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Relationships between air dose rates and radionuclide concentrations in agricultural plants observed in areas affected by the Fukushima Dai-ichi accident

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ABSTRACT

Most of environmental monitoring programs include measurements of the air dose rates and the radionuclides activity concentration in plants. Both these parameters depend on deposition density of radionuclides. Therefore, measurements of one parameter can (with some supplementary information) be used as an indicator for the other parameter. After the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, the Emergency Operation Centre (EOC) operated by the Environmental Radioactivity Monitoring centre of Fukushima and Ministry of Agriculture, Forestry and Fisheries (MAFF) of Japan carried out large sampling programme over different distances from the NPP. The sampling programme was focused on the usage of the weed leaves as a proxy for the prediction of radionuclide transfer to some cultivated plants. The MAFF monitoring programme in 2011–2016 was addressed mainly to agricultural crops. In both cases, the air dose rates were measured at the sites of the sampling. The paper addresses the assessments of relationship between radionuclide activities concentrations in plants and ambient dose rates. The time—dependent relationships were quantified based on weed, buckwheat, brown rice and soybean data obtained in 2011–2016. The recommendations on optimizing emergency sampling programmes based on use of the data of ambient dose rates are also presented.

1. Introduction

Large-scale nuclear accidents present big challenges for authorities responsible for the protection of humans and the environment. Lesser known, but just as pertinent, are the challenges in remediation of affected agricultural lands. The Fukushima prefecture is renowned for its high quality agricultural products and the remediation of the contaminated areas in the prefecture is often considered as a top priority in the recovery phase. Monitoring of agricultural lands is a key activity that provides necessary information for emergency response and remediation planning of contaminated environments (Fesenko et al., 2006).

The air (ambient) dose rate[1] measured at the height of 1 m above top soil in agricultural land is an important parameter that can be used for such a purpose. Air dose rates are easily measurable, readily available during a nuclear or radiological emergency. In case of lack of capacities to provide measurements of radionuclide activity concentrations in the soils and crops, the data on air dose rate can be used for prompt decisions on emergency response in agriculture (Khomutinin et al., 2020a, 2020b). Another advantage is that the air dose data integrate quants coming from rather large area (with radius of 50–100 m) whereas point samples representative only for the small areas where samples were taken.

Shortly after the accident, a large sampling programme of wild plants (the weed leaves) was initiated out by the Emergency Operation Centre (EOC) operated by the Environmental Radioactivity Monitoring centre of Fukushima. Soil and vegetation samples in different locations at the different directions and distances from the Fukushima Dai-ichi NPP were collected and measured (Onda et al., 2015, 2020). Further, in 2012–2017, MAFF organized a sampling campaign for agricultural crops on selected agricultural fields. The soil, weed and crop sampling was supplemented with air dose rate measurements. Some data on the weed contamination, used in this research, were also analysed by Renaud et al. (2012) and Onda et al. (2020). In particular, Renaud et al. (2012) provided a discussion of the site selection and the data for study site, whereas Onda et al. (2020) provides more details of the sites and the sampling campaign. However, no data on radionuclides half-lives of radionuclide concentrations in the weed leaves were provided based on these data. Therefore, data evaluation and analysis presented here are different from those presented in the above publications.

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[1] For purposes of this paper, the terms "air dose rate" and "ambient dose rate" are used interchangeably.

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The IAEA introduced a set of operational intervention level (OILs) based on data that are readily available during a nuclear or radiological emergency. Two of such operational intervention criteria directly addressed to the contamination of agricultural produce. In particular, OIL3γ that is equal 1 μSv/h air gamma dose rate above background recommends the public to stop consumption, distribution and sale of non-essential local produce, until the activity concentrations have been assessed by using operational intervention criteria based on the ^{131}I and ^{137}Cs concentrations in the food products (IAEA, 2017).

At the early stage after the deposition when most of radionuclides can be associated with surface of the plants, the ambient dose rate reflects contamination of soil and plants, especially at the early stage after the deposition when most of radionuclides can be associated with surface of the plants. At the later stage, both the plant contamination and air dose rates are assumed proportional to the radionuclide activity in the soil. Therefore, the relationship between activity concentration in plants and the air dose rate can be speculated. The relation between radionuclide concentrations in the soil or plants and the air dose rate values is time-dependent and depends on many other factors including radionuclide depth distribution in the soil. These constrains in use of air dose data should also be considered.

The Ministry of Agriculture, Forestry and Fisheries (MAFF), reported that more than 96% of the total amount of Cs remained in the 0–5 cm of the top soil of paddy fields (MAFF, 2013), whilst Yamaguchi et al. (2015) reported that for virgin, undisturbed soil, most of the radiocaesium was retained within the 0–0.5 cm soil layer. It is also reported by that 71 out of 82 sites showed that the radioactive Cs downward migration was limited within 5 cm (Matsuda et al., 2015). Similar data for the early stage after the depositions were obtained in numerous studies carried out in the aftermath of the Chernobyl accident (Alexakhin et al., 1996; Prister et al., 1996; IAEA, 2006; Almgren and Isaksson, 2006; Fesenko et al., 2006Alexakhin et al., 2007) and in some research performed after the global fallout (IAEA, 2009).

Measurement of the ambient dose rate is an important step in sampling of both soil and plants (Khomutinin et al., 2020a, 2020b) because the dose rate can be, with some uncertainty, used for reconstruction of the soil contamination (Takata et al., 2014, 2015a,b). This uncertainty can be low in the case of the known geometry of irradiation especially if the area of concern is cleared from vegetation, depth distribution is known, and biomass of plants is not very high. Additionally, the air dose rate data can be applied to identify areas where food monitoring is not of importance or can be used for delineating highly contaminated areas where additional sampling of agricultural produce is required.

The objective of the paper was to analyse the relationships between radionuclide activity concentrations in wild plants, soil and air dose rate depending on time lapsed after the Fukushima Dai-ichi accident to investigate a possibility to use this relationship in delineation of areas with different crop contamination. The assessments presented in this paper are based on data of radionuclide concentrations in the weed and brown rice in 2011 provided by Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the Fukushima Agricultural Technology Centre, respectively. Data from 2012 to 2017 of brown rice, soybean and buckwheat in selected areas of Fukushima prefecture were used to define the relationships between the air dose rates and radiocaesium activity concentrations in the soil and plants at the time of harvest. Additionally, the feasibility using air dose rate as an indicator of the necessity for food monitoring during emergencies were evaluated.

2. Materials and methods

2.1. Sampling of weed and site description

The EOC began to collect the soil and weed samples as early as March 18, 2011 and continued until March 2013 (Onda et al., 2020). The weed species were of the Rosette plants family, including *Rumex obtusifolius*, *Dandelion, Plantain, Goldenrod, Papaver dubium* (NARO, 2015). The locations of the sampling sites ranged from 24 to 84 km from the FDNPP (Onda et al., 2020) (Table 1). The data obtained by this programme were presented on the Nuclear Radiation Authority (NRA) website (Ministry of Education, Culture, Sports, Science and Technology, 2012a, b).[2] The data presented at the NRA website were organized as a set of individual rows (or lines) with unique information. Each line consists of the site attributes, sampling date/time, the air dose rate and radionuclide concentrations in the soil and plants sampled together during that sampling act. Altogether, the website provides 776 rows for the March–May and 416 rows for the June–December sampling campaign (MEXT, 2012a; MEXT, 2012b). The air dose rates were determined at a height of 1 m above ground with the aid of a CsI (TI) scintillation detector from March to December 2011 (Onda et al., 2015).

Detailed information on the weed development (biomass of leaves, fraction of died and newborn biomass) was not available. However, as the area just entered the spring season, it can be assumed that newly emerged weed leaves may appear from April onwards. Both the top soil and existing plant biomass were contaminated directly at the time of deposition. After the deposition on surface of vegetation, a part of radionuclides are lost from plants due to removal by wind and rain, either through leaching or by cuticular abrasion. The increase of biomass during growth does not cause a loss of activity; but it does lead to a decrease in activity concentration due to effective dilution. These processes should be considered to explain the uncertainty in relationship between air dose rate and radionuclide concentration in the plant.

Activity concentrations of ^{131}I, ^{134}Cs, and ^{137}Cs were measured by γ-spectrometry in the fresh weed levels samples taken from March to May 2011, whereas data for ^{134}Cs and ^{137}Cs in the fresh weed leaves samples were measured until December 2011. Radionuclides other than ^{131}I, ^{134}Cs, or ^{137}Cs were also detected in a few samples (NARO, 2015). These data were not presented in the MEXT database MEXT (2012a, 2012b).

2.2. Radionuclides in crops and external dose rate in 2011–2016

Most of the crops were not being sown yet at the time of the direct Fukushima depositions, and thus these crops were not affected by direct contamination of the edible parts of plants. In 2011, it was not possible to investigate the radionuclide concentrations in all fields before the rice

Table 1
Characteristics of the sampling sites near the Fukushima Daiichi NPP in 2011 (Onda et al., 2020).

Site ID	Distance from the FDNPP	Date of depositions	Start of sampling	Contamination density with ^{137}Cs, kBq/m^2
2–1	42 km North/West	2011/03/16	2011/03/18	961
2–2	45 km North/West	2011/3/16	2011/3/18	168
2–3	41 km West	2011/3/16	2011/3/18	20
2–4	24 km North	2011/3/12	2011/3/18	67
2–5	39 km West/South/West	2011/3/15	2011/3/18	24
2–6	43 km South/South/West	2011/3/15	2011/3/18	28
2–7	34 km West/North/West	2011/3/16	2011/3/25	459
2–8	50 km North/West	2011/3/16	2011/3/25	212
2–9	57 km West/North/West	2011/3/16	2011/3/26	214
4–3	60 km West/North/West	2011/3/16	2011/4/7	100

[2] http://radioactivity.nsr.go.jp/ja/list/483/list-1.html.

cultivation. Therefore, only representative fields based on expert judgments were selected, and the radioactivity of the soil was determined. When any rice bag exceeded the provisional regulation value,[3] thorough investigation of the soil survey on each field in the area was carried out. Therefore, data of radionuclide activity concentrations in crops at the time of harvest and corresponding data on air dose rate for 2011 are rather limited. The only information available is the report from Fukushima prefecture that presented some data for radionuclide activity concentrations in brown rice and in the soil (Fukushima prefecture, 2011). The corresponding database consists of 99 values on radiocaesium in brown rice samples and 99 results of the air dose rate measurements in the fields where the rice was produced.

In 2012, the MAFF started a large-scale sampling programme for assessing radiocaesium activity concentrations in crops, namely, in rice, wheat, soybean and buckwheat produced in the Fukushima prefecture. The air dose rates were also measured during sampling at each sampling site. The data used for assessments are given in Table 2.

Altogether 1151 values on radiocaesium in crops, 1287 values for radiocaesium in the soil and air dose rate values were initially used to assess relationships between radiocaesium concentrations in plants, soils and ambient dose rates.

2.3. Quality control and data processing

The data for each sampling point included the information on radiocaesium and radioiodine (for the first two month after the deposition) concentrations in the weed leaves, the air dose values and other fields attributes, such as the field location and the sampling date. The rows (lines) that had uncomplete sets of attributes, such as missed data were eliminated from the further analysis. Therefore, the number of the data for every sampling point used for further assessments were normally less than that of given for same sampling points published in the MEXT database (2012a, 2012b). In particular, in 135 samples, the concentrations of both caesium isotopes were not presented. In 41 samples ^{134}Cs was not detected, although ^{137}Cs was measured, and in 6 cases. ^{134}Cs was detectable but Cs^{137} was not (MEXT, 2012a, 2012b).

As the radionuclides activity concentrations in crops decreased with the time lapsed after the accident, the number of samples with ^{134}Cs and/or ^{137}Cs concentrations below detection limits increased. This was more the case for ^{134}Cs, which has a shorter half-life (2.07 years) than that of ^{137}Cs, (with a half-life of 30.17 years). Another challenge in the samples measurement was application for such a purpose of different detectors and dissimilar conditions of the measurements in same cases. Therefore, the detection limits assigned to the samples were not the same for all data obtained. To avoid bias in the assessments of the basic statistics related to the data, concentrations below the detection limits were removed from the dataset used for identification of the air dose rates and radiocaesium concentrations in crops (see Table 2). The ratio of ^{134}Cs–^{137}Cs in samples of soil and plants was served as an additional tool for data quality check as there is no disequilibrium in the environment of these isotopes.

The Microsoft Excel and Statistica 9.0 and Grapher 9.6 standard software packages were used for statistical analysis and graphical presentation of the data reported in the paper.

3. Results and discussion

3.1. Relationship between concentration of radionuclides in weed and air dose rate

I-131, ^{134}Cs and ^{137}Cs dominated in the depositions during the first weeks of the accident, whereas ^{134}Cs and ^{137}Cs also dominated afterwards, when most of the iodine had decayed (with a half-life of 8 days). The activity ratio of ^{134}Cs–^{137}Cs was close to 1.0 whereas for activity ratio of ^{131}I to ^{137}Cs was around 12.0. These values are different from those measured after the Chernobyl accident where activity ratios of ^{134}Cs–^{137}Cs was around 0,5 and those for ^{131}I to ^{137}Cs was around 20.

Information on correlation of ^{131}I, ^{134}Cs and ^{137}Cs concentrations in the weed leaves can contribute to an assessment of the environmental contamination in areas where data for all dose-forming radionuclides are missing. Correlation analysis applied to all the databases showed that correlation coefficient of ^{131}I in relation to ^{137}Cs amounted to 0.56; whilst correlation coefficient of ^{134}Cs in relation to ^{137}Cs was nearly 1.0 (Fig. 1). These differences can be explained by different physicochemical properties of radionuclides affecting atmospheric dispersion of ^{131}I and radiocaesium and dry/wet deposition coefficients specific for the above radionuclides.

Table 2
General information on the data on radionuclide concentrations in crops used for assessments.

Year	Buckwheat		Rice		Soybean	
	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs	^{134}Cs	^{137}Cs
Number of samples with activity above minimum detectable level						
2012	17	21	51	86	39	42
2013	60		41	63	55	59
2014	19	32	50	117	32	50
2015	10	29	21	85	19	46
2016	14	31	27	103	18	43
Total number of samples						
2012	25		162		43	
2013	77		81		60	
2014	40		160		58	
2015	38		148		58	
2016	36		122		43	
All	216		673		262	

[1]136 samples of soil.

The main conclusion following these data is that there is a strong linear correlation between two caesium radioisotopes, and data for one isotope can be substituted by data for another one, considering differences in radioactive decays and dose conversion coefficients. The ^{134}Cs concentration can be calculated in any environmental media as:

$$Q(^{134}Cs) = Q(^{137}Cs) \times e^{-(\lambda_{134}-\lambda_{137})\times t} \quad (1)$$

[3] The Provisional Regulation Values for both radioiodine and radiocaesium were set by the Ministry of Health, Labour and Welfare on 17th March 2011 (shortly after the accident) as the temporary permissible levels in food and drinking water. For radiocaesium, these values were 200 Bq kg^{-1} for drinking water and dairy products and 500 Bq kg^{-1} for the other foodstuffs.

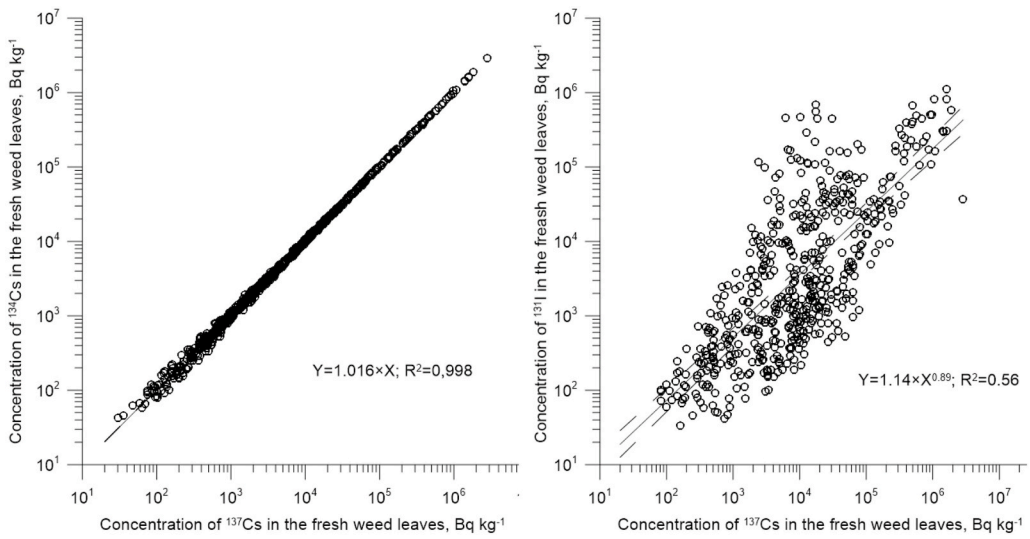

Fig. 1. Relationships between concentrations of ^{134}Cs and ^{137}Cs (left figure) and ^{131}I, and ^{137}Cs (right figure) in the weed leaves. Dashed lines represent 95% confidence interval. The data represent individual data points. Both Y-and X-axes are log-scaled.

where $Q(^{134}Cs)$ is the concentration of ^{134}Cs in the plants or the soil and $Q(^{137}Cs)$ is the concentration of ^{137}Cs in the corresponding environmental compartment, t is the time lapsed after the depositions. Another important point is that the linear correlation between concentrations of ^{131}I and ^{137}Cs was less strong as that for two caesium isotopes.

Major "emergency" radionuclides specific for the Fukushima accident, namely, ^{131}I, ^{134}Cs and ^{137}Cs, as well as natural radionuclides can contribute to the air dose rate. These radionuclides emit radiation of varying energies, providing different contributions to the external dose rate, even though their ambient concentrations are similar.

The average level of the terrestrial radiation in Japan before the Fukushima accident turned out to be about 0.056 μSv h^{-1} (Abe et al., 1980). Thus, natural radiation background can also contribute to the dose rate readings, especially at the sites with low deposition after the FDNPP accident (IAEA, 2015a). Indeed, the contribution of natural outdoor radioactivity can reach up to 30% at sites 2-3, 2-5, 2-6. Substantial contributions of natural radioactivity to the air dose rates (12–16%) can also be observed at sites 2-4, 2-10, 4-4.

As composition of major dose-forming radionuclides in the depositions is changes with time after the fallout, the effective radionuclide concentration (Q_e *(t)*) can be expressed as:

$$Q_e(t) = k_{131} \times q_{131}(t) + (k_{134} \times e^{-(\lambda_{134}-\lambda_{137})\times t} + k_{137}) \times q_{137}(t) \quad (2)$$

where Q_e is the effective radionuclide concentration, (Bq kg^{-1}), q_i – is the concentration of radionuclide in the weed leaves (fresh weight), (Bq kg^{-1}), k_i – are the contributions of the ^{131}I, ^{134}Cs and ^{137}Cs to the air dose rate (based on IAEA, 2001). Thus, the expression of:

$$(k_{131} \times q_{131}(t) + (k_{134} \times e^{-(\lambda_{134}-\lambda_{137})\times t} + k_{137}) \times q_{137}(t) \quad (3)$$

represents the effective concentration that reflects the contributions of major dose-forming radionuclide to the air dose rate.

$$k_i = \frac{DC_i}{\sum DC_i} \quad (4)$$

and can be considered as a proxy for air dose rate.Where DC_i is the dose conversion coefficient for the plane source, (μSv h^{-1})/(kBq m^{-2}). These values amount to 0.13 × 10^{-2}, 0.54 × 10^{-2} and 0.21 × 10^{-2} (μSv h^{-1})/(kBq m^{-2}) for ^{131}I, ^{134}Cs and ^{137}Cs, respectively (IAEA, 2001).

The examples of the relationship between the air dose rate and the

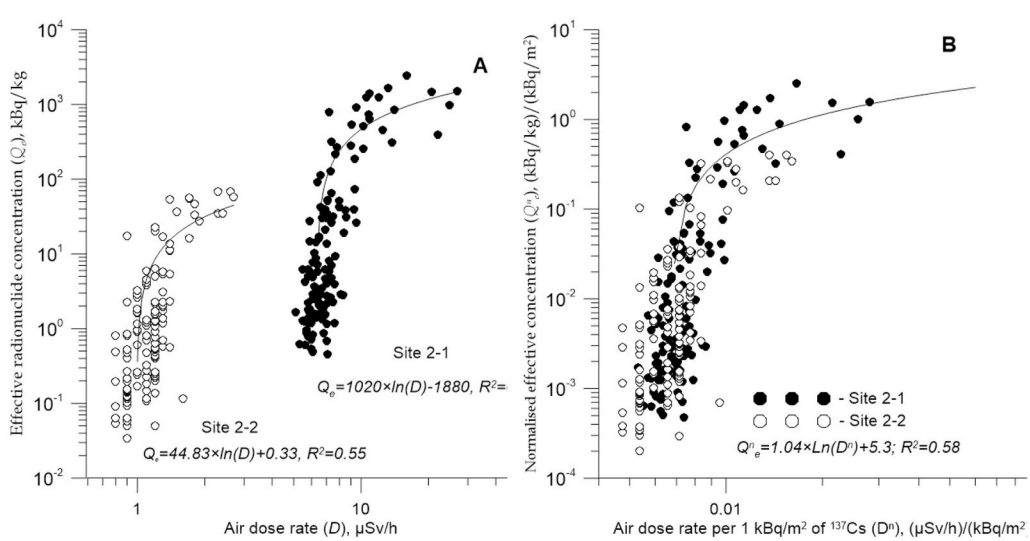

Fig. 2. Relationships between air dose rates and effective radionuclide concentrations in the fresh weed leaves measured at sites 2-1 and 2-2 (A). The same data normalised on the mean deposition densities of ^{137}Cs (kBq/m^2) (B). The data represent individual data points. Both Y-and X-axes are log-scaled.

effective radionuclide concentrations in the weed are shown based on the data of two sites (2-1 and 2-2) with different deposition levels of ^{137}Cs (see Table 1). The data for both sites were best approximated by the exponential fit with the coefficient of discrimination (R^2) of 0.59 for site 2-1 and 0.56 for site 2-2 (Fig. 2A). The data of Fig. 2A, normalised to the deposition densities of ^{137}Cs, namely, on 961 and 168 kBq/m,^{2} for sites 2-1 and 2-2, respectively, are given in Fig. 2B. It can be seen that the normalised data for two sites can be approximated by the same equation with similar parameters. Thus, these data demonstrate a possibility to use them for estimating the threshold level of the air dose rate above which the concentrations of radionuclides in plant can exceed not acceptable limit.

The values of the air dose, ($D(t)$, μSv h^{-1}), normalised to the effective concentration of radionuclides in the weed leaves ($R_d(t)$, (μSv h^{-1})/(Bq kg^{-1})) can be defined as follows:

$$R_d(t) = \frac{D(t)}{Q_e(t)} \tag{5}$$

$Rd(t)$ values, calculated based on the whole data set derived for all monitoring sites, are given in Fig. 3.

For March-December 2011, the time dependent ratios of the air dose rate and effective contamination can be fitted by the power function ($R^2 = 0.8$):

$$R_d(t) = a \times t^{1.6} \tag{6}$$

For March–May, the relationships ($R_d(t)$) can be also be well approximated ($R^2 = 0.92$) by the ordinary exponential equation:

$$R_d(t) = a \times e^{bt} \tag{7}$$

with the following parameters: $a = 2.0 \times 10^{-5}$ (μSv h^{-1})/(Bq kg^{-1}), and b – 0.0652 (d^{-1}) ($R^2 = 0.65$). The same equation is applicable also for most of other sampling sites with the parameter "a" ranging 7.6×10^{-6} - 3.0×10^{-5} and "b" ranging 0.05–0.07 d^{-1}.

It is worth noting that equation (7) can be transformed to simple relationship:

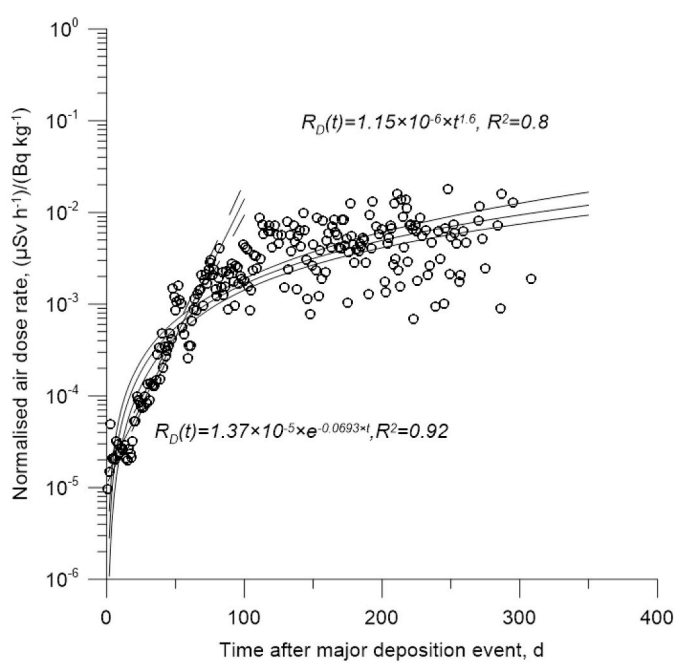

Fig. 3. Variation with time in the air dose rate normalised to the effective concentration. Individual points represent geometric means of the data available for every date. Approximations are supplemented with 95% confidence limits. Doted lines represent the exponential fit of data for March–May. Y-axis is log-scaled.

$$D(T) = a \times Q_e(t) \times e^{bt} \tag{8}$$

that can be used as relationship between air dose rate and effective concentrations in plants at the early stage after the deposition. The R_d values for March–May, demonstrate an exponential growth with the time lapsed from the deposition because of the decay of ^{131}I in the weed and the radionuclides weathering from the leaf surfaces, whereas during June–December this increase become negligible (Fig. 3). A few months after the deposition, the radionuclides were totally transferred to the surface of the topsoil that consisted of mainly decomposed plant residues (the sod, or root mat) and during June–December the radiocaesium concentrations in the weed leaves were governed by the radionuclide concentrations in the sod. The leaching of radiocaesium from the sod is much slow compared to weathering and therefore, the radiocaesium transfer to the weed leaves sampled in June–December was approximately at the same level. Thus, a linear relationship between the logarithm of air dose rate normalised to the effective radionuclide concentrations in the weed and the time after the deposition was observed during March to May, and this dependence became obscure for the samples obtained during June to December.

The correlation coefficients between radionuclide activity concentrations of ^{131}I, ^{134}Cs, ^{137}Cs in the sampled weed leaves and air dose rates are given in Table 3. In most cases, a correlation between the ^{131}I concentrations in the weed leaves and the air dose rate was higher than that of between other radionuclides and air dose rates. This was because of short decay, short radionuclide half-lives on the weed leaves and, hence, faster reduction of iodine at the surface of leaves. Another reason is that for calculation of the correlation coefficient for iodine, only values higher than the detection limit were analysed, i.e. the data for iodine covered much shorter time than for radiocaesium. This was crucially important for the sites where time-series of the measurements were relatively short. In such cases, identification of the parameters of the air dose reduction, as well as radionuclide concentrations in plants may had higher variations.

A general point, relevant to all sites is that all correlation coefficients calculated for June–December for individual sampling sites were much lower than those calculated for March–May and demonstrate a lack of the linear dependence between the air dose and radiocaesium

Table 3
Correlations between major dose forming radionuclides in the weed leaves and the ambient dose rate.

Sampling site	Correlation coefficient			
	^{131}I	^{134}Cs	^{137}Cs	$Q_e(t)$[1]
Sampling period: March–May				
2–1	0.90	0.66	0.66	0.69
2–2	0.81	0.44	0.43	0.51
2–3	0.69	0.76	0.76	0.76
2–4	0.79	0.46	0.46	0.62
2–5	0.86	0.78	0.77	0.82
2–6	0.64	0.65	0.65	0.68
2–7	0.93	0.71	0.71	0.74
2–8	0.81	0.63	0.63	0.66
2–9	0.65	0.54	0.54	0.55
4–3	0.72	0.52	0.51	0.54
All sites	0,59	0.66	0.66	0.68
Sampling period: June–December				
2–1		0.16	0.15	0.15
2–2		0.17	0.15	0.15
2–3		0.00	0.00	0.00
2–4		0.25	0.25	0.25
2–6		0.00	0.00	0.00
2–7		0.03	0.04	0.04
2–8		0.37	0.36	0.36
2–9		0.16	0.17	0.17
4–3		0.50	0.50	0.50
All sites		0,46	0.46	0.46

[1]Effective radionuclide concentration (See Eq. (2)).

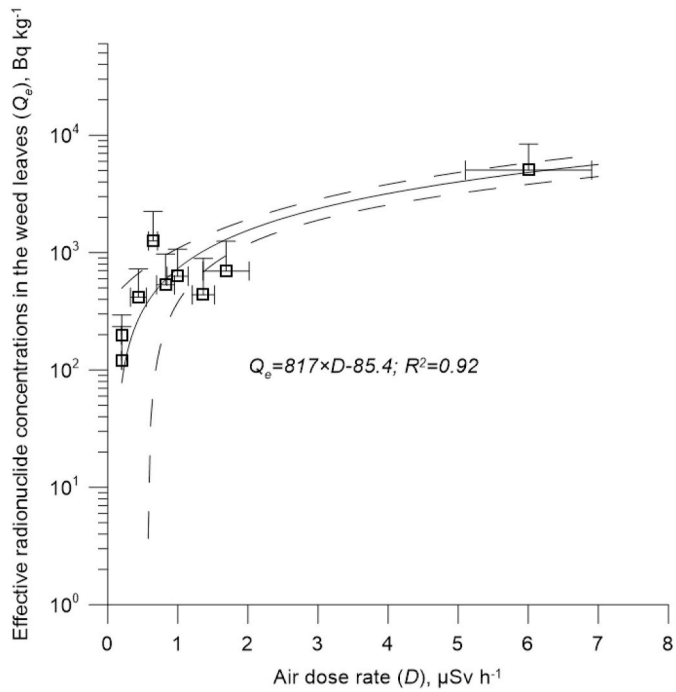

Fig. 4. Relationships between air dose rate and effective radionuclide concentration in June–December 2011. Dashed lines are 95% confidence intervals. Points represent mean data calculated for sites described in Table 1. Vertical bars are standard deviations for effective concentrations and horizontal bars are standard deviations of air dose rates. Fig. 1. Relationships between concentrations of ^{134}Cs and ^{137}Cs (left figure) and ^{131}I, and ^{137}Cs (right figure) in the weed leaves. Dashed lines represent 95% confidence interval. Y-axis is log-scaled.

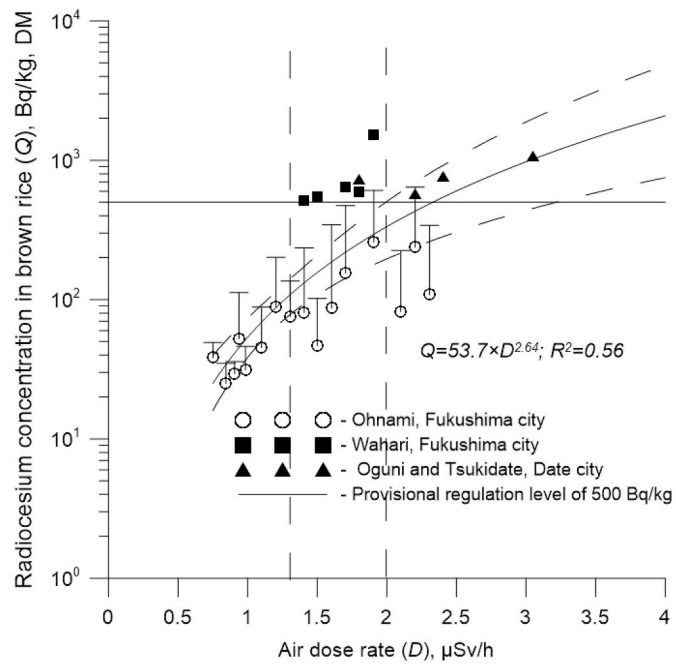

Fig. 5. Relationship between air dose rate and radioactive caesium isotopes (^{137}Cs+^{134}Cs) concentrations in brown rice in the area where relatively high level of brown rice was reported in 2011. The data for Wahari and Oguni/Tsukidate represent individual data points. The data for Onhami are mean data with standard deviations. The fit with the confidence intervals are based on all data. Y-axis is log-scaled.

concentrations in the weed leaves. The high variations of radionuclide concentrations in the weed and much lower variations observed for air dose rates at the sites were governed by some site-specific random factors including top soil properties and local inhomogeneity of the sampling sites contamination. In particular, the lowest correlation coefficients were calculated for sites 2-3, 2-6 where air dose rates measured during June–December were totally at the same level and amounted 0.2 μSv h^{-1} (MEXT, 2012b).

To avoid effect of site-specific inhomogeneity in the deposition density, the relationship the relationship between air dose rates and effective concentrations in the weed leaves were calculated based on the mean data obtained for the sampling sites during the whole period (Fig. 4). As can be seen from the data of Fig. 4, there is a statistically significant linear relationship between mean air dose rates and effective radiocaesium concentrations in plants and this dependence can be described based on the simple linear equation.

Thus, the concentrations of radiocaesium representative for the weed leaves contamination at the sampling sites can be calculated based on the air dose data and vice versa. The obtained relationships could be in use for prompt estimating concentration of radionuclides in plants that can be considered for analogues for leaf vegetables or green fodder for animal at the early stage of the emergency when there is a lack of resources for precise measurements.

3.2. Comparison of radionuclides concentrations in crops, soil and the air dose rate

3.2.1. Relationship between air dose rate and radiocaesium concentration in brown rice in 2011

As it was mentioned earlier, the data on the air dose rates and the radionuclide activity concentrations in brown rice for Ohnami, Watari, Oguni and Tsukidate areas of the Fukushima prefecture were derived from the report of the Fukushima prefecture and Ministry of Agriculture, Forestry and Fisheries of December 2011. The radiocaesium activity concentrations in brown rice sampled around the Fukushima affected area in relation to the air dose rates measured at the same fields in 2011 are presented in Fig. 5.

The provisional regulation level of radioactivity in staple food introduced in 2011 was set as 500 Bq kg^{-1} and corresponding constraint on field-based agricultural activities was set as 5000 Bq kg^{-1} in dry soil (Nuclear Emergency Response Headquarters, 2011). Despite the restrictions on agricultural activities on contaminated lands, nine 30 kg-bags with rice, or approximately 1.5% of brown rice produced in 2011 were found to be with the radiocaesium concentration above the provisional regulation value. This rice was harvested from the fields with radiocaesium in soil that was with the radiocaesium concentrations below 5000 Bq kg-1, i.e. where rice production was permitted (Fukushima Association for Securing Safety of Agricultural Products, 2012). That was because the soil radioactivity was not the only factor that determines the radionuclide accumulation in the crops. There were some other factors, such as the level of exchangeable potassium in soil, pH of the soil solution and content of the clay minerals in the soil also affected the radionuclide transfer to plants (Zhu and Smolders, 2000; Nisbet and Woodman, 2000; IAEA, 2009; Matsunami et al., 2016).

The data reported in Fig. 5 allow delineation of the areas where the concentration of radiocaesium in rice bags could exceeded 500 Bq kg^{-1}. Based on the data given in Fig. 5, it can be speculated that if the air dose rate exceeded some value (i.e. 1.4 μSv/hr), radiocaesium activity concentration in the rice samples can exceed 500 Bq kg^{-1}. The default OILγ3 of the IAEA recommends the value of 1 μSv h^{-1} at 1 m above background for delineation of the areas where agricultural produce are not suitable for consumption after the severe reactor accident. The value 1.3 μSv/hr obtained here is close to the default OILs value, indicating a need to adopt the OILs to the site-specific conditions as it is recommended by the IAEA document (IAEA, 2017). Thus, the data on air dose rate can be used for justification in a need of more comprehensive monitoring in the area

with ambient dose rate above 1.3 μSv hr^{-1} for the first year after the depositions.

As rice cultivation was initiated after May, the soil was paddled before planting in most cases and subsequent uptake radioactive Cs by plants was from relatively homogeneously contaminated soil. Additionally, some remedial options, such as application of potassium fertilisers, can provide substantial reduction of the crop contamination without effect on the value of the air dose rate by physical removal of radionuclides from top soil. In 2011, potassium concentrations in the soil of all fields where concentrations of radiocaesium in the rice exceeded the provisional regulation value were measured. The results of this study showed that the fields with low potassium concentrations in the soil had a higher probability of producing rice with activity concentration higher than the regulation value (Kohyama et al., 2015; Kato et al., 2015; Shinano, 2016). The reduction of ^{137}Cs transfer to crops can be observed even in the areas where no remediation was applied because of the so-called "aging" of radiocaesium in soil (Fesenko et al., 1997; IAEA, 2009; Takeda et al., 2013; Tsukada, 2014; Saito et al., 2016).

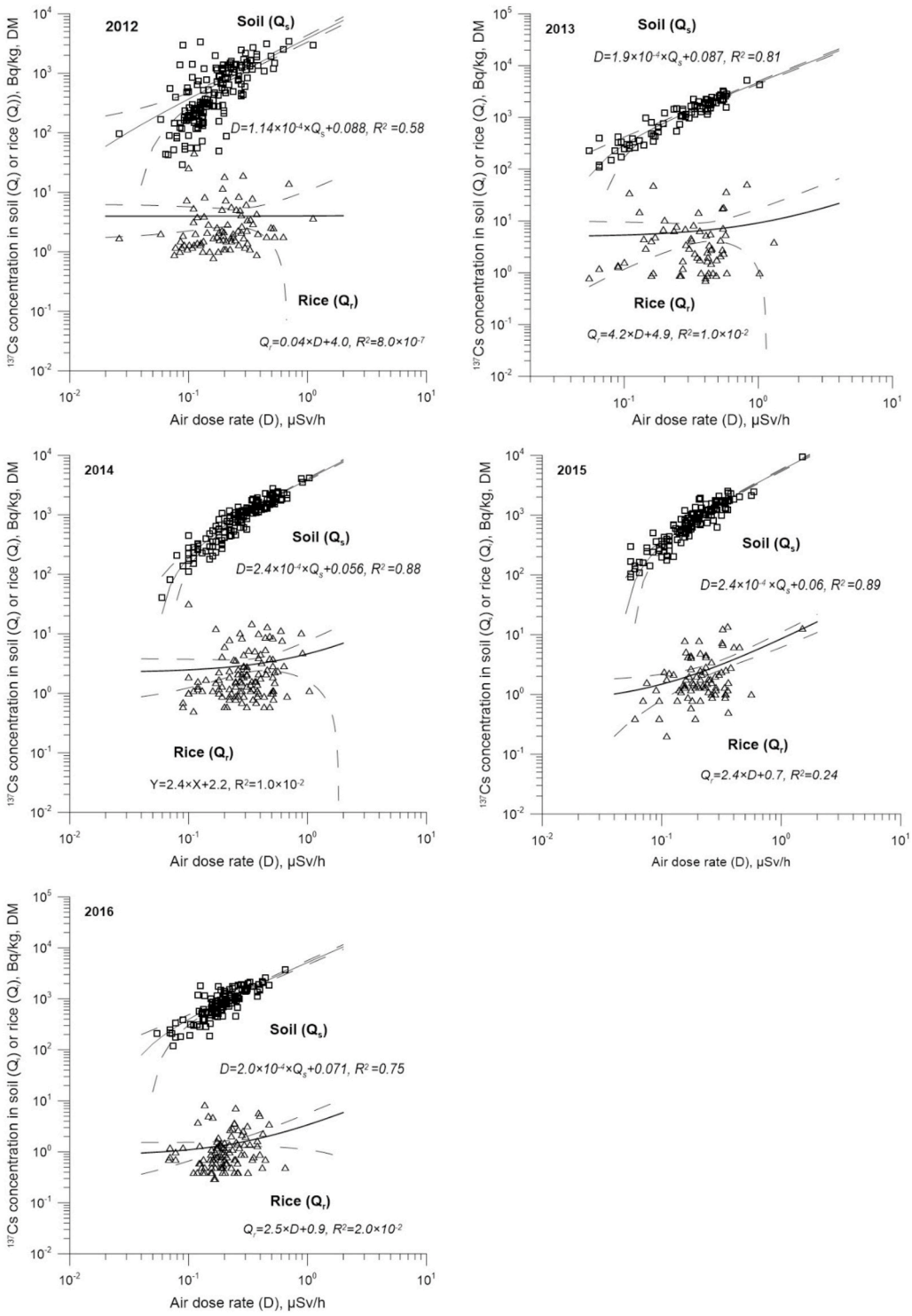

Fig. 6. Relationship air dose rate and activity concentrations of ^{137}Cs in the soil and brown rice. The data represent individual data points. Both Y-and X-axes are log-scaled.

Recently, such effects received statistical justifications based on measured rice products in Japan (Yamamura et al., 2019). Thus, the concentrations of radiocaesium in the plant reflect an influence of several factors and relationship between radionuclide concentration in plants and air dose rates should account for these co-factors.

3.2.2. Relationship between air dose rate and radiocaesium concentration in soil and crops in 2012–2016

The relationships between air dose rates for 2012–2016 and corresponding concentrations of ^{137}Cs in the soil and brown rice are outlined in Fig. 6.

As it was mentioned earlier, in case of radiological emergency the ambient dose rate consists of two components reflecting the contribution of "emergency" radionuclides and natural radiation background. That contribution of radiation background should be considered when assessing correlation between external dose rate and radiocaesium concentration in plants. Although it can be assumed that the mean contribution of the radiation background in Japan was 0.056 μSv h^{-1} without account of cosmic radiation (Abe et al., 1980), the contributions of natural radionuclides can vary in some range, depending on local variations of concentrations of natural radionuclides in the soil.

The data presented in Fig. 6 provide estimates of the natural radiation background. These values are generally higher than the value of 0.037 μSv h^{-1} given in the IAEA report (IAEA, 2015b) as average external dose rate to the public, and ranged 0.06–0.1 μSv h^{-1}. Such differences can reflect the shielding effect used for estimating external dose rate in radiological protection of humans and variations in natural background across different locations.

It can be seen from Fig. 6 that the relationship between ^{137}Cs in the soil and the air dose rate includes two components. The first component represents linear relationship between air dose rate and radiocaesium activity in the soil, reflecting contamination of the soil by radiocaesium of the Fukushima origin with small contribution of the radiation background. The second one (constant value) provides contribution to the measured air dose rate by the natural radiation background. The contributions of natural background at open places, assessed based on the data of individual years are given in Fig. 6, varied from 0.056 to 0.088 μSv h^{-1} with a mean value of 0.078 μSv h^{-1} and a standard deviation around that mean of 0.014 μSv h^{-1}. It is worth noted that these values complies with the pre-Fukushima assessments for whole Japan radiation background value of 0.056 μSv h^{-1} presented in Abe et al. (1980).

The deviations of the air dose rate values from the fit curve were higher for the year 2012 (Fig. 6). This tendency can be explained by the application of the soil based remedial options. The fraction of the fields with high air dose rates was much higher for the year 2012 compared to those observed for 2013–2016 when the soil-based remediation activity on fields used for agriculture after 2011 was mainly completed. If the data for 2012 are excluded from this analysis, the contribution of the natural background can be assessed as 0.075 ± 0.007 μSv h^{-1}. This value can be used for assessing external doses to the farmers considering time they spend doing agricultural work. Additionally, the account for contribution of natural background is especially important when external dose rates are getting below 0.075 μSv h^{-1} and use of ambient dose rate for assessments may result in misinterpretation of the field data.

No clear relationship between radiocaesium concentrations in the rice and the air rates was found to be in 2012–2016 (Fig. 6). Radiocaesium concentrations in the brown rice were much lower compared with those measured in 2011. In 2011, some fields with lower potassium levels produced relatively higher concentration of radioactive Cs in the brown rice, and it seems that the soil radioactivity directly affected contamination of the brown rice only where the soil potassium level was low.

The data reflecting relationships between radiocaesium activity concentrations in rice and air dose rates provided by the MAFF along with some examples demonstrating on effect of potassium application to the crop contamination for 2012 and 2016 are presented in Fig. 7.

The air dose rate in 2012 were in the range from 0.1 to 10 μSv h^{-1}, whereas potassium concentration in the soil varied from 20 to 80 mg per 100 g of K_2O. In 2012, high concentrations in brown rice were found to be at the rather low air dose rates and the concentration of available K_2O in soil well below 20 mg per 100 g of dry soil. The fraction of the data with K_2O in soil higher 20 mg per 100 g, show similar levels of radiocaesium in plants, thus reflecting effect of potassium application as a remedial option mentioned above. The data on role of potassium in regulating in radiocaesium transfer to plants were also presented in some other publications (Kato et al., 2015; MAFF, 2016; Kubo et al., 2017).

Rice is a staple crop in Japan and priorities in remediation of agricultural fields in the areas affected by the Fukushima Dai-ichi accident were given to paddy fields. After 2012, an additional application of potassium to decrease the radiocaesium transfer from the soil to plants were implemented and these actions resulted in a lack of the relationship between radiocaesium in plant and the air dose rate. Additional

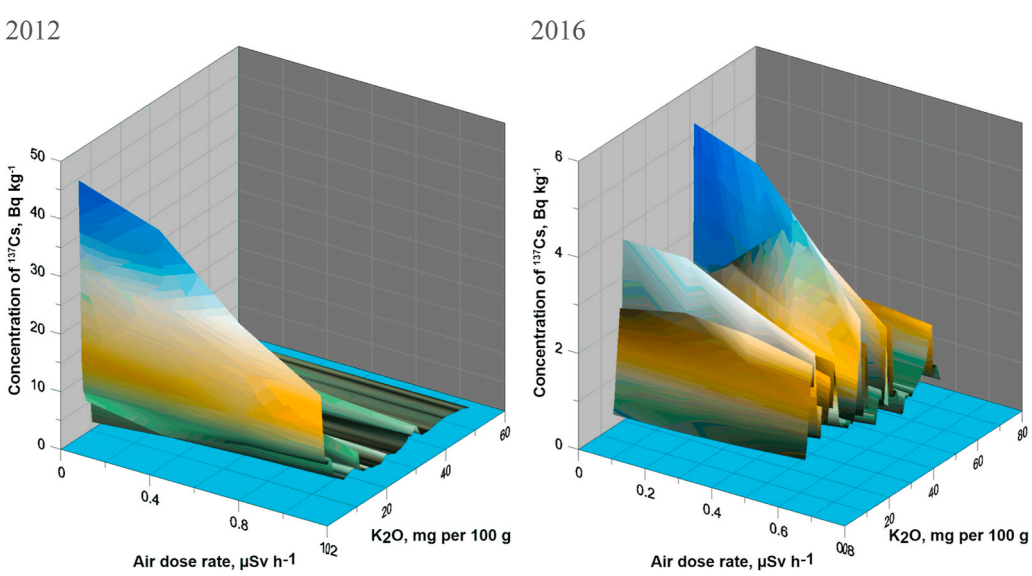

Fig. 7. Relationship between ^{137}Cs concentrations in brown rice (Bq kg^{-1}, dry matter), exchangeable (or available for 2012) potassium (K_2O, mg per 100 g of dry soil) and ambient dose rate in μSv h^{-1}. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

potassium fertilizers were applied in the rates maintaining some required exchangeable potassium level in the soil. The concentration of potassium ions at the root surface of the plant can substantially affect transfer of both potassium and radiocaesium due to the competitive character of uptake of these chemical elements. Thus, puddling of soil is an important soil management process that mixes the soil thoroughly and allows potassium ions easy access the root surface due to the saturated soil condition (Yamaguchi et al., 2012). These features of the rice planting make the soil to plant concentration ratios for rice relatively low and could mask dependence of radiocaesium activity concentration in plants on the soil radioactivity. Indeed, application of elevated potassium rates based on the status of exchangeable potassium in the soil provides different reduction of ^{137}Cs concentration in the crop. In 2012, the rice contamination in the fields with higher exchangeable potassium in soil reduced to the minimum levels and varied randomly (Fig. 7a). In 2016 (Fig. 7b), because of wide application of remediation to the whole affected areas, the concentrations of ^{137}Cs in crops was rather low and reflected site-specific difference in ^{137}Cs transfer to rice.

A similar effect was observed in the areas affected by the Chernobyl accident, where the contribution of the remediation to the reduction of ^{137}Cs concentrations in agricultural products was higher than 60% (Fesenko et al., 1995, 1997, 2001). This option substantially decreased the number of samples exceeding the standard limit value from 2012 (100 Bq kg^{-1} for the most of agricultural products), and from 2016 no brown rice samples with radiocaesium concentrations above that limit were observed.

The lack of statistically significant relationship between air dose rate and other crops, namely, buckwheat and soybean was also observed Fig. 8.

Elevated potassium rates to the fields with buckwheat and soybean has been implemented in areas affected by the accident since 2013, i.e. one year after potassium application for reduction of radiocaesium in rice. In 2012, the conventional fertilization and soil management processes carried out in the case of upland crops, but the fertilization of certain fields were insufficient to decrease concentrations of radiocaesium in the crop. The soil management differences could affect the relationship between concentration of radiocaesium in plants and air dose rates because puddling is not applied to the upland crops, and water saturation may occur only during heavy rains.

It is worth mentioning that there are some differences in locations of paddy and upland fields. Paddy fields are normally placed on the flat area, while upland fields are placed on a sloped section and closer to the non-decontaminated area (such as forest). The surrounding environment is also an important factor that may affect the air dose rate and, in some areas, the data can reflect these situations. Additionally, attachment of soil to harvested grain was reported in many cases for buckwheat. It had could affect the radioactivity in crops, if the grain was not subjected polishing to before measurements (Kubo et al., 2016).

3.2.3. *Effects of radiocaesium concentrations below MDA*

A quantity of the samples with radiocaesium concentrations below minimum detectable activities (MDA) were growing with the time lapsed after the depositions and such samples were found irrespective of the air dose rate in the dose range 0–0.5 μSv h^{-1}. (Fig. 9 and Table 1).

The fractions of samples with the ^{134}Cs concentrations below MDA were generally higher than those for ^{137}Cs (Table 1). This is anticipated results, as concentrations of ^{134}Cs in plants were reduced faster than that of ^{137}Cs because of the shorter half-life. These data also show that the sampling programme can be optimized based on the air dose rate data avoiding (or decreasing) overabundant sampling in areas where samples with concentrations of radiocaesium below MDA can dominate.

It is worth also noting that the derived relationships are not as straightforward and reflect differences in the soil treatment of the affected areas. Additionally, scattering of the data in areas of high concentrations of radiocaesium in the soil demonstrates the effectiveness of different soil-based management options resulting in redistribution of radionuclides in the soil. Such effects should be accounted for evaluation of the data where concentrations of radiocaesium can be below MDA.

The MDA values and the activity concentrations in plants are different for ^{134}Cs and ^{137}Cs. Therefore, the data on frequency distribution for these radionuclides with the radiocaesium concentrations below MDA were analysed separately (Fig. 9). The account for the contribution of natural background (right column) show that the maximum of the samples with the concentrations below MDA can be observed at external dose rates below 0.07–0.09 μSv h^{-1}, and these values can be considered as lower limits, below which the monitoring of the air dose rates cannot provide useful information.

4. Conclusions

A robust environmental monitoring programme is the essential tool to ensure food safety in case of radiological emergencies. Such programmes were developed and implemented in Japan shortly after the

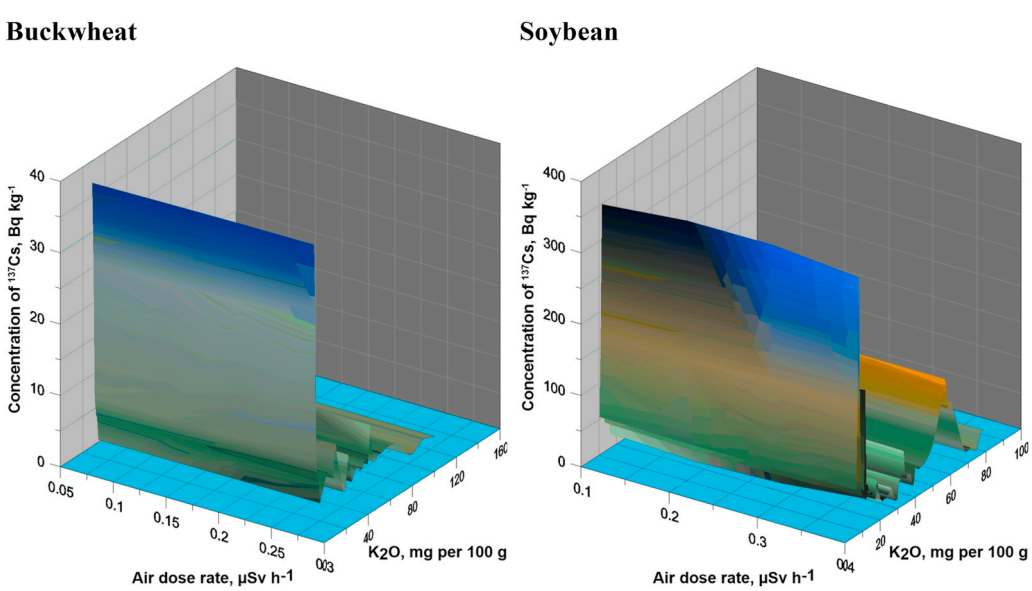

Fig. 8. Relationship between ^{137}Cs concentrations in buckwheat grain and soybean (Bq kg^{-1}, DM), exchangeable potassium and ambient dose rate in 2012.

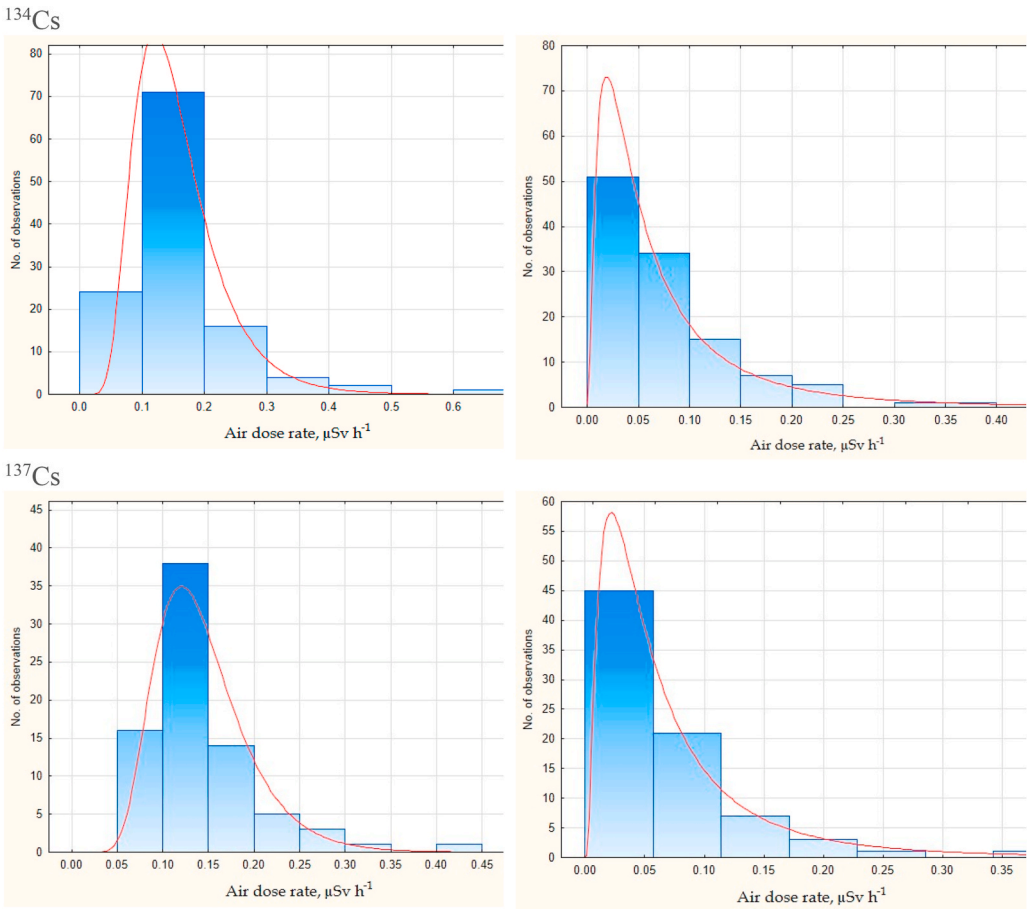

Fig. 9. Frequency distribution of the samples taken in 2012 with concentrations of ^{134}Cs and ^{137}Cs below MDA in respect to the corresponding air dose rates. The left column displays the measured data. The data in the right column were corrected with the radiation background.

Fukushima Dai-ichi NPP accident. A specific feature of the environmental monitoring program implemented in Japan was a combination of the data on radionuclide activity concentrations in the soil, plants and measurement of the air dose rates. Relationships among these radiological parameters are useful in optimizing monitoring programmes after a radiological or nuclear emergency. The statistically significant relationship between external dose rates and radionuclide concentrations in plants during March–May 2011 was nonlinear because of differences in reduction of the air dose and radionuclide concentrations in the plants, whereas for June–May a simple linear relationship between mean radiocaesium concentrations in plants and mean air dose rate was identified. It was shown that activity concentration in rice was higher the regulation limit during the first year after the accident in areas where air dose rate at the time of harvest exceeds 1.3–1.4 μSv h^{-1}. This value is close to the OIL value of 1.0 μSv h^{-1} suggested by the IAEA as a default operational intervention criterion for the case of reactor accidents. Assessments made indicated the air dose rate of 1.3 μSv h^{-1} as a threshold value for the fields, where radiocaesium in the crop could potentially exceed the regulatory value of 500 Bq kg^{-1}. In 2012–2016, the effect of implementation of remedial options (potassium application, deep ploughing, reversal tillage and top soil removal) drastically affected a dependence between radiocaesium concentration in plants and external dose rates and no reliable relationship was found between radiocaesium in crops and air dose rate. Such effect should be considered for environmental monitoring in areas where remediation was applied in response to radiological emergency. Overall, presented results can provide substantial input in the optimization of emergency monitoring. These allows the conclusion that for the sites with ambient dose rate below 1.0 μSv h^{-1}, recommended by the IAEA as OIL$_{\gamma}$3, the concentrations of radiocaesium in the crops should be applied with caution taking into account local farming conditions and regulation limits.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2020.106359.

References

Abe, S., Fujitaka, K., Fujimoto, K., 1980. Natural radiation in Japan. In: Gesell, T.F., Lowder, W.M. (Eds.), Conference on Natural Radiation Environment III; Houston, TX, USA; 23 - 28 Apr 1978. Department of Energy, Oak Ridge, TN (USA). Technical Information Center, pp. 1034–1048, 1980; pp. 1034–1048.

Alexakhin, R.M., Fesenko, S.V., Sanzharova, N.I., 1996. Serious radiation accidents and the radiological impact on agriculture. Radiat. Protect. Dosim. 64, 37–42.
Alexakhin, R.M., Sanzharova, N.I., Fesenko, S.V., Spiridonov, S.I., Panov, A.V., 2007. Chernobyl radionuclide distribution, migration, and environmental and agricultural impacts. Health Phys. 93 (5), 418–426.
Almgren, S., Isaksson, M., 2006. Vertical migration studies of Cs-137 from nuclear weapons fallout and the Chernobyl accident. J. Environ. Radioact. 91, 90–102.
Fesenko, S.V., Colgan, P.A., Sanzharova, N.I., Lissianki, K.B., Vazquez, C., Guardans, R., 1997. The dynamics of the transfer of caesium-137 to animal fodder is areas of Russia affected by the Chernobyl accident and resulting doses from the consumption of milk and milk products. Radiat. Protect. Dosim. 69, 289–299.
Fesenko, S.V., Alexakhin, R.M., Balonov, M.I., Bogdevich, I.M., Howard, B.J., Kashparov, V.A., Sanzharova, N.I., Panov, A.V., Voigt, G., Zhuchenka, Y.M., 2006. Twenty years' application of agricultural countermeasures following the Chernobyl accident: lessons learned. J. Radiol. Prot. 26, 351–359.
Fesenko, S.V., Alexakhin, R.M., Sanzharova, N.I., Spiridonov, S.I., 1995. Dynamics of ^{137}Cs concentration in agricultural products in areas of Russia contaminated as a result of the accident at the Chernobyl nuclear power plant. Radiat. Protect. Dosim. 60, 155–166.
Fesenko, S.V., Jacob, P., Alexakhin, R., Sanzharova, N.I., Panov, A., Fesenko, G., Cecille, L., 2001. Important factors governing exposure of the population and countermeasure application in rural settlements of the Russian Federation in the long term after the Chernobyl accident. J. Environ. Radioact. 56, 77–98.
Fukushima Association for Securing, 2012. Safety of agricultural products. https://fukumegu.org/ok/contents/#.
Fukushima prefecture, 2011. Fukushima prefecture and Ministry of Agriculture, Forestry and Fisheries. Interim report on the Analysis of the factor with radioactive caesium, which exceeded a provisional regulation value. http://konstantin.cocolog-nifty.com/blog/files/240112_tyukan.pdf.
IAEA, 2001. Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment, IAEA Safety Reports Series No. 19. International Atomic Energy Agency, Vienna.
IAEA, 2006. Environmental Consequences of the Chernobyl Accident and Their Remediation: Twenty Years of Experience, Report of the Chernobyl Forum Expert Group "Environment." STI/PUB/1239. International Atomic Energy Agency, Vienna.
IAEA, 2009. Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments, IAEA TECDOC No. 1616. International Atomic Energy Agency, Vienna.
IAEA, 2015a. The Fukushima Dai-Ichi Accident Technical Volume 5, Post-accident Recovery. International Atomic Energy Agency, Vienna.
IAEA, 2015b. The Fukushima Dai-Ichi Accident, Technical, 4/5. Radiological Consequences. International Atomic Energy Agency, Vienna.
IAEA, 2017. Operational Intervention Levels for Reactor Emergencies and Methodology for Their Derivation. EPR-NPP-OILs, vol. 2017. International Atomic Energy Agency, Vienna.
Kato, N., Kihou, N., Fujimura, S., Ikeba, M., Miyazaki, N., Saito, Y., Eguchi, T., Itoh, S., 2015. Potassium fertilizer and other materials as countermeasures to reduce radiocesium levels in rice: results of urgent experiments in 2011 responding to the Fukushima Daiichi Nuclear Power Plant accident. Soil Sci. Plant Nutr. 61, 179–190. https://doi.org/10.1080/00380768.2014.995584.
Khomutinin, Yu, Fesenko, S., Levchuk, S., Zhebrovska, K., Kashparov, V., 2020a. Optimising sampling strategies for emergency response: soil sampling. J. Environ. Radiact. This issue.
Khomutinin, Yu, Fesenko, S., Levchuk, S., Holiaka Kashparov, V., 2020b. Optimising sampling strategies for emergency response: sampling of vegetation. J. Environ. Radiact. This issue.
Kohyama, K., Obara, H., Tanaka, Y., Saito, T., Sato, M., Yoshioka, K., Taniyama, I., 2015. Soil properties for analysing cause of high radiocesium concentration in brown rice produced in 2011 in Fukushima prefecture. Bull. Natl. Inst. Agro-Environ Sci. 34, 63–73. In Japanese with English summary.
Kubo, K., Fujimura, S., Kobayashi, H., Ota, T., Shinano, T., 2017. Effect of soil exchangeable potassium content on cesium absorption and partitioning in buckwheat grown in a radioactive cesium-contaminated field. Plant Prod. Sci. 20, 396–405. https://doi.org/10.1080/1343943X.2017.1355737.
Kubo, K., Kobayashi, H., Nemoto, K., Hirayama, T., Matsunami, H., Ichihashi, Y., Ota, T., Keitoku, S., Shinano, T., 2016. Decreasing radioactive cesium in lodged buckwheat grain after harvest. Plant Prod. Sci. 19, 91–95.
Matsuda, N., Mikami, S., Shimoura, S., Takahashi, J., Nakano, M., Shimada, K., Uno, K., Hagiwara, S., Saito, K., 2015. Depth profiles of radioactive cesium in soil using a scraper plate over a wide area surrounding the Fukushima Dai-ichi Nuclear Power Plant. Japan. J. Environ. Radioact. 139, 427–434.
Matsunami, H., Murakami, T., Fujiwara, H., Shinano, T., 2016. Evaluation of the cause of unexplained radiocaesium contamination of brown rice in Fukushima in 2013 using autoradiography and gamma-ray spectrometer. Sci. Rep. 6, 20386 https://doi.org/10.1038/srep20386 published online 4th February 2016.
MAFF, 2013. Ministry of Agriculture, Forestry and Fisheries. Technical documents for decontamination of agricultural field. http://www.maff.go.jp/j/nousin/seko/josen/.
MAFF, 2016. Ministry of Agriculture, Forestry and Fisheries (MAFF), 2016. Factors that radioactive cesium concentration is high and its countermeasures in soybean. http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/youin_daizu_3.pdf.
MEXT, 2012a. Ministry of education, culture, Sports, science and Technology. Compiled in Nuclear Regulation Authority website. https://radioactivity.nsr.go.jp/ja/contents/6000/5081/24/Readings_of_environmental_monitoring_sample_All_Results_For_May2011_20120912.pdf.
MEXT, 2012b. Ministry of education, culture, Sports, science and Technology. Compiled in Nuclear Regulation Authority website. https://radioactivity.nsr.go.jp/en/contents/6000/5706/24/Readings_of_environmental_monitoring_sample_June1,2011-%20December31,2011_20121101.pdf.
NARO, 2015. Guideline for the usage of weed in the levee, actual situation and the points for the usage after nuclear power plant accident. http://www.naro.affrc.go.jp/publicity_report/publication/files/narc_keihansou_cs.pdf.
Nisbet, A.F., Woodman, R.F.M., 2000. Soil-to-plant transfer factors for radiocaesium and strontium in agricultural systems. Health Phys. 78, 279–288.
Nuclear Emergency Response Headquarters, 2011. http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/ine_sakutuke.pdf.
Onda, Y., Kato, H., Hoshi, M., Takahashi, Y., Nguyen, M.-L., 2015. Soil sampling and analytical strategies for mapping fallout in nuclear emergencies based on the Fukushima Dai-ichi Nuclear Power accident. J. Environ. Radioact. 139, 300-107.
Onda, Y., Swank, L., Shinano, T., Dercon, G., Lee Zhi Yi, A., Kato, H., Ohse, K., 2020. Soil and vegetation sampling approaches for agricultural systems during early stage of nuclear emergencies. J. Environ. Radioact. this issue.
Prister, B.S., Belli, M., Sanzharova, N.I., et al., 1996. Behaviour of radionuclides in meadows including countermeasures application. In: Karaglou A, A., Desmet, G., Kelly, G.N., Menzel, H.G. (Eds.), The Radiological Consequences of the Chernobyl Accident. EUR 16544 EN. Brussels: European Commission, pp. 59–68.
Renaud, P.H., Simon-Cornu, M., Gonze, M.-A., Morlon, C., Parache, V., Orsakissok, I., Navarro, E., Cessac, B., Rannou, A., Champion, D., 2012. Contamination of Japanese foodstuffs of terrestrial origin after the Fukushima nuclear accident and related dose assessments Part 1: foodstuff contamination. Radioprotection 48, 493–509.
Saito, T., Takahashi, K., Murakami, T., Shinano, T., 2016. Analysis of factors causing high radiocesium concentrations in Brown rice grown in minamisoma city. In: Radiological Issues for Fukushima's Revitalized Future, ISBN 978-4431558477, pp. 189–198. https://doi.org/10.1007/978-4-431-55848-4_17.
Shinano, T., 2016. Mitigation of radioactive contamination from farmland environment and agricultural products. Mod. Environ. Sci. Eng. 2, 454–461. https://doi.org/10.15341/mese(2333-2581)/07.02.2016/003.
Takata, Y., Kohyama, K., Obara, H., Maejima, Y., Ishitsuka, N., Saito, T., Taniyama, I., 2014. Spatial prediction of radioactive Cs concentration in agricultural soil in eastern Japan. Soil Sci. Plant Nutr. 60, 393–403. https://doi.org/10.1080/00380768.2013.879528.
Takata, Y., Koyama, K., Obara, H., Maejima, Y., Hiradate, S., Kihou, N., Saito, T., Taniyama, I., 2015a. Emergency survey for spatial mapping of radioactive caesium concentration in agricultural soil. Bull. Natl. Inst. Agro-Environ Sci. 34, 43–51. In Japanese with English summary.
Takata, Y., Koyama, K., Obara, H., Maejima, Y., Ishitsuka, N., Saito, T., Taniyama, I., 2015b. Spatial prediction of radioactive Cs concentration in agricultural soil in East Japan. Bull. Natl. Inst. Agro-Environ Sci. 34, 53–61. In Japanese with English summary.
Takeda, A., Tsukda, H., Nakao, A., Takaku, Y., Hisamitsu, S., 2013. Time-dependent changes of phytoavailability of Cs added to allophanic Andosols in laboratory cultivations and extraction tests. J. Environ. Radioact. 122, 29–36. https://doi.org/10.1016/j.envrad.2013.02.005.
Tsukada, H., 2014. Behaviour of radioactive caesium in soil with aging. Jpn. J. Soil Sci. Plant Nutr. 85, 77–79. In Japanese.
Yamaguchi, N., Eguchi, S., Ikeba, M., Fujiwara, E., Makino, T., Taniyama, I., 2015. Radiocesium extraction from arable soils at the initial stage after deposition of radionuclides. Bull. Natl. Inst. Agro-Environ Sci. 34, 53–61. In Japanese with English summary.
Yamaguchi, N., Takada, Y., Hayashi, K., Ishikawa, S., Kuramata, M., Eguchi, S., Yoshikawa, S., Sakaguchi, A., Asada, K., Wagai, R., Makino, T., Akahane, I., Hiradate, S., 2012. Behaviour of radiocaesium in soil-plant systems and its controlling factor: a review. Bull. Natl. Inst. Agro-Environ Sci. 31, 75–129. In Japanese with English summary.
Yamamura, K., Fujimura, S., Saito, T., Ishikawa, T., Arai, M., Ota, T., Shinano, T.A., 2019. A statistical model for estimating the radiocesium transfer factor from soil to brown rice using the soil exchangeable potassium content. J. Environ. Radioact. 195, 114–125.
Zhu, Y.-G., Smolders, E., 2000. Plant uptake of radiocaesium: a review of mechanisms, regulation and application. J. Exp. Bot. 51, 1635–1645. https://doi.org/10.1093/jexbot/51.351.1635.