

Canadian Experiences in Development of Critical Loads for Sulphur and Nitrogen

Shaun Watmough, biogeochemist, Department of Environmental and Resource Studies,
Trent University, Peterborough, Ontario K9J 7B8, Canada

Julian Aherne, biogeochemist, Department of Environmental and Resource Studies,
Trent University, Peterborough, Ontario, Canada

Paul Arp, Professor, Faculty of Forestry and Environment Management, University of
New Brunswick Fredericton, NB, Canada

Ian DeMerchant, GIS specialist, Canadian Forest Service, Atlantic Forestry Centre,
Fredericton, NB, Canada

Rock Ouimet, forest soil science scientist, Direction de la recherche forestière,
Forêt Québec, Ministère des Ressources Naturelles du Québec, Sainte-Foy, QC, Canada

Abstract—Critical loads are a broad-scale modelling approach designed to assess the potential risk of pollutants to ecosystems. A description of the methodology for estimating critical loads (sulphur and nitrogen) for acid deposition (CL(A)) for upland forests in eastern Canada is presented, using a case study in central Ontario. In eastern Canada, CL(A) have been calculated for upland forests, with the objective of maintaining the molar ratio of base cations to aluminium in soil solution above 10. In the current approach, nitrogen (N) dynamics including N fixation, N immobilisation and denitrification have been set to zero. Further, critical load estimates presented in this study do not include nutrient removals through harvesting, and dry deposition input is estimated to be 20 percent of wet (1994 to 1998) deposition. Critical loads were calculated separately for Ontario, Québec and the Maritime Provinces (New Brunswick, Nova Scotia and Newfoundland) using the same methods, but using different soil and forest databases. Mean area-weighted critical loads among provinces are similar, ranging between 273 eq ha⁻¹ yr⁻¹ (Newfoundland) and 512 eq ha⁻¹ yr⁻¹ (Ontario). Preliminary estimates indicate that more than 50 percent of the upland forest area in Ontario and Québec and between 10 (Newfoundland) and 33 percent (Nova Scotia) of upland forest in the Maritimes receive acid deposition in excess of the critical load. Current efforts are being directed toward improving the accuracy of critical load estimates and current exceedances using better estimates of dry deposition and harvesting removals, and investigating the linkage between exceedance of the critical load and adverse biological effects.

Introduction

In Europe and increasingly in other parts of the world, the concept of critical loads has been applied to estimate acceptable levels of acid deposition (Posch and others 2001). A critical load is 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). Critical loads have been most commonly applied to sulphur and nitrogen deposition, although critical loads for trace metals, ozone and organic chemicals are also being developed (Posch and others 2001). Critical load models have been an important and successful tool for

the development of control strategies for transboundary air pollution in Europe (Gregor and others 2001), and critical loads for acid deposition (sulphur (S) and nitrogen (N)) have recently been calculated for regions in eastern North America (Arp and others 1996, Ouimet and others 2001, Watmough and Dillon 2003a). The most common method for calculating steady state critical loads of acidity for forest soils is the Simple Mass Balance (SMB) model (Posch and others 2001).

Upland forests in the Northeastern United States and Eastern Canada are particularly sensitive to acidic deposition because of high local S and N deposition rates, and because shallow and poorly-buffered upland soils are common throughout the region. Critical loads for upland forests in eastern Canada were estimated under the

NEG/ECP (New England Governors/Eastern Canadian Premiers) working group on critical loads (Québec and Maritime Provinces) or through a complimentary project funded by Environment Canada. In this paper we present a description of the methodology adopted for critical load calculations using a case study in Ontario. Preliminary critical load estimates for Eastern Canada are summarised also.

Methods

The Simple Mass Balance Model

The simple mass balance model assumes a simplified, steady state input-output description of the most important biogeochemical processes that affect soil acidification. Potential ecosystem inputs include: 1) atmospheric deposition of S, N, calcium (Ca), magnesium (Mg), and potassium (K); 2) soil base cation weathering rate; and 3) N fixation. Ecosystem outputs and consumption include: 1) net nutrient accumulation in the soil; 2) net nutrient storage in above-ground biomass by uptake; 3) net removal of nutrients by forest harvesting or other disturbance; 4) nutrient loss through soil leaching; and 5) denitrification or N immobilisation.

$$CL(S+N)=BC_{dep}-Cl_{dep}+BC_w+N_i+N_u+N_{de}-ALK_{le(crit)} \quad (1)$$

Equation (1) describes the critical load calculation (units are in eq ha⁻¹ yr⁻¹), where BC_{dep} = base cation (Ca + Mg + K + sodium (Na)) deposition, Cl_{dep} = chloride deposition BC_w = base cation weathering, Bc_u = net base cation (Ca + Mg + K) uptake by trees, N_i = net nitrogen immobilisation rate in soil, N_u = net nitrogen uptake by trees, N_{de} = net denitrification rate, and ALK_{le(crit)} = critical alkalinity leaching rate. In the current assessment, harvesting removals were excluded, in other words, Bc_u and N_u were set to zero. In addition, N_i and N_{de} were assumed to be negligible. Further details on the SMB model are given by (UBA 1996) and the NEG/ECP Forest Mapping Group (NEG/ECP Environment Task Group 2001).

A key component of the critical load calculation is the critical alkalinity leaching rate. This critical chemical criterion ultimately determines the acceptable level of base saturation in forest soils. Among the many chemical criteria proposed in the European modelling exercise, the UN-ECE developed a model to calculate the critical ALK_{le} for forest soils based on two criteria: the molar Bc:aluminium (Al) ratio in soil leachate; and the gibbsite dissolution constant which controls Al solubility (UBA 1996). The following equation summarises the calculation:

$$ALK_{le(crit)} = 1.5 \times \left(\frac{Bc_{dep} + Bc_w - Bc_u}{(Bc / Al)_{crit}} \right) + Q^{2/3} \times \left(1.5 \times \frac{Bc_{dep} + Bc_w - Bc_u}{(Bc / Al)_{crit} \times K_{gibb}} \right)^{1/3} \quad (2)$$

where Bc = K + Ca + Mg (*dep*, *w*, and *u* represent deposition, weathering, and net uptake, respectively; eq ha⁻¹ yr⁻¹), Na is excluded since it provides no protection against Al toxicity for plants, (Bc/Al)_{crit} = molar soil solution threshold criterion, Q = water flux from the bottom of the rooting zone (m³ ha⁻¹ yr⁻¹), and K_{gibb} = gibbsite dissolution constant criterion (m⁶ eq⁻²). A range of threshold values have been proposed for Bc/Al and K_{gibb}; the current assessment is based on Bc/Al = 10 and log₁₀(K_{gibb} in (L mol⁻¹)²) = 9.0. These values were chosen with the overall objective of maintaining soil base saturation at an acceptable level for forest health and productivity.

Data Requirements

Critical loads for upland forests and related exceedances were compiled and mapped separately for Newfoundland, Nova Scotia, New Brunswick (Ballard and others 2004), Québec (Ouimet 2004), and Ontario (Watmough and others 2004) following the same methodology, but using different data sources. Input data for critical load calculations (table 1) were collected from multiple sources although the majority of the data were obtained from the Canadian Soil Information System (CANSIS) and Environment Canada. More detailed land polygon (CANSIS) data were available for the Maritimes compared with Québec and Ontario. All atmospheric wet deposition data were provided by Environment Canada (Ro and Vet 2002), and represent the average wet deposition (1994 to 1998), multiplied by a constant weighting factor of 20 percent to account for dry deposition. Updated estimates of dry deposition are currently being included into revised critical load maps.

Table 1. Input parameters for calculation of critical loads.

Theme	Map attribute (units)
Climate	Runoff or soil percolation (m)
	Mean annual temperature (°C)
	Atmospheric deposition Ca, Mg, K, Na, Cl, SO ₄ , NO ₃ , NH ₄ (eq ha ⁻¹)
Land polygons	Ecological region, CANSIS soil landscapes, etc.
Soil data	Substrate acidity type: acid, intermediate or basic
	Average soil depth (m)
	Average clay content (percent)

Base Cation Weathering Rate

Base cation weathering rates for each soil polygon were estimated based on the weighted clay content of soil, parent substrate, soil depth and temperature (Sverdrup and others 1990):

$$\text{Acid substrate: } W = (56.7 \times \% \text{clay}) - (0.32 \times \% \text{clay}^2) \quad (3)$$

$$\text{Intermediate substrate: } W = 500 + (53.6 \times \% \text{clay}) - (0.18 \times \% \text{clay}^2) \quad (4)$$

$$\text{Basic substrate: } W = 500 + (59.2 \times \% \text{clay}) \quad (5)$$

where W = base cation weathering rate ($\text{eq ha}^{-1} \text{yr}^{-1}$) for 1 m soil depth. Weathering estimates were corrected for differences in mean annual temperature:

$$W_c = W \times e^{((A/(2.6+273)) - (A/(273+T)))} \quad (6)$$

where W_c is the temperature-corrected weathering rate ($\text{eq ha}^{-1} \text{yr}^{-1}$) for 1 m soil depth; A is the Arrhenius constant (3600°K) and T is the mean annual temperature ($^\circ \text{C}$). The maximum soil depth used in the calculations was 0.75 m, based on the assumption that this represented the maximum rooting depth for upland forests.

The application of this steady state model within the geographic mapping context has been described in detail by the (NEG/ECP Environment Task Group 2001) and (Arp and others 2001). The case study presented here, describes the calculation of critical load and current exceedance for 10 soil mapping units situated in the Manitoulin Island region, central Ontario (fig. 1). Preliminary critical load estimates for eastern Canada, that are included in the 2004 Canadian Acid Rain Assessment report to be released in early 2005 (Environment Canada in prep), are summarised also.

Results

Case Study: Manitoulin Island

The weighted percent clay content of the 10 land polygons centered around Manitoulin Island was between 2.5 and 33.4 percent; soil depth for all polygons was set to the maximum depth of 0.75 m (table 2). By incorporating substrate type and average annual temperature for each land polygon, mean annual base cation weathering rates were estimated to be between 113.9 and 2303.7 $\text{eq ha}^{-1} \text{yr}^{-1}$ (table 2). Higher weathering rates are associated with higher clay content (table 2) and substrate acidity. Annual base cation deposition was generally higher in the southern part of the region, reflecting the closer

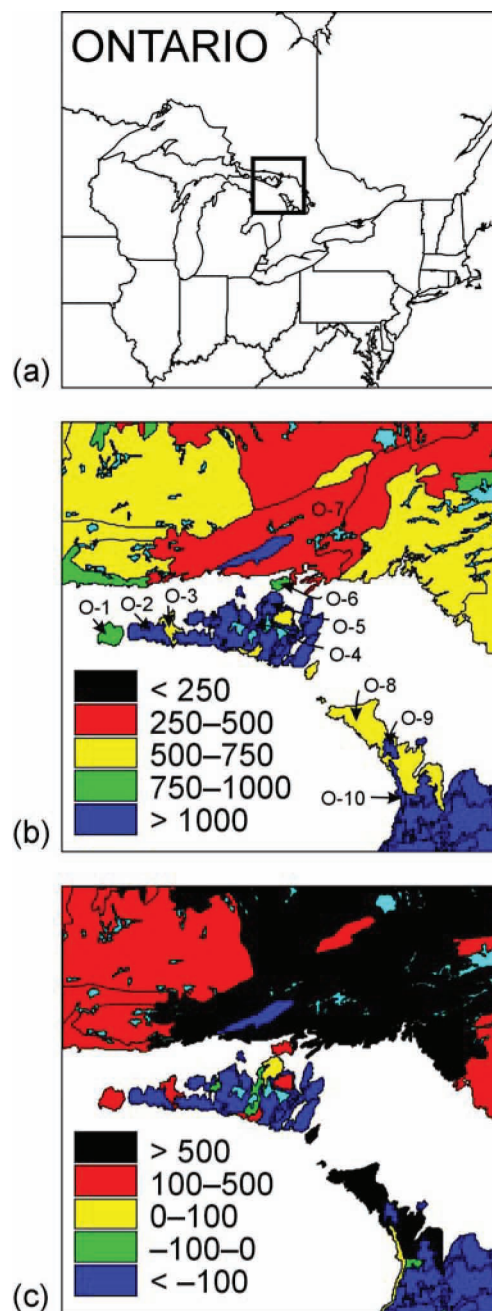


Figure 1. (a) Case study location, centred around Manitoulin Island and the Bruce peninsula (b) critical loads for acid deposition ($\text{eq ha}^{-1} \text{yr}^{-1}$) and (c) current exceedance ($\text{eq ha}^{-1} \text{yr}^{-1}$) based on average 1994 to 1998 deposition.

proximity to industrial activities in southern Ontario and the greater dominance of calcareous soils. Base cation deposition ranged from a low of 145.9 $\text{eq ha}^{-1} \text{yr}^{-1}$ at the north-west tip of Manitoulin Island, to a high of 214.5 $\text{eq ha}^{-1} \text{yr}^{-1}$ at the most southerly land polygon on the Bruce Peninsula (table 2; fig. 1). Base cation inputs from deposition represented between 8 (O-9) and 60 percent (O-7) of the total annual base cation weathering inputs for each land polygon.

Table 2. Selected land polygon characteristics (percent clay, weathering rate, base cation deposition, critical alkalinity leaching, and sulphur and nitrogen deposition) for critical load calculations and estimates of current exceedance (negative values indicate no exceedance).

ID	Clay (percent)	W _c ¹	BC _{dep}	ALK _{le(crit)} (eq ha ⁻¹ yr ⁻¹)	S _{dep}	N _{dep}	CL(A)	Exceedance
O-1	15.6	528.8	145.9	-165.9	334.2	495.9	869.8	126.3
O-2	19.4	1045.2	163.2	-287.1	364.6	543.9	1528.2	-438.0
O-3	11.1	290.4	163.2	-113.0	364.6	543.9	599.3	490.9
O-4	10.8	783.6	178.1	-230.7	389.8	576.0	1228.0	-69.1
O-5	14.8	704.3	187.6	-207.5	416.8	613.0	1137.0	98.7
O-6	12.1	552.2	187.7	-172.7	417.1	613.3	950.1	286.4
O-7	2.5	113.9	176.2	-73.6	393.2	577.7	398.8	766.2
O-8	13.1	278.6	207.4	-118.6	443.9	656.8	646.1	674.7
O-9	33.4	2303.7	205.3	-574.7	439.5	648.7	3124.7	-1818.8
O-10	7.8	774.8	214.5	-231.8	454.5	671.8	1264.0	87.5

¹ W_c: temperate corrected weathering rates were normalized to a maximum depth of 75 cm.

Land polygons with higher base cation inputs (and no harvesting) will have a greater ALK_{le(crit)} (equation 2). In this example, annual ALK_{le(crit)} was between -73.6 (O-7) and -574.7 eq ha⁻¹ yr⁻¹ (O-9). All N dynamic parameters (fixation, immobilisation and denitrification) were set to 0 and we assumed that there was no forest harvesting. Critical loads for acid deposition for each land polygon are simply a function of the base cation weathering rate, base cation deposition, and the ALK_{le(crit)} (equation 1). Critical loads for acid deposition are between 398.8 (O-7) and 3124.7 eq ha⁻¹ yr⁻¹ (O-9) (table 2; fig. 1).

Sulphate deposition is between 334.2 and 454.5 eq ha⁻¹ yr⁻¹ (table 2) and follows a similar pattern to base cation deposition with higher values occurring in the southern part (O-10) of the study region. Similarly, N (nitrate + ammonium) deposition follows the same spatial pattern as both sulphate and base cations and ranges from 495.9 to 671.8 eq ha⁻¹ yr⁻¹ (typically ~60 percent of total acid deposition), with the highest values occurring in the southern region of the island. Acid deposition exceeds the critical load at 7 of the 10 land polygons, with exceedance values ranging between 87.5 (O-10) and 766.2 eq ha⁻¹ yr⁻¹ (O-7), indicating that reductions in acid (S + N) deposition of up to 78 percent (O-7) are required to

prevent the Bc:Al molar ratio in soil solution from falling below 10 (table 2).

Eastern Canada

Approximately half of the mapped area in Ontario and Québec currently receives acid deposition in excess of the critical load (table 3). Newfoundland has the lowest areal exceedance, with 10 percent of the mapped area exceeding the critical load. The percentage area of exceedance in New Brunswick and Nova Scotia is intermediate between Newfoundland and Québec/Ontario. Comparison between the provinces, however, should be made with caution, since the mapping units (polygon area) in Ontario and Québec are much larger (≥ 300 km² on average) than in the Maritime provinces (1.5 km² on average). On average, almost 48 percent of the mapped region of eastern Canada exceeds the critical load of acidity. The greatest exceedance occurs where atmospheric acid deposition rates are high and where the critical soil acidification loads are calculated to be low, and occur in central and eastern Ontario, southern Québec, especially in the Lower-Laurentide region north of the St. Lawrence River, and southeastern Nova Scotia.

Table 3. Summary statistics for critical load estimates and current exceedances in eastern Canada (2004 Canadian Acid Rain Assessment [Environment Canada, in press]).

Province	Mean critical load ¹ (eq ha ⁻¹ yr ⁻¹)	Mean exceedance ¹	95 th percentile exceedance ¹	Terrestrial area in exceedance (percent)
Newfoundland	273	153	292	9.9
Nova Scotia	429	192	296	33.1
New Brunswick	459	148	426	10.6
Ontario	512	202	602	50.6
Québec	416	353	736	51.5
Total	439	265	427	48.3

¹ Area-weighted.

Discussion

Steady state critical loads for acid deposition calculated using the SMB model provide a method of estimating the potential risk from acid deposition over a large geographical area. They highlight areas that are potentially most at risk from acid deposition, but do not provide an indication of when adverse effects will occur. The current approach also assumes that N immobilisation is zero, which does not generally reflect the present situation (Watmough and Dillon 2003b), but assumes that “at steady state” there will be no net N immobilisation (or denitrification and fixation). Critical load estimates are also based upon the assumption that molar Bc:Al ratios in soil solution below 10 will result in adverse effects on forest health. In this study we have adopted a precautionary approach with respect to both the critical chemical limit and N dynamics. However, the critical chemical limit in soil, soil weathering rates, and particularly N dynamics in forest soils are areas of active research, so it is possible that critical load maps will be updated as new information becomes available.

In general, the greatest exceedance of the critical load occurs in regions that are estimated to have the lowest weathering rates and are therefore likely to have the lowest exchangeable base cation pools in soil. Recent studies in eastern Canada have provided evidence for a link between exceedance of the critical load and adverse biological effects (Ouimet and others 2001; Moayeri 2001; Watmough and others 2004), although a causal relationship between poor forest health and exceedance of the critical load has yet to be established. Current research efforts are being directed toward improving the accuracy of critical load estimates and current exceedance calculations by using more accurate estimates of dry deposition and soil weathering rates, and by including land-use changes (for example, harvesting and fire).

Acknowledgments

The authors gratefully acknowledge the Canadian National Atmospheric Chemistry (NAtChem) Precipitation Database and its data contributing agencies/organisations for the provision of the wet deposition data for 1994 to 1998, used in this publication. The agencies/organisations responsible for data contributions in 1994 to 1998 to the NAtChem/Precipitation Database include Environment Canada, and the provinces of Ontario, Québec, New Brunswick, Nova Scotia and Newfoundland, the United States Environmental Protection Agency, and the United States National Atmospheric Deposition Program/National Trends Network.

References

- Arp, P.A.; Leger, W.; Moayeri, M.H.; Hurley, J.E.; 2001. Methods for mapping Forest Sensitivity to Acid Deposition for northeastern North America. *Ecosystem Health*. 7: 35–47.
- Arp, P.A.; Oja, T.; Marsh, M. 1996. Calculating critical S and N loads and current exceedances for upland forests in southern Ontario, Canada. *Canadian Journal of Forest Research*. 26: 696–709.
- Balland, V.; Arp, P.A.; Hurley, J.E.; DeMerchant, I.; Bourassa, Y. 2004. Estimating potential acid-rain induced base-cation depletion economics for Nova Scotia and New Brunswick. in *Pollutants Across Boundaries, Third Canadian Workshop on Air Quality*, Québec, QC, Canada.
- Environment Canada. 2004 Canadian acid rain assessment. Environment Canada, Ottawa, ON, Canada. (in prep).
- Gregor, H.-D.; Nagel, H.-D.; Posch, M. 2001. The UN ECE international programme on mapping critical loads and levels. *Water, Air and Soil Pollution: Focus*. 1: 5–19.
- Moayeri, M.H. 2001. Mass balance related to sustainability of forest biomass production: concepts and applications. Ph.D. Thesis, University of New Brunswick, Fredericton, N.B., Canada.
- NEG/ECP Environment Task Group. 2001. Critical load of sulphur and nitrogen assessment and mapping protocol for upland forests. NewEngland Governors and eastern canadian Premiers, Acid Rain Action Plan, Halifax, Canada. 21 p.
- Nilsson, J.; Grennfelt, P. 1988. Critical loads for sulphur and nitrogen. *NORD 1988:97*, Nordic Council of Ministers, Copenhagen, Denmark. 418 p.
- Ouimet, R. 2004. Cartographie des secteurs forestiers du Québec vulnérables aux précipitations acides : première approximation. Rapport interne No. 482, Gouvernement du Québec, Ministère des Ressources naturelles, de la Faune et des Parcs, Québec.
- Ouimet, R.; Duchesne, L.; Houle, D.; Arp, P.A. 2001. Critical loads of atmospheric S and N deposition and current exceedances for Northern temperate and boreal forests in Québec. *Water Air and Soil Pollution: Focus*. 1(1/2): 119–134.
- Posch, M.; de Smet, P.A.M.; Hettelingh, J.-P.; Downing, R.J. 2001. Modelling and mapping of critical thresholds in Europe. Status report 2001, Working Group on Effects of the Convention on Long-range Transboundary Air Pollution, RIVM, Bilthoven, Netherlands. 165 p.
- Ro, C. U.; Vet, R. J. 2002. Analyzed data fields from the National Atmospheric Chemistry Database (NAtChem) and Analysis Facility. Air Quality Research Branch, Meteorological Service of Canada, Environment Canada, 4905 Dufferin St., Toronto, Ontario, Canada.
- Sverdrup, H.; de Vries, W.; Henriksen, A. 1990. Mapping critical loads: a guidance to the criteria, calculations, data collection and mapping of critical loads. *Miljörappport (Environmental Report) 1990:14.*, Copenhagen: Nordic Council of Ministers. *NORD: 1990:98*. 124 p.
- UBA. 1996. Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded. UN ECE Convention on Long-range Transboundary Air Pollution, Berlin, Germany. 144 p.
- Watmough, S. A.; Dillon, P. J. 2003a. Do critical load models adequately protect forests? A case study in south-central Ontario. *Canadian Journal of Forest Research*. 33: 1544–1556.

- Watmough, S. A.; Dillon, P. J. 2003b. Base cation and nitrogen budgets for seven forested catchments in central Ontario, 1983–1999. *Forest Ecology and Management*. 117: 155–177.
- Watmough, S. A.; Aherne, J.; Dillon, P. J. 2004. Critical loads Ontario: relating exceedance of the critical loads with biological effects at Ontario forests. Report 2. Environmental and Resource Studies, Trent University, Ontario, Canada. 22 p.