A review of anthropogenic sources of nitrogen and their effects on Canadian aquatic ecosystems

DAVID W. SCHINDLER^{1,*}, PETER J. DILLON² and HANS SCHREIER³

¹Department of Biological Sciences, University of Alberta, Edmonton, AB, T6G 2E9, Canada; ²Environmental and Resource Studies, Trent University, 1600 West Bank Drive, Peterborough, ON, K9J 7B8, Canada; ³Institute for Resources, Environment and Sustainability, University of British Columbia, 2202 Main Mall, Vancouver, BC, V6T 1Z4, Canada; *Author for correspondence (e-mail: d.schindler@ualberta.ca; phone: +780-492-1291; fax: +780-492-9234)

Key words: Acid rain, Agriculture, Eutrophication, Water pollution

Abstract. Nitrogen releases to air and water are low in most of Canada, but in southern areas with rapid development there are telltale signs of the problems from releases to air and water that are described elsewhere in this volume. These include higher nitrogen in water and releases to the atmosphere from urban areas, industry and agriculture. As a result, in parts of Ontario and Quebec underlain by Precambrian geology, nitrogen deposition is near the critical loads found for geologically similar areas of Europe. In particular, combined inputs of sulphuric and nitric acids are causing base cation depletion in forest soils and keeping some lakes at pH values too low to allow the recovery of biological communities. In southern Ontario, Alberta and British Columbia, rapidly expanding human populations, industry and agriculture are causing high concentrations of nitrate in surface and groundwaters. At present, there is little sign of estuarine eutrophication in Canada, but it appears to be imminent on the Pacific coast, as the result of expanding human populations and intensifying agriculture in the lower Fraser Valley and Puget Sound. Steps should be taken now to prevent the widespread problems caused by nitrogen pollution that have occurred in Europe, the USA, and other populous and industrialized regions.

Introduction

Most areas of Canada still have low human population densities, and are relatively free from industrial activity. As a result, much of the country has escaped the problems associated with nitrogen contamination that have plagued more industrialized countries. These include the contamination of precipitation, surface and ground water, soil saturation with nitrate, acidification of receiving waters, smog formation and pollution of surface and groundwaters to beyond guidelines to protect human health and ecosystem integrity. The problem of managing nitrogen in most of Canada is thus still largely one of prevention, ideally profiting from the mistakes made in more populous and industrialized countries.

Most Canadian freshwaters still contain low concentrations of inorganic nitrogen. In a survey of provincial databases in eastern Canada, Jeffries (1995) found that almost all lakes contained less than 10 μ eq l⁻¹ of NO₃. Provincial mean concentrations for nitrate in lakes were all less than 10 μ eq l⁻¹, except for Prince Edward Island, where four of six lakes examined had over 100 μ eq l⁻¹ and New Brunswick, where about 10 percent of lakes had nitrate concentrations of 10–40 μ eq l⁻¹. Much of the land in these two small Atlantic provinces is used for agriculture, and most of the soil is sandy. Heavily developed parts of southern Ontario, Quebec, southern Alberta and coastal southern British Columbia are other areas with higher than background nitrogen. Lakes and streams show the expected symptoms of overfertilization with nutrients, including nitrogen. In the remainder of this review, we will focus largely on these areas.

Canada-wide trends in airborne emissions of nitrogen

Chambers et al. (2001) estimate that human activities discharge 1.4 million tonnes of nitrogen to the atmosphere in Canada, with ammonia releases from fertilizer and manure being the biggest source. Release of N_2O was evenly split among transportation, industry and agriculture. NO_x emissions from industry and transportation were also important. No NO_x data were available for agriculture, but it was assumed to contribute about as much as industry. More detailed information is given below.

Emissions of NO_x in Canada nearly doubled from 1970–1985. From 1985 to 2000 emissions were relatively constant. A 17% decrease in eastern Canada caused by regulation of emissions from vehicles and smelters was approximately balanced by a 29% increase from electrical generation, vehicles and oil and gas industries in western Canada (Figure 1). Countrywide emissions are projected to decrease 17% by 2020, a balance between a 39% reduction in eastern Canada and a 5% increase in the west, where increased emissions from oil and gas extraction, and coal-fired electrical power are expected.

Ammonia emissions increased by 9% between 1995 and 2000, largely in agricultural areas of the west. They are expected to increase by 50% between 2000 and 2020 largely as the result of the increasing intensity of livestock and poultry production, commercial fertilizer and pesticide manufacturing (Figure 1, Vet et al. 2005).

American emissions of NO_x and ammonia are also of concern, because prevailing winds carry the gases from areas of intensive electricity generation and agriculture in the US Midwest over southeastern Canada. US and eastern Canadian NO_x emissions have declined slightly and a downward trend is expected to continue. In contrast, ammonia emissions increased by 15% between 1990 and 2000. They are expected to continue to increase slightly until 2020 (Vet et al. 2005).

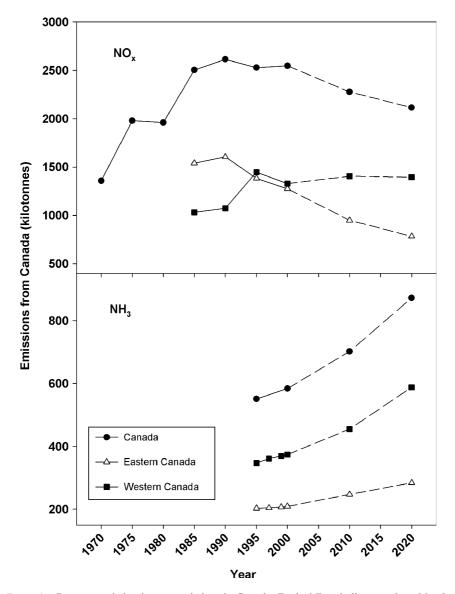


Figure 1. Recent trends in nitrogen emissions in Canada. Dashed lines indicate projected levels. Data from Vet et al. (2005).

The sources of NO_x in Canada are somewhat different from more urbanized and industrialized countries, with off-road vehicles contributing almost as much NO_x as on-road vehicles. In total, vehicles emit almost 50% of NO_x in Canada. The oil and gas industries also contribute disproportionately to Canadian emissions. In contrast, electrical power generation is a smaller percentage than in many countries, as a result of heavy reliance in most of Canada

on hydroelectricity rather than fossil fuel burning. Agriculture contributes most of the ammonia emissions. Livestock culture is the predominant source, followed by manufacturing of commercial fertilizer (Table 1). Both gases contribute to soil acidification, water pollution with nitrate and ammonium, and urban smog.

Atmospheric deposition of nitrogen and its impact on ecosystems

As a land area, Canada is only slightly smaller than all of Europe. Most of the population, industry and agriculture are in a band a few hundred kilometres wide along the US border. Very few studies of deposition have been done north of 60° N. latitude.

Eastern Canada has been studied much more thoroughly than the west, due to concerns about the effects of acidifying deposition on geologically-sensitive terrain. For convenience, we will treat eastern and western Canada separately, dividing the country at the Ontario–Manitoba border.

Eastern Canada

Annual wet nitrate deposition in most of eastern Canada north of 52° N. latitude and east of the Manitoba–Ontario border is <5 kg NO₃ ha⁻¹ y⁻¹ (Figure 2). Only in populous industrial regions of southern Ontario and Quebec does nitrate deposition exceed 10 kg NO₃ ha⁻¹ y⁻¹ over large areas. An even smaller area along the US border with those provinces has nitrate deposition of 20–25 kg NO₃ ha⁻¹ y⁻¹ (Figure 2; Vet et al. 2005). The elevated nitrate concentrations in these regions are the result of NO_x emissions from industrial and automotive sources in southern Canada and the northern USA (Whelpdale and Galloway 1994; Vet et al. 2005). Fortunately, the highest

Table 1. Percentage contribution of major emitting sectors to total nitrogen emissions in Canada. Data from Vet et al. 2005.

Sector	NO_x	NH ₃
Non-ferrous mining and smelting	-	
Electrical power generation	11.4	_
Upstream oil and gas	13	_
On-road vehicles	32.6	3.1
Off-road vehicles	26.9	_
Industrial fuel combustion	=	_
Other fuel combustion	_	_
Agriculture (animals)	_	55
Pesticides and fertilizer	-	34.6
Chemicals and products	_	_

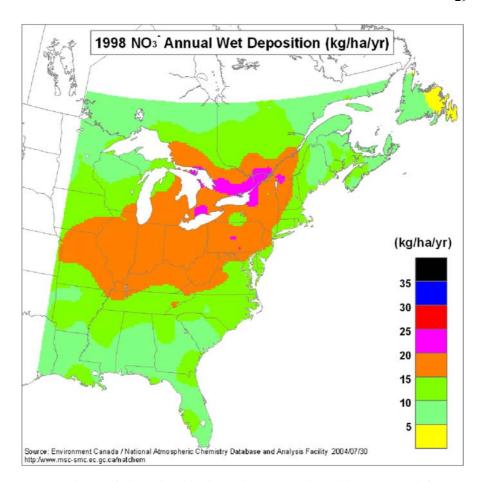


Figure 2. A map of nitrate deposition in southeastern Canada and the eastern USA for 1998. Reprinted with permission from Vet et al. 2005. Note that in this and subsequent figures, deposition values have been left in their original units. For convenience in converting, 1.00 mg l⁻¹ $NO_3 = 0.224$ mg l⁻¹ N = 16 μ eq l⁻¹.

nitrate deposition occurs in areas where soils and water are well buffered. Moreover, deposition has remained relatively constant for some time.

Preliminary wet ammonium deposition maps of eastern Canada show a somewhat different pattern. Here, the highest values observed, of 5–6 kg NH $_3$ ha $^{-1}$ y $^{-1}$ extend through southern parts of far western Ontario, near the Manitoba border (R. Vet, pers. comm.). There is little population, agriculture or industry in the area, which is largely underlain by Precambrian rock with little overburden. The elevated values probably reflect the proximity to agricultural areas of the US Midwest and southern Manitoba. North of 52° N latitude, values return to a near-pristine <2.5 kg NH $_3$ ha $^{-1}$ y $^{-1}$.

Dry deposition is significant at the eight sites where it has been measured, all south of 52° N. It ranges from 17 to 41 percent of total nitrogen deposition. It

is highest near industrial areas along the US border. Combining with wet deposition to obtain total N deposition, values range from 2.6 kg N ha⁻¹ y⁻¹ at the western edge of Ontario to 7.5 kg N ha⁻¹ y⁻¹ at several sites in the East.

Dry deposition values have only been measured for 5 years, so there are no detectable long-term trends. Peroxyacetyl nitrate (PAN) and NO₂ are not included in dry deposition, so total deposition estimates are likely underestimated by as much as 40% in industrial regions, and about 10% for most of Canada (Vet et al. 2005).

Wet and bulk deposition measurements of inorganic nitrogen species have been made since the early 1980s at several sites. In Dorset, in central Ontario, near the center of the steep gradient from high southern deposition values to near-pristine northern ones, 26 years of precipitation record show no statistically-significant trend in either nitrate or ammonium. In contrast, sulfate in rainfall has declined by 45-50% during the same period as the result of reduced sulfur oxide emissions from smelters and coal-fired power plants in the region, and recent reductions in the USA (Figure 3). Similarly, the Experimental Lakes Area in northwestern Ontario has collected wet, dry and bulk precipitation for many years. There are no significant trends in deposition of sulfur or nitrogen, although there is a suggestion of a recent upturn in wet deposition of nitrogen (Linsey et al. 1987; Vet et al. 2005). The major source areas for the site are the Midwestern USA, followed by western Canada. Total nitrogen and sulfur deposition at the ELA site are only 2.6 kg N ha⁻¹ y⁻¹ and 2.9 kg S ha⁻¹ y⁻¹ (Vet et al. 2005). Management of nitrogen deposition in eastern Canada cannot be done in isolation from the USA. Canada emits only 7 percent of the combined nitrogen emissions in eastern Canada and the USA,

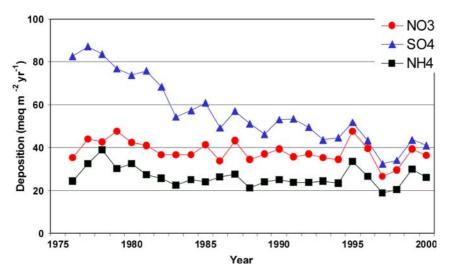


Figure 3. Twenty five years of sulfate, nitrate and ammonium concentrations in bulk deposition at Dorset Ontario.

but receives 32 percent of the resulting wet deposition of nitrogen, because of being downwind of areas of high NO_x production in the American Midwest.

Impact of high nitrogen deposition on ecosystems

Compounds of both sulfur and nitrogen contribute to the acidification of soils and aquatic systems, and with respect to the effects of acidification, the elements cannot be treated in isolation. Various models have been used to predict whether the capacity of terrestrial and aquatic ecosystems to tolerate acidifying substances have been exceeded in eastern Canada. Critical load models combining the deposition of acidifying sulfur and nitrogen compounds similar to those used in Europe have generally been used for these assessments (Critical load is defined here as in the UN-ECE as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" following Nilsson and Grennfelt 1988).

One concern in base-poor soils such as those overlying Precambrian rock has been that continued loading of acidifying substances is leaching calcium in excess of the rate that it is being replenished by deposition and weathering (Likens et al. 1996). Vet et al. (2005) used critical load models for soils and freshwaters to project the extent of acidification damage under "best case" and "worst case" emissions control scenarios for Canada and the USA. It was estimated that between 0.5 and 1.8 million km² or 21 to 75 percent of eastern Canada continues to receive acid deposition in excess of critical loads. The effect of high acid deposition is particularly severe for forest soils, which continue to export base cations in excess of inputs from weathering and deposition. Calcium is particularly badly affected, with soil pools in south-Central Ontario depleted by up to 30% since the early 1980s (Watmough and Dillon 2003a, b). Declines in sugar maple growth (Watmough 2002) and hardwood forest production (Oimet et al. 2001) have been attributed to calcium depletion caused by exceedance of critical loads.

An additional concern is the combination of acid deposition and forest harvesting. Watmough and Dillon (2003b) calculate that the combination of calcium removal by forest harvesting and leaching could leave soils unable to support new forest growth within a few decades because of calcium deficiency. In brief, it appears that controlling acid deposition will be an important consideration in sustainable forestry in eastern Canada.

Aquatic critical load models also indicate widespread acidification damage. Vet et al. (2005) estimate that about 15% of lakes in eastern Canada south of 52° N that historically had pH values > 6 (a critical threshold for many aquatic species) will be incapable of maintaining such pH values under the current acid deposition regime. The total number of lakes in eastern Canada south of 52° N that are affected according to this criterion is 500,000 to 600,000 (Vet et al. 2005). As acidifying emissions continue, the continuing depletion of base ca-

tions in catchment soils will require greater and greater reductions in emissions of sulfur and nitrogen to reach target pH values in receiving waters. Climate warming and drought have also caused the reoxidation of sulfur deposited in catchments during previous periods of high deposition, exacerbating the acidification problem (Bayley et al. 1992; Lazerte 1993).

In the Dorset, Ontario area, where long-term chemical records are available for several lakes and their inflow streams, Henriksen et al. (2002) SSWC steady-state model indicates that the proportion of lakes where critical loads are exceeded has declined from >90 percent in the late 1970s to <40 percent in the late 1990s, largely the result of reductions in sulfur oxide emissions. As the result of these reductions, total inorganic nitrogen deposition (nitrate plus ammonium) in equivalent units is now about 50 percent higher than sulfur deposition (Figure 3). Despite the documented depletion of soil nutrients and nitrogen deposition that exceeds long-term critical loads in analogous parts of Europe, there are no detectable trends in stream or lake concentrations of inorganic nitrogen in central Ontario during the last two decades. Nitrogen export varies considerably between catchments and years, ranging from 0.1 to 1.5 kg N ha⁻¹ y⁻¹ (Watmough and Dillon 2003a).

Dean Jeffries (pers. comm.) has also analyzed 1081 lakes in eastern Ontario using Henriksen's SSWC model. Current deposition exceeds critical loads in 45 percent of the lakes. The nitrogen component of the exceedances is, however, very small.

An exception to the above is Lake Superior, which receives little phosphorus from either the atmosphere or via inflow streams, but receives moderate loading of airborne nitrogen from industries and agriculture in the USA. As a result, nitrate has nearly doubled in the latter half of the 20th century, from 200 μ g l⁻¹ in 1950 to almost 360 μ g l⁻¹ in 2001 (Hugh Dobson, National Water Research Institute, pers. comm.).

Watmough et al. (2004) analyzed trends over a 16-year period in sixteen forested catchments in Ontario. They found that there were no uni-directional long-term trends in stream nitrate concentration, but that there were common long-term patterns. Two stream groupings were identified. Those with shallow soils, moderate slopes and large wetlands had low nitrate concentrations, while those with deeper soils, steeper slopes and little wetland area had high nitrate concentrations. These differences were attributed to various climate factors including greater impact of droughts in the latter group. The importance of the watersheds' characteristics in determining nitrate concentration is streams was also noted by Schiff et al. (2002), who observed nitrate exports differing by a factor of 10 from two neighbouring streams with different slopes and wetland components.

Nitrate plays a role in lowering pH during spring snowmelt in eastern Canada. During snowmelt, concentrations of nitrate and hydrogen ion in streams and lakes can increase several-fold. At some sites, nitrate can exceed $20~\mu eq~l^{-1}$ for short periods in the spring, enough to contribute to depression of pH and alkalinity in lakes where alkalinity is less than $100~\mu eq~l^{-1}$. Alkalinity

and pH depressions also occur in the autumn in streams and the littoral zones of lakes, particularly following dry summers. Such events are also a major problem for the biota (Dillon et al. 1997; Eimers and Dillon 2002). Although sulphate was the dominant factor in most watersheds in these events, in one of nine watersheds an increase in nitrate concentration was the cause of the decline in alkalinity (Laudon et al. 2004).

Kaste and Dillon (2003) compared input and output for several terrestrial catchments in eastern Ontario and Norway, and so far, deposited nitrogen has been more efficiently retained in Canadian forested ecosystems than in similar European catchments. The eight Ontario sites are in an area that receives near-maximum nitrogen deposition, averaging 9 kg TIN ha⁻¹ y⁻¹ for 19–23 years. They all retained over 90% of input. In contrast, four Norwegian catchments that had deposition roughly twice as high retained only 43–71% of input. An exception was Langtjern, which had a 97% TIN retention, but a deposition equal to the Canadian sites. From these data it appears that there is a major breakpoint in terrestrial saturation in northern forests, occurring somewhere between 9 and 18 kg TIN ha⁻¹ y⁻¹, above which leakage from catchments increases rather rapidly. This is within the range estimated by Grennfelt and Hultberg (1986) for nitrate leaching at 10–15 kg N ha⁻¹ y⁻¹ (Reviewed by Jeffries 1995).

At the Turkey Lakes Watershed (TLW), a few hundred km northwest of Kaste and Dillon's sites in Ontario, nitrogen retention was only 65%, at average TIN deposition of about 9 kg ha⁻¹ y⁻¹. There is some evidence that the source of the nitrate in outflow is nitrification of organic nitrogen in forest soils (Spoelstra et al. 2001), and there has been little change in the efficiency of retention over time. Hazlett and Foster (2002) showed that nitrate concentrations in subsurface throughflow draining toward Little Turkey Lake in the TLW varied greatly in relation to slope, reflecting the soil horizons through which the water had passed. In summary, there appears to be stable, and usually high retention of nitrogen at the forested monitoring sites in eastern Canada, even in those that receive the highest nitrogen deposition.

Wetlands are ubiquitous in the boreal ecozone. Beaver ponds (Devito and Dillon 1993a) and *Sphagnum*-conifer swamps (Devito and Dillon 1993b), the two dominant types of wetlands, are effective sinks for inorganic N, although they release an equivalent amount of organic N. They therefore contribute substantially to the effectiveness of landscapes in retaining nitrate and ammonium, although they are in balance with respect to total N.

However, it is still unclear how close to critical loads these systems may be. The FAB model (Posch et al. 1997) has been used in Ontario to calculate the combined critical load of S and N (Aherne et al. 2004). This model takes into account the major biogeochemical processes that involve nitrogen in a lake and its terrestrial catchment, including uptake by vegetation, immobilisation in the soil and denitrification, and in-lake retention. The model assumes that at least some of the N leaks from catchments. Results suggest that the combined S and N deposition exceeds the critical load to a higher proportion of lakes than if

critical loads based on only S were considered. In Europe, there seems to be a several year lag before breakthrough begins at intermediate nitrogen deposition, even at constant rate. A long lag in reversal is also observed at the one site where nitrogen deposition was experimentally decreased (Wright et al. 1994). It would be wise to err on the side of caution, regulating nitrogen emissions to keep deposition in Canada at current values or lower.

Kaste and Dillon (2003) also found that the terrestrial catchments of lakes were the predominant sink for nitrogen because they tended to be large relative to the area of the lake to which they drained (drainage area/lake area ratios of 3–49). On the other hand, lakes were more efficient sinks per unit area, retaining 42–70% of deposition compared to 20–27% for terrestrial catchments. The most important sink in oligotrophic boreal lakes is believed to be denitrification in lake sediments (Kelly et al. 1990; Molot and Dillon 1993).

Western Canada

So far, deposition of nitrogen has been relatively low in most of western Canada. Deposition values are $<2 \text{ kg NO}_3 \text{ ha}^{-1} \text{ y}^{-1}$ for most of the area west of Manitoba (Jeffries et al. 2003), but these are based on relatively few stations. Esther, Alberta, near the center of the western prairies, has a total deposition of only 1.8 kg N ha⁻¹ y⁻¹. One exception is the lower Fraser Valley in B.C., where rates of more than 10 kg N ha⁻¹ y⁻¹ have been reported (Belzer et al. 1998; Schreier et al. 1999). NO_x emissions have increased by 29 percent in western Canada between 1985 and 2000, in contrast to the declines in eastern Canada. As of 2000, NO_x emissions in the west exceeded those in eastern Canada for the first time. Emissions are predicted to increase by another 5 percent between 2000 and 2020. This estimate may be low, due to rapid population growth and massive developments in the Alberta Oil Sands. As described later, these developments are expected to cause enormous increases in nitrogen emissions to the atmosphere (Allen 2004).

There are only measurements of dry deposition at two sites in western Canada, an area half the size of Europe. The climate of this region varies greatly, making it is impossible to generalize about either dry or total deposition. Given rapid increases in population, agriculture and industry in the west, more deposition monitoring stations must be considered as an urgent priority.

There is a slight suggestion of recently increased deposition of nitrate in southern Alberta, at the only site in the Rocky Mountains with long term data. Although annual values are scattered, deposition appears to have doubled in about 15 years, from approximately 2 to about 4 kg NO₃ ha⁻¹ y⁻¹ (Alberta Environment, unpublished data). The region is not immediately downwind of any large urban or industrial development. However, rapidly-growing industrial areas near Calgary Alberta occupy a position on the eastern slopes of the Rocky Mountains similar to those of Colorado, where nitrogen deposition has

increased rapidly as industry and population have expanded (Williams et al. 1996). Dated ice samples taken in 1995 from glaciers at the highest point in the Canadian Rockies showed low concentrations and no trend in ammonia and nitrate deposition in 50 years (D.W. Schindler, unpublished data), but this situation may have changed in the ensuing decade. There is some evidence that N deposition is increasing downwind of major population centers, as described in the next section.

Increasing humans, livestock and commercial fertilizer as sources of atmospheric nitrogen

Rapidly increasing populations of humans, industrial development, livestock, fertilizer manufacturing, and increasing use of manure and commercial fertilizer in southern British Columbia and Alberta are causing increased atmospheric concentrations of NO_x and ammonia (Schreier et al. 2003; Schreier and Brown 2004). Vet et al. (2005) estimate that ammonia emissions in western Canada increased by 8 percent between 1985 and 2000. Most of the emissions were in southern parts of the prairie provinces and the lower Fraser River watershed, where agriculture is concentrated. Highest values were in central Alberta and coastal British Columbia, where both agriculture and fossil fuel combustion are greatest. Ammonia emissions in western Canada are projected to increase by 57 percent between 2000 and 2020, largely the result of livestock and poultry culture and other agricultural activities. Nitrogen released by centers of human population is causing increased airborne emissions and deposition of nitrogen well outside the geographical bounds of cities. Kochy and Wilson (2001) found that deposition of nitrogen was higher in national parks downwind of prairie cities than in more remote parks of the prairies. Cheng (1994) predicted such results from modelling studies in Alberta. His models predicted deposition of up to 4 kg N ha⁻¹ y⁻¹ near Edmonton and Calgary, cities of approximately 1 million people each. However, these deposition values are lower than those reported by Kochy and Wilson, using resin bags rather than precipitation collections. Their results show deposition of 8 kg N ha¹ y⁻¹ for the most pristine sites in Jasper National Park of the Rocky Mountains, and 22 kg N ha⁻¹ y⁻¹ for the highest sites downwind of Edmonton. Intercalibration of such methods with direct analysis of precipitation samples is in order before results of the two methods can be realistically compared.

The Athabasca Oil Sands

Another potential new source of nitrogen to the atmosphere is rapid development in the oil sands of northeastern Alberta. So far, the two oil sands plants that have been in operation for almost 30 years have not produced excessive emissions of nitrogen. But with rapidly-increasing oil prices, a shortage of

secure sources of oil in the Middle East, and recent hurricane damage to oil rigs and refineries around the Gulf of Mexico, expansion has escalated. A third oil sands plant is now operating, two more are under construction, and several others are in the approval process. Huge trucks capable of carrying hundreds of tonnes each are expected to produce NO_x that will cause nitrogen emissions to increase by 359% over 1998 values in the near future (Environment Canada 2003). Nitrate deposition in the mid-1990s was generally less than 2 kg ha⁻¹ y⁻¹ (Anonymous 2000). Near the center of oil sands activity, nitrogen deposition is projected to be as high as 65 kg N ha⁻¹ y⁻¹ (Allen 2004), and deposition greater than 20 kg N ha⁻¹ y⁻¹ is expected over thousands of square kilometres (Anonymous 2000). Fortunately, most of the region around the oil sands is not geologically-susceptible to acidification. The nearest areas of concern from acidification is in northern Saskatchewan, over 100 km east of the oil sands, and in a few parts of the Birch Mountains, about 100 km to the northwest. Prevailing winds in the area are from the east, followed by the west. So far, modelled emission and deposition scenarios have not been verified with precipitation measurements, and the establishment of a precipitation monitoring network should be given high priority. Emissions of CO₂ and SO₂ are also expected to increase substantially as the result of oil sands development.

In summary, little information is available on sources or deposition of atmospheric nitrogen in much of western Canada. There have been no analyses of where critical loads of acidifying substances may be exceeded at present or in the future. Such studies are urgently required in order to plan for ecologically sustainable development in several areas of rapid population and industrial growth, including the lower Fraser River Valley of British Columbia, the corridor from Calgary to Edmonton in Alberta, and the Athabasca Oil Sands in northern Alberta.

Agriculture and urbanization as direct sources of nitrogen to water

The most rapidly increasing source of nitrogen in Canada is from agriculture. Much of the increase in dissolved nitrogen results from direct movement of nitrogen to water, rather than via the atmosphere. Both the use of commercial fertilizers and the production of manure from livestock husbandry have increased very rapidly. The increases in nitrogen are much more rapid than those in phosphorus and potassium, the other two main elements in commercial fertilizer. Since records began in 1950, nitrogen fertilizer production in Canada has increased about 75-fold, while use of phosphorus and potassium have increased by only 5 and 7-fold, respectively (Korol and Rattray 1999).

In some cases it is difficult to distinguish losses of nitrogen by agriculture to water from losses to air. Releases can be as N_2O , a potent greenhouse gas and precursor of urban smog, or as ammonia, which produces odors at high

concentrations, is a NO_x precursor, and can both acidify sensitive soils and act as a fertilizer.

Increased demand for meat, particularly in the USA and the Orient, and decreased financial returns per animal have caused an explosive increase in livestock culture in Canada. In several provinces, populations of cattle, hogs and poultry have increased several fold since the mid-20th century. In the five years 1996–2001, Canadian cattle increased by 4.4%, hogs by 26.4%, and chickens by 23.4%, despite a human population growth of only 4.0% (Stats-Can, 2001). Alberta currently has the most cattle, 6.4 million. They increased by 11.3% in the 1996–2001 period. The stated goal of the Alberta government is to double that number. Hog increases have been greatest in Quebec and Manitoba, but numbers in Alberta, Saskatchewan and other provinces have also increased substantially. Chicken number have increased most rapidly in British Columbia, by 37% over the same 5 year time period.

Chambers et al. (2001) calculate that 4.3 million tonnes of nitrogen were added to Canadian farm land in 1996 from anthropogenic sources. Of this, 1.97 million tonnes were applied to agricultural land as manure, fertilizer and biosolids, 0.77 million tonnes were fixed by domestic legumes, and 0.043 million tonnes as atmospheric deposition of nitrate and ammonium. Only 58 percent of the amount added to agricultural lands was removed in the form of crops. A total of 0.3 million tonnes of nitrogen were estimated to enter fresh, ground and coastal waters as a result of human activity.

Most of the reported exceedances of groundwater standards for human consumption of nitrate in Canada were in agricultural areas. Seventeen percent of agricultural land in Ontario, 6% in Quebec, and 3% in Atlantic provinces would be expected to produce runoff or seepage with >14 mg l⁻¹ N, largely as nitrate (Chambers et al. 2001). In Ontario, a significant proportion of both shallow and deep wells have nitrate in excess of drinking water standards of 10 mg NO_3 l⁻¹ (Goss et al. 1998; Rudolph et al. 1998).

According to Canadian statistics, hogs are particularly large sources of phosphorus and nitrogen, with approximately 10 and 5 times the per capita output of humans, respectively (Chambers et al. 2001). The application of manure to land is high in parts of Canada that have high populations of livestock. Most notably, southern Alberta, southern Ontario and Quebec, and the lower Fraser Valley and Okanagan regions of southern B.C. have manure applications of 1000 kg N ha⁻¹ and more.

Eighty percent of nitrogen consumed by farm animals is excreted or egested, after which 4–95% is lost to the atmosphere. Agriculture emitted an estimated 132 thousand tonnes of N₂O–N in Canada in 1996, with the largest sources in Alberta and Saskatchewan. The analogous figure for NH₃–N was 849 thousand tonnes from agriculture (1990 values). Fifty-five percent of the emissions were from fertilizer. Anhydrous ammonia is the most popular fertilizer in Canada, followed by urea. Both can have high losses to the atmosphere and acidify the soils. Fifty five percent of the nitrogen emissions from livestock are emitted by cattle (Desjardins and Keng 1999). Some of the highest atmospheric

ammonia deposition has been measured in the most intensively used agricultural area in the lower Fraser Valley in B.C. where annual total atmospheric ammonia deposition rates of 9.5 kg N ha⁻¹ y⁻¹ were measured in 1997 (Schreier et al. 1999).

The lower Fraser River Valley in British Columbia is one of the most rapidly developing areas of Canada, with human populations and intensive agriculture increasing very rapidly (Schreier et al. 2003). The region has the highest density of livestock in Canada. Livestock culture is also changing rapidly from traditional methods to industrial confined feeding operations (CFOs). Nitrogen applications of both manure and commercial fertilizer are high, with over 50% supplied by manure in many of the subregions. Total applications in most areas are 200 to more than 300 kg N ha⁻¹ y⁻¹. In most cases, these applications are in excess of plant needs, and excesses of application/demand are typically over 100 kg N ha⁻¹ y⁻¹, with calculated excesses in some areas exceeding 300 kg N ha⁻¹ y⁻¹. While populations of cattle, hogs and horses appear to be relatively stable, there has been an explosion in poultry culture, with chicken populations nearly doubling in between 1991 and 2001 (Schreier et al. 2003).

Commercial fertilizer use in Canada has also increased, by 20–30% every 5 years, according to Statistics Canada data (Statscan 2001). Using a nutrient budget modelling approach, Schreier et al. (2003) calculated that average annual surplus applications for the arable area of the lower Fraser Valley were more than 50 kg N ha⁻¹ y⁻¹. Seven of the twenty subregions of the area had application rates exceeding 100 kg N ha⁻¹ y⁻¹ (Figure 4, Schreier and Brown 2004). In the Sumas River watershed, which is located in the most intensively used agricultural area in the valley, surplus levels of up to 300 kg N ha⁻¹ y⁻¹ were reported by Berka et al. (2001) and Schreier et al. (2003). Berka (1996) showed a significant positive relationship between surplus application rates and ammonia concentrations in surface and groundwater of the Sumas River watershed during the winter season and a significant negative relationship between surplus N and dissolved oxygen. Nitrate concentrations in the river have gone up steadily for over 30 years (Figure 5). Smith (2004) showed positive correlations between animal stocking density and surplus nitrogen applications within stream buffer zones (100 m wide on both sides of the river, and 500 m long stream segments) and ammonia and nitrate values in the Sumas River during the winter. Smith was also able to show how the rapid increase in animal stocking density between 1973 and 2003 has impacted the nitrate levels in the streamwater during the winter, when active uptake by vegetation is low.

Urbanization presents other problems. Decreased infiltration of water, coupled with lawn fertilizer, pet excrement, street dust, etc. can make storm water quality quite similar to that of sewage. Destruction of wetlands (80% in the lower Fraser, 70% in the western prairies), channelization of streams, and destruction of riparian buffer zones lead to the efficient delivery of all chemicals to water.

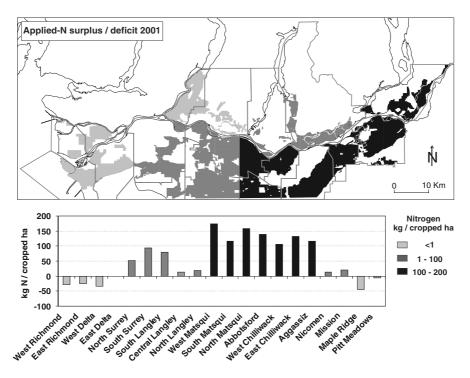


Figure 4. Annual surplus/deficit nitrogen application in 20 enumeration areas in the Lower Fraser Valley in B.C. in 2001. Values are in kg N $\rm ha^{-1}~y^{-1}$ and represent application rates above crop nutrient requirements.

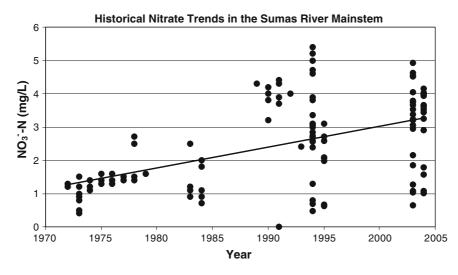


Figure 5. Trends in nitrate in the Sumas River, British Columbia, showing the effects of increasing agricultural intensity between 1972–2004.

In most regions of Canada, regulations for discharge of animal wastes are rudimentary. Most wastes are simply spread or sprayed on the land. Hog operations usually discharge to surface lagoons, which have high ammonia losses to the atmosphere. Timing and rates of manure application are not well-regulated, with application on frozen ground being common. Riparian corridors and wetlands are filled and destroyed in many regions. On average, 70% of wetlands in southern Canada have been destroyed, with little regard for their role as important nitrogen sinks or sites of denitrification. As a result, nitrogen has increased greatly in many surface waters. Phosphorus has also increased, and as most waters in the western prairies are phosphorus rich to begin with, one result is rapidly increasing eutrophication.

Eutrophication of freshwaters and marine ecosystems

Most Canadian freshwaters are phosphorus limited, and in general phosphorus released from human activity has increased even more rapidly than nitrogen (Chambers et al. 2001; Schindler 2001). In Lake Winnipeg, concentrations of total nitrogen have more than doubled in less than 10 years, from about 350 to over 800 μ g l⁻¹. Phosphorus and chlorophyll have also increased, from 30 to 55 μ g l⁻¹ and 5 to over 20 μ g l⁻¹, respectively. The current state of the lake with respect to nutrient concentrations and algal blooms is roughly the same as Lake Erie was at the height of the eutrophication problem in the early 1970s (M.P. Stainton, Freshwater Institute, pers. comm.).

Increasing trends for phosphorus, nitrogen and algal populations are commonly identified in paleoecological studies of western lakes (Blais et al. 2000; Schindler unpublished data). In general, phosphorus has increased more rapidly than nitrogen, resulting in the proliferation of nitrogen-fixing Cyanobacteria. In the west, increasing temperatures (1–3° C in the western prairies), and drought conditions (which have caused huge declines in water flows in rivers and water renewal times in lakes) have exacerbated problems with water quantity and quality (Schindler and W. Donahue, unpublished data).

Land-use change has been responsible for some of the nutrient increases in lakes in Canada. The prairies have been largely converted to agricultural land, so that new lands for agriculture are taken from areas that were originally forested. Studies in Ontario (Dillon and Kirchner 1975) and Alberta (Neufeld and Schindler unpublished) indicate that simply clearing a small parts of forested catchments for agriculture can cause yields of nutrients to increase by at least two-fold. Once cleared, pastures or fertilized croplands usually follow, causing additional increases in nutrient runoff. Many of the lakes in western Canada have undergone moderate to severe eutrophication as a result of these changes. In Alberta, with its rapid population growth, rapid lakeshore development has contributed to eutrophication in many of the same lakes.

So far, there is little sign of coastal eutrophication in Canada even though large coastal cities like Halifax, Victoria, and part of Vancouver discharge

sewage directly into coastal waters. Perhaps the most threatened area in the country is the Georgia Basin, which receives the discharge of the Fraser River as mentioned above, but is surrounded by 4.5 million people and their enterprises, if the combined populations of the Vancouver Area, Victoria on Vancouver Island, and the American city of Seattle are included. The population of the area is increasing rapidly, with the 2010 population predicted to be 28% higher than at present. Eutrophication is presently confined to small protected bays, because of high exchange rates with the main Pacific Ocean via the Strait of Juan de Fuca (Chambers et al. 2001).

On the east coast of Canada, Halifax Harbor has received the effluents from the cities of Halifax, Dartmouth and Bedford for many decades. Raw sewage is discharged, and although the human population is much smaller (343000) than the Georgia Basin, the Bedford Basin and Halifax Harbor form a small, protected marine estuary. Fortunately, very high tides produce a rapid exchange with the main Atlantic Ocean. While it is difficult to dissect the effects of nitrogen from other components of raw sewage, dinoflagellate blooms and occasional summer fish kills have been observed (Chambers et al. 2001).

Summary

While Canada has so far escaped major problems with airborne or waterborne nitrogen, many of the problems experienced by other countries are beginning to emerge as human populations and industry increase and livestock culture intensifies. Potential problems with acid rain, increased nitrate in surface and ground water, and eutrophication, especially of marine estuaries, are problems that are expensive and difficult to control once they have reached critical stages. It would be economically astute to limit nitrogen releases before the problem becomes acute.

Acknowledgements

We are grateful to Dean Jeffries for reviewing an early draft of the manuscript and for providing unpublished data. Bob Vet supplied copies of nitrogen deposition maps for eastern Canada. Michael Stainton shared his recent chemical data for Lake Winnipeg. Research by DWS was supported by a Natural Sciences and Engineering Research Council Discovery Grant # 89673.

References

Aherne J., Posch M., Dillon P.J. and Henriksen A. 2004. Critical loads of acidity for surface waters in south-central Ontario, Canada: regional application of the First-order Acidity Balance (FAB) model. Water Air Soil Pollut. Focus 4: 25–36.

- Allen E. 2004. Effects of nitrogen deposition on forests and peatlands: A literature review and discussion of the potential impacts of nitrogen deposition in the Alberta Oils Sands Region. Wood Buffalo Environmental Association, Fort McMurray, AB.
- Bayley S.E., Schindler D.W., Parker B.R., Stainton M.P. and Beaty K.G. 1992. Effects of forest fire and drought on acidity of a pase-poor boreal forest stream: similarities between climate warming and acidic precipitation. Biogeochemistry 17: 191–204.
- Belzer W., Evans C. and Poon A. 1998. Atmospheric Nitrogen Concentrations in the Lower Fraser Valley. Environment Canada, Aquatic and Atmospheric Science Division, Vancouver, B.C. DOE FRAP 1997: 1–23.
- Berka C., Schreier H. and Hall K. 2001. Linking Water Quality with agricultural intensification in a rural watershed. Water Air Soil Pollut. 127(1–4): 389–401.
- Berka C. 1996. Relationships between agricultural land use and surface water quality using GIS: Sumas River Watershed, Abbotsford, B.C. M.Sc. Thesis. Resource Management and Environmental Studies, University of British Columbia. June 1996, 174 pp.
- Blais J.M., Duff K.E., Schindler D.W., Smol J.P., Leavitt P.R. and Agbeti M. 2000. Recent eutrophication histories in Lac Ste Anne and Lake Isle, Alberta, Canada, inferred using paleo-limnological methods. Lake and Reserv. Manage. 16: 292–304.
- Chambers P.A., Guy M., Roberts E.S., Charlton M.N., Kent R., Gagnon C., Grove G. and Foster N. 2001. Nutrients and their impact on the Canadian environment. Agriculture and Agri-Food Canada, Environment Canada, Fisheries and Oceans Canada, Health Canada and Natural Resources Canada, Ottawa. 241 p.
- Cheng L. 1994. Concentration and Deposition of Anthropogenic Pollutants in Alberta: Summary Report. Alberta Research Council, Edmonton, p. 10.
- Desjardins R.L. and Keng J.C. 1999. Nitrous oxide emissions from agricultural sources in Canada. In: Desjardins R.L. Keng J.C. and Haugen-Kozyra K. (eds) Proceedings of the International Workshop on Reducing nitrous Oxide Emissions from Agroecosystems, Banff, Alberta March 3–5 1999. Conservation and Development Branch, Alberta Agriculture, Food and Rural Development, Agriculture and Agri-Food Canada, Research Branch, Edmonton, Alberta.
- Devito K.J. and Dillon P.J. 1993a. The importance of runoff and winter anoxia to the P and N dynamics of a beaver pond. Can. J. Fish. Aquat. Sci. 50: 2222–2234.
- Devito K.J. and Dillon P.J. 1993b. The influence of hydrologic conditions and peat oxia on the phosphorus and nitrogen dynamics of a conifer swamp. Wat. Resourc. Res. 29: 2675–2685.
- Dillon P.J. and Kirchner W.B. 1975. The effects of geology and land use on the export of phosphorus from watersheds. Water Res. 9: 135–148.
- Dillon P.J., Molot L.A. and Futter M. 1997. A note on the effect of El Niño-related drought on the recovery of acidified lakes. Int. J. Environ. Mon. Assess. 46: 105–111.
- Eimers M.C. and Dillon P.J. 2002. Climate effects on sulphate flux from forested catchments in south-central Ontario. Biogeochemistry 61: 337–355.
- Environment Canada 2003. Submission to the Alberta Energy and Utilities Board. Canadian Natural Resources Ltd. Horizon Oil Sands Mine Application 1273113. Sept. 2003.
- Goss M.J., Barry D.A.J. and Rudolph D.L. 1998. Contamination in Ontario farmstead domestic wells and its association with agriculture: 1. Results from drinking water wells. J. Contam. Hydrol. 32: 267–293.
- Grennfelt P. and Hultberg H. 1986. Effects of nitrogen deposition on the acidification of terrestrial and aquatic ecosystems. Water Air Soil Pollut. 30: 945–963.
- Hazlett P.W. and Foster N.W. 2002. Topographic controls of nitrogen, sulfur and carbon transport from a tolerant hardwood hillslope. Water Air Soil Pollut. Focus 2: 63–80.
- Henriksen A., Dillon P.J. and Aherne J. 2002. Critical loads of acidity for surface waters in south-central Ontario, Canada: regional application of the Steady-State Water Chemistry (SSWC) model. Can. J. Fish. Aquat. Sci. 59: 1287–1295.
- Jeffries D.S. 1995. Freshwater acidification in Canada caused by atmospheric deposition of nitrogen pollutants: A preliminary assessment of existing information. A draft discussion paper. National Water Research Institute, Burlington, Ontario contribution 95–116, 68 pp.

- Jeffries D.S., Clair T.A., Couture S., Dillon P.J., Dupont J., Keller W., McNicol D.K., Turner M.A., Vet R. and Weber R. 2003. Assessing the recovery of lakes in southeastern Canada from the effects of acid deposition. Ambio 32: 176–182.
- Kaste O. and Dillon P.J. 2003. Inorganic nitrogen retention in acid-sensitive lakes in southern Norway and southern Ontario, Canada-a comparison of mass balance data with an empirical N retention model. Hydrol. Processes 17: 2393–2407.
- Kelly C.A., Rudd J.W.M. and Schindler D.W. 1990. Acidification by nitric acid future considerations. Water Air Soil Pollut. 50: 49–61.
- Kochy M. and Wilson S.D. 2001. Nitrogen deposition and forest expansion in the northern Great Plains. J. Ecol. 89: 807–817.
- Korol M. and Rattray G. 1999. Canadian Fertilizer Consumption, Shipments and Trade, 1997–1998. Agri-Foods and Agriculture Canada, Ottawa.
- Laudon H., Dillon P.J., Eimers M.C., Semkin R.G. and Jeffries D.S. 2004. Climate induced episodic acidification of streams in central Ontario. Environ. Sci. Technol. 38: 6009–6015.
- Lazerte B.D. 1993. The impact of drought and acidification on the chemical exports from a minerotrophic conifer swamp. Biogeochemistry 18: 153–175.
- Likens G.E., Driscoll C.T. and Buso D.C. 1996. Long-term effects of acid rain: response and recovery of a forest ecosystem. Science 272: 244–246.
- Linsey G.A., Schindler D.W. and Stainton M.P. 1987. Atmospheric deposition of nutrients and major ions at the Experimental Lakes Area in northwestern Ontario. Can. J. Fish. Aquat. Sci. 44(Suppl. 1): 206–214.
- Molot L.A. and Dillon P.J. 1993. Nitrogen mass balances and denitrification rates in central Ontario Lakes. Biogeochemistry 20: 195–212.
- Nilsson J. and Grennfelt P. 1988. Critical Loads for Sulfur and Nitrogen. Nordic Council of Ministers, Copenhagen, Denmark, p. 31.
- Oimet R., Duschesne D., Houle D. and Arp P.J. 2001. Critical load and exceedances of acid deposition and associated forest growth in the northern hardwood and boreal coniferous forests in Quebec, Canada. Water Air Soil Pollut. Focus 1: 119–134.
- Posch M., Kamari J., Forsius M., Henriksen A. and Wilander A. 1997. Exceedance of critical loads for lakes in Finland, Norway and Sweden: Reduction requirements for acidifying nitrogen and sulfur deposition. Environ. Manage. 21: 291–304.
- Rudolph D.L., Barry D.A.J. and Goss M.J. 1998. Contamination in Ontario farmstead domestic wells and its association with agriculture: 2. Results from multilevel monitoring well installations. J. Contam. Hydrol. 32: 295–311.
- Schiff S.L., Devito K.J., Elgood R.J., McCrindle P.M., Spoelstra J. and Dillon P.J. 2002. Two adjacent catchments: dramatically different NO₃ export. Wat. Resourc. Res. 38: 1292, doi: 10.1029/2000WR000170.
- Schindler D.W. 2001. The cumulative effects of climate warming and other human stresses on Canadian freshwaters in the new millennium. Can. J. Fish. Aquat. Sci. 58: 18–29.
- Schreier H., Bestbier R. and Derksen G. 2003. Agricultural nutrient management trends in the Lower Fraser Valley, BC (Based on Agricultural Census 1991–2001). (Compact Disc) IRES, University of British Columbia.
- Schreier H. and Brown S. 2004. Multiscale approaches to water management: land-use impacts on nutrient and sediment dynamics. In: Tchiguirinskaia I., Bonell M. and Hubert P. (eds), Scales in Hydrology and Water Management. International Association of Hydrological Sciences (IAHS), Wallingord, GB, pp. 61–75 (IAHS publication 287).
- Schreier H., Hall K., Brown S.J., Wernick B. and Berka C. 1999. Agriculture: an important non-point source of pollution. In: Gray C. and Tuominen T. (eds), Health of the Fraser River Aquatic Ecosystem, Vol. II. Environment Canada, DOE-FRAP, Vancouver, 83–99.
- Smith I. (2004) Cumulative effects of agricultural intensification on nutrients and trace metal pollution in the Sumas River watershed. M.Sc. Thesis, Resources Management and Environmental Studies, University of British Columbia, Vancouver, 275 pp.

- Spoelstra J., Schiff S.L., Elgood R.J., Semkin R.G. and Jeffries D.S. 2001. Tracing the sources of exported nitrate in the Turkey Lakes Watershed using ¹⁵N/¹⁴N and ¹⁸O/¹⁶O isotopic ratios. Ecosystems 4: 536–544.
- StatsCan 2001. Agricultural Census Data. Statistics Canada, Ottawa, On.
- Vet R., Brook J., Ro C., Shaw Narayan J., Zhang L., Moran M. and Lusis M. 2005. Atmospheric response to past emission control programs. In: Canadian Acid Rain Science Assessment 2004, Chapter 3. Environment Canada, Ottawa Ontario, pp. 15–98.
- Watmough S.A. 2002. A dendrochemical survey of sugar maple in south-central Ontario. Water Air Soil Pollut. 136: 165–187.
- Watmough S.A. and Dillon P.J. 2003a. Base cation and nitrogen budgets for seven forested catchments in central Ontario 1983–1999. Forest Ecol. Manag. 177: 155–177.
- Watmough S.A. and Dillon P.J. 2003b. Calcium losses from a forested catchment in south-central Ontario, Canada. Environ. Sci. Technol. 37: 3085–3089.
- Watmough S.A., Eimers M.C., Aherne J. and Dillon P.J. 2004. Climate effects on nitrate export from forested catchments in south central Ontario. Environ. Sci. Technol. 38: 2383–2388.
- Whelpdale D.M. and Galloway J.N. 1994. Sulfur and reactive nitrogen oxide fluxes in the North Atlantic atmosphere. Global Biogeochem. Cycles 8: 481–493.
- Williams M.W., Baron J., Caine N., Summerfeld R. and Sanford R. 1996. Nitrogen saturation in the Rocky Mountains. Environ. Sci. Technol. 30: 640–646.
- Wright R.F., Cosby B.J., Ferrier R.C., Jenkins A., Bulger A.J. and Harriman R. 1994. Changes in acidification of lochs in Galloway, southwestern Scotland, 1979–1988: The MAGIC model used to evaluate the role of afforestation, calculate critical loads, and predict fish status. J. Hydrol. 161: 257–285.