1×10^{17}	2.4×10^{11}
^{238}Pu	87.7 a	4.2×10^{13}	
^{239}Pu	2.41 E4 a	9.7×10^{12}	
^{240}Pu	6560 a	1.4×10^{13}	
^{241}Pu	14.4 a	2.8×10^{15}	
^{242}Pu	3.73 E5 a	3.3×10^{10}	
^{241}Am	432 a	3.6×10^{12}	4.9×10^{12}
^{243}Am	7370 a	2.3×10^{11}	
^{242}Cm	162 d	6.9×10^{14}	4.5×10^{14}
^{244}Cm	18.1 a	1.6×10^{13}	

Table 4

Activity concentrations relative to ^{137}Cs in fuel, measured in Europe (Table 2), and their corresponding release ratios.

Activity ratio	Fuel ^a	Atmosphere (median)	Release ratio
^{131}I (total): ^{137}Cs	3.6	94	26
^{132}Te : ^{137}Cs	1.6	2.5	1.5
^{95}Nb : ^{137}Cs	12.9	0.9	0.07
^{140}La : ^{137}Cs	9.9	$\sim 0.2^b$	$\sim 0.02^b$

^a 26.7 GWd/t burn-up.

^b Estimated fallout isotope ratio highly uncertain.

refractory elements are present. This could indicate that the fuel temperatures remained well below 2400 °C which were reached during the Chernobyl core heat-up phase. At such temperatures releases of refractory elements (Sr, Mo, Zr, Ru, Ba) still are limited to a few percent when compared to those of caesium (Powers et al., 1987; Feuerstein, 1987). Therefore we can not exclude that due to atmospheric dispersion, the concentrations in Europe of such isotopes have decreased so much that they were no longer detectable.

4. Conclusions

The analyses of activity ratios of the emissions from the Fukushima Dai-ichi nuclear accident which were transported into air over Europe allow to estimate the mean core burn-up and decay time of the fuel from which they originated. The isotopic composition observed in Europe corresponds to emissions from a core with an effective burn-up of approximately 26.7 GWd/t being in operation until the date of the earthquake, i.e. from units 1 to 3. Emissions from the core of unit 4 which had been unloaded prior to the accident are confined to volatile radionuclides.

In cases of severe nuclear accidents the methodology proposed here may allow to use measurements of environmental radionuclides for getting information on the on-site situation which may be difficult to receive otherwise.

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