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Contrasting stream nitrate and sulfate response to recovery from experimental watershed acidification

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Abstract Improvements in air quality have led to ecosystem recovery from acidic deposition, but the mechanisms and trajectories of this recovery are not fully understood. Here, we present long-term stream response and recovery data for paired watersheds at the Bear Brook Watershed in Maine (BBWM) during declining ambient SO_4 and NO_3 in precipitation. East Bear (EB) received ambient deposition from 1989 to 2018; West Bear (WB) received artificially elevated $N + S$ from 1989 to 2016. The WB treatment was discontinued after 2016, the beginning of the recovery from both the experimental $N + S$ and ambient decline. Stream SO_4 in WB gradually declined after the treatment ended, from ~ 147 µeq L^{-1} in 2010–16 to ~ 126 µeq L^{-1} in 2017–18. The declining S inputs induced desorption of SO_4 from soil phase surfaces, with stream loss far exceeding precipitation input. At the current rate of recovery, it will be many decades before the WB stream returns to pre-treatment SO_4 concentrations. In contrast, NO_3 is only weakly adsorbed in soil, and WB stream NO_3 concentrations rapidly declined from ~ 39 µeq L^{-1} in 2010–16 to ~ 5 µeq L^{-1} in 2017–18, comparable to the N-limited EB stream. The acid anions are strongly coupled to base cation chemistry in streams, and there was a distinct hysteretic response of Ca and Mg to the chronic acidification, as $(Ca + Mg)$ increased rapidly during the initial years, followed by declining values due to depletion of the soil exchange complex. This 30-year monitoring study (1989–2019) provides insights into recovery mechanisms from acidic deposition and highlights the role of abiotic processes in soil that mediate nutrient cycling and retention. Documenting the rapid response of N alongside the slower recovery for S identifies the temporal resolution necessary for other whole-watershed recovery studies.

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Introduction

Elevated acidic deposition in the form of reactive nitrogen (N) and sulfur (S) has altered biogeochemical processes in northeastern USA forests, beginning with industrialization and accelerating in the twentieth century. Forests that were typically N-limited received high N inputs, resulting in increased N losses via leaching and denitrification (Aber et al. 2003; Patel and Fernandez 2018). Acidic deposition accelerated base cation and aluminum (Al) loss from soils, acidifying the soil, increasing tree stress and mortality, and suppressing soil organic matter decomposition (Fernandez et al. 2003; Duarte et al. 2013; Ouimet et al. 2017; Oulehle et al. 2018). Successful implementation of the Clean Air Act in 1970 and its Amendments in 1990 has reduced acidic deposition in these ecosystems over the last 4+ decades (Kahl et al. 2004; Strock et al. 2014; Gavin et al. 2018; Sullivan et al. 2018; Shao et al. 2020). Recent studies have reported ecosystem recovery from elevated nutrient and acidic deposition, including decreased acid anion concentrations and increased base cations, dissolved organic carbon (DOC), acid neutralization capacity, and pH in soils, streams, and lakes, in many, although not all, ecosystems (Strock et al. 2014; Fuss et al. 2015; Lawrence et al. 2015; Armfield et al. 2019; Gilliam et al. 2019). Many conceptual models suggest hysteretic patterns in ecosystem recovery from acidic deposition with respect to base cation and nitrate leaching, and adsorption-desorption processes in soils (Fernandez et al. 2003; Gilliam et al. 2019). But because site-specific factors influence many of these processes, it is currently unclear how different systems respond to these improvements in air quality.

The Bear Brook Watershed in Maine (BBWM) is a long-term experimental watershed established to study the effects of whole-watershed acidification and N-enrichment (Patel et al. 2019a). Established in the late 1980s, the West Bear (WB) watershed received nearly three decades of experimentally elevated N and S deposition, whereas the reference East Bear (EB) watershed received only ambient atmospheric deposition. The treatment increased stream NO_3, SO_4, base

cations, Al, and phosphorus (P) leaching in WB (Fernandez et al. 2003, 2010). The experimental chemical manipulation of West Bear was discontinued in November 2016, marking the onset of the "recovery phase" from long-term experimental acidification and N enrichment, plus the longer-term more gradual decline in ambient N + S deposition. Here, we describe initial stream chemistry response to termination of the long-term acidification treatment of WB, focusing on NO_3 and SO_4.

Methods

Site description

The Bear Brook Watershed in Maine (44°52'N, 68°06'W) consists of two paired first order forested watershed streams. East Bear brook (EB, 11.0 ha), the reference, received ambient deposition. West Bear brook (WB, 10.3 ha) received bimonthly applications of ammonium sulfate $[(NH_4)_2SO_4]$ fertilizer from above the canopy, at the rate of 28.8 kg S ha^{-1} $year^{-1}$ and 25.2 kg N ha^{-1} $year^{-1}$, from November 1989 to October 2016. Both watersheds are drained by first-order nearly perennial streams. Soils, developed from till, are coarse-loamy, mixed, frigid Typic and Aquic Haplorthods. Vegetation is similar in both watersheds, with lower elevations dominated by hardwood species, primarily *Fagus grandifolia* Ehrh. (American beech), *Acer saccharum* Marsh. (sugar maple), *Acer rubrum* L. (red maple), and *Betula alleghaniensis* Britt. (yellow birch); higher elevations are dominated by softwood species, *Picea rubens* Sarg. (red spruce) and *Abies balsamea* L. (balsam fir). Additional site information is in Norton et al. (1999), Patel et al. (2019a), and the publications cited therein.

Stream chemistry and input-output fluxes

Stream samples were collected manually as grab samples during baseflow conditions, and with ISCO™ automated samplers during snowmelt and rain high flow events from EB and WB. During the first half of the manipulation period, stream samples were collected weekly (baseflow) and during high stream-flow events (up to 300 samples per year). During the second half of the study period, sampling frequency was reduced; >50 samples per year were collected

biweekly or monthly, and more frequently during selected hydrologic events. Stream samples were analyzed for NO_3^- and SO_4^{-2} (ion chromatography, IC), NH_4^+ (colorimetry), and base cations (Ca^{+2}, Mg^{+2}, Na^+, K^+) by inductively coupled plasma atomic emission spectrometry (ICP-AES) at the University of Maine's Sawyer Environmental Research Center. Measured stream chemistry from baseflow samples plus hydrologic events were interpolated between collections and coupled with hourly stream discharge to develop annual stream fluxes (Kahl et al. 1999; Norton et al. 2010).

Wet-only precipitation was collected using an AeroChem-Metrics^{TM} precipitation collector (Norton et al., 1999). Collections were made at the site until 2014. From 2014 onwards, we used annual wet deposition values from the CASTNET Howland station (HOW132), located ~60 km northwest of BBWM (CASTNET 2018). The annual wet deposition overlapping values for HOW132 and BBWM were strongly correlated (Pearson's r = 0.7735, $p < 0.0001$). Similarly, dry deposition was not measured at the site, so for this analysis we used reported estimates of annual dry deposition from the CAST-NET Howland station (Patel et al. 2019a). Wet-only precipitation samples were analyzed for NH_4^+, NO_3^-, and SO_4^{-2} using ion chromatography at the University of Maine's Sawyer Environmental Research Center.

Annual mass-balance nutrient budgets were calculated for EB and WB using annual input and output fluxes. Annual ecosystem nutrient retention was calculated as wet + dry deposition (+ treatment for WB) *input* – stream *output*. A watershed was considered *leaky* when outputs exceeded inputs and was considered *retentive* when inputs exceeded outputs. All annual concentrations and fluxes were computed for water years from October 1 to September 30.

Results and discussion

Recovery phase for WB: Contrasting patterns in sulfate and nitrate

The $(NH_4)_2SO_4$ treatment increased WB exports of stream SO_4 and NO_3 during the treatment period (1989–2016) (Fig. 1). Stream SO_4 concentrations rapidly increased (1989–1996) and then gradually declined (1996–2016). In contrast, NO_3 concentrations increased even more rapidly, but then stabilized at elevated concentrations in WB but with much higher interannual variability and inconsistent patterns in the second and third decades. Noteworthy was the initial behavior in recovery for WB (2016–18), when SO_4 and NO_3 showed very different responses.
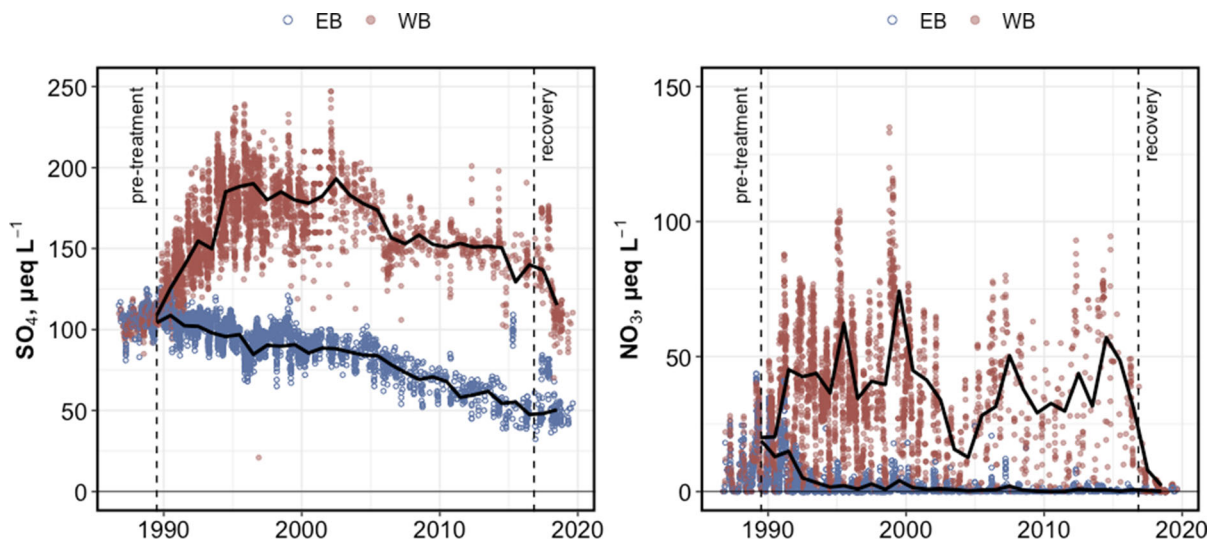

Fig. 1 Stream concentrations of SO_4 and NO_3. *EB* East Bear (reference); *WB* West Bear (treated). Solid lines represent annual volume-weighted mean concentrations

Long-term, gradual declines in S continue post-recovery

Stream SO_4 concentrations for WB averaged 163 µeq L^{-1} during the entire treatment, ~ 2 times the average concentration in the reference EB (80 µeq L^{-1}). There was a gradual decline in WB stream SO_4 concentrations from 1995 to the end of treatment, at a rate similar to that of EB for the same period (~ 2 µeq L^{-1} year^{-1}). This pattern of decline is widely reported in northeastern USA watersheds and is attributed to the declining atmospheric SO_4 deposition as a result of the CAA and its Amendments. Post-treatment (2017–2018), SO_4 in WB declined substantially, ~ 25 µeq L^{-1} from 2016 to 2018 (Fig. 1).

Treatment recovery triggers abrupt N decline to near reference conditions within two years

Stream NO_3 concentrations for the treated WB were ~ 10 times the values in EB during the early years of manipulation (Fig. 1), as N-enrichment stimulated N mineralization (nitrification) in WB soils (Nadelhoffer et al. 1999). This ratio increased to ~ 95 by 2005, driven by the experimental additions in WB and the declining concentrations in the reference EB (Patel and Fernandez 2018). Following treatment termination, WB NO_3 volume-weighted concentrations abruptly decreased by nearly 75% to 8.0 µeq L^{-1} in 2017 and to 2.2 µeq L^{-1} in 2018, approaching the concentration in the reference EB (0.2 µeq L^{-1}).

Intra-annual patterns of stream NO_3 provide insight on mechanisms governing WB stream N recovery (Fig. 2). High streamflow events during snowmelt typically have higher concentrations of NO_3 compared to baseflow conditions, because NO_3 produced under the snowpack in winter is flushed from the soils with meltwater (Casson et al. 2014). This effect was magnified in the treated WB stream during the manipulation period. The magnitude of difference in snowmelt-dominated vs. baseflow WB concentrations declined almost exponentially over the two post-treatment years. Snowmelt-period NO_3 concentrations in WB steadily declined from mean values of 11.0 µeq L^{-1} in 2017 to 4.3 µeq L^{-1} in 2018 and 2.9 µeq L^{-1} in 2019. Baseflow NO_3 concentrations during the 2017–2018 were below-detection in WB,
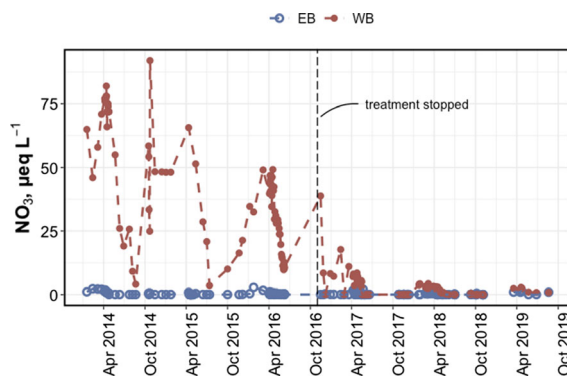

Fig. 2 Stream NO_3 concentrations in EB (East Bear, reference) and WB (West Bear, treated) for all stream samples (base flow and hydrologic event) during 2014–2019. Snowmelt typically occurs in April in both watersheds. The dashed line represents cessation of treatment in November 2016

comparable to EB. By spring of 2019, snowmelt-dominated stream concentrations in WB did not differ significantly from EB (Fig. 2).

Reversible adsorption in soils drives sulfate response

Research in northeastern USA forests has found relatively high specific and non-specific adsorption of SO_4 in Spodosols (Fuller et al. 1985). Although S is an essential plant nutrient, nutrient requirements are typically fulfilled by unpolluted atmospheric deposition; ambient deposition at Bear Brook has been greater than demand (Mitchell et al. 2011). This results in SO_4 leaching losses from soils after the specific adsorption capacity of the soil is exceeded. Ecosystem S retention is dominated by SO_4 adsorption on colloidal surfaces with a portion of the S slowly incorporated into the organic phase by biological uptake and litter inputs to the soil (Novák et al. 2005; Mitchell et al. 2011). In a quasi-steady state, leaching equals input.

As the $(NH_4)_2SO_4$ treatment increased SO_4 concentrations in soil solution, SO_4 adsorption in WB soils increased, as did stream SO_4 exports (Fernandez et al. 1999). Partitioning between soil and soil solution changes, favoring adsorption as declining pH increases the positive surface charge of Al and Fe hydroxides, and consequently anion adsorptive capacity (Nodvin et al. 1986; Stumm 1992). When experimentally elevated inputs of S to WB abruptly ceased in late 2016, soil S dynamics switched from net

adsorption to net desorption, reflecting lower soil solution SO_4 concentrations (Kahl et al. 2004; Mitchell et al. 2011). Cessation of treatments is presumed to have increased soil pH, which would cause hydrolysis of primarily the abundant solid-phase Al hydroxide complexes, reducing SO_4 adsorption capacity of soils, thereby releasing H^+ and SO_4 (Dahlgren et al. 1990; Nodvin et al. 1986).

In 1987–1991, Rustad et al. (1996) added HNO_3 and H_2SO_4 to experimental plots near the BBWM watersheds, followed by the study of soil (1991–1993) and soil solution (1991–1994) recovery. The soils adsorbed SO_4 during their four-year acidification treatment; thereafter, soil SO_4 desorption commenced, gradually approaching a new equilibrium with soil solutions. We see a similar response at the watershed scale in our current study, with gradual release of SO_4 from the soils into the stream.

Nitrogen response is driven by mineralization/ immobilization dynamics

Unlike SO_4, there is little abiotic NO_3 adsorption in the soils at BBWM (Rustad et al. 1996; Strahm and Harrison 2007). Most of the N retention over the study period is attributable to biological immobilization with accumulated N stored in biota and soil organic matter (Nadelhoffer et al. 1999; Patel et al. 2019a). Thus, upon cessation of elevated N inputs in 2016, there was no significant abiotically adsorbed N for desorption, and N immobilized in soil organic matter would remain tightly cycled with little loss to leaching.

The reduced stream NO_3 export is consistent with soil NO_3 patterns. In soil samples collected in 2017, NO_3 concentrations did not differ between the two watersheds, although extractable NH_4 was ~6 times greater in WB than in EB (Appendix A1). Long-term soil records indicate that the N + S additions significantly increased NO_3 concentrations in WB soils during the treatment period (Patel and Fernandez 2018). Various mechanisms have been described to explain increased NO_3 availability, including (a) increased nitrification due to availability of labile NH_4 substrate, and (b) suppressed plant and mycorrhizal NO_3 uptake (Högberg et al. 2007; Emmett 2007). Possibly the sudden loss of NH_4 input may have reversed one or more of these mechanisms, although additional research is needed for a definitive explanation.

Response of stream cations

The acidification treatment has historically reduced the acid neutralizing capacity (ANC) of the WB stream (Laudon and Norton 2010; Navrátil et al. 2010). ANC represents the difference between the base cations (BC) (Ca^{+2}, Mg^{+2}, Na^+, K^+) and the acid anions (NO_3^-, SO_4^-, Cl^-) (Laudon and Norton 2010). Acidic DOC and ionic Al species are not included for simplicity. In the charge balance in these low-DOC streams, Na^+ is almost balanced by Cl^-, with a small contribution from weathering; K^+ is typically <5–6 μeq L^{-1} with little inter-or intra-annual variation (Navrátil et al. 2010). Thus, here we use (Ca^{+2} + Mg^{+2}) – (SO_4^{-2} + NO_3^-) as an index of ANC in runoff that allows us to track the evolution of base cations over time.

The temporal trends of charge balance in EB and WB generally follow the conceptual model reported in Fernandez et al. (2003) describing stream base cation chemistry and soil chemistry during periods of acid deposition and recovery (Fig. 3). The 1:1 line in Fig. 3 indicates a balance between base cations and strong acid anions, and points below this line represent an acidified stream. The EB stream showed declines in (SO_4 + NO_3) as well as (Ca + Mg) during the study period and remained close to the 1:1 line, indicating
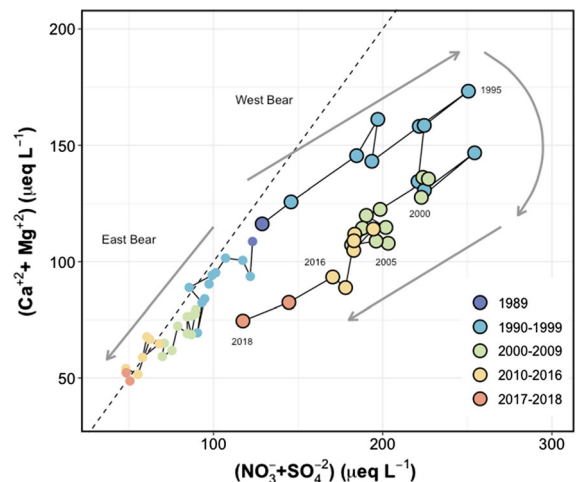

Fig. 3 Progression of stream (Ca + Mg) versus (NO_3 + SO_4) volume-weighted concentrations for East Bear and West Bear streams. The dashed line represents the 1:1 line where base cations and strong acid anions are balanced; points below this line represent an acidified stream. Arrows describe a generalized pattern in the evolution of stream chemistry for both watersheds

only slightly acidic pH (Fig. 3). In WB, from 1990 to 1995, stream (SO_4 + NO_3) increased with the experimental treatment, accompanied by a dramatic increase in (Ca + Mg) leached through cation desorption from the acidifying WB soils (Fernandez et al. 2003). This response is consistent with coupled NO_3-Ca leaching at the Hubbard Brook, NH (Likens et al. 1996) and Fernow, WV (Edwards et al. 2006) watersheds in eastern USA. Gilliam et al. (2020) recently summarized the results of 25 years of treatments with $(NH_4)_2SO_4$ at the Fernow Experimental Forest in West Virginia, also reporting declines in soil base cations from accelerated leaching losses due to treatment, and higher concentrations of base cations in the stream. WB stream (Ca + Mg) at BBWM declined after 1995, attributed to depleted exchangeable base cations in the soil being unable to provide charge balance to increasing strong acid anion export which was increasingly dominated by aluminum (Al) (Norton et al. 2010). As ambient SO_4 deposition declined, the annual data for WB moved toward the 1:1 line, as strong acid anions and base cations declined. This coupled pattern accelerated during the 2017–2018 recovery period. Both (Ca + Mg) and (SO_4 + NO_3) in 2018 were lower than the initial values in 1989, as is true for EB. For WB, (Ca + Mg) is expected to continue to decrease and then slowly increase as the soil exchangeable base cations recover from weathering (i.e. increasing BC adsorption and base saturation) (Lawrence et al. 2015). Eventually, stream BC concentrations will reach a new dynamic equilibrium, and climatic factors may influence that trajectory (e.g. Houle et al. 2010; Kopáček et al. 2017b). EB may have reached its lowest (Ca + Mg) concentrations, stabilized at 50 µeq L^{-1} for 4 years. If so, BC concentrations will increase if (SO_4 + NO_3) continues to decline.

Ecosystem N + S retention and implications for ecosystem recovery

Although initially leaky with respect to SO_4 (i.e., inputs < outputs), WB became a more SO_4-retentive watershed during the treatment period, and retained, on average, ∼21% of the annual inputs, and in some years, even >50% (Fig. 4; Appendix A2). Over the 27 years of treatment, WB had a cumulative net retention of 195 kg S ha^{-1}, compared to EB, which experienced a cumulative net loss of 188 kg S ha^{-1}

(Fig. 4, Appendix A2). When the WB treatment stopped, desorption dominated soil SO_4 dynamics, as reflected in the declining but still elevated SO_4 export. During the WB recovery period reported, WB had a net loss of ∼21 kg S ha^{-1} $year^{-1}$ for 2017–2018.

If soil SO_4 desorption continues at the current rate, it would take 19 years for complete SO_4 recovery, if defined as the loss of the cumulative excess S in WB (compared to EB). However, the rate of S desorption from soils will decline over time (Edwards 1998; Kopáček et al. 2014), and complete stream recovery may take many decades. Recovery is further complicated by climatic influences on S retention such as increased SO_4 leaching during drought periods due to the oxidation of organically bound S (Eimers et al. 2004; Strock et al. 2016).

West Bear had the opposite pattern from SO_4 with respect to N retention. The elevated N-inputs reduced ecosystem N retention in WB, due, at least in part, to increased nitrification in soils and subsequent stream exports. In total, WB retained ∼82% of the annual inputs during the treatment period, compared to EB, which had ∼95% ecosystem N retention. After 27 years of experimental additions, WB retained a cumulative 682 kg N ha^{-1}, compared to the cumulative retention of 146 kg N ha^{-1} in EB, mainly in the aboveground biomass and the surface organic soil (Patel et al. 2019a). Gaseous N losses are minimal at BBWM, and stream export is the major pathway of N loss (Patel et al. 2019a). Because WB stream NO_3 concentrations declined almost to ambient (∼1 µeq L^{-1}) within the initial recovery period, even during typically N-enriched snowmelt periods, we see a switch to a more N-retentive WB, and we expect the amount of N retained in the watershed will only increase with time (Schmitz et al. 2019). The difference in N storage between the two watersheds will likely continue until a new steady state is achieved, or the N cycle is disrupted (e.g., fire, harvests, ice storms, insects, and disease), which would result in major losses of accumulated N from the system (Ryan et al. 1992; Houlton et al. 2003; Kopáček et al. 2017; Oulehle et al. 2019; Patel et al. 2019b).

This unique multi-decadal, whole watershed manipulation experiment provides evidence for mechanisms of ecosystem acidification, N and S enrichment, and initial evidence and mechanisms of recovery. While site-specific factors govern many ecosystem processes, the initial stream recovery
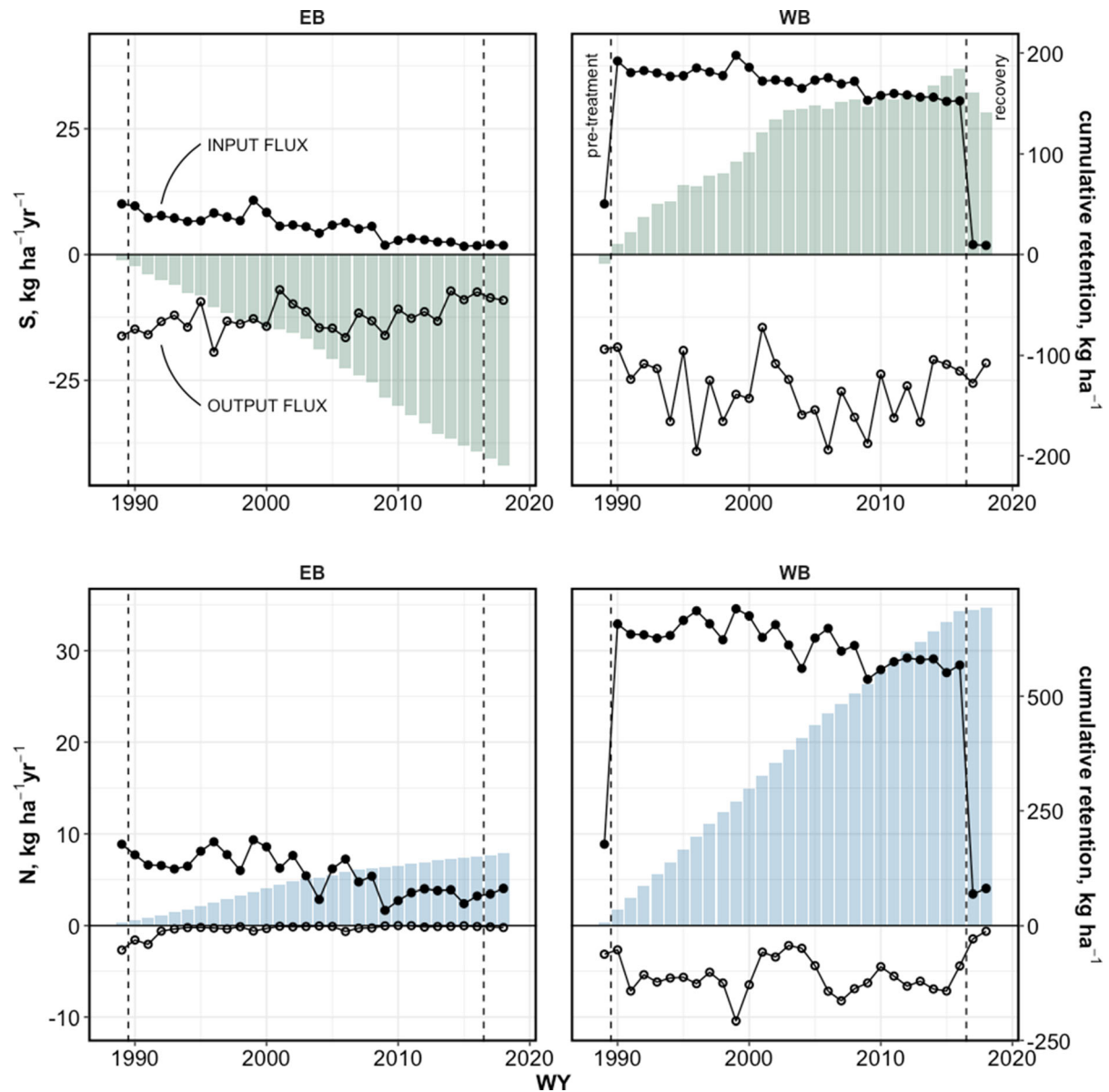
Fig. 4 Input-output fluxes (solid circles = inputs, open circles = outputs) and cumulative ecosystem retention (bars, secondary y-axis) of S and N. A positive retention indicates that inputs > outputs, and a negative retention indicates inputs < outputs. *EB* East Bear (reference); *WB* West Bear (treated)

patterns of NO_3 and SO_4 export, coupled with base cation export, are generally consistent with mechanisms reported for other experimental forested watersheds in eastern North America (Watmough et al. 2005; Lawrence et al. 2015; Hazlett et al. 2020). In addition, our data show the hysteretic nature of stream chemical recovery from acidification, consistent with other evidence in the literature (e.g., Gilliam et al.

2019). The dominant acid anions, NO_3 and SO_4, have notably different trajectories and timescales of ecosystem retention and recovery. While stream NO_3 recovery from N-enrichment was rapid, recovery of SO_4, which is dominated by the rate of abiotic soil SO_4 desorption, should persist for decades or more, delaying stream recovery.

Appendices

Appendix A1: Soil extractable inorganic nitrogen

Methods: Sampling and extraction

A field campaign was conducted in July 2017 to collect soil samples from the hardwood and softwood stands of both watersheds. Soils were sampled from

Table 1 Extractable inorganic N (NH_4-N and NO_3-N) in soils sampled in 2017

	EB	WB
NH_4-N, mg N kg^{-1}	3.63 ± 0.15	23.40 ± 1.88*
NO_3-N, mg N kg^{-1}	0.68 ± 0.12	0.79 ± 0.15

Values reported as mean ± standard error. Asterisks denote significant differences between watersheds at $\alpha = 0.05$

existing ecosystem monitoring plots located 60 m apart along east-west transects (18 plots in EB, 20 plots in WB), in order to representatively capture elevation, aspect, and forest composition gradients in each watershed. The surface litter was removed and soil samples were collected with a push corer from the top 15 cm, integrating the organic and mineral horizons. Soil samples were sieved through a 2 mm screen and homogenized.

Inorganic N (NH_4 and NO_3) was extracted from 5 g subsamples of field-moist soil in 25 mL of 2.0 M KCl. Samples were shaken for 1 hour, allowed to settle, then filtered through Whatman Grade 1 filter paper. NH_4-N and NO_3-N were then analyzed using a Lachat QuickChem® 8500 Series Flow Injection Analyzer (Lachat Instruments, Loveland, CO) at the University of Virginia (Table 1).

Appendix A2: Annual ecosystem mass balance for N and S

A positive value in the "retention" column indicates net ecosystem retention (inputs > outputs), and a negative value indicates net ecosystem loss (inputs < outputs). Percentage retention is calculated with respect to inputs, for positive balances only.

| WY | East Bear | | | | | | | |
| | Sulfur kg S ha^{-1} year^{-1} | | | | Nitrogen kg N ha^{-1} year^{-1} | | | |
	Input	Output	Retention	% retention	Input	Output	Retention	% retention
1989	10.05	16.22	−6.17	–	8.52	2.67	5.85	68.66
1990	9.39	14.85	−5.46	–	8.03	1.57	6.46	80.45
1991	7.88	15.94	−8.06	–	6.84	2.05	4.79	70.03
1992	7.39	13.33	−5.94	–	6.43	0.60	5.83	90.67
1993	7.35	12.08	−4.73	–	6.12	0.37	5.75	93.95
1994	7.48	14.46	−6.98	–	7.22	0.22	7.00	96.95
1995	5.69	9.39	−3.70	–	6.58	0.20	6.38	96.96
1996	7.91	19.39	−11.48	–	8.45	0.27	8.18	96.80
1997	8.95	13.26	−4.31	–	9.62	0.37	9.25	96.15
1998	5.99	13.82	−7.83	–	5.94	0.12	5.82	97.98
1999	10.62	12.78	−2.16	–	8.91	0.59	8.32	93.38
2000	8.99	14.30	−5.31	–	9.55	0.32	9.23	96.65
2001	5.26	7.02	−1.76	–	4.93	0.07	4.86	98.58
2002	6.68	9.84	−3.16	–	9.03	0.14	8.89	98.45
2003	4.35	11.38	−7.03	–	4.58	0.10	4.48	97.82
2004	5.49	14.58	−9.09	–	4.36	0.04	4.32	99.08
2005	5.22	14.66	−9.44	–	5.19	0.09	5.10	98.27
2006	6.35	16.53	−10.18	–	7.24	0.64	6.60	91.16
2007	5.63	11.65	−6.02	–	4.05	0.28	3.77	93.09
2008	5.62	13.20	−7.58	–	5.72	0.26	5.46	95.45
2009	2.25	16.10	−13.85	–	1.89	0.03	1.86	98.41
2010	2.51	10.86	−8.35	–	2.46	0.00	2.46	100.00
2011	3.77	12.70	−8.93	–	4.20	0.01	4.19	99.76
2012	2.71	11.39	−8.68	–	3.99	0.16	3.83	95.99
2013	2.60	13.25	−10.65	–	3.76	0.10	3.66	97.34
2014	2.58	7.26	−4.68	–	3.87	0.08	3.79	97.93
2015	1.74	8.99	−7.25	–	2.20	0.02	2.18	99.09
2016	1.86	7.46	−5.60	–	3.10	0.10	3.00	96.77
2017	2.06	8.61	−6.55	–	3.35	0.14	3.21	95.82
2018	1.91	9.10	−7.19	–	4.01	0.21	3.80	94.76
Cumulative retention		−208.12					158.32	

| WY | West Bear | | | | | | | |
| | Sulfur kg S ha^{-1} year^{-1} | | | | Nitrogen kg N ha^{-1} year^{-1} | | | |
	Input	Output	Retention/loss	% retention	Input	Output	Retention/loss	% retention
1989	10.05	18.83	−8.78	–	8.52	3.13	5.39	63.26
1990	38.19	18.42	19.77	51.77	33.23	2.65	30.58	92.03
1991	36.68	24.78	11.90	32.44	32.04	7.15	24.89	77.68
1992	36.19	21.70	14.49	40.04	31.63	5.37	26.26	83.02
1993	36.15	22.66	13.49	37.32	31.32	6.17	25.15	80.30
1994	36.28	33.20	3.08	8.49	32.42	5.73	26.69	82.33
1995	34.49	19.09	15.40	44.65	31.78	5.65	26.13	82.22
1996	36.71	39.14	−2.43	–	33.65	6.35	27.30	81.13
1997	37.75	24.99	12.76	33.80	34.82	5.10	29.72	85.35
1998	34.79	33.18	1.61	4.63	31.14	6.28	24.86	79.83
1999	39.42	27.79	11.63	29.50	34.11	10.42	23.69	69.45
2000	37.79	28.59	9.20	24.35	34.75	6.47	28.28	81.38
2001	34.06	14.51	19.55	57.40	30.13	2.90	27.23	90.38
2002	35.48	21.68	13.80	38.90	34.23	3.43	30.80	89.98
2003	33.15	24.82	8.33	25.13	29.78	2.18	27.60	92.68
2004	34.29	31.86	2.43	7.09	29.56	2.48	27.08	91.61
2005	34.02	30.88	3.14	9.23	30.39	4.38	26.01	85.59
2006	35.15	38.83	−3.68	–	32.44	7.17	25.27	77.90
2007	34.43	27.18	7.25	21.06	29.25	8.22	21.03	71.90
2008	34.42	32.34	2.08	6.04	30.92	6.90	24.02	77.68
2009	31.05	37.58	−6.53	–	27.09	6.27	20.82	76.85
2010	31.31	23.76	7.55	24.11	27.66	4.49	23.17	83.77
2011	32.57	32.48	0.09	0.28	29.40	5.52	23.88	81.22
2012	31.51	26.10	5.41	17.17	29.19	6.62	22.57	77.32
2013	31.40	33.30	−1.90	–	28.96	6.08	22.88	79.01
2014	31.38	20.88	10.50	33.46	29.07	6.93	22.14	76.16
2015	30.54	21.81	8.73	28.59	27.40	7.15	20.25	73.91
2016	30.66	23.14	7.52	24.53	28.30	4.41	23.89	84.42
2017	2.06	25.57	−23.51	–	3.35	1.45	1.90	56.72
2018	1.91	21.55	−19.64	–	4.01	0.62	3.39	84.54
Cumulative retention			143.24				692.87	

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