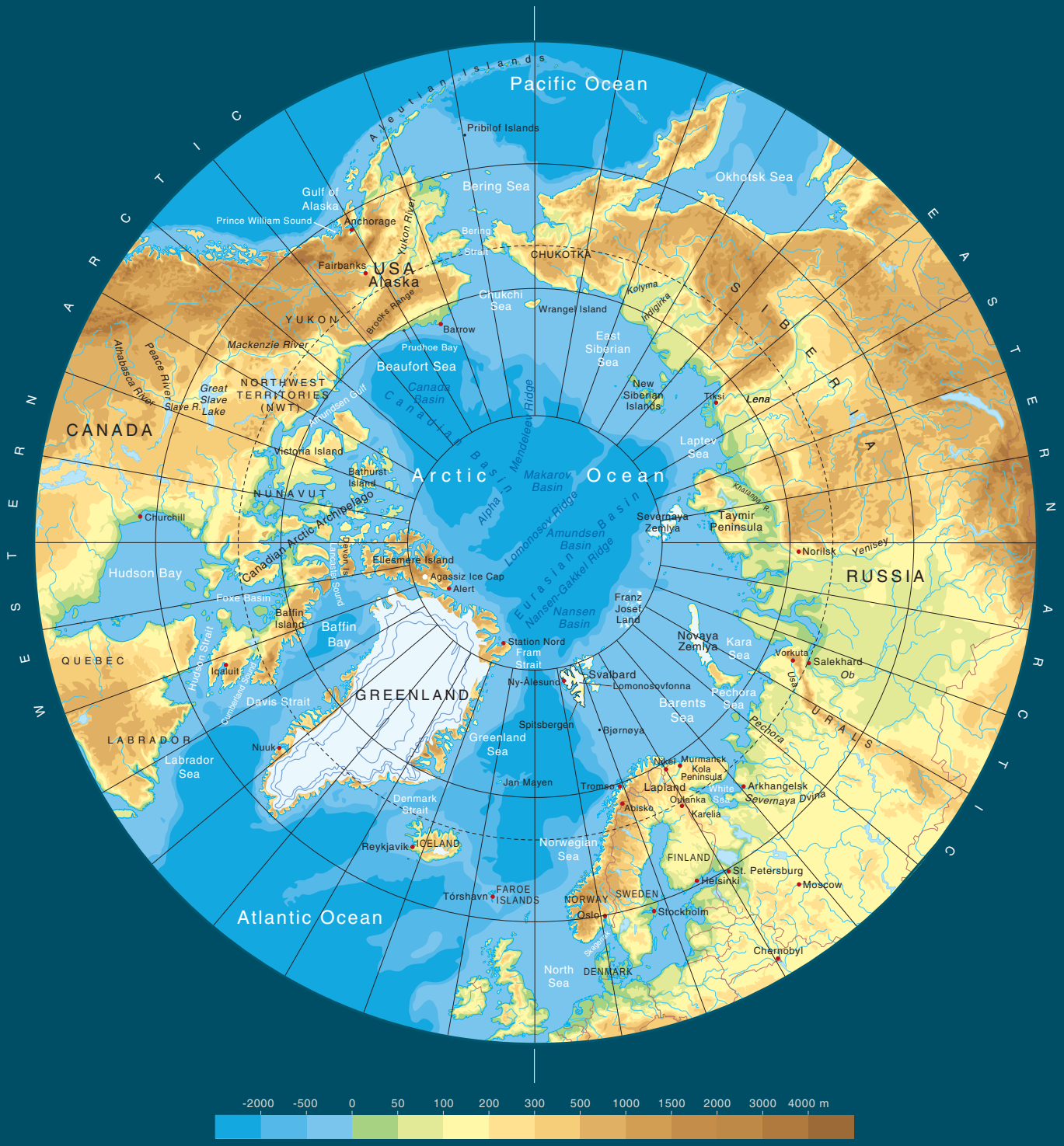
AMAP Assessment 2006:

Acidifying Pollutants, Arctic Haze, and Acidification in the Arctic

Arctic Monitoring and Assessment Programme (AMAP)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Contents

Preface

This report details the results of the 2006 AMAP assessment of Acidifying Pollutants, Arctic Haze, and Acidification in the Arctic. It builds upon the previous AMAP acidification assessment presented in 'AMAP Assessment Report: Arctic Pollution Issues' that was published in 1998.*

The Arctic Monitoring and Assessment Programme (AMAP) is a group working under the Arctic Council.

The Arctic Council Ministers have requested AMAP:

- *to produce integrated assessment reports on the status and trends of the conditions of the Arctic ecosystems;*
- *to identify possible causes for the changing conditions;*
- *to detect emerging problems, their possible causes, and the potential risk to Arctic ecosystems including indigenous peoples and other Arctic residents; and*
- *to recommend actions required to reduce risks to Arctic ecosystems.*

*This report provides the accessible scientific basis and validation for the statements and recommendations made in the AMAP Overview report, 'Arctic Pollution 2006: Acidification and Arctic Haze'** that was delivered to Arctic Council Ministers at their meeting in Salekhard, Russia in October 2006. It includes extensive background data and references to the scientific literature, and details the sources for figures reproduced in the Overview report. It also includes conclusions and recommendations of a scientific nature, such as proposals for filling gaps in knowledge, and recommendations relevant to future monitoring and research work. Some of these are taken up in the Overview report, although that report focuses more on recommendations that specifically focus on actions aimed at improving the Arctic environment.*

To allow readers of this report to see how AMAP interprets and develops its scientifically-based assessment product in terms of more action-oriented conclusions and recommendations, the 'Executive Summary of the Arctic Pollution 2006 overview report' is reproduced in this report on pages ix to xi.

The AMAP assessment is not a formal environmental risk assessment. Rather, it constitutes a compilation of current knowledge about the Arctic region, an evaluation of this information in relation to agreed criteria of environmental quality, and a statement of the prevailing conditions in the area. The assessment presented in this report was prepared in a systematic and uniform manner to provide a comparable knowledge base that builds on earlier work and can be extended through continuing work in the future.

The AMAP scientific assessments are prepared under the direction of the AMAP Working Group. The product is the responsibility of the scientific experts involved in the preparation of the assessment. The lead country for the AMAP Acidification and Arctic Haze Assessment under AMAP phase II was Finland. The assessment is based on work conducted by a large number of scientists and experts from the Arctic countries (Canada, Denmark/Greenland/Faroe Islands, Finland, Iceland, Norway, Russia, Sweden, and the United States), together with contributions from indigenous peoples' organizations, from other organizations, and from experts in other countries.

AMAP would like to express its appreciation to all of these experts, who have contributed their time, effort, and data; and especially to the lead experts who coordinated the production of this report, and to referees who provided valuable comments and helped ensure the quality of the report. A list of the main contributors is included in the acknowledgements on pages vi - vii of this report. The list is not comprehensive. Specifically, it does not include the many national institutes, laboratories and organizations, and their staff, which have been involved in the various countries. Apologies, and no lesser thanks, are given to any individuals unintentionally omitted from the list. Special thanks are due to the lead authors responsible for the preparation of the various chapters of this report.

The support of the Arctic countries is vital to the success of AMAP. AMAP work is essentially based on ongoing activities within the Arctic countries, and the countries also provide the necessary support for most of the experts involved in the preparation of the assessments. In particular, AMAP would like to express its appreciation to Finland for undertaking the lead role in supporting the Acidification and Arctic Haze assessment. Special thanks are also offered to the Nordic Council of Ministers for their financial support to the work of AMAP, and to sponsors of other bilateral and multilateral projects that have delivered data for use in this assessment.

The AMAP Working Group that was established to oversee this work, and the AMAP Acidification and Arctic Haze assessment group are pleased to present its assessment.

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Oslo, July 2006

* AMAP, 1998. AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. xii+859 p.

** AMAP, 2006. Arctic Pollution 2006: Acidification and Arctic Haze. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.

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Executive Summary to the *Arctic Pollution 2006* Ministerial Report

The first AMAP assessment – *Arctic Pollution Issues: A State of the Arctic Environment Report* – documented direct evidence of acidification effects on the Kola Peninsula and in limited areas of northern Norway and Finland, and around Norilsk in the Taymir region of Russia, mainly related to emissions from smelters in or close to these arctic areas. Acidification effects were also seen in some sensitive low-deposition areas of the European Arctic receiving pollutants from long-range transport. Data for areas of the North American Arctic and eastern Siberia that, due to their geology, are potentially vulnerable to acidification were generally lacking. So although the assessment did not find evidence of acidification effects in these areas, it concluded that improved information on possible acidification effects in these regions of the Arctic was desirable.

The present assessment builds on information in the first assessment and fills several gaps in knowledge. In particular it examines information on trends over the ten-year period since the first assessment was completed. It also addresses the need for more information on local sources of acidifying pollutants within the Arctic that were previously unknown or insufficiently quantified; the need for more information on contaminant levels and trends in some areas; the need to integrate physical and biological models with information on environmental measurements of sources and pathways; and the need for more information on the combined effects of climate change and contaminant pathways on acidification in the Arctic and arctic haze, including improvements of models for assessments. This assessment also considers links to hemispheric pollution issues.

Arctic Acidification

Arctic acidification is a subregional issue, and is only of major concern in areas with both sensitive geology and levels of acid deposition elevated to a point that exceeds the system's acid neutralizing capacity. Arctic haze is a visible manifestation of long-range transported air pollution. Arctic haze is largely composed of sulfate aerosol and particulate organic matter, which builds up in the arctic atmosphere during wintertime and appears in springtime over large regions of the Arctic, both in North America and Eurasia as haze layers with reduced visibility.

Sulfur is the most important acidifying substance in the Arctic, with nitrogen of secondary importance. Significant anthropogenic sources of sulfur emissions, and to a lesser extent nitrogen emissions, exist within the arctic region. In addition, long-range transported air pollutants contribute to acidification and arctic haze in the Arctic. Emissions from natural sources within the Arctic (volcanoes, marine algae, and forest fires) are very difficult to quantify and almost impossible to project.

Studies to date have been unable to show any significant health effects that are directly associated with emissions from the smelters that are the main sources of sulfur pollution within the Arctic. Epidemiological studies indicate that differences in health status of populations in areas of the Arctic with some of the highest levels of acidifying air pollutants, the Norwegian and Russian border populations, are more associated with socio-economic conditions than environmental pollution.

Trends

Some air and precipitation monitoring stations have now generated time series datasets that are long enough to show whether concentrations are increasing, decreasing, or staying the same over time. Sulfate concentrations measured in air at monitoring stations in the High Arctic (Alert, Canada; and Ny-Alesund, Svalbard) and at several monitoring stations in subarctic areas of Fennoscandia and northwestern Russia show decreasing trends since the 1990s. In contrast, levels of nitrate aerosol are increasing during the haze season at Alert (Canada), and possibly also at Barrow (Alaska) but longer data series are needed to confirm this trend. The increasing trends in nitrate are particularly apparent in recent years indicating a decoupling between the trends in sulfur and nitrogen. These observations are supported by modeling results.

Although further improvement in the acidification status of the terrestrial and freshwater ecosystems of the Arctic can be expected during the period until 2020, this is dependent on the implementation of existing international agreements to reduce emissions of acidifying substances. The Gothenburg Protocol to the UN ECE LRTAP Convention is the most important agreement in this connection. However, model projections based on full implementation of the Gothenburg Protocol indicate that the decreasing trends in deposition observed between 1990 and 2000 are likely to level off. Measurement data indicate that downward trends in concentrations may already be leveling off at some sites.

It is therefore recommended that:

- **All arctic countries are encouraged to ratify the UN ECE LRTAP protocol to Abate Acidification, Eutrophication, and Ground-level Ozone (the 'Gothenburg Protocol') and to support its implementation.***
- **Arctic countries look into the need to strengthen the provisions of the existing international agreements, and consider the need for new instruments to reduce emissions of acidifying substances.**

Significant reductions in emissions from the non-ferrous metal smelters on the Kola Peninsula, and to a lesser extent the Norilsk smelters, in the Russian Arctic have been achieved over the past ten years. Chemical monitoring data show that lakes in the Euro-Arctic Barents region are showing clear signs of a regional-scale recovery from acidification. Lakes close to the sources on the Kola Peninsula are showing the clearest signs of recovery.

*** The Protocol entered into force on 17 May 2005. As of July 2006, Denmark, Finland, Norway, Sweden and the United States have both signed and ratified, accepted, or approved the Protocol, Canada has signed but not yet ratified the Protocol, and Iceland and the Russian Federation have neither signed nor ratified the Protocol.**

However, non-ferrous metal production remains the dominant source of emissions of acidifying gases to the atmosphere within the Arctic. Other significant anthropogenic sources of sulfur emissions within or close to the Arctic include energy production plants and mining industries. Sources of nitrogen emissions within the Arctic include transportation, in particular shipping, and oil and gas activities. Detailed information on all of these sources is generally lacking.

It is therefore recommended that:

- **Information on emissions from arctic point sources in Russia, in particular information on emissions from the non-ferrous metal smelters on the Kola Peninsula and at Norilsk should continue to be made available. Information on emissions in other arctic areas should be improved.**
- **The impacts of acidification from arctic shipping and oil and gas activities, including future scenarios for emissions associated with these sources should be assessed.**

Links between Acidification, Arctic Haze, and other Environmental Issues

The causes and the effects of acidifying air pollutants and arctic haze are closely linked to other environmental problems. It is not clear how climate change will influence future acidification and arctic haze pollution in the Arctic. The effects of haze aerosols on the arctic climate are complicated by feedbacks between aerosols, clouds, radiation, snow and ice cover, and vertical and horizontal transport processes. Whether the pollutant aerosols cause an overall warming or an overall cooling is not yet known.

The amount of haze precursors (haze-inducing substances) reaching Alaska and the Canadian Arctic appears to have increased since the late 1990s. The frequency, severity, and duration of boreal forest fires appear to be increasing and the pollution plumes from these summer fires can extend over vast areas. In intense fire years, boreal forest fires may be the dominant source of black carbon (soot) for the Arctic. The importance of Asian sources to acidification and arctic haze pollution in the Arctic is not yet clear.

It is therefore recommended that:

- **Future AMAP assessments view acidification and arctic haze in the wider context of air pollution and climate change. The issues addressed in this more integrated type of assessment should include hemispheric transport of air pollutants, emissions from forest fires, particulate matter, and climate change effects.**

Gaps in Knowledge – Monitoring, Research, and Modeling

Atmospheric monitoring

Acidification is not known to have serious impacts in the Arctic outside the Kola/Fennoscandia region and the Taymir region in the vicinity of Norilsk. However, knowledge of acidification status in the Arctic is far from complete, particularly in relation to future effects. While Fennoscandia has several background air monitoring stations for acidification parameters, most areas of the Arctic have few, if any, background air monitoring stations.

Remote stations that are not affected by local or regional air pollutants are useful for studying trends in the levels of pollutants transported into the Arctic from long-range sources. Under AMAP, a network of arctic air monitoring stations has been established to assess trends in a range of pollutants, including acidifying substances, persistent organic pollutants, and metals such as mercury; however in recent years the overall coverage of this network has been reduced such that coverage is limited, particularly in Russia and the United States.

It is therefore recommended that:

- **A critical review of the existing arctic air monitoring network be conducted to identify the optimal number and location of long-term background monitoring stations for air and precipitation chemistry.**
- **To the extent possible, this network should be integrated with other monitoring and research planning, with the aim of developing a network of 'multi-purpose' background air monitoring stations in the Arctic.**

Episodic events

Short-term events of high atmospheric concentrations of sulfur dioxide are responsible for direct damage to vegetation at varying distances from the smelters. At many sites a large proportion of the annual acid deposition is accumulated in just a few days.

Similarly, pollutants deposited onto the snow pack accumulate throughout the polar winter and are released rapidly into rivers and lakes with snowmelt in spring. These pulses of very acidic water can cause short periods of very toxic conditions. Freshwater biota can be critically affected during acidic episodes and therefore assessments need to address both average conditions and conditions that may occur during episodic events.

It is therefore recommended that:

- **Further studies, with high temporal resolution, be conducted on the ecological impact of pulses or episodic events.**

Effects on terrestrial and freshwater ecosystems

In the European Arctic there are clear direct effects of sulfur dioxide emissions on trees, dwarf shrubs, and epiphytic lichens. The present deposition of acidifying compounds resulting from long-range transport of anthropogenic emissions at lower latitudes does not appear to be a threat to terrestrial ecosystems in most of the Arctic. In terms of their effects on plants, it is difficult to differentiate between the effects of acidifying air pollutants and elevated heavy metal levels in soils. Habitat destruction and possible changes in food availability are strongly reducing biodiversity in the immediate vicinity of the smelters.

It is therefore recommended that:

- **Future studies be conducted on terrestrial ecosystems to address the combined effects of acidifying sub-**

stances and heavy metals and other relevant factors in an integrated manner.

Available terrestrial and freshwater monitoring data provide irregular and incomplete coverage of the Arctic, even in acid-sensitive regions. Similarly, assessments of biological effects of acidification in arctic surface waters are largely based on sparse and isolated data.

It is therefore recommended that:

- **Coordinated monitoring and research be carried out to provide more chemical and biological data on effects and trends in terrestrial and freshwater ecosystems in the most impacted areas of the Arctic.**

Modeling

Modeling is one of the most important tools available for gaining insight into the possible pollution status of the extensive areas of the Arctic where the observational networks are absent or poorly developed. Models also allow investigation of scenarios for future trends, and for linkages between contaminant pathways and, for example, climate change.

It is therefore recommended that:

- **Existing air transport and deposition models be improved and further validated using measurements of sulfur compounds, nitrogen compounds, and black carbon in the Arctic, including measurements conducted during field campaigns.**
- **Studies be conducted to identify and provide estimates of sources of black carbon to the Arctic.**
- **Data sets gathered during aircraft and ground-based surveys, in particular, long-term data sets, be integrated for use in three-dimensional arctic climate models designed to evaluate climate forcing by arctic haze.**

Cooperation on monitoring

Close cooperation between AMAP and other international organizations involved with monitoring and modeling deposition and effects of acidifying pollutants within the European Arctic, such as programs under the UN ECE LRTAP Convention, have proven mutually beneficial. The new EANET (Acid Deposition Monitoring Network in East Asia) initiative represents an opportunity to develop similar cooperation in relation to monitoring in the Far East of Asia.

It is therefore recommended that:

- **AMAP continues to develop its cooperation with relevant international organizations, in particular to obtain more precise data on emissions from southeast Asia and to investigate the possible impact of these emissions on the Arctic.**
- **Resources be made available to ensure that relevant existing and future national data on acidification parameters, in particular from arctic monitoring stations, are reported to the AMAP database at NILU according to agreed procedures.**

Chapter 1
Introduction

Martin Forsius

As a consequence of the intense scientific and environmental policy work on acidification, the atmospheric emissions of acidifying pollutants, the atmospheric processes, and the environmental impacts of these pollutants on different ecosystems are now well understood. The acidifying compounds – sulfur dioxide (SO_2), nitrogen oxides (NO_X), and ammonia (NH_3) – have different sources. Sulfur dioxide emissions are mainly associated with point sources such as power plants, smelters, pulp and paper mills, and oil and gas processing. As well as these point sources, NO_X emissions are also derived from diffuse sources such as vehicles. Ammonia emissions are almost entirely derived from agricultural sources and so are more difficult to quantify. A full understanding of the acidification problem requires information on the emissions and processes of neutralizing compounds such as base cations. Neutralizing compounds are derived from anthropogenic and natural sources.

Rapidly increasing scientific evidence on acidification during the 1970s and 1980s was the starting point for international negotiations on controlling emissions of compounds that undergo long-range transport. The Convention on Long-Range Transboundary Air Pollution (UN ECE CLRTAP) and the air pollution work and directives of the European Commission have been key international activities in this respect. Large international emissions databases, pollutant transport models, environmental impacts monitoring and assessment programs, and integrated assessment models have been created within these frameworks and have served as technical infrastructures for negotiations (e.g., Sliggers and Kakebeeke, 2004; UN-ECE, 2004b). Work on the acidification issue has been one of the great environmental 'success stories' and SO_2 emissions were reduced by around 67% in Europe between 1980 and 2000; with many countries having reductions of almost 90% (EMEP, 2004).

Since the 1970s, the focus on air pollution has widened considerably and is now moving toward issues such as eutrophication, small particulates and health effects, ground-level ozone, heavy metals, and the interactions between these issues and with climate change. Rapid industrial growth and increasing energy use in several world regions (including heavily populated countries such as China and India) have added a new dimension. There is also increasing evidence of the importance of hemispheric transport of pollutants, such as ozone, mercury, and persistent organic pollutants.

In the 1970s it was recognized that even remote parts of the Arctic are influenced by air pollution, mainly due to anthropogenic activities at lower latitudes. Arctic haze was first reported in the 1950s by pilots flying in the Canadian and Alaskan Arctic, but it was not until the mid-1970s that its anthropogenic origin was established (e.g., Rhan *et al.*, 1977). Arctic haze is a varying mixture of sulfate, particulate organic matter, nitrogen compounds, dust and black carbon, as well as trace elements such as heavy metals and other contaminants. Arctic haze aerosol particles thus provide a transport pathway for contaminants to the Arctic. The impact of arctic haze on climate forcing is receiving increasing attention.

Within the Arctic itself there are a few but very significant sources of air pollutants. Production of copper, nickel, and other non-ferrous metals from sulfur-bearing ores creates the largest emissions of acidifying compounds (mainly sulfur) and heavy metals. Most of these smelter emissions come from the Nikel, Zapolyarnyy, and Monchegorsk complexes on the Kola Peninsula and from Norilsk on the Taymir Peninsula in northwestern Siberia. There are also emissions from several large cities, notably Murmansk with around 400 000 inhabitants. Consequently, the regions surrounding the large smelter complexes in northern Russia, as well as the northeastern areas in the neighboring countries of Norway and Finland, are the areas where most acidification and other air pollution impact studies have been undertaken over the last few decades.

At the First Ministerial Conference on the Arctic Environmental Protection Strategy (AEPS), held in Rovaniemi, Finland in June 1991, Ministers of the Arctic States established the Arctic Monitoring and Assessment Programme (AMAP) to 'monitor the levels of anthropogenic pollutants in relevant compartments of the Arctic environment'. Ministers further identified persistent organic pollutants, heavy metals, and radioactivity as the key pollutants that should be a priority during the first phase of AMAP (1991–1997). The Ministerial Conferences in Nuuk, Greenland (1993) and Inuvik, Canada (1996) extended this list to include: acidification and arctic haze, and petroleum hydrocarbon pollution, in a subregional context; and environmental consequences of, and biological effects due to global climate change and stratospheric ozone layer depletion, relevant to the Arctic.

An assessment of acidification and arctic haze was carried out within the AMAP framework and reported in the extensive and fully-referenced AMAP Assessment Report: Arctic Pollution Issues (AMAP, 1998). This concluded that there was direct evidence of acidification effects on the Kola Peninsula and in a limited area of the Norwegian part of eastern Finnmark. Direct damage to forests, fish, and invertebrates was documented near the Kola smelters. There were no indications of any acidification impacts in the North American Arctic although large areas were considered potentially vulnerable to acidification.

This assessment updates information in the previous assessment. The main aim has been to include new knowledge of processes, sources, and pathways, to fill gaps in knowledge on contaminant levels and trends and to correct possible errors in the previous assessment. Interactions with other issues (mainly climate change and heavy metals) and connections to hemispheric pollution issues are also considered.

The structure of this report follows the pollution pathways from emission sources (chapter 2), through transport and deposition processes (chapter 3), and impacts and processes of aerosols (chapter 4), to the present status and trends in the chemical and biological responses to acidifying pollutants in terrestrial and aquatic ecosystems (chapters 5 and 6). Chapter 7 reviews the human health impacts of acidifying air pollutants in the Arctic, while chapter 8 summarizes the main conclusions of this assessment and presents recommendations for further work.

Chapter 2
Sources of Acidifying Pollutants and Arctic Haze Precursors

Lars R. Hole, Jesper Christensen, Martin Forsius, Marjut Nyman, Andreas Stohl, and Simon Wilson

2.1. Sources within the Arctic

Within the Arctic, the major anthropogenic emissions of nitrogen oxides (NO_X) and sulfur dioxide (SO_2) are associated with sources located in a limited number of areas where industrial enterprises and/or population centers occur. With the exception of oil and gas related activities, these emission sources are almost exclusively located in the northern territories of the Russian Federation. According to the Russian Federal Statistical Committee (Goskomstat), emissions of SO_2 in arctic regions of Russia accounted for 33% of the total SO_2 emissions from all territories of the Russian Federation.

Despite generally very low population densities, large towns and cities do exist within the (sub-)Arctic, notably Murmansk with its population of 400 000. There are also natural sources located within the Arctic; these include volcanoes that emit SO_2 in the volcanically active areas of, for example, Kamchatka, Alaska, and Iceland. However volcanic emissions are not addressed in this assessment. Natural biogenic emissions associated with forest fires are another source of natural emissions and these are increasingly prevalent in arctic areas as a consequence of climate change (ACIA, 2004). These emissions are described further in section 2.1.5.

Energy production and transport in and around arctic urban population centers are responsible for reduced air quality at the local scale, including pollution by NO_X, SO_2, and fine particulates, which can have negative health implications (see chapter 7). However, industrial sources are responsible for the major emissions of concern within the Arctic. Although few in number, some of these industrial sources, in particular those associated with non-ferrous metal smelting operations, are significant, both at a regional scale and in terms of global ranking of individual sources.

The first AMAP assessment (AMAP, 1998) concluded that NO_X makes a negligible contribution to the acidification effects observed within the Arctic, and that arctic sources are insignificant compared with the amount of NO_X carried into the Arctic via long-range transport. The present assessment confirms this finding, but due to the differences achieved in reductions of SO_2 relative to NO_X, and the potential for increased development of the Arctic that may enhance NO_X emissions within the region, this assessment addresses NO_X in more detail than the first AMAP assessment. There is also increasing evidence documenting intercontinental transport of air pollutants, including NO_X, SO_2, fine particles, and black carbon.

2.1.1. Stationary sources: industry and energy

The first AMAP assessment clearly identified the non-ferrous metal smelters at Nikel, Monchegorsk, and Zapolyarnyy on the Kola Peninsula and at Norilsk in northern Siberia, as the largest (anthropogenic) sources of acidifying air pollutants within the Arctic (AMAP, 1998). Although emissions reductions have been achieved over the last decade, these smelters remain the largest sources within the Arctic and so this chapter also focuses on these sources.

Non-ferrous metal production is one of the greatest sources of environmental pollution in Russia, second only to energy production in accounting for 'harmful emissions' (SO_2, dust, NO_X, etc.) to the atmosphere. Sulfur dioxide, originating from the roasting and smelting of sulfur-containing minerals, makes up the bulk (ca. 80%) of the emissions from non-ferrous metal processing plants. Around 75% of the SO_2 emissions are associated with the production of nickel in the nickel-cobalt sector, with most of the rest from the copper production sector. Emissions from aluminum plants are mainly 'dust' (particulates) and carbon dioxide (CO_2).

Based on information (1998–2001) from reports of companies involved in non-ferrous metal production in Russia, ten companies are responsible for around 85% of the total 'harmful emissions' to the atmosphere. The non-ferrous metal smelters at Norilsk, Zapolyarnyy, and Monchegorsk (all operated by MMC Norilsk Nickel) are all within or close to the Arctic and together account for 68% of the total 'harmful emissions' from non-ferrous metal production in Russia. The Norilsk Nickel consortium is the largest producer in the Russian non-ferrous metal sector and one of the world's largest producers of nickel, palladium, platinum, cobalt, and copper.

Time series of SO_2 emissions from the non-ferrous metal smelter at Nikel (on the Kola Peninsula) between 1980 and 2002 (Figure 2.1) illustrate the substantial reductions that have occurred over this period. Decreased emissions during the early 1990s are largely due to economic decline following the break-up of the former Soviet Union, although by 1995 emissions at Nikel had returned to their 1990 levels (ca. 180 kt SO_2). The main emissions reductions have been achieved since 1996.

In recent years, the MMC Norilsk Nickel plants have also considerably reduced their emissions of SO_2. Abatement measures include the (by-)production of sulfur (ca. 260 kt in 2000) and sulfuric acid (ca. 30 kt in 2000) from captured SO_2, although this is not a profitable activity due to high transportation costs and distance from markets. Emissions from the Pechenganickel (Zapolyarnyy/Nikel) and Severonickel (Monchegorsk) combines have been reduced from around 255 kt in 1991 to around 150 kt in 2000, and from around 180 kt in 1992 to around 45 kt in 2000, respectively, as a result of introducing emission control technologies and the closure of the Severonickel smelting plant.

In 1999, MMC Norilsk Nickel announced extensive reconstruction of its production facilities, including the introduction of technologies to reduce emissions of harmful substances. Measures include modernizing the processing of copper-nickel ores using new flotation reagents in order to reduce sulfur minerals prior to roasting and smelting, and reconstruction of facilities to produce and utilize technical-grade sulfur on a profitable basis.

Part of the reconstruction of facilities on the Kola Peninsula is being funded through environmental cooperation agreements between Russia, Norway, and Finland, and emission reductions from these sources are being imple-

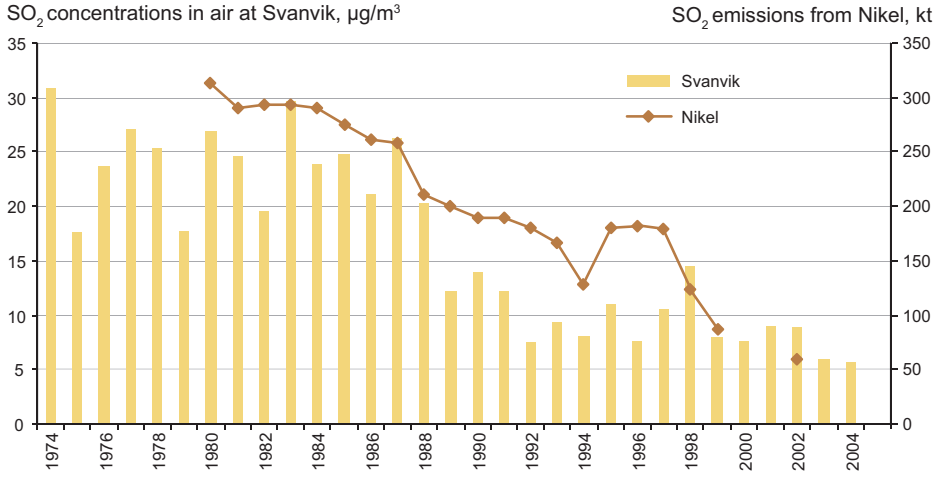


Figure 2.1. Annual SO_2 concentration in air at ground level at the Svanvik monitoring station and SO_2 emissions from the non-ferrous metal smelters at Nikel (after Hagen *et al.*, 2005).

mented in accordance with agreements under the LRTAP Convention to reduce SO_2 emissions relative to 1980 levels by 40% by 2005 and 50% by 2010.

In addition to the smelter complexes, other industries also emit SO_2 to the atmosphere including the energy sector. An inventory of the 130 largest coal-fired power stations in the Russian Federation (VTI/IVL, 2004) lists nine plants within the Arctic (see Figure 2.2). These had combined SO_2 emissions of around 80 kt in 2002 (85% from the plants at Severodvinsk, Vorkuta, and Apatity), which is less than 5% of the combined SO_2 emissions from the Kola and Norilsk smelters in the same period.

Total SO_2 emissions from the five regions of northwestern Russia – Murmansk Oblast, Republic of Karelia; Archangelsk Oblast, Nenets Autonomous Okrug and Komi Republic – were 240 (ca. 85% from the Kola smelters), 77, 89, 3.8, and 65 kt, respectively in 2002. Additional industry related sources are the pulp and paper industry in the Republic of Karelia and Archangelsk Oblast and the oil and gas industry in the Nenets Autonomos Okrug and Komi Republic.

With the possible exception of oil and gas related sources (see section 2.1.3), there are few significant point sources of SO_2 within the Arctic outside the territories of the Russian Federation. The few sources that do exist include mining activities and small-scale power generation or waste incineration plants located in population centers.

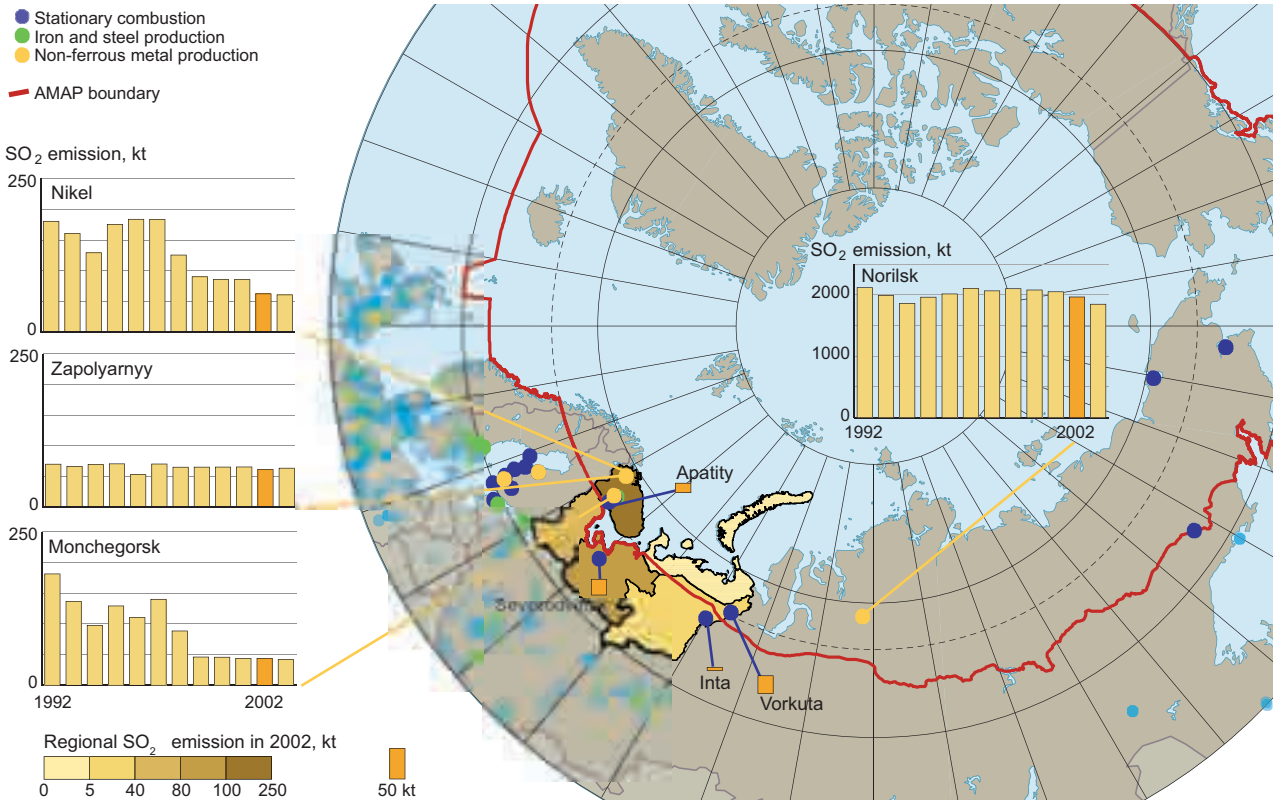


Figure 2.2. Sulfur dioxide emissions from metallurgical industry sources and major coal-fired power plants in Russia (there are no significant point sources north of 60° N in other Arctic countries; pale coloured symbols are sources south of 60° N). Also shown are total emissions of SO_2 in 2002 from the five regions of northwestern Russia. Subplots show trends in emissions from Russian non-ferrous metal smelters. Orange columns on the subplots, and on the map (for the four power plants with the largest SO_2 emissions), represent emissions in 2002 (note: the scale on the Norilsk subplot is different from all other data shown). Sources: smelter emissions: Ministry of Natural Resources official statistics; coal-fired power plant emissions: VTI/IVL, 2004.

2.1.2. **Local air pollution in Russian cities**

Urban air monitoring data from Russian cities can help assess the influence of the main pollution sources (Figure 2.3). However, it is important to remember that such data represent average and maximum concentrations in ambient urban air, and that as well as industrial emissions these concentrations also reflect other urban activities including vehicle emissions.

The highest NO_X concentrations among cities in the Russian Arctic region were observed at Norilsk and Murmansk up to 1998, but since then high concentrations have also occurred in Salekhard and Nikel. Concentrations of SO_2 in Russian cities have been consistently highest in Nikel and Norilsk.

Between 1990 and 2003 there was a clear increase in NO_X concentrations in Russian cities. This is particularly obvious at Nikel, Norilsk, and Salekhard after 1997. Average and maximum NO_X concentrations are now two to three times higher in Nikel than they were between 1990 and 1997, and one and a half to two times higher in Monchegorsk, Norilsk, and Salekhard. Maximum NO_X concentrations in Norilsk increased until 1999, but have decreased significantly since then. These increases also reflect the big increase in the number of private vehicles in the Russian Arctic well as in the rest of Russia over the last ten years.

There was a general decrease in SO_2 concentrations between 1990 and 2003 in Nikel, Monchegorsk, and Norilsk. However, interannual variability is very high (in Norilsk annual average concentrations can vary by a factor of two). This variability makes it impossible to detect clear trends in SO_2 concentration.

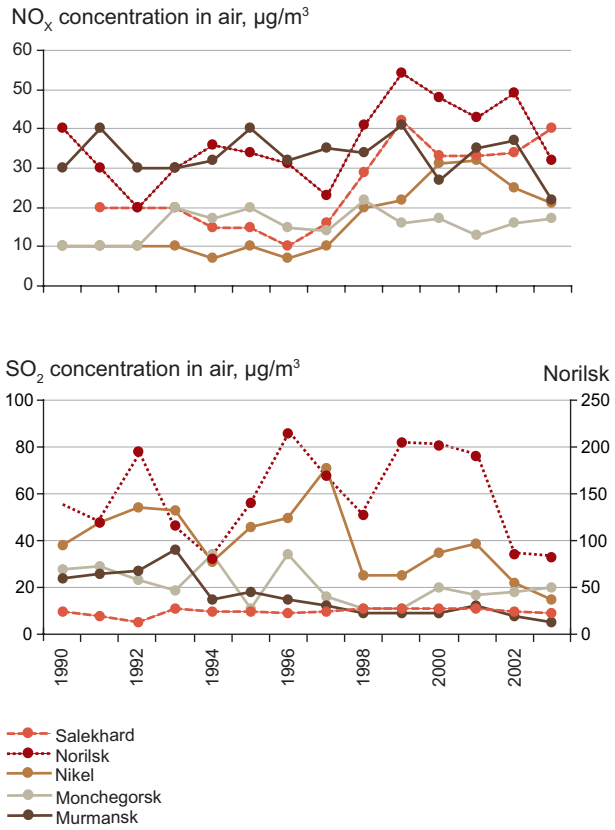


Figure 2.3. Trends in NO_x and SO_2 concentrations in air in various cities in the Russian Arctic.

2.1.3. **Oil and gas activities**

The Arctic is estimated to contain at least 25% of the world's undiscovered petroleum resources (Ahlbrandt, 2002). It has been suggested that by 2010, annual volumes of 150 million tonnes of oil may be shipped by sea as a result of the development of oil production and transportation infrastructure in the Barents region alone (Frantzen and Bambulyak, 2003).

As energy consumption and air pollution are directly linked, any increase in oil and gas related activities is likely to result in increased pollutant emissions. According to WWF (2005) oil and gas development will pose a major threat to the Barents Sea region.

From the exploration phase to final closure of the production field, oil and gas production involves emissions and discharges to air and water. Emissions to air include exhaust gases containing CO_2, NO_X, sulfur oxides (SO_X), methane (CH_4) and non-methane volatile organic compounds (nmVOC) from various types of combustion equipment and sources including gas turbines, engines and boilers; gas flaring; and oil and gas burning in connection with well testing and well maintenance work. Other sources of hydrocarbon gases (CH_4 and nmVOC) include gas ventilation, minor leaks, and diffuse emissions and boiling down of hydrocarbon gases (largely nmVOC) from the storage and loading of crude oil. Power generation, using natural gas and diesel oil as fuel, is the predominant reason for CO_2 and NO_X emissions, followed by gas flaring. The main cause of SO_X emissions is the combustion of sulfur containing hydrocarbons.

The Arctic Offshore Oil and Gas Guidelines prepared by the Arctic Council Working Group on Protection of the Arctic Marine Environment (PAME, 2002) provide an overview of potential impacts of offshore development in the Arctic and how these impacts should be taken into consideration in offshore oil and gas activities. The guidelines cover air emissions, which mainly originate from the combustion of fuels for power generation, and direct emissions from the production, treatment, storage, and transportation of oil and gas. According to PAME, "the activities will entail considerable inputs of gases into the atmosphere from power generation, flaring, well testing, leakage of volatile petroleum components, supply activities and shuttle transportation. These air emissions may have effects on the climate and they may cause acidification on nearby land and contribute to emissions of any number of hazardous substances." Methane, which is a powerful greenhouse gas, is also released to the atmosphere by gas drilling, from leaky pipelines, and by venting and flaring activities on oil and gas rigs (AMAP, 1998).

The ongoing AMAP assessment of petroleum hydrocarbons in the Arctic will address the environmental impact of oil and gas developments in the Arctic, including the associated effects on human health and social and economic consequences. It will address emissions from oil and gas activities, however comprehensive coverage of all air emissions from the many activities related to oil and gas production may warrant a separate assessment at some point in the future.

At present, the main areas of oil and gas exploitation in the Arctic include the North Slope of Alaska, the offshore areas in the Norwegian and Barents Sea, and the Yamalo-Nenets and Komi areas of Russia. Extensive petroleum resources also exist in the Canadian Arctic, which may be developed if the economic situation is favorable. Oil and

gas exploration activities are being actively pursued in other parts of the Arctic, including the offshore areas west of Greenland, around the Faroe Islands, and in the Kara, Chukchi, and East Siberian Seas.

Although detailed information on emissions associated with oil and gas activities in the Arctic is lacking, available information can be used as an example. The petroleum sector makes a considerable contribution to the national total emissions for Norway. For example, a large part of Norway's VOC emissions are from petroleum-related emissions in the North Sea. Similarly, an estimated 20% of Norwegian NO_X emissions in 2000 were derived from oil and gas resource development (this figure excludes related shipping activities). For SO_X, total emissions by the Norwegian oil and gas industry in 2003 were around 600 tonnes; the low emissions figures reflect the low sulfur content of the fuel gas used in gas turbines and engines. As a result, diesel combustion was the main source of SO_X emissions from the oil industry. Annual Norwegian SO_2 emissions were 23 000 tonnes in 2003; thus, the Norwegian oil industry accounts for approximately 2.5% of total emissions in Norway (OLF, 2003).

A study into the possible impact of hydrocarbon activities in the Lofoten-Barents Sea area involved development scenarios with three levels of activity. Air emissions in each scenario included emissions from offshore and inshore facilities and from transportation. The areas affected by emissions from hydrocarbon production in the Barents Sea-Lofoten area are Finnmark, Troms, and northernmost Nordland (Guerreiro *et al*., 2003). In each scenario, the total increase in air emissions relative to national emissions in 2001 is 3 to 8% for CO_2 and 0.7 to 2.8% for NO_X. Transport contributes 50% for NO_X (DNV, 2003). However, the projected increase in deposition (e.g., for nitrogen) relative to that at present seems higher than the relative increase in emissions; up to 25% at sea and 5 to 15% on land owing to the currently very low deposition values (around 400 mg/m^2 per year).

The extensive impact assessment of year-round petroleum activities in the Lofoten-Barents Sea by Norwegian authorities indicates that there will be emissions to air, which although mostly insignificant in terms of impact, may in some cases exceed critical limits.

According to DNV (2003) the normal level of activity in the oil industry does not cause serious effects in the marine environment. However, in some cases, there may be significant CO_2 emissions to air. For example, increases of 15 to 20% in total CO_2 emissions may occur due to increased energy use by the discharge of produced waters. Transportation would cause most of the nitrogen emissions. The contribution from the oil industry to acidification is considered to be low and emissions are thought not to exceed critical limits (DNV, 2003). None of the scenarios led to exceedance of critical loads for terrestrial ecosystems. However, critical loads for northern ecosystems are not well established and more careful mapping of ecosystem types is required in the vicinity of planned/prospective land-fast constructions or facilities related to the oil and gas industry. The DNV report concluded that oil-field specific investigations should be made of the effects on the local environment of increased emissions to air. Also, that it is difficult to estimate the effect of increased air pollution from oil and gas activities on the arctic haze phenomenon, but that this may be significant since the haze is known to consist of anthropogenically produced impurities (see chapter 4).

While the impact of oil and gas activities on climate in the arctic areas is a less well-covered issue, a warming climate has several consequences for oil and gas production in the Arctic. Climate change is projected to lead to less sea ice. The use of oil and gas resources in remote arctic locations will increase as the ice-free season increases and ice cover decreases. Increased transportation activities will increase the risk of oil spills. Emissions to the atmosphere are also likely to increase. Transport in general uses more energy than industrial, agricultural, commercial, and institutional sectors combined. In addition to enhancing oil and gas activities, climate warming will increase risks in winter operations such as the construction of ice roads (Thurston, 2003).

In many cases, international agreements commit signatories to limiting their air emissions to certain levels. Greenhouse gas emissions may be increased by oil and gas activities, Norway can meet its commitments to reducing greenhouse gas emissions through the Kyoto mechanisms using national reductions or international emissions trading. Other gases (NO_X, SO_2, and VOC) causing regional environmental problems, such as acid rain, eutrophication, and ground-level ozone, are regulated by the LRTAP Convention. Of the countries with prospective oil and gas fields on the continental shelf, the Gothenburg Protocol has been signed and ratified by Norway and the USA and signed by Canada. Russia has not signed the protocol.

2.1.4. **Shipping activities**

Emissions from shipping are an important component of total global emissions of SO_2 and NO_X and are of concern in the Arctic because they can influence nearby landmasses. According to the EDGAR database (Emission Database for Global Atmospheric Research), international marine transport contributed around 7300 kt of SO_2 and around 9600 kt of nitrogen dioxide (NO_2) to global emissions in 2000; corresponding to around 5 and 7.5% of total global emissions, respectively. For both SO_2 and NO_X, the eight arctic countries are responsible for around 25% of the estimated global emissions due to international marine transport.

Within the Arctic, emissions to the air from shipping in 1998 were estimated at 30 kt of SO_2 and 105 kt of NO_X (Table 2.1). These estimates are based on a statistical emissions modeling approach using available data on ship-

Table 2.1. Emissions to air in the Arctic and fuel consumption by arctic shipping in 1998 (kt) (Norwegian Maritime Directorate, 2000).

	NO_X	CO	nmVOC	SO_2	CO_2	Particulate matter	CH_4	N_2O	Fuel
Wet cargo	6.4	0.6	0.2	3.2	276	0.44	0.03	0.01	87
Dry cargo	40.1	4.9	1.6	10.6	2101	1.38	0.20	0.05	663
Fishing vessels	47.6	5.6	1.8	13.9	2412	1.83	0.23	0.06	761
Icebreakers/Tugs	11.3	1.4	0.5	2.4	610	0.30	0.06	0.02	192
Total	**105.4**	**12.5**	**4.1**	**30.0**	**5399**	**3.95**	**0.51**	**0.14**	**1703**

nmVOC: non-methane volatile organic compounds.

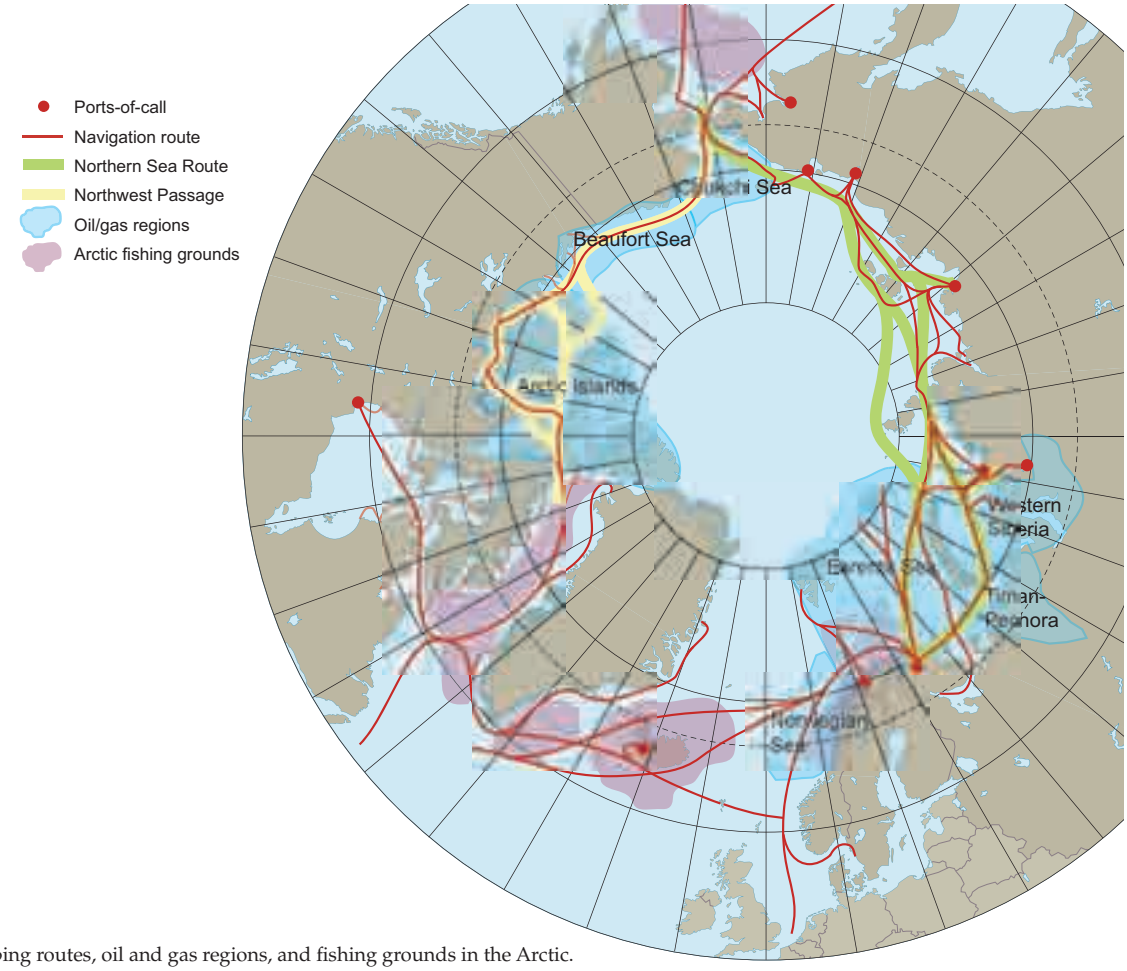


Figure 2.4. Shipping routes, oil and gas regions, and fishing grounds in the Arctic.

ping volume, estimated fuel consumption, and relevant emission factors (DNV, 1999). In addition to commercial shipping (cargo transport and cruise shipping), these estimates also include emissions from fishing vessels. The DNV study noted inadequacies in the available shipping statistics, however. For example, the estimates do not fully address emissions from shipping on inland waterways, which is a very significant shipping activity in the Arctic and in northern Russia in particular. Also, the estimates do not include emissions from military shipping. Although military activity in the Arctic (such as operations by the Russian Northern Fleet) have been reduced considerably since the end of the Cold War, emissions associated with naval vessels and other military installations may be a significant additional component.

The most extensive shipping activities in the Arctic take place in Russia, with the Northern Sea Route (NSR) carrying the largest volume of traffic of any arctic seaway. The NSR connects the Barents Sea and the Bering Strait and serves ports at the mouths of the major Russian rivers (Figure 2.4) providing a connection with the extensive ship and barge transport along the major inland waterways of northern Russia. The NSR was officially opened for international transit trading in 1991; although there has been little commercial utilization by non-Russian vessels to date. The NSR is restricted by sea ice and open for as little as two and a half months of the year at some points. Annual traffic along the NSR peaked at around 6600 kt in 1987 but by 2000 had decreased to around 1500 to 2000 kt, reflecting declining economic activity in this part of Russia following the break-up of the former Soviet Union. Recently, however, traffic volumes have increased and are projected to continue to increase over the next 10 to 15 years, with part of this increase associated with the transport of oil from northern Russia to markets in Europe and Asia (Brigham and Ellis, 2004). Transport of petroleum products from northern Russia, together with increasing cruise traffic is also resulting in increased shipping activity in the Barents Sea and the Norwegian Sea. There are also extensive maritime activities in Canadian arctic waters (i.e., the Beaufort Sea, Northwest Passage, and Hudson Bay), including deliveries of general cargo and fuel through the Mackenzie River system, and the supply of cargo and petroleum products to eastern arctic communities. Projected reductions in sea-ice thickness and extent associated with climate change, and a resulting increase in the length of the season during which the NSR and Northwest Passage are open for navigation have raised the possibility that shipping activities in arctic waters may increase substantially in future (ACIA, 2004). According to a recent study (Lee Behrens, 2000) the most significant emissions from merchant shipping in 2000 were associated with the transport of dry cargo.

The other major source of emissions was fishing vessels, reflecting the large size of the fleets, high fuel consumption by (in particular larger) fishing vessels, and the long periods spent at sea. Because fishing is a largely seasonal activity, at least for the smaller fishing vessels (<30 m) that make up 80% of the fishing fleets, emissions occur primarily during the summer. Large-scale commercial fishing activities are concentrated in the Barents and Bering Seas.

Although estimates of emissions from shipping in the Arctic are subject to considerable uncertainty due to the current lack of detailed information on shipping statistics, several observations are possible. Taken together, emissions from shipping in the Arctic are currently small compared with global emissions from marine traffic (<1% for SO_2 and approximately 1% for NO_X). Sulfur dioxide emissions from arctic shipping are also minor compared to emissions from other sources within the Arctic (e.g., <5% of the emissions from Russian smelters), however this is not the case for NO_X with shipping emissions more than double the reported emissions from Russian non-ferrous metal production.

The environmental impact of NO_X and SO_2 emissions from shipping in the Arctic will depend upon their contribution relative to existing background concentrations in the areas concerned. The seasonal nature of some inputs must also be considered. Increased shipping traffic density may contribute significantly to the load of acidifying gasses in some areas (e.g., close to ports), however available evidence suggests that shipping is not currently a major contributor to overall levels of acidifying pollution in the Arctic. An increase in arctic shipping traffic is expected during the next 10 to 15 years, as a result of increasing oil transport (particularly in the Barents Sea) and a longer navigation season due to reduced ice cover. However, emissions associated with this increased shipping activity will probably be partly compensated by improved technology, and possibly also by reduced levels of fishing activity. Thus, dramatic changes in sulfur and nitrogen emissions from shipping are not expected in this period. In the longer term, a more significant increase in shipping, for example due to ice-free conditions in the NSR, could lead to more substantial emissions of acidifying pollutants in the region.

2.1.5. Natural sources within the Arctic: wildfires

Wildfires are a large episodic source of black carbon and other pollutants (Lavoué *et al*., 2000). It was realized only recently that boreal wildfire emissions affect the atmosphere at a hemispheric scale (Wotawa *et al*., 2001). Pollution plumes originating from boreal fires have been observed over downwind continents (Forster *et al*., 2001) and can circle the entire northern hemisphere (Damoah *et al*., 2004). An aircraft campaign frequently found aerosol plumes from Alaskan and maybe also Siberian forest fires over the Alaskan Arctic (Shipham *et al*., 1992) and a PhD thesis suggests a link between boreal forest fires and black carbon observations in Greenland and at other arctic sites (Lavoué, 2000). The increase in areas burned over recent decades (Lavoué *et al*., 2000; Kasischke *et al*., 2005), which is probably due to a warming in the boreal region, is a matter of concern. There are speculations that black carbon deposits from boreal forest fires could enhance the melting of arctic glaciers and sea ice (Kim *et al*., 2005). Boreal fires are a summer-time phenomenon that occur when removal mechanisms (wet and dry deposition) are relatively efficient and the arctic troposphere is generally much cleaner than in winter. But precipitation (and hence wet removal) is suppressed by the high particle numbers in the vicinity of the fires (Andreae *et al*., 2004), possibly allowing much of the black carbon to reach the Arctic where it is then deposited.

2.2. Sources outside the Arctic and atmospheric transport to the Arctic

Because of its remoteness, the arctic troposphere was long believed to be extremely clean. Arctic haze was described for the first time in the 1950s and in the 1970s it became obvious that anthropogenic sources outside the Arctic strongly contributed to pollution of the arctic atmosphere (see chapter 4). The haze phenomenon, accompanied by high levels of gaseous air pollutants (e.g., hydrocarbons, Solberg *et al*., 1996), has been observed regularly since then, especially in the lower troposphere, and is due to the special meteorological situation in the Arctic in winter and early spring (Shaw, 1995).

The extreme dryness of the arctic troposphere minimizes wet deposition, thus leading to a very long lifetime for aerosols in the Arctic in winter. Surfaces of constant potential temperature form closed domes over the Arctic, with minimum values in the arctic boundary layer (Klonecki *et al*., 2003). This isolates the lower troposphere from the rest of the atmosphere by a transport barrier, the so-called 'Arctic Front'. Meteorologists realized that in order to facilitate isentropic transport, a pollution source region must have the same low potential temperatures as the arctic haze layers (Carlson, 1981; Iversen, 1984; Barrie, 1986). This rules out most of the world's pollution source regions because they are too warm, and leaves northern Eurasia as the main source region for the arctic haze (Rahn, 1981; Barrie, 1986). There, the Arctic Front can be located as far south as 40° N on average in January (see Figure 4.1). Furthermore, northern Eurasia is on a preferred pathway into the polar dome that involves diabatic cooling of air traveling over snow-covered land. This transport is highly episodic and often related to large-scale blocking events (Raatz and Shaw, 1984; Iversen and Joranger, 1985). In contrast, air masses leaving North America's densely populated east coast are heated diabatically (Klonecki *et al*., 2003) because of the frequent warm conveyor belts over the downwind North Atlantic Ocean (Stohl, 2001; Stohl *et al*., 2002; Eckhardt *et al*., 2003). Southeast Asia is located at even higher potential temperatures than North America and so was also rejected as a source of arctic haze, although an Asian desert origin was suggested for the elevated haze layers (Rahn *et al*., 1977) that are also quite frequent (Leiterer *et al*., 1997).

Pollution transport to the Arctic varies considerably (see section 3.7.3 on the effects of natural climate variations on long-range transport to the Arctic). During the 'positive' phase of the North Atlantic Oscillation (NAO), transport from all three northern hemisphere continents (Europe, North America, and Asia in order of significance) into the Arctic is enhanced, resulting in higher arctic pollution levels (Eckhardt *et al*., 2003; Duncan and Bey, 2004). Given the long-term variability in the NAO, its status must be considered when studying arctic pollution trends (Macdonald *et al*., 2005).

Recently, new issues have attracted scientific interest. Anthropogenic emissions in southern and eastern Asia have grown rapidly over the past few decades, especially black carbon emissions. These are now much larger than in Europe, North America, and Russia combined. Black carbon is important because it absorbs solar radiation and leads to a possibly strong albedo reduction if deposited onto snow or ice (Hansen and Nazarenko, 2004). In a recent model study, Koch and Hansen (2005) suggested that

southern Asia is now the dominant source of black carbon in the arctic upper troposphere and is comparable to the European source near the arctic surface. However, if the thermodynamic argument holds that in winter it is virtually impossible for a southern Asian air mass to reach the arctic lower troposphere (Carlson, 1981; Iversen, 1984; Bowling and Shaw, 1992), then it is not clear how Asian black carbon, given black carbon's short atmospheric lifetime of 6±2 days (Park *et al.*, 2005), can intrude into the polar dome in the model of Koch and Hansen (2005).

There is also controversy regarding transport from the stratosphere to the arctic troposphere. It is known that tropopause folds, which are frequent at the Polar Front, also occur at the Arctic Front (Rao and Kirkwood, 2005). Several studies suggest a strong influence of transport from the stratosphere on the ozone concentrations in the arctic free troposphere (Dibb *et al.*, 2003; Allen *et al.*, 2003). Other studies suggest a much lower frequency of stratosphere-troposphere-transport events reaching the lower troposphere in the Arctic than at mid-latitudes (James *et al.*, 2003; Sprenger and Wernli, 2003), and their influence on surface ozone at Alert seems small (Dibb *et al.*, 1994). In fact, because potential temperatures in the lower stratosphere are higher than in pollution source regions at mid-latitudes, even more diabatic cooling is required for stratospheric air to penetrate the polar dome.

In a recent paper Stohl (2006) used the Lagrangian particle dispersion model FLEXPART to construct a global data set of 1.4 million continuous trajectories. At the model start, particles were distributed homogeneously in the atmosphere and were then transported for 5.5 years using both resolved winds from ECMWF (European Centre for Medium-range Weather Forecasts) analyses and parameterized turbulent and convective transport. Based on this data set, a climatology of transport in and to the Arctic was developed. It was found that the time air resides continuously north of 70° N – its 'arctic age' – is highest near the surface in the North American sector of the Arctic. North of 70° N and near the surface, the mean arctic age of air is about a week in winter and two weeks in summer (Figure 2.5). Arctic age decreases rapidly with altitude to about

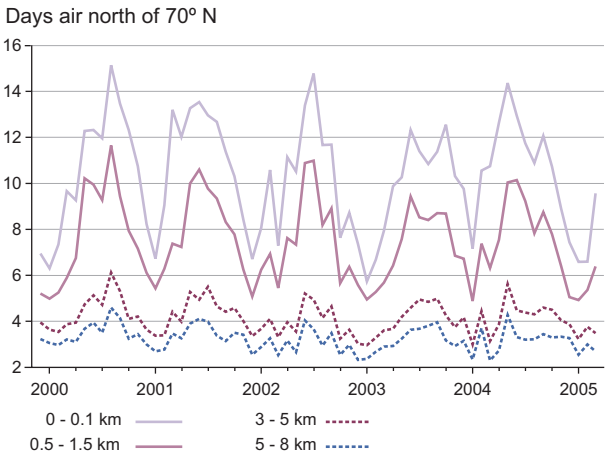


Figure 2.5. Time series of monthly mean arctic age of air, averaged over the central Arctic (north of 70° N) at different altitudes.

three days in the upper troposphere. In the most isolated regions of the Arctic, air is exposed to continuous darkness for, on average, 10 to 14 days in December (Figure 2.6). Transport from the stratosphere to the lower troposphere is much slower in the Arctic than at mid-latitudes. In the central Arctic, for instance, the probability that air near the surface was transported from the stratosphere within 10 days is only about 1% in winter and 0.3% in summer. Air pollution can be transported into the Arctic along three pathways: low-level transport followed by ascent in the Arctic, low-level transport alone, and uplift outside the Arctic, followed by descent in the Arctic. Only the last pathway is frequent for pollution originating from North America and Asia, whereas European pollution can follow all three pathways in winter, and pathways one and three in summer.

Sensitivities of arctic air masses to emissions of air pollutants were calculated for transport times of up to 30 days before the air masses reached the Arctic. They were highest over Siberia and Europe in winter and over the oceans in summer. Using an inventory for anthropogenic

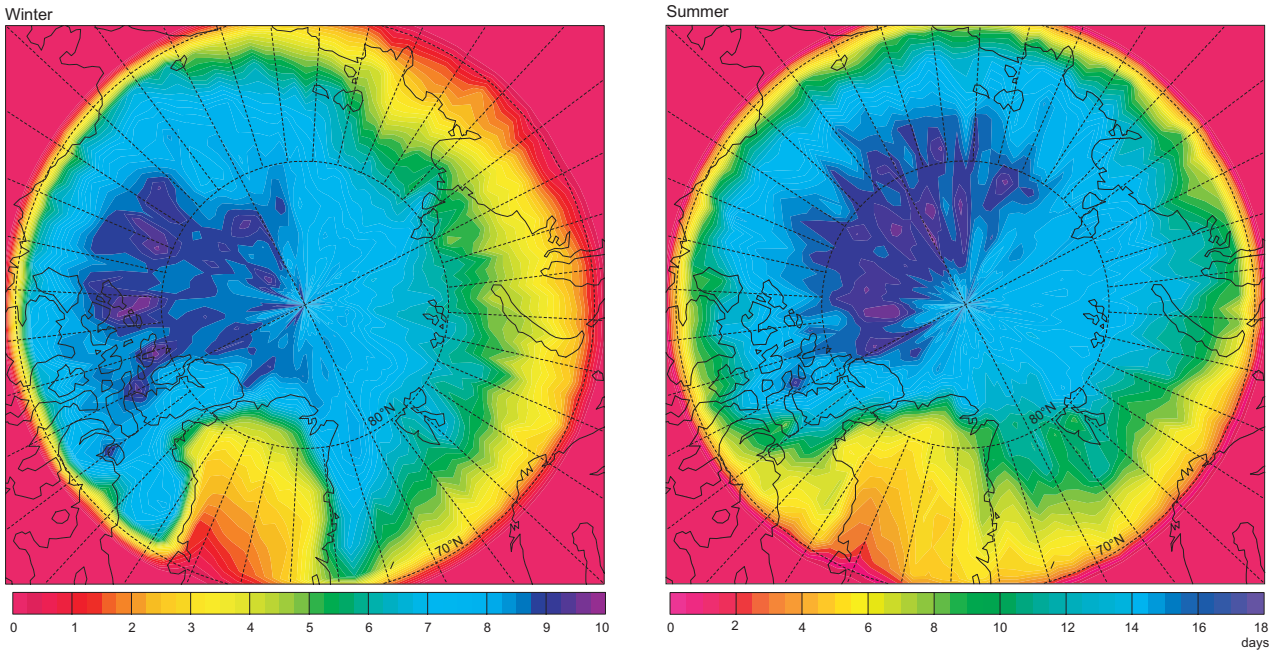


Figure 2.6. Mean arctic age of air in the lowest 100 m of the atmosphere in January (winter) and July (summer).

black carbon emissions, it was found that black carbon potential source contributions from Asia to the Arctic are much lower than those from central Europe and Russia (Figure 2.7), despite much higher emissions in Asia. The Eurasian origin of black carbon in the Arctic is also confirmed by the Danish Eulerian Hemispheric Model (DEHM) (Figure 2.8). For time scales of five and ten days, south Asian black carbon potential source contributions near the arctic surface are only 1.6 and 10% of the corresponding European values. Using an inventory for black carbon emissions from boreal and temperate forest fires, black carbon potential source contributions to the Arctic (particularly from fires in Siberia) were much larger than anthropogenic black carbon potential source contributions in summer (see also section 2.1.5). Boreal forest fires may even dominate the annual total black carbon budget in the Arctic in years of strong burning. Measurements of black carbon (or light absorption by aerosols) at the surface and aloft from aircraft combined with modeling studies such as that of Stohl (2006) are required to further define sources of black carbon to the Arctic.

2.3. Emissions estimates used in modeling

Several emissions areas in the northern hemisphere contribute to air pollution in the Arctic. Emissions scenarios for the model studies in this assessment were based on global emissions on a 1° x 1° grid from the EDGAR database for SO_2, NO_X, and VOC for 1990 and 1995. Emissions of ammonia (NH_3) and NO_X from lightning and soil emissions from the global GEIA (which provides gridded data, aggregated according to various categories at global or regional scales, from several inventories) were also used. Modified gridded EDGAR emissions were obtained for 2000, 2010, 2020, and 2030 based on IIASA emissions scenarios (Frank Dentener, European Commission Joint Research Centre, pers. comm.). For 2010 and 2020 there were two types of emissions scenarios: The 'Current LEgislation' (CLE) scenario, which reflects the current perspectives of individual countries on future economic development and takes into account the effects of presently agreed emission control legislation in the individual countries. The other scenario is the 'Maximum technically Feasible Reduction' (MFR) scenario. This projects the scope for emission reductions offered by full implementation of presently available emission control technologies, while maintaining the projected levels of anthropogenic activities. Global emissions are redistributed to the model grid, which is an extension of the old 150 km resolution EMEP grid. The global emissions scenarios are combined with the EMEP expert emissions of SO_2, NO_X, NH_3, and VOC for 1985, 1990, 1995 to 2003, and expert emissions estimates for 2010 and 2020 (see http://webdab.emep.int). Specific information on emissions from the Russian non-ferrous metal smelters is also used (see section 2.1.1).

Figure 2.9 shows emissions of SO_x-S and NO_X-N for 2000. Figure 2.10 projects emissions of SO_x-S for 2020 for the CLE and MRF emissions scenarios. For SO_x-S, a comparison of the two figures indicates that the CLE scenario results in only small changes in emissions, while the MRF scenario results in relatively large emissions reductions. This is also evident in Figure 2.11, which projects total emissions of both SO_x-S (95 % of which is SO_2) and NO_x-N between 1990 and 2020.

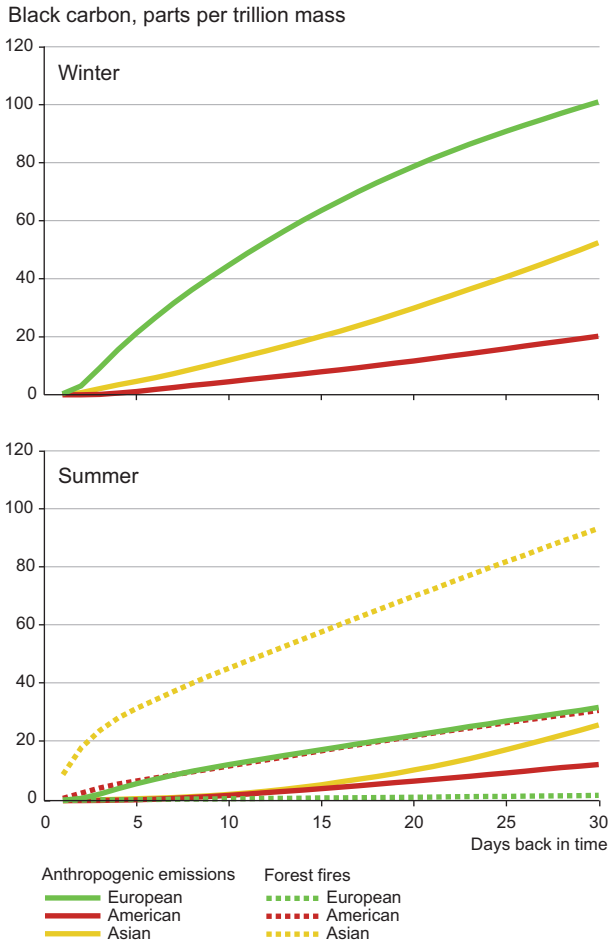


Figure 2.7. Black carbon potential source contributions from continents as a function of transport time to the Arctic, for the subset that also reaches a minimum arctic altitude below 1000 m, for winter and summer.

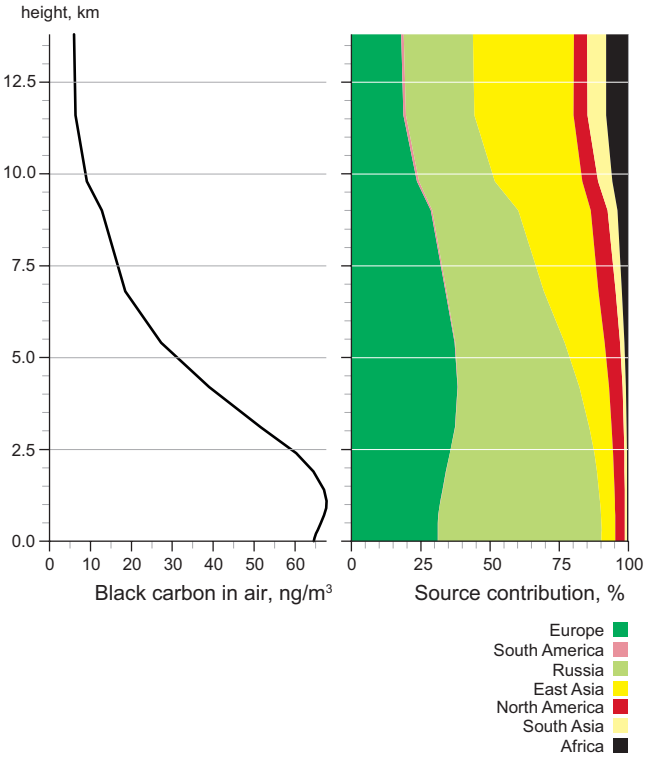


Figure 2.8. Concentration and origin profile of black carbon north of the Arctic Circle as calculated by the Danish Eulerian Hemispheric Model (see section 3.7). Average for 1991 to 2001.

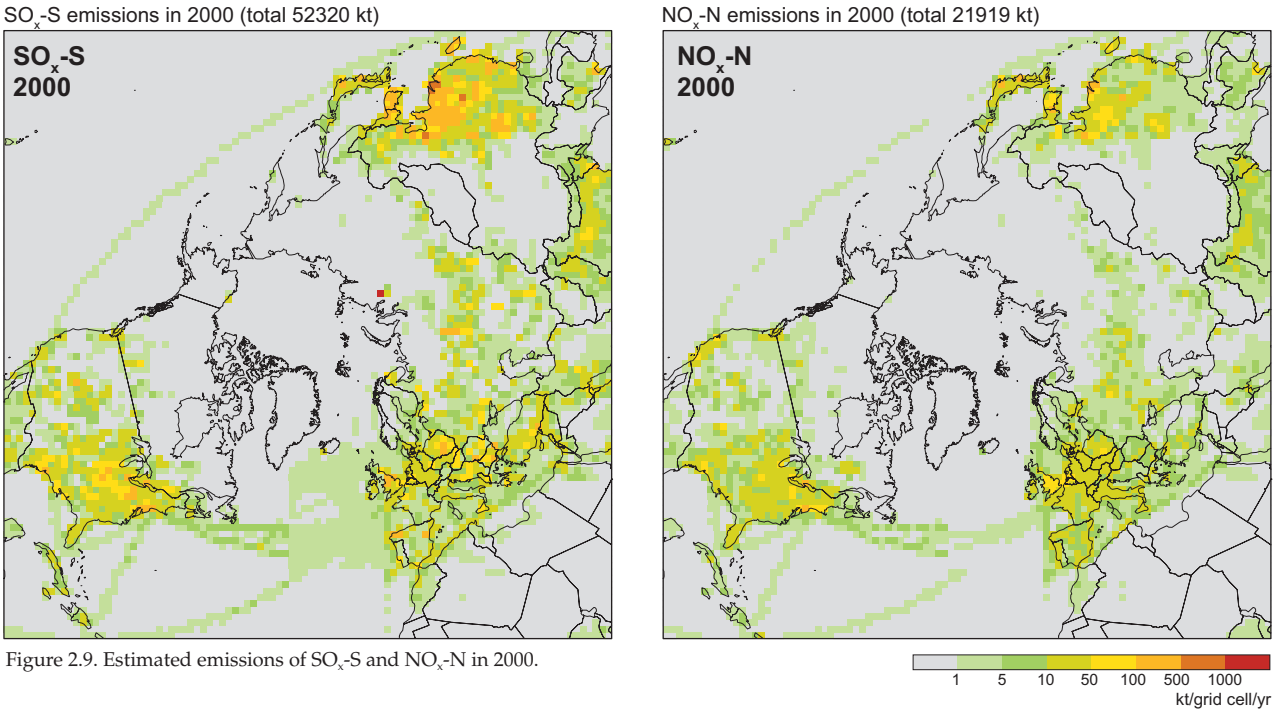


Figure 2.9. Estimated emissions of SO_x-S and NO_x-N in 2000.

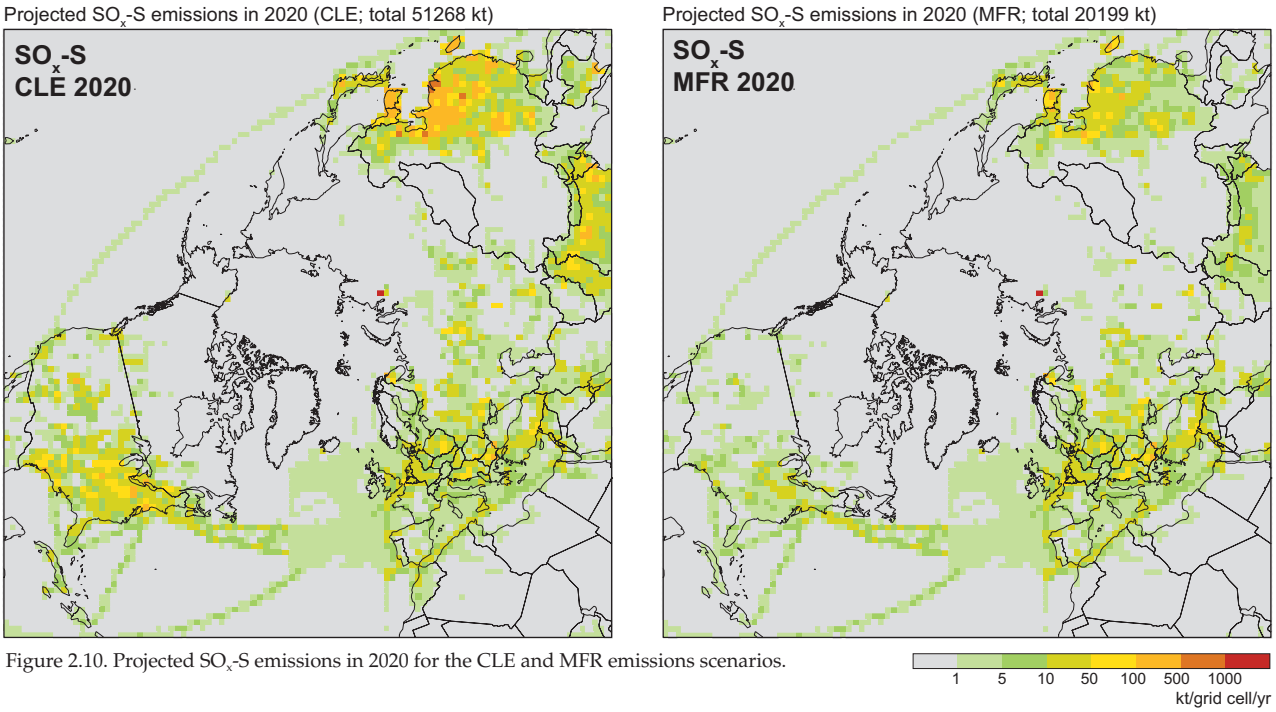


Figure 2.10. Projected SO_x-S emissions in 2020 for the CLE and MFR emissions scenarios.

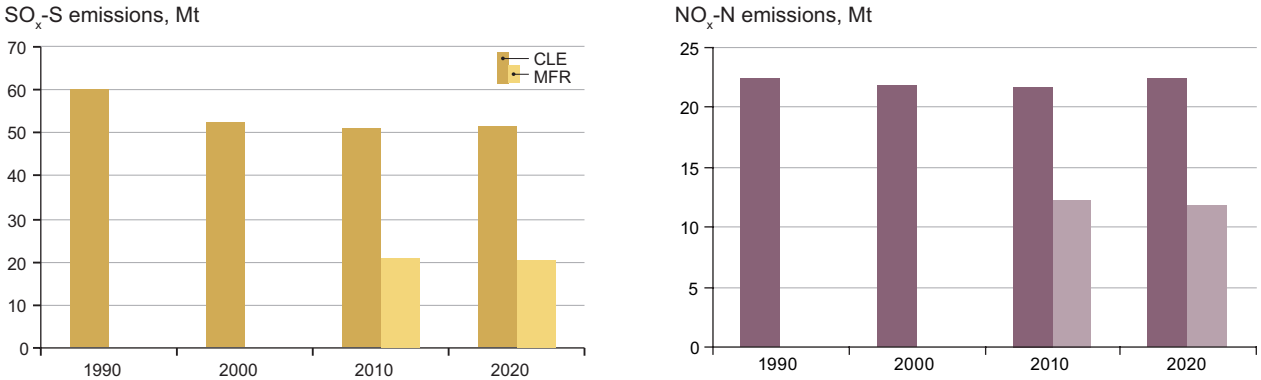


Figure 2.11. Projected total emissions of SO_x-S and NO_x-N between 1990 and 2020 for the area shown in Figure 2.10.

Chapter 3

Concentrations and Deposition of Acidifying Pollutants

Lars R. Hole, Jesper Christensen, Veronika A. Ginzburg, Vladimir Makarov, Natalia A. Pershina, Alla I. Polischuk, Tuija Ruoho-Airola, Peotr Ph. Svistov, and Vitaly N. Vasilenko

The first AMAP assessment on acidification in the Arctic (Kämäri, 1998) highlighted that although there are few sources of acidifying air pollutants within the Arctic, high-latitude ecosystems are particularly sensitive to pollution and that some effects of acidification are evident even in low deposition areas. Adverse effects of acidifying pollutants were noted in fish populations in acidified lakes and in forests and natural vegetation, including lichens. The assessment described the few but significant point sources of acidifying air pollutants in Norilsk (on the Taymir Peninsula in northern Siberia) and on the Kola Peninsula. These were the areas of greatest concern, in addition to the northern regions of Norway and Finland. The assessment focused on sulfur as this was considered to dominate the acidification issue.

The assessment concluded that there had been no trends in atmospheric concentrations of acidifying compounds in either Canada or Alaska since the early 1980s, but that there were decreasing trends on Svalbard. There were no background data for Russia. It was considered that around 75% of the deposition could occur in the form of dry deposition, but there were insufficient observations to confirm this. Model results for sulfur dioxide (SO_2) and sulfate (SO_4) compared well with observations for times series at Station Nord (Greenland) and for long-term averages at several EMEP stations. The assessment concluded by recommending that air chemistry stations be established in Alaska and eastern Russia, and that further resources were required to support model verification (inter-comparison), particularly in relation to dry deposition.

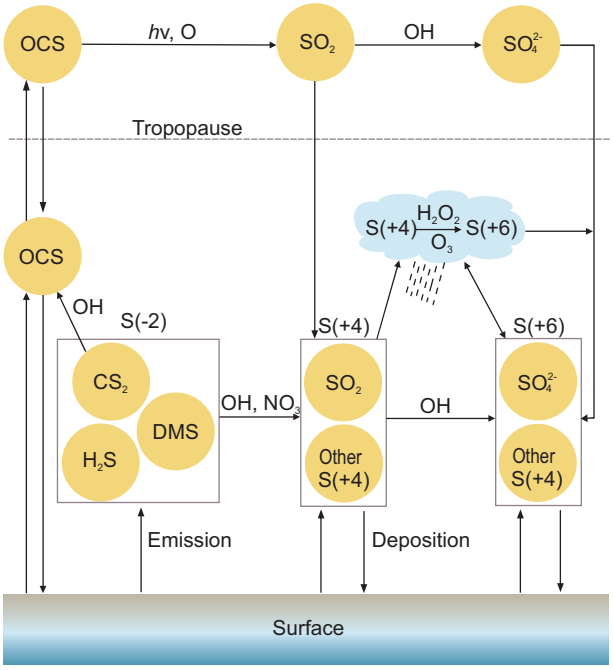


Figure 3.1. Atmospheric sulfur cycle (Berresheim *et al.*, 1995).

3.1. Atmospheric and transport processes for air pollutants in the Arctic

The atmosphere is an effective oxidizing medium, where the emitted sulfur and nitrogen compounds readily undergo reactions to oxidize further to sulfate and nitrate. Both gas phase and liquid phase reactions in the atmosphere are important routes to the acidic sulfate and nitrate products. The oxidized sulfur and nitrogen compounds can exist in gaseous form, bound in particles, or dissolved in cloud droplets and rainwater. The atmospheric chemistry associated with the acid deposition process, especially the part starting with nitrogen oxides, is complex.

Emitted oxides and their reaction products are transported with the airflows and simultaneously removed from the atmosphere by dry and wet deposition processes. Dry deposition rates of gaseous and particulate compounds are controlled by physical and chemical characteristics of the compounds, the properties of the ground surface, and the state of the atmospheric boundary layer (Sehmel, 1980; Wesely and Hicks, 2000). Wet deposition is highly affected by the amount of precipitation, but also by the type of aqueous phase (e.g., liquid water, snow, or ice). Wet deposition removes the acid components from the atmosphere by several processes, which occur both within and below clouds. Although atmospheric lifetimes of SO_2, nitrogen oxides (NO_X), and their oxidation products are of the order of a few days (Schwarz, 1979; Levine and Schwarz, 1982; Logan, 1983), in the High Arctic during winter the atmospheric half-life of sulfate has been reported to reach as much as two weeks or more (Barrie, 1986). Transport distances range from hundreds to thousands of kilometers (Seinfeld and Pandis, 1998). Thus, besides the actual emissions, many additional factors affect the observed concentrations and trends of the compounds involved in the acid deposition process, including their relative concentrations in the atmosphere, the reversible nature of some reactions, and meteorological conditions.

3.1.1. Sulfur

Figure 3.1 shows schematically the atmospheric cycle for sulfur compounds. H_2S, DMS, CS_2, and OCS are sulfidies. The reactions are controlled by many factors, both chemical and physical, for example the concentrations of oxidizing agents (elemental oxygen, O; OH radicals; and ozone, O_3) or photons (*hv*), concentration ratios of different air pollutants, and the acidity of liquid water associated with particles and humidity (Seinfeld and Pandis, 1998). The atmospheric chemistry of the sulfur cycle is dominated by OH radical reactions in the gas phase which lead to the production of gaseous sulfuric acid (H_2SO), and by gaseous and aqueous phase reactions between SO_2 and hydrogen peroxide (H_2O_2) and O_3. The lack of sunlight in the Arctic for large parts of the year (see section 2.2 on arctic age of air) limits the production of the OH radical and hydrogen peroxide (Barrie, 1986). The seasonality of SO_2 oxidation to

sulfate (SO_4^{2-}) is important in prolonging the presence of sulfate aerosols in the Arctic into April and May (Barrie and Hoff, 1984; Barrie, 1996). S(-2), S(+4) and S(+6) in Figure 3.1 illustrate different oxidation levels of sulfur.

3.1.2. Nitrogen

Figure 3.2 shows schematically the atmospheric cycle for nitrogen compounds. Important nitrogen-containing compounds are molecular nitrogen (N_2), nitrous oxide (N_2O), nitric oxide (NO), nitrogen dioxide (NO_2), nitric acid (HNO_3), inorganic and organic nitrates (NO_3^-), ammonium (NH_4^+), and ammonia (NH_3). The same oxidizing agents as for sulfur control the reactions.

The previous AMAP assessment (AMAP, 1998) reported that, 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 has a considerable 'dark' component, with night-time reactions between NO_2 and O_3 to form NO_3 and N_2O_5 (nitrogen pentoxide) (Seinfeld and Pandis, 1998). The reaction of N_2O_5 with water produces gas phase nitric acid (HNO_3) and particulate nitrate. The formation of the peroxy acetyl nitrate (PAN) 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 to 80% of the airborne oxidized nitrogen (NO_y) (Bottenheim *et al.*, 1993; Singh *et al.*, 1992).

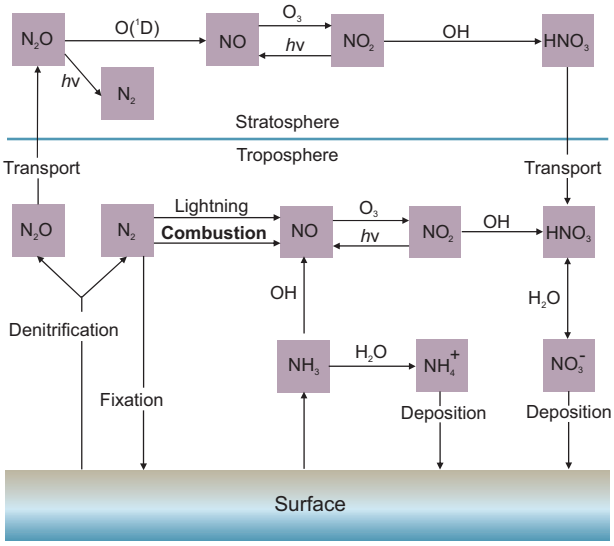


Figure 3.2. Atmospheric nitrogen cycle (Seinfeld and Pandis, 1998). The important anthropogenic nitrogen oxide emissions are illustrated as combustion, which oxidizes molecular nitrogen (N_2) to nitric oxide (NO), and further oxidation by ozone (O_3) to NO_2.

3.2. Distribution of monitoring stations

The AMAP atmospheric monitoring network consists of a number of stations distributed throughout the Arctic. Most of these are EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air pollutants in Europe) stations that also report to the AMAP database at NILU (Norwegian Institute for Air Research). In addition, a few national stations report data to AMAP. Some stations have reported data since the mid-1970s. Figure 3.3 and Table 3.1 identify the stations for which data have been used in this assessment. Most stations are located in the European Arctic. Very few measurements from North America were available for this assessment (data were available from Snare Rapids, Alert, and Barrow). The only station in northern Greenland (Station Nord) was closed in 2002.

The EMEP network was established to support the 1979 Geneva Convention on Long-range Transboundary Air Pollution (LRTAP). The LRTAP Convention provides a broad framework for cooperative action on controlling and reducing the impact of transboundary air pollution and establishes a process for negotiating measures to control emissions of air pollutants through legally binding

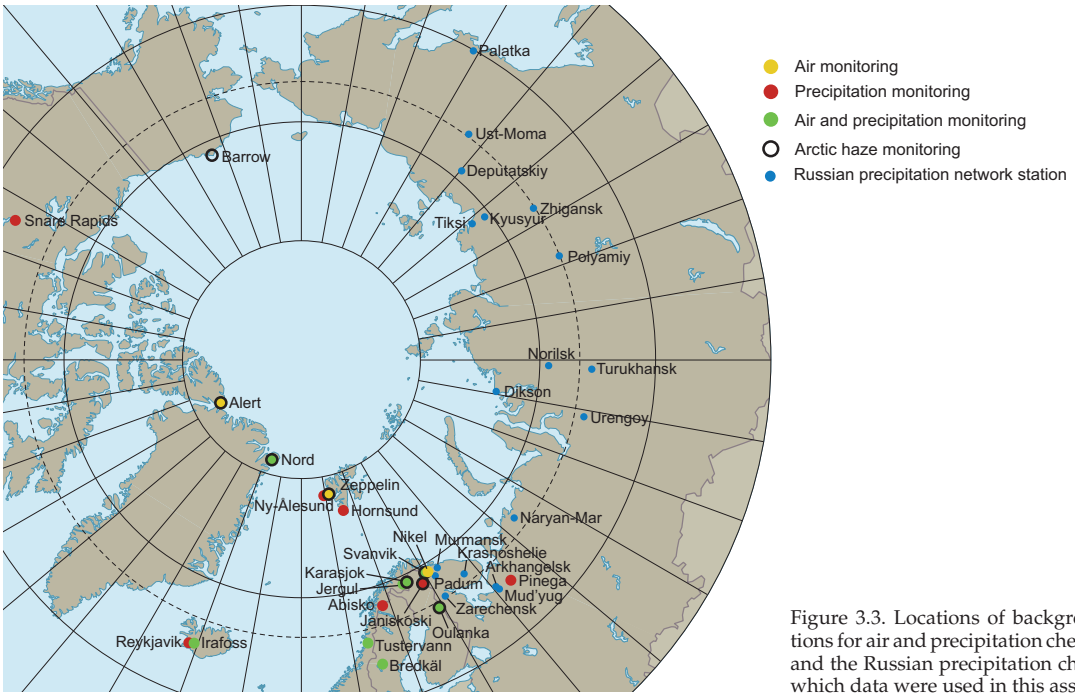


Figure 3.3. Locations of background monitoring stations for air and precipitation chemistry, and arctic haze and the Russian precipitation chemistry stations from which data were used in this assessment.

Table 3.1. AMAP stations included in this assessment.

Station name	Latitude	Longitude	Media[a]	Operating period	Height above sea level (m)
Alert	82° 28′ N	62° 30′ W	A	1980-	210
Nord	81° 36′ N	16° 40′ W	A + P	1990-2002	20
Oulanka	66° 19′ N	29° 24′ E	A + P	1990-	310
Irafoss	64° 5′ N	21° 1′ W	A + P	1980-	61
Reykjavik	64° 8′ N	21° 54′ W	P	1994-	61
Jergul (with Karasjok)	69° 27′ N	24° 36′ E	A + P	1977-1997	255
Tustervann	65° 50′ N	13° 55′ E	A + P	1977-	439
Zeppelin (Ny-Ålesund)	78° 54′ N	11° 53′ E	A	1989-	474
Svanvik	69° 27′ N	30° 2′ E	A + P	1986-	30
Karasjok (with Jergul)	69° 28′ N	25° 13′ E	A + P	1997-	333
Ny-Ålesund	78° 55′ N	11° 55′ E	P	1982-	1
Janiskoski	68° 56′ N	28° 51′ E	P	1990-	118
Pinega	64° 42′ N	43° 24′ E	P	1979-	28
Bredkäl	63° 51′ N	15° 20′ E	A + P	1979-	404
Abisko	68° 21′ N	18° 49′ E	P	1992-	390
Nikel	69° 24′ N	30° 12′ E	A		
Snare Rapids	63° 31′ N	116° 00′ W	P		
Barrow	71° 19′ N	156° 40′ W			
Hornsund	77° 00′ N	15° 33′ E	P	1988-	7

[a] A: air; P: precipitation

protocols. Within this process, the main objective of the EMEP program is to provide signatories and subsidiary bodies to the LRTAP Convention with qualified scientific information to support the development and further evaluation of the international protocols on emission reductions negotiated within the convention. The EMEP program is undertaken in collaboration with a broad network of scientists and national experts that contribute to the systematic collection, analysis, and reporting of emission data, measurement data, and integrated assessment results. Since 2003, EMEP has had stations in 35 countries, varying from one to 25 stations in each country.

The Acid Deposition Monitoring Network in East Asia – EANET – was established in 1998. Twelve countries participate in the network, which stretches from the Russian–Mongolian border to Indonesia. Regular monitoring activity under EANET began in 2001. The EANET monitoring program includes, among others, measurements of SO_2, NO_2, and O_3 in air; acidity, electrical conductivity, sulfate, and nitrate in precipitation; soils and vegetation, and freshwater systems. Measurements are made at urban, regional, and remote sites. Data from Mondy, the Russian remote monitoring site situated near the Russian–Mongolian border, can be used to estimate long-range atmospheric transport from East Asia to the Arctic.

In 2002, 24 stations reported data relevant to acidification and eutrophication to the AMAP database at NILU. Most of these stations are located in Fennoscandia (see Table 3.1).

The Russian national monitoring network for precipitation chemistry has 130 monitoring stations. Precipitation samples are analyzed at twelve regional analytical laboratories for between eight and eleven compounds, including SO_4^{2-}, Cl^-, NO_3^-, HCO_3^-, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, the sum of ions, and pH. In addition to these stations, there are 105 monitoring sites where pH only is analyzed. The monitoring stations are unevenly distributed, with less than 40% situated in the vast Siberian region. Data have been recorded for up to 45 years at some stations. This assessment uses data from 18 monitoring stations within the Russian Arctic (see Table 3.2). The Russian Arctic may be sub-divided into three sectors on the basis of climate: the Atlantic, Siberian, and Pacific sectors.

Monitoring data from the Zarechensk, Padun, Krasnoshelie, Naryan-Mar, Urengoy, Turukhansk, Zhigansk, Deputatskiy, Ust-Moma and Palatka sites were used to represent background levels. Except for the two EMEP-stations reported here (Janiskoski and Pinega), there are no monitoring sites in the Russian Arctic representing regional or background conditions for air quality. Urban monitoring sites in the industrial areas of five Russian cities – Nikel, Monchegorsk, Murmansk, Salekhard and Norilsk – were used to estimate pollutant levels in air at the nearest industrial center.

Pollutant levels and acidity of snow cover in the Russian Arctic are monitored at 99 stations.

Table 3.2. Russian precipitation stations.

Station name	Start	Latitude	Longitude
Atlantic sector			
Zarechensk	1990	66.7° N	31.4° E
Padun	1991	68.6° N	31.8° E
Murmansk	1991	69.0° N	33.1° E
Krasnoshelie	1990	67.4° N	37.1° E
Mud'yug	1958	64.9° N	40.3° E
Arkhangelsk	1991	64.6° N	40.5° E
Naryan-Mar	1962	67.7° N	53.0° E
Siberian sector			
Urengoy	1989	66.0° N	78.4° E
Dikson	1980	73.5° N	80.4° E
Turukhansk	1962	65.8° N	87.9° E
Norilsk	1991	69.3° N	88.3° E
Polyarniy	1990	66.7° N	112.4° E
Zhigansk	1991	66.8° N	123.4° E
Kyusyur	1990	70.6° N	128.0° E
Tiksi	1990	71.6° N	128.9° E
Pacific sector			
Deputatskiy	1990	69.3° N	139.7° E
Ust-Moma	1990	66.5° N	143.2° E
Palatka	1962	60.1° N	150.9° E

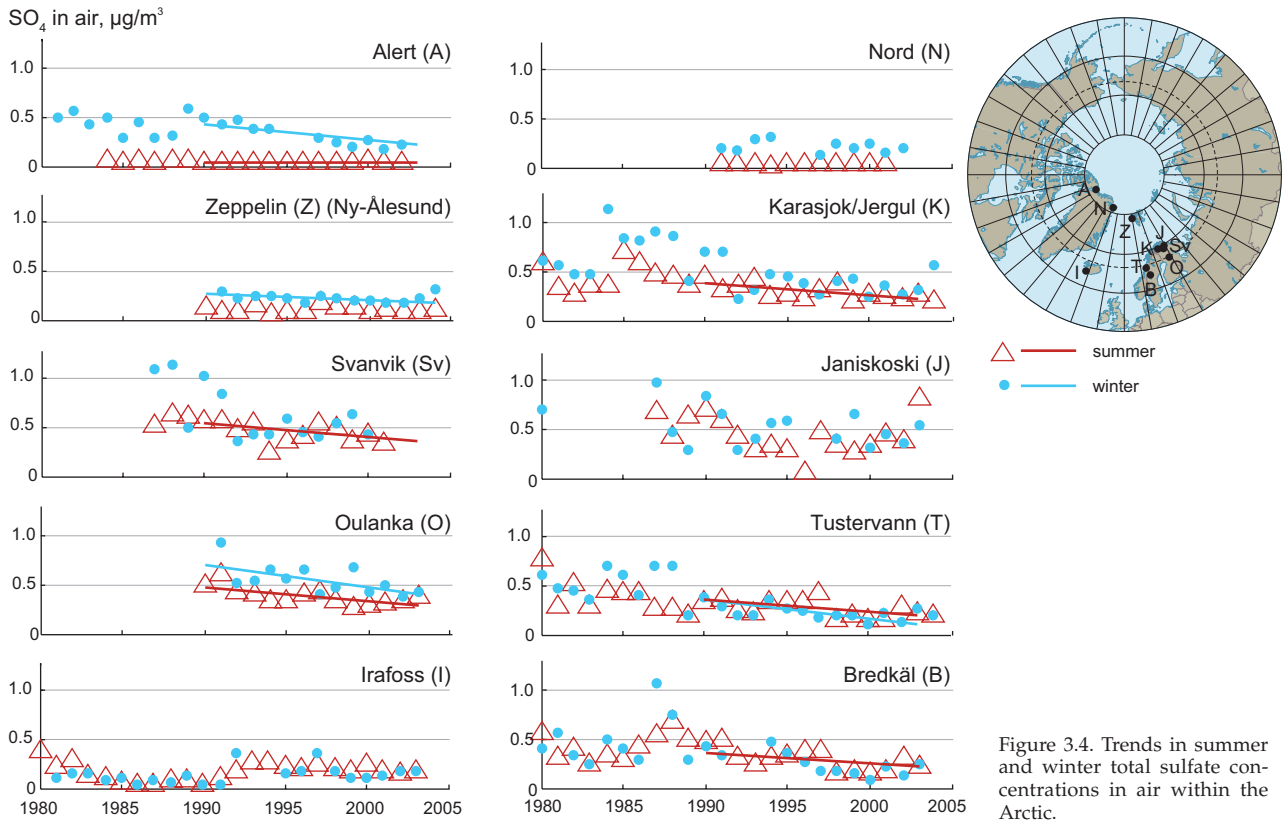


Figure 3.4. Trends in summer and winter total sulfate concentrations in air within the Arctic.

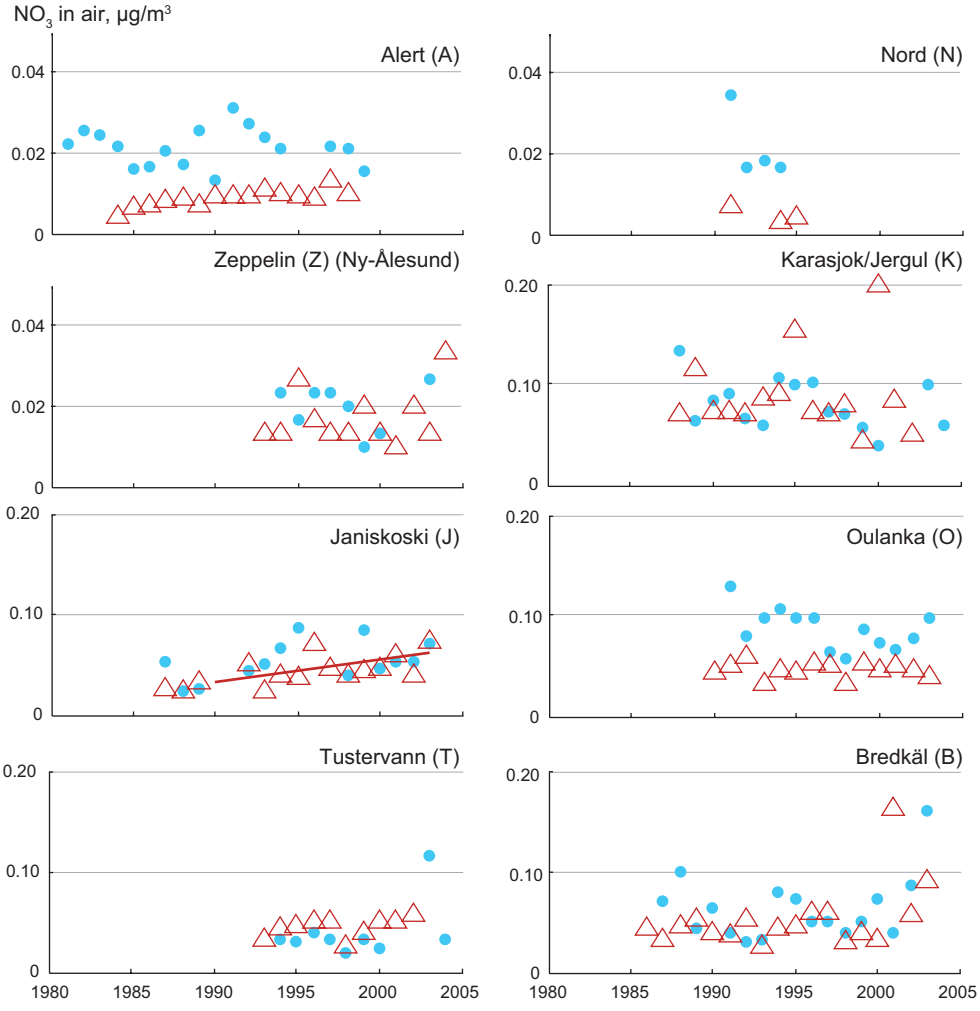


Figure 3.5. Trends in summer and winter nitrate concentrations in air within the Arctic.

3.3. Concentrations, distribution, and trends in air and precipitation

There are few good time series for the main atmospheric compounds in the High Arctic. There are also few stations that monitor both air and precipitation. Precipitation and air chemistry data from the AMAP monitoring stations are presented in this assessment as average winter (December, January, February) and summer (June, July, August) values. In cases where a monthly value was missing, the whole season was omitted from the analysis. Precipitation data were weighted according to precipitation amount. Existence of a monotonic increasing or decreasing trend in the time series after 1990 was tested using the non-parametric Mann-Kendall test at significance levels of $p<0.1$, $p<0.05$, and $p<0.001$ as a two-tailed test (Gilbert, 1987). The estimate for the slope of a linear trend was calculated using the non-parametric Sen's method (Sen, 1968). The Mann-Kendall test is suitable for cases where the trend may be assumed to be monotonic, and thus with no seasonal cycle or cycle of any other type present in the data. Missing values are allowed in the Mann-Kendall test and the data need not conform to any particular distribution. Sen's slope estimator is the median of the slopes calculated from all pairs of values in the data series. The Sen's method is not greatly affected by data outliers and can be used when data are missing (Salmi *et al.*, 2002).

3.3.1. Air

Figures 3.4 and 3.5 show time series for seasonal SO_4 and NO_3 concentrations in air for the stations with the best air quality time series in the Arctic. Table 3.3 summarizes the trend statistics for these datasets as well as for SO_2 and NH_4 in air. Significant downward trends are evident for SO_4 and SO_2 in air at many stations, both in summer and winter. Interestingly, it is only in summer that a significant reduction in sulfate levels in air is seen in Svanvik (but not Nikel) despite emissions in the area having been significantly reduced. For NO_3 and NH_4 there is no clear pattern, although NH_4 is decreasing at some stations.

An annual air quality study is undertaken in the border areas between Norway and Russia (Hagen *et al.*, 2005). Figure 2.1 showed the annual average SO_2 concentrations in air at Svanvik together with the total annual SO_2 emissions from the non-ferrous metal smelters at Nikel. Although the decreasing trend in air concentration after 1990 is not statistically significant, reductions have occurred since 1980. Despite a flat or downward trend in SO_2 concentrations in air at most sites, concentrations at Svanvik are still relatively high and periodic peaks in concentration ('episodes') well above the recommended health limit (an hourly value of 350 µg/m^3) are still common (Hagen *et al.*, 2005). It is obvious that these high values can largely be attributed to the non-ferrous metal smelters in Nikel (Figure 3.6).

3.3.2. Precipitation

3.3.2.1. General pattern

A decreasing trend in annual average sulfate concentrations is also evident in the precipitation data (Figure 3.7). The data also show seasonal differences. At Ny-Ålesund (Svalbard) there are higher concentrations in winter, while at Zeppelin (approximately 1 km from Ny-Ålesund) the seasonal difference in SO_4 concentrations in air is smaller.

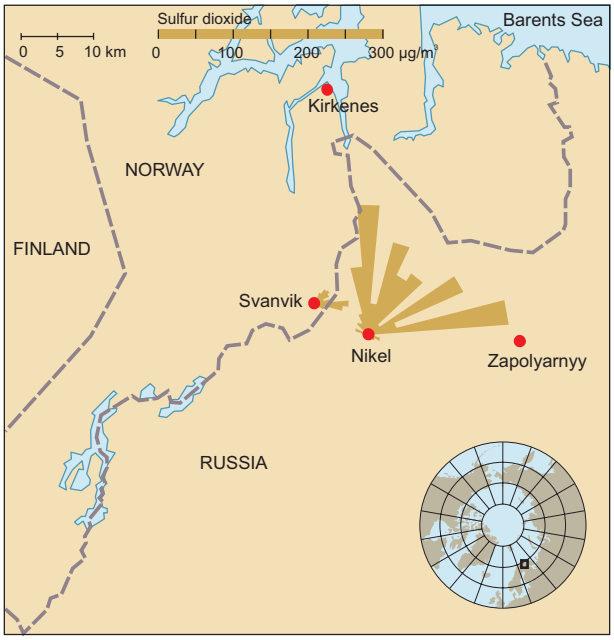


Figure 3.6. Mean concentrations of SO_2 in air at Svanvik and Nikel for different wind sectors for October 2004 to March 2005 (Hagen *et al.*, 2005).

Table 3.3. Trend statistics for acidifying pollutants in arctic air, 1990–2003.

	Summer				Winter			
	SO_2	SO_4	NO_3	NH_4	SO_2	SO_4	NO_3	NH_4
Alert	NA	*-			NA	***-		**-
Oulanka	**-	**-				**-		
Irafoss	NA		NA	NA	NA		NA	NA
Janiskoski		NA	*+				NA	
Pinega							**+	
Nikel		NA	NA	NA		NA	NA	NA
Tustervann		**-			*-	*-		
Zeppelin (Ny-Ålesund)				*-	**-	**-		*-
Svanvik		*-						
Karasjok/Jergul	**-	**-		**-				**-
Bredkäl	**-	*-			**-	**-		
Nord			NA		**-		NA	

NA: not available; significance level: * P<0.1, ** P<0.05, *** P<0.001; +/- indicate positive and negative trends respectively; an empty cell indicates no significant trend.

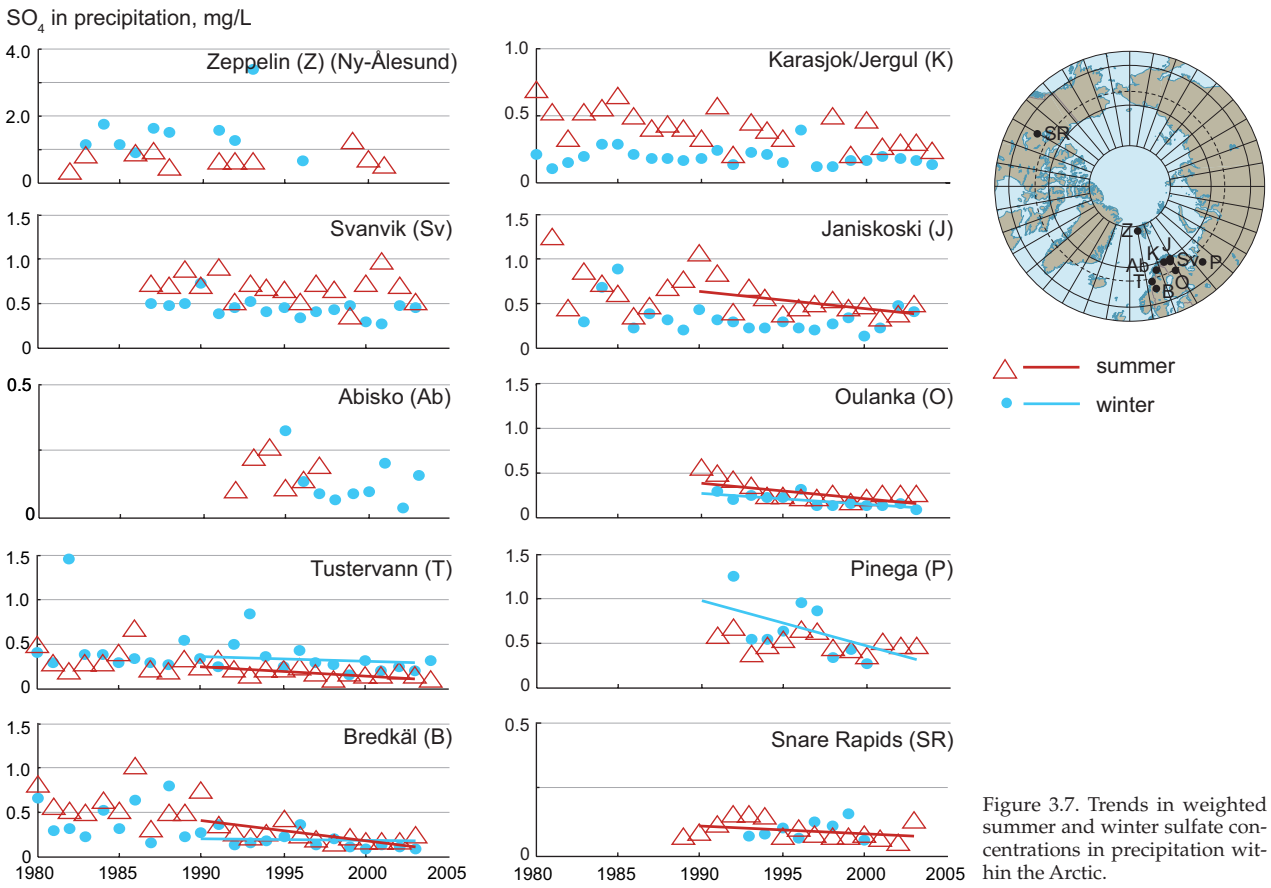


Figure 3.7. Trends in weighted summer and winter sulfate concentrations in precipitation within the Arctic.

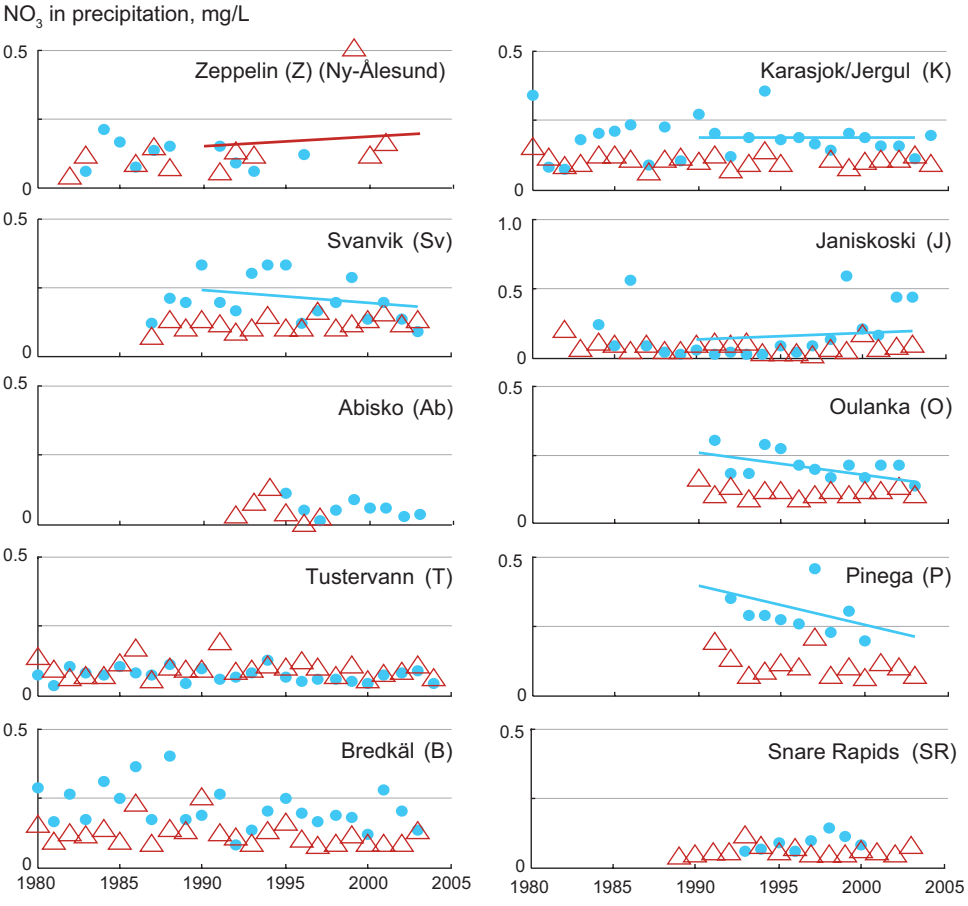


Figure 3.8. Trends in weighted summer and winter nitrate concentrations in precipitation within the Arctic.

It is also interesting to note that the significant increase in pH at most stations is not necessarily coupled to a significant decrease in SO_4 concentrations (Table 3.4). There is a notable marine sulfur contribution at several sites, in particular at Ny-Ålesund (not shown).

There is no consistent pattern for NO_3 in precipitation (Figure 3.8) and for the summer data the only significant increasing trend is at Ny-Ålesund, although the data for this site are sparse. More observations for nitrate both in air and precipitation are required to better understand the development of nitrate pollution in the Arctic.

3.3.2.2. **Russian Arctic**

A significant amount of information on acidifying pollutants in precipitation is now available for the Russian Arctic. The Russian Arctic is usually subdivided on the basis of climatic conditions into the Atlantic, Siberian, and Pacific sectors. Long-term data show that these regions also differ in terms of precipitation chemistry and acidity. Trend statistics for a number of Russian stations are shown in Table 3.5. Trends are compared for the period since 1990, although some measurements started as early as 1958 (Table 3.2).

Table 3.4. Trend statistics for acidifying pollutants in arctic precipitation, 1990–2003.

	Summer						Winter					
	Precip.	pH	SO_4	SO_4*	NO_3	NH_4	Precip.	pH	SO_4	SO_4*	NO_3	NH_4
Oulanka	*-		**-	**-					**-	**-	*-	*-
Irafoss		*+			NA	NA		*+		*-	NA	NA
Janiskoski			**-	**-						**-	**+	
Pinega		*+							*-	*-	**-	**-
Tustervann		**+	***-	***-				***+	*-	***-		
Karasjok/Jergul		**+						***+		**-	*-	*-
Svanvik		**+						**+		*-	*-	
Zeppelin (Ny-Ålesund)		**+			*+							
Bredkäl		***+	**-	*-		**+			**-	**-		**+
Abisko									*-			
Snare Rapids	**+		**-									
Hornsund			NA	NA	NA		*-	*+	NA	NA	NA	NA

SO_4*: non-marine sulfate; NA: not available; significance level: * P<0.1, ** P<0.05, *** P<0.001; +/- indicate positive and negative trends respectively; an empty cell indicates no significant trend.

Table 3.5. Trend statistics for Russian precipitation stations. Only significant trends are shown.

		Winter			Summer		
		Mann-Kendall statistic	*p*-value	Sen-slope	Mann-Kendall statistic	*p*-value	Sen-slope
Atlantic sector							
Zarechensk	Precipitation				-1.831	0.067	-1.678
	pH				2.337	0.019	0.050
	NH_4-N				-1.736	0.083	-0.010
Padun	NH_4-N	-1.960	0.050	-0.019	-1.870	0.062	-0.010
Murmansk	SO_4-S	-2.741	0.006	-0.252	-2.303	0.021	-1.538
	NO_3-N	-2.141	0.032	-0.030	-1.916	0.055	-0.993
	NH_4-N	-3.558	0.000	-0.088	-2.901	0.004	-0.347
Krasnoshelie	-						
Mud'yug	SO_4-S	-1.831	0.067	-0.230			
Arkhangelsk	Precipitation	1.952	0.051	0.968			
	SO_4-S	-2.873	0.004	-0.198			
	NO_3-N	-1.650	0.099	-0.022			
	NH_4-N	-2.440	0.015	-0.035			
Naryan-Mar	pH	2.146	0.032	0.038	2.198	0.028	0.043
	SO_4-S	-2.326	0.020	-0.043			
	NH_4-N	2.623	0.009	0.043			
Siberian sector							
Urengoy	Precipitation	-2.562	0.010	-2.588	-1.830	0.067	-4.003
	pH	1.837	0.066	0.071	-1.877	0.061	-0.059
	SO_4-S				-1.783	0.075	-0.032
Dikson	SO_4-S	-2.728	0.006	-1.958	-2.524	0.012	-0.248
	NO_3-N				1.788	0.074	0.012
	NH_4-N				-2.180	0.029	-0.052
Turukhansk	Precipitation				-2.728	0.006	-1.958
Norilsk	-						
Zhigansk	-						
	pH				2.658	0.008	0.040
Tiksi	pH				1.930	0.054	0.909
	SO_4-S				1.930	0.054	0.082
Pacific sector							
Deputatskiy	-						
Ust-Moma	SO_4-S	2.573	0.010	0.068			
Palatka	-						

Atlantic sector

The average monthly SO_4-S concentration in precipitation in the Atlantic sector of the Russian Arctic for 1990 to 2004 was 1.8 ± 1.6 mg/L. Concentrations did not exceed 2.0 mg/L in more than 70% of samples. The highest monthly concentration in the industrialized regions (Murmansk, Arkhangelsk, Mud'yug) was 23.3 mg/L. In background areas (Zarechensk, Padun, Krasnoshelyie, and Naryan-Mar) concentrations ranged from 0.3 to 0.8 mg/L.

The average monthly NO_3-N concentration in the Atlantic sector for 1990 to 2004 was 0.31 ± 0.3 mg/L. In 62% of samples concentrations did not exceed 0.3 mg/L. The highest monthly concentration was 3.61 mg/L. Concentrations at background stations (Zarechensk, Padun, Krasnoshelyie) were between 0.1 to 0.7 mg/L in more than 85% of samples. The minimum nitrogen concentration corresponded to the maximum amount of precipitation. The average monthly concentration of NH_4-N in precipitation was 1 ± 0.71 mg/L. Concentrations of NH_4-N in precipitation did not exceed 1.4 mg/L in more than 86% of samples. The highest monthly concentration in precipitation was 16.6 mg/L.

The distribution of average monthly pH values in precipitation is shown in Figure 3.9. The most frequent pH values fell in the range 4.6 to 6.7 and their distribution mostly follows a normal distribution curve. The average pH value was close to 5.6. Values of 3.6 occurred in two samples only, while values above 7.4 occurred in 27 samples. Acidity was highest during the spring and summer. Precipitation became less acidic from west to east. In contrast to the Atlantic sector as a whole, for which precipitation is mostly neutral, precipitation at the Murmansk station was mostly acidic (ranging from 3.2 to 6.5). Over 80% of precipitation events at Murmansk had a pH of less than 5.4.

The total amount of monthly precipitation in the Atlantic region varied from 25 to 78 mm, with the greatest quantities falling during spring and autumn. An inverse correlation was observed between pollutant levels and the total amount of precipitation per month.

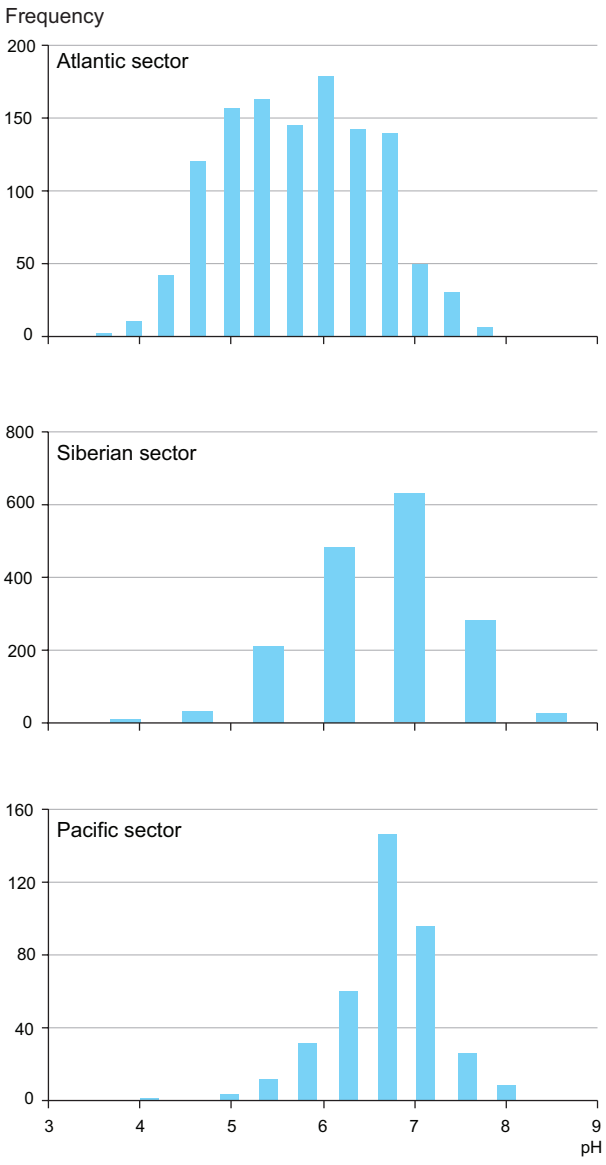


Figure 3.9. Frequency distribution for monthly pH values in precipitation within the Russian Arctic for the Atlantic, Siberian, and Pacific sectors for 1990 to 2004.

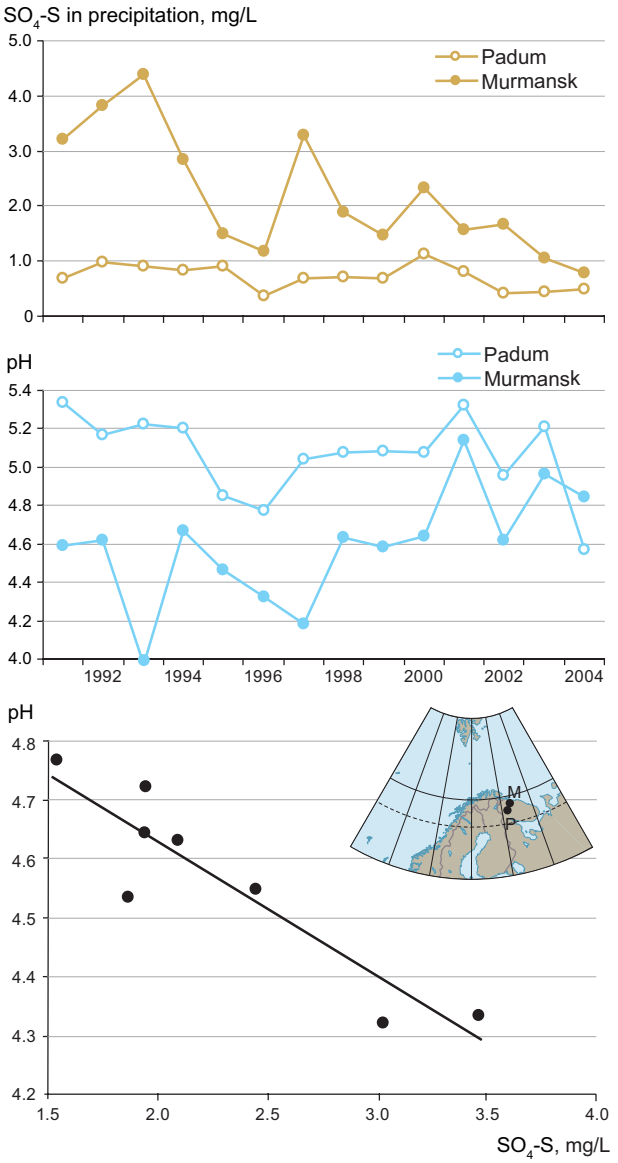


Figure 3.10. Annual average sulfate sulfur concentrations and pH in precipitation within the Russian Arctic at a heavily impacted site (Murmansk) and a background site (Padun) since 1991, and the correlation between sulfur and pH in samples from Murmansk.

Table 3.6. Concentrations of sulfur (mg/L), nitrogen (mg/L) and pH at background monitoring stations in the Russian Arctic, 1990-2004.

	Sulfur (SO_4)			Nitrogen (NO_3)			Nitrogen (NH_4)			ΣN	$N(NH_4)/N(NO_3)$	S/ ΣN	pH		
	average	min	max	average	min	max	average	min	max				average	min	max
Winter (D, J, F)															
Atlantic	0.43	0.020	1.40	0.16	0.01	0.68	0.35	0.03	1.21	0.51	2.19	0.84	5.70	4.35	6.60
Siberian	0.60	0.120	2.49	0.13	0.01	0.61	0.55	0.02	3.20	0.68	4.23	0.88	6.10	4.90	7.10
Pacific	0.68	0.230	2.23	0.14	0.01	0.67	0.56	0.04	2.62	0.70	4.00	0.97	6.50	5.10	7.50
Summer (J, J, A)															
Atlantic	0.41	0.040	1.67	0.07	0.00	0.42	0.28	0.04	1.25	0.35	4.00	1.17	5.63	4.64	6.59
Siberian	0.40	0.130	1.49	0.12	0.01	0.34	0.35	0.01	1.70	0.47	2.92	0.85	6.20	5.20	7.20
Pacific	0.84	0.200	4.00	0.10	0.02	0.33	0.37	0.06	1.62	0.47	3.70	1.79	6.40	5.60	7.10
Year (12 months)															
Atlantic	1.10	0.02	1.78	0.40	0.00	0.59	0.80	0.00	1.36	1.20	2.00	0.92	5.50	4.20	7.10
Siberian	0.70	0.12	2.65	0.20	0.01	0.68	0.30	0.01	3.20	0.50	1.50	1.40	6.00	4.70	7.30
Pacific	0.80	0.20	3.88	0.25	0.01	1.73	0.80	0.03	2.62	1.05	3.20	0.76	6.40	5.00	7.70

Since 1990 there has been a clear tendency for decreasing SO_4 concentrations in precipitation together with an increase in pH, especially near the main sources of anthropogenic pollution. However, this was not the case for precipitation in the Murmansk area which continued to be acidic (Figure 3.10). Acidity of precipitation within the Atlantic sector shows a strong correlation with sulfur concentration. There are no clear trends in NO_3 for this period. Trends in background areas of the Atlantic sector reflect changes in neighboring industrialized regions but with lower absolute values and less variability.

Siberian sector

Concentrations of SO_4-S in the Siberian sector of the Russian Arctic ranged from zero to 143.06 mg/L in Norilsk (which had the highest concentration throughout Russia). The average level for the region (excluding Norilsk) did not exceed 0.89 mg/L and there was a maximum concentration of 16.85 mg/L. Concentrations were less than 0.5 mg/L in 67% of samples. Background concentrations (Urengoy, Turukhansk, and Zhigansk, Table 3.6) ranged from 0.2 to 1.2 mg/L with the minimum values in warm or spring months. Concentrations below 1 mg/L were observed in 85% of samples only in Turukhansk. In contrast, concentrations at Norilsk were between 30 and 60 mg/L in 87% of samples. Average monthly concentrations at Norilsk were 50 to 60 times higher than at Turukhansk. Precipitation at Norilsk was most polluted in May and September (Figure 3.11). Seasonal variation at the background station in Turukhansk followed the same pattern as at Norilsk.

Concentrations of NO_3-N in precipitation within the Siberian sector were less than 2.3 mg/L in 94% of samples. The maximum (about 6.5 mg/L) was observed at the Dikson and Kyusyur stations. The background level at Turukhansk did not exceed 0.2 mg/L in 73% of samples (SD 0.22 mg/L). The concentration of NH_4-N in precipitation at this station was higher than NO_3-N in 90% of samples. Background areas were characterized by an average NH_4-N level of 0.3 mg/L (SD 0.25 mg/L). The highest concentrations (over 5 mg/L) mostly occurred at Kyusyur and the average value in this area was less 0.5 mg/L.

There have been no significant trends in sulfur (Figure 3.11) or nitrogen levels in precipitation at Norilsk over the last 15 years. Although concentrations decreased in the mid-1990s they have now resumed the levels seen in the early 1990s.

Precipitation over the Asian part of the Russian Arctic is generally less acidic than in the European part (Figure 3.9). Atmospheric precipitation in the Siberian sector is

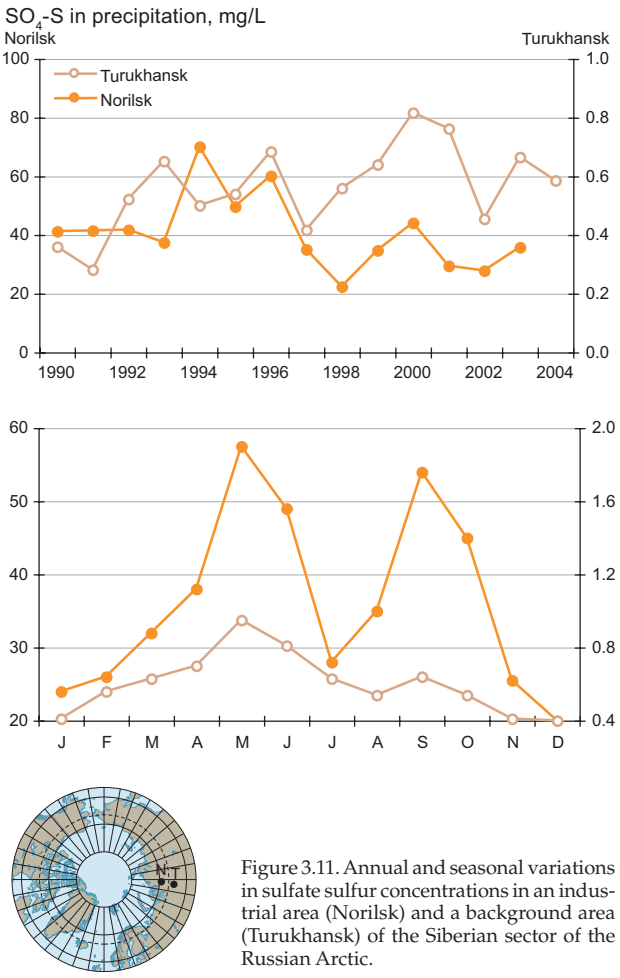


Figure 3.11. Annual and seasonal variations in sulfate sulfur concentrations in an industrial area (Norilsk) and a background area (Turukhansk) of the Siberian sector of the Russian Arctic.

generally alkaline. The average pH is 6.7. pH values were between 6.2 and 7 in 80% of samples. Values of 3.9 were observed twice and were over 7.7 in 20 samples. Precipitation at Turukhansk had an average pH of 5.9.

Pacific sector

The Pacific sector of the Russian Arctic was characterized by three stations: Deputatskaya, Ust-Moma, and Palatka. The average SO_4-S concentration in precipitation was 1.4 ± 1.7 mg/L. The highest concentration was 18.27 mg/L and the lowest was around the detection limit. Concentrations did not exceed 1.7 mg/L in 85% of samples.

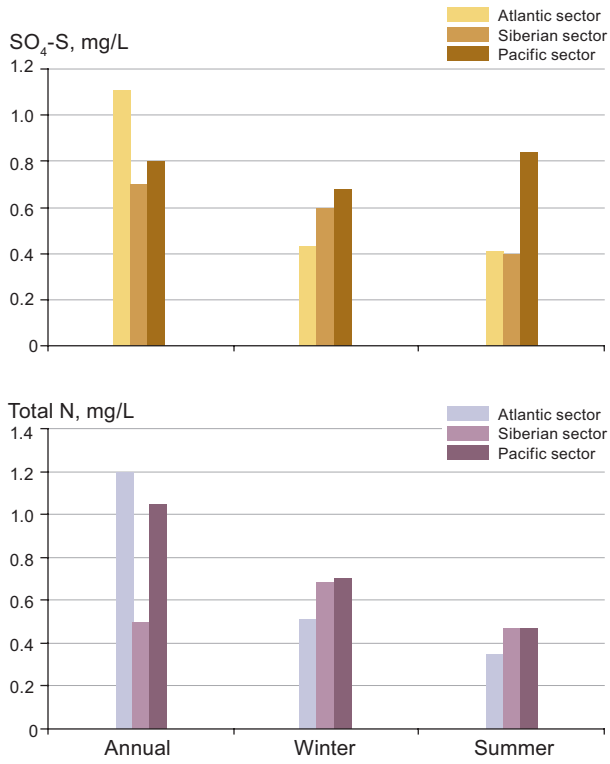


Figure 3.12. Annual and seasonal variations in average background levels of sulfate sulfur and total nitrogen in precipitation across the Atlantic, Siberian, and Pacific sectors of the Russian Arctic.

The average NO_3-N concentration in precipitation was 0.2 ± 0.2 mg/L. Concentrations did not exceed 0.14 mg/L in 50% of samples. The highest concentration was 1.5 mg/L and the lowest was below the detection limit. The average NH_4-N concentration was around 0.64 ± 0.8 mg/L. Concentrations did not exceed 0.7 mg/L in 75% of samples. The highest value was 8.01 mg/L and the lowest was below 0.01 mg/L.

Precipitation in the Pacific sector has continued to decrease in acidity over the last few years. The pH was between 6.7 and 7.2 in 60% of samples and in eight samples the monthly pH was 7.6. The lowest value (5.0) was observed twice. The annual precipitation was 400 to 500 mm, with 5 to 65 mm per month.

A comparison of background pollutant levels in precipitation across the Russian Arctic is presented in Figure 3.12 and Table 3.6.

The acidity of precipitation within the Russian Arctic did not increase over the study period (1990 to 2004). Acidic precipitation was only observed on the Kola Peninsula, where 60% of samples had pH values of 5.0 and episodic precipitation events may have had values as low as 3.2. Precipitation acidity within the Atlantic and Siberian sectors decreased over the study period, while levels in the Pacific sector remained the same.

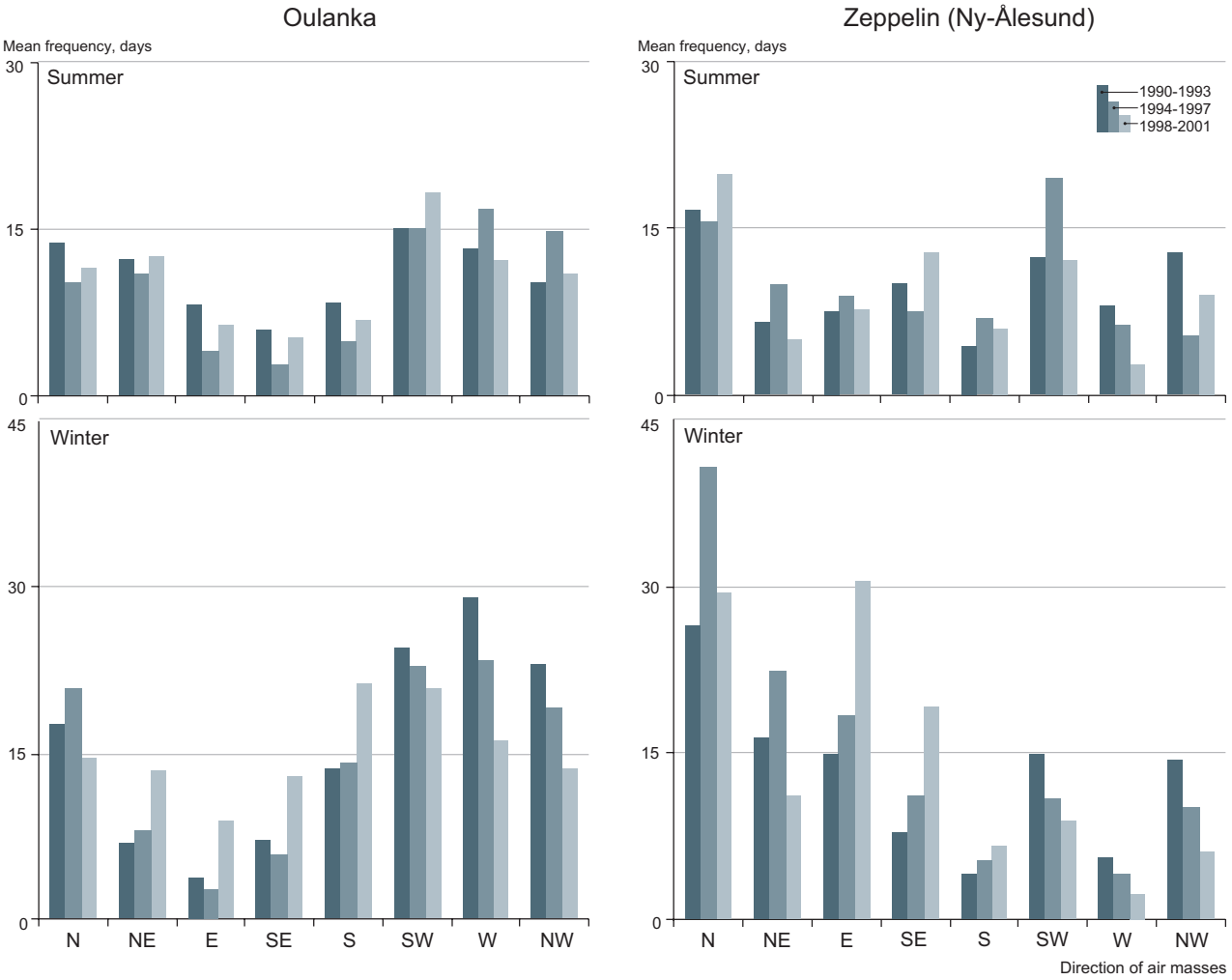


Figure 3.13. Frequency and variation in the direction of air masses arriving at Oulanka and Zeppelin over 4-year periods during the 1990s.

3.4. Episodes and exposure to sulfur and nitrogen

Atmospheric transport and deposition data are usually presented as mean values owing to the enormous amount of data collected. However, a characteristic feature of observations in atmospheric concentration, particularly at remote sites, is the occurrence of extreme peaks in concentration. The 2002 AMAP assessment on the influence of global change on contaminant pathways to, within, and from the Arctic (Macdonald *et al.*, 2003) stated that events and short-term variations are important for the delivery of contaminants to the Arctic. The assessment also stated that the frequency of extreme events is likely to increase with climate change.

A large proportion of the annual acid deposition is often accumulated in just a few days. For example, a study in the 1990s found that the five worst days at Finnish background stations could bring 20 to 30% of the annual bulk sulfate load (Ruoho-Airola and Salmi, 2001). Days on which the deposition exceeded the annual median value for the station by a factor of 10 were counted as episode days. The number of episode days in central and northern Finland was significantly more frequent. The episodes were not just due to high levels of precipitation since the mean sulfate concentration in episodic rain was higher than the annual mean concentration. The episodes were mostly imported: the air mass arriving at the background stations had passed over high emission areas outside Finland. High episodicity has been detected in large areas of northern Europe (Smith and Hunt, 1978) and eastern North America (Brook, 1995).

Sectoral exposure to atmospheric sulfur and nitrogen compounds in the 1990s was examined at a Finnish arctic station (Oulanka) and a Norwegian station (Zeppelin) located in Spitsbergen (see Figure 3.13). Variations in the atmospheric circulation over Fennoscandia have an important effect on the composition of the atmosphere. Daily concentrations of gas phase and particulate sulfur and nitrogen compounds together with estimates of air transport routes (2-dimensional 925hPa trajectories obtained from the EMEP MSC-W; http://www.emep.int/index_assessment.html) enabled a sectoral examination of pollution exposure. Sectoral exposure means the sum of the daily loads arriving from a specific sector over the period assessed. The sectoral distributions roughly illustrate the relative importance of different transport directions to the dry deposition of sulfur and nitrogen (Ruoho-Airola *et al.*, 2004).

Transport from the southwest and west dominated the airflow at Oulanka, whereas transport was most frequent from the north at Zeppelin. Figure 3.13 shows four-year mean values for the different transport sectors for each station. The results are supported by an analysis of atmospheric circulation during different states of the Arctic Oscillation (Macdonald *et al.*, 2003). In the 1990s, during a positive phase in the AO index, southwesterly winds predominated at Oulanka, whereas at Spitsbergen the winds were more from the north and east, particularly in winter. However, the frequency of westerly airflow decreased at Oulanka during the 1990s (Moberg *et al.*, 2005).

At Oulanka, the SO_2 concentrations in summer were highest in air masses arriving from the north and northeast, in winter concentrations were highest in air masses from sectors between the northeast and southwest. At Zeppelin,

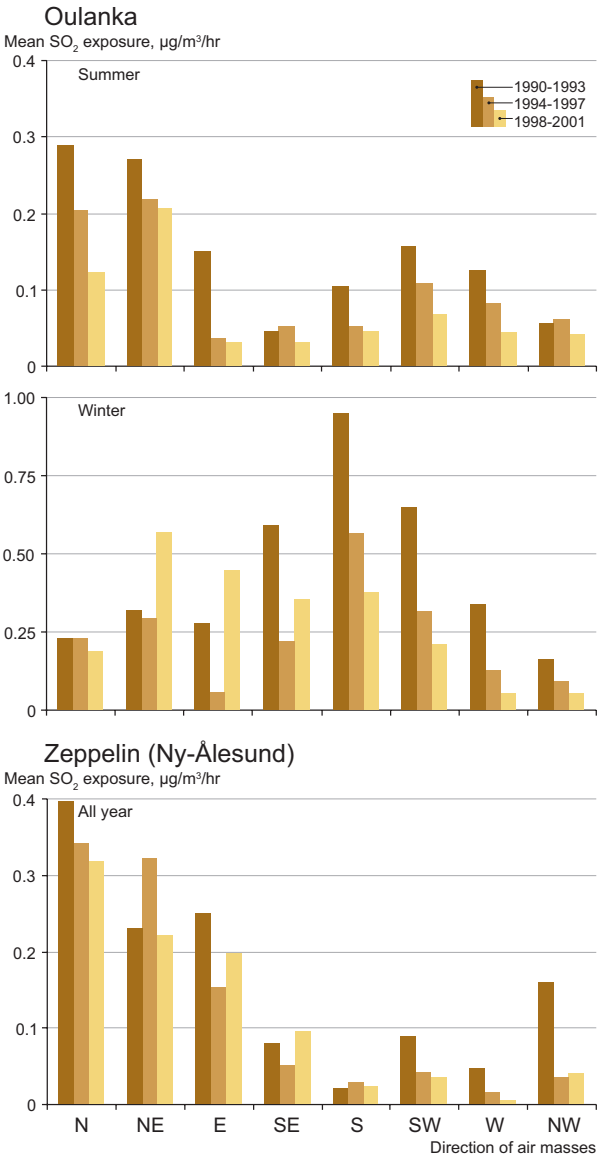


Figure 3.14. Mean exposure to sulfur dioxide at Oulanka and Zeppelin in relation to the direction of air masses arriving at the stations in the 1990s.

the highest concentrations occurred in air masses from the north.

In summer, exposure to SO_2 at Oulanka is highest with the arrival of air masses from the northern sectors – which is where the high emissions on the Kola Peninsula are located. In winter, exposure is dominated by transport from the southern and southwestern sectors, and since the end of the 1990s from eastern sectors as well. At Zeppelin exposure is dominated by transport from the north and northeast throughout the year. Figure 3.14 shows the sectoral distribution for SO_2 exposure during the 1990s as four-year mean values.

Atmospheric sulfate is highest at Oulanka in air masses arriving from between the southeast and southwest. At Zeppelin the highest sulfate concentrations occur in air masses arriving from the northeast and east. Exposure to sulfate closely follows the pattern for SO_2 with the exception that at Oulanka the winter distribution is also evident during summer.

The patterns of concentration and exposure for total atmospheric ammonium (i.e., the sum of gaseous ammonia and particulate ammonium) and total atmospheric nitrate

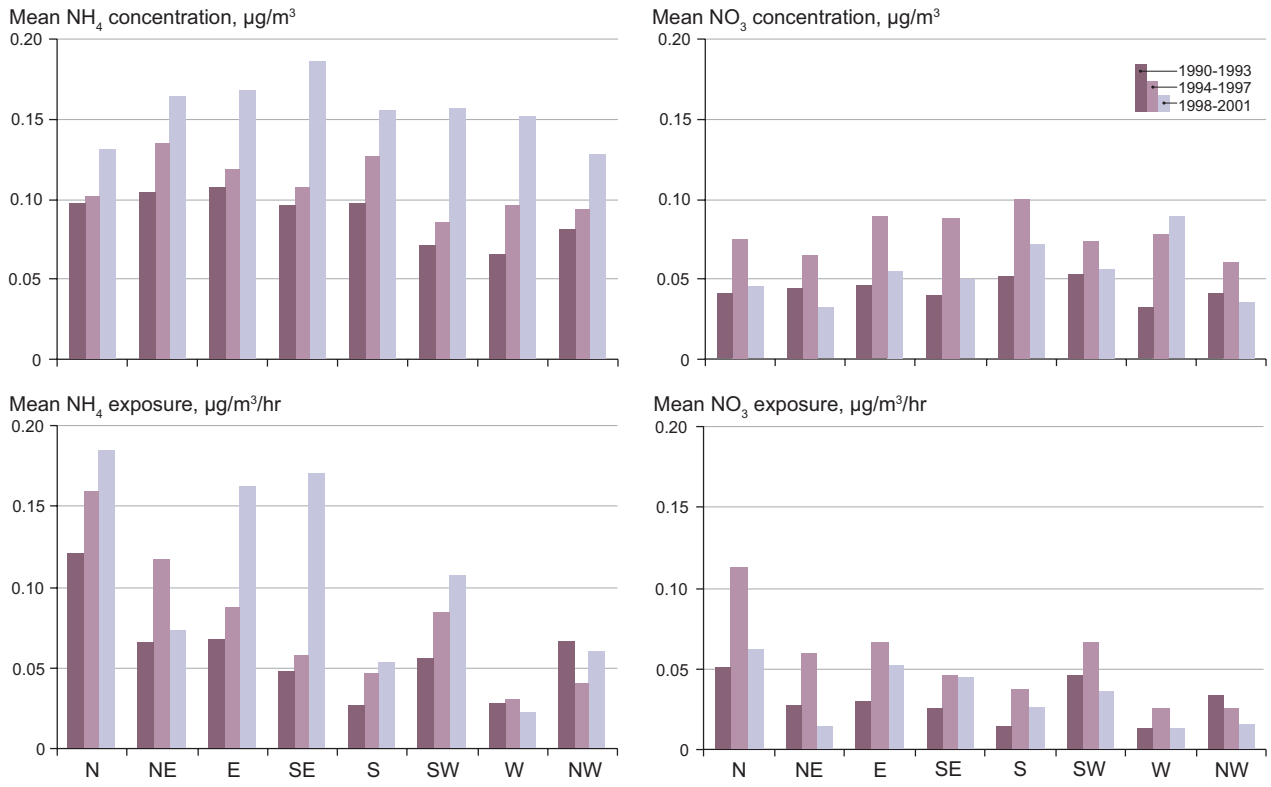


Figure 3.15. Mean concentration and exposure to nitrogen compounds at Zeppelin in relation to the direction of air masses arriving at the station in the 1990s.

(i.e., the sum of gaseous nitric acid and particulate nitrate) at Oulanka were similar to those for sulfate. At Zeppelin the difference in concentration for the various air masses was small (Figure 3.15), thus exposure was highest from the northern sector as air masses were most frequent from the north (Figure 3.13). However, recent research indicates that the main source area for nitrate deposited on Svalbard is Western Europe (Julin, 2003).

3.5. Concentrations in seasonal snow cover

3.5.1. General pattern

A recent study of snow samples from the end of winter 1996/1997 provides additional information about the acid deposition pattern in the Arctic (de Caritat *et al.*, 2005). The study was based on a total of 21 snow samples from 17 arctic locations, including sites in Norway, Sweden, Finland, Svalbard, Russia, Alaska, Canada, Greenland, and Iceland. Major element concentrations in the melted snow indicate that the composition of most samples was consistent with diluted seawater. Deviations indicate additional SO_4 and Cl relative to seawater, suggesting an anthropogenic contribution (Alaska, Finland, Sweden, Svalbard). The samples with the highest Na and Cl content (Canada, Russia) also have higher Na:SO_4 and Cl:SO_4 ratios than seawater, suggesting a slight contamination from (probably local) de-icing activities. Local soil or rock dust inputs to the snow are indicated by 'excess' calcium (Alaska, Svalbard, Greenland, Sweden). No overall relationship was found between pH (range: 4.6–6.1) and total or non-marine SO_4 (SO_4*), but the data are too limited (with only one season sampled) to draw firm conclusions on long-range SO_2 transport. In a few samples (Alaska, Finland, Sweden, Svalbard), a significant proportion (>50%) of SO_4 was non-marine in origin. A few samples from northern Europe showed a weak trend of decreasing pH with increasing SO_4.

3.5.2. Russian Arctic

The pollution load and acidity of snow cover in the Russian Arctic is monitored at 99 stations. Atmospheric deposition loads of sulfur and nitrogen compounds per unit area are presented in Table 3.7. Average data for the European and Asian parts of Russia are also given for comparison (Belikova *et al.*, 1984). Estimated atmospheric deposition loads for the continental Russian Arctic (see Table 3.8) (Reviews of the state of environment in Russian Federation, 1995) include estimates of the proportions of the sulfur and nitrogen derived from sources within the Russian Arctic and from transboundary sources. Deposition on the open sea areas of the Arctic Ocean is presented in Table 3.9 and the distribution of measured pH values in snow cover within the Russian Arctic is shown in Figure 3.16. Not all snow monitoring sites reported data each year owing to difficulties with the transport and storage of samples under arctic conditions.

Several conclusions can be drawn from the data presented in Tables 3.7, 3.8, and 3.9 and Figure 3.16. It is clear that the average level of atmospheric deposition in background areas of the Russian Arctic (i.e., outside the areas affected by the major industrial centers) is much lower than in the European and Asian parts of Russia to the south. Also, that atmospheric deposition fluxes for both sulfur and nitrogen decrease from west to east across the Russian Arctic. The highest levels of sulfur deposition occur within

Table 3.7. Average atmospheric deposition loads for sulfur and nitrogen in the Russian Arctic.

	Atmospheric loads, kg/km²/yr			
	S	NO_X-N	NH_4-N	Total N
Kola Peninsula				
Eastern part of peninsula (minimum values)	300	60	130	190
Entire region	452	66	137	203
50 km zone around smelters	800-3000	60-90	100-140	160-230
Arkhangelsk region (northern part)	180	100	142	242
Yamal Peninsula (entire region)	130	150	160	310
Taymir Peninsula	100	70	90	160
Entire region				
50 km zone around Norilsk smelter complex	450-4000	70-120	80-120	150-220
Arctic Ocean coastal area and Yakutsk region	75	35	65	100
Arctic Ocean islands				
Zemlya Frantsa Iosifa	140	42	125	167
Severnaya Zemlya and islands	90	50	100	150
Novosibirsk islands	40	10	65	75
European territory of Russia	810	180	450	630
Asian territory of Russia	350	90	180	270

Table 3.8. Atmospheric deposition loads for sulfur and nitrogen emitted from sources within the Russian Arctic and from transboundary sources.

	Total deposition		Sources within the Russian Arctic		Transboundary sources	
	t/yr	%	t/yr	%	t/yr	%
Kola Peninsula						
Sulfur	65500	100	46100	70.5	19400	29.5
Nitrogen (NO_X)	9600	100	8000	8.3	8800	91.7
Total nitrogen	30000	100				
Arkhangelsk region and Komi (Arctic part)						
Sulfur	48400	100	5890	12.2	42510	87.8
Nitrogen (NO_X)	13100	100	840	6.4	12260	93.6
Total nitrogen	46500	100				
Tyumen region (Arctic part)						
Sulfur	58600	100	1600	2.8	57000	97.2
Nitrogen (NO_X)	13900	100	6600	47.5	7300	52.5
Total nitrogen	72600	100				
Krasnoyarsk region (Arctic part)						
Sulfur	286200	100	222000	77.5	64200	22.5
Nitrogen (NO_X)	15900	100	1060	6.7	14840	93.3
Total nitrogen	142590	100				
Yakutia, Magadan region and Chukotka (Arctic part)						
Sulfur	70720	100				
Nitrogen (NO_X)	36400	100				
Total nitrogen	104000	100				

Table 3.9. Atmospheric deposition loads for sulfur and nitrogen compounds on the open sea areas of the Arctic Ocean.

	Sulfur		NO_3-N	
	Average flux , kg/km²/yr	Amount, t/yr	Average flux , kg/km²/yr	Amount, t/yr
Barents Sea	180 (110-220)	256000	60 (40-70)	85440
Kara Sea	90 (60-120)	79470	40 (25-65)	35300
Laptev Sea	30 (20-40)	19860	12 (5-25)	7900
East Siberian Sea	25 (20-30)	22800	10 (5-20)	9100
Chukchi Sea	30 (10-30)	11900	15 (10-20)	9000

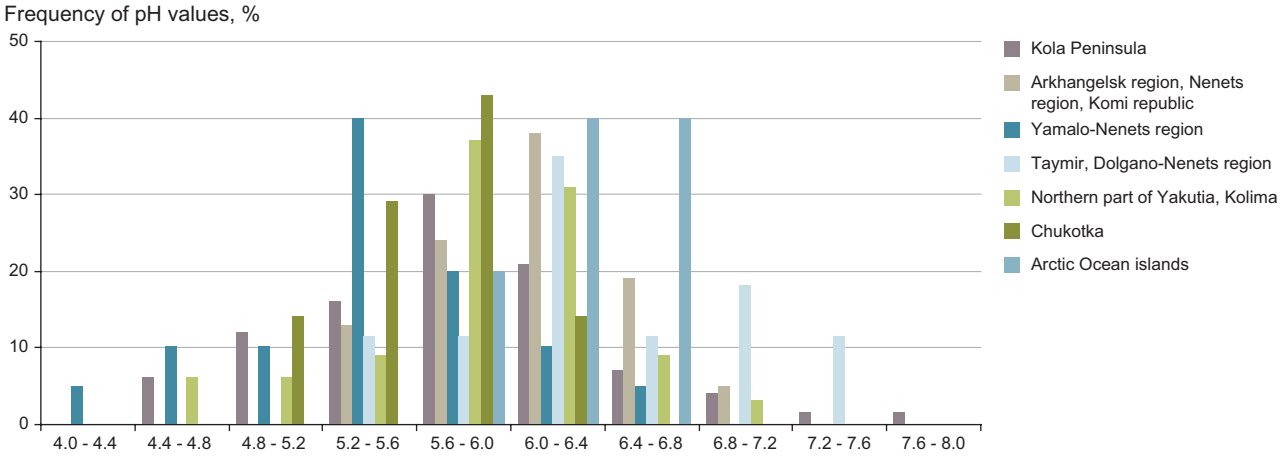


Figure 3.16. Acidity of snow cover across the Russian Arctic, 2001–2004.

concentric zones extending approximately 60 km from the non-ferrous metal smelters on the Kola Peninsula and at Norilsk. The tables and figure do not indicate acidic precipitation in winter over large areas of the Russian Arctic. The average pH level in snow cover accumulated during the polar winter (6-7 months) is typically above 5.0, although acidic precipitation is sometimes seen in Taymir at the outer limit of the area affected by industrial activities in Norilsk.

The tables show that around 530 kt of sulfur, 89 kt of nitrogen as nitrogen oxides, and 382 kt of total nitrogen are deposited on the continental region of the Russian Arctic each year. The proportion estimated to have originated from Russian sources ranged from 12% (Arkhangelsk region, Komi, Tyumen region) to 70–77% (Murmansk region, the northern Krasnoyarsk region). Annual deposition to the seas of the Russian sector are estimated at 390 kt of sulfur and 147 kt of nitrate nitrogen (Table 3.9).

Figure 3.16 shows that acidic snow cover occurred in all regions. Acidic snow (snow with a pH of less than about 5.6) was most frequently observed in the Yamalo-Nenetckiy region (65% of measurements). The lowest frequency of acidic snow samples (11–13%) was observed in the Arkhangelsk region, Nenetckiy region, Komi republic, and Taymir.

In seasonal snow in the area closest to the non-ferrous metal smelters on the Kola Peninsula (Nikel, Zapolyarnyy, and Monchegorsk) pH levels are relatively high, at up to 6.0–6.8. This reflects neutralization by alkaline dust, which is a very important process in such areas. No seasonal trends were evident in the pH values. Acidic snow with pH values of 4.6 to 5.2 is evident more than 100 km from the non-ferrous smelters in the direction of the prevailing winds.

The effects of emissions from Norilsk enterprises on snow acidity are unclear. A snow acidity level of pH 6.0 dominates the area up to 250 km from Norilsk. Measurements at 12 monitoring sites up to 800 km to the south and southeast of Norilsk found no pH values below 5.6. However, Norilsk emissions may have decreased the pH value for snow cover to the west of Norilsk in the direction of Yamal.

The influence of sea salt on precipitation acidity is evident in coastal areas of the Kola Peninsula, in the Arkhangelsk region, and in Chukotka. This is characterized by a relatively small decrease in pH (to 5.0) compared to the background level of 5.6.

Precipitation chemistry in Yakutia is mainly influenced by salts of continental origin. This is clear from the high calcium bicarbonate content, while salts of marine origin comprise only 16% of the total salt content. Precipitation in Yakutia has a pH close to 5.6 which is typical of unpolluted precipitation (Figure 3.17). Emissions from the industrial sources (e.g., non-ferrous metal smelters) commonly produce an alkaline reaction; thus precipitation and snow cover in the vicinity of these sources have higher pH values than those further away. On the other hand, acidification of precipitation and snow cover occurs in areas remote from the sources (due to long range transport of sulfur and nitrogen compounds) and the snow water here may have low pH values.

The chemical composition of the snow cover is directly dependant on the chemical composition of the precipitation. Snow chemistry is also influenced by gas exchange between the snow cover and the atmosphere during the long polar winter, by the transport of material from the

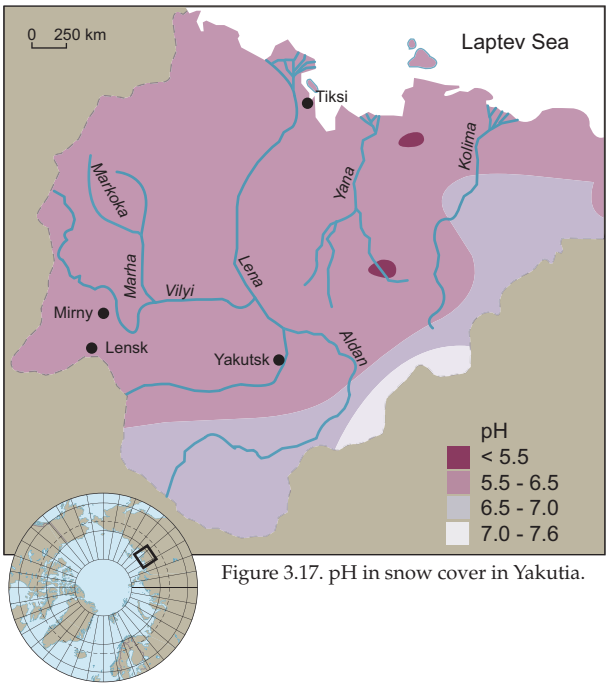


Figure 3.17. pH in snow cover in Yakutia.

underlying surface to the snow, and by the dissolution of contaminants in the snow cover during snowmelt.

Relatively high pH values (i.e., values near neutrality; 6.5–7.2) in snow cover in southeastern and eastern Yakutia reflect the arrival of cold air masses from the Pacific basin. Snow acidification due to local sources has not been observed in Yakutia.

Because the pH of meltwater depends on the anion:cation ratio and on the presence of alkaline compounds such as calcium and magnesium oxides which predominate in emissions, high pH values occur in snowmelt waters. High dust levels in the air in industrial regions and a predominance of carbonates and calcium and magnesium oxides in the emissions, have lead to a sharp increase in pH values in some industrial areas: in the heavily impacted areas of western Yakutia snow pH values as high as 7.5 to 8.5 occur in some urban regions (Udachny: 7.85; Mirnyi: 8.2) and 8.0 to 9.4 in mining areas (the Mir diamond pipe: 9.45).

3.6. Pollution history from ice cores and lake sediments

Whereas ice cores from Antarctic and Greenland ice sheets have been useful for extracting pollution history with high time resolution, ice cores from arctic ice caps have been considered difficult to use because summer melting and percolation of melt water are thought to blur the signal (Głowacki, 1997). However, it has recently been shown that by including a model for percolation effects (elution of ions), it is possible to reconstruct the 'pristine' chemical composition of ice caps such as Lomonosovfonna (78°51' N, 17°25' E, 1255 m above sea level) on Svalbard (Moore *et al.*, 2005). Even with 80% melting there is little disturbance in chemical stratigraphy. This suggests that ionic records from arctic ice caps are almost as reliable as those from the large ice sheets where melting hardly occurs. This new type of knowledge and improved and more objective

techniques (e.g., Rasmussen *et al*., 2002) make it possible to interpret and compare trans-Arctic ice core chemical records. However, nitrate and sulfate profiles from ice caps in areas such as the Russian Arctic are still not available in the open literature. Spatial variations in macroelements in Severnaya Zemlya glaciers (Evseev *et al*., 2000) indicate that averaged concentrations deposited on snow and ice during warm stages of the Late Holocene and historical epoch are 1.8 to 3 times higher than the corresponding values for cold stages.

To interpret chemical signals in ice cores, knowledge about deposition and post-depositional processes for the different atmospheric compounds is required. Sharp *et al*. (2002) studied the snow pack chemistry at the John Evans Glacier at Ellesmere Island, Canada (79°40′ N, 74°23′ W) and showed that the seasonal cycle in snow chemistry closely reflects changes in the composition of the atmospheric aerosol at Alert. There was some modification of nitrate concentrations by post-depositional processes. Mean water-weighted solute concentrations in the snow pack are largely independent of accumulation, while atmospheric deposition tends to increase with accumulation. This suggests that, for most species, wet deposition is the dominant depositional process throughout the year. However, concentrations of calcium and potassium increase with both accumulation and elevation, implying an enhanced input from dry deposition of soil dust above 800 m elevation. Concentrations of sulfate are inversely related to accumulation, especially in the winter layer, suggesting a significant input from non-precipitating events, such as dry deposition or riming, during this period of very limited snowfall.

Results of ice core drillings from Svalbard have been published over the last ten years or so (e.g., Kekonen *et al*., 2002). Even though summer melting and percolation of melt water diffuses the signals, it is evident that useful information about climate and pollution history can be obtained from these studies (Isaksson *et al*., 2003). The anthropogenic influence on the Svalbard environment is illustrated by increased levels of non-sea-salt sulfate, nitrate, acidity, fly ash, and organic contaminants, particularly during the latter half of the 1900s (Figure 3.18). Decreased concentrations of some components in recent decades probably reflect emissions reductions.

At Penny Ice Cap (67°15′ N, 65°46′ W) in the Canadian Arctic, Goto-Azuma *et al*. (2002) showed that summer melting resulted in disturbance of the seasonal signal. Sulfate and nitrate concentrations at this site started to increase around 1900 and 1960 respectively. Bigler *et al*. (2002b) showed that, over the last 1200 years, significant changes in sulfate levels were present only during the industrial era. Their Greenland ice core was taken at B20 (78°50′ N, 36°30′ W, 2150 m above sea level).

To date, lake sediments have been considered of limited value as archives of the atmospheric pollution history since around 1800 because the temporal resolution has been too low. However, recent research shows that human activity has induced climate-driven regime shifts in lakes across the Arctic (Smol *et al*., 2005) and the effect of increased nitrogen deposition cannot be excluded. Increased nitrate concentrations throughout the 1990s also correspond well with changes in diatom populations found in lake sediments on Svalbard and at other arctic sites (Wolfe, University Centre on Svalbard pers. comm., 2005). Increased nutrient deposition and climate change are possible explanations for the consistent pattern of biological change observed (e.g., relative appearance of species).

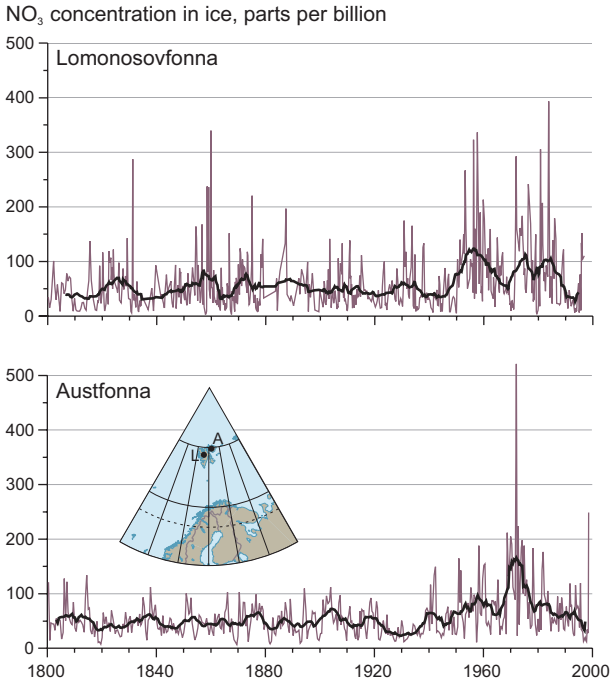


Figure 3.18. Nitrate concentrations in ice cores from glaciers on Svalbard (Isaksson *et al*., 2003). The nitrate records from Lomonosovfonna (adapted from Kekonen *et al*., 2002) and Austfonna (adapted from Watanabe *et al*., 2001) are for the period from 1800 to the present. The black line is a 25-year running mean.

3.7. Modeling

The Danish Eulerian Hemispheric Model (DEHM) system consists of a weather forecast model, the PSU/NCAR Mesoscale Model version 5 (MM5) modeling subsystem (see Grell *et al*., 1994), which is driven by meteorological data from ECMWF (the European Centre for Medium-range Weather Forecasts), and a 3-dimensional atmospheric transport model, the DEHM model. The model has a horizontal resolution of 150 x 150 km and 20 vertical layers and the coverage is close to hemispheric from nearly 10° N at the corners and 25° N at the midpoints of the model domain boundaries.

The model system has been used to study transport of air pollution to the Arctic since 1991. There are several versions of this system. The original version of the DEHM model was developed for studying the long-range transport of SO_2, SO_4 and lead (Pb) to the Arctic (Christensen, 1997, 1999). The sulfur version was used in the first AMAP assessment (see Kämäri, 1998) and the Pb version in the AMAP heavy metals assessment (AMAP, 2005). The model was further developed to study transport, transformation, and deposition of reactive and elemental mercury, and this version was also used in the heavy metals assessment, see also Christensen (2004) and Heidam *et al*. (2004). Other versions calculate the concentrations and deposition of various pollutants (Frohn *et al*., 2002, 2003) through the inclusion of an extensive chemistry scheme, and transport and exchange of atmospheric carbon dioxide (Geels *et al*., 2004) and persistent organic pollutants (Hansen *et al*., 2004).

This assessment used both the original version of the model and the extensive chemical version of the model. The original version includes two species: SO_2 and SO_4 and the chemical transformation of SO_2 to SO_4 is described by a simple linear transformation depending on time of year and latitude. The extensive chemical version includes 63

species and more than 120 reactions that describe the chemistry of sulfur oxides, nitrogen oxides, reduced nitrogen (NH_X), volatile organic compounds, and ozone. The simple version of the DEHM model was used for the long-term calculations because it is up to 30 times faster than the version with the large chemical scheme. Emissions used as input to the models are described in section 2.3.

3.7.1. Validation of the system for temporal trend analysis

The chemical version of the DEHM model was run for the period 1991 to 2002 using actual emissions estimates for each year. Mean air concentrations of SO_X (i.e., SO_2+SO_4) and NO_3 are shown in Figure 3.19 for 2000. The SO_X results show clearly the very high concentrations around Norilsk,

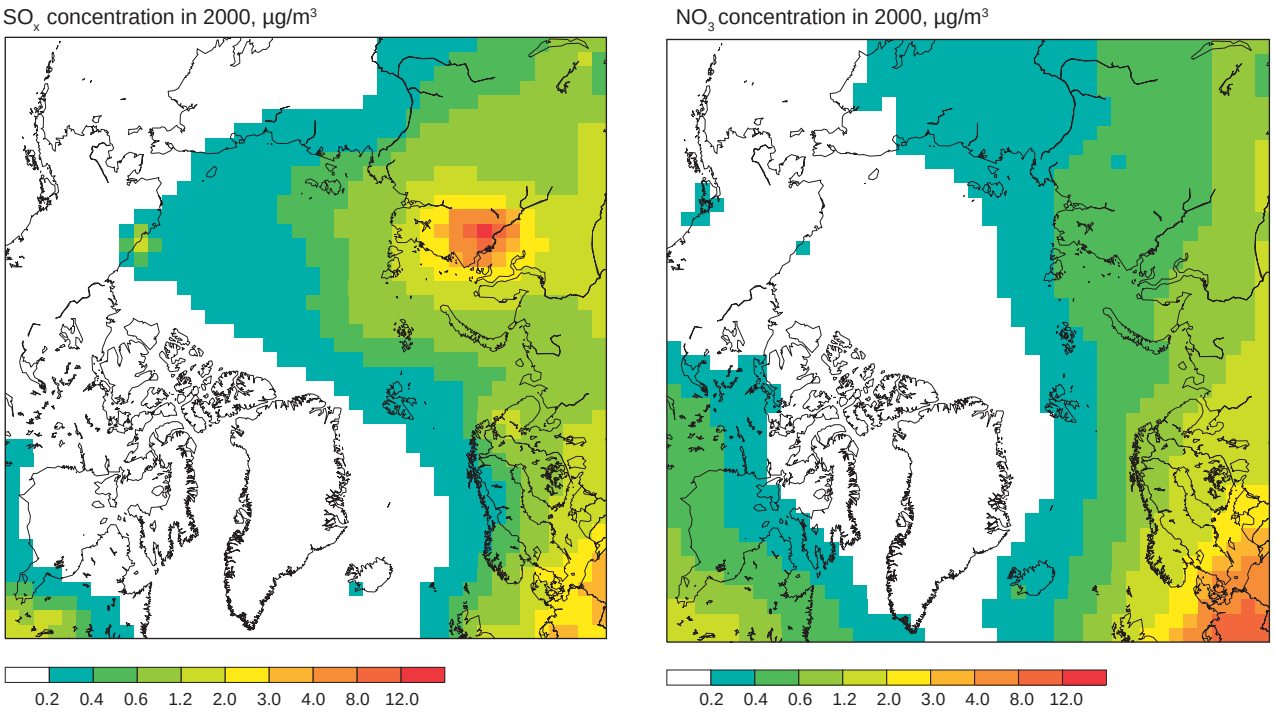


Figure 3.19. Mean surface air concentrations of sulfur oxides (i.e., sulfur dioxide plus sulfate) and nitrate in 2000.

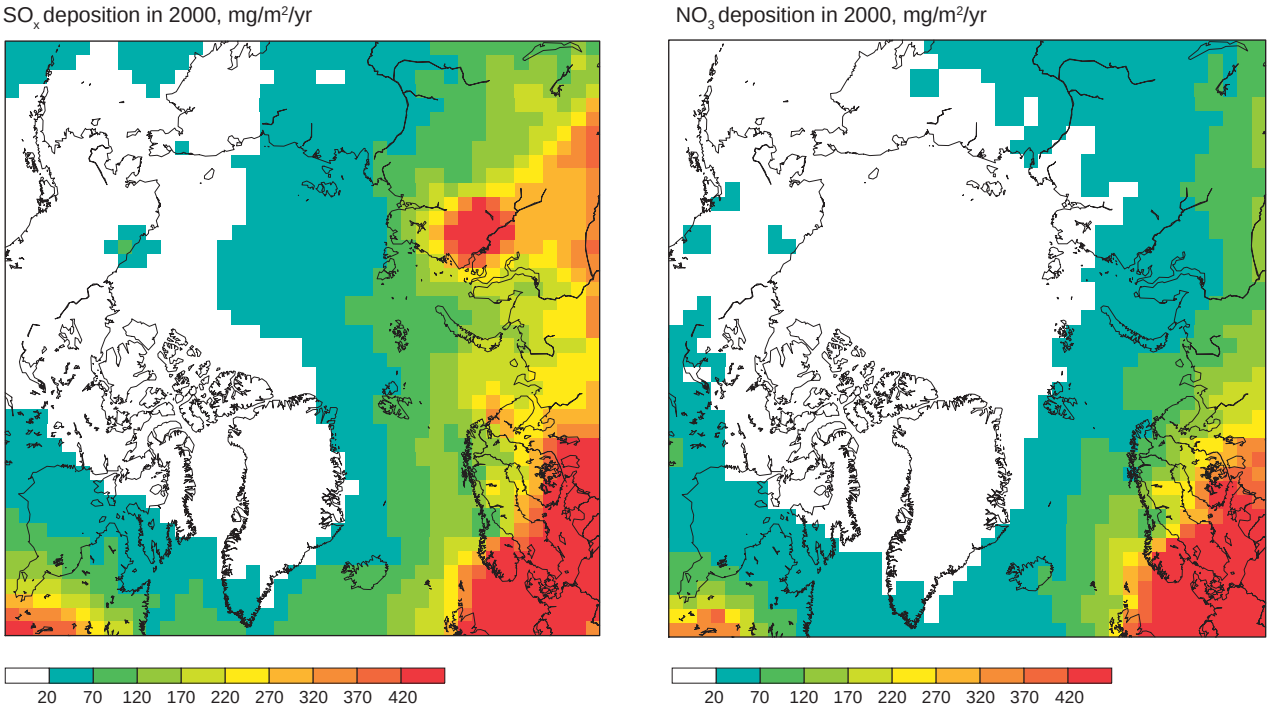


Figure 3.20. Total deposition of sulfur oxides (i.e., sulfur dioxide plus sulfate) and nitrate in 2000.

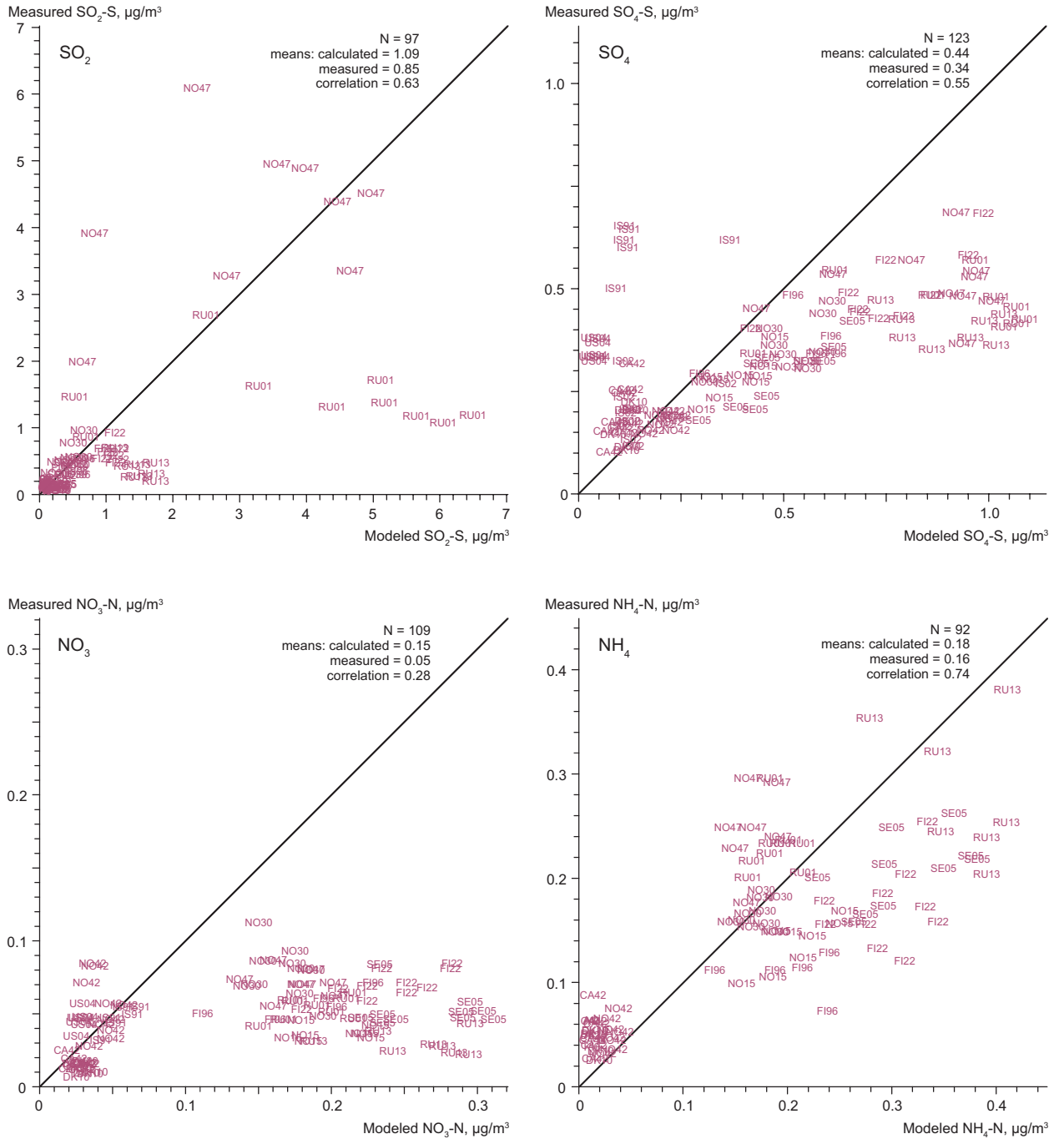


Figure 3.21. Scatterplots comparing measured and modeled annual average concentrations of sulfur dioxide, sulfate, nitrate and ammonium at arctic monitoring stations for 1991 to 2000. Labels on graphs refer to EMEP station codes.

and the high concentrations across the Kola Peninsula, and the elevated concentrations associated with oil-related activities at Barrow. The NO_3 results do not indicate any local hot spots within the Arctic and that concentrations decrease from the source areas in Europe and North America towards the Arctic. In Figure 3.20 total deposition of SO_X and NO_3 for 2000 are shown, and the general patterns are similar to those for air concentrations in Figure 3.19.

For arctic areas the model results were compared with measurements from a range of background monitoring stations (Figure 3.21). A scatter plot comparing measured and modeled annual average concentrations of SO_2, SO_4, NO_3 and NH_4 for all arctic stations for each year between 1991 and 2000 shows a reasonable correspondence between the two datasets for SO_2 and SO_4, and a particularly good correspondence for NH_4. The model predicts the annual averages for most arctic stations well, but over predicts NO_3 concentrations for stations in the European Arctic.

Figures 3.22, 3.23 and 3.24 show time series of measured and modeled monthly mean concentrations of SO_2, SO_4 and NO_3 for some arctic monitoring stations. For Tustervann (Figure 3.22) there is reasonable agreement between the modeled and measured data for both SO_2 and SO_4, but poor agreement for NO_3 concentrations. This is also the case for Zeppelin (Figure 3.23). The results for Janiskoski (Figure 3.24) show that for this station the model overestimates the SO_2 concentrations for most years, except after 1999. The main reason for this is that the spatial distribution

of official EMEP expert emissions for the Kola Peninsula was changed in 1999 – before 1999 high emissions were attributed to the grid cell in which Janiskoski is placed whereas after 1999 these emissions were allocated to the neighboring grid cell. These changes in the distribution of emissions also result in a better performance of the model for SO_4. The model is able to predict well monthly variations in SO_2 and SO_4 for a large part of the Arctic, with most difficulties associated with the Russian stations and at Alert and Denali in North America. Common to all stations are poor results for NO_3. The model predicts very well the higher NO_3 concentrations at mid-latitudes, but over predicts lower NO_3 concentrations at high latitudes in the European Arctic.

3.7.2. Trend analysis based on measurements at Station Nord and DEHM model results

Total sulfur (the sum of sulfur measured as sulfur dioxide and sulfate) measured at Station Nord shows a strong seasonal variation with high values in the winter/spring and very low values in the summer. In addition, the partition of the two components changes during the winter/spring season from a high content of sulfur dioxide during the dark winter period to a very low content after polar sunrise in early spring.

Station Nord is virtually unaffected by local or regional air pollution and can be considered a remote watchtower from where to follow the average emissions from a huge emissions area in the eastern part of Europe and Russia.

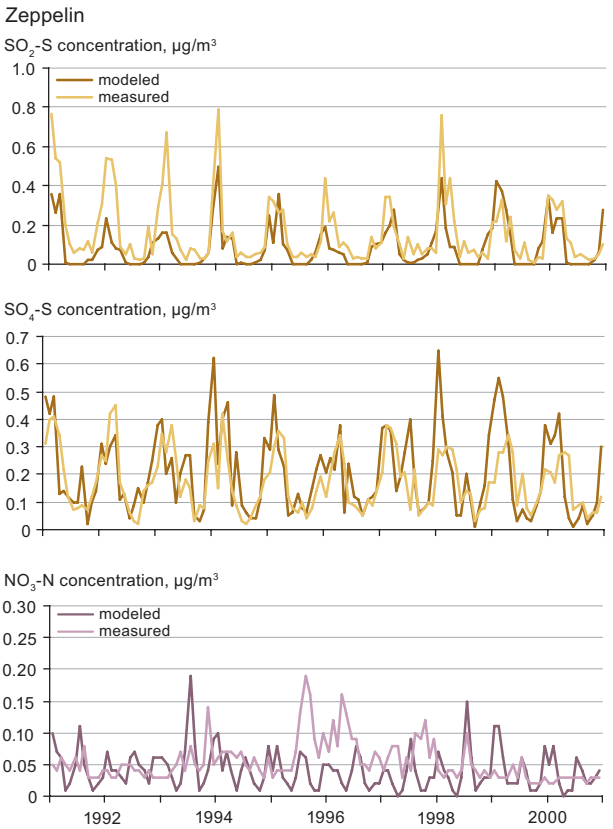


Figure 3.23 Time series of measured and modeled monthly concentrations of sulfur dioxide, sulfate, and nitrate at Zeppelin (Spitsbergen).

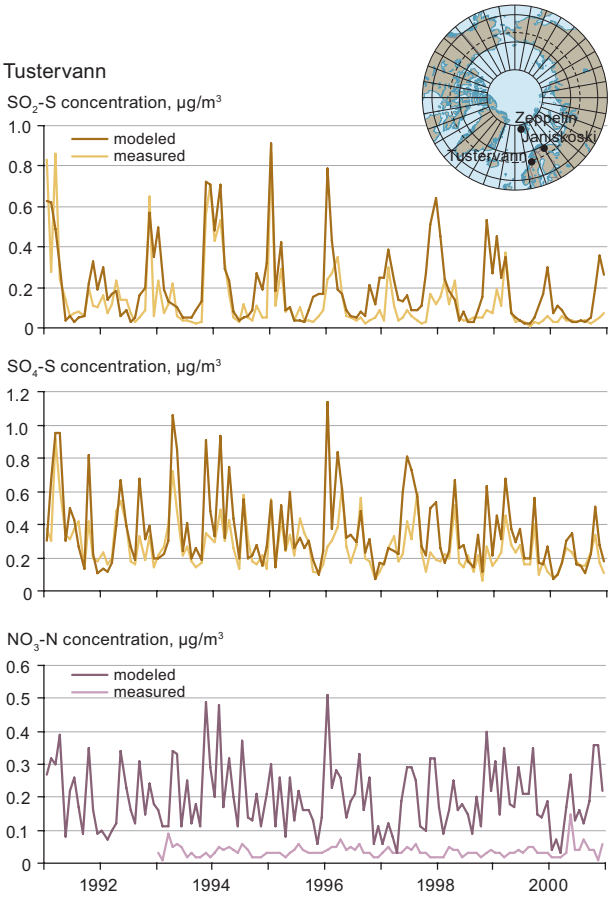


Figure 3.22. Time series of measured and modeled monthly concentrations of sulfur dioxide, sulfate, and nitrate at Tustervann (Norway).

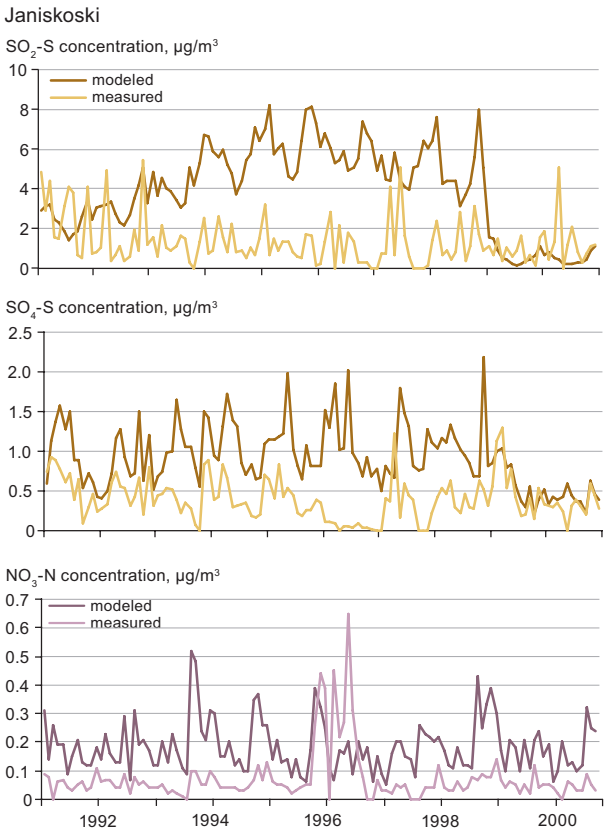


Figure 3.24. Time series of measured and modeled monthly concentrations of sulfur dioxide, sulfate, and nitrate at Janiskoski (Russia).

A study of the general emissions trends is therefore possible using the long time series of sulfur measurements made over the last decade. But, unfortunately, the measured concentrations depend strongly upon meteorological conditions along the transport pathway to the Arctic. In summer, emissions never get to the Arctic, whereas in winter, transport times can change greatly from week to week. Therefore, single measurements provide a poor reflection of emissions in the source areas and a trend analysis on the raw concentration data can give misleading results. One way round this problem was to use the results of the hemispheric Eulerian model DEHM.

The DEHM model with simple SO_2-SO_4 chemistry was run for the 11-year period from 1991 to 2001, with emissions kept constant by using emissions data for 1990 only. All variations in modeled concentrations at Station Nord were therefore due to daily changes in meteorological conditions. Figure 3.25 shows the ratio SO_x(measured): SO_x(modeled), calculated by weighted regression analysis for each year between 1991 and 2000 (Wåhlin *et al.*, 2002). The values were fitted by an exponential function of time. It is evident that this trial function does not disagree statistically with the data points when the uncertainties (standard deviations) are taken into account. The fitted function has an exponential decay of 11 years with an uncertainty of 2 years. If it is assumed that the changes in the ratio between the measured and modeled concentrations of total SO_X are due to changes in emissions only, these analyses indicate that the emissions, which contribute to the measured concentrations at Station Nord, decreased by a factor of two between 1991 and 2000.

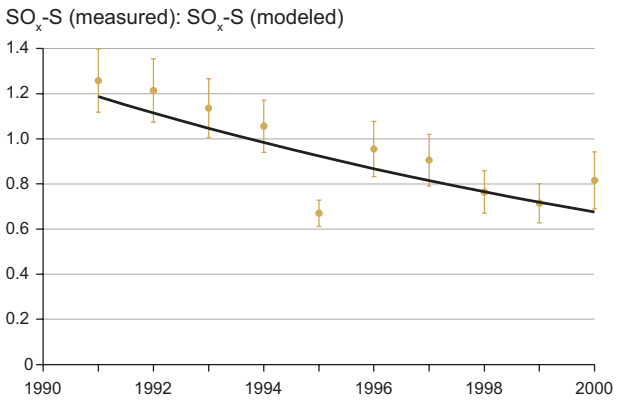


Figure 3.25. Ratios determined by regression analysis of measured values of sulfur (SO_x-S) and constant emission model values for each year between 1991 and 2000. The values are fitted by an exponential function of time with a half-value period of 11±2 years.

3.7.3. **Effects of natural climate variations on long-range transport to the Arctic**

Climate change may alter the atmospheric transport of contaminants to and within the Arctic. One way to investigate this is to study the influence of present natural climate variations (such as the North Atlantic Oscillation (NAO) and/or the Arctic Oscillation (AO)) on transport of contaminants to and within the Arctic using an atmospheric transport model. The DEHM model has been used to study natural climate variations related to the AO and NAO by carrying out 24 years of model calculations.

A large part of the climate variability over the northern hemisphere is associated with variability in the NAO index (Figure 3.26). The NAO index is defined as the difference between surface pressures of the subtropical highs at the Azores and the subpolar lows at Iceland. The AO is defined as the surface-level pressure anomalies for the North Pole. As the NAO and AO are highly correlated only the NAO index was used in this assessment.

When the NAO is in a positive phase, negative low-pressure anomalies over the Icelandic region and throughout the Arctic, together with positive high-pressure anomalies across the subtropical Atlantic, tend to produce stronger westerly winds at mid-latitudes. The negative low-pressure anomalies throughout the Arctic trigger transport episodes from the Russian Arctic into the atmosphere over the Arctic Ocean.

For the European Arctic the total mean column level of SO_2+SO_4 for January to April is negatively correlated with the NAO index (Figure 3.27), which means that during periods with a negative NAO index there is higher sulfur transport from Europe to the Arctic. For the Canadian-USA-eastern Siberian Arctic the total mean column level of SO_2+SO_4 for January to April is positively correlated with the NAO index (Figure 3.27), which means that during periods with a positive NAO index there is more sulfur transport from eastern Russia into the Arctic and more transport from North America towards the area between Canada and Greenland.

Climate models project that large positive NAO events will occur more frequently in future, and that the rest of the

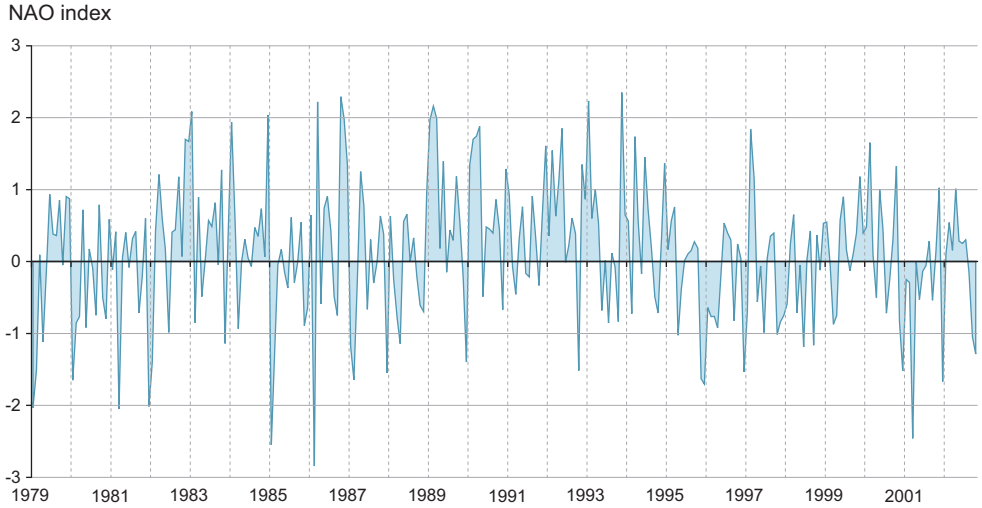


Figure 3.26. NAO index for the period 1979 to 2002.

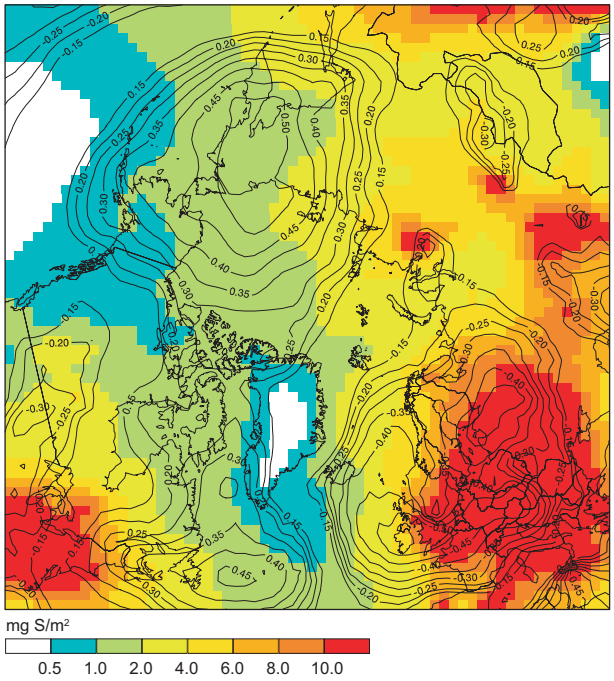


Figure 3.27. The total mean column level of SO_2+SO_4 for the period January to April for 1979 to 2002, with contour lines showing the correlation between the different monthly mean total column levels and the NAO index.

NAO frequency distribution will change little (see Coppola *et al.*, 2005). This would result in less transport from European sources and more transport from sources in eastern Russia and North America, resulting in lower concentrations in the European Arctic and higher concentrations in the Russian and North American Arctic.

3.7.4. **Scenarios**

The DEHM model with extensive chemistry was run for six emissions scenarios. These emissions scenarios are for the years 1990, 2000, 2010 and 2020, with 2010 and 2020 having two different emissions scenarios: the 'Maximum technically Feasible Reduction' (MFR) scenario and the 'Current LEgislation' (CLE) scenario (see section 2.3 for further details). For each emissions scenario the DEHM model was run using the same meteorological input for the period 1991 to 1993 in order to reduce the effects of meteorological variations on the model results.

The model results (see Figure 3.28) confirm the decrease in SO_x concentrations calculated for Station Nord (discussed in section 3.7.2) and estimate that both the mean SO_X concentrations and the total sulfur deposition almost halved between 1990 and 2000. The results for NO_y are similar to those for SO_x.

The outcome projected for the two future emissions scenarios (CLE and MFR) is a small decrease in both concentrations and deposition. Another important point is

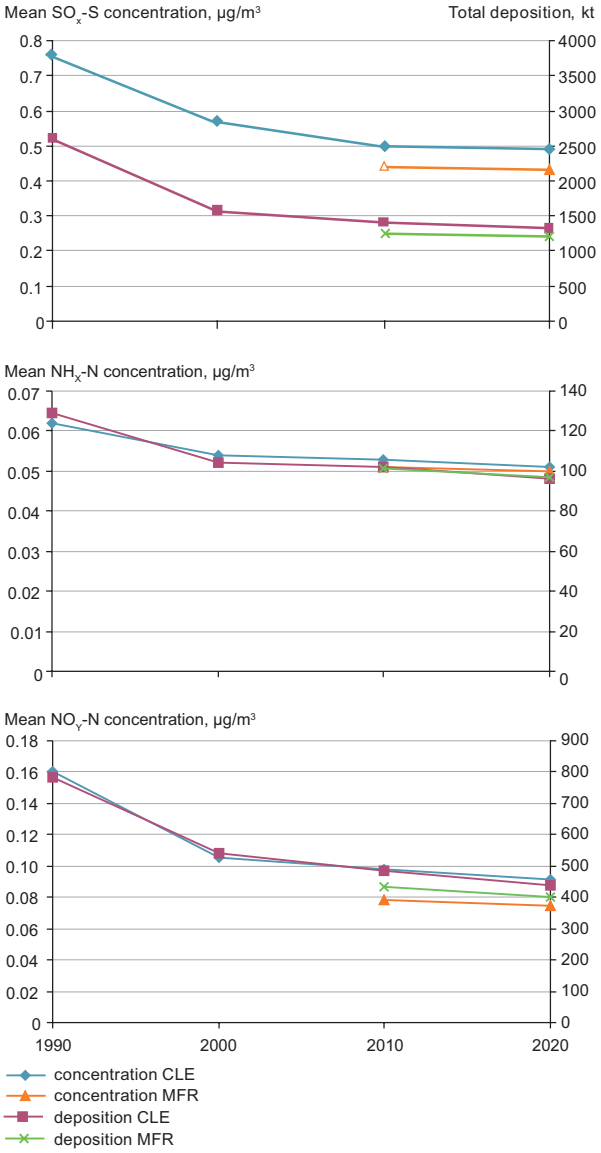


Figure 3.28. Total mean concentrations and total depositions of sulfur oxides, reduced nitrogen (NH_X) and airborne oxidized nitrogen (NO_y) for the area north of Arctic Circle for six emissions scenarios.

that even though the total emissions in the MFR scenario are a factor of 2 less than those in the CLE scenario this has only a minor effect on the total concentrations and deposition of SO_x and NO_x in the Arctic. Since most of the difference between the two scenarios is in the Asian emissions, this indicates that future emissions in Asia are likely to have only a small impact on acidification in the Arctic. Pollutants emitted in warmer regions such as Asia are not transported directly to the cold Arctic (see section 2.2). It is emissions in Eurasia that contribute most to acidification in the Arctic and so it is future changes in these emissions that are likely to have the greatest impact. These findings confirm the results in the previous AMAP assessment (AMAP, 1998).

Chapter 4
Arctic Haze

Patricia Quinn, Betsy Andrews, Ellsworth Dutton, Glenn Shaw, and Tuija Ruoho-Airola

4.1. The arctic haze phenomenon

It has been more than 50 years since observations of a strange haze, of unknown origin, were reported by pilots flying in the Canadian and Alaskan Arctic (Greenaway, 1950; Mitchell, 1956). Based on measurements at McCall Glacier in Alaska, Shaw and Wendler (1972) noted that the turbidity maximized in spring. First measurements of the vertical structure of the haze were made in an Alaskan 'bush' airplane with a hand-held sunphotometer (Shaw, 1975). At that time the origin of the haze was uncertain and was attributed to ice crystals seeded by open leads or blowing dust from riverbeds. It was only through 'chemical fingerprinting' of the haze that its anthropogenic source was revealed (Ottar *et al.*, 1986; Rahn *et al.*, 1977; Rahn and McCaffrey, 1979; Rahn, 1989). By the late 1970s the anthropogenic origin was clear but surprising since it was widely believed that aerosol was generally not transported more than a few hundred kilometers from its source regions. Experts from Europe and America convened at the first Arctic Air Chemistry Symposium at Lillestrom, Norway in 1978 and an informal measurement network was agreed upon. Spatial gradients soon showed the direction of flow and the surprisingly large extent of this anthropogenic cloud of pollution. A combination of intensive field programs and long-term measurements extending over the past thirty years confirmed the early conclusions that the haze is anthropogenic in origin due to emissions from Europe and the former Soviet Union that are transported to and trapped in the arctic air mass during the winter and early spring (Figure 4.1).

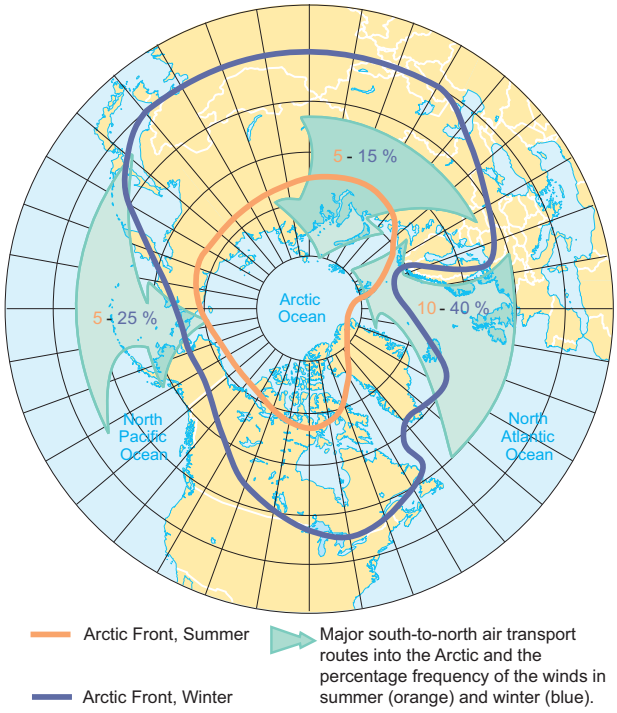


Figure 4.1. Mean position of the arctic air mass in winter (January) and summer (July), superimposed on the percentage frequency of major south-to-north transport routes into the Arctic in summer and winter (AMAP, 1998).

The haze comprises a varying mixture of sulfate and particulate organic matter and, to a lesser extent, ammonium, nitrate, dust, and black carbon (e.g., Li and Barrie, 1993; Quinn *et al.*, 2002). It is also rich in certain heavy metals which has allowed for the identification of particular industrial sources (e.g., Shaw, 1983; Rahn, 1989). Particles within the haze are well-aged with a mass median diameter of about 0.2 µm or less (e.g., Heintzenberg, 1980; Hoff *et al.*, 1983; Pacyna *et al.*, 1984; Shaw, 1984; Clarke, 1989; Leaitch *et al.*, 1989; Trivett *et al.*, 1989; Hillamo *et al.*, 1993). This particle size range is very efficient at scattering visible solar radiation since the peak in the particle surface-area size distribution is near the maximum efficiency for Mie scattering (Waggoner and Weiss, 1980; Shaw, 1987). The haze is also weakly absorbing due to the presence of black carbon (e.g., Hansen and Rosen, 1984; Noone and Clarke, 1988; Kahl and Hansen, 1989; Hopper *et al.*, 1994). The result of the strong scattering and weaker absorption is a noticeable reduction in visibility to a few kilometers or less. The 'weak' absorption may have large climatic influences when the dark colored haze spreads out over the highly reflecting snow and ice pack of the Arctic. The highly reflecting surface enhances aerosol-radiative interactions due to multiple scattering between the surface and the haze.

Several seasonally-dependent mechanisms contribute to the formation of arctic haze. Strong surface-based temperature inversions form in the polar night causing the atmosphere to stabilize. This cold and stable atmosphere inhibits turbulent transfer between atmospheric layers as well as the formation of cloud systems and precipitation; the major removal pathway for particulates from the atmosphere (Barrie *et al.*, 1981; Shaw, 1981, 1995; Heintzenberg and Larssen, 1983). In addition, meridional transport from the mid-latitudes to the Arctic intensifies during the winter and spring (Iversen and Joranger, 1985). The combination of these factors results in the transport of precursor gases and particulates to the Arctic and the trapping of the pollutant haze for up to 15 to 30 days (Shaw, 1981, 1995).

Aircraft and lidar measurements throughout the 1980s and 1990s revealed that the haze occurs primarily in the lowest five kilometers of the atmosphere and peaks in the lowest two kilometers (Leaitch *et al.*, 1984; Hoff, 1988; Pacyna and Ottar, 1988; Barrie, 1996). Throughout the haze season, the pollution layers are highly inhomogeneous both vertically (tens of meters to 1 km thick) and spatially (20 to 200 km in horizontal extent) (Radke *et al.*, 1984; Brock *et al.*, 1989).

Recent aircraft measurements of sulfate aerosol using a high time resolution technique revealed detailed information about the evolution of the vertical structure of the haze between February and May (Scheuer *et al.*, 2003). During early February, significant enhancements in sulfate aero-

sol are confined near the surface (< 2 km) as long-range transport from northern Eurasia occurs along low level, sinking isentropes (Klonecki *et al*., 2003). As the haze season progresses, enhanced sulfate occurs at higher altitudes (up to at least 8 km). Since vertical mixing is prohibited by the persistent low-level inversion (Kahl, 1990), the higher altitude haze layers are thought to be due to transport into the Arctic along vertically higher isentropes tracing back to increasingly warmer source regions in northern Eurasia. During early April, sulfate layers below 3 km begin to dissipate due to the beginning of solar heating and resulting mixing near the surface. However, more stable isentropic transport continues at higher altitudes. By the end of May, both the lower and higher altitude sulfate enhancements are significantly decreased due to the continued break-up of the inversion and the return of wet deposition.

Recent studies have provided evidence for an influence of natural climate variability on interannual changes in levels of arctic haze. Modeling the dispersion of anthropogenic emissions from northern hemisphere continents, Eckhardt *et al*. (2003) found that the North Atlantic Oscillation (NAO) influences pollution transport into the Arctic during the winter-spring haze season (see also section 3.7.3). During positive phases of the NAO, surface concentrations of modeled tracers in the arctic winter were found to be elevated by about 70% relative to negative phases. This difference was mainly due to a change in pathways of European pollution and, to a lesser extent, North American pollution to the Arctic both of which are enhanced during positive NAO phases. In addition, during positive NAO phases, significant positive correlations between the NAO and measured carbon monoxide concentrations were found at three arctic monitoring stations (Spitsbergen, Barrow, and Alert) confirming enhanced poleward transport of pollution from Europe, Asia, and North America. Similar but weaker correlations between the NAO and measured carbon monoxide concentrations were found for spring. Low correlations were found during summer and autumn.

During transport from the source regions to the Arctic, the pollutant-containing air masses have a high probability of reaching saturation and nucleating and precipitating clouds. It is not understood how so much material gets through a strongly scavenging system (Bowling and Shaw, 1992).

Arctic haze has been the subject of much study because of its potential to change the short and longwave radiation balance of the Arctic, to affect visibility, and to provide a source of contaminants to arctic ecosystems. The near surface concentration of aerosols at most places in the Arctic are about an order of magnitude lower than those found at more polluted and industrialized locations. At the same time, however, the affected areas are much larger in size and the affected ecosystems in the high Arctic are thought to be quite sensitive to gaseous and aerosol contamination.

It is not known what fraction of the arctic haze contaminants leave the Arctic and what fraction is deposited within the Arctic on land and sea surfaces. As the polar night ends, some of the pollution that has accumulated is released to the mid-latitudes (Penkett *et al*., 1993; Heintzenberg *et al*., 2002). It is known that haze contaminants (e.g., acidic sulfate and organics) end up in arctic ecosystems (Meijer *et al*., 2003; Wania, 2003) but the timing and mechanism of the scavenging from the atmosphere is not well understood. Measurements from a 5 m snow pit and 75 m ice core in Greenland indicated that there is a maximum in sulfate and black carbon deposition to the surface during the arctic haze season although there can be significant deposits throughout the year (Masclet *et al*., 2000). Similarly, sulfate concentrations as well as concentrations of other arctic haze tracers (lead, cadmium, and arsenic) in snow at sixteen sites across northwest Alaska were found to be highest in the later winter snow pack (Douglas and Sturm, 2004). The aerosols are removed from the atmosphere either through dry deposition or wet deposition with the mechanism of the latter most likely involving nucleation followed by precipitation in the form of ice crystals (Masclet *et al*., 2000). Since the timing of the buildup of the haze and the snow pack is similar and haze concentrations decline before the snow has fully melted, it is likely that haze contaminants first enter the ecosystem by being deposited in the snow (Douglas and Sturm, 2004). Contaminants deposited in the snow then end up in the tundra and rivers as the snow melts. The network of snow measurements across northwest Alaska showed spatially homogeneous concentrations of sulfate and trace elements suggesting little variability in atmospheric concentration or scavenging efficiency within the confines of the sampled region. By contrast, the acidity of the snow was much patchier indicating that competing acidifying and buffering sources determine the local pH. For example, pH was consistently higher in the Brooks Range than elsewhere due to mechanical weathering of carbonate rocks.

4.2. Trends in arctic haze

4.2.1. Chemical composition

Arctic haze is marked by a dramatic increase in concentrations of several key particulate pollutants during winter and early spring. The seasonal trend in the haze has been detected at several monitoring sites in the Arctic including Alert (82.46° N) in the Canadian Arctic, Station Nord in Greenland (81.4° N), Zeppelin (79° N) on the island of Svalbard, Barrow in Alaska (71.3° N), Karasjok (69.5° N) and Svanvik (69.45° N) in northern Norway, Oulanka (66.3° N) in northern Finland, and Janiskoski (69° N) in western Russia (see Figure 3.3). A time series for particulate sulfate concentrations is shown for these eight monitoring sites in Figure 4.2. Each site has a similar winter/early spring increase in sulfate with maximum concentrations reaching up to about 1 µg S/m^3. Monthly average concentrations in summer are less than 0.03 µg S/m^3. Non-marine sulfate (SO_4*) makes up about 30% of the submicron mass during the haze season (Barrie *et al*., 1981; Quinn *et al*., 2000, 2002). Figure 4.3 shows the time series for particulate nitrate concentrations at Alert and Barrow, two sites which have a clear seasonal pattern for this species. Maximum concentrations are near 0.04 µg N/m^3. Other species also indicative of continental sources (ammonium and non-marine potassium for biomass burning and magnesium and calcium for dust) have maximum concentrations in winter and spring indicating long range transport to the Arctic (Quinn *et al*., 2002).

Natural aerosol chemical components display seasonal cycles quite different from anthropogenic components. Sea salt concentrations are highest at Alert and Barrow from November through February (Quinn *et al*., 2002). The winter maximum has been attributed to seasonally high winds

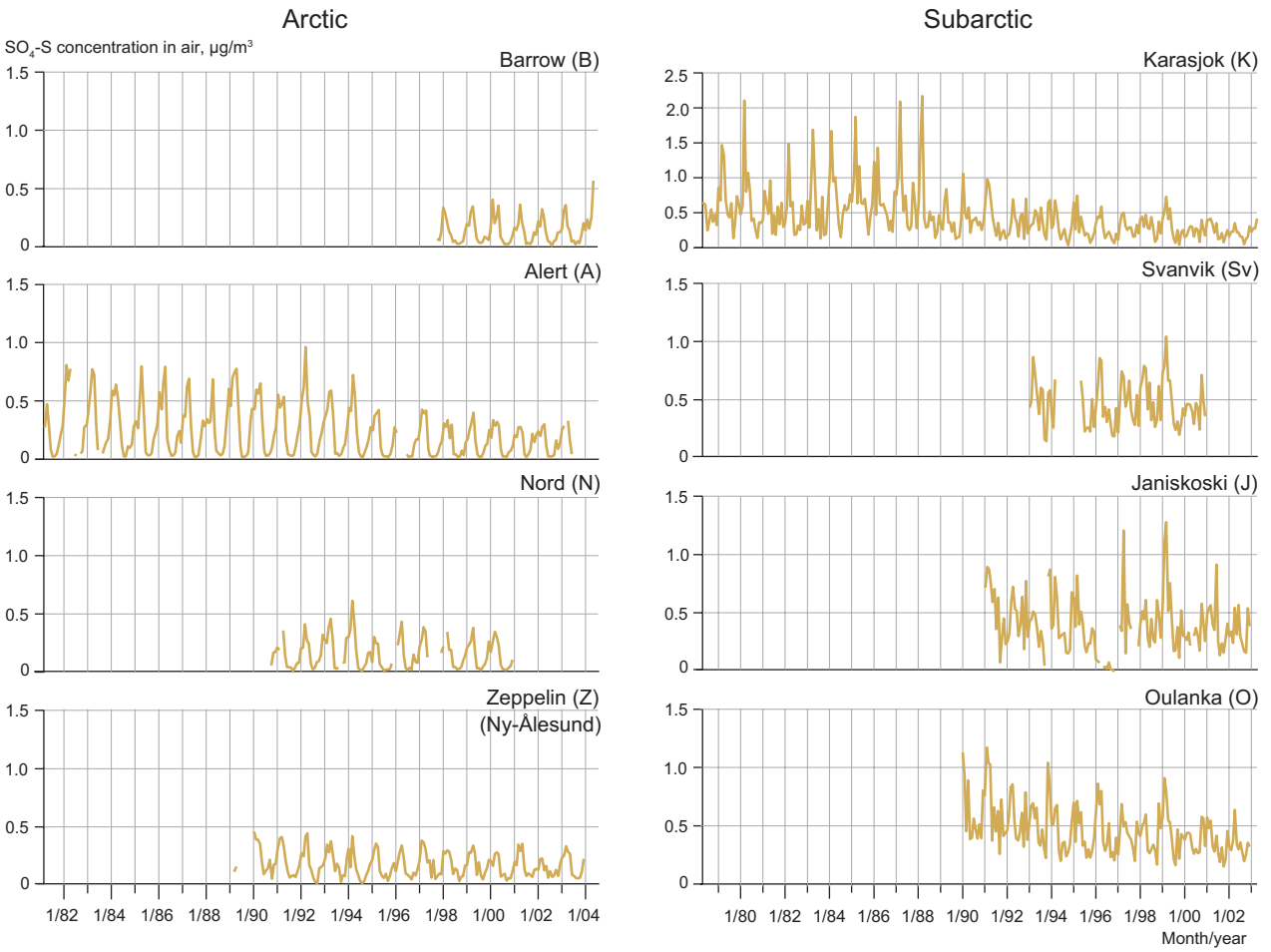
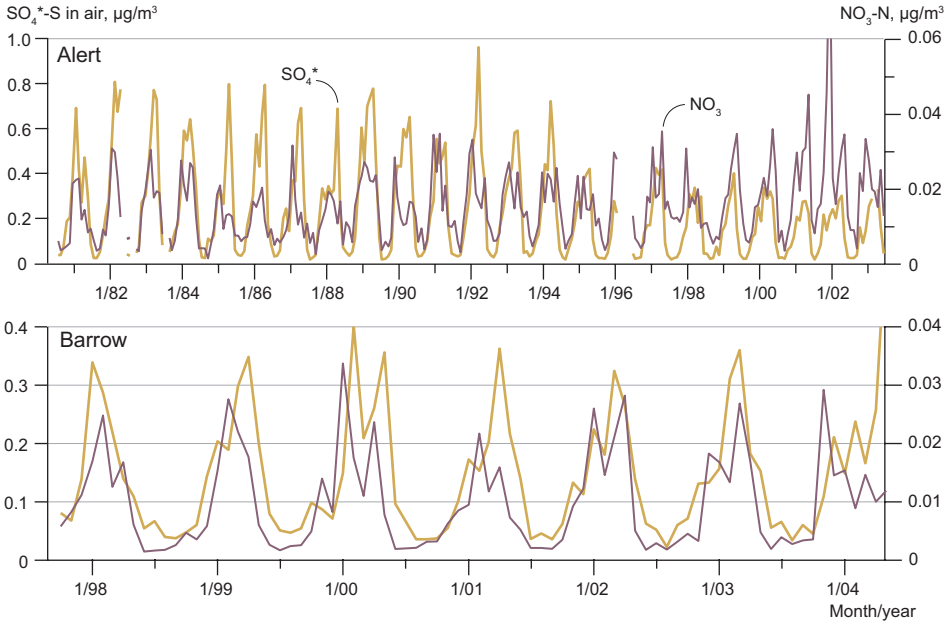


Figure 4.2. Time series of monthly averaged particulate sulfate concentrations at eight arctic monitoring sites. Data made available for Alert by the Canadian National Atmospheric Chemistry (NAtChem) Database and Analysis System, for Barrow by NOAA PMEL (http://saga.pmel.noaa.gov/data/), and for the other stations by EMEP (http://www.emep.int/).



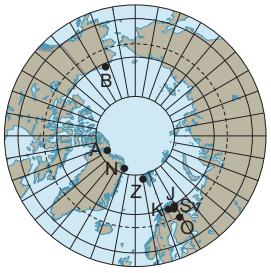


Figure 4.3. Time series of monthly averaged particulate sulfate and nitrate concentrations at Alert (Canada) and Barrow (Alaska). Data sources as for Figure 4.2.

in high-latitude source regions of the Pacific and Atlantic Oceans and long-range transport to the Arctic (Sturges and Barrie, 1988; Sirois and Barrie, 1999). Supermicron sea salt aerosol peaks during the summer months at Barrow when the ice pack extent is at a minimum (Quinn *et al.*, 2002). Atmospheric methanesulfonic acid (MSA^-) is derived solely from the oxidation of biogenically produced dimethylsulfide (DMS). Concentrations of MSA^- begin to increase in late June as the sea ice recedes and phytoplankton productivity in surface waters begins. This is also when DMS that has been trapped under the ice is released (Ferek *et al.*, 1995).

Two years of measurements at a site in northern Finland revealed that particulate organic matter made up, on average, 22% of the total fine aerosol mass (range 3 to 69%) (Ricard *et al.*, 2002). Correlation of particulate organic

matter with sulfate* was low indicating that they had different sources. Since particulate organic matter displayed a seasonal cycle with maximum concentrations in summer, it is most likely to result from biogenic emissions and/or enhanced oxidation processes. A small increase in organic acids as early as February/March may indicate photooxidation at polar sunrise, as was pointed out for Alert by Kawamura *et al*. (1996).

The longest record of sulfate concentrations in the Arctic (1980 to present at Alert, Canada) revealed no change in sulfate concentrations during the 1980s (Sirois and Barrie, 1999). This lack of a trend was attributed to little change in emissions in the former Soviet Union between 1985 and 1990. Beginning in 1991, sulfate and other measured anthropogenic constituents (lead, zinc, copper, excess vanadium and manganese, and ammonium) began to decline suggesting that reduced industrial activity in the early years of the new Eurasian republics had led to lower levels of pollutants reaching the Arctic.

A combined modeling and measurement analysis of sulfate concentrations at Station Nord in northern Greenland indicated a decreasing trend throughout the 1990s (Heidam *et al*., 2004). The analysis was able to account for scatter in measured concentrations due to changing meteorology. With the meteorological variability removed, it was possible to attribute the decrease in concentrations to a reduction in emissions. The model that was used estimated that more than 70% of the sulfur measured at Station Nord was emitted from the area making up the former Soviet Union which indicated that emissions from the region decreased significantly during the 1990s. This result is supported by the 50% decrease in Russian sulfur emissions reported to EMEP during the 1990s (Vestreng, 2003). It is not clear from this analysis how reductions from Western Europe and North America influenced sulfate concentrations at Station Nord.

Sulfate concentrations also decreased during the 1990s at several other sites in the Arctic. Data for updating trend analyses through the first few years of the 21st century are now available at many arctic sites.

As pointed out by Macdonald *et al*. (2005), the detection of recent trends in the Arctic is difficult due to the combination of short or incomplete data records at some sites and interference from natural variations on seasonal, annual, and decadal time scales. To remove seasonal variability from the trend analyses, this assessment has focused on average monthly concentrations for March and April. The existence of a monotonic increasing or decreasing trend in the time series was tested with the nonparametric Mann-Kendall test at significance levels $p<0.001$, $p<0.01$, and $p<0.1$ as a two-tailed test (Gilbert, 1987). The estimate for the slope of a linear trend was calculated with the nonparametric Sen's method (Sen, 1968a). The Mann-Kendall test is suitable for cases where the trend may be assumed to be monotonic such that no seasonal or other cycle is present in the data. In the Mann-Kendall test, missing data values are allowed and the data need not conform to any particular distribution. The Sen's slope is the median of the slopes calculated from all pairs of values in the data series. The Sen's method is not greatly affected by data outliers and can be used when data are missing (Salmi *et al*., 2002). Trends are only reported for a significance level of <0.1, i.e., when the probability of no trend is 10% or less.

Average monthly concentrations of sulfate for March and April are shown in Figure 4.4 along with the Sen's slope estimates for a significance level of <0.1. The significance of the trend, slope estimate, and percentage change over the measurement period are given in Table 4.1. For sites with a significant trend (based on the Mann-Kendall test), sulfate concentrations decreased by 30 to 60% between 1990 and present. The decreasing trend in sulfate at Alert detected through the 1990s has continued into the present century. In addition, sulfate has decreased significantly at Zeppelin, Karasjok, and Oulanka.

Corresponding data are shown for nitrate in Figure 4.5 and Table 4.2. In contrast to sulfate, nitrate increased during April at Alert for the long-term period of 1981 to 2004 and during March for the shorter term more recent period of 1990 to 2004. Between 1990 and 2004, nitrate concentrations increased by about 50%. More measurements are needed at both Alert and Barrow to verify these trends, however.

4.2.2. **Optical properties**

The seasonality and trends in arctic haze are clearly seen in time series data of light absorption and scattering by

Table 4.1. Change in monthly average sulfate concentrations at a range of arctic monitoring stations for March and April. Significance of the trend, Sen's slope estimate, and percentage change per period are listed.

			α[a]	Slope[b]	Percentage change over period
Alert	SO_4*	March 1981–2002	0.01	-0.066	- 66
		April 1981–2003	0.001	-0.079	- 71
		March 1990–2002	0.001	-0.088	- 59
		April 1990–2003	0.01	-0.086	- 63
Nord	SO_4	March 1994–2002			
		April 1994–2002			
Zeppelin	SO_4	March 1990–2003	0.1	-0.094	-33
		April 1990–2003	0.1	-0.079	-27
Barrow	SO_4*	March 1998–2004			
		April 1998–2004			
Karasjok	SO_4	March 1978–2003	0.001	-0.044	- 80
		April 1978–2003	0.001	-0.021	- 59
		March 1990–2003	0.1	-0.017	- 40
		April 1990–2003	0.01	-0.025	- 48
Svanvik	SO_4	March 1993–2000			
		April 1993–2000			
Janiskoski	SO_4	March 1991–2002			
		April 1991–2002			
Oulanka	SO_4	March 1990–2002	0.1	-0.034	- 45
		April 1990–2002	0.01	-0.043	- 56

[a] Significance level, α, of the Mann-Kendall test. A significance level of 0.001 indicates a 0.1% probability of no trend. No value indicates a significance level of >0.1; [b] Sen's nonparametric method was used to estimate the slope of the existing trend as change per year (μg m^3/yr).

Table 4.2. Change in monthly average nitrate concentrations at Alert (Canada) and Barrow (Alaska) for March and April. Significance of the trend, Sen's slope estimate, and percentage change per period are listed.

			α[a]	Slope[b]	Percentage change over period
Alert	NO_3	March 1981–2000			
		April 1981–2000	0.1	0.0006	67
		March 1990–2000	0.05	0.0008	50
		April 1990–2000			
Barrow	NO_3	March 1998–2004			
		April 1998–2004			

[a] Significance level, α, of the Mann-Kendall test. A significance level of 0.001 indicates a 0.1% probability of no trend. No value indicates a significance level of >0.1; [b] Sen's nonparametric method was used to estimate the slope of the existing trend as change per year (μg m^3/yr).

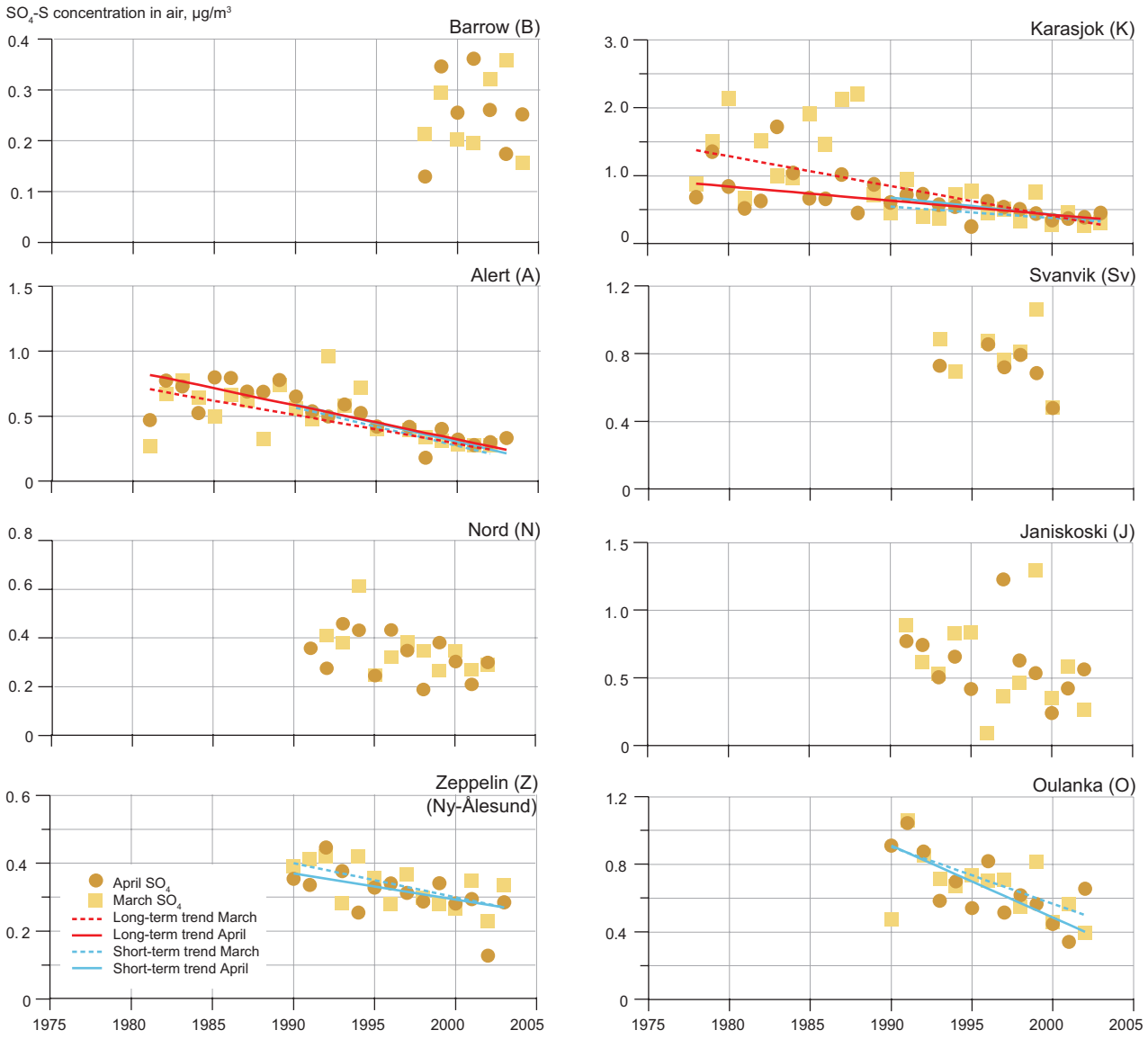
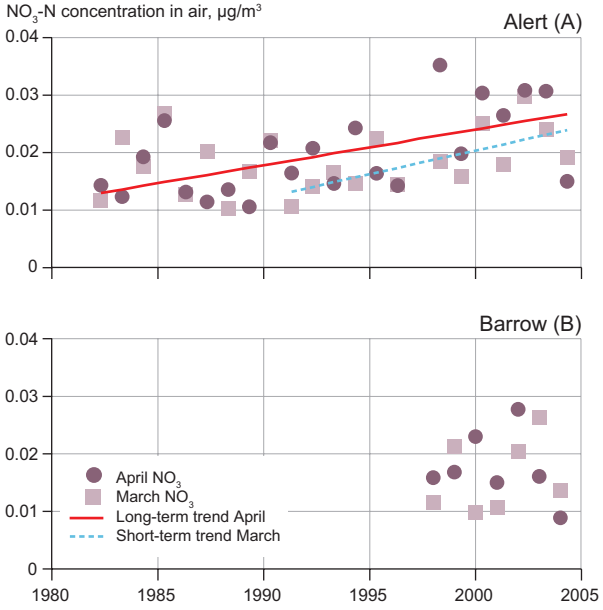


Figure 4.4. Monthly averaged sulfate concentrations for March and April at eight arctic monitoring sites and Sen's slope estimates for the long term (approximately 1980 through the available data) and short term (approximately 1990 through the available data) trends. Trend lines are not shown for $\alpha > 0.1$. Data sources as for Figure 4.2.



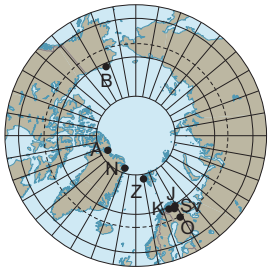


Figure 4.5. Monthly averaged nitrate concentrations for March and April at Alert (Canada) and Barrow (Alaska) and Sen's slope estimates for the long term (approximately1980 through the available data) and short term (approximately 1990 through the available data) trends. Trend lines are not shown for $\alpha > 0.1$. Data sources as for Figure 4.2.

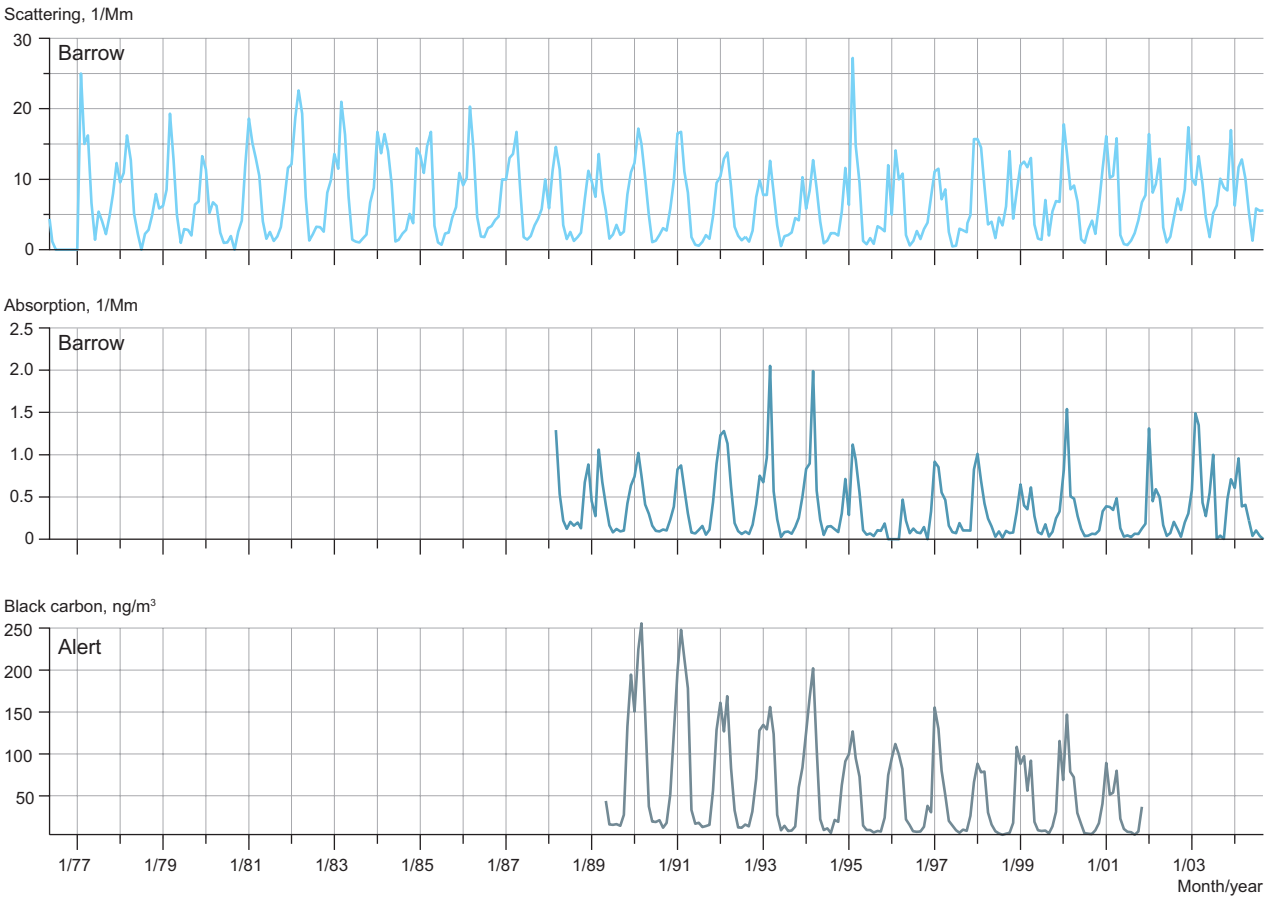


Figure 4.6. Monthly averaged light scattering and absorption at 550 nm by <10 µm aerosol at Barrow (Alaska) and black carbon concentrations at Alert (Canada). Data sources as for Figure 4.2.

aerosols measured at the surface (Figure 4.6) (Bodhaine, 1989) and in total column aerosol optical depth (Dutton *et al*., 1984; Herber *et al*., 2002). Bodhaine and Dutton (1993) reported that both aerosol scattering and optical depth measurements at Barrow peaked in 1982 followed by a factor of two decrease between 1982 and 1992. The decrease was only apparent during March and April corresponding to the time of year when arctic haze is most pronounced. It was hypothesized that the decrease in the haze was most likely to be due to a combination of a reduction in the output of pollution aerosol by Europe and the former Soviet Union and stricter pollution controls in Western Europe. The decreases in aerosol scattering and optical depth at Barrow during this ten-year period are not equal to the known reductions in sulfate emissions, however, indicating that other factors such as changes in transport processes could have played a role (Jaffe *et al*., 1995). Since the NAO was in a positive phase between the early 1980s and 1990s, implying enhanced transport of pollutants to the Arctic (Hurrell and van Loon, 1997), it appears not to have been responsible for the observed decrease in arctic haze during this period.

An update of the monthly averaged light scattering data analysis originally performed by Bodhaine and Dutton (1993) is shown in Figures 4.7 and 4.8 with data at Barrow extending through 2005. Also shown are Sen's slope estimates for three periods: 1977–2005, 1982–1996, and 1997–2005. The significance of the detected trends, slope estimates, and percentage change in scattering for these periods are given in Table 4.3. For March, there is a significant decreasing trend in scattering over the entire measurement period of 1977 to 2005. Breaking the period into two smaller intervals reveals a decreasing trend from 1982 to 1996 which reverses to yield an increasing trend from 1997 to 2005. The increasing trend for March is significant at the 0.05 level indicating that there is a probability of 95% that the trend exists.

A similar analysis for light absorption at Barrow indicates an overall significant decrease between 1988 and 2005 for March. Trends between 1997 and 2005 are unclear (Figures 4.7 and 4.8; Table 4.3). Sharma *et al*. (2006) reported an increasing trend in absorption for the winter months (defined as January through April) at Barrow between 2000 and 2003.

Black carbon concentrations measured at Alert show a decreasing trend for both March and April over the measurement period of 1990 through 2001 (Figures 4.7 and 4.8; Table 4.3). Sharma *et al*. (2004) reported a decrease in black carbon (the main light absorber in the arctic atmosphere) of 56% for the winter/spring season from 1989 to 2002 at Alert. However, Sharma *et al*. (2006) reported an increase during winter (January to April) beginning in 2000.

A recent study using a general circulation model suggested that one of the major sources of arctic soot today is southern Asia (Koch and Hansen, 2005) due to increasing emissions from industrial and biofuel combustion. This suggestion has been refuted by Stohl (2006), however, based on the long passage from pollution sources in southern and eastern Asia to the Arctic relative to more rapid transport from Europe and northern Asia. Using a particle dispersion model, it was found that, for a transport time of five days, the southern Asia black carbon

contribution is about 1 to 2% of the European source contribution near the surface and 3 to 5% for the total air column.

Results reported by Stohl (2006) identify boreal and temperate forest fires, especially Siberian fires, as a significant source of black carbon during the summer. In intense fire years, boreal forest fires may be the dominant source of black carbon for the Arctic. Measurements at Alert and Barrow show there is a strong seasonal cycle in potassium*, a tracer for biomass burning, with minimum values in the summer and maximum values in the winter (e.g., Quinn *et al*., 2000). Based on these data, the impact of summertime forest fire emissions on low altitude surface sites within the Arctic is relatively small compared to winter emissions. There is evidence, however, of pyrocumulonimbus injection of smoke from boreal forest fires to the upper atmosphere (Fromm *et al*., 2005). In addition, biomass burning signatures have been observed in the snow at the high altitude (3200 m) site of Summit, Greenland (Dibb *et al*., 1996). The fraction of this material that is deposited to lower elevations throughout the Arctic is unknown. More measurements coupled with modeling studies are required to identify sources of black carbon to the Arctic and to assess trends in black carbon and light absorption by aerosols.

An extension of the Barrow aerosol optical depth (AOD) data through 2002 shows a continued decrease through the mid-1990s (Figure 4.9). Monthly averaged values of AOD anomalies (relative to a base of non-volcanic years) for March show a continued decline through 2002. However, the AOD anomalies for April indicate an increase between

Table 4.3. Change in monthly average light scattering and absorption at Barrow (Alaska) and black carbon at Alert (Canada) for March and April. Significance of the trend, Sen's slope estimate, and percentage change during the period are listed.

			α[a]	Slope[b]	Change during the period %
Barrow	σ_{sp}[c]	March 1977–2005	0.01	-2.4 E-07	- 41
		April 1977–2005			
		March 1982–1996	0.01	-5.7 E-07	- 42
		April 1982–1996	0.01	-6.6 E-7	- 56
		March 1997–2005	0.05	7.8 E-07	46
		April 1997–2005			
Barrow	σ_{ap}[c]	March 1988–2005	0.1	-3.9 E-08	-61
		April 1988–2005			
		March 1988–1996			
		April 1988–1996			
		March 1997–2005			
		April 1997–2005			
Alert	BC[d]	March 1990–2001	0.001	-17	- 88
		April 1990–2001	0.05	-8.3	-63
		March 1997–2001	0.1	-3.9	-20
		April 1997–2001			

[a] Significance level, α, of the Mann-Kendall test. A significance level of 0.001 indicates a 0.1% probability of no trend. No value indicates a significance level of > 0.1; [b] Sen's nonparametric method was used to estimate the slope of the existing trend as change per year (mm/yr for scattering and absorption, ng m^3/yr for black carbon); [c] <10 µm aerosol; [d] BC: black carbon.

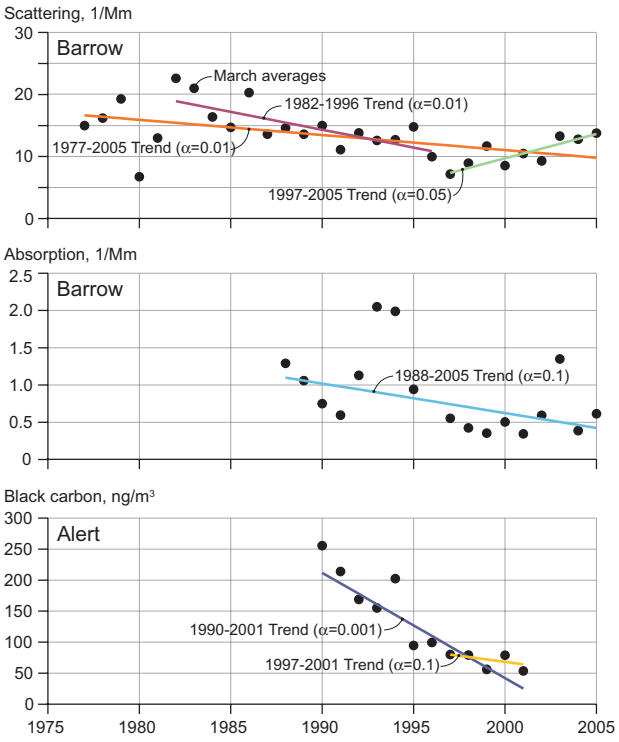


Figure 4.7. Monthly averaged concentrations for March of light scattering and light absorption at 550 nm for <10 µm aerosol at Barrow (Alaska) and black carbon concentrations at Alert (Canada). Lines indicate the Sen's slope estimate for the periods indicated and the α value indicates the significance level of the trend (α = 0.001 indicates there is a 0.1% probability that the trend does not exist). Trend lines are not shown for α >0.1. Data sources as for Figure 4.2.

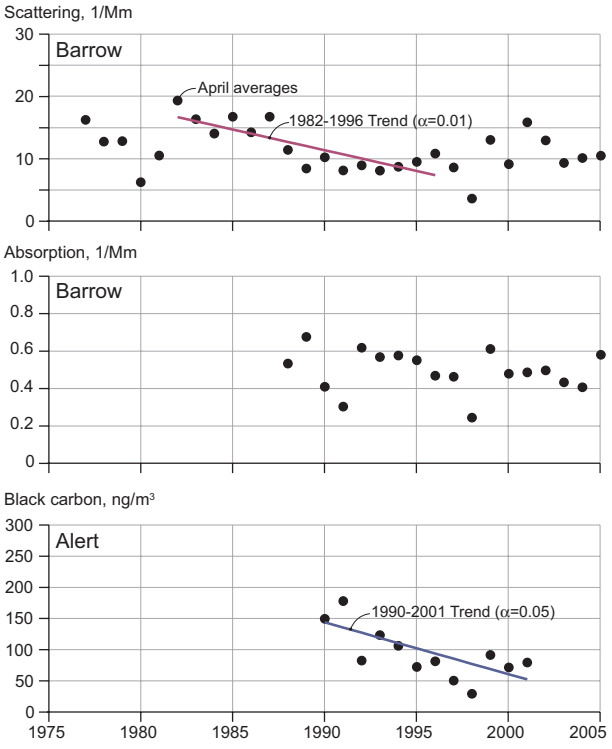


Figure 4.8. Monthly averaged concentrations for April of light scattering and light absorption at 550 nm for <10 µm aerosol at Barrow (Alaska) and black carbon concentrations at Alert (Canada). Lines indicate the Sen's slope estimate for the periods indicated and the α value indicates the significance level of the trend (α = 0.001 indicates there is a 0.1% probability that the trend does not exist). Trend lines are not shown for α >0.1. Data sources as for Figure 4.2.

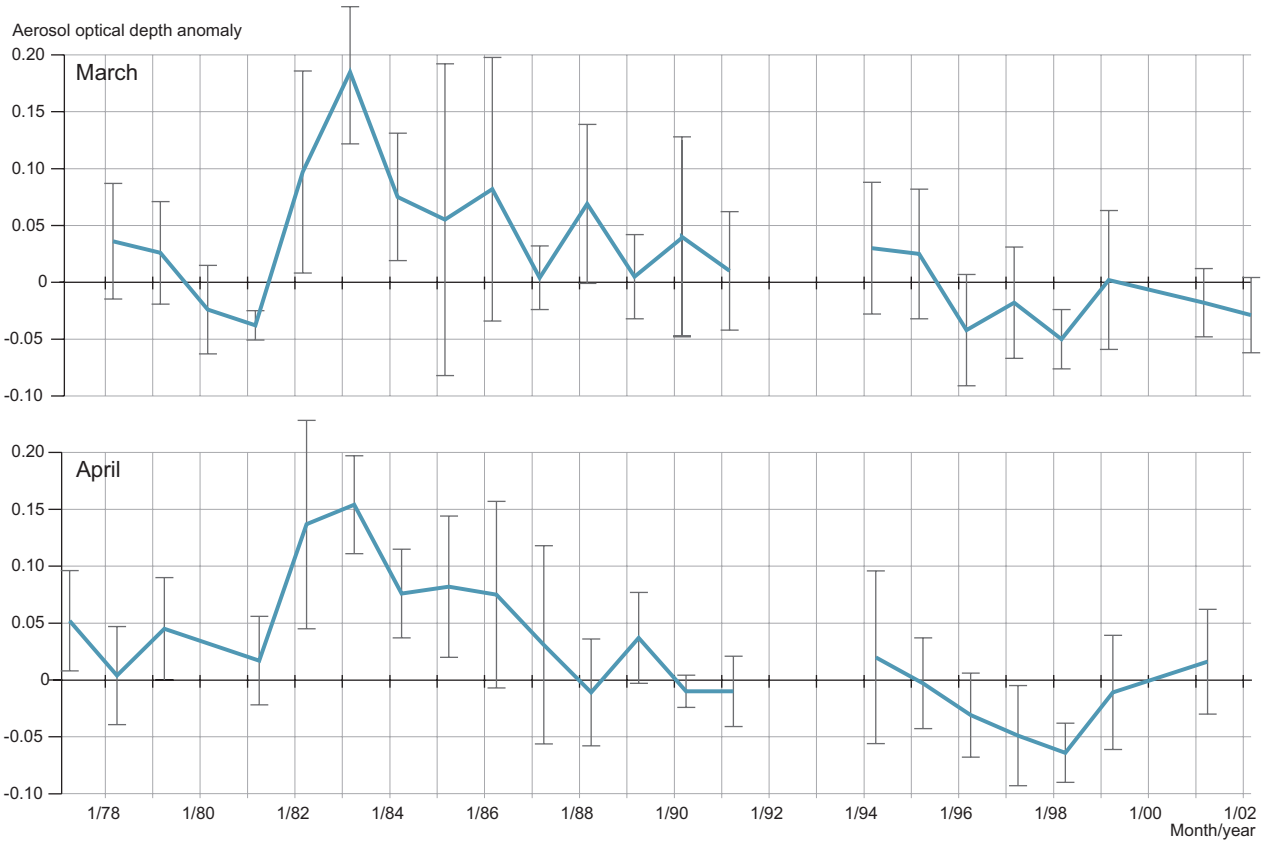


Figure 4.9. Monthly averaged aerosol optical depth anomalies at Barrow (Alaska) for March and April. The anomalies are relative to a base of non-volcanic years. Data from 1992 and 1993 were removed due to stratospheric aerosol influx from the Pinatubo eruption in 1991. Vertical lines represent 1 standard deviation of the monthly mean (data provided by NOAA CMDL).

1998 and 2001 where the currently available data record ends. In contrast to the Barrow trend through the 1990s, Herber *et al*. (2002) reported a slightly increasing trend in AOD (1% per year) at Koldewey station in Ny-Ålesund, Spitsbergen, between 1991 and 1999.

4.3. Effects of aerosol on the climate system in the Arctic

4.3.1. Direct effects

The direct effects of aerosols on the radiation balance in the Arctic are due to the absorption and scattering of radiation by the aerosol. The Arctic is thought to be particularly sensitive to changes in radiative fluxes imposed by aerosols because of the small amount of solar energy normally absorbed in polar regions (Valero *et al*., 1989). Arctic haze is present as a layer of light-absorbing material over a highly reflective ice/snow surface. Shaw and Stamnes (1980) first realized that the absorbing nature of arctic haze would have a significant impact on the energy balance of the Arctic. Several early calculations using 1-D radiative transfer models estimated that the diurnally averaged atmospheric warming due to the layer ranged between 2 and 20 W/m^2, with a corresponding depletion of the solar flux at the surface of 0.2 to 6 W/m^2 (Porch and MacCracken, 1982; Leighton, 1983; Blanchet and List, 1987; Emery *et al*., 1992; Shaw *et al*., 1993). These estimates agreed with direct measurements from wideband sun photometers (Mendonca *et al*., 1981). Heating rates of about 0.1 to 0.2 K/day were measured by Valero *et al*. (1989) during AGASP (Arctic Gas and Aerosol Sampling Program) II and by Treffeisen *et al*. (2004) during the ASTAR (Arctic Study of Tropospheric Aerosols, Clouds, and Radiation) 2000 campaign in Svalbard. The AASE (Airborne Arctic Stratospheric Expedition) II flights in winter 1992 revealed soot-contaminated arctic aerosols at altitudes of 1.5 km. Pueschel and Kinne (1995) calculated that this layer of aerosols could heat the earth–atmosphere system above surfaces of high solar albedo (ice/snow) even for single scattering albedos as high as 0.98. Hence, a modest amount of black carbon in the haze layers can result in a measurable contribution to diabatic heating.

MacCracken *et al*. (1986) estimated that the cooling of the surface due to absorption of solar radiation by the haze layers would be compensated by infrared emission from the atmosphere to the surface. The infrared emission is expected to dominate during the polar night when longwave radiation controls the energy budget of the Arctic. If the haze particles deliquesce thereby taking up water and growing to cloud droplet or ice crystal size, their longwave impact will be enhanced. Measurements made on Svalbard when the sun was below the horizon indicate that arctic haze can have a measurable direct thermal radiative forcing altering the flux of downward longwave radiation by up to +3 to +4.7 W/m^2 and the outgoing longwave radiation by -0.23 to +1.17 W/m^2 (Ritter *et al*., 2005).

The vertical distribution of the absorbing haze layers does not affect the radiation budget at the top and bottom of the atmosphere (Cess, 1983) but may affect atmospheric circulation and climate feedback processes.

4.3.2. **Indirect effects**

The indirect effects of aerosols on irradiances in the Arctic result from the impact of aerosol particles on the microphysical properties of clouds. Enhanced aerosol particle concentrations increase solar cloud albedo by increasing the number concentration and decreasing the average size of cloud droplets provided the liquid water content in the clouds remains constant (Twomey, 1977). An increase in the number of pollution aerosol particles that act as cloud condensation nuclei will affect arctic stratus and stratocumulus by increasing the cloud droplet number concentration which results in more radiation being reflected back to space (Albrecht, 1989; Twomey, 1991). The relatively low aerosol number concentrations in the Arctic results in a large percentage of particles activating during cloud formation. Hence, changes in aerosol properties are likely to have a significant impact on microphysical and optical cloud properties. As the cloud droplet number concentration increases, cloud droplet size decreases which reduces drizzle formation and increases cloud coverage and lifetime potentially leading to less deposition of haze contaminants within the Arctic (Hobbs and Rangno, 1998).

Garrett *et al*. (2004) showed that low-level arctic clouds are highly sensitive to particles that undergo long-range transport during winter and early spring. The sensitivity was detected as higher cloud droplet number concentrations and smaller cloud droplet effective radii compared to summertime clouds exposed to particles nucleated in the Arctic from local biogenic sources. In addition, arctic stratus appears to be more sensitive to pollutant particles than clouds outside the Arctic. The most significant effect of the change in cloud properties due to arctic haze may be on cloud emissivity. A decrease in droplet effective radius in these optically thin clouds will increase the infrared optical depth and thus the infrared emissivity (Curry and Herman, 1985; Garrett *et al*., 2002). The result is expected to be an increase in downwelling infrared irradiances from the cloud and an increase in the rate of springtime snow pack melting (Zhang *et al*., 1996).

According to observations during the SHEBA (Surface Heat Budget of the Arctic Ocean) experiment, supercooled cloud droplets are common in the Arctic even at temperatures of -20 °C or lower (Curry, 1995). The sulfate-containing pollution aerosol within arctic haze is also thought to affect ice nucleation. Models estimate that aerosols containing sulfuric acid produce fewer ice nuclei than nearly insoluble aerosols (Blanchet and Girard, 1995). Measurements corroborate this finding. Borys (1989) reported that arctic haze aerosol had lower ice nuclei concentrations, a lower ice nuclei to total aerosol fraction, and slower ice nucleation rates than aerosol from the remote unpolluted troposphere. The reduction in ice nuclei leads to a decrease in the ice crystal number concentration and an increase in the mean size of ice crystals (Girard *et al*., 2005). As a result, the sedimentation and precipitation rates of ice crystals increase leading to an increase in the lower troposphere dehydration rate and a decrease in the downwelling infrared irradiances from the cloud. Using a 1-D simulation and observations from Alert (Canada), Girard *et al*. (2005) found that a cloud radiative forcing of -9 W/m^2 may occur locally as a result of the enhanced dehydration rate produced by sulfate aerosol. The mechanism by which ice nuclei concentrations are decreased in the presence of sulfuric acid aerosol has yet to be explained and warrants further research. If this mechanism applies to much of the Arctic, it could explain the cooling tendency in the eastern high Arctic during winter.

Because of the combination of the static stability of the arctic atmosphere, the persistence of low level clouds, and the relatively long lifetime of aerosols during the haze season, the impact of aerosols on cloud microphysical and optical properties may be greater in the Arctic than elsewhere on earth (Curry, 1995; Garrett *et al*., 2004). The winter/spring occurrence of arctic haze events enables the study of anthropogenic influences against a very clean atmospheric background. In other regions of the globe, a reliable distinction between natural and anthropogenic effects is more difficult. In this sense, the Arctic is a natural laboratory for studying the anthropogenic portion of the aerosol–cloud–radiation interactions.

4.3.3. **Surface albedo**

Surface albedo affects the magnitude and sign of climate forcing by aerosols. Absorbing soot deposited onto the surface via wet and dry deposition affects the surface radiation budget by enhancing absorption of solar radiation at the ground and reducing the surface albedo (Warren and Wiscombe, 1980) (Figure 4.10). Clarke and Noone (1985) found a 1 to 3% reduction in snow albedo due to deposited black carbon with another factor of three reduction as the snow ages and black carbon becomes more concentrated. Hansen and Nazarenko (2004) estimated that soot con-

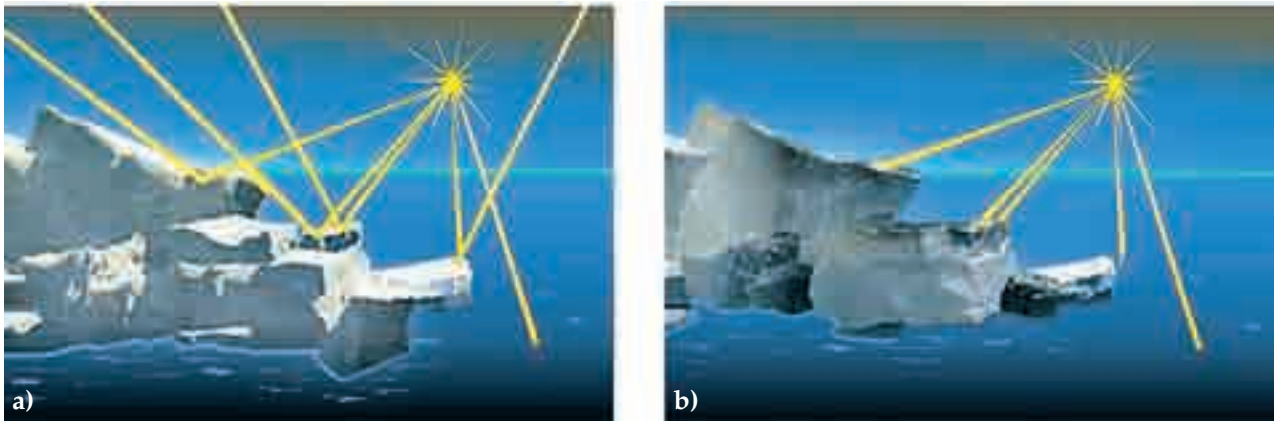


Figure 4.10. Impact of soot deposited onto snow and ice surfaces in the Arctic. Polar ice reflects light from the sun back to space (a). As the ice begins to melt, less light is reflected and more is absorbed by the oceans and surrounding land leading to an increase in overall temperature and further melting. Darker, soot-covered ice reflects even less light and, thus, enhances the warming (b) (Source: NASA).

tamination of snow in the Arctic and the corresponding decrease in surface albedo yields a positive hemispheric radiative forcing of +0.3 W/m^2. The resulting warming may lead to the melting of ice and may be contributing to earlier snowmelts on tundra in Siberia, Alaska, Canada, and Scandinavia (Foster *et al*., 1992).

Clearly, the radiative impacts of pollutant aerosols in the Arctic are complex. Complex feedbacks between aerosols, clouds, radiation, sea ice, and vertical and horizontal transport processes complicate the impact as do potentially competing effects of direct and indirect forcing. As a result, the magnitude and sign of the forcing for the Arctic are not yet well understood.

4.4. Summary

Based on measurements of sulfate aerosol – a main constituent of arctic haze – and light scattering and extinction, the amount of haze and haze precursors reaching the Arctic was either relatively constant or decreasing between the 1980s and early 1990s (Bodhaine and Dutton, 1993; Sirois and Barrie, 1999; Heidam *et al*., 2004). The updated trends in light scattering presented here show a continued decrease through the late 1990s with an increase in the first years of the 21st century at Barrow (Alaska). There also is evidence, although not as strong, of an increasing trend in black carbon over this period at Alert (Canada). Sulfate appears to have continued decreasing into the 21st century based on measurements at Alert in the Canadian Arctic, Zeppelin on the island of Svalbard, Karasjok in northern Norway, and Oulanka in northern Finland. On the other hand, nitrate appears to be increasing at Alert with an unclear trend at Barrow. Continued measurements coupled with chemical transport models are required to better define emerging trends and to assess their causes.

Arctic haze is generally understood to consist of anthropogenically-generated material and has often been attributed to sources in central Eurasia (Shaw, 1983). There are examples, however, of Asian dust entering the Alaskan sector of the Arctic from as long ago as the mid-1970s (e.g., Rahn *et al*., 1981). Recent modeling studies yield conflicting results on whether southern Asia is a significant source of pollutants to the Arctic or not. Given the rapid industrialization of China, the increasing amounts of pollution being transported over long distances, and indications of increasing concentrations of absorbing aerosol in the Arctic more research is warranted to document the contribution of this source to arctic haze and to determine its climate impact on the Arctic. A warming climate has been forecast to result in large increases in the areal extent of fires within Russian and Canadian boreal forests (Stocks *et al*., 1998). Hence, boreal forest fires are another source to be monitored to determine their impact on black carbon concentrations in atmospheric aerosol as well as black carbon that is deposited onto snow and ice surfaces.

Other key atmospheric species have a distinct seasonality in the Arctic. There is evidence of the enrichment of halogens in arctic air masses in late winter and spring. Since these compounds tend to peak later in the year, it is thought that they are produced photochemically. More research is required to partition their sources (e.g., anthropogenic, especially coal combustion vs. marine), to investigate their numerous and complex chemical pathways, and to assess their environmental impacts. Of special note is iodine, which shows a bimodal seasonal behavior, peaking in both spring and autumn (Sturges and Shaw, 1983).

The direct radiative effect of arctic haze has been estimated with 1-D radiative transfer models which find a warming in the atmosphere due to absorption of solar radiation and a concurrent cooling at the surface. These estimates are highly sensitive to the assumed properties of the aerosol in the haze. Despite the many research activities devoted to the characterization of arctic haze since the 1970s, measurements of arctic aerosols are not extensive or well distributed in space or time, which limits the accuracy of the estimates of both the direct and indirect radiative forcing. Treffeisen *et al*. (2004) designed an approach based on cluster analysis for integrating aircraft, ground-based, and long-term data sets for use in 3-D climate models. The accurate evaluation of climate forcing by arctic haze requires such data sets coupled with 3-D climate models that consider both direct and indirect effects. In particular, 3-D models are required to assess the complex feedbacks between aerosols, clouds, radiation, sea ice, and dynamic transport and to quantify climate forcing due to arctic haze (Girard *et al*., 2005).

Chapter 5
Effects on Terrestrial Ecosystems

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The first AMAP assessment of acidification (AMAP, 1998) addressed the processes involved in the acidification of arctic soils, and the direct effects of sulfur dioxide (SO_2) and nitrogen oxides (NO_X), as well as acidifying deposition, on the biotic components of terrestrial ecosystems. At that time, however, there was little empirical evidence to suggest that soil acidification was anything more than a local problem in very limited parts of, for example, the Kola Peninsula. The serious damage to flora and fauna reported in the area was mainly attributed to the direct toxic effects of SO_2, combined with the accumulation of toxic heavy metals in the arctic environment. It is extremely difficult to distinguish between the direct effects of SO_2, the indirect effects of the deposition of acidifying compounds (SO_2 and sulfate (SO_4)), and the direct toxic effects of heavy metals on terrestrial ecosystems in the areas around the smelters. In the Arctic, the cumulative effects of acidifying emissions and the deposition of toxic heavy metals can be disastrous for ecosystems which are already subject to extreme climatic conditions. In this, the latest acidification assessment, a large amount of new empirical data are presented and discussed.

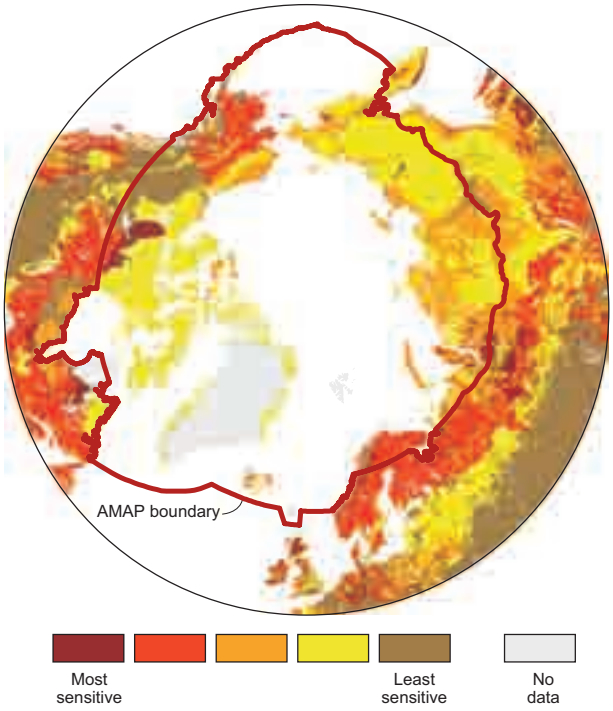


Figure 5.1. Sensitivity of arctic ecosystems to acid deposition (Kuylenstierna *et al.*, 2001).

5.1. Effects on soils

Kuylenstierna *et al.* (2001) have mapped the relative sensitivity of terrestrial ecosystems to acidic deposition at the global scale. An overview of the sensitivity of arctic ecosystems is provided in a circumpolar map extracted from the global sensitivity map. The sensitivity classes are based on base saturation and cation exchange capacity data applied to FAO (Food and Agriculture Organization) soil types. The soils most sensitive to acidic deposition have a low base saturation (<40%) and a low cation exchange capacity (<10 meq/100 g); the implication is that these soils have low rates of long-term mineral weathering and a limited base cation content and may therefore be subject to rapid changes in base saturation and pH. The most sensitive areas occur in Fennoscandia, in parts of Russia, and in parts of Canada and Alaska. Much of the Canadian Arctic seems not to be sensitive to acidic deposition (Figure 5.1).

AMAP has identified three regions in the Arctic that may be susceptible to acidification caused by the deposition of acidifying compounds (sulfur and nitrogen): the Kola Peninsula in northwestern Russia, the Taymir Peninsula in northern Russia, and the Chukotka region in eastern Siberia. The Kola Peninsula and the Taymir Peninsula both receive acidifying compounds from major local point sources; deposition in these areas has been, and is still, high. There is a large amount of information about the effects of acidic deposition on soils on the Kola Peninsula but much less about effects on soils in the Norilsk area. Owing to the very high SO_2 emissions from the Norilsk smelter complex, the acidifying effects are potentially many times higher than on the Kola Peninsula. Although the Chukotka region may receive large amounts of acidifying pollutants via long-range transport from industrial sources in China, India, and other parts of the Far-East, reliable deposition data and information about the soil acidification status of the region are extremely difficult to obtain. Other parts of the Arctic receive acidifying compounds via long-range transport from North America, Europe and the Far-East but, according to deposition measurements made in the Arctic, the levels are not likely to cause widespread soil acidification in background areas in the near future.

5.1.1. Acidity status of soils on the Kola Peninsula

The results of two regional studies – the Kola Ecogeochemistry Project (www.ngu.no/Kola) and the Barents Ecogeochemistry Project (www.gsf.fi/Barents) – have recently become available. These cover the distribution area of SO_2 emissions from the Cu-Ni smelters in Nikel, Zapolyarnyy and Monchegorsk on the Kola Peninsula, and almost pristine areas of northern Finland (Reimann *et al.*, 1998a; Salminen *et al.*, 2004). The survey carried out as part of the Kola Ecogeochemistry Project had a sampling density of approximately 1 site per 300 km^2 and generated information about soil acidity status and the most important natural and anthropogenic factors contributing to soil acidity in this region (Reimann *et al.*, 1998a, 2000a; Kashulina *et al.*, 1998a,b, 2003; Kashulina and Reimann,

2001; Kashulina, 2002). Figure 5.2 shows the area discussed here. The results of the Barents Ecogeochemistry Project, undertaken five years later and with a lower sampling density, confirmed the overall conclusions of the Kola Ecogeochemistry Project, and in addition, provided information about temporal trends in soil acidity in the region.

According to the Kola Ecogeochemistry Project, the pH(H_2O) of the organic (O) horizon of podzolic soils in 1995 ranged from 3.2 to 5.6. Although the region has received high levels of acidifying deposition for about 60 years, this wide range in pH cannot be related to the effects of two of the world's largest SO_2 emission sources, which are located on the Kola Peninsula, since an even greater pH range (3 to 6) has been reported as typical for the O-horizon of podzols in background areas (Targulian, 1971).

5.1.1.1. **Natural factors affecting soil acidity**

Marine input of base cations

There is a relatively strong decreasing trend (0.6 pH units) in the acidity of the O-horizon along a south–north gradient running through the background area (from the Arctic Circle to the coast of the Barents Sea) of northern Finland (Figure 5.3) (Reimann *et al.*, 2000b; Kashulina *et al.*, 2003). The sharpest increase in pH (0.4 pH units) occurs on moving north through the 100 km wide coastal zone. There is a 10-fold increase in sodium (Na) and a 2-fold increase in magnesium (Mg) in ground mosses and the O-horizon over the same coastal zone (Figure 5.3). This indicates that marine aerosols are probably the main factor explaining the pH increase near the coast. The figure also indicates that natural precipitation chemistry can have a significant effect on acidity parameters in the O-horizon.

Bioclimatic factors

The distribution of Na concentrations (Na is a major element in marine aerosols, but not an important plant nutrient) in mosses and the O-horizon (Figure 5.3) suggests, however, that the influence of marine aerosols is only responsible for the south–north pH gradient in the O-horizon within a distance of 200 km from the coast. The minor increase in pH between 200 and 500 km from the coast coincides with a slight increase in Mg and calcium (Ca) concentrations (Mg and Ca are both important plant nutrients) (Figure 5.3). The southward decrease in exchangeable Ca and especially Mg concentrations in the O-horizon is probably due to increasing uptake and utilization by plants. Thus, the change in bioclimatic factors (climate + vegetation) on moving from south to north may also affect the distribution of acidity in the O-horizon in the region (Kashulina *et al.*, 2003).

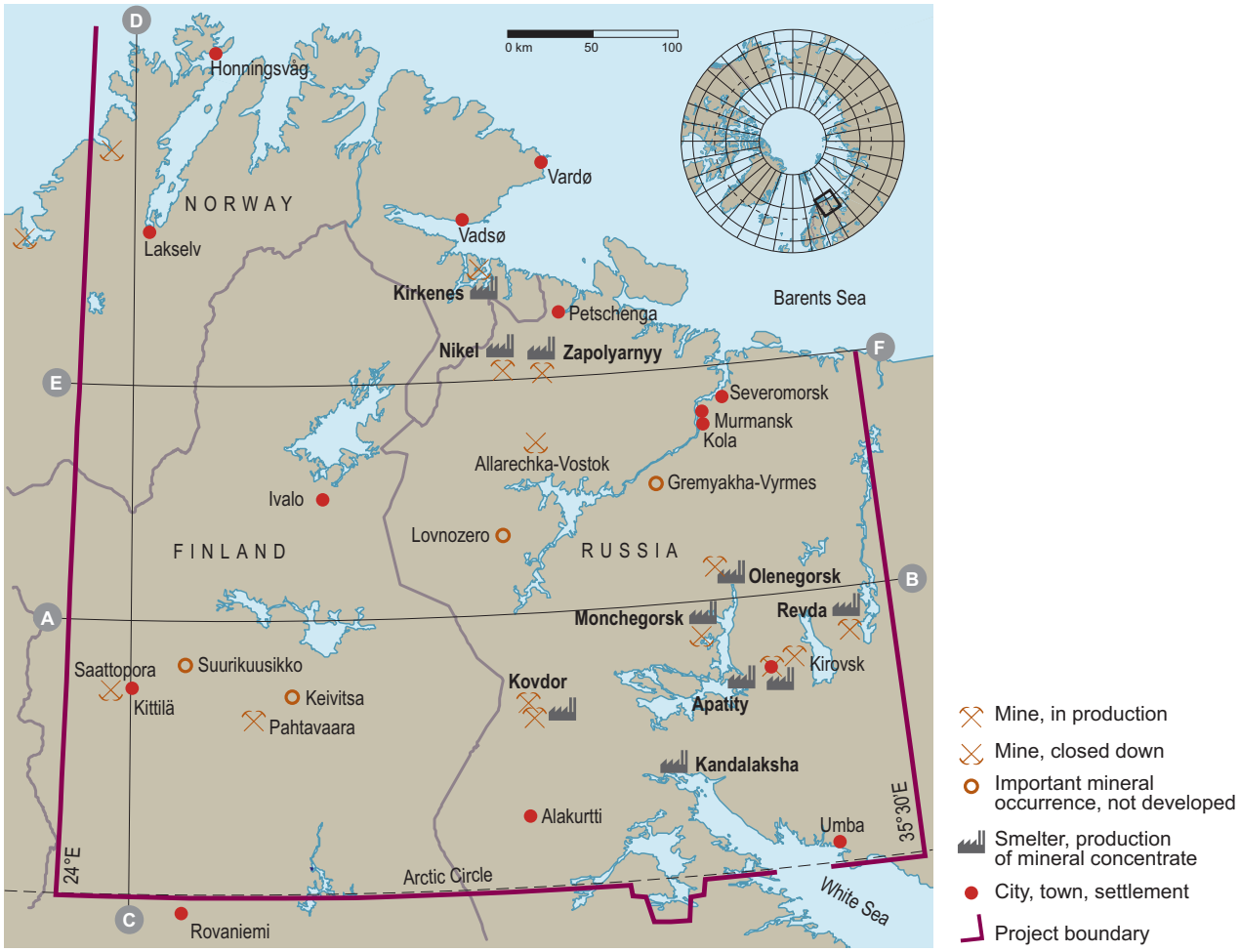


Figure 5.2. The Kola Ecoregion survey area and major industrial centers. The lines A–B, C–D, and E–F mark the transects shown in Figures 5.3, 5.4, 5.5, 5.7, and 5.9.

Site-specific and temporal variation

All the soil acidity parameters showed very high site-specific variation (Figure 5.3). For instance, pH at two adjacent sites in background areas differed by more than one pH unit. Acidity in the O-horizon can vary within the same site by more than one pH unit over the year (Levina, 1969). Thus, spatial and temporal variation in pH in the O-horizon may be greater than any natural trends at the regional level. Site-specific characteristics and/or processes can have a stronger deterministic effect on soil acidity status at an individual site than any other regional factors (Kashulina *et al*., 2003).

5.1.1.2. **Sulfur dioxide emissions and soil acidity**

The O-horizon of podzols is directly influenced by the input of ions and cations in precipitation. In the study area the organic material in the O-horizon has a lifetime of around 20 to 50 years. As a result, it 'integrates' the effects of pollutant deposition over a relatively long period, and the cumulative effect of acidifying pollutants can be considerable. According to Kashulina *et al*. (2003), precipitation acidity is affected by SO_2 emissions only within a radius of 30 km around the smelters. In other parts of the Kola Peninsula, the emissions of base cations (derived from the smelters, marine aerosols, and basal and alkaline dust from other industrial sources) supply enough base cations to maintain precipitation at a less acidic level than that recorded in northern Finland. Current emissions of nitrogen dioxide on the Kola Peninsula are low and have little or no effect on precipitation acidity; nitrate concentrations in precipitation are low and relatively uniform throughout the area (Tikkanen and Niemelä, 1995; Kashulina *et al*., 1998a). Thus, the area where the soil can be affected by acidified rain on the Kola Peninsula is limited to restricted zones around the smelters. The contribution played by the direct adsorption of SO_2 on the soil surface on the Kola Peninsula needs to be investigated, however.

Monchegorsk area

The west–east transect of pH in the O-horizon, running through the area occupied by the Monchegorsk smelter (Figure 5.4), indicates that emissions from industrial activities at Apatity (mining and processing of alkaline rocks mined at Kirovsk near Apatity), located 40 km south-east of Monchegorsk, have increased the pH by 0.3 units. These emissions also affect the pH of the surface soil close to the Monchegorsk smelter, and are still detectable at the Russian/Finnish border to the west. The atmospheric origin of this increase in pH is supported by the distribution of base cations in moss (Figure 5.4).

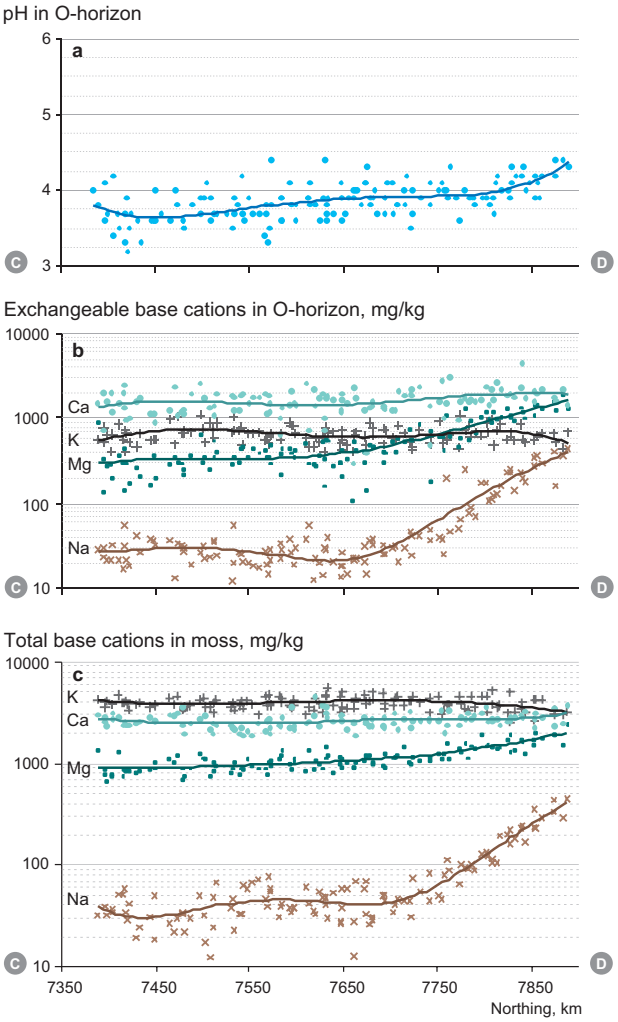


Figure 5.3. Transect south–north across the survey area near the western boundary in 1995 (C–D on Figure 5.2), showing: (a) pH in a water extract of the O-horizon; (b) ammonium acetate extractable (pH 4.5) base cation concentrations in the O-horizon; and (c) total base cation concentrations in mosses (Kola Ecogeochemistry Project, Kashulina *et al*., 2003).

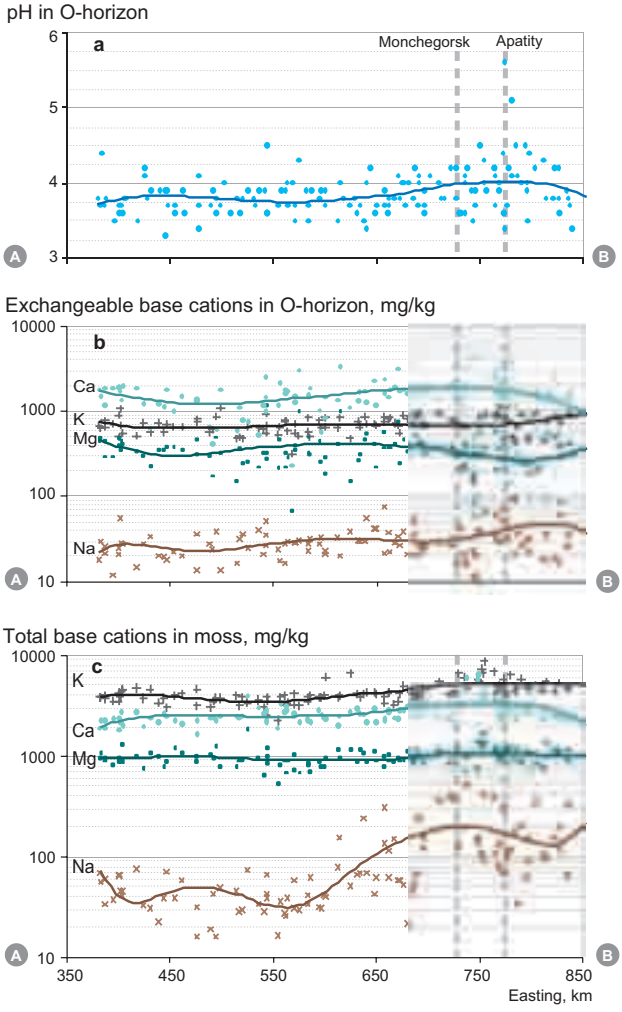


Figure 5.4. Transect west–east across the survey area and through the industrial zone centered on Monchegorsk and Apatity in 1995 (A–B on Figure 5.2), showing: (a) pH in a water extract of the O-horizon of podzol soils; (b) exchangeable base cation concentrations in the O-horizon of podzol soils; and (c) total base cation concentrations in mosses (Kola Ecogeochemistry Project, Kashulina *et al*., 2003).

The distribution of exchangeable Ca (the dominant base cation) in the O-horizon along the transect follows the same pattern as for pH (Figure 5.4). However, there is no corresponding trend for potassium (K) or Mg. This could be partly due to the increased leaching into stream water, observed within 30 km of Monchegorsk (Kashulina *et al*., 2003). However, the increase in total Mg in mosses (Figure 5.4) and the O-horizon near Monchegorsk (Reimann *et al*., 1998a) is in agreement with the increased deposition of Mg in the area. The decrease in the exchangeable Mg concentration may be associated with low solubility of Mg in anthropogenic particulate material deposited on the O-horizon.

A low pH (although within the natural range of pH variation) and extremely low base cation concentrations (Figure 5.4) are characteristic of soils at a number of sampling sites in the immediate vicinity of the Monchegorsk smelter where the vegetation cover has been completely destroyed. Low pH values were also reported in case or gradient studies within a distance of 5 km from the smelters on the Kola Peninsula (Chertov *et al*., 1993; Koptsik and Muchina, 1995; Kashulina *et al*., 2003). No changes in soil acidity were found in a gradient study within a distance of 10 to 80 km from Monchegorsk (Tikkanen and Niemelä, 1995).

Nikel/Zapolyarnyy area

Further north, the variation in pH in the O-horizon along an east–west transect through Nikel and Zapolyarnyy is less than 0.2 pH units (Figure 5.5). There is a very slight (<0.1 pH units) decreasing trend near Nikel, due to a small number of low pH values near Nikel. Correspondingly, the slightly increasing trend near Zapolyarnyy is due to a number of relatively high pH values. The eastern end of the transect has slightly higher pH values than the western end. The distribution of marine-derived cations (Na and Mg) in mosses and the O-horizon indicates an increasing maritime influence towards the east, starting near Nikel (Figure 5.5). The western section of the transect is more inland, while the eastern section ends at the Barents Sea coast. In addition to marine-derived deposition, the Zapolyarnyy area receives alkaline dust from opencast mining (Reimann *et al*., 1997).

Generally, it appears that the sources of base cations are sufficient to prevent acidification of the O-horizon, and to maintain relatively constant pH and base cation concentrations in the vicinity of Nikel and Zapolyarnyy. As is the case for Monchegorsk, only a small number of low values were observed in the immediate vicinity of Nikel/Zapolyarnyy, especially at sites where the vegetation cover has been severely damaged.

5.1.1.3. **The role of overburden and bedrock chemistry**

The simultaneous emission of fly ash by the smelters and associated power plants, and the alkaline nature of the overburden and bedrock in some areas near the emission sources, are frequently used to explain the lack of widespread soil acidification on the Kola Peninsula, despite the very high SO_2 emissions (Koptsik and Muchina, 1995; Moiseenko, 1997; Tikkanen and Niemelä, 1995). This raises the question of whether the relatively high base cation content of the parent material in some areas near both emission sources (Monchegorsk and Nikel/Zapolyarnyy) can influence the chemistry of the O-horizon and counteract the acidifying effect of the emissions. Relatively poor correlation has been found between the element concentrations in the parent material (C-horizon) and the O-horizon immediately around the Nikel/Zapolyarnyy smelters (Reimann *et al*., 1998b). Base cation concentrations in the O-horizon at all the sampling points show a much closer relationship with the corresponding concentrations in mosses (the chemistry of which is primarily determined by atmospheric inputs) (Figures 5.3 to 5.6), than with those in the C-horizon (Kashulina *et al*., 2003). Thus, the distribution of acidity in the O-horizon in the region is mainly determined by the atmospheric input, and the geological influence is hardly detectable in the relatively high atmospheric base cation deposition gradient on the Kola Peninsula. Fly ash from the smelter complexes is not the most important source of base

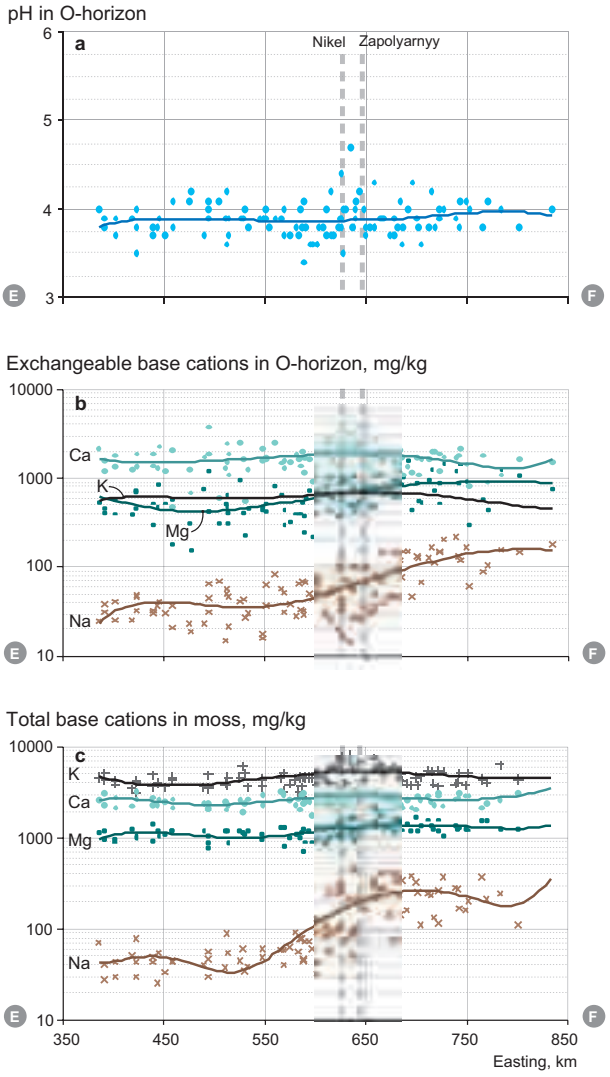


Figure 5.5. Transect west–east across the survey area and through the industrial zone centered on Nikel and Zapolyarnyy in 1995 (E–F on Figure 5.2), showing (a) pH in a water extract of the O-horizon of podzol soils; (b) exchangeable base cation concentrations in the O-horizon of podzol soils; and (c) total base cation concentrations in moss (Kola Ecogeochemistry project, Kashulina *et al*., 2003).

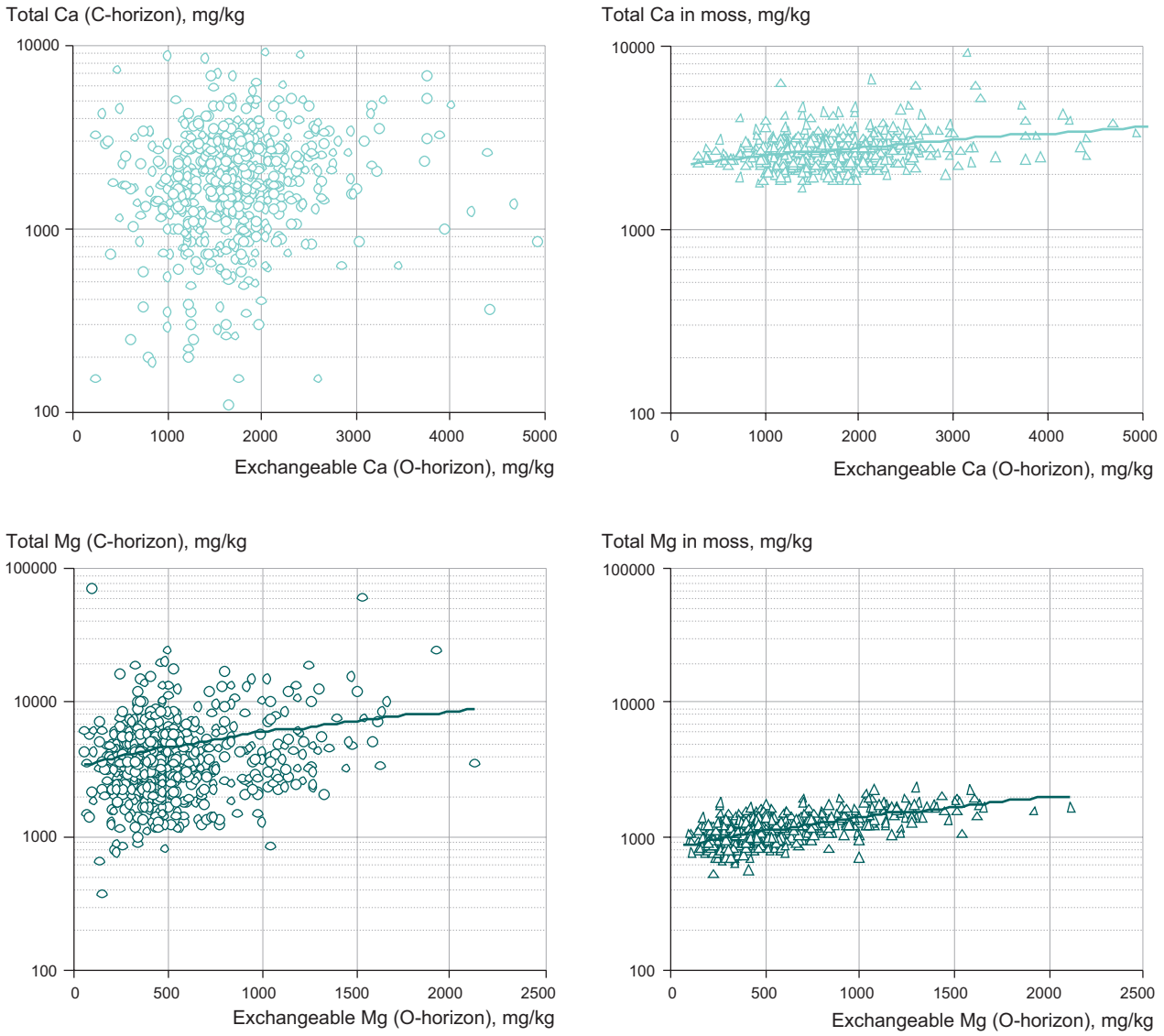


Figure 5.6. Ammonium acetate extractable Ca and Mg in the O-horizon of soils (x-axis) vs. their total concentrations (aqua regia extractable) in the C-horizon of soils and in moss in 1995 (Kola Ecogeochemistry project, Kashulina *et al*., 2003).

cations in deposition on the Kola Peninsula (Kashulina *et al*., 2003). Alkaline dust from the apatite fertilizer plant, marine aerosols, and alkaline dust from open-cast mining appear to be the major sources of base cation deposition on forest soil in the western part of the Kola Peninsula. The current rate of base cation emissions is sufficient to maintain soil acidity parameters at levels that are even less acidic than, for example, in northern Finland. The only areas with increased soil acidity and exchangeable base cation depletion are in the severely damaged ecosystems immediately around the smelters.

Exchangeable aluminum

The major factor governing the distribution of exchangeable aluminum (Al) in the O-horizon on the Kola Peninsula appears to be the bedrock geology (Kashulina *et al*., 2003). Aluminum concentrations in the C-horizon along the transect running through the major SO_2 emission source areas (Figure 5.7) are more similar to the concentrations in the O-horizon than to the moss concentrations (Figure 5.4). The Al concentrations in mosses along the transect do, however, suggest that there is elevated deposition of Al into the O-horizon near Nikel/Zapolyarnyy (alkaline rock dust from the opencast mine) and near Apatity (apatity-nepheline dust). The Al concentrations in precipitation increase to such an extent immediately around the smelters (Reimann *et al*., 1997; de Caritat *et al*., 1998), that Al is a major cation (in equivalent terms) in rain and snow (Kashulina *et al*., 2003). Thus, the relatively high exchangeable Al concentrations in the soil and percolation water reported near Monchegorsk (Motova and Nikonov, 1993; Lukina and Nikonov, 1996) are more likely to be associated with basal lithology and increased deposition than to the impacts of SO_2 emissions.

The aluminum:base cation (Al:BC) ratio in the O-horizon (Figure 5.7) shows extremely high spatial variation. The highest values occur around Apatity (apatite-nepheline, syenite) and in soils overlying the meta-sedimentary rocks of northern Norway. The Al:BC ratio in the O-horizon shows a good correlation with the total Al concentration in the C-horizon ($r = 0.73$), and so the geology is therefore the main factor determining the Al:BC ratio in the O-horizon

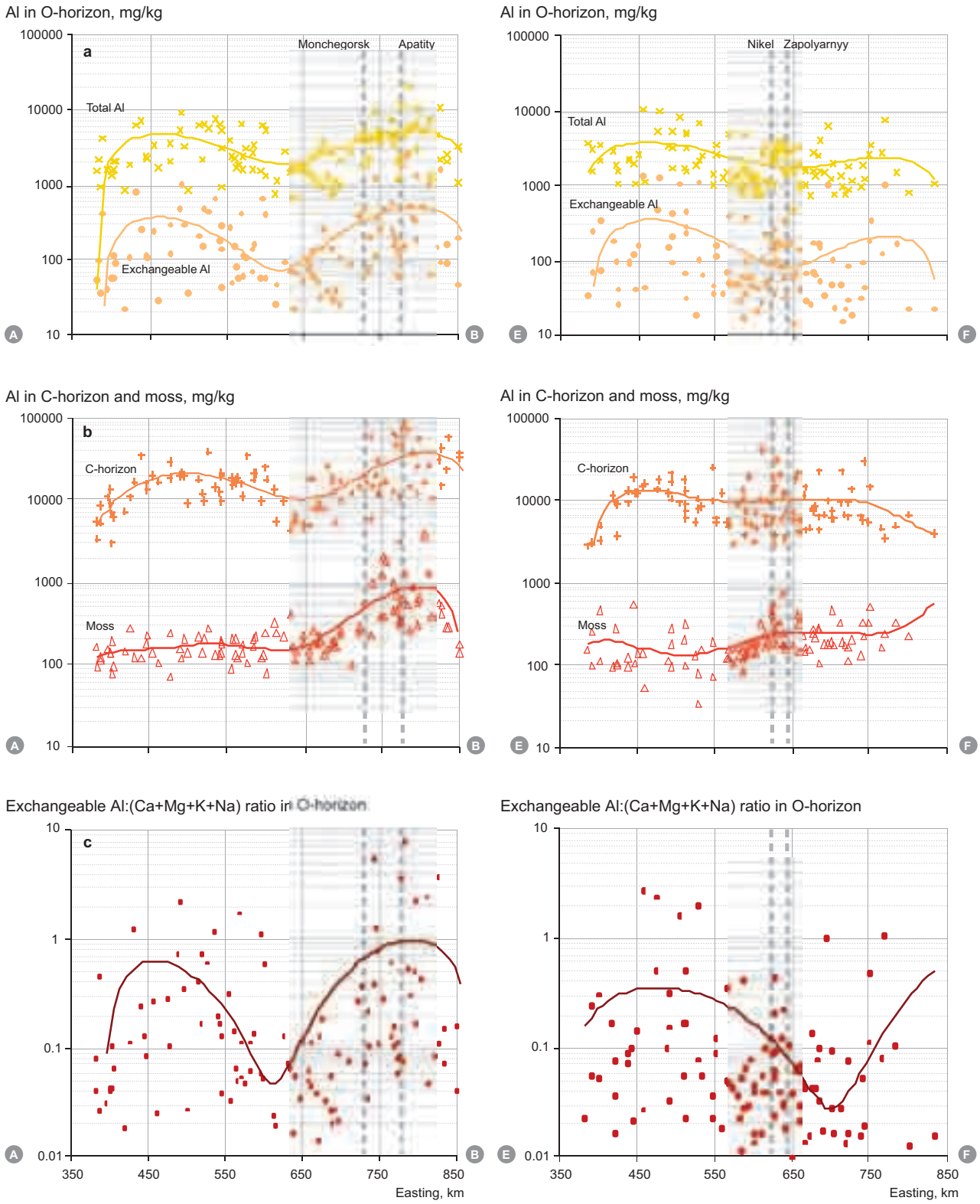


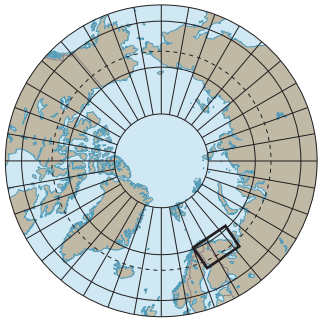
Figure 5.7. A comparison of the two west–east transects across the survey area in 1995 (A–B and E–F on Figure 5.2), showing changes from east to west in (a) total Al (nitric acid) and exchangeable Al concentrations in the O-horizon; (b) total Al concentrations in moss and Al (aqua regia extracts) concentrations in the C-horizon; and (c) the exchangeable Al:(Ca+Mg+K+Na) ratio in the O-horizon (Kola Ecogeochemistry Project, Kashulina *et al.*, 2003).

(Kashulina *et al.*, 2003). Emissions affect the Al:BC ratio in the soil only within the immediate vicinity of the smelters. Furthermore, this appears to be an indirect effect of pollution via damage to the ecosystem. Elevated Al:BC ratios also occur in the completely destroyed ecosystems (Figure 5.8).

5.1.1.4. **Connections between soil condition and ecosystem quality**

Soil acidity

The distribution of the major acidity parameters in the various classes of ecosystem damage (Kashulina *et al.*, 2003)



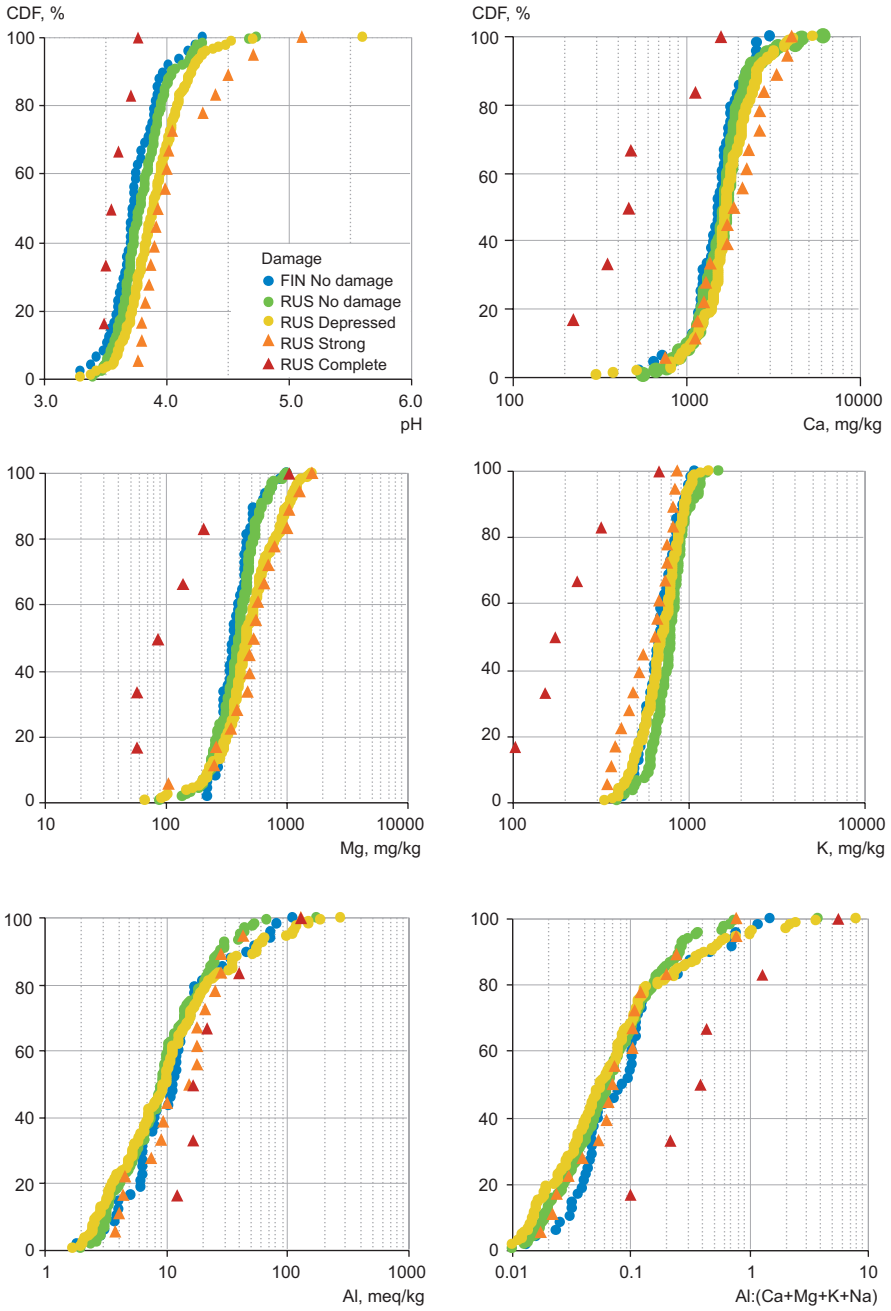


Figure 5.8. Cumulative distribution functions (CDF, %) of pH (water extraction), exchangeable Ca, Mg, K, and Al concentrations and the exchangeable Al:(Ca+Mg+K+Na) ratio in the O-horizon in the 'no visual damage' zone in Finnish Lapland and for various classes of ecosystem damage in the western part of the Kola Peninsula in 1995 (Kola Ecogeochemistry Project, Kashulina *et al.*, 2003).

shows that the onset of ecosystem damage (and even of serious ecosystem damage) on the Kola Peninsula occurs where there is a more favorable (higher pH and base cation concentrations) acidity status in the O-horizon compared with background areas in northern Finland (Figure 5.8). In contrast, severe ecosystem damage appears to be a prerequisite for soil acidification to appear on the Kola Peninsula.

Plant nutrients

In addition to base cations (Ca, Mg, K) and other important nutrients (e.g., nitrogen and phosphorus), depletion of some other nutrients (e.g., manganese (Mn) and zinc (Zn)) is regarded as one of the detrimental consequences of acidifying deposition on ecosystems (Galloway, 1995). A sharp decrease in some nutrients (Mn, Zn, Mg) has been reported in severely damaged ecosystems around the smelters on the Kola Peninsula (Lobersli and Venn, 1995; Tikkanen and Niemelä, 1995). A deterioration in the nutrient status of the soil on the Kola Peninsula has occurred only in the completely destroyed ecosystems around the smelters. There is also a decrease in phosphorus concentrations in the seriously damaged ecosystems within a radius of 30 km around the smelters. Manganese concentrations below the deficiency level only occurred at sites with serious or complete destruction of the ecosystems (Kashulina, 2002). Furthermore, the depletion of major nutrients is not a specific reaction of the ecosystem to acidifying deposition.

Thus, the indirect effect of acidifying pollutants on the condition of ecosystems via soil acidification and nutrient depletion does not appear to be the main factor affecting forest ecosystem condition on the Kola Peninsula. The direct effects of SO_2 on the vegetation (Aamlid *et al.*, 1995; Tikkanen and Niemelä, 1995; Kashulina *et al.*, 2003) is a much more likely explanation for the widespread damage to ecosystems on the Kola Peninsula. The significant

increases in the concentrations of some other pollutants (e.g., nickel, cobalt, cadmium, silver, arsenic, lead, copper and others) of up to a few orders of magnitude in the soil near the major emission sources (Räisänen *et al*., 1997; Reimann *et al*., 1998a; Äyräs and Kashulina, 2000) is obviously contributing to the widespread ecosystem damage.

5.1.1.5. **Temporal trends in soil acidity**

A comparison of soil acidity parameters near the Monchegorsk smelter in 1995 (Reimann *et al*., 1997; Kashulina *et al*., 2003) and 2000-2001 (Salminen *et al*., 2004) showed no clear changes in the exchangeable Ca concentration in the O-horizon (Figure 5.9). The values obtained in the sparser sampling network in 2000 are within the range of spatial variability reported for 1995.

5.1.2. **Acidification and the acidity status of soils in the Norilsk area**

The Norilsk mining area is one of the largest point sources of sulfur and certain heavy metals in the northern Arctic (AMAP, 1998). Annual sulfur emissions were around 1 million tonnes between 1985 and 2000 (Ekimov *et al*., 2001), while annual nickel and copper emissions were up to 1300 and 2800 tonnes, respectively. These emissions are more than ten times higher than emissions reported for the Monchegorsk smelter on the Kola Peninsula (MRCENR, 1995; Ekimov *et al*., 2001). Since 2000, there have been no reported changes in the volume of metal production or any investments to decrease emissions. SO_2 emissions for Norilsk were 2118 kt for 1992 and 1847 kt for 2003.

The effects of emissions on the soil are partly determined by climate. Owing to the presence of permafrost in the region, chemical processes can only occur in the uppermost part of the soil, and because of the long winter (9 to 10 months) the period favorable for chemical changes is short. However, the uppermost part of the soil profile is strongly affected by the emissions of sulfur and heavy metals.

Basalts with a basic chemical composition characterize the bedrock in the Norilsk area and in the neighboring Putorana Mountains. According to Naldrett *et al*. (1992), the sulfide ore deposits in the Norilsk area are hosted by a large flood basalt formation that erupted in the Devonian and Carboniferous sedimentary rocks. The age of the basalt is 251 million years. It consists of picritic, basaltic, and tholeitic lavas. Today the basalts form the mountains (the Putorana Mountains) and sedimentary rocks (which are not metamorphosed but eroded deeper) fill the valleys (e.g., in the Norilsk area). These sedimentary rocks include Devonian calcareous and dolomitic marls, and sulfate-rich evaporates and lower Carboniferous shallow water limestones. Westward from Norilsk the bedrock changes and mainly includes intermediate and more acidic types of rock.

In 2001, water and soil samples were collected from ten sites in the Norilsk industrial area and in the uncontaminated area to the east of the industrial zone. Stream water samples were collected from each site and complete soil profiles were collected from two sites – one in the industrial area and the other in the clean background area. Organic layer samples were also taken from two sites. The sampling and analytical procedures are as reported by Gregorauskiene *et al*. (2000) and Salminen and Gregorauskiene (2002).

The minerogenic soils that have developed on the weathering crust of basalt (Lake Lama) have pH values ranging from 5.1 to 6.2, and the soils that have developed on calcareous sedimentary rocks (Norilsk) from 7.1 to 8.6. These values are considerably higher than the respective values reported for Monchegorsk (Table 5.1). The buffering capacity, as indicated by Ca and Mg concentrations, is much higher in Norilsk than in Monchegorsk. The soils in the Norilsk area cannot be considered sensitive to acidification; the pH of the organic layer in the Norilsk industrial area was as high as 6.4. However, the Cu and Ni values of the organic layer are at the same level as in Monchegorsk (1370-2820 mg/kg), due to the smelter emissions. The corresponding concentrations in the Lake Lama area are much lower (72-82 mg/kg). However, even the concentrations in the Lake Lama area are higher than the median values (5.9-7.9 mg/kg) reported for large areas of the Kola Peninsula (Salminen *et al*., 2004).

5.1.3. **Effects on soil micro-organisms**

Soil acidification and the deposition of sulfur and heavy metals can influence soil micro-organisms via several routes. First, a decline in pH and the accumulation of pollutants in the soil may directly reduce microbial growth and activity (Bååth, 1989). Second, pollutants may indirectly harm the symbiotic and rhizosphere micro-organisms by decreasing the amount of photosynthesizing foliage of their hosts. This, in turn, reduces carbon flow to the roots and mycorrhizal fungi (which are dependent on carbon received from the host) and to rhizosphere microbes (which are negatively affected both by a reduction in the availability of suitable environment due to reduced root biomass and by a decline in the amount of carbohydrate exudates produced by the roots). Direct and indirect impacts of acidification

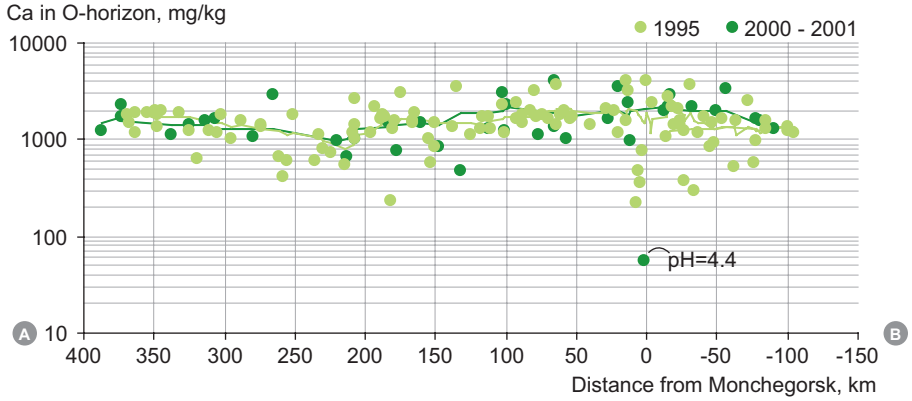


Figure 5.9. Transect west–east across the survey area (A–B on Figure 5.2) showing exchangeable Ca in the O-horizon of podzol soils in 1995 (Kola Ecogeochemistry Project) and from 2000 to 2001 (Barents Ecogeochemistry Project).

Table 5.1. Element concentrations in soil horizons in the Monchegorsk (Kashulina and Gregorauskiene, 2000), Norilsk, and Lake Lama areas.

Sites/ horizon	Lowest depth (cm)	pH	LOI (%)	C (%)	Ca (mg/kg)		Mg (mg/kg)		Cu (mg/kg)	Ni (mg/kg)	S (mg/kg)
					Total	AR	Total	AR	AR	AR	Total
Monchegorsk											
Oer	0	3.90	20.50	11.60		792		532	1370	2030	446
OEer	3	4.39	5.79	3.11	39800	614	37300	443	366	541	122
Bs	20				42400						
BC1	40	4.78	3.72	0.92	49900	3310	48200	4850	52.0	72.9	175
2BC2	82	5.42	2.10	0.47	54900	3110	43100	4540	62.2	60.3	95.4
Norilsk											
O	5-6	6.40	42.90	22.90		23600	7340		2180	2820	3580
B1	13	7.10	7.96	1.61	43100	12800	24200	8390	64.5	45.0	274
B2	25	7.60	9.36	2.32	44200	13100	27400	8290	50.3	37.0	173
BC	49	8.60	5.35	0.81	62900	25600	29200	9410	50.6	31.0	122
BCk2	70	8.60	5.96	1.07	73900	32900	30900	11100	53.5	32.5	<100
Lake Lama											
O	4	4.00	65.20	32.30		3900	2980		81.9	72.4	1080
E	3	4.30	25.10	12.80		4140		3560	30.9	18.4	
B	7	5.10	20.30	5.31	31600	7980	22800	8650	47.9	41.6	198
BC	19	5.40	13.70	2.91	37500	9750	23000	10100	58.1	42.3	198
BC2	41	6.00	7.68	1.10	43800	13400	25800	10900	66.5	39.7	124
C	70	6.20	6.00	0.70	50700	15800	29000	10800	56.1	36.6	157

LOI: loss in weight on ignition; AR: aqua regia extractable concentration.

and heavy metals include species-specific effects, leading to shifts in fungal species composition. Third, pollutants often reduce the amount of litter produced by vegetation, and also change its composition. In addition, the direct effects of acidifying pollutants on soil invertebrates, resulting for example in the disappearance of earthworms in polluted areas (section 5.3.2), can affect the mechanical degradation of litter. Both the amount and composition of the litter, as well as changes in the degradation processes of soil organic matter, may have several impacts on soil decomposer activity and diversity.

In the Arctic, relatively few studies have been carried out on the impacts of acidifying pollutants on soil micro-organisms. The adverse effects on soil microbial biomass and activity seem to be concentrated in relatively restricted areas around the nickel-copper smelters where there are high levels of heavy metal deposition. For instance, Ohtonen and Väre (1996) reported no detectable changes in soil microbial activity and biomass parameters at distances of no more than 14 km from the smelter complex at Nikel. On the other hand, soil microbial activity and biomass were reported to be extremely reduced near Monchegorsk. For example, basal respiration of micro-organisms at sites 8 and 10 km from the Monchegorsk smelter was 5 and 8 µg carbon dioxide (CO_2) per gramme of organic matter (OM) per hour, respectively, while at sites 36 km or more the basal respiration was 16 to 40 µg CO_2/g OM/hr. Microbial biomass was 2.2 and 2.4 C/g OM at nearby sites, and 2.4-5.6 C/g OM at more distant sites (Ohtonen and Väre, 1996). Also, the diversity of microfungi (Lebedeva, 1993; Evdokimova, 2000), algae, and prokaryotes (non-spore forming gram negative bacteria, cyanobacteria and streptomycetes) (Evdokimova, 2000) in forest soils decreased moving towards the pollution source. In another study (Nikonov *et al*., 2001), populations of prokaryotes in the soils of Norway spruce stands were found to increase, and populations of eukaryotes to decrease, moving towards Monchegorsk. Deterioration of the forest soil, leading to losses in soil organic matter and nutrients, was assumed to be connected to the adverse effects of heavy metals on soil fungal populations in drier Scots pine forests (Polyanskaya *et al*., 2001). Changes in the composition of soil microbial communities have also been found in taiga forests around the Kostamuksha iron pellet plant (Zaguralskaya and Ziabchenko, 1994) and in subtundra forests of the Taymir Peninsula affected by the Norilsk nickel-copper smelter (Kirtsideli *et al*., 1995). Intriguingly, rhizosphere of severely damaged larches near Norilsk contained 10 to 100 times more micro-organisms than healthy larches in unpolluted regions; this phenomenon presumably resulted from the development of saprotrophic microbiota that benefit from root decline (Raguotis, 1989).

Fluoride deposition originating from the Kandalaksha aluminum smelter caused slight alkalinization of the soil and altered the community composition of soil fungi, although no effects on the biomass of soil bacteria and fungi were found (Evdokimova, 2001). However, a more detailed investigation revealed a significant (by a factor of eight) decline in fungal biomass near Kandalaksha (Evdokimova *et al*., 2004).

Acidification may not necessarily affect the basidiomycete and ascomycete ectomycorrhizal fungal symbionts of forest trees, as they are well adapted to naturally acidic conditions in forest soil (Smith and Read, 1997). However, a decrease in ectomycorrhizal colonization in the roots of forest trees has been found in response to soil acidification (Danielson and Visser, 1989; Holopainen *et al*., 1996; Brunner, 2001). Emissions from the Monchegorsk smelter have been reported to negatively affect the diversity of ectomycorrhizal fungi (Isaeva, 2004).

Studies on the effects of acidifying pollution on arbuscular mycorrhizal (AM) colonization in herbs and grasses in the Arctic are even fewer than those dealing with ectomycorrhizal symbionts of trees. There appears to have been only one survey (Ruotsalainen *et al*., 2006) on root colonisation in *Deschampsia flexuosa*, a common perennial grass that is tolerant of both acidification and heavy metals. There was a decrease in hyphal colonization of both AM and DSE (dark septate) fungi with increasing pollution levels. In contrast, DSE microsclerotia were more abundant in highly polluted sites, possibly indicating the strategy of these fungi to survive and disperse from unfavorable environments. Severe damage to field layer vegetation creates a patchy environment, such as the industrial barrens around

Monchegorsk, and under these conditions AM fungi may have difficulties dispersing from one plant host to another. This may have further impacts on fungal communities in the soil.

The cover of *Cladina* lichens, which is assumed to be especially effective at filtering out heavy metals, has deteriorated and even disappeared in industrial barrens and declining forests (Tømmervik *et al*., 1995), and this may accelerate the adverse impacts of acidifying pollutants on the activity of soil micro-organisms. The structure of the organic layer has been damaged severely in these areas (Rigina and Kozlov, 1999). This is partly due to the loss of living fungal hyphae which, together with the fine roots, normally play an important role in maintaining the structure of this layer.

Thus, changes in ground and field layer vegetation due to acidifying pollution in the Arctic are linked to soil deterioration and changes in soil microbiology. On the basis of sparse information on soil micro-organisms in the Arctic, the adverse effects of acidifying pollutants seem to be concentrated in restricted areas around the nickel-copper smelters and to be more strongly linked to the excessive deposition of heavy metals rather than to acidification. The diversity of soil micro-organisms may be affected to a greater extent or at lower levels of pollution, than the biomass and activity of the soil microbial communities.

5.2. Effects on vegetation in the European Arctic

The damage zones around Nikel and Monchegorsk on the Kola Peninsula reported in the previous AMAP assessment (AMAP, 1998) ranged from forest death to a zone with changes in the microscopic structure of epiphytic lichens only. The corresponding range in annual average atmospheric SO_2 concentrations was from >40 to 2-4 µg/m^3 (Tikkanen and Niemelä, 1995; Tømmervik *et al*., 1995). Nikel, although having a much smaller smelter than Monchegorsk, has had (for technical reasons) considerably higher SO_2 emissions (but lower metal emissions) than Monchegorsk. It is also further north and so, having a harsher climate, the vegetation is dominated by subarctic birch forest rather than the boreal forests around Monchegorsk (Kashulina *et al*., 1997). Satellite data showed that the total area affected by air pollution (average SO_2 concentrations >10 µg/m^3) around Nikel increased from 400 km^2 in 1973 to more than 3900 km^2 in 1988, and remained at this level during the early 1990s (Høgda *et al*., 1995; Tømmervik *et al*., 1995; see also Aamlid *et al*., 1995; Gytarsky *et al*., 1997). Furthermore, episodes of high SO_2 emissions had caused changes in the lichen and dwarf shrub communities over an additional 1100 km^2 in 1988, the area affected by severe air pollution near Nikel thus increasing to more than 5000 km^2.

The size of the forest-death area was over 400 km^2 in the Nikel-Zapolyarnyy and Varanger regions; the area near the border between Russia and Norway studied in 1988 (Tømmervik *et al*., 1995). At Monchegorsk, the forest-death area covered between 400 and 500 km^2 and extended more than 10 km to the south and more than 15 km to the north of the smelter complex (Mikkola, 1996). The average modeled SO_2 concentrations exceeded 40 µg/m^3 in this area (Tuovinen *et al*., 1993). Marked defoliation of conifers and an absence of epiphytic lichens occurred in the area with annual average modeled SO_2 concentrations of 15 to 40 µg/m^3. External damage on the needles of Scots pine (e.g., tip necrosis) and changes in species composition of the lichen communities occurred in areas with annual average modeled SO_2 concentrations of 8 to 15 µg/m^3, and the damage area extended to the eastern part of Inari in Finland. The total area covered by visible damage zones surrounding Monchegorsk, Nikel, and Zapolyarnyy was estimated to be around 39 000 km^2. Around Monchegorsk, chlorosis of pine needle stomata occurred in areas with annual average modeled SO_2 concentrations of 4 to 8 µg/m^3, and there were changes in the microscopic structure of pine needles and epiphytic lichens where the annual average modeled SO_2 concentrations were 2 to 4 µg/m^3 (Tikkanen and Niemelä, 1995).

In the mid-1990s, the proportion of damaged ecosystems in the European Arctic increased steadily from the northern boreal forest zone to the tundra. Furthermore, the same stages of ecosystem disturbance (severe, damaged, depressed, no damage) coincided with lower concentrations of the major pollutants in humus in tundra and in subarctic birch forest ecosystems than in forested areas (Kashulina *et al*., 1997). On the other hand, a vegetation study from South-Varanger in eastern Finnmark, Norway (Aarrestad and Aamlid, 1999) showed that the 'pollution variables' Cu and Ni in humus, Ni in *Cladina*, and the modeled atmospheric SO_2 concentration, together only explained around 9% of the species variation after taking into account variation due to natural environmental variables. The comparisons between sulfur concentrations in moss and Ni concentrations in humus at various levels of ecosystem damage demonstrate that the state of the ecosystems in the mid-1990s in Norway and Finland had, with few exceptions, no correlation with the deposition of pollutants from the Russian smelters. In Russia, however, pollution strongly influenced ecosystem degradation. There, even the 'no damage' class had a higher median value for Ni and sulfur than for all damage classes in Finland or Norway (Kashulina *et al*., 1997).

As there have been very few studies on acidification effects on vegetation in the Arctic since the last AMAP assessment, this section reviews the few new studies together with the results of studies based on data collected in the early 1990s, but mainly published in the late 1990s or early 2000s. The studies include data on direct and indirect effects of air pollutants, especially SO_2, and both critical levels and critical loads are addressed.

5.2.1. Lichen-dominated and mountain birch (tundra) ecosystems

The main vegetation types in the northernmost and high altitude areas of northern Norway and the Kola Peninsula consist of lichen and dwarf shrub communities. Changes in the area with reindeer lichen (*Cladina* spp.)-dominated vegetation (heaths and forests with dwarf birch and mountain birch, *Betula nana* and *B. pubescens* ssp. *czerepanovii*) around the Cu-Ni smelters in Nikel and Zapolyarnyy correlated with changes in emissions of SO_2 between the 1970s and late 1990s (Høgda *et al*., 1995; Tømmervik *et al*., 1995, 1998, 2003). Thus, there was a reduction in the areas of lichen-dominated mountain heaths and forests from 37% in 1973 to 10% in 1992, followed by a slight increase to 12% in 1999 in the border areas of Norway and Russia (Tømmervik *et al*., 2003). Reindeer lichens had completely disap-

peared from around two Ni-Cu smelter complexes on the Kola Peninsula by the 1990s (Kalabin, 1991; Tømmervik *et al*., 1995), their growth rates reaching normal levels at a distance of 50 to 60 km from the smelters (Helle and Kojola, 1992). In addition to *Cladina stellaris*, the arctic lichen *Cladonia rangiferina* is also sensitive to SO_2 and/or metals (Koptsik *et al*., 2003). Kapitsa and Golubeva (1997) ranked the lichens *Cladina mitis* and *Cladonia cornuta* among the most sensitive species to anthropogenic pressure, while the lichen *Cetraria nivalis* was ranked as less sensitive, and the low bushes *Phyllodoce caerulea* and *Salix glauca* were among the most tolerant species. Gaseous uptake measurements on lichens are limited, but according to Winner *et al*. (1988, cited in Nash and Gries, 1995), SO_2 uptake by *C. rangiferina* is at least an order of magnitude greater than in a typical vascular plant. The lichen-dominated vegetation types were, however, not just changed into barrens or partly damaged vegetation entities, but were also changed into heath (and woodland) vegetation with sparse lichen cover and were dominated by dwarf shrubs such as bilberry (*Vaccinium myrtillus*) in the Nikel–Zapolyarnyy and South-Varanger areas in the early 1990s (Tømmervik *et al*., 1995, 1998).

The gradual change of lichen-dominated vegetation between 1973 and 1999 into barrens or sparsely vegetated areas or into other vegetation formations: partly damaged heather-dominated vegetation, bilberry forests/heaths, and meadow forests (on better soils in lowland areas) occurred mainly between 5 and 30–40 km from the smelters at Nikel and Zapolyarnyy, depending on prevailing wind directions during the growing season. For example, the previous sparsely vegetated coastal mountains have become more barren due to large emissions of SO_2 and their location within the path of the prevailing winds. On the other hand, the slight increase from around 10 to 12% in the lichen-dominated communities (*Betula nana*-lichen heath and *Empetrum*-lichen types) during the 1990s indicates an improvement in air quality and environmental conditions for these vegetation formations (Tømmervik *et al*., 2003); for example, a decrease from 20 to 10 µg/m^3 in the summer SO_2 concentrations at Svanvik in the Norwegian–Russian border area (SFT, 2002). The significant positive relationship between lichen (*Cladina* sp.)-dominated forests (*Empetrum*-lichen type with birches; r=0.84, p=0.018) and the significant negative relationship between changes in the area of industrial barrens (r=-0.95, p=0.001), and the changes in SO_2 emissions between 1973 and 1999 may be explained by the response times of the different land cover classes to SO_2 emissions. There is always a lag in the vegetation response to changes in SO_2 emissions and in this case the area of lichen-dominated vegetation types decreased and the barrens increased at the same time as emissions decreased (but not reaching 1973 levels before the mid-1990s) (Tømmervik *et al*., 2003).

Average SO_2 concentrations for the lichen-dominated vegetation (*Betula nana*-lichen heath and *Empetrum*-lichen types) were 3.5 and 9.2 µg/m^3, respectively, and were 13.6 µg/m^3 for heather woodland/partly damaged heather (*Empetrum-Vaccinium* type) with a sparse/reduced lichen cover (Tømmervik *et al*. 1995, 1998). There were associations between the damaged land cover, the industrial barren land, and the SO_2 concentration in air at ground level and Ni and sulfur concentrations in reindeer lichens. Average sulfur concentrations in reindeer lichens (*C. stellaris* and *C. mitis*) from *Empetrum*-lichen type forests and *Empetrum-Vaccinium* type heather woodland/partly damaged heather were 489 and 765 mg/kg, respectively, in the border areas between Norway (Southern-Varanger) and Russia (Nikel–Zapolyarnyy) in the late 1980s to early 1990s. In the bilberry forests/heaths (*V. myrtillus-D. flexuosa* and *Cornus suecica* types), the average SO_2 concentration was 14.9 µg/m^3 and the lichen sulfur concentration was 644 mg/kg (Tømmervik *et al*., 1998). The vegetation damage observed and the slightly elevated sulfur concentrations in lichen thalli may be attributed to both high short-term SO_2 concentrations as well as long-term exposure to elevated sulfur deposition based on the proposed critical level of 10 µg SO_2 /m^3 (annual mean) to be adopted for cyanobacterial lichens (UNECE, 1993, 2004a).

One of the nine catchments where Reimann *et al*. (2001b,c,d, 2003) studied total sulfur concentrations in the leaves of various species was an arctic tundra catchment in Berlevåg on the Barents Sea coast of northern Norway. They did not report air quality and/or deposition data and so could not calculate correlations between air quality and/or deposition and plant and soil sulfur concentrations. At any rate, their results did not show any effects of changes in SO_2 emissions from the Kola Peninsula, and/or SO_4 in marine aerosols, on the sulfur concentrations of plant samples collected in summer 1999. For example, the total sulfur concentrations of *Hylocomium splendens* and *Pleurozium schreberi* ranged from 716 to 863 mg/kg and for *V. myrtillus* from 1720 to 2230 mg/kg. They concluded that total sulfur in moss is largely governed by the input of particulate material (dust), such as metal sulfides (Gregurek *et al*., 1999), and not by atmospheric SO_2 concentrations. The total sulfur concentrations of plant and soil samples in Berlevåg were similar to those in the Russian catchments of Vorkuta and Naryan-Mar, with all three areas showing large variations in the total sulfur concentrations in leaves both between species within each area as well as between areas (Figure 5.10, Table 5.2).

Particulate emissions that contain heavy metals are deposited closer to the emission sources than SO_2 and

Table 5.2. Median sulfur concentrations (mg/kg dry weight) in plant leaves collected from four northern European catchments in summer 1999 (Reimann *et al*., 2001c, 2003).

Catchment	Berlevåg (arctic tundra)	Monchegorsk (northern taiga)	Naryan-Mar (southern tundra with birch and shrub)	Vorkuta (moss-lichen shrub tundra with birch and willow)
Moss (*Hylocomium splendens*)	754	1090	889	824
Blueberry (*Vaccinium myrtillus*)	2025	1710		
Cowberry (*Vaccinium vitis-idaea*)	1350	2095	1300	1480
Crowberry (*Empetrum nigrum*)	1220	1260	1130	1360
Birch (*Betula pubescens*)		1775	1890	
Willow (*Salix* spp.)	3765	2335	2350	3180
Pine (*Pinus sylvestris*)		1190		
Spruce (*Picea abies*)		1985	547	

Figure 5.10. Location of the catchments included in the Barents Project (Reimann *et al*., 2003; reprinted from Reimann *et al*., 2001d).

SO_4 (e.g., Gytarsky *et al*., 1995) and therefore the relative importance of the direct and indirect harmful effects of sulfur compounds increases with increasing distance from the emissions sources. The occurrence of visible SO_2 injuries in the leaves (*B. pubescens*, *B. nana*, *V. myrtillus*, *V. uliginosum*) and Scots pine needles (*Pinus sylvestris*) in northeastern Norway close to the Russian border (Aamlid, 1993; Aarrestad and Aamlid, 1999) (Figure 5.11) has mainly been attributed to episodes with high SO_2 concentrations that occur under specific meteorological conditions (Jerre, 1994). Hilmo and Larsen (1994), in turn, reported morphological differences (growth form, fertility, apothecial morphology, discoloration) in the epiphytic lichens *Hypogymnia physodes* and *Melaniella olivacea* at sites exposed to high concentrations of SO_2 in Sør-Varanger compared to lichens at an unpolluted control site. As *M. olivacea* had a larger mean cover on birch stems than *H. physodes* in the border area between Norway and Russia, and was more frequently found at the plots in the 1991 and 1993 studies (Aamlid and Skogheim, 2001), it may even be considered more tolerant to the direct and indirect effects of sulfur deposition than *H. physodes*. The zero zone of lichen cover coincided approximately with the areas where the modeled annual mean SO_2 concentrations reached 25 µg/m^3. However, sulfur compounds are not the only acidifying compounds; Hilmo and Larsen (1994) found morphological changes (discoloration, growth form) in *Parmelia sulcata* at an industrial site in Glomfjord that may be related to elevated nitrogen deposition.

The results of Tømmervik *et al*. (1998, 2003) on vegetation changes in the area of Pasvik and Zapolyarnyy/Nikel at the Norwegian–Russian border show an increase from about 7% in 1973 to about 20% in 1979, with a stabilization at approximately 30% by 1999 in bilberry and low herb vegetation formation and meadow vegetation (bilberry forests/heaths and meadow forests with mountain birch). This change coincided with the decrease in the areas of lichen-dominated vegetation and was mainly due to the transformation of dwarf shrub (*Empetrum hermaphroditum*) and lichen-dominated vegetation cover types into bilberry and *Deschampsia flexuosa*-dominated formations which appear up to 5 to 40 km from the smelters (Deyeva and

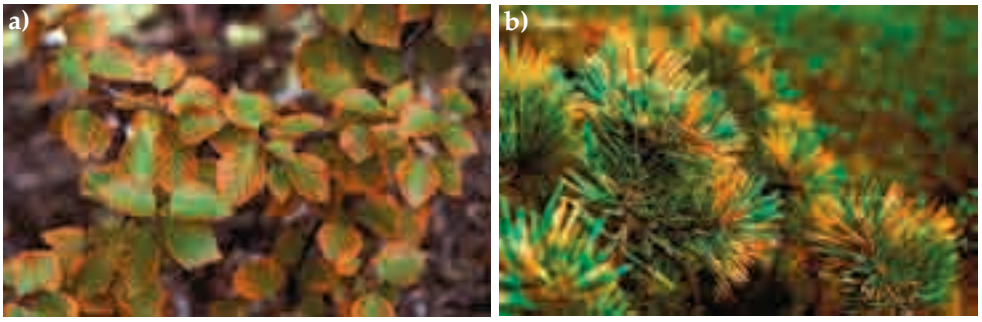


Figure 5.11. Visible (acute) SO_2 injuries on (a) birch (*Betula pubescens*) leaves and (b) Scots pine (*Pinus sylvestris*) needles at Svanvik, Norway, approximately 10 km northwest of Nikel (Photos: Dan Aamlid).

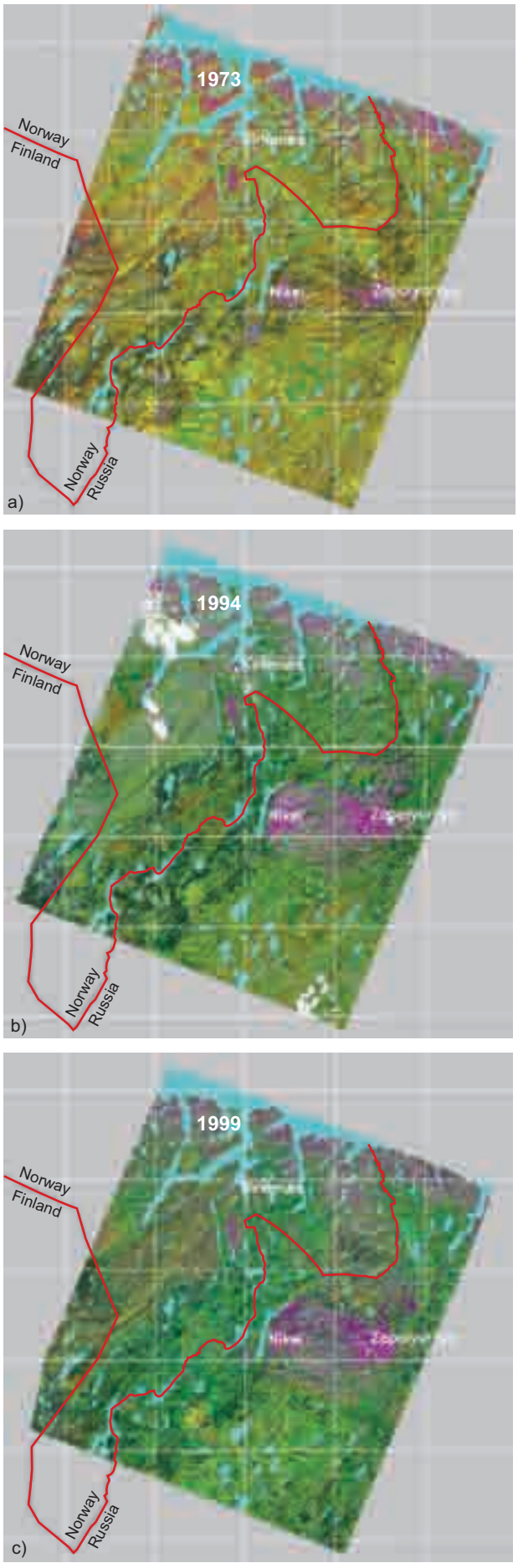


Figure 5.12. Land cover maps for the Pasvik–Nikel area for (a) 1973, (b) 1994, and (c) 1999 (Tømmervik *et al.*, 2003).

Figure 5.13. Location of joint Norwegian/Russian sites for intensive terrestrial monitoring (SFT, 2002).

Maznaja, 1993; Tømmervik *et al.*, 1995, 1998, 2003). With regard to pollution resistance of mountain birch, studies in the vicinity of the smelters at Monchegorsk may suggest phenotypic acclimatization of mature trees to a gradual increase in pollution as well as selection for pollution-resistant genotypes that may have occurred due to the exceptionally high emissions pressure over the past few decades (Valkama and Kozlov, 2001; Kozlov and Zvereva, 2004; Kozlov, 2005).

5.2.2. **Coniferous forest ecosystems**

According to Tømmervik *et al.* (2003), the vegetation cover that had the highest association (r=-0.94, p=0.001) with changes in SO_2 emissions in the Pasvik and Nikel–Zapolyarnyy areas was the mixed birch-pine forests with lichen content (Figure 5.12). The increase in the area of mixed pine-birch forests, especially from 1988 to 1999, meant that the area affected by air pollution was reduced. The most important differences in species composition in the Nikel–Pasvik area along the Norwegian–Russian border (see Figure 5.13 for the location of the Russian–Norwegian vegetation monitoring sites) were related to the loss of species richness, i.e., a lower abundance of bryophytes (e.g., *H. splendens*, *P. schreberi*, *Dicranum* species) and lichens (several *Cladonia* species) (Aamlid *et al.*, 2000), and a concomitant increase in the dominance of air pollution resistant species such as *D. flexuosa* and *E. hermaphroditum* (SFT, 2002; Tømmervik *et al.*, 2003; see also Vassilieva *et al.*, 1995) (Table 5.3). These differences are explained on the basis of both the direct impacts of pollution and the indirect soil-mediated processes leading to, for example, soil

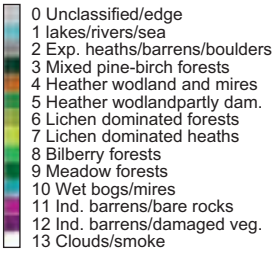


Table 5.3. Average number of species within 1 m^2 plots in pine forest along a western gradient from Nikel (SFT, 2002). The highest values for each species group (bold font) indicate the change from lichen- and moss-dominated vegetation to small shrub- and grass-dominated vegetation near the emissions sources.

Species group	Sample plots and their distance from Nikel						
	45 km south	25 km west	16 km west	12 km west	9 km northwest	6 km north	6 km west
Small shrubs	3.5	2.9	3.1	3.1	3.4	**4.2**	4.0
Herbs	1.9	1.9	1.7	0.8	1.9	**2.0**	1.1
Mosses	**5.1**	4.6	4.7	4.7	4.6	1.2	2.1
Liverworts	1.3	1.5	**2.0**	1.5	1.6	0.7	1.2
Lichens	3.7	4.1	7.5	**8.2**	3.9	2.3	3.1
All species [a]	15.4 (24)	14.9 (30)	**19.2 (32)**	18.7 (31)	15.6 (26)	10.9 (23)	11.5 (17)

[a] maximum number of species in brackets

acidification and nutrient imbalances (Tømmervik *et al*., 1995; Aarrestad and Aamlid, 1999; Koptsik *et al*., 1999a,b, 2003; SFT, 2002). Furthermore, for example, *Pohlia nutans* is a moss species that has increased its cover in the most polluted area. This species, characterized by wide ecological amplitude, colonized the emptying litter in industrial barren land in the absence of competition from other (moss) species more sensitive to pollution (Koptsik *et al*., 2003).

According to Kashulina *et al*. (1997, 2003), direct exposure to SO_2 is the most likely, although not the only mechanism of vegetation damage in the eight catchments located on the Kola Peninsula and in northeastern Finland and Norway. The highest sulfur concentrations in plant samples in the most polluted sites were usually 2- to 4-fold higher than at control sites (Gytarsky *et al*., 1995; Äyräs *et al*., 1997; Aamlid *et al*., 2000; Steinnes *et al*., 2000; Reimann *et al*., 2001a,c). Steinnes *et al*. (2000) reported a decrease from 4270 to 2000 mg/kg and from 2820 to 1420 mg/kg in the mean total sulfur concentration of *B. pubescens* and *E. hermaphroditum* leaves, respectively, on moving from the most polluted study sites around Nikel to the southernmost sites more close to background conditions in 1991. A decrease from around 3500 to 1000 mg/kg, and from around 3400 to 1500 mg/kg in the foliar sulfur concentration of *Betula* sp. and *V. myrtillus*, respectively, between the areas closest to the Nikel smelters (<5 km) and background areas (about 30 km) was, in turn, reported by Gytarsky *et al*. (1995). Sulfur was also the major element in moss (*H. splendens* and *P. schreberi* 2090-543 mg/kg) and the O and B soil horizons in the 1995 survey in northern Finland, Norway, and Russia (Äyräs *et al*., 1997; Kashulina *et al*., 1997, 2003; Kashulina and Reimann, 2001, 2002; Reimann *et al*., 2001a).

Reimann *et al*. (2001b,c,d, 2003) measured total sulfur concentrations in the leaves of several plant species collected from nine catchments in northern Europe (Finland, Norway and Russia) in summer 1999. Only Norway spruce (*Picea abies*) needles showed significantly higher sulfur concentrations in all samples collected near Monchegorsk. Correlation between 'available' sulfur in surface soil and observed foliar sulfur concentrations for all plants growing in the soils was in general very low. Furthermore, Reimann *et al*. (2003) concluded that macroscopic and microscopic evidence of leaf damage are more reliable indicators of pollution impact than foliar sulfur concentration. In a study carried out in the early 1990s on Scots pine, Rautio and Huttunen (2003) also found a weak relationship between element concentrations in the soil and internal foliar concentrations. A factor representing high foliar levels of Ni, Cu, and sulfur, and low levels of Zn and Mn, was found to explain most of the variation in the number of needle age classes and tip necrosis. The macroscopic injury variables (including stomatal chlorosis and other discolorations) correlated clearly with the modeled SO_2 concentration in air. However, the cell injuries studied did not show any distinct relationships with element concentrations in either foliage or deposited particles (Rautio *et al*., 1998c).

High variation in the total sulfur concentrations in plant samples collected during the growing season may be explained (in addition to species-specific characteristics and differences in gaseous and particulate sulfur deposition) by nutrient status, pH, and other soil characteristics, as well as by climate, composition of the vegetation layer and altitude (Reimann *et al*., 2001b, 2003). For example, despite higher SO_2 emissions from the Nikel smelter compared to the Monchegorsk smelter, plants accumulate more sulfur near the latter because of the specific combination of geomorphological and meteorological conditions (Koptsik *et al*., 1999a). It is also well known that plant nutrient concentrations (including sulfur) vary considerably during and between growing seasons. For example, the average sulfur concentrations of current-year Scots pine needles collected in August-October between 1996 and 1999 were 755 to 901 mg/kg at Värriö in the eastern part of Finnish Lapland and 806 to 988 mg/kg at Sammaltunturi (i.e., Pallas) in the western part of Finnish Lapland (Sirkku Manninen, University of Helsinki, unpubl. data). This is why needle samples for bioindicator studies mapping the dispersal of sulfur emissions, as well as the area possibly affected by sulfur deposition, are usually collected in winter, i.e., when emissions are at their highest and the trees are physiologically relatively inactive (see e.g., Manninen *et al*., 1997a, 1998; Rautio *et al*., 1998a,b).

The latitude-related decrease in the total sulfur concentration on moving northward (Reimann *et al*., 2003) may be related to the length of the periods during which the plants have stomatal uptake of SO_2 versus the time when they are exposed to non-stomatal deposition only, as the proportion of (winter) non-stomatal deposition increases towards the north. The dry deposition of sulfur dominates in northern areas due to the low precipitation, and dry deposition accounts for about 80% of the total deposition (Tuovinen *et al*., 1993). On the other hand, as most of the plants have a snow-cover for six to seven months of the year, more than 50% of the total annual deposition will deposit onto snow (Kashulina and Reimann, 2002). This means that the sulfur concentrations of mosses and lichens which are thallophytes (i.e., they have no roots but obtain all their nutrients through their above-ground parts as ions dissolved in water – rain and melting snow), may give a different picture about the rate and distribution of sulfur emissions compared to that given by foliar sulfur analyses of evergreen conifers. Steinnes (1995) pointed out that it may, however, be feasible to achieve representative results (at least for heavy metals) by carefully selecting the sampling site for moss in order to avoid interference from wind erosion or surface water flow. On the other hand,

the lower growth rate of mosses in arctic regions may be expected to lead to greater uptake of airborne trace elements per unit weight than in mosses at more southerly latitudes. Furthermore, the different growth structure of the plant may make it more difficult to accurately define the exposure period.

In addition to the total sulfur concentration of Scots pine needles, statistically significant correlations ($p<0.001$) were found between the annual mean SO_2 concentration and needle sulfate concentration and S:N ratio at the sites close to Monchegorsk in the early 1990s (Manninen *et al*., 1998) (Table 5.4). Koptsik *et al*. (1999a, 2001) also found a marked gradient in the needle total sulfur concentration and S:N ratio of Scots pine with increasing distance from the Nikel smelters in the early 1990s. The increase in the S:N ratios towards the Nikel smelters is not just explained by the increasing needle total sulfur concentrations, but by lower nitrogen supply from the soil observed as decreasing concentrations of nitrogen in soil organic horizons toward the smelter. In addition to nitrogen concentrations, those of ammonium acetate-extractable Mg and K in organic horizons tended to be lower within several kilometers of the Nikel smelters compared to remote sites (Koptsik *et al*., 1999b, 2001, 2003) as they also were around the Monchegorsk smelters (Lukina and Nikonov, 1999, cited in Zvereva and Kozlov, 2005). With regard to temporal trends related to the decrease in SO_2 emissions and, consequently, in the ambient SO_2 concentrations in the border areas of Norway and Russia (Hagen *et al*., 2005), the results of Koptsik *et al*. (G. Koptsik, Moscow State University, pers. comm.) suggested a decrease in the total sulfur concentration of current-year Scots pine needles from August 1991-1994 to August 2002. At any rate, as there are differences in foliar responses to elevated sulfur and nitrogen deposition between coniferous tree species (Manninen and Huttunen, 2000; Luyssaert *et al*., 2005), more research is needed on foliar sulfur and nitrogen fractions and ratios, as well as on Mg:N and Ca:Al ratios, in the northernmost areas for use as indicators of acidification.

5.2.3. **Reindeer grazing, climate change, nitrogen deposition, and other factors**

Reindeer grazing is one factor that modifies or interacts with the effects of sulfur deposition on vegetation, especially in northern Norway. According to Gaare and Tømmervik (2000), reindeer have grazed down the new 'reindeer' lichens in the period 1992 to 1999, i.e., after the emissions reductions at the smelters on the Kola Peninsula. There has also been extensive forestry in the southern part of the Pasvik area and this has led to reductions in lichen cover (succession) and pine forests. Severe desiccation of the forest vegetation close to the smelters at Nikel and Zapolyarnyy has led to increasing amounts of dead needles and woody debris, which in turn has led to more frequent wild forest fires (Tømmervik *et al*., 2003). Moreover, outbreaks by caterpillars such as *Epirrita autumnata* have been followed by successions from lichen-dominated vegetation to more dwarf shrub- and grass-dominated vegetation (Lehtonen, 1987; Tenow and Bylund, 2000).

Over most of northernmost Europe, the trend towards a shift in lichen-dwarf shrub dominated forests to more grass- and herb-dominated forests, may reflect a situation where slow-growing species such as lichens, crowberry (*E. hermaphroditum*), and cowberry (*V. vitis-idaea*) are declining due to intensive reindeer grazing (Olofsson *et al*., 2001). Tømmervik *et al*. (2004) also gave climate change (increased precipitation) and long-range transported nitrogen pollution (increasing nitrogen deposition is also linked to increasing precipitation) as possible accentuators for the increase of birch forests and vegetation types dominated by bilberry, wavy hair-grass (*D. flexuosa*), the dwarf cornel (*C. suecica*), and mosses (*P. schreberi*, *H. splendens*) in northern Norway over the last 40 years.

Increased nitrogen deposition through precipitation and/or deposition of manure (feces) from the reindeer may create unfavorable living conditions for slow-growing species. Bobbink *et al*. (2002) considered an empirical critical load of 5 to 10 kg N/ha/yr, based on field manipulations in which exceedance was indicated by changes in biomass and in the moss layer, reliable for tundra ecosystems. They gave the same critical load for arctic, alpine, and sub-alpine shrub habitats (with exceedance indicated by a decline in lichens, mosses, and evergreen dwarf-shrub), but its reliability was referred to as 'expert judgment' (see also UNECE, 2004a). Recent results from field experiments in an area of northern Sweden with low background nitrogen deposition and from a large-scale monitoring study, show that important vegetational changes start to occur when adding low nitrogen doses and that recovery of the vegetation after nitrogen inputs stop is a very slow process. Based on data from the Swedish research program Abatement Strategies for Transboundary Air Pollution, Nordin *et al*. (2005) suggested that the critical load should be lowered from the current 10 to 15 kg N/ha/yr to a level of 6 kg N/ha/yr to protect the biodiversity of the boreal forest understorey vegetation. The impact of elevated nitrogen

Table 5.4. Concentrations (mg/kg) and ratios of sulfur and nitrogen in current-year Scots pine needles collected at different distances from the smelters on the Kola Peninsula showing change along the pollution gradients and seasonal variation in needle chemistry.

	Sampling	Total S[a]	Organic S	SO_4-S	SO_4-S/organic S[a]	Total N	Total S/Total N[a]	Reference
Finnish Lapland[b]	Sep-Oct 1990, Sep 1992	720-1153	479-862	141-361	0.20-0.53	9200-13000	0.032-0.045	Manninen *et al*., 1997b
Monchegorsk-Jena[c]	Apr 1991 Jul 1992	856-2548 794-1474	672-1599 619-971	105-1297 168-656	0.14-1.04 0.21-0.80	8100-13800 8400-14200	0.032-0.089 0.034-0.055	Manninen *et al*., 1998
Norwegian-Russian border[b]	Aug 1991-1994 Aug 2002	499-1917 896-1248				13400-21300	0.031-0.046	Koptsik *et al*., 1999a; 2001; pers. comm.
Monchegorsk-Finnish border[d]	Sep 1990 Apr 1991	1112±471 1321±499						Rautio *et al*., 1998b

[a] The background total sulfur concentration is around 600 mg/kg, SO_4-S concentration 100 to 200 mg/kg, and total sulfur:total nitrogen ratio 0.028 (on a gramme-atom basis) (Manninen, 1995 and references therein); [b] min–max site means , [c] min–max values for individual tree, [d] mean±SD for all sites.

deposition was seen as decreased abundance of *V. myrtillus* and *V. vitis-idaea* and increased abundance of *D. flexuosa* in particular, as well as decreased abundance of *P. schreberi* and *H. splendens*.

Care must be taken when drawing conclusions about nitrogen deposition on the basis of plant nitrogen analyses. For example, Walker *et al*. (2003a) found marked regional gradients in terricolous lichen nitrogen concentrations in the Usa river basin, northeast European Russia, suggesting that there were concomitant gradients in nitrogen deposition. On the 240 km south-to-north transects through the town Inta, values of $[N]_{apex}$ (the apical 5 mm part) in *Cladonia stellaris* decreased from 0.57±0.01 mmol/g to 0.43±0.01 mmol/g. Moreover, elevated $[N]_{apex}$ in *C. arbuscula*, and to a lesser extent in *Flavocetraria cucullata*, in the Vorkuta area suggested elevated nitrogen deposition due to local industrial pollution. Variation in $[N]_{apex}$ in *C. arbuscula* correlated well with other pollutant signals in snow ($[SO_4^{2-}]_{snow}$, $[Ca^{2+}]_{snow}$, $[K^+]_{snow}$, pH) (Walker *et al*., 2003b) as well as thalli, but as on the other transects, it was not correlated with $[NO_3^-]_{snow}$. Molar concentrations of organic nitrogen were broadly similar to those of nitrate, except at sites close to Vorkuta where $[organic\ N]_{snow}$ values were greater than $[NO_3^-]_{snow}$ by a factor of 2 or 3, varying spatially in a manner similar to that of $[SO_4^{2-}]_{snow}$. Thus Walker *et al*. (2003a) concluded that a higher deposition rate of organic nitrogen at the sites close to Vorkuta might have contributed to elevated $[N]_{apex}$ in this area. In addition, they did not exclude that impaired lichen growth at sites close to Vorkuta, perhaps due to phytotoxic air pollutants, might have resulted in higher tissue nitrogen concentrations. They emphasized that data for snow pack chemistry were for a single winter period, whereas $[N]_{apex}$ values probably gave an indication of nitrogen deposition integrated over a period of at least one to two years. Furthermore, that these interpretations were made in the absence of commensurate data on $[NH_4^+]_{snow}$.

According to Walker *et al*. (2003a), mat-forming lichen cover was generally poor in the Vorkuta region due to heavy grazing and trampling by reindeer; for example, *C. stellaris* was largely absent at most sites. Virtanen *et al*. (2002), in turn, identified two impact zones around Vorkuta on the basis of remote sensing data: (1) a pollution zone (150–200 km^2), where most of the lichen species are absent and changes in vegetation communities' species composition in all main plant groups are obvious (willows especially being more dominant than in unpolluted sites), and (2) a slight pollution/disturbance zone (600–900 km^2), where vegetation changes are mainly similar but less so than the changes in the first zone – in particular the amount of herbs and grasses is increased relative to unpolluted areas. The upper soil layers (15–30 cm) in Vorkuta, where atmospheric pollution is mainly caused by dust from open coalmines, emissions from coal combusting power plants and a cement factory, and burning of waste rock near the coalmines, have become chemically modified with pH ranging from 6.7 to 8.9 indicating strong alkalinization, and concentrations of exchangeable Ca and total nitrogen are 10 to 20 and 2 to 4 times higher, respectively, than at comparable non-polluted sites. Heavy metal levels in soils are also increased in the area (Getsen *et al*., 1994, cited in Virtanen *et al*., 2002). Emissions data for the 1990s suggested that SO_2 emissions averaged around 40 000 tonnes per year from the Vorkuta industrial complex in the Komi Republic (Virtanen *et al*., 2002). The only information found on the effects of emissions on terrestrial vegetation in the Norilsk area was that by Tutubalina and Rees (2001) who reported a loss of vegetation from the city of Norilsk between 1961 and 1995 and the growth of landscape damage over a larger area between 1972 and 1995. Several studies on the spatial and temporal dynamics of forest decline in this area were published in Russian.

5.2.4. Needs and recommendations for future research and monitoring

In the multi-medium ecogeochemical mapping carried out in the European Arctic (Russia, Finland and Norway) in the mid-1990s, the only sample material in which the highest bulk sulfur deposition figures were directly reflected in a subarctic birch forest catchment close to Zapolyarnyy was vegetation (Äyräs *et al*., 1997; Kashulina *et al*., 1997, 2003; Kashulina and Reimann, 2001, 2002; Reimann *et al*., 2001a). However, the almost 10-fold increase in the bulk sulfur deposition on moving from a coniferous forest catchment (0.17 g S/m^2/yr) at Pallas in the western part of Finnish Lapland to the subarctic birch forest catchment near Zapolyarnyy (1.5 g S/m^2/yr) resulted in only a 2-fold increase in the sulfur concentration of moss samples (from a median of 800 to 1500 mg/kg).

Gytarsky *et al*. (1997) calculated that the annual dry deposition of sulfur is about 2-fold higher in deciduous forests compared to coniferous forests on the Kola Peninsula. Taking into account the high proportion of dry-deposited sulfur, they expected that the values of total sulfur deposition over the slightly damaged area (based on the classification by Boltneva *et al*., 1982; Karaban *et al*., 1988), where the modeled growing season mean SO_2 concentration was 21 µg/m^3, would increase to 0.6 to 1.0 g/m^2/yr in coniferous forests and 1.2 g/m^2/yr in deciduous forests (Gytarsky *et al*., 1995, 1997). In general, dry deposition to coniferous forest is considered to be higher than that in deciduous forest. The dry deposition velocity of 1.1 cm/s for SO_2 on deciduous forests during the vegetation period (McMahon and Denison, 1979) versus that of 0.3 cm/s on coniferous forests (Garland, 1977) used by Gytarsky *et al*. (1997) in their calculations may explain the high sulfur deposition in deciduous forest. At any rate, the values given by Gytarsky *et al*. (1995, 1997) are 4-fold higher than the proposed SO_2 critical level of 5 µg/m^3 as a growing season mean (Manninen, 1995; Manninen and Huttunen, 2000; see also Tsvetkov, 1993), as well as the critical load of 0.32 g/m^2/yr derived for the highly sensitive forest ecosystems of northern Europe (Downing *et al*., 1993). The summertime (April–September) mean SO_2 concentrations have been around 5 to 10 µg/m^3 at Svanvik (about 10 km west of Nikel) and reached 150 µg/m^3 at Nikel in the late 1990s (Hagen *et al*., 2001, cited in SFT 2002). Thus acute and chronic impacts of SO_2 and sulfur deposition still occur in the area. There is a need for measurements of dry deposition velocities on different vegetation types.

According to Alexeyev (1995), the least studied but most important stage of air pollution impacts on arctic and subarctic terrestrial ecosystems is the initial damage, when the negative irreversible processes of ecosystem destruction are still preventable. Simultaneous determination of several signals can provide an early warning of damage to terrestrial vegetation by low-level airborne pollutants. It is recommended that studies be carried out at permanent plots (in cooperation with air quality, deposition, soil and faunal monitoring) in order to assess the state and

functioning of the tundra, mountain birch and coniferous forest ecosystems. These studies should focus on the biomass (growth) and species composition and coverage of field and ground layer vegetation, the condition and frequency of epiphytic lichens, visible foliar injuries, foliar sulfur and nitrogen concentrations, and ratios of S:N, Mg:N and Ca:Al using standardized methods (including different sampling dates for the coniferous, deciduous and thallophytic key species). In particular, more attention should be paid to nitrogen-related issues and interactions. Lichens (e.g., *Lobaria*, *Peltigera* and *Stereocaulon* species and *Nephroma arcticum*) containing cyanobacteria are abundant in arctic ecosystems and may account for a major part of the nitrogen fixed in these systems. Their nitrogen fixation may be even more sensitive to acidity than photosynthesis and may thus be decreased by decreasing pH (Nash and Gries, 1995). Climate change and elevated nitrogen deposition are expected to affect negatively the slow-growing lichen vegetation in particular (e.g., Cornelissen *et al*., 2001; Gordon *et al*., 2001; Fremstad *et al*., 2005) and higher CO_2 concentrations and UV-B radiation may reduce nutrient cycling and so may potentially reduce ecosystem productivity (Callaghan, 2005 and references therein).

On the other hand, if the most sensitive tundra vegetation is at least partly replaced by more tolerant forest vegetation over the next 100 years (Kaplan *et al*., 2003) and SO_2 emissions do not increase, the state of the vegetation may (continue to) improve – also taking into account that plant compensatory responses as well as allocation to reproduction are species specific (e.g., Zvereva and Kozlov, 2001, 2005). Alekseev and Soroka (2002) reported that a higher growth rate occurred in Scots pine on the northwestern Kola Peninsula despite significant air pollution. Generally, the younger the trees, the greater the growth increase. The most probable reasons for the marked increase in radial increment growth of Scots pine in the region are climate warming (including an increase in growth season length) and higher levels of CO_2 as well as changes in forest fire frequencies (increased frequency and, consequently, a shift towards younger forests). The lack of significant differences in diameter growth between trees in different damage classes is explained by Alekseev and Soroka (2002) by the low number of samples on severely damaged trees, which are most susceptible to growth suppression by pollution.

5.3. Effects on fauna

5.3.1. Effects on birds and mammals

Acidification and acidic deposition typically affect terrestrial vertebrates indirectly. There are only a few studies concerning the direct effects of acidification on mammals and birds, but indirect effects mediated through exposure to metals in food or water have been reported (Dudley and Stolton, 1996). On the Kola Peninsula, all the vertebrate species studied, for example, root vole (*Microtus oeconomus*), grey-sided vole (*Clethrionomys rufocanus*), common shrew (*Sorex araneus*), willow grouse (*Lagopus lagopus*) and common frog (*Rana temporaria*), had elevated heavy metal concentrations in liver, bone, muscle, and skin. The accumulation of heavy metals seems to depend on trophic level (AMAP, 2005).

5.3.1.1. Mammals

Mitotic activity of spleen and cornea cells in root voles of different age and sex was higher 4 km from the Monchegorsk smelter than 28 km from the smelter. Mature root vole males had a lower hemoglobin content at 4 km than at 28 km. The mean frequency of chromosome aberrations in spleen, cornea, and marrow cells of root voles ranged from 34 to 50%. In spleen and cornea cells there were no differences in aberration frequency between 4 and 28 km, but in marrow cells the frequency of aberrations was significantly higher at 4 km than at 28 km (Kataev and Popova, 1993).

Males of the field vole (*Microtus agrestis*) collected 4 km south of the Monchegorsk smelter had lighter livers and heavier spleens than males collected 28 km away (Kataev and Popova, 1993). However, correlation analysis based on data from eight sites located 3 to 52 km south of Monchegorsk did not show any relationship between body mass of adult grey-sided voles and distance from the smelter (Kataev *et al*., 1994). The proportion of pregnant females was higher in unpolluted sites, but the number of fetuses per female was the same along the pollution gradient (Kataev *et al*., 1994). However, resorption of embryos was much higher at the most polluted site (36.8%) than at the control site (3.8%) (Kataev and Makarova, 1984).

The sex ratio of grey-sided voles near the Monchegorsk smelter was around 1:1, while males made up 65% of the population at an unpolluted forest site. The age structure of the populations was also different: no overwintered individuals were found in the polluted sites compared to 5.9 to 7.7% of the population overwintering in unpolluted forests (Kataev and Makarova, 1984). The proportion of adult voles in catches from the moderately polluted sites around the Monchegorsk smelter was higher than that at unpolluted sites (Kataev *et al*., 1994), suggesting that juvenile mortality is higher at moderately polluted sites than unpolluted sites.

The density of grey-sided voles, the most abundant vole species, was lowest close to the Monchegorsk smelter and increased with distance to the farthest, less-polluted trapping sites. The bank vole, red vole (*Clethrionomys rutilus*) and Norway lemming (*Lemmus lemmus*) were absent from the most severely damaged area, and were also scarce at the moderately polluted area 28 km south of the smelter (Kataev *et al*., 1994). Snap trapping in the surroundings of Monchegorsk revealed six species of small mammals; the common shrew and grey-sided vole were the most common in unpolluted forests. Only the grey-sided vole and root vole were captured in industrial barrens (Kozlov *et al*., 2005a).

A long-term study by Kataev (1995) revealed a monotonous decline in the population of small mammals on approaching the Monchegorsk smelter: the biomass decreased from 87 kg/km^2 at the unpolluted forest site to 69 kg/km^2 at the moderately polluted site to 15 kg/km^2 at the most polluted site. However, the pattern in 2001 (Kozlov *et al*., 2005a) differed from earlier data (Kataev, 1995): the density decline in the most polluted sites was much smaller (51.8% of the background value) and, most importantly, the highest densities were associated with slightly to moderately polluted forest. This result may be partially explained by the presumably higher plant quality in moderately polluted sites: grey-sided voles preferred bilberry shoots at distances of 20 to 40 km from Monchegorsk compared to shoots from sites both closer and further away (Suomela and Palokangas, 1993).

Although pollution may have suppressed the cyclic density fluctuations of the bank vole (Figure 5.14), this and many other results obtained near Monchegorsk should be viewed as tentative due to the non-replicated experimental design (one polluted vs. one or two unpolluted sites).

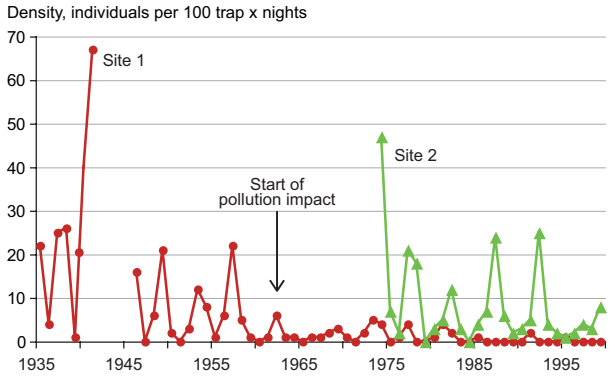


Figure 5.14. Effects of pollution on density of bank vole (*Clethrionomys glareolus*) in Lapland Biosphere reserve (the break in observations is due to the Second World War). Site 1 is located 30 km south of Monchegorsk; it was considered unpolluted until 1958–1960, when the first signs of forest damage by emissions were detected. Site 2 is located 30 km west of the smelter and represents an unpolluted control site (protected from emissions by the Monche-tundra mountain range) (after Kozlov and Barcan, 2000).

Hörnfeldt (2004) listed several hypotheses to explain the long-term decline in vole numbers and changes in vole dynamics. One hypothesis attributed the decline to the decrease in quantity or quality of food, or to the decrease in availability of natural shelter for voles. Lichens are an important winter food for bank voles (*Clethrionomys glareolus*), and another hypothesis explained the changes in regular 3- to 5-year, synchronous, high amplitude northern Fennoscandian vole dynamics by the decline in pendulous lichens (Hansson, 1999). Thus, acidification might indirectly affect vole dynamics. Voles, especially *Microtus* voles, the grey-sided vole, and the bank vole, are key species in northern vertebrate communities, and changes in their dynamics may affect many of the avian and mammalian predators dependent on voles as prey (e.g., Hanski *et al*., 2001). The supply of bilberry (*Vaccinium myrtillus*) may also affect vole dynamics (Hörnfeldt, 2004). Bilberry is sensitive to the direct effects of SO_2 (SFT, 2002) and, together with birch, is regarded as one of the most suitable bio-indicators for monitoring pollutants (Steinnes *et al*., 2000). Bilberry is an important food item for the grey-sided vole (Kalela, 1957) and bilberry stands provide shelter for grey-sided voles against predators (Löfgren, 1995).

The densities of large mammals also declined on approaching the Monchegorsk smelter. Although this pattern seems trivial, no data on the effects of pollution on the abundance of large mammals had ever been published prior to the study by Kozlov *et al*. (2005a). During the winter the tracks of eight species of large mammal were recorded. Mountain hare (*Lepus timidus*) and red fox (*Vulpes vulpes*) were the most common, and only these two species were recorded in industrial barrens near Monchegorsk (Kozlov *et al*., 2005a).

5.3.1.2. **Birds**

Changes in reproduction have been documented for three abundant hole-nesting bird species in the impact zone of the Monchegorsk smelter: redstart (*Phoenicurus phoenicurus*), pied flycatcher (*Ficedula hypoleuca*), and Siberian tit (*Parus cinctus*). Up to 80% of breeding attempts by these species failed in the most polluted habitats; as a whole, reproductive success in industrial barrens was five times lower than in background areas. In severely polluted sites, 46% of nests were abandoned in the nest building phase (9% in an unpolluted site), with 36% of nests abandoned before completing the clutch (3% in an unpolluted site) (Gilyazov, 1993).

In the 1980s, the densities of breeding birds in moderately and severely polluted habitats along the Monchegorsk pollution gradient were estimated to be 60 and 13%, respectively, of the density in the unpolluted forests (Gilyazov and Kataev, 1990; Gilyazov, 1993). The results of recent counts are in good agreement with these earlier data: bird biomass declined monotonously on approaching the pollution source and was reduced to 12.7% in the industrial barrens relative to unpolluted forests (Kozlov *et al*., 2005a).

In the zone of declining forests (about 15 km south of Monchegorsk), bird abundance was reduced to around two-thirds of that in unpolluted forests. In particular, the densities of typical northern taiga species like tree pipit (*Anthus trivialis*), redstart, redwing (*Turdus iliacus*), song thrush (*T. philomelos*), willow warble, pied flycatcher, and Siberian tit were lower than in control sites. In this zone, the abundances of capercaillie (*Tetrao urogallus*), hazel grouse (*Bonasia bonasia*), willow grouse, and black grouse (*Tetrao tetrix*) had been reduced by 1988 to approximately half the numbers observed in 1963 (2.5 versus 4.9 ind/km^2, respectively), while no reduction was recorded in unpolluted forests (7.4 versus 7.3 ind/km^2) (Gilyazov, 1993). Since the 1970s, hazel grouse, *Strix* owls, eagle owl (*Bubo bubo*), Tengmalm's owl (*Aegolius funereus*), and treecreeper (*Certhia familiaris*) have not been recorded closer than 40 km from Monchegorsk and birds of prey did not nest in this area (Gilyazov, 1993).

Counts of 16 bird species around the Monchegorsk smelter demonstrated that, in spring, birds migrate to heavily polluted areas, possibly because of the earlier disappearance of the continuous snow cover, which is reduced to a third of the background depth (Kozlov, 2001). Later in the season, however, they return to the surrounding (moderately contaminated) forest habitats. As a result, during the spring migration the density of birds in heavily polluted habitats was, on an average, 1.3 times higher than in moderately polluted sites. In contrast, during the nesting period bird density in heavily polluted sites was 2.5 times lower, and during the post-nesting period 15 to 20 times lower than in moderately polluted sites (Gilyazov, 1993).

Species richness of birds in an industrial barren south of Monchegorsk, estimated by observations repeated during 1977 to 1990, was reduced by two-thirds relative to two unpolluted sites (Gilyazov, 1993). Similarly, the number of bird species recorded in 2001 by standard surveys (three 1 km-long transects per site) decreased from 11 to 15 in unpolluted forests to 3 to 4 in industrial barrens near Monchegorsk (Kozlov *et al*., 2005a).

Species characteristic of tundra and forest-tundra, such as yellow wagtail (*Motacilla flava*), meadow pipit (*Anthus pratensis*), bluethroat (*Luscinia svecica*), wheatear (*Oenanthe oenanthe*), little bunting (*Emberiza pusilla*), and redpoll (*Carduelis flammea*), all of which are migratory species, constituted about 40% of the records at a barren site. Willow warble represented 50 to 80% of the northern taiga species complex (redstart, redwing, willow warble, pied

flycatcher, Siberian tit and brambling (*Fringilla montifringilla*)) in undisturbed conditions, but at a barren site its share did not rise above 15%. Both willow warble and meadow pipit were relatively abundant, possibly because of habitat changes (replacement of coniferous forests by birch and willow bushes, along with replacement of ericaceous dwarf shrubs by grasses) that favor these species. Also, the abundance of crows (*Corvus corone*) increased in polluted habitats relative to unpolluted forests (Gilyazov, 1993).

5.3.1.3. **Concluding comments on birds and mammals**

The most likely explanation for the observed changes in vertebrate populations near Monchegorsk is habitat deterioration. Forest decline results in a decrease in the number of nesting sites for birds; however, the increase in secondary open habitats favors certain bird species. Changes in the abundance of invertebrates (see section 5.3.2) may influence food availability for insectivorous birds – birds in the polluted area were reported to be suffering from nutritional stress (Framstad, 2002; Eeva *et al*., 2003). The low number of microtine rodents in the damaged and moderately polluted areas is due to a decrease in the quantity of important food plants: epiphytic lichens for bank vole and possibly also for red vole, mosses for Norway lemming, and vascular plants, especially bilberry, for grey-sided vole (Kalela, 1957; Kataev *et al*., 1994; Löfgren, 1995; Hansson, 1999; Hörnfeldt, 2004).

Although there is almost no direct evidence of pollution-induced mortality of vertebrates in the Arctic, data on the population densities of a carrion-feeding beetle, *Nicrophorus vespilloides*, indicate an increase in the mortality rates of vertebrates in moderately polluted forest habitats around Monchegorsk (Kozlov *et al*., 2005a). Even if pollutants do not kill the birds and mammals, mortality may increase due to a reduced ability to sustain environmental stresses such as disease, low winter temperatures, and shortage of food (Heinz, 1989; Hörnfeldt, 2004). Winter mortality (e.g., Novikov, 1981) may be especially high in the contaminated habitats.

The increase in mortality, in the absence of a compensatory increase in reproduction (as reported in earlier studies), suggests that populations in the polluted (low quality) habitats are supported by migration from the surrounding unpolluted territories, and that migrants are not able to assess habitat quality in terms of toxic contamination before breeding. Moderately polluted habitats may be attractive for dispersing birds and mammals not only because of the permanent vacation of territories due to high mortality, but also because some resources are more abundant there than in the surrounding unpolluted forests. The contaminated area of about 1000 km^2 around the Monchegorsk smelter complex acts like a death trap for dispersing birds and mammals, and the moderately contaminated forested part of this area can possibly be classified as an attractive sink habitat (Kozlov *et al*., 2005a).

5.3.2. **Effects on invertebrates**

The effects of acidifying pollutants on invertebrates have been explored to a much lesser extent than many of the other components of terrestrial ecosystems, and the previous AMAP assessment (AMAP, 1998) contained no information on invertebrates. However, recent studies conducted mainly in the vicinity of the Monchegorsk nickel-copper smelter on the central part of the Kola Peninsula provide some information on the responses of several groups of invertebrates, primarily insect herbivores, to pollution.

Empirical data collected in the vicinity of the point sources, which emit an extremely wide range of metals and other compounds, can not be used to unequivocally attribute observed effects to a specific pollutant. Nevertheless, many pollutants are considered to have a minor impact on biota around point sources. In particular, there has been no correlation found between larval weight for the moth *Eriocrania* and nickel concentrations in the larval body (Kozlov *et al*., 2000), which agrees with the reported resistance of many insects to heavy metals (Heliövaara and Väisänen, 1993; Boyd and Martens, 1994). In contrast, both direct and indirect effects of SO_2 on Lapland leaf beetle (*Chrysomela lapponica*) and autumnal moth (*Epirrita autumnata*) have been confirmed experimentally (Kozlov *et al*., 1996c; Kozlov and Selikhovkin, 1997). Therefore in this assessment changes observed in invertebrate communities are linked to both the direct and indirect effects of acidifying pollutants, primarily to changes in habitat structure following forest decline (Rigina and Kozlov, 1999). This supports the conclusion in the previous AMAP assessment (AMAP, 1998), that the deterioration of ecosystems around the smelter complexes on the Kola Peninsula is mainly due to pollution by high concentrations of SO_2, leading to extreme cases of acidification.

5.3.2.1. **Size, individual performance, and population structure**

Insect size frequently declines with an increase in pollution levels (Heliövaara and Väisänen, 1993). However, this trend has not been found in the Monchegorsk area: weight of the Lapland leaf beetle, *Chrysomela lapponica* (Zvereva *et al*., 1995a), femur length in the autumnal moth (Ruohomäki *et al*., 1996), and the length of the hind tibia in black fly (*Simulium pusillum*) (Kozlov *et al*., 2005b), were independent of pollution load. The absence of a pollution effect in *S. pusillum*, together with an extremely low abundance of black fly near the Monchegorsk smelter, implies that at least some of the specimens from the most heavily polluted sites may represent occasional migrants rather than locally breeding populations. However, this explanation is not valid for *C. lapponica* or the autumnal moth as their larvae were collected in heavily polluted sites.

Despite some biochemical adaptations to pollution (Zvereva *et al*., 2003), the individual performance of *C. lapponica* beetles collected in heavily polluted, industrial barrens was lower than in unpolluted forests; females laid less eggs and egg hatchability was significantly lower (Zvereva *et al*., 1995a).

Although an increase in the rate of melanic specimens with an increase in pollution is well documented (Brakefield, 1987), this has not been reported for the Arctic. There were no melanic specimens among geometrid and noctuid moths collected around the Monchegorsk smelter by bait-traps during 1991 to 1994 (Kozlov, 1997). Similarly, frequencies of melanic morphs of the Lapland leaf beetle *C. lapponica* were independent of pollution load, despite pronounced variation in morph frequencies among study sites and differences between dark and light morphs in both size and performance (Zvereva *et al*., 2002).

Changes in spatial structure of insect populations are limited to observations on their distribution among and

within host-plant individuals. Distribution of mines of two moths, *Eriocrania* spp. on birch and *Phyllonorycter strigulatella* on elder, is more aggregated near the pollution source (Kozlov, 1987, 2003). For *Eriocrania*, an increase in aggregation can be explained by higher wind speed (Kozlov, 2002) in industrial barrens, which forces females of tiny moths to walk rather than to fly between the oviposition sites. However, this explanation is not valid for *P. strigulatella*, due to much lower habitat disturbance near the power plant in Apatity than in the industrial barrens around the Monchegorsk smelter. Instead, the latter may simply concentrate on longer, presumably more vigorous, shoots, because the preference for longer shoots increased towards the emission source (Figure 5.15).

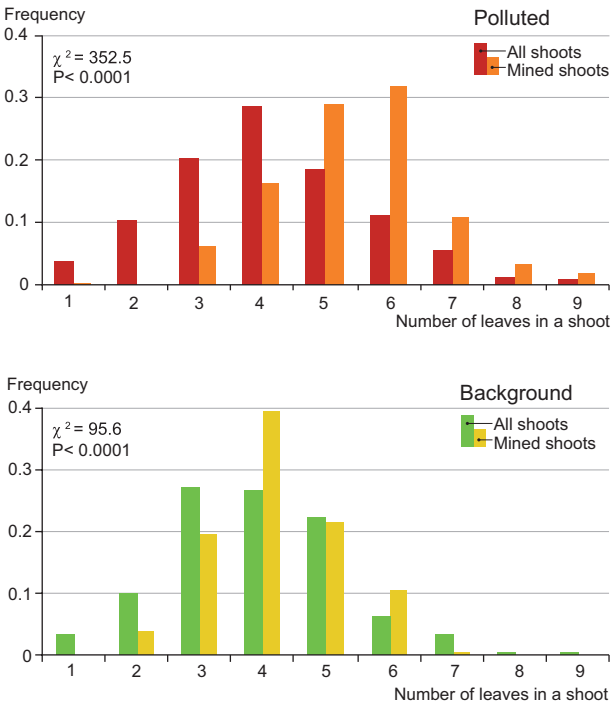


Figure 5.15. Distribution of *Phyllonorycter strigulatella* mines among shoots of its host, *Alnus incana*, with different number of leaves relative to distribution of shoots in a random sample in polluted and background sites around the power plant at Apatity. Sample sizes: polluted sites, all shoots: N=500, mine number: N=1250; background sites, all shoots: N=300, mine number: N=1000. Statistics (χ^2) reflect the difference between the distributions of all shoots and mined shoots (after Kozlov, 2003).

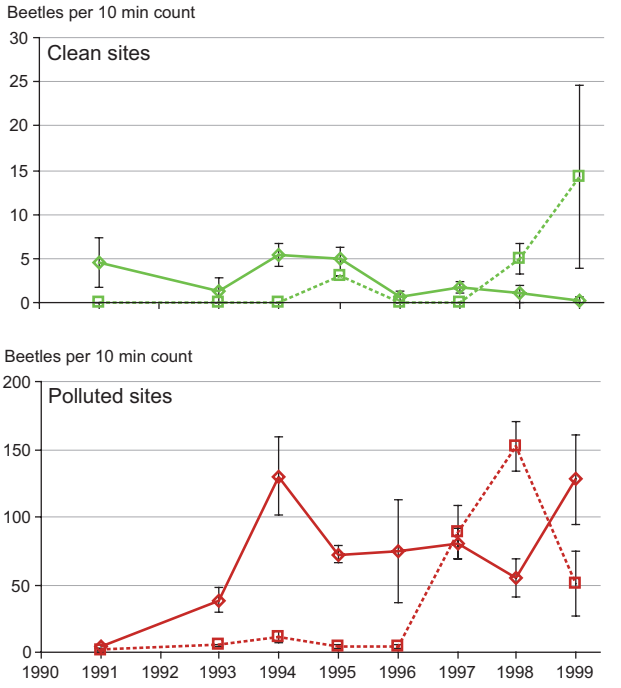


Figure 5.16. Population dynamics of Lapland leaf beetle (*Chrysomela lapponica*) at two relatively clean and two heavily polluted sites near the copper-nickel smelter in Monchegorsk. Density was assessed by the number of beetles found during 10 minute counts (conducted in three replicates at each site); bars indicate standard errors (after Zvereva *et al.*, 2002).

5.3.2.2. **Changes in population densities**

Some species clearly benefit from the impact of pollution. In particular, the willow feeding Lapland leaf beetle (*Chrysomela lapponica*) is generally infrequent in subarctic forests; however, outbreaks of this species, sometimes resulting in complete defoliation of willow bushes, have been detected in the vicinity of Monchegorsk and Nikel (nickel-copper smelters), and Vorkuta (a cement factory and a power plant) (Figure 5.16) (Zvereva *et al.*, 1997b, 2002; Virtanen *et al.*, 2002). Long-term studies on the Lapland leaf beetle suggest that pollution favors this species in a number of ways. First, the abundance of *Salix borealis*, which is the most favorable host plant of the leaf beetle, is much higher in the willow- and birch-dominated secondary communities around the Monchegorsk smelter than in virgin coniferous forests located at considerable distances from Monchegorsk (Zvereva *et al.*, 1995a,b). This may be especially important in relation to the findings concerning a decrease in feeding niche breadth with increasing pollution levels: near the smelter, beetles concentrated on *S. borealis*, while in unpolluted habitats they used other willow species as well (Zvereva *et al.*, 1995b). Second, the death of large trees in polluted habitats makes the forest more open, well illuminated, and with higher summer temperatures (Kozlov and Haukioja, 1997); these microclimate changes may be beneficial for many insects in northern areas. In particular, larval survival of *C. lapponica* increased with an increase in mean June temperature (Zvereva, 1999). Third, the impact of moderate concentrations of SO_2 improves host willow quality for *C. lapponica* (Kozlov *et al.*, 1996c), an effect that seems to be general for herbivores (Hughes, 1988; Riemer and Whittaker, 1989). Fourth, the overall impact of pollution creates enemy-free space for *C. lapponica* (Zvereva and Kozlov, 2000) by decreasing the abundance of some predators, especially syrphid flies, birds, and ants; in contrast, fly parasitoids are not sensitive to pollution (Figure 5.17). Thus, it seems that the release from predation pressure allows leaf beetles to increase their abundance, despite a decrease in individual performance and to utilize high-quality host plants up until the point where overgrazing causes a deterioration in host quality (termed Delayed Inducible Resistance), with a subsequent crash in the outbreak (Zvereva *et al.*, 1997a).

Pollution may interact with the natural density fluctuations of some insects and, in these situations, the effects of pollution become visible only in high-density years. This is the case with a tiny moth, *Phyllonorycter strigulatella*, the larvae of which develop in the leaves of *Alnus incana*. This was monitored around a coal-fired power plant (emitting 11 000 to 29 000 tonnes of SO_2 each year) near Apatity during 1991 to 2001. The periodicity in density fluctuation was

not affected by pollution; peak densities of the leafminer were observed at both polluted and unpolluted sites in 1993 and 1999 (Figure 5.18). Densities of *P. strigulatella* showed no correlation with pollution levels between outbreaks, but increased strongly near the power plant during outbreaks. At polluted sites the density increased by a factor of 15 to 20, whereas at unpolluted sites it increased by a factor of 3 to 4, relative to the density between outbreaks (Kozlov, 2003).

Increase in density with increasing proximity to the Monchegorsk smelter complex has also been reported in leafmining moths (*Eriocrania* spp.) feeding on birches, in the moth *Argyresthia pygmaeella* the larvae of which live in willow buds (Kozlov, 1997), and in saprophagous flies (families Calliphoridae, Scatophagidae, Calobatidae, Otitidae, Sepsidae) (Zvereva, 1993a,b). Around Nikel, a density increase was found for the autumnal moth (Ruohomäki *et al*., 1996).

The opposite – a decline in density near the pollution source – has been reported more frequently than a density increase; however, this may have resulted from the strong publication bias against the reporting of 'positive' results. This trend seems most common for soil mesofauna around all Kola Peninsula polluters (Koneva, 1993; Zenkova, 1999; Zenkova and Zainagutdinova, 2002). In particular, the density of spiders in the most polluted sites was around 10% of that observed in unpolluted forests around Monchegorsk (Gilyazova, 1993) and Nikel (Koneva and Koponen, 1993). Abundance of larvae of two families of soil-inhabiting beetles – click beetles (Elateridae) and soldier beetles (Cantharidae) – near the Kandalaksha aluminum smelter decreased by a factor of 10 to 20 (Zenkova and Zainagutdinova, 2002). The most pronounced decline – to 0.43% of the background density – was reported for soil mites near Monchegorsk (Zenkova, 1999). Ants also decline near Monchegorsk, as revealed by direct censuses (Gilyazova, 1993) and estimates of their predatory activity against leaf beetle larvae (Zvereva and Kozlov, 2000). Among herbivores, this pattern was evident in the birch-feeding leafroller (*Eulia ministrana*) (Kozlov, 1997) and several noctuid moths (*Xestia rhaetica, X. speciosa, Eurois occultus*) (Kozlov *et al*., 1996a). A decrease in the abundance of the autumnal moth was reported near Monchegorsk (Ruohomäki *et al*., 1996); this, however, contradicts the density increase of the autumnal moth reported near Nikel (Ruohomäki *et al*., 1996). Catches of human-biting mosquitoes (Culicidae) and black flies (Simuliidae) decreased near the Monchegorsk smelter by a factor of 10 to 100, presumably due to the combined action of pollutant toxicity, pollution-induced forest degradation, and a decline in vertebrate density (Kozlov *et al*., 2005b). Among other fly species, the density decline was most pronounced in predatory groups such as Empididae (Zvereva, 1993a,b).

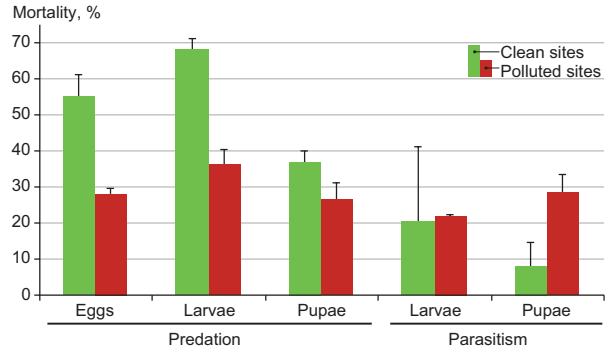


Figure 5.17. Mean mortality of different stages of Lapland leaf beetle (*Chrysomela lapponica*) due to predators and parasitoids in two relatively clean and two heavily polluted sites near the copper-nickel smelter at Monchegorsk. Bars indicate standard errors (after Zvereva and Kozlov, 2000).

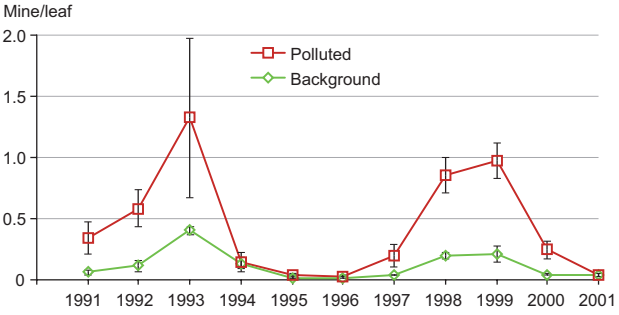


Figure 5.18. Density fluctuations for the leafmining moth *Phyllonorycter strigulatella* in polluted and background areas around the power plant at Apatity on the Kola Peninsula. Means are based on five (polluted sites) or four (background sites) site-specific values; bars indicate standard errors (after Kozlov, 2003).

Although detection of a non-linear (e.g., dome-shaped) response to pollution is more difficult than a linear response, this kind of density pattern has been found in several species, such as noctuid moths (*Acronicta auricoma, Hyppa rectilinea, Diarsia mendica, Xestia alpicola, Sympistis heliophila*), geometrid moths (*Rheumaptera subhastata, Ematurga atomaria*), and butterflies (*Clossiana euphrosyne, Vacciniina optilete*) (Kozlov *et al*., 1996a,b). The density of frit flies (Chloropidae) peaked at 7 to 17 km from Monchegorsk, being equally low in industrial barrens and in unpolluted forests (Zvereva, 1993b). Population density of the burying beetle (*Nicrophorus vespilloides*) determined by bait-trapping, peaked at 15 to 30 km from Monchegorsk. Since the abundance of vertebrates decreases towards the pollution source, this pattern indicates an increase in the beetle's food supply (carcasses of vertebrates), i.e. an increase in the mortality rates of vertebrates even in moderately polluted forest habitats (Kozlov *et al*., 2005a).

Finally, densities of some groups of invertebrates do not seem to change along pollution gradients. This pattern was detected for the willow-feeding leaf beetle (*Phratora vitellinae*) (Kozlov, 1997), springtails (Collembola), and soil-dwelling beetle larvae (Zenkova, 1999).

5.3.2.3. **Changes in species richness, diversity, and community structure**

Species richness of different taxa or ecological groups, including moths and butterflies, ants, flies (including human-biting flies), birch-feeding insects and some other insects, does not decline with decreasing distance to the smelter at Monchegorsk (Kozlov, 1996, 1997; Kozlov *et al*. 1996a,b, 2005b; Kozlov and Whitworth, 2002). Moreover, some rare or endangered species of moths and butterflies, such as *Sympistis zetterstedti* and *Clossiana freja*, have been recorded in severely polluted habitats (Kozlov, 1996). The occurrence on the Kola Peninsula of the lunar hornet clearwing (*Sesia bembeciformis*) which had been considered extinct in Finland for decades, is only associated with industrial barrens adjacent to the Monchegorsk smelter (Kozlov and Jalava, 1994). Interestingly, both species of parasitic fly that develop in larvae of the Lapland leaf beetle, *C. lapponica*, were considered extremely rare prior to the mass occurrence reported in the impact zone around the Monchegorsk smelter (Richter and Zvereva, 1996; Disney *et al*., 2001).

In contrast, the diversity (measured by the Shannon-Weaver index) of several groups of insect, for example moths and butterflies, or all birch-feeding insects, was reported to decrease with increasing levels of pollution, while the diversity of other groups, such as flies, did not change (Kozlov, 1997; Kozlov and Whitworth, 2002; Kozlov *et al.*, 2005b). These data suggest that acidification modifies insect communities primarily via changes in abundance, rather than through selective removal of some species.

The few studies that have been undertaken on soil mesofauna suggest that several taxa, such as earthworms (Lumbricidae) and millipedes (Myriapoda), may have completely vanished in severely polluted habitats (Koneva, 1993; Gilyazova, 1993; Zenkova, 1999). Only two species of spider, *Steatoda phalerata* and *Agyneta gulosa*, were caught in the industrial barren 2.5 km from Monchegorsk, compared to 18 species collected in forests 20 and 30 km from Monchegorsk (Koponen, 2005). However, the conclusion concerning an overall decline in the taxonomic diversity of soil mesofauna (Zenkova, 1999) near the Monchegorsk smelter must be viewed as tentative, due to deficient sampling design and because the diversity data were not corrected for sample size.

5.3.2.4. **Concluding comments on invertebrates**

Much circumstantial evidence suggests that acidic precipitation is responsible for a decline in populations of some insects in Europe and North America (Heliövaara and Väisänen, 1993). However, densities of many insects significantly increased near point polluters, thus pollution impacts on invertebrates can not be seen as purely adverse. Although the causal reasons behind the reported changes in insect populations are far from clear, the effect is more likely to be linked to changes in host plant abundance and physiology, or with changes in habitat structure, rather than to toxicity (Kozlov, 1997). Data collected in heavily contaminated industrial barrens show that these habitats are relatively rich in insect fauna and can even serve as refugia for some rare and endangered species.

5.4. Critical loads of acidity and their exceedance

Several protocols under the Convention on Long-range Transboundary Air Pollution (LRTAP) require signatories to reduce sulfur and nitrogen emissions, the latest being the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone which was signed in Gothenburg in 1999 and entered into force in May 2005. This is an effects-based protocol, meaning that ecosystem vulnerabilities were used in negotiations of emission reduction targets. The vulnerability of ecosystems to the deposition of sulfur and nitrogen is quantified by critical loads, 'the quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge' (Nilsson and Grennfelt, 1988). Under the auspices of the LRTAP Convention, critical loads are calculated at national focal centers of many European countries following agreed methodologies (UBA, 2004). The data are collected, vetted, and collated by the Coordination Centre for Effects, which produces European maps and makes the data available for integrated assessments, for example in support of emission reduction negotiations (Hettelingh *et al.*, 2001).

Parties to the LRTAP Convention mostly compute critical loads for terrestrial ecosystems (forests and semi-natural vegetation) following agreed procedures laid down in the Mapping Manual (UBA, 2004). Although the LRTAP Convention covers the region of the UN Economic Commission for Europe (UNECE) – i.e. Europe, the area of the former Soviet Union, the United States, and Canada – critical loads data for North America have not been available to date. Only recently a level-0 approach has been used to compute critical loads for terrestrial ecosystems in Canada.

The resulting critical load map for Canada represents the long-term buffering capacity (or weathering rate) in the top 60 cm (at most) of soil (Aherne and Watmough, 2005). The principal data set underlying the current map is the Soil Landscapes of Canada (SLC; scale of 1:1,000,0000) database. The SLC map (and associated databases) was produced by generalizing detailed soil survey data. The level of mapping was designed to be used for broad, regional-scale assessments. Using semi-quantitative methods, soil buffering capacities (derived from percent clay and substrate type) have been allocated to soil types. Buffering capacities were vertically and spatially weighted for each soil type to derive a single average value for each mapping unit. This level-0 approach does not yet consider vegetation uptake, atmospheric inputs, or soil leaching, and represents an initial attempt at mapping critical loads for Canada. Figure 5.19 shows the acidity critical loads for terrestrial ecosystems above 60° N, both for Canada and northern Europe (note: terrestrial critical loads for Alaska and Greenland have not been mapped).

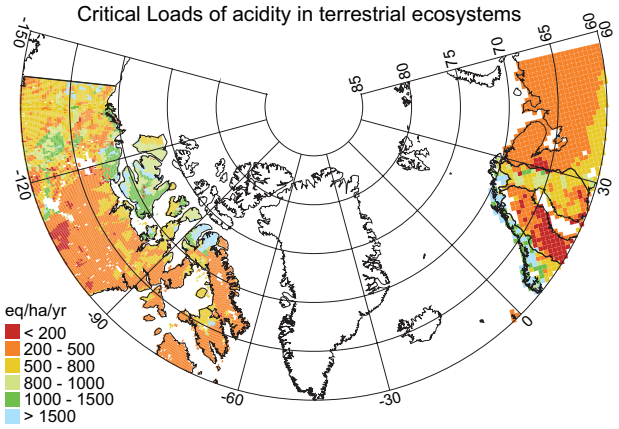


Figure 5.19. Critical loads of acidity for terrestrial ecosystems in northern Europe and Canada north of 60° N.

Exceedances of critical loads were calculated by combining the critical load maps with modeled deposition data, using the hemispheric Eulerian model DEHM. Three emission/deposition scenarios were used for the exceedance calculations (see section 2.3 for details): (i) 1990 emissions, (ii) the Current Legislation (CLE) scenario for 2010 (the CLE scenario reflects the current perspectives of individual countries on future economic development and takes into account the effects of presently agreed emission control legislation in the individual countries), and (iii) the Maximum technically Feasible Reduction (MFR) scenario for 2020 (the MFR scenario projects the scope for emission reductions offered by full implementation of presently available emis-

sion control technologies, while maintaining the projected levels of anthropogenic activities).

Estimated exceedances of the critical loads for the three scenarios are shown in Figure 5.20. The results indicate that for Canada there is no exceedance (north of 60° N) under the three deposition scenarios (i.e., not even in 1990). Although critical loads are comparable in Canada and northern Europe, deposition of sulfur and nitrogen is much smaller in Canada. The minimum critical load is about 84 eq/ha/yr and the maximum rates of sulfur and nitrogen deposition are about 30 to 40 eq/ha/yr each (not in the same place). Thus not even the combined sulfur and nitrogen deposition exceeds any critical load in this region.

In northern Europe large regions show critical load exceedances for 1990 (Figure 5.20). However, this area is much reduced assuming implementation of currently agreed emission reduction measures (CLE 2010), and would almost completely disappear assuming the MFR 2020 scenario.

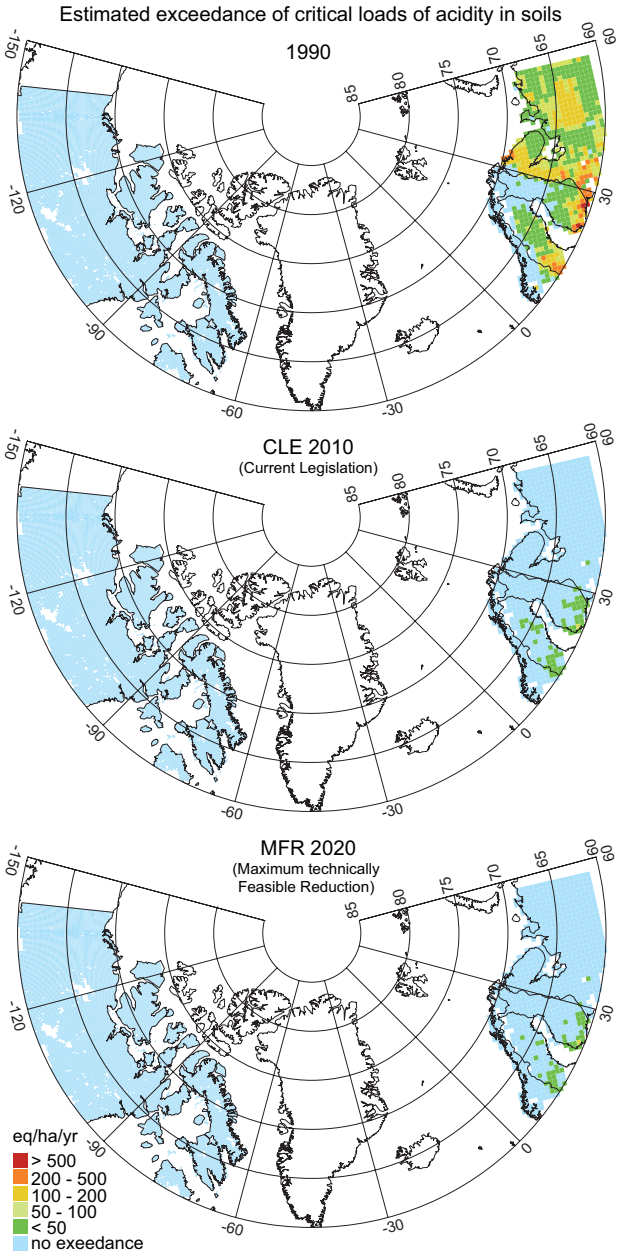


Figure 5.20. Estimated exceedance of critical loads of acidity for soils for three emission/deposition scenarios: 1990 emissions, the CLE scenario for 2010 (CLE, 2010), and the MFR scenario for 2020 (MFR, 2020).

Chapter 6
Effects on Freshwater Ecosystems

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6.1. Evidence from water quality monitoring

The previous AMAP assessment of acidification of surface water in the Arctic focused on northern Fennoscandia and the Kola Peninsula (AMAP, 1998). There were no reports of acidified lakes and rivers in Arctic Canada and Alaska. Although lakes in some areas of North America were reported to be acid sensitive there were no indications of acidification.

The geographical extent of surface water acidification was based on national lake surveys in Finland, Norway, and Sweden between 1986 and 1990 (Henriksen *et al*., 1992). These surveys showed that the impacts from high levels of sulfur deposition were mostly limited to a distance of about 50 km from the large pollution sources. Nitrate concentrations were very low over the whole area, due to very low nitrogen deposition.

The border areas between Norway, Finland, and Russia were reported to be heavily polluted. Studies of lakes in the eastern parts of Finnmark in 1986 (Traaen, 1987) showed that the sulfate concentration had more than doubled since 1966 and was at the same stage of acidification as the most acidified lakes in southern Norway. Numerous small mountain lakes in the area were chronically acidic (pH<5). Even large lakes had little of their original buffering capacity left. Some small lakes, in particular at Jarfjordfjellet, were too acidic to support fish. Studies in 1987 to 1989 (Traaen, 1991) indicated that large areas in Sør-Varanger would become increasingly damaged, with a loss of fish stocks, if the acid deposition increased further.

On the Kola Peninsula acidified lakes were reported around the industrial centers, and along the northern and eastern parts of the peninsula. However, at Kola, pollution from nickel and copper was a bigger problem in lakes than acidification (Moiseenko *et al*., 1995).

An extensive inventory of small lakes in northern Finland between 1987 and 1991 showed that acidic and poorly buffered lakes were widely found in northeastern Lapland near the Norwegian and Russian borders. Sulfate concentrations here were generally higher than in other parts of Lapland (Kähkönen, 1996). A survey of small mountain lakes and brooks in northeastern Lapland in 1991 to 1993 found that the alkalinity of surface waters was lowest (mostly <50 µeq/L) in the Vätsäri area 40 to 50 km west of the Nikel smelter (Lappalainen *et al*., 1995).

Areas sensitive to acidification and areas possibly acidified by acid deposition were quantified by calculating the critical loads for acidification of surface waters (see Box). Sulfur was regarded as the only acidifying agent as nitrate concentrations in lakes and streams in this area were very low. Critical loads for northern Fennoscandia, the Kola Peninsula, and the Spitsbergen Archipelago (Henriksen *et al*., 1994; Lien *et al*., 1995) were very variable, ranging from <300 to >1300 mg S/m^2/yr. Most areas of Finland

Definitions, terms, and calculations

Water chemistry

The chemical composition of surface waters in an undisturbed ecosystem is mainly determined by the contribution of ions from weathering and ion exchange in the catchment and from atmospheric deposition. Atmospheric deposition is influenced by sea salts, soil dust, and long-range transported air pollutants. The chemical composition of precipitation depends on distance from the sea and anthropogenic pollution sources. It is modified by a number of processes when passing through a catchment. Such processes are biological (microbial activity, uptake by plants, release of ions through decomposition, etc.) and chemical (weathering, ion exchange, adsorption and desorption, redox processes, precipitation etc.). In general, bedrock composed of gneisses and granitic gneisses has low weathering rates, yielding waters with a low content of base cations and bicarbonate that consequently have a low buffering capacity toward acidification. Bedrock with a more basic character such as gabbro, greenstone, and schist has higher weathering rates and releases more base cations and bicarbonate, yielding surface waters with a higher buffering capacity. Biological processes usually result in modification of ion composition or removal of ions, while weathering gives a net contribution. The total sum of ions from deposition and weathering together with all the various processes occurring within a catchment determine the chemistry of the runoff water. Additional, but often similar, in-lake processes can further modify the ion composition.

Acidification and naturally acidified lakes

The pH of most natural, mineral-bearing waters falls within the range 6 to 9. pH 6 is a threshold value below which lake biology is detrimentally affected by acidification (Baker et al., 1990; Doka et al., 2003; Holt and Yan, 2003). pH in a lake is affected by several natural processes within the catchment, such as the rate of weathering and production of bicarbonate, mobilization and leaching of organic acids derived from humic substances (organic anions, typically indicated by dissolved organic carbon/ total organic carbon levels), and input of sulfate from natural sulfide-bearing minerals. Low weathering rates, high input of organic anions, and high input of natural sulfate help to depress pH in the lake or river. Large inputs of anthropogenically derived sulfur and nitrogen that exceed the critical loads for the lake can also depress the pH to biologically harmful levels. It is not obvious, by measuring pH only, if a low pH value in a water body is due to anthropogenic or natural processes.

Alkalinity

Alkalinity is a measure of the buffering capacity of water, or the capacity of bases to neutralize acids. Alkalinity is a measure of the water's ability to resist change in pH and to neutralize

acid inputs. Alkalinity is a more integrative indicator of lake acidification than pH. The most important buffering materials in natural waters in the Arctic are primarily bicarbonate (HCO_3^-) and organic acids. Waters with low alkalinity (<20 µeq/L) are very susceptible to changes in pH. Waters with high alkalinity (>200 µeq/L) are able to resist major shifts in pH. As increasing amounts of acid are added to a water body, the buffering capacity of the water is consumed, and the pH of the water decreases (acidification). At pH 5.5, only very weak buffering ability remains, and at pH levels below 4.5 there is no alkalinity left. Alkalinity is measured by titration. An acid of known strength (the titrant) is added to a volume of a treated sample of water. The volume of acid required to bring the sample to a specific pH level reflects the alkalinity of the sample. Alkalinity can be measured in the laboratory in many different ways, making it difficult to compare results from different investigations.

Acid neutralizing capacity

Calculated Acid Neutralizing Capacity (ANC) is an equivalent to measured alkalinity. ANC is an even more integrative and robust parameter than alkalinity in establishing good dose/response relationships between water chemistry and damage to the biological community. ANC is the parameter used as the critical chemical criterion for sensitive indicator organisms in surface waters within the international critical loads work. ANC is defined as (Reuss and Johnson, 1986): equivalent sum of base cations minus the equivalent sum of strong acid anions $ANC = ([Ca^{2+}] + [Mg^{2+}] + [Na^+] + [K^+] + [NH_4^+]) - ([Cl^-] + [SO_4^{2-}] + [NO_3^-])$*. Waters with low ANC (<50 µeq/L) indicate possible damage to biota.*

Biologically relevant chemistry

The ultimate goal of emissions control programs is biological recovery, or the return of sensitive species that have been eliminated during the course of acidification. An assessment of biologically-relevant chemical trends can only suggest that biological recovery is possible (or expected), not that it has occurred. When surface water trends are shown to be moving in the correct direction (e.g., decreases in sulfate, or increases in pH), they indicate improvement in the acid–base chemistry of lakes and streams. It is important to note that these improvements do not necessarily equate to recovery. The term 'recovery' implies that the chemistry has returned to some pre-acidified status, such as pre-industrial levels of sulfate or alkalinity; trends indicate only that surface waters are moving toward this recovered status, not that they have reached it. In the absence of good data on biological recovery, it is common to assume that biological recovery will eventually occur, after a sufficient time lag, when key chemical variables have recovered their pre-acidification levels. These key chemical variables are those that have direct toxic effects on biota (primarily hydrogen ion and aluminum) and those that ameliorate some of the toxic effects (primarily base cations like calcium). For these reasons, evaluations of chemical recovery are often focused on acidity (pH and alkalinity), aluminum, and base cations (calcium).

Non-marine concentrations

All calculations and presentations of sulfate and base cations (sum of Ca + Mg) in this work are non-marine fractions, i.e. the seasalt contributions have been subtracted from the total levels measured. For sulfate, what is left represents the natural background input from weathering (which is normally very low) and the anthropogenic contribution from deposition. For base cations the remaining fraction is derived from weathering. Non-marine fractions (denoted by an asterisk) of sulfate and base cations* in lake and river water are calculated (see below) under the assumptions that all chloride (Cl) is of marine origin (cyclic sea salts) and is accompanied by other ions in the same proportions as in seawater. Base cations* are in this assessment taken as the sum of non-marine Ca + Mg. All units are in µeq/L.*

$[Ca^*]$	=	$[Ca]$	-	0.037	·	$[Cl]$
$[Mg^*]$	=	$[Mg]$	-	0.198	·	$[Cl]$
$[SO_4^*]$	=	$[SO_4]$	-	0.103	·	$[Cl]$

Acid sensitive lakes and critical loads

Lake water chemistry gives information on sensitivity to acidification. An extremely sensitive lake typically has an alkalinity of <20 µeq/L, and a less sensitive lake from 20 to 50 µeq/L. A lake with an alkalinity of >200 µeq/L is considered to be insensitive to acidification. The concentration of base cations can also give an indication of the acid sensitivity of the water, as the base cation concentrations directly reflect the weathering rate and bicarbonate production rate within the catchment. Surface water with low concentrations of base cations (BC <100 µeq/L) indicates sensitivity to acidic atmospheric inputs. Concentrations of base cations from 100 to 400 µeq/L indicate moderate sensitivity and values >400 µeq/L indicate general insensitivity.*

To evaluate anthropogenic acidification of lakes, the water chemistry must be evaluated together with deposition input. This is done by calculating the critical load of acidity (CL_{Ac}) and the exceedance of the critical load based on atmospheric inputs of sulfur and nitrogen. The critical load concept is a method of estimating ecosystem sensitivity to acidic inputs (i.e., sulfur and nitrogen), and was used to prepare the two protocols to the LRTAP Convention for reducing emissions of sulfur and nitrogen in Europe: the Oslo Protocol in 1994 (UNECE, 1994) and the Gothenburg Protocol in 1999 (UNECE, 1999). Similarly, critical loads were used to guide the distribution of sulfur dioxide emissions reductions in southeastern Canada during the 1980s and 1990s (Jeffries, 1997). The CL_{Ac} is a property of the lake and its catchment and is primarily based on weathering rates in the catchment. Since weathering is a function of bedrock geology, the sensitivity to acidification of surface waters can also be determined from geological maps. Exceedance of critical loads compares the critical load with deposition – actual or expected. When the deposition is greater than the critical load the aquatic ecosystem is expected to become damaged.

The geographical extent of surface water sensitivity to acidification in the AMAP region can also be determined from a map prepared by the Stockholm Environmental Institute based on soil type, land cover, and soil moisture. This is mainly assumed to hold for assessment of soil sensitivity, but will in general also hold for surface water sensitivity.

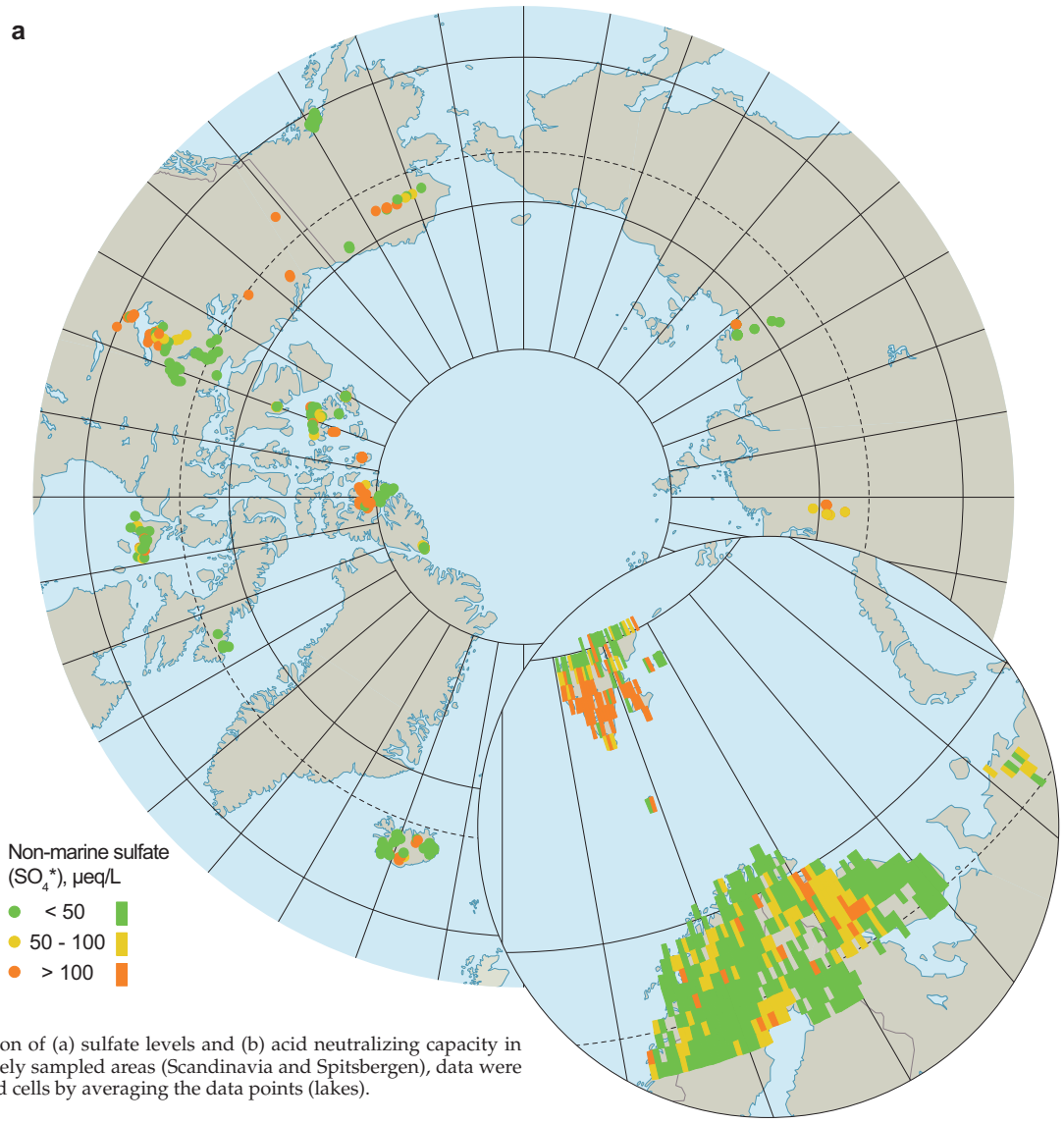


Figure 6.1. Distribution of (a) sulfate levels and (b) acid neutralizing capacity in arctic lakes. For densely sampled areas (Scandinavia and Spitsbergen), data were summarized into grid cells by averaging the data points (lakes).

and Norway near the smelters were quite sensitive, and critical loads for the Kola Peninsula were low. Exceedance of critical loads (based on 1990 sulfur deposition), which indicates possible surface water acidification, occurred in 70% of the county of Sør-Varanger in Norway. Critical loads were exceeded in 48% of lakes on the Kola Peninsula (Moiseenko, 1994). Small exceedances (based on 1990 sulfur deposition) were recorded in 5% of the ice-free area of Svalbard and Bear Island, but only in the northern parts (Lien *et al.*, 1995).

Long-term trends in surface water chemistry in Finland, Norway, Sweden, and on the Kola Peninsula, showed that between the mid-1980s and early 1990s, acidification had stabilized and may even have reduced slightly in some lakes. This was assumed to have resulted from decreased sulfur emissions in Europe. Some of the lakes and rivers had enough buffering capacity that they were not affected by high acid inputs.

6.1.1. **Current status**

The current status of acidification of surface waters in the Arctic has been determined from a compilation of regional and sub-regional lake surveys. Most were undertaken in northern Fennoscandia and the Kola Peninsula, but areas of Arctic Canada, northern Russia, Alaska, Iceland, and the Spitsbergen Archipelago were also covered. Since the previous AMAP assessment on acidification (AMAP, 1998) there has been one large lake survey in the arctic part of the Fennoscandian region. This was undertaken in 1995 and included the Kola Peninsula, the northern part of Fennoscandia, and Iceland (Henriksen *et al.*, 1997a,b; Skjelkvåle *et al.*, 2001c).

This assessment compiles data from several arctic regions. Sources of data and median values for key chemical variables for different arctic regions are summarized in Table 6.1. The geographical distribution of sulfate and acid neutralizing capacity (ANC) in arctic lakes is shown in Figure 6.1. It should be remembered that compiling data in this way means that the sample population is not a statistical subset of the overall lake population. Chemical data from 605 lakes within the Canadian Arctic were compiled for this assessment. Most of the data were collected in the 1990s (Table 6.1). The variables include: pH, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}), potassium (K^{+}), alkalinity, sulfate (SO_4^{2-}), chloride (Cl^{-}), nitrate (NO_3^{-}), plus dissolved organic carbon (DOC) and specific conductance. Some datasets contained dissolved inorganic carbon (DIC) values rather than alkalinity, and in such cases, the latter was estimated by assuming that the DIC exists entirely as bicarbonate (HCO_3^{-}).

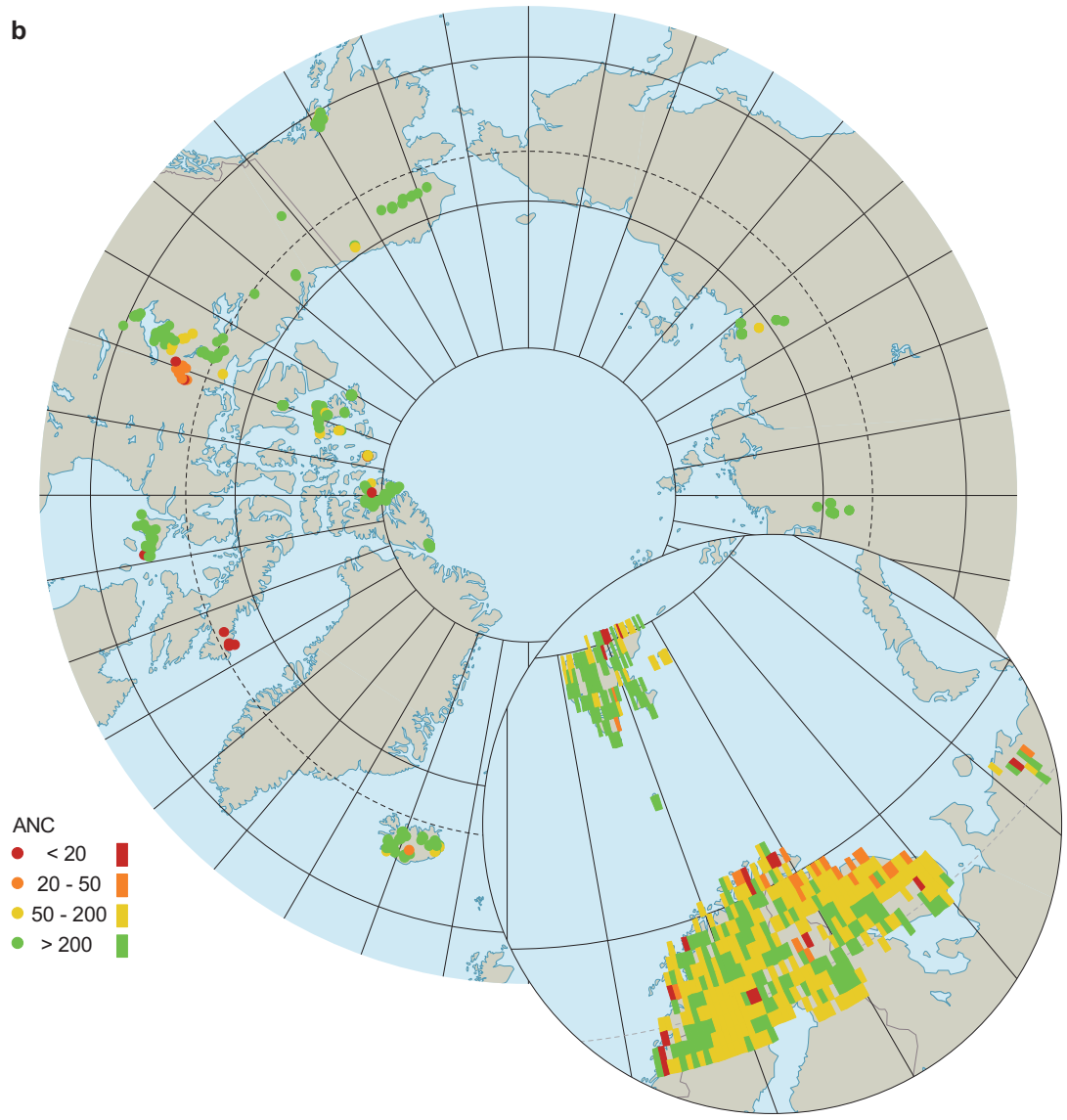


Table 6.1. Median values for a selection of key chemical variables including the calculated critical load of acidity CL_{Ac} and exceedance.

		n	% of total lake population	pH	BC*, µeq/L	Alkalinity, µeq/L	ANC, µeq/L	SO_4*, µeq/L	DOC, mg C/L	NO_3, µeq/L	CL_{Ac}, meq/m²/yr	% of area exceeded by sulfur deposition
Northern Russia [a]	Taymir area	23		7.97	490		445	80	4.0			
Northern Russia [a]	Pechora River basin	29		6.75	196		195	53	7.7			
Northern Russia [a]	Lena River basin	31		7.40	393		312	37	4.0			
Northern Russia [b]	Kola Peninsula	460	2.3	6.45	172	79	144	36	7.6	0.1	66	14
Finland [b]	Lapland	184	2.1	6.81	196	116	175	35	5.3	0.2	64	8
Norway [b]	Nordland, Troms and Finnmark	205	1.4	6.80	145	88	106	27	1.4	0.2	101	12
Sweden [b]	Norbotten	641	3.5	6.84	188	114	165	30	4.0	0.4	64	3
Iceland [c]		39	2.1	7.30	369	441	477	10	1.0	>0.1	546	0
Svalbard and Bear Island [c, d]		167	30	7.54	541	492	429	82	0.5	1.9	193	5
US, Alaska [e]	Kenai	59		6.88	101	96	144	3	6.4	0		
US, Alaska [f]	CABAL	22		7.79	457	459	448	89	4.7	0		
Canada [g]	Yukon (YK)	96		8.09	972	692	756	55	5.6	0.6		
Canada [g]	Northwest Territories (NT)	167		7.89	1866	1525	1519	83	3.0	0.7		
Canada [g]	Nunavut (NU)	3		7.90	1577	1200	1098	275	10.6	0.8		

CL_{Ac} is calculated by using the catchment dependent ANC_{limit} (Henriksen and Posch, 2001) and the Norwegian background sulfate concentration for all countries, except Sweden (Wilander, 1994). Exceedance CL is calculated with S-deposition from different sources, for each country year of deposition and name of the institute providing the deposition numbers are given: Norway (1990, NILU Norwegian Institute for Air Research), Sweden (1994, SMHI Swedish Meteorological and Hydrological Institute), Finland (1990, SYKE), Kola and Iceland (1992, EMEP European Monitoring and Evaluation Programme).

Data sources: [a] Duff *et al.* (1998); [b] Henriksen *et al.* (1997b); [c] Skjelkvåle *et al.* (2001c); [d] Lien *et al.* (1995), [e] Newell and Mitch (1992), Eilers *et al.* (1993); [f] Allen-Gil *et al.* (1997) [g] Nahanni (NT), Tuktoyaktuk (NT), Yellowknife (NT) and Wager Bay (NU) from Environment Canada, unpub. data; region between Coronation Gulf and Great Slave Lake (both NT and NU) from Rühland and Smol (1998), Rühland *et al.* (2003); region between southern YK and Tuktoyaktuk (NT) from Pienitz *et al.* (1997a,b); Wood Buffalo National Park (NT) from Moser *et al.* (1998); Melville Island (both NT and NU) from Bronwyn Keatley (Queen's University, unpub. thesis data); Prince Patrick Island (NT), Ellef Ringnes Island (NU) and Ellesmere Island (NU) from Antoniades *et al.* (2003a,b); Victoria Island and Axel Heiberg Island (NU) from Michelutti *et al.* (2002a,b); Baffin Island (NU) from Joynt and Wolfe (2001); Southhampton Island (NU) from Mallory *et al.* (2006).

6.1.1.1. **Northern Fennoscandia and the Kola Peninsula**

The Euro-Arctic Barents region consists of two major geological provinces: the northern part of the Precambrian shield and the Caledonian fold belt toward the north and west. Within the framework of European geology, the Norwegian, Swedish, Finnish, and Kola Precambrian is a part of the Fennoscandian or Baltic Shield. The Caledonian fold belt includes Precambrian and Paleozoic sediments. The Precambrian rocks are dominated by gneisses and granitic gneisses with low weathering rates, while the Caledonian and the Archean Karelian province also include rocks of more basic character with higher weathering rates (Lidmar-Bergström and Näslund, 2005).

The results from the 1995 Nordic Lake Survey were reported by Henriksen *et al*. (1997a,b), Henriksen (1998), and Skjelkvåle *et al*. (2001c) and form the basis of this section.

Lakes with relatively high concentrations of base cations are found in areas with basic/mafic rocks and metasediments; south-central Kola, central Finnmark, and central Lapland as well as scattered areas in the Swedish mountains. Low base cation concentrations, giving the greatest sensitivity to acidification, are found scattered all over the area and are most abundant in areas with bedrock composed of granite and granitic gneisses, i.e., the northern part of Kola, Norwegian coastal areas, the northeastern and southern parts of Lapland, and the western part of Norrbotten county. Northern Norway had the highest percentage of lakes with low concentrations of base cations; in Norway 40% of lakes had <100 µeq/L, while Finnish Lapland had only 10%. In Norrbotten and Kola respectively, 21 and 26% of lakes were acid sensitive. On the other hand, Norway also had the highest number of lakes with very high concentrations of base cations (11% of lakes had >500 µeq/L compared to 4 to 6% in the other countries).

Low alkalinity lakes (<20 µeq/L) were most abundant in the northern parts of the Russian Kola. Here, the higher content of organic matter in lakes in the eastern parts and the higher anthropogenic sulfate content in the western parts lowers the alkalinity. In the northern parts of the Nordic countries both the geological conditions and the precipitation amounts determine the alkalinity; depending on the weathering rate of the minerals in the soil and dilution by precipitation, respectively. In some regions with a high proportion of peatland, low alkalinity values in lakes are largely attributable to organic acidity. In general, low alkalinity lakes are rare in the northern Nordic countries and are scattered throughout the region as a whole.

Sulfate is considered to be the major acidifier of surface waters. High sulfate concentrations were most common in the western part of the Kola Peninsula, particularly around the smelters in Nikel and Monchegorsk. More than 10% of the lakes on the Kola Peninsula had >100 µeq/L non-marine sulfate (SO_4*). High concentrations were also found scattered around the whole Euro-Arctic Barents region. Most of these lakes probably had a significant input of sulfate from geological sources, reflecting the diverse geology of the area. The eastern Kola Peninsula represented the largest area with consistently low (<20 µeq/L) sulfate levels, reflecting the low sulfur deposition in this area.

Nitrate concentrations in the lakes of the Euro-Arctic Barents region were generally very low. More than 75% of lakes had <1 µeq/L NO_3. It was therefore concluded that nitrogen deposition was an insignificant acidifier of these lakes.

pH values of <5.5 were most abundant in the northern part of the Russian Kola (19% of lakes). Russian Kola also had the highest frequency of lakes with pH levels of <6.0 (28% of lakes), followed by northern Norway (14%), Norrbotten (7%), and Lapland (7%). In general, the lakes of the Euro-Arctic Barents region are less acid than lakes further south in these countries.

There was a pronounced west to east gradient in total organic carbon (TOC) concentrations. The coastal areas of Norway usually had <2 mg C/L, while lakes with >8 mg C/L were abundant in Norrbotten, Lapland, and the eastern parts of the Kola Peninsula. The topographically flat areas with peatland accumulation on the eastern Kola Peninsula and around Bothnian Bay are reflected in high TOC levels. However, despite the high proportion of peatland areas in the north, TOC concentrations in lakes in northern regions were found to be lower than in the countries as a whole, due to the colder climate, longer soil frost period, and lower peat decomposition rate (Kortelainen, 1993). Low TOC values in mountain lakes in the border area between Norway and Sweden are due to less organic soils in the catchments and high precipitation leading to dilution.

The estimated critical loads of acidity were exceeded in all countries. The highest percentage of exceeded lakes occurred in the Russian Kola and Norway, while Sweden had the lowest percentage of exceeded lakes. The calculations were based on sulfur deposition for 1990 (Finland and Norway), 1992 (Kola), and 1994 (Sweden) (see Henriksen *et al*., 1997a for full references of source data). In Norrbotten, the mean exceedance for 47 NILU-grids (95 percentile) using the FAB model with respect to deposition in 1990, 1997, and 2010 (in accordance with the Gothenburg Protocol) was 117, 38, and 31 eq/ha/yr, respectively.

6.1.1.2. **Iceland**

Iceland was created by volcanic activity along the Mid-Atlantic Ridge during the last 20 million years. The volcanic rocks in Iceland are predominantly mafic. Owing to the vesicular nature of the lava with glassy crusts, they weather readily. The lake water chemistry clearly reflects this geology. Results from the 1995 lake survey (Skjelkvåle *et al*., 2001c) showed that Icelandic lakes generally had high concentrations of base cations and high values of ANC and pH. Sulfate and nitrate concentrations were in general very low, due to low anthropogenic deposition of sulfur and nitrogen. High sulfate lakes were found in the active geological zones. Owing to the basic conditions of Icelandic lakes, they are not sensitive to acidification.

Owing to the high critical loads and the low sulfur deposition in Iceland, critical loads were not exceeded in any of the lakes studied. Hence, acidification of lakes is not a problem in Iceland.

6.1.1.3. **Svalbard and Bear Island**

The sedimentary rocks at Svalbard and Bear Island have a diverse mineralogy that includes limestone, dolomite, gypsum/anhydrite and shales containing phosphatic nodules, and yields waters with highly variable ionic composition. The lakes were sampled in 1990 to 1992 (Lien *et al*., 1995; Skjelkvåle *et al*., 2001c). High sulfate lakes were related to gypsum/ anhydrite in the soils and bedrock. In the northern parts of Svalbard some lakes were very acid sensitive, and in this area the critical loads were exceeded given the 1990 sulfur deposition (Lien *et al*., 1995).

6.1.1.4. New critical loads and exceedance calculations for the Euro-Arctic Barents region

Critical load calculations are used for assessment and policy work under the Convention on Long-range Transboundary Air Pollution (LRTAP) and the European Union and so are regularly updated (see section 5.4). This assessment focuses on data for northern Europe submitted to the Coordination Centre for Effects in 2004 (Hettelingh *et al.*, 2004) and new data for 104 lakes in the Kola region. The data on individual lake catchments are aggregated to the 'EMEP 50x50 km grid', which is used under the LRTAP Convention for deposition and exceedance calculations. Figure 6.2 shows the 5 percentile of the critical load of acidity on this grid system. Also shown are the critical loads for surface waters in Svalbard, which were computed and documented by Lien *et al.* (1995).

Exceedance of critical loads was calculated by combining the critical load maps with modeled deposition data, using the hemispheric Eulerian model DEHM (see section 3.7.4). Three emission/deposition scenarios were used in the calculations (see section 2.3): (i) 1990 emissions, (ii) the 'Current Legislation' scenario for 2010 (CLE 2010), and (iii) the 'Maximum technically Feasible Reduction' scenario for 2020 (MFR 2020). The same scenarios were used for calculating critical load exceedance for soils (see section 5.4).

Estimated exceedance of critical loads for the surface waters in northern Europe are shown in Figure 6.3 for the three emissions scenarios. The results indicate that implementation of presently agreed emissions reductions will reduce both the area and magnitude of exceedance substantially. However, the results also show that there is still clear exceedance of critical loads for surface waters in parts of the Kola region even after implementation of maximum technically feasible emissions reductions.

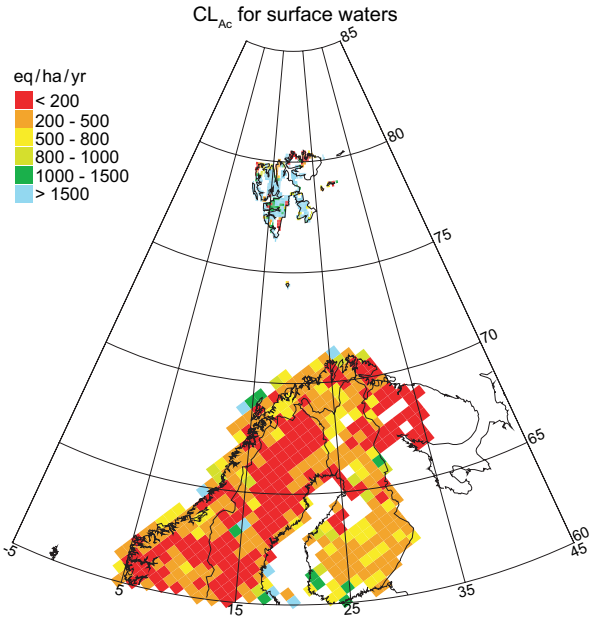


Figure 6.2. Critical loads of acidity for surface waters in northern Europe.

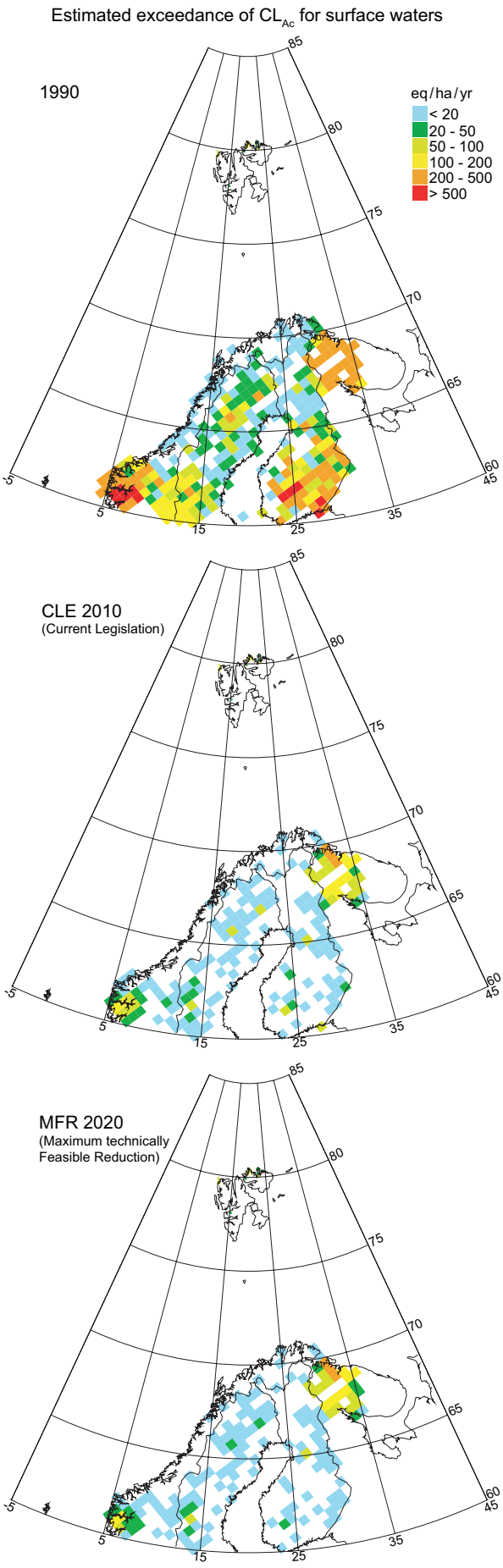


Figure 6.3. Estimated exceedance of critical loads of acidity for surface waters in northern Europe for three scenarios: 1990 emissions, Current Legislation for 2010 (CLE 2010), Maximum technically Feasible Reductions for 2020 (MFR 2020).

6.1.1.5. **Current status in Arctic Canada**

The area north of 60° N in Canada (the southern boundary of the AMAP domain) contains three politically defined territories: Nunavut, the Northwest Territories and the Yukon, plus the northernmost part of the province of Quebec. The area (more than 4 million km^2) comprises eight terrestrial ecozones spanning the boreal, taiga, arctic, and cordillera types that encompass the full spectrum of geological sensitivity to acidification. The largest expanses of acid-sensitive terrain (defined by a combination of bedrock and surficial geology) occur on Baffin Island and the continental mainland west of Hudson Bay. Most of the islands of the Arctic Archipelago have carbonate geology and so are not acid-insensitive.

Owing to the low solubility of the soils and bedrock characteristic of acid-sensitive terrain, associated surface waters are typically low in dissolved minerals and so the concentration of base cations indicates aquatic sensitivity. Fifteen percent of lakes investigated had base cation levels of <100 µeq/L, 20% of 100 to 400 µeq/L, and 65% of >400 µeq/L. The only extremely acid-sensitive lakes occur on Baffin Island and the central mainland straddling the border of Nunavut and the Northwest Territories. It should be noted, however, that the sample population excluded a large proportion of the acid-sensitive terrain.

Sulfur dioxide has long been acknowledged as the main air pollutant acidifying Canadian lakes (Jeffries, 1995, 1997) and is ultimately deposited on aquatic and terrestrial landscapes as SO_4. Sulfate deposition in the Canadian Arctic is very low (~3 meq/m^2/yr) so if atmospheric deposition is the main source of SO_4 in lakes, then lake concentrations should also be uniformly low. Figure 6.1 shows that lake SO_4 concentrations in Canadian arctic lakes are in fact spatially variable indicating the presence of geological sources in some areas, particularly Axel Heiberg Island.

Dissolved organic carbon levels in artic lakes exhibit a broad range in concentration (Table 6.1). Approximately 25% of lakes in Arctic Canada had substantial levels of DOC (>10 mg/L), which may contribute to some acidification by naturally occurring organic acids.

Lakes having alkalinity concentrations of <200 µeq/L have traditionally been considered sensitive to acidic deposition and 26% of the samples fell into this category. The spatial distribution of low alkalinity lakes tends to confirm the sensitivity interpretation using base cations. Unfortunately, there are no alkalinity data for Baffin Island lakes.

Of the lakes sampled, 251 had sufficient data to permit a calculation of critical loads for acidity (CL_{Ac}) using the Steady-State Water Chemistry Model (Henriksen and Posch, 2001) with an acid neutralizing capacity threshold value (ANC_{limit}) of 40 µeq/L. The median CL_{Ac} was 127 meq/m^2/yr and 8% of lakes had values of ≤10 meq/m^2/yr. The spatial distribution of three CL_{Ac} classes confirmed that the sample lakes mostly likely to acidify occur on Baffin Island and the central mainland. Whether or not the Steady-State Water Chemistry Model should be applied in a desert environment (annual surface runoff for almost all lakes was estimated to be 150 mm or less) merits further consideration.

6.1.1.6. **Naturally acidic lakes in Arctic Canada**

Most of the Canadian lakes fell within the pH range 6 to 9, which is assumed as the normal range for most natural, mineral-bearing waters. Only 3% of sample lakes had a pH of <6. Furthermore, only three of 605 lakes were strongly acidic, having a pH of <5. Two of the three strongly acidic lakes are on Axel Heiberg Island (pH 3.6 and 3.8) and both had extremely high SO_4 concentrations (Michelutti *et al*., 2002a). Schiff *et al*. (1991) used isotopic techniques to identify the source of the acid in the most acidic of these lakes (Colour Lake). They showed that oxidation of pyrite in the surrounding basin and oxidation of ferrous iron from an anoxic zone of the lake following spring ice-off and water column overturn caused the observed low pH and high SO_4 concentrations. It is not clear whether the arctic setting of these lakes is an important determinant in this acidification process. Certainly, lakes and rivers in temperate climates that are affected by acid mine drainage become acidic through similar processes. In natural temperate settings, however, acid generation by sulfide mineral oxidation tends to be self-limiting because the iron oxide weathering product coats the mineral surface rendering it less reactive. The occurrence of a naturally extremely acidic lake such as Colour Lake (and probably the other Axel Heiberg lake) is quite rare. Given the widespread occurrence of sulfide and to a lesser extent sulfate minerals, it seems reasonable to assume that some and perhaps even many lakes have been affected by acid-generating oxidation processes. The best evidence of this is probably the SO_4 data rather than the pH data. The third extremely acidic lake in the Canadian sample population was on Baffin Island (pH 4.66), but there are no SO_4 data to confirm oxidation of local sulfide minerals as the acid source.

A unique ecosystem acidification occurs at the Smoking Hills along the sea shore of Cape Bathurst in the Northwest Territories (70°14′ N, 127°10′ W; Havas and Hutchinson, 1983). Bituminous shales appear to have been burning for thousands of years thereby producing ground-level acidic fumigations that strongly influence the local tundra. Ponds in the area (not included in the data compilation described above) typically have pH levels of >8, but those within the fumigation zone have been acidified, sometimes to a pH of <2. The ponds exhibit elevated metal concentrations (aluminum, iron, zinc, nickel, manganese, and cadmium) and biota that are characteristic of acidic environments elsewhere. Biota in adjacent non-acidified ponds are typical of other arctic environments.

6.1.1.7. **Alaska**

There are very few lake or stream chemistry data available from acid-sensitive parts of the Alaskan arctic. The Kenai Lakes Investigation Project (Newell and Mitch, 1992; Eilers *et al*., 1993) characterized the major ion chemistry of over 800 lakes on the Kenai Peninsula from a statistical survey of 59 lakes in 1988. The results showed two groups of lakes: those with an alkalinity of <300 µeq/L (78% of lakes) and those with an alkalinity of >700 µeq/L. Low-alkalinity lakes had significantly lower concentrations of base cations and silica and significantly higher average concentrations of DOC than high-alkalinity lakes. Despite widespread acidic soils and bog vegetation, and resulting high DOC levels, none of the lakes sampled were acidic (minimum alkalinity 20 µeq/L).

The Chemistry and Biology of Arctic Lakes (CABAL; Allen-Gil *et al*., 1997) project sampled 22 lakes on the North Slope of Alaska in 1992 (D. Landers, U.S. Environmental Protection Agency, unpubl. data). The results showed only two lakes with an alkalinity of <200 µeq/L (minimum 156 µeq/L); none were considered to be acidified.

6.1.1.8. **Northern Russia, Siberia**

Lakes were sampled in three regions of Siberia, northern Russia, in 1993 to 1995 (Duff *et al*., 1998). The lakes were situated in the Pechora River basin, in the Yenisey River basin on the Taymir Peninsula, and in the Lena River basin. Only the lakes in the Taymir region were located in the same area as the large emissions source at Norilsk, but these were still around 200 km away. All the lakes (with the exception of a few in the Pechora River basin) were small, dilute, and oligotrophic with a neutral to slightly alkaline pH. Forested lakes near the mining center of Norilsk had higher concentrations of the major ions and metals; sediments in these lakes also had elevated metal concentrations (Blais *et al*., 1999). However, all the sample lakes in this area had ANC levels of >200 µeq/L and so were well buffered towards acidification. Some acidic lakes in the Pechora River basin had very high levels of organic carbon or high sulfate concentrations, probably due to geological sources.

6.1.2. **Temporal trends**

Temporal trends in water chemistry can be identified using data from regular monitoring over many years or from a series of repeated lake surveys. This assessment is based on both types of data but is limited to the Euro-Arctic Barents region (i.e., the northern part of Finland, Norway, Sweden and the Kola Peninsula of Russia). Data for other areas are not available.

The Euro-Arctic Barents region is affected by long-range transboundary air pollution and by emissions from industrial centers on the Kola Peninsula. Although the pollution exhibits relatively large year-to-year variations, it has still been possible to identify a general decrease in sulfur deposition and consistently low nitrogen deposition (see section 3.3).

6.1.2.1. **Lakes in Finland, Norway, and Sweden**

Acidification has been monitored in Finnish lakes since 1990. In northern Finland, there are estimated to be around 2100 small (4-100 hectare) headwater or seepage lakes susceptible to acidification (Forsius *et al*., 2003). A subset of 35 small forest or mountain lakes has been monitored annually in Finnish Lapland. In Norway, 24 lakes have been monitored on an annual basis since the 1986 lake survey (Henriksen *et al*., 1988). The monitoring is focused on areas of eastern Finnmark near the Russian border. The monitoring network includes six lakes in the Jarfjord area and 11 lakes in the county of Sør-Varanger. In addition there are six lakes in the counties of Nordland and Troms (SFT, 2005). In Sweden, eight 'reference' lakes have been sampled three to four times each year (Wilander, 1998).

The data from these 60 lakes for 1990 to 2004 were compiled to assess surface water trends in non-marine sulfate (SO_4*), non-marine base cations (Ca+Mg)*, alkalinity, ANC, and pH. The lakes were sub-divided into three groups based on geographical location: (1) Lapland, Finland, (2) eastern Finnmark, Norway, (3) northern Norway and Sweden (Figure 6.4). Trends for each lake and each variable were analyzed using the non-parametric tests given by Hirsch *et al*. (1982). These are equivalent to a non-seasonal Mann-Kendall trend test and the Sen slope estimator (cf. Helsel and Hirsch 1995). Trend slopes for each region were calculated using the same test, but in this case each lake was treated as a 'season'. The results give the direction of the trend from the trend slope. The detected trends are monotonic, i.e., proceed in only one direction.

Non-marine sulfate

Most sites showed a significant decrease in sulfate between 1990 and 2004 (Figures 6.4 and 6.5, Tables 6.2 and 6.3). The exception was a single site in northern Sweden (Abiskojaure) in which concentrations increased due to sulfur-containing minerals within the drainage basin. Lakes in eastern Finnmark near the emissions sources showed the most pronounced decrease with a trend slope of -1.4 µeq/L/yr (Table 6.3) based on concentration levels of 78 µeq/L in 1990 to 55 µeq/L in 2004 (representing a 30% decrease). Lakes in Finnish Lapland showed a trend slope of about -1.0 µeq/L/yr based on concentrations of 32 µeq/L to 18 µeq/L (a 43% decrease). Lakes in southern and central Lapland are more affected by long-range transported air pollutants from the south and so have benefited from the decrease in total European sulfur emissions. Lakes in northern parts of Norway and Sweden showed very slight decreases with an overall trend slope of -0.4 µeq/L/yr (Table 6.3) based on concentrations of about 20 µeq/L to 13 µeq/L (a 30% decrease).

Non-marine base cations

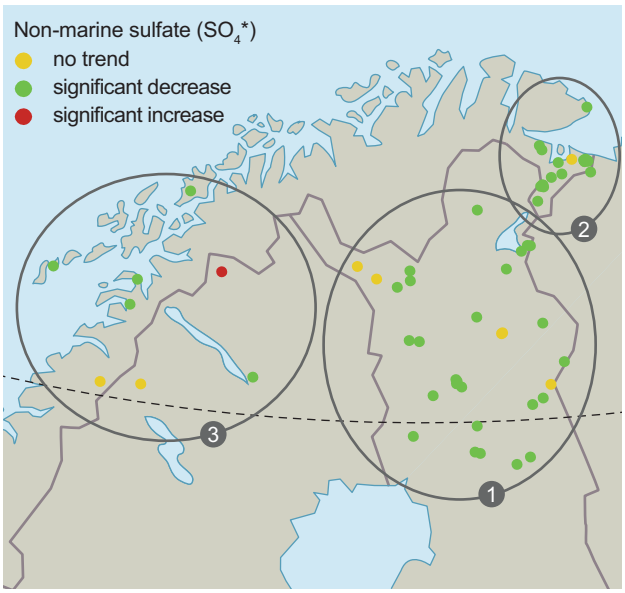
Base cations showed very small changes in concentration and the trends were both positive and negative (Figures 6.4 and 6.5, Table 6.3). Of the 60 lakes in this assessment, 25% had slightly significant increasing trends while 12% had significant decreasing trends. Trends for two of the three regions showed significant increases with average trend slopes of +0.5 µeq/L/yr for northern Norway and Sweden and +0.8 µeq/L/yr for Lapland. The increase in the first group was strongly dependant on the increase in a single lake (Lake Abiskojaure). Small changes and even increases in BC* concentrations compared to SO_4* con-

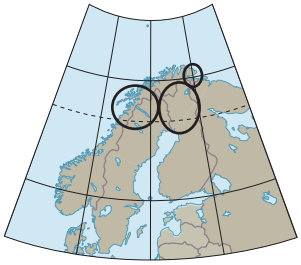
Table 6.2. Decrease in sulfate concentrations (µeq/L) between 1990 and 2004 for the three sub-regions. Values are calculated from linear regressions of average values.

	SO_4*, µeq/L		Change
	1990	2004	(%)
Lapland, Finland	32	18	-43
Eastern Finnmark, Norway	78	55	-30
Northern Norway and Sweden	20	13	-30

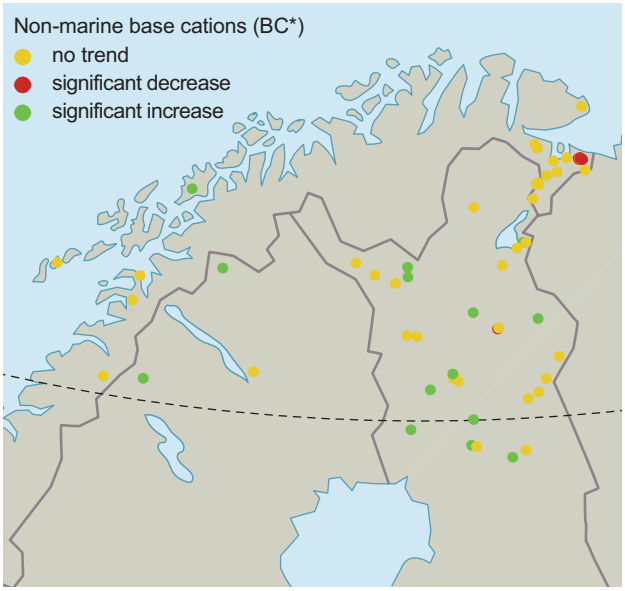
Table 6.3. Regional trends for 1990 to 2004. Values are median slopes with significant results (p<0.05) in bold. Units are µeq/L/yr.

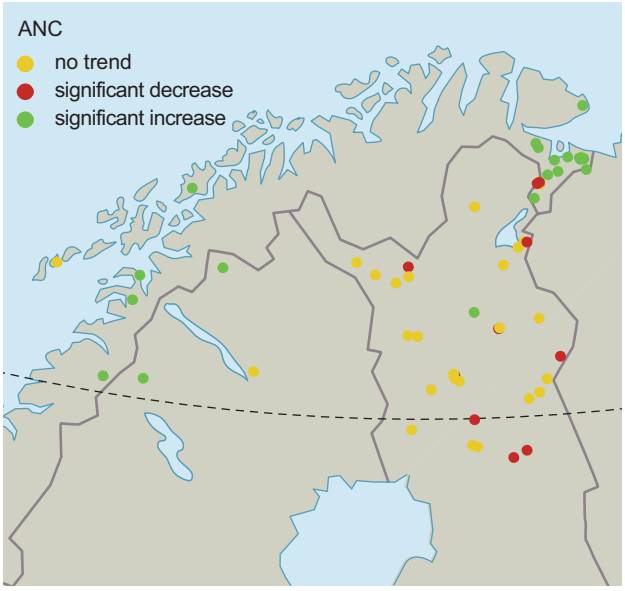
		Alkalinity		ANC		BC*		H^+		SO_4*	
	n	p	Theil slope	p	Theil slope	p	Theil slope	p	Theil slope	p	Theil slope
Lapland, Finland	35	**0.002**	**1.0**	**0.04**	**-0.3**	**0.006**	**0.8**	0.2	0.00	**0.0003**	**-1.0**
Eastern Finnmark, Norway	17	**0.004**	**0.3**	**0.0003**	**1.9**	0.05	-0.4	**0.0004**	**-0.04**	**0.0001**	**-1.4**
Northern Norway and Sweden	8	**0.007**	**0.9**	**0.001**	**1.6**	**0.007**	**0.5**	**0.002**	**-0.01**	**0.001**	**-0.4**

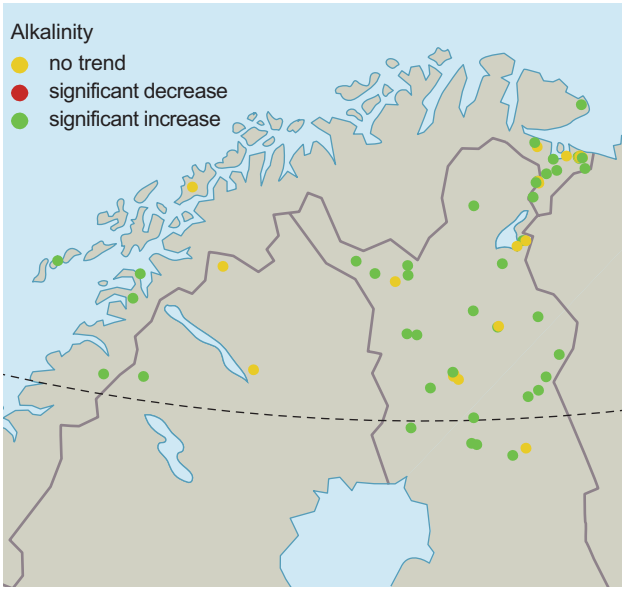




1 Lapland, Finland
2 Eastern Finnmark, Norway
3 Northern Norway and Sweden







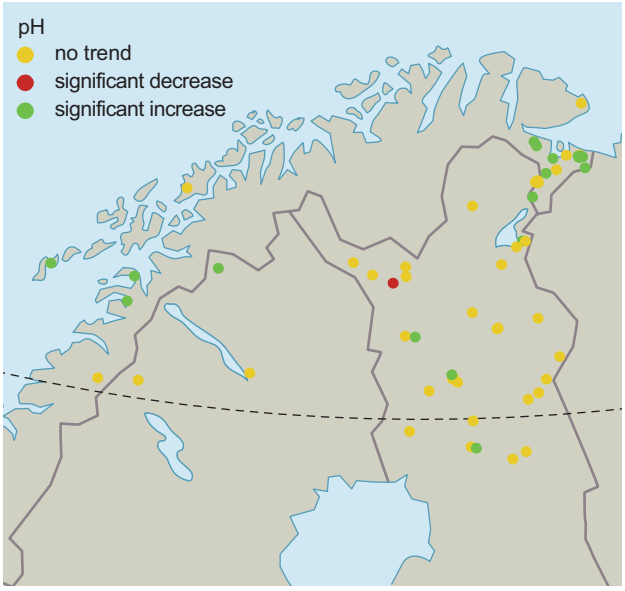


Figure 6.4. Trends in non-marine sulfate, non-marine base cations (Ca+Mg), alkalinity, acid neutralizing capacity, and pH across the Euro-Arctic Barents region for 1990 to 2004. Large circles denote the three sub-regions.

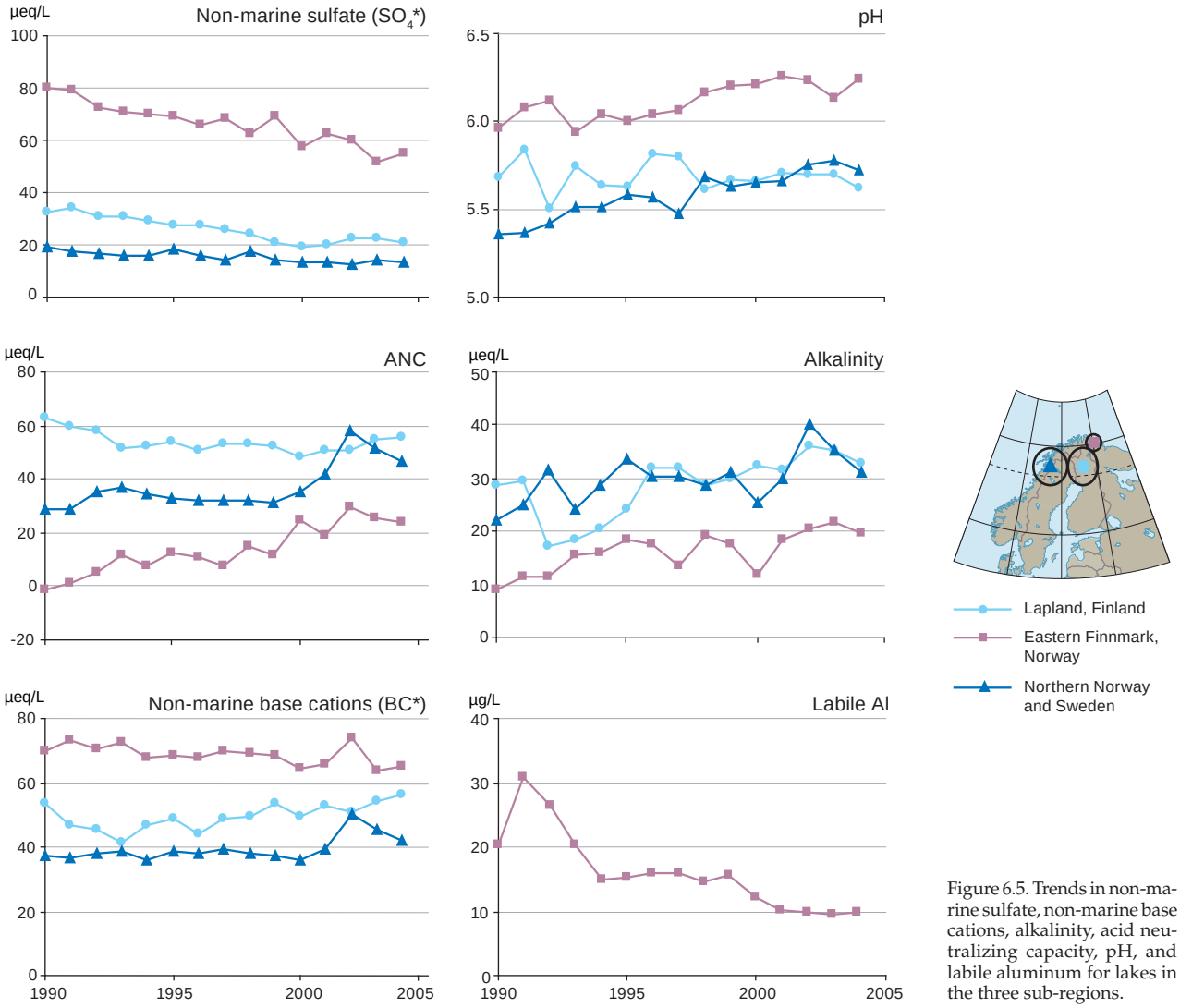


Figure 6.5. Trends in non-marine sulfate, non-marine base cations, alkalinity, acid neutralizing capacity, pH, and labile aluminum for lakes in the three sub-regions.

centrations enabled alkalinity to increase (see Figures 6.4 and 6.5). The median annual change in alkalinity ranged from +0.3 to +1.0 µeq/L/yr for the three regions (Table 6.3). The signs of chemical recovery are not reflected in the same way in calculated ANC (Figures 6.4 and 6.5). For Lapland, the median annual change in ANC was a decrease of -0.3 µeq/L/yr (Table 6.3). The small lakes in northern Finland are low ionic, dilute lakes (with BC* typically <100 µeq/L) that have had only modest impacts from air pollutants. Therefore, a high interannual variation in ion concentrations in these lakes reflects hydrological variations.

Alkalinity and pH

Increases in alkalinity are at an early stage and not really reflected in the pH values. In 37% of lakes, pH showed a significant increase (Figures 6.4 and 6.5, Table 6.3 (calculated as decrease in concentration of H^+)). This increase was most pronounced in eastern Finnmark. There is a large interannual variation in pH in lakes in eastern Lapland, due to variations in hydrology and natural organic acids. So a consistent and corresponding increase in pH cannot be expected. There is also pronounced seasonal variation, with the highest values in late summer and the lowest at the end of winter or at snowmelt.

Aluminum

There are few data on aluminum. In Finland, when sample pH was >6.2, the laboratories did not analyze aluminum. Moreover, at many sites inorganic bound aluminum was below the detection limit (<10 µg/L). The 1995 lake survey showed that a median value for lakes in northern Finland was <10 µg/L (with a 90 percentile of 10 µg/L), and the median in northern Norway was also <10 µg/L (90 percentile of 13 µg/L). Lakes in eastern Finnmark show a clear decrease from around 20 to 30 µg/L in the early 1990s to 10 µg/L in 2004 (Figure 6.5).

6.1.2.2. **Lakes on the Kola Peninsula**

A lake survey has been conducted every five years on the Kola Peninsula (1990, 1995, 1998, 2000, 2005). Six lakes with different atmospheric loads were chosen to illustrate long-term changes. Two are near (10 km) the large point sources at Nikel and Monchegorsk while the other four are over 80 km away (Figure 6.6).

Water quality in the two lakes near the point sources had improved, with decreased concentrations of sulfate (Figure 6.6), nickel, and copper. Base cation levels have also decreased at these sites. As the concentrations of sulfate, nickel and copper also decreased in the four lakes located

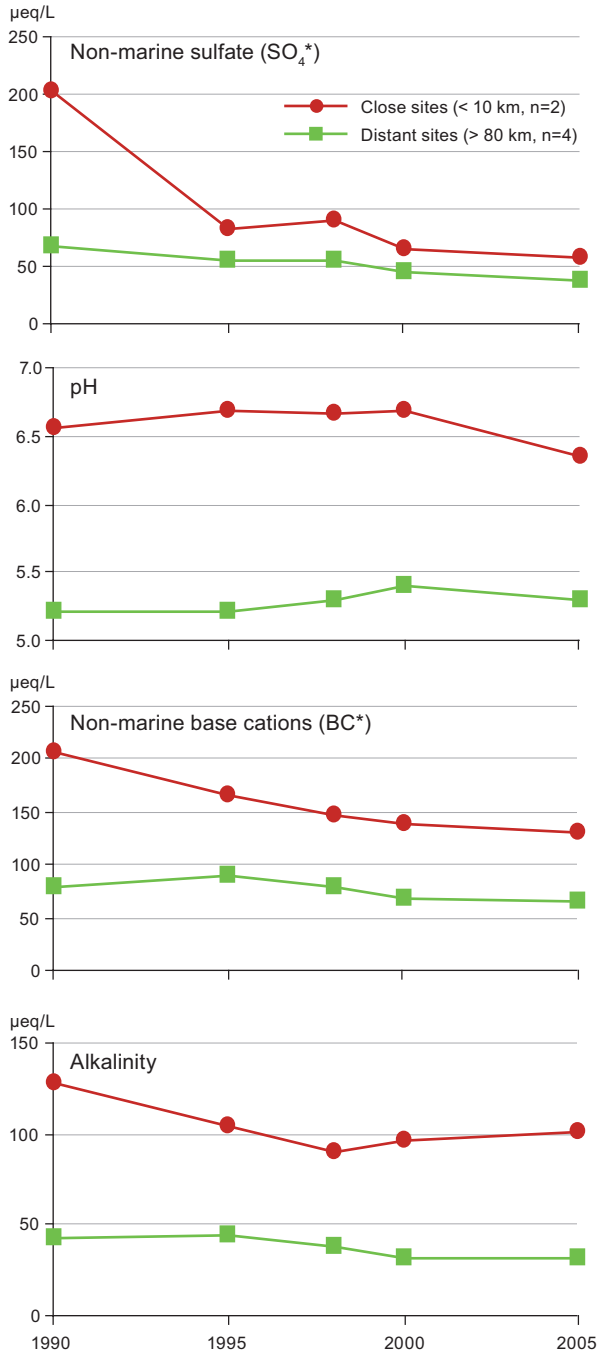


Figure 6.6. Trends in non-marine sulfate, alkalinity, non-marine base cations, and pH for lakes on the Kola Peninsula.

some distance from Nikel and Monchegorsk it appears likely that these decreases represent a decrease in regional sulfate and metal loads in the Kola North. There was no tendency for pH or alkalinity to increase despite the large decrease in sulfate.

6.1.2.3. **Swedish repeated lake survey**

In each of the Swedish lake surveys (1990, 1995 and 2000) 315 lakes were sampled in Norrbotten county. An evaluation using Theil's slope showed that SO_4 concentrations fell in 96% of lakes, which supports the results of other studies presented here. Alkalinity and ANC decreased in 91 and 69% of lakes, respectively. So this study shows no anticipated response of the lakes to the decreased sulfur deposition. Seven Norrbotten reference lakes studied several times each year also had decreasing trends for alkalinity, but in all seven the ANC increased (median 0.0015 meq/L/yr). A comparison of these two studies shows that changes observed on just a few occasions may be more indicative of variation between years than a trend. Trends, as evident from more intense monitoring, are very small and so surveys with few observations over time may be insufficient to identify the trends found using more intensive studies (Wilander, 1998).

6.1.2.4. **Concluding comments on trends**

Long-term monitoring indicates that lakes in the Euro-Arctic Barents region are recovering from acidification and that this is due to reduced sulfur deposition. However, even if the lakes have received acidic inputs and have consequently experienced a decrease in ANC, alkalinity, and pH, the lake water has not necessarily been acidified to a level at which visible damage to the biota can be expected. Lakes in eastern Finnmark near the non-ferrous metal smelters on the Kola Peninsula show the clearest signs of recovery due to reduced sulfur deposition. Other studies from the Finnish-Norwegian-Russian border area also show signs that lakes there are recovering from acidification. Surveys of small mountain lakes and brooks in northeastern Lapland in 1993 and 2000 found a significant increase in alkalinity and a decrease in sulfate concentrations. The increase in alkalinity was pronounced in the most acidic lakes (i.e., those with an alkalinity of <20 µeq/L) in 1993 (Lappalainen *et al.*, 1995; Tammi *et al.*, 2003b). The recovery of lakes further from these major pollution sources (that experienced only modest impacts from air pollutants) are affected by a high interannual variation in ion concentrations and so trends are not easily detected. Although the changes seen in Swedish surface waters are thus less pronounced than in northern Norway, they are still significant evidence of improvement in the Barents region.

6.2. Effects of acidification on arctic biota

The first AMAP assessment (AMAP, 1998) reviewed data on the effects of acidification on the freshwater biota of arctic surface waters. One of the key conclusions of the assessment was that arctic biota live in an extreme environment – the limited temperature range, ice-free season, and productivity of the Arctic produce biotic assemblages that may require very little additional stress before they are affected. The assessment also described those characteristics of arctic biota that make them especially sensitive to acidification. The assessment concluded that, while the taxa found in arctic waters are also likely to occur in sub-artic (and even temperate) waters, the number of different taxa in any one arctic lake or stream is relatively low – low taxa richness and low productivity create the potential for small changes in biotic assemblages to have relatively large ecosystem effects (Hobbie, 1984). Within the Arctic, the likelihood of low productivity and paucity of species increases with latitude. The assessment found that crustacean zooplankton may be absent from ultra-oligotrophic arctic lakes due to lack of food and that benthic invertebrate assemblages are generally dominated by chironomid larvae in lakes, and in flowing waters by the same sensitive insect taxa (e.g., mayflies) that dominate in northern temperate areas. The assessment also found that there are a small number of fish

species in most arctic waters – fish life spans may be very long (e.g., 25 to 40 years for Arctic char) due to slow growth and the slow accumulation of sufficient energy reserves for reproduction. Thus, arctic fauna is considered sensitive or vulnerable to anthropogenic alterations, including acidification (Hammar, 1989), principally because arctic assemblages occupy simple and labile ecosystems that undergo extreme climatic conditions and fluctuations.

Although there are relatively few (compared to temperate zones) published studies on arctic biota and acidification, research and monitoring prior to 1996 supports the characterization of arctic biota as susceptible to acidification (AMAP, 1998). The few documented effects include sensitive zooplankton (e.g., some species of *Daphnia*) being rare or absent from acidified lakes in the Jarfjord area of Norway (Nøst *et al*., 1992); there are no other reports of acidification effects on plankton in the Arctic. Also in the Jarfjord area, Nøst *et al*. (1992) reported low abundance of acid-sensitive mayflies (Ephemeroptera) in acidified lakes; in the Dalelva catchment (Norway) the loss of sensitive benthic invertebrate taxa was attributed to low pH and elevated aluminum concentrations (Bækken and Aanes, 1990). In the Murmansk region of Russia, the abundance of zoobenthos in small acidified lakes was as low as that in lakes contaminated by heavy metals (Yakovlev, 1992). In northeastern Finland, no acidification effects were reported for the benthos, with sensitive taxa (e.g., *Baetis rhodani*, *B. lapponicus*) found throughout the area. Many researchers concluded that fish populations in the Arctic were relatively unaffected by acidification (e.g., Erkinaro *et al*., 1992; Lappalainen *et al*., 1995; Nøst *et al*., 1992); deleterious effects of acidification on fish may only be found in the Jarfjord area of Norway (Hesthagen *et al*., 1992).

6.2.1. **Current status**

More recent data support the conclusions of the first AMAP assessment (AMAP, 1998), namely that acidification effects are rare. These data are all for the areas immediately adjacent to the Kola Peninsula – there continues to be no biological data for any acid-sensitive areas of the North American Arctic.

6.2.1.1. **Phytoplankton and periphyton**

Planktonic and epilithic diatoms are considered some of the best biotic indicators of acidification; the disappearance of acid-sensitive diatom species, and the dominance of acid-tolerant species, has long been used to quantify the long-term progression of acidification (see section 6.4). The current species composition of diatoms and chrysophytes can also be used to assess whether acidification has altered the current status of lakes and streams. There is currently no evidence of altered diatom assemblages due to acidification in the Arctic. Sorvari *et al*. (2002), for example, examined sediment cores from five lakes in Finnish Lapland and concluded that, although diatom assemblages have changed over the past two centuries, there was no evidence of changing diatom-inferred lake water pH. Similarly, Korhola *et al*. (1999) reported stable diatom assemblages from three lakes in northeastern Finnish Lapland, and concluded from their pH reconstructions that 'no substantial changes in the acidification status of lakes have occurred within the last century despite the very high local acidic deposition' (see also section 6.4).

Lakes in northeastern and northwestern Finnish Lapland have been studied for their contemporary phytoplankton fauna as a part of the European-wide project EMERGE (European mountain lake ecosystems: regionalization, diagnostics & socio-economic evaluation). One purpose of the project was to set the baseline of the ecological conditions of pristine mountain/arctic lakes in Europe for future monitoring. Even though the lakes do not appear to be anthropogenically acidified, variations in the species composition of phytoplankton indicate different responses and sensitivities to pH.

Phytoplankton were studied from 33 lakes in northern Finland as part of EMERGE. All the lakes had low biomass and low species numbers and are classified as oligotrophic or ultraoligotrophic from their chlorophyll a concentrations and phytoplankton biomass (Vollenweider and Kerekes, 1982). Most lakes were dominated by chrysophytes, which are characteristic of oligotrophic lakes with cool summer water temperatures, low alkalinity and conductivity, and neutral or slightly acid pH (Sandgren, 1988). Several lakes showed a pronounced dominance pattern, where only a few phytoplankton taxa contributed up to 80% of total phytoplankton biomass. This pattern is considered to be a consequence of stress, such as harsh climate (Willén, 2003). In the EMERGE study this pattern seemed to be more common in lakes with some additional stress (other than climate), such as high altitude or low pH. Although the studied lakes are widely distributed across Finnish Lapland, representing different environmental, altitudinal, and geomorphological settings, site-specific lake chemical properties (e.g., pH, conductivity, calcium, magnesium, sodium, alkalinity) seem to be the best predictors of species composition.

6.2.1.2. **Macroinvertebrates**

Data on benthic invertebrates from lakes and streams suggest that, while demonstrable acidification effects are rare, sensitive taxa are common in the Arctic, and the potential for subtle (or future) effects is high.

Nearly 400 small lakes and their inlet/outlet streams have been studied in northeastern Fennoscandia (Finnish Lapland, northern Norway, and the Murmansk region). Data collected between 1990 and 1997 suggest that both anthropogenic (mineral) and natural (organic) acidification have effects on the structure of benthic assemblages (Yakovlev, 1999). These effects (e.g., declines in species diversity, relative abundance and biomass in acid-sensitive invertebrates such as *Gammarus*, as well as snails, mayflies, and stoneflies) also vary strongly with natural abiotic factors of the landscape, including lake size, hydrological type, and morphology. Impoverished benthic fauna, typical of acidified lakes, was found in roughly 25% of the acid and/or humic lakes in central and northern Lapland, and attributed to decreased pH and the toxic effects of elevated aluminum (Yakovlev, 1999). Yakovlev (2000) also reported increased dominance of the benthos by predators, and a decrease in primary consumers, with decreasing pH in Fennoscandia. Both Yakovlev (1999) and Erkinaro *et al*. (2001) reported no observable acidification effects on macroinvertebrate assemblages in northernmost Finland.

Benthic assemblages may also be strongly affected by acidic episodes in the Arctic. Hämäläinen and Huttunen (1998) used a set of 17 test streams in northeastern Finland to construct a weighted averaging (WA) model predicting minimum pH from invertebrate assemblage data, and then

applied the WA model to an additional 37 streams in the region. They found that the predicted pH values correlated most closely with the minimum pH values observed during spring snowmelt. They also found a strong correlation with longitude, suggesting stronger effects (and lower minimum pH values) near the Kola region.

Midge larvae (chironomids; Diptera: Chironomidae) have been sampled extensively in the EMERGE lakes in Finnish Lapland. The composition of benthic littoral chironomid assemblages in this region appears to be strongly influenced by pH, but the assemblages do not exhibit signs of acidification. Nyman *et al*. (2005) found that the variation in chironomid communities of 50 shallow lakes across western Finnish Lapland was explained by the factors that are likely to respond to future climate change (e.g., sediment organic content, total organic carbon, pH, and mean July air temperature). These subarctic lakes are not anthropogenically acidified, but are highly vulnerable to multiple stresses caused by climate change and ultraviolet radiation (ACIA, 2004; Rautio and Korhola, 2002) (see section 6.5).

6.2.1.3. **Fish**

Recent data from acid-sensitive regions of the Arctic suggest little evidence of widespread effects on fish assemblages, but significant effects in some highly affected areas. Hesthagen *et al*. (1998) analyzed questionnaire data from 401 lakes (236 with Arctic char, 293 with brown trout) in northern Norway near the Russian border. They concluded that only three populations of Arctic char had been lost due to acidification, while three populations of char and eight of brown trout were reduced at least to some degree. This is consistent with the larger context of acidification (and recovery) in the Nordic countries; results from the project 'Fish status of Nordic Lakes' indicate that 'fish population losses were most frequent in the most highly acidified region of southern Norway and least common in eastern Fennoscandia' (Rask *et al*., 2000; Tammi *et al*., 2003a). Similarly, a study of 13 rivers in northernmost Finland found no signs of acid-induced failure in salmonid reproduction and/or recruitment (Erkinaro *et al*., 2001). Importantly, many studies of fish in acid-sensitive regions of the Arctic have focused on salmonids (e.g., Arctic char, brown trout) which may be relatively tolerant of low pH and elevated aluminum (Poléo and Bjerkely, 2000).

In a fish and water chemistry survey carried out in northeastern Finnish Lapland in 1991-1993, Lappalainen *et al*. (1995) found that the buffering capacities of small lakes and brooks were lowest in the Vätsäri area. The surface waters of the area consist of an exceptionally high number of small headwater lakes and brooks, which are sensitive to acidification due to the geochemistry of the local soil (Kähkönen, 1996). Owing to the relatively large differences in altitude over a short distance, natural obstacles to migration are common and fish populations in many lakes and brooks are isolated from each other. In some of the small lakes in the Vätsäri area, the alkalinity values were critically low in 1993, and studies revealed the first signs of acid-induced fish population damage in Finnish Lapland in local minnow (*Phoxinus phoxinus*) populations. In continuing this research and monitoring, special attention has been focused on the minnow and its reproduction because of the sensitivity of the species to acidification and its frequent occurrence in the study area (see section 6.2.2.2).

6.2.2. **Temporal trends**

6.2.2.1. **Invertebrates**

Temporal trend data are available for 12 arctic lakes in Norway (included in the Norwegian national monitoring program on long-range transported air pollution (SFT, 2005)). The microcrustacean assemblages of six lakes situated in Nordland and Troms counties were surveyed in 1999, and the data indicate no or only minor impacts of acidification. One of these lakes has been followed with annual sampling and shows no signs of changing acidification status. Six lakes in the eastern part of Finnmark (Varanger Peninsula) were surveyed in 2000 and 2004 (additional data are available for 1990 to 1997 for most of the lakes); these data indicate minor to moderate impacts of acidification in the microcrustacean communities.

Lake Dalvatn is an acid-sensitive lake in Finnmark that has been monitored for zooplankton since 1990. Signs of improvement in the acidification status of Lake Dalvatn are given by the increased abundance of the acid-sensitive cladoceran *Daphnia longiremis* (Figure 6.7). This species, which is absent in acidified lakes (Keller *et al*., 2002), was first recorded in Lake Dalvatn in 1996, and has been found each year since 1999 with increasing dominance. The presence of other acid-sensitive species of microcrustaceans has also increased in the last six years.

6.2.2.2. **Fish**

Tammi *et al*. (2003b) reported on fish data collected as part of a re-sampling of 20 lake and stream sites in the Vätsäri area of northeastern Finland in 2000; these were the same sites as were sampled in 1993 by Lappalainen *et al*. (1995),

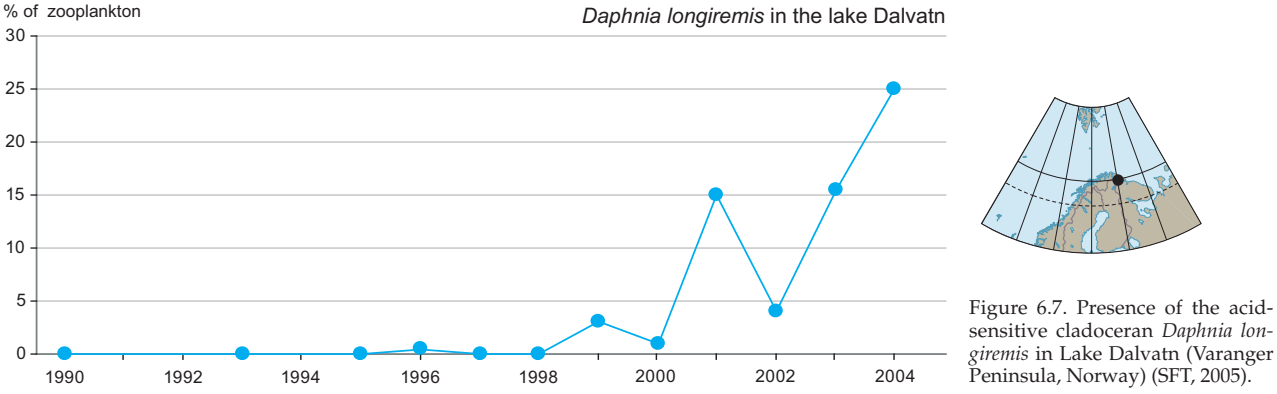


Figure 6.7. Presence of the acid-sensitive cladoceran *Daphnia longiremis* in Lake Dalvatn (Varanger Peninsula, Norway) (SFT, 2005).

who found evidence of acidification in the age-structure of minnow populations. Comparisons of the 1993 and 2000 data are shown for four sites in Figure 6.8. In all brooks and lakes, the alkalinity values were significantly higher, and the sulfate concentrations and conductivity values significantly lower, in 2000 than in 1993. The increased densities of minnow, and changes in the length distribution of the sampled fish, indicate reproductive success and recruitment of young fish at most sampling sites in the late 1990s. Although fish samples were taken in single years, the catches of electrofishing included several age-classes – the combined data on abundance and length distribution provided convincing evidence of the reproductive success of fish in the late 1990s. Despite the improvement in water quality, sparse minnow populations at some of the study sites appear to have disappeared completely during the 1990s. At these sites, the catch for 1993 consisted of only a few, rather large (60–80 mm) individuals and no minnows were found at all by electrofishing in 2000.

Lake Otervatn in eastern Finnmark has been monitored for water chemistry and brown trout populations (using benthic gill nets) since 1986 (Figure 6.9). The lake has shown a dramatic recovery in both acid/base status and brown trout populations over the last 20 years. Brown trout catch per unit effort shows a significant increase over time, and is significantly correlated with the observed increase in alkalinity. These data are strongly suggestive of recovery from acidification.

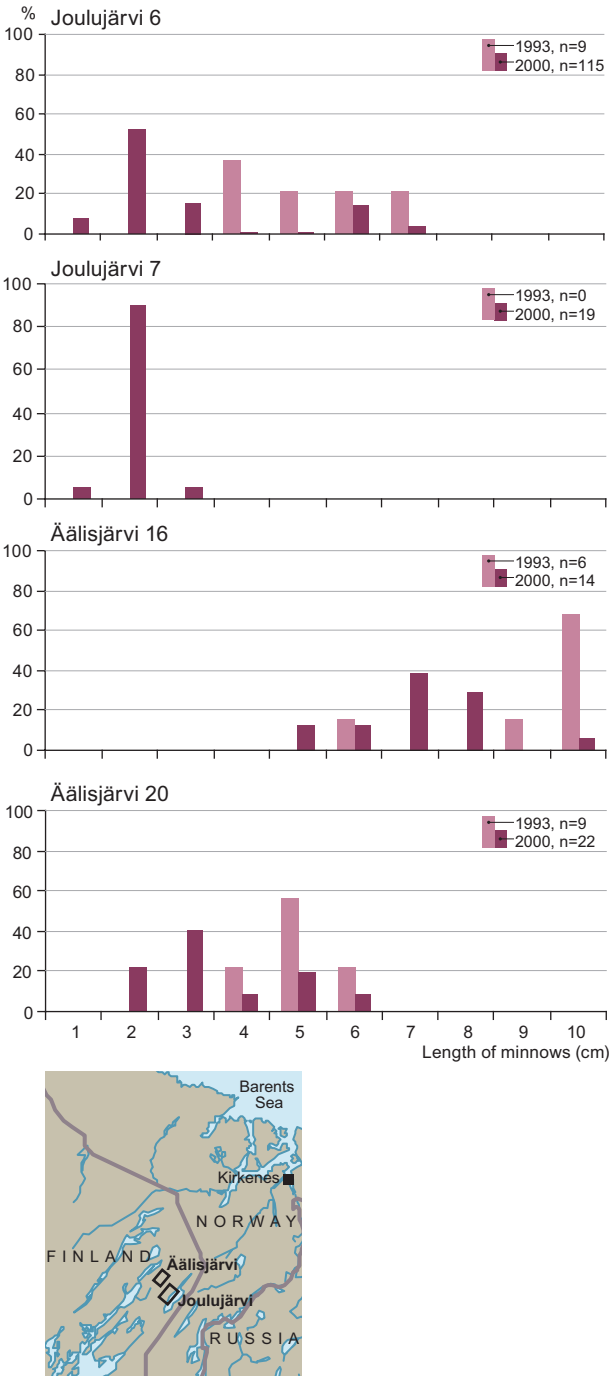


Figure 6.8. Length distributions of minnows caught by electrofishing in the Joulujärvi area (sites 6 and 7), and in the Äälisjärvi area (sites 16 and 20) in 1993 and 2000 (re-drawn from Tammi *et al.*, 2003b).

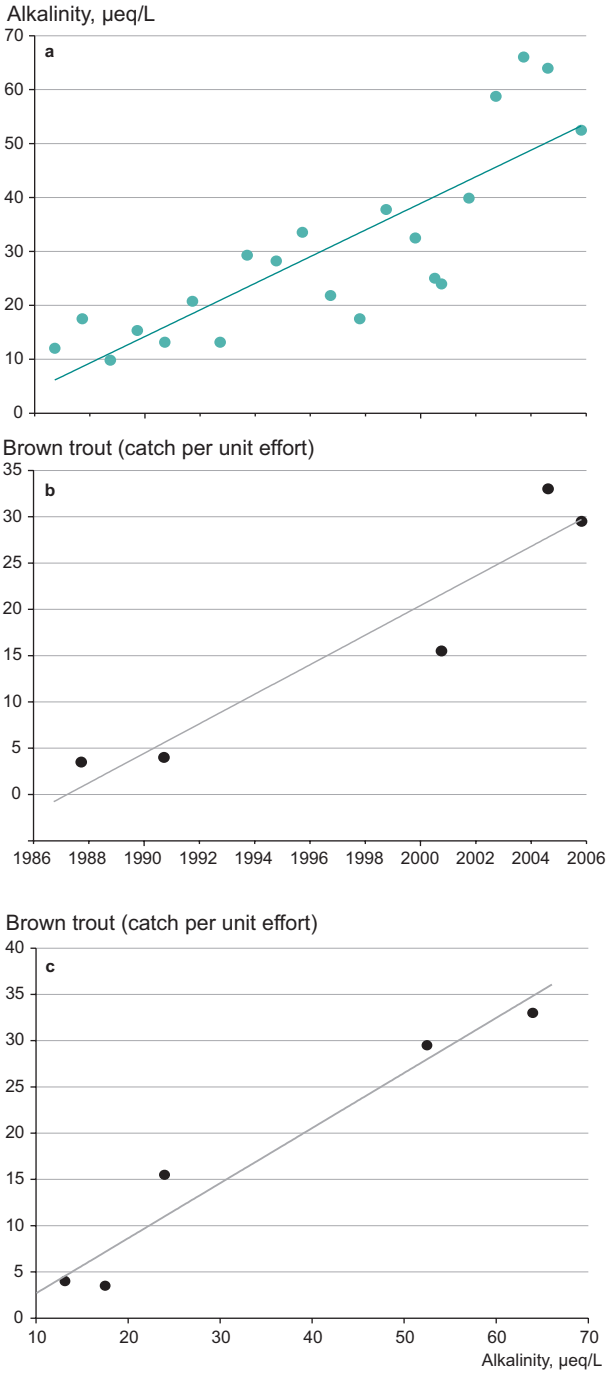


Figure 6.9. Data for Lake Otervatn in eastern Finnmark, showing (a) alkalinity (µeq/L) and (b) brown trout catch per unit effort (CPUE, expressed in number of fish caught per 100 m^2 of gill net area) since 1986, and (c) brown trout populations in relation to changes in alkalinity.

6.3. Episodic acidification

Episodic acidification of surface streams during spring floods is a ubiquitous phenomenon all over the world, including arctic regions. Acid episodes are especially dangerous in the Arctic; contaminants deposited from the atmosphere accumulate in the snowpack during the long polar winter and are released rapidly into drainage basins during the short spring flood in acidified polluted snowmelt waters that cause a sharp decrease in pH and alkalinity. Simultaneous pulses of protons and metals may lead to episodes with extremely strong toxic effects in streams and rivers. pH in rivers often drops during spring or rain floods under natural conditions because of base-flow dilution by atmospheric precipitation, although in some cases this phenomenon can be due to the leaching of natural organic acids from soils in forested or wetland catchments (Laudon and Bishop, 1999). However, this natural pH decline can also be dramatic and may affect biota. The accumulation of anthropogenic acids (SO_4, NO_3) in catchments abruptly intensifies the episodic acidification in flood periods. Arctic ecosystems are especially vulnerable to acid pulses during spring floods. After polar night, the vulnerability of arctic biota to acid and toxic impacts is much higher. Situations in which two or more stressors occur simultaneously, thereby multiplying the risk for aquatic life, are dealt with in more detail in section 6.5. Episodic pH decline has been observed in several arctic regions, for example, Sweden, Finland, and Russia (Kinnunen, 1992; Moiseenko, 1999; Laudon *et al.*, 2000; Moiseenko *et al.*, 2001).

How pH episodes form depends on the conditions of water formation. Catchments in the Arctic are mostly of the tundra or forest type, although hilly and mountainous catchments also occur. The mechanism of pH depression in surface waters is determined, as a rule, by the interaction of several factors capable of causing acidification. Published reports emphasize five factors that contribute to depressed ANC during flood time: (1) dilution from increased discharge; (2) H_2SO_4 and (3) HNO_3 derived from precipitation or natural sources; (4) organic acids derived from watershed soils or wetlands; and (5) HCl derived from 'salt-effect', i.e., reactions within catchment soils (e.g., Jeffrey *et al.*, 1992).

6.3.1. Acidic episodes in the Kola region

Acidic episodes have been studied in the Kola North, where a high level of anthropogenic sulfur deposition has been recorded. The decrease in pH during spring flood relative to the pre-episode period (i.e., the winter low-water period) was examined in 21 streams (Moiseenko, 1999; Moiseenko *et al.*, 2001). The decrease was most noticeable in streams where the pre-episode pH was high, while in streams that were chronically acidified (with pH values during the low-water period of <6) the pH drop was insignificant owing to the similarity between the chemistry of water in acidified streams and atmospheric precipitation (Figure 6.10). This is in general agreement with data collected in Europe and North America (Wigington *et al.*, 1992). The period of pH depression during spring flood in this part of the Arctic is short and rarely exceeds five to seven days.

Acidic episodes facilitate the discharge of metals into streams and rivers. It is well known that water acidification causes an increase in the concentrations of labile metal forms (Dillon *et al.*, 1988; Nelson and Campbell, 1991; Jeffries, 1997). In arctic regions, an abrupt drop in water pH in a short flood period is accompanied by a pulse of metals, especially in their ionic forms, and it is this that generates the greatest hazard for aquatic organisms. The leaching of metals during flood (2–3 weeks) can account for up to 75% of their total annual load (Moiseenko, 1999; Moiseenko *et al.*, 2001). Data on streams of the Kola North showed that in the periods of pH depression during floods, the total metal concentration increases in all types of creek, notwithstanding dilution by snowmelt water. The total concentration of aluminum in water increased by 50–88%. In the most acidified tundra stream studied, an increase in the concentration of its labile form was accompanied by a drop in the form bound with organic complexes.

6.3.2. Acidic episodes in the Dalelva catchment in eastern Finnmark, Norway

Dalelva is a small (3.2 km^2) undisturbed catchment dominated by heathland and mountains, in Jarfjord in eastern Finnmark, Norway (69°45′ N, 30°23′ E). Vegetation comprises birch forest to an elevation of about 150 m with heath and moorland above. Small lakes cover about 15% of the catchment. The catchment is usually covered with snow for six to seven months of the year, approximately from mid-October to late May, and receives relatively little precipitation (500–600 mm/yr). At the adjacent meteorological station (Lanabukt) about 45% of the annual precipitation was accumulated as snow during 1990 to 2000 (Kaste and Skjelkvåle, 2002). The extensive snow accumulation has a large impact on the seasonal runoff pattern, which is characterized by very low flow during winter and a distinct snowmelt flood in May to June.

The large seasonal variations in streamwater flow also have big impacts on water chemistry (Kaste and Skjelkvåle, 2002; SFT, 2005). During the long cold winters, when precipitation is accumulated as snow, the stream is dominated by baseflow with relatively high solute concentrations

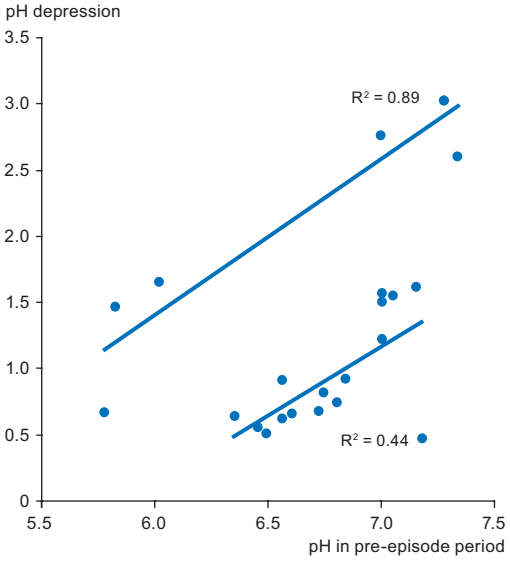


Figure 6.10. Water pH depression during spring flood versus pH during the low-flow period in streams of the Kola North. The upper line shows the relationship in streams where the minimum pH during high flow was <5, and the lower line where the minimum pH during high flow was >5.

(Figure 6.11). Concentrations of major ions and nitrogen compounds (not shown) tend to build up during winter and then decrease rapidly to initial levels at the start of the snowmelt flood. This dilution of base cations during snowmelt also causes a rapid decrease in streamwater pH, from the normal level of around 6.0 to 6.5, to values of around 5.5 (Figure 6.11). Sea-salts accumulated in the snowpack during winter are eluted from the snow in the early melting phase, resulting in an annual peak in chloride concentrations immediately before the onset of the main snowmelt flood (Figure 6.11).

After snowmelt, streamwater flow and solute concentrations return rapidly to normal summer levels. The Dalelva catchment usually experiences a short and intense growing season that lasts approximately four months. During summer, air temperatures can be relatively high and owing to the relatively low precipitation amounts, streamwater flow is usually very low at this time.

The overall water chemistry at Dalelva is characterized by relatively high concentrations of non-marine sulfate owing to industrial SO_2 emission sources on the Russian side of the border. During 1990 to 2000, however, streamwater concentrations of non-marine sulfate declined by 35% as a result of reduced SO_2 emissions (SFT, 2005). Dalelva is moderately affected by humic substances; average concentrations of total organic carbon between 2001 and 2003 were in the range 3.7 to 4.4 mg C/L. The greatest influence of total organic carbon is often associated with the onset of snowmelt. Streamwater concentrations of nitrate are relatively low, with annual peaks up to 70 to 100 µg N/L prior to snowmelt, and growing season values typically below 5 µg N/L.

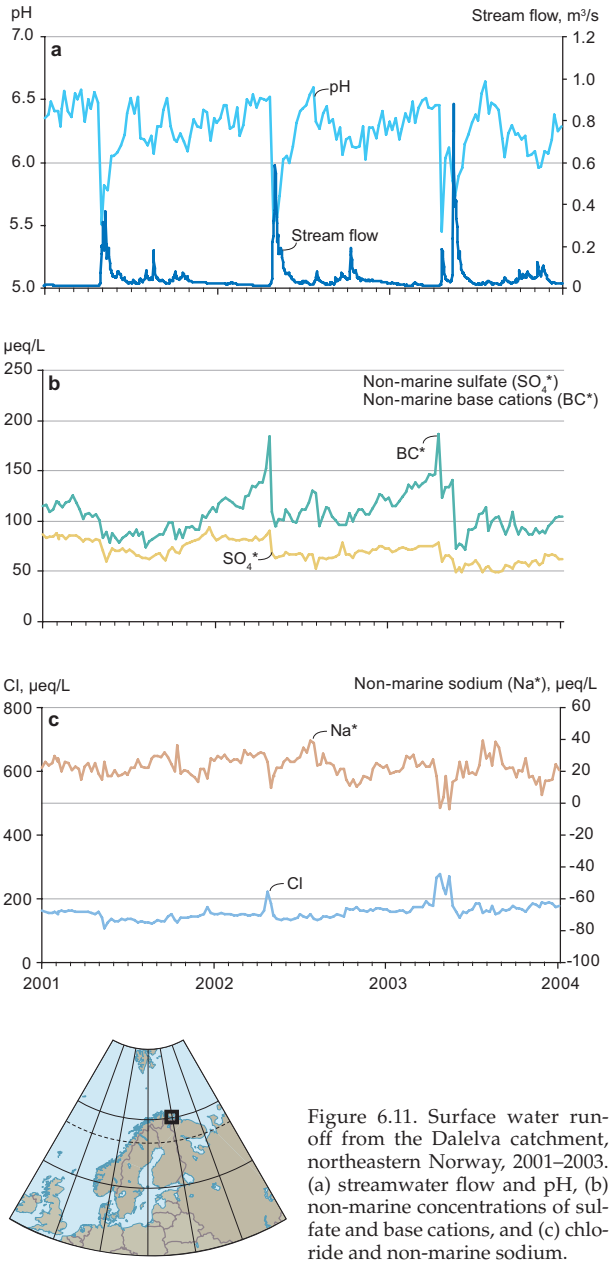


Figure 6.11. Surface water runoff from the Dalelva catchment, northeastern Norway, 2001–2003. (a) streamwater flow and pH, (b) non-marine concentrations of sulfate and base cations, and (c) chloride and non-marine sodium.

6.3.3. **Acidic episodes in northern Sweden**

A large, multi-investigator project in northern Sweden developed the Boreal Dilution Model (BDM) to quantitatively distinguish the natural and anthropogenic mechanisms that drive episodic decline of ANC and pH during hydrological events (Bishop *et al.*, 2000; Laudon *et al.*, 2000). The BDM identifies the anthropogenic component of episodic ANC decline (ΔANC_{poll}) from relative differences in the runoff dynamics of base cations and anthropogenic acid anions during episodes. Snow chemistry data are not used in the BDM. However, comparison of snow chemistry with the results from over 50 applications of the BDM in catchments from the alpine zone to the coast of northern Sweden sampled between 1991 and 1999 revealed a strong relationship between SO_4 in snow and the anthropogenic impact on the subsequent spring flood. This suggests an immediate and proportional response in spring flood acidification to changes in winter SO_4 deposition. Nitrogen is not a factor in the spring flood of this nitrogen-poor region (Laudon *et al.*, 2000).

Trends in anthropogenically driven episodic acidification were analyzed in five streams from northernmost Sweden between 1990 and 1999 using the BDM (Laudon and Hemond, 2002). Although there was no significant change in annual average stream water chemistry, the anthropogenically driven episodic acidification associated with spring flood runoff decreased by 40 to 80%. A strong correlation between winter SO_4 deposition and the anthropogenic component of episodic acidification in these five streams suggests that future reductions of acid deposition will further improve the spring flood acidification situation in northern Sweden. These results also indicate that reduced emissions of acid precursors have generated significant improvements in the surface water chemistry during episodes associated with spring runoff in northern Sweden.

While the data requirements of the BDM are too big for regional assessments, the correlation between snow SO_4 and ΔANC_{poll}, together with a relatively consistent amount of snowmelt in the peak of spring flood creates the basis for the more empirical 'one point BDM' (pBDM, Laudon *et al.*, 2004). This model uses widely available lake chemistry measurements, and can thus provide a more synoptic view of how human impact on spring melt ANC in northern Sweden has responded to changed SO_4 deposition. This is a region where many surface waters experience low ANC and pH in conjunction with hydrological episodes despite a relatively low annual acid deposition load. The largest episodic ANC declines and greatest biological effects in the region are associated with spring flood (Laudon *et al.*, 2000).

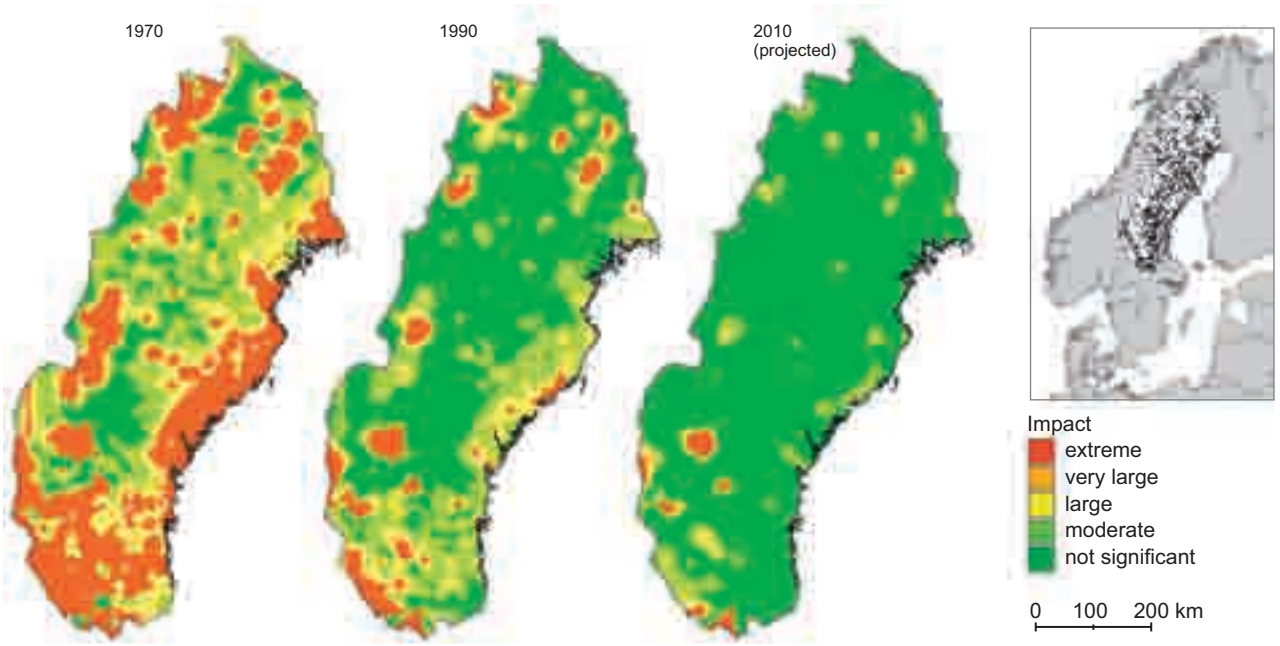


Figure 6.12. Intensity of anthropogenic episodic acidification in northern Sweden during the 1970 spring flood and the 1990 spring flood, and projected for the 2010 spring flood. Assessed using the acidification index of the Swedish Environmental Protection Agency and the pBDM model (Laudon and Bishop, 2002).

Sulfur deposition in northern Sweden peaked in 1970 (Mylona, 1996) and had declined by 65% by 1990 (S. Mylona, Norwegian Pollution Control Authority, unpubl. data). A further 55% reduction relative to the 1990 level is expected by 2010 in accordance with the 1999 Gothenburg Protocol to the LRTAP Convention. Winter chemistry data suitable for use in the pBDM was available for 1240 lakes sampled in conjunction with national monitoring (Figure 6.12). Application of the pBDM to these lakes using the 1990 and 2010 data projects a clear recovery (Figure 6.12, Table 6.4). In 1970, 54% of lakes in Sweden were subject to significant human impact (classes 3–5, Table 6.4). By 1990, only 14% of the lake population was more than moderately impacted by acid deposition during spring. By 2010 that number is projected to be 3%. Reductions in acid deposition have led to rapid and substantial chemical recovery from spring flood acidification in arctic waters, including 70 000 lakes and 1 000 000 km of watercourses in northern Sweden (Laudon and Bishop, 2002). These results are based on the strong correlation between SO_4 concentration in snow and the anthropogenic component of spring flood ANC decline (Laudon *et al*., 2004). Laudon *et al*. (2004) estimated that the 65% reduction in sulfur deposition between 1970 and 1990 reduced the area of acidified spring floods across 250 000 km^2 of northern Sweden by 75%.

A large and rapid reduction in the anthropogenic influence on spring flood acidity does not mean that spring ANC/pH decline disappears. A large spring flood ANC decline (equivalent to ca. 50%) is a natural feature of aquatic ecosystems in northern Sweden. The relative increase in organic acids during spring flood also contributes to a natural pH decline (Ivarsson and Jansson, 1995; Laudon *et al*., 1999).

The difficulties in assessing episodic acidification have contributed to the focus of previous acidification recovery assessments on changes in average lake conditions. The failure to account for an episodic response may greatly underestimate the immediate benefits of reducing acid deposition. Furthermore, since spring flood responds directly to snow acidity in this region that is not chronically acidified, delays in deposition reduction translate directly into recovery delays. Important as these results are for northern Sweden where large investments are made each year to remediate acidification in spring flood by liming, it is very likely that extensive regions in the boreal zones of North America, Europe and Asia may be similarly sensitive to changes in winter SO_4 deposition.

Table 6.4. Extent of anthropogenic acidification in northern Sweden (Laudon and Bishop, 2002).

			Percentage cover		
Acidification class	ANC_p / ANC_{pi}	Acidification index	1970	1990	2010
1	1.0 - 0.75	not significant impact	20	63	91
2	0.75 - 0.50	moderate impact	26	23	6
3	0.50 - 0.25	large impact	18	6	1
4	0.25 - 0.10	very large impact	7	1	1
5	<0.10	extreme impact	29	7	1
1-2			46	86	97
3-5			54	14	3

ANC_p: peak in ANC; ANC_{pi}: pre-industrial peak in ANC

6.3.4. Concluding comments on episodic acidification

In arctic regions, episodic acidification is a ubiquitous natural phenomenon during spring flood that has been intensified by acidic deposition. It develops swiftly due to the pollutants accumulated in the snowpack over winter being released rapidly into drainage basins during snow melt. The dominant factor in the acidification mechanism is the type of stream catchment. Replacement of hydrocarbonates by stronger acids and dilution are the most apparent and well-known factors and these affect acid episodes in all streams. In forest and wetland streams, organic acids also contribute strongly to pH decline. In remote coastal tundra areas, HCl is the dominating factor because of ion-exchange processes in catchments (adsorption of marine aerosol Na^+).

A general pattern in the behavior of metals induced by acidification is an increase in their concentration, redistribution toward the most toxic ionic form, and pulses of metals during the period of episodic acidification. An abrupt increase in the ionic form, which is the most toxic form for biological systems, together with low pH causes a toxic stress for water inhabitants. In polar regions, the maximum stress for biota occurs during spring flood periods, when pH is at a minimum and the concentration of ionic forms of metals is at a maximum.

A model based assessment on data from five streams in northern Sweden indicated that reduced emissions of acid precursors have generated significant improvements in surface water chemistry during episodes associated with spring runoff. Although there was no significant change in the annual average stream water chemistry at these sites, the anthropogenically driven episodic acidification associated with spring flood runoff decreased by 40 to 80% between 1990 and 1999. A regional scale model application indicated that the 65% reduction in sulfur deposition between 1970 and 1990 has reduced the area of acidified spring floods across 250 000 km^2 of northern Sweden by 75%. It is also likely that future reductions in acid deposition will further improve the spring flood acidification situation in this region.

The difficulties of assessing episodic acidification have contributed to many previous acidification recovery assessments having focused on changes in *average* lake conditions. The failure to account for an episodic response may greatly underestimate the immediate benefits of reducing acid deposition. Also, since spring flood responds directly to snow acidity in those northern regions that are not chronically acidified, delays in deposition reductions translate directly into recovery delays.

6.4. Evidence from paleolimnological studies

Monitoring of northern lakes often started after industrial pollution had been ongoing for tens of years. In the absence of long-term data on water quality, as is often the case in the Arctic and subarctic, paleolimnological reconstructions using microfossils preserved in lake sediments provide a powerful chronology of acidification history and recovery and a tool for separating anthropogenic impact from the natural pH succession. Microfossils are surprisingly accurate proxy sources for reconstructing past environmental conditions. Modern distributions of organisms that preserve well in lake sediments can be related to the hydrochemistry of the water body using a large set of lakes ('training set') and environmental optima and tolerance ranges for the individual species. These ecological optima and tolerance ranges can then be applied to fossil communities by means of mathematical calibration (transfer) functions, which enable changes in the hydrochemistry of a lake to be quantitatively determined from the fossil assemblages deposited in the sediment over a certain time period. In particular, the strong linkage between diatoms and lake-water pH has long been recognised (for a historical review, see Battarbee *et al*., 2001), which is why diatoms have been widely used as indicators in bio-monitoring present and past changes in the acidification status of surface waters. Using paleolimnological techniques, it is possible to obtain a temporal perspective on the lake ecosystems with regard to acidification (Smol, 1992).

In the previous AMAP assessment (AMAP, 1998), paleolimnological assessments of acidification in arctic Canada and Alaska showed that the lakes had been unproductive throughout their entire existence and that long-term natural acidification was still occurring. However, anthropogenically-induced acidity was reported to have affected the present-day acid-sensitive plankton species, invertebrates, and fish on the Kola Peninsula and in the neighboring areas of Finland and Norway (AMAP, 1998). Critical loads were reported to have been exceeded in large areas of northern Finland and Norway due to the low critical load values in these systems (the lakes in the region are characterized by a low buffering capacity and are sensitive to acidification, see section 6.1) and the influence of emissions from industrial areas on the Kola Peninsula. At present, the diatom-based pH-reconstructions cover extensive areas of arctic Fennoscandia, the Kola Peninsula, Siberia (Norilsk), Svalbard, and arctic Canada.

6.4.1. Millennial trends in lake acidification

6.4.1.1. Fennoscandia and the Kola Peninsula

Natural long-term acidification is a common feature of lakes in cold environments with thin soils and acid bedrock in the catchment. Most of the available pH reconstructions are based on changes in the subfossil diatom flora, since diatoms are very responsive to changes in acidity (e.g., Weckström *et al*., 1997; Bigler and Hall, 2002). Seppä and Weckström (1999) studied the acidification history of Lake Tsuolbmajavri, located just above the present pine treeline in northwestern Finnish Lapland. The current pH of the lake is around 7.4. Diatom-inferred pH (hereafter referred to as DI-pH) was observed to have decreased slowly and gradually throughout the Holocene (i.e., the last 10 000 years) (Figure 6.13). The periods of slightly more evident drops in DI-pH values were related to climate-driven changes in soil-forming processes and catchment vegetation patterns, for example the immigration of pine and the initiation of paludification. However, the influence of peatlands on the acidification status of lakes is probably less clear in northern Fennoscandia than in more southerly boreal environments owing to the less acidic nature of the mire vegetation and water in the northern rich fens (Korhola *et al*., 2002; Sjörs and Gunnarsson, 2002). A similar slow and gradually decreasing pH trend was reconstructed from another lake (Lake Toskaljavri) in the barren tundra region of Finnish Lapland (Figure 6.13), with a pH decrease of ca. 0.3 to 0.4 pH units during the Holocene (Seppä *et al*., 2002).

An even weaker trend in natural acidification was evident in data from Solovieva and Jones (2002), who studied the Holocene history of a small upland lake (Lake Chuna) on the Kola Peninsula (Figure 6.13). The lake water is currently dilute, clear and slightly acidic with a pH of around 6.4. The lake experienced slow natural acidification in the early Holocene, with the acid-base balance achieved about 4000 years ago after which there appeared to have been no further acidification. By applying the diatom models developed by Weckström *et al*. (1997) and Solovieva (2000), Grönlund and Kauppila (2002) recorded a similar slight trend of progressively declining DI-pH towards the present in Lake Soldatskoje, a small tundra site located in the northern coastal area of the Kola Peninsula. Similarly,

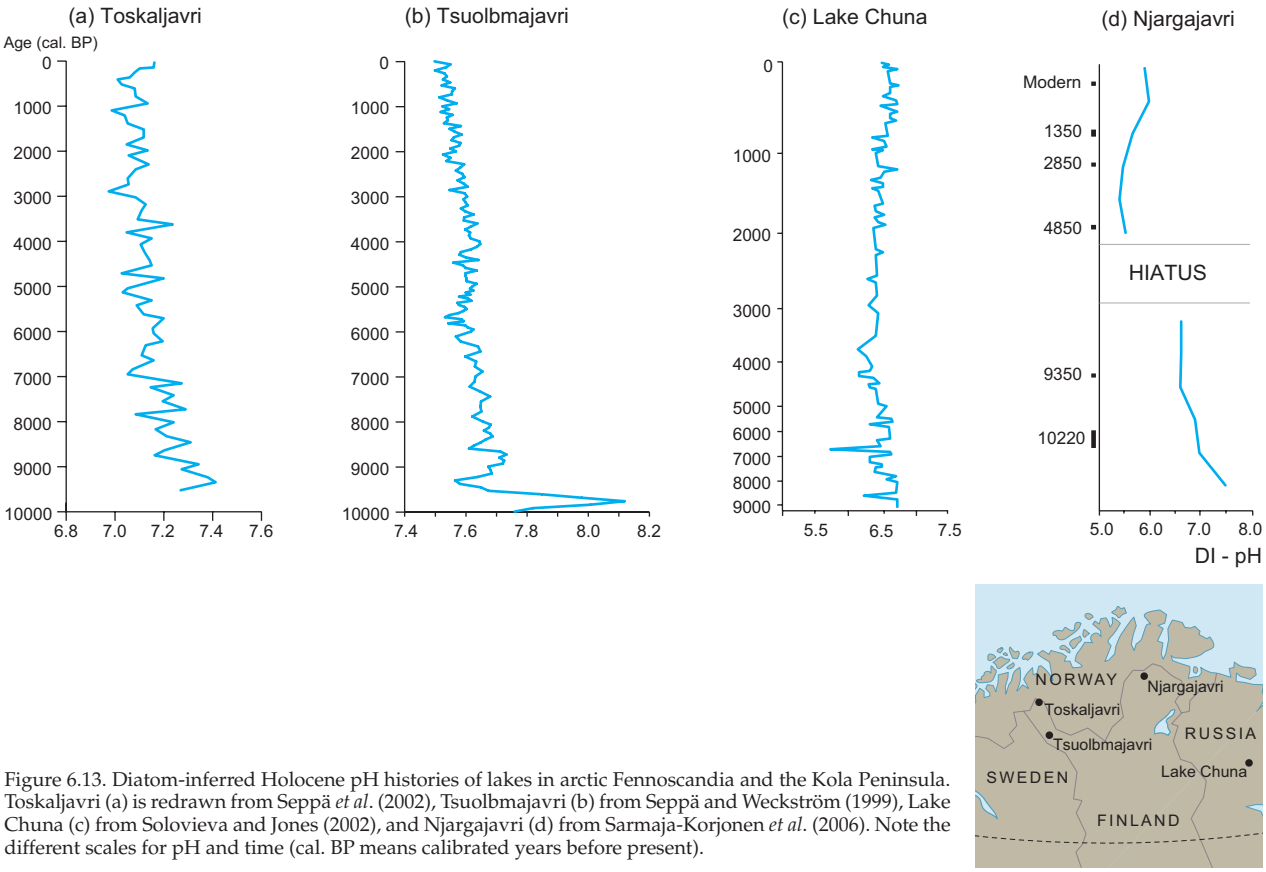


Figure 6.13. Diatom-inferred Holocene pH histories of lakes in arctic Fennoscandia and the Kola Peninsula. Toskaljavri (a) is redrawn from Seppä *et al*. (2002), Tsuolbmajavri (b) from Seppä and Weckström (1999), Lake Chuna (c) from Solovieva and Jones (2002), and Njargajavri (d) from Sarmaja-Korjonen *et al*. (2006). Note the different scales for pH and time (cal. BP means calibrated years before present).

Bigler *et al*. (2002a, 2003) found that two small subarctic lakes near Abisko in northern Sweden had gone through a slow long-term acidification during the Holocene from about pH 7.2 to 6.8 and 7.2 to 6.5, respectively. No evidence was found that changes in land-use or reindeer herding would have affected the acidity status of these lakes during the last 1000 years.

Korsman (1999) reconstructed the late Holocene pH history from five currently acidic lakes in northern Sweden using diatoms and reported a slight natural acidification trend beginning thousands of years ago as a result of soil-forming processes and natural changes in vegetation. One of these lakes is within the AMAP region. In a recent study, Sarmaja-Korjonen *et al*. (2006) reported relatively strong mid-Holocene acidification of around two pH units from 7.5 to 5.5 in Lake Njargajavri, a small shallow lake in Finnish Lapland (Figure 6.13). The reasons for the rapid decline in pH may be due to specific characteristics of this poorly buffered, acid (pH 5.3) and oligotrophic lake and its catchment that made the system particularly susceptible to climatically-induced changes in pH. Climate is known to modulate lake acidity through links between lake ice cover, primary productivity, and DIC dynamics (Wolfe, 2002, see section 6.5.1).

6.4.1.2. **Concluding comments on millennial-scale acidification**

Millennial-scale changes in lake water acidity in arctic Fennoscandia and the Kola Peninsula are mainly slow or non-existent during the Holocene. Excluding the initial transient alkaline period following deglaciation evident at some sites, Korhola and Weckström (2004) estimated that the long-term natural rate of pH decline in arctic lakes in this region has been around 0.005 to 0.01 pH units per 100 years, which is considerably less than the corresponding 0.01 to 0.02 pH units per 100 years in boreal lakes. Some arctic lakes may, however, be especially vulnerable to changes in climate, which can modulate pH.

6.4.2. **Recent acidification**

6.4.2.1. **Fennoscandia and the Kola Peninsula**

In 1990, critical loads for surface waters in northern Europe were exceeded almost everywhere. Henriksen *et al*. (1997a,b) reported that critical loads of acid deposition were exceeded in 50 to 70% of lakes on the Kola Peninsula and in the Norwegian–Russian border areas. Regional geochemical mapping from 1992 to 1998 demonstrated that although the local influence of industry on the Kola Peninsula can be seen, distance from the coast dominates the distribution of pH and sulfur trends in lake water at the regional scale (Reimann *et al*., 2000a).

A quick and efficient paleolimnological means to establish the extent of lake acidification at the regional level is the so called 'top–bottom approach'. From a number of lakes a single sediment sample is analysed (e.g., for diatoms) from the top of the sediment layer, to represent present-day conditions, with another taken from deeper down the core to represent pre-industrial conditions. By comparing these samples it is possible to estimate the extent of the change in species assemblages and thus the DI-pH values since pre-industrial times. This method was applied to 118 northern Swedish lakes (Korsman, 1999) and to 32 lakes around the smelters on the Kola Peninsula (Weckström *et al*., 2003). The results indicate that in the majority of the Swedish lakes DI-pH was unchanged, while on the Kola Peninsula the decrease in DI-pH occured only within the immediate vicinity of the smelters. These diatom data do not sup-

port the hypothesis of large-scale modern acidification in northern Sweden, nor the widespread acidification of arctic lakes due to sulfur pollution from the Kola smelting and mining industries. However, in four lakes out of 15 studied by Dauvalter (1997) in Finnish Lapland, acidification was postulated to have altered the geochemical cycling of potentially harmful metals by dissolution from sediments back to water or by reducing the adsorption of metals onto sedimenting particles.

In addition to these broad-scale top-bottom studies, several downcore pH reconstructions covering the past 200 to 300 years have been made for a number of individual lakes (see Figure 6.14). Collectively, these studies indicate that no substantial changes in DI-pH have taken place. The next paragraph summarises some of these studies.

Data from three small, potentially acid-sensitive (autumn alkalinity values of 20, 20, and 60 µeq/L) lakes, two in eastern Finnish Lapland 40 km west and 150 km southwest from the Nikel smelter and a reference lake in western Lapland, suggested no substantial changes in DI-pH despite the relatively high acid deposition in the east (Korhola *et al*., 1999) (Figure 6.14, lakes A-C). The other lakes (Figure 6.14, lakes D-N), dilute clearwater lakes with varying geology, were not chosen to evaluate their acidification history and so their sensitivity to acidification was not a major criterion for selection. Sorvari *et al*. (2002) did not find change in DI-pH over the last 200 years in five tundra lakes in northwestern Finnish Lapland (Figure 6.14, lakes H-L) investigated using high-resolution sediment sampling. Similarly, Weckström (Environmental Change Research Unit, University of Helsinki, unpubl. data) found no decrease in DI-pH in two lakes in western Finnish Lapland (Figure 6.14, M-N), in fact one even showed a slight increase in DI-pH since 1800 AD.

Sediments representing the industrial time of Lake Chuna, a small upland lake on the Kola Peninsula about 30 km from the Monchegorsk smelter, were studied by Moiseenko *et al*. (2000) and Ilyashuk and Ilyashuk (2001). Moiseenko *et al*. (2000) found that toward present times the acidobiontic diatom taxa (as well as abnormal forms of some diatom species) increased as species diversity decreased, these changes running in parallel with the start of heavy metal accumulation due to industrial development of the region. Ilyashuk and Ilyashuk (2001) studied changes in the subfossil benthic invertebrate (Diptera: Chironomidae) communities in the sediments of Lake Chuna. They concluded that compositional changes in the assemblages were due to inputs of airborne contaminants and climate change. According to their interpretation, the first changes that took place in chironomid communities around 1950 were caused by the decrease in pH and the accumulation of heavy metals in bottom sediments. In the uppermost sediment layers climate change may have lead to the decrease in the predominant species, to increased taxon evenness, and thus to an increase in species diversity over the last two decades. Solovieva and Jones (2002) studied the same lake using modern quantitative approaches but did not find any significant decline in DI-pH over recent decades/centuries. This discrepancy between the pH reconstructions for Lake Chuna from the two different diatom studies is still to be resolved. It may in part result from methodological differences in reconstructing pH. Nevertheless, over the last 100 years the sediments have accumulated some toxic elements (lead, nickel, copper, cobalt, and cadmium) despite their

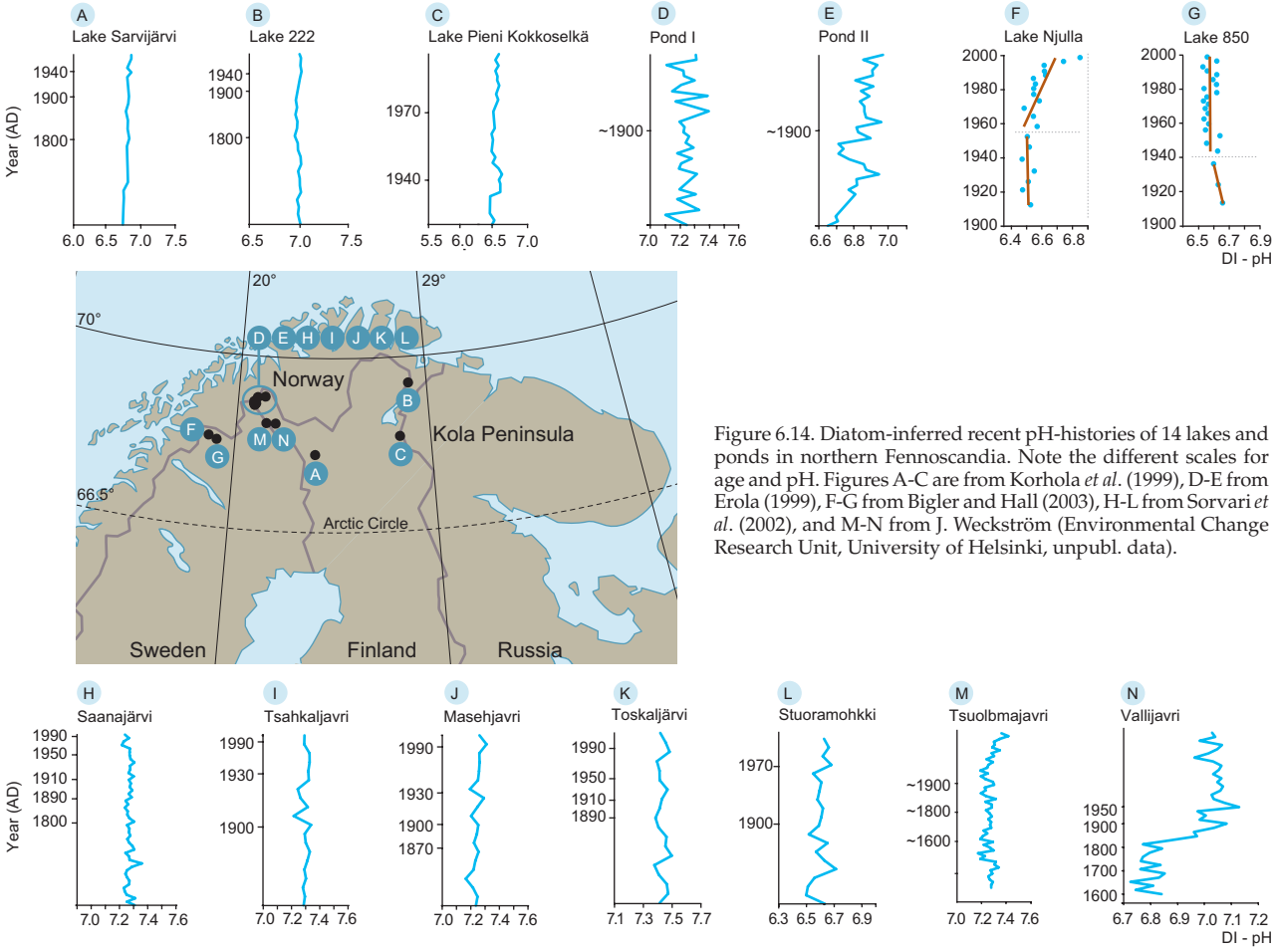


Figure 6.14. Diatom-inferred recent pH-histories of 14 lakes and ponds in northern Fennoscandia. Note the different scales for age and pH. Figures A-C are from Korhola *et al*. (1999), D-E from Erola (1999), F-G from Bigler and Hall (2003), H-L from Sorvari *et al*. (2002), and M-N from J. Weckström (Environmental Change Research Unit, University of Helsinki, unpubl. data).

low concentrations in water. The increasing appearance of deformations in diatom and chironomid specimens towards recent times may indicate that the accumulation of heavy metals has had an adverse effect on the lake biota.

In their study of two bays of Lake Imandra on the Kola Peninsula (Monche Bay in the near vicinity of the Monchegorsk copper-nickel smelter and Kunchast Bay, an internal reference site located about 90 km southwest from the smelter), Ilyashuk *et al*. (2003) found an increase in the metal contamination of the sediments in Monche Bay that was accompanied by marked changes in chironomid communities towards more toxic-tolerant species and a decline in diversity and total abundance. Also, the presence of increasingly frequent morphological deformities among the detritus-eating *Chironomus* and the decreased Benthic Quality Index (BQI) both served to indicate increasingly toxic conditions for the biota. At the reference site (Kunchast Bay) no obvious changes in chironomid communities were noted that could be connected to metal contamination and acidification. The mouthpart deformities of *Chironomus* may reflect a switch from non-genetic stress in midge larvae to an increasing amount of stress at genetic level under prolonged stress conditions (Ilyashuk *et al*., 2003).

Dauvalter and Rognerud (2001) studied the impact of heavy metals on the watershed of the Pasvik River, the largest river system in northern Fennoscandia. They found increased concentrations of heavy metals in recent sediment layers in the lower reaches of the river. This was presumably due to the atmospheric emissions of nickel, copper, cobalt, zinc, cadmium and mercury from the smelters and to wastewaters from tailing dams and mines. In the upper river reaches no significant changes in vertical distribution of heavy metals were apparent.

The geochemical and biological data from the Kola Peninsula suggest localized effects of pollution from the smelter industries within a few tens of kilometers from the actual emission sources. However, no precise dates were available for the sediment cores studied by Dauvalter and Rognerud (2001) and Ilyashuk *et al*. (2003) and the time resolution of the samples (1 cm sediment slices) was not high enough to allow estimates of any recovery.

6.4.2.2. **Siberia**

Using the top–bottom approach, Michelutti *et al*. (2001) found that diatom assemblages in the sediments of 17 lakes had experienced relatively little change since pre-industrial times in the Norilsk area, in Russian Siberia. Lakes seemed well buffered against acidification due to the surrounding alkaline bedrock and overlying glacial deposits. According to their investigations, the effects of the massive mining activities on the water quality of these lakes have been minimal, and the alkaline nature of the lake water has resulted in the incorporation of the insoluble metallic complexes into the lake sediments. However, the mining activities may have caused increased erosion, which had altered the species assemblages to some extent (Michelutti *et al*., 2001).

There is no evidence of widespread lake acidification during the industrial period in the Usa Basin of the East-European Russian Arctic according to the subfossil diatom assemblages studied by Solovieva *et al*. (2002). However, they did find evidence of alkalinization of the lakes due to atmospheric deposition, as shown by the increased DI-pH values towards the present. Similarly, Solovieva *et al*. (2005) did not find any detectable signs of acidification in two lakes within this region studying both diatoms and chironomids from the high-resolution sediment record extending back to about 1800.

6.4.2.3. **Svalbard**

Elevated levels of atmospheric contaminants such as spheroidal carbonaceous particles (from fossil fuel combustion), polyaromatic hydrocarbons, polychlorinated biphenyls, and possibly lead from both long-range and local sources are recorded in Svalbard lake sediments for the past 30 to 40 years (Rose *et al*., 2004). Lakes in Svalbard are potentially particularly sensitive to acid deposition as they are located on deep permafrost and there is little groundwater interaction (Betts-Piper *et al*., 2004). Exceedance of critical loads for acidity (Lien *et al*., 1995) also suggests that acidification effects are possible, but no ecological impacts on diatom (Jones and Birks, 2004) or chironomid (Brooks and Birks, 2004) assemblages from the lake sediments have been observed. Trends in DI-pH values in five Svalbard lakes have been stable over the past few centuries. However, top–bottom analysis of chrysophycean stomatocysts showed marked shifts in assemblages in most of the lakes studied. Lakes on granitic bedrock, which are presently acidic, may have become more acidic over time. On the other hand, lakes on carbonate bedrock that are presently alkaline appear to have increased their pH in recent times. These changes may be related to atmospheric contamination from local and remote sources, but climate change may also play a role (Betts-Piper *et al*., 2004).

6.4.2.4. **Concluding comments on recent acidification**

Broad-scale recent acidification of arctic lakes is not apparent through DI-pH reconstructions. This may be partly because most of the lakes studied are not particularly sensitive to acidification or are not located in areas of high deposition. Also, there are no accurately dated high-resolution DI-pH reconstructions from the near vicinity of the local emission sources to allow a paleolimnological evaluation of acidification status in these lakes. However, within a few tens of kilometers from the local emission sources there is clear evidence of the effects of metal accumulation on the lake biota as a function of time, as indicated by the increased proportion of deformed diatoms and chironomids towards the core top. As documented in a recent Arctic-wide study (Smol *et al*., 2005), climate change is already affecting lakes and ponds and their biota in arctic areas. This is evident from the marked recent change in algal and zoological paleolimnological indicators that is inconsistent with atmospheric acidification or nutrient input but instead indicates the widespread impacts of climatic change (e.g., Douglas *et al*., 1994; Sorvari *et al*., 2002; Michelutti *et al*., 2003).

6.5. Interaction between acidification and other environmental issues

Both the causes and effects of air pollutants are closely linked to other environmental problems and human activities. Many of the traditional air pollutants and green-

house gases have common sources. They interact chemically and physically in the atmosphere and cause a variety of interrelated environmental effects at different spatial scales (Swart *et al*., 2004; Table 6.5). Politically these different air pollutants and greenhouse gases have been treated separately; they also have different spatial and temporal scales. Others have a more local deposition pattern (e.g., many heavy metals) while others are spread over hundreds of kilometers or even globally (sulfur, greenhouse gases). The different spatial scale of each pollutant and its effects makes georeferenced modeling necessary, as this can handle different scales and resolutions simultaneously (van Rompaey, 1995 and references therein). There is also a need to seek synergies in emissions controls for air pollution and climate change to gain economic and political benefits (Swart *et al*., 2004). Furthermore, the projected climate change-related alterations in temperature, wind patterns, and precipitation can change the routes of contaminant entry and the locations and amounts of deposition in the Arctic (AMAP, 2002; Macdonald *et al*., 2003; ACIA, 2004).

Table 6.5. Impacts of various substances emitted to the air (redrawn from Seip and Aunan, 2002).

		Regional impacts		Local impacts		
	Climate change	Acidification	Ground level ozone	Health	Vegetation	Materials
CO_2	x					
CH_4	x		x			
SO_2	x	x		x	x	x
NO_x	x	x	x	x	x	x
NH_3	x	x		x	x	x
NMVOC[a]	x		x	x	x	?
PM[b]	x	x		x	x	x

[a] non-methane volatile organic compounds; [b] particulate matter

Climate warming, acid deposition, the effects of toxic chemicals, and increasing exposure to ultraviolet (UV) radiation are all regarded as widespread problems in northern ecosystems. In the past, climate warming, acidification, and UV radiation were treated as if they had distinct and separate effects on ecosystems. It is now clear that these major stresses caused by man's alteration of the atmosphere cannot be studied in isolation (Schindler *et al*., 1996; Schindler and Curtis, 1997). The recent Arctic Climate Impact Assessment (ACIA, 2004) concluded that the total impact of contaminants, excess UV radiation, and climate warming is greater than the sum of its parts. Thresholds of tolerance of biodiversity or of the structural and functional attributes of ecosystems to changes in atmospheric stressors are largely unknown (Freedman and Beauchamp, 1998). Multiple stresses, for example, concomitant changes in the atmosphere and land use, make these thresholds even more difficult to define.

This section summarizes the possible interactions between acidification and other environmental issues in high-latitude aquatic ecosystems in two key sectors: climate change and UV radiation, and heavy metals/contaminants.

6.5.1. Interactions concerning climate change and UV radiation

6.5.1.1. Anticipated changes in climate

The overarching stress on ecosystems in the future will be global climate change, which is projected to be greatest in the Arctic due to various feedback mechanisms (Chapman and Walsh, 1993; Overpeck *et al*., 1997). The Arctic is highly likely to be warmer and moister in future. According to the Arctic Climate Impact Assessment (McBean, 2005), the average surface temperature in the Arctic is currently increasing by approximately 0.09 ºC per decade and it is probable that there was an increase in arctic precipitation of 1% per decade during the past century. The Arctic Climate Impact Assessment projected (Kattsov and Källén, 2005) that the annual precipitation will increase by 7.5 to 18.1% by 2071-2090, depending on the model used. The projected increase is generally greatest in autumn and winter and smallest in summer. Temperature is projected to increase by 2.8 to 4.6 ºC in the Arctic (north of 60º N) by 2071-2090. Climate change will influence water quality by altering the balance between the atmospheric, terrestrial, and aquatic processes in watersheds, and the effects of human resource use on these processes. Among the most important physical and chemical changes projected for freshwater ecosystems are: increasing water temperatures, thawing permafrost, changes in ice cover on rivers and lakes, and increasing levels of contaminants (ACIA, 2004).

6.5.1.2. Anticipated changes in hydrology and water quality

Effects of increasing temperature and moisture on water quality

Murdoch *et al*. (2000) summarized some of the potential water quality effects of increasing temperature and changing moisture conditions. If air temperatures increase, fewer arctic lakes and streams will freeze to the bottom and lakes will have an increased number of ice-free days. In both cases these changes will increase nutrient cycling and productivity. A shallower depth of freezing in arctic lakes during warm years has been shown to cause increased productivity through a lengthening of the growing season. If sufficient nutrient sources are available, lakes in arctic and alpine regions will also experience increasing productivity as a result of more frequent mixing and deeper thermoclines with increases in temperature. Longer thaw seasons will enhance decomposition and greenhouse gas releases from northern wetlands and peatlands. Increased moisture is predicted to decrease permafrost thawing compared to warm-dry conditions, but increased erosion from high runoff may yield greater nutrient, DOC, and sediment loads from thawing permafrost terrain.

Dissolved organic carbon is derived from terrestrial vegetation, especially wetland vegetation. Concentrations in high latitude lakes are low. In small boreal lakes DOC is the most important determinant of thermocline depth (Perez-Fuentetaja *et al*., 1999). Changes in the levels of DOC are likely to be one of the most important changes of climate warming in surface waters (Schindler, 2001). The effects of climate change on treeline lakes are likely to be strong since shifts in vegetation cover near the treeline will affect DOC in lake waters as well as hydrology (Rouse *et al*., 1997). The projected increase in supply of organic material and nutrients will enhance primary production

in arctic freshwaters dramatically (Schindler, 1997; Hobbie *et al*., 1999; Flanagan *et al*., 2003).

Thawing permafrost increases the flow into groundwater, which can lead to the disappearance of water bodies in some areas but an increase the area of wetlands and small ponds in others. Longer ice-free periods and the northward advance of vascular plants will increase evapo(transpi)ration, leading to lower water levels. On the other hand, increased precipitation and cloudiness may counteract this. However, lakes, ponds, and wetlands are more likely to dry out during summer (Wrona *et al*., 2005). Earlier ice break-up affects supplies of nutrients, sediments, and water quality, but increases the amount of harmful UV-radiation during spring. These multiple stressors will impact on the aquatic biota at a time when they are having to compete with new species moving north from lower latitudes.

pH changes in high-latitude and high-altitude lakes

Thawing permafrost and a deepening of the active layer are very likely to increase geochemical weathering and nutrient release (Wrona *et al*., 2005). Warmer wetter conditions will also favor erosion and greater runoff of weathering products. This could increase the buffering capacity of lakes against strong acids.

As previously described, climate change probably alters the chemical properties of high-latitude lakes, potentially influencing their susceptibility to anthropogenic atmospheric deposition. A small change in temperature can have a profound impact on aquatic environments, especially in areas where the duration of snow and ice cover has a dominant role in determining ecosystem functioning. It has been suggested that climate (e.g., temperature and precipitation) indirectly controls the pH of oligotrophic alpine lakes (Psenner and Schmidt, 1992) as well as poorly buffered arctic lakes (Wolfe, 2002). In the Arctic, this is explained by dissolved inorganic carbon (DIC) dynamics, which are affected by lake ice cover and primary productivity (Wolfe, 2002). Thus DIC regulates the pH of dilute lakes. Globally, lakes are supersaturated by carbon dioxide (CO_2) and under prolonged ice conditions this situation is exaggerated, which leads to a decline in pH. With a warmer climate and longer ice-free season CO_2 supersaturation is eliminated and enhanced algal production further reduces limnetic CO_2, shifting the balance in inorganic forms of dissolved carbon away from CO_2 and toward HCO_3, resulting in an increase in pH (Wolfe, 2002). If the catchment to lake area ratio is small (e.g., < 10) in-lake processes become especially important in determining the lake pH status. Otherwise changes in the catchment, such as vegetation, land use, erosion, and weathering, would mainly drive changes in lake water quality.

Climate change may also affect the seasonality of the stress. A rise in algal productivity under warmer conditions may lead to larger seasonal fluctuations in pH as organic matter production influences the redox potential and consequently the acid-base equilibrium (Psenner and Schmidt, 1992), thus acting as an additional stress on the biota with pulses of strong acids into lakes in spring (see section 6.3). The relationship between climate and pH is, however, dependent on the unbuffered character of remote high-latitude lakes, which allows the direct control of pH by DIC dynamics. In general, it seems that a warmer and moister future climate would increase the alkalinity of arctic lakes.

At present, water below the winter ice in subarctic lakes is warmer than in lakes further south due to the rapid ice over. This sustains efficient decomposition on the sediment surface and causes anoxia in lakes (especially mesopolyhumic lakes) at the end of winter. Climate warming may remove this problem by shortening the ice-on period and increasing wind mixing in shallow lakes. On the other hand, increased production and DOC concentrations in shallow lakes may strengthen thermal stratification and sedimentation of organic matter to the bottom, thus increasing oxygen demand. Deep lakes are projected to be more strongly stratified. Changing the redox situation near the bottom will affect the alkalinity of the lakes.

Multiple stressors and effects on biota

The combined effects of multiple stressors may be antagonistic or synergistic, i.e., smaller than expected or larger than expected, respectively (Folt *et al*., 1999). The cumulative impacts of anthropogenic stressors on ecosystems are especially worrying in relation to a loss of biodiversity and to changes in ecosystem functioning (Sala *et al*., 2000; Schindler, 2001; Vinebrooke *et al*., 2004). When the critical load for multiple environmental stresses is exceeded, the ecosystem may change abruptly. Sala *et al*. (2000) projected very little change in biodiversity in arctic areas if the interactions between driving factors (land use, climate, nitrogen deposition, biotic exchange and atmospheric CO_2) are synergistic, and much change if the interactions are antagonistic.

For example, acidification typically shifts phytoplankton communities towards larger dinoflagellates and filamentous algae, while suppressing cyanobacteria and smaller chrysophytes and larger eukaryotic algae that are less well adapted to higher temperatures, suggesting that acidification and environmental warming have synergistic negative impacts on phytoplankton. It has been hypothesized that since acidification favors smaller zooplankton species, such as rotifers and certain copepods, the effects of several other stressors on zooplankton will be reduced in acidified lakes. For example, smaller zooplankton species experience lower metabolic costs per capita than larger species during warming, and are less vulnerable to UV radiation, visually feeding vertebrate predators, and pesticides. On the other hand, some planktonic crustaceans, which are UV-tolerant due to their ability to produce photo-protective pigmentation, are likely to face an increased risk of predation by visually feeding planktivorous fish, especially if large populations of alien sportfish are introduced into clear lakes (Vinebrooke *et al*., 2004). In their experiments on multiple stress effects on daphnids, Folt *et al*. (1999) concluded that effects of toxins and low food supply would probably be enhanced by thermal stress. Hypothesized antagonistic and synergistic effects of anthropogenic acidification and other major abiotic and biotic stressors on phytoplankton and zooplankton are shown in Table 6.6.

6.5.1.3. **Recovery from acidification in surface waters**

There is good documentation of a large-scale chemical recovery process from surface water acidification in Europe and North America (Stoddard *et al*., 1999; Forsius *et al*., 2001; Evans *et al*., 2001; Skjelkvåle *et al*., 2001a). Recovery is also documented in the northern regions (section 6.1.2). Modeling studies based on current emission reduction

Table 6.6. Hypothesized antagonistic (+) and synergistic (-) effects of major abiotic and biotic stressors (first column) and anthropogenic acidification on phytoplankton and zooplankton (redrawn from Vinebrooke *et al*., 2004).

	Phytoplankton	Zooplankton	Rationale
Elevated temperature	-	+	Acid-sensitive cyanobacteria are thermophilic; small acid-tolerant zooplankton (e.g., rotifers) experience lower metabolic costs than large acid-sensitive cladocerans.
Ultraviolet radiation	+	+	Acid-tolerant species must also be UV-tolerant as they experience elevated UV exposure during lake acidification and loss of UV-attenuating DOC. Small acid-tolerant rotifers show higher UV-tolerance than other zooplankton.
Eutrophication	-	?	Acid-sensitive cyanobacteria are better competitors for nutrients than other groups of phytoplankton.
Toxins	+	+	Acid-tolerant, small zooplankton species are more resistant to contaminants than large species.
Predation	+	-	Large, acid-tolerant phytoplankton (e.g., cyanobacteria, dinoflagellates) are less edible; smaller zooplankton are more susceptible to predatory invertebrates like *Chaoborus*, while pigmented UV- and acid-tolerant zooplankton are easily detected by introduced fish.

plans project further chemical recovery (Jenkins *et al*., 2003; Wright *et al*., 2005). Uncertainties in these scenarios are mainly related to the effects of climate change and future behavior of nitrogen in the ecosystem. Other uncertainties are related to the biological response.

Present-day climatic conditions are commonly assumed in model projections of future scenarios. However, as previously discussed large changes in climate are anticipated for the Arctic and the direction and degree of change may significantly affect the dynamics of terrestrial and aquatic ecosystems. Skjelkvåle *et al*. (2003) identified four key climate-related factors that may influence recovery from acidification: (a) increased frequency and severity of sea-salt episodes, (b) increased frequency and severity of drought, (c) increased turnover of organic carbon, and (d) increased mineralization of nitrogen. Although most empirical evidence is from boreal or temperate regions, it is likely that these processes are also relevant for most arctic environments.

Increased frequency and severity of sea-salt episodes

The 'sea-salt effect' in surface waters is important in areas receiving substantial inputs of sea-salts, in particular coastal areas of Norway, the United Kingdom and the United States. The sea-salt effect may temporarily increase the acidity of runoff. However, a prerequisite for the lake and stream acidification effect to occur from sea-salt episodes is that the catchment soil is acidic. Recent climate forecasts project a dramatic increase in the North Atlantic Oscillation index (see section 3.7.3) over the next 80 years, implying that warm, westerly conditions in winter may become more prevalent. A greater frequency and intensity of sea-salt episodes may therefore be expected in coastal surface waters. Sea-salt episodes may have important regional biological effects. Massive regional fish kills were reported for the first time after the episode in southwestern Norway during winter 1993 (Barlaup and Åtland, 1996). The fish deaths occurred in moderately acidified systems that suddenly became extremely acid.

Increased frequency and severity of drought

In parts of North America, the reduction and storage of sulfate in wetlands, and its subsequent re-oxidation and release, have been shown to have a major impact on runoff water quality and hence recovery (Dillon and LaZerte, 1992; Jeffries *et al*., 2003). Immobilization and re-mineralization of sulfur within soil organic matter are both important in European soils. Both stores of sulfur are sensitive to drought. Relationships between sulfate pulses and drought associated with El Niño events have been shown to occur in lakes in Ontario, Canada (Dillon *et al*., 1997). In the UK, large flushes of sulfate were widely observed in streams following a drought in 1995 (e.g., Harriman *et al*., 2001). Climate-regulated sulfur retention and release represent 'noise' within an overall recovery trend. Release of stored sulfur will delay recovery where pools are large. Also, sulfate flushes following drought (particularly if droughts become more severe with climate change) may continue to generate acidic episodes, despite improvements in baseline water quality. These drought-driven episodes may be more extreme or frequent in future climate scenarios and may, like sea-salt episodes, contribute to a delay in chemical and biological recovery in surface waters.

Increased turnover of organic carbon

Regional trends of increasing DOC concentrations over the last two decades have been documented across substantial parts of northern and central Europe (Evans and Monteith, 2001; Skjelkvåle *et al*., 2001a,b, 2005; Evans *et al*., 2005; Vuorenmaa *et al*., 2006) and eastern North America (Stoddard *et al*., 2003). For Canada the picture is less straightforward (Jeffries *et al*., 2003). The widespread occurrence of these trends indicates a regional cause and various hypotheses have been put forward to explain them. It has been proposed that these increases may be coupled to change in potential drivers, such as increasing temperature (Freeman *et al*., 2001; Hejzlar *et al*., 2003), changes in hydrological regimes (Tranvik and Jansson, 2002; Hejzlar *et al*., 2003), increasing atmospheric CO_2 concentration (Freeman *et al*., 2004), airborne nitrogen enrichment in soils (Findlay, 2005), or decreasing sulfur deposition (Stoddard *et al*., 2003; Evans *et al*., 2005; Vuorenmaa *et al*., 2006). More research is needed to establish the relative significance of the various drivers in the different regions.

Elevated DOC concentrations in surface waters have raised concerns that terrestrial carbon stores may be becoming unstable, with unpredictable consequences for the global carbon cycle and with complex consequences for surface waters. Recovery from acidification along with decreasing acid deposition is being partially offset by increasing organic acidity. Increasing DOC concentrations may also cause increased buffering of changes in pH, increased water coloration, and decreased visible light and UV-B

penetration within the water column (see sections 6.5.1.2 and 6.5.1.4). While increased organic acidity may delay chemical recovery from acidification in surface waters, the other factors may influence biological recovery.

Increased mineralization of nitrogen

Additional uncertainties with regard to nitrogen processes relate to the influence of climate (short and long term) on nitrate leaching, which may alter the long-term trend, or simply add 'noise' to the anthropogenic signal. Because internal ecosystem cycling of nitrogen greatly exceeds system inputs and outputs, any disturbance of this cycle has the potential to completely obscure the relationship between nitrogen deposition and runoff. Both in the UK and the US, large pulses of nitrate have been observed in surface waters following severe winters possibly as a result of soil freezing (Mitchell *et al.*, 1996; Monteith *et al.*, 2000). The frequency of such pulses may change in future in response to altered climate. Results from the CLIMEX project, where ambient air and soil temperature was increased over three years, show increased leaching of inorganic nitrogen, probably due to increased mineralization and nitrification rates in soils (Wright and Jenkins, 2001). Continued high deposition of nitrogen is likely to increase nitrogen saturation and generate increased nitrate concentrations in runoff, thereby delaying recovery due to reductions in sulfur emissions. Increased temperature due to climate change may increase nitrate in runoff and thereby contribute to delay in recovery. The role of nitrogen in acidification and recovery remains uncertain.

6.5.1.4. Impacts of DOC changes on UV radiation in lakes

Climate effects on exposure to UV radiation were discussed extensively by Wrona *et al.* (2005). Humic substances strongly absorb UV radiation and act as a screen, thus protecting organisms from its detrimental effects. Under normal conditions all wavelengths of solar radiation in freshwater ecosystems, including UV radiation, are attenuated to some degree by DOC (Schindler, 1999) and long term variation in underwater UV irradiance is primarily controlled by the amount of dissolved organic material. Long lasting ice cover has a similar protective role, protecting organisms from UV radiation during the most intensive radiation period in spring. Interactions between climate change and UV radiation potentially modify the underwater UV radiation regime via: (a) changes in stratospheric ozone levels, (b) changes in snow- and ice-cover duration, and (c) changes in DOC levels in natural waters (Wrona *et al.*, 2005). A reduction in snow and ice cover on the surface of rivers, lakes, or oceans is likely to increase the exposure of many organisms to UV radiation (Weatherhead *et al.*, 2005). On the other hand, UV penetration in dilute lakes that are presently within catchments dominated by tundra and forest-tundra may be significantly reduced if climate warming induces a northward shift in the treeline or increases in the density of forest cover, leading to increased DOC inputs (Pienitz and Vincent, 2000; Figure 6.15). Lakes with very high catchment to lake area ratios are likely to be most affected by vegetation changes in treeline areas (Pienitz and Vincent, 2000). Export of humic substances is related to climate and to the balance between production and decomposition. However, climatic as well as deposition impacts on DOC export are under debate (Evans *et al.*, 2002; section 6.5.1.3).

Thus, both acid precipitation and climate warming may enhance the exposure of aquatic organisms to increased UV radiation, which is usually attributed only to depletion of stratospheric ozone (Schindler *et al.*, 1996; Gorham, 1998; Schindler, 1999). Levels of DOC decrease rapidly when lakes become acidified. Lower pH values lead to increased protonation of functional groups and cause the dissolved materials to become hydrophobic and to settle out of the water column onto bottom sediments. Increased photodegradation also seems to occur. Acidification, climate warming, and stratospheric ozone depletion may thus act together in a 'three-pronged attack' of UV radiation on aquatic systems, particularly in dry conditions (Schindler *et al.*, 1996; Schindler, 1999; Figure 6.15).

6.5.2. Interactions concerning heavy metals/contaminants

In principle, the atmospheric transport routes for acidic compounds should be similar to the pathways of other contaminants, such as heavy metals. Trace metals can catalyze the oxidation of SO_2 to sulfate by sunlight (Kellogg, 1995). Acid precipitation contains a variety of trace metals. However, the deposition pattern of heavy metals is different from SO_2 in that they (usually) deposit within

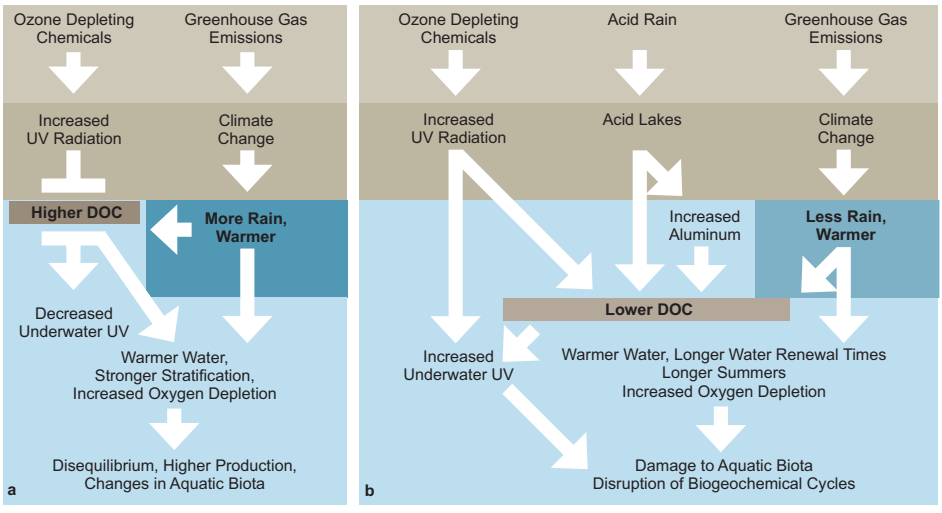


Figure 6.15. Climate warming will induce environmental changes in the Arctic that alter the exposure of aquatic organisms to the increasing levels of UV radiation caused by stratospheric ozone depletion: (a) a warmer wetter climate is likely to result in higher DOC levels in arctic lakes which, because humic substances strongly absorb UV radiation, will offset the effects of increased UV radiation from stratospheric ozone depletion (modified from Schindler, 1999), whereas (b) in arctic areas with a warmer drier climate the effects of stratospheric ozone depletion, acid rain, and climate warming can combine to increase the exposure of aquatic biota to UV radiation by causing a decrease in the DOC levels in lakes (redrawn from Schindler, 1999).

the immediate vicinity of the sources (mercury being a clear exception). Therefore, the interactive effects of metals and acid precipitation are especially visible locally near the emitters. The co-emissions of the smelters (base cations) may be adequate to prevent environmental acidification at the regional scale (e.g., Kashulina *et al*., 2003). The AMAP heavy metals assessment (AMAP, 2005) included little information on the potential interactions of heavy metals with other pollutants, for example sulfate, or the cumulative impacts of multiple pollutants. This section addresses potential interactions of heavy metal/contaminant pollution with other air pollution issues, including climate change. The complementary AMAP assessment on contaminant pathways (Macdonald *et al*., 2003) addressed recent global changes and their effects on the distribution of various contaminants.

6.5.2.1. **Processes in air**

Warming, increased precipitation, and thawing permafrost will increase the transport and deposition of contaminants to the Arctic. This was discussed in detail by Wrona *et al*. (2005) in relation to mercury and persistent organic pollutants. The timing of weather events also affects the transfer of contaminants to the Arctic. For example, snowfall occurring at the time of arctic haze would increase the transfer of contaminants to the ground at that time of year.

The northward movement of toxic metals and organic pollutants by cold condensation was confirmed by evidence from tree bark (Simonich and Hites, 1995) and lake sediments (Muir *et al*., 1995). Differential distillation, which pollutants undergo, allows them to be released in vapor form from warm areas and to re-condense in colder regions. However, Givelet *et al*. (2004) concluded that there is no evidence for the 'cold condensation hypothesis' and that the Arctic was not an important natural sink for mercury during pre-industrial times. Moreover, while the anthropogenic emissions of mercury are thought to have decreased by 30% in the past 20 years (Pacyna and Pacyna, 2002), there is evidence that mercury fluxes may have doubled over the past 100 years in the Arctic (Lockhart *et al*., 1995, 1998; Jackson, 1997) and that the average concentration of heavy metals has more than doubled on the Kola Peninsula over the last 20 years (Dauvalter, 2003). According to Givelet *et al*. (2004), springtime mercury depletion events are the chief mechanism for transferring atmospheric mercury to the arctic environment. Climate warming enhances mercury depletion events, which in turn increases the transfer of mercury into the food webs.

Synergistic interactions seem to occur between acidification, climate warming, and stratospheric ozone depletion that enhance the global mercury cycle (reviewed by Schindler, 2001). Global change and air pollutant levels influence the scale of mercury depletion (mercury transport and deposition from the air) (AMAP, 2002). The release of mercury to the atmosphere might be expected to increase as lakes become clearer (due to changes in DOC concentrations), particularly if increased methyl mercury is available due to warming lakes, and as reservoirs are constructed (Schindler, 1999).

6.5.2.2. **Processes in terrestrial areas**

Terrestrial processes have a key role in determining the impacts of heavy metals on surface waters. On the Kola Peninsula, direct exposure to SO_2 seems to be the main reason for ecosystem deterioration (Kashulina *et al*., 2003). The pine ecosystems there act as a biogeochemical barrier against metals by accumulating them in plant tissue and humus horizons (Goryainova and Nikonov, 1997). The function of forest ecosystems as a regulator of the cycles of these heavy metals (copper and nickel) is disrupted if the vegetation is lost (e.g., by acidification) and more toxic substances can then drain into watercourses.

Significant interactions between acidic rain and metal exposure were also observed in the lichen *Bryoria fuscesens* by Tarhanen *et al*. (1999). They established that although the metal load had a very important role in the decline of epiphytic lichen cover, acidic precipitation further disturbs the symbiosis between the photobiont and mycobiont of a lichen. Kashulina *et al*. (2003) concluded that the whole spectrum of emitted elements needs to be studied in the Kola region, in order to understand the effect of anthropogenic activities, including acidification.

6.5.2.3. **Processes in surface waters**

Changes in the timing of spring freshet, ice melt, and productivity are very likely to alter the efficiency of arctic lakes in capturing contaminants, for example mercury. Episodes of high contaminant levels are likely to occur in freshwaters when stored contaminants are released due to warming climate (Wrona *et al*., 2005). A wetter climate will expand the spatial extent of direct runoff to surface waters thus significantly increasing pollutant loads from point and non-point sources that are hydrologically isolated or filtered through groundwater aquifers under current flow conditions (Murdoch *et al*., 2000). Inundation of wetlands, riparian zones, and low-lying soils results in increased mobilization of trace metals and organic compounds from soils, increased mobilization and methylation of mercury, and greater anaerobic activity in saturated soils (sulfate reduction, denitrification) (Murdoch *et al*., 2000).

A change in flow patterns can alter the capability of a lake to receive and restore contaminants. At present, most of the contaminant load is carried through the lake surface layers under the ice before lake turnover and peak primary production and little is retained in the lake itself (Macdonald *et al*., 2000). In a warming climate the main runoff pulse may couple with lake mixing and primary production and hence cause more contaminants to be captured within the lake. In the case of acidifying compounds, lakes in catchments with thin soils, especially on slowly weathering bedrock, may thus face strong effects as the substances remain in the lake, rather than being carried rapidly through the lake.

Acidification and heavy metal contamination often work synergistically because the solubility of heavy metals in water increases as pH falls. As a result, heavy metals leach more quickly from contaminated soils in contact with acidic water. Dauvalter (1995) studied 15 lakes in Finnish Lapland. In four acidic lakes, the low pH had changed the geochemical cycling of potentially harmful metals by increasing desorption of metals from sediments back to water and/or decreasing the adsorption of metals onto settling sediment particles. Cadmium and nickel mobilization from sediments during acidification was observed by Nuorteva *et al*. (1987) and Fjeld *et al*. (1994).

Heavy metals are absorbed by plankton at the base of the food web and biomagnified to significant amounts at higher trophic levels. The influence of acidification and eutrophication on metal behavior must therefore be consid-

ered. It is well known that inorganic monomeric aluminum acts synergistically with pH to cause embryo mortality (e.g., Clark and Hall, 1985; Clark and LaZerte, 1985; Freda and McDonald, 1990; Blaustein *et al*., 2003). UV radiation also may affect the toxicity of contaminants (Wrona *et al*., 2005).

Water chemistry, especially acidity, and food web structure also affect mercury availability and uptake. Acidification can greatly enhance methylation, producing a higher proportion of bioavailable methylmercury (Miskimmin *et al*., 1992; Gilmour and Henry, 1991). Lake acidification is known to favor higher methylation to demethylation ratios, both directly and via effects on DOC (see references in Schindler *et al*., 1995). Climate warming alters the thermal conditions of lakes in a way that increases the methylation of mercury (Ramlal *et al*., 1993; Schindler *et al*., 1995). On the other hand, increased penetration of shortwave solar radiation due to decreases in DOC may cause increased conversion of methylmercury to elemental mercury, which is then released back to the atmosphere (Sellers *et al*., 1996). Once back in the atmosphere, mercury is susceptible to long-range transport and biomagnification in distant food chains (Schindler, 1999).

Future climate change, in interaction with other environmental problems, is thus thought to influence the distribution patterns and mobility of organic pollutants and toxic metals in freshwater systems and to lead to changes in the uptake and accumulation of these substances in freshwater food chains. This is one of the main hypotheses tested in an ongoing EU project EURO-LIMPACS (http://www.eurolimpacs.ucl.ac.uk/index.php).

Acidification of the oceans

Owing to increasing atmospheric CO_2 levels, CO_2 levels in the oceans have increased since pre-industrial times. Added CO_2 decreases the CO_3^{2-} (carbonate) ion concentration and makes the ocean more acidic. Ocean pH has decreased by 0.1 units, equivalent to a 30% increase in hydrogen ion concentration over the last two centuries (Caldeira and Wickett, 2003; The Royal Society, 2005). If global emissions of CO_2 continue to rise on current trends, then the average pH of the oceans could fall by 0.5 pH units by 2100 (a 0.5 unit decrease in ocean acidity means a three-fold increase in hydrogen ions). This could have substantial effects on the biological processes in surface oceans. Recent studies indicate that future undersaturation of aragonite and calcite, especially in polar oceans, may have potential biological impacts on calcifying organisms, especially shelled pteropods, the densities of which are high in cold water regions (Orr et al., 2005). Shell dissolution rates of pteropods increase in waters that have become undersaturated with aragonite (e.g., Byrne et al., 1984). Also, benthic calcareous organisms such as cold-water corals are in danger of becoming surrounded by water masses that are undersaturated with aragonite. Orr et al. (2005) suggested that some high latitude surface waters will probably become undersaturated within the next 50 years, leading to detrimental conditions for many organisms.

Chapter 7
Air Pollution and Health Impacts in the Arctic

Jon Øyvind Odland

In addition to the detrimental ecosystem effects of acidifying compounds and haze precursors described in previous chapters, elevated concentrations of air pollutants may also cause serious human health impacts. Concern over health effects is currently one of the most important factors driving international air pollutant emission reduction policies (Sliggers and Kakebeeke, 2004; UNECE, 2004b).

7.1. Major air pollutants of health concern

The sources, health effects, and vulnerable groups for major air pollutants are summarized in Table 7.1. It is very difficult to isolate the health effects of individual pollutants. It is more useful to consider each of the major pollutants as 'indicators' of the mixture of air pollution created by motor vehicles, home heating, and industry. Continuous monitoring of particulate matter, nitrogen oxides, ozone, and carbon monoxide is now established in the big cities (Simpson *et al.*, 2001), but these data provide only general estimates of actual exposures in individuals. Carbon monoxide is the only major air pollutant for which a biomarker of exposure (carboxyhaemoglobin in erythrocytes) is available.

Researchers are currently investigating the importance of the size and chemical composition of particles as a causal factor for cardiorespiratory effects (Brunekreef and Holgate, 2002). The focus is now on the very small particles; PM2.5 and PM1 (i.e., particles smaller than 2.5 µm or 1 µm, respectively). As very small particles penetrate further into lungs than larger particles, they are believed to be more strongly associated with adverse health effects. Diesel engine emissions contribute disproportionately to the very-small-particle fraction of urban air pollution (WHO, 1996).

The recent documentation of lung cancer as an effect of long-term exposure to urban air pollution (Pope *et al.*, 2002) highlights carcinogenic chemicals in the smallest air particles and in carcinogenic gases (e.g., benzene; benz[a]pyrene) as possible causal agents.

Carbon dioxide (CO_2), another air pollutant created by fuel combustion, has no direct health effects at the concentrations occurring in the ambient environment. However, it is the main 'greenhouse gas' causing global climate change (McMichael *et al.*, 1996) and, as such, indirectly contributes to the global health impact of such change. Efforts to reduce urban air pollution by reducing the use of cars would have the added benefit of reducing CO_2 emissions.

Health effects of the major air pollutants are listed in Tables 7.1 and 7.2. In any particular study, establishing

Table 7.1. Selected outdoor air pollutants and their effects on health (adapted from WHO, 2001).

Source	Known health effects	Contributing or potentiating factors	Vulnerable populations
Particulate matter			
Biomass and fossil fuel combustion in home heating, industry and motor vehicle engines; cigarette smoke	Upper respiratory tract irritation and infection; exacerbation of and increased mortality from cardiorespiratory diseases	Sulfur dioxide, sulfuric acid; cold, heat, humidity	Elderly people with respiratory and cardiovascular diseases; children with asthma
Sulfur dioxide and acid aerosols			
Fossil fuel combustion; metal smelting and petrochemical industries; home heating/cooking with coal	Throat irritation; exacerbation of cardiorespiratory diseases, including asthma	Exercise, particulates, asthma	People with respiratory diseases (e.g., children with asthma); elderly people with respiratory and cardiovascular diseases
Oxides of nitrogen			
Fuel combustion at high temperature (e.g., from vehicle engines, gas cooking and heating	Eye irritation; upper respiratory tract infection (especially in children); exacerbation of asthma; irritation of bronchi	Exercise, respiratory tract infection, asthma	People with respiratory diseases (e.g., children with asthma)
Ozone			
Reaction product of sunlight and vehicle pollutants; hydrocarbons and oxides of nitrogen	Eye and throat irritation; reduced exercise capacity; exacerbation of respiratory disease	Exercise, respiratory tract infection, asthma	People with respiratory diseases (e.g., children with asthma)
Carbon monoxide			
Biomass and fossil fuel combustion; cigarette smoke and vehicle exhaust	Headache, nausea, dizziness, breathlessness, fatigue, visual disturbance, confusion; angina, coma, death; low birth weight (after maternal exposure during pregnancy)	Coronary artery disease	People with ischemic heart disease
Lead			
Smelting; leaded petrol	In children: neuropsychological and cognitive effects. In adults: hypertension, classic lead poisoning	Other sources of lead; iron deficiency	Children, pregnant women
Other pollutants; 'air toxics'			
(hydrocarbons, aldehydes, other organic compounds, asbestos)	Eye irritation; lung cancer; asthma	Smoking, occupational exposures	Smokers, asbestos workers, people with asthma, children

Table 7.2. Adverse respiratory health effects of air pollution (American Thoracic Society, 2000).

- Increased mortality
- Increased incidence of lung cancer
- Increased frequency of symptomatic asthma attacks
- Increased incidence of lower respiratory tract infections
- Increased exacerbation of chronic cardiopulmonary or other diseases, reflected in various ways, including reduced ability to cope with daily activities, increased hospitalization, increased physician visits and medication, and decreased pulmonary function
- Reduction of FEV1 (forced expiratory volume in one second) or FVC (forced vital capacity) associated with clinical symptoms
- Increased prevalence of wheezing unrelated to colds, or wheezing on most days or nights
- Increased prevalence or incidence of chest tightness
- Increased prevalence or incidence of cough/phlegm production requiring medical attention
- Increased incidence of acute upper respiratory tract infections that interfere with normal activity
- Acute upper respiratory tract infections that do not interfere with normal activity
- Eye, nose, and throat irritation that may interfere with normal activities (e.g., driving a car), if severe

whether there is an association between air pollution and one or more of the effects listed depends on exposure level, the background health status of the population exposed, and their age.

7.2. Key epidemiological findings

Most recent epidemiological studies of air pollution and mortality have used time-series analysis to relate daily mortality rates to daily air pollution levels (on the same day or previous days). However, this approach cannot be used to ascertain whether increased mortality reflects a significant reduction in life expectancy (Brunekreef and Holgate, 2002). A few studies have documented associations between mortality and air pollution exposure over longer periods. The Harvard 'six cities' study involved a 14–16-year prospective cohort of more than 8000 adults in the United States, and the American Cancer Society (ACS) study collected data on over 500 000 people living in 51 different US metropolitan areas between 1982 and 2000 (Pope *et al*., 1995, 2002). These studies reported significant associations between annual average particle pollution levels (PM10 or PM2.5) and annual all-cause mortality rates: an average increase of 10 μg/m^3 of PM10 or PM2.5 was associated with a 3–4% increase in mortality. The ACS study also found a significant association between levels of PM2.5 and deaths due to cardiorespiratory diseases and lung cancer (Pope *et al*., 2002). A 'natural intervention' study using long-term exposure and effect data analyzed mortality in Dublin before and after coal-burning was banned in 1990 (Clancy *et al*., 2002). The annual average particle pollution level declined by 36% after the ban, while adjusted mortality rates decreased by 15.5% and 10.3% for respiratory and cardiovascular deaths, respectively.

A review of epidemiological studies concluded that there may be a relationship between acute ozone exposure and increased mortality, especially in elderly people (Thurston and Ito, 2001). However, the concurrence of high ozone levels with hot weather makes it difficult to separate the effect of heat from the effect of ozone on mortality.

Acute time-series studies have shown associations between particle pollution and daily hospital admissions, mainly for respiratory diseases (especially asthma and chronic obstructive pulmonary disease) but also for cardiovascular diseases (Morgan *et al*., 1998; McGowan *et al*., 2000; Petroeschevsky *et al*., 2001; Denison *et al*., 2001; Department of Environment, 2003).

7.3. The arctic perspective

The arctic perspective is, in general, the same as the global perspective but with the addition of point sources related to heavy industrial complexes, especially in the area of the former Soviet Union (Smith-Sivertsen *et al*., 1998). The associations between air pollution and human health in different arctic areas are briefly described in the recent AMAP assessment on human health (AMAP, 2003), and in more detail in many publications related to the Russian–Norwegian Health Group's reports (Smith-Sivertsen *et al*., 1998, 2001, 2003; Dotterud *et al*., 2001). The results reported were mostly very optimistic. Studies to date have been unable to show any significant health effects that are directly associated with emissions from the nickel refineries. No

Table 7.3. Reflections and predictions (after Kjellström *et al*., 2002).

Circa 1900
Cities being rapidly industrialized. Large amounts of sulfur dioxide and particulates being emitted in heavily populated areas from inefficient combustion of coal in power stations and industrial and domestic furnaces.
Air pollution and resultant lack of sunshine in industrialized areas causing widespread lung damage, high mortality, and an upsurge in diseases such as rickets in children. Indoor air pollution, from cooking with coal and wood, even worse for health than outdoor pollution.
Circa 2100 (a pessimistic view)
Increasing air pollution in developing countries.
Limited progress on motor vehicle emissions in developed countries.
Circa 2100 (an optimistic view)
Intelligent global stewardship of our natural resources has led to a major shift towards alternative energy sources such as wind power, solar energy, and fuel-cell engines for vehicles.
More wood burning to heat houses, as this creates less greenhouse gas emissions than fossil fuel.
Coal burning in super efficient and clean-burning electric power stations is still continuing in countries with large coal reserves (e.g., China, India, United States of America).
Many governments have put pressure on motor vehicle manufacturers to produce less-polluting cars.
High-quality public transport systems and advanced telecommunications systems have made daily commuting in private cars largely obsolete.
Private vehicles are mainly used for leisure activities.

reduction in lung function between exposed and non-exposed groups could be demonstrated (Smith-Sivertsen *et al*., 2001). Sensitization described by serum IgE-levels was more common in Russia than Norway, but the Russians did not report any more atopic diseases (Smith-Sivertsen *et al*., 2003). Atopic diseases seem to be even less prevalent in the Russian study groups than in studies of other northern European countries (Dotterud *et al*., 2001). Urinary nickel excretion was considerably higher in the Russian populations, but only partly associated with vicinity to the nickel smelters or to occupational exposure (Smith-Sivertsen *et al*., 1998). Differences in health status of the Norwegian and Russian border populations seem to be more associated with socio-economic conditions than to environmental pollution (Odland *et al*., 2004).

7.4. The shifting panorama

From a global and arctic perspective, many countries are at the same stage of industrial and urban development as Western European countries 50 to 80 years ago, when high levels of ambient air pollution from coal-burning were common. At the household level, promoting energy-efficient and less-polluting cooking stoves constructed from local materials would be an important step in reducing air pollution (Table 7.3). Switching the energy source for cooking to less-polluting kerosene, gas, and electricity is another solution, often out of reach for poor communities in the short term (Kjellström *et al*., 2002). Worldwide, a major change in priorities is needed to steer economic development towards low-pollution policies.

Chapter 8
Conclusions and Recommendations

Martin Forsius, John Derome, Lars R. Hole, Sirkku Manninen, Marjut Nyman, Patricia Quinn, Brit Lisa Skjelkvåle, and John L. Stoddard

8.1. Sources of acidifying pollutants and arctic haze precursors

AMAP assessments have clearly documented that air pollutants, including persistent organic pollutants, heavy metals such as mercury, and acidifying substances can reach the Arctic as a result of long-range transport from source regions in Europe, North America, and Asia. In relation to acidifying substances, the industrial areas of northern Europe and parts of Russia and the northeastern United States are responsible for most of the pollution exported to the Arctic. There are also significant sources of acidifying substances within the Arctic, including industrial sources (the metallurgical industry, power plants, oil and gas related activities) and diffuse sources associated with, for example, shipping. Emissions from natural sources within the Arctic (volcanoes, marine algae, and forest fires) are very difficult to quantify and almost impossible to project. However, the frequency, severity, and duration of boreal forest fires do appear to be increasing, possibly related to the influence of changing climate.

Nonferrous metal production remains the dominant source of emissions of acidifying gases to the atmosphere within the Arctic. Non-ferrous metal smelters located at Norilsk, and at Nikel, Zapolyarnyy, and Monchegorsk on the Kola Peninsula (all operated by MMC Norilsk Nickel) together account for 68% of the total 'harmful emissions' (including sulfur dioxide, dust, and nitrogen oxides) from non-ferrous metal production in Russia. These smelters are all located within or close to the Arctic. Sulfur dioxide emissions from these smelters have reduced substantially due to changes in production and better technology for controlling emissions. The main reductions have occurred since 1995 and have been considerably greater on the Kola Peninsula than at Norilsk.

It is recommended that improved information be obtained on emissions from arctic point sources, in particular for the non-ferrous metal smelters on the Kola Peninsula and at Norilsk.

Recent modeling results suggest that southeast Asia is not likely to be a major source of acidification-related atmospheric pollution at ground level in the Arctic. Efforts are, however, needed to confirm this finding. Future changes in atmospheric transport patterns due to climate change could also alter this conclusion. Europe (Northern Russia in particular) and to a lesser degree North America continue to be the main source regions for acidifying pollutants carried into the Arctic by long-range transport.

Shipping activities in arctic waters may increase substantially in the future. Projected reductions in sea-ice thickness and extent due to climate change have raised the possibility that shipping activities in arctic waters may increase substantially over the coming decades (as the navigation season lengthens and new sea routes open). Emissions associated with this increased activity are difficult to project.

The impacts of acidification from arctic shipping should be addressed, and future scenarios for emissions associated with this source should be included in this work.

The Arctic is thought to contain at least a quarter of the world's undiscovered petroleum resources: most of these in Russia, Alaska, Canada, Greenland, and Norway (offshore). A significant increase in arctic oil and gas activity can be expected, including activity in offshore areas.

The impacts associated with oil and gas related activities on acidification and arctic haze in the Arctic should be assessed together with the assessment of the impacts of oil and gas development.

8.2. Trends in air concentrations and deposition

Ice core drillings on Svalbard show that significant changes in atmospheric pollution within the Arctic have only occurred since the beginning of the industrial era, indicated by increased levels of non-sea-salt sulfate, nitrate, acidity, fly ash, and organic contaminants. Acidifying pollutants in rain, snow, dust, and gases have been monitored regularly at purpose-built stations in some parts of the Arctic, mostly since the 1980s. Fennoscandia has several background monitoring stations, while the vast Siberian region and the Canadian Arctic and Alaska have relatively few. Some of the stations have now generated time series datasets that are long enough to show whether concentrations are increasing, decreasing, or staying the same over time.

8.2.1. Air and precipitation

Sulfate concentrations measured at those air monitoring stations with the best air quality time series in the High Arctic (Alert, Canada; and Ny-Ålesund, Svalbard) and at several monitoring stations in subarctic areas of Europe (Oulanka, Finland; Tustervann, Svanvik, Karasjok/Jergul, Norway; Bredkälen, Sweden) show decreasing trends since the 1990s. Although there are relatively few background stations within the Arctic measuring sulfate concentrations, most record a decrease in concentrations since the 1990s. These observations are supported by modeling results. There are few signs of significant trends in precipitation for the period studied. However, expected future occurrence of rain events in both summer and winter will result in increasing wet deposition in the Arctic.

For nitrate and ammonia the pattern is unclear, with increases at some stations and decreases at others. The increasing trends in nitrate are particularly apparent in recent years indicating a decoupling between the trends in sulfur and nitrogen. Time series of sulfur and nitrogen concentrations in precipitation at Norilsk since 1990 do not show any significant trends.

In general, sulfur deposited originates from local point sources and long-range transport. In the AMAP region, high levels of deposition only occur close to large point sources in the vicinity of the Nikel and Monchegorsk smelters on the Kola Peninsula and in Norilsk in northwestern Siberia. With the exception of areas affected by emissions from the industrial centers, average atmospheric deposition loads are much lower in arctic Russia than in more southerly parts of the country. Background sulfate levels in precipitation decrease from west to east across the Russian Arctic. There is a similar pattern for background levels of nitrate in precipitation across the Russian Arctic. A trans-Arctic snow study found no overall relationship between pH and levels of sulfate from human activities.

Short-term episodes of winds carrying very high concentrations of air pollutants can result in pulses of pollution entering the arctic environment. Studies at Finnish background stations showed that the five worst days of the year can bring 20 to 30% of the annual sulfate load of bulk deposition.

8.2.2. **Model projections**

The decreasing trends in levels of acidifying pollutants observed at many sites throughout the Arctic are supported by model results. Models indicate that mean concentrations of sulfur oxides and total sulfur deposition within the Arctic almost halved between 1990 and 2000. These decreasing trends are supported by empirical data from the several monitoring stations around the Arctic (see above). The modeled results for airborne oxidized nitrogen are similar to those for sulfur. The models also confirm earlier findings that emissions in Eurasia continue to make the greatest contribution to acid deposition within the Arctic.

Model projections based on future emissions scenarios indicate that the decreasing trends observed between 1990 and 2000 are likely to level off and that only small reductions in concentrations and deposition can be expected after 2020, even if maximum feasible reductions in emissions are achieved. Although further recovery and continuing improvement in the acidification status of the Arctic can be expected during the period until 2020, this is dependent on the implementation of existing international agreements to reduce emissions of acidifying substances. The Gothenburg Protocol is the most important agreement in this connection.

All arctic countries should be encouraged to ratify the UN ECE LRTAP protocol to Abate Acidification, Eutrophication, and Ground-level Ozone (the 'Gothenburg Protocol') and to support its implementation.

8.3. **Arctic haze**

Based on measurements of sulfate aerosol and light scattering by aerosols, the amount of arctic haze reaching the Arctic was either relatively constant or decreasing between the 1980s and early 1990s. Levels of sulfate aerosol have decreased since the late 1990s and the indications are that levels are still decreasing. In contrast, nitrate aerosol appears to be increasing in Alert (Canada), with an unclear trend at Barrow (Alaska). However, trend analysis on light scattering data collected since the late 1990s indicates an increase in the amount of haze reaching the Alaskan Arctic (Barrow). There is also evidence of increasing light absorption, due to black carbon aerosols at Barrow (Alaska) and Alert (Canada). Forest fires are a possible source of this black carbon. The frequency, severity and duration of boreal forest fires seem to be increasing and the pollution plumes from these summer fires can extend over vast areas. Boreal forest fires may even dominate the black carbon budget in the Arctic in years of strong burning.

8.4. **Effects**

8.4.1. **Human health**

Epidemiological studies indicate that differences in health status of the Norwegian and Russian border populations are more associated with socio-economic conditions than environmental pollution. Studies to date have been unable to show any significant health effects that are directly associated with emissions from the nickel refineries.

8.4.2. **Terrestrial ecosystems**

The present deposition of acidifying compounds from long-range, transboundary transport of anthropogenic emissions at lower latitudes does not appear to be a threat to terrestrial ecosystems in most of the Arctic.

The non-ferrous metal smelters are responsible for the vast majority of acidification-related effects in plants and soils within the European Arctic. Information on the effects of emissions from the smelters on the Kola Peninsula on terrestrial ecosystems is accumulating rapidly and is more accessible to the international scientific community than information on the situation in the Norilsk region, which is less detailed and published mostly in Russian.

Acidified soils on the Kola Peninsula mostly occur immediately around the smelters and coincide with the areas where the vegetation has been completely destroyed. Strongly acidic precipitation only falls within about 30 km of the smelters; outside this zone lower sulfur dioxide levels and the presence of alkaline particles in the atmosphere prevent the precipitation becoming acidic. Sparse data on soil microorganisms show that adverse effects are also concentrated in restricted areas around the smelters but are more associated with excessive heavy metal deposition than soil acidification.

Outside the area immediately around the smelters, there is no clear evidence of soil acidification due to sulfur dioxide emissions (and subsequent deposition of acidifying compounds) on the Kola Peninsula. The lack of widespread soil acidification despite high sulfur dioxide emissions appears to result from the simultaneous emission of alkaline fly ash from the power plants and the apatite fertilizer complex, the low interception of acidifying compounds by the sparse cover of coniferous trees, and the low rate of conversion of sulfur dioxide to sulfuric acid in the Arctic. Soils in the Norilsk area are not considered acid sensitive.

In the European Arctic there are direct effects of sulfur dioxide on trees, dwarf shrubs, and epiphytic lichens. Effects on vegetation, some of which are evident at a regional level, are due to changes in air quality and in most cases are not related to changes in soil acidification. Direct effects of sulfur dioxide include visible leaf damage, decrease in needle life span in conifers, and elevated sulfur concentrations in plant tissues. The observed changes in plant community structure (species composition and coverage) also indicate the indirect effects of sulfur and nitrogen deposition. In terms of their effects on plants, it is difficult to differentiate between the effects of acidifying air pollutants and elevated heavy metal levels in soils. Large quantities of heavy metals are emitted by the non-ferrous metal smelters.

The most likely explanation for the changes observed in vertebrate populations near Monchegorsk is habitat deterioration. Winter mortality may be especially high in the contaminated habitats. Changes in insect populations are mostly due to changes in host plant abundance and physiology, accompanied by a decline in some natural predators, and to changes in habitat structure.

The critical loads of acidity for terrestrial ecosystems have not been exceeded in Canada north of 60° N. In the European Arctic the widespread exceedance of critical loads of acidity in 1990 should have almost disappeared by 2010 if the agreed emissions reduction policies are fully implemented. However, the critical levels of sulfur dioxide and critical loads for acidity in highly sensitive forest ecosystems are expected to be exceeded locally and regionally near the non-ferrous metal smelters. (In this context, 'critical level' refers to the atmospheric concentration above which direct adverse effects will occur on plants, while 'critical load' refers to the maximum amount that can be deposited on an ecosystem without causing that ecosystem to become damaged). It must be remembered however, that non-exceedance of critical loads/levels does not necessarily mean that there will be no environmental change.

8.4.3. **Freshwater ecosystems**

Anthropogenic acidification of lakes is only of concern in areas with both sensitive geology and elevated levels of acid deposition. Because the areas with the highest levels of acid deposition, for example Norilsk, are less geologically sensitive the biggest impacts on lake chemistry and biology have mostly been observed in small sensitive ecosystems in local areas.

Available lake chemistry data provide irregular and incomplete coverage of the Arctic. Only a few of the lakes monitored in the Canadian Arctic are acid sensitive; these are mostly on Baffin Island or the central mainland. There are no lake chemistry data for large areas of the North American Arctic. Acid sensitive lakes are scattered throughout the European Arctic but are most abundant in northern Fennoscandia and the Kola region. Acidification by natural organic acids is important in some lakes.

Chemical monitoring data show that lakes in the Euro-Arctic Barents region are showing clear signs of a regional-scale recovery from acidification caused by long-range transported sulfur. Lakes close to the pollution sources on the Kola Peninsula are showing the clearest signs of recovery. There are insufficient data to make similar conclusions about regional-scale biological recovery.

Assessments of biological effects of acidification in arctic surface waters are largely based on sparse and isolated data. There are far more chemical data available than biological data. The best biological effects data are from the impacted areas in northeastern Norway and Finland – outside this area it is impossible to draw robust conclusions about biological effects in surface waters. It is important to note that lakes subjected to acidic inputs, even those with measured decreases in acid neutralizing capacity, alkalinity, and pH, have not necessarily been acidified to the point where measurable damage to the biota can be observed.

Biological recovery is occurring in at least two isolated areas. Both sites are directly downwind of point sources on the Kola Peninsula. A comparison of minnow populations in lakes and streams in the Vätsäri area of northeastern Finland between 1993 and 2000 and increasing abundance of the acid-sensitive cladoceran *Daphnia longiremis* since 1999 in Lake Dalvatn, an acid-sensitive lake in Finnmark, both indicate improvement in the acidification status of these waters. A clear recovery of the brown trout population of lake Otervatn in Eastern Finnmark has occured.

Changes in the microscopic fossil record in lake sediments indicate that most arctic lakes have experienced very slow natural acidification over the last 10 000 years. These paleolimnological studies do not support the hypothesis of recent and widespread acidification of arctic lakes due to sulfur pollution from the smelting and mining industries. Many of the lakes studied are outside areas of high acid deposition or are not particularly sensitive to acidification. Within a few tens of kilometers from the local emission sources there is however clear evidence of the effects of metal accumulation on the lake biota as a function of time, as indicated by the increased proportion of deformed specimens of diatoms and chironomids towards the top section of the sediment. The algal and zoological paleolimnological indicators also show that the global warming is affecting the lakes and ponds in the Arctic.

Sulfur deposition has important effects on the severity of acidic episodes in spring. Data on streams of the Kola North showed that in the periods of pH depression during floods (acidic episodes) the total concentration of toxic aluminum in water increased by 50 to 88%. A model based assessment on data from streams in northern Sweden indicated that reduced emissions of acid precursors have generated significant improvements in the surface water chemistry during episodes associated with spring runoff, and that episodic acidification decreased by between 40 and 80% between 1990 and 1999. It is also likely that expected future reductions in acid deposition will further improve the spring flood acidification situation in the northern regions.

In 1990, critical loads of acidity for surface waters in the European Arctic were exceeded almost everywhere. Implementation of agreed emission reduction policies will reduce the area in which critical loads are exceeded and, to a greater extent, the amount by which the critical loads

are exceeded. But even with the implementation of the maximum feasible emissions reductions (the 'MFR 2020' scenario) there are still very likely to be areas where the critical loads for surface waters are exceeded. The required deposition reduction is up to 3.2 kilograms of sulfur per hectare per year in the most exposed area of the Kola Peninsula.

8.5. Links between acidification, arctic haze, and other environmental issues

The effects of haze aerosols on the arctic climate are complicated by feedbacks between aerosols, clouds, radiation, sea ice, and vertical and horizontal transport processes. Whether the pollutant aerosols cause an overall warming or an overall cooling is not yet known. Changes in the light scattering and absorbing properties of the haze directly affect the amount of solar radiation passing through the haze.

The causes and the effects of air pollutants are closely linked to other environmental problems. For example, climate change, changes in exposure to ultraviolet radiation, the effects of toxic chemicals, and land-use. Complex interactions between these factors can strongly affect ecosystem responses to air pollutants.

Recent studies indicate that if global emissions of carbon dioxide continue to rise at current rates, then the average pH of the surface oceans could fall by 0.5 pH units by 2100. This could cause detrimental conditions for calcifying organisms, especially in high latitude surface oceans, within the next 50 years.

Future AMAP assessments should view acidification and arctic haze in a wider context. Particularly in relation to toxic pollutants, particulate matter, and climate change and climate feedback effects (i.e., reduced snow albedo as a result of increasing black carbon deposition).

Chemical and biological variables should be monitored and studies conducted to assess the risk of acidification of high latitude surface oceans due to increased carbon dioxide emissions.

8.6. Gaps in knowledge and recommendations concerning monitoring and research needs

8.6.1. Geographical gaps

New information from the areas around Norilsk has provided valuable insight into the sensitivity, and potential vulnerability of this region. The few samples taken, however, are not sufficient to enable an overall assessment of the geographic extent or trends in environmental effects in the Norilsk region.

Continued effort should be made to collect data and information for the Norilsk region. This should involve both MMC Norilsk Nickel and the relevant local and regional authorities.

While Fennoscandia has several background air monitoring stations for acidification parameters, most areas of the Arctic have few, if any, background air monitoring stations. The vast regions of Siberia east of the Urals, Alaska, and the Canadian Arctic are covered by relatively few stations, and some of these are being closed. A better distribution of background monitoring/research stations will enable a more complete and accurate circumpolar assessment to be made of acidification and arctic haze, as well as of other air pollution issues.

Additional background monitoring stations for air and precipitation chemistry should be established in northern Canada and Alaska. More regional and background air monitoring and precipitation chemistry stations should be established in the Russian Arctic. These background air monitoring stations should be extended to form 'multi-purpose' monitoring stations that monitor acidification parameters together with, for example, persistent organic pollutants and heavy metals, ozone and UV, and_precipitation chemistry and relevant meteorological and ecosystem effects parameters. This will help to ensure cost-effective monitoring at remote sites that are expensive to operate and maintain, and will allow data to be assessed in a more multidisciplinary context.

8.6.2. Data availability

Both AMAP and EMEP have benefited from a close cooperation with respect to monitoring levels of acidifying pollutants in air and precipitation within the European Arctic. The new EANET (Acid Deposition Monitoring Network in East Asia) initiative represents an opportunity to develop similar cooperation in relation to monitoring in the Far East of Asia, including assessment of the acidification potential and arctic haze impacts of long-range transported air pollutants from southeast Asia. Programs addressing acidification issues also exist in North America, although their coverage of arctic areas is limited.

AMAP should continue to develop a better linkage with programs such as EMEP and EANET and with appropriate national experts (particularly in Russia and North America) to extend the geographical areas and time series datasets available for use in AMAP assessments.

Improved efforts should be made to ensure that relevant data, both existing and future, from North American monitoring stations are reported to the AMAP database at NILU according to agreed procedures. Including more North American data in the AMAP database would enable a better understanding of hemispheric pollutant transport to the Arctic and would be a useful source of data for groups engaged in hemispheric transport modeling.

Monthly data from Russian precipitation chemistry monitoring sites should be reported to the AMAP database at NILU according to agreed procedures. This will enable a better understanding of acid deposition at the regional level within the European Arctic.

8.6.3. Trends in air and precipitation

Long-term monitoring is extremely important, even when trends are decreasing. For example, background monitoring stations that measure sulfate have mostly recorded a decrease in concentrations since the 1990s. In contrast, many stations have recorded an increasing trend in nitrate

over recent years. This indicates a decoupling between the trends in sulfur and nitrogen. There is also evidence of increasing light absorption due to black carbon aerosols at Barrow (Alaska) and Alert (Canada). Natural climate fluctuations (such as the North Atlantic Oscillation) affect the concentrations of acidifying air pollutants and levels of arctic haze. Time series datasets need to be long enough to resolve the effects of natural climate fluctuations and to identify the underlying trends. Measurements linked to chemical transport models are particularly important for helping to establish the causes of trends.

Monitoring of trends in acidic species in the Arctic and other high latitude stations should continue and sulfur dioxide should be added to the list of chemicals monitored at Barrow (Alaska).

Monitoring of aerosol composition, light scattering, and light absorption should continue at Barrow (Alaska) and Alert (Canada), as well as at other locations to better define changes in arctic haze over time.

8.6.4. **Pathways**

Recent modeling studies yield conflicting results as to whether southern Asia is a source of pollutants to the Arctic. Rapid industrialization in this region has caused an increase in the emissions of many air pollutants and these increases are likely to continue. The transport pathways to the Arctic are likely to be affected by the projected changes in the climate system.

Co-operation with relevant international organizations, such as EMEP, the LRTAP Convention Task Force on Hemispheric Transport of Air Pollutants, and EANET should be enhanced to obtain more precise data on emissions from southeast Asia and to investigate the possible impact of these emissions on the Arctic.

Further research is needed into pollution transport pathways to the Arctic. In particular, the potential effects of future changes in transport pathways due to climate change should be assessed.

8.6.5. **Effects monitoring and research**

The work on ecosystem effects has clearly shown that it is very difficult to differentiate the effects caused by different pollutants, for example effects caused by acidification, poor air quality, and toxic metals. Data on effects on biota are sparse and isolated and key chemical data for cause-effect assessments are often incomplete. Episodic effects are very important in the Arctic due to the extreme climate and the large emission point sources. Surface waters are generally more sensitive to inputs of acidifying pollutants than soils. For this reason there is a greater and more extensive exceedance of critical loads to lakes than soils. Recovery will initially be observed as decreases in acidity followed by recovery of aquatic biota. The latter is the ultimate proof for successful action.

More biological data from sites close to the main emission point sources and less affected sites as reference are needed to assess effects and verify recovery. Systematic studies (monitoring programs) should be conducted on key species and variables that indicate changes in the structure and function of terrestrial ecosystems (including possible recovery) in very polluted areas as well as in 'control' areas. Similarly, more biological data on effects and trends in surface waters should be collected in the most impacted areas of the Arctic. The effects of acidic episodes on biota should be studied with high temporal resolution. In these contexts, AMAP should co-operate and co-ordinate with other relevant programs and projects, such as CAFF and CBMP in obtaining biological data. Systematic lake surveys should be conducted in acid-sensitive regions of the Arctic.

Sites for effects monitoring studies should be located close to air quality, deposition, and climate monitoring stations ('multi-purpose stations', see above). The study sites should be representative of the different terrestrial ecosystem types (tundra, mountain birch, and coniferous forest ecosystems) because these differ in their sensitivity to air pollution impacts.

Limnological and accurately-dated high-resolution paleolimnological studies are needed in lakes near emission sources in the environmentally sensitive areas of the Kola Peninsula and Norilsk to assess recovery.

8.6.6. **Models**

Modeling is one of the most important tools available for gaining insight into the possible pollution status of the extensive areas of the Arctic where the observational networks are absent or poorly developed. Models also allow investigation of scenarios for future trends, and for linkages between contaminant pathways and, for example, climate change.

Measurements of sulfur species, nitrogen species, and black carbon should be used to further validate and improve existing air quality and deposition models. Measurements should be conducted during field campaigns to improve models, for example determining dry deposition velocities during summer.

There is a need to integrate aircraft, ground-based, and long-term data sets for use in three-dimensional arctic climate models designed to evaluate climate forcing by arctic haze. Existing data are primarily long-term ground-based measurements of aerosol composition and, at a few sites, scattering and absorption. To estimate aerosol radiative forcing in the Arctic requires additional data on the vertical and horizontal distributions of composition and amount of the haze as well as its variability. Such data are only derived from periodic aircraft measurements and long-term ground-based observations.

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Abbreviations

*	of non-marine origin
1-D	one-dimensional
3-D	three-dimensional
Al	aluminum
AMAP	Arctic Monitoring and Assessment Programme
ANC	acid neutralizing capacity
AO	Arctic Oscillation
AOD	aerosol optical depth
BC	black carbon / base cation
C	carbon
Ca	calcium
C-horizon	parent material of soil
CH_4	methane
Cl	chloride
CL_{Ac}	critical load of acidity
CLE scenario	emissions scenario based on current legislation
CO	carbon monoxide
CO_2	carbon dioxide
Cu	copper
DEHM	Danish Eulerian Hemispheric Model
DIC	dissolved inorganic carbon
DI-pH	diatom-inferred pH
DOC	dissolved organic carbon
EANET	Acid Deposition Monitoring Network in East Asia
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
K	potassium
LRTAP Convention	Convention on Long-Range Transboundary Air Pollution
MFR scenario	emissions scenario based on maximum technically feasible reductions
Mg	magnesium
Mn	manganese
N	nitrogen
Na	sodium
NAO	North Atlantic Oscillation
N_2O	nitrous oxide
NH_3	ammonia
NH_4	ammonium
NH_4-N	ammonium nitrogen
Ni	nickel
NILU	Norwegian Institute for Air Research
nmVOC	non-methane volatile organic compounds
NO_2	nitrogen dioxide
NO_3	nitrate
NO_3-N	nitrate nitrogen
NO_X	nitrogen oxides
NO_X-N	nitrogen oxides nitrogen
NSR	Northern Sea Route
O-horizon	organic horizon of soil
S	sulfur
SO_2	sulfur dioxide
SO_4	sulfate
SO_4-S	sulfate sulfur
SO_X	sulfur oxides
UN ECE	UN Economic Commission for Europe
UV	ultraviolet
Zn	zinc

AMAP
Arctic Monitoring and Assessment Programme

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