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A survey of human exposure to mercury, cadmium and lead in Greenland

Jens C. Hansen



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# A survey of human exposure to mercury, cadmium and lead in Greenland

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Analyses of lead, mercury and cadmium in tissues from seal and fish have shown high concentrations of mercury and cadmium. A toxicological evaluation of the actual concentrations has revealed that in some districts of Greenland, the population may exceed the provisionally tolerable weekly intake (WHO, 1977) of cadmium with from 2 to 20 times and of mercury with from 2 to 40 times. Lead intake was below the provisionally tolerable weekly intake. As these high dietary intakes might have adverse health effects in the consumers, an investigation was undertaken in order to evaluate the human exposure as reflected in blood and hair concentrations. Five districts in Greenland and a control group of Greenlanders living in Denmark have been examined.

A total of 144 persons (including the control group) have participated.

Samples were taken in September and October 1979.

Mercury. Strong evidence was found for a connection between mercury exposure and seal-eating. The mercury levels found indicate that the exposure calculated from food analyses is overestimated, but still the most highly exposed groups are on an

exposure level where subclinical effects may be anticipated.

Cadmium. In general the blood cadmium concentrations are higher in Greenland than in Denmark, but the groups in Greenland were found to be very similar. In hair concentrations no differences between the groups were observed. Separation of data on blood cadmium between smokers and non-smokers showed the differences between the mean values to be highly significant. In spite of the presumably higher dietary intake, no influence on blood concentrations could be observed. Contrary to blood, hair reflected dietary intake but not smoking. The results indicate that neither blood nor hair as only parameter reflects total cadmium exposure.

A positively significant correlation was demonstrated between lead and cadmium

concentrations in hair, but not in blood.

Lead. Blood concentrations were found to be at the same level as found in Western European countries, but all to be below the limit of 35  $\mu$ g/100 ml which is the upper individual limit in the EEC-countries.

The highest blood-values were found in the two northern districts, where the level is significantly higher than the level in the two southern districts. The difference was found to be related to varying eating habits, also smoking habits were found to be reflected in blood and hair. Blood was found to be a better index medium than hair for evaluating lead exposure.

Selenium. A potentially toxicity-modifying micronutrient selenium was determined in a limited number of hairsamples. No evidence of a high selenium intake could be

provided.

Further research is needed especially concerning mercury exposure. Concerning lead and cadmium, the levels found are well below what is regarded a critical level. As, however, the concentrations are on the same level as those found in industrialized countries, follow-up studies seem to be needed in order to observe trends of exposure.

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#### 1. Introduction

Due to the technological development human populations are increasingly exposed to environmental pollutants that are potentially determinant to health. Many of these, e.g. the heavy metals mercury, cadmium, and lead, have already proved to cause diseases among the most heavily exposed groups. As elements occurring naturally in the earth crust they are ubiquitous and thus also found in human tissues and excreta in natural

background concentrations. These background concentrations may vary according to geographical, geological, climatological and cultural conditions.

The distribution of pollutants in a population can be estimated through biological monitoring programmes, which are systematic collections of human or other biological specimens, for which analyses of pollutant concentrations, metabolites and products of biotransformation are of immediate application. Carried out in

areas not expected to be polluted, these programmes can provide information on background levels, and when this is known, provide direct evidence of undue exposure.

In arctic areas such as Greenland, which are remote from industrial areas, pollution should be expected to be minimal. Still, analyses of tissues from marine mammals caught along the coast of Greenland, have revealed surprisingly high concentrations of mercury and cadmium. As marine mammals in certain districts of Greenland form a major food item, there is in fact a risk of adverse effects due to high dietary intake of those two metals. Earlier sporadic analyses on human blood and hair have shown high mercury concentrations (Carstensen and Poulsen, 1973; Hansen and Nygaard, 1974). So, it seemed relevant and necessary to carry out a biological monitoring programme, and through this provide data to evaluate the dietary exposure to mercury and cadmium in the population of Greenland, and to determine to what extent the metals in the food are reflected in human index media such as blood and hair.

Beside mercury and cadmium, lead was included in the programme even though there was no evidence of specific exposure; but it was done because it made it possible to get an impression of lead exposure in arctic areas, which would be of a general scientific interest.

The programme was started in August 1979 and finished in December 1979.

# 2. Objective and rationale

A toxicological evaluation of existing data on concentrations of mercury, cadmium and lead in fish and marine mammals caught at Greenland has been carried out by the Danish National Food Institute (Miljøministeriet, 1978). In the report an estimate of exposure was made of both a normal and an excessive (normal  $+2 \times$  standard deviation) level of food intake on a low and on a high level of concentration. The results are summarized in table 1, in which the WHO provisional, tolerable weekly intake (PTWI) is also indicated (WHO, 1972).

The table shows that the cadmium intake from a normal food intake on a low level of concentration exceeds PTWI by a factor of 2, and by a factor of 4 at the high level of concentration. In cases of extreme food intake (normal  $+ 2 \times$  standard deviation), the factors are 10 and 25, respectively. For mercury the excess factors are 2 and 8 from a normal food intake and 9 and 45 from an extreme food intake. Regarding lead, the estimated intake even in cases of extreme food intake was below recommended values.

For comparison, in a number of countries dietary cadmium exposure has been estimated to vary between 0.056 and 0.392 mg per person per week (CEC, 1978). Dietary mercury intake varies considerably. In unpolluted areas with no tradition for eating fish, a person

Table 1. Calculated dietary exposure to mercury, cadmium, and lead in Greenland, mg/person/week.

Element	Low exposure	High exposure	PTWI*)
Mercury			
Normal food intake	0.59	2.39	0.3
Normal $+ 2 \times S.D.$	2.57	13.67	(max. 0.2 as methyl-Hg)
Cadmium			
Normal food intake	0.95	1.94	
Normal $+ 2 \times S.D.$	5.20	12.22	0.4-0.5
Lead			<u></u>
Normal food intake	0.38	1.05	
Normal $+ 2 \times S.D.$	1.01	2.86	3

<sup>\*)</sup> Provisional tolerable weekly intake.

will digest between 0 and 0.14 mg per week. In polluted areas like Sweden and Japan where much fish is eaten, the exposure will be considerably higher; from Sweden are reported weekly intakes between 0.07 and 0.399 mg and between 0.35 and 1.4 mg in Japan (CEC, to be published). The average lead intake via food is reported to be between 1.4 and 2.1 mg per person per week (WHO, 1977).

# Comments on toxicological significance of dietary metal intake with special regard to the estimates for Greenland

#### 3.1 Cadmium

Of cadmium taken via food approx. 5% will be absorbed (CEC, 1978). Cadmium accumulates in the kidney cortex, and it reaches a maximum at the age of 50 (Schroeder and Balassa, 1961). Based on animal experiments and existing epidemiological data it has been suggested that 200 mg/kg is a critical concentration (Friberg and Piscator, 1972), at higher concentrations tubular damages with proteinuria occur. The critical level is estimated to be reached at the age of 50 by 50% of a population with an average daily absorption of 20 µg, but 1% will still show symptoms at a daily average absorption of 2.7 µg (Kjellström, 1977b).

The estimated dietary intake in Greenland (table 1) corresponds with a daily absorption from 6.8 to  $87.3~\mu g$  which suggests that a considerable proportion of the population can be supposed to reach the critical level in kidneys at the age of 50. Cadmium-induced kidney damage is irreversible and is a precondition to secondary diseases. The intake level estimated for the population in Greenland should therefore be considered with great severity.

It should, in this connection, be underlined that in the above mentioned calculations, only dietary intake is considered and not cadmium taken from cigarette smoking which for the non-occupationally exposed part of the population is the dominating source.

#### 3.2 Mercury

Experiences from Minamata in Japan have indicated that signs of methyl mercury intoxication occur at a daily intake of 4  $\mu$ g/kg or 0.28 mg per 70 kg, or 1.96 mg per week/person (Berglund et al., 1971). The WHO recommended weekly maximum intake is thus based on a factor of safety of approx. 10.

The estimates in table 1 of mercury intake thus suggest that groups of the population in Greenland risk chronic mercury intoxication. In this connection, special attention should be paid to pregnant women, as in contrast to cadmium mercury (methyl mercury) is able to pass the placental barrier; and intrauterin mercury intoxication has been reported (Engelson and Herner, 1952; Harada, 1968; Matsumoto et al., 1965; Snyder, 1971).

#### 3.3 Lead

With regard to lead there is no reason to be concerned, as the estimated level of dietary intake must be regarded well below the critical value. As lead is a metal of great concern as an environmental pollutant trends of larger intake should, however, be followed.

There are many uncertainties in the above described calculations and estimates of dietary intake of the metals. Firstly, the analytical data on fish and marine mammals are not representative of the whole of Greenland. Secondly, the seasonal variation in food items eaten has not been taken into account, and finally, estimates of the total amount of food eaten by each person as well as the proportion of the individual items are extremely difficult.

The figures in table 1 are therefore only rough estimates, but still a serious indication of the existence of a potential risk to those of the Greenland population depending primarily on local seafood.

The objective of the programme was to investigate whether or not the dietary intake of mercury and cadmium is reflected in humans, and if so to an extent which should be regarded critical. Lead was included to obtain a basis level for lead in blood and hair from an arctic area as this information is of general scientific interest and furthermore should be valuable in evaluating levels found in industrialized areas.

# 4. General comments on biological monitoring

Systematic sampling and analysis of human or other biological specimens may help to understand the relationship between exposure to pollution and health hazards. Biological monitoring can make existing environmental (physical and chemical) monitoring systems more cost-effective by establishing direct or indirect links to humans. Pollution concentrations in the object of impact, the human being, can be related to sources of environmental concentration, and the populations at risk can be defined. The effectiveness of control measures may be assessed through subsequent periodical biological monitorings which will detect changes in levels of pollutants in man and thus be an early warning system.

#### 4.1. Index media

The ideal parameter for assessing human exposure is a measurement of the concentration of a pollutant in the critical organ (dosis). The critical organ or target organ is the organ or group of cells which accumulate the toxicant to the highest degree or are the most sensitive. The first signs and symptoms of intoxication can thus be expected to be due to malfunctioning of the critical organ.

In the case of the heavy metals, the critical organ for methyl-mercury is the central nervous system, for cadmium the kidneys and for lead the bone marrow and the peripheral nerves. This means that the target organs are not in all cases accessible to biopsies, and for this reason other easily obtainable tissues, body liquids or excreta, must be used on the condition that the concentration in these reflects bodyburden or dosis, i.e. a concentration in a critical organ. These specimens supposed to reflect dosis are index media. Under biological monitoring programmes, excreta are difficult to obtain, and therefore blood and hair are often the preferred index media.

#### 4.2. Blood

As to lead, blood is regarded as an index of bodyburden and as such is used as an index medium for evaluating the lead exposure in the general population within the European communities as fixed in the directive on Biological Screening of the Population for Lead of 27/3–1977 (CEC, 1977). Concerning mercury and cadmium, there is more doubt about the significance of blood concentrations, but for mercury the blood level is supposed to reflect recent exposures.

As such, blood data will be of value in surveys concentrated upon estimation of exposure. If the exposure is continuous and constant, bodyburden and health risks can also be predicted; while in case of discontinuous exposure at various exposure levels, the value of blood as an only parameter must be questioned. Regarding blood cadmium, only a limited information is available as to its use as an index medium. It is, however, supposed to be an indicator of recent exposure (Kjellström, 1977a).

#### 4.3 Hair

Hair has so far not been so widely used as an index medium as has blood, and at the moment is still a question of controversy. Analysis of scalp hair seems, however, with regard to contamination to be a suitable method of primary screening of a population, for trace element pollutants including the heavy metals (IAEA, 1978; Chattopadhyay, 1977; Henkins, 1979).

Several authors have demonstrated correlations between fish consumption and hair concentrations (Shishido and Suzuki, 1974; Yamaguchi et al., 1971, 1975). So, in a non-occupationally exposed population the level of mercury in hair is supposed – under normal conditions – to reflect mercury intake with food, in particular where the consumption of fish and other marine animals is of importance.

As most mercury in fish is in a methylated state (Westöö and Ohlin, 1975; Bache et al., 1975) hair mercury in the general population is believed to be mainly methyl mercury. Most studies on hair mercury do not distinguish between various compounds (inorganic-methyl mercury) but only report total mercury concentrations.

In a study of islanders from the Pacific, Shishido and Suzuki (1974) conclude that organic mercury increases the total mercury content in human hair. Yamaguchi et al. (1975) have reached the same conclusion.

That exposure to inorganic mercury can also contribute to hair concentrations has been shown in studies on Italian furnace workers producing metallic mercury and on mercury miners (Cigna Rossi et al., 1976; Gutemann et al., 1973), on personnel in dental clinics (Dale et al., 1975) and also on chemists working with polarography (Hansen, 1976). Creason et al. (1975) demonstrated that Ba, Hg, Pb and V environmental exposure gradients (as measured in dust-fall or house dust) were significally associated with concentrations in scalp hair of both children and adults.

Cadmium seems to be the element least examined as to the usefulness of hair as a monitoring medium. A few studies have, however, indicated the value of determining hair cadmium (Hammer et al., 1971, Aurand and Sonnenborn, 1973).

The advantages of using hair as an index medium are 1) that hair concentrations of the elements are higher than in other tissues (often several orders of magnitude). Obviously this gives a solution to the analytical problem involved by analysing near the detection limit which is often the case in analyses performed on blood from non-occupationally exposed persons. 2) Hair samples can be easily obtained without inconvenience for the donor, and 3) hair samples are easily posted and stored

The disadvantages are first of all the risk of exogen contamination. Secondly, there is a considerable intraindividual variation as all hair does not grow equally quickly. Some hairstrains are in a resting state (telogen phase) while others are in a growing state (anagen phase). Jaworowski et al. (1966) have shown that only hairstrains in the anagen phase incorporate lead.

The drawbacks can, however, be counteracted: the exogen contamination by a thorough preanalytical cleaning (Petering et al., 1971) and the intraindividual variation by using a standardized sampling technique by taking several subsamples each containing strains from different part of the scalp as e.g. described in the IAEA monograph on this topic (IAEA, 1978).

## 5. Programme design

Biological monitoring programmes may be designed for at least three purposes:

- to assess a general population exposure to one or several pollutants, thus obtaining baseline levels,
- to evaluate the situation where high exposure is indicated or suspected,
- to evaluate exposure in situations where toxic effects are present or suspected.

The present study was designed as a study of the first type. An evaluation of the results obtained will answer whether or not follow up studies of types 2 or 3 are needed.

As stressed by The international workshop on: The Use of Biological Specimens for the Assessment of Human Exposure to Environmental Pollutants organized by CEC, EPA and WHO (Berlin et al., 1979), in designing a monitoring programme the following general principles and guidelines need consideration:

- Studies should only be started where the overall purpose and objectives have been clearly defined, as it will determine the programme. – In casu the purpose and objectives were well defined through the demonstration of high dietary metal intake in Greenland which demanded a general survey of the exposure in the population.
- 2) The donors and biological specimens selected should be representative of the target population and bodyburden of interest. – The selection of donors will be discussed in a later section. As to the specimens selected (blood and hair) these have been discussed in the section on index media.
- 3) Detailed pertinent information regarding the donors and environmental conditions should be collected and recorded for each sample. – To fulfil this demand in the present study, an interview questionaire was filled out for each donor at the same time as samples were taken.
- 4) Special attention should be paid to specimen collection as regards the use of trained staff, choice of

container and transport conditions. – All samples in the Greenland survey were taken by trained laboratory technicians at local hospitals in the districts. Prior to sampling the technicians were especially instructed in the use of bloodsampling device (Terumo® Venoject) and in filling out the questionaire. The vacutainer system for bloodsamples was chosen because it minimizes the risk of contamination, and because the vials are resistant to changes in temperature and atmospheric pressure occurring during transportation by air. Hair samples were placed in clean polyethylene bags.

- 5) Storage conditions for the specimen will be determined by the pollutant being studied. Metal content in hair will not be changed by time, but still all hair samples were analysed within 2 months from sampling. Blood samples will be stable for at least one month when kept in refrigerator. The blood samples were sent to the analytical laboratory immediately after they had been received and analysed within a few days.
- 6) Suitable analytical methods should be chosen and careful attention paid to the availability of laboratory service. This prerequisite will be discussed in detail in a following section, "Analytical procedures and control".

#### 5.1 Ethical and legal considerations

There are differences among various countries as to regulation costums and restrictions that must be considered in implementing human monitoring programmes. Above all it should be recalled that the Nurenberg code of 1947 and later the Helsinki declaration of 1964 as revised in the Tokyo declaration from 1975 preclude any experiment on a human subject without his "Voluntary Consent", and this principle has remained absolutely unchallenged. More recently the term, "informed consent" has been widely used. It is generally agreed that children, mental defectives and mentally deranged cannot give valid consent, although in certain circumstances their parents or other legal guardians may give consent on their behalf.

While the format of the "informed consent" to be used may vary from country to country according to the circumstances and the type of index media to be collected the following base elements should be considered:

- 1) procedure to be followed for taking specimens;
- description of any attendant discomfort and risk which might be expected;
- description of the purpose of the study, and of the benefits to be reasonably expected for the individual and the community;
- an assurance that the individual results will be kept confidential but that the individual (and/or his physician) will be informed, if he/she so wishes,

- about results with comments regarding their significance:
- in case of a study requiring repeated sampling, the person is free to withdraw his consent and to discontinue participation at any time without prejudice to the subject;
- as far as possible informed consent should be given in writing.

Informed consent is especially needed when a biological specimen is to be obtained with invasive techniques (e.g. blood sampling), whereas non-invasive sampling such as hair, nails and urine are usually free of legal and ethical complications (Berlin et al., 1979).

All scientific studies in Greenland need legal permission from the Ministry for Greenland after recommendation from the Commission for Scientific Research in Greenland. This study was recommended and financed by the Commission.

Concerning the ethical problems, in the first place the population was informed about the project through radio and newspapers. When contacted and prior to sampling each individual participant was informed according to the elements of "informed consent" mentioned above. The consent was not obtained in writing as the risk involved in this particular programme was regarded minimal.

Furthermore the project was described after the planning phase in the popular scientific journal "Forskning i Grønland" (Hansen, 1979), and equally after the project has been finished, the results have been summarized in the same journal (Hansen and Harvald, 1980).

#### 5.2 Project design for this study

The project was intended to take geographical as well as demographical differences into consideration. The first question to arise was how big a number of donors was necessary to obtain a valid estimate of the exposure.

The variance, in the population will determine the number of samples; as, however, this is not known a priori, the decision must be based on a guess. The total variance of data will consist of an analytical variance which is known and the population variance which consists of the following elements:

- a) variance related to geographical location (natural environmental background level and degree of pollution),
- b) individual variance (genetic status, eating and smoking habits, occupational exposure),
- c) time related variance (day to day, according to season).

The ideal monitoring programme will consist of a representative sample of the population to investigate. Representativity in any absolute sense is, however, an

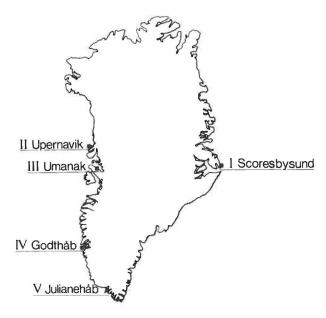


Fig. 1. Map of Greenland with sampling districts indicated.

utopia. The best approximation would be a group selected at random through the civil personnel register, a principle used in a monitoring programme in Denmark (Bach, 1979, 1980). Due to the specific traffical conditions in arctic areas, this principle is not practicable in Greenland, and it was therefore decided to use a cluster technique instead and select 5 districts with towns which could be reached easily and with a population large enough to provide the needed number of samples.

Population surveys will often involve from 0.001–0.01% of the total population; but the number should not be less than 100. The population of Greenland is 49.719 (1977), and it was decided to examine 150 individuals corresponding to 0.3% plus 25 Greenlanders living permanently in Denmark, who were to function as a control group.

As the primary purpose of the survey was to estimate the dietary exposure, it was decided to use a stratified sampling and sample from males in the age group 15–29 years. The rationale for this choice was as follows: children should be avoided because of ethical reasons. Women were excluded because of the possibility that pregnancies could introduce an uncontrollable parameter. The age group was chosen partly because men at this age have the highest relative food intake and as such form the group with the highest dietary exposure and partly because it is the age group best represented in Denmark in educational institutions and as such the best suited for the control group. An additional reason to choose a fixed age group was to avoid a possible age dependency in blood and hair concentrations.

#### 5.3 Choice of districts

In Greenland five districts were chosen for investigation, and from each it was intended to get 25 samples except from Godthåb (district IV) which is the greatest town with approx. 10.000 inhabitants, where 50 samples were planned as a greater heterogenity can be expected in this district. The geographical situation of the sampling districts is seen from fig. 1.

Upernavik (II) and Umanak (III) represent northern locations with traditional eating habits which means that marine mammals form a major part of the food. Furthermore, most of the analytical data on heavy metals in seals originate from animals caught in these districts. Godthåb (IV) represents modern Greenland where most imported food is eaten. Julianehåb (V) was chosen because of its southern location, and Scoresbysund (I) was chosen as an east coast district. The size of population in all towns was large enough to obtain a sample size of 25 men of the age of 15–29.

#### 5.4 Sampling

From each participant two blood samples were taken by venous puncture (Plica cubiti). At each sample of 5 ml one was stabilized by EDTA the other was taken in a plain vial to obtain serum. Hair samples were obtained from each person by cutting five subsamples as near to the scalp as possible. The subsamples were taken in the frontal, left and right temporal, and from the nuchal region.

An interview questionaire has been filled out containing information of present addresses, and if the participant had not lived at least two years at the present address, earlier addresses were recorded, civil personal register number (which contain data of birth and age), present occupation and previous one(s) if changes had taken place during the last two years. Housing conditions (living in town or village, house or flat), smoking habits, drinking habits, permanent intake of medicine, eating habits (mainly local food or mainly imported food or equal quantities of both and an indication of the number of meals of seal, whale and fish taken per week).

Prior to the sampling procedure, informed consent was obtained in each case according to the above described principles.

After sampling the hematocrit was determined at the local hospital laboratory (in district III Umanak hemoglobin was determined) to check for a state of anemia.

Immediately after sampling samples were sent by air to the laboratories for analysis, all samples were coded to allow identification but the origin hid to the laboratory staff.

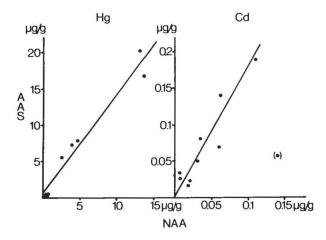


Fig. 2. Intercomparison between results on hair mercury and cadmium determined by two different methods.

AAS: atomic absorption spectrophotometry

NAA: neutronactivation analysis.

#### 5.5 Analytical procedure

#### 5.5.1. Blood

All blood samples were analysed at "Medicinsk Laboratorium" in Copenhagen. (This laboratory participates in the European Analytical Quality programme carried out under the auspices of the Commission of the European Countries).

The technique used was atomic absorption spectrometry. Hair analysis were carried out at the Institute of Hygiene, University of Aarhus.

#### 5.5.2. Procedure for hair analysis

From each sample 5 cm from the scalp-end was used. The sample was homogenized by being cut into pieces of approx. 2 mm. To eliminate possible external contamination the homogenized sample was washed according to the principle described by Petering et al. (1971). A wet-ashing was performed in a teflon decomposition bomb with conc. nitric acid. (p.a.), and the elements were determined by atomic absorption spectrometry (AAS). As an analytical control, 10 samples of hair (5 from Greenland and 5 from Denmark) were sent to EEC Joint Research Center, Ispra Italy for double analysis by a neutronactivation technique (NAA). The NAA technique is, however, not suited for lead analysis, and control has been performed only for mercury and cadmium. The results of this intercomparison programme are shown in fig. 2. The figure shows a very good correlation between the two techniques; but the AAS technique has a tendency to give higher values than does the NAA technique, which indicates the existence of a systematic error in one or both of the techniques. This will, however, not affect the validity of

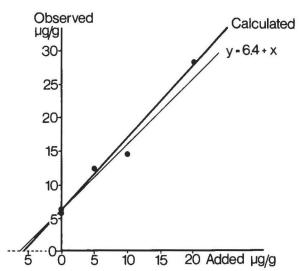


Fig. 3. Recovery of lead added to a hair sample.

the AAS technique used for relative comparisons between the groups in the study.

On six samples selenium was also determined by the two methods. All samples showed very uniform concentrations, and a very good agreement was found to exist between the methods: AAS 0.47  $\mu$ g/g  $\pm$  23.6%, NAA 0.42  $\mu$ g/g  $\pm$  19.7%.

Concerning analytical control for lead for comparison, 3 samples have been analysed at the Institute of Hygiene, University of Copenhagen, and a good agreement of results was obtained (sample 1: CPH 1.8  $\mu$ g/g AAR 1.4  $\mu$ g/g, sample 2: CPH 2.5  $\mu$ g/g AAR 3.3  $\mu$ g/g, sample 3: CPH 2.3  $\mu$ g/g AAR 2.1  $\mu$ g/g). The number of samples in this comparison is too small to be regarded as an intercalibration but shows, however, that the two laboratories reach results on the same level. It has not been possible to establish contact with a laboratory sufficiently experienced in hair-lead analysis to establish an intercalibration, and for this reason analytical control has been limited to internal control based on analysis in duplo and additions.

The analytical error based on 127 in duplo determinations was calculated to

$$s = 0.3781$$
  
( $\Sigma d^2 = 36.3290 \quad s^2 = \frac{\Sigma d^2}{2k} = 0.1430 \quad k = 127$ )

Additions of lead standards to a hair sample corresponding to 5, 10 and 20  $\mu$ g/g are seen in fig. 3.

#### 5.6 Practical experience of the sampling procedure

The sampling took place in the period September to October 1979. Unfortunately it was not possible to

Table 2. Number of samples from the districts.

	District	No. of samples wanted	No. of samples obtained
	Scoresbysund	25	26*)
II	Upernavik	25	14
III	Umanak	25	22
IV	Godthåb	50	50
V	Julianehåb	25	25
VI	Aarhus	25	7

<sup>\*)</sup> Hair samples only.

obtain the scheduled number of samples from all districts as seen from the table 2.

From district I (Scoresbysund) blood samples were taken in the wrong vials and thus had to be rejected. For this reason hair data only have been obtained from this district. The most difficult group to approach was the control group consisting of Greenlanders living in Denmark as only seven volunteered; as, however, the analytical data from this small group definitively differed from all groups from Greenland and furthermore were in agreement with results obtained in Denmark, in other population surveys (Bach, 1979, 1980), it was decided to accept the group as control and use other Danish data for comparison when available. The reduced number obtained in districts II and III was accepted as the analytical results showed a clear tendency, and because supplementary samples collected at a later time could introduce a seasonal variance.

#### 6. Results

The number of participants, their age and distribution between towns and villages are given in table 3.

All hematocrit and hemoglobin values were found to be within a normal range except 1 (sample no. II-1) which showed a hematocrit of 22.

As reported in the following, the data obtained from this survey have shown to fulfil neither requirements to a normal distribution nor a logarithmic distribution, but as it is often the case in biological monitoring programmes, data show a distribution skewed to the right. For

Table 3. Distribution of donors according to districts, age and residence in town or village.

	District	No. of samples	Age $\tilde{x} \pm S.D.$ Years		ence in village
I	Scoresbysund	26	21.8±4.5	25	1
11	Upernavik	14	$22.8 \pm 5.3$	11	3
III	Umanak	22	21.4±2.9	15	7
IV	Godthåb	50	$19.8 \pm 3.3$	50	
V	Julianehåb	25	$19.0 \pm 3.7$	4	21
VI	Aarhus	7	$21.3 \pm 2.4$	7	

this reason the numerical values have been used for estimates of general trends, while more detailed analyses have been made on the values transformed logarithmically. In the tables both arithmetic means and mean values of the logarithmically transformed data are indicated.

None of the participants in this programme could be regarded as occupationally exposed and in accordance with this no occupational differences for any of the elements under investigation have been noted.

#### 6.1. Mercury

#### 6.1.1. Wholeblood Concentrations

Wholeblood samples were analysed for total mercury (inorganic + methyl mercury). The frequency distribution district by district is shown in fig. 4, where the upper "normal" level and TLV (threshold limit value) are also indicated for inorganic and for organic (alkyl mercury) compounds (Valentin, 1979).

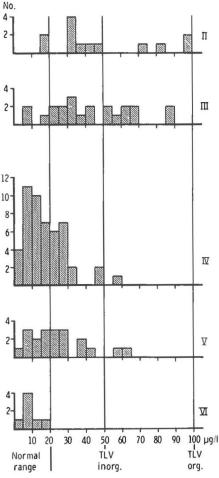


Fig. 4. Frequency distribution of mercury in wholeblood in districts II – VI, with indication of normal range and TLV (Threshold limit value) for inorganic and organic mercury.

It is to be seen that in the control group (VI) all values are within the normal range which was also the case in a Danish population survey, where the highest recorded value was  $17 \mu g/l$  (Bach, 1980).

In all districts in Greenland, values between the TLV for inorganic and organic compounds (Valentin, 1979) were recorded. The highest concentration was 95.8 µg/l found in district II (Upernavik). In fig. 5 the accumulated percentages are shown with indication of median values. The figure indicates a clear geographical difference with higher medians in the two northern districts than in the southern ones. The medians, arithmethic, and geometric means and standard deviations are given in table 4. The control group is significantly lower than other groups. The two northern (II and III) districts, which are traditionally hunting districts, are not different but as a group significantly higher than the groups from the two southern districts (IV and V) between which no difference is observed.

# % accumulated

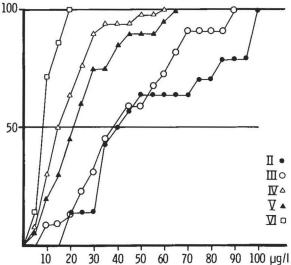


Fig. 5. Accumulated frequencies of wholeblood mercury concentrations in districts II-VI.

Table 4. Mercury in wholeblood, serum and hair according to sampling district. (Units: wholeblood and serum μg/l), hair μg/g).

	District	No.	C	Median -	Arith	nm. mean	Geomet	ric mean
	District	140.	Sample	Median	x	S.D.	х (log)	S.D. (log)
I	Scoresbysund	26	Hair	11.8	12.43	4.63	1.0479 (11.2)	0.1648
		13	Wholeblood	37.6	49.1	28.37	1.6237 (42.0)	0.2547
II	Upernavik	14	Serum	5.5	7.1	4.09	0.7795 (6.0)	0.2728
		14	Hair	9.3	12.4	8.86	1.0000 (10.0)	0.2913
		22	Wholeblood	40.0	43.5	23.05	1.5650 (36.8)	0.2821
III	Umanak	22	Serum	5.0	5.8	3.92	0.6744 (4.8)	0.2928
		22	Hair	11.5	12.8	8.95	0.9915 (9.8)	0.3444
		50	Wholeblood	15.0	17.4	11.99	1.1393 (13.8)	0.3140
IV	Godthåb	50	Serum	3.3	3.4	1.71	0.4778 (3.0)	0.2143
		50	Hair	6.4	8.0	6.19	0.7772 (6.0)	0.3554
		20	Wholeblood	22.5	24.6	15.88	1.2952 (19.7)	0.3168
V	Julianehåb	25	Serum	4.4	4.8	1.70	0.6433 (4.4)	0.1907
		25	Hair	6.8	7.2	3.43	0.8114 (6.5)	0.2451
		7	Wholeblood	7.6	8.5	4.51	0.8768 (7.5)	0.2265
VI	Aarhus	7	Serum	2.1	2.2	0.92	0.3105 (2.0)	0.1928
		7	Hair	1.8	2.0	1.35	0.2416 (1.7)	0.2521
	- 10	95	Wholeblood	1.5	1.9			
	Denmark*)	95	Hair	0.5	0.61			

<sup>\*:</sup> Data after Bach, 1980.

Figures in brackets antilog x.

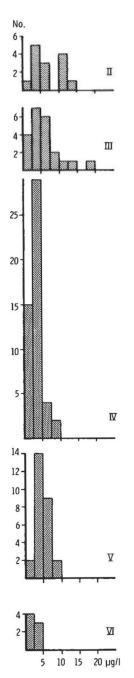


Fig. 6. Frequency distribution of serum mercury concentrations in districts II - VI.

#### 6.1.2. Serum

The values found in serum are represented in fig. 6 and 7. The same pattern as the one found for wholeblood data is observed. The values are relatively low and all below  $50 \mu g/l$  which could be regarded as a tentative toxic concentration of serum mercury (Hansen, 1976).

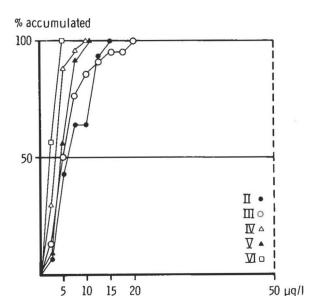


Fig. 7. Accumulated frequencies of serum mercury concentrations in districts II – VI.

In table 4 the control group is again shown to be the lowest in mean mercury and as for wholeblood the northern districts are the highest. The differences between districts are, however, not so pronounced as they are for wholeblood concentrations.

#### 6.1.3. Hair

The observed concentrations in hair are presented in table 4. and fig. 8a, b and 9a, b. No normal range is indicated here, as this can hardly be done at the moment due to insufficient data on hair as an index medium to evaluate exposure to mercury. It will be noticed that the same pattern as the one observed for wholeblood and serum is also seen in the hair data.

The hair mercury concentrations found in Scoresbysund (I) (conf. fig. 8b and 9b) are at the same level as in districts II and III. So, it is likely that blood values would also be on the same level as found in the other two districts.

#### 6.1.4. Mercury levels in relation to eating habits

The data have demonstrated a clear geographical difference between the northern (II + III) and southern districts (IV + V). The data have been divided into three groups in each district according to the amount of seal eaten per week.

- 1. Little or no seal: less than 3 meals per week.
- 2. Medium seal: 3-6 meals per week
- 3. Much seal: more than 6 meals per week

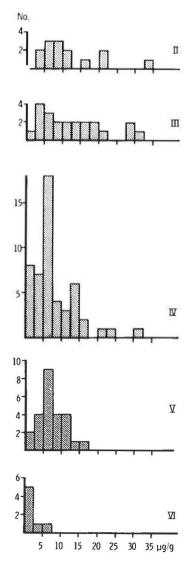


Fig. 8a. Frequency distribution of hair mercury concentrations in districts II – VI.

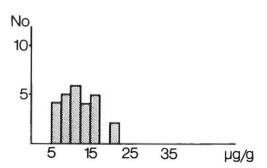


Fig. 8b. Frequency distribution of hair mercury concentrations in districts I.

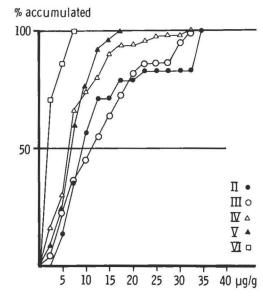


Fig. 9a. Accumulated frequencies of hair mercury concentrations in districts II-VI.

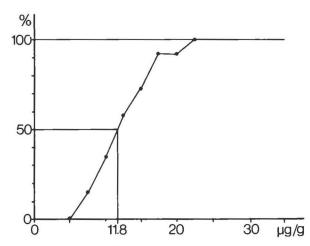


Fig. 9b. Accumulated frequencies of hair mercury concentrations in district I.

The logarithmic mean values and variances appear in table 5. Significant differences between districts are indicated in table 6. Fig. 10–12 gives the logarithmic mean values and their 95% confidence intervals, according to eating habits in the three parameters measured district by district. The figures clearly demonstrate the higher level in the northern districts, especially in wholeblood concentrations and that all three parameters show higher mercury concentrations in seal-eaters than in persons with no or low intake of seal.

Table 5. Mercury concentrations in blood, serum and hair, according to seal-eating habits.  $\bar{x}_{(log)}$  and variance indicated. Figures in brackets antilog  $\bar{x}$ .

			6 or more meals per wee	k			meals week			3 or less meals per w	
District	Sample	N	Mean x	variance v	N	Me x		variance v	N	Mean x	variance v
I	Blood Serum Hair	6	1.1902 (15.5)	0.0091	7	0.9861	(9.7)	0.0262	13	1.0171 (10.	4) 0.0241
	Blood	3	1.8073 (64.2)	0.0639	8	1.5805	(38.1)	0.0732	2	1.5210 (33.	2) 0.0014
11	Serum	3	0.9137 (8.2)	0.0733	9	0.7412	(5.5)	0.0937	2	0.7507 (5.	0.0024
	Hair	3	1.1952 (15.7)	0.1145	9	0.9359	(8.6)	0.0874	2	0.9820 (9.	6) 0.0162
	Blood	6	1.7729 (59.3)	0.0203	11	1.6069	(40.4)	0.0335	5	1.2233 (16.	7) 0.0930
III	Serum	6	0.9060 (8.1)	0.0386	11	0.6918	(4.9)	0.0556	5	0.3582 (2.	3) 0.0565
	Hair	6	1.1627 (14.5)	0.0971	11	0.9818	(9.6)	0.1266	5	0.8076 (6.	4) 0.0985
	Blood			-	3	1.3381	(21.8)	0.0297	47	1.1266 (13.	4) 0.1010
IV	Serum				3	0.5913	(3.9)	0.0990	47	0.4705 (3.	0.0476
	Hair				3	0.8115	(6.5)	0.0019	47	0.7745 (5.	0.1350
	Blood	9	1.4234 (26.5)	0.0657	3	1.4021	(25.2)	0.0075	8	1.1110 (12.	0.1305
V	Serum	13	0.6994 (5.0)	0.0164	4	0.7066		0.0062	8	0.5203 (3.	3) 0.0685
	Hair	13	0.9076 (8.1)	0.0263	4	0.9212	(8.3)	0.0080	8	0.5989 (4.	0.0810

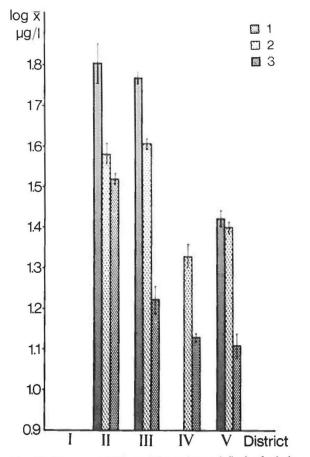


Fig. 10. Mean and 95% confidence interval (log) of whole-blood mercury concentrations in districts II – V, according to eating habits.

1: less than 3 meals of seal per week

2: 3-6 meals of seal per week

3: more than 6 meals of seal per week.

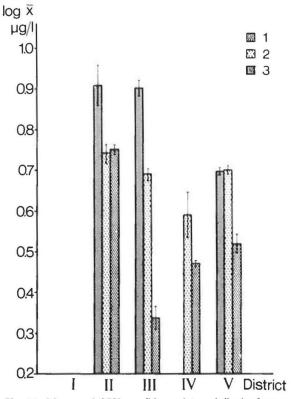


Fig. 11. Mean and 95% confidence interval (log) of serum mercury concentrations in districts  $\rm II-V$ , according to eating habits.

1: less than 3 meals of seal per week

2: 3-6 meals of seal per week

3: more than 6 meals of seal per week.

Table 6. Significance of differences in mercury concentrations in wholeblood, serum and hair between districts.

			SEI	RUM										
w		II	III	IV	V	VI			I	II	III	IV	V	VI
H O	II		-	+	+	+	H A	I						
L E	III	-		+	-	+	R	II	-					
В	IV	+	+		+	+		III	-	-				
L	V	+	+			+		IV	+	+	+			
O O D	VI	+	+	+	+			V	+	+	+	-		
ט								IV	+	+	+	+		

<sup>+ =</sup> significant different 5%

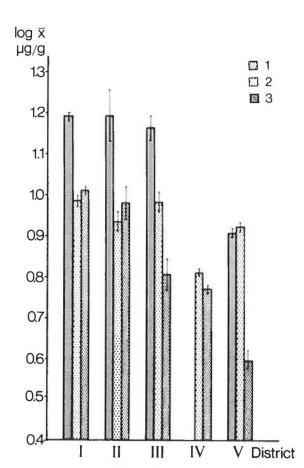


Fig. 12. Mean and 95% confidence interval (log) of hair mercury concentrations in districts I-V, according to eating habits.

- 1: less than 3 meals of seal per week
- 2: 3-6 meals of seal per week
- 3: more than 6 meals of seal per week.

#### 6.1.5. The relationship between the parameters

A good correlation was found between wholeblood and serum concentrations (r = + 0.8940 significant at 0.1%). The corresponding values are plotted in fig. 13, which also indicates the calculated regression line (wholeblood Hg =  $-2.55 + 6.68 \times$  serum Hg).

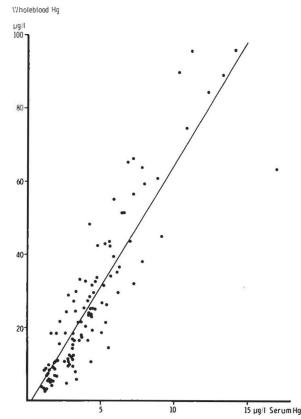


Fig. 13. Relationship between wholeblood and serum mercury concentrations from all districts.

<sup>- =</sup> not significant different.

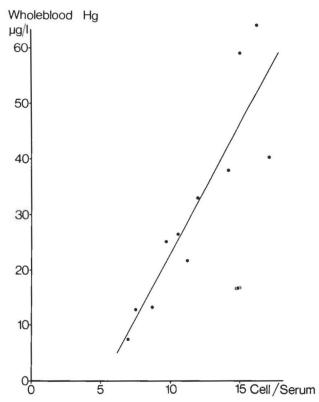


Fig. 14. Relationship between mean wholeblood mercury concentration and the calculated mean cell/serum ratios from each district (II - V), when data are grouped according to eating habits (conf. table 6).

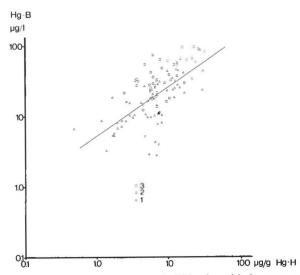


Fig. 15. Relationship between wholeblood- and hair mercury concentration from all districts (log scale).

1: less than 3 meals of seal per week

2: 3-6 meals of seal per week

3: more than 6 meals of seal per week.

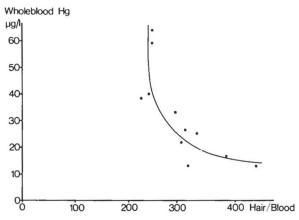


Fig. 16. Relationship between mean wholeblood mercury concentration and the mean hair/blood ratios from each district (II – V), when data are grouped according to eating habits (conf. table 6).

In fig. 14 the mean cell/serum ratios are plotted against mean wholeblood concentration for each group from table 6. A positive correlation is demonstrated. This means that mercury is accumulated more in cells than in plasma. Cell concentration (c) was calculated from the formula

$$c = \frac{[w - s (1 - h)]}{h}$$

where w is the wholeblood concentration, s the concentration in serum and h is the hematocrit. Exposure to methyl mercury is reported to give a cell/plasma ratio about 10 whereas exposure to inorganic mercury compounds gives a lower ratio around 1 (Gerstner and Huff, 1977). The ratios observed in this study, especially in the most heavily exposed groups, are thus atypically high.

Wholeblood and hair concentrations were also found to be positively correlated when the data were transformed logarithmically. A plot of the corresponding values, and of the calculated regression line, is given in fig. 15.

The ratios between hair and wholeblood concentrations given as means for the groups in table 6 are plotted in fig. 16. The figure indicates a negative non-linear correlation. At increasing blood values the ratio proceeds asymptomatically towards a value between 220 and 250, while the ratio increases at decreasing blood values.

#### 6.1.6. Discussion of mercury data

The close connection between mercury in index media and food of marine origin indicates that the major part of human mercury exposure in Greenland is to be found in the state of methyl mercury, which is supported by the finding of high cell/serum ratios (Gerstner and Huff, 1979).

Methyl mercury is an accumulative compound, and the body-burden revealed by continuous exposures is related to the amount taken in daily via food. A relation between body-burden in a steady state and daily ingested methyl mercury is given by Kitamura et al. (1976). This relationship is illustrated in fig. 17. Referring to the estimate of average intake of mercury in the population in Greenland given in table 1, the average daily intake should vary between approx. 0.1-2 mg/day. According to the model given by Kitamura et al. (1976) fig. 17, this would mean that Greenlanders (on average, and supposed that all mercury is in the methylated state) should be on a level of between maximum-no-effect and the toxic level (subclinical intoxication); and in the case of extremely high food intake in areas with a high contamination level the resulting body-burden should be expected to be on the toxic level.

Birke et al. (1972), have calculated the relationship between wholeblood concentration and daily methyl mercury intake to be described by the equation.

$$Hg-B \mu/l = -1.2 + 0.80 \times \mu g Hg/day$$

According to this, the most highly exposed group of seal-eaters in districts II and III (with a mean blood concentration of approx. 60  $\mu$ g/l) should have a mean daily intake of 75  $\mu$ g (0.075 mg) methyl mercury, and the highest recorded value 95.8 accordingly corresponds to a daily intake of approx. 125  $\mu$ g/day (0.125 mg). In a more recent publication Kershaw et al. (1980) found the relationship to be expressed by the equation

Hg-B 
$$\mu$$
g/ml = 0.9  $\mu$ g Hg/day.

The estimate of daily intake based on blood concentrations actually found would indicate that the estimates based on analysis of seal and fish are somewhat overrated. It must, however, be underlined that models describing the relationship between body-burden as well as wholeblood concentrations in relation to daily uptake are extremely difficult and should not be regarded as absolute values but instead as approximate values.

A fact is, however, that as a whole mercury levels found in Greenland are high compared to what is normally found in most parts of the world, and the concentrations found in wholeblood, serum, and hair are closely related to intake of marine mammals. That the level in the non-sealeating groups is also higher than that found in Denmark must be attributed to the fact that in general more fish is eaten in Greenland than in Denmark.

The geographical differences demonstrated show that the group in district V having 6 or more meals of seal

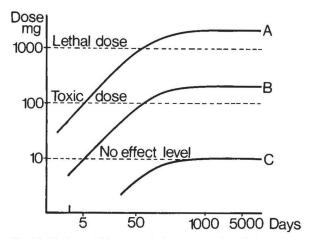


Fig. 17. Estimated bioaccumulation curves of methyl mercury in the human body (modified after Kitamura et al., 1976). A: daily intake 20 mg; B: daily intake 2 mg; C: daily intake 0.1 mg.

per week, had significantly lower concentrations in the three index media than the corresponding groups in districts II and III (and district I in hair). This indicates that seals in the south are probably less contaminated than seals in the northern districts. A factor which may play a role in this connection is, that a bigger quantity of seal is probably eaten per meal in the northern districts, where hunting is traditional, than in Julianehåb where eating habits are more influenced by imported food, especially among young people.

The finding of atypically high cell/serum ratios which increase by increasing wholeblood concentrations are not in agreement with the normally recorded ratio of around 10 in case of exposure to methyl mercury (Gerstner and Huff, 1977). In experiments with rats Magos and Webb (1977) have, however, shown that mercury distribution between the blood compounds is altered by simultaneous administration of selenium, so that cells take up more methyl mercury in relation to plasma when selenium is coadministered than when mercury is given alone. A similar reaction has not been demonstrated in humans, but together with mercury a concomitant high selenium intake from sea food could provide a ready explanation of the ratios found in this study.

The hair/blood ratios found in the most heavily exposed groups are in accordance with the values reported by Amin-Zaki et al. (1976) in their investigation in Iraq on persons intoxicated by methyl mercury. From Sweden Birke et al. (1972) report a ratio of 300 in persons exposed through fish comsumption. This value is also in accordance with the finding in this study due to the dose dependency of the ratio (fig. 16).

The results have indicated that at a constant exposure, blood, specially blood cell concentration, as well as hair mercury give useful indicators of human exposure

to mercury and that the cell/serum ratio can provide information on the influence of specific modifying factors.

The adverse effects of methyl mercury become detectable at blood concentrations of 200–600  $\mu g/l$  (Clarkson et al., 1975). Subclinical changes may, however, occur at a lower level, and the results of this study indicate that the most heavily exposed individuals in Greenland approach a critical level of exposure where subclinical symptoms of mercury intoxication can be anticipated in the most sensitive individuals. Special attention should be paid to the possibility of intra-uterine exposure.

Observations of the Minamata epidemic in Japan and more recent reports on experimental animals suggest that prenatal life is the most sensitive to methyl mercury intoxication (Khera, 1973). If this is the case, the calculations and models for dose-response relationships in adults have only a limited value in determining safe levels of methyl mercury for human beings, and future studies on dose-response should be directed towards the question of effects on early stages in the human life cycle.

An investigation from Alaska (Galster, 1976) on Eskimo mothers and their children exposed to mercury intake via seal-meat and -organs showed proportionality between intake and concentrations in blood and placenta. A correlation was also shown to exist between red cell concentrations in mothers and their newborn children. The relationship between these concentrations could be described as the equation

Hg (child) = 
$$1.8 \times$$
 Hg (mother) –  $4.5$ .

This means that the child obtains a higher cell concentration than the mother, which is true even if there is a correction for differences in hematocrit. If the highest blood values found in this study can be found also in pregnant women – using the above mentioned equation and supposing a hematocrit of mother of 40, and 50 in child, and supposing same cell/serum ratio – the assumption grows that as much as 200  $\mu$ g/l wholeblood can be found in newborn children in Greenland. This value is close to what can be expected to give clinical poisoning in adults. This points strongly to the absolute need for supplementary investigations to clarify possible adverse effects of the dietary mercury intake in the most heavily exposed population groups in Greenland.

#### 6.2. Cadmium

#### 6.2.1. Blood

In fig. 18 the frequency distribution of cadmium concentrations in blood is shown with indication of the highest value registered (by the same analytical laboratory) in non-occupationally exposed persons in Denmark and with indication of a supposed toxic level (Lauwerys et al., 1979). Only in Godthåb (IV) and

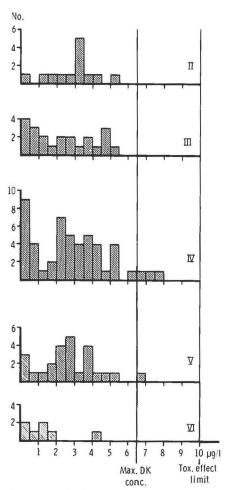


Fig. 18. Frequency distribution of blood cadmium concentrations in districts II – VI, with indication of highest concentration found in non occupationally exposed Danes and limit for toxic effect.

Julianehåb (V) blood concentrations are found exceeding those found in Denmark. The cumulated distribution of percentages is seen from fig. 19. The median value of the control group is lower than the medians in the groups from Greenland which are very close to each other.

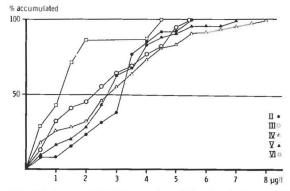


Fig. 19. Accumulated frequencies of blood cadmium concentrations in districts II – VI.

Table 7. Cadmium distribution in blood and hair according to geographical distribution. (Units: blood μg/l, hair μg/g).

	District	No. of samples	Sample	Median	Arithm. mean $\bar{x}$	Geometric mean $\bar{x}$ (log)	S.D.
I	Scoresbysund	26	Blood Hair	0.14	0.31	0.7679 (0.17)	0.4355
II	Upernavik	14	Blood Hair	3.15 0.24	2.82 0.56	0.3746 (2.37) 0.4984 (0.32)	0.3231 0.4834
III	Umanak	22	Blood Hair	2.50 0.36	2.30 0.51	0.1773 (1.50) 0.4354 (0.37)	0.4807 0.3592
IV	Godthåb	50	Blood Hair	2.75 0.26	2.80 0.44	0.2802 (1.91) 0.5831 (0.26)	0.4494 0.4456
v	Julianehåb	25	Blood Hair	2.70 0.36	2.59 0.47	0.2794 (1.90) 0.5453 (0.28)	0.4755 0.4297
VI	Aarhus	7	Blood Hair	1.15 0.30	1.34 0.65	0.0377 (0.92) 0.3989 (0.40)	0.4026 0.4701

Figures in brackets antilog x.

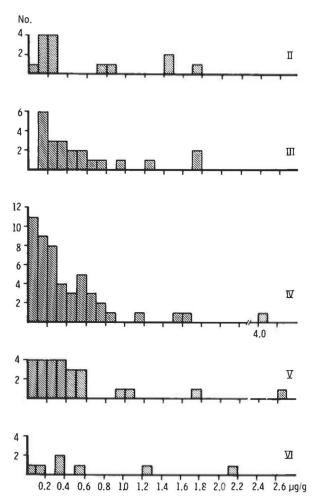


Fig. 20a. Frequency distribution of hair cadmium concentration in districts II – VI.

In table 7 the median arithmetic and geometric means and standard deviation are given for each district. No significant differences can be observed between the districts except for the fact that district II (Upernavik) is significant higher (5%) than district IV(Aarhus).

#### 6.2.2. Hair

The distribution of hair concentration is seen in fig. 20a and b. As for blood the highest individual values are found in the Godthåb and Julianehåb districts. The lowest mean value was found in Scoresbysund, but no significant differences could be demonstrated between the districts. No significant correlation was found between concentrations of blood cadmium and hair cadmium.

#### 6.2.3. Smoking

As cigarette smoking is regarded as the most important source of cadmium (Friberg et al., 1971) the data were separated into two groups according to smoking habits. Smokers are defined as persons who smoke more than 5

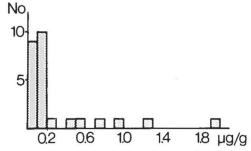


Fig. 20b. Frequency distribution of hair cadmium concentration in district I.

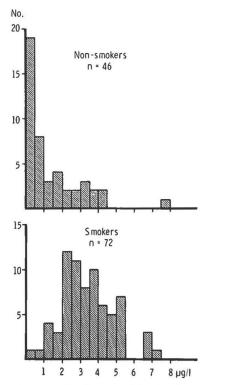
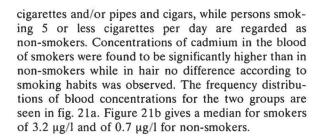


Fig. 21a. Frequency distribution of blood cadmium concentrations in smokers and non-smokers.



#### 6.2.4. Background blood concentration

The overall distribution of blood cadmium (fig. 22) is inhomogeneous. In more than 50% of non-smokers the concentration is found to be below 1  $\mu$ g/l. If this group is analysed separately as seen in fig. 23 a median value of 0.32  $\mu$ g/l is found, which must be regarded as a background level. This is in accordance with reports on "normal" blood values for non-smokers (Ulander and Axelson, 1974).

#### 6.2.5. Eating habits

As smoking is shown to influence cadmium concentration in blood, the data have been separated according to smoking and to seal-eating. Seal-eaters are defined as persons having more than 3 meals of seal per week and non-seal-eaters as having 3 or less. The mean values (calculated on logarithms) and 95% confidence inter-

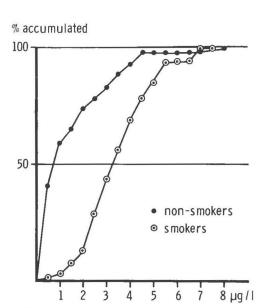


Fig. 21b. Accumulated frequencies of blood cadmium in smokers and non-smokers.

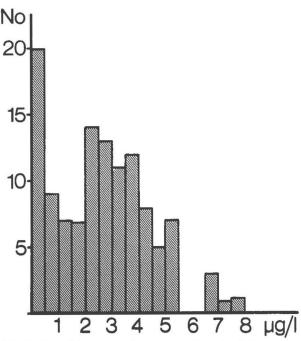
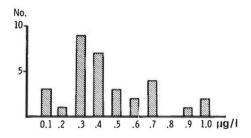


Fig. 22. Overall frequency distribution of blood cadmium concentrations.



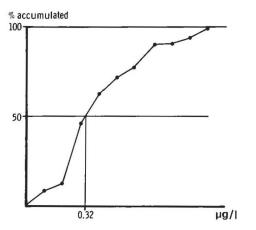


Fig. 23. Frequency distribution and accumulated frequencies of blood cadmium concentrations  $\leq 1 \mu g/l$ .

vals of both blood and hair are seen in fig. 24. The figure shows that smoking is reflected in blood, whereas in spite of the supposedly high dietary intake (conf. table 1) there is no difference according to seal-eating. Contrary to blood, hair seems to reflect dietary intake but not smoking.

The two index media seem to reflect exposure from different absorption sites. Blood reflects pulmonary uptake, while hair reflects dietary intake. This could be explained by a different metabolism of intestinally and pulmonarily absorbed cadmium.

Via the lymph- and portal-vein system intestinally absorbed cadmium is carried to the liver where it is bound to metallothionein (Piscator, 1964; Nordberg, 1971; Cempel and Webb, 1976); and from the liver the protein bound cadmium is partly transported to the kidneys, where it accumulates, and is partly excreted via the bile. The low reflection of dietary cadmium is in accordance with the fact, that only a small part of dietary cadmium is absorbed, namely about 5-6% (CEC, 1978) and is in accordance with the finding of fecal cadmium roughly equivalent to oral intake. Much more cadmium is taken up at pulmonary exposure, about 30% of the inhaled amount (CEC, 1978). Purely a priori, it can be predicted that body distribution of a foreign compound may depend on rate and route of administration.

If the idea of a different metabolism is valid, it rules

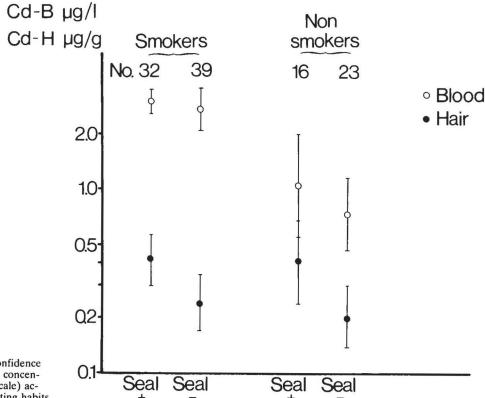


Fig. 24. Mean and 95% confidence intervals of blood and hair concentrations of cadmium (log scale) according to smoking and eating habits.

out dietary cadmium as an important factor in human exposure in Greenland in comparison to pulmonary exposure e.g. from cigarettes. This idea has, however, to be confirmed through an analysis of the kidneys which are the target organs and the only indication of bodyburden and risk. Not until kidney and index media concentrations have been compared will it be possible to draw definite conclusions of their use. This study has, however, shown that neither blood nor hair alone is sufficient to evaluate cadmium exposure, as they obviously reflect different rates of exposure. Analysis made on kidneys from Danish traffic casualties have shown that tobacco smoking has caused most of the cadmium present in kidneys (Østergaard, 1977). Future investigations will show if the same is the case in Greenland, or if the high dietary intake of cadmium, which is not reflected in index media, is anyhow accumulated in kidneys, and as such form a risk to the health.

#### 6.2.6. Discussion on cadmium data

The results of blood investigations have shown to be highly dependent on smoking habits and are found to be on a level comparable to what is indicated in literature as "normal" values (Ulander and Axelson, 1974) and all values are far below a critical level (Lauwerys et al., 1979). Hair concentrations are found a little below what is considered "normal" which is generally supposed to be  $0.24-2.7~\mu g/g$  (Iyengar et al., 1978). This together with the finding that hair reflects the dietary intake, supports the above given theory that dietary cadmium is a source of minor importance. Proof of this, however, cannot be given until an analysis of kidneys from seal-eating Greenlanders has been performed.

Cadmium accumulates in kidney cortex and reaches a maximum at the age of approx. 50. A critical concentration is suggested by Friberg and Piscator (1977) to be around 200 p.p.m. at which level proteinuria occurs.

This critical kidney concentration is calculated by Kjellström and Nordberg (1978) to be reached at the age of 45 with a daily intake of 300-450 µg cadmium per day. Compared to table 1, this limit should be exceeded by a considerable part of the seal-eaters in Greenland. Supposing an intestinal absorption of 5%, a daily absorption of 6.8-87.3 µg cadmium can be expected (conf. table 1). According to a dose-response model by Kjellström (1977b), a daily absorption of approx. 7 µg will produce proteinuria in 10% of 50 year old people and 20 µg/day in 50%, and an absorption of 87µg daily will give clear signs of clinical intoxication. As these conditions are clearly contradicted by the findings of "normal" concentrations in hair and blood, which are not in general significantly different from what is found in other areas, the figures in table 1 must either be overestimated or cadmium is present in a binding which is not readily absorbed from the intestines.

Analysis of cadmium in kidney cortex performed in Denmark on casualties have shown a mean value of approx. 40 mg/kg (Østergaard and Clausen, 1974; Østergaard, 1977). At this level dose-response studies have indicated that due to individual variations and variations in exposure, between 0.1 and 2.5% of the total population at an age of 50 have passed the critical level.

Exposure through cigarette smoking is supposed to be the same in Greenland as it is in Denmark, so a similar response rate can be expected. The contribution of dietary cadmium to kidney level and because of this to risk of chronic intoxication is still unknown. Supplementary investigations are therefore urgently needed, before a final assessment of risk from cadmium exposure is possible.

#### 6.3. Lead

#### 6.3.1. Blood

The frequency distribution of blood lead concentrations is shown district by district in fig. 25.

The figure also indicates that no single value exceeds what was recorded as the highest value in a Danish population survey (Bach, 1979), and that no value exceeds the individual upper limit for blood lead in the general, nonoccupationally exposed population of 35  $\mu$ g% given by the EEC directive of 1977 (EEC, 1979).

In figure 26 are given the accumulated frequencies, and median values are indicated as well. It is seen that the northern districts (Upernavik II and Umanak III) have higher medians than the southern ones (Godthåb IV and Julianehåb V). The control group has median value lower than all Greenland districts.

Tables 8–9 show that there are no significant differences between districts II and III and between IV and V, but significantly higher values are found in II and III as a group compared to IV and V (conf. fig. 28). The southern groups are found to be on the same level as in a comparable group (age and sex) in the Danish population survey (Bach, 1979). There seems to be a geographical difference in blood levels in Greenland. This finding will be discussed later.

#### 6.3.2. Hair

Fig. 27a and b show the distribution of hair concentrations with indication of the upper "normal" limit (20  $\mu g/g$ ) (Grandjean, 1978). The figures show that the major part of the results are below 20  $\mu g/g$ , only in Umanak (III) and Godthåb (IV) some values are found between 20 and 70  $\mu g/g$ . The median values are found very similar (table 8), and thus the hair medians do not follow the blood medians. Table 8 also shows that the mean hair value does not differ to the same degree as blood mean values do. The geographical difference is only found to be reflected in blood, whereas hair values from north and south do not differ (fig. 28).

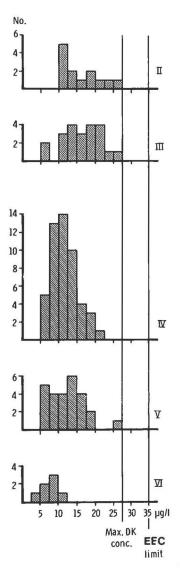


Fig. 25. Frequency distribution of blood lead concentrations in district II - VI, with indication of highest concentration recorded in Denmark and the upper individual confidence EEC limit.

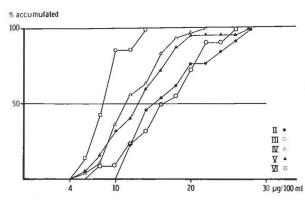


Fig. 26. Accumulated frequencies of blood lead concentrations in districts II-VI.

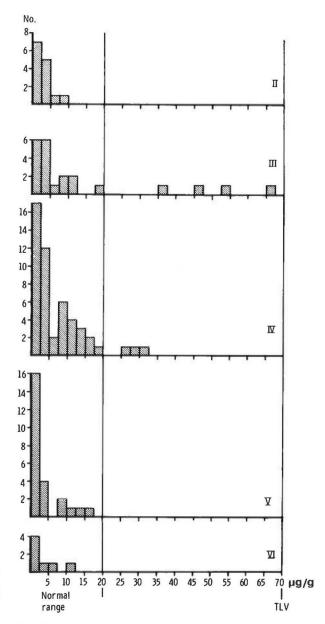


Fig. 27a. Frequency distribution of hair lead concentrations in districts  $\Pi-VI$ , with indication of normal range.

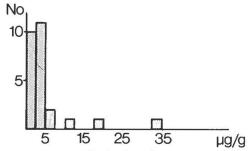


Fig. 27b. Frequency distribution of hair lead concentrations in district I.

Table 8. Lead concentrations in blood and hair according to geographical distribution. (Units: blood µg/100 ml, hair µg/g).

	District	No. of samples	Sample	Median	Arithm. mean $\bar{x}$	Geometric mean $\tilde{x}$ (log)	S.D.
I	Scoresbysund	26	Blood Hair	3.3	5.0	0.5157 (3.28)	0.3581
II	Upernavik	14	Blood Hair	15.2 3.0	16.1 3.0	1.1829 (15.24) 0.4154 (2.61)	0.1464 0.2322
III	Umanak	22	Blood Hair	16.0 4.5	16.0 13.6	1.1795 (1512) 0.7756 (5.96)	0.1567 0.5578
IV	Godthåb	50	Blood Hair	11.4 4.0	11.4 7.0	1.0363 (10.87) 0.6436 (4.40)	0.1398 0.4335
V	Julianehåb	25	Blood Hair	12.9 2.5	12.2 4.0	1.0631 (11.56) 0.4394 (2.75)	0.1775 0.3587
VI	Aarhus	7	Blood Hair	8.4 3.0	7.9 3.9	0.8737 (7.48) 0.4563 (2.86)	0.1526 0.3641
	Denmark*)	33	Blood Hair	10.9	11.6	1.04 (11.00)	0.1660

<sup>\*)</sup> Data after Bach, 1979. Figures in brackets antilog x.

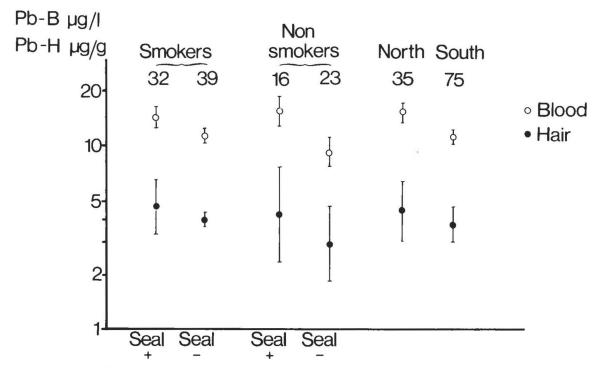
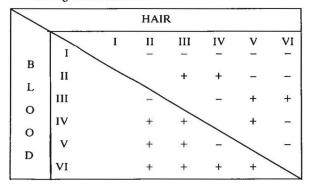


Fig. 28. Mean and 95% confidence intervals of blood and hair concentrations of lead according to eating and smoking habits and comparison between the northern (I and III) and southern districts (IV and V).

Table 9. Significance of difference between districts.

- + = significant different 5%
- = not significant different.



A weak, but significant correlation was found between blood and hair lead r = 0.3076 (significant on the 0.5% level).

#### 6.3.3. Smoking and eating habits

In fig. 28 are given the means (calculated on logarithms) and 95% confidence interval for blood and hair according to information on eating and smoking.

In the blood a significant difference was found between seal-eaters (more than 3 meals per week) and non-seal-eaters. This was true for both smokers and non-smokers, and between smoking and non-smoking non-seal-eaters (level of significance 5%), whereas no difference was recorded between smoking and non-smoking seal-eaters. The same tendencies are seen in hair, but the variances in hair concentrations are so large that the differences are non-significant. The results indicate that both intake of lead via food and via smoking is reflected in bloodlead concentrations, and that blood is a better index medium for evaluating lead exposure than hair.

According to table 10 the group from the northern districts has more seal-eaters (80%) than the southern group (27%), whereas smokers and non-smokers are equally distributed between the groups. When the results are corrected for this difference in group composi-

tion, the mean values for the northern and southern districts are the same, so there are no real geographical differences but only differences in eating habits.

Concerning the high hair lead value found in the Umanak district where 4 out of 22 have concentrations above 20  $\mu$ g/g, no explanation has been formed. Three members of the group have been found in villages near the leadmine in Marmorilik (conf. fig. 29) but none of the donors showing high hair concentration have been working in the mine.

Contamination from the mine is not likely to occur. It has therefore been considered whether the high values in this area could be related to geological characteristics of the area.

Godthåb and Julianehåb are situated in areas dominated by crystalline gneiss. The mineral content of this gneiss varies but can roughly be compared to granodiorite, which on a global scale contains on average 15 p.p.m. lead (Taylor, 1968a). Gneiss from the westcoast of Greenland has shown 14 p.p.m. (mean value of 6 analysis) (Baadsgaard et al., 1976).

Umanak is situated in a metasedimentary area consisting of limestone and slate. On a global scale, slate contains an average of 20 p.p.m. of lead, somewhat higher than gneiss. In limestone from Uvkusigssat and Mārmorilik in the Umanak district lead values as high as 2470 p.p.m. are found (Frank Pedersen, Institute of Geology, University of Aarhus, person. comm.).

There are also big areas in the Umanak district with lava rocks. The composition of these corresponds to that of basalts. In corresponding rocks in Iceland an average lead concentration of 23 p.p.m. has been found (Kraushopf, 1976). Upernavik is situated close to a granite intrusion. No analysis on this specific granite exist, but corresponding types of granite from other parts of the world have been reported to contain 19 p.p.m. (Turekian and Wedepohl, 1961) and 30 p.p.m. of lead (Taylor, 1968a and 1968b). It is therefore reasonable to expect the granite in Upernavik to contain more lead than the gneiss in Southwest Greenland.

The information on naturally occurring lead concentrations could support the idea that there is a direct relationship between concentrations between man and his geological environment. The analytical data permit, however, no further elucidation of the question which

Table 10. Distribution of seal-eaters (more than 3 meals per week), and smokers (more than 5 cigarettes per day) between the northern and southern districts.

	Total	Seal +		Seal -		Smokers		Non smokers	
	No.	No.	%	No.	%	No.	%	No.	%
Northern districts (II + III)	36	29	80.6	7	19.4	20	55.6	16	44.4
Southern districts (IV + V)	75	20	26.7	55	73.3	52	69.3	23	30.7

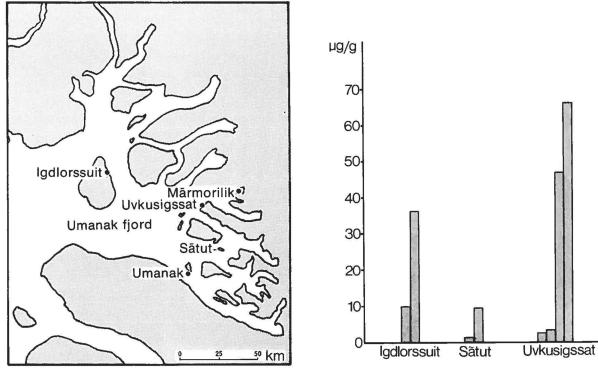


Fig. 29. Map of the Umanak district (III) with indication of villages where samples are taken in relation to the leadmine in Mārmorilik. Columns indicate individual hair lead values recorded in samples from the villages.

for the moment is purely theoretical, but as it will be of importance in assessing biological standards for human environmental exposure it might be worthwhile to investigate.

#### 6.3.4. Discussion of lead data

The lead values found in blood which are normally accepted as the best parameter of human exposure were below the limits fixed in the EEC directive of 35  $\mu$ g% for individuals, fifty per cent of the population examined should be below 20  $\mu$ g%, and as such there is no reason for concern. The few high hair lead values are, however, an indication that periodical peak exposures may take place in certain areas. The finding of equal or in seal-eaters of higher mean blood concentrations than in Denmark where the population in contrast to what is true in Greenland is under the influence of heavy traffic and industries, points to a future need to follow trends in blood lead especially in children, who form a risk group due to their sensibility to lead exposure, which is higher than that of adults (Lin-Fu, 1975).

#### 6.4. Selenium

Analysis of selenium was not included in the original objectives where it would have been highly relevant as selenium is in interaction with mercury (Parizek and Ostadalova, 1967; Ganther et al., 1972), cadmium (Kar et al., 1960) and possibly also with lead (Cerklewski and Forbes, 1976; Rastogi et al., 1976). The reason was that no Danish laboratory was able to trace selenium in the low concentrations normally found in blood and hair. After termination of the analytical programme, the Institute of Hygiene, University of Aarhus, has got analytical equipment which makes selenium analysis possible. At this time all blood samples were destructed and only hair samples remained. It was then decided to analyse 5 samples chosen at random from each district and 5 Danish hair samples for comparison. Ten of the samples were divided into two, and the one subsample sent to the EEC joint Research Laboratory at Ispra for analytical control as reported in section 5.5.2.

The results of the hair analysis from each district are shown in fig. 30. No differences between districts and between Greenland and Denmark were observed; neither between seal-eaters nor non-seal-eaters. The values found are in agreement with reported normal values of  $0.3 - 0.6 \mu g/g$  (Valentine et al., 1978; Schroeder et al., 1970). Valentine et al. (1978) report hair selenium to be significantly positively correlated to intake via drinking water and suggest that hair be used as a reliable parameter of dietary selenium intake. The results of hair analysis do not support the idea that the supposed high selenium intake in Greenland due to the high consuming of marine food leads to a higher ab-

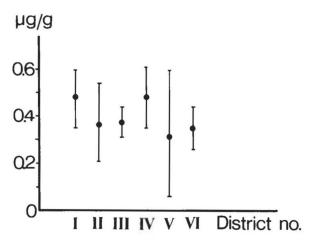


Fig. 30. Mean and 1 S.D. of hair selenium concentrations from districts I – VI.

sorption of selenium than in Denmark, which is regarded as a low selenium area (Gissel-Nielsen, 1977).

In livers from marine mammals Koeman et al. (1973, 1975) have shown selenium to occur together with mercury in a molar ratio of 1:1, obviously bound together in a biologically non-active complex which is not readily absorbed from the intestines (Kristensen and Hansen, to be published). In mammals the liver is the organ with the highest selenium retention. In experiments with minks fed with pike containing 6 mg Hg as methyl mercury and 0.2 mg selenium/kg Jernelöv et al. (1976) found a considerable accumulation of mercury in muscle up to 100 days, which was not followed by selenium as was the case in liver and kidney.

This means that mercury exists in muscles in a non-selenium bound state. A similar pattern is observed in mice exposed to inorganic mercury and selenium (Kristensen and Hansen, to be published). There is no information available on the distribution of the two elements in seal, but it will most likely be identical with that of minks and mice. According to this, seal muscle tissue will be the main source of mercury to humans. The main source of selenium in the diet is fish, where selenium generally occurs in high concentrations compared to mercury, mean molar ratio Hg/Se 1:16 (Koeman et al., 1975).

In experiments with mice injected with methyl mercury and selenium in various molar ratios Satoh and Suzuki (1979) found that simultaneously administered selenium increased the blood concentration of mercury which, however, was only due to increase of the concentration in red blood cells. The reaction was dose-dependent in such a way that the more selenium was dosed in relation to mercury, the higher the concentration in the cells became. A high exposure of biologically active selenium will thus raise the cell to plasma ratio.

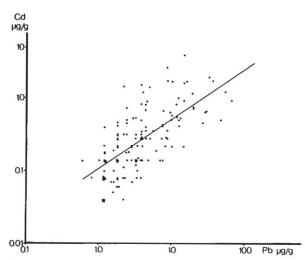


Fig. 31. Relationship between hair cadmium and -lead (log) scale).

The use of hair as index medium for evaluating selenium status needs further investigations, and final conclusions cannot be drawn upon the few results reported here.

Thus the observation of very high cell to plasma ratios in the most heavily exposed groups could be an indication of a high absorption of active selenium, which could mean that a protection against adverse health effects of methyl mercury could be present. The aleviating effect of selenium on mercury toxicity is thought to be due to a binding to stable complexes, which are stored in liver, kidney and spleen. Therefore when concomitant with high mercury intake, a high selenium intake is not necessarily reflected in hair. It must, however, be strongly underlined that this explanation is purely theoretical and needs further investigation, as selenium is in interaction not only with mercury but also with cadmium and probably lead.

#### 6.5. Relationships between elements

No relationship between mercury and selenium could be found in hair. In both marine mammals (Koeman et al., 1975) and in humans exposed to mercury (Kosta et al., 1975) mercury and selenium have been reported to be found in a molar ratio 1:1 in various organs. This shows that hair does not accumulate these elements in the same proportions as do other organs; this is in agreement with animal experiments (Kristensen and Hansen, to be published) and shows the need to investigate the use of hair for evaluating selenium status. Mercury in blood as well as in hair was not found to correlate with cadmium and lead in blood or hair.

Cadmium and lead in blood were found to vary independently, whereas cadmium and lead in hair were correlated as shown in fig. 31. The correlation coeffi-

Table 11. Relationship between lead and cadmium in hair according to smoking and eating habits.

	No. of samples	Regression equation	Coefficient of regression r <sup>2</sup>	Mean lead $\bar{x} \log \pm S.D.$	Mean cadmium $\bar{y} \log \pm S.D.$
Smokers + seal	19	log Cd = -0.7469 + 0.6896 log Pb	0.3167	0.5996±0.3396	-0.3334±0.4168
Non smokers + seal	8	$\log Cd = -0.7574 + 0.5021 \log Pb$	0.4120	0.8291±0.5695	$-0.3411 \pm 0.4454$
Smokers  – seal	50	$\log Cd = -1.1098 + 0.7979 \log Pb$	0.5277	0.5559±0.4083	$-0.6663 \pm 0.4485$
Non smokers  – seal	25	$\log Cd = -1.0082 + 0.5408 \log Pb$	0.4821	0.4854±0.4857	-0.7457±0.3783

cient is + 0.6770 which is significant on the 0.1% level. Correlation in hair between cadmium and lead have earlier been reported by Petering et al. (1973).

When the paired data are grouped according to smoking and eating habits a significant correlation is observed in each group, as seen in table 11 and fig. 32. The figure shows, as mentioned earlier, that dietary but not inhaled cadmium is reflected in hair concentrations, whereas there are no differences in lead concentrations. It should be noticed that the slope of the regression lines are different, depending on smoking or not smoking, while the intercept (the cadmium level) is determined by the seal-eating. This points to the existence of an interaction, direct or indirect, between the elements and furthermore to suggesting that the elements are metabolized differently according to the route of absorption leading to different reaction patterns.

Direct interactions between lead and cadmium have been indicated by Exon et al. (1979) and Thawly (1977), while both metals are known to interact with a number of micronutrients such as iron, copper,

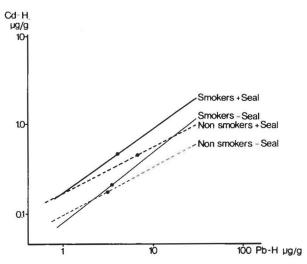


Fig. 32. Regressionlines with indication of mean values for the relationship between hair cadmium and -lead in groups separated according to eating and smoking habits.

selenium, zinc, calcium, vitamin E, vitamin C and vitamin D. Thus there are several possibilities of indirect interactions through one or more of these compounds. Studies of interaction are causing increasing interest, as an understanding of these reactions is of importance both to the understanding of metal toxicity and to assessing optimal quantities of essential micronutrients.

## 7. Concluding remarks

Most biological surveys have an unfortunate tendency to leave more questions unanswered than those included in the objectives, and this survey is no exception.

The primary question was to evaluate human exposure to mercury, cadmium and lead in relation to dietary intake and to evaluate the exposure level found in relation to health risks.

Mercury was found to be strongly related to intake of marine mammals, and in the part of the population where these are not eaten, the mercury level found in hair and blood corresponds with what is found in other populations with a relatively high consumption of fish.

As to health risks, the results have indicated that in the districts where traditional local food is eaten, especially in the northern districts, the exposure level reaches a limit where subtoxic effects may be anticipated.

Regarding cadmium, it is concluded that in spite of a supposedly high dietary intake, only a small reflection of this is found in hair and blood, whereas smoking habits in Greenland as in other parts of the world seem to be the major cadmium source. Cadmium exposure is to be regarded more as a "civilization factor" than as an environmental factor. The influence of dietary cadmium on kidney accumulation still has to be investigated.

As it was to be expected from food analysis lead was found to be within normal ranges but surprisingly, the levels are comparable with the level found in an industrialized society, e.g. Denmark; this in spite of the loca-

tion of Greenland far from industries and the very modest traffic intensity in the towns of Greenland.

The questions which have appeared during the survey concern mercury: do the high blood concentrations in the northern districts lead to placental transfer, or are toxicity modifying factors present (e.g. selenium) to prevent fetal exposure? Is the high exposure through seal-eating as it is observed in several arctic areas a result of a global mercury pollution, or is it a result of a natural accumulation of background mercury through the food chain?

Regarding cadmium, the surprising finding of a very little influence of dietary intake of hair and blood concentrations raises the question if cadmium present in natural bindings in seal is absorbed in a proportion less than 5% normally considered as typical through the gastroentestinal tract. An analysis on human kidneys is, however, necessary to decide whether or not dietary cadmium from marine mammals form any health risk.

The finding that blood lead levels in Greenland are on the same level as in West European industrialized countries raises the question whether this is caused by natural geological lead concentrations or whether there are unknown sources of pollution? This should be checked through investigations of blood lead especially in children.

The correlation between lead and cadmium in hair, but not in blood, raises the question if there is a direct interaction between these two elements, or if the interaction is an indication of one or more metabolic functions.

The above mentioned questions are still unanswered, and for sure, together with them, many others which have not revealed themselves yet, are of great importance, as a relevant interpretation of biological monitoring depends heavily on their solution. In this connection it must be stressed that the heavy metals in question are in interaction with many essential micronutrients as calcium, copper, iron, vitamin D, vitamin E, selenium, ascorbic acid, etc. The traditional diet in Greenland is deficient of some of these e.g. calcium, vitamin E and ascorbic acid (P. Helms Institute of Hygiene, Aarhus, pers com.). Especially ascorbic acid is a problem in the traditional diet (Uhl, 1955), and due to this the population may be vulnerable to an environmental pollution. Continuous investigations in Greenland of these environmental problems are of importance for several reasons:

- They will allow determination of trends in exposure level. Increasing median values form a more serious observation than the finding of few individuals to pass a biological standard.
- Elucidation of interactions between toxic and essential elements and the interference of toxic metal in metabolic processes will provide a better understanding of nutritional problems and risks from environmental pollutants.

The population groups in Greenland are ideal for studying invironmental exposure to heavy metals, because firstly, the population is stationary and only minimally influenced from outside, and secondly the sources of environmental exposure are only few compared to the industrialized societies. This simple environmental situation provides a good possibility to perform exposure/dose-response studies, which are otherwise extremely difficult.

Continued studies in Greenland are therefore very important from an international scientific point of view, but first of all it is important to be able to take appropriate measurements to protect the population in Greenland from adverse health effects from the environment. Awareness of potential health effects from environmental factors as the heavy metals is especially important in arctic areas because of the isolation of populations with a high degree of self-sufficiency in food supply, which can lead to a suboptimal intake of certain essential micronutrients and due to interactions may have pronounced effects on metal toxicity.

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# 8. Addendum

#### Individual Results

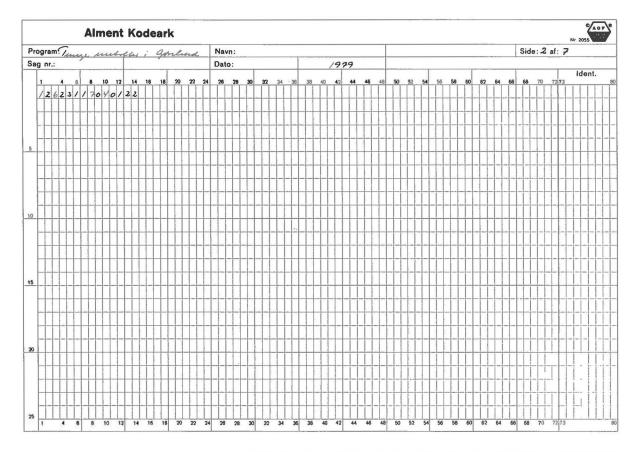
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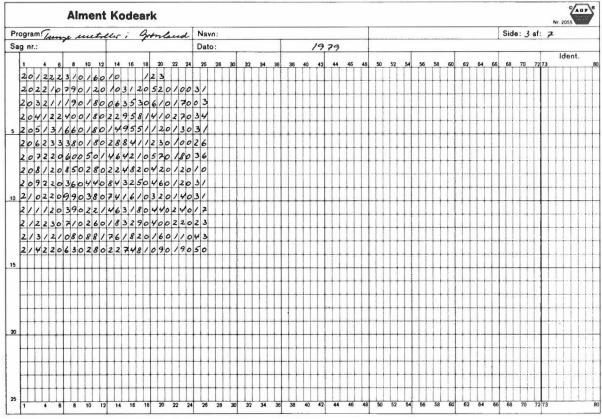
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- 2.3. Personal code no. serial no.
  - 4. Smokers 2, non smokers 1
  - 5. More than 6 meals of seal per week 3
    3-6 meals of seal per week 2
    less than 3 meal of seal per week 1
- 6.7.8. Mercury concentration in hair  $\mu g/g \times 10$
- 9.10.11. Lead in hair  $\mu$ g/g x 10
- 12.13.14. Cadmium in hair  $\mu g/g \times 100$
- 15.16.17. Mercury in blood  $\mu$ g/1 x 10
- 18.19.20. Mercury in serum μg/1 x 10 21.22.23. Lead in blood μg/100 ml
- 21.22.23. Lead in blood μg/100 ml 24.25.26. Cadmium in blood μg/1 x10

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Alterations against the ms. will be charged to the author(s). Twenty five offprints are supplied free. Order form, quoting price, for additional copies accompanies 2nd proof. Manuscripts (including illustrations) are not returned to the author(s) after printing unless especially requested.

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All Greenland place names in text and illustrations must be those authorized. Therefore sketch-maps with all the required names should be forwarded to the Secretary for checking before the ms. is submitted.

Language. – Manuscripts should be in English (preferred language), French, or German. When appropriate, the language of the ms. must be revised before submission.

Title. – Titles should be kept as short as possible and with emphasis on words useful for indexing and information retrieval.

Abstract. – An English abstract should accompany the ms. It should be short, outline main features, and stress novel information and conclusions.

Typescript. – Page 1 should contain: (1) title, (2) name(s) of author(s), (3) abstract, and (4) author's full postal address(es). Large mss. should be accompanied by a Table of Contents, typed on separate sheet(s). The text should start on p. 2. Consult a recent issue of the series for general lay-out.

Double space throughout and leave a 4 cm left margin. Footnotes should be avoided. Desired position of illustrations and tables should be indicated with pencil in left margin.

Underlining should only be used in generic and species names. The use of italics in other connections is indicated by wavy line in pencil under appropriate words. The editor undertakes all other type selection.

Use three or fewer grades of headings, but do not underline. Avoid long headings.

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Book: Marsden, W. 1964. The lemming year. – Chatto & Windus, London: xxx pp.

Chapter (part): Wolfe, J. A. & Hopkins, D. M. 1967. Climatic changes recorded by Tertiary landfloras in northwestern North America. – In: Hatai, K. (ed.), Tertiary correlations and climatic changes in the Pacific. – 11th Pacific Sci. Congr. Tokyo 1966, Symp.: 67–76.

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