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Time trends in radiocaesium in the Japanese diet following nuclear weapons testing and Chernobyl: Implications for long term contamination post-Fukushima

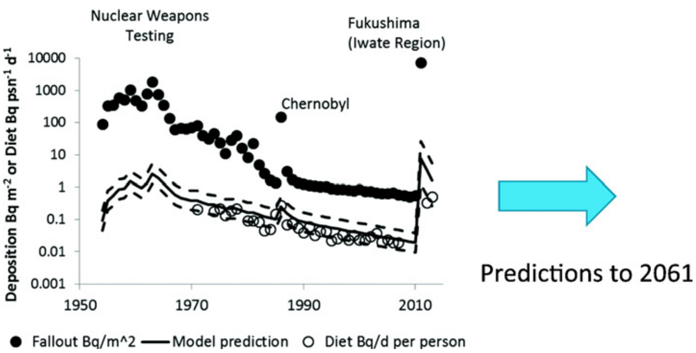
Jim T. Smith [a,*], Keiko Tagami [b], Shigeo Uchida [b]

[a] School of Earth and Environmental Sciences, University of Portsmouth, Burnaby Building, Burnaby Road, Portsmouth PO1 3QL, UK
[b] National Institutes for Quantum and Radiological Science and Technology, 4-9-1 Anagawa, Inage-ku, Chiba-shi, Chiba 2638555, Japan

HIGHLIGHTS

- Cs is the main contaminant following the Fukushima Daiichi nuclear accident.
- We analysed >4000 measurements of Cs in Japan over 50 years pre-Fukushima.
- Time trends in changes in Cs in foodstuffs and diet were identified.
- Relatively consistent time trends were seen in diet pre and post-Fukushima.
- Models of 1959–2009 data can inform dose predictions and management.

GRAPHICAL ABSTRACT



50 years of radiocaesium in the Japanese diet.

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ABSTRACT

Estimation of time changes in radiocaesium in foodstuffs is key to predicting the long term impact of the Fukushima accident on the Japanese diet. We have modelled >4000 measurements, spanning 50 years, of ^{137}Cs in foodstuffs and whole diet in Japan after nuclear weapons testing (NWT) and the Chernobyl accident. Broadly consistent long term trends in ^{137}Cs activity concentrations are seen between different agricultural foodstuffs; whole diet follows this general trend with remarkably little variation between averages for different regions of Japan. Model blind tests against post-NWT data for the Fukushima Prefecture showed good predictions for radiocaesium in whole diet, spinach and Japanese radish (for which good long term test data were available). For the post-Fukushima period to 2015, radiocaesium in the average diet followed a declining time trend consistent with that seen after NWT and Chernobyl. Data for different regions post-Fukushima show a high degree of mixing of dietary foodstuffs between regions: significant over-estimates of average dietary ^{137}Cs were made when it was assumed that only regionally-produced food was consumed. Predictions of mean committed effective internal doses from dietary ^{137}Cs (2011 to 2061) in non-evacuated parts of the Fukushima Prefecture show that average internal dose is relatively low. This study focused on average regional ingestion dose rates and does not attempt to make site specific predictions. However, temporal trends identified could form a basis for site specific predictions of long term activity concentrations in agricultural products and diet both outside and (to assess potential re-use) inside currently evacuated areas.

* Corresponding author.
E-mail addresses: jim.smith@port.ac.uk (J.T. Smith), tagami.keiko@qst.go.jp (K. Tagami), uchida.shigeo@qst.go.jp (S. Uchida).

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1. Introduction

Following the 2011 Fukushima Daiichi (FDNPP) nuclear accident, the Japanese government put in place a large scale contamination monitoring system designed to prevent sale and consumption of foodstuffs with >100 Bq kg^{-1} of radiocaesium, though the initial provisional regulation value was up to 500 Bq kg^{-1} (Hamada et al., 2012). Despite the unprecedented scale of food monitoring, and the relatively low predicted radiation dose rates to the Japanese population (UNSCEAR, 2013; WHO, 2013), the continued presence of relatively long-lived ^{137}Cs in foodstuffs – understandably – remains of major concern to Japanese people. For example, rice grown in the more contaminated parts of the Fukushima Prefecture is currently traded at an approximately 20% lower price than the national average (MAFF, 2016) due to consumer concerns over its safety. This concern is heightened by uncertainty in how dose rates from consumption of contaminated food will evolve over coming decades. This study focuses on ^{137}Cs, the most important long term contaminant from the FDNPP accident: although initially important, ^{134}Cs decays with a 2.07 year half-life, so is not significant compared to ^{137}Cs in the long term (>10 years) after fallout.

In response to global fallout from Cold War atmospheric nuclear weapons testing (NWT), the Japanese government instigated the world's most intensive programme of monitoring of radioactivity in the environment (1959–2009). All these data are available from the Japanese Environmental Radioactivity Database (NRA, 2016). Spanning a fifty year period from 1959 to 2009, thousands of measurements were made of ^{137}Cs in fallout, wheat, rice, and whole diet, including in 1986 when small amounts of Chernobyl fallout reached Japan. The present study models these unique historical data to evaluate long term availability of ^{137}Cs in Japanese agricultural systems.

On long time scales (years), the processes that determine radiocaesium transfers to and from many different ecosystem components (for example, soil-plant and between plants and herbivorous mammals) are fast in comparison with the slow decline in radiocaesium availability in soil (Shiozawa, 2013; Smith et al., 1999). Laboratory studies have shown that the sorption of ^{137}Cs by soils and sediments has a slow kinetic component, in which ^{137}Cs is transferred to less available sites in the mineral lattice (Evans et al., 1983), a process often referred to as "fixation".

After NWT fallout, Aarkrog (1971) observed an initial exponential decline in ^{137}Cs activity concentrations in milk and rye with effective ecological half-life (half time of reduction in activity concentration in an environmental compartment, including physical decay) of 1–2 years. In a study of radiocaesium levels in the human body after NWT, Newton et al. (1977) hypothesised a pool of bio-available ^{137}Cs in the environment that declined even more slowly, with half-life 7.5 to 30 years. Following the Chernobyl accident, Smith et al. (1999) analysed many long term field studies of temporal changes in radiocaesium in three different ecosystem components: vegetation, surface waters (dissolved phase), and milk. Over the first five years after Chernobyl, these workers observed similar rates of decline of ^{137}Cs in all three ecosystem components, all having effective ecological half lives in the range 1–4 years, with mean approximately 1.7 years. In the longer term (>5 years post-accident), a slower component to the decline in Chernobyl-derived ^{137}Cs activity concentrations in surface water, terrestrial vegetation, and fish was observed (Jonsson et al., 1999; Smith et al., 2000b) with effective ecological half-life in fish, water and terrestrial vegetation increasing to 6–30 years. From the observed persisting mobility of radiocaesium in the environment, and particularly the increase in effective ecological half-life over time, it was concluded (Smith et al., 2000b) that the sorption-desorption process of radiocaesium in soils and sediments tends towards a reversible steady-state. These post-Chernobyl results confirmed the hypothesis of Newton and coworkers (Newton et al., 1977), based on NWT fallout, that there was no complete fixation of radiocaesium to soils, but a continued contamination of foodstuffs that declined only slowly in the decades after fallout.

The weapons test fallout occurred at differing rates over a number of years whilst the vast majority of the Chernobyl fallout was deposited during only a few days. Once the different input functions are accounted for, however, the long term changes in environmental mobility of radiocaesium can be compared for both events (Smith et al., 2000a). Thus, a study of the extensive measurements of radiocaesium in Japanese foodstuffs (including rice, the key staple food in Japan) after NWT and the Chernobyl accident should be able to inform predictions of the long term consequences of food chain contamination post-Fukushima. In particular, accounting for the long term decline in ^{137}Cs bioavailability should give a more accurate and less conservative estimate of future ingestion doses and contamination of products than was possible in previous studies. For example, Harada et al. (2014) assumed only physical decay in assessing future doses from Fukushima. Further, an assessment of research relevant to long term dose assessment post-FDNPP (UNSCEAR, 2015) highlighted the need for studies of "dynamics of the variation over time of the concentration of caesium radionuclides in various foods".

2. Materials and methods

2.1. Data sources

Fallout of ^{137}Cs from atmospheric nuclear weapons testing (NWT) and the Chernobyl accident was estimated using monthly measurements of ^{137}Cs deposited (Bq m^{-2}) in 9 regions (Akita, Fukuoka, Hokkaido, Ibaraki, Ishikawa, Miyagi, Niigata, Osaka, Tokyo) where long time series data were available (SM: Table S1). Deposition prior to April 1957 (the beginning of measurements at Tokyo) was estimated using fallout data from 1954 to 85 (Cambray et al., 1989). The Tokyo data (scaled so that total fallout from beginning of sampling to 1985 was equivalent to that from the given region) were used to provide estimates for missing data from before sampling began (1959 or 1963) at other sites. Missing data within the sampling period (to February 2011) were estimated by linear interpolation to provide a complete data set of monthly deposition in each region. Using the empirical fallout data for each region reduces model uncertainty since it does not require assumptions to be made about the precipitation-fallout relationship. Models were run both with monthly and annually averaged deposition data, though no clear improvement was observed by using monthly data so the results presented here use annual data.

Activity concentrations of ^{137}Cs in wheat, brown and white rice (Bq kg^{-1} d.w.), and whole diet (Bq d^{-1} per person) were assembled from (Komamura et al., 2006) and data in vegetables from (NRA, 2016). Whole diet data were obtained using the duplicate diet method (one additional portion of each meal was taken for measurement) for five households in each prefecture. The wheat and rice data are summarised in Table S1. These data typically spanned a time period from 1959 to 63 up to 2001–09, though in four regions, whole diet measurements did not begin until 1971/72. For vegetables (radish root and leaf, spinach leaf), individual samples were measured at a large range of sites across Japan: 1343 measurements from >100 sites for radish root; 364 measurements from 44 sites for radish leaf and 1200 measurements from >100 sites for spinach. The wheat, rice and whole diet data were analysed using deposition data for the corresponding region; data for vegetables were from >30 prefectures across Japan, and were analysed as a single dataset using mean national deposition data.

For wheat, rice, and vegetables the year of Chernobyl fallout (1986) was excluded from model fits since it was not expected that these annual data would reproduce the short term Chernobyl peak. For whole diet data, which responds on a longer timescale, data from 1986 were included.

Associated site-specific measurements of soil type, soil density, pH, Cation Exchange Capacity (CEC), and particle size distribution (coarse and fine sand, silt and clay) were available (Komamura et al., 2006) for measurements of radiocaesium in wheat and rice. These data were used to investigate possible associations between soil characteristics and long term (1985–99) ^{137}Cs activity concentrations in wheat and rice. Data were log-transformed where necessary to ensure normality, tested using the Shapiro-Wilk test.

2.2. Modelling time changes in ^{137}Cs

Time changes in ^{137}Cs concentrations from weapons testing and Chernobyl can be characterised using an exponential "transfer function" model (Smith et al., 2000b):

$$C(t) = \int_{-\infty}^{t} D(\tau)\left(Ae^{-(\lambda+k_1)(t-\tau)} + Be^{-(\lambda+k_2)(t-\tau)} + Ce^{-(\lambda+k_3)(t-\tau)}\right)d\tau \quad (1)$$

where $C(t)$ is the concentration of the radionuclide in the environmental compartment of interest (in this case, wheat, rice, vegetables, and whole diet) at time t and $D(t)$ (Bq m^{-2}y^{-1}) is the (time dependent) radionuclide deposition. A, B, and C (m^2 kg^{-1} for foodstuffs; m^{2}d^{-1} for diet) are coefficients representing, respectively, a fast decline in activity concentrations after fallout (at rate k_1), a slow decline (at rate k_2) as a result of soil fixation processes, and the very long term component which declines due to vertical migration, erosion and further slow reductions in bioavailability. λ is the physical decay constant of ^{137}Cs.

For fitting to annual data in food products and whole diet, we used the discrete version of this equation:

$$C(j) = A'D(j) + \sum_{i=1}^{i=j} D(i)\left(Be^{(-(k_2+\lambda)(j-i))} + Ce^{(-(k_3+\lambda)(j-i))}\right) \quad (2)$$

where the first short-timescale component (rapid washoff from plant surfaces and initial sorption to soil) is replaced by $A' = A / k_1$ since we are here considering annual data and $k_1 \gg 1$ year^{-1}. Not all parameters in Eqs. (1) and (2) are required to describe the time-dependent variation in foodstuffs and whole diet. Therefore, eight different variations of Eq. (2) with different combinations of parameters included or omitted were fitted to the measured data using the proc. NLIN function in SAS (Version 9 of the SAS System. Copyright © 2015 SAS Inst. Inc.). The best fit model was chosen using the corrected Akaike Information Criterion (Burnham and Anderson, 2003). Model fits were carried out for wheat, rice and whole diet from the 9 individual regions and for a set of averaged measurements over the 9 regions, as well as for average activity in spinach leaf, Japanese radish leaf, Japanese radish root (all model fits excluded data from the Fukushima Prefecture to allow model blind testing against these data).

Model uncertainty was evaluated by comparing predictions of competing models, and by studying the variance between model estimates and individual site measurements: this is discussed in detail in the Supplementary material. Note that in this study we do not physically interpret parameter values, rather use them to determine the best fit to empirical data; this is discussed further in Supplementary material. Where appropriate, model averaging was carried out to test whether an averaged (based on Akaike weights) model gave significantly different results to the "best" model. Model averaging was not necessary for the key diet predictions since, for diet, the "best" model had an Akaike weight of 0.999.

2.3. Predicting 50 year cumulative internal doses

Cumulative committed effective doses following the FDNPP accident were estimated for whole diet for the period 2011–2061 using the equation:

$$50 \text{ year integrated dose (Sv)} = \sum_{i=1}^{50} C_{diet}(i).e_{ing}.365 \quad (3)$$

where C_{diet} is the average daily ^{137}Cs intake in whole diet in year i (Bq d^{-1}) and e_{ing} is the effective dose coefficient for ingestion of ^{137}Cs by adults (1.3×10^{-8} Sv Bq^{-1} (ICRP, 2013)). The 50 year period was chosen since it gives a representation of the dose expected to be accumulated by a young adult (from 2011 to 2061) during their lifetime. This period is also coherent with the 45 year range of post-NWT measurements in whole diet (1963–2008) from which the model was calibrated. We caution against extrapolation beyond a 50 year period as these models are not designed to be extrapolated beyond the range of calibration data.

3. Results

3.1. Nuclear weapons test fallout

Time series models of radiocaesium in wheat, rice, vegetables and whole diet gave good fits to measurements during the NWT fallout period (see SM for summary of available measurements and Figs. S2 and S3). Best fit models (based on the corrected Akaike Information Criterion, AIC$_c$) for each of the environmental compartments are given in SM: Table S2 and illustrated in Fig. 1. These show that a large proportion (78–98%) of the variation is explained. Akaike weight for the "best" models varied from 0.35 for wheat to 0.99 for whole diet. For all foodstuffs, model predictions for the second and third best models showed only minor differences to those for the best model (discussed in SM and see Fig. S1). Using the models developed from the post-NWT period, Fig. 2 compares long term trends in different foodstuffs for a nominal fallout (in the first year) of 1000 Bq m^{-2} (starting in 2011) simulating a delta-function (or "spike") type deposition as seen after the FDNPP accident. Fig. 2 shows a high degree of consistency in time changes between different products in the very long term (15–50 years) after fallout. Also shown in Fig. 2 is the time change in radiocaesium in river water (dissolved phase) as was determined in a previous study (Smith et al., 2004) of European rivers following the Chernobyl accident, showing consistency in long term time trends between the NWT and Chernobyl events, and between water (dissolved phase) and bioavailability to agricultural foodstuffs.

3.2. Chernobyl fallout

Following the 1986 Chernobyl accident, a small amount of radiocaesium reached Japan, as shown in the deposition data in Fig. 1. The model fit for whole diet for the years 1980–90 is shown in more detail in SM (Fig. S4). Since the influence of Chernobyl on radiocaesium deposition and activity concentrations in Japan was minor compared to NWT, the good fit to the small peak in mean ^{137}Cs in whole diet during 1986 and 87 can be considered a strong test of the model (model fitted parameters are determined by the strong NWT signal rather than the much weaker Chernobyl signal). Activity concentrations in wheat, rice, and vegetables during the first year after a fallout event are more influenced by short term processes than whole diet. Therefore (for wheat, rice and vegetables), the model was not expected to predict the influence of Chernobyl since the activity in 1986 is strongly dependent on seasonal timing of the fallout. For example, the relatively good prediction of the peak in ^{137}Cs in wheat in 1986 (Fig. 1) is a coincidence caused by the Chernobyl fallout in May and June coinciding with the spring wheat harvest. It is expected that, for rice, incorporation and "fixation" of ^{137}Cs in soil would have occurred prior to the autumn harvest (young rice plants are transplanted to paddy fields in May or June), so no Chernobyl peak is seen in the rice data. This also of course applies to early phase FDNPP release (also in Spring); we can expect good predictions of whole diet in the first year after fallout, but high uncertainty in predictions for other foodstuffs in the first year.

3.3. Influence of soil characteristics on radiocaesium transfers

In the longer term after fallout, uptake to the food chain is determined primarily by root uptake from soil (Hinton et al., 1996). Measurements of soil characteristics were available at sites where wheat and brown rice activity concentrations were monitored. We evaluated, by

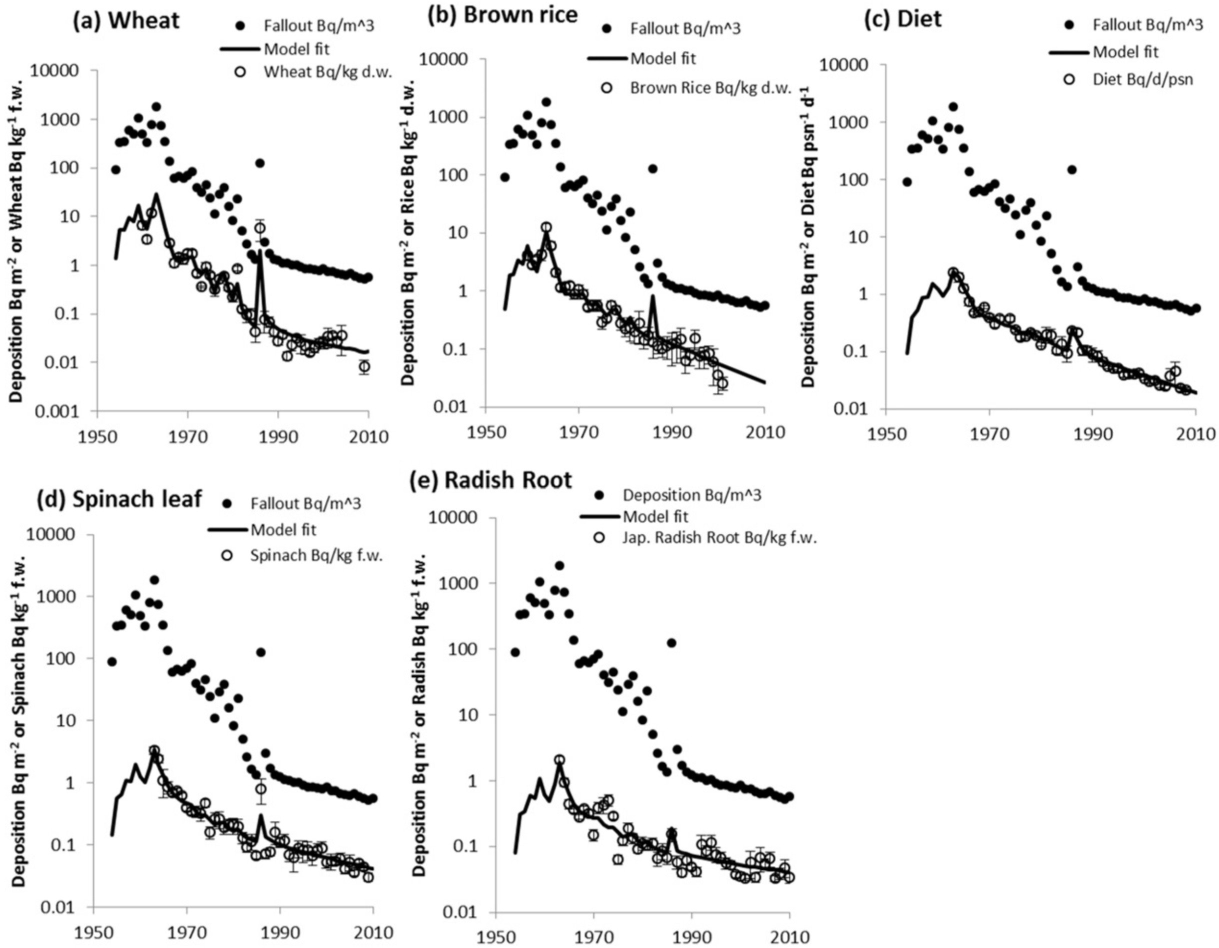


Fig. 1. Model best fits to the mean measurements of ^{137}Cs in (a) wheat, (b) brown rice, (c) whole diet, (d) spinach leaf and (e) Japanese radish (root) in Japan during the period of NWT fallout (1954–2010). Error bars show ±SE. For the 1986 Chernobyl peak, the model was only expected to give good predictions for the whole diet data (see text). Note that wheat and brown rice are measured per kg dry weight (d.w.), vegetables per kg fresh weight (f.w.). On a d.w. basis vegetable activity concentrations would be much higher.

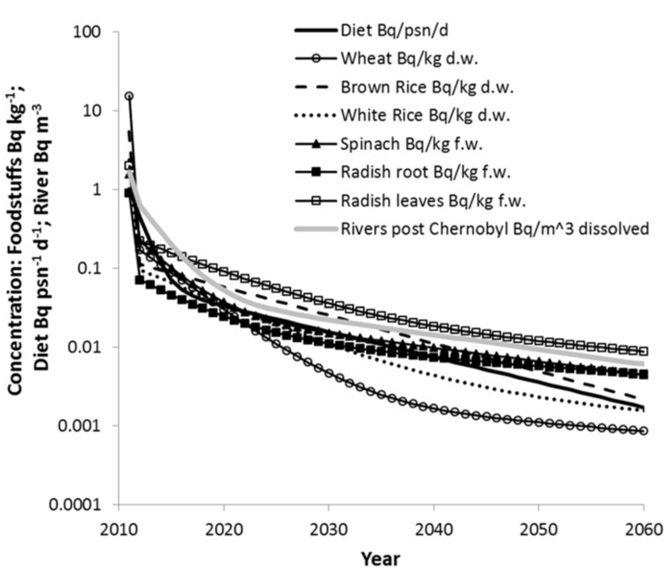
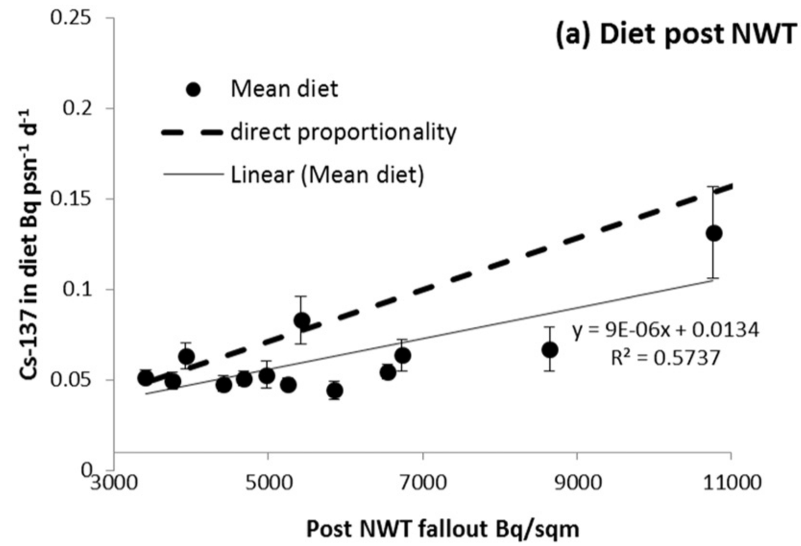


Fig. 2. Best fit modelled concentration in wheat, brown rice, white rice, vegetables and whole diet for a nominal fallout of 1000 Bq m^{-2}. In the long term (decades), these time trends in radiocaesium bioavailability are consistent with that predicted (thick grey line) in previous studies of radiocaesium in 12 large European rivers (dissolved phase) post-Chernobyl (Smith et al., 2004).

linear correlation, associations between soil density, pH, CEC, composition, and long term activity concentrations in wheat and rice following NWT (SM Fig. S5). No correlation was found between soil characteristics at each site and rate of decline (1980–99) in activity concentration in brown rice (the 1980–99 period, compared to 1985–99 used in subsequent analyses, allowed a better estimate of slope). The log-transformed mean (1985–99) activity concentration in brown rice was, however, significantly (two-tailed $p < 0.05$; $N = 13$; $r^2 = 0.43$) positively correlated with total fallout (1959–80) in each region. There was also a significant ($p < 0.05$; $N = 13$; $r^2 = 0.33$) positive correlation with percent silt and weakly significant ($p < 0.1$) positive correlation with percent clay. For wheat, there was a significant ($p < 0.05$; $N = 9$; $r^2 = 0.57$) positive correlation of the long term activity concentration with soil CEC, but no significant correlation with total NWT fallout (1959–80). Clearly, it was not possible to link whole diet with soil characteristics, but, as expected, mean concentration in whole diet (1985–99, excluding Chernobyl) was significantly ($p < 0.05$; $N = 13$; $r^2 = 0.44$) correlated with log total fallout to each site (1959–80); Fig. 3(a).

3.4. Predictions of ^{137}Cs in foodstuffs from Fukushima prefecture after NWT

Models for radiocaesium in vegetables (developed using data in >30 other prefectures of Japan) were tested against available measurements in spinach and Japanese radish root made in a range of sites in the Fukushima Prefecture during the years after NWT fallout. Fig. 4 shows that the time trend in these foodstuffs in the Fukushima Prefecture is consistent with that determined from other regions in Japan. The



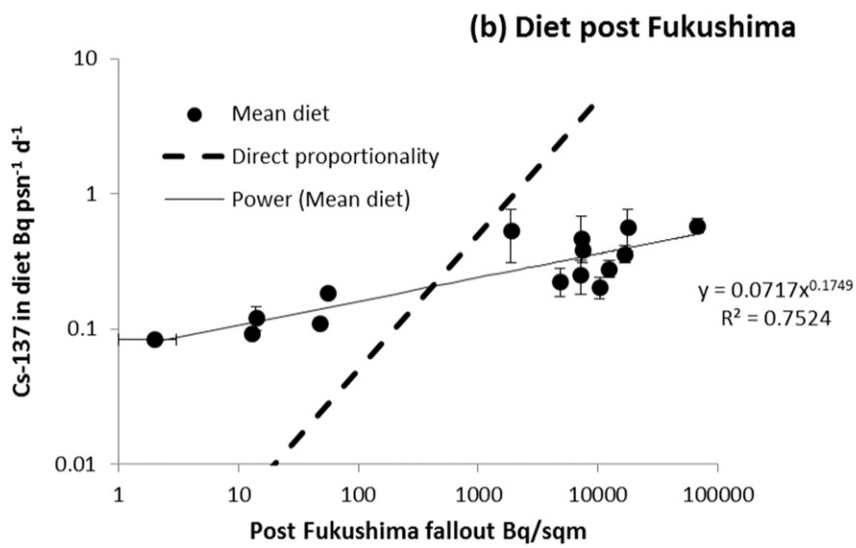


Fig. 3. Positive relationship between ^{137}Cs in diet and deposition in different regions of Japan following (a) NWT and (b) the FDNPP accident. Post-accident fallout and diet for the Fukushima Prefecture are averages for each of the three regions (Aizu, Nakadori and Hamadori) excluding the evacuated areas. The dashed lines show the expected relationship if there were direct proportionality between ^{137}Cs in whole diet and deposition. Error bars show ±SE.

models make good predictions of measured values, though variability in measured values is high. This high variability, as seen in Fig. 4 (and discussed further in SM), is due to a combination of within-site and between-site variation due to environmental factors (soil type, mineralogy, chemistry); the measurements were made at 14 (Japanese radish) and 18 (spinach) different sites of differing soil characteristics.

3.5. Predictions for ^{137}Cs in whole diet pre- and post-Fukushima

Data on average diet from five regions (Fukushima, Saitama, Kanagawa, Kochi, Nagasaki) were available from the NWT period, and had not been used in model development, so these sites provided a "blind" test of the model for NWT fallout in whole diet. Post-Fukushima data for 2012–2015 presented in (Fukushima, 2016; Uekusa et al., 2014) was used to test the model predicted time trend for the Fukushima event. Blind predictions for the NWT period showed good agreement with measured data, and the small Chernobyl peak was reproduced well (Fig. 5). For the post-FDNPP accident data, however, the model tended to over-predict ^{137}Cs in whole diet in regions of low Fukushima fallout, and under-predicted in regions of high Fukushima fallout. Only in the Iwate region ("medium" - 1884 Bq m^{-2} fallout post-Fukushima) did the model give an accurate prediction of ^{137}Cs in whole diet. Fig. 3(b) shows that the post-Fukushima regional diet activity concentrations are highly significantly ($p < 0.01$), but not strongly correlated with regional ^{137}Cs fallout. As discussed in Section 4 below, we believe that this model over-prediction in high deposition areas, and under-prediction in low, is primarily due to significant mixing of food products between regions. It is likely that food restrictions also played a role: in the early period, activity concentrations in rice, for example, approached or exceeded the 100 Bq kg^{-1} limit in areas of high contamination (Nihei et al., 2015).

Data from dietary surveys after the Fukushima accident (2012–15) were used to test the time trend in activity concentration predicted from the NWT data (Fig. 6). Good agreement is seen between the NWT predicted trend and that observed post-Fukushima; it should be noted that these dietary data only test the trend, not the predicted absolute concentrations, since the dietary data are not linked to fallout data.

3.6. Predicting cumulative effective doses from whole diet

Cumulative committed effective internal doses from radiocaesium were estimated by running the whole diet model for a 50 year period

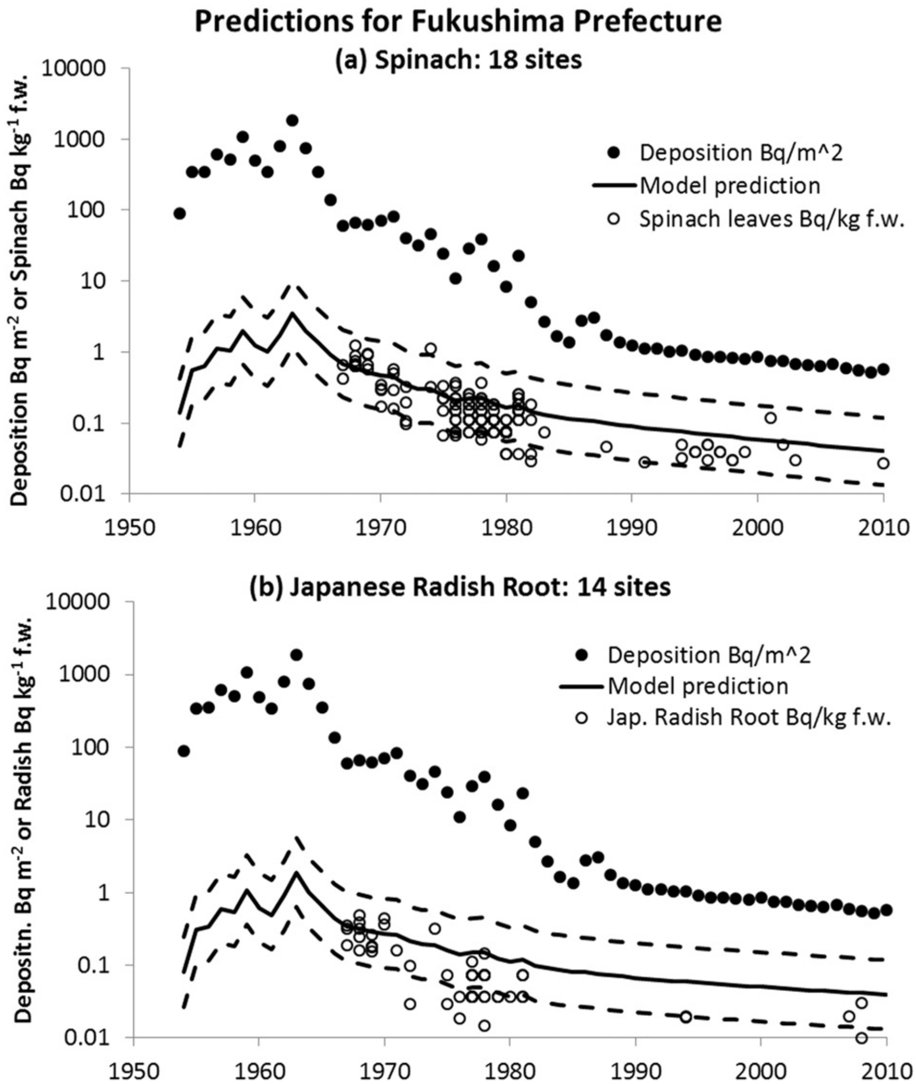


Fig. 4. Model blind prediction of ^{137}Cs in (a) spinach; (b) Japanese radish root at a range of sites in the Fukushima Prefecture after NWT. Measurement error is minor in comparison with between site variation so sample error bars are not shown.

from 2011 to 2060 for the regions relatively more contaminated by Fukushima fallout, and for the period 1954–2003 for NWT fallout. Table 1 shows estimated internal doses (2011–2061) predicted by the model under two assumptions: the first that a large proportion of produce is consumed from the local region (i.e. that is no mixing of diet between regions, and that there are no controls on consumption of foodstuffs) and the second, that significant mixing occurs between more- and less-contaminated regions (as is seen in the empirical whole diet data, and in the evidence from the Fukushima City Food Market, discussed in Section 4).

4. Discussion

The good agreement of the generic models to data in foodstuffs and diet post NWT (Figs. 1, 4, 5 and see SM Table S2) shows that time trends in activity concentrations are relatively consistent between sites, though a tendency to over-predict the low very long term activity concentrations for wheat and brown rice can be seen in SM Fig. S2. Variation from model predicted values (SM Figs. S2, S3) is also influenced by variability in deposition (as well as site-specific environmental factors) since these generic model fits did not in this case account for differences in fallout between regions. It should be noted that the models for individual foodstuffs are not expected to give accurate predictions for the short term (first year) due to differences in fallout timings between NWT and Chernobyl or Fukushima. Due to temporal and spatial averaging in whole diet (i.e. diet is composed of a range of foodstuffs from different areas and harvested at different times) the response of the whole diet variable is slower: as shown in Fig. 1 (c), the model calibrated using NWT data gives good predictions for the increase following the Chernobyl release. The spatial and temporal averaging of whole diet also leads to significantly less variation in diet data than in individual foodstuffs (Supplementary material Fig. S2).

It should be noted that a significant proportion of the Japanese diet is imported from overseas, increasing from about 35% in the late 1960′s to 60% in the late 1990′s, though it has remained constant for the last 20 years. This could potentially have influenced the observed time trends in dietary contamination, however:

(1) fallout from NWT contaminated the whole Northern Hemisphere. Imported foodstuffs (the vast majority of which are from the Northern Hemisphere) would also have been contaminated. There is strong evidence (Smith et al., 2002) that mean dietary time trends following NWT were similar in different temperate countries. The change in dietary ^{137}Cs for Japan from 1967 to 85 (the period when rates of imported foodstuffs were changing significantly) can also be compared with that in Germany for the

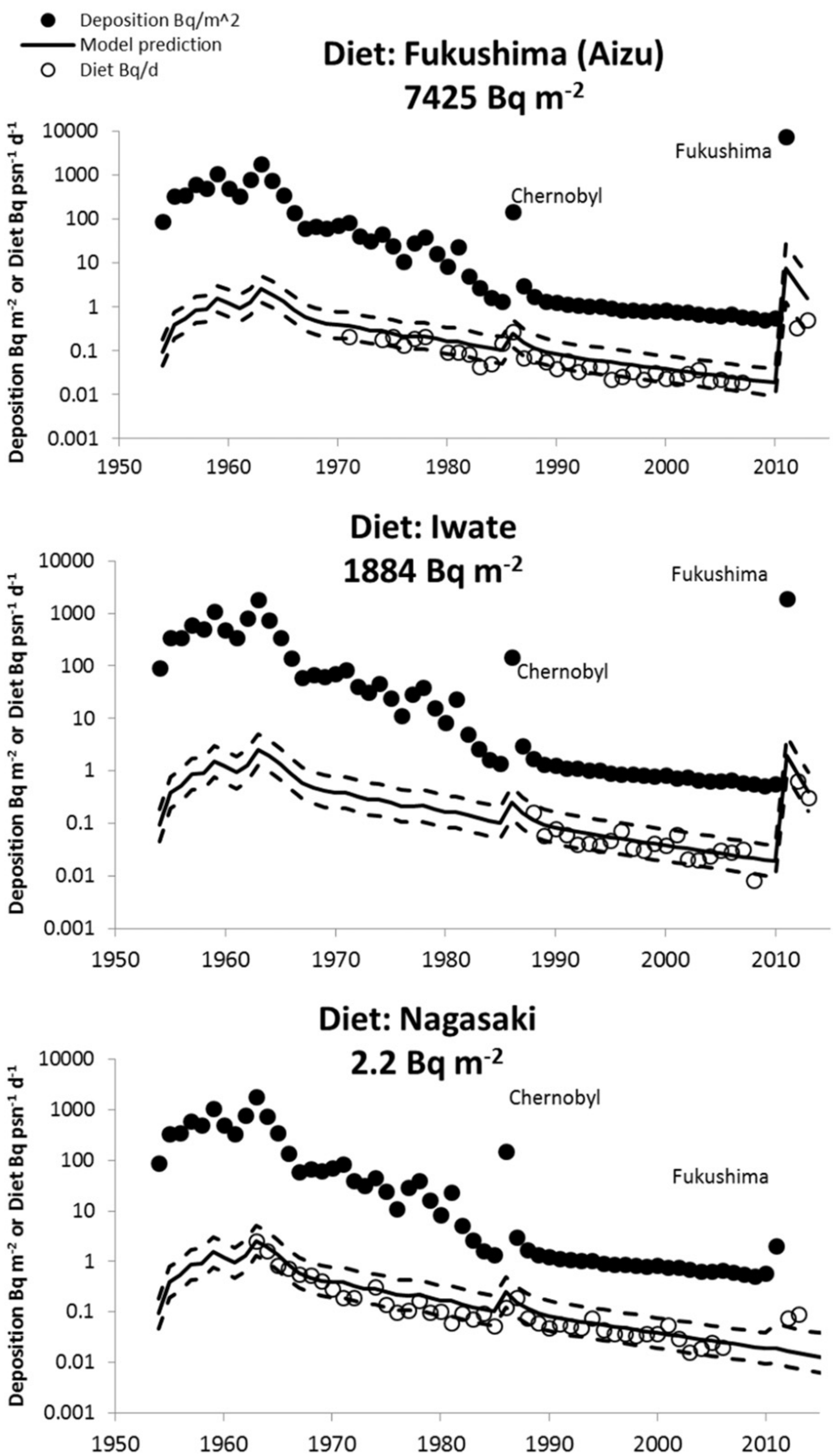


Fig. 5. Model blind prediction of ^{137}Cs in whole diet in three regions (a) Fukushima (Aizu); (b) Iwate; (c) Nagasaki, pre- and post-Fukushima. Average ^{137}Cs deposition following the Fukushima accident is also shown for each region. The small peak resulting from Chernobyl fallout is predicted well, but the model over-estimates in high fallout areas, and under-estimates in low fallout areas following Fukushima due to inter-regional mixing of foodstuffs.

same period. For the average whole diet presented here for Japan an effective ecological half-life of 7.9 years is observed for this period, close to 8.1 years observed for the same period in Germany by Pröhl et al. (2006);

(2) the empirical data show time trends in dietary Cs-137 in nine different regions of Japan to be quite consistent (Fig. S2), and long term trends in total diet are consistent with those in individual foodstuffs grown in Japan (Fig. 2). The <1:1 relationship between deposition and diet (Fig. 3a) is explained by imports of foodstuffs from other regions and countries. Nevertheless, despite a factor of three variation in fallout between regions and factor of two variation in average dietary Cs-137 (Fig. 3 a), the time trends in diet in different regions are quite consistent (Supplementary material; Fig. S2).

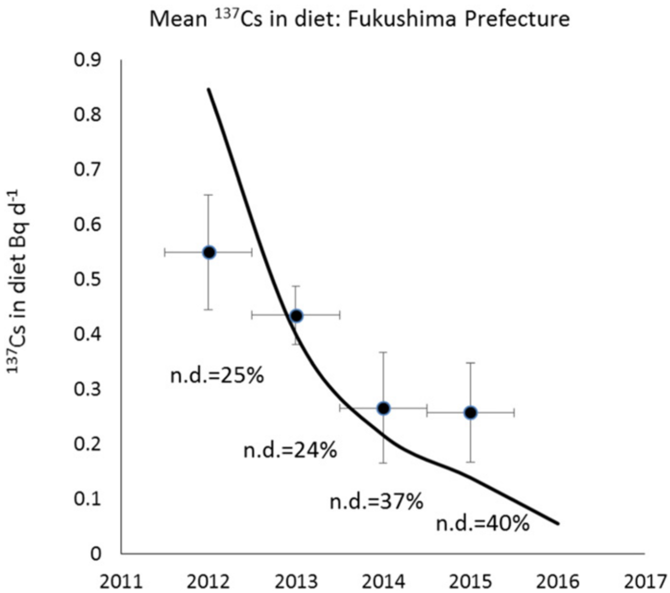


Fig. 6. Model blind predicted rate of decline in ^{137}Cs in whole diet compared to measurements in the Fukushima Prefecture 2012–15. The percentage of samples below the detection limit ("n.d.") is also shown for each year. Means were calculated based on n.d. values equal to the detection limit (L.O.D.). Y-axis error bars represent ±2 S.E., x-axis error bars represent range in sampling period.

Relatively consistent long term time trends in radiocaesium activity have previously been observed in both surface waters (dissolved phase) and milk following NWT and Chernobyl (Smith et al., 1999), the similarity between the different environmental compartments being attributed to a slowly declining pool of chemically available radiocaesium in the surface layers of soils. Fig. 2 also shows the decline in ^{137}Cs in European rivers (dissolved phase) following Chernobyl, as compared with that observed in Japanese foodstuffs and whole diet post-NWT. Though there are clear differences in rates of decline during the first 5–10 years after fallout, in the longer term, declines are similar between different foodstuffs. Wheat appears to show a more rapid rate of decline than rice, for example, as observed in a study of effective ecological half-lives (Merz et al., 2016) in these products. Comparison of long term rates (k_2 value SM Table S2) between wheat, white rice and radish (for which the "best" model was the same) show a more rapid rate of decline for wheat, but the model estimated uncertainty ranges overlapped. Rates of decline in average diet (representing an integrated measure of ^{137}Cs in foodstuffs) are highly consistent between different sites (Fig. S2), though, as expected, inter-site variability is significantly greater for individual foodstuffs.

Though time changes in activity concentration in particular agricultural foodstuffs are relatively consistent, individual site characteristics clearly affect long term root uptake. Somewhat surprisingly, higher long term (1985–99) average brown rice activity concentration was *positively* associated with % silt (SM: Fig. S5b) and % clay, though the latter was of weak significance ($p < 0.1$). A priori, one would expect a negative association with silt and clay fractions and a positive association with % sand. However Ishikawa and coworkers (Kamei-Ishikawa et al., 2008) reported that sorption sites with a high affinity for Cs ("Frayed Edge Sites") depend on the clay type so total clay content does not have a strong correlation with Cs sorption. Further, these researchers (Ishikawa et al., 2007) found that illite content was more important than total clay for fixation of Cs on Japanese paddy soils; it is known (Comans et al., 1991) that illite clays have a high affinity for Cs. Higher long term wheat activity concentrations were not associated with a particular soil fraction, but were associated with total CEC (SM Fig. 5c); high CEC may indicate the presence of higher organic matter content, and hence higher radiocaesium availability (Sanchez et al., 1999).

This evaluation of the influence of soil characteristics on transfer has identified some key influencing factors for these data, but must be considered a limited assessment since a relatively low number of sites were studied. Further, no detailed soil mineralogical analysis was available, nor was detailed soil chemistry (for example [K^+] concentration) both of which influence ^{137}Cs uptake to plants (Fesenko et al., 2007; Saito et al., 2014; Uematsu et al., 2015; Uematsu et al., 2016; Yamaguchi et al., 2016).

For post-NWT measurements, there was, as expected, a significant positive association between mean long term (1985–99) activity concentration in whole diet and fallout (Fig. 3a) with the relationship less strong than the expected directly proportional relationship, due to mixing of foodstuffs between regions of different fallout level and the high degree of imported foodstuffs. For post-Fukushima whole diet data, there was also a (statistically highly significant) correlation between average ^{137}Cs in daily diet and deposition in 2011 to a given region. However, this relationship was even weaker than for NWT: as shown in Fig. 3(b), the slope is much less steep than the line of direct proportionality. This weak correspondence between whole diet and fallout causes the model to over-predict in areas of low fallout and under-predict in areas of high fallout (Fig. 5) post-2011. This implies (for average regional diets studied here) significant mixing of produce between regions, as might be expected in Japan's highly-developed food production system. This hypothesis is supported by data on provenance of fruit and vegetables at the Fukushima City Food Market (http://www.

Table 1
Predicted and best-fit regional average cumulative doses from diet in the more contaminated regions following the Fukushima accident. Model predicted doses (assuming no food controls and mixing between areas) are significantly above the cumulative 50-year dose from NWT (98.9 μSv), but best-fit doses are less than half of those from NWT fallout. Numbers in italics show uncertainty ranges.

Prefecture	City or town	Deposition, 2011	50 year cumulative dose, μSv		Model predicted	Best fit
		Mean Bq m^{-2}	Predicted (no inter-region mixing of diet)	Best fit (significant mixing of diet between regions)	As % of NWT dose 1954–2003	As % of NWT dose 1954–2003
Miyagi	Sendai, Onagawa, Ishinomaki	12,370	146	12.8	148	13.1
		10,640–13,330[a]	*62.8–315*[b]	*6.46–25.8*[b]		
Fukushima	Hamadori Area Iwaki	17,710	209	47.8	211	48.3
		1200–40,000	*7.1–944*	*23.9–95.5*		
	Nakadori Area Fukushima	67,500	796	35.4	805	35.8
		25,000–190,000	*147–4480*	*17.7–70.8*		
	Aizu	7425	87.6	28.7	88.6	29.0
	Aizuwakamatsu	*2500–13,000*	*14.7–307*	*14.3–57.3*		
Ibaraki	Mito, Hitachinaka	16,770	198	16.9	200	17.0
		12,951–20,579	*76.4–486*	*8.43–33.7*		
Tokyo	Shinjyuku	10,310	122	8.99	123	9.1
		8759–11,867	*51.7–280*	*4.49–18.0*		

[a] Range in estimated deposition.
[b] Range in model dose estimates based on variability in deposition and model uncertainty.

city.fukushima.fukushima.jp/soshiki/24/1030.html) showing that, in 2012 and 2013, only 16% of vegetables and 25% of fruit were sourced from the Fukushima region. To a lesser extent, the weak relationship between fallout and dietary ^{137}Cs is likely also due to the effects of countermeasures in the more affected areas, and the 100 Bq kg^{-1} limit on foodstuffs.

The model-predicted rate of decline in ^{137}Cs in diet was further tested using whole diet measurements for the Fukushima region (Fukushima, 2016; Uekusa et al., 2014) for 2012–15. As shown in Fig. 6, the measurements during this period are consistent with the relatively rapid decline in radiocaesium in whole diet predicted by the model. It should be noted that the number and location of diet sample measurements varied between years, so sampling errors may exceed those shown in Fig. 6. Three high-value outliers (all in 2012/13) were removed from the set of 408 measurements. It should also be noted that that these dietary data only test the trend, not the predicted absolute concentrations, since the dietary data are not linked to fallout data. The rapid early declines in dietary activity concentration predicted from the NWT data are further supported by a study (Tsukada et al., 2016) reporting that the average concentration of radiocaesium in agricultural plants in 2013 was approximately one quarter of that observed in 2012.

As shown in Table 1, even in the more contaminated (but not evacuated) parts of the Fukushima region, 50 year cumulative doses via intake of foods are significantly below those from NWT, because of measures to protect the food chain and the high degree of mixing between regions. If no mixing between regions is assumed, cumulative doses in the more contaminated areas are significantly greater than those from the NWT period (Table 1). A previous study (Harada et al., 2014) of three areas in the Hamadori region of Fukushima Prefecture observed mean dietary ^{137}Cs from 0.68 to 2.2 Bq/d in 2012 that compares with the value of 0.85 Bq/d for 2013 used in the present study for Hamadori (Uekusa et al., 2014). The annual ingestion dose rates of several μSv year^{-1} in 2012/13 estimated by (Harada et al., 2014) for the Hamadori region are in broad agreement with those estimated in the present study which estimated 2–8 μSv year^{-1} for 2013. However, predicted ingestion dose rates (Harada et al., 2014) in 2060 are significantly higher than in the present study since the former (Harada et al., 2014) only considered physical decay of ^{137}Cs with half-life 30.2 years, whereas the present study determined a more rapid (changing) rate of decline with, in the long term, an effective ecological half-life of order 8 years. The study of effective ingestion doses by the UN Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2013) estimated, using the FARMLAND model (Brown and Simmonds, 1995), a 10 year cumulative dose to adults of the non-evacuated parts of the Fukushima Prefecture of 1380 μSv, assuming that all food was locally sourced. This compares with 150–600 μSv (10 year cumulative dose) calculated by the model presented here, assuming an average fallout of 31 kBq m^{-2} for the Fukushima Prefecture. These two assessments can be considered to be in broad agreement, given the inherent uncertainties in dose assessment, and that the present study did not account for other radionuclides, particularly ^{134}Cs.

It should be noted that these predicted doses are regional averages and do not include, for example, higher doses to people who consume "wild" foodstuffs such as mushrooms, berries, game animals (Merz et al., 2015; Tagami et al., 2016), and freshwater fish. After the Chernobyl accident, in some areas, rates of decline in ^{137}Cs in foodstuffs from semi-natural ecosystems were observed to be very slow, in some cases being close to the physical decay half life of 30.2 years (Smith and Beresford, 2005). Nor, of course, do the predictions include doses which would arise from consumption of foodstuffs from highly localised areas of higher (or lower) fallout, or consumption of produce from evacuated areas. The two scenarios outlined (assuming both the current high degree of mixing, and assuming only locally produced food is consumed) give a best estimate and upper bound prediction for the regional average integrated dose. Potential future changes in regional imports of foodstuffs could alter the best estimate ingestion dose, but the maximum would be expected to remain the same. Finally, it should be noted that these doses are only calculated for ^{137}Cs and do not include potential doses from other isotopes released by the Fukushima accident.

It is noted that there is evidence of discharges of spherical cesium particles from the Fukushima accident (Abe et al., 2014; Adachi et al., 2013; Satou et al., 2016). These particles had a relatively high concentration of radiocaesium, however, only a small number of particles were reported (Abe et al., 2014; Adachi et al., 2013; Satou et al., 2016), so we assumed that the activity contribution from the Cs particles to the total Cs deposited on land surface would be negligible. Moreover, the particles were less soluble in water which means less is available to plants through the root uptake process. Hence the presence of particles, if significant, would lead to model over-prediction of internal dose rates.

5. Conclusions

This study developed models for prediction of an acute fallout of radiocaesium (such as those from the Chernobyl and FDNPP accidents) using time changes in ^{137}Cs following NWT. The model fitting used a procedure which accounted for the fact that the NWT fallout occurred over many years allowing predictions to be made for the relatively short deposition event from the Chernobyl and FDNPP accidents. Good consistency in time trends for dietary ^{137}Cs between different regions post NWT gives confidence in predictions of ingestion doses for 50 years post-Fukushima. The identified time trends were in broad agreement with whole diet data post-Fukushima, though it is acknowledged that these test data are limited in their time span, and may have been influenced by food restrictions.

Total ^{137}Cs fallout from NWT of 2.8 PBq (estimated from data in (Aoyama et al., 2001)) to the land surface of Japan is similar to 2.75 PBq (Bailly du Bois et al., 2014) from the FDNPP accident. Obviously, the much greater impact of Fukushima is due to the much more highly spatially concentrated nature of the fallout compared to NWT. However, given the measures taken to protect the food chain following Fukushima, and apparently highly interconnected nature of the Japanese food system, it appears that the long term impact of radiocaesium from Fukushima on the average Japanese diet is similar to that from NWT, even in the most contaminated (not evacuated) regions. It should be emphasised that variation in whole diet amongst individuals with different food consumption habits is not within the scope of this study of regional average diet. However, the consistency in time trends in radiocaesium in diet between NWT, Chernobyl and the first years after Fukushima gives confidence that the wealth of information obtained by Japan during the period of NWT fallout is likely to be a good guide to predicting the long term consequences of the Fukushima accident. This study could form a basis for site specific predictions of long term activity concentrations in agricultural products both inside and outside currently evacuated areas.

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Conflict of interest statement

Keiko Tagami and Shigeo Uchida work for a research institute supported by the Japanese government. Jim Smith has a project funded by the UK Natural Environment Research Council, Radioactive Waste Management Ltd. and the Environment Agency of England and Wales. He has also carried out small (<£5 k) consultancy projects for a range of clients including the Japan Atomic Energy Agency and UK Radioactive Waste Management Ltd.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.05.240.

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