



**HAL**  
open science

## Accumulation of trace elements in cattle from rural and industrial areas in Belgium

Nadia Waegeneers, Jean-Christophe Pizzolon, Michel Hoenig, Ludwig de Temmerman

► **To cite this version:**

Nadia Waegeneers, Jean-Christophe Pizzolon, Michel Hoenig, Ludwig de Temmerman. Accumulation of trace elements in cattle from rural and industrial areas in Belgium. *Food Additives and Contaminants*, 2009, 26 (03), pp.326-332. 10.1080/02652030802429096 . hal-00577333

**HAL Id: hal-00577333**

**<https://hal.science/hal-00577333v1>**

Submitted on 17 Mar 2011

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



**Accumulation of trace elements in cattle from rural and industrial areas in Belgium**

Journal:	<i>Food Additives and Contaminants</i>
Manuscript ID:	TFAC-2008-156.R1
Manuscript Type:	Original Research Paper
Date Submitted by the Author:	06-Aug-2008
Complete List of Authors:	Waegeneers, Nadia; Veterinary and Agrochemical Research Centre, Ecochemistry Department Pizzolon, Jean-Christophe; Veterinary and Agrochemical Research Centre, Ecochemistry Department Hoenig, Michel; Veterinary and Agrochemical Research Centre, Ecochemistry Department De Temmerman, Ludwig; Veterinary and Agrochemical Research Centre, Ecochemistry Department
Methods/Techniques:	Metals analysis - ICP/MS
Additives/Contaminants:	Copper, Heavy metals - arsenic, Heavy metals - cadmium, Lead
Food Types:	Animal products - liver, Animal products - kidney, Animal products - meat

SCHOLARONE™  
Manuscripts

1  
2  
3  
4  
5  
6  
7 **Accumulation of trace elements in cattle from rural and industrial**  
8 **areas in Belgium**  
9

10  
11  
12  
13  
14  
15 N. Waegeneers\*, J.-C. Pizzolon, M. Hoenig & L. De Temmerman  
16  
17

18  
19  
20  
21  
22  
23 Veterinary and Agrochemical Research Centre (VAR-CODA-CERVA), Ecochemistry  
24  
25 Department, Leuvensesteenweg 17, B-3080 Tervuren (Belgium)  
26  
27

28  
29  
30  
31  
32  
33 \* Corresponding author. E-mail: [nadia.waegeneers@var.fgov](mailto:nadia.waegeneers@var.fgov)  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

---

**Abstract**

Trace element concentrations were determined in meat, kidney and liver tissue of cattle, which had resided more than 18 months in areas historically contaminated by emissions from the non-ferrous metal industry or in regions with normal background metal contamination levels in Belgium. Trace element levels were determined by inductively coupled plasma-mass spectrometry. Lead, arsenic and cadmium concentrations in meat were low for all cattle. The concentrations in kidneys were respectively 1.8-, 2.2- and 2.5-fold higher in cattle from the contaminated areas compared to animals from reference areas. The European maximum level for cadmium in kidney was exceeded by 75% of the kidneys from contaminated areas and 47% of the kidneys from reference areas. Lead, arsenic and cadmium concentrations in bovine livers from contaminated areas were increased by about a factor of 2.3. Zinc levels in livers and kidneys were within normal ranges, but 20% higher in the contaminated areas compared to reference areas. Copper levels in livers were high in all areas. Dairy cattle accumulated significantly higher levels of cadmium in kidneys and of lead in liver and kidneys than did beef cattle.

**Keywords :** *cadmium, lead, arsenic, zinc, copper, kidney, liver, meat*

## Introduction

Industrial and agricultural processes have resulted in the release of toxic trace elements into the environment, although for some specific elements relatively high concentrations can naturally occur in the soil. Cadmium, lead and arsenic are causing most concern because of their transfer through the food chain, bearing a potential risk to animals and humans (Friberg et al., 1979; López-Alonso et al., 2002). The major sources of toxic trace elements in agricultural soils include atmospheric deposition (De Temmerman et al. 2003), animal manure, agrochemicals and mineral fertilizers (Nicholson et al., 1999). Cattle originating from farms located within a 20-km radius around zinc refinery plants in the Netherlands, had 2.5 times higher average cadmium levels and 1.5 times higher average lead levels in organs when compared to controls (Spierenburg et al., 1988). Cadmium and lead levels in calves from a polluted area in northern Spain, were significantly higher than those in calves raised in a rural area, whereas arsenic levels in no case differed between calves from both areas (Miranda et al., 2005). In Belgium, some rural areas have been contaminated by emissions of the non-ferrous metal industry (Lauwerys et al., 1990). This has led to increased trace element levels in soils and humans (Staessen et al., 1994; Cornelis et al., 2006). Trace element concentrations in tissues of animals raised in these regions have, however, not yet been studied.

The objective of this study was to determine toxic and trace element concentrations in tissues of cattle that resided for more than 18 months in areas close to non-ferrous metal plants, and to compare the results with cattle from areas at ambient metal levels in the environment.

## Materials and methods

### *Sampling*

Kidneys, livers and muscle tissue of bovine animals were sampled at random by the Belgium Federal Agency for the Safety of the Food Chain (FASFC) in 2005. The samples were taken as part of the official control program of the FASFC in three contaminated areas and in regions with normal background metal levels in the environment, which will be further called the reference area. The contaminated areas were (i) *site A* - a rural area in the north-eastern part of the Kempen region near three primary zinc smelters, of which one has been closed since the 1970s, (ii) *site B* - a rural area south of the city of Antwerp near a precious metals refining facility, which processes secondary raw materials and wastes to extract and refine non-ferrous metals such as lead, copper and precious metals, and (iii) *site C* - a rural area in the south of the Antwerp Kempen region near a copper refining facility and a cobalt production plant (Figure 1). The so-called reference area consists of several rural areas throughout Belgium, away from point sources of toxic and trace elements. Belgium is divided in 10 major agricultural regions, from which seven were included in the current research (Zandstreek, Zandleemstreek, Leemstreek, Kempen, Weidestreek, Condroz and Ardennen). The division is based on differences in soil geochemical properties.

[insert Figure 1 about here]

Besides sites A, B & C there are other contaminated areas in Belgium, but no samples were taken in those areas because of their negligible cattle or milk production activities. The number of samples taken by the FASFC was determined in the official

1  
2  
3  
4 control program. The methodology developed for the program is based on risk  
5 evaluation, statistical tools and current scientific knowledge (Maudoux et al., 2006). In  
6  
7 total, 99 animals from the reference area and 53 animals from the contaminated areas  
8  
9 were investigated. Each bovine animal in Belgium is obligatory marked by an ear tag  
10  
11 and has a bovine passport. Together with each sample, a copy of the passport was  
12  
13 received, containing information on the date of birth, sex and type of the animal and on  
14  
15 the farms where the animal resided. Farms where the animals had resided for more than  
16  
17 18 months, were contacted and the farmers were asked to further participate voluntarily.  
18  
19 A questionnaire was filled out with the farmers, rendering information about, amongst  
20  
21 others, the feed supplied to the animals. In total, 19 farmers in the reference area  
22  
23 (covering 20 animals) and 34 farmers in the contaminated areas (covering 38 animals)  
24  
25 were visited.  
26  
27  
28  
29  
30  
31  
32  
33  
34

### 35 *Analytical methods*

36  
37 The animal tissues were sampled by officially appointed veterinarians and according to  
38  
39 the rules outlined in the control program of the FASFC. The animal tissues were  
40  
41 immediately deep frozen and stored in a freezer until analysis. At the time of analysis,  
42  
43 the tissues were partially thawed and mixed in a home mixer to homogenize the  
44  
45 samples. Consequently, the kidney cortex and medulla, which contain unequal trace  
46  
47 element concentrations, were homogeneously mixed and metal concentrations are  
48  
49 expressed on the whole organ. For each tissue, two homogenized sub-samples were  
50  
51 mineralised by acid digestion (nitric acid) in pressure vessels of an X-PRESS  
52  
53 microwave digestion system (CEM Corporation, Matthews, NC, USA). Trace element  
54  
55 concentrations in all digests were quantified by inductively coupled plasma-mass  
56  
57  
58  
59  
60

1  
2  
3  
4 spectrometry (ICP-MS, VG PQ-ExCell, TJA, US). Each batch of 20 samples  
5  
6 additionally included one procedure blank, one certified reference material (IAEA-407  
7  
8 Fish tissue) and one laboratory control material. In case of deviations from a set of  
9  
10 common criteria for tolerable variations of blanks, and for values obtained for the  
11  
12 reference materials, the analyses in a batch had to be repeated. The analyses of As, Cd  
13  
14 and Pb in meat and organs were performed under accreditation, following the 17025  
15  
16 ISO norm. The limit of quantification (LOQ) for the analyzed trace elements was  
17  
18 calculated as 10 times the standard deviation of 10 procedure blanks, multiplied with  
19  
20 the dilution factor. The LOQ values, expressed on a fresh weight basis, equalled to  
21  
22 0.002 mg Cd kg<sup>-1</sup>, 0.006 mg Pb kg<sup>-1</sup>, 0.030 mg As kg<sup>-1</sup>, 0.04 mg Cu kg<sup>-1</sup> and 0.90 mg Zn  
23  
24 kg<sup>-1</sup>, respectively.  
25  
26  
27  
28  
29  
30  
31  
32

### 33 *Statistical analysis*

34  
35 Statistical analyses were performed with UNISTAT Statistical Package, Version 5.5.06  
36  
37 (UNISTAT Ltd, London, UK). Measured values were used for samples below the LOQ,  
38  
39 unless stated otherwise, so that distributions would not be distorted. The normal  
40  
41 distribution of data was verified by the Kolmogorov-Smirnov test with Lilliefors  
42  
43 correction. Basic statistics were calculated on untransformed data. Data that were log-  
44  
45 normally distributed, were log-transformed before further analysis. Data were analysed  
46  
47 by one-way analysis of variance (ANOVA) followed by Duncan's multiple range test  
48  
49 ( $\alpha = 0.05$ ). Two animals from the reference areas were excluded from statistical  
50  
51 analysis because they were highly contaminated with metals and the source of  
52  
53 contamination could not be traced.  
54  
55  
56  
57  
58  
59  
60



## Results and discussion

Many animal studies have shown that in chronic exposure experiments, the largest amounts of Cd are found in the liver and the kidneys (e.g. Kotsonis and Klaassen, 1978). Muscles, on the other hand, contain a relatively small part of the total body Cd (Neathery et al., 1974). Lead is distributed to both soft tissues (blood, kidney, liver, ...) and mineralising systems (bones, teeth). Among soft tissues, the liver and kidney contain the highest Pb concentrations in mammals while muscles contain the lowest concentrations. In the current study, As, Cd, Pb and Cu concentrations in meat were low compared to their respective concentrations in kidney and liver (Table I), which is in line with the earlier findings stated above. In the reference areas, the majority (57-82%) of the As, Cd and Pb concentrations in meat were below the LOQ, while in the contaminated areas 91% of the As concentrations, 28% of the Cd concentrations and 77% of the Pb concentrations were below the LOQ. Mean Cd and Cu concentrations in meat were significantly higher in the contaminated areas when compared to the reference area. For As, Pb and Zn, no significant differences could be observed between both areas. Although mean Cd concentrations are significantly different between the contaminated and the reference area, the Cd concentrations in all meat samples were far below the European maximum level of 0.05 mg kg<sup>-1</sup> fresh weight (EU, 2006) and fell within previously reported ranges in other countries (e.g. Langlands et al., 1988; Jorhem et al., 1991; López-Alonso et al., 2000; Olsson et al., 2001). The Pb concentrations in meat samples from the reference and contaminated areas were as well far below the European maximum level of 0.1 mg kg<sup>-1</sup> fresh weight (EU, 2006).

1  
2  
3  
4 Arsenic showed a higher accumulation in meat than did Cd and Pb, whereas an opposite  
5  
6 tendency could be seen in liver and kidney (Table I). Arsenic, Cd and Pb concentrations  
7  
8 were higher in kidney than in liver. It is known that kidney Cd concentrations exceed  
9  
10 the liver concentration following prolonged exposure (Kotsonis and Klaassen, 1978),  
11  
12 except at very high exposures (Bernard et al., 1980). In case of repeated exposure, liver  
13  
14 Cd levels increase rapidly and a re-distribution of Cd to the kidney occurs over a period  
15  
16 of time. The major source of renal Cd in chronic exposure is derived from hepatic Cd,  
17  
18 which is transported in the form of Cd-metallothionein (Chan and Cherian, 1993).  
19  
20  
21  
22

23 [Insert Table I around here]  
24

25  
26 Within the reference area there were no significant differences in As and Cd  
27  
28 concentrations in kidney between the different agricultural regions within the reference  
29  
30 area. The geometric mean values for As in kidney ranged from 0.026 to 0.044 mg kg<sup>-1</sup>  
31  
32 while the Cd concentrations ranged from 0.661 to 0.894 mg kg<sup>-1</sup> between the seven  
33  
34 agricultural regions. The Pb concentrations in kidney were significantly different  
35  
36 between the Zandstreek on the one hand (geometric mean: 0.246 mg kg<sup>-1</sup>) and the  
37  
38 Kempen, Weidestreek, Condroz and Ardennen on the other hand (range of geometric  
39  
40 means: 0.077-0.122 mg kg<sup>-1</sup>). The Zn concentrations in kidney differed significantly  
41  
42 between the Ardennen (geometric mean: 20 mg kg<sup>-1</sup>) and the Kempen and Weidestreek  
43  
44 (geometric mean: 14 and 15 mg kg<sup>-1</sup> respectively). There were no significant differences  
45  
46 in Cd or Zn concentrations in liver within the reference area. The geometric means for  
47  
48 Cd in liver ranged from 0.116 to 0.190 mg kg<sup>-1</sup>, those for Zn ranged from 28 to 41 mg  
49  
50 kg<sup>-1</sup>. The Pb concentrations in liver significantly differed between the Weidestreek  
51  
52 (geometric mean: 0.025 mg kg<sup>-1</sup>) and the Zandstreek, Zandleemstreek and Leemstreek  
53  
54 (of geometric means: 0.060-0.090 mg kg<sup>-1</sup>). Mean As, Cd, Pb and Zn concentrations in  
55  
56  
57  
58  
59  
60

1  
2  
3  
4 liver and kidney were all significantly higher in the contaminated areas than in the  
5 reference area (Table I). The mean concentrations in liver were 2.2-, 2.3- and 2.4-fold  
6  
7 higher for As, Cd and Pb, respectively, in the contaminated areas when compared to the  
8  
9 reference area. The average Cd concentration in livers of cattle around several zinc  
10  
11 refinery plants in the Kempen area (The Netherlands) was 0.370 mg kg<sup>-1</sup> fresh weight,  
12  
13 whereas it was 0.170 mg kg<sup>-1</sup> fresh weight in a control area (Spierenburg et al., 1988;  
14  
15 original data given on a dry weight basis, recalculated to fresh weight by means of the  
16  
17 given dry matter content of liver). These values are comparable to the data found in the  
18  
19 current study (0.446 and 0.190 mg Cd kg<sup>-1</sup> fresh weight). The mean concentrations in  
20  
21 kidneys were 1.8-fold, 2-fold and 2.5-fold higher for Pb, As and Cd, respectively, in the  
22  
23 contaminated areas when compared to the reference area. Average Cd concentrations in  
24  
25 kidneys in the Dutch Kempen area were 0.83 and 2.04 mg kg<sup>-1</sup> fresh weight in the  
26  
27 control and polluted area respectively (Spierenburg et al, 1988), which is slightly lower  
28  
29 than 1.14 and 2.86 mg Cd kg<sup>-1</sup> fresh weight found in this study. Such pronounced Cd  
30  
31 concentrations in kidneys have not been found in other EU countries so far (Antoniou et  
32  
33 al., 1989; López-Alonso et al., 2000; Miranda et al., 2001; Olsson et al., 2001). These  
34  
35 Cd levels in kidneys do cause a lot of concern as they frequently exceed the European  
36  
37 maximum level (EU, 2006). In the contaminated areas, 75% of the kidneys were above  
38  
39 the European maximum level of 1 mg kg<sup>-1</sup> fresh weight and should not enter the food  
40  
41 chain. The situation for kidneys in the reference area also causes concern as 47% of the  
42  
43 samples exceed the European maximum level. Cadmium concentrations in livers  
44  
45 exceeded less frequently the European maximum level of 0.5 mg kg<sup>-1</sup> fresh weight,  
46  
47 although still 25% of the livers from the contaminated areas and 4% of the livers from  
48  
49 the reference area did not comply with the legislation. The Pb concentrations were  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

1  
2  
3  
4 above the European maximum level of 0.5 mg kg<sup>-1</sup> fresh weight for 19% of the kidney  
5  
6 samples from the contaminated areas, 8% of the kidney samples from the reference  
7  
8 area, 6% of the liver samples from the contaminated areas and 1% of the liver samples  
9  
10 from the reference area. The age of the animals is an important factor in the  
11  
12 accumulation of trace elements, and especially Cd, in organs (Andersen and Hovgård  
13  
14 Hansen, 1982; Antoniou et al., 1989). There was, however, no significant difference in  
15  
16 the mean age of animals from the contaminated areas when compared to the reference  
17  
18 area, neither were there any significant differences in age within the reference area  
19  
20 (Table II). Zinc levels in livers and kidneys were within normal ranges (Korsrud et al.,  
21  
22 1985; Spierenburg et al., 1988; López Alonso et al., 2000 and references therein), but  
23  
24 20% higher in the contaminated areas compared to other regions. Zinc is known to be  
25  
26 widely distributed in body tissues, but reaches its highest concentrations in liver, bones,  
27  
28 muscles and kidneys (Eisler, 1973). Liver copper levels varied widely among all  
29  
30 animals (1.9-395 mg kg<sup>-1</sup> fresh weight; Table I) and mean levels were high compared to  
31  
32 literature data as well in the reference areas as in the contaminated areas (Spierenburg et  
33  
34 al, 1988; López Alonso et al., 2000 and references therein).

41  
42 [Insert Table II around here]

43  
44 Interactions between trace element uptake and accumulation may be synergistic  
45  
46 or antagonistic. Cadmium raises for example Zn levels in kidneys and liver because of  
47  
48 active transport from the blood and the formation of metallothioneins, which bind both  
49  
50 Cd and Zn (Thijs et al., 1992). Tabel III presents Pearson correlation coefficients  
51  
52 between trace element concentrations in kidney and liver. These correlations show  
53  
54 indeed increased Zn levels with Cd accumulation in kidney and liver, both in the  
55  
56 reference area as in the contaminated areas. Also Pb, Cu and Zn were positively  
57  
58  
59  
60

1  
2  
3  
4 correlated to each other in kidneys. Cadmium and Cu in kidneys were not correlated  
5  
6 with each other in the reference area. In liver, Cd and Cu were negatively correlated.  
7  
8 Antagonistic interactions have been reported for Cd and Cu (Rice et al., 1973).  
9  
10 Cadmium causes a depression in the serum level of ceruloplasmin, the protein which is  
11  
12 responsible for Cu transport. Such interactions might lead to a Cu deficiency, which  
13  
14 might explain the very low liver Cu concentrations ( $< 5 \text{ mg kg}^{-1}$ , which is definitive for  
15  
16 Cu-deficiency) that have been found in some animals (Table I). Copper levels in liver  
17  
18 were also negatively correlated with Pb in liver. This negative correlation was also  
19  
20 observed by Spierenburg et al. (1988). It is not clear if a competition such as between  
21  
22 Cu and Cd can also explain the negative correlation between Cu and Pb. Increasing  
23  
24 trace element concentrations in kidneys were also accompanied by increased  
25  
26 concentrations of the same element in liver, except for Cu. The liver is known to be the  
27  
28 major storage organ for Cu.  
29  
30  
31  
32  
33

34  
35 [Insert Table III around here]  
36

37  
38 Dairy cattle from the reference area accumulated significantly higher levels of  
39  
40 Cd in kidneys than did beef cattle (Table IV). Similarly, the Pb concentration in kidneys  
41  
42 and in livers of dairy cattle was significantly higher than its concentration in kidneys  
43  
44 and livers of beef cattle. López-Alonso et al. (2003) found higher levels of Cd and Pb in  
45  
46 livers of dairy cattle compared to beef cattle, but not in kidneys. They suggested that the  
47  
48 differences in metal accumulation could be associated with a higher metal dietary intake  
49  
50 and a higher hepatic metabolism associated with milk production. Beef cattle in the  
51  
52 study of López-Alonso et al. (2003) received a diet essentially based on pasture whereas  
53  
54 the dairy cattle received a supplementation with corn silage and concentrates, in  
55  
56 addition to pasture. Differences in feed between beef and dairy cattle in the current  
57  
58  
59  
60

1  
2  
3  
4 study were less pronounced, although beef cattle received in general about 60% corn  
5 silage and 40% pasture (fresh pasture and silage) whereas beef cattle received in general  
6  
7 50% corn silage and 50% pasture. The diet was supplemented with concentrates, both  
8  
9 for beef and dairy cattle. The ratio of concentrates/roughage differs between dairy and  
10  
11 beef cattle. In Belgium, this ratio is about 40/60 for dairy cattle and 50/50 for beef  
12  
13 cattle. Concentrates are generally purchased and therefore comply with the Belgian  
14  
15 legislation concerning maximum allowable trace element levels. Roughage is, on the  
16  
17 other hand, very often locally grown by the farmer and not submitted to any compliance  
18  
19 control. Whether there are differences in the metal content of the roughage, will be  
20  
21 further investigated. The major difference between beef and dairy feed is, however, the  
22  
23 daily dry matter intake. The daily dry matter intake by dairy cattle is 16-20 kg, while it  
24  
25 is only about 10 kg for beef cattle (Pauwels, 1998; Römkens et al., 2007). Given the fact  
26  
27 that differences in Cd accumulation between dairy and beef cattle are only significant in  
28  
29 the kidney, suggests that the difference in dietary exposure is the most important reason  
30  
31 for the larger Cd accumulation in dairy cattle kidneys. The higher Pb concentrations in  
32  
33 liver and kidney of dairy cattle compared to beef cattle may be explained by a higher  
34  
35 dietary exposure and by Pb mobilisation from bones during dairy cattle lactation, as  
36  
37 suggested by López-Alonso et al. (2003).  
38  
39  
40  
41  
42  
43  
44  
45

46  
47 [Insert Table IV around here]  
48

49 Cattle originating from site A had higher average Cd concentrations in kidney  
50  
51 and liver than cattle from site C and the reference areas (Figure 2). Site A is well known  
52  
53 to be contaminated with Cd (Lauwerys et al., 1990; Staessen et al., 1994). The average  
54  
55 Cd concentrations in kidney and liver at site B were also higher than in the reference  
56  
57 areas. There are no differences in Pb concentrations in kidney and liver of cattle from  
58  
59  
60

1  
2  
3  
4 the three contaminated sites although the yearly average air Pb concentration in PM<sub>10</sub>  
5 dust at site B (173 ng m<sup>-3</sup> in 2006) was increased compared to air Pb emissions at site A  
6 (33 ng m<sup>-3</sup> in 2006) and ambient background concentrations (13 ng m<sup>-3</sup> in 2006), and  
7 the yearly average air Pb deposition level at site B (274 μg m<sup>-2</sup> d<sup>-1</sup> in 2006) was  
8 increased compared to ambient background concentrations (8 μg m<sup>-2</sup> d<sup>-1</sup> in 2006)  
9 (MIRA, 2007; VMM, 2007). Copper concentrations were elevated in liver of cattle  
10 originating from site C, near a Cu refining plant, but not in kidney (Figure 3).  
11  
12  
13  
14  
15  
16  
17  
18  
19

20  
21 [Insert Figure 2 around here]  
22

23 [Insert Figure 3 around here]  
24  
25  
26  
27

## 28 **Conclusion**

29  
30 Toxic and trace element concentrations have been measured in meat, kidney and liver  
31 samples of cattle from contaminated and reference areas in Belgium. Although As, Cd  
32 and Pb concentrations in meat were low in all areas, Cd and Pb concentrations in kidney  
33 and liver regularly exceeded the European maximum levels, both in the contaminated  
34 and the uncontaminated areas. Zinc levels in livers and kidneys were within normal  
35 ranges, while liver copper levels varied widely among all animals and mean levels were  
36 high compared to literature data. Dairy cattle accumulated significantly more Cd in  
37 kidneys and Pb in liver and kidneys compared to beef cattle, which is probably due to a  
38 larger dietary exposure compared of dairy cattle and by Pb mobilisation from bones  
39 during lactation. It is important to pay attention to the fact that a considerable  
40 contamination of toxic elements takes place in cattle originating from Belgium, as  
41 heavy metals can be transferred through the human food chain and cause food safety  
42 problems.  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

## References

- 1  
2  
3  
4  
5  
6  
7  
8  
9 Andersen A, Hovgård Hansen H. 1982. Cadmium and zinc in kidneys from Danish  
10 cattle. *Nord Vet Med.* 34: 340-349.  
11  
12  
13 Anatoniou V, Tsoukali-Papadopoulou H, Epivatianos P, Nathanael B. 1989. Cadmium  
14 concentrations in beef consumable tissues in relation to age of animals and area  
15 of breeding. *B Environ Contam Tox.* 43:915-919.  
16  
17  
18  
19  
20  
21 Bernard A, Goret A, Buchet JP, Roels H, Lauwerys R. 1980. Significance of cadmium  
22 levels in blood and urine during long-term exposure of rats to cadmium. *J*  
23 *Toxicol Environ Health.* 6: 175-184.  
24  
25  
26  
27  
28 Chan HM, Cherian MG. 1993. Mobilization of hepatic cadmium in pregnant rats.  
29 *Toxicol Appl Pharm.* 120: 308-314.  
30  
31  
32  
33 Cornelis C, Berghmans, P, Van Sprundel M, Van der Auwera J-C. 2006. Use of the  
34 IEUBK model for determination of exposure routes in view of site remediation.  
35 *Hum Ecol Risk Assess.* 12: 963-982.  
36  
37  
38  
39  
40 De Temmerman L, Vanongeval L, Boon W, Hoenig M, Geypens M. 2003. Heavy metal  
41 content of arable soils in northern Belgium. *Water Air Soil Poll.* 148: 61-76.  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60 Eisler R. 1997. Zinc hazards to plants and animals with emphasis on fishery and  
wildlife resources. In Cheremisinoff PN, editor. *Ecological issues and  
environmental impact assessment.* Houston: Gulf Publishers Company. p 443.  
EU 2006. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting  
maximum levels for certain contaminants in foodstuffs. *Off J Eur Union* L364/5.  
Friberg L, Nordberg GF, Vouk VB. 1979. *Handbook on the toxicology of metals.*  
Elsevier/North-Holland Biomedical Press, Amsterdam.



- 1  
2  
3  
4 Jorhem L, Slorach S, Sundström B, Ohlin B. 1991. Lead, cadmium, arsenic, and  
5 mercury in meat, liver and kidney of Swedish pigs and cattle in 1984-88. Food  
6 Addit Contam. 8:201-212.  
7  
8  
9  
10  
11 Korsrud GO, Meldrum JB, Salisbury CD, Houlahan BJ, Saschenbrecker PW, Tittiger F.  
12 1985. Trace element levels in liver and kidney from cattle, swine and poultry  
13 slaughtered in Canada. Can J Comp Med. 49: 159-163.  
14  
15  
16  
17  
18 Kotsonis FN, Klaassen CD. 1978. The relationship of metallothionein to the toxicity of  
19 cadmium after prolonged administration to rats. Toxicol Appl Pharm 46: 39-54.  
20  
21  
22  
23 Langlands JP, Donald GE, Bowles JE. 1988. Cadmium concentrations in liver, kidney  
24 and muscle in Australian sheep and cattle. Aust J Exp Agr. 28:291-297.  
25  
26  
27  
28 Lauwerys R, Amery A, Bernard A, Bruaux P, Buchet J-P, Claeys F, De Plaen P,  
29  
30 Ducoffre G, Fagard R, Lijnen P, Nick L, Roels H, Rondia D, Saint-Remy A,  
31 Sartor F, Staessen J. 1990. Health effects of environmental exposure to  
32 cadmium. Objectives, design and organization of the Cadmibel Study: a cross-  
33 sectional morbidity study carried out in Belgium from 1985 to 1989. Environ  
34 Health Persp. 87: 283-289.  
35  
36  
37  
38  
39  
40  
41  
42 López-Alonso M, Benedito JL, Miranda M, Castillo C, Hernández J, Shore RF. 2000.  
43 Toxic and trace elements in liver, kidney and meat from cattle slaughtered in  
44 Galicia (NW Spain). Food Addit Contam. 17:447-457.  
45  
46  
47  
48  
49 López-Alonso M, Benedito JL, Miranda M, Castillo C, Hernández J, Shore RF. 2002.  
50 Interactions between toxic and essential trace metals in cattle from a region with  
51 low levels of pollution. Arch Environ Con Tox. 42: 165-172.  
52  
53  
54  
55  
56  
57  
58  
59  
60

- 1  
2  
3  
4  
5 López-Alonso M, Prieto-Montaña F, Miranda M, Castillo C, Hernández J, Benedito JL.  
6  
7 2003. Cadmium and lead accumulation in cattle in NW Spain. *Vet Hum Toxicol.*  
8  
9 45: 128-130.  
10
- 11 Maudoux JP, Saegerman C, Rettinger C, Houins G, Van Huffel X, Berkvens D. 2006.  
12  
13 Food safety surveillance through a risk based control programme: approach  
14  
15 employed by the Belgian Federal Agency for the Safety of the Food Chain. *Vet*  
16  
17 *Quart.* 28: 140-154.  
18
- 19 MIRA. 2007. State of the Environment Report on the Flemish region of Belgium.  
20  
21 Available:  
22  
23 [http://www.milieurapport.be/default.aspx?pageID=86&ChapID=2611&NodeID](http://www.milieurapport.be/default.aspx?pageID=86&ChapID=2611&NodeID=2617)  
24  
25 [=2617](http://www.milieurapport.be/default.aspx?pageID=86&ChapID=2611&NodeID=2617) via the INTERNET. Last accessed 2008 April 15. In Dutch.  
26  
27  
28  
29
- 30 Miranda M, López Alonso M, Castillo C, Hernández J, Benedito JL. 2001. Cadmium  
31  
32 levels in liver, kidney and meat in calves from Asturias (North Spain). *Eur Food*  
33  
34 *Res Technol.* 212:426-430.  
35  
36
- 37 Miranda M, López Alonso M, Castillo C, Hernández J, Benedito JL. 2005. Effects of  
38  
39 moderate pollution on toxic and trace metal levels in calves from a polluted area  
40  
41 of northern Spain. *Environ Int.* 31: 543-548.  
42  
43
- 44 Neathery MW, Miller WJ, Gentry RP, Stake PE, Blackmon DM. 1974. Cadmium-109  
45  
46 and methyl mercury-203 metabolism, tissue distribution and secretion into milk  
47  
48 of cows. *J Dairy Sci.* 57: 1177-1183.  
49  
50
- 51 Nicholson FA, Chambers BJ, Williams JR, Unwin RJ. 1999. Heavy metal contents of  
52  
53 livestock feeds and animal manures in England and Wales. *Bioresource*  
54  
55 *Technol.* 70: 23-31.  
56  
57  
58  
59  
60

- 1  
2  
3  
4  
5 Olsson I-M, Jonsson S, Oskarsson A. 2001. Cadmium and zinc in kidney, liver, muscle  
6  
7 and mammary tissue from dairy cows in conventional and organic farming. J  
8  
9 Environ Monitor. 3:531-538.  
10
- 11 Pauwels, H. 1998. Rundvleesproductie: voeding van vleesstieren. Ministerie van  
12  
13 Middenstand en Landbouw, Bestuur Onderzoek en Ontwikkeling - DG 6, Dienst  
14  
15 Ontwikkeling Dierlijke productie, Hasselt. pp. 40. (in Dutch)  
16  
17
- 18 Rice DP, Murthy L, Shirly T, Menden E, Petering HG. 1973. The impact of low level  
19  
20 cadmium feeding on blood chemicals in male, Sprague-Dawley rats. Trace Subst  
21  
22 Environ Health. 7: 305-311.  
23  
24
- 25 Römken PFAM, Zeilmaker MJ, Rietra RPJJ, Kan CA, van Eijkeren JCH, van  
26  
27 Raamsdonk LWD, Lijzen JPA. 2007. Blootstelling en opname van cadmium  
28  
29 door runderen in de Kempen: een modelstudie. Alterra-rapport 1438. Alterra,  
30  
31 Wageningen, pp 70. (in Dutch)  
32  
33
- 34 Spierenburg TJ, De Graaf GJ, Baars AJ, Brus DHJ, Tielen MJM, Arts BJ. 1988.  
35  
36 Cadmium, zinc, lead, and copper in livers and kidneys of cattle in the  
37  
38 neighbourhood of zinc refineries. Environ Monit and Assess. 11: 107-114.  
39  
40
- 41 Staessen JA, Lauwerys RR, Ide G, Roels HA, Vyncke G, Amery A. 1994. Renal  
42  
43 function and historical environmental cadmium pollution from zinc smelters.  
44  
45 Lancet. 343: 1523-1527.  
46  
47
- 48 Thijs L, Staessen J, Amery A, Bruaux P, Buchet JP, Claeys F, De Plaen P, Ducoffre G,  
49  
50  
51 Lauwerys R, Lijnen P, Nick L, Remys AS, Roels, H, Rondia D, Sartor F. 1992.  
52  
53 Determination of serum zinc in a random population of four Belgian towns with  
54  
55 different degrees of environmental exposure to cadmium. Environ Health  
56  
57 Perspect. 98: 251-258.  
58  
59  
60

1  
2  
3  
4 VMM. 2007. Luchtkwaliteit in het Vlaamse Gewest - 2006. Vlaamse  
5  
6  
7 Milieumaatschappij, Aalst. pp. 148 + bijlagen (in Dutch).  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

For Peer Review Only

1  
2  
3  
4  
5  
6 Figure 1. Geographical location of the sampling areas. The contaminated sites A, B and C are  
7 indicated as white ovals. Shaded areas represent sampling areas at ambient environmental  
8 metal levels, which are considered as reference areas.  
9  
10  
11

12  
13  
14  
15 Figure 2. Average cadmium and lead concentration in kidney and liver as a function of the  
16 origin of the cattle. Bars with the same letter on top are not significantly different ( $p < 0.05$ )  
17 following Duncan multiple range test on log-transformed data. The data are retransformed  
18 into natural numbers.  
19  
20  
21  
22  
23  
24  
25  
26

27 Figure 3. Average copper concentration in kidney and liver as a function of the origin of the  
28 cattle. Bars with the same letter on top are not significantly different ( $p < 0.05$ ) following  
29 Duncan multiple range test on log-transformed data. The data are retransformed into natural  
30 numbers.  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60

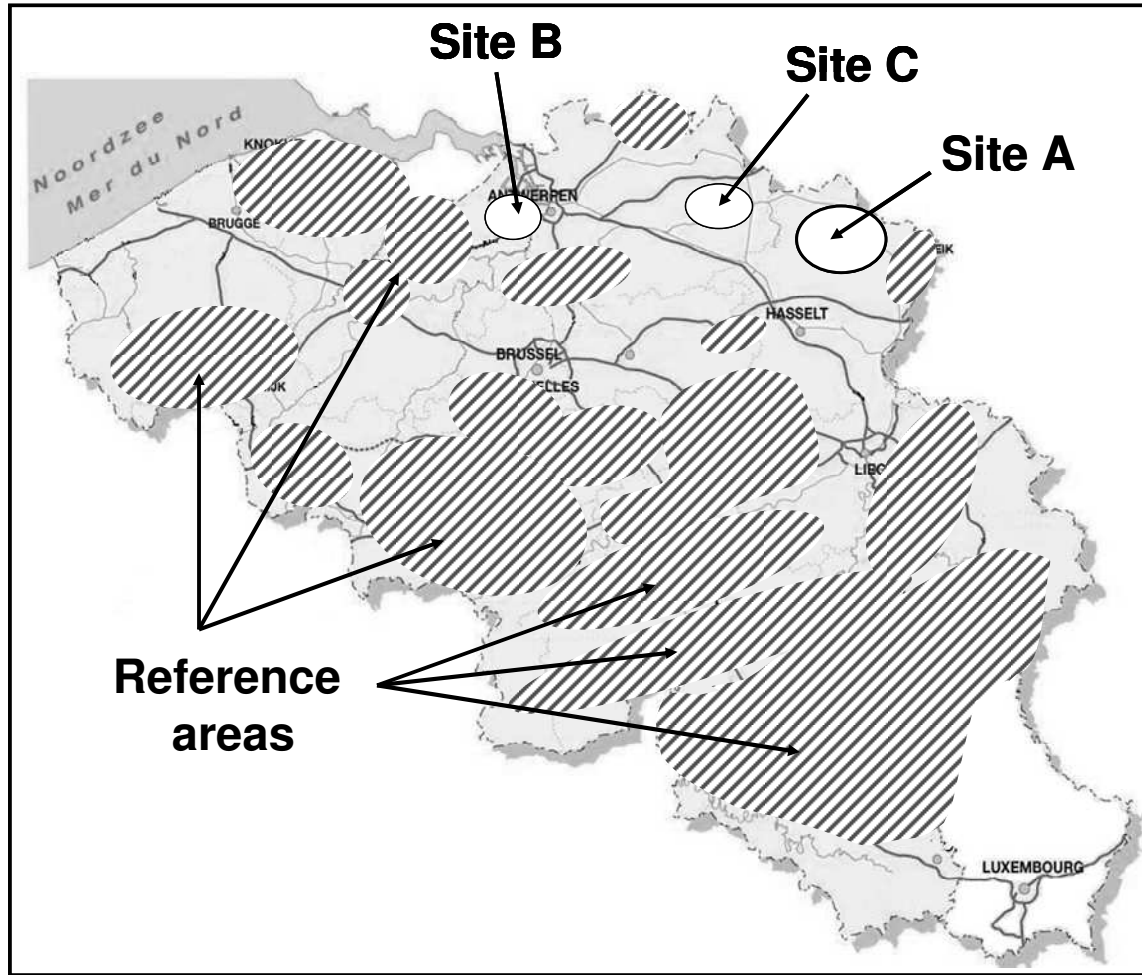


Fig. 1

1  
2  
3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47

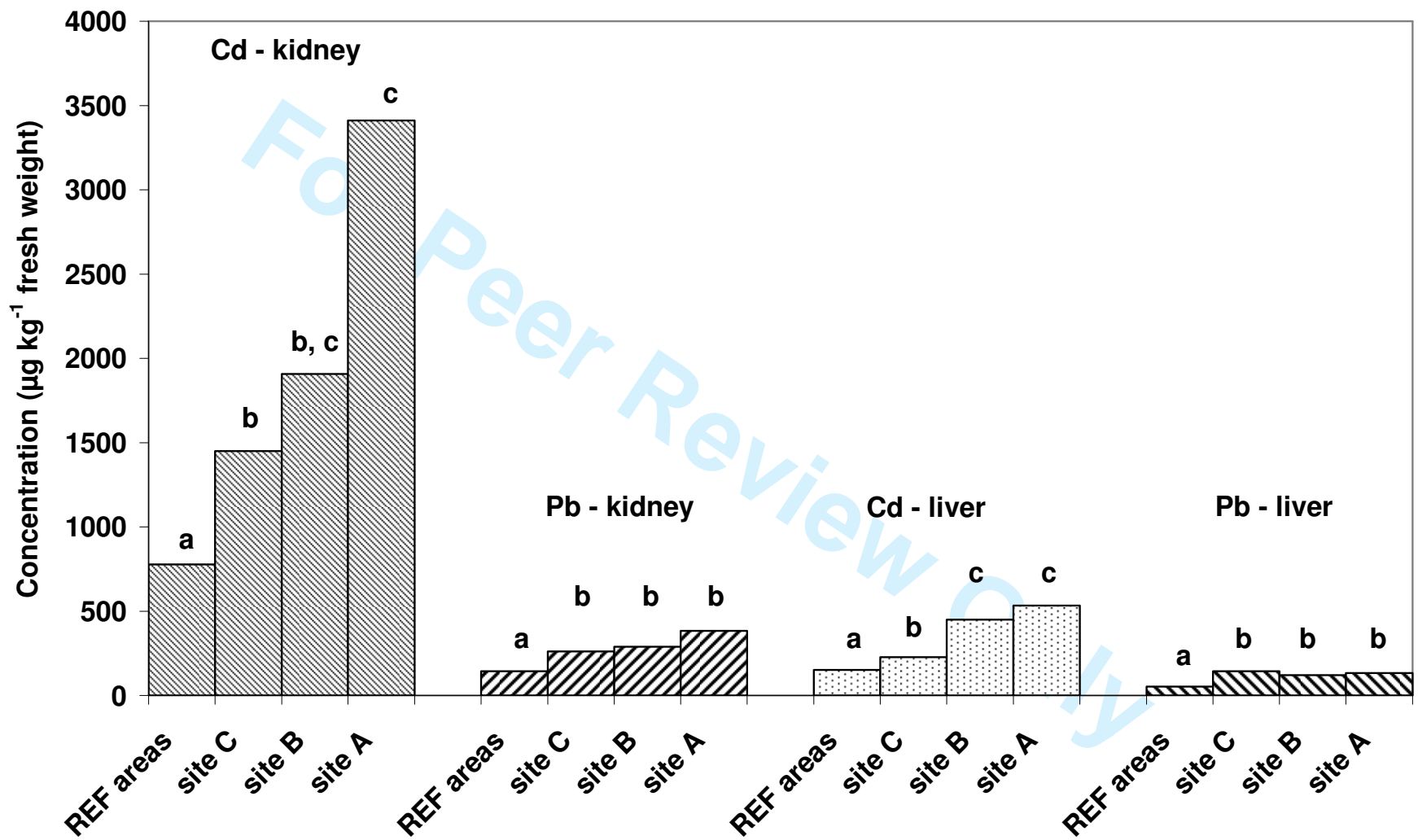


Fig. 2

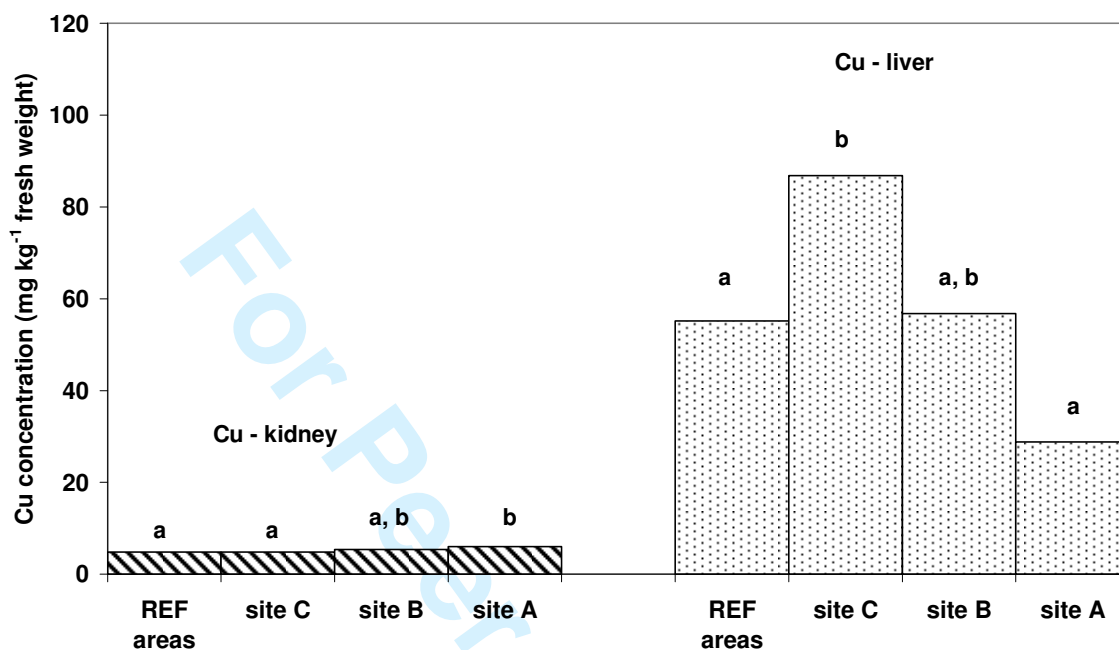


Fig. 3



**Table I.** Descriptive statistics of trace element concentrations in bovine kidney, liver and meat collected in a reference area (n = 97) and in polluted areas (n = 53). The concentrations are expressed on a fresh weight basis.

Tissue	Area		As mg kg <sup>-1</sup>	Cd mg kg <sup>-1</sup>	Pb mg kg <sup>-1</sup>	Cu mg kg <sup>-1</sup>	Zn mg kg <sup>-1</sup>
		LOQ	0.030	0.002	0.006	0.04	0.90
Kidney	<i>Reference area</i>	Mean ± stdev	0.043 ± 0.023	1.142 ± 0.922	0.212 ± 0.209	4.97 ± 1.67	18.3 ± 5.8
		Geometric mean	0.035	0.778	0.144	4.75	17.5
		Min-max	0.001 <sup>*</sup> -0.147	0.093-4.22	0.022-1.21	2.60-12.5	9.20-44.2
	<i>Polluted area</i>	Mean ± stdev	0.093 ± 0.078	2.862 ± 3.159	0.373 ± 0.318	5.31 ± 1.26	22.0 ± 5.7
		Geometric mean	0.074	1.842	0.290	5.17	21.4
		Min-max	0.006 <sup>†</sup> -0.543	0.193-15.3	0.061-1.91	2.20-9.20	14.2-41.6
Liver	<i>Reference area</i>	Mean ± stdev	0.017 ± 0.011	0.191 ± 0.136	0.082 ± 0.126	80.1 ± 66.7	40.3 ± 22.2
		Geometric mean	0.014	0.152	0.053	55.2	36.5
		Min-max	0.001 <sup>‡</sup> -0.064	0.029-0.642	0.009-1.149	1.86-395	18.1-164
	<i>Polluted area</i>	Mean ± stdev	0.037 ± 0.025	0.446 ± 0.473	0.194 ± 0.181	92.7 ± 71.8	53.9 ± 68.4
		Geometric mean	0.029	0.317	0.136	62.7	43.2
		Min-max	0.006 <sup>¶</sup> -0.111	0.055-2.655	0.017-0.984	3.1-279	21.4-518
Meat	<i>Reference area</i>	Mean ± stdev	0.023 ± 0.024	0.002 ± 0.002	0.003 ± 0.003	1.6 ± 0.5	43.3 ± 9.3
		Geometric mean	0.016	0.002	0.002	1.5	42.4
		Min-max	0.001 <sup>§</sup> -0.208	0.001 <sup>  </sup> -0.012	0.001 <sup>#</sup> -0.018	0.7-2.7	27.2-77.3
	<i>Polluted area</i>	Mean ± stdev	0.017 ± 0.009	0.004 ± 0.004	0.004 ± 0.003	2.2 ± 0.5	41.2 ± 6.6
		Geometric mean	0.015	0.003	0.003	2.1	40.7
		Min-max	0.001 <sup>**</sup> -0.047	0.001 <sup>††</sup> -0.019	0.001 <sup>‡‡</sup> -0.013	0.7-3.0	25.5-60.4

\* n < LOQ = 32; † n < LOQ = 3; ‡ n < LOQ = 88; ¶ n < LOQ = 28; § n < LOQ = 70; || n < LOQ = 55; # n < LOQ = 80; \*\* n < LOQ = 48; †† n < LOQ = 15; ‡‡ n < LOQ = 41.

**Table II.** Detailed information on the age of the animals in the seven agricultural regions of the reference area and of the animals from the three contaminated areas.

Area	Mean $\pm$ stdev	Geometric mean	Min-max	n <sup>o</sup> of samples
Zandstreek	3.4 $\pm$ 1.6	3.1	1.4-7.3	16
Zandleemstreek	5.1 $\pm$ 3.1	4.1	1.5-11.4	17
Leemstreek	4.6 $\pm$ 2.7	4.0	1.6-11.6	26
Kempen	3.8 $\pm$ 1.3	3.6	2.0-5.3	7
Weidestreek	5.4 $\pm$ 2.1	5.0	2.4-8.3	10
Condroz	3.9 $\pm$ 2.5	3.2	1.0-8.1	9
Ardennen	5.4 $\pm$ 2.4	4.9	1.6-9.7	12
Site A	4.0 $\pm$ 1.7	3.7	1.7-7.2	11
Site B	3.7 $\pm$ 1.7	3.4	2.0-7.2	12
Site C	4.9 $\pm$ 2.1	4.4	0.6-10.9	30

**Table III.** Pearson correlations between trace element contents in organs for cattle from polluted areas and from the reference area. Correlation coefficients were calculated on log-transformed trace element concentrations in fresh kidneys and livers.

	Kidney				Liver			
	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn
Kidney-Cd	-	0.59 <sup>***</sup>	0.31 <sup>*</sup>	0.61 <sup>***</sup>	0.75 <sup>***</sup>	0.25 <sup>*</sup>	-0.27 <sup>*</sup>	0.00
Pb	0.63 <sup>***</sup>	-	0.27 <sup>*</sup>	0.41 <sup>**</sup>	0.60 <sup>***</sup>	0.68 <sup>***</sup>	-0.33 <sup>**</sup>	0.06
Cu	0.07	0.24 <sup>**</sup>	-	0.50 <sup>***</sup>	0.17	-0.33 <sup>**</sup>	-0.15	0.04
Zn	0.44 <sup>***</sup>	0.50 <sup>***</sup>	0.73 <sup>***</sup>	-	0.43 <sup>***</sup>	0.15	-0.16	0.36 <sup>**</sup>
Liver-Cd	0.82 <sup>***</sup>	0.52 <sup>***</sup>	-0.09	0.26 <sup>**</sup>	-	0.51 <sup>***</sup>	-0.33 <sup>**</sup>	0.06
Pb	0.57 <sup>***</sup>	0.88 <sup>***</sup>	0.10	0.39 <sup>***</sup>	0.58 <sup>***</sup>	-	-0.21	0.30 <sup>*</sup>
Cu	-0.36 <sup>***</sup>	-0.36 <sup>***</sup>	0.14	-0.14	-0.21 <sup>*</sup>	-0.32 <sup>***</sup>	-	0.19
Zn	0.32 <sup>***</sup>	0.51 <sup>***</sup>	0.26 <sup>**</sup>	0.47 <sup>***</sup>	0.44 <sup>***</sup>	0.56 <sup>***</sup>	0.02	-

Upper right half: cattle from polluted areas.

Lower left half: cattle from the reference area.

\* p < 0.05

\*\* p < 0.01

\*\*\* p < 0.001

**Table IV.** Cadmium and lead concentrations (in mg kg<sup>-1</sup> fresh weight) in kidney and liver of dairy and beef cattle from the reference area.

	Cadmium		Lead	
	Dairy cattle	Beef cattle	Dairy cattle	Beef cattle
Kidney	1.21*	0.62	0.31*	0.11
Liver	0.20	0.13	0.12*	0.04

\* Significant differences between dairy and beef cattle at  $p < 0.05$  following Duncan multiple range test on log-transformed data. The data are retransformed into natural numbers.