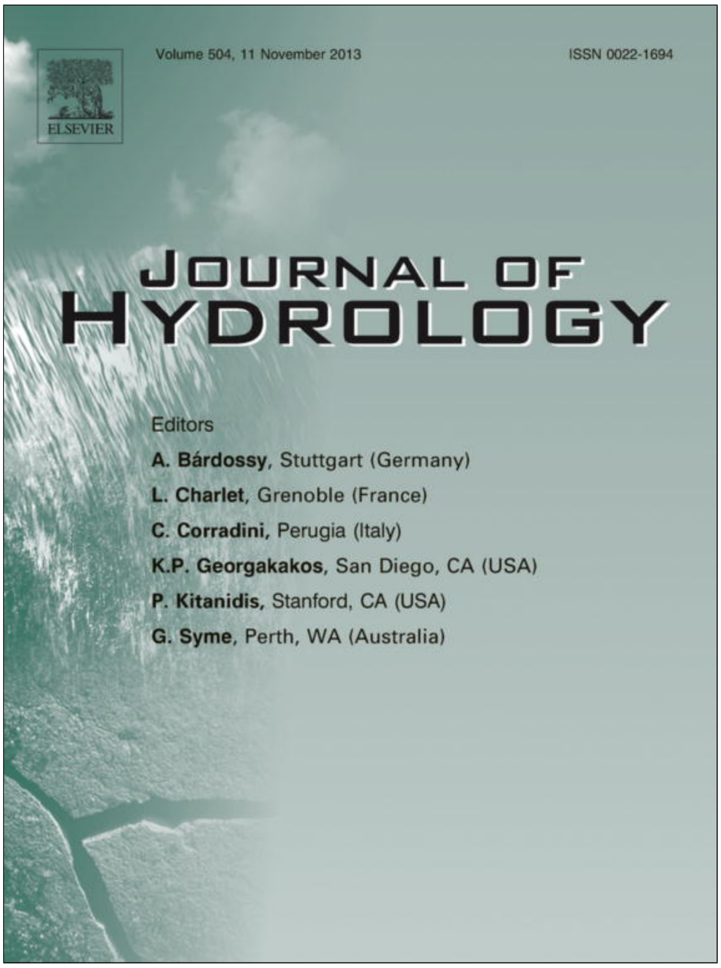
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A predictive geospatial approach for modelling phosphorus concentrations in rivers at the landscape scale

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SUMMARY

Enrichment by phosphorus (P) constitutes a significant pressure on river systems, and is one of the main causes of freshwater pollution globally. Catchment environmental conditions influence the timing and magnitude of P release and transfer to water bodies, and therefore can potentially provide a basis for identifying water bodies vulnerable to impairment by P and/or resistant to restoration efforts. The current research involved construction of a geospatial database, comprising monthly values for flow-weighted concentrations of molybdate reactive phosphorus (fwMRP) sampled in rivers from 2006 to 2008 together with spatially-expressed environmental data relating to 18 different variables for 54 catchments in the Republic of Ireland. A regression–kriging modelling methodology within a landscape-scale, geospatial approach was tested. Environmental conditions relating to hydrological transportation and connectivity (slope, degree of surface saturation, soil water content) were found to exert greater influence over concentrations of P in rivers than direct proxies of sources of P (e.g. human population level or land use). Geospatial models provided greater explanation of P variance than regression models (an improvement in predictive capability of up to 8.5%). Data for fwMRP were segregated sub-annually into two periods, one focused on summer and the other on winter months. A geospatial model for the period including winter months was found to have a better predictive capability than the one that centred upon the summer, with the latter routinely overestimating fwMRP when compared with observed (test) data. Geospatial models potentially provide a means of optimising monitoring regimes for river water quality, and can also be used as a screening tool to focus management and remediation measures where they are likely to prove most effective.

1. Introduction

Eutrophication of waterbodies, as a result of inputs of anthropogenically-derived nutrients, largely phosphorus (P) (Ulén and Kalisky, 2005) but also nitrogen (N) (Sutton et al., 2011), is a widespread and a growing problem (Smith, 2003; Liu et al., 2012). For example, eutrophication accounts for up to 60% of impaired freshwater bodies in the USA (USEPA, 2009), while eutrophic or hyper eutrophic conditions exist in 33% of monitored rivers and lakes in Europe (EEA, 2010). Responding effectively to eutrophication is complicated by the transboundary nature and separation, in terms of location, scale and timing, of causes and effects (Sharpley et al., 2009; Doody et al., 2012). The Water Framework Directive (European Parliament, 2000) (WFD) seeks to address these issues in European (EU) member states, with the latter aiming to achieve and maintain good water quality for all waterbodies by 2015 (Anon, 2005). The WFD incorporates measures listed under previous water quality directives, such as the Nitrates Directive (European Parliament, 1991b), Urban Waste-water Treatment Directive (European Parliament, 1991a) and Habitats Directive (European Parliament, 1997). Measures previously implemented to meet the requirements of these directives form part of the WFD Programme of Measures (POMs); these are the management actions that will be implemented by EU member states to achieve improvements in water quality.

Natural sources of P in waterbodies include atmospheric deposition, natural weathering of soil and detritus from riparian vegetation. Sources of anthropogenically-derived P are often categorised into point or diffuse (Pieterse et al., 2003; Jarvie et al., 2006; Liu and Chen, 2008). Agricultural activities have been shown to be largely responsible for diffuse-sourced P (Hahn et al., 2012; Ulén et al., 2007). However, P from small point sources in rural areas, such as minor wastewater treatment plants (WWTPs) and septic tanks, which was previously often attributed to diffuse sources

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(Jordan et al., 2007), can also be important, especially during periods when the concentrating effects of low water levels in a catchment are maximised (Greene et al., 2011; Evans, 2012). Water passing over or through agricultural soils mobilises P; inputs of P from diffuse sources are therefore closely related to runoff (Edwards and Withers, 2008), and can vary greatly, both spatially and temporally (Stutter et al., 2008; Withers and Jarvie, 2008; White and Hammond, 2009; Rothwell et al., 2010). Moreover, if fertiliser or livestock waste applications coincide with extreme rainfall events, especially on impermeable soils, large amounts of P can be mobilised (Preedy et al., 2001). By comparison, levels of P from point sources, such as WWTPs, are relatively constant throughout the hydrological year, are independent of flow, and are largely in soluble form (Jordan et al., 2007).

The risk of P transfer to freshwaters increases when high loads of P from point or diffuse sources are coupled with high P transport factors determined by environmental conditions (e.g. soil type) in the catchment (Greene et al., 2011). Understanding the inherent relationships between P in waterbodies, anthropogenic sources of P and environmental conditions in the corresponding catchment can help delineate these critical source areas (CSAs) responsible for the majority of P loadings (Doody et al., 2012) and thus identify waterbodies that are likely to prove insensitive to attempts at restoration. While many site-specific studies, generated at relatively small, field-based experimental scales, have provided valuable data informing process understanding (e.g. Brennan et al., 2012; Hahn et al., 2012), they are not immediately suited to management at the whole catchment or landscape scale. For policy makers seeking to develop management and mitigation programmes at catchment and broad landscape scales there are clear advantages to deploying a geospatial, holistic modelling approach that takes into account the spatial variation of P and the interrelationships of multiple stressors over many catchment types.

Increasing interest in assessing the spatial distribution of nutrients in freshwaters has been facilitated by recent advances in geospatial modelling techniques. For example, in the United States (US) Yuan (2004) and Peterson and Urquhart (2006) found the prediction accuracy of respectively, nitrate (NO_3^-) and dissolved organic carbon (DOC) concentrations in rivers increased when spatial autocorrelation was considered. Yang and Jin (2010) compared the effectiveness of ordinary least square regression and spatial regression in evaluating the impacts of catchment characteristics, such as land use and hydrological properties, on oxidised N in streams in the US; predictive capability was improved through the incorporation of geospatial techniques and information. In the current study, the spatial distribution of concentrations of P in rivers is explored and defined according to potential catchment sources and within the context of geographically varying environmental conditions. This spatiality, once defined, is then used to construct sub-annually resolved predictive models of P concentrations in rivers. These models can potentially be used in optimising the current monitoring regime for river water quality, and to focus remediation efforts on areas that are likely to be most responsive.

2. Methods

2.1. Study area

Georeferenced information on molybdate reactive phosphorus (MRP) concentrations in river water and a range of environmental conditions in associated catchments was compiled for 54 river monitoring sites in the Republic of Ireland (RoI) (Fig. 1, Table S1). The most recent and comprehensive data from river monitoring sites were selected on the basis that they were representative of sources of P, environmental conditions, and Strahler stream order in the RoI. The RoI, bordering the eastern North Atlantic, comprises a land area of 68,889 km^2 and has a temperate climate (Love and O'Brien, 2003; Favis-Mortlock, 2006) characterised by high levels of rainfall, averaging 750–1250 mm per annum but exceeding 2500 mm in mountainous parts of the west of the country, and low rates of evapotranspiration (20–50% of rainfall). Typically, winter and summer display the most distinct seasonality extremes, with the highest levels of rainfall and lowest air temperatures reported during the winter months. Synoptic-scale changes in conditons from winter to spring or from summer to autumn tend to occur gradually.

Poorly drained peats and gleys, including peaty podzols, are predominant in northern and western parts of the RoI, whereas the south and east typically comprise free-draining brown earths and brown or grey-brown podzols (Brogan et al., 2002). Over 64% of land cover is improved agricultural land, of which approximately 80% is devoted to grasslands (silage, hay and pasture), the remaining being allocated to rough grazing and crop production (CSO, 2000).

2.2. Data sources and preparation

The sources of data that underpin this research and their pretreatment prior to analysis, including target and predictor variable generation, are described in the Supplemental Information. Here we focus on describing analysis and interpretation.

2.3. Data analysis

2.3.1. Multiple regression

All variables were analysed for normality to meet statistical requirements using histograms, normal Q–Q plots and the Sharpiro–Wilk test for normality. All identified non-normal data were $\log(x + 1)$ transformed. A paired t-test was applied to flow weighted molybdate reactive phosphorus (fwMRP) data to test the hypothesis that the means of the two sub-annually resolved datasets (one centred upon the summer months, but including late spring and early autumn (May–October) – hereafter referred to as 'summer', the other upon the winter months, but including late autumn and early spring (November–April) – hereafter referred to as 'winter') were equal ($p < 0.05$). Simple linear regression analysis was applied to explore fwMRP, as a function of the individual variables that directly or indirectly describe environmental conditions that may influence river P concentrations (Table 1) in 49 of the 54 study catchments. Data for the remaining five catchments selected randomly were withheld for use in a subsequent model testing exercise. A correlation matrix was developed to identify relationships between variables. Initial multiple linear regression models were fit to describe fwMRP as a function of a combination of significant predictors. The resulting residual plots and variance inflation factors (VIFs) of each regression model were examined to ensure that multiple regression assumptions were met and to identify multicollinearity.

Principal component analysis (PCA) (Hotelling, 1933) is a means of reducing the complexity of data to enable identification of the main causes, or principal components, of variation (Field, 2009) and was applied here to the intercorrelated variables to yield standardised, independent predictors of fwMRP. Three principal components were estimated (principal component axes 1–3). The database was then imported into R v 2.12 (Venables and Smith, 2010). Stepwise multiple linear regression of fwMRP, as a function of a combination of the principal components, was carried out. Optimum combinations of the predictors were selected and used to generate two seasonal PCA-based regression models.

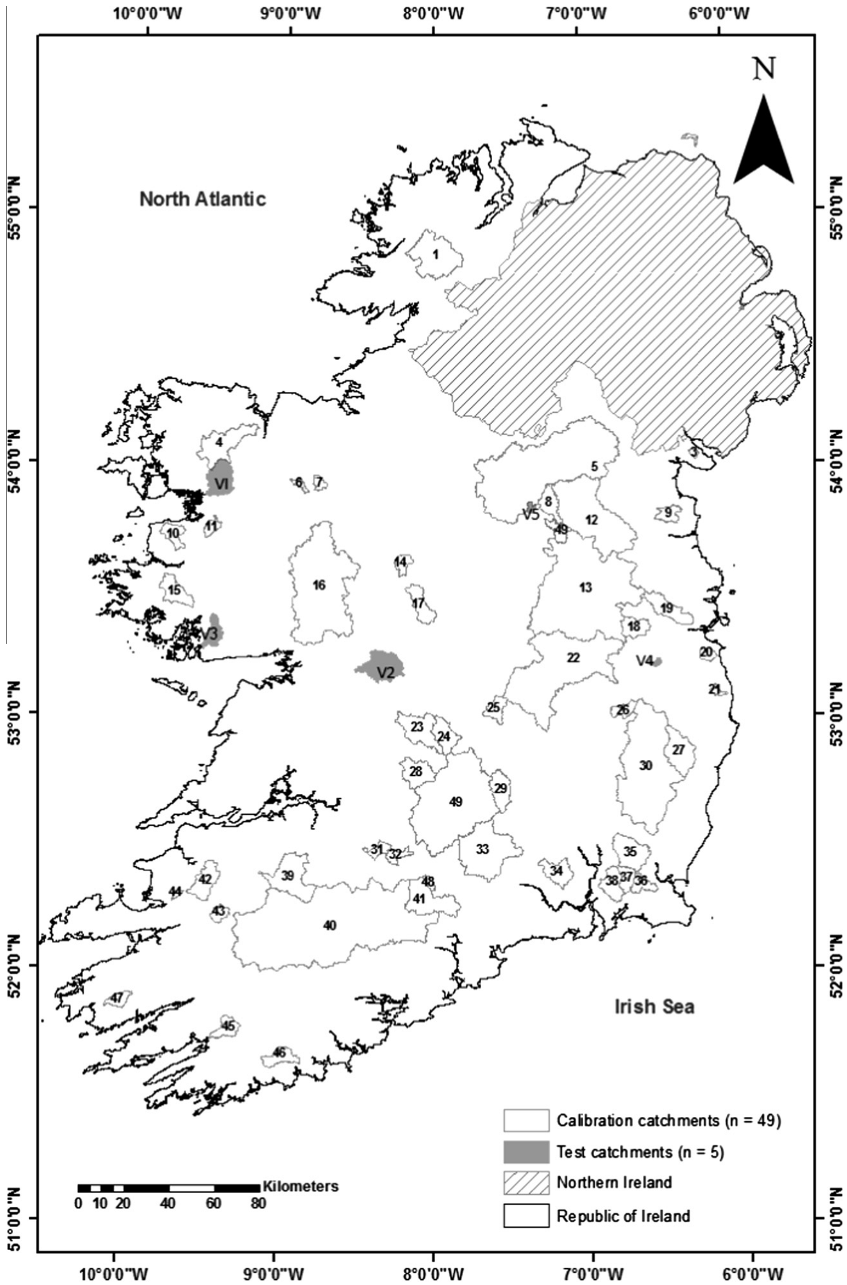

Fig. 1. Locations of the 54 river water quality monitoring sites and corresponding catchment boundaries used in the study. Numbers for catchments correspond to those listed in Table S1 (1–49 indicates sites used for analysis and V1–5 signifies sites used for model testing purposes). Catchment boundaries were delineated in ArcGIS 9.3 using a DEM (20 m) and geographical coordinates (easting and northing) of river monitoring sites provided by the Environmental Protection Agency in Ireland. Information on environmental variables associated with the river water quality monitoring sites is provided in Table S2.

2.3.2. Geospatial analysis

Conventional statistical methods for modelling P export from catchment attributes to water, such as regression, are based on the assumption that spatial correlation between points is random. A geostatistical approach takes this further by accounting for the random and structured nature of spatial variables that exists in many environmental datasets and assuming that sites situated close to one another in space share more similarities than those placed farther apart (Tobler, 1970; Webster and Oliver, 2007).

Geospatial analyses typically comprise two stages; variogram modelling to develop a spatial structure, followed by prediction of the target variable based on spatial interpolation by kriging. Kriging can be used to provide reliable estimates of unobserved variables where environmental conditions are relatively homogeneous. Where conditions vary over relatively short distances, however, predictions to reasonable levels of confidence can be difficult. Variations of local topography, land use, nutrient sources and transport mechanisms may not display an identifiable spatial structure. In such cases, the strength of the kriged estimates decreases, since catchment specific measurements are impacted by strong local variation. Adaptations of the kriging technique have been developed to surmount this weakness, with the adapted technique referred to as regression-kriging (RK) (Hengl et al., 2007). Instead of kriging the target variable directly, the residuals left over after initial multiple regression analysis are examined for spatial autocorrelation and a combination of the regression model and the variogram model of residuals are kriged and interpolated to predict the target variable at unobserved locations.

Table 1
The catchment-specific variables associated with the 54 study river monitoring sites.

Variable name	Variable description
DrainageDensity	Length of river network per unit area of catchment (km)
MeanSlope	Mean slope of catchment (°)
TWI	Mean topographic wetness index of catchment
Bed[1-3]	Bedrock extremely vulnerable (1), moderately vulnerable (2) or resistant to weathering (3) (%)
Artificial	Artificial surfaces in catchment (%)
Pasture	Pasture land in catchment (%)
DesorpRisk	P desorption risk of soils
RiverLength	Length of river network (km)
CatchmentLakeRatio	Ratio of catchment area to lake area
CatchmentArea	Catchment area (km^2)
Urban	Urban area in catchment (%)
Forest	Forestry area in catchment (%)
Arable	Arable area in catchment (%)
RiverOrder	Strahler stream order of river containing monitoring point
Farmyards	Farmyards per unit area of catchment (farmyard km^{-2})
RunoffRisk	P runoff risk index of gley soils in catchment
Cattle	Cattle density in catchment (cattle km^{-2})
Humans	Human density in catchment (person km^{-2})

The RK methodology was used in this study. In order to explain the degradation of spatial correlation between two georeferenced monitoring sites when the separation distance is increased (Fig. 2), variograms were modelled using the gstat package of R (Hengl et al., 2007). First, experimental variogram models were fit to fwMRP with initial variogram parameters comprising nugget = 0, sill parameter = the overall variance in the dataset, and the range = one-quarter of the diagonal of the bounding box (maximum distance of spatial autocorrelation). Variogram functions relate the semivariance (half the expected squared difference between paired data values $z(x_i)$ and $z(x_i + h)$) to the lag distance h, by which sample sites are separated (Webster and Oliver, 2007):

$$\gamma(h) = 1/2E[z(x_i) - z(x_i + h)]^2 \tag{1}$$

For discrete sampling sites, such as those used to collect water quality samples, the function for the variogram was estimated as:

$$\gamma(h) = 1/2m(h)\sum_{i=1}^{m(h)}[z(x_i) - z(x_i + h)]^2 \tag{2}$$

where $\gamma(h)$ is the experimental semivariance estimation at a lag distance h; $z(x_i)$ is the value of the measured samples at sample location x_i; $m(h)$ is the total number of sample sites within the distance h.

Second, a variogram model was then fitted to the residuals of the two PCA-based regression models. Diagnostics and the curve followed by the data in the experimental variograms indicated that a theoretical spherical model would best fit the residual variogram structures; a progressive decrease of spatial autocorrelation (or an increase of semivariance) until a specified sill value was reached, beyond which autocorrelation was equal to zero. The fitted variogram models provided information about the spatial structure of the data for spatial interpolation by kriging. Kriging involves the application of least-square linear regression algorithms to predict the value of a variable at unobserved locations based on the spatial structure (variograms model) of a dataset. Predictions are linear sums of weighted observations within a given neighbourhood:

$$\check{z}(x_0) = \sum_{i=1}^{n}\lambda_i z(x_i) \tag{3}$$

where $\check{z}(x_0)$ is the predicted value of z at location x_0, λ_i is the weight associated with the ith observation and meet the condition needed for unbiased estimates with minimum estimation variances and $z(x_i)$ is the value of the measured samples at sample location x_i.

For RK:

$$z(x) = z^*(x) + \varepsilon(x) = \sum_{i=0}^{n}\beta k q_k(x) + \varepsilon(x) \tag{4}$$

where $z(x)$ is the target variable; $x = (x,y)$ represents the two-dimensional spatial coordinates, $z^*(x)$ is the spatial field that can be estimated for each site x where $qk(x)$ is known, βk are fitted regression coefficients, q_k are the external predictor variables and $\varepsilon(x)$ is a normally distributed residual with zero-mean.

The krige function was applied with an estimated variance (95% confidence interval). Both models generated were tested for accuracy using leave-one-out cross-validation and prediction of fwMRP values for the five river monitoring (test) sites that had been excluded from the dataset used in model construction.

3. Results

Flow weighted MRP concentrations in the rivers ranged from 3 μg l^{-1} in winter months to 135 μg l^{-1} in summer months. Owing to a non-normal distribution of data, a $\log(x + 1)$ transformation was applied to fwMRP data and all potential predictor variables (except cattle density and pasture). The paired t-tests on fwMRP data showed that the means of the two sub-annually resolved datasets were statistically different ($p < 0.05$).

3.1. Regression analysis

Results of the linear regression analyses indicated that 11 of the 18 environmental variables tested were significant predictors of seasonal fwMRP (Table 2). Urban and other artificial land uses, the extent of pasture, cattle density, runoff risk, human density and TWI were positively related to fwMRP. The extent of bedrock resistant to weathering, mean slope, drainage density and forest cover were inversely related to fwMRP. Initial multiple linear regressions of fwMRP as a function of a combination of all significant predictors yielded large VIFs, ranging from 1.5 to 20.8, indicating the presence of multicollinearity between predictor variables and thus a need for PCA.

3.2. Principal component analysis

The first three principal components accounted for 74% of the total variation in the predictor data (Fig. S1, Fig. 3, Table S3). The main component, principal component 1, explained 37.5% of total variation and was mainly influenced by variation in drainage density, forest cover, mean slope and the extent of weathering-resistant bedrock (drivers with negative influences on fwMRP). Variations in urban and other artificial land uses and in human population density (drivers with positive influences on fwMRP) largely explained principal component 2, which accounted for 25% of total variation. Principal component 3 mainly accounted for variations in cattle density, extent of pasture and the degree of P runoff risk and TWI (drivers with positive influences on fwMRP). The optimal combination of environmental predictors (in the form of the three principal components) of fwMRP was selected by stepwise multiple linear regression. No multicollinearity between predictor variables in the two regression models produced was evident (VIF = 1.0), and the residuals did not significantly depart from a normal distribution. The adjusted R^2 values ($p < 0.05$) of the regression model for the summer and winter data were 0.68 and 0.66, respectively.

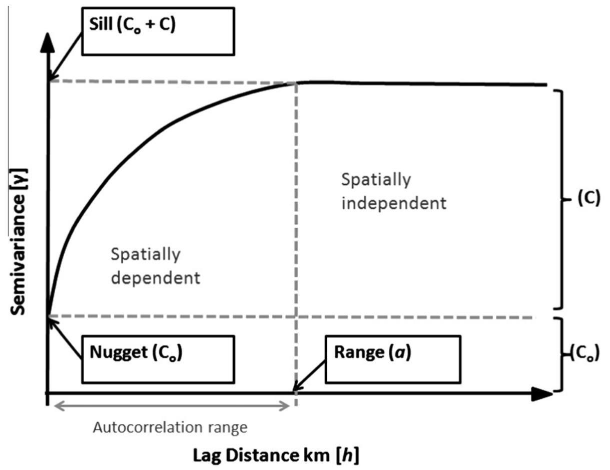

Fig. 2. Schematic of the characteristics of a typical variogram model. Semivariance (γ) typically increases with increased lag distance (h), until it eventually reaches a maximum (the sill variance, ($C_o + C$)) at which point the graph flattens out. Such a variogram is termed bounded. The distance at which the variogram reached its sill is called the autocorrelation range (a), which is the limit of spatial dependence over which two sites are correlated. Sites separated by greater distances than the range are spatially independent. The nugget (C_o), the intercept of the variogram, occurs when a spatial process is discontinuous as the distance h approaches zero lag. If the semivariance increases with distance but does not reach a maximum value at the largest distance measured, the variogram is termed unbounded. In the event of no spatial dependence in the data (e.g. owing to measurement errors and variation that occurs over distances less than the shortest sampling interval), the variogram is referred to as pure nugget.

3.3. Geospatial analysis

The experimental variograms detected spatial dependence in the sub-annually resolved fwMRP data and the residuals of the PCA-based multiple regression models. All variogram models were tested for isotropic, Euclidian distance correlation, and were fitted with a spherical model (Fig. 4). Overall, values for the nugget, sill and range parameters of the residuals were generally smaller than the values estimated by direct variogram analysis on the dependent variable (MRPLX1 in Fig. 4). Models showed a large decline in the sill value from the dependent to residual model parameters. The distances at which fwMRP values were no longer spatially correlated were 93 km and 89 km for summer and winter data, respectively. However, the residuals of the regression models had spatial dependence over an area of up to 102 km diameter.

Generation of geospatial models (by the application of RK on a combination of the PCA-based multiple regression and variogram models) yielded R^2 values of 0.70 (summer) and 0.71 (winter). These R^2 values were higher than those for the corresponding multiple regression models. Based on the difference between these R^2 values, the winter-based geospatial model explained 8.5% more variance in the P data than the non-spatial regression model, while the predictive accuracy of the summer model was improved by 3%. Leave-one-out cross-validation analysis produced individual predicted fwMRP values for each river monitoring site. The R^2 values of the linear relationship between predicted and observed were 0.72 and 0.70 for winter and summer, respectively. The results of model testing using data from river monitoring sites and catchments kept separate from the datasets used in model construction show that the geospatial model based on winter data (RMSE = 7 μg l^{-1}) was more robust than the one based on summer data (RMSE = 18 μg l^{-1}). Standardising the RMSE (RMSE divided by the standard deviation of the observed values) provides a useful basis for comparing the levels of skill of different models: a standardised RMSE of 0.4 or less indicates good prediction accuracy (Hengl et al., 2004). The winter model has a standardised RMSE of 0.3. The lower accuracy of the summer model is also evident in its standardised RMSE, which at 1.9 is higher than that of the winter model and higher than the 0.4 threshold. The summer model was found to over-predict P concentrations when compared with actual levels at the river monitoring sites used in model testing.

4. Discussion

4.1. Multiple stressors of P in rivers

Urban areas can exert adverse pressures on water quality through acting as sources of large quantities of pollutants, including nutrients, the alteration of hydrological transport pathways, and reduced water residence times (Barron et al., 2013; Carey et al., 2013). The impact of urbanisation on concentrations of P is evident here in the significant positive relationships observed between fwMRP and human population density in particular, and also the extent of urban and other artificial land uses. The relationship between fwMRP and human population density was slightly stronger in winter than in summer, suggesting peak anthropogenic influence during what is often hydrologically the most variable part of the year. However, in-stream retention of P during summer could have caused the decreased explanation of variance (de Klein and Koelmans, 2011; Jarvie et al., 2011). The proportion of variance of fwMRP explained by the extents of urban and other artificial land uses was greater in summer than winter, indicating that the risk of nutrient enrichment from these land covers is increased during the ecologically sensitive summer period, presumably because of the concentrating effects of low-flows.

The influence of geology on P concentrations in rivers is widely known (Sliva and Dudley Williams, 2001; Rothwell et al., 2010; Nasr and Bruen, 2013) and has been shown here to have a small, but notable influence. The extent of bedrock resistant to weathering in the study catchments was inversely related to fwMRP (summer R^2 = 0.28, p = 0.05 and winter R^2 = 0.21, p = 0.05). The type of bedrock has a direct impact on environmental variables, such as hydrology, soils and relief, which are expected to influence P transfer (Skinner et al., 2004; Mellander et al., 2012). Thus, poorly drained (gley) soils – characterised by waterlogging, extensive across the island of Ireland (Diamond and Sills, 2001), comprise a major contributing factor to runoff risk. Runoff risk was positively related to fwMRP in the current research, with little seasonal variation in R^2 values evident. Geology also plays an indirect role in P concentrations in surface waters through, for example, influencing human settlement and agriculture (Legg and Taylor, 2006; Jaroslaw and Hildebrandt-Radke, 2009).

Mean catchment slope was inversely related to fwMRP, as has previously been reported for Ireland (Donohue et al., 2005; Taylor

Table 2
Coefficient of determination (R^2) values for linear regression of significant predictors of fwMRP.

Variable	Winter months		Summer months	
	R^2	Rank	R^2	Rank
MeanSlope	0.401 (–)	1	0.383 (–)	1
TWI	0.384 (+)	2	0.315 (+)	2
Human	0.328 (+)	3	0.269 (+)	5
RunoffRisk	0.248 (+)	4	0.252 (+)	6
Pasture	0.234 (+)	5	0.270 (+)	4
Forestry	0.232 (–)	6	0.222 (–)	8
Cattle	0.225 (+)	7	0.174 (+)	10
Bedrock[3]	0.213 (–)	8	0.279 (–)	3
Urban	0.154 (+)	9	0.251 (+)	7
Artifical	0.133 (+)	10	0.208 (+)	9
DrainageDensity	0.082 (–)	11	0.143 (–)	11

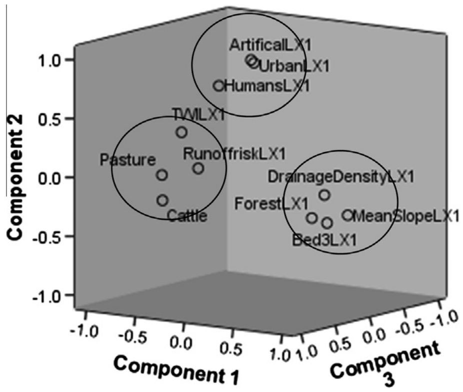

Fig. 3. Principal component analysis plot in rotated space (varimax rotation) showing significant predictors (environmental conditions) of concentrations of fwMRP. Three principal components axes were extracted (shown in the circles). The main component, principal component 1, explained 37.5% of total variation and was mainly influenced by variation in drainage density, forest cover, mean slope and weather resistant bedrock (drivers with negative influences on fwMRP). Differences in urban and other artificial land covers and in human population density (drivers with positive influences on fwMRP) largely explained principal component 2, which accounted for 25% of total variation. Variations in cattle density, extent of pasture, P runoff risk and TWI (drivers with positive influences on fwMRP) were mainly accommodated by principal component 3.

et al., 2012). A combination of higher precipitation, thinner soils, less vegetation, resistant bedrock, groundwater contribution and a decreased influence of anthropogenic and agricultural pressures in upland, high relief areas of the study catchments may explain the low P concentration (Soulsby et al., 1998, 2002; Bowes et al., 2003; Thompson et al., 2012). Lower catchment slope also indicates longer groundwater residence times, thereby facilitating larger areas of saturated soil leading to increased dissolved nutrient concentrations in the water (D'Arcy and Carignan, 1997). An inverse relationship between P concentration and a combination of high drainage density, high slope and weather resistant geology has also been reported for rivers in England and Wales (Evans, 2002). As subsequent research by Johnes et al. (2007) has demonstrated, this inverse relationship may be because the prevalence of low intensity, relatively extensive forms of land use in many upland areas in western Europe means that a high P export potential remains largely unrealised.

Mean slope was the strongest predictor of fwMRP, while TWI (a proxy for overland flow owing to saturation-excess) displayed the second strongest positive relationship to fwMRP in the study database. Catchments can contain relatively small but well-defined areas of saturation (typically lowlands) that can be responsible for a large proportion of overland flow; where such areas overlap with high levels of soil P, substantial exports of P to freshwaters can occur (Reaney et al., 2011; Wall et al., 2011; Dahlke et al., 2012). High loadings of P are a function of high rainfall intensity, coupled with low catchment slope, impermeable soil type, high soil water content and high degree of surface saturation. In the current research, these parameters were encapsulated in both hydrologically related environmental conditions (mean slope and TWI) found to be most influential in predicting P concentrations in river water.

Of the agricultural variables investigated, the percentage of pasture and cattle density in a catchment related directly to fwMRP. Positive relationships between pasture grasslands and P in waterbodies have been identified in other studies (Wood et al., 2005; Withers et al., 2007; Miller et al., 2010; Cournane et al., 2011). Grass-based agricultural production systems in Ireland are known to contribute a significant proportion of P to freshwaters (Kurz et al., 2005). High inputs of fertilisers and subsequent leaching of P from fertilised soils to freshwaters are the most likely explanation for the direct relationship between P concentrations at the monitoring sites and extent of pasture in the associated catchments (Withers et al., 2007; Nash and Hannah, 2011; Jordan et al., 2012; Buckley and Carney, 2013). Concentrations of P in river water may also reflect the legacy effect of past management practices (Schulte et al., 2010), notably past applications of fertilisers. Furthermore, cattle are associated with both point and diffuse sources of P (McGechan et al., 2005), either through direct addition of their faeces and urine or indirectly as a result of their destruction of riverbanks and re-suspension of river sediments (Jarvie et al., 2010; Miller et al., 2010; Smith et al., 2013). Arable land use in the catchments was not identified as a significant influence on P concentrations, despite a strong correlation found in a study of river catchments in England and Wales (Evans, 2002). This difference may reflect the dominance of grassland agriculture in Ireland or a low rate of P export owing to the landscape type typically used for arable production (Johnes et al., 2007).

The extent of forestry is inversely related to fwMRP in the study catchments. Previous studies have shown that the presence of tree cover, buffer filter zones and associated natural processes leads to a decline in P export to freshwaters (Evans, 2002; Ballester et al., 2003; Anbumozhi et al., 2005; Mouri et al., 2011). Forests utilise nutrients in a largely closed cycle (Abelho, 2001), resulting in relatively low losses to the catchment (McElarney et al., 2010). Moreover, forested areas typically have wider and more continuous riparian buffer zones (Booth, 1991) that protect aquatic systems by controlling runoff (Rodgers et al., 2010). However, harvesting of trees in forest plantations can increase the delivery of P to freshwaters (Piirainen et al., 2007; Kreutzweiser et al., 2008; Rodgers et al., 2010; Drinan et al., 2013): the fact that this does not appear to be a problem in the study catchments, at least according to the current study, is perhaps due to the largely moribund state of commercial forestry in the RoI at present.

The outputs generated here provide information on the hierarchy of environmental conditions that influence seasonal P loss at the landscape scale across the RoI. In particular, the results stress the importance of environmental conditions relating to hydrological transportation and connectivity (i.e. slope, TWI) as the fundamental control of concentrations of P in rivers. According to the data, these environmental conditions exert a greater influence over concentrations of P in rivers than factors that are expected to be more directly associated with sources of P (e.g. human population or pasture). There is great uncertainty related to identifying high risk sources of P at the catchment scale, particularly owing to variations of soil moisture and rainfall levels (Doody et al., 2012). Isolating these high risk areas is difficult because diffuse sources of P do not originate from all parts of a catchment, or even where P is available in excess. Rather, P is transported to waterbodies when CSAs coincide with hydrologically active areas (variable source areas, VSAs) that are directly linked to a waterbody by hydrological pathways (Frey et al., 2009; Reaney et al., 2011; Jordan et al., 2012; Shore et al., 2013). The VSAs occur as a function of high rainfall intensity, coupled with catchment slope, soil type, soil water content and degree of surface saturation, and vary seasonally and spatially as a result. To date, empirical studies of the factors influencing loadings of P in rivers at the landscape scale have generally neglected the role of hydrological transport, focusing primarily on sources of P. However, the current research suggests that a greater proportion of the variability of P can be explained through using simple measures of hydrological connectivity based on high resolution digital elevation model data. Although the CSA concept works most optimally at the field or hillslope level (e.g. Reaney et al., 2011; Wall et al., 2011), catchment scale assessment provides a useful means of identifying high risk areas for P loss in the absence of detailed field data.

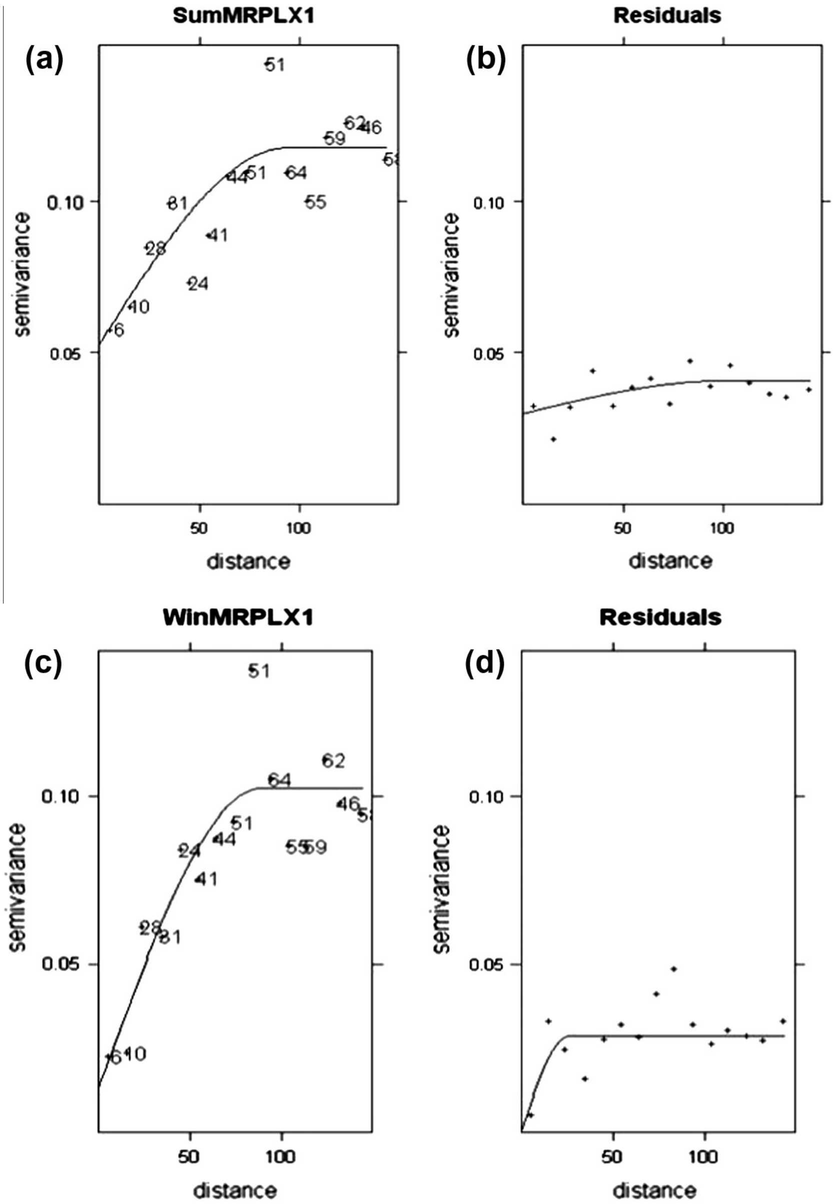

Fig. 4. Variogram models: (a) based on the dependent variable (MRPLX1) for summer; (b) based on the residuals of the regression models (using principal components) for summer; (c) based on the dependent variable (MRPLX1) for winter; (d) based on the residuals of the regression models (using principal components) for winter. The semivariogram values (y-axis) are plotted against the lag distance values (x –axis), i.e., the separation distances of individual pairs of measurements (h). The numbers inside the variograms represent the numbers of pairs of observations for each h (lag distance) value.

4.2. Practical applicability

The geospatial models improved predictive capability by up to 8.5% when compared with equivalent non-spatial regression models. However, the improvement in proficiency was most noticeable for the winter model; the process of model testing, based on data from a set of five catchments not used previously, also revealed that the winter model had a greater predictive ability than the summer model, with the latter tending to over-estimate MRP concentrations. Increased biological activity and the partial loss of connectivity within the drainage network at low flows causing greater in-stream retention of P (Jordan et al., 2005; Rothwell et al., 2010; Jarvie et al., 2011) are likely explanations for the behaviour of the geospatial model calibrated with data from the low-flow summer months. Enhancement of the prediction accuracy and utility of the geospatial models produced here should follow with the emergence of higher quality data, together with accommodation of the entire Irish Ecoregion (i.e. including river catchments in Northern Ireland, UK).

Models are employed in environmental policy and management because they provide practically useful outputs (van Daalen et al., 2002). Model transparency, simplicity, user friendliness and data availability are crucial factors for their successful use in policy and management. Proven accuracy of predictions is also vital. To increase user-friendliness, the geospatial models constructed here can be made available as a computer software package in the R computing environment. Furthermore, the concept of the Environmental Virtual Observatory (Emmett et al., 2011), currently linking environmental data and model structures in a cloud-based resource for the UK, is highly appropriate for the effective use of the models by a wider and more diverse audience.

Specifically, the geospatial models may be used as a screening tool to guide decision making, targeting limited resources to areas where mitigation would be the most effective, i.e. in the catchments identified as most vulnerable to P pollution. Indeed, geospatial models in general are potentially valuable in environmental management as they can permit patterns and relationships developed under data-rich conditions to be extended to regions and systems where there is limited availability of suitable data, such as unmonitored rivers or lakes. For example, monitoring of freshwater quality in the RoI currently targets a sample of sites that are regarded as representative, leaving many rivers and lakes unmonitored (EPA, 2006). However, obligations for WFD compliance include submission of an up-to-date report of the chemical and ecological status of both monitored and unmonitored waterbodies in 2015. Using readily-available data describing environmental conditions in all river catchments in the RoI, geospatial models can be used to identify waterbodies that are vulnerable to external loadings of P, and this information can be used as a guide to both their relative chemical and ecological status and in designing monitoring campaigns that target the most at-risk rivers and catchments, before implementing mitigation measures. Furthermore, scenario analysis, such as the type conducted by Johnes et al. (2007) and Glavan et al. (2012) in which model parameters are altered to explore the impact of changing the sources and management of P in catchments, could further guide decision-making at the landscape scale.

5. Conclusion

Effective management of aquatic pollution from nutrients and its impacts requires a sound understanding of the full range of influencing mechanisms and processes in catchments. Landscape-scale modelling provides a cost-effective, practical way of testing hypotheses, and augmenting current understanding. Here, non-spatial regression analysis indicates that statistically significant relationships exist between biologically available P and indicators of catchment conditions, reflecting the influence of environment on concentrations of P in waterbodies. Moreover, the research described provides new information on the hierarchy of environmental conditions that influence sub-annual variations in losses of P at the landscape scale. In particular, the results highlight the importance of catchment conditions relating to hydrological transportation and connectivity (i.e. slope, TWI) in influencing concentrations of P in rivers, particularly when compared with the effects of proxies of sources of P (e.g. human population level and extent of pasture). Moreover, the results highlight that, even after controlling for the environmental variables mentioned above, unexplained variation in P concentrations can be explained, in part, by a spatial relationship between monitoring sites across the landscape. Geospatial models have great potential in environmental management at the landscape scale, for example in the *a priori* identification of rivers with a high likelihood of vulnerability to impairment by biologically-available P.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jhydrol.2013.09.040.

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