Assessing the Impacts of Long-Range Sulfur and Nitrogen Deposition on Arctic and Sub-Arctic Ecosystems

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Abstract For more than a decade, anthropogenic sulfur (S) and nitrogen (N) deposition has been identified as a key pollutant in the Arctic. In this study new critical loads of acidity (S and N) were estimated for terrestrial ecosystems north of 60° latitude by applying the Simple Mass Balance (SMB) model using two critical chemical criteria ($Al/Bc = 1$ and $ANC_{le} = 0$). Critical loads were exceeded in large areas of northern Europe and the Norilsk region in western Siberia during the 1990s, with the more stringent criterion ($ANC_{le} = 0$) showing the larger area of exceedance. However, modeled deposition estimates indicate that mean concentrations of sulfur oxides and total S deposition within the Arctic almost halved between 1990 and 2000. The modeled exceeded area is much reduced when currently agreed emission reductions are applied, and almost disappears under the implementation of maximum technically feasible reductions by 2020. In northern North America there was no exceedance under any of the deposition scenarios applied. Modeled N deposition was less than $5 \text{ kg ha}^{-1} \text{ y}^{-1}$ almost across the entire study area for all scenarios; and therefore empirical critical loads for the eutrophying impact of nitrogen are unlikely to be exceeded. The reduction in critical load exceedances is supported by observed improvements in surface water quality, whereas the observed extensive damage of terrestrial vegetation around the mining and smelter complexes in the area is mainly caused by direct impacts of air pollution and metals.

Keywords Arctic · Critical loads · Exceedance · Acidity · Nitrogen · Modelling

INTRODUCTION

Since 1970s it has been recognized that remote parts of the Arctic are influenced by air pollution from anthropogenic

activities at lower latitudes. Although arctic haze was first noticed by whalers and explorers during the 1750s, it was not until the mid-1970s that its anthropogenic origins were established (Rahn et al. 1977). Arctic haze is a mixture of sulfate (SO_4^{2-}), particulate organic matter and nitrogen (N) compounds, as well as trace elements, and contaminants from industrial emissions. There are few but significant sources of air pollution within the Arctic; production of copper, nickel, and other non-ferrous metals from sulfur (S)-bearing ores, are the largest sources of acidifying compounds with emissions dominated by the Nikel, Zapolyarny, and Monchegorsk smelter complexes on the Kola Peninsula and Norilsk on the Taymir Peninsula in north-western Siberia (Hole et al. 2006, 2009). Norilsk is the largest single S emission source in the Arctic region. Consequently, the regions surrounding the smelter complexes (in northern Russia and north-eastern areas of Norway and Finland) are those where most Arctic air pollution impact studies have been undertaken (AMAP 1998, 2006).

During the 1970s and 1980s, scientific evidence on the impacts of air pollution led to international negotiations to control emissions of compounds that undergo long-range transport. The UNECE Convention on Long-range Transboundary Air Pollution and EU National Emission Ceiling Directive, primarily based on effects-based ‘critical loads’, have been key international instruments in this respect. Critical loads are defined as ‘a 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). Sulfur emission reductions have been one of the great environmental ‘success stories’; SO_2 emissions have been reduced by ~67% in Europe between 1980 and 2000, with reductions of almost 90% in many countries (EMEP 2004).

Box 1

The critical load of sulfur (S) and nitrogen (N) acidity is not a single value, but a trapezoidal function in the N–S-deposition plane characterized by three quantities, $CL_{\max}(S)$, $CL_{\min}(N)$, and $CL_{\max}(N)$ that are illustrated in Fig. 1. Under several simplifying assumptions (Posch and De Vries 1999; De Vries and Posch 2003; UBA 2004) the maximum critical load of S, $CL_{\max}(S)$, is derived as (in $\text{eq ha}^{-1} \text{a}^{-1}$):

$$CL_{\max}(S) = BC_{\text{dep}} - Cl_{\text{dep}} + BC_w - Bc_u - ANC_{\text{le,crit}}$$

where the subscripts dep, w, u, and le refer to deposition, weathering, uptake, and leaching, respectively, BC are base cations ($BC = Bc + Na$, $Bc = Ca + Mg + K$), Cl are chloride ions. $ANC_{\text{le,crit}}$ is the critical (or acceptable) leaching of ANC (Acid Neutralizing Capacity) defined as:

$$ANC_{\text{le}} = HCO_{3,\text{le}} + Org_{\text{le}} - H_{\text{le}} - Al_{\text{le}}$$

where HCO_3 are bicarbonate ions, Org organic anions, H protons, and Al aluminum. As long as $N_{\text{dep}} \leq CL_{\min}(N) = N_i + N_u$, where N_i is the immobilized N in the soil, all deposited N is consumed by uptake and immobilization, and S can be considered alone. For $N_{\text{dep}} > CL_{\min}(N)$ the critical load is a function of both S and N deposition. And in case of zero S deposition the maximum critical load of N is derived as:

$$CL_{\max}(N) = CL_{\min}(N) + CL_{\max}(S)/(1 - f_{\text{de}})$$

where f_{de} is the fraction of the net N input which is denitrified ($0 \leq f_{\text{de}} < 1$). Critical loads are linked to ecosystem vulnerability via a chemical criterion which is then used to calculate $ANC_{\text{le,crit}}$ (via chemical equilibria). Here we investigate two criteria: (a) the widely used molar ratio of [Al] and [Bc] in soil solution ($Al/Bc = 1$), and (b) zero leaching of ANC ($ANC_{\text{le}} = 0$). In most systems the latter criterion is (much) stricter, since it requires that the net acidity input is balanced by base cation weathering and (non-anthropogenic) deposition.

The exceedance of a critical load for a given sulfur or nitrogen deposition, which indicates the increased risk for ‘harmful effects,’ is calculated according to the procedure depicted in Fig. 1. For mapping purposes, individual exceedances are aggregated, mostly within deposition grid cells, to the so-called *average accumulated exceedance* (AAE) (Posch et al. 2001):

$$AAE = \frac{A_1 \cdot Ex_1 + A_2 \cdot Ex_2 + \dots + A_n \cdot Ex_n}{A_1 + A_2 + \dots + A_n}$$

where Ex_i is the exceedance and A_i the area of the i -th ecosystem ($i = 1, \dots, n$). If there is non-exceedance for an ecosystem (i.e., depositions fall into the gray area in Fig. 1), then $Ex_i = 0$. In addition, the area where critical loads are exceeded—mostly expressed as percentage of the total ecosystem area—is often used to evaluate deposition scenarios.

The Ministers of the Arctic States established the Arctic Monitoring and Assessment Programme (AMAP) in 1991 to ‘monitor the levels of anthropogenic pollutants in the Arctic environment’ (Arctic Environmental Protection Strategy 1991). Since 1998, acidic deposition and arctic haze have been identified as key pollutants. Consequently, two assessments of acidification and arctic haze have been carried out under the AMAP framework (AMAP 1998, 2006). This study presents new (harmonized) critical loads of acidity (S and N) for terrestrial ecosystems (forests and semi-natural vegetation) in the arctic and sub-arctic regions with latitude north of 60° , and exceedance estimates (i.e., areas where deposition is greater than the critical load) for the years 1990, 2000, 2010, and 2020. Furthermore, the potential eutrophying impact of N deposition is discussed.

MATERIALS AND METHODS

Critical Loads for Terrestrial Ecosystems

Critical loads of acidity were estimated with the so-called Simple Mass Balance (SMB) model (Box 1), which links deposition to a chemical variable (the ‘chemical criterion’) in the soil, or soil solution, associated with ecosystem effects. The violation of a specific value (the ‘critical limit’) for the chemical criterion is associated with ecosystem damage. In this way deposition(s) are linked to ‘harmful effects.’ Consequently, the selection of the chemical criterion (and its critical value) is a crucial step in

deriving a critical load. The most widely used soil chemical criterion is based on the ratio of aluminum and base cations (Al/Bc ; where Bc is the sum of calcium (Ca^{2+}), magnesium (Mg^{2+}), and potassium (K^+)) in soil solution. Uncertainties in the SMB model have been widely discussed (Skeffington 2006; Li and McNulty 2007). However, one of the largest sources of uncertainty in critical load calculations is the relationship between the critical limit and the ‘harmful effect’ (Løkke et al. 1996; Reinds

Fig. 1 Critical load (CL) function of sulfur (S) and acidifying nitrogen (N) for an ecosystem defined by $CL_{\max}(S)$, $CL_{\min}(N)$, and $CL_{\max}(N)$, derived from site properties and the chosen chemical criterion. The gray area below the CL function denotes deposition pairs resulting in an ANC-leaching below $ANC_{\text{le,crit}}$ and thus non-exceedance of critical loads. The *critical load exceedance* is calculated by adding the N and S deposition reductions needed to reach the critical load function via the shortest path ($E \rightarrow Z2$): $Ex = ExN + ExS$. Z1 and Z3 indicate alternative deposition reduction paths to achieve non-exceedance

et al. 2008). Further details on the methodology for calculating critical loads can be found in Posch and De Vries (1999), De Vries and Posch (2003), and UBA (2004).

In the current study, critical loads of acidifying S and N deposition for terrestrial ecosystems were estimated using an Al/Bc = 1 mol/mol. The Al/Bc ratio has been documented to be an indicator for damage to the fine roots of vegetation (Sverdrup and Warfvinge 1993). However, due to the large uncertainty and criticisms associated with the Al/Bc ratio (Løkke et al. 1996; Cronan and Grigal 1995), critical loads were also estimated using acid neutralizing capacity ($ANC_{le} = 0$) as the critical chemical criterion (Reinds et al. 2008; Holmberg et al. 2001) (see also Box 1). This criterion preserves the long-term balance of base cations in soils. Exceedance of critical loads, estimated as the ‘average accumulated exceedance’ (AAE, see Box 1), was calculated under four emission–deposition scenarios: (i) 1990 emissions, (ii) 2000 emissions, (iii) Current Legislation (CLE) emissions for 2010, and (iv) the Maximum technically Feasible Reduction (MFR) emissions for 2020.

Data for Critical Loads

The determination of critical loads of acidity for forests and semi-natural vegetation requires (spatial) input data describing climatic variables (e.g., temperature and precipitation), base cation deposition and weathering, nutrient uptake and N transformations (e.g., immobilization, denitrification, see Box 1). A combined map with the information to derive these input data was generated by overlaying maps of land cover, soils, climate, and forest growth regions. The methodology is described in detail in Reinds et al. (2008) for Europe and northern Russia; modifications for North America are described below.

Land Cover and Soil Data

The distribution of forests and semi-natural vegetation north of 60° was described using three land cover databases: the harmonized land cover map for the European region (Slootweg et al. 2005) (250 m resolution); the Global Land Cover 2000 project map for northern Russia (Bartholome et al. 2002) (1 km resolution); and the US Geological Surveys ‘North America Land Cover Characteristics’ database (Anderson et al. 1976) (1 km resolution). Greenland and some Eurasian islands were excluded from the current study as they are almost entirely covered by continuous ice cover.

The distribution of soil types was described using the European Soil Database (JRC 2006) (scale 1:1,000,000) for the European and Russian regions (see Reinds et al. 2008 for further details), and the Soil Landscapes of Canada (SLC; scale 1:1,000,000) for Canada. Furthermore, soil

properties were vertically and spatially weighted for each soil type in the SLC to derive an average value for each mapping unit (for consistency with the European soil database). A similar approach was used for Alaska using the US Department of Agriculture’s State Soil Geographic (STATSGO) database (scale 1:250,000).

Meteorology and Hydrology

The annual water flux through the soil at the bottom of the rooting zone (soil percolation) is required to compute the leaching of compounds. For Europe and northern Russia, the rooting zone was set at 50 cm, except for lithosols, which were assumed to have a depth of 10 cm. For North America, soil depth was set to a maximum of 50 cm (or less based on the aggregated soil horizons). The leaching rate was estimated from meteorological data and soil properties. Long-term (1961–1990) average monthly temperature, precipitation, and cloudiness were derived from a high resolution (10' × 10') European database (Mitchell et al. 2004). For sites outside Europe (North America and Russia), a coarser 0.5° × 0.5° global database from the same source was used.

Evapotranspiration was calculated following (Prentice et al. 1993): potential evapotranspiration was computed from precipitation, temperature, and cloudiness. The effect of snow cover was included by simulating accumulation and melting of a snow layer using temperature and precipitation. Actual evapotranspiration was then computed using a reduction function for potential evapotranspiration based on the available water content in the soil (Federer 1982). Soil water content was in turn estimated using a simple ‘bucket-like’ model that used water holding capacity and precipitation data.

Base Cation Deposition and Weathering

Base cation deposition for Europe was obtained from an atmospheric dispersion model (Van Loon et al. 2005). For northern Russia and northern America, Ca^{2+} deposition was derived from a global mineral dust model (Tegen and Fung 1995) using soil Ca^{2+} content, as described in Bouwman et al. (2002). A consistent map of Bc deposition between Europe and Russia was generated using relationships between Ca^{2+} deposition and Mg^{2+} and K^{+} deposition at monitoring stations in Europe (Reinds et al. 2008). A similar approach was used for North America; the relationship between observed Bc deposition in eastern Canada and modeled Ca^{2+} deposition was used to estimate a consistent field for Alaska and northern Canada.

Total base cation ($BC = Bc + \text{sodium } (Na^{+})$) weathering was estimated as a function of parent material and texture, corrected for temperature (UBA 2004). For Europe

and Russia, parent material was obtained from the European Soil Database. A similar approach was used for North America; base cation weathering rates were derived from percent clay and substrate type for each soil horizon, and (vertically and spatially) weighted to derive an average value for each mapping unit. The weathering rates of Ca^{2+} , Mg^{2+} , K^+ , and Na^+ , from the total BC weathering, were estimated as a function of clay and silt content (Reinds et al. 2008).

The ranges in estimated base cation sources vary significantly between the three regions (northern America, Europe, and Russia); in general depositions and weathering are lower in Canada, with the largest depositions in Europe, and largest weathering rates in Russia (Fig. 2).

Nutrient Uptake, Nitrogen Immobilization, and Denitrification

The net nutrient uptake of Bc and N by forests was computed by multiplying the estimated annual average growth of stems and branches with their elemental contents of Bc and N (Jacobsen et al. 2002). For semi-natural vegetation, net uptake was set to zero assuming no net removal. Forest growth in Europe was derived from the European Forestry Institute database (Schelhaas et al. 1999), which provided measured growth data for about 250 regions for various species and age classes. Growth was assessed by computing the area-weighted average growth over all age classes for each combination of region and tree species group. Forest growth in Russia was estimated from data in Alexeyev et al. (2004) (see also Reinds et al. 2008). For

Alaska and northern Canada, the net nutrient uptake was set to zero under the assumption that no removal of Bc or N occurred, i.e., there is limited or no forest harvesting. However, uptake of Bc and N often balance each other and the net effect of ignoring nutrient uptake on the critical loads of acidity is thus limited.

The denitrification fraction for the entire study area was computed as a function of the soils' drainage status (Reinds et al. 2001) and varied between 0.1 for well drained soils to 0.8 for peaty soils. The long-term net N immobilization was estimated as a function of temperature (Hettelingh et al. 2008) derived from data on long-term soil build-up of N in Sweden (Rosén et al. 1992).

Emissions and Deposition Scenarios

Anthropogenic emission scenarios were developed for 1990, 2000, 2010 (CLe), and 2020 (MFR) using several global or large-scale inventories (EDGAR database, GEIA, RETRO, and EMEP; see Hole et al. 2009 for further details). The CLe scenario reflected the current perspectives of individual countries on future economic development, taking into account already agreed emission control legislation. The MFR scenario reflected full implementation of presently available emission control technologies, while maintaining the projected levels of anthropogenic activities.

The total (wet and dry) deposition of S and N to terrestrial ecosystems north of 60° was estimated using the Danish Eulerian Hemispheric Model (DEHM) system, which has been used to study the transport and dispersion of air pollution in the Arctic since 1991 (Christensen 1997).

Fig. 2 Cumulative distribution functions for estimated base cation ($\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+$) deposition (blue lines) and weathering (brown lines) for the entire study area and three sub-regions (Northern America, Europe, and Russia)

The model has a horizontal resolution of $150 \text{ km} \times 150 \text{ km}$ (extension of the EMEP grid), 20 vertical layers and covers most of the northern hemisphere. The DEHM includes 63 chemical species and more than 120 reactions that describe the chemistry of sulfur oxides, nitrogen oxides, reduced nitrogen, volatile organic compounds, and ozone. Further details on DEHM and the deposition scenarios used in the current study are given in Hole et al. (2009). That paper also compares model results with measurements and discusses their uncertainty.

RESULTS AND DISCUSSION

Critical Loads for the Arctic

The lowest critical loads (and most sensitive regions) occur in northern Fennoscandia and eastern Siberia (based on 5th percentiles of the maximum critical load of S, $CL_{\max}(S)$, on a $50 \text{ km} \times 50 \text{ km}$ grid resolution; Fig. 3). In northern North America, the lowest critical loads occur in eastern Canada. The general pattern of sensitivity (based on $CL_{\max}(S)$) is broadly similar to the empirical sensitivity map in Kuylenstierna et al. (2001). In general, low critical loads are found in areas with low weathering rates associated with coarse soils on acidic parent material. The critical loads of acidity are based on a large amount of information; the total number of records (ecosystems), for

which critical loads were estimated is ca. 700,000. Further, despite the large number of data sources (maps), the critical load of acidity for terrestrial ecosystems is broadly consistent north of 60° . The choice of critical chemical criteria had a significant impact on the critical load; $Al/Bc = 1$ is less stringent than $ANC_{le} = 0$. The median critical load value is about $700 \text{ eq ha}^{-1} \text{ a}^{-1}$ for the Al/Bc criterion compared to $300 \text{ eq ha}^{-1} \text{ a}^{-1}$ for the ANC_{le} criterion (Fig. 3), the latter resulting in lower critical load values everywhere.

In Reinds et al. (2008) six different soil critical chemical criteria for calculating critical loads of acidity for terrestrial ecosystems (for Europe and northern Asia) were evaluated. The ANC_{le} criterion gave the lowest median critical load values and the Al/Bc criterion represented the other extreme. These two criteria thus give a good indication of the critical load range associated with the selection of the limit value. While the Al/Bc criterion protects (in theory) against fine root damage, the ANC_{le} criterion more strictly preserves existing soil base cation pools. The $Al/Bc = 1$ is the most widely used criterion (Reinds et al. 2008; Posch et al. 2005); as a consequence the current study focused on this criterion; however, the ANC_{le} criterion may be more suitable for protecting arctic systems. For the study area, $Al/Bc = 1$ is equivalent to a negative ANC_{le} for most ecosystems (Reinds et al. 2008).

Critical loads of acidity for regions with extreme climate should be treated with caution as the presence of frozen

Fig. 3 Critical load of acidity (5th percentile of $CL_{\max}(S)$) for areas north of 60° using two different critical limits: $Al/Bc = 1$ (left) and $ANC_{le} = 0$ (right)

Fig. 4 Areas with monthly mean temperatures $<0^{\circ}\text{C}$ for a period of 8–12 months (1961–1990 average meteorology)

soils during most of the year might violate some of the assumptions used in the critical load calculations. While there is an obvious gradient in climatic conditions, there is no absolute boundary where the critical load approach is not feasible. The area with shallow permafrost reported in FAO-UNESCO (2003) corresponds well with areas with an average monthly temperature below zero for at least eight months of the year. This area (monthly mean temperatures less than

zero for 8–9 months year^{-1}) potentially indicates where the current critical loads approach is not feasible (Fig. 4).

Deposition in the Arctic

The highest S deposition values in the study region occurred around the Norilsk Nickel smelters in western Siberia (see open circle in Fig. 5). This smelter complex is the largest single S emission source in the Arctic with an annual SO_2 emission of 1847 kt in 2003, exceeding the total annual emissions of any EU country. There are large point sources in the Kola Peninsula, but emissions have been substantially reduced since 1990 (50–80%). These reductions are much larger than at Norilsk, where emission decreased by only about 13% between 1992 and 2003 (Hole et al. 2006). Higher S depositions also occur in south-eastern Finnish regions and the St. Petersburg area. The highest N deposition values were observed in southern Fennoscandia (Fig. 5); mainly due to long-range transport from central Europe.

The DEHM results indicated that both the total S deposition and air concentration almost halved between 1990 and 2000 in the Arctic region (Hole et al. 2009). This is primarily due to emission reduction measures implemented during recent decades (EMEP 2004). The two future emissions scenarios (CLE and MFR) predicted a small decrease in both concentration and deposition compared with 2000 values. Although the total emissions in the MFR scenario are a factor of two less than those in the CLE scenario they had only a minor effect on the total concentration and deposition of S and N in the Arctic. This

Fig. 5 Total deposition of sulfur and nitrogen in 2010 (CLE scenario) calculated with the DEHM system. The white circle indicates the Norilsk mining and smelter complex ($69^{\circ}29' \text{ N}$, $88^{\circ}12' \text{ E}$)

is because the largest potential reductions in SO_2 emissions are in China and SE Asia; and these regions have little influence on the pollution of the Arctic (Stohl 2006). In contrast, emissions in Europe and Russia contribute most to the deposition in the Arctic and therefore future changes in these emissions will have the greatest impact in this region (Hole et al. 2009).

Exceedance of Critical Loads of Acidity

In 1990, critical loads of acidity were exceeded in large areas in northern Europe and the Norilsk region in western Siberia (Fig. 6, Table 1). However, the ecosystem areas showing exceedance were significantly reduced assuming implementation of currently agreed emission reduction measures (CLe 2010; Fig. 6); further, for $\text{Al/Bc} = 1$ the exceeded area almost disappeared under the MFR 2020 scenario (Table 1). As critical loads of acidity for $\text{ANC}_{\text{le}} = 0$ were much lower than for $\text{Al/Bc} = 1$ (Fig. 3), the exceeded area was larger. For $\text{ANC}_{\text{le}} = 0$ more than 50% of the ecosystem area was exceeded in the northern European region (Nordic countries) during 1990 and 10% of the area was still exceeded in 2020 under the MFR scenario (Table 1); some exceedance also remained in northern Russia. It should be recognized that the exceedance maps (Fig. 6) display the average accumulated exceedance (AAE, see Box 1), but give no information of the ecosystem area exceeded, which also diminished with declining deposition.

In North America there was no exceedance (north of 60°) under any deposition scenario (i.e., not even in 1990).

Table 1 Exceedance of critical loads (% ecosystem area exceeded) in the different regions assuming four different emission–deposition scenarios and two different chemical criteria and critical limits for the critical load calculations

Ecosystem area	Criteria and limit	Northern Europe 735,000 km^2	Northern Russia 6,618,000 km^2	Northern America 4,047,000 km^2
1990	$\text{Al/Bc} = 1$	17.6	4.9	0.0
2000	$\text{Al/Bc} = 1$	2.0	0.4	0.0
2010 (CLe)	$\text{Al/Bc} = 1$	1.4	0.3	0.0
2020 (MFR)	$\text{Al/Bc} = 1$	1.0	0.2	0.0
1990	$\text{ANC}_{\text{le}} = 0$	54.4	15.4	0.0
2000	$\text{ANC}_{\text{le}} = 0$	23.2	5.2	0.0
2010 (CLe)	$\text{ANC}_{\text{le}} = 0$	15.1	4.5	0.0
2020 (MFR)	$\text{ANC}_{\text{le}} = 0$	10.7	2.5	0.0

The total mapped ecosystem area in the regions is also given

Although critical loads are generally comparable between the three main regions (Northern Europe, Russia, and America, Fig. 3), deposition of S and N is much smaller in North America (Fig. 5). The minimum critical load is about $130 \text{ eq ha}^{-1} \text{ a}^{-1}$ and the maximum rates of S and N deposition are about $30\text{--}40 \text{ eq ha}^{-1} \text{ a}^{-1}$ (not at the same location). Thus not even the combined S and N deposition exceed critical loads in northern North America.

Although most of the regions with frozen soils (Fig. 4) do not show critical load exceedance (Fig. 6), climate

Fig. 6 Average accumulated exceedance (see Box 1) of the critical loads of acidity (with chemical criterion and critical limit $\text{Al/Bc} = 1$) north of 60° in 1990 and 2010 (CLe scenario)

predictions indicate that warming will be most pronounced in the Arctic (ACIA 2005). This will not only reduce the area of permafrost, but also will reduce ice cover and lead to increased and new shipping traffic and oil, gas and mineral exploration in the Arctic. This may result in greater depositions of S and N in the region, which are not considered in the deposition scenarios used in this study. Also an increase in wet deposition is expected in the Arctic due to climate change (Hole et al. 2009).

Potential Eutrophying Impact of N Deposition

Nitrogen is the limiting nutrient for plant growth in many natural and semi-natural ecosystems. Most plants are adapted to nutrient-poor conditions, and can only survive or compete successfully on soils with low N availability. In addition, the N cycle in ecosystems is complex and strongly regulated by biological and microbiological processes, and thus many changes may occur in plant growth, inter-species relationships and soil-based processes as a result of increased N deposition. The eutrophying impact of N deposition on the European scale is also evaluated using critical loads and their exceedance as a basis for integrated assessment modeling (Hettelingh et al. 2007). To this end information on empirical critical loads for N deposition (based on field studies and experiments) has been collected (UBA 2004; Achermann and Bobbink 2003). The estimated empirical critical loads for boreal forests are in the range of 10–20 kg N ha⁻¹ a⁻¹ (5–10 kg N ha⁻¹ a⁻¹ in low deposition areas; 1 kg N = 71.4 eq). For heathland, scrub, and tundra habitats the range is 5–25, for grasslands and tall forb habitats 5–25, and for mire, bog, and fen habitats 5–25 kg N ha⁻¹ a⁻¹. Based on experimental and monitoring data for northern Sweden (Nordin et al. 2005), it was concluded that the empirical critical load for boreal forest understorey vegetation should be at most 6 kg N ha⁻¹ a⁻¹. Higher values caused increased growth of *D. flexuosa* and decreasing abundance of *V. myrtillus* and *V. vitis-idaea*.

Nitrogen deposition in the Arctic region is low (Fig. 5), and annual deposition under CLe exceeds 5 kg N ha⁻¹ a⁻¹ in only 1% of the study area. Further, deposition is less than 1 kg N ha⁻¹ a⁻¹ in about 80% of the region (Fig. 7). Given this low deposition, empirical critical loads of nutrient N are hardly exceeded; accordingly, critical loads of nutrient N using a mass balance approach were not assessed in the present study.

Empirical Evidence of Damage and Recovery in the Arctic

Long-term measurements of air quality, deposition, and ecosystem effects in the Arctic region confirm the diminishing impact of air pollutants. Sulfate concentrations

Fig. 7 Cumulative distribution of total nitrogen deposition in the study area for the CLe 2010 scenario (1 kg N = 71.4 eq)

measured at air pollution stations in the high Arctic and at several monitoring stations in the sub-arctic areas in Europe show decreasing trends since the 1990s. For nitrate and ammonia the pattern is unclear with increases at some stations and decreases at others. In contrast, time series of S and N concentrations in precipitation at Norilsk do not show any significant trends (Hole et al. 2009; AMAP 2006).

Chemical monitoring data indicate that lakes in the Barents region are showing clear signs of a regional-scale recovery from acidification owing to reductions in long-range transported S. Lakes close to pollution sources on the Kola Peninsula show the clearest signs of recovery. However, there are insufficient data to make similar conclusions about regional-scale biological recovery (Skjelkvåle et al. 2006; Vuorenmaa and Forsius 2008). There is no evidence of any temporal trends in soil acidity in the Kola region (Derome et al. 2006).

There are large damage zones of vegetation around the smelter complexes in Nikel and Monchegorsk in the Kola region. The size of the area with dead forests was over 400 km² at the Nikel and Varanger area and between 400 and 500 km² at Monchegorsk. It has been estimated that the total vegetation area affected by air pollution exceeds 5000 km² around Nikel (Derome et al. 2006; Tømmervik et al. 1995, 2003). Acidified soils on the Kola Peninsula mostly occur immediately around the smelters and coincide with the areas where the vegetation has been completely destroyed. 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). 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, as well as the low rate of conversion of sulfur dioxide to sulfuric acid in the Arctic (Derome et al. 2006).

The major observed effects on vegetation in the Kola region, some of which are evident at a regional level, are mainly due to changes in ambient air quality and metal concentrations in soils. Direct effects of SO₂ include visible leaf damage, decrease of needle life span in conifers, and elevated S concentrations in plant tissue. The observed changes in plant community structure (species composition and coverage) also indicate the effects of indirect soil-mediated processes. 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 (Derome et al. 2006; Forsius et al. 2006).

There is much less empirical evidence available from the Norilsk area. Basalts with a basic chemical composition characterize the bedrock in the Norilsk area and the adjacent Putorana mountains. The buffering capacity, as indicated by Ca²⁺ and Mg²⁺ 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 has been found to be as high as 6.4. However, the copper and nickel values of the organic layer are at the same level as in Monchegorsk (1370–2820 mg/kg) and the uppermost part of the soil profile is strongly affected by the emissions of S and heavy metals (Derome et al. 2006; Salminen et al. 2004).

Critical load and exceedance estimates represent a long-term steady-state situation and thus ecosystem damage is unlikely to occur immediately in exceeded areas. Dynamic biogeochemical modeling is required to assess the time-scales of ecosystem damage and recovery (Aherne et al. 2008). However, the recovery observed in sensitive lake catchments studied in northern Europe (Skjelkvåle et al. 2006; Vuorenmaa and Forsius 2008) clearly confirm the decreasing risk of damage by soil acidification shown by the critical load exceedance calculations in the present study. The exceedance calculations indicate rather modest exceedances of critical loads around the large point sources at the Kola Peninsula and Norilsk, even during the 1990s (Fig. 6). This is consistent with the above empirical evidence, indicating limited soil acidification and more direct impacts of changes in air quality and heavy metals on the terrestrial vegetation in these regions.

CONCLUDING REMARKS

The critical loads of S and N acidity for terrestrial ecosystems north of 60° indicate that the most sensitive

regions occur in northern Fennoscandia and eastern Siberia. In northern North America, the lowest critical loads generally occur in eastern Canada. In contrast, the highest S deposition values in the study region occurred around the Norilsk mining and smelter complex in western Siberia, but there are also large point sources on the Kola Peninsula. Model results indicate that mean concentrations of sulfur oxides and total S deposition within the Arctic almost halved between 1990 and 2000. These decreasing trends are supported by empirical data from monitoring stations around the Arctic.

In a policy context, exceedances of critical loads are more important than the information on the sensitivity of the different regions per se. Critical loads were exceeded in large areas in northern Europe and the Siberian Norilsk region in 1990. However, the ecosystem area showing exceedance was much reduced when the currently agreed emission reduction measures were modeled; the exceeded area is estimated to disappear under the MFR 2020 scenario (Al/Bc = 1). Due to the lower deposition of S and N in northern North America, there is no exceedance north of 60° under any of the deposition scenarios. Furthermore, empirical critical loads for the eutrophying impact of N are unlikely to be exceeded in the study area owing to the low N deposition.

Emission reductions have decreased the area and magnitude of the critical load exceedances in the study region. The observed extensive damage of terrestrial vegetation around the mining and smelter complexes in the area is mainly caused by direct impacts of air pollution and metals. The available empirical data and the estimates of exceedances provide a consistent view on the recovery of sensitive ecosystems north of 60°. However, the large changes in climate predicted for the Arctic region may potentially cause an increase in air pollution emissions in this sensitive region owing to the increased exploitation of natural resources and shipping as well as changes in precipitation patterns. Therefore, the continued assessment of the risk of impacts to the most sensitive ecosystems will remain an important issue.

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