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Heavy-Metal Contents of Dutch Arable Soils

By W. VAN DRIEL and K. W. SMILDE*)

Introduction

In densely populated countries like The Netherlands, gradual pollution of the environment makes it necessary to determine the current, normal levels of trace elements in the soil. In particular, it is important to control the levels in arable soils, used for the production of food for human and animal consumption, to prevent accumulation of hazardous elements to intolerable concentrations. At the Institute for Soil Fertility two actions have been undertaken for establishing background levels of heavy metals in the soil:

1. A selection was made from the large collection of soil samples of the Institute. As most soil types are represented in this collection, only restricted additional sampling was required to obtain a fair survey of the levels of trace elements. The selected soil samples originate from the period 1960-1980; sampling depth was 0-20 cm and the elements analyzed were As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb and Zn. The general soil characteristics were also investigated: organic matter content, % fraction < 16 μ m, pH, and CaCO₃ content. About 1000 samples were analyzed for these constituents. The results of this survey will be presented in this paper.

2. In 1978 a survey of the concentrations of four toxic elements (As, Cd, Hg, Pb) in Dutch crops and soils was started. Apart from arable crops, some glasshouse crops and fodder crops have been also included in this survey. The results of this survey will be published within a few years.

Soil Types

In the Netherlands nearly all soils originate from materials which, after erosion of the bedrock, were transported by fluvial, glacial or aeolian processes. In the genesis of most Dutch soils, human impact has played an essential part. Already in the middle-ages the rivers Rhine and Meuse were dammed to protect the adjacent areas from flooding. The extensive fen-peat deposits in the western and northeastern parts of the country were dug out.

The resulting lakes and pools have been partly reclaimed. In the eastern part of the country the top layer of the original peat moss was mixed with the underlying sandy soil, resulting in the 'formation' of sandy soils with high organic-matter contents. Pleistocene, sandy soils (cover sands) have been improved locally with farmyard manure and heather sods, resulting in a 'black' layer of up to one metre.

About forty percent of the land lies below the present mean sea level, and would be inundated but for the protection by dikes. It is clear that the levels of trace elements in the Dutch arable soils have been largely dependent on human activities during the last ten centuries.

In this paper we distinguish five main soil types: sandy soils, clay soils (> 16 μ m < 12 %), fen-peat soils, peat moss/sand mixtures and loess soils. The data available do not provide sufficient information to distinguish more detailed subtypes:

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Materials and Methods

Sampling

To determine the normal levels of trace elements in Dutch soils, samples have been selected from soils all over the country. The samples come from the 0–20 cm layer of arable soils, each composed of about 40 subsamples per location. The fen-peat soil samples mainly originate from the 0–20 cm layer of grassland, as no arable peat soils were available. The samples were dried at 30–40°C; the 0–2 mm fraction was used for analysis. During treatment of the samples due care was taken to avoid metal contamination.

Methods

Organic matter, pH, fraction < 16 μm , and CaCO_3 were estimated according to the usual procedures of the Institute (VIERVEIJZER et al., 1979).

The metals Cr, Cu, Fe, Mn and Zn were analyzed by flame atomic absorption spectrometry after digestion with $\text{H}_2\text{SO}_4\text{-HNO}_3$ 1:1. For analysis of Cd, Ni and Pb, samples were digested with HNO_3 (five times fuming to dryness) and the elements determined in methylisobutylketone environment with flame atomic absorption spectrometry, after liquid-liquid extraction with sodium diethyldithiocarbamate, according to the procedure developed by BALRAADJISING (1974).

The elements As, Hg and Sb were determined by neutron activation analysis at the Central Laboratory TNO, Delft, according to the procedure of TJIÖE et al. (1976).

The digestion methods used do not yield total metal contents in the samples. The small difference between the actual content and the content obtained with concentrated acid extraction is due to differences in the mineral composition of the samples. The undissolved fraction of the elements is considered less relevant in terms of biological availability. In this paper we will use the determined metal contents as if they were total contents. All results are based on dry weight (105°C),

Results and Discussion

In this section a summary of the analytical results will be given. As we concentrate on the elements that are important to the environment, the results for iron and manganese will be omitted, as well as the data on the general soil properties.

Tab. 1
Mean heavy-metal contents of Dutch arable soils
Mittlere Schwermetallzusammensetzung von niederländischen Böden
($\text{mg} \cdot \text{kg}^{-1}$)

Element (Element)	Clay Soil (Tonböden)	Sandy soil (Sandböden)	Fen-peat soil (Niedermoor)	Moss peat/ sand mixtures (Sandgemischtes Hochmoor)	Loess soil (Lößböden)
Zn	117	44	101	25	86
Cu	23	11	28	21	13
Cr	78	26	63	20	68
Ni	33	5	26	3	13
Pb	43	31	71	32	30
Cd	0,5	0,3	0,9	0,3	0,9
Hg	0,2	0,2	0,2	0,1	0,1
As	14	5	12	2	8
Sb	0,8	0,4	1,6	0,4	0,7
n =	248	63	40	22	8

Table 1 presents a survey of the mean contents of As, Cd, Cr, Cu, Hg, Ni, Pb, Sb and Zn in apparently uncontaminated, arable soils belonging to the five soil types mentioned before. Comments will be given in the sections for the separate soil types.

Clay soils

The frequency distribution of nine elements in clay soils is shown in Figure 1. Dutch clay soils are rather diverse in age and origin. Five different areas can be distinguished: In the northern, western and southwestern parts of the country marine clay is predominant. In some reclaimed lakes old marine clays occur. In the centre of the country, the basin of the rivers Rhine and Meuse, young fluvial clay soils occur.

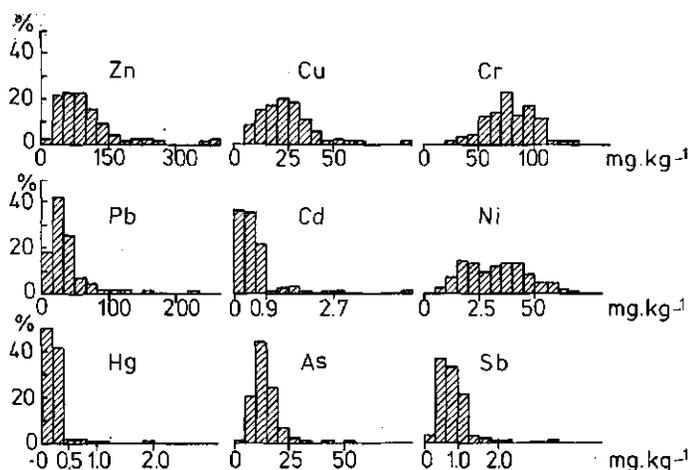


Fig. 1

Frequency distribution of nine elements in 248 Dutch clay soils
Häufigkeitsverteilung von neun Elementen in 248 Niederländischen Tonböden

The interpretation of the results is hindered by two factors. The preferential sorption of As and the heavy metals by the finer soil particles results in a linear relationship between the (total) metal contents of the sample and the % fraction $< 16 \mu\text{m}$ (DE GROOT, 1964). In the same area, a lighter clay soil will have lower metal contents than a heavier one. This can be overcome by normalizing the contents to a constant particle size composition (e.g. 50% $< 16 \mu\text{m}$). As this was not possible in this study, the range of metal concentrations is relatively large.

The second factor is the effect of the strongly polluted rivers Rhine, Meuse and Scheldt. In polders reclaimed in the 18th and 19th century, occasionally higher metal concentrations have been found, resulting from flooding in recent times at extreme tide levels. The higher values (e.g. $> 140 \text{ mg/kg Zn}$) in the frequency distribution may be explained by this phenomenon. If the data from these suspectedly polluted areas are excluded, the mean values for clay soils presented in Table 2 are obtained. Apart from these effects, the heavy metal levels in the five areas do not differ significantly, in spite of the large variations in age and origin. The mean heavy-metal levels in Dutch clay soils are of the same order of magnitude as those in fossil shales (Tab. 3) (TUREKIAN and WEDEPOHL, 1961).

Tab. 2
Effect of local pollution on the mean element contents of clay soils
Effekte von regionaler Kontamination auf mittlere Elementgehalte von Tonböden
 (mg.kg⁻¹)

Element (Element)	Mean values including suspected areas (Mittlere Werte inklusiv verdachte Gebiete)	Mean values without suspected areas (Mittlere Werte ohne verdachte Gebiete)
Zn	117	76
Cu	23	20
Cr	78	73
Ni	33	26
Pb	43	30
Cd	0,5	0,34
Hg	0,2	0,2
As	14	13
Sb	0,8	0,6

Sandy soils

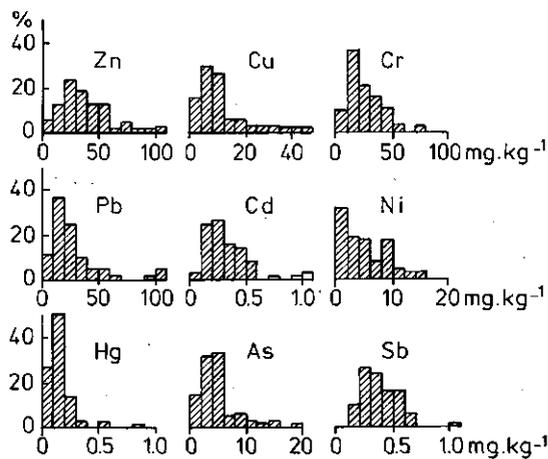


Fig. 2

Frequency distribution of nine elements in 63 Dutch sandy soils
Häufigkeitsverteilung von neun Elemente in 63 Niederländischen Sandböden

The sandy soils (Fig. 2) generally have lower levels of arsenic and heavy metals than the clay soils. The differences among the northern, eastern and southern areas are rather small. In the southern sandy soils enhanced zinc and cadmium levels have been observed in the vicinity of zinc smelters, situated along the Belgian-Dutch frontier. The relatively low levels in this soil type tend to be higher than those in sandstones (Tab. 3).

Fen-peat soils

Most of the fen-peat, originally covering a large part of this country, has been dug off. The remaining peat area is mainly in use as grassland. Two areas are included in this study: the western and the northern (Friesland). In the western area, arsenic and heavy-

metal concentrations are about twice as high as in the northern area. In the frequency distribution pattern (Fig. 3) this results in two distinct peaks for nearly all elements. The levels of the elements are higher than those of the sandy and clay soils. However, because of the low bulk density of peat soils, the concentrations based on equal volumes are about the same.

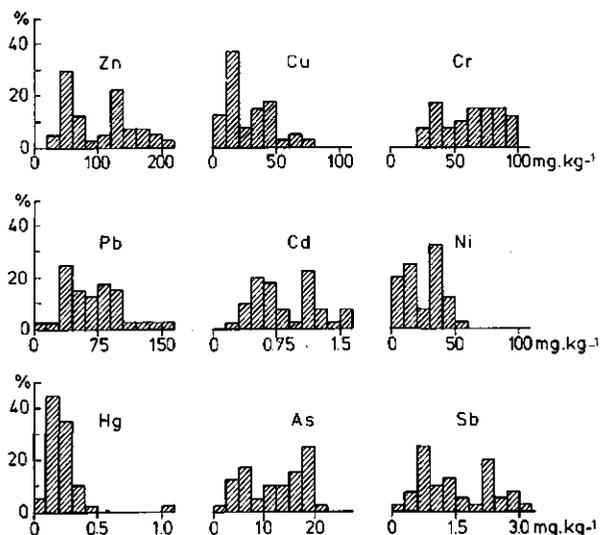


Fig. 3

Frequency distribution of nine elements in 40 Dutch fen-peat soils
 Häufigkeitsverteilung von neun Elementen in 40 Niederländischen Niedermoorböden

Peat moss/sand mixtures

These soils, situated in the northeastern part of the country, are man-made soils: after removal of the peat layer, the top 50 cm layer of the original moss peat soil was mixed with the underlying sand layer, and with sand from the canals. The levels of the elements studied are lower than those of the fen-peat and sandy soils (Fig. 4). This was to be expected because of the extremely low metal contents of the sandy subsoil. In the past, these soils gave serious problems with trace element deficiencies (Cu).

Loess soils

The southeastern part of the country has a limited area of loess soils.

The arsenic and heavy-metal contents are similar to those of the clay soils (Tab. 1).

Comparison of the mean element levels of Dutch soils with reference soils

Table 3 shows the average element concentrations in the earth's crust, shales and sandstones. The mean concentrations found in Dutch arable soils agree with these values. KLOKE (1980) proposed acceptable concentrations of some elements in agricultural soils: As 20, Cd 3, Cr 100, Cu 100, Hg 2, Ni 50, Pb 100, Sb 5, Zn 300 mg·kg⁻¹. The As and heavy-metal concentrations in sandy soils, fen-peat soils and peat moss/sand mixtures do not exceed these levels, apart from some high Pb-levels in sandy and fen-peat soils. Some 5–10 per cent of the clay soil samples, likely to be polluted by flooding, had a Cr-, Ni-, Pb-, or Zn-concentration in excess of the limit suggested by Klope. However, the meaning of KLOKE's critical values for crop quality remains to be verified.

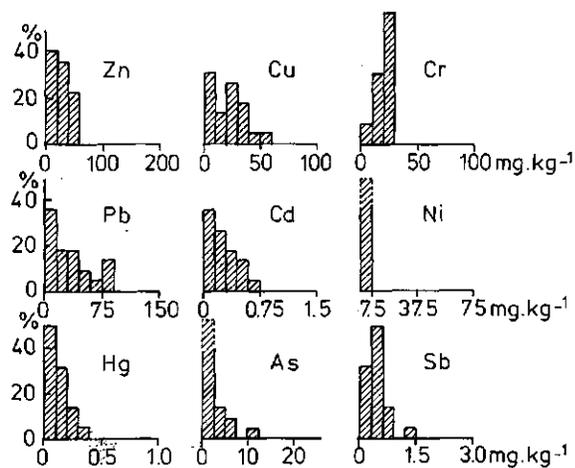


Fig. 4

Frequency distribution of nine elements in 22 Dutch moss peat/sand mixtures
 Häufigkeitsverteilung von neun Elemente in 22 Niederländischen sandgemischten Hochmoorböden

Tab. 3

Heavy metal contents of clay, sandy soils, shales, sandstones and the earth's crust
 Schwermetallzusammensetzung von Ton- und Sandböden, von Sedimentgesteinen, Sandsteinen
 und von der Erdkruste
 (mg.kg⁻¹)

Element (Element)	Shales ¹⁾ (Sediment- gesteine)	Clay soils ²⁾ (Tonböden)	Sandstones ³⁾ (Sandsteine)	Sandy soils ⁴⁾ (Sandböden)	Earth's crust ⁵⁾ (Erdkruste)
Zn	95	117	16	44	83
Cu	45	23	1-9	11	47
Cr	90	78	35	26	83
Ni	68	33	2	5	58
Pb	20	43	7	31	16
Cd	0,3	0,5	0,01-0,09	0,3	0,13
Hg	0,4	0,2	0,3	0,2	0,08
As	13	14	1	5	1,7
Sb	1,5	0,8	0,01-0,09	0,4	0,5

1), 3) TUREKIAN and WEDEPOHL, 1961

2), 4) This study

5) VINOGRADOV, 1962

Polluted soils

As mentioned before, the rivers Meuse, Rhine and Scheldt transport heavily polluted sediments into this country. This is illustrated by comparing the composition of the 0-20 cm layer of polders in the fresh-water tidal area of the rivers Rhine and Meuse, reclaimed in 1759 and 1927, with the composition of the sediments deposited in the same area in 1957 (Tab. 4). The increase in the concentrations of nearly all elements (except nickel) illustrates the dramatic effects of the industrial activities in Western Europe (SALOMONS

and DE GROOT, 1977). This holds especially for the river flood plains, which are flooded at high water tides almost every year. Moreover, river sediments are also deposited in the harbour basins in Rotterdam.

Tab. 4
Heavy-metal contents of polder soils
Schwermetallgehalte von Polderböden
 (mg.kg⁻¹)

Element (Element)	Reclaimed in (Trockengelegt in)		
	1759	1927	1957
Zn	93	460	2070
Cu	29	53	247
Cr	94	112	406
Ni	41	32	58
Pb	31	130	540
Cd	0,3	1,9	13,4
Hg	0,3	0,7	10,0
As	15,4	21,4	144
n =	23	36	24

„Safe“ disposal of the contaminated dredged materials offers serious problems for the harbour authorities. Possible adverse effects of these dredge spoils when used as a substrate for growing crops have been studied extensively at the Institute for Soil Fertility.

Table 5 demonstrates the high metal concentrations as observed in the river flood plains of the three large rivers.

Tab. 5
Polluted soils (river flood plains)
Kontaminierte Böden (Flußauen)
 mg.kg⁻¹

Element (Element)	River (Fluß)		
	Rhine (Rhein)	Meuse (Maas)	Scheldt (Schelde)
Zn	2360	2040	570
Cu	295	191	81
Cr	530	162	139
Ni	68	63	37
Pb	660	610	147
Cd	16	31	3
Hg	12	4	2
As	205	61	51
Sb	11	5	3

Summary

A survey of the heavy-metal levels in Dutch arable soils was made. About a thousand samples, representing the main soil types in The Netherlands and the contaminated areas along the large rivers, were analyzed for the elements As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb and Zn. The results for Fe and Mn are not reported in this paper.

The heavy-metal levels in the clay soils are similar to those of fossil shales. The gradual pollution of the river sediments is demonstrated by comparing soils in polders reclaimed from the fresh-water tidal area in 1759 and 1927 with material deposited in a similar area in 1957. The heavy-metal contents of the sandy soils tend to be higher than those of the sandstones. The fen-peat soils in the northern part of the country have lower levels of the elements studied than those in the west.

It is concluded that there is a similarity in chemical composition between the Dutch arable soils, their parent materials and the earth's crust. Only in recent river sediments, As and heavy-metal concentrations grossly exceeded the levels considered acceptable for cultivated soils.

Zusammenfassung

DRIEL, W. VAN and SMILDE, K. W.: *Heavy-metal contents of Dutch arable soils (Schwermetallgehalte der niederländischen Böden)*.

Landwirtsch. Forsch., Sonderh. 38, Kongreßband Trier 1981

Rund 1000 Bodenproben aus den wichtigsten Niederländischen Kulturböden, und aus dem Flußbereich, sind untersucht worden auf den Gehalt an den Elementen As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb und Zn. Die Proben stammen aus den Jahren 1960–1980. Die Ergebnisse von den weniger umweltbelastenden Metallen Eisen und Mangan werden hier nicht berichtet.

Die mittlere Elementgehalte in Tonböden sind denen in Sedimentgesteinen ähnlich. Die zunehmende Belastung der Flußsedimente durch Schwermetalle wird illustriert durch die Gehalte von Polderböden, eingedeicht in die Jahren 1759 und 1927 zu vergleichen mit denen von Sedimenten aus dem gleichen Bereich in 1957. Die Belastung der Sandböden ist etwas höher wie die der Sandgesteine. Die Schwermetallgehalte von den nördlichen Niedermooren sind niedriger wie den westlichen.

Es wird konkludiert, daß die Niveaus der untersuchte Elemente in den niederländischen Kulturböden nur wenig abweichen von denen der vermutlichen Muttergesteine und der Erdkruste. Nur die rezenten Flußsedimenten überschreiten die Orientierungswerten von KLOKE vielmals.

Résumé

DRIEL, W. VAN and SMILDE, K. W.: *Heavy-metal contents of Dutch arable soils (Les teneurs en métaux lourds des sols arables de la Hollande)*.

Landwirtsch. Forsch., Sonderh. 38, Kongreßband Trier 1981

Environ mille échantillons de sol représentant les principaux types de sols de la Hollande, y compris les bassins pollués bordant les grands fleuves, ont été analysés pour la détermination des éléments suivants: As, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, Sb et Zn. Les données de Fe et Mn ne sont pas présentées ici.

Les teneurs en métaux lourds des sols argileux sont de l'ordre de celles des schistes fossilisés. En comparant la couche supérieure du sol des polders récupérés de l'estuaire des grands fleuves en 1759 et 1927 avec les sédiments récemment déposés (1957) dans les bassins fluviaux, la pollution croissante des sédiments fluviaux saute aux yeux. Les sols sableux contiennent un peu plus de métaux lourds que les grès. Les sols tourbeux de l'ouest sont plus riches en métaux lourds que ceux du nord-est.

Il n'y a pas de grandes différences entre la composition chimique des sols arables et celle de leurs roches mère et de la croûte terrestre. Seulement les teneurs en As et en métaux lourds dépassent considérablement les limites proposées pour les sols cultivés.

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