

ESTIMATION OF CRITICAL LOADS OF ACIDITY FOR LAKES IN NEW ENGLAND AND EASTERN CANADA

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Summary

The New England Governors and Eastern Canadian Premiers (NEG/ECP) adopted the *Acid Rain Action Plan* in June 1998, and issued a series of action items to support its work toward a reduction of SO₂ and NO₃ emissions in northeastern America. One of these action items was to prepare an updated map of lake sensitivity for the NEG/ECP area. The Water quality monitoring workgroup opted to work with the critical load concept, which convey both the lake sensitivity issue and the deposition effects on lake bodies. Combined sulfur and nitrogen critical loads and deposition exceedances were computed using Henriksen's Steady-State Water Balance model (SSWC). This model was used to assess lake critical loads in several other provinces of Canada. As expected, results show that critical loads in the NEG/ECP area are very low in general over low-carbonate content bedrock, and higher in sedimentary terrain. They are often exceeded in many parts of Eastern Canada and northeastern United States. The results indicate that regional low target loadings are needed to reduce damages to lakes or help recover from acidification.

Keywords : Critical load, sensitivity, acid rain, deposition, SSWC model, lakes

Introduction

Acid rain has been in the news for the past 40 years, and yet we are still debating to what extent emissions has to be reduced to efficiently protect ecosystems. This question is often brought up by policy-makers. Back in the 70's and 80's, the main question was about proving the reality of acid rain and its impacts on ecosystems. When scientist agreed acid rain was a reality, the policy-makers wanted to know if recovery from acidification was possible and what should be the deposition threshold under which lakes could be protected from acidic deposition. In Canada, the concept of a unique target load for sulfate in wet deposition to protect moderately sensitive waters (20 kg/ha/year) was proposed. With emission reductions under way, scientists questioned the adequacy of this target load. In the 90's, numerous studies showed that acidification was occurring under lower deposition fields, and that 20 kg/ha/year was not enough to protect sensitive waters. Moreover, nitrogen was not even considered in the equation. In 1997, the Acidifying Emissions Task Group, under the National Air Issues Coordinating Committee of the Canadian Council of Ministers of the Environment (NAICC/CCME), made a point that the earlier

target load was not enough and that we should embrace the critical load concept that can be scientifically-derived for individual lakes and forests. Based on that, models were applied and the results show that additional emission reductions were needed to reach critical loads in Eastern Canadian lakes. Again, nitrogen was left out of the equation. The 2000's may now be a stepping stone. Both scientists and policy-makers are now aware that sulfur and nitrogen both need to be considered in critical load assessments.

The critical load (CL) concept varies slightly according to authors, but one definition provided by the United Nations Economic Commission for Europe (UN ECE) and reported by Nilsson and Grennfelt (1988) is still the most widely accepted by the scientist and policy-makers community : *“The highest deposition of acidifying compounds that will not cause chemical changes leading to long term harmful effects on ecosystem structure and function according to present knowledge”*.

Critical load models used for assessment of lake critical loads have been around for the last 15 years. Aherne *et al.* (2000) identified 3 approach levels to critical loads assessment : 1) Level 0 for semi-quantitative assessment approaches based on sensitivity classes of soils and waters, 2) Level 1 approaches based on steady-state mass balance methods, and 3) Level 2 approaches based on dynamic and process-based models. Level 1 approaches includes Canadian models such as SIGMA/SLAM (Dupont and Grimard, 1989) and the Integrated Assessment (IAM) Model (Jeffries *et al.* 1999). These models only consider sulfate as an acidifying pollutant. Both models generate pH-based critical loads, where a pH of 6 is considered as the threshold. Scandinavian and US models rather consider alkalinity-based thresholds. The Steady-State Water Chemistry (SSWC) model, also know as the F-factor model develop by Henriksen, is probably the more widely used and the simplest to adapt (Henriksen, 1979). The Modification of the Steady-State (MOSS) Method (Shaffer *et al.* 1991) is another derived form of the Henriksen's model. Both models use a very limited number of geochemistry and lakewater data to compute CLs. Other very popular level 1 methods include the Steady-State Mass Balance (SMB) model (deVries, 1991; Posch *et al.* 1993, 1997), the PROFILE method (Warfinge and Sverdrup, 1992), and the FAB or First-order Acidity Balance (FAB) model (Hindar and Henriksen, 1998). Finally, level 2 models such as the MAGIC (Cosby *et al.* 1985; Foster *et al.* 2001), MAGIC-WAND model (Ferrier *et al.* 1995), SAFE, SMART and MERLIN models (UN ECE, 1996) consider more complex geochemistry processes and data, and can be use to dynamically simulate the fate of water quality from lake watersheds.

The critical load concept is widely accepted in the UN ECE country members, with the exception of the United States where the US EPA indicated it was not comfortable with the use of an acid deposition standard such as a critical or target load (US EPA, 1995), due to the large amount of scientific uncertainty regarding the role of nitrogen in acidification and watershed nitrogen saturation (Alberta Environment, 1999). US EPA may have a point about the level of precision these models have. It can be seen debatable by some to set stringent standards while the values generated by these tools can be highly variable. On the other hand, critical loads with their unprecision can still be useful environmental indicators to assess the ecosystem vulnerability and pinpoint those areas more exposed to acidification. It can also give an idea of the amount of acidic deposition to be reduced in order to favor acidification recovery. It is in this state on mind that the critical load concept was considered for the NEG/ECP area. The main goals are:

- Identify sensitive lake regions inside the NEG/ECP territory;
- Identify lake regions that could benefit from additionnal reduction in deposition;
- Identify those lake regions that will probably not recover following emission controls.

The Water monitoring work group chose to generate a map of the critical load of acid deposition of sulphur + nitrogen (S + N) based on the Steady State Water Chemistry (SSWC) model (Henriksen and Posch., 2001). This map was supposed to assess the maximum level of acidic deposition that would not induce biological damages in the long term to sensitive lake ecosystems. At the same time, this critical load map would provide an index of the surface water sensitivity to acidification. This approach has been used in other countries such as Great Britain (Ullyett *et al.* 2001). The work group members agreed to work together by pooling lake data from provinces and states into an integrated database.

Material and methods

Model selection

One of the major constraints underlying the application of the SSWC model to the NEG/ECP area was the data heterogeneity amongst jurisdictions, and also the lack of geochemistry data to support the more robust critical load models available. The FAB model was initially considered, but was set aside due the almost total lack of geochemistry data in our lake database. The SIGMA/SLAM was also set aside because it is now considered outdated. The IAM model was also considered, but was rejected because of the lack of a nitrogen component in the actual version of the model. The choice of SSWC over FAB was conformed by the paper by Hindar and Henriksen (1998) who applied both models of a set of Ontario lakes and the one by Henriksen *al.* (2002) who used it successfully on 1495 Ontario lakes. Both models generated output that were mostly in agreement even if the SSWC model is cruder in nature. The effectiveness of the SSWC model was also confirmed at a recent workshop on critical loads in Copenhagen in 1999 (Lokke *et al.* 2000). The Holdren *et al.* (1993) paper also showed the SSWC model ranked amongst the best in its category. Attendies at the reported workshop were more confident in CL estimates from surface water models than their soils and forests counterparts.

The SSWC model is built around critical loads derived from alkalinity or ANC (acid neutralizing capacity) values and estimates of pre-industrial concentrations of sulphates and basic cations (see Hindar *et al.* (1998) and Henriksen *et al.* (2002) for a description of the SSWC equations and processes). Because the model is ANC-based, critical load thresholds are not directly expressing pH levels. It is possible, however, to approximate the desired pH by defining a value of ANC-limit. Sverdrup *et al.* (1990) were amongst the first to suggest a pH of 6 as the chemical criterion best suited to protect most aquatic organisms. The latest federal-provincial 5-years assessment report (Jeffries, 1997) made a round-up of biological effects on aquatic organisms and plants and conclude that significant biological damages start to occur around a pH of 6 and increase with decreasing pH. The use of this pH 6 threshold in the IAM model was again described in Jeffries *et al.* (1999). Holt *et al.* (2002) recently confirmed that a pH of 6 is the threshold to use for zooplankton species when modelling community changes in Central-Ontario. This criterion of pH 6 was also discussed by Henriksen *et al.* (2002) for Ontario lakes

Data integration

A worksheet version of the SSWC model was sent to all jurisdictions to compile data from each province and state. This project involved using "available data" on "clean lakes" (lakes where no significant agricultural, industrial or urban activities are present). For this reason, there are some heterogeneity among lake sets. There were guiding principles involved. It was agreed to share the most recent lake data available for these clean lakes. For most states and provinces, samples were collected between 1996 and 2002 (New Hampshire, Vermont, Maine, Connecticut, Nova Scotia,

New Brunswick, New-Ffoundland. There were two exceptions : Massachussetts, wich lakes were sampled between 1988 and 1992, and Quebec, where lakes were visited between 1986 and 1990. Quebec lakes came mostly form the Quebec Lake Survey (Dupont, 1992a), where lake selection was statistically-derived to enable quantitative extrapolation to the entire lake population. Most samples were collected during fall, but summer samples were considered when no other data were available. Late winter samples were used for Quebec lakes. Winter samples collected under ice-cover were found to be the most representative of yearly conditions for Quebec lakes (Dupont, 1986). Fall samples were found to overestimate the organic acidity component. Correction for sea-salts was performed for jurisdictions near the sea coast. All samples are integrated and most were taken between 0 and 5 meters from the lake surface or in the photic zone. All lakes used in this study were georeferenced and had pH, SO₄, ANC, NO₃ or total alkalinity, calcium, magnesium, and other minor anions and cations data. Lake data from Massachussetts, Connecticut, Vermont, Maine, New Hampshire, Nova Scotia, Newfoundland, New Brunswick and Quebec were gathered by the representative of each jurisdiction and sent in Spring 2002 to the University of Maine (New England data) and Environment Quebec (eastern Canadian province data). A total of 1703 lakes were considered in this study. Annual runoff was obtained from hydrological atlases (Atlantic provinces) and 30-year annual average runoff derived from climatology data (Quebec). Annual runoff ranged mostly from 400 to 900 mm/year, but some extreme runoff was encountered for some high-altitude or sea-coast lakes (up to 1300 mm/year). All other coefficients were borrowed from the Henriksen *et al.* (2002) paper (e.g. S=400 meq/m²/year, etc.)

Analytical methodology

Laboratory methodologies were compared to check if there were any major compatibility issues. All methodology used among the jurisdictions are derived from *Standard Methods* (Clesceri *et al.* 1998), with minor differences in detection limits or laboratory protocols. A small committee of scientists examined the protocols and methodologies used by each state or province and concluded they were compatible. No quality assurance/quality control was performed however. pH was measured everywhere with electrometry. ANC was measured with GRAN titration. Total alkalinity was measured with conductivimetric titration. Nitrate was measured by automated colorimetry. Sulfate was measured by plasma emission ion chromatography, with the exception of Quebec who relied on a modified colorimetric methodology generating results in concordance with the IC (the method makes correction for high DOC levels that can induce a bias when using standard colorimetry for SO₄). All basic cations were analysed with ion chromatography. All parameters were expressed as ueq/L, but CL results are expressed as fluxes (meq/m²/year).

Model assumptions

Models represent a simplification of a complex reality. For this reason, we have to assume that weathering rates don't change over the years, which is not the case according to recent studies showing a decreasing trend in base cation concentrations in soils and water (Jeffries *et al.* 1999). For this reason, critical load values computed from older lake data sets by the SSWC model will probably be overestimated as noted by Henriksen *et al.* (2002). Another factor to take into account is the fact that not all lakes had a pH of 6 in the beginning. Some lakes may have already been acidic or may have shown a pH < 6 in the past. For this reason, many authors use a statistical limit to exclude these cases. In Canada, a 5 % damage level (or the minimum-affected-area criterion in Holdren *et al.* 1993 paper) was set for the pH 6 threshold criterion. This damage-level excludes highly sensitive lakes that may be acidic even without an anthropogenic contribution to acidic deposition. In this paper, we don't rely on such a damage level because the main outputs are maps and not quantitative estimates of lake proportion below or above pH 6.

From the 1703 lakes studied, 22 (1.2 %) showed a negative critical load value. These negative values were set to zero. However, such lakes may never have been able to withstand a pH of 6 in the past.

Another factor to consider is the model error and uncertainty. Henriksen *et al.* (1992) listed 3 major uncertainty factors for the SSWC model : 1) the model structure and parameters, 2) the input data due to measurement errors and calculation methods, and 3) the relationships between lake chemistry and aquatic biota. Several papers do acknowledge that CL models involve uncertainties, but few try to assess the model error. Jeffries *et al.* (1999) mentioned a 25-30 % error for the CL estimates. The amount of error may again be higher or lower for the SSWC and other steady-state models depending of the input data quality. It is often suggested to avoid calculating a CL for an individual lake and favor probability distributions of CL for given groups of lakes (Posch *et al.* 1993, 1997). Because this paper does not intend to specifically assess specific lake CL, we relied on CL classes for our maps. We also made the assumption that CL estimates were of the correct order of magnitude. Comparison of SIGMA/SLAM, IAM and SSWC results obtained for the Quebec lake data set showed similar results and spatial variations, thus comforting us in the representativity of critical load estimates for NEG/ECP lakes. Holdren *et al.* (1993) did similar comparisons and came to the same conclusions that different models generated similar CL. Hokke *et al.* (2000) reported from a UN ECE workshop that critical loads should be considered as separation between deposition levels with different risk/probability of damage, instead of being regarded as thresholds. The maps in this paper are in agreement with this viewpoint.

Another basic assumption underlying the use of the SSWC model is the steady-state component. All CL calculated should be seen as long-term *sustainable* deposition (Skeffington, 1999). We thus have to assume that steady-state will occur eventually. This explain why there is often a lack of correlation between actual water quality data and prediction by the SSWC model (Curtis *et al.*, In press). Only dynamic modelling could ascertain the time needed to reach this steady state. The amount of damages may vary from lake to lake depending on the biota community. Some species may be tolerant to low pH, while others are not. In this study, we assume that damages start with a pH of 6 for all lakes. We also have to assume that all lake data are compatible and uniform, which is probably not the case because of differences in sampling seasons, sampling methodologies, analytical methodology, etc. The fact that the SSWC model does not consider organic anions also demands that we assume that humic acid concentrations in all NEG/ECP lakes are similar, which is surely untrue.

The authors in the scientific literature often consider a ANC-limit value (tolerance criterium for aquatic organisms) of 20 ueq/L as a threshold to consider for critical load computations (Henriksen *et al.*, 1995 ; Posch *et al.* 1997; Hindar *et al.*, 1998). Lien *et al.* (1992) found that brown trout populations in Norway were generally intact above 20 ueq/L. This roughly translates to a pH of 5.5. This observation stands true in Canada where similar fish populations seem intact above this threshold. However, as mentioned earlier, several sport and non sport fish species (e.g. minnows) are affected below a pH of 6. Sverdrup and DeVries (1994) did adopt this line of reasoning and adopted a 50 ueq/L ANC criterion. The ANC-limit should in fact take into account the inherent sensitivity of the local area. Where 20 ueq/L. is fine for Norway, it may prove to be too low or too high for other countries (Henriksen *et al.* 1992). For this reason, Henriksen *et al.* (2002) opted for a 40 ueq/L ANC-limit to be on the safe side. Protection of most aquatic organisms being considered important, the workgroup chose a similar protection threshold Use of the pH-alkalinity relationship developed by Small and Sutton (1986) confirmed that a 40 ueq/L

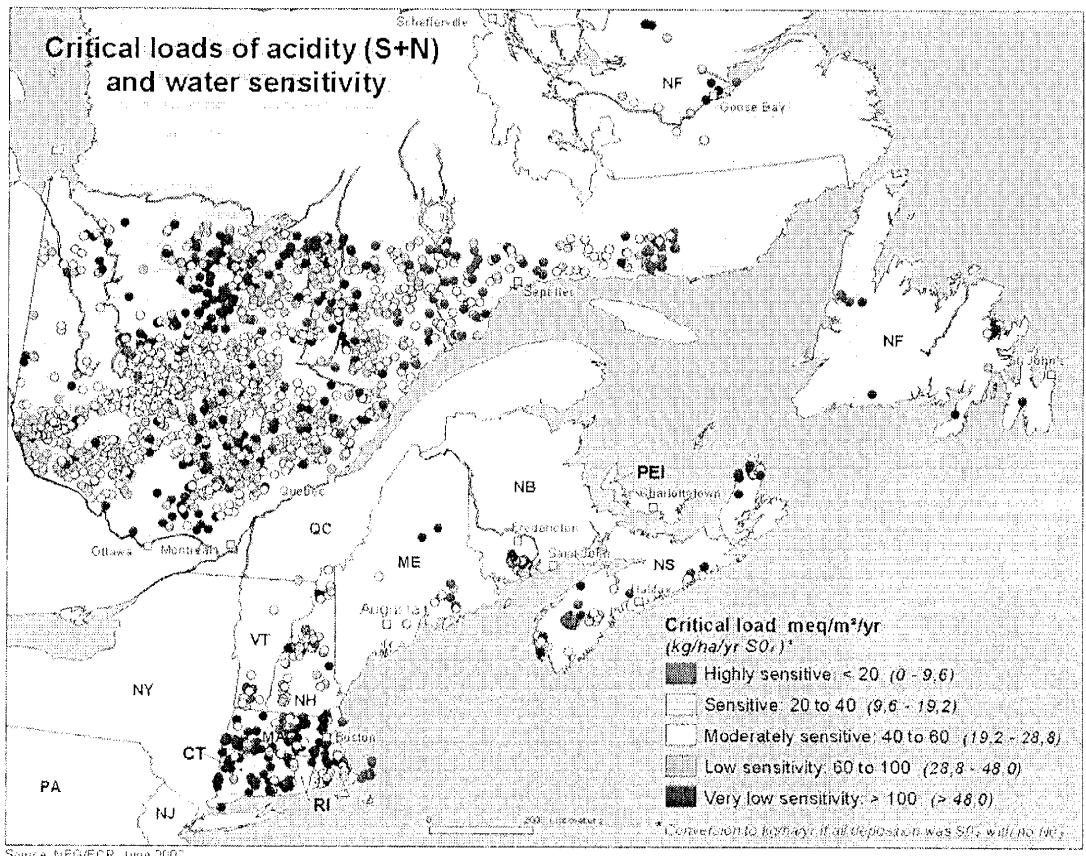
ANC value corresponds to a pH 6 for Quebec lake waters. This value is the range of ANC values (0-50 $\mu\text{eq/L}$) discussed by Raddum and Skjelkdale (2001).

Maps

Henriksen's model enables us to generate two series of maps. The first series shows the critical load map with values expressed in $\text{meq/m}^2/\text{year}$ (with consideration of a ANC-limit of 40 $\mu\text{eq/L}$). The map can be expressed as point sites (Figure 1) or extrapolated spatially using a density function approach in the GIS Arcview Spatial analyst to highlight hot spots and protected areas (Figure 2). A 30-km radius area of influence was used for the density function. Interpolation is generated when sites are less than 60 km apart. Use of GIS geostatistical krigging methods or square-grid pattern (used in UN ECE reports) to generate a grid-based map of critical loads were considered but were set aside because of large gaps on the NEG/ECP territory. The gaps would have induced "spikes" or "singularities" in areas devoid of lakes. Many cells would also have been empty with a square-grid map. These gaps are generally related to data unavailability. Most of the Canadian Shield region and New England states are adequately covered but gaps are observed for the southern portion of Quebec (south of the St. Lawrence River), large parts of New Brunswick, Newfoundland and Maine, and all of Prince Edward Island. Many lake areas were not considered because of local sources of pollution or because lakes are not sensitive. Other areas such as the St. Lawrence lowlands do not support lakes.

Results and discussion

Five classes of critical loads were defined (< 20, 20 to 40, 40 to 60, 60 to 100 and \geq 100 $\text{meq/m}^2/\text{year}$ of S + N in deposition). These values can be translated roughly to < 9.6, 9.6 to 19.2, 19.2 to 28.8, 28.8 to 48, and > 48 kg/ha/year of total sulphate-only deposition. The first three classes indicate environments that are extremely sensitive to acidification, and therefore lakes that are potentially vulnerable to acidic deposition. According to these maps, highly sensitive waters are found in northwestern Quebec (near Rouyn-Noranda), Quebec's north coast, southern Vermont, eastern Maine, southwestern Nova Scotia and New Brunswick, Cape Cod area, northern New Hampshire and western Newfoundland. Low sensitivity areas are located near Ottawa in Canada, in Connecticut, in Massachusetts, northern Maine, and southern Newfoundland. Most locations in Quebec, northern New England (Vermont, New Hampshire, and Maine), Nova Scotia, and Labrador exhibit moderately elevated critical load values.



Source: NEECEP, June 2002

Figure 1. Critical loads of acidity (S + N) and the relative sensitivity of surface waters to acidification

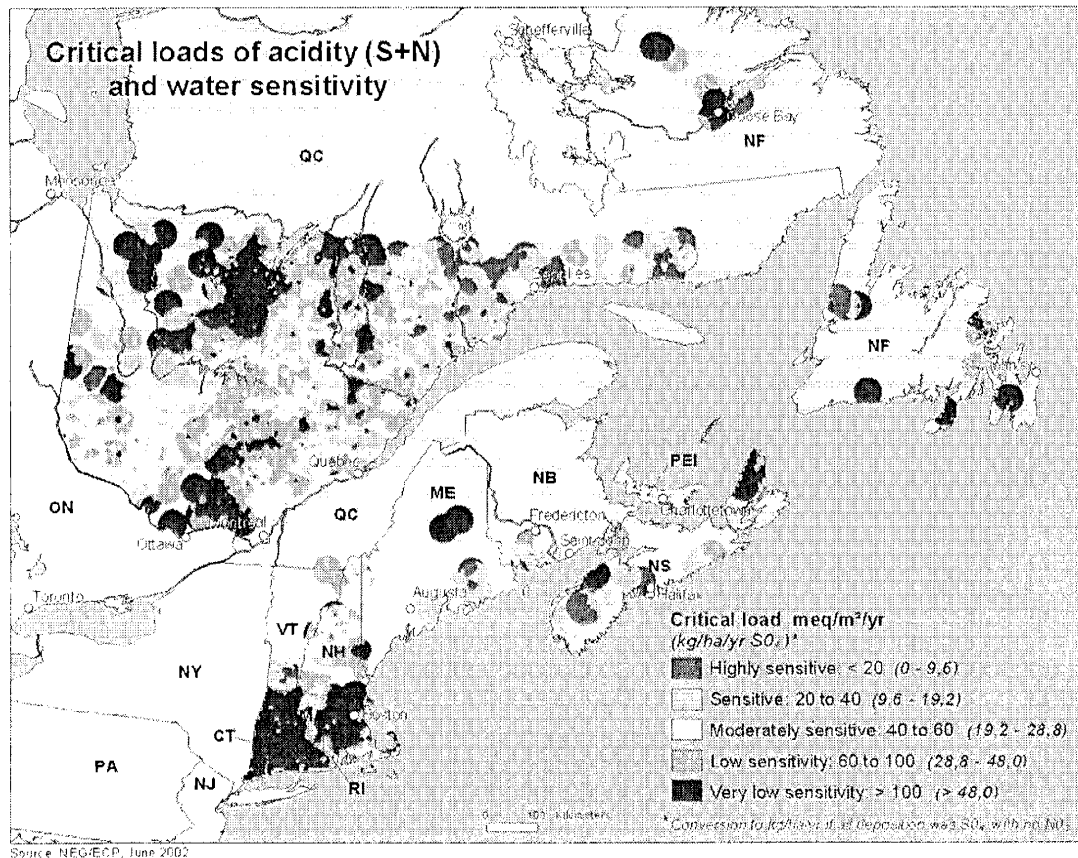


Figure 2. Critical loads of acidity (S + N) and the relative sensitivity of surface waters to acidification (spatial variability interpolated with a density function in Arcview Spatial Analyst)

Table 1 presents the proportions of lakes according to 5 critical load categories for individual jurisdictions, New England and Eastern Canada and the entire NEG/ECP area. As for the maps (Figure 1 and 2), the lakes associated with the ≤ 20 and 20-40 meq/m²/year categories are the most likely to be affected by present day acidic deposition. These 2 classes represent 28 % of all NEG/ECP lakes (varying from 7.2 % in Connecticut to 56.7 % in Nova Scotia). The mean critical load for NEG/ECP lakes is 56 meq/m²/year (54.5 meq/m²/year for Eastern Canadian lakes and 77.5 meq/m²/year for New England lakes). This value is similar to one found by Henriksen *et al.*(2002) for Ontario lakes, and those values reported by the authors for Finland (63 meq/m²/year), Norway (56 meq/m²/year), and Sweden (61 meq/m²/year). Nova Scotia, New Brunswick, Maine and Vermont host very diluted lakes. These jurisdictions have respectively 56.7%, 56.4%, 56.3% and 50% of their lake sets showing an anual N+S critical load below 40 meq/m²/year. On the other hand, some states such as Connecticut, have a fewer percentage of lakes with low critical loads. Finally, Massachussets has both lakes with very low or very high critical loads. The geology, terrain morphology and altitude may explain those differences. These physiographic factors can vary much on the local scale where geology is very complex (mix of sedimentary rocks and local ignous intrusive bedrock), and where altitude is highly variable.

Table 1. Proportion of lakes in 5 critical load classes for individual jurisdiction, region or NEG/ECP area.

Jurisdiction	n	Critical loads (meq/m ² /year)				
		≤ 20	20 - 40	40 – 60	60 – 100	> 100
Connecticut	56	1.8	5.4	7.1	14.3	71.4
Maine	16	25.0	31.3	25.0	6.3	12.4
Massachussets	125	20.0	9.6	10.4	9.6	50.4
New Hampshire	47	6.4	21.3	25.5	38.3	8.5
Vermont	12	25.0	25.0	33.3	16.7	0.0
New England (total)	256	19.9	6.3	9.0	5.5	58.6
New Brunswick	39	25.6	30.8	25.6	10.3	7.7
Newfoundland – Labrador	29	13.8	0.0	20.7	20.7	44.8
Nova Scotia	97	37.1	19.6	13.4	13.4	16.5
Quebec	1282	6.4	19.1	30.2	27.5	16.8
Eastern Canada (total)	1447	9.1	18.3	28.7	25.9	18.0
NEG/ECP area (total)	1703	9.9	18.1	26.5	24.4	21.1

Figures 3 and 4 show critical exceedances expressed as 5 value classes. The orange (exceedances ranging between 0 and 10 meq/m²/yr) and red classes (over 10 meq/m²/yr) show areas where critical loads are actually exceeded or near-exceeded by current levels of acidic deposition. These areas are mostly located on Quebec's north coast, western Quebec, southwestern Nova Scotia and New Brunswick, western Newfoundland, central and coastal Massachusetts, southern Vermont, several areas in New Hampshire and southern Maine. A large portion of the lake water areas presents high critical load values protecting them from acidic deposition. It is the case in Connecticut, Rhode Island, the greater part of Massachusetts, some sectors of the Atlantic Provinces and central Quebec. High critical loads would also be observed on the south shore of the St. Lawrence in Quebec if lake data had been included.

Figure 3 and 4 show that acidic deposition does not exceed critical loads for a large portion of the NEG/ECP region. This means that, in general, the lake waters in these areas should be able to maintain a pH of 6 or an ANC value of 40 ueq/L (green and blue areas). Acidic lakes may be present in these parts but their number or proportion should be very low compared with non acidic ones.

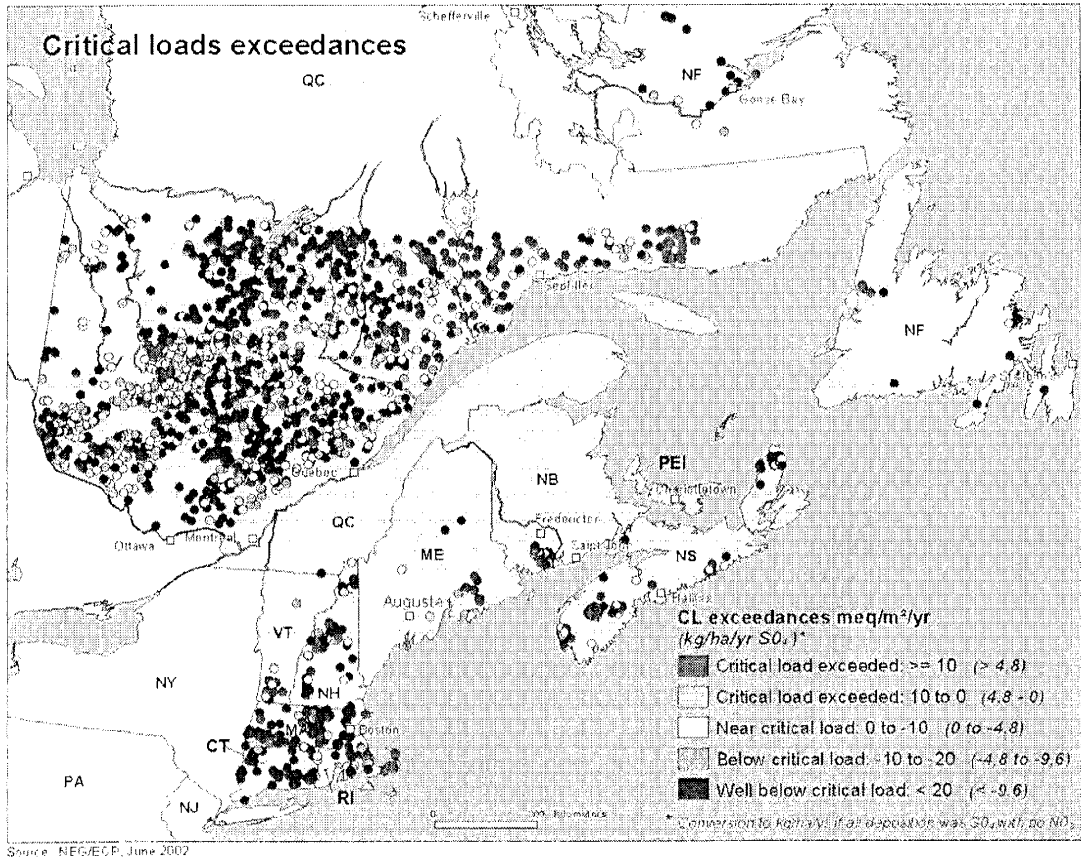


Figure 3. Critical load exceedances.

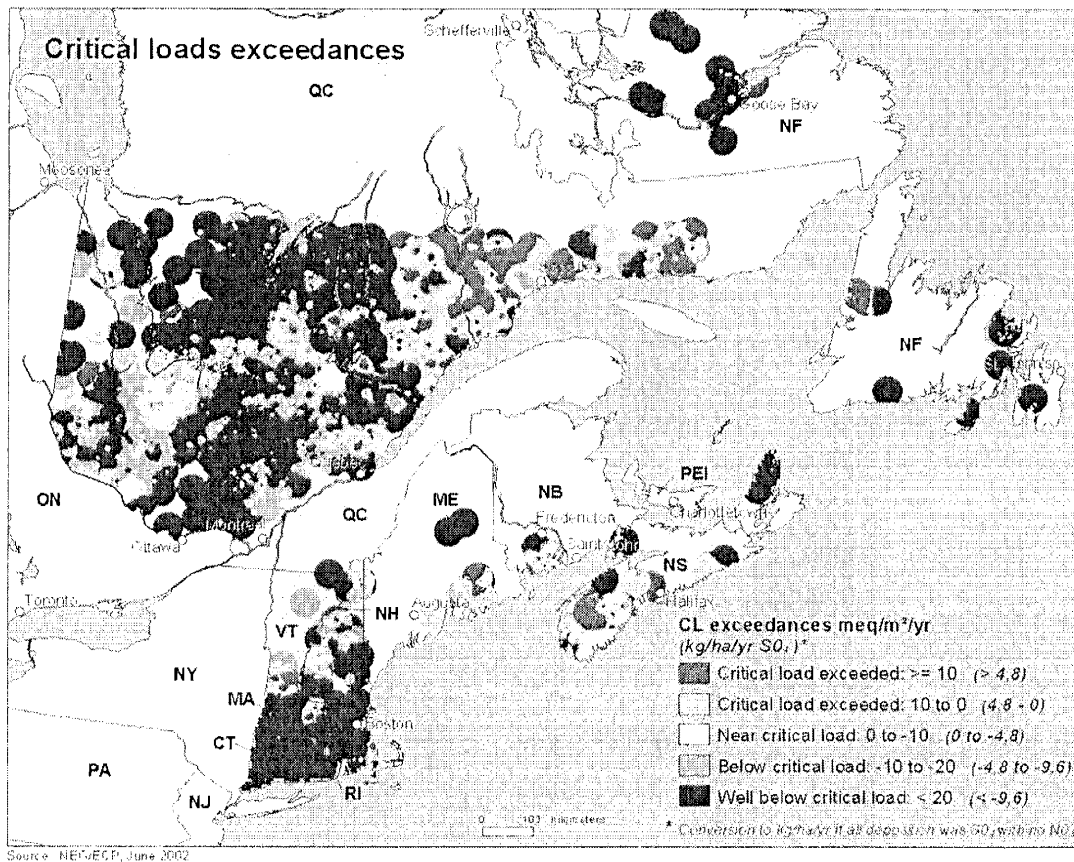


Figure 4. Critical load exceedances interpolated spatially with a density function in Arcview Spatial Analyst.

There are many hot spots showing exceedances above $10 \text{ meq/m}^2/\text{year}$. The largest by far is located in northeastern Quebec where critical loads are often above $10 \text{ meq/m}^2/\text{year}$. Other major hot spots are located in Nova Scotia, north of Quebec City, near Rouyn-Noranda in northwestern Quebec, Maine, southern Vermont and a few isolated spots in other jurisdictions. For these lakes, the potential for recovery seems very farfetched because the anthropogenic contribution to deposition would need to be completely removed, which is very unlikely. In many cases, those lake hot spots host very diluted lakes or brownwater lakes that never supported a pH of 6. This is the case for North Coast lakes in Québec, southwestern lakes in Nova Scotia and some westernmost lakes in Newfoundland. The North Coast lakes represent a very special case. 33 % of the lakes in this area show a pH lower than 5.5 and a large number of these never supported a sustainable fish population (Dupont, 1992b). These lakes may have been acidic for millenia. Acidic deposition in these remote area is very near the natural background levels. In some lakes, even removing all deposition would not suffice to achieve critical load.

Table 2 represents the critical load exceedances for individual jurisdictions, New England, Eastern Canada and NEG/ECP area. Proportions are very similar to the table 1 statistics. Overall, critical loads are exceeded for 29.6 % of the lakes with another 14.4 % near the exceedance threshold. The proportion of lakes showing exceedances varies from 7.2% in Connecticut to 59% in New Brunswick, with most jurisdictions in the 25-30%. New England and Eastern Canada exhibit almost identical percentages (26.9% for Canada and 26.2% for New England). Again,

Nova Scotia (56.7%), New Brunswick (59%), Maine (56.3%) and Vermont (50%) show the highest proportions of exceedance. These values are greater than those reported in Henriksen *et al.* (2002) for south-central Ontario (19%), Finland (9%), Sweden (9%), Denmark (9%), but are similar to Norway (26%). The greater proportions of lakes exhibiting exceedances can be explained by the fact that very sensitive lake areas have been included in the study, while less sensitive lakes have been left out (New Brunswick, Nova Scotia, etc.). This is not the case for Quebec lakes where a statistical survey was conducted over the entire Canadian Shield area, and in Massachusetts, where all parts of the state have been covered.

Table 2. Proportion of lakes in 5 critical load exceedance classes for individual jurisdiction, region or NEG/ECP area.

Jurisdiction	n	Exceedances (meq/m ² /year)				
		≥ 10	10 to 0	0 to -10	-10 to -20	< 20
Connecticut	56	1.8	5.4	7.1	14.3	71.4
Maine	16	43.8	12.5	18.7	6.3	18.7
Massachusetts	125	24.0	5.6	6.4	4.0	60.0
New Hampshire	47	14.9	14.9	12.8	10.6	46.8
Vermont	12	50.0	0.0	25.0	8.3	16.7
New England (total)	256	18.1	11.5	14.4	11.7	44.3
New Brunswick	39	46.2	12.8	12.8	10.3	17.9
Newfoundland – Labrador	29	13.8	0.0	10.3	10.3	65.6
Nova Scotia	97	45.4	11.3	11.3	2.1	29.9
Quebec	1282	14.6	12.9	15.8	13.9	42.8
Eastern Canada (total)	1447	14.0	12.9	14.5	16.0	42.6
NEG/ECP area (total)	1703	18.1	11.5	14.4	11.7	44.3

It is important to note that these critical loads combine both S and N contributions. Reduction of emissions should thus be achieved by reducing either sulphate or nitrate or a given amount of both. In the early 80's, a target load of 20 kg/ha/year only addressed wet sulphate. The actual critical load computed with the SSWC model, however, takes into account bulk deposition and both S and N. Considering that nitrate is almost equivalent to sulphate in deposition and even greater in some areas, this means that a large number of lakes would need a reduction in deposition of sulfate to under 10 kg/ha/year and a similar reduction of nitrate to insure that acidification will not occur.

The National Acid Rain Strategy conclusions (AETG, 1997) mention that a 75% reduction (with reference to 1995 levels) in sulfate deposition would be required from Canada and the US to achieve critical loads. With such a reduction in emissions, the report notes that all recently acidified lakes should be restored. But this prediction does not take into account nitrogen deposition nor basic cation depletion in soils and lakes. This means that some very sensitive acidified lakes may not recover from acidification even with such a reduction. The Eulerian *Acid Deposition and Oxidant Model* (ADOM) was applied to generate deposition fields under various emissions-reduction scenarios. These simulated loadings were feeded into the IAM model to predict sulfate critical loads and exceedances for southeastern Canadian lakes (Kaminski, 2002). One of the more efficient reduction scenario would require a 50% additional reduction from both Canada and US SO₂ emitters based on the AQA 2010 initial emission targets.

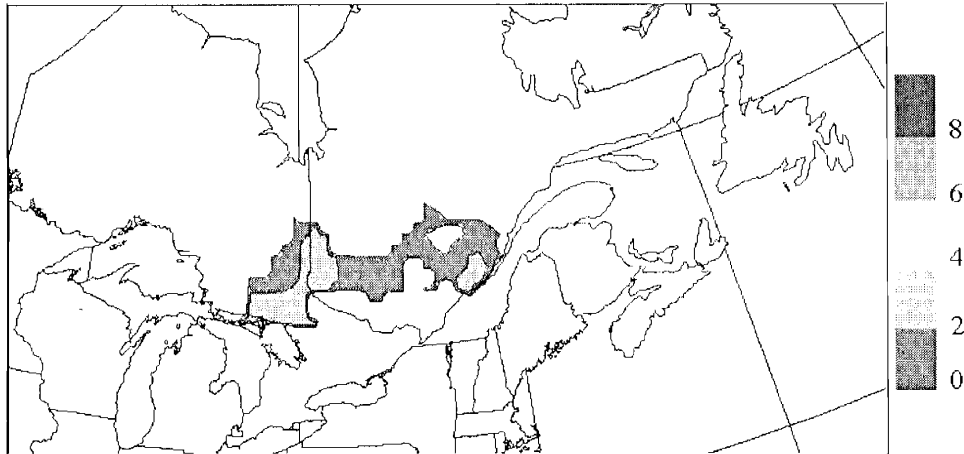


Figure 5. ADOM/IAM model prediction for a 50 % additional decrease in emission from USA and Canada with reference to emission targets set in 1995 for 2010 in the Air Quality Accord (Source : Kaminski, 2002).

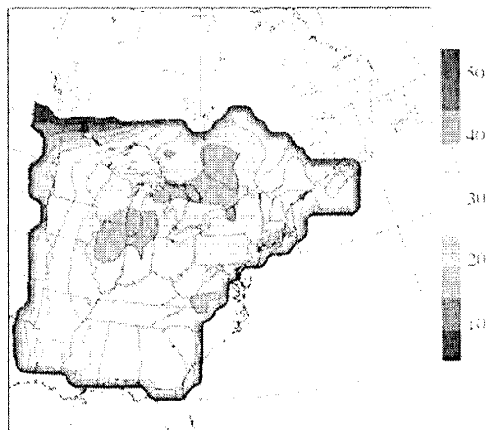


Figure 6. Percent difference field between the 1995 target emission scenario and a 50 % additional reduction in emission from US and Canada for annual ambient SO_4 (source : Kaminski, 2002)

The additional 50 % emission reduction scenario described in Kaminski (2002), show that exceedances from critical loads in deposition (accounting for a 5% damage level) would range from 0 to 4 kg/ha/year of sulfates. The potential percentage SO_4 deposition reduction would range from above 40% in western Quebec, around 30% range for New England and southwestern Nova Scotia and New Brunswick, and in the 20% range and lower elsewhere to the northeast. This means that lakes present in the orange shaded areas on Figure 4 (0-10 meq/m²/year), which may roughly translate as a 0-4.8 kg/ha/year of sulfate deposition, would probably recover from acidification following an additional 50 % reduction in emission.

Because of the model uncertainties and the fact that lakes near survey sites may be more or less acidic, the critical loads calculated by the SSWC model may be over or underestimated, and thus appear in the wrong class on Figure 6. For this reason, it is plausible to assume that both orange (critical loads exceeded) and yellow lake areas (near critical load level) may benefit from additional reductions and be recovered.

Apart from this recovery suggested with the emission reduction scenarios, which can be regarded as arguable depending if you are or are not a modeller or a believer, the critical load and exceedances maps still represent useful tools to assess lake sensitivity in a jurisdiction. Critical load maps are better suited to assess the vulnerability of surface waters exposed to acidic deposition. ANC and alkalinity maps are also useful to show which lake areas are most or least sensitive, but they do not convey the exposure factor to acidic deposition (e.g., is the lake affected by acidic deposition or what amount of acidic deposition would be needed to insure that the biota will not be affected in the long term?). With such a tool, decision-makers can pinpoint what lake areas are most susceptible to acidification or to recovery, and make plans accordingly to monitor these lake area more intensively over time to assess the impacts of future reductions.

Conclusion

The Water Quality Working Group, under the auspices of the New England Governors and Eastern Canadian Premiers Conference, has been issued to produce a sensitivity map for the NEG/ECP territory that would replace the one produced in 1988. The Group opted instead for the production of a critical load map (and exceedance map) to show where vulnerable areas were located and to assess the maximum level of acidic deposition required to sustain adequate water quality conditions over the long term. The Henriksen's Steady State Water Chemistry model (SSWC) was selected for that task and lake data from all NEG/ECP jurisdictions were pooled together and used to calculate combined nitrogen and sulfur critical loads and deposition exceedances. Model results show that a large portion of the NEG/ECP is well protected from acidification, but a significant part of it receive deposition in exceedance of lake critical loads. Maps were used to assess the extent of these lake areas affected by deposition. In turn, these same maps should help decision-makers highlight hot spot locations and maximise water quality monitoring in these areas in order to assess the recovery of recently acidified lakes.

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References :

- AETG, 1997. *Towards a National Acid Rain Strategy*. National Air Issues Coordinating Committee, The Acidifying Emission Task Group, 98 p.
- Aherne, J., D. Ryan, Y. de Kluzenaar, R. van den Beuken and E.P. Farrell. 2000. *Critical loads and levels – Literature review of the current state of knowledge regarding the calculation and mapping of critical loads : Determination and mapping of critical loads for sulphur and nitrogen and critical loads for ozone in Ireland*. Forest Ecosystem Research Group Report No. 45, 40 pages.
- Alberta Environment 1999. *Application of Critical, Target, and Monitoring Loads for the*

- Evaluation and Management of Acid Deposition*. Clean Air, Strategic Alliance, Alberta Environment, Publication no T/472, 67 p.
- Cosby, B.J., R.F. Wright, G.M. Hornberger and J.N. Galloway. 1985. Modelling the effects of acid deposition: assessment of a lumped parameter model for soil water and stream chemistry. *Water Research* **21**:51-63.
- Clesceri, L.S., A.D. Eaton and A.E. Greenberg (Eds.). 1998. *Standards Methods for Examination of Water and Wastewater, 20th Edition*. APHA, AWWA, WEF.
- Curtis, C.J., B. Reynolds, T.E.H. Allott and R. Harriman. 2003. The link between the exceedance of acidity critical loads for freshwaters, current chemical status and biological damage : A re-interpretation. *Water, Air and Soil Pollution* **In press**:399-413.
- DeVries, W. and J. Kros. 1991. *Assessment of critical loads and the impact of deposition scenarios by steady state and dynamic soil acidification models*. Report no. 36, DLO The Winand Staring Centre for Integrated Land, Soil and Water Research, Wageningen, The Netherlands.
- Dupont, J. 1986. *Evolution spatiale et temporelle de l'acidité de 14 lacs de la région de Portneuf pour la période 1982-1983*. Quebec Ministry of the Environment, Report No. PA-21, 97 p.
- Dupont, J. and Y. Grimard. 1989. A simple dose-effect model of lake acidity in Quebec (Canada). *Water, Air and Soil Pollution* **44**:259-272.
- Dupont, J. 1992a. Quebec Lake Survey : I. Statistical assessment of surface water quality. *Water Air and Soil Pollution* **61**:107-124.
- Dupont, J. 1992b. Quebec Lake Survey : II. Origin and extent of acidification. *Water Air and Soil Pollution* **61**:125-137.
- Ferrier, R.C., W. De Vries and P. Warfvinge. 1995. *The use of dynamic models for the determination of critical loads for nitrogen – Developments since Løkeberg*. Proceedings of the UN ECE Workshop on nitrogen deposition and its effects : Critical loads mapping and modelling, Grange-Over-Sands, 24-26 October 1994, Department of the Environment, London.
- Foster, H.J., M.J. Lees, H.S. Wheeler, C. Neal and B. Reynolds. 2001. Dynamic modelling of spatially variable catchment hydrochemistry for critical loads assessment. *Water, Air and Soil Pollution* **130**:1283-1288.
- Henriksen, A. 1979. A simple approach for identifying and measuring acidification of freshwater. *Nature* **278**:542-545.
- Henriksen, A., J. Kamari, M. Posch and A. Wilander. 1992. Critical loads of acidity : Nordic Surface Waters. *Ambio* **21**:356-363.
- Henriksen, A. et M. Posch. 2001. Steady-State Models for Calculating Critical Loads of Acidity for Surface Waters. *Water, Air and Soil Pollution Focus* **1**, 375-398.
- Henriksen, A., P.J. Dillon and J. Aherne. 2002. Critical loads of acidity for surface waters in south-central Ontario, Canada : regional application of the Steady-State Water Chemistry (SSWC) model. *Canadian J. Fisheries and Aquatic Sciences* **59**:1287-1295.
- Hindar, A. and A. Henriksen. 1998. *Mapping of Critical Loads and Critical Load Exceedances in the Killarney Provincial Park, Ontario, Canada*. Norwegian Institute for Water Research, report no. O-97156, 36 p.
- Hindar, A., A. Henriksen, S. Sandoy and A.J. Romundstad. 1998. Critical load concept to set restoration goals for liming acidified norwegian waters. *Restoration Ecology* **6**:353-363.
- Holdren jr, G.R., T.C. Strickland, P.W. Shaffer, P.F. Ryan, P.L. Ringold and R.S. Turner. 1993. Sensitivity of critical load estimates for surface waters to model selection and regionalization schemes. *J. of Environmental Quality* **22**:279-289.
- Holdren jr, G.R., T. C. Strickland, B.J. Cosby, D. Marmorek, D. Bernard, R. Santore, C.T. Driscoll, L. Pardo, C. Hunsaker and R.S. Turner. 1993. *Environmental management* **17**:355-363.

- Holt, C.A., N.D. Yan and K.M. Somers. 2002. pH 6 as the threshold to use in the critical load modeling for zooplankton community change with acidification in lakes of south-central Ontario : accounting for morphometry and geography. *Canadian J. Fisheries Aquat. Sci.* **60**:151-158.
- Jeffries, D.S. (Ed.). 1997. *1997 Canadian Assessment Acid Rain Assessment -- Vol III : The Effects on Canada's Lakes, Rivers and Wetlands*. Environment Canada.
- Jeffries, D.S., D.C.L. Lam, M.D. Moran and I. Wong. 1999. The effects of SO₂ emission controls on critical load exceedances for lakes in southeastern Canada. *Water Science & Technology* **39**:165-171.
- Lien, L., G.G. Raddum and A. Fjellheim. 1992. *Critical Loads for Surface Waters : Invertebrates and Fish*. Norwegian Institute for Water Research, Oslo, Norway, Acid Rain Research Report no.21.
- Lokke, H., J. Bak, R. Bobbink, K. Bull, C. Curtis, U. Falkengren-Grerup, M. Forsius, P. Gundersen, M. Hornung, B.L. Skjelkvale, M. Starr and K. Tybirk. 2000. *Critical Loads Copenhagen 1999*. Danmarks Miljøundersøgelser, Arbejdsrapport fra DMU nr. 121, 48 p.
- Kaminski, J.W. 2002. *Emissions-Scenario Simulations of New Provincial SO₂ Reduction Targets Using the Acid Deposition and Oxidant Model*. ARM Consultants, prepared for the anadian Council of Ministers of the Environment, inc., 23 p.
- Nilsson, J. and P. Grennfelt (Eds.). 1988. Critical loads for sulphur and nitrogen. Nordic Council of Ministers and the United Nations Economic Commission for Europe, Miljørapport 1988:16.
- Posch, M., M. Forsius and J. Kamari. 1993. Critical loads of sulfur and nitrogen for lakes I : model description and estimation of uncertainty. *Water, Air and Soil Pollution* **66**:173-192.
- Posch, M., J. Kamari, M. Forsius, A. Henriksen and A. Wilander. 1997. Exceedance of critical loads for lakes in Finland, Norway and Sweden : Reduction requirements for acidifying nitrogen and sulfur deposition. *Environmental management* **21**:291-304.
- Raddum, G.G. and B.L. Skjelkdale. 2001. Critical limit of acidifying compounds to invertebrates in different regions of Europe. *Water, Air and Soil Pollution* **130**:825-830.
- Shaffer, P.W., B. Rosenbaum, G.R. Holdren jr, T.C. Strickland, M.K. McDowell, P.L. Ringold, R.S. Turner, P.F. Ryan and D. Bernard. 1991. *Estimating critical loads of sulfate to surface waters in the northeastern United States : A comparative assessment of three procedures for estimating critical loads of sulfate for lakes*. United States Environmental Protection Agency, Corvallis, Oregon, US EPA Report 600-3-91-062.
- Skeffington, R.A. 1999. The use of critical loads in environmental policy making : A critical appraisal. *Environmental Science & Technology* **June 1999**: 245A-252A.
- Small, M.J. and M.C. Sutton. 1986. A regional pH-alkalinity relationship. *Water Research* **20**:335-343.
- Sverdrup, H., W. De Vries and A. Henriksen. 1990. *Mapping critical loads*. Nordic Council of Ministers, Copenhagen, Denmark, Nord 1990:98.
- Sverdrup, H. and W. De Vries. 1994. Calculating critical loads for acidity with the simple mass balance method. *Water, Air and Soil Pollution* **72**:143-160.
- Ulllyett, J.M., J.R. Hall, M. Hornung and M. Kernan. 2001. Mapping the potential sensitivity of surface waters to acidification using measured freshwater critical loads as an indicator of acid sensitive areas. *Water, Air and Soil Pollution* **130**: 1241-1246.
- UN ECE. 1996. *Manual on Methodologies and Criteria for Mapping Critical Levels/Loads and Geographical Areas Where They Are Exceeded*. UN ECE Commission on Long-Range Transboundary Air Pollution, prepared Task Force on Mapping with assistance of the Coordination Center for Effects (CCE), 204 p.
- US EPA. 1995. *Acid Deposition Standard Feasibility Study Report to Congress*. Report no. EPA 430-R095-001a, Office of Air and Radiation, Acid Rain Division.

Warfvinge, P. and H. Sverdrup. 1992. Calculating critical loads of acid deposition with PROFILE: A steady state soil chemistry model. *Water, Air and Soil Pollution* **63**:119-143.