Supplementary Information

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Section S1

Estimation of hourly rainfall time series for the Wylye catchment

Rainfall data were obtained from the Demonstration Test Catchment (DTC) Programme and the Environment Agency (EA) as follows:

Source	Site	Easting (m)	Northing (m)	Data format
DTC	Brixton Deverill	385600	137900	Daily, 9am to 9am
DTC	Norton Ferris	379000	136466	Daily, 9am to 9am
EA	Frome	377364	148748	Raw tipping bucket times with QA flags
EA	Gillingham	380310	125840	Raw tipping bucket times with QA flags
EA	Penridge	375350	131860	Raw tipping bucket times with QA flags
EA	Tisbury	395694	129843	Raw tipping bucket times with QA flags
EA	Walters Farm	372649	137298	Raw tipping bucket times with QA flags

The EA gauges were all outside the Wylye catchment; locations are shown in Figure 1c (main manuscript). Only data flagged G (good) was used from tipping bucket data, analysed to give hourly time series. For periods where tips were not marked 'G', hourly time series were filled with NaN (not a number), to signify missing data rather than no rain. For the hourly time series, cross correlation between sites indicated that the sites with highest cross correlation were Gillingham, Penridge and Walters Farm (cross correlation 0.73 – 0.80, at zero time lag). Hourly datasets were aggregated to daily (9am to 9am) and regressed with the daily datasets from Brixton Deverill (catchment outlet) and Norton Ferris (in W of catchment) to assess total rainfall volume and how representative each one was of rainfall in the catchment. The datasets most closely aligned with Brixton Deverill and Norton Ferris were Gillingham, Penridge and Walters Farm (R^2 between 0.79 and 0.90). As all the datasets had some missing data, a combined dataset taking the mean (discounting missing data) of sites Gillingham, Penridge and Walters Farm was used to create an hourly dataset from 1 January 2011 – 31 May 2014. This dataset was used for transfer function modelling.

Section S2

Model assessment criteria

Model fit was assessed according to R_t^2 (akin to Nash Sutcliffe Efficiency):

$$R_t^2 = 1 - \frac{\hat{\sigma}^2}{\sigma_y^2}; \qquad \text{(S1)}$$

where $\hat{\sigma}^2 = \frac{1}{N}\sum_{i=1}^{N}[\hat{y}_i - y_i]^2$; $\sigma_y^2 = \frac{1}{N}\sum_{i=1}^{N}[y_i - \bar{y}]^2$; $\bar{y} = \frac{1}{N}\sum_{i=1}^{N} y_i$ (S2)

$\hat{y}_i$ are the model estimations, y_i are the observations, $\hat{\sigma}^2$ is the variance of the model residuals and σ_y^2 is the variance of the observations.

Systematic over- or under-prediction of the model was evaluated with model bias:

Model bias = $100 * \Sigma[\hat{y}_i - y_i] / \Sigma y_i$ (S3)

A balance of model fit and over-parameterisation was sought using the Young Information Criterion (YIC) (Young, 1984) and visual inspection of the model fit to the monitoring data.

$$YIC = \log_e \frac{\hat{\sigma}^2}{\sigma_y^2} + \log_e \{NEVN\} \qquad \text{(S4)}$$

where NEVN is the normalised error variance norm defined as:

$$NEVN = \frac{1}{np}\sum_{i=1}^{np} \frac{\hat{\sigma}^2 \hat{P}_{ii}}{\hat{a}_i^2} \qquad \text{(S5)}$$

np is the number of parameters estimated, $\hat{P}_{ii}$ is the ith diagonal on the parameter covariance matrix, $\hat{a}_i^2$ is the square of the ith parameter. The first term in YIC is based on the coefficient of determination and is a measure of how well the model explains the data (the smaller the model residuals, the more negative this term becomes). The second term is a measure of the over-parameterisation; generally, a higher order model will capture more of the dynamics of the system, but with higher uncertainty in the parameter estimates. In that case the second term in YIC will dominate. Thus YIC is a compromise between the fit of the model and model complexity.

Table S1 Study catchment characteristics

Catchment	Newby Beck at Newby	Blackwater at Park Farm	Wylye at Brixton Deverill
Part of DTC catchment	Eden, Cumbria	Wensum, Norfolk	Avon, Hampshire
Sampling location at catchment outlet	54.59° N, 2.62° W	52.78° N, 1.15° E	51.16° N, 2.19° W
Elevation of sampling location (m a.s.l.)	233	43	189
Size of catchment (km^2)	12.5	19.7	50.2
Aspect (° from North)	28°	144°	106°
Mean (and standard deviation) annual rainfall[a] (mm)	1262 (220)	995 (142)	714 (109)
Baseflow index[b]	0.39	0.80	0.93
Soils[c]	Clay loam and sandy clay loam soils; Brickfield 3, Waltham and Clifton soil associations	Chalky boulder clay and sandy loam soils; Beccles 1, Burlingham 1 and Wick 2 and 3 soil associations	Sandy loam and silty clay loam soils; Ardington, Blewbury, Coombe 1, Upton 1, and Icknield soil associations
Geology	Glacial till over Carboniferous limestone	Quaternary glacial till, sands and gravels over Pleistocene Crag and Cretaceous Chalk	Cretaceous Chalk and Upper Greensand
Land use	Livestock	Arable crops	Livestock and cereals

[a] From UKCP Gridded Observation Data, 1981 – 2011 (Met Office, 2009)
[b] From Flood Estimation Handbook (Robson and Reed, 1999)
[c] From Soil Survey of England and Wales (Soil Survey of England and Wales, 1983)

Table S2 Notation

a_f, a_s	Parameters in the denominator polynomials of the partial fraction expansion into parallel, first order transfer functions (see SI Table S3)
b_f, b_s	Parameters in the numerator polynomials of the partial fraction expansion into parallel, first order transfer functions (see SI Table S3)
β	A constant exponent in the rainfall non-linearity (see Eq. 4)
δ	Pure time delay in a discrete-time model (see SI Table S3, Eq. S6 and S7)
m	Order of the numerator polynomial
n	Order of the denominator polynomial
NSE	Nash Sutcliffe Efficiency (see also R_t^2)
Q(*t*)	Discharge at time *t*
R(*t*)	Rainfall at time *t*
*R*e(*t*)	Effective rainfall at time *t*
R_t^2	(also NSE) Model fit = 1 – variance of model residuals/variance of observations
σ_y^2	Variance of observations $= \frac{1}{N}\sum_{i=1}^{N}[y_i - \bar{y}]^2$
$\hat{\sigma}^2$	Variance of model residuals $= \frac{1}{N}\sum_{i=1}^{N}[\hat{y}_i - y_i]^2$
TPload(t)	Total phosphorus load during time step ending at time t
τ	Time delay in a continuous-time model (see SI Table S3, Eq. S8 and S9)
y_i	Observation at ith time step
$\bar{y}$	Mean of observations $= \frac{1}{N}\sum_{i=1}^{N} y_i$
$\hat{y}_i$	Model prediction at ith time step
YIC	Young Information Criterion (see SI Section S2, Eq. S4)

Table S3 Structure of models and relationship between parameters from discrete-time and continuous-time models (from Ockenden et al. (in press))

Structure: Discrete time A second-order discrete linear transfer function with no noise model, denoted by [2, 2, δ] takes the form: $$y(t) = \frac{b_1 + b_2 z^{-1}}{1 + a_1 z^{-1} + a_2 z^{-2}} u(t - \delta) \quad \text{(S6)}$$ where *y(t)* is model output at time *t*, *u(t)* is model input, z^{-1} is the backwards step operator i.e. $z^{-1}y(t) = y(t\text{-}1)$. b_1, b_2, a_1, a_2 are parameters determined during model identification and δ is the number of time steps of pure time delay. For a physical interpretation, models are only accepted it they can be decomposed by partial fraction expansion into two first order transfer functions with structure [1, 1, δ] representing fast and slow pathways, with characteristic time constants and steady state gains, i.e. $$y(t) = \frac{b_\mathrm{f}}{1 - a_\mathrm{f} z^{-1}} u(t - \delta) + \frac{b_\mathrm{s}}{1 - a_\mathrm{s} z^{-1}} u(t - \delta) \quad \text{(S7)}$$ where b_f and b_s are gains on the fast and slow pathways, respectively, and a_f and a_s are parameters characterising the time constants of the fast and slow pathways respectively. a_f and a_s are roots of the denominator polynomial in the second order transfer functions above (Eq. S6).
Structure: Continuous-time A second order continuous-time linear transfer function with no noise model takes the form: $$Y(s) = \frac{b_1 s + b_2}{s^2 + a_1 s + a_2} e^{-s\tau} U(s) \quad \text{(S8)}$$ where, *Y(s)* and *U(s)* represent the Laplace transforms of the output and input, respectively. b_1, b_2, a_1, a_2 are parameters in the denominator and numerator polynomials in the derivative operator $s = \frac{\mathrm{d}}{\mathrm{d}t}$ that define the relationship between the input and the output, and τ represents the delay. Models are only accepted if they can be decomposed by partial fraction expansion into two parallel, first-order transfer functions i.e. $$Y = \frac{b_\mathrm{f}}{s + a_\mathrm{f}} e^{-s\tau} U + \frac{b_\mathrm{s}}{s + a_\mathrm{s}} e^{-s\tau} U \quad \text{(S9)}$$ where a_f and a_s are direct reciprocals of the fast and slow time constants respectively, which define the fast and slow components of the response. b_f and b_s are parameters which determine the gain of the fast and slow components, respectively.

Relationship between parameters for discrete-time and continuous-time models

Parameters b_1, b_2, a_1, a_2 (and parameters b_f , b_s , $\mathrm{a}_{\mathrm{f},}$ a_s) have different interpretation, and therefore different values between discrete-time and continuous-time models. The relationship between the parameters (see most Control Engineering textbooks, (e.g. Franklin et al., 2002) between discrete model denoted by superscript d and continuous time model denoted by superscript c is as follows:
for instance, for denominator parameter a_f

$$a_\mathrm{f}^\mathrm{d} = e^{-a_\mathrm{f}^\mathrm{c}\Delta t} \tag{S10}$$

while for b_f we have:

$$b_\mathrm{f}^\mathrm{d} = \frac{b_\mathrm{f}^\mathrm{c}}{a_\mathrm{f}^\mathrm{c}}(1 - e^{-a_\mathrm{f}^\mathrm{c}\Delta t}) \tag{S11}$$

Table S4 Definition of time constants, steady state gains and fraction on each pathway for discrete-time and continuous-time models, e.g. for second order model, following partial fraction decomposition according to SI Eq. S7 (discrete-time) or SI Eq. S9 (continuous-time)

	Discrete-time	Continuous-time
Time constants (fast, slow)	$\frac{\Delta T}{-log_{\mathrm{e}}(a_{\mathrm{f}}^{\mathrm{d}})}$; $\frac{\Delta T}{-log_{\mathrm{e}}(a_{\mathrm{s}}^{\mathrm{d}})}$	$\frac{1}{a_{\mathrm{f}}^{\mathrm{c}}}$; $\frac{1}{a_{\mathrm{f}}^{\mathrm{c}}}$
Steady state gains	$SSG_1 = \frac{b_{\mathrm{f}}^{\mathrm{d}}}{1-a_{\mathrm{f}}^{\mathrm{d}}}$; $SSG_2 = \frac{b_{\mathrm{s}}^{\mathrm{d}}}{1-a_{\mathrm{s}}^{\mathrm{d}}}$	$SSG_1 = \frac{b_{\mathrm{f}}^{\mathrm{c}}}{a_{\mathrm{f}}^{\mathrm{c}}}$; $SSG_2 = \frac{b_{\mathrm{f}}^{\mathrm{c}}}{a_{\mathrm{f}}^{\mathrm{c}}}$
Fraction on each pathway	$\frac{SSG_1}{SSG_1+SSG_2}$; $\frac{SSG_2}{SSG_1+SSG_2}$	$\frac{SSG_1}{SSG_1+SSG_2}$; $\frac{SSG_2}{SSG_1+SSG_2}$

Table S5

Model structure and parameters identified, including uncertainty from 10,000 Monte Carlo realisations (from Ockenden et al. (in press))

Model structures and parameters for DBM models used in simulations								
Site	Model output	Model input	Model structure	β	a1	a2	b1	b2
Newby, Eden	Discharge Q	Rainfall R	Continuous [2, 2, 1]	0.37	0.3474 ± 0.0064	0.0023 ± 0.0001	0.1646 ± 0.0026	0.0026 ± 0.0001
Newby, Eden	Total P load TP	Effective rainfall Re	Continuous [1, 1, 1]		0.6429 ± 0.0191		2.0086 ± 0.0562	
Blackwater, Wensum	Discharge Q	Rainfall R	Discrete [2, 2, 6]	0.65	-1.9324 ± 0.0021	0.9325 ± 0.0021	0.0526 ± 0.0012	-0.0521 ± 0.0012
Blackwater, Wensum	Total P load TP	Rainfall R	Continuous [2, 2, 4]		0.0826 ± 0.0018	0.00021 ± 0.00003	0.0335 ± 0.0012	0.00016 ± 0.00002
Wylye, Avon	Discharge Q	Rainfall R	Discrete [2, 2, 6]	0.59	-1.7785 ± 0.0109	0.7790 ± 0.0108	0.0440 ± 0.0016	-0.0428 ± 0.0015
Wylye, Avon	Total P load TP	Effective rainfall Re	Continuous [2, 2, 6]		0.1660 ± 0.0080	0.00029 ± 0.00003	1.3015 ± 0.0506	0.0054 ± 0.0006

Figure S1

Hourly streamflow (Q) against total phosphorus (TP) concentration for the Newby Beck catchment, with the rising limb of storm hydrographs in blue and the falling limb of hydrographs in red.

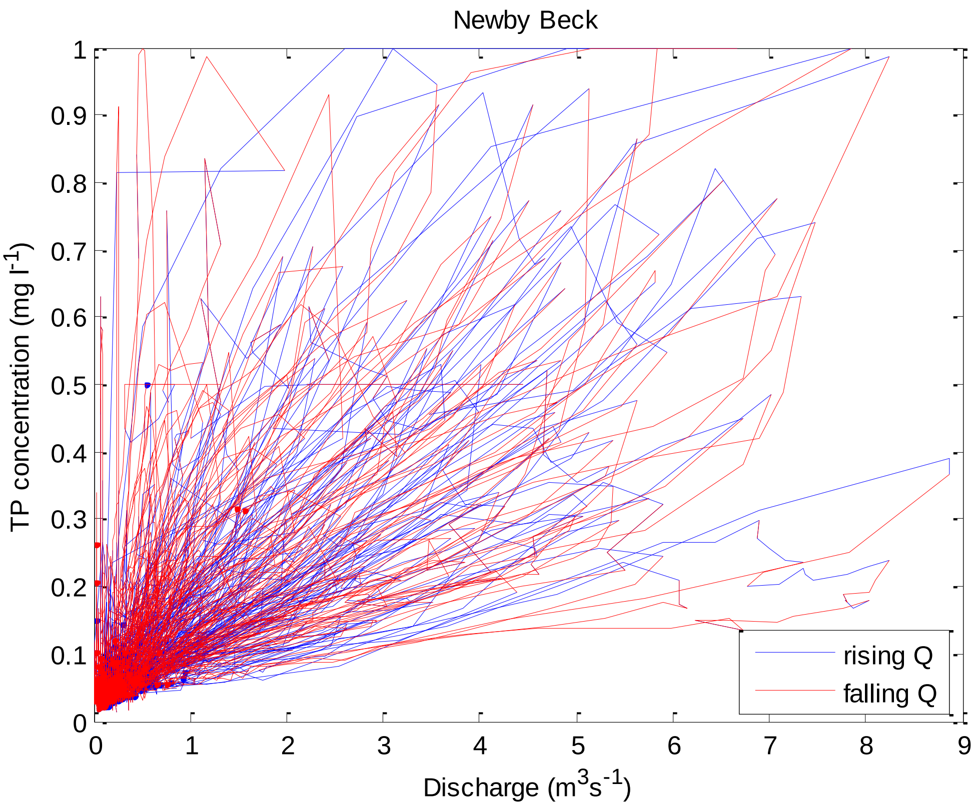


Figure S2

Hourly streamflow (Q) against total phosphorus (TP) concentration for the Blackwater catchment, with the rising limb of storm hydrographs in blue and the falling limb of hydrographs in red.

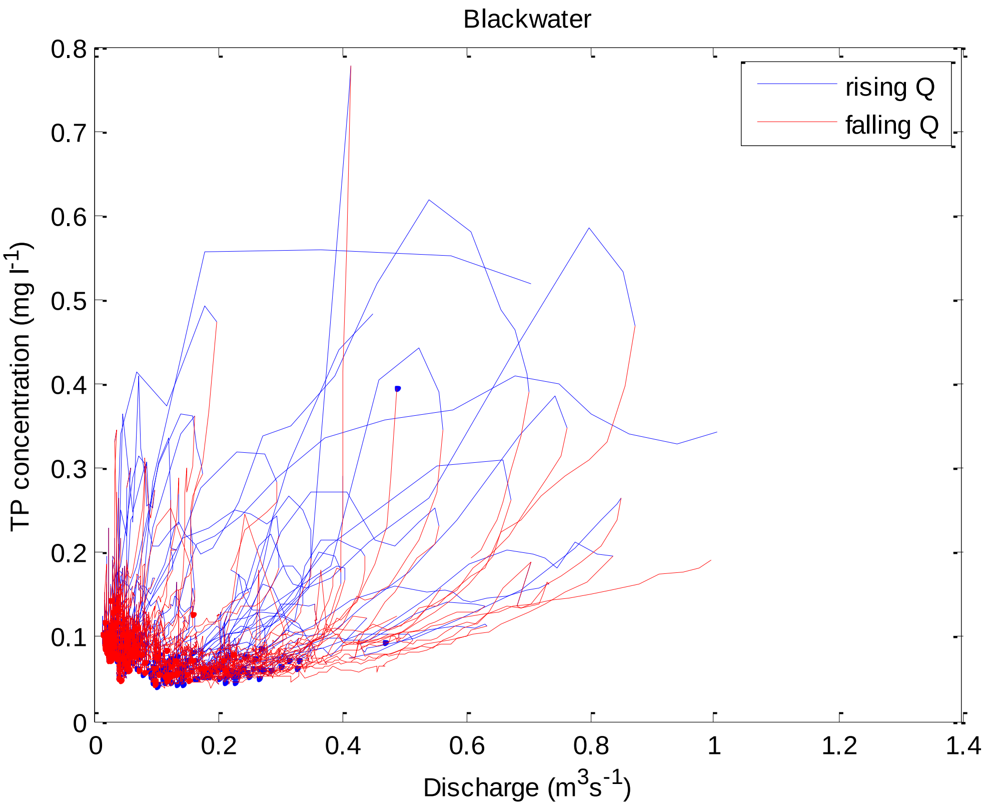
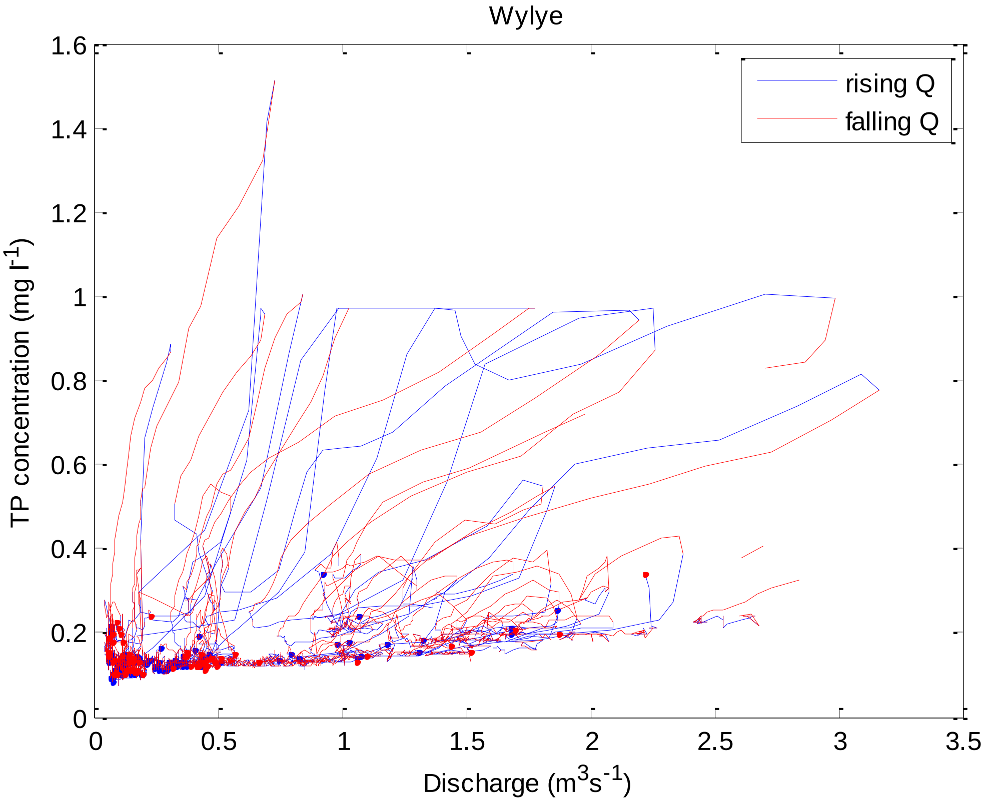


Figure S3

Hourly streamflow (Q) against total phosphorus (TP) concentration for the Wylye catchment, with the rising limb of storm hydrographs in blue and the falling limb of hydrographs in red.



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