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Contamination of soil and street dust with lead and cadmium near a lead smelter at Arnhem, Netherlands

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Summary

As part of an epidemiological study, an investigation was carried out on the lead and cadmium pollution of soil and street dust in the vicinity of a secondary lead smelter in the town of Arnhem, Netherlands. Samples of soil and street dust were taken in residential quarters at different distances to the smelter. A significant correlation was found between Pb content of soil and distance to the smelter. In the sampling area 1 km around the smelter the average Pb content in the soil layer 0-5 cm was 370 mg/kg and in the soil layer 5-20 cm 256 mg/kg. Mean Cd contents were 1.60 and 1.28 mg/kg respectively. Correlation between the contents in both layers was low for Pb and high for Cd, due to different degree of solubility of both heavy metals in the soil. A low but significant relation was found between Pb and Cd contents of the soil, suggesting that only part of the metals originated from the same source. Mean heavy metal concentrations in street dust (fraction <0.3 mm) were 761 mg Pb/kg and 3.81 mg Cd/kg. Correlation between both heavy metals in street dust was high, suggesting same sources of pollution (mainly traffic). The most important sources of pollution for Pb and Cd in the studied area were the lead smelter and traffic.

Introduction

Food, ambient air, soil, street dust, house dust, drinking water and paint have all been described as pathways for human lead uptake from the environment. In the vicinity of lead smelters emphasis has been laid on ambient air, soil and house dust (Yankel et al., 1977).

In 1978 an epidemiological study was performed in parts of Arnhem, a town in the Netherlands with a population of ca. 150 000 inhabitants, in order to establish the major pathways for lead uptake by children (aged 1-3 years) living in the close vicinity of a secondary lead smelter. This study revealed the uptake of contaminated soil and dust as the most important contribution (Brunekreef et al., 1981). Main lead pollution sources in the area studied are emissions from the smelter and from traffic. The lead smelter has been in operation for the past 30 years. Until recently the emission of lead into the air amounted to an estimated value of 500 kg per day but now it has been decreased to ca. 50 kg per day. However, lead deposited in the past could still cause future problems as lead tends to be rather immobile in soil. In order to evaluate the possibility of future uptake and to develop means for deminishing this, soil and dust in the surroundings of the smelter were analysed for lead. Traffic being a source for emission of both lead and cadmium, the collected samples were also analysed for cadmium. Results of this survey are presented here. Possible treatments for decontamination of soil polluted with lead will be discussed in forthcoming papers.

Materials and methods

Samples of soil and street dust were collected at different distances from the smelter. For reference purposes sampling was also performed in a residential area of Wageningen, where no industrial air pollution with lead or cadmium occurs and traffic is less intensive.

The survey area in Arnhem, where no predominant wind direction occurs, can be divided into the following parts (cf. Fig. 1):

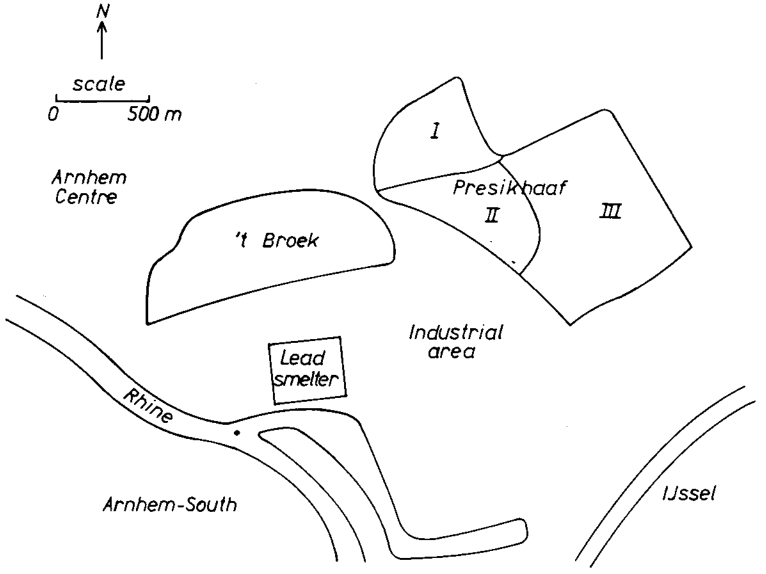

Fig. 1. Sketch-map of smelter and sampling area in Arnhem with location of the residential quarters.

a) area of ca. 1 km around the smelter, including;
b) the residential quarter 't Broek at 500-1000 m north of the smelter;
c) the residential quarters Presikhaaf I, II and III at 1000-2000 m northeast of the smelter;
d) incidental samplings at ca. 2 km from the smelter in northwest, northeast, southeast and southwest directions.

Distribution of samplings was regularly spread over each of the areas but sampling density differed considerably (cf. Table 1).

In the 1-km area, including 't Broek, soil samples were taken at depths of 0-5 and 5-20 cm below surface. In all other areas only the top-layer 0-5 cm was sampled. Each sample, consisting of ca. 1 kg moist soil, was composed of ca. 30 subsamples collected over a surface area of about 10 m^2. After drying at the air the soil was passed through a 2-mm sieve.

In 't Broek and in Wageningen also street dust samples were taken. The dust was collected after sweeping ca. 25 m^2 of street surface for each sample. The dust was dried at the air and passed through a 0.3-mm sieve.

5 g of air-dried, sieved sample were extracted with 40 cm^3 3 *M* HCl for 3 hours in a boiling water-bath. After making up with de-ionized water to 100 cm^3 the extract was filtered through a hard ash-free filter; Pb and Cd were measured in the filtrate by atomic absorption spectrophotometry (AAS). Results are expressed on the basis of air-dry material.

The efficiency of the above extraction procedure was examined by comparison for a number of samples with the results of a digestion with $HClO_4$ and HNO_3 in a ratio of 1:4 by volume. Digestion did not yield significantly higher contents of Pb and Cd.

Results and discussion

Analytical results are presented in Tables 1-3 and Figs. 2-6. The data are given as arithmetic means whereas also the variation is mentioned. These variations are rather high, partly due to inhomogenities of the materials analysed, partly due to different degrees of contamination.

Soil

As is shown in Table 1 the average lead content in the top-layer of the area 1 km around the smelter is elevated as compared to the residential area Presikhaaf and considerably higher than at the town of Wageningen. Uncontaminated soils usually have Pb contents of less than 100 mg/kg (Allaway, 1968). For urban areas values of 100-300 mg/kg have been reported (Roberts et al., 1974; Linzon et al., 1976). Near roads with high traffic density higher values can be found (up to 300-400 mg/kg, or even more). According to Linzon et al. (1976) soils with Pb contents exceeding 600 mg/kg should be indicated as severely polluted. Especially such high levels can contribute to an increased lead level in the blood of little children (Yankel et al., 1977). In the residential areas at greater distance from the smelter (Presikhaaf I, II, III) Pb contents decrease. Wageningen shows

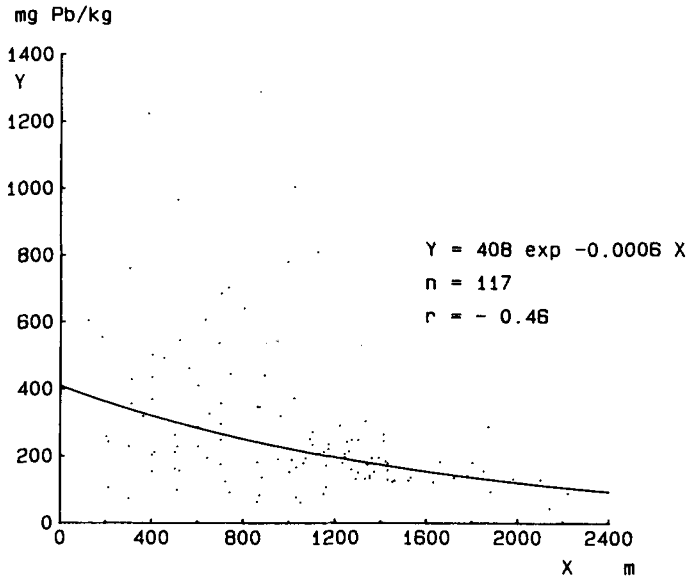

Fig. 2. Lead content (mg/kg) of the soil layer 0-5 cm vs. distance (m) to the smelter.

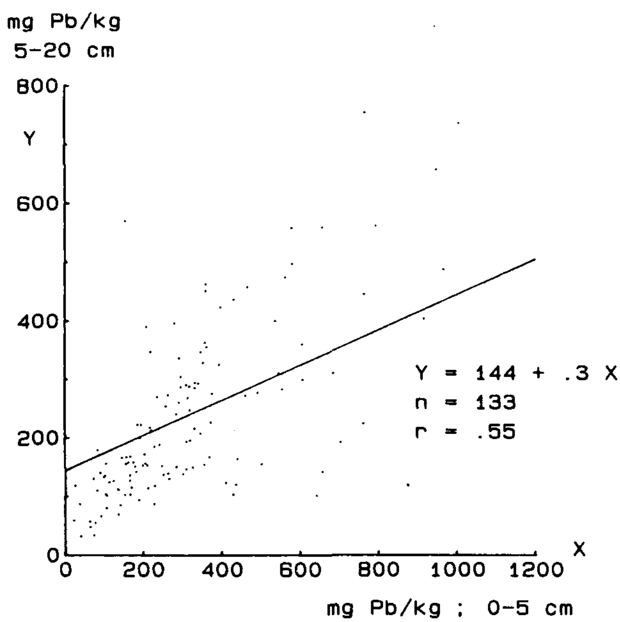

Fig.3. Lead content (mg/kg) of the soil layer 5-20 cm vs. 0-5 cm. Sampling area: 1 km around smelter.

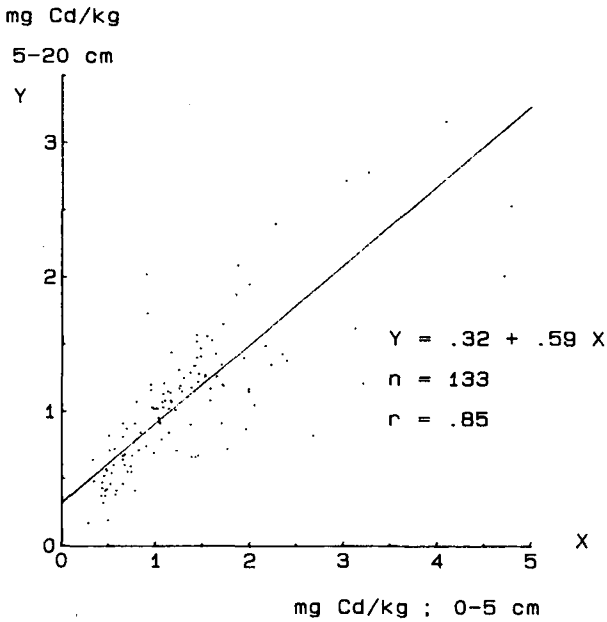

Fig. 4. Cadmium content (mg/kg) of the soil layer 5-20 cm vs. 0-5 cm. Sampling area: 1 km around smelter.

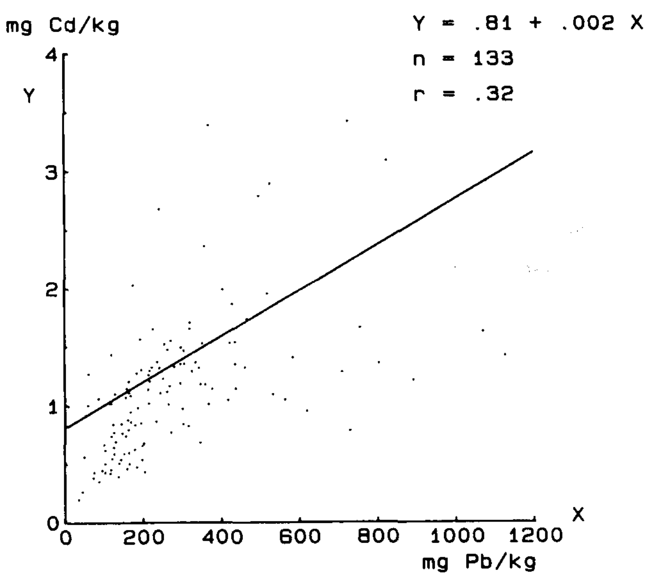

Fig. 5. Cadmium content (mg/kg) vs. lead content (mg/kg) for the soil layer 0-20 cm. Sampling area: 1 km around smelter.

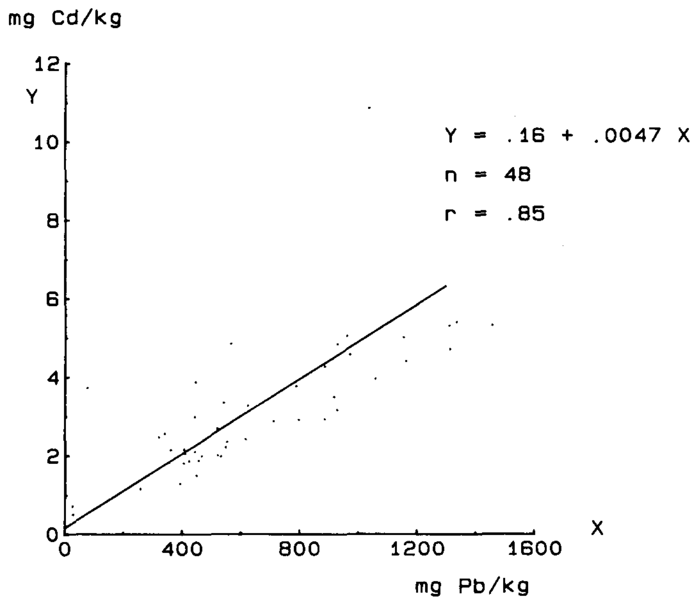

Fig. 6. Cadmium content vs. lead content in street dust (mg/kg). Sampling area: residential quarter 't Broek.

a low average lead content of soil.

From Fig. 2 it appeas that there is a low but significant correlation between lead content of the soil (layer 0-5 cm) and the distance to the smelter ($r = 0.46$ and $P = 0.005$). An exponential fit gave the best results as is often found for this kind of pollution sources. For cadmium no such correlation could be found. The average background concentration for cadmium in soils is ca. 0.15 mg/kg (Page et al., 1972). Close to traffic ways higher values have been measured, up to 1-2 mg Cd/kg (Lagerwerff et al., 1970). Table 2 gives the results for Pb and Cd for the different soil layers in the 1 km area. Also the average value for the 0-20 cm layer, as calculated from the 0-5 and 5-20 cm values (weighted arithmetic mean) is presented. The values are slightly higher than usually found in urban areas. Figs. 3 and 4 present a plot of the top-layer values against the 5-20 cm values for each sampling spot in the 1 km area, for lead and cadmium respectively. These figures give some interesting information about the mobility of both heavy metals in the soils involved. In general solubility of Pb in soils is very low, depending on factors like pH, complexation with dissolved organic matter or Cl^-, and the adsorption capacity of the soil. Fig. 3 still shows a rather good correlation between both layers ($r = 0.55$), probably due to mechanical translocation (biological activity or tillage). Cd solubility in soils tends to be high. The high correlation ($r = 0.85$) resulting from Fig. 4 is ascribed to leaching in addition to mechanical translocation.

From Fig. 5 it appears that a significant although low correlation exists

Table 1. Number of samplings and Pb content (mg/kg) of the soil layer 0-5 cm in different sampling areas.

Sampling area	Number of samples	Arithmetic mean	Range
Arnhem:			
– 1 km around lead smelter	143	370	21-2309
– 't Broek*	71	327	21-1126
Presikhaaf	47	182	93- 303
– Presikhaaf I*	11	164	121- 264
– Presikhaaf II*	27	203	128- 203
– Presikhaaf III*	9	140	93- 182
Wageningen	43	60	27- 117

* Sub-area.

(P = 0.05 and r = 0.32) between Pb and Cd in soil (0-20 cm). Considering this result, in combination with the relationships between Pb and Cd contents and distance to the smelter, it may be concluded that different sources, in part, are responsible for the Pb and Cd contamination. In this case traffic probably contributes to both Pb and Cd, and the smelter to Pb only.

Street dust

Finger-attached street dust, especially the small dust particles, is one of the possible pathways of lead ingestion by little children (Barltrop, 1972; Lepow et al., 1974). Street dust in big cities can contain large amounts of lead. Farmer & Lyon (1977) found an average Pb content of 960 mg/kg with a range of 150-2300. Lepow et al. (1974) mention a mean content of 1200 mg Pb/kg, whereas Roberts et al. (1974) report a value of 1002 mg/kg with a variation of 500-1561. In all these cases the analyses were performed on the fraction $<$2mm. Contents found are dependent on the fraction size studied, since highest concentrations have been found in the smallest fractions.

The mean Pb and Cd contents of street dust in Arnhem were much higher than for soil (cf. Tables 2 and 3). This is attributed in part to fraction size differences. Compared to the values reported in literature Pb content of street dust in

Table 2. Pb and Cd content (mg/kg) of different soil layers in the area 1 km around the lead smelter.

Depth (cm)	Number of samples	Pb		Cd	
		arithmetic mean	range	arithmetic mean	range
0- 5	143	370	21-2309	1.60	0.28-10.47
5-20	133	256	32-1167	1.28	0.17- 7.89
0-20*	133	286	34-1126	1.37	0.20- 8.42

* As calculated from the 0-5 and 5-20 cm samples.

Table 3. Pb and Cd content (mg/kg) of street dust (fraction <0.3 mm) in different sampling areas.

Area	Number of samples	Pb		Cd	
		arithmetic mean	range	arithmetic mean	range
Arnhem, 't Broek	35	761	25-2667	3.81	0.5-16.17
Wageningen	6	350	148- 720	1.43	0.6- 2.19

't Broek-Arnhem is low; this is the more so as here the fraction <0.3 mm was taken. In Wageningen values both for Pb and Cd are much lower than in Arnhem due to lower traffic density and lack of other pollution sources. The high correlation (r = 0.85) between Pb and Cd in dust as derived from Fig. 6 is caused by the fact that both heavy metals in this material predominantly originate from the same source (traffic). Long-term accumulation by direct emission of the smelter is less probable because the street dust is regularly removed by cleaning and washing away into the street sewer system after rainfall.

Measures to prevent the continuation of lead uptake

In order to prevent further uptake of lead by children, contaminated dust and soil should be removed or their Pb contents decreased.

A cleansing operation of the streets by sweeping and rinsing temporarily deminished the amount of dust and the degree of contamination. These effects, however, were temporary only because the main sources of pollution (traffic, surrounding contaminated soil) remained unaffected.

A decrease of the lead content of the (top) soil can be achieved in different ways. Most effective is removal of the top-soil (0-20 cm) and replacement by uncontaminated soil. The material removed has to be disposed of somewhere else where no adverse effects are to be expected. As an alternative displacement of lead within the soil profile can be considered. This can be achieved by acidification, entailing dissolution of insoluble lead compounds and exchange from the adsorption complex, or by complexation with chelating agents like EDTA or DTPA, causing formation of soluble lead complexes.

Soil removal, although being immediately effective, is very expensive. Therefore it can be considered in severe cases only, where lead contents are so high that direct harmful effects to the health of children are to be feared. In the area studied at Arnhem the top layer (0-20 cm) was removed from all gardens with a Pb content exceeding 400 mg/kg in the upper 20 cm, and replaced with uncontaminated soil.

In-situ treatment of the soil by acidification or complexation have not well been studied so far. Therefore these methods were not yet applicable to situations occurring in practice. Research on these approaches, of which the results will be presented in forthcoming papers, has been initiated.

Conclusions

In the area 1 km around the lead smelter slightly elevated mean Pb contents in the soil were found compared to other urban areas. Lead contamination of soil could be ascribed mainly to Pb emissions from the smelter. Traffic, an important pollution source for both Pb and Cd, was the main source for Pb and Cd contamination of street dust. Mean lead and cadmium contents of street dust were not high compared to other big cities. To prevent further uptake of lead by little children contaminated dust and soil had to be removed. At present other methods, for instance displacement of lead in the soil profile, are not applicable in practice.

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References

Allaway, W. H., 1968. Agronomic controls over environmental cycling of trace elements. *Adv. Agron.* 20: 235.

Barltrop, D., 1972. Sources and significance of environmental lead for children. Int. Symp. Environm. Health Aspects Lead (Amsterdam). Eur. 5004 d-e-f: 657-681.

Brunekreef, B., S. J. Veenstra, K. Biersteker & J. S. M. Boley, 1981. The Arnhem lead study. I. Lead uptake by 1 to 3-year-old children living in the vicinity of a secondary lead smelter in Arnhem, The Netherlands. *Environm. Res.* 25: 441-448.

Farmer, J. G. & T. D. B. Lyon, 1977. Lead in Glasgow street dirt and soil. Science Total Environm. 8: 89-93.

Lagerwerff, J. V. & A. W. Specht, 1970. Contamination of roadside soil and vegetation with cadmium, nickel, lead and zinc. *Environm. Sci. Technol.* 4: 583-586.

Lepow, M. L., L. Bruckman, R. A. Rubino, S. Markowitz, M. Gilette & J. Kapish, 1974. Role of airborne lead in increased body burden of lead in Hartford children. *Environm. Health Persp.* 7: 99-101.

Linzon, S. N., B. L. Chai, P. J. Temple, R. G. Pearson & M. L. Smith, 1976. Lead contamination of urban soils and vegetation by emissions from secondary lead industries. *J. Air Pollution Control Ass.* 26: 650-653.

Page, A. L. & F. T. Bingham, 1972. Cadmium residues in the environment. *Residue Rev.* 48: 1-44.

Roberts, T. M., W. Gizijn & T. C. Hutchinson, 1974. Lead contamination of air, soil, vegetation and people in the vicinity of secondary lead smelters. Trace substances in environmental health VIII. Columbia, Mo., p. 155.

Yankel, A. J., L. H. von Lindern & S. D. Walter, 1977. The Silver Valley lead study: The relationship between childhood blood lead levels and environmental exposure. *J. Air Pollution Control Ass.* 27: 763-767.