Worldwide isotope ratios of the Fukushima release and early-phase external dose reconstruction

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Measurements of radionuclides (RNs) in air made worldwide following the Fukushima accident are quantitatively compared with air and soil measurements made in Japan. Isotopic ratios RN:137Cs of 131I, 132Te, 134,136Cs, are correlated with distance from release. It is shown, for the first time, that both within Japan and globally, ratios RN:137Cs in air were relatively constant for primarily particle associated radionuclides (134,136Cs; 132Te) but that 131I shows much lower local (<80 km) isotope ratios in soils relative to 137Cs. Derived isotope ratios are used to reconstruct external dose rate during the early phase post-accident. Model "blind" tests show more than 95% of predictions within a factor of two of measurements from 15 sites to the north, northwest and west of the power station. It is demonstrated that generic isotope ratios provide a sound basis for reconstruction of early-phase external dose rates in these most contaminated areas.

Following the accident at the Fukushima Dai-ichi (I) Nuclear Power Station (NPS), many measurements were made worldwide of radioisotopes in air. In Japan, the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) established daily measurement of gamma dose rate and deposited activity at a number of sites around the Fukushima I NPS, and within the highly contaminated area to the north and west (W, NW, N) of the plant. There were, however, relatively few sites where gamma-ray dose and deposited radioactivity monitoring were available to evaluate early-phase external dose rates, particularly in the Fukushima Prefecture1. ("Early-phase" is here defined as within the first 30 days following the accident, before external dose was dominated by relatively long-lived 134,137Cs.) Since external dose rates declined rapidly due to decay of short-lived radionuclides2, and external dose rates in other countries could not be measured directly, models are required for reconstruction of early-phase dose rates. This is of particular importance for the highly contaminated region to the northwest of Fukushima I NPS, some parts of which were still inhabited during the weeks after the accident3.

This study represents the first systematic review of available air and soil monitoring data both in the highly contaminated areas of Japan and worldwide. Potential isotopic fractionation is evaluated over the long distances (from 10's of km to thousands) travelled, as was observed for a number of radionuclides after Chernobyl7. The study of the variation in isotope ratio with distance provides supporting evidence for a model to reconstruct early-phase external dose rates. Since there are relatively few measurements of early-phase dose rates, radionuclide depth profiles and isotopic composition, it is necessary to parameterise models for external dose rate based on generalized radionuclide isotope ratios. The model presented here, for the first time, generalises external dose models5,6 to sites at which dose rate, isotopic composition and depth-distribution in soils data are not available. It is noted that, whilst isotope ratios of primarily particulate bound radionuclides were relatively even in the area to the north and west (W, NW, N) of Fukushima prefecture (formed largely by the 15 March release and rapid washout in rainfall), fallout to the south and southwest was also influenced by the March 21–23 rainfall event and characterized by more variable isotope ratios7. We will here focus on the area to the north, northwest and west for which external dose rate data are available for model testing and calibration, though in principle the approach could be used for other areas.

Results

Mean isotope ratios. The change in radionuclide (RN) ratios RN:137Cs in air (particulate phase) and soil with distance from the Fukushima I NPS are shown in Figure 1 and Table 1. As shown in Table 1, 131I/137Cs isotope...
Figure 1 | Average corrected ratios to $^{137}$Cs of $^{131}$I, $^{134,136}$Cs and $^{132}$Te in (a) soils and (b) air particulates (data for gaseous-phase $^{131}$I are shown in Figure 3). Many of the $^{131}$I measurements in air taken in the vicinity of Fukushima NPS were excluded from (b) as sampling did not begin until some days after the radionuclide release.

Table 1 | Average corrected mean ratios to $^{137}$Cs from release of Fukushima ($\pm 2$ S.E., based on variation of means between sites)

<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Soil samples in near-zone (&lt;80 km)</th>
<th>Air samples in near-zone (&lt;80 km)</th>
<th>Particulate</th>
<th>Air samples in Japan (80–2,000 km)</th>
<th>Particulate</th>
<th>Air samples in Pacific Ocean and US (2,000–12,000 km)</th>
<th>Particulate</th>
<th>Air samples in EU (&gt;12,000 km)</th>
<th>Particulate</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{131}$I</td>
<td>22.5 ± 3.7</td>
<td>31.8 ± 12.9 *</td>
<td>70.9 ± 63.5</td>
<td>69.5 ± 26.9</td>
<td>77.1 ± 16.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sites = 145</td>
<td>Sites = 22</td>
<td>Samples = 90</td>
<td>Samples = 234</td>
<td>Samples = 457</td>
<td>Samples = 196</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Samples = 1,844</td>
<td>18.3 ± 1.7</td>
<td>No data</td>
<td>17.5 ± 2.1</td>
<td>17.1 ± 4.2</td>
<td>22.0 ± 6.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{132}$Te</td>
<td>0.09 ± 0.01</td>
<td>1.03 ± 0.10</td>
<td>0.99 ± 0.21</td>
<td>0.87 ± 0.06</td>
<td>0.91 ± 0.07</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sites = 146</td>
<td>Sites = 17</td>
<td>Samples = 68</td>
<td>Samples = 217</td>
<td>Samples = 455</td>
<td>Samples = 196</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Samples = 1,866</td>
<td>0.22 ± 0.01</td>
<td>No data</td>
<td>0.19 ± 0.03</td>
<td>0.22 ± 0.04</td>
<td>0.20 ± 0.03</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Note that these measurements began 4–5 days after the passage of the initial plumes so, because the $^{131}$I/$^{137}$Cs ratio varied with release time, cannot be directly compared with other data and are not included in Figure 1.
ratios in soil deposits in the area within 80 km of Fukushima I NPS (mean ratio $^{131}$I/$^{137}$Cs = 22.5) were low compared to those in air particulates worldwide (ratio ca. 70–80). The measurements in soil in the key region to the NW of Fukushima I NPS (Figure 2) show a highly significant inverse correlation between the $^{131}$I:$^{137}$Cs ratio and the $^{137}$Cs activity concentration (Spearman’s correlation coefficient $-0.54; N = 89; p < 0.001$). The best fit equation (using the Akaike Information Criterion $^a$) to these data was of the form

$$^{131}I : ^{137}Cs \text{ ratio} = a(C_{^{137}Cs})^b + c \quad (1)$$

as shown in Figure 2 in comparison with a standard power law fit (i.e. excluding the intercept $c$).

Parameter values for the fitted equations are shown in Figure 2. [Model fits were carried out on log-transformed data of the concentration of each isotope in soil ($C_{^{131}I}$ vs $C_{^{137}Cs}$) then back-transformed and expressed as a ratio $^{131}$I:$^{137}$Cs for presentation of results.] Note that Equation (1) is only applicable in the high fallout areas ($>c\alpha$, 300 Bq kg$^{-1}$ of $^{137}$Cs) to the north, northwest and west of Fukushima I NPS. For lower $C_{^{137}Cs}$ fallout areas, a generic value of 76 could be used (the value of the ratio at 300 Bq kg$^{-1}$ of $^{137}$Cs). The value of 300 Bq kg$^{-1}$ is the lower bound of available $^{131}$I:$^{137}$Cs measurements in soil (see fig. 2). The choice of this lower bound value ($^{131}$I:$^{137}$Cs ratio $= 76$ at 300 Bq kg$^{-1}$ of $^{137}$Cs) is supported by the far field particulate air monitoring data in Japan, Europe and North America (ratio $= 70–80$, Table 1). Given efficient washout of the particulate form of $^{131}$I the peak air concentrations from the early stages of the accident (15/16 March) were only available at two sites: highly significant inverse correlation between the $^{131}$I:$^{137}$Cs ratio and particulates worldwide (ratio ca. 70–80). The measurements in soil (see fig. 2). The choice of this lower bound value ($^{131}$I:$^{137}$Cs ratio $= 76$ at 300 Bq kg$^{-1}$ of $^{137}$Cs) is supported by the far field particulate air monitoring data in Japan, Europe and North America (ratio $= 70–80$, Table 1). Given efficient washout of the particulate form of $^{131}$I the far-field air particulate ratio is likely to be more accurate, though the higher gaseous phase ratio (discussed below) could be used if conservative estimates of dose are required. It is noted that fallout to the south and southwest of Fukushima I NPS was also influenced by the March 21 rainfall event and characterized by different isotope ratios$^c$; more site-specific ratios may be required for these areas.

Measurements of gaseous and particulate $^{131}$I in Japan in the early stages of the accident (15/16 March) were only available at two sites: Nuclear Science Research Institute, Tokai-mura$^a$ and the High Energy Linear Accelerator Research Centre, Tsukuba$^a$ (respectively, 120 and 160 km south of Fukushima I NPS). Measurements from other sites worldwide began later, but due to longer travel times, captured the peak air concentrations from the early stages of the accident. Figure 3 shows the available mean $^{131}$I:$^{137}$Cs ratios of both gaseous and particulate phase $^{131}$I as a function of distance.

**External dose model.** Equation (2) (Methods section, below) was used to quantify the time change in external dose rate at each of six sites to the north and west (W, NW, N) of Fukushima I NPS (for map see Supplementary Fig. S1). The measured radioactivity concentration in soil at each site was used as the independent, the measured dose rate as the dependent variable in a linear regression model (Eq. 2, Methods section). Activity concentrations of other radioisotopes were estimated using the generic mean RN:$^{137}$Cs ratios in Table 2. For $^{131}$I, the relation between $^{131}$I:$^{137}$Cs ratio and measured $^{137}$Cs activity concentration was used (Eq. 1; Figure 2). For other radionuclides, mean isotope ratios measured in soil within 80 km were used (longer-range isotope ratios were observed to be similar). Model fits to measured external gamma dose rate are shown in Figure 4 for the six sites for which “good” time-series of dose rate were available; “good” means that the investigated period is exhaustively covered by a high number of observations.

Given that only one site-specific input parameter ($^{137}$Cs activity concentration) and one fitting parameter ($A$, a measure of absorption of gamma rays in the soil) are used in the model, the fits show good agreement with observations. This confirms the accuracy of the derived mean isotope ratios. A slight tendency to under-predict dose rates in the period from 20–80 days is likely due to differences in absorption in soil at different sites, and (relatively small) local variation in isotope ratios. The model sensitivity to uncertainty in $^{131}$I:$^{137}$Cs ratio, and in the value of $A$ is evaluated in Figures 5 and 6.

In Fig. 5, the data are shown together with the model using the mean value of $A$, the mean radionuclide ratios (Table 2) (bold line), and the model with the mean $^{131}$I:$^{137}$Cs ratio $r$ (Eq. 1 parameterized as in Table 2) replaced by $r \pm 2\sigma(R)$ (dotted curves). Sigma ($\sigma$) represents the standard deviation in the residuals of the fit of Eq. 1 to observed data (std. deviation of $r_{\text{measured}}/r_{\text{modelled}}$ where $r$ is the $^{131}$I/$^{137}$Cs ratio: these were normally distributed; Kolmogorov-Smirnov test in IBM SPSS Statistics 20). In Fig. 6, the data are again shown with the model, using the mean value of $A$ and the mean radionuclide ratios (Table 2) (bold line), and the model with the minimum and maximum observed $A$ values (6.0–14.2) instead (dotted lines).

**Blind testing of the model.** The external dose model was blind tested by comparing predictions of the change in external gamma dose rate with time against measurements at a further fifteen sites. Figure 7 plots model blind predicted dose rates against those measured at four of the fifteen test sites (see Supplementary Figure S4 for the remainder). Figure 8 compares model predictions against data
from six calibration sites plus the fifteen test sites. Ninety-five percent of measured values are within a factor of two of predictions. The pattern of data in Figure 8 (b) appears “stepwise” because of short term variability in the measurements at a given site (likely due primarily to soil moisture content and measurement uncertainty). The model does not (and does not aim to) reproduce this variability. This results, for longer (>30 days) time periods, in variation in the measured value with little variation in the model prediction for a given site, giving the apparent “stepwise” pattern. The uncertainty in the measurement of external dose rate is expected to be small in comparison with uncertainties introduced by varying isotope ratios at individual sites (particularly 131I:137Cs ratio) and varying early-phase depth distributions.

Discussion
As was observed after Chernobyl, ratios of the radiocaesium isotopes (134Cs:137Cs, 136Cs:137Cs) show low variability and are relatively constant with distance (as observed in Japan). It should be noted, however, that there appears to be a slightly higher 134Cs:137Cs ratio in air within 80 km of Fukushima I NPS than 80 km (Table 1), potentially due to differences between release events: 134Cs:137Cs ratios may vary according to the source of contamination depending on fuel age in a given reactor. A shift of this isotope ratio had been noted in Europe and attributed to the small fraction of 137Cs released during the Chernobyl accident or nuclear weapons test fallout still present in the atmosphere. However, radiocaesium concentrations in Japan were too high to be significantly influenced by atmospheric 137Cs from these sources. The mean 132Te:137Cs ratio shows relatively little difference between air concentrations and soil deposits, and no evidence of fractionation with distance (though there is significant variability in values from different sites). This is likely because both are mainly transported as fine particulates. The slight variations in ratios of primarily particulate-bound radionuclides would not significantly affect external dose rate predictions.

In contrast to the primarily particulate-borne radionuclides, the (particulate) 131I:137Cs ratios in air are significantly more variable, and are much higher, on average, than those measured in soil near to the Fukushima I NPS. Apparently lower ratios in air are observed within 80 km of the Fukushima I NPS (Table 1) compared to those measured in the U.S. and Europe. These data, however, must be interpreted with caution since significant air monitoring in the Fukushima area only began 4–5 days after passage of the initial

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Table 2 | Summary of the parameter values used in the predictive model*

<table>
<thead>
<tr>
<th>Radioelements</th>
<th>Decay constant [d(^{-1})]</th>
<th>Emitting gamma energy [MeV/nT]</th>
<th>Ratio to 137Cs</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>131I</td>
<td>8.64E-02</td>
<td>0.3828</td>
<td>4.89 (\times) 10(^{2}) (\times) C(_{137Cs})(^{-0.76}) + 10.22</td>
<td>9.17 (\times) 10(^{-5})</td>
</tr>
<tr>
<td>132Te</td>
<td>2.16E+01</td>
<td>0.2344</td>
<td>18.30</td>
<td></td>
</tr>
<tr>
<td>132I</td>
<td>2.16E+01</td>
<td>2.2645</td>
<td>0.90</td>
<td></td>
</tr>
<tr>
<td>134Cs</td>
<td>9.20E+04</td>
<td>1.5551</td>
<td>0.22</td>
<td></td>
</tr>
<tr>
<td>134Cs</td>
<td>5.27E+02</td>
<td>2.1283</td>
<td>1.00</td>
<td></td>
</tr>
<tr>
<td>132Cs</td>
<td>6.29E+05</td>
<td>0.5963</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*\(R_{131I}\) is the ratio of 131I:137Cs and \(C_{137Cs}\) is the mean activity concentration of 137Cs (Bq/kg) in soil. The relationship for \(R_{131I}\) is only valid in areas of relatively high fallout \((C_{137Cs} > 300 \text{ Bq kg}\(^{-1}\))\). For areas of lower fallout, a value of \(R_{131I} = 76\) should be used.

\(132I\) is in secular equilibrium with \(132Te\) hence follows the \(132Te\) decay curve.
contamination plumes. Thus much of the local (<80 km) $^{131}$I/$^{137}$Cs air monitoring data are not shown in Figure 1b.

The limited data for two Japanese sites on 15 March (peak release period) suggests lower $^{131}$I/$^{137}$Cs ratios in both particulate and gaseous phases at this time, as illustrated in Figure 3 (mean of two sites: 14.6 for particulate $^{131}$I/$^{137}$Cs and 10.3 gaseous $^{131}$I/particulate $^{137}$Cs), and much lower fraction of $^{131}$I in the gaseous phase. The observed low ratios of $^{131}$I relative to $^{137}$Cs in soil deposits in areas of high fallout (Fig. 2) are consistent with these limited data in air in Japan on 15th March (Fig. 3) and with the expected ratio in the source term at this time. Thus, the constant in the regression equation (Eq. 1, Fig. 2: $c = 10.2$ with 95% confidence interval 7.7–12.8) for the iodine ratio in soil is similar to the limited measurements in air in Japan on 15th/16th March. This likely reflects the fact that the contamination plume to the northwest of Fukushima I NPS was formed from deposition with low $^{131}$I:$^{137}$Cs isotope ratio from the peak release on 15th March. Rain

Figure 4 | Model fits to measured gamma dose rates obtained using generic isotope ratios for radiocaesium isotopes and $^{132}$Te, and using a regression Equation (Eq. 1) for $^{131}$I; the A parameter is fitted to the data using SAS (SAS Institute Inc.).
was observed in the Fukushima prefecture during the night from 17:00 on March 15 to 04:00 on March 16: this time corresponded with significant emissions.

The relatively low $^{131}$I:$^{137}$Cs isotope ratios observed in the areas of high fallout and in the 15th March air monitoring data in Japan (Figs. 2 and 3) are not seen in the measurements in other countries. The $^{131}$I:$^{137}$Cs ratios in both phases at longer distances were much higher, even in the peak corresponding to the early (12–15 March) releases from Fukushima. Mean values were 73.1 (N=48 range 12–290) for $^{131}$I:$^{137}$Cs particulate and 269 (N=13 range 78–853) for the ratio of gaseous $^{131}$I to particulate $^{137}$Cs. The much higher $^{131}$I:$^{137}$Cs isotope ratios observed at long distances suggest highly efficient deposition of the 15th March release (characterised by low I/Cs ratios$^{13}$) in the area to the northwest of Fukushima I NPS.

There is no clear evidence of a significant change in $^{131}$K:$^{137}$Cs ratio over long distances in either airborne particulate or gaseous phases (Fig. 3). But there appears to be a (non-significant) trend of increasing ratio with distance potentially implying more efficient deposition of the 15th March release (characterised by low I/Cs ratios$^{13}$) in the area to the northwest of Fukushima I NPS.

The external dose model shows good agreement with data from the fifteen blind test sites (Figures 7, 8 and Supplementary Fig. S4), implying that the model and parameter values are generally applicable to other sites in areas to the north, northwest and west of Fukushima I NPS. Given the consistency of isotope ratios at greater distances (and good agreement between “low fallout” $^{131}$I/$^{137}$Cs ratio of 76 and the mean of 70–80 observed in the long distance data in Table 1), the model could be used to estimate the very much lower external dose rates in other countries.

was based on relatively few data and, in the Chernobyl near-zone, radionuclide deposition rates may have exceeded $^{131}$I due to fallout of larger airborne particulates including fuel particles$^{14}$.

Mean fractions of gaseous phase $^{131}$I were relatively constant with distance (Fig. 3), with mean gaseous fraction 80.4% (standard error (S.E.) 1.3%; range 49.2–93.4%). The consistency of gaseous fractions of $^{131}$I over long distances implies relatively little transfer from gaseous to particulate form. As discussed by Masson et al.$^{15}$, “$^{131}$I [from Fukushima] remains mainly in its gaseous form during transport. Thus, the transfer from gaseous to particulate form, if it exists, was not sufficient within the two-week interval to counterbalance the decrease of particulate $^{131}$I due to its deposition, mainly by rain.”

The external dose model shows good agreement with data from the fifteen blind test sites (Figures 7, 8 and Supplementary Fig. S4), implying that the model and parameter values are generally applicable to other sites in areas to the north, northwest and west of Fukushima I NPS. Given the consistency of isotope ratios at greater distances (and good agreement between “low fallout” $^{131}$I/$^{137}$Cs ratio of 76 and the mean of 70–80 observed in the long distance data in Table 1), the model could be used to estimate the very much lower external dose rates in other countries.

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**Figure 5** | Model results using a generic mean $A$ value and isotope ratios (bold lines) with ±2σ uncertainty in estimation of I-131/Cs-137 ratio (dotted lines) compared with the measurement data at two of the six calibration sites. The values of $A$ and isotope ratios were the generic values from Table 2: not fitted to the data. The graphs for the other four sites are shown in Supplementary Information Figure S2.

**Figure 6** | Model results using generic $A$ value and isotope ratios (Table 2) (bold lines) with ±2σ uncertainty in estimates of the A-parameter (dotted lines) compared with the measurement data at two calibration sites. The graphs for the other four sites are shown in Supplementary Information, Figure S3. The values of $A$ and isotope ratios were the generic values from Table 2: not fitted to the data.
The fitted values of the $A$ parameter (primarily a measure of absorption of gamma rays in soil) at the six calibration sites were in a relatively narrow range ($6.0–14.2 \times 10^{-2}$) with mean $9.2 \times 10^{-2}$. This relatively narrow range gives confidence that values of this unknown parameter can be extrapolated to reconstruct external dose rates at other sites, on the basis only of the measured $^{137}$Cs activity concentration. It also implies that, in the early phase, soil self-shielding was broadly similar across the different sites.

The results of the model sensitivity analysis (Figures 5, 6 and Supplementary Figures S2, S3) are consistent with the factor of two uncertainty range of model predictions in blind testing, implying that the value of the $A$ parameter, representing to a large extent soil shielding, and of the $^{131}$I:$^{137}$Cs ratio, are the greatest source of model uncertainty. Under conditions of high rainfall, radioceasium can penetrate relatively deeply into the soil profile, however in most areas affected by Chernobyl and at Fukushima, the majority of radioactivity remained near the soil surface, limiting gamma-ray absorption within the soil. Over years to decades after fallout, gamma dose rates will decline due to radioactive decay and further slow penetration of radioceasium into the soil profile.

In summary, uncertainty in model predictions results from: attenuation and shift of the energy spectrum by penetration of fallout into the soil; geometry of the dose rate monitors (i.e. response to given contamination); uncertainty of the I/Cs ratio, and possibly different fallout episodes with different radionuclide ratios. Nevertheless, the model shows good agreement with measured dose rates against test data, providing strong evidence for the ability of such models to extrapolate dose rates at sites for which early phase dose rates and isotopic composition are not available. In principle, the model could be applied to other areas of Japan, outside the areas to the north and northwest considered here, by adapting in particular to take account of the important second deposition event (21 March) to the south and southwest.

**Methods**

**Isotope ratios and key radionuclides.** Measurements of radionuclides in air and soil worldwide were collated. In addition, we use measurements at fourteen sites in Europe summarised in Masson et al. and one site in Ukraine (this study, see Supplementary Table S1). Most of the available measured data are of the relatively long-lived isotopes of radioceasium ($^{134}$Cs, $^{137}$Cs), so ratios of all radionuclides to $^{137}$Cs were determined. In total, we collected 1866 measurements from 147 soil sampling sites and 1112 measurements from 114 air sampling stations. All ratios were corrected to 11 March 2011 at 14:46 when the earthquake occurred and the reactors of Fukushima I NPS were shutdown, so the generation of fission products was effectively stopped. The total gamma decay energies of all radionuclides and their decay products were determined from ICRP Publication 107 along with their decay constants. These data were then analysed (on the basis of emission energies, half-life and information on contamination levels) to determine eight radionuclides (Supplementary Table S2) which could potentially have made a significant contribution to external gamma-ray dose rate in the weeks and months after the accident. In the first days after the accident, external dose was dominated by $^{132}$Te and $^{132}$I, followed by $^{131}$I for a period of several weeks, after which the radioceasium isotopes ($^{134,136,137}$Cs) were dominant.

Note that the contribution of $^{132}$I is due to ingrowth from $^{132}$Te; the direct release from the reactor does not significantly contribute due to its short half-life (2.295 h), $^{132}$Te and $^{132}$I were also found around Fukushima I NPS, and are included in...
Figure 8 | Comparison of predicted dose rates (uSv/h) and measured data from (a) first measurement up to 30 days and (b) more than 30 days after the accident when the gamma dose rate were dominated by short-lived ($^{132}$Te/$^{132}$I and $^{131}$I) and long-lived ($^{134}$/$^{137}$Cs) radionuclides respectively. Dotted lines show a factor of two over- and under-estimation of model predictions. As expected, model predictions at a site to the south of the NPS are poorer than those to the north and northwest due to the March 21 fallout event to the south.
Model. In the initial period, before significant penetration of radionuclides into the soil, the external gamma ray dose rate is determined primarily by level of radioisotope deposition and decay energy. We therefore estimate the external gamma dose rate, $D (\mu S v \cdot h^{-1})$, at 1 m above the soil surface by:

$$D(t) = A \cdot C_{\text{est}} \cdot \sum \frac{E_i}{R_i} \cdot e^{-t/d}$$

(2)

Where $E_i$ is total gamma decay energy of each nuclide (MeV) (see Supplementary Table S2), $C_{\text{est}}$ is the activity concentration of $^{137}$Cs in the surface 5 cm of soil, $R_i$ is ratio of deposition of each nuclide to $^{137}$Cs, $\lambda$ is the decay constant of each nuclide ($d^{-1}$), and $d$ is time after the earthquake. $A (\mu S v \cdot h^{-1} \cdot$ MeV $^{-1} \cdot$ Bq $^{-1}$) is an empirically-determined constant to convert gamma decay energy to gamma dose, being dependent on absorption in soil and air. Since, in the early phase, the majority of radioactivity is on or close to the soil surface, the model neglects energy-dependence of absorption, but the $A$ parameter accounts for potentially different distributions of radionuclides with depth at different sites.

Using mean ratios of each radionuclide to $^{137}$Cs, Equation (2) was fitted to measurement of changes in external gamma-ray dose rate with time at six sites at a distance 24-40 km from Fukushima I NPS. Model fits were carried out in SAS (PROC NLIN; SAS Institute Inc.) to determine the value of the one unknown parameter, $A$. The model was then “blind” tested against data from a further fifteen sites for which (more limited) dose rate measurements were available.

3. Tokonami, S. The model was then “blind” tested against data from a further fifteen sites for which (more limited) dose rate measurements were available.

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