



Unearthing a stream-wetland floodplain system: increased denitrification and nitrate retention at a legacy sediment removal restoration site, Big Spring Run, PA, USA

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Abstract Nitrogen (N) retention is a common goal of stream-wetland restoration projects in systems with excess nitrate (NO_3^-), however N retention depends on habitats with high denitrification and uptake rates that interact with NO_3^- . Legacy sediments deposited along formerly impounded streams bury and disconnect historic floodplain-wetland systems. This disconnection limits sediment-water interactions,

decreases N retention and increases N delivery. Restoration with legacy sediment removal should lead to greater N retention due to the reestablishment of wet habitats that interact with NO_3^- -rich water, but the formation of biogeochemically retentive soils under modern conditions of high NO_3^- , N retention rates, and recovery time are unclear. An experimental restoration approach undertaken at Big Spring Run in Lancaster, PA, USA was used to test the hypothesis that reconnection of a stream to its historic floodplain with legacy sediment removal enhances N processing and retention. We describe changes in sediment and water concentrations of N and organic carbon

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(C) along with the changes in sediment biogeochemical processing rates of denitrification, nitrification, and C mineralization, before and for five years following restoration. Our results show that biogeochemical processing increased and higher NO_3^- retention developed following stream-wetland restoration. NO_3^- retention improved after several years as organic matter accumulated to ultimately support higher rates of denitrification that transitioned from organic C limitation to NO_3^- limitation. We conclude that, in systems with high contemporary NO_3^- , restoration via legacy sediment removal and floodplain reconnection can lead to the accumulation of organic matter and improved biogeochemical NO_3^- retention over time

Keywords Nitrogen · Organic carbon · Legacy sediment · Floodplain · Groundwater · Restoration

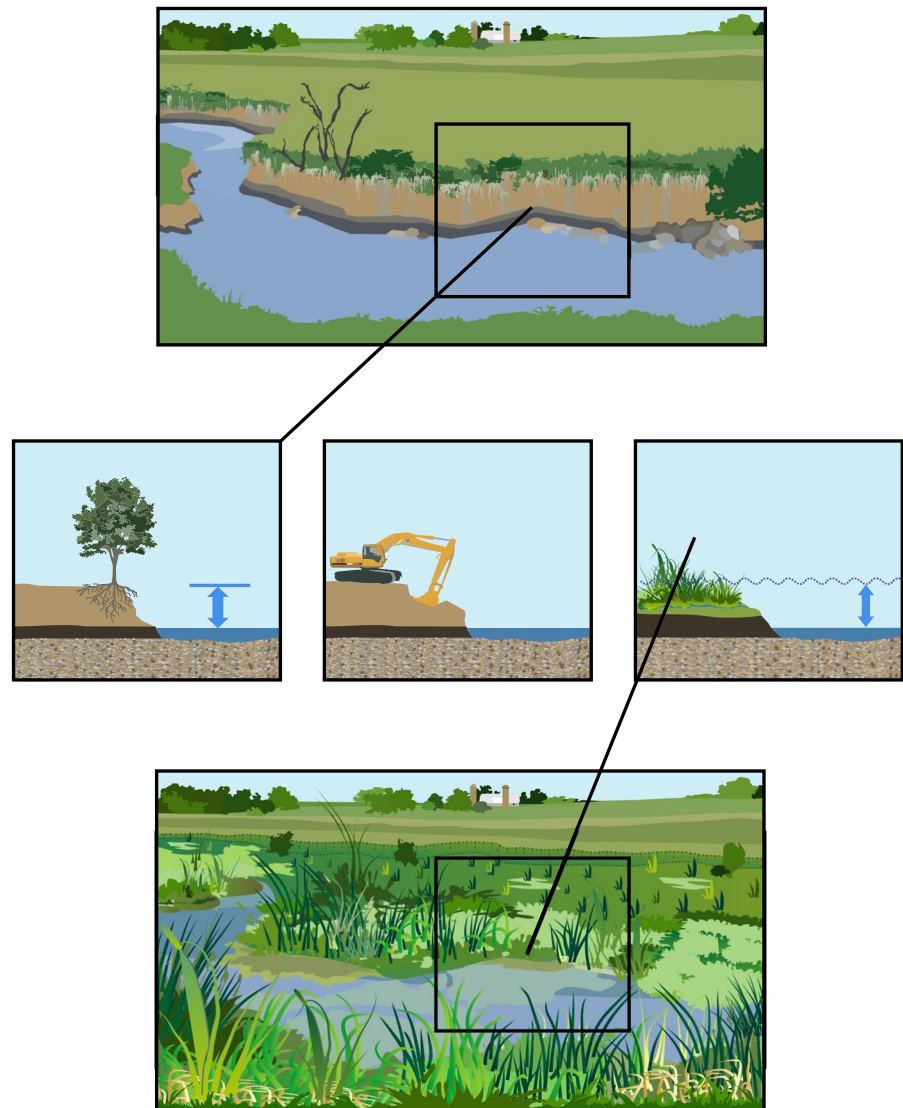
Introduction

Excess nitrogen (N) pollution from various anthropogenic sources degrades water quality in watersheds globally (Carpenter et al. 1998; Paul and Meyer 2001; USEPA 2002; Galloway et al. 2004; Compton et al. 2011). Restoration of floodplain, wetland, and stream habitats support the reduction of N pollution in watersheds (Bernhardt et al. 2007; Craig et al. 2008; Bernhardt and Palmer 2011; McMillan and Noe 2017). Connected streams and functional floodplains (Forshay and Stanley 2005; Roley et al. 2012) and riparian zones (Peterjohn and Correll 1984; Hill 1996; Mayer et al. 2007) are known to retain nitrate (NO_3^-), where retention includes the storage and removal of N in a floodplain-stream system through physical and biological pathways that decrease downstream discharge of N. However, clear evidence for restoration techniques that establish N reduction, along with predictable expectations of the controls of N retention, are needed to plan future investments in stream and floodplain restoration. Here, we report the formation of a highly retentive system, N and C changes in a restored floodplain-stream previously buried under the sediments of a once dammed stream valley bottom, and provide a description of how this restored system functions to retain N as well as the factors that influence retention in the system.

During the 18th century, rapid expansion of agriculture across the mid-Atlantic Piedmont of the United States (Jacobson and Coleman 1986) led to the deposition of alluvium and colluvium throughout riparian zone floodplains, including extensive sediment deposition in mill-dam ponds (Walter and Merritts 2008b). Ecosystems once characterized by shallow, anabranching stream networks and floodplain wetlands (Morgan 1867; Walter and Merritts 2008a; Brush 2009) that frequently flowed overbank onto broad riparian floodplains are now sediment-fill terraces buried with, up to, several meters of legacy sediment (Walter et al. 2007; Walter and Merritts 2008a). As dams were breached due to decay or intentionally removed, these thick, legacy sediment deposits dramatically transformed stream-wetland systems into persistent, incised stream channels disconnected from their floodplains when streams eroded and cut down through the sediment (Doyle et al. 2003; Walter and Merritts 2008b), which diminished nutrient processing (Stanley and Doyle 2002).

Retention of N in incised streams with floodplains laden with legacy sediment is hindered because high denitrification rates that typically occur at the sediment surface, such as in soils of a fill terrace covered floodplain (Weitzman et al. 2014), in the plant-dominated sediments in and around a channel (Forshay and Dodson 2011), or in the associated wetlands (Richardson et al. 2011; Wohl et al. 2021) are separated from NO_3^- -rich surface and shallow groundwaters in the fill terrace condition. The high stream channel walls restrict overbank flooding (Walter et al. 2013) which results in vertical and horizontal separation between a stream and its floodplain. This condition effectively limits pulses of nutrients and sediments from depositing on the floodplain (sensu Junk et al. 1989; Tockner et al. 1999), impedes much of the rapid NO_3^- removal that is possible with flooding into backwaters (e.g. Forshay and Stanley 2005), and bypasses microbially active sediments of floodplains and wetlands (Burt et al. 1999; Filoso and Palmer 2011). The incised and often scoured channels limit the accumulation of organic matter (Groffman et al. 2005), microbial biomass (Myrold and Tiedje 1985) and formation of macrophyte beds where enhanced denitrification is known to occur (Forshay and Dodson 2011). Legacy sediment has also been shown to act as a potential source of NO_3^- with relatively high potential nitrification rates, whereas buried soils, even

Fig. 1 Restoration at the Big Spring Run site included removal of legacy sediment and construction of multiple channels, leading to saturated conditions that promote re-establishment of wetlands and wetland plants. The terraced and disconnected floodplain (top panel) prevented stream and groundwater interaction in the surface soils in the pre-restoration condition. With legacy sediment removal and a dramatic change in soil elevation relative to groundwater and surface water flow, the floodplain is hydrologically connected (center, middle panel). The blue arrows (right and left center panels) represent the typical flood stage, the three layers of sediment represent the legacy sediments on top, a buried hydric layer, and basal gravels at the bottom. After floodplain reconnection and legacy sediment removal (bottom panel), the floodplain develops habitat conducive to greater and more intense biogeochemical processes and the potential for enhanced biogeochemical activity



when organic-rich, are inefficient at denitrification (Weitzman et al. 2014; Weitzman and Kaye 2017; Wade et al. 2020). However, stream restoration with removal of legacy sediment may reconnect stream-floodplain systems and form biogeochemically active sediments that allow interaction of NO_3^- -rich groundwater and surface water. Removal of legacy sediment can re-establish the hydrologic transport of NO_3^- into biogeochemically active soils, support the formation of saturated surface soils, and promote shallow groundwater or hyporheic interaction with organic-rich sediments (Fig. 1).

Legacy sediment removal in valley bottoms is a management action (Fleming et al. 2019; Altland

et al. 2020) that restores connectivity by lowering the legacy sediment terrace elevation along a stream and enables stream water to expand across the lower floodplain surface (Walter et al. 2007). Removing the sediment may also unearth historic wetlands and floodplain soils, expose wetland seed banks (Merritts et al. 2011; Wegmann et al. 2012; Walter et al. 2013), promote hydrophytic plant growth, and contribute to the accumulation of organic matter and hydric conditions (Voli et al. 2009; Voosen 2020) that can enhance N removal in the shallow groundwater and surface waters. Investigation of restoration approaches that include sediment removal are needed to better elucidate the formation of retentive habitats and influence

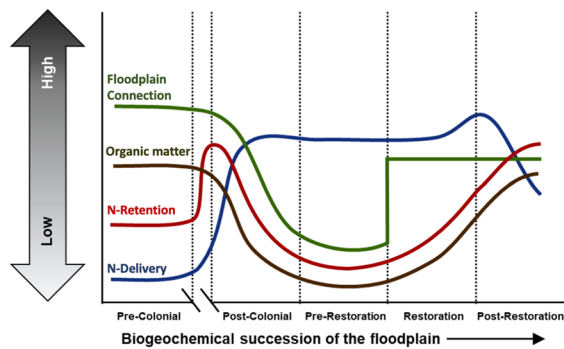


Fig. 2 Proposed floodplain connectivity and relative biogeochemical succession patterns of organic matter and nitrogen (N) over time from the pre-colonial to post-restoration periods. The period of pre-restoration represents the period of least N-retention and, effectively, the lowest soil organic matter which leads to higher N-delivery. As the legacy sediment removal restoration is implemented, connectivity is immediately restored, N-retention begins to gradually improve as organic matter accumulates and, ultimately, N-delivery decreases

of these restoration practices on nutrient delivery (Inamdar et al. 2021). (Fig. 2)

Here we evaluate how restoration of a stream-wetland system with legacy sediment removal and floodplain reconnection changes N processing and retention. We measured changes in denitrification, nitrification, ammonification, and carbon mineralization rates in soils as well as N concentrations in surface water and groundwater before and after restoration to test the hypothesis that restoration via legacy sediment removal enhances biogeochemical processing rates and N retention. We hypothesized that legacy sediment removal and the re-establishment of hydrophytic plants and subsequent increase in sediment organic matter will increase potential denitrification rates. Further, we anticipated that denitrification would be C limited immediately following restoration and, later, NO_3^- -limited as organic C content in sediment and water at the restoration site increases. NH_4^+ , and thus, nitrification rates, should decrease due to increased plant uptake and detrital organic matter accumulation increases carbon mineralization that limits oxygen availability. We describe how surface water and groundwater N concentrations are affected by biogeochemical processes in soils brought about by geomorphologic changes after restoration that support the accumulation of organic matter and soil C, and the lag times in biogeochemical

and NO_3^- response based on rates of stoichiometric change favorable for N retention. Results from this study are intended to provide insights into whether restoration of stream-wetland systems based on legacy sediment removal and stream-floodplain reconnection can support predictable biogeochemical responses that enhance N retention.

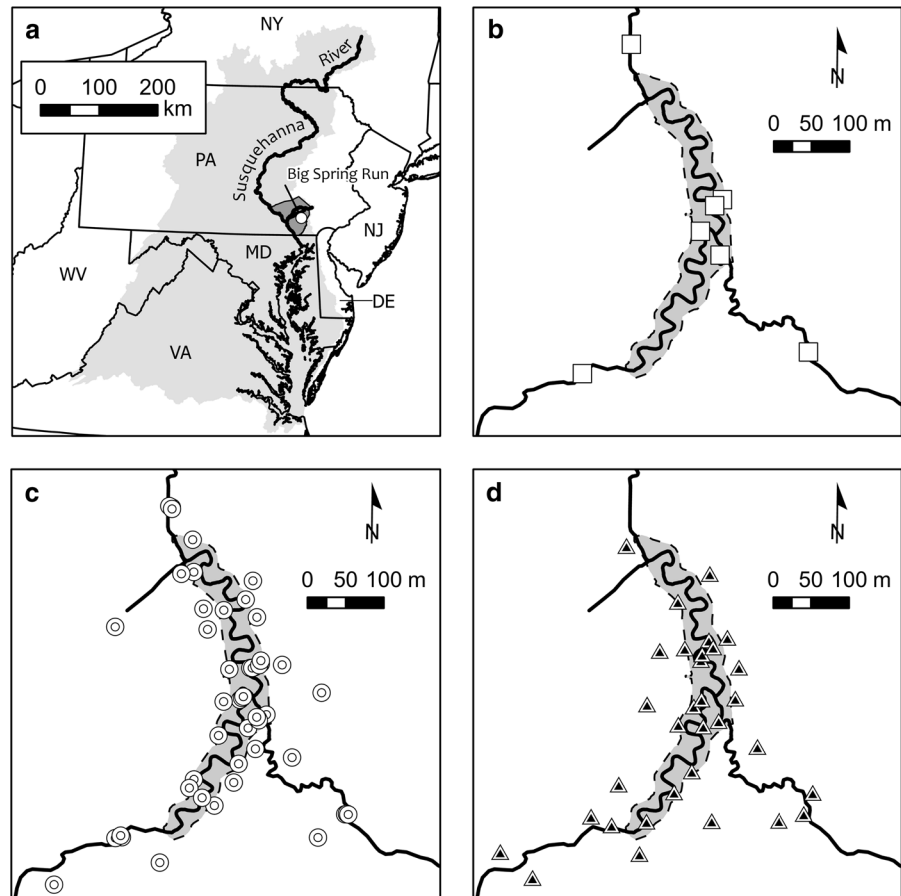
Methods

Study site

Big Spring Run (BSR) ($39^\circ 59' \text{N}$, $76^\circ 15' \text{W}$) is a northward-flowing second-order stream in Lancaster County, PA. BSR that is a tributary to Mill Creek and is a headwater in the Conestoga River watershed. The BSR watershed is a mix of agricultural and urban landuse that is typical of the low-relief mid-Atlantic Piedmont region. The Conestoga River watershed flows into the Susquehanna River, which provides ~50% of the freshwater entering the Chesapeake Bay (Schubel and Pritchard 1986; Amoros and Bornette 2002; Hirsch 2012) (Fig. 3a and Online Resource Fig. S1). The area has a humid temperate climate, with annual precipitation ~1040 mm (with highest monthly averages occurring from April through September) and annual temperatures ranging from -5 to 29°C (Langland et al. 2020). Soils along BSR are silty loams derived from the Cambro-Ordovician Conestoga limestone (Merritts et al. 2005). Along the length of BSR, incision into the legacy sediment terrace followed the breaching of a milldam during a storm in the 1930s. This incision resulted in stream banks between 1 and 3 m thick in the main channel and historic floodplain zone of BSR (Merritts et al. 2006; Walter and Merritts 2008b).

In September 2011, restoration of a portion of BSR began to test a new best management practice (BMP) based on a natural aquatic ecosystem restoration design (USEPA 2000; Hartranft et al. 2011). Specifically, to reconnect the original floodplain and restore the hydrology of the site, legacy sediment in a segment of the BSR watershed was removed from a 1.9-hectare area down to approximately the level of a historic wetland sediment layer (The BSR Project 2021). To test the restoration approach at BSR, over 20,000 t of legacy sediment was removed from an area covering ~19,000 m^2 . The valley length of

Fig. 3 The Big Spring Run (BSR) restoration study site **a** in southeastern Pennsylvania. Sample collection sites at BSR for **b** surface water, **c** groundwater, and **d** soil included locations inside (IN) and outside (OUT) of the legacy sediment removal. The gray shading around the BSR channel represents samples IN the removal area, and samples OUT of the removal area fall in the white space. The three X's on subfigures a-c represent temporary USGS gaging sites, with the northernmost downstream gage used for discharge measurements in the load estimates



the restoration was approximately 454 m, with an average depth of legacy sediment removed of 1.4 m and width of 35 m, respectively (e.g., from valley margin to valley margin along that length). Sediment was removed down to the top of the buried hydric soil, restoring an accommodation space that permitted the re-establishment of hydraulic conditions suitable for wetland development (Online Resource Fig. S2). Our analyses and previous evaluations of the natural valley bottom aquatic ecosystem characteristics buried under legacy sediment served as a guide and target for the BSR restoration design and informed the restoration of this restoration site (Walter and Merritts 2008a; Hartranft et al. 2011). Previous monitoring results indicate that stream-wetland characteristics, such as plant establishment and anastomosing channel form, were restored during the study period (Hartranft 2013). The location of the site and individual samples are located in Online Resource (Fig. 1; Table 1).

Soil sampling and analysis

Soils were collected annually at 30 sites throughout BSR restoration study area from 2010 to 2016 (Fig. 3d), excluding the restoration year of 2011. Pre-restoration soil cores were collected to depth of refusal in April 2010 over four days (5–8 April) as reported in Weitzman et al. (2014) including only the uppermost to 5–20 cm section of the surface A horizon for this study. The core sampling locations were determined using a stratified random sampling design that allowed for collection across the landscape at the BSR restoration site, characterized by an upland zone (not impacted by legacy sediment) and a legacy sediment zone near and along the stream. Post-restoration soil samples (surface only) were collected at the same locations on 6/21/2012, 4/29/2013, 7/23/2014, 3/25/2015, and 5/26/2016. While all post-restoration sampling sites were still within the general restoration site, they were further

Table 1 Soil annual mean concentrations and processing rates measured inside (IN) and outside (OUT) the Big Spring Run (BSR) legacy sediment removal area. Values within parentheses represent one standard error of the mean

	Units	2010		2012		2013		2014		2015		2016	
		IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT
Organic Matter	%	6.82 (0.23)	6.72 (0.15)	4.68 (0.38)	10.45 (0.59)	5.35 (0.42)	10.20 (0.72)	6.00 (0.43)	11.36 (0.74)	4.96 (1.17)	10.12 (0.89)	7.42 (0.41)	9.80 (0.69)
Carbon	%	2.29 (0.10)	2.11 (0.08)	1.59 (0.18)	3.72 (0.23)	1.84 (0.22)	4.20 (0.33)	2.29 (0.18)	4.34 (0.38)	2.09 (0.13)	3.32 (0.32)	2.84 (0.13)	3.75 (0.30)
Nitrogen	%	0.24 (0.01)	0.21 (0.01)	0.14 (0.02)	0.37 (0.02)	0.16 (0.02)	0.40 (0.03)	0.18 (0.02)	0.42 (0.04)	0.20 (0.01)	0.35 (0.03)	0.22 (0.01)	0.36 (0.03)
C:N	mol:mol	10.94 (0.36)	11.71 (0.26)	13.95 (0.75)	11.94 (0.20)	14.28 (0.42)	12.50 (0.58)	15.54 (0.65)	12.13 (0.14)	12.06 (0.41)	10.99 (0.26)	15.05 (0.52)	12.32 (0.31)
NO ₃ ⁻ -N	mg kg ⁻¹ soil	5.20 (0.71)	3.66 (0.49)	0.75 (0.26)	3.60 (1.26)	0.21 (0.15)	10.31 (5.90)	1.45 (0.30)	4.45 (1.14)	1.82 (0.63)	4.78 (1.42)	2.16 (0.32)	5.11 (1.19)
NH ₄ ⁺ -N	mg kg ⁻¹ soil	7.92 (0.93)	6.01 (0.54)	3.37 (0.39)	10.47 (1.37)	3.71 (0.67)	6.40 (1.28)	4.68 (0.35)	8.40 (0.88)	5.32 (0.80)	7.06 (1.06)	9.59 (0.76)	13.19 (3.44)
Nitrification	g m ² year ⁻¹	44.77 (6.89)	38.39 (7.13)	16.53 (5.49)	61.44 (8.72)	12.79 (6.61)	58.91 (12.70)	19.71 (6.35)	49.20 (7.66)	10.59 (4.00)	44.04 (6.19)	27.24 (12.98)	62.10 (12.77)
Ammonification	g m ² year ⁻¹	- 26.08 (3.78)	- 14.69 (3.34)	- 8.96 (1.27)	- 34.86 (5.33)	- 9.20 (2.51)	- 14.14 (5.06)	40.56 (4.58)	55.43 (6.10)	- 1.42 (3.22)	- 3.71 (3.63)	- 29.50 (2.75)	- 42.95 (13.66)
Carbon Mineralization	g m ² year ⁻¹	716.16 (59.72)	698.47 (90.63)	308.22 (66.74)	1117.82 (165.28)	997.41 (165.99)	1492.86 (201.40)	1073.25 (173.82)	1205.91 (165.49)	953.29 (92.79)	1072.81 (134.16)	1558.92 (154.32)	1720.38 (234.73)

categorized as inside (IN) (gray shaded area) or outside (OUT) (white area) of the legacy sediment removal area (Fig. 3). Post-restoration surface soil samples were collected to a depth of 5 cm using a hand trowel that was cleaned with water and acetone between each sampling location. Samples were placed in ice-filled coolers in the field and transported to the Kaye Biogeochemistry Laboratory at The Pennsylvania State University (PSU) in University Park, PA, USA where they were refrigerated at 4 °C. Within 6 h of collection, all soil samples were split, with ~75 g remaining at the Penn State lab for soil nutrient analysis, and ~75 g sent to the Robert S. Kerr Environmental Research Center of the U.S. Environmental Protection Agency (EPA) in Ada, OK, USA for organic matter composition determination and denitrification analysis.

The fraction of each soil sample remaining at the PSU lab was homogenized by hand and then subsampled for the following analyses: gravimetric water content (GWC), total nitrogen (N) and carbon (C), potential net N and C mineralization, and maximum potential nitrification following Weitzman et al. (2014). Briefly (see Online Resource Text S1 for details), GWC was determined by oven drying soil to constant mass, total N and C concentrations were determined by dry combustion elemental analysis, potential net N and C mineralization rates were estimated using 7-day laboratory incubations (Binkley and Hart 1989; Hart et al. 1994; Hart and Stark 1997) and colorimetric analysis for ammonium ($\text{NH}_4^+\text{-N}$) (Sims et al. 1995) and nitrate ($\text{NO}_3^-\text{-N}$) (Doane and Horwath 2003), and maximum potential nitrification (an index of nitrifier population size) was quantified using the shaken soil-slurry method (Belser 1979; Belser and Mays 1980) adapted from Hart et al. (1994). Soil fractions sent to the EPA lab were homogenized and analyzed for potential net denitrification and percent organic matter (see Online Resource Text S1 for details). Potential net denitrification bioassays were conducted using the acetylene block technique modified for sediments (Tiedje et al. 1989; Holmes et al. 1996; Groffman et al. 1999), with limitation by $\text{NO}_3^-\text{-N}$ and/or organic C determined using nutrient-amended media. The organic matter fraction of the sediment (%OM) was determined as the change in mass after combustion.

Water sampling and analysis

Water samples were collected from six stream surface locations corresponding to six shallow groundwater piezometer transects (Fig. 3b). Each of the six piezometer transects were comprised of one mid-channel piezometer and two piezometers on each side of the channel (within 5 m of the pre-restoration channel). An additional 30 shallow groundwater wells were installed to screen depths below the water table and soil surface to refusal to collect groundwater IN and OUT of the legacy sediment removal area (Fig. 3c). Piezometers and wells were sampled on a quarterly basis, excluding the period of construction in 2011. Water samples were shipped on ice to the EPA lab for analysis of $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$, which were measured via flow injection analysis, and dissolved organic carbon (DOC), which was determined on a TOC analyzer (see Online Resource Text S2 for details.) A simplified load estimate for $\text{NO}_3^-\text{-N}$ in water was calculated based on discharge at the site and the downstream nutrient concentrations based on surface water samples collected at the two most downstream sites multiplied by the mean daily discharge (USGS gaging station: 015765195) at the time of sampling. These load estimates were aggregated to compare pre- and post-restoration $\text{NO}_3^-\text{-N}$ loads in kg per day to provide a description of the effects of this restoration activity on water quality. The samples collected were not storm weighted or dictated by flow dynamics, but rather the non-storm flow conditions.

Statistical analysis

Denitrification rates were natural log transformed ($\ln x + 1$) and OM, C, and N fractions were transformed using $\sin^{-1}\sqrt{x}$ to normalize variance. Differences among sampling date, location within the legacy sediment removal (IN and OUT), and nutrient amendment were compared using a univariate model in SPSS followed by Tukey's post-hoc test with $\alpha=0.05$ on factors identified as significant in the ANOVA (IBM Corp 2017). In cases where significant two-way interactions were found, post-hoc tests of the individual single subject effects are presented to show the influence of time since restoration and nutrient treatments to summarize the main effects. Figures presented include the data and samples collected within the

legacy sediment removal area to show the magnitude of the changes from the restoration.

Water sample data were analyzed by grouping groundwater wells IN and OUT of the legacy sediment removal area and compared across years. Surface water samples were grouped based on location either as upstream or downstream of the main restoration area. Both surface water and groundwater were compared across years using univariate general linear model (GLM), ANOVA and where appropriate Tukey's HSD or compared as pre- and post-restoration using paired t-tests. Figures were prepared with R (R Core Team 2020) or SPSS (IBM Corp 2017).

Results

Our results show significant changes in sediment nutrient composition, sediment N processing rates, and water chemistry following legacy sediment removal.

Sediment chemistry and processing rates

Prior to restoration, in April of 2010, sediment nutrient composition was not generally different between samples collected IN and OUT of the legacy sediment removal area for OM, %C, extractable NO_3^- -N, extractable NH_4^+ -N, and C:N ($p > 0.05$). However, there were small but significant differences in soil %N, $0.049\% \pm 0.003$ (mean \pm s.d.) IN vs. $0.046\% \pm 0.004$ OUT ($p < 0.05$, $F_{(1, 29)} = 7.9$). Following restoration in 2011, OM, extractable NH_4^+ -N, extractable NO_3^- -N, %C, and %N were significantly less than pre-restoration levels but then steadily increased across the five-year time span to regain similarity to initial C and N stocks (Fig. 4; Table 1). However, post-restoration C:N was significantly higher than pre-restoration levels and remained high, with the exception of 2015 (Fig. 4d).

Sediment nutrient processing rates changed significantly following restoration (Fig. 5). Initial denitrification potential rates showed a significant interaction between location (IN vs. OUT: $p < 0.05$, $F_{(5, 188)} = 9.85$) and time since restoration ($p < 0.05$, $F_{(5, 188)} = 6.82$), indicating a different response to the removal of legacy sediment IN and OUT over time. Prior to restoration, denitrification potential was limited by available organic C as indicated by the

similarity in rates between C-amended and potential-amended (C + NO_3^- -N) sediments (Fig. 5d). After restoration, soils became NO_3^- -N-limited by 2016: there was no organic carbon limitation in samples collected within the legacy sediment removal area (IN) and there was an overall increase in total denitrification potential. Soil carbon mineralization rates were also significantly different across sampling years ($p < 0.05$ $F_{(6, 96)} = 1149.5$), with a pre-restoration rate of 716.2 ± 59.7 g C m^{-2} yr^{-1} IN. There was an immediate decrease in carbon mineralization after restoration (308.2 ± 66.7 g C m^{-2} yr^{-1}) with subsequent increase to an eventual high in 2016 (1558.9 ± 154.3 g C m^{-2} year^{-1}) within the legacy sediment removal area. Soil net nitrification prior to restoration showed no significant differences between IN versus OUT ($p > 0.05$, $F_{(1, 28)} = 0.36$), but samples collected within the legacy sediment removal area (IN) decreased by over 50% immediately after restoration and remained lower than pre-restoration measurements throughout the study (ANOVA $p < 0.05$, $F_{(6, 96)} = 9.23$; Fig. 5a; Table 1).

Water chemistry

The mean groundwater NO_3^- -N concentrations across the overall study area, including IN and OUT, decreased by 14% from 8.3 ± 0.2 mg N L^{-1} (mean \pm se) pre-restoration (2009 through 2011) to 7.1 ± 0.2 mg N L^{-1} post-restoration (2012 to 2016) (ANOVA $p < 0.05$, $F_{(2, 1306)} = 322$). We note that the upstream surface water NO_3^- -N concentrations decreased following restoration (Fig. 6) and caution is warranted in attribution of the magnitude of concentration changes to groundwater solely to legacy sediment removal restoration. Sample collection year and location (IN or OUT) were significant factors within the model ($p < 0.05$) with a significant interaction between annual NO_3^- -N concentration and groundwater well location, IN vs. OUT (GLM ANOVA $p < 0.01$ $F_{(7, 1306)} = 4.7$). Pre-restoration groundwater NO_3^- -N concentrations IN the sediment removal area were initially lower than OUT of the sediment removal area (6.1 ± 0.6 vs. 10.2 ± 0.5 mg N L^{-1} , respectively) ($p < 0.05$, $F_{(2, 241)} = 235$). Although concentrations IN remained lower than OUT post-restoration (7.2 ± 0.2 vs. 7.6 ± 0.3 mg N L^{-1} , respectively) ($p < 0.05$, $F_{(2, 1065)} = 925$), the difference was small. We found a significant difference among

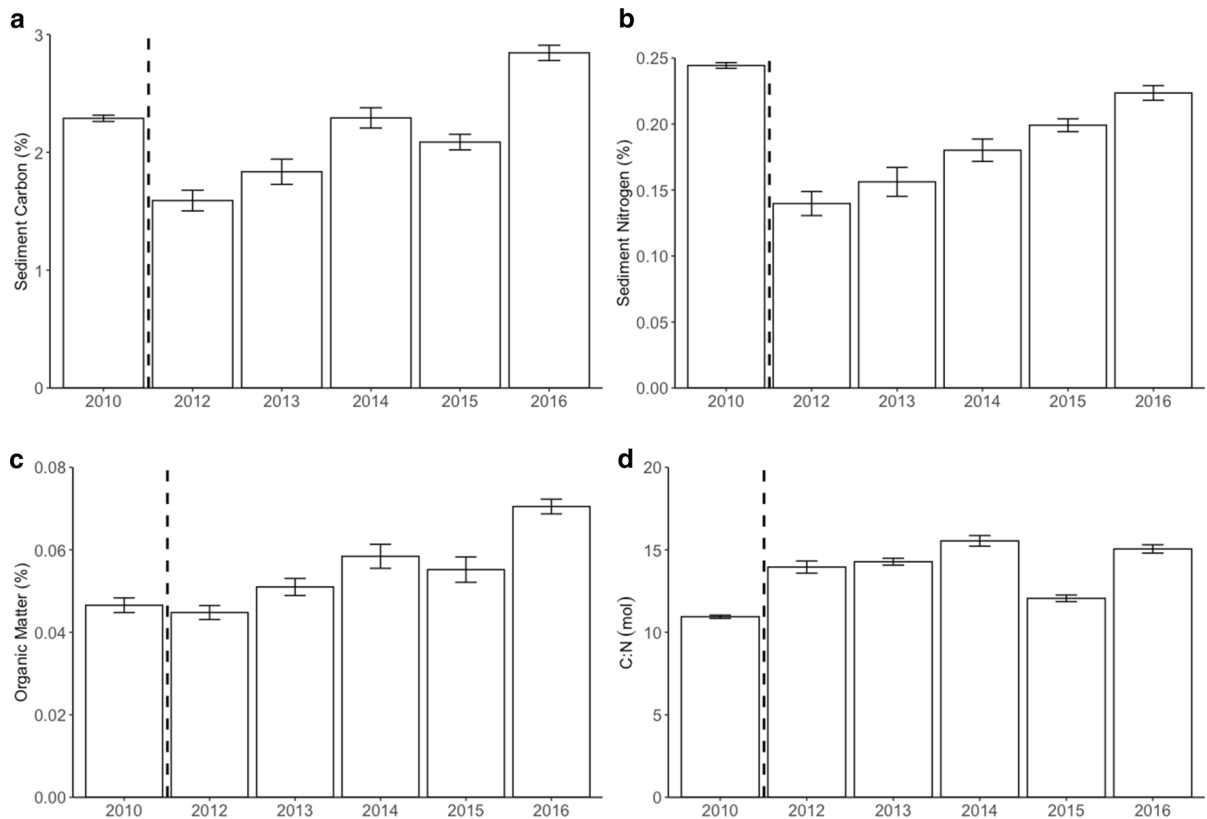


Fig. 4 Soil nutrient composition IN the Big Spring Run (BSR) legacy sediment removal area, including **a** sediment carbon, **b** sediment nitrogen, **c** organic matter fraction, and **d** carbon:nitrogen. Bars represent mean values and whiskers represent one standard error of the mean. The dotted line rep-

resents the year of restoration and legacy sediment removal, 2011, when no annual data were collected, and distinguishes between pre-restoration and post-restoration samples. See Table 1 for further detail

sampling years in groundwater NO_3^- -N concentration IN the legacy sediment removal area (ANOVA $p < 0.01$, $F_{(8, 773)} = 151.3$) as mean annual concentrations were highly variable (Fig. 6b). Despite this high variability, there was a significant decrease of 1.9 mg N L^{-1} (Table 2) in groundwater NO_3^- -N concentration observed in the final two years of study (i.e., 2015 and 2016), a 23% decrease compared to 2014 (Tukey's HSD < 0.5). Annual groundwater NO_3^- -N concentration of OUT samples of the legacy sediment removal area was also significantly different (ANOVA $p < 0.01$, $F_{(8, 533)} = 150.5$) with the greatest NO_3^- -N concentrations observed in the years prior to restoration with a maximum in 2010 of $12.0 \pm 0.9 \text{ mg N L}^{-1}$ compared to a low of $6.9 \pm 0.6 \text{ mg N L}^{-1}$ in 2015 (Fig. 6a).

Groundwater NH_4^+ -N concentration decreased significantly ($p < 0.05$, $F_{(7, 1306)} = 29.7$) following

restoration for both samples IN and OUT of the legacy sediment removal area, from a mean annual high of $71.4 \pm 18.5 \mu\text{g L}^{-1} \text{NH}_4^+$ -N in 2009 to a low of $39.0 \pm 14.5 \mu\text{g L}^{-1} \text{NH}_4^+$ -N in 2015 and $7.1 \pm 26.1 \mu\text{g L}^{-1} \text{NH}_4^+$ -N in early 2016 (Table 2). No significant differences in NH_4^+ -N concentrations were observed between well locations IN or OUT, nor between location and year (Table 2).

Groundwater DOC was highly variable across sites and years with significant interactions between location and year IN and OUT of the legacy sediment removal area (GLM ANOVA $p < 0.05$, $F_{(7, 1325)} = 2.3$; Table 2). The maximum annual DOC of $2.6 \pm 0.5 \text{ mg C L}^{-1}$ was observed IN the legacy sediment removal area during the abbreviated sampling immediately prior to restoration in 2011, whereas the minimum of $1.2 \pm 0.2 \text{ mg C L}^{-1}$ was observed OUT of the legacy sediment removal area during pre-restoration in

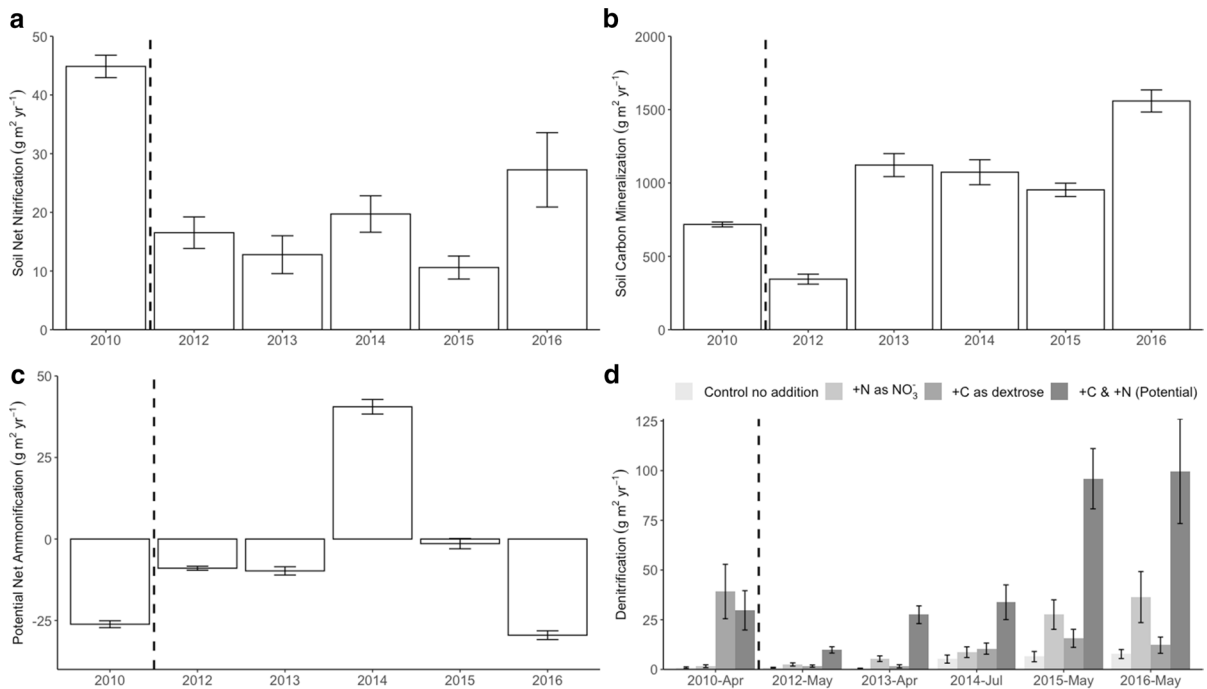


Fig. 5 Soil processing rates IN the Big Spring Run (BSR) legacy sediment removal area, including **a** soil net nitrification, **b** soil carbon mineralization, **c** potential net ammonification, and **d** denitrification by acetylene block with amendments in a factorial design of control (no addition), with nitrogen (N) as nitrate (NO_3^-), with carbon (C) as dextrose, and with C+N

(potential). Bars represent mean values and whiskers represent one standard error of the mean. The dotted line represents the year of restoration and legacy sediment removal, 2011, when no annual data was collected, and distinguishes between pre-restoration and post-restoration samples

2009. Groundwater DOC concentration from wells inside the legacy sediment removal area varied annually (ANOVA $p < 0.01$, $F_{(8,781)} = 6.67$) with the lowest concentrations of $1.2 \pm 0.1 \text{ mg C L}^{-1}$ observed the year after restoration in 2012, which then increased to a high of $1.4 \pm 0.1 \text{ mg C L}^{-1}$ by 2015. The DOC concentrations OUT of the legacy sediment removal area were similarly variable with significant differences between years (ANOVA $p = 0.04$, $F_{(8,544)} = 2.1$).

Surface water NO_3^- -N concentration varied depending on sampling locations (Fig. 6c, d; Table 3). The greatest NO_3^- -N concentrations were observed in the East Branch of BSR (SW 789 and SW 131,415) with overall mean concentrations exceeding 11.0 mg N L^{-1} . Surface water NO_3^- -N in the most downstream locations (SW 123 and SW 456 and SW 192,021: note that SW 192,021 is a relocated site within 20 m of SW 456 after channel modification) showed a significant decrease following restoration (ANOVA $p < 0.01$, $F_{(7,61)} = 1006.5$) with a mean of $10.9 \pm 0.6 \text{ mg NO}_3^-$ -N L^{-1} in 2010 decreasing to

$8.6 \pm 0.8 \text{ mg N L}^{-1}$ in 2015 and $8.4 \pm 1.4 \text{ mg L}^{-1}$ in 2016, a 23% difference. Please note that the upstream NO_3^- -N decreased following restoration (Fig. 6) and caution is warranted in attribution of the concentration changes solely to legacy sediment removal restoration (Table 3).

Similar to groundwater patterns, the highest concentration of surface water NH_4^+ -N were observed prior to restoration in 2009 (mean annual = $13.3 \pm 1.1 \mu\text{g NH}_4^+$ -N L^{-1}). These concentrations decreased dramatically over the years following restoration to a low of $0.7 \pm 2.4 \mu\text{g L}^{-1}$ mean annual concentration in 2016. Surface water NH_4^+ -N concentrations were significantly different across years (GLM ANOVA $p < 0.01$ $F_{(6,140)} = 27.5$), but notably not different between sampling locations or position up or downstream within the site $p > 0.05$ (Table 3).

The highest surface water DOC concentrations occurred immediately after restoration in 2012 and 2013 at 1.1 ± 0.1 and $1.2 \pm 0.1 \text{ mg C L}^{-1}$, respectively,

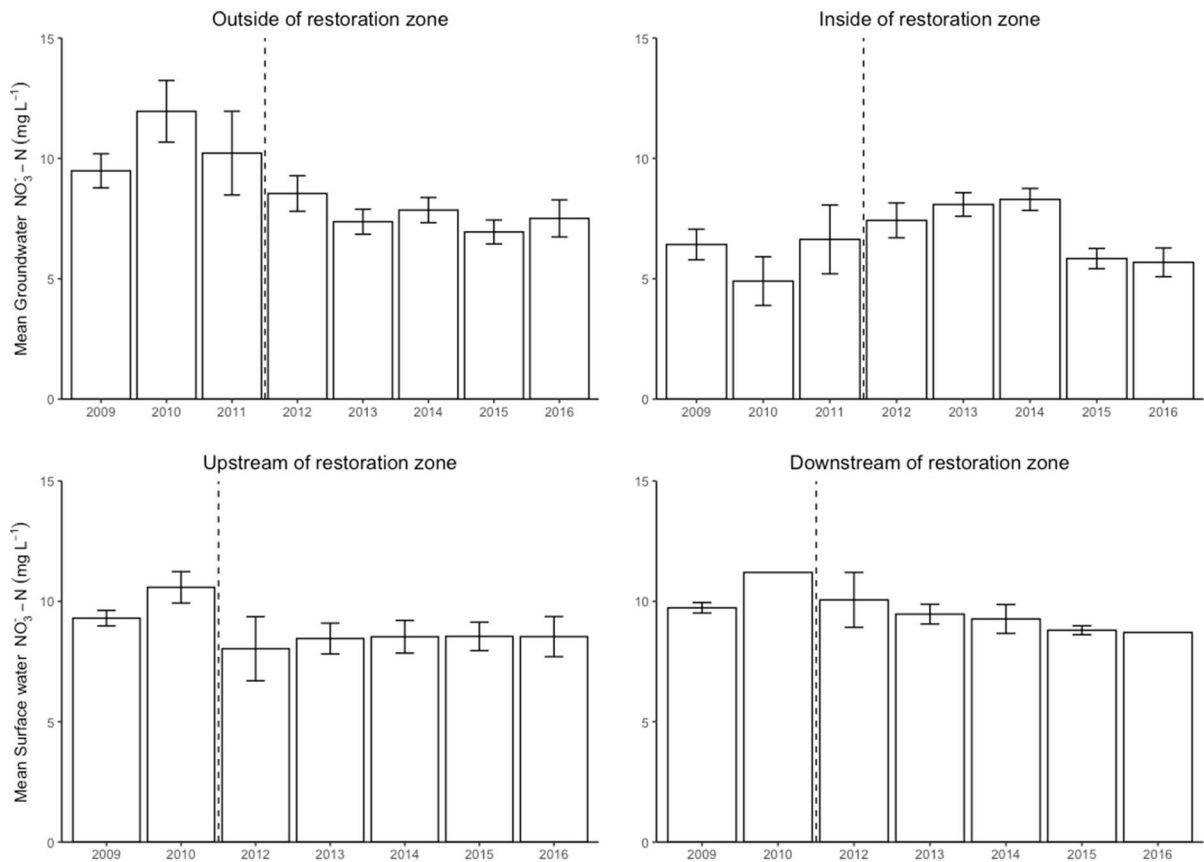


Fig. 6 Mean annual nitrate (NO_3^- -N) concentrations for groundwater **a** outside (OUT) and **b** inside (IN) and surface water **c** upstream and **d** downstream of the Big Spring Run (BSR) legacy sediment removal area. Bars represent mean val-

ues and whiskers represent one standard error of the mean. The dotted line represents the year of restoration and legacy sediment removal, 2011, when no data were collected, and distinguishes between pre-restoration and post-restoration samples

then fell to the lowest concentrations in the final years of observations in 2015 and 2016 to 0.9 ± 0.1 and $0.6 \pm 0.2 \text{ mg C L}^{-1}$, respectively. Surface water DOC concentrations were significantly different across years (GLM ANOVA $p < 0.01$, $F_{(6, 186)} = 3.2$; Table 3) and between individual sample sites, but no significant interactions were found between factors.

The highest molar ratios of organic C to inorganic N (C:N) in surface water occurred in the years immediately after restoration, culminating in an annual high mean of 0.46 ± 0.06 (C:N \pm s.e.) in 2012, while the lowest observed molar ratios of 0.10 ± 0.08 occurred in 2015 and 2016 (Tukey's HSD < 0.5). There were significant differences over time (GLM ANOVA $p < 0.01$, $F_{(6, 136)} = 4.5$) and between sampling location ($p < 0.01$, $F_{(6, 136)} = 12.9$; Table 3). The highest C:N was observed in the western branch of

BSR with a mean of 0.48 ± 0.05 and the lowest ratios were observed in the eastern branch with a mean of 0.06 ± 0.05 .

Nitrate loads in stream water at BSR changed significantly over time with a drop in total load in the years immediately following restoration ($48.8 \pm 3.6 \text{ kg day}^{-1}$) compared with load estimates before restoration ($53.3 \pm 3.7 \text{ kg day}^{-1}$) ($p < 0.01$, $F_{(1, 57)} = 154.8$). When comparing years ($p < 0.01$, $F_{(7, 54)} = 66.0$) the load decreased and remained lower throughout the post-restoration period, except for 2014 (Fig. 7) with the lowest loads occurring in 2015 at $38.1 \pm 3.7 \text{ kg day}^{-1}$. These loads are based on the mean discharge (Online Resource Fig. S3) and NO_3^- -N measured at our two most downstream sites on sampling dates. It should be noted that the upstream NO_3^- -N concentrations decreased following restoration (Fig. 6) and

Table 2 Groundwater chemistry annual mean values measured inside (IN) and outside (OUT) the Big Spring Run (BSR) legacy sediment removal area

	2009		2010		2011		2012		2013		2014		2015		2016	
	IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT	IN	OUT
DOC (mg L ⁻¹)	1.49 (0.13)	1.17 (0.12)	1.93 (0.31)	1.65 (0.39)	2.62 (0.99)	1.07 (0.25)	1.17 (0.11)	1.56 (0.27)	1.38 (0.08)	1.74 (0.15)	1.30 (0.07)	2.11 (0.29)	1.41 (0.07)	1.88 (0.29)	1.21 (0.09)	1.34 (0.23)
NO ₃ ⁻ -N (mg L ⁻¹)	6.42 (0.64)	9.48 (0.71)	4.90 (1.01)	11.95 (1.28)	6.63 (1.43)	10.22 (1.74)	7.42 (0.72)	8.54 (0.74)	7.94 (0.44)	7.40 (0.47)	8.30 (0.46)	7.85 (0.52)	5.84 (0.42)	6.94 (0.50)	5.68 (0.60)	7.51 (0.77)
NH ₄ ⁺ -N (µg L ⁻¹)	78.82 (15.34)	64.06 (22.65)	48.04 (18.08)	49.27 (36.92)	28.5 (21.26)	118.71 (71.44)	9.62 (2.19)	78.09 (27.71)	15.66 (1.93)	93.31 (29.30)	32.52 (8.07)	129.21 (57.61)	23.25 (5.54)	54.80 (20.28)	8.79 (2.94)	5.35 (3.22)
C:N (mol:mol)	13.89 (6.42)	1.82 (1.25)	29.06 (24.59)	64.73 (60.55)	13.27 (10.02)	2.44 (1.92)	29.09 (10.99)	9.61 (8.25)	24.11 (13.21)	13.62 (6.88)	6.33 (2.16)	3.94 (1.15)	19.75 (5.69)	6.71 (3.49)	5.17 (2.21)	1.50 (0.94)

Values within parentheses represent one standard error of the mean

caution is warranted in attribution of the magnitude of load or concentration changes solely to legacy sediment removal restoration. These individual load estimates are based on discharge and concentration on sampling days. The samples collected do not represent storm weighted loading and are heavily influenced by the episodic nature of sample collection and discharge of the date collected. This approach represents the non-storm flow conditions during our sampling events and provides an indicator of the effects of the restoration.

Discussion

Unearthing buried stream-wetlands and reconnecting a formerly incised stream with its floodplain as part of a legacy sediment removal restoration approach enhances N retention and decreases nitrogen concentrations in groundwater NO₃⁻-N and NH₄⁺-N within the immediate vicinity of legacy sediment removal and also decreases surface water NO₃⁻-N and NH₄⁺-N downstream of the restoration area. These changes appear to be driven by a combination of increased denitrification and plant uptake as well as a decrease in localized loading from changes in land use due, in part, to improved stream-floodplain connection with groundwater and surface water and accumulation of organic matter in the floodplain.

The soil measurements at BSR demonstrated an apparent stoichiometric control of denitrification (Hedin et al. 1998) particularly within the legacy sediment removal area. The site dramatically shifted from a system starved of organic C, the terminal electron donors for denitrification, to a sediment matrix capable of denitrifying excess NO₃⁻-N as the terminal electron acceptor (Fig. 5d). The observations made at BSR support the hypothesis that when organic matter accumulates over time (Fig. 4c) and interacts over a biogeochemically active plant sediment matrix, restoration can drive a system to shift past a state of excess NO₃⁻-N with low organic C, to lower NO₃⁻-N with higher C:N due to higher processing rates under conditions of high C availability (Taylor and Townsend 2010). This shift of organic matter accumulation over the course of several years agrees with our observations of soil carbon mineralization and denitrification potential (Fig. 5b and d, respectively). Initially, these values dropped to below pre-restoration rates

Table 3 Surface water chemistry annual mean values measured at different positions relative to the Big Spring Run (BSR) legacy sediment removal area

Stream position	Sample ID		2009	2010	2012	2013	2014	2015	2016	
Downstream	SW 123	DOC (mg L ⁻¹)	0.98 (0.05)	0.75 (0.08)	0.71 (0.14)	1.11 (0.14)	1.16 (0.23)	0.88 (0.03)	0.78 (0.12)	
		NO ₃ ⁻ -N (mg L ⁻¹)	9.73 (0.22)	11.20 –	10.06 (1.14)	9.49 (0.34)	9.27 (0.60)	8.82 (0.22)	8.70 (0.01)	
		NH ₄ ⁺ -N (µg L ⁻¹)	8.00 (0.00)	7.00 –	7.04 (0.30)	15.84 (4.06)	10.81 (6.23)	2.66 (1.53)	0.68 (0.00)	
		C:N (mol:mol)	0.12 (0.01)	0.07 –	0.09 (0.03)	0.14 (0.02)	0.16 (0.05)	0.12 (0.01)	0.11 (0.02)	
		SW 192021	DOC (mg L ⁻¹)	N/A –	N/A –	0.68 (0.14)	1.13 (0.14)	1.10 (0.18)	0.89 (0.04)	0.67 (0.02)
		NO ₃ ⁻ -N (mg L ⁻¹)	N/A –	N/A –	9.81 (1.29)	8.93 (0.31)	9.03 (0.52)	8.46 (0.19)	8.06 (0.09)	
		NH ₄ ⁺ -N (µg L ⁻¹)	N/A –	N/A –	5.01 (1.49)	11.50 (2.61)	9.56 (5.00)	1.14 (0.00)	0.68 (0.00)	
		C:N (mol:mol)	N/A –	N/A –	0.08 (0.03)	0.15 (0.02)	0.15 (0.03)	0.10 (0.02)	0.10 (0.00)	
		SW 456	DOC (mg L ⁻¹)	0.94 (0.07)	0.76 (0.13)	N/A –	N/A –	N/A –	N/A –	N/A –
			NO ₃ ⁻ -N (mg L ⁻¹)	9.37 (0.16)	10.60 –	N/A –	N/A –	N/A –	N/A –	N/A –
			NH ₄ ⁺ -N (µg L ⁻¹)	13.71 (3.71)	7.00 –	N/A –	N/A –	N/A –	N/A –	N/A –
			C:N (mol:mol)	0.12 (0.01)	0.07 –	N/A –	N/A –	N/A –	N/A –	N/A –
	Midstream	SW 101112	DOC (mg L ⁻¹)	1.18 (0.03)	1.23 (0.14)	2.16 (0.97)	1.66 (0.21)	1.80 (0.21)	1.43 (0.09)	1.03 –
			NO ₃ ⁻ -N (mg L ⁻¹)	7.55 (0.11)	7.86 (0.90)	3.59 (2.02)	5.31 (0.43)	4.73 (0.45)	4.74 (0.14)	5.16 (0.39)
			NH ₄ ⁺ -N (µg L ⁻¹)	11.67 (2.38)	7.00 (0.00)	8.58 (2.42)	14.76 (4.64)	9.56 (3.67)	4.70 (1.91)	0.68 (0.00)
C:N (mol:mol)			0.18 (0.01)	0.19 (0.05)	1.28 (1.04)	0.40 (0.09)	0.48 (0.09)	0.36 (0.03)	0.22 –	
		SW 789	DOC (mg L ⁻¹)	0.64 (0.06)	0.76 (0.16)	0.91 (0.37)	0.72 (0.12)	0.69 (0.11)	0.52 (0.01)	0.44 (0.02)
			NO ₃ ⁻ -N (mg L ⁻¹)	11.18 (0.23)	11.83 (0.22)	10.50 (0.30)	11.22 (0.26)	11.88 (0.39)	11.17 (0.00)	10.13 (0.18)
			NH ₄ ⁺ -N (µg L ⁻¹)	18.00 (3.87)	7.00 (0.00)	4.67 (2.02)	10.54 (3.09)	5.41 (3.27)	1.14 (0.00)	0.68 (0.00)
			C:N (mol:mol)	0.07 (0.01)	0.07 (0.02)	0.10 (0.04)	0.08 (0.01)	0.07 (0.01)	0.05 (0.00)	0.05 (0.00)
Upstream	SW 131415	DOC (mg L ⁻¹)	0.65 (0.08)	0.68 (0.14)	0.43 (0.07)	0.78 (0.11)	0.65 (0.13)	0.50 (0.03)	0.42 (0.04)	
		NO ₃ ⁻ -N (mg L ⁻¹)	11.47 (0.34)	12.53 (0.46)	12.50 (1.20)	12.03 (0.35)	12.60 (0.16)	11.77 (0.36)	10.80 (0.10)	

Table 3 (continued)

Stream position	Sample ID	2009	2010	2012	2013	2014	2015	2016
	NH ₄ ⁺ -N (μg L ⁻¹)	14.43 (3.39)	7.00 (0.00)	1.20 (0.00)	4.20 (0.78)	5.08 (2.43)	1.14 (0.00)	0.68 (0.00)
	C:N (mol:mol)	0.07 (0.01)	0.06 (0.02)	0.04 (0.01)	0.08 (0.01)	0.06 (0.01)	0.05 (0.00)	0.05 (0.01)
	DOC (mg L ⁻¹)	1.46 (0.14)	1.16 (0.18)	1.89 (0.75)	2.02 (0.31)	1.69 (0.24)	1.37 (0.17)	N/A –
	NO ₃ ⁻ -N (mg L ⁻¹)	7.24 (0.16)	9.13 –	3.76 (2.30)	4.79 (0.46)	4.40 (0.88)	4.63 (0.30)	N/A –
	NH ₄ ⁺ -N (μg L ⁻¹)	14.13 (4.56)	7.00 –	2.64 (1.44)	10.92 (2.84)	8.43 (3.11)	3.52 (2.3850)	N/A –
	C:N (mol:mol)	0.24 (0.02)	0.12 –	1.16 (0.94)	0.58 (0.18)	0.63 (0.19)	0.39 (0.08)	N/A –

. The midstream positions represent samples collected inside (IN) the legacy sediment removal area, while the downstream and upstream positions represent samples collected outside (OUT) the legacy sediment removal area. Values within parentheses represent one standard error of the mean. Note that SW 456 was replaced with SW 192,021 within 20 m of the same location due to construction see Online Resource Text S2 for details

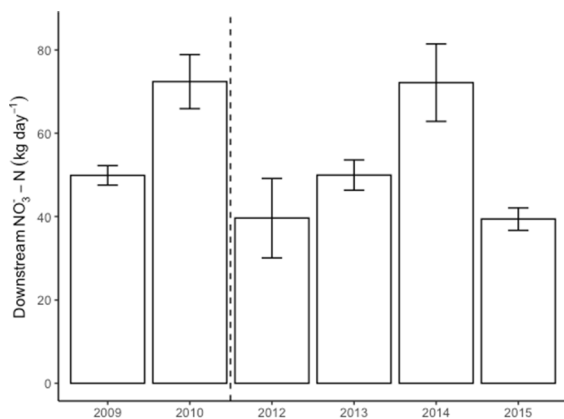


Fig. 7 Nitrate (NO₃⁻-N) loads measured at the most downstream surface water sites at Big Spring Run (BSR). Bars represent mean values and whiskers represent one standard error of the mean. These are calculated loads based on individual observed concentrations and discharge at the most downstream locations. The dotted line represents the year of restoration and legacy sediment removal, 2011, when no data were collected, and distinguishes between pre-restoration and post-restoration samples

after legacy sediment removal and restoration when organic material was lacking (Fig. 5b and d), but then climbed to substantially higher rates with a corresponding drop in NO₃⁻-N concentration years after restoration due to higher organic C relative to inorganic N.

With legacy sediment removal we also observed a decrease in nitrification rates (Fig. 5a), with an overall increase in microbial activity indicated by soil carbon mineralization rates (Fig. 5b) that decreases the internal loading of NO₃⁻-N and prevent de novo NO₃⁻ production. Prior to legacy sediment removal, most of the biogeochemically active soils were separated from the NO₃⁻-N carried by surface and groundwater. These higher and drier sediment terraces in the pre-restoration condition tended to support greater nitrification rates which act as an additional source of NO₃⁻-N pollution (Fig. 5a) (Groffman et al. 2002, 2003; Weitzman et al. 2014; Weitzman and Kaye 2017). After sediment removal, the floodplain elevation was lowered and became hydrologically reconnected to the main channel, which created saturated conditions and introduced plant material conducive to organic C accumulation (Fig. 4c) and higher denitrification potential (Fig. 5d).

These sediment processes and water chemistry dynamics suggest that the combination of hydrologic connectivity, NO₃⁻-N, and accumulation of organic matter from allochthonous and autochthonous sources controls NO₃⁻-N retention in the BSR stream-wetland system. This relationship is likely due to the more saturated conditions of the sediments, which along with plant material (Online Resource Fig. S4) support greater denitrification in the soils and sediment (Forshay and Dodson 2011), but also slows aerobic

respiration and consumption of organic matter. In this system, denitrification is enhanced after legacy sediment removal through the reconnection of streams to their floodplains and associated wetlands (Fink and Mitsch 2007; Hammersmark et al. 2008; Harrison et al. 2014; Palmer et al. 2014). BSR is at the confluence of two streams in the restoration reach, one of which was generally high in NO_3^- -N (East Branch) while another was relatively high in organic carbon (West Branch). This combination of streams that meet at a more biogeochemically active floodplain instead of trapped between deeply incised channel banks also helped show the dramatic effects of legacy sediment removal on biogeochemical processes, particularly in the subsurface. Studies elsewhere that describe restorations resulting in stream-wetland systems also showed significant N reductions and, in particular, Filoso and Palmer (2011) observed that during periods of high stream flow, only those restoration projects that converted lowland streams to “stream-wetland complexes” were effective at reducing N fluxes because of the “spillover” of stream flow onto adjacent floodplains and wetlands. In a synthesis of N removal across global studies of stream restoration, Newcomer Johnson et al. (2016) found best results among strategies that reconnected floodplains and streambeds and increased the reactive surface area of flowing water and connected wetlands. The restoration at BSR appear to follow a similar pattern of reconnection of stream to floodplain wetlands resulting in greater N reduction potential.

Our results show that denitrification and retention are enhanced, particularly when surface soils are placed in contact with N-rich surface and groundwater, with legacy sediment removal in a restored area, but research is needed to test this approach in other legacy sediment-rich watersheds to determine the scalability and effective longevity of this restoration approach. Selection of locations with buried Holocene wetlands or floodplains in wet valley bottoms also may be important factors in predicting restoration success when development of nitrogen retention is desired because these soils, when functional, are particularly good at denitrification, even in modern systems with high NO_3^- -N loads (Fig. 5d). Regardless, likely success factors for enhancing N retentive processes in floodplains buried by legacy sediment here include removing the legacy sediment, lowering the floodplain, creating the proper hydrology, and

reestablishing surface water and groundwater connections because these are known drivers of beneficial biogeochemical processes (Parola and Hansen 2011; Wohl et al. 2021). These results may not be typical in systems where there was no buried wetland or wet valley bottom, or where a site is not engineered to support stable, valley bottom wetland characteristics. Whether the relict wetland soil microbial community was resurrected to support the observed biogeochemical processes or a new community developed under the favorable conditions created by the legacy sediment removal and floodplain restoration is outside of the scope of this study, but observable N retentive processes did develop in the soils. Restoration of the conditions that formed the relict wetland soils in a contemporary floodplain stream-wetland also support high denitrification rates in contemporary high N systems. The continuous accumulation of organic material in the sediments coinciding with the establishment of plants in the system (Online Resource Fig. S4) are likely critically linked to restoration success and longevity but changes to soil composition can take years to sufficiently relieve organic C limitation of denitrification as well as other biogeochemical processes (Fig. 5). We did not observe improvements to NO_3^- -N retention or denitrification until nearly five years after restoration, probably due to the initial lack of available C for microbial activity and gradual accumulation of soil OM, C, and N (Fig. 4) that eventually came to support higher biogeochemical processes rates, like denitrification.

Increasing soil rates of biogeochemical N retention processes, like denitrification, depend upon geomorphic and hydrologic conditions that enhance organic matter availability (Hedin et al. 1998) as well as contact with N-rich waters. Establishment of saturated conditions can lead to slower aerobic respiration and accumulation of organic matter. This combination is critical because the stoichiometric excess of C relative to NO_3^- -N is associated with decreased NO_3^- -N concentrations in surface and groundwater and higher processing rates (Taylor and Townsend 2010). It is likely that stream restoration activities that, like legacy sediment removal, support conditions conducive to N retention may experience a time lag as a common characteristic while organic matter and microbial biomass accumulates (Hamilton 2012) (Fig. 4). In some cases, stream and floodplain restoration may fail to generate the appropriate conditions

for enhanced denitrification (e.g. Orr et al. 2007) or the duration of observation may be insufficient to observe enhanced retention (McMahon et al. 2021). A functional stream-floodplain system and associated wetlands should retain N and promote relatively high denitrification rates when organic matter accumulates, reduced conditions form that inhibit nitrification, and N-rich waters meet.

If we consider the historic biogeochemical succession of the historical floodplain stream-wetland at BSR that experienced colonial land development and damming (Fig. 2), we can begin to evaluate the relationships and controls of N retention in similar systems to understand how a combination of geomorphic stream restoration and floodplain reconnection may function under modern restoration. The pre-colonial connectivity between stream and floodplain was likely quite high, with beaver and treefall aggrading organic matter and causing diverse hyporheic flow in the near stream and valley bottom (Naiman et al. 1988; Briggs et al. 2013). The soils were hydric and organic matter was high. N retention was likely limited by N availability that was controlled by mineralization and small amounts of allochthonous N (relative to current inputs), which led to high potential retention and ultimately low N delivery downstream. As trees were removed, land was developed or cleared, soil aggraded in channels and the delivery of N to the stream may have begun to increase along with N retention to potentially exceed initial retention rates with the installation of a mill dam, but as the dam silted in, dams failed and incised channels formed. The surface soils became aerobic and disconnected from both the stream and shallow groundwater. N delivery to these incised streams could no longer retain N and the most organic matter and carbon rich sediments were buried or existed at an elevation that likely supported more nitrification than denitrification. In effect the floodplain was buried and disconnected, with diminishing N retention and minimal accumulation of organic matter the stream simply conveyed any excess N downstream. With the onset of intensive agriculture and fertilizer use, concentrations of NO_3^- -N increased substantially in the Piedmont and across the world in both surface and groundwaters over time (Galloway et al. 2004) to levels that far exceed what would have been experienced prior to burial. With the implementation of restoration approaches like legacy sediment removal

that lower bank elevation, connect and restore wetland floodplain to their streams allowing organic matter to accumulate allowing retention to improve on the floodplain. Based on our observations of organic matter accumulation and resulting NO_3^- -N reduction under contemporary high NO_3^- -N concentrations, the removal of legacy sediments and restoration can be effective at producing N retentive soils via denitrification but will take time to develop. During this transition we observed that nitrification did decrease immediately but decreases in groundwater NO_3^- -N concentrations did not occur until denitrification was no longer C limited. Floodplains restored via legacy sediment removal have the potential to regain their original function, and, in conditions of high contemporary NO_3^- -N concentrations, do promote N retention that ultimately exceeds historical rates that may decrease N delivery downstream. The site observed here is a historic valley bottom with clearly documented evidence of hydric soils and wetland conditions (Hartranft et al. 2011). Other floodplain restoration sites, for example, those that may have dammed a more energetic or steeper system, may not develop the same enhancement of N retention or may not support the plant communities or hydrological characteristics needed to accumulate organic material in a way that allows NO_3^- -N-rich waters to interact with these biogeochemically retentive soils, resulting in a different biogeochemical succession following restoration. It is also possible that N inputs to this site may eventually overwhelm the retention capacity of the system (Bertot and Dodds 2005) or shift the system out of the current retentive state (Peterjohn et al. 1996). Clearly, further observation at BSR and additional studies at other sites are warranted to better characterize and predict future outcomes.

Our results show changes in groundwater concentration throughout the restoration area both outside and inside the legacy sediment removal zone after restoration (Fig. 7), which could be due to the effects of surface planting, infiltration or changes in practices on the immediate surface outside of the legacy sediment removal area in addition to the clear evidence we provide of improved denitrification and decrease in nitrification (Fig. 5a) in the legacy sediment removal area. The dynamics of concentration observed in groundwater suggests that the potential for complex surface and groundwater relationships occur at BSR, as in many stream-floodplain systems,

will ultimately require careful consideration in evaluation of efficacy long term.

In this study we observed stream concentrations change in the restoration area of BSR, but limitations of sample frequency and design limit the certainty of the magnitude of N retention estimates. Estimates of total retention or load can be difficult to observe or quantify on even a stream reach basis (McMahon et al. 2021). At BSR we observed that external inputs to the system are dynamic as upstream N in surface water decreased immediately following restoration and a similar pattern occurred in downstream waters (Fig. 6c, d). Further, BSR is composed of two tributaries that differ in N composition and hydrology that add complexity to downstream evaluation of N retention (Table 3). We do see that there is a change in NO_3^- -N load following restoration (Fig. 7) which may be driven by upstream changes. The load estimate approach, based on relatively low flow episodic samples, may show how legacy sediment removal and a restored stream-wetland systems can enhance N retention but discerning the actual load reduction across hydrologic events is potentially obscured by diverse flow and load inputs from surface and groundwater. For example, in another study, storm and discharge triggered event estimates above and below this restoration site showed little change in N loads before and after restoration (Langland et al. 2020). The Langland study shows that there is little effect on load observations during the triggered flows of storm or runoff events on the ascending limb which could be dominated by overland flow, flushing effects, and larger watershed N sources entering the system. Considering both the simplified regular flow load estimates with change in the upstream NO_3^- -N observations following restoration made in this study and the obvious limitations of event driven load studies of NO_3^- -N by others, suggests that careful consideration of expectations and measurement approaches are needed to accurately represent the benefits of legacy sediment removal and any restoration practice that intends to enhance N retention. Because of the complex nature of flow and load estimates with respect to discharge and N concentrations, the management approaches that account for size, groundwater flow, uptake, and denitrification estimates within a hyporheic zone are likely to provide better estimates of N retention than load based estimates during flow events and more closely reflect a conservative estimate of the

total N retained or attributable to restoration based on legacy sediment removal (Altland et al. 2020).

Thousands of streams in the Mid-Atlantic Piedmont of the United States have Holocene floodplain-wetlands buried by legacy sediments (Walter and Merritts 2008b). Many of these streams experience modern high NO_3^- -N loads that can be addressed with restoration to retain pollution, but these practices still require continued study to ensure that the desired outcomes are achieved. Investigation and observation to understand how restoration investments can best function as pollution sinks and development of a better understanding of design limitations and long-term efficacies should bolster the growth of stream-wetland and floodplain restoration into the future.

Conclusion

Reconnecting streams and floodplains through legacy sediment removal, which promotes floodplain-wetland development, can increase NO_3^- retention and decrease downstream N loads. The critical factors that can lead to higher NO_3^- retention are: (1) restoring hydrologic regimes conducive to floodplain-wetland development; (2) lowering surface soil and sediment elevations relative to water level to form wetter conditions that facilitate hyporheic exchange and delivery of NO_3^- -rich water to biogeochemically active floodplain sediments; (3) plant and organic matter development to support organic carbon accumulation in wet sediments; and (4) time for the formation of stoichiometrically favorable conditions (high C:N) for denitrification that overcome an apparent threshold of organic carbon limitation. The restoration shifted the system into NO_3^- limitation with high denitrification potential, decreased nitrification, and enhanced NO_3^- retention. Based on these observations, one should expect similar results in similar situations, but attention should be given to the hydrological, biogeochemical, geomorphological, and temporal factors that control these ecological processes when evaluating a site or monitoring progress for successful NO_3^- retention or deeming a restoration successful (Parola and Hansen 2011; Hawley 2018). It is likely that floodplain reconnection efforts that fail to achieve the wetted conditions, biogeochemical nexus of organic C and NO_3^- , or attainment of

favorable stoichiometric conditions over time for denitrification will not achieve high NO_3^- load reductions. As a corollary, incorporating these factors of NO_3^- retention where feasible in other stream modification or restoration activities may provide additional NO_3^- retention (Kaushal et al. 2008; Klocker et al. 2009; Mayer et al. 2022). The restoration effort at BSR demonstrates that floodplain reconnection and legacy sediment removal can support NO_3^- reductions and should garner consideration as a nutrient BMP (Altland et al. 2020) in streams impaired by legacy sediment.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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