Human Health Risk Assessment: A Case Study Involving Heavy Metal Soil Contamination After the Flooding of the River Meuse during the Winter of 1993–1994

Harma J. Albering,[1] Sandra M. van Leusen,[1] Edwin J.C. Moonen,[1] Jurian A. Hoogewerff,[2] and Jos C.S. Kleinjans[1]

[1]Department of Health Risk Analysis and Toxicology, Maastricht University, Maastricht, The Netherlands; [2]Geotechnical Institute, OEFPZ Arsenal, Vienna, Austria

At the end of December 1993 and also at the end of January 1995, the river Meuse, one of the major rivers in Europe, flooded and river banks were inundated. We investigated the possible health risks of exposure to heavy metal concentrations in river bank soils resulting from the flooding of the river Meuse at the end of 1993. Soil and deposit samples and corresponding arable and fodder crops were collected and analyzed for heavy metals. Although the soils of the floodplain of the river Meuse appeared severely polluted mainly by Cd and Zn, the heavy metal concentrations in the crops grown on these soils were within background ranges. Incidentally, the legal standard for Cd as endorsed by the Commodities Act was exceeded in wheat crops. The main exposure pathways for the general population were through the consumption of food crops grown on the river banks and through the direct ingestion of contaminated soils. For estimating potential human exposure in relation to soil pollution, we used a multiple pathway exposure model. For estimating the actual risk, we determined metal contents of vegetables grown in six experimental gardens. From this study, it can be concluded that there is a potential health risk for the river bank inhabitants as a consequence of Pb and Cd contaminations of the floodplain soils of the river Meuse, which are frequently inundated (averaged flooding frequency once every 2 years). *Key words:* exposure assessment, heavy metals, plant uptake, river Meuse, river water pollution, soil pollution. *Environ Health Perspect* 107:37–43 (1999). [Online 8 December 1998] *http://ehpnet1.niehs.nih.gov/docs/1999/107p37-43albering/abstract.html*

The river Meuse is located in western Europe; it originates in France, flows through Belgium, and enters into the North Sea in The Netherlands. The river Meuse is a rain river, which means that the river reacts very quickly to rainfall in its catchment area. At Borgharen (Fig. 1) near the Dutch-Belgian border, the annual average flow rate is around 250 m^3/sec (*1*). However, this flow rate can range from 25 m^3/sec during summertime periods of drought to 2,000 m^3/sec or more during ongoing precipitation (*2*). High peak flows repeatedly cause floodings of the river Meuse, predominantly in wintertime. The southern part of The Netherlands is particularly vulnerable to precipitation upstream in the Meuse basin in the Belgian Ardennes and northern France. This is mainly due to the geomorphologic composition of the river system, which is characterized by a steep narrow valley in the Belgian Ardennes, with only a few storage possibilities for suddenly rising water levels of the river (*2*). A discharge of 1,500 m^3/sec at Borgharen occurs on average once every 2 years and may result in flooding of the river Meuse at a few locations in the southern part of the Dutch Province of Limburg. At 2,000 m^3/sec some villages in this area become inundated. For the first time since 1926, extremely large-scale inundation of the river Meuse occurred in December 1993 and January 1995 (*2,3*). The river flooded its banks at several sites in The Netherlands and in Belgium and northern France (*2*). For example, 21,000 ha was inundated during the flood of December 1993 in the province of Limburg (10% of the total province) in The Netherlands (*4*). In time, this frequent flooding of the river in the riparian countries implies a more or less serious contamination by river pollutants of the soil in these areas. Important chemical contaminants of the water phase, and also of the sediment, are heavy metals including Zn, Pb, and Cd, as well as multiple organic compounds such as pesticides and polycyclic aromatic hydrocarbons (PAHs) resulting from the historical and actual industrial and agricultural processes in the catchment area (*1,5*). A study performed in The Netherlands after the flooding of the river Meuse in 1984 indicated that the flood deposits are highly contaminated by heavy metals (*5*). Various reports published in The Netherlands and Belgium after the previous floodings of the river Meuse in 1980 and 1984 evaluated heavy metal levels in flood deposits and topsoil and determined that the soils of the floodplain are enriched with heavy metals.

In The Netherlands as well as in France and Belgium, the soils of the floodplain are commonly used for agriculture (*6,7*). In general, the total surface of arable land is increasing with decreased flooding frequency of the river banks, while the forelands, which are regularly flooded (approximately once every 2 years), are mainly used as pasture land (*6*). Crops grown on the floodplain of the river banks of the Meuse may be consumed by man and/or by livestock. Incidental studies conducted in Belgium and The Netherlands have shown that high levels of Cd and/or Pb may be present in vegetables (strawberries, lettuce, spinach, endive, and kale), arable crops (barley and wheat), and fodder crops (ensiled grass and hay produced from flooded grass) grown on the floodplain of the river Meuse, although the major part of the samples taken from vegetables and arable and fodder crops grown on the river banks show heavy metal levels in background range (*6*).

The objective of this study was to evaluate heavy metal exposure risks for inhabitants of the river banks in relation to the flooding of the river Meuse during the winter of 1993–1994. The heavy metal contents (e.g., As, Cd, Cu, Pb, and Zn) of the topsoil and flood deposits and the corresponding food and feed crops were evaluated. A general multiple pathway exposure model (HESP) was used to estimate potential human exposure in relation to soil contamination of the floodplain of the river Meuse (*8*). This model relates the soil concentration of a pollutant to various environmental media and predicts the concentration of a pollutant in vegetation, beef, and dairy products. In this respect, it is of relevance that uncertainties in modeling the food chain are dominated by uncertainty of transfer factors, for example, from soil to plant (*9,10*). The uptake of heavy metals by plants appears to be influenced by different factors specific for both soil and plant, such as pH, temperature, cation exchange capacity of the soil, the presence of other heavy metals in the soil, chemical speciation, the age of the plant, plant species, etc. (*11–15*). To assess these uncertainties in the present study, we investigated the metal uptake by three different crop groups, e.g., legumes (beans), leafy vegetables (lettuce), and root vegetables (potatoes), in six experimental gardens located on

Address correspondence to H.J. Albering, Department of Health Risk Analysis and Toxicology, Maastricht University, PO Box 616, 6200 MD Maastricht, The Netherlands.

Received 13 April 1998; accepted 12 August 1998.

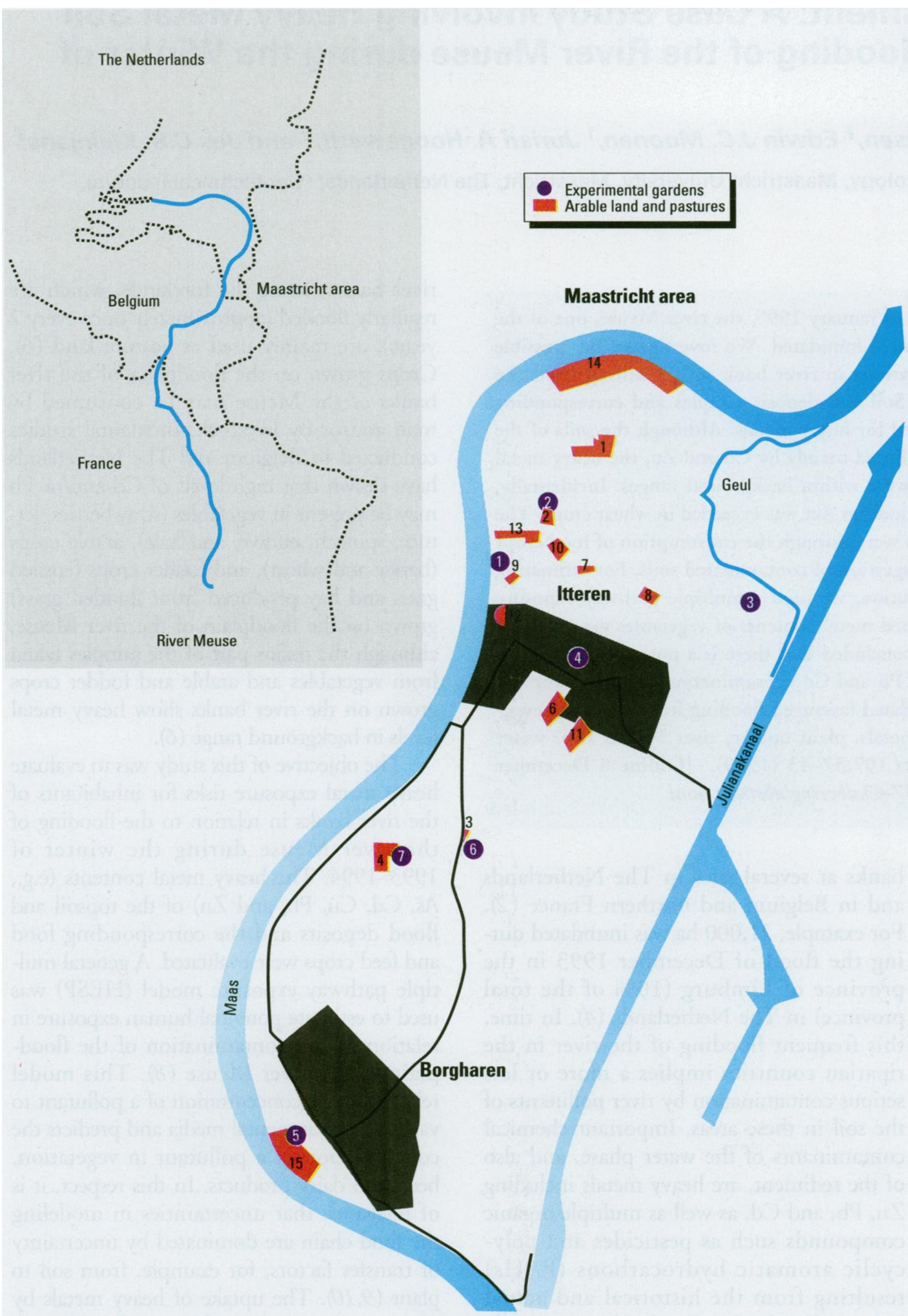

Figure 1. Map of the river Meuse and the area under study.

the floodplain of the river Meuse, and we calculated exposure risks as a consequence of the consumption of locally grown vegetables.

Methods

Study Area

The study area (see Fig. 1) is located in the province of Limburg in the southern part of The Netherlands. Two villages are located in this area. The lowest parts of the two villages are predisposed to flooding because they have little protection from the river and are built close to the river bank at the site where the steep slope of the river becomes more moderate (*2*). In general, the soils of the floodplain of the river Meuse in this area are used for vegetable gardens as well as for pasture and arable farming. The range of flooding frequencies in these areas is from once every 2 years to once every 3,000 years.

Sampling

Immediately after the flooding in January 1994, soil samples were collected from 15 agricultural fields (at least two samples, with a maximum of five samples per field), a total of 48 soil samples. Seven samples were taken from soils with a flooding frequency on average of once every 2 years; 25, 15, and 1 samples were taken from soils with a flooding frequency on average of once every 10, 50, and 3,000 years, respectively. Each individual soil sample was composed of five pooled subsamples taken from the topsoil layer (1 m^2) at a depth of 0–20 cm. The underlying assumption for this sample practice was that the soil contamination was homogeneously deposited by the river. Additionally, samples from the flood deposits and the underlying subsoils were collected from five fields. The thickness of the flood deposits varied from 5 to 10 cm. From these fields, crops such as silage maize (*Zea mays*), potatoes (*Solanum tuberosum* L.), English ryegrass (*Lolium perenne*), and wheat (*Triticum aestivum* L.) were sampled just prior to harvest in late July, August, and September 1994.

Furthermore, we established six experimental gardens on the floodplains of the river Meuse in March 1994. Before planting, soil samples were collected as described above. The soil was turned over to a depth of 25 cm, and a solid commercial fertilizer (27% nitrogen) was used in an application rate of 4 kg/ha. Potatoes, lettuce (*Lactuca sativa acephala*), and beans (*Phaseolus vulgaris*) were sown in these gardens in April and May 1994. Weeding, watering, and loosening of the soil was done manually as needed. A net was spread over the garden to protect the crops from birds and rabbits. Additionally, metaldehyde (6.4%) was applied following label recommendations once during the season of growth to prevent slugs. Lettuce was harvested in late June or early July, and potatoes and beans were harvested during late July and early August.

Chemical Analysis

Soil and deposits. Soil and flood deposit samples were dried at 105°C, sieved (2 mm), and subsequently ground in an agate mortar and stored until analysis. For metal analysis, 0.5 g of the ground samples were digested with aqua regia for 2 hr and filtered. After evaporation to a moist residue of 1 ml, the residue was dissolved in 100 ml of 0.1% HNO_3. Concentrations of As, Cd, Cu, and Pb were measured by graphite-furnace atomic absorption spectrometry (AAS) with Zeeman background corrections, and concentrations of Zn were measured by flame AAS. All glass and plastic labware was previously washed in diluted nitric acid and deionized water to prevent contamination.

Soil parameters (pH-KCl, organic matter, and clay fraction) were determined according to Dutch Normalization Institute instructions. In short, the pH was estimated potentiometrically after equilibration with

KCl, and organic matter was determined by loss-on ignition at 550°C for 2 hr. The clay fraction (particulate fraction <2 µm) was determined by treatment with H_2O_2 and HCl, followed by sieving over a 38 µm sieve. Upon settling, the clay fraction was subsequently separated. For both the agricultural and experimental fields, we determined the pH and organic fraction of the soil. We determined the clay content of the soil only in the experimental gardens.

Crops. All portions of lettuce, beans, and potatoes produced during the entire growing season in the experimental gardens were cumulatively harvested. Crops were thoroughly washed with demineralized water to remove surface dust and soil. A subsample was taken prior to the washing procedure to determine the dry weight. In addition, the ends of the beans were trimmed and the potatoes were peeled with a stainless steel peeler and sliced. The edible material was bulked, dried at 70°C, and ground in an agate mortar. To prevent slime formation, the lettuce samples were previously dried at 35°C.

Agricultural crop samples were randomly collected from the fields. To determine dry weight, a subsample was taken from the agricultural crops. Ryegrass samples were taken with ceramic scissors up to 5 cm above the topsoil, sliced, dried at 70°C, and ground in a stainless steel mill. From the wheat plants, only the ears were collected. The grains were separated, dried at 70°C, and ground. The total silage maize plants were collected, air-dried, bulked, and ground. The same procedure was applied to the agricultural potatoes as described for the potatoes grown in the experimental gardens.

Each sample (0.5 g) was treated with 20 ml of a 1:1 mixture of concentrated HNO_3 and water purified by a MilliQ water purification system, and boiled for 30 min. After filtration, the filtrate was evaporated to a moist residue (2–3 ml) and subsequently diluted in 50 ml of 0.1% HNO_3. Concentrations of Cd, Cu, Pb, and Zn were measured by AAS as described above.

Soil guideline values in The Netherlands. At present, the Dutch government uses two guideline values, e.g., intervention and target value, to assess the degree of pollution in the soil and to decide on remediation strategies (*16*). The target value refers to an acceptable or natural concentration in the soil. The intervention value indicates an unacceptable risk to man or environment due to soil contamination and is based on human toxicological and ecotoxicological data. The values are standardized for the clay fraction (25%) and organic matter fraction (10%) of the soil. Exceeding the intervention value implies a potential risk, taking all possible exposure pathways into account. Subsequently, an actual risk analysis (taking into account only the relevant exposure pathways) must be performed in order to determine the priority for clean up. In the present study, heavy metal levels of soil and sediment samples were normalized for the organic fraction (average 8.5%) and clay fraction (average 2%) of the floodplain soils and compared to these target and intervention values.

Commodities Act. We compared the heavy metal concentrations in the vegetables and arable crops to the legal standards according to the Commodities Act in The Netherlands (*17*). We also compared these values to generally observed concentrations in vegetables in The Netherlands and to baseline values of heavy metals in crops grown in uncontaminated areas (*18,19*). Commodities Act standards are based on generally observed concentrations in the edible part of the crops as well as on established acceptable daily intake standards. No standards for crops have been set for Cu and Zn. The concentrations of heavy metals in fodder crops were compared to the prescribed limits according to the Commodity Board on animal feed in The Netherlands (*20*).

Exposure Assessment

For quantification of potential human exposure in relation to the contamination by heavy metals of the banks after the flooding of the river Meuse, we used a multiple pathway exposure model (Human Exposure to Soil Pollutants; HESP) (*8*). This model describes all relevant exposure pathways and transfer processes in relation to the physicochemical characteristics of the soil. The model uses a general equation to estimate exposure to contaminants:

$$\text{Exposure} = \frac{C \times IR \times EF \times FI \times AF}{BW}, \quad (1)$$

where C = concentration of the contaminant in different media, IR = ingestion rate, EF = exposure frequency, FI = fraction contaminated, AF = absorption factor, and BW = body weight.

In the present study, human exposure was assessed by two different methods. First, we applied the standard HESP model, the standard model in soil quality assessments in The Netherlands, which solely uses data on soil pollution as the input parameter. Relevant exposure pathways in relation to the agricultural function of the floodplain of the river Meuse were the ingestion of soil, the ingestion of crops grown on the contaminated floodplains, and ingestion of meat and dairy products from cattle, pigs, and poultry fed with feed crops grown on the contaminated floodplains. By modeling these pathways, we assumed that adults and children spent 24 hr/day every day of the year on the contaminated site. Moreover, the annual average fraction of time that adults and children spent outdoors on the site was 0.32 and 0.12, respectively. Furthermore, we assumed that 10% of the ingested vegetables, meat, and dairy products came from the contaminated site. The ingestion rates of soil and dust by adults and children were estimated to be 295 and 150 mg/day, respectively (*8*).

Secondly, in view of the uncertainties regarding plant–soil transfer, the indirect exposure pathway of homegrown consumed vegetables was quantified by using the results of the heavy metal analyses in the crops grown in the experimental gardens on the soils of the floodplain, so that more specific data could be applied as input parameters to the HESP model. We assumed that the crops cultivated in the experimental gardens represent the range of crops generally grown in vegetable gardens. Subsequently, we assumed that the heavy metal uptake by potatoes, beans, and lettuce represented the metal uptake in all root vegetables, legumes, and leafy vegetables. Further in contrast to HESP conditions, we assumed that 100% of the consumed vegetables were homegrown. The indirect exposure pathway was determined by multiplying the daily consumption rate of homegrown vegetables with the contamination of the heavy metals in the corresponding crops. The daily consumption rate of homegrown vegetables was derived from a study performed by home gardeners in The Netherlands (*21*). The daily consumption rates for potatoes, leafy vegetables, root vegetables, legumes, kale, and other vegetables were 158, 80, 41, 37, 29, and 68 g for adults and 75, 25, 16, 19, 9, and 21 g for children, respectively. The heavy metal concentration in kale and other vegetables were assumed to equal the heavy metal concentration in leafy vegetables. To estimate actual human exposure, ingestion of soil was also taken into account. The ingestion rates of soil for adults and children were assumed to be 50 and 150 mg/day, respectively (*8*). The soil ingestion rate for adults was less than 295 mg/day, as used for the potential human exposure assessment, because the fraction of time that adults spent outdoors on the site was assumed to be 0.05 on annual average instead of 0.32 (*8*). Furthermore, in the study area, it was questionable whether the locally produced dairy and meat products were actually consumed by inhabitants. Therefore, this pathway was excluded.

Total exposure levels were calculated for both children and adults and compared to the established tolerable daily intake values (TDI). The TDI refers to the dose of a substance that can be taken in daily without identifiable risk to lifelong exposure. Additionally, the hazard quotient was calculated, which refers to the ratio of the calculated lifetime daily exposure devided by the reference dose (TDI). Daily exposure (mg/kg/day) averaged over a lifetime (e.g, 70 years) was calculated by

$$\frac{6 \times \text{daily exposure}_{\text{child}}}{70} + \frac{64 \times \text{daily exposure}_{\text{adult}}}{70}. \qquad (2)$$

If the hazard quotient is below 1, no health risk may occur. For Pb, different TDIs have been established for adults and children because children appear to be more sensitive to Pb than adults (*22*). The TDIs for heavy metals were taken from Veerkamp and ten Berge (*8*).

Statistical Analysis

Spearman rank correlation coefficient analysis was used to explore the relationship between the heavy metal content in soil and soil characteristics such as pH, organic matter, and clay fraction. Furthermore, this analysis was used to correlate mutual data. Statistical differences between the heavy metal content in soils of the floodplain of the river Meuse with different flooding frequencies were evaluated by means of the Mann-Whitney *U*-test. Furthermore, the Mann-Whitney *U*-test was used to evaluate the heavy metal uptake by the different plant species. For all statistical analyses, $p<0.05$ was considered significant.

Results

Soil and Flood Deposits

Heavy metal analysis in deposits and underlying soil taken from five arable fields showed that the levels of Cd, Zn, Pb, Cu, and As in the deposits, (82%, 51%, 42%, 36%, and 17%, respectively) were markedly higher on average than in the underlying soil. Concentrations of heavy metals in soil samples collected at the arable fields are presented in Figure 2. We observed the highest heavy metal concentrations in soils from the most frequently flooded areas. The wide range illustrates the variation in heavy metal concentrations in the floodplain soils. For example, the heavy metal concentrations in soils with a flooding frequency of once every 2 years on average varied from 11.1 to 17.7 mg/kg dry weight (dw) for As, 5.6 to 14.0 mg/kg dw for Cd, 62 to 116 mg/kg dw for Cu, 157 to 326 mg/kg dw for Pb, and from 705 to 1,239 mg/kg dw for Zn. Soils of the floodplain that are inundated on average once every 50 years were significantly less contaminated by heavy metals (except As) compared to soils which are flooded at higher frequency (Mann Whitney *U*-test, $p<0.05$). Moreover, in soils inundated on average once every 10 years, the Cd levels were significantly lower in comparison to the soils that are inundated on average once every 2 years.

Comparison of the results with the soil quality guidelines as set by the Dutch government indicated that the intervention value for Zn was exceeded in 80% of the soil samples. Incidentally, the intervention value for Cd and Cu was exceeded.

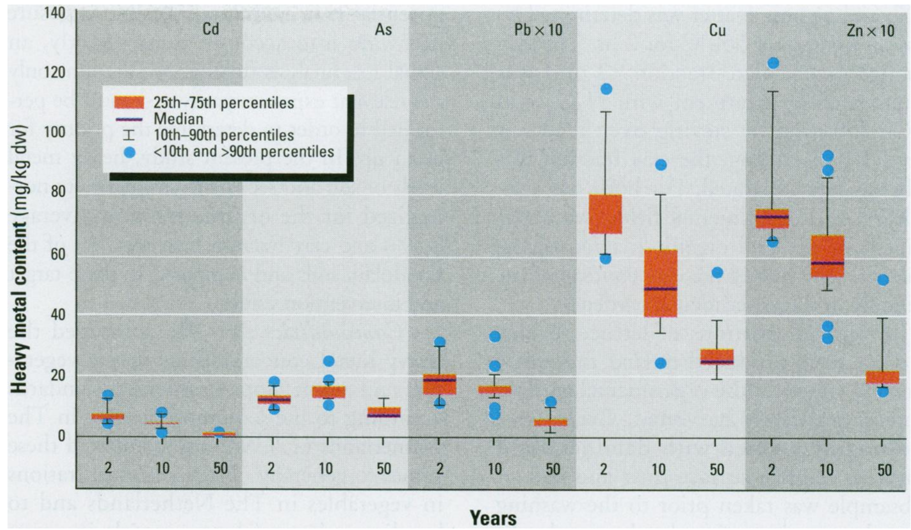

Figure 2. Heavy metal content (mg/kg dry weight) in soil samples from the floodplain of the river Meuse, The Netherlands, in relation to the flooding frequency of once every 2, 10, and 50 years on average. The Pb and Zn concentrations have been multiplied by 10.

Spearman rank correlation coefficient analysis demonstrated a significant correlation between the organic matter fraction of the soil and the heavy metal content in the soil (except As). Subsequently, the pH of the soil significantly correlated with heavy metal content; the heavy metal levels were significantly interrelated ($p<0.05$).

Crops and Animal Tissue

The ranges of heavy metals in arable and fodder crops cultivated in the agricultural fields are shown in Table 1, as compared to the guideline value according to the Commodities Act and to the Commodity Board on animal feed in The Netherlands. The permissible level for Cd (0.15 mg/kg fresh weight) was exceeded in only two wheat samples, despite the fact that these plants were cultivated in soils with a relatively low Cd content (1.1 and 4.4 mg/kg dw). The heavy metal content observed in the other arable and fodder crops were within normal background levels (Table 2).

Table 1. Heavy metal content (mg/kg fresh weight) in arable and fodder crops

Arable and fodder crops	No.	Cd		Cu		Pb		Zn
		Range	Standard[a]	Range	Standard	Range	Standard	Range
Wheat	4	0.08–0.28	0.15	3.5–4.9		0.08–0.17	0.5	38–45.2
Potatoes	3	0.02–0.03	0.1	1.3–1.4		–[b]	0.2	2.1–5.5
Silage maize	2	0.15–0.33	1	4.2	35[c]	1.4	10	39–42
Ryegrass	4	0.11–0.12	1	6.2–7.6	35[c]	0.9–1.5	10	35–55

[a]Standard according to the Commodities Act (wheat, potatoes, kidney, and liver) and maximum permissible concentrations in single feedstuffs (silage maize and ryegrass) according to the Commodity Board on animal feed. The heavy metal concentrations in maize and grass have been calculated assuming a moisture content of 12%.
[b]Below the detection limit.
[c]Mixed feedstuffs.

Table 2. Background values for heavy metals in vegetables and arable and fodder crops in The Netherlands (mg/kg fresh weight)[a]

Source	Cd	Cu	Pb	Zn
Vegetables				
Lettuce	0.01–0.19	0.23–1.6	0.01–0.03	1.2–4.3
Potatoes	0.01–0.09	0.3–2.9	0.01–0.08[b]	1.9–11
Beans	–[c]	0.3–0.8	–	1.9–10
Arable crops				
Wheat	0.01–0.26	1.4–4.5	0.02–0.69	19–41
Potatoes	0.01–0.09	0.3–2.9	0.01–0.08[b]	1.9–11
Fodder crops				
Silage maize	0.14–6.8[b]	–	1.0–4.1[b]	–
Ryegrass	0.03–0.84[b]	–	0.7–9.1[b]	–

[a]Data from Staarink and Hakkenbrak (*18*).
[b]Data from Wiersma et al. (*19*).
[c]No information available.

Heavy metal levels of vegetable crops cultivated in the experimental gardens are shown in Table 3. No detectable concentrations of Pb were found in beans and potatoes. In addition, the Cd level in beans was below the detection limit. The heavy metal concentrations in these crops were also within normal background ranges except for the Pb and Zn levels in lettuce, in which samples with relatively high levels were observed. Moreover, in one garden located in an area with a flooding frequency of once every 2 years, the Cd concentration in lettuce and potatoes exceeded the Commodity Act standard (0.1 mg/kg fresh weight). Mann-Whitney *U*-test analysis showed a significant difference between the Cu content of potatoes and the Cu content of beans and lettuce. A significantly higher concentration of Cd was observed in lettuce in comparison to the Cd content in potatoes. In addition, the Zn content in lettuce significantly differed from the Zn content of potatoes and beans (Mann-Whitney *U*-test, $p<0.05$).

Exposure Assessment

We used two different methods to estimate human exposure in relation to soil pollution of the banks of the river Meuse. First, the standard HESP model was applied, which solely uses data on soil pollution. Human exposure to heavy metals in soil of the floodplain of the river Meuse might occur directly through the ingestion of soil or indirectly through consumption of locally grown vegetables and dairy and meat products from locally raised farm animals. Taking background exposure into account, the TDI for Pb and Cd appeared to be exceeded by children at all sample locations. For adults, the TDI was exceeded at 65% of the locations; this resulted in a hazard quotient for Cd varying from 1.4 to 5.5. Only at one location was the TDI for As and Zn exceeded by children; however, the hazard quotient was below 1. The ingestion of dairy and meat products from locally raised farm animals appeared to be the primary source of potential human exposure and accounted for more than 80% (adults) or 90% (children) of total human exposure.

Furthermore, human exposure was assessed by taking into account the location-specific data of heavy metals in vegetables grown in the established experimental gardens. Table 4 summarizes the results of the actual exposure assessment for adults and children exposed to contaminants in the floodplain soil, which floods an average of once every 2 years. Only one experimental garden (No. 1) was located in this area (see Fig. 1). The heavy metal concentrations in vegetables cultivated in the other experimental gardens (Nos. 2–6), were relatively low, and the results of the exposure assessment indicate that there was no potential human health risk (data not shown). Cd and Pb intake in excess of TDI values was indicated only for soils with a flooding frequency of once every 2 years on average (experimental garden No. 1). The most important exposure pathway for Cd appeared to be the ingestion of locally grown vegetables. Specifically for children, a health risk of Pb was indicated, with the main exposure pathway being the direct ingestion of soil.

Discussion

Soil and Flood Deposits

In the area under study, the heavy metal concentrations in the soil of the floodplain of the river Meuse significantly increased with increased flooding frequency of the river. The area was frequently inundated in the past decades. The results showed an enrichment of the floodplain soil by heavy metals, predominantly Cd and Zn. In general, low levels of As and Cu were found in these soils; the concentration of As was comparable to background concentrations in Dutch agricultural clay soils (*6*). The wide range (Fig. 2) illustrates the variation of heavy metal concentrations in the soil, which is probably related to differences in sedimentation, mineralogy, and particle size characteristics as well as nonfluvial supply of heavy metals (*6*). The measured values were well in agreement with reported literature values (*4,6*). In comparison to Dutch soil quality guideline values, the intervention value for Zn was frequently exceeded.

After our 1994 study, widespread inundation of the river Meuse again occurred in January 1995. Large parts of The Netherlands as well as parts of Belgium and northern France became inundated (*2*). The flood had a longer duration than the flood of December 1993. Settled sediments along the Dutch part of the

Table 3. Heavy metal content of the vegetables harvested in the experimental gardens on the floodplain of the river Meuse (mg/kg fresh weight)

Heavy metal	Vegetable	Median	Range	No. of samples exceeding the standard value
Cd	Lettuce	0.10	0.03–0.21	1
	Potatoes	0.04	0.02–0.12	1
	Beans	–[a]	–	0
Cu	Lettuce	0.70	0.3–1.1	
	Potatoes	1.40	1.36–1.65	
	Beans	0.66	0.3–1.38	
Pb	Lettuce	0.10	0.04–0.13	0
	Potatoes	–	–	0
	Beans	–	–	0
Zn	Lettuce	6.8	5.4–9.1	
	Potatoes	3.5	1.8–6.1	
	Beans	4.6	4.3–6.3	

[a]Below the detection limit; the detection limit for Cd and Pb in beans is 0.003 and 0.03 mg/kg fresh weight, respectively; the limit for Pb in potatoes is 0.065 mg/kg fresh weight.

Table 4. Calculated heavy metal doses (mg/kg/day) for adults and children exposed to contaminants in the floodplain soil with an inundation frequency of once every 2 years

Heavy metal	Pathway	Adult		Child
Cd	Ingestion of soil	0.10×10^{-5}		0.14×10^{-3}
	Ingestion of vegetables			
	Root vegetables	0.34×10^{-3}		0.72×10^{-3}
	Leafy vegetables[a]	0.53×10^{-3}		0.77×10^{-3}
	Legumes[b]	0.16×10^{-7}		0.38×10^{-5}
	Total[c]	0.11×10^{-2}		0.18×10^{-2}
	Hazard index		1.2	
Cu	Ingestion of soil	0.85×10^{-4}		0.12×10^{-2}
	Ingestion of vegetables			
	Root vegetables	0.44×10^{-2}		0.94×10^{-2}
	Leafy vegetables	0.24×10^{-2}		0.34×10^{-2}
	Legumes	0.35×10^{-3}		0.85×10^{-3}
	Total	0.26×10^{-1}		0.34×10^{-1}
	Hazard index		0.19	
Pb	Ingestion of soil	0.24×10^{-3}		0.33×10^{-2}
	Ingestion of vegetables			
	Root vegetables[b]	0.18×10^{-3}		0.39×10^{-3}
	Leafy vegetables	0.33×10^{-3}		0.48×10^{-3}
	Legumes[b]	0.16×10^{-6}		0.38×10^{-4}
	Total	0.10×10^{-2}		0.44×10^{-2}
	Hazard index	0.14		1.2
Zn	Ingestion of soil	0.85×10^{-3}		0.12×10^{-1}
	Ingestion of vegetables			
	Root vegetables	0.011		0.024
	Leafy vegetables	0.022		0.033
	Legumes	0.002		0.005
	Total	0.22		0.25
	Hazard index		0.22	

[a]Including kale and other vegetables.
[b]For values below the detection limit, the detection limit has been used.
[c]Including background exposure.

river after the flooding in January 1995 were, on average, of better or equal quality as compared with the situation in 1986, 1988, and 1993 (*23*). The heavy metal concentrations in settled sediment collected in the area under study were in accordance with the values reported in our study after the flooding of the river during the winter of 1993–1994 (*23*). Wolterbeek et al. (*24*) found higher metal concentrations in settled sediment in comparison to the topsoil after the flooding of the river in 1995. However, the higher concentrations in settled sediment did not result in a distinct increase in the metal content of the topsoil.

Heavy metal concentrations in surface water and suspended matter of the river Meuse have decreased during the last decades; however, the chemical quality of the sediment and soils of the floodplain has changed only to a minor degree (*1*). This means that the quality of the flood deposits of the river Meuse did not change profoundly during the past decades.

Soil characteristics such as pH, organic matter fraction, and clay fraction were considered in this study. In general, the pH and organic matter of the soil decreased with the decrease of flooding frequency of the calciferous sediments of the river Meuse (*6*). However, in this study, no relationship was observed between the soil level of heavy metals and the clay fraction (<2 μm). Relatively low clay fractions were measured in this study (from 1 to 3.1%). This might be due to the high flow velocities in the study area during the flood of the river in December 1993. The transport capacity of water at high discharges is large, and erosion of the riverbed and the streambank may result in a large supply of coarse material (*5,6*). A decrease of flow velocities results in sedimentation of more finer material (*23*). The topsoil samples collected in this study immediately after the withdrawal of the flood might consist of coarse material (e.g., low clay fraction). A positive correlation was demonstrated between the inundation frequency of the river Meuse and the clay fraction in the soil (Spearman rank correlation coefficient = 0.83, $p<0.05$). This implies that regularly inundated soils have a lower clay fraction in the soil.

Crops

Various studies have been conducted to evaluate the heavy metal uptake by plants in relation to soil pollution and atmospheric deposition on the surface of soils (*6,11, 13,15,25–27*). Variable results are reported. Larsen et al. (*27*) found elevated concentrations of Cr and As in soils and plants around a wood preservation factory in Denmark. Around a Cd processing factory in Germany, very high Cd levels were found in soils and in the banks of the Grumbach brook, which resulted in very high Cd levels in lettuce, onions, and parsley that exceeded the limit values (*25*). In contrast, Ward and Savage (*11*) observed no high values of trace metals in crops located near a superhighway in London, despite the fact that the Pb content of the surface soil was significantly increased. Heavy metal concentrations in crops grown on the enriched floodplain soils of the river Meuse are within the range of concentrations found in crops grown on uncontaminated soils, but incidentally high Cd and Pb values in crops have been observed (*6*).

Uptake of heavy metals from the soil by plant species is influenced by the physicochemical characteristics of the soil and the plant species, and can be changed by different environmental and human factors (*11–15*). For example, a low soil pH appears to increase the bioavailability of metals and enhances the uptake by plants (*10,28*). The area under study can be characterized as an agricultural area. In this study, most heavy metal concentrations in various arable and fodder crops grown in the area were within background values. In general, the relatively high pH of the floodplain soil (6.9 ± 0.6) reduced the availability of the metals for plant uptake (*6*). However, relatively high levels of Cd in wheat were observed in relation to the relatively low Cd levels in arable soils. An earlier study in 1988 on heavy metals in arable crops grown on the floodplain of the river Meuse showed similar results and indicated that the Cd levels in wheat may exceed the permissible level of 0.15 mg/kg fresh weight if the corresponding floodplain soil is pH 7 or lower and the Cd content of the soil is 1 mg Cd/kg dw or higher (*6*). In this study, samples of wheat exceeded the permissible levels. The Cd levels in the soil were 1.1 and 4.4 mg/kg dw and the pH levels of the soil were 5.5 and 7.3, respectively.

Our results confirm that the relative uptake of heavy metals differs between vegetable crops (*13*). In the experimental gardens located on the floodplain of the river Meuse, the highest concentrations of Cd, Pb, and Zn were found in lettuce, whereas the highest concentrations of Cu were observed in potatoes. Lettuce and potatoes cultivated on a soil with an inundation frequency of once every 2 years on average exceeded Commodities Act standards. In the other experimental gardens, the heavy metal contents of the vegetables were within background values (*18*). In a few cases, a significant correlation was observed between the metal uptake by the plant and the metal content in the soil. The Cd levels in lettuce and potatoes and the Zn levels in lettuce were significantly correlated with the Cd and Zn levels in the soil (Spearman rank correlation coefficient, $p<0.05$). However, due to the limited number of experimental gardens, no relationship could be assessed between the metal levels in vegetables and different soil factors such as pH, organic matter, and clay fraction.

The bioconcentration factor (BCF) is widely used in modeling human exposure in relation to exposure through ingestion of vegetables via the soil–plant–human pathway (*9,10,29,30*). The BCF is defined by the equilibrium concentration of the pollutant in plant tissue (dry weight) divided by the equilibrium concentration of the pollutant in the soil on a dry weight basis. As expected, the highest BCF values were observed for lettuce. The BCF values were 0.28–0.98 for Cd, 0.14–0.54 for Cu, 0.001–0.016 for Pb, and 0.14–0.26 for Zn. Moreover, the highest BCF value for potatoes and beans was found for Cu. The mean BCFs ± standard deviations of Cu for potatoes and beans were 0.14 ± 0.065 and 0.116 ± 0.06, respectively.

Exposure Assessment

In using food chain exposure models, one has to be aware of the uncertainties that are inherent to the model parameters (*9,10,29,30*). McKone and Ryan (*10*) performed an uncertainty analysis of a food chain model in relation to As and TCDD exposure and found that the overall uncertainty in exposure was due to the uncertainty in biotransfer factors which limits the precision of food chain exposure prediction to a 90% confidence range of roughly two orders of magnitude. By contrast, validation of the EPA food chain model based on background concentrations in air, water, and soil by Travis and Blaylock (*30*) indicated that the predicted total human exposure to contaminants in the food chain is within a factor of two of the measured exposure in market basket studies. However, the distribution of exposure in the population (variability) has not been assessed. McKone (*9*) indicated that both uncertainty and variability must be taken into account to estimate the overall variance in the prediction of exposure. For example, biotransfer factors (from soil to plant) can be seen as uncertain, whereas the daily vegetable intake is characterized by variability, which also may have an important influence on the variance of total exposure (*29*).

The concentration in the plant depends on two processes: uptake through the roots with subsequent internal transport, and deposition of dust on the leaves and subsequent uptake (*8*). Bioconcentration factors are used in the HESP exposure model to estimate the uptake through the root and the subsequent internal transport. In this study, the uncertainty in soil–plant transfer factors was reduced by measuring the heavy metal content ratios in root and leafy vegetables. The default BCF values for Cd (leafy vegetables) and Cu (root vegetables) used in the HESP model were within the range of the calculated BCF on the basis of the heavy metal content in the vegetables (dry weight) cultivated in the experimental gardens. For the other heavy metals, the calculated BCFs for leafy vegetables as well as root vegetables were remarkably lower. An exception was Cu (leafy vegetables) for which the calculated BCF values were higher. Ratios of the predicted (on basis of the default BCF value) versus measured heavy metal content in leafy vegetables were 3.5, 1.2, 6.6, and 4.2 for Cd, Cu, Pb, and Zn, respectively. The ratios for root vegetables were 2.7, 0.8, and 3.5 for Cd, Cu, and Zn, respectively. This implies that by using the HESP model, human exposure through the ingestion of vegetables was overestimated by 70% for Cd, Pb, and Zn and underestimated by 27% for Cu.

An assessment of human exposure risks, based on actually measured contaminant concentrations in locally grown vegetables as an extension to the standard HESP exposure model, indicated a possible health hazard for Cd and Pb (only for children) in soils that become inundated an average of once every 2 years. The calculation suggested that under certain circumstances consumption of homegrown vegetables might be a major contribution to total Cd exposure. The calculations were based on average crop consumption rates of the home gardeners population in The Netherlands (*21*). In the experimental gardens, lettuce, potatoes, and beans were grown as representative crops cultivated in domestic gardens. Lettuce, potatoes, and beans are grown by 93%, 67%, and 98%, respectively, of the home gardeners in The Netherlands (*21*). No insight was available into the variability of the consumption rate of domestically grown vegetables in the area under study. The exposure calculation was based on the worst-case assumption that all vegetables consumed were home grown and that the consumption of any group was fixed in relation to the other groups. However, the actual consumption of homegrown leafy vegetables, potatoes, and legumes appeared to be on average, 57%, 12%, and 22%, respectively, for the home gardeners population in The Netherlands (*21*). Taking into account these percentages, the calculated Cd exposure in relation to soil contamination of the floodplain with an inundation frequency of once every 2 years appeared to be decreased and resulted in an hazard quotient of 0.24.

Müller and Anke (*25*) investigated the relationship between high Cd levels in soil and plants and the Cd levels in hair, blood, and urine of people living in a contaminated area in Germany. The values were within the normal range, suggesting that exposure through the consumption of homegrown vegetables may be excluded. By contrast, Staessen et al. (*31*) found that 2–4% of the variance in the long-term body burden of Cd was due to the consumption of vegetables grown in domestic gardens on sandy acidic soils, implying that the ingestion of locally grown vegetables is an important source for human Cd exposure.

In summary, although the soils of the floodplain of the river Meuse appeared to be enriched with heavy metals, the heavy metal contents in crops grown on these soils were within normal background values. Incidentally, high Cd values were observed in wheat, lettuce, and potatoes. The human health risk associated with the heavy metal contamination of the soil, and indirectly the food chain, seemed very low. The most important exposure risks were associated with Cd and Pb levels in soils that have a flooding frequency of once every 2 years. In the case of Pb, the most important exposure pathway was ingestion of soil, whereas for Cd, ingestion of locally grown vegetables was the predominant exposure pathway.

In general, the applied exposure model can serve as an assessment tool to identify the critical exposure pathways in situations of soil pollution. However, the results of this study suggest that current exposure assessment models are likely to both overestimate and underestimate exposure through the ingestion of vegetables to a considerable degree. Therefore, location-specific data are necessary to calculate actual human exposure through the ingestion of vegetables.

References and Notes

1. Ploco SS. Using modular planning and modular modelling towards sound water quality management of the river Meuse. Eur Water Pollut Control 5:41–50 (1995).
2. van Meijgaard E, Jilderda R. The Meuse flood in January 1995. Weather 51:39–45 (1996).
3. van de Ven G. The Netherlands and its rivers. Tijdsch Econ Soc Geogr 87:364–370 (1996).
4. Delft Hydraulics. Survey after the Flooding Disaster of the River Meuse [in Dutch]. Delft, The Netherlands: Delft Hydraulics, 1994.
5. Leenaers H, Schouten CJ, Rang MC. Variability of the metal content of flood deposits. Environ Geol Water Sci 11:95–106 (1988).
6. Institute for Soil Fertility. Research Group Heavy Metals in River Bank Soils of the Meuse and Tributaries. Heavy Metal Concentrations in River Bank Soils and Corresponding Crops in the Meuse, Geul and Roer Basin in the Province of Limburg [in Dutch]. Haren, The Netherlands:Institute for Soil Fertility, 1987–1988.
7. Micha JC, Borlee MC. Recent historical changes on the Belgian Meuse. In: Historical Change of Large Alluvial Rivers: Western Europe (Petts GE, Moeller H, Rouse AL, eds). Chichester, UK:John Wiley 1989;269–293.
8. Veerkamp W, ten Berge W. Human Exposure to Soil Pollutants (HESP). Shell Internationale Petroleum Maatschappij b.v. The Hague:Shell, 1994.
9. McKone TE. Uncertainty and variability in human exposure to soil contaminants through home-grown food: a Monte Carlo assessment. Risk Anal 14:449–463 (1994).
10. McKone TE, Ryan PB. Human exposures to chemicals through food-chains: an uncertainty analysis. Environ Sci Technol 23:1154–1163 (1989).
11. Ward NI, Savage JM. Metal dispersion and transportational activities using food crops as biomonitors. Sci Total Environ 146/147:309–319 (1994).
12. Haghiri F. Plant uptake of cadmium as influenced by cation exchange capacity, organic matter, zinc and soil temperature. J Environ Qual 3:180–183 (1974).
13. Haghiri F. Cadmium uptake by plants. J Environ Qual 2:93–96 (1973).
14. Abdel-Sabour MF, Mortvedt JJ, Kelsoe JJ. Cadmium-zinc interactions in plants and extractable cadmium and zinc fractions in soil. Soil Sci 145:424–431 (1988).
15. Voutsa D, Grimanis A, Samara C. Trace elements in vegetables grown in an industrial area in relation to soil and air particulate matter. Environ Pollut 94:325–335 (1996).
16. van den Berg R, Denneman CAJ, Roels JM. Risk assessment of contaminated soil: proposals for adjusted, toxicologically based Dutch soil clean-up criteria. In: Contaminated Soil 93. (Arendt F, Annokkée GJ, Bosman R, van den Brink WJ, eds), Amsterdam:Kluwer Academic Publishers, 1993; 349–364.
17. Commodities Act; Regulation of Heavy Metals. Rijswijk, The Netherlands:SDU, 1985.
18. Staarink T, Hakkenbrak P. Book of Contaminants [in Dutch]. Rijswijk, The Netherlands:Commodity Inspection Department, 1991.
19. Wiersma D, van Goor BJ, van der Veen NG. Cadmium, lead, mercury and arsenic concentrations in crops and corresponding soils in The Netherlands. J Agric Food Chem 34:1067–1074 (1986).
20. Animal Feed Act. Product Board Animal Feed. The Hague:SDU, 1988.
21. Hulshof PJM. Vegetable Consumption Rate by Homegardeners [in Dutch]. Rijswijk, The Netherlands: Inspectorate for Health Care, 1988.
22. Bowers TS, Beck B, Karam HS. Assessing the relationship between environmental lead concentrations and adult blood lead levels. Risk Anal 14:183–189 (1994).
23. Hoogewerf MR, van Hout HRA. Sampling of the flood deposits of the river Meuse 1995 [in Dutch]. Maastricht, The Netherlands:Public Works and Water Management Directorate Limburg, 1995.
24. Wolterbeek HT, Verburg TG, van Meerten TG. On the 1995 flooding of the rivers Meuse, Rhine and Waal in The Netherlands: metal concentrations in deposited river sediments. Geoderma 71:143–156 (1996).
25. Müller M, Anke M. Distribution of cadmium in the food-chain (soil-plant-human) of a cadmium exposed area and the health risks of the general population. Sci Total Environ 156:252–258 (1994).
26. Davies BE. Plant-available lead and other metals in British garden soils. Sci Total Environ 9:243–262 (1978).
27. Larsen EH, Moseholm L, Nielsen M. Atmospheric deposition of trace elements around point sources and human health risk assessment. II: Uptake of arsenic and chromium by vegetables grown near a wood preservation factory. Sci Total Environ 126:263–275 (1992).
28. Garcia WJ, Blessin CW, Inglett GE, Kwolek W, Carlisle JN, Hughes LN, Meister JF. Metal accumulation and crop yield for a variety of edible crops grown in diverse soil media amended with sewage sludge. Environ Sci Technol 15:793–804 (1981).
29. Stern AH. Monte Carlo analysis of the U.S. EPA model of human exposure to cadmium in sewage sludge through consumption of garden crops. J Expo Anal Environ Epidemiol 3:449–469 (1993).
30. Travis CC, Blaylock BP. Validation of a terrestrial food chain model. J Expo Anal Environ Epidemiol 2:221–239 (1992).
31. Staessen JA, Vyncke G, Lauwerys RR, Roels HA, Celis HG, Claeys F, Dondeyne F, Fagard RH, Ide G, Lijnen PJ, et al. Transfer of cadmium from a sandy acidic soil to man: a population study. Environ Res 58:24–34 (1992).