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Episodic sulphate export from wetlands in acidified headwater catchments: prediction at the landscape scale

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Abstract. Sulphate (SO_4^{-2}) concentrations in 34 intensively measured Canadian Shield streams near the Dorset Research Centre, central Ontario, were used to test a hydrogeologic model that uses simple measures of wetland area and till depth to identify catchments that produce SO_4^{-2} pulses. Mean annual measured maximum SO_4^{-2} concentrations were significantly greater in shallow till (<1 m depth) catchments containing wetlands than catchments covered with deeper tills (>1 m depth) containing wetlands or catchments with no wetlands. Average maximum SO_4^{-2} concentrations in wetland catchments during years with dry summers were >20 mg/L in 19 of 20 catchments with average till depths of <1 m, whereas concentrations were <20 mg/L in 5 of 6 watersheds with average till depths of >1 m. Peaks in mean annual maximum SO_4^{-2} concentrations from wetland catchments with shallow till occurred during summers with rain fall 150–200 mm less than potential evaporation estimates. There were no significant differences in mean average annual SO_4^{-2} concentration among the different catchments during wet summers, with SO_4^{-2} concentrations ranging from 6 to 13 mg/L. These observations suggest that a large portion of the temporal and spatial variation in SO_4^{-2} chemistry and export can be predicted in headwater catchments of the Canadian Shield and perhaps in other landscapes where till depth influences upland-wetland hydrologic connections.

Introduction

Valley bottom wetlands represent critical interfaces between uplands and streams and influence sulfate (SO_4^{-2}) dynamics in headwater landscapes. Many wetlands which receive high SO_4^{-2} inputs in acid precipitation have a large capacity to retain SO_4^{-2}, but substantial oxidation of reduced sulfate

(S) may occur during water table drawdowns (van Haesebroeck et al. 1997). Downstream acidification caused by episodic SO_4^{-2} release from wetlands following droughts has been reported (Braekke 1981; Wieder 1985; Bayley et al. 1986; Van Dam 1988) and acidification of surface waters has been linked to fish kills (Gosling & Baker 1980; Holopainen & Oikari 1992) and more recently to water clarity and biologically effective UV-B penetration (Schindler et al. 1996; Yan et al. 1996). Increased SO_4^{-2} release following dry summers can change annual wetland budgets from a pattern of retention to export (LaZerte 1993; Devito 1995) and may contribute to the persistence of downstream acidification despite reductions in SO_4^{-2} deposition in catchments (Dillon & LaZerte 1992; Wright & Schindler 1995; Dillon et al. 1997). Furthermore, SO_4^{-2} pulses and resulting acidification may become more frequent and widespread if the climate warms and the frequency of drought in eastern temperate North America hold (Manabe & Wetheralds 1986). Increasing our understanding of the controls of SO_4^{-2} pulses and predicting which landscapes are susceptible to prolonged acidification from atmospheric input is necessary to adequately evaluate the success of recent reductions in S emissions and direct policy in future amendments to S emission control programmes in eastern North America. Current knowledge is inadequate for estimating how widespread this SO_4^{-2} pulse phenomenon is or predicting which wetlands are susceptible to water table drawdown and episodic SO_4^{-2} release following drought.

The rate and magnitude of water flow to wetlands and the maintenance of surface saturation are key to temporal and spatial variations in peat anoxia (Sparling 1966; Devito & Dillon 1993), which regulates cyclic reduction and oxidation and long term S accumulation in wetland ecosystems (Brown & MacQueen 1985; Wieder & Lang 1988, Urban et al. 1989; Feijtel et al. 1989). A landscape framework, which examines wetland biogeochemistry in the context of catchment hydrology, is needed to develop generalizations about the biogeochemical function of a wetland so that knowledge can be transferred from studied sites to other wetlands.

The hydrology of headwater catchments in the shallow till-rock ridges physiographic region of central Ontario is characterized as seasonal groundwater in local flow regimes due to shallow till and impervious bedrock (Winter & Woo 1990). Large regions of the Canadian Shield and other areas in North America and northern Europe have similar characteristics. Small valley bottom wetlands are common features in these landscapes and are likely to experience large water table draw down and autumn SO_4^{-2} pulses following dry summers, particularly in areas of acidic deposition. A recent study of several headwater catchments that represent the range of till depths typical of the southern Canadian Shield has shown that small increases in till depth from <1 m to 1–3 m result in a shift from discontinuous to continuous upland-wetland groundwater connections and contrasting patterns of wetland water table fluctuations during dry summers (Devito 1994; Devito et al. 1996). During years with low summer precipitation large water table draw down and increased SO_4^{-2} mobilization and export occurred in the conifer swamps located in catchments with shallow till (<1 m), but the peat remained water saturated and SO_4^{-2} immobilization and retention occurred during the same period in the conifer swamps located in catchments covered with deeper till (>1 m) (LaZerte 1993; Devito 1994, 1995; Devito & Hill 1997, 1998). The conceptual model, developed on the basis of these sites, assumes that simple measures of till depth integrate primary controllers of the upland wetland-groundwater linkage and can be used to estimate relative differences in the seasonal amplitude and duration of water table fluctuation (Devito et al. 1996, Devito & Hill 1997). The water table elevation in turn controls peat anoxia, water flow path and thus patterns of SO_4^{-2} transport and transformation in the valley bottom wetlands (Sparling 1966; Brown & McQueen 1985; Devito & Dillon 1993; Devito & Hill 1997, 1998).

This paper tests the predictive value of the hydrogeologic conceptual model to explain the spatial and temporal variation in SO_4^{-2} dynamics in catchments of the shallow till physiographic region characteristic of the southern Canadian Shield landscape. Predicting the occurrence of episodic SO_4^{-2} pulse following dry summers is emphasized. This is done by relating the percent wetland area and the proportion of established categories of till depth (Chapman 1975) to annual minimum and maximum SO_4^{-2} concentrations collected from 34 intensively monitored headwater streams near the Dorset Research Centre, Ontario.

Study site

The 34 headwater streams are located within 50 km of Dorset, Ontario (lat. 45° N, long. 78° W). The two wetland catchments used to develop the conceptual model (Devito et al. 1996; Devito & Hill 1997) were included in this analyses. The catchments vary in size from 1–191 ha with till depths ranging from <1 to >10 m (Table 1).

The physiography, geology and hydrological and geo-chemical studies of Harp and Plastic Lake catchments have been reported in Devito (1994, 1995), Devito et al. (1996), Devito and Hill (1997, 1998) and for the other headwater catchments in Dillon et al. (1991), Girard et al. (1985) and Jeffries and Snyder (1983). The headwater catchments are underlain by effectively impermeable Precambrian metamorphic silicate bedrock covered with a thin basal till. Due to the shallow till, only local groundwater aquifers develop and the hydrology

Table 1. Some physiographic characteristics and stream sampling periods of the headwater catchments

Lake inflow	Catchment area (ha)	% of catchment area		Years of sampling
		Wetland	Minor till (>1 m)	
Blue Chalk 1	20	0	94	13, 1980–92
Chub 1	60	3	24	13
Chub 2	126	8	17	13
Dickie 5	30	25	0	13
Dickie 6	22	22	0	13
Dickie 8	67	8	14	13
Dickie 10	79	17	0	13
Dickie 11	17	21	0	13
Harp 3	26	13	60	13
Harp 3A	20	3	97	13
Harp 4-03	15.5	2	88	13
-08	7.2	0	50	11, 1982–92
-13	62	19	45	11
-14	3.7	0	54	11
-18	38	19	32	11
-21	4.1	0	100	10, 1983–92
-23	23	7	63	7, 82–86, 90–92
Harp 5	191	13	35	13, 1980–92
Harp 6	10	10	34	13
Harp 6A	15	8.5	7	13
Heney 1	29	2	16	8, 1985–92
Heney 2	14	18	19	8
Heney 3	–	0	0	7, 1985–91
Heney 4	–	0	0	7
Plastic 1-03	21	13	11	11, 1980–1991
-07	3.1	13	0	11
-08	3.5	0	0	11
Plastic 2	4	6	16	12, 1980-91
Plastic 2A	1	0	50	12
Plastic 4	4.8	5	95	13, 1980–92
Plastic 5	5.2	11	5	12, 1981–92
Plastic 6	12	13	35	13, 1980–92
Red Chalk 2	27	13	0	13
Red Chalk 3	70	15	82	13

varies considerably over a year (Devito et al. 1996). The surficial geology of the area ranges from exposed bedrock, thin till-rock ridges with depths <1 m to minor till plains 1–10 m plus in depth. These are representative of the shallow till-rock ridges physiographic region of central Ontario (Chapman 1975).

The wetlands often occupy central bedrock depression and valley bottoms. Conifer, mixed and thicket swamps represent the dominant peatland type in this region (Riley 1988). Peaty, humic mesisols and cumula humisols in excesses of 5 m depth often overlay layers of gyttja and pockets of silt, clay, sand and gravel (Devito 1994; Bunting et al. 1996).

Methods

For the general landscape analysis, the stream chemistry from 34 head-water inflows and tributaries to 7 lakes were used (Table 1). Catchments in which beaver ponds were the dominant wetland type were excluded from this analysis. Water was sampled approximately weekly or biweekly from 1980 to 1992 and analysed for SO_4^{-2} as described by OME (1984). For most sites the water chemistry has been monitored for 13 years, but for all sites the SO_4^{-2} chemistry was monitored for a minimum of seven years which included at least four dry summers and two summers with above average summer precipitation (Table 1).

To observe the different response in SO_4^{-2} dynamics, data for each stream sampling site were split into years with dry summers and wet summers. The 13 year (1980–1992) mean (±SD) for June–September rainfall at the Dorset meteorologic station was 340±96 mm. From 1980 to 1992 there were 5 years in which the summer precipitation was <300 mm (1983 and 87–90) and 5 years with summer precipitation of >350 mm (1980, 82, 85, 86 and 92). The minimum and maximum SO_4^{-2} concentration measured in each year were used to determine the average minimum and maximum concentration of the 5 dry summer and wet summer years (see Table 1). Wetland and catchment areas and surficial geology data were obtained for each stream sampling location (Table 1). Surficial geology was determined from local surficial geology maps (1:10,000 scale) and maps in Jeffries & Snyder (1983) and Girard et al. (1985). These data were checked against soil pit information in some catchments. Wetland and pond areas were obtained from air photos and ground surveys.

Given the general nature of till depth data that is available at the regional scale for areas such as the Precambrian Shield, a simple approach was used to classify catchments. Catchment were categorized as: 1) predominantly deep till if >50% of the catchment was covered with minor till (>1 m depth), and

2) predominantly shallow till if >50% of the catchment was covered with thin till-rock ridges (<1 m depth). Differences in mean annual maximum and minimum stream SO_4^{-2} concentrations among catchment categories during dry and wet summers were analysed using multi-factor ANOVA performed on Data Desk, version 5.0 (Velleman 1995).

Results

Sulphate dynamics, wetland area and till depth

In years with dry summers, the average maximum SO_4^{-2} concentrations where significantly greater in streams draining wetland catchments with shallow till than the other three catchment types (Figure 1, Table 2). 19 of 20 streams draining watersheds with shallow till (<1 m) and >2% wetland area had mean annual measured maximum SO_4^{-2} concentrations >20 mg/L during years with dry summers (Figure 1a). In contrast, 5 of 6 streams in catchments with deeper till (>1 m) and >2% wetland area had mean annual maximum SO_4^{-2} concentrations which were <20 mg/L, although wetland area varied from 3–19%. Sulphate concentrations in streams draining non-wetland catchments with shallow till were not significantly different from non-wetland catchments with deeper till. Maximum SO_4^{-2} concentration ranged from 10–17 mg/L in both catchments types (Figure 1a).

During years with wet summers there was no significant difference in the average maximum SO_4^{-2} concentration of stream draining the four catchment types (Figure 1b, Table 2). The average maximum SO_4^{-2} concentration ranged from 6–13 mg/L.

The average maximum SO_4^{-2} concentration of years with dry summers rapidly increased from 10–15 mg/L to >20 mg/L in all but 1 stream draining wetland catchments as the % cover of deeper till in the non-wetland portion decreased to ≤50% (Figure 2). These data provide support for the initial categorization of catchment based on < or ≥50% of the area as minor till plain.

The average minimum SO_4^{-2} concentrations in streams draining wetland catchments were significantly less than concentrations in streams draining catchments with <2% wetland area (Figure 3, Table 2). The trend was similar both for years with dry and wet summers and in catchments with predominantly shallow till and deeper till.

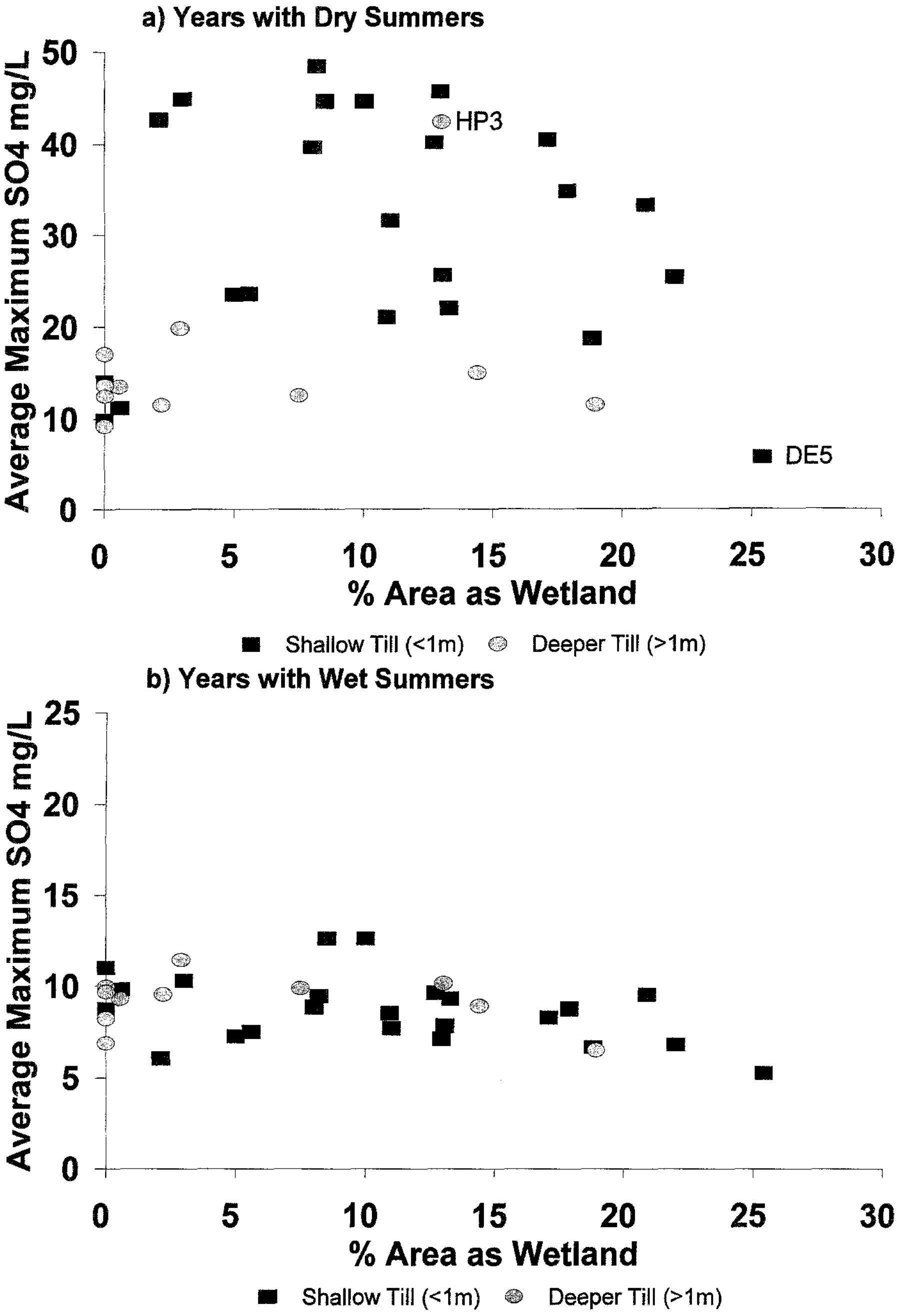


Figure 1. The average annual measured maximum SO_4^{-2} concentrations during years with a) dry summers; and b) wet summers in relation to the % wetland area of the catchment. Catchments were categorized as: 1) deeper till if $\geq$50% of the catchment was covered with till >1 m depth, and 2) shallow till if >50% of the catchment as covered with thin till-bedrock ridges <1 m depth. Dry summers had <300 mm and wet summers >350 mm for June–September rainfall.

Table 2. Analysis of variance for the mean annual maximum and minimum sulphate concentrations in streams draining catchments with or without wetlands, deep and shallow till depth during dry (<300 mm rainfall) and wet summers (>350 mm) from 1980 to 1992, Dorset, Ontario.

Source of variation	Mean annual maximum SO_4 concentration				Mean annual minimum SO_4 concentration			
	df	ms	Fs	Probability	df	ms	Fs	Probability
Between summers	1	972	12.7	0.0007**	1	2.9	0.9	0.3604
Between catchment	1	553	7.2	0.0092**	1	115	34	<0.0001**
Between till depth	1	325	4.3	0.0433*	1	17	5.1	0.0281*
Summer by catchment	1	371	4.9	0.0312*	1	0.2	0.05	0.8225
Summer by till depth	1	315	4.1	0.0466*	1	0.4	0.11	0.7421
Catchment by till depth	1	164	2.1	0.1481	1	9.6	2.8	0.0971
Summer by catchment by till depth	1	245	3.2	0.0783	1	0.04	0.01	0.9136
Residuals	60	76			60	3.4		
Contrasts:								
Between catchment in dry summers	1	915	6.2	0.0185*	1	53	18	0.0002**
Between till depths in dry summers	1	640	4.3	0.0459*	1	11	3.8	0.0608
Between catchment in wet summers	1	9	1.8	0.1891	1	62	16.4	0.0003**
Between till depths in wet summers	1	0.04	0.01	0.9295	1	6.2	1.6	0.2092
Between summers in wetland catchments	1	2201	25	<0.0001**	1	3.8	1.1	0.3071
Between till in wetland catchments	1	821	9.2	0.0039**	1	45	13	0.0009**
Between summer in non-wetland catchments	1	50	2.1	0.1721	1	0.6	0.2	0.6368
Between till in non-wetland catchments	1	9.6	0.4	0.5354	1	0.4	0.2	0.7002
Between till in wetland catchments during dry summers	1	1784	10.3	0.0037**	1	28	8.3	0.0082**

df = degrees of freedom, ms = mean square error, Fs = sample statistic of F distribution

** Significant, $\alpha = 0.01$

* Significant, $\alpha = 0.05$

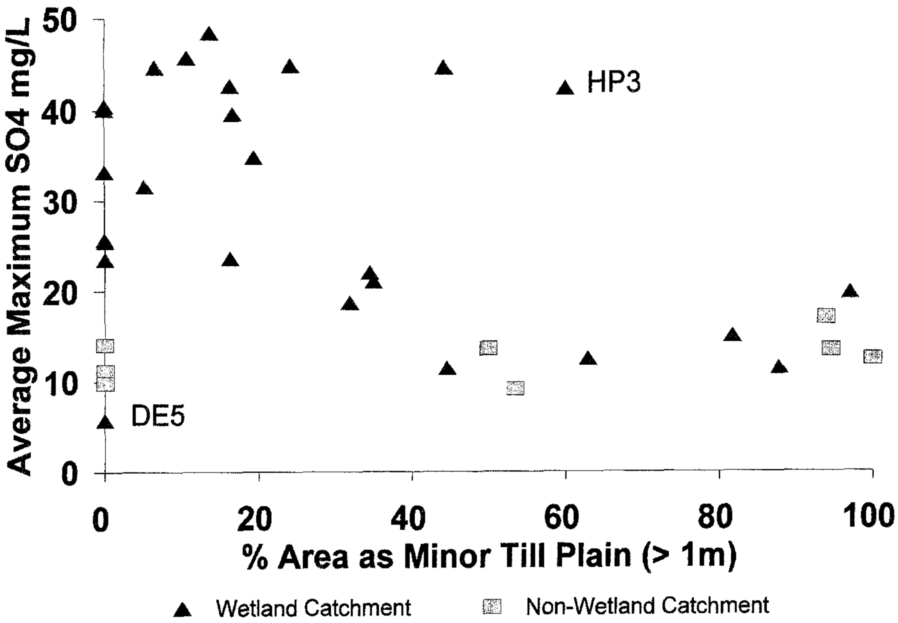
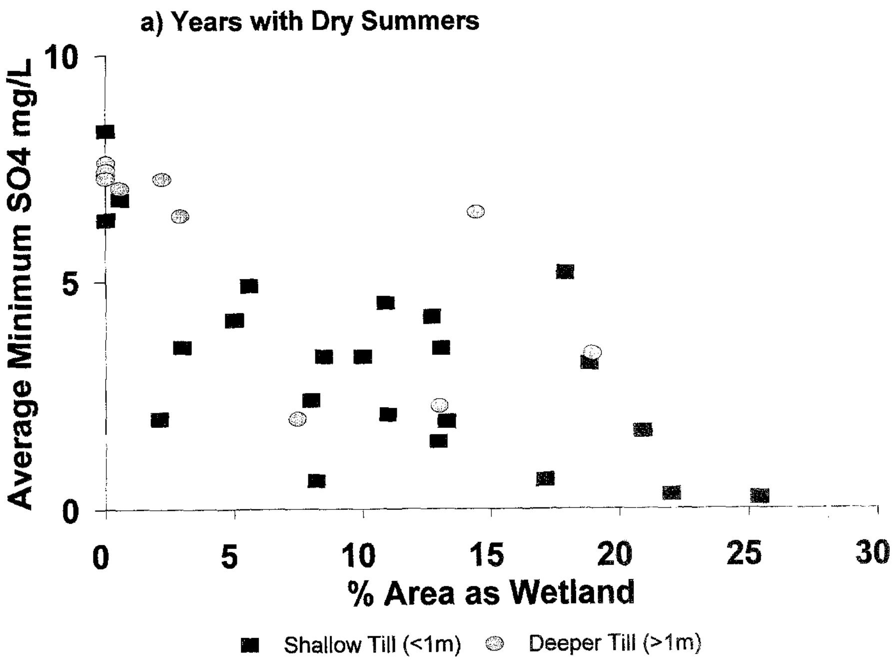


Figure 2. The average annual maximum SO_4^{-2} concentrations of 5 years with dry summers at stream sites relative to the % area of the headwater catchments covered with minor till plain (>1 m depth). Wetlands catchments had ≥2% of the area covered with wetlands.

Sulphate dynamics and summer rainfall

During 1980–1992 the average measured maximum SO_4^{-2} concentration of all streams draining shallow till catchments containing wetlands increased rapidly to values >25 mg/L when summer precipitation dropped below 300 mm, whereas the average of all streams draining shallow till catchments with <2% wetland area remained within 9–15 mg/L in all years (Figure 4). The average measured maximum SO_4^{-2} concentrations in streams draining deeper till wetland and non-wetland catchments increased to only 15–20 mg/L in years with summer precipitation <300 mm. The average maximum SO_4^{-2} concentrations of streams draining the four types of catchments were similar (8–12 mg/L) in years with >300 mm of summer precipitation.

Although the average minimum SO_4^{-2} concentrations varied by as much as 3 mg/L between years there was no relationship between average minimum SO_4^{-2} concentrations of streams draining any type of catchment and summer precipitation (Figure 5). The average minimum concentration in catchments with <2% wetland cover generally ranged between 6–8 mg/L. The average minimum concentration in streams draining wetland catchments was about 4–5 mg/L in catchments with deeper till and 2–3 mg/L in catchments with



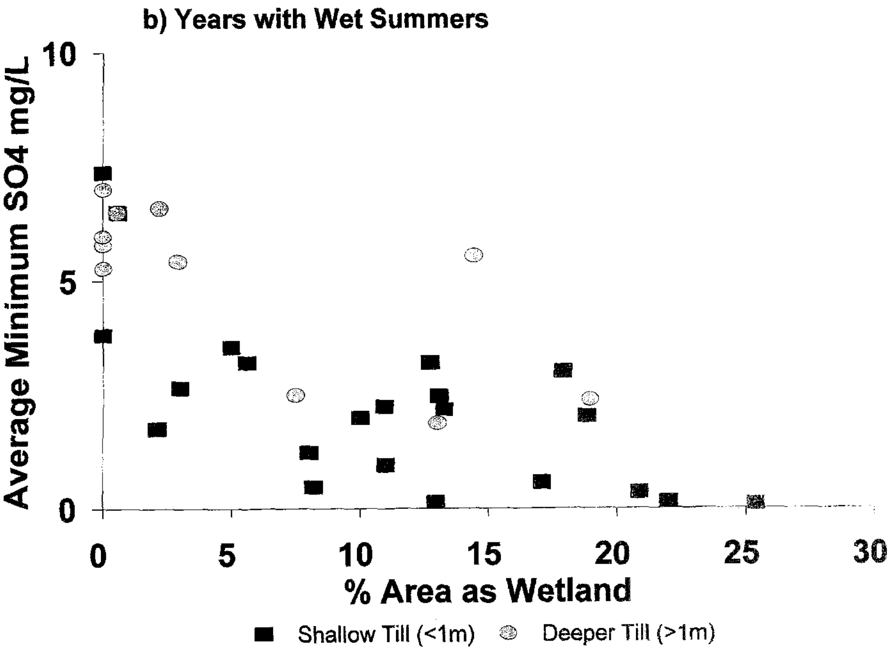


Figure 3. The average annual measured minimum SO_4^{-2} concentrations during years with a) dry summers; and b) wet summers in relation to the % wetland area of the catchment. Catchments were categorized as in Figure 1.

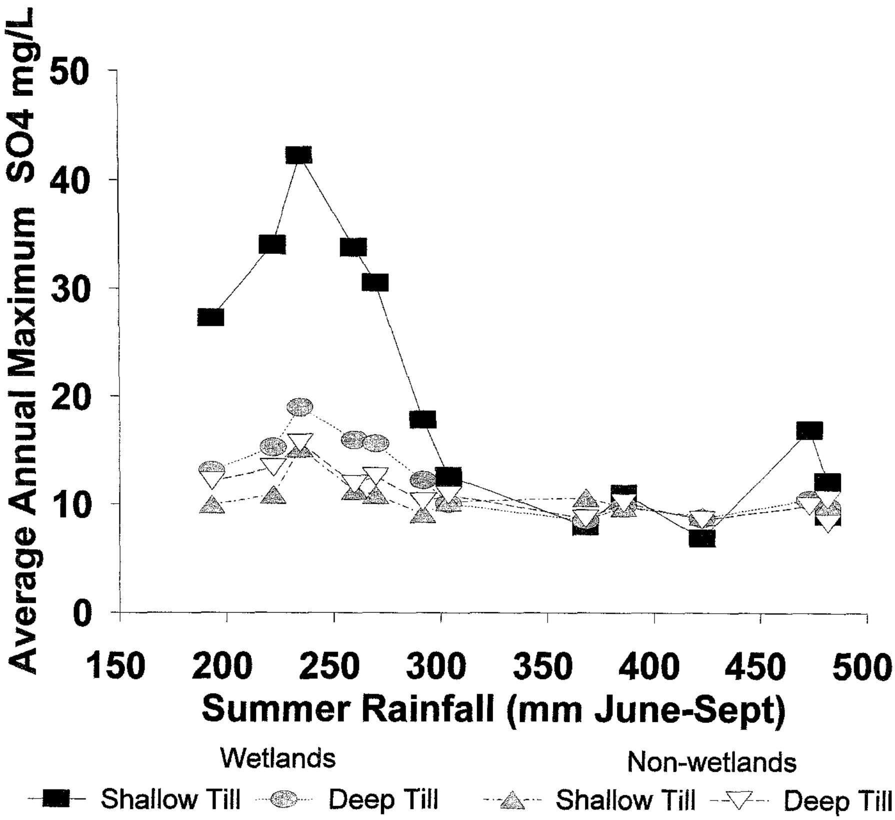


Figure 4. Average annual measured maximum SO_4^{-2} concentrations of all streams in each of four classes in relation to summer rainfall 1980–1992. Wetland catchments with shallow till ($n = 20$) and deeper till ($n = 6$). Non-wetland catchments with shallow till ($n = 3$) and deeper till ($n = 5$).

shallow till. The seasonal differences in SO_4^{-2} concentration was, therefore, much greater in streams draining wetland catchments relative to streams draining catchments with <2% wetland cover (Devito 1994). The seasonal differences in SO_4^{-2} concentration were greater in streams draining wetland catchments with shallow till relative to streams draining wetland catchments with deeper till.

Discussion

This analysis shows that catchment hydrogeology is strongly related to SO_4^{-2} transformation and transport in wetlands and headwater streams at the landscape scale. Classifying catchments based on < or ≥50% coverage of till

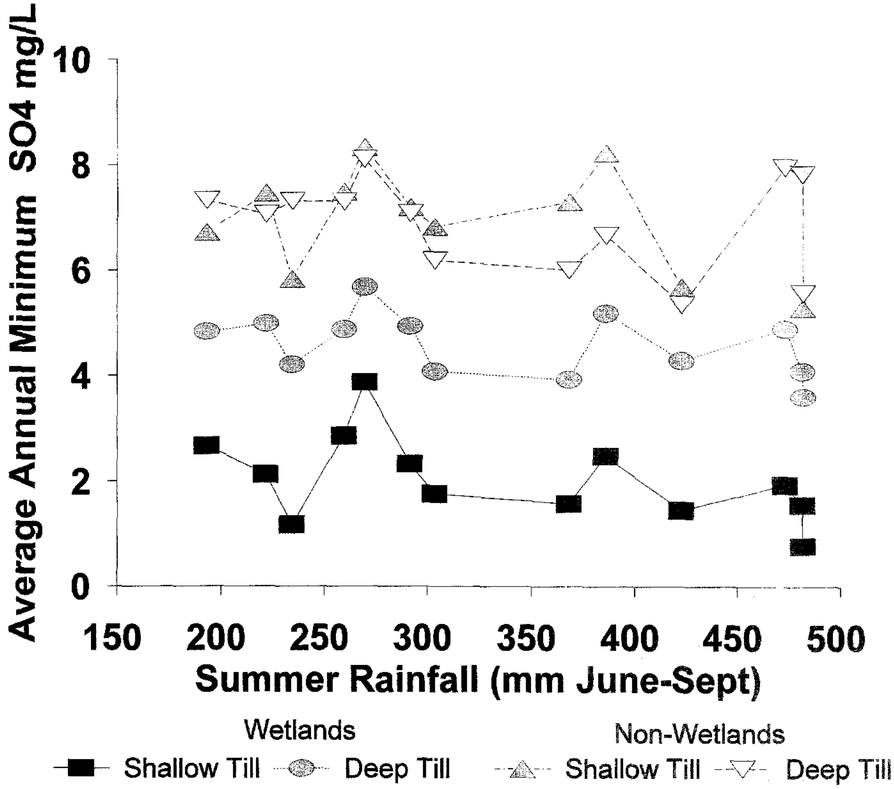


Figure 5. Average annual measured minimum SO_4^{-2} concentrations of streams in four catchment classes in relation to summer rainfall 1980–1992.

($>$1 m) in the non-wetland portion of the catchment appears to provide a useful method for distinguishing which wetlands can produce large SO_4^{-2} export following dry summers in this landscape. Peak SO_4^{-2} concentrations, indicative of a large water table draw down and re-oxidation of accumulated S (Devito 1994; Devito & Hill 1997, 1998), occurred in wetland catchments covered predominantly with shallow tills ($<$1 m). The upland-wetland hydrologic connection appears to change from a transient to a continuous link when the catchment is covered predominantly ($\geq$50%) with surficial deposits classified as minor till plains ($>$1 m depth; Devito et al. 1996).

The range of till depths observed in the catchments used in this study are typical of the shallow till-rock ridges physiographic region of the Canadian Shield (Chapman 1975). Consequently the range in biogeochemical behaviour observed in these catchments maybe considered typical of this region which covers a large portion of the Canadian Shield. Twenty of the 34 catchments were covered predominantly in shallow till and contained wetlands.

Assuming this proportion is representative of other areas, about 60% of the catchments in the southern Canadian Shield are susceptible to a large export of SO_4^{-2} and potential acidification downstream from the valley swamps in years with dry summers. The remaining % of catchments with wetlands will potentially have greater long term SO_4^{-2} retention.

Although wetlands may occupy only a small percentage of the catchment area, these data illustrate their large impact on the SO_4^{-2} dynamics of head-water catchments. This reflects the stream side position of wetlands in the landscape and their anoxic organic sediments (eg. Wieder 1985; Urban et al. 1989; Devito & Hill 1997). The lack of large SO_4^{-2} pulses in streams draining non-wetland watersheds indicates that this episodic release following dry summers originates mainly from the wetland portion of the watershed. The stores of reduced S in organic peats are the primary source of re-oxidized SO_4^{-2} exported from the watersheds following dry summers (Lazerte 1993; Devito 1995; Devito & Hill 1998). The maximum SO_4^{-2} concentrations during years with wet summers occurred during the high flow period from fall to spring. The lack of a relationship between maximum SO_4^{-2} concentration during years with wet summers and % wetland area indicates that upland sources of SO_4^{-2} have little interaction with anoxic soils during these periods because of the high water tables in the valley wetlands (Devito & Hill 1997).

Two of the 26 wetland catchments show maximum SO_4^{-2} concentrations during dry summers that would not be expected based on estimates of the distribution of catchment till depth. A stream entering Harp Lake (HP3, Figure 1) drains a wetland catchment (13% wetland) covered with 60% minor till plain but the average maximum SO_4^{-2} concentrations during the 5 dry years was above 40 mg/L. The outlet stream of HP3 catchments ran dry for more than a month during the dry summers (unpublished data) indicating that perhaps the surficial geology maps overestimate the percent cover of minor till plan in this catchment. Furthermore, road construction at the base of the catchment and earlier logging and debris dams observed in the stream near the wetland may have altered the drainage patterns and SO_4^{-2} dynamics in upstream soils.

A stream entering Dickie Lake (DE5, Figure 1) drains a wetland catchment (25% wetland) with exposed bedrock and patches of thin till but the average maximum SO_4^{-2} concentration during the dry summers was <6 mg/L. The stream outlet to the swamp ceased and visual observations showed no evidence of peat saturation during the dry summers (L. Scott, pers. comm.). It is unknown why no S re-oxidation and export occurs when this wetland dries out. The relatively large wetland area (25%) does not appear to be a factor. Discharge ceased during dry summers and high peak SO_4^{-2} concentrations were measured in the fall in the streams of four other headwater catchments to Dickie Lake with wetlands covering 17–22% of the area.

These results provide insight into the climatic conditions necessary to initiate S mobilization and SO_4^{-2} export from wetlands in this landscape. Although summer rainfall varied between 150–500 mm, potential evapotranspiration (PET) rates generally ranged between 425–475 mm over the same time period (Devito et al. 1996). A response in SO_4^{-2} export from the wetlands in catchment with shallow till occurs when the summer rainfall for June to September is <300 mm. Thus, a summer water deficit of at least 150–200 mm results in the cessation of upland runoff and water table drawdown in the valley swamps. This threshold response to summer precipitation suggests that upland and wetland water storage potential is relatively small in headwater catchments with shallow till. However, till depths do not need to be very deep to produce continuous upland-wetland connections. Groundwater discharge of 4 mm/day (equivalent to PET) throughout the summer from 2–3 m depth of minor till plains has been shown to be sufficient to maintain the water table near the peat surface in two wetlands in the Dorset region (Devito 1994; Devito et al. 1996).

Cessation of outlet stream discharge from all streams draining catchments with predominantly shallow till occurred during the dry summers. Cessation of stream outflow was concurrent with a water table drop of 0–10 cm below the peat surface in several of the study wetlands sites (Devito 1994; Devito et al. 1996) indicating that the potential depression storage of surface water is low. The specific yield of peat decreases with depth from about 0.5 at 0–10 cm to <0.1 at 30–40 cm (Devito et al. 1996). A water deficit of 10 cm would result in 20 cm, or greater, drop in water table below the peat surface. Observations from several basin swamps in this study (Devito 1994; Devito & Hill 1998) and other experiments (van Haesebroeck et al. 1997) indicate that a minimum water table drawdown of 20 cm is require for aeration and re-oxidation of accumulated S. Summer precipitation of 300 cm or less and a water deficit of 150–200 mm appears to result in a water table drawdown of 20 cm below the surface required for mobilization of reduced S followed by SO_4^{-2} peaks in headwater streams draining shallow till catchments in this landscape.

The physiography of the study area is representative of large areas of the Canadian Precambrian Shield (Chapman 1975). Our model which uses till depth as an indicator of upland-wetland-stream hydrologic and biogeochemical behaviour may also prove effective in predicting the SO_4^{-2} dynamics of wetlands in similar headwater catchments with thin tills and impermeable bedrock in the northern U.S. and Scandinavia which have historically received high atmospheric deposition rates of sulphur. The model described here may also prove useful in predicting at the landscape scale the dynamics of other elements controlled by water table fluctuation and redox processes in peat such as carbon (Roulet et al. 1992), nitrogen (Dillon et al. 1991; Devito & Dillon 1993) and mercury (Mierle 1990). These elements have significant environmental impact and are currently the focus of increased research.

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