Chapter 9

Acidifying Pollutants, Arctic Haze, and Acidification in the Arctic

Contents

9.1. Introduction

The emissions of acidifying compounds, sulfur dioxide (SO_2) and nitrogen oxides (NO_x), have increased since pre-industrial times as fuel combustion, population and industrial activity have increased worldwide. These pollutants occur in considerable quantities and cause adverse effects on human health and the natural environment. Sulfur emissions are mainly associated with point sources such as power plants, smelters, pulp and paper mills, and oil and gas processing. In contrast, nitrogen oxides are derived not only from power generating stations but also from diffuse emissions, which are less easy to quantify.

During the 1970s, it was discovered that even remote parts of the Arctic were influenced by air pollution. Part of this is due to human activities at lower latitudes. There is a strong link between the regional acid deposition problem and the Arctic haze phenomenon, which was first observed in the 1950s and further studied in the mid-1970s. Within the Arctic itself, there are only a few, but significant, sources of acidifying air pollutants: the non-ferrous smelters on the Kola Peninsula and in Norilsk, which have very large emissions of sulfur in particular. Consequently, the areas of greatest concern with respect to acidification in the Arctic are the northern areas of Finland and Norway and the regions in Russia surrounding the large smelter complexes. At the Second Ministerial Conference on the Arctic Environment, Ministers agreed to accord acidification a priority status under AMAP in a subregional context.

Due to the sensitivity of the ecosystems in the Arctic, some effects of acidification can also be found in low-deposition areas dominated by long-range transported pollutants. Adverse effects of acidifying pollutants have been noted in fish populations in acidified lakes, and in forests and natural vegetation, including lichens. Sulfate and black carbon aerosols from emissions of acidifying compounds have the potential to alter the radiative forcing of the Arctic atmosphere, and hence the climate.

Critical loads of deposition, or critical concentration levels are terms which quantitatively describe the sensitivity of natural systems to anthropogenic exposure. The critical load concept is presently used to formulate strategies for air pollution control within the United Nations Economic Commission for Europe (UN ECE) Convention on Long-Range Transboundary Air Pollution (LRTAP). The application of the critical load concept to northern environments has to be done with caution because the criteria or data upon which the critical load calculations are based may not properly take into account the conditions and relationships prevailing in the Arctic. The data from the AMAP program allow the methods for calculating critical loads to be adapted to accommodate the characteristics of the extreme Arctic climatic conditions.

General climatic, biological and biogeochemical conditions specific to the acidification phenomenon in the Arctic and subarctic environments have been discussed by Nenonen (1991). In addition, a review of the environmental consequences of anthropogenic acidification in Fennoscandia and the Kola Peninsula has previously been given by Kinnunen and Nenonen (1993).

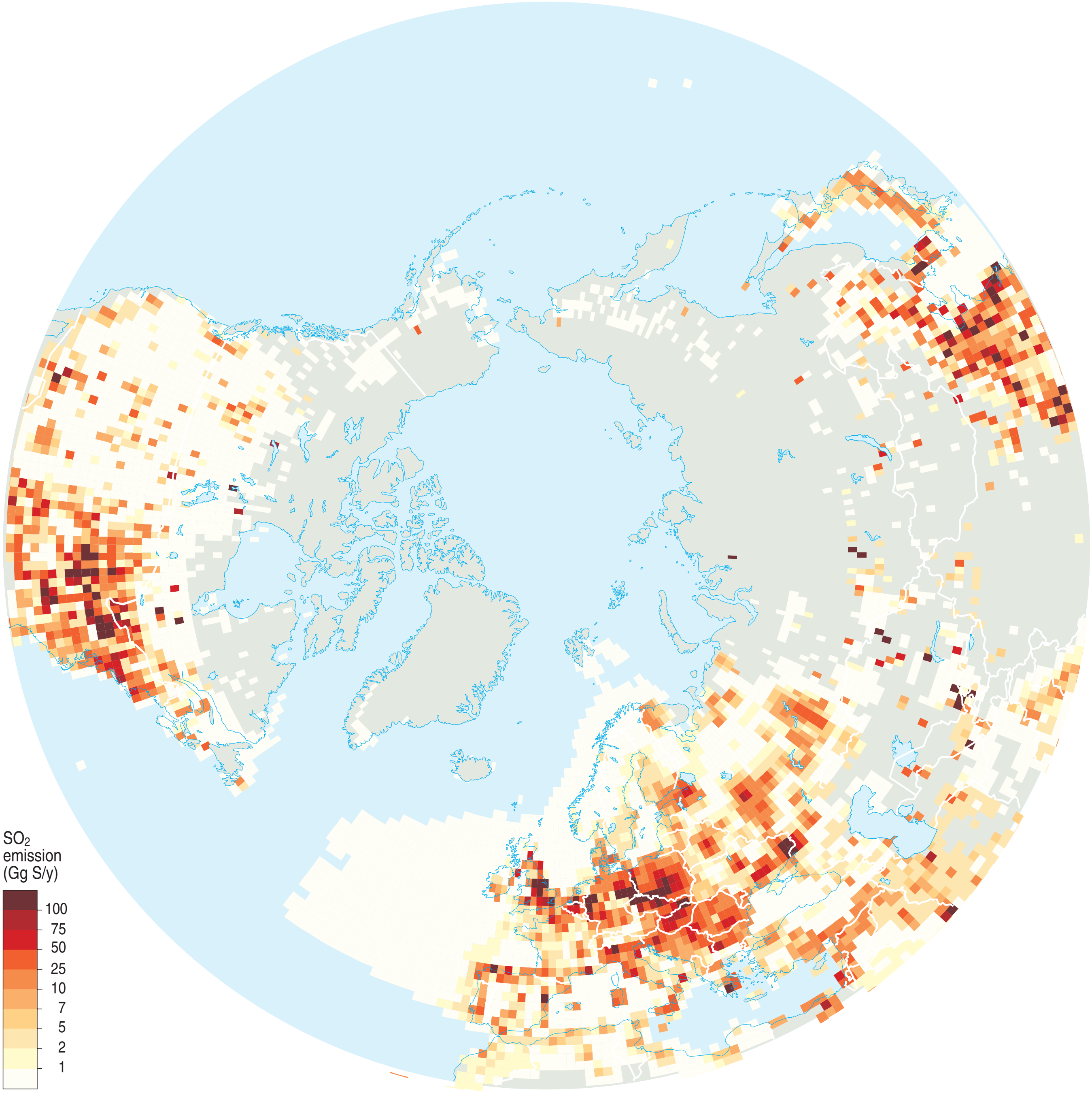

Figure 9·1. Anthropogenic SO_2 emissions for 1985 north of 30°N (Benkovitz *et al.* 1995).

In this chapter, available data on acidifying air pollutants, their emissions, transport, concentrations, deposition, and effects on the Arctic environment, as well as the role of sulfate-bearing particles in the Arctic haze phenomenon, and their occurrence, are assessed. In addition to sulfur and nitrogen oxides, ammonia also has an acidifying effect on the environment. Since the levels of nitrogen compounds, in particular those of ammonia, are at background levels in the Arctic natural environment, the emissions and effects of ammonia are not included in the present chapter, and this assessment concentrates on sulfur.

9.2. Sources of the acidifying compounds

It has been shown that anthropogenic activities account for 70 Tg S (range 62-80 Tg S) of the 98 Tg S (range 80-147 Tg S) global emissions of sulfur each year (Whelpdale and Kaiser 1997). In addition, it has been estimated that 21 Tg N of the 39 Tg N global nitrogen oxide emissions each year are of anthropogenic origin (Whelpdale and Kaiser 1997).

Approximately half of the natural emissions of sulfur are volcanic and most of these are injected into the stratosphere. The rest is mainly from biogenic marine sources. Biomass burning is a significant source of sulfur and part of this source could arguably be classified as anthropogenic rather than natural. It is not surprising that acidification from atmospheric deposition in remote regions is heavily dominated by the long-range transport of these anthropogenic acidic compounds.

9.2.1. Sources outside the Arctic

9.2.1.1. Anthropogenic sources

The distributions of anthropogenic sulfur dioxide and nitrogen oxide emissions over the northern hemisphere are strongly localized to the highly populated and industrialized regions of eastern North America, Europe and south-

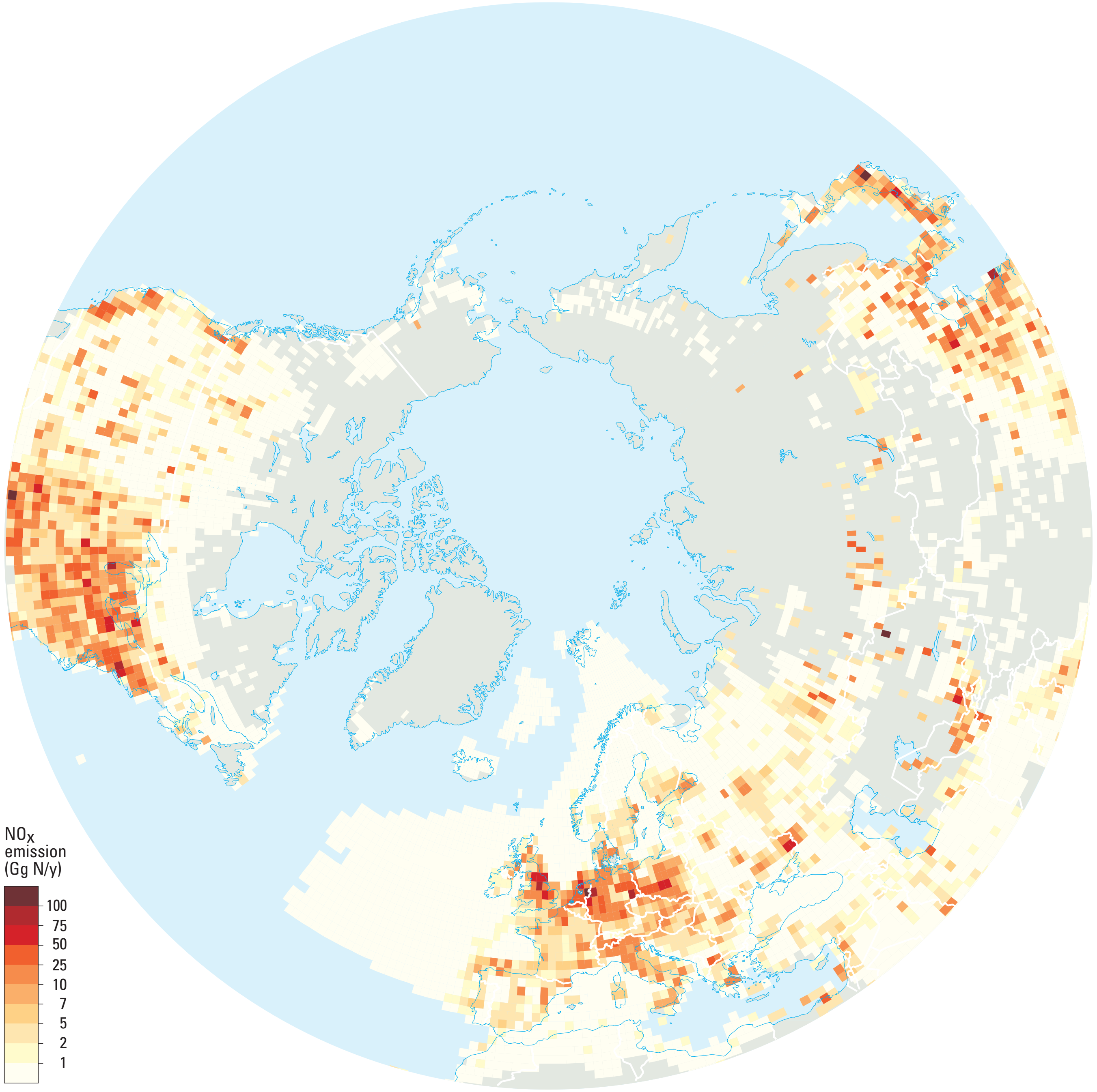

Figure 9·2. Anthropogenic NO_x emissions for 1985 north of 30°N (Benkovitz *et al.* 1995).

eastern Asia (Figures 9·1 and 9·2) as depicted by data of Benkovitz *et al.* (1995). Outside of these high-emission areas, there are smaller regions with elevated emissions, either in connection with densely populated areas or with the exploitation of fuel or mineral reserves. North America contributes about 20% and Europe about 40% of the estimated global emissions of sulfur dioxide from anthropogenic sources.

A large part of the remaining global emissions occur in the far East, particularly in China, where large amounts of coal are used to produce heat and energy. Industrial emissions in central Siberia (eastern Ural, Kuznetsk, Karaganda and the Irkutsk region) are also significant on the hemispheric scale. Figure 9·3 shows the sources of SO_2 north of 50°N with major emission sources on the Kola Peninsula and at Norilsk in Russia.

Detailed information on the relative amounts of sulfur dioxide and nitrogen oxide emissions from different activities is available (USEPA 1995, EEA 1995). The combustion of fossil fuel in electric power plants is the most important source of sulfur dioxide emissions, and mobile sources (e.g., automobiles) are the most important sources of nitrogen oxides (Table 9·1).

Table 9·1. Annual emissions of sulfur and nitrogen in Europe by sector (EEA 1995).

Source	Tg S	%	Tg N	%
Power generation	14.9	54	3.8	21
Commercial and residential combustion	3.0	11	0.8	4
Industrial combustion	7.0	25	2.4	13
Production processes	0.9	3	0.4	2
Fossil fuel extraction	0.05	0	0.08	0
Road transport	0.7	3	7.8	44
Other mobile sources	0.6	2	2.3	13
Waste treatment/disposal	0.09	0	0.2	1
Agriculture	0.001	0	0.05	0
Natural	0.6	2	0.05	0
Total	27.8		17.9	

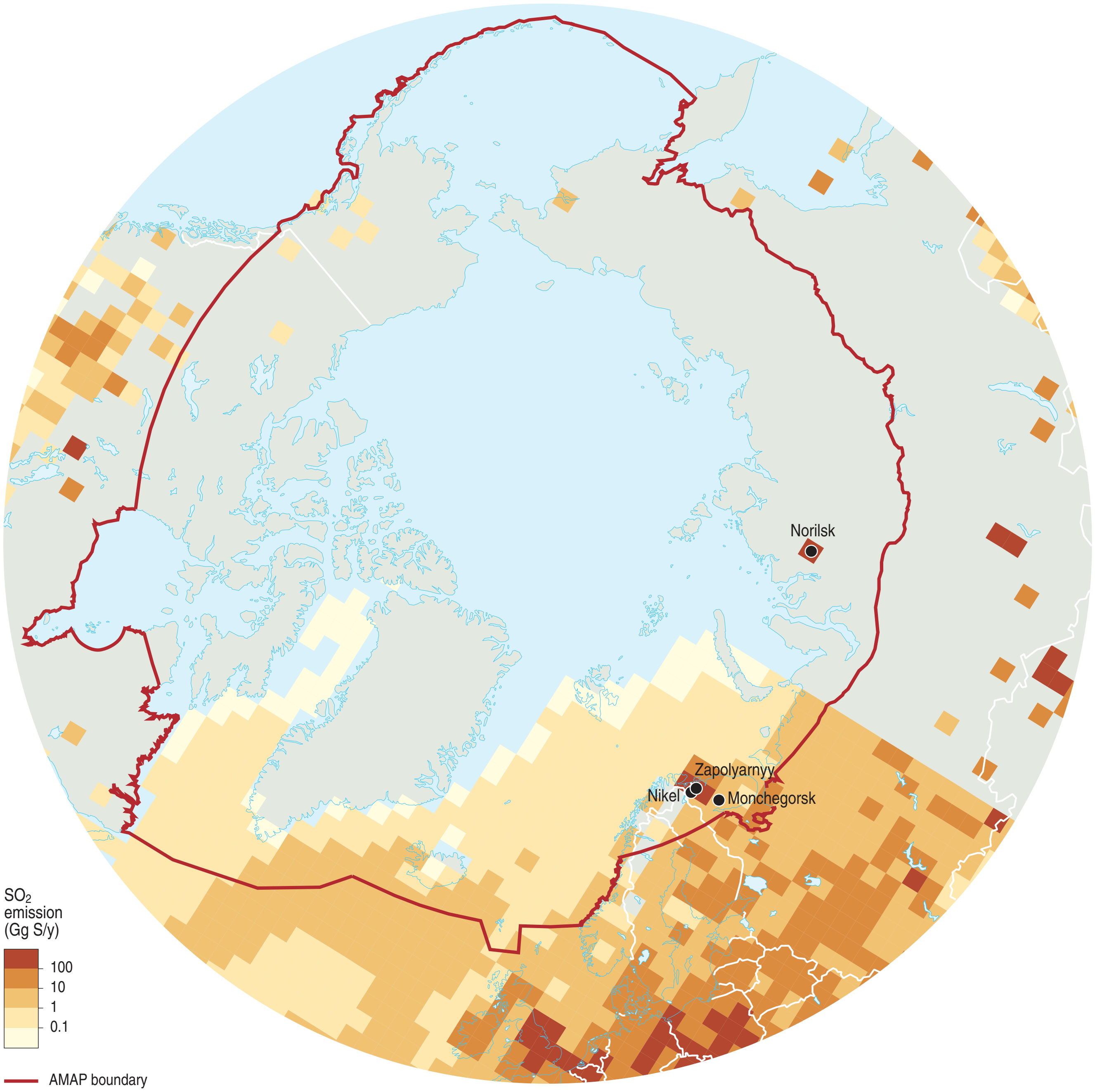

Figure 9·3. SO_2 emissions north of 50°N (modified after Benkovitz *et al.* 1995, see Figure 9·1) showing point source emissions from Arctic and subarctic non-ferrous smelters and including natural DMS sources from the North Atlantic Ocean. Area sources are not included for the Asian part of Russia. This emissions data set for 1988 (total inventory of 50 892 900 tonnes S per year) was used as input for modeling transports of sulphur, results of which are shown in Figures 9·11, 9·12 and 9·20. (Data source: Norwegian Meteorological Institute).

The data given in Figures 9·1 and 9·2 are derived from 1985. Since then, emissions of sulfur dioxide have decreased by about 25% in Europe (UN ECE 1994) (Figure 9·4) and will be reduced further in accordance with the Second Sulfur Protocol. The most important measures to reduce the emissions of sulfur have been flue-gas cleaning in coal-fired power plants, substitution by fuels with a low sulfur content, such as natural gas, energy conservation, and increased sulfur recovery in petroleum refining processes. Flue-gas cleaning has been implemented in several power stations, and is already responsible for a substantial reduction of the sulfur dioxide emissions in Europe (10%) and Japan. Emissions in North America have also decreased somewhat during the same period. However, sulfur emissions in China have not decreased since 1988 and without stringent measures for energy saving and SO_2 control, SO_2 emissions are predicted to increase rapidly within the next 30 years (Qi *et al.* 1995).

Emissions of nitrogen oxides increased rapidly during the period 1950-1975, but remained relatively constant over the period 1980-1990 in Europe and North America (USEPA 1995). One of the control measures introduced to reduce nitrogen oxide emissions has been the installation of three-way catalytic converters on automobiles. However, in Europe, the resulting reduction has been largely offset by the increase in traffic volume. Smelters and other process industries may have relatively small emissions of nitrogen oxides. While the ratio of nitrogen oxide emissions to sulfur dioxide emissions is typically 0.5 in many European countries, it is less than 0.3 in some countries in Eastern Europe.

9.2.1.2. **Natural sources**

The algae in ocean surface waters are a source of sulfur to the atmosphere in the form of dimethylsulfide (DMS), which is oxidized in the atmosphere to sulfur dioxide, sulfate and

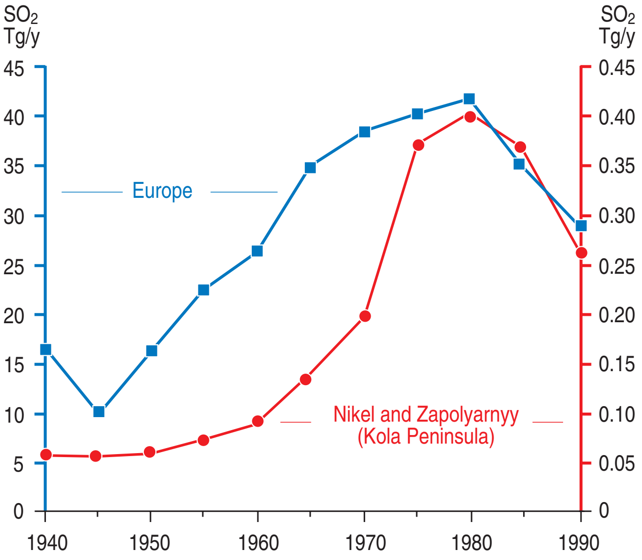

Figure 9·4. Time series of emissions of SO_2 from European sources (Mylona 1993) and two major sources on the Kola Peninsula (Tuovinen 1993).

methyl sulfonic acid (MSA). Emissions of DMS from the world oceans have been estimated to be 15-30 Tg S/y (Andrae 1990). DMS emissions are seasonal, and are determined both by biological productivity and by diffusion and transport through the sea-air interface (Tarrason *et al.* 1995).

Emissions of reduced sulfur compounds from terrestrial environments and vegetation are about one order of magnitude smaller than the marine emissions. Volcanic emissions of sulfur include both hydrogen sulfide (H_2S), elemental sulfur and sulfur dioxide. The emissions are located in areas of volcanic activity and are extremely variable from one year to another. Annual emissions of sulfur from volcanoes between 1964 and 1972 have been estimated at 7.8 Tg S/y (Spiro *et al.* 1992). On the other hand, the 1991 eruption of Mount Pinatubo in the Philippines alone resulted in emissions of 20 Tg sulfur.

Ammonia (NH_3) is also involved in acidification processes; it is a neutralizing compound in the atmosphere, but acts as a net acidifying agent in soils. Ammonia combines with sulfuric acid in the atmosphere to form $(NH_4)_2SO_4$, NH_4HSO_4 and other semi-neutralized sulfates. The total annual emissions of ammonia in Europe within the European Monitoring and Evaluation Programme (EMEP) area are estimated to be 7-8 Tg, and are mainly from animal husbandry and other agricultural activities (Buijsman *et al.* 1987). There are few sources of ammonia in the Arctic, and aerosols in the Arctic are typically highly acidic having a composition largely comprised of NH_4HSO_4 (Barrie and Barrie 1990).

Acidification is not solely a function of sulfate (or nitrate) deposition but is also controlled by the base cations (Ca^{2+}, Mg^{2+}, K^+, Na^+) contained in aerosols or precipitation. There is considerable evidence that recent declines in sulfate levels have occurred together with an accompanying decrease in base cations (Hedin *et al.* 1994). While some of these base cations can be argued to have a natural source (e.g., soil dust), European decreases in base cations have been attributed to an anthropogenic decrease (Hedin *et al.* 1994). For the latter reason, decreases in emissions of sulfur species may not result in an equivalent decrease in acidity.

9.2.2. Sources within the Arctic

9.2.2.1. Metal smelters

The largest emissions of acidifying substances within the Arctic are caused by the production of copper, nickel and other non-ferrous metals from sulfidic ores. Traditional methods involve roasting of the ore to remove part of the sulfur as sulfur dioxide and to oxidize iron before further smelting and refining. In most cases, recovery of sulfur dioxide is technically feasible, but only economically so when the sulfuric acid produced by the recovery process can be utilized in other chemical industries, such as superphosphate fertilizer production or the production of gypsum or other inorganic chemicals.

Table 9·2 gives the main products and estimated sulfur emissions for the largest non-ferrous smelters in the Arctic. One smelter south of the Arctic Circle has been included for comparison and completeness. The right-hand axis of Figure 9·4 shows the time series of the emissions of Nikel and Zapolyarnyy.

Table 9·2. Emissions from non-ferrous smelters in the Arctic in 1992.

Location	Coordinates	Production	Emissions, Gg S/y
Nikel	69°24'N, 30°13'E	Copper, nickel	90
Zapolyarnyy	69°25'N, 30°47'E	Copper, nickel	36
Monchegorsk	67°57'N, 33°00'E	Copper, nickel	91
Norilsk	69°57'N, 88°09'E	Copper, nickel	1100
Sulitjelma*	67°10'N, 15°10'E	Copper	30 (1985)
Rönnskär*	64°45'N, 21°11'E	Lead, zinc	5

* The copper smelter in Sulitjelma closed down in 1985. The smelter at Rönnskär in northern Sweden is south of the Arctic Circle, but has been included because of its proximity to acidified areas. In the early 1970s, emissions from this smelter were close to 65 Gg/y.

9.2.2.2. Exploitation and usage of fossil fuels

Within the Arctic, there is coal mining on Spitsbergen (Norway), in Vorkuta (Russia) and in the Tiksi region (northeastern Siberia). Moreover, there is a large coal mining area in the Pechora Basin, which lies just south of the Arctic Circle in northern Russia. Oil exploitation in the Arctic is described in chapter 10.

Because of the small number of inhabitants in much of the Arctic, fuel and energy consumption is low and the emissions from the usage of fossil fuels are mainly located in towns. For example, there are coal-fired power plants in Vorkuta and Inta (Russia), which serve the local settlements, and coal mining and oil and gas exploration in these areas. Other examples include the mining settlements on Spitsbergen which are also served by small, coal-fired electric power plants in Longyearbyen and Pyramiden.

Shipping and fishing activities are also sources of air pollutants in the Arctic. For example, extensive deep-sea fishing for cod, capelin and prawns takes place in the Barents Sea. Marine transport, particularly of timber and timber products, is also important along the Siberian coast and on the Siberian rivers.

9.2.2.3. Natural emissions

There are areas of volcanic activity in the North Atlantic and Bering Sea regions, and in southern Alaska, however, the associated sulfur emissions are relatively low and sporadic. The volcano that erupted on Jan Mayen Island in 1978, is not presently active. A recent eruption, in late September, 1996, occurred under the Vatnajökull glacier in central Iceland. Most of the volcanoes in the Aleutian range are located to the south of the Arctic Circle, as are the active volcanoes with more continuous sulfur emissions on the Kamchatka Peninsula and Iceland. A notable area with respect to natural emissions of sulfur is the Smoking Hills area in Canada, where the 'natural' combustion of pyrite-bearing bituminous shale, releasing sulfur dioxide and sulfuric acid mist and aerosol, has caused phytological damage within 500 m of the source (Freedman *et al.* 1990). However, these emissions are relatively low compared to anthropogenic inputs (Barrie 1986).

The Arctic Ocean and adjacent seas are generally quite productive and hence the biogenic production of dimethylsulfide (DMS) and transport of DMS through the sea-air interface must be considered a potential source of atmospheric sulfur. These emissions occur mainly between June and August and emission rates may approach 50 mg S/m^2/y in the North Atlantic/Barents Sea region (Tarrason *et al.* 1995). On the basis of S-isotope analyses, Li and Barrie (1993) conclude that, in winter, anthropogenic sources account for almost all of the sulfur in the Arctic atmosphere, whereas in summer about 30% of the sulfur is from natural sources. Li *et al.* (1993) report 13 years of airborne MSA observations at Alert peaking in spring (April-May) and in summer (July-August). Maenhaut (1993) has shown that biogenic sulfur may account for more than 50% of the total airborne sulfur in airborne particles at Spitsbergen in July-August, but that the biogenic contribution to the annual mean concentration level is only about 2%.

9.3. Atmospheric processes

The chemical cycle of acidic compounds in the atmosphere is comprised of sources, chemical transformations in the atmosphere, atmospheric transport, and wet and dry removal processes. The pathways by which gases and aerosols move through the atmosphere and enter the Arctic are discussed in chapter 3 of this assessment. In principle, atmospheric transport routes for acidic compounds should be similar to the pathways of other contaminants, such as non-volatile POPs and heavy metals. In this section, the chemistry of sulfur and nitrogen-bearing compounds, the air concentration data recorded in the Arctic, and, finally, Arctic haze as a visible phenomenon are discussed

9.3.1. Arctic air chemistry

9.3.1.1. The sulfur cycle

The atmospheric chemistry of the sulfur cycle is dominated by OH radical reactions in the gas phase with H_2S, DMS, and SO_2, all of which lead to the production of gaseous sulfuric acid (H_2SO_4), and by gaseous and aqueous phase reactions between SO_2 and hydrogen peroxide (H_2O_2) and ozone (O_3). Once sulfate is produced, its removal is relatively rapid with an atmospheric half-life in the order of 3 to 7 days at mid-latitudes and about two weeks or more in the High Arctic during winter (Barrie 1986). The atmospheric emission, production, transport and deposition cycle of sulfate aerosol (whether sulfuric acid or ammoniated sulfate compounds such as $(NH_4)_2SO_4$ and NH_4HSO_4) has been the subject of intense research activity during the last 20 years (Barrie and Barrie 1990, NAPAP 1990, Levy *et al.* 1990, Whelpdale 1992).

There are transport and chemical processes in the sulfur cycle that are strongly latitude-dependent. The lack of sunlight in the Arctic for large parts of the year limits the production of the OH radical and H_2O_2. The former is produced from the photodissociation of ozone in clean air and, incrementally, from hydrocarbon radicals in more polluted areas (Ehhalt *et al.* 1991). Lower OH and H_2O_2 concentrations in winter slow the sulfate production cycle and increase the SO_2/SO_4^{2-} ratio observed in the Arctic (Barrie and Hoff 1984, Möhler and Arnold 1992). This photochemical mechanism is critical to the timing of the Arctic haze maximum (see section 9.5). The seasonality of SO_2 oxidation to sulfate is important in prolonging the presence of sulfate aerosols in the Arctic into April and May (Barrie and Hoff 1984, Barrie 1995).

9.3.1.2. Nitrogen chemistry

As air masses move from mid-latitudes to the Arctic, the set of chemical species available to drive nitrogen chemistry changes. The nitrogen chemical cycle (Seinfeld 1986) has a considerable 'dark' component, with night-time reactions between NO_2 and O_3 to form NO_3 and N_2O_5. The reaction of N_2O_5 with water produces gas phase nitric acid (HNO_3) and particulate nitrate. Lack of sunlight slows the photochemical destruction of HONO back to NO. HONO has been observed in the Arctic troposphere (Adrian *et al.* 1992, Li 1994, Hausmann and Platt 1994). The RO_3 (peroxyacyl radical) reaction with NO_2 in the formation of peroxyacetyl nitrate (PAN) (and other C_3-C_7 alkyl nitrates) is especially important in the Arctic, since the alkyl nitrate chemical removal mechanism is also dependent on light and temperature. PAN is an atmospheric reservoir for nitrogen in the Arctic winter, and these alkyl nitrates may contain as much as 75-80% of the airborne NO_y (Bottenheim *et al.* 1993, Singh *et al.* 1992).

Over large parts of the mid-latitudes, ammonia emissions are generally sufficient to neutralize, at least partially, the sulfuric acid formed as a result of the oxidation of sulfur dioxide. Neutralization is greatest in summer near the ground and least in winter aloft in the atmosphere. Ammonia can also react with nitric acid to form ammonium nitrate. Ammonium nitrate has a much lower rate of deposition than gaseous nitric acid, so the result is an enhanced transport of both ammonium and nitrate out of the emission areas (Hov and Hjøllo 1994).

9.3.2. Arctic geophysical properties affecting acidic compounds

Insolation at high latitudes is extremely variable, ranging from complete darkness in winter to complete daylight in summer. Long periods of Arctic night shut off photochemical activity responsible for the transformation of SO_2 to H_2SO_4 and NO_x to HNO_3. Thus, the relative importance of photochemical and 'dark' reactions in the Arctic atmosphere can be expected to be quite different to that in mid-latitudes. As an indication of the intensity of solar radiation in the High Arctic summer, mid-day intensities at 70°N are comparable to those at 7 a.m. at latitude 45°N.

The low Arctic temperatures modify the chemistry of heterogeneous reactions between gases and aerosols. Reaction and sorption mechanisms peculiar to low temperatures have been suggested for SO_2 on snow and ice (Conklin *et al.* 1993). Ice and sulfuric acid catalyzed halogen destruction of O_3 at polar sunrise in the Arctic boundary layer has been demonstrated by Barrie *et al.* (1988, 1994). Low temperatures are largely responsible for the appreciable PAN reservoir in the Arctic.

Pollutants transported from mid-latitudes will tend to follow surfaces of constant potential temperature (isentropic surfaces). A rapidly moving air mass will rise as it moves into the Arctic, forming pollutant layers at higher altitudes (Carlson 1981, Iversen 1984, Raatz *et al.* 1985, Schnell *et al.* 1989, Radke *et al.* 1989). The vertical dispersion of surface-produced pollutants within the Arctic, however, is limited by strong inversions in the Arctic boundary layer (frequently greater than 20°C in the first few hundred meters above ground). Despite the fact that high altitude haze-layers are observed, there is considerable evidence to show that the main mass of the Arctic aerosol and gaseous pollutants are confined predominantly to altitudes below 2 km in the

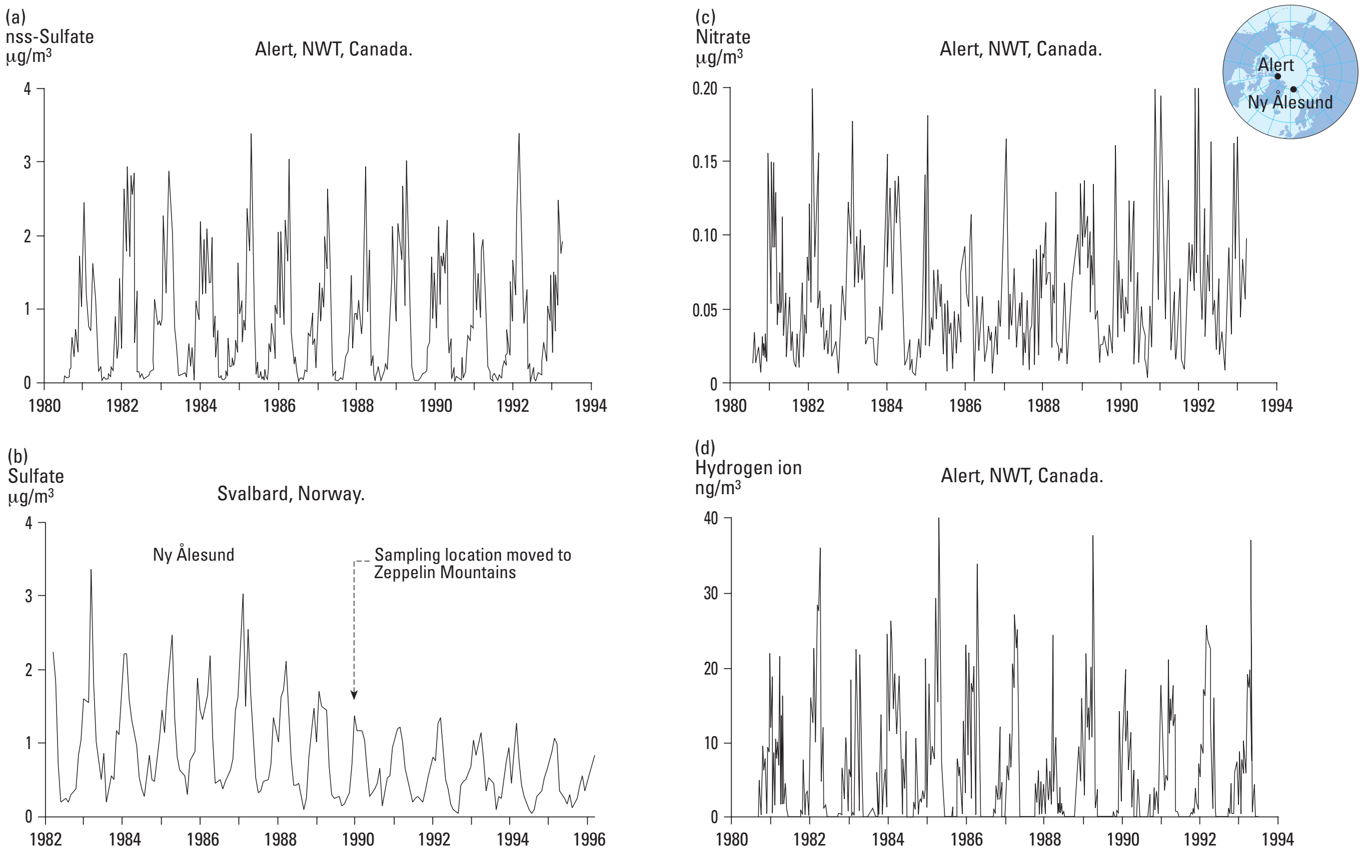

Figure 9·5. The time series of a) weekly-averaged non-seasalt (nss) sulfate aerosol at Alert, NWT, Canada (Barrie pers. comm.), b) monthly-averaged sulfate aerosol at Ny-Ålesund, Norway (data source: NILU), c) weekly-averaged nitrate at Alert (Barrie pers. comm.), and d) weekly-averaged hydrogen ion at Alert (Barrie pers. comm.).

atmosphere (Shaw 1975, Hansen and Rosen 1984, Schnell and Raatz 1984, Leaitch *et al.* 1984, 1989, Hoff 1988, Pacyna and Ottar 1988). The atmospheric injection of anthropogenic and other contaminants to the Arctic has pronounced seasonal and latitudinal variations (see chapter 3, Figures 3·4 and 3·6). Iversen (1989a) showed that these variations can explain much of the variance in sulfate concentrations measured at Spitsbergen. Thus, it appears that, in addition to seasonal variations in physical and chemical properties of Arctic air, transport determines much of the observed seasonality of Arctic haze.

A predominant factor in the special behavior of acidic pollutants in the Arctic is the lack of precipitation. In fact, some parts of the Arctic meet the definition of a desert, with an annual precipitation of less than 100 mm at many Arctic sites (see chapter 3). Much of the precipitation which does occur takes the form of drizzle from summer stratus clouds during June to August. At this time, the washout of pollutants is at its most efficient in the Arctic, the meridional exchange is at its minimum, and the extension of the Arctic air mass does not include the large Eurasian emission sources (see chapter 3, Figures 3·4 and 3·5).

While the amount of precipitation is limited in the Arctic, the Arctic atmosphere contains enough water vapor to influence gas phase reactions. Relative humidity over ice is nearly always supersaturated during the winter months. Relative humidity with respect to water is often as high as 90%. This leads to rapid nucleation of ice crystals from ice nuclei (Borys 1989, Jaffrezo and Davidson 1993). Speculation about the role of this mechanism as a removal pathway for sulfates arose in the early-1980s, but it was concluded that sulfate aerosols are less efficient ice nuclei than other remote aerosols (Borys 1989). The contention is that this process tends to keep aerosols aloft, rather than forming ice crystals which would precipitate out. More recent work on Greenland, however, concluded that although the riming of ice crystals and snow may only contribute 5% of the water mass deposited on the Arctic snowpack, over 30% of the chemical deposition may be derived from these events (Jaffrezo and Davidson 1993). Morphological studies of single aerosol particles support this view, since aerosol droplets containing liquid and solid inclusions were observed as well as sulfuric acid droplets (Parungo *et al.* 1993).

The Earth's surface in the North differs from that at lower latitudes. Both the land and water surfaces are covered with ice or snow for a large part of the year. Deposition on these surfaces is very inefficient and this leads to long residence times for particles and gases in the Arctic atmosphere. The lack of vegetation in the High Arctic leads to low deposition on the surface, because vegetation generally has a higher uptake of pollutants than a smooth surface. The deposition velocity will be strongly related to the surface characteristics of the Arctic. These transport and deposition processes are described in more detail in chapter 3.

9.4. Concentrations of acidifying compounds

9.4.1. Measurements of atmospheric concentrations

Our knowledge of Arctic air chemistry comes from extensive field campaigns (some are discussed in sections 9.3 and 9.4) as well as from routine observations at a few sites over longer time periods, starting in the late-1970s.

The time series of weekly-averaged sulfate aerosol concentrations measured at Alert, NWT, Canada since 1980 is shown in Figure 9·5a (Barrie pers. comm.) and the time series measured at Ny-Ålesund, Svalbard since 1982 in Figure

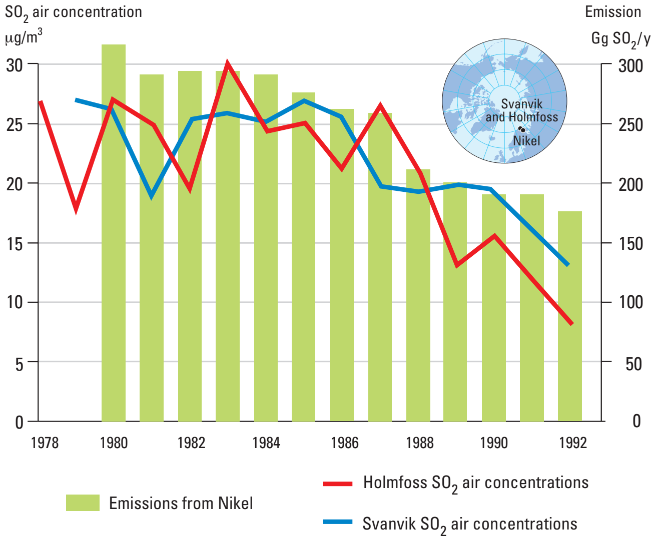

Figure 9·6. Emission rates of sulfur dioxide from Nikel together with annual average SO_2 air concentrations measured at two sites in Norway (1978-1992) (Sivertsen 1996).

9·5b. The peak sulfate concentration at Ny-Ålesund was 3-4 µg/m^3 in the 1980s, but is clearly lower in the 1990s, showing a decreasing trend since 1987. The peak concentrations occur in the months of March and April. The concentration time series for Alert does not show any statistically significant trends (Barrie pers. comm.). It is difficult to determine a trend in a dynamically variable time series without measurements over many years (Blanchard *et al.* 1995).

The yearly average concentrations of sulfur dioxide show a decreasing trend after 1987 at three measuring stations in northern Norway: Jergul (SFT 1994), Svanvik (Sivertsen *et al.* 1994) and Holmfoss (Sivertsen 1996). The data from the latter two stations clearly reflect the reduction in SO_2 emissions from 310 kt in 1980 to about 180 kt in 1992 from the nearby Nikel smelter (Figure 9·6) (Sivertsen 1996).

Figure 9·5c shows the atmospheric concentration time series for nitrate at Alert, NWT. The nitrate concentration is lower by a factor of about twenty by mass (by a factor of about ten on a molar basis) compared to sulfate. This is consistent with observations in Scandinavia. In a latitude-dependent survey of nitrate and sulfate in precipitation in northern Europe and the Arctic, Joranger and Semb (1989) showed that the nitrate/sulfate molar ratio decreased monotonically from south to north (Figure 9·7). In northern Scandinavia (Joffre *et al.* 1990, Joranger and Semb 1989, Tuovinen *et al.* 1993), the nitrate/sulfate molar ratio ranges from about 0.4 to 1 (mass ratio of 0.2-0.5).

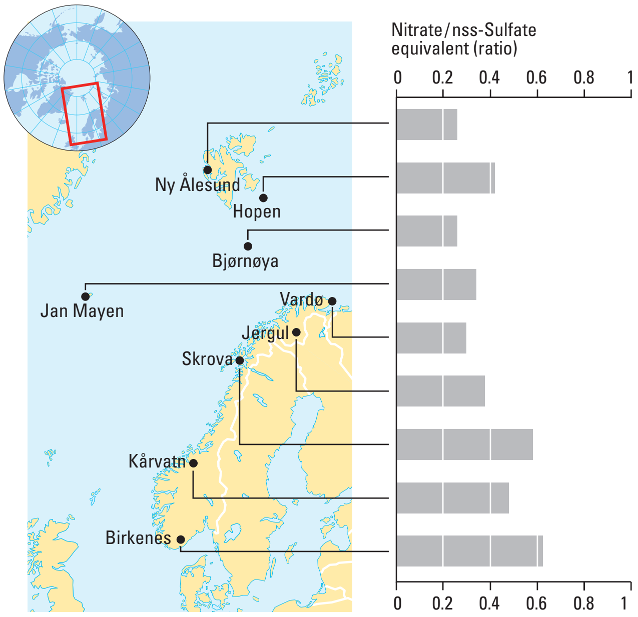

Figure 9·7. Variation of the nitrate to non-seasalt (nss) sulfate equivalent ratio in precipitation at Norwegian recording stations based on measurements between August 1982 and July 1984 (Joranger and Semb 1989).

Figure 9·5d shows the time series for the concentration of hydrogen ions on particulate filters at Alert, NWT (Barrie pers. comm.). The time series is very similar to that for sulfate, with the maximum in the spring of each year. In fact, the molar percentage ratio of ammonium plus hydrogen ions to sulfate is almost constant throughout the year, varying from 0.40 to 1.75 with an average near 1. This strongly suggests that the aerosol is, on average, predominantly NH_4HSO_4, which is consistent with observations of Barrie and Barrie (1990).

The representativeness of measurements made at only one site in the Arctic obviously needs close examination. Barrie (1986) presented the relative sulfate concentrations throughout the Arctic for the spring months of 1980 and 1981. The conclusion drawn from this work is that the mean springtime sulfate concentrations in the Norwegian Arctic are 1.5-3.9 µg/m^3 and in the North American Arctic 1.2-2.2 µg/m^3. The results suggest an intrusion of sulfate from Europe and the former Soviet Union into the High Arctic.

Areas near Norilsk in Siberia, the Kola Peninsula and northern Scandinavia are likely to suffer from high levels of ambient air concentrations. Not only do these regions bear the brunt of the European and Asian emissions which find their way to the Arctic, but they are also subjected to local and regional emissions of considerable magnitude. For example, at the air quality monitoring station at Sevettijärvi, northeastern Finland, the highest measured hourly average SO_2 concentrations approached 400 µg/m^3 in 1990 (Virkkula *et al.* 1992), and at Viksjøfjell, northern Norway, close to the Cu-Ni smelters on the Kola Peninsula, they have exceeded 1000 µg/m^3 (Sivertsen *et al.* 1994).

In terms of chronic exposure to sulfur dioxide, the monthly-averaged SO_2 concentrations at Sevettijärvi reached 20 µg/m^3 in November 1992 (Tikkanen and Niemelä 1995). Closer to the smelters on the Kola Peninsula, Norwegian measurements have shown air concentrations at Kirkenes, Holmfoss, and Svanvik to exceed the 15 µg/m^3 half-year forest-damage threshold for the winter season (see section 9.10.2.1; Sivertsen *et al.* 1992). Figure 9·8a shows measured annual average sulfur dioxide concentrations and Figure 9·8b average annual sulfate aerosol concentrations across the Arctic. Andruhov and Pudovkina (1993) and Tsaturov (pers. comm.) report yearly air SO_2 concentrations in industrial regions near Nikel of 30-60 µg/m^3, Zapolyarnyy about 70 µg/m^3, Monchegorsk about 20 µg/m^3 and Norilsk 140-160 µg/m^3. The concentrations in Greenland, the Canadian Arctic, and Svalbard are much lower compared to these polluted regions.

9.4.2. Precipitation chemistry

There are many problems in collecting reliable and representative precipitation samples in the Arctic (see chapter 3, Figure 3·10). Precipitation collectors and gauges have low capture efficiencies for snow, particularly at wind speeds above 4-5 m/s. Low precipitation intensities and drifting snow make the collection of reliable samples from the surface snow layer difficult. Because of this, and also because of the relative inaccessibility of many Arctic areas, precipitation chemistry data are largely missing from the areas of the High Arctic, where the concentrations of acidifying compounds are believed to be low.

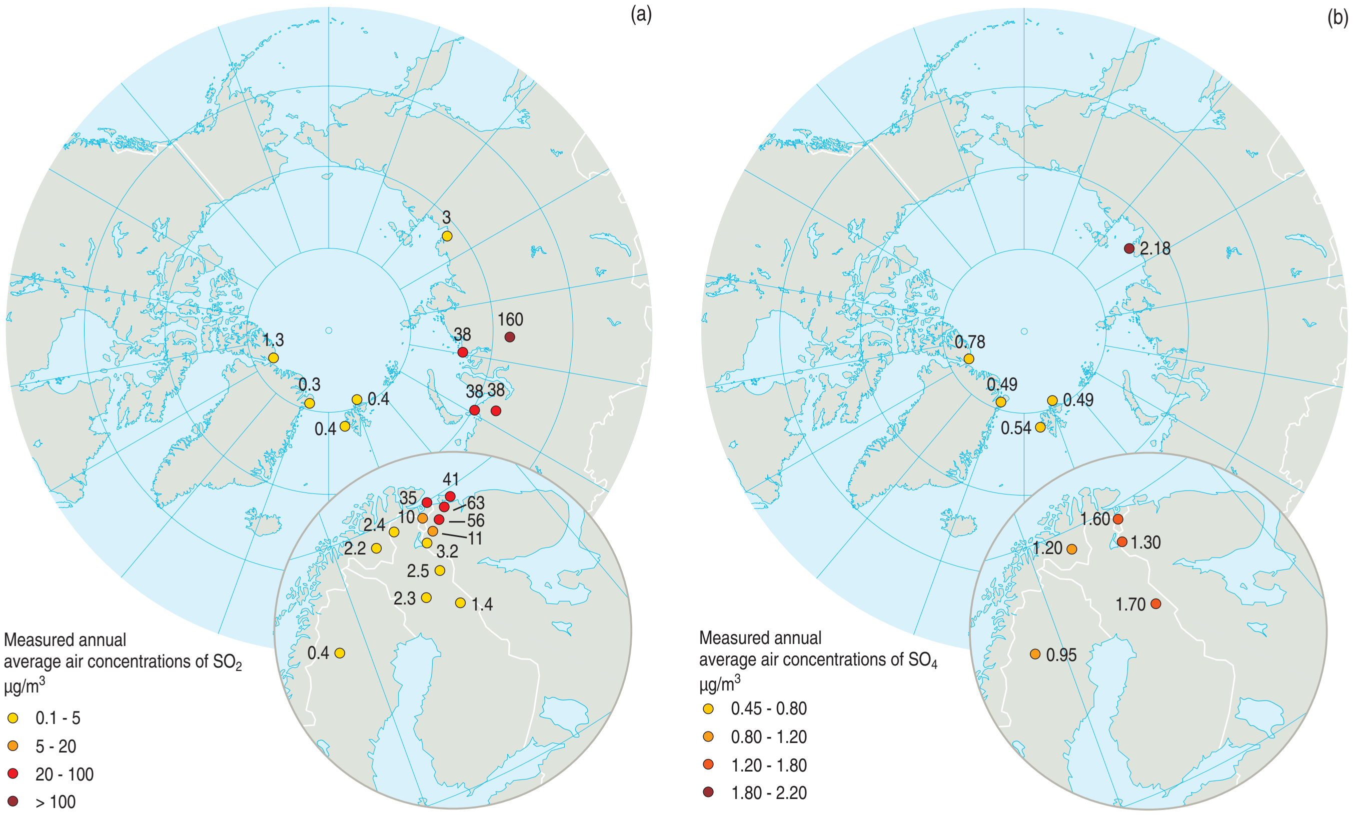

Figure 9·8. a) Yearly average sulfur dioxide air concentration measurements in the Arctic (Tsaturov pers. comm., Sivertsen 1996, Iversen *et al.* 1991, Sandnes and Styve 1992, Sandnes 1993, Tuovinen *et al.* 1994, Larsen 1994, Tikkanen and Niemelä 1995, SFT 1994, Heidam pers. comm., Tørseth 1996, Barrie pers. comm.). b) Yearly average sulfate air concentration measurements in the Arctic (Iversen *et al.* 1991, Sandnes and Styve 1992, Sandnes 1993, Tuovinen *et al.* 1994, Heidam pers. comm., Tørseth 1996, Barrie pers. comm.).

Figure 9·9, displaying some representative snow chemistry data in the Arctic, shows that concentrations of sulfate and nitrate in northern Canada, Alaska, and Greenland are mostly low. High concentrations of sulfur occur in the Barents region and in Siberia. The snow chemistry in northern Russia and northern Siberia show a pronounced influence of the Norilsk smelter (Tsaturov pers. comm.). The very low concentrations at the Greenland ice cap may be partly due to low scavenging efficiencies under extremely cold conditions, as discussed by Davidson *et al.* (1987, 1989).

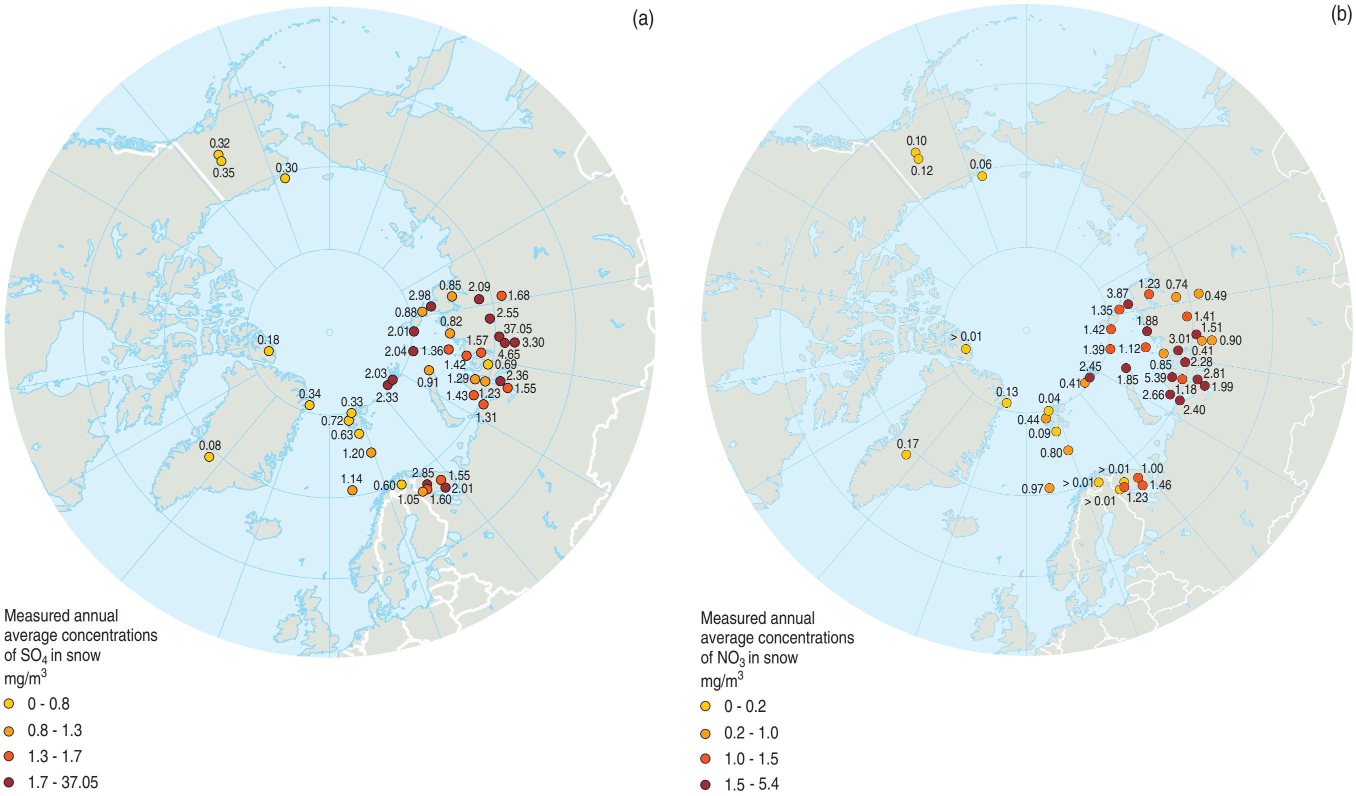

Figure 9·9. Concentrations of a) sulfate and b) nitrate in snow (Tsaturov pers. comm., Dayan *et al.* 1985, Barrie *et al.* 1985, Shaw *et al.* 1993, Galloway *et al.* 1982, National Environment Research Institute pers. comm., Davidson *et al.* 1987, Semb *et al.* 1984, Joranger and Semb 1989, Tikkanen and Niemelä 1995, Jaffe *et al.* 1995).

The snow samples collected on Spitsbergen (Semb *et al.* 1984) indicate a clear southeast-northwest gradient, suggesting that the main transport direction during the deposition of sulfate and nitrate was from the southeast. The concentrations of both sulfate and nitrate were generally much less than the mean values obtained from regular precipitation samples by Joranger and Semb (1989) from the same area.

The nitrate concentration in snow on Greenland has increased markedly during the last 100 years, but the highest concentrations occur later in the year than is the case for sulfate. The reason for this is not fully understood. At Spitsbergen, and in the Arctic generally, the contribution of nitrate to precipitation acidity is generally much lower than at lower latitudes.

Ryaboshapko *et al.* (1994) present some data on the concentrations of sulfate and the deposition of sulfate by precipitation from an extensive network of stations in Russia. While most of these sites are south of the Arctic Circle, concentrations of 0.8-1.0 mg SO_4-S/L may be found in both northern Russia and northwestern Siberia.

More detailed information on precipitation chemistry is available for northern Fennoscandia and the Kola Peninsula, from a number of precipitation sampling stations (Derome *et al.* 1992a, Lövblad *et al.* 1992, Sivertsen *et al.* 1993) and snowpack sampling stations (Derome *et al.* 1992b, 1993, Baklanov *et al.* 1994, Makarova *et al.* 1994, Buznikov *et al.* 1995, Jaffe *et al.* 1995). In general, these measurements show only a moderate influence of the large sulfur dioxide emissions on the Kola Peninsula on the precipitation and snow chemical composition, except within 20-50 kilometers from the sources (see Tikkanen and Niemelä 1995).

9.4.3. **Glacial measurements**

Another important source of information for assessing the temporal development of Arctic air pollution is measurements on glacial ice. Records from glacial ice cores provide reconstructions of trends in the contamination of Arctic air by acidic and other pollutants. Accumulated snow exhibits changes in chemistry with depth, reflecting the chemistry of the atmosphere at the time of deposition (Barrie *et al.* 1985). Measurements of snow and ice composition have been made on Greenland (Busenberg and Langway 1979, Hammer *et al.* 1980, Herron 1982, Davidson *et al.* 1981, 1985), on northern Ellesmere Island, Canada (Koerner and Fisher 1982, Barrie *et al.* 1985) and on Mount Logan in northwestern Canada (Holdsworth and Peake 1984).

The historical record from Greenland ice at the Dye 3 site indicates that there has been an increase in the concentrations of the strong acid anions SO_4^{2-} and NO_3^- in this century (Herron 1982) (Figure 9·10). Measurements in southern Greenland (Neftel *et al.* 1985) confirm Herron's observations depicting a significant increase since the late-1930s (Figure 9·10). Ice core acidity at another location in Greenland displayed no trend between 1850 and 1950, but showed a gradual slow increase since 1950 (Hammer *et al.* 1980).

A historical record for conductivity (which is well correlated with acidity), inferred from the ice core at Agassiz ice cap on northern Ellesmere Island, indicates little variation in the magnitude of air pollution in the first half of this century, but a marked increase of about 75% between 1956 and 1977. This increase reflects the increase in European SO_2 and NO_x emissions (Barrie *et al.* 1985). Current levels of winter acidity are 10-12 µeq/L compared to 7-9 µeq/L prior to 1956.

Measurements of recently accumulated ice layers indicate that the maximum concentrations of sulfates and fine parti-

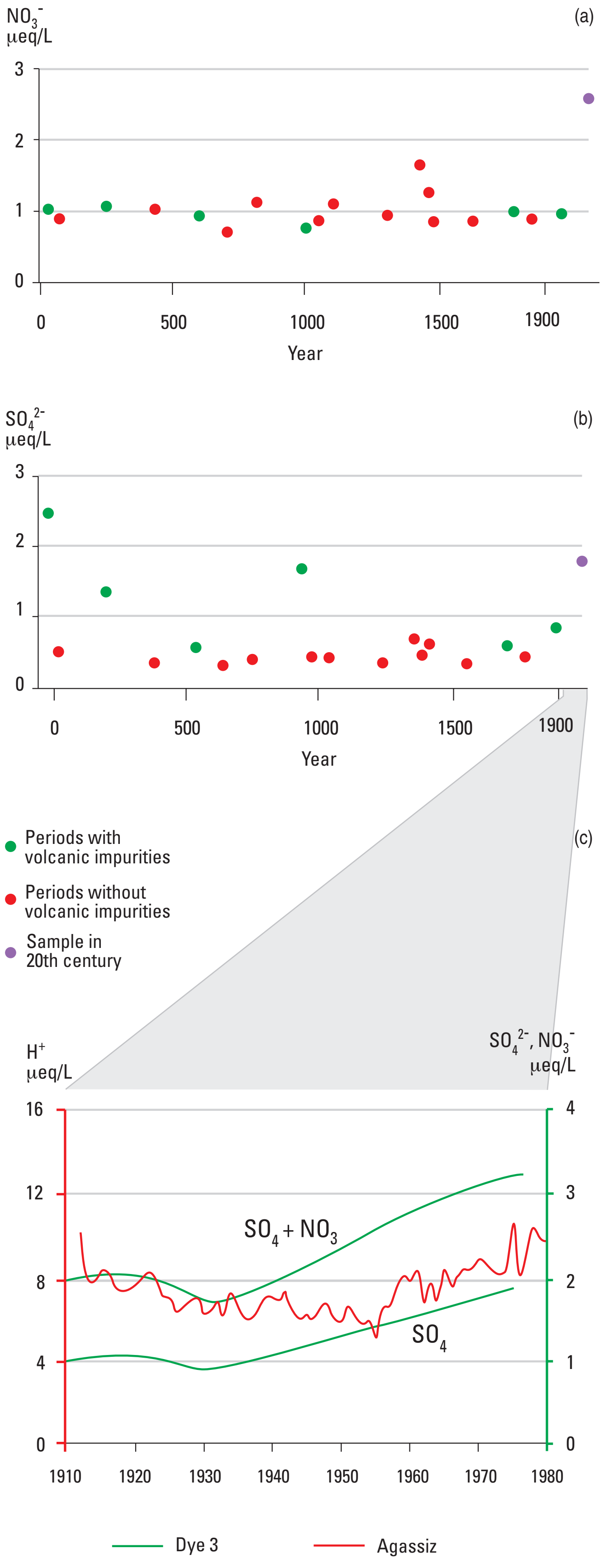

Figure 9·10. Historical records of a) NO_3^- and b) SO_4^{2-} concentrations from the Greenland icecap at Dye 3 (data source: Herron 1982; modified from Barrie 1986). Each point is a multi-year average over a period of 2-37 years. The same sample in this century is emphasized with a cross. c) A comparison of trends in the average SO_4^{2-} and SO_4^{2-} plus NO_3^- concentrations (data source: Neftel *et al.* 1985) at Dye 3 in Greenland and in the annual mean H^+ concentration from Agassiz icecap, Canada (inferred from conductivity by Barrie *et al.* 1985). The Neftel curves represent cubic spline fits reported by the authors (modified from Barrie *et al.* 1985).

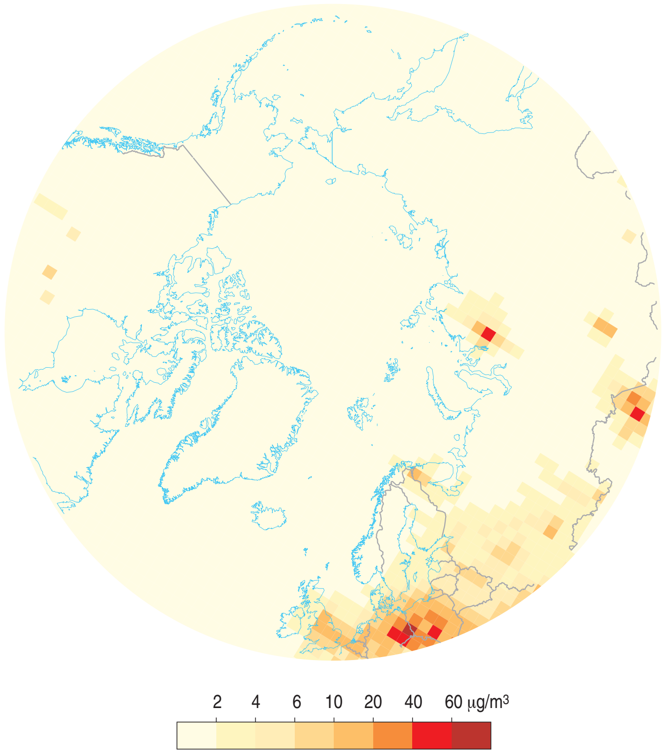

Figure 9·11. The predicted (1988) average surface air sulfur dioxide concentrations in the Arctic (Iversen and Tarrason 1995, Tarrason and Iversen 1996, Tarrason 1995).

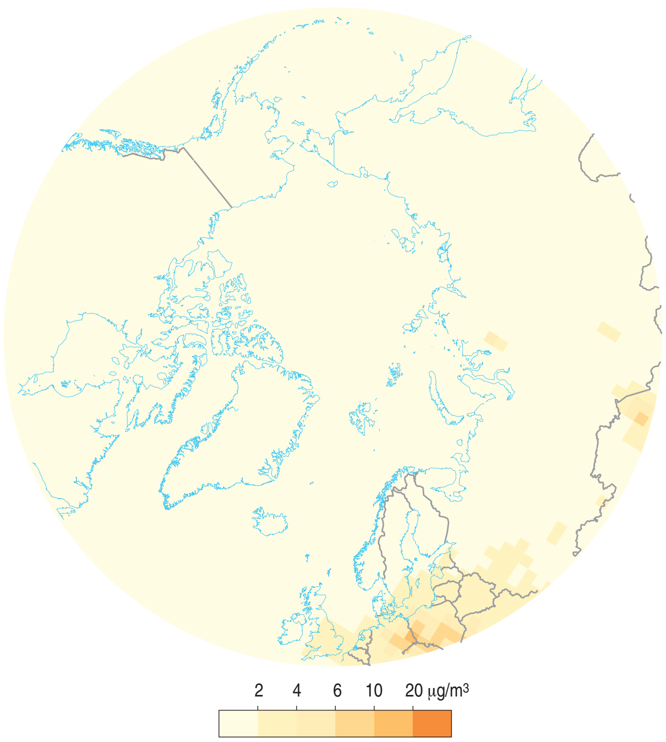

Figure 9·12. The predicted (1988) average surface air sulfate concentrations in the Arctic (Iversen and Tarrason 1995, Tarrason and Iversen 1996, Tarrason 1995).

cles were observed in the beginning of the 1980s. No major changes in concentrations have been observed during the 1980s. A decrease occurred at some stations at the end of the decade. These trends can be partially related to the documented reductions in sulfur emissions in Europe, and partially to the replacement of coal by natural gas in the production of heat and electricity, particularly in Russia.

9.4.4. **Modeling results**

The ability to predict the impacts on the Arctic of known sulfur sources at mid-latitudes is a good test of the state of our knowledge about the processes involved. The sophistication of models used to make predictions have varied from the simpler two-dimensional box models to three-dimensional Eulerian models. Predictions of Arctic air pollution have benefited from the development of mid-latitudinal regional acid deposition models (see chapter 3 for general description).

The state-of-the-art for atmospheric modeling of pollution in the Arctic is highly developed compared to models for other compartments, largely because of the development over the years of 1) weather forecast models, which provide considerable information about the physical processes in the atmosphere and reliable meteorological data for the atmospheric transport models, 2) comprehensive regional acid deposition models for the mid-latitudes, which have detailed descriptions of both the physical and chemical processes in the atmosphere, and finally 3) atmospheric models specifically for the Arctic.

The state-of-the-art for atmospheric long-range transport modeling for the Arctic is comprised of three-dimensional Eulerian models, which have detailed descriptions of the physical processes, while the chemical processes are simplified (first order sulfur chemistry), in order to be able to perform long-term calculations. Examples of such models include the three-dimensional hemispheric model developed by Iversen (1989b) and Tarrason and Iversen (1992, 1996), the three-dimensional global sulfur model by Dastoor and Pudykiewicz (1996), and the three-dimensional hemispheric model developed by Christensen (1995, 1997).

The model by Iversen (1989b) was used for calculating the transport of sulfur to the Arctic for March and July, reproducing the seasonal differences observed for Arctic haze. More recently, Tarrason and Iversen (1992, 1996) have applied a further developed version of the model for a full year. Figures 9·11 and 9·12 show the spatial distribution of the predicted ground level sulfur dioxide and sulfate air concentrations (Tarrason and Iversen 1996, Iversen and Tarrason 1995, Tarrason 1995). The predicted air concentrations clearly reflect the SO_2 sources on the Kola Peninsula, Norilsk, and the industrial parts of Europe.

An example of the application of the model by Christensen (1995, 1997) is given in Figure 9·13, where the calculated concentrations of SO_2 and SO_4^{2-} have been compared with measurements at Station Nord in northeastern Greenland (81°36'N, 16°40'W). The figure shows very good agreement between the calculated and observed weekly mean concentrations of sulfur dioxide and, even more so, of sulfate. Scatter plots depicted in Figure 9·14, demonstrate the general tendency of the model to reproduce the observed SO_2 concentrations with reasonable accuracy for European monitoring stations.

The different models have been used to estimate the contribution from different sources to Arctic sulfur pollution. An example of this is shown in Figure 9·15, where the vertical distribution of SO_x (SO_2+SO_4^{2-}) concentrations, averaged over the area north of 75°N, and the contributions from the different sources are shown. Figure 9·15a shows that the surface air concentration is about 0.56 ppbv (1 ppbv = 1.3 µg S/m^3), with a maximum concentration of about 0.60 ppbv at 600 m. Above 600 m, the concentration decreases, dropping to 0.06 ppbv at 7000 m. Figure 9·15b shows that sources at Norilsk contribute about 35% and European sources about 17% to the surface air concentrations.

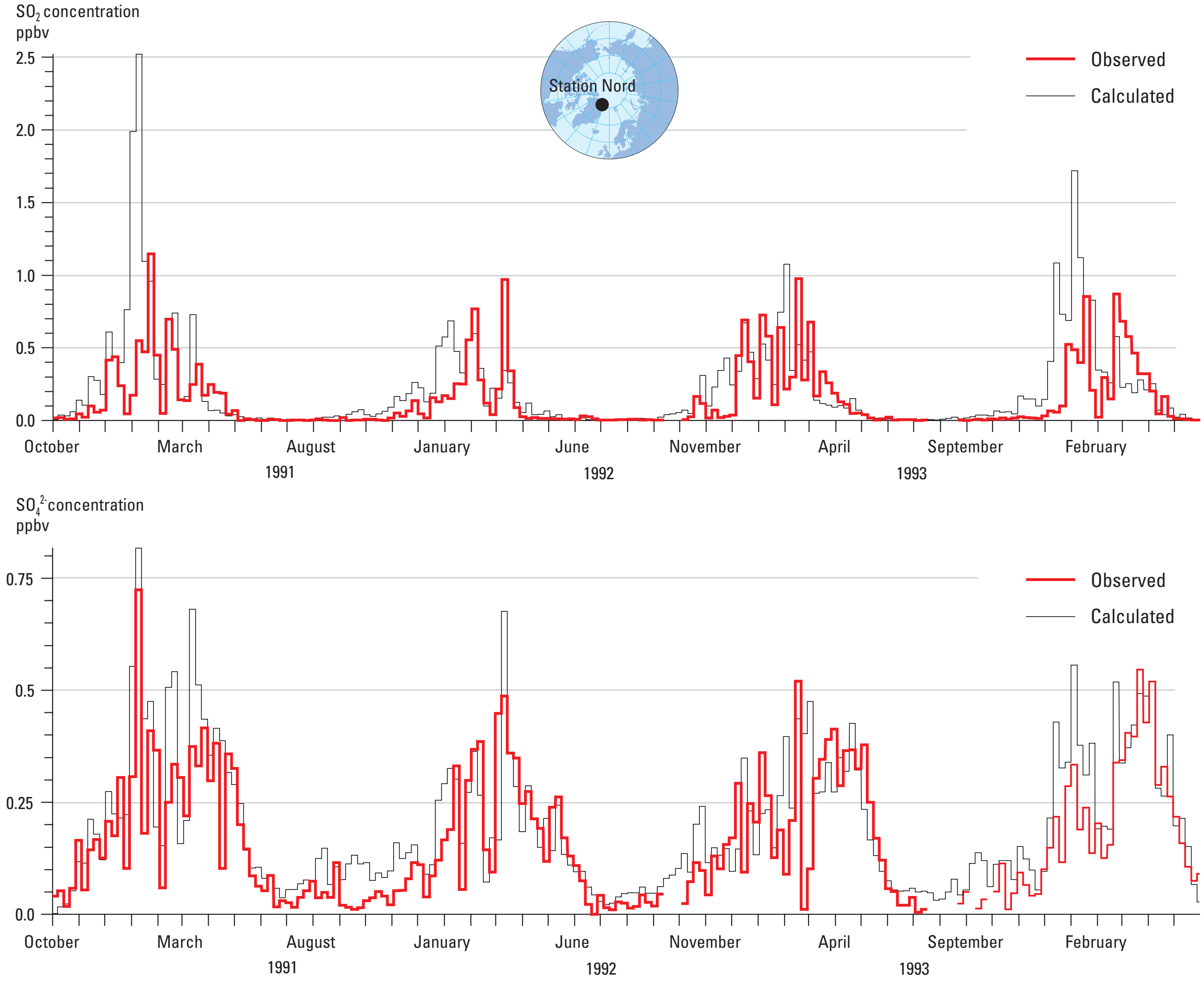

Figure 9·13. Calculated concentrations of SO_2 and SO_4^{2-} compared with measurements at Station Nord in northeastern Greenland (Christensen 1995, 1997).

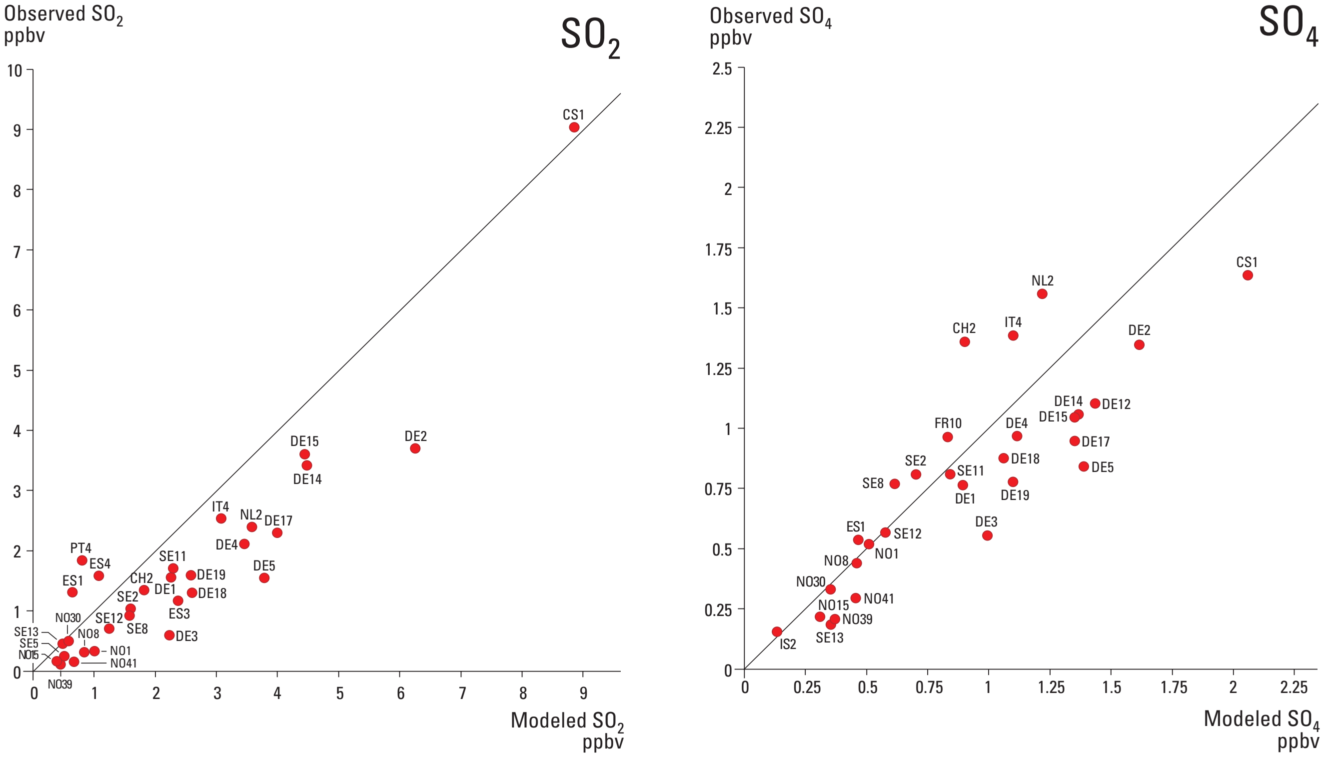

Figure 9·14. Scatterplots for comparisons between the average concentrations of SO_2 and SO_4 in air from October 1990 to December 1993 as monitored by the European Monitoring and Evaluation Programme (EMEP) and calculated by Christensen (1997). Point labeling refers to EMEP station numbers.

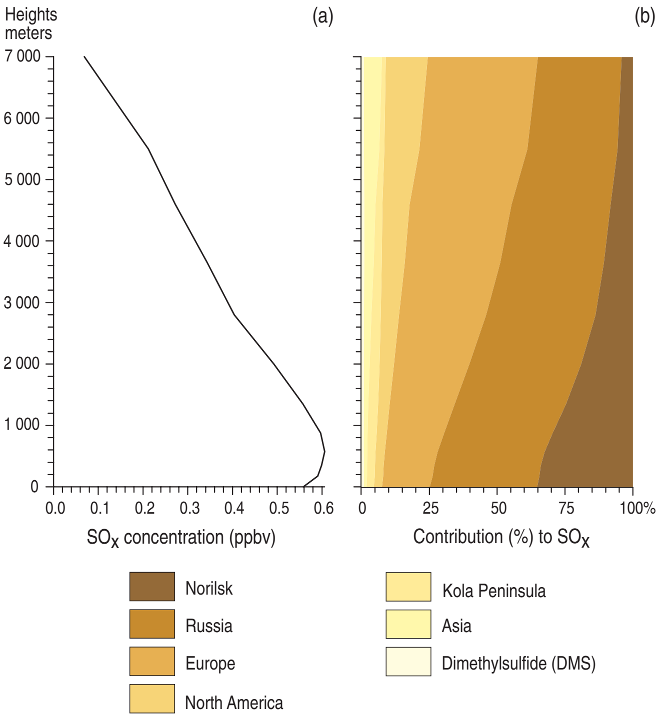

Figure 9·15. a) Vertical distribution of SOx ($SO_2 + SO_4^{2-}$) concentrations averaged over the area north of 75°N, and b) contributions from the different sources (Christensen 1995, 1997).

The contributions from the Kola Peninsula and North America are both about 3%, while DMS emissions and the sources in Asia both contribute about 1%. At higher elevations, the contributions from local sources in the Arctic, Norilsk and Kola Peninsula, decrease, while the long-range transported contributions from Europe, North America and Asia increase. The contributions from sources in the rest of the former Soviet Union remain at a level of about 40% throughout the vertical profile. As a whole, the contribution from the entire former Soviet Union to the concentration of SO_x in the Arctic boundary layer is about 78%.

In the three-dimensional global sulfur model by Dastoor and Pudykiewicz (1996), the time integration is performed using a semi-Lagrangian approach. The results of this model show that high levels of pollution are evident in the Arctic in winter months from all sources, confirming the qualitative picture outlined by Barrie (1986). There is a strong meridional transport in winter, which leads to high concentrations in high latitudes, particularly over the Arctic. In summer, however, the sulfur is mostly confined to mid-latitudes. Moreover, in summer, the sulfate concentrations over the Atlantic Ocean are higher than in winter, which is evidence of a stronger zonal flow and weaker precipitation scavenging over the North Atlantic.

On a more local scale, half-yearly average SO_2 concentrations of the air in Norwegian/Russian border areas have been reported by Sivertsen *et al.* (1992) and Sivertsen (1996). The results indicate that the high SO_2 concentrations extend more toward the northeast than to the south and southwest. However, the largest air pollution problems in the vicinity of the smelters are dominated by pollution episodes linked to adverse meteorological conditions. For Viksjøfjell, northeast of the smelters of Nikel and Zapolyarnyy, the modeled annual average SO_2 concentration is 20 µg/m^3, which agrees well with the measured annual average value, whereas the measured monthly averages vary between 10 and 55 µg/m^3 (Sivertsen 1996).

The areal distribution of average sulfur dioxide concentrations between July 1990 and July 1991 on the Kola Peninsula and Finnish Lapland has also been computed by a model developed by the Russian Institute of Global Climate and Ecology and the Finnish Meteorological Institute (see Tikkanen and Niemelä 1995). The results, shown in Figure 9·16, depict the distribution of the emissions from three smelters on the Kola Peninsula, demonstrating the significant local influence of these sources.

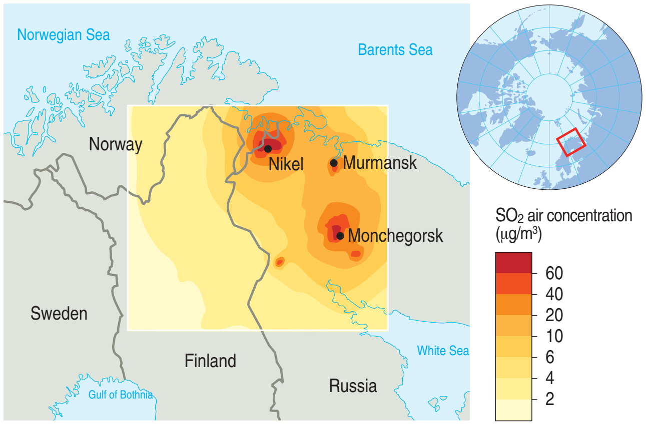

Figure 9·16. Model calculations of ground level yearly average sulfur dioxide air concentration on the Kola Peninsula (data source: Tuovinen, Finnish Meteorological Institute; in Tikkanen and Niemelä 1995).

9.5. Arctic haze

9.5.1. The Arctic haze phenomenon

In the mid-1950s, pilots flying in the Canadian Arctic reported a phenomenon of deep hazes which obscured visibility (Greenaway 1950, Mitchell 1956). The depth and coloration of the hazes suggested that the obscured visibility was not natural and was caused by micrometer-size particles. The effect was merely viewed as a curiosity and not analyzed in any great detail until the mid-1970s when the early studies of Shaw (1975) noted strange haze layers at Barrow, Alaska. At the same time, a Danish research group made observations of the haze in Greenland (Flyger *et al.* 1976). This obscured visibility focused attention on Arctic air pollution, and much of the work discussed in this assessment arose because of the abnormal nature of these hazes in the Arctic. In an evaluation of Arctic air pollution, Ottar *et al.* (1986) concluded that emissions from eastern Europe and the former Soviet Union were largely responsible for the Arctic haze that they observed in the Scandinavian Arctic. By the mid-1980s, chemical fingerprinting methods had determined that Eurasia and North America were the primary source regions for this general pollution (Rahn 1989).

From the ground, Arctic haze appears as a whitening of the sky near the horizon, with a reduction in visibility to a few kilometers or less. Viewed horizontally, from aircraft, the haze layers have a brownish tinge (Schnell and Raatz 1984, Radke *et al.* 1984, Raatz *et al.* 1985). This tinge or discoloration encourages the interpretation that the haze is absorbing light, and contains significant levels of organic particles. However, from what we have learned in the preceding sections, the tropospheric aerosol burden near the surface is largely sulfate with minor amounts of nitrates, soil, and organic material. The horizontal visual observations should be regarded with some caution since an aerosol that is totally scattering, with no absorption, will appear dark when viewed from certain angles.

The haze consists of a well-aged submicron aerosol with a mass median diameter of 0.2 µm or less (Heintzenberg

1980, Hoff *et al.* 1983, Pacyna *et al.* 1984, Shaw 1984, Leaitch *et al.* 1989, Trivett *et al.* 1989, Clarke 1989, Covert and Heintzenberg 1993, Hillamo *et al.* 1993). This particle size is well suited for scattering light (Waggoner and Weiss 1980) since the peak in the particle surface-area distribution is near the maximum efficiency for Mie scattering. Estimates of the chemical composition of Arctic aerosol range from 30% to 90% sulfate, the remaining material consisting of carbonaceous soot (<20%), soil (<30%) and marine aerosols (<10% in winter and <35% in summer) (Li and Barrie 1993). Nearly all of the light scattering of the haze aerosol can be attributed to the sulfate concentration, the ambient humidity and the presence of frozen water (ice crystals or snow) in the optical path. Nearly all of the light absorption at visible wavelengths appears to be due to carbonaceous material ('black carbon', 'soot' or 'graphitic carbon') (Hansen and Rosen 1984, Noone and Clarke 1988, Clarke 1989, Kahl and Hansen 1989, Hansen *et al.* 1992, Hopper *et al.* 1994).

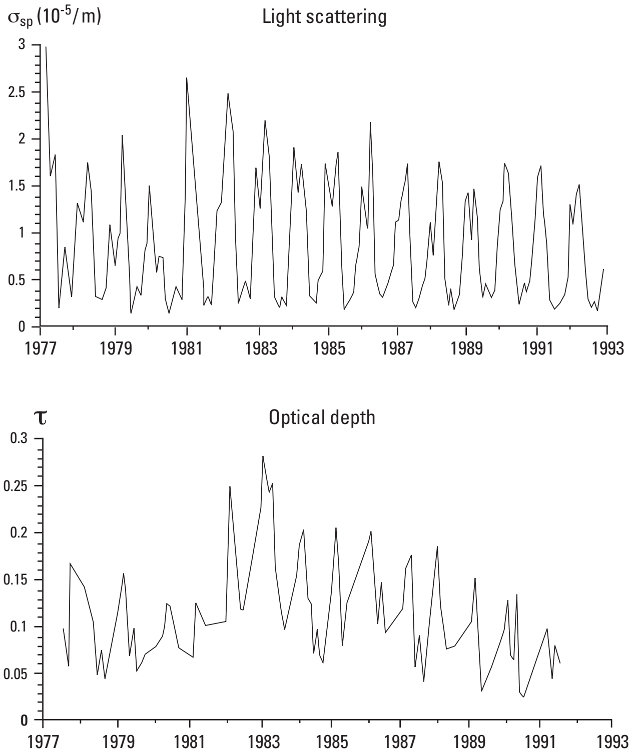

Figure 9·17. The time series of light scattering (σ_{sp}) and optical depth (τ) at 500 nm at Barrow, Alaska (Bodhaine and Dutton 1993).

Most of the information on light scattering in the Arctic has come from nephelometry (Heintzenberg 1980, Bodhaine 1983, 1989, Barrie *et al.* 1981, Barrie and Hoff 1985, Trivett *et al.* 1989, Heintzenberg *et al.* 1991, Bodhaine and Dutton 1993). Figure 9·17 shows the time series of light scattering and optical depth at 500 nm at Barrow, Alaska (Bodhaine and Dutton 1993). The aerosol scattering coefficient (σ_{sp}) has been related to horizontal visibility (V) by Koschmieder's formula (Koschmieder 1925) $V = 3.91/\sigma_{sp}$. During the AGASP II study of 1986, Trivett *et al.* (1989) reached the conclusion that the minimum short-term visibility which might be expected from Arctic aerosols in clear weather conditions is about 30 km. They attributed the much lower visibilities which occasionally occurred during their study to additional scattering from ice crystals in the atmosphere. Meyer *et al.* (1991) have reached a similar conclusion.

Valero *et al.* (1989) found the vertical optical thickness of clear-sky Arctic haze to be about 0.1-0.2 at maximum. This is consistent with Bodhaine and Dutton's results in Figure 9·17. Optical depth maxima on an annual basis obtained from hemispheric models have been predicted to be 0.02-0.04 (Iversen and Tarrason 1995). Optical depths in ice crystal layers, which are visually transparent, can reach 0.9 or greater (Hoff and Leaitch 1989).

Absorption in the haze has been measured using an aethelometer to be less than 6×10^{-7}/m (Hopper *et al.* 1994), which is about 10% of the average scattering in Figure 9·17. This is consistent with the estimated single scatter albedo of 0.86-0.96 (Hansen and Rosen 1984, Clarke 1989).

Vertical profile information on Arctic haze aerosols has been widely obtained by lidar (Hoff 1988, Radke *et al.* 1989, Trivett *et al.* 1989, Brock *et al.* 1990, Schnell *et al.* 1989, Morley *et al.* 1990). In the vertical direction, the main mass of Arctic aerosols is limited primarily to the lowest five kilometers, peaking in the lowest two kilometers of the atmosphere, due to the strong inversions present in the Arctic (Leaitch *et al.* 1984, Barrie 1996, Hoff 1988, Pacyna and Ottar 1988).

More recently, Arctic thin cloud and ice crystal scattering have been observed by satellite remote sensing (Kergomard and Tanre 1989, Ebert and Curry 1992). The main conclusions of these studies are that Arctic clouds are underestimated in climatologies of the Arctic and may have greater radiative impacts than previously predicted.

As early as 1981, it was speculated that such hazes might affect the Arctic climate (Barrie *et al.* 1981). Climate modeling studies reveal that there might be measurable effects, but that the effects would be small (Cess 1983, Blanchet 1989, Maxwell and Barrie 1989). Recent speculations and Global Circulation Model (GCM) predictions of the impact of sulfate aerosols on climate (Charlson *et al.* 1991, 1992, Kiehl and Briegleb 1993, Taylor and Penner 1994, Box and Trautman 1994, Iversen and Tarrason 1995) have renewed interest in this possible impact of Arctic haze.

9.5.2. Occurrence of Arctic haze

Arctic haze is predominantly a thin sulfate haze which is augmented by humidity effects and by ice crystals. The formation of Arctic haze is a result of unusual physical and meteorological conditions in the Arctic, as described by Shaw (1995). Strong surface-based temperature inversions form in the polar night, causing the atmosphere to stabilize. Turbulent transfer between the atmospheric layers is inhibited, and the removal of aerosols and gases is blocked. Moreover, this stable and cold atmosphere in the polar regions inhibits the formation of cloud systems that can give rise to precipitation. Suppression of these removal mechanisms for trace constituents in the Arctic atmosphere is one important cause of the contamination of Arctic air in winter and spring. Other important mechanisms contributing to the contamination are the atmospheric meridional transport and the slow rate of chemical transformation in Arctic air during winter.

In a recent study of visibility observations in Canada, it was found that haze production in winter is widespread throughout Canada, even at relatively low latitudes (Hoff 1994). Figure 9·18 shows the contours of median visibilities in Canada for the summer and winter months during 1951-1993. The results shown are computed only from cases where the relative humidity (RH) is less than 80% in order to exclude poor visibility cases caused by moist aerosols, fog or precipitation. These visual ranges should correspond to dry aerosol obscuration of visibility. The 35-km contour extends down from the Arctic to below Hudson Bay, a shape that is very similar to the position of the Arctic front in winter (see chapter 3, Figure 3·4).

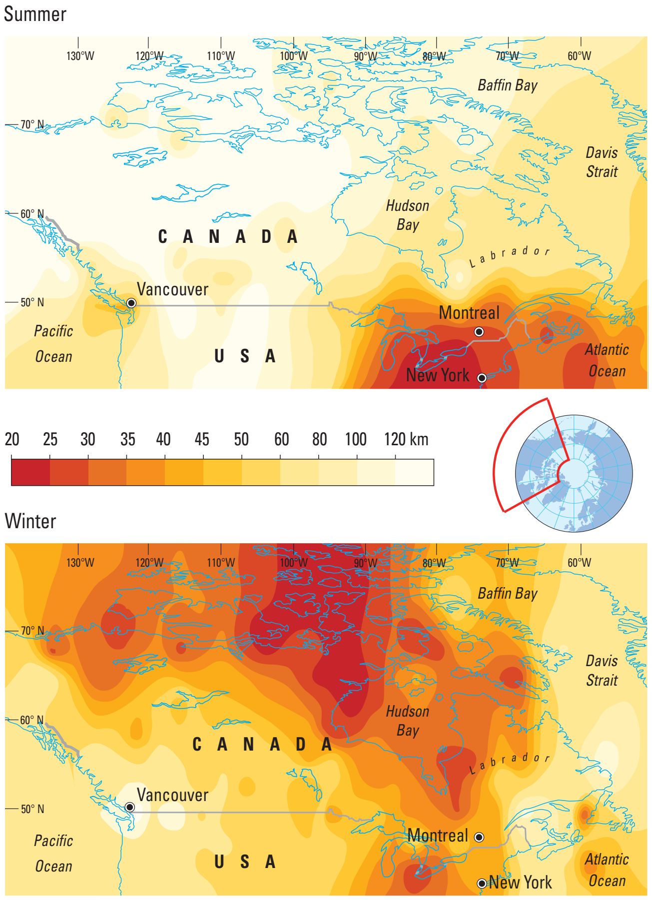

Figure 9·18. Summer and winter visibility observations (for relative humidities less than 80%) for the period 1951 to 1993 in North America (Hoff 1994).

European sources contribute most of the Arctic haze in winter, but in summer, when haze is at a minimum, the North American and Asian sources produce 30-70% of the haze. According to model results, the natural production of DMS (dimethylsulfide) contributes 1-9% of the Arctic haze in summer but is insignificant in the winter (<1%) (Tarrason 1995, Tarrason and Iversen 1996) (Figure 9·19). This estimate is clearly lower than the one by Li and Barrie (1993), who conclude that in summer about 30% of the sulfur is from natural sources.

In winter, the concentrations of sulfate are highest (0.14-0.21 µg/m^3) in the atmosphere at altitudes below 2 km, while in summer the differences between altitudes are not so distinct (Figure 9·19). With respect to the vertical distribution averaged over the whole year, there is a less sharp decline towards lower altitudes than in winter. The concentration is, on average, 0.10 µg/m^3, being slightly higher at altitudes less than 1500 meters (up to 0.15 µg/m^3). Europe contributes about 70% to the concentrations at altitudes higher than 1000 m, while North America contributes less than 10%.

9.6. Deposition of acidifying compounds

9.6.1. Wet and dry deposition

Sivertsen *et al.* (1992, 1993) have mapped the deposition of sulfate in precipitation in northern Fennoscandia, using available precipitation sampling stations. Also, Jaffe *et al.* (1995) have surveyed snowpack samples on the Kola Peninsula. These procedures are probably not satisfactory for mapping total deposition in the area close to the large emission sources on the Kola Peninsula. The measured sulfate deposition by precipitation or snow collected at the sites closest to the emission sources are only about twice as high as the general levels in the area (Jaffe *et al.* 1995, Sivertsen *et al.* 1993), the average annual deposition by precipitation on the Kola Peninsula and adjoining parts of Norway and Finland being only about 200 mg S/m^2. It is concluded, therefore, that most of the deposition occurs in summer as dry deposition to vegetation. The low deposition velocity of SO_2 to snow, and its slow oxidation to sulfuric acid, lead to enhanced transport of sulfur away from the sources (Sivertsen *et al.* 1992, Tuovinen *et al.* 1993, Barrie 1995, Jaffe *et al.* 1995).

The amount of accumulation of acidic species in surface media resulting from dry deposition depends on the concentration of the compound in the boundary layer, its transport toward the active surface by irregular air movements in the turbulent boundary layer close to the ground, diffusion

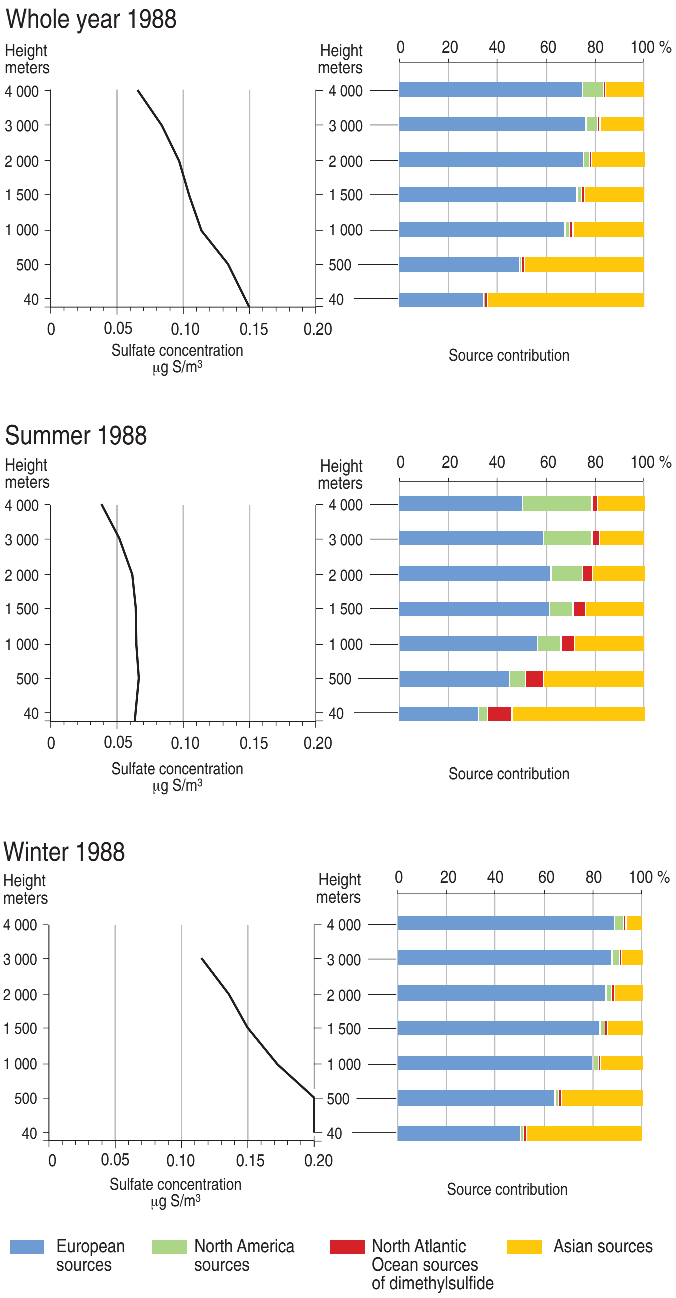

Figure 9·19. Vertical distribution of Arctic haze in summer, winter, and the whole year. Sulfur air concentrations and relative source-contribution (%) for different heights above the ground are shown. The concentrations are averages over the Arctic area north of 70°N (modified from Tarrason 1995, Tarrason and Iversen 1996).

through the laminar surface layer, and diffusion and chemical reactions within the surface elements. Theoretical and experimental studies of dry deposition have mainly been carried out over arable land, agricultural crops and forest, and have shown typical deposition velocities for sulfur dioxide in the range 0.5-1 cm/s, while the deposition rate for sulfate particles, which are typically in the submicron size, is much less (Lövblad *et al.* 1992). In general, deposition rates are lower for areas with sparse vegetation, and in the dormant season for vegetation.

Deposition rates on snow and snow-covered surfaces are generally very low, as has been found in several experimental studies. This is partly due to the high aerodynamic resistance (r_a) over snow surfaces (Dovland and Eliassen 1977), and partly to the fact that snow crystals do not take up sulfur dioxide directly. However, at temperatures above –17°C, a liquid phase consisting of ionic impurities is also present in snow. This liquid phase is responsible for the uptake by snow of SO_2 and H_2O_2.

Dry deposition rates of SO_2 and SO_4 are low in the Arctic because of the high fraction of snow and ice-covered surfaces. This is indirectly demonstrated by the relatively high sulfur concentrations during winter and spring in the High Arctic, which is only possible because of the low deposition rates during transport. However, because of the very low precipitation rates, dry deposition may still account for a significant part of the total deposition in some areas. More detailed studies are possible around large emission sources where concentrations of sulfur dioxide may also be high in the summer. The dry deposition of sulfur is estimated to be about 75% of the total deposition on the Kola Peninsula (Tuovinen *et al.* 1993).

Table 9·3. Monthly mean air concentrations of sulfur dioxide and sulfate measured at Svanvik, together with deposition velocities and inferred dry deposition from canopy throughfall measurements.

	Measured concentrations, µg/m^3		Deposition velocities[a], cm/s		Inferred dry deposition mg/m^2
	SO_2-S	SO_4	Sulfur dioxide	Sulfate particles	
January	8.21	0.75	0.2	0.05	45
February	4.95	1.05	0.2	0.05	28
March	24.3	1.73	0.2	0.05	132
April	4.02	0.94	0.2	0.05	23
May	3.44	0.58	0.8	0.1	75
June	7.81	0.59	1	0.15	212
July	4.08	0.45	1	0.15	111
August	4.76	0.69	1	0.15	130
September	2.54	0.27	1	0.15	69
October	1.84	0.32	0.8	0.1	40
November	4.18	0.2	0.2	0.05	23
December	2.48	0.37	0.2	0.05	14
Year					902
Dry deposition indicated from canopy throughfall measurements[b]			Mellesmo (spruce) Svanhovd (pine)		1200 745

a. Tuovinen *et al.* 1993. b. Aamlid 1992a.

Canopy throughfall measurements made at Svanvik, northern Norway, give clear evidence that dry deposition of sulfur dioxide is a dominating acidifying process in this area (Aamlid 1993a, SFT 1992). Two stands, one of spruce (*Picea abies*) and one of pine (*Pinus silvestris*), have been monitored. The spruce stand gives somewhat higher dry deposition values than the pine stand, which may be a result of the generally higher leaf-area index and growth rate of spruce relative to pine. The measured concentrations of sulfur dioxide and sulfate using the same deposition velocities as were used in the computations by Tuovinen *et al.* (1993) give the estimated dry deposition of sulfur dioxide shown in Table 9·3.

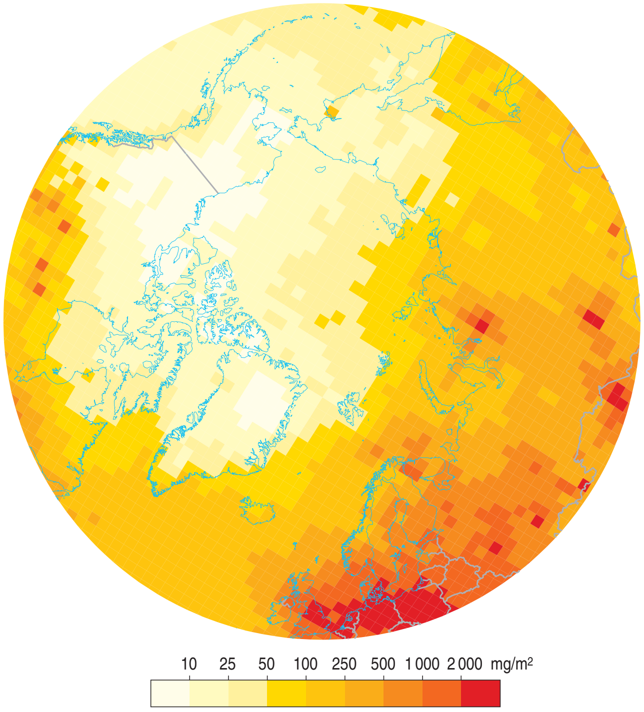

Figure 9·20. Model calculations of total (wet and dry) deposition estimates of sulfur for 1988 (data source: Norwegian Meteorological Institute).

9.6.2. **Modeling deposition**

In Europe, the EMEP program carries out dispersion and deposition modeling at its two Meteorological Synthesizing Centers, in Oslo and in Moscow (MSC-W and MSC-E, respectively). The results of the hemispheric transport model developed by the Norwegian Meteorological Institute (Tarrason 1995, Tarrason and Iversen 1996, Iversen and Tarrason 1995) predict that, within the Arctic, there are two main areas with annual sulfur deposition rates in excess of 500 mg S/m^2/y (Figure 9·20). One of these is northern Fennoscandia and the Kola Peninsula, and the other is around Norilsk in northwestern Siberia. MSC-E has also carried out preliminary model calculations for the northern hemisphere, including estimated deposition fields of sulfur and nitrogen compounds (Galpierin *et al.* 1994). These calculations also indicate very low deposition rates in the Arctic in general.

The total deposition of sulfur, calculated from the model of Christensen (1995, 1997), averaged over the area north of 75°N for the four winters of 1990-1994, is about 6.7 mg S/m^2/month. The contributions from the different sources to the total deposition are shown in Figure 9·21a. The figure shows that the Norilsk area and the Kola Peninsula contribute 18% and 17%, respectively, while the rest of the former Soviet Union and Europe both contribute about 28%. It is interesting to note that although the contribution of the Kola Peninsula to surface air concentrations is as low as 3% (see Figure 9·15), the contribution to the total deposition is 17%.

There is a seasonal variation in the total deposition of SO_x, from 3-6 mg S/m^2/month in summer to 8-12 mg S/m^2/month in winter (see Figure 9·21b). The total deposition shows smaller seasonal variation than the mean concentrations of the Arctic boundary layer, where the winter concentrations are about a factor of 10 higher than the summer concentrations, indicating that in the summer period the atmospheric transport to the Arctic occurs at higher altitudes (National Environmental Research Institute 1995). The sources from Russia have a large seasonal variation, contributing about 15% in the summer period to more than 50% in the winter period (Figure 9·21c).

Calculations on a more local scale, including deposition both from distant sources and from sources on the Kola Peninsula, have been carried out by Tuovinen *et al.* (1993) for northern Fennoscandia. Seasonally averaged dry deposition velocities were used to estimate the dry deposition of sulfur dioxide and sulfate aerosol, taking into account the long period of permanent snow cover, from October to April. Seasonally averaged washout coefficients were used to estimate the deposition by precipitation, assuming the rate of deposition to be proportional to the washout coefficient, the square root of the precipitation intensity, and the mean concentrations of sulfur dioxide and sulfate in the mixing layer. The calculated total deposition of sulfur is shown in Figure 9·22. Tuovinen *et al.* (1993) conclude that within the area of annual deposition in excess of 0.3 g S/m^2/y, dry deposition accounts for more than 60% of the total deposition, and the contribution of the emissions from the sources on Kola may be more than 50% at distances of up to 200 km. In spite of the very high emissions from the non-ferrous metallurgical smelters in this area, these sources only dominate the deposition in the 150 × 150 km^2 grid cells where these emission sources are located, and their contribution to deposition in the neighboring grid cells is typically around 20% (Tuovinen *et al.* 1993). Similar calculations have been performed for the dispersion of sulfur dioxide and other pollutants in the immediate surroundings of the smelters by Sivertsen *et al.* (1993) with similar results.

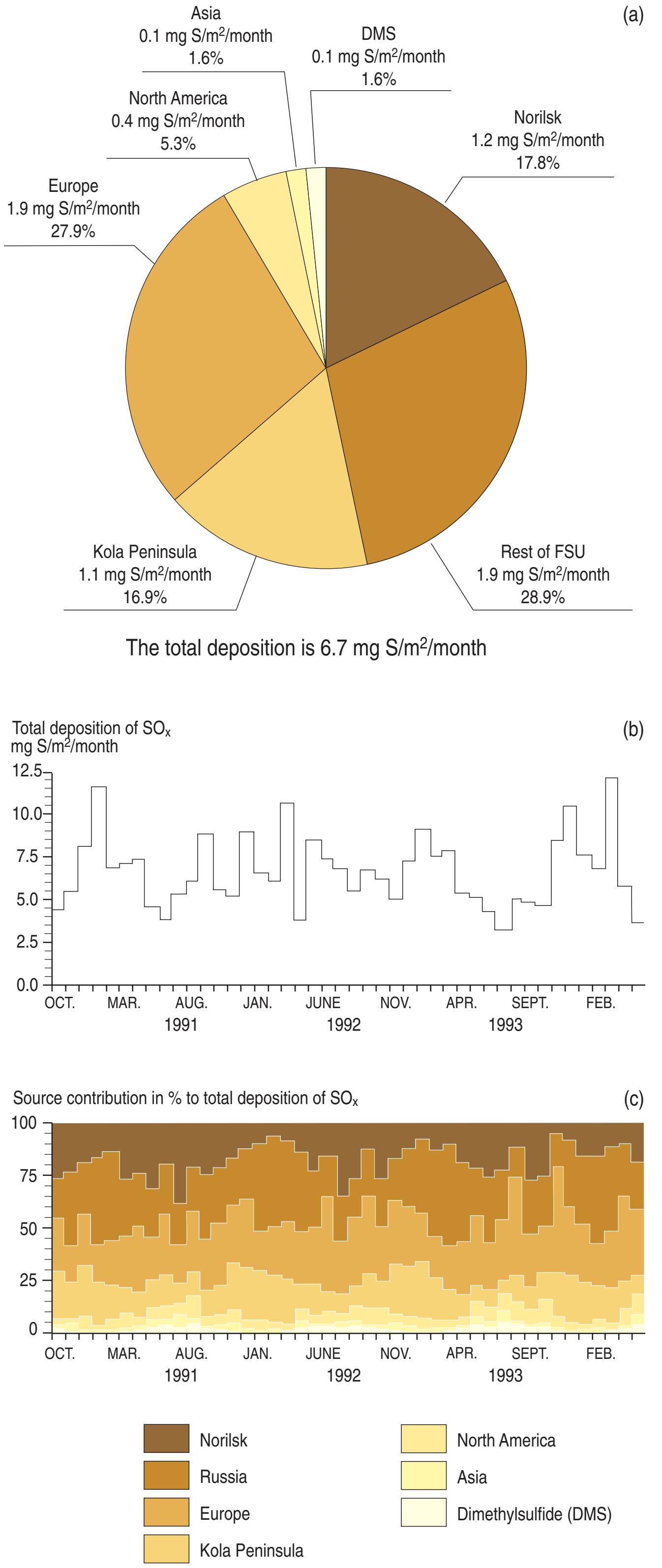

Figure 9·21. a) The contribution from different sources to the total deposition, averaged over the area north of 75°N from October 1990 to May 1994 (Christensen 1995, 1997), b) the calculated total monthly depositions of SO$_x$ averaged over the area north of 75°N, and c) the contributions from different sources to the total monthly deposition (Christensen 1995, 1997).

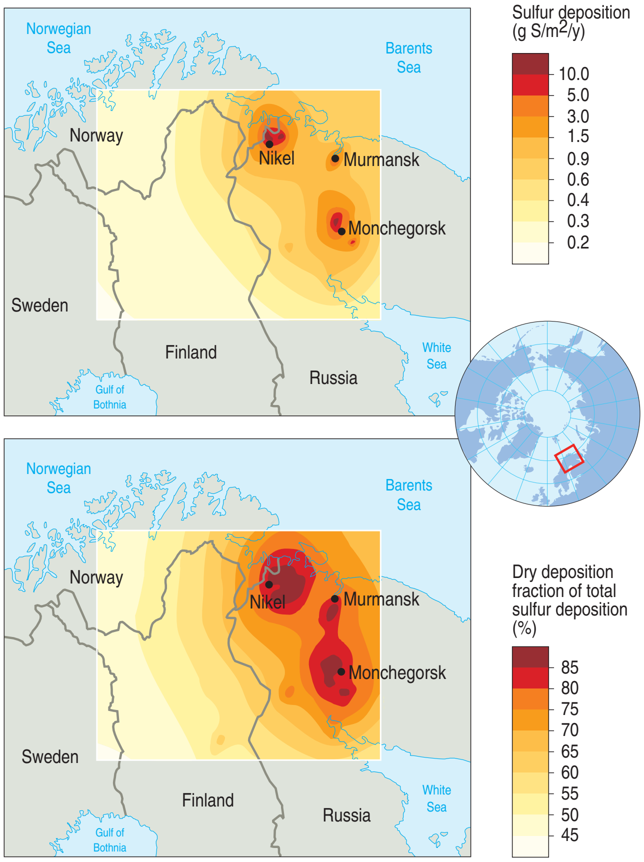

Figure 9·22. Model calculations of sulfur deposition and fraction of dry deposition for the period July 1990 to June 1991 in northern Fennoscandia and the Kola Peninsula (modified from Tuovinen *et al.* 1993).

9.7. Processes regulating terrestrial and aquatic acidification

9.7.1. Soil acidification

9.7.1.1. Chemical processes

Arctic regions belong to the area of glaciated terrain. In presently non-glaciated areas of the Arctic, glaciogenic drift, mainly till, covers much of the bedrock. Tundra soils are predominantly poorly drained, consisting of a mineral soil overburden covered by an organic layer (humus).

The predominant soil-forming processes in forested and treeless (tundra) regions of the Arctic and subarctic are podzolization on mineral soil sites and peat accumulation in waterlogged areas. The cold, moist climate (precipitation exceeding evaporation), and in many areas a thick permafrost layer, are the main reasons for the development of extensive wetland areas and paludified or semi-paludified mineral soils. Many peat soils are naturally rather acidic, and their extremely high organic content means that they are strongly buffered and, therefore, relatively unaffected by acidic deposition levels that can strongly increase soil acidification in mineral soils. The transport of dissolved and particulate organic material by percolation water and surface runoff has a considerable effect on water quality in the watercourses, as well as on the properties of the underlying mineral soil.

Podzol horizons have mainly developed in till and sorted, coarse soils, i.e., those containing a high proportion of sand and gravel. Podzolic soils are characterized by a light-colored eluvial horizon located immediately below the organic layer, and below this, a dark-colored illuvial horizon. The eluvial horizon has been strongly leached (weathered) by acidic and dilute (i.e., low in minerals or electrolytes) percolation water. The pH of the percolation water gradually increases as it passes down through the reddish-brown illuvial horizon. Aluminum, iron and silica compounds and organic matter dissolved in the percolation water are precipitated in the eluvial horizon as aluminum-iron-silicon hydroxides (Rose *et al.* 1979, De Cornick 1980, Farmer *et al.* 1980, Anderssen *et al.* 1982). The generally high soil moisture content, commonly anaerobic conditions, and slow decomposition rate of plant litter result in the gradual accumulation of an organic layer consisting of acidic, partially decomposed litter and humic material in the surface soil layer (Strahler 1970, Jacks *et al.* 1984).

In podzolic soils, the equilibrium between the chemical composition of the soil and soil water is controlled by biological and geochemical processes: organic matter decomposition, nutrient uptake, weathering, leaching, cation exchange, and aluminum, iron and silicate buffering. The chemically modified, surficial horizons of podzolic soils are underlain by relatively unaltered parent material. The geochemical composition of this material determines to a considerable extent the elemental composition of the upper soil horizons, as well as the type of vegetation that develops on the site (Andersson 1988, Binkley and Richter 1987, Boyle and Voigt 1973).

Minerals in soil and bedrock differ markedly in their susceptibility to weathering and solubility of elements (Rose *et al.* 1979). In glaciated regions, the soil and bedrock are mainly composed of silicates. The weathering of silicate minerals is the neutralization process within soils; this is limited by the slow weathering rates of the most common minerals in Arctic soils. In Europe, carbonate-bearing minerals, which are readily weathered, play only a local role, and hence the main femic minerals (amphiboles, pyroxenes and micas) are the most important in the neutralizing process.

Secondary clay minerals have a large reaction surface and play an important role in buffering processes and in, e.g., sulfate adsorption. Adsorption may immobilize a significant fraction of incoming sulfur and delay acidification by decades or even centuries (Eriksson 1988, Warwood *et al.* 1986, Courchense and Hedershot 1989). Young soils in glaciated terrain typically have small sulfate adsorption capacities, and as a consequence, the time-lag between increasing sulfate deposition and increasing sulfate runoff is much shorter (Rochelle *et al.* 1987). However, sulfate mobility in the surface soil may be much lower in podzolic soils in the North than is generally realized. The aluminum and iron hydroxides in the illuvial horizon form anion binding sites that rather effectively remove sulfate from the soil solution (Jonsson and Cole 1977, Parfitt and Smart 1978, Jonsson *et al.* 1986, McDonald 1987).

Cation exchange on the surfaces of soil particles in the humus and eluvial horizons is the most important buffering process, capable of retarding the acidification process (Boyle and Voigt 1973, Petersen 1976, Evans 1980, Brady 1984, Andersson 1988). The exchangeable cations include Ca^{2+}, Mg^{2+}, K^+, and Na^+, which are replaced during the acidification process mainly by Al^{3+} and H^+ ions. Cation exchange is controlled by soil pH, grain size, moisture content and the organic matter content and is affected by the abundance and type of clay minerals. In the mineral soil, cation exchange takes place primarily in the fine fraction, owing to the large, negatively charged surface area of fine particles. In coarse-grained soils, e.g., sands and gravels, cation exchange is low owing to the low clay content; however, this is somewhat offset by the presence of relatively large amounts of organic material (Birkeland 1974).

9.7.1.2. **Hydrological processes**

The equilibrium reactions between soil and runoff water are greatly affected by precipitation patterns. The rate at which water passes down through the surface soil, eventually reaching the watercourses or groundwater, depends on the soil depth, grain size, organic matter and moisture content, as well as on topography, vegetation cover and season. The higher the content of fine material (inorganic and organic) in the soil, the higher the water-retention capacity. The retention time of water in such soils is long, and sufficient for the slow buffering and neutralization reactions between percolation water and soil particles to reach equilibrium.

In the case of very fine-textured and compact soils, e.g., clay, or in permafrost areas, water cannot infiltrate the subsurface soil layers. Although some of the water is lost as surface runoff, such sites usually become paludified or develop into wetlands. In coarse-grained soils (gravel and sand formations, eskers), with a relatively low organic matter content, the water percolates rapidly down into the groundwater, and the effect of overburden properties on percolation-water quality is minimal. In the case of surface runoff, the water has only very short-term contact with the organic layer or exposed bedrock, and chemical modification (e.g., neutralization) is slight. A high proportion of exposed bedrock and rough topography increase the proportion of surface runoff.

9.7.1.3. **Biological processes**

Microbiological activity in the soil in Arctic and subarctic regions is, in general, rather low. This is a result of the low mean soil temperature during the short summer period, the presence of permafrost in many areas, the low proportion of easily decomposable material in the plant litter, and the restrictions on microbial activity set by the high soil moisture content following snowmelt in the spring (Boyle and Voigt 1973, Binkley and Richter 1987, Andersson 1988). Anaerobic conditions in waterlogged soils are also a frequent occurrence. Saprophytic fungi, which are adapted to acidic conditions and which also contribute to the maintenance of low soil pH through the exudation of organic acids and formation of humus substances, are predominant.

9.7.1.4. **Natural and anthropogenic soil acidification**

Soil acidification, i.e., a gradual decrease in soil base saturation and a consequent decrease in pH over time, is a natural process in both mineral and peat soils in the Arctic region. Peat formation and podzolization are associated with a gradual accumulation of protons in the surface soil. In those parts of the Arctic and subarctic region with a natural forest cover, the gradual accumulation of protons is interrupted by the recurrent outbreak of forest fires; fire destroys much of the surface acidic organic matter, and the ash which is produced has a strong neutralizing effect. In such ecosystems, there is a clear acidification/neutralization cycle. However, this cycle of events is not so pronounced on peatlands and in the Arctic tundra.

In addition to acidic deposition, anthropogenic soil acidification can be caused by a number of factors, many of which are associated with commercial forestry practices. The cultivation of even-aged stands results in the gradual accumulation of base cations in the biomass and a decrease in soil pH over time. These base cations are lost from the ecosystem to a varying extent in timber harvesting. The theoretical soil acidification due to cation removal in connection with whole-tree harvesting, for instance, is of the same order of magnitude as the soil acidification caused by acidic deposition (Nilsson *et al.* 1982).

Elevated levels of acidifying deposition (sulfate and nitrate) can result in an increase in the proton pool and displacement of base cations, i.e., decrease in base saturation (BS) from the humus layer and uppermost mineral soil layers, and also increase the proportion of aluminum and heavy metals on exchange sites in the mineral soil and in the soil solution. Base saturation and exchangeable aluminum concentrations are the most widely used soil acidification parameters.

9.7.2. Aquatic acidification

9.7.2.1. Processes governing aquatic acidification

Chemical weathering and the ion exchange processes occurring in catchments supply the majority of base cations found in surface waters. Calcium and magnesium are generally the dominant cations in clear-water lakes located in areas subject to low levels of acidic deposition. In areas near the marine coast, sodium, chloride and other marine aerosol salts contribute significantly to the ionic composition of surface waters. In clear-water lakes, with low dissolved mineral content, located in areas of low levels of acidic deposition, bicarbonate is normally the dominant anion, while in lakes receiving high deposition of sulfate, sulfate is the dominant anion. Organic anions are important contributors to the ionic composition in humic lakes having high concentrations of dissolved organic carbon (DOC).

The key to surface water acidification lies in soil chemical processes; most of the incoming strong acids from the atmosphere are deposited on terrestrial catchments, and the soil processes alter the chemical composition of runoff. Soil processes are thus central to understanding the changes introduced by acid deposition (Reuss and Johnson 1986).

Water acidification can be determined as the loss of alkalinity. Acidification of soil and waters requires the presence of mobile anions to mobilize and transport acid cations (Seip 1980). Only six anions need to be considered: the strong acid anions Cl^-, F^-, NO_3^- and SO_4^{2-}, organic anions and bicarbonate (HCO_3^-). Although NO_3^- is extremely mobile in soils, nitrogen is often a growth-limiting nutrient in northern terrestrial ecosystems, and thus most of the incoming nitrogen is usually taken up by the terrestrial biomass. Acidic deposition increases the external source of SO_4^{2-}, and the accompanying cations are mainly acidic (H^+ and NH_4^+). Sulfate in neutral cyclic seasalts is normally the only other source of importance. Atmospheric inputs generally far exceed the biological sulfur requirement.

Sulfate reduction and retention in lake sediments are the most important processes within lakes (in-lake processes) that may speed up the reversal of lake acidification (Wright and Hauhs 1991). The importance of the in-lake processes depends on the flushing time of the lake: for lakes with rapid water renewal, in-lake processes are of minor importance. The ability of a lake to generate alkalinity by in-lake processes is called in-lake alkalinity generation. The most important in-lake processes in the lake are microbial sulfate reduction, microbial denitrification, algal uptake of nitrate and the release of calcium, magnesium, potassium, iron and manganese from lake sediments (Schindler *et al.* 1991).

Although the soil processes within the catchment control the long-term acidification of surface waters, the hydrological processes are central in determining the daily to monthly acidity dynamics. The occurrence of acid pulses during snowmelt is a widespread phenomenon in small streams in steep terrain draining watersheds with thin or no soil cover, combined with large acid snow accumulation and a rapid thaw. Initially during snowmelt, impurities, e.g., acids and metals, from the whole snowpack migrate through the snow and are released with the first meltwater. This can result in an acid pulse with a pH below 5.5, which can directly affect the draining streams due to low interaction with the frozen soil (Bjärnborg 1983, Jacks *et al.* 1986, Olofsson *et al.* 1995). Later during snowmelt, the water quality improves as soil water predominates in the meltwater, i.e., when the ground is no longer frozen. The lowering of pH that can occur in this phase is mainly due to dilution and the contribution of natural weak organic acids from the humus layer.

9.7.3. Calculating critical loads

Critical load (CL) is defined as a quantitative estimate of the loading of one or more pollutants below which significant harmful effects on specified sensitive elements of the environment are not likely to occur according to current knowledge (Nilsson and Grennfelt 1988). The methods used to calculate critical loads for air pollutants have been under intensive development. The methodology has evolved in several workshops (e.g., Nilsson 1986, Nilsson and Grennfelt 1988, Grennfelt and Thörnelöf 1992), and the regional characteristics of the critical loads have been demonstrated in numerous scientific publications (e.g., Henriksen *et al.* 1990, Kämäri *et al.* 1993). In Europe, data on critical loads have been collected at the Coordination Center for Effects (located in the Netherlands) for use in negotiations on air pollution control within the context of the Convention on Long-Range Transboundary Air Pollution (see Hettelingh *et al.* 1991, Downing *et al.* 1993, Posch *et al.* 1995). Moreover, a manual has been produced for guiding the European mapping exercise (ECE 1995).

The critical load of acidity to surface waters is estimated on the basis of present water chemistry. The aim is to calculate the flux of base cations in preindustrial conditions on the basis of present base cation concentrations. The critical load of acidity is then the base cation flux exceeding the selected acid-neutralizing capacity (ANC) threshold. This so called ANC-limit is derived on the basis of tolerance of fish in Norwegian lakes (Henriksen *et al.* 1990). The ANC is in turn defined as the difference between non-marine base cations and strong acid anions.

For soils, the critical loads are calculated by quantifying the various fluxes that in the long-run contribute to the neutralization of the soil (base cation weathering or uptake), or to the retention of the acidity within the soil (sulfur and nitrogen uptake, immobilization). The critical load is then

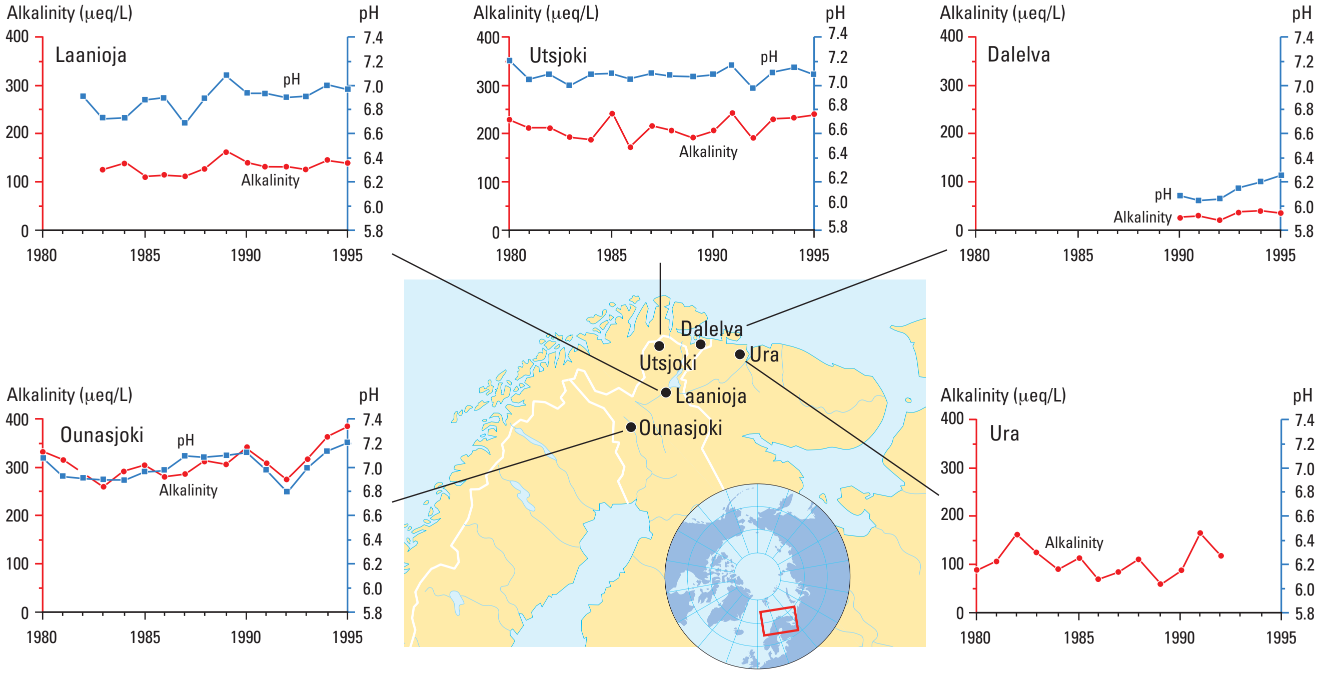

Figure 9·23. Annual average of alkalinity and pH as measured in rivers in northern Fennoscandia and the Kola Peninsula (data sources: Ounasjoki, Utsjoki and Laanioja – Finnish Environment Institute, unpubl.; Dalelva – Traaen pers. comm.; Ura – Moiseenko pers. comm.).

determined by the residual of these fluxes in excess over a selected threshold which is assigned for the leaching of ANC out of the soil.

Lately, the critical load of both surface waters and soils has been shown to be a function between sulfur and nitrogen deposition (e.g., Posch *et al.* 1993a, 1995, 1997). In other words, the critical load of one of these pollutants cannot explicitly be determined without making assumptions about the other. The shape and values of the function are determined by the above-mentioned fluxes.

9.8. Trends indicating acidification

9.8.1. Terrestrial parameters

9.8.1.1. Soil solution and percolation-water quality

The monitoring of soil solution and percolation-water quality is an effective means of detecting early signs of anthropogenic soil acidification. Continuous non-destructive sampling can be performed at the same point, thus eliminating many of the problems associated with the extremely high spatial and temporal variation in many soil properties. Sampling below the different genetic soil horizons also provides information about the capacity of the soil to buffer and neutralize acidifying deposition as it passes down through the soil profile.

Most of the monitoring work carried out on soil solution and percolation-water quality has been performed in Fennoscandia, as well as to some extent on the Kola Peninsula. Many of the monitoring points in the Arctic and subarctic areas were originally established as ‘clean’ background stations in the national monitoring network of individual countries. There is a clear south-north gradient in acidifying deposition across Fennoscandia and no signs of accelerated acidification have yet been detected in surface soil water in northernmost Norway or Finland. Elevated Ni, Cu, Al and S concentrations in soil water in Norway, Finland and Russia are restricted to the immediate vicinity (<50 km) of the Cu-Ni smelters at Monchegorsk and Nikel on the Kola Peninsula (Tikkanen and Niemelä 1995). Maximum reported total Al concentrations in soil water are about 2 mg/L, which is below values generally considered to be thresholds for the onset of Al-induced root damage (Lindroos *et al.* 1996). The relatively high organic matter content of podzolic soils in the area also means that a high proportion of the total Al is in complexed form, i.e., non-toxic to plants (Lindroos *et al.* 1994).

According to the ion balance studies that have been carried out on precipitation, canopy throughfall and soil water chemistry, it would appear that many of the soils in Arctic and subarctic areas are, for the present, well capable of counteracting current levels of acidifying deposition. However, the gradual enlargement of the proton pool in the surface soil will, at some time in the future, undoubtedly pose a threat to forest and tundra ecosystems that are not subjected to natural forest fire/regeneration cycles.

9.8.2. Aquatic parameters

9.8.2.1. Streams and rivers

An assessment of national monitoring data on river water chemistry from 1963 to 1989 has revealed trends indicating acidification in the rivers of northern Finland (Kinnunen 1990, 1992). According to new regional and temporal assessments, including observations for the years 1970-1995 (Vuorenmaa, unpubl.), acidification in northern Finland has stopped; the buffering capacity of the larger rivers has remained high, and there has been no consistent downward trend in water alkalinity and pH values since the beginning of the 1970s. Instead, there are signs of increasing alkalinity values during the years 1980-1995, a relatively wet period during which more strategic sampling produced a more homogenous data set, compared to that sampled during the relatively dry 1970s. There are no signs of proceeding acidification detectable in either the river Utsjoki or the Laanioja stream (included in the national monitoring program for small drainage basins) on the basis of a 16-year monitoring period (Vuorenmaa, unpubl.) (Figure 9·23).

The western part of Finnish Lapland is less exposed to the sulfur load than the northeastern part, and the influence of sulfur emissions outside the Arctic is more detectable in western Lapland. This region lies in the ore-rich central

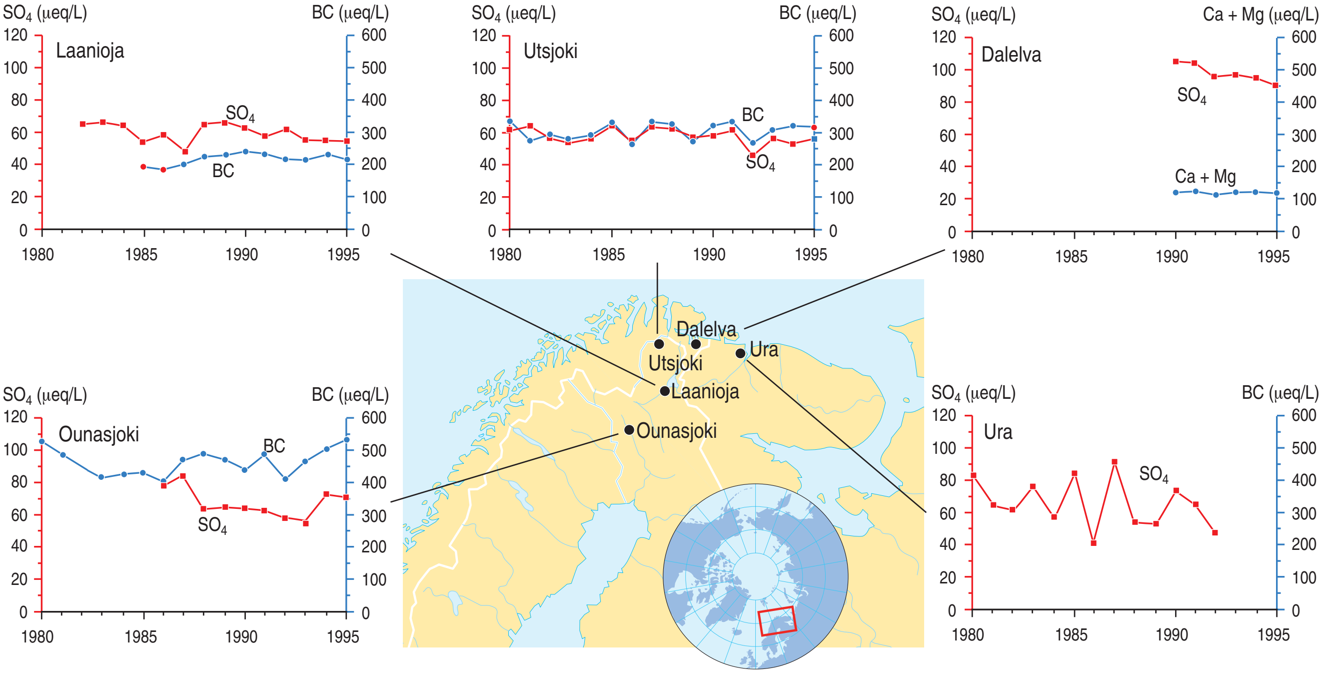

Figure 9·24. Annual average of sulfate and base cation concentrations as measured in rivers in northern Fennoscandia and the Kola Peninsula. Note: SO_4 (non-seasalt) estimated for river Utsjoki from total sulfur (data sources: Ounasjoki, Utsjoki and Laanioja – Finnish Environment Institute, unpubl.; Dalelva – Traaen pers. comm.; Ura – Moiseenko pers. comm.).

Lapland schist area, where some high regional sulfate anomalies in waters are caused by lithological sources, i.e., sulfide minerals in the soil and bedrock (Kähkönen 1996). However, the overall decline in sulfate emissions in Europe also seems to be reflected in a lowering of sulfate concentrations in the rivers of this area (Figure 9·24). In northeastern Lapland, the sulfate concentrations have remained more stable. However, a slight decline is seen in sulfate in, e.g., the Laanioja stream and the river Utsjoki (Figure 9·24), which is in good agreement with the observed reduction in sulfur deposition (wet + dry) in the Sør-Varanger area during the past few years (SFT 1994).

In northern Norway, river water acidification has been regularly monitored since the late-1980s. In eastern Finnmark, water chemistry has been measured in the river Dalelva and the Norwegian-Russian border rivers, the Pasvik and Jakobselva. Data from 1979/80 for the Pasvik are available for comparison. Despite the vicinity of the Ni-Cu smelters at Nikel, the buffering capacity of the River Pasvik has been such that the water chemistry has remained stable and there have been only very small changes in acidification parameters from 1979/80 to 1990/91. However, the maintenance of stable water chemistry throughout the year is attributable to the regulated flow of water from Lake Inari in Finland. In the unregulated, small river Jakobselva, the sulfate concentrations are higher and alkalinity is lower than in River Pasvik, and there is a marked depression of alkalinity in the spring. The remaining alkalinity (ca. 60 µeq/L) has still been sufficient to avoid acid water (Traaen *et al.* 1993). The river Dalelva clearly suffers from acidic pulses during spring flows. As annual values, sulfate concentrations in the river Dalelva have slightly declined and pH values have increased during the past few years (Figures 9·23 and 9·24).

The regular monitoring of river water chemistry in northern Sweden (Norrbotten county) started in 1969. The monitoring results for larger rivers demonstrate that sulfate concentrations started to decrease in the late-1970s. The buffering capacity of the rivers has remained largely unchanged since the 1970s (Bernes 1991). Recent monitoring results, e.g., from the River Råne, show that the alkalinity and base cation concentrations have remained stable and no decrease in the buffering capacity has been detected (Wilander pers. comm.). The pH value of the River Råne has declined since the 1960s. It has been suggested that the fall in pH values observed in northern Sweden between the hydrologically different decades, the 1970s (dry period) and 1980s (wet period), is primarily due to the slight increase in the concentration of organic matter (Bernes 1991).

Based on the analysis of river water samples collected from the large rivers of the Kola Peninsula during the period 1953-1992, a decrease in alkalinity occurred during both flood and winter periods (Moiseenko 1994a, 1994b). For the Kola region, the decreasing trend in alkalinity for the period 1953-1992 is significant. However, based on monitoring data available for, e.g., the River Ura, it seems that a slight decline in sulfate concentration, and a consequent rise in alkalinity has occurred during recent years (Figures 9·23 and 9·24).

9.8.2.2. Lakes

Regular monitoring throughout Lapland of small lakes sensitive to acidification was initiated after the Finnish lake survey conducted in 1987. The data from the lake monitoring network, however, do not show continuing acidification during recent years (Mannio and Vuorenmaa 1995).

Water samples collected from the Sør-Varanger region in 1986 demonstrated that small lakes in the area were markedly acidified (Traaen 1987). Sulfate concentrations in the lakes east of Kirkenes have more than doubled during the period from 1966 to 1986. Regular monitoring of lake water chemistry in the Sør-Varanger region started in 1987. Water analysis data from 1986 to 1990 indicate that acidification had stopped and stabilized at the level prevailing in 1986 (Traaen 1991). The water chemistry of the monitored lakes has remained relatively stable between 1986-1992, although, due to the hydrological conditions, the base cation and sulfate concentrations declined slightly in the Jarfjord region (Traaen *et al.* 1993). Recent monitoring results have demonstrated that sulfate concentrations have decreased significantly in lakes both east of Kirkenes (1.9 μeq/L/y) and in the Jordfjord region (4.4 μeq/L/y) during the period 1988/89 to 1994. As a response to reduced sulfate concentrations, the pH-values of the monitored lakes have correspondingly increased (Traaen 1994).

The Swedish lake surveys carried out during 1972-1990 have shown that the acidification status of the northern lakes has changed remarkably little since the early-1970s; neither pH nor alkalinity values have exhibited any long-term trends (Bernes 1991). In Norrbotten county, regular sampling of reference lakes, where liming has not been permitted, was initiated in 1985. Based on recent results, e.g., for Lake Vaimok, it appears that a slight rise in water alkalinity and a decline in sulfate concentration has occurred, especially since 1988/89 (Wilander pers. comm.). The observed decrease in base cation concentration is partly attributable to the reduced sulfate load. Changes in base cation concentrations, including large seasonal and inter-annual variations, are also caused by varying hydrological conditions, mainly the amount of precipitation (Wilander 1994).

9.8.2.3. **Groundwaters**

The water chemistry data of the national groundwater monitoring network (Finnish Environment Institute, unpubl.) for northern Finland are in good agreement with the observed state of acidification in surface waters. In the monitored groundwater basins, alkalinity has remained largely unchanged, and no signs of continuing acidification have been detected during 1979-1994. Even at Nellim, the groundwater station located in northeastern Lapland near to the Finnish-Russian border, there have been no appreciable changes in alkalinity, pH values and base cation concentrations during the 15-year monitoring period. The observed reduction in sulfur deposition during the last few years has not been reflected in a corresponding decrease in the sulfate concentrations.

The Norwegian Groundwater Monitoring Network (LGN) data (unpubl.) for stations north of the Arctic Circle show that the non-marine sulfate concentrations generally remained unchanged until the late-1980s, but have declined slightly since then. In stations located farther inland, the alkalinity has clearly remained equal to hardness, indicating that the groundwater in areas concerned has not been appreciably affected by strong acids. In coastal areas, on the other hand, the variation in alkalinity, H^+ and total Al concentrations in groundwater has been striking and there were sharp decreases in alkalinity and pH in the latter part of the 1980s and during the early-1990s. These variations were primarily accompanied by highly elevated Na^+ and Cl^- concentrations, a decrease in the Na:Cl ratio and non-marine Na^+ concentrations close to zero, indicating severe seasalt episodes.

The alkalinity of groundwater in inland areas of Norrbotten county in Sweden is roughly equal to hardness, which indicates that groundwaters have not been appreciably affected by strong acids (Bernes 1986, 1991). This state has prevailed over an extended time period. Long-term groundwater acidification has not been detected in northern Sweden (Andersson and Stokes 1988).

9.8.2.4. **Episodic acidic pulses**

Acid pulses are difficult to monitor as they are usually of short duration and occur in the smallest streams. In the Swedish Scandes (subarctic), acid pulses may be the reason for declining fauna in small streams (Ahlström *et al.* 1995). These streams all have a pH above 6 for the whole year, except for just a few days or weeks at the beginning of snowmelt.

The Norwegian rivers Jakobselva and Dalelva, in eastern Finnmark close to the Russian border, are subjected to acid pulses with expected damage to sensitive organisms (Traaen *et al.* 1993). In the latter river, automatic pH-monitoring started in 1988. During spring 1990, the pH drop was from 6.2 to 5.0 in just one day at the onset of the snowmelt. When the spring flood peaked 10 days later, pH had risen to 5.5 (Traaen *et al.* 1991).

Episodic acidification during the spring flood has also been studied in seven streams on the Kola Peninsula 1993-1994. Moiseenko *et al.* (1995) noted severe acid pulses in some streams and concluded that 'in the Arctic regions the maximum peak stress for the biota is created during the spring flood period'.

9.9. Spatial scales of terrestrial and aquatic acidification

9.9.1. Geographic extent of soil acidification

9.9.1.1. Regional sensitivity of soils to acidification

The impact of acidic deposition on the natural environment depends on two factors, the ability of the ecosystem to resist changes caused by loading of acidifying compounds, i.e., the inherent sensitivity, and the magnitude of deposition. The sensitivity normally refers to the susceptibility of an ecosystem to changes in structure and function. Sensitivity mapping has predominantly been carried out in North America (e.g., McFee 1980, Norton 1980, Environment Canada 1988), in Russia (Snakin and Meichenko, unpubl.), and in Europe (Kämäri 1986, Kämäri and Holmberg 1989, Kuylenstierna and Chadwick 1989, Kämäri *et al.* 1990).

For Canada, the national soil sensitivity calculations are based on a model employing different soil and bedrock characteristics, e.g., carbonate content and soil depth. The terrain has been classified using three sensitivity levels. A map prepared by Environment Canada shows that 43% of Canada's land area is sensitive to acidic deposition. These sensitive areas are mainly located on the Canadian Shield, south of the Arctic areas (Environment Canada 1988). However, large areas of the Northwest Territories, Yukon Territory, and Baffin Island are rated to have low potential of soils and bedrock to reduce the acidity of atmospheric deposition.

In Russia, the sensitivity of soils has been calculated from a range of factors, e.g., the amount of precipitation, the degree of humification, the radiation balance, seismicity, neotectonic activity, signs of prevalent tectonic movement, type of topography, the nature of the rock's structural ties, and freezing processes. Soil sensitivity is classified on the basis of these factors into seven groups, ranging from extremely low to extremely high susceptibility to acidic deposition. The extremely sensitive and sensitive soils are mainly situated in the northern parts of Russia (Snakin and Meichenko, unpubl.).

The most sensitive areas in the northern parts of the Kola Peninsula are situated in the northeastern tundra areas, where acidic rock components which are resistant to weathering predominate. The area around the Severonikel and Pechenganikel smelters is composed of basic and ultrabasic rocks which have a high buffering capacity against acid loads (Moiseenko 1991).

Recently, the sensitivity map by Kuylenstierna and Chadwick (1989) has been extended to a global map, prepared by the Stockholm Environment Institute, applying the methodology described by Kuylenstierna *et al.* (1995). An overview of the sensitivity of Arctic ecosystems is provided in a circumpolar map extracted from this global sensitivity map

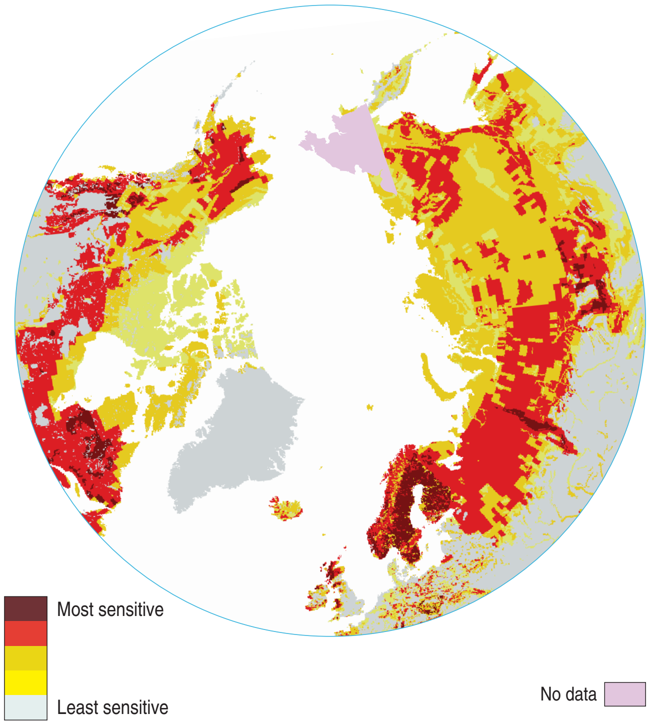

Figure 9·25. Sensitivity of Arctic ecosystems to acid deposition (data source: Stockholm Environment Institute, unpubl. preliminary data).

(Figure 9·25). The sensitivity distribution defined here combines three factors: soil type, land cover and a measure of soil moisture. Each data layer provides information concerning the buffering ability of the terrestrial ecosystem. The map indicates that most Arctic areas belong to those classes being the most sensitive areas to the effects of acidic deposition. The most sensitive areas are situated in Fennoscandia, in parts of Russia and in parts of Alaska. Much of the Canadian Arctic seems not to be sensitive to acidic deposition.

9.9.1.2. Evidence of soil acidification in the Arctic

With the exception of the immediate vicinity (<ca. 30 km) of the two Ni-Cu smelters on the Kola Peninsula, base saturation levels reported in the Arctic region are high, which is normal for upland forest soils and tundra soils consisting of overburden with varying base cation content. In four locations in the Chukotka region and Wrangel Island, more than 150 soil profiles were studied, representing the most characteristic ecotypes with varying geochemical settings. These shallow Arctic and tundra soils, with weakly developed humus horizons, had very high base saturation (BS) values, between 81and 96% for acid parent materials, and between 94 and 99% for hyperbasic rock types and limestone (Alexeeva-Popova *et al.* 1995).

In northern Fennoscandia and northwestern Russia, the base saturation values in the humus layer range from 15% to 80% (Tikkanen and Niemelä 1995). There is no clear spatial variation in the gradient that could be attributed to the corresponding sulfur deposition gradient (see Figure 9·22). However, there is a clear south-north decrease in sulfate concentrations in both the humus layer and winter snowpack, with locally elevated sulfate concentrations close to the Ni-Cu smelters at Nikel and Monchegorsk in northwestern Russia (Tikkanen 1995). This south-north decreasing gradient is a result of wet sulfur deposition originating from distant sources, such as Central Europe, southern Sweden and Finland, the St. Petersburg area, and northeastern Estonia. Although calculations suggest that the sulfur load in certain parts of northern Fennoscandia exceeds the critical load for forest soils (see Figure 9·27), actual sulfur concentrations in the humus layer in the region appear to be only slightly elevated. Humus sulfur concentrations to the west of Nikel and Monchegorsk are high up to a distance of about 30 km from the point emission sources, and are presumably partly attributable to the emission of sulfate-containing particulate material (Tikkanen and Niemelä 1995).

In the immediate vicinity of the smelters in northwestern Russia, very low base saturation values have been reported – ca. 15% (Tikkanen and Niemelä 1995). This loss of base cations has been attributed to displacement by the high load of protons or heavy metals (Cu and Ni) in local deposition, or a combination of both (Lukina and Nikonov 1995). Although sulfate concentrations in the snowpack close to the smelters are high, snow pH is well above 5 (Derome *et al.* 1992b). Elevated proton concentrations in the humus layer have not been found (Tikkanen 1995). No soil data are available from the regions close to the Norilsk smelter.

There is normally an inverse relationship between base saturation and exchangeable-Al levels. No abnormally high exchangeable-Al levels have been reported for soils in northern Fennoscandia and Chukotka, except for close to the Nikel and Monchegorsk smelters (Lukina and Nikonov 1995). This is in good agreement with the base saturation values.

In Alaska and the Canadian Arctic, there is no evidence of ongoing soil acidification (Barrie 1986).

9.9.2. Geographic extent of surface water acidification

9.9.2.1. Acidification of surface waters in northern Norway, Sweden, Finland and Russia

National lake surveys to assess the acidification status of surface waters have been carried out in Finland, Norway and Sweden between the years 1986 and 1990. In the fall of 1996, a new joint survey, based on random selection of lakes, was conducted in the entire Fennoscandia, Finland, Norway, and Sweden, as well as on the Kola Peninsula and parts of Karelia. The data from this survey will be reported during 1997. The databases available from large-scale surveys of this type are mainly designed to provide nation-wide or provincial assessments on particular problems. More detailed surveys can provide information on the geographic extent of the effects due to a point source, e.g., a smelter complex.

The data used for preparing maps showing the extent of surface water acidification in northern Norway, Sweden and Finland, and in northwestern Russia (Figure 9·26) were derived from the above-mentioned national lake surveys. The surveys were carried out in the whole of Finland in 1987 (Forsius *et al.* 1990), in northern Finland in 1988-1991 (Kähkönen 1992, 1996), in Norway in 1986 (Henriksen *et al.* 1988, 1994), in Sweden in 1990 (Bernes 1991), and in Russia in 1989-1992 (Moiseenko 1994a, 1994b).

The sulfate and nitrate concentration levels in Figure 9·26 reflect the loading of acidifying pollutants, and the base cation concentrations reflect the ability of the catchments to neutralize acidifying loading through weathering processes. The figure demonstrates that the geographic extent of high levels of sulfur pollution is limited to within a distance of some 50 km from the pollution sources. The nitrate concentrations are very low in the whole of northern Fennoscandia and Kola. Base cation levels are very low, especially in the mountain and coastal areas of Norway and Russia when the contribution of seasalts is subtracted.

Extensive surface water acidification has taken place, particularly on the Norwegian side of the Russian-Norwegian

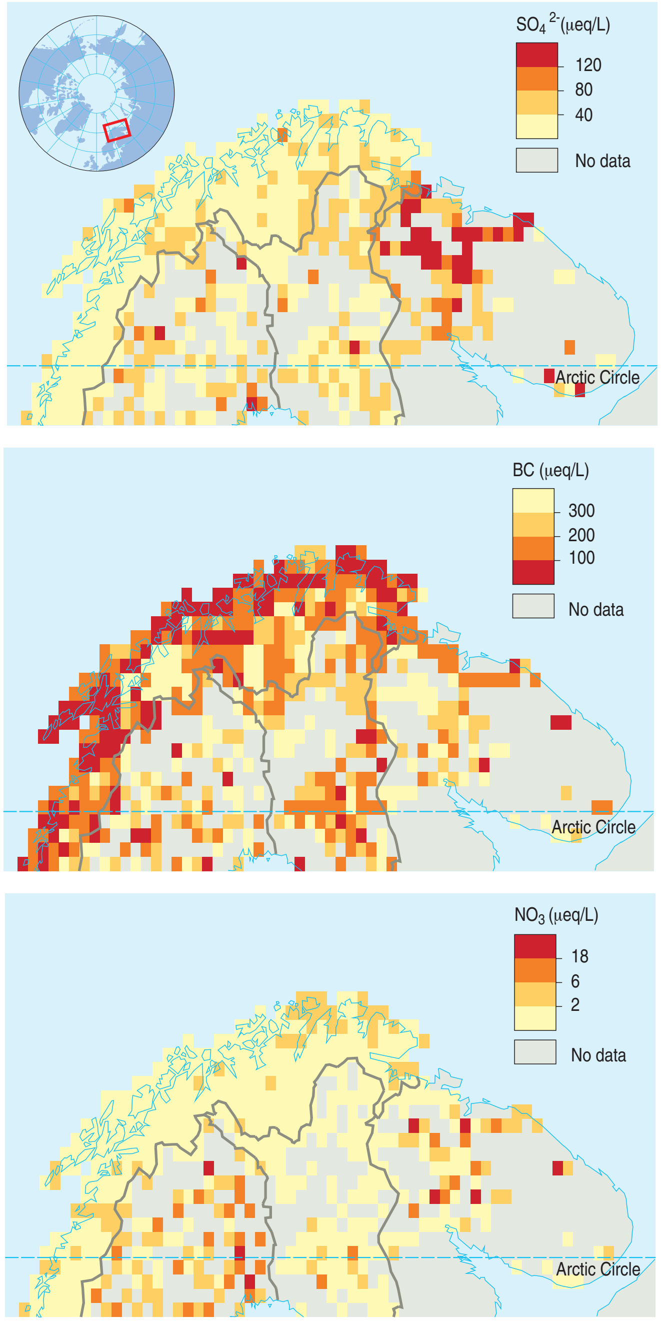

Figure 9·26. Median concentrations of sulfate, base cations, and nitrate in lakes of northern Fennoscandia and the Kola Peninsula (Finnish, Swedish and Norwegian data published in Henriksen *et al.* 1994; Kola data from Moiseenko, unpubl.)

border. Of the lakes examined in the Sør-Varanger region, 87% had an alkalinity of less than 50 µeq/L, and 60% less than 20 µeq/L. Lakes with zero alkalinity can be found in most parts of Sør-Varanger. Only 17% of the examined lakes (n = 30) in Sør-Varanger had a pH exceeding 6.5; 47% of the lakes had pH less than 6.0, of which 10% were lower than 5.0. The strongly acidified lakes are mainly found in the Jarfjord mountain area, east of Kirkenes (Traaen *et al.* 1991).

On the Kola Peninsula, 5.6% of the examined lakes (n = 370) had pH less than 5, and 13.5% less than 6 (Moiseenko 1994b). Acidified lakes occur throughout northern Kola, and there are strongly acidified small lakes with pH less than 5.0 located in the mountains within the Kuorpukas, Chuna, Volchii and Monche regions especially (Moiseenko 1994b). Sulfate values for lakes on the Kola Peninsula range from 50 to 250 µeq/L. Around the industrial centers, the sulfate concentrations in the lakes are greater than 200 µeq/L. The background sulfate concentrations in the Pechenga area are not more than 38 µeq/L (Gurevitch 1966). Northward from Pechenga, along the Barents Sea coast, where acidic granite-gneiss rocks are predominant, the condition of lakes can be characterized as critical, i.e., pH range is between 6.0 and 6.5, HCO_3^- to SO_4^{2-} ratio is less than 1, alkalinity is less than 50 µeq/L, and in some cases ANC is only 8 µeq/L (Traaen *et al.* 1991). A high sensitivity to water acidification was also noted in the northern and eastern tundra areas (Moiseenko 1994b). On the Russian side, the pollution of lakes by heavy metals (Ni and Cu) is considered to be a greater threat than acidification (Moiseenko *et al.* 1995).

The sulfate concentrations of clear-water lakes are elevated in the Inari region, northeastern Finland (median 60 µeq/L) and the buffering capacity of the waters is low (Kähkönen 1992, 1996). Sulfate is the main anion in the acid lakes. There are areas of Lapland where the lakes are very sensitive to acidification; 6% of the lakes examined in Lapland had an alkalinity less than 0 µeq/L and 20% of the lakes an alkalinity of between 0 and 50 µeq/L (Kähkönen 1992, 1996). Most of the lakes sensitive to acidification in Lapland are situated in northeasternmost Finland, adjacent to the Russian border, in the Enontekiö region of northwestern Finland, and in the areas of Ranua and Posio in southern Lapland.

9.9.2.2. Acidification of waters in Arctic Canada and Alaska

According to the studies conducted in northern Canada, no significant acidification has taken place, although many areas are highly sensitive to acidic precipitation (McNeely and Gummer 1984, Tibbats *et al.* 1987, Edwards *et al.* 1987, Jeffries *et al.* 1994, Jeffries pers. comm.). The paleolimnological data from some lakes with low pH indicated that the lakes have been unproductive throughout their existence, and that the natural process of gradual acidification is apparently continuing.

Data from the lakes and ponds in the Prudhoe Bay area in Arctic Alaska do not show any evidence of acidification (Snyder-Conn pers. comm.). There are also data for 1988 and 1989 from the ponds and small lakes of the Arctic National Wildlife Refuge. These data do not show any acidification, but the very low alkalinity of some refuge ponds does indicate poor buffering capacity and susceptibility to acidification (Snyder-Conn and Lubinski 1993).

9.9.3. Critical loads of acidity and their exceedance

Under the framework of the UN ECE, international legal instruments are developed to reduce the emissions of the most detrimental pollutants. The focus has initially been on reducing the effects of acid rain, through the control of sulfur emissions, but the activities have been widened to include nitrogen compounds, volatile organic compounds, photochemical oxidants, heavy metals and persistent organic pollutants. The signatories of the LRTAP Convention also cooperate on research into environmental effects of pollution.

The work organized under the LRTAP Convention includes the collection of new and updated data from national focal centers, using best estimates for regions with no supplied data and providing negotiating bodies and the scientific community with relevant critical load and exceedance maps. The critical load mapping for acidifying deposition is based on vulnerable ecosystems, comprised mostly of forest soils and surface waters, selected by national focal centers. In northern Europe, the simple steady state mass balance approach

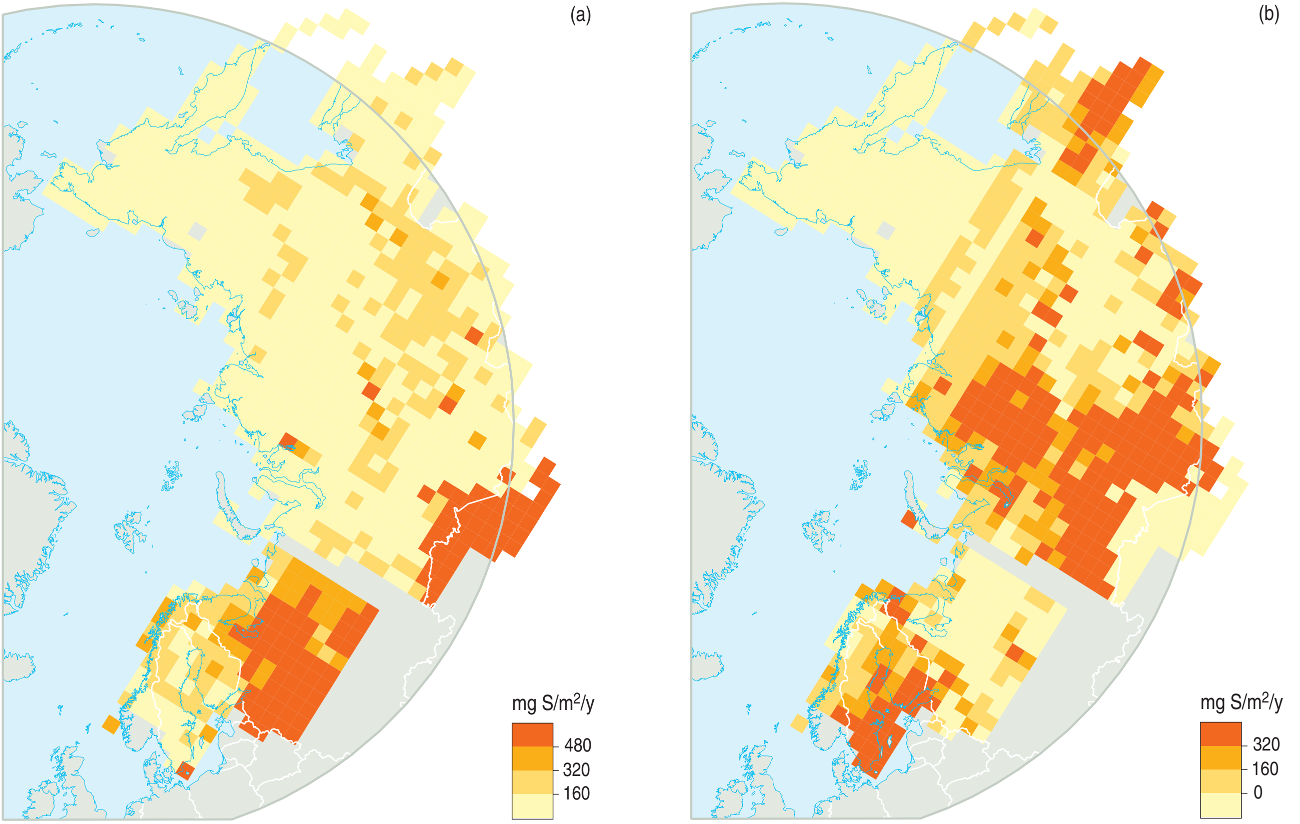

Figure 9·27. a) Critical deposition of sulfur, and b) its exceedance, in forest soils of northern Europe and Russia (5th percentile of the calculation points in each EMEP grid cell). The critical load values for sulfur have been derived from critical load of acidity assuming the acidifying nitrogen deposition of 1990. (Data for northern Europe originates from relevant National Focal Centers, compiled by the Coordination Center for Effects in the Netherlands. Data for the Asian part of Russia is by Bashkin *et al.* 1995).

has been used to estimate ecosystem sensitivities (Figure 9·27). The details of national data vary from country to country, and for emission reduction strategy assessments the target maps, following agreed statistical methods, are presented at the EMEP grid level, where continent-wide deposition model results are available.

The lake survey databases available in Finland, Norway, Sweden and the Kola Peninsula have been used for national critical load calculations and mapping, and for a common assessment of the three Nordic countries (Henriksen *et al.* 1990, 1994). In the northern areas, sulfur is presently the only acidifying agent, as the nitrate concentrations in lakes and streams are very low. Deposition estimates of sulfur in Finland were obtained from model calculations of the Finnish acidification model (Johansson *et al.* 1990) for the year 1990. For Norway and Sweden, land-use weighted sulfur deposition values, estimated by Lövblad *et al.* (1992), were used.

According to these calculations, there is large spatial variability in the critical load values, ranging from less than 300 to more than 1300 mg S/m^2/y (Henriksen *et al.* 1994). Most of the northeastern Finnish and Norwegian regions close to the large emission sources in the Kola region (especially the smelter at Nikel) are rather sensitive. Scattered areas with very low critical loads occur in the counties of Nordland, Troms and Finnmark in Norway and several areas in Sweden and Finland (Henriksen *et al.* 1994) (Figure 9·28a). Cal-

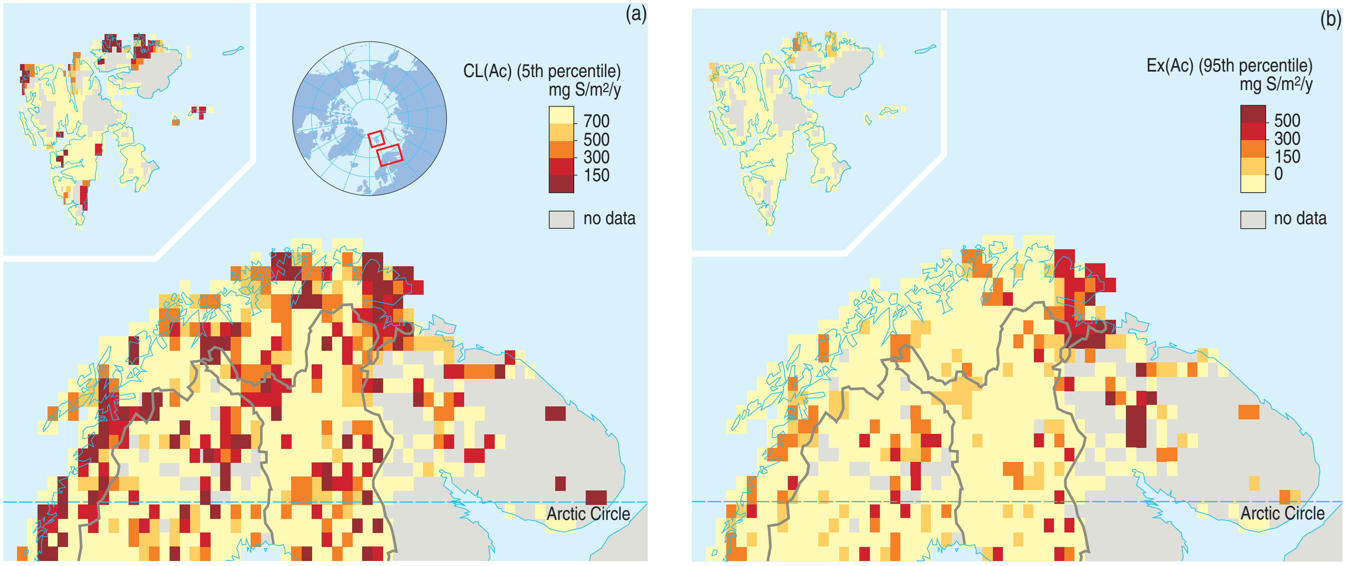

Figure 9·28. a) Critical loads of acidity expressed as sulfur (5th percentile presented for each grid) for lakes in northern Fennoscandia and surface waters on Svalbard, and b) their exceedance (95th percentile) (Henriksen *et al.* 1994, Lien *et al.* 1995).

culations made by Traaen *et al.* (1993) show that 90% of the area in Sør-Varanger has a critical load value less than 800 mg S/m^2/y.

The largest area where the critical load was exceeded is located in northern Finland and Norway due to the low critical load values and the influence of the Kola emissions. The highest exceedance values are in the range 300-800 mg S/m^2/y, and exceedance occurred in about 11% of the observations (Henriksen *et al.* 1994) (Figure 9·28b). In Sør-Varanger, the critical loads are exceeded in 70% of the area with the current level of sulfur deposition (Traaen *et al.* 1993). These values are low in comparison with those for the most affected areas in southern Norway and Sweden, where more than 50% of the lakes show an exceedance of more than 800 mg S/m^2/y (Henriksen *et al.* 1990).

The surface waters of northern Kola are highly sensitive to acid deposition. The results of Moiseenko (1994b) show that minimum values of critical load, CL < 300 mg S/m^2/y, occur in over 50% of the territory. The lowest median values are typical for the northern and eastern parts of the Kola Peninsula. The exceedance of critical load to surface waters occurred in 48% of the lakes on the Kola Peninsula.

Lien *et al.* (1995) have mapped the critical loads of acidity to surface waters on Svalbard, Norway. They found that 12% of the ice-free area of northern Svalbard has very low critical loads of acidity (<400 mg S/m^2/y), whereas 67% of Svalbard is well protected against acidic precipitation (critical load of acidity up to 1600 mg S/m^2/y) (Figure 9·28a). A minor exceedance of critical load was recorded in 5% of the ice-free area of Svalbard, and only in the northern parts (Figure 9·28b).

9.10. Effects of acidification on Arctic ecosystems

9.10.1. Effects on soils

9.10.1.1. Soil fertility

Soil fertility is primarily a function of the macronutrient (N, Ca, Mg, K, P) and trace element (Cu, Zn, B, etc.) concentrations. Soil moisture and other physical factors (e.g., grain size) also have an effect on soil fertility, but both the chemical and physical properties of the soil are usually inter-correlated to a varying degree, e.g., the more fertile the site, the better the water-holding properties and the higher the macronutrient and trace element contents. On peat soils, the limiting factor determining fertility, and associated tree growth, is usually the P and K levels, and on upland soil the N content. Acidifying deposition can both increase fertility in the short term (N deposition) and decrease it in the long term (loss of base cations, e.g., Ca and Mg, and the onset of nitrification). According to the analyses carried out on a wide range of peat, humus and mineral soil samples, there are no signs of a deterioration in soil fertility in northern Fennoscandia (Tikkanen and Niemelä 1995). The results of soil analyses are somewhat difficult to interpret owing to the high spatial and temporal variation in soil chemical properties. However, the above conclusion is supported by the results of needle chemistry surveys, which are less susceptible to spatial variation and, furthermore, reflect actual nutrient uptake by trees. The variation in needle chemistry rather closely follows that of the overburden, i.e., geochemical variation.

A severe decrease in soil fertility has been reported close to the smelters at Monchegorsk and Nikel, and this has undoubtedly contributed to tree death in the area. Soil and needle Ca, Mg, K, Zn and Mn concentrations are extremely low (Raitio 1992, Tikkanen 1995). The main cause of this loss of macronutrients and trace elements (with the exception of Cu, of course) appears to be displacement of plant nutrients from the exchange sites as a result of the very high Cu and Ni deposition levels. No signs of soil acidification (e.g., low pH) have been reported (Lukina and Nikonov 1995).

9.10.1.2. Soil microbiology and decomposition

Only a few isolated studies have been carried out on soil microbiology and decomposition in northern Fennoscandia. Marfenina and Grigoryev (1992) have studied the soil microfungi near the Pechenga Ni-Cu smelter on the Kola Peninsula. Closest to the plant (within 1.5 km), there was a decline in species richness and diversity compared to the control site (60 km). In stressed conditions, the composition of microfungal species of fungal soil communities and epiphytic communities become more simplified. The species most resistant to acid deposition is *Penicillium spinulosum*, and the most sensitive are dark-colored species.

Most of the effects of air pollution on soil microfungi on the Kola Peninsula appear to have been caused by heavy metal pollution (Lebedeva 1993). However, it is difficult to separate the effects of heavy metals on the soil microfungi from the effects of acidification. Contamination of the soil by sulfur compounds, together with heavy metals, has had a dramatic effect on the area around the smelters where the humus layer has disappeared (Chertov *et al.* 1993).

9.10.2. Effects on vegetation

9.10.2.1. Effects on forests

Sulfur dioxide causes damage to foliar tissues, affects the metabolism of trees, and accelerates their decline and eventual death (Bäck *et al.* 1992, Huttunen 1994). Episodes of high sulfur dioxide concentrations cause direct injury to growing plant tissues, resulting in chlorosis or necrosis of exposed foliage (Aamlid 1993b). In areas where the average concentration in the air is normally low, gaseous air pollutants can constitute a chronic stress on the vegetation, predisposing trees and other plants to a range of injuries of climatic origin (Aamlid and Venn 1992, Sutinen 1993). The deposition of air pollutants, especially sulfate, can increase the leaching of essential nutrients (base cations) from the foliage and soil, resulting in visible deficiency symptoms and reduced growth. The degradation of soils through acidification and the mobilization of toxic aluminum ions can occur. Continued deposition of heavy metals can cause problems for nutrient uptake by the roots, possibly by interfering with the mycorrhizal associations (Aamlid 1993b).

The critical sulfur dioxide levels in air for forest ecosystems and natural vegetation at low temperatures are 15 μg SO$_2$/m^3 (annual mean and half-year mean) (UN ECE 1996) and 150 μg SO$_2$/m^3 (1-hour mean) (UN ECE 1992). Measurements and calculations have shown that both the long-term and short-term critical levels are exceeded over large areas on the Kola Peninsula and also on the Norwegian side of the border. At Viksjøfjell in Norway, hourly mean concentrations of above 3000 μg/m^3 have been measured. According to calculations, the concentrations can exceed 1000 μg/m^3 (1-hour) at distances of 50 km from Nikel under certain meteorological conditions (Jerre 1994).

Permanent ice or tundra vegetation covers vast areas of the northern Arctic, and forests only grow in the southern regions. In Arctic Canada, Alaska and Greenland, the vegetation in the ice-free areas is mostly tundra consisting of lichens, mosses, shrubs and small bushes. As a consequence of the warming effect of the Atlantic Gulf Stream, the cli-

Table 9·4. Zones of vegetation degradation around Monchegorsk and Nikel according to Tikkanen and Niemelä (1995) (see Figure 9·30). Damage zone classifications in parenthesis by Tømmervik *et al.* (1995) (see Table 9·5).

Name of the zone	Vegetation	Annual average estimated atmospheric SO$_2$ concentration, µg/m^3
Forest death area (1. Total damage)	Dead	> 40
Inner visible damage zone (2. Severe damage)	No epiphytic lichens Marked defoliation in conifers	15-40
Outer visible damage zone (3. Intermediate damage)	Changes in species composition of lichens and dwarf shrubs External damage on the needles of pine *(Pinus sylvestris)*, birch *(Betula bubescens)* and aspen *(Populus tremula)*	8-15
Inner non-visible damage zone (4. Moderate damage)	Chlorosis of the stomata of pine needles Changes of species composition of lichens	4-8
Outer non-visible damage zone (5. Minor damage)	Changes in the microscopic structure of pine needles and epiphytic lichens	2-4

Table 9·5. The studied area (12 703 km^2) near the border between Russia and Norway divided into damage zones of varying severity (Tømmervik *et al.* 1995).

Zone description	Area, km^2	%
1. Total damage	415	3.3
2. Severe damage	343	2.7
3. Intermediate damage	1995	15.7
4. Moderate damage	1887	14.6
5. Minor damage	770	6.1
6. Slight/no damage	7293	57.4

mate in northern Europe is moderate. Therefore, most of the coniferous forests found north of the Arctic Circle are growing in western Russia, Finland and Norway.

The total tundra area damaged by anthropogenic factors (a region with a relatively low coverage of forest), extending from the Kola Peninsula to Chukotka, is about 470 000-500 000 km^2 (Kryuchkov 1990). Most of the problems occurring on the Kola Peninsula and near Norilsk in western Siberia are not caused by acidifying pollutants alone. They are frequently associated with heavy metals, other toxic substances, and with land disturbance. Excessive forest felling is also seen as one of the main reasons for environmental problems in the taiga zone.

There are zones of increasing pollution and ecosystem degradation around the industrial areas in Norilsk, Nikel and Monchegorsk (Vlasova *et al.* 1991, Kryuchkov 1991, Aamlid 1992b, 1992c, Mikkola 1996, Gytarsky *et al.* 1995, Tømmervik *et al.* 1995) (Tables 9·4 and 9·5; Figures 9·29 and 9·30).

The area of total vegetation damage around Norilsk, caused by sulfur and heavy metal emissions from the Cu-Ni smelters at Norilsk, extends about 80 km south of the city, and covers an area of ca. 5000 km^2 (Vlasova *et al.* 1991) (Figure 9·29). According to a report by the Taimyr Ecology and Natural Resources Committee, ‘perished woods stretch over 120 km south, 180 km southeast and 90 km east of the

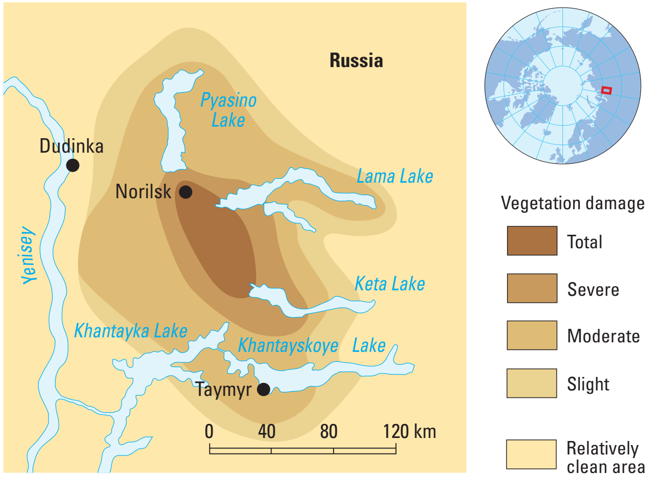

Figure 9·29. Vegetation damage zones around Norilsk (modified from Vlasova *et al.* 1991).

Figure 9·30. Approximate forest damage zones in the vicinity of Monchegorsk and Nikel and the visible-damage and non-visible-damage zones on the Kola Peninsula and Finnish Lapland (Tikkanen and Niemelä 1995). The inset shows coverage (%) of lichens (*Hypogymnia physodes* and *Parmelia olivacea*) on birch trunks in the Norwegian-Russian border area (Aamlid 1992b).

city of Norilsk' (Ministry for Environment Protection and Natural Resources of the Russian Federation 1994), comprising an area of about 15 000 km^2.

In the Nikel-Pechenga and Varanger regions, the size of the forest-death area is greater than 400 km^2 (Tømmervik *et al.* 1995). The area affected by air pollution in these regions increased from approximately 400 km^2 in 1973 to more than 5000 km^2 in 1988 (Tømmervik *et al.* 1992). In terms of distance from the emission source, the affected area extended by about 5 km per year in the period 1973-1979, 4 km per year between 1979 and 1985, and 1 km per year from 1985 to 1988 (Tømmervik *et al.* 1995, Kalabin 1991). The prevailing winds carry most of the pollutants from the Nikel and Zapolyarnyy smelters to the northeast and east. Therefore, only the outer visible-damage zone around Nikel extends into the eastern parts of Inari in Finland (Tikkanen and Niemelä 1995) (Figure 9·30).

At Monchegorsk, on the Kola Peninsula, the forest-death area covers an area of 400-500 km^2 and extends more than 10 km to the south and more than 15 km to the north of the smelter complex (Mikkola 1996) (Figure 9·30). Forest death in the Monchegorsk region is estimated to be advancing at a rate of ca. 0.5 km per year (Mikkola 1996).

The main problem in the Norwegian-Russian border area seems to be the high concentrations of sulfur compounds in the air together with the impact of heavy metals (nickel and copper). High, long-term average concentrations of these pollutants are mainly observed on the Russian side of the border. On the Norwegian side, the long-term average ambient concentrations are rather low, and the main problem seems to be the episodes with high SO_2 concentrations that occur under specific meteorological conditions (Jerre 1994). On several occasions, damage to vegetation and trees has been observed in the Norwegian forests close to the Russian border. The damage has mainly been identified as partially brown leaves and needles (Aamlid 1992c, 1993b). The most severe vegetation damage on the Norwegian side of the border is in the vicinity of the Jarfjord Mountain (Tømmervik *et al.* 1989, 1991).

The critical levels of sulfur dioxide are exceeded for sensitive ecosystem compartments on more than 3200 km^2 of Russian and Norwegian territories (Aamlid *et al.* 1995). A considerably larger area is probably influenced by air pollution, because elevated pollutant levels can be traced by chemical analysis of plants and soils sampled far outside the affected area.

Only a small area in eastern Inari, northeastern Finland, is under a direct and immediate threat of pollution damage. Aside from this area, the emissions from the Kola Peninsula are not expected to cause significant damage to forest ecosystems in Finland in the near future (Tikkanen and Niemelä 1995).

9.10.2.2. Effects on lichens and shrubs

The main vegetation types in the northern areas of northern Norway and the Kola Peninsula consist of lichen and dwarf shrub communities. The area with lichen-dominated vegetation (heaths and forests) around the Cu-Ni smelters in Nikel and Zapolyarnyy decreased from ca. 2700 km^2, to ca. 700 km^2 in 1988, with a subsequent increase to ca. 1050 km^2 in 1994. This is correlated with an increase in emissions of SO_2 and other compounds from 1973 to 1988 and a subsequent decrease of emissions from 1988 to 1994 from these smelters (Høgda *et al.* 1995, Tømmervik *et al.* 1995). The critical SO_2 levels for lichen-dominated vegetation types and for epiphytic lichens in cold climates (annual average <10 µg SO_2/m^3 (UN ECE 1996)) have been exceeded in the eastern parts of the Varanger and Nikel-Pechenga areas (Tømmervik *et al.* 1995). The epiphytic lichens (*Hypogymnia physodes*, *Parmelia olivacea*) on birch stems are absent in rather large areas in the eastern part of the Norwegian forests close to the Russian border and there is decreased coverage over even larger areas (Aamlid 1992b) (Figure 9·30).

Studies of morphological parameters in epiphytic lichens show that several parameters are relevant in terrestrial monitoring. A morphological analysis of *Hypogymnia physodes* and *Melanelia olivacea* from three sampling sites in Sør-Varanger, exposed to high concentrations of sulfur dioxide, shows morphological differences (growth form, fertility, apothecial morphology, discoloration) compared to lichens sampled from an unpolluted control site (Hilmo and Larsen 1994). Samples of *Parmelia sulcata* were taken at an industrial site in Glomfjord and from an unpolluted control area to study morphological effects of nitrogen pollution. Morphological differences were detected (discoloration, growth form), and substantially higher concentrations of chlorophyll were found in samples from sites with high nitrogen load than in samples from the control area (Hilmo and Larsen 1994).

Air pollution from the Kola Peninsula has affected the growth rates and weight-to-length ratio of reindeer lichens (*Cladina* spp.). Growth rates reached their normal level at a distance of 50-60 km from the smelters (Helle and Kojola 1992). Reindeer lichens have completely disappeared from around two Ni-Cu smelter complexes on the Kola Peninsula (Kalabin 1991, Tømmervik *et al.* 1991). This is an extremely important matter for reindeer management because the carrying capacity of the most important ranges is primarily determined by the growth rate of reindeer lichens (Helle and Kojola 1992).

In areas with high SO_2 and heavy metal deposition, where the tree cover is completely destroyed and the ground vegetation severely damaged, the field vegetation layer is the only living vegetation component. The field vegetation is important in inhibiting soil erosion processes that would otherwise lead to the complete degradation of the forest ecosystems. The proportion of *Vaccinium myrtillus* (L.) has decreased near the emission sources, but that of *Empetrum hermaphroditum* (*Lge*) Hagerup has increased (Bakkal 1992, Tømmervik *et al.* 1995). The bilberry (*Vaccinium myrtillus*)-dominated vegetation has, however, increased in a sector within 5 to 30 km from the smelters, mainly due to the transformation of lichen-dominated vegetation into a bilberry and *Deschampsia flexuosa*-dominated formation (Deyeva and Maznaya 1993, Tømmervik *et al.* 1995). The proportion of other species in the field layer has not changed significantly. Moreover, some species of mosses increase markedly if the SO_2 concentration in the air is high (Andreyeva 1990).

9.10.3. Effects on freshwater ecosystems

Negative effects on biota usually first appear in small streams and in the littoral zones of lakes; larger water bodies have a naturally higher capacity to buffer an acid inflow. Extensive disappearance of fish can occur in streams before the effects of acidification are detected in lakes (Bergquist 1991). It is important to distinguish between these phases of acidification. The first phase, characterized by occasional acid flushes in streams in spring during the snowmelt, is hard to detect with routine water sampling, because the decrease in pH occurs for a period lasting only for a few days or weeks (McComas *et al.* 1990, Moiseenko 1994b). The second phase has started when small lakes also contain acidic water during periods of high discharge. While the water quality might improve during summer, several small lakes in base-poor catchments remain acidic throughout the year. The final phase occurs when several larger lakes have acidic conditions.

9.10.3.1. **Arctic freshwater biota**

Tundra waters may be open for a few weeks, while some High Arctic lakes are permanently covered with ice. Low temperatures and a large-amplitude photocycle are characteristic of Arctic waters. Most polar lakes and streams contain life which is adapted to long, dark winters with ice cover, and short summers. Although extreme, the aquatic environment may be regarded as less harsh than the terrestrial habitat. In the water, the organisms are protected from extreme temperatures, low humidity, strong winds and erosion (Hobbie 1984, Hammar 1989).

Most of the taxa of phytoplankton, zooplankton, invertebrates and fish that exist in the Arctic are also found in subarctic and even in temperate waters. Nevertheless, the number of species in the Arctic is low and the production is often very small (Hobbie 1984, Hammar 1989). Arctic food chains are generally short due to the low natural production and the harsh environment.

There are gradual differences in freshwater biota as one moves north (Hobbie 1984, Hammar 1989). Phytoplankton production is normally small and often surpassed by the production of attached algae and higher plants. The phytoplankton are dominated by small taxa, often flagellates. Small size and motility allow the organisms to choose the most favorable light level under the ice cover. Vascular plants are surprisingly common in the Low Arctic, although the flora is greatly reduced compared to that of the subarctic.

Zooplankton may be lacking in some ultra-oligotrophic lakes, due to lack of food. Normally in smaller lakes, they over-winter as eggs, and quickly develop as the temperature rises. In deeper waters, zooplankton can pass the winter as free-swimming forms.

The dominant benthic invertebrates of the Arctic are the chironomid larvae. In shallow ponds, the larvae may be completely frozen from October until June, but when conditions improve emergence takes place, if necessary through holes in the ice cover. As for plankton, smaller forms of invertebrates dominate in the Arctic. In several lakes, even chironomids are lacking and small taxa of Protozoa, Rotifera, Tardigrada and Nematoda dominate. In the southern parts of the Arctic, the invertebrate fauna is comparatively rich, dominated by the same groups as in northern temperate regions. Above the forest line there is normally a drastic decline in the number of freshwater taxa, primarily due to harsh climatic conditions, loss of nutrients and loss of habitat (Huru 1992, Nauwerck 1994).

Fish are present in most Arctic waters deeper than 3-4 m (McPhail and Lindsay 1970). Under severe climatic conditions, fish have more fat reserves by winter and slower growth than in lower latitudes. Maturation is also slower and reproduction is not undertaken every season as it takes a longer time to build up energy reserves (Moiseenko and Yakovlev 1990a). Arctic fish tend to have an extremely long life span; 25-40 years have been recorded for Arctic char (*Salvelinus alpinus*) and Lake trout (*Salvelinus namaycush*) (Hobbie 1984, Hammar 1989).

Most authors describe Arctic fauna as sensitive or vulnerable to anthropogenic alterations (e.g., Hammar 1989). Several species live under constant environmental stress and are therefore more susceptible to additional stress. High mountain lakes and Arctic lakes are thought to be particularly sensitive to environmental changes, due to extreme climatic conditions, slow chemical processes, and the fact that they are simple and labile ecosystems (Muniz and Wallö 1990, Rosseland 1990).

9.10.3.2. **Effects on decomposition**

In the extremely oligotrophic Arctic freshwater systems, the release of nutrients through decomposition of higher plant material is essential. It has been shown in temperate regions that acidification causes oligotrophication of waters due to several processes (Nyman 1990, Wilander *et al.* 1995). McKinley and Vestal (1982) experimentally studied the microbial decomposition of *Carex* leaves in microcosms with water from an ultra-oligotrophic Alaskan Arctic lake. They found that reduction of the pH below 5 caused a 36% reduction of the microbial activity as compared to the ambient control (pH 7.4).

9.10.3.3. **Effects on plankton**

Only a few studies of acidification effects on plankton have been performed. In the Kola Peninsula region, there has been a decline in the diversity of plankton species resulting from the disappearance of sensitive species. In small acidified lakes, the number of species and the abundance of phyto- and zooplankton are as low as in lakes contaminated by heavy metals (Yakovlev 1992). A low abundance of acid-sensitive daphnids (Cladocera) was also reported in acidified lakes in the Jarfjord area (Norway) (Nøst *et al.* 1991).

Outside the above-mentioned regions, no reported signs of acidification effects on plankton exist in the Arctic. Nauwerck (1994) has reported the results of extensive surveys of water chemistry, phytoplankton and zooplankton in Abisko, northern Sweden (68°N). He found a low number of plankton species in lakes with a pH range of 5.1-5.8. He does not explain the low pH in the lakes, so it is not known if acidification is the cause.

9.10.3.4. **Effects on periphyton and higher vegetation**

Aquatic vegetation has not been studied in the Arctic in relation to acidification. In Norway, a nationwide interview survey was carried out in 1992 to identify recent developments of periphyton (attached) algae. Out of the 592 questionnaires, 57% reported increased periphyton growth, mainly in remote and high altitude areas (Lindström 1995). In the Swedish high mountains (subarctic), such observations have also been frequent (Degerman *et al.* 1992). Lindström (1995) suggests that increased nitrogen deposition could be the reason.

9.10.3.5. **Effects on invertebrates**

A likely sign of the effects of acidification on bottom fauna is evident in the streams of Sør-Varanger and the Varanger Peninsula (Norway) (Huru 1992). In acidic streams, the number of species was low and the sensitive species were missing. In the rivers farther away from the Cu-Ni smelters in the northern part of the Kola Peninsula, e.g., in the Lakselv catchment (Huru 1992), no effect of acidification on the bottom fauna was noted. Nøst *et al.* (1991) have also reported a low abundance of the acid-sensitive mayflies (Ephemeroptera) in acidified lakes in the Jarfjord area (Norway). Moreover, in the Dalelva catchment (Norway), damage to bottom fauna has occurred, which might be connected with the low pH and high concentrations of labile aluminum (Bækken and Aanes 1990). Both studies above are from more or less the same geographical area (eastern Finnmark).

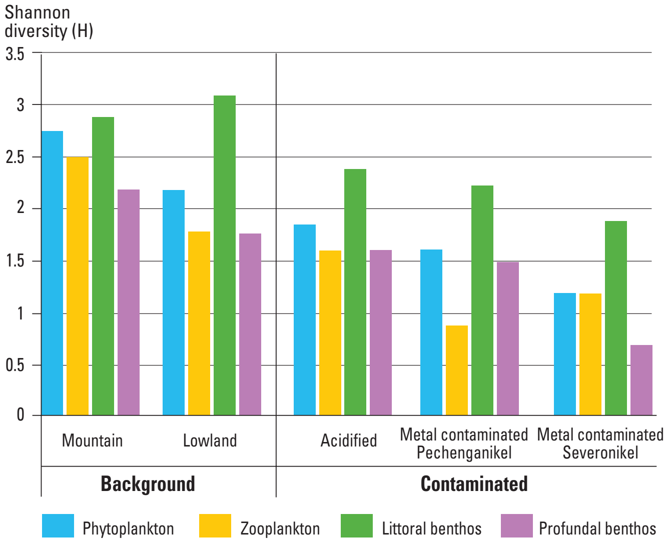

Figure 9·31. Diversity indices (Shannon-Weaver means) for phytoplankton, zooplankton and zoobenthos in lakes and streams in the Murmansk region (Yakovlev 1992).

Yakovlev (1992) presented data for lakes and streams in the Murmansk region. There was a decline in species diversity as a result of the loss of sensitive species. The abundance of zoobenthos in small acidified lakes was as low as that in the lakes contaminated by heavy metals (Figure 9·31). The acidity has the most prominent effect on invertebrates in remote lakes situated on base poor bedrock (Yakovlev 1995).

Field experiments with the freshwater pearl mussel (*Margaritifera margaritifera*) in the River Pana (Murmansk region) showed that this species can withstand short periods (hours) of pH values down to 4 (Moiseenko and Yakovlev 1990b). The status of freshwater pearl mussel populations in this river has remained unchanged since 1936 at which time this region was not directly contaminated by heavy metals.

In northeastern Finland (67-68°N), there were no indications of acidification in the benthos of streams and rivers; many acid-sensitive species, e.g., the mayfly *Baetis lapponicus* and the caddis fly *Arctopsyche ladogensis* (Engblom and Ligdell 1983), were still present throughout the area (Erkinaro *et al.* 1992). Accordingly, Hämäläinen and Huttunen (1994) found only little, if any, signs of acidification effects on biota in streams of northeastern Finnish Lapland, as judged by the distribution of invertebrate taxa. Among other acid-sensitive species, the mayfly *Baetis rhodani* was found in all streams except one. However, the stream minimum pH inferred from macroinvertebrate assemblages by a mathematical model decreased from west to east in parallel with the increasing modeled sulfur deposition (Tuovinen *et al.* 1994). Consequently, it was suggested that the acid load from the Kola emissions might have some effect on the biological state of the streams in the eastern border of Finnish Lapland.

In the area around the Cu smelter at Sulitjelma in Norway, the most severe negative effects on stream benthos were evident within a distance of 30 km to the east of the smelter (Degerman *et al.* 1992). Although the pH of the streams was low, the sulfur and metal concentrations were not determined. It is therefore not possible to distinguish the effects of metal contamination from those of acidification, nor to exclude natural acidity. However, because the waters had a low humus content and the low pH area was situated downwind from the smelter, anthropogenic acidification was considered to be probable. The smelter was closed down in 1987. It is possible that there has been a subsequent recovery of the benthos similar to that observed near the Cu-Ni smelter in Sudbury, Canada (46°N) (Keller *et al.* 1986, Keller and Yan 1991, McNicol *et al.* 1995). The Sulitjelma area should be a focus for further investigations as it could show the recovery of the fauna after a smelter shut-down in the Arctic region.

9.10.3.6. Effects on fish

The rivers of the northern part of the Kola Peninsula and Sør-Varanger are of great importance for salmonid reproduction (Moiseenko and Yakovlev 1990b). Salmonids are most sensitive to low pH during hatching, as fry and later on in the lifecycle as smolts. In the river Tenojoki, the pH during the snowmelt in spring 1988 ranged from 4.7 to 6.3 (mean 5.5) (McComas *et al.* 1990). These values are clearly detrimental to both brown trout (*Salmo trutta*) and salmon (*Salmo salar*), especially if combined with elevated concentrations of labile metal ions. Although McComas *et al.* (1990) did not take any biological samples, they suggested that the timing of the low pH was fortunate, since the salmon eggs were not ready to hatch at the time when water quality was at its worst.

Traaen *et al.* (1991, 1993) found several small lakes in the Sør-Varanger area (Norway) with low pH and labile aluminum concentrations considered harmful to fish. This was especially pronounced in the Dalelva catchment, where damage to bottom fauna has also been reported (Bækken and Aanes 1990). The labile aluminum concentrations during spring flood were 30-50 µg/L (Traaen *et al.* 1993).

No effects of acidification were detected on the populations of Arctic char in 20 small lakes in eastern Finnmark (Hesthagen *et al.* 1992). The authors suggested that the lake-spawning Arctic char is exposed to better water quality than the brown trout, which spawns in streams. A similar finding has been reported by Nøst *et al.* (1991) for the lake Limgarnbergtjern in the Jarfjord area. The latter study found unaffected fish recruitment in several lakes investigated in the Murmansk area. This was explained by the base-rich geology. The high acid-neutralizing capacity of the bedrock close to the metallurgical industries has been pointed out by several authors as the reason why relatively few waters are acidified in the proximity, whereas granite/gneiss bedrock in more remote areas provides less protection for the fauna (Traaen *et al.* 1991, Nøst *et al.* 1992). Hesthagen *et al.* (1995) conclude that in northern Norway, fish populations damaged by acidification are only common in the Jarfjord area.

Erkinaro *et al.* (1992) electrofished in rivers and streams farther south in Finland, in the area immediately to the east, west and south of Lake Inari (68° N). They found reproduction of brown trout in the whole area, and also the occurrence of acid-sensitive species such as burbot (*Lota lota*) and European minnow (*Phoxinus phoxinus*). Together with observations of acid-sensitive benthos (see above), they concluded that ‘no symptoms of acid-induced reductions could be seen’. Lappalainen *et al.* (1995) electrofished acid-sensitive streams and lakes in northeastern Finnish Lapland, but did not find any signs of damage among the fish populations. The absence of minnows in some small waters might be the first sign of acid-induced fish population damage in the area.

The status of freshwater fish in 1900 lakes throughout the county of Nordland, northern Norway, was recorded by interviews of local fishermen. The data were compared with earlier information on water quality (Halleraker and Hesthagen 1995). Less than 10% of the salmonid populations were recorded as reduced or extinct. The major causes were overexploitation and effects of damming for hydroelectric power. In some areas, acidification could be imminent, in spite of low deposition. It was suggested that sea spray deposition could be an important factor.

Thus, outside of the Kola Peninsula and the neighboring areas in Finland and Norway, there are no reports of acidification affecting fish populations in the Arctic.

9.10.4. Effects on birds and mammals

Birds are affected both indirectly through changes in food availability brought about by acidification (Eriksson 1991, Gilyazov 1993) and directly through increased exposure to metals and lowered uptake of calcium. Gilyazov (1993) reports that terrestrial birds close (less than 40 km) to the smelters on the Kola Peninsula were negatively affected. Bird density, especially birds of prey, decreased during 1977-1990. It was not possible to distinguish effects of acidification from the effects of heavy metals.

There are no studies reported from the Arctic on the effects of acidifying pollutants on mammals.

9.11. Scenarios for future acidification

9.11.1. Available geochemical models

Steady-state and dynamic models have been developed to predict the acidification of soils, lakes, streams and groundwater (e.g., Cosby *et al.* 1985, 1986, De Vries *et al.* 1989, 1993). While the former are used to estimate the steady state of a system for a given load by neglecting time-dependent processes and finite pools, dynamic models are used to predict the gradual chemical response of a receptor to a changing deposition level by including various buffer and adsorption/desorption mechanisms. There are several widely used and tested models available for analyzing long-term acidification trends and the effects of acidification on soil and surface water quality. Applications of two models on two sites are available for the Arctic region near the Kola smelters.

The SMART model (Simulation Model for Acidification's Regional Trends; De Vries *et al.* 1989) was developed to estimate long-term chemical changes in soil and soil water in response to changes in atmospheric deposition. The model is designed for applications on a regional scale, including entire catchments and soil regions. Its output includes base saturation and the concentrations of the major anions and cations in soil solution and runoff water. SMART consists of a set of mass balance equations, which describe the soil input-output relationships (weathering, adsorption, leaching) for the cations (Ca, Mg, Al) and strong acid anions (SO_4, NO_3, Cl), and a set of equilibrium equations (cation exchange, Al dissolution), which describe the soil processes. The soil model, originally described by De Vries *et al.* (1989), has recently been enhanced by the inclusion of simple descriptions of i) sulfate adsorption/desorption, ii) dissociation of organic anions, iii) denitrification, iv) N immobilization (Posch *et al.* 1993b, De Vries *et al.* 1994), and v) lake water processes (Kämäri *et al.* 1995).

The MAGIC model (Model for Acidification of Groundwater in Catchments) is an intermediate-complexity process-oriented model for constructing acidification history and predicting future acidification over time periods of decades to centuries (Cosby *et al.* 1985). Like SMART, MAGIC makes use of aggregated parameters on a catchment scale and focuses on chemical changes in the soil caused by atmospheric deposition, vegetation and leaching. The processes in MAGIC include atmospheric deposition, sulfate adsorption, cation exchange, CO_2 dissolution, precipitation and dissolution of aluminum, chemical weathering, uptake and release of cations by vegetation, and export in runoff.

Both models have been extensively used for a variety of applications in North America and Europe. Application of MAGIC to whole-catchment studies shows that the model is able to predict the response of water and soil acidity to large and rapid changes in acid deposition (Wright *et al.* 1990). These results reinforce other evaluations of MAGIC, such as comparison with the paleolimnological reconstruction of lake acidification (Jenkins *et al.* 1990, Neal *et al.* 1988) and changes in regional lake chemistry in southern Norway (Wright *et al.* 1991). The credibility of SMART has increased following its application to historical soil data (Posch *et al.* 1989) and comparison with other soil/water models (Wright *et al.* 1991, Warfvinge *et al.* 1992).

9.11.2. Potential development of Arctic acidification

Two calibrated applications of acidification models are available for the Arctic region. MAGIC has been used to predict the responses of the river Dalelva in northeastern Norway (Wright and Traaen 1992), and SMART has been applied to data on the Christmas lakes in northeastern Finland (Kämäri *et al.* 1995). The time-step used in these applications is one year. The reason for the use of a long time-step is the focus on long-term effects. The models are calibrated to present soil and water quality data, using the best estimates for deposition patterns for the past 100-140 years, obtained by scaling using the historical records of sulfur emissions at the nearby smelters (see Wright and Traaen 1992, Kämäri *et al.* 1995).

The model runs demonstrate the potential vulnerability of the region. As the soils are coarse and thin, there is a rapid depletion of the cation exchange capacity even with fairly low rates of sulfur deposition. The model results suggest that the soil base saturation should have been 10-25% higher 100 years ago than it is at present in order to reproduce the present-day base saturation values and the present-day surface water concentrations when driven with the assumed historical sulfur deposition for the area.

The gradual decrease of base saturation will continue both at the river Dalelva and the Christmas lakes if the deposition is assumed to remain at its present level. The sulfate adsorption assumed for the Christmas lakes acts only as a temporary sink for sulfur, and a gradual increase in sulfur concentrations takes place after the year 2000 (Figure 9·32).

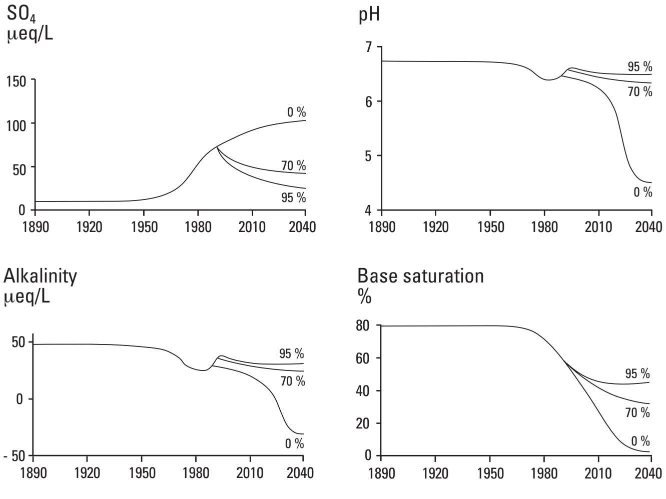

Figure 9·32. SMART model results for selected soil and lake output variables at Christmas lakes for the past 90 years and for three future scenarios assuming a 0%, 70% and 95% reduction of the present sulfur deposition levels (modified from Kämäri *et al.* 1995).

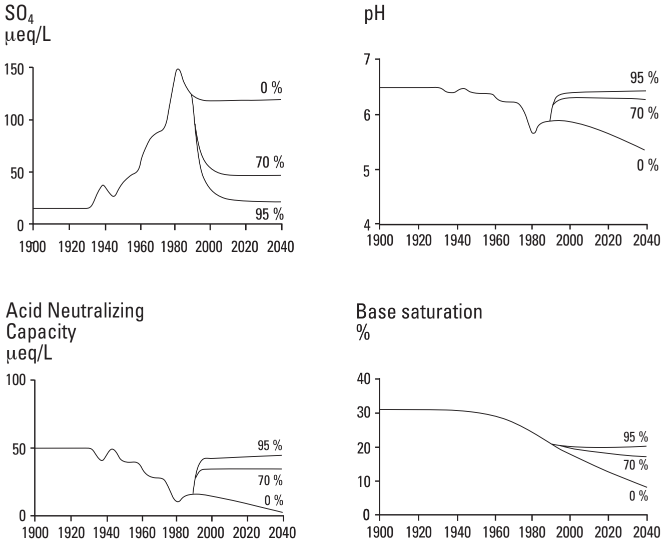

Figure 9·33. MAGIC model results for the streamwater at Dalelva for the past 90 years and for three future scenarios assuming a 0%, 70%, and 95% reduction of the present sulfur deposition levels (Wright and Traaen 1992).

The base saturation under this highest sulfur deposition scenario decreases rapidly to near zero (Figure 9·32), and a rapid acidification of the lake water results after the year 2010. A new steady state is achieved by the year 2035 with a negative alkalinity and a lake pH of around 4.5.

For Dalelva, the high sulfur deposition scenario, assuming a present sulfur deposition level of 66 meq/m^2/y, implies a continuing acidification and loss of base saturation in the soil, and loss of ANC in the surface water. According to the model results, base saturation would fall below 10%, ANC will be completely lost, and pH of values less than 5.5 will be reached by 2040 (Figure 9·33).

All scenarios at the river Dalelva and the Christmas lakes, assuming a drastic reduction in sulfur deposition to background levels by the year 2000, show a recovery for the soil and surface waters, and no adverse effects are expected. The recovery of the Christmas lakes for the low scenario, involving a decrease to background values, is slower than for the river Dalelva, as some of the adsorbed sulfate is slowly being leached out from the catchment soils into the surface water (see Figure 9·32).

The model results indicate that the future sulfur deposition will have to be very low in order to stop and reverse the ongoing acidification. However, since the present base saturation is fairly high (above 20-50%), there is still time to reduce the sulfur deposition to values which the system can tolerate. The model runs suggest that if strong enough reductions in sulfur deposition take place within 10-20 years, the changes in the soils will not result in adverse effects in the terrestrial or aquatic ecosystems.

9.12. Conclusions and recommendations

9.12.1. Conclusions

Sources of the acidifying compounds

Anthropogenic sources of sulfur and nitrogen are widely known on a global basis with sufficient certainty to allow modeling of the transport of these chemicals to the Arctic. Less well known are the natural sources of sulfur, e.g., DMS, within the Arctic.

Anthropogenic sources on the Kola Peninsula and at Norilsk, Russia, are the dominant sources of sulfur north of 65°N.

Decreasing emissions of sulfur have been seen in Europe. This decrease of emissions may not be identically reflected in the improvement of the acidification status because of a concomitant decrease in base cations as well.

Atmospheric processes

Arctic air chemistry of the sulfur- and nitrogen-bearing species is relatively well known due to continuous monitoring since the mid-1970s.

In the High Arctic of Canada and in Alaska, air concentrations of sulfates and nitrates have not changed appreciably since the early-1980s. On Svalbard, however, there is a decreasing trend attributable to reductions in European sulfur emissions.

Arctic Haze, as a visible phenomenon, brought attention to the acidification processes in the High Arctic, but like acidification, little environmental impact from the haze aerosol is likely to occur in the High Arctic.

Climatic effects from changes in atmospheric radiative forcing by haze aerosols is possible; however, the predicted degree of such impact varies among models.

Deposition of acidifying compounds

In the High Arctic, wet deposition may constitute up to 50% of the total sulfur and nitrogen deposition. Close to major smelter sources, dry deposition is about 75% of the total sulfur deposition.

Dry deposition velocities in the Arctic are not well known and estimates of removal rates will suffer from this lack of knowledge.

Modeling of concentrations and deposition of long-range transported acidifying compounds has been carried out for the Arctic. Verifications of models against observations have shown good agreement between model output and observed concentrations.

Environmental impacts

There is direct evidence of acidification effects on the Kola Peninsula and in a limited area in the Norwegian eastern Finnmark. Direct damage to forests, fish and invertebrates has been documented near the Kola smelters. Only restricted forest damages are noted outside the Kola Peninsula. In the immediate vicinity of the smelters, the negative effects of heavy metals are added to the acid stress.

Extensive vegetation damage has been reported from the region surrounding the smelter of Norilsk.

In adjoining regions of northern Finland and Norway, the critical loads for acidifying substances are exceeded in large areas. There exist indications of effects of acid deposition on stream fauna and small lakes, probably related to acid pulses during snowmelt. There is some evidence of decreasing alkalinity compared to earlier periods, which can be related to deposition.

There are no indications of anthropogenic acidification of surface waters in the North American Arctic. However, large areas of the North American Arctic are considered to be vulnerable to acidification.

Data on biological impacts are not available from North America, Greenland and most parts of Russia, e.g., Norilsk and the Taimyr Peninsula.

There are generally few monitoring stations in small streams, where acidification is considered most likely to first occur.

Modeling results indicate that the future sulfur deposition in Arctic Scandinavia and the Kola Peninsula will have to be very low to stop and reverse the ongoing acidification. However, strong reductions within 10-20 years will be beneficial as the soil base saturation is still fairly high.

9.12.2. Recommendations

Sources of the acidifying compounds

Emissions reductions of major sources in the Arctic have been modeled to show improvements to the environment. Such reductions should be carried out.

Atmospheric processes

Monitoring for trends of acidic species in the Arctic should continue at the high-latitude stations, Alert, Station Nord and Ny-Ålesund, and near regions where significant sources exist (the Kola Peninsula and Fennoscandia).

There is a lack of air chemistry data from eastern Russia and Alaska.

Better climate models are needed to assess the potential impacts of aerosols on the Arctic.

Deposition of acidifying compounds

Defining the parameters for dry deposition in the transport models should be improved in order to provide better estimates of deposition.

The development of the transport models for different spatial scales should continue, model intercomparison should be performed, and more emphasis should be placed on careful model verification.

Environmental impacts

Little quantitative information is available around the Norilsk smelter, and the impact of this source on the surrounding ecosystems should be investigated.

The effects of acid surges should be studied and modeled in Arctic conditions, when appropriate, using biological indicators.

More information is needed on the integrated soil-water relationships in the Arctic. Monitoring of small streams should be given high priority.

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