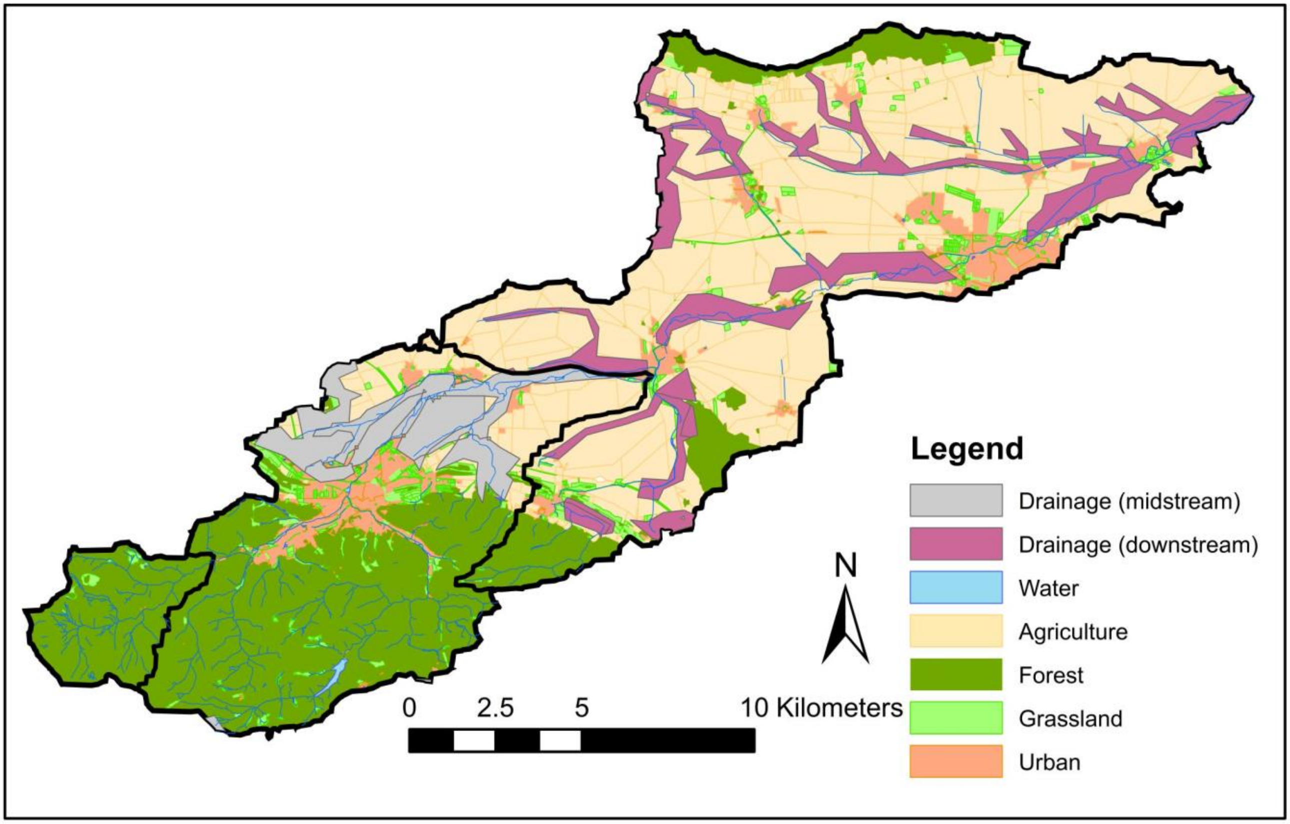
Supplement

S1 Addition to material and methods

S1.1 Catchment characteristics



5 **Figure S1.1: Map of the catchment highlighting the agricultural area that is artificially drained**

S1.2 Water quality time series of NH_4-N and NO_2-N

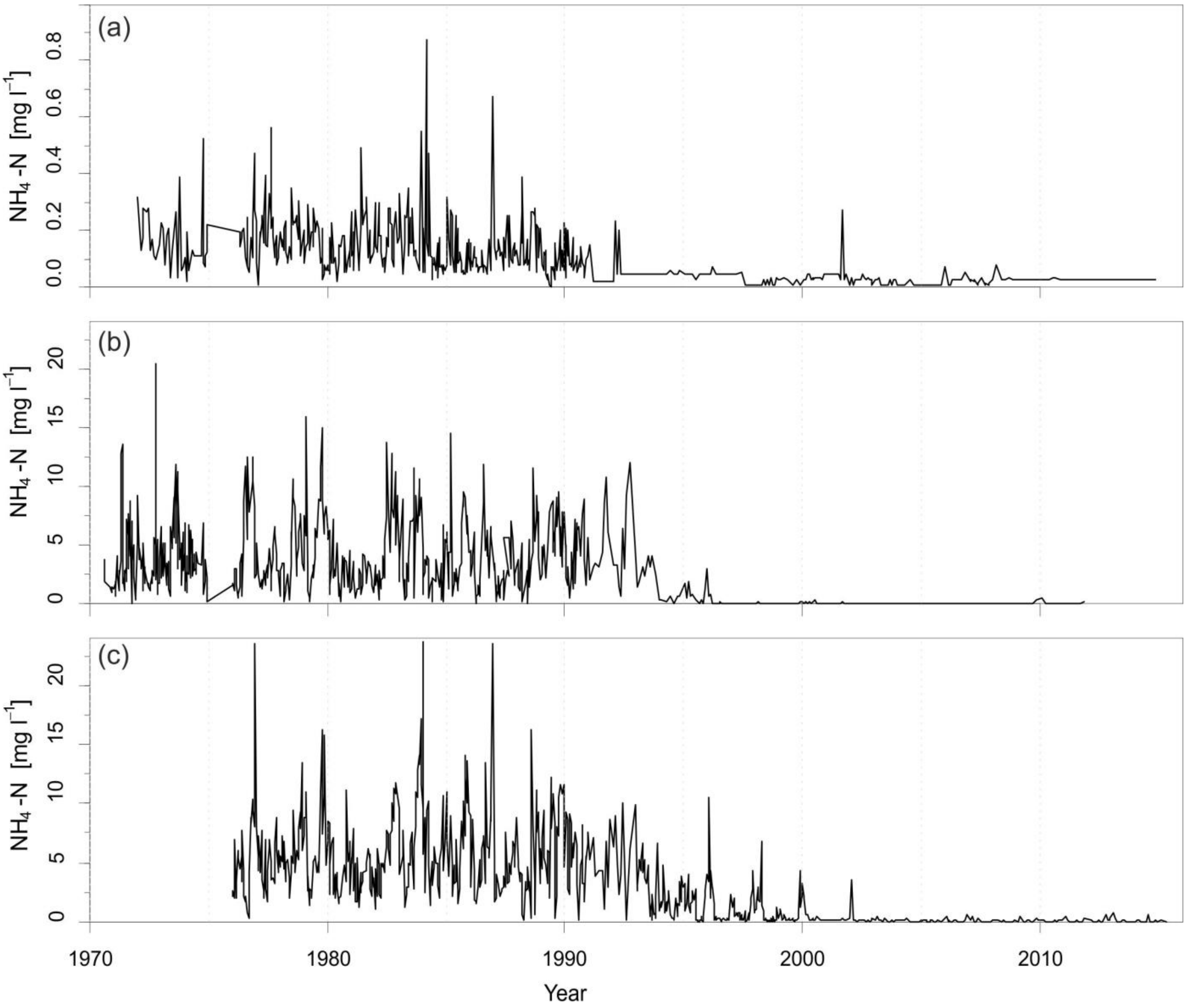


Figure S1.2.1: Time series of NH_4-N concentrations. (a) Upstream; (b) Midstream; (c) Downstream.

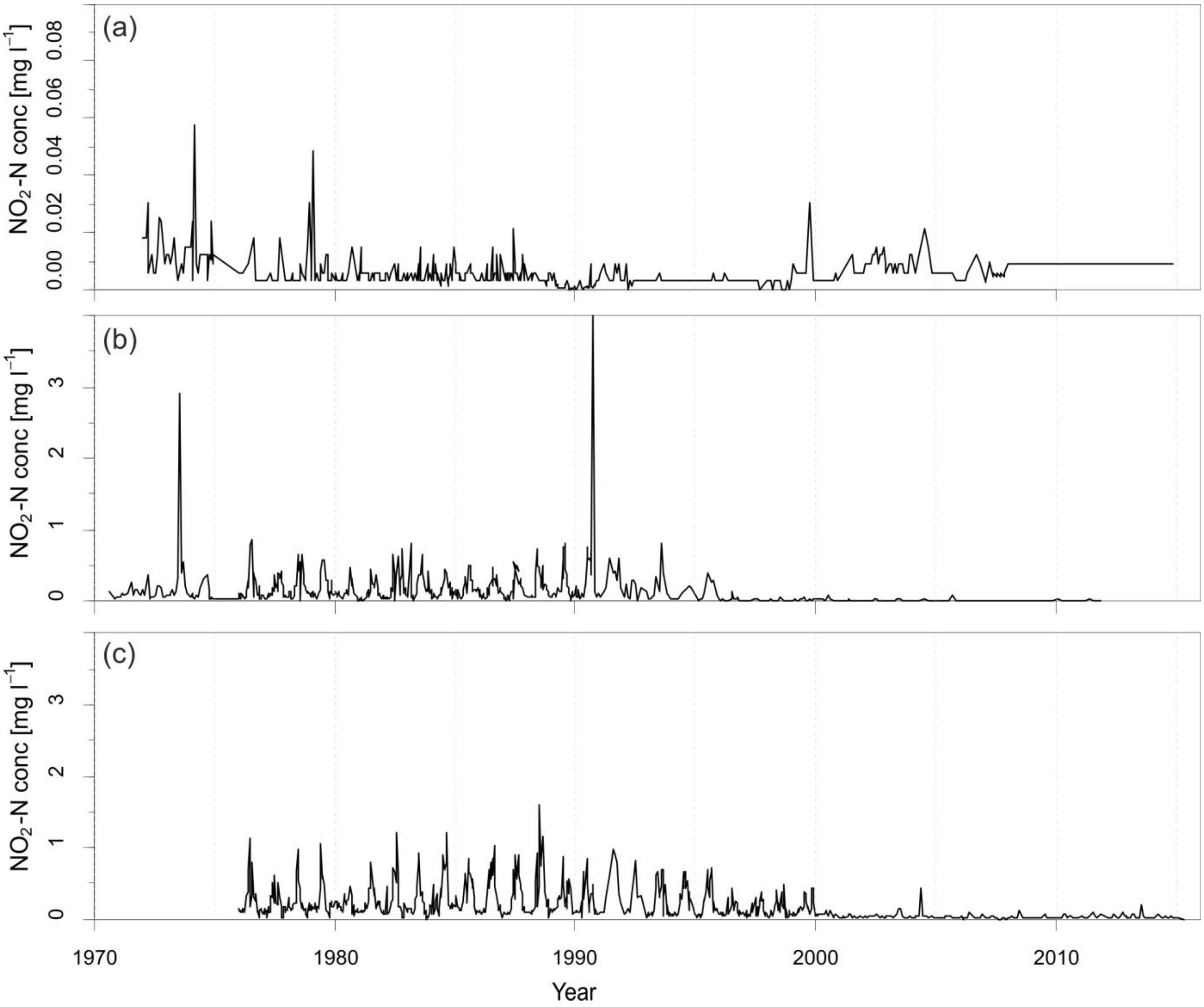


Figure S1.2.2: Time series of NO_2-N concentrations. (a) Upstream; (b) Midstream; (c) Downstream.

S.1.2.3 Estimation of NO_3-N contribution from wastewater borne NH_4-N

The assumption that agricultural, diffuse input was the main source of NO_3-N, needed an investigation of the possible amount of NO_3-N stemming from nitrified NH_4-N released by the WWTPs.

We estimated the maximal amount of waste water born NO_3-N from the highest nitrification rates taken from other studies (Webster et al., 2003; Mulholland et al., 2000, Tank et al., 2000). The highest rate was 0.19 g m^{-2} d^{-1} (Tank et al., 2000). Note that we assume that this rate is constant over the year and that NH_4-N was always unlimitedly available.

To calculate the river area, we used the stream length und mean stream width between the two WWTPs and corresponding stations Mid- and Downstream (Table S.1.2.3). This estimation took the relocation of the WWTP Wernigerode in 1995 into account. The estimation of waste water born NO_3-N revealed a maximal contribution of 5.2 % stemming from nitrification over the years. Hence, in-stream nitrification of NH_4-N release from point sources did not contributed significantly to riverine NO_3 loads or concentrations.

Table S.1.2.3: Estimation of NO_3-N contribution by in-stream nitrification of waste water born NH_4-N in the anthropogenically impacted subcatchments

	Midstream before 1995	Midstream after 1995	Downstream
NH_4-N uptake (Tank et al. 2000; g m^{-2} d^{-1})	0.19		
River area from WWTP to station (m^2)	58 456	12 431	46 092
Nitrified NH_4-N (t a^{-1})	4.1	0.9	3.2
Years of selected time series	1972–1995	1995–2011	1976–2011
Total uptake over time (t N)	107.0		111.9
Total NO_3-N export over time (t)	3 697		2 146
Total uptake according to export (%)	2.9		5.2

S2 Addition to results

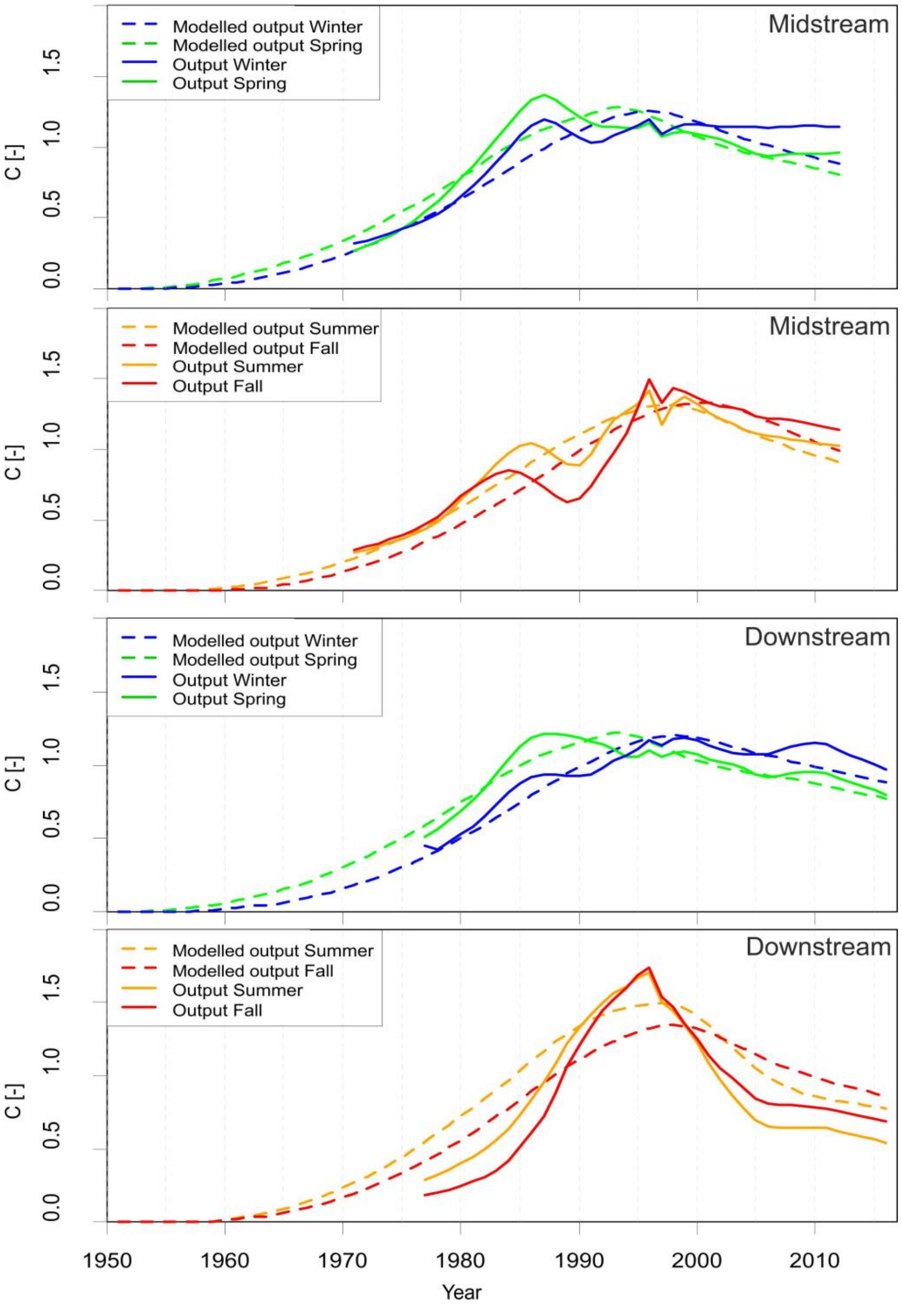


Figure S2.1: Modelled N-output concentrations derived from the N-input convolved with the log-normal travel time distributions (dashed lines) and the scaled observed flow-normalized seasonal NO_3-N concentrations (solid lines) over time.

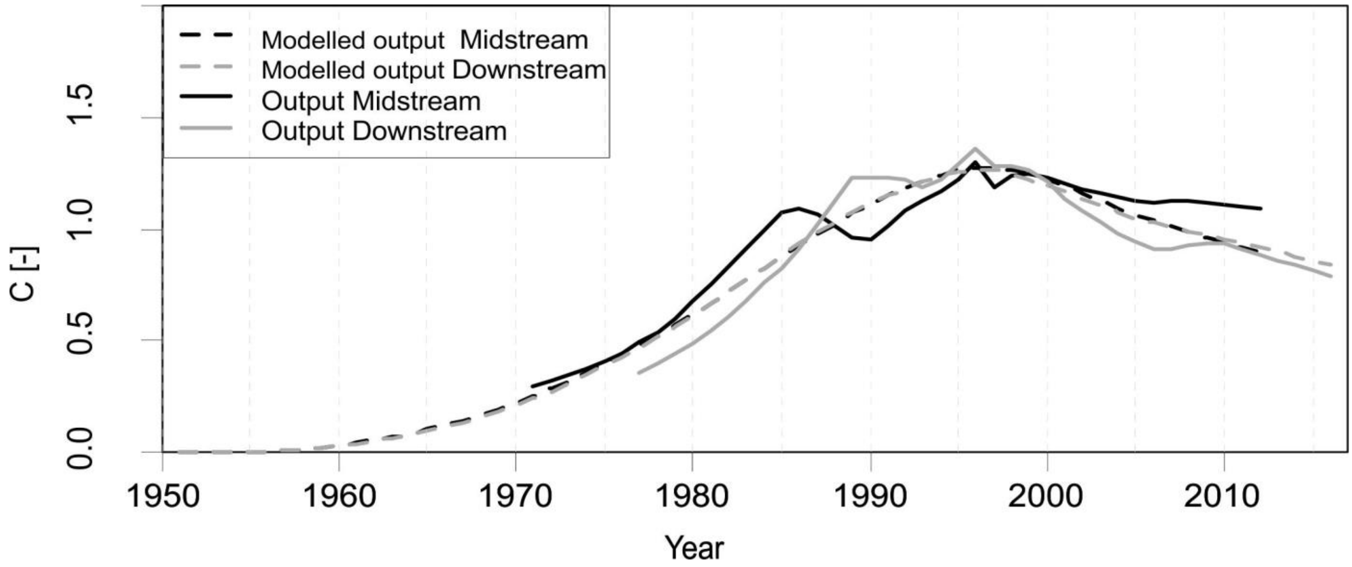


Figure S2.2: Modelled N-output concentrations derived from the N-input convolved with the log-normal travel time distributions (dashed lines) and the scaled observed flow-normalized annual NO_3-N concentrations (solid lines) over time.

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