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Patterns of nitrogen and sulfur accumulation and retention in ombrotrophic bogs, eastern Canada

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Abstract

Nitrogen (N) and sulfur (S) play important roles in peatlands, through their influence on plant production and peat decomposition rates and on redox reactions, respectively, and peatlands contain substantial stores of these two elements. Using peat N and S concentrations and dry bulk density and ^{210}Pb dating, we determined the rates of N and S accumulation over the past 150 years in hummock and hollow profiles from 23 ombrotrophic bogs in eastern Canada. Concentrations of N and S averaged 0.80% and 0.18%, respectively, generally increased with depth in the profile and there was a weak but significant correlation between N and S concentrations. Rates of N and S accumulation over the past 50–150 years ranged from 0.5 to 4.8 g N m^{-2} yr^{-1} and from 0.1 to 0.9 g S m^{-2} yr^{-1}. There were significant but weak correlations between C, N and S accumulation rates over 50-, 100- and 150-year periods. Over the last 50 years, rates of S accumulation showed little differentiation between hummocks and hollows, whereas the pattern for N accumulation was more variable (hummock minus hollow rate ranged from -1 to $+1.5$ g N m^{-2} yr^{-1}), with hummocks generally having a larger N accumulation rate, correlated with the rate of carbon (C) accumulation. There was a modest but significant positive correlation between 50-year rates of N accumulation and wet atmospheric deposition of N measured between 1990 and 1996, with accumulation rates about four times that of wet deposition. The difference between deposition and accumulation of N is attributed to organic N deposition, dry deposition and N_2 fixation. A weaker, but still significant, correlation was observed between 50-year S accumulation and 1990–1996 wet atmospheric S deposition, with about 75% of the deposited S accumulating in the peat. A laboratory experiment with peat cores exposed to varying water table position and simulated N and S deposition, showed that on average 87% and 98% of the deposited NH_4^+ and NO_3^-, respectively, and 58% of the deposited S were retained in the vegetation and unsaturated zone of the cores, supporting the results from the field study.

Keywords: atmospheric deposition, bog, carbon, nitrogen, peat, sulfur

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Introduction

The role of northern peatlands as major global sinks of carbon (C) has been well established: they have accumulation rates of between 10 and 30 g C m^{-2} yr^{-1}, and contain between 5 and 250 kg C m^{-2} with a global C mass of between 250 and 450 Pg (Gorham, 1991; Turunen *et al.*, 2001). The accumulation and retention of nitrogen (N) and sulfur (S) in peatlands are less established. N is an important, growth-limiting nutrient in many peatlands (e.g. Aerts *et al.*, 1992; Berendse *et al.*, 2001) and can be added to the peat surface through dry and wet atmospheric deposition and fixed from

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atmospheric N_2 by microbes (e.g. Hemond, 1983; Urban & Eisenreich, 1988). S plays an important role in the oxidation–reduction biogeochemistry of peatlands, influencing the production and emission of methane to the atmosphere (e.g. Gauci *et al*., 2002, 2004; Blodau & Moore, 2003; Vile *et al*., 2003) and the mineralization of C (e.g. Wieder & Lang, 1988; Vile *et al*., 2003). In addition to a supply from groundwaters, S can be supplied to peatlands through dry and wet atmospheric deposition. Based on common C : N and C : S ratios in peat of 40 : 1 and 250 : 1, northern peatlands contain up to 10 Pg N and 2 Pg S, stored in the last 10 000 years.

The rates of atmospheric deposition of N and S have increased in the past 150 years in North America and Europe, as a result of acid precipitation, and may have influenced rates of plant production and decomposition and C cycling. The concentration of N in peat has been reported in many studies (0.04–0.29%, see Turetsky *et al*., 2000) and one can estimate long-term rates of N accumulation of 0.3–1.0 $g\,N\,m^{-2}\,yr^{-1}$, based on C accumulation rates of 10–30 $g\,C\,m^{-2}\,yr^{-1}$ (Gorham, 1991) and a C : N ratio of 30 : 1 to 40 : 1. Kuhry & Vitt (1996) reported long-term N accumulation rates of 0.2–0.5 $g\,m^{-2}\,yr^{-1}$ in peatlands of western Canada. Urban & Eisenreich (1988) calculated short-term accumulation rates of about 2 $g\,N\,m^{-2}\,yr^{-1}$ over 20–85 years in a Minnesota bog, similar to those observed in southern Sweden (Malmer & Holm, 1984).

Fewer studies report the S concentration in peat or the rate of S accumulation. Peat S concentrations range greatly from 10 to 100 $\mu g\,g^{-1}$ in a Minnesota bog (Urban *et al*., 1989), to 800–7000 $\mu g\,g^{-1}$ in peatlands in West Virginia, Czechoslovakia and Alberta (Wieder & Lang, 1986, 1988; Novák & Wieder, 1992; Turetsky *et al*., 2000) and 700–1400 $\mu g\,g^{-1}$ in *Sphagnum* moss samples from Britain (Bottrell & Novák, 1997). Estimates of S accumulation in peatlands over 20–100 years range from 0.2 to 0.5 $g\,m^{-2}\,yr^{-1}$ in a Minnesota bog (Urban *et al*., 1989), 0.06–0.16 $g\,m^{-2}\,yr^{-1}$ in Alberta peatlands (Turetsky *et al*., 2000) and 0.3–1.5 $g\,m^{-2}\,yr^{-1}$ in a variety of sites (Novák *et al*., 1994).

To measure the recent ($<$150 years) accumulation rates of N and S and to establish the influence of atmospheric deposition on these rates, we sampled 23 ombrotrophic bogs along a transect from north-western Ontario to the Maritimes, Canada, representing a variation in climate and of atmospheric deposition of N and S. In each bog, we sampled a hummock and a hollow and analyzed the upper 50–80 cm for C, N and S concentration. Using these concentrations and dry bulk density, we used the ^{210}Pb dating method to estimate the accumulation rates of these elements over the past 150 years. To estimate retention rates of wet deposition we carried out a greenhouse experiment on peatland cores adding N and S to mesocosms at different loads and with different water table positions.

In this paper, we:

(i) examine the rates of peat accumulation of N and S over 50, 100 and 150 years and their relationship with C accumulation over that period;
(ii) examine differences in peat hummock and hollow accumulation of C, N and S at the bogs;
(iii) test the hypothesis that rates of N and S accumulation in peat over the past 50 years are correlated with rates of wet atmospheric deposition of N and S measured from 1990 to 1996;
(iv) estimate rates of atmospheric N and S deposition over the past 150 years and compare these values with peat accumulation rates of N and S; and
(v) determine whether N and S added to intact peatland mesocosms under experimental green house conditions are retained under different long- and short-term deposition rates and water table levels.

Materials and methods

Study area and sampling

Twenty-three ombrotrophic bogs were selected (Table 1) based on the work of Damman & Dowhan (1981), Glaser & Janssens (1986), Damman (1988), and Gorham *et al*. (1985). All peatlands were characterized by an open canopy of *Picea mariana*, with *Chamaedaphne calyculata, Ledum groenlandicum, Kalmia angustifolia, Kalmia polifolia, Vaccinium uliginosum, Vaccinium myrtilloides, Vaccinium oxycoccos, Andromeda polifolia* and scattered *Eriophorum spissum*. In coastal regions, the shrub cover was less dominant than in more continental areas but the same species were found in all bogs. A few *Pinus banksiana* and *Larix laricina* were found as a mixture with *Picea mariana* in several peatlands of Ontario and Québec. Hummocks were dominated by *Sphagnum fuscum*, with scattered *S. magellanicum, S. capillifolium, Polytrichum* spp., *Cladina* spp., *Empetrum nigrum* and *Sarracenia purpurea*, and hollows by *S. rubellum*. and *S. angustifolium*. In coastal regions, *S. flavicomans* was commonly found in the sampled hummocks as a mixture with *S. fuscum*. Water table position during the summer months ranged from 30 to 60 cm beneath the surface in hummocks and 5–20 cm in hollows.

Recent rates of C, N and S accumulation (RERCA, RERNA and RERSA, respectively) were measured by collecting two short cores (50 cm) using a box sampler (85 × 85 × 1000 mm) at a hummock and a hollow site in each bog. Each short core was double-wrapped in polythene bags, and stored at −4 °C. Additional peat samples were collected down to 80 cm for each core in case they were needed for further analysis.

Additional peat cores to study the retention process of added S and N in mesocosms experiments were taken from hollows in two peatlands at the low and high end of the sulfur and nitrogen deposition gradient. The low deposition peatland ($<0.4\,g\,N\,m^{-2}\,yr^{-1}$ and $<0.2\,g\,S\,m^{-2}\,yr^{-1}$) was located in the Experimental Lakes Area (ELA), near Kenora, north-western Ontario; the mean annual temperature is 1.8 °C and the mean annual precipitation is 678 mm. It is a small acidic and oligotrophic peatland located in the north-western watershed of Lake 239 on the Precambrian Shield (Bayley *et al*., 1986). The peatland is dominated by *P. mariana* and *S. magellanicum, S. angustifolium* and *S. fuscum*. The high deposition peatland was Mer Bleue, located east of Ottawa, eastern Ontario (Table 1). It is a large ombrotrophic bog with a shrub layer of *C. calyculata* and *K. angustifolia* and mosses dominated by *S. magellanicum* in hollows (Moore *et al*., 2002).

Laboratory analyses

Measured volumes of the peat samples were dried to a constant mass at 70 °C, weighed and the dry bulk density calculated. Ten samples of 1 cm thickness were taken from each peat core at 5 cm intervals and a dry 0.5 g subsample from each peat layer was used for analysis. Peat C and N concentrations were analyzed using a LECO CHN analyzer (Mississauga, Ontario, Canada), and S concentration was determined by an Elementar vario EL analyzer (Hanan, Germany).

^{210}Pb activity, as estimated by the α emitting ^{210}Po granddaughter, was determined after spiking with a ^{209}Po yield tracer. The theory of ^{210}Pb dating ($t_{1/2}$ = 22.3

Table 1 Location and characteristics of 23 ombrotrophic peatlands sampled in eastern Canada with climatic characteristics and estimated atmospheric wet deposition of nitrogen (N) and sulfur (S) and N and S deposition zones

Province	Peatland	North (latitude)	West (longitude)	Mean annual temperature (°C)	Mean annual precipitation (mm)	N deposition ($g\,m^{-2}\,yr^{-1}$)	N deposition class	S deposition ($g\,m^{-2}\,yr^{-1}$)	S deposition class
Nova Scotia	Petite Bog	45°09′	63°56′	6.5	1175	0.34 (0.08)	1	0.41 (0.09)	2
	Western Head	43°41′	65°08′	5.9	1217	0.33 (0.04)	1	0.37 (0.07)	1
	Cape Sable	43°28′	65°36′	5.9	1217	0.33 (0.04)	1	0.37 (0.07)	1
	Fourchu	45°42′	60°14′	5.5	1480	0.29 (0.03)	1	0.35 (0.05)	1
P. E. Island	Foxley Moor	46°43′	64°02′	5.0	1090	0.32 (0.08)	1	0.42 (0.12)	2
New Brunswick	Point Sapin	46°59′	64°51′	4.6	1087	0.32 (0.08)	1	0.42 (0.12)	2
	Point Escuminac	47°04′	64°49′	4.6	1087	0.27 (0.06)	1	0.36 (0.08)	1
	Miscou Island	47°56′	64°30′	4.6	1018	0.28 (0.07)	1	0.38 (0.12)	1
	Savoy Bog	47°47′	64°36′	4.6	1018	0.28 (0.07)	1	0.38 (0.12)	1
Quebec	Lac à la Tortue	46°32′	72°40′	4.6	1042	0.76 (0.14)	3	0.75 (0.17)	3
	Yellow Lake	48°54′	71°54′	1.5	835	0.32 (0.06)	1	0.42 (0.07)	2
	Mirabel	45°41′	74°03′	4.8	1030	0.81 (0.12)	3	0.78 (0.17)	3
	Despinassy Bog	48°44′	77°44′	1.0	913	0.47 (0.10)	2	0.51 (0.13)	2
	Ilets-Jeremie Bog	48°54′	68°49′	1.5	996	0.35 (0.06)	1	0.38 (0.09)	1
	Mai Bog	49°57′	67°02′	1.0	1128	0.33 (0.05)	1	0.43 (0.07)	2
	Port Cartier Bog	50°02′	66°56′	1.0	1128	0.29 (0.05)	1	0.41 (0.07)	2
	Escoumins Bog	48°24′	69°21′	3.0	998	0.36 (0.05)	1	0.42 (0.07)	2
Ontario	Mer Bleue	45°25′	75°31′	5.8	910	0.81 (0.13)	3	0.82 (0.15)	3
	Baker Bog	49°08′	90°45′	1.5	763	0.35 (0.06)	1	0.22 (0.06)	1
	Norembego	48°59′	80°42′	0.6	920	0.52 (0.12)	2	0.56 (0.12)	2
	Nellie Bog	48°46′	80°50′	0.9	793	0.52 (0.12)	2	0.56 (0.12)	2
	Hislop Bog	48°28′	80°23′	1.5	876	0.52 (0.12)	2	0.56 (0.12)	2
	Holtrye Bog	48°32′	80°48′	0.9	793	0.52 (0.12)	2	0.56 (0.12)	2

years) assumes that ^{210}Pb concentrations in peat decrease exponentially with depth, approaching a low constant value taken to represent the supported ^{210}Pb fraction formed within soil as opposed to that deposited from the atmosphere (e.g. Turetsky *et al*., 2000). The constant rate of supply model of Appleby & Oldfield (1978) was applied to calculate the ages of peat layers. The cumulative dry mass of peat on an aerial basis ($g\,m^{-2}$) was calculated as layer thickness weighted averages from the dry bulk density profile and converted into C, N and S, based on C, N and S concentration analyses for surface cores. The accumulated mass of C, N and S above the oldest ^{210}Pb-datable horizon was divided by the age of this horizon to give the RERCA, RERNA and RERSA rates ($g\,m^{-2}\,yr^{-1}$), for the 0–50-, 0–100- and 0–150-year periods.

Atmospheric deposition

Rates of atmospheric wet N and S deposition were calculated using the 7-year mean (1990–1996) results for eastern North America (R. Vet, C.U. Ro, and D. Ord, National Atmospheric Chemistry Database and Analysis Facility, Atmospheric Environment Service, Environment Canada, Ontario, SOE Bulletin No. 99-3), using the station nearest each bog. The bogs were allocated to three deposition classes: class 1, <0.4; class 2, 0.4–0.6; and class 3, $>0.6\,g\,N$ or $S\,m^{-2}\,yr^{-1}$ (Table 1). The climate data (Table 1) were derived from the Canadian Climate Normals (Meteorological Service of Canada, http://www.msc-smc.ec.gc.ca/climate/climate_normals/index_e.cfm), using the station nearest each bog.

To estimate atmospheric wet N and S deposition over the 0–50-, 0–100- and 0–150-year time periods, we used estimates of SO_2 and NO_x emission for these periods from the United States (US Environmental Protection Agency, 2000) and applied these to the observed 1990–1996 deposition values.

Mesocosm experiments

To determine the fate of N and S added to peat profiles, 16 peat cores including intact vegetation, 20 cm diameter and 75 cm length, were extracted in PVC tubes from hollows in late summer to fall and a drainage mesh and cap attached at the bottom. Pore water samplers (7 mm outer diameter, 3 mm inner diameter) were inserted horizontally at 2 cm depth increments. The water table was initially adjusted with distilled water to 2–6 cm below the moss surface. After 60 days, the water table was lowered to about 36 cm in eight of the cores and constant water tables kept for another 220 days. The temperature was initially 22 °C during the day and 8 °C at night and after day 50, 12 °C during the day and 8 °C at night. Relative humidity was maintained at 70% and light intensity was adjusted to $250\,\mu mol\,m^{-2}\,s^{-1}$. Solution was added to the peat cores with a sprinkler 5–6 days a week and water manually retrieved at an average outflow of $1.8\,mm\,day^{-1}$. The inflowing solute contained H_3O^+, sulfate (26 or $104\,mmol\,L^{-1}$), nitrate and ammonium (40 or $120\,\mu mol\,NH_4NO_3\,L^{-1}$) according to a fractional factorial design with two replicates per treatment (Table 2, Box *et al*., 1978). These concentrations were chosen to represent approximate background (assuming 30% dry deposition) and three- to fourfold increased deposition levels than at Mer Bleue. In addition, the inflow contained dissolved calcium ($30\,\mu mol\,L^{-1}$), magnesium ($15\,\mu mol\,L^{-1}$), sodium ($50\,\mu mol\,L^{-1}$), potassium ($5\,\mu mol\,L^{-1}$) and chloride ($150–250\,\mu mol\,L^{-1}$). Sulfate

Table 2 Fractional factorial design matrix of the mesocosm experiment and retention of added nitrogen (N) and sulfur (S)

Treatment				Retention at water table (%)			Retention at outflow (%)		
Location*	WT[†]	N[‡]	S[§]	NO_3^-	NH_4^+	SO_4^{2-}	NO_3^-	NH_4^+	SO_4^{2-}
MB	Low	High	Low	>99	97	0	>99	96	81
MB	Low	Low	High	97	81	16	>98	86	82
MB	High	Low	Low	97	82	76	>98	85	56
MB	High	High	High	>99	97	89	>99	91	97
ELA	Low	Low	Low	96	88	44	>98	58	21
ELA	Low	High	High	>99	90	69	>99	85	85
ELA	High	High	Low	>99	97	77	>99	97	21
ELA	High	Low	High	>98	60	94	>98	53	97

*MB, Mer Bleue; ELA, Experimental Lakes Area 239.
[†]Water table (WT) – high 2 cm and low 35 cm below surface.
[‡]N additions – low 1.12 and high $3.36\,g\,N\,m^{-2}\,yr^{-1}$ as NH_4NO_3.
[§]S additions – low 0.83 and high $3.33\,g\,m^{-2}\,yr^{-1}$ as SO_4^{2-}.

was determined by ion chromatography (Dionex, Metrosep Anion Dual 1, (Dionex Corporation, Sunnyvale, CA, USA) at 0.5 mL min^{-1} flow rate and chemical suppression). Dissolved inorganic nitrogen (DIN) was measured as NH_4^+ and $NO_2^- + NO_3^-$ using colorimetric methods on a Latchat FIA$^+$ 8000 series continuous flow auto-analyzer (Latchat Instruments, Milwaukee, WI, USA). Nutrient retention was calculated by subtracting the mass flux of an element at the water table and outflow, respectively, from the mass flux through deposition into the mesocosm. The vegetation grew during the experiment, with estimated rates of photosynthesis equal to or larger than ca. 200 g C m^{-2} yr^{-1} (Blodau *et al.*, 2004).

Results

Concentrations of peat N ranged from 0.35% to 2.25% with an average of 0.80% and standard deviation of 0.35%, and S concentrations ranged from 0.07% to 0.55% with an average of 0.18% and standard deviation of 0.09%. There was a weak but significant overall correlation between peat N and S concentration ($r^2 = 0.19$, $n = 466$, $P < 0.001$). Ratios of C : N, C : S and N : S averaged 65 : 1, 318 : 1 and 5 : 1, respectively. In general, concentrations of N and S increased with depth in the peat profiles, as illustrated by hummock and hollow examples from the three deposition classes (Fig. 1), with a pronounced increase in S concentration in the basal samples collected from near the water table.

Using N and S concentration and dry bulk density results and ^{210}Pb dating, the cumulative mass of C, N and S in the profiles was estimated for the period up to 200 years ago. For each of the three N and S deposition zones, average values were 11.5, 14.5 and 16.3 kg C m^{-2}, 210, 360 and 450 g N m^{-2} and 55, 58 and 110 g S m^{-2}; the accumulation increased with an increase in deposition by zone (Fig. 2).

Rates of N and S accumulation over the 50-, 100- and 150-year periods averaged 1.86, 1.65 and 1.46 g N m^{-2} yr^{-1}, and 0.38, 0.37 and 0.32 g S m^{-2} yr^{-1}, respectively (Fig. 3). There was, however, considerable variation among sites for the rates for each time period, with coefficients of variation averaging 47% for N and 43% for S.

There was a strong relationship between C accumulation over the 50-, 100- and 150-year periods with the accumulation of N ($r^2 = 0.65$–0.68, $P < 0.001$, Fig. 4). Positive but weaker relationships ($r^2 = 0.40$–0.44, $P < 0.001$) also existed between C and S accumulation over these periods. In both cases, the position of the 50-year regression line between C and N or S accumulation rates was lower than for the other time periods, indicating a higher C : N or C : S ratio.

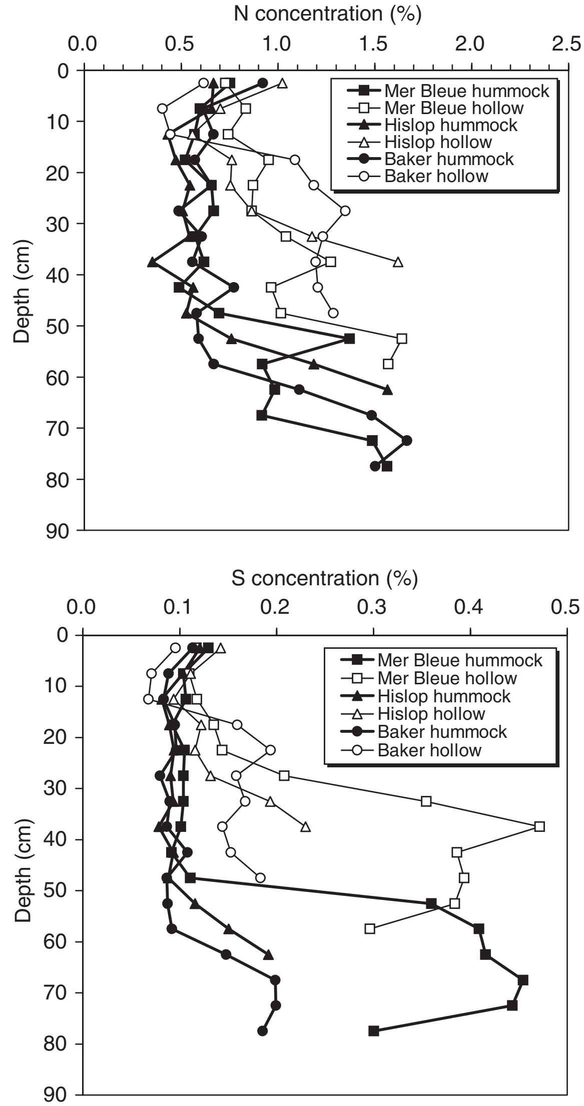

Fig. 1 Concentrations of nitrogen (N) and sulfur (S) in hummock and hollow profiles from sites representing deposition classes 1 (Baker), 2 (Hislop) and 3 (Mer Bleue).

In nearly all cases, the 50-year RERCA rates in hummocks were larger than those in hollows, the difference ranging from −40 to +75 g C m^{-2} yr^{-1} (Fig. 5). There was a relatively small differentiation in the RERSA between hummocks and hollows, generally less than 0.2 g S m^{-2} yr^{-1}, with an increase as the difference in hummock–hollow RERCA increased. Differentiation between hummock and hollow N accumulation (RERNA) was larger, ranging from −0.9 to +1.5 g N m^{-2} yr^{-1}, with a strong relationship of increasing N differentiation with increasing C differentiation.

We examined the influence of atmospheric N and S deposition on RERNA and RERSA by comparing annual wet deposition rates averaged over the period

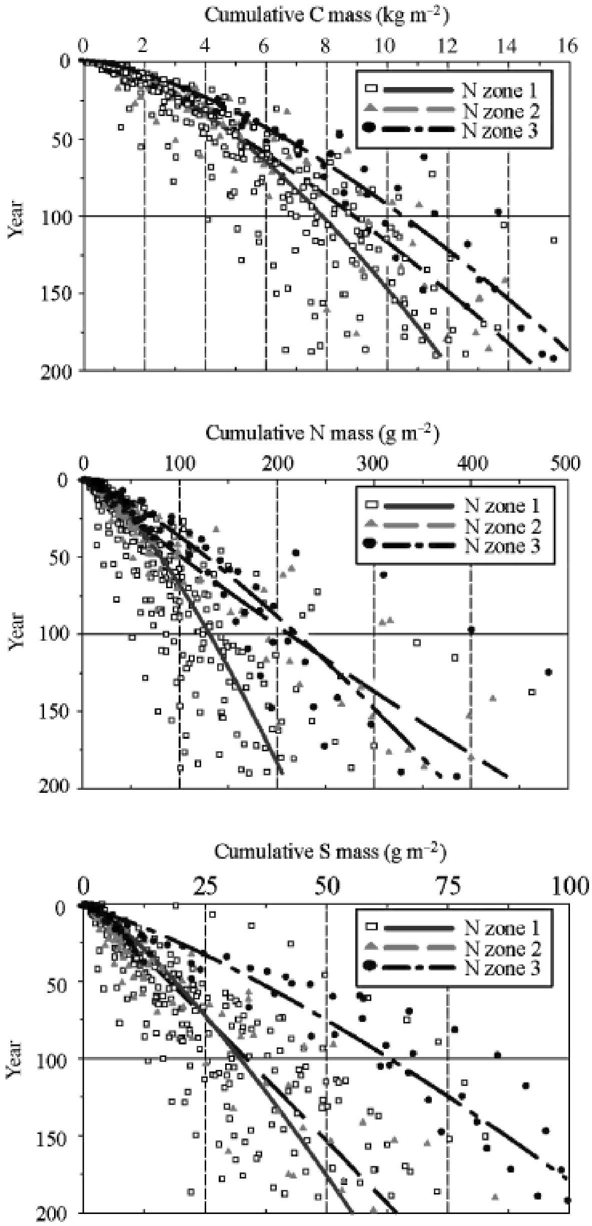

Fig. 2 Cumulative mass : age relationships for carbon (C), nitrogen (N) and sulfur (S) and the average relationship for profiles in the three N or S deposition zones.

1990–1996 for the sites with the 50-year RERNA and RERSA (Fig. 6). There was a modest but significant ($r^2 = 0.30$, $P < 0.001$) positive correlation between RERNA and N deposition, and accumulation rates were about four to five times that of wet deposition. A weaker, but still significant correlation ($r^2 = 0.21$, $P < 0.001$) was observed between RERSA and S deposition, with about 75% of the deposited S accumulating in the peat (Fig. 6). Many profiles showed substantially lower S accumulation rates than the estimated atmospheric S deposition rate. Generally, there was a large variation in RERNA and RERSA at sites exposed to low rates of atmospheric N and S deposition. The small number of sites sampled in areas with high rates of atmospheric N and S deposition limits the strength of the relationship between deposition and accumulation.

Based on estimated emissions of SO_2 and NO_x from the United States (US Environmental Protection Agency, 2000), we calculated that the overall N and S wet deposition rates for the 50-, 100- and 150-year periods would be 83%, 53% and 38% for N and 108%, 92% and 71% for S of the observed 1990–1996 wet deposition rates. Using these estimated deposition corrections applied equally to all the sites' 1990–1996 deposition rates, there was a significant correlation between deposition and accumulation over the 0–50-, 0–100- and 0–150-year periods, with the best-fit regressions for the longer periods being above those for the 50-year period (Fig. 7).

In the mesocosm experiments, nitrate was nearly completely retained (> 97%) above the water table in all treatments (Table 2) and below the moss canopy, concentrations were always $< 3\,\mu mol\,L^{-1}$. Ammonium was retained as well, although to a lesser extent (81–97%) and below the water table ammonium was on average released (Table 2). Between 75% and 96% of the added sulfate was retained by the unsaturated vegetation cover when the water table was close to the peat surface (Table 2). Sulfate was generally also retained below the water table (Table 2), probably because of sulfate reduction, although cases of net release occurred. In treatments with a low water table, the retention of sulfate by the moss layer was reduced from 0% to 69% (Table 2). In these treatments, sulfate concentrations only decreased to ca. $10–30\,\mu mol\,L^{-1}$ below the water table (Fig. 8) and as outflow concentrations were similar, the retention was larger in the low S treatment (Table 2). The significantly negative correlation between sulfate concentrations at the water table and rates of photosynthesis determined simultaneously ($r^2 = 0.53$, $P < 0.05$; Blodau *et al.*, 2004) suggested that the uptake of sulfate was related to the photosynthetic activity of the plant cover. Estimated average rates of photosynthesis decreased from 48 to $27\,mmol\,m^{-2}\,day^{-1}$ under high and low water table treatments (Blodau *et al.*, 2004).

Discussion

N and S cycling in peatlands is dominated by organic forms and transformations. Atmospheric deposition of N is primarily in inorganic forms, ammonium (NH_4^+) and nitrate (NO_3^-), which can be rapidly and efficiently absorbed by the moss layer (Li & Vitt, 1997; Aldous,

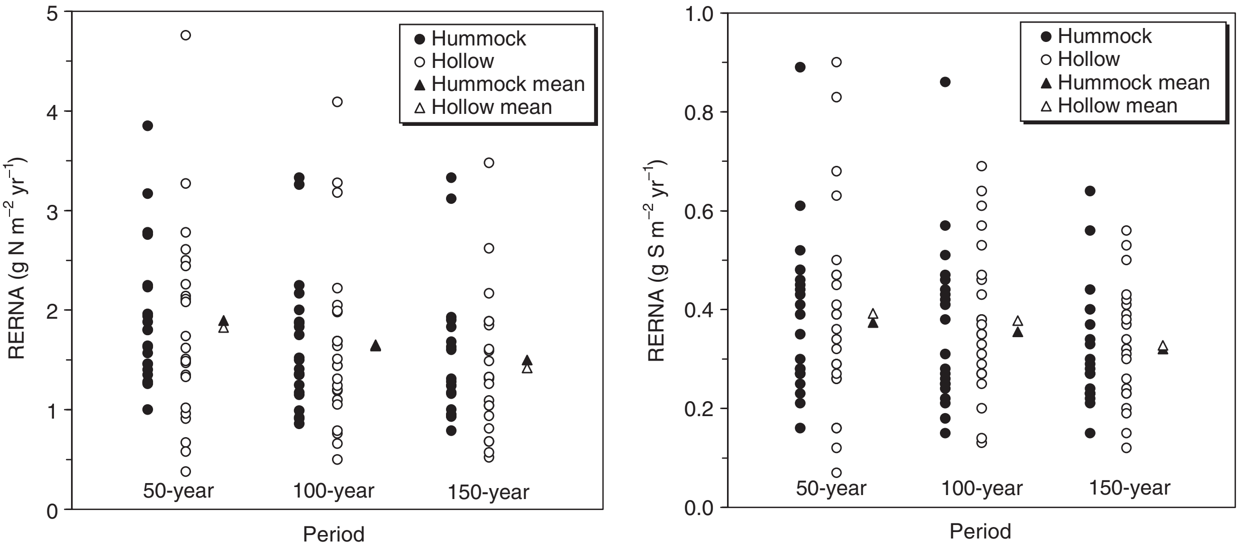

Fig. 3 Rates of nitrogen (N) (RERNA) and sulfur (S) accumulation (RERSA) in peat profiles over the 50-, 100- and 150-year periods.

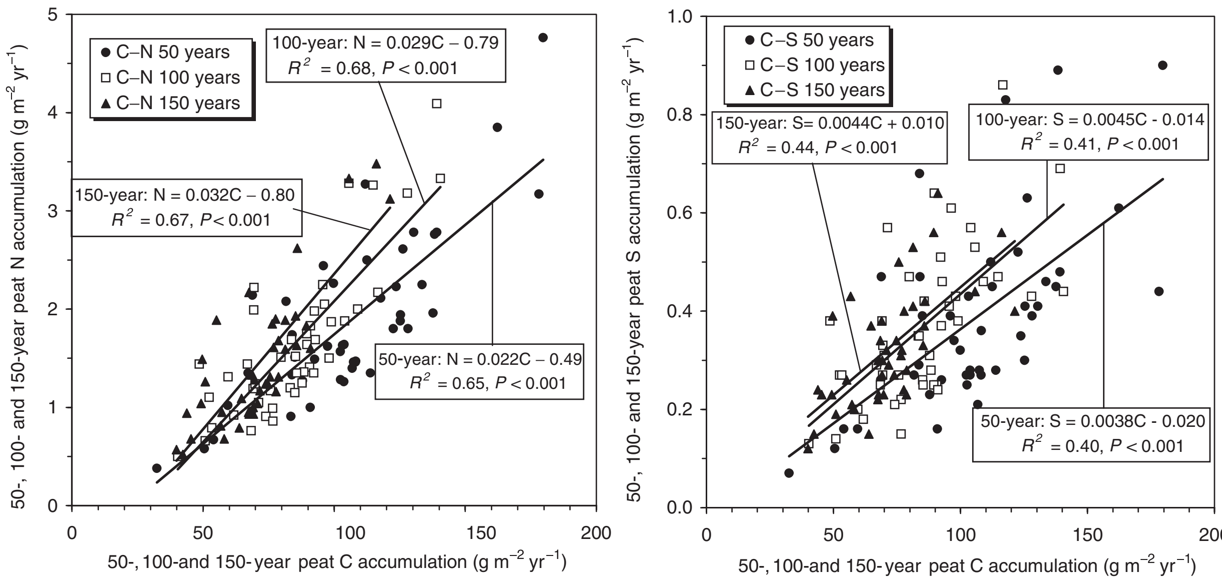

Fig. 4 Relationship between rates of nitrogen (N) (RERNA) and sulfur (S) accumulation (RERSA) and C accumulation (RERCA) in peat profiles over 50-, 100- and 150-year periods.

2002a, b; Nordbakken *et al.*, 2003) and this was confirmed by the results of our mesocosm experiments. Bog porewater usually contains small concentrations of NH_4^+, almost no NO_3^- and the dominant soluble form is dissolved organic N (DON): at the Mer Bleue bog, porewater concentrations averaged 0.47, 0.04 and 1.88 mg L^{-1} of NH_4^+–N, NO_3^-–N and DON, respectively (Basiliko & Moore, personal communication). Peatland plants may be able to take up organic forms of N (Nordbakken *et al.*, 2003). The absence of porewater NO_3^- and strongly reducing conditions in the upper part of bog profiles mean that rates of denitrification are small (e.g. Regina *et al.*, 1996).

Although S inputs from the atmosphere are dominantly in the form of sulfate (SO_4^{2-}), additions of SO_4^{2-} to peat porewater result in a rapid disappearance of SO_4^{2-} (e.g. Bayley *et al.*, 1986) and in bog profiles, most of the S is found in organic forms (e.g. Wieder & Lang, 1986, 1988; Urban *et al.*, 1989; Novák *et al.*, 1994; Vile *et al.*, 2003). In ombrotrophic bog profiles, the accumulation of organic matter over the past 100 years occurs primarily within the acrotelm, the generally oxidized surface peat layer above the water table, so that reduction–oxidation reactions that are important in overall S biogeochemistry in wetlands (e.g. Urban *et al.*, 1989; Mandernack *et al.*, 2000; Blodau *et al.*, 2002) may play a minor role in the results that we present here, although there may be anaerobic microsites within the acrotelm (Vile *et al.*, 2003).

Our results show that significant accumulation of N and S has occurred over the past 150 years in bogs in eastern Canada. The range of N accumulation rates is large (0.5–4.8 $g\,N\,m^{-2}\,yr^{-1}$) over the 50–150-year periods but the average (1.7 $g\,N\,m^{-2}\,yr^{-1}$) is similar to the 2 $g\,N\,m^{-2}\,yr^{-1}$ over 20–85 years in a Minnesota bog reported by Urban & Eisenreich (1988) and similar to values reported by Malmer & Holm (1984) in Sweden. In Norway, Ohlson & Økland (1998) reported a similar large spatial variation of C (20–100 $g\,C\,m^{-2}\,yr^{-1}$) and N (0.6–2.1 $g\,N\,m^{-2}\,yr^{-1}$) accumulation in a bog over the last 125 years. In northern Alberta, Turetsky *et al.* (2000) observed N accumulation rates of 0.7 and 0.8 $g\,N\,m^{-2}\,yr^{-1}$ in two bogs. N accumulation rates were generally larger in hummocks than hollows, following the pattern of C accumulation.

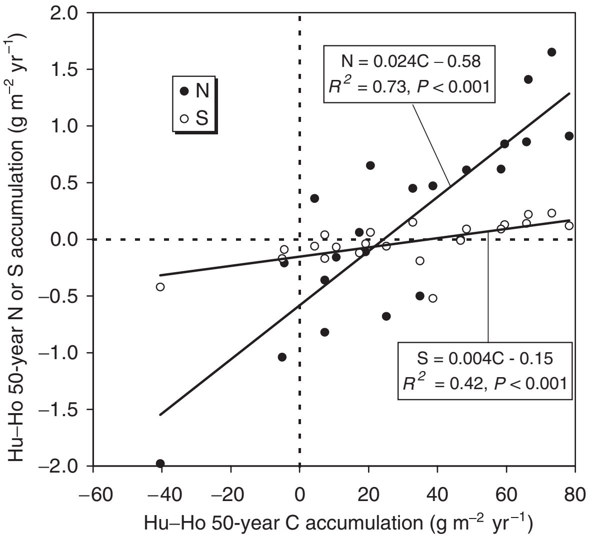

Fig. 5 The difference between hummock and hollow accumulation of nitrogen (N) and sulfur (S) (RERNA and RERSA) and C (RERCA) over the past 50 years.

A fraction of the N accumulation can be explained by retention of deposited inorganic N. The results of the mesocosm experiments suggest that during the growing season inorganic N can be fully retained at short-term deposition rates of up to 4.7 $g\,N\,m^{-2}\,yr^{-1}$. N saturation, including export of mineral N, has recently been proposed to occur in peatlands that receive more than ca. 1.8 $g\,N\,m^{-2}\,yr^{-1}$ in the long term (Lamers *et al.*, 2000), similar to the largest estimated total deposition rate along the Canadian transect. This proposal was based on a simple mass balance consideration of annual biomass synthesis, N deposition level and C/N ratios of 40 : 1 to 50 : 1 occurring in *Sphagnum* and is to some extent supported by other experimental studies (Williams *et al.*, 1999). At Mer Bleue, at the high deposition of the Canadian transect, the concentration of N in the capitulum of *S. capillifolium* (hummock) and *S. magellanicum* (hollow) averaged 0.83% and 0.86%, respectively, in line with observations of N deposition and *Sphagnum* N collated by Lamers *et al.* (2000). Based on studies in Alberta, Vitt *et al.* (2003) suggested that the growth of *S. fuscum* increased with increasing atmospheric N deposition, reaching a critical total deposition value of 1.4–3.4 $g\,N\,m^{-2}\,yr^{-1}$.

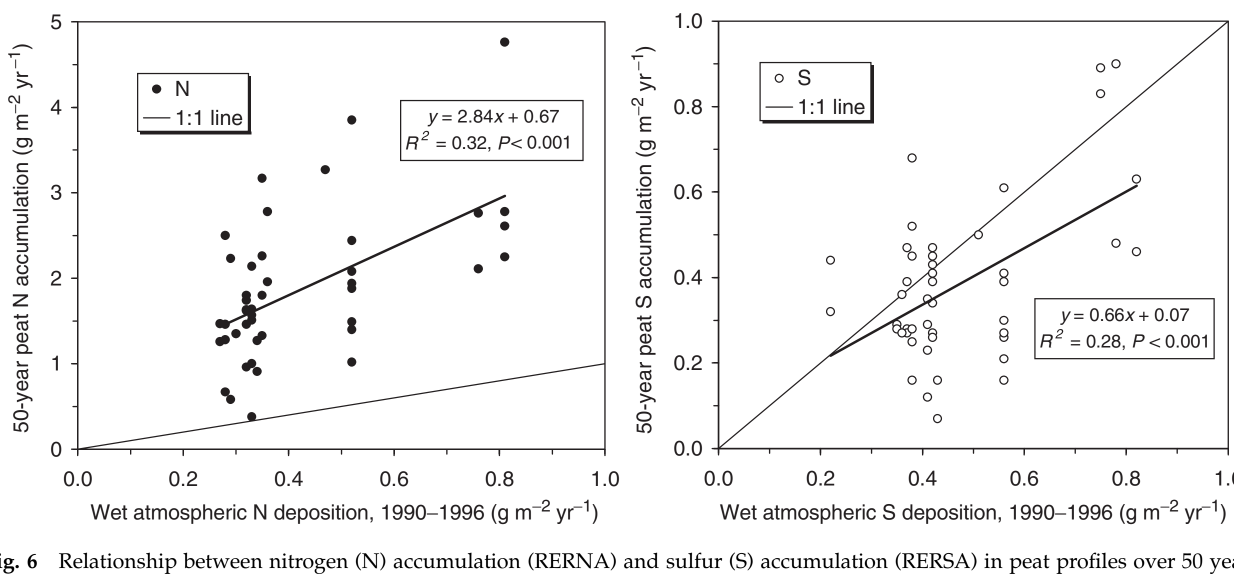

Fig. 6 Relationship between nitrogen (N) accumulation (RERNA) and sulfur (S) accumulation (RERSA) in peat profiles over 50 years and atmospheric wet deposition of N and S between 1990 and 1996.

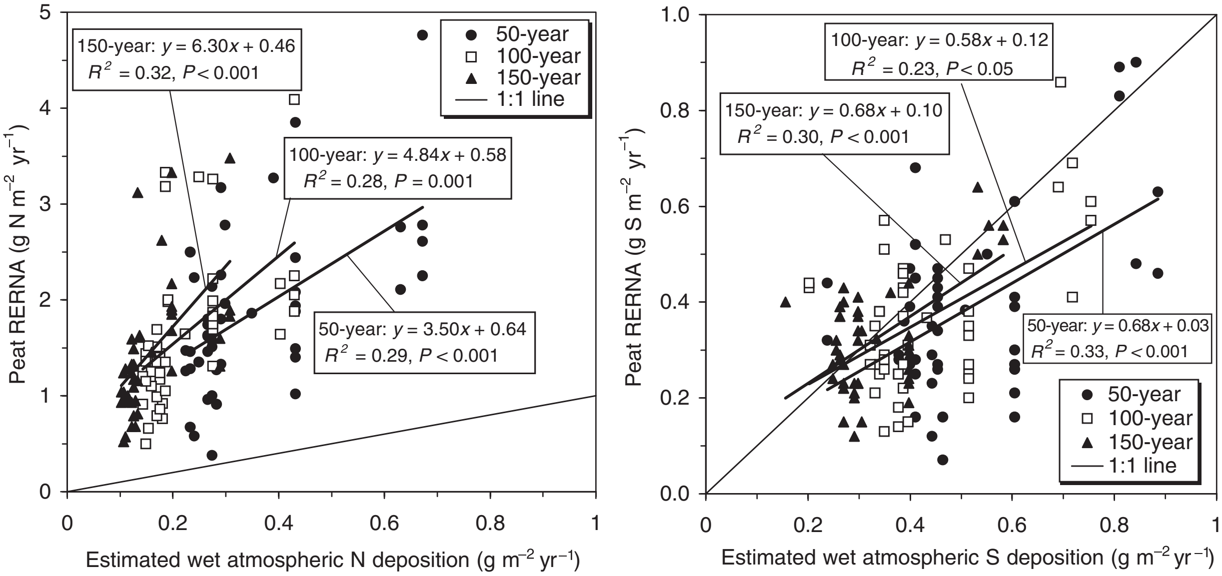

Fig. 7 Relationship between nitrogen (N) accumulation (RERNA) and sulfur (S) accumulation (RERSA) in peat profiles over 50-, 100- and 150-year periods and estimated atmospheric wet deposition of N and S over those periods.

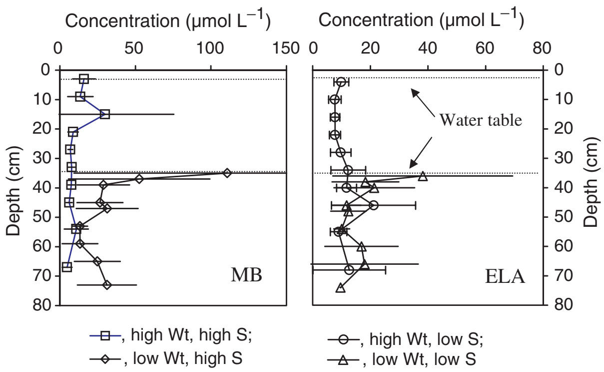

Fig. 8 Pore water concentrations of SO_4^{2-} in the mesocosm treatments for the Mer Bleue (MB) (left) and Experimental Lakes Area (ELA) (right) peat cores. Corrected for the evapotranspiration from the mesocosms, inflow concentrations were about 75 and 220 μmol L^{-1} NH_4NO_3 and 45 and 190 μmol L^{-1} SO_4^{2-}. Bars indicate standard deviations.

On average, N accumulation in the bog cores of this study is about four times that in wet atmospheric deposition. Other sources of N therefore have to be considered. These include organic N deposition, dry deposition and N_2 fixation. DON forms a variable proportion of atmospheric N deposition and may increase the overall deposition by about 50% above the wet deposition (Neff *et al*., 2002; Cornell *et al*., 2003). Dry deposition is difficult to measure and estimate and may be smaller in bog systems than forests, because of the small leaf surface area: at Mer Bleue, for example, the vascular leaf area index is 1.3 (Moore *et al*., 2002). Thus, contemporary total N atmospheric deposition may be 75–100% larger than wet deposition, amounting to 0.4–1.6 g N m^{-2} yr^{-1} at these bog sites in eastern Canada. This range is close to the critical N deposition value of about 1.5 g m^{-2} yr^{-1} for *Sphagnum* growth and N retention cited by Lamers *et al*. (2000) and Vitt *et al*. (2003).

Annual N_2 fixation estimates in ombrotrophic bogs range from 0.1 and 1.0 g N m^{-2} yr^{-1} in Minnesota and Massachusetts, respectively (Chapman & Hemond, 1982; Hemond, 1983; Urban & Eisenreich, 1988). Measurements have been infrequent and sparse and rates may be underestimated, as has been recently found in boreal forests (DeLuca *et al*., 2002).

Losses of N from the peat may arise from denitrification, although rates are small (<0.2 g N m^{-2} yr^{-1}) in acidic bogs with a generally low water table (Hemond, 1983; Urban & Eisenreich, 1988; Regina *et al*., 1996). Aquatic export of N occurs as NH_4^+, NO_3^-, DON and particulate N. Losses of NH_4^+ may account for 0.1–0.2 g N m^{-2} yr^{-1} and NO_3^- losses are small, probably <0.05 g N m^{-2} yr^{-1} (Hemond, 1983; Urban & Eisenreich, 1988). Export of DON may be larger, between 0.1 and 0.3 g N m^{-2} yr^{-1}, based on DOC export of 5–10 g C m^{-2} yr^{-1} and DOC : DON ratios of 30 : 1 to 50 : 1 (Hemond, 1983; Urban & Eisenreich, 1988; Fraser *et al*., 2001).

Thus, the bogs along our transect receive 0.4–1.6 g N m^{-2} yr^{-1} in precipitation and 0.1–1.0 g N m^{-2} yr^{-1} in N_2 fixation, and lose through denitrification (<0.2 g N m^{-2} yr^{-1}) and aquatic export (0.2–0.5 g N m^{-2} yr^{-1}). These net gains, ranging from 0 to 2.3 g N m^{-2} yr^{-1}, are similar to the N accumulation rates over the 50–150-year periods, which average 1.5–2.0 g N m^{-2} yr^{-1}. There was a correlation between increasing atmospheric N deposition and N accumulation rates, although there is great variability among sites with similar N deposition. In part, this variation may be related to the hummock–hollow microtopography: hummocks had larger C and N accumulation rates than hollows along our transect (Fig. 5). In a Norwegian bog, Ohlson & Økland (1988) recorded coefficients of variation of 56% and 57% for recent C and N accumulation rates in peat samples taken within a 20 m × 20 m area. This variability may be associated with higher rates of plant production and N fixation in hummocks, whereas atmospheric N deposition rates are unlikely to be very different between hummocks and hollows. Leaf biomass in bog hummocks averaged 156 g m^{-2}, while the average for hollows was 106 g m^{-2}, although *Sphagnum* capitulum mass was similar at 176 and 172 g m^{-2} (Turunen *et al.*, 2004). In Alberta, Vitt *et al.* (2003) noted that production of *S. fuscum* increased with increased atmospheric N deposition, within the range encountered in eastern Canada, without a major increase in N concentration in the underlying peat. Although there are changes in climate, particularly mean annual temperature (Table 1) and growing season, along the transect, it appears that the primary cause of increased N accumulation in the peat cores is increased atmospheric N deposition.

The range of S accumulation rates in the eastern Canadian bogs (0.1–0.9 g S m^{-2} yr^{-1}) encompassed that found in other wetlands noted earlier (Urban *et al.*, 1989; Novák *et al.*, 1994; Turetsky *et al.*, 2000). Our results suggest that, on average, the bogs retained about 75–90% of the recent wet atmospheric deposition of S in the acrotelm. Inputs of dissolved organic S (DOS) are probably insignificant (Park *et al.*, 2003), but dry deposition of SO_4^{2-} may increase total S deposition by up to 50%. There can be substantial S losses (0.1–0.3 g m^{-2} yr^{-1}) from wetland catchments, as both SO_4^{2-} and DOS (Devito *et al.*, 1989; Urban *et al.*, 1989; Devito, 1995; Gorham *et al.*, 1998). Gaseous losses of H_2S are probably negligible since concentrations in the pore water of bogs seem to remain in the lower micromolar range (Steinmann & Shotyk, 1997; Blodau *et al.*, 2002). The retention rate observed in these eastern Canadian bogs is similar to that noted by Hemond (1983) at Thoreau's bog (77%) and by Urban *et al.* (1989) at bogs in Minnesota and north-western Ontario (58% and 91%, respectively).

Our estimates of S retention rates were also in agreement with the results of the mesocosm experiment, which showed similar retention efficiencies with the exception of combined low water tables and high deposition. In contrast to NO_3^-, SO_4^{2-} concentrations in the pore water did not decrease below 10–30 μmol L^{-1}. This concentration may be because of either the release of sulfates from the peat matrix (Mandernack *et al.*, 2000) or kinetic limits to microbial utilization of SO_4^{2-} and was responsible for the lower S retention rates in the mesocosm experiments. The results of both transect and mesocosm approach, nevertheless, suggest that North American bogs are sinks for S. This sink function may be lost under more severe S pollution in parts of Europe and the Appalachian mountain range, where SO_4^{2-} has been found in peat porewater at concentrations of 50–300 μmol L^{-1} (Wieder & Lang, 1988; Wieder *et al.*, 1990; Nedwell & Watson, 1995; Watson & Nedwell, 1998). Large continuous SO_4^{2-} export in streams was also reported from a central European peatland (Hruska *et al.*, 1996). The retention capacity for S has also been partially lost in North American swamps affected by acidic precipitation and large input rates of N and S from upland ecosystems (LaZerte, 1993).

The maintenance of the stoichiometric relationship between C and N in these bogs (Fig. 4) suggests that elevated rates of atmospheric deposition of N have led to increased rates of C accumulation. Turunen *et al.* (2004) observed a significant relationship (partial regression, $r^2 = 0.28$ and 0.38 for hummocks and hollows, Fig. 3) between wet atmospheric N deposition during 1990–1996 and 50-year C accumulation, along this Canadian bog transect. As noted above, this may arise from N being the major nutrient limiting plant productivity in boreal bogs, with increased *Sphagnum* production associated with N deposition rates encountered here (e.g. Malmer, 1990; Aerts *et al.*, 1992; Berendse *et al.*, 2001; Vitt *et al.*, 2003). The increased C accumulation rate with elevated N deposition may also arise from a slowing in the rate of organic matter decomposition, as observed in European forests by Berg & Matzner (1997) and slower heterotrophic respiration in the uppermost 5 cm of the peat profile (Williams & Silcock, 1997).

Conclusions

Ombrotrophic bogs in eastern Canada have accumulated N and S over the past 150 years at rates of 0.5–4.8 g N m^{-2} yr^{-1} and 0.1–0.9 g S m^{-2} yr^{-1}. There was a high degree of spatial variability in accumulation rates, particularly between hummocks and hollows. The N and S accumulation rates were correlated with recent rates of atmospheric N and S deposition over the range 0.3–0.8 g N m^{-2} yr^{-1} and 0.2–1.0 g S m^{-2} yr^{-1}. The ratio between accumulation and wet atmospheric deposition averaged about 4 : 1 and 0.75 : 1 for N and S, respectively. The results confirm that in eastern Canadian bogs, the plant canopy and upper peat control the subsurface dynamics of S and N through their filter function, converting mineral N and S into organic forms.

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