Journal of Food Protection, Vol. 71, No. 12, 2008, Pages 2504–2513

A Chain Modeling Approach To Estimate the Impact of Soil Cadmium Pollution on Human Dietary Exposure

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MS 08-050: Received 25 January 2008/Accepted 2 June 2008

ABSTRACT

Cadmium in soil poses a risk for human health, due to its accumulation in food and feed crops. The extent of accumulation depends strongly on soil type and the degree of pollution. The objective of the present study was to develop a predictive model to estimate human dietary cadmium exposure from soil characteristics. This chain model consists of three basic steps: (i) calculation of plant cadmium levels from soil contamination levels and soil characteristics, (ii) calculation of animal transfer from consumption and contamination levels, and (iii) human exposure from both plant and animal products. Six soil scenarios were assessed, reflecting a specific contaminated region and ranging from 0.5 mg/kg of Cd (pH 4.5) to 2.5 mg/kg of Cd (pH 5.5). Cadmium levels in feed crops and vegetables were estimated with regression and mathematical models. Animal exposure and transfer to cattle kidneys, livers, and meat were calculated using a consumption database and a parameterized linear simulation model. Human exposure was estimated by Monte Carlo simulation, using a consumption database. The median human exposure for the different scenarios ranged from 0.24 to 0.98 μg/kg of body weight per day, which is comparable to results obtained from exposure levels based on observed field contamination data. The study shows that a chain model approach from soil contamination to human exposure, including animal exposure and transfer to animal products, can successfully be applied. The model can be used for fast evaluation of dietary cadmium exposure and the identification of risk areas based on soil conditions.

Awareness has grown that soil is an important factor in the feed and food supply chain in order to deliver safe and high-quality products. Whether dealing with home-grown or industrially processed vegetables or animal feed, the transfer of contaminants from soil to a specific product is of importance regarding the quality and safety of food and feed. The role of the soil in maintaining food safety is explicitly mentioned in the European Thematic Strategy for Soil Protection *(35)*. The presence of heavy metals like cadmium in soils used to produce food or feed poses a public health risk due to the accumulation of the contaminant in food and feed and the subsequent accumulation in the human body over time. This accumulation can result in a variety of health problems, with impairment of kidney function being the main adverse effect *(19)*. The degree to which cadmium is available for plant uptake and further accumulation in edible plant parts as well as animal target organs (such as kidneys, liver, meat) depends strongly on the degree of pollution and soil characteristics *(24, 26, 41)*.

In 2001, the European Union introduced food quality standards for cadmium contamination levels in crops for feed and food production, which are implemented to protect consumers from an unacceptable dietary exposure to cadmium as a result of intake of food *(12)*. In some areas in The Netherlands and other European countries, diffuse pollution with cadmium has resulted in elevated levels in soil. Regional differences in cadmium levels and soil characteristics will lead to differences in cadmium levels in feed and food crops and eventually to differences in human dietary exposure to cadmium.

In the Kempen area on the border of The Netherlands and Belgium, soil cadmium concentrations are relatively high (0.5 to 5 mg/kg) as compared with other regions in The Netherlands (average is below 0.3 to 0.5 mg/kg) *(5, 26)*. The main reason for the elevated cadmium (as well as lead and zinc) levels in the soil of this specific area is the presence of a zinc smelter near the town of Budel. In the period 1882 to 1983, emission estimates from this factory ranged from 66 to 178 tons of cadmium, which resulted in a polluted area of approximately 350 km^2 *(7)*. Although the levels of cadmium in soil are not extremely high and are below the level at which further investigation is required (the Dutch intervention for sandy Kempen soils is approximately 7 mg kg^{-1} in soil), the soils are mostly acidic, with pH levels below 5.5. At these low pH levels, the availability of cadmium in soil is high, which can lead to high uptake by plants, despite the rather low total cadmium content in the soil *(10)*. Indeed, crop samples from private gardens and agricultural soils with cadmium levels exceeding the food quality standard *(2)* occurred primarily in soils with a pH lower than 5.5 and cadmium concentrations between 0.2 and 0.6 mg kg^{-1} *(26)*.

Human risks of soil cadmium contamination are related to the degree to which the metal enters the food chain. Uptake by crops, fodder, animals, and ultimately the con-

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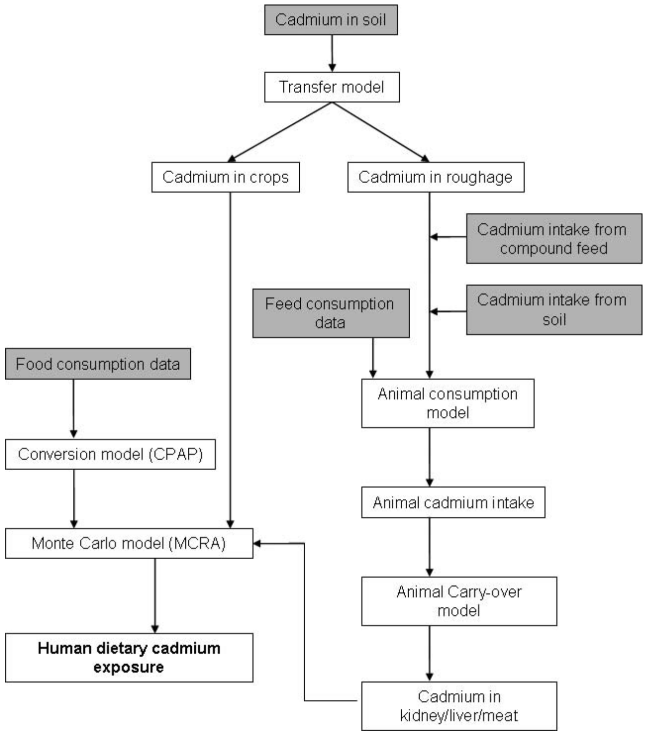

FIGURE 1. *Flow chart of the chain model to estimate human dietary exposure to cadmium from animal and plant products, based on soil cadmium levels and soil quality parameters.*

sumer depends on soil properties, animal properties, plant properties, and consumption patterns. Most human exposure assessments concerning the dietary intake of heavy metals like cadmium are based on measured concentrations in food products *(6, 18, 28, 42–44)*. However, for specific regions, monitoring data are often not available. The objective of the present study was to estimate human dietary exposure to cadmium on a regional level, using a predictive modeling approach taking into account the supply chain from soil to the consumer. An exposure model was developed that links soil cadmium levels and soil characteristics to intake in plants, dietary cadmium transfer by animals, and finally, consumer exposure from dietary intake of plant and animal products. The model is subsequently applied to different soil scenarios, reflecting the conditions in the Kempen area. The approach is designed in such a way that the model can also be applied to other regions, using region-specific data.

MATERIALS AND METHODS

Model overview. To assess the extent to which cadmium in soil poses a risk for human health by food consumption, a human-exposure chain model was developed. The model integrates soil cadmium levels, soil characteristics, soil-to-plant transfer, animal intake, transfer to animal products, and human consumption patterns to estimate human cadmium exposure from dietary intake. (A schematic overview of the model is given in Figure 1.) As a baseline scenario, the average exposure of the Dutch population was determined based on an average consumption pattern and cadmium levels in food products, as measured in national monitoring programs. As a regional case, the human dietary exposure to cadmium of the Kempen population was assessed for six situations reflecting typical conditions of soils in the Kempen area, with a variation in pH and soil cadmium levels as observed throughout the region in arable soils (Table 1). For each of these soil scenarios, the cadmium concentrations in cattle organs and vegetables were calculated with the models described below. These predicted cadmium levels were subsequently used to estimate the dietary cadmium exposure of the Kempen population. It was assumed that vegetable products and cattle organs and meat consumed were produced locally. For vegetables and food products for which no predictive model estimates were available, contamination data from the national monitoring programs were used (average national contamination level). The model and data used are described in more detail in the following sections.

TABLE 1. *Description of the different soil scenarios evaluated in the exposure assessment*

Soil scenario	Cadmium level soil (mg/kg)	pH	Description
A	0.5	4.5	Clean: low pH
B	0.5	5.5	Clean: high pH
C	1	4.5	Average: low pH
D	1	5.5	Average: high pH
E	2.5	4.5	Contaminated; low pH
F	2.5	5.5	Contaminated: high pH

Model description. The model consisted of an animal module, a soil-plant module, and a human module. The soil-plant model was applied to the relationship between soil and roughage, and between soil and vegetables independently. Within the animal module, the cadmium levels in cattle organs were estimated using predictive models for cadmium uptake by grass and maize, cattle consumption patterns, and transfer characteristics. Within the crop module, the cadmium levels in different products were estimated using predictive models based on soil cadmium levels and soil characteristics (pH, organic matter content, clay content). The human module estimated the human dietary exposure to cadmium by Monte Carlo simulation, based on cadmium levels in food products and consumption data.

Modeling cadmium levels in cattle kidneys and liver. (i) Quantification of cadmium intake. The levels of cadmium in livers and kidneys of 6-year (1,825 days)-old cattle were estimated using the method described in Römkens et al. *(27)*. The daily intake (DI, expressed in milligrams per day) of cadmium by cattle in the Kempen area was calculated as the sum of the cadmium intake by soil ingestion, roughage (grass and maize in a ratio of 70 to 30%), and compound feed (feedstuffs that are blended from various raw materials and additives):

$$DI = \sum (Cd_{soil} \times F_{soil}) + (Cd_{compound\ feed} \times F_{compound\ feed}) + (Cd_{roughage} \times F_{roughage}) \quad (1)$$

where Cd_{soil}, $Cd_{compound\ feed}$, and $Cd_{roughage}$ are the cadmium levels (in milligrams of Cd per kilogram of dry matter) in, respectively, soil, compound feed, and roughage; and F_{soil}, $F_{compound\ feed}$, and $F_{roughage}$ ($=F_{grass} + F_{maize}$) are the cattle consumption levels of soil, compound feed, and roughage (in kilograms of dry matter per day). The cadmium intake by contaminated water was neglected due to the low contribution to the total cadmium intake *(27)*. The level of soil ingestion (F_{soil}) by cows was estimated to be 3% of the total roughage (grass and maize) consumption *(14, 34)*. The levels of cadmium (in milligrams of Cd per kilogram of

TABLE 2. *Regression parameters for the crops used in the present study to calculate cadmium levels (25, 32)*

Crop	n	INT	α	β	γ	δ	R^2	Dry matter (%)
Grass	14	1.45	NS[a]	NS	1.22	−0.38	0.63	88
Maize	39	0.90	NS	−0.32	1.08	−0.21	0.50	88
Potato	60	0.97	−0.41	−0.20	0.81	−0.21	0.78	24
Endive	87	2.35	−0.44	−0.18	0.58	−0.28	0.66	6
Leek	15	2.52	−1.22	−1.00	1.40	−0.24	0.48	10
Lettuce	69	2.55	−0.39	−0.19	0.85	−0.33	0.71	5
Spinach	36	2.19	−0.40	NS	0.77	−0.29	0.49	6
Tomato	40	1.52	−0.75	NS	0.51	−0.21	0.41	5
Carrot	100	1.00	NS	NS	0.29	−0.20	0.43	11
Cucumber	45	−0.86	NS	NS	0.74	NS	0.57	3
French beans	47	0.44	NS	NS	1.08	−0.33	0.69	11
Scorzonera	52	2.25	NS	NS	0.49	−0.44	0.74	23
Celery	103	1.29	NS	NS	0.65	−0.20	0.54	10
Radish	39	1.03	−0.39	−0.20	0.67	−0.11	0.74	8

[a] NS, not significant.

dry matter) in grass (Cd_{grass}) and maize (Cd_{maize}) were estimated with the following regression equation:

$$\log(\text{Cd}) = \text{INT} + \alpha \times \log(\text{OM}) + \beta \times \log(\text{clay}) + \gamma \times \log(\text{Cd}_{soil}) + \delta \times \text{pH} \quad (2)$$

where OM is the organic matter content (in percentage, fixed on 3% for all six soil scenarios, based on measurements in the Kempen area), clay is the fraction of clay particles (in percentage, fixed on 3% for all six soil scenarios), Cd_{soil} is the cadmium level in the soil (in milligrams per kilogram of dry matter), and pH is the pH of the soil *(5, 10).* The regression parameters INT (intercept), α, β, γ, and δ in equation 2 were estimated by regression analysis (stepwise multiple regression) with two national and one local (Kempen area) data sets *(25)* (Table 2). The data sets consisted of paired measurements, i.e., soil cadmium levels, soil characteristics, and plant cadmium levels, were measured simultaneously at the same location. The relations and individual parameter estimates presented in Table 2 were significant at the $P < 0.05$ level.

The level of cadmium in compound feed was obtained from the Dutch KAP (Quality of Agricultural Products) database, where the results of national monitoring programs for chemical compounds in foods and feed are stored (http://www2.rikilt.dlo.nl/kap/index.html) *(38).* Hereby, it was assumed that the compound feeds were obtained from nationally operating manufacturers, which produce their feeds predominantly from imported ingredients. The median cadmium level in compound feeds was 0.05 mg kg^{-1} of dry matter.

The average daily cattle consumption pattern was estimated for three different age classes (0 to 1, 1 to 2, and >2 years). Total roughage consumption ($F_{roughage}$) was estimated to be 4, 8, and 14 kg dm day^{-1}, respectively. This resulted in, respectively, 0.12, 0.24, and 0.42 kg of dry matter per day of soil ingestion (F_{soil}); 2.8, 5.6, and 9.8 kg of dry matter per day of grass consumption (F_{grass}); and 1.2, 2.4, and 4.2 kg of dry matter per day of maize consumption (F_{maize}). Compound feed consumption ($F_{compound\ feed}$) was estimated to be 0.24, 0, and 2.32 kg of dry matter per day *(25).*

Modeling cadmium levels in cattle kidneys and liver. (ii) Carry-over of cadmium to kidneys and liver of cattle. In practice, cadmium irreversibly accumulates in cattle organs *(4),* and estimates of biological half-life times of cadmium range from 40 days to several years or even infinite in animals *(39),* and from 10 to 30 years in humans *(21).* A linear assumption is supported by experimental data on the accumulation of cadmium in cattle *(3, 8, 30, 31),* contrary to other studies suggesting the development of a steady state *(39).* Considering some recent evaluations *(25, 40),* the transfer of cadmium to the kidneys and liver was calculated using a linear biotransfer rate (BTR), assuming an irreversible cadmium accumulation without excretion from the target organ. The latter implicates a slight overestimation at the end of the 5-year period, and can therefore be considered worst-case scenario. The cadmium concentration in the organs (C, in milligrams per kilogram), after period T (days), was calculated using the BTR (per 1 kg of tissue) and the DI:

$$C = \text{BTR} \times \text{DI} \times T, \quad \text{with BTR} = \frac{dC/dt}{dA/dt} \quad (3)$$

where dC/dt is the increase of the cadmium concentration in the organ tissue per day whereas dA/dt is the additional cadmium intake per day. Values of the BTR were estimated from literature data of experiments and were found to range between 0.4 and 5.7 $\times 10^{-4}$ for kidneys and between 0.12 and 3.2×10^{-4} for livers *(3, 30).* Here, values of 9.0×10^{-5} for kidneys and of 1.7×10^{-5} for liver were used. These data are derived from the generic bioconcentration factors (BCF) given by van Hooft *(37).* The BCF is a dimensionless overall coefficient relating the cadmium levels in organs and that of the average food (both in milligrams per kilogram):

$$\text{BCF} = \frac{\text{Cd}_{organ}}{\text{Cd}_{average\ in\ food}} \quad (4)$$

($Cd_{average\ in\ food}$) is equal to DI/M_{total}, where M_{total} stands for the total amount of products consumed (in kilograms per day). Using equations 3 and 4, the relation between the BCF and BTR then can be reduced to:

$$\frac{\text{BCF}}{M_{total}} = \text{BTR} \times T \quad (5)$$

For cows of 6 years of age, T is equal to 2,190 days, whereas the total food intake over that period equals 33,230 kg, which is the sum of intake of soil, grass, maize, and compound feed *(16).* The BCF for kidneys is equal to 2.99 kg^{-1} *(37),* which is then equivalent to a BTR of 9.0×10^{-5} kg^{-1}. This value is within the previously reported range and very close to the calculated BTR of 9.1×10^{-5} kg^{-1}, using data from an experimental study *(8).* For liver, the reported BCF by van Hooft is 0.554, which is then equivalent to a BTR of 1.7×10^{-5}, using the same assumptions as for the kidney. The model estimated the cadmium levels in cattle kidneys and livers. However, the animal product most consumed is meat. In order to get an estimation of cadmium levels in meat, we used the weighted mean cadmium concentration ratio kidneys/liver/meat of 134/31/1 to calculate concentrations in meat *(13, 20, 44).*

Modeling cadmium levels in vegetables. The levels of cadmium in various vegetables were calculated with equation 2. The regression parameters INT, α, β, γ, and δ in equation 2 were estimated by regression analysis (stepwise multiple regression) with two national data sets and one local (Kempen area) data set *(25)* (Table 2). The data sets consisted of paired measurements, i.e., soil cadmium levels, soil characteristics, and plant cadmium levels, and were measured at the same time at the same location. The relations and individual parameter estimates listed in Table 2 were significant at the $P < 0.05$ level.

Food consumption data. Food consumption data of the Dutch National Food Consumption Survey of 1997 and 1998 *(17)* were used to asses dietary cadmium exposure. This is the most recent measurement of consumption patterns of the Dutch population at the national level. In this survey, the food consumption of 6,250 individuals aged 1 to 97 years (of which 530 were young children aged 1 to 6 years) from 2,564 households was recorded on two consecutive days. The unit of intake for the calculations was 24 h in order to obtain random daily consumption patterns. This resulted in 12,500 daily consumption patterns (consumed amount of food during 1 day by an individual) for the total Dutch population, and 1,060 for young children. The consumption patterns of processed and compound food were transformed to amounts of primary food products by the RIKILT Conversion of Primary Agriculture Products program *(36).* Food consumption patterns of the Kempen population were assumed equal to those of the national population.

Monitoring data on cadmium levels in food products. For the baseline exposure scenario and for food products other than cattle organs and meat and the vegetables listed in Table 2, data on concentrations of cadmium levels in food products were obtained from the KAP database. The data originated from the Food and Consumer Product Safety Authority and the Institute for Marine Resources and Ecosystem Studies. The animal food products most sampled were kidneys and livers, because they are known to contain elevated cadmium concentration due the accumulation in these specific organs. However, the animal food product most consumed is meat, which is not regularly tested for cadmium presence. Hence, the cadmium levels in meat are estimated from the available data on kidneys and livers, as described earlier in the ''Materials and Methods'' section.

Modeling of human exposure. Human long-tem (chronic) dietary exposure to cadmium was estimated in a probabilistic way using the Monte Carlo risk assessment program MCRA, version 6 *(9).* MCRA simulates daily consumptions by sampling randomly drawn food consumption patterns from the food consumption database (Dutch National Food Consumption Survey) and combining these with random samples from the contamination database (KAP). The result is a full variability distribution of short-term intakes rather than a traditional deterministic point estimate of exposure. For chronic dietary exposure assessment, MCRA estimates the intake as the consumption on each day of each consumer multiplied by the average value of the compound cadmium concentration levels divided by body weight. With the Monte Carlo simulation, days of consumption are randomly sampled from the consumer database. Each time a consumption day is sampled, it contributes to the probability distribution of intakes (each individual contribution is a simulation). The total number of simulations was 100,000. Compound concentration data were presented as full data (as opposed to the use of mean, percentiles, or maxima) and presented by a nonparametric (empirical) distribution. In order to account for heterogeneity of variance (e.g., some individuals are more variable than others are with respect to their consumption patterns), the discrete and semiparametric method (Iowa State University Foods) was used *(11, 22),* with logarithmic transformation of intake data. The transformation to normality was improved by a spline fit to the transformed intakes. The transformed human daily intake was tested for normality with the Anderson-Darling test. The uncertainty (presented as 2.5 to 97.5% uncertainty percentiles) around the percentiles of the intake distribution was assessed by resampling consumption and concentration data sets with the bootstrap method (100 resampled sets, with each set resampled 10,000 times). The exposure was modeled for the total population (1 to 97 years, mean age of 36.3 years, mean weight of 62.8 kg, $n = 6{,}250$) and for children (1 to 6 years, mean age of 3.6 years, mean weight of 17.1 kg, $n = 530$).

Model validation. The predicted cadmium concentrations in the organs for each soil scenario were validated against monitoring data of cadmium levels in cattle organs from the Kempen area *(31, 33).* The predicted cadmium levels in vegetables were validated against contamination levels as measured during independent surveys (i.e., data other than those used to construct the predictive models) of vegetables produced in the Kempen area *(1, 32).* The level of human exposure as estimated for the different scenario's was validated with the dietary exposure to cadmium of the Kempen population, using MCRA, based on actual measured contamination levels of food products from the Kempen area *(1, 31, 33).*

RESULTS

Cadmium levels in cattle organs. The estimated cadmium levels in roughage ranged between 0.16 and 1.50 mg kg^{-1} of fresh weight (FW) for maize, and between 0.09 and 1.47 mg kg^{-1} of FW for grass, depending on the soil scenario. Only for the most extreme scenario did cadmium levels exceed the maximum tolerated level of 1.0 mg kg^{-1} of FW. Calculated cadmium levels in cattle kidneys and livers for the six different soil scenarios in the Kempen area ranged from 0.37 to 4.03 mg kg^{-1} of FW (average of 1.55 mg kg^{-1} of FW) for kidneys, and from 0.07 to 0.75 mg kg^{-1} of FW for livers (Table 3). The maximum tolerated levels for both organs were exceeded for soil scenarios E and F. The estimated levels were comparable to measured values obtained in 1988 and 2005, which ranged from 0.14 to 7.66 mg kg^{-1} of FW (average of 2.06) *(33)* and from 0.75 to 3.4 mg kg^{-1} of FW (average of 1.53) *(31)* for kidneys, respectively, and between 0.06 to 1.24 mg kg^{-1} of FW (average of 0.37) *(33)* for livers. Hence, the calculated values for the each of the six different soil scenarios can be considered realistic and suitable for further application.

Cadmium levels in vegetables. Estimated cadmium levels in 12 vegetable types for six different soil scenarios representative for the Kempen area are presented in Table 4. Most of the vegetable cadmium levels exceeded maximum tolerated levels for three out of the six scenarios (me-

TABLE 3. *Calculated cadmium levels in roughage, total cadmium intake by cattle, and final cadmium levels in the liver and kidneys of 6-year-old cattle for the six defined soil scenarios*

	Cd content roughage		Total Cd intake cow (0−6 yr)				Cadmium in organs		
Soil scenario	Maize (mg/kg)	Grass (mg/kg)	Maize (g/cow)	Grass (g/cow)	Compound feed (g/cow)	Soil (g/cow)	Avg content (mg/kg)	Kidney (mg/kg FW)	Liver (mg/kg FW)
Maximum	1.0[a]	1.0[a]						1.0[a]	0.5[a]
A	0.26	0.21	0.31	4.1	0.5	0.4	0.22	0.66	0.12
B	0.16	0.09	0.19	1.7	0.5	0.4	0.12	0.37	0.07
C	0.56	0.48	0.66	9.5	0.5	0.7	0.47	1.42[b]	0.26
D	0.34	0.20	0.41	4.0	0.5	0.7	0.25	0.75	0.14
E	1.50[b]	1.47[b]	1.77	29.2	0.5	1.9	1.35	4.03[b]	0.75[b]
F	0.92	0.61	1.11	12.2	0.5	1.9	0.69	2.05[b]	0.38[b]

[a] Maximum tolerated level.
[b] Calculated value exceeds the maximum tolerated level.

dium soil cadmium level combined with low pH, and both the high soil cadmium scenarios). Comparison of the predicted vegetable cadmium levels, and the median values of monitoring data from the Kempen *(1, 32)* were in close agreement (Table 4). Obviously, the predicted values from the worst-case scenarios (high cadmium, low pH: scenarios C and E) exceeded the measured median value. The measured data originate from well-maintained arable soils mostly, and the comparison of model predictions from scenario B or D (pH 5.5 soils) with the median values of measured cadmium levels in crops is therefore the most realistic. Apart from tomato and endive, the measured median values are close to the predicted levels.

Dietary exposure levels of the Dutch population to cadmium. For the total Dutch population (1 to 97 years), the transformed daily intake data showed no significant deviation from normality (Anderson-Darling test value of 0.32, $P = 0.75$). The day-to-day variability in cadmium intake was equal to the variability between individual consumers. The long-term median dietary exposure to cadmium for the total Dutch population, as calculated by the MCRA program, was 0.16 μg/kg of body weight (BW) per day (2.5 and 97.5% uncertainty bound of 0.15 and 0.17 μg/kg of BW per day, respectively) (Figs. 2 and 3A). This means that 50% of the total population had an intake equal or less than 0.16 μg/kg of BW per day (Fig. 2A). The uncertainty bounds around the calculated percentiles were small and not overlapping (Fig. 3A). Considering a Tolerable Daily Intake (TDI) level of 0.5 μg/kg of BW^{-1} day^{-1} *(2),* this means that the TDI was exceeded by approximately 0.25% of the total Dutch population (2.5 and 97.5% uncertainty bound of 0.20 and 0.70% of the population, respectively). The food groups contributing most to the cadmium intake included wheat (45%), potatoes (28%), and vegetables (20%) (Fig. 4). Within the group of vegetables, spinach (5.5%) and carrots (3.3%) especially contributed most to the predicted exposure. Meat products contributed approximately 4% (of which 2% was contributed by beef), fruits for 2%, seafood for 1%, and kidneys and livers for only 0.2%.

Also for children (1 to 6 years), the transformed daily intake data showed no significant deviation from normality (Anderson-Darling test value of 0.14, $P = 0.99$). In contrast to the total population, the within-individual variability in cadmium intake was 2.33 times larger than the between individual variability. The long-term median dietary exposure to cadmium for Dutch children of 1 to 6 years was 0.36 μg/kg BW per day (2.5 and 97.5% uncertainty bound of 0.33 and 0.38 μg/kg of BW per day, respectively) (Fig. 2B and 3B). Uncertainty of the percentile distribution was considerably larger compared with that of the 1- to 97-year-old population (Fig. 3B). The TDI (0.5 μg/kg of BW per day) was exceeded by approximately 10% of 1- to 6-year-old population (2.5 and 97.5% uncertainty bound of 6 and 18% of the population, respectively). As for the total population, the food groups that contributed most to the intake of children included wheat (47%), potatoes (27%), and vegetables (16%). Identical to the total population, within the group of vegetables, spinach (6.5%) and carrots (3.6%) were the most important contributors. Meat products contributed approximately 2.5% (of which beef contributed 1.6%) and fruits 2.9%.

Dietary exposure levels for different regional soil scenarios. With respect to the total Kempen population (1 to 97 years), the transformed daily intake data showed no significant deviation from normality for scenarios A, B, C, D, E, and F (Anderson-Darling test values of 0.42, 0.27, 0.24, 0.49, 0.21, and 0.55, respectively; *P* values of 0.42, 0.75, 0.90, 0.25, 0.90, and 0.25, respectively). For all scenarios, the within-individual variation in cadmium intake was higher than the between-individual variation (ratios of 2.20, 1.58, 2.57, 1.96, 3.45, and 2.69, respectively, for each of the different soil scenarios). The long-term median dietary exposure to cadmium for the Kempen population of 1 to 97 years for the different soil scenarios was 0.33, 0.24, 0.50, 0.36, 0.98, and 0.63 μg/kg of BW per day, respectively (Fig. 2A). The TDI (0.5 μg/kg of BW per day) was exceeded by approximately 10, 3, 50, 25, 80, and 60% of the Kempen population for the soil scenarios A to F. With an increase in the degree of soil pollution, the relative contribution of potatoes and vegetables to the total intake of cadmium increased, whereas that of wheat decreased (Fig. 4).

With respect to the young children (1 to 6 years old),

TABLE 4. *Calculated cadmium levels in vegetables for the six different soil scenarios, the maximum tolerated level for each vegetables as set by the European Union (12) and the median of measured cadmium levels in vegetable produced in the Kempen area, according to the meta-analyses of Smolders et. al. (32)*

Soil scenario	Cd levels in vegetable (in mg/kg FW)											
	Potato	Endive	Leek	Lettuce	Spinach	Tomato	Carrots	Cucumber	French beans	Scorzonera	Celery	Radish
Maximum[a]	0.10	0.20	0.10	0.20	0.20	0.05	0.10	0.05	0.05	0.10	0.10	0.10
A	0.06	0.24[b]	0.09	0.17	0.17	0.05	0.11[b]	0.00	0.00	0.31[b]	0.16[b]	0.09
B	0.04	0.13	0.05	0.08	0.09	0.03	0.07	0.00	0.00	0.11[b]	0.10	0.07
C	0.11[b]	0.37[b]	0.24[b]	0.31[b]	0.29[b]	0.07[b]	0.14[b]	0.00	0.01	0.44[b]	0.25[b]	0.14[b]
D	0.07	0.19	0.14[b]	0.14	0.15	0.05	0.09	0.00	0.00	0.16[b]	0.15[b]	0.11[b]
E	0.23[b]	0.62[b]	0.87[b]	0.67[b]	0.59[b]	0.12[b]	0.18[b]	0.01	0.03	0.69[b]	0.45[b]	0.26[b]
F	0.14[b]	0.33[b]	0.50[b]	0.31[b]	0.30[b]	0.07[b]	0.12[b]	0.01	0.01	0.25[b]	0.28[b]	0.21[b]
Median measured	0.03	0.06	0.06	0.10	0.14	0.01	0.08	0.02	0.01	0.18	0.27	NA[c]

[a] Maximum tolerated level.
[b] Calculated value exceeds the maximum tolerated level.
[c] NA, not analyzed.

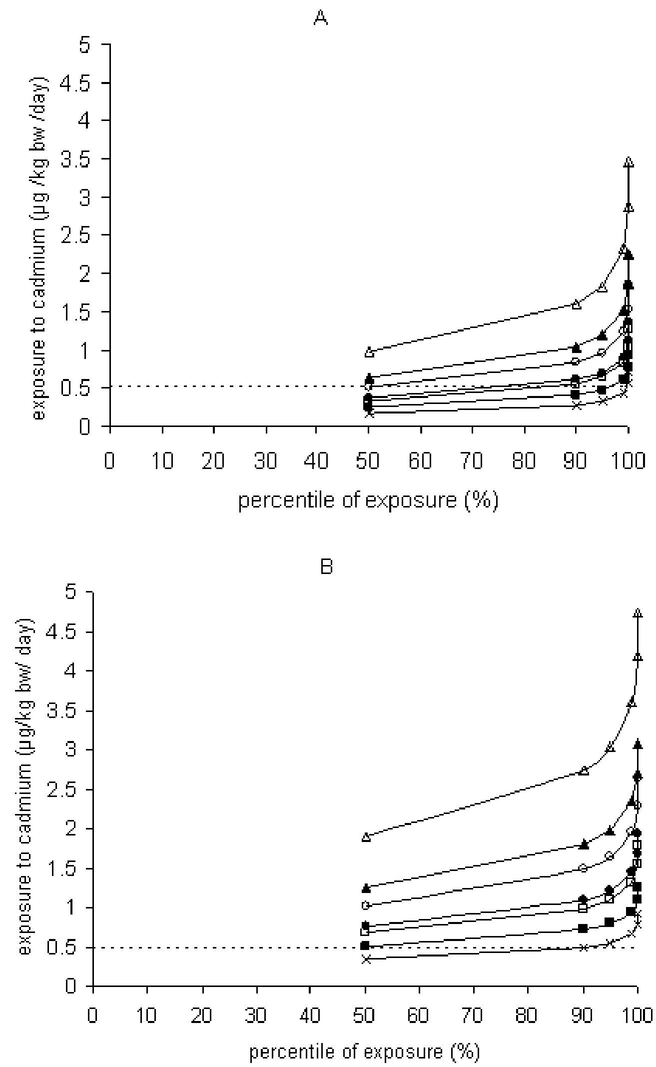

FIGURE 2. *Percentiles of chronic exposure to cadmium in (A) the total population (1 to 97 years) and (B) young children (1 to 6 years) for different scenarios: baseline exposure (×) and Kempen soil scenarios A (□), B (■), C (○), D (●), E (△), and F (▲).*

the transformed daily intake data showed no significant deviation from normality for scenarios A, B, C, D, E and F (Anderson-Darling test values of 0.50, 0.21, 0.12, 0.16, 0.47, and 3.2, respectively; *P* values of 0.99, 0.90, 0.99, 0.99, 0.25, and 0.60, respectively). For all scenarios, the within-individual variation in cadmium intake was higher than the between individual variation (ratios of 4.21, 3.20, 4.73, 3.62, 6.11, and 4.89, respectively for soil scenarios A to F). The long-term median dietary exposure to cadmium for the young children in the Kempen for the soil scenarios A to F were 0.68, 0.51, 1.02, 0.76, 1.9, and 1.26 µg/kg of BW per day, respectively (Fig. 2B). The TDI (0.5 µg/kg of BW per day) was exceeded by approximately 70, 50, 90, 80, 100, and 100% of the young children in the Kempen area. With an increase in the degree of soil pollution, the relative contribution of potatoes to the total cadmium intake increased, that of vegetables was stable, whereas that of wheat decreased.

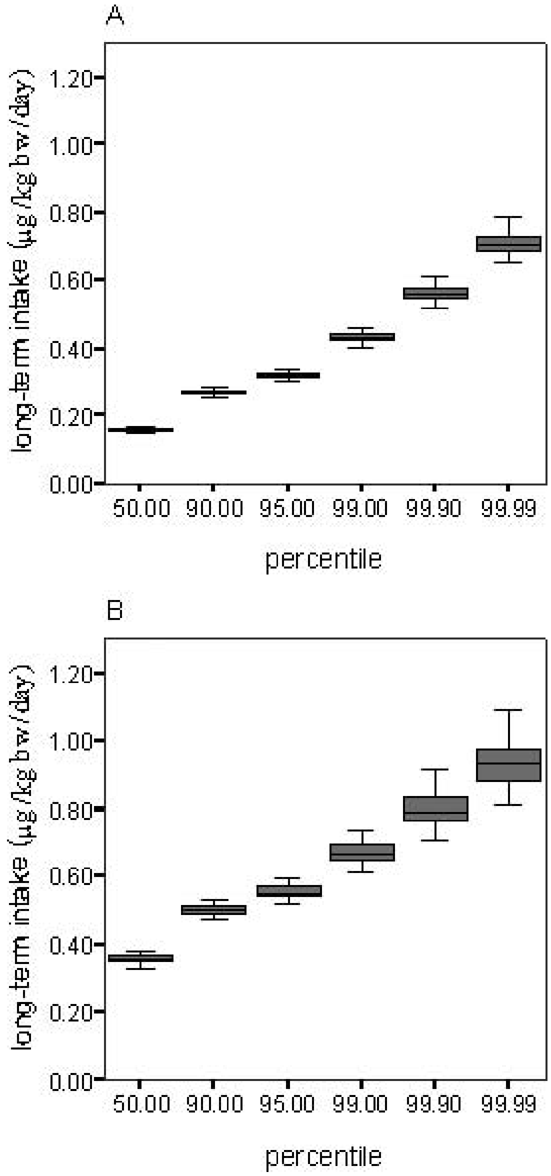

FIGURE 3. *Box plot representing the uncertainty of the estimated percentiles distribution of exposure for long-term human dietary cadmium exposure. (A) Total Dutch population 1 to 97 years. (B) Total Dutch population children 1 to 6 years.*

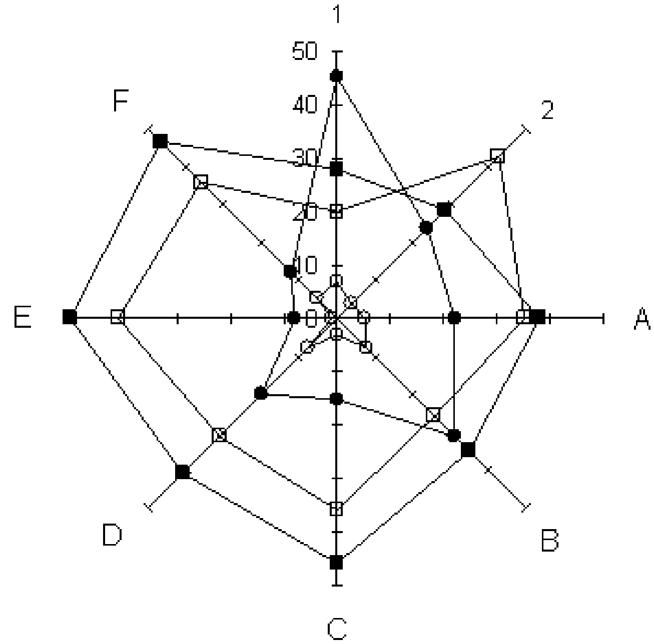

FIGURE 4. *Contribution of the most important food groups (■, potatoes; ●, wheat; □, vegetables; ○, rest) to the dietary exposure to cadmium for the total Dutch population (1 to 97 years), based on monitoring data (1); the Kempen inhabitants (1 to 97 years), based on measured contamination data on Kempen food products (12); and for Kempen inhabitants (1 to 97 years), according to soil scenarios A, B, C, D, E, and F (here presented as 3 to 8).*

Validation of the scenario exposure assessment. As a validation for the level of human exposure as calculated for the different soil scenarios (using the predictive models), the dietary exposure to cadmium of the Kempen population was estimated using measured contamination levels of food products from the Kempen area. The transformed human daily intake showed no significant deviation from normality (Anderson-Darling value of 3.33, P value of 0.45), and the ratio between within individual variation and between individual variation was 3.74. The long-term median dietary exposure to cadmium was 0.28 μg/kg of BW per day for the 1- to 97-year-old population and 0.58 μg/kg of BW per day for the 1- to 6-year-old population. The TDI (0.5 μg/kg of BW per day) was exceeded by approximately 10% of the 1- to 97-year-old population and 45% of the 1- to 6-year-old population. The food groups contributing most to the cadmium intake were vegetables, potatoes, and wheat (respectively, 39, 29, and 26% for the 1- to 97-year-old population [Fig. 4], and 43, 29, and 24% for the 1- to 6-year-old population). The exposure values calculated fall within the range of the calculated exposure for the scenarios (Fig. 5).

DISCUSSION

The presence of heavy metals like cadmium in soils used for agriculture poses a potential public health risk due to the accumulation in food and feed. The soil is increasingly recognized as an essential part of a systems approach to achieve the delivery of safe foods to the consumer. Regional differences in soil cadmium levels and soil characteristics may lead to differences in human dietary exposure to cadmium. The objective of this study was to develop a model that links soil cadmium levels and soil characteristics to human dietary exposure. Cadmium levels in vegetables, roughage, feed and cattle organs were estimated by empirically derived predictive models based on soil cadmium levels and soil characteristics. Applying Monte Carlo simulations, these contamination data were combined with food consumption patterns in order to estimate human dietary exposure to cadmium. The chain of models was applied to six different soil scenarios, varying in soil cadmium level and pH, and reflecting the range of soil conditions to be found in the cadmium polluted Kempen area.

The estimated cadmium concentrations in vegetables and cattle organs showed acceptable agreement with measured levels in food products sampled in the Kempen area. The median of the measured values for potato, endive, leek,

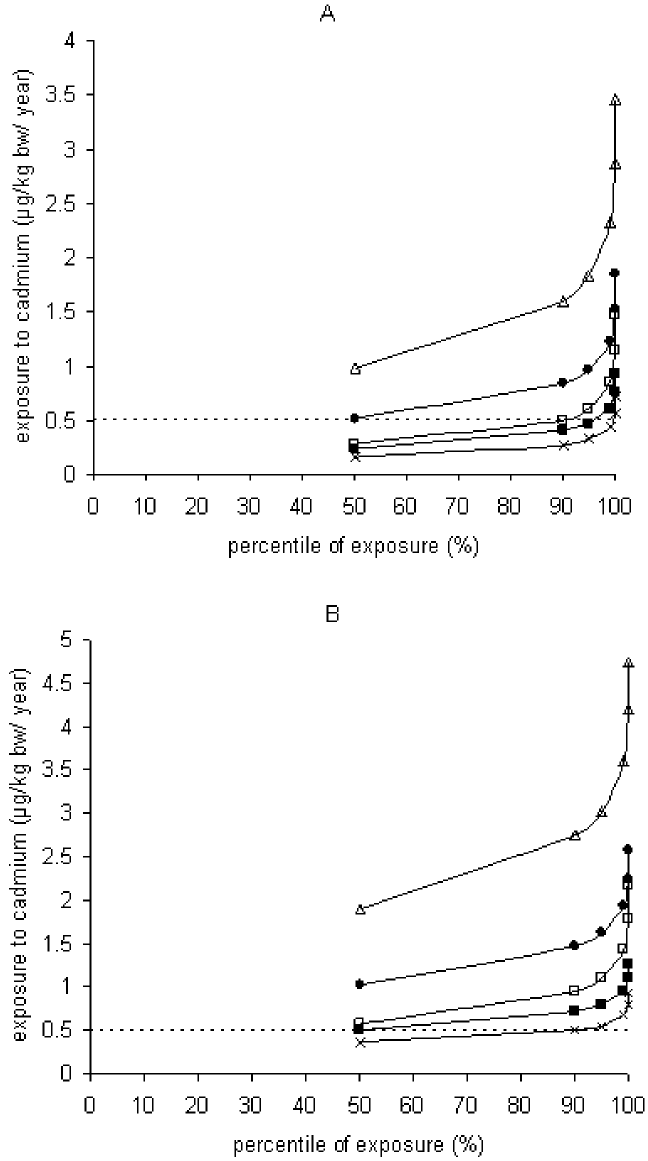

FIGURE 5. *Percentiles of chronic exposure to cadmium in (A) the total population (1 to 97 years) and (B) young children (1–6 years) for the baseline exposure, based on national monitoring data (×); exposure of Kempen inhabitants as calculated by soil scenario B (lowest exposure of all scenario) (■); exposure of Kempen inhabitants, based on measured contamination data from Kempen food products (validation) (□); exposure of Kempen inhabitants, according to the average calculated scenario values (●); and exposure of Kempen inhabitants, as calculated by soil scenario E (highest exposure of all scenarios) (△).*

lettuce, spinach, tomato, carrots, cucumber, French beans, and scorzonera were close to the predicted values of soil scenario A (0.5 mg cadmium kg^{-1} soil and pH 4.5) and B (0.5 mg cadmium kg^{-1} soil and pH 5.5) *(1)*. For endive and tomato, the cadmium levels seem to be either overestimated by the predictive models, or underreported by the surveys. However, maximum reported measured values are in range with the predicted values (0.21 mg kg^{-1} for endive and 0.03 mg kg^{-1} for tomato). From the predictive models, it can be concluded that with soil, cadmium levels of 1 mg kg^{-1} (which is the maximum tolerable level in soil), and a pH below 5.5, most vegetables of concern will exceed present food quality standards set by the European Union *(1)*. The predicted cadmium concentrations in cattle organs also were in line with measured concentrations from cattle raised in the Kempen area *(31, 33)*. These agreements between measured and predicted cadmium concentrations justify the use of the predictive models to estimate cadmium concentrations in vegetables.

The median long-term dietary exposure to cadmium in The Netherlands was estimated at 0.16 μg/kg of BW per day for the total Dutch population and at 0.36 μg/kg of BW per day for children (1 to 6 years). The levels of cadmium exposure for the Dutch population are comparable with other European countries *(15, 18, 29, 42)*. It was estimated that 0.25% of the total Dutch population would exceed the TDI level of 0.5 μg/kg of BW per day, which is roughly equal to 40,000 consumers. For children (1 to 6 years), this was estimated to be 10%, which is considerably higher. However, it should be noted that the uncertainty bounds were relatively large (6 to 18%).

The chain model presented with this study clearly shows that the effects of different regional soil scenarios (cadmium levels and soil characteristics) on human dietary exposure are large. It was estimated that, depending on the scenario selected, 3 to 80% of the Kempen population will exceed the TDI. A comparison of the results obtained here with a recent risk assessment study for the Kempen area *(23)* shows that the average exposures are in good agreement. Median exposure levels of 0.18 μg/kg of BW per day for adults to 0.41 μg/kg of BW per day for children were obtained based on fixed levels of cadmium in food products *(23)*. This is close to the median exposure levels of 0.24 μg/kg of BW per day for adults and 0.51 μg/kg of BW per day for children calculated in scenario B. Results for scenario B can be considered representative for the entire Kempen, whereas those of scenario D can be considered as the realistic upper boundary. Little or no arable crops are grown on soils with a pH at or below 4.5 or cadmium levels far beyond 1 mg kg^{-1} in the soil. The results from these scenarios (C, E, and F) nevertheless clearly indicate that, indeed, exposure levels may reach unacceptable levels under certain circumstances. In the study by Oomen et al. *(23)*, the impact of soil quality on exposure was not accounted for, and exposure was based on average data on the quality of food and animal products. This study clearly shows that differences in soil acidity and contamination levels, and the interaction between both leads to rather elevated levels of human exposure.

It should be noted that it was assumed that for the vegetables listed in Table 2, only locally produced vegetables were consumed, which can be considered a worse-case scenario. The same applies for the validation exposure assessment. The exposure assessment based on measured contamination data from vegetables produced in the Kempen area resulted in values that fall between the exposure estimates of soil scenarios 1 and 2 (cadmium level of 0.5 mg kg^{-1}, pH values of 4.5 and 5.5), indicating the validity of the predictive chain model. Furthermore, the cadmium bioavailability from the various products may be different depending on the nature of the cadmium present in the product.

In the present study, a chain modeling approach in order to estimate human dietary exposure to cadmium is presented. With this approach, cadmium levels in vegetables and cattle organs are estimated using predictive models. Subsequently, these contamination levels are combined with consumption patterns to get to an estimate of human exposure, using Monte Carlo simulations. Validation of the models showed that the results based on predicted contamination levels reflect the results based on measured data. The calculated median exposure levels are in good agreement with existing estimates. The applicability of a food production chain model to estimate human exposure is demonstrated successfully. To our knowledge, this is the first attempt to link soil quality directly to human contaminant exposure, using intermediate models of soil–plant and feed–animal products relationships. This enables the user to assess whether or not a specific combination of soil properties, soil acidity, and land use lead to unacceptable levels of human exposure. As such, the results indicate for the Dutch Kempen area that exposure due to consumption of food from areas with cadmium levels below 1 mg kg^{-1} and well-maintained arable soils remains below the TDI. The advantage of the model presented here is that it can be used for a relatively fast evaluation of cadmium exposure for specific regions, without the necessity of sampling large amounts of local food products differing in soil cadmium levels and soil characteristics. Since the concept used here has a modular structure, it is rather easy to adapt certain model parts, like the transfer of metals from soils to crops to be used in different regions, for different crops or different contaminants not included here.

ACKNOWLEDGMENTS

The authors acknowledge the contribution of Kees Kan (Animal Science Group, Wageningen University) for his contribution to the consumption patterns of cattle. This study was (partly) financed by INTERREG IIIC, the Dutch Ministry of Agriculture, Nature and Food Quality, and foundation Actief Bodembeheer de Kempen. We thank Ron Hoogeboom (RIKILT) for his critical review of the manuscript.

REFERENCES

1. Anonymous. 2005. Cadmium en lood in landbouwgewassen in de kempen, najaar 2005. Report of the Food and Consumer Product Safety Authority. Available at: http://www.vwa.nl. Accessed 25 July 2008.
2. Baars, A. J., R. M. C. Theelen, P. J. C. M. Janssen, J. M. Hesse, M. E. v. Apeldoorn, M. C. M. Meijerink, L. Verdam, and M. J. Zeilmaker. 2001. Re-evaluation of human-toxicological maximum permissible risk levels. RIVM report 711701025. Available at: http://www.rivm.nl/bibliotheek/rapporten/711701025.html. Accessed 25 July 2008.
3. Baxter, J. C., B. Barry, D. E. Johnson, and E. W. Kienholz. 1982. Heavy metal retention in cattle tissues from ingestion of sewage sludge. *J. Environ. Qual.* 11:616–620.
4. Beresford, N. A., R. W. Mayes, N. M. J. Crout, P. J. Maceachern, B. A. Dodd, C. L. Barnett, and C. S. Lamb. 1999. Transfer of cadmium and mercury to sheep tissues. *Environ. Sci. Technol.* 33:2395–2402.
5. Brus, D. J., J. J. De Gruijter, D. J. J. Walvoort, F. De Vries, J. J. B. Bronswijk, P. F. A. M. Römkens, and W. De Vries. 2002. Heavy metals in the environment: mapping the probability of exceeding critical thresholds for cadmium concentrations in soils in The Netherlands. *J. Environ. Qual.* 31:1875–1884.
6. Brussaard, J. H., W. V. Dokkum, C. G. Van der Paauw, R. H. De Vos, W. L. De Kort, and M. R. Löwik. 1996. Dietary intake of food contaminants in The Netherlands (Dutch Nutrition Surveillance System). *Food Addit. Contam.* 13:561–573.
7. Copius Peereboom-Stegeman, J. H. J., and J. W. Copius Peereboom. 1989. The intake of cadmium in the Kempen, an area in the south of The Netherlands. *Ecotoxicol. Environ. Saf.* 18:93–108.
8. Crout, N. M. J., N. A. Beresford, J. M. Dawson, J. Soar, and R. W. Mayes. 2004. The transfer of 73As, 109Cd and 203Hg to the milk and tissues of dairy cattle. *J. Agric. Sci.* 142:203–212.
9. de Boer, W. J., and H. van der Voet. 2007. MCRA, release 6, a web-based program for Monte Carlo risk assessment. Available at: http://mcra.rikilt.wur.nl. Accessed 25 July 2008.
10. de Vries, W., P. F. A. M. Römkens, and G. Schütze. 2007. Critical soil concentrations of cadmium, lead and mercury in view of health effects on humans and animals. *Rev. Environ. Contam. Toxicol.* 191:91–130.
11. Dodd, K. W. 1996. A technical guide to C-SIDE. Technical report 96-TR 32. Department of Statistics and Center for Agricultural and Rural Development, Iowa State University, Des Moines.
12. European Commission. 2001. Commission regulation (EC) no 466/2001 of 8 March 2001: setting maximum levels for certain contaminants in foodstuffs. *Off. J. Eur. Communities* 77:1–13.
13. Farmer, A. A., and A. M. Farmer. 2000. Concentrations of cadmium, lead and zinc in livestock feed and organs around a metal production centre in eastern Kazakhstan. *Sci. Total Environ.* 257:53–60.
14. Fries, G. F., G. S. Marrow, and P. A. Snow. 1982. Soil ingestion by dairy cattle. *J. Dairy Sci.* 65:611–618.
15. Jarup, L., M. Berglund, C. G. Elinder, G. Nordberg, and M. Vahter. 1998. Health effects of cadmium exposure—a review of the literature and a risk estimate. *Scand. J. Work Environ. Health* 24:1–51.
16. Kans, K. 2007. Personal communication.
17. Kistemaker, C., M. Bouman, and K. F. A. M. Hulshof. 1998. Consumption of separate products by Dutch population groups—Dutch National Food Consumption Survey 1997–1998 (in Dutch). TNO (Nutrition and Food Research Institute). Zeist, The Netherlands.
18. Larsen, E. H., N. L. Andersen, A. Møller, A. Petersen, G. K. Mortensen, and J. Petersen. 2002. Monitoring the content and intake of trace elements from food in Denmark. *Food Addit. Contam.* 19:33–46.
19. McLaughlin, M. J., D. R. Parker, and J. M. Clarke. 1999. Metals and micronutrients—food safety issues. *Field Crops Res.* 60:143–163.
20. Miranda, M., M. L. Alonso, C. Castillo, J. Hernandez, and J. L. Benedito. 2001. Cadmium levels in liver, kidney and meat in calves from Asturias (North Spain). *Eur. Food Res. Technol.* 212:426–430.
21. Nordberg, G. F., and T. Kjellstrom. 1979. Metabolic model for cadmium in man. *Environ. Health Perspect.* 28:211–217.
22. Nusser, S. M., A. L. Carriquiry, K. W. Dodd, and W. A. Fuller. 1996. A semiparametric transformation approach to estimating usual daily intake distributions. *J. Am. Stat. Assoc.* 91:1440–1449.
23. Oomen, A. G., P. J. C. M. Janssen, J. C. H. van Eijkeren, M. I. Bakker, and A. J. Baars. 2007. Cadmium in the Dutch Kempen area: an integrated risk assessment. RIVM report 320007001/2007. RIVM (National Institute for Public Health and the Environment), Bilthoven, The Netherlands. (In Dutch with English summary.)
24. Peijnenburg, W., R. Baerselman, A. De Groot, T. Jager, D. Leenders, L. Posthuma, and R. Van Veen. 2000. Quantification of metal bioavailability for lettuce (*Lactuca sativa* L.) in field soils. *Arch. Environ. Contam. Toxicol.* 39:420–430.
25. Römkens, P. F. A. M., J. E. Groenenberg, R. P. J. J. Rietra, J. E. Groenenberg, and W. de Vries. 2007. Onderbouwing LAC2006-waarden en overzicht van bodem-plant relaties ten behoeve van de Risicotoolbox; een overzicht van gebruikte data en toegepaste methoden. Alterra-rapport 1442. Alterra Wageningen, The Netherlands.
26. Römkens, P. F. A. M., G. W. Schuur, J. P. A. Lijzen, R. P. J. J. Rietra, and E. M. Dirven-van Breemen. 2005. Risico's van cadmium en lood

in moestuinen in de Kempen. Alterra-rapport 1129. Alterra Wageningen, The Netherlands.

27. Römkens, P. F. A. M., M. J. Zeilmaker, R. P. J. J. Rietra, C. A. Kan, J. C. H. van Eijkeren, L. W. D. van Raamsdonk, and J. P. A. Lijzen. 2007. Blootstelling en opname van cadmium door runderen in de Kempen: een modelstudie. Alterra-rapport 1438. Alterra Wageningen, The Netherlands.
28. Schrey, P., J. Wittsiepe, U. Budde, B. Heinzow, H. Idel, and M. Wilhelm. 2000. Dietary intake of lead, cadmium, copper and zinc by children from the German North Sea island Amrum. *Int. J. Hyg. Environ. Health* 203:1–9.
29. Seifert, M., and M. Anke. 1999. Daily intake of cadmium in Germany in 1996 determined with the duplicate portion technique. *J. Trace Microprobe Tech.* 17:101–109.
30. Sharma, R. P., J. C. Street, J. L. Shupe, and D. R. Bourcier. 1982. Accumulation and depletion of cadmium and lead in tissues and milk of lactating cows fed small amounts of these metals. *J. Dairy Sci.* 65:972–979.
31. Slob, W. 2005. Advies inzake de relatie tussen de herkomst van runderen en het gehalte cadmium in hun nieren. RIVM-RIKILT Front Office, Wageningen, The Netherlands.
32. Smolders, E., G. Jansson, L. Van Laer, A. Ruttens, J. Vangronsveld, P. Römkens, L. De Temmerman, N. Waegeneers, and J. Bries. 2007. Teeltadvies voor de landbouw in kader van het Interreg project BeNeKempen. Internal report OVAM. Mechelen, Belgium.
33. Spierenburg, T. J., G. J. De Graaf, A. J. Baars, D. H. J. Brus, M. J. M. Tielen, and B. J. Arts. 1988. Cadmium, zinc, lead, and copper in livers and kidneys of cattle in the neighbourhood of zinc refineries. *Environ. Monit. Assess.* 11:107–114.
34. Thornton, I., and P. Abrahams. 1983. Soil ingestion—a major pathway of heavy metals into livestock grazing contaminated land. *Sci. Total Environ.* 28:287–294.
35. Van-Camp, L., B. Bujarrabal, A.-R. Gentile, R. J. A. Jones, L. Montanarella, C. Olazabal, and S.-K. Selvaradjou (ed.). 2004. Reports of the Technical Working Groups established under the Thematic Strategy for Soil Protection, Volume III, Organic Matter. European Commission, Director General, Joint Research Centre, European Environmental Agency, Ispra-Copenhagen EUR 21319 EN/3. Office for Official Publications of the European Communities, Luxembourg.
36. van Dooren, M. M. H., I. Boeijen, J. D. v. Klaveren, and G. v. Donkersgoed. 1995. Conversie van consumeerbare voedingsmiddelen naar primaire agrarische produkten [Conversion of consumed foods into raw agricultural commodities]. RIKILT report 95.17. RIKILT-DLO, Wageningen, The Netherlands.
37. van Hooft, W. F. 1995. Risico's voor de volksgezondheid als gevolg van blootstelling van runderen aan sporenelementen bij beweiding. Rapport nr. 693810 001. Rijksinstituut voor Volksgezondheid en Milieuhygiëne, Bilthoven, The Netherlands.
38. van Klaveren, J. D. 1999. Residue monitoring in The Netherlands 1998—Program for the Quality of Agricultural Products. Available at: http://www2.rikilt.dlo.nl/kap/index.html. Accessed 25 July 2008.
39. van Raamsdonk, L. W. D., G. A. L. Meijer, H. Bouwmeester, C. A. Kan, W. Beek, and P. A. Kemme. 2007. Schadelijke stoffen in de dierlijke productieketen, niveasu van voorkomen, normen en overdracht. Achtergrond en input voor ketenmodellering. RIKILT rapport 2007.011. RIKILT, Wageningen, The Netherlands.
40. van Raamsdonk, L. W. D., G. A. L. Meijer, C. A. Kan, H. Bouwmeester, H. van der Voet, P. Adamse. W. Beek, and P. A. Kemme. 2007. Schadelijke stoffen in de dierlijke productieketen: overdracht gemodelleerd in ketenperspectief. RIKILT rapport 2007.009. RIKILT, Wageningen, The Netherlands.
41. Wang, G., M. Y. Su, Y. H. Chen, F. F. Lin, D. Luo, and S. F. Gao. 2006. Transfer characteristics of cadmium and lead from soil to the edible parts of six vegetable species in southeastern China. *Environ. Pollut.* 144:127–135.
42. Wilhelm, M., J. Wittsiepe, P. Schrey, U. Budde, and H. Idel. 2002. Dietary intake of cadmium by children and adults from Germany using duplicate portion sampling. *Sci. Total Environ.* 285:11–19.
43. Wilhelm, M., J. Wittsiepe, P. Schrey, A. Hilbig, and M. Kersting. 2005. Consumption of homegrown products does not increase dietary intake of arsenic, cadmium, lead, and mercury by young children living in an industrialized area of Germany. *Sci. Total Environ.* 343:61–70.
44. Winter-Sorkina, R. d., G. v. Donkersgoed, M. I. Bakker, and J. D. v. Klaveren. 2003. Dietary intake of heavy metals (cadmium, lead and mercury) by the Dutch population. RIVM report 320103001/2003. RIVM and RIKILT, Wageningen, The Netherlands.