Global Environmental Monitoring System (GEMS)

Assessment of Human Exposure to Lead: Comparison between Belgium, Malta, Mexico and Sweden

Prepared for
United Nations Environment Programme
and
World Health Organization

by

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KAROLINSKA INSTITUTE, STOCKHOLM
1985

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Liber Tryck AB Stockholm 1985

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Chapter 1

Introduction

In 1978 an international UNEP/WHO project was initiated to assess human exposure to lead and cadmium through biological monitoring. The development and results of the project have been published in "Assessment of Human Exposure to Lead and Cadmium through Biological Monitoring" (Vahter, 1982; Friberg and Vahter, 1983). The following countries participated in the project: Belgium, India, Israel, Japan, Mexico, People's Republic of China, Peru, Sweden, USA and Yugoslavia. Scientific responsibility for the implementation of the project was delegated to a Coordinating Institution: the National Institute of Environmental Medicine and the Department of Environmental Hygiene of the Karolinska Institute, Stockholm, Sweden.

About 200 teachers from one urban area in each country constituted the target group. Lead and cadmium were measured in samples of venous blood. The programme was carried out in conjunction with a rigid quality control programme.

The results showed that there is considerable variation in metal exposure between the areas studied. For example, for lead in blood median values ranged from about 60 µg Pb/l in Beijing and Tokyo to 220 µg Pb/l in Mexico City. The 90-percentile values ranged from 89 µg Pb/l in Tokyo to 346 µg Pb/l in Mexico City.

The project did not study the different sources of observed differences between countries. The project concluded with a recommendation that biological monitoring should also be carried out in other areas. Such an expansion of the project should include integrated monitoring of pollutants in different environmental media, e.g. food, drinking water and air. The use of integrated monitoring was subsequently endorsed by a UNEP/WHO Government Expert Group in 1982.

The present paper describes a follow-up study of the above mentioned UNEP/WHO Pilot Project on Assessment of Human Exposure to Lead and Cadmium through Biological Monitoring, carried out in 1983/84.

The main objectives of the follow-up study were:

- To study whether differences in blood lead levels found between countries in the Biological Monitoring Project were primarily due to exposure via ingested lead or via inhaled lead;
- To study the relationship between exposure to lead and concentrations of lead in blood; and
- To make a preliminary survey, in selected areas, of the possible sources of high exposure.

The study was planned to include Belgium, Mexico, Peru and Sweden. Judging from concentrations of lead in blood found during the 1981 Biological Monitoring Project, Belgium was an "intermediate exposure" country, Mexico was a "high

exposure" country, and Peru and Sweden "low exposure" countries. For various reasons it was not possible to include Peru in the follow-up study.

An earlier collaborative study between Belgium and Malta (Bruaux et al., 1983) had, however, shown that Malta might prove to be a very useful participant for this study. Blood lead levels in Malta appeared to be even higher than those in Mexico, with median values of about 290 μg Pb/l and 90-percentile values of about 450 μg Pb/l.

The final study included Belgium, Malta, Mexico and Sweden. Participating institutions were:

- The Institute of Hygiene and Epidemiology (IHE), Brussels, Belgium;
- The Department of Health, Valletta, Malta;
- General Directorate of Research on Health, Vice Ministry of Research and Development, Ministry of Health, Mexico City, Mexico; and
- The National Institute of Environmental Medicine (NIEM) and the Department of Environmental Hygiene of the Karolinska Institute (KI), Stockholm, Sweden.

WHO delegated the overall responsibility for the project to NIEM and KI, Stockholm, Sweden.

The exposure to lead was evaluated primarily by analyses of lead in blood, feces and air. The present study did not include air analyses in Mexico and Sweden. In Mexico City there are results from a National Monitoring Program indicating high air lead levels. In Sweden, previous studies had shown that air lead levels are low in this country. In Belgium, air monitoring for heavy metals has been routinely performed for several years, while in Malta three air sampling stations, technically similar to the Belgium ones, were installed for one full year for the purposes of the present study.

Blood lead levels were considered to provide the best indication of total exposure to lead, while lead in feces, due to the relatively low absorption (on an average 10%) of ingested lead, provides an indication of the actual exposure via ingestion (including lead cleared from lungs and swallowed). In Malta, in addition to analysis of lead in blood, feces and air, further studies were carried out to explore possible explanations for the high levels of lead in blood. These additional studies were on an exploratory level only and were carried out to supply the appropriate authorities with as much information as possible and, thus to enable them to implement protective measures and to form a basis for further, more detailed studies. The target groups were drawn from those in the above mentioned UNEP/WHO Biological Monitoring Project.

The IHE carried out the analyses for Malta and also assisted in the actual field studies in that country. During the planning stage of the project it was agreed that the NIEM/KI should take responsibility for analyses of lead in blood and feces from Mexico. In practice, the studies were carried out in close collaboration between NIEM/KI and the Mexican counterpart.

A quality assurance programme for the analysis of lead in blood and feces was implemented throughout the project.

Although the main responsibility for the preparation of this report has rested on the Belgian and Swedish institutions, a draft report, prepared by the IHE, was discussed at a meeting in Malta, 30 May—4 June 1984, with participants from Belgium, Malta, Mexico and Sweden. A first draft of the final report was discussed

at a meeting in Brussels with participants from Belgium and Sweden, 10—13 September 1984. The revised final draft report, based on a discussion, and prepared during a meeting in Stockholm, 23—26 October 1984, with participants from Belgium and Sweden, was circulated to the participating institutions for comments.

Chapter 2

Background

2.1 General background

Lead in humans may originate from many different sources. Lead is present in the working environment, in ambient air, in drinking water, in food, and in nonfood items such as, for example, paint. The best way to estimate total exposure is, therefore, through biological monitoring. This is possible because sufficient information on metabolism is available. The lead level in blood is the best indicator of current exposure. Blood lead level reflects a dynamic equilibrium between exposure, absorption, distribution and elimination of lead (WHO, 1980).

Lead in the environment may enter the body through either inhalation, oral intake or percutaneous absorption. The latter route is considerably less significant than the respiratory and gastrointestinal routes for uptake of inorganic lead (WHO, 1980).

Intake via inhalation is dependent on particle size and breathing pattern. WHO (1980) proposed that $30 \pm 10\%$ of inhaled lead is absorbed through the lung. Chamberlain et al. (1978) investigated the deposition rate of lead from car exhaust. They found that deposition rate in lungs was 60% for inhaled lead from "fresh" car exhaust and 50% for inhaled lead from other urban air. This small difference may be due to differences in particle size. In "fresh" car exhaust the particles are smaller than those inhaled from other urban air or during occupational exposure. Solubility of the particles is of less importance for exposure via inhalation, since virtually all particles deposited in the lungs will be absorbed (Chamberlain et al., 1978; Morrow et al., 1980).

Oral intake of inorganic lead is another exposure route of importance. Oral intake mainly involves uptake from food and beverages, including water. Particles deposited and cleared from the upper respiratory tract will, to some extent, contribute to oral exposure. Oral intake also includes nonfood substances such as lead-containing paint. Most of the absorbed lead is primarily accumulated in the bone system where it has a very long biological half-time. Lead is excreted mainly via urine (80%) and gastrointestinal secretion (WHO, 1980).

2.2 Lead uptake

Several factors may influence the rate of uptake and retention of lead. Presence of food in the stomach seems to be the most important factor. Chamberlain et al. (1978) found a mean retention of $PbCl_2$, after 5 days, of 7% with food and 45% when fasting. Corresponding figures for PbS were 6% and 12%, respectively. Rabinowitz et al. (1980) found that the absorption of a lead tracer was 8.2% in the presence of food and 35% when no food was present in the stomach.

Calcium and phosphate, especially in combination, decrease lead uptake (Bar-

ton et al., 1978; Quarterman, 1978a; Moore et al., 1979; Heard and Chamberlain, 1982). Low protein diet, high lipid intake and undernourishment increase lead uptake (Quarterman et al., 1978b; Moore, 1979; Aungst and Fung, 1981). However, the effect of iron deficiency on the absorption and retention of lead in man is ambiguous (Flanagan et al., 1982). It is poorly understood to what extent these different factors influence absorption and retention of lead.

Absorption from the gastrointestinal tract is often given as 10% for adults (WHO, 1977). Experimental data on absorption are presented, for example, in the Kehoe lead balance studies (1961a, b and c). In these studies median dietary lead estimate, based on duplicate diets, was 159 μg Pb/day; average daily fecal lead was 109% of dietary lead with considerable variation (Gross, 1981). Oral supplementation experiments were performed on four subjects who received supplements of 300, 1000, 2000 and 3000 μg Pb/day. Fecal lead dropped to 83% of dietary lead and Gross (1981) estimated lead balance to be 0.13 μg Pb/day/microgram increase of lead in diet for these four subjects. Rabinowitz et al. (1976) studied absorption in five subjects using a stable lead ^{204}Pb isotope. Absorption ranged from 6.5 to 13.7% of intake. Moore et al. (1979) using oral intake of ^{203}Pb and a whole-body counter found that the mean whole-body retention in 11 subjects after four days was 21.3% with a range 10 to 48%. It is not known how much lead remained in the gastrointestinal tract.

In a lead balance study on healthy children, 3 months to 8 years of age, Alexander et al. (1974) found mean absorption of ingested lead to be 53%. In another balance study, Ziegler et al. (1978) found a net absorption of 42% of intake, in children less than two years of age. These data, even though uncertain, indicate that children might have a higher gastrointestinal absorption rate than do adults. The figure of 10%, proposed by WHO (1977), for gastrointestinal absorption in nonfasting adults must, therefore, be considered as a rough estimate only, with large interindividual variations. Oral intake of 100 to 200 μg Pb would be in the same order of magnitude as, e.g. exposure to 1 to 2 μg per m^3 of lead in air, if it is assumed that 20 m^3 air is inhaled per 24 hours and that 50% of the lead inhaled with air enters the body. Oral intake of lead between meals, from any source, may be of significance due to higher absorption rates of lead during fasting.

Several studies have attempted to relate oral intake of lead to blood lead levels. To estimate external lead exposure due to oral intake, three approaches are used: (1) duplicate meals, (2) market basket surveys, and (3) fecal lead determinations.

Duplicate Diet In duplicate diet studies, estimated lead exposures are assessed by having subjects put aside, for a limited period of time, a duplicate portion of the food consumed at each meal. These studies probably provide a good, but short-term estimate of oral intake. The techniques available for the analysis of lead in food have, historically, been subject to inaccuracies, hence, the validity of data obtained using the duplicate diet approach has not been confirmed.

Market Basket Approach The market basket approach uses the observed lead concentration for a variety of food items coupled with estimated dietary consumption of the particular food items. Some studies base estimated exposure on national estimates of typical consumption patterns. Other studies actually record daily dietary intakes. This approach is subject to analytical problems similar to those found in the duplicate diet approach. It also faces the additional problem of obtaining an accurate estimate of dietary intakes.

Fecal Lead Determination Fecal lead determination, faces two major difficulties. Firstly, the procedure involves the use of a mathematical estimate of the coefficient of overall absorption from the gut, to estimate the external exposure. As discussed earlier in this section, this coefficient is probably not constant. Secondly, it is difficult to collect a representative sample. One advantage of fecal lead determination is, however, that all substances ingested via the oral route are taken into account, even nonfood items.

2.3 Epidemiological studies

Most studies have focused on the correlation between lead in water and blood lead levels. This is, probably, due to the fact that lead in water was an important contributor to exposure in the areas where blood lead levels were found to be high. To predict lead exposure, on the basis of concentrations in water, is complicated, since consumption varies greatly and the actual concentration of lead in water depends on whether "first draw water" or running water is consumed.

Pocock et al. (1983) approximated oral intake to concentration of lead in "first draw water". In this particular study lead in water was the major source of lead exposure. Pocock and co-workers found a linear regression for "first draw water" lead levels of less than 100 μg Pb/l. At levels above 100 μg Pb/l the slope was considerably less; 23% of the first regression line. Moore et al. (1982) found a curvilinear cubic root relationship. Sixty percent of the subjects had "first draw water" lead concentrations below 50 μg Pb/l. Both these studies indicate a nonlinear relationship when lead concentrations in water are high.

Sherlock et al. (1982) considered exposure to both dietary lead and drinking water lead. Concentrations of lead in drinking water were determined for 114 dwellings. A duplicate diet survey was conducted on a nonrandom sample of 31 out of a population of 114 adult women. Eleven subjects had a weekly intake of lead in diet above 3 mg/week, 9 subjects had a weekly intake between 3 and 1 mg/week and 11 subjects a weekly intake below 1 mg/week. It was found that a curvilinear regression fitted the obtained data better than a linear one, $PbB = -14 + 36\sqrt[3]{PbD}$, $r^2 = 0.52$, PbB = blood lead in μg/l, PbD = diet lead μg/day.

The Department of the Environment Central Directorate on Environmental Pollution (1982) found a curvilinear relationship between dietary lead and blood lead levels, with calculated PbB increases of about 50 μg Pb/l, when oral intake increases from 100 to 200 μg Pb/day. These studies were, however, performed on subjects with higher than normal dietary lead intake only, >300 μg Pb per day.

2.4 Experimental studies

Aungst et al. (1981) showed that gastrointestinal absorption in rats is dose-dependent. Experimental studies in humans have generally used high intake of lead from supplements to the diet. For example, Stuik (1974) gave a lead supplement of 20–30 μg Pb/kg and day; for a 70 kg man this corresponds to 1400–2100 μg Pb/day, and Kehoe's experiment (Gross, 1981) included supplements of 300–3000 μg Pb/day. The Stuik (1974) and Gross (1981) studies showed a relationship between oral intake to blood lead level. An increase of 100 μg Pb/day will increase blood lead

level by 20 μg Pb/1. The slope was lower at high oral intake. However, the blood lead levels were generally high.

Ryu et al. (1983) performing a study on 17 infants, 8 to 196 days of age, fed milk with different lead content (average dietary intake 16 μg Pb/day for infants fed milk supplied in cartons and 61 μg Pb/day for infants fed milk or formula supplied in cans). They found a linear relationship, $PbB = 46 + 1.6\ PbD$ (PbD = μg Pb/day).

In the report of the Nutrition Foundation's Expert Advisory Committee "Assessment of the Safety of Lead and Lead Salts in Food" 1982, available data from the literature, on the relationship of exposure to lead via the gastrointestinal route to blood lead levels, were combined and resulted in the equation; $PbB = 43.3 + 10.5\sqrt{PbD}$. An increase in dietary intake from 100 to 200 μg Pb/day would, according to this equation, result in an increase in blood lead levels from 148 to 192 ug Pb/1.

Laxen (1983) proposed the following equation; $PbB = 28.3\sqrt{(uptake)}$ based on a wide range of data. An increased oral intake from 100 to 200 μg Pb/day, assuming a 10% absorption, would increase blood lead levels from 90 μg Pb/1 to 127 μg Pb/1, i.e. an increase of 37 μg Pb/1 blood.

2.5 Background to the present study

This study was implemented using biological monitoring of lead in blood and lead in feces, and monitoring of environmental samples of lead in drinking water and dust. Lead in blood is a measure of the total "internal" exposure to lead, lead in feces a measure of exposure via the gastrointestinal route. To evaluate possible sources of lead exposure samples of lead in drinking water and indoor and outdoor dust were analyzed. Other possible sources of lead exposure are working environment, ambient air, food, and ingestion of nonfood substances.

The working environment was not an objective of the present study. We believe that our choice of target group, nonsmoking males, mainly teachers, eliminates occupational exposure as a source of uptake in the group studied. Analysis of samples of ambient air and food did not generally fall within the scope of this study. For both Belgium and Sweden several studies are available which deal with these specific exposure sources. In Sweden levels of lead in air and lead in food are known to be low. In Belgium lead levels in both these sources of exposure are somewhat higher than the levels in Sweden. Some preliminary data on levels of lead in food and air are available for Mexico, indicating high concentrations.

Malta, where lead exposure probably was higher than average (Bruaux et al., 1983), was selected for a preliminary survey of possible sources of exposure. This survey included analysis of environmental samples of ambient air, food items, soil and street dust. For comparison equivalent samples were analyzed in Belgium.

2.6 General information about the four countries

2.6.1 Belgium

The population of Belgium in 1982 was 9 855 000 (333 persons/km²); 995 000 inhabitants (6149 persons/km²) lived in the capital.

Brussels, which is located at a latitude of 51° north and a longitude of 2° east, is the administrative center of the country. Belgium has a wide variety of industries as well as several printworks, articles such as laces, gloves, furniture, car parts, musical instruments, jewellery, and lithography as well as several printworks are produced. There are also breweries and distilleries. The climate is of the temperate-maritime type with four seasons, spring (March—May), summer (June—August), autumn (September—November) and winter (December—February). The average daily temperature is about 16° C in summer and 3° C in winter. The average annual precipitation is about 780 mm.

Traffic density and lead emission

In August 1982 the number of private cars (excluding trucks, buses, motorcycles, etc.) was 3 234 951, corresponding to about 1 car for every 3 inhabitants. In Brussels there were 359 952 private cars in August 1982, corresponding to about 1 car for every 3 inhabitants.

Between 1970 and 1977 the maximum allowable lead concentration was 0.84 g Pb/l petrol. Concentrations decreased progressively from 0.84 g Pb/l in 1970 to 0.75 g Pb/l in 1977. The maximum allowable concentration of lead in petrol decreased to 0.45 g Pb/l in 1978 and 0.40 g Pb/l in 1982.

Annual petrol (diesel oil excluded) consumption increased from 2.2 million metric tons in 1970 to 2.7 million metric tons in 1982 (i.e. from 2.9 million to 3.6 million cubic metres). Based on the maximum allowable lead concentrations quoted above it can be calculated that the lead emissions from car traffic dropped from 2505 metric tons in 1970 to 1443 tons in 1982. One tenth of the total number of private cars are located in Brussels within an area of 162 km^2. For 1982 lead emission from cars can be estimated to be about 1.1 metric tons Pb/km^2 and year in Brussels.

2.6.2 Malta

The Maltese Islands, which comprise Malta, Gozo, Comino and two other small uninhabited islands, are situated in the middle of the Mediterranean Sea. Total area 316 km^2. The population in 1981 was 323 000 inhabitants (1022 persons/km^2). The capital, Valletta, is located at a latitude of 35° north and a longitude of 14° east. The Islands can be considered as a whole unit, there being no real differences between rural and urban areas. The weather of the Maltese Islands is characterized by long spells of fine, calm weather with only brief periods of storm and rough weather. The summer is fine and hot with maximum average temperatures reaching about 30° C, however, the sea breeze prevents the coastal regions from becoming too hot. The onshore sea breeze during the day and the offshore breeze at night occur when a slack pressure gradient covers the Central Mediterranean; this is a common occurrence during the summer months. The annual average precipitation is about 559 mm.

Traffic density and lead emission

In mid-1983 the total number of vehicles on the road was 109 319 of which 71 380 were private cars; i.e. approximately 1 private car for every 4.5 inhabitants. About 75% of the private cars are of class A type (less than 1500 cc).

In Malta lead content in petrol is not regulated by legislation. The usual content is reported to be 0.4 μg Pb/l, but occasionally somewhat higher concentrations have been observed.

From 1973 to 1982 the annual consumption of petrol did not vary appreciably. In 1973 there was a maximum consumption of 43 752 metric tons and in 1974 a minimum of 32 716. The mean for the 10-year period was 38 794 metric tons, i.e. 52 370 m^3. Based on the mean lead concentration quoted above this implies a lead emission, from car traffic only, of about 21 metric tons per year. This corresponds to about 0.07 metric tons Pb/km^2 and year for Malta.

2.6.3 Mexico

The population of Mexico in 1982 was 72 118 000 inhabitants (37 persons/km^2). Mexico City has a population of 9 400 000 inhabitants, while the population of the metropolitan area is 14 500 000 inhabitants; the mean population density is 6500 persons/km^2. Mexico City is located at a latitude of 19° north and a longitude of 99° west. Mexico City is situated 2240 m above sea level and has a highland climate. The minimum temperature is −2°C in the winter and the maximum +29°C in summer. The annual precipitation is about 560 mm.

Traffic density and lead emission

The total number of vehicles in the Republic of Mexico is 5 827 823 distributed as follows:

Cars	4 254 880
Trucks	1 489 143
Buses	83 000

The number of private cars is about 1 car per 17 inhabitants. Total number of cars in Mexico City is 1 600 000.

Annual petrol consumption is about 20 million cubic metres. The lead concentration in the petrol has been decreasing slowly from 0.98 g Pb/l in 1981 to 0.64 g Pb/l in 1982 and 0.2 g Pb/l in 1984. Based on these lead concentrations in petrol it can be calculated that annual lead emission from car traffic was about 19 600 tons before 1981, 12 800 tons in 1982 and 4 000 tons in 1984.

In 1982 Mexico City with an area of 1500 km^2 and slightly less than 30% of the total number of cars had a lead emission from car exhaust of about 2.3 metric tons/km^2 and year.

The present investigation was carried out in 1983, i.e. before the last decrease in the concentration of lead in petrol.

2.6.4 Sweden

The population of Sweden in 1982 was 8 327 000 inhabitants; population density 17.1 persons/km^2. Stockholm, an administrative center, is located on the coast of the Baltic Sea at a latitude of 59° north and a longitude of 18° east. The metropolitan area has 1 402 000 inhabitants (286 persons/km^2). There are no lead-emitting industries in the metropolitan area. The climate is of the temperate type

with an average temperature of +18°C in July and −3°C in February. Annual precipitation is around 550 mm.

Traffic density and lead emission

There are about 3 million cars in Sweden, i.e. about 1 car for every 3 inhabitants. In metropolitan Stockholm there are 444 000 cars, about 1 car for every 3 inhabitants.

Since the first of July 1981 the maximum allowable lead concentration content is 0.15 g Pb/l for both regular and premium petrol.

About one seventh of the total number of private cars are located in the metropolitan area of Stockholm; total area 4900 km^2. In 1982 the lead emission from car exhaust was about 0.02 metric tons/km^2 and year.

Since 1968 the lead content in petrol has evolved according to the data presented in Table 2:1.

Table 2:1 Average lead content in petrol in Sweden

Year	All types of petrol Pb g/l	Petrol consumption m^3	Lead content metric tons
1968	0.63	3 824 950	2410
1969	0.52	3 752 547	1950
1970	0.50	3 984 649	1990
1971	0.51	4 111 561	2099
1972	0.43	3 946 204	1689
1973	0.34	4 449 921	1507
1974	0.37	4 206 154	1540
1975	0.34	4 403 515	1497
1976	0.35	4 675 939	1637
1977	0.33	4 612 154	1522
1978	0.32	4 476 247	1432
1979	0.29	4 651 163	1349
1980	0.24	4 599 505	1100
1981	0.18	4 367 972	800
1982	0.15	4 712 000	705

Chapter 3

Methodology and Study Implementation

3.1 Target group

Nonsmoking males, from Belgium, Mexico and Sweden, were selected from the groups studied within the first UNEP/WHO Biological Monitoring Project. As Malta did not participate in the UNEP/WHO project, 36 nonsmoking male teachers were selected from that country.

A total of 107 male subjects participated in the study including:

- Belgium: 25 teachers from Brussels aged 26 to 42 years; most of whom participated in the first biological monitoring project.
- Malta: 36 teachers aged 30 to 39 years.
- Mexico: 19 subjects from Mexico City (16 teachers) aged 35 to 51 years; most of whom participated in the first project.
- Sweden: 27 subjects from Stockholm aged 24 to 50 years, all of whom participated in the first project.

3.2 Questionnaires

Two detailed questionnaires were prepared. The first one, a general questionnaire, was designed to provide detailed information about identity, personal history, consumption of food and beverages, health status, recreation activities, and housing conditions.

The second questionnaire was more specific for the test period. This questionnaire was completed by subjects the day before and during the 3 days of feces collection; it provides detailed information on food and beverages consumed during this period. This questionnaire was also used as the basis for selection of food samples collected in Malta.

3.3 Biological samples

3.3.1 Blood

3.3.1.1 Sampling

Sampling was carried out from November 1983 to January 1984. One blood sample was taken from the cubital vein of each of the 107 subjects. In order to avoid clotting the sample was taken not sooner than two hours after a major meal. Blood lead concentrations were determined for each sample.

In Belgium and Malta, 10 ml samples of blood were collected for metal analysis and transferred to two 5 ml tubes containing 5 mg (K_2Mg — EDTA) and one 10 ml plain tube. The samples were then mixed by inverting the tubes ten times. The samples were stored in a refrigerator until analysis. Time of sampling to time of analysis was never more than seven days.

In Sweden and Mexico, 10 ml blood samples were collected in green stoppered Venoject evacuated blood collection tubes T-200KA, each containing about 140 units of sodium heparin (Terumo Corp., Japan). The skin was cleaned before sampling using Medi Swab (Medi-Pack Ltd., Surrey, U.K.) The blood/heparin was mixed by inverting ten times, transferred into three acid-washed polyethylene tubes and stored at $-20°C$.

3.3.1.2 Analysis

Lead was determined by atomic absorption spectrophotometry (AAS). The samples from Belgium and Malta were analyzed at the IHE in Brussels. Mexico and Sweden analyzed their own samples.

The Institute of Hygiene and Epidemiology (IHE), Brussels, used a Perkin-Elmer, model 5000, atomic absorption spectrophotometer equipped with a Zeeman background corrector, a HGA 500 graphite furnace (ETA), AS-40 autosampler, EDL power supply, model 056 strip-chart recorder and PRS printer. Fifty microliters of whole blood were diluted in 450 μl Triton X-100 (0.2%). The analyses were performed using l'Vov's platform in a pyrolytic graphite tube and peak height evaluation. Calibration solutions were prepared by additions of known amounts of lead to human blood. For details of the method see Claeys (1982).

Karolinska Institute (KI), Stockholm, used a Perkin-Elmer 373 AAS equipped with a deuterium background correction system, HGA 500, AS-40, PRS 10 printer and a two-channel recorder model 056. The Institute in Mexico used a Perkin-Elmer 703 AAS apparatus equipped with a deuterium background correction system, HGA 400 (ETA) unit, AS-1 automatic sample injector and a recorder.

Sample treatment in Mexico and Sweden was carried out according to the method described by Stoeppler et al. (1978) and Stoeppler and Brandt (1980), modified by Lind (Elinder et al., 1983; WHO, 1983). A sample of 0.3 ml blood was deproteinized by the addition of 0.5 ml 0.8M nitric acid. The supernatant was analyzed for lead using standard graphite tubes and displayed peak area evaluation. Calibration solution was prepared by addition of known amounts of lead to cow blood.

3.3.2 Feces

3.3.2.1 Sampling

Each subject submitted the total feces amount from three consecutive days (72 hours) in conjunction with the blood sampling. Seventy-two-hour samples were chosen, since 72 hours is the average fecal transit time for Europeans (Burkitt et al., 1972). White plastic bags were put near the toilet seat and subjects were asked to defecate straight into them. The subjects were instructed to avoid urinating into the bags. The plastic bags were sealed with a rubber band and immediately put into a

plastic jar with a tight push-on lid as well as a tight screw-on lid and stored in a cool place until transportation. A record of the time of day defecation took place was kept by each subject. On arrival at the participating laboratories the total weight of the plastic bags and their contents were determined before storage. The Belgian and Maltese samples were stored at +4°C until analysis, Mexican and Swedish samples at −20°C.

3.3.2.2 Analysis

Belgian and Maltese samples were analyzed at IHE, Brussels, according to the following procedure. The feces sample was carefully mixed by squeezing the bag by hand, from the outside. A hole was then cut using scissors and four 10 to 15 g subsamples were placed into 4 different crucibles. These samples were dried to constant weight at 110°C.

Wet Ashing The dried matter of each sample (from the 4 crucibles) was pulverized and homogenized in a Fritsch apparatus. Each sample was then stored in a plastic container. After redrying of the powder, 100 mg of each sample were weighed in a Teflon tube to which 2 × 1 ml of concentrated nitric acid (Aristar grade, BDH, U.K.) was added. The Teflon tube was put in a programmable heater block (Coreci Palcor type RNS 2 R4) and progressively heated to 80°C; when the powder was completely dissolved, 0.8 ml of hydrogen peroxide was added to each tube and the temperature progressively brought to 100−110°C until complete dryness (about 20 to 24 hours).

ETA-AAS The dry residue was dissolved in 2 ml nitric acid 0.5%. An adequate dilution (usually tenfold) of this solution was injected into the graphite tube of the AAS. The apparatus and the conditions for lead determination were the same as those used for the blood analysis at IHE. The lead concentration of each sample is determined by internal standard additions of lead 10, 20 and 40 μg Pb/l. Two independent standard additions are made to each sample on the same day. Each sample is then analyzed at least three times on independent days.

Sweden and Mexico analyzed their own samples according to a method derived from Kjellström et al. (1978). Mixing of feces was carried out as described for Belgium. Three subsamples of 10 g each were placed into crucibles.

Dry-Ashing The samples were dried at 105°C and then dry-ashed overnight in a time/temperature programmable muffle furnace (Carbolite, model food-ashing) at 450°C after which the ash was dissolved in 15 ml 1M HNO_3. The solution was transferred to glass tubes and stored covered in a refrigerator at +4°C until analysis. In Mexico, the temperature of the muffle furnace (Thermolyne F-A1730) was slowly raised, manually, up to 450°C to simulate the time/temperature programme used in Sweden.

ETA and Flame AAS All the samples in Sweden were first analyzed by ETA-AAS, using a method of standard addition and with the same equipment as described for the PbB determinations. Due to high background absorption and very strong interferences, which depressed the signal by at least a half, all samples were also analyzed by flame AAS utilizing a Perkin-Elmer 403 instrument with a carefully optimized deuterium background correction system and a recorder (PE 56). Detection limit ($\bar{x}_{blank} + 3\ SD_{blank}$) for flame AAS was between 49−59 ng Pb/g acid

solution, resulting in a detection limit in the subsample of about 75 ng Pb/g wet weight. Accuracy and precision were much better using flame AAS than with ETA-AAS, hence, Swedish data are based on flame AAS. Mexico used mainly flame AAS due to the high concentrations of lead in feces.

3.3.2.3 Calculation of daily excretion

The concentration of lead in each feces sample is deduced from the standard curve. This allows calculation of the total lead content in feces of each subject for each day.

In most cases there was at least one feces sample for each of the three days of collection, and the mean daily feces lead content was easily calculated, by dividing the sum by three.

A few special cases were observed. For example, if there were feces samples from day 1 or day 1 and 2 only, these were considered as a one-day and two-day sampling period, respectively. On the other hand, if the samples were obtained on days 2 and 3 only, these were still considered to represent a three-day sampling period.

3.4 Environmental samples

Drinking water and dust samples were taken in all 4 countries and analyzed for lead content. In addition, air samples of suspended and sedimented particles, street dust, soils, food and beverages were taken in Malta in order to explore the reasons for the high blood lead levels observed (Bruaux et al., 1983). This will be discussed in Chapter 6 "Extended study in Malta".

3.4.1 Water

3.4.1.1 Sampling

Water samples were taken during the period of feces collection. The first 250 ml of water drawn in the morning from the cold water tap in the kitchen of each subject's home in Belgium, Malta and Sweden was collected in an acid-washed polyethylene bottle.

In Mexico most people have both municipal water supplies and their own reserve systems. Average results of several morning samples, from both supplies, were used.

3.4.1.2 Analysis

For Belgium and Malta, all samples were acidified with nitric acid and then analyzed by direct ETA-AAS. Results are taken from direct reading of a standard curve (0.5% nitric acid).

In Sweden and Mexico, levels of lead in water were determined by direct ETA-AAS using the method of standard addition.

3.4.2 Dust

3.4.2.1 Sampling

At least two samples were taken from each participant's home, one from outdoors and one indoors.

In Belgium and Malta, indoor dust samples were taken from the vacuum cleaner. The material collected was often mainly composed of fibres, thread, hairs or fur. The outdoor sample was taken from the window sill using a brush.

In Sweden and Mexico, outdoor samples were taken from the window sill or balcony (mean 0.10 g) and the indoor samples were taken at the top of the inner doors (mean 0.12 g). The dust was wiped off using two separate brushes, one for indoor and one for outdoor samples. The brushes were cleaned after taking each sample. In Sweden, the dust was collected in acid-washed and pre-weighed 50 ml glass beakers and after determining wet weight stored covered until analysis.

In Mexico, the dust was collected in plastic bags and transferred to crucibles before analysis.

3.4.2.2 Analysis

In all four countries, the lead concentration in dust were determined by flame AAS.

In Belgium and Malta, after drying, the samples were homogenized in a mortar and sieved through a stainless steel sieve of mesh size 500 micrometres. Wet ashing was carried out according to the method described for feces analysis.

The analysis was made without deuterium background correction, using a standard curve (0.5% nitric acid).

In Sweden the samples, together with four empty beakers as blanks, were dried overnight in an oven at +100°C; dry weights were determined the following morning. To all the beakers 0.5 ml concentrated HNO_3 were added and the samples were allowed to evaporate in an oven overnight.

The samples and blanks were then dry-ashed, treated and analyzed in the same way as described for feces with flame AAS.

For Mexico, samples were analyzed as described for Sweden, although no acid was added to the dust samples before the thermal treatment in the furnace. The same technique was used as described for feces analysis by flame AAS.

3.5 Statistical methods for evaluation of the relationship between lead in feces and lead in blood

3.5.1 Linearity

In order to evaluate lead intake by means of the relationship between daily feces lead and lead in blood, different approaches might be used. Most previous investigators found a curvilinear relationship. The first step for this study then will be to exclude a linear relationship between lead in feces and lead in blood. This was done by calculating the arithmetic means and standard deviations for lead in blood and lead in feces for the four countries. In this way four points, one for each country, could be identified with their respective uncertainty intervals in all direc-

tions (see Section 5.1.3 and Figure 5.3). The best line through these four points was computed taking into account the different standard errors and correlations. If there is a linear relationship the sum of the squared differences from the line to the country averages is chi-square distributed with two degrees of freedom which can be tested.

3.5.2 Regression analysis

Both PbB (lead in blood) and PbF (lead in feces) are often considered to be lognormally distributed. From a purely mathematical point of view a multiplicative power relationship giving a straight line on a log-log scale might be used. If, in the future, the x-values are determined in the same way and if they are derived from the same population, ordinary classical linear regression can be used. A line relating a hypothetical "true" x-value, measured without any random measurement error, to the y-value cannot be determined without further assumption on the model. This line describes to what extent the blood concentration changes as the daily intake of lead changes. The function can lie anywhere between the regression line of y on x and the regression line of x on y. One method to estimate the function is to minimize the total mean square residual in x and y, i.e. orthogonal regression. This method assumes that the errors in the estimations of x- and y-values are of the same magnitude and is, therefore, of particular use for intercomparative studies. In our study it is obvious that there are errors in the estimation of x-values (feces lead), both analytically and in the representativeness of our three-day samples. It is also obvious that there are errors in y-values (blood lead levels). At a given "true" intake the effect on blood lead can vary due to individual differences in, for example, metabolism, hematocrit, mineral deficiencies, or as a result of exposure via, e.g. inhalation.

Without knowledge of the errors in x and y it is impossible to choose the correct regression model. Earlier studies have used classical regression. In this study we present data from both classical and orthogonal regression analyses.

Chapter 4

Quality Assurance

4.1 Preanalytical quality control

Biological samples may be contaminated from various sources, e. g. through use of unsuitable blood collecting tubes and/or anticoagulants (Zief and Mitchell, 1976; Nise and Vesterberg, 1978). Furthermore, contamination may originate from the skin if not properly cleansed or from contaminated swabbing solutions (Bratzel and Reed, 1974). To avoid sample contamination all persons involved in sampling and analysis were given precise instructions in order to minimize the risk for contamination. All material used for sampling was checked for heavy metals contamination before use, and if necessary, washed with diluted nitric acid and deionized water.

4.2 Blood

Blood lead analyses were performed in accordance with the analytical quality control (QC) methods used in the biological monitoring programme for heavy metals exposure (Vahter, 1982; Friberg and Vahter, 1983). QC samples were prepared at KI. Internal QC samples were incorporated in each run of monitoring samples. At least one set of external QC samples was analyzed at each laboratory. All were accepted (see Figures 4.1—4.3).

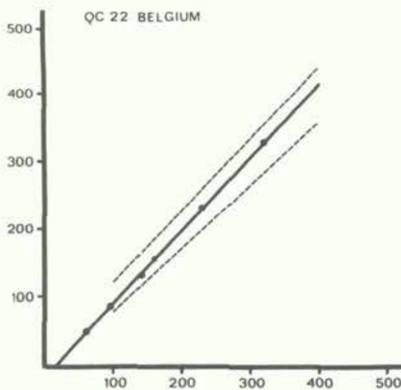


Figure 4.1. Results (μg Pb/l) from blood quality control run in Belgium.
y-axis: reported values; x-axis: reference values. Solid line is calculated regression line. Dotted lines indicate acceptance interval. Accepted.

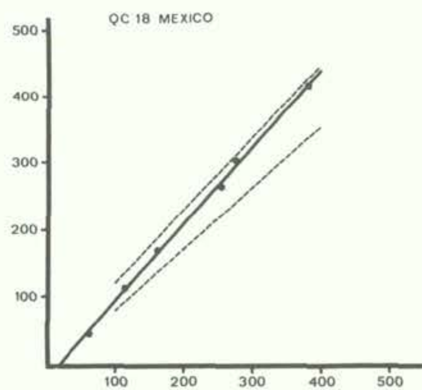


Figure 4.2. Results (μg Pb/l) from blood quality control run in Mexico.
y-axis: reported values; x-axis: reference values. Solid line is calculated regression line. Dotted lines indicate acceptance interval. Accepted.

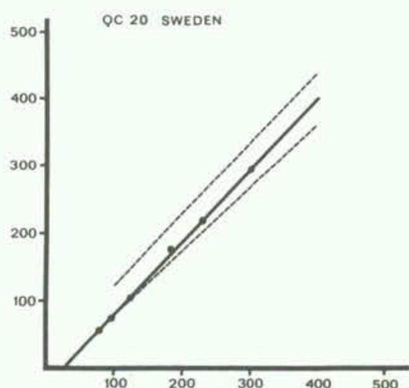


Figure 4.3. Results (µg Pb/l) from blood quality control run in Sweden.
y-axis: reported values; x-axis: reference values. Solid line is calculated regression line. Dotted lines indicate acceptance interval. Accepted.

Intercomparison of Monitoring Samples About 20 % of the blood samples were exchanged between KI and IHE. The agreement between the two laboratories was very good (see Figure 4.4).

An intercomparison was also made between KI and the Mexican laboratory; one of the three aliquots of each Mexican blood sample was analyzed at KI. The agreement was very good (see Figure 4.5).

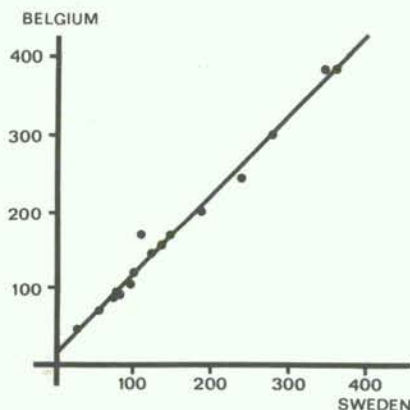


Figure 4.4. Results (µg Pb/l) of an intercomparison of monitoring blood samples between Sweden and Belgium. Line represents orthogonal regression analysis. Belgium = 1.04 x Sweden + 15. r = 0.99, n = 16.

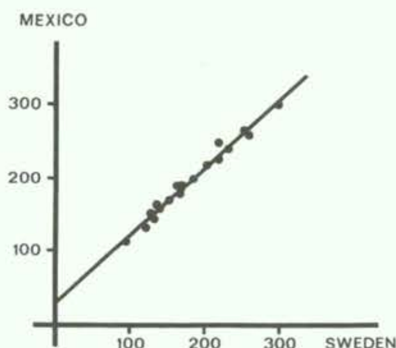


Figure 4.5. Results (µg Pb/l) of an intercomparison of monitoring blood samples between Sweden and Mexico. Line represents orthogonal regression analysis. Mexico = 0.92 x Sweden + 29. r = 0.99, n = 19.

It was decided to accept the results of the analyses for lead in blood found at each participating laboratory.

4.3 Feces

No standard quality assurance methods are available.

Intercomparison of Monitoring Samples About 10% of the samples were exchanged between IHE and KI (dried feces) and between KI and Mexico (dry-ashed feces).

It must be stressed that the methods used were ETA-AAS for IHE and flame AAS for KI. In Sweden, as discussed in Section 3.3.2.2, accuracy and precision were found to be better using the flame rather than ETA-AAS. There was strong interference giving depression of signals with ETA-AAS as well as a high background absorption. At IHE, different equipment for ETA-AAS was used including l'Vov's platform, Zeeman background corrector and better optics. The intercomparison between IHE and KI was very good (see Figures 4.6—4.7), we concluded, therefore, that the results are both reliable and comparable.

Mexico used mainly flame AAS due to anticipated high concentrations. There was a systematic difference between Mexico and Sweden (see Figure 4.8). Mexican

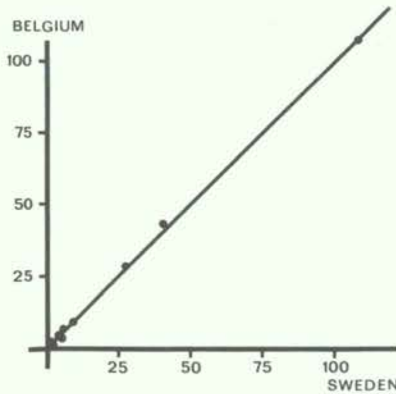


Figure 4.6. Results (µg Pb/g dry weight) of an intercomparison of monitoring feces samples between Sweden and Belgium. Line represents orthogonal regression analysis. Belgium = 0.99 x Sweden + 0.5. r = 1.00, n = 15.

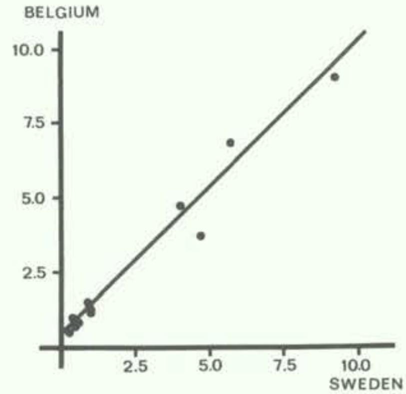


Figure 4.7. Results (µg Pb/g dry weight) of an intercomparison of monitoring feces samples between Sweden and Belgium. Close-up figure on low level concentrations. Line represents orthogonal regression analysis. Belgium = 0.99 x Sweden + 0.5. r = 1.00, n = 15.

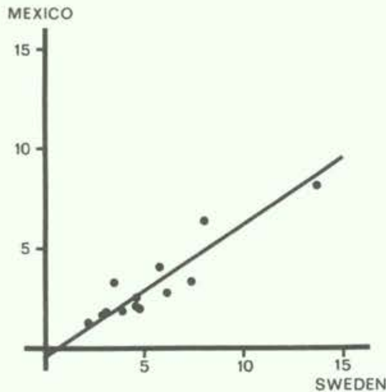


Figure 4.8. Results (µg Pb/g dry weight) of an intercomparison of monitoring feces samples. Line represents orthogonal regression analysis. Mexico = 0.66 x Sweden — 0.4. r = 0.91, n = 13. Note the systematic difference between Mexico and Sweden.

data on feces lead concentrations were adjusted to agree with the results from Sweden, as the Swedish data in the interlaboratory comparison with Belgium had proven to be reliable.

4.4 Water

Intercomparison. For the Belgian and Maltese samples the results of water analyses are those of direct readings on a standard curve. For Sweden and Mexico, the results given are derived from the method of standard addition, which resulted in somewhat higher values of lead concentration. Both these methods were used in Sweden and Mexico to arrive at this conclusion. KI analyzed about 1/3 of the Mexican water samples and good agreement was found. The levels of lead found in Belgian and Maltese samples should be considered minimum levels only.

4.5 Dust

The method of sampling for outdoor dust was the same in all four countries, but differed for indoor dust between Belgium-Malta and Sweden-Mexico.

Thirteen samples of dust were exchanged and analyzed by KI and IHE. The agreement between IHE and KI values is relatively poor (see Figure 4.9). The reason for this poor agreement is, possibly, lack of homogeneity of the samples; moreover, samples were small and, therefore, in some cases analysis had to be performed on subsamples which were far too small for accuracy. Flame AAS was also performed differently. No intercomparisons between Mexico (flame AAS) and Sweden were performed. For these reasons the results of analyses of lead in dust are to be considered as uncertain.

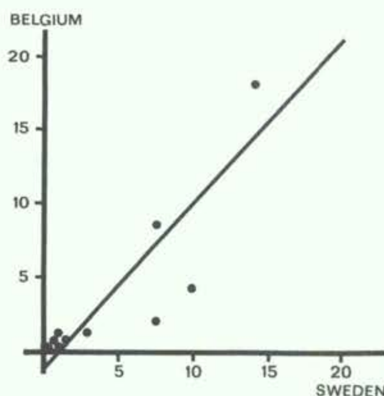


Figure 4.9. Results (µg Pb/g dry weight) of an intercomparison of monitoring dust samples between Sweden and Belgium. Line represents orthogonal regression analysis. Belgium = 1.12 x Sweden − 1.1. $r = 0.87$, $n = 13$.

Chapter 5

Results

5.1 Biological samples

5.1.1 Blood (PbB)

The results of blood lead analyses are given in Table 5.1 and Figure 5.1. For comparison, results for 1981 from Vahter (1982), from Bruaux et al. (1983) and from Claeys et al. (1983) are all included in the table. The results are comparable, since the analyses in 1981 and 1983 were carried out by the same laboratories with the same quality control programme and on comparable subjects; nonsmoking adult males. The only differences are related to age of subjects and time of year when sampling was carried out. All age groups were included in the 1981 study. In 1983 age groups were limited to include only subjects 26—42 years of age in Belgium, 30—39 years of age in Malta, 35—51 years of age in Mexico and 24—50 years of age in Sweden.

Blood lead levels are significantly different in the four countries. Median values for 1983 were 53 μg Pb/l in Sweden, 135 μg Pb/l in Belgium, 188 μg Pb/l in Mexico and 247 μg Pb/l in Malta. Blood lead levels appear to be 15—30 % lower in 1983 compared to the results obtained in 1981. Individual blood lead results for 1983 are listed in the Appendix.

Table 5.1. Blood lead concentration (μg Pb/l). A comparison between Belgium, Malta, Mexico and Sweden, 1981—1983.*

Country	Year	*n*	Median	P90	Mean	SD	GM	GSD
Belgium	1981	50	162	217	165	39	160	1.3
	1983	25	135	165	137	19	136	1.2
Malta	1981	195	293	455	307	119	288	1.4
	1983	36	247	343	243	70	232	1.4
Mexico	1981	21	255	401	269	77	259	1.3
	1983	19	188	263	195	48	189	1.3
Sweden	1981	31	77	147	83	38	75	1.6
	1983	27	53	114	59	31	51	1.7

* for explanation see text

Abbreviations:

n number of subjects
P90 90th percentile
SD standard deviation
GM geometric mean
GSD standard deviation of geometric mean

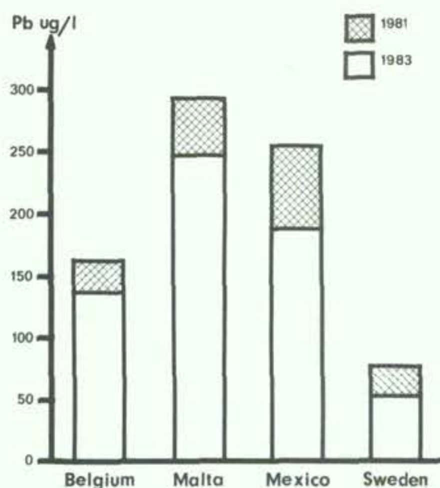


Figure 5.1. Lead in blood (μg Pb/l; (median values). Comparison between Belgium, Malta, Mexico and Sweden (1981—1983). Nonsmoking males.

5.1.2 Feces (PbF)

Total feces weight in g/day fresh weight (mean ± SD) and the ratio dry to wet weight (mean ± SD) were 140 ± 55 and 0.22 ± 0.04 for Belgium; 162 ± 73 and 0.21 ± 0.06 for Malta; 198 ± 75 and 0.20 ± 0.05 for Mexico; and 162 ± 62 and 0.24 ± 0.05 for Sweden. Feces lead was measured to estimate oral exposure. The results of analyses of lead in feces μg Pb/g wet weight, μg Pb/g dry weight and μg Pb/day are given in Tables 5.2—5.4 and Figure 5.2. As can be seen there are

Table 5.2. Lead in feces (μg Pb/g feces wet weight). A comparison between Belgium, Malta, Mexico and, Sweden, 1983.

Country	*n*	Median	P90	Mean	SD	GM	GSD
Belgium	25	0.79	1.34	0.84	0.38	0.76	1.6
Malta	36	2.63	6.16	3.33	2.92	2.48	2.2
Mexico	19	0.89	1.62	0.91	0.34	0.86	1.4
Sweden	27	0.13	0.44	0.18	0.12	0.15	1.8

Table 5.3. Lead in feces (μg Pb/g feces dry weight). A comparison between Belgium, Malta, Mexico and Sweden, 1983.

Country	*n*	Median	P90	Mean	SD	GM	GSD
Belgium	25	3.2	5.3	3.7	1.9	3.4	1.5
Malta	36	11.1	32.3	14.8	10.8	11.8	2.0
Mexico	19	4.7	6.3	4.8	1.6	4.6	1.3
Sweden	27	0.6	1.7	0.8	0.6	0.7	1.8

Table 5.4. Lead in feces (μg Pb/day).
A comparison between Belgium, Malta, Mexico and Sweden, 1983.

Country	*n*	Median	P90	Mean	SD	GM	GSD
Belgium	25	82	252	116	101	92	1.9
Malta	36	361	1 281	505	434	372	2.2
Mexico	19	159	258	170	66	157	1.5
Sweden	27	22	63	31	27	23	2.1

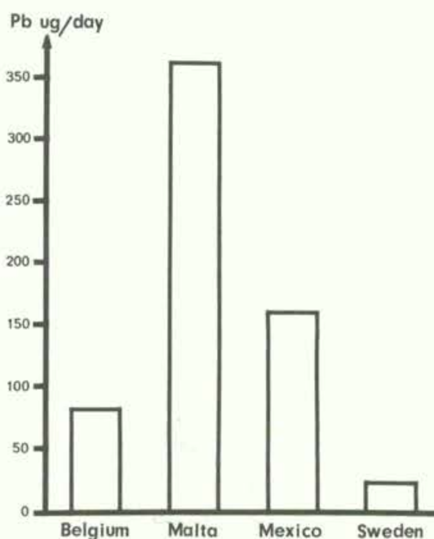


Figure 5.2. Lead in feces (μg Pb/day; (median values). Comparison between Belgium, Malta, Mexico and Sweden. Nonsmoking males.

considerable differences in feces lead levels between the four countries, with median values of 22 μg Pb/day in Sweden, 82 μg Pb/day in Belgium, 159 μg Pb/day in Mexico and 361 μg Pb/day in Malta. These differences are statistically significant. Individual levels of lead in feces in μg Pb/g dry weight feces and μg Pb/day are listed in the Appendix.

5.1.3 The relationship between lead in feces and lead in blood

To exclude a linear relationship between lead in feces and lead in blood the method described in Section 3.6.1 was used. A chi-square value of 14.3 with two degrees of freedom was obtained. Thus, the average for the four different countries does not lie on a straight line, $p < 0.01$ (Figure 5.3). This may be due to differences between the four countries or a nonlinear relationship between PbF and PbB.

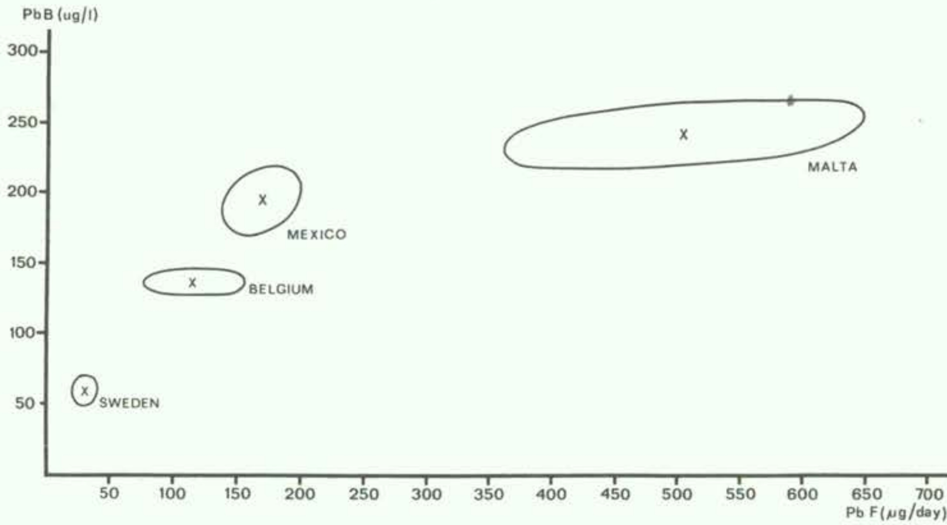


Figure 5.3. Arithmetic averages for lead in feces (µg Pb/day; PbF) and lead in blood (µg Pb/l; PbB) in all countries and 95% values in all directions. The ellipses cover the true average with probability 86%.

The basic fit of the curves tested was; $y = a + bx$, $y = ae^{bx}$ $(a > 0)$, $y = a + b \ln x$ and $y = ax^b$ $(a > 0)$, respectively. The best correlation found was for $y = ax^b$, where a and b are constants, y = log PbB and x = log PbF.

To monitor oral intake we used lead in feces per day.

In Tables 5.5—5.8 the equations for the orthogonal and classical regression lines

Table 5.5. Relationship between feces lead (µg Pb/day) and blood lead (µg Pb/l) using orthogonal regression analysis.

$y = A + Bx$, where A and B are constants;
y = log PbB; x = log PbF.

Country	*n*	r	A	SD_A	B	SD_B
Belgium	25	0.05^{NS}	1.69	0.09	0.22	0.05
Malta	36	0.43^{2}	1.31	0.17	0.41	0.06
Mexico	19	0.36^{NS}	0.89	0.32	0.63	0.14
Sweden	27	0.46^{1}	0.72	0.18	0.73	0.13
All countries combined	107	0.81^{3}	1.00	0.07	0.55	0.03

1. $p < 0.02$
2. $p < 0.01$
3. $p < 0.001$

NS. Nonsignificant

Table 5.6. Relationship between feces lead (µg Pb/g dry weight) and blood lead (µg Pb/l) using orthogonal regression analysis.

y = A + Bx, where A and B are constants;
y = log PbB; x = log PbF.

Country	n	r	A	SD_A	B	SD_B
Belgium	25	-0.07^{NS}	1.96	0.04	0.32	0.07
Malta	36	0.55^{2}	1.86	0.08	0.48	0.07
Mexico	19	0.58^{1}	1.69	0.12	0.88	0.17
Sweden	27	0.53^{1}	1.86	0.05	0.99	0.17
All countries combined	107	0.86^{2}	1.81	0.02	0.58	0.03

1. $p < 0.01$
2. $p < 0.001$

NS. Nonsignificant

Table 5.7. Relationship between feces lead (µg Pb/day) and blood lead (µg Pb/l) using classical regression analysis.

y = A + Bx, where A and B are constants;
y = log PbB; x = log PbF.

Country	n	r	A	SD_A	B	SD_B
Belgium	25	0.05^{NS}	2.11	0.09	0.01	0.05
Malta	36	0.43^{2}	1.92	0.17	0.18	0.06
Mexico	19	0.36^{NS}	1.78	0.32	0.23	0.14
Sweden	27	0.46^{1}	1.26	0.18	0.33	0.13
All countries combined	107	0.81^{3}	1.21	0.07	0.45	0.03

1. $p < 0.02$
2. $p < 0.01$
3. $p < 0.001$

NS. Nonsignificant

Table 5.8. Relationship between feces lead (µg Pb/g, dry weight) and blood lead (µg Pb/l) using classical regression analysis.

y = A + Bx, where A and B are constants;
y = log PbB; x = log PbF dry feces.

Country	n	r	A	SD_A	B	SD_B
Belgium	25	-0.07^{NS}	2.14	0.04	−0.02	0.07
Malta	36	0.55^{2}	2.09	0.08	0.26	0.07
Mexico	19	0.58^{1}	1.94	0.12	0.51	0.17
Sweden	27	0.53^{1}	1.79	0.05	0.53	0.17
All countries combined	107	0.86^{2}	1.85	0.02	0.50	0.03

1. $p < 0.01$
2. $p < 0.001$

NS. Nonsignificant

between PbB and PbF micrograms in feces per day and PbB and PbF (dry weight) are presented.

Figures 5.4—5.6 present orthogonal and classical regression lines between PbF (per day) and PbB. The correlations between PbF (wet weight) and PbB are

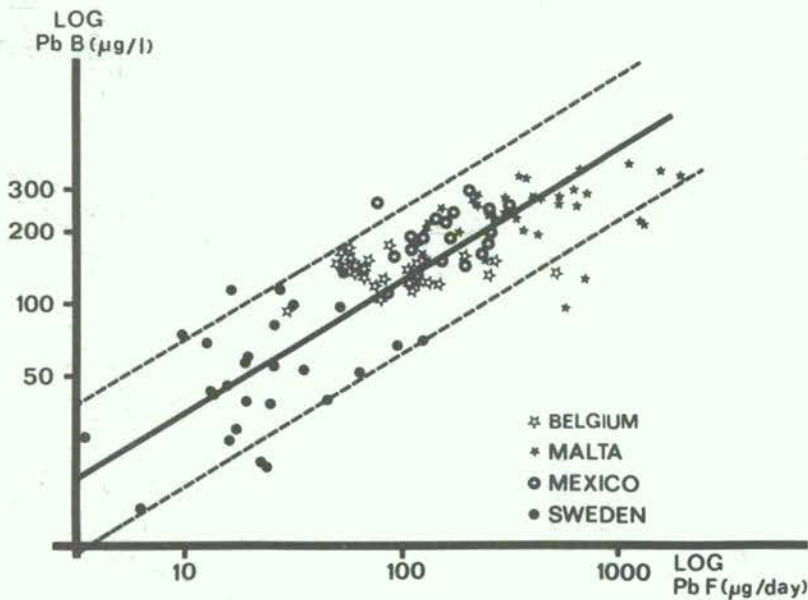


Figure 5.4. Correlation between lead in feces (μg Pb/day; PbF) and lead in blood (μg Pb/l; PbB) in Using orthogonal regression analysis and 90% confidence range (broken lines). r = 0.81, n = 107. Log PbB = 1.00 + 0.55 log PbF.

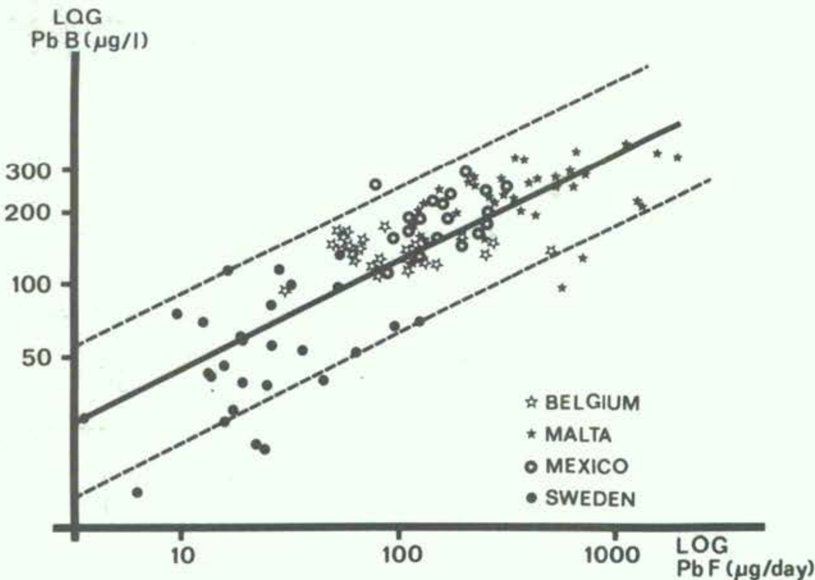


Figure 5.5. Correlation between lead in feces (μg Pb/day; PbF) and lead in blood (μg Pb/l; PbB) Using classical regression analysis and 90% confidence range (broken lines). r = 0.81, n = 107. Log PbB = 1.21 + 0.45 log PbF.

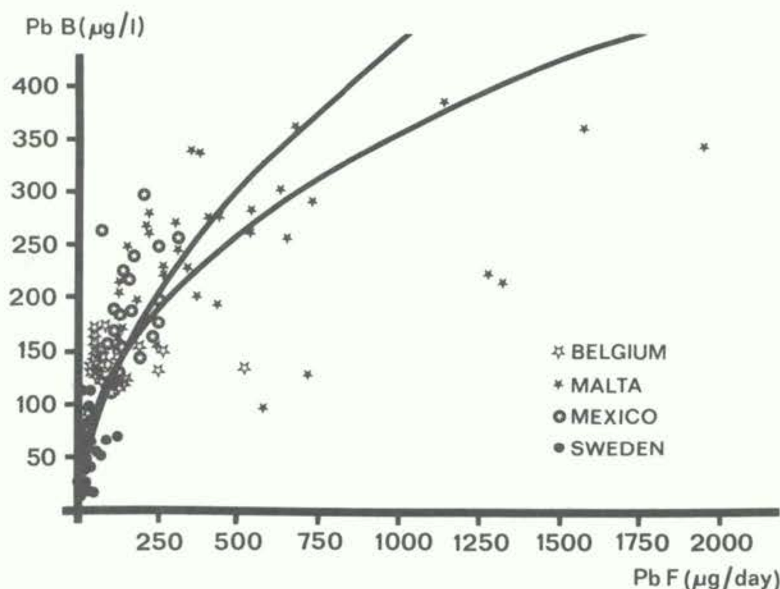


Figure 5.6. Lead in blood (µg Pb/l; PbB) as a function of lead in feces (µg Pb/day; PbF) Lines represent, the left, orthogonal regression analysis; the right, classical regression analysis. Correlation between PbB and PbF. r = 0.81, n = 107.

generally poorer than those for PbF (dry weight) and PbB and PbF (µg Pb/day) and PbB.

The best correlation coefficients are found for PbF (dry weight) and PbB. This could be due to incomplete collection of feces or the composition of diet influencing the rate of lead absorption.

5.2 Environmental samples

5.2.1 Water

At least one water sample was collected from each participant's home in all four countries. This sample was taken mainly from the kitchen cold water tap, as "first draw water", in the morning. The results of the water lead analyses are given in Table 5.9.

Table 5.9. Lead concentration in "first draw" water (µg *Pb/l).*
A comparison between Belgium, Malta, Mexico and Sweden, 1983.

Country	*n*	Median	P90	Mean	SD	GM	GSD
Belgium	25	9	30	14	15	8	3.0
Malta	36	1	4	2	1	2	1.8
Mexico	19	1	3	2	1	1	1.7
Sweden	27	⩽3	⩽3	⩽3	—	—	—

In Malta, Mexico, and Sweden, the lead concentrations in water are very low, less than 3 ug Pb/l. In Belgium, water lead does, to a minor extent, contribute to the increased blood lead levels.

5.2.2 Dust

Dust samples were collected from each participant's home, one from inside the house or flat (indoor dust) and one from the outside (outdoor dust). It should be recalled that the sampling method was the same for outdoor dust (window sill or balcony), but different for indoor dust in Belgium/Malta (vacuum cleaner) and in Sweden/Mexico (sweepings from the top of the inner doors).

Quality control by intercomparison of analyses between KI and IHE showed that analytical problems still remain, probably due to lack of homogeneity and to too small samples as well as differences in the AAS technique used (see Sections 3.4.2 and 4.5).

The results of indoor dust and outdoor dust lead analyses are given in Table 5.10.

Table 5.10. Indoor and outdoor dust lead concentrations (µg Pb/g). A comparison between Belgium, Malta, Mexico and Sweden, 1983.

Country	*n*	Median	P90	Mean	SD	GM	GSD
*Indoor dust**							
Belgium	25	146	448	281	500	132	3.2
Malta	35	380	4 178	1 468	3 552	416	4.0
Mexico	16	533	1 034	587	303	516	1.7
Sweden	27	360	850	440	263	362	1.9
Outdoor dust **							
Belgium	24	367	6 072	1 795	3 083	582	4.1
Malta	35	555	2 050	1 035	1 555	577	2.7
Mexico	19	733	1 531	822	418	725	1.7
Sweden	27	1 096	2 263	1 231	897	993	2.0

* Sampling method different for Belgium/Malta — Mexico/Sweden (see text)
** Same method of sampling

Table 5.10 shows that dust lead concentrations differ greatly, not only between the four countries, but also among the samples taken in each country, as indicated by the high standard deviation. This fact, as well as the remaining analytical problems, make interpretation of the data somewhat hazardous. However, a comparison of the results from the four countries shows that the lead concentrations in indoor dust are lower than the concentrations in outdoor dust.

Chapter 6

Extended Study in Malta

This study was carried out as preliminary survey to identify the possible environmental sources of the high blood lead levels found. In order to investigate some environmental sources samples of ambient air, street dust, soil and food were analyzed. Lead levels in these samples were compared to those of corresponding Belgian samples.

6.1 Suspended and sedimenting particles in ambient air

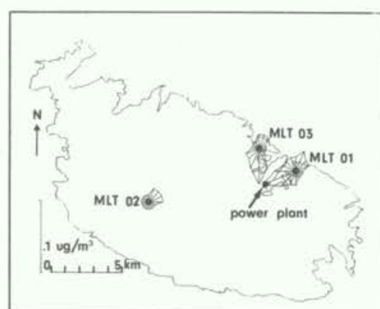
6.1.1 Sampling

Three air sampling stations similar to those used in Belgium were installed in Malta, one (MLT 01) monitoring a residential/industrial area, one (MLT 02) monitoring background levels and one (MLT 03) monitoring an urban area with heavy traffic. Twenty-four-hour samples of suspended particles were taken using low volume samplers at breathing height. The flow volume was 12 liters/min through a Sartorius 0.45 μm cellulosenitrate membrane filter (Ralph et al., 1982). The diameter of the collected particles was below 15 μm with the equipment used and, thus, corresponds to the respirable fraction. On top of each sampling station deposit gauges of NILU type (IBN, 1976) were installed. Thirty-day samples of sedimenting particles were taken.

To evaluate the influence of a single source of emisson the individual mean values of metal concentrations for each wind direction were calculated. The calculation procedure called "pollution roses" is described in Barnes and Egleton (1977). The length of the radius of each sector of the "pollution rose" is proportional to the average air metal concentration in that particular wind direction.

6.1.2 Analysis

For suspended particles wavelength dispersive X-ray fluorescence is used without any pretreatment of the sample filters. A number of elements (As, Cd, Cr, Cu, Mn, Ni, Pb, V, Zn and SO_4^{2-}) were analyzed. Figures 6.1 and 6.2 present results for lead and vanadium. (Other results are available from IHE upon request). Sedimented particles were analyzed using a wet chemical technique followed by atomic absorption spectrophotometry.



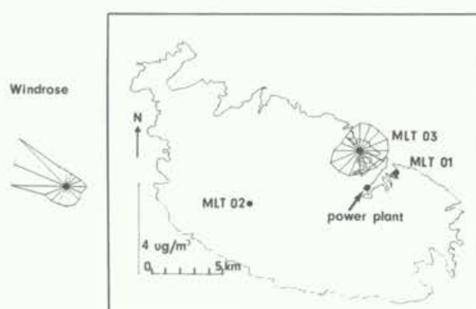


Figure 6.1. Pollution roses for vanadium in Malta, Sept. 1, 1983—August 31, 1984, windrose to the right.

Figure 6.2. Pollution roses for lead in Malta, Sept. 1, 1983—August 31, 1984,windrose to the left.

6.1.3 Results

Table 6.1 presents results of the measurement of lead in suspended particles for a one-year period, September 1, 1983 to August 31, 1984 for Maltese and Belgian sampling stations using similar equipment at comparable locations. The classification residential/industrial corresponds in Malta to a residential area close to a shipyard. No precisely comparable station was to be found in Belgium. Figures 6.1 and 6.2 are "pollution roses" for vanadium and lead. The different sampling stations in Malta are indicated in the figures as well as the only power plant. From Figure 6.1 it is clear that the power plant is the main source of vanadium. As can be seen in Figure 6.2 no single source of lead can be pinpointed. A single source close to station MLT 03 is still a possibility. The most reasonable explanation is, probably, that car exhaust accounts for the high lead levels found around station MLT 03.

Table 6.2 presents the one-year mean (September 1, 1983 to August 31, 1984) results for samples of sedimenting particles in Maltese and Belgian sampling stations using similar equipment at corresponding locations.

Table 6.1. Air lead concentrations as suspended particles (daily means and 95-percentiles in µg Pb/m³). A comparison between Malta and Belgium.

	Malta			Belgium		
Sampling area	Station	Mean	P95	Number of stations	Mean	P95
Residential/ industrial	MLT 01	0.17	0.52	18	0.37	0.89
Background	MLT 02	0.08	0.17	4	0.12	0.33
Heavy traffic	MLT 03	1.17	2.15	3	1.02	1.98

Table 6.2. Air lead concentrations as sedimenting particles, deposit gauges (monthly means in µg Pb/m² and day). A comparison between Malta and Belgium.

	Malta		Belgium	
Sampling area	Station	Mean	Number of stations	Mean
Residential/ industrial	MLT 01	162	11	229
Background	MLT 02	148	1	109
Heavy traffic	MLT 03	570	1	1860

6.2 Soil

6.2.1 Sampling

Eleven topsoil samples were analyzed for Malta, five of them taken less than one meter from the edge of a road and six 5—40 meters from the edge of a road.

6.2.2 Analysis

The analyses were performed at the Belgian Ministry of Agriculture using flame atomic absorption spectrophotometry (flame AAS) on a strong acid extract of the soil samples.

6.2.3 Results

Lead concentration in the soil samples ranged from 29 to 367 µg Pb/g dry material, mean 92 µg Pb/g, geometric mean 70 µg Pb/g, geometric SD 1.96 (Table 6.3). Five topsoil samples in Malta were taken less than one meter from the edge of a road; mean lead content 138 µg Pb/g. Six samples were taken 5—40 meters from the edge of a road; mean lead content 54 µg Pb/g. As Table 6.3 shows soil close to roads is more contaminated than soil at a distance from roads.

Table 6.3. Lead concentrations in topsoil from Malta (µg Pb/g dry material) taken at different distances from roads.

	Less than 1 meter from the edge of the road	5 to 40 m from the edge of the road
		29
	367	54
	64	42
	81	39
	116	79
	60	79
Mean	138	54

6.3 Street dust

6.3.1 Sampling

Eight surface dust samples were collected in Malta and ten comparable samples were taken in Belgium.

6.3.2 Analysis

Analysis was carried out at IHE using flame atomic absorption spectrophotometry (flame AAS) as described for dust (see Section 3.4.1).

6.3.3 Results

Table 6.4 presents levels of lead in street dust at heavy (n = 4, mean 1828 μg Pb/g) and low (n = 4, mean = 502 μg Pb/g) traffic intensity sites in Malta. Two samples, from a wall and a balcony in an urban area with heavy traffic, gave a mean of 2655 μg Pb/g dry material. Table 6.5 presents lead levels in street dust samples taken in Belgium at heavy traffic intensity sites (n = 3, average 2255 μg Pb/g) and low traffic intensity sites (n = 7, average 371 μg Pb/g).

Table 6.4. Lead concentrations in street dust in Malta (μg Pb/g dry material) from areas with heavy and low traffic.

	Heavy traffic	Low traffic
	1925	631
	1545	925
	2240	366
	1600	86
Mean	1828	502

Table 6.5. Lead concentrations in street dust in Belgium (μg Pb/g dry material) from areas with heavy and low traffic.

	Heavy traffic	Low traffic
	1650	452*
	2015	297
	3099	291
		613*
		118
		558*
		267
Mean	2255	371

* Rather medium than low; moderate traffic in the morning and at night.

6.4 Food

6.4.1 Sampling

In order to investigate whether food items are a source of lead exposure in Malta, samples of several foodstuffs were collected. Sampling was carried out in two phases. In the first phase about 40 samples were collected using the market basket approach. Samples were collected, based on the answers to the questionnaire on food consumption, which was answered the day before and during the feces collection period. The food basket consisted of 5 samples of flour and pastas, 2 of butter, 18 of different drinks, 5 of meat, 4 of fish, 4 of vegetables and 3 of milk. Since preliminary results from analyses of lead content in vegetables in Malta showed higher levels compared to those found in Belgium, the second food sampling phase focused on vegetables including potatoes and carrots. In this phase 84 vegetable samples (cabbages, carrots, potatoes, lettuce, snowball cabbages and tomatoes) were obtained from three sources in Malta: from the fields, from a vegetable market and from shops. For comparison, 56 equivalent samples were taken in Belgium.

6.4.2 Sample treatment

6.4.2.1 Solid food samples

The unwashed samples were first cut into small pieces using a stainless steel surgical scalpel and were then distributed into crucibles in aliquots of 30 – 60 g. The wet weight was determined and the samples were dried to constant weight at 110°C. After cooling the dry weight was determined and the dry residue was homogenized in a Fritsch homogenizer and stored, as a fine powder, until analyzed.

After redrying, wet ashing was carried out according to the technique described for feces (see Section 3.3.2.2).

6.4.2.2 Liquid samples

Liquid samples were diluted in 0.5% nitric acid.

6.4.2.3 Analysis

Analysis was carried out using electrothermal atomic absorption spectrophotometry (ETA-AAS). Samples with high lead concentrations were analyzed by flame AAS. The lead concentration of each sample was determined by the method of internal standard addition. Each sample was analyzed at least twice on two different days.

6.4.3 Quality assurance

Internal quality assurance was performed by introducing three NBS standards: Citrus leaves (No. 1572), spinach leaves (No. 1570) and tomato leaves (No. 1573) (see Table 6.6). For intercomparison some vegetables were also analyzed at two

Table 6.6. Results of IHE lead analysis (µg Pb/g dry weight) of NBS standard reference material.

	NBS reference values Mean ± SD	IHE values
Spinach No. 1570	1.2 ± 0.2	1.2, 1.0
Citrus leaves No. 1572	13.3 ± 2.4	12.1, 9.3
Tomato leaves No. 1573	6.3 ± 0.3	5.5, 4.3, 8.5

other Belgian laboratories. The first intercomparison was made with the laboratory of "Faculté des Sciences Agronomiques de l'Etat à Gembloux". This laboratory performed wet ashing using nitric and perchloric acid and measured lead content by differential pulse anodic stripping voltammetry (DPASV). Figures 6.3–6.4 present the results of this intercomparison. The equation appear to indicate that the results obtained using DPASV are systematically lower than those obtained with AAS, if the highest value is excluded this difference becomes even more pronounced. The agreement between the two laboratories is poor. It is not clear which results are most correct.

The second intercomparison was made with the laboratory of "Institute de Recherches Chimiques—Tervueren" (IRC). The sample treatment, wet ashing, was made at IHE for all samples and the comparison presented is simply a comparison between flame AAS at IRC and ETA-AAS at IHE. The regression line is presented in Figure 6.5. There was good agreement between flame AAS and ETA-AAS.

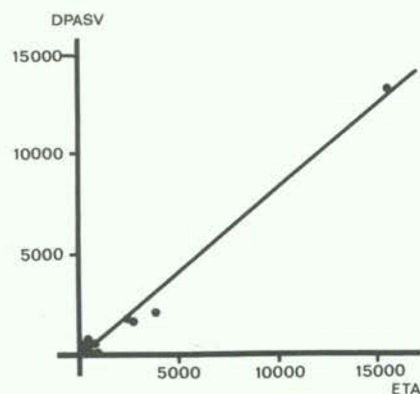


Figure 6.3. Results (µg Pb/g dry weight) of an intercomparison between ETA-AAS and Differential Pulse Anodic Stripping Voltammetry (DPASV) on food samples. Line represents orthogonal regression analysis. DPASV = 0.83 x ETA – 74. r = 0.99, n = 18.

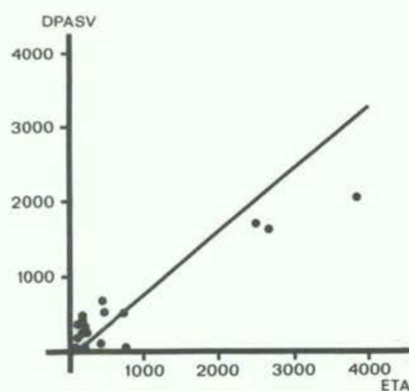


Figure 6.4. Results (µg Pb/g dry weight) of an intercomparison between ETA-AAS and Differential Pulse Anodic Stripping Voltammetry (DPASV) on food samples. Close-up on the low level concentration. Line represents orthogonal regression analysis. DPASV = 0.83 x ETA – 74. r = 0.99, n = 18.

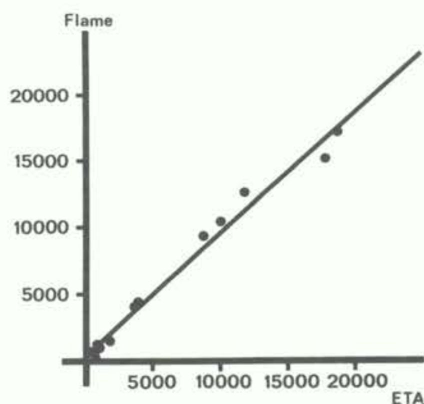


Figure 6.5. Results (µg Pb/g dry weight) of an intercomparison between flame AAS and ETA-AAS on food samples. Line represents orthogonal regression analysis. Flame = 0.91 x ETA + 417. r = 0.99, n = 12.

These intercomparisons seem to indicate that there are still a number of analytical problems in food analysis. The problems are probably more often related to the ashing method than to the final analysis. From these quality assurance results it is obvious that caution must be exercised in the interpretation of results from the food analyses.

6.4.4 Results

Table 6.7 presents data on lead concentrations in the market basket samples collected in Malta during the first phase of food sampling. The table also contains some data on concentrations in corresponding Swedish food samples (Jorhem et al., 1984).

Some preliminary results, which revealed high lead concentrations in vegetables, prompted us to perform a second sampling phase focused on vegetables.

Table 6.7. Lead concentrations (means, µg Pb/kg or µg Pb/l fresh weight) in Maltese and Swedish foodstuffs. (Swedish data derived from Jorhem et al.,1984).

Sample	n[1]	Malta	Sweden
Flour	5[2]	110[2]	9 (mixed flour)
Pastas			16
Butter	2	31	63
Wine	6	105	79
Soft drinks	10	23	6
Beer	2	2	2–16
Canned fish	3	76	580 (average)
Fresh fish	1	34	50 (herring, cod)
Canned meat	3	377	410 (corned beef)
Fresh meat	2	27	100 (round beef)
Milk	3	10	6

1. Malta Only
2. Flour and pasta taken together for Malta

Table 6.8. Lead concentrations (means and ranges, μg Pb/kg fresh weight) in vegetables from Malta, Belgium and Sweden. (The Swedish data are from Jorhem et al., 1984 and are based on peeled and washed samples).

	Malta			Belgium			Sweden		
Food	*n*	Mean Pb	Range	*n*	Mean Pb	Range	*n*	Mean Pb	Range
Cabbages	12	38	6–162	8	93	22–153	24	12	3–27
Carrots	12	163	25–389	12	55	25–118	47	21	4–14
Potatoes	12	94	35–267	8	34	14–72	63	15	6–39
Lettuce	12	371	53–1276	16	405	142–559	31	43	3–160
Snowball cabbages	12	77	9–259	–	–	–	–	–	–
Tomatoes	24	12	6–26	12	12	8–15	30	15	6–44
Total	84	110		56	150			21	

Equivalent samples were collected in Belgium. Table 6.8 presents analytical results for the second sampling phase in Malta and Belgium as well as data from the study by Jorhem et al. (1984) on Swedish samples. The Swedish results are based on peeled and washed vegetables while those in Malta and Belgium are based on unpeeled and unwashed vegetables. Washing and peeling decreases lead concentrations in vegetables by 25–75% (Larsen, 1980; Slorach, personal communication). Table 6.7 shows lead concentrations in samples from the Maltese market basket. It is obvious that most food items seem to have lead concentrations in the same range or somewhat lower than those in Sweden. However, the most important food ingredients in Maltese diets are flour and pastas, both of which had higher lead concentrations than corresponding Swedish samples. Table 6.8 shows lead concentrations in vegetables and it can be seen that there is, in general, no difference in lead concentrations for Malta and Belgium, although potatoes and carrots have lead levels that are three times higher in Malta than in Belgium. Examination of the literature shows that Swedish values are generally much lower, but these values are based on analysis of peeled and washed samples and are thus not directly comparable.

6.5 Summary

As can be seen from the results of analyses of the lead content in suspended and sedimenting particles, street dust and soil (Tables 6.1–6.5), no major differences between Malta and Belgium were revealed. However, the tendency of the Maltese to lead a more outdoor life might result in higher exposure from outdoor sources. Some lead concentrations found in samples of flour and pastas, as well as in potatoes and carrots seem to be higher in Malta. This is of interest since all these foodstuffs are major ingredients in the Maltese diet. As indicated by the quality assurance results, considerable difficulties still remain in the analysis of lead in food. The comparison of vegetable lead levels between Malta and Belgium, on the one hand, and Sweden on the other hand is further hampered by the fact that in Malta and Belgium the samples analyzed were unwashed and unpeeled while the Swedish samples were washed and peeled. Washing decreases lead concentrations in vegetables by 25–75% (Larsen, 1980; Slorach, personal communication). On

the basis of the available data we are unable to identify a single environmental source for the high levels of lead in feces and blood. There is, however, an indication that food is the principal exposure source. The reason for the relatively high lead content in certain food items is unknown. Further analyses of lead in food samples are needed.

Chapter 7

Discussion

7.1 The relationship between lead in feces and lead in blood

In the present study we compared blood lead levels and feces lead levels in four countries, Belgium, Malta, Mexico and Sweden. Blood lead is the best indicator of recent exposure and feces lead is a measure of oral intake. One venous blood sample and all feces from three consecutive days were collected from each subject. The feces samples were collected in plastic bags which had been checked for lead contamination. The representativeness of the feces values as an indication of individual exposure is limited due to: (1) limited sampling period (3 days) and (2) possible differences in absorption rate, depending on composition of ingested material as well as possible individual factors. However, feces lead provides a fairly good measure of oral intake due to the relatively low absorption of ingested lead, at least in adults. It should be kept in mind though that the scatter occasionally leads to large uncertainties.

Blood lead levels for the different countries are given in Table 5.1 and Figure 5.1. The median values were quite different for the different countries, with lowest values in Sweden followed by Belgium, Mexico and Malta. The values for feces lead content in μg Pb/g wet weight and dry weight and μg Pb/day are given in Tables 5.2–5.4. Again the median values varied greatly between the countries, increasing in the same sequence as for blood lead (Figure 5.2). If we assume a gastrointestinal absorption of 10%, the daily oral intake in Belgium would be about 90 μg. This is in agreement with Buchet et al. (1981), who reported a daily intake via food of 96 μg Pb/day. Using the same assumption for Sweden, the daily intake would be about 24 μg Pb/day, which is in agreement with data from Schütz (1979) who found dietary lead content to be 30 μg Pb/day, and data from Slorach et al. (1983) who found values of 27 μg Pb/day in a typical Swedish diet. In Mexico, the oral intake would be 177 μg Pb/day. A preliminary estimate made by the National Mexican Food Monitoring Program (unpublished data, provided by General Directorate of Research on Health, Vice Ministry of Research and Development, Ministry of Health) is close to 200 μg Pb/day. A similar validation cannot be made for Malta as there is no direct estimate of lead intake via food.

Both PbB and PbF are often considered to be lognormally distributed. In our study we tried to compare exposure to lead between the four countries and to relate exposure, through the oral route, to blood lead levels in the four different populations taken together. Using different statistical approaches a curvilinear relationship was obtained, thus, the increase in blood lead levels did not increase proportionally to the increase in feces lead levels. This indicates a dose-related metabolism, but only on the assumption that the populations are comparable, i.e. without differences in other exposure sources, genetics, metabolism, hematocrit, mineral

deficiencies, or any other factors that would affect the relationship between exposure and blood lead levels. Individual exposure to lead via air was unknown. Inhalation leads to differences in exposure since there are different air lead levels in the four countries. However, inhalation as a source of exposure is of less significance than exposure via the oral route.

This study tried to select test subjects that were as comparable as possible. However, we had no knowledge of differences in genetics or metabolism between the populations studied. Analyses of serum iron, hematocrit and blood-hemoglobin were performed on the Belgian and Maltese populations. No systematic differences were revealed between those two countries. No such analyses were performed in Mexico or Sweden. The same tendency towards a curvilinear relationship between PbF and PbB as that found in the four populations taken together was found in the Maltese subpopulation alone. In the Belgian, Mexican and Swedish populations the spread in exposure levels is small within the different groups, which makes these population groups unsuitable for individual regression analysis.

Our results indicate, thus, a dose-related metabolism. The results are in agreement with earlier published results by, for example, Department of the Environment Central Directorate on Environmental Pollution (1982); Sherlock et al. (1982) and the Nutrition Foundation's Expert Advisory Committee (1982), i.e. the relationship between PbF and PbB is curvilinear and $\frac{\Delta \text{PbB}}{\Delta \text{PbF}}$ is dependent on where on the curve the subject falls. Our study has the advantage of including groups with both low, intermediate and high exposure levels. No data on low exposure levels are available from, for example, the studies mentioned above. Furthermore, the design of our study included an extensive quality assurance programme.

Without knowledge of the errors in feces and blood lead it is impossible to choose the correct regression model for analysis of the relationship. Earlier studies have used classical regression. In this study we present data both from classical and orthogonal regression between log PbF and log PbB, as we do not know the magnitude and distribution of random errors in either PbF or PbB (see Section 3.5.2).

7.2 Specific sources as contributors to exposure

7.2.1 Lead in water

Lead levels in "first draw water" are given in Table 5.9. The median values were 9, 1, 1, and <3 μg Pb/l for Belgium, Malta, Mexico, and Sweden, respectively. In Belgium, lead concentration in water is several times higher than in the other countries and may be a significant source of exposure. In the other countries water contributes very little to exposure.

7.2.2 Lead in dust

As discussed in Chapter 4, Section 5, Quality assurance, there were serious problems with dust lead analysis. The intercomparison was poor, probably due to the limited amount sampled and the lack of homogeneity within the sample.

Furthermore, for indoor dust, different sampling techniques were used for Belgium and Malta, on the one hand, and Mexico and Sweden, on the other hand. Results are given in Table 5.10.

Due to these analytical problems no individual comparisons of lead in dust can be made. Caution must be exercised also in the interpretation of averages. Median values of lead in outdoor dust were 367, 555, 733 and 1096 μg Pb/g for Belgium, Malta, Mexico, and Sweden, respectively. The highest median value was found in Sweden. Therefore, in this study, outdoor dust seems to have been of no importance for blood lead levels. Lead content in outdoor dust appears to be higher than in indoor dust. For indoor dust two comparisons between median lead values can be made, Belgium—Malta 146 and 380 μg Pb/g, respectively, and Mexico—Sweden 533 and 360 μg Pb/g, respectively. Levels of lead in indoor dust seem to be higher in the country with higher blood lead levels in each comparison. This factor may be one of several contributors to exposure.

7.2.3 Lead in air

Air lead analyses within this project were carried out only in Malta and Belgium (Section 6.1). Similar concentrations, about 1 μg Pb/m^3, were observed in areas with heavy traffic in both countries. These values may be compared with some data from Sweden and Mexico, obtained from routine monitoring programmes.

Data on suspended particle analyses from Stockholm 1982—1983 (unpublished data, provided by the Municipal Environmental Health Administration) show a range of 0.1 to 1.2 μg Pb/m^3. Results from a National Monitoring Program in Mexico City (unpublished data, provided by General Directorate of Research on Health, Vice Ministry of Research and Development, Ministry of Health) indicate a yearly average, for 1982, of 2.4 μg Pb/m^3 and a range of 0.6 to 5.7 μg Pb/m^3. Sedimenting particles were analyzed only in Malta and Belgium (Section 6.1). There were no tendencies towards higher levels in Malta than in Belgium.

Chamberlain (1983) suggested, based on epidemiological as well as experimental data, that an increase in air lead of 1 μg Pb/m^3 would result in an increase of 15—30 μg Pb/l blood. Assuming air lead levels to be 1.0 μg Pb/m^3 for Belgium, 1.2 for Malta, 2.4 for Mexico and 0.45 for Sweden, the contributions to blood lead would be 15—30, 18—36, 36—72 and 7—14 μg Pb/l, respectively. These values are equivalent to 11—22% of the median blood lead values for Belgium, 7—15% for Malta, 19—38% for Mexico, and 12—25% for Sweden.

7.2.4 Lead in soil and street dust

In normal noncontaminated soil lead content ranges, according to Shacklette (1971), from less than 10 μg Pb/g to 70 μg Pb/g. Swedish soils contain 2—364 μg Pb/g with a mean of 16 μg Pb/g (Andersson, 1977).

The soil and dust samples taken in Malta indicate lead concentrations higher than those in noncontaminated soil and dust, however, in eleven soil samples the concentration of lead ranged from 29 to 367 μg Pb/g, with a geometric mean of 70 μg Pb/g (Section 6.2). As could be expected soil close to roads was more contaminated than soil further away from roads. No firm conclusion on the magnitude of contamination of soil in Malta can be drawn.

Four street dust samples (heavy traffic) from Malta (Section 6.3) had concentrations ranging from 1545—2240 μg Pb/g dry material, mean 1828 μg Pb/g. Four street dust samples (low traffic) had concentrations ranging from 86—925 μg Pb/g dry material, mean 502 μg Pb/g. Lead concentrations in the analyzed street dust samples are in the same order of magnitude as those for Belgium; mean value for low traffic 371 μg Pb/g and for high traffic 2255 μg Pb/g dry material. In Lancaster, U.K. (Harrison, 1979) the corresponding figures were 570 and 2130 μg Pb/g, respectively.

The contribution of soil and dust to total exposure for adults is difficult to evaluate. Based on data from Stark et al. (1982) soil and indoor dust would contribute 20 μg Pb/l blood for every 1000 μg Pb/g, in children. For adults, these figures must be considerably lower. The contribution from street dust is even more difficult to evaluate.

7.2.5 Lead in food

The major exposure route for individuals not occupationally exposed is via the gastrointestinal route. This does not necessarily imply that gastrointestinal intake is solely due to exposure via food. Other possible sources of exposure, besides food, are water and other beverages, ingestion of nonfood lead substances, such as paint, or leaching of lead from pottery.

As can be seen from Table 6.7 "Lead concentrations in Maltese and Swedish foodstuffs" the lead concentrations in most Maltese samples are in the same range or lower than those for Sweden, except for flour and pastas where Malta had values of about 110 μg Pb/kg compared to only about 9 and 16 μg Pb/kg in Sweden. In Table 6.8 lead concentrations in vegetable samples are compared for Malta, Belgium and Sweden. Concentrations of lead are derived from unwashed and unpeeled samples in Belgium and Malta, but from washed and peeled samples in Sweden. Lead concentrations are higher in all vegetable samples from Malta and Belgium than in samples from Sweden, with the exception of tomatoes. This result could be due to the washing techniques. However, this would not explain why, for Malta, lead levels in potatoes and carrots are much higher than those for both Belgium and Sweden. Potatoes, carrots, flour and pastas are all major ingredients in the Maltese diet.

Based on the available data, it is assumed that food is the most probable source of the high blood lead levels in Malta, even though this cannot be proven. Further studies in Malta should investigate specific sources of exposure more thoroughly.

Chapter 8

Conclusions

This study confirmed the differences in blood lead levels found earlier between the four countries (Vahter, 1982; Bruaux, 1983; Friberg and Vahter, 1983). The study also indicates 15—30% lower levels than the results from previous studies carried out in these four countries. The median values found were 135, 247, 188 and 53 μg Pb/l blood, for Belgium, Malta, Mexico and Sweden, respectively. The range for blood lead values was 14—385 μg Pb/l blood.

The median values of lead in feces were also quite different with 82, 361, 159 and 22 μg Pb/day for Belgium, Malta, Mexico and Sweden, respectively. The range of feces lead for the four countries was 3.5—1950 μg Pb/day. Lead in feces is a good measure of oral intake since, in adults, only about 10% of ingested lead is absorbed by the gastrointestinal tract.

The relationship between feces lead and blood lead was not linear. Using different statistical approaches, classical and orthogonal regression analysis, curvilinear functions were obtained for the combined data from the four countries. For Malta, where the exposure as expressed by feces lead levels covered a wide range, a similar relationship was observed.

The data are in agreement with the results published earlier by, e.g. Department of the Environment, Central Directorate on Environmental Pollution (1982); Sherlock et al. (1982); and the Nutrition Foundation's Expert Advisory Committee (1982), i.e. the relationship between PbF and PbB is curvilinear and $\frac{\Delta \text{PbB}}{\Delta \text{PbF}}$ is dependent on where on the curve the subject falls. Our study has the advantage of including groups with both low, intermediate and high exposure levels. No data on low exposure levels are available from the other studies. Furthermore, the design of our study has included an extensive quality assurance programme.

Possible reasons for the curvilinear relationship are discussed. It is concluded that the results indicate a dose-related metabolism of lead.

The other major exposure route is through inhalation. Using the relationship between air lead levels and blood lead levels proposed by Chamberlain (1983), i.e. 15—30 μg Pb/l blood per μg Pb/m^3 in air, and the air levels found in urban traffic areas, inhalation would contribute 15—30 μg Pb/l blood in Belgium, 18—36 μg Pb/l blood in Malta, 36—72 μg Pb/l blood in Mexico and 7—14 μg Pb/l blood in Sweden. These values are equivalent to 11—22%, 7—15%, 19—38% and 12—25% of the median blood lead values found in the respective countries.

In the extended study in Malta we could not pinpoint a single environmental source for the high lead level in feces and, consequently, could not account for the high blood lead levels found. The most probable source, based on the data collected, is food. There are indications that certain foodstuffs are of particular importance. At this stage it is not possible to explain the cause of the relatively high

lead content in feces. Future studies in Malta should investigate possible sources of exposure more thoroughly.

This follow-up study, although partly of a pilot nature, has clearly shown the value of an integrated monitoring programme where biological monitoring forms the core. The study has also pointed to the difficulties involved in making a quantitative evaluation of the contributions from specific sources by routine environmental monitoring. More sophisticated environmental monitoring is needed as a complement to the biological monitoring.

Acknowledgements

Other participants in the project were:

Belgium: G. Ducoffre, M. Legrand, S. Hallez and E. Bonnyns; Institute of Hygiene and Epidemiology (IHE), Ministry of Health, Brussels

Malta: M. Sammut and L. Vella; Ministry of Health, Valletta

Mexico: A. Carboney, B. Silva, M.T. Roldan and S. Becera; General Directorate of Research on Health, Vice Ministry of Research and Development, Ministry of Health, Mexico City

Sweden: C. G. Elinder and R. Åberg; National Swedish Institute of Environmental Medicine and Department of Environmental Hygiene, Karolinska Institute (KI), Stockholm; Present address: National Board of Occupational Safety and Health, Solna, and LKB-Produkter AB, Bromma, respectively

The editors are also indebted to Daniel Thoburn, Department of Statistics, University of Stockholm, for his suggestions and statistical analysis.

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Appendix

Individual levels of lead in blood (μg Pb/1) and in feces (μg Pb/g dry weight and μg Pb/day) in Belgium, Malta, Mexico and Sweden

BELGIUM

No.	PbB (μg Pb/l)	PbF (μg Pb/g dry weight)	PbF (μg Pb/day)
1	120	4.4	146.7
2	128	2.2	63.0
3	125	3.7	82.0
4	135	4.5	110.7
5	169	1.8	55.1
6	159	3.9	57.6
7	154	4.3	194.3
8	137	1.6	54.1
9	124	3.0	111.4
10	131	7.4	252.4
11	94	2.0	30.0
12	118	3.1	76.0
13	145	5.2	120.5
14	117	3.4	112.2
15	134	3.5	60.2
16	165	2.6	54.7
17	111	3.1	83.5
18	174	2.3	87.8
19	150	5.3	266.9
20	123	4.7	132.6
21	142	3.1	57.7
22	146	2.2	50.4
23	140	2.4	64.3
24	150	3.2	69.6
25	134	10.4	518.1

MALTA

No.	PbB (μg Pb/l)	PbF (μg Pb/g dry weight)	PbF (μg Pb/day)
1	171	2.9	112.9
2	270	30.5	409.5
3	198	5.6	184.7
4	122	3.8	114.6
5	96	16.6	577.6
6	385	35.5	1137.3
7	359	49.6	1578.1
8	267	6.8	214.3
9	261	8.1	221.3
10	301	25.6	627.6
11	202	7.7	370.9
12	222	32.3	1281.6
13	339	10.9	350.7
14	156	5.7	243.6
15	262	18.7	536.9
16	290	16.5	727.4
17	158	4.9	126.9
18	343	37.8	1949.9
19	228	9.1	340.7
20	256	14.1	653.5
21	270	9.9	305.0
22	245	4.9	310.6
23	337	14.3	380.1
24	362	19.2	672.5
25	155	4.9	130.8
26	194	9.0	433.2
27	206	6.7	127.5
28	276	12.4	433.2
29	222	11.2	268.9
30	214	21.3	1325.0
31	248	7.8	153.3
32	283	17.8	538.8
33	215	10.9	133.3
34	279	14.9	222.9
35	229	8.6	263.3
36	127	17.3	717.4

MEXICO

No.	PbB (µg Pb/l)	PbF (µg Pb/g dry weight)	PbF (µg Pb/day)
1	239	6.3	173.5
2	263	4.2	77.1
3	198	5.7	258.4
4	163	5.3	234.1
5	217	4.3	159.3
6	188	4.7	168.5
7	190	3.3	111.5
8	248	5.2	253.0
9	225	4.5	143.4
10	152	3.4	150.8
11	177	5.9	250.3
12	157	3.8	93.1
13	297	5.3	204.2
14	144	3.6	196.6
15	257	9.8	314.3
16	131	4.8	119.3
17	187	5.1	125.7
18	168	3.2	110.5
19	112	2.8	86.0

SWEDEN

No.	PbB (µg Pb/l)	PbF (µg Pb/g dry weight)	PbF (µg Pb/day)
1	14	0.3	6.2
2	82	0.6	25.7
3	46	0.5	15.7
4	22	0.6	22.4
5	75	0.6	9.6
6	60	0.5	19.4
7	42	0.5	13.6
8	97	1.2	52.1
9	52	1.1	63.3
10	28	0.3	3.5
11	134	1.7	53.7
12	58	0.4	19.0
13	70	2.6	125.0
14	38	0.5	24.7
15	30	0.5	17.4
16	99	1.5	31.7
17	55	0.5	26.1
18	21	0.6	24.1
19	40	1.0	45.0
20	67	1.8	94.9
21	43	0.4	13.2
22	114	0.7	16.4
23	39	0.5	19.1
24	53	1.5	35.7
25	27	0.6	15.9
26	114	0.8	27.8
27	69	0.4	12.6