SCIENTIFIC REPORTS

OPEN

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:^{137}Cs of ^{131}I, ^{132}Te, ^{134,136}Cs, are correlated with distance from release. It is shown, for the first time, that both within Japan and globally, ratios RN:^{137}Cs in air were relatively constant for primarily particle associated radionuclides (^{134,136}Cs; ^{132}Te) but that ^{131}I shows much lower local (<80 km) isotope ratios in soils relative to ^{137}Cs. 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 Prefecture[1]. ("Early-phase" is here defined as within the first 30 days following the accident, before external dose was dominated by relatively long-lived ^{134,137}Cs.) Since external dose rates declined rapidly due to decay of short-lived radionuclides[2], 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 accident[3].

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 Chernobyl[4]. 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 models[5,6] 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 ratios[7]. 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:^{137}Cs 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, ^{131}I/^{137}Cs isotope
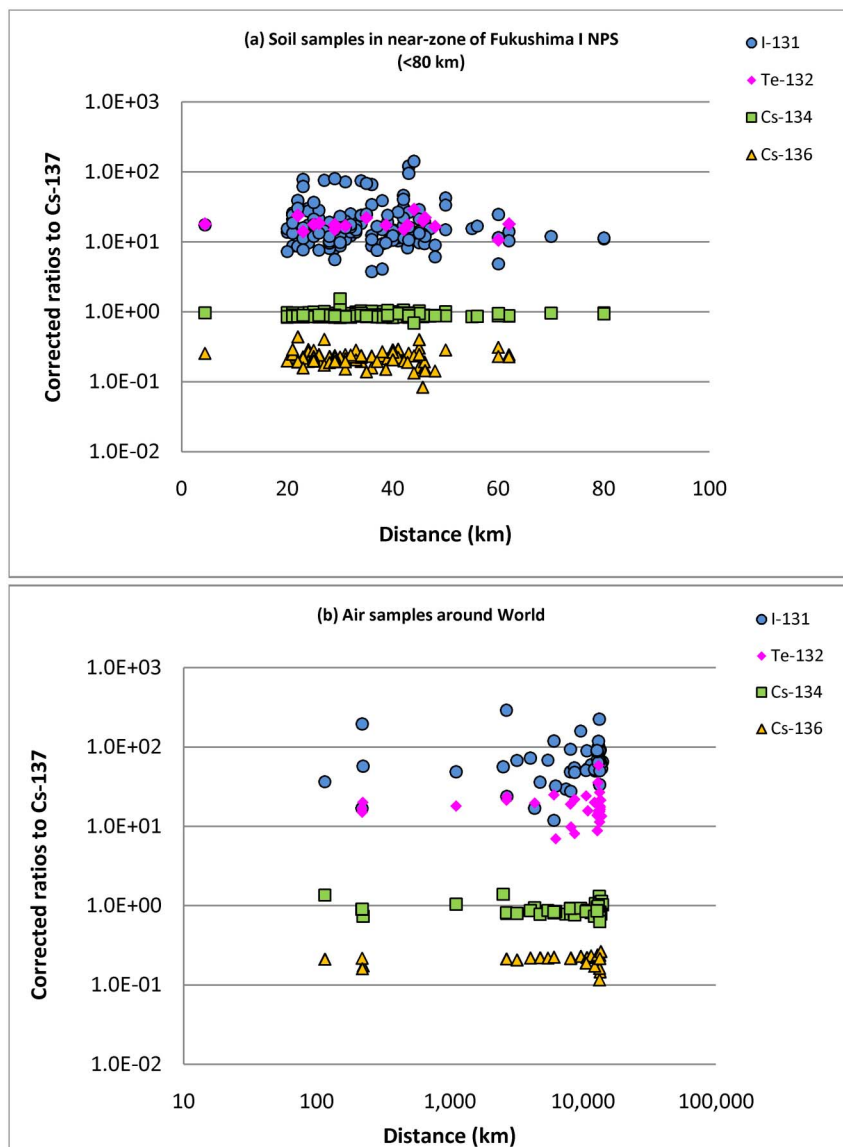
Figure 1 | Average corrected ratios to ¹³⁷Cs of ¹³¹I, ¹³⁴,¹³⁶Cs and ¹³²Te in (a) soils and (b) air particulates (data for gaseous-phase ¹³¹I are shown in Figure 3). Many of the I-131 measurements in air taken in the vicinity of Fukushima NPS were excluded from (b) as sampling did not begin until some days after the radionuclide release.

Radioisotope	Soil samples in near-zone (<80 km)	Air samples in near-zone (<80 km) Particulate	Air samples in Japan (80–2,000 km) Particulate	Air samples in Pacific Ocean and US (2,000–12,000 km) Particulate	Air samples in EU (>12,000 km) Particulate
Table 1 | Average corrected mean ratios to ¹³⁷Cs from release of Fukushima (±2 S.E., based on variation of means between sites)					
¹³¹I	22.5 ± 3.7 Sites = 145 Samples = 1,844	31.8 ± 12.9 * Sites = 22 Samples = 90	70.9 ± 63.5 Sites = 5 Samples = 234	69.5 ± 26.9 Sites = 20 Samples = 457	77.1 ± 16.6 Sites = 22 Samples = 196
¹³²Te	18.3 ± 1.7 Sites = 20 Samples = 20	No data	17.5 ± 2.1 Sites = 5 Samples = 226	17.1 ± 4.2 Sites = 21 Samples = 455	22.0 ± 6.0 Sites = 22 Samples = 196
¹³⁴Cs	0.90 ± 0.01 Sites = 146 Samples = 1,866	1.03 ± 0.10 Sites = 17 Samples = 68	0.99 ± 0.21 Sites = 5 Samples = 217	0.87 ± 0.06 Sites = 20 Samples = 420	0.91 ± 0.07 Sites = 22 Samples = 193
¹³⁶Cs	0.22 ± 0.01 Sites = 88 Samples = 297	No data	0.19 ± 0.03 Sites = 4 Samples = 105	0.22 ± 0.04 Sites = 11 Samples = 184	0.20 ± 0.03 Sites = 9 Samples = 27

*Note that these measurements began 4–5 days after the passage of the initial plumes so, because the ¹³¹I/¹³⁷Cs 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[8]) to these data was of the form

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

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_{131I} vs C_{137Cs}) 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 (>ca. 300 Bq kg^{-1} ^{137}Cs) to the north, northwest and west of Fukushima I NPS. For lower C_{Cs137} 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 I^{131}:^{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} ^{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[7]; 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[9] and the High Energy Linear Accelerator Research Centre, Tsukuba[10] (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 radiocaesium activity 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 (σ) represents the standard deviation in the residuals of the fit of Eq. 1 to observed data (std. deviation of $r_{measured}/r_{modelled}$ 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) $\times$ 10^{-5}) 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
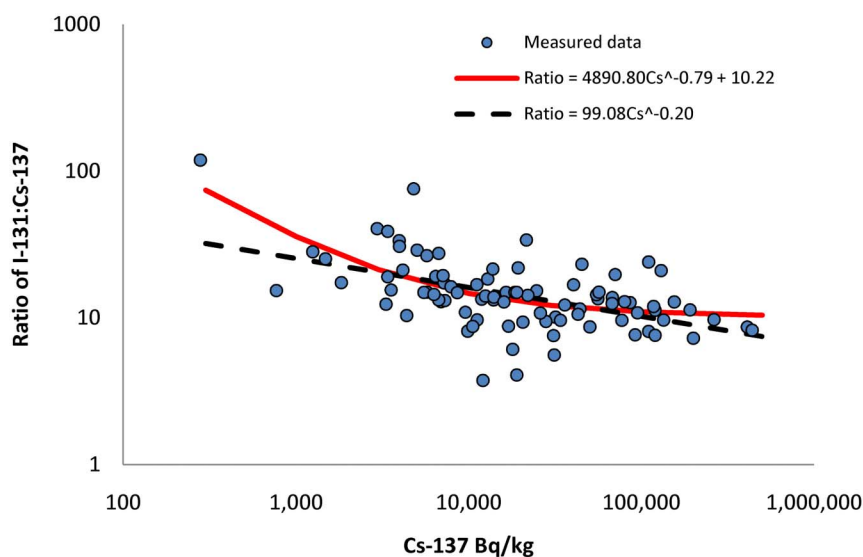

Figure 2 | Scatter plot of ratio of I-131 to Cs-137 against activity of Cs-137 in soil around Fukushima I NPS. In areas of very high radiocaesium fallout, the ratio ^{131}I/^{137}Cs is much lower than in areas of lower radiocaesium fallout.
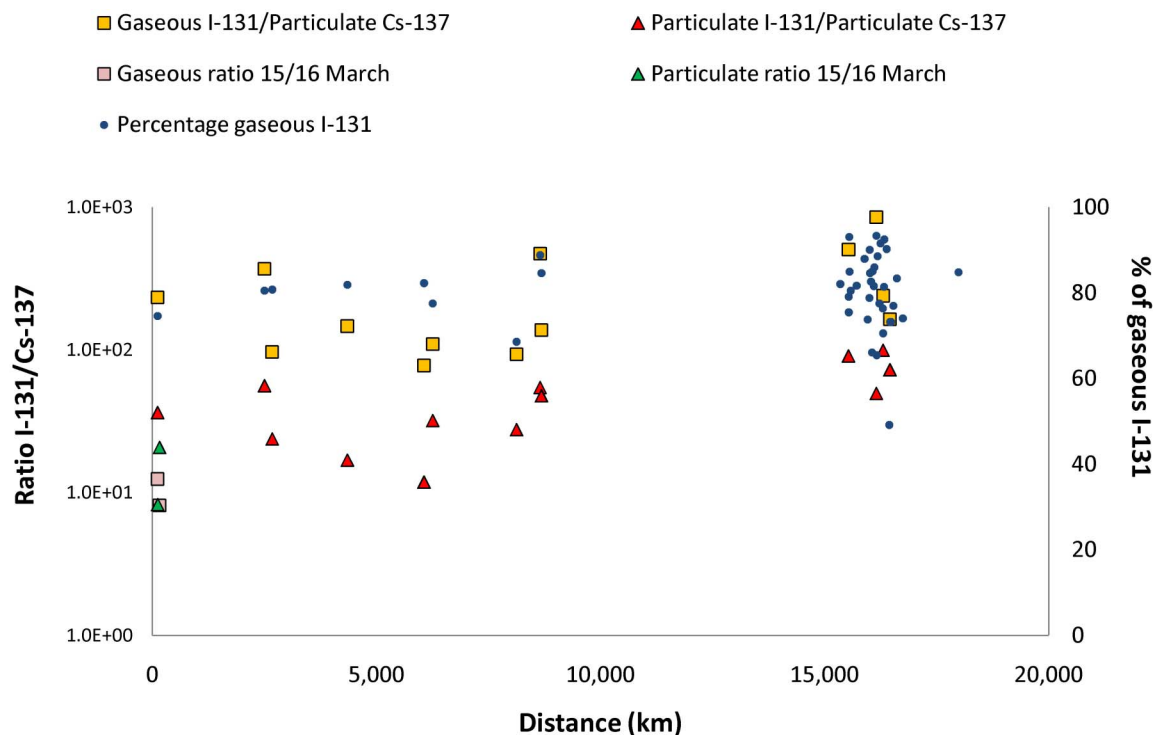
Figure 3 | Ratios of particulate and gaseous ^{131}I to particulate ^{137}Cs with distance from Fukushima I NPS. At two sites 120 and 160 km to the south of the accident site, lower ^{131}I:^{137}Cs ratios were observed in both phases on 15 March.

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 ^{131}I:^{137}Cs ratio) and varying early-phase depth distributions.

Discussion

As was observed after Chernobyl[11], ratios of the radiocaesium isotopes (^{134}Cs:^{137}Cs; ^{136}Cs:^{137}Cs) show low variability and are relatively constant with distance (as observed in Japan[7]). It should be noted, however, that there appears to be a slightly higher ^{134}Cs/^{137}Cs ratio in air within 80 km of Fukushima I NPS than >80 km (Table 1), potentially due to differences between release events: ^{134}Cs:^{137}Cs ratios may

vary according to the source of contamination depending on fuel age in a given reactor[12]. A shift of this isotope ratio had been noted in Europe and attributed to the small fraction of ^{137}Cs released during the Chernobyl accident or nuclear weapons test fallout still present in the atmosphere[12]. However, radiocaesium concentrations in Japan were too high to be significantly influenced by atmospheric ^{137}Cs from these sources. The mean ^{132}Te:^{137}Cs 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) ^{131}I:^{137}Cs 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

Table 2	Summary of the parameter values used in the predictive model*			
Radioelements	Decay constant (d^{-1})	Emitted gamma energy (MeV/nt)	Ratio to ^{137}Cs	A
^{131}I	8.64E-02	0.3828	$R_{131I} = 4.89 \times 10^3 \times C_{137Cs}^{-0.76} + 10.22$	$A = 9.17 \times 10^{-5}$
^{132}Te	2.16E-01	0.2344	18.30	
^{132}I	2.16E-01+	2.2645		
^{134}Cs	9.20E-04	1.5551	0.90	
^{136}Cs	5.27E-02	2.1283	0.22	
^{137}Cs	6.29E-05	0.5963	1.00	

*R_{131I} is the ratio of ^{131}I:^{137}Cs and C_{137Cs} is the mean activity concentration of ^{137}Cs (Bq/kg) in soil. The relationship for R_{131I} is only valid in areas of relatively high fallout (C_{137Cs} > 300 Bq kg^{-1}). For areas of lower fallout, a value of $R_{131I} = 76$ should be used.
+^{132}I is in secular equilibrium with ^{132}Te hence follows the ^{132}Te decay curve.
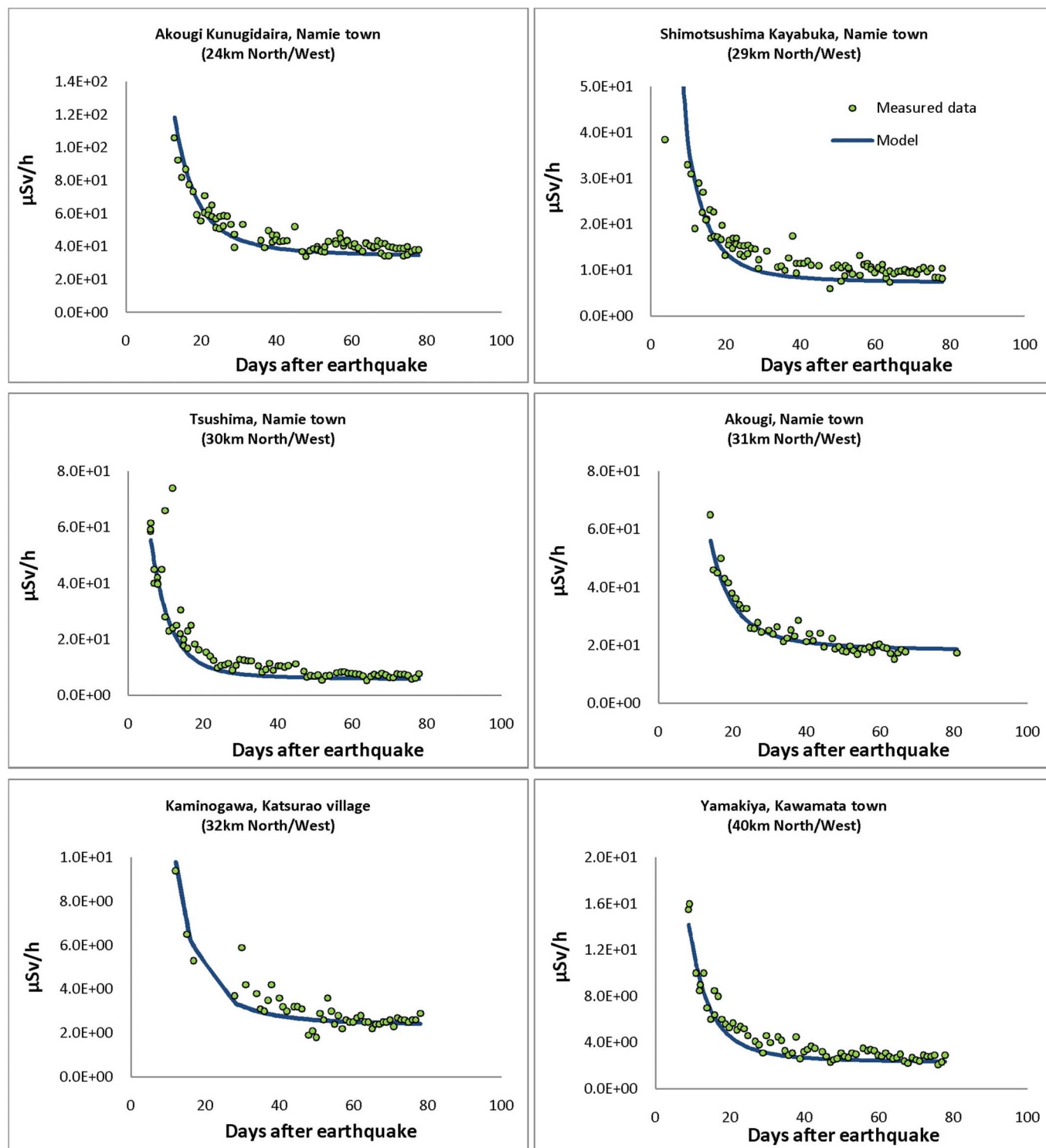
Figure 4 | Model fits to measured gamma dose rates obtained using generic isotope ratios for radiocaesium isotopes and ^{132}Te, and using a regression Equation (Eq. 1) for ^{131}I; the A parameter is fitted to the data using SAS (SAS Institute Inc.).

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[13]. 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
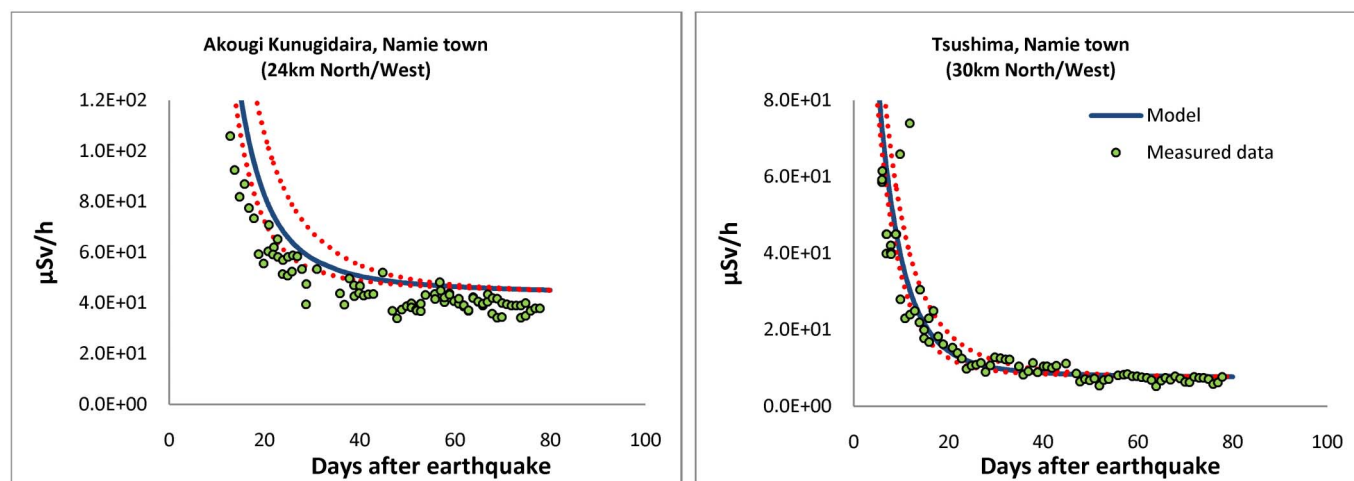
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Figure 5 | Model results using a generic mean _A_ value and isotope ratios (bold lines) with ±2σ uncertainty in estimation of I-131/Cs-137 ratio (dotted lines) compared with the measurement data at two of the six calibration sites. The values of _A_ 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.

was observed in the Fukushima prefecture during the night from 17:00 on March 15 to 04:00 on March 16[7]: 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 ^{137}Cs. This appears to be in contrast to observations following the Chernobyl accident[4] which showed generally _decreasing_ ^{131}I:^{137}Cs ratio with distance due to greater dry deposition of gaseous and particulate ^{131}I. It should be noted, however, that this observation

was based on relatively few data and, in the Chernobyl near-zone, radiocaesium 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.
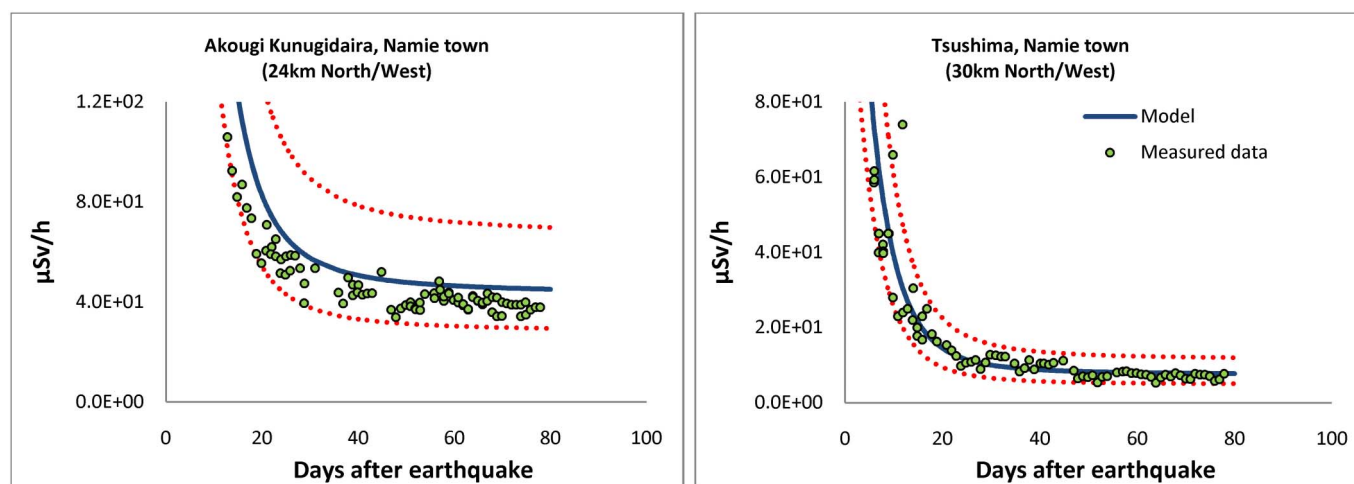

Figure 6 | Model results using generic _A_ value and isotope ratios (Table 2) (bold lines) with ±2σ uncertainty in estimates of the A-parameter (dotted lines) compared with the measurement data at two calibration sites. The graphs for the other four sites are shown in Supplementary Information, Figure S3. The values of _A_ and isotope ratios were the generic values from Table 2: not fitted to the data.
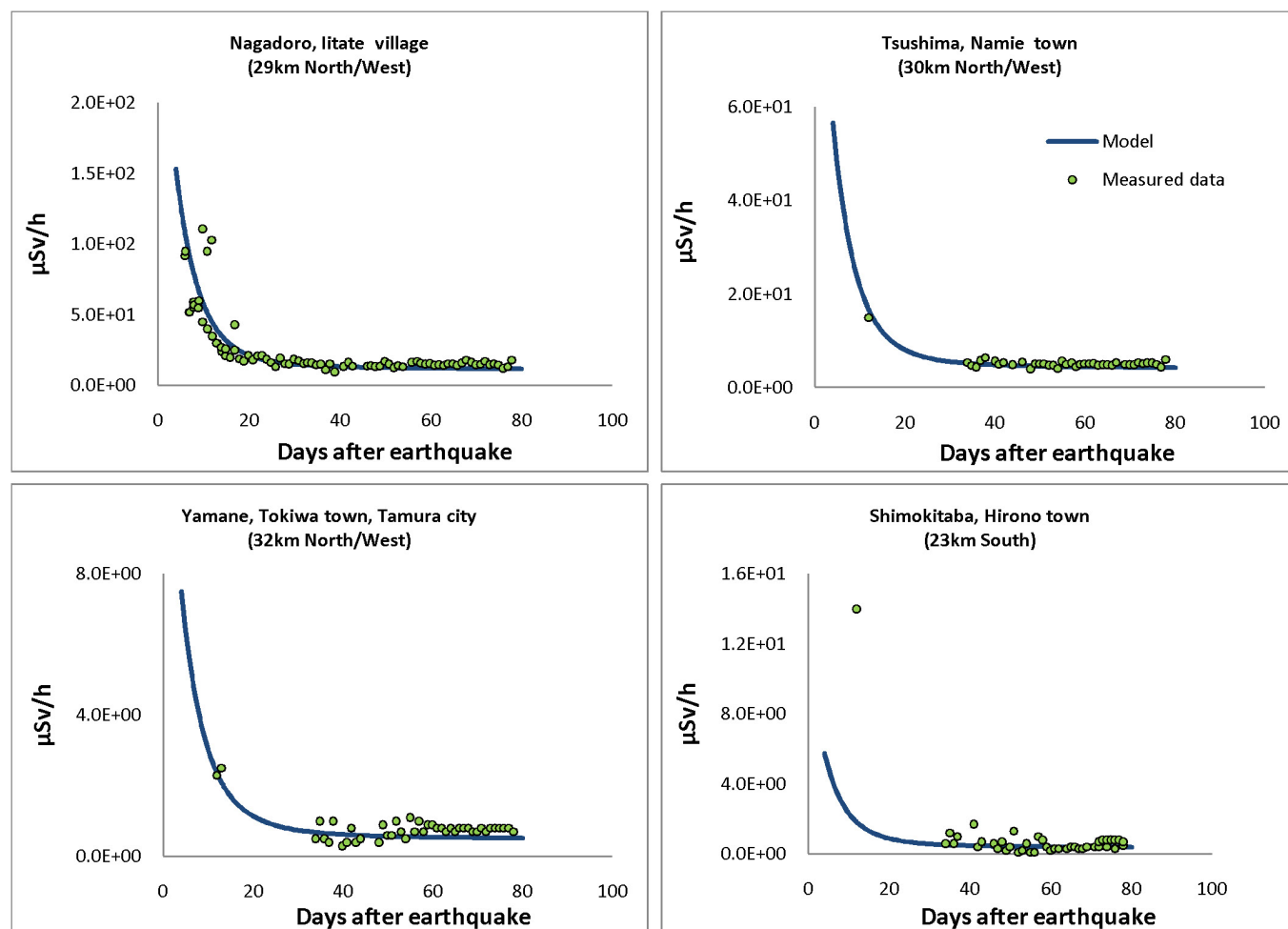
Figure 7 | Blind prediction of external gamma dose vs days since the earthquake using only measured Cs-137 concentration at each of four sites as an input variable. The graphs illustrate the good fit in regions to the North and West, but, as expected, relatively poorer fit at the one site to the south. For blind predictions at all 15 test sites, see Supplementary Information Figure S4.

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 ($(6.0–14.2) \times 10^{-5}$) with mean 9.2×10^{-5}. 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, radiocaesium can penetrate relatively deeply into the soil profile[16], however in most areas affected by Chernobyl[14], and at Fukushima[17], 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 radiocaesium into the soil profile[14,16].

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[5,9,10,18–36]. In addition, we use measurements at fourteen sites in Europe summarised in Masson et al.[15], Bossew et al.[37] 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 radiocaesium (^{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[38] 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 radiocaesium 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). ^{129m}Te and ^{129}Te were also found around Fukushima I NPS[7], and are included in
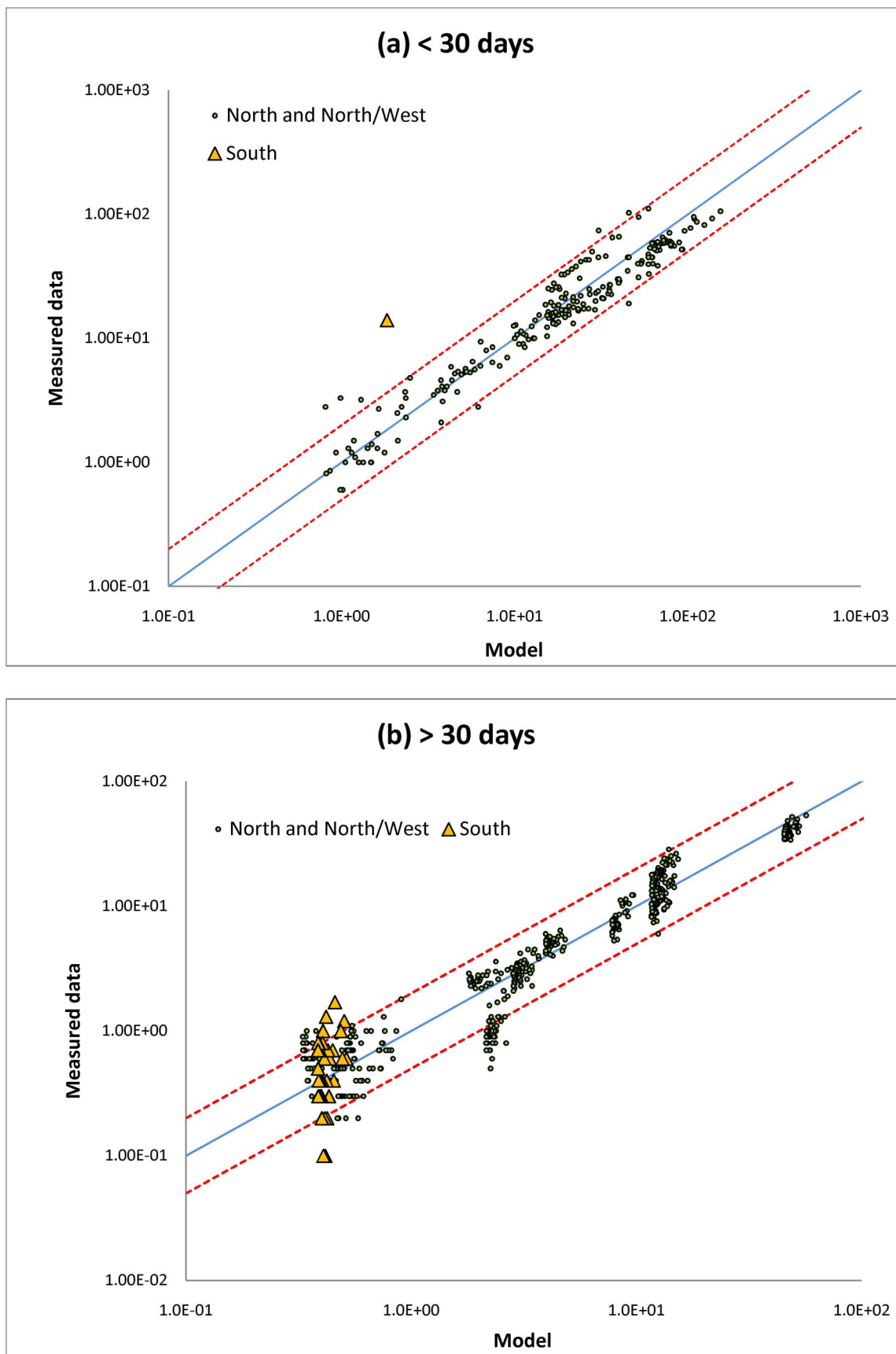
Figure 8 | Comparison of predicted dose rates (uSv/h) and measured data from (a) first measurement up to 30 days and (b) more than 30 days after the accident when the gamma dose rate were dominated by short-lived (^{132}Te/^{132}I and ^{131}I) and long-lived (^{134,137}Cs) radionuclides respectively.
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.

Table S2 for completeness, but these nuclides have only a minor effect on gamma-ray dose rate because of their low emitted gamma energy at 0.0376 and 0.0625 MeV for ^{129m}Te and ^{129}Te respectively. In addition, Tagami[39] show very low activity concentrations of other radioelements including ^{95}Nb, ^{110m}Ag and ^{140}Ba + ^{140}La, all of which can be ignored for evaluating gamma dose in this model.

Model. In the initial period, before significant penetration of radionuclides into the soil[16,17], 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 (μSv h^{-1}), at 1 m above the soil surface by:

$$D(t) = A.C_{137Cs}.\sum_i E_i.R_i e^{-\lambda_i t} \qquad (2)$$

Where E_i is total gamma decay energy of each nuclide (MeV) (see Supplementary Table S2), C_{137Cs} (Bq kg^{-1}) is the mean activity concentration of ^{137}Cs in the surface 5 cm of soil, R_i is ratio of deposition of each nuclide to ^{137}Cs, λ_i is the decay constant of each nuclide (d^{-1}), and t (d) is time after the earthquake. A [μSv hr^{-1} MeV^{-1} kg 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 measurements 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.

1. Hirose, K. Fukushima Dai-ichi nuclear power plant accident: summary of regional radioactive deposition monitoring results. *Journal of Environmental Radioactivity* **111**, 13–17.
2. Hosoda, M. et al. The time variation of dose rate artificially increased by the Fukushima nuclear crisis. *Sci. Rep.* **1**, 87, dx.doi.org/10.1038/srep00087 (2011).
3. Tokonami, S. et al. Thyroid doses for evacuees from the Fukushima nuclear accident. *Sci. Rep.* **2**, 507 dx.doi.org/10.1038/srep00507 (2012).
4. Mück, K. et al. A Consistent Radionuclide Vector After the Chernobyl Accident. *Health Physics* **82**, 141–156 (2002).
5. Endo, S. et al. Measurement of soil contamination by radionuclides due to the Fukushima Dai-ichi Nuclear Power Plant accident and associated estimated cumulative external dose estimation. *Journal of Environmental Radioactivity* **111**, 18–27 (2012).
6. Beck, H. L. Exposure Rate Conversion Factors for Radionuclides Deposited on the Ground. (U.S. Department of Commerce, Springfield, 1980).
7. Kinoshita, N. et al. Assessment of individual radionuclide distributions from the Fukushima nuclear accident covering central-east Japan. *Proceedings of the National Academy of Sciences* **108**, 19526–19529 (2011).
8. Bergman, R. N., Ider, Y. Z., Bowden, C. R. & Cobelli, C. Quantitative estimation of insulin sensitivity. *American Journal of Physiology - Endocrinology And Metabolism* **236**, E667–677 (1979).
9. JAEA. Emergency Monitoring of Environmental Radiation and Atmospheric Radionuclides at Nuclear Science Research Institute, JAEA Following the Accident of Fukushima Daiichi Nuclear Power Plant. A. <http://jolissrch-inter.tokai-sc.jaea.go.jp/pdfdata/JAEA-Data-Code-2012-010.pdf> (accessed 07 March 13).
10. KEK. *Measurement result of airborne nuclide and air radiation level in Tsukuba area* <http://legacy.kek.jp/quake/radmonitor/GeMonitor-e.html> (accessed 04 May 2011).
11. Mück, K. et al. A Consistent Radionuclide Vector After the Chernobyl Accident. *Health Physics* **82** (2002).
12. Kirchner, G., Bossew, P. & De Cort, M. Radioactivity from Fukushima Dai-ichi in air over Europe; part 2: what can it tell us about the accident? *Journal of Environmental Radioactivity* **114**, 35–40 (2012).
13. Chino, M. et al. Preliminary Estimation of Release Amounts of ^{131}I and ^{137}Cs Accidentally Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere. *Journal of Nuclear Science and Technology* **48**, 1129–1134 (2011).
14. Smith, J. T. & Beresford, N. A. *Chernobyl – Catastrophe and Consequences.* (Springer-Praxis, 2005).
15. Masson, O. et al. Tracking of Airborne Radionuclides from the Damaged Fukushima Dai-Ichi Nuclear Reactors by European Networks. *Environmental Science & Technology* **45**, 7670–7677 (2011).
16. Bunzl, K. et al. Cs-137 mobility in soils and its long-term effect on the external radiation exposure. *Radiation and Environmental Biophysics* **36**, 31–37 (1997).
17. Kato, H., Onda, Y. & Teramage, M. Depth distribution of ^{137}Cs, ^{134}Cs, and ^{131}I in soil profile after Fukushima Dai-ichi Nuclear Power Plant Accident. *Journal of Environmental Radioactivity* **111**, 59–64 (2012).
18. Biegalski, S. R. et al. Analysis of data from sensitive US monitoring stations for the Fukushima Daiichi nuclear reactor accident. *Journal of Environmental Radioactivity* (2011).
19. Carvalho, F. P., Reis, M. C., Oliveira, J. M., Malta, M. & Silva, L. Radioactivity from Fukushima nuclear accident detected in Lisbon, Portugal. *Journal of Environmental Radioactivity* **114**, 152–156 (2012).
20. EPA. Japanese Nuclear Emergency: Radiation Monitoring. (2011). <http://epa.gov/japan2011/rert/radnet-sampling-data.html#air> (accessed 19 March 2013).
21. Gudelis, A. et al. Radionuclides in the ground-level atmosphere in Vilnius, Lithuania, in March 2011, detected by gamma-ray spectrometry. *Journal of Environmental Radioactivity* **109**, 13–18 (2012).
22. HPA. *Update on the Fukushima Incident: 14 July 2011*, <http://www.hpa.org.uk/web/HPAweb&HPAwebStandard/HPAweb_C/1309968917992> (accessed 21 August 2012).
23. Imanaka, T. et al. Early Radiation Survey of Iitate Village, Which Was Heavily Contaminated by the Fukushima Daiichi Accident, Conducted on 28 and 29 March 2011. *Health Physics* **102**, 680–686 (2012).
24. Kanai, Y. Monitoring of aerosols in Tsukuba after Fukushima Nuclear Power Plant incident in 2011. *Journal of Environmental Radioactivity* **111**, 33–37 (2012).
25. Leon, J. D. et al. Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA. *Journal of Environmental Radioactivity* **102**, 1032–1038 (2011).
26. Loaiza, P. et al. Air radioactivity levels following the Fukushima reactor accident measured at the Laboratoire Souterrain de Modane, France. *Journal of Environmental Radioactivity* **114**, 66–70 (2012).
27. MacMullin, S. et al. Measurement of airborne fission products in Chapel Hill, NC, USA from the Fukushima Dai-ichi reactor accident. *Journal of Environmental Radioactivity* **112**, 165–170 (2012).
28. Perrot, F. et al. Evidence of ^{131}I and ^{134,137}Cs activities in Bordeaux, France due to the Fukushima nuclear accident. *Journal of Environmental Radioactivity* **114**, 54–60 (2012).
29. Pham, M. K. et al. Detection of Fukushima Daiichi nuclear power plant accident radioactive traces in Monaco. *Journal of Environmental Radioactivity* **114**, 131–137 (2012).
30. Piñero García, F. & Ferro García, M. A. Traces of fission products in southeast Spain after the Fukushima nuclear accident. *Journal of Environmental Radioactivity* **114**, 146–151 (2012).
31. RIKEN. *Report on the γ-ray measurement after the accident of Fukushima I Nuclear Power Station,* <http://www.radiochem.org/kinkyu/49.pdf> (accessed 05 May 2011).
32. Stoehlker, U., Nikkinen, M. & Gheddou, A. Detection of radionuclides emitted during the Fukushima nuclear accident with the CTBT radionuclide network. *Monitoring Research Review: Ground-Based Nuclear Explosion Monitoring Technologies,* 715-724 (2011).
33. UCB. *Air Sampling Results* <http://www.nuc.berkeley.edu/UCBAirSampling/AirSamplingResults> (21 August 2012).
34. MEXT. *Readings of dust sampling (All Results for May 2011)* <http://www.mext.go.jp/component/english/__icsFiles/afieldfile/2011/06/15/1306621_053110.pdf> (accessed 21 August 2012).
35. MEXT. *Readings of soil monitoring (All Results for May 2011),* <http://www.mext.go.jp/component/english/__icsFiles/afieldfile/2011/06/15/1306622_053110.pdf> (accessed 10 July 2011).
36. MEXT. *Readings of environmental monitoring sample (All Results for May 2011),* <http://www.mext.go.jp/component/english/__icsFiles/afieldfile/2011/06/15/1306623_053110.pdf> (accessed 10 July 2011).
37. Bossew, P. et al. Radioactivity from Fukushima Dai-ichi in air over Europe; part 1: spatio-temporal analysis. *Journal of Environmental Radioactivity* **114**, 22–40 (2012).
38. ICRP. Nuclear Decay Data for Dosimetric Calculations. ICRP Publication 107. (International Commission on Radiological Protection, 2008).
39. Tagami, K. et al. Specific activity and activity ratios of radionuclides in soil collected about 20 km from the Fukushima Daiichi Nuclear Power Plant: Radionuclide release to the south and southwest. *Science of the Total Environment* **409**, 4885–4888 (2011).

Acknowledgements

Author contributions

K.C. and J.T.S. wrote the main manuscript text, with contributions from P.B. and G.K. K.C. collated data G.L. supplied data, and K.C., J.T.S., G.K. and P.B. analysed data. All authors reviewed the manuscript.

Additional information