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Key Points:
- Streamflow is the most important factor that drives water quality temporal variability in most catchments
- Antecedent flows, soil moisture, vegetation cover, and water temperature also help to explain temporal variability
- Relationships between temporal variability of water quality and its driving factors vary between catchments

Supporting Information:
- Supporting Information S1

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Key Factors Affecting Temporal Variability in Stream Water Quality

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Abstract Understanding the factors that influence temporal variability in water quality is critical for designing water quality management strategies. In this study, we explore the key factors that affect temporal variability in stream water quality across multiple catchments using a Bayesian hierarchical model. We apply this model to a case study data set consisting of monthly water quality measurements obtained over a 20-year period from 102 water quality monitoring sites in the state of Victoria (Southeast Australia). We investigate six water quality constituents: total suspended solids, total phosphorus, filterable reactive phosphorus, total Kjeldahl nitrogen, nitrate-nitrite (NO_x), and electrical conductivity. We find that same-day streamflow has the greatest effect on water quality variability for all constituents. Additional important predictors include soil moisture, antecedent streamflow, vegetation cover, and water temperature. Overall, the models do not explain a large proportion of temporal variation in water quality, with Nash-Sutcliffe coefficients lower than 0.49. However, when considering performance on a site-by-site basis, we see high model performance in some locations, with Nash-Sutcliffe coefficients of up to 0.8 for NO_x and electrical conductivity. The effect of the temporal predictors on water quality varies between sites, which should be explored further for potential spatial patterns in future studies. There is also potential for further extension of these temporal variability models into a predictive spatiotemporal model of riverine constituent concentrations, which will be a useful tool to inform decision making for catchment water quality management.

Plain Language Summary Water quality in rivers can change greatly over time. Understanding the causes of these changes is important for managing water quality. In this study, we used a statistical modeling approach to identify the influences of these temporal changes across 102 catchments in Victoria, Australia. The models were based on monthly measurements of water quality indicators (sediments, nutrients, and salts) obtained over 20 years. We find that the streamflow is the most important influence on temporal changes in water quality. Additional important drivers include soil moisture, recent streamflow, vegetation cover, and water temperature. The effects of these influences on the temporal patterns of water quality vary between catchments. Catchment managers could use the results to identify catchments and periods with poor water quality and thus to develop localized management strategies.

1. Introduction

Rivers and streams around the world are experiencing increasing levels of contamination (Liu et al., 2012; Loucks et al., 2005; Nash, 1993; National Water Commission, 2005; Schwarzenbach et al., 2010; van Vliet et al., 2013). Increasing contamination can make water unsuitable for human uses and thus result in greater costs associated with treating the water to a suitable standard (Jiang, 2009). Furthermore, degraded riverine water quality has negative impacts on aquatic ecosystems (Smith et al., 1999; Vorosmarty et al., 2010). As such, there is a pressing need for mitigation strategies that can combat the deterioration of riverine water quality.

There can be substantial differences in stream water quality over both space and time (Ai et al., 2015; Bengraïne & Marhaba, 2003; Chang, 2008; Vega et al., 1998). For example, across monitoring stations, average suspended sediment concentrations can vary between less than 10 and up to 1,700 mg/L (Meybeck &

Helmer, 1989). Constituent concentrations can vary at different time scales including event (Saraceno et al., 2009), daily (Pellerin et al., 2012), seasonal (Arheimer & Lidén, 2000), and interannual (Larned et al., 2004). For example, Vaishali and Punita (2013) reported variations in average electrical conductivity (EC) levels from 1,517 to 5,423 μS/cm, when comparing conditions before and after winter at the same location. Therefore, understanding and modeling these spatiotemporal variabilities is critical to developing effective strategies to improve and manage stream water quality.

Central to understanding the spatiotemporal variabilities in riverine water quality is the identification of key physical processes and controls of these variabilities. The large spatial and temporal variability witnessed in riverine water quality is driven by three key processes that affect stream water quality, which can vary greatly over both time and space: (1) source: the amount of the constituent within the catchment; (2) mobilization: the detachment of these constituents from the source, due to weathering, erosion or biogeochemical processing; and (3) delivery: the transport of mobilized constituents from the catchment to receiving waters (Granger et al., 2010). Regarding controlling factors, spatial variability in water quality can be affected by both human activities in the catchment (e.g., land use, vegetation cover, and land management) and natural catchment characteristics (e.g., climate, geology, soil type, topography, and hydrology; Onderka et al., 2012; Tramblay et al., 2010). At the same time, temporal shifts in water quality can be influenced by factors such as streamflow (Ahearn et al., 2004; Mellander et al., 2015; Sharpley et al., 2002), which influences seasonal variability in the delivery of the constituent to receiving waters, and rainfall (Fraser et al., 1999) and air temperature (Lecce et al., 2006; Robson, 2014), which affect the mobilization and transport of constituents in the catchment. Water temperature can also control biogeochemical processing of nutrients in streams (Roberts & Mulholland, 2007). In addition, the amount of the constituent in the catchment (source) is influenced by antecedent dry weather period (Arheimer & Lidén, 2000; Lecce et al., 2006), vegetation cover (Kaushal et al., 2014; Ouyang et al., 2010), and seasonal variability in human activities in the catchment (Stutter et al., 2008).

While both spatial and temporal variability are important in riverine water quality, this study focuses on the temporal variability in water quality across a region of approximately 200,000 km^2, guided by a previous study of the spatial variability in the same region (Lintern et al., 2018). In the existing literature, conceptual or physically based distributed models (Argent et al., 2009; Arnold & Fohrer, 2005) have been widely used to explore temporal variability in water quality. However, these models implicitly apply a wide suite of model assumptions, some of which are strongly contested, for example, those relating to representation of transport pathways and transit times (Hrachowitz et al., 2016). They also rely on extensive data sets and efforts for model calibration, particularly when considering large regions with high heterogeneity in catchment conditions and the key physical processes that govern water quality (Abbaspour et al., 2015). This has largely limited the scales of obtained understanding on water quality variability, both spatially (Luo & Zhang, 2009) and temporally (Boskidis et al., 2010; Poudel et al., 2013). On the other hand, data-driven statistical models can allow more flexible structures to account for spatial heterogeneity. However, most existing applications of statistical water quality models focus on temporal variability at only a single location (Kisi & Parmar, 2016; Kurunç et al., 2005; Parmar & Bhardwaj, 2015). There are a few examples of temporal analyses of riverine water quality that have examined behavior across multiple locations (Chang, 2005; Mei et al., 2014). However, these studies have had limited spatial extents. Consequently, there is a general lack of understanding of the catchment characteristics influencing spatial differences in the temporal dynamics of water quality across large spatial scales in the existing literature.

Targeting this knowledge gap, this study aims to understand the key factors that influence temporal water quality variability across large scales. We use a statistical modeling approach, which enables us to account for the abovementioned spatial heterogeneity. A hierarchical structure was chosen for the model, due to its capacity to simultaneously account for temporal trends at multiple locations (Vietz et al., 2018; Webb & King, 2009). The insights provided by our investigation can potentially lead to methods that assist catchment managers in designing water quality mitigation and restoration strategies.

2. Materials and Methods

We investigate six water quality constituents that are common concerns for stream water quality in Australia, namely, total suspended solids (TSS), total phosphorus (TP), filterable reactive phosphorus (FRP), total Kjeldahl nitrogen (TKN), nitrate-nitrite (NO_x), and electrical conductivity (EC). To investigate the key drivers of temporal water quality variability across catchments, we used a Bayesian hierarchical modeling framework to describe relationships between temporal variations of water quality (predictands) and a number of temporally varying driving factors including climatic and hydrologic conditions (predictors) across large number of catchments (section 2.1). At each catchment, linear regression was used to describe how water quality constituents change according to their key driving factors. We chose to use linear models with assuming normality of residuals, because this improved convergence rates of the models within the computationally intensive variable selection and fitting process (Gelman et al., 2013). To suit the linear model structure, data for both the temporal variables and the water quality were acquired and then transformed to maximize symmetry (as detailed in sections 2.2 and 2.3.1, for the predictands and predictors, respectively). For each water quality parameter, the optimal linear model structure was determined via two steps: (1) selection of the best set of temporal predictors and (2) estimation of the best fit values of corresponding model parameters (section 2.3). Postmodeling analyses were also carried out to further assess (a) the model performance and (b) the key factors driving temporal variability of water quality (section 2.4).

2.1. Overarching Model Structure

The concentration of a constituent (TSS, TP, FRP, TKN, NO_x, and EC) at time i and site j (C_{ij}) is assumed to be normally distributed with a mean μ_{ij} and a global standard deviation σ (equation (1)). The mean, μ_{ij}, is modeled as the sum of the observed time-averaged mean constituent concentration at site j ($\overline{C}_{j\text{obs}}$) and the deviation from the mean at time i at the same site (Δ_{ij}) (equation (2)). Here we focus on modeling of temporal variability (represented by deviations from mean at different time steps). This temporal variability is described by subtracting the observed site-level mean, $\overline{C}_{j\text{obs}}$, from the constituent concentration at each time step at that site, μ_{ij} (left-hand side of equation (3)). Subtracting the site-level mean allows us to eliminate the impacts of spatial variability in stream water quality. We can then model temporal variability at each site as a function of the observed site-level standard deviation of the constituent (σ_{j_obs}), and a linear effect of n temporal variables, T_1 to T_n (e.g., climate condition, streamflow, and vegetation cover; right-hand side of equation (3)). We refer to equation (3) from here on as the *temporal variability model* of water quality.

$$C_{ij} \sim N\left(\mu_{ij}, \sigma\right) \tag{1}$$

$$\mu_{ij} = \overline{C}_{j\_\text{obs}} + \Delta_{ij} \tag{2}$$

$$\Delta_{ij} = \mu_{ij} - \overline{C}_{j\_\text{obs}} = \sigma_{j\_\text{obs}}\left(\beta\_T_{1,j} \times T_{1,ij} + \cdots + \beta\_T_{n,j} \times T_{n,ij}\right) \tag{3}$$

Temporal variability at each site is represented by terms in the bracket in the right-hand side of equation (3), which is scaled up by the site-level standard deviation. The scaling was applied so that variabilities would be on a comparable scale between sites, and thus, the parameter values ($\beta\_T_{1,j}, \ldots, \beta\_T_{n,j}$) would also be comparable between sites.

To account for variations of the key controls for water quality temporal variability across sites, we used a hierarchical model structure which allows site-specific parameter values that describe the effects of each temporal variables ($\beta\_T_{1,j}, \ldots, \beta\_T_{n,j}$). A Bayesian approach is selected for parameter estimation (calibration) due to its high capacity for resolving complex model structures (e.g., Borsuk et al., 2001; Obenour et al., 2014). The Bayesian hierarchical model assumes that site-specific values of each temporal parameter $\beta\_T_{n,j}$ are from a common prior distribution defined by parameters referred to as *hyperparameters*. During the model calibration, increasing numbers of samples are drawn, which allows the prior distribution to gradually converge into the corresponding posterior distribution that maximizes the likelihood of model fit. The Bayesian hierarchical model enables us to model explainable processes as well as stochastic unexplainable processes (Clark, 2005). Furthermore, such a hierarchical model structure can utilize time series data at multiple locations to strengthen the site-specific temporal models (also known as *borrowing strength*; Webb & King, 2009).

2.2. Data Collection and Preprocessing

Water quality data were extracted from the Victorian Water Quality Monitoring Network database (Department of Environment, Land, Water and Planning Victoria, 2016). This database contains monthly ambient water quality data measured at approximately 400 sites across the state of Victoria (Southeast

Australia). These water quality data date back to 1990 for some sites. From this study we extracted monthly water quality data sampled between 1994 and 2014 for 102 sites (Figure 1), which are the sites that contain continuous monthly measurements for the longest consistent period. The choice of calibration data set was influenced by two factors. First, to maximize the comparability of fitted coefficients between sites and to avoid the possible influence of decadal state-wide climatic fluctuations, we chose to concentrate on sites with good data availability. Second, to maximize both the record length and the number of sites included. Together, this provides us with higher confidence in identifying large-scale patterns in water quality temporal dynamics. The catchments corresponding to the water quality monitoring sites were identified using the Geofabric tool (Bureau of Meteorology, 2012), with areas ranging from 5 to 16,000 km^2.

The water quality constituents considered in this study were as follows: TSS, TP, FRP, TKN, nitrate-nitrite (NO_x), and EC. All water quality records that were flagged as potential errors or below the limits of reporting (LOR) were removed from the data set. It is worth noting that although there are well-established approaches to treat data below the LOR in water quality analyses (Helsel, 1990), we decided to remove these data. This is because that in this study, we are primarily concerned with understanding they key factors contributing to poor water quality (i.e., high constituent concentrations). Furthermore, data below the LOR (even the post-treatment records) are unlikely to represent the true constituent concentrations and would thus affect the accurate identification of temporal water quality variability.

To assist fitting of the linear relationship explaining temporal variation in water quality to observed data (equation (3)), all constituent concentrations were Box-Cox transformed using the *car* package in R (Fox & Weisber, 2011) to maximize symmetry in their distributions. For each constituent, the optimal Box-Cox parameter λ was first identified using the concentration data at each site; the average λ across all sites was then used to transform the constituent at once. This transformation approach ensures that all sites were being transformed using a consistent transformation parameter. The transformed water quality data were checked visually for symmetry (Figure S1 in the supporting information).

Model explanatory variables for each catchment were compiled from streamflow monitoring and climate data. Instantaneous flows (mm/day) and water temperature (°C) for each of the 102 sites between 1994 and 2014 were also extracted from the Water Quality Monitoring Network database. In addition, we used gridded weather data and normalized difference vegetation index (NDVI) data to calculate the catchment average daily rainfall (mm), daily evapotranspiration (ET; mm), daily average temperature (°C), daily root zone (less than 1-m depth) and deep (more than 1-m depth) soil moisture (both as % of available water holding capacity), and monthly NDVI (dimensionless; Table 1). The model does not consider the temporal impact of land use explanatory variables (e.g., measures of agricultural activity), as the temporal resolution of available data did not match that of the water quality monitoring data.

2.3. Selection of Optimal Model Structures

2.3.1. Selection of Model Predictors

Temporal variations in the concentrations of individual constituents are assumed to be driven by different combinations of temporal variables (T_1 to T_n in equation (3)). Therefore, we performed an exhaustive exploration of a large number of potential temporal predictors to identify the best combination of predictors for each constituent.

The search started from a range of potential temporal predictors including conditions of streamflow, climate, and land cover (Table 2). Temporal variability in constituent concentrations is largely a result of changes in (i) the amount of the constituent in the catchment (i.e., source), (ii) the amount of the constituent mobilized, and (iii) the amount of the constituent transported to receiving waters (Lintern et al., 2018). The amount of the constituent in the catchment is influenced by seasonal changes in human activities (e.g., fertilizer application; Hill, 1978; Houser & Richardson, 2010) and the amount of the constituent that has built up in the catchment since the previous flow event (Arheimer & Lidén, 2000; Stutter et al., 2008). In addition, specifically for nutrients, the rate and extent of vegetative uptake (Groffman et al., 2009; Mattsson et al., 2003) and transformation due to microbial activity during the antecedent dry weather period can be important (Lloyd et al., 2016; Robson, 2014). As such, hydroclimatic variables such as temperature and antecedent dry weather period (represented by antecedent average streamflow, water and air temperature, rainfall, and preceding continuous period of dry days) can influence temporal variability in stream water quality. In addition, variables such as seasonality (represented by day of the year), vegetation cover (represented by NDVI), temperature, and soil

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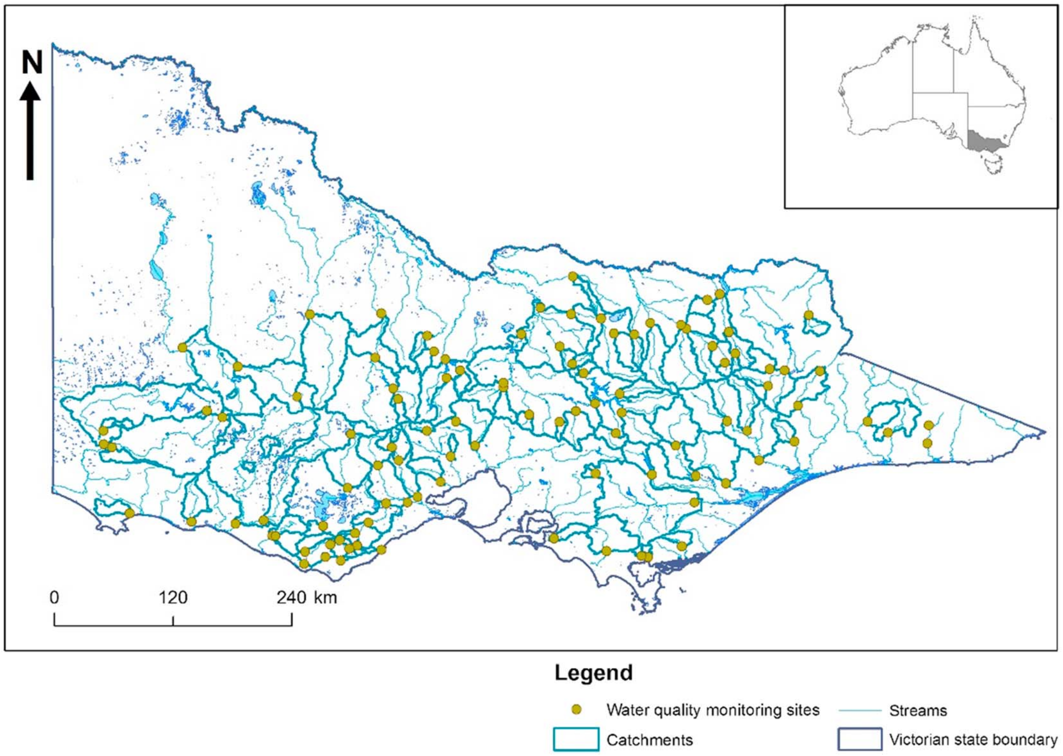

Figure 1. Maps of study sites. 102 water quality monitoring sites and their catchment boundaries used for analysis. Inset shows location of the state of Victoria in Australia.

moisture can be related to agricultural activities within the catchment (e.g., land clearing, cropping, and fertilization), as well as nutrient uptake by plants and transformation by microbial processing.

Variations over time in constituent mobilization can be influenced by the amount of erosion due to rainfall and runoff, particularly for sediments (Granger et al., 2010). The extent of erosion of particulates can be also be affected by human activities (e.g., tillage; Skaggs et al., 1994), vegetation cover (Meybeck et al., 1989), soil desiccation (Prosser et al., 2000), and mineralization of nutrients from soils (Arheimer & Lidén, 2000). As such NDVI, soil moisture and temperature (both water and air) could be important predictors of constituent mobilization.

Table 1
Sources for Climate and Land Cover Data

Data		Source	Grid size
Daily rainfall Daily average temperature		Australian Water Availability Project (Raupach et al., 2009, 2012) Available from http://www.csiro.au/awap; http://www.bom.gov.au/jsp/awap/index.jsp	5 km by 5 km
Daily actual ET Daily average root zone soil moisture Daily average deep soil moisture		Australian Water Resources Assessment (Frost et al., 2016) Available from http://www.bom.gov.au/water/landscape	5 km by 5 km
Monthly NDVI	January 1994 to December 1999	Advanced Very High Resolution Radiometer product (Eidenshink, 1992) Available from https://earthdata.nasa.gov/	1 km by 1 km
	January 2000 to December 2013	Moderate Resolution Imaging Spectroradiometer; MOD13A3 (NASA LP DAAC, 2017) Available from https://earthdata.nasa.gov/	1 km by 1 km

Note. ET = evapotranspiration; NDVI = normalized difference vegetation index.

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Table 2

Categories and Names of the 19 Potential Temporal Predictors for the Temporal Variation of Catchment Water Quality

Rainfall	Streamflow	Other same-day climate conditions	Vegetation
Rainfall on the same day	Streamflow on the same day	Water temperature	NDVI for the sampling month
Rainfall averaged over 1, 3, 7, 14 and 30 preceding days	Streamflow averaged over 1, 3, 7, 14, and 30 preceding days	Air temperature	
Dry spell length in the past 14 days		Evaporation	
		Root zone soil moisture	
		Deep soil moisture	

Note. NDVI = normalized difference vegetation index.

Transport of the constituents to receiving waters is generally influenced by streamflow generation processes (Lintern et al., 2018). Both particulate (e.g., TSS, TP, and TKN) and dissolved constituents (FRP and NO_x) are transported to receiving waters by surface flows. The dissolved constituents can also be transported by subsurface flows (Blanco et al., 2010; Mellander et al., 2015; Wood, 1977).

We included all predictors that represent instantaneous hydroclimatic conditions on the day of the water quality measurement: streamflow, rainfall, water and air temperature, ET, and soil moisture. We also considered the average streamflow and rainfall over different preceding periods, namely, 1, 3, 7, 14, and 30 days, which were calculated from the corresponding daily data set. The effect of antecedent dry spells was considered by counting the number of continuous dry days (daily rainfall = 0 mm) within the past 14 days. In addition, we also considered the vegetation cover with the monthly NDVI data. In all, a total of 19 temporal predictors was considered as potential predictors for water quality temporal variability (Table 2).

For each constituent, we explored all possible combinations (4,980,736 combinations) of these 19 potential predictors, which is referred to as *exhaustive search* from now on (Guyon & Elisseeff, 2003; May et al., 2011). Specifically, for each constituent, a linear regression was built between water quality temporal variability and each possible combination of temporal predictors (equation (3)). Considering the intensive computational requirement, for each combination of predictors, we fitted 102 site-specific linear regressions for water quality temporal variation instead of a full hierarchical model. Considering the variability of coefficients for each predictor across sites, identification of the key predictors cannot be done through conventional significance tests on individual predictors across all study catchments. Instead, we used the Bayesian information criterion (BIC) to summarize model performance and complexity and thereby select the best common set of predictors across all catchments (Burnham & Anderson, 2007; Saft et al., 2016; Schwarz, 1978). Specifically, for each site we first calculated the BIC scores for the different linear regressions fitted to all possible combinations of predictors and identified the best predictor set for that site, which led to the lowest BIC score. This approach allowed us to convert the absolute BIC scores for all possible predictor sets to differences to the lowest BIC (i.e., ΔBIC) at each site, which represents the *distance from the best model* at each site. As such, we were then able to average the ΔBIC scores for each possible predictor set across sites, and then to select the best predictor set based on the lowest average ΔBIC across sites.

It is worth noting that the data of the above-mentioned temporal predictors were highly skewed and relationships between untransformed variables were nonlinear; hence, transformations were used to satisfy the linearity assumption in the temporal variability model (equation (3)). To resolve this, before fitting the linear models we transformed each predictor using the Box-Cox transformation within the *car* package in R (Fox & Weisber, 2011), in a similar way to the transformation of the constituent concentrations (section 2.2). The symmetry of the transformed data for each temporal variable was checked visually (Figure S2 in the supporting information).

To account for any additional seasonal patterns in catchment water quality that are independent of these hydroclimatic and vegetation effects, we considered day of the year for all the dates of measurement for each constituent (i.e., day of the year J, $1 \leq J \leq 366$) as additional predictors to the temporal predicators for each constituent. Considering the seasonal effects was not part of the exhaustive search, but after the *best performing* models had been determined from the exhaustive searches. This is due to the potentially high cross correlations between measurement dates and hydroclimatic and vegetation conditions. Specifically, the

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sine and cosine of the day of the year of all measurement dates were calculated, which can together define a sinusoidal function to describe seasonality as equation (4):

$$a\ \sin\left(\frac{2\pi J}{365}\right) + b\ \cos\left(\frac{2\pi J}{365}\right) = A\ \sin\left(\frac{2\pi J}{365} + P\right) \tag{4}$$

where a and b are coefficients, with the phase shift and amplitude of seasonal variation defined by P and A, respectively. With inclusion of both the seasonality predictors (transformed and standardized as per other predictors), the average ΔBIC value was calculated again for each constituent, which was then compared with the average ΔBIC for the functions containing only the temporal predictors selected from the exhaustive search. This additional analysis for seasonal effects informs whether additional seasonality effects can improve the model fit without adding unnecessary complexity (for which the additional day of the year predictors will lead to a decrease in ΔBIC) and, thus, whether the seasonality predictors should be included in the model in addition to the selected temporal predictors.

2.3.2. Estimation of Model Parameters

Following the identification of the best set of predictors for each constituent, the temporal component of the spatiotemporal model (equation (3)) was fitted across all monitoring sites for each constituent, within a Bayesian hierarchical structure. To achieve this, we used the R package *rstan* (Stan Development Team, 2018), which enables both the sampling of parameter values from prior distributions with Markov chain Monte Carlo and model evaluation. It is worth noting that prior to model calibration for each constituent, the data of all predictors were standardized. In this way, the calibrated parameter values are indicative of the relative importance of effects of those temporal predictors.

Constituent standard deviation (σ) was assumed to be drawn from a minimally informative zero-truncated normal distribution with mean of 0 and standard deviation of 10 (Gelman, 2006; Stan Development Team, 2018). The site-level regression coefficients of the climate predictors ($\beta\_T_{1,j}$, $\beta\_T_{2,j}$, …, $\beta\_T_{n,j}$ in equation (3)) were assumed to be drawn from the corresponding hyperparameter normal distribution with means of $\mu.\beta\_T_1$, $\mu.\beta\_T_2$, …, $\mu.\beta\_T_n$ and standard deviations of $\sigma.\beta\_T_1$, $\sigma.\beta\_T_2$, …, $\sigma.\beta\_T_n$. These hyperparameters were further assumed to be drawn from minimally informative normal distributions with means of 0 and standard deviations of 5 (for the means) and minimally informative zero-truncated normal distribution with mean of 0 and standard deviation of 10 (for the standard deviations).

In each model run there were three independent Markov chains. A burn-in of 1000 iterations and total iterations of 2,000 were used for each chain. Convergence of the chains was checked using the *Rhat* value (Sturtz et al., 2005). We assessed the model fit using the Nash-Sutcliffe coefficient (NSE; Nash & Sutcliffe, 1970) as per Obenour et al. (2014).

2.4. Analyses of Model Outputs

2.4.1. Assessment of Model Performance

We first assessed the performance of the calibrated models by the fit of the temporal variability component of each constituent (i.e., Δ_{ij} in equation (3)) across all 102 sites and calculated a NSE (Nash & Sutcliffe, 1970) for all data. The site-specific NSE values were also calculated to assess how model performance varies with site. As an alternative way of assessing the model fit, we also compared the observed and simulated concentration of each constituent, which is obtained by adding the observed site-level mean concertation to both the fitted and observed temporal variability component (i.e., yielding μ_{ij} in equation (2)). Inclusion of the site-level mean concentration informs the impact of the temporal model performance on the concentration of each constituent. Lastly, the model residuals were checked for normality, to inform the sufficiency of the linear model structure.

2.4.2. Assessment of Key Factors Driving Temporal Water Quality Variability

To assess the importance of the key factors that explain temporal variability in water quality across the 102 sites, for each constituent we investigated the calibrated regression coefficients for each predictor variable within the temporal variability model (i.e., eff. $T_{1,\ j}$ to eff. $T_{n,\ j}$ in equation (3)), for their magnitudes and consistency across sites. To further understand the relative importance of those key driving factors of temporal variability, we also compared the temporal variability model of each constituent with an additional model, which was fitted between water quality temporal variability and only the most important predictor for each constituent.

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3. Results

3.1. Model Performance

Temporal variability in water quality remains largely unexplained by temporal changes in climate, streamflow, and vegetation cover (Figure 2). Among all of the constituents, we have the highest predictive power for the NO_x model, with a NSE of 0.49. While temporal variability explained by the models is relatively small when considering all sites together (Figure 2), the site-specific NSEs show that performance varies across sites, with up to 80% temporal variation explained for NO_x and EC at some sites (Figure 3).

Although the models had relatively low NSEs for the prediction of temporal variability in water quality, model fitting for the true constituent concentrations (i.e., when the observed site-level mean is added to the modeled temporal variability to simulate μ_{ij} in equation (2)) was generally satisfactory. This is evidenced with NSE values exceeding 0.53 (Figure 4), suggesting that with known site-level mean concentration for each constituent, the models can explain more than half of the observed spatiotemporal variability. The comparison between Figure 4 and Figure 2 suggests that NO_x is the only constituent for which temporal predictors can explain a substantial proportion of the spatiotemporal variability (NSE = 0.486 for temporal variability compared with NSE = 0.703 for true concentration). However, for the remaining constituents, the majority of spatiotemporal variability can be explained by the site-level mean constituent concentrations, since the low NSEs in fitting the temporal variability component (ranging between 0.135 and 0.33) substantially improve when considering the concentration instead (ranging between 0.536 and 0.883).

Analyses of residuals of the temporal variability models suggested that there is no clear trend in the residuals over time (Figure S3 in the supporting information). The residuals also do not appear to follow the temporal trend in observed concentrations (Figure S4 in the supporting information). In addition, these residuals are not correlated with any of the key temporal predictors—streamflow, water temperature and deep soil moisture (Figure S5 in the supporting information).

3.2. Key Driving Factors of Temporal Water Quality Variability and Their Effects

Results of the exhaustive search for model predictors suggested that streamflow (both on the same-day and over preceding days), water temperature, and soil moisture were important predictors for all constituents. Vegetation cover (as represented by the NDVI) is influential only for nitrogen species. Rainfall variables (both on the same-day and over preceding days), air temperature, and ET were not key predictors for any constituent (Table 3). In addition, when seasonality (sine and cosine of the day of the year) was added to the six best sets of predictors, all the ΔBIC values increased, suggesting that considering seasonality effects in addition to hydroclimate and vegetation conditions cannot provide a substantial improvement in model performance to compensate the increase in model complexity. Thus, we did not include these seasonality predictors in our models.

Figure 5 presents the effects of the important temporal predictors for each of the six constituents, as the regression coefficients for each predictor ($\beta\_T_{1,j}$, $\beta\_T_{2,j}$, …, $\beta\_T_{n,j}$ in equation (3)). Note that since we allowed the values of each coefficient to vary across sites (as detailed in section 2.1), each box in Figure 5 is used to represent the variability across all 102 sites and does not relate to statistical significance at any particular site. As all variables were standardized, the overall magnitudes of the effects are indicative of the relative importance of the temporal parameters in the model. It is clear that same-day streamflow is the most influential predictor of temporal changes in water quality for all constituents. The key role of instantaneous streamflow is in line with many studies that have modeled water quality changes as a function of streamflow (e.g., Sharpley et al., 2002; Wood, 1977). We provide some more in-depth discussion on the effects of each temporal variable, as well as the possible physical processes that they inform in section 4.1.

Using only streamflow as a predictor of water quality leads to NSEs of 0.06 (TSS) to 0.31 (EC; Table 4). In comparison, inclusion of other hydroclimatic predictors in the models (i.e., the full temporal variability models, with NSEs shown in Figure 2) leads to overall improvement in the model performance, ranging from 29% (EC) to 232% (TP) increase in NSE. The greatest increase in model performance is seen for TP (232%) and TKN (177%). The improvements in model performance from including variables other than streamflow may indicate a direct causal effect of these variables or that they are surrogates for other important (but unobserved) driving processes.

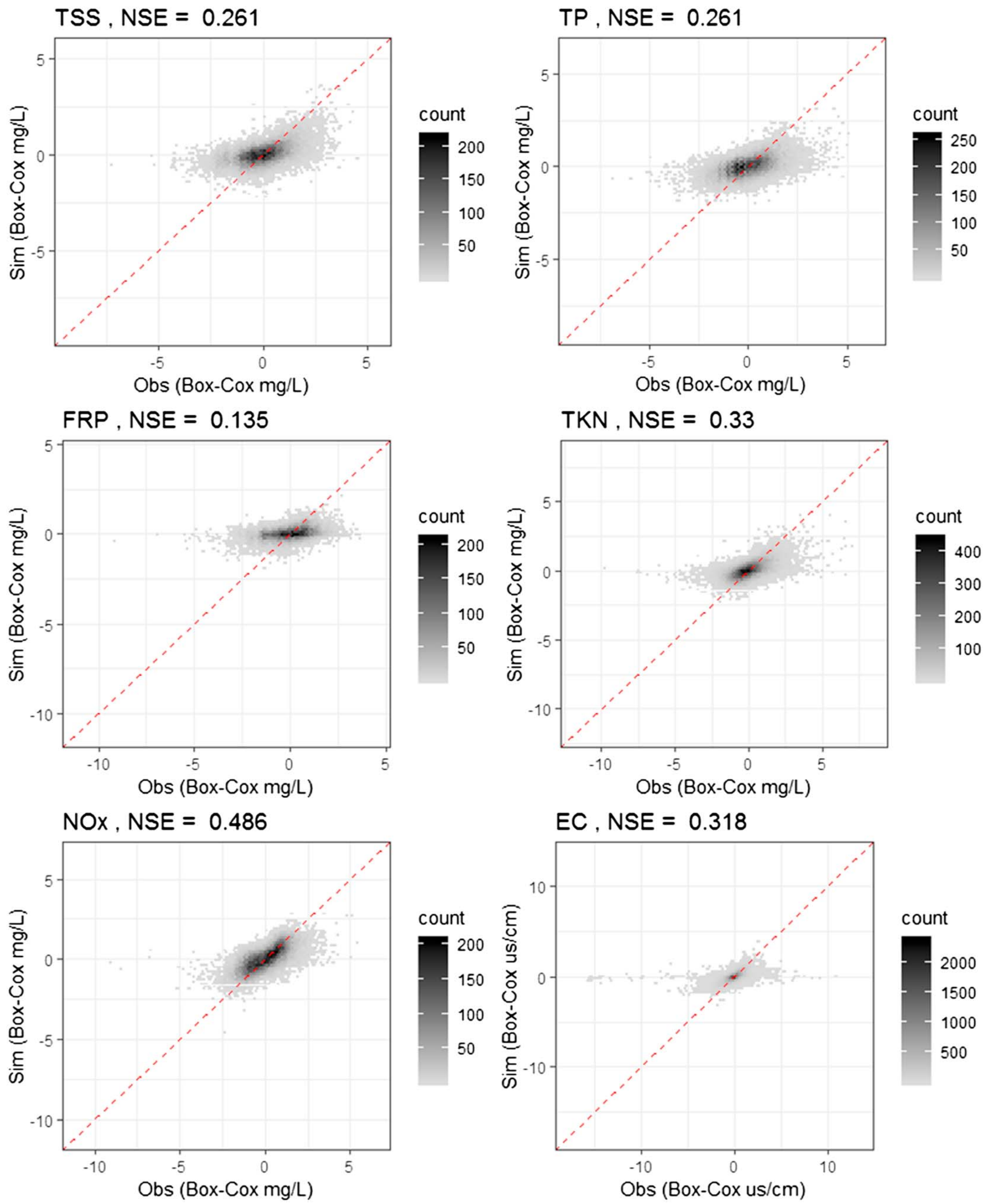

Figure 2. Performance of the Bayesian hierarchical models for the temporal variability of the six constituents across 102 sites, represented by the simulated and observed temporal variations across all sites, away from the corresponding site means. Darker regions represent denser distribution of values of simulations and observations. The Nash-Sutcliffe coefficients for each constituent are also shown. Dashed red lines represent the 1:1 lines. TKN = total Kjeldahl nitrogen; NSE = Nash-Sutcliffe coefficient; FRP = filterable reactive phosphorus; TP = total phosphorus; TSS = total suspended solid; EC = electrical conductivity.

4. Discussion

Section 3.2 presents the effects of the key temporal variables that affect temporal variability in catchment water quality, which we obtained using long-term monitoring data with a Bayesian hierarchical model. We concluded that streamflow is the most important control on temporal variability. In this section we provide a more in-depth discussion on the effects of streamflow, as well as other important factors for temporal variability, and suggest some possible physical processes that are related to these key factors (section 4.1). Further

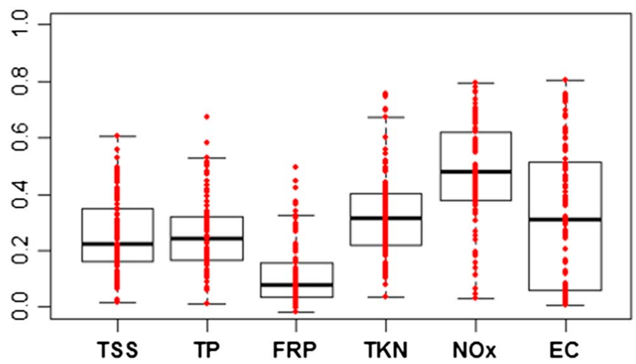

Figure 3. Distribution of the Nash-Sutcliffe coefficients at individual sites, for the six constituents. TSS = total suspended solid; TP = total phosphorus; FRP = filterable reactive phosphorus; TKN = total Kjeldahl nitrogen; EC = electrical conductivity.

to these understandings, we discuss how the key findings and experiences from this modeling exercise can inform future analyses and sampling that can help catchment water quality management, these include (a) design of predictive models for catchment water quality (section 4.2) and (b) design of more efficient sampling strategies (section 4.3).

4.1. Key Factors and Processes that Drive Temporal Variability in Water Quality

Streamflow. For all constituents, the effect of same-day streamflow is the largest in magnitude. For most constituents (TSS, TP, FRP, TKN, and NO_x in Figures 5a–5e), same-day streamflow has a positive impact on water quality at most locations (i.e., leading to increasing concentrations). These highlight the key role of surface flow in driving the temporal variability of sediments and nutrients and also indicate higher importance of the mobilization and delivery processes (for which constituent increases with streamflow) compared with dilution (for which constituent decreases with streamflow). Some potential pathways via which increasing streamflow can enhance the delivery of sediments and nutrients include (1) higher streamflow being able to transport more constituents to streams (Ahearn et al., 2004; Mellander et al., 2015) and (2) increased erosive power of greater streamflow, which can mobilize sediments and adsorbed nutrients in the catchment and/or channels (Donohue et al., 2005; Drewry et al., 2006).

For EC on the other hand, there is a negative effect of flow on EC (Figure 5f) at most sites. This relationship can be explained by the fact that salts (i.e., general ions) are generally transported to receiving streams through subsurface flows, so that when streams are dominated by subsurface flow contributions during dry periods, the concentrations in streams are likely to increase (Ahearn et al., 2004; Trowbridge et al., 2010). During wet periods on the other hand, the finite supply of salts, which are being transported to receiving streams by subsurface flows, is diluted by surface runoff containing lower salt levels (Vaze et al., 2004). It is worth noting that approximately 10% of sites exhibit positive relationships between EC and streamflow (Figure 5f). It is possible that these relationships are a result of the long-term changes in streamflow that have occurred at some sites due to the Millennium Drought throughout southeastern Australia between 1997 and 2009 (Saft et al., 2015). The decreasing trend in streamflow during the drought leads to more frequent low flow conditions, which might be influencing the relationship between EC and streamflow to become more positive.

Antecedent streamflow. Antecedent streamflow (7, 14, and 30 days) was the second most important predictor for most constituents. At most sites, TSS, TP, TKN, and EC are affected negatively by the antecedent streamflow (Figures 5a, 5b, 5d, and 5f). This suggests that during prolonged high flow periods, the concentrations of TSS, TP, TKN, and EC decrease, possibly due to the dilution of an existing supply of these constituents in the catchment (Warner et al., 2009), while supply builds up in the catchment during dry periods.

NO_x has positive regression coefficients for antecedent streamflow (30 days) at about half of all sites (Figure 5e). This relationship indicates increased NO_x concentrations following wet periods at these sites. NO_x can be transported to receiving streams by subsurface flows, especially along shallow pathways that leach nitrate from the soil profile. These shallow pathways are activated during wet conditions (Saffarpour et al., 2016). Indeed, increased leaching of nitrate to receiving waters through subsurface flows has been identified after wet years (Aubert et al., 2013; Bende-Michl et al., 2013).

Soil moisture. Soil moisture in the deep soil is important for all constituents, and soil moisture in the root zone is important for all constituents except FRP (Figures 5a, 5b, and 5d–5f). The importance of soil moisture is possibly a result of (i) the fact that it can affect the proportion of rainfall that is converted to surface runoff (Jiang et al., 2014), (ii) the activation of shallow subsurface pathways, and (iii) its influence on vegetation growth (and nutrient uptake) and the rate of soil erosion (Wood, 1977). Soil moisture can also govern the level of microbial activity and nutrient processing occurring in the soil (Arheimer & Lidén, 2000; Christopher et al., 2008).

However, the effect of soil moisture on water quality varies between constituents. For TSS and EC, the median effect of root zone and deep soil moisture is approximately 0 but ranges from negative at some sites to

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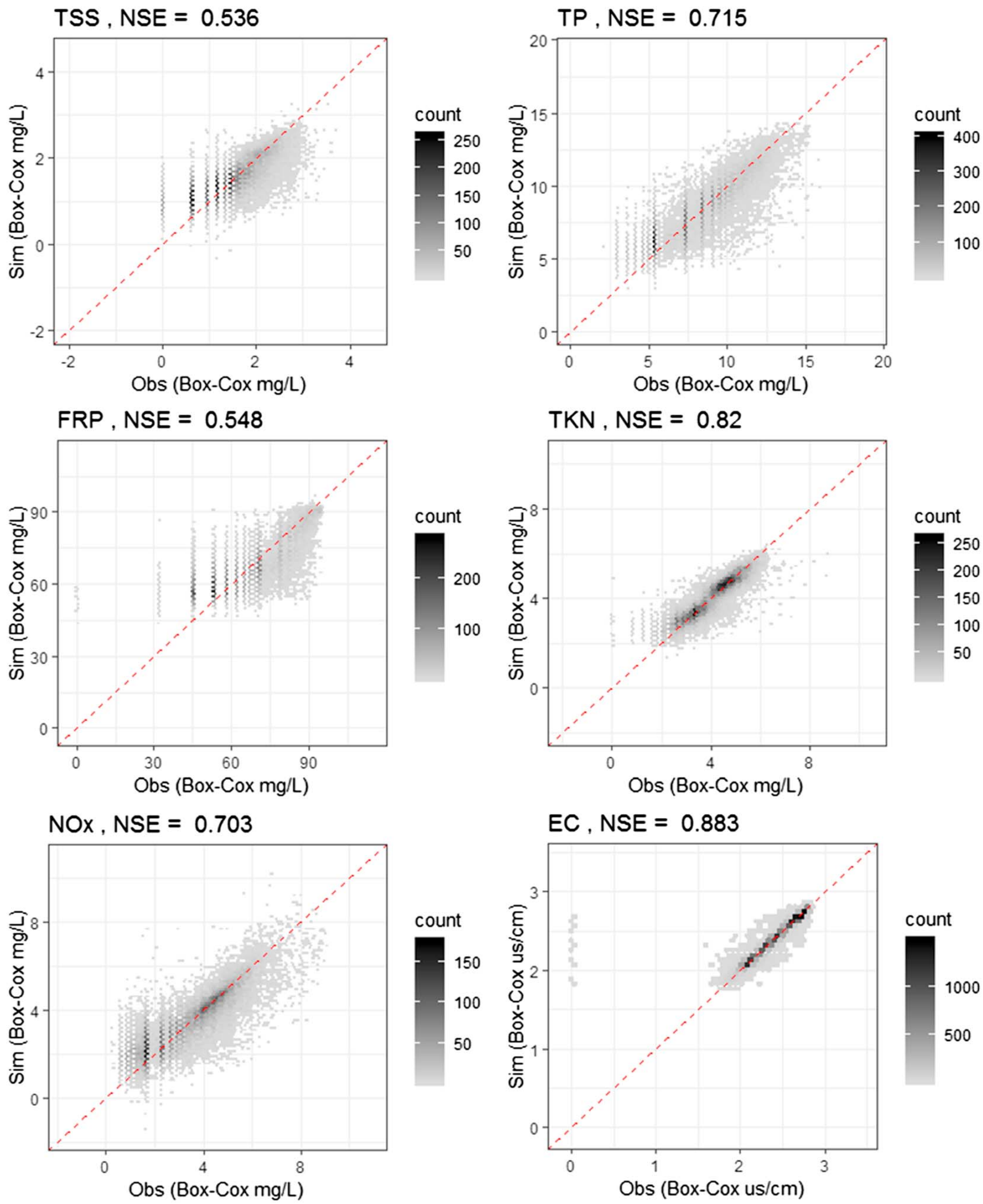

Figure 4. Performance of the Bayesian hierarchical models for the temporal variability of the six constituents across 102 sites, represented by the simulated and observed true concentration across all sites. These simulated true concentrations were obtained by adding the observed site-mean concentrations to the simulated temporal variability. Darker regions represent denser cluster of points. The Nash-Sutcliffe coefficients (NSEs) for each constituent are also shown. Dashed red lines represent the 1:1 lines. TSS = total suspended solid; TP = total phosphorus; FRP = filterable reactive phosphorus; TKN = total Kjeldahl nitrogen; EC = electrical conductivity.

positive at others (Figures 2a and 2f). For the nutrients for most sites (except FRP; Figures 2b, 2d, and 2e), there is a positive effect of root soil moisture on nutrient concentrations and a negative effect of deep soil moisture on water quality. Higher root soil moisture could lead to higher nutrient concentrations at most sites because of the greater surface and shallow subsurface runoff that is produced when the root zone soil moisture is high.

Table 3
Temporal Predictors Selected for the Bayesian Hierarchical Modeling of Each Constituent, From an Exhaustive Search Based on ΔBIC (Highlighted Cells)

	Rain preceding day(s)						Drysp	Flow preceding day(s)							Water	Air		SM	SM
WQ	0	1	3	7	14	30	14d	0	1	3	7	14	30	NDVI	*T*	*T*	ET	root	deep
TSS								■			■				■			■	■
TP								■					■		■			■	■
FRP								■							■				■
TKN								■					■	■	■			■	■
NO_x								■					■	■	■			■	■
EC								■				■			■			■	■

Note. BIC = Bayesian information criterion; WQ = water quality; NDVI = normalized difference vegetation index; ET = evapotranspiration; SM = soil moisture; TSS = total suspended solid; TP = total phosphorus; FRP = filterable reactive phosphorus; TKN = total Kjeldahl nitrogen; EC = electrical conductivity.

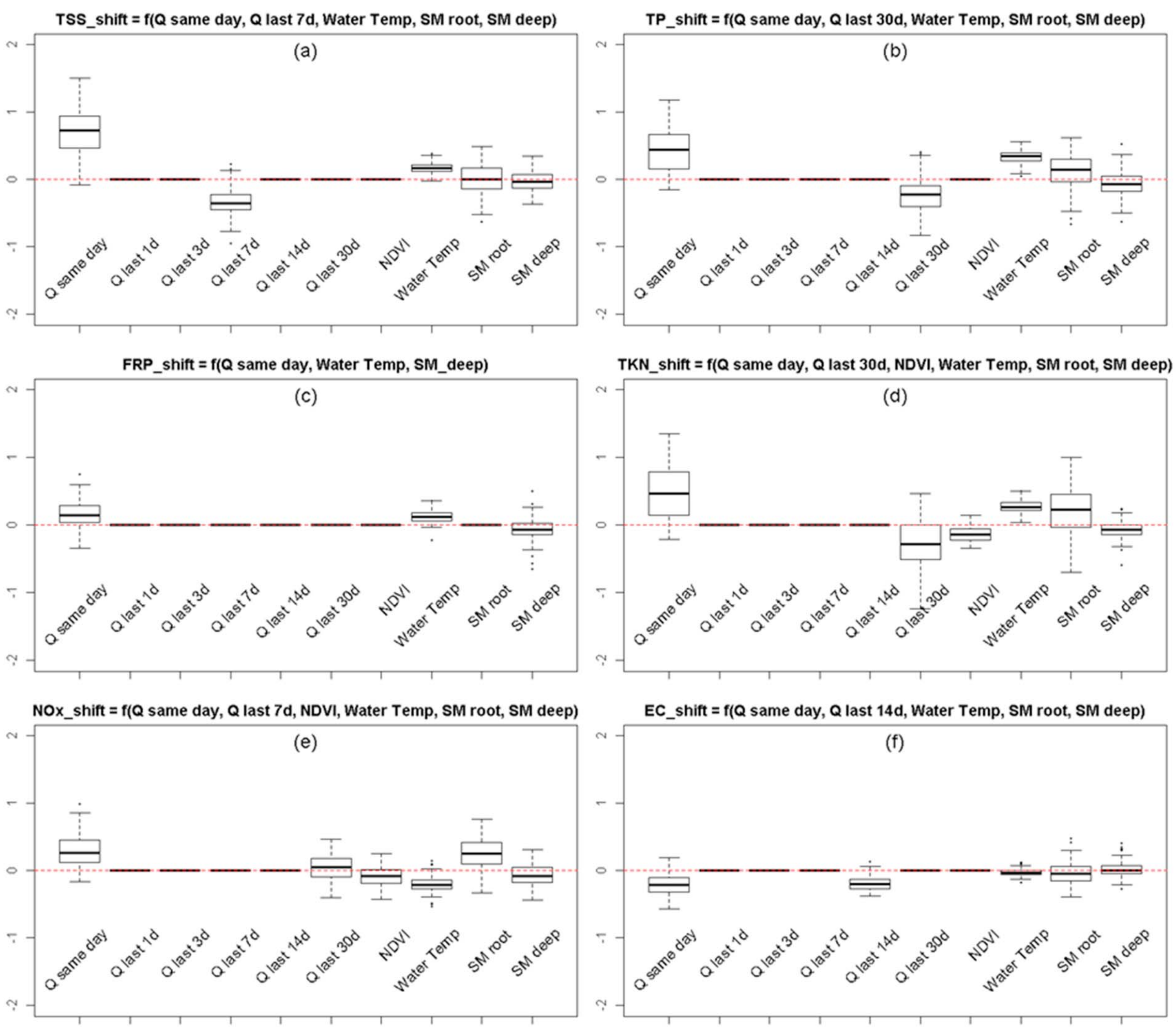

Figure 5. Effects of hydroclimatic predictors on the temporal variability of each constituent across 102 sites, summarized by the posterior mean of the calibrated parameter values for each predictor. *Y* axis shows the effect as the number of standard deviations away from site mean, as per equation (3). Note that only predictors identified for at least one constituent during the selection of predictors are shown on the *X* axis. TSS = total suspended solid; SM = soil moisture; TP = total phosphorus; FRP = filterable reactive phosphorus; TKN = total Kjeldahl nitrogen; NDVI = normalized difference vegetation index; EC = electrical conductivity.

Table 4

Comparison Between Model Performance Between Using (i) the Most Important Predictor (Streamflow-Only Models) and (ii) All of the Important Predictors (Full Models)

Constituent	NSE streamflow-only models	NSE full models	NSE improvement
TSS	0.15	0.26	70%
TP	0.08	0.26	232%
FRP	0.06	0.14	107%
TKN	0.12	0.33	177%
NO_x	0.31	0.49	57%
EC	0.25	0.32	29%

Note. Model performance is summarized by Nash-Sutcliffe coefficients (NSEs) in fitting the temporal variability in water quality (see Figure 2 for those for the full models). TSS = total suspended solid; TP = total phosphorus; FRP = filterable reactive phosphorus; TKN = total Kjeldahl nitrogen; EC = electrical conductivity.

NDVI. The negative regression coefficients for the effect of NDVI on TKN and NO_x (Figures 5d and 5e) suggest that greater vegetation cover results in lower concentration of nitrogen species. This overall negative relationship could be due to uptake of nitrogen by plants during the growth season (Arheimer & Lidén, 2000). However, the fact that vegetation cover is not an important predictor for phosphorus suggests that nitrogen species may be more influenced by plant uptake than phosphorus species. It was expected that NDVI would be important in influencing the extent of erosion in the catchment and would therefore influence the trends in the particulate compounds such as TSS and TP. However, NDVI does not show as a key predictor for either TSS or TP (Figures 2a and 2b), which may indicate that vegetation is not as important as other factors in influencing the mobilization and transport of particulates from the catchment into receiving waters. However, the capacity of NDVI to accurately reflect temporal changes in vegetation cover might also be limited by factors like sensor resolutions and data noise (Pettorelli et al., 2005).

Temperature. Water temperature was an important predictor for all constituents, which has positive effects at most sites on TSS, TP, FRP, and TKN (Figures 5a–5d). This positive relationship could result from the cross correlation between air and water temperature across both space and time (Figure S3), and warmer periods are likely associated with greater concentrations of sediments and nutrients due to the enhancement of the source and mobilization processes: (1) soil desiccation and greater soil erodibility (Wood, 1977), (2) agricultural activities that can occur during warmer periods such as tillage, or (3) lower plant canopy cover in drier and warmer months.

Warmer periods are also correlated with periods with lower rainfall and streamflow. So another possible explanation of the positive temperature effects is that the lower surface flow in warmer periods reduces dilution of sediments and nutrients in streams (Houser & Richardson, 2010; Miller et al., 2014; Mulholland, 2004). However, this is less likely as our previous results suggest a relatively minor role of these dilution effects compared with flushing and erosion caused by surface flow (section *Streamflow*).

On the other hand, water temperature had negative effects on NO_x and EC at most sites (Figures 5e and 5f). This negative relationship could be because NO_x and EC transport to streams can be dominated by subsurface flows. There may be a buildup of groundwater with high salinity and high NO_x concentrations in summer months and flushing in winter months (e.g., Costelloe et al., 2009). It is also possible that when there are higher water temperatures, denitrification processes (both in soil and water) are enhanced due to increased reaction kinetics under higher temperatures (Whitehead et al., 2009).

Summary and management implications. It is important to note that the key predictors for temporal variability in water quality (Table 3 and section 3.2) are identified based on only statistical results (BIC). The BIC aims to maximize model performance while minimizing the number of model parameters. An unselected predictor does not necessarily mean that the predictor represents a process that is not important. It could be due to (i) cross correlations between the temporal predictors or (ii) the particular predictor being a poor surrogate for a process. As an example, air temperature was not recognized as an important predictor, possibly due to its high correlation with water temperature (Figure S6 in the supporting information), which has more direct influences on nutrient processing in streams (Roberts & Mulholland, 2007). Similarly, rainfall was not identified as a key predictor of temporal variability in water quality. This could be due to the strong cross correlation between rainfall and streamflow, or could be indicating that runoff is a stronger surrogate of the constituent mobilization and transport process.

Nevertheless, identifying the key predictors for temporal variability in riverine constituent concentrations provides valuable information for catchment management. For example, these understandings can inform whether specific constituent concentrations are expected to increase or decrease over time, based on seasonality of the driving hydrologic, climatic, and vegetation conditions; these can also assist in identifying critical seasons and/or longer periods for which management actions should be taken. In some situations, it may not be practical to obtain data for all the key temporal predictors that we considered in our models (e.g., due to additional cost and labor efforts). In these cases, a potential alternative is to use only the data for the most important driving factor for temporal variability, that is, streamflow, with a sacrifice of predictive power compared with models that use all the key temporal predictors (Table 4 and section 3.2).

4.2. Future Development Toward a Predictive Fully Integrated Spatiotemporal Model

For many of the constituents, only a small proportion of the temporal variability in water quality is explained by these hierarchical models. However, once combined with observed spatial variability (i.e., site mean concentrations), these temporal variability models can assist in explaining the majority of spatiotemporal variability (Figure 4). This improvement in model performance suggests that the hierarchical models used for exploring temporal variability in this study could be adapted into a fully integrated predictive model for spatiotemporal variability in riverine constituent concentrations. A previous study which focused on the same region and the same set of water quality constituents as this study has identified the key controls for spatial variability (Lintern et al., 2018). Different catchment characteristics, including climate, hydrology, geology, topography, and land use, were identified as important controls for spatial water quality variability. These key spatial controls can be combined with the key temporal controls identified in this study, to inform the design of an integrated spatiotemporal water quality model, which allows prediction across both time and space.

A key remaining task required for developing the spatiotemporal predictive model is the ability to predict the site-specific effects of key temporal predictors. Our results clearly illustrate a wide range in relationships between water quality temporal variability and its key predictors across catchments. For example, Figure 5 shows that, while the majority of catchments exhibit positive effects of streamflow on the concentrations of all six constituents (i.e., increase in concentrations with flow), there were negative effects for 0.98% (TSS) to 19.6% (NO_x) of catchments. In some cases, this could be due to outliers, which might explain why where only 1% of catchments show negative streamflow effects for TSS. However, there is still large variability in the temporal effects across catchments (e.g., effects of antecedent streamflow and soil moisture). Further investigations are required to identify the key driving factors of the spatial variability of these temporal relationships across catchments.

4.3. Recommendations for Future Monitoring of Water Quality Predictors and Responses

This research illustrates the value of using collected data to understand the key factors and possible processes that can affect catchment behaviors over time. In the current model, temporal variation in water quality is represented by monthly monitoring data of sediments, nutrients, and salts. The representation and understanding of these temporal variabilities can be potentially strengthened by utilizing data with higher temporal resolution. Alternatively, high-frequency sampling of proxy data could also be utilized to enhance the learning from existing monthly samples. For examples, continuous monitoring data of EC and turbidity can be good proxies for salts and sediments and are readily available from state agencies, such as the Victorian Water Quality Monitoring Network database (Department of Environment, Land, Water and Planning Victoria, 2016) and the NSW Water Information database (NSW Government, 2018).

The modeling approach in this study highlights the potential to extend this data-driven approach to improve understanding of other riverine pollutants, such as toxicants (e.g., heavy metals) and other emerging chemicals (e.g., pesticides) that can have substantial impacts on the aquatic systems and human health (Barceló, 2007; Lippmann, 2000). However, these investigations are currently limited by data availability for toxicants and emerging pollutants. Existing data sets are generally highly localized with diverse focus on different pollutants (Department of Primary Industries, 2016; Environment Protection Authority Victoria, 2013, 2015), which makes it difficult to obtain an understanding across larger scales. The scarcity of data on toxicants and emerging chemicals identifies an important gap in the monitoring of riverine water quality.

As a limitation of this study, the temporal variability model does not consider the temporal changes in land use management (e.g., seasonality in agricultural activity, the amount and timing of fertilizer regimes, and use of crop rotations) as predictors of water quality. This is due to the lack of data available for capturing land use management. However, changes in these land use management practices can lead to increases in the sources of pollutants as well as changes to surface and subsurface runoff generation processes (e.g.,

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DeFries & Eshleman, 2004; Tang et al., 2005). Therefore, further improvements on the temporal model performance is likely warranted by improved capacities in monitoring of temporal patterns in land use management. A good example is the monitoring of adaptation for improved management practices in the agricultural areas within the Great Barrier Reef region, to minimize sediments and nutrients inputs to the marine ecosystems (Queensland Government, 2016; Star et al., 2015).

5. Conclusions

We lack understanding of the drivers of temporal variability in water quality across large spatial scales. Therefore, this study aims to improve the understanding of the key factors and controls that drive the temporal variability in riverine water quality, with long-term data set over 102 catchments in Australia. To achieve this, Bayesian hierarchical models were developed for six water quality constituents including sediments, nutrients, and salts. Using the modeling results, we not only identified the key predictors that could be used to explain temporal variability in riverine water quality but also demonstrated that the influence of these predictors on water quality can be site specific. Catchment managers should consider this site specificity of the relationship between hydroclimatic variables and temporal variability in water quality, and to identify those site-specific critical time periods and spatial variables to inform further detailed investigations and/or investment priorities.

Overall, we find that same-day streamflow has the greatest effect on water quality temporal variability for all constituents. Additional important predictors include soil moisture, antecedent streamflow, vegetation cover, and water temperature. This understanding of the key controls of temporal water quality variability can inform the design of a fully integrated, predictive spatiotemporal model. Such a model could assist catchment managers in designing and identifying key site-specific and regional management strategies, particularly for riverine systems with elevated constituent levels.

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