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Lead in Deciduous Teeth of Children Living in a Non-ferrous Smelter Area and a Rural Area of the FRG*

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Summary. Lead concentrations were measured in the deciduous teeth (incisors) of 302 children living in a lead-smelter area in the FRG (Stolberg, Rheinland) and of 86 children living in a nonpolluted rural area (Gummersbach, Bergisches Land). Blood lead levels were determined in 83 of the children living in the lead-smelter area. On average, tooth lead levels of children living in the smelter area (mean: 6.0 µg/g; range: 1.49–38.5 µg/g) were significantly higher than those of children living in the rural area (mean: 3.9 µg/g; range: 1.6–9.4 µg/g). Blood lead levels were 6.8–33.8 µg/100 ml (mean: 14.3 µg/100 ml). Children of lead-workers had on average higher tooth lead and blood lead levels than children of people who were not lead-workers. Tooth lead levels increased with increasing duration of residence in the lead-smelter area and with the degree of local environmental pollution by lead, as indicated by the lead content of the atmospheric dust fall-out around the children's homes. The correlation coefficient of tooth lead vs blood lead was 0.47. The intra-individual variability of tooth lead levels was low ($r = 0.86$), and tooth lead levels of brothers and sisters were similar ($r = 0.75$), suggesting that tooth lead may be used as a representative and reliable indicator of long-term lead exposure.

Key words: Tooth lead levels – Blood lead levels – Current lead exposure – Long-term lead exposure

Introduction

Tooth lead levels have been used as a parameter to assess the long-term lead exposure of children and adults in various studies (Barry 1978; Ewers et al. 1979; Fosse and Justesen 1978; Khandekar et al. 1978; Shapiro et al. 1978; Needleman et

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al. 1979; Fergusson et al. 1980; Steenhout and Pourtois 1981). In 1979 we carried out a survey to determine lead levels in the deciduous teeth of children living in a non-ferrous smelter area at Stolberg, FRG. The Stolberg area is known to be highly polluted by lead and other heavy metals (arsenic, cadmium, copper, nickel, tin and zinc) due to the presence of ore-mining and non-ferrous smelters dating back to Roman times. Whereas ore-mining ceased at the beginning of the twentieth century, a large, primary non-ferrous smelter and several secondary non-ferrous smelters are at present in operation. In 1974 the industrial emissions of lead were estimated to be about 33.4 tons per year (Ministerium für Arbeit, Gesundheit und Soziales NW 1975). Since 1970 various preventive measures have been taken to reduce the emissions. Present industrial lead emissions are estimated to be about 17 tons per year (Ministerium für Arbeit, Gesundheit und Soziales NW 1975).

From 1973 to 1976 several surveys and epidemiological investigations were carried out in the area to evaluate the degree of population exposure to lead, cadmium and zinc and to define the relative contribution of air, food, water and other exposures to the total intake of these metals among the population (Einbrodt et al. 1974a, b; Rosmanith et al. 1975a, b, 1976; Einbrodt et al. 1976a, b). Since children were assumed to constitute a group particularly at risk, the investigations were mainly directed at children living in various parts of the city of Stolberg. The investigations comprised a determination of lead, cadmium and zinc levels in blood and hair as well as a determination of the urinary levels of cadmium and zinc.

With the exception of cadmium in urine (Buchet et al. 1980), all these parameters mainly reflect current or recent levels of exposure, but do not provide an indication of long-term exposure to these metals. Since lead is accumulated in the skeleton (bones and teeth) and tooth lead levels may be considered as an index of the lead body burden, an investigation of the tooth lead concentrations of children living in the area was carried out to assess their long-term exposure to lead. The study was combined with a neuropsychological investigation of children with varying tooth lead levels (Winneke et al. 1981; Winneke et al. 1982). The objective of the present communication is to report on:

1. Tooth lead levels of children living in the Stolberg area
2. Tooth lead levels of a reference group of children living in an area without industrial emissions of lead
3. The relationship between tooth lead levels and various indicators of exposure
4. The relationship between tooth lead levels and current blood lead levels

Subjects and Methods

Study Areas and Their Environmental Pollution by Lead

A description of the industrial area of Stolberg (about 57000 inhabitants) and data reflecting the degree of environmental pollution by lead and other heavy metals (air, soil, dust fall-out, water, food and fodder plants) have been published previously (Ministerium für Arbeit, Gesundheit und Soziales NW, 1975). The lead, cadmium and zinc concentrations of suspended particles in the ambient air have been recorded by five monitoring stations located in various parts of the city of Stolberg since 1973. The 1-year average levels of airborne lead ranged 1.28–7.61 $\mu g/m^3$ in 1973 and could be reduced by various preventive measures to 0.36–2.03 $\mu g/m^3$ in 1980. In 1971 the mean lead content of the atmospheric dust fall-out was up to 10 mg $Pb/m^2/day$ at 1–2 km from the lead-smelter and fluctuated around 0.5–2.0 mg Pb/m^2/day in various other parts of the city. In 1979 the atmospheric dustfall in the vicinity of the primary lead-smelter averaged around 1–3.6 mg Pb/m^2/day[1].

As regards the exposure of the population, particularly of children, it is important to mention that a number of slag dumps containing relatively high concentrations of a wide variety of non-ferrous metals are located at various parts of the city. Former investigations indicated that children playing on or near those dumps were highly exposed (Einbrodt et al. 1974b). Furthermore, it is assumed that in the past transportation of slag materials by lorries contributed significantly to the dust fallout along the transport routes.

Gummersbach (about 48000 inhabitants) is a rural and partially industrial area, which is not polluted by lead and other heavy metals because non-ferrous smelters do not exist in this area. Unfortunately, no data on airborne lead levels and the lead content of the atmospheric dust fall-out are available for this area. Motor vehicle exhaust fumes are considered to be the major source of lead emissions. Air monitoring performed by our laboratory in a rural area near Gummersbach showed a 1-year average of airborne lead levels of 0.14 µg/m^3 in 1976.

Study Populations

The parents of all Stolberg children born between July 1, 1968 and June 30, 1973 ($n = 3669$) were asked to submit, if available, the shed deciduous teeth of their child/children. For this purpose a letter, including a questionnaire and a small plastic bag to take up the teeth, was sent to these families. The questionnaire was designed to register the child's name, date of birth, present residence, duration of residence at the last dwelling-place and father's and mother's present and former occupation(s). In 1979 we collected 857 deciduous teeth from 338 children. Molars, canines, rotten teeth and teeth with fillings were discarded.

All in all, specimens from 302 children (143 boys; 159 girls) were analysed. The age distribution of these children was as follows:

Age group[a]	Number of children investigated	Total number of children living at Stolberg represented in the age group
1967/1968	5	1005
1968/1969	41	890
1969/1970	41	806
1970/1971	60	727
1971/1972	70	608
1972/1973	82	638
1973/1974	2	544

[a] The age groups comprise all children born between July 1 and June 30 of the following year. The teeth of seven children born in 1967/68 and 1973/74 were included in the study

Blood samples for blood lead analysis were collected on a voluntary basis from a subgroup of 83 children participating in the neuropsychological investigations from August to December 1980. The child's finger was carefully cleaned and the blood sample taken using heparinized

1 The data were kindly provided by Mr. Brehm, Ministerium für Arbeit, Gesundheit und Soziales NW, Düsseldorf

capillary tubes (44.7 µl; Brand, Wertheim, FRG), lead-free Lancets (Environmental Sciences Associates, Bedford, MA, USA), and a special vinyl plastic for sealing the capillary tubes (Microhematocrit Sealant; Brand, Wertheim, FRG). Two blood samples were obtained from 67 children.

For comparison, tooth lead levels of 86 children living at Gummersbach (Bergisches Land, FRG) were also analysed. The teeth from this group of children were collected in 1976, and the children had been born between 1965 and 1971.

Metal Analyses

Tooth lead levels were measured by electrothermal atomic absorption spectrophotometry (Perkin Elmer, model 400) equipped with a heated graphite furnace atomizer (Perkin Elmer, model HGA 500) purged with argon, a hollow cathode lamp for lead (Rank Hilger, Westwood, Kent, UK) and an automatic sample dispenser (Perkin Elmer, model AS 1). The sample preparation and determination of lead was performed as follows. Each specimen was washed with warm, double-distilled water and dried overnight at 110° C. The dried specimen was weighed and transfered into a specially cleaned, calibrated 50-ml glass flask containing 4 ml HNO_3 (65%; Merck, Suprapur, Darmstadt, FRG). Then the flask was heated until the tooth had completely dissolved (about 5–10 min) and after cooling the residue was diluted to a final volume of 50 ml. A 0.5-ml aliquot portion of this solution was diluted with 1 ml 0.01 *N* HCl (Brand Diluette, Wertheim, FRG) and the diluted solution transfered into specially cleaned Teflon vials of the automatic sample dispenser. The following working conditions were selected for the AAS analysis: drying 35 s at 120° C (ramp time 15 s); ashing: 24 s at 650° C (ramp time 20 s); atomization: 3 s at 1400° C (ramp time 0; internal flow: 10 ml Ar/min); injection volume: 0.050 ml. Each determination was carried out in duplicate. The quantitative determination of lead was performed with the aid of an average calibration curve based on a pooled 'tooth solution'. In order to check the precision of analyses, 0.5 ml aliquot portions obtained from a pooled tooth solution, which was prepared as described above and kept in a Teflon bottle at 4° C, were examined with each analytical series. The analytical coefficient of variation (including the day-to-day variation) was 5% at tooth lead levels of about 6.0 µg/g. Over the entire concentration range the reproducibility of analyses, as calculated on the basis of analyses of 604 pairs of subsamples, was on average 6%. Under the working conditions described, the detection limit of the method is 0.15 µg Pb/g.

Blood lead levels were measured by the method of Stoeppler et al. (1978), which was modified for small volumes of blood. Calibration was obtained by a standard addition calibration curve on the basis of blood. The accuracy of blood lead analyses was ascertained by regular participation in quality control programmes organized by the Health and Safety Directorate of the CEC. Over the entire concentration range, the reproducibility of analyses, as calculated on the basis of 64 duplicate samples independently pre-treated and analysed, was about 15%.

Statistical Analyses

Data were normally distributed after logarithmic transformation. Logarithmic data were used, therefore, for all statistical analyses. Multiple step-wise regression analysis was used to evaluate the effect of various independent variables on tooth lead levels. The regression assumptions were tested by examining the residuals in the subgroups defined by the variables applied (Bartlett's test of homogeneity of variances; examination of outliers; test for the equal distributions of signs). Differences in the cumulative frequency distributions were statistically ascertained by the χ^2 test.

Results

The data on tooth lead (PbT) and blood lead (PbB) levels of the Stolberg and Gummersbach children are summarized in Table 1. The geometric mean value of the tooth lead levels of the Stolberg children is significantly higher than that of the

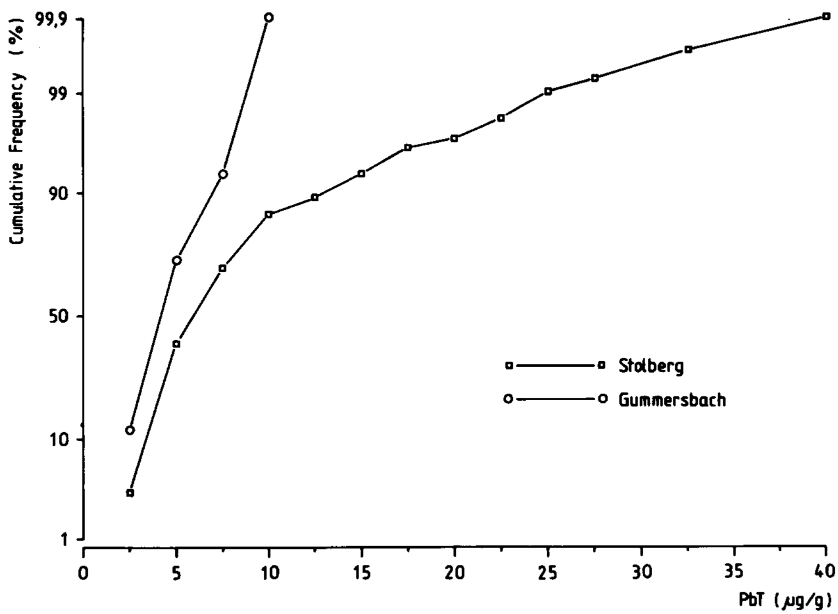

Fig. 1. Cumulative frequency distributions of tooth lead levels *(PbT)* of children living in a non-ferrous smelter area *(Stolberg)* and in a rural area *(Gummersbach)*

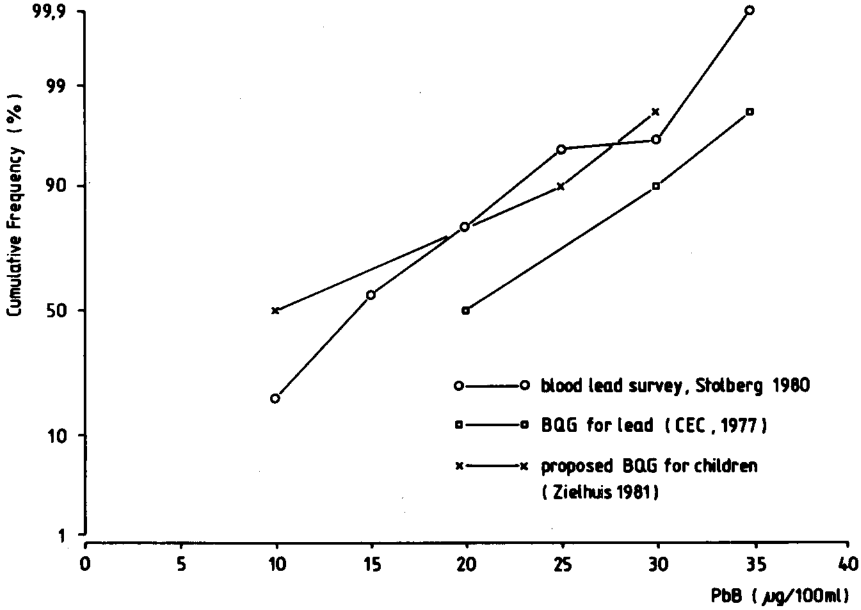

Fig. 2. Cumulative frequency distribution of blood lead levels *(PbB)* of children living in a non-ferrous smelter area *(Stolberg)*

Table 1. Tooth lead and blood lead levels in children living at Stolberg and Gummersbach

Parameter	Stolberg					Gummersbach				
	n	Mean[a]	Median	Coefficient of variation	Range	n	Mean	Median	Coefficient of variation	Range
Tooth lead (μg/g)	302	6.0 (7.1)	5.9	0.72	1.5–38.5	85	3.9[b] (4.2)	3.8	0.49	1.6–9.4
Blood lead (μg/100 ml)	83	14.3 (15.3)	14.1	0.44	6.8–33.8					

[a] Geometric means; arithmetic means in parenthesis
[b] Significant difference vs the geometric means of the reference group at the $P<0.001$ level (test of separate variance estimate; $t=8.05$; $df=182$)

Gummersbach children ($P<0.001$). Figures 1 and 2 present cumulative frequency distributions of the tooth lead and blood lead levels. The distribution of the blood lead levels of the Stolberg children compares favourably with the Biological Quality Guidelines (BQG) for screening of the general population for lead (CEC, 1977), but significantly exceeds ($P<0.001$) the proposed BQG for pre-school children (Zielhuis 1981).

Table 2 presents the results of a multiple step-wise regression analysis of the effect of various variables and indicators of exposure on the variation of the tooth lead levels of the Stolberg children. The results show that tooth lead levels are significantly affected by two of the variables applied: (a) father's present and/or former occupational exposure to lead and (b) duration of residence at Stolberg.

The effect of these variables on the tooth lead levels are shown in Table 3. On average, children have higher tooth lead levels if their fathers are occupationally exposed to lead (or have been) than children whose fathers have never been lead-workers. Furthermore, it is obvious that tooth lead levels increase with increasing duration of residence at Stolberg.

Children of unskilled manual workers have, on average, significantly ($P<0.001$) higher tooth lead levels (mean: 8.54 μg/g; range: 3.0–23.8 μg/g; $n=30$) than children of 'white-collar' and skilled manual workers (mean: 5.45 μg/g; range: 1.5–31.2 μg/g; $n=236$). Children of lead-workers were not considered in this evaluation.

Brothers and sisters have similar tooth lead levels (Fig. 3). The correlation coefficient, calculated on the basis of 21 pairs of sisters and brothers, is 0.75 ($P<0.001$). The intra-individual variability of tooth lead levels was evaluated on the basis of 89 children who had submitted more than one incisor (Fig. 4). The correlation coefficient is 0.86 ($P<0.001$).

In order to study the relationship between environmental lead pollution and tooth lead levels, we selected those children who have lived in their present homes

Table 2. Multiple step-wise regression analysis of the effect of various variables on tooth lead levels in children ($n = 287$[a]) living in a non-ferrous smelter area (Stolberg)

Source of variation	*df*	SQ	*F*	Percent of the total variation explained
Sex	1	0.01	0.14	0.05
Age[b]	1	0.09	1.54	0.54
Father's occupational exposure to lead (two categories[c])	1	1.53	29.05[e]	9.25
Duration of residence at Stolberg (three categories[d])	2	0.50	4.94[f]	3.05
Not explained	281	14.42		

[a] Children with questionnaire incompletely filled out were not considered in this calculation
[b] Age at the time this study was conducted (1979)
[c] Category A: father occupationally exposed to lead at present or formerly ($n = 23$); category B: father occupationally not exposed to lead at present or formerly ($n = 264$)
[d] Category A: children who moved to Stolberg within the first 4 years of their lives; category B: children who moved to Stolberg after the 4th year of life; category C: children who have been living at Stolberg all their lives
[e] Significant at the $P < 0.001$ level
[f] Significant at the $P < 0.01$ level

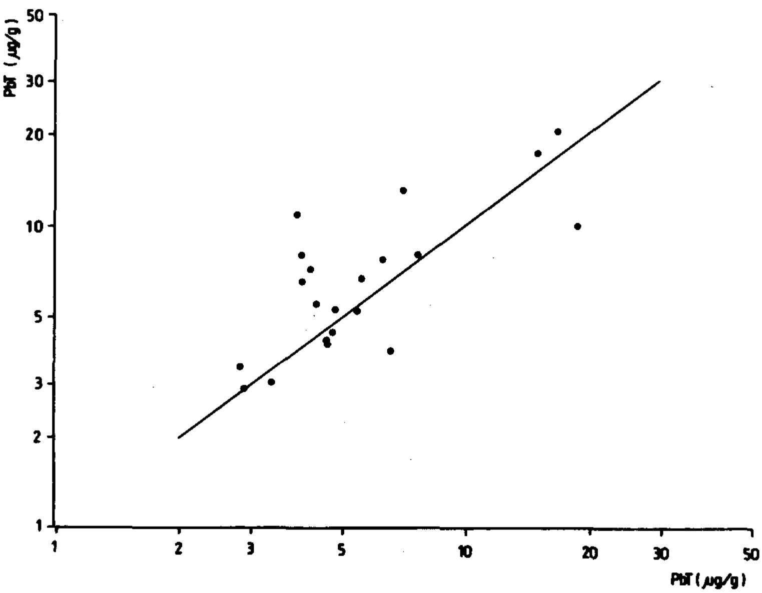

Fig. 3. Tooth lead levels of brothers and sisters ($n = 21$; $r = 0.75$)

Table 3. Tooth lead levels (µg/g) in relation to father's occupational exposure to lead and the duration of residence in the non-ferrous smelter area (Stolberg)

	Father occupationally not exposed to lead			Father occupationally exposed to lead		
	n	Mean	Coefficient of variation	*n*	Mean	Coefficient of variation
Children who moved to Stolberg after the 4th year of life	12	4.24	0.91	1	(29.3)	
Children who moved to Stolberg before the 4th year of life	28	5.14	0.55	1	(8.21)	
Children living at Stolberg all their lives	224	5.91	0.68	21	11.36	0.80

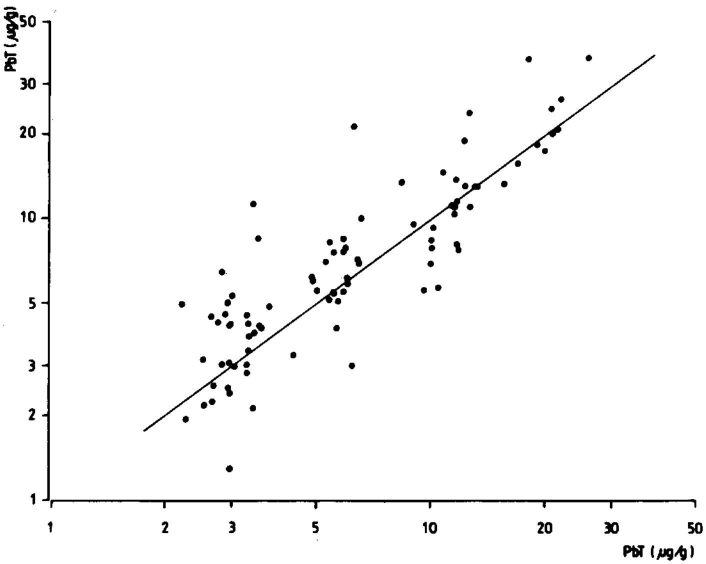

Fig. 4. Intra-individual variability of tooth lead levels *(incisors)*; $n = 89$; $r = 0.86$

all their lives. The lead content of the atmospheric dust fall-out was used as a parameter to characterize the local environmental pollution by lead. Data on the lead content of the atmospheric dust fall-out in 1973–1976 were available for 23 quadrants (1 km × 1 km areas), which covered the environment of 1–2 km around the primary lead-smelter as well as the central parts of the city of Stolberg. In total 95 children who lived in this area all their lives were considered.

A multiple step-wise regression analysis, including the lead content of the atmospheric dust fall-out (means from the period 1973–1976) as a further variable, was calculated for this group. The results summarized in Table 4 show that the variation of tooth lead levels is significantly affected by three of the variables applied: (a) age; (b) father's occupational exposure to lead; (c) mean lead content of

Table 4. Multiple step-wise regression analysis of the effect of various variables on the tooth lead levels in children living in a non-ferrous smelter area. In this calculation only children were considered who had been living in their present homes all their lives ($n = 95$)

Source of variation	*df*	SQ	*F*	Percentage of the total variation explained	Level of significance
Sex	1	0.01	0.02	0.02	NS
Age	1	0.32	5.29	5.41	$P<0.05$
Father's occupational exposure to lead[a]	1	1.13	22.56	18.82	$P<0.001$
Pb content of the atmospheric dust fall-out[b]	1	0.48	10.69	8.05	$P<0.01$
Not explained	90	4.05			

[a] Category A: father occupationally exposed to lead at present or formerly ($n = 11$); category B: father occupationally not exposed to lead at present and formerly ($n = 84$)
[b] The means of 1-year average levels in 1973–1976 were used

the atmospheric dust fall-out during 1973–1976. Tooth lead levels increased with the age of the child and the lead content of the atmospheric dust fall-out (PbD) in the quadrant of the child's home (regression equation: log PbT = 4 0.076 PbD + 0.71; $r = 0.33$).

The effect of the variable 'father's occupational exposure to lead' amounts to 18.8% of the total variation explained, since there are relatively more children of lead-workers represented in this subgroup than in the total sample examined (see Table 2).

Tooth lead levels were correlated with blood lead levels. There is a significant linear correlation between these parameters (log PbB = 0.268 log PbT + 0.942; $n = 83$; $r = 0.47$; $P < 0.001$). Children of lead-workers tend to have on average higher blood lead levels (geometric mean: 19.7 µg/100 ml; range: 13.6–31.9 µg/100 ml; $n = 5$) as compared to the other children in the survey (geometric mean: 14.2 µg/100 ml; range: 6.8–33.8 µg/100 ml; $n = 73$) (*t*-test; $P = 0.05$).

Discussion

This investigation shows that the children living in the lead-smelter area (Stolberg) usually have a significantly higher lead body burden than otherwise comparable children living in a non-polluted area (Gummersbach). Tooth lead levels of the Stolberg children are affected at least by four partially interdependent variables reflecting the degree and duration of environmental lead exposure: (a) father's occupational exposure to lead; (b) duration of residence at Stolberg; (c) age; (d) local environmental pollution by lead as indicated by the lead content of the atmospheric dust fall-out around the children's homes. Unfortuntely, no data regarding the lead content of house dust and domestic waters or the occurrence of lead pipes in aged houses, etc. were available to assess the influence of various other factors of possible relevance.

An outstanding finding of this study is that children of present and/or former lead-workers have, on average, higher tooth lead levels than otherwise comparable children. This is consistent with various reports in the literature showing that children of lead-workers have higher mean blood lead levels than children of people who are not lead-workers (Baker et al. 1977; Center for Disease Control 1977, as quoted by Elwood et al. 1977; Millar 1978; Bundesgesundheitsamt 1980). To explain this effect it has been suggested that it is the lead-worker himself who brings the lead home, presumably in the form of dust on his person or clothing. Although a number of hygienic measures have benn taken at the lead-works to prevent this form of 'domestic pollution', the results shown in Tables 2 and 3 demonstrate that in relation to the other factors considered, the presence of a lead-worker in the household appears to be an important factor. This is further confirmed by the results of the blood lead survey of this study, which indicate that even under today's conditions the 'lead-worker effect' appears to be of relevance.

Another important factor investigated in this study is the effect of the duration of residence in the smelter area on the tooth lead levels. It has been demonstrated by numerous studies that children living in non-ferrous smelter areas are more heavily exposed to environmental lead and other toxic metals such as cadmium, arsenic and manganese than children living in non-polluted rural or in urban areas (Roels et al. 1980; Buchet et al. 1980). Since tooth lead levels are a cumulative function of earlier exposure, it is reasonable to presume that with long-term elevated exposure, tooth lead levels increase with increasing duration of residence in the smelter area.

Children of unskilled manual workers usually have significantly higher tooth lead levels than those of skilled manual and non-manual workers. This finding parallels recent observations showing increased blood lead levels in children of unskilled manual workers compared to children of skilled workers (Yule et al. 1981). The cause of these differences might be a higher degree of 'domestic pollution' in the families of low socio-economic status or other factors unknown at present, which need to be clarified.

Tooth lead levels of those children who lived at Stolberg for their whole life are significantly affected by three of the variables applied in this study: father's occupational exposure to lead; age (which in this group reflects the duration of exposure); lead content of the atmospheric dust fall-out. The lead content of the atmospheric dust fall-out may be considered an indicator of the degree of environmental pollution on the playgrounds, which generally are located near the children's homes. Excessive contact with soil and dust when playing outdoors as well as indoors has been demonstrated to constitute an important factor regarding excessive exposure of children in non-ferrous smelter areas (Sayre et al. 1974; Roels et al. 1980; Buchet et al. 1980).

It is unlikely that different dietary habits could account for the differences of tooth lead levels of the Stolberg and the Gummersbach children. Former investigations carried out in the Stolberg area did not show dietary habits to have an effect on the blood lead levels of children, although high lead concentrations have been found in vegetables as well as in meat, livers and kidneys of domestic animals grown in the Stolberg area (Schröder et al. 1978). It may be concluded, therefore, that the differences in tooth lead levels of the Stolberg and the Gummersbach children observed in this study are mainly due to the different degree of environmental lead pollution (air, dust) in the two areas. The blood lead levels of the Stolberg children observed in this survey indicate, however, that the current exposure of these children is only moderately increased. The distribution of blood lead levels compares favourably with the reference values of the Council Directive on Biological Screening of the Population for Lead (CEC 1977). With regard to the increased susceptibility of young children it has been proposed, however, that BQG should be lowered for children, particularly pre-school children: 98% < 30 μg/100 ml; 90% < 25 μg/100 ml; 50% < 10 μg/100 ml (Zielhuis 1981). In The Netherlands the Ministry of Health and Environmental Hygiene has accepted this new BQG as an official guideline for the population survey in 1979 and 1981, particularly with respect to the screening of pre-school children (Zielhuis 1981). Compared to these reference values, there is still an unacceptable degree of exposure for some of the Stolberg children examined in this study. Further investigations appear to be warranted to evaluate the exposure of the population groups most at risk, i.e. infants, young children and pregnant women. From our study it can be concluded that one way to reduce the total exposure of some children could be the reduction of 'domestic pollution' in the lead-worker families.

Tooth lead and blood lead levels were correlated moderately well in the children examined in this study. Whereas blood lead levels change rapidly with changing conditions of exposure (total intake), tooth lead concentrations are a cumulative function of long-term lead intake and allow undetected cases of undue lead absorption to be identified when other indices of exposure have returned to normal. A high correlation between tooth lead and blood lead, therefore, cannot be expected. Similar correlations have been obtained recently by Ernhart et al. (1981) who correlated pre-school and school-age blood lead levels of children with their dentine lead levels. Shapiro et al. (1978) did not find a correlation between blood lead and tooth lead levels; however, when tooth lead concentrations were divided by the child's age, a significant correlation was obtained between these parameters.

Finally, it should be pointed out that the results of our study support the idea of using the lead concentration of deciduous teeth as a representative and reliable indicator of long-term lead exposure of children. The intra-individual variability of tooth lead levels appears to be small when the same type of tooth is considered. Moreover, there is a high correlation between the tooth lead levels of siblings, indicating that the tooth lead levels are representative of specific environmental conditions for a family.

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