HEAVY METAL CONTENT OF ARABLE SOILS IN NORTHERN BELGIUM

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(Received 6 February 2001; accepted 3 April 2003)

Abstract. More than 600 arable soils from Flanders (Northern-Belgium) were analysed for their total acid extractable contents of As, B, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn in order to determine any increase in the natural background values. Samples were taken at random in several municipalities throughout Flanders. Areas with known historical or actual sources of trace element emissions were omitted although in some cases the distance between those sources and the sampling locations was not more than 20 km. The main soil types were, ranging from north to south: sand, loamy sand, light sandy-loam, sandy-loam and loam. In the coastal area, clay soils (sea polders) were sampled. In addition to the analysis of trace elements, the soil texture class, the pH and the carbon content were determined. Macro- or mesonutrients, Ca, K, P, Mg and Na, were determined from a weak acid extract of the soil samples. Correlations between trace elements and macronutrients provide some information about fertilisation practices and heavy metal sources. A limited number of soils showed slightly enhanced levels for As, Be, Cu, Co, Cr, Mn, Ni Pb and Zn. In most cases, this could be linked to the regional industrial activities. However, a clear increase for Cu and Zn, above the natural background could be distinguished in areas with low atmospheric heavy metal deposits. In these cases, the excessive use of animal manure in the past may be the reason for this enrichment. However there was no indication that the Cd content of the soil was raised by the use of large amounts of pig slurry and/or by other common agricultural activities.

Keywords: arable soils, heavy metals, total acid extractable contents, trace elements

1. Introduction

During 1995, the arable soils of North Belgium (Flanders) were sampled and analysed for their total acid extractable contents of As, B, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb and Zn. The sampling points were spread throughout the area in order to represent the major soil types. Natural contents of heavy metals in arable soils depend primarily on their contents in the geological parent material (Bowen, 1966; Kabata-Pendias and Pendias, 1984). However, changes in composition are possible during the weathering process. Climate driven physical and chemical processes can influence the natural contents of the soil and the anthropogenic impact can be very important. The soils sampled are all from Quaternarian parent material and the textural classes range from sand to loam and clay (polder). The low natural

Water, Air, and Soil Pollution **148:** 61–76, 2003.

contents for most of the elements are typical for the Quaternarian deposits from the Pleistocene (Eolian) period. For some elements (As, B, Be, Co, Cr, Cu, Mn, Ni and Zn) there are increasing amounts as a function of increasing clay contents (De Temmerman *et al.*, 1982, 1984). 'Normal' contents of a whole series of mineral elements have been determined in several countries i.e. in Germany (Kloke, 1980) and Belgium (De Temmerman *et al.*, 1984). These normal contents can be influenced on the long term by several inputs of heavy metals. Sources of metal accumulation are long and mid-range transport of heavy metal containing dust particles and, for some elements, also agricultural practices (Kloke *et al.*, 1984). Mineral fertilizers, primarily phosphate fertilizers, and animal manure can increase the soil contents of certain mineral elements. Moreover additions of sewage sludge and some kinds of compost are able to enrich soils with heavy metals (Kloke *et al.*, 1984).

For practical reasons, the selection of sampling sites was done on the municipality level omitting areas adjacent to known sources of heavy metal containing dust. Within the selected municipalities, sampling points were selected randomly taking into account the predominant texture. The northern part of Flanders, an area of predominantly sandy and sandy-loam soils, has been industrialised since the end of the 19th century. Lead, zinc and copper smelters have or had been active for many decades and contamination of arable and forest soils were clearly shown in that area by Bosmans and Paenhuys (1980). Taking into account the effects of local sources we sought to determine eventual effects, at a distance of more than 20 km from those sources.

Accumulation of heavy metals in arable soils is important because of the potential transfer of heavy metals through crops to animals (feed crops) and humans (food crops and vegetables). To this respect Cd is an important element, not only because of the long term accumulation in humans but also because of the high potential for root uptake and accumulation in above ground plant parts. Soil conditions such as pH and texture play a very important role in the availability of cadmium in the soil (Wiersma *et al.*, 1986; Öborn *et al.*, 1995; Puschenreiter and Horak, 2000).

The aim of this study was to determine the 'normal' contents of a series of elements in arable soils and to detect whether agricultural practices (use of phosphate fertilizers, liming, application of heavy metal containing pesticides, application of sewage sludge and/or manure) and atmospheric deposition of heavy metals already had a noticeable effect on the heavy metal contents of the soil. It is not expected that enrichment with heavy metals by normal agricultural practices will be much different among fields with comparable soil texture. However, in some areas, an increased impact of atmospheric deposits is likely because of the vicinity (at 20 km distance or more) of industrial plants emitting heavy metals. Other areas were influenced by a massive use of animal wastes such as pig manure. Indeed, in these areas there was since several decades a very intensive animal husbandry and in some cases excessive amounts of manure were used in the past.

2. Materials and Methods

Samples of arable soils were selected at random in a number of municipalities where no known emissions of heavy metals occur. Only the top soil layer (0–23 cm) was sampled at 20 to 25 places per field to obtain an average sample. For each sample, the soil texture was determined by palpation by two well trained experts independently. If any differences in texture class were noticed, a third expert determined the texture class. For practical reasons the soils were grouped into 4 main texture classes: sand (sand and coarse sand), sandy-loam (loamy sand, light sand-loam, sandy-loam), silt loam, and clay (mainly polder clay). The samples were air dried in the laboratory and sieved with a 2 mm grid sieve. A standard soil analysis was carried out to determine the pH in a KCl solution (with a glass electrode) and the organic carbon content (% OC) by means of a modified Walkley & Black method (Jackson, 1958). The air-dried soil (1 g) was extracted with 20 mL ammonium lactate (0.1 N ammonium lactate and 0.4 N acetic acid; pH 3.75) to determine the K, Mg, Ca, and Na contents with atomic absorption spectrometry (AAS) (Egner *et al.*, 1960). Phosphorus was determined by colorimetry (445 nm) on the same extract after addition of a mixture of ammonium vanadate, nitric acid and ammonium molybdate (Teichner, 1967; Csato and Kadar, 1992). The results are expressed as mg kg^{-1} air-dried soil. The soil samples were further dried at 105 °C during 16 hr and extracted with a mixture of HNO_3-HCl (1:3) under reflux followed by determination of trace elements by inductively coupled plasma atomic emission spectrometry with ultrasonic nebulization (ICP-AES-USN) which allows to measure the lowest values. Low contents of Cd and Pb were confirmed by using electrothermal AAS (ETAAS). Validation of the whole method was done using reference materials CRMS 7001–7004 (light sandy soils, silty clay loam, loam) from Analytica (Czech Republic). These reference materials are certified for total, aqua regia (our case) and hot and cold HNO_3 extractable contents. For the elements tested, the recovery was in very good agreement with the certified value. The obtained results are considered as total acid extractable contents expressed in mg kg^{-1}.

The contents of trace elements in arable soils were compared to the 'normal' ranges, which were earlier determined for non-arable soils (De Temmerman *et al.*, 1984). Moreover they were compared to background values as they appear in the Flemish legislation (VLAREBO, 1996). Background values (Upper limits) for As, Cd, Cr, Cu, Pb, Ni and Zn in arable soils are defined for a standard soil containing 10% clay and 2% organic matter (OM) (% organic carbon (OC) = 0.58 $*$ (% OM)). Those values N(10,2) are 19, 0.8, 3.7, 17, 40, 9, 62 mg kg^{-1} dry matter DM, respectively. The formula $N(x,y) = N(10,2) * ((A + b * x + C * y)/(A + B * 10 + C * 2))$ allows to calculate the background value as a function of the % clay (x) and OM (y). The values for A, B and C are, respectively, 14, 0.5 and 0 for As, 0.4, 0.03 and 0.05 for Cd, 31, 0.6 and 0 for Cr, 14, 0.3 and 0 for Cu, 33, 0.3 and 2.3 for Pb, 6.5, 0.2 and 0.3 for Ni and 46, 1.1 and 2.3 for Zn. The background values

TABLE I

Trace element contents in sandy soils in mg kg^{-1} DM – 225 samples

Ele-ment	Aver.	Median	Min–max	Percentiles			'Normal' range[a]	Background value[b]
				90 pct	95 pct	99 pct		
As	4.7	3.8	0.93–34.4	8.6	10.4	15.2	0.3–8	15
Be	0.15	0.12	<0.03–0.96	0.31	0.38	0.56	0.01–0.3	
B	2.7	0.8	<0.1–32.6	7.8	13.9	29.0	2.0–15	
Cd	0.28	0.26	0.07–0.72	0.43	0.52	0.65	0.1–0.5	0.69
Cr	9.8	8.5	3.1–39.3	15.4	21.3	30.2	0.5–10	32
Co	1.44	1.10	0.06–7.80	2.68	4.10	5.28	0.3–2	
Cu	10.3	9.3	1.48–32.0	17.3	22.5	29.1	3.0–15	15
Pb	21	18	3.2–191	34	46	104	5–40	43
Mn	151	131	18.3–513	265	295	465	10–300	
Ni	3.1	2.5	0.19–14.4	5.3	7.1	11.3	0.5–10	8.2
Zn	37	32	12.7–207	59	71	118	25–70	59

[a] De Temmerman *et al*., 1984.
[b] VLAREBO, 1996 (Average 2% clay; 3.9% OM).

are defined as a 90 percentile of the contents in 'background' samples and not as an average value (VLAREBO, 1996).

The background values for arable soils represented in the Tables I–IV are calculated on the basis of the average content of clay of the texture class considered and average content of organic matter. As such they have only and indicative value. The average clay content was derived from the texture triangle for each texture class. For individual samples it is necessary to know exactly the clay and organic matter content in order to be able to calculate the real background or threshold limit for arable soils.

The occurrence of enriched soils becomes clear by comparing the average and median values. Indeed a group of undisturbed soils will show a normal frequency distribution of the heavy metal contents and consequently an equal mean and median value. The higher percentiles show the occurrence of high values due to inputs from external sources. Frequency distribution graphs were drawn and normal distribution was tested at a 95% significance level (One Sample Kolmogorov-Smirnov test) by using Unistat 4.5, Statistical Package for Windows.

The discussion of results is done by major soil texture class. However, differences can occur per province. The province of Antwerp is characterised mainly by sandy soils; the Flemish part of Brabant by sandy-loam and loam soils. On the other hand, in West- and East-Flanders and in Limburg all texture classes occur from sandy soils in the north to loam soils in the south. As industrial sources of heavy metal pollution occur mainly in the north of the provinces East-Flanders,

TABLE II

Correlation matrix for sandy soils and sandy-loam soils

Sandy soils – ns: not significant; r > 0.09 for $P = 0.1$; r > 0.11 for $P = 0.05$; r > 0.16 for $P = 0.01$

	As	Be	B	Cd	Cr	Co	Cu	Pb	Mn	Ni	Zn	C	P	K	Mg	Ca	Na
As		0.36	0.40	ns	0.51	0.54	0.16	ns	0.27	0.46	ns	ns	–0.19	0.20	0.16	0.23	ns
Be	0.35		0.15	0.24	0.37	0.51	ns	ns	0.25	0.55	0.13	ns	ns	ns	0.42	0.45	ns
B	0.48	0.11		ns	0.80	0.31	ns	–0.10	ns	0.20	ns	ns	ns	0.15	ns	ns	–0.11
Cd	0.39	0.34	0.23		0.18	0.12	ns	0.16	0.31	0.15	0.35	0.34	ns	ns	ns	ns	ns
Cr	0.63	0.43	0.49	0.39		0.59	ns	ns	0.37	0.50	ns	ns	–0.13	0.16	0.11	0.11	ns
Co	0.36	0.56	0.19	0.45	0.61		0.18	ns	0.47	0.73	0.17	–0.24	ns	0.26	0.30	0.25	ns
Cu	–0.09	ns	–0.09	–0.17	ns	ns		0.24	ns	0.13	0.32	ns	0.61	0.54	0.22	ns	0.13
Pb	0.11	ns	ns	ns	ns	ns	0.36		0.12	0.11	0.14	ns	0.15	0.17	ns	ns	ns
Mn	0.31	0.27	0.21	0.46	0.52	0.65	–0.12	0.08		0.50	0.23	–0.21	ns	0.12	0.16	0.15	ns
Ni	0.53	0.63	0.26	0.52	0.71	0.80	–0.10	ns	0.56		0.19	–0.24	ns	0.26	0.47	0.48	0.19
Zn	0.17	0.27	0.19	0.38	0.31	0.44	0.24	ns	0.39	0.37		ns	ns	0.26	0.14	ns	ns
C	ns	0.09	ns	0.12	ns	–0.11	0.12	0.17	–0.22	ns	0.15		ns	–0.10	ns	ns	ns
P	–0.26	–0.17	–0.08	–0.34	–0.21	–0.25	0.58	–0.05	–0.31	–0.33	0.11	0.15		0.62	0.33	0.17	0.22
K	ns	–0.21	ns	–0.15	0.09	ns	0.45	ns	ns	ns	0.21	0.15	0.66		0.40	0.18	0.34
Mg	0.27	0.39	ns	0.08	0.26	0.24	0.16	ns	–0.14	0.31	0.12	0.21	0.26	0.34		0.76	0.44
Ca	0.48	0.52	0.17	0.22	0.43	0.32	–0.11	–0.13	ns	0.48	0.11	0.10	ns	ns	0.70		0.40
Na	0.09	0.13	ns	ns	0.10	0.12	0.35	ns	–0.18	0.17	0.08	0.14	0.42	0.37	0.50	0.41	

Sandy loam soils – ns: not significant; r > 0.07 for $P = 0.1$; r > 0.10 for $P = 0.05$; r > 0.14 for $P = 0.01$

TABLE III

Trace element contents in sandy-loam soils in mg kg^{-1} DM – 274 samples

Element	Aver.	Median	Min–max	Percentiles			'Normal' range[a]	Background value[b]
				90 pct	95 pct	99 pct		
As	5.8	5.2	2.2–19.6	8.7	10.1	14.3	1–10	17–20
Be	0.20	0.15	<0.03–1.00	0.44	0.53	0.76	0.02–0.5	
B	2.8	2.6	<0.1–25.4	4.8	5.7	10.4	5–30	
Cd	0.30	0.28	0.08–1.33	0.43	0.51	0.75	0.1–0.5	0.73–0.88
Cr	14	14	4.4–30	18	20	25	1–20	35–38
Co	3.8	3.7	0.15–8.9	6.0	6.5	8.3	0.3–5	
Cu	12	11	2.5–43	19	23	34	5–20	16–17
Pb	21	19	10–83	29	38	65	5–40	40–42
Mn	302	292	47–619	462	480	524	100–500	
Ni	7.6	6.9	1–19.1	12.7	13.8	17.4	1–25	8.5–9.5
Zn	49	45	16.8–209	67	79	118	25–100	60–65

[a] De Temmerman *et al*., 1984.
[b] VLAREBO, 1996 (Average 6–11% clay; 2.6% OM).

TABLE IV

Trace element contents in loam soils in mg kg^{-1} DM – 115 samples

Element	Aver.	Median	Min–max	Percentiles			'Normal' range[a]	Background value[b]
				90 pct	95 pct	99 pct		
As	6.8	6.6	3.6–21.9	8.2	10.9	15.9	5–12	21
Be	0.31	0.29	<0.03–0.090	0.57	0.63	0.83	0.05–1.2	
B	3.7	3.7	<0.1–15.2	5.8	6.3	8.5	10–45	
Cd	0.36	0.35	0.11–0.66	0.52	0.57	0.60	0.1–0.5	0.95
Cr	18	18	8.1–36	22	25	33	5–30	39
Co	5.6	5.7	0.15–8.9	7.7	8.3	8.8	1–10	
Cu	12	11	5.7–88	16	19	33	2–25	18
Pb	21	19	9.6–106	27	33	63	15–40	43
Mn	403	398	129–711	520	534	613	200–600	
Ni	12	12	4.2–21	17	18	20	5–25	10.0
Zn	64	54	28–264	86	138	216	50–100	68

[a] De Temmerman *et al*., 1984.
[b] VLAREBO, 1996 (Average 14% clay; 2.6% OM).

Antwerp and Limburg, a comparison shows clearly the influence of atmospheric deposits of heavy metals (De Temmerman *et al.*, 2000).

3. Results and Discussion

The results have been grouped per major texture class in sandy soils, sandy-loam soils, loam soils and clay soils and are discussed per group.

3.1. Sandy soils

The sandy soils consist of the texture classes course sand and fine sand. On average the pH is rather low (5.5) and ranging from 3.8 to 7.6. The average and range (minimum–maximum) of organic carbon was: 2.3% (0.8–3.8) and of several nutrient elements such as P: 354 mg kg^{-1} (50–1130), K: 157 mg kg^{-1} (30–420), Mg: 95 mg kg^{-1} (10–720), Ca: 1010 mg kg^{-1} (80–15 700) and Na: 13 mg kg^{-1} (1–65). The results of the analysis of sandy soils for minor elements are shown in Table I. On average the As and Be contents of the soils were normal, but 10% of the soils showed an increased content compared to the normal range. However for As only the 99-percentile was higher than the calculated background value. The As and to some extent also Be enriched soils were all found in the provinces of Antwerp, Limburg and East-Flanders and can be linked to the occurrence of lead, copper and zinc smelters (De Temmerman *et al.*, 2000).

Several sandy soils showed small B contents that could probably cause problems of deficiency for some crops. This confirmed the findings of Vanongeval *et al.* (1996). In some cases there was also a clear enrichment, which could be due to air pollution and to the application of boron fertiliser. In general, the Cd contents in arable sandy soils were rather small and were rarely above the calculated background value. This is rather surprising because most of the present and historical industrial activities releasing cadmium are located in those areas. Apparently there was not much long-term accumulation in such well-drained and mostly acidic soils. From the frequency distribution, however, it appears that there was a number of soils clearly enriched with Cd, as there was no normal distribution (Figure 1). In West-Flanders, where no known industrial sources of Cd occur, the contents showed a normal distribution with an average and median value almost the same at 0.18 mg kg^{-1} (De Temmerman *et al.*, 2000). From those results it is obvious that other sources (fertilisers and manure) have no clear impact on the Cd contents of arable soils. Chromium and Co showed slightly increased levels, chromium being in most cases below the calculated background value. The contamination of both elements was primarily located in the provinces of Antwerp and Limburg and can be linked to industrial sources (De Temmerman *et al.*, 2000).

The Cu background levels were clearly increased; the 90-percentile was higher than the calculated background value. That means that more than 10% of the

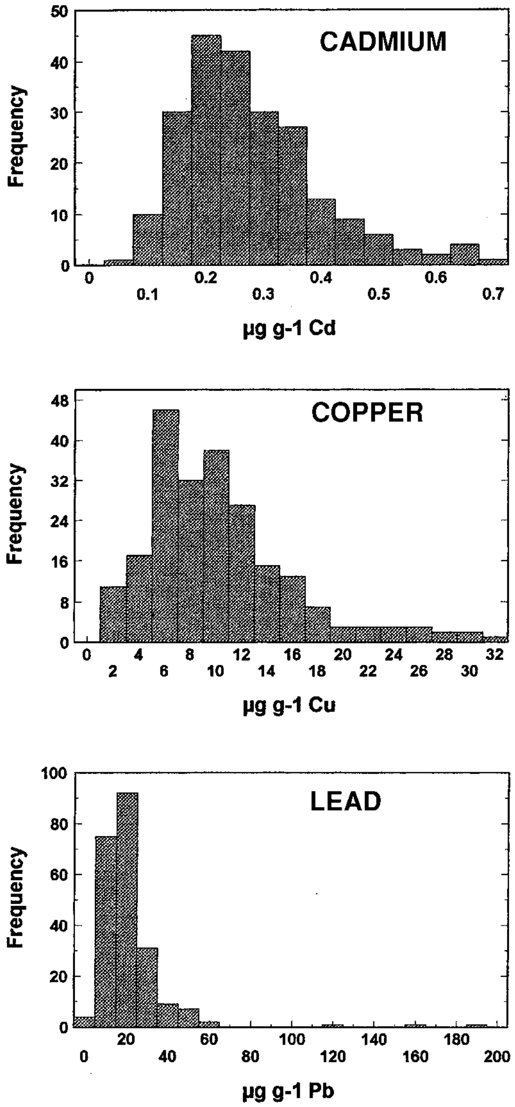

Figure 1. Frequency distributions of the cadmium, copper and lead contents in sandy soils. Total number of samples: 225. The cadmium, copper and lead contents are not normally distributed; null hypothesis rejected at 95% significance level.

soils investigated had an increased Cu content. This is clearly illustrated by the frequency distribution for Cu (Figure 1). A very limited number of soils showed increased Pb levels (Figure 1). This was indicated by the non normal distribution and by the difference between the average and the median value. The 95-percentile was above the background value. The contaminated soils were found in the province of Antwerp where historical pollution and mid-range transport are the source, and in

West-Flanders where the source was not clear. The Mn and Ni values were, on average, rather normal but in both cases, contents higher than the normal range can occur. This was also the case for Zn. The 90-percentile was equal to the calculated background value. Zinc enrichment was primarily found in Limburg and can be related to the historically pollution from the zinc smelters. An increase above background was observed in West-Flanders, which could have been due to the extensive use of liquid manure (pig slurry). In addition, the Cu and Zn contents were well correlated in that province (Table II).

The industrial impact due to mid-range transport of heavy metal-containing dust is illustrated by the good correlation among the elements As, Be, Cr, Co, Ni and to some extent also with B and Mn (Table II). Several of these elements have common sources. Zn and Cd were less strongly correlated in spite of emissions from zinc smelters over many decades. As those factories are located in an area with very poor sandy soils, a large part of the deposits are likely to have been transported to deeper soil layers.

In addition to industrial development, an intensive animal husbandry industry has developed in those areas with poor sandy soils, producing a large amount of animal manure per ha of agricultural land. It is not surprising that the enhanced Cu levels in those soils were well correlated with P and to some extent also with Zn. A good correlation between P and Cu was not always obvious, as extractable P was not the best indicator of long-term accumulation in arable soils. The total content is a better reflection of the history of the soil (Vanongeval *et al.*, 1995).

In general, Pb was poorly correlated with most of the other elements in spite of the occurrence of some common industrial sources. The use of Pb in fuel has made it the most widespread pollutant.

3.2. Sandy loam soils

The sandy loam soils consist of texture classes of loamy sand, light sand-loam, sand-loam and light loam. On average the pH was 6.1 ranging from 3.9 to 7.8. The average and range (minimum–maximum) content of carbon was: 1.5% (0.7–2.7) and of several nutrient elements such as P: 298 mg kg^{-1} (60–990), K: 187 mg kg^{-1} (60–590), Mg: 138 mg kg^{-1} (30–670), Ca: 1730 mg kg^{-1} (320–13 450) and Na: 17 mg kg^{-1} (2–58).

The results on minor elements in sandy loam soils are summarised in Table III. In spite of the slightly increased contents, the As concentrations fitted well within the normal range. The soil contents were always smaller than the calculated background value. The Be contents were, in some cases, slightly increased compared to the normal range. This occurred primarily in the provinces of East-Flanders and Limburg. Beryllium was correlated with Cd, Cr, Co, Mn, Ni and Zn (Table II). The B contents were in the normal range, but as with the sandy soils, many of the sandy-loam soils were poor in boron and there was a risk of deficiency for some crops.

The Cd contents were comparable to those in sandy soils. In some cases the levels were increased, and some soils could be considered as contaminated in a very few cases, as shown in the frequency distribution (Figure 2). Locations with slight contamination were primarily found in the provinces of Limburg and Brabant. Chromium contents were normal and were not larger than the calculated background. In general Co showed a slightly increased, and Cu a markedly increased level (Figure 2). The 90-percentile was higher than the calculated background value. The increase of the Cu content was primarily clear in West-Flanders where it can be attributed to the use of large amounts of manure in the past (De Temmerman *et al.*, 2000). The same conclusion could be drawn for Zn as both elements were well correlated (Table II).

The Pb and Mn contents fitted well within the ‘normal’ range. However, from the frequency distribution, it can be seen that a limited number of soils showed increased contents above the background (Figure 2). For Ni, all the data fitted well in the ‘normal’ range determined before (De Temmerman *et al.*, 1984). However, a lot of samples were above the calculated background.

In general, the elements As, Be, Cr, Cd, Co, Mn, Ni and Zn were correlated with each other and in most cases they were also correlated with B (Table II). Lead was not correlated with most of the other elements except Cu, indicating that primarily other sources of pollution played a role. This was also the case for Cu: it was well correlated with Pb and Zn, and to some extent with Mn and As. The correlation with Zn indicated that extensive use of animal manure played an important role in the enrichment of soils. However, important common industrial sources also exist for both elements. Remarkably, the Cd content was not linked to that of Cu. It was thought that the use of animal manure might have also increased the Cd levels. However, this was clearly not the case, and could be proven by a more detailed analysis of the data obtained in West-Flanders, an area without important industrial sources of Cd but with an important development of intensive animal husbandry (De Temmerman *et al.*, 2000).

Copper and Zn contents were also correlated with carbon and Cu was well correlated with extractable P and Na. These are important indications that the increase in Cu content was linked to the use of animal manure. Be, Cd, Cr, Co, Ni and Zn were well correlated with Mg and Ca. For some of these elements, this is an indication that an increase in the pH reduced the leaching of these elements.

3.3. Loam soils

The loam soils sampled have on average a pH of 6.5, ranging from 4.4 to 8.0. The average and range (minimum–maximum) content of carbon was: 1.5% (0.6–2.5) and of several nutrient elements such as P: 241 mg kg^{-1} (60–490), K: 186 mg kg^{-1} (90–350), Mg: 136 mg kg^{-1} (40–640), Ca: 2070 mg kg^{-1} (550–8700) and Na: 20 mg kg^{-1} (4–32). In general the loam soils showed normal contents of heavy metals, as there are no known major pollution sources in these areas (Table IV).

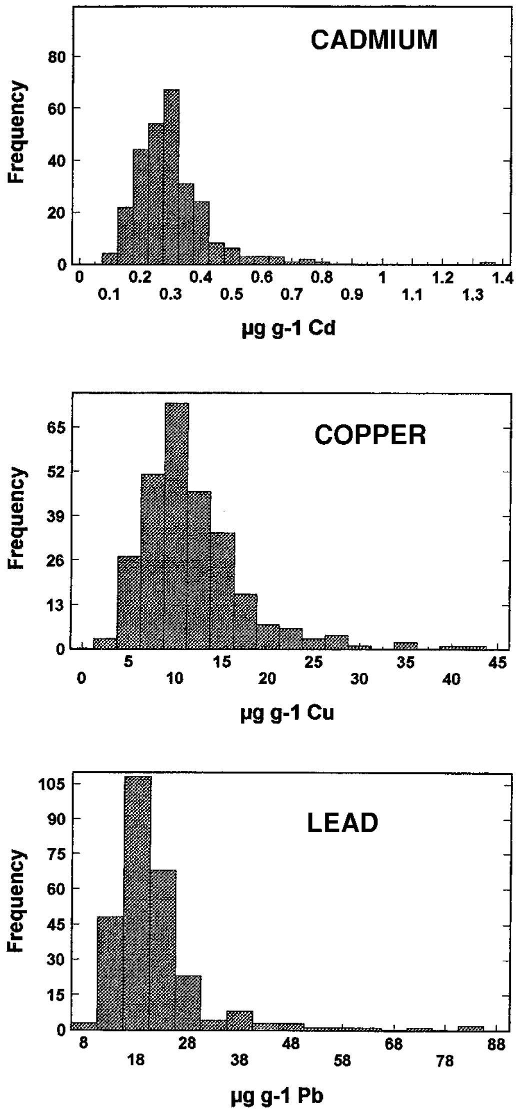

Figure 2. Frequency distributions of the cadmium, copper and lead contents in sandy-loam soils. Total number of samples: 274. The cadmium, copper and lead contents are not normally distributed; null hypothesis rejected at 95% significance level.

The As contents were in less than 5% of the samples, above the 'normal' ranges and were mostly below the calculated background value. Beryllium and B contents were normal; the latter being fairly small in some cases. Cadmium was below the calculated background level but, especially in Limburg, there was a clearly increased concentration (De Temmerman *et al.*, 2000). This could be due to a mid-range transport of Cd (and Zn) containing dust particles originating from industrial activities in the Meuse valley around Liège. However, in spite of the increased level in Limburg, the general Cd contents show a normal frequency distribution (Figure 3). Chromium had a perfectly normal range but the Co contents were slightly increased. The Cu and Pb contents were mostly in the normal range, however, in more than 5% of the cases they were larger than the calculated background. Also Mn and Ni were mostly in the normal range but Ni was in some cases, above the calculated background.

The same correlations occurred in the loam soils as for the sandy-loam soils (Table V) i.e. As, Be, Cr, Cd, Co, Mn, Ni and Zn were well correlated with each other. Lead was correlated with Cd but Cu did not show any positive correlation with any of the other elements. However, there were correlations with the P, K and Mg contents, but not with organic carbon and Na. Only in a very few cases, an influence of liquid manure use could have influenced the soil contents.

3.4. Clay soils

The clay soils consist mainly of polder clay. The pH is rather high, on average 7.1 ranging from 4.9 to 7.7. The average and range (minimum–maximum) content of organic carbon was: 1.9% (1.2–2.9) and of several nutrient elements such as: P: 355 mg kg^{-1} (80–910), K: 224 mg kg^{-1} (50–470), Mg: 393 mg kg^{-1} (50–650), Ca: 11780 mg kg^{-1} (590–36 600) and Na: 55 mg kg^{-1} (14–108).

For the clay soils, the obtained results were all in the normal range and below the calculated background value. The exception was Ni, but for this element the calculated background seemed to be unrealistic (Table VI). The As and Be contents were within a normal range or were slightly increased.

The elements Cr, Co, Mn, Ni, Zn and Cu were correlated with each other and some of these elements showed good correlations with As, B and Be (Table V). There might be a geochemical relationship in the parent material. Cadmium showed completely normal contents and was not correlated with any of the other elements while lead, showing some slight increase in the background, was only correlated with copper. Arsenic and B showed a correlation with the organic carbon content, and none of the elements were correlated with P. An influence of liquid manure could not be shown here.

4. Conclusions

The arable soils that were not in the vicinity of sources of heavy metal containing dust, showed rather normal values of heavy metal contents. However for most of the elements there was some slight increase above the background values and in a very few cases the soils can be considered as slightly contaminated. The influence of industrial activities on the soil contents was clearly demonstrated. In some regions, the Cu and Zn contents were slightly raised, possibly due to excessive

TABLE V

Correlation matrix for loam and clay soils

Loam soils – ns: not significant; r > 0.12 for $P = 0.1$; r > 0.15 for $P = 0.05$; r > 0.21 for $P = 0.01$

	As	Be	B	Cd	Cr	Co	Cu	Pb	Mn	Ni	Zn	C	P	K	Mg	Ca	Na
As		0.34	0.61	0.28	0.61	0.33	ns	ns	0.24	0.34	0.31	ns	ns	ns	0.13	0.45	ns
Be	ns		0.28	ns	0.22	0.49	–0.19	ns	ns	0.53	0.47	0.22	ns	0.14	0.40	0.49	ns
B	0.46	–0.45		0.23	0.52	0.20	–0.19	ns	0.22	0.30	0.26	0.18	ns	0.13	ns	0.21	ns
Cd	ns	ns	ns		0.28	0.33	ns	0.16	0.41	0.31	0.39	0.14	ns	0.14	–0.13	0.14	ns
Cr	0.48	ns	0.65	ns		0.54	ns	ns	0.48	0.62	0.27	0.27	ns	0.18	ns	0.37	ns
Co	0.65	0.39	0.42	ns	0.81		ns	ns	0.36	0.77	0.43	0.23	–0.19	ns	0.19	0.33	ns
Cu	ns	0.37	ns	ns	0.37	0.53		ns	–0.16	ns	ns	ns	0.19	0.14	0.21	ns	ns
Pb	ns	ns	ns	ns	ns	ns	0.54		0.18	ns	ns	ns	–0.13	ns	ns	ns	ns
Mn	ns	ns	ns	ns	0.40	0.48	0.39	ns		0.26	0.29	ns	ns	0.15	–0.28	ns	ns
Ni	0.58	0.36	0.46	ns	0.80	0.80	0.49	ns	0.61		0.43	0.24	–0.22	ns	0.19	0.46	ns
Zn	0.47	ns	0.44	ns	0.74	0.65	0.63	ns	0.66	0.84		0.13	ns	ns	ns	0.25	ns
C	0.61	ns	0.50	ns	ns	ns	ns	ns	ns	ns	ns		0.24	0.29	0.19	0.22	0.17
P	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns		0.67	0.19	0.13	0.14
K	ns	ns	ns	ns	ns	ns	ns	–0.43	ns	ns	ns	ns	0.48		0.19	ns	0.27
Mg	0.59	ns	0.38	ns	0.35	ns	ns	ns	ns	0.44	ns	0.46	ns	ns		0.48	ns
Ca	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	0.55		0.25
Na	ns	ns	0.38	ns	ns	ns	ns	ns	ns	0.36	0.35	ns	0.40	ns	0.67	0.62	

Clay soils – ns: not significant; r > 0.34 for $P = 0.1$; r > 0.41 for $P = 0.05$; r > 0.51 for $P = 0.01$

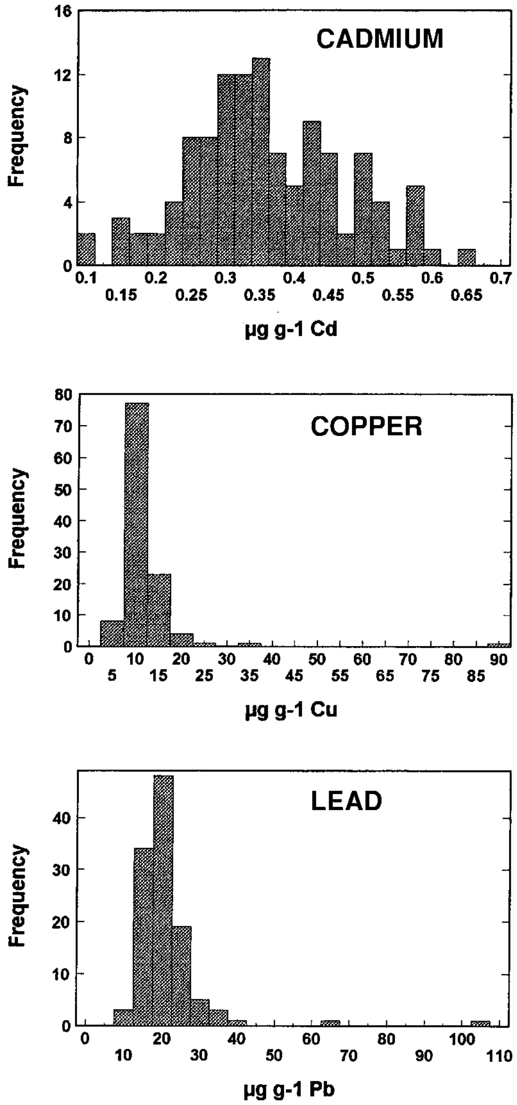

Figure 3. Frequency distributions of the cadmium, copper and lead contents in loam soils. Total number of samples: 115. The cadmium, contents are normally distributed; null hypothesis accepted at 95% significance level. The copper and lead contents are not normally distributed; null hypothesis rejected at 95% significance level.

fertilisation with animal manure in the past. However, none of these soils can be considered as really contaminated. In spite of the very long history of pollution in the northern part of Flanders (Kempen area), the increase in the soil concentration (at more than 20 km from known sources) was rather limited for elements such as Cd and Zn. Apparently, the well-drained acidic sandy soils do not allow much accumulation of these elements.

TABLE VI

Trace element contents in clay soils in mg kg^{-1} DM – 24 samples

Element	Aver.	Median	Min–max	Percentiles			'Normal' range[a]	Background value[b]
				90 pct	95 pct	99 pct		
As	8.2	7.7	2.6–12.4	10.9	11.2	12.1	5–12	27
Be	0.35	0.24	<0.03–1.33	0.98	1.10	1.28	0.05–1.2	
B	9.2	9.1	2.6–15.5	14.0	14.5	15.3	10–45	
Cd	0.26	0.28	0.09–0.45	0.32	0.37	0.43	0.1–0.5	1.20
Cr	18	19	7.2–30	26	28	30	5–30	46
Co	5.1	5.3	0.91–8.3	7.7	7.8	8.2	1–10	
Cu	8.9	8.9	2.4–15.5	12.2	12.7	14.9	5–25	22
Pb	18	17	7.9–58	22	22	50	15–40	44
Mn	251	228	143–471	398	431	462	200–600	
Ni	12	12	3.3–19	17	18	19	5–25	12.0
Zn	48	49	21.5–78	63	68	76	20–100	78

[a] De Temmerman *et al.*, 1984.
[b] VLAREBO, 1996 (Average 25% clay; 3.3% OM).

In general, loam and clay soils showed rather normal contents. In the area where these soils were sampled there are only a few minor industrial sources and also less manure was used in the past. However, in the south eastern part of Flanders (province of Limburg), some slight Cd and Zn enrichment was noticed, probably due to mid or long-range transport originating from sources in the Meuse valley around Liège.

Acknowledgements

The authors are grateful to Paul Coosemans for carrying out the analytical work.

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