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REPORT

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Abstract Calculations of critical loads for inputs of strong acids to lakes and exceedances of critical load can be a useful tool for the management of polluted areas. In Sudbury, Canada, large emissions of sulfur dioxide have resulted in widespread lake acidification in parts of northeastern Canada. Due to its vicinity to Sudbury, extensive lake acidification and fish loss has occurred in the Killarney Provincial Park. We have linked measured water quality to critical loads and strong acid deposition. Total S deposition decreased from an estimate of 70 meq m ⁻² yr ⁻¹ in 1981 to 47 meq m ⁻² yr ⁻¹ in 1997. The critical load was estimated to be exceeded in 57 % of the Killarney park area in 1981, whereas 38 % of the area was exceeded in 1997. A target load for total S deposition may be set at 30 meq m ⁻² , corresponding to 14.4 kg SO ₄ ha ⁻¹ yr ⁻¹ . This deposition level corresponds to a 2010-scenario according to the Clean Air Act Amendments with an additional 50 % emission reduction, and will protect the park area almost completely from exceedances of the critical load.

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Northern Lakes Recovery Study

Killarney Park Restoration Program

**Mapping of Critical Loads and Critical Load
Exceedances in the Killarney Provincial Park,
Ontario, Canada**

Preface

The Killarney Provincial Park south of Sudbury in Canada has suffered from the effects of acid deposition for several decades, and a restoration program has been established. The program asked the Norwegian Institute for Water Research (NIVA) (letter of October 4, 1996) to prepare a proposal for critical load mapping of the park. The proposal was mailed by November 19, 1996 and the project was confirmed by a letter of June 4, 1997 from the Cooperative Freshwater Ecology Unit at the Ontario Ministry of Natural Resources.

Our contacts in Sudbury have been Dr. John M. Gunn and Ed Snucins at the Cooperative Unit, Laurentian University in Sudbury, and we wish to thank them and their staff for providing the necessary data and for fruitful discussions and all help during the project period. We also thank Dr. Michael D. Moran at the Atmospheric Environment Services, Ontario for providing deposition data for Killarney.

The project is part of The Northern Lakes Recovery Study (NLRS) a cooperative study between Canada and Norway, designed to assess factors that affect the recovery of acid stressed lakes. It has received external funding from Falconbridge Limited in Sudbury. NIVA has also supported the project (E-88411).

Grimstad, June 12, 1998

Atle Hindar

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Summary

Calculations of critical loads for inputs of strong acids to lakes and the exceedance of critical loads has been shown to be a useful tool for the management of areas polluted by acid deposition. Mapping of these data visualize the extension of polluted areas and how seriously affected they are. In Sudbury, Canada, large emissions of sulfur dioxide have resulted in widespread lake acidification in parts of north-eastern Canada. The Sudbury emissions have been substantially reduced in the period 1970-1988, and even further thereafter. Due to its vicinity to Sudbury, extensive lake acidification and fish loss has occurred in the Killarney Provincial Park. After substantial reductions in emissions from the Sudbury smelters many lakes recovered during the 1980's. We have linked measured water quality to critical loads and strong acid deposition. These results may therefore be useful for evaluation of the potentials for further improving the conditions in the lakes.

Total S deposition was estimated to have decreased from about $70 \text{ meq m}^{-2} \text{ yr}^{-1}$ in 1981 to $47 \text{ meq m}^{-2} \text{ yr}^{-1}$ in 1997. A deposition scenario for 2010 predicts a decrease to $43 \text{ meq m}^{-2} \text{ yr}^{-1}$, and three other scenarios relate to this decrease. They represent an additional 25, 50 or 75 % emission reduction, respectively.

The critical loads for sulfur was estimated to have been exceeded in 57 % of the Killarney park area in 1981, whereas 38 % of the area was exceeded in 1997. Depending on which of the 2010-scenarios will be fulfilled, the critical load will be exceeded in 0 to 31 % of the park area.

A target load for total S deposition may be set at 30 meq m^{-2} , corresponding to $14.4 \text{ kg SO}_4 \text{ ha}^{-1} \text{ yr}^{-1}$. This deposition level corresponds to a 2010-scenario according to the Clean Air Act Amendments with an additional 50 % emission reduction, and will protect the park area almost completely for exceedances of the critical loads. A target load of $40 \text{ meq m}^{-2} \text{ yr}^{-1}$ will result in almost complete elimination of critical load exceedances above $15 \text{ meq m}^{-2} \text{ yr}^{-1}$. This load corresponds to a combination of the expected 2010 and the 2010 + 25 % - scenario.

To get a more complete picture of how the deposition scenarios will impact the species and the biodiversity of the park, two additional approaches may be taken. One is to study the possibility of developing dose-response relationships for different biological species analysed in the Killarney lakes. These relations could then be used for modifying critical load formulations specifically adapted to the conditions and the species occurring in the Killarney lakes. Thus, the critical load data would thereby be more directly related to biological species found in the park.

Another is simultaneous calculation of critical loads of acidifying N and S deposition and their potential exceedances. This may be done if specific data for nitrogen immobilisation and uptake are available. Whereas the Steady-State Water Chemistry (SSWC) model allows the calculation of critical loads of acidity (S+N) and their present exceedances, the First-order Acidity Balance (FAB) model (see Appendix A) is suitable for the extended work.

Nitrogen was not included in the calculations given in this first report because the nitrate lake-concentrations seemed relatively low and would not significantly change the exceedances. However, since the FAB-model seems to be applied as the basis for the second N-protocol, an assessment of the potential effects of N-deposition should also be carried out for the Killarney Provincial Park.

1. Introduction

Calculations of critical loads for inputs of strong acids to lakes and exceedances of their critical loads can be a useful tool for the management of polluted areas (Hettelingh et al. 1995). Mapping of these data visualize the extension of polluted areas and how seriously affected they are. In Europe this approach has been used for both surface water and soils (Henriksen et al. 1992; Hornung et al. 1995; Sverdrup et al. 1995), and as a basis for negotiations of emission reductions (UN/ECE 1994). The calculated exceedances are expressed as excess acid, and may therefore also be used to estimate the amount of limestone material necessary to neutralize acidified surface waters (Hindar et al. 1998).

In Sudbury, Canada, large emissions of sulfur dioxide have resulted in widespread acidification in parts of north-eastern Canada (Beamish and Harvey 1972). In the 1960's annual averages for the total Sudbury SO₂ emissions were about 2.2 million tons, which is of the same order of magnitude as the emissions from the Norilsk complex in Siberia, Russia. However, the Sudbury emissions were substantially reduced during the years 1970-1988 (Keller and Pitlabo 1986; Gunn and Keller 1990; Keller et al. 1992), and even further thereafter (Gunn et al. 1995).

During the period 1980-1993, US emissions contributed 86 % to the total sulfur emissions in eastern North America (Ro et al. 1997). Due to large reductions in emissions at Sudbury, one should expect the US emissions to have increased its relative impact on the Sudbury area in the last two decades.

Due to its vicinity to Sudbury, extensive acidification and fish loss has occurred in the Killarney Provincial Park located south of Sudbury. Fish loss was first documented by Beamish and Harvey (1972) for Lumsden Lake together with a substantial reduction in pH also in several other lakes in the Park. Today it is known that many of the over 200 lakes and ponds of the Killarney Park lost their populations of fish, plankton, benthic invertebrates, amphibians and aquatic birds as the lakes acidified in the 1960's and 1970's (Snucins and Gunn 1998).

After substantial emission reductions from the Sudbury smelters many lakes recovered during the 1980's (Keller and Pitlabo 1986; Gunn and Keller 1990), but after the emission reductions leveled off thereafter no further improvements were registered by Keller et al. (1992).

The critical load concept is a useful tool to predict future conditions in acidified lakes based on given deposition scenarios. This report assesses the past, present and future conditions of the lakes in the Killarney Park, based on given deposition scenarios.

2. The study area

2.1 Killarney Park Area

Killarney Provincial Park (46°3'N, 81°21'W) is situated approximately 60 km south-west of Sudbury, Ontario (

Figure 1). This 48,500 ha wilderness area is part of the La Cloche Mountains located close to the Georgian Bay in the north-eastern part of Lake Huron.

The over 200 lakes and ponds in the park are widely different in size, elevation, catchment characteristics and water chemistry, and they have therefore reacted differently to acid deposition (Snucins and Gunn 1998). Most of the park is underlain by ridges of quartzite (

Figure 2), which is extremely weathering resistant. In these areas many of the lakes are acidified to water quality levels below threshold limits for many fish species.

The park area is now part of a national ecological monitoring and assessment network (EMAN), and will continue to be the site of long-term monitoring of the impacts of a variety of environmental stressors.

The Sudbury area has a long-lasting winter. The combination of a large fraction of the precipitation falling as snow and low annual runoff have impacts on flushing patterns and retention times of the lakes in the area. Mean annual runoff rates for the Sudbury area is 0.4 m yr^{-1} (Keller and Carbone 1997) and 0.35 m yr^{-1} for Killarney (Government of Ontario 1984).

2.2 Sulfur Emissions

Total S-emissions in eastern Canada and eastern United States declined from 10.6 MT/yr in 1980 to 8.6 MT/yr in 1986, followed by fairly constant emissions until 1993. 86 % of the total emission was from the United States (Ro et al. 1997). Correspondingly, wet deposition decreased by 18.2 % in the ten-year period 1980-83 to 1990-93. 40 % of the total deposition to this area occurred in Canada.

Emissions of SO_2 from the Sudbury smelters declined from 2.2 million tons yr^{-1} in the 1960's to 0.72 million tons yr^{-1} in 1988 (Gunn and Keller 1990), a reduction of almost 70 %. Thereafter, further reductions have occurred. Combined 1995 SO_2 -emissions from the INCO Ltd. and Falconbridge Ltd. smelters were not allowed to exceed 0.365 million tons annually (Gunn et al. 1995). The Sudbury emissions are thus reduced by more than 80 % since the 1960's.

The total emissions from INCO Ltd. and Falconbridge Ltd. smelters in Sudbury used to be of the same magnitude as from the Norilsk smelter in the Siberia part of Russia. After the emission reductions the annual emission from the INCO smelter is now about equal to the largest emitters in Europe. The 1997-emissions of SO_2 from the 10 largest emitters in Europe were in the range 0.28-0.70 million tons yr^{-1} .

After the substantial emission reductions in Sudbury and with the prevailing wind directions, US emissions are the most significant contributors to present-day acidification status in the Killarney park area. The future acidification status of the park will therefore be strongly dependent on further emission reductions both in Canada and the US.

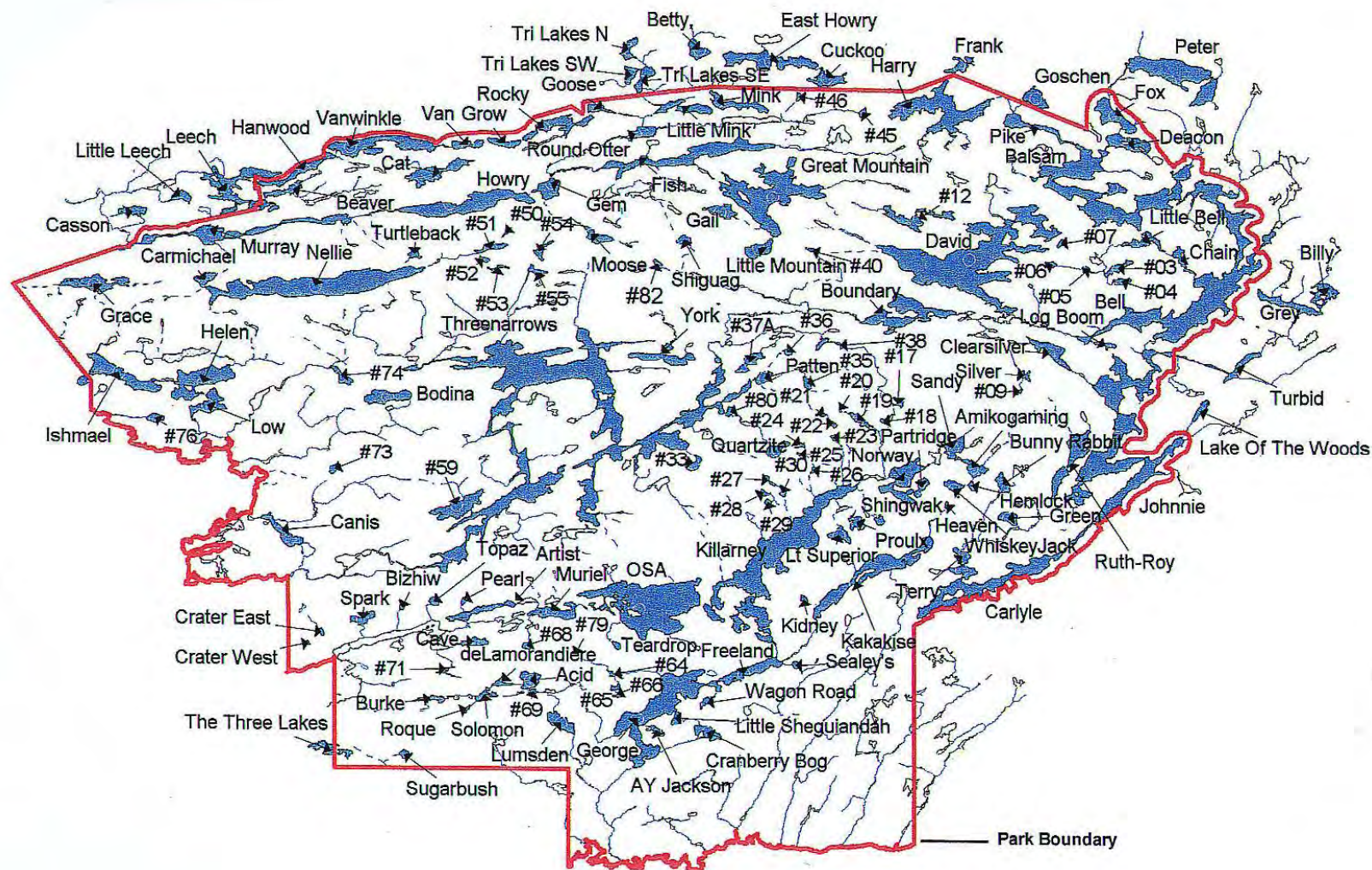


Figure 1. Location of water sampled lakes of the Killarney Provincial Park.

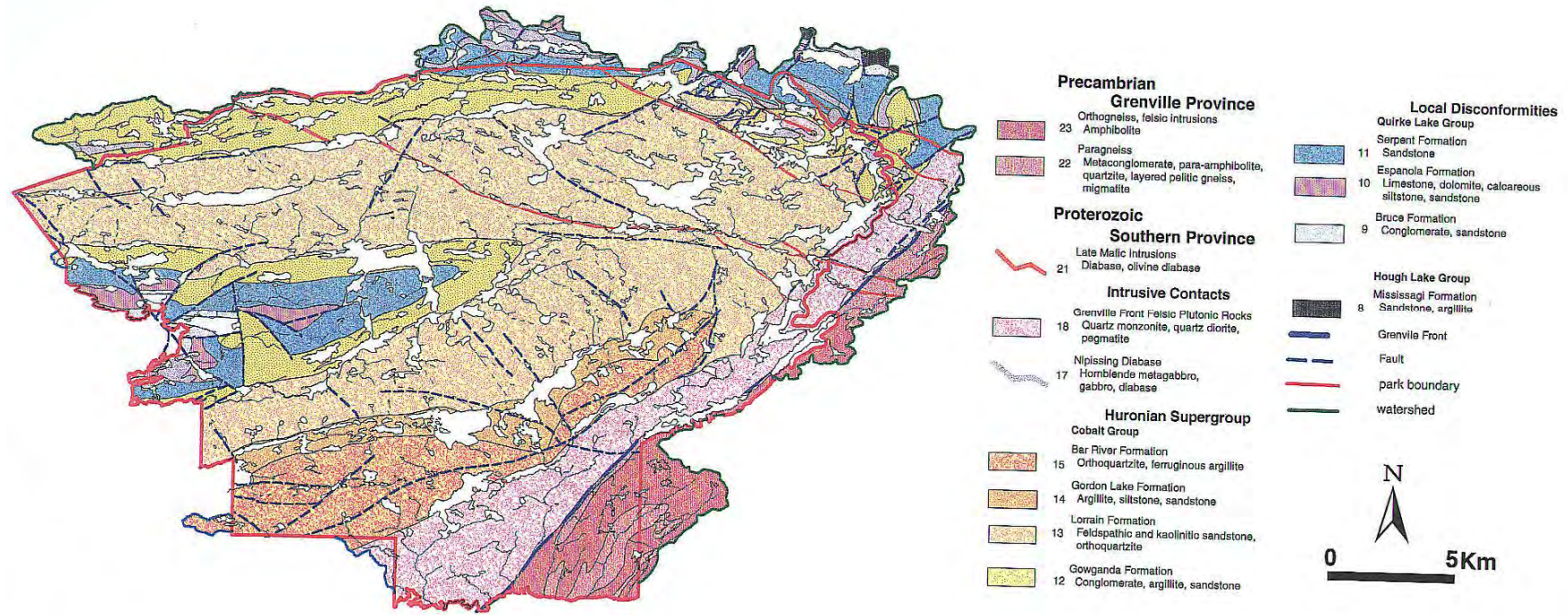


Figure 2. Geological map of Killarney (Adapted from Ontario Dept. of Mines and Northern Affairs maps by Snucins and Gunn 1998).

3. Methods and Material

3.1 Lake sampling and chemical analyses

A synoptic survey of 151 lakes in the Killarney Provincial Park was carried out during the period 23 January to 13 February, 1996 (Snucins and Gunn 1998). Tube composite water samples were collected from each lake by lowering a 2.5 cm diameter plastic Tygon tubing through the ice to a depth of 5 m, or if shallower, to 1 m above the substrate.

pH was analyzed at the Coop Unit in Sudbury. The pH data were checked by comparing split samples of pH from the Coop Unit and the Ontario Ministry of Environment laboratories in Dorset and Toronto. Samples for analyses of pH, major ions, dissolved carbon and nutrients were submitted to the Ontario Ministry of Environment Laboratory in Toronto.

Water chemistry data together with a mean runoff of 0.35 m yr^{-1} (Government of Ontario 1984) was used to calculate the critical load for each lake. The Killarney Park area is probably too small to have large variability in run-off. However, the run-off may vary considerably from year to year. No data exists, however, from the area to document patterns and between-year differences.

Time trends for pH and sulfate from eight intensively sampled lakes in the park for the period 1980-1996 was estimated by simple regression analysis. The lakes are: David, Bell, Johnnie and Ruth-Roy in the Johnnie Lake catchment, Killarney and George in the George Lake catchment, O.S.A. Lake and Nellie Lake.

All morphometric data of the lakes and their watersheds are from the Coop Unit in Sudbury (Snucins 1996).

3.2 Critical loads and their exceedances

The critical load of acidity to surface waters is equal to the weathering rate of the lake catchment less a given amount of buffer to keep water quality conditions acceptable to specified aquatic organisms (e.g. fish). The weathering rate is estimated from present day water chemistry (see Appendix A for details). At present two steady state models are used for calculating critical loads of acidity to surface waters; the Steady State Water Chemistry (SSWC) method and the First-order Acidity Balance (FAB) model. Steady state models predict the final effect of induced changes, but do not consider the time span to reach steady state. To estimate the time aspect the more complex dynamic models must be used. These models, however, requires much more data to be operative than the simple steady state models. The SSWC-model is the simplest one and requires weighted yearly average water chemistry or an estimate thereof and yearly runoff to calculate critical loads, whereas the FAB-model also requires lake and catchment data. (see Appendix A).

The exceedance of critical loads is calculated by deducting the critical load from the deposition of sulfur (and nitrogen). If the deposition is higher than the critical load, then it is exceeded. In this report we have used historic, present and future deposition scenarios.

The deposition scenarios used are based on the 1989-emission and deposition data. Deposition scenarios for 1994, 1997 and 2010 were calculated by Michael D. Moran at the Atmospheric Environment Service, Ontario, Canada. In order to estimate the 1981-deposition we used the 1989-deposition and lake sulfate trends for eight Killarney lakes. We found a significant ($r^2=0.69$; $p=0.0001$) mean trend of $-0.158 \text{ mg SO}_4 \text{ L}^{-1} \text{ yr}^{-1}$. This trend was used to estimate the SO_4 deposition in

the year 1981, see Table 1. Although this approach may be questionable we obtained a figure that corresponds well with observed regional-scale deposition and changes in emissions over this period (Moran, pers. comm.). All deposition scenarios used are presented in Table 1.

We have calculated the critical load values based on sub-catchments instead of whole lake catchments. This approach leads to several options in selecting appropriate sub-catchments. Here are some examples of the approaches that have been used;

- If CL is based on the water chemistry of a lake with a large catchment, much of the nuances in the catchment may be hidden. To increase the resolution, smaller lake catchments were used as a basis. If CL could be calculated for upstream catchments of a lake, data from the lake itself was used to calculate CL for the sub-catchments not accounted for.
- If a lake catchment consisted of two or more totally different geological formations with respect to weathering capacity, misleading CL-estimates may be the result. Therefore, part of the catchment (the central 71 km²) of Lake Threenarrows (LTN) was separated into three sub-catchments, which then were represented by three different CL-values; LTN-north by CL of Carmichael (downstream Nellie), LTN-south by CL of O.S.A. and LTN-middle by a calculation based on LTN water chemistry and CL of the other sub-catchments of LTN.
- Another situation emerges for catchments without lakes or without lake-water chemistry data. Such catchments may be represented by the CL of a nearby lake. Care should be taken to secure that this "stand in" lake is situated on the same type of bedrock as the catchment involved. This approach was used for the triangle north of Collins inlet in the southernmost part of the park.
- Adjacent lakes may have almost identical CL-values. Some of the catchments were therefore combined.
- Some of the water input to the park area is from catchments and lakes outside the park border. Fortunately, lake water samples were also collected from the most important ones of these lakes. To obtain an optimal resolution these areas are included in the calculations and also in the sum of sub-areas. This explains why the calculated total area of the park was temporarily increased from 45000 ha to 52700 ha.

If the chemistry data for Lake Threenarrows had been used for calculating CL for its whole catchment (103 km²), we would not have found any exceedances based on the 1994-deposition scenario and the predicted future scenarios. By splitting the whole catchment of Lake Threenarrows into 11 separate sub-catchments, however, a much more detailed picture emerged.: half the lake catchment area will be exceeded even in 2010 according to the CCUSA2-scenario (Table 1), as presented below. This illustrates how important the catchment resolution is for the results.

Table 1. Emission and deposition scenarios used in this report. Most of the data are from Moran (1997) and Moran (pers. comm.). The deposition based on a declining trend in lake-sulphate and 1989 as a base (Lakes-trend, see text) is by the authors.

Scenario	Year	Alternative emiss.red.	Emission				Deposition		
			Can. KTonnes SO2/yr	U.S. SO2/yr	Tot. SO2/yr	Frac. of 1989	Moran, wet kg SO4/ha/yr	Moran, total kg SO4/ha/yr	Total meq/m2
Lakes-trend	1981								69.0
BASE	1989		2688	17511	20199	1.00	21	26.3	54.7
CCONLY*	1994		1939	17862	19801	0.98	19.8	24.8	51.6
CCUSA1**	1997		1939	14865	16804	0.83	18.1	22.6	47.1
CCUSA2**	2010		1939	12446	14385	0.71	16.7	20.9	43.5
25FCAP	2010	- 25%	1739	9335	11074	0.55	14.4	18.0	37.5
5CCUS2	2010	- 50%	969	6223	7192	0.36	11	13.8	28.6
75FCAP	2010	- 75%	579	3112	3691	0.18	7.4	9.3	19.3

*Referring to the 1985 Eastern Canada Acid Rain Control Program

**Referring to the 1990 U.S. CAAA (Clean Air Act Amendments)

BASE	1989
CCONLY*	Canadian controls only
CCUSA1**	First phase of the US SO2 emissions reductions
CCUSA2**	Second phase of the US SO2 emissions reductions
25FCAP	2010 emissions reduced by 25 % (2030)
5CCUS2	2010 emissions reduced by 50 % (2030)
75FCAP	2010 emissions reduced by 75 % (2030)

4. Results and Discussion

4.1 Lake water chemistry

The Killarney lakes are obviously different in many respects. Three main factors seem to be important, deposition of SO_4 , geology and soil type in the lake catchments.

The S-deposition is high and results in elevated concentrations of SO_4 in the lakes (Figure 3 and Table 2). Depending on the geology of the catchment, this strong external acid impact may lower the pH. Lakes that are situated on easy weathered bedrock, as in the central-western part and along the park-border in south, east and north, may nevertheless have pH above 6.0 and up to the maximum of 7.3.

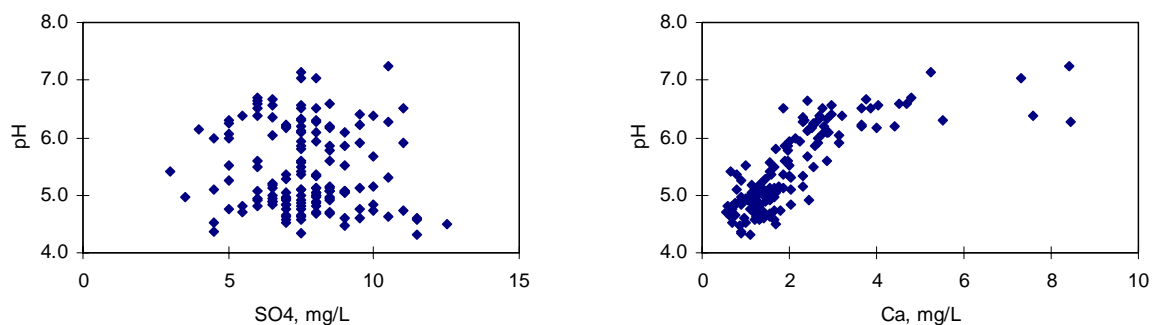


Figure 3. Relationship between pH and sulfate and calcium, respectively, in Killarney lakes. One observation ($> 10 \text{ mg L}^{-1} \text{ Ca}$) is omitted.

Figure 3 shows that the SO_4 concentrations in the park lakes are high; $4\text{--}12 \text{ mg L}^{-1}$ ($80\text{--}250 \mu\text{eq L}^{-1}$). Due to the high SO_4 -deposition, two groups of lakes are found in the park. The first group consists of lakes in the areas with easy weathered bedrock and thus relatively high concentrations of base cations. In this area pH is acceptable in spite of the strong acid load. In the second group, those in the horseshoe-shaped quartzite area, Ca is low and pH is well below 5.0 in many lakes due to low weathering rates of the bedrock. If the Ca-concentration is below 1 mg L^{-1} ($50 \mu\text{eq L}^{-1}$), pH is usually below 5.5. If the Ca-concentration is above 3 mg L^{-1} ($150 \mu\text{eq L}^{-1}$), pH is above 6.0.

Dissolved organic material may ameliorate the toxicity of aluminum, and may therefore be important for the structure of the biological community. The park lakes are more colored than acidified lakes in e.g. Norway, and half of them have concentrations above $3.4 \text{ mg L}^{-1} \text{ DOC}$. DOC tends to form in catchments rich in bogs. High DOC-concentrations may thus be more characteristic for lakes in the lower part of the park, typical for the areas of more favorable geology. If lakes with $\text{DOC} > 3 \text{ mg L}^{-1}$ are omitted, most of the high-pH and high-Ca lakes disappear (Table 2), confirming this assumption.

As a result of the marked reduction of SO_4 during the last 15 years (Figure 4) one may expect the pH to improve. This has also been documented (Figure 5). In this way the two lake-pH groups may be closer now than a few decades ago and will probably be even more close in the future.

Table 2. Summary of lake-chemistry characteristics. Data from Snucins and Gunn (1998).

Variable	Unit	Number of lakes	of 90 % of lakes less than	50 % of lakes less than (median)	Mean
SO ₄	mg SO ₄ L ⁻¹	151	10.0	7.5	7.63
pH		151	6.40	5.14	
pH (DOC<3)		66	5.51	4.98	
Ca	mg L ⁻¹	151	3.65	1.65	2.15
Ca (DOC<3)	mg L ⁻¹	66	2.0	1.38	1.41
DOC	mg L ⁻¹	151	8.1	3.4	4.15
TAI	µg L ⁻¹	151	500	169	217

The trend in SO₄ for single lakes is mainly a result of a gradually reduced S-deposition. Relatively large lakes, with long retention times, tend to respond slower to changes in external inputs. The decreasing SO₄-trends of the eight Killarney-lakes were all significant (p<0.05) and were tested for retention-dependent lags. However, no significant contribution from the lake-retention time to the variance in trends was found. This may be explained by the gradual decrease in deposition of SO₄ over many years which has resulted in a more or less steady decrease in lake-SO₄ for all lakes in the region.

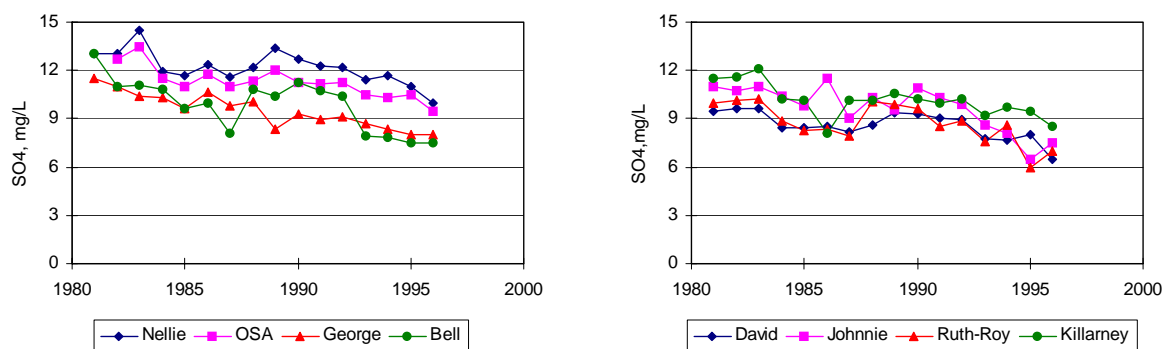


Figure 4. Time trends for sulfate concentrations in eight Killarney lakes. Data from the ARG long term monitoring program (W. Keller, unpubl. data).

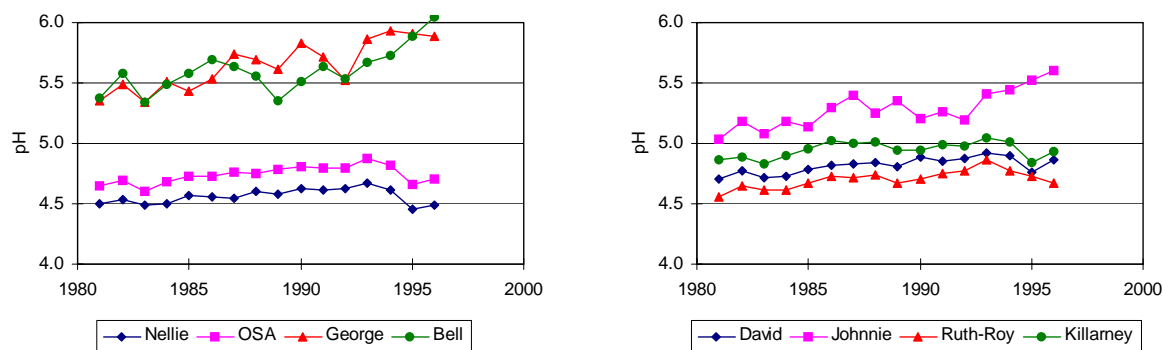


Figure 5. Time trends for pH in eight Killarney lakes. Data from the ARG long term monitoring program (W. Keller, unpubl. data).

4.2 Critical load and their exceedances

The calculated critical load for a lake is considered as an inherent property of the lake and its catchment. Very high values indicate acceptable water quality for sensitive organisms regardless of deposition scenarios, whereas low values indicate sensitivity to acidification. Figure 6 shows the distribution of critical loads of sub-catchments in Killarney, and Figure 7 to Figure 14 show the critical load exceedances of the different deposition scenarios. Almost 50 % of the Killarney area belongs to a high critical load class, whereas only a small part of the area (2 %) has a critical load less than $25 \text{ meq m}^{-2} \text{ yr}^{-1}$ (Figure 15). Figure 16 shows that there is a reasonable relationship between pH in the lakes and their calculated critical load exceedances. There is also a clear distinction between low-pH lakes in the quartzite-region (exceeded) and higher-pH lakes of the central-west and the park-edge area in north and south-east (not exceeded).

The critical load methods for surface waters are based on ANC as the critical chemical criteria and brown trout (*Salmo trutta*) as the biological indicator, see Appendix A. This introduces some uncertainties for the Killarney-calculations because lake trout (*Salvelinus namaycush*) is the target organism of Killarney, and no regional datasets link ANC to the status of lake trout populations. However, a pH of about 5.5 is considered a critical limit for lake trout (Baker et al. 1988; Gunn and Keller 1990; Conlon et al. 1992) and pH 6.0 for zooplankton (Keller et al. 1990; Havens et al. 1993). Non-exceedance of the critical load corresponded to a pH close to 5.5 for the whole dataset (Figure 16). However, if the higher-DOC lakes ($\text{DOC} > 4 \text{ mg L}^{-1}$) are removed, non-exceedance corresponds to a pH closer to 6.0. Time-lags in the response to deposition reductions may occur, but using the 1989 deposition-scenario has an effect on these pH-values of only +0.1 pH unit.

ANC is defined here as the difference between base cations and strong acid anions. According to the electrical neutrality requirement ANC also expresses the concentrations of H^+ , organic anions, inorganic Al-ions and bicarbonate, and is thus related to the toxic components. The reason why ANC is preferred rather than pH is the fact that toxicity to acidified waters is strongly related to the presence of inorganic aluminum, not only pH. In colored waters that are not anthropogenically acidified pH may be low (5.0-5.5) and no toxic Al may be present. Acidified clear-waters of the same pH may be highly toxic due to the presence of inorganic Al. Acidified colored lakes with elevated concentrations of Al may not be toxic due to the complexing ability of the humic substances. Even clearwater lakes with very low base cation concentrations may have $\text{pH} < 5.5\text{-}6.0$ at unpolluted conditions.

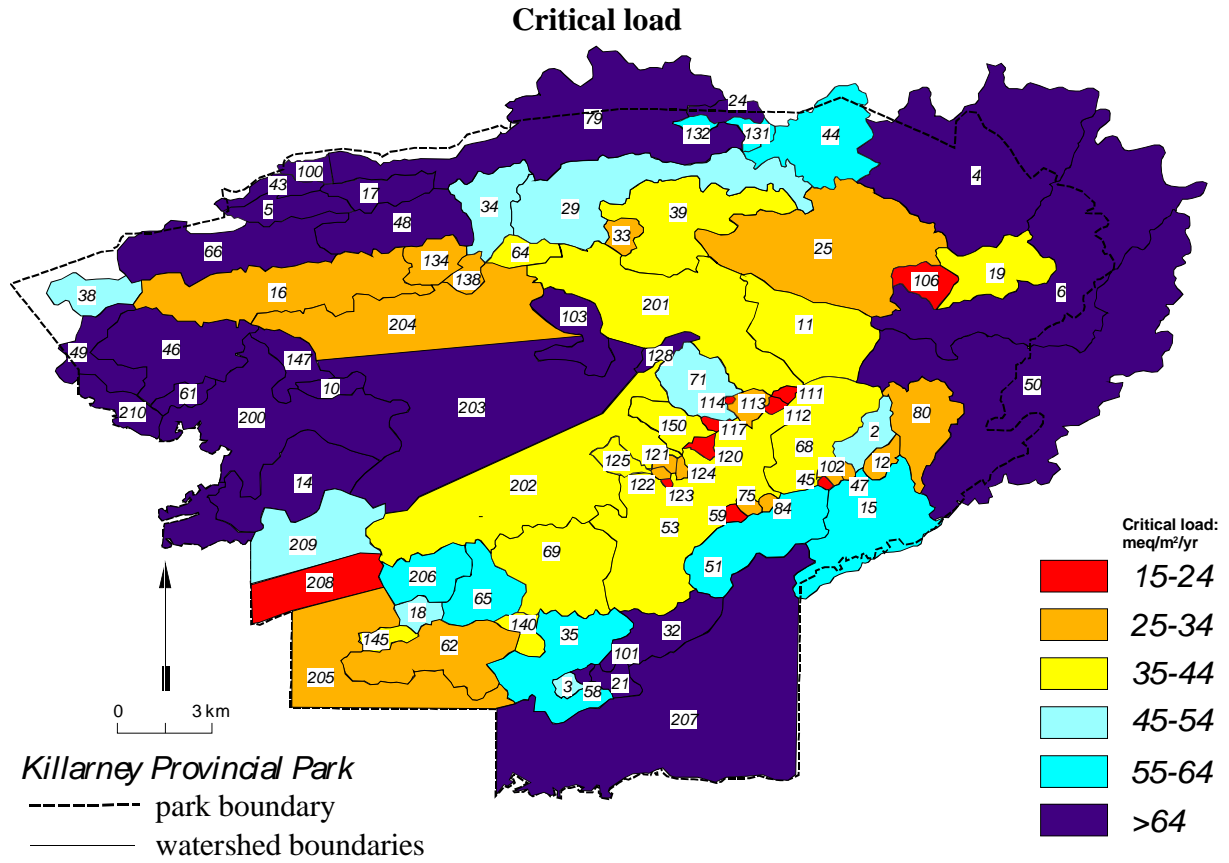


Figure 6. Critical load of sub-catchments of the Killarney Provincial Park.

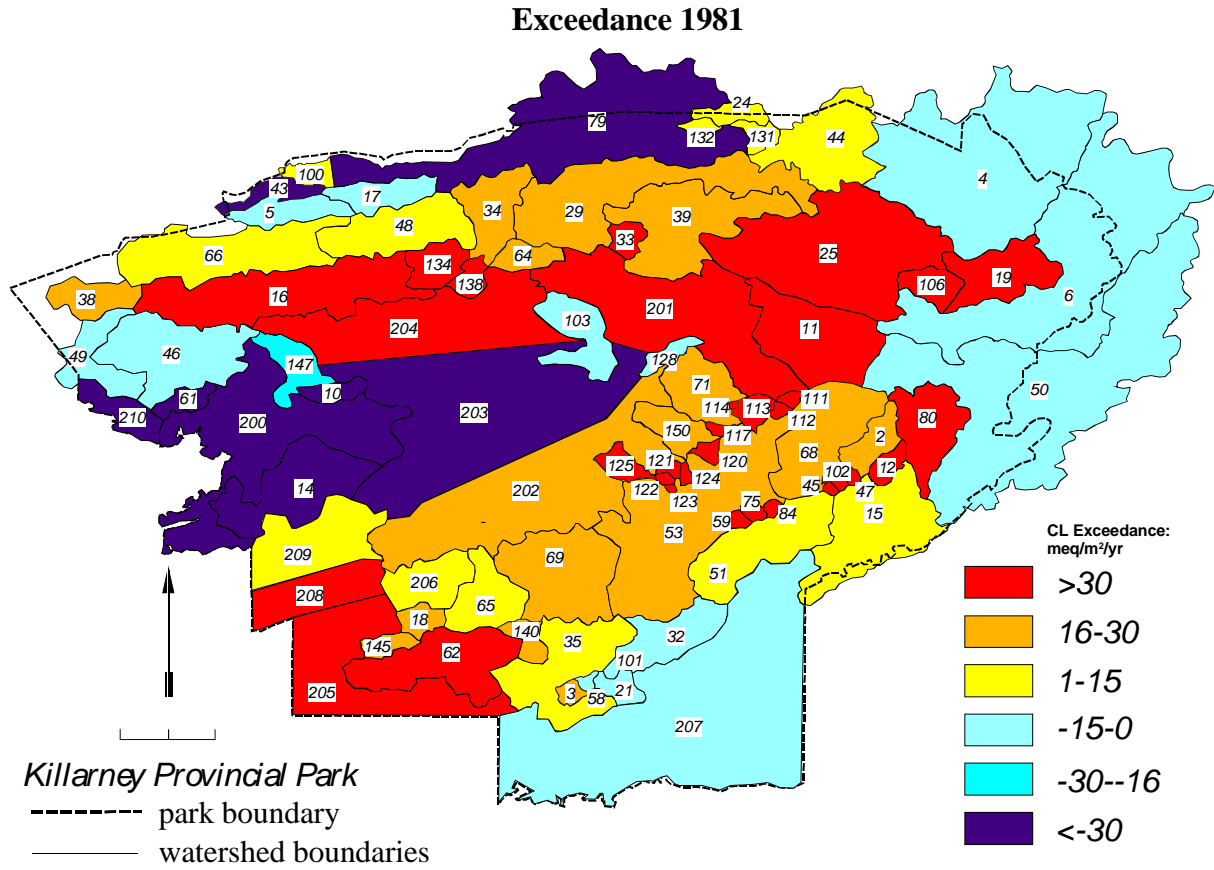


Figure 7. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 1981.

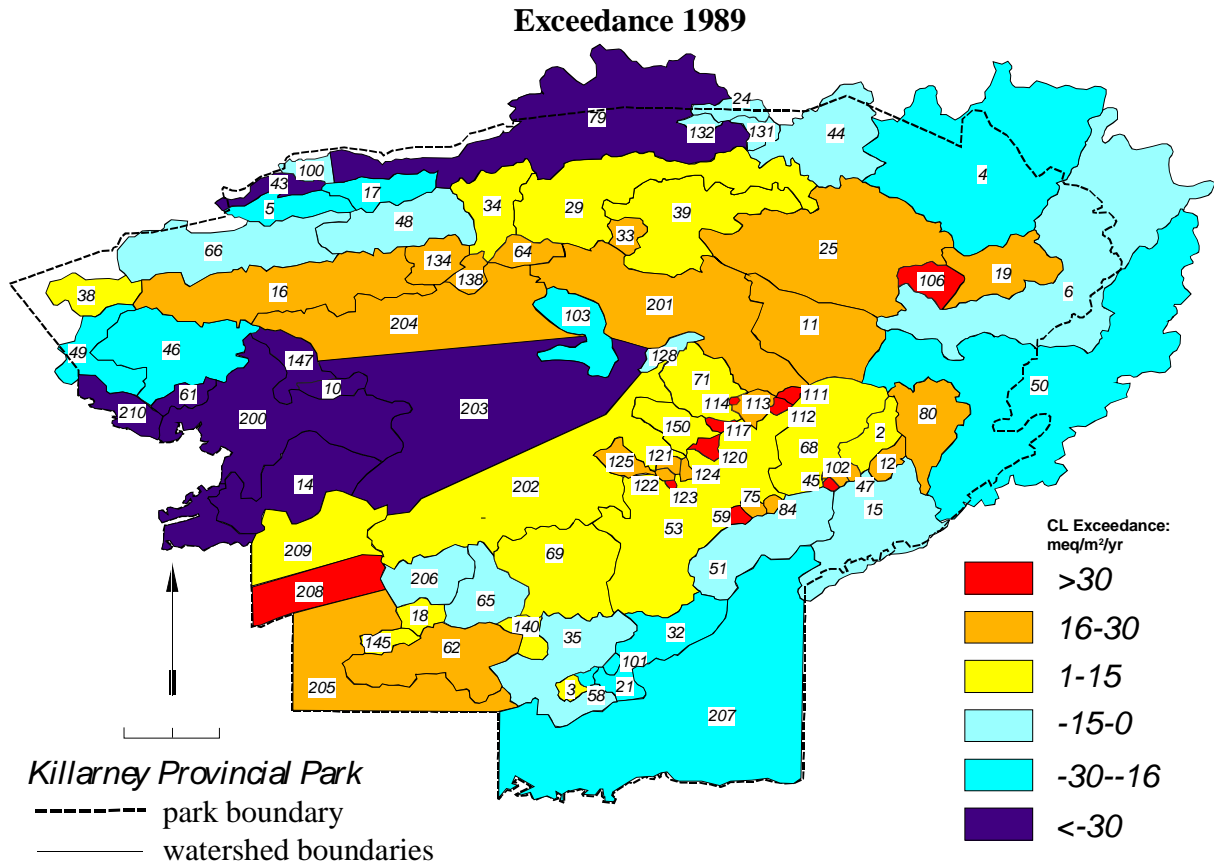


Figure 8. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 1989.

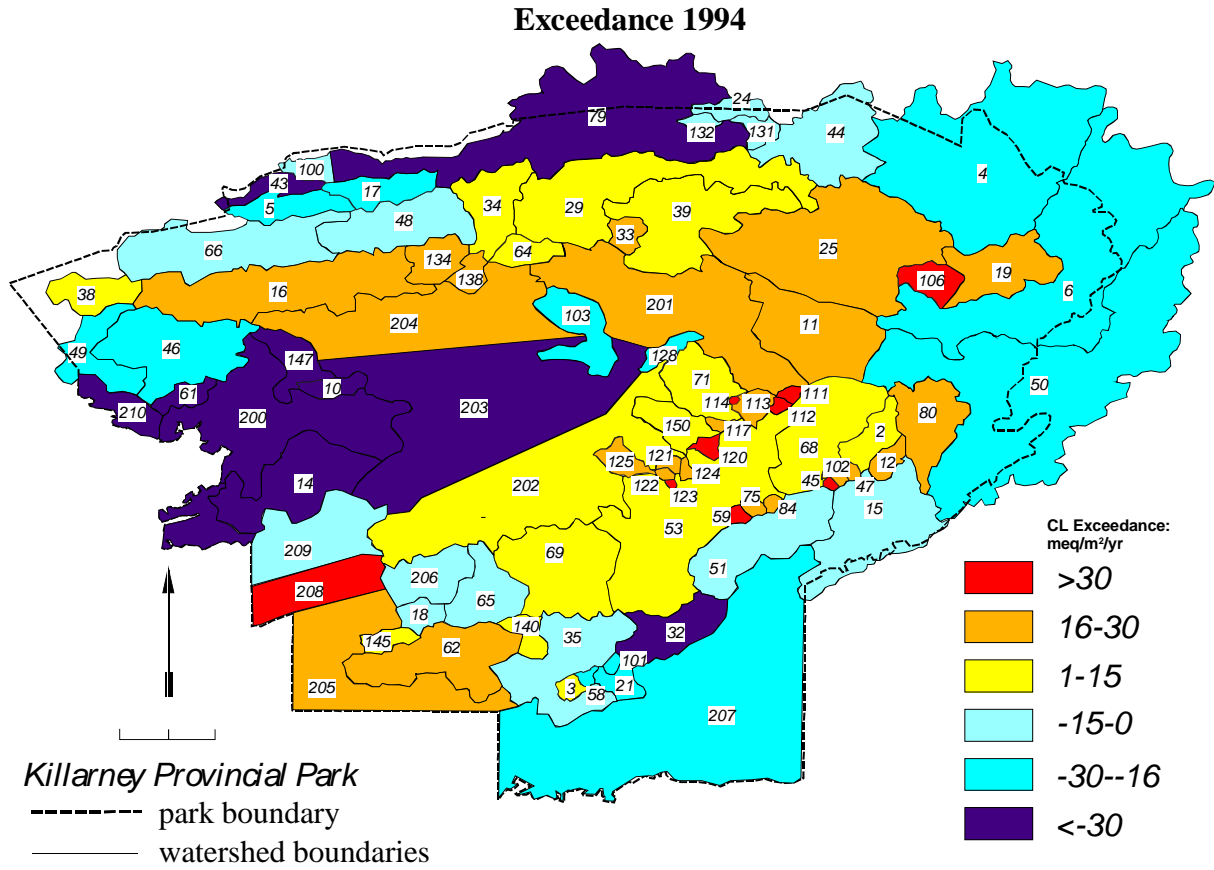


Figure 9. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 1994.

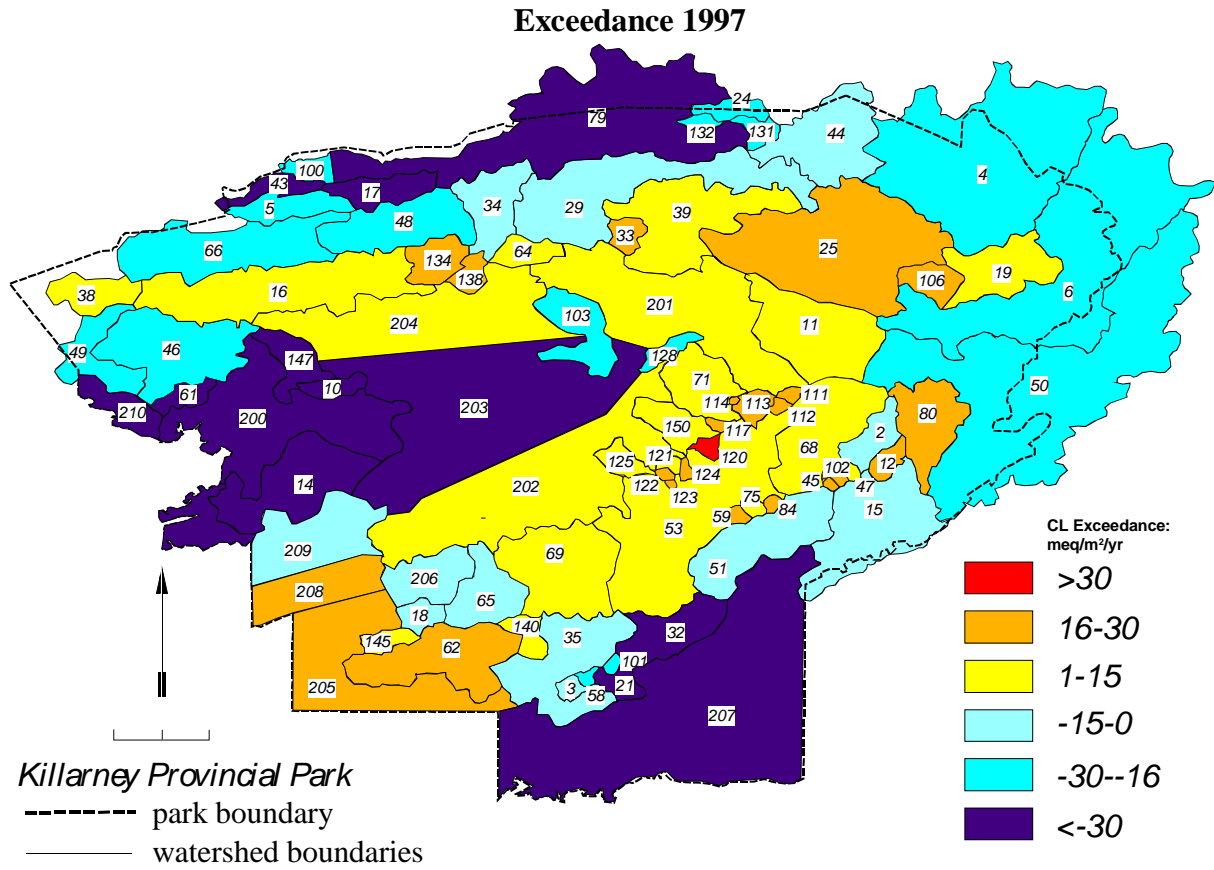


Figure 10. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 1997.

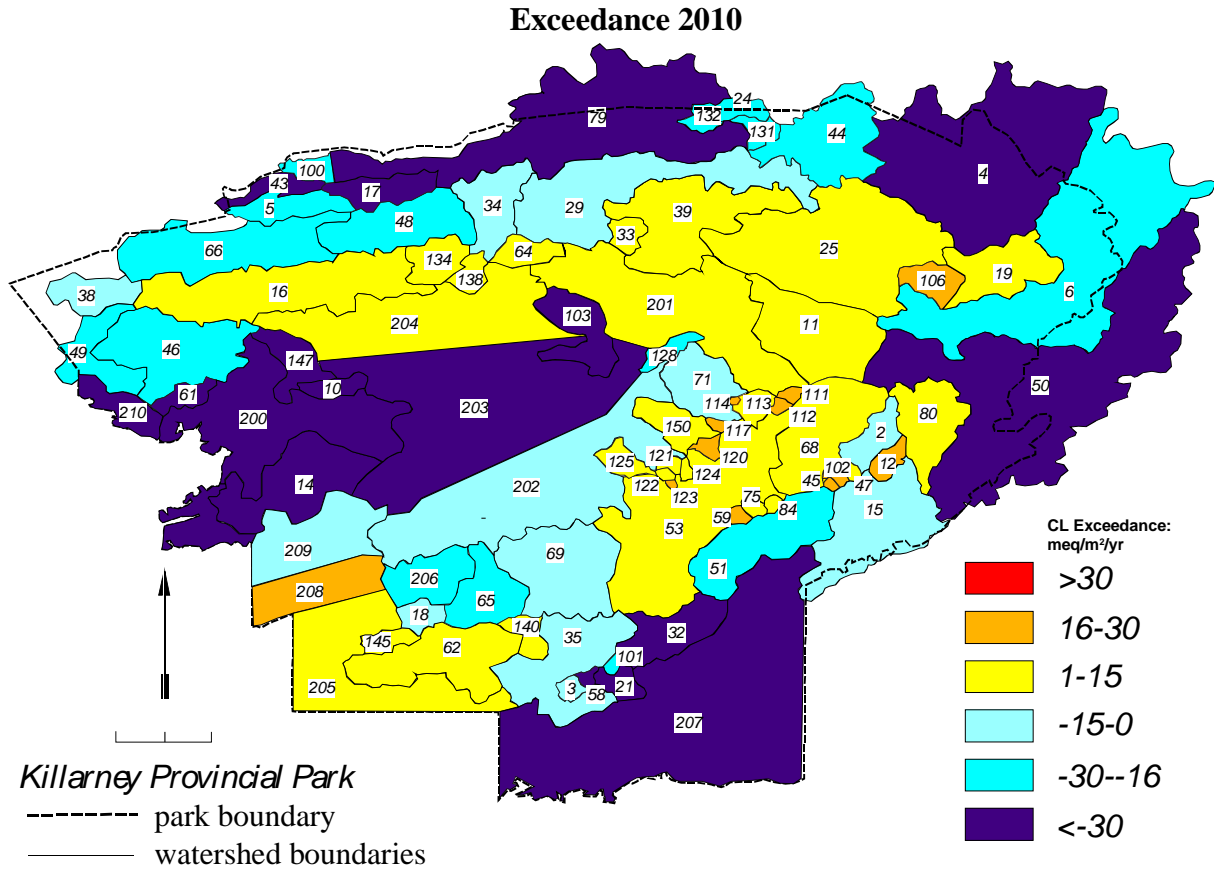


Figure 11. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 2010.

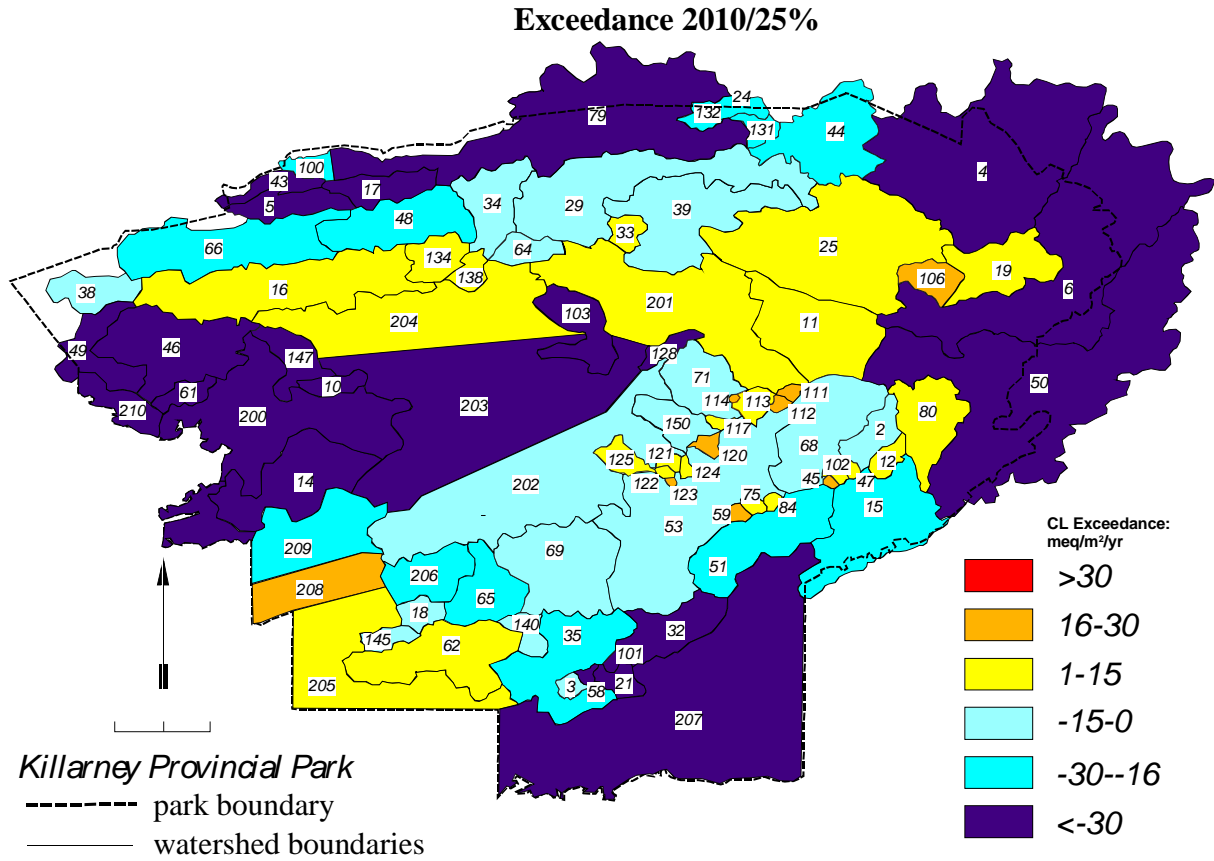


Figure 12. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 2010 with an additional 25 % emission reduction.

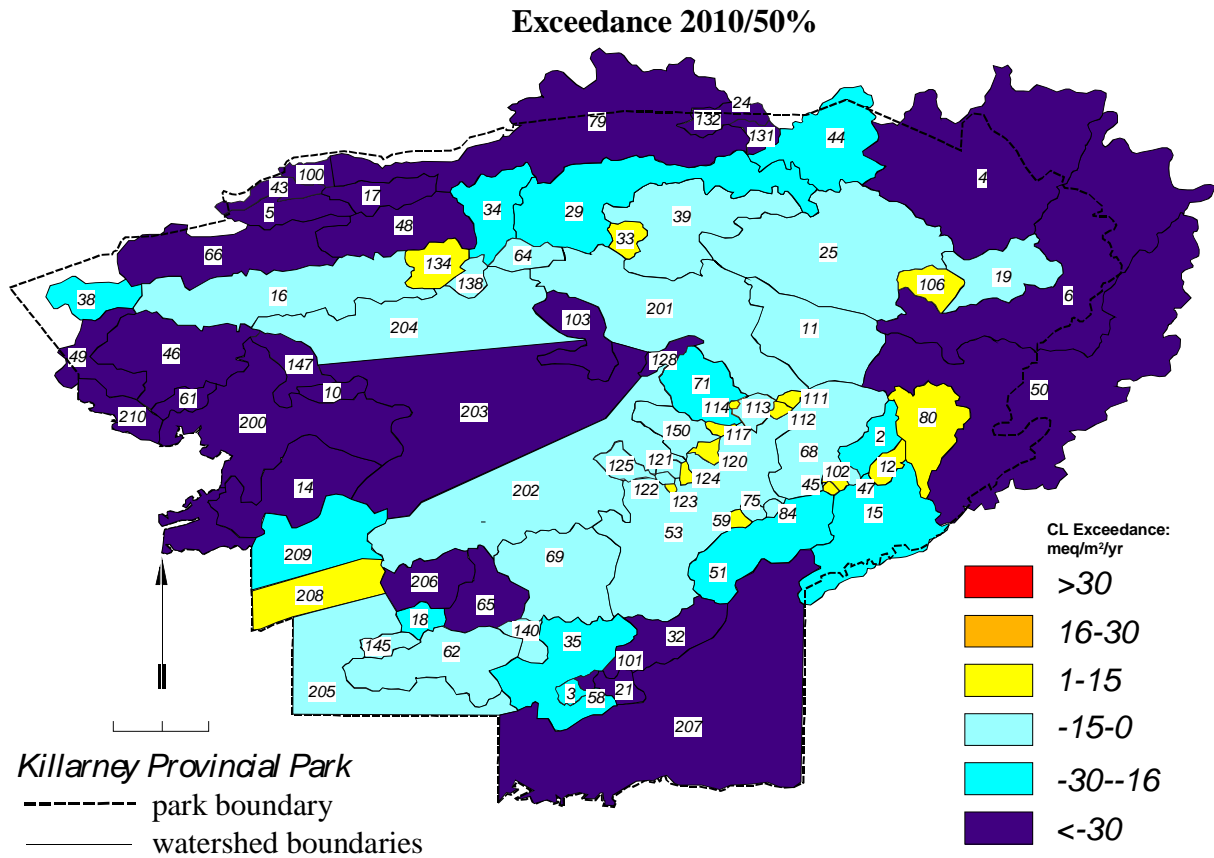


Figure 13. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 2010 with an additional 50 % emission reduction.

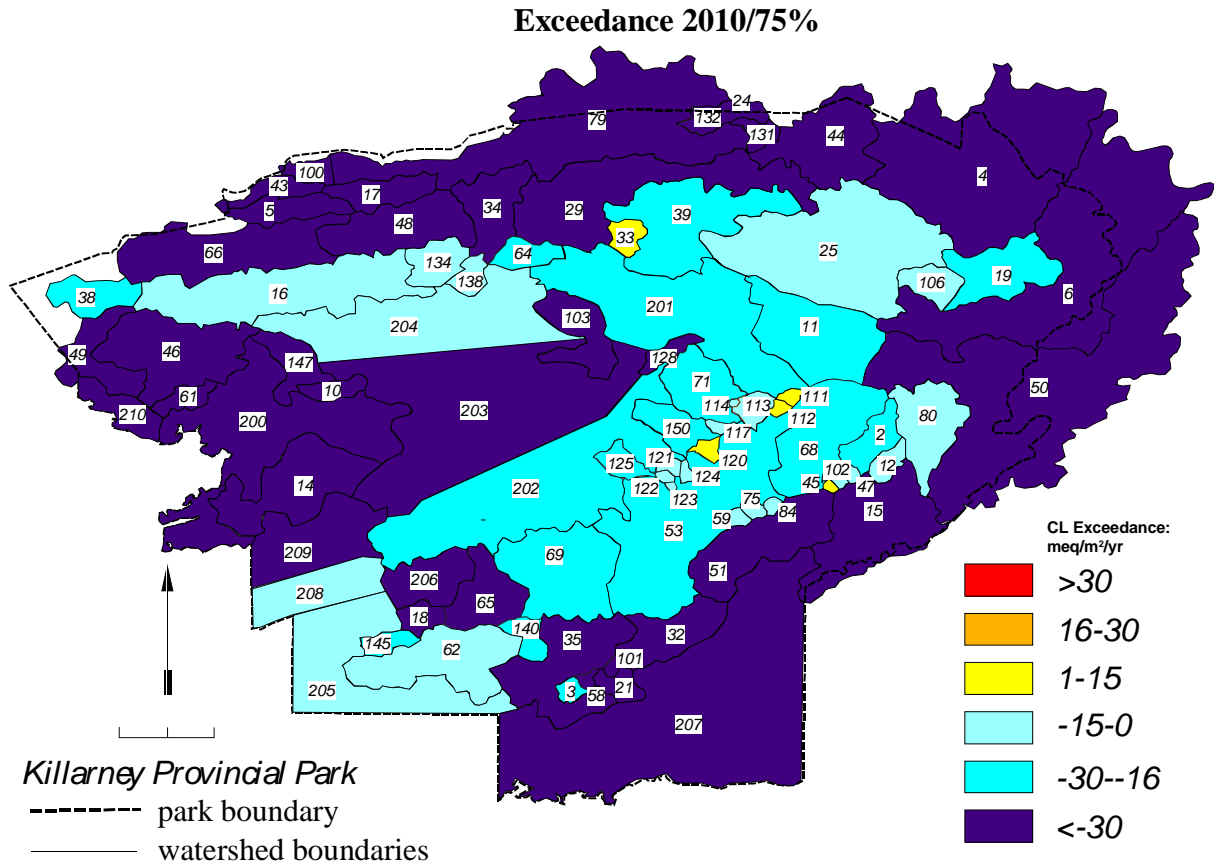


Figure 14. Critical load exceedance of sub-catchments of the Killarney Provincial Park in 2010 with an additional 75 % emission reduction.

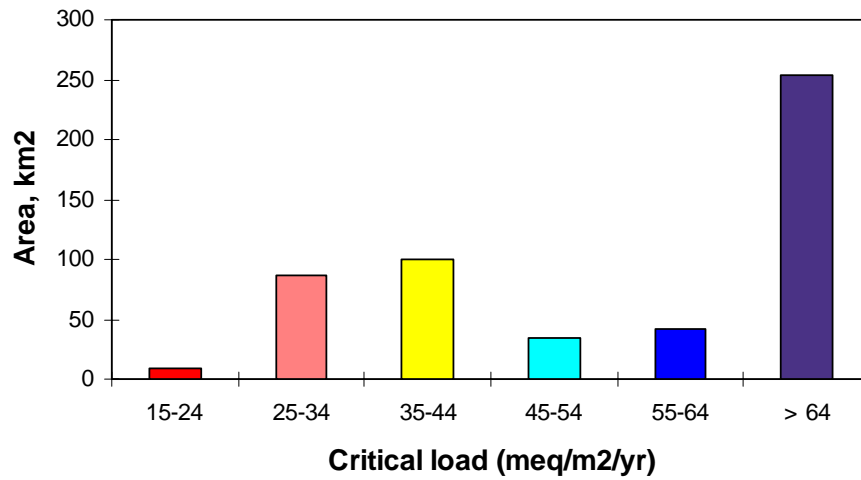


Figure 15. The Killarney Park area divided into different critical load classes. Almost 50 % of the area is in the highest critical load class ($> 64 \text{ meq m}^{-2} \text{ yr}^{-1}$), which is supposed to secure acceptable water quality for sensitive aquatic species at any realistic deposition scenarios, see later sections.

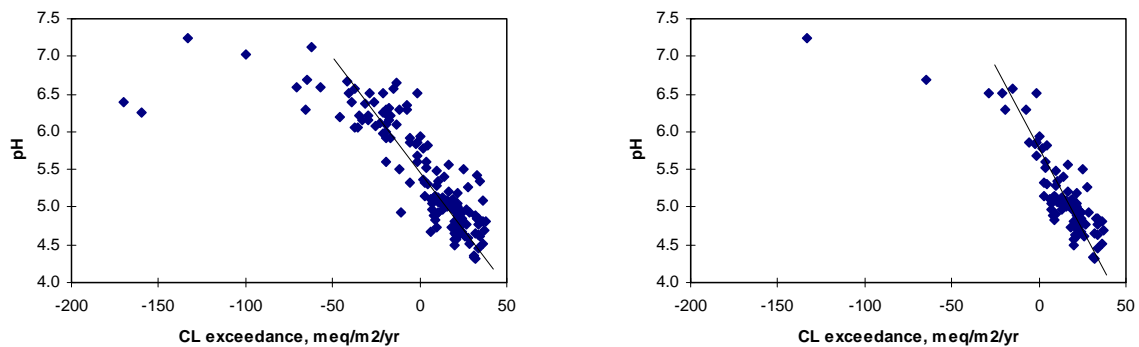


Figure 16. Relationship between critical load exceedance (1994-scenario) and pH (1996-survey) in 151 Killarney lakes (left panel). Lakes with DOC-concentrations above 4 mg L^{-1} have been removed from the dataset of the right panel. Lines are drawn by hand through data with CL-exceedance $> 50 \text{ meq m}^{-2} \text{ yr}^{-1}$.

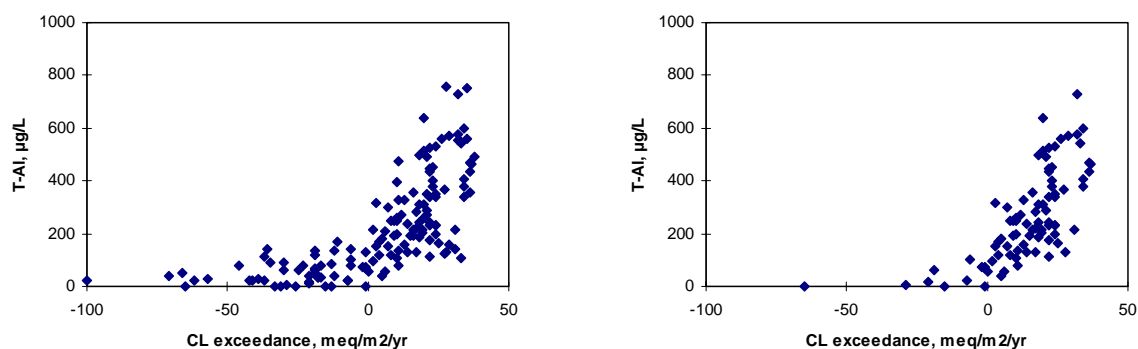


Figure 17. Relationship between critical load exceedance (1994-scenario) and total aluminum (T-Al; 1996-survey) in 151 Killarney lakes (left panel). Lakes with DOC-concentrations above 4 mg/L have been removed from the dataset of the right panel. Three data-points ($<-100 \text{ meq m}^{-2} \text{ yr}^{-1}$ are omitted).

Low critical loads combined with high strong acid deposition will normally lead to mobilization of aluminum in the soils, as seen in Figure 17. The relatively high concentrations of total aluminum may be related to the high acid deposition and to the relatively low run-off. High values may also be associated with high concentrations of dissolved organic matter. In lakes with $\text{pH} > 6.0$, higher T-Al than $50 \mu\text{g L}^{-1}$ is almost entirely found in lakes with $\text{DOC} > 4 \text{ mg L}^{-1}$.

As pointed out above the exceedance of critical loads are calculated by subtracting the critical load from the deposition, thus giving positive values for exceedance. The S-deposition to the Killarney Park has decreased significantly during the last 15 years Figure 18 (left panel). According to the control programs of Eastern Canada and the Clean Air Act of the U.S., further reductions are expected. Four scenarios for 2010 are also given in Figure 18 (left panel). From 1981 to 1997 the area of the Killarney Provincial Park where the critical loads are exceeded was reduced from almost 60% to nearly 40%. In the year 2010 the exceeded area will be reduced to about 30% at steady state (Figure 18, right panel). If S-emissions are reduced by additionally 25%, the exceeded area will be further reduced to about 20%, whereas 50% and 75% additional reductions will secure almost complete non-exceedance situations in the Park. The two diagrams in Figure 18 indicate that a reduction in S-deposition to about $30 \text{ meq m}^{-2} \text{ yr}^{-1}$ can be considered as a target load for the area if complete recovery is desired.

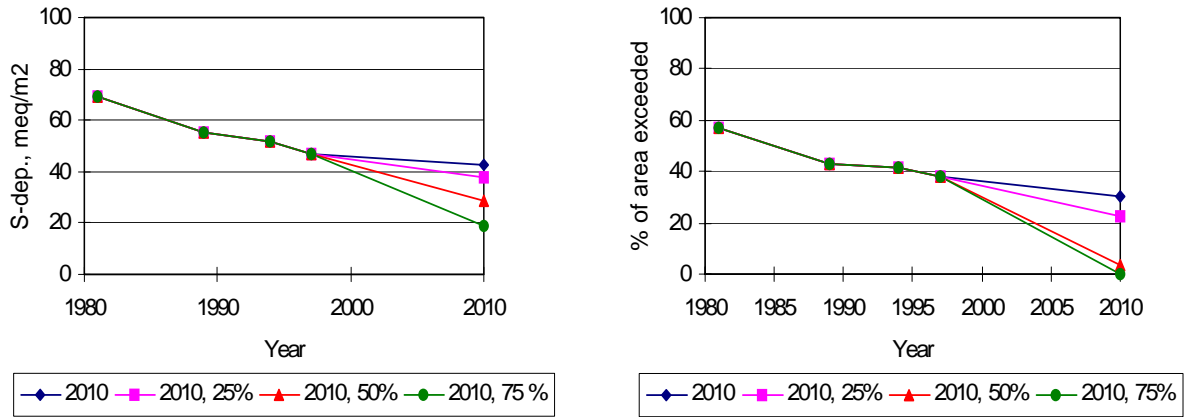


Figure 18. S-deposition scenarios (see also Table 1; left panel), and percent of the park area where critical loads are exceeded at the given S-deposition scenarios (right panel).

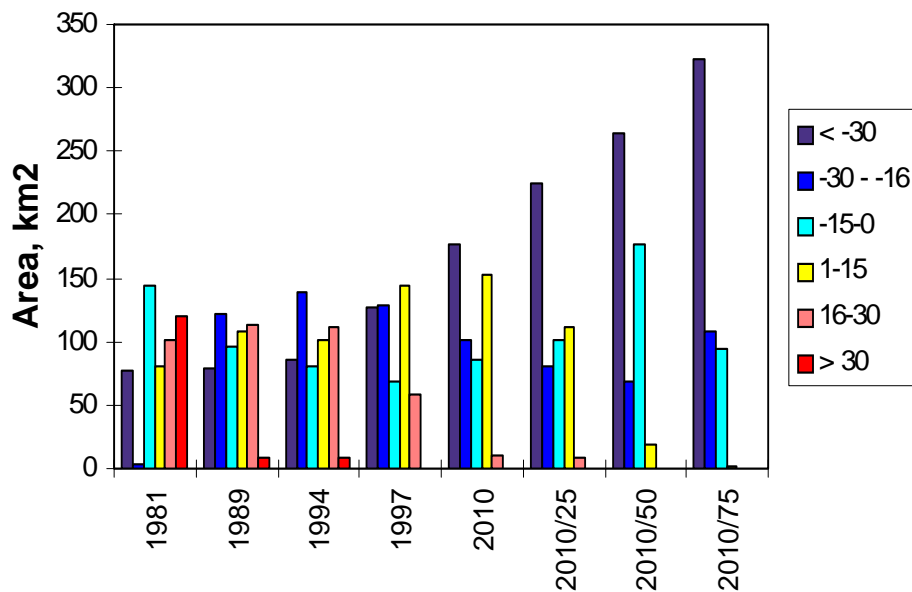


Figure 19. Critical load exceedances in the Killarney park area distributed according to year and exceedance classes. Exceedance unit: meq m⁻² yr⁻¹. Negative values reflect non-exceedance. The 2010-scenarios with an additional 50 % or 75 % reduction in S-emission secure almost complete non-exceedance situations.

Exceedances may vary from high to low. At high exceedances most of the acid-sensitive aquatic organisms are expected to be heavily affected. At decreasing exceedances the degree of damage is also

assumed to decrease. It may therefore be of interest to consider the distribution of the park area according to the degree of exceedance. Figure 19 shows that a broad specter of exceedance classes was represented in 1981. In 1997 exceedances larger than $20 \text{ meq m}^{-2} \text{ yr}^{-1}$ were almost absent, and the situation may be slightly improved in 2010. It is worth while to note that, according to these calculations, a target load of $40 \text{ meq m}^{-2} \text{ yr}^{-1}$ (a combination of the 2010 and the 2010 + 25 % reduction scenario) will almost completely eliminate exceedance classes larger than $15 \text{ meq m}^{-2} \text{ yr}^{-1}$.

According to Moran (1997) critical loads in eastern Canada have been estimated to vary from $8 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ($17 \text{ meq m}^{-2} \text{ yr}^{-1}$) to $20 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ($42 \text{ meq m}^{-2} \text{ yr}^{-1}$) of non-sea-salt SO_4 wet deposition (which turned out to be a surrogate for total deposition). These figures are from the Canadian 1990 Assessment and were determined relative to a pH damage threshold of 6.0. The model did not take NO_3 leaching into account. Our figure of $30 \text{ meq m}^{-2} \text{ yr}^{-1}$ as a target load for total strong acid deposition corresponds to $14.4 \text{ kg SO}_4 \text{ ha}^{-1} \text{ yr}^{-1}$, which is in the middle of the eastern Canada critical load range. Our figure secures protection of almost all the park area from critical load exceedances. If the target load is increased to $40 \text{ meq m}^{-2} \text{ yr}^{-1}$, large exceedances will be avoided, but 25 % of the park area will have exceedances in the range $1\text{-}15 \text{ meq m}^{-2} \text{ yr}^{-1}$ and an additional 10 km^2 will have exceedances above $15 \text{ meq m}^{-2} \text{ yr}^{-1}$.

5. Concluding remarks

The results presented in this report show that use of the critical load approach in the Killarney Provincial Park is fruitful. This is partly due to the variety of lake characteristics in this relatively small area. The site is therefore of great value from a modelling point of view.

Although uncertainties are pointed out concerning the coupling between target species of the park and critical water quality (ANC or pH), critical load exceedances divide the lakes in two at a pH of 5.5 or 6.0 depending on whether the colored lakes are part of or not part of the lake population. For lakes of DOC <math>< 4 \text{ mg L}^{-1}</math> the result correspond well with the target water quality (pH 6.0) that has been agreed upon in Ontario.

The calculations also show that considerable improvements in water quality have occurred and that further improvements may be expected if the commitments of the Clean Air Act is fulfilled. Scenarios for 2010 and for 2010 with further emission reductions show that the sulphur deposition may reach the critical load in the future. If the target load, the politically acceptable load, is set to $30 \text{ meq m}^{-2} \text{ yr}^{-1}$, one might expect that the water quality will improve more or less completely.

To obtain a more complete picture of how the deposition scenarios will impact the species and the biodiversity of the park, two additional approaches may be taken.

1. To study the possibility of developing dose-response relationships for different biological species analysed in the Killarney lakes, using a similar approach as used in Norway for brown trout (*Salmo trutta*) and other fish and invertebrate species (Lien et al. 1992). These relations could then be used for modifying critical load formulations specifically adapted to the conditions and the species occurring in the Killarney lakes. Thus, the critical load data would thereby be more directly related to biological species found in the park. This will, in turn, be a valuable tool for managing the park.

2. Calculation of critical loads of both acidifying N and S deposition and their potential exceedances. This may be done if specific data for nitrogen immobilisation and uptake are available. Whereas the Steady-State Water Chemistry (SSWC) model allows the calculation of critical loads of acidity (S+N) and their present exceedances, the First-order Acidity Balance (FAB) model (see appendix) is suitable for the extended work. The FAB-model is based on the steady-state mass balance approach, widely used in many models for computing critical loads for forest soils (see UN/ECE 1996, Posch et al. 1997a,b).

Nitrogen was not included in the calculations given in this report because the nitrate lake-concentrations seemed relatively low and would not contribute to a large change in exceedances. However, since the FAB-model seems to be applied as the basis for the second N-protocol, an assessment of the potential effects of N-deposition should also be carried out for the Killarney Provincial Park. This may also be justified by the possibility of future climatic changes that might interact with nitrogen processes of the soil system. Increased mineralisation due to elevated temperatures may result in increased N runoff from the terrestrial compartments.

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Appendix A. Critical Load methods

The critical load concept

The concept of critical loads has been widely accepted as a basis for designing control strategies to reduce regional air pollution. In order to gain more insight in the magnitude and spatial variation of critical loads, the UN/ECE Executive Body of the Convention on Long-range Transboundary Air Pollution (LRTAP) has set up a Task Force on Mapping Critical Levels/Loads under the Working Group on Effects. Critical loads data from individual countries are collected, mapped and reported by the Coordination Center for Effects (CCE), located at the National Institute of Public Health and the Environment (RIVM) in Bilthoven, the Netherlands. The results are reported biannually (see Posch et al. 1997a for the latest report).

The calculation of critical loads for various receptors (surface water, forest soils) is an approach which seeks to link emissions abatement strategies to the capacity of ecosystems to withstand and buffer the effects of acid deposition. A critical load is defined as (Nilsson and Grennfelt 1988):

“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”.

Until 1994 the international work focused on the development of methodologies and the synthesis of national data to produce European maps of critical sulphur deposition used in the negotiations of the Second Sulphur Protocol, signed in Oslo in June 1994 (UN/ECE 1994). In order to prepare the scientific support for the negotiations of a revised protocol on the reduction of nitrogen emissions, a new methodology was developed to take into account multiple effects (i.e. eutrophication and acidification) of multiple pollutants (sulphur and nitrogen). The critical loads of S and N are interrelated and therefore a single critical load for one pollutant cannot be defined without making assumptions about the other.

Calculating Critical Loads for Surface Waters

Two steady state models – one empirical and one process-oriented – for calculating critical loads of acidifying deposition (both S and N) for surface waters are in current use and these are summarized below: The (modified) Steady-State Water Chemistry (SSWC) model allows the calculation of critical loads of acidity (S+N) and their present exceedances. The First-order Acidity Balance (FAB) model allows the simultaneous calculation of critical loads of acidifying N and S deposition and their exceedances. The FAB-model is based on the steady-state mass balance approach, widely used in many models for computing critical loads for forest soils (see UN/ECE 1996, Posch et al. 1997a). Here only the model formulations are presented, and for a derivation of the models we refer to the cited literature.

The modified SSWC-model

The critical load of acidity is calculated as (see, e.g., Henriksen et al. 1990):

$$(1) \quad CL(Ac) = Q ([BC^*]_0 - ANC_{limit})$$

where Q is the runoff, $[BC^*]_0$ is the original base cation concentration, and ANC_{limit} is the chosen critical ANC threshold.; the star refers to sea salt corrected quantities. $[BC^*]_0$ is estimated from the present leaching of base cations and the long-term changes in the inputs of strong acid anions using the so-called F-factor (Henriksen 1984, Brakke et al. 1990):

$$(2) \quad [BC^*]_t - [BC^*]_0 = F ([SO_4^*]_t + [NO_3]_t - [SO_4^*]_0 - [NO_3]_0)$$

where the subscripts 0 and t refer to the original (background) and present concentrations, resp. $[\text{SO}_4^*]_0$ is estimated from a linear regression with $[\text{BC}^*]_t$ using data from Norwegian background lakes, whereas $[\text{NO}_3]_0=0$. The F-factor is calculated following Brakke et al. (1990), but have recently been modified to account for catchments with high and low runoff:

$$(3) \quad F = \sin((\pi/2) Q [\text{BC}^*]_t / S) \quad \text{for} \quad Q [\text{BC}^*]_t < S$$

where S is the annual base cation flux above which $F=1$. The critical ANC limit is calculated by a model suggested by Henriksen et al. (1995) and results in values between 0 and $50 \mu\text{eq l}^{-1}$ depending on the catchment characteristics. The formula used for Norway is:

$$(4) \quad \text{ANC}_{\text{limit}} = 0.25 Q [\text{BC}^*]_0 / (1 + 0.25 Q)$$

where Q is runoff in m yr^{-1} .

The SSWC-model has been modified to include both S and N acidity by considering the present (measured) N-leaching (N_{leach}) in the calculation of the present exceedance of the critical load (Kämäri et al. 1992):

$$(5) \quad \text{Present Ex(Ac)} = S_{\text{dep}} + N_{\text{leach}} - \text{CL(Ac)}$$

where S_{dep} is the present deposition of sulphur. The N leaching term describes the balance between N deposition and the N processes in the catchment such as uptake, immobilization, denitrification and in-lake retention of nitrogen.

The FAB-model

When considering the effects of both sulphur and nitrogen simultaneously, one cannot, as pointed out above, expect to obtain unique critical loads of S and N, since a reduction in the deposition of sulphur might allow a higher deposition of acidifying nitrogen compounds without causing 'harmful effects'. From an acidity balance one can derive the following equation, describing the trade-off between sulphur and nitrogen critical loads (Posch et al. 1997b):

$$(6) \quad a_N \text{CL(N)} + a_S \text{CL(S)} = b_1 N_{\text{upt}} + b_2 N_{\text{imm}} + \text{BC}_{\text{le,crit}}$$

where N_{upt} and N_{imm} are the net growth uptake (harvested N) and immobilization of N, resp., and a_N , a_S , b_1 and b_2 are dimensionless constants depending on lake and catchment properties alone:

$$(7a) \quad a_N = (1 - f_{\text{de}}(1-r))(1 - \rho_N)$$

$$(7b) \quad a_S = 1 - \rho_S$$

$$(7c) \quad b_1 = f(1 - f_{\text{de}})(1 - \rho_N)$$

$$(7d) \quad b_2 = (1-r)(1 - f_{\text{de}})(1 - \rho_N)$$

where f is the fraction of forest area within the catchment and r is the lake:catchment area ratio. In deriving eq.5 not only the uptake and immobilization of N have been taken into account, but also denitrification and the in-lake retention of N and S, all three as linear functions of the net input of N (resp. S) with proportionality coefficients f_{de} , ρ_N and ρ_S , leading to the coefficients above. The in-lake retention coefficient ρ_N is modeled by a kinetic equation (Kelly et al. 1987, Dillon and Molot 1990):

$$(8) \quad \rho_N = s_N / (Q/r + s_N)$$

where s_N is the net mass transfer coefficient for N (m/yr). An analogous equation holds for ρ_S (Baker and Brezonik 1988). Finally, the critical base cation leaching from the catchment is computed from water quality data by the steady-state model introduced above (eq.1):

$$(9) \quad \text{BC}_{\text{le,crit}} = Q ([\text{BC}^*]_0 - [\text{ANC}]_{\text{limit}})$$

In addition to eq.5 the critical load of S is limited by the following constraint:

$$(10) \quad CL(S) \leq CL_{\max}(S) = BC_{le,crit} / a_S$$

Below a value of

$$(11) \quad CL_{\min}(N) = (b_1 N_{upt} + b_2 N_{imm}) / a_N$$

all N deposition is taken up, retained or immobilized in the catchment. On the other hand, the highest critical load of N (in the absence of S deposition) is given by

$$(12) \quad CL_{\max}(N) = CL_{\min}(N) + BC_{le,crit} / a_N$$

Eq.6, together with these constraints, determines the so-called critical load function (Figure 1), separating the N- and S-deposition values which cause 'harmful effects' (exceedance) from those which do not (non-exceedance).

As mentioned above, unique critical loads for S and N cannot be specified; however, this may be an advantage, since it allows to determine (cost-)optimal deposition reductions. If the deposition of one of the compounds is fixed (prescribed), a unique critical load for the other can be computed from eq.5.

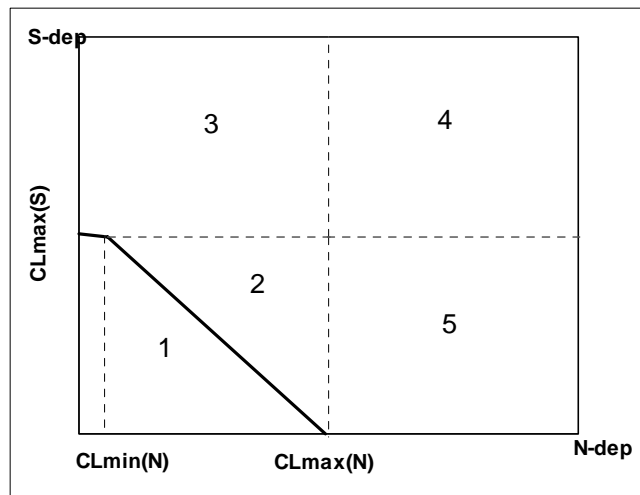


Figure 1. The critical load function. Each lake has its own diagram. The shaded area (1) indicates those N and S deposition values not causing exceedance. The critical load function (thick line) is given by the calculated values for $CL_{\max}(S)$, $CL_{\min}(N)$ and $CL_{\max}(N)$. The location of the lake's N and S deposition in the diagram gives the reduction requirements for the lake to give non-exceedance: 1. no reductions required, 2. N or S reduction, 3. mandatory S-reduction, 4. mandatory S and N reduction, 5. mandatory N-reduction.

The FAB-model gives us the reduction requirements to reach non-exceedance for a given deposition scenario and can as mentioned be used to achieve optimal reduction conditions. In order to estimate the total critical load exceedance (both from N and S) the following equation can be used.

$$(13) \quad \text{Pot Ex} = -CLAC + S_{dep} + (N_{dep} - CL_{\min}(N))$$

Input data

SSWC-model:

Data necessary for the calculation of critical loads of acidity for lakes are:

Variable	Value
Base cations: Ca, Mg, Na, K	Yearly weighted average values, or estimates thereof
Anions: SO ₄ , NO ₃ , Cl	Yearly weighted average values, or estimates thereof
Runoff	Yearly mean runoff

The FAB-model:

Requires in addition to the data for the SSWC-model the following data:

Variable	Value
Lake area	Measured from maps
Catchment area	Measured from maps
Forest fraction in catchment	Measured from maps
Soil fraction in catchment	Measured from maps
N _u : nitrogen uptake in catchment	Dependent on tree species and harvesting practice For non-productive forests and non-forested areas N _u = 0
N _{imm} : N immobilization	2-5 kg/ha/yr (=142-357 eq/ha/yr)
f _{de} : denitrification fraction	=0,1+0,7•f _{peat}
S _N : mass transfer coefficient for N	2-8 m/yr (Dillon and Molot 1990)
S _S : mass transfer coefficient for S	0,2-0,8 m/yr (Baker and Brezonik 1988)
r. lake/catchment ratio	0-1

In this report only the Steady state Water Chemistry model has been used.

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