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The influence of hot particle contamination on ^{90}Sr and ^{137}Cs transfers to milk and on time-integrated ingestion doses.

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Abstract

Most models for transfers of radionuclides through the food chain typically assume that the radioactivity is initially deposited in chemically available forms. It is known, however, that releases of radionuclides in the form of hot particles may significantly influence their environmental transfers and uptake to the food chain. This study presents models for time changes in ^{90}Sr and ^{137}Cs in milk which incorporate hot particle contamination using observed rates of hot particle dissolution following the Chernobyl accident. A general equation is presented for the influence of hot particles on overall ingestion doses. As expected from previous work, fallout of hot particles significantly influences time changes in radionuclide activity concentrations in foodstuffs. It is also shown that incorporation of radionuclides in hot particles influences time-integrated ingestion doses. For a situation in which a large proportion (90-100%) of fallout is in slowly-dissolving hot particles, time-integrated ingestion doses from ^{90}Sr and ^{137}Cs are reduced by a factor of approximately two compared to the case where all radioactivity is deposited in bioavailable forms. However, the influence of rapidly-dissolving hot particles on time-integrated ingestion doses is relatively minor. Remaining significant uncertainties in dose estimates are discussed.

Keywords: hot particle, fuel particle, dissolution, radiocaesium, radiostrontium, Chernobyl, milk, grass, cow, dose.

1. Introduction

Most models for transfers of radionuclides through the food chain assume that the radioactivity is initially deposited in chemically available forms. However, it is known that the accidental release of radionuclides in the form of hot particles may significantly influence their environmental transfers and uptake to the food chain (e.g. Konoplev and Bobovnikova, 1991; Salbu et al., 1994; Krouglov et al., 1997). Release of a proportion of fallout in chemically less available hot particles was observed in releases from the Windscale reactor in the UK during 1952-57 (Salbu et al., 1994) as well as from the Chernobyl accident (e.g. Bobovnikova et al., 1991; Kashparov et al., 1999).

Salbu et al. (1994) describe the influence of hot particles on food chain transfers as follows:

> "The transfer of radionuclides through food chains might initially be overestimated if inert fuel particles or radionuclides irreversibly associated with condensed particles … are present. Conversely, the long-term transfer would be underestimated if mobilization of radionuclides from fuel particles due to weathering is not taken into account."

In this study, historical data on time changes in ^{90}Sr and ^{137}Cs in milk will be modelled for cases where fuel particle contamination is not significant. Using observed rates of hot particle dissolution following the Chernobyl accident (Kashparov et al., 1999) the model will be extended to quantify the influence of hot

particle contamination on radionuclide uptake via the food chain.

The initial explosion and subsequent fire at Chernobyl deposited fuel particles principally within an area of radius of 30 km around the reactor. Within this area, the majority of fallout was in the form of these hot particles (Bobovnikova et al., 1991). Kashparov and coworkers (1999) distinguish two forms of particles released at different stages of the accident: "non-oxidised" particles of uranium dioxide fuel released during the initial explosion and "oxidised" particles released during the subsequent reactor fire. This was confirmed by subsequent measurement of the oxidation state of particles emitted in different directions (North and West) following the accident (Salbu et al., 2001). More than 90% of the release of ^{90}Sr, ^{141,144}Ce, Pu isotopes and ^{241}Am was in the form of fuel particles of average diameter around 10 μm (Kashparov et al., 1999; Mück et al., 2002), and within 30 km of the plant most (approximately 50-75%) of the ^{137}Cs was in fuel particles (Krouglov et al., 1998; Kashparov et al., 1999), though the (relatively volatile) caesium isotopes were also dispersed much further afield.

Even at large distances from Chernobyl, radionuclides could be deposited in particluate form. Fuel particles of < ~1 μm diameter were found as far away as Norway and Sweden (Devell et al., 1986; Salbu et al., 1994). Approximately 75% of the ^{137}Cs in Norwegian rainwater was associated with high molecular weight particles (> 10,000 Da), though this may in part have been a result of attachment to and subsequent washout of atmospheric dust particles. Nine fuel particles found in North Eastern Poland contained from 4 990-139 000 Bq of ^{103}Ru and 1 050-28 000 Bq of ^{106}Ru in each particle (decay corrected to April 26, 1986) (Schubert and Behrend,

1987). Concentrations of ^{137}Cs in these particles were much lower. In the UK, at a large distance from Chernobyl, the majority of radionuclides were deposited in relatively chemically available forms (Hilton et al., 1992; Smith and Beresford, 2005).

The rates of fuel particle decay estimated by Kashparov et al. (1999) imply that at present the majority of Chernobyl-derived fuel particles in terrestrial systems have dissolved. By 1997, less than 20% of ^{90}Sr remained in fuel particle form (Kashparov et al., 2004).

Even at large distances from Chernobyl, the chemical form of radioactivity influenced uptake of radionuclides to foodstuffs. After Chernobyl a number of studies (Beresford et al., 1989; Ward et al., 1989; Hansen and Hove, 1991; Voigt et al., 1996) reported that transfer coefficients for recently deposited radiocaesium were lower than for plant incorporated radiocaesium via root uptake. For example, Hansen and Hove (1991) found increasing transfer coefficients to goats' milk from 0.042 d l^{-1} in 1986 to 0.124 d l^{-1} in 1988, the latter value being similar to that observed for a (bioavailable) ^{134}Cs tracer.

2. Modelling

Rates of degradation of fuel particles can be estimated (Konoplev et al. 1992; Krouglov et al.. 1997; Kashparov et al., 1999) using a simple decay equation:

$$A(t) = A_0 \exp(-k_p t) \quad (1)$$

where $A(t)$ and A_0 are the radionuclide activities in particles at time t and at the time of deposition respectively. The half-life of fuel particle degradation, $T_{1/2}^{p}$, is given by ($T_{1/2}^{p} = \ln 2/k_p$).

Let the fraction of the surface contamination density D [Bq m^{-2}] which is in fuel particles be f_p. The surface contamination density of radioactivity which is not in fuel particle form, D_a [Bq m^{-2}] is therefore given by:

$$D_a(t) = D(1 - f_p \exp(-(k_p + \lambda)t)) \quad (2)$$

where λ is the physical decay constant of the radionuclide.

Radiocaesium and radiostrontium activity concentrations in vegetation change significantly over time due to changes in their inventories and bioavailability in the soil. Activity concentrations of radiocaesium decline over the years after fallout as a result of "fixation" to clay minerals (e.g. Cremers et al., 1988; Smith et al., 1999; 2000) as well as (initially, to a lesser extent) erosion from and redistribution within the soil column. Fixation is believed to be much less important for radiostrontium (Coughtrey & Thorne, 1983) but activity concentrations of radiostrontium in surface waters, vegetation and foodstuffs still slowly decline over the years after fallout (e.g. UNSCEAR, 1977, Mück et al. 2001; Cross et al. 2002). In the absence of fuel particle contamination, the change in ^{137}Cs and ^{90}Sr annual average activity concentrations in vegetation, C_v(t) [Bq kg^{-1} d.w.] following a deposition, D [Bq m^{-2}], may be modelled using a series of exponential functions (e.g. Mück, 1997; Travnikova et al., 1999; Smith et al. 1999; 2000; 2002; Mück et al., 2001):

$$C_v(t) = D\left[\alpha \exp(-(k_1 + \lambda)t) + \beta \exp(-(k_2 + \lambda)t) + \gamma \exp(-(k_3 + \lambda)t)\right] \quad (3)$$

where α, β, γ [m^2 kg^{-1}] and k_1, k_2, k_3 [y^{-1}] are empirically determined constants. The exponential terms represent, respectively, the fast initial decline in activity concentrations due to washoff processes, slower declines over the first years after fallout and very slow declines in the long term. In the present study, annual average activity concentrations are being modelled, so the first rapid washoff term will be neglected.

In the presence of fuel particle contamination, the fraction of radioactivity in the soil not in fuel particle form will be taken up by vegetation according to Equation 3. It is assumed that, over time, newly available radioactivity leached from fuel particles will also be taken up by vegetation in subsequent years according to Equation 3. Thus, in year 5 after fallout, for example, radioactivity newly leached from fuel particles will be significantly more strongly accumulated in vegetation than radioactivity which was not initially deposited in fuel particle form and has had 5 years to become less bioavailable to vegetation.

In the presence of fuel particle contamination, the contamination density per unit area of newly bioavailable radioactivity in year j, $\delta_a(j)$ [Bq m^{-2}] is given by:

$$\begin{aligned} \delta_a(j=0) &= (1-f_p)D \\ \delta_a(j>0) &= f_p D(e^{-(k_p+\lambda)(j-1)} - e^{-(k_p+\lambda)j}) \end{aligned} \quad (4)$$

The activity concentration of the radionuclide in vegetation in year *i* is, from Equations 3 and 4:

$$C_v(i) = \sum_{j=0}^{j=i} \delta_a(j)[\beta \exp(-(k_2 + \lambda) j) + \gamma \exp(-(k_3 + \lambda) j] \quad (5)$$

The transfer of radionuclides from an animal's diet to milk or meat is most often expressed as the equilibrium transfer coefficient (F_f or F_m for meat or milk respectively, units: d kg^{-1}), defined as the ratio of the activity concentration in a tissue to the rate of radionuclide ingestion:

$$F_m = \frac{Activity\ conc.\ in\ milk,\ Bq\,kg^{-1}}{Radionuclide\ ingestion\ rate,\ Bq\,d^{-1}} = \frac{C_m}{C_v I_f} \quad (6)$$

or

$$C_m = F_m C_v I_f \quad (7)$$

where C_m is the activity concentration in the milk (Bq kg^{-1}) (C_f for meat; Bq kg^{-1} fresh weight), C_v is the activity concentration in vegetation (Bq kg^{-1} dry matter) and I_f (kg d^{-1} dry matter) is the feed intake rate.

Equation 6 can be written in terms of the equilibrium Concentration Ratio (*CR*, kg kg^{-1}), the ratio of activity concentration in milk to that in feed:

$$CR = \frac{Activity\ conc.\ in\ milk,\ Bq\,kg^{-1}}{Activity\ conc.\ in\ \ feed,\ Bq\,kg^{-1}} = \frac{C_m}{C_v} = F_m I_f \quad (8)$$

By definition,

$$C_m = C_v \times CR \quad (9)$$

2.1 Empirical data and model calibration

Assuming that approximately 100% of ^{90}Sr in the Chernobyl 30-km Zone was in fuel particles at the time of fallout, Kashparov and coworkers (1999) found that rates of dissolution were related to the type of fuel particle deposited and the pH of the soil. Particles deposited to the west of the reactor during the initial release phase took much longer to dissolve ($T_{1/2}^{p}$ = 4.95 – 17.33 years) than those deposited during the subsequent fire in a North-South plume ($T_{1/2}^{p}$ = 1.65 – 4.95 years) (Kashparov et al., 1999). This difference was attributed by these workers to the lower degree of UO_2 oxidation of the particles released to the west of the reactor during the initial explosion. In this study the influence of fuel particle dissolution on rates of radionuclide uptake to vegetation and milk will be assessed using the range of dissolution rates observed by Kashparov et al. (1999). Thus, model runs were carried out for low dissolution rates ($T_{1/2}^{p}$ = 17.33 years, k_p = 0.04 y^{-1}: low degree of oxidation, soil pH: 5.6) and high dissolution rate ($T_{1/2}^{p}$ = 1.65 years, k_p = 0.42 y^{-1}: high degree of oxidation, soil pH: 4.5).

The model for uptake of ^{90}Sr and ^{137}Cs in milk in the absence of fuel particles

(Equation 3) was calibrated using time series measurements of these radionuclides in milk from areas with low fallout of radioactivity in fuel particle form. For ^{137}Cs, time series measurements in vegetation and milk from the Bryansk region of Russia after the Chernobyl accident were used (Travnikova et al. 1999). For ^{90}Sr, post-Chernobyl data is relatively sparse for areas without significant fuel particle contamination. Therefore, time series measurements of ^{90}Sr in milk at a number of different sites following fallout from atmospheric nuclear weapons testing (NWT) was used (UNSCEAR, 1977; 1982). In the UNSCEAR (1977) report, these data were presented as pCi of ^{90}Sr per gram of calcium in milk. The data were therefore converted to Bq l^{-1} using an average Ca content in milk of 1.2 g l^{-1} (UNSCEAR, 1982). Time series measurements in milk were obtained for sites in Denmark, Finland, UK, USA (New York) and Russia (Moscow) for the period 1955-1980. Deposition of ^{90}Sr to each of these sites was estimated according to latitude from data in UNSCEAR (1977) and the relative amount deposited in each year was estimated using data on annual deposition to the Northern Hemisphere presented in Cambray et al. (1989), see Figure 1.

For ^{137}Cs, parameter values for Equations 3 and 9 were determined using a chi-squared fit to the measured data of activity concentrations in both vegetation and milk. For ^{90}Sr, activity concentrations in vegetation were not available, so only data for milk were fitted. Since, on a timescale of years, the activity concentration in milk is a constant multiple of that in vegetation (Equation 9), the time-dependent parameters in Equation 3 can be fitted against milk data alone.

3. Results

The results of the model fits against the measurements of ^{137}Cs in vegetation and milk in the Bryansk region of Russia following the Chernobyl accident are shown in Figure 2. As expected, the time changes in activity concentrations in milk closely follow those in vegetation, showing that the two components are in dynamic equilibrium. Fitted parameter values for ^{137}Cs are given in Table 1.

The results of the model fits against the measurements of ^{90}Sr in milk from several countries following fallout from nuclear weapons testing are shown in Figure 3. The model shows a good fit to the measurements and fitted parameter values for ^{90}Sr are given in Table 1.

Using parameter values presented in Table 1, predictions were made of the activity concentration in vegetation assuming different fractions, f_p, of activity initially deposited in fuel particles. Figure 4 shows the results of these model predictions for values of f_p in the range 0 – 0.99 and for low ($T_{1/2}^{p}$ = 17.33 years, k_p = 0.04 y^{-1}: low degree of oxidation, soil pH: 5.6) and high hot particle dissolution rates ($T_{1/2}^{p}$ = 1.65 years, k_p = 0.42 y^{-1}: high degree of oxidation, soil pH: 4.5). For both low and high hot particle dissolution rates, there is a pronounced difference in the temporal change in activity concentrations in milk if a significant proportion of the activity is initially deposited in hot particles (compared to zero initial deposition in hot particles). The difference is particularly pronounced for the case of low hot particle dissolution rate. In the first years after fallout, predicted activity concentrations are significantly lower when a significant proportion of the contamination is in hot particle form. But, after a

period of 20 years following fallout, activity concentrations of ^{137}Cs in vegetation and milk are estimated to be 2.9-3.9 times higher for $f_p > 0.5$ compared to $f_p = 0$. Activity concentrations of ^{90}Sr after the 20 year period are estimated to be 1.5 – 2.0 times higher for $f_p > 0.5$ compared to the case of no fuel particle contamination ($f_p = 0$).

Doses from consumption of milk were calculated for a 20 year period after fallout assuming a milk consumption rate for adults of 370 kg y^{-1} as observed in a village in Ukraine (Beresford and Wright, 1999) in 1997. Time-integated doses to adults (for the 20 year period) were calculated using committed effective doses per unit intake given in NRPB (1998). Time-integrated doses are shown in Figure 5 for different fractions, f_p, of activity initially deposited in fuel particles. It is clear from Figure 5 that, over this time period, the presence of fuel particles has little influence on time integrated dose if the particles release their radioactivity rapidly. For the slow release case, however, the dose via consumption of milk is significantly lower in the situation where a significant fraction of radioactivity is unavailable in hot particle form.

4. Discussion

There are relatively few empirical data for sites at which a significant proportion of fallout is in the form of fuel particles. A study of ^{137}Cs and ^{90}Sr transfers to crops within the 30 km exclusion zone was carried out by Krouglov et al. (1997). At the sites studied, almost 100% of ^{90}Sr and 50-70% of ^{137}Cs was in the form of hot particles. The measurements in grains (Krouglov et al., 1997) generally followed the same pattern as that predicted for milk in Figure 4, with the maximum ^{90}Sr activity concentration being observed 2-3 years after the maximum ^{137}Cs activity

concentration. This was primarily because a much higher fraction of ^{90}Sr was initially in the form of hot particles.

4.1 Influence of hot particles on total ingestion doses

As shown in Figure 4, deposition of radioactivity in fuel particle form tends to reduce the activity concentration in milk and hence the predicted dose via the ingestion pathway (though note remaining uncertainties due to inadvertent ingestion of fuel particles, discussed below). The influence on dose is dependent on the fraction of radioactivity initially deposited in fuel particle form and the rate of hot particle degradation. If a significant proportion of radioactivity is deposited as hot particles (and hence is unavailable for root uptake) then the time-integrated dose is reduced by that fraction of radioactivity which decays whilst still in unavailable forms. Thus, that fraction of deposited radioactivity not contributing to dose via the ingestion pathway, ø [dimensionless], is given by:

$$\phi = f_p \lambda \int_0^\infty \exp(-(k_p + \lambda)t)\, dt = \frac{f_p \lambda}{k_p + \lambda} \qquad (10)$$

where ø is the dose integrated from time zero to infinity. For the rates of fuel particle degradation considered here (k_p = 0.04 – 0.42), the fraction not contributing to dose is given in Table 2. The values presented in Table 2 apply to ingestion of any foodstuff (grains, milk, meat etc.) where the contamination enters the foodstuff via root uptake. It therefore does not account for potential direct ingestion of hot particles adhering to foodstuffs, or inadvertent ingestion of hot particles in animals' feed.

The data in Table 2 implies that (for the range of fuel particle degradation rates considered here), the ingestion dose (via root uptake) is reduced by up to 36% for ^{137}Cs and by up to 38% for ^{90}Sr for the case in which a large proportion of hot particles are present compared to the case where radionuclides are all deposited in available form.

4.2 Uncertainties

The model presented here requires estimates of radionuclide deposition as an input parameter: the accuracy of such estimates depends on the quality of deposition mapping and estimation. In addition, for beta and alpha emitting radionuclides (e.g. for ^{90}Sr), where fuel particles are present, activity depositions of may be underestimated if the extraction procedure is less than 100% efficient. In addition, for ^{137}Cs, the presence of hot particle deposition in the Bryansk region (ca. 200 km from Chernobyl), though much less important than in the 30 km zone, potentially adds additional uncertainty to the model as the calibration assumed this component to be negligible. The quality of the ^{90}Sr calibration data is believed to be high: even though it is converted from data on ^{90}Sr in milk per gram of calcium, the data from the different countries is very consistent (Figure 3) both in activity concentration and in temporal pattern.

Variation in radionuclide soil-plant concentration ratio is a major source of uncertainty in predicting transfers to foodstuffs. For example, it has long been known that areas most vulnerable to radiocaesium contamination of milk and meat were

those with nutrient poor soils of low clay mineral content (e.g. Aarkrog, 1979; Desmet et al., 1990). Thus, model predictions for transfers of radionuclides to milk (Figure 4) will vary according to various environmental and animal management factors. The time-changes in activity concentrations in various foodstuffs are, however, less variable (e.g. Mück, 1997; Smith et al., 1999; Mück et al., 2001) so Figure 4 should be representative of the influence of hot particles on time changes in activity concentrations. Note also that predictions are made on an annual average basis – the models were calibrated using annual average data, so the initial "spike" in activity concentrations due to ingestion by animals of radioactivity adhering to plant surfaces is not modelled.

A further, important, source of uncertainty in models is the ingestion of hot particles either by farm animals or directly by humans. For example, Salbu et al. (1995) fed fuel particles released by Chernobyl to six goats. In one of the goats, a particle was retained in the second stomach compartment for 3.5 months. Hot particles ingested by animals could dissolve in the gastro-intestinal tract and thus "unavailable" radioactivity could be transferred to milk and meat. There is insufficient data at present to assess the uncertainty which this introduces into models. Studies of transfer coefficients suggest that (at least for radionuclides attached to small aerosol particles) this may not have a major influence on activity concentrations in animal-derived foodstuffs. As discussed above, a number of post-Chernobyl studies (at sites a long distance from the reactor) (Beresford et al., 1989; Ward et al., 1989; Hansen and Hove, 1991; Voigt et al., 1996) reported that transfer coefficients for recently deposited radiocaesium (adhered to plant surfaces with a proportion in the form of small particulates) were significantly lower than for radiocaesium incorporated in

plants via root uptake. This implies that, in these cases, the radioactivity ingested in the form of small particulates was less bioavailable than that incorporated in plants via root uptake.

It should also be noted that the models described above do not account for inadvertent ingestion by humans of hot particles attached to plant surfaces.

5. Conclusions

- As expected from previous work into the influence of hot particles on biological uptake (Salbu et al., 1994; Krouglov et al., 1997), fallout of hot particles significantly influences time changes in radionuclide activity concentrations in foodstuffs. These time changes have here been quantified for the grass-cow-milk pathway.
- The influence of hot particles on time-integrated ingestion doses from ^{90}Sr and ^{137}Cs is also quantified. For a large proportion (90-100%) of fallout in slowly-dissolving hot particles, ingestion doses are reduced by a factor of approximately two compared to the case where all radioactivity is deposited in bioavailable forms. For rapidly dissolving hot particles, however, the influence on ingestion doses is relatively minor.
- Significant uncertainties in dose estimates remain, particularly concerning the role of direct ingestion of hot particles by animals and humans.

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Figures

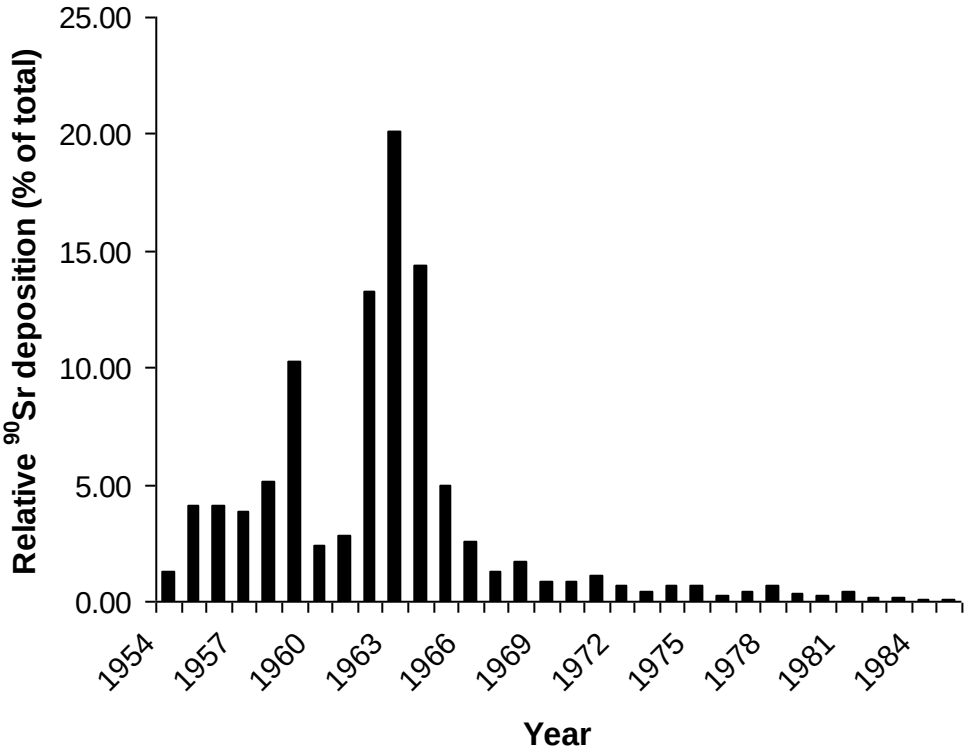

Figure 1. Relative deposition of nuclear weapons test ^{90}Sr in the Northern Hemisphere, expressed as a percentage of total fallout to 1985. From data presented in Cambray et al. (1989).

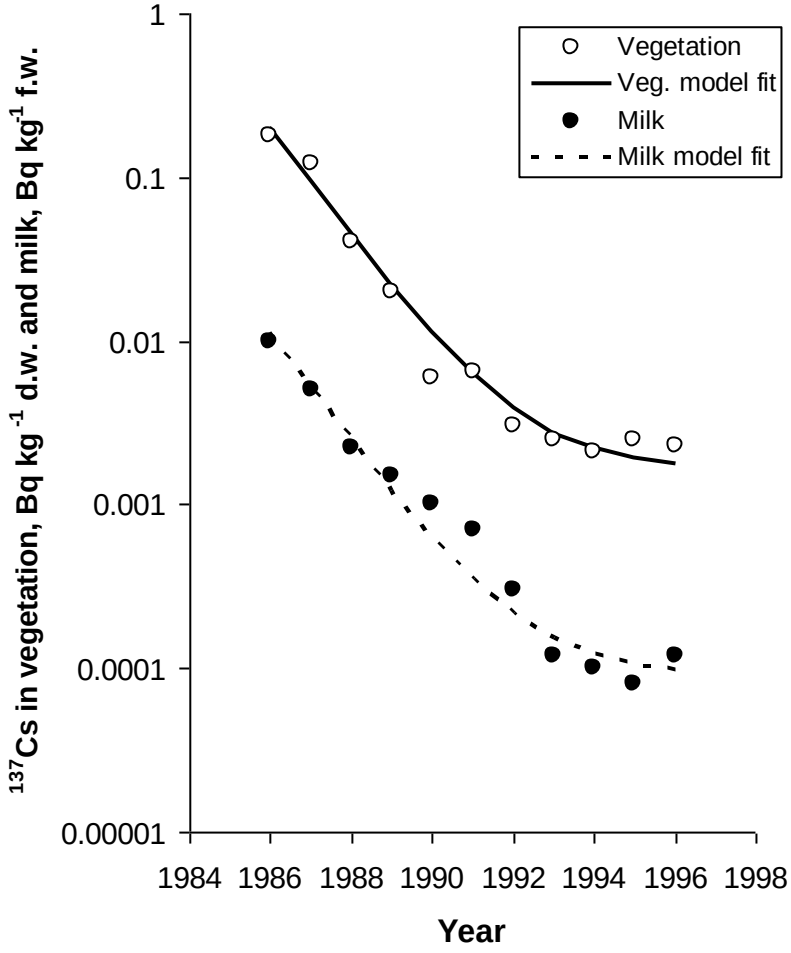

Figure 2. Model fits to annual mean ^{137}Cs activity concentration in grassy vegetation and milk after the Chernobyl accident (from data in Travnikova et al., 1999). Data has been normalised to 1 Bq m^{-2} total ^{137}Cs deposition.

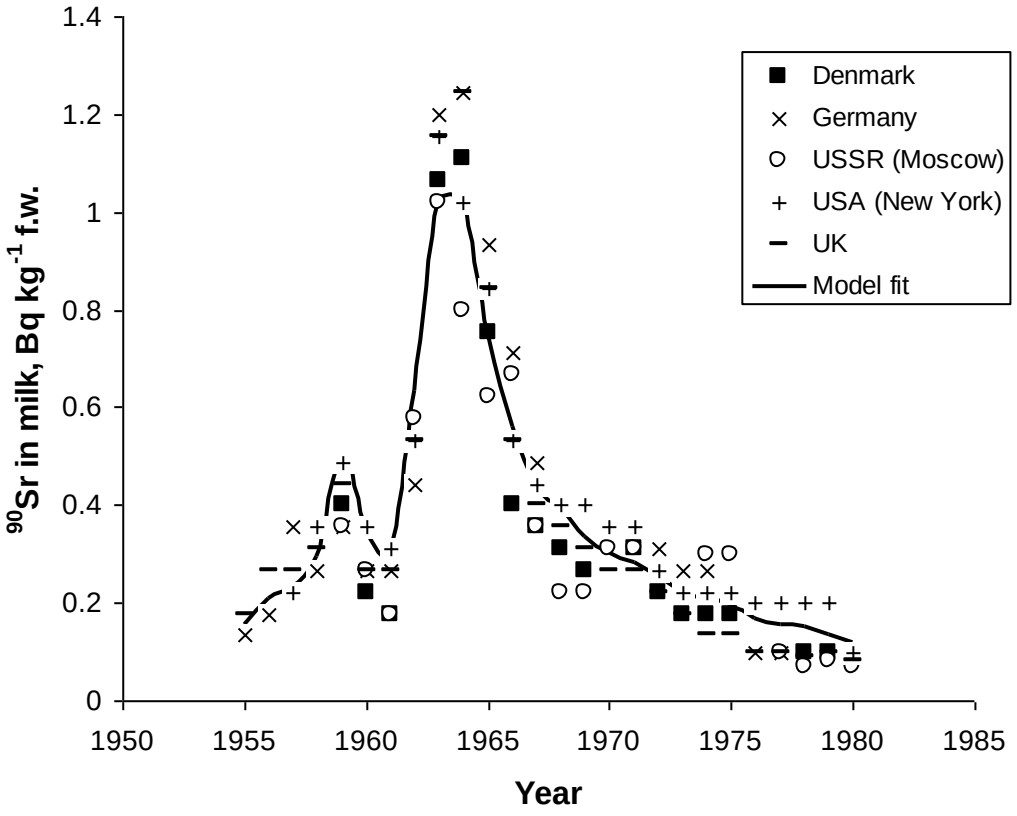

Figure 3. Model fit to annual mean ^{90}Sr activity concentration in milk in various Northern Hemisphere countries after nuclear weapons testing (from data in UNSCEAR, 1977, 1982). Fallout for the period 1954-85 in these areas was approximately 2900 Bq m^{-2} (from data in UNSCEAR, 1977; Cambray et al., 1989).

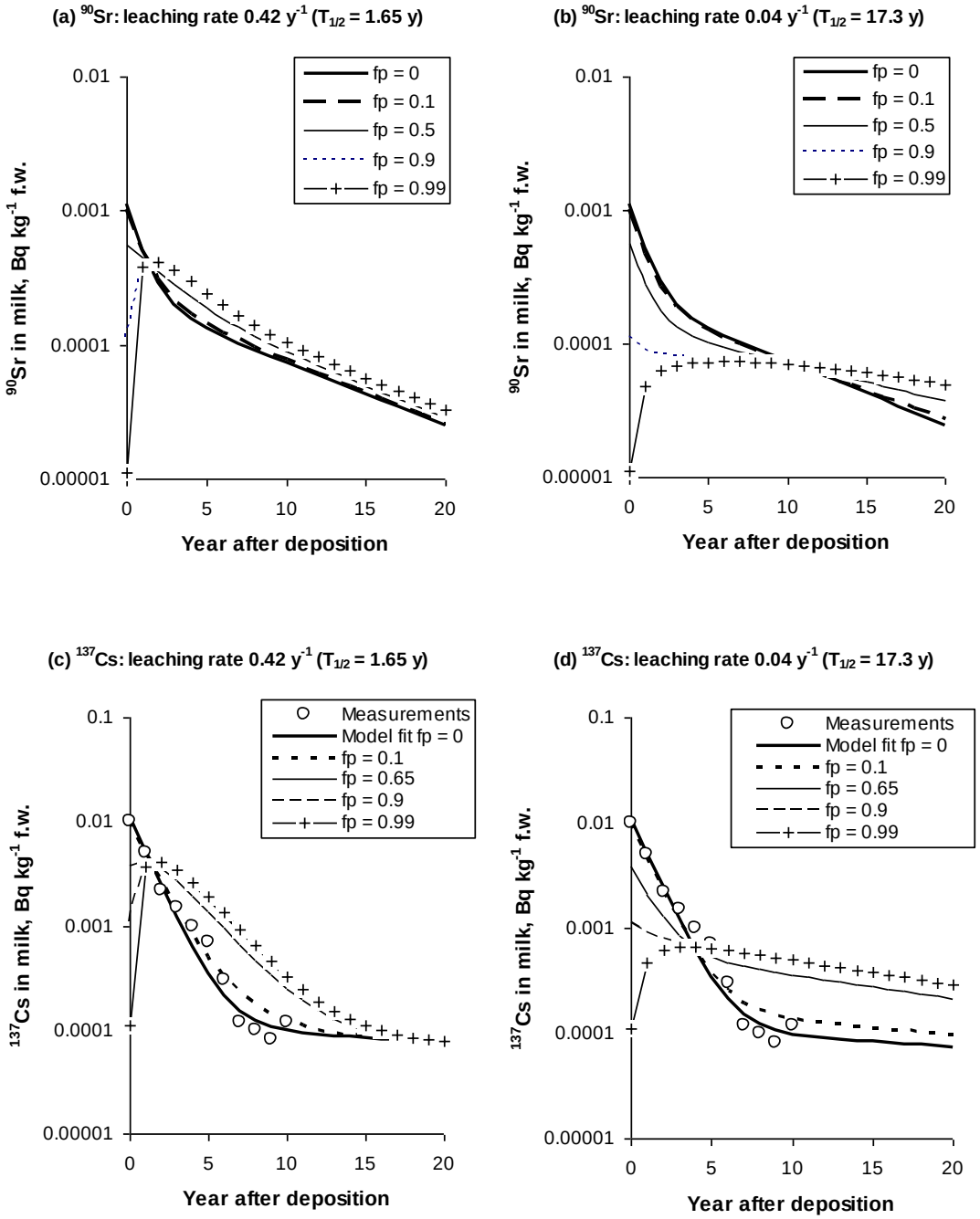

Figure 4. Model predicted activity concentrations of ^{90}Sr and ^{137}Cs in milk following a deposition of 1 Bq m^{-2} of each radionuclide. Predictions are given for different fractions, f_p, of radionuclide in unavailable fuel particle form and for different fuel particle dissolution rates.

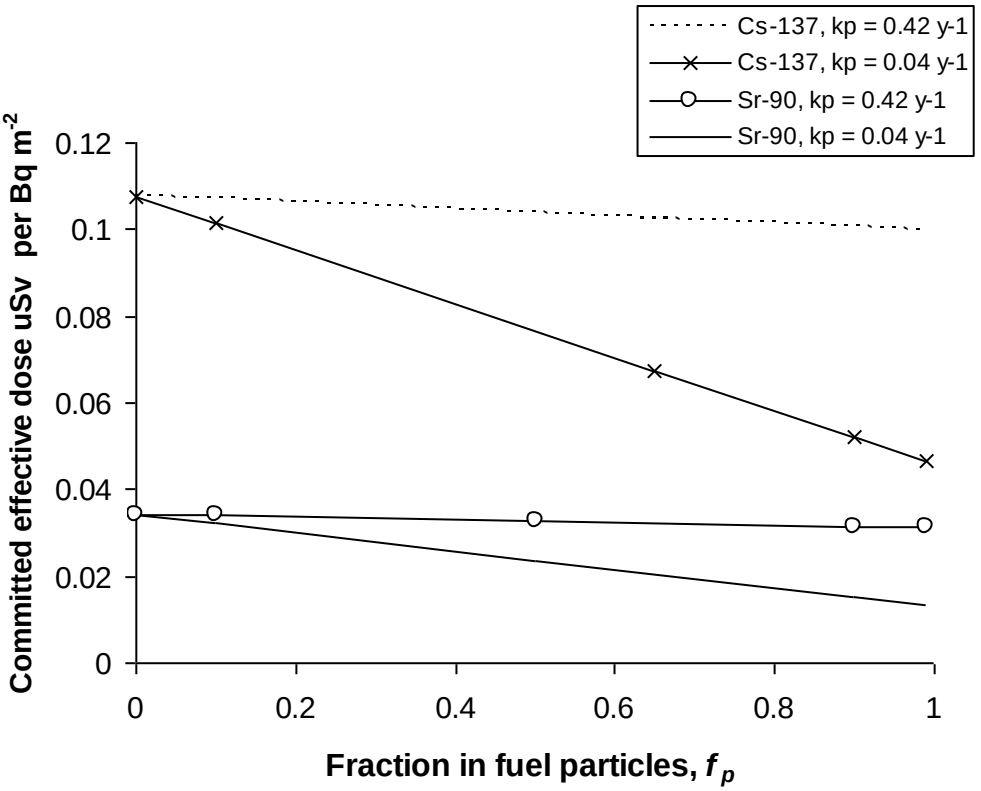

Figure 5. Time-integrated (over a 20 year period after fallout) committed effective dose to adults via the milk pathway vs. the fraction of ^{90}Sr and ^{137}Cs deposited in fuel particles. Relationships are shown for different values of the fuel particle dissolution rate, k_p.

Tables

Table 1. Fitted parameter values for the model for transfers of ^{90}Sr and ^{137}Cs to milk.

Radionuclide	**Fitted parameter value**	
Cs-137	$\beta = 0.198$ m^2 kg^{-1}	$k_2 = 0.741$ y^{-1}
	$\gamma = 0.0021$ m^2 kg^{-1}	$k_3 = 0.0$ y^{-1}
	$CR = 0.055$ kg kg^{-1}	
Sr-90*	$\beta \times CR = 0.00903$ m^2 kg^{-1}	$k_2 = 1.00$ y^{-1}
	$\gamma \times CR = 0.00218$ m^2 kg^{-1}	$k_3 = 0.085$ y^{-1}

* Since the model was fitted directly to the measurements of activity concentration in milk, the fitted parameters β, γ were determined as the product of these parameters and the milk-vegetation concentration ratio.

Table 2. Fraction of total deposited activity not contributing to time-integrated dose as a result of radioactive decay whilst being chemically unavailable in hot particles. The values shown are for different values of f_p and k_p and represent the fraction by which the ingestion dose is reduced compared to the case of zero hot particle depositon ($f_p = 0$).

	Cs-137		**Sr-90**	
f_p	$k_p = 0.04$	$k_p = 0.42$	$k_p = 0.04$	$k_p = 0.42$
0	0	0	0	0
0.1	0.036	0.005	0.040	0.005
0.3	0.11	0.015	0.11	0.016
0.5	0.18	0.026	0.19	0.027
0.7	0.26	0.036	0.26	0.038
0.9	0.33	0.047	0.34	0.049
1.0	0.36	0.052	0.38	0.054