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Stream restoration can reduce nitrate levels in agricultural landscapes

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12 Abstract

The EU Water Framework Directive (WFD) has emphasized that altered stream/river morphology and 13 diffuse pollution are the two major pressures faced by European water bodies at catchment scales. 14 15 Increasing efforts have been directed toward restoration to meet WFD standards for ecological health, 16 but this work has achieved limited success. One challenge is that little is known about how 17 morphological changes (i.e., re-meandering) may affect nitrate retention within whole stream 18 networks. We investigated this issue in the well-monitored Bode catchment (3,200 km²) in central 19 Germany. First, we implemented a fully distributed process-based mHM-Nitrate model, exploring its 20 performance over the period from 2015 to 2018. Second, we simulated the effects of restoring more 21 natural stream morphology (i.e., increasing sinuosity) on nitrate retention. The mHM-Nitrate model 22 performed well in replicating daily discharge and nitrate concentrations (median Kling-Gupta values of 23 0.78 and 0.74, respectively). Within the stream network, gross nitrate retention efficiency was 5.1% 24 and 67.2% in the winter and summer, respectively; this measure took into account both denitrification 25 and assimilatory uptake. In the summer, the denitrification rate was about two times higher in a lowland 26 sub-catchment dominated by agricultural lands than in a mountainous sub-catchment dominated by forested areas (204.1 and 102.4 mg N m⁻² d⁻¹, respectively). Similarly, in the same season, the 27 assimilatory uptake rate was approximately five times higher in streams surrounded by lowland 28 29 agricultural areas than in streams in higher-elevation, forested areas (200.1 and 39.1 mg N m⁻² d⁻¹, respectively). In the late summer, denitrification always peaked after assimilatory uptake. In our 30 31 simulation, restoring stream sinuosity was found to increase net nitrate retention efficiency by up to 32 25.4%; greater effects were seen in small streams. Taken together, our results indicate that restoration 33 efforts should consider augmenting stream sinuosity to increase nitrate retention and decrease nitrate 34 concentrations at the catchment scale.

35 Keywords:

36 River restoration; sinuosity; mHM-Nitrate model; stream denitrification; assimilatory uptake

37 Highlights:

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1. The denitrification rate and assimilatory uptake rate are higher in agricultural-dominant areas than in forest areas.

40 2. Increasing stream sinuosity improves net nitrate retention efficiency more in small streams

41 than in large streams.

42 3. Small streams in agricultural areas should be given priority in restoration efforts.

3

43 1. Introduction

Excess nitrogen in surface waters represents a major threat to aquatic ecosystems (Birgand et al., 2007). 44 In streams, inorganic nitrogen largely occurs in the form of nitrate (NO_3) , a highly water soluble ion that 45 can easily enter streams and rivers from both diffuse and point sources. To reduce nitrate levels in 46 47 Europe's streams and rivers, extensive management strategies have been deployed over the past three 48 decades (European Commission, 1991a; b). Their success remains limited, as around 60% of Europe's surface water bodies still have not attained ecological health (European Environment Agency, 2018). 49 The EU Water Framework Directive (WFD) has emphasized that, in Europe, altered stream/river 50 51 morphology and diffuse pollution are two key pressures acting on water bodies at the catchment scale (Carvalho et al., 2019). For example, more than 37% of Germany's rivers are classified as heavily 52 53 modified, as a result of channelization or straightening (Pander et al., 2017). The loss of stream bottoms has shortened water residence times and limited hyporheic exchanges, resulting in lower levels of 54 55 nutrient retention and greater rates of downstream transport (Baker et al., 2012; Doyle et al., 2003; 56 Gucker and Boechat, 2004; Opdyke et al., 2006). Attention has turned to stream restoration as a 57 management tool for increasing nitrate retention (Craig et al., 2008; Newcomer Johnson et al., 2016; 58 Wohl et al., 2015). Multiple techniques have been tested out in headwater streams and large lowland rivers (Flávio et al., 2017), such as re-meandering (Lorenz et al., 2009; Pedersen et al., 2014) and 59 60 reconnecting streams with floodplains (Roley et al., 2012) and ponds (Passy et al., 2012) in agricultural 61 zones.

Within streams, nitrate retention is the result of temporary retention by plants (i.e., assimilatory uptake) and permanent removal by bacteria (i.e., denitrification) (Groffman et al., 2009; Ye et al., 2012). In general, stream restoration is thought to promote both processes (assimilatory uptake: Huang et al. 2022; denitrification: Craig et al., 2008). To date, research has largely focused on the effects of remeandering at the reach scale and has found contrasting results (Bukaveckas, 2007; Craig et al., 2008; Kaushal et al., 2008; Klocker et al., 2009; Lin et al., 2021; Veraart et al., 2014; Wagenschein and Rode,

68 2008). For example, denitrification was seen to be higher in restored streams (i.e., after reconnection 69 with floodplains) than in unrestored streams (Kaushal et al., 2008; Roley et al., 2012). Furthermore, 70 restored reaches may display higher levels of gross primary productivity and ecosystem respiration 71 (Kupilas et al., 2017). In contrast, Klocker et al. (2009) found no difference in denitrification rates 72 between restored and unrestored streams, and Veraart et al. (2014) observed that denitrification rates 73 were highly variable: for some streams, rates were significantly higher in unrestored versus restored 74 sections, while, in other streams, rates did not vary among sections. The researchers attributed these 75 results to differences in hydrological conditions and levels of sedimentary organic matter. Thus, we 76 presently have a limited understanding of how restoration could affect nitrate retention at broader 77 scales.

78 Within stream networks, nitrate retention is shaped by complex interactions between hydrological, 79 geomorphological, and biogeochemical processes (Ensign and Doyle, 2006; Yang et al., 2019; Ye et al., 80 2012). While the effects of hydrological and biogeochemical processes have been explored to some 81 degree (Alexander et al., 2009; Covino et al., 2010; Marcé et al., 2018), there has been no systemic 82 research on networks with contrasting morphologies and, more notably, on the effects of restoration 83 efforts (i.e., re-meandering). This gap in knowledge likely results from three key challenges. First, it is 84 difficult to disentangle how nitrate retention is affected by geomorphology versus other factors (Lin et al., 2016). Second, we lack detailed historical information on stream morphology (i.e., natural 85 86 conditions) within catchments (Guzelj et al., 2020). Third, uncertainty arises when attempts are made 87 to parse out the influences of lateral terrestrial flows versus in-stream processes (Helton et al., 2018; Helton et al., 2011). 88

Scenario analysis holds promise for addressing these challenges because it can be implemented by combining fully distributed catchment modeling with detailed spatiotemporal data from monitoring programs. In particular, simulations can explore how re-meandering could affect nitrate retention at the network scale. Recently, Yang et al. (2018) developed a fully distributed grid-based hydrological nitrate model (mHM-Nitrate) that can provide detailed spatial information on terrestrial nitrate inputs
within stream networks. This model has successfully described terrestrial and aquatic processes (i.e.,
assimilatory uptake) (Yang et al., 2019) across different catchments (Wu et al., 2022; Yang et al., 2018;
Zhou et al., 2022). In addition, researchers have been extensively characterizing the denitrification rates
associated with different land-use types (Böhlke et al., 2009; Mulholland et al., 2009). This has yielded
abundant opportunities for evaluating how morphological changes in streams can spatially and
temporally impact nitrate transport and retention.

Here, we looked at how stream morphology affects spatiotemporal nitrate retention dynamics within a stream network—the Bode catchment in central Germany. To this end, we used the mHM-nitrate model, which can handle large gradients in catchment characteristics. More specifically, we aimed to i) evaluate assimilatory uptake and denitrification within the entire catchment; ii) characterize spatiotemporal variability in retention dynamics and identify the key factors at play in two subcatchments; and iii) simulate the effects of re-meandering on nitrate concentrations and retention efficiency for a stream network.

107 2. Study area and methods

108 2.1 Study area

Covering around 3,200 km² in central Germany, the Bode catchment is closely monitored and thus 109 110 serves as a rich source of hydrological and hydrochemical data (Mueller et al., 2016; Wollschläger et 111 al., 2016). The catchment extends from the Harz Mountains, a low, rocky mountain range, to the 112 northeastern lowlands of central Germany. Annual precipitation follows an elevational gradient within 113 the catchment, ranging from more than 1,500 mm in the upper Harz Mountains to less than 500 mm 114 in the vast lowland plains (Figure 1a). Mean annual air temperature ranges from 5 °C at Brocken, the 115 mountain's highest peak, to 9.5 °C in eastern Magdeburg Börde (Wollschläger et al., 2016). The Harz 116 Mountains have steep slopes with shallow, less fertile soils that are predominantly covered by forests.

All agricultural activity is associated with the region's plateaus and lower-elevation areas. Within the catchment, 66% of the land is arable; 26% is forested or composed of semi-natural habitat; 7% is urban or dedicated to open-cast mining; and 1% is covered by water bodies and wetlands (CORINE 2012 land cover map, https://gdz.bkg.bund.de/index.php/default/open-data.html, last accessed 1 June 2020; Figure 1b).



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Figure 1. Bode catchment: (a) elevational map showing the gauging stations, (b) land use map, and (c) stream order obtained from the mHM-Nitrate model (1-km routing grid) with a stream mask derived from the observed network.

126 We chose two sub-catchments with different representative landscapes to investigate retention

- 127 processes in greater detail. Upper Selke is a sub-catchment located in the Harz Mountains (Meisdorf
- 128 outlet; Figure 1); it is dominated by forests (73% of 177.7 km²) and contains natural streams. Großer
- 129 Graben is a sub-catchment in the intensively farmed lowlands (Oschersleben outlet; Figure 1); it is
- dominated by arable land (87.4% of 435.4 km²) and contains heavily modified streams (Figure 1b).

About 80% of the lowland stream network is heavily modified or completely changed (State Agency for Flood Protection and Water Management of Saxony-Anhalt, LHW; <u>http://gldweb.dhi-wasy.com/gld-</u> <u>portal/</u>, last accessed 10 April 2020; Figure S1). Stream order analysis showed that there were two times more streams in Großer Graben than in Upper Selke (Table S1), the result of artificial drainage in the former. Additionally, the total stream length in Großer Graben was twice that in Upper Selke, except in the case of 1st-order streams (Table S1). Small streams (1st-3rd order) accounted for a high percentage of total stream length: 81% and 88% in Großer Graben and Upper Selke, respectively.

138 Mean daily nitrate concentrations (data collected at 15-min intervals) were available for the Meisdorf, 139 Hausneindorf, Hadmersleben, and Stassfurt stations (Helmholtz Centre for Environmental Research – 140 UFZ; Rode et al., 2016). Monthly and biweekly nitrate data (obtained via grab samples) were available for the Wegeleben, Nienhagen, and Oschersleben stations (LHW, http://gldweb.dhi-wasy.com/gld-141 142 portal/, last accessed 10 April 2020). Daily discharge data were available for all seven stations (LHW, 143 http://gldweb.dhi-wasy.com/gld-portal/, last accessed 10 April 2020). Monthly nitrate concentrations 144 were available for the Wegeleben and Nienhagen stations between 2007 and 2014. For the Wegeleben 145 station, nitrate concentrations were unavailable in 2015 and between 2017 and 2018. However, they 146 were available at two-month intervals at the Nienhagen station from 2015 to 2018 and at monthly intervals at the Oschersleben station from 2010 to 2018. 147

148 2.2 mHM-Nitrate model

The mHM-Nitrate model is a fully distributed process-based model of nitrate dynamics at the catchment scale (Yang et al., 2018). It was developed from the mesoscale Hydrological Model (Samaniego et al., 2010) and the Hydrological Predictions for the Environment model (Lindström et al., 2010). The mHM-Nitrate model simultaneously characterizes the hydrological and nitrate processes associated with terrestrial and stream environments for individual grid cells using a daily time step. For the terrestrial environment, the model considers the following key hydrological processes: interception, snow accumulation, snow melting, evapotranspiration, infiltration, groundwater 156 recharge, and runoff generation. The nitrate processes considered are the sources of nitrate (i.e., wet atmospheric deposition, application of fertilizer and manure, and presence of plant/crop residues), 157 158 transports (i.e., infiltration through multiple soil layers and percolation to the deep groundwater layer), sinks (i.e., denitrification and uptake by plants/crops), and transformation among the four nitrogen 159 pools (i.e., dissolved inorganic nitrogen, dissolved organic nitrogen, active solid organic nitrogen, and 160 161 inactive solid organic nitrogen) for each soil layer. For the stream environment, the model considered nitrate transformation (i.e., denitrification, assimilatory uptake, and remineralization) for each reach. 162 163 More detailed descriptions of the mHM-Nitrate model can be found in Yang et al. (2018) and Yang et al. (2019); the source code can be found in Yang and Rode (2020). 164

165 Gross nitrate assimilatory uptake within streams (F_{assim} ; kg N d⁻¹) was calculated using the new 166 regionalization approach proposed by Yang et al. (2019):

167
$$F_{assim} = U_{assim} \times f_L \times W \times L \times H \times \Delta t \tag{1}$$

where U_{assim} is the assimilatory uptake rate (mg N $m^{-3} d^{-1}$); $f_L \in [0,1]$ is a light availability coefficient that reflects the combined impact of global radiation and riparian shading on assimilatory uptake; and W, L, H are stream width (m), length (m), and depth (m), respectively.

171 Net assimilatory uptake within streams (F_{net} ; kg N d⁻¹) was calculated by subtracting remineralization 172 from gross assimilatory uptake (Yang et al., 2019), which was determined by multiplying gross 173 assimilatory uptake and a temperature factor (f_T),

$$F_{net} = F_{assim} \times f_T \times f_{net} \tag{2}$$

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$$f_T = \frac{T}{20} \times \frac{T_{10} - T_{20}}{5} \tag{3}$$

where $f_{net} \in [0,1]$ is a land-use coefficient that reflects the fraction of gross assimilatory uptake; T is water temperature (°C); and T_{10} and T_{20} are mean water temperature at 10 and 20 days, respectively. 178 The amount of denitrification within streams (F_{den} ; kg N d⁻¹) was calculated based on the relationship 179 between the denitrification rate and nitrate concentrations:

$$F_{den} = U_{den} * W * I$$

181
$$U_{den} = U_{max} * \frac{C_{NO_3^-}}{C_{NO_3^-} + ks} * f_{temp}$$

182
$$f_{temp} = \begin{cases} 0 & T < \\ \frac{T}{5} \times 2^{\frac{T-20}{10}}, & 0 < T < \\ 2^{\frac{T-20}{10}}, & T > 5 \end{cases}$$

where U_{max} is the maximum potential denitrification rate (mg N m⁻² d⁻¹) and U_{den} is the denitrification rate (mg N m⁻² d⁻¹) adjusted for $C_{NO_3^-}$ (nitrate concentration; mg N L⁻¹) and ks (nitrate concentration at half saturation; mg N L⁻¹). The latter has a default value of 1.5 mg N L⁻¹ in the mHM-Nitrate model (Yang et al., 2018).

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The net nitrate retention efficiency (Eff_{net}) of a stream is the amount of nitrate retained by net assimilatory uptake and denitrification (sum of F_{den} and F_{net}) divided by the total nitrate input load (L_{input} ; the sum of lateral terrestrial imports and upstream loads). The gross nitrate retention efficiency (Eff_{gross}) for a stream is the amount of nitrate retained by gross assimilatory uptake and denitrification (sum of F_{den} and F_{assim}) divided by the total nitrate input load (L_{input}) (Wollheim et al., 2008).

192 In previous studies using the mHM-Nitrate model, stream networks were generated using the digital 193 elevation model (DEM), and nitrate retention processes within streams (both assimilatory uptake and 194 denitrification) were considered for all reaches (i.e., grid cells). This approach may generate a high 195 degree of uncertainty around retention levels because of the uncertainty around the quantity of benthic surface area within the stream network. Thus, we employed a high-resolution digital elevation 196 197 model (DEM; 25 × 25 m) in combination with the observed stream network (LHW, http://gldweb.dhi-198 wasy.com/gld-portal/, last accessed 10 April 2020) (Figure 1c) to generate a more representative model 199 stream network.

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(5)

(6)

200 To this end, we first created a fishnet grid polygon with a routing resolution of 1 imes 1 km using the 201 Create Fishnet tool in the Data Management Toolbox in ArcMap 10.8. Second, the fishnet polygon was transposed onto the observed stream network (Figure 1c). If the real stream occurred within a grid cell 202 203 of the fishnet polygon, the grid cell was assigned a value of 1; if not, the grid cell was assigned a value 204 of 0. Third, the fishnet was converted to raster using the Polygon to Raster tool in the Conversion 205 Toolbox and exported into an ASCII file using the Raster to ASCII tool, also in the Conversion Toolbox. 206 Fourth, the ASCII file was imported into the mHM-Nitrate model, where the routing source code was modified to consider an additional routing mask-representing the observed stream network. Stream 207 retention processes were only activated within the routing mask (Figure 1c). 208

209 2.3 Model setup

210 We only briefly summarize the setup of the mHM-Nitrate model for the Bode catchment because it is 211 described in detail elsewhere (Zhou et al., 2022). The model was run using daily time steps over the 212 period from 2006 to 2018. It was calibrated using data from 2010 to 2014. The results were validated 213 using data from 2015 to 2018, namely discharge and nitrate concentrations at seven gauging stations 214 that reflected key features of the Bode catchment (e.g., land use, stream order, and nitrate 215 concentration). To model processes within terrestrial and stream environments, the grid resolution was set to 1 km. To calibrate the model, we used sensitivity analysis to identify the 15 most sensitive 216 217 parameters—the top 10 hydrological parameters and the top 5 nitrate parameters. The latter were the 218 stream denitrification rate, the soil denitrification rate in arable and non-arable areas, and the 219 assimilatory uptake rate within streams in arable and non-arable areas. The range for the assimilatory 220 uptake rate (100–500 mg N m⁻² d⁻¹) was defined using high-frequency sensor measurements from 221 previous research (Rode et al., 2016; Yang et al., 2019). The range for the stream denitrification rate (10-700 mg N m⁻² d⁻¹) was defined using studies on lowland streams in central Germany (Huang et al., 222 2022; Kunz et al., 2017a; Kunz et al., 2017b; Zhang et al., 2023). 223

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224 2.4 Simulating stream restoration

We designed a model scenario to explore the effects of stream sinuosity on nitrate retention. It simulated a situation in which the current stream network had been restored, such that its sinuosity was greater than that seen in networks of channelized streams. Stream sinuosity (S_n) in each grid cell was calculated by dividing stream length (determined from LHW data) by thalweg length (determined

calculated stream power (*SP*; kg m s⁻³) in each grid cell as follows (Rhoads, 2010):

$$SP = \rho g Q S$$

where ρ is water density (1,000 kg m⁻³), g is the gravitational acceleration (9.8 m s⁻²), Q is the discharge rate (m³ s⁻¹), and S is the channel slope.

using the DEM). We estimated the sinuosity of natural streams using a series of equations. First, we

Second, we calculated natural stream sinuosity for the grid cells in which stream power was greater than 10 kg m s⁻³. We drew on the work of Harnischmacher (2007), which showed that the sinuosity of lowland streams was correlated with stream power when stream power was between 10 and 100 kg m s⁻³ (correlation coefficient = 0.946, p=0.001). This analysis used data for 11 undisturbed stream sections that served as references; these streams displayed similar geological conditions to natural streams in the lower Bode catchment (Harnischmacher, 2007). Sinuosity (*S_n*) was calculated as follows:

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$$S_n = 0.043 + \log_{10}^{SP} \tag{8}$$

For grid cells in which stream power was less than 10 kg m s⁻³, we calculated natural stream sinuosity by estimating mean potential sinuosity based on stream type (Briem et al., 2003; Ministerium für Umwelt und Naturschutz Landwirtschaft und Verbraucherschutz des Landes Nordrhein-Westfalen, 2010). The lowlands of the Bode catchment are largely characterized by small loess and loamdominated rivers (Type 18) (Figure S2), and potential natural sinuosity within the entire stream network ranges from 1.01 to 2.0, depending on stream type (Table S2). To simplify our calculations, we used the mean potential natural sinuosity for each stream type. Finally, we only focused on simulating the

(7)

restoration of streams in arable areas, since the streams in forested areas were less affected by human activity (Figure S1). The riparian zone was sufficiently large in the lowland arable areas to allow for increases in stream sinuosity.

- 251 In the baseline scenario (i.e., the actual state of the Bode stream network), stream sinuosity was low
- 252 (mean = 1.04 for $1^{st}-3^{rd}$ order streams and mean = 1.07 for $4^{th}-6^{th}$ order streams; Table 1). However,
- there was a certain degree of variability (range: 1.00–2.73), with high levels of sinuosity seen exclusively
- in very short stream sections (Figure S3a). In the restoration scenario, mean sinuosity increased by 0.35
- relative to the baseline, with the biggest augmentation seen in 6th-order streams (Table 1 and Figure
- 256 S3b).
- The effects of stream restoration were simulated over the validation period because the years between 258 2015 and 2022 were relatively dry and warm (Zhou et al., 2022), which likely represents future 259 conditions under climate change (Huang et al., 2015).
- Table 1. Stream sinuosity range (and mean) for each stream order in the baseline and restorationscenarios.

| Stream order | Sinuosity | | | |
|-----------------|------------------|------------------|--|--|
| | Baseline | Restoration | | |
| 1 st | 1.00-2.53 (1.03) | 1.00–2.53 (1.30) | | |
| 2 nd | 1.00-2.60 (1.04) | 1.00–2.60 (1.34) | | |
| 3 rd | 1.00-2.28 (1.04) | 1.00–2.53 (1.44) | | |
| 4 th | 1.00–2.73 (1.06) | 1.00-3.11 (1.50) | | |
| 5 th | 1.00-2.09 (1.08) | 1.00–2.54 (1.29) | | |
| 6 th | 1.00–2.65 (1.07) | 1.00–3.62 (1.55) | | |

262 3. Results

263 3.1 Model performance

The mHM-Nitrate model generally performed well when simulating discharge, nitrate concentrations, and nitrate loads at the seven gauging stations (Figure 2 and S4-S5 and Table 2). Nash-Sutcliffe efficiency (NSE) exceeded 0.72 and 0.80 for the calibration and validation periods, respectively (Table

20. For the validation period, the ranges of Kling-Gupta efficiency (KGE) were 0.52–0.93, 0.22–0.80, and 0.60–0.91 for discharge, nitrate concentrations, and nitrate loads, respectively (Table 2). The model did overestimate nitrate concentrations at the Oschersleben station (Figure 2d) for the whole modeling period (2010–2018); percent bias (PBIAS) was 11.0% and 28.8% for the calibration and validation periods, respectively. In contrast, nitrate loads were accurately estimated at all seven stations during both the calibration and validation periods (PBIAS range = -18.7–23.2%).

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Figure 2. Performance of the mHM-Nitrate model: discharge and nitrate concentrations at (a-b) Meisdorf, (c-d) Oschersleben, and (e-f) Stassfurt during the calibration period (2010–2014) and validation period (2015–2018).

278

| 279 | Table 2. Model evaluation metrics for daily discharge (Q; m ³ s ⁻¹), nitrate concentrations (mg N L ⁻¹), and | |
|-----|---|--|
| 280 | nitrate loads (kg N d ⁻¹) at the seven gauging stations during the calibration and validation periods. | |
| 281 | Metric abbreviations: NSE = Nash-Sutcliffe efficiency, KGE = Kling-Gupta efficiency, and PBIAS = percent | |

282 bias.

| Station | Metric | Calibr | Calibration (2010–2014) | | | Validation (2015–2018) | | |
|--------------|--------|--------|-------------------------|-------|-------|------------------------|-------|--|
| | | Q | NO₃⁻ | Load | Q | NO ₃ - | Load | |
| Meisdorf | NSE | 0.84 | 0.48 | 0.67 | 0.78 | 0.60 | 0.72 | |
| | KGE | 0.77 | 0.70 | 0.74 | 0.71 | 0.76 | 0.82 | |
| | PBIAS | 4.10 | -6.40 | -3.60 | 23.8 | 4.10 | 11.8 | |
| Hausneindorf | NSE | 0.85 | -0.56 | 0.68 | 0.82 | 0.42 | 0.77 | |
| | KGE | 0.87 | 0.44 | 0.75 | 0.78 | 0.72 | 0.83 | |
| | PBIAS | 11.1 | -21.4 | 4.40 | 16.2 | 3.30 | 12.4 | |
| Wegeleben | NSE | 0.93 | -0.36 | 0.79 | 0.93 | - | - | |
| | KGE | 0.96 | 0.58 | 0.72 | 0.88 | - | - | |
| | PBIAS | -0.50 | -17.0 | -16.2 | 3.30 | - | - | |
| Nienhagen | NSE | 0.72 | -0.08 | 0.75 | 0.37 | -0.46 | 0.77 | |
| | KGE | 0.80 | 0.50 | 0.78 | 0.52 | 0.22 | 0.70 | |
| | PBIAS | 13.3 | -23.2 | -15.5 | 36.1 | -2.40 | 23.2 | |
| Oschersleben | NSE | 0.75 | 0.54 | 0.65 | 0.80 | 0.41 | 0.78 | |
| | KGE | 0.85 | 0.60 | 0.61 | 0.71 | 0.61 | 0.60 | |
| | PBIAS | -8.70 | 11.0 | -13.3 | -8.30 | 28.8 | -10.7 | |
| Hadmersleben | NSE | 0.87 | 0.52 | 0.85 | 0.93 | 0.61 | 0.93 | |
| | KGE | 0.89 | 0.66 | 0.79 | 0.93 | 0.80 | 0.91 | |
| | PBIAS | 0.80 | -8.30 | -8.50 | 4.80 | 6.30 | 5.80 | |
| Stassfurt | NSE | 0.85 | 0.50 | 0.82 | 0.92 | 0.63 | 0.91 | |
| | KGE | 0.87 | 0.71 | 0.72 | 0.91 | 0.80 | 0.91 | |
| | PBIAS | 0.50 | -13.7 | -18.7 | 6.20 | 2.60 | 3.10 | |

3.2 Spatiotemporal dynamics of nitrate retention 283

3.2.1 Annual and seasonal nitrate retention in the Bode catchment 284

| 285 | The mHM-Nitrate model suggested that the Bode stream network experienced the highest total nitrate |
|-----|--|
| 286 | input loads (L_{input}) in the winter from 2015 to 2018 (annual mean = 3.68 kg N ha ⁻¹ y ⁻¹ ; Table 3). For this |
| 287 | same period, annual net and gross nitrate retention efficiency (Eff_{net} and Eff_{gross}) were 12.9% and |
| 288 | 24.6%, respectively (Table 3). Both peaked in the summer (34.9% and 67.2%, respectively), which is |
| 289 | when input loads were lowest and assimilatory uptake and denitrification were highest. In contrast, the |
| 290 | two types of efficiency had lower values in the winter and spring (Table 3). Within the stream network, |

the assimilatory uptake rate (U_{assim}) reached its greatest value in the spring and summer (104.9 and

133.9 mg N m⁻² d⁻¹, respectively), while the denitrification rate (U_{den}) was highest in the summer and

autumn (158.2 and 116.7 mg N $m^{-2} d^{-1}$, respectively; Table 3).

Table 3. Variables describing mean annual and seasonal nitrate input loads and retention for the entire stream network from 2015–2018. Variable values were estimated using the mHM-Nitrate model; the only exception was L_{obs} , which was calculated from the observed data. Abbreviations: L_{input} = total nitrate input load; L_{obs} = observed exported nitrate load; L_{out} = estimated exported nitrate load; F_{assim} = amount of gross assimilatory uptake; U_{assim} = assimilatory uptake rate; F_{den} = amount of denitrification; U_{den} = denitrification rate; Eff_{net} = net nitrate retention efficiency; and Eff_{gross} = gross nitrate retention efficiency.

| Variable | Winter | Spring | Summer | Autumn | Annual |
|--|--------|--------|--------|--------|--------|
| L_{input} (kg N ha ⁻¹ | 1.49 | 1.06 | 0.52 | 0.56 | 3.68 |
| season ⁻¹ / kg N ha ⁻¹ y ⁻¹) | | | | | |
| L_{obs} (kg N ha ⁻¹ y ⁻¹) | 1.47 | 0.97 | 0.32 | 0.38 | 3.14 |
| L_{out} (kg N ha ⁻¹ y ⁻¹) | 1.44 | 1.09 | 0.34 | 0.40 | 3.27 |
| F_{assim} (kg N d ⁻¹) | 58.1 | 565.7 | 512.7 | 165.5 | 321.0 |
| U_{assim} (mg N m ⁻² d ⁻¹) | 12.7 | 104.9 | 133.9 | 45.9 | 74.7 |
| F_{den} (kg N d ⁻¹) | 169.7 | 320.9 | 536.7 | 362.1 | 348.4 |
| U_{den} (mg N m ⁻² d ⁻¹) | 45.4 | 77.1 | 158.2 | 116.7 | 99.5 |
| Eff_{net} (%) | 3.8 | 10.2 | 34.9 | 21.4 | 12.9 |
| Eff_{gross} (%) | 5.1 | 27.7 | 67.2 | 31.2 | 24.6 |

301 3.2.2 Nitrate retention within two representative sub-catchments

For the period from 2015 to 2018, we investigated nitrate retention within two sub-catchments— Upper Selke and Großer Graben. They were chosen because they represented certain landscape profiles within the Bode catchment (Figure 1). To characterize daily rates of denitrification (U_{den}) and assimilatory uptake (U_{assim}), we determined the median values for all the streams in the network. Both rates were highly variable among seasons and years in the two sub-catchments (Figure 3). At the annual scale, the denitrification rate (U_{den}) was nearly two times higher in Großer Graben than in Upper Selke (126.4 vs. 69.1 mg N m⁻² d⁻¹, respectively; Table 4). Seasonally, U_{den} was high in the summer and autumn and low in the spring and winter in both sub-catchments (Figure 3 and Table 4). Furthermore, there was seemingly an influence of land-use type. In the summer, U_{den} was two-fold greater in Großer Graben (median = 204.1 mg N m⁻² d⁻¹), which is dominated by arable land, than in Upper Selke, which is dominated by forests (median = 102.4 mg N m⁻² d⁻¹).

At the annual scale, the assimilatory uptake rate (U_{assim}) was more than three times higher in Großer Graben than in Upper Selke (102.0 vs. 27.6 mg N m⁻² d⁻¹, respectively; Table 4). This rate was also high in the spring and summer and low in the autumn and winter in both sub-catchments (Figure 3 and Table 4). In the summer, the U_{assim} was five times greater in Großer Graben than in Upper Selke (median ± SD = 200.1 ± 27.1 vs. 39.1 ± 8.7 mg N m⁻² d⁻¹, respectively). In Upper Selke, it always peaked in April and then rapidly decreased (Figure 3a).

Compared to assimilatory uptake, denitrification accounted for a higher proportion of gross nitrate uptake in Upper Selke than in Großer Graben across all seasons, except for the spring (range in Upper Selke: 72–88% vs. Großer Graben: 51–87%; Table 4). A similar pattern was seen at the annual scale (Upper Selke: 71% vs. Großer Graben: 55%; Table 4).

Gross nitrate retention efficiency (Eff_{gross}) demonstrated clear annual and seasonal patterns in both 323 sub-catchments (Figure 4); high values of Eff_{gross} were seen during low-flow periods in the summer 324 325 and autumn (Table 4). It decreased rapidly in July 2017 at Großer Graben and Upper Selke, the result 326 of peak flow events causing high nitrate loads in streams (Figures 2a and 2c). At the annual scale, gross 327 efficiency displayed a similar median value in both sub-catchments (Upper Selke: 26.0% and Großer Graben: 35.2%). In the summer, the median was higher for Großer Graben than for Upper Selke (82.2% 328 329 vs. 58.8%, respectively). It is worth noting that the sub-catchments had similar median benthic surface areas in the summer (Großer Graben: 0.28 km² and Upper Selke: 0.24 km²). 330



331

Figure 3. Daily denitrification rates, gross assimilation rates, and gross nitrate retention efficiencies in the sub-catchments of (a) Upper Selke and (b) Großer Graben from 2015 to 2018.

| 334 | Table 4. Median ± SD daily denitrification rates, assimilatory uptake rates, and gross nitrate retention |
|-----|--|
| 335 | efficiency at seasonal and annual scales in the Upper Selke and Großer Graben sub-catchments from |
| 336 | 2015 to 2018. Abbreviations: U_{den} = denitrification rate; U_{assim} = assimilatory uptake rate; and Eff_{gross} |
| 337 | = gross nitrate retention efficiency |

| Variable | Sub-catchment | Winter | Spring | Summer | Autumn | Annual |
|---|---------------|--------------------|---------------------|---------------------|---------------------|--------------------|
| | | | | | | |
| U_{den} | Upper Selke | 25.9 <u>+</u> 22.8 | 61.8 ± 28.1 | 102.4 ± 22.1 | 81.1 ± 25.6 | 69.1 <u>±</u> 36.5 |
| (mg N m ⁻² d ⁻¹) | Großer Graben | 70.3 ± 33.3 | 111.1 <u>+</u> 41.3 | 204.1 <u>+</u> 22.6 | 148.9 <u>+</u> 34.8 | 126.4 ± 61.6 |
| U _{assim} | Upper Selke | 5.0 ± 8.4 | 63.9 <u>+</u> 22.7 | 39.1 ± 8.7 | 18.3 ± 6.1 | 27.6 ± 24.3 |
| (mg N m ⁻² d ⁻¹) | Großer Graben | 9.9 ± 14.7 | 148.9 <u>+</u> 51.1 | 200.1 ± 27.1 | 56.8 ± 40.6 | 102.0 ± 77.5 |
| Eff_{gross} (%) | Upper Selke | 2.0 ± 3.7 | 22.7 <u>+</u> 14.0 | 58.8 <u>+</u> 17.4 | 29.3 <u>+</u> 17.2 | 26.0 ± 22.4 |
| | Großer Graben | 5.4 ± 7.0 | 22.0 ± 17.2 | 82.2 ± 23.3 | 42.6 ± 30.4 | 35.2 <u>+</u> 31.7 |

338 3.2.3 Spatiotemporal patterns in nitrate retention efficiency

- 339 We plotted accumulated net retention efficiency (ANRE) along an upstream to downstream gradient
- 340 for the two main stems of the Upper Selke and Großer Graben sub-catchments (Figure 4: heatmap;

Figure S6: stream networks). ANRE is the ratio between total nitrate retention within streams and the 341 total nitrate input load attributable to terrestrial sources. Spatiotemporal patterns in ANRE varied with 342 catchment size, which increased from headwater to outlet. Dynamics were similar across seasons and 343 344 years in both sub-catchments: ANRE was higher in the summer and autumn but lower in the winter and 345 spring (Figure 4). ANRE remained high (>60%) between late June and late November of 2018 in Upper 346 Selke and between late July and early December of 2018 in Großer Graben, periods of drought in the sub-catchments. Between 2015 and 2018, ANRE hit its minimum value in the summer of 2017, due to 347 348 peak flows in July 2017 (Figure 4 and Figures 2a and 2c).

349 However, some differences were apparent between the two sub-catchments for the period between 2015 and 2018. In Upper Selke, which is dominated by forests, ANRE was lowest in headwater streams, 350 351 as a result of their low assimilatory uptake rates (U_{assim}) and denitrification rates (U_{den}) (Figure 4a). In contrast, in streams near the outlet (<36 km away), ANRE reached high values (>60%) during the 352 353 summer and autumn (Figure 4a). In Großer Graben, which is dominated by arable land, ANRE was 354 highest in headwater streams, which experienced lower total nitrate input loads (L_{input}) than higher 355 order streams (Figure 4b). In this sub-catchment, high ANRE values (>60%) only occurred in headwater streams that were about 40 km upstream from the outlet (Figure 4b). 356

357 In Upper Selke, there was a correlation between ANRE values in the summer and catchment size (Figure 358 4a). This pattern could have two explanations: 1) the relative surface area covered by forest increased 359 from upstream to downstream and 2) there is a positive correlation between nitrate retention and 360 benthic surface area. In the summer of 2018, ANRE was lower in the sub-catchment's middle region, namely in streams located 24.8 and 18.0 km from the outlet (Meisdorf station, Figure 4). Such was due 361 362 to the large nitrate input loads (L_{input}) from nearby arable land (Figure S6a). Similarly, ANRE was dramatically lower in intermediate sections of Großer Graben (Figure 4b) because of the large nitrate 363 364 input loads (L_{input}) from tributaries that joined the main network stem around 38.9 and 25.5 km from 365 the outlet.



Figure 4. Patterns of accumulated net retention efficiency (ANRE) in the (a) Upper Selke and (b) Großer
 Graben sub-catchments. The y-axis depicts the direction of flow (headwater to outlet = top to bottom),
 while the x-axis depicts changes over time.

370

371 3.3 Simulated effects of stream restoration

In the baseline scenario, summer net nitrate retention efficiency (Eff_{net}) was slightly higher in streams in forested areas versus agricultural areas (median = 6.2% vs. 3.9%, respectively). Within areas with similar land use, this variable had larger values in small streams than in large streams. For example, in the summer in Großer Graben, the values of Eff_{net} ranged from 0.1 to 34.0% for 1st-3rd order streams and from 0.1 to 2.0% for 4th-5th order streams (Figure 5a). In the restoration scenario, in the summer,
greater improvements in net retention efficiency were seen for small streams in Großer Graben (1st3rd order streams: increase of 0.1–16.8%, 4th-5th order streams: increase of 0–11.8%) (Figure 5b). The



383



Figure 5. Spatial patterns of net nitrate retention efficiency in the summer in the a) baseline scenario
 and (b) in the restoration scenario (% increase over baseline).

384 In the baseline scenario, within areas with similar land use, mean summer nitrate concentrations were higher in small streams (1st-3rd order) than in large streams (4th-6th order) (Figure 6a). For instance, in 385 386 Großer Graben, the range for small streams was 3.3–4.8 mg N L⁻¹, while the range for large streams was 2.3–2.6 mg N L⁻¹. Additionally, these concentrations were also higher in small streams found in lowland 387 agricultural areas (range = 0.9–17.0 mg N L⁻¹) than in small streams in mountainous areas (range = 0.6– 388 389 5.2 mg N L⁻¹) (Figure 6a). In the restoration scenario, nitrate concentrations declined more sharply in 390 the former than in the latter areas; the largest decrease occurred in the tributaries of the lower Bode 391 river (-1.3 mg N L⁻¹; Figure 6b). In lowland areas, summer nitrate concentrations dropped more for large streams (4th-6th order) than for small streams (1st-3rd order) (Figures 6b and S7b). For example, in 392 393 Großer Graben, these concentrations declined by 0.1–0.3 mg N L⁻¹ and 0.3-0.5 mg N L⁻¹ for small and 394 large streams, respectively.



Figure 6. Spatial patterns of mean summer nitrate concentrations in the a) baseline scenario and (b) in
 the restoration scenario (absolute decrease from baseline).

398

399 4. Discussion

In this study, we investigated the potential effects of stream restoration on nitrate retention dynamics 400 401 via a combined approach. We utilized the detailed monitoring data available for the Bode catchment in 402 a well-calibrated, process-based mHM-Nitrate model to explore network-scale patterns. According to 403 established criteria for evaluating watershed model performance (Moriasi et al., 2015; Moriasi et al., 2012), the mHM-Nitrate model generally performed well in capturing the dynamics of both discharge 404 405 and nitrate concentrations. Although the model struggled somewhat with discharge at the Meisdorf 406 and Nienhagen stations (PBIAS: 23.8% and 36.1%, respectively; Table 2), the absolute differences 407 between the observed and estimated values were small (Meisdorf: 36.1 mm year¹ and Nienhagen: 23.5 mm year-1). Nitrate concentrations were overestimated at the Oschersleben station for the entire 408 modeling period (2010–2018) because discharge and nitrate concentrations were overestimated 409 410 (PBIAS: 26.8% and 18.4%, respectively) in the summer during the validation period. The differences corresponded to 3.34 mm year⁻¹ and 0.31 mg N L⁻¹, respectively. A detailed discussion of model 411 412 performance for the Bode catchment is provided by Zhou et al. (2022).

4.1 Modeling nitrate retention processes in stream networks

The mHM-Nitrate model estimated that, in the summer, daily denitrification rates (U_{den}) ranged from 414 100.9 to 198.5 mg N m⁻² d⁻¹ (mean \pm SD = 151.1 \pm 19.1 mg N m⁻² d⁻¹ for the entire stream network). 415 These figures fit with those obtained by Mulholland et al. (2009), who used ¹⁵N isotope analysis to 416 417 estimate daily denitrification rates (U_{den}) at the reach scale. They found that values ranged from 0 to 418 220.1 mg N m⁻² d⁻¹ for small streams in areas with different land uses and climatic conditions. Using a reach-scale N₂ method on N-enriched streams, Böhlke et al. (2009) estimated that daily denitrification 419 rates (U_{den}) in the Iroquois River basin (USA) ranged from 48.4 to 677.0 mg N m⁻² d⁻¹; stream nitrate 420 421 concentrations were similar between their study and ours. In addition, Zhang et al. (2023) used highfrequency measurements to quantify daily denitrification rates (U_{den}) in the summer in the lower Bode 422 River. These observed values fell between 72.3 and 253.0 mg N m⁻² d⁻¹ and were thus reasonably similar 423 424 to our model's estimated values of 81.8 to 188.2 mg N m⁻² d⁻¹ for the same reaches.

425 The model estimated that daily assimilatory uptake rates (U_{assim}) were 27.6 ± 24.3 and 102.0 ± 77.5 mg N m⁻² d⁻¹ for streams in forested and agricultural areas, respectively (Table 4). Using high-frequency 426 427 measurements, Rode et al. (2016) found maximum daily assimilatory uptake rates in a Selke subcatchment (streams in forested areas: 97.5 mg N m⁻² d⁻¹ and streams in agricultural areas: 270 mg N 428 429 $m^{-2} d^{-1}$) that are consistent with our results (Figure 3 and Table 4). Applying the same model to the Selke catchment, Yang et al. (2019) reported values (mean \pm SD) of 86.4 \pm 1.9 mg N m⁻² d⁻¹ for streams in 430 forested areas and 18.8 ± 6.2 mg N m⁻² d⁻¹ for streams in agricultural areas. Kunz et al. (2017b) 431 determined that mean daily assimilatory uptake rates (U_{assim}) were 120 mg N m⁻² d⁻¹ and 239 mg N m⁻² 432 433 ² d⁻¹ for channelized and natural streams, respectively. This work took place in the lowlands associated 434 with the Weiße Elster River, which is near to our study area and thus provides additional support for 435 the reliability of our model's estimates.

436 4.2 Relationships between catchment characteristics and nitrate retention

Nitrate retention is determined by both denitrification and assimilatory uptake (U_{den} and U_{assim}). These 437 variables displayed pronounced seasonal variability among different land-use categories (Figure 3). 438 Denitrification (U_{den}) was higher in the summer and autumn and lower in the spring and winter for both 439 440 sub-catchments. This pattern matches those seen in previous studies (Alexander et al., 2009; Wollheim 441 et al., 2008), where retention dynamics were correlated with nitrate concentrations and temperature. This seasonality could also be related to levels of sediment and dissolved oxygen (Christensen et al., 442 443 1990; Inwood et al., 2005; Uusheimo et al., 2018) and to levels of organic carbon (Arango et al., 2007; 444 Comer-Warner et al., 2020; Tatariw et al., 2013). However, we were unable to include these variables 445 in our mHM-Nitrate model because the lack of observed data made it impossible to construct empirical 446 equations that would have allowed us to scale up to the entire stream network.

From 2015 to 2018, denitrification rates (U_{den}) were higher in Großer Graben than in Upper Selke across all seasons (Figure 3 and Table 4). This pattern likely resulted from the higher nitrate⁻ concentrations and water temperatures in Großer Graben, a sub-catchment dominated by agricultural activity. Our findings concur with those of past studies (Böhlke et al. (2009); Inwood et al. (2007); Mulholland et al. (2008)), which observed that denitrification rates (U_{den}) were positively correlated with nitrate concentrations across land use types.

453 Assimilatory uptake rates (U_{assim}) were similarly higher in Großer Graben than in Upper Selke across all seasons (Figure 3 and Table 4). These results align with those of previous research that utilized high-454 frequency measurements (Rode et al. 2016) and that applied the mHM-Nitrate model to the Selke 455 456 catchment (Yang et al., 2019a). The latter two studies reported that assimilatory uptake (U_{assim}) occurred at a higher rate in streams in open-canopy environments (i.e., agricultural areas) versus 457 458 closed-canopy environments (i.e., forests). Arango et al. (2008) found similar results. The assimilatory 459 uptake of nitrate is mainly controlled by primary productivity (Heffernan and Cohen, 2010; Roberts and 460 Mulholland, 2007), which is affected by light, a resource whose availability is higher in agricultural 461 versus forested areas (Yang et al., 2019). Denitrification always peaked after assimilatory uptake, in the 462 second half of July and August. This pattern likely arises because denitrification is more sensitive to 463 water temperature, while assimilatory uptake is more sensitive to light availability (Heffernan and 464 Cohen, 2010, Kunz et al. 2017b). Dynamics were consistent across streams and years.

465 Although the rates of both processes varied in space and time for the two sub-catchments, denitrification surpassed assimilatory uptake across all seasons, except for spring. Our observation fits 466 with the work by Böhlke et al. (2004), who noted that denitrification accounted for more than 50% of 467 468 gross nitrate uptake in a stream with high nitrate concentrations (i.e., occurring in an agricultural area). 469 Similarly, Potter et al. (2010) reported that denitrification accounted for 1–97% of gross nitrate uptake and that this figure exceeded 35% for five out of the nine streams studied. Kunz et al. (2017b) found 470 471 that, in July, the denitrification rate was about five times higher than the assimilatory uptake rate in a 472 natural reach of the Weiße Elster River. In contrast, Mulholland et al. (2008) indicated that 473 denitrification made a relatively limited contribution (16%) to gross nitrate uptake (mainly in low-nitrate 474 streams); Ribot et al. (2017) arrived at a figure of 0.15%. These low values may have resulted from 475 specific site conditions. They were obtained using ${}^{15}NO_3$ tracers that were added to streams with low 476 nitrate concentrations. It appears that denitrification makes a greater contribution to total nitrate uptake when nitrate concentrations are higher. 477

478 Upper Selke and Großer Graben likely had similar temporal patterns of gross nitrate retention efficiency 479 and ANRE (Figures 3 and 4) because of similarities in interactions among land use, nitrate concentrations, temperature, and discharge. Both variables had low values in the winter and spring due 480 481 to elevated terrestrial inputs during high flow periods, and seasonally low water temperatures resulted 482 in reduced rates of assimilatory uptake and denitrification (Alexander et al., 2009). Terrestrial inputs 483 likely had more influence on the above temporal dynamics because they were an order of magnitude 484 larger. While terrestrial inputs were higher in Großer Graben than Upper Selke, median gross nitrate 485 retention efficiency in the summer was higher in Großer Graben than Upper Selke (Table 4), a pattern

that can be explained by the former's higher rates of assimilatory uptake and denitrification. The sub-486 487 catchments displayed significantly different spatial patterns of ANRE (Figure 4). ANRE was low in Upper 488 Selke headwaters because assimilatory uptake and denitrification were low (Figure 4a); it was higher in 489 Großer Graben headwaters than downstream reaches (Figure 4b) because the elevated nitrate levels 490 caused by terrestrial inputs in the downstream reaches far exceeded the amounts of nitrate removed 491 by assimilatory uptake and denitrification. This result indicates that land use around the nitrate source (e.g., arable land) can strongly influence retention efficiency at the network scale. Previous research 492 493 arrived at a similar conclusion: land use, and notably the location of arable lands within catchments, 494 can strongly affect the nitrate removal and export from the catchment (Casquin et al., 2021; Dupas et 495 al., 2019; Mineau et al., 2015).

496 4.3 Potential effects of restoring sinuosity on nitrate retention

In our baseline scenario, summer net nitrate retention efficiency was higher in streams in forested versus agricultural areas (Figure 5a). Indeed, in the latter, retention capacity was overwhelmed by large nitrate inputs from terrestrial sources. Adjusting for stream length, net nitrate retention efficiency per km was lower for large than small streams (Figure 5a), as seen in previous studies (Wollheim et al., 2008; Wollheim et al., 2006).

502 Our restoration scenario specifically explored the effects of increasing stream sinuosity. It found that in 503 areas with arable land, the changes to stream morphology increased net nitrate retention efficiency 504 more in the lowlands than in the mountains (Figure 5b); in the latter area, small streams already display 505 pronounced meandering (Figure S3). As stream length increased, so did the benthic surface areas, also 506 augmenting retention. These gains have also been observed at the reach scale in prior research 507 (Wagenschein and Rode, 2008).

508 Furthermore, in our restoration scenario, net nitrate retention efficiency improved more for small 509 streams (1st-3rd order) than for large streams (4th-6th order), as seen in Großer Graben (Figure 5b). This 510 pattern emerges because terrestrial nitrate input greatly exceeds nitrate retention in larger streams, which suggests that increasing sinuosity could have a greater impact on retention efficiency in small streams. Across the entire stream network, increased sinuosity more dramatically reduced nitrate concentrations in large streams than in small streams (Figure 6b), likely because large streams have experienced cumulative downstream retention and harbor larger benthic surface areas (Alexander et al., 2009). Consequently, restoration regimes that increase sinuosity could be powerfully deployed in small streams in agricultural areas, acting to increase nitrate retention efficiency and decrease nitrate transport downstream.

518 Our study adds to research looking at how alterations in stream morphology could affect nitrate 519 retention dynamics. Past work has shown that re-meandering can induce transient storage, which impacts the denitrification rate (Baker et al., 2012; Opdyke et al., 2006). Such may result mechanistically 520 521 from lower flow velocities and higher water residence times in the hyporheic zone (Bukaveckas, 2007; 522 Gomez et al., 2012; Pinay et al., 2009; Zarnetske et al., 2011). Additionally, denitrification could 523 experience greater increase in vertical hyporheic zones compared to meanders because vertical 524 exchanges beneath stream bedforms are considerably more pronounced than are lateral exchanges 525 through stream bars and meander banks (Gomez-Velez and Harvey, 2014; Gomez-Velez et al., 2015). 526 Modifications to stream morphology can directly or indirectly affect nutrient dynamics by increasing 527 spatiotemporal variability in the composition and activity of aquatic communities (Lin et al., 2016). However, it is hard to arrive at any generalizations because we continue to lack field studies comparing 528 529 denitrification rates in modified versus natural streams-data that are essential for model 530 parameterization. We have interpreted our results conservatively because we did not explicitly establish any links between these processes and natural stream morphology, which shapes rates of 531 nitrate uptake in the stream bed. 532

533 4.4 Implications for stream restoration

Although stream restoration projects are abundant, they often focus on the reach scale (Newcomer Johnson et al., 2016). This study highlights the need for developing methods that act at the network scale, such as increasing stream sinuosity. In our simulation, increasing stream sinuosity improved nitrate retention efficiency more in streams in agricultural areas (Großer Graben) than in streams in forested areas (Upper Selke). Moreover, this strategy more dramatically reduced nitrate concentrations in large streams than in small streams because of the accumulative retention in the upper streams. This finding indicates that restoration efforts should prioritize small streams in highly polluted, agricultural areas, such as our study area in the lowlands of central Germany.

Encouraging investment in stream restoration (e.g., re-meandering) can be challenging for two key reasons: first, it is costly and technically difficult and, second, the benefits are only significant during periods of low flow and low terrestrial inputs. Realistically, stream restoration alone cannot reduce nitrate concentrations to desired levels. Instead, systems exploiting a combination of terrestrial and stream-targeted measurements could be used to effectively and sustainably manage river basins (Lammers and Bledsoe, 2017). However, we must first conduct further research on how such combined measurements can affect nitrate retention at the stream network scale.

549 5. Conclusion

550 In this study, we used observed data from Germany's Bode catchment in combination with a fully 551 distributed process-based mHM-Nitrate model to investigate how re-meandering could affect nitrate 552 retention dynamics within a heavily modified stream network. There was pronounced spatiotemporal 553 variability in rates of assimilatory uptake and denitrification within stream networks with different land-554 use types and morphological characteristics. Both rates were higher in streams in more agricultural 555 versus more forested areas. At the network scale, increased stream sinuosity had a greater positive 556 impact on nitrate retention efficiency in small streams. However, nitrate concentrations decreased 557 more dramatically in large streams due to accumulative retention in upper streams. Our findings 558 underscore that major benefits could arise from re-meandering small streams in agricultural areas. It is 559 important to acknowledge that our stream restoration regime was somewhat simplified—we increased 560 sinuosity without considering the resulting effects on rates of nitrate denitrification and assimilatory

561 uptake. Thus, this work is a conservative first step along a lengthy research pathway. For example, 562 future research should explore whether nitrate retention efficiency could be enhanced even more by 563 combining stream-based strategies (e.g., re-meandering, improved floodplain connectivity) with land-564 based strategies (e.g., buffer strips, construction of wetlands).

565 Our results highlight the dominant role of denitrification in gross nitrate uptake across all seasons 566 (excluding the spring). They also showed that, regardless of stream size or nearby land use, the 567 denitrification rate always peaked after the assimilatory uptake rate, in the second half of July and 568 August.

Taken together, our findings suggest that stream restoration efforts should prioritize small streams in highly polluted, agricultural areas. To optimally design restoration strategies, we must characterize denitrification and assimilatory uptake rates in stream networks in the field before and after restoration; these data could then be used in distributed hydrological water quality models (such as an mHM-Nitrate model) to further improve understanding of these dynamics.

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