ORNL-5786

OAK RIDGE
NATIONAL
LABORATORY

MARTIN MARIETTA

A Review and Analysis of Parameters for Assessing Transport of Environmentally Released Radionuclides through Agriculture

C. F. Baes III
R. D. Sharp
A. L. Sjoreen
R. W. Shor

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

5. MISCELLANEOUS PARAMETERS

Other default parameters included in the TERRA code are the weathering removal constant, λ_w, the metabolic removal rate constants from milk and beef, λ_m and λ_f, respectively, and the lifetime grain and forage requirements of cattle on feed, Q_g^{fc} and Q_f^{fc}, respectively. The weathering removal constant is extremely important in calculating surface plant concentrations due to direct deposition processes, and the latter four parameters are utilized in calculating beef and milk concentrations.

5.1 The Weathering Removal Loss Constant, λ_w

After radionuclides are initially deposited on vegetation surfaces environmental processes (in addition to radiological decay) will begin to remove the deposited material. Miller and Hoffman[225] have reviewed the literature on weathering removal of radionuclides from vegetation. They classify the environmental removal processes as wind removal, water removal, growth dilution, and herbivorous grazing. Wind removal may be very effective in removal of freshly deposited large particles (> 1 µm diameter), but not nearly as effective after the first few days. Submicron particles may be released from plant surfaces during periods of rapid growth and high transpiration rates. Also, surface abrasion from wind action may dislodge salt particles, wax, and other surface fragments. Radioactivity associated with these components would also be removed from the vegetation.

Precipitation, fog, dew, and mist—all may remove surface-deposited radionuclides via direct washoff and leaching. Leaching, in addition, may remove radionuclides incorporated into plants through root uptake. Wash-off, like wind removal, seems to be most effective on freshly deposited material. Precipitation falling as a light, continuous drizzle is more efficient than a large quantity of precipitation falling over a much shorter period.[225]

Removal due to growth dilution and grazing by herbivores may vary considerably by plant and location. Produce growth characteristics may be quite varied. Slow-growing varieties may be expected to be less affected by growth dilution than faster growing varieties. Grazing by herbivores may be particularly hard to predict. Weathering removal tends to occur in an exponential manner with a characteristic half-time, T_w.[225] From this half-time a weathering removal constant, λ_w, may be derived according to

$$\lambda_w = \frac{ln2}{T_w}. \qquad (47)$$

In the TERRA code the value of λ_w adopted by the USNRC[6] of 5.73×10^{-7} (equal to a T_w of 14 d) is used for all radionuclides (except for iodine) on all plant surfaces. This value is somewhat arbitrary, but is within the range of reported values in the literature. In their literature review Miller and Hoffman[225] found measured values of T_w to range between 2.8 to 34 days with a geomeiric mean of all reported values of 10 days. For I_2 vapor, iodine particulates, and other particulates on herbaceous vegetation the geometric means of reported values of T_w are 7.2, 8.8, and 17 days, respectively. The value of T_w used in TERRA is 1.0×10^{-6} s^{-1}, which corresponds to a T_w of 8 days.

5.2 The Metabolic Turnover Constant For Milk, λ_m

In the TERRA code radionuclide transfers to beef and milk are modeled via a single compartment model whereby the radionuclide is transferred from feed directly to milk and beef. This approach differs from the approach taken by the USNRC[6] in that isotopes of the same element with significantly different half-lives may yield different milk and beef concentrations, even though the milk and beef transfer coefficients (F_m and F_f, respectively) are the same for the isotopes. Such one-compartment models require quantification of all inputs and outputs from the compartment. For milk and beef the metabolic removal constants must be known.

The model for radionuclide transfer to milk is given by

$$C_m = \frac{C_{feed} Q_{feed} f_{tm} (1 - \exp - (\lambda_m t_m))}{m_p \lambda_m}, \tag{48}$$

where

C_m	=	the radionuclide concentration in milk (Bq or Ci/kg),
C_{feed}	=	the radionuclide concentration in feed (Bq or Ci/kg),
Q_{feed}	=	the ingestion rate of feed (kg/s),
f_{tm}	=	the fractional transfer from ingested feed to milk (unitless),
λ_m	=	the metabolic turnover constant for milk (s^{-1}),
t_m	=	the time at which milk is sampled (s), and
m_p	=	the quantity of milk collected per milking (kg).

At equilibrium Eq. (48) reduces to

$$C_m = \frac{C_{feed} Q_{feed} f_{tm}}{m_p \lambda_m}. \tag{49}$$

Since by the USNRC[6] approach,

$$C_m = 86{,}400 C_{feed} Q_{feed} F_m , \tag{50}$$

where 86,400 = the number of seconds in a day, then

$$f_{tm} = 86{,}400 F_m m_p \lambda_m . \tag{51}$$

Since F_m and m_p are already known (from reference 7 m_p = 13.4 kg), then the only parameter which needs to be defined is λ_m.

Ng and his associates[145] have determined values of metabolic halftimes, T_m, for various elements in milk (Fig. 5.1: note that these values of T_m are in terms of days rather than seconds). They consider a value of T_m of 0.693 d (equal to ln 2) to be conservative. Such a value of T_m is equivalent to a λ_m of 1.0/d or 1.16×10^{-5}/s. This latter value is adopted for calculation of milk concentrations in the TERRA code. Using this value in Eqs. (49) and (51) allows for an equilibrium milk concentration to be achieved within approximately seven days.

5.3 The Metabolic Turnover Constant For Beef, X,

The metabolic turnover constant for beef is determined in a manner similar to that for milk by substituting the fractional transfer to beef, f_{tf}, the time to slaughter, t_s, the muscle mass of beef cattle, m_m, the metabolic turnover constant for beef, λ_f, and the beef transfer coefficient, F_f for the respective parameters f_{tm}, t_m, m_p, λ_m, and F_m in Eqs. (49)-(51). However, estimates of λ_f do not appear to be available in the literature. In fact, the question of whether equilibrium beef concentration ever occurs for some radionuclides has never been completely resolved. As default in

	I A	II A	III B	IV B	V B	VI B	VII B	VIII	VIII	VIII	I B	II B	III A	IV A	V A	VI A	VII A
II	Li 0.693	Be 0.80											B 0.693		N 0.693		F 0.693
III	Na 17	Mg 0.693											Al 0.693	Si 0.693	P 1.97	S 0.693	Cl 0.693
IV	K 5.3	Ca 1.01	Sc 0.693	Ti 0.693	V 0.693	Cr 0.693	Mn 0.693	Fe 0.693	Co 0.693	Ni 0.693	Cu 0.693	Zn 2.71	Ga 0.693	Ge 0.693	As 0.693	Se 2.21	Br 0.693
V	Rb 0.54	Sr 2.11	Y 0.693	Zr 0.693	Nb 0.693	Mo 0.89	Tc 0.693	Ru 0.693	Rh 0.693	Pd 0.693	Ag 0.693	Cd 0.693	In 0.693	Sn 0.693	Sb 0.693	Te 1.35	I 1.01
VI	Cs 0.93	Ba 1.58		Hf 0.693	Ta 0.693	W 0.863	Re 0.67	Os 0.693	Ir 0.693	Pt 0.693	Au 0.693	Hg 0.693	Tl 2.19	Pb 3.33	Bi 0.693	Po 1.15	At 0.693
VII	Fr 0.693	Ra 1.97															

Lanthanides	La 0.693	Ce 0.693	Pr 0.693	Nd 0.693	Pm 0.693	Sm 0.693	Eu 0.693	Gd 0.693	Tb 0.693	Dy 0.693	Ho 0.693	Er 0.693	Tm 0.693	Yb 0.693	Lu 0.693
Actinides	Ac 0.693	Th 0.693	Pa 0.693	U 0.693	Np 0.693	Pu 0.693	Am 0.693	Cm 0.693							

Key: Li — Symbol; 0.693 — Transfer Coefficient, T_m

Figure 5.1. Metabolic half-times for the elements in milk (days), based on reference 145.

TERRA we have assumed that equilibrium does, indeed, occur, and a λ_f of 5.73×10^{-7}/s (equal to a T_f of 14 d) is reasonable. Such a turnover rate constant allows for equilibrium to be achieved after approximately 90 days.

5.4 Lifetime Grain and Forage Requirements For Cattle On Feed, Q_g^{fc} and Q_f^{fc}, Respectively

In calculating radionuclide transport into beef the average annual lifetime feeding schedule of the cattle is combined with the predicted radionuclide concentrations in the feed to predict average annual intake of radionuclides by the cattle. For milk cows and "all other" cattle the inventory feeding schedules may be used in the calculation because slaughtered individuals from these categories may be assumed to have always resided in their respective category. However, lifetime grain and forage requirements for cattle on feed are different from the inventory grain and forage requirements (discussed in the report by Shor, Baes, and Sharp[7]) which are used in the calculation of pasture production (Sect. 4.1) because they take into account the movement of the individuals from one inventory category to another. These lifetime average feeding rates are used in the calculation of beef concentrations in the TERRA code.

Since the cattle in feedlots are slaughtered after an average occupancy of six months, and since they enter and leave the feedlot throughout the year, the lifetime feeding rate of grain and forage is a mix of the feeding schedules in the inventory categories "all other cattle" and "cattle on feed." For example, an animal entering the feedlot at the beginning of the year would have been fed on the feedlot schedule only before slaughter, but those entering thereafter until the end of the year would have been fed a combination of the feedlot and "all other cattle" schedules before slaughter. In determining the lifetime feeding schedule of slaughtered cattle from feedlots, we assume that entry and exit from the feedlot is at a constant rate equal to $s_g/365$ or $n_g/182.5$. The ideal animal entering the lot is 9 months old and is fed for 6 months or 182.5 days. In order to find an average feeding rate for this animal, his feed is added over the last 13.5 months of his life (the first 1.5 months is assumed to be on milk) and 12/13.5 of this amount is his annual rate of feeding. From Table 17 of reference 7 the daily grain consumption rate for cattle on grain is 5.0 kg/d (equal to 1820/365). The comparable rate for forage is 2.7 kg/d. The respective rates for the "all other cattle" category are 0.4 kg/d for grain and 8.3 kg/d for forage. Therefore the totals for grain and forage for the last 13.5 months of life are 910 kg and 1003 kg, respectively. The annual rates are 891 kg and 2108 kg for grain and forage, respectively. These rates are used in the TERRA code in the calculation of radionuclide concentrations in beef from slaughtered feedlot cattle.

5.5 The Carbon and Water Content of Foods

In the TERRA code concentrations of tritium (H-3) and carbon-14 in foods are calculated according to a model which assumes that the specific activities of tritium and carbon-14 in foods at a given location are the same as the specific activities of H-3 and C-14 in atmospheric H_20 and CO_2, respectively (equilibrium is assumed). Thus, the first step in calculating activity concentrations of tritium and carbon-14 in food is calculating their respective activity concentrations in atmospheric water vapor and carbon dioxide. For tritium, this calculation is made by utilizing the SITE parameter, absolute humidity, H, by the equation

$$C_{wv}^{H3} = 1000\,\frac{C_a^{H3}}{H}\,, \tag{52}$$

where

C_{wv}^{H3} = the activity concentration of tritium in atmospheric water vapor (Bq or Ci/kg),
C_a^{H3} = the activity concentration of tritium in air based on the atmospheric dispersion calculation (Bq or Ci/m^3), and
H = the absolute humidity (g/m^3.

Once the specific activity of H-3 in atmospheric water vapor is calculated, then the same activity in the atmospherically derived water of vegetable produce, beef, and milk is assumed. That is

$$C_{food}^{H3} = C_a^{H3} \cdot f_w^a \; , \tag{53}$$

where

C_{food}^{H3} = The tritium activity concentration in food (Bq or Ci/kg) and
f_w^a = the fraction of water in food derived from atmospheric sources (unitless).

Traditionally, the tritium concentration in food has been assumed to be 50% of tritium concentration in air (f_w^a = 0.5) based on a model by Anspaugh, et a1.[226] However, recent empirical evidence suggests that tritium concentration in vegetation under chronic exposure conditions is nearly equal to the tritium air concentration (f_w^a = 1.0).[227] In the TERRA code the default is the latter assumption.

The water content of the produce categories may be derived from the dry-to-wet weight conversion factors presented in Table 2.3. The value (1.0 – the listed conversion factor) gives the kilograms of H_20 per kilogram fresh produce. For beef and milk, reference 14 yields 0.615 and 0.87 kilograms of water per kilogram of fresh, uncooked food, respectively. The water content of leafy vegetables is assumed to be 0.934 (Table 5.1).

A specific activity approach, analogous to that for tritium, is used for carbon-14. The specific activity of C-14 in atmospheric CO_2 is given by

$$C_{cd}^{C14} = 1000 \frac{C_a^{C14}}{0.18} \; , \tag{54}$$

where

C_{cd}^{C14} = the activity concentration of carbon-14 in atmospheric CO_2 (Bq or Ci/kg),
C_a^{14} = the activity concentration of carbon-14 in air based on the atmospheric dispersion calculation (Bq or Ci/m^3), and
0.18 = the average concentration of CO_2 in the atmosphere (g/m^3), corresponding to 330 ppm by volume.[228]

The carbon content of the food categories in TERRA, based on a recent review by Killough[229] and supplemental information from reference 14, is given in Table 5.2.

5.6 Coarse (2.5 - 15 m) Suspended Particulate Matter

Resuspension of material deposited on surface soils is calculated in TERRA via a mass loading approach.[230] In such an approach the specific activity of a radionuclide in resuspended material is assumed to be the same as the specific activity of surface soil. Thus, the calculation of surface soil concentration is used together with the quantity of resuspended material in the air (mass loading) to calculate an air concentration due to resuspension. This air concentration is given by

$$C_a^r = \frac{C_s^s P_{sus}}{1 \times 10^9} \; , \tag{55}$$

Table 5.1. Water content of produce, beef, and cow's milk

Food	Water content[a]	Weighting factor[b]	Food	Water content
Leafy vegetables			Beef	
Broccoli	0.899	3.7	Chuck	0.65
Brussel sprouts	0.849	0.6	Flank	0.61
Cabbage	0.924	22.0	Hamburger	0.55
Cauliflower	0.917	2.8	Liver	0.697
Celery	0.937	15.5	Porterhouse	0.58
Escarole	0.866	1.1	Rib roast	0.59
Green onions	0.876	2.6	Round	0.69
Lettuce	0.948	46.0	Rump	0.55
Spinach greens	0.927	5.7	Sirloin	0.62
Weighted average	0.934		Average	0.615
Exposed produce[c]	0.874		Whole cow's milk	0.870
Protected produce[c]	0.778			
Grain foods[c]	0.112			

[a]Kilograms of water per kilograms fresh, unprepared produce or edible portions of uncooked food (reference 14)

[b]Relative importance based on production in kilograms (% of total) in the conterminous United States.

[c]Based on values given in Table 2.3.

where

C_s^s = surface soil (depth = 1 cm) concentration (Bq or Ci/kg),
1×10^9 = the number of micrograms per kilogram (μg/kg),
C_a^r = resuspension air concentration (Bq or Ci/m^3), and
P_{sus} = suspended particulate matter (μg/m^3).

In TERRA the mass loading value P_{sus} is based on data reported by the EPA.[231] This parameter represents the 2.5-15 μm diameter particle fraction collected by either the Size-Selective Inlet (SSI) hi vol or the dichotomous samplers operated as part of the Inhalable Particulate Network (IPN) operated by EPA's Environmental Monitoring and Support Laboratory, Research Triangle Park. Inhalable suspended particulate matter appears to be bimodally distributed into fine and coarse particle sizes. The fine fraction (<0.1-2 μm) are mostly generated by fossil fuel combustion and atmospheric photochemistry processes. The coarse fraction (2.5-l5 μm) is primarily a result of windblown dusts, mechanical processes, and pollen.

The value of P_{sus} of 15.5 μg/m^3 used as default in TERRA is the geometric mean of values taken from the April 1979-June 1980 IPN summary (Fig. 5.2). The data are reported for 46 sampling locations in the conterminous United States, and represent annual arithmetic averages for each station. As shown in Fig. 5.2, the parameter P_{sus} is lognormally distributed. The range of measured values is from 3.2 to 52.4 μg/m^3.

Table 5.2. Water content of produce, beef, and cow's milk

Food	Carbon content[a]	Weighting factor[b]	Reference
Leafy vegetables			
Broccoli	0.042	3.7	229
Brussel sprouts	0.065	0.6	229
Cabbage	0.032	22.0	229
Cauliflower	0.035	2.8	229
Celery	0.024	15.5	229
Escarole	0.056	1.1	14
Green onions	0.053	2.6	14
Lettuce	0.020	46.0	229
Spinach greens	0.028	5.7	229
Weighted average	0.026		
Exposed produce[c]			
Apple	0.070	15.4	229
Asparagus	0.030	0.6	229
Bushberries	0.070	1.6	229
Cherry	0.074	0.7	14
Cucumber	0.016	4.0	14
Eggplant	0.031	0.1	14
Grape	0.083	20.2	229
Peach	0.056	6.9	229
Pear	0.076	3.5	229
Plums and prunes	0.062	3.1	229
Sweet pepper	0.033	1.3	14
Snap bean	0.047	0.7	229
Squash	0.021	1.8	229
Strawberry	0.044	1.3	229
Tomato	0.025	38.8	229
Weighted average	0.050		
Beef	0.228		229
Protected produce			
Bean (dry	0.198	2.2	229
Cantaloupe	0.025	1.1	229
Carrot	0.049	2.4	229
Grapefruit	0.048	5.5	14
Lemon	0.047	2.4	14
Onion	0.054	3.6	14
Orange	0.055	22.8	229
Peanut	0.574	3.4	229
Peas	0.114	0.4	14
Potato	0.095	33.7	229
Sugarbeet	0.051	6.5	14
Sugarcane	0.438	5.5	229
Sweet corn	0.118	6.0	229
Sweet potato	0.137	1.5	229
Tree nuts	0.659	0.4	229
Watermelon	0.034	2.6	14
Weighted average	0.116		
Grains			
Barley	0.395	10.1	229
Corn (for meal)	0.118	37.7	229
Oats	0.431	2.3	229
Rye	0.396	0.5	229
Soybean	0.465	5.3	229
Wheat	0.391	44.0	229
Weighted average	0.293		
Whole cow's milk	0.069		14

[a]Kilograms of carbon per kilograms fresh, unprepared produce. Based on protein, fat, and carbohydrate content of 50, 76, and 44%, respectively.

[b]Relative importance based on production in kilograms (% of total) in the conterminous United States.

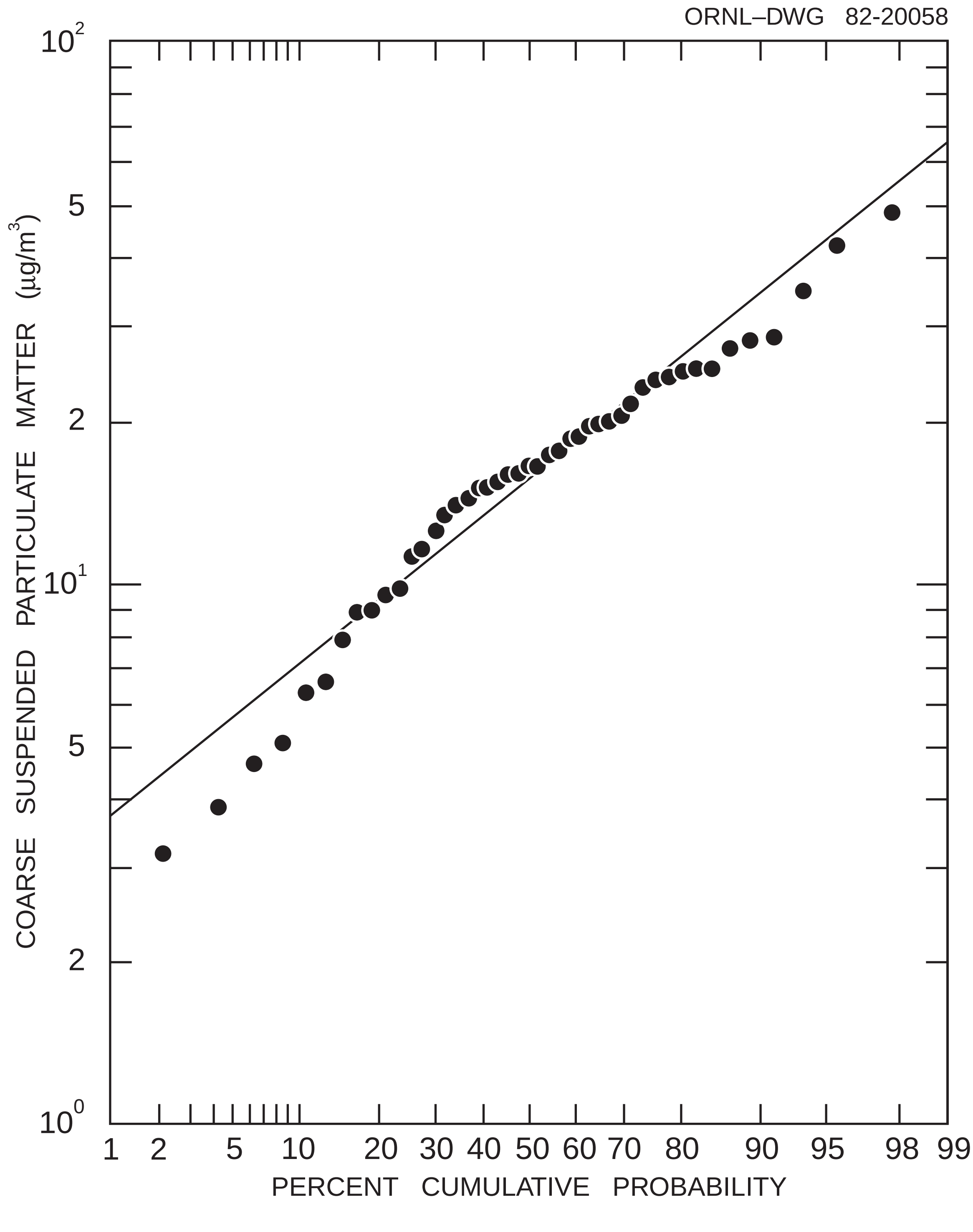

Figure 5.23. Lognormal probability plot of coarse suspended particulate matter (2.5 – 15 μm).

Resuspended material may contribute to plant surface concentrations before and after termination of the atmospheric source term. In TERRA a deposition rate of the resuspended activity is calculated according to

$$D_r^r = \frac{C_a^r V_d^r}{100} , \tag{56}$$

where

D_r^r = the deposition rate of resuspended material (Bq or Ci/m^2/s),
V_d^r = deposition velocity of the resuspended material (cm/s), and
100 = the number of centimeters in a meter (cm/m).

The value of V_d^r used in TERRA is 0.1 cm/s, which is a reasonable estimate for particle diameters between 2 and 15 µm, a friction velocity of 30 cm/s, and particle densities >l g/cm^3 as shown by Sehmel[232] (Figure 5 in reference 232).