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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.

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 1952–1957 (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 et al. (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.

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Even at large distances from Chernobyl, radionuclides could be deposited in particulate form. Fuel particles of $<\sim 1\ \mu$m 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 4990–139,000 Bq of ^{103}Ru and 1050–28,000 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 those 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:

$$A(t) = A_0 \exp(-k_p t) \tag{1}$$

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, $T^{p}_{1/2}$, is given by ($T^{p}_{1/2} = \ln 2/k_p$).

Let the fraction of the total 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:

$$D_a(t) = D\left(1 - f_p \exp(-(k_p + \lambda)t)\right) \tag{2}$$

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 and 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):

$$\begin{aligned} C_v(t) = {} & D[\alpha \exp(-(k_1 + \lambda)t) + \beta \exp(-(k_2 + \lambda)t) \\ & + \gamma \exp(-(k_3 + \lambda)t)] \end{aligned} \tag{3}$$

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 Eq. (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 Eq. (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:

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

The activity concentration of the radionuclide in vegetation in year i is, from Eqs. (3) and (4):

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

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:

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

or

$$C_m = F_m C_v I_f \tag{7}$$

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.

Eq. (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:

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

By definition,

$$C_m = C_v \times \text{CR} \tag{9}$$