Chapter 7

Heavy Metals

Contents

7.0. Introduction

Heavy metals occur naturally in all ecosystems, but with large variations in concentration. They are also released to the environment from many different anthropogenic sources. This chapter identifies the metals that are relevant in a pollution context and considers the chemical forms in which they are found. It describes natural and man-made sources, pathways, and levels of heavy metals in the atmosphere and in the marine, freshwater, and terrestrial environments. Toxicological characteristics of heavy metals, e.g., uptake, accumulation, and effects in organisms, are also considered.

A focus of this chapter is the description of concentrations of heavy metals found in the Arctic in terrestrial, freshwater, and marine ecosystems, because these data provide the basis for assessing geographical differences and temporal trends in levels of heavy metals. Although a substantial amount of heavy metal data has been compiled, detailed conclusions are difficult to make because 1) the area covered by the AMAP assessment is very large; 2) few areas have been monitored on a regular basis (the best covered areas have data for less than five sampling years, and temporal trend sampling seldom covers up to two decades); 3) geographical coverage of available sampling data is poor; and 4) analytical results are often not completely comparable due to sampling, analytical, and reporting differences.

These limitations can be severe. A number of publications have addressed the problem of what constitutes adequate biological sampling in connection with ecotoxicological investigations (Bignert *et al.* 1993, 1994, Olsson 1995). These works stress that substantial individual variation occurs in biological systems and that many samples have to be analyzed before values are available which reliably describe bioaccumulation, seasonal variation, or spatial and temporal trends. According to Olsson (1995), from 11 to >20 years of annual sampling is needed to discover a 5% annual change in the concentrations of Cd, Pb, and Hg in the muscle and liver of Swedish reindeer.

There are also problems linked to analytical quality. Over the last 25 years a number of different techniques (including differences in sample preservation and preparation, analysis, and equipment) have been used to measure heavy metal concentrations. The available data are thus not always of uniform quality. In a review of heavy metals in the Greenland marine environment (Dietz *et al.* 1996), it was evident that some of the older data were incorrect (too high) because techniques had not been sufficiently tested at that time, particularly for Pb and Cd. It should be stressed that this chapter reports the data as they were published in the literature, without adjusting for the different analytical techniques, except in cases where raw data or analytical information was available to the authors (some data which were clearly erroneous were omitted). It is obvious that considerable attention must be directed at intercalibration of laboratories in the future if coherent data sets for circumpolar heavy metal concentrations are to be obtained.

Another factor that makes it difficult to compare existing heavy metal data is the way in which data are reported, i.e., the statistical parameters reported, and the level of detail available in the reported data and its supporting information. For example, much of the available data consists of arithmetic means, geometric means, or median values; statistical parameters which can differ considerably when calculated for any given set of data. This complicates comparison of data from different sources. For biota, the species and tissues analyzed, age/size, sex, and year of sampling also differ among the available data and make comparisons complex.

The ideal situation when comparing data for such a wide area and from diverse sources would be to have access to all raw data (individual sample concentration values) and any necessary supporting information (sample characterization, biological information, full methodological descriptions). This was not possible for much of the data used in preparing this chapter, e.g., much of the data available in the published literature. For all the new AMAP data, however, appropriate reporting procedures have been implemented. Notwithstanding these limitations, a substantial understanding of the status of metals in the Arctic can be reached using the results of already-published investigations.

Methods used in the preparation of this chapter for the selection, handling, and assessment of data are described in the appropriate subsections.

7.1. Physical/chemical characteristics

7.1.1. Identification of metals to be considered

Heavy metals can create adverse effects on environmental and human health due to their toxicity and their bioaccumulation in various environmental compartments. A number of studies have been carried out to assess the behavior of these pollutants in the environment (e.g., review in Pacyna *et al.* 1993a). The results of these studies are summarized in Table 7·1 (after Nriagu 1984 and Pacyna and Winchester 1990). Environmental concentrations of many of the metals listed in this table are often higher than the concentrations expected from their natural occurrence in terrestrial and aquatic environments. Emissions of heavy metals, mostly on fine particles, during various human activities are the major cause of these increased concentrations, resulting in alterations of geochemical cycles of these metals.

The increase in concentration of a given metal, measured in a certain reference material, such as crustal rocks or soils, in relation to a certain reference metal, such as Al, Ti, or Sc, can be defined as the enrichment factor of this metal (EF). Most often, metals are enriched on a local scale, but some are enriched on regional and global scales. Regional scale is often defined as continental (1000-2000 km), whereas global scale is usually regarded as intercontinental, e.g., Northern Hemisphere. Episodes of long-range transport of pollutants within air masses result in the enrichment of metal concentrations far from source regions; the Arctic is a receptor of such transport (Pacyna and Winchester 1990).

The environmental and health effects of heavy metals depend greatly upon on the mobility of each metal through environmental compartments and the critical pathways through which the metals reach the human body. Almost all metals in Table 7·1 are either water/lipid soluble or volatile. Some metals are water/lipid soluble and volatile. Ingestion with food is the major pathway for many metals entering humans; however, quantitative information is incomplete for many metals.

Finally, the degree of concern about human and environmental health varies with each metal. Some metals are clearly toxic. Others are known to be essential micronutrients for humans and animals. The true importance of some metals to human and animal health is not known due to incomplete information. The general indication of potential health concern for some heavy metals is also noted in Table 7·1.

Taking into account the available information on the behavior and effects of heavy metals, most studies focus on Hg, Cd, and Pb. Fewer studies target As, Cu, Cr, Ni, V, Se, and Zn. These two groups of heavy metals have been pro-

Table 7·1. Perturbations of the geochemical cycles of trace metals by society (Nriagu 1984, Pacyna and Winchester 1990).

Element	Scale of perturbation[a]			Most diagnostic environments[b]	Mobility[c]	Health concern[d]	Critical pathway[e]
	Global	Regional	Local				
Pb	+	+	+	A, Sd, I, W, H, So	v, a	+	F, A[f]
V	+	+, c	+	A	[g]	(+)	A?
As	+	+	+	A, Sd, So, W	v, s, a	+	A, W
Sn	+	+	+	A, Sd, W	v, a	+[8]	F
Zn	+	+	+	A, Sd, W, So	v, s	E	F
Cd	+	+	+	A, Sd, So, W	v, s	+	F
Hg	+	+	+	A, Sd, Fish, So	v, a	+[h]	F, (A)
Sb	+	+	+	A, Sd	v, s	(+)	F, W, A?
Cu	+	+	+	A, Sd, W, So	v, s	E	F?
Ag	+	+	+	A, Sd, W	(v)	(+)	?
Se	+	(+)	+	A	v, s, a	E	F
Ge	?	+	+	A, So, W?	v, s, a	(+)[h]	?
Ni	(+)	+	+	A, Sd	–	E	F, W, A?
Cr	–	+	+	A, Sd, W, Gw	s, v[i]	E	W, F
B	–	(+)	+	A, Sd, Gw	v, s	E	W
K	–	(+)	+	A	s	E	F
Pt	?	?	+	A, Sd	s	(+)	?
Pd	?	?	+	Sd	s	(+)	?
Mo	?	?	+	A, W, So, Sd	s	E	F, W
Tl	?	?	+	Em, So	v, s	(+)	A, F?
In	?	?	+	A, So, Em	v	(+)	?
Bi	?	?	+	A, So, Em	v	(+)	?
Be	?	?	+	A, So, Em	–	(+)	A
Ga	?	?	+	Em	v	(+)	?
Te	?	?	(+)	So	v, a?	(+)	?

a. +: significant perturbation; (+): possible perturbation; – : no perturbation; ?: not enough information; c: enhanced due to mobilization of crustal materials (soil, dust).
b. A: air; Sd: sediments (coastal, lake); So: soils; I: ice cores; W: surface waters; Gw: groundwaters; H: humans; Em: emission studies (only listed when little geochemical information is available).
c. v: volatile; s: soluble; r: soluble only under reducing conditions; a: mobile as alkylated organometallic species; –: not mobile.
d. +: toxic in excess; (+): toxic, but little data available; E: essential, but toxic in excess.
e. F: food; W: water; A: air;
f. Exposure through hand-to-mouth activity is critical for lead in children.
g. Enriched relative to crustal abundance from fuel oil combustion (vanadium porphyrins).
h. Organometallic forms only.
i. Hexavalent form volatile and toxic, trivalent form essential.

posed by the United Nations Economic Commission for Europe (UN ECE), as well as by international programs involved in studies of the transport of pollutants to the North Sea and the Baltic Sea, as a priority list for emission reduction policies for heavy metals.

The Arctic region is a major receptor of heavy metals generated in other regions of the Northern Hemisphere (Rahn and Lowenthal 1984, Maenhaut *et al.* 1989, Barrie 1991, Shaw 1991b, Cheng *et al.* 1993). The aims of this chapter are 1) to assess emission sources and fluxes of heavy metals to and in the Arctic, 2) to describe the concentration of metals in various environmental compartments, 3) to assess the degree of bioaccumulation and biomagnification of heavy metals in Arctic biota, and 4) to assess the environmental effects of these pollutants in the Arctic. A major focus is placed on the priority heavy metals noted above, particularly Cd, Pb, Hg, and Se.

7.1.2. Speciation of metals in the environment

Atmosphere

Information on the chemical speciation of heavy metals emitted to the atmosphere is limited. For Hg, most of the emissions from combustion of fuels occurs in the gaseous phase. In the combustion zone, Hg present in coal or other fossil fuels evaporates in elemental form. Some of it is then oxidized while in the flue gases. The oxidized forms of Hg can be retained in modern flue gas cleaning systems. Mercury retained in fly ash (as well as in bottom ash) is often disposed of on land, after which some is transported to the aquatic environment. While it is difficult to quantify these transport processes, Nriagu and Pacyna (1988) estimated that Hg in coal fly ash and bottom ash contribute up to 40% of the direct releases of the element to the terrestrial environment. The emission generation process for Hg during the incineration of wastes is similar to that during combustion of fossil fuels. However, more Hg in the oxidized form is expected from incinerators due to the higher content of chlorine in waste matter than in fossil fuels.

The major chemical forms of As, Cd, and Pb created by the main emission source categories are presented in Table 7·2. The inorganic forms of As (particularly trivalent As), from sources including smelters or coal-fired power plants, dominate in the air over emission areas. It is important to observe that inorganic compounds of arsenic generally are more toxic than organic arsenic compounds, and that the trivalent forms are more toxic than the other forms. Methylated forms of As are probably of minor significance.

Elemental Cd (Cd(0)) and its oxide are the predominant chemical forms of the metal emitted from major sources. These two forms seem to be the most toxic Cd species, together with Cd chloride, which is found in emissions from waste incineration. For Pb, inorganic forms are the most widely released chemical species, particularly Pb oxide, Pb chloride, and Pb sulfates.

The relative volatility of the chemical species presented in Table 7·2 is as follows: elemental As, its trioxide and chloride, elemental Cd and its chloride, and elemental Pb are volatile; Pb chloride is intermediate; and the oxides of Cd and Pb are non-volatile. The volatile species occur in a vapor phase during emission generation, whereas the non-volatile compounds are emitted largely as fly ash, even at higher temperatures.

Aquatic systems

A complete understanding of chemical speciation is essential for gaining a comprehensive understanding of the chemical status of aquatic ecosystems. This is in turn essential for evaluating the risk to the health of the ecosystems and individuals within them as a result of exposure to metals, and for being able to predict how changes in environmental parameters will influence bioavailability, bioaccumulation, and the toxic effects of metals. An overview of metal speciation in aquatic systems is provided below. Greater detail is pro-

Table 7·2. Major chemical species created evolved during fossil fuel combustion and industrial processes. Me(O): elemental form of a given heavy metal.

Process	As	Cd	Pb
Coal combustion	As(0), As_2O_3, As_2S_3	Cd(0) , CdO, CdS	$PbCl_2$, PbO, PbS, Pb
Oil combustion	As(0), As_2O_3, Organic arsines	Cd(0), CdO	PbO
Non-ferrous metal production	As_2O_3	CdO, CdS	PbO, $PbSO_4$, PbO, $PbSO_4$
Iron and steel manufacturing		CdO	PbO
Refuse incineration	As(0), As_2O_3, $AsCl_3$	Cd(0), CdO, $CdCl_2$	Pb(0), PbO, $PbCl_2$

vided in a number of good reviews (Sadiq 1992, Webb 1979, Nriagu 1980, Furness and Rainbow 1990, Singer 1973, NRCC 1979a, 1979b, 1979c, 1979d, 1981a, 1981b, Elinder 1984, Moore and Ramamoorthy 1987, Linnik and Nabivanets 1986)

The speciation of heavy metals in aquatic systems is controlled by a number of factors including ionic strength, pH, redox potential, presence of organic ligands, and temperature. Metals occur broadly in particulate and soluble forms. Possible species include free aquated ions; complex ionic entities; inorganic ion-pairs and complexes; organic complexes, chelates and compounds; metals bound to high molecular weight organic materials; highly dispersed colloids; metals sorbed on colloids; precipitates; and metals present in live and dead biota. Various species are in –dynamic equilibrium with each other and thus the relative proportion of each species in an aqueous medium is determined by thermodynamic and kinetic relationships. Using Cd as an example, the following is a comparison of the dissolved chemical species present in 'ideal' seawater and river water at 25°C. In seawater, over 90% of the Cd is in the form of chloro complexes; of these $CdCl_2$ represents more than 50% ($CdCO_3 < Cd^{2+} < CdCl_3^- < CdCl^+ < CdCl_2$).

Equilibrium conditions in aquatic systems are often extremely complicated and difficult to measure; consequently, chemical species in the system are often estimated from thermodynamic solubility relationships.

Mercury

Mercury exists in natural waters in three oxidation states: Hg(0), Hg(I), and Hg(II). Hg(II) forms hydroxocomplexes even at low pH values. These complexes predominate among inorganic forms of Hg under natural conditions ($pH > 6$), even in the presence of considerable concentrations of chloride ions. Mercury can also form stable complexes with many organic ligands, especially those containing sulfur (amino acids, oxycarbonic acids etc.) and high molecular natural compounds like fulvic and humic acids. In natural waters, Hg compounds are strongly bound to particulate matter. In particulates, Hg is readily transported by river and accumulates in bottom sediments.

In addition to hydrophobic organic complexes, Hg(II) can form water- and lipid-soluble alkylderivative compounds, primarily methylmercury. There are two pathways of methylation: microbiological and chemical. The later mechanism usually involves natural organic substances, mostly fulvic acids. Varshal and Buachidze (1983) report that 12 hours after discharging Hg(II) into water containing high concentrations of fulvic acid, up to 34% of the Hg is transformed into alkyl-mercury compounds.

Cadmium

Cadmium is usually present in the environment as Cd(II) and starts to be hydrolyzed at pH 9. The complexing ability of Cd, even with humic and fulvic acids, is not as strong as other heavy metals; in natural waters it is predominantly present in dissolved (Cd(II)) form. Cadmium forms low solubility compounds with several anions (primarily carbonate and phosphate) which control the concentration of Cd(II) under natural conditions (pH 8-9). Low concentrations of these anions favor the adsorption of Cd(II) by particulate matter, clay particles in particular, and Cd is transported in this form. Adsorption of Cd(II) by sediments increases with increasing pH; at $pH > 7$, nearly all Cd is in the adsorbed phase (Moore and Ramamoorthy 1987).

Lead

Lead usually exists in natural waters as Pb(II). It reacts readily with some major anions (CO_3^{2-}, SO_4^{2-}) at pH values typical of natural waters. Lead has a high complexing ability and forms stable complexes with S-, P-, O-, and N-containing organic ligands which leads to its accumulation in live and dead aquatic biota. At low concentrations of soluble organic ligands, Pb exists primarily in particulate form at $pH > 6$. The proportion of particle-bound Pb to total Pb in the world's river waters varies from 15 to 83% depending on water composition. Like Hg, Pb can be microbiologically methylated in bottom sediments.

The speciation of metals in aquatic systems has important consequences for both bioaccumulation and toxicity. In freshwater, free metal ions of Pb, Cd, and Hg are much more abundant than in natural waters containing chloride ion and other complexing anions; in general these divalent cations are considered to be more toxic than the soluble complexed forms of Pb, Cd, or Hg. Consequently, the toxic effects of Pb, Cd, or Hg are frequently more pronounced in freshwater systems than in estuarine or marine systems, even through the concentrations of dissolved metals are higher in the latter (complexation increases solubility, but usually reduces toxicity).

7.2. Sources of pollution

Heavy metals are emitted to the atmosphere from both natural and anthropogenic sources. Very few of the sources that directly result in the contamination of the Arctic environment are located in the Arctic. Metals released to the environment outside the Arctic are transported to the Arctic via air currents, rivers, and ocean currents. This section reviews the sources and fluxes of heavy metals within the Arctic as well as their sources outside and pathways to the Arctic.

7.2.1. Natural sources

An accurate inventory of heavy metal sources and emissions to the atmosphere from natural processes is needed to make a complete assessment of the extent of regional and global pollution by heavy metals in the Arctic. It is generally presumed that the principal natural sources of heavy metals include wind-borne soil particles, volcanoes, seasalt spray, and wild forest fires (Nriagu 1989). Recent studies have shown, however, that particulate organic matter is the dominant component of atmospheric aerosols in non-urban areas (Talbot *et al.* 1988, Artaxo *et al.* 1988) and that over 60% of the airborne heavy metals in forested regions can be attributed to aerosols of biogenic origin (Zoller 1983).

A global assessment of natural sources of atmospheric heavy metals has been made by Nriagu (1989). A summary of these estimates is presented in Figure 7·1. Biogenic sources can account, on average, for over 50% of the Se, Hg, and Mo, and from 30 to 50% of the As, Cd, Cu, Mn, Pb, and Zn, released annually to the atmosphere from natural sources. Volcanic emissions can account for 40-50% of the Cd and Hg and 20-40% of the As, Cr, Cu, Ni, Pb, and Sb released annually from natural sources. Seasalt aerosols seem to account for <10% of atmospheric heavy metals from natural sources. Finally, soil-derived dusts can account for over 50% of the total Cr, Mn, and V emissions, as well as for 20-30% of the Cu, Mo, Ni, Pb, Sb, and Zn released annually to the atmosphere. As the accuracy of emission estimates for natural sources is low, these percentage contributions should be considered as approximations only.

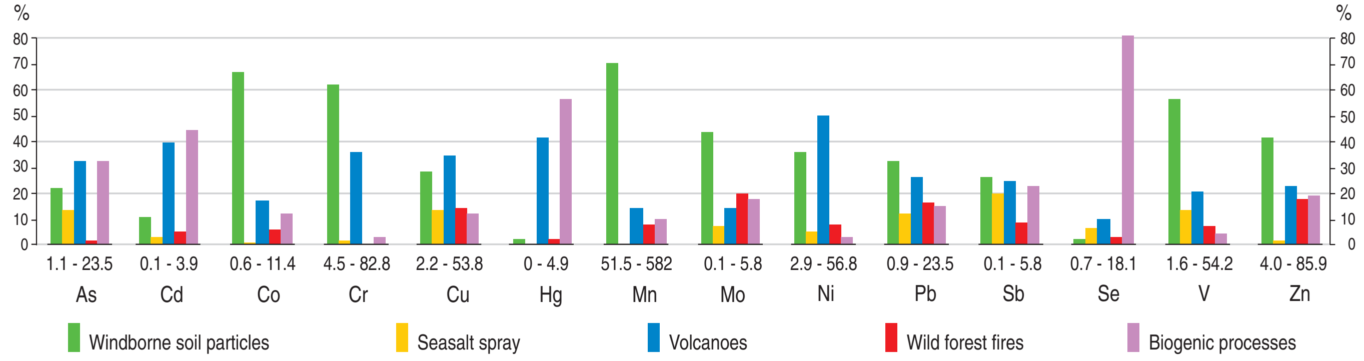


Figure 7·1. Global emissions of trace metals to the atmosphere from natural sources (after Nriagu 1989). Numbers under the columns are the range of estimates of the emissions in thousands of tonnes per year. The percentages shown by the bars are calculated using the maximum value of the range of the total and individual source category estimates.

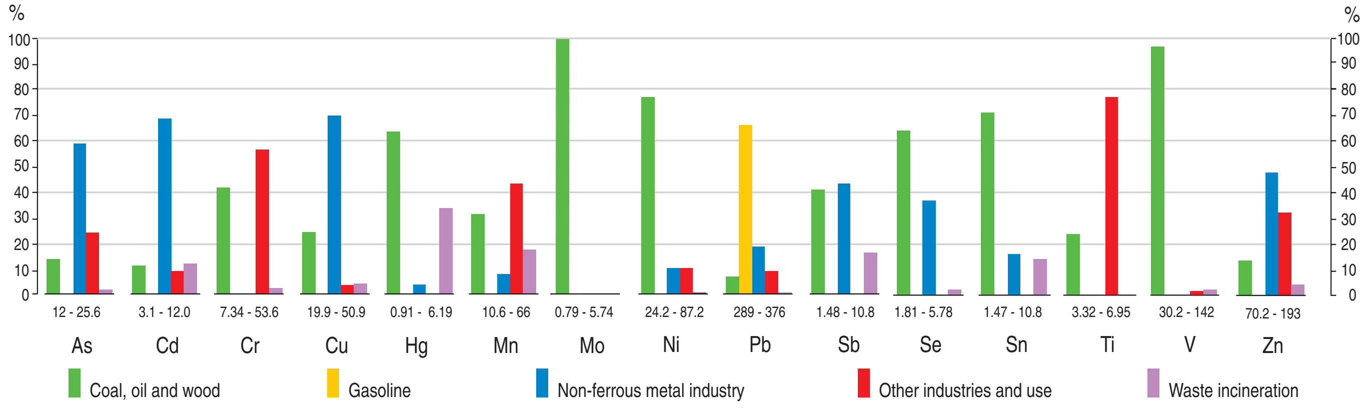


Figure 7·2. Global emissions of trace metals to the atmosphere at the beginning of the 1980s from anthropogenic sources (after Nriagu and Pacyna 1988). Numbers under the columns are the range of estimates of the emissions in thousands of tonnes per year. The percentages shown by the bars are calculated using the maximum value of the range of the total and individual source category estimates.

The natural sources of heavy metals which influence the freshwater, terrestrial, and marine environment are even more difficult to assess than the atmospheric sources. In general, soils and sediments tend to reflect the composition of their parent material. Soils and sediments in mineralized areas, therefore, usually have the highest concentrations of the corresponding metals. For example, rocks with high Hg content usually occur in areas of crustal instability where volcanic and geothermal activity are high.

It is also very difficult to assess the extent to which emissions from natural processes affect the contamination of the Arctic environment. In general, fluxes from these processes within the Arctic are regarded as less significant than anthropogenic releases, both within and outside the Arctic. However, very long range transport within air masses of soil particles from deserts in Asia and Africa to the High Arctic has been postulated by Pacyna and Ottar (1988). A series of haze bands over Barrow, Alaska in April and May 1976 were found to consists of dust (Rahn *et al.* 1981). The bulk elemental composition of the particles was crustal or near-crustal and their mass-median radius of about 2 μm indicated that they could have originated more than 5000 km from Alaska. Trajectory analysis showed that these particles could have passed over the arid and semi-arid regions of eastern Asia during intense dust storms which had occurred there. This hypothesis has been confirmed by measurements in the Norwegian Arctic (Pacyna and Ottar 1989) and in the Canadian Arctic (Welch *et al.* 1991). The origin and evolution of dust clouds in central Asia has recently received consideration. The existence of natural constituents in the Arctic aerosol in central Asia was explained by long-range transport of eroded dust from the deserts in Asia and Africa during dust storms. However, no quantitative assessment has been made of how much of the eroded dust and attached heavy metals is transported from the Asian and African deserts to the Arctic.

7.2.2. Anthropogenic sources

High temperature processes generate various heavy metals. These processes include coal and oil combustion in electric power stations and heating and industrial plants, gasoline combustion, roasting and smelting of ores in non-ferrous metal smelters, melting operations in ferrous foundries, refuse incineration, and kiln operations in cement plants. The metals enter the atmosphere and the aquatic and terrestrial ecosystems; virtually every industry discharges heavy metals into these ecosystems. The assessment presented here is focused on the principal industrial and commercial users of raw materials and water, and on producers of solid wastes.

7.2.2.1. Sources and fluxes of atmospheric input on a global scale

The first quantitative worldwide estimate of the annual industrial input of 16 heavy elements into air, soil, and water was published by Nriagu and Pacyna (1988). The summary of the estimate of atmospheric emissions is presented in Figure 7·2. Pyrometallurgical processes in the primary non-ferrous metal industries are the major source of atmospheric As, Cd, Cu, In, Sb, and Zn, and an important source of Pb and Se. Combustion of coal in electric power plants and in-

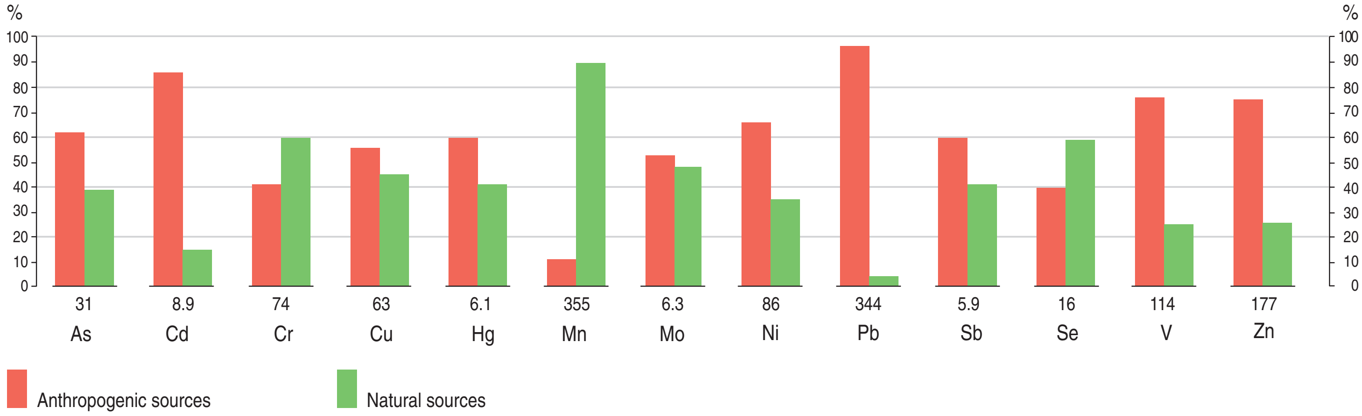


Figure 7·3. Comparison of global emissions of trace metals to the atmosphere from natural and anthropogenic sources in 1983. Numbers under the columns are the median values of estimates of total emissions in thousands of tonnes per year. The percentages shown by the bars are calculated from the median values of the ranges of the estimates for natural and anthropogenic sources.

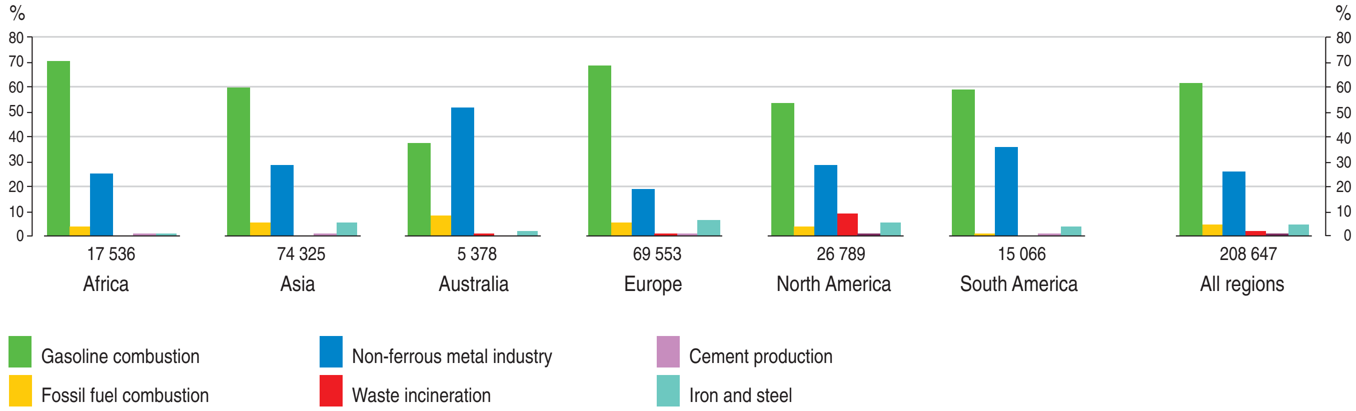


Figure 7·4. Global emissions of Pb to the atmosphere from various sources and source regions in 1989. Numbers under the columns are maximum estimates of the total emissions in tonnes.

dustrial, commercial, and residential burners is the major source of anthropogenic Hg, Mo, and Se and a significant source of As, Cr, Mn, Sb, and Ti. Combustion of oil for the same purpose is the most important source of V and Ni.

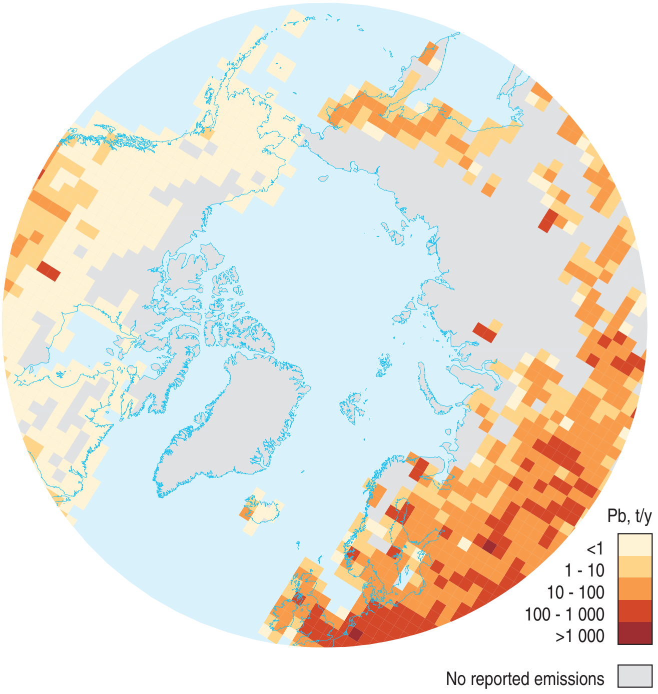


Figure 7·5. Spatial distribution of emissions of Pb within the 150 km × 150 km EMEP grid system for the area north of latitude 50°N in 1989. (Source of data: Norwegian Meteorological Institute, after Pacyna *et al.* 1993b).

Combustion of leaded gasoline is still the major source of Pb. Chromium and Mn are derived primarily from the iron and steel industry. Little information is available on the emission of heavy metals from various diffuse (fugitive) sources.

A comparison of the median values of worldwide emissions of heavy metals from natural and anthropogenic sources (Figure 7·3) suggests that human activities generate emissions of heavy metals that exceed those from natural sources. Therefore, anthropogenic emissions result in significant alterations of the natural biogeochemical cycling of many heavy metals in the global environment.

Recently, a revision of the global emission inventory of Pb was prepared for the reference year 1989 (Pacyna *et al.* 1993b) as a part of the Global Emission Inventory Activities (GEIA) operated within the IGBP International Global Atmospheric Chemistry (IGAC) program. The results show that in 1989 the maximum emission was about 209 000 tonnes of Pb, of which 62% came from gasoline combustion, followed by 26% from non-ferrous metal production (Pacyna *et al.* 1995). The summary of the results showing the maximum emission estimates is presented in Figure 7·4. One-third of the total estimated emissions of the element originates in Asia and Europe. The spatial distribution of these emissions within the 150 × 150 km EMEP grid system for the area north of latitude 50°N is presented in Figure 7·5.

An estimate of global anthropogenic emissions of Hg has also been completed for AMAP (Pacyna and Pacyna 1996). The spatial distribution of these emissions within a 1° × 1° grid is presented in Figure 7·6, and the contributions from different continents and different sources are presented in Figure 7·7. These preliminary data suggest that between 1300 and 2150 tonnes of Hg are emitted annually to the at-

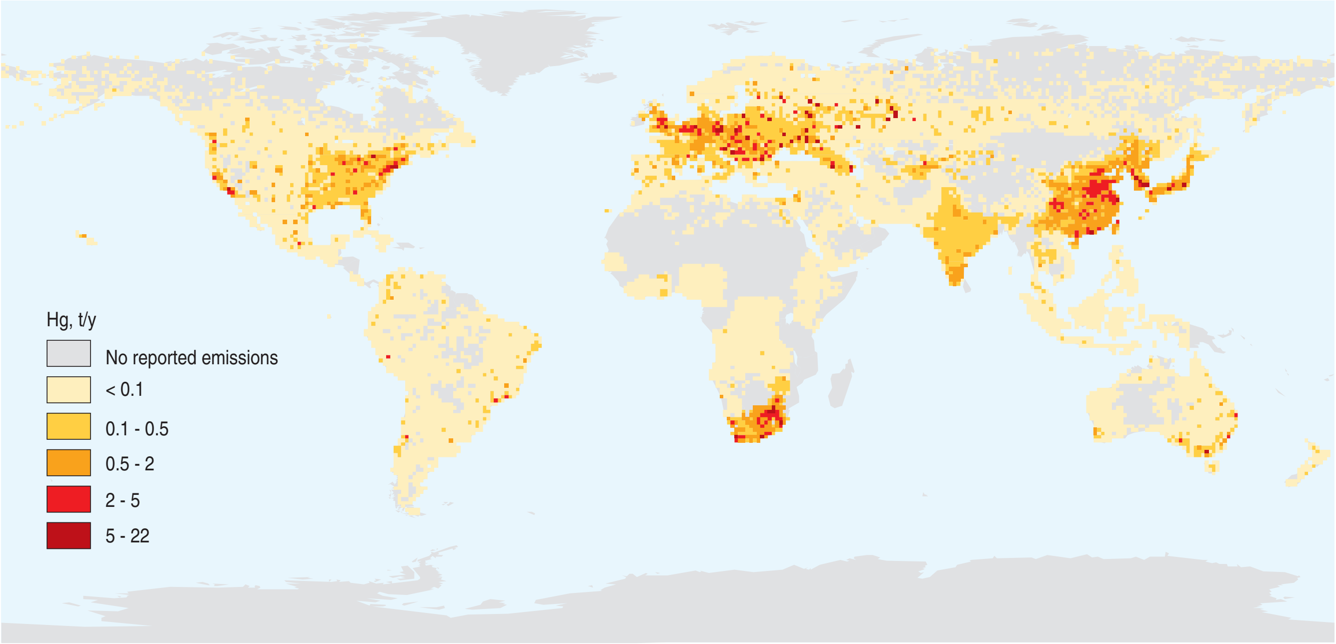


Figure 7·6. Spatial distribution of global emissions of Hg in 1990 within a 1° × 1° grid. The total emission inventory is 2144 tonnes Hg. (Source of data: Jozef Pacyna pers. comm., Canadian Global Emmissions Interpretation Centre (CGEIC)).

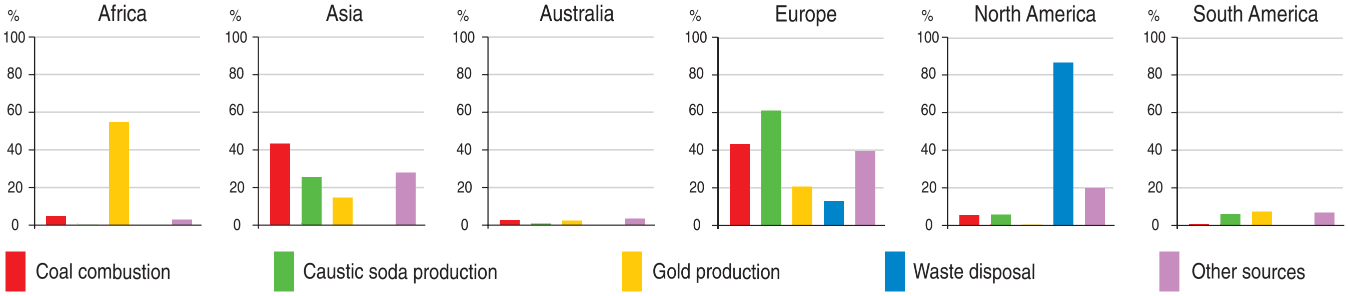


Figure 7·7. Contributions from different continents and from different sources to the global emissions of Hg to the atmosphere.

mosphere at the present time. The major sources of these emissions are combustion of coal to produce electricity and heat (60%), followed by gold production and waste disposal.

The above-mentioned studies on global releases of heavy metals are based on a number of research projects conducted to estimate the atmospheric emissions of heavy metals in Europe, North America, and Asia. Emissions from these areas relevant to the Arctic are reviewed below.

7.2.2.2. **Sources and fluxes of aquatic input on a global scale**

The major sources of anthropogenic heavy metal contamination of aquatic ecosystems (including the ocean) include domestic wastewater effluents (especially As, Cr, Cu, Mn, and Ni), coal-burning power plants (As, Hg, and Se in particular), non-ferrous metal smelters (Cd, Ni, Pb, and Se), iron and steel plants (Cr, Mo, Sb, and Zn), and the dumping of sewage sludge (As, Mn, and Pb). The atmosphere is the major route of Pb entry into natural waters and also accounts for over 40% of the V loading. The results of the worldwide assessment of anthropogenic inputs of 13 heavy metals into the aquatic ecosystem is presented in Figure 7·8 (Nriagu and Pacyna 1988).

A comparison of data in Figures 7·2 and 7·8 indicates that for most of the heavy metals, the annual anthropogenic inputs into water exceed the quantities emitted to the atmosphere. If it is assumed that only 25% of the industrial effluents are discharged into lakes and rivers, the average concentrations in these waters should reach levels several-fold

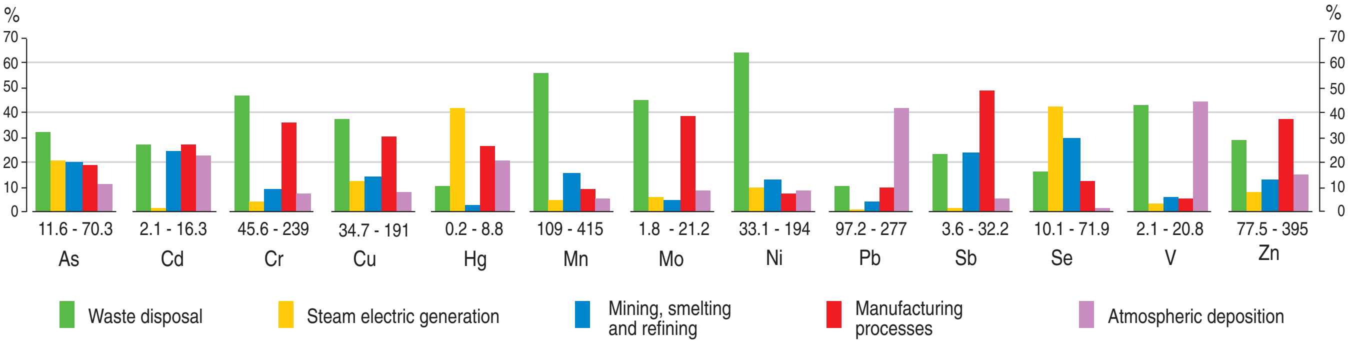


Figure 7·8. Global anthropogenic inputs of trace metals to aquatic ecosystems at the beginning of the 1980s (after Nriagu and Pacyna 1988). Numbers under the columns are the range of estimates of the inputs in thousands of tonnes per year. The percentages shown by the bars are calculated using the maximum value of the range of the total and individual source category estimates.

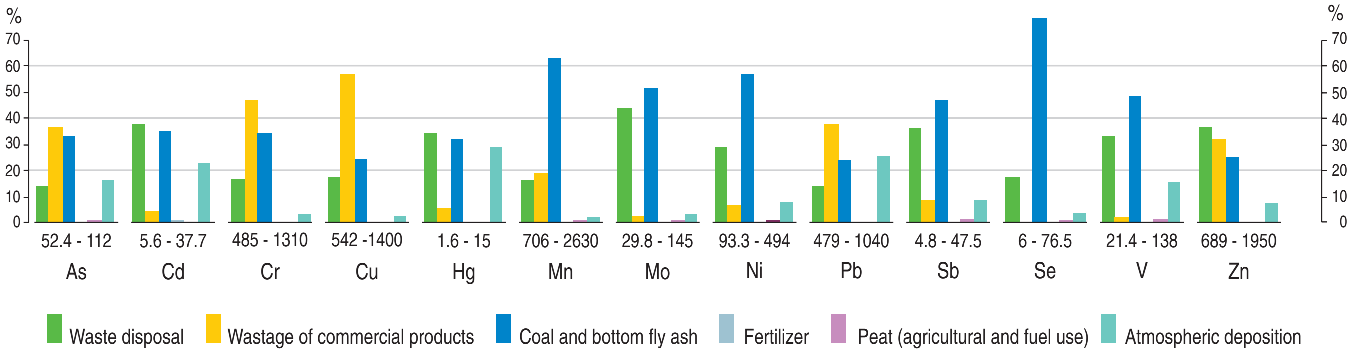


Figure 7·9. Global anthropogenic inputs of trace metals to soils at the beginning of the 1980s (after Nriagu and Pacyna 1988). Numbers under the columns are the range of estimates of the inputs in thousands of tonnes per year. The percentages shown by the bars are calculated using the maximum value of the range of the total and individual source category estimates.

higher than those in unpolluted lakes and rivers. In other words, the current rate of worldwide industrial inputs greatly exceeds the baseline burdens of heavy metals in the average lake and river. Most of the effluent discharges occur in Europe (including Russia), North America, and some Asian countries, implying that the contamination of the freshwater resources in these regions may be much more severe than is generally realized.

7.2.2.3. Terrestrial input and output of heavy metals on a global scale

The first quantitative assessment of worldwide fluxes of heavy metals into soils was prepared by Nriagu and Pacyna (1988) for the reference year 1983. A summary of this work is presented in Figure 7·9. The estimates suggest that soils are receiving large quantities of heavy metals from disposal of a variety of industrial wastes. The two principal sources of heavy metals in soils worldwide are the disposal of ash residues from coal combustion and the general breakdown and weathering of commercial products on land. Urban refuse represents an important source of Cu, Hg, Pb, and Zn with notable contributions of Cd, Pb, and V also coming via the atmosphere. The large volumes of wastes associated with animal husbandry, logging, and agricultural and food production can significantly affect the heavy metal budget of many soils. Although municipal sewage sludge may not be a particularly important source on a global scale, it can be one of the most important sources of metal contamination of soils on a local scale.

If the estimated metal inputs are distributed uniformly over the cultivated land area, the annual rates of metal application are not very significant because of the large background reservoir of heavy metals. Nevertheless, each soil has a limited retention capacity for heavy metals and there is growing concern that, at the current rate of anthropogenic input, many soils in various parts of the world (e.g., central Europe and Japan) either have become or will soon become overloaded with heavy metals (Kabata-Pendias 1984, Asami 1988).

Soils, like waters, can be a source of atmospheric contamination of some heavy metals, particularly Hg. These emissions result from various out-gassing of Hg laden rock and from volatilization of Hg from soils, vegetation, and water bodies. Current data suggest that these emissions of Hg are of the same order as emissions from anthropogenic sources (Pacyna and Keeler 1994).

It should be noted that emissions from natural sources are difficult to distinguish from so-called secondary emissions and diffusive re-emissions from anthropogenic sources. These include re-emissions of previously deposited Hg as well as emissions resulting from discharge into water bodies and from contaminated soils. Hence, it is more appropriate to differentiate between pre-industrial and post-industrial diffuse sources (Lindqvist 1991) than between natural and anthropogenic re-emission.

Only sparse information is available on re-emission of other heavy metals from soils and water surfaces. Oceans can be an important source of Se emissions to the atmosphere on a global scale, contributing as much as 25% to the total emissions of the element (Nriagu 1989).

7.2.3. Emission inventories for sources within and outside the Arctic

Obviously, only a part of worldwide emissions is responsible for the contamination of the Arctic environment by heavy metals. It is critical to identify sources important to the Arctic and to quantify the amount of emissions from these sources that reaches the Arctic region. The results of source-receptor studies, summarized in Pacyna (1991), indicate that emissions from sources in Eurasia contribute more than half of the air pollution measured in the Arctic. The major source regions include the Urals, the Kola Peninsula, the Norilsk area, and the industrial regions in Central and Eastern Europe (Rahn and McCaffrey 1980, Rahn and Lowenthal 1984).

The contributions of European and North American emissions to Arctic air pollution seem to be smaller than the contribution from the Russian sources. European and North American emissions are, however, major contributors to the

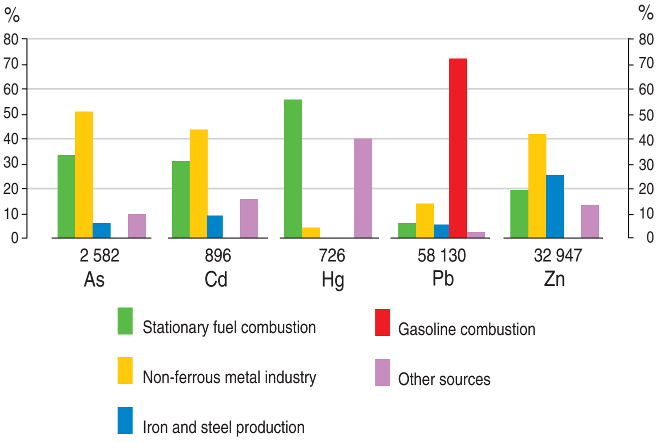


Figure 7·10. Emissions of selected heavy metals to the atmosphere in Europe at the beginning of the 1990s. Numbers under the columns are emissions in tonnes per year.

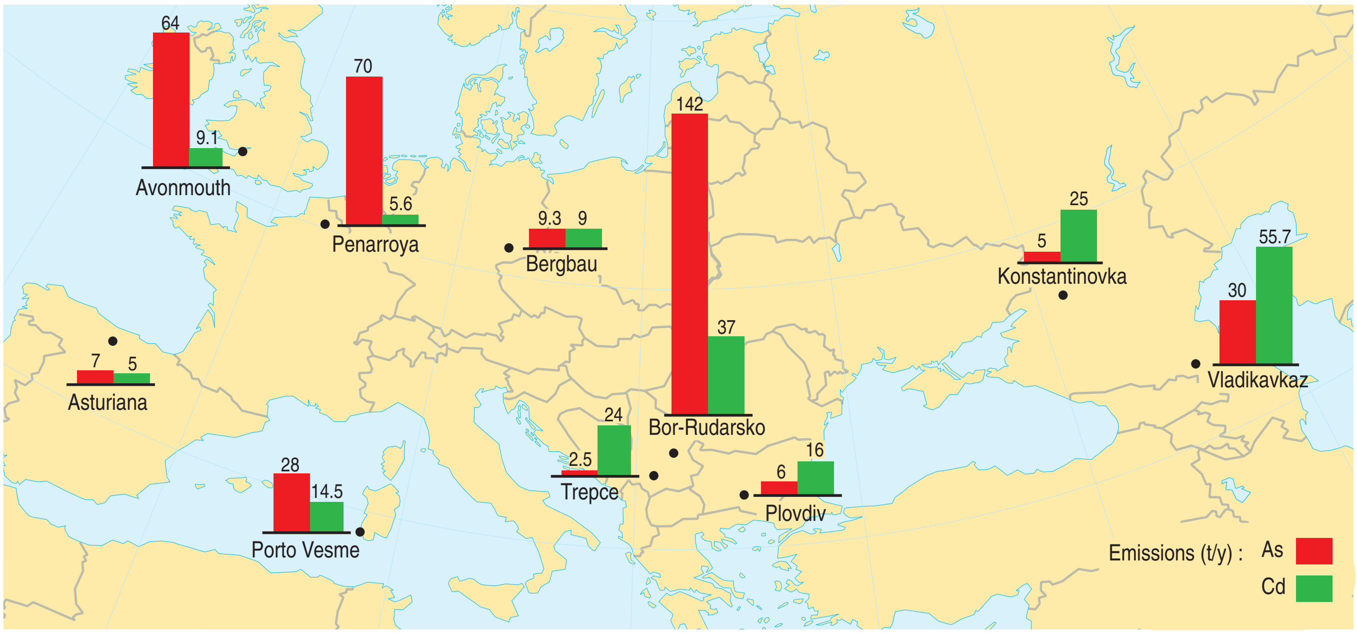


Figure 7·11. The ten major point sources, and their emissions of As and Cd to the atmosphere, in Europe at the beginning of the 1990s. Numbers above the columns are emissions in tonnes per year.

contamination of the subarctic regions, such as northern Scandinavia (European emissions) and the northern part of Canada (North American emissions).

These sources, located outside the Arctic, are important in discussing the contamination of the High Arctic environment, which remains largely unaffected by local industrial activities. It is necessary, therefore, to review the emissions from these outside sources in order to assess quantitatively their contribution to Arctic contamination.

7.2.3.1. **Atmospheric emissions from sources outside the Arctic and their trends**

The first attempt to estimate atmospheric emissions of heavy metals from anthropogenic sources in Europe was completed at the beginning of the 1980s (Pacyna 1984). This European survey has since been updated, completed, and emission gridded (Axenfeld *et al.* 1992, a review by Pacyna 1994). National emission inventories have only recently become available in the European countries. These emission data, together with the international expert estimates, were used to compile current European emission estimates for As, Cd, Hg, Pb, and Zn (Figure 7·10). A spatial distribution of the heavy metal emission estimates in Europe is available within the EMEP 150 km by 150 km grid system (Axenfeld *et al.* 1992). The ten major European point sources of heavy metal emissions to the air at the beginning of the 1990s are shown in Figure 7·11.

Changes of heavy metal emissions to the atmosphere from sources in Europe from the 1950s until present have also been studied. The results are presented in Figure 7·12 (Olendrzynski *et al.* 1995). A decreasing trend of Hg levels in atmospheric deposition in Scandinavia has been observed during the last few years (Munthe *et al.* 1994). This trend was related to a possible decline of Hg emissions, particularly in Central and Eastern Europe. These emissions can be expected to decrease by up to 30%. The economic decline in this part of Europe at the beginning of the 1990s, related to the transition from centrally planned economies to market oriented ones, was suggested as the major reason for the possible changes in Hg emissions. In addition, the lower consumption of fuels in Eastern Europe in recent years is an important factor which has caused the decrease of Hg emissions.

Pacyna *et al.* (1991) project that by using best available technology, As and Cd emissions in Europe should decrease by a factor of 3 and 2, respectively, by the year 2000. A prompt switch to unleaded gasoline should reduce Pb emissions in Europe by a factor of 4-10.

Pursuant to the requirements of the 1990 US Clean Air Act Amendment, an interim toxic emission inventory has

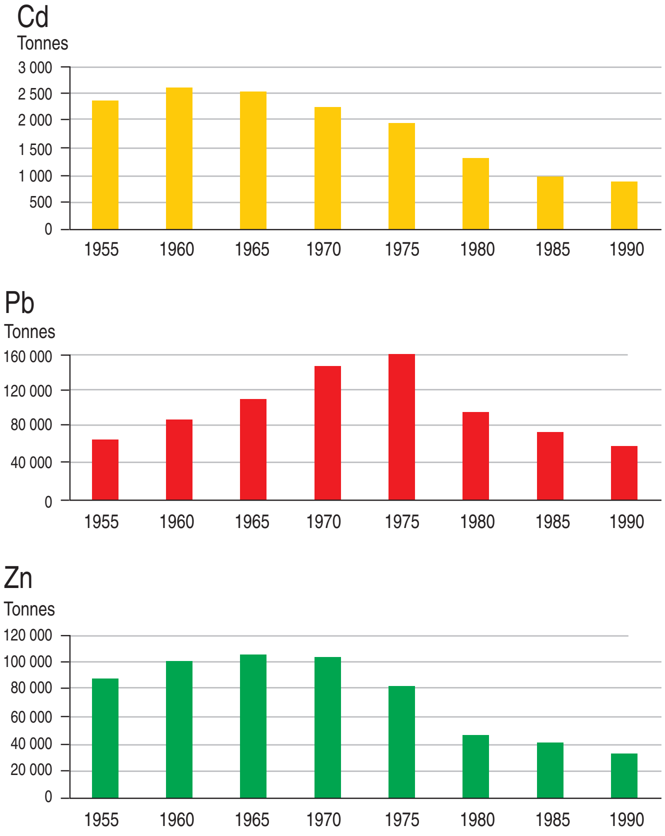


Figure 7·12. Changes with time in European atmospheric emissions of Cd, Pb and Zn.

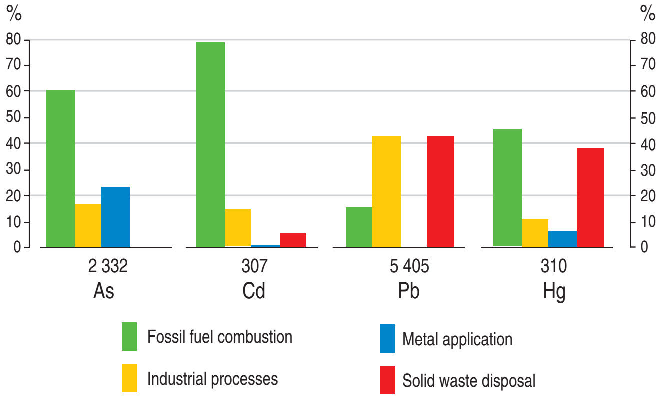


Figure 7·13. Estimates of emissions of As, Cd, Pb and Hg to the atmosphere from major source categories in the United States. Numbers under the columns are emissions in tonnes per year. (Source of data: As and Pb, Voldner and Smith 1989; Cd, US EPA 1993a; Hg, US EPA 1993b).

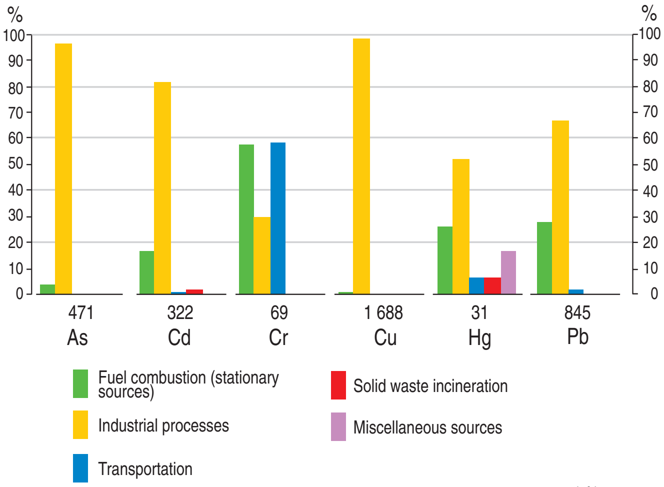


Figure 7·14. Estimates of emissions of As, Cd, Cr, Cu, Hg and Pb to the atmosphere from major source categories in Canada in 1982 (after Jaques 1987). Numbers under the columns are emissions in tonnes per year.

been developed for the continental United States. Emission estimates for As, Pb (both after Voldner and Smith 1989), Cd (after US EPA 1993a), and Hg (after US EPA 1993b) from major source categories in the United States are presented in Figure 7·13.

Environment Canada has initiated several projects on emission inventory development for heavy metals in Canada. In Figure 7·14, the 1982 emissions of As, Cd, Cr, Cu, Hg, and Pb from major source categories are presented on the basis of data from Jacques (1987). Emission estimates for Hg and Pb have been revised to account for major changes in consumer patterns in recent years.

It is believed that the reliability of emission data from Europe and North America decreases in the following order:

Pb > Hg and Cd > remaining heavy metals.

An accuracy of <25% was suggested for the emission estimates of Pb, 50% or less for Cd and Hg, and 100% for the rest of the metals (Pacyna 1994).

7.2.3.2. Atmospheric emissions from sources within the Arctic

In addition to outside sources, there are also sources of heavy metals within the Arctic. Combustion of fossil fuels to produce electricity and heat is one of the major source categories present in the region, followed by industrial processes in the Russian Arctic.

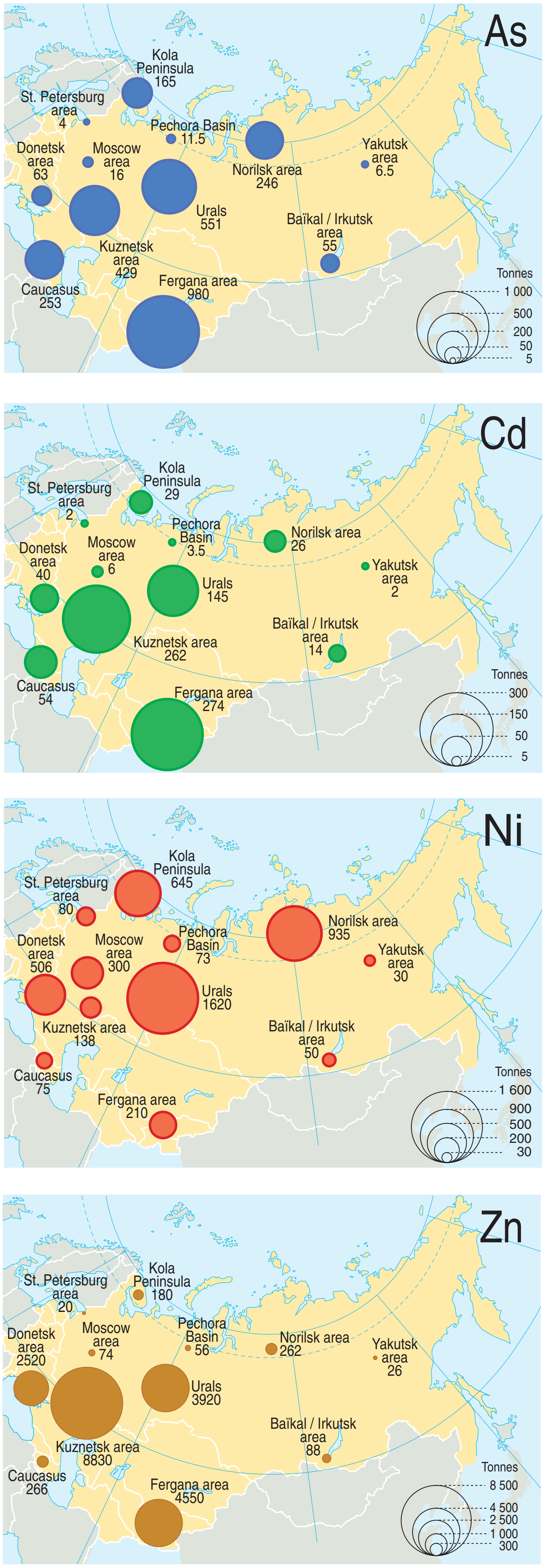


Figure 7·15. Emissions of As, Cd, Ni and Zn to the atmosphere from major sources in the former Soviet Union in 1979/80. (After NILU 1984).

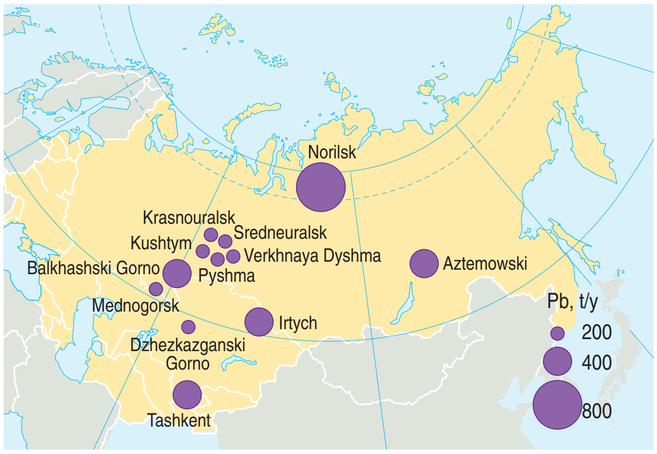


Figure 7·16. Major point sources (non-ferrous metal smelters) of Pb to the atmosphere in the Urals and the Asian part of the former Soviet Union. (After Pacyna *et al.* 1993b).

Preliminary estimates of atmospheric emissions of As, Cd, Cu, Cr, Mn, Ni, Pb, Sb, Se, V, and Zn for major source regions in the former Soviet Union, including the areas outside of the Arctic, have been prepared by NILU (1984) on the basis of emission factors and statistical data for the reference year 1979/80; data for selected metals are shown in Figure 7·15. Emissions from non-ferrous and ferrous metal production, fossil fuel combustion, and gasoline combustion were estimated to dominate the total emissions in those regions entirely or partly located within the Arctic, such as the Kola Peninsula, the Norilsk area, and the northern Urals. The inventory of sources in the European part of the former Soviet Union is included in the European emission survey noted above. A list of major point sources of heavy metal emissions to the atmosphere in the Urals and the Asian part of the former Soviet Union is presented in Figure 7·16 (Pacyna *et al.* 1993b).

Sivertsen *et al.* (1992, 1994) studied the level of air pollution in 1989 in the border areas of Norway and Russia related to the Cu and Ni emissions from the sources on the Kola Peninsula. With respect to emissions of heavy metals, there are two major source regions on the Kola Peninsula: the Pechenganikel industrial complex consisting of the Nikel and Zapolyarnyy Cu-Ni smelters, and the Severonikel smelter complex. The emissions of Cu and Ni in the Pechenganikel smelter complex are estimated to be approximately 310 and 510 tonnes, respectively. However, very recent information (e.g., Pozniakov 1993, Lyangusova 1990) suggests that actual emissions could be about one order of magnitude higher. By contrast, the official Russian data place the 1994 emissions from Nikel and Zapolyarnyy at about 163 tonnes of Cu and 297 tonnes of Ni (CENR 1995).

During the 1980s, the Severonikel smelter complex became the largest Ni producer in the world with an annual output of 140 000 tonnes. Preliminary estimates of Cu and Ni emissions to the atmosphere from this source are approximately 3000 and 2700 tonnes, respectively (Pozniakov 1993). These emissions appear high, particularly when compared with the historical estimates of emissions from the Pechenganikel complex. The official Russian data place the 1994 emissions from Severonikel at about 934 tonnes of Cu and 1619 tonnes of Ni (CENR 1995). The high variability among various estimates for emission, however, emphasizes the need to verify the information on heavy metal emissions on the Kola Peninsula because these emissions contribute substantially to the contamination of the Norwegian Arctic and perhaps to that of a broader area.

7.2.3.3. Aquatic emissions from sources outside the Arctic

Compared with atmospheric emissions, much less information is available on emission inventories reporting discharges of heavy metals to the aquatic environment. In one of very few approaches in Europe, discharges of Cu, Pb, Ni, and Zn from major point sources were estimated for the Commission of the European Communities (EC) (Daamen *et al.* 1990). These estimates are presented in Figure 7·17. Ship building (including ship maintenance) is estimated to be the major industry discharging Cu to the aquatic environment in Europe. Nickel discharges in the EC region originate mainly from manufacturing of electrical equipment and the secondary transformation of metals. The manufacture of ceramics is by far the most important industry discharging Pb to the aquatic environment. Finally, Zn discharges in the EC region are mainly due to the manufacturing of basic industrial chemicals. Daamen *et al.* (1990) conclude that discharges of heavy

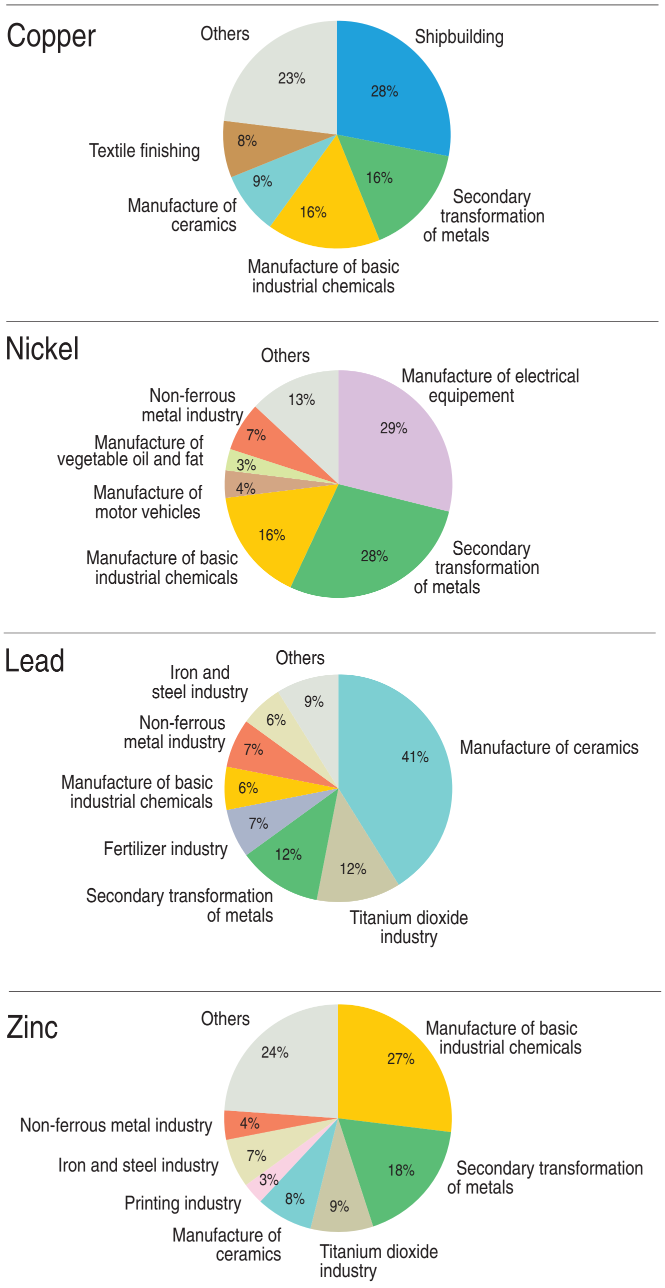


Figure 7·17. The relative importance of different branches of industry in discharges of Cu, Ni, Pb and Zn to the aquatic environment from major point sources in the European Community (EC). (After Daamen *et al.* 1990).

metals from the iron and steel industry, non-ferrous metal manufacturing, and mining in the CEC region are underestimated in the above-mentioned inventory due to lack of information.

7.2.3.4. Aquatic emissions from sources within the Arctic

In Greenland, mining has been an important source of local heavy metal pollution of the sea. Asmund *et al.* (1991) have made an estimate of quantities of Zn, Cd, and Pb released to the sea from two mines in Greenland. One of these mines, the Black Angel Pb-Zn mine in Uuummannaq, West Greenland, operated from 1973 to 1990. During production, it was estimated that 8-16 tonnes of Zn, 6-12 tonnes of Pb, and 50-120 kg of Cd were released annually to seawater. Dumping of waste rock at the time the mine was closed in 1990 added about 10 tonnes of Zn, 1 tonne of Pb, and 60 kg of Cd to this total (Asmund 1992a, 1992b). After mine closure, the amounts released have declined drastically, to 10% or less of the amounts measured during production (Asmund 1992a, 1992b). At a closed cryolite mine in Ivittuut in south Greenland, Asmund *et al.* (1991) estimate an annual input to the sea of 1-2.5 tonnes of Zn, 0.4-1 tonnes of Pb, and 4-10 kg of Cd from deposited waste rock in the intertidal zone. No estimates are available from a closed Pb mine at Mestersvig in East Greenland, which operated from 1956 to 1963, but environmental studies of seaweed, mussels, and fish indicate that significant amounts of Pb and Zn have entered the marine environment (Johansen *et al.* 1985).

A comparison of the Greenland mines with the Pb-Zn mine at Nanisivik in the Canadian Arctic shows that the Canadian mine has impacted the sea to a much lesser extent (Asmund *et al.* 1991). Information on the releases of metals from the Red Dog Pb-Zn mine in Alaska and their impact on the environment is not yet available.

The major mining and metallurgical activities on the Kola Peninsula and in the Norilsk region are sources of heavy metals into the aquatic environment (e.g., Falk-Petersen *et al.* 1992). For example, the Severonikel smelter in Monchegorsk discharged approximately 24 million m^3 of wastewater containing 54 tonnes of Ni during 1993 (NEFCO 1995). The waters around Novaya Zemlya are important dumping areas for various wastes, including toxic wastes. However, no quantitative assessment exists for the dumping of heavy metals contained in these wastes within this region. Heavy metals are also transported by the large rivers, such as the North Dvina, Pechora, Ob, Yenisey, and Lena, contributing to the contamination of the Arctic environment.

7.2.3.5. International agreements on emission reduction

A number of international efforts have been initiated to try to reduce heavy metal emissions to the environment. These include:

1. Paris Commission (PARCOM) Decisions. A variety of actions have been agreed to through the auspices of the Paris Commission to reduce heavy metal emissions to the air and marine environment.
2. Third North Sea Conference 1990. Many countries undertook to implement reduction of discharges of heavy metals.
3. Organization for Economic Co-operation and Development (OECD). The OECD is preparing a series of documents examining risk reduction measures for a variety of substances under the OECD Chemical Programme, including documents for heavy metals.

In 1988, the UN Economic Commission for Europe (UN ECE) established a Task Force on Heavy Metals with the aim of providing a state-of-the art report on heavy metals, examining the following issues: emission inventories, atmospheric dispersion and deposition, analytical problems, and abatement techniques including their economic aspects (UN ECE 1994). The ultimate goal of the Task Force is to prepare a substantiation document for a protocol on the reduction of heavy metal emissions in the ECE region as well as in the United States and Canada.

7.3. Special issues of pathways of metals

Pathways of metals reaching the Arctic are studied with the use of transport models. This section presents models simulating long-range transport of these compounds to the Arctic.

7.3.1. Atmospheric transport

Following release into the atmosphere, heavy metals can be either deposited in the vicinity of the emission source or subject to long-range transport via air masses. In most cases (except Hg and to some extent Se), emission of heavy metals occurs on particles. The size of emitted particles containing heavy metals, as well as the temperature and speed of exhaust gases and the height of the emission source, are the major factors influencing the relative proportion of metals transported locally over long distances.

During the past two decades, there have been several applications of various long-range transport models to study the origin of atmospheric heavy metals measured in large industrial and residential areas, such as the entire European continent, and in remote locations, such as the northern parts of Scandinavia and some parts of the Arctic. One focus has been assessing the degree to which atmospheric deposition of these pollutants contributes to the contamination of certain regions, including the North Sea, the Baltic Sea, and the Arctic. Despite intense interest in using transport models in relation to PARCOM and HELCOM activities (studies of the transport and deposition of pollutants to the North Sea and the Baltic Sea, respectively) and other projects concerning deposition over the entire European continent and over the drainage basins of the rivers Rhine and Elbe, only a few operational models validated through comparison with experimental data are currently available in Europe. These models have been designed to simulate long-range and long-term transport of inert, particle-bound heavy metals and include 1) a combined trajectory-climatologic approach (Alcamo *et al.* 1992), 2) statistical approaches of a Gaussian plume model and a trajectory model (van Jaarsveld *et al.* 1986), 3) an EMEP-type Lagrangian trajectory model (Petersen *et al.* 1989, Petersen and Kruger 1992), and 4) a Eulerian model (Galperin *et al.* 1994a).

7.3.1.1. Dispersion models to study the impact of sources outside the Arctic

The first model of long-range transport of heavy metals in Europe (Pacyna *et al.* 1984) was presented soon after the first European emission survey was completed. The model was similar to that used in the OECD study on long-range transport of sulfur compounds (OECD 1979). The model results were used to verify emission data through a comparison of measured and calculated concentrations of heavy metals in ambient air. Later, the model was modified and used to study the long-range transport of heavy metals to

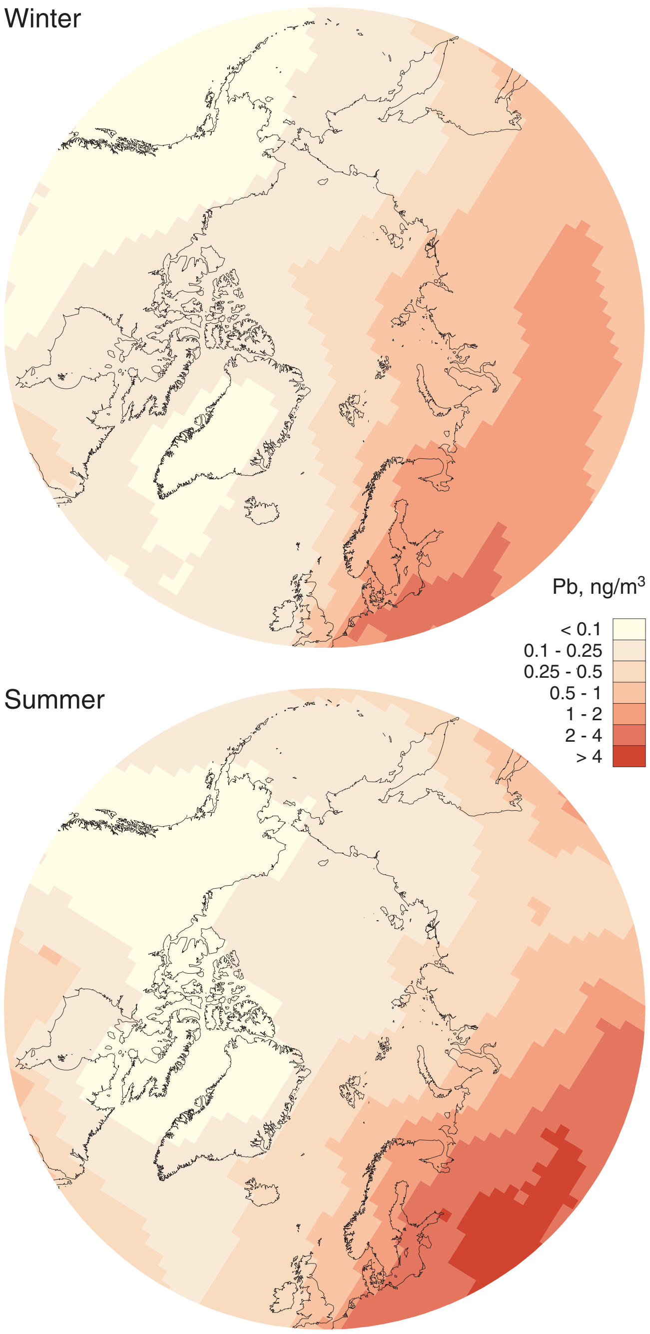


Figure 7·18. Averaged upper (3000 m) air concentrations of Pb in winter (December-February) and summer (June-August) as modeled by the updated (1996) hemispheric EMEP transport model in a simulation for the reference year 1988. The gridded Pb emissions data used in the model simulation are shown in Figure 7·5. (Source of data: Norwegian Meteorological Institute, Leonor Tarrasón pers. comm.).

remote sites in Scandinavia (Pacyna *et al.* 1989) and finally to the Norwegian Arctic. It was concluded that the emission estimates for As, Sb, V, and Zn from anthropogenic sources in Europe (as applied in the model) can be related to the concentrations measured in the air over Scandinavia and the Arctic, and that the agreement between measured and modeled concentrations was within a factor of two.

A Eulerian model has been developed at the Meteorological Synthesizing Centre - East (MSC-E) of EMEP (Galperin *et al.* 1994a). The model is used to calculate source-receptor relationships for Pb, Cd, As, and Zn in the UN ECE region. An important improvement in this model was the inclusion of a particle size spectrum to the parameterization of the model. Particle size greatly affects the removal of heavy metals from the atmosphere. This model was then applied to study long-range transport of Pb, Cd, Zn, and As from sources in Eurasia and North America to the Arctic (Galperin *et al.* 1994b). It was concluded that the simulation of the deposition of studied metals in the Arctic demonstrated an appreciable impact of emissions from sources in these regions.

The impact of emissions from Eurasian sources on the quality of air in Barrow, Alaska during 1985-1992 was also studied using an isentropic air trajectory model, developed by Harris and Kahl (1994). During the Arctic haze season (wintertime), trajectories suggested that the transport of pollution from north central Russia occurs near the surface, whereas that from northern Europe occurs at higher altitudes.

In a more quantitative approach, a long-range aerial transport Lagrangian model of Olson and Oikawa (1989) was used to compute the annual flux of Sb, As, Cd, Pb, Zn, and V into the Arctic from Eurasia (Akeredolu *et al.* 1994). Comparison of the model-predicted concentrations of heavy metals with a set of limited observations at existing sampling stations close to the Arctic Circle showed agreement within a factor of 2 to 3.

Recently, a model simulation of Pb transport to the Arctic has been performed for AMAP on the basis of the global emission inventory mentioned earlier (Figure 7·5, Tarrason 1996). Air concentrations (Figure 7·18) and deposition (Figure 7·19) of this metal within the Arctic were estimated for the reference year 1988. In general, the results of this simulation confirm the impact of emission sources in Eurasia on the contamination of Arctic air.

The models used in Europe, their attributes, and main area of application have been reviewed by Petersen (1993). A similar comparison for the North American models has been made by Voldner (in Pacyna *et al.* 1993a).

In summary, various modeling studies on the origin of Arctic air pollution support the conclusion that during win-

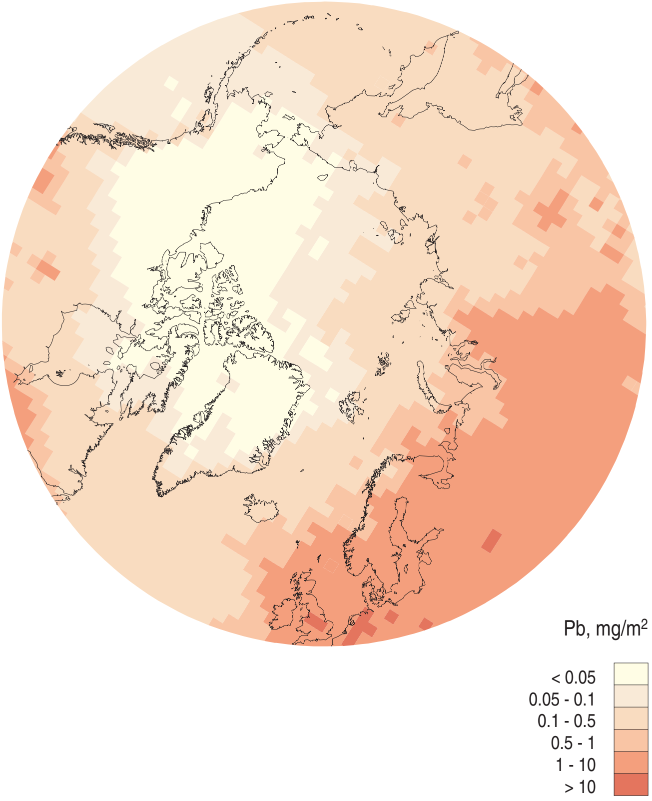


Figure 7·19. Lead deposition as modeled by the updated (1996) hemispheric EMEP transport model in a simulation for the reference year 1988. The gridded Pb emissions data used in the model simulation are shown in Figure 7·5. (Source of data: Norwegian Meteorological Institute, Leonor Tarrasón pers. comm.).

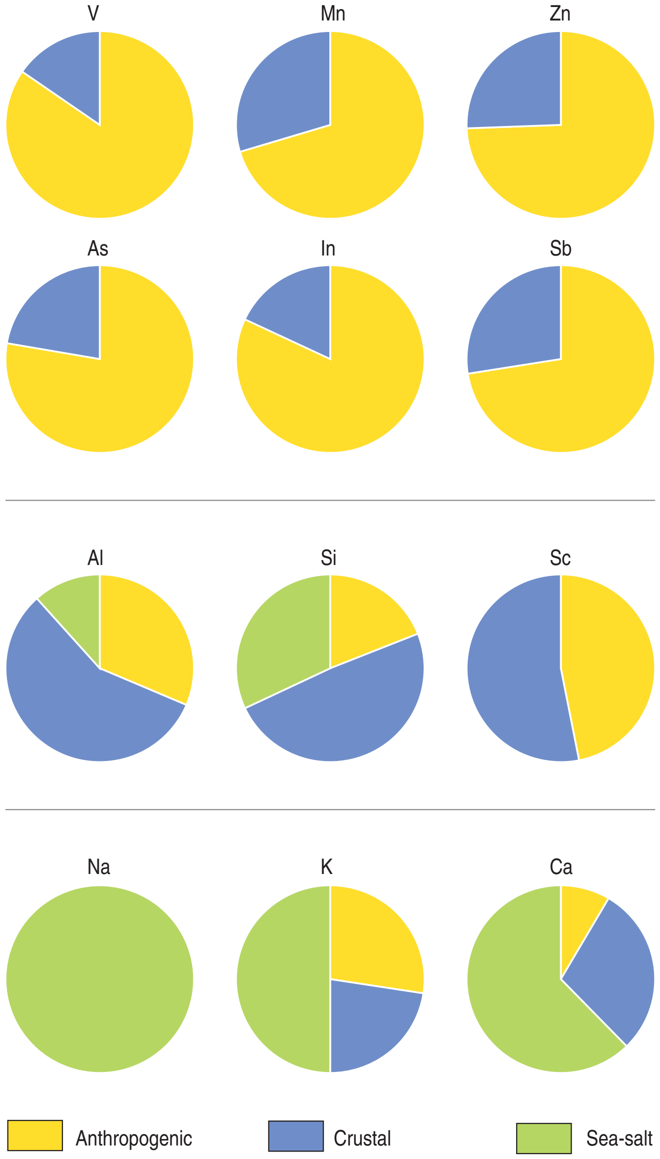


Figure 7·20. Average contributions from various sources to the atmospheric concentrations of elements in winter air samples at Ny-Ålesund, Svalbard (after Maenhaut *et al.* 1989). The source contributions are expressed as the percentage of the average air concentrations; these did not always sum to 100% due to inaccuracy of methods.

ter, 60 to 70% of heavy metal input is transported to the region from sources in the former Soviet Union, with the rest from Europe and North America (Raatz 1984, Lowenthal and Rahn 1985, Maenhaut *et al.* 1989). In summer, the contribution from sources in Europe can be as high as 75%. Up to 6% of the total emissions of As, Cd, Pb, Zn, V, and Sb in all of Eurasia is deposited in the Arctic (Akeredolu *et al.* 1994).

It is important to note that the meteorological conditions in the High Arctic during winter do not favor the deposition of pollutants. Relative to other global locations, there are fewer and smaller cloud droplets or ice crystals for particles to collide with or to diffuse and attach to in the polar air. The Arctic air mass has great dynamic stability and is characterized by laminar flow. Washout of pollution is low throughout most of the polar air mass. Stable stratification in winter prevents strong vertical mixing (Shaw 1986, Benson 1986). Under such conditions, pollutants (including heavy metals) transported to the Arctic can be trapped for several weeks. Indeed, the episode of 'mega-haze' in the Arctic in the late winter of 1986 seems to confirm this hypothesis (Li and Winchester 1989). On some occasions, however, the episodes persist no longer than a few hours. Thus, is the deposition process efficient enough to retain small particles with heavy metals within the Arctic region, or are they carried out of the region with air masses? Although the answer to this question has is of fundamental importance when assessing the impact of industrial emissions on the quality of the environment in the Arctic region, there are only a few measurements of wet deposition of heavy metals and other pollutants, and even fewer of dry deposition. Generally, these processes in the Arctic are poorly understood.

In general, models of long-range transport, deposition, and modification of heavy metals in the Arctic are incomplete at the present. Bowling and Shaw (1992), for example, have indicated that when physically reasonable constraints are placed on sources and sinks of heat and water mass, and on the relative humidity of near-surface air, it becomes apparent that the assumption of isentropic flow without precipitation is incompatible with observed water mixing ratios in layers of particles in the Arctic.

7.3.1.2. Receptor models to study the impact of sources outside the Arctic

A number of statistical source-receptor techniques have been used to study the contribution of various sources or even source regions to the contamination of the Arctic air by heavy metals. Maenhaut *et al.* (1989) applied the absolute principal component analysis (APCA) and the chemical mass balance method (CMB) to assess this contribution. The APCA indicated that there were three source components contributing to the chemical composition of measured aerosols: 1) anthropogenic activity, 2) soil dust, and 3) sea-salt. The average contributions of these components to the atmospheric concentrations of several heavy metals in winter aerosols measured in the Norwegian Arctic is given in Figure 7·20 (after Maenhaut *et al.* 1989). The APCA does not, however, produce a fine resolution of the contributions of emissions from various source regions. To attempt this, the CMB analysis of source apportionment was performed using a set of elemental signatures obtained by Lowenthal and Rahn (1985). Average regional apportionments of metals from Europe and the Asian part of Russia to aerosols in the Norwegian Arctic are shown in Figure 7·21 (after Maenhaut *et al.* 1989).

The North American path has been described in source-receptor studies by Maenhaut *et al.* (1989). There are also

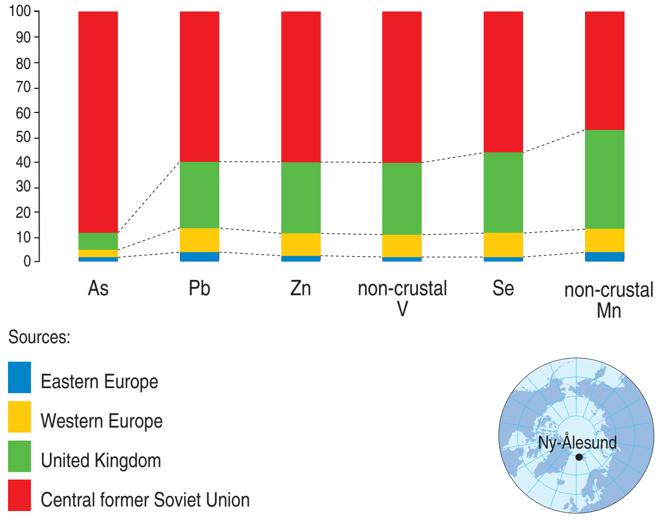


Figure 7·21. Average regional source apportionment of metals in winter aerosol at Ny-Ålesund, Svalbard from Europe and the central part of the former Soviet Union (including the Urals and Norilsk). (After Maenhaut *et al.* 1989).

flows of air masses from the northern Pacific to Alaska (Shaw 1991b). This air system contains mean concentrations of heavy metals two to three times lower than the air mass system associated with lobes of the polar air mass that influence the area of central Alaska.

Information on the origin of heavy metals and other pollutants in the Arctic as described above has been confirmed through the application of potential source contribution function (PSCF) to the sets of data from the Norwegian Arctic (Li *et al.* 1996).

Other methods have been applied to study the origin of pollution in Arctic air and include:

1. A tracer system based on ratios between air concentrations of various trace elements (Lowenthal and Rahn 1985);
2. Factor analysis techniques (Barrie and Hoff 1985);
3. Individual particle analysis (Anderson *et al.* 1992, Sheridan 1989); and
4. Analyzes using information on isotope ratios in the Arctic air (Sturges and Barrie 1989), halogens (Sturges and Barrie 1988), graphitic carbon (Rosen *et al.* 1981, Heintzenberg 1982), and organic compounds (Pacyna and Oehme 1988).

These studies have proved that the Eurasian source regions contribute most of the contamination measured in the Arctic, particularly for heavy metals.

7.3.1.3. Modeling the dispersion of emissions from sources in the Arctic

A Gaussian model has also been used to study the local impact of emissions from sources within the Arctic. Baklanov and Rodjushkina (1993) studied the impact of emissions from the Severonikel smelter complex on the Kola Peninsula on the surrounding environment. The modeled average concentrations of Cu and Ni were higher than the measured values by factors of 2 and 8, respectively. This rather high disagreement can be partly explained by overestimation of emissions. As mentioned earlier, significant variability exists among emission estimates for this region.

7.3.2. Rivers

As noted in chapter 3, rivers are one of the major pathways of contaminants 1) to the Arctic region from the lower latitudes and 2) to the Arctic marine ecosystem from the Arctic terrestrial ecosystem. The speciation of heavy metals in rivers is controlled by the physico-chemical properties of individual heavy metals and by river chemical conditions, particularly the presence of complexing ligands such as natural organic compounds. Concentrations of natural organic matter in the Mackenzie River and in Siberian rivers are similar but much lower than those of the Russian Euroarctic rivers (North Dvina and Pechora). Though water discharges by the Siberian rivers are much higher than those of the North American rivers, sediment transport by Siberian rivers is significantly lower (except the Yana, Indigirka, and Kolyma rivers in Eastern Siberia). For example, the Mackenzie River sediment load is seven times higher than that of the Yenisey River.

Sedimentation processes play an important role in the fate of heavy metals in river transport, particularly if the rivers contain reservoirs or pass through lakes (e.g., Mackenzie River-Great Slave Lake, Pyasina River-Pyasino Lake). In this case, lakes provide a sink for heavy metals transported by river flow. This situation is especially important in the case of Pyasino Lake, where heavy metals discharged with waste waters from the Norilsk smelter tend to accumulate.

For pollution sources situated in the upper reaches of large Arctic rivers (e.g. the Urals industrial complex in the upper reaches of the Ob River basin) the transport of particle-bound heavy metals to the Arctic is either reduced or eliminated by deposition occurring along the river bed.

Rivers are the largest source of freshwater to the Arctic. North-flowing rivers drain an area of 10 000 000 km^2 of northern Asia, northern Europe, and North America as far south as latitude 50°N (Barrie *et al.* 1992). Most major north-flowing rivers are found in Asia and Europe with only one, the Mackenzie, in North America. Cattle (1985) estimated that the total annual river discharge into the central Arctic Basin ranges from 2700 to 5000 km^3 and varies from year to year. Three Russian rivers (the Yenisey, Lena, and Ob) account for almost 70% of the total water input to the Arctic Ocean. Very few mass balance studies between the different pathways are available. The relative importance of the different pathways depends on the metals considered and on whether figures for emissions or actual deposited values are used. In addition, the season and the distance from the sources will strongly influence the estimates. A comparison of the river fluxes of metals to the Arctic Ocean, presented in Table 7·18, with atmospheric emission data from Akeredolu *et al.* (1994) shows great variation in the relative significance of these pathways. For metals such as Cd and Pb, riverine transport toward the Arctic is approximately half as great as the atmospheric contribution (Cd: 47 tonnes in atmosphere, 25.6 tonnes in river; Pb: 2400 tonnes in atmosphere, 859 tonnes in river). Other metals show the reverse: for example, rivers carry five times more Zn than atmospheric emissions (1350 tonnes in atmosphere, 6660 tonnes in river).

Although the amount of freshwater flowing into the Arctic Ocean equals only about 1% of the water masses entering through Fram Strait, rivers have an important effect on the oceanography of the region. In draining vast areas of land, they may also carry significant amounts of contaminants to the ocean surface layer.

The flow of freshwater to the Arctic is strongly seasonal. For the Ob and Yenisey, the effects of spring thaw are evident in May, with a sudden increase to maximum flow in June. The relatively high monthly flow rates and lower but more sustained flow of the Mackenzie, peaking in July, reflect the moderating effects of the large lakes in the drainage system.

7.3.3. Estuaries

As noted in chapter 3, estuarine zones function as filters for terrigenous matter entering the ocean (marginal filter) and combine the characteristics of both the catchment areas and the receiving marine basin. To assess a net flux of trace elements to the open ocean, the processes that control their transport through an estuary must be understood.

The Ob, Yenisey, and Lena estuaries are salt wedge estuaries with minimal (generally < 1 m) tidal range, comparatively short freshwater residence-time, and high seasonal variability and biological productivity. Such estuaries provide favorable conditions for highly variable chemical and biochemical reactions which strongly influence the behavior of many elements. Metals also exhibit individual tendencies. This is illustrated by the distribution of concentrations of dissolved Cd, Cu, Ni, Pb, and Zn in the Lena mixing zone, based on data obtained during a September 1989 expedition (Martin *et al.* 1993). Cu, Ni, and Zn concentrations changed insignificantly with salinity. Lead concentrations showed strong salinity related variations and a tendency to be removed from solution, whereas Cd concentrations increased with increasing salinity.

More detailed data on the fate of dissolved metals in the Lena and Yenisey estuaries, which confirmed the conclusions of the previous studies, were obtained during an international expedition to the Kara Sea in autumn 1993 (Lisitsin and Vinogradov 1994, Kravtsov *et al.* 1994, Dai and Martin 1995). In these studies it was shown that colloids controlled the fate of dissolved metals in these estuaries as well as their net river flux to the open part of the Kara Sea.

The behavior of Fe (removal) and Cd (desorption) was consistent for the estuarine zones of all three largest Siberian rivers. Behavior of the other metals was specific for each estuary. Changes of metal speciation in estuaries drastically affects their fluxes in dissolved and suspended forms. For Fe, Pb, and Zn for example, only 10-20% of total river flux reaches the open ocean. In the case of Cu and Ni, the net flux is approximately the same as the total river input. The flux of dissolved Cd to the sea tends to increase due to the desorption of Cd from suspended particles.

7.3.4. Oceans

Contaminants, including heavy metals in water or ice in the Arctic marine environment, are transported directly by ocean currents. Ocean circulation is driven by a combination of forces. A particular force can dominate in a particular geographical area; for example, tidal forces are dominant in many channels of the Arctic Archipelago whereas wind stress is most important for surface currents in the Canadian Basin (Barrie *et al.* 1992).

The major water exchange between the Arctic Ocean and other oceans occurs through Fram Strait. The West Spitsbergen Current flows northward off the west coast of Spitsbergen, transporting Atlantic water from the Nordic seas into the Arctic Ocean (Swift and Aagaard 1981). Relative to Arctic Ocean water, the inflowing Atlantic water can be enriched with heavy metals, such as Pb, Cu, Ni, Hg, As, V, Cd, and Cr, as seen from measurements carried out within the Oslo and Paris (OSPAR) Convention on the reduction of the contamination of the North Sea (ATMOS 1993).

Approximately half of the West Spitsbergen Current flows across the strait and recirculates southward at the surface as modified (cooled) Atlantic water. The remainder flows northward, divides into various branches, cools on contact with the polar ice pack, and subsides beneath the Arctic surface water forming the deep ocean layer. Beneath the Western Spitsbergen Current lies northward-flowing bottom water. Together, these currents make up to 78% of the inflow to the Arctic Basin. On the western side of the Fram Strait, the East Greenland Current flows southward at the surface carrying sea ice to the North Atlantic. Beneath the East Greenland Current are the southward-flowing modified Atlantic water and bottom water. Together, these currents constitute about 83% of the outflow from the Polar Basin (Barrie *et al.* 1992). Exchange with the Pacific Ocean across the Bering Strait is one-way into the Arctic, contributing about 20% of the total inflow. Again, large temporal and spatial variabilities occur in this flow (Coachman and Aagaard 1981).

Within the Arctic Ocean itself, the main surface circulation features are the clockwise circulation of the Beaufort Gyre and the transpolar drift. The transpolar drift flows from Siberia, across the pole, then southward to exit as the East Greenland Current. This drift can be contaminated by heavy metals released from discharge sources in Siberia. Within the Arctic Archipelago, the net transport is from the Arctic Ocean to Baffin Bay.

7.3.5. Ice

Various pollutants, including heavy metals, deposited on the sea ice from the atmosphere can be transported long distances in the ice or in blowing snow and then released to the atmosphere or the ocean during the melt processes. In addition, there is also a vertical transport, because growing sea ice rejects salt from the ice matrix in the form of dense brine which drains into the surface waters beneath. This vertical convection thus enhances exchange between the surface and deep ocean compartments (Gade *et al.* 1974).

Spring breakup of ice becomes evident from late April to mid-May as flaw leads and polynyas expand. Under the influence of river runoff, landfast ice begins to deteriorate in June. In late summer, sea ice forms as the surface water cools to its freezing point.

According to Russian observations, Pb, Fe, Cu, and Cd typically are elevated in sea ice/snow compared with surface ocean water (Melnikov 1991), perhaps due to atmospheric deposition and infreezing of particulate matter. Under-ice observations in the Laptev Sea indicate that starting in March, Pb, Fe, and Cu are released from the ice, apparently due to brine migration, resulting in concentrations in the surface water that are two to three times higher than the initial values (Pb increased from 0.1 to >0.2 µg/kg, Fe from <0.5 to >1.0 µg/kg, and Cu from <0.1 to 0.2 µg/kg) (Melnikov 1991).

Campbell and Yeats (1982) concluded that, during melting, ice contributed Fe, Cu, and Cd to surface waters in northwest Baffin Bay. In their study, sea ice with notably high particulate concentrations (4.75 mg/L) collected off Bylot Island yielded concentrations of these metals significantly in excess of the levels observed in surface waters, as shown in Table 7·3.

Table 7·3. Concentrations of metals in ice and water, Baffin Bay (Campbell and Yeats 1982).

	Concentrations, in ice and water, µg/kg				
	Fe	Ni	Cu	Cd	Hg
Ice	1.32-1.47	25.28-59.90	0.37	8.22-7.29	0.31
Water	0.10-0.56	0.60-3.07	0.17-0.28	0.18-0.60	0.020-0.075

The polar ice moves continuously, under the influence of the wind and ocean currents. The polar pack in the Beaufort Sea drifts in a clockwise direction, whereas in the rest of the Arctic Ocean the transpolar drift carries ice from the Siberian coast toward Fram Strait where it exits the Arctic Ocean (Gordienko 1958).

7.4. Toxicological characteristics

7.4.1. Toxicokinetics: general principles

Heavy metals occur naturally in the Earth's crust and are ubiquitous in the environment. Some heavy metals (e.g., Zn, Cu, Se) are biologically essential, playing an integral role in enzyme structure or as catalytic co-factors. Over 50 enzymes require Zn as a co-factor for example, and no life can exist without it. Other metals, such as Pb, Cd, and Hg, do not appear to have any biological function in organisms and are accordingly termed non-essential metals. Only trace amounts of essential heavy metals are required physiologically; in increased doses, these metals become toxic, as do non-essential metals. Animals have developed a variety of homeostatic mechanisms for the regulation of levels of essential metals (Clarkson 1986), which usually involve the control of gastrointestinal absorption. Toxic effects are therefore less likely to be experienced with these metals than is the case with non-essential heavy metals.

The three non-essential heavy metals that have been identified as being of greatest concern in the Arctic, because of their consequences for human health (see chapter 12) are Pb, Cd, and Hg. Selenium has also been included for discussion because it is essential in enzymes and proteins, and can reduce the toxic effects of some heavy metals (e.g., Cd, Hg, and As – see below). Similarly, the well-defined toxic effects of Se can be reduced by some of these metals. In contrast to most organic chemicals (see chapter 6) that can be eliminated from body tissues via metabolic degradation, elemental metals cannot be degraded. The only mechanism by which metals can be removed from the body is by excretion. Heavy metals, therefore, have the potential to accumulate in the body, leading to acute and chronic effects.

The high reactivity of heavy metals has a direct bearing on their bioavailability, distribution, and ultimate toxicity. The major factor underlying the biochemical properties of heavy metals with regard to their transportation, distribution, and elimination in organisms is their high affinity to sulfur and sulfhydryl groups of proteins. Sulfhydryl groups are ubiquitous in organisms, occurring on plasma proteins, membrane proteins, and enzymes.

Factors influencing the toxicity of heavy metals in solution are summarized in the following scheme (after Bryan 1976):

- Form of metal in water
  - Inorganic
  - Organic
    - Soluble
      - Ion
      - Complex ion
      - Chelate ion
      - Molecule
    - Particulate
      - Colloidal
      - Precipitated
      - Adsorbed
- Presence of other metals or poisons
  - Joint action
  - No interaction
  - Antagonism
    - More-than additive
    - Additive
    - Less-than-additive
- Factors influencing physiology of organisms and possibly form of metal in water
  - Temperature
  - Ph
  - Dissolved oxygen
  - Light
  - Salinity
- Condition of organism
  - Stage in life history (egg, larva, etc.)
  - Changes in life cycle (e.g. molting, reproduction)
  - Age and size
  - Sex
  - Starvation
  - Activity
  - Additional protection (e.g. shell)
  - Adaptation to metals
- Behavioral response
  - Altered behavior

7.4.2. Uptake

Uptake of heavy metals is directly related to bioavailability. Factors affecting bioavailability include both those specific to the organism (age, feeding habits, and nutritional status) and those specific to the metal (chemical form, particle size) (Beck *et al.* 1995). In general, the uptake of metals by biota involves crossing a biological membrane. This is most readily achieved by divalent cations. Consequently, any of the physico-chemical variables (e.g., pH, redox potential, ionic strength, presence/absence of dissolved organic matter and other complexing ligands, suspended particulate matter, temperature) influencing the concentration of divalent cations in solution will affect bioavailability. In animals, the major routes of heavy metal uptake in order of efficiency are ingestion, inhalation, and dermal absorption (see Table 7·5).

Heavy metals tend to accumulate in so-called 'storage' compartments as long as the rate of uptake exceeds the rate of excretion. Because rates of excretion or elimination of metals are often different in different parts of the body, these storage compartments represent the lumping together of various parts of the body where rates of specific metal elimination are the same. Accumulation often continues throughout life, or in some cases, at least until metal toxicity is experienced. Pb, for example, is typically redistributed from various organs into two tissue compartments: an exchangeable compartment (blood and soft tissue) and a storage compartment (bone). While accumulation occurs in both compartments, Pb levels tend to stabilize at a maximum level in the exchangeable compartment with time, or in some cases even to decrease. Lead levels in bone, however, tend to increase throughout the lifetime of an organism.

7.4.2.1. Bioaccumulation and biomagnification: general principles

The ability of organisms to accumulate heavy metals to concentrations of one or more orders of magnitude greater than concentrations in their food usually represents the major pathway leading to chronic toxicity. However, accumulated metal may be present in tissues in a relatively non-toxic or inert form even if it was originally toxic, because the toxicity of the metal can be modified through interactions between metals or through biotransformation by the organism. In contrast to POPs which are highly lipophilic and therefore accumulate primarily in body lipids, heavy metals are preferentially accumulated in proteinaceous tissues. The degree to which metals are accumulated varies greatly depending both on the metals involved and on the organ or tissue. The concepts of bioaccumulation and biomagnification used in studies of the environmental distribution of heavy metals are slightly different than those used for other persistent pollutants and therefore need to be clarified to avoid confusion.

The term bioaccumulation refers to the net accumulation of metals within an organism from both biotic (other organisms) and abiotic (soil, air, and water) sources. Bioaccumulation (or the bioaccumulation factor, BAF) is expressed as the relationship between the concentration of a metal in the tissue of an organism and the concentration of the metal in the air (terrestrial) or water (aquatic). This unitless ratio represents the steady state between the rate of uptake and the rate of loss of metal by the organism, and is largely dependent on the bioavailability of different metals to an organism, and the tissue type selected for measurement. Bioaccumulation can vary naturally between individual organisms and within populations just as metal concentrations can vary among individuals of the same species; it depends partly on an organism's food habits and overall physiological condition.

Generally speaking, the BAF is more appropriately used to describe accumulation in the aquatic environment, as the uptake of heavy metals by higher organisms in the terrestrial environment occurs primarily via ingestion. Heavy metal concentrations in the surrounding terrestrial environment (soil and air) play a less important role in bioaccumulation than is the case in water. A concept closely related to bioaccumulation, 'bioconcentration' (or bioconcentration factor, BCF) describes the concentration of a metal, derived only from water (through gills or epithelial tissue), in a tissue relative to the concentration of the metal in the water (Macek *et al.* 1979). Bioconcentration can, realistically, only be measured in the laboratory under specific conditions. Although dermal absorption may play an important role in aquatic systems, most organisms (except phytoplankton) do not necessarily accumulate metals in tissues directly and solely from water.

The progressive bioaccumulation of heavy metals by successive trophic levels is termed biomagnification. The biomagnification factor (BMF) is generally expressed as the ratio of two heavy metal concentrations (C_n and C_{n+1}) present in the tissue at two trophic levels (n and n + 1). In particular, it can be described as the concentration ratio in a tissue of a predator organism to that in a prey organism (i.e., C_{n+1}/C_n). The BMF is not restricted to contaminant movement over a single trophic level, and can in fact refer to concentrations in organisms one or more trophic levels apart.

Although heavy metals have been found in most tissues of mammals, certain metals tend to accumulate preferentially in one or more tissues. For instance, in most mammals experiencing chronic exposure to Cd, concentrations occur primarily in the kidney. Only small, relatively insignificant concentrations are present in muscle. Similarly, Hg is accumulated largely in the liver, and Pb in bone. It is therefore important to compare metal concentrations from the same tissue compartment for both predator and prey in order to obtain consistent BMFs. Inconsistencies can arise in the comparison of biomagnification factors due to the selection of different tissues for contaminant measurements. The BMF for Cd in polar bears (*Ursus maritimus*) relative to seals is < 1 if based on liver tissue of both animals, but is 77 if based on blubber, the tissue (along with skin) preferentially consumed by bears (Muir *et al.* 1992).

7.4.2.2. **Terrestrial ecosystem: bioaccumulation/biomagnification**

Background levels of heavy metals in the soil vary regionally, and are primarily affected by the geology of local bedrock. The anthropogenic contribution to heavy metal contamination through long range atmospheric fallout (from precipitation and dry deposition) exceeds the natural component (from degassing of the earth's crust) (Pacyna 1994). In the 1950s and 1960s, the use of heavy metals in agricultural chemicals (the use of alkylmercury fungicides as seed dressings in particular) resulted in intoxication of the terrestrial food chain. Industrial sources of contamination (discussed in section 7.6.2.1), so-called 'hot spots,' are generally localized; deposition rates are markedly elevated nearest the source and decrease rapidly with distance.

Most heavy metals deposited in soil-water systems are rapidly bound by organic particulate matter. The availability of these metals to terrestrial microorganisms depends largely on the their oxidation state, the organic content of the soil, and pH. In the Arctic, soil acidification due to acid rain is an important factor contributing to heavy metal bioaccumulation, as most metals are more readily accumulated as divalent cations, and acidification generally enhances the concentration of divalent cations in solution. Mercury behaves in the opposite way: decreasing pH enhances the absorption of Hg onto organic matter (Bergkvist 1986, Lodenius 1990). At pH 4.8 Bergholm *et al.* (1985) found the following binding strength sequence based on the solubility of metals: Fe > Al > Cu > Pb > Zn > Mn = Cd. Similarly, Andersson *et al.* (1991) suggested the following sequence in the pH range 3.5-5.0: Hg > Pb > Cu > Zn > Cd > Mn > Ca > Mg, with Hg being bound most strongly.

Terrestrial lichens, which lack root systems, take up metals directly from the air. While absorbing a small proportion of heavy metals through their leaves, vascular plants generally absorb metal cations through their roots from soil water. Fungi accumulate metals differently than vascular plants, showing some degree of selectivity. Most macrofungi species contain significantly more Zn and Cd than plants, and certain species strongly accumulate Hg and Cd (Lodenius 1990). Direct uptake of metals from the air through the lungs is of no significance in the Arctic environment, except in the immediate vicinity of major pollution sources.

7.4.2.3. **Freshwater ecosystem: bioaccumulation/biomagnification**

The main sources of heavy metal contamination in the freshwater environment include atmospheric fallout, smelting processes, mining activities, and runoff from terrestrial drainage systems. Aquatic systems differ from terrestrial ones in the proportion of metals they receive from natural sources (background concentration) versus anthropogenic sources.

The acidification of terrestrial soils by acid rain increases the solubility of certain metal ions including Mn, Zn, Pb, and Cd, thereby increasing leaching into surface waters. In a study affecting heavy metal bioaccumulation in pristine headwater lakes, Iivonen (1992) concluded that the bioaccumulation of several metals, particularly Pb and Cd, was increased with increasing acidification of the water column. Furthermore, there was some evidence that the presence of organic matter reduced heavy metal bioaccumulation, probably due to complexation of heavy metals with dissolved humic matter. The stabilities of metal-humic complexes in natural waters are higher than those of most corresponding inorganic metal complexes.

Heavy metal uptake by aquatic biota begins with the lower trophic levels. Unicellular organisms (bacteria, algae) and aquatic plants take up metals exclusively from the water (pore water in the case of rooted plants) by passive surface absorption. Organisms at higher trophic levels generally accumulate metals via both passive absorption directly from the water and via food items. Zoobenthic and zooplanktonic organisms take up metals largely by feeding on algae, particulate organic bottom and suspended matter, and on other zoobenthic and zooplanktonic organisms. The extent of heavy metal uptake over body surfaces is not known, but is likely related to metal concentration and chemical speciation in the water column.

Carnivorous fish, at the highest trophic level in the freshwater ecosystem, absorb metals from the water partly through body surfaces, primarily the gills, and partly from food (zooplankton, zoobenthos, and smaller fish). The proportion of uptake attributable to either source varies according to the species of fish, the metal, and the concentrations in the water and in the food. In one species of fish, 90% of the inorganic Hg burden was absorbed directly from water (Jernelow 1972), whereas in another species, more than 80% of MeHg was absorbed from food (Hall *et al.* 1997).

7.4.2.4. **Marine ecosystem: bioaccumulation/biomagnification**

Heavy metals enter the marine environment through various pathways including atmospheric deposition, transport by rivers, leaching from agricultural land, weathering of rocks and soils, and from point sources (e.g., mining, spills, incineration of wastes on the open sea, and various accidental or deliberate discharges). Studies monitoring heavy metal contamination in European seas indicate that atmospheric pathways (atmospheric particles, gas exchange) play a dominant role in the transport of Pb, Cd, As, Cu, Zn, and other elements. In the North Sea and the Baltic Sea approximately 50% of these elements is deposited from the atmosphere (ATMOS 1993, Pacyna 1992). Inputs of metals to coastal waters via surface runoff from north-flowing rivers are important contributors to the sea in the summer, where they accumulate in sediments and in marine biota.

The food webs of the Arctic depend mainly on primary production by planktonic organisms and ice algae. This production is highly seasonal and depends on local conditions (such as temperature regime and nutrient enrichment). Phytoplankton blooms assimilate heavy metals from the water column and either transfer metals directly to higher trophic levels via grazing (e.g., zooplankton), or transport them to the ocean floor via processes of bodily absorption and sinking. Similarly, as upwelled water ages, organisms gradually enrich the water with organic compounds, some of which act as chelating agents binding to metals, thereafter transporting them to the sea floor as particulate matter. In this way most of the sediments in areas of upwelling are generally rich in metals.

Once on the bottom, metals may remain at the surface as a result of biological mixing (bioturbation) of the sediments by benthic invertebrates, which in turn support bottom feeding animals such as bearded seal (*Erignathus barbatus*), walrus (*Odobenus rosmarus*), and some whale species. Consequently, metals may remain in contact with benthic biota for longer periods of time than if buried. Dead detrital organic particles are mineralized and returned to the water column where they again become available for bioaccumulation.

The High Arctic food web can have as many as five steps, from algae through invertebrates to fishes, seals, and finally, polar bears (Welch *et al.* 1992). Arctic cod (*Arctogadus glacialis*) is a pivotal species in the food web, and the polar bear is the apex predator.

In Table 7·4, BMFs for some key species in the Greenlandic marine ecosystem have been listed. At the lower trophic end, it is obvious that Cd is not accumulated from parathemisto to Arctic cod, even when comparing liver of Arctic cod with the whole parathemisto. On the other hand, a clear accumulation takes place in birds, seals, and whales, where the BMF may be as high as 64.4. for muscle in fulmar (*Fulmarus glacialis*) versus Arctic cod. The BMF is also low between polar bears and ringed seals (*Phoca hispida*) because polar bears prefer to eat seal blubber, which is low in Cd (Muir *et al.* 1992, Dietz *et al.* 1995). Muir *et al.* (1992) also reported similar BMF for *Parathemisto libellula*/Arctic cod and Arctic cod/narwhal (*Monodon monoceros*) from the Canadian High Arctic.

Mercury increases from 6.4 to 15.6 times in liver and muscle of Arctic cod relative to whole parathemisto. The biomagnification is much higher between Arctic cod and ringed seal, where a BMF can reach 2823. In general the Se tends to biomagnify, but to a much lesser degree than does Hg.

Insufficient data were available to produce similar figures for Pb from the same areas. However, data from Muir *et al.* (1992) and the data compiled in this assessment indicate no biomagnification, i.e., BMF < 1.

Table 7·4. Biomagnification factors of cadmium, mercury, and selenium in the Greenlandic marine food chain.

		Biomagnification factor, wet weight basis		
Species/tissue	Area	Cd	Hg	Se
Parathemisto vs. Arctic cod				
Whole vs. liver	Avanersuaq	0.9	6.4	3.8
Whole vs. muscle	Avanersuaq	0.04	15.6	2.3
Blue mussel vs. common eider[a]				
Soft tissue vs. liver	Qeqertarsuaq	5.1	49.8	5.8
	Nanortalik	4.1	47.6	6.7
Soft tissue vs. kidney	Qeqertarsuaq	12.7	16.6	6.0
	Nanortalik	17.1	18.4	5.6
Soft tissue vs. muscle	Qeqertarsuaq	0.07	8.1	1.0
	Nanortalik	0.13	10.1	0.7
Arctic cod vs. black guillemot				
Liver vs. liver	Avanersuaq	4.1	46	3.0
	Ittoqqortoormiit	9.8	63	2.7
Muscle vs. muscle	Avanersuaq	6.1	5.6	1.7
	Ittoqqortoormiit	22	6.8	2.9
Arctic cod vs. Brünnichs guillemot				
Liver vs. liver	Avanersuaq	7.9	56	2.3
	Ittoqqortoormiit	24.6	118	3.1
Muscle vs. muscle	Avanersuaq	11	5.9	1.5
Arctic cod vs. northern fulmar				
Liver vs. liver	Avanersuaq	16.3	112	10.7
	Ittoqqortoormiit	18.9	170	5.9
Muscle vs. muscle	Avanersuaq	20.6	8.1	5.0
	Ittoqqortoormiit	64.4	10.2	12.4
Arctic cod vs. ringed seal[b]				
Liver vs. liver	Avanersuaq	19.9	115	1.8
	Nanortalik	20.8	636	3.5
	Kong O. Fjord	6.8	2823	10.9
	Ittoqqortoormiit	30.6	840	2.7
Arctic cod vs. narwhal[c]				
Liver vs. liver	Avanersuaq	13.3	178	3.5
Muscle vs. muscle	Avanersuaq	1.9	6.6	0.4
Ringed seal vs. polar bear[d]				
Liver vs. liver	Avanersuaq	0.13	8.9	3.6
	Ittoqqortoormiit	0.06	1.8	1.3
Kidney vs. kidney	Avanersuaq	0.29	14.4	3.3
	Ittoqqortoormiit	0.28	9.2	1.3
Muscle vs. muscle	Avanersuaq	0.13	0.28	1.8
	Ittoqqortoormiit	0.20	0.24	0.85

a. Means of two size groups (30-40 and 40-50 mm) of blue mussels from Nanortalik and only one size group (40-50 mm) from Qeqertarsuaq were used for calculations.
b. Means of 4-5 age groups of ringed seals were used for calculations.
c. Means of juvenile, mature and adult narwhals were used for calculations.
d. Means of 4-5 age groups of seals and 3 age groups of polar bear were used for calculations.

7.4.3. Transport, biotransformation, and distribution

Once absorbed, heavy metals are distributed in the body by the circulatory system, irrespective of their chemical form (Foulkes 1995). The fraction of transported metal absorbed by various organs and the fraction subsequently excreted vary greatly for each metal. The mechanisms involved in selective metal uptake in organs are not well understood (Foulkes 1995). Metals are initially distributed to a variety of organs and tissues, and subsequently redistributed to other tissues for storage and inactivation. For example, Cd in the blood (bound to high molecular weight proteins) initially accumulates in the liver (where efficient metallothionein synthesis probably takes place), and is subsequently redistributed to the kidney as a Cd-metallothionein complex. In the latter form, Cd can be readily filtered through renal glomerelli and reabsorbed in the tubuli where it binds to the renal tubular cells (Friberg *et al.* 1986, Norberg and Norberg 1987). A similar mechanism is found with Cu, which is initially bound to albumin and some amino acids and transported in the plasma to liver, brain, and muscle tissue. Under certain circumstances Cu can be remobilized by binding to a high molecular weight protein, ceruloplasmin, and released back into circulation to be transported to various other tissues.

The formation or breakdown of metal-carbon bonds or a change in the oxidation state of a metal within an organism (biotransformation) will affect the chemical activity of heavy

Table 7·5. Routes and efficiency of intake of Pb, Cd, Hg, and Se for different organisms. Information was compiled from reviews by WHO (1976, 1987, 1989a, 1989b, 1990, 1991, 1992a, 1992b), unless specified otherwise.

Metal	Organism	Route	Compound	Efficiency	Reference
Pb	Terrestrial plants	Roots		Readily absorbed	
		Shoots		Lesser extent	
	Birds	Intestine		Dependent on fiber in diet	
	Animals	Intestine	Airborne Pb	5-10%	1
	Humans	Lungs	Airborne Pb	30-50%	1
		Intestine		10%	1
Cd	Fish	Intestine		1%	
		Gills		0.1%	
	– Rapidly exchanging tissue	Intestine		1-2%	2
	Cows	Intestine		16%	2
	Rats, mice, and monkeys	Intestine		1-6%	3
	Animals	Lungs		7-40%	3
	Humans	Lungs		25-50%	3
		Intestine		5-7%	3
Hg	Shrimp	Intestine	Methyl Hg	70-80%	
		Intestine	Inorganic Hg	38%	
	Fish	Gills	methyl Hg	High. at low temp. wat. hardn.	
		Intestine	methyl Hg	Higher at higher temp.	
	Birds	Intestine		Organic > inorganic	
	Mammals	Intestine	Methyl Hg	>95%	
		Intestine	Inorganic Hg	>15%	
	Humans	Lungs	Hg vapor	80%	4
		Intestine	Metallic Hg	Poorly absorbed	4
		Skin		Likely; no qual. data	4
Se	Rats	Lungs	H_2Se0_4	94%	4
		Lungs	Elemental Se	57%	4
		Intestine	Selenite	92%	
		Intestine	Selenomethionine	91%	
		Intestine	Selenocystine	81%	
		Intestine	Se in rabbit kidney	87%	
		Intestine	Se in fish muscle	64%	
		Skin	Selenite	10%	
	Sheep and cows	Intestine		30-35%	4
	Mammals	Intestine		Reg. homeostatically	4
	Humans	Intestine	Selenite	60%	
		Intestine	Selenate	94%	
		Intestine	Selenomethionine	96%	

1. Tsuchiya (1986); 2. Paludan-Müller (1993); 3. Friberg *et al.* (1986); 4. Berlin (1986); 5. Högberg and Alexander (1986).

metal compounds, and therefore their toxicity. Changes in the oxidation state influence the ability of a metal to interact with various tissue ligands. Hg, for example, exists in three oxidation states: elemental (Hg(0)), the mercurous ion (Hg_2(II)), and the mercuric ion (Hg(II)). Hg(0) easily penetrates biological membranes because of its high lipid solubility. The mobility of Hg_2(II) and Hg(II) are much more restricted due to their tendency to form salts and their high affinity for sulfhydryl groups on proteins (Clarkson 1986).

Two biotransformation processes are important to the toxicity of heavy metals:

1. Methylation/demethylation of certain heavy metals and metalloids (e.g., As, Hg, and Se). In some cases cleavage may serve as a detoxification pathway, whereas in others the metabolite is the more toxic species. For example, methylation (the formation of metal-carbon bonds) of inorganic As and Se has been seen to lead to reduced toxicity in a number of animals and to form the basis for excretable metabolites (e.g., methylated selenides), while the reverse is true for Hg. In the case of methylmercury exposure, processes of demethylation are important for detoxification. In the case of Se, biotransformation in the liver seems to be the major mechanism by which homeostasis is maintained.
2. Formation of inert complexes also plays an important role in heavy metal detoxification. The biochemical relationship between metallothionein and such heavy metals as Cd, Zn, Cu, and Hg, for example, is fundamental to their toxicity. Metals binding with metallothionein form inert complexes which can be retained in body tissues (Clarkson 1986). Similarly, Se can reduce the toxicity of certain metals such as As, Cd, and Hg by forming inert compounds (Högberg and Alexander 1986) which usually accumulate within organisms.

The fact that many Arctic animals at high trophic levels (e.g., seals, whales, seabirds) are consumed by Arctic people has provided the impetus for analyzing tissue concentrations of various pollutants. Until now, measurements have been primarily carried out on those organs and tissues that are consumed. As a result of, the literature on metals and other contaminants in the tissues of Arctic animals may or may not represent targets of toxicological action in the animals themselves. Arctic fish and marine mammals accumulate relatively high levels of methylmercury in kidney, liver, and muscle. Although knowledge of these concentrations is important for assessing the potential exposure of human consumers to methylmercury, in terms of evaluating the toxic effect to mammals the most important concentration is that in the brain, which is generally not measured. A considerable amount of data has been collected for the critical organ of chronic Cd intoxication, the kidney. Even though Cd concentrations exceeding the threshold for toxic effects in various marine mammals have been measured, no clinical toxicological investigations have been carried out.

Table 7·6. Biological half-life of Pb, Cd, Hg, and Se for different organisms and tissue groups. Information was compiled from reviews by WHO (1976, 1987, 1989a, 1989b, 1990, 1991, 1992a, 1992b), unless specified otherwise.

Metal	Organism/tissue	Half-life (t½)	Ref.
Pb	Blue mussel	Trimethyl/ethyllead: 3-4 days	
	Insects:		
	– Fast body burden	7 days	
	– Slow body burden	22 days	
	Fish tissue:	9 days (received from water) 40 days (received from diet) Trimethyl/ethyllead: > 41 days	
	Humans:		
	– Rapidly exchanging tissue	35 days	1
	– Soft tissue compartment	40 days	1
	– Bone	20 years	1
Cd	Invertebrate tissue	2-53 days	
	Isopods and mollusks	Little or no excretion	
	Fish tissue	24-63 days	
	Mice and rats	200-700 days	
	Squirrel monkeys	> 2 years	
	Other mammals:		
	– Liver	10-50% of lifespan	
	– Slowest component	> 20% of lifespan	
	Humans:		
	– Kidney	10-30 years	2, 3
	– Liver	5-15 years	3
Hg	Leguminous plants	Up to 75% as elemental Hg	
	Invertebrates	$t^{1}/_{2}$ inorganic > $t^{1}/_{2}$ organic	
	Blue mussel	53-293 days	
	Fish:	Metallic and inorganic Hg: 45 days (received in water) Organic Hg: 323 days (received from diet) Inorganic Hg: 61.6 days (received from diet)	
	Mice and rats	12 days	
	Seals	Methyl Hg: 500 days	4
	Dolphins	Methyl Hg: 1000 days	4
	Humans:		
	– Phase I: blood	Metallic Hg: 2-4 days (received as vapor) Methyl Hg: 39-70 days	
	– Phase II	Metallic Hg: 15-58 days (received as vapor)	
	– Whole body	Methyl Hg: 52-93 days Inorganic Hg: 40 days	
	– Kidney	Inorganic Hg: 64 days	
	– Brain	19-26 days	
	– Hair	Bimodal: 35-100 days and 110-120 days	
Se	Rats:		
	– Kidney	38 days	
	– Muscle	74 days	
	– Whole body	55 days	
	Animals:		
	– Phase I	1-3 days	5
	– Phase II	30-70 days	5
	Humans:		
	– Phase I	1 day	5
	– Phase II	8-20 days	5
	– Phase III	65-116 days	5

1. Carlson 1986 cited in WHO 1989a; 2. Chowdhury and Chandra 1987; 3. Nordberg *et al.* 1985; 4. André *et al.* 1990; 5. Högberg and Alexander 1986.

7.4.4. **Excretion**

The most important excretory pathways for metal compounds in animals are gastrointestinal and renal. Gastrointestinal excretion includes excretion of metals into bile (and pancreatic fluid) and excretion by the intestinal mucosa. Excretion of most organic metal compounds occurs primarily by the bile, while inorganic compounds are excreted in the gastrointestinal tract. Considerable quantitative differences have been reported for different animal species. Metals excreted in the bile may be reabsorbed farther down the intestinal tract, and subsequently re-excreted into bile. Intestinal reabsorption can be prevented if excreted metals form bonds with non-absorbable compounds. In this way a net gastrointestinal excretion can occur, with the heavy metals being eliminated from the body with feces. The other mechanism of gastrointestinal excretion involves the removal of metal compounds in association with the rapid turnover of cells of the intestinal mucosa. Small quantities of certain heavy metals (e.g., inorganic Cd and Hg) are eliminated from the body when the cells are shed.

Urinary excretion is probably the second most important excretory route for animals. The glomerula membrane acts as a filter, allowing only those molecules with relatively low molecular weights to pass through into the renal tubules. Thus, metals bound to low-molecular weight proteins such as insulin or metallothionein may be cleared from the blood plasma in this way, although a proportion of this is subject to reabsorption. For example, Cd bound to metallothionein is efficiently reabsorbed, with only a small proportion being ultimately excreted with the urine.

7.4.5. **Uptake, accumulation, and loss in biota**

The following is a brief overview of the toxicokinetic characteristics of the four heavy metals identified as most significant to Arctic biota: Pb, Cd, Hg, and Se. The details presented below are derived primarily from data contained in the International Programme on Chemical Safety (IPCS) of the World Health Organization (WHO).

An important factor in the process of bioaccumulation (which also has implications for toxicity) is the biological half-time. This is the time it takes for the concentration of an absorbed pollutant to be reduced to half of its initial value, and consequently biological half-times generally refer to rates of elimination from specific storage compartments. Rates of excretion vary between metals and their compounds, and may have a major influence on chronic toxicity. Most of the data on the half-time of many heavy metals have been obtained from actual measurements of elimination rates following exposure.

A summary of the primary routes of uptake and biological half-times for Pb, Cd, Hg, and Se in various organisms are given in Tables 7·5 and 7·6, respectively. Invertebrates and fish take up only a small proportion (0.1% for Cd) of the metals in the water through the gills. Intestinal intake by invertebrates and fish is larger; the most efficient intake is for methylmercury (70-80%) and a much smaller uptake for Cd (1%). Excretion by invertebrates and fish compared with higher trophic levels is rather fast, with biological half-times of 3-40, 2-63, and 53-323 days for Pb, Cd, and Hg, respectively. This means that the major route of metals is through food, where the levels also are higher than in the surrounding water.

For higher organisms, metals are efficiently (7-94%) absorbed through the lungs, with the degree of absorption varying by animal species, metal, and chemical form of the metal. Mercury vapor and H_2Se0_4 are taken up most efficiently (80-94%), but Pb and Cd readily cross the lung epithelium (7-50%). Air levels of these metals, however, are rather low in most Arctic areas, so this pathway is of only minor importance. The uptake through the intestine depends on the organism, metal type, and chemical form. Only 1-16% of Pb and Cd are taken up by various species, whereas 57-95% of Hg and Se are absorbed through the intestine. Methylmercury is taken up more than six times as efficiently as inorganic Hg. Lead and Se are readily excreted, which results in moderate levels of these metals in internal organs. Lead, however, is deposited in bone, where its half-time can be up to 20 years. Cadmium is excreted slowly once it is taken up by organisms, with a half-time of 10-50% of the organism's life span (up to 30 years), whereas Hg has intermediate half-time (12-1000 days). Differences in half-time between the different species partly explain the generally higher observed metal levels in longer-lived species. The different half-times also explain differences between the observed levels of the different metals in the various tissue compartments. One might speculate that the excretion of Hg in seals and whales is so inefficient because of the abundance of Se in the environment. Se detoxifies Hg by forming the biologically inert compound mercuric selenide, which is not destroyed by the proteolytic enzymes and is thus a terminal phase in the reactions of methylmercury detoxification in the liver (Martoja and Berry 1980, cited in Andre *et al.* 1990).

7.4.5.1. **Lead**

Lead in the environment is strongly absorbed onto sediment and soil particles, and is therefore largely biologically unavailable. Many of the inorganic salts formed (Pb oxides and sulfides) are not readily soluble in water and are sequestered in sediments. In aquatic systems, uptake is influenced by various environmental factors such as temperature, salinity, pH, and the presence of organic matter. Among all the chemical species, Pb(II) is considered to be most readily absorbed by biota.

It has been shown that in terrestrial plants, Pb is taken up through the roots and to a lesser extent through the shoots. Although there has been some evidence of translocation, it appears that this plays a very limited role in distribution. Some of the Pb entering plant root cells becomes incorporated in the cell wall. Studies of aquatic plants have shown that submerged species often retain greater concentrations of Pb than floating species, with both shoots and roots evidently playing an important role in absorption.

For many animals it is not known for certain whether Pb is absorbed through the skin or actually taken up via inhalation or contaminated food. Accumulation in mussels (*Mytilus edulis*) has been demonstrated to occur in all tissues, but highest concentrations are seen in the kidney. In fish, Pb accumulates primarily in the gill, liver, and kidney, though it is not known whether accumulation in the gills represents uptake into the tissue or absorption onto exterior surfaces. Birds dosed with lead shot show signs of tissue accumulation in liver, muscle, and bone, and appear to be influenced by the amount of fiber in their diet. Lead also accumulates in eggs and embryos.

In humans, Pb is initially distributed to various organs and tissues and is gradually redistributed into two compartments: an exchangeable compartment, comprising blood and soft tissues, and a storage compartment, essentially bone. Lead levels in bone continue to increase throughout life while stabilizing, or in some cases decreasing, in soft tissues (Tsuchiya 1986, see chapter 12 for further details).

7.4.5.2. **Cadmium**

In aquatic systems, Cd is most readily absorbed by organisms directly from the water (primarily over gills and body surfaces) in its free ionic form (Cd(II)). Although many of its inorganic salts are soluble in water (e.g., acetate, chloride, and sulfate), they do not appear to be taken up. In fact, Cd toxicity is generally found to be lower in marine waters, and this has been attributed to chloride complexation. In marine waters, Cd is mainly present as soluble chloride complexes. Cadmium is also easily complexed with some organic compounds, which has an important bearing on its bioavailability. Increased organic content of the water generally decreases uptake and toxicity. Complexes with EDTA, NTA, and DPTA, for example, render Cd unavailable, and complexes with fulvic acids of low molecular weight and citric acid make Cd less available than in its free ionic form. The formation of hydrophobic complexes with compounds such as xanthates (used in mining for the enrichment of metals from sulfide ores) and dithiocarbamates, on the other hand, enhance uptake by acting as carriers over biological membranes, and are therefore of particular environmental concern. In terrestrial systems, the major source of exposure to animals is via inhalation and ingestion.

Uptake in plants depends on the availability of ionic Cd, which is affected by soil composition (i.e., particle size, mineralogy, organic matter content, and pH). Studies with lettuce and chard reveal dose-related uptake from soil into leaves. Red maple, white pine, and Norway spruce seedlings cultured in contaminated soil contained more Cd in the roots than in the leaves. Cadmium is usually found bound to the cell wall in roots.

Most Cd present in mollusks is taken up directly from the water. Uptake in oysters depends largely on the rate of water flow through the body, which is linked to feeding activity and water temperature. Accordingly, oysters can accumulate twice as much Cd in summer as in winter. Cadmium accumulates in both soft tissues and the shell, although localized accumulation can occur in the gills, heart, mantle, and muscles. Terrestrial pulmonate snails were found to absorb Cd from the chloride administered in their diet. In the terrestrial slug (*Arion ater*) most of the Cd retained in the body was found associated with the digestive gland. Specific metallothionein-like Cd binding proteins have been isolated from both marine and terrestrial gastropods. Differences in the ability of marine crustaceans to absorb Cd directly from the water are attributed to variations in cuticle permeability and lifestyle. Accumulation is greatest in the hepatopancreas, with lower concentrations occurring in the exoskeleton, muscle, and serum. There is no evidence that these organisms possess any mechanisms for Cd regulation. Isopods and mollusks retain Cd in their tissues with little or no excretion. Elimination half-lives in other species have been estimated to be between 2 and 53 days.

As with mollusks, metabolic rate and feeding are important factors affecting uptake of Cd in fish. This occurs primarily through gill lamellae, and is reduced with increasing water hardness (i.e., Ca(II) and Mg(II) concentrations). Two mechanisms are thought to be involved in this reduction in Cd uptake: 1) inhibition of Cd(II) uptake into gill tissue by Ca(II), and 2) physiological adaptive responses to increased Ca(II) concentrations. After exposure to Cd chloride in water, uptake is greatest in the gill, kidney, and liver. Fish exposed to Cd contaminated food accumulate Cd principally in the kidney, gut, and liver. One study suggests that retention is greater for Cd present in food than in water (1% compared with 0.1%). On a subcellular level, Cd has been found to be distributed both in the cytosol and in the nucleus bound to metallothioneins and other Cd-binding proteins rich in cysteine. Biological half-times for Cd in fish have been estimated to range between 24 and 63 days. Birds exposed to dietary levels of Cd chloride accumulate Cd in the liver and the kidney.

Animal data suggest that absorption over pulmonary surfaces is higher than gastrointestinal absorption. Depending on particle size, deposition, and solubility in biological fluids, up to 50% of inhaled Cd may be absorbed, compared with (on average) 5% of ingested Cd. The extent of dietary uptake is reportedly affected by nutritional status, ranging from 1 to

20%. The importance of the protein metallothionein for both the distribution and toxicity is described in section 12.2.3.2. Additional information can be found in Nordberg and Nordberg (1987). Essentially, once in the blood stream Cd is bound to high and low molecular weight proteins and distributed around the body. Binding to metallothioneins (low molecular weight components in the blood) invariably leads to transportation to the kidney, where its primary toxic effects are experienced. Excretion of Cd from muscle, kidney, liver, and the body overall is very slow. In rats, the half-time is 200-700 days, while in humans, half-times reported for muscle, kidney, and liver range from 10 to 30 years.

7.4.5.3. **Mercury**

Background concentrations of Hg are generally low, except in the immediate vicinity of mining sites, etc. Mercury occurs naturally as elemental Hg, and as organic and inorganic compounds (e.g., Hg vapor, Hg salts, short- and long-chain alkylmercury compounds). This speciation of Hg is of great importance in relation to uptake from soil and water, as the different chemical forms differ greatly in their physico-chemical properties. Much of the Hg in the environment is unavailable to organisms, as it is strongly bound to sediment or organic material. Inorganic forms can be methylated by microorganisms and transformed to methylmercury, which is much more readily taken up and accumulated in both aquatic and terrestrial organisms.

Uptake in aquatic plants increases with increasing concentrations of Hg salts, with greater levels occurring in roots than in shoots. However, some of this may be attributed to differences in surface absorption and not uptake *per se*. The possibility of some active uptake has been suggested for marine diatoms (e.g., *Chaetoceros costatum*). A similar pattern of absorption and distribution is found in studies with terrestrial plants. Soil type strongly influences the extent of uptake. For example, Hg absorption is inversely related to organic content. As with aquatic plants, the highest Hg concentrations occur in the roots, though there is some evidence of translocation to other parts of the plant including the leaves. It has been suggested that several leguminous plants appear to be able to eliminate Hg by a process termed 'biovolatilization' involving the loss of at least 75% of their tissue burden as elemental Hg vapor.

Most of the studies of Hg uptake in invertebrates do not differentiate between external absorption and actual uptake. In the case of methylmercury compounds, uptake usually correlates with surface absorption capacity. Blue mussels readily accumulate Hg, and have been found to have a biological half-life of 293 days following chronic exposure, and only 53 days following acute exposure. Approximately 75% of the Hg accumulated was immobilized in an inorganic form. Another study with a sediment-feeding bivalve (*Macoma balthica*) revealed that only 6% of the total Hg accumulated was present as methylmercury, although this percentage was somewhat lower than for mussels in the same area.

Assimilation of Hg in arthropods depends on both form (organic or inorganic) and source of exposure, and is highly variable between taxa (Zauke *et al.* 1996). When presented in the diet of shrimp (*Hyalella azteca*), net absorption of methylmercury was 70-80%, compared with 38% of inorganic Hg. Studies of the uptake of radioactive isotopes of Hg by grass shrimp (*Palaemonetes pugio*) indicate that 24 hours after exposure, methylated Hg is concentrated mainly in the ventral nerve chord, whereas assimilated inorganic Hg is found predominantly in the gills. However, after 72 hours the tissue distribution changed; concentration decreases in the ventral nerve chord corresponded with a general increase in other tissues (e.g., exoskeleton, foregut, and remaining tissues). Mercury levels in the gills remained relatively constant. Fiddler crabs (*Uca pugilator*) are able to transfer Hg from the gills to the hepatopancreas with increasing effectiveness when temperatures are increased. Few reports are available on methylation or excretion of Hg in arthropods. Blow flies (*Lucilia illustris*) reportedly tend to eliminate inorganic Hg compounds with greater ease than methylated forms.

Environmental variables such as temperature, pH, and redox potential are particularly important for Hg uptake in fish. Data on uptake suggest that absorption increases with higher temperatures and lower pH. Tissue concentrations of Hg increase with age for both marine and freshwater fish. Mercury accumulated in fish is usually in the form of methylmercury, whereas the source is usually inorganic. It has been proposed that methylation of inorganic forms is facilitated by microbial metabolism, either in the environment prior to assimilation or by microorganisms associated with bronchial mucosa. It has also been suggested that inorganic Hg is methylated in fish liver. Mosquito fish (*Gambusai affinis*) have been shown to absorb metallic Hg five times faster than inorganic Hg. This is presumably related to the high lipid solubility of the metallic form, which allows it to pass through gill membranes while salts become tightly bound to mucoproteins. High Hg levels in the gills have been observed in a number of species. Methylmercury levels in rainbow trout (*Salmo gairdneri*) steadily increase in the muscles and brain, whereas other Hg compounds accumulate primarily in the kidneys, spleen, and liver. In brook trout (*Salvelinus fontinalis*), this steady increase continued over a long period until a steady-state tissue level was achieved. During this period, no significant elimination was observed. Elimination rates reported for metallic and inorganic Hg (absorbed from water) are similar in mosquito fish, having a half-life of 45 days. The half-lives for inorganic and organic Hg taken up by the thornback rays (*Raja clavata*) from diet are 61 and 323 days, respectively.

As with fish, birds can assimilate organic forms of Hg more readily than inorganic compounds. Experiments with Japanese quail (*Coturnix coturnix japonica*) indicate that uptake is unaffected by route of administration. Methylmercury is distributed evenly in tissues while inorganic compounds accumulated primarily in the liver and kidneys of adult birds. The highest concentrations following chronic Hg exposure are generally found in the kidneys and liver. Accumulations may also occur in reproductive organs (e.g., goshawks, *Accipter gentilis*). Excretion appeared to be enhanced by egg laying, with concentrations of methylmercury occurring in the egg white and other Hg compounds typically in the yolk. Overall, inorganic forms are more rapidly excreted than methylmercury. Plumage and other keratinized structures represent an important excretion route for both forms, though especially for methylmercury.

The primary uptake route of Hg in marine and terrestrial mammals is though diet. This is related to the relatively high concentrations of methylmercury in food items (e.g., fish), which is more effectively taken up than inorganic forms. Studies with mice, sheep, cows, and humans have shown that over 95% of methylmercury given either in drinking water or food is taken up. Possible uptake in the lungs and over the skin is possible, but it is unlikely to be of much importance due to low environmental levels. Tissue levels have been reported from wild populations of seals, dolphins (*Stenella coeruleoalba*), and common or harbour porpoise (*Phocoena phocoena*) (Andre *et al.* 1990). Highest levels generally occur in the liver. Levels present in bone tissue are believed to be correlated with age.

7.4.5.4. **Selenium**

The metalloid Se is widely distributed in the environment, existing naturally in the form of selenide (Se (II–)), elemental Se, selenite (Se (IV+)), and selenate (Se (VI+)). Most Se compounds are water soluble and are thus readily available for dermal and gastrointestinal uptake. Under acidic conditions or in biological systems, Se compounds may be reduced to selenide, which may replace sulfur in amino acids (e.g., selenomethionine and selenocysteine). Many Se compounds are volatile and relatively unstable at room temperature (e.g., hydrogen selenide). Under certain conditions, therefore, the lungs can play an important role in the uptake of soluble as well as insoluble Se compounds.

Little or no information is available on the characteristics of uptake, distribution, and excretion of Se in plants, invertebrates, fish, and birds. Limited data are available for mammals from laboratory studies with rats, mice, dogs, monkeys, and ruminants. Certain plant groups accumulate Se to very high levels when grown in highly contaminated soils (e.g., several species of *Astragalus*, and some *Haplopappus* and *Stanleya*), whereas other plants (e.g., *Aster, Atriplex, Castelleja, Grindelia, Gutierrezia, Machaeranthera*, and *Menzela*) concentrate Se to lesser levels. Only low-level accumulation occurs in grasses, grains, and weeds. The ability of plants to accumulate Se from soils with low Se content is also variable (Högberg and Alexander 1986).

Results from animal studies suggest that Se compounds are effectively absorbed in the gastrointestinal tract. The rates of absorption were 90% for selenite and 80% for selenomethionine or selenocysteine administered orally. The degree of uptake was found to be much lower in sheep and cows (30-35%), although this probably reflects the reduction of selenite to its elemental form by bacterial action in the gastrointestinal tract (Högberg and Alexander 1986). Although little is known of the physiological processes governing absorption of simple Se compounds, there is some evidence that selenomethionine can be transported against a concentration gradient. Corresponding data for Se uptake in the lungs are unavailable. In the lungs of rats, uptake of selenious acid (H_2SeO_4) was 94%, and elemental Se, 57%. No data are available for dermal uptake, although it has been observed in rats.

Once absorbed, water soluble Se is rapidly distributed to most major organs, with highest concentrations in the liver and kidney. Little is known about Se transport. It has been suggested that after accumulation in red blood cells, Se compounds are translocated to plasma proteins (selenoproteins), which in turn may play an important role in transporting Se to the liver. In humans, Se binding to plasma lipoproteins has been demonstrated. The form of Se has been observed to have a marked influence upon accumulation; selenite-derived Se accumulates more rapidly in liver and kidney than selenate-derived Se (Högberg and Alexander 1986).

Mammals appear capable of maintaining Se homeostasis at low levels of exposure. Many Se compounds are biotransformed within the liver to excretable metabolites (e.g., methylated selenides such as trimethylselenide and dimethylselenide). Studies on rats have revealed that the predominant excretory pathway is via urine; the fraction excreted depends on nutritional status and dosage. At high levels, up to 67% of Se dosage is excreted in urine compared with 10% in feces (via bile). In contrast to the general pattern, ruminants can excrete up to 66% of the total Se dosage in feces. This increased fecal excretion has been attributed to poor gastrointestinal absorption rather than a greater rate of elimination. Significant quantities of methylated metabolites, such as dimethylselenide, can also be excreted from the body by exhalation. Experiments with rats, however, have demonstrated that this occurs only with dosages approaching lethal concentrations.

Two elimination phases are evident from animal studies. In the first rapid phase, the biological half-life of Se ranges from one to three days, depending on the compound and dose. The second phase of elimination, which has been found to be independent of dose, ranges from 30 to 70 days in most species. This pattern is slightly different in humans, where three phases have been identified with biological half-lives of about 1 day, 8-20 days, and 65-116 days (Högberg and Alexander 1986).

7.5. Toxicological effects

Numerous laboratory experiments have been carried out to assess the toxicological effects of acute and chronic doses of heavy metals. Intoxicants are often administered via inhalation, ingestion, injection, or absorption through the skin, so that effects can be directly related to dosage, forming the basis of dose-effect and dose-response relationships (Pfitzer and Vouk 1986). Most experiments focus on clinical signs and symptoms of lethal and sublethal toxicity. In contrast to this, the data currently available for heavy metal levels in tissues of Arctic organisms are generally obtained from environmental monitoring of natural populations. In this case, the emphasis is on exposure rather than dosage, and the focus is on the circumstances and exposure levels that may elicit effects (Elinder 1984). In the Arctic, metal concentrations, and hence dosages, generally do not change quickly (except in the event of a spill or an accidental release), and therefore information in relation to acute toxicity is less applicable than information relating to chronic toxicity.

Although many reliable measurements of metal levels in organisms have been obtained, there are relatively few measurements of metals in the environment in which the organisms live. Consequently, in order to evaluate the effects of heavy metal exposure on Arctic biota, it is necessary to extrapolate from results obtained under specific, laboratory exposure conditions. As little experimental toxicology has been carried out with Arctic organisms to date, in the majority of cases conclusions have to be drawn from counterparts in temperate climates. The extent to which results obtained for temperate species are applicable to Arctic species is not entirely clear, especially in light of the intrinsic differences between species in terms of sensitivity and responses to heavy metal exposure. Furthermore, environmental conditions associated with changing seasons are more extreme in polar regions, and there has been almost no experimental study of how seasonal variations influence toxicity.

The effects of heavy metal intoxication are many and varied, depending on the metal, the organism involved, the route of uptake, metabolism or biotransformation, and rates of elimination. The following is a brief overview of information on this subject contained in the WHO's International Programme on Chemical Safety (IPCS). This information includes the typical characteristics of and toxic effects attributable to each of the four heavy metals of greatest concern in the Arctic environment, Pb, Hg, Cd, and Se. Where possible, results from studies relating heavy metal burdens to toxicity will be cited for possible comparison with levels reported in Arctic flora and fauna (see section 7.7.3 and Table 7·22).

Although there are numerous data available for heavy metal concentrations in various Arctic biota tissue, there are few reports of toxic effects of heavy metals in relation to actual tissue levels. Most of the data contained in the IPCS regarding heavy metal toxicity are presented in the form of experimental concentrations (i.e., lethal concentrations – LC_{50}) or dosage administered (i.e., lethal dose – LD_{50}), rather than resulting tissue burdens. Therefore, in the absence of other data, only potential effects of heavy metal intoxication can be described below.

7.5.1. Lead

According to WHO (1989a), Pb levels present in the environment are unlikely to affect aquatic plants. Reductions in photosynthesis and respiration have been reported in sunflowers along with reductions in overall growth in lettuce and carrots after exposure to very high Pb concentrations. In was concluded that Pb is only likely to affect terrestrial plants at very high environmental concentrations (e.g., >0.005 mg/L).

The tolerance of invertebrates to Pb salts is variable, particularly where populations have experienced prior exposure. Symptoms of toxicity reported for nematodes and caterpillars include impaired reproduction and development. Caterpillars (*Scotia segetum*) fed contaminated food throughout development showed decreased survival to adult stage, and those that did survive were visibly deformed and unable to produce eggs. Mature mussels were able to tolerate large amounts of Pb in their tissues without apparent toxic effects (by enclosing the metal in membrane-bound vesicles). Settlement of oyster larvae was found to be significantly reduced for *Crassostrea gigas*, as exposure to >0.01 mg/L Pb brought about reduced larval development and therefore delayed peak settlement time. Reduced oxygen consumption has been reported for shrimp exposed to high concentrations of Pb. A dose-related decrease in oxygen consumption (0-2.0 mg/L) was observed with crayfish (*Orconectes virilis*) exposed to Pb acetate for a period of ten days, although long-term acclimatization was evident after 20, 30, and 40 days. Although Pb apparently reduces the capacity for oxygen uptake through the gills, the crayfish were able to compensate by increasing the flow of water over gill surfaces. Growth suppression has been reported for larval brine shrimp (*Artemia*) with exposure to concentrations exceeding 5 mg/L.

Chronic exposure of adult fish to inorganic Pb can cause sublethal effects on morphology, enzyme activity, and avoidance behavior. Juvenile stages are more susceptible to Pb than adults or eggs. In a study designed to establish acceptable toxicant limits for inorganic Pb, Davies *et al.* (1976, cited in WHO 1989a) observed blackening of the tail, followed by spinal curvature and eroded caudal fins in response to exposure to sublethal concentrations of Pb. These sublethal concentrations were not related to tissue burdens. Other studies have also reported anemia and basophilic stippling of erythrocytes. The presence of Pb is also believed to result in reduced larval survival due to skeletal malformation, erosion of fins, outgrowths from the fry, and poor absorption of yolk.

A number of studies reporting Pb toxicity in birds, mainly gallinaceous species, have revealed that both Pb salts and metallic Pb are not toxic except at very high concentrations. Comparable sensitivities are assumed for other bird species. For chickens, Japanese quail, and mallard ducks (*Anas platyrhynchos*) a variety of sublethal acute effects have been reported, including lethargy, weakness followed by anorexia and anemia, and an overall reduction in weight gain. High Pb levels in blood have been correlated with suppression of the activity of the enzyme delta-ALAD in erythrocytes, which is involved in heme synthesis. With low doses of Pb acetate, egg production in Japanese quail was depressed, and this effect was enhanced with increased levels. Where dosages were high, egg production was almost completely depressed and the eggs which were produced were either soft shelled or without shells. Hatch rates were also reduced.

Birds can be exposed to high dosages of metallic Pb through the ingestion of pellets of lead shot. Over a period of 20 days, adult mallard ducks showed clear symptoms of Pb poisoning in response to sublethal dosages, including green diarrhea, anorexia, and weakness. Short-term Pb toxicosis involving varying degrees of paralysis and abnormal locomotor function has been reported as being transitory, disappearing after eight days. Intoxication with metallic Pb also brought about chronic inhibition of erythrocyte delta-ALAD activity. Results also indicate that trialkyllead compounds are very toxic, but effects have only been reported for starlings (*Sturnus vulgaris*). The symptoms included severe lack of co-ordination, resulting in decreased feeding.

In mammals, the major effects of Pb are related to the hematopoietic, nervous, gastrointestinal, and renal systems. Symptoms and effects of inorganic Pb poisoning are described for humans in chapter 12. It is assumed that the principal effects of Pb intoxication in mammals are likely to be similar to those described for humans. The primary effects of Pb intoxication include 1) interference with the synthesis of red blood cells which are necessary for oxygen transport (anemia), 2) damage to both the central and peripheral nervous system leading to encephalopathy and neurological dysfunction, and 3) gastrointestinal symptoms such as loss of appetite, diarrhea, constipation, and in severe cases colic (Tsuchiya 1986).

7.5.2. Cadmium

Few reports are available concerning the toxic effects of Cd on plants. Reduced growth and the loss of chlorophyll-*a* from leaves have been reported for two floating weeds (*Lemna minor* and *Salvinia natans*) and water hyacinth plants exposed to Cd. Plants grown in soil are generally insensitive to Cd except at high concentrations. Two tree species, white pine (*Pinus strobus*) and red maple (*Acer rubrum*), responded differently to high Cd exposure. Red maple seedlings developed interveinal chlorosis and stunting of the leaves, wilting, and death, while pines experienced inhibition of needle expansion. Effects were also observed on root development. At high dosages, there was both a reduction in the number of new roots and a stunting of those already present. Stunted growth and similar toxic symptoms have also been observed in lettuce, cabbage, carrot, and radish plants. Physiological studies have demonstrated that increasing Cd concentrations reduce stomatal opening, leading to a reduction in both transpiration and photosynthesis.

Laboratory studies of acute toxicity suggest that Cd is moderately to highly toxic to aquatic invertebrates, depending on a variety of environmental variables (temperature, salinity, organic content of water, and chelating agents). Effects of long-term exposure can include larval mortality and temporary reduction in growth. In an investigation of sublethal concentrations and effects on adult shrimp (*Palaemonetes pugio*), Cd body burdens of 40 µg/g were found to inhibit molting. At more moderate concentrations of 23 µg/g and 10 µg/g, molting was stimulated. Histological effects at near-lethal concentrations included blackening and damage to gill filaments, although no body residue levels were given. Terrestrial invertebrates, on the other hand, are relatively insensitive to Cd exposure, probably due to the presence of effective sequestration mechanisms in specific organs. Nevertheless, nematodes exposed to sublethal doses of Cd do not grow as large as controls and have significantly reduced reproductive success. Similarly, growth and reproduction are the parameters found to be most sensitive to Cd in the ollembolan (*Orchesella cincta*) and the oribatid mite (*Platynorthrus peltifer*). Few studies are available on the toxic effects of Cd on mollusks. Some work has been carried out on effects on early development stages of the mud snail (*Ilyanassa obsoleta*) and the sea star (*Asterias rubens*). In both cases, development was adversely affected. Subadult garden snails fed Cd responded with decreasing food consumption with increasing dosage. At high levels, both growth and reproductive activity were suppressed.

Acute toxicity of Cd has been studied in a variety of fish with respect to lethal concentrations. LC_{50} values have not been related to tissue burdens. Sublethal effects have been reported for a number of fresh water and saltwater species. Eggs and larvae are most sensitive to Cd exposure, responding to exposure with either delayed hatching or non-emergence and inferior larval growth. The most sensitive indicator of Cd toxicity in Atlantic salmon (*Salmo salar*) was found to be inhibition of growth of alevins. When three generations of brook trout were intoxicated, significant numbers of first and second generation males died during spawning. Cadmium toxicity in fish is characterized by ionic imbalance with reduced plasma Ca(II), Na(II), and Cl^-. The probable mechanism underlying this toxicity is inhibition of ion-transporting enzymes by Cd(II) present in gill membranes. Furthermore, Cd has been shown to inhibit Na/K-ATPase in the gills, thereby having an influence on ATP production in the gill. Chronic exposure may also bring about prolonged hypoglycemia and reduced ability to absorb glucose and fructose in the gut. Cadmium has also been shown to interfere with calcium metabolism. Minnows (*Phoxinus phoxinus*) exposed to various concentrations of Cd developed fractured vertebrae in the caudal region. Spinal abnormalities were also observed with newly hatched medaka fry (*Oryzias latipes*) after eggs were exposed. Fish showing malformations of the spine after exposure to Cd had significantly less Ca(II) in the vertebral column than did control fish.

Kidney damage has been reported in wild colonies of pelagic sea birds having Cd residues of 60-480 µg/g. Similar results have also been obtained in laboratory studies in which birds were dosed so that Cd levels in kidney ranged from 95 to 240 µg/g. The damage included cell necrosis, nuclear pyknosis, mitochondrial swelling, and some tubulorrhexis, although there was some indication of regeneration. Spermatogenesis was reduced in wood pigeons (*Columba palumbus*) and mallards exposed to Cd.

Chronic Cd exposure produces a wide variety of similar acute and chronic effects in mammals and humans. These are discussed in detail in section 12.2.3.2. Kidney damage and lung emphysema are the primary effects of high levels of Cd in the body. No laboratory investigations have been carried out on the toxic effects of high Cd burdens in Arctic mammals. However, based on human toxicology, it is reasonable to assume that Cd concentrations of 100-200 µg/g in the kidneys may represent a risk. At these concentrations, the first signs of renal damage (proteinuria) have been observed in land mammals and humans. Accumulation of Cd in the kidney can also cause disturbance in Vitamin D and calcium metabolism, and in severe cases can lead to osteomalaci and osteoporosis. Experiments with rats have shown that Cd concentrations in the range of 50-170 µg/g can cause an inhibition in the change of Vitamin D to the active form 1,2,5 DHCC, which has an important regulating effect on calcium metabolism and stimulates the uptake of calcium from the intestine. Cadmium is seldom the cause of liver damage in mammals; however, experiments with rats involving long-term exposure (six months), with final Cd concentrations of 188 µg/g, caused changes in liver cell morphology and enzyme activity (Elinder 1985).

7.5.3. **Mercury**

Susceptibility to toxic effects from high Hg accumulation varies greatly. The degree of toxicity is related to a number of environmental parameters including temperature, salinity, dissolved oxygen, and water hardness. Organic forms of Hg are generally more toxic than inorganic forms in both aquatic and terrestrial organisms. This is related to the lipophilic properties of fine Hg compounds. As a protective mechanism, some microorganisms biotransform inorganic Hg complexes assimilated from their surroundings into organic forms. This process of biomethylation renders Hg much more accessible to higher organisms.

A number of adverse effects have been reported for plants exposed to a wide range of Hg concentrations. High concentrations of inorganic Hg affect macroalgae (*Laminara hyperborea*) by reducing gametophytic germination. Adverse effects on the growth of the red algae (*Plumaria elegans*) have been reported, with reductions of up to 50% following acute exposure. Significant reductions in growth have also been observed in cauliflower, lettuce, carrots, and potato tubers. Water cabbage (*Pistia stradiotes*) exposed to various concentrations of inorganic Hg responded by increasing levels of free amino acids and decreasing chlorophyll content, protein, RNA, and enzymatic activities (catalase and protease). These changes typically lead to senescence. A closer look at the toxic effects at the tissue level in *Elodea densa* revealed differences between tissues. Apical cells and roots developed abnormal nuclear and mitochondrial characteristics, although roots were less sensitive. Mitotic activity was inhibited in root meristems. In the presence of methylmercury, mitosis was promoted in bud meristems, although cell divisions were abnormal. Methylmercury inhibited the development of both root and bud initials in *Elodea densa*. Finally, a study with roots and shoots of barley plants indicates that inorganic Hg may also significantly reduce K^+ and PO_4 uptake and translocation.

There are no reports of the effects of chronic exposure on invertebrates. Most studies have focused on acute and short-term effects. Invertebrates in general are more susceptible to Hg toxicity during the larval stages. Acute exposure inhibits embryogenesis of developing American oyster larvae. Studies of the effects of Hg on crustacean larvae have revealed extended periods of metamorphosis and decreased survival (blue crabs, *Callinectes sapidus*), as well as reduced settling (barnacles, *Balanus balanoides*). Larval shrimp (*Palaemonetes vulgaris*) did not show immediate responses to acute exposure. Instead, delayed effects were noted including reduced post-larval survival, delayed molting, extended development times, increased number of larval instars, and an increase in the occurrence of abnormalities.

There is much less information available on potential effects in adult invertebrates. Significant reductions in growth have been observed in some species (e.g., blue mussels and terrestrial slugs). Adult crabs (*Carcinus maenus*) exposed to high Hg concentrations displayed reduced cardiac activity and oxygen consumption, and in severe cases a loss of the ability to osmoregulate. Mercury is also believed to decrease the rate at which limb regeneration occurs in crabs (e.g., *Uca pugilator*). A single study of behavioral responses of grass shrimps (*Palaemonetes pugio*) to acute intoxication demonstrated that the probability that prey organisms would be caught by predators increased after exposure, indicating a general decrease in responsiveness.

Physiological biochemical abnormalities have been described for a number of fish species following exposure to sublethal concentrations of Hg. These include depressed olfactory response with increasing concentrations and exposure duration (e.g., rainbow trout and *Anabas scandens*); reduced gill filament respiration due to damage to secondary lamellae (e.g., roach, *Leuciscus rutilus*); blindness; reductions in hemoglobin content, erythrocyte count, body weight, and body protein (*Anabas scandens*); and, finally significant decreases in intestinal absorption rates of glucose, fructose, glycene, and trytophan. Osmoregulatory effects have also been observed in some fish (rainbow trout), particularly in response to exposure to inorganic Hg. It has been suggested that this symptom arises as a result of enhanced mucus production on the gills caused by Hg exposure. There is also some suggestion that immune response can be affected.

In addition to the effects already described for fish, Hg has an adverse effect on reproductive performance in many species. Chronic exposure was seen to suppress oocyte development in freshwater teleosts (e.g., *Channa punctatus*). This effect corresponded with the suppression of gonadotrophs in the pituitary, inducing a gonadal 'resting phase'. Furthermore, studies investigating the effects of exposure on rainbow trout sperm indicate that Hg in the water column has the potential to significantly decrease fertilization. Environmental contamination also has the potential to inhibit hatching success (e.g., zebra fish, *Brachydanio rerio*) and promote embryo deformation. Killifish (*Fundulus heteroclitus*) exposed to inorganic Hg during embryonic development showed an increasing incidence of spinal curvature, although the susceptibility of eggs was variable.

Most of the effects identified for inorganic Hg intoxication in birds have been reported for gallinaceous species, which are considered to be unrepresentative of other birds. In general, organic Hg is more toxic than inorganic salts. The primary effect, which is evident from various studies (e.g., chickens and Japanese quail) is that birds refuse both food and water, leading to subsequent poor growth. Therefore the clinical effects may be both direct and indirect. It has been estimated that a period of at least two weeks of exposure is required before the direct effects are likely to occur.

A number of more subtle effects have also been identified from laboratory studies, including effects on enzyme systems, cardiovascular function, blood parameters, immune response, and kidney function and structure. The activity of two enzymes in particular, cholinesterase and lactate dehydrogenase, can be altered by oral Hg exposure in Japanese quail. Cholinesterase activity decreased with dosage while lactate dehydrogenase activity increased to three times its original level irrespective of dose. In chickens, Hg intake induced cardiovascular disturbances in the form of myocardial changes and alterations in the number of red blood cells, hematocrit, and corpuscular volume. Chronically exposed chickens showed suppressed primary and secondary immune responses. Damage to the proximal tubules of the kidney has also been reported to result from oral exposure (e.g., juvenile starlings). Some reproductive effects have been reported (e.g., reduced fertilization and weakening of the egg shell), although these are not as prominent for inorganic Hg salts as for organic Hg.

Sublethal doses of organic Hg result in neurological impairment, reproductive effects, and weight loss in birds. Neurological symptoms including impaired co-ordination of muscle movements and ataxia have been observed in a number of birds (domestic fowl; penned pheasant, *Phasianus colchicus*; red-tailed hawk, *Buteo jamaicensis*; goshawk). On a physiological level, effects have been described as resulting from degeneration of cerebellum and medulla oblongata and from demyelination of the spinal cord and peripheral nerves. In red-tailed hawks, observed effects on feeding and eventual weight loss were also partly attributable to feeding difficulties resulting from impaired muscle co-ordination. However, autopsies of goshawks exposed to Hg revealed that the dominant effect leading to weight loss was likely to be muscular atrophy. In many cases, reductions in egg production and hatchability have also been reported. Some studies have described significantly reduced egg production, chick weight, and survival. In addition, large numbers of thin-shelled and shell-less eggs were seen in some studies (e.g., penned pheasant, Japanese quail). Mercury concentrations above 30 μg/g ww in liver and kidney and 2-3 μg/g ww in eggs may result in detrimental effects on birds (see Table 7·22).

Although Hg is primarily a neurotoxin in mammals, it is also responsible for damaging reproductive capacity as it interferes with spermatogenesis, causing chromosome damage in connection with mitosis. Developing fetuses, and breastfeeding offspring in particular, are sensitive to Hg in the form of methylmercury. WHO has concluded that the developing fetus is especially susceptible to Hg due to intense cell division and formation of the central nervous system. An effective placental barrier exists against the transfer of inorganic Hg, but not methylmercury which readily transfers from maternal to fetal tissue and accumulates to some extent in the fetal brain (Bremner 1974).

As outlined in Table 7·22 (section 7.7), liver and kidney Hg concentrations above 25-60 μg/g ww may constitute a risk for Arctic mammals. Experiments on other animals have shown that the organs most susceptible to damage from methylmercury and inorganic Hg are the central nervous system and kidney, respectively. There are no guideline concentration limits available for these organs in humans, as the parameter used for determining Hg burden is blood concentration. Marine mammals with Hg levels in the blubber could potentially be at risk at times when food is scarce. However, concentrations in blubber of Arctic marine mammals are generally low.

7.5.4. Selenium

Overall, there is little information on the toxicity of Se. There is no information on the effects of acute and chronic exposure to plants, invertebrates, fish, or birds. Limited data are available for animals and humans. Under acute exposure, the critical organ of Se toxicity appears to be the central nervous system. Pathological studies on animals suffering from acute Se poisoning have revealed liver and kidney congestion, endocarditis, myocarditis, and peticheal hemorrhages of the epicardium. Selenium intoxication in dogs and rats reportedly induces vomiting, dyspnea, tetanic spasms, and finally death by respiratory failure. In a study investigating the long-term effects of Se, rats fed on seleniferous sesame meal suffered from hepatic lesions with 7.34 μg/g Se in the liver, whereas rats with 0.72 μg/g had no lesions.

Toxic effects resulting from long-term Se exposure through consumption of accumulatory plants have been reported for livestock. The so-called 'Blind staggers' syndrome, which can take several weeks to develop, is characterized by impaired vision, reduced appetite, and a tendency to wander. The effects of chronic poisoning on livestock (the so-called 'alkali disease') include emaciation, hair loss from the mane and tail, deformation and shedding of hooves, and erosions of the joints of long bones. Liver cirrhosis may develop in severe cases. Similar effects have also been reported in sheep and dogs (Högberg and Alexander 1986).

Anecdotal reports suggest that excess Se may also decrease reproductive performance in farm livestock and hatchability in chickens. This has also been reported to be the case with mice. In fact, Se accumulates in mammalian testes to a greater extent than in other tissues (see review by Hansen and Deguchi 1996), and influences spermatozoal motility. Selenium is incorporated in the mitochondrial capsule of sperm, and in this way is likely to affect its behavior and function. In humans, both high and low levels have negative effects on the number and motility of spermatozoa. No data are available for the effects of Se on female fertility.

Studies of this type are difficult to interpret and extrapolate to other mammal species. Two independent studies have demonstrated that, under certain circumstances, Se compounds are less toxic to animals which normally have a high dietary intake of Se.

7.6. Regional and circumpolar levels and trends of metal contaminants

7.6.1. Atmosphere

7.6.1.1. Air concentrations in the High Arctic

Air contamination from anthropogenic sources has been measured throughout the Arctic air mass (Heidam 1984, Lowenthal and Rahn 1985, Ottar *et al.* 1986, Shaw 1988, Barrie *et al.* 1989, Djupstrøm *et al.* 1993) (Figure 7·18). Vertical profiles of air concentrations obtained during several research programs have added to the understanding of the physical and chemical characteristics and origin of Arctic air contamination. Of particular interest in this connection are the results of the three Arctic Gas and Aerosol Sampling Program (AGASP) campaigns in 1983 (Schnell 1984), in 1986 (Herbert *et al.* 1989) and in 1989 (Schnell *et al.* 1991); the 1988 Canadian Polar Sunrise Experiment (Barrie 1991); and the British Petroleum (BP) program in the Norwegian Arctic from 1982 to 1984 (Ottar *et al.* 1986). The following description of Arctic air pollution is based on measurements carried out within these programs. In the lower troposphere (up to 3 km) particle layers are frequently strongly banded (Ottar and Pacyna 1986, Shaw 1986). The temperature and wind profiles measured through these layers suggest that their thermal stability is sufficient to maintain these laminar structures, although there is often considerable wind shear between the layers (Radke *et al.* 1989). Aerosol concentrations in the lower troposphere show significant variations depending on meteorological conditions, primarily temperature inversions.

The measurements of vertical profiles revealed layers of particles in the upper part of the High Arctic troposphere, although particle concentrations were much lower than those in the lower troposphere. Aerosol size measurements in the lower (up to 2-3 km) and upper (from 3 to 4-5 km) parts of the troposphere during winter were highly variable, with fine particles (< 1.0 µm) dominating the lower part and coarser particles in the upper part. The larger concentrations of small particles in the lower part of the troposphere appeared to be associated with air masses transported directly from emission regions. They may also have been a result of gas-to-particle conversion (Bodhaine 1989).

During summer, aerosol concentrations are usually very low throughout the High Arctic. However, vertical profiles measured in the Norwegian Arctic sporadically indicated layers of polluted air with a well defined lower boundary at about 2 km (Pacyna and Ottar 1988). Particles in the upper part of the troposphere in summer were of similar size to those measured in winter.

Fine particles contain high concentrations of several anthropogenic heavy metals, particularly during episodes of long-range transport of air masses passing over the major emission source regions (Ottar *et al.* 1986). Coarse particles, not correlated with haze, consist mainly of clay minerals, other soil constituents, and, to a lesser extent, seasalt compounds. The chemical composition of particles measured in the Norwegian Arctic during a winter flight is shown in Figure 7·22. Data obtained on this and other research flights in the Norwegian Arctic (Pacyna *et al.* 1985) indicate the presence of high concentrations of several anthropogenic heavy metals and natural compounds in the lower layer of the Arctic troposphere (up to 2-3 km). The upper layer (from 3 to 5 km) contains a mixture of natural compounds and anthropogenic heavy metals with the latter group in small concentrations.

Several ground-measurement campaigns have been carried out in the High Arctic to assess the level of heavy metals and other chemicals and to use these data to identify the origin of Arctic air pollution. Knowledge of the subject ex-

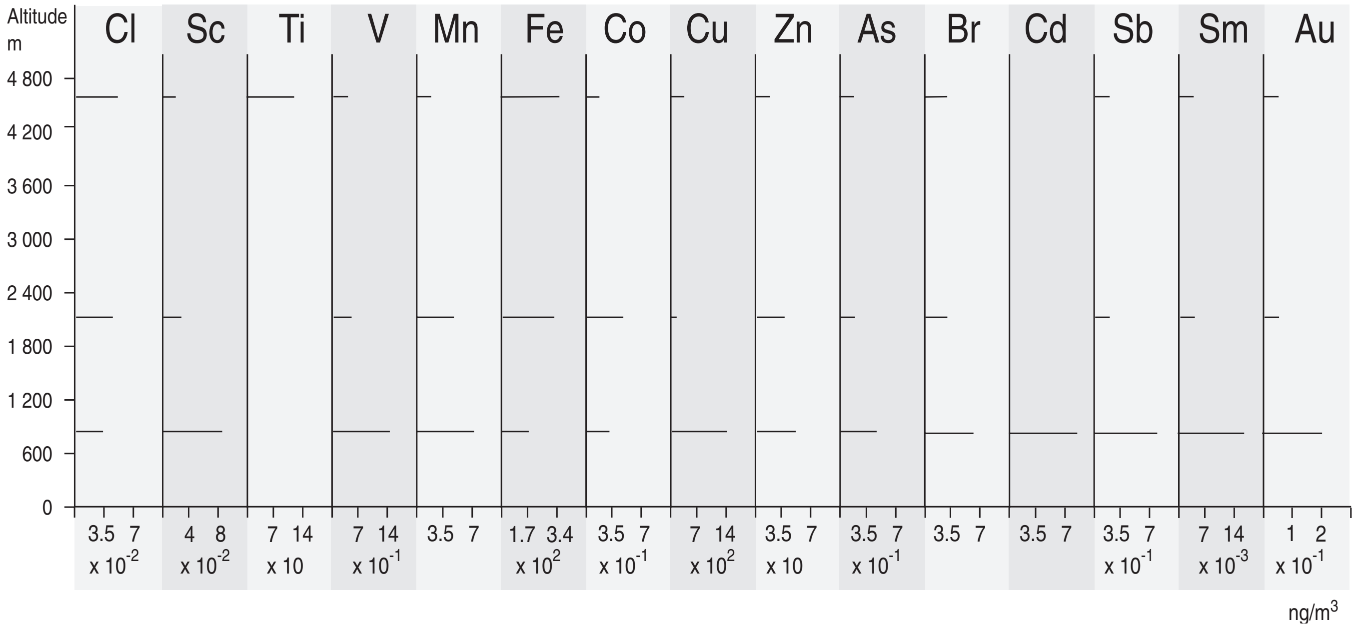


Figure 7·22. Composition of particles measured at different altitudes during winter flights in the Norwegian Arctic. (After Pacyna *et al.* 1985).

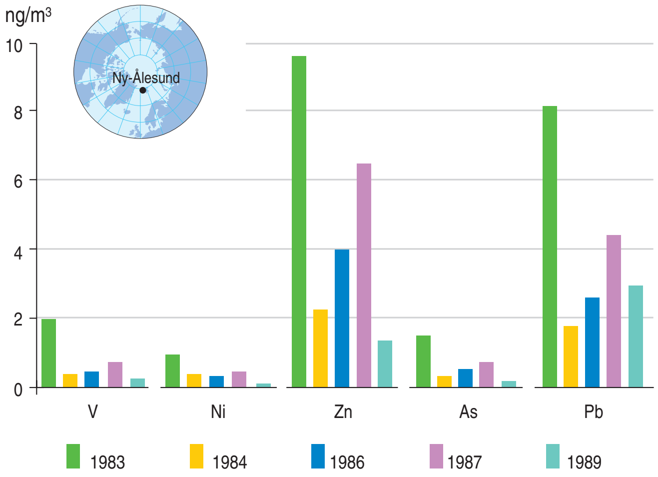


Figure 7·23. Atmospheric winter concentrations (median values) of metals measured in different years in the < 2.5 µm aerosol fraction at Ny-Ålesund, Svalbard. (After Maenhaut *et al.* 1996).

Table 7·7. Winter and summer median concentrations (ng/m^3) of heavy metals at Ny-Ålesund (Svalbard) and Alert (Canadian Arctic) in 1984.

Metal	Ny-Ålesund[a] Winter	Ny-Ålesund[a] Summer	Ny-Ålesund[a] Winter: Summer ratio	Alert[b] Winter	Alert[b] Summer	Alert[b] Winter: Summer ratio
Cu	0.9	0.3	3.0	1.52	0.55	2.8
Ni	0.38	0.1	3.8	0.25	0.07	3.6
Pb	1.76	0.20	8.8	2.15	0.22	9.8
Zn	2.3	0.15	15	3.88	0.97	4.0

a. Data from Norwegian Institute of Air Research (NILU).
b. Data from L. Barrie pers. comm.

panded greatly after North American and Western European research agencies started a coordinated research network in 1977. This effort resulted in extensive data sets of concentrations in surface air for Greenland (Heidam 1984), the Norwegian Arctic (Larssen and Hanssen 1980, Heintzenberg *et al.* 1981), northern Canada (Barrie and Hoff 1985, Barrie *et al.* 1989), and northern Alaska (Rahn and Heidam 1981, Rahn *et al.* 1981, Shaw 1988). Since then, information on heavy metals in the background aerosol in the Russian Arctic has become available (Vinogradova *et al.* 1993).

Median winter concentrations of heavy metals and other compounds measured in 1983, 1984, 1986, 1987, and 1989 at Ny-Ålesund, Spitsbergen, are presented in Figure 7·23 (after Maenhaut *et al.* 1996). The median values in the 1989 campaign are generally of the same order as those in the previous winter campaigns, but for some heavy metals having strong anthropogenic sources (such as V, Mn, Ni, Zn, As, Mo, Sb, and Pb), they are clearly lower.

Much less information exists on heavy metal concentrations during summer. A comparison of median concentrations measured at Ny-Ålesund and Alert in the Canadian Arctic in winter and summer of 1984 are shown in Table 7·7 (Maenhaut *et al.* 1989). The ratios of winter concentrations to summer concentrations range from 3 to 15. The ratios for individual metals are generally similar for the Alert and Ny-Ålesund locations.

Results of the three concurrent measurement campaigns at Poker Flat (PKF) and Point Barrow (BRW) in Alaska and Ny-Ålesund carried out in 1986 have been compared (Djupstrøm *et al.* 1993). Data for As and V are presented in Figure 7·24. It was concluded that polluted aerosols transported to the High Arctic within air masses from source regions at lower latitudes can be trapped in the polar region, particularly during winter. Having rather low potential for deposition due to their small particle size, aerosols can circulate with the Arctic air masses over the region. Therefore, a load of air pollution originating, for example, in northern Eurasia can under certain conditions reach Alaska first and then travel farther in a return flow over the North Pole to

Figure 7·24. Concentrations of As and V in aerosols measured during concurrent measurement campaigns on Svalbard and two locations in Alaska during April 1986. (After Djupstrøm *et al.* 1993).

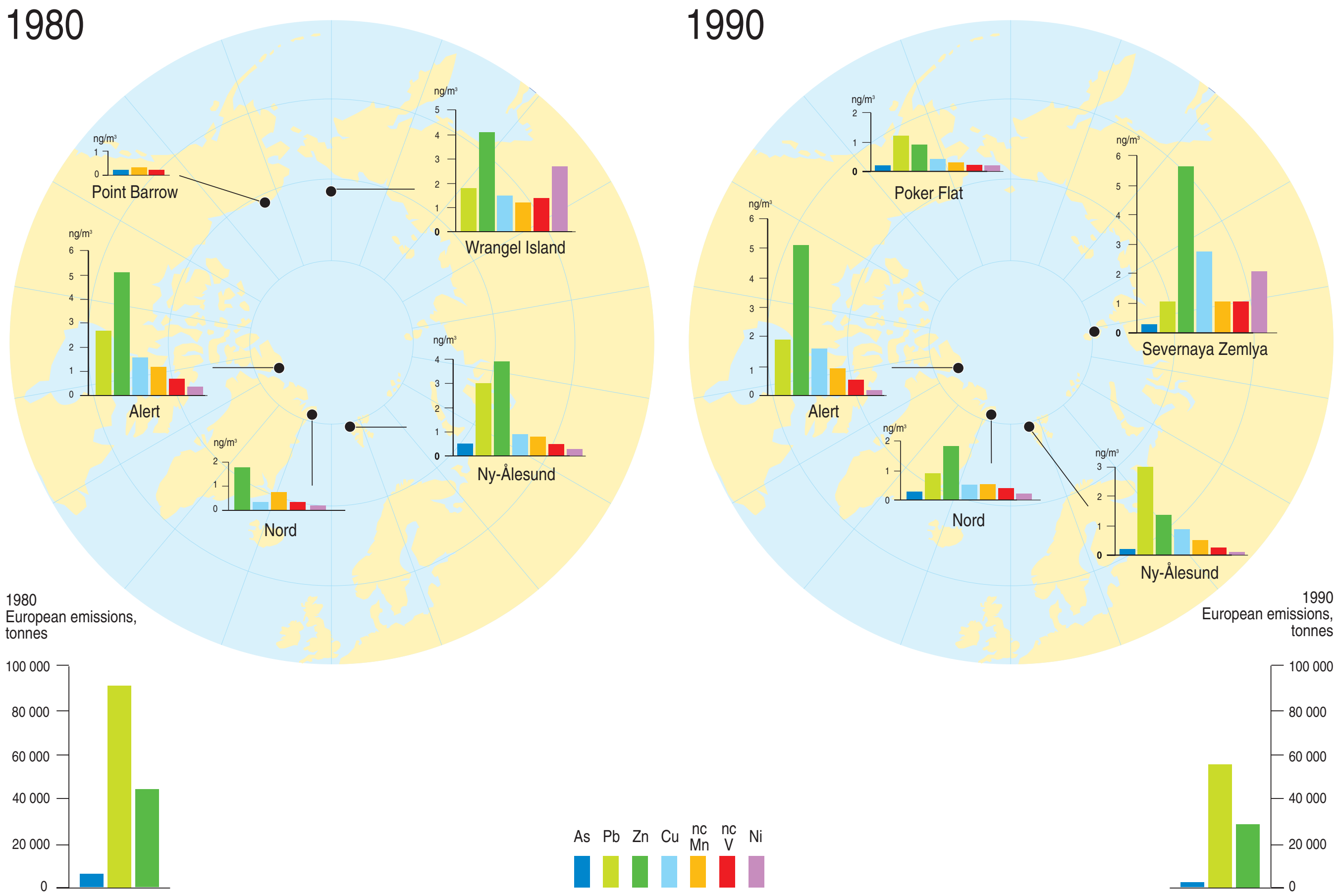


Figure 7·25. Winter concentrations of metals in air at remote locations in the Arctic at the beginning of the 1980s and 1990s, and European emissions of As, Pb and Zn during the same periods. (Data sources: Ny-Ålsund, Norwegian Institute of Air Research; Alert, Len Barrie pers. comm.; Nord, Niels Heidam pers. comm. and Danish Environmental Protection Agency 1997; Severnaya Zemlya and Wrangel Island, Russian State Committee for Hydrometeorology Assessment 'Heavy metals in the Arctic'; Point Barrow and Poker Flat, Glen Shaw pers. comm. and (Pb, Zn, Ni from 'Gates of the Arctic' station, Fairbanks near Poker Flat) Polissar *et al.* 1996).

the Norwegian Arctic. In this way concentrations measured at various stations in the High Arctic are intercorrelated. Indeed, analysis of a 4-year chemical data set from Alaska (Shaw 1991b) has proved this hypothesis. Concentrations of several heavy metals measured in the air over Greenland (Heidam 1984) and at stations in Canada, such as Alert, Igloolik, and Mould Bay (Barrie and Hoff 1985, Barrie *et al.* 1989), are similar to those measured in the Norwegian Arctic and Alaska. The concentrations of V, Co, Zn, and Sb in these parts of the Arctic are also in good agreement with the concentrations measured at the stations in Severnaya Zemlya and Wrangel Island in the Russian Arctic (Vinogradova *et al.* 1993). A lack of data made the comparison for other heavy metals impossible.

Winter air concentrations of several heavy metals in the Norwegian Arctic, Alaska, Greenland, the Canadian Arctic, and Severnaya Zemlya and Wrangel Island in the Russian Arctic in the 1980s and the 1990s, together with European emissions of As, Pb and Zn during the same periods, are presented in Figure 7·25. Concentrations at the beginning of the 1980s are somewhat higher than those at the beginning of the 1990s. This is particularly true for Pb due to the reduction in the use of leaded gasoline in various regions of the Northern Hemisphere. However, the available data cannot provide enough information to assess quantitatively the decrease in concentrations from the 1980s to 1990s.

The concentrations of heavy metals in Arctic air in winter are generally one order of magnitude higher than the winter concentrations measured in Antarctic air. Summer concentrations in the Arctic and Antarctica are similar.

7.6.1.2. **Concentrations of heavy metals in subarctic air**

Air concentrations of heavy metals in the High Arctic are much lower than the concentrations measured around major point sources of emissions located mostly in the subarctic region. For example, concentrations of Ni, Cu, and As measured at several stations in northern Norway (Figure 7·26) and on the Kola Peninsula are at least one order of magnitude higher than the concentrations measured at Ny-Ålesund (Figure 7·25). Maximum daily concentrations of these metals on the Kola Peninsula are at least two orders of magnitude higher than the concentrations measured during the episodes of air pollution transport to Ny-Ålesund. Concentrations of several heavy metals near the Severonikel smelter stacks on the Kola Peninsula are about three orders of magnitude higher than the maximum concentrations at Ny-Ålesund (Panichev *et al.* 1993). Concentrations decrease by one order of magnitude within 20 km distance from the stacks.

7.6.1.3. **Atmospheric deposition in the Arctic**

Although there have been a number of successful studies directed at the origin of Arctic air pollution by heavy metals, a quantitative assessment of the portion of the pollution load actually deposited in the High Arctic has not been made. Information on deposition of heavy metals in the Arctic can be obtained from studies by Davidson (1991) who confirmed that atmospheric chemistry over the Greenland Ice Sheet differs greatly from the patterns observed at coastal sites. This observation is very important when assessing the magnitude

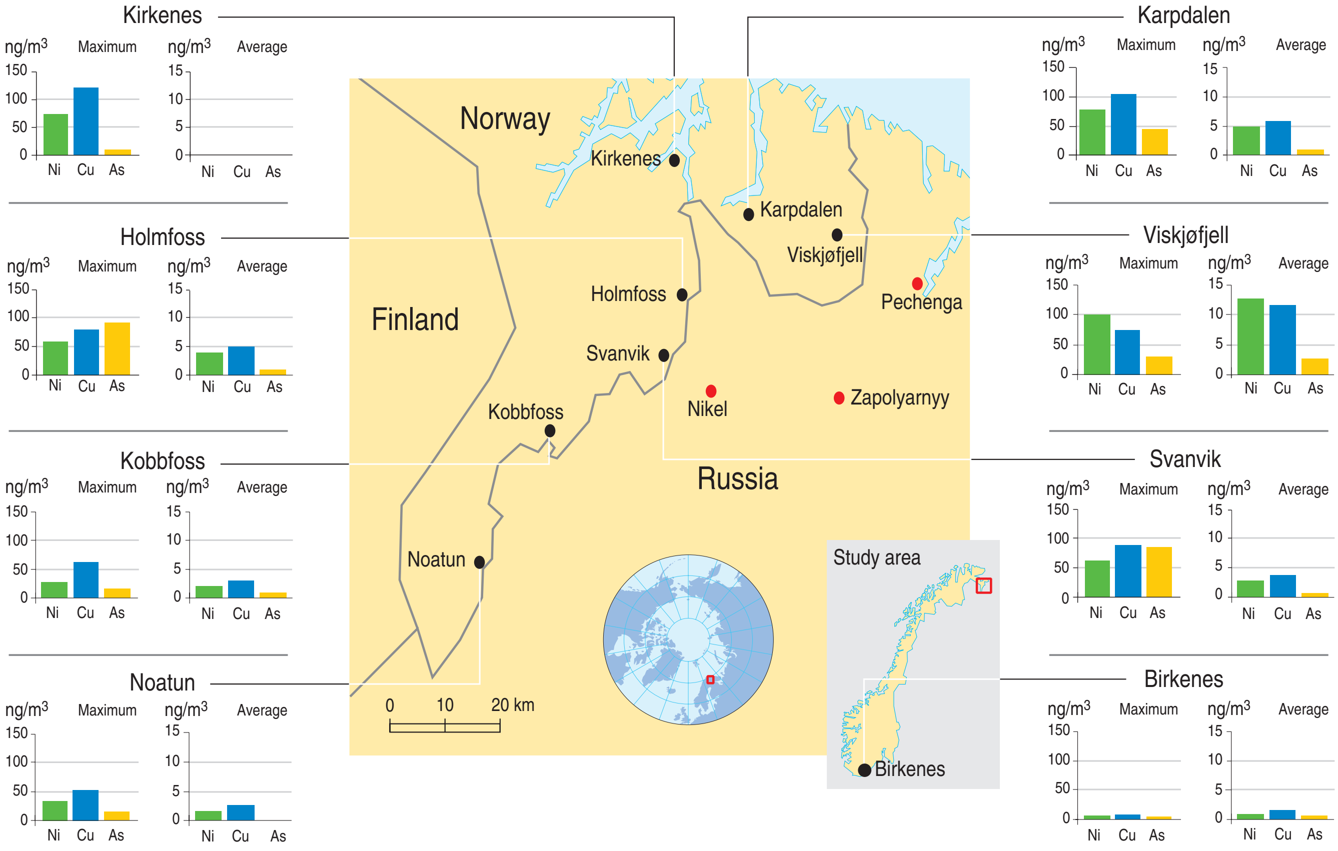


Figure 7·26. Average (15-month) and 24-h average maximum air concentrations of Ni, Cu and As measured at several stations in northern Norway near to major point sources on the Kola Peninsula. (Source of data: Norwegian Institute for Air Research).

of atmospheric deposition of heavy metals on the basis of measurements.

The past and present atmospheric deposition fluxes of heavy metals in the Arctic can be estimated by combining the heavy metal concentrations measured in the snow or ice with the snow/ice accumulation rates. Candelone *et al.* (1996) have made these calculations for Summit in Greenland for different times: 7760 years BP, which gives the pre-human activity Holocene fluxes; 1773, which corresponds to the onset of the first Industrial Revolution; 1850, which is the time when concentrations of some heavy metals started to increase; and 1992. The results of flux estimates for Pb, Zn, Cd, and Cu at Summit are presented in Table 7·8 for these periods, as well as for the 1960s and 1970s when the metal concentrations reached maximum values. The authors then estimated the cumulative anthropogenic deposition flux of heavy metals to the Greenland ice cap from 1773 to 1992, i.e., from the onset of the first Industrial Revolution to present. The cumulative deposition flux during this period was 3200 tonnes for Pb, which is one order of magnitude higher than the deposition of the metal calculated for Greco-Roman times (Hong *et al.* 1994). For Zn, Cd, and Cu, the estimated fluxes were about 2500, 60, and 200 tonnes, respectively. There has been a reduction in the use Pb in gasoline over the last two decades. A corresponding decrease can be monitored in the Greenland Ice Sheet (e.g., Boutron *et al.* 1991, 1995, Hong *et al.* 1994).

Table 7·8. Heavy metal deposition fluxes at Summit, central Greenland. (After Candelone *et al.* 1996).

Year	Cd	Flux, ng/cm^2/y Cu	Pb	Zn
7760 BP	0.006	0.039	0.013	0.53
1773	0.006	≤ 0.064 [a]	0.18	0.37
1850	0.006	0.053	0.35	0.7
1992	0.018	0.17	0.39	1.2
1960s/1970s maximum	0.041	0.22	2.5	0.041

a. No satisfying plateau of concentration was obtained for Cu in the 1773 sample.

In Norway, surveys of Pb concentrations in moss have been repeated since 1975 to estimate the latitudinal gradient in Pb deposition (Figure 7·27, next page). These studies also indicate a decreasing trend in Pb deposition (Steinnes *et. al.* 1994, Steinnes 1995).

Other deposition measurements have also been carried out in subarctic areas, such as the Kola Peninsula, northern Scandinavia, and Canada. The results of measurements on the Kola Peninsula carried out by Sivertsen *et al.* (1992) and Jaffe *et al.* (1995) as well as those summarized by Alexeyev (1993) are in good agreement, suggesting that the total annual deposition of Cu and Ni can reach a few hundred mg/m^2 in the direct vicinity of the smelter stacks and decrease to a few mg/m^2 within a few tens of kilometers. An example of Ni deposition on snow in the Kola Peninsula in April 1990 is presented in Figure 7·28 (next spread) (Sivertsen *et al.* 1992). Wet deposition is far greater than dry deposition in the area.

Rainwater composition was measured at eight Arctic catchment areas in northern Europe: four in Russia, three in Finland, and one in Norway (Reimann *et al.* 1997). Close to industrial sources in Russia, most of the heavy metals measured (e.g., Co, As, Cu, Mo, Ni, Sb) show enrichment of two to three orders of magnitude in their median levels, compared with background levels measured in Finland.

The 1980s deposition of heavy metals on the Kola Peninsula was at least one order of magnitude higher than the deposition in the 1960s (in Kryuchkov and Makarova 1989),

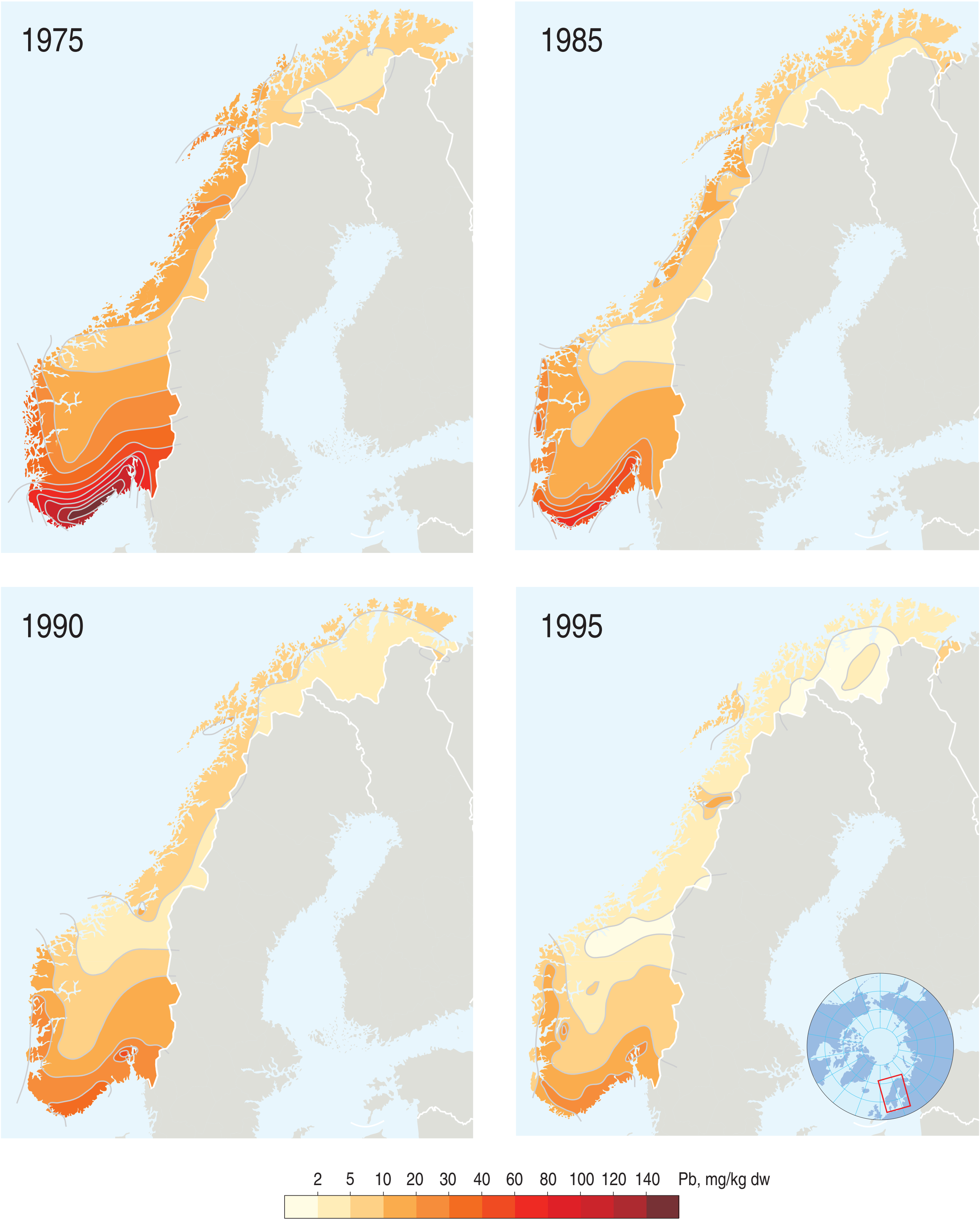


Figure 7·27. Latitudinal gradient of deposition of Pb in Norway in different years, as reflected by Pb concentrations in moss. (After Steinnes *et al.* 1994, Steinnes 1995).

with the major change occurring in the 1970s. The deposition trend over the last 30 years in the region reflects the emission trend discussed earlier in this chapter.

The levels of annual deposition of Ni, Cu, and other heavy metals in northern Finland, reported by Selin *et al.* (1992), are at the level of a few mg/m^2 and agree well with measurements on the Kola Peninsula outside the immediate area of the smelter stacks (Figure 7·28). Deposition of heavy metals in northern Scandinavia, also affected by emissions from smelters on the Kola Peninsula, is similar to the deposition in southern Scandinavia resulting mostly from the long-range transport of pollution from emission sources in Europe (Juntto 1992). Lead seems to be an exception, showing higher deposition in the south.

Figure 7·28. Nickel deposition to snow on the Kola Peninsula in April 1990. (After Sivertsen *et al.* 1992).

7.6.2. Terrestrial ecosystems

The data upon which this section is based are provided in Annex Tables 7·A1-7·A4.

7.6.2.1. Soil

Somewhat limited data for metals in soils are available for Greenland, Norway, Sweden, and Russia. General information on concentrations of Pb, Cu, Cr, Ni, V, and Zn in the soils of northern Fennoscandia are summarized in the Geochemical Atlas of Northern Fennoscandia (Geological Surveys of Finland, Norway and Sweden 1986). Figure 7·29 shows an example of available information for Norway. Anomalously high concentrations of these metals are noted; these correlate with known geological provinces. The range of natural concentrations can be up to two orders of magnitude.

Large amounts of data have been collected in the Kola Peninsula and in neighboring areas of Fennoscandia in an attempt to quantify the effects of emissions from the metallurgical complexes in the area (Pechenganikel in Nikel and Zapolyarnyy, and Severonikel in Monchegorsk) and to study the mobility of metals in soils in areas influenced by acidification. The principal contaminants studied are Cu and Ni. Barkan *et al.* (1993) and Evdokimova and Mozgova (1993a, 1993b) measured Cu in various soil horizons in areas up to six kilometers from the Severonikel smelter over the period 1976 and 1986. Concentrations were highly variable (30-4262 µg/g) and indicate that even in an area receiving exceedingly high loadings, some soil samples reflected near-background concentrations.

Contamination by both metals is clearly detectable throughout the Kola region and, as already noted, is particularly evident in the water and sediments of aquatic environments within 30 kilometers of the smelters. Soil samples at Karasjok, Norway are clearly and heavily contaminated by Cu (300-7400 µg/g). The overall effects attributable to the smelters, constructed approximately 50 years ago, are devastating. The cause is a combination of gaseous emissions causing acid rain and a huge increment to the aerial deposition of metals. Kryuchkov (1991) notes 'industrial deserts', entirely or almost entirely void of vegetation, surrounding the smelter towns for hundreds of square kilometers. According to the 1989 State of the Environment Report for Russia (State Committee for the Protection of Nature 1991) the Kola Peninsula qualified as one of eight most seriously polluted eco-catastrophe areas of the former Soviet Union.

Låg *et al.* (1970) have described the situation at Karasjok as an example of natural heavy-metal-poisoned soil (and vegetation). This phenomenon is thought to be fairly common in areas where sulfide mineralization occurs in bedrock. The 'poisoning' results when heavy metals are extracted from crystalline mineralization in bedrock and overburden, then transported downslope in groundwater solution, and finally reprecipitated in humus-rich soils where the solutions containing heavy metals emerge at the surface (Bølviken *et al.* 1977). Areas affected by this phenomenon often led to a patchwork pattern where barren soil patches range in size from 10^2 to 10^3 m^2. Bølviken *et al.* (1977) have shown that such poisoned areas can be mapped using the LANDSAT multispectral scanner system.

Metal concentrations in Greenland soils from four remote locations were: <12-37 µg/g, <12-13.8 µg/g, 8-13 µg/g, <0.04-0.10 µg/g, and <0.01-0.03 µg/g for Cu, Zn, Pb, Cd, and Hg, respectively. These concentrations probably represent background conditions. Results for Russian soils in Yamal, Taimyr, and the Lena Reserve appear much lower by comparison. Part of the explanation for this difference may be natural. Another factor is analytical bias: results of the joint Norwegian-Russian laboratory intercalibration (Akvaplan-niva 1996) suggest that the Russian metal data could be low by 30-50%. Notwithstanding analytical variability,

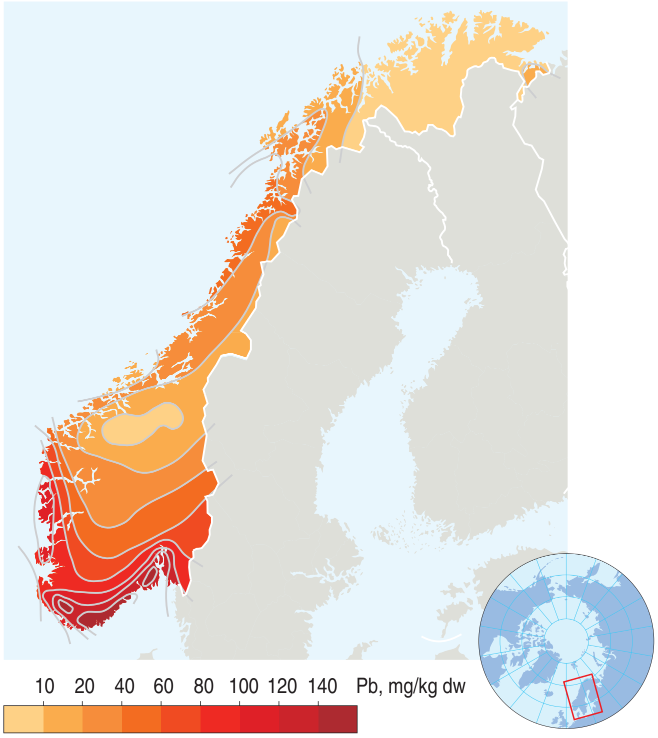


Figure 7·29. Lead concentrations in soil in Norway in 1975.

Table 7·9. Comparison of concentrations of metals in Arctic soils with soil quality guideline values. 'Most stringent guideline value' is derived from Annex Table 7·A17.

Metal	Most stringent guideline value, µg/g dw	USA (Alaska)	Canada	Greenland	Iceland	Norway	Sweden	Finland	Russia
Pb	25	n.d.	+	+	n.d.	+	n.d.	n.d.	n.d.
Cd	0.35	n.d.	++	+	n.d.	+++	n.d.	n.d.	++
Hg	0.1	n.d.	n.d.	+	n.d.	n.d.	n.d.	n.d.	+++
Se	0.7	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

+++ Virtually all reported values exceed the guideline. ++ Some (up to 50%) reported values exceed the guideline. + No reported values exceed the guideline.

it is probably valid to compare the 1994 and 1995 Russian data as they likely comprise an internally consistent dataset. These data suggest no change in the metal content of soils in Taimyr and Yamal as the differences in the values from 1994 to 1995 are generally less than the range of analytical error. When the available circumpolar data are compared with the most stringent soil quality guideline values for metals in soil (Table 7·9) (BKH Consulting Engineers 1995), the frequency of exceedence occurs in the order Cd > Hg > Pb.

The background composition of soils is affected primarily by bedrock geology, the hydraulic environment, weathering processes, and biological processes. Deposited metals are distributed through the soil-water systems and trapped in the soil profile with varying retention times according to their physico-chemical properties. The mobility of metals in the soil-water system is the result of a complex interaction of many processes. For many metals such as Al, Cd, and Zn, acidification of soils and waters is by far the most important factor regulating the concentrations and transport of metals from soils to running waters (Johansson *et al.* 1991). In contrast to most other metals, the solubility of Hg in soils and waters does not increase with decreasing pH. The adsorption of Hg on humic matter actually increases at lower pH values (Lodenius 1987).

The bioavailability of metals is related to chemical speciation. The divalent form of metals is usually the most readily bioaccumulated because of its mobility across cell membranes. Consequently any factor which favors the enhancement of divalent metal cations in the terrestrial/aquatic system will lead to greater bioaccumulation. In the Arctic, the most important factor in this regard in soil systems is acidification (pH). Other important factors include redox potential, presence of complexing ligands and competing ions, and temperature.

7.6.2.2. **Microorganisms**

No data were found for the metal content of microorganisms in the Arctic terrestrial ecosystem.

7.6.2.3. **Vegetation**

The most abundant data on the metal content of vegetation in the Arctic exists for mosses and lichens, particularly in Scandinavia.

Much of the data on lichens has been obtained in studies of the lichen→reindeer→wolf food chain. Data on the metal content of mosses are abundant because mosses have been identified as useful monitors of atmospheric deposition of metals, Figure 7·27. The technique was introduced in 1968 by Rühling and Tyler (1968) and is now being used routinely for large-scale deposition studies in several countries. The basis of the moss monitoring technique is that mosses lack a root system and therefore depend on surface uptake of chemical substances. They satisfy almost all their water and nutrient requirements directly from the air, and translocate them to their thin leaves. A dense moss carpet accumulates nearly all the mineral material deposited (Rühling *et al.* 1992).

A limitation of the technique is that factors other than air pollution contribute significantly to the element distribution observed in mosses. For instance, windblown soil dust deposited on the moss surface can be exceedingly difficult to remove prior to analysis (Steinnes and Jacobsen 1994). This can lead to a relatively large sampling variability and ultimately to highly variable results. These problems pose the greatest difficulty for interpreting and comparing results for metal loading with various receiving environments. It is not known to what extent surface contamination of mosses affects each dataset compiled in Annex Table 7·A2. In Table 7·10, the content of

Table 7·10. Metals in samples of the feather moss (*Hylocomium splendens*) collected in various regions of the Arctic.

Region	Year	Pb, µg/g dw	n	Cd, µg/g dw	n	Hg, µg/g dw	n	Se, µg/g dw	n
USA	1990-1992	0.35-1.6	21	< 0.03-0.98	21	< 0.016-0.112	–	–	–
Greenland	–	1.2-9.9	40	0.09-0.49	39	0.08-0.17	21	0.29-15.7	24
Norway									
– Svalbard	–	2.0-9.6	5	0.12-0.52	–	–	–	0.30-9.97	5
– North Norway	1977	15	–	–	–	–	–	0.37	–
– North Norway	1985	11	–	0.17	–	–	–	0.43	–
Russia		0.6-3.6	≅6	0.02-0.06	≅6	< 0.01-0.05	≅6	–	–

Table 7·11. Metals in samples of lichen collected in various regions of the Arctic.

Region	Species	Pb, µg/g dw	n	Cd, µg/g dw	n	Hg, µg/g dw	n	Se, µg/g dw	n
Canada									
– Bathurst	Unspecified	0.01-0.15	12	0.01-0.09	12	0.01-0.08	12	–	–
– Cambridge Bay	Unspecified	0.05-0.41	3	0.06-0.24	3	0.05-0.15	3	–	–
– Inuvik	Unspecified	n.d.-0.08	8	0.01-0.08	8	n.d.-0.04	8	–	–
Greenland	*Cetraria nivalis*	0.8-6.4	62	0.07-0.16	62	0.03-0.05	24	0.01-0.22	40
Northern Finland	*Hypogymnia physodes*	17.2	154	0.57	154	–	–	–	–
Russia									
– Taimyr	*Cetraria islandica*	0.57-1.5	2	0.15-0.18	2	0.03-0.05	2	–	–
– Taimyr	*Cetraria delisei*	0.80	1	0.08	1	0.02	1	–	–
– Various	Lichen/Lichenophyta	0.9-2.7	≥7	0.03-1.7	≥7	< 0.01-0.05	≥7	–	–

Pb, Cd, Hg, and Se in the species *Hylocomium splendens* are compared for samples collected in Alaska, Greenland, Norway, and Russia. Large ranges are inherent in the data and are in part related to sampling variability for the reasons noted above. Values for Russian samples appear to be low. This may be due to an analytical bias to underrecovery as noted in the results of the joint Norwegian/Russian laboratory intercalibration (Akvaplan-niva 1996). Differences in concentrations between samples collected in 1977 and 1985 in the Norwegian monitoring study (Steinnes *et al.* 1994) suggest no substantial temporal trend. Mäkinen (1994) measured the metal content of several moss species along two transects (one north-south, the other east-west) in northern Lapland and the Kola Peninsula. For each metal studied, the transects effectively mapped the zone of influence of emissions from various smelters and mining operations. Elevated values were found at distances of more than 100 km from the smelters. The highest values close to Monchegorsk were 263 times higher for Cu and 1328 times higher for Ni than the levels found in mosses in Finnish Lapland (background). Ranges of metals in mosses found along the transects were as follows: 0.1-0.95 µg/g, 1-70 µg/g, 5-1360 µg/g, 3-3100 µg/g, 2-25 µg/g, and 20-50 µg/g for Cd, Cr, Cu, Ni, Pb, and Zn, respectively.

Metals in various lichen species are compared in Table 7·11 for samples collected in Canada, Greenland, Finland, and Russia. The lowest values of Pb and Cd are reported for samples collected in Canada, and the highest for Finnish moss. The Hg content of lichens was similar (generally 0.01-0.10) for all regions.

Ford and Vlasova (1995 in press) measured Pb and Cu concentrations in the forage lichen *Cetraria cucullata* in Arctic Alaska and in the Taimyr Peninsula region of Russia north and south of the huge Cu-Ni smelter at Norilsk. The data indicate a zone of influence of about 200 km around Norilsk where Cu concentrations are approximately 1 to 2 orders of magnitude higher than those measured at locations 150 to 500 km north of Norilsk. Lead concentrations in *C. cucullata* were also higher to the south but the enrichment was only about a factor of 5-10 relative to the lichens from the northern sites. The data show that values for Pb and Cu are generally low in both Arctic Alaska and the Taimyr Peninsula relative to industrialized locations like Norilsk, but that samples of *C. cucullata* from Prudhoe Bay and along the Dalton Highway (the land transportation corridor between Fairbanks and the Prudhoe Bay oilfields) Alaska contain concentrations of Pb and Cu similar to those found in the industrialized regions of Siberia and western Russia.

7.6.2.4. **Terrestrial birds**

There is only a limited database on metal concentrations in terrestrial bird species in the Arctic. The absence of time series measurements on any given species and of any systematic spatial sampling generally precludes evaluation of Arctic-wide spatial and temporal trends. In general, concentrations in all birds decrease in the order kidney > liver > muscle.

The highest metal concentrations occur in grouse and ptarmigan in all areas, and the highest concentrations for these species occur in Canada (Annex Table 7·A3). The high concentrations of Cd in willow ptarmigan (*Lagopus lagopus*) kidney tissue of individual birds in Canada (1020 µg/g dw) and in Norway (121 µg/g dw) are notable. These are among the highest values for Cd reported for wild birds.

Wren *et al.* (1994) examined the concentrations of Cd, Cu, and Zn in willow ptarmigan from Arctic Norway and found that Cd concentrations were consistently three to ten times higher in the kidneys of adults than of juvenile birds from the same location. Zinc concentrations tended to be higher in adults as well but the difference was only about 10-30%. There was no difference in Cu concentrations between adult and juvenile willow ptarmigan. Wren *et al.* concluded that the bioaccumulated Cd was natural in origin.

RCMA (1996) provide summary data for Cd, Pb, and Hg in muscle and liver tissues of carnivorous, herbivorous, and omnivorous birds across Siberia. In muscle tissue, concentrations for all metals decrease in the order carnivores > omnivores > herbivores; in liver tissues there is no clear pattern. Some spatial trends are evident in the data. The muscle and liver tissues of herbivores contain more Pb in the east than in the west of Siberia. A similar trend occurs for Cd in the muscle and liver of omnivores, and for Cd and Pb in the liver tissues of carnivores.

Ptarmigan provide the most diverse dataset at the present time. The data summarized below (and in Table 7·12, and Figure 7·30 next page) provide comparative values for Pb, Cd, Hg, and Se in the willow ptarmigan and the rock ptarmigan (*Lagopus mutus*).

Willow ptarmigan tend to contain more Cd in kidney tissue than do rock ptarmigan. This observation has also been reported in these birds in Norway (Mykelbust *et al.* 1993, Pedersen and Myklebust 1993) and in Russia (RCMA 1996). Different accumulation patterns of metal uptake among different bird species (and even among individuals of the same species) are usually caused by differences in geological, di-

Table 7·12. Concentrations of Pb, Cd, Hg, and Se in willow ptarmigan (*Lagopus lagopus*) and rock ptarmigan (*Lagopus mutus*) for samples collected in the Arctic regions of the USA, Canada, Norway, and Russia. Figures in brackets shows number of samples. (Sources of data: see Annex Table 7·A3).

		Concentration µg/g, dry weight (except Russia, wet weight)											
		Lead			Cadmium			Mercury			Selenium		
Area	Species	Liver	Kidney	Muscle	Liver	Kidney	Muscle	Liver	Kidney	Muscle	Liver	Kidney	Muscle
USA, Alaska	Rock ptarmigan	<1-2 (18)	–	–	3-18 (18)	21-109 (18)	–	<0.02 (18)	–	–	0.48-1.3 (18)	–	–
Canada, Yukon Territory	Willow ptarmigan	0.08-1.5 (9)	0.1-7 (9)	–	20-122 (9)	9-1020 (9)	–	<0.05-0.18 (9)	<0.05-0.3 (9)	–	0.9-1.8 (9)	0.3-4 (9)	–
	Rock ptarmigan	0.08-2.8 (4)	0.9-176 (4)	–	7-54 (4)	28-887 (4)	–	<0.05-0.21 (4)	0.07-0.23 (4)	–	<0.05-1.4 (4)	1.5-4.9 (4)	–
Norway	Willow ptarmigan	0.43-1.47 (64)	0.45-0.9 (49)	–	0.75-9.1 (64)	46-121 (49)	–	0.019-0.046 (64)	0.062-0.093 (49)	–	0.48-0.93 (64)	–	–
Sweden	Willow ptarmigan	0.37-0.8 (9)	0.73-2.1 (9)	–	8-13 (9)	56-126 (9)	–	0.2-0.4 (9)	0.08-0.1 (9)	–	–	–	–
Russia	Willow ptarmigan	0.3 (1)	–	0.12 (1)	0.50 (1)	–	0.36 (1)	0.06 (1)	–	0.04 (1)	–	–	–
	Rock ptarmigan	<0.05 (1)	–	<0.05 (1)	0.05 (1)	–	0.05 (1)	0.04 (1)	–	0.02 (1)	–	–	–

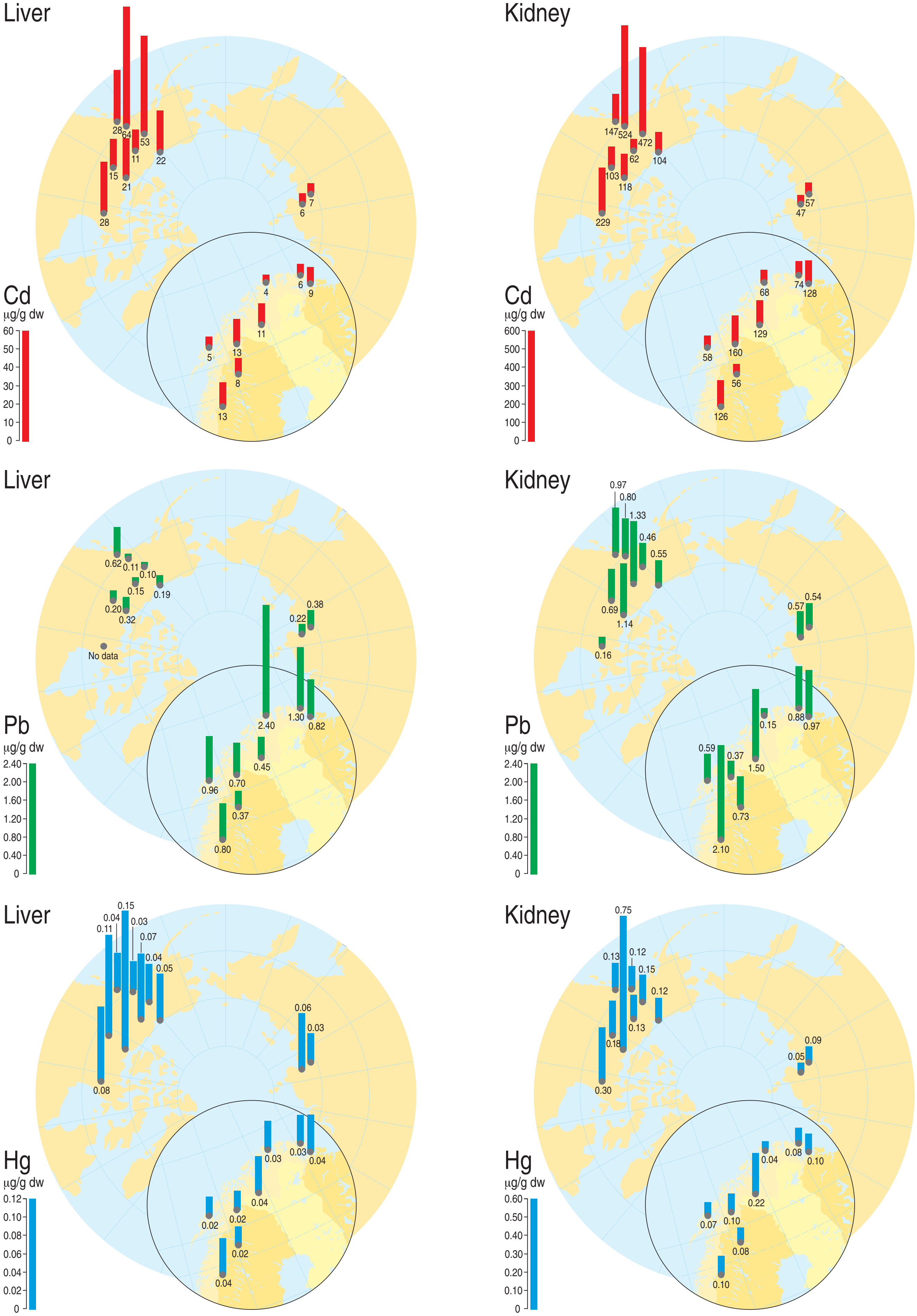


Figure 7·30. Circumpolar distribution of Cd, Pb and Hg levels in liver and kidney tissue of willow ptarmigan (*Lagopus lagopus*).

Table 7·13. Cadmium in liver, kidney, and muscle of *Rangifer tarandus* from various locations in the Arctic. (Sources of data: see Annex Table 7·A4).

Concentration µg/g, dry weight (except Finnish Lapland and Russia, wet weight)

	Liver			Kidney			Muscle		
Location	Range	n	Mean	Range	n	Mean	Range	n	Mean
Canada									
Finlayson Herd	3-64	49	11.4	51-882	50	121	–	–	–
Bonnet Plume Herd	1.1-8.3	21	3.2	7-108	21	38	–	–	–
Porcupine Herd	1.1-15.2	81	4.6	7-176	80	41	–	–	–
Tay Herd	1.7-19.6	20	9.6	44-473	20	121	–	–	–
Northwest Territories, Bathurst Herd	0.05-2.7	20	1.96	5-26	20	9.68	–	–	–
Arviat	1.2-6.6	10	3.69	10-63	10	33.9	–	–	–
Southampton Island	–	–	–	12-45	10	18.8	–	–	–
Cape Dorset	0.7-4.7	10	2.24	4-24	10	14.1	–	–	–
Lake Harbour	1.1-7.9	10	3.88	5-58	10	32	–	–	–
Inuvik, NWT	1.1-11	10	5.83	6-89	10	42.7	–	–	–
Beverly, NWT	2.1-4.7	10	3.42	14-59	10	31	–	–	–
Cambridge, NWT	0.6-4	10	1.35	4-19	10	9.4	–	–	–
Taloyoak, NWT	0.68-1.5	10	1.06	5-12	10	7.4	–	–	–
Pond Inlet, NWT	0.3-1.6	10	0.98	10-19	10	14.5	–	–	–
Denmark/Greenland	0.32-2.33	13	–	–	–	–	0.001-0.009	14	–
Finnish Lapland									
Southern Lapland	0.190-0.402[a]	60	–	0.525-1.72[a]	90	–	< 0.001-0.002[a]	59	–
Western Lapland	0.233-0.758[a]	60	–	0.650-4.62[a]	90	–	< 0.001-0.003[a]	62	–
Eastern Lapland	0.388-0.958[a]	51	–	0.938-4.25[a]	97	–	< 0.001-0.003[a]	66	–
Northern Lapland	0.310-0.546[a]	48	–	1.03-2.84[a]	48	–	0.002-0.006[a]	30	–
Norway	0.1-1.7	52	0.4	0.3-10	52	1.5	–	–	–
Russia	0.05-0.08[a]	5	–	0.05-0.45[a]	4	–	<0.02-0.10[a]	8	–
USA	–	–	–	38-61	3	51.5	–	–	–

a. Range of means, µg/g ww.

etary, and physiological factors. Myklebust *et al.* (1993) measured Cd in the food of willow and rock ptarmigan. The Cd content of items in the willow ptarmigan diet (willow shrubs, *Salix* spp.; and birches, *Betula pubescens*, *B. nana*) was generally higher than that of the rock ptarmigan diet (more crowberries, bilberry, and mountain avens), suggesting diet as an important factor in explaining the difference in body burden between willow and rock ptarmigan.

Pedersen and Myklebust (1993) showed that there is no net accumulation of Cd in the kidney or liver of ptarmigan after their first winter, suggesting that once a threshold is attained, uptake and excretion of Cd are balanced. A major mechanism for excretion of Cd could be through shedding of feathers during molting. (It is known, for example, that a significant excretion mechanism of Hg is through feather molting (Braune and Gaskin 1987)). The evidence provided by Pedersen and Myklebust (1993) supports this. They note that ptarmigan molt 3-4 times per year and replace 100-120 g feathers which amounts to 20-25% of their body weight. Furthermore, in willow ptarmigan, the lowest levels of Cd in juvenile birds occur during the spring/summer molt (2-3 months), whereas the build-up of Cd in the liver and kidneys is seen during winter when the ptarmigan have had the same plumage for eight months.

The Nordic Council of Ministers (1992) has proposed maximum Cd concentrations for the kidney, liver, and muscle tissue of meat (pigs and cattle) for human consumption of 1 µg/g, 0.5 µg/g, and 0.05 µg/g (ww), respectively. By this measure, most birds in Annex Table 7·A3 exceed the guidelines.

7.6.2.5. **Mammals**

The most comprehensive data set for metals in Arctic mammals exists for caribou/reindeer (*Rangifer tarandus*). To lesser extents both spatially and temporally, measurements are compiled for a variety of other mammals in Annex Table 7·A4. The data set for *Rangifer tarandus* is large because this species is known to bioaccumulate high concentrations of Cd in liver and kidney tissue and consumption of these tissues may constitute a human health risk (Crête *et al.* 1989). There is no published evidence, however, that demonstrates that the Cd burdens found in caribou liver or kidney constitute a health risk to the animals themselves. Elliot *et al.* (1992) and Nicholson *et al.* (1983) report that significant kidney dysfunction occurs for most mammals and birds when Cd concentrations exceed 100-200 µg/g ww (approximately 400-800 µg/g dw). Values have been reported at these levels for caribou kidney in the Arctic only infrequently.

Concentrations of Cd in caribou tissues decrease in the order kidney > liver > muscle in all regions. There is also a strong positive correlation between Cd concentration in kidney and age in all regions (Gamberg and Scheuhammer 1994, Rintala *et al.* 1995, Frøslie *et al.* 1986). The correlation is much weaker in the case of Cd in liver, and a leveling off of Cd concentrations tends to occur in older animals (> 10 years). Gamberg and Scheuhammer (1994) noted also that concentrations of Cd in liver and kidney of caribou tended to be higher in samples collected in spring than those collected in autumn. If a conversion factor from wet weight to dw is four to one, then the Canadian, Finnish Lapland, Norwegian, and Greenland data fall within similar ranges. The Russian data appear to be low by comparison. It may be that the Russian data are low due to the analytical differences discussed above (Akvaplan-niva 1996). Frøslie *et al.* (1986) also report on Svalbard reindeer based on the unpublished data of Norheim *et al.* There, median Cd concentrations in liver and kidney were 0.6 µg/g ww (n = 55) and 3.3 µg/g ww (n = 44), respectively.

A summary of Cd concentrations in liver, kidney, and muscle tissue of various caribou herds is given in Table 7·13 (data extracted from Annex Table 7·A4). Data for the Canadian herds located in the Northwest Territories indicate that there are clear differences in levels of Cd in the tissues among herds. The variation in Cd concentrations in the animals is thought to be strongly linked to natural sources, namely that local vegetation (e.g., *Salix* spp.) becomes enriched with Cd when it grows in soils derived from Cd-rich mineral zones known to exist in the NWT and the Yukon (Muir *et al.* 1996). There is, however, no systematic geographical pattern in these differences. The values for Cd in kidney of caribou of the Yukon herds however, were higher and more variable than those of the NWT. In Norway, there

Figure 7·31. Changes with time in concentrations of Cd, Pb and Hg in liver and muscle tissue of reindeer (*Rangifer tarandus*) sampled in three districts in eastern Sweden. (Source of data: Mats Olsson pers. comm.).

is a striking (approx. 3-fold) increase in Cd burdens in the kidneys of reindeer from Arctic Norway to southern Norway (Frøslie *et al.* 1986). A similar spatial trend has been noted in Sweden. It is consistent with the gradients for acid precipitation (Overrein *et al.* 1980), atmospheric fallout (Hanssen *et al.* 1980), and the concentration gradients of Cd in Norwegian surface soils (Allen and Steinnes 1980). In Russia, the concentration of Pb in the muscle and liver of reindeer are generally higher in the east than in the west; there is no similar trend for Cd (Melnikov *et al.* 1996).

The only substantial temporal data set examining metal concentrations in caribou is provided by the Swedish Environmental Monitoring Programme (SEMP 1995) through which samples of reindeer have been collected continuously since the early-1980s in three districts along the Swedish easternmost mountain chain, encompassing the Saami villages of Gabna, Laevas, Girgas, Iran, Ran, Handölsdalen, and Mittådalen. Analysis of the accumulated data (see Figure 7·31) indicates that during the past ten years, Cd, Pb, and Hg concentration in the muscle and liver of Swedish reindeer show no significant log-linear or linear change. Furthermore, SEMP (1995) calculated that the number of years required to detect an annual change of 5% ranged between 10 and 21 years depending upon element and tissue.

As noted above, the Nordic Council of Ministers (1992) has proposed a maximum concentration for Cd in kidney, liver, and muscle tissue of meat (pigs and cattle) for human consumption of 1 µg/g, 0.5 µg/g and 0.05 µg/g (ww), respectively. According to this proposed guideline, all caribou in Canada would fail. Far fewer exceedances would occur for Finnish reindeer. Rintala *et al.* (1995) note that Cd concentrations in the muscle samples of reindeer from Lapland were at the same level as those in muscle samples of Finnish pigs and cattle. The same trend has been seen for Pb and Hg in muscle tissue of Finnish reindeer.

Lead and Se in caribou liver, kidney, and muscle tissues generally fall within the ranges considered to represent background conditions. Mercury is high in Canadian animals and is generally attributed to natural geological sources in the Canadian Shield. Partitioning of metals among the various tissues varies with metal. For example, Hg tends to decrease in the same order as does Cd (kidney > liver > muscle), whereas Pb generally decreases in the order liver > kidney > muscle.

Among the other terrestrial mammals tested, the greatest enrichment of metals, particularly for Cd, occurs in moose (*Alces alces*). Extremely high values (up to 1380 µg/g dw) have been reported in the Yukon. In Norway, moose kidneys and liver were also enriched with Cd (max. value 19 µg/g ww (kidney), and 3 µg/g ww (liver)) and showed the same north-south gradient as did reindeer. Frøslie *et al.* (1985) point out that the same geographical differences for liver Cd concentrations are seen in lambs grazing in natural pastures.

Among the smaller mammals, examination of the data in Annex Table 7·A4 led to the following observations:

1. Lead, Hg, and Se concentrations tend to be low compared with those reported for similar tissues in reindeer and moose.
2. Some high kidney Cd values occur for beaver (130 µg/g), porcupine (326 µg/g), snowshoe hare (*Lepus americanus*) (166 µg/g), ground squirrel (538 µg/g), and european or mountain hare (*Lepus timidus*) (99 µg/g). Most of the high values occur in animals from the Yukon. Some high values are seen in hares in Norway. The high values probably are related to the diet of the animals, most of which have feeding preferences for and a corresponding accessibility to vegetation that accumulates Cd.
3. Data for the lichen→reindeer→wolf food chain indicate no biomagnification of Cd, Pd, Hg, or Se.
4. The data for Russia again appear to be the lowest of all. The reason(s) may be related to analytical biases as noted above.

The ecological significance of the high Cd concentrations measured in the kidney of various mammals is currently un-

Table 7·14. Occurrence of cadmium concentrations exceeding 100, 200, 600, and 800 µg/g (dw) in the kidney tissue of mammals and birds of selected regions of the Arctic. Y indicates Yukon.

Species/area	n	n >100 µg/g	n >200 µg/g	n >600 µg/g	n >800 µg/g	Max. value µg/g
Caribou						
Yukon						
– Bonnet Plume Herd	22	1	0	0	0	108
– Finlayson Herd	52	30	9	2	1	882
– Porcupine Herd	76	3	0	0	0	176
– Tay Herd	20	10	23	0	0	473
Alaska						
– 40 Mile Herd	3	0	0	0	0	61
NWT						
– Beverley Herd	10	0	0	0	0	59
– Bathurst Herd	20	0	0	0	0	26
– Arviat	10	0	0	0	0	63
– Southampton Island	10	0	0	0	0	45
– Cape Dorset	10	0	0	0	0	24
– Lake Harbour	10	0	0	0	0	58
– Inuvik	10	0	0	0	0	89
– Cambridge Bay	10	0	0	0	0	19
– Taloyoak	10	0	0	0	0	12
– Pond Inlet	10	0	0	0	0	19
Norway	204	0	0	0	0	18
Finland	325	0	0	0	0	15
Russia	5	0	0	0	0	4
Wolf						
Yukon	21	0	0	0	0	22
NWT	30	0	0	0	0	8
Moose, Y	51	35	10	2	1	1380
Lynx, Y	1	0	0	0	0	6
Red fox, Y	1	0	0	0	0	1
Marten, Y	7	0	0	0	0	8
Mink, Y	3	0	0	0	0	0.5
Hoary marmot, Y	1	0	0	0	0	46
Muskrat, Y	13	0	0	0	0	8
Short-tailed weasel, Y	8	0	0	0	0	7
Dall sheep, Y	4	0	0	0	0	30
Mountain goat, Y	4	0	0	0	0	16
Porcupine, Y	6	4	1	0	0	326
Snowshoe hare, Y	29	1	0	0	0	155
Ground squirrel, Y	19	1	1	0	0	538
Red squirrel, Y	26	1	0	0	0	155
Beaver, Y	14	5	4	0	0	256
Spruce grouse, Y	38	2	2	1	0	760
Ruffed grouse, Y	7	3	2	1	1	1020
Blue grouse, Y	3	0	0	0	0	11
Willow ptarmigan, Y	9	7	5	4	4	1020
Rock ptarmigan, Y	4	1	1	1	1	887
White-tailed ptarmigan,Y	2	2	0	0	0	161
Misc. ptarmigan, Y	6	5	3	0	0	232
Mallard duck, Y	2	0	0	0	0	4
Scaup, Y	2	0	0	0	0	9

known. No clearly demonstrated effects on wildlife in the Arctic related to Cd burdens have ever been reported. It is thought, however, that concentrations of Cd of 200 µg/g ww (approx. 600 µg/g dw) in kidney tissues of birds and mammals are capable of causing renal dysfunction (Nicholson *et al.* 1983). Long-term exposure to lower concentrations is perhaps capable of causing a number of sublethal effects.

In Table 7·14, the data for Cd in kidney tissues are arranged to show the number of samples exceeding 100 µg/g, 200 µg/g, 600 µg/g, and 800 µg/g (dw) for all data in Annex Table 7·A4. The 100 µg/g level identifies animals significantly above circumpolar ‘background’ concentrations; the 600 µg/g level identifies animals possibly at risk of kidney dysfunction; the 800 µg/g level identifies animals having extreme body burdens of Cd.

The data clearly indicate that the greatest bioaccumulation of Cd in animals occurs in caribou, moose, squirrels, grouse, and ptarmigan of the western Canadian Arctic (Yukon). This is opposite to the trend noted for the marine ecosystem (see section 7.4.4.8). It indicates that local natural sources of Cd probably are the primary factor in controlling bioaccumulation in the terrestrial environment of the Canadian Arctic.

7.6.3. Freshwater ecosystems

7.6.3.1. Metals in freshwater

Data on the background metal content of freshwater in the Arctic are very limited both temporally and geographically. Erickson and Fowler (1987) report data for 1985 and 1986 in various channels of the Mackenzie River Delta during winter. Mean values were approximately 1.1-1.4 µg/L, 0.5-2.5 µg/L, 0.15-0.99 µg/L, 10-123 ng/L and 5-8 ng/L for Cu, Zn, Pb, Cd, and Hg, respectively. Average values published for the same metals during the period 1960-1974 (Environment Canada 1981) were 5, 8, 2, 1, and 0.005 µg/L, respectively. Rovinsky *et al.* (1995) report values for Russian rivers in generally the same ranges as noted for the Mackenzie River, namely 0.3-3 µg/L, 1.8-2.6 µg/L, 0.15-0.99 µg/L, 20-290 ng/L, and 20-66 ng/L for Cu, Zn, Pb, Cd, and Hg, respectively. Recent data for 11 rivers in Arctic Canada indicate average concentrations of <1-2 µg/L, <1-2 µg/L, <0.7-1.3 µg/L, <0.2 µg/L and <0.2 µg/L for Cu, Zn, Pb, Cd, and Hg, respectively (Jeffries and Carey 1994). Much higher values are reported for lakes and ponds of the Arctic National Wildlife Refuge, Alaska) (Snyder-Conn and Lubinski 1993) (Annex Table 7·A7). The values are suspect because even the detection limits noted in the data usually exceed the maximum values measures for metals in other Arctic freshwaters.

There are numerous examples of localized enrichment of metals in freshwater in the Arctic. Most of these enriched areas are associated with mining operations. Garrow Lake, NWT is used as a tailings pond at a Pb-Zn mine in the Canadian Arctic and concentrations of Cu, Zn, Pb, Cd, and Hg are typically in the order of 28 µg/L, 360 µg/L, 1.8 µg/L, 0.7 µg/L and 0.03 µg/L, respectively (INAC 1994). In Arctic Russia and the Kola Peninsula, such mining operations have caused enrichment of metals in the freshwater ecosystem on a much larger scale. Values exceeding 10 µg/L Cu and Ni generally occur to about a 30 km radius around metallurgical complexes in the Murmansk Region but effects of these metals in aquatic systems is easily detected hundreds of kilometers from the source (ACOPS 1995). The highest-recorded concentrations of Cu and Ni in waters of the Murmansk Region are summarized in Table 7·15. During the period 1991-1994, Cu attained concentrations of 11-2524 times the maximal allowable concentration (MAC), and Ni 2-135 times the MAC. The ecosystems of at least five water bodies are considered to be completely destroyed. Similar severe and widespread contamination of water bodies has been reported for the Norilsk region, where intensive metallurgical activities also occur.

Data for metals in lakes of northern Scandinavia are generally within the lower end of the range reported here for rivers (Annex Table 7·A7).

Table 7·15. Cu and Ni contamination of water bodies in Murmansk Region (Hydrochemical Institute 1992, 1993, 1994, 1996).

		Highest recorded level, µg/L[a]			
Water Body	Metal	1991	1992	1993	1994
Kolos-Yoki River, mouth	Cu	47	14	29	27
	Ni	102	60	195	53
Luotn-Yoki River	Ni	56	38.5	32	17
Hayki-Lampi-Yoki River	Ni	32	43	24	24
Nyuduay River	Cu	2524	300	168	518
	Ni	1347	409	465	400
Monche Lake	Cu	225	260	176	113
Imandra Lake (Monche-Guba)	Cu	105	35	20	11
	Ni	195	6	37	5

a. The maximum allowable concentrations (MAC) for these metals are: MAC_{Cu} = 1 µg/L; MAC_{Ni} = 10 µg/L.

Table 7·16. Comparison of concentrations of metals in Arctic freshwaters with water quality guideline values. 'Most stringent guideline values' are from Annex Table 7·A17.

Metal	Most stringent guideline value, µg/L	USA (Alaska)	Canada	Greenland	Iceland	Norway	Sweden	Finland	Russia
Pb	0.1	++	+++	n.d.	n.d.	n.d.	n.d.	++	++
Cd	0.045	++	+	n.d.	n.d.	n.d.	n.d.	+	++
Hg	0.012	++	+	n.d.	n.d.	n.d.	+	n.d.	+
Se	5	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

+++ Virtually all reported values exceed the guideline.
++ Some (up to 50%) reported values exceed the guideline.
\+ No reported values exceed the guideline.

When the available metal data are compared with the most stringent water quality guideline value for metals in freshwater (Table 7·16), the frequency of exceedence occurs in the order Pb > Cd > Hg.

7.6.3.2. **Metals in freshwater sediments**

7.6.3.2.1. **River and lake bottom sediments**

Data for metals in freshwater sediments are compiled in Annex Table 7·A5, and include samples from the USA, Canada, Greenland, Norway, Finland, Sweden, and Russia. The greatest quantity of data have been produced in Scandinavia, where comprehensive studies have been completed to characterize rates of metal accumulation in forest lakes, to quantify the effects of long-range transport, and to describe the mobility of metals throughout forest ecosystems. Unfortunately, the number of samples analyzed and their geographical distribution are insufficient to be used to evaluate or describe spatial or temporal trends.

Within Scandinavia, the data show similar trends. Concentrations of Cu, Zn, Pb, Cd, and Hg can be highly variable even in adjacent water bodies. An important factor affecting metal concentrations is pH of the water body. In general, a decrease in pH results in a release of metals from sediments. In the case of Zn and Cd, it inhibits deposition because retention times for these metals in the water column are increased. Mercury behaves differently from the other metals. A decrease in pH usually decreases the solubility of Hg because the adsorption of Hg on humic matter appears to increase at lower pH values. The Hg adsorbed on to humic substances is not easily displaced.

Skotvold *et al.* (1996) studied heavy metals in sediments from 91 lakes in northern and Arctic regions of Norway. They concluded that Pb, Cd, Zn, and Hg deposition was primarily from long-range air transport sources, whereas Cu was the result of local sources. These conclusions were in agreement with the data reported by Steinnes and Henriksen (1993). On the northern Norwegian mainland, gradients for Pb, Cd, and Hg decreased south to north, while Cu and Zn had the opposite trend. The range of mean values for Cu, Zn, Pb, Cd, and Hg were 25-110 μg/g, 80-175 μg/g, 17-85 μg/g, 0.45-0.75 μg/g, and 0.06-0.20 μg/g, respectively. On Spitsbergen and Bear Island, the average concentrations of Pb (25-90 μg/g) and Cd (0.5-1.3 μg/g) were generally higher than those found on the northern Norwegian mainland, but the mean concentrations of Hg (0.062-0.098 μg/g) were lower.

Johansson (1989) reported similar trends for metals measured in 54 lakes in Sweden (14 were located within the AMAP region). Gradients for Pb, Zn, Cu, and Cd all decreased south to north. Concentrations in the surface layer were 3-25 μg/g, 5-40 μg/g, 20-65 μg/g, and 0.07-1.3 μg/g for Pb, Cu, Zn, and Cd, respectively. In the south, vertical gradients of the metals along cores suggested enrichment factors of about 50, 7, 4, and 2 for Pb, Cd, Zn, and Cu, respectively. In the north, Zn and Cu showed no vertical gradient and the enrichment of Pb and Cd in the surface sediment was only about a factor of 2.

Verta *et al.* (1989) provide profiles for Pb, Zn, Cd, and Hg in sediment cores from 18 small headwater drainage and seepage lakes in Finland. The trends are in general agreement with the data of Skotvold *et al.* (1996) and Johansson (1989), namely a gradient decreasing south to north.

Lockhart *et al.* (1995) provide data for Hg in dated cores from ten lakes in Arctic Canada. Recent fluxes of Hg varied from 5 μg/m^2/y to approximately 50 μg/m^2/y compared with fluxes of 0.7 to 31 μg/m^2/y estimated from the deepest portions of the cores. The enrichment factors for Hg (ratio of concentration of Hg at top of each core divided by that at the bottom) ranged from 1.1-7.0 (mean, 2.4). These values are consistent with data reported in temperate areas of North America. Furthermore, Mannio (1996) provides data for sediments in northern Finnish lakes where surficial Hg concentrations are 40-180 ng/g and enrichment factors are 1.8-2.2. Similar concentrations and trends have been noted in northern Norway and Sweden (e.g., Rognerud *et al.* 1993). Viewed as a whole, the data suggest a widespread and continuing input of Hg into Arctic sediments. Indeed, the results shown in Figure 7·32 for Finnish and Canadian sediment cores are remarkably similar despite the wide geographical separation between the sampling areas.

This may be related to the rather unique properties of Hg as a metal. Specifically, it is highly mobile, particularly in the gas phase, and is capable of being re-emitted from sediment and water. Successive deposition/re-emission cycles combined with decreasing average temperatures at higher latitudes could lead to enrichment of Hg in Arctic regions.

Mannio (1996) measured the accumulation of Cu, Zn, Pb, Cd, and Hg in four lakes located in Arctic Finland. The surface layers of each core were generally enriched relative to deeper layers and the maximum values usually occurred within the top 5 cm.

Rognerud *et al.* (1993) provide data on historical trends on accumulation rates of Cu, Cd, Co, Ni, Pb, and Zn for two Norwegian lakes (Dalvatn and Durvatn) from cores collected during a joint Norwegian – Russian expedition in 1992. The lakes are located downwind of Russian smelters. Rognerud *et al.* (1993) showed that the sediment accumulation rates for each lake were constant during the past 50-60 years. Consequently, any changes in the rate of accumulation of individual metals should be attributable primarily to changes in atmospheric deposition. The ratio between the accumulation rates of metals in the most recent settled sediment and a reference sediment from the 1920s prior to the start of smelting operations indicate that the accumulation rates of Ni, Cu, and Pb have increased by factors of 2-4. The increase in Ni and Cu are attributed to the smelters as the deposition of these metals elsewhere in Norway are low (Rognerud and Fjeld 1990, Sivertsen *et al.* 1991). The increase in Pb accumulation rates are thought primarily to be related to the general increase of Pb in the atmosphere of the

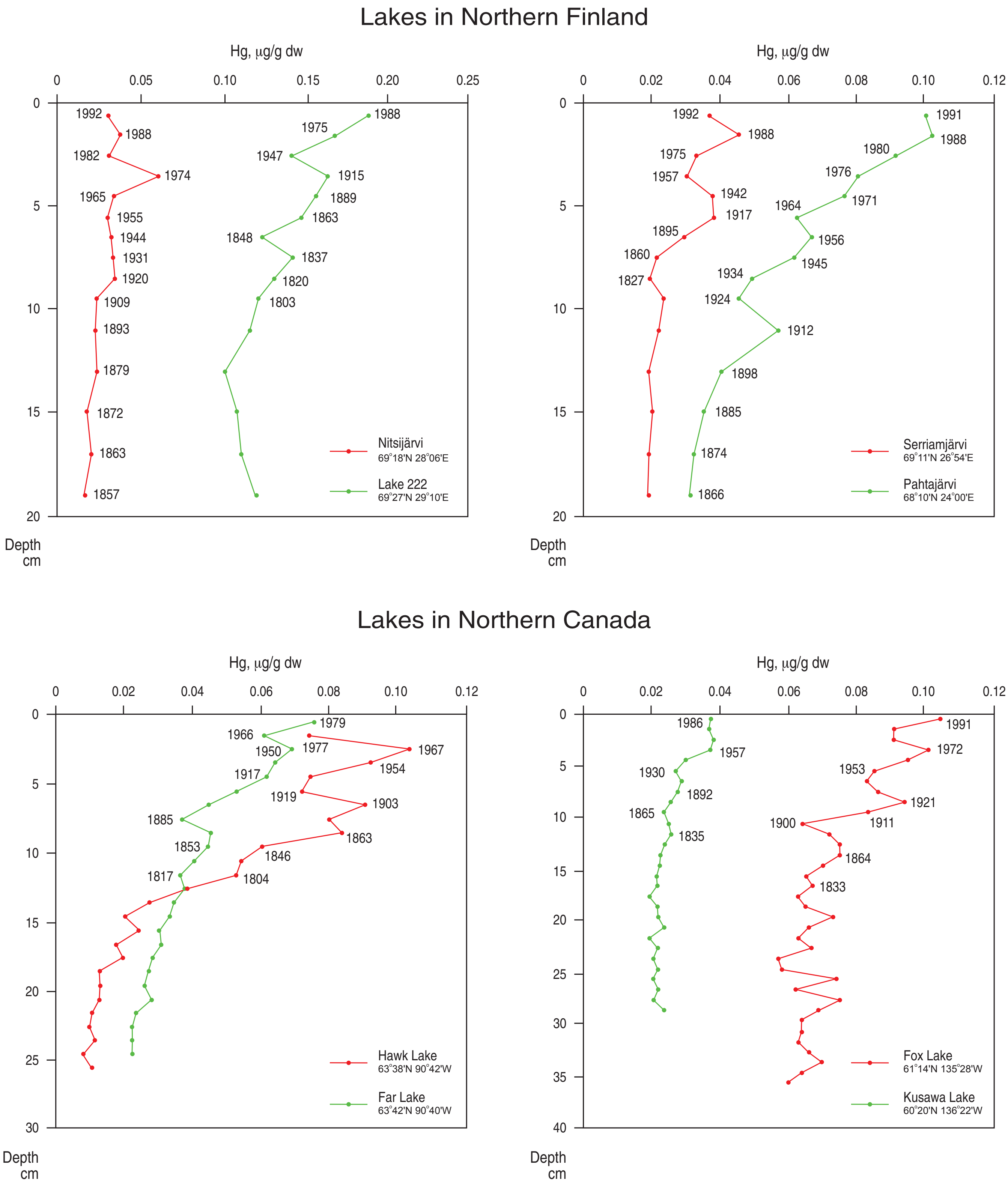


Figure 7·32. Concentrations of Hg in dated sediment cores from lakes in Arctic Finland and Canada. (Source of data: Lockhart *et al.* 1995, Northern Canada; Mannio 1996, Northern Finland).

northern hemisphere (Norto and Kahl 1991), but a small contribution is related to the smelters (Hagen *et al.* 1991). Rognerud *et al.* (1993) also conclude that there is no obvious contribution of atmospheric deposition of Cd and Zn from the smelters.

Dahl-Hansen and Evenset (1995) measured relatively low concentrations of Cu (4.4-12 µg/g), Zn (76-114 µg/g), Hg (0.026-0.092 µg/g), Cd (0.11-0.89 µg/g), and Pb (2.9-8.5 µg/g) in Lakes Nyulay, Kotyol, and Kapylty in Arctic Russia. Far greater pollution by metals is seen in areas of Arctic Russia influenced by metallurgical complexes such as those on the Kola Peninsula and around Norilsk. In these areas, the concentrations of Ni, Cu, Co, Cd, and Hg in the surface sediments of lakes located up to 40 kilometers from the source of pollution exceed background values by 10-380 times (ACOPS 1995). As lake sediments are excellent storage reservoirs for metals, enhanced metal concentrations are likely to exist in these areas for many decades.

The metal content of cores from four locations in Greenland (Riget *et al.* 1997c), four lakes in the Yukon Territory,

Table 7·17. Comparison of concentrations of metals in Arctic freshwater sediments with sediment quality guideline values. 'Most stringent guideline values' are from Annex Table 7·A17.

Metal	Most stringent guideline value, µg/g dw	USA (Alaska)	Canada	Greenland	Iceland	Norway	Sweden	Finland	Russia
Pb	15	++	++	++	n.d.	+++	++	+++	++
Cd	0.6	++	++	+	n.d.	++	++	++	+++
Hg	0.15	n.d.	+	+	n.d.	++	++	++	+
Se	1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

+++ Virtually all reported values exceed the guideline.
++ Some (up to 50%) reported values exceed t
+ No reported values exceed the guideline.

Canada (Lockhart and Muir 1996), and Lake Kyusyur, Russia (Rovinsky *et al.* 1995) all were similar and probably indicative of the range of natural background values (Annex Table 7·A5).

When the available metal data are compared with the most stringent sediment quality guideline value for metals in freshwater sediments (Table 7·17), the frequency of exceedence occurs in the order Pb > Cd > Hg.

7.6.3.2.2. **Freshwater particulates**

The two main datasets on suspended particulates/suspensions in freshwater systems (see Annex Table 7·A6) are provided by Erickson and Fowler (1987) (Mackenzie River) and Melnikov (1991) (four Russian rivers). Values for the Mackenzie River (44-77 µg/g, 134-225 µg/g, 17-32 µg/g, 0.57-0.68 µg/g, and 0.090-0.210 µg/g for Cu, Zn, Pb, Cd, and Hg, respectively) are somewhat higher than the values reported by Melnikov for the Mezen, North Dvina, Ob, and Pechora Rivers (3-28 µg/g, 19-50 µg/g, and 4-29 µg/g for Cu, Zn, and Pb, respectively). There is no obvious reason for the difference, but it is probably related to the particle size of the suspended fraction, its organic content, sample size, and sampling methods. The data for the Russian rivers seem to be low in that the values are much lower than the river sediments themselves. Given that the metal content usually varies inversely with particle size and that suspended sediments are usually finer or no coarser than riverbed sediments, this is somewhat puzzling.

The anthropogenic contribution of metal concentrations in the particles of riverine suspended matter can be evaluated by using the concept of an enrichment factor (EF) in which the ratio of metals in suspended matter of a given river is compared with the ratio of the same metals in suspended matter of global rivers:

$$EF = \frac{(Me/Sr)_{susp}}{(Me/Sr)_{susp.global}}$$

The elements Sr, Al, or Li, which exist almost entirely as a part of mineral crystal structure, are taken as reference elements (Thomas and Martin 1982). By using this index and a large number of samples, the waters of most Russian rivers fall within global natural variability Gordeev and Tsirkunov (in press).

7.6.3.2.3. **River heavy metal fluxes**

An assessment of heavy metal fluxes by Eurasian rivers to the Arctic Ocean has been completed in recent years, to a considerable extent as the result of the International Programme 'SPASIBA'. The research (Cossa and Coquery 1993, Martin *et al.* 1993, Kravtsov *et al.* 1994, Coquery *et al.* 1995, Gordeev and Shevchenko 1995, Guieu *et al.* 1996, Rachold 1995) showed that downstream sections of the major Siberian rivers are characterized by low concentrations of dissolved and particulate forms of heavy metals. Dai and Martin (1995) showed that colloidal material (10^4 Dalton – 0.4 µm) made a significant contribution to the so-called 'dissolved' fraction of heavy metals in the Ob and Yenisey rivers (20-50% Pb, 40-70% Cd and Ni, up to 70-80% Cu). The authors conclude that there is a fundamental role of colloidal fractions in determining the behavior of trace metals in the estuarine zone as well as in the control of their net discharge into the Kara Sea. A comparison of some dissolved and particulate inputs by the Ob, Yenisey, and Lena rivers into the Kara and Laptev Seas is presented in Table 7·18. As data concerning seasonal changes in concentrations are absent, the values in this table should be considered qualitative only.

A comparison of gross and net river fluxes of dissolved metals to the Eurasian Basin (Guieu *et al.* 1996) and of fluxes from the other sources to the Arctic Ocean (inflow with saline waters, organic matter decomposition) are shown in Table 7·19 (Gordeev and Tsirkunov in press). The data show that net river flux exceeds gross river flux 5-6 times for Cd, 2.5 times for Ni and 1.5 times for Cu mainly due to the desorption from particles in estuarine zones. Net and total fluxes of Zn were approximately equal. The data also show that riverine input of heavy metals into the Eurasian sector of the Arctic Ocean appears to be rather small compared with other sources (Cd, 4%; Cu, 27%; Ni, 11%; Zn, 2%). Similar data for North America was not available for the assessment.

Table 7·18. Riverine fluxes of dissolved and particulate heavy metals to the Arctic Ocean (t/y) [a].

River	Cu	Pb	Zn	Ni	Cd	As	Hg	Fe×10^3
Ob								
Dissolved	850	6	160	530	0.3	–	0.2	12
Particulate	840	260	1700	630	3.3	–	0.8	940
Yenisey								
Dissolved	1000	4	820	340	1.0	–	0.2	10
Particulate	650	180	1300	450	13.6	–	0.3	320
Lena								
Dissolved	300	9	180	160	2.8	80	0.4	12
Particulate	490	400	2500	550	4.6	–	4.0	590

a. Ob and Yenisey: Dissolved metals (except Hg) – Dai and Martin 1995, Kravtsov *et al.* 1994; particulate metals (except Hg) – Gordeev *et al.* 1995. Lena: Dissolved and particulate metals (except Hg) – Martin *et al.* 1993. Dissolved and particulate Hg in all rivers – Cossa and Coguery 1993, Coguery *et al.* 1995.

Table 7·19. Gross river flux and net river flux of dissolved heavy metals to the Eurasian (EA) Basin with other inputs to the Arctic Ocean (mol/y).

Metal	Gross river flux to EA Basin[a]	Net river flux to EA Basin[b]	Other fluxes to Arctic Ocean[c]	Total fluxes to Arctic Ocean[b]	Riverine input as % of total
Cd	2.0×10^5	1.1×10^6	2.9×10^7	3.0×10^7	4
Cu	5.0×10^7	7.4×10^7	2.0×10^8	2.7×10^8	27
Ni	1.6×10^7	4.4×10^7	3.6×10^8	4.0×10^8	11
Zn	4.4×10^6	4.4×10^6	2.7×10^8	2.8×10^8	2

a. Gordeev and Tsirkunov submitted; b. Guieu *et al.* 1996;
c. Yeats and Westerlund 1991.

7.6.3.3. **Microorganisms**

No data were found for the metal content of microorganisms in the Arctic freshwater system.

7.6.3.4. **Algae and plants**

Very few data have been collected on the metal content of aquatic plant species in the Arctic. Iivonen *et al.* (1992) report on the Pb, Cd, Cu, Zn, and Ni content of two floating water-plant species, *Nuphar luteum*, and *Sparganium* sp., from three lakes in Arctic Finland. Concentration mean values are summarized in Table 7·20.

Table 7·20. Metal concentrations in two floating water-plant species, *Nuphar luteum*, and *Sparganium* sp., from three lakes in Arctic Finland (µg/g dw), (Iivonen *et al.* 1992).

Species	Pb	Cd	Cu	Zn	Ni
Nuphar luteum	0.1, 0.2	0.06, 0.09	1.35, 2.75	2.4-31	0.3-0.7
Sparganium sp.	0.57-4.48	0.15-1.14	2.2-5.6	35-128	0.8-2.0

7.6.3.5. **Metals in freshwater invertebrates**

Very few measurements have been made on the concentrations of metals in the lower food chain, including freshwater invertebrates. The few data that have been reported are summarized in Annex Table 7·A8. Most values for Pb, Cd, and Hg are at or near the detection limits.

7.6.3.6. **Fish**

Metal concentrations in various freshwater fish are compiled in Annex Table 7·A9. In general, there are few instances of comprehensive data for a given species that will allow comparison of values across the Arctic or as a time series. A possible exception is Hg, for which the largest set of values exists. This emphasis on Hg in the environment and in the food chain reflects the fact that it clearly bioaccumulates and that the effects Hg has on fish and on human consumers are well documented (see sections 7.5.3 and 12.2.3.1).

Table 7·21. Mercury concentrations in Arctic freshwater fish.

	Mercury concentration, µg/g wet weight					
Region	Arctic char (*Salvelinus alpinus*)	N	Whitefish (*Coregonus* spp.)	N	Burbot (*Lota lota*)	N
Northern Canada	0.01-0.57	226	0.01-2.49	102	0.11-0.30	4
Greenland	0.17-0.99	86	–		–	
Finnish Lapland	0.09-0.32	55	0.06-0.28	18	0. 23	8
Iceland	0.02-0.03	2	–		–	
Norway	0.03-0.25	39	n.d.-0.18	99	–	
Russia	0.01	2	0.01-0.11	60	0. 01	1
Sweden	0.10	?	0.28	?	–	

Data for Hg in the muscle tissue of Arctic char (*Salvelinus alpinus*), whitefish (*Coregonus* spp.), and burbot (*Lota lota*) are relatively consistent for all Arctic regions as shown in Table 7·21 (data extracted from Annex Table 7·A9).

The recent analysis of archived samples of pike (*Esox lucius*) muscle tissue in Sweden under the Swedish Environmental Monitoring Programme provides the most comprehensive time series for Hg in Arctic freshwater fish. A total of 237 samples of pike caught in 21 different years between 1968 and 1996 were analyzed (Annex Table 7·A9). Geometric mean values (weight adjusted) varied between 194 and 459 ng/g ww. No clear temporal trend is evident in the data (see Figure 7·33); variations around a mean for the entire dataset of 306 ng/g ww probably represent natural year-to-year fluctuations and analytical variability (Olsson pers.comm.).

General trends in the data for Hg in fish tissue include the following:

1. Highest concentrations tend to be in predatory fish such as pike and perch (*Perca fluviatilis*).
2. There is a tendency for concentration to correlate positively with length (age) for pike and perch and no correlation with whitefish and Arctic char.
3. The highest concentrations of Hg in fish are not necessarily correlated with the presence of high Hg concentrations in sediments. There are several reasons for this. First, Se is known to reduce the uptake of Hg by fish (and biota in general) so the Se content of sediments must be factored into any model involving correlation. Second, the concentration of calcium and magnesium affect the bioaccumulation of Hg by altering competition equilibria. Finally, pH and buffer capacity have an impact on the bioaccumulation of Hg by fish because humic substances can form strong complexes with Hg thereby effectively reducing Hg availability (bioaccumulation of metals other than Hg is usually enhanced in acidified low calcareous lakes).
4. The fish from Arctic Canada generally have the highest body burdens of Hg found in the Arctic. This is thought to reflect the widespread naturally elevated baseline for Hg in the Canadian Arctic and may be related to Hg-organic matter associations). Fish exceeding the guideline level of 0.5 µg/g for human consumption occur most frequently in Arctic Canada and Greenland (range of mean muscle values 0.17-1.0 µg/g (n = 86)).

The bioaccumulation of metals in fish is highly variable in the Arctic freshwater ecosystem, as it is in other ecosystems. The partitioning of metals among fish tissues varies with species, age, sex, and season. Lake or river chemistry, the presence of other contaminants, humic substances and other complexing agents, and feeding habits of the fish also play significant roles in the dynamics of bioaccumulation.

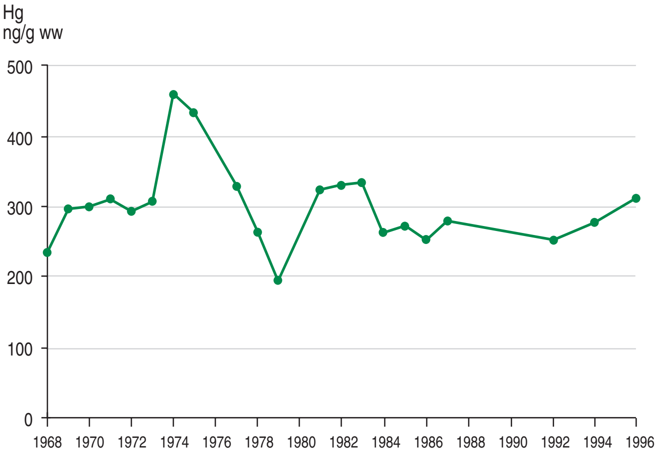


Figure 7·33. Changes with time in (weight adjusted, geometric mean) concentrations of Hg in muscle tissue of pike (*Esox lucius*) in Sweden. (Source of data: Mats Olsson pers. comm.).

7.6.3.7. Metals in aquatic birds

Based upon a limited number of measurements (Annex Table 7·A3), the bioaccumulation of metals by aquatic birds appears to be at least an order of magnitude less than that of terrestrial birds. The distribution among tissues is the same as for terrestrial birds, however: kidney > liver > muscle. There are too few data to delineate any spatial, temporal, or species trends. The largest dataset for metals in aquatic birds is for a group of ten species at various locations in Arctic Canada (NWT, Yukon, Quebec, and Labrador, CWS 1996). For Cd, Pb, and Hg, many values were at or near the detection limit. Ranges for values exceeding the detection limit in breast muscle tissue were as follows: Cd, 0.02-0.87 µg/g ww (n = 134); Hg, 0.2-1.93 µg/g ww (n = 158); and Se, 0.04-2.1 µg/g ww (n = 418).

7.6.3.8. Mammals

The data for metals in freshwater mammals consist of Pb, Cd, and Hg measurements in the hair of ringed seals living in Lake Saimaa, Finland (Hyvärinen and Sipilä 1984). Although Lake Saimaa is located just outside the AMAP assessment area, it is nonetheless relevant to include this dataset here because the boreal environment of this lake is similar to that for lakes in the AMAP area. Ringed seals exists in the Arctic and the data provide one of the very few observations of possible links between exposure to metals and biological effects.

Hair samples collected around 1983 had the following ranges of concentration means (n=32): 0.5-0.7 µg/g dw, 3.6-8.5 µg/g dw, and 3.2-20.7 µg/g dw for Cd, Pb, and Hg, respectively. According to data obtained for museum samples, the concentration of Hg in the hair of adult ringed seals was about the same as found by Hyvärinen and Sapilä (1984) in yearlings. In 1965 the concentration in hair was approximately 50 µg/g. The authors speculated that the sharp decline in the ringed seal population of Lake Saimaa during the 1960s and 1970s could be related to the insufficient availability of Se in the lake, which made the seals more susceptible to the toxic effects of Hg and premature (still) births.

7.6.4. Wetland ecosystems

Wetlands are a transitional compartment between the terrestrial and aquatic environments and play an important role in fate of contaminants, heavy metals in particular. In areas of strong anthropogenic impact, wetlands can serve as accumulators of heavy metals and as sources of significant secondary contamination of river waters. The survey of more than 250 wetlands in the Russian sector of the Arctic (Annex Table 7·A16) has shown that concentrations of heavy metals in wetland ecosystems are generally low, with the exception of the areas where intensive heavy industry occurs. Three areas with high levels of local heavy metal pollution of wetland ecosystems have been identified:
1) Kola Peninsula in the vicinity of non-ferrous smelters,
2) the Vorkuta area on the north of the Komi Republic, and
3) the Norilsk area in Central Siberia. In the same areas, increased concentrations of heavy metals in river waters have also been observed. Metal concentrations in remote parts of the same regions are close to background levels, with the exception of Far North-East Asia where increased concentrations of some metals including Hg are observed. This area is situated in a zone of mercury ore belts, and increased concentrations of Hg in this area are thought to originate primarily from natural sources.

7.6.5. Marine ecosystem

The aim of this section is, for the Arctic marine ecosystem, to compile baseline data on the toxic metals Hg, Pb, Cd, and Se; to present a detailed summary of the current state of knowledge of heavy metal levels; and to use this information to identify temporal and geographic trends. Reports of levels of heavy metal contamination likely to have been affected by local anthropogenic sources (e.g., mining) were not included in the tables of this review, but levels found at these point sources are described in the text.

A review of the state of knowledge of contaminants in the Arctic marine ecosystem (to early 1990) was presented by Muir *et al.* (1992). This review focused mainly on the North American part of the Arctic as well as Greenland. No data were presented from Iceland, Scandinavia, or the Russian part of the Arctic, and no data were presented for marine birds. The following section provides an update to the review by Muir *et al.* (1992), and adds data from recent publications as well as data obtained during the AMAP monitoring program. Data from Finland and Sweden have not been included in the marine review as the Baltic and the Gulf of Bothnia are not within the agreed boundary of the AMAP Arctic marine area.

In preparing this updated review, a thorough survey of the literature has been carried out. Literature published since 1991 was included in a search profile for heavy metals in the Arctic environment. Data tables presented in 'Arctic marine ecosystem contamination' by Muir *et al.* (1992) have been incorporated into this report, and provide the basis for much of the literature published prior to 1991 for the Canadian Arctic. The eight Arctic countries also provided some literature for the AMAP process. National assessment reports from Greenland and Canada, recently compiled, have been included (Dietz *et al.* 1996, 1997a, Riget *et al.* 1997a, Muir *et al.* 1996). Electronic data searches were made in the Northern Aquatic Food Chain Contamination Database (Version 1.0 E; October 1993) and in the databases of the Department of Fisheries and Oceans (Canada) in Winnipeg.

All data extracted from available literature are presented in tabular form using a format similar to that used by Muir *et al.* (1992). Species are presented in systematic order; geographical areas are given from north to south; and data are arranged by year. Different tissue compartments (where applicable) are listed in a consistent order (liver, kidney, muscle, bone, blubber).

It is important to note that the level of information available for specific metals varies considerably between locations and species and is related largely to the ease of sampling. Most samples are collected near shore or in areas of human activity. For example, there are more data available for metals in seals than in whales because it is easier to collect seal samples.

An attempt was made to standardize the data where possible. In the older literature, little emphasis was placed on statistical requirements such as normal distribution, homogeneity of variance, etc. In order to facilitate statistical comparison, therefore, data are given primarily as geometric means (GM) and relative standard deviations (rel. SD). Unlike sediment and water data, heavy metal data for biota are usually not normally distributed.

Where possible, all concentrations are related to wet weight of tissues; where the concentrations given in the literature were given on a dry weight basis, they were converted to a wet weight basis, using the moisture content presented in each report for individual samples. Where such information was not available, appropriate conversion factors were used.

Where values for both arithmetic means (AM) and geometric means (GM) were given, but not relative SD, only AM and SD are given in the table. Where raw data were given, GM and rel. SD are calculated. Where two or fewer samples were analyzed, no rel. SD is calculated. Where the given values were means of subsamples taken from the same animal, no SD is given.

Statistical analyzes involving tests of the significance of parameters and of differences of metal levels have not been performed for this assessment, but such analyzes have in many cases been carried out in connection with previous reporting and publishing of the data. References to these analyzes are given in the text.

7.6.5.1. **Seawater**

Information on the concentrations of metal levels in seawater from the Arctic is available mostly through Canadian and German programs. Concentrations of as many as 29 elements are reported from various parts of the Arctic. Arctic data on baseline concentrations have been compiled at the Institute of Ocean Sciences, Department of Fisheries and Oceans in Canada for the Beaufort Sea (Thomas *et al.* 1990), Northwest Passage (Thomas *et al.* 1991), Queen Elizabeth Islands (Thomas *et al.* 1986a), and Canada Basin-Arctic Ocean (Thomas *et al.* 1986b).

Data from the Barents Sea, the White Sea and the Arctic seas have been presented by the Ministry for Environment Protection and Natural Resources of the Russian Federation (MEPNR 1994). It was concluded that many pollutants, including heavy metals, enter Arctic waters through river input from north-flowing rivers such as the Pechora, Yenisey, Lena, Ob, and Kolyma. Most of the pollutants accumulate in river mouths and estuaries and then are spread across the Arctic basin toward Alaska by the circumpolar current. Recent data are available from Pechora, Kara, and Laptev Seas (Rosgidromet 1995).

Contaminants, including heavy metals in seawater, have been given low priority in the AMAP programs, because the concentrations of most pollutants in seawater are close to the detection limits of most laboratories and are very costly to measure.

Lead

Mart and Nurnberg (1984) reported Pb concentrations in eastern Arctic ocean water of 15 ng/L at the surface and 3-4 ng/L at 1500-2000 m. Background concentrations for Pb in Beaufort Sea shelf waters appeared to be in the range of <20-40 ng/L (Thomas *et al.* 1982). Recent seawater analyzes from the Pechora, Kara, and Laptev Seas in Russia show very high Pb levels in the range 160-500 ng/L (Rosgidromet 1995), but these data should be confirmed as they may be unrepresentative. By comparison, Pb levels ranged from 29 to 41 ng/L in the surface waters of the North Atlantic and the Norwegian Sea (Mart and Nurnberg 1984). In the North Pacific between Hawaii and California, Pb in surface water ranged from 5 to 15 ng/L, decreasing with depth to approximately 1 ng/L (Schaule and Patterson 1981). Lead concentrations are consistently higher in surface water than in deeper layers. In coastal waters, concentrations can reach 50 ng/L, and can be much higher in heavily polluted waters (Burnett *et al.* 1980).

Probably more than any other metal, Pb has been enriched, particularly in the northern hemisphere, because of anthropogenic inputs. An indirect estimate of 0.6 ng/L of Pb in prehistoric oceanic surface water has been reported (Schaule and Patterson 1981). Surface waters of the North Pacific contain ten times more Pb now, and those of the Arctic even more, than in prehistoric times. In the Greenland Ice Sheet, modern Pb concentrations are 300 times higher than those characteristic of prehistoric times (Ng and Patterson 1981). However a recent decrease has been observed due to the use of unleaded gasoline (Boutron *et al.* 1991, 1995, Hong *et al.* 1994).

There is clear evidence that mining in the Arctic has increased local Pb concentrations in seawater. At the Black Angel Pb-Zn mine at Maarmorilik in northwest Greenland, which operated from 1973 to 1990, Pb levels in seawater as high as 200 000 ng/L were reported in bottom fjord water, where mine tailings were discharged (Asmund 1992a, 1992b). In surface waters close to the mine site, Pb concentrations up to 42 000 ng/L were measured (Asmund *et al.* 1991). Close to a cryolite mine in Ivittuut in south Greenland (closed in 1986), Pb concentrations in surface waters reached 18 000 ng/L (Asmund *et al.* 1991). Lead concentrations in the range of 40-100 ng/L were reported by Thomas *et al.* (1984) for the waters of Strathcona Sound, a northern Baffin Island fjord at the Nanisivik Pb-Zn mine; these concentrations were approximately one to two orders of magnitude higher than open ocean background concentrations.

Cadmium

Moore (1981) reported Cd concentrations of 70 ng/L for the Central Arctic Ocean for the upper 100 m and then 20 ng/L to a depth of 2500 m, remaining approximately constant over the entire interval. Campbell and Yeats (1982) measured Cd for the eastern Arctic Ocean; concentrations ranged from 13 to 21 ng/L at depths of 1500-2000 m, but surface values were lower (8 ng/L). In the Norwegian Sea, a Cd concentration of 22 ng/L was measured in surface water (Danielsson *et al.* 1985). Rosgidromet (1995) reports Cd levels between 20 and 260 ng/L for waters of the Pechora, Kara, and Laptev Seas. A comparison of values for the North Atlantic and the Norwegian Sea shows no significant mid-oceanic differences for Cd (Mart *et al.* 1984). In relatively warm surface waters, e.g., the northeast Atlantic and more southerly latitudes, Cd concentrations are lower in surface water than in deep water, indicating uptake of Cd by organisms at the surface, and release of Cd from sinking organic matter in subsurface layers.

Similar to the case for Pb, elevated Cd concentrations have been observed locally as a result of mining activities. In bottom water at the point of tailings discharge from the Black Angel mine in Greenland, Cd concentrations of up to 2500 ng/L were found (Asmund 1992a, 1992b). At the Nanisivik mine on Strathcona Sound in the Eastern Canadian Arctic, Thomas *et al.* (1984) reported concentrations in surface water from 30 to 130 ng/L. Close to Ivittuut in south Greenland, surface water concentrations of Cd were between 18 and 252 ng/L (Johansen *et al.* 1995).

Mercury

Very little information is available on the concentrations of Hg in Arctic Ocean water. Weiss *et al.* (1974) reported mean Hg concentrations of 11-22 ng/L (0-400 m) for the southern Beaufort Sea. Thomas (1983) reported background values of 1-15 ng/L for total Hg in samples from the Beaufort Sea shelf. The dissolved Hg concentrations were strongly and negatively correlated with the concentrations of particulate organic carbon. In Puget Sound (Washington, USA), the Hg concentration was 2-10 ng/L (Bothner and Robertson 1975). By comparison, the average Hg concentration was 4.1 ng/L in the Gulf Stream (over a depth of 250-4500 m), 8 ng/L (0-750 m) in the Sargasso Sea, and 3-4 ng/L in waters around the United Kingdom (Mukherji and Kester 1979). In the temperate zone in the North Pacific, total Hg was 14 ng/L in surface waters and 2 ng/L between 500 and 5000 m (Miyake and Suzuki 1983). Off the Swedish west coast, the values ranged from 5 to 12 ng/L (Gustavsson and Edin 1985). There appears to be no great difference in the concentration of Hg in oceanic water around the world, except in polluted waters where concentration can exceed the values noted above by an order of magnitude or more.

The older data have, however, been questioned in a recent NATO workshop (NATO-ARW 1996). Current consensus is that total dissolved Hg in unpolluted ocean water is between 0.2 and 1 ng/L with somewhat higher concentrations in coastal waters and in density interfaces where accumulation due to particulate dissolution is enhanced (up to 2 ng/L). Methylated Hg (when it can be measured) is generally less than 10% of the total Hg. Dimethylmercury is often the major methylated species.

7.6.5.2. **Sediments**

Concentrations of trace elements in sediments depend on local geology, particle size, the amount of organic matter, and the degree of various kinds of anthropogenic influence. The relationship between sediment metal concentration and grain size, with higher metal concentrations found in fine grained sediments than in sandy sediments, is a common feature related to the geochemical distribution of total metals with particle size. The anthropogenic influence might be estimated by the use of normalizers, such as percent organic matter, Al (Windom 1989), percent grain size less than 63 microns (Allan 1971), other grain size fractions (Loring 1990), and Li (Loring 1990). Although it is generally agreed that some kind of normalization is necessary when trace element concentrations in sediments from different environments are compared, or where anthropogenic influence is

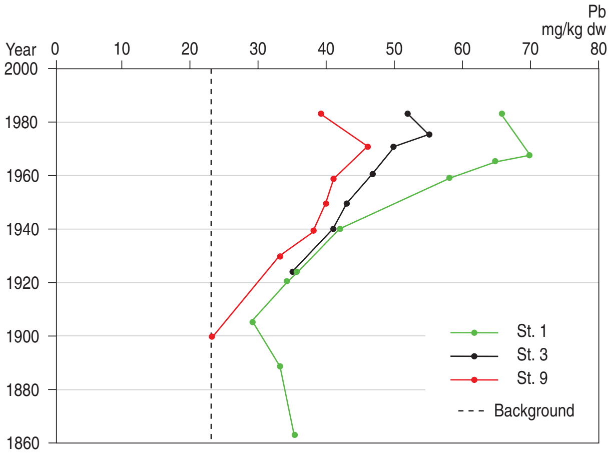


Figure 7·34. Lead in sediments near the cryolite mine at Ivittuut, south Greenland (after Johansen *et al.* 1995). Stations 1, 3 and 9 are located at distances of 800 m, 1600 m and 5600 m, respectively, from the mine.

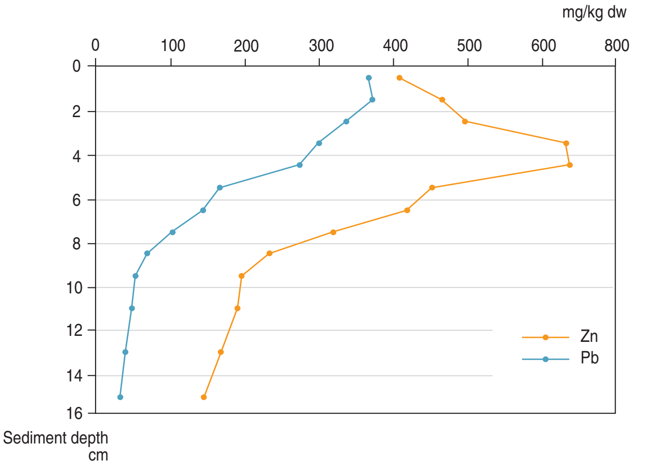


Figure 7·35. Lead and zinc in a sediment core sampled in 1991, 1.5 km from the Black Angel mine in West Greenland. (Source of data: G. Asmund pers. comm.).

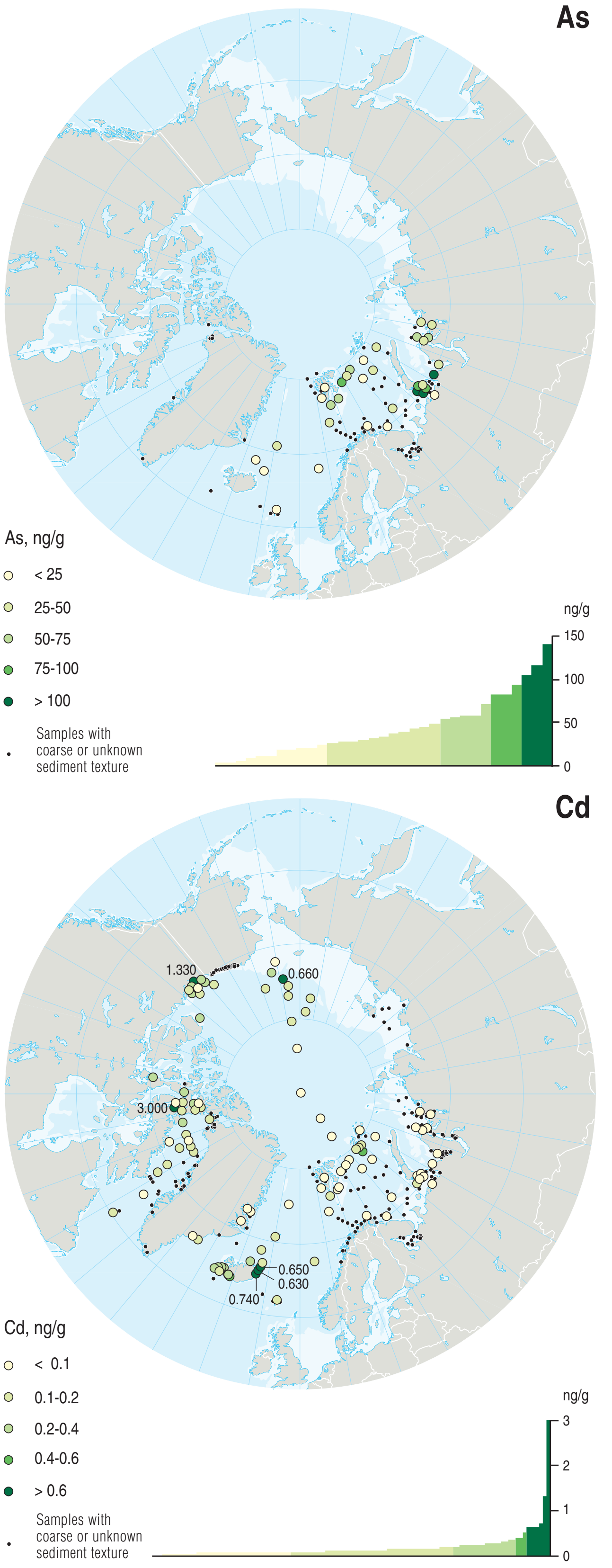
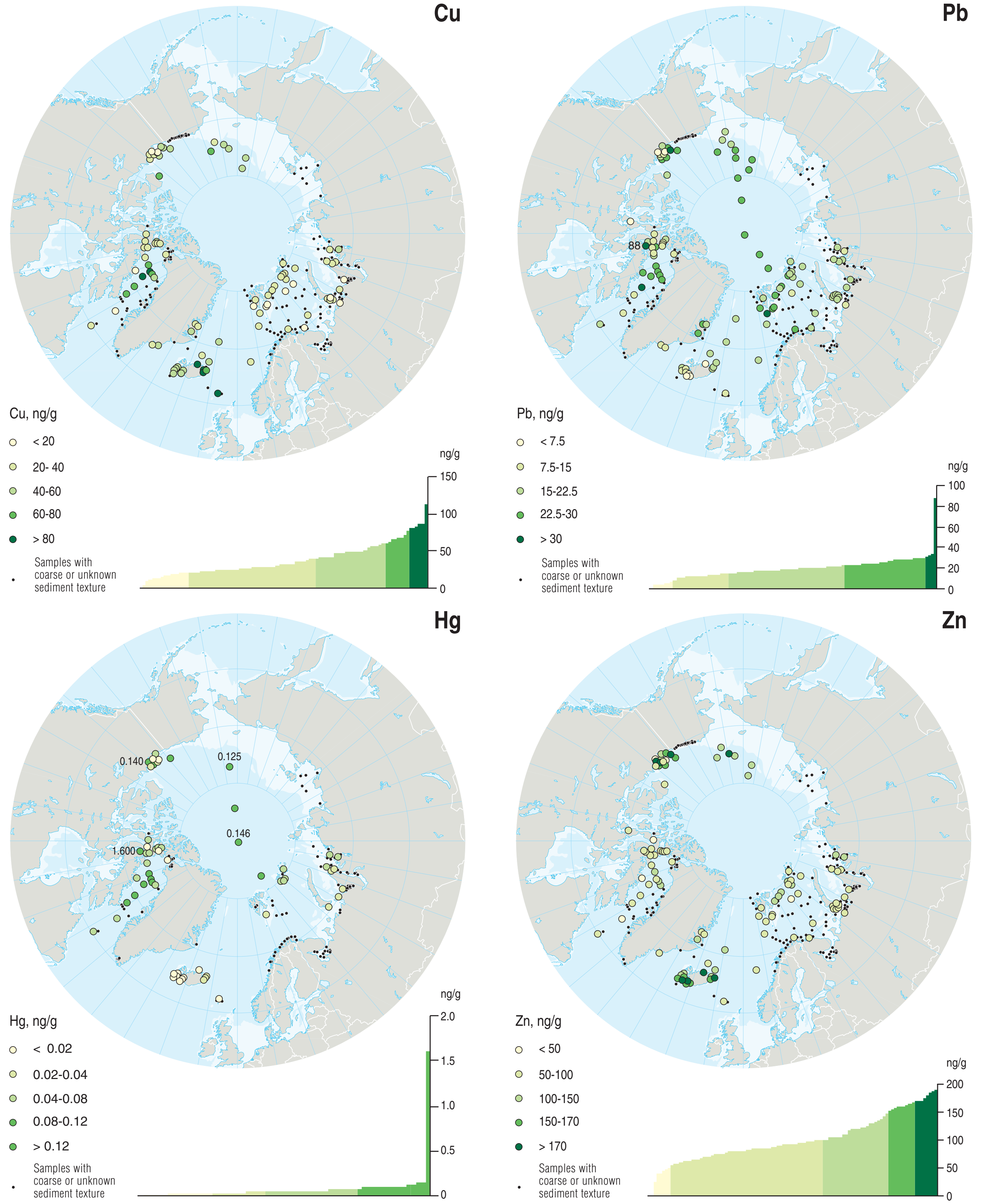


Figure 7·36. Continued on next page →

Concentrations of metals in fine-grained surface marine sediments in the Arctic. (Sources of data: see Annex Table 7·A10).



the subject of concern, the method of normalization that should be used is still a matter of vigorous debate. Also excluded from the overview are severely polluted sediments near local point sources, e.g., Maarmorilik (Loring and Asmund 1987), Strathcona Sound (Fallis 1982, Thomas and Erickson 1983), and Orkdalsfjorden (OSPARCOM/NIVA unpubl. data). Other examples of contaminated sediments are given in Smith and Loring (1981). Some examples of the influence on sediments of mining in the Arctic are shown in Figures 7·34 and 7·35.

The Norwegian data in Annex Table 7·A10 often represent sediments with significant anthropogenic influence. Wherever possible, concentrations are given both in surface sediments and in deeper layers expected to reflect natural background concentrations.

Data for heavy metals in surficial Arctic marine sediments are shown in Figure 7·36. Only those data for fine sediments (i.e. grain size reported as less than 63 microns or other information indicating this, cf. Annex Table 7·A10) are included in the plots in order to make the data comparable.

On this basis, the geographical coverage is limited and there is no systematic distribution evident in the data. The concentrations of metals in the sediments correspond generally with bedrock geology, and any anomalies can be explained in terms of known mineralization or anthropogenic activities. If regional processes such as long-range air transport were a primary factor in controlling sediment metal distribution, then less variability would be expected in the data and distribution, and an emission-related systematic pattern would have been observed.

Copper

The lowest and highest reported values of Cu are 4 mg/kg in sandy sediments from the Canadian Arctic and 137 ng/kg at Vaigat in West Greenland. The data reported for Arctic Russia are near the lower range of these values. These Russian results may, however, be low by 30-40% according to a Norwegian-Russian laboratory intercalibration (Akvaplan-niva 1996). Copper concentrations in whole sediments from Greenland are higher (90-140 mg/kg) in areas containing tertiary volcanic rocks than in areas of non-volcanic sediments (10 to 40 mg/kg) (Loring and Asmund 1996). The concentration of Cu in Arctic marine sediments increases with increasing content of fine grained sediment. Concentrations can be as low as 4 mg/kg in sandy sediments. In sediments containing more than 95% mud, the Cu concentration in Arctic sediments was 61±18 mg/kg (Loring 1984).

As the concentrations of Cu in recent (surficial) sediments are comparable to those in deeper and older sediments, the levels summarized in Annex Table 7·A10 are believed to represent true natural background levels.

Zinc

The lowest reported Zn concentration in sediments from the Arctic is 9.5 mg/kg found in a sandy Barents Sea sediment. The highest, 181±22 mg/kg, is reported for 50 samples from the Beaufort Continental Shelf. The data for marine sediments from the Russian Arctic, 25-75 mg/kg, could be up to 42% lower based on the results of the Norwegian-Russian laboratory intercalibration (Akvaplan-niva 1996). Loring and Asmund (1996) found no significant differences between Zn concentrations in sediments from various geological provinces of Greenland. The concentrations of Zn in fine-grained sediment ranged between 40 and 100 mg/kg.

Both Loring (1984) and Macdonald and Thomas (1991) found that Zn concentrations were higher in fine grained sediments than in sandy sediments. Zinc concentrations ranged from 22 to 160 mg/kg in sediments containing 0% mud and 100% mud, respectively. As there is generally no trend in the concentrations of Zn with depth, the values shown in Annex Table 7·A10 are believed to be true natural background levels, with the exception of Norwegian surface sediments.

Lead

The lowest reported levels are 1.4±0.2 mg/kg (Thomas *et al.* 1983, cited in Muir *et al.* 1992) in 124 sediment samples from the southern Beaufort Sea continental shelf, and 4.17 ±0.72 in 26 sediment samples from Hudson Bay. In sediments from the Beaufort Sea shelf, concentrations ranged from 9.1 to 20.1 mg/kg, corresponding to 0% and 100% mud, respectively. The highest concentration of Pb in Arctic sediments far from point sources is 53.0±2.2 mg/kg in the surface layer of three sediment cores from Ålesund, Norway. This is probably of anthropogenic origin as the same cores had a concentration of only 22 mg/kg at a depth of 40-45 cm. The highest reported concentration in sediments believed to be natural is 43.5±2.2 in sediments from southern Beaufort Sea continental shelf. The wide range of values evident in the southern Beaufort Sea is probably related primarily to sediment texture and mineralogy. The data for the Russian Arctic (7.7±1.5 to 17±5 mg/kg) could be low by 0% and 61% based on the results of the Norwegian-Russian laboratory intercalibration exercise (Akvaplan-niva 1996).

Loring and Asmund (1996) found that the concentrations of Pb in Greenlandic sediments from areas containing tertiary volcanic rocks and from the Nagssugtoqidian mobile belt were lower (12-14 mg/kg) than in sediments from other geological provinces (21-24 mg/kg). In contrast, Loring (1984) found no dependence of concentrations on mud content for a suite of Arctic sediments that contained 18±6 to 21±8.5 mg/kg of Pb. The lack of enrichment of Pb in the upper one or two centimeters of Arctic marine sediments suggests that long range transport of aerosol particles is not an important process in the Arctic.

Cadmium

The lowest reported concentration of Cd in Arctic surface sediments is 0.02 mg/kg in a sample from the Barents Sea. High values of 4.6±0.3 and 1.33±0.54 have been reported in the southern Beaufort Sea and the Mackenzie Delta. These values probably include Cd from barite used in drilling fluids during oil and gas drilling activities (Thomas, pers. comm.). Other high values include 0.32±0.31 for Beaufort Sea, 0.31 ±0.08 for the MacCormick Fjord, Greenland, 0.23±0.21 for the Ob Gulf, Russia, and 0.52 for one sample out of 43 in the Barents Sea. There are, however, several examples showing that Cd accumulates at the interface between oxidized and reduced sediment, where Mn also has a discontinuity (e.g., Gobeil and Macdonald unpubl.). For this reason, many sediment cores cannot be used for temporal trend studies.

No significant differences were found in Cd concentrations between the geological provinces in Greenland (Loring and Asmund 1996); this is probably related to the relatively high analytical uncertainty in Cd measurements coupled with the fact that most concentrations are low or at the detection limit. The average concentration reported for deep water Greenlandic sediments was 0.l23±0.048 mg/kg. Loring (1984) found a strong dependence between Cd concentrations and mud content; Cd concentrations ranged from 0.058 to 0.144±0.030 mg/kg corresponding to mud content of <5% to >95%, respectively. There is no indication that Cd accumulates in the upper layers of Arctic sediments.

Mercury

Reported values for the concentration of Hg in Arctic sediments range from below the detection limit of 0.01 mg/kg in subsurface sediments from Roroy and Skrova, Norway, and 0.009±0.004 mg/kg in sediments from Uummannaq, Greenland to 0.243±0.043 mg/kg in southern Beaufort Sea continental shelf sediments and 1.6±1.2 mg/kg in sediments from Strathcona Sound, Canada collected in 1975. The latter very high value may reflect the influence of the nearby sulfide ore body (the Nanisivik mine started operation in October 1976). No dependence of Hg concentration on source rock geology was detected from Greenlandic sediments (Loring and Asmund 1996), where the average concentration was 0.045 ±0.045 mg/kg.

Hg concentrations generally correlate negatively with sediment grain size. On the Beaufort Sea shelf, Hg ranges from 0.01 to 0.1 mg/kg in sediments containing 0% to 100% mud (Macdonald and Thomas 1991). Similarly, Loring (1984) reported a grain size-Hg dependence for sediments from the Canadian Arctic in the concentration range 0.04±0.014 to

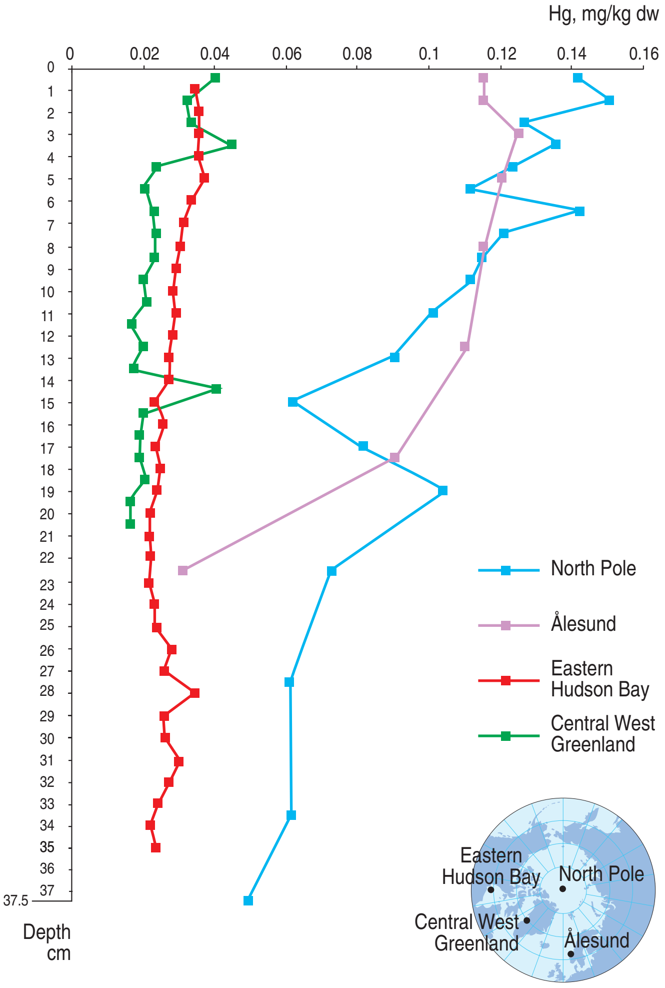


Figure 7·37. Concentrations of Hg in Arctic marine sediment cores. (Source of data: Gobeil and Macdonald unpubl., North Pole; Norwegian Institute for Water Research, Ålesund; Lockhart unpubl., eastern Hudson Bay; Dietz *et al.* 1997b, central West Greenland).

0.07±0.039 mg/kg. There are several data sets that indicate widespread accumulation of Hg in surficial Arctic sediments (Lockhart unpubl., Dietz *et al.* 1997b, OSPARCOM/NIVA unpubl. data, Gobeil and Macdonald unpubl.; Figure 7·37). The enrichment of Hg occurs in the upper 2 to 10 cm of the sediments, even at the North Pole. This phenomenon could indicate global scale input of Hg to the marine environment in recent times. More comprehensive investigations of Arctic sediments are required, however, before definite conclusions can be drawn about the nature and source of the observed enrichment.

Arsenic

Arsenic is not a part of the AMAP sediment monitoring program, but data on As has often been reported for Arctic sediments. Loring *et al.* (1995) and Maage *et al.* (1996) report values for As in sediments of the Pechora Sea and the Barents Sea. The concentrations ranged from 2.0 to 308 mg/kg. Loring *et al.* (1995) found high As concentrations in sediments contaminated with radionuclides from local sources (e.g., near the nuclear weapons test site at Novaya Zemlya). However, subsequent work by both Maage *et al.* (1996) and Loring (pers. comm.) shows that As enrichment is a common feature in sediments of the Barents and Pechora Seas. The explanation for this enrichment is probably natural and geological, however some attention might be given to the potential effects of As accumulation in higher avians and mammals.

7.6.5.3. **Microorganisms**

No data were found on the concentrations of heavy metals in microorganisms from the Arctic.

7.6.5.4. **Algae**

Few data have been published on heavy metals in Arctic algae. Data, primarily for Cd and to a lesser extent Pb, Hg, and Se, are available for five intertidal species in Greenland, but the geographical extent is very limited (Annex Table 7·A11). Additional data in algae are available only on Pb for Canada and Norway.

Lead

The Greenland data do not indicate differences in Pb levels between the three species analyzed for Pb. Lead concentrations vary by a factor of about two, with geometric mean values in the growing tips ranging from 0.175 to 0.360 µg/g dw. No geographical differences within Greenland are indicated. In algae from Canada and Norway (Svalbard), Pb concentrations are 3-5 times higher than those from Greenland. It is suspected that these differences do not reflect real geographical differences of baseline levels, but rather differences in sample preparation, analytical techniques, or species composition.

Lead concentrations are higher in whole algae plants than in growing tips (Annex Table 7·A11). No significant within-year variations have been found in areas not affected by local pollution in Greenland (Riget *et al.* 1995). Brown algae have been used as bioindicators of heavy metal pollution from mining operations in Greenland, and the algae have shown to be very good indicators of local geographic differences and temporal trends of Pb pollution (Figure 7·38, Johansen *et al.* 1991). At the Black Angel mine in northwest Greenland, Pb in seaweed was elevated in a wide area (up to 30 km from the mine), with values up to 50 µg/g near the mine (Johansen *et al.* 1997). Local elevations in Pb concentration were also observed at the cryolite mine in south Greenland (Johansen *et al.* 1995), at the Pb-Zn mine in East Greenland (Johansen *et al.* 1985), and at the Nanisivik Pb-Zn mine at Strathcona Sound in the Canadian Arctic (Asmund *et al.* 1991).

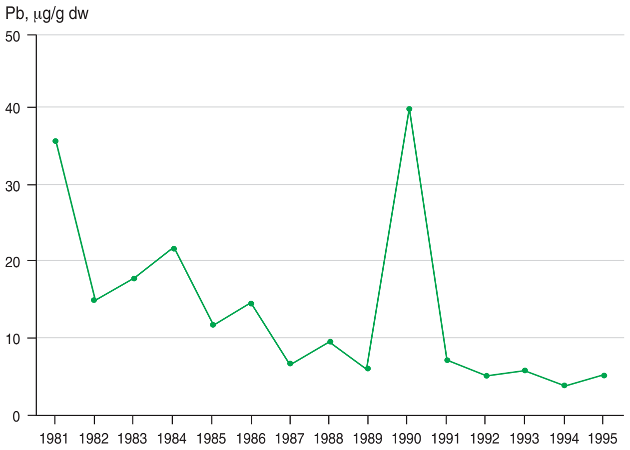


Figure 7·38. Temporal trend of Pb concentrations in growing tips of seaweed (Fucus vesiculosus) at the Black Angel lead-zinc mine in West Greenland. The background value is estimated to be 0.2 µg/g dw. (After Johansen *et al.* 1997).

Cadmium

The concentration of Cd found in algae from Canada range from 0.7 to 1.8 µg/g dw. The more comprehensive dataset from Greenland shows a wider range, from 0.142 to 3.503

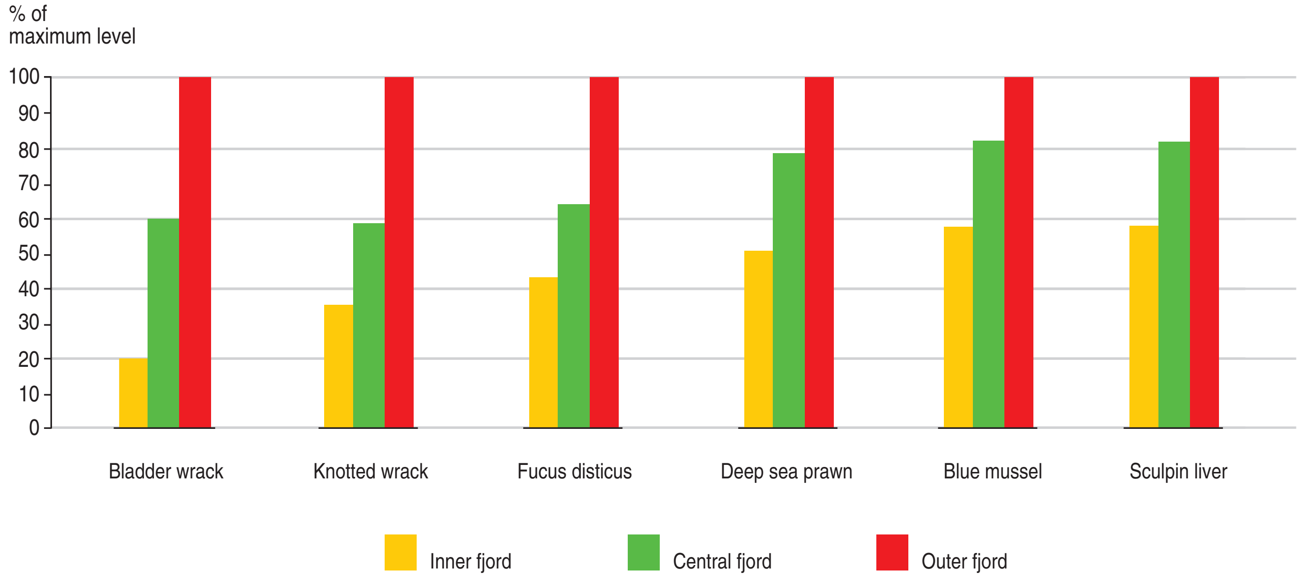


Figure 7·39. Local differences in Cd concentrations in sedentary and stationary algae and biota from Nuuk and Uummannaq fjords, central West Greenland (after Dietz 1995). Levels are plotted as the percentage of the maximum concentrations.

µg/g dw (Annex Table 7·A11). Concentrations in knotted wrack (*Ascophyllum nodusum*) are from three to six times lower than in bladder wrack (*Fucus vesicolosus*) and the other wrack species, *Fucus distichus*, but are similar to the concentration reported for an unidentified kelp species from Svalbard.

No geographical differences on a regional scale are apparent. However, important local geographical differences in Cd concentrations were detected in Greenland. The highest concentrations were found near the open sea and the lowest concentrations in the inner reaches of large fjords, with levels differing by up to a factor of 5 (Riget *et al.* 1993) (Figure 7·39). This agrees with Russian observations showing that the dissolved Cd concentration increases with the salinity in the Ob and Yenisey estuaries (Dai and Martin 1995). Cadmium concentrations have also been found to vary seasonally. In Greenland, Cd concentrations in bladder wrack in February were about a factor of 3 higher than in August (Riget *et al.* 1995).

At the three former mine sites in Greenland, Cd levels in seaweed were not elevated above background levels, although Cd was higher than background in ore and mine waste (Johansen *et al.* 1985, 1991, Hansen and Asmund 1986).

Mercury and selenium

Few data are available on Hg and Se concentrations in algae (Annex Table 7·A11). Selenium levels seem not to differ much (concentrations range from <0.2 to 0.6 µg/g dw), whereas larger differences have been reported for Hg (<0.01 µg/g dw in kelp from Canada to more than 1 µg/g dw in kelp from the Kara Sea, Russia). Data, however, are too few and the uncertainty in the Kara Sea data is too high to reach firm conclusions about possible geographical differences.

7.6.5.5. **Invertebrates**

Data (Annex Table 7·A12) are available mainly for bivalves, amphipods, and decapods. The following description of metal concentrations in invertebrates will concentrate on these groups.

Lead

In bivalves, most data are available for the blue mussel which has been selected as an indicator species in the AMAP program. Data from Greenland show that the Pb concentration in blue mussels increases slightly with increasing shell length (Riget *et al.* 1996). Older data (1983-1990) from southwest Greenland show Pb concentrations from 0.142 to 0.476 µg/g. Recent AMAP data from Greenland are similar to the lowest value of the older Greenland data, ranging from 0.072 to 0.155 µg/g at Qeqertarssuaq (central West Greenland) and from 0.124 to 0.188 µg/g at Nanortalik (south Greenland). Norwegian samples of blue mussels collected from 1984 to 1993 are in the range of 0.101-0.483 µg/g, which is similar to the range observed in the same period in Greenland. Old Icelandic data (1978) are relatively high (0.56-0.805 µg/g), whereas Icelandic data from 1990 to 1992 are lower (range 0.019 to <0.195 µg/g). Older Greenland data (1984-1985) for other bivalve species are in the same range (0.069-0.186 µg/g) as the 1994 AMAP data for blue mussels. Data on bivalves from areas outside Greenland, except Iceland, cannot be directly compared with the Greenland data, as they appear to correspond to a different tissue. Concentrations in Canada and Russia seem to be higher than in Greenland, but these data may not be reliable and require confirmation and intercalibration.

At the Black Angel mine in northwest Greenland, Pb concentrations in blue mussels were elevated over background levels in a wide area (up to 30 km from the mine), with values exceeding 1000 µg/g nearest the mine (Figure 7·40; Johansen *et al.* 1991). Local elevations were also observed in blue mussels at the cryolite mine in south Greenland (Johansen *et al.* 1985), in three bivalve species at the Pb-Zn mine in East Greenland (Hansen and Asmund 1986), and in clams at

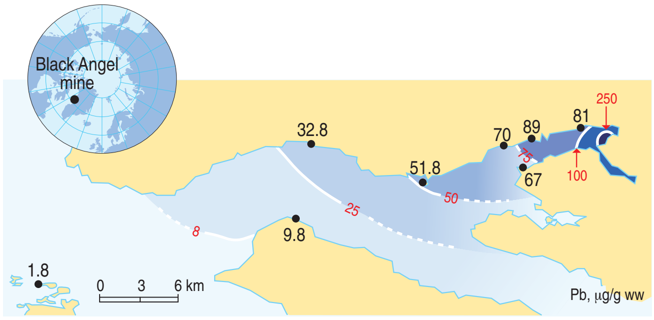


Figure 7·40. Lead levels in blue mussels (*Mytilus edulis*) in the fjord outside the Black Angel lead-zinc mine in West Greenland in 1986-87. Values on the fjord coast are measured concentrations. Estimated concentrations in the fjord are indicated by isolines. (After Johansen *et al.* 1991).

the Nanisivik Pb-Zn mine at Strathcona Sound in the Canadian Arctic (Asmund *et al.* 1991). At the Black Angel mine, local elevations of Pb were also observed in the deep-sea prawn (Johansen *et al.* 1991).

Data on Pb concentrations in crustacea, primarily Greenlandic deep-sea prawn, indicate higher values in heads and shells than in muscle by a factor of 2 to 10, and higher in small than in large prawns. Geographical differences were not apparent, and Pb levels were generally low, with geometric mean values ranging from 0.008 to 0.050 μg/g in muscle, and 0.070-0.181 μg/g in heads and shells. Comparable Pb concentrations were found in another shrimp species (*Eualus belcher*) from Greenland.

Data on Pb in crustacea from other Arctic countries are generally very limited. The few available Russian data were higher than other values, except for recent Norwegian data on meat from the deep-sea prawn from Canada and Norway. Here levels are similar to what is found in Greenland. Lead concentrations in Arctic zooplankton from the Greenland Sea and Fram Strait appear high in some cases.

For other invertebrate groups, data are limited but include a few results on polychaetes, echinoderms, and ascidians from Russia. They cannot be compared with other areas directly, but some of the Pb values appear to be relatively high.

Cadmium

Most of the mollusk data obtained prior to 1994 are for blue mussels from three areas in West Greenland and Norway. Concentrations range from 0.458 μg/g in small mussels from Uummannaq to 1.065 μg/g in large mussels from Paamiut. There is no indication of regional differences within Greenland (Annex Table 7·A12); however, local geographical differences have been found. Similar to the situation for algae, the highest Cd concentrations in blue mussels are seen in the open sea and the lowest in the inner regions of large fjords (Riget *et al.* 1996) (Figure 7·39). The highest Cd concentrations are found in the largest mussels (Riget *et al.* 1996).

The 1994 AMAP data from south Greenland (Nanortalik) range from 0.561 to 0.763 μg/g and are at the same level as the older Greenland data, whereas the new data from central West Greenland (Qeqertarssuaq) are somewhat higher (0.692-1.254 μg/g).

The data on blue mussels from Iceland indicate that Cd concentrations there are similar to or lower than those in Greenland, whereas they are lower in blue mussels from Norway (0.124-0.353 μg/g).

In two other bivalve species (a cockle, *Serripes groenlandicus,* and green crenella from northern and East Greenland), Cd concentrations are similar to those found in the blue mussel, whereas levels in Iceland scallop from northwest Greenland are significantly higher, by a factor of 3-6.

Cadmium data in other bivalve species and in gastropods from other countries are sparse, but the levels generally fall within the ranges observed in Greenland.

Many data are available on Cd in crustacea, especially amphipods and decapods, but almost all are from Canada and Greenland (Annex Table 7·A12). Some of the data were reported on a dry weight basis and some on a wet weight basis; a conversion factor of 0.25 for whole crustacea (Dietz *et al.* 1996) has been used to convert to wet weight basis for comparisons.

In general, high Cd concentrations are found in crustacea, but with large variations, even within the same species. In amphipods, Cd concentrations in whole animals range from 0.11 to 4.60 μg/g ww, with most values exceeding 1 μg/g. There are no systematic geographical trends in the data.

Cd concentrations in crustacea increase with the size (age) of the animals. In the amphipod *Parathemisto libellula*, Cd concentrations are approximately twice as high in large as in small animals (Annex Table 7·A12).

For decapods, data are available mainly for Greenland. Five species have been analyzed. Analyzes were either carried out on whole animals or on the muscle and remaining parts (heads and shells) separately. Cadmium levels are highest in heads and shells (referred to as shell), intermediate in whole animals, and lowest in muscle. Shell values range between 0.344 and 3.93 μg/g ww, with lowest levels in small deep-sea prawns from Nuuk and highest in large individuals of the same species from Uummannaq. Whole animal values range between 1.28 and 7.91 μg/g, with lowest levels in deep-sea prawns from Maniitsoq and highest in large Bikini prawns from Avanersuaq.

Concentrations of Cd in muscle range between 0.013 and 0.046 μg/g. The lowest concentrations are found in deep-sea prawns from Nuuk (both large and small) and small prawns from Paamiut. The highest muscle levels are seen in large deep-sea prawns from Uummannaq. The limited data from Canada and Norway indicate Cd levels similar to those in Greenland.

The local geographical differences of Cd levels that have been found for seaweed and bivalves are also observed in crustaceans. In deep-sea prawns the highest Cd concentrations occur in samples from the open sea and the lowest in samples collected in the inner regions of large fjord systems in Greenland (Figure 7·39); the observed levels differed approximately by a factor two (Johansen *et al.* 1991).

For the remaining groups of crustacea (copepods, ostracods, mysids, isopods, and euphausiids) few data are available (Annex Table 7·A12). In general they also have high concentrations, but with large variations, from <0.015 μg/g in euphausiids to 6.62 μg/g ww in copepods.

For other invertebrate groups, annelids, echinoderms, ascidians, and chaetognaths from Russia, Canada, and Norway, Cd data are sparse, and comparisons cannot be made, except to note that Cd levels in these groups seem to be lower than in mollusks and crustacea.

At the three former mine sites in Greenland, Cd concentrations in invertebrates in most cases do not exceed background levels (Johansen *et al.* 1985, 1991, Hansen and Asmund 1986).

Mercury

Most data are for bivalves from Greenland and Norway and for decapods from Greenland.

In bivalves from Greenland, Hg levels are similar among species and regions (Annex Table 7·A12). In soft tissue, the range is from 0.011 μg/g in a cockle to 0.020 μg/g in an Iceland scallop. The 1994 AMAP data from Greenland fit well into this range with levels from 0.014 to 0.017 μg/g, and similar values are found in blue mussels from Iceland. Concentrations of Hg in bivalve samples from Norway are similar (<0.009-0.033 μg/g) to those found in the Greenland samples.

Although tissues and species are not similar, Hg values for bivalves from Russian and Canada are similar to those found in Greenland and Norway. In general, Hg concentrations in bivalves are low compared with those found in fish, seabirds, and marine mammals from the Arctic.

Although higher than in bivalves, Hg levels are low in crustacea. In decapods from Greenland whole animal values range between 0.023 and 0.258 μg/g, with the lowest levels occurring in small Bikini prawns from Avanersuaq and the highest in large deep-sea prawns from Baffin Bay.

The Hg concentration is higher in whole large decapods than in small animals. No systematic geographical differences in Hg levels in decapods have been reported.

In copepods and ostracods, Hg concentrations seem to be at the same level as in decapods, whereas in the remaining crustacea groups (isopods, amphipods, and euphausiids) and in annelids, echinoderms, and ascidians, data are few but Hg levels appear lower than in the other crustacea groups (Annex Table 7·A12).

Selenium

Most results are from Greenland, and almost all are for bivalves and decapods. In three bivalve species (Iceland scallop, green crenella, a cockle), all from the same area in northern Greenland, Se concentrations range from 0.326 to 0.869 µg/g (Annex Table 7·A12). The level in the green crenella is about twice as high as in the two other species.

The 1994 AMAP data on Se in blue mussels from two regions in Greenland are at the same level as in green crenella, ranging from 0.53 to 0.94 µg/g. The highest Se concentrations are found in small blue mussels, decreasing by one third in the largest mussels.

Selenium concentrations are higher in decapods than in bivalves. As is the case for Cd, Se appears to be concentrated in the heads and shells and not in the muscle of the animals. The concentrations of Se in whole decapods from Greenland range between 1.09 and 3.91 µg/g. The lowest concentrations are found in small Bikini prawns from Avanersuaq and the highest in large Sclerocrangons from Ittoqqortoormiit. Although Se concentrations are higher in whole large crustaceans than in small, differences are not large. No geographical differences on a regional scale are evident in the existing data.

The few Se data in other animal groups (gastropods, amphipods, and ascidians) fall within the range of those found in bivalves and decapods.

7.6.5.6. **Fish**

The concentrations of Pb, Cd, Hg, and Se in marine fish species are presented in Annex Table 7·A13.

Lead

Pb data are available for several fish species. Most data are for muscle and liver tissue, a few are for bone, and one is for spleen and gonads. Generally, Pb concentrations in fish muscle are very low (<0.002-0.05 µg/g). Where muscle and liver have been analyzed, the levels in liver are higher than those in muscle – up to 0.2 µg/g, mainly in fish species from Orkdalsfjorden/Trossavika, Norway which may be contaminated. The few data of Pb in bone tissue indicate levels similar to those in liver. Many of the older data are higher than newer results; they are considered to be overestimates because of analytical problems which were prevalent when the samples were analyzed (Dietz *et al.* 1996, 1997b).

Age accumulation. The data available for fish are too few to allow analysis for age/size dependence. Dietz *et al.* (1997a), Riget *et al.* (1997b), and Bohn and Fallis (1978) found no correlation between fish size (age) and Pb concentrations.

Geographical differences. The limited data available for Pb in marine fish do not allow for general conclusions regarding possible geographical differences. However, recent investigation carried out under the AMAP program showed that Pb levels in livers of Arctic cod (*Boreogadus saida*) and shorthorn sculpin (*Myoxycephalus scorpius*) were higher in East Greenland than in West Greenland (Riget *et al.* 1997a).

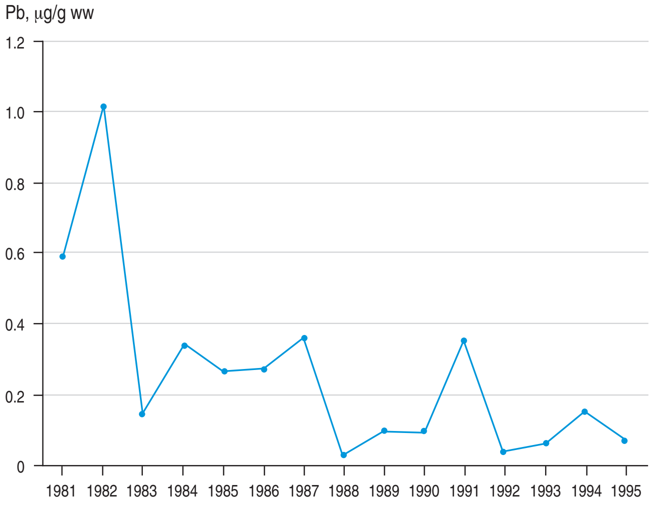


Figure 7·41. Temporal trend of Pb concentrations in liver tissue of spotted wolffish (*Anarhichas minor*) at the Black Angel lead-zinc mine in West Greenland. (After Johansen *et al.* 1996).

Point sources. At the Black Angel mine in northwest Greenland, local elevated Pb concentrations were measured in the bone of shorthorn sculpin and spotted wolffish (*Anarhichas minor*). Johansen *et al.* (1985) report Pb concentrations of 5 µg/g in the bones of capelin (*Mallotus villosus*); concentrations were lower in liver and muscle tissue. A general decline has been observed for a number of species due to improved treatment of mine waste (e.g., Figure 7·41). Elevated levels of Pb in fish were not observed at the cryolite mine in south Greenland (Johansen *et al.* 1985), whereas Agger *et al.* (1991) report enrichment of Pb concentrations in the Pb-Zn mine in East Greenland.

Cadmium

Cadmium data are available for a substantial number of fish species in muscle, liver, kidney, bone, bile, spleen, and gonads, but most are for muscle and liver tissue. The concentrations of Cd in fish muscle are generally below the detection limit (<0.001-0.05 µg/g). Cadmium levels in liver (generally up to 1.0 µg/g) were higher than in muscle. For wolffish, cusk (*Brosme brosme*), redfish (*Sebastes marinus*), shorthorn sculpin, and Greenland halibut (*Rienhardtius hippoglossoides*), the liver values were even higher, between 1-12 µg/g. One of the reasons for these high Cd levels may be the long life span of some of these fish species, especially wolffish, Greenland halibut, and redfish. Stange *et al.* (1996) also found higher levels in redfish than in fish species with a shorter life span. Extremely high values were found in livers of Pacific herring (*Clupea harengus*) and broad whitefish (*Coregonus nasus*) from Tuktoyaktuk Harbour (30.6 and 40.3 µg/g). The levels of Cd in kidney were lower than in liver for most of 13 Greenland species, below or just above a value of 0.1 µg/g. The highest value was found in the kidney of a cusk (4.2 µg/g).

Cadmium levels in fish bone from Greenland were all <0.015 µg/g. The bile and spleen of a small number of Greenlandic fish species contained mean Cd concentrations from <0.001 to 0.4 µg/g. The highest individual values were found in an eelpout (*Lycodes eudipleurostichus*) from Ittoqqortoormiit (0.643 µg/g and 2.06 µg/g in spleen and bile, respectively). Gonads from six fish species were analyzed, and ranged from < 0.001 to 0.33 µg/g in Arctic cod from Pangnirtung.

Age accumulation. There is no solid evidence that the concentration of Cd increases with size of fish. However, Dietz *et al.* (1997a) and Riget *et al.* (1997b) found a tendency of increasing Cd concentrations with size in a limited portion of the Greenlandic data, as did Bohn and Falls (1978) for shorthorn sculpin. On the other hand, Hellou *et al.* (1992b) found Cd concentrations in Atlantic cod (*Gadus morhua*) were negatively correlated with size.

Geographical differences. Cd levels in liver of Atlantic cod, Greenland cod (*Gadus ogac*), redfish, shorthorn sculpin, and especially Arctic cod can be compared over broad geographical areas. No clear geographical differences are evident as the variability of the data on a local scale is often similar to that on a regional scale. However, the data indicate that Arctic cod from the Barents Sea have lower concentrations of Cd than Arctic cod from other areas, Figure 7·42. Samples from Arctic cod and shorthorn sculpin collected in 1994 and 1995 showed higher Cd contents in livers of individuals from northwest Greenland compared with other Greenland areas (Riget *et al.* 1997a). Local differences have been found in the Cd content of livers of shorthorn sculpin from some Greenland fjords: samples collected in the open sea can contain Cd concentrations up to three times higher than those in sculpin collected from the inner regions of large fjords (Figure 7·39) (Asmund *et al.* 1988).

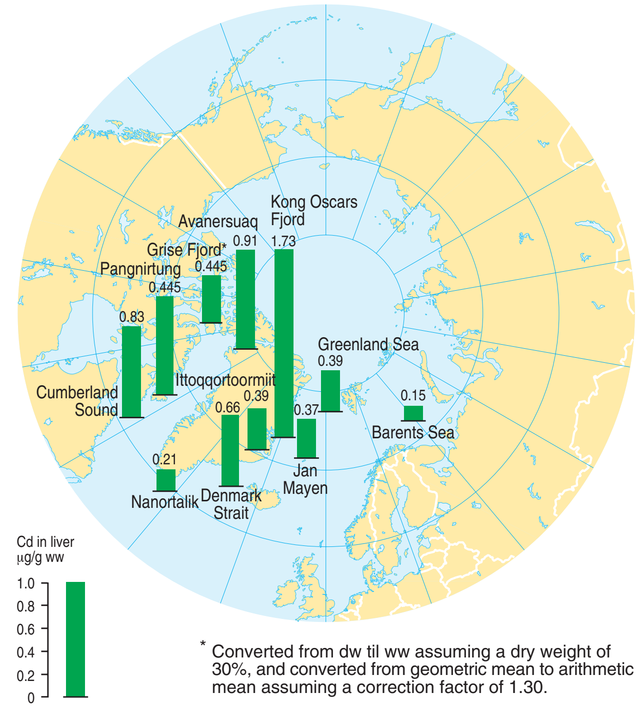
Point sources. At three mine sites in Greenland (Black Angel Pb-Zn mine, cryolite (Al) mine in south Greenland, and Pb-Zn mine in East Greenland), monitoring data suggest no elevation of Cd in fish above background (Johansen *et al.* 1991, Hansen and Asmund 1986, Agger and Johansen 1992, Johansen *et al.* 1995, Asmund *et al.* 1988).

Mercury

Data are available for Hg in muscle, liver, kidney, bile, spleen, and gonads of fish; most data are for muscle and liver. In muscle the concentrations of Hg are generally in the range of 0.01-0.1 µg/g. In some species, such as Greenland halibut, two eelpout species, Arctic cod, and shorthorn sculpin, Hg concentrations are higher, between 0.1 and 0.2 µg/g. The highest values in fish muscle were found in Arctic cod from Kong Oscars Fjord in East Greenland (0.882 µg/g). Concentrations in liver were lower than in muscle, between 0.01 and 0.06 µg/g for most species. The highest value of Hg in liver tissue was found in samples of Arctic cod from Upernavik (0.192 µg/g).

In kidneys, Hg concentrations ranged between 0.013 and 0.078 µg/g, except in Greenland cod from Cambridge Bay (0.17 µg/g). The concentration of Hg in the spleen and bile of Greenlandic fish ranged from 0.011 to 0.122 µg/g and from <0.005 to 0.067 µg/g, respectively. In gonads of nine fish species Hg concentrations ranged from 0.01 to 0.09 µg/g.

Age accumulation. Dietz *et al.* (1997a) and Riget *et al.* (1997b) found a clear, positive correlation between Hg concentrations in muscle, liver, and kidney and size of fish. Stange *et al.* (1996) report a similar relationship for fish from the North Atlantic.



* Converted from dw til ww assuming a dry weight of 30%, and converted from geometric mean to arithmetic mean assuming a correction factor of 1.30.

Figure 7·42. Circumpolar distribution of Cd levels in liver tissue of Arctic cod (*Boreogadus saida*). Arithmetic mean concentrations. (Sources of data: see Annex Table 7·A13).

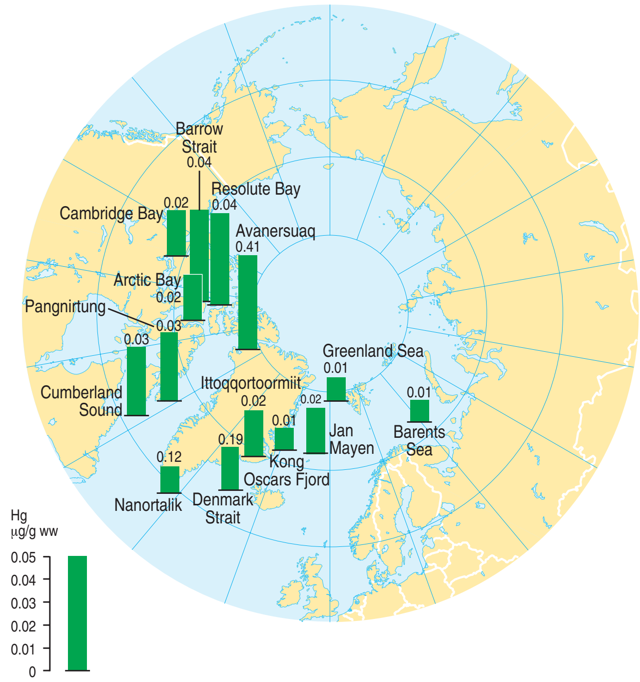


Figure 7·43. Circumpolar distribution of Hg levels in muscle tissue of Arctic cod (*Boreogadus saida*). Arithmetic mean concentrations. (Sources of data: see Annex Table 7·A13).

Geographical differences. There are very few fish species which have been sampled on a circumpolar scale and analyzed for Hg. An exception is Arctic cod for which the concentration of Hg in muscle tissue is available for the eastern Canadian Arctic, West and East Greenland, Jan Mayen, the Greenland Sea, and the Barents Sea. The concentrations of Hg in fish from the Barents Sea and Greenland Sea are lower than in the other areas; however, the sampling periods differed considerably among areas, which may have influenced the levels found (Figure 7·43). Some of the highest values were found in fish from the northern areas of eastern Canada as well as western and eastern Greenland.

Temporal trends. Very limited information is available on temporal trends in marine fish. A study from two locations in the Baltic (Landsort and Utlängen) south of the Arctic area (58°45'N), however, have shown an increasing trend over the period from 1980 to 1993 (Bignert *et al.* 1995).

Selenium

Data on the Se content of 20 fish species are available. In muscle, concentrations range from below detection (0.2 μg/g in the Greenland data) to 1.01 μg/g in shorthorn sculpin from East Greenland. In liver, concentrations are clearly higher than in muscle; the highest values are reported for Atlantic wolffish, Greenland halibut, and golden redfish, all from Nuuk, West Greenland (~3 μg/g), for redfish from Denmark Strait (4.87 μg/g), and for Pacific herring from Tuktoyaktuk Harbour (3.26 μg/g). The concentrations of Se in kidneys are similar to or marginally higher than those found in liver. Long rough dab from south Greenland had the highest concentration, in kidney tissue, 6.30 μg/g. The Se concentration in spleen of nine Greenlandic fish species ranged from 0.39 μg/g in cusk from south Greenland to 5.25 μg/g in Arctic cod from Ittoqqortoormiit, East Greenland. In four fish species, all from Nuuk, West Greenland, the levels of Se in bile range from below detection (<0.2 μg/g) to 0.74 μg/g. Levels of Se in gonads were generally low except for Arctic sculpin from Tuktoyaktuk Harbour.

Age accumulation. Concentrations of Se in liver tended to decrease with increasing length of fish. Stange *et al.* (1996) report a similar trend for Atlantic cod but not for redfish. No correlation between the concentration of Se in muscle or kidney tissue and fish length is apparent in the data (Dietz *et al.* 1997a, Riget *et al.* 1997b).

Geographical differences. Data for the Se concentration in liver of Arctic cod exist for the eastern Canadian Arctic, West and East Greenland, Jan Mayen, the Greenland Sea, and the Barents Sea, but no clear geographical differences are apparent.

7.6.5.7. **Seabirds**

Lead

Several seabird species were analyzed for Pb, mostly from Greenland, Canada, and Norway (Annex Table 7·A14). Few data are available from Iceland and Russia. In seabird muscle, some extreme high values (up to 26 μg/g) are found in several species from Svalbard, Norway. Whether these high values are real is unknown. It is necessary to evaluate the data for Pb in the breast tissue of birds with caution, particularly if they were killed by lead shot. All other Pb values are below 0.4 μg/g, except for one specimen of king eider (*Somateria spectabilis*) from Holman Island, Canada which contained 1 μg/g of Pb. The levels in liver and kidney ranged from <0.009 to 0.8 μg/g. The highest concentrations in kidney are found in seabirds from Svalbard. The analyzes of bone from five species of seabirds from Lancaster Sound, Canada and for the common eider (*Somateria molissima*) from Uummannaq, West Greenland suggest that the Pb concentrations in Lancaster Sound are abnormally high, possibly because these data are old (1977).

Age accumulation. In general, few data on the Pb concentrations in seabirds are suitable for evaluating the relationship between age and accumulation. Data for 25 gulls sampled in 1994 in four areas of Greenland as part of the AMAP program, however, were found suitable (Riget *et al.* 1997a) for addressing this question. The Pb levels in livers of these gulls were not correlated with the age of the birds. Limited data for composite samples of common eider from Pangnirtung, Sanikiluaq, and Uummannaq, long-tailed duck and red-breasted merganser (*Mergus serrator*) from Inukjuak, and Iceland gull (*Larus glaucoides*) from Uummannaq support this conclusion.

Geographical differences. No seabird species has been analyzed for Pb on a circumpolar basis. In general, however, seabirds from Russia have a higher Pb level than seabirds from other areas. This situation may reflect differences in sampling and analytical methodology rather than real differences in concentrations. The 1994 AMAP results on gulls from four regions in Greenland indicate higher Pb levels in East Greenland than in other regions of Greenland (Riget *et al.* 1997a).

Point sources. At the Black Angel mine in northwest Greenland, Pb levels in muscle, liver, and kidney from seabirds (common eider; glaucous gull, *Larus hyperboreus*; Iceland gull; and black guillemot, *Cepphus grylle*) were not elevated, whereas Pb concentrations in bone were two to ten times higher than at reference sites (Agger and Johansen 1992).

Cadmium

Most data for Cd in seabirds are from Greenland, Canada, and Norway. Some data are available for Iceland and Russia (Annex Table 7·A14). In muscle tissue, some extreme high values (up to 28 μg/g) are found in fulmar from Svalbard, Norway. High levels in muscle tissue are also found in seabirds from Lancaster Sound (northern fulmar, 2.59 μg/g; kittiwake, *Rissa tridactyla*, 1.62 μg/g; Brünnichs guillemot, *Uria lomvia*, 1.55 μg/g). Most other values of Cd in muscle are below 0.6 μg/g.

Cadmium concentrations generally decrease in the order of kidney > liver > muscle. In liver, Cd concentration ranges from below 1 μg/g in yearlings of most seabird species to about 10 μg/g in older birds, except for several seabird species from Svalbard where values up to 30 μg/g in fulmar have been reported. High values (approx. 20 μg/g) were also found in northern fulmar and Brünnichs guillemot from Lancaster Sound, Canada and in glaucous gulls from Avanersuaq, northwest Greenland. In kidney tissue, yearling seabirds have low levels, ~0.1 μg/g. The highest values are found in older birds with levels up to 80 μg/g in glaucous gull from the northern part of West Greenland. In five seabird species, Cd levels in bone tissue are low, ranging from below detection to 0.112 μg/g in black guillemot from Uummannaq, West Greenland.

Age accumulation. In cases where Cd concentrations are given for different age groups (mostly from Greenland), the levels generally increase with age (Nielsen and Dietz 1989). In the 1994 AMAP results from Greenland, Riget *et al.* (1997a) found that Cd concentrations in liver of glaucous gull and Iceland gull clearly increased with the age of the gulls. However, Norheim (1987) found no differences between juvenile and adult birds from Svalbard, including glaucous gull, Brünnichs guillemot, little auk (*Alle alle*), and common eider.

Geographical differences. No clear geographical differences are evident. However, in all cases the concentrations of Cd in birds from Lancaster Sound are higher than that in birds from Greenland. The levels in the birds from Svalbard are for some species lower or similar to those from Greenland and for other species higher than those from Greenland. Several comparisons of seabirds of the same age can be made for the Greenland area. The highest levels were found in northwest Greenland (Nielsen and Dietz 1989, Dietz *et al.* 1996). In contrast, the levels in gulls sampled as part of the 1994 AMAP program were very similar from north to south in West Greenland (Riget *et al.* 1997a).

Point sources. At three mine sites in Greenland, results of a monitoring program indicate no elevation of Cd in seabirds relative to background levels (Hansen and Asmund 1986, Asmund *et al.* 1988, Johansen *et al.* 1991, 1995, Agger and Johansen 1992).

Mercury

Hg concentrations generally decrease in the order liver > kidney > muscle (Annex Table 7·A14). The highest concentration in muscle tissue (2.19 µg/g) was found in great cormorant from Kangaatsiaq, West Greenland. This value is based on one specimen, which also had the highest concentration in liver and kidney. Other values range from below detection to about 1 µg/g in red-breasted merganser from Kangiqsualujjuaq, Canada, and glaucous gull from Avanersuaq, northwest Greenland. Several seabird species have been analyzed for Hg in liver tissue. Levels in liver range from 0.1 to 3 µg/g. Except for a high value of 28.1 µg/g in kidney tissue of one specimen of great cormorant from West Greenland, the levels were intermediate between the values in liver and muscle, ranging from 0.053 µg/g in one-year-old kittiwake to 2.33 µg/g in older glaucous gulls. Seven seabird species have been analyzed for Hg in feathers. The highest levels were found in kittiwake (5.5 µg/g fresh weight).

Age accumulation. Few data are available for assessing age accumulation of Hg in seabirds. Levels in older kittiwake, Brünnichs guillemot, and black guillemot from Lancaster Sound, Canada and black guillemot from West Greenland were generally higher than those in younger birds. No accumulation with age was found in livers of glaucous and Iceland gulls from Greenland in the new AMAP data (Riget *et al.* 1997a), or in five seabird species from Greenland (Nielsen and Dietz 1989).

Geographical differences. No clear conclusion can be drawn about geographical differences of Hg in seabirds across the Arctic. Mercury levels in seabirds from Arctic Canada are similar to those from Greenland (Nielsen and Dietz 1989), Svalbard, and northern Norway. Thompson *et al.* (1992a) found higher Hg concentration in feathers of birds from northwest Iceland than those from northern Norway. Within Greenland, Nielsen and Dietz (1989) and Dietz *et al.* (1996) found higher concentrations in birds from northwest and northeast Greenland than in those from south Greenland. Riget *et al.* (1997a) presented new AMAP data, and found higher concentrations in livers of glaucous gulls from East Greenland but no elevated levels in northwest Greenland compared with south Greenland.

Temporal trend. Appelquist *et al.* (1985) used feathers to test for a historical trend of Hg concentration in black guillemot and Brünnichs guillemot from Greenland, and found the levels to be almost constant during this century, though slightly increasing in black guillemot.

Selenium

Concentrations of Se generally decrease in the order kidney > liver > muscle. Several seabird species have been analyzed for Se in muscle, liver, and kidney tissues (see Annex Table 7·A14). The levels in muscle range from below detection to 4.95 µg/g in older kittiwake. In seabird liver, the levels ranged from 1.03 in little auk to 21.4 µg/g in common eider, both from Uummannaq, West Greenland.

Selenium levels in kidney ranged from 1.24 µg/g in ivory gull from Uummannaq, West Greenland to 27 µg/g in glaucous gull from Ittoqqortormiit, East Greenland.

Age accumulation. Very few data exist for assessing possible age accumulation of Se in seabirds. Riget *et al.* (1997a), using new AMAP data, found an increase in Se with age in glaucous and Iceland gulls.

Geographical differences. Few data are available for comparing Se levels in seabirds from around the Arctic. Selenium and Hg concentrations are highly correlated and, as is the case for Hg, the levels of Se in seabirds from Canada, Greenland, and Svalbard are similar. Within Greenland, Nielsen and Dietz (1989) and Riget *et al.* (1997a) found a tendency toward higher concentrations in birds from northwest Greenland and northeast Greenland than in those from south Greenland.

7.6.5.8. **Marine mammals**

A substantial amount of data is available on the concentrations of metals in Arctic marine mammals. Because Cd and Hg bioaccumulate strongly and because Hg biomagnifies, much more emphasis has been placed on obtaining body burdens of metals in biota of higher trophic levels.

Lead

Data on tissue concentrations of Pb in marine mammals have been produced for ten species including polar bear. Most data are from Canada and Greenland, some are from Alaska and Norway, and almost none are from Iceland and Russia. Concentrations of Pb range from below detection (<0.010 µg/g) to 0.083 µg/g in tissues of ringed seals. For other pinniped species (harp seal, *Pagophilus groenlandicus*; fur seal, *Callorhinus ursinus*; and walrus), they range between 0.010-0.328 µg/g. High mean values (up to 1.0 µg/g) are found in some polar bear tissue from Svalbard (Norheim *et al.* 1992), but some of these results are old and questionable. A recent study by Wagemann *et al.* (1996) includes data for ringed seals, beluga whales (*Delphinapterus leucas*), and narwhals and shows no Pb levels higher than 0.083 µg/g. Dietz *et al.* (1996) reviewed previous Pb data from Greenland and found that mean Pb concentrations for seals did not exceed 0.058 µg/g. Differences between tissues are not as apparent for Pb as for other heavy metals (Annex Table 7·A15).

Age accumulation. Wagemann *et al.* (1983, 1990, 1996) found no correlation between Pb levels and the size or age of narwhals and belugas from Canada. A similar lack of correlation has been reported for white-beaked dolphins (*Lagenorhynchus albirostris*) and pilot whales (*Globicephala melaena*) from Newfoundland by Muir *et al.* (1988). The data by Norheim *et al.* (1992) for Pb in liver and kidney of adult and juvenile polar bears from Svalbard also show no correlation between age and tissue Pb concentrations.

Geographical differences. Possible geographical differences are difficult to evaluate as analytical problems may well obscure true differences because Pb levels in marine mammals are low, and frequently below or close to the detection limit. Wagemann *et al.* (1996), however, found significantly higher Pb concentrations in muscle and liver tissues of ringed seals from the western Canadian Arctic compared with those from the eastern Canadian Arctic, but no differences in muscle and liver of belugas from the northern Canada (Wagemann *et al.* 1990, 1996). The levels of Pb were higher in belugas from Hudson Bay than those from the Mackenzie Delta. Also, belugas from St. Lawrence River, Quebec, were significantly higher (0.104-0.159 µg/g) than belugas from the Arctic as a whole, which was attributed to anthropogenic input of Pb. Lead levels in liver of dolphins off Newfoundland (Muir *et al.* 1988) are of the same order of magnitude, but are lower in muscle. In liver of ringed seals from the northern part of central West Greenland, Pb levels were half those in seals from central East Greenland (Dietz *et al.* 1996).

Temporal trends. Few reliable data are available to evaluate temporal trends of Pb in marine mammal tissues. Wagemann *et al.* (1996) found no temporal trends in muscle and liver of narwhal from the eastern Canadian Arctic over a span of 14 years (1979-1993).

Cadmium

Data for Cd in marine mammal tissues are available for six seal species, six whale species, and polar bears (Annex Table 7·A15). The concentrations vary considerably among tissue types, but generally decrease in the order kidney > liver > muscle.

In ringed seals, the highest Cd concentrations (geometric means) are found in animals from northwest Greenland (111 µg/g in kidneys, 36.7 µg/g in livers). There were no clear differences among seal species. In whales, the highest concentrations (63.5 µg/g) were found in kidneys of narwhals from Pond Inlet (Wagemann *et al.* 1983, 1996). Minke whales (*Balaenoptera acutorostrata*) had the lowest kidney concentrations (4.24 µg/g), whereas belugas, pilot whales, harbour porpoises, and white-beaked dolphins had intermediate concentrations. Few data are available for bowhead whales (*Balaena mysticetus*). Two adult specimens (Becker *et al.* 1995b) had considerable Cd in the liver (17.5-18.9 µg/g). Other tissues such as blubber and skin (mattak) are very low in Cd, and in most cases the levels are close to or below detection.

The concentrations of Cd in polar bears are remarkably low compared with seals and whales, even though the polar bear is a top predator. This has been explained by the feeding habits of the polar bear. It feeds virtually exclusively on ringed seal blubber, which has a very low Cd concentration (e.g., Norstrom *et al.* 1986, Braune *et al.* 1991, Dietz *et al.* 1995). Cadmium concentrations in kidneys, liver, and muscle of polar bears are 19.7, 1.67, and 0.024 µg/g, respectively.

Age accumulation. In Annex Table 7·A15, metal levels are compiled for a large range of ages. For geographical and temporal comparisons, it is of major importance to assess the relationship between age and metal levels. Cadmium is virtually absent from the body at birth, both in humans and marine mammals (WHO 1992a, 1992b, Dietz *et al.* 1995, 1997a). The increase of Cd concentration with age in seals has been documented by several authors (Sergeant and Armstrong 1973, 1980, Wagemann *et al.* 1996). Dietz *et al.* (in press) also found a highly significant correlation of Cd with age, but the concentration increased only until the seal reached adulthood and decreased thereafter. Consequently, Cd concentrations in old seals were one third lower than in young adult seals.

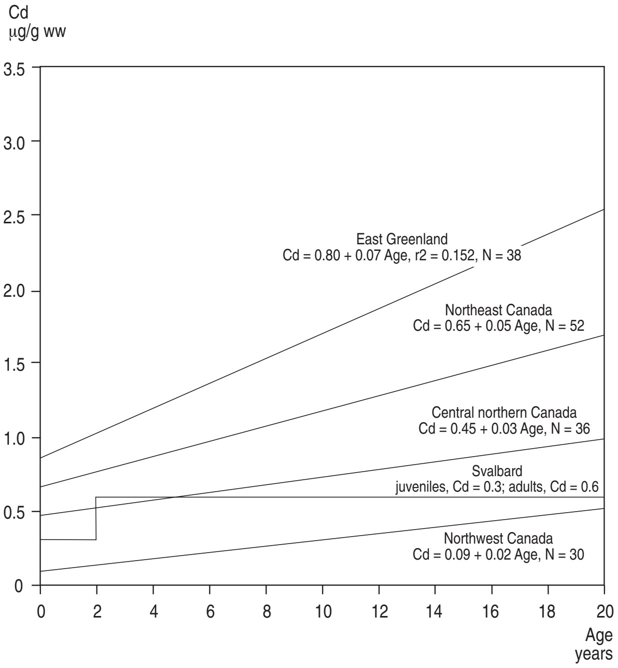


Figure 7·44. Age accumulation of Cd in liver of polar bears from different Arctic regions. (After Dietz *et al.* 1995a).

Both Wagemann *et al.* (1983, 1990, 1996) and Muir *et al.* (1988) have noted the increase in Cd concentrations with age in whales from Canadian waters. In a study on minke whales, belugas, and narwhals from Greenland, Hansen *et al.* (1990) showed that in eight out of 18 animals, Cd concentrations increased significantly with age and that middle-aged whales had the highest values. Paludan-Müller *et al.* (1993) found that Cd increased until the age of four years in harbour porpoises from Greenland waters. In animals older than four years, muscle and liver concentrations reached a constant level, whereas kidney levels decreased.

An increase of Cd concentrations in polar bear tissues with age has been documented by several authors (Norstrom *et al.* 1986, Braune *et al.* 1991, Dietz *et al.* 1995) (Figure 7·44), but no evidence of a decrease in old bears has been reported.

Geographical differences. Concentrations of Cd in marine mammal tissues increase from west to east in the Canadian High Arctic for ringed seal, beluga, and polar bear (Norstrom *et al.* 1986, Braune *et al.* 1991, Wagemann *et al.* 1996). This trend could be extended to cover West Greenland, for ringed seals and polar bears, but not for belugas (Annex Table 7·A15 and Figures 7·45 to 7·47). Wagemann *et al.* (1996) explain the geographical trend within Canada in terms of geological differences between the western and the eastern Canadian Arctic. The eastern part of Canada consists essentially of Precambrian igneous and metamorphic rocks with a higher Cd concentration than is found in the Postcambrian unmetamorphosed sedimentary rocks characteristic of the western Canadian Arctic. The correlation between regional geology and levels in marine biota indicates that natural sources may be the primary cause of the high levels in the eastern Arctic. In Greenland, no significant differences in the Cd levels of bottom sediment were found for the different geological structures (Loring and Asmund 1996). On the other hand, Cd levels are generally highest in ringed seals and polar bears from northwest Greenland compared with areas farther south (Dietz *et al.* 1996).

A five-fold difference in Cd tissue concentrations occurs across the Arctic and depends on the areas, species, and tissues examined. Cadmium levels in ringed seals and polar bears in central East Greenland are somewhat lower than in northwest Greenland (Avanersuaq); they are even lower around Svalbard. On the east cost of Greenland (Ittoqqortoormiit, Danmarkshavn, and Kong Oscars Fjord), only minor differences in the concentrations of Cd found in ringed seal occur between areas, with no discernible north-south trend.

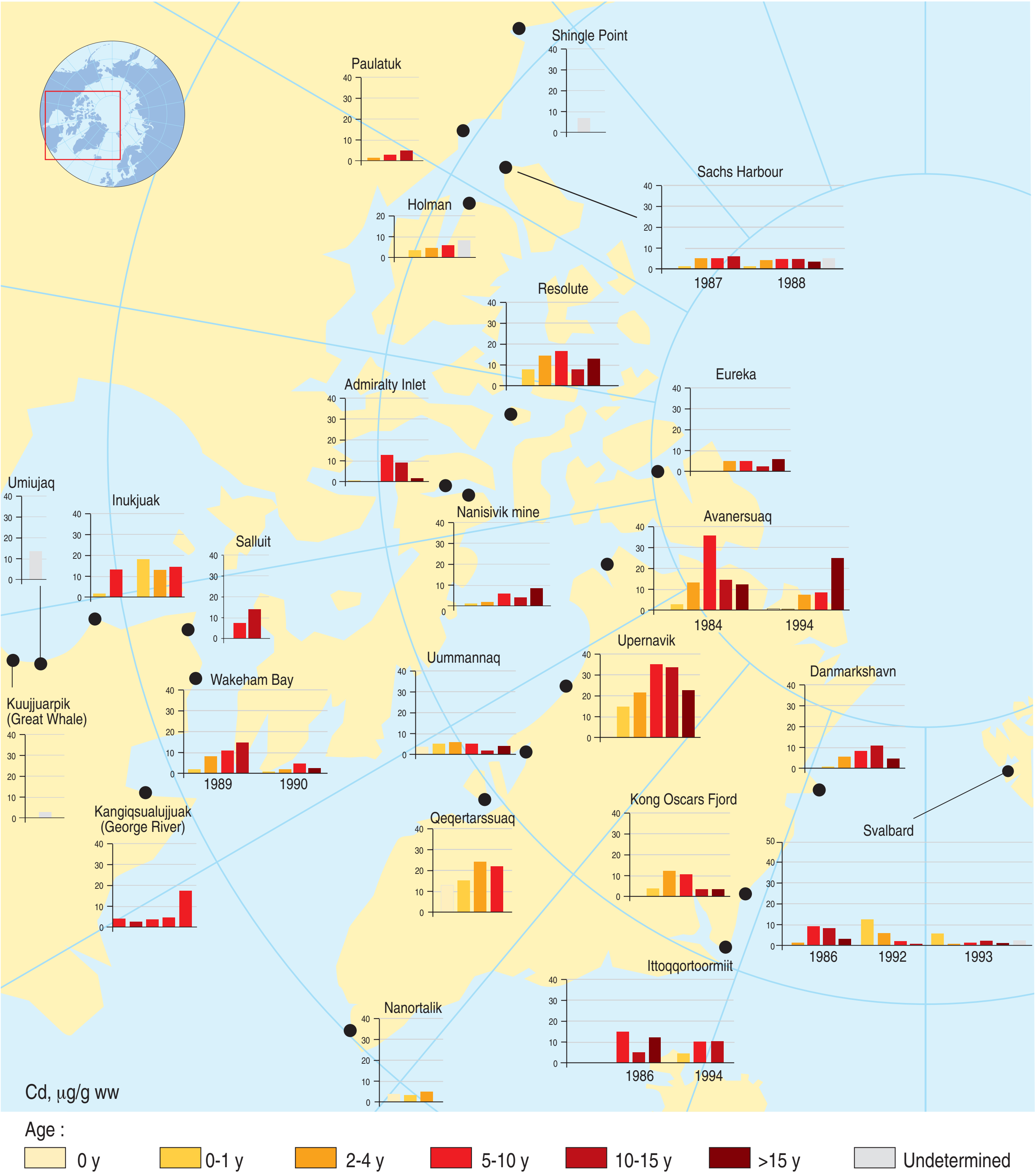


Figure 7·45. Distribution of Cd levels in liver tissue of ringed seal (*Phoca hispida*) of different ages (years). Plots show selected data (geometric mean values) from Annex Table 7·A15. (Sources of data: see Annex Table 7·A15).

Cadmium concentrations in ringed seals from the Arctic are many times higher than those reported for ringed seal from the Gulf of Finland and the Gulf of Bothnia (Helle 1981, Perttilä 1986, Frank *et al.* 1992; data not included in Annex Table 7·A15). Specifically, concentrations are approximately 15 times higher in muscle, 16-75 times higher in liver, and 24-42 times higher in kidney. Johansen *et al.* (1980) also concluded that Cd was highest in Arctic seals. Concentrations of Cd in harbour porpoises from Greenland waters also had ten times higher Cd levels than did those from European waters (Paludan-Müller *et al.* 1993). These differences may be partly explained by differences in available food items. Species such as parathemisto, other crustaceans, and Arctic cod may be important Cd sources in the Arctic. The higher levels in Arctic marine mammals may also be a consequence of slower growth rates in the Arctic. Slow growing poikilothermic organisms accumulate metals over a longer period of time before being eaten (Muir *et al.* 1996). In Svalbard, conditions seem different. Although situated far north, Svalbard is strongly influenced by the relatively warm Gulf Stream, leading to faster growth of the lower food chain

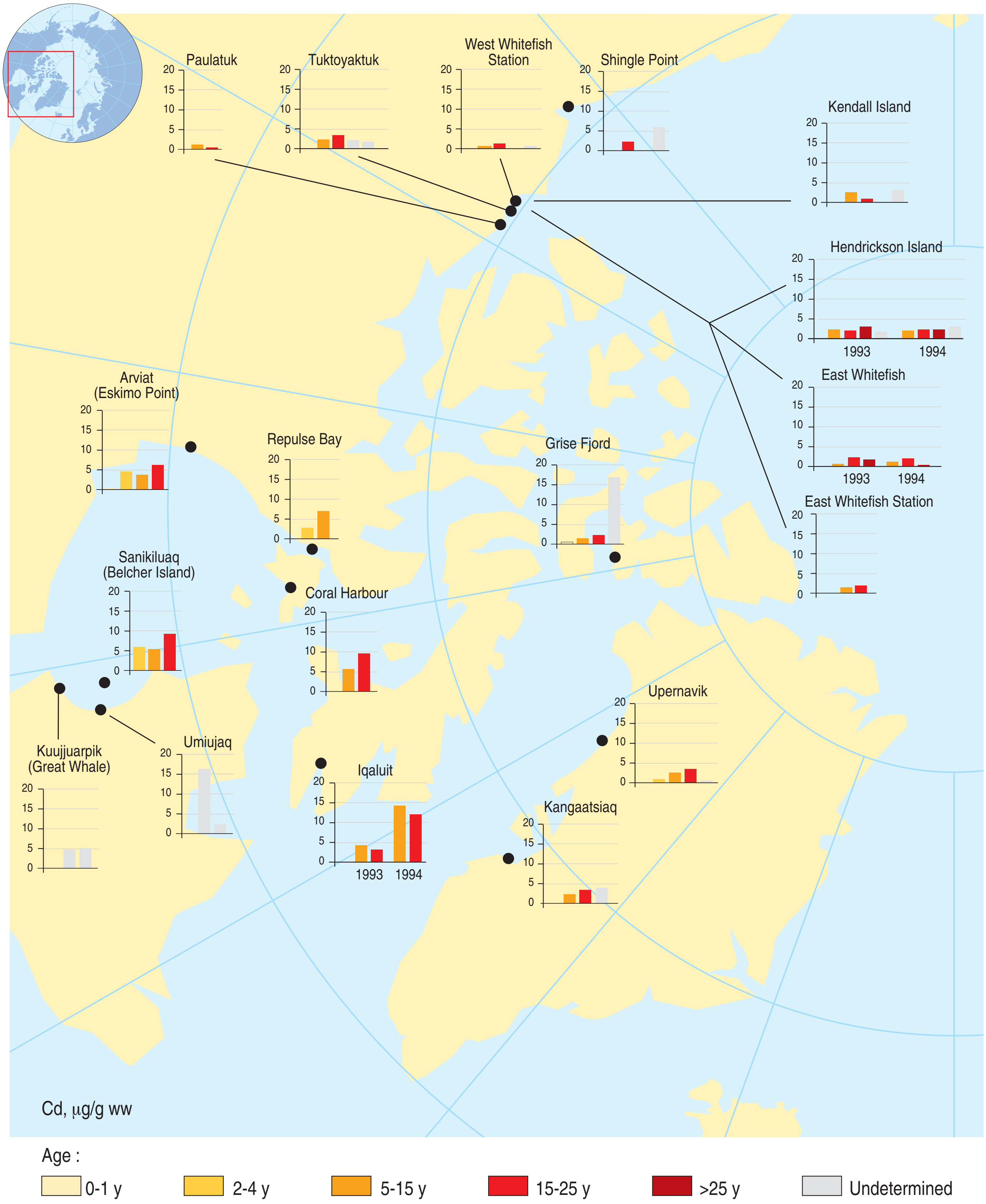


Figure 7·46. Distribution of Cd levels in liver tissue of beluga whale (*Delphinapterus leucas*) of different ages (years). Plots show selected data (geometric mean values) from Annex Table 7·A15. (Sources of data: see Annex Table 7·A15).

organisms, which ultimately results in lower body burdens of metals.

Temporal trends. Limited information is available on temporal trends in marine mammals. Wagemann *et al.* (1996) is the only study addressing this question. They found no temporal trends of Cd in tissues of narwhal and beluga sampled over approximately 14 years in the Canadian High Arctic. Hansen (1988) analyzed hair samples from seals and humans from the 15th century and found the levels in humans were not significantly different from those today, whereas levels in seal furs were significantly higher (2.6 times) now than in the old samples. However, the value of hair as a satisfactory indicator of Cd exposure was questioned by the author.

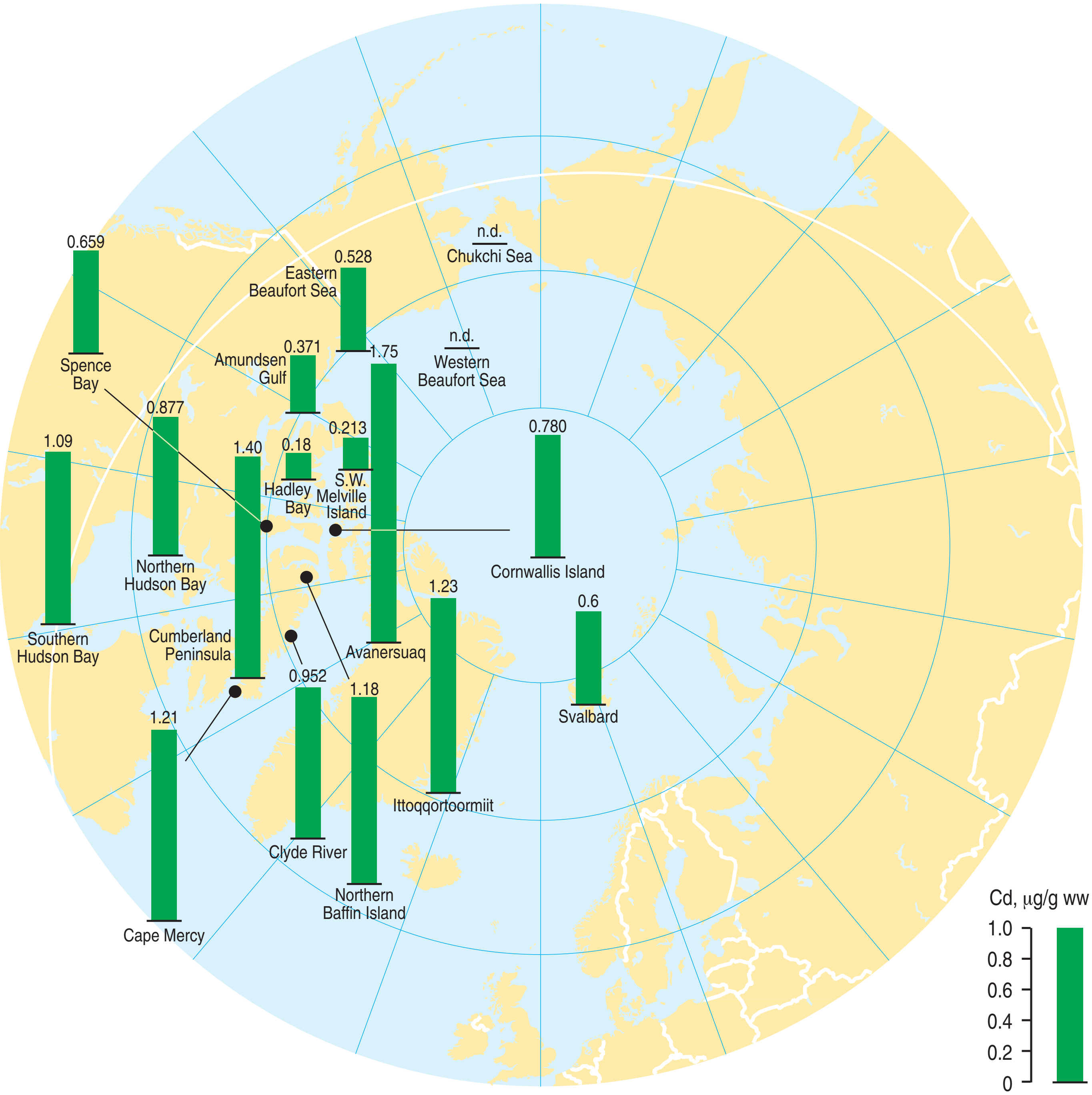


Figure 7·47. Distribution of Cd levels (age adjusted to 6.9 years, Svalbard adults) in liver tissue of polar bear (*Ursus maritimus*). (Source of data: Braune *et al.* 1991, Norheim *et al.* 1993, Dietz *et al.* 1995, 1996).

Point sources. Ringed seals caught near the Nanisivik Pb-Zn Mine in Strathcona Sound and at a reference site in Admiralty Inlet had comparable Cd levels (Wagemann 1989). Similarly, at the three mine sites in Greenland noted earlier, Cd has been monitored in ringed seals, and at no site was it elevated relative to reference sites (Hansen and Asmund 1986, Asmund *et al.* 1988, Johansen *et al.* 1991, 1995, Agger and Johansen 1992).

Mercury

Mercury data from eight seal species, eight whale species, and polar bears are compiled in Annex Table 7·A15, showing, as for Cd, considerable differences among tissue types. Concentrations generally decrease in the order liver > kidney > muscle. In polar bears, however, the highest Hg levels are found in the kidney.

Concentrations of Hg in seal and whale muscle frequently exceed 0.50 µg/g ww, particularly in older individuals because of the accumulation of Hg with age. The highest mean Hg concentrations (0.72 µg/g in muscle, 32.6 µg/g ww in liver) are found in ringed seals in the western Canadian Arctic (Wagemann *et al.* 1996). Up to 219 µg/g have been measured in the liver tissue of a ringed seal from Sachs Harbour. This is also the population where the highest mean values have been recorded (Wagemann *et al.* 1996; see Figure 7·48, next page). Very high levels, 143 µg/g on average, have been reported for bearded seal from the Amundsen Gulf (Smith and Armstrong 1975, 1978). Mercury can be transferred from the mother to the fetus during gestation, which is apparently not the case for Cd (Wagemann *et al.* 1988). Almost all the Hg in muscle tissue is present as methylmercury, whereas in liver tissue methylmercury seldom ex-

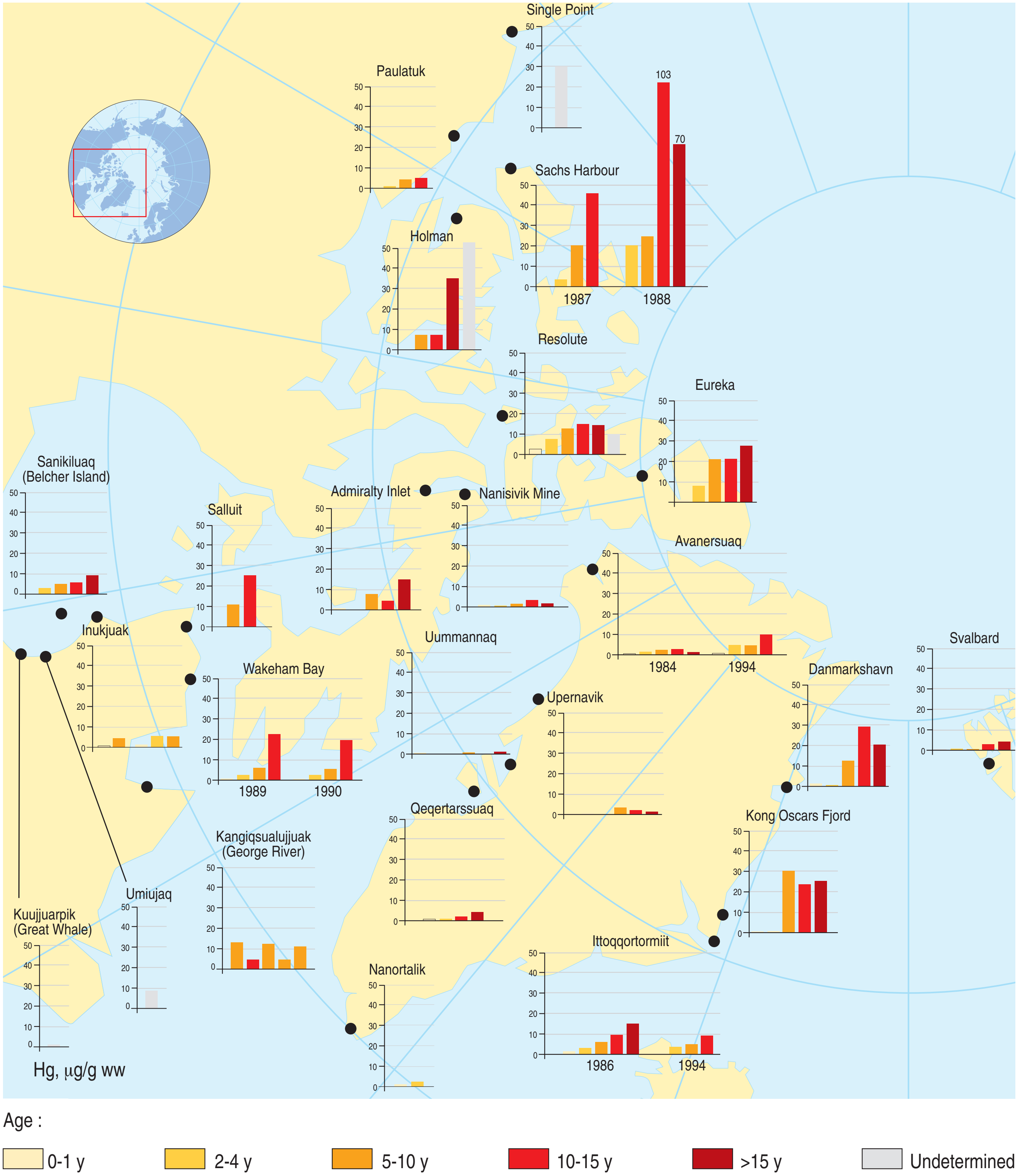


Figure 7·48. Distribution of Hg levels in liver tissue of ringed seal (*Phoca hispida*) of different ages (years). Plots show selected data (geometric mean values) from Annex Table 7·A15. (Sources of data: see Annex Table 7·A15).

ceeds 2 µg/g, even when the total Hg concentration is high (Dietz *et al.* 1990, Figure 7·49). In the kidney tissue of adult seal and whale, organic Hg is 10-20% of total Hg, whereas in polar bears it is 6% (Dietz *et al.* 1990, Figure 7·49). This fraction is consistently higher, up to 70%, in young seals and walrus (Born *et al.* 1981, Wagemann *et al.* 1988, Dietz *et al.* 1990).

In general, Hg levels are low in baleen whales. Maximum levels in livers of bowhead, fin (*Balaenoptera physalus*), and minke whales do not exceed 0.30, 0.546, and 0.452 µg/g respectively. In contrast, Hg levels in most toothed whales are comparatively high. In livers of white-beaked dolphins, harbour porpoises, narwhals, belugas, and pilot whales, maximum means are 0.831, 8.21, 10.8, 42.4, and 280 µg/g, respectively. The extremely high values of total Hg in pilot whales were from samples taken in 1977 near the Faeroe Islands, and are well above the concentrations measured subsequently in that area (38 to 84 µg/g). In those 1977 samples, methylmercury levels up to 35±10 µg/g were reported (Julshamm *et al.* 1987).

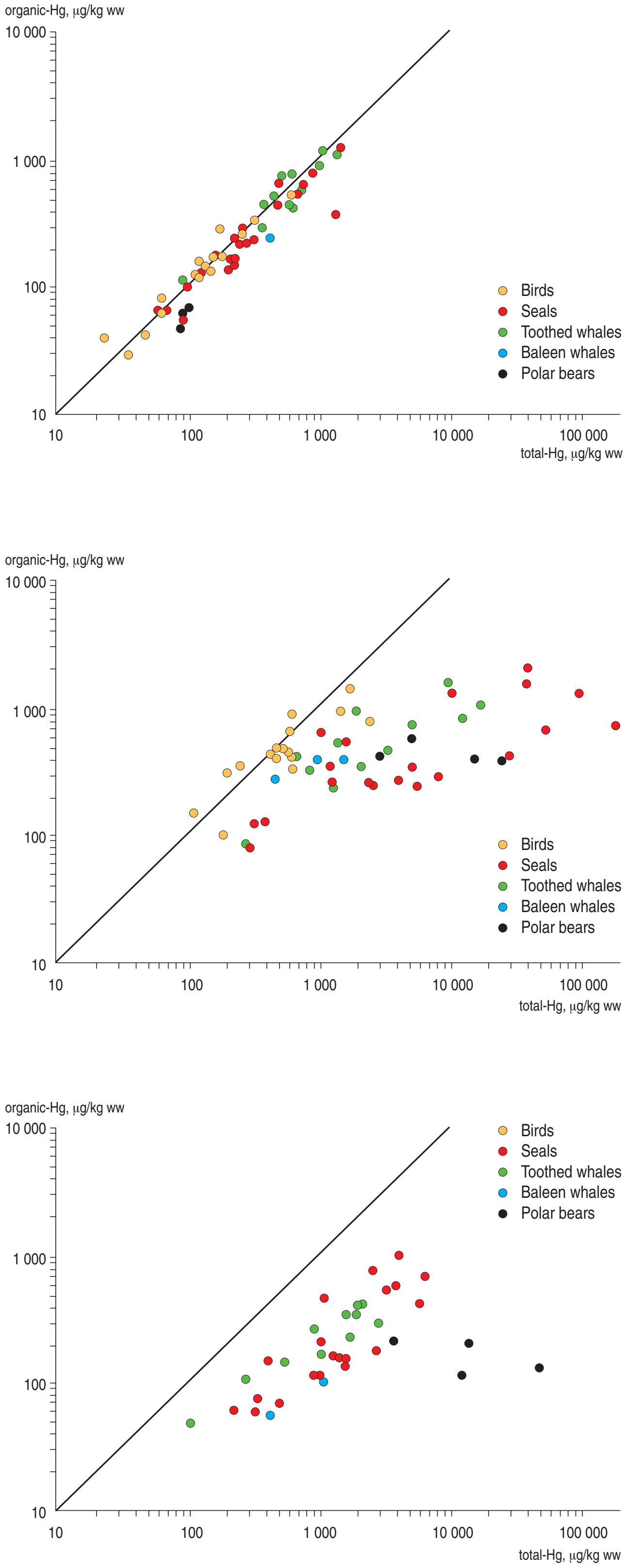


Figure 7·49. Organic mercury versus total mercury in muscle, liver and kidney tissue of Greenlandic marine animals (after Dietz *et al.* 1990). The lines indicate 100% organic mercury.

Polar bears also have quite high Hg levels in their livers. Mean concentrations up to 53.0 µg/g have been reported in polar bear livers from the eastern Beaufort Sea (Norstrom *et al.* 1986, Braune *et al.* 1991).

Age accumulation. As was the case for Cd, the concentration of Hg increases in marine mammals with age. This has been well documented for seals (Smith and Armstrong 1975, 1978, Wagemann *et al.* 1996) and for whales in the Canadian Arctic (Gaskin *et al.* 1972, 1979, Wagemann *et al.* 1983, 1990, 1996, Muir *et al.* 1988). Hansen *et al.* (1990) found that Hg was positively correlated with age in muscle, liver, and kidney of narwhals, in liver and kidney of belugas, and in liver of minke whales. Paludan-Müller *et al.* (1993) found that in harbour porpoises from Greenland waters, Hg increased with age in muscle, skin, and perhaps also in kidney until four years of age. By contrast, Hg appears to increase in whale liver throughout the entire lifetime of the animal. Julshamm *et al.* (1987) found that Hg concentration in both the muscle and liver of pilot whales reached a plateau once the whales reached a certain size. An increase of Hg concentrations in the liver and kidneys of polar bears with age has also been documented (Norstrom *et al.* 1986, Braune *et al.* 1991, Dietz *et al.* 1995; Figure 7·50).

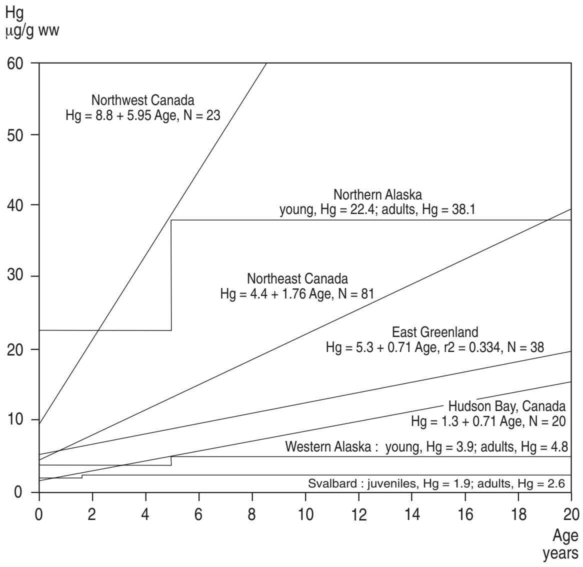


Figure 7·50. Age accumulation of Hg in liver of polar bears from different Arctic regions. (After Dietz *et al.* 1995).

Geographical trends. Based on a sample size of two, the concentration of Hg in ringed seals in Alaska appears to be high (1.52 and 3.52 µg/g in a one- and a two-year-old ringed seal, respectively) (Zeisler *et al.* 1993). These levels correspond well with concentrations found in the western Canadian Arctic, West Greenland, Svalbard, and northern Norway (Smith and Armstrong 1975, 1978, Johansen *et al.* 1980, Carlberg and Boler 1985, Wagemann 1989, Wagemann *et al.* 1996, Skaare 1994, Dietz *et al.* 1996, 1997a; Annex Table 7·A15). Smith and Armstrong (1978) found no significant differences in methylmercury in liver or muscle tissue between areas as widely separated as Holman in the western Canadian Arctic and Pond Inlet on northern Baffin Island. However, Eaton and Farant (1982) point out, based on the results from Smith and Armstrong (1978), that the age accumulation of Hg in liver of ringed seals increased from east to west. The increasing trend of Hg in ringed seal liver from eastern to western Canada is supported by a recent study of Wagemann *et al.* (1996), but no similar trend was found in kidney and muscle (Annex Table 7·A15 and Figure 7·48).

Juvenile ringed seals from Jarfjord in Norway had lower Hg concentrations in liver and kidney than did ringed seals from Canadian and Greenland waters. Ringed seals from the Gulf of Bothnia and Gulf of Finland (data not included in Annex Table 7·A15), on the other hand, exceed even the highest values reported for the Arctic (Helle 1981, Perttilä *et al.* 1986, Frank *et al.* 1992). Higher levels in seals from northwestern European waters than in those from the Arctic

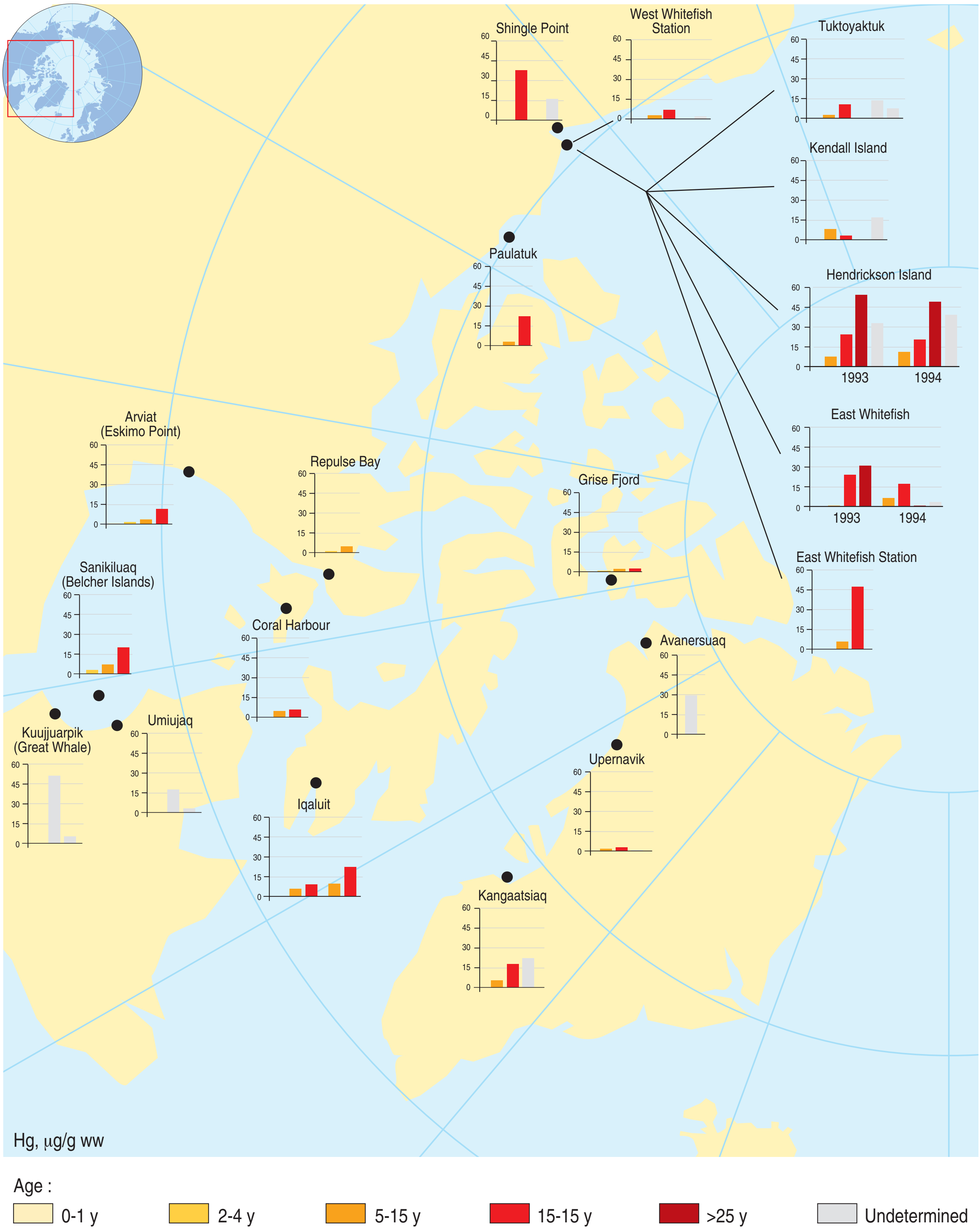


Figure 7·51. Distribution of Hg levels in liver tissue of beluga whale (*Delphinapterus leucas*) of different ages (years). Plots show selected data (geometric mean values) from Annex Table 7·A15. (Sources of data: see Annex Table 7·A15).

are also indicated by studies involving grey seal (*Halichoerus grypus*) and harbour seal (*Phoca vitulina*) (Law *et al.* 1991, Frank *et al.* 1992), and in harbour porpoise from the two areas (Paludan-Müller *et al.* 1993). In seals found dead off the coast of the Netherlands, the Hg concentration in liver was extremely high, 257-326 µg/g (Koeman *et al.* 1972), most likely due to anthropogenic sources. Yamamoto *et al.* (1987) presented data for two adult Weddell Sea seals from the more pristine Antarctic. They found 0.11-0.16 and 3.1-8.5 µg/g Hg in muscle and liver, respec-

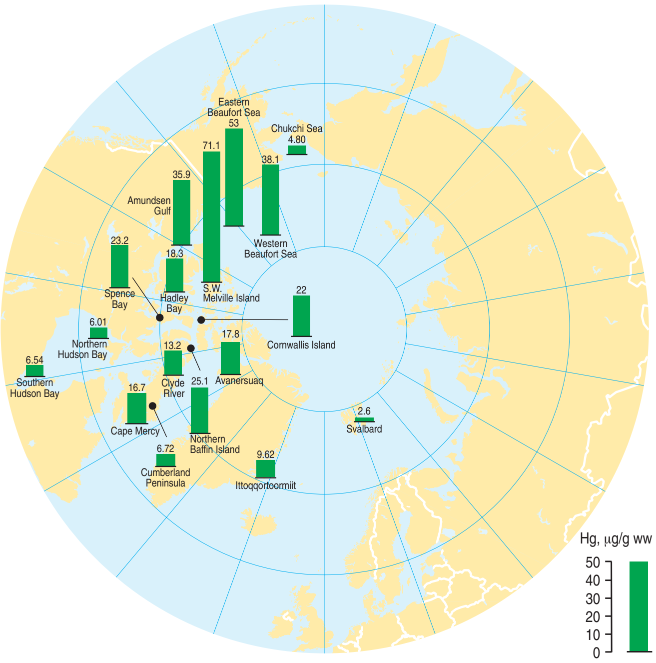


Figure 7·52. Distribution of Hg levels (age adjusted to 6.9 years, Svalbard adults) in liver tissue of polar bear (*Ursus maritimus*). (Source of data: Braune *et al.* 1991, Norheim *et al.* 1993, Dietz *et al.* 1995, 1996).

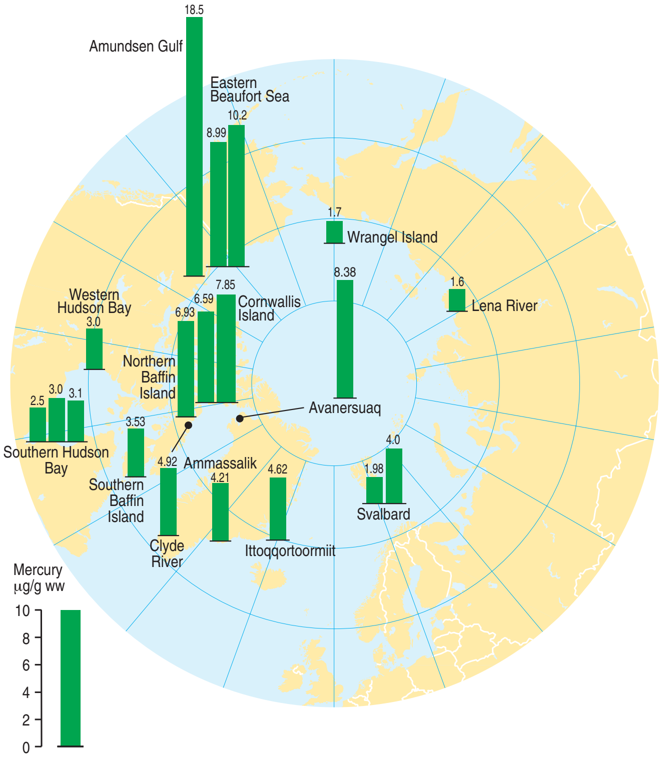


Figure 7·53. Distribution of Hg levels in hair of polar bear (*Ursus maritimus*). (Sources of data: Eaton and Farant 1982, Renzoni and Norstrom 1990, Born *et al.* 1991).

tively, which is consistent with background concentrations in the Arctic.

The high Hg levels in the western part of the Canadian Arctic have also been documented in beluga whale tissue (muscle, liver, and kidney) and are most clearly seen in liver (Annex Table 7·A15 and Figure 7·51). The different ages of the animals in the two areas were a confounding factor in the comparison between the western and the eastern Arctic. However, comparisons of the accumulation rates of Hg in liver (data were not given for muscle and kidney) of belugas from western and eastern Canada showed that the accumulation rate was more than three times higher in western Canada (Wagemann *et al.* 1996).

Polar bears from northern Alaska have higher levels of Hg in liver and muscle tissue than do those in western Alaska (Lentfer and Galster 1987). Bears from western Arctic Canada accumulated Hg faster in their livers than did polar bears from eastern Arctic Canada, whereas bears from Hudson Bay had slightly lower concentrations (Norstrom *et al.* 1986, Braune *et al.* 1991). The eastward decreasing trend in Hg extended to Greenland and Svalbard (Norheim *et al.* 1992, Dietz *et al.* 1995; Annex Table 7·A15; Figure 7·52). The observed geographic trend has also been documented for Hg levels in polar bear hair (Eaton and Farant 1982, Renzoni and Norstrom 1990, Born *et al.* 1991; Figure 7·53). Norstrom *et al.* (1986) suggested that the differences found between western Arctic Canada and eastern Arctic Canada were most likely caused by higher Hg levels in the ringed seal food chain caused by higher natural levels in the sediments (and consequently in the lower food chain) of the Melville Island area. The evidence presented above indicates a geographic trend in ringed seal liver, but these differences are not apparent in kidney and muscle. Too few data are available to evaluate geographical trends in ringed seal blubber, which is the preferred food of polar bears.

Temporal trends. Few investigations on temporal trends have been conducted for marine mammals. In a recent study, Wagemann *et al.* (1996) concluded that livers of recently collected (1987-1993) ringed seals from western Canada accumulating Hg approximately three times faster than those collected in 1972-1973 (from Smith and Armstrong 1975, 1978; Figure 7·54). In contrast, muscle levels were significantly lower (43%) in the recent samples, but so was the mean age (42% lower), which means that muscle levels were approximately the same in the two samples. A comparison of liver values in ringed seals from Avanersuaq, northwest Greenland taken in 1984 and 1994 (for age groups: 2-4, 5-10 and >15 years) showed increasing Hg levels, with concentrations 2.3-6.9 times higher in the recent samples. Over the same time period, differences were minor in one-year old seals from Avanersuaq and Nanortalik, south Greenland. This was true for all age groups from Ittoqqortoormiit, central East Greenland as well. A recent study on Atlantic walruses did not show any temporal trend (Wagemann *et al.* 1995, 1996), but the authors suggest that the low Hg levels in walruses and the relatively short sampling time (six years) may explain the lack of trend. The same study shows that the Hg accumulation rate in livers of belugas from the western Canadian Arctic increased by a factor of 1.7-1.8 from 1981 to 1993 (Figure 7·54). Kidney levels were also significantly higher (1.74 times), but muscle samples were only 25% higher, which was not significant.

Total Hg concentrations were also higher in tissues of narwhals in 1992-1993 compared with 1978-1979, although the size of the animals indicated that the recent group of animals was somewhat older (1978-1979, 376 cm; 1992-1994, 420 cm). Pilot whales from the Faeroe Islands have been sampled from 1977 to 1987 (Julshamm *et al.* 1987, Caurant *et al.* 1994). Regressions of Hg concentration in liver against time showed no significant trend. To evaluate differences over longer periods of time, hair samples from seal fur clothing from the 15th century have been analyzed and compared with values obtained for recent fur samples, both from Greenland. The recent values are approximately four times higher (Hansen 1988). When hair concentrations are compared between Inuit mummies (1470±50 AD) and contemporary Greenlandic residents, a three-fold increase in Hg concentration is suggested (Hansen 1988). As the Inuit in the 15th century lived almost exclusively on the marine food chain, as

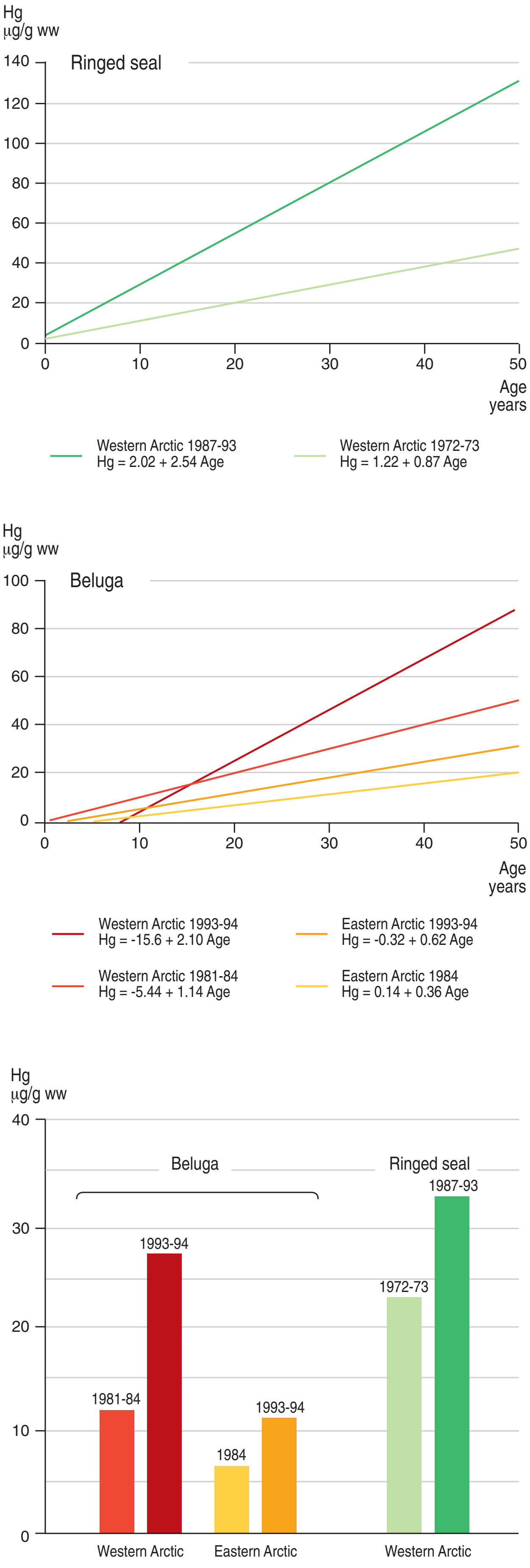


Figure 7·54. Age accumulation and temporal comparison of mean concentrations of Hg in liver tissue of ringed seal (*Phoca hispida*) and beluga whale (*Delphinapterus leucas*) from the Canadian Arctic. (After Wagemann *et al.* 1996).

shown by the stable isotope ^{13}C/^{12}C ratios, and today's population lives on a mixed diet of Arctic and imported food, the increase could have been even larger if compared on a same-food basis. Analysis results from Ammassalik, East Greenland from the 19th century were all lower than those from the 15th century, which was attributed to a severe famine in the areas where the samples were taken. The 15th century hair samples are approximately five times higher in Hg than the present-day Danish level (Hansen 1988).

Selenium

Selenium analyzes have been completed for most of the species examined for Hg because Se is regarded as an antagonist to Hg, and data interpretation is consequently most valid when both concentrations are known. In most Arctic samples, Se is present in a substantial surplus compared with Hg on a molar basis (Dietz *et al.* 1997a). In ringed seals, Se concentrations in liver and kidney are similar, ranging between 0.93 and 18.4 µg/g over different age groups and areas. The levels in muscle are approximately ten times lower than in liver and kidney. In whales, Se concentrations decrease in the order liver > kidney > muscle, whereas in polar bears the order is kidney > liver > muscle.

Mean liver concentrations of Se have been recorded as high as 18.4 µg/g in ringed seals, and the highest levels among the other seven seal species are found in bearded seal (mean = 34.4 µg/g). Walrus livers contain lower Se concentrations (highest mean 3.14 µg/g) than those of ringed seal. Baleen whales are generally low in Se with a highest mean of 1.78 µg/g. In toothed whales, the Se level may be as high as 24.2 µg/g in belugas. In pilot whales that are known to feed on cephalopods, values are even higher. Samples from 1977 (the year in which high Hg levels were measured) indicate Se concentrations as high as 172 µg/g in liver, whereas in other years they were only 12-23 µg/g. Selenium levels up to 23.4 µg/g have been recorded in the livers of polar bears.

Age accumulation. The molar ratio between Se and Hg in liver tissue of marine mammals is frequently close to 1, at least at concentrations above approximately 3 µg/g (Koeman *et al.* 1973, 1975, Hansen *et al.* 1990, Braune *et al.* 1991, Becker *et al.* 1995a, Dietz *et al.* 1995). Selenium acts as a detoxifying agent by binding Hg as mercuric selenide (HgSe – tiemannite) (Iwata *et al.* 1981, Joiris *et al.* 1991). It is, therefore, not surprising that Se and Hg accumulate with age in liver tissue. Selenium is generally not found to accumulate to the same degree with age in muscle and kidney as in liver (Wagemann *et al.* 1983, 1990, Hansen *et al.* 1990, Paludan-Müller *et al.* 1993, Dietz *et al.* 1995). The concentration of Se in the skin of harbour porpoises is positively correlated with age (Paludan-Müller *et al.* 1993).

As outlined above, Hg and Se are associated in liver. The same geographical trends are therefore found for Se in liver as for Hg, whereas no clear geographical differences can be detected in other tissues. Hence, Wagemann *et al.* (1996) found higher Se levels in ringed seals and belugas from the western Canadian Arctic than in those from the eastern Canadian Arctic. This study found no geographical trends for muscle or skin tissue. Concentrations of Se in marine mammals have always been found to be higher in northwest Greenland compared with other Greenland areas, except for ringed seals, where higher levels in central East Greenland have been reported (Dietz *et al.* 1996).

Temporal trends. No information has been published on temporal trends for Se. However, it can be expected that the trends found for Hg in liver tissue of ringed seal, belugas, and narwhals will also be found for Se, since these two elements are highly correlated. Comparisons of ringed seals from Avanersuaq, northwest Greenland showed an increase from 1.6 to 4.0 times for the age groups above two years. The younger age groups (0 and 1 years), however, showed a decrease from 0.6 to 0.9 (Annex Table 7·A15).

7.7. Biological effects (acute, short-, and long-term toxicity; reproductive, physiological, and behavioral effects; etc.)

A generalized overview of reported effects threshold levels for Hg, Cd, Pb and Se in tissues of main animal groups is presented in Table 7·22. These thresholds, together with information presented in section 7.5, have been compared with concentrations of metals observed in Arctic biota in the following assessment of the potential for effects in different Arctic ecosystems.

7.7.1. Effects on terrestrial ecosystems

No biological effects attributable specifically and unambiguously to heavy metal pollution in the Arctic terrestrial ecosystem have been reported. There is concern, however, over the high concentrations of Cd in the kidneys of some Arctic mammals (primarily reindeer) and some gamebirds (ptarmigan). Even though some concentrations exceed threshold values believed to cause kidney dysfunction, no such effects have ever been observed or reported.

Cases of severe ecological damage, however, have been reported in the Arctic terrestrial system, specifically around the huge metal smelter complexes of the Kola Peninsula, Russia. The complete collapse of healthy ecosystems for tens of kilometers around these smelters is primarily the result of pollution by high concentrations of SO_2 and NO_x leading to extreme cases of acidification. Although the concentrations of metals can be very high in these areas, they are only a contributing factor (along with hydrocarbons, anthropogenic chemicals, and non-chemical factors) to the damage observed.

7.7.2. Effects on freshwater ecosystems

The only biological effect thought to be attributable specifically and unambiguously to heavy metal pollution in the Arctic freshwater ecosystem is the decline in the ringed seal population of the Lake Saimaa, Finland. There it is thought that Hg contamination (and lack of sufficient Se to detoxify it) has rendered the seals more prone to premature and still births.

It is the view of Russian ecologists that severe ecological damage has occurred along portions of major Siberian rivers

Table 7·22. Overview of reported effects threshold levels for metals in tissues of main animal groups.

Metal	Group	Tissue	Concentration mg/kg ww[a]	Symptoms	Reference
Pb	Waterfowl	Blood	20-50	Subclinical poisoning	Pain 1996
			50-100	Clinical poisoning	
			> 100	Severe clinical poisoning	
		Liver	2-6	Subclinical poisoning	
			6-15	Clinical poisoning	
			> 15	Severe clinical poisoning	
		Bone	10-20 (dw)	Subclinical poisoning	
			10-20 (dw)	Clinical poisoning	
			> 20 (dw)	Severe clinical poisoning	
	Terrestrial birds	Blood	0.2-3	Subclinical poisoning	Franson 1996
			> 1-5	Toxic	
			> 5-10	Lethal	
		Liver	2-6	Subclinical poisoning	
			> 3-6	Toxic	
			> 5-20	Lethal	
		Kidney	2-20	Subclinical poisoning	
			> 3-15	Toxic	
			> 5-40	Lethal	
	Mammals	Liver	> 30 (dw)	Clinical signs	Ma 1996
		Kidney	> 90 (dw)	Clinical signs	
		Blood	> 10	Adverse effect on cognitive and behavioral performance	
Cd	Birds	Liver	> 40	Cadmium poisoning	Furness 1996
		Kidney	> 100	Cadmium poisoning	
	Marine mammals	Liver	> 20-200	Potential renal dysfunction	Law 1996
		Kidney	> 50-400	Potential renal dysfunction	
Hg	Birds	Liver	> 30	Lethal level in free ranging birds	Thompson 1996
		Kidney	> 30	Lethal level in free ranging birds	
		Egg	> 3.0	Detrimental effect upon free ranging bird hatching	
		Liver	> 30	Laboratory succumbed animals due to Hg intoxication	
		Kidney	> 30	Laboratory succumbed animals due to Hg intoxication	
		Egg	> 2.0	Detrimental effect upon experimental bird hatching	
	Terrestrial mammals	Liver	> 30	Lethal or harmful in free ranging wildlife	Thompson 1996
		Kidney	> 30	Lethal or harmful in free ranging wildlife	
		Liver	> 25	Laboratory succumbed animals due to Hg intoxication	
		Kidney	> 25	Laboratory succumbed animals due to Hg intoxication	
	Marine mammals	Liver	> 60	Liver damage	Law 1996
Se	Birds	Eggs	> 3	Deformed embryos	Heinz 1996
		Liver	> 9	Deformed embryos	
		Kidney	> 9	Deformed embryos	
		Blood	> 5-14	Lethal effect	
	Terrestrial mammals	Liver	> 7	Hepatic lesions	WHO 1997

a. Unless indicated as dw.

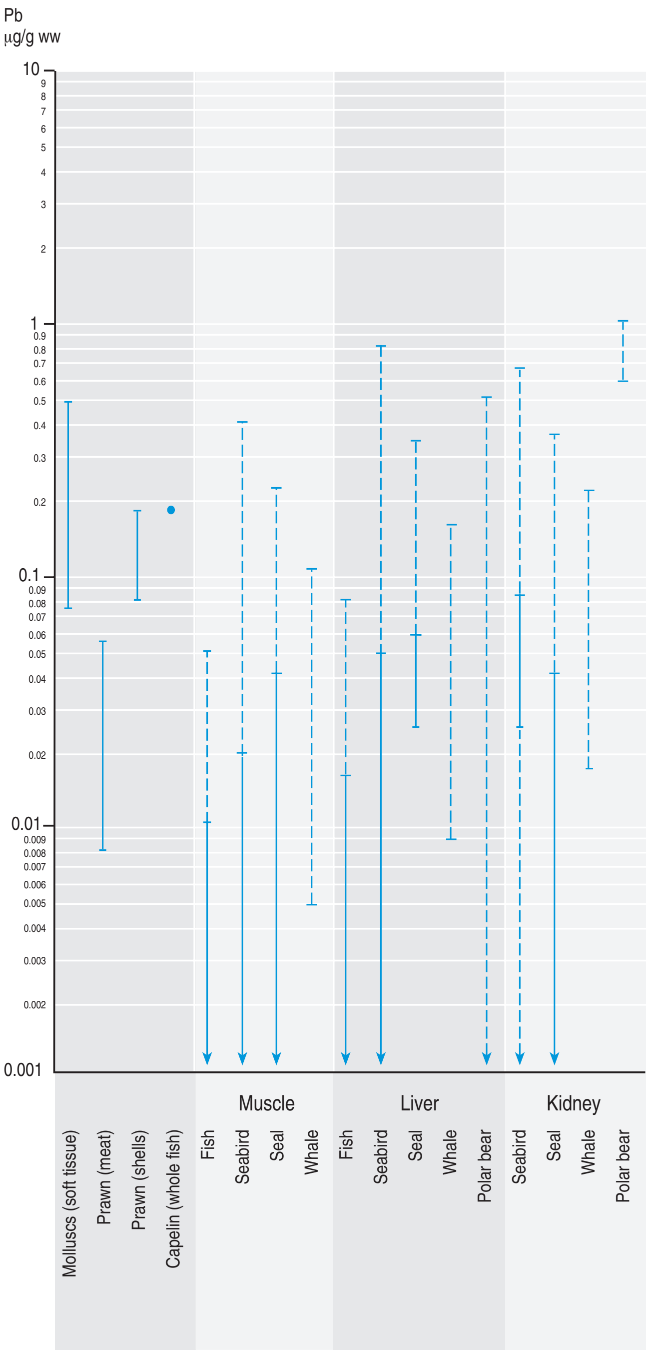


Figure 7·55. Summary of ranges of Pb levels (mean values) in Arctic marine organisms. Solid parts of the lines indicate ranges for Greenlandic data from Dietz *et al.* (1996), where the Pb analyses have been critically evaluated and only data for samples with n ≥ 8 were included.

and watercourses. As noted above for the terrestrial ecosystems, however, it is difficult to conclude that metal contamination is any more than one of many factors which have caused the problem. In many of the most-affected parts of the aquatic ecosystem, large inputs of sewage, petroleum hydrocarbons, sulfactants, refined petroleum products, acidifying compounds, and other anthropogenic chemicals are common. These substances are capable of causing the damage even without the additional negative consequences of metal pollution.

7.7.3. **Effects on marine ecosystems**

Lead

The overall Pb baseline levels in the Arctic are low. There is no indication that Pb levels increase in higher trophic levels. As lethal and sublethal effects of Pb have not been related to tissue burdens, potential effects cannot be derived from the levels measured. The only places where effects are likely to occur are hot spots such as mining areas and possibly some Russian estuaries.

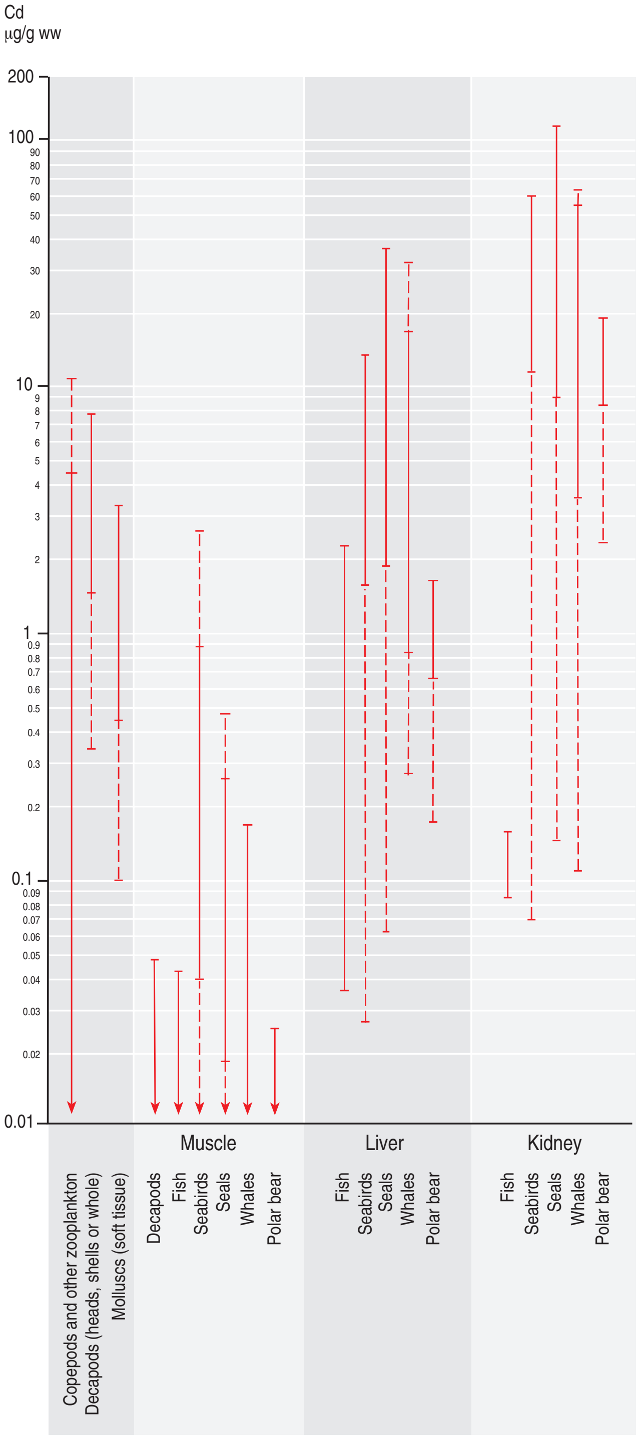


Figure 7·56. Summary of ranges of Cd levels (mean values) in Arctic marine organisms. Solid parts of the lines indicate ranges for Greenlandic data from Dietz *et al.* (1996), where the Cd analyses have been critically evaluated and only data for samples with n ≥ 8 were included.

Cadmium

According to the information from the literature summarized in section 7.5.2, effects on molting might occur in shrimps when body burdens exceed 10-40 µg/g. However, Cd in Arctic invertebrates is lower than this limit and therefore no effects are expected in this animal group. No information relating biological effects to body concentrations is available for fish; consequently, effects can not be evaluated

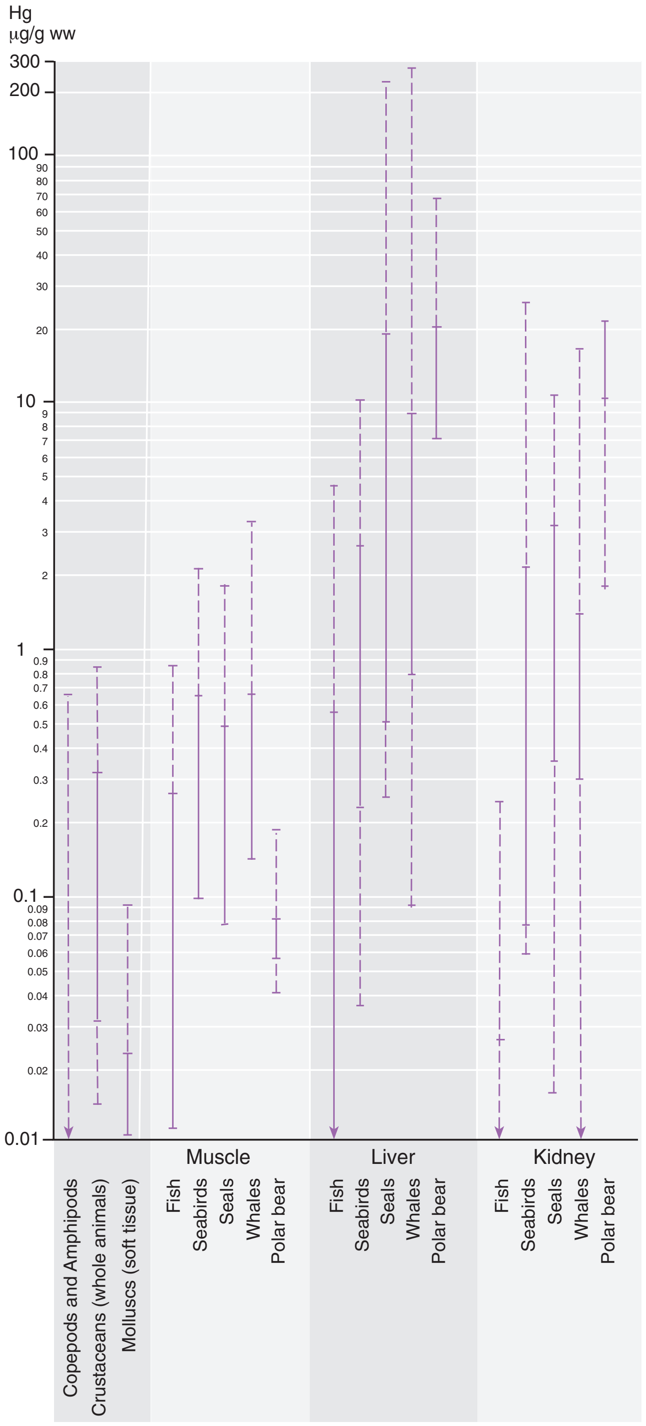


Figure 7·57. Summary of ranges of Hg levels (mean values) in Arctic marine organisms. Solid parts of the lines indicate ranges for Greenlandic data from Dietz *et al.* (1996), where the Hg analyses have been critically evaluated and only data for samples with n ≥ 8 were included.

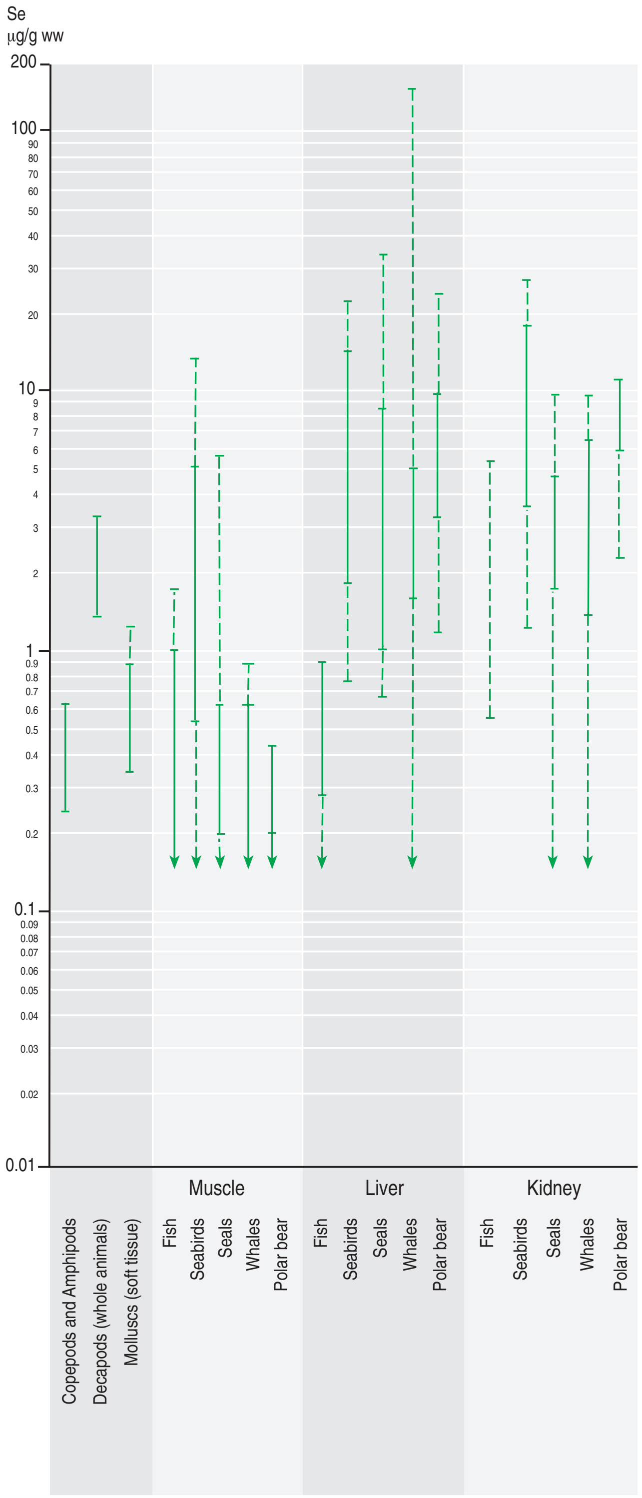


Figure 7·58. Summary of ranges of Se levels (mean values) in Arctic marine organisms. Solid parts of the lines indicate ranges for Greenlandic data from Dietz *et al.* (1996), where the Se analyses have been critically evaluated and only data for samples with n ≥ 8 were included.

for this animal group. Kidney damage can occur in pelagic sea birds at Cd concentrations of 60 µg/g, whereas the corresponding limit appears to be approximately 100 µg/g for marine mammals. The frequencies of occurrence of Cd concentrations above these limits were calculated for marine birds and seals from the Greenland area, where raw data were available and geographical comparisons indicated the highest Cd levels. In no cases were kidney levels above 60 µg/g for seabirds at ages 0 and 1 year. For birds older than one year, 7.3% of the birds had concentrations higher than 60 µg/g (see also Figure 7·59, next page). This percentage increased northward with 7.9, 13.3, and 18.8% in Uummannaq, Upernavik, and Avanersuaq, respectively. For glaucous gulls from Avanersuaq and kittiwakes from Upernavik, as many as 50% of the birds contained Cd exceeding the 60 µg/g limit.

A similar pattern was observed for seals in Greenland waters. The limit of 100 µg/g was exceeded in 10.3% of the ringed seals and in 5% of hooded seals (*Cystophora cristata*) analyzed, whereas no values above this threshold were reported for harp seals. Again the high values seem to be found in northwest Greenland, where as many as 37.0 and 35.2% of the ringed seal measurements from Avanersuaq and Uper-

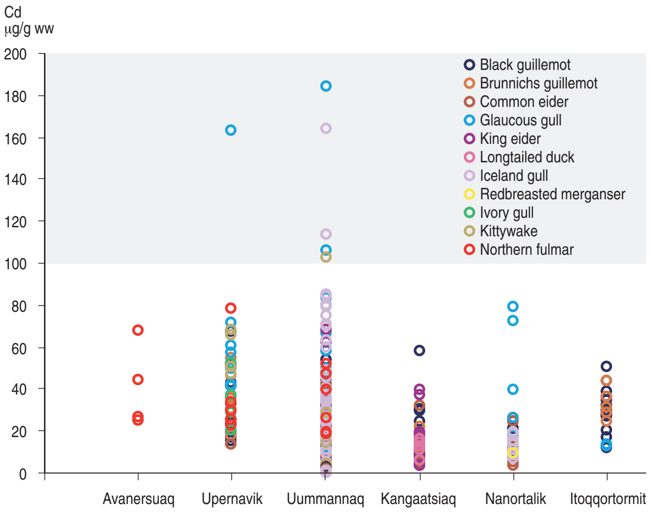


Figure 7·59. Cadmium levels in kidney of adult marine birds from Greenland. Levels associated with potential for kidney damage are above 100 µg/g ww. (Source of data: R. Dietz unpubl.).

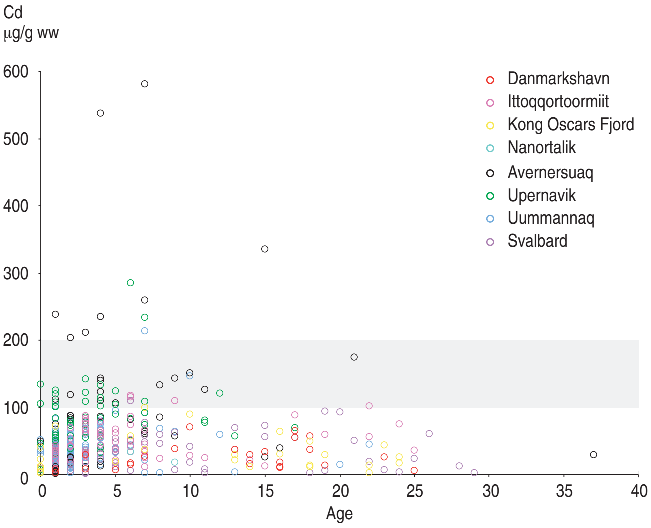


Figure 7·60. Cadmium levels in kidney of adult ringed seals from Greenland and Svalbard. Levels associated with potential for kidney damage are above 100-200 µg/g ww. (After Dietz *et al.* 1998 in press).

navik, respectively, were above the 100 µg/g limit (see Figure 7·60). In the human health chapter (chapter 12) the authors claim that effects may be found in humans even at 50 µg/g as the 'healthy worker effect' may cause threshold limits to be set too high. In the 10-15 year-age-group, as many as 66.7% of the seals from Avanersuaq exceeded the 100 µg/g limit, as did 50% of the 5-10 year-age-group from both Avanersuaq and Upernavik. In order to determine whether these apparently high levels pose a problem to the seals, samples of kidneys from this area in Greenland were examined in a pilot study by skilled pathologists. Fifthteen kidney samples were selected from three different concentration ranges (<2, 80-100, and >200 µg/g ww) for histopathological examination. The examination did not reveal any kidney damage even in the highest concentration group, suggesting that seals have an ability to detoxify substantial quantities of Cd (Dietz *et al.* 1998 in press).

In conclusion, Cd concentrations in kidneys of some species of marine birds and mammals, especially in northwest Greenland, are high enough to cause concern. Such high values may be seen in other Arctic areas as well. As only one recent pilot study has focused on possible biological effects related to high Cd tissue burdens (such as morphological damage at the microscopic level), it is at present impossible to make firm conclusions as to whether these animals are suffering from any effects of the high Cd levels. A preliminary conclusion of the pilot study is that Arctic seal populations may have developed strategies to effectively detoxify Cd.

Mercury

As no studies of the effects of chronic exposure to Hg have been related to tissue burdens, it is not possible to determine what tissue levels constitute a risk in marine biota. Experiments on certain animal groups have shown that the central nervous system and kidney are the organs most susceptible to damage from methylmercury and inorganic Hg, respectively. Even for humans, no guideline concentration limits are available for internal organs because the parameter most widely used to measure Hg burden is blood concentration.

It would therefore be relevant to carry out morphological effect studies in populations known to have high body burdens of Hg, for example animals from the western Canadian Arctic. Physiological and biochemical abnormalities include neurological impairment, reproductive effects, liver damage, and significant decreases in intestinal absorption. These effects of chronic exposure to Hg may appear at tissue burdens above 25-60 µg/g ww (see Table 7·22).

Selenium

Few studies of the effects of chronic exposure to Se have been related to the respective tissue burdens. In a previously cited experiment where hepatic lesions were found in rats fed on an Se-rich diet, the liver Se concentration was 7.34 µg/g. A substantial proportion of Arctic marine biota have liver concentrations above this limit, but Se in the marine ecosystem in general is higher than in the terrestrial ecosystem. The observed 1:1 molar ratio between Se and Hg commonly found in liver tissue of marine mammals suggests that these metals act as antagonists for each other and are detoxified as inert mercuric selenite (HgSe, or tiemannite).

7.8. Conclusions and recommendations

7.8.1. Conclusions

Details are given in Tables 7·23 and 7·24.

1. The distribution of heavy metals among the various environmental compartments of the Arctic is dynamic and driven by natural sources, processes, and environmental factors. Significant anthropogenic inputs of metals are detectable against the highly variable natural background on local scales, commonly in the order of tens of kilometers or less.
2. Metals are taken up by Arctic biota and their levels often reflect local geology or local anthropogenic activities.
3. The most important metals in the Arctic biosphere are Cd and Hg because they occur in some biota at concentrations that may have health implications for individual animals or may have implications for human consumers.
4. Few spatial or temporal trends are apparent in the existing data, largely due to poor temporal or spatial coverage or to unresolvable artifacts in the data related to differences in sampling, analytical, and reporting protocols.

7.8.1.1. Sources and transport of metals

1. During winter, about two-thirds of the heavy metals in air in the High Arctic are transported from Eurasia, particularly from the Kola Peninsula, the Norilsk region, the

Table 7·23. Overview of major conclusions related to terrestrial/aquatic ecosystems.

	Air and atmospheric deposition	Freshwater	Sediment	Soil	Vegetation	Birds	Fish	Mammals
1. *Concentrations of metals exceeding average global background*								
1.1. Regional	None	None	None	None	None	High Cd in kidney of ptarmigan	None	High Cd in reindeer kidney
1.2. Local	Kola Peninsula	At ‘hot spots’ of human/industrial activity such as smelter complexes of the Kola Peninsula (scale 10-100 km)	At ‘hot spots’ of human/industrial activity such as smelter complexes of the Kola Peninsula (scale 10-100 km)	At ‘hot spots’ of human/industrial activity such as smelter complexes of the Kola Peninsula (scale 10-100 km)	At ‘hot spots’ of human/industrial activity such as smelter complexes of the Kola Peninsula (scale 10-100 km)	Cd in kidney of ptarmigan high in Yukon/NWT	High Hg in fish of NWT	Unknown
2. *Spatial trend*								
2.1. Regional	South to north decrease	None	High Hg concentrations in Arctic lakes	Kola/Northern Scandinavia enrichment in Cu, Ni	Kola/Northern Scandinavia enrichment in Cu, Ni	None	None	None
2.2. Local	Decrease with distance from the source region	Enrichment near point sources (Russian rivers, lakes of Kola Peninsula)	Gradients near point sources (Cu, Ni)	Gradients near point sources (Cu, Ni)	Gradients near point sources (Cu, Ni)	Cd in ptarmigan high in Yukon Pb in herbivores in Russia greater in east than in west	Highest values for Hg occur in Canada	Cd in caribou kidney higher in Yukon than in NWT In Norway Cd in reindeer/moose kidney increases along N-S gradient In Russia Pb in reindeer liver/muscle higher in east than in west
3. *Temporal trends*								
3.1. Regional	A decrease over last 2 decades. Strong seasonal variation; the highest values seen in winter	Unknown	Recent Arctic-wide increase in surficial Hg concentrations in Arctic lakes	Unknown	Unknown	Unknown	Unknown	Unknown
3.2. Local	A decrease over the last 5 years in the Kola Peninsula. Winter concentrations higher than in summer	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown	Unknown
4. *Observed biological effects attributable to metals*								
4.1. Regional	Not applicable	Not applicable	Not applicable	Not applicable	Combined	Combined	Combined	Combined
4.2. Local	Not applicable	Not applicable	Not applicable	Not applicable	Combined	Cd in some ptarmigans from Yukon is high enough to cause kidney damage	Combined	Cd in some moose and reindeer from Yukon are high enough to cause kidney damage. Possibly still births of Lake Saimaa ringed seal due to Hg contamination.

Unknown: Insufficient data to reach a conclusion.
None: No trend or effect documented from a fair amount of data.
Combined: Contamination by metals may, and probably does contribute to some effects caused primarily by other factors.

Urals, and the Pechora Basin. Five to ten percent of these emissions are deposited in the High Arctic. The remaining one third of the heavy metals in High Arctic air in winter is transported from industrial regions in Europe and North America. In summer, local sources dominate the contamination of the High Arctic.

2. The highest concentrations of atmospheric heavy metals in Arctic air occur in the vicinity of smelter complexes on the Kola Peninsula and at Norilsk and result from emissions from these smelters.
3. Near point sources such as mine sites and some Russian estuaries, heavy metals exceed background levels up to 30 km from the source.
4. Riverine transport of heavy metals toward the Arctic Basin is approximately half the atmospheric contribution for metals like Cd and Pb, while for others such as Zn the riv-

Table 7·24. Overview of major conclusions related to the marine ecosystem.

	Seawater	Sediment	Algae	Invertebrates	Fish	Seabirds	Marine mammals
1. *Concentrations of metals exceeding average global background*							
1.1. Regional	None	None	None	Cadmium is higher in Arctic mussels and prawns	None	None i	Cadmium is higher n some Arctic marine mammals
1.2. Local	At point sources such as mining areas in Canada and Greenland as well as some Russian estuaries (scale <30 km)	At point sources such as mining areas in Canada and Greenland as well as some Russian estuaries (scale <30 km)	At point sources such as mining areas in Canada and Greenland as well as some Russian estuaries (scale <30 km)	At point sources such as mining areas in Canada and Greenland as well as some Russian estuaries (scale <30 km)	At point sources such as mining areas in Canada and Greenland as well as some Russian estuaries (scale <30 km)	None	None
2. *Spatial trends within the Arctic*							
2.1. Regional	None	Sediments seem to follow geographical provinces over the Arctic	None	Cadmium is higher in Greenland compared to Norway	Arctic cod from the Barents Sea are lower in Cd than the rest of the Arctic. The highest level for Cd and Hg are seen at several populations from northern latitudes in Canada and W Greenland. Hg is low in fish from the Greenland Sea	Seabirds from Lancaster Sound have high Cd levels, whereas birds from the Svalbard region have low	The highest Cd levels are recorded in northwest Greenland and the lowest are from Svalbard. Hg levels are highest in western Canada and in pilot whales from the Faeroe Islands
2.2. Local	Enrichment near point sources. Increasing natural Cd from inner fjords toward the sea	Enrichment near point sources. Increasing natural Cd from inner fjords toward the sea	Enrichment near point sources. Increasing natural Cd in individuals from inner fjords toward the sea	Enrichment near point sources. Increasing natural Cd in individuals from inner fjords toward the sea	Enrichment near point sources. Increasing natural Cd in stationary fish from inner fjords toward the sea	Not likely, due to the migratory behavior of birds	Not likely, as few marine mammals are stationary
3. *Temporal trends within the Arctic*							
3.1. Regional	Higher Pb levels than in prehistoric time	Hg in sediment from most Arctic areas show levels elevated or increasing in recent sediments	Unknown	Unknown	Unknown	Moderate to no increase in Hg	Hg in seals from northern Canada and Greenland as well as toothed whales are increasing
4. *Observed biological effects attributable to metals*							
4.1. Regional	Not applicable	Not applicable	Not likely	Not likely	Not likely	Cadmium is high enough in some areas to pose a threat for kidney damage	Cadmium is high enough in some areas to pose a threat for kidney damage
4.2. Local	Not applicable	Not applicable	Possible combined effect in some Russian estuaries	Possible combined effect in some Russian estuaries	Possible combined effect in some Russian estuaries	Cd is high enough in some areas to pose a threat for kidney damage	Cd is high enough in some areas to pose a threat for kidney damage

Unknown: Insufficient data to reach a conclusion.
None: No trend or effect documented from a fair amount of data.
Combined: Contamination by metals may, and probably does contribute to some effects caused primarily by other factors.

ers are more important, carrying five times the atmospheric load. Such mass balance calculations will change considerably with the distance from the sources and the time of year, since the source contributions are strongly seasonal.

7.8.1.2. Arctic metal concentrations relative to global background

1. Heavy metal concentrations in air in the High Arctic are one order of magnitude lower than concentrations in other remote locations and about two orders of magnitude lower than the concentrations around major point sources in the Kola Peninsula. Air concentrations measured on the Kola Peninsula are comparable with the concentrations in the most polluted regions of Europe and North America.
2. Background levels in soil, lakes, rivers, and oceans generally fall within the global ranges.
3. Cadmium levels in some terrestrial birds and mammals are high compared with global background, as are Hg levels in some freshwater fish. Cd levels in marine organisms from large parts of the Arctic exceed global background. Mercury and Se levels in marine mammals are high, but do not exceed the highest global levels. Lead levels in large parts of the Arctic are at the lower end of global background.

7.8.1.3. Spatial trends within the Arctic

1. The concentrations in surface deposition around the sources, e.g., on the Kola Peninsula, decrease by between one and two orders of magnitude within 10-100 km from the emission source. The concentrations within the large area of the High Arctic are uniformly distributed, varying by a factor of 2-3.
2. The concentrations of trace elements in marine sediments are dependent on local geology, particle size, the amount of organic matter, and anthropogenic influence. The background geographical distribution of Pb, Cd, Hg, and Cu in marine sediments is related to the geological provinces of the Arctic.
3. Regional geographical differences in metal concentrations of benthic flora and fauna as well as in those of fish are not very apparent. Seasonal and local trends for a metal like Cd for some stationary marine species are larger than regional differences in baseline data. No geographical differences can be observed in fish species. For marine birds, only Cd shows a geographical trend, seeming to be highest in northwest Greenland and the Lancaster Sound area. For ringed seals, beluga whales, and polar bears, Cd levels have been shown to be highest in eastern Arctic Canada and northwest Greenland. Mercury levels for the same group of animals have proven to be highest in the western Canadian Arctic, decreasing toward south and east. Geology, food constitution, and growth processes linked to temperature are possible explanations for these differences.

7.8.1.4. Temporal trends within the Arctic

1. The concentration of heavy metals measured in subarctic air has decreased during the last two decades. All the heavy metals show strong seasonal variation in the High Arctic.
2. Mercury in Arctic sediments shows an increase over time, indicating a widespread regional process. As the anthropogenic fluxes does not show the same pattern, further investigations are needed before firm conclusions can be drawn.
3. Temporal trend data are scarce in Arctic biota. There is some evidence of Hg increasing by a factor 2-3 in some marine mammals within the last two decades. Only liver, and in certain cases kidney, shows such increase. It remains uncertain, however, whether this is a real increase or reflects year-to-year variation. Mercury concentrations in human and seal hair from the 15th century are 2-3 times lower than present-day samples.

7.8.1.5. Observed biological effects and health aspects attributable to metals

7.8.1.5.1. Observed biological effects

1. Health effects have so far not been investigated in Arctic biota. However, Cd levels in some caribou, moose, and ptarmigan from the Yukon Territory as well as those in seabirds and marine mammals from northwest Greenland may be high enough to cause kidney damage. It is likewise uncertain whether Hg poses a health threat to the most highly exposed groups of marine mammals, those in the western Canadian Arctic as well as pilot whales from the Faeroe Islands. However, there are indications that Se is present in concentrations that can protect against Hg poisoning.

7.8.1.5.2. Tissue burdens of metals relative to national standards

1. Relative to the concentration limits proposed by the Nordic Council of Ministers for Cd in kidney, liver, and muscle tissue, all caribou in Canada as well as most game birds and marine mammals across the Arctic have excessive levels of Cd. In almost all cases, Pb levels in marine organisms from the Arctic are well below food standard limits; however, this is not the case for hot spot areas such as mining areas and some Russian estuaries. No standard limits are given for Se in food, but in some cases, human intake of Se is estimated to be high.

7.8.2. Recommendations

1. No additional heavy metal data should be collected for AMAP applications until unified, standard protocols for sampling, analysis, and reporting are established for the AMAP countries or unless it can be clearly shown in advance that data to be obtained are sufficiently tied, through a comprehensive QA/QC protocol, to coherent standards or methods that ensure final intercomparability of datasets.
2. Mass balance studies for heavy metals in the air, rivers, and ocean across the Arctic should be conducted. Increased source receptor modeling is required to develop quantitative strategies for the basis of formulating policy decisions among states.
3. The data gaps for biota should be filled, but priority should be given to metals and organisms for which there are concerns for biological effects. The lack of data from e.g. marine mammals of Russian waters should be addressed.
4. Studies on the processes behind the geographical differences and trends observed in the Arctic should be initiated and supported.
5. Additional data should be acquired to elucidate Cd and Hg time trends in a wide number of Arctic ecosystem compartments. Processes behind potential trends should be studied to resolve whether the changes are natural or man-made.
6. Health effects should be studied in Arctic species having body burdens containing Cd and Hg levels of concern.

For recommendations on tissue burdens of metals relative to national standards see chapter 12 (Human Health).

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Annex

Contents