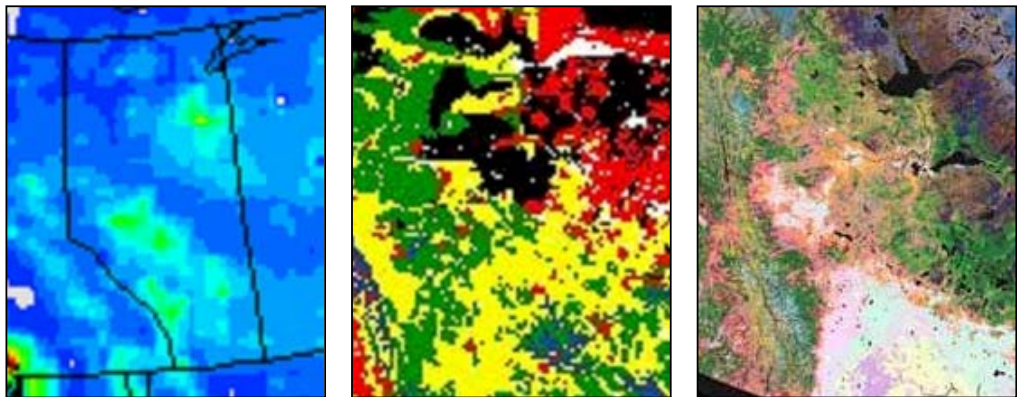


Development of an Approach to Assess Uncertainty in Terrestrial Critical Load and Exceedance



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Table of contents

Summary	ii
Definitions	iii
1. Background	1
2. Objectives	1
3. Methodology	1
3.1. STAGE I Key data-set compilation	2
3.2. STAGE II Monte Carlo sampling	4
3.3. STAGE III Spatial aggregate level	5
3.4. STAGE IV AURAMS level	5
3.5. STAGE V Regional or national level	5
4. Results	5
4.1. Mineral soils and spatially unique units	5
4.2. Critical load uncertainties	6
4.3. Exceedance uncertainties	7
5. Summary and recommendations	8
Acknowledgements	9
References	9
Appendix I. The Steady-State Mass Balance model	11
Appendix II. Data sources	12
Appendix III. Supporting maps and illustrations	13
Appendix IV. Uncertainty distribution types and ranges	14
Appendix V. Application of the uncertainty framework to the Athabasca Oil Sand Region, Alberta	15

Critical load. ‘a quantitative estimate of 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

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Summary

A framework for estimation of uncertainty in critical loads (CL) and exceedances (EX) of acidity (sulphur and nitrogen) was developed for forested mineral soils in Canada. This report describes the framework and its application to a study (test) area (420 km × 420 km) located on the Ontario-Quebec border. The study area was subdivided into 100 squares (42 km × 42 km) based on the spatial structure of modelled sulphur and nitrogen deposition (AURAMS) used for EX calculation. The uncertainty estimates were summarized for each square (referred to as AURAMS GRID).

Monte Carlo (MC) simulations of the Steady-State Mass Balance (SSMB) model were used to assess uncertainty in CL and EX estimates. Under the MC scheme, SSMB model input values were repeatedly randomly sampled from parameter distributions. Depending on model parameter, a normal or rectangular distribution was assigned, with values sampled within the range of up to ±50 % of the parameter mean. The model was run repeatedly (n = 1000), using a new (random) set of parameters for each model run, producing a distribution of output values (i.e., CL and EX) for each spatial aggregate (an area with a unique combination of input parameter values, based on spatial coincidence of key data-sets (maps) used to derive input variables). The median CL and EX values were selected from each resultant frequency distribution. In addition, the coefficient of variation for CL and probability (percent of total simulations) of EX (values > zero) were calculated. All four estimates were summarized for each AURAMS GRID using area weighted cumulative distributions. Each (output) variable was independently sorted in ascending order; then the cumulative area of their corresponding spatial aggregate was calculated and converted into probability to produce a cumulative distribution function (CDF). Fifth percentile CL (5CL) and 95th percentile coefficient of variation of CL (95CL_{CV}) were estimated from the CDF. The 95th percentile EX (95EX) and 95th percentile of exceedance probability (95EX_{PROB}) were also estimated. The resultant values were mapped for each AURAMS grid within the study area. A map representing three categories (high, potential and low risk) of exceedance uncertainty was also presented.

The application of the uncertainty framework to study area revealed varying confidence (increasing or decreasing uncertainty) for 5CL and 95EX estimates. For example, AURAMS GRIDS characterized by low CL_{CV} values (least deviation from the 5CL value) were expected to have low uncertainty. In contrast, higher CL_{CV} lowered the confidence of the mapped 5CL estimate, owing to the larger dispersion of values around the median CL for each AURAMS GRID. Similarly, AURAMS GRIDS with low (95EX_{PROB} < 25 %) or high probability of exceedance (95EX_{PROB} > 75 %) were assumed to have high certainty in their negative or positive 95EX values, respectively. Those characterized by potential exceedance risk (25 % > 95EX_{PROB} < 75 %) were considered less certain, as their 95EX value could shift between exceeded and non-exceeded.

The uncertainty framework is particularly relevant to the assessment of deposition scenarios. A national application of the uncertainty assessment framework is recommended with a few amendments, including revision of the input data uncertainty ranges (based on data sources and expert recommendations) and further differentiation of spatial aggregates (to incorporate forest types).

Definitions

Monte Carlo (MC): repeated (n times) random sampling from predefined frequency distributions. In the current study, a new set of inputs parameters was selected during each run to produce a distribution of output values.

CL: critical load, “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). See Appendix I for CL calculation.

AURAMS GRID: spatial unit (42 km × 42 km in size) used for modelled total sulphur and nitrogen deposition based on North American emission inventories using AURAMS model (A Unified Regional Air quality Modelling System).

Spatial aggregates: a set of unique input values (such as deposition, base cation weathering rate, runoff) based on spatial coincidence defined using an overlay of mapped data layers within each AURAMS GRID.

5CL: area weighted 5th percentile critical load, ensures that 95 % of the area (within an AURAMS GRID) are protected. The 5CL is derived from the area weighted cumulative distribution of CL for all spatial aggregates within each AURAMS GRID. The CL for each spatial aggregate was set at the median from 1000 MC simulations.

95CL_{CV}: area weighted 95th percentile coefficient of variation (CV) of critical load within each AURAMS GRID. The coefficient of variation was derived as the ratio of the standard deviation over the mean (both calculated from 1000 MC simulations for each spatial aggregate). The CV indicates the spread around the median CL for the 1000 simulations.

EX: exceedance was estimated as sulphur and nitrogen deposition minus CL. Negative values indicate non-exceeded CL while positive values indicate that current sulphur and nitrogen deposition exceed the CL

95EX: area weighted 95th percentile of exceedance indicates the S and N deposition exceedance of CL for 95 % of the area within an AURAMS GRID. The 95EX was derived from a cumulative distribution function (CDF). The CDF was created by plotting sorted (in ascending order) median EX values (calculated from 1000 MC simulations) of each spatial aggregate within an AURAMS GRID against cumulative area (represented in %) of each corresponding spatial aggregates.

95EX_{PROB}: area weighted 95th percentile of exceedance probability, indicates the chance that sulphur and nitrogen depositions will exceed the CL (i.e., exceedance values larger than zero) for the majority (95 %) of the area. Probability for each spatial aggregate was estimated as the percent of MC simulations (n = 1000) that were exceeded, sorted in ascending order and plotted against corresponding cumulative area (%). The resultant cumulative distribution function (CDF) was used to estimate the 95th percentile exceedance value.

GEM GRID: Global Environmental Multi-scale (GEM) grid at a resolution of 35 km × 35 km representing the average annual total (wet plus dry) base cation (BC) and chloride (Cl⁻) atmospheric deposition data for Canada during the period 1994–1998.

1. Background

Critical loads (CL) of acidity (sulphur [S] and nitrogen [N]) and exceedance (EX) have been determined and mapped for upland forests across Canada (Carou et al. 2008) following guidelines established by the New England Governors and Eastern Canadian Premiers (NEG-ECP) Environmental Task Group on Forest Mapping (NEG-ECP Environment Task Group, 2001). While there is general consensus that current critical load modelling approaches are adequate, it is also recognised that there is a high level of uncertainty associated with the model (data) inputs (Acid Deposition Critical Load workshop, Ottawa, March 2009).

Uncertainties in CL and EX for forest soils have been investigated for many regions, e.g., Sweden (Barkman 1995, Barkman et al. 1997, Barkman and Alveteg 2001a, b), the United Kingdom (Heywood et al. 2006a, b, c, Skeffington et al. 2006, 2007), Germany and Austria (Suuturi et al. 2001) and the United States of America (Li and McNulty 2007). Uncertainty assessments have been performed at several spatial scales ranging from site specific (e.g., Skeffington et al. 2006, 2007a) to national (e.g., Heywood et al. 2006a, b, c) and larger-scale applications (182 monitoring plots across Europe, Reinds and de Vries 2009).

In general, a single CL or EX value (e.g., 5th percentile CL and 95th percentile EX) is presented for each study site (Barkman and Alveteg 2001b). However, incorporating uncertainty, typically employing a Monte Carlo (MC) approach, results in probability distributions of output variables based on quantified uncertainty in data inputs (Skeffington 2006).

2. Objectives

The primary objective was to develop an approach to assess uncertainty in critical load and exceedance estimates for mineral soils of upland forests in Canada and apply the approach to a study (test) area.

3. Methodology

The effects of input data uncertainty on CL and EX estimates for Canadian forest soils were assessed using MC simulations of the Steady-State Mass Balance (SSMB) model (Sverdrup and De Vries, 1994; see Appendix I). Monte Carlo analysis is a well-established method for assessing the effects of uncertainty in model parameters on model outputs (e.g. Rubinstein, 1981, Barkman and Alveteg 2001a, b, Skeffington et al. 2006). Within a MC scheme, parameter values are randomly sampled from a predefined frequency distribution. The model is run repeatedly, with a new set of parameters for each model run, resulting in a distribution of output values.

To assess uncertainty in CL and EX for forest mineral soils in Canada a framework was developed that incorporated five stages (see Figure 1 and description below). STAGE I involved compilation of key input data-sets, creation of spatial aggregates and the definition of input data ranges and parameter distribution types for those spatial aggregates. STAGE II employed MC sampling to generate distributions of CL and EX for the spatial aggregates. In STAGE III, median CL and EX values were selected from the distributions. Furthermore, for each spatial aggregate the coefficient of variation of CL and the percentage of exceedance values larger than zero were calculated (i.e., EX probability). In STAGE IV area weighted cumulative distribution functions were derived for each output variable (see STAGE III) to estimate the 5th percentile CL (5CL), the 95th percentile coefficient of variation of CL (95CL_{CV}), the 95th percentile EX (95EX) and EX probability (95EX_{PROB}). The 5CL, 95CL_{CV} and 95EX, 95EX_{PROB} were mapped separately in STAGE V.

The uncertainty framework was applied to a study area (420 km × 420 km) located across the Ontario and Quebec border (Figure 2).

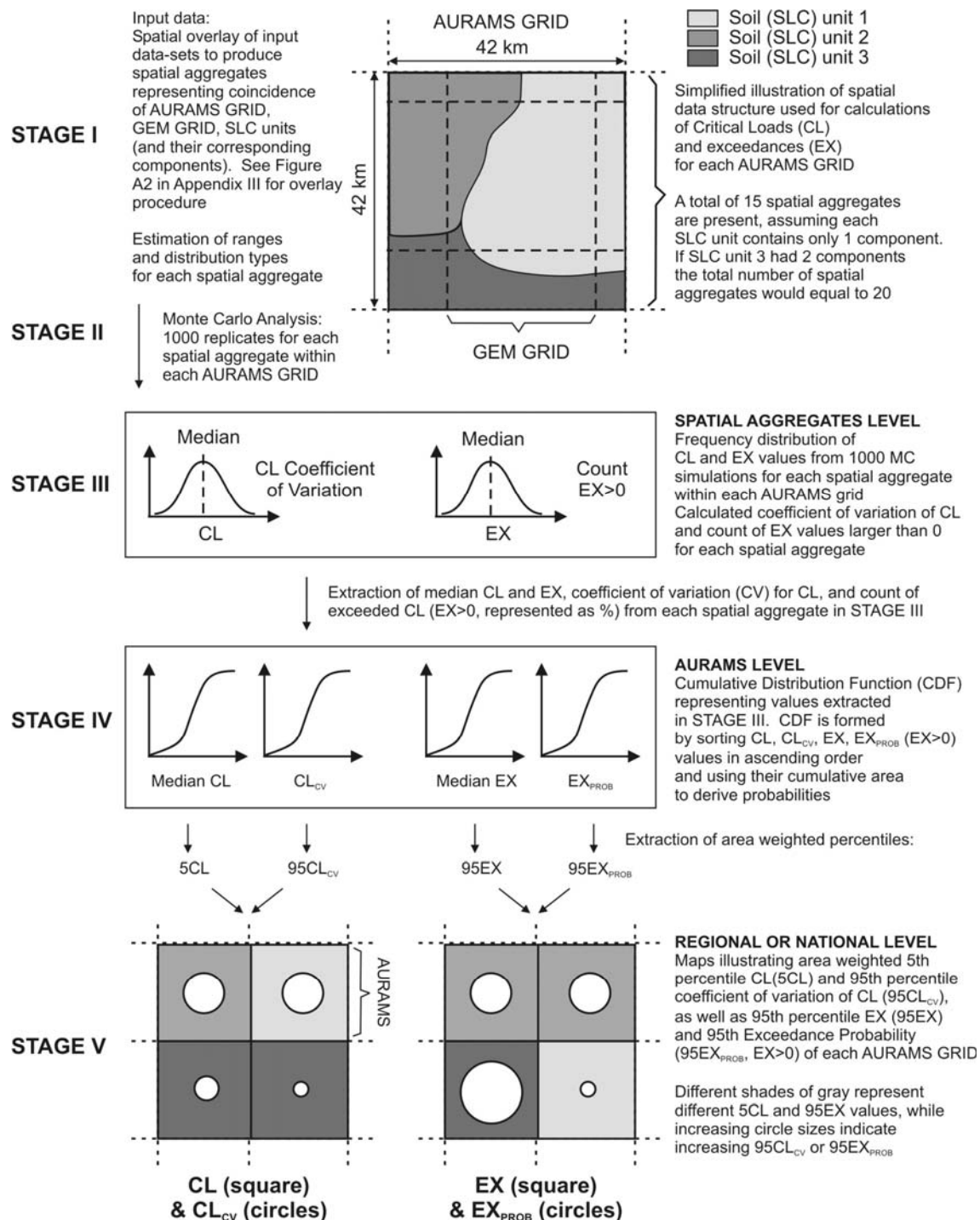


Figure 1. Five stages in the uncertainty assessment framework (see text for description). Note: STAGE V was only applied to the ‘test’ area level in the current study.

3.1. STAGE I Key data-set compilation

Several key data-sets were compiled to derive the necessary (input) variables for CL and EX calculation (Appendix II). The Soil Landscape of Canada (SLC) map was the principal database underlying the estimation of base cation (BC) weathering rate. Gridded (GEM GRID: 35 km × 35 km) total (wet plus dry) atmospheric base cation and chloride (Cl⁻) deposition data were provided by Environment Canada. Modelled total S and N deposition for the entire country were available for 2002 (AURAMS GRID: 42

km × 42 km). Forested areas were delineated using the USGS Global Land Cover Characterization database (GLCC Version 2 Simple Biosphere Model categories).

Base cation weathering rates were estimated using a soil type-texture approximation method (Ouimet 2005). The approach estimated weathering rate from texture (clay content) and parent material class, derived from SLC map (the best resolution database available for estimating national base cation weathering). The most recent version (3.1.1) was used where available¹ and version 2.1 elsewhere (Appendix III, Figure A1). Only soil map components representative of mineral soils (with > 1 % coverage) within each SLC unit were used to produce weathering estimates. To estimate weathering, percent clay was vertically weighted to produce a value for each component (i.e., soil type) within a mapping unit on the SLC map.

Critical alkalinity leaching is typically estimated from a critical molar base cation to aluminium (Bc:Al) ratio in soil solution and the gibbsite dissolution constant (K_{gibb}). The choice of critical chemical limit (receptor and level of protection) has a significant impact on the resultant critical load. However, the selection of the critical limit reflects ethical considerations, a decision made to ensure adequate protection of the selected biological indicator and consequently the chosen receptor ecosystem. To illustrate the influence of the selected chemical criteria, CL and EX estimates for the Ontario-Quebec region were determined using Bc:Al of 1 (the most common protection limit used in Europe [and elsewhere]) and 10 (a more stringent threshold, previously used in Canada) for mineral forest soil (maximum soil depth was set to 50 cm).



Figure 2. Map of Canada with the location of the uncertainty framework ‘study area’. The area covered 420 km × 420 km and was located across the Ontario and Quebec border.

Spatial aggregates

Several steps were undertaken (Figure 1 and Appendix III, Figure A2) to produce unique spatial aggregates of CL and EX input variables for mineral forest soils. Mineral soils were selected from SLC map, while forested areas were identified using USGS Land Cover Characterization database². Forest types representing three categories 1) broadleaf deciduous trees, 2) deciduous and evergreen trees, and 3)

¹ SLC v3.1.1 (August 2007) is the latest revision of the Soil Landscapes of Canada map. However, the map coverage is incomplete: soil information is available for the province of Alberta, Nova Scotia, and Newfoundland and for the major agricultural regions of British Columbia, New Brunswick, Manitoba, Ontario, Quebec and Saskatchewan.

² URL: edc2.usgs.gov/glcc/na_int.php

evergreen needle leaf trees were used to define a single forest cover (however, the three cover types were not differentiated in the study application of uncertainty framework).

Subsequently, an overlay procedure was used to derive a unique combination of parameter values representing spatial coincidence of S and N deposition (uniform per AURAMS GRID), BC and Cl⁻ deposition (GEM GRID) and BC weathering (from SLC units and their corresponding components). The proportion of forest mineral soils as well as count of unique areas per AURAMS GRID were calculated and mapped separately.

Input data distributions

The nature of the distribution for each required input parameter (see Appendix I for SSMB data requirements) reflects knowledge of the particular parameter. Ranges of input parameters and their uncertainties may be based on collected data, literature survey, or expert judgment (e.g., Heywood et al 2006a).

In the current study, data distribution types and ranges were primarily based on literature recommendations (Table 1 and Appendix IV, Table 2A). Depending on the uncertainty associated with each model parameter, a normal (i.e., greater confidence) or rectangular (i.e., less confidence, Skeffington et al. 2007b) distribution was assigned, with values sampled within the range of up to $\pm 50\%$ of the parameter mean. Normal distribution was utilized for BC, N, S, and Cl⁻ deposition, as well as runoff (Q) and temperature (T). Rectangular distribution was used for BC weathering (BC_w), N immobilization (N_i) and denitrification (N_{de}). Wider ranges were used for less certain parameters, while narrower ranges were utilized for parameters characterized by more certainty. For example, in the current study S deposition was assigned $\pm 20\%$ uncertainty while N deposition was given $\pm 40\%$; justification stems from estimated reliability of the two data sources. Sulphur deposition estimates are considered to be more reliable than N, as model processes better capture observed S deposition compared to N deposition.

Table 1. List of Steady-State Mass Balance model parameters randomly varied within the Monte Carlo scheme to estimate critical load and exceedance frequency distribution. Each parameter was assigned a distribution type (D [R is rectangular, N is normal]), based on previous literature recommendations (see Appendix IV). Parameter values were randomly sampled within each distribution of specified ranges (up to $\pm 50\%$ of the mean).

Parameter	Variable description	Unit	D	Range
BC _w	Base cation weathering (depth weighted)	eq ha ⁻¹ yr ⁻¹	R	$\pm 40\%$
S _{dep}	Sulphur deposition	eq ha ⁻¹ yr ⁻¹	N	$\pm 20\%$
N _{dep}	Nitrogen deposition	eq ha ⁻¹ yr ⁻¹	N	$\pm 40\%$
BC _{dep}	Base cation deposition	eq ha ⁻¹ yr ⁻¹	N	$\pm 20\%$
Na _{dep}	Sodium deposition	eq ha ⁻¹ yr ⁻¹	N	$\pm 20\%$
Cl _{dep}	Chloride deposition	eq ha ⁻¹ yr ⁻¹	N	$\pm 20\%$
N _i	Nitrogen immobilization (mean set to 0.5 kg ha ⁻¹)	eq ha ⁻¹ yr ⁻¹	R	$\pm 50\%$
N _{de}	Nitrogen denitrification (mean set to 0.5 kg ha ⁻¹)	eq ha ⁻¹ yr ⁻¹	R	$\pm 50\%$
T	Temperature	°C	N	$\pm 10\%$
Q	Runoff	M	N	$\pm 20\%$
K _{gibb}	Gibbsite dissolution constant	m ⁶ eq ⁻²	R	8.5–9.5
Bc:Al	Base cation to aluminium ratio			1, 10

3.2. STAGE II Monte Carlo sampling

Monte Carlo approaches utilize repeated (random) sampling from a predefined distribution to produce output ranges. The number of model simulations range typically between hundreds to thousands (Skeffington 2006). Barkman et al. (1995) employed 500 simulations to estimate uncertainty in 128 forest sites in southern Sweden. Barkman and Alveteg (2001b) used 1200 replicate calculations per site for Swedish forest soils; Skeffington et al. (2006) used a sample size of 1000, while Heywood et al. (2006c)

employed 1000 and 2000 model simulations to ensure uncertainty estimates were stable for sites in the United Kingdom. In the current study 1000 replications, following Skeffington et al. (2006), were used.

3.3. STAGE III Spatial aggregate level

Median values of CL and EX estimates were extracted from frequency distributions representing the 1000 MC simulations for each spatial aggregate. Subsequently, the coefficient of variation (CV), the ratio of standard deviation over the mean, of CL was calculated (CL_{CV}) for each aggregate (from 1000 simulations). The CV was utilized as a measure of uncertainty (dispersion of values) around the selected CL for each spatial aggregate. The probability of exceedance (EX_{PROB}) for each spatial aggregate was expressed as percent of positive EX values ($EX > 0$) of all 1000 MC simulations.

3.4. STAGE IV AURAMS level

All four estimates derived in STAGE III were summarized for each AURAMS GRID within the study area. Median CL and EX values, CL_{CV} and EX_{PROB} for each spatial aggregate were sorted independently in ascending order within each AURAMS GRID; the cumulative area of their corresponding spatial aggregate was calculated and converted into percent (%). The median CL and EX, CL_{CV} and EX_{PROB} values were then plotted against the proportion of area they represented. These area weighted cumulative distribution functions (CDF) then allowed calculation of the 5th percentile CL (5CL), 95th percentile CL_{CV} (95 CL_{CV}), 95th percentiles of EX (95EX) and 95th percentile EX probability (95 EX_{PROB}). Levels (percentiles) were set to protect 95 percent of the area of each AURAMS GRID.

3.5. STAGE V Regional or national level

The 5CL, 95 CL_{CV} , 95EX and 95 EX_{PROB} were mapped for the study area. In order to facilitate visual uncertainty assessment three map-types were created. In first map, categories for coefficient of variation (represented by different circle sizes) were plotted against ranges of critical loads values (indicated by different colours) for each AURAMS GRID. The 95EX and 95 EX_{PROB} were plotted in a similar manner. The second map represented value ranges of 95EX and were indicated by a colour, while circle sizes identified the 95 EX_{PROB} category. The following five categories (based on methods by Skeffington et al. 2007b) of 95 EX_{PROB} were used (1) 0–5 % probability: unlikely to be exceeded; (2) 5–25 % probability: relatively low risk of exceedance; (3) 25–75 % probability: potential risk of exceedance; (4) 75–95 % probability: relatively high risk of exceedance; and (5) > 95 % probability: highly likely to be exceeded. These five categories were redefined into three levels of exceedance risk: low (< 25 %), potential (25–75 %) and high (> 75 %) to indicate uncertainty in exceedance for the study area (third map-type).

4. Results

4.1. Mineral soils and spatially unique units

The proportion of mineral forest soils and count of spatial aggregates in the Ontario-Quebec region varied for the AURAMS GRIDS within the study area. The proportion of mineral soils ranged between 14 and 95 % of the grid in the study area (Figure 3A). Smaller proportions were observed in the southwest of the study area, with mineral soils occupying less than 50 % of AURAMS GRIDS. The number of spatial aggregates in the study area ranged between 4 and 230, with the majority of AURAMS GRIDS containing 50 or fewer spatial aggregates (Figure 3B).

Quantification of the proportion of mineral forest soil (the receptor) was deemed important for presentation of CL and EX estimates and their aerial implication. For example, the same (high) EX value for an AURAMS grid with 15 % of mineral soils as compared to a grid with 90 % mineral soil may differ in significance for emission reduction considerations.

4.2. Critical load uncertainties

The area weighted 95th percentile coefficient of variation and 5th percentile CL varied among the AURAMS GRIDS (Figure 4). AURAMS GRIDS stretching from the southwest to the northeast of the study area were characterized by the smallest deviation from the mean (i.e., the lowest 95CL_{CV} values) and typically lower 5CL values. Consequently, the confidence in the 5CL estimates plotted for those GRIDS was high. In contrast, decreased confidence or increased uncertainty in 5CL estimates was observed for AURAMS GRIDS characterized by high 95CL_{CV} values. The grids with higher uncertainty (larger 95CL_{CV} values) were primarily located in the northwest and southeast portion of the study area and represented a range of low to high 5CL.

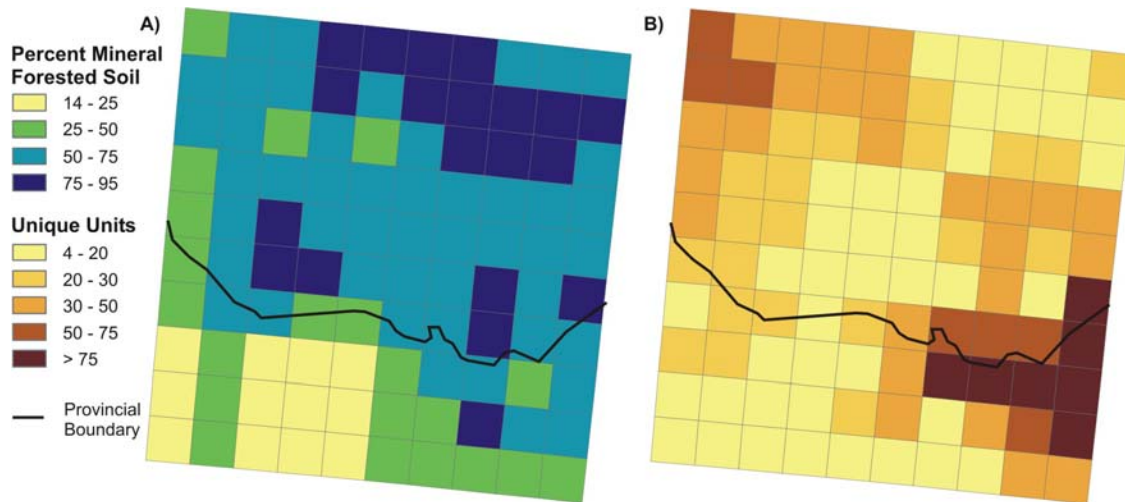


Figure 3. Map of the study area representing (A) the proportion (%) of mineral forest soils within each AURAMS GRIDS, and (B) the number of spatial aggregates, defined as unique combination of input values (based on spatial coincidence of data-sets used to calculate CL and EX). The number incorporates the count of all mineral soil components (soil types) within each Soil Landscape of Canada unit. The line indicates the provincial boundary.

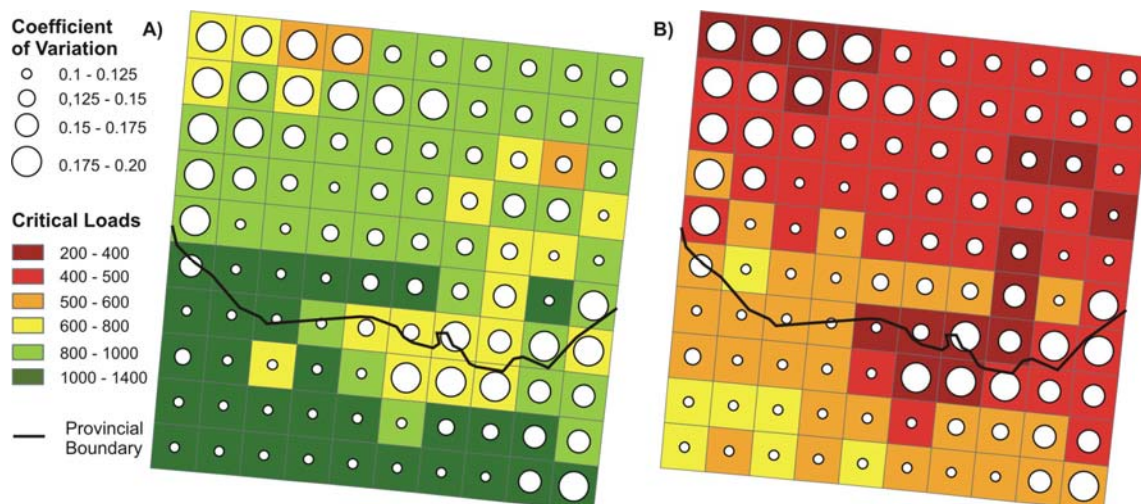


Figure 4. The area weighted 5th percentile critical load (5CL) estimated using a critical alkalinity leaching rate based on a Bc:Al ratio of (A) 1 and (B) 10. Area weighted 95th percentile coefficient of variation (95CL_{CV}) is also mapped. The coefficient of variation was derived using ratio of standard deviation over mean representative of 1000 simulations for each spatial aggregate within an AURAMS GRID.

The $95CL_{CV}$ values represent a relative assessment of uncertainty (classification of high versus low uncertainty) for the study area. The absolute ranges of the coefficient of variation, and consequently confidence level of CL estimates, will differ depending on region. For regional comparison another study (test) area was selected in the northeast portion of Alberta, the Athabasca Oil Sand Region (AOSR; the framework application to AOSR is presented in Appendix V).

As expected the 5CL were higher when estimated using a critical chemical criterion (Bc:Al) for forest soil protection of 1 compared to 10. The 5CL ranged between 535–1400 eq ha⁻¹ yr⁻¹ (Figure 4A) and between 292–677 eq ha⁻¹ yr⁻¹ (Figure 4B) for estimates based on Bc:Al of 1 and 10, respectively

In addition to incorporating uncertainty, the framework differs from previous approaches to estimate CL and EX in Canada (e.g., Carou et al. 2008). In the current study, the data were summarized based on the spatial structure of sulphur and nitrogen deposition (AURAMS GRID) and therefore the assessment is particularly relevant to deposition scenario analysis. Furthermore, a more complete representation of soil units was accommodated in the estimation of the 95th aerial percentile defined for ecosystem protection. A similar pattern was observed between median CL estimates in the current study (Figure 5) and previous studies (Carou et al. 2008). In contrast, the 5th percentile CL in both studies (Figure 4B and e.g., Carou et al. 2008, Figure 1) were quite different, with the current study showing lower critical loads.

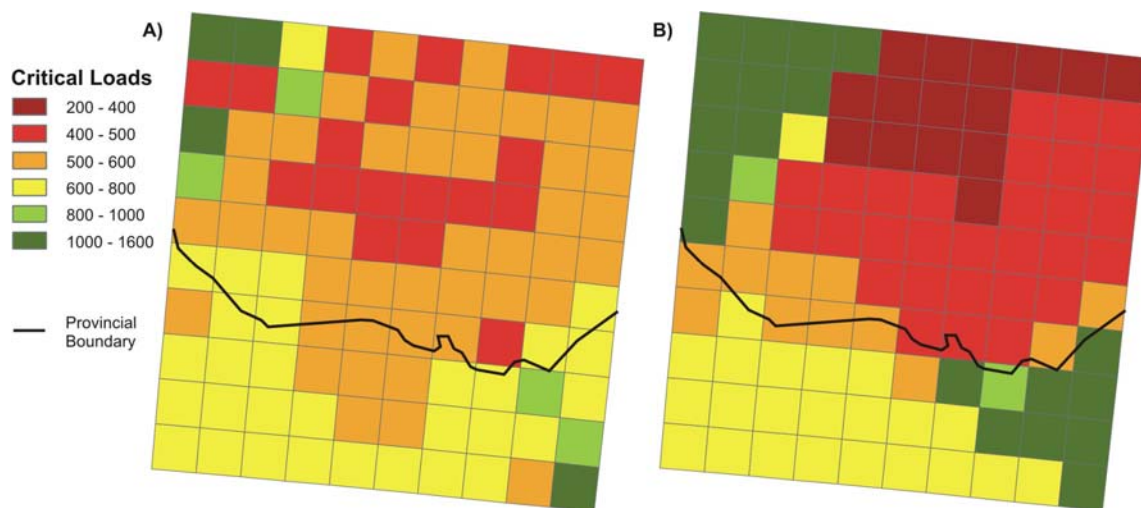


Figure 5. Comparison of the median critical loads (estimated using Bc:Al of 10) for each AURAMS GRID based on (A) estimates derived in the current study (the area weighted median of spatial aggregates) and (B) medians derived for Canada-wide CL assessment (Carou et al. 2008). The primary difference in methods of CL estimates is the treatment of spatial units (e.g., (B) average of all soil components to represent soil unit, compared to spatial weighting in this study).

4.3. Exceedance uncertainties

The exceedance values (95EX) as well as the probability of exceedance ($95EX_{PROB}$) varied for different AURAMS GRIDS and the level of protection limit (Figure 6). The more stringent critical threshold (Bc:Al of 10) resulted in exceedance values for all GRIDS (95EX range between 22 and 800 eq ha⁻¹ yr⁻¹). Furthermore, all but one of the grids had a high probability of being exceeded under a Bc:Al threshold of 10, i.e., $\geq 75\%$ chance that the majority of the area within each AURAMS GRID would receive S and N deposition in excess of CL. In contrast, setting the protection limit to that commonly used in Europe (Bc:Al ratio of 1) indicated that only 41 of the 100 AURAMS GRIDS received S and N deposition levels in excess of their CL ($95EX > 0$ eq ha⁻¹ yr⁻¹), while the remaining GRIDS ($n = 59$) were expected to have no exceedance ($95EX < 0$ eq ha⁻¹ yr⁻¹). The exceedance probability derived using the critical ratio of 1 for AURAMS GRIDS ranged between 0 and 100 % for the study area. However, a comparison of exceedance values and their corresponding exceedance probabilities (Figure 7) indicated that certain grids were likely

to shift between exceeded and non-exceeded categories. Assignment of each AURAMS GRID to one of three exceedance uncertainty categories, based on the $95EX_{PROB}$ values (Figure 7), revealed that 58, 24, and 18 GRIDS had low (< 25 %), potential (25–75 %) and high (> 75 %) risk of being exceeded, respectively.

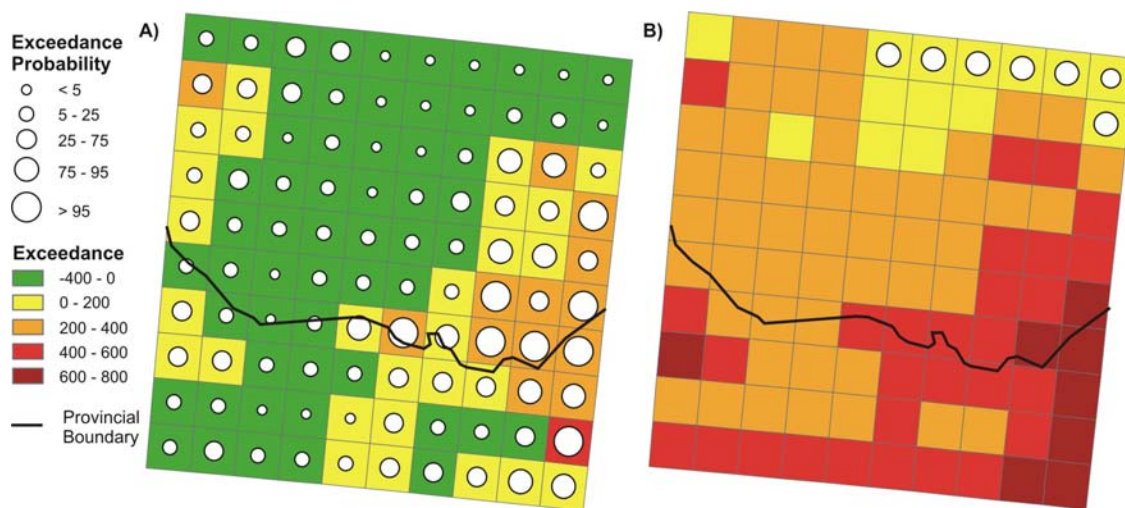


Figure 6. The area weighted 95th percentile exceedance ($95EX$) estimated using a critical alkalinity leaching rate based on a Bc:Al ratio of (A) 1 and (B) 10. The associated exceedance probability ($95EX_{PROB}$), indicating the likelihood of exceedance, is also shown (using 5 categories suggested by Skeffington et al. 2007b, see text for description). Unless otherwise shown, $95EX_{PROB}$ is > 95 %.

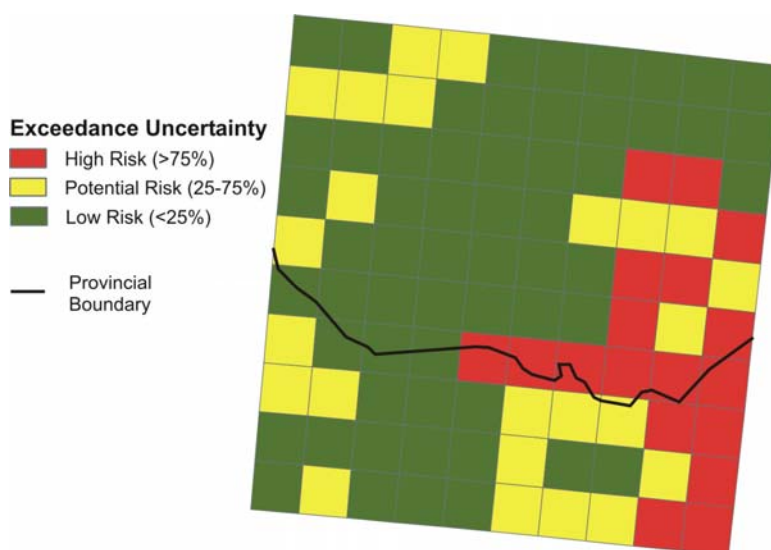


Figure 7. Uncertainty in exceedance (using $95EX$ values based on Bc:Al ratio of 1) under AURAMS 2002 sulphur and nitrogen deposition. The likelihood of exceedance is expressed by categorizing exceeded ($95EX > 0 \text{ eq ha}^{-1}\text{yr}^{-1}$) and not exceeded grids ($95EX < 0 \text{ eq ha}^{-1}\text{yr}^{-1}$) according to their exceedance probability ($95EX_{PROB}$). The $95EX_{PROB}$ categories were reduced to three (from five listed by Skeffington et al. 2007b, see description in text) categories that included low risk (< 25 %), potential risk (25–75 %) and high risk (> 75 %) of exceedance.

5. Summary and recommendations

The framework provided an assessment of the uncertainty for 5CL and $95EX$ estimates based on frequency distributions of input parameters; both the current state (CL and EX values) and deviation from that state ($95CL_{CV}$ and $95EX_{PROB}$) relative to current deposition level (2002) were presented. The maps

demonstrated that confidence in 5CL and 95EX estimates varied across the study region. The 5CL, 95EX and their associated uncertainties will differ depending on mapping grid spatial summary structure (e.g., summarizing values on GEM GRID rather than AURAMS GRID), or the uncertainty range specified for the SSMB model parameters. The framework allows for such changes, if deemed appropriate, without compromising the focus of the uncertainty assessment.

Quantification of any chemical threshold designed to protect desired ecosystem is intrinsically uncertain (e.g., Skeffington et al. 2007b). As a result, incorporation of such threshold in decision making will be associated with a level of risk (Barkman 1998). Explicit presentation of uncertainty in critical load estimates (rather than specifying a single value) is therefore considered more appropriate (Skeffington et al. 2007b). Policy makers may account for the risk by assessing the degree of confidence in estimates and plan according to the desired level of protection (Larssen et al. 2007).

The proposed framework is applicable to a Canada-wide uncertainty assessment of CL (acidity) and EX estimates for terrestrial mineral forest soils. A national application of the framework should incorporate several amendments. Firstly, the uncertainty ranges should be revised to accommodate regional differences, as confidence in input parameters (based on data sources) varies across Canada. For example, wider uncertainty ranges should be implemented for base cation and chloride deposition in British Columbia, where 35 km × 35 km deposition maps are not currently available. Narrower uncertainty ranges may be specified for areas where higher resolution soil data is available and spatial aggregates may be further delineated to incorporate forest types (instead of single forest cover).

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Appendix I. The Steady-State Mass Balance model

Long-term critical load of sulphur (S) and nitrogen (N) to forest soils may be estimated using the Steady-State Mass Balance (SSMB) model. The SSMB model assumes a simplified, steady state input-output description of the most important biogeochemical processes that affect soil acidification. Potential ecosystem inputs include atmospheric deposition of sulphate (SO_4^{2-}), N (nitrate and ammonium), chloride (Cl^-), sodium (Na^+), calcium (Ca^{2+}), magnesium (Mg^{2+}) and potassium (K^+), and soil base cation weathering rate. Ecosystem outputs and consumption include net removal of nutrients by forest harvesting, nutrient loss through soil leaching, denitrification and N immobilisation. The SSMB model, described in detailed by the ICP M&M (UBA 2004), estimates critical load of sulphur, $\text{CL}(\text{S})$, and nitrogen, $\text{CL}(\text{N})$:

$$(A.1) \quad \text{CL}(\text{S}) + \text{CL}(\text{N}) = \text{BC}_{dep} - \text{Cl}_{dep} + \text{BC}_w - \text{BC}_u + \text{N}_i + \text{N}_u + \text{N}_{de} - \text{Alk}_{le,crit}$$

where BC_{dep} = base cation ($\text{BC} = \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+ + \text{Na}^+$) deposition, $\text{Cl}_{dep} = \text{Cl}^-$ deposition, BC_w = base cation weathering, BC_u = net base cation ($\text{BC} = \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+$) uptake by trees (harvesting removal), N_i = net N immobilisation rate in soil, N_u = net N uptake by trees, N_{de} = net denitrification rate, and $\text{Alk}_{le,crit}$ = critical alkalinity leaching rate. Units of $\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$. This formulation has been rewritten by the NEG-ECP to estimate the long-term critical loads of sulphur plus nitrogen $\text{CL}(\text{S+N})$:

$$(A.2) \quad \text{CL}(\text{S} + \text{N}) = \text{BC}_{dep} - \text{Cl}_{dep} + \text{BC}_w - \text{BC}_u + \text{N}_i + \text{N}_u + \text{N}_{de} - \text{Alk}_{le,crit}$$

Under the NEG-ECP protocol, harvesting removals were not considered; therefore, long-term net uptake of N and BC_u were set to zero. The long-term net denitrification was considered negligible in well-drained upland forest ecosystems. Similarly, the net N immobilisation in soils was also assumed to be negligible in the long-term since this process can be negative or close to zero with stand dynamics and natural disturbances such as fire (NEG-ECP 2001). The final model under the NEG-ECP protocol can therefore be simplified to:

$$(A.3) \quad \text{CL}(\text{S} + \text{N}) = \text{BC}_{dep} - \text{Cl}_{dep} + \text{BC}_w - \text{Alk}_{le,crit}$$

The critical alkalinity leaching rate for forest soils is estimated from a critical molar base cation to (inorganic) aluminium (Bc:Al) ratio in soil leachate and the gibbsite dissolution constant (K_{gibb}) which controls aluminium solubility in mineral soils (UBA 2004):

$$(A.4) \quad \text{Alk}_{le,crit} = -Q^{2/3} \cdot \left(1.5 \cdot \frac{\text{BC}_{dep} + \text{BC}_w - \text{BC}_u}{K_{gibb} \cdot (\text{Bc:Al})_{crit}} \right)^{1/3} - 1.5 \cdot \frac{\text{BC}_{dep} + \text{BC}_w - \text{BC}_u}{(\text{Bc:Al})_{crit}}$$

where Q is soil runoff rate or precipitation surplus ($\text{m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$), The NEG-ECP protocol used a Bc:Al ratio of 10, a $\log K_{gibb}$ of 9.0 and $\text{BC}_u = 0$ (as above); K_{gibb} is expressed as $\text{m}^6 \text{ mol}_c^{-2}$. In practice, the NEG-ECP protocol uses a function based on the total acid input to derive a critical chemical limit equivalent to a Bc:Al ratio of 10.

Exceedance (EXC) of steady-state critical load of S and N to upland forest soils, is calculated as the current total deposition flux of S plus N (nitrate plus ammonium) minus critical load:

$$(A.5) \quad \text{EXC} = \text{S}_{dep} + \text{N}_{dep} - \text{CL}(\text{S} + \text{N})$$

where S_{dep} = total (wet plus dry) S deposition and N_{dep} = total N deposition. Unit of $\text{mol}_c \text{ ha}^{-1} \text{ yr}^{-1}$; negative exceedance values represent regions that are 'not exceeded', i.e., soils will not acidify to a level where forest soil damage is expected.

Appendix II. Data sources

Table A1. List of variables, sources and data resolution for critical load model.

Variable category	Data source	Resolution
Climate (Q, T)	Long-term mean annual temperature (°C) and precipitation surplus or runoff (m) during the period 1961–1990 (Posch 2006 and New et al 1999)	35 km grid
Deposition (BC, Cl)	Environment Canada, data period 1994–1998. Annual average total deposition of ions to land cover type. The area covered by each land use type is derived from the USGS Global Land Cover Characterization database (GLCC Version 2 Simple Biosphere Model categories)	35 km grid
Deposition (S, N)	Annual sulphur and nitrogen deposition predicted by Environment Canada's AURAMS (A Unified Regional Air-quality Modelling System) acid-deposition model. NAESI second base case run 2002	42 km grid
Soil (weathering)	Soil Landscapes of Canada Working Group. 2005. Soil Landscapes of Canada v3.1.1 and v2.1. Agriculture and Agri-Food Canada	1:1,000,000
Forest cover	The area covered by each land use type is derived from the USGS Global Land Cover Characterization database (GLCC Version 2 Simple Biosphere Model categories)	1 km grid

Appendix III. Supporting maps and illustrations

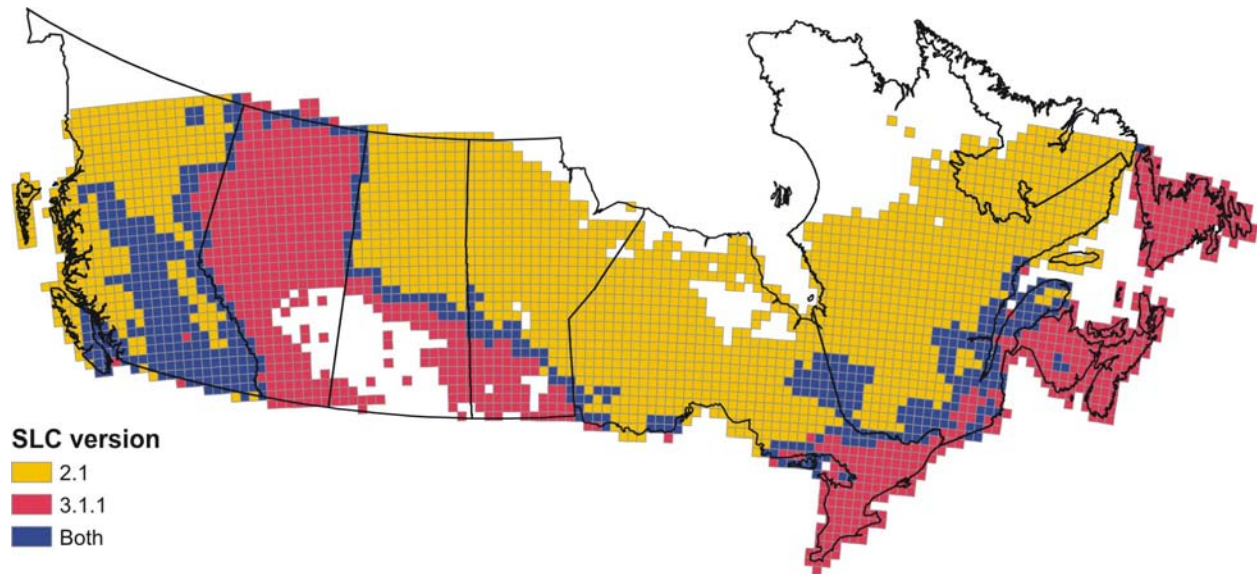


Figure A1. Spatial distribution of the availability of two versions of Soil Landscape of Canada (SLC) used in the current study. The map indicates the occurrence of each, or coincidence of both SLC versions within each AURAMS grid for each province.

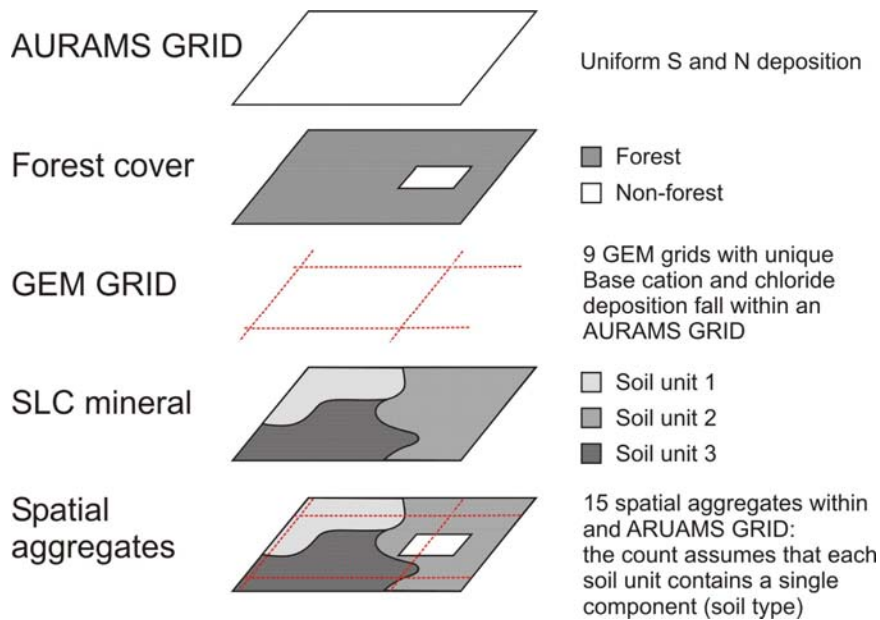


Figure A2. Illustration of the overlay procedure used to derive spatial aggregates to produce unique set of input variables for critical load and exceedance calculations for forested mineral soils.

Appendix IV. Uncertainty distribution types and ranges

Table A2. Summary of uncertainty estimates, distribution (D) and ranges, for inputs to the critical load (and exceedance) calculations from selected studies.

Source	Heywood et al. 2006c		Suutari et al. 2001		Reinds and de Vries 2009		Barkman and Alveteg 2001b	
Country	United Kingdom		Germany	Austria	Europe		Sweden	
Parameter	D	Range	Range		D	Range	D	Range
S _{dep}	N	25 % ^a					R+T	15 %
N _{dep}	N	25 % ^a			N	40 % ^a	R+T	20–30 % ^a
BC _{dep}	N	25 % ^a	±20 %	±30 %	Nt	40 % ^a	R+T	40 %
BC _w	R	±33–100 % ^b	±20 %	±40 %	Nt	10 % to 75 % ^b		20–100 % ^b
Bc _u	N	23 % ^a	±15 % ^a	±20 %			T	45 %
Q	N	23 % ^a	±15 %	±50 %	Nt	15 %	n.v.	n.v.
BC:Al	R	±50 % ^b	±10 %	– ^b			R	50 %
K _{gibb}	R	±50 % ^b	±20 % ^d	±20 % ^d			R	0.5 ^c
N _i	R	±50 % ^b	±5 %	– ^b	Nt	0.24		
N _{uptake}	N	27 % ^a	±15 % ^a	±20 %			T	45 %
N _{de}	R	±20–50 % ^{b,c}	±20 % ^c	– ^b	Nt	0.17–0.38 ^b		
N _{le(acc)}	T	–80 % to +25 % ^d						

Distribution type: N (normal), R (rectangular), T (triangular) and t (truncated). NOTE: n.v. is not varied.

Notes on selected studies (Table A2).

Heywood et al. (2006c): Steady-State Mass Balance model applied to a coniferous forest ecosystem; ^a Coefficient of variation $x\%$ = (mean value/standard deviation) \times 100; ^b Minimum value = mean value $- x\% \times$ mean value, Maximum value = mean value $+ x\% \times$ mean value; ^c Most likely values of the distribution have been assumed to be the default values; ^d Depending on soil type.

Suutari et al. (2001): Variables for Steady-State Mass Balance model; uniform distribution was assumed around median values for all variables; Parameters were assumed to be uncorrelated with the exception of base cation and nitrogen uptake (full correlation was assumed); ^a Derived from 10 % variation in BC/N content and biomass; ^b No value given; German values have been used; ^c Value originally provided was 10 %; ^d Assigned by the Coordination Centre for Effects (CCE: www.pbl.nl/en/themasites/cce).

Reinds and de Vries (2009): Very Simple Dynamic model in steady state (equivalent to Steady-State Mass Balance model) application to multiple sites across Europe; Range in parameter values identified as sigma (σ : standard deviation); ^a values measured at the site; ^b as a function of texture class and parent material.

Barkman and Alveteg (2001b): Used PROFILE to estimate uncertainty in forest soils in Sweden; ^a NO₃⁻ 20 %, NH₄⁺ 30 %; ^b derived from PROFILE, dependent on mineral type; ^c range in absolute value.

Appendix V. Application of the uncertainty framework to the Athabasca Oil Sand Region, Alberta

Study site

Athabasca Oil Sand Region (AOSR) is located in the northeast portion of Alberta (Figure A3). The region contains the largest deposits of recoverable oil in North America. As a result of industrial development (oil extraction) in the region, large increases in emission of S and N have been observed in the province (Environment Canada 2005). The impacts of acid deposition on forest soils in the region are of concern as a large proportion of northern Alberta has been classified as sensitive to acid deposition (Holowaychuk and Fessenden 1987) and further emission increases are expected due to continued industrial activity (Whitfield et al. 2009).

Critical load, coefficient of variation, exceedance and exceedance probability for the AOSR were determined using a critical threshold of $Bc:Al = 1$, following the uncertainty framework as previously outlined. It should be noted that uncertainty ranges for input data were consistent across both regions (Ontario-Quebec and AOSR); the objective of the current study was to develop an uncertainty framework and allow for a comparison between regions based on this approach. It is recognised that uncertainty ranges need to be revised for a more robust national-scale application.

Mineral soils and spatially unique units

The proportion of mineral forest soils and the number of spatial aggregates varied across the AOSR study area (Figure A4, Table A3). The forested mineral soils occupied between 9 and 83 % of the AURAMS GRIDS ($n = 32$) within the region. All but one grids contained ≥ 25 % mineral forest soil and over half ($n = 22$) of the grids contained ≥ 50 % mineral forest soil. The number of spatial aggregates ranged between 33 and 175, with the majority of grids ($n = 29$) with more than 50 spatial aggregates.

Critical load and uncertainties

Compared to the Ontario-Quebec test area (Figure 4A), forest soils in the AOSR appeared more sensitive to acid deposition (i.e., lower 5CL values, Figure A5, Table A3). In Alberta, 11 and 9 of the AURAMS GRIDS were categorized to have 5CL in ranges of 400–500 and 500–600 $\text{eq ha}^{-1} \text{yr}^{-1}$, respectively. In Ontario-Quebec, none of the grids had 5CL values $< 500 \text{ eq ha}^{-1} \text{yr}^{-1}$ and only 3 grids fell in the range of 500–600 $\text{eq ha}^{-1} \text{yr}^{-1}$ (Figure 4A). Note: map categories are consistent across the two study regions to facilitate comparison (see Figure 4A and Figure A5).

Although, the CL uncertainty estimates were similar for both regions (95CL_{CV} of 0.118–0.195 in AOSR, Table A3, and 0.100–0.200 in Ontario-Quebec, Figure 4A), a larger number of grids had higher coefficient of variation in the AOSR. The majority ($n = 26$ of 32) of AURAMS GRIDS had 95CL_{CV} ≥ 0.15 (Figure A5, Table A3) compared to less than half ($n = 34$ of 100) grids in the Ontario-Quebec region (Figure 4A).

Exceedance uncertainties

Despite generally lower 5CL compared to the Ontario-Quebec, few grids in the AOSR were classified at high risk of exceedance (Figure A5, Table A3). Sulphur and nitrogen deposition exceeded CL values with high exceedance probability ($EX_{\text{PROB}} \geq 75$ %) for 5 grids in the AOSR. The remaining AURAMS GRIDS in the region were classified to either have low risk of exceedance (i.e., $n = 24$ with $95EX_{\text{PROB}} < 25$ % and all grids with $95EX < 0 \text{ eq ha}^{-1} \text{yr}^{-1}$), or potential risk of exceedance (i.e., $n = 3$ with $25 \% \geq 95EX_{\text{PROB}} < 75$ %). Furthermore, 7 and 1 AURAMS GRIDS in the AOSR had 95EX value lower than the minimum ($< -400 \text{ eq ha}^{-1} \text{yr}^{-1}$, Figure 5, Table A3) and higher than the maximum ($\geq 800 \text{ eq ha}^{-1} \text{yr}^{-1}$, Figure 5, Table A3) reported for the Ontario-Quebec region, respectively.

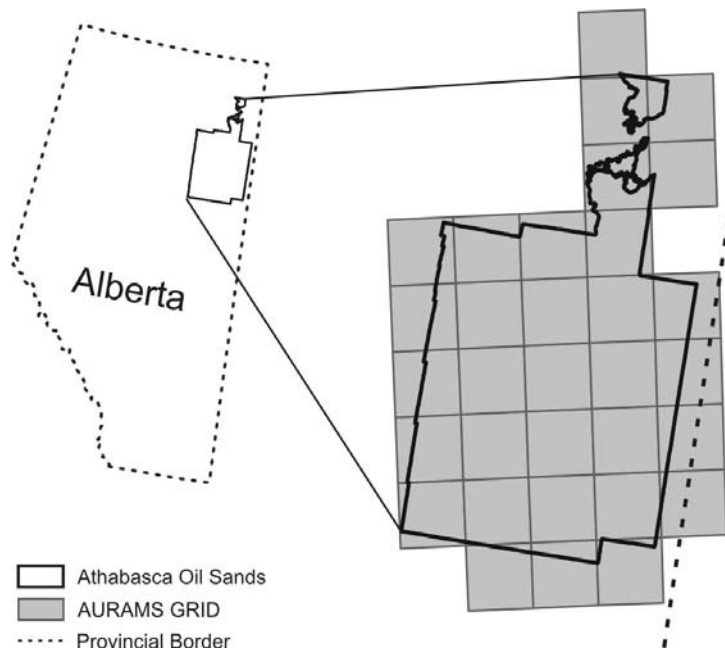


Figure A3. Map of Alberta with the location of the Athabasca Oil Sands Region (AOSR). The AOSR occupied 32 AURAMS GRIDS (map inset).

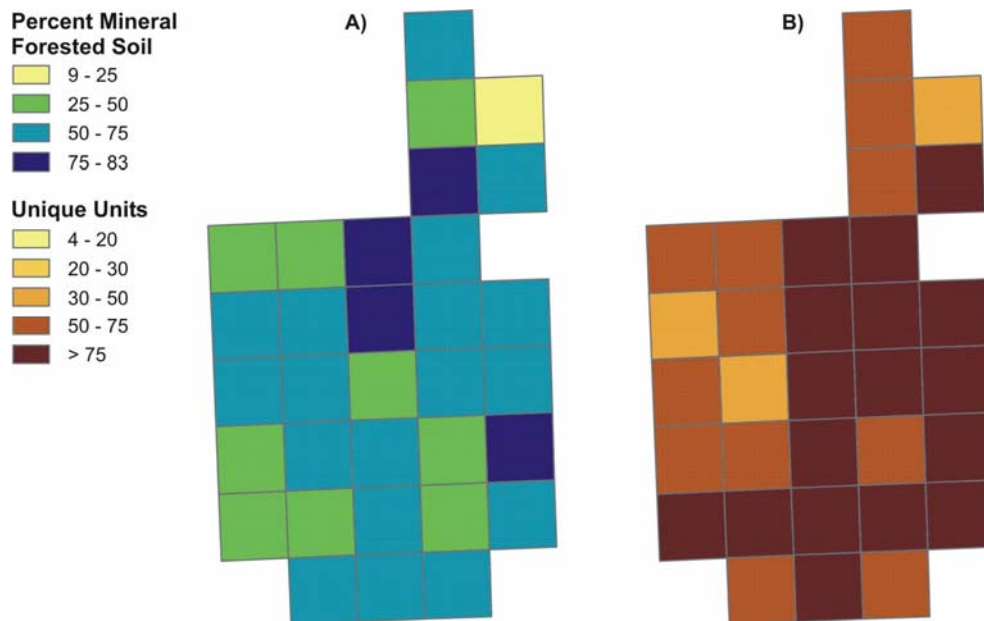


Figure A4. Map of the AOSR representing A) the proportion (%) of mineral forest soils within each AURAMS GRIDS, and B) the number of spatial aggregates, defined as unique combination of input values (based on spatial coincidence of data-sets used to calculate CL and EX). The number incorporates the count of all mineral soil components (soil types) within each Soil Landscape of Canada (SLC) unit.

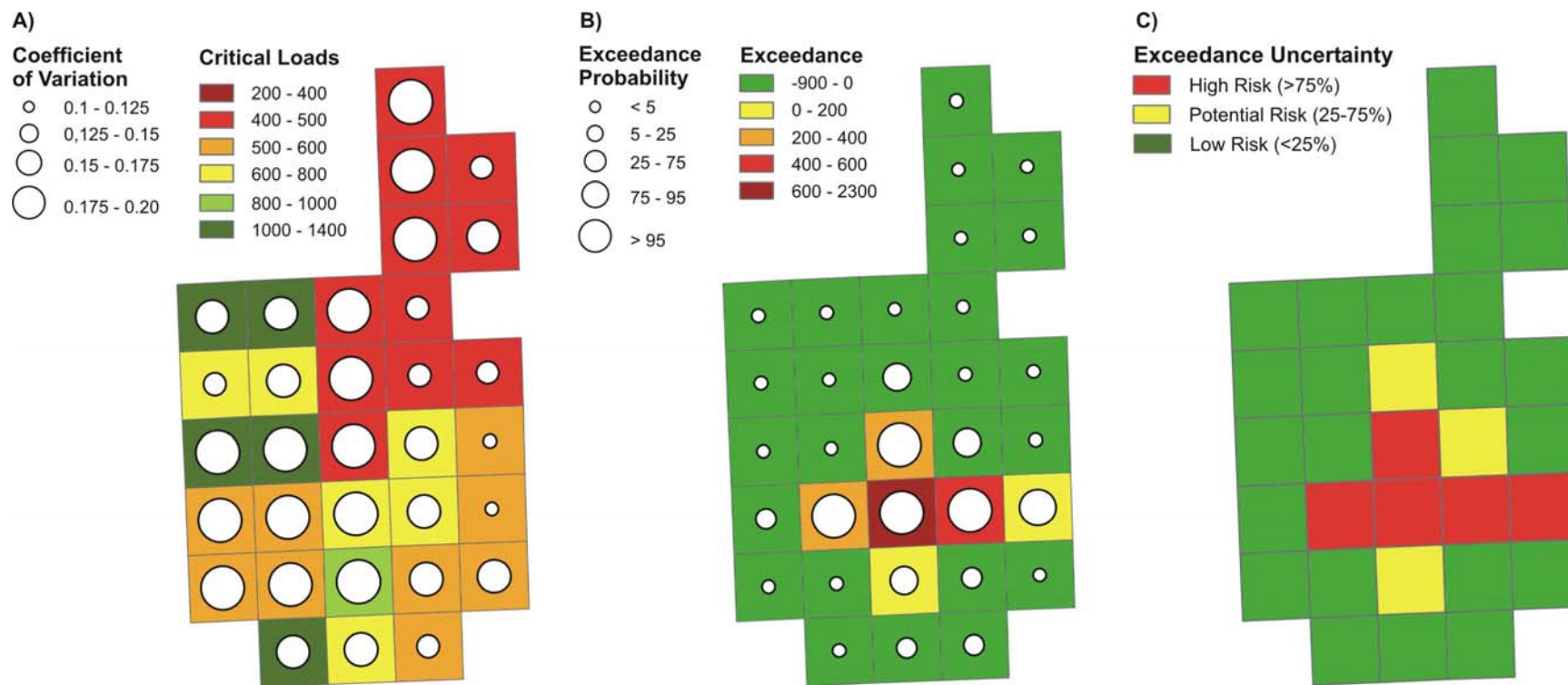


Figure A5. The critical load, coefficient of variation, exceedance and exceedance probability for AOSR was calculated using a critical alkalinity leaching based on a Bc:Al ratio of 1. The maps represent [A] the area weighted 5th percentile critical load (5CL) and 95th percentile coefficient of variation ($95CL_{CV}$); [B] the area weighted 95th percentile exceedance ($95EX$), and five categories of exceedance probability ($95EX_{PROB}$), indicating the likelihood of exceedance; [C] uncertainty in exceedance under AURAMS 2002 sulphur and nitrogen deposition. The likelihood of exceedance is expressed by categorizing exceeded ($95EX > 0 \text{ eq ha}^{-1}\text{yr}^{-1}$) and non-exceeded grids ($95EX < 0 \text{ eq ha}^{-1}\text{yr}^{-1}$) according to their exceedance probability ($95EX_{PROB}$). The $95EX_{PROB}$ categories were reduced to three categories that included low risk (< 25 %), potential risk (25–75 %) and high risk (> 75 %) of exceedance.

Table A3. Data summaries for each AURAMS GRID (42 km × 42 km) in the Athabasca Oil Sand Region: proportion of mineral forest soil (% Min. Soil), number of unique spatial aggregates (Count Units), 95th percentile exceedance (95EX) and exceedance probability (95EX_{PROB}), 5th percentile critical load (5CL) and 95th percentile coefficient of variation of the critical load (95CL_{CV}).

AURAMS	% Min. Soil	Count Units	95EX	EX _{PROB}	5CL	95CL _{CV}
48-93	41	111	-249.1	0	517.7	0.183
48-94	33	66	-89.6	8	507.0	0.189
48-95	50	58	-834.6	0	1087.6	0.190
48-96	65	40	-909.9	0	755.6	0.160
48-97	43	58	-918.2	0	1067.0	0.177
49-92	57	59	-790.8	0	1112.6	0.162
49-93	44	81	-178.4	0	527.0	0.183
49-94	51	74	216.0	99	508.4	0.193
49-95	66	50	-849.4	0	1280.4	0.189
49-96	66	59	-508.6	0	683.1	0.170
49-97	34	56	-889.7	0	1068.1	0.165
50-92	67	106	-109.9	21	775.7	0.165
50-93	61	175	34.0	57	922.6	0.186
50-94	65	140	2290.9	100	608.0	0.189
50-95	49	85	394.2	100	471.6	0.195
50-96	81	159	-33.4	30	462.8	0.191
50-97	80	109	-186.8	0	462.7	0.191
51-100	54	67	-296.9	0	418.5	0.194
51-92	62	72	-100.6	7	553.1	0.157
51-93	49	103	-104.3	24	533.0	0.176
51-94	49	67	418.1	100	702.1	0.170
51-95	59	102	-10.3	46	676.4	0.162
51-96	65	90	-135.3	1	471.4	0.131
51-97	74	85	-249.6	0	476.8	0.131
51-98	76	64	-303.6	0	467.3	0.196
51-99	46	65	-362.3	0	461.0	0.192
52-93	59	112	-171.3	1	598.0	0.174
52-94	83	98	108.0	89	590.3	0.118
52-95	74	98	-191.7	0	582.9	0.129
52-96	63	103	-169.3	0	471.2	0.130
52-98	65	91	-275.3	0	448.4	0.177
52-99	9	33	-311.7	0	436.8	0.134