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Cadmium in the food chain near non-ferrous metal production sites

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Abstract
Dietary cadmium (Cd) exposure was estimated for adults living in Cd-contaminated areas close to non-ferrous metal plants and compared with dietary Cd exposure in the general Belgian adult population. To evaluate the contamination levels of locally produced food items, 35 fruit samples, 97 vegetable samples, 98 samples of potatoes and 53 samples of meat, liver and kidney of cattle, which had resided for more than 18 months in the contaminated area, were analyzed for Cd. Mean Cd concentrations in fruit and vegetables were 1.1- to 9-fold higher than in samples from other regions at ambient Cd levels. Mean Cd concentrations in bovine meat, liver and kidney were 2-fold higher compared to samples from animals in other regions of Belgium. The estimated dietary intake was 31.3 and 63.3 μg day^{-1} for average and large consumers, respectively, in the contaminated area, compared to 17 and 38.3 μg day^{-1}, respectively, for the general adult population. Excessive consumption of locally produced food items in areas close to non-ferrous metal plants could result in Cd intake levels exceeding the provisional tolerable weekly intake (PTWI).

Keywords: *Cadmium, dietary intake, risk assessment, pollution, cattle, vegetables*

Introduction

The level of cadmium reflects the level of industrial pollution in the local environment (Saegerman et al. 2006). In previous decades, numerous non-ferrous metal production plants were constructed in industrialized countries. In Belgium, many industrial sites were developed for the production and treatment of non-ferrous metals, with several zinc and copper smelters being built in the northern part of the country (Campine region). An adverse consequence of these activities is the mainly historical, local emission into air, water and soil of large amounts of contaminants, such as cadmium (Cd), of which Belgium was one of the principal producers in Europe (Monography 2004). Cadmium can also occur naturally in the environment from the gradual erosion and abrasion of rocks and soils or from singular events, such as forest fires and volcanic eruptions. It is, therefore, naturally present in air, water, soils and foodstuffs (http://www.cadmium.org/introduction.html). Over the last decades, considerable attention has focused on the toxicological effects of contamination of the local environment by heavy metals (Marshall and Mellinger 1980; Mench et al. 1989; Saegerman et al. 2006). As a consequence, progress has been made in controlling emissions of these toxic elements by industry and significant efforts were (and are being) devoted to remedial action. Remedial action mainly involves the site itself and nearby private gardens where high metal concentrations have been measured. Less attention has been paid to areas a few kilometers away from the industrial sites where lower but increased metal levels have been detected in several

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environmental compartments (IHE 1986; Staessen et al. 1994). These areas are typically situated within a few tens of kilometers radius of the industrial sites and may pose a contamination risk for the food chain and, consequently, human health. Residents of six contaminated districts close to three smelters in the Campine region had higher urinary Cd excretion, a biomarker of lifetime exposure, than residents from four other districts (Staessen et al. 1994; 1999). Furthermore, residents in the more contaminated districts had increased risk of bone fractures (Staessen et al. 1999). Recently, it was found that overall cancer risk was significantly associated with a doubling of 24-h cadmium excretion (Nawrot et al. 2006). In the 10 districts surveyed by Staessen et al. (1994), Cd in the soil was positively correlated with Cd in vegetables and residents' urine.

The major routes of Cd transfer to humans via the food chain are illustrated in Figure 1. Soils are contaminated by Cd through wet and dry deposition of Cd emissions. Contaminated soil, in turn, can be the origin of groundwater pollution (up to 400 $\mu g\,l^{-1}$ in wells located in the Campine region; IHE 1986) and of contamination of locally grown crops (fodder crops, such as maize and grass, as well as vegetables, such as potatoes, carrots and cabbages). The fodder crops are fed to cattle, which accumulate Cd in specific organs, such as kidneys and liver (López Alonso et al. 2000). At the top of the food chain, the consumer of locally produced foodstuffs in particular will be exposed principally via the consumption of animal products (especially the organs) and vegetables (mostly broad-leaf and root vegetables).

The objective of this study was to estimate human Cd exposure through the food chain in areas of northern Belgium close to non-ferrous metal plants. Cd concentrations were recorded in vegetables and animal products from these areas and other regions in Belgium. Dietary Cd intake was calculated for consumers of these locally produced food items and compared to the dietary intake of the general adult population in Belgium.

Materials and methods

Sampling

The contaminated area is situated in the north of Belgium, around several non-ferrous metal production sites with known Cd emissions. The surface area contaminated with Cd is about 1200 km^2 or 4% of the total Belgian surface area.

Vegetable and animal food products were sampled in the period 2004–2005 by the Belgian Federal Agency for the Safety of the Food Chain (FASFC), both in contaminated and "uncontaminated" areas

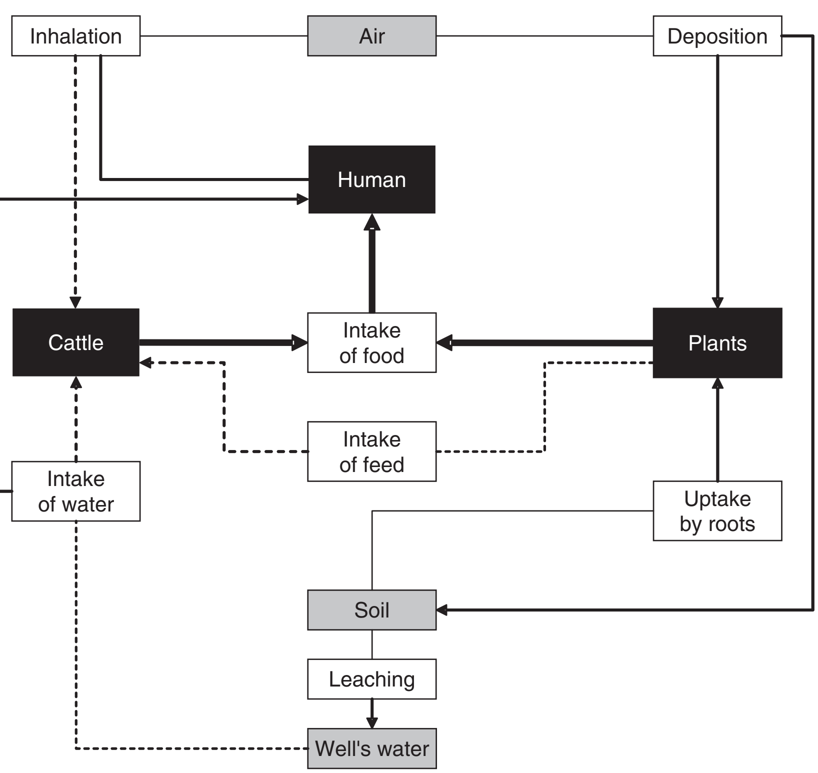

Figure 1. Main routes for entry of cadmium into the food chain and uptake by humans. (----: Contamination of cattle from the environment; ——: Contamination of humans through food consumption).

of Belgium, i.e. areas at ambient environmental Cd levels. The number of samples taken by the FASFC was determined in the official control program. The methodology developed for the program is based on risk evaluation, statistical tools and current scientific knowledge (Maudoux et al. 2006). Vegetable food products included strawberries, berries, cabbages, beans, asparagus, carrots, scorzonera, celery, leek and potatoes. Field sampling of the fruit and vegetable crops was carried out just before harvest. Samples were taken close to the emission sources and at increasing distances (within a radius from 4 up to 10 km). Animal food products (kidney, liver and meat) were sampled at random in slaughterhouses by FASFC in 2005. Tissues were sampled from animals that had resided more than 18 months in the contaminated areas and animals from areas of ambient environmental Cd levels.

Analytical methods

Vegetable samples were ground before analysis and only edible parts were used. Potato tubers, carrots and scorzonera roots were washed thoroughly and peeled. The analytical method for vegetable samples is derived from EN 14084:2002 (Foodstuffs. Determination of trace elements. Determination of lead, cadmium, zinc and iron by atomic absorption spectrometry (AAS) after microwave digestion). Homogenized samples were microwave-digested with nitric acid and hydrogen peroxide and the concentration of cadmium quantified by means of an AAS graphite furnace. Each batch of eight samples in duplicate included two procedural blanks and at least two certified reference materials. In the case of deviation from a set of common criteria for the blank and reference material values, analysis of the batch was repeated.

The limit of quantification (LOQ) for Cd was derived from inter-laboratory reproducibility tests on actual samples with very low concentrations of cadmium and calculated as $LOQ = \sqrt{2}.6.s_R$, where s_R represents the intra-reproducible standard deviation (VITO 2005) and equals $0.01\,mg\,kg^{-1}$. Analytical results below the LOQ were (reported <LOQ) set equal to the LOQ/2 for data analysis purposes (half weight bound principle). Analyses were carry out in the laboratory of the FASFC at Gentbrugge.

Animal tissues were stored in a freezer until analysis. The tissues were mixed in a domestic food-mixer to homogenize the samples. Consequently, the kidney cortex and medulla, which contain unequal trace element concentrations, were homogeneously mixed and the Cd concentration expressed for the whole organ. For each tissue, two homogenized subsamples were microwave-digested with nitric acid at 180°C and Cd concentrations in all samples were quantified by inductively coupled plasma–mass spectrometry (ICP–MS; VG PQ-ExCell, TJA, US). Each batch of 20 samples additionally included one procedural blank, one certified reference material (IAEA 407) and one laboratory reference material. As above, in the case of deviation, analysis was repeated. The limit of quantification (LOQ) for Cd was calculated as 10 times the standard deviation of 10 procedure blanks multiplied by the dilution factor and equals $0.002\,mg\,kg^{-1}$. Analytical results below the LOQ were set equal to LOQ/2 for data analysis purposes (half weight bound principle). All Cd concentrations are expressed on a fresh weight basis.

Estimation of cadmium intake

Average and large Cd intake were calculated both for adults living in contaminated areas (local adult population in a contaminated area) and those in areas of ambient environmental Cd levels (general adult population). Differences in Cd intake between adults from both areas are based on differences in measured Cd concentrations in locally produced food items. Differences between average and large Cd intake are based on differences in daily food consumption amounts.

Food products were classified in 13 major food groups (Table I). Vegetable subdivisions conformed to definitions for the European maximum levels settings (European Commission 2001). These subgroups are defined as: (i) vegetables, excluding leafy vegetables, fresh herbs, all fungi, stem vegetables, root vegetables and potatoes, (ii) stem vegetables and root vegetables, excluding celeriac, and (iii) leafy vegetables, fresh herbs, celeriac and all cultivated fungi. The European maximum levels of Cd for the respective categories are (i) $0.05\,mg\,kg^{-1}$, (ii) $0.1\,mg\,kg^{-1}$, and (iii) $0.2\,mg\,kg^{-1}$. Daily consumption of each of the major food groups and subgroups was estimated for adults between the ages of 19 and 59 years. Several food consumption models are available and a Belgian food consumption survey was carried out in 2004 (Devriese et al. 2006). The survey, based on food frequency questionnaires and two 24-h dietary recall inquiries of more than 3200 persons, reports the median and 25th, 50th, 75th and 97.5th percentile consumption of major food groups. The results are not sufficiently detailed, however, to differentiate consumption within the food groups outlined in Table I. The UK Pesticides Safety Directory has published a new chronic intake calculation model, the National Estimate of Dietary Intake (NEDI) model, with mean and 97.5th percentile consumption data

Table I. Average and 97.5th percentile consumption (kg day^{-1}) of different food groups by Belgian adults between 19 and 59 years of age.

Commodity	Consumption		Data source
	Average	97.5 percentile	
Fruit (all)	0.111	0.288	Devriese et al. 2006
Berries	0.006	0.015	NEDI 2006
Other fruit	0.106	0.273	Calculated from Devriese et al. 2006 and NEDI 2006
Vegetables (all)	0.141	0.257	Devriese et al. 2006
Vegetables (standard* 0.05 mg kg^{-1})	0.102	0.186	Calculated from Devriese et al. 2006 and NEDI 2006
Vegetables (standard 0.1 mg kg^{-1})	0.023	0.042	Calculated from Devriese et al. 2006 and NEDI 2006
Vegetables (standard 0.2 mg kg^{-1})	0.016	0.030	Calculated from Devriese et al. 2006 and NEDI 2006
Potatoes	0.281	0.466	Devriese et al. 2006
Meat (all)	0.143	0.285	Devriese et al. 2006
Poultry	0.019	0.053	Devriese et al. 2006
Cattle	0.068	0.128	Calculated from WHO 2006 and Devriese et al. 2006
Pig	0.056	0.104	Calculated from WHO 2006 and Devriese et al. 2006
Offal (all)	3.0 10^{-4}	0.003	Devriese et al. 2006 and NEDI 2006
Cattle offal	1.2 10^{-4}	0.001	Calculated from WHO 2006 and NEDI 2006
Kidney	2.0 10^{-5}	1.7 10^{-4}	NEDI 2006
Liver	9.7 10^{-5}	8.4 10^{-4}	NEDI 2006
Other offal	1.8 10^{-4}	0.002	Calculated from WHO 2006 and NEDI 2006
Cereals	0.194	0.402	Devriese et al. 2006
Eggs	0.010	0.028	Devriese et al. 2006
Milk & milk products (excluding cheese)	0.164	0.483	Devriese et al. 2006
Cheese	0.032	0.075	Devriese et al. 2006
Fish	0.017	0.050	Devriese et al. 2006
Seafood	0.006	0.025	Devriese et al. 2006
Oils and fats	0.021	0.073	Devriese et al. 2006
Other (e.g. sweets, sugar)	0.295	0.987	Devriese et al. 2006
Total	1.417	3.421	

*Standard refers to food or food group according to the European maximum limit.

of different food groups and subgroups (National Estimate of Dietary Intake 2006). Consumption of fish and seafood is, however, lacking in this model. The GEMS/Food regional dietary pattern of raw and semi-processed food commodities database provides 5-year average intakes for most foods and for clusters of countries (World Health Organization 2006). Although dietary consumption patterns in Belgium do not differ to any extent from consumption patterns in neighbouring countries, countries with different dietary habits are included within the same cluster, e.g. Mediterranean countries, which makes the GEMS/Food database less suitable for Cd intake calculations in Belgium. Therefore, the daily consumption of the major food groups is based on the Belgian food consumption survey (Devriese et al. 2006). NEDI consumption data (National Estimate of Dietary Intake 2006) correspond relatively well to the Belgian food consumption survey data and were used to calculate the relative intake of food from subgroups within a major food group and these relative values were applied to the Belgian food consumption survey data. Remaining data gaps were filled with GEMS/Food consumption data (World Health Organization 2006) and the resulting food consumption pattern is presented in Table I.

Mean Cd concentrations were calculated for the major food groups or their subgroups. All measured Cd concentrations of individual food samples were averaged per (sub)group. The Cd content of food groups not sampled within the framework of this study (e.g. cereals, fish, milk) were collected from literature (Beernaert et al. 1990; SCOOP 2004) or from FASFC (unpublished results).

Average dietary Cd intake (µg day^{-1}) was calculated by multiplying the average Cd concentration in each food (sub)group (µg kg^{-1}) by the average daily consumed weight of that food (sub)group (kg day^{-1}). Large dietary Cd intake was calculated by using the 97.5th percentile consumption value for each food (sub)group. In general, average Cd concentrations and the half weight bound principle (i.e. concentrations <LOQ are set equal to LOQ/2) were used in intake calculations as this methodology gives a realistic and appropriate estimate of long-term exposure. To make comparisons possible, some calculations were also carried out using the median Cd concentration and the high weight bound principle (i.e. results <LOQ are considered equal to the LOQ).

Daily dietary Cd intake was compared with the provisional tolerable weekly intake (PTWI) of

Table II. Average cadmium concentration (mg kg^{-1} fresh weight) in fruits, vegetables, bovine muscle, liver and kidney from the contaminated area *vs.* other regions in Belgium with ambient environmental Cd levels (number of samples from which the mean was calculated is indicated in brackets).

	Contaminated area			Regions with ambient Cd levels		
Commodity	Mean (Number)	S.D.	Range	Mean (Number)	S.D.	Range
Berries (standard* 0.05 mg kg^{-1})	0.012 ($n=35$)	0.0106	0.005–0.043	0.0061 ($n=35$)	0.0026	0.005–0.014
Strawberries	0.012 ($n=28$)	0.011	0.005–0.043	0.006 ($n=28$)	0.0026	0.005–0.014
Other berries	0.011 ($n=7$)	0.01	0.005–0.03	0.006 ($n=7$)	0.0027	0.005–0.011
Vegetables (standard 0.05 mg kg^{-1})	0.008 ($n=25$)	0.0054	0.005–0.026	0.0072 ($n=90$)	0.0047	0.005–0.025
Cabbage	0.009 ($n=21$)	0.006	0.005–0.026	0.006 ($n=30$)	0.0026	0.005–0.018
Others (onion, bean, tomato…)	0.007 ($n=4$)	0.004	0.005–0.013	0.0079 ($n=60$)	0.0053	0.005–0.025
Vegetables (standard 0.1 mg kg^{-1})	0.085 ($n=41$)	0.0767	0.005–0.320	0.0207 ($n=69$)	0.018	0.005–0.088
Asparagus	0.009 ($n=11$)	0.004	0.005–0.015	0.011 ($n=10$)	0.009	0.005–0.032
Leek	0.065 ($n=5$)	0.043	0.011–0.13	0.0143 ($n=10$)	0.008	0.005–0.025
Carrot	0.124 ($n=15$)	0.073	0.043–0.32	0.03 ($n=29$)	0.02	0.005–0.088
Others (celery, fennel, scorzonera …)	0.12 ($n=10$)	0.073	0.029–0.28	0.015 ($n=20$)	0.016	0.005–0.065
Vegetables (standard 0.2 mg kg^{-1})	0.277 ($n=31$)	N.A$.	0.07–1.20	0.0304 ($n=68$)	0.054	0.005–0.36
Lettuce	N.A.	N.A.	N.A.	0.015 ($n=27$)	0.017	0.005–0.065
Others (spinach, parsley, mushroom …)	N.A.	N.A.	N.A.	0.04 ($n=41$)	0.067	0.005–0.36
Potatoes (standard 0.1 mg kg^{-1})	0.051 ($n=98$)	0.0297	0.011–0.15	0.0209 ($n=26$)	0.014	0.005–0.067
Meat (standard 0.05 mg kg^{-1})	0.004 ($n=53$)	0.004	0.001–0.019	0.002 ($n=99$)	0.003	0.001–0.021
Liver (standard 0.5 mg kg^{-1})	0.446 ($n=53$)	0.473	0.055–2.66	0.203 ($n=99$)	0.159	0.029–0.854
Kidney (standard 1.0 mg kg^{-1})	2.862 ($n=53$)	2.655	0.193–15.3	1.250 ($n=99$)	1.187	0.093–6.96

*Standard refers to food or food group according to the European maximum limit.
$N.A.: not available. The Cd content of this food group has not been determined in the framework of this study but is derived from unpublished data of the Federal Agency for the Safety of the Food Chain.

7 μg kg^{-1} bw per week (World Health Organization 2001) for a 60-kg adult as:

$$\%\mathrm{PTWI} = \frac{\text{Daily dietary Cd intake } (\mu\text{g day}^{-1}) \cdot 7(\text{day})}{7(\mu\text{g kg}^{-1}\text{ bw}) \cdot 60(\text{kg})} \cdot 100\% \quad (1)$$

Due to the absence of recent, publicly available, individual consumption data representative of the Belgian population, a deterministic approach has been applied only. This approach gives a realistic fraction of the PTWI to which consumers of food products produced in the vicinity of non-ferrous metal production sites are exposed.

Statistical analysis

The mean of the quantitative parameters in each group with unequal variance was compared via Welch's test (Dagnelie 1998). The limit of statistical significance of the conducted tests was defined as $p \leq 0.05$.

Results

Cadmium in vegetable and animal food products

Mean Cd concentrations in fruits, vegetables and potatoes from the contaminated area were significantly greater (1.1- to 9-fold) than samples from other regions in Belgium with ambient environmental Cd concentrations (Welch' test; $p \leq 0.001$) (Table II). The greatest increase in Cd content was found for stem, root and leafy vegetables. The average Cd concentration in leafy vegetables from the contaminated area was above the European maximum level of 0.2 mg kg^{-1} fresh weight. About 60% of vegetables (standard 0.2 mg kg^{-1}) from the contaminated area were above the European maximum level. About 3% of vegetables (standard 0.2 mg kg^{-1}) exceeded the European maximum level in other regions in Belgium. About 40% of vegetables (standard 0.1 mg kg^{-1}) from the contaminated area exceeded the maximum level. Also, 6% of the potatoes from the contaminated area were above the European maximum level. About 3% of the vegetables (standard 0.2 mg kg^{-1}) exceeded the European maximum level in other regions in Belgium. Potatoes and vegetables (standard 0.1 mg kg^{-1}) from other regions in Belgium were below the European maximum level.

The mean Cd concentrations in meat, liver and kidney from the contaminated area were significantly greater (2-fold) than from other regions in Belgium with ambient environmental Cd concentrations (Welch' test; $p \leq 0.001$). The average Cd concentration in bovine kidneys, both from contaminated and uncontaminated areas, exceeds the European maximum level of 1.0 mg kg^{-1} fresh

Table III. Local adult population in a contaminated area: mean Cd concentration (mg kg^{-1} fresh weight) of food groups, daily Cd intake (μg day^{-1}) for a 60-kg adult with an average or 97.5th percentile consumption pattern and percentage PTWI for each pattern and food group.

		Average consumption		97.5th percentile consumption	
Commodity	Mean Cd concentration (mg kg^{-1})	Daily Cd intake (μg day^{-1})	%PTWI	Daily Cd intake (μg day^{-1})	%PTWI
Fruit (all)					
Berries	0.012	0.07	0.11	0.18	0.29
Other fruit	0.004	0.42	0.70	1.09	1.82
Vegetables (all)					
Vegetables (standard* 0.05 mg kg^{-1})	0.008	0.82	1.36	1.48	2.47
Vegetables (standard 0.1 mg kg^{-1})	0.085	1.95	3.24	3.54	5.90
Vegetables (standard 0.2 mg kg^{-1})	0.277	4.55	7.57	8.27	13.8
Potatoes	0.051	14.3	23.9	23.8	39.6
Meat (all)					
Poultry	0.005	0.09	0.16	0.26	0.44
Cattle	0.004	0.27	0.45	0.51	0.85
Pig	0.007	0.39	0.65	0.73	1.22
Offal (all)					
Cattle offal					
Kidney	2.862	0.06	0.09	0.49	0.82
Liver	0.446	0.04	0.07	0.37	0.63
Other offal	0.063	0.01	0.02	0.10	0.17
Cereals	0.023	4.47	7.45	9.25	15.4
Eggs	0.003	0.03	0.05	0.08	0.14
Milk & milk products (excluding cheese)	0.4×10^{-3}	0.07	0.11	0.19	0.32
Cheese	0.9×10^{-3}	0.03	0.05	0.07	0.11
Fish	0.007	0.12	0.20	0.35	0.58
Seafood	0.136	0.86	1.43	3.41	5.69
Oils and fats	0.004	0.07	0.12	0.26	0.44
Other (e.g. sweets, sugar)	0.009	2.66	4.43	8.88	14.81
Total		31.3	52.2	63.3	105.5

*Standard refers to food or food group according to the European maximum limit.

weight (Table II). About 75% of the kidneys from the contaminated area were above the maximum level and were unsuitable to enter the food chain. Also, 50% of the kidneys from other regions in Belgium surpassed the limit value. The average Cd concentration in liver from the contaminated area was close to the European maximum level of 0.5 mg kg^{-1} fresh weight, 25% of the samples were above the limit value. Six percent of liver from other regions in Belgium were above the European maximum level. Cadmium concentrations were, however, below the European maximum level of 0.05 mg kg^{-1} fresh weight in all meat samples from Belgian cattle.

Dietary Cd intake

Dietary Cd intake was calculated for adult populations (19–59 years; assumed mean weight 60 kg) living in the Cd-contaminated area or elsewhere in Belgium, i.e. at ambient environmental Cd levels. It was assumed that the vegetables, potatoes and small fruit (berries) consumed by adults living in the contaminated area were locally produced. As a large number of cattle farms are situated in the contaminated area, it was also assumed that the consumed cattle meat and offal was locally produced. In general, this meat is nationally distributed, but farmers might have slaughtered some of their cattle for private consumption (e.g. Saegerman et al. 2002). At average food consumption levels (Table I), adults living in the Cd-contaminated area had an estimated daily Cd intake of 31 μg day^{-1} (Table III), which is almost double the estimated intake of the general adult population (17 μg day^{-1}; Table IV). Potato consumption was responsible for 46% of the daily Cd intake in the contaminated area and accounted for 24% of the PTWI (Table III). The Cd content of potatoes was lower at ambient environmental Cd levels; only contributing to 10% of the PTWI in those areas (Table IV). Other major contributors to the average daily Cd intake were vegetables and cereals. Despite the large Cd concentrations found in cattle offal, their contribution to the daily Cd intake was negligible (Tables III and IV). The percentage of the PTWI due to cattle offal consumption was less than 0.1%, regardless of environmental Cd levels.

Table IV. General adult population: mean Cd concentration (mg kg^{-1} fresh weight) of food groups, daily Cd intake (µg day^{-1}) for a 60–kg adult with an average or 97.5th percentile consumption pattern and percentage PTWI for each pattern and food group.

Commodity	Mean Cd concentration (mg kg^{-1})	Average consumption: Daily Cd intake (µg day^{-1})	Average consumption: %PTWI	97.5th percentile consumption: Daily Cd intake (µg day^{-1})	97.5th percentile consumption: %PTWI
Fruit (all)					
Berries	0.006	0.04	0.06	0.09	0.15
Other fruit	0.004	0.42	0.70	1.09	1.82
Vegetables (all)					
Vegetables (standard* 0.05 mg kg^{-1})	0.007	0.73	1.22	1.34	2.23
Vegetables (standard 0.1 mg kg^{-1})	0.021	0.47	0.79	0.86	1.44
Vegetables (standard 0.2 mg kg^{-1})	0.031	0.50	0.83	0.91	1.51
Potatoes	0.021	5.88	9.80	9.74	16.2
Meat (all)					
Poultry	0.005	0.09	0.16	0.26	0.44
Cattle	0.002	0.14	0.23	0.26	0.43
Pig	0.007	0.39	0.65	0.73	1.22
Offal (all)					
Cattle offal					
Kidney	1.250	0.03	0.04	0.22	0.36
Liver	0.203	0.02	0.03	0.17	0.28
Other offal	0.063	0.01	0.02	0.10	0.17
Cereals	0.023	4.47	7.45	9.25	15.4
Eggs	0.003	0.03	0.05	0.08	0.14
Milk & milk products (excluding cheese)	0.4×10^{-3}	0.07	0.11	0.19	0.32
Cheese	0.9×10^{-3}	0.03	0.05	0.07	0.11
Fish	0.007	0.12	0.20	0.35	0.58
Seafood	0.136	0.86	1.43	3.41	5.69
Oils and fats	0.004	0.07	0.12	0.26	0.44
Other (e.g. sweets, sugar)	0.009	2.66	4.43	8.88	14.81
Total		17.0	28.4	38.3	63.8

*Standard refers to food or food group according to the European maximum limit.

The influence of the variability in food consumption on Cd intake was taken into account by calculating the daily Cd intake based on a 97.5th percentile consumption pattern (Table I). With a daily Cd intake of 38.3 µg day^{-1}, the general adult population in Belgium reached 63.8% of the PTWI (Table IV). Adults consuming locally produced food items in the Cd-contaminated area had an estimated daily Cd intake of 63.3 µg day^{-1} (Table III). This is more than the provisional tolerable intake level for a 60-kg person, as 105.5% of the PTWI was reached at this exposure level. Potatoes and cereals remain the most important sources of Cd intake – together they account for 32% of the PTWI in the general population and 55% of the PTWI for the local population in the contaminated area.

Discussion

The dietary Cd intake of the general adult population in Belgium, estimated at 17 µg day^{-1}, is comparable to earlier published mean intake data of 23.1 µg day^{-1} (range 13.4–41.8 µg day^{-1}; Van Cauwenbergh et al. 2000) and 18 µg day^{-1} (range 2.1–88.1 µg day^{-1}; Buchet et al. 1983). Although these Cd intake values are based on duplicate meals, they confirm that the Cd intake for the general adult population in this study is a realistic estimate. The dietary exposure estimate is also close to recent estimates from the UK: 14 µg day^{-1} (Ysart et al. 2000); Denmark: 16 µg day^{-1} (Larsen et al. 2002); Tarragona, Spain: 14.3 µg day^{-1} (Bocio et al. 2005); Catalonia, Spain: 12.03/15.73 µg day^{-1} for females/males (Llobet et al. 2003), but 6-fold larger than the most recent dietary exposure estimate in France: 2.7 µg day^{-1} (Leblanc et al. 2005).

Potatoes, cereals and vegetables are the main sources of Cd for the Belgian adult population – together providing 71% of the daily Cd intake. The large consumption of these food groups and the greater Cd levels in potatoes and vegetables from the contaminated area results in a large average daily Cd intake in the contaminated area, corresponding to 44% of the PTWI. Although the mean Cd concentration in potatoes and vegetables produced in the contaminated area was below the European maximum levels, several samples exceeded this value. The high Cd concentrations in cattle offal are relatively unimportant in terms of Cd intake due to its low consumption, even within the large consumption scenario. However, consumption data are based on the general population. Assuming that offal is eaten as a replacement for meat, i.e. 143 g meat is replaced by an equal amount of liver or kidney, it can be calculated that the high figures for cattle liver and kidney consumption presented in Table I correspond to two portions of cattle liver per year and one portion of cattle kidneys every 2 years. These figures are realistic for average Belgian adults between the ages of 19 and 59 years. It is possible, however, that some people, especially cattle farmers, consume liver or kidneys more regularly. Where cattle liver originating from the contaminated area is eaten at a ration of once per month, this would increase the average weekly intake of Cd to 14.9 µg or 3.5% of the PTWI. One portion of cattle kidneys per month would increase the average weekly Cd intake to 95.5 µg or 22.7% of the PTWI. Thus, the high Cd concentrations in cattle offal are significant. Action has been taken by the federal government, involving a ban on bringing on the market, for human consumption, kidneys from bovine animals that resided at least 18 months in contaminated areas and a ban on the export of such bovine animals. Furthermore, offal, suspected of containing contaminants at such risk levels, are confiscated.

For the local adult population in a contaminated area, as well for the general adult population, intake is 2-fold higher for the large compared to the average consumption scenario. Hence, the large consumption scenario in the contaminated area reveals Cd intake at more than 100% of the PTWI. Not all food groups would be consumed in such large amounts at the same time; however, a Cd intake of 38.3 $\mu g\ day^{-1}$ for the general population is within the range reported by Van Cauwenbergh et al. (2000). Cd intake estimation in the contaminated area assumes that 100% of several food groups are produced in the area, which is an extreme but realistic assumption.

Less than 50% of results were below the LOQ. According to WHO (1995), if the proportion of <LOD results is $\leq$60%, LOD/2 should be used. LOD is meaningless in determinations via ICP–MS; hence, for intake calculations, <LOQ results were set equal to LOQ/2 (i.e. half weight bound principle). If the high weight bound principle is applied (i.e. <LOQ results are considered equal to the LOQ), the percentage PTWI reached is slightly higher (+4%). Moreover, mean Cd concentration was used for the intake calculations. The percentage PTWI reached when mean Cd concentration is replaced by median concentration in intake calculations is slightly lower (−7%), giving a PTWI of 98.1 vs. 105.5% for a large intake scenario in the contaminated area. When both the median and high weight bound principles are applied together, the PTWI reaches 101.1% for a large intake scenario in the contaminated area. Intake estimations could be improved by applying a probabilistic approach, taking into account individual variations in food consumption and/or by using a more extensive database for Cd concentrations in food items.

Overall, residual pollution from defunct, non-ferrous smelters continues to present a serious health hazard, necessitating targeted preventive measures (Nawrot et al. 2006).

Conclusion

A deterministic approach to estimating Cd intake for an adult population living in a Cd-contaminated area close to non-ferrous metal plants revealed the possible intake of almost twice as much Cd as the general adult population in Belgium (large consumption scenario assuming the local population consumes foodstuffs exclusively produced in the contaminated area). Cadmium intake by adults consuming large amounts of food surpassed the PTWI. Average Cd concentrations in fruit, vegetables, potatoes, meat and offal were higher in the contaminated area than in other regions of Belgium. A probabilistic approach is recommended to improve Cd intake estimation, taking into account (i) the distribution of individual consumption data for the sub-population living within the contaminated vicinity of non-ferrous metal plants and for the general Belgian population, and (ii) geographic distribution of Cd concentrations in an extended array of food items. Such data are not available at present.

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