A Monte Carlo approach to the inverse problem of diffuse pollution risk in agricultural catchments

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Abstract

The hydrological and biogeochemical processes that operate in catchments influence the ecological quality of freshwater systems through delivery of fine sediment, nutrients and organic matter. Most models that seek to characterise the delivery of diffuse pollutants from land to water are reductionist. The multitude of processes that are parameterised in such models to ensure generic applicability make them complex and difficult to test on available data. Here, we outline an alternative – data-driven – inverse approach. We apply SCIMAP, a parsimonious risk based model that has an explicit treatment of hydrological connectivity. We take a Bayesian approach to the inverse problem of determining the risk that must be assigned to different land uses in a catchment in order to explain the spatial patterns of measured in-stream nutrient concentrations. We apply the model to identify the key sources of nitrogen (N) and phosphorus (P) diffuse pollution risk in eleven UK catchments covering a range of landscapes. The model results show that: 1) some land use generates a consistently high or low risk of diffuse nutrient pollution; but 2) the risks associated with different land uses vary both between catchments and between P and N delivery; and 3) that the dominant sources of P and N risk in the catchment are often a function of the spatial configuration of land uses. Taken on a case by case basis, this type of inverse approach may be used to help prioritise the focus of interventions to reduce diffuse pollution risk for freshwater ecosystems.

Keywords: Diffuse pollution; Hydrological connectivity; Land cover; Nutrients; Nitrogen; Phosphorus; Risk; Modelling.

1. Introduction

The source-mobilisation-delivery conceptualisation of diffuse pollution transfer from land to water is widely accepted (Heathwaite, 2010) and forms the basis of several existing diffuse pollution models that seek to explain the variation in river water quality over time in terms of the processes and pathways of delivery operating within a catchment. Much effort has been focused on characterising the variation in water quality timeseries (Burt *et al.*, 2011; Howden *et al.*, 2010; Kirchner *et al.*, 2004; Neal *et al.*, 2010a) or producing a good fit between modelled processes and measured water quality (Lazar *et al.*, 2010; Whitehead *et al.*, 2007) but such effort does not elucidate the spatial signals in a catchment. Not all locations in a river catchment (even if they have the same land use) contribute equally to the delivery of sediment or nutrients and hence to in-stream sedimentation and water quality degradation (Heathwaite *et al.*, 2000; Lane, 2008; Page *et al.*, 2005; Pionke *et al.*, 2000). Critical source areas (CSAs) in catchments are characterised by the capacity to entrain material and to connect and hence deliver it to the drainage network (Sharpley *et al.*, 2008; 2009). The environmental degradation associated with diffuse nutrient and sediment losses from land to water may be redefined as comprising a series of spatially-distributed sources of varying size (often fields, or even parts of fields) where particularly risky uses of land combine with a high probability of connection of those risks to the river network (Heathwaite et al., 2000; Lane *et al.*, 2006; Lane 2008). Focusing intervention measures on reducing diffuse pollution delivery from these risky land uses should maximise the return on investment in terms of improvements to ecological quality (Collins and McGonigle, 2008; Heathwaite, 2010; Living With Environmental Change, 2011).

A range of modelling approaches have been developed to meet the challenge of identifying the locations within a catchment that have the greatest probability of contributing high diffuse pollution loads to receiving waters. Lane et al. (2006) classify diffuse pollution models into three main groups: (1) transfer function modelling – which predicts nutrient export on the basis of simple empirical transfer functions driven by known fertiliser and manure inputs coupled with soil nutrient status (e.g., Ekholm *et al.*, 2005; Heathwaite *et al.*, 2003a; Johnes, 1996; Johnes *et al.*, 2007; Jordan *et al.*, 1994; Khadam and Kaluarachchi, 2006); (2) land unit modelling – which applies physically-based (sometimes called mechanistic) models of nutrient cycling to individual land units in order to determine export (e.g. Matthews, 2006; Priess *et al.*, 2001; Vatn *et al.*, 2006); (3) land transfer modelling – which combines the kind of analysis described in (2) with a physically-based, sometimes dynamic, treatment of how material is transferred across the landscape (e.g. Easton *et al.*, 2008; Neumann *et al.*, 2010; Wade *et al.*, 2002). The latter (see Radcliffe *et al.*, 2009) ought to capture effectively the delivery of diffuse pollutants. The main difficulty is whilst they are physically-based they contain parameters or require data whose values either: (i) cannot be determined from available data; or (ii) need to be adjusted, calibrated, so as to force the model to fit known system response (Oreskes *et al.*, 1994). The information demanded in terms of data and model parameters may exceed the information content of calibration data (Heathwaite, 2003;

2010; Kirchner, 2006) and different model realisations (i.e. model runs with different parameter combinations) may have very similar levels of success (i.e. equifinality, Beven, 1993).

Mathematical models are constructed through a process where, in response to a perception of what matters to the system of interest, the processes that need to be modelled are identified (e.g. rainfall, evapotranspiration, infiltration, runoff generation, biogeochemical processing, mobilisation of material into solution and its subsequent transformation in transit, etc). A suitable model to represent these processes will then be identified, modified or developed. This process is implicitly reductionist and points to the development of ever more complex models given the multitude of processes that could be included to guarantee that the model can be applied in many situations. There are two responses to this challenge. The first couples conventional predictive models with differing levels of process complexity at different scales (e.g. Hewett *et al.*, 2009; Quinn, 2004). Each level contains process complexity that is appropriate to the information available to that scale of enquiry. Information is then exchanged between scales as a means of scaling up. The second, which we focus on here, uses a risk-based analysis with a single simplified model to represent all scale ranges. These approaches have proved very effective in diffuse pollution modelling (e.g. Heathwaite *et al.*, 2003a, b; Johnes and Heathwaite, 1997; Jordan *et al.*, 1994; Munafo *et al.*, 2005; Siber *et al.*, 2009; Weld and Sharpley, 2007). Their primary assumption is that the amount of material that is exported from a land unit can be traced to the properties of that land unit (e.g. physical attributes like slope and soil type) and how it is managed (e.g. levels of fertiliser application). Measurements have allowed identification of associated export coefficients, which in many cases have some kind of *a priori* or logical basis (e.g. export coefficients for a pollutant that is eroded whilst bound to fine sediment are greater for land uses where vegetation cover is bare for part of the year). We label this 'forward modelling'.

This paper presents an alternative conceptualisation, in which we consider the problem and use a Bayesian approach to determine the weightings that must be given to different land uses in order to explain spatial patterns of measured in-stream nutrient concentrations. Following Mosegaard and Tarantola (2002), inverse modelling involves using a physical theory (or set of theories) to connect a set of model parameters to a set of observations. In an inverse model, the forward model is inverted so as to predict the model parameters that reproduce those observations. In some cases this inversion is tractable using maximum likelihood methods but not in all cases (Mosegaard and Tarantola, 2002). The inverse problem can also be approached by pseudo-randomly generating a large collection of (forward) models, then analyzing and displaying the models to convey information on the relative likelihoods of model properties (Mosegaard and Tarantola, 1995). This can be accomplished using a Monte Carlo method even in cases where no explicit formula for the a priori distribution is available (Mosegaard and Tarantola, 1995). It is this latter approach that we adopt here, with the objective of making as few *a priori* assumptions as possible about what might be driving river water quality patterns (Lane 2008).

Following observations regarding critical source areas, we retain the assumption that locations will vary spatially in their ability to generate and deliver diffuse pollution risk. It is clear that in trying to understand the relative contribution of diffuse pollutants in catchments, model assumptions have a material impact upon the way a system is modelled (e.g. the assumed contribution of point and diffuse pollution sources will fundamentally impact upon the extent to which a model must focus upon point source delivery of discharges from sewage treatment plants and urban drainage). By taking an inverse approach, we 'train' each model to the local characteristics of each catchment, avoiding the need for a generic model in which many model parameters may end up being superfluous and where training (or calibration) is difficult because there is rarely enough data to distinguish between different model formulations (Beven, 1989). The aim of this paper is to present our approach to the inverse problem for two key nutrients associated with diffuse pollution: phosphorus (P) and nitrogen (N). Both nutrients are particularly important controls on the ecological quality of freshwater systems (Heathwaite, 2010). We use the results of our analysis to show that policy interventions designed to reduce the risk of diffuse pollution need to be sensitive to the relationship between nutrients, relative land use dominance and catchment characteristics, including land use configuration and hydrological properties.

2. Methods

2.1. The SCIMAP model

We use SCIMAP to produce a risk-based estimation of diffuse pollution (see Lane *et al.*, 2006 and Reaney *et al.*, 2011) and a full description is provided in the Supplementary Online Material that accompanies this manuscript (Appendix 1). SCIMAP conceptualises catchments as comprising a collection of flow paths that accumulate spatially distributed sources that may result in the pollution of receiving waters from across the landscape and deliver them into the river corridor. It is within the river corridor that diffuse pollution may become ‘visible’, either through detection of temporal changes in water quality via routine monitoring (e.g. elevated nitrate concentrations) or through the more limited, evidence from physical water quality deterioration (e.g. algal blooms, Hilton et al., 2006; or long-term changes in ecological quality, Reaney et al., 2011). Given an observed change in catchment water quality a primary challenge is to attribute this to its sources, whether point source or diffuse. If the latter, the challenge becomes over which locations are likely to be the significant CSAs. SCIMAP’s approach is relative in that, subject to data availability, the model can be applied at any scale, with the predictions relative to the scale at which the model is used. It allows successive identification (in relative terms) of the catchments that merit prioritisation, followed by the sub-catchments and then eventually the associated fields. A full description and application of the model is provided in Reaney *et al.* (2011) who show how SCIMAP can be used to understand the relationships between land use, hydrological connectivity and the spatial distributions of salmonid populations. In this paper, we use an inverse approach to estimate the generation risk by inferring how land uses need to be weighted to optimise the explanation of spatial patterns of measured water quality parameters. We use an informal Bayesian likelihood estimation procedure conceptually similar to the Generalised Likelihood Uncertainty Estimation approach (Beven and Binley, 1992; Vrugt *et al.*, 2009).. We use water quality data that are available through the Environment Agency for England and Wales (EA) General Quality Assessment (GQA) monitoring network (see: http://bit.ly/EA-GQA). These datasets are described in more detail later in the paper.

2.2. Application

The SCIMAP model framework has five general steps; the focus of the inverse approach reported in this paper is Step (1), which is described in full below. Further detail of Steps (2) to (5) and full justification is given in Reaney *et al.* (2011) and in the Supplementary Online Material (Appendix 1); only a brief summary is given here. Step (1) seeks to identify, in relative terms and for each location in a landscape, the risks of diffuse pollution generation. Step (2) determines the risk of delivery, the delivery index, for each location. This is based upon the assumption that the driest point along a flow path between a location, *i*, and the river is the one that is most likely to control the extent to which material moving over the surface or the shallow subsurface moves vertically as opposed to laterally and, in so doing, becomes hydrologically-disconnected from the surface water system. Lane *et al.* (2009) show that using this measure to determine a delivery index can explain a significant proportion of the tendency towards hydrological connection. We derive the delivery index from 10 m resolution digital elevation models collected using airbourne Interferrometric Synthetic Aperture Radar (see Reaney *et al.* 2011). We emphasise that the analysis makes a specific assumption that topography exerts a primary control on the spatial structure of soil moisture in agricultural catchments. Each location then has a risk of diffuse pollution generation and a risk of delivery. These are scaled to give relative generation and delivery risks for each location in the catchment and multiplied together (Step 3). In Step 4, the resultant location risks are routed through the catchment to the river network, using the same topographic data used in Step 2. Step 4 results in a monotonically increasing level of risk with distance downstream in the river network and so in Step 5 we correct this by dividing the result by the upslope contributing precipitation for each location in the river network, using annual average precipitation data, based on the UKCP09 baseline (Perry and Hollis, 2005).

2.2.1. Generation Risk

In this paper, we use a sampling approach to the inverse problem in Step (1). Step (1) is underpinned by the assumption that some land use and/or land management combinations are more likely to generate diffuse pollution risk than others, and we can use land cover as a first approximation of this risk. There are well-established approaches for determining the risk of diffuse pollution generation from land cover, ranging from the simpler export coefficient models (e.g. Heathwaite *et al.*, 2003a; Johnes, 1996) through to more complex models of sediment entrainment and nutrient cycling (e.g. Vatn *et al.*, 2006). Here, we use an informal Bayesian inference methodology to infer the risk weighting that needs to be given to each land cover in order to optimise a spatially-distributed set of water quality observations. Our analysis is focused on P and N risk as two of the key consequences of agricultural diffuse pollution and drivers of the deterioration of ecological quality in freshwaters. Here, we assume that: different land covers generate different diffuse pollution risks; within land cover risk variability is small relative to that between land covers; and the pattern of land covers is fixed over the observation period. The aim of Step (1) is then to infer the optimum land cover risk weighting in the SCIMAP framework, so as to maximise the level of explanation in a spatially-variable, measured, risk indicator.

2.2.2. Land cover

Our starting point for Step 1, the estimation of the generation risk, is identification of land cover classes from the UK Land Cover Map 2000, a digital map derived from a computer classification of satellite scenes obtained mainly from Landsat satellites and with a resolution of 25 m (Fuller *et al.*, 2002). We use the data in raster format (converted from the original vector database) resampled to the same resolution as the elevation data (10 m) using a nearest neighbour algorithm. These data represent the finest resolution and most precise UK wide land cover dataset that was available at the time of analysis. The Foresight Land Use Futures Report (2010) highlighted the difficulties in obtaining accurate and current land use data for the UK, partly as a result of the way the data is collected but also because synthesising very different data sources to produce a UK land use map remains a challenge. The Land Cover Map records 16 classes and 27 subclasses within the 'Broad Habitats' classification (Jackson, 2000). We grouped these broad habitats and their subcomponents into ten land cover classes that have potential to contribute varying magnitudes of diffuse pollutants to receiving waters. The ten classes chosen with respect to their likely linkage to diffuse pollution sources are: improved grassland, rough grass, moorland, bog, urban, cereals, horticulture, non-rotational horticulture, woodland, and 'other', which was set to represent those land covers (e.g. lakes) with zero risk. Table 1 gives the relationship between the broad habitat classes and the ten SCIMAP classes. Improved grassland is regularly re-seeded and receives significant nutrient inputs usually as slurry and/or fertiliser; the dominance of palatable grasses gives these areas a distinct spectral signature (Fuller *et al.*, 2002). Rough grass land covers are dominated by very low productivity grasses, they are not normally improved by reseeding or fertilizer applications because the land tends to be physically-limiting (e.g. too wet, too steep, too rocky) and can include areas dominated by *Pteridium aquilinum* at the height of the growing season. Moorland cover is characterised by large expanses (> 25%) of ericaceous species and gorse. Bogs are either upland or lowland areas that are permanently, seasonally or periodically waterlogged defined based on both vegetation (ericaceous, herbaceous and mossy swards) and peat depth (> 0.5 m from peat drift maps). Urban land covers range from single buildings to large towns or cities and include: roads, derelict ground, and gardens. Cereals include spring and winter crops; horticulture includes arable bare ground and non-cereal spring crops; and non-rotational horticulture includes orchards and non-grass setaside. Woodland includes both broad-leaved and coniferous woodland.

<Table 1 near here>

2.2.3. Risk indicator

The inverse approach requires time-integrated, spatially-distributed datasets that can provide an indication of water quality. Here, we use the General Quality Assessment (GQA) data collected by the EA, which has over 7000 observation sites across England and Wales. The EA GQA scheme does not routinely determine total P. Most samples are analyses for the inorganic dissolved P fraction with P determined as orthophosphate on *unfiltered* water samples with a limit of detection of 0.0082 mg l^{-1} PO_4^{3-}-P and a reporting limit of < 0.02 mg l^{-1} PO_4^{3-}-P. Like P, total N is not routinely analysed in the GQA scheme. The most robust data are records of nitrogen as nitrate (NO_3^--N) for unfiltered water samples calculated by the difference between Total Oxidised Nitrogen (TON) and NO_2^--N. The limit of detection is 0.0294mg l^{-1} NO_3^--N and the reporting limit is < 0.2mg l^{-1} NO_3^--N.

The viability of the GQA dataset for our application is governed by the spatial distribution of the observations. The GQA sample locations are not chosen exclusively to monitor the impact of diffuse pollution on water quality; legislative drivers are particularly important (e.g. monitoring point source discharges and water abstraction). Consequently, there is some bias towards sampling sites above and below point sources such as sewage treatment works. Here, we take advantage of this bias: rather than excluding urban land cover from the analysis and taking measured nutrient concentrations and trying to apportion them into ‘point’ and ‘non-point’ sources, we use inference (see below) to work out the required risk weighting, and hence indicate the relative importance of ‘urban’ and ‘non-urban’ sources to explaining spatial patterns of water quality. Thus, a catchment where urban land covers are inferred to need a high weighting will be one where the spatial structure of measured water quality is influenced strongly by pollution associated with urban sources rather than agricultural sources (Davies and Neal, 2007). The inference works on the assumption that the location of a sewage works is approximated by the flow paths identified through an urban setting. Whilst urban drainage is commonly gravity driven, urban drainage is complex and hence there is a possibility for error arising from deviation between the flow paths inferred from topographic data in urban areas and the actual areas of the landscape (i.e. urban drainage) that contribute to a sewage works.

The GQA scheme is designed to collect one sample per month and data were available for a 15 year period, 1990-2005, with a mean of 155 observations per site. Following Davies and Neal (2007) and

Rothwell *et al.*, (2010a), and given that the analysis is at the scale of England and Wales we use the arithmetic mean rather than flow weighted GQA concentrations from each site. This allows us to take advantage of the large number of available sites that are critical to our approach, despite the lack of flow data with which to develop rating curves at these sites. The highly episodic nature of nutrient transport within rivers (Burt *et al.*, 2011; Doyle, 2005; Edwards and Withers, 2008; Walling and Webb, 1985), suggests that the lack of flow weighting may introduce error in the mean concentration estimates (Johnes, 2007) and although our own tests suggest that the number of concentrations samples is large enough to capture the range of observed discharges (see Appendix 2), the results should be considered in the light of this limitation. However, our approach is more robust to this measurement error than others, since the relative rather than absolute magnitude of the concentrations is more important in a risk based framework.

2.2.4. Inference of land cover risk weightings

The inference of land cover risk weightings used a Monte Carlo sampling framework. We undertook 5,000 model simulations, randomly selecting weightings in the range 0 to 1 for each land cover for each simulation (see Appendix 3 in the Supplementary Online Material for details on our choice of 5000 simulations). No *a priori* likelihood is assigned to these weightings. For each simulation, an objective function is determined, in this case a correlation coefficient, that describes the level of association between the water quality indicator (the spatially-distributed, mean GQA P and N concentrations) and their spatially-corresponding risk estimates.

In philosophical terms, our approach mirrors that adopted in Generalized Likelihood Uncertainty Estimation and recognised in associated problems of equifinality in hydrological models (Beven and Binley, 1992; Beven 1993). Equifinality refers to a situation where different combinations of model parameters can result in the same or similar model predictions. Most commonly, it is identified when model predictions are compared to independent measurements, and those measurements cannot distinguish between different model realisations. Our null hypothesis is that there is no systematic variation in model performance between model realisations as a function of the values of a given land use weighting. If we can reject the null hypothesis for a given land use, we can infer that a particular land use weighting influences model performance and therefore influences instream nutrient concentrations.

If we accept the null hypothesis for a land use then the influence of its weighting on model performance and therefore instream nutrient concentrations cannot be identified.

We suggest four possible reasons why the null hypothesis might be true and these are both methodological and substantive. First, it may be because a given land use has little or no coverage in a catchment. The inverse approach uses the influence of the land use on observations to define its risk so a land use must be present in order to exert an influence. If the influence is subtle and the coverage is limited the signal from this land use will be very weak. Second, high risk weightings for one land cover may offset low risk weightings in another such that optimum performances can be attained with a range of weightings for these land covers. This situation arises when the fraction of the landscape in a given land cover class (weighted by the average delivery index for that class) for each of the water quality measurement points covaries with one or more other landscape fractions. To some extent, this is a function of the available water quality data: i.e. the equifinality will becomes less severe with more measuring points, if the fractions become progressively more differentiated. However, if present, we cannot resolve this problem without additional data that avoid the covariance problem. Third, a land use class may be too broad to have a single coherent weighting if it encompasses a range of management practices and therefore of nutrient availability. Fourth, the model may not represent processes that are important in explaining the variability in observed nutrient concentrations (e.g. instream uptake). This problem is inherent to all models since they necessarily simplify the system; it tempts the developer toward ever more complex models, as discussed above. One way of establishing the models suitability may be its ability to explain the variance in the observations.

The inverse approach provides information on three properties: identifiability, influence and importance. The identifiability of a particular land cover weighting defines the extent to which we can identify an optimum risk weighting given the uncertainty associated with those individual model realisations that give the best results. We use here the standard deviation of the risk weighting for those best results and where this is lower, we can conclude that the risk weighting is more identifiable. If the risk weighting is more identifiable, we conclude that it has a greater influence over the model's performance than where it is less identifiable. However, the link between identifiability and influence can be disrupted by equifinality so that while we can infer influence from an identifiable weight, we cannot infer a lack of influence from a less identifiable weight.

The importance of a particular land cover for instream nutrient concentrations is then defined by its risk weighting (assuming that the model representation is suitable and the weighting identification is without error). Land covers with below average weightings (0.5) will lower (or dilute) the diffuse pollution risk whereas those with above average weightings will increase the risk. For weightings less than 0.5, the land cover is a 'diluter' of the diffuse pollution signal with increasing importance as the risk tends to zero. For weightings greater than 0.5, the land cover is a contributor to the diffuse pollution signal with increasing importance as the risk tends to one. Identifiability remains relevant here since it informs the degree of certainty with which the weighting can be identified. Errors in model structure (e.g. process representation) will affect the extent to which the identified weightings reflect true risk associated with a given land cover but are not represented within the identifiability since these errors can disrupt identifiability or alter identified weightings (e.g. by inflating a land cover's risk weighting to account for more efficient delivery).

To visualise the relationship between model realisations and model parameters, we plot land cover risk weighting against objective function to create 'dotty plots' (Figure 1a). These plots contain considerable scatter as a result of parameter interactions. For example, if the best objective function value is associated with a risk weighting of 0.8 for improved grassland, not all simulations with the improved grassland weighting close to 0.8 will produce good objective function values as the full set of simulations are considered, within which the weightings assigned to other land covers will not be optimal. However, pattern in the scatter (e.g. trend) suggests that the improved grassland weighting exerts an influence on the model's performance and the form of this trend gives some indication as to the range of reasonable weightings that might be assigned to that land cover class. In the illustrative example, Figure 1a shows that simulations with a high weighting for improved grassland are more likely to generate high correlations with the water quality data being used to judge each simulation. The plots also identify rough grass, horticulture and urban land covers as influential with rough grass and horticulture requiring a low risk weighting and urban areas a weighting of ~0.4. The other land covers show no clear pattern in their dotty plots, and therefore we are unable to identify their influence on the models performance or their importance as a source of nutrients in this catchment. As discussed above, not all land covers will necessarily have identifiable weightings, especially since some land covers are absent in some catchments (e.g. non-rotational horticulture in Figure 1a) and are included only to maintain a consistent approach across the catchments.

The cloud of points in the dotty plots is not uniformly dense. We convert the dotty plots into two dimensional probability density functions (pdfs) using non-overlapping bins of length 0.05 in x and y. These pdfs (Figure 1b) show the probability of achieving a given value of the objective function conditional on a particular risk weighting for the land cover class in question, and assuming a random attribution of weightings for all the other land cover classes. This allows us to understand the relationship between risk weighting and model performance better than if we look only at the trend in the upper or lower limits to the dotty plot, or view the cloudiness as an indication that land cover effects cannot be resolved.

Ranking each simulation by its correlation coefficient gives a ranked simulation list that is a measure of the likelihood of each simulation having the most correct set of weightings. Starting with the *x* most likely simulations, we determine the mean and standard deviation of the risk weightings associated with those *x* simulations, then progressively increase *x;* each time recalculating the mean and the standard deviation. The minimum value of *x* considered was 0.5% of the total number of simulations (25 simulations in this application) to enable stable calculations of means and standard deviations (after testing their stability for x = 5 : 100 for a single catchment). The plot of mean and standard deviation of weighting against correlation, which we term an 'optimisation plot' (Figure 1c) shows: (1) the land covers with clearly identifiable risk weightings, characterised by a narrow range of weightings, or a small standard deviation; and (2) the magnitude of the weighting associated with a given correlation, which determines the importance of a land cover in contributing high (weightings closer to one, diffuse pollution sources) or low levels of risk (weightings closer to zero, diffuse pollution dilution). A narrow standard deviation implies that the weighting of that land cover is identifiable; a high mean weighting (e.g. improved grassland in Figure 1c) implies that it is an important source of risk relative to other land covers; a low weighting (e.g. rough grass or horticulture in Figure 1c) implies an important source of dilution (i.e. it acts to dilute the nutrient concentrations in the catchment). High standard deviations (e.g. ~0.3 for bog, moorland, non-rotational horticulture) indicate that we cannot identify the risk weighting for that land cover either because it is uninfluential or because its influence is not identifiable due to equifinality. Finally, we use the mean and standard deviation of the optimum risk weightings in a t-test to identify the confidence with which we can reject the null hypothesis that the mean weightings are a result of random noise. Where the risk weightings are important and identifiable the null hypothesis will be rejected. Where they are either not important (mean risk close to 0.5) or not identifiable (standard deviation close to 0.3) the null hypothesis will be accepted.

Note that: (1) areal effects (i.e. large area, low magnitude) are implicitly corrected for as the estimate of risk is calculated with an upslope contributing precipitation weighting; and (2) differential connectivity effects are corrected for through the delivery index. This latter point is important as the likelihood of delivery will interact with the spatial distribution of a given land cover to determine the propensity with which a location can both generate and deliver risk. Here, we are finding the land cover weighting required for generation risk taking into account that different locations within the landscape have different likelihoods of delivery.

<Figure 1 near here >

2.3. Case study catchments

The approach has been applied to eleven catchments across England and Wales (Figure 2a) that are relatively data rich, either as a result of additional EA monitoring as part of the Catchment Sensitive Farming programme (http://bit.ly/EA-CSF) or through ongoing academic research. Land cover and land management (including livestock) data provide important indicators as to the potential source of pollutants in a catchment. In particular, the relative percentage of agriculture versus urban may be important with respect to the pathways and forms of delivery of contaminants from land to water. For each of the eleven catchments, the relative balance between agriculture (arable and improved grassland) and urban land covers is shown in Figure 2b. Rough grassland, woodland, moorland and bog are grouped together as other in this graph. For agricultural land covers and diffuse pollution risk, the ratio of improved grassland (pasture) to arable may be particularly important in characterising different forms of diffuse pollution risk. We use a pasture-arable index (PAI) to reveal the difference (% area) normalised to the total agricultural area: PAI = (A – P) / (A + P); where: P is the percentage of the catchment that is pasture; and A is the percentage of the catchment that is arable. Index values can range from -1 (all pasture) to 1 (all arable). This index is plotted relative to the hydrological regime in Figure 2c. The hydrological regime defines the connectivity between pollutant and river and is influenced by the topographic gradients within a catchment and the properties of its soil and rock and can be broadly evaluated in terms of the mean baseflow index. SCIMAP's hydrological treatment is most suited to a surface and shallow subsurface flow regime, where residence times are short and flows are predominantly lateral rather than vertical (i.e. a low base flow index; BFI).

<Figure 2 near here>

Catchments are distributed across England and Wales and range from: surface water to groundwater dominated (captured through the catchment average BFI); pasture to arable dominated (captured through the PAI); and predominantly upland to predominantly lowland. Upland catchments have higher mean elevation, with more variability in elevation and (due to orographic rainfall enhancement) tend to have higher catchment mean annual precipitation (MAP) and more variability in annual precipitation over the catchment. To generalise the findings from these catchments we have compared the means and standard deviations of the risk weightings for each land cover for N and P with a set of independent variables chosen to describe the catchments' characteristics. We use: the Ordnance Survey coordinates of the catchment centre point to represent their relative location; catchment area to define their size; mean and standard deviation of elevation to describe their topography; mean and standard deviation of mean annual precipitation to capture their rainfall conditions; mean base flow index to quantify the relative dominance of ground water or surface water; the pasture arable index to capture the relative dominance of pasture or arable land use; and the percentage cover of each land cover to establish the influence of percentage cover on mean and standard deviation of risk weighting. We regressed these catchment characteristics against means and standard deviations of risk weightings for each catchment recording the best fit from linear, power, exponential and logarithmic least squares analysis.

3. Results

A detailed analysis of the results is given below for one of the catchments – the Hampshire Avon – followed by more general analysis of the data across all eleven catchments. The detailed analysis serves to demonstrate the methodology and to illustrate how the inferred land cover weightings can be used to interpret diffuse pollution sources. The general analysis seeks to make substantive conclusions regarding diffuse pollution processes across the 11 catchments considered.

3.1. Example catchment – the Hampshire Avon

The inverse approach provides information on the influence of land cover on in-stream nutrient concentrations in the form of: (1) scatter plots (Figure 3a) and pdfs (Figure 3b) of the relationship between model performance and risk weighting assigned to each land cover; (2) optimisation plots (Figure 3c) showing the mean and standard deviation of the weightings for model runs which performed better than a given threshold; (3) optimised mean weightings and their standard deviations from the best 0.5% of model runs (Table 2); and (4) t-test results to identify the confidence with which we can reject the null hypothesis that the mean weightings are a result of random noise (Table 2). These pieces of evidence then need to be interpreted to establish the contribution of each land cover to diffuse pollution risk. In the following section we will: interpret these four outputs for woodland as an example; then show how the outputs can be combined for all the Avon land covers; and finally interpret these to draw some general conclusions about land cover types and diffuse pollution in the Avon.

<Figure 3 near here>

The dotty plots and pdfs (Figure 3a and b) show a strong negative relationship between model performance and the risk weighting assigned to woodland areas (Table 2). The optimisation plots (Figure 3c) show a consistent decline in both the mean weighting and its standard deviation as the model runs are refined to include only the best performances. For the optimum model performances: (1) mean weightings are very low suggesting that woodland should be assigned a low risk to achieve the best results; and (2) the standard deviation of the weightings is also very low suggesting that this weighting is identifiable and can be assigned with a higher degree of certainty. Table 2 shows that woodland weightings for the Avon are significantly different from the expected random weighting, at 99.9% confidence for both P and N. These outputs from the inverse approach are summarised in Table 2 and the outputs support one another to show that woodland is clearly identifiable as a low risk land cover and is an important source of dilution for both P and N.

<Table 2 near here>

Table 2 shows that the different outputs from the inverse approach are often consistent in their indication of a land cover's risk and the identifiability of that risk for a particular nutrient (e.g. rough grassland, moorland, urban, cereals, horticulture) although they may differ between nutrients. In some cases there are differences between the outputs for a single nutrient, such as improved grassland for P, where the dotty plot and pdf indicate that it exerts some identifiable influence on model performance but the significance test shows that its optimum risk weighting is not significantly different from the null case. These cases highlight a need for careful examination of the outputs together. Often these conflicting results suggest that the risk associated with a land cover is not identifiable but in some cases (e.g. improved grass for P) they can highlight important information not captured in one or more of the outputs.

Other land covers have no identifiable influence in all the outputs (dotty plots, pdfs, optimisation plots and optimised values). Bog and non-rotational horticulture do not show identifiable patterns, for P or N (Figure 3a and b), they have mean values close to 0.5 (Table 2) and standard deviations ~0.3, the expected values for a random sample from a uniform distribution. This may be because they have little or no coverage in the Avon, highlighting an important property of our approach: it cannot make predictions on the consequences of introducing new land cover types into the catchment. This is a function of the inverse approach, which uses the land cover's influence on observations to define its risk rather than defining it *a priori* (as in the forward approach).

Other land covers such as improved grassland cover a large proportion of the catchment but still display considerable scatter in their dotty plots. This may reflect: (1) equifinality due to covariance in its coverage with other land covers across the sub-catchments; (2) within class variability in the available nutrients; or (3) unrepresented processes that disrupt the signal from this land cover.

In general, the results for the Avon suggest that woodland, rough grass and moorland are low risk land covers for both P and N (Table 2). These land covers might be considered as sources of water with low P and N concentrations that dilute rather than generate diffuse pollution. Woodland in particular is consistently important as a 'diluting' land cover in the Avon. The differences in the strengths of relationships between rough grass and moorland (Table 2) suggest that rough grass can be more confidently identified as a source of dilution for P while moorland is more identifiable for N. It is important to stress that the risk weightings that we have identified for land covers in the Avon are relative (rather than absolute). As such, no matter how low the instream concentrations in a catchment, there will always be some land covers contributing more nutrients and others contributing less (acting to dilute).

Land covers identified as high risk by the inverse approach appear to have differing signals according to the nutrient being considered. Urban land cover appears an important source for P but not for N in the Avon (Table 2, Figure 3), potentially linked to point source P inputs at sewage treatment works. Likewise, horticulture appears to be an important source for P but is less important for N (Table 2). This may be associated with excess nutrient applications to horticultural crops. Unpublished data using the Phosphorus Indicators Tool (Heathwaite *et al.* 2003a) identified horticultural land cover as a high source of P with a high delivery potential. Haygarth (2004) suggests that glass house and nursery stock pose a high risk of nutrient loss through excess use of liquid fertiliser. Relative to the value of the crop the cost of nutrient fertilizers is low. Cereals in the Avon have a low P risk (Table 2, Figure 3c) but very high N risk and may be associated with locations that favour subsurface nutrient flux over surface runoff. The spatial distribution of cereal crops in the Avon, which is predominantly chalk with a high Base Flow Index (BFI, Table 8) tends to be focused on plateau areas some distance from receiving waters. Thus, hydrological connection will be infrequent and delivery risk may be overestimated for nutrients which rely on surface pathways for delivery, while those that can be effectively transported by groundwater flow will remain connected.

3.2. Extensive analysis of all 11 study catchments

3.2.1. Phosphorus

For P, cereals and horticulture are an important source of risk (i.e. risk weighting significantly greater than average 0.5) in only one catchment and three catchments respectively. Despite its limited coverage non-rotational horticulture is an important source of risk in two other catchments (Table 3). This suggests that arable land cover is sometimes a source of P in UK catchments. However it is neither the most consistent nor the most dominant source. Urban land covers require a high risk weighting in nine out of eleven catchments, with seven of these requiring a weighting greater than 0.77 (Table 3). Improved grassland is important (i.e. has risk weightings that are significantly different from the null case) in six of the eleven catchments but has an above average weighting in only three of these (Table 3). The risk weightings for land covers associated with extensive land use practices (e.g. rough grass, moorland and woodland) are clearly identifiable (they have low standard deviations) and are generally important sources of dilution (they have low mean risks). Rough grass has significant weightings in ten of the eleven catchments and needs to be given a low weighting (<0.23) in all of these (Table 3). Moorland and woodland weightings are significant in fewer catchments (3 for each) but almost always require a low weighting. Bogs cover only a very small proportion of any catchment and have no significant weightings.

< Table 3 near here>

3.2.2. Nitrogen

For N, one or more arable land covers are an important source of risk in eight out of eleven catchments. Of the remaining three, urban land cover is important for two catchments and improved grassland for the other. Cereals, horticulture and non-rotational horticulture appear to represent an important source of risk in five, five and two catchments respectively (Table 4). Improved grassland appears to be a more important land cover for N than for P, although its influence is still highly variable. It has significant weightings in eight of the eleven catchments but has an above average weighting in only four of these (Tables 3 and 4). For the extensive land uses (rough grass, moorland, woodland) the results for N are broadly similar to P. Rough grass has a significant weighting in fewer catchments for N as compared with P (eight out of eleven) but must be given a low weighting in all of these catchments. Woodland has a significant weighting in seven catchments and is generally low risk (<0.19 for five catchments) but occasionally a source of risk (>0.69 for two catchments). Moorland has more significant weightings in more catchments for N than for P, and almost always requires a low weighting with seven catchments in which its risk weighting must be set to <0.36 (although it has a high weighting for the Frome).

<Table 4 near here>

3.2.3. Land cover areal extent effects

Tables 3, 4 and 5 show that, notwithstanding the upslope contributing precipitation weighting, the land covers that exert a significant influence on model performance are often those that cover the largest proportion of the catchment. We explored this observation by considering the standard deviation (SD) of the risk weightings from the best 0.5 % of model runs for each land cover for each nutrient. Lower standard deviations imply that a particular land cover has a risk weighting that is clearly identifiable using the inverse approach. Generally, as the area covered by a given land cover increases, so the standard deviation becomes narrower (Figure 4). Non-rotational horticulture, which has generally low catchment cover, has a range of standard deviation values, suggesting that its identifiability varies between catchments, and is generally uninfluenced by the proportion of the catchment it covers (Figure 4a and c). Urban land cover accounts for <10% of all catchments studied but often has a low standard deviation (indicating that its risk weighting is often clearly identifiable) and appears to be independent of percentage cover. Cereals and horticulture vary in percentage cover from 2% to 36% but the standard deviations are negatively correlated with the percentage cover (**Error! Reference source not found.**), suggesting that where these land covers have a high spatial coverage they tend to be clearly identifiable (Figure 4a and c). Improved grassland often has a very large share of the catchment but there is no negative relationship between percentage cover and standard deviation. Even high percentage covers have high standard deviations. This suggests that the weighting associated with improved grassland is often very difficult to identify even when it makes up a very large proportion of the catchment. As in the specific case of the Avon, this may reflect: equifinality due to covariance in its coverage with other land covers; within class variability in the available nutrients; or unrepresented processes that disrupt its influence.

<Table 5 and Figure 4 near here>

3.2.4. Catchment Characteristics

We have tested the ability of a set of independent variables that represent catchment characteristics to predict the variance in both the mean (representing importance as a source of risk or dilution) and the standard deviation (representing identifiability) of the risk weightings assigned to each land cover.

Percentage cover is consistently the most effective predictor of the standard deviation of the weightings (**Error! Reference source not found.**). For P, it is the only significant predictor for 3 land covers (horticulture, non-rotational horticulture and woodland) and the dominant predictor (highest r^2) for one other (cereals). For N, it is the only significant predictor for one land cover (urban) and the dominant predictor for two others (rough grass and moorland). In every case identifiability improves as percentage cover increases. The other catchment characteristics have a less consistent influence on identifiability, either in terms of the number of land covers that they influence or the direction of their influence (i.e. to improve or reduce identifiability). There are only two land covers for P (rough grass and moorland) and two for N (cereals and woodland) where another variable is a better predictor of identifiability than percentage cover (**Error! Reference source not found.**).

For P, moving north across the UK, risk weightings for rough grass become more identifiable (negative trend between northing and standard deviation of risk weighting r^2=0.53). The weightings for moorland become less identifiable as catchments become more pasture rather than arable dominated; they become more identifiable in upland catchments (where elevation and average annual rainfall are both higher and more variable). For both N and P, the risk weighting for cereals becomes less identifiable in upland catchments and more identifiable with distance east and in pasture dominated catchments (**Error! Reference source not found.**). For P these relationships are weak relative to the strong influence of percentage cover. Whereas for N the influence of percentage cover is weaker.

Percentage cover exerts little influence on mean weightings (it is a significant predictor in only four land covers, all for N, and dominant in only one); instead, mean weightings are more effectively predicted by other catchment characteristics (

Table 6). For P, moving north across the UK, mean risk weightings for rough grass tend to decrease while those for improved grassland increase (
Table 6). Upland catchments have higher weightings for rough grass, improved grassland, urban areas and horticulture. This is reflected in strong correlations with mean elevation and its variability and with catchment average annual rainfall and its variability. The relationships between catchment characteristics and the risk weighting assigned to improved grassland are particularly strong. Improved grassland risk weightings increase significantly with distance north (r^2=0.52;
Table 6) and with increased variability in rainfall and elevation (r^2=0.57) suggesting that in upland-dominated northern catchments, improved grassland constitutes a more important source of risk than for lowland catchments in the south. However, the dominant control on improved grassland risk weighting for P appears to be catchment average BFI (r^2=0.91). This suggests that improved grassland represents a lower risk land cover for P in ground water than in surface water dominated catchments. These relationships are much stronger than the very limited control that percentage cover exerts on the improved grassland risk weighting (r^2=0.05).

<
Table 6 and 7 near here>

There are more significant relationships between catchment characteristics and mean risk weightings for N than for P (

Table 6) suggesting that the mean risk weightings for N are more sensitive to these characteristics. This is perhaps unsurprising given the dominance of urban point sources for P.

For N, northern catchments have lower risk weightings for rough grass (r^2=0.43) and high weightings for woodland (r^2=0.42) while eastern catchments have high weightings for rough grass (r^2=0.43) but lower for cereals and horticulture (r^2 of 0.44 and 0.57 respectively in

Table 6). As catchments become increasingly dominated by upland areas the risk weightings for rough grass fall while the weightings for cereals and horticulture increase (Table 6). This is unlikely to be related to differences in the percentage cover of these land covers in upland and lowland catchments since percentage cover is a relatively weak predictor of weighting for these covers. As catchments become increasingly groundwater rather than surface water dominated, the weightings for woodland and improved grassland reduce (r^2 of 0.52 and 0.41 respectively in Table 6).

3.2.5. Overall Model Performance

Table 8 shows the correlation coefficients of the best model performances for P and N. In general, the results are encouraging and do suggest that this approach can reconcile a significant amount of the spatial variability in the statutory water quality data used here. The results also suggest that SCIMAP is calibrated more effectively for N than P.

Figure 5 suggests that SCIMAP performs well for upland catchments (positive correlation with mean and standard deviation of catchment elevation) that have higher and more variable annual rainfall; while it performs less well for catchments that are groundwater dominated (negative correlation with base flow index) and predominantly arable (negative correlation with pasture arable index). The model is more sensitive to these controls for N than P, and the model performance for Nitrate appears to be particularly strongly controlled by variability in annual rainfall across the catchment and average elevation. These two variables are likely to be strongly related to one another through the control that orographic rainfall enhancement exerts on the spatial variability in rainfall across UK catchments. Maximum correlation coefficients for N are very low for lowland catchments with little rainfall variability but rise very rapidly so that they are all >0.8 for catchments with mean elevation >100 m and standard deviation of annual rainfall >100 mm/a. These controls may reflect the hydrological basis of the analysis of connectivity used here. In particular, the model's assumptions are best suited to catchments where steeper topographic and hydraulic gradients and impermeable bedrock encourage lateral surface or subsurface flow rather than deep infiltration and ground water flow. The recharge of streams by groundwater sources may both dilute but also re-introduce N and P in ways that are not represented in our analysis.

<Table 8 and

Figure 5 near here>

4. Discussion

The relative risk weighting assigned to each land cover is not consistent across the 11 catchments investigated here, suggesting that the importance of a particular land cover in contributing to a given in-stream water quality parameter is both geographically-variable (across the study catchments considered) and nutrient dependent (e.g. Table 3). This was confirmed by comparison of inferred risk weightings with catchment characteristics (

Table 6). It implies that it will be difficult to identify universal nutrient availability risks for particular land covers that can be applied to all catchments as in the export coefficient modelling approach (e.g. Johnes, 1996; Johnes *et al.*, 2007) and the phosphorus indicators tool (Heathwaite *et al.*, 2003a, 2005a). Thus, a careful consideration of catchment characteristics will be needed *a priori* in defining risky land covers. Inverse approaches have a role to play in identifying for a given catchment and parameter (e.g. nutrient) those land covers that are most likely to be sources. Approaching the inverse problem for water quality without both a risk generation and a risk connection treatment is likely to be difficult because spatial variability in connectivity and dilution effects may impart significant spatial variability on the water quality data in ways that make finding the land cover signal particularly difficult.

The level of variability between catchments is surprising since we might expect the risk associated with each land cover to be similar (at least in relative terms). To some extent, the variability may reflect differences in risk availability on these land covers resulting from different land management practices between catchments. However, it is also likely to reflect the spatial structure of the catchment in terms of the dominance of particular land cover types (Davies and Neal, 2007;

Figure 4). Further, at least some of the variability in risk weighting may be related to an implicit parameterisation of hydrological processes that are not currently represented in SCIMAP. This is illustrated in the detailed analysis for the Hampshire Avon and the differences general improvement in model performance in surface water dominated catchments (

Figure 5). Our results illustrate the importance of inverse approaches in situations such as this, where there are known and possible process inadequacies that can't be dealt with easily through model reformulation. Such inadequacies will be manifest as reductions in the extent to which good results (here correlations with the water quality data) can be found.

We can be more certain about the land covers that represent low risks (rough grass, moorland and woodland). These land covers are important since they act to dilute nutrient fluxes from other sources in the catchment. This is perhaps unsurprising since they are areas expected to have little or no nutrient application (Jackson, 2000). However, it is encouraging for the model that these land cover types are consistently identified as low risk without any kind of *a priori* tuning. In fact the model highlights an important point: that land uses like rough grazing, moorland and woodland have an important contribution to make to the overall spatial signal of instream nutrient concentrations whilst they are maintained in a low input state. If their status changes and their capacity to dilute is affected this may have important consequences for water quality downstream.

In general terms, urban land covers tend to need high weightings for in-stream P concentrations in many of the catchments. This is consistent with recent UK studies linking P concentrations to the percentage urban cover (Rothwell *et al.*, 2010a), population density (Davies and Neal, 2004, 2007) and number of known point sources (Rothwell *et al.*, 2010b) in catchments. It suggests the continued importance of point source pollution for in-stream P concentrations, particularly from sewage treatment works (Jarvie *et al.*, 2006; Muscutt and Withers, 1996; Neal *et al.*, 2010b). A key advantage of the inverse approach used here (taking account of possible bias of sample sites to urban areas) is that it is not necessary to separate *a priori* a possible point signal from a non-point signal, so resolving the dilemma regarding the relative significance of point and non-point sources. This significance is revealed through the analysis, which identifies when non-urban sources are dominant.

For N, we found that arable areas were a more important source of risk. This is consistent with the results of statistical analysis by Ferrier *et al.* (2001), who found nitrogen concentrations in Scottish rivers to be highly correlated with the extent of arable land. Other UK studies also found that the extent of arable land was a significant predictor for N but much less significant for P concentrations (Davies and Neal, 2007; Rothwell *et al.*, 2010a, 2010b).

The optimum risk weightings do not identify improved grassland as a dominant driver for N or P, this is surprising given the high levels of N and P application associated with this land cover (Johnes, 1996; Johnes and Butterfield, 2002); and results from export coefficient modelling (Johnes *et al.*, 2007), which highlight livestock waste as an important contributor to diffuse agricultural nutrient loading. However, Davies and Neal (2007) also found no relation between grassland cover and P concentration suggesting that the strong urban signal masks the contribution from improved grassland. The dotty plots for individual catchments often show complex patterns for improved grassland. In some cases high risk weightings producing reasonable model performances but low to medium risks were required for the best model performances; and those low-medium risks then have the best and some of the worst model performances. This may be in part a result of the true relative risk associated with improved grasslands, which have intermediate nutrient application and availability. However, it may also reflect equifinality due to covariance in its coverage with other land covers; the influence of unrepresented processes; or limits to the available land cover data which are unable to distinguish between grasslands that are managed in very different ways and as a result should be assigned different risk weightings (the model's data requirements and the limits to current data are discussed in detail below).

An important distinction between our approach and others is our simple but explicit representation of the probability that material will be delivered to the river network. There is a recognition in the literature: that this probability of delivery is important in defining diffuse pollution risk (Beven *et al.*, 2005; Haygarth *et al.*, 2005); that some parts of the catchment are more connected than others both in terms of the frequency and duration of connection (Bracken and Croke, 2007; Jensco *et al.*, 2009; Lane *et al.*, 2009); and that representing this connectivity is essential to effectively capturing delivery (Frey *et al.*, 2008; Heathwaite *et al.*, 2005b; Neumann *et al.*, 2010). There is also a recognition that attempts to capture connectivity and delivery resort to spatially explicit models that are data hungry and computationally intensive (Neumann *et al.*, 2010; Radcliffe *et al.*, 2009); and that these models are too complex to provide predictions at a fine enough resolution over areas large enough to be relevant for decision making (Heathwaite, 2003; Neumann *et al.*, 2010). Our approach, using a simple static metric for hydrological connectivity, which has been shown to capture both the frequency and duration of connection (Lane *et al.*, 2009), enables us to run the model over large areas (e.g. ~2300 km^2 for the River Eden catchment) at fine resolutions (<20m). Figure 3 shows that there is some uncertainty in the inferred weightings that optimize our water quality measures. As it is easy to propagate these uncertainties through the risk analysis, it provides the basis of balancing: the feasibility of the kind of interventions that might reduce risk; their locations; their spatial extents; and the number of interventions; given the uncertainties associated with particular land use effects. Any intervention will, equally, be sensitive to changes in the spatial scale of impact: locations with a more certain land use weighting; that are well-connected; and larger in extent are more likely to have effects that propagate through to larger spatial scales. Changing small patches of land of a few 100 m^2 is unlikely to be detectable given these uncertainties, but this approach still provides a means of delivering decision maker needs in relation to the prioritization of sub-catchments and reaches (Heathwaite, 2003; Johnes *et al.*, 2007) and the spatial extent over which prioritized reaches might impact downstream. We emphasise the need, however, to be careful regarding spatial scale effects below. The model proceeds by time-integrated, rather than a dynamic treatment of the system and a risk based rather than explicit nutrient balance approach. We feel that these simplifications are appropriate in developing a tool for prioritization where time integrated relative risk is the crucial factor; and given the available data. The difficulty with our approach is the underpinning assumption of what element of the hydrological cycle drives water quality response, in this case surface and shallow subsurface flows, and this is reflected in the poorer optimisation for catchments with higher BFI and hence groundwater impacts.

Our model requires three sets of spatial information. Firstly, it requires information on the connectivity of each location in the catchment to the river network. This is derived from fine resolution topographic data under a set of assumptions and is used to define the delivery index. Given the availability of fine resolution topographic data it is likely to be the assumptions (e.g. exponential decline in hydraulic conductivity with depth, surface parallel water table; Lane *et al.*, 2006) rather than the data that limit this component.

Secondly, the model requires a data set that identifies units of land that we expect to be similar in terms of their nutrient availability (these are not limited to the land cover classes used here). The more internally consistent these units are (i.e. the more between class rather than within class variability) the more effective an inverse approach will be at identifying the risks associated with them. Therefore, the suitability of our land cover based classes will be defined by: 1) the strength of the association between land cover, land use and management; and 2) the spatial resolution at which the land cover can be resolved, this will be particularly important in heterogeneous landscapes where there is a patchwork of different land covers with different availabilities. This highlights an issue for diffuse pollution modelling if the data on nutrient availability is limited both in spatial resolution and level of detail. In our case, satellite derived land cover data are the best available data for the UK but are unable to distinguish between grassland areas with very similar spectral signatures but very different management practices (e.g. silage vs. permanent grazing). Some land cover classes (e.g. horticulture and cereals) are spectrally very different but may reflect differences for that year rather than in the long term where both sets of fields may be managed in the same way (e.g. crop rotation); the land use in these areas and as a result the long term nutrient availability may be similar or even the same. In other settings, with better land cover measurement systems, this may be less of an issue.

Finally, in-stream observations drive the risks assigned to these availability units (land covers), the choice of observation will define the risks that are assigned. For example, different risk weightings assigned to N and P in this study. These data need to reflect the interest for the catchment (e.g. N or P in this study); they need to be time-integrated (in our case using the mean of the observations) but representative (this can be particularly difficult for nutrients mobilized in storm events). The observations need to be spatially rich and the inverse approach hinges on having observations sites that contain a mix of land cover types and that mix being different from one observation point to the next. Large numbers of observation points (e.g. >100 for the Eden catchment) will enable both a good mix of upstream land covers and redundancy between points, ensuring that no single observation is defining the identified risk weightings. Importantly though, this method does not require that the observation points are independent of one another and as a result nested catchments and sub-catchments can all provide useful data for the inverse approach.

In using this approach, we emphasise that there are three critical elements of the approach that may be problematic. First, as we explained in the methodology, the observation that there is no systematic relationship (e.g. in a ‘dotty plot’) between an Objective Function describing model performance and the values of a land cover weighting has four different interpretations. First, the land cover does not exert an influence on the model's performance and as such is not important either as a source of risk or dilution. Second, the land cover does exert an influence on model performance but it is not identifiable as a result of equifinality due to covariance in the coverage of land cover classes. Third, the data are inadequate to resolve the influence of the land cover class (e.g. where nutrient availability is highly variable within a class). Fourth, the model may not represent processes that are important in explaining the variability in observed nutrient concentrations. In the second case, the available water quality data are unable to resolve the influence of this land cover. If the analysis was undertaken over a smaller spatial scale, with a very high density of monitoring sites, then this parameter might be shown to be important. This analysis is, in effect, a relative one in that its findings apply to the spatial extent over which the water quality data are available. Care must be shown in considering spatial units very much smaller or very much larger than suggested by these data. However, we emphasise that when a systematic relationship is found then this provides very important information at the scale of analysis over what is contributing to the observed spatial variation in a water quality parameter. The second critical element of the approach is related to the spatially-distributed water quality data themselves. Such datasets are rare and even where available may not have sufficient temporal resolution to be representative of the actual water quality signal at a point in a catchment. It is necessary to consider: (1) the representativeness of the spatial distribution of sites; and (2) the representativeness of the temporal variability in water quality; through a careful analysis of those data; before deciding to use them in the manner we demonstrate in this manuscript. Third, some of the variability in the risk weightings between catchments may be related to an implicit parameterisation of unrepresented hydrological processes (e.g. groundwater flow) arising from incorrect assumptions in SCIMAP. Such assumptions are likely to be manifest in lower levels of optimal agreement between predictions and observations and this level of agreement, in a Bayesian approach, may be a useful wider indicator of the extent to which the assumptions being made in the model are acceptable. Such an evaluation needs to be catchment-by-catchment, and checked against other contextual data such as BFIs.

5. Conclusion

The inverse approach developed in this paper allows us to draw four broad conclusions. First, the relative risk weightings assigned to each land cover are not consistent across all catchments, suggesting that the importance of a particular land cover in contributing to river water quality is variable between catchments. Second, some of this variability is due to catchment properties suggesting that diffuse pollution policy needs to be carefully tuned on a catchment-by-catchment basis to reflect both the land cover mix and catchment characteristics. Third, trends differ between the two nutrients, P and N, considered here. For P, urban land covers are often high risk; rough grass and moorland are generally low risk; improved grasslands are intermediate risk and arable land covers do not always require a high risk weighting, although this may be partly because the measured water quality data are unable to resolve arable land cover effects due to equifinality resulting from covariance in the coverage of arable land covers. Point source pollution reflected in the weightings given to urban land covers appears to exert a dominant control on in-stream P concentrations in many, but not all, catchments. For N, urban land covers are less dominant and often low risk; rough grass and moorland remain low risk; and arable land covers are generally important N sources in many catchments. This highlights the ability of our analysis to identify when non-urban sources are dominant, resolving the dilemma regarding the relative significance of point and non-point sources and negating the need for their *a priori* separation. Finally, differences in the dominant pollution source depending on the pollutant raise intriguing questions about whether they result from differences in nutrient availability or in delivery. Improved model performance for N relative to P suggests that this is at least partly related to delivery and may reflect differences between nutrients in: hydrologic flow paths; the extent to which they are conservative during transport; and / or the ease with which they can be measured.

One final theme emerges from this paper: the kinds of generalisation that might be possible in relation to possible diffuse pollution causes. Ideally, we would have shown that it is possible to isolate a subset of predictors that can be used to profile diffuse pollution risks in any one catchment. Such a generalisation would then allow the refinement of diffuse pollution policy. Work with further predictors might lead to such a generalisation but such work may also be misplaced as it assumes that all the possible candidates for a more complex generalisation are both known and quantifiable. The generalizable element of this paper is not the relative importance of land covers, but rather a methodology that combines a relatively simply model with spatially-distributed extant measurements, to infer the parameters that matter. The simplicity of the model (and the associated Monte Carlo method) means that the analysis is not computationally demanding, can be fully automated, and yields information on the uncertainty of model findings simultaneously with model predictions. As such, it may be a preferable approach than using a more complex model where the data and computational demands of a more complex model cannot be readily met.

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Tables

Table 1: Centre for Ecology and Hydrology Land Cover Map for 2000 classes and their translation to SCIMAP classes. Land covers that were either absent from the catchments or expected to contribute zero risk are classed as other (modified from Jackson, 2000 and Fuller *et al.*, 2002).

Land cover		Description	SCIMAP Class
Broad-leaved woodland	1.1	deciduous, mixed, open birch, scrub	Woodland
Coniferous woodland	2.1	conifers, felled, new plantation	Woodland
Arable cereals	4.1	barley, maize, oats, wheat, cereal (spring), cereal (winter)	Cereals
Arable horticulture	4.2	arable bare ground, carrots, field beans, linseed, potatoes, peas, oilseed rape, sugar beet, mustard, non-cereal (spring)	Horticulture
Non rotational horticulture	4.3	orchard, arable grass (ley), setaside	Non Rotational Horticulture
Improved grassland	5.1	intensive, grass (hay/ silage cut), grazing marsh	Improved grassland
Set aside grass	5.2	grass set aside	Rough grass
Neutral grass	6.1	rough grass (unmanaged), grass (neutral / unimproved)	Rough grass
Calcareous grass	7.1	calcareous (managed), calcareous (rough)	Rough grass
Acid grass	8.1	acid, acid (rough), acid with Juncus, acid with *Nardus/Festuca/Molinia*	Rough grass
Bracken	9.1	Bracken	Rough grass
Dwarf shrub heath	10.1	dense ericaceous, gorse	Moorland
Open dwarf shrub heath	10.2	ericaceous, gorse	Moorland
Fen, marsh, swamp	11.1	swamp, fen/marsh, fen willow	Bog
Bog	12.1	bog: shrub, grass/shrub, undifferentiated (all on deep peat)	Bog
Water (inland)	13.1	water (inland)	Other
Montane habitats	15.1	Montane	Moorland
Inland Bare Ground	16.1	despoiled, semi-natural	Other
Suburban/rural developed	17.1	suburban/rural developed	Urban
Continuous Urban	17.2	urban residential/commercial, urban industrial	Urban
Supra-littoral rock	18.1	Rock	Other
Supra-littoral sediment	19.1	shingle, shingle (vegetated), dune, dune shrubs	Other
Littoral rock	20.1	rock, rock with algae	Other
Littoral sediment	21.1	mud, sand, sand/mud with algae	Other
Saltmarsh	21.2	saltmarsh, saltmarsh (grazed)	Other
Sea / Estuary	22.1	Sea	Other

Table 2: a summary of the influence of each land cover type on Orthophosphate (PO_4^{3-}-P) and Nitrate (NO_3^--N) risk inferred from inverse modelling for the Hampshire Avon including: a description of the relationship between risk weighting and model performance from the dotty plots and pdfs; the mean and standard deviation of the optimum risk weightings; and whether these mean risks were significantly different from those expected from random sampling. Bog land cover is not listed since it is not present in the Avon.

		Orthophosphate				Nitrate			
Land cover type	Cover (%)	Correlation risk v model fit	Optimum mean/ standard deviation	Signif	Summary of Influence on P risk	Correlation risk v model fit	Optimum mean/ standard deviation	Signif	Summary of Influence on N risk
Improved grassland	29	weak (+)	0.61/0.23	no	medium risk	none	0.57/0.25	no	not influential
Rough grass	11	weak (-)	0.24/0.16	99%	low risk	v weak (-)	0.27/0.19	95%	low risk
Moorland	2	weak (-)	0.28/0.21	95%	low risk	strong (-)	0.09/0.09	99.9%	low risk
Urban	8	weak (+)	0.65/0.26	90%	high risk	none	0.44/0.29	no	not influential
Cereals	22	strong (-)	0.16/0.08	99.9%	low risk	v strong (+)	0.78/0.18	99%	v high risk
Horticulture	14	v strong (+)	0.83/0.13	99%	v high risk	weak (+)	0.69/0.19	95%	High risk
Non rotational Horticulture	1	none	0.45/0.30	no	not influential	none	0.53/0.29	no	not influential
Woodland	13	strong (-)	0.12/0.09	99.9%	v low risk	v strong (-)	0.05/0.04	99.9%	v low risk

Table 3: optimised land cover risk weightings from SCIMAP based on the GQA in-stream Orthophosphate (PO_4^{3-}-P) measurements in 11 UK catchments. Mean risk weightings that are significantly different from those expected based on random sampling with >90% (bold), 95% (*), 99% (**) and 99.9% (***) confidence are in red where they are high and blue where they are low risks. Blank entries result where that land cover is absent from a catchment.

Land cover	Avon	Deben	Eden	Frome	Rother	Slapton	Till	Wensum	Wye	Wyre	Yealm
Improved grassland	0.61	0.40	0.38	**0.06*****	0.57	0.56	**0.71***	**0.13*****	**0.65**	**0.75****	**0.22****
Rough grass	**0.24****	**0.09*****	**0.16*****	**0.23****	0.42	**0.31***	**0.07*****	**0.16*****	**0.34***	**0.13*****	**0.16*****
Moorland	**0.28***	0.36	**0.19****	0.50		0.51	0.41		**0.12*****	0.52	0.37
Bog		0.48	0.62	0.50			0.59	0.46	0.55	0.45	0.48
Urban	**0.65**	**0.13*****	**0.82****	**0.69***	**0.82****	**0.77****	**0.77****	**0.85*****	**0.78****	**0.30***	**0.87*****
Cereals	**0.16*****	**0.84*****	0.65	**0.34***	**0.09*****	**0.32***	**0.24****	**0.18****	**0.29***	0.55	**0.31***
Horticulture	**0.83****	**0.28***	0.51	**0.13*****	**0.15*****	**0.69***	0.53	**0.24****	**0.78****	**0.16*****	0.40
Non Rotational Horticulture	0.45	**0.31***	**0.65**	**0.68***	0.44			0.53	0.53		
Woodland	**0.12*****	**0.31***	**0.14*****	**0.71***	**0.11*****	0.46	0.51	0.47	**0.35**	0.60	0.45

Table 4: optimised land cover risk weightings from SCIMAP based on the GQA in-stream Nitrate (NO_3^--N) measurements in 11 UK catchments. Mean risk weightings that are significantly different from those expected based on random sampling with >90% (bold), 95% (*), 99% (**) and 99.9% (***) confidence are in red where they are high and blue where they are low risks. Blank entries result where that land cover is absent from a catchment.

Land cover	Avon	Deben	Eden	Frome	Rother	Slapton	Till	Wensum	Wye	Wyre	Yealm
Improved grassland	0.57	**0.66**	0.56	**0.02*****	**0.26****	**0.77****	**0.72***	**0.06*****	0.42	**0.71***	**0.37**
Rough grass	**0.27***	**0.21****	**0.12*****	0.43	**0.29***	0.62	**0.10*****	0.52	**0.13*****	**0.16*****	**0.14*****
Moorland	**0.09*****	**0.36**	**0.15*****	**0.65**		0.44	**0.09*****		**0.22****	**0.19****	**0.08*****
Bog		**0.66**	0.53	0.52			0.49	0.54	0.64	0.49	0.54
Urban	0.44	**0.83****	0.40	0.56	**0.87*****	**0.25***	0.58	0.45	0.58	**0.06*****	**0.85*****
Cereals	**0.78****	**0.63**	**0.73***	0.56	**0.11*****	**0.64**	0.56	**0.04*****	**0.69***	0.52	0.56
Horticulture	**0.69***	**0.14*****	0.63	0.49	**0.26****	**0.85*****	**0.87*****	**0.77****	**0.80****	0.64	0.41
Non Rotational Horticulture	0.53	0.42	**0.72***	**0.80****	0.52			0.43	0.61		
Woodland	**0.05*****	**0.14*****	**0.10*****	0.59	**0.19****	0.35	**0.69***	0.55	**0.17*****	**0.71***	0.44

Table 5: the percentage area covered by each of the SCIMAP land cover classes for each of the catchments under consideration.

Land cover	Avon	Deben	Eden	Frome	Rother	Slapton	Till	Wensum	Wye	Wyre	Yealm
Improved grassland	28.6	4.4	44.3	29.1	30.4	35.5	20.3	7.9	32.8	42.9	36.0
Rough grass	11.2	12.5	25.2	4.8	10.0	8.3	14.8	6.5	30.1	13.8	17.5
Moorland	1.6	0.8	7.6	4.9	0.0	0.1	5.1	0.0	4.8	6.0	5.7
Bog	0.0	0.6	2.8	0.3	0.0	0.0	0.8	0.1	0.2	1.9	0.5
Urban	8.1	7.2	2.6	7.5	5.3	7.1	1.3	6.9	3.9	10.8	7.5
Cereals	22.2	31.3	2.9	21.1	15.6	24.5	25.8	36.0	4.8	3.6	13.5
Horticulture	13.9	28.4	1.6	14.4	18.0	12.9	23.5	32.9	8.5	13.4	7.1
Non Rotational Horticulture	1.3	2.4	2.9	2.8	0.3	0.0	0.0	0.1	0.1	0.0	0.0
Woodland	12.8	8.3	8.8	12.5	19.1	6.9	7.5	9.2	14.2	4.7	10.9

Table 6: r^2 values for significant correlations (>95% confidence) between catchment characteristics and the mean risk weighting for each catchment for a given land cover. Red text indicates positive correlations blue text indicates negative correlations. Bold text indicates r^2 values significantly different from zero at 99.9% confidence.

Land cover	Northing	Easting	MAP	MAP sigma	Elevation	Elevation Sigma	BFI	% cover
Improved grassland	**0.52 (P)**			0.42 (P)		**0.57 (P)**	**0.91 (P)** 0.41 (N)	0.41 (N)
Rough grass	0.41 (P) 0.43 (N)	0.43 (N)		0.43 (N)	0.43 (N)	**0.76 (N)**		**0.56 (N)**
Urban					0.32 (P)			**0.66 (N)**
Cereals		0.44 (N)			0.33 (N)	0.31 (N)		
Horticulture		**0.56 (N)**	0.35 (N)	0.39 (N)	0.45 (P) **0.56 (N)**			
Woodland	0.42 (N)						**0.52 (N)**	**0.47 (N)**

Table 7: r^2 values for significant correlations (>95% confidence) between catchment characteristics and the standard deviation of risk weighting for each catchment for a given land cover. Red text indicates positive correlations blue text indicates negative correlations. Bold text indicates r^2 values significantly different from zero at 99.9% confidence.

Land cover	Northing	Easting	Area	MAP	MAP sigma	Elevation	Elevation Sigma	PAI	% cover
Rough grass	**0.53 (P)**				0.42 (N)	0.46 (N)	**0.51 (N)**		**0.88 (N)**
Moorland				0.40 (P)	**0.66 (P)**	**0.64 (P)**	**0.54 (P)** 0.41 (N)	**0.54 (P)**	**0.47 (P)** **0.52 (N)**
Urban									0.33 (N)
Cereals		**0.50 (N)**		0.42 (P) 0.38 (N)	0.39 (P) 0.36 (N)		0.34 (P) 0.57 (N)	0.46 (P) 0.42 (N)	**0.77 (P)** 0.39 (N)
Horticulture									0.44 (P)
Non rotational horticulture									0.35 (P)
Woodland			0.36 (N)						0.41 (P)

Table 8: correlation coefficients for the optimum SCIMAP model performance based on the GQA in-stream measurements of Orthophosphate (PO_4^{3-}-P), and Nitrate (NO_3^--N) in 11 UK catchments with the values for independent variables used to represent catchment characteristics. The ordnance survey grid reference of the catchment centre point; mean and standard deviation of mean annual rainfall; mean and standard deviation of elevation; catchment area; the pasture arable index; and mean base flow index. Correlation coefficients are labelled where they are significant with 95% (*) and 99.9% (**) confidence.

Catchment	OS Grid Reference	Mean (σ) annual rainfall (mm/a)	Mean (σ) elevation (m)	Area (km^2)	Pasture -Arable Index	Base Flow Index	Optimum Correlation Coefficients	
							Orthophosphate	Nitrate
Avon	407200,136491	806 (47)	120 (47)	1716	0.13	0.87	0.71^{**} (n=54)	0.89^{**} (n=50)
Deben	629571,254581	582 (10)	29 (18)	745	0.87	0.58	0.68^{**} (n=32)	0.57^{**} (n=32)
Eden	355981,534572	1162 (328)	242 (153)	2274	-0.71	0.48	0.71^{**} (n=80)	0.86^{**} (n=80)
Frome	377582,92295	892 (50)	88 (62)	867	0.14	0.70	0.36^{*} (n=48)	0.60^{**} (n=40)
Rother	580450,126894	775 (52)	47 (42)	571	0.05	0.42	0.63^{**} (n=41)	0.72^{**} (n=38)
Slapton	277209,44517	1023 (69)	81 (46)	135	0.03	0.61	1.00^{**} (n=6)	0.99^{**} (n=6)
Till	393953,634235	725 (86)	137 (129)	1286	0.42	0.46	0.77^{**} (n=25)	0.93^{**} (n=25)
Wensum	599667,320954	661 (20)	49 (16)	699	0.79	0.64	0.78^{**} (n=17)	0.28 (n=17)
Wye	321295,243964	1063 (288)	242 (140)	3049	-0.42	0.54	0.83^{**} (n=108)	0.92^{**} (n=107)
Wyre	347216,444615	1099 (173)	77 (112)	561	-0.43	0.44	0.87^{**} (n=37)	0.92^{**} (n=37)
Yealm	261514,55322	1300 (212)	144 (123)	215	-0.27	0.57	0.66^{*} (n=16)	0.96^{**} (n=16)

Figures

Figure 1: Illustrative outputs from SCIMAP. Figure 1a shows the ‘dotty plots, the correlation achieved for each simulation associated with the land cover risk weighting in that simulation. Figure 1b expresses the dotty plots in Figure 1a as two dimensional probability density functions. Darker areas indicate a high density of points within the ‘dotty plot’ and therefore a high probability that SCIMAP predictions with that land cover risk weighting will fit the observations with that correlation coefficient (assuming random sampling for all other land cover weightings). Figure 1c shows the mean and standard deviation of weightings associated with correlations at or above the values shown on the y-axis; the lines are bold where the risk weighting is significantly different from the null (no influence) case with 95 % confidence. In each plot the red horizontal lines show correlation values required for 95 % (dashes), and 99 % (solid) confidence in the correlation.

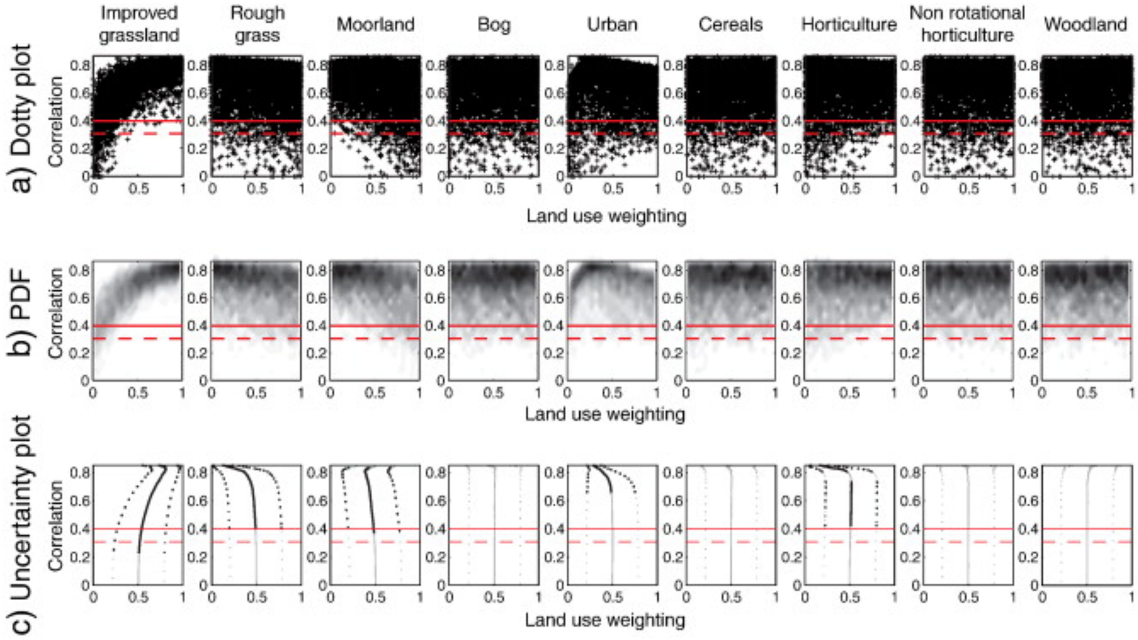


Figure 2: Catchment properties for the eleven study catchments. Figure 2a shows the location of each catchment in the UK superimposed on a topographic map. Figure 2b shows the percentage area of each catchment that is managed for agriculture (arable and improved grassland) compared to that which is urban to provide some indication of the relative impact of diffuse relative to point source pollution. The rest of the land cover classes are grouped together as 'other'. Figure 2c shows the hydrologic and agricultural setting for each catchment by plotting base flow index as an indicator of hydrologic regime against the pasture-arable index as an indicator of the dominant agricultural regime within the catchment.

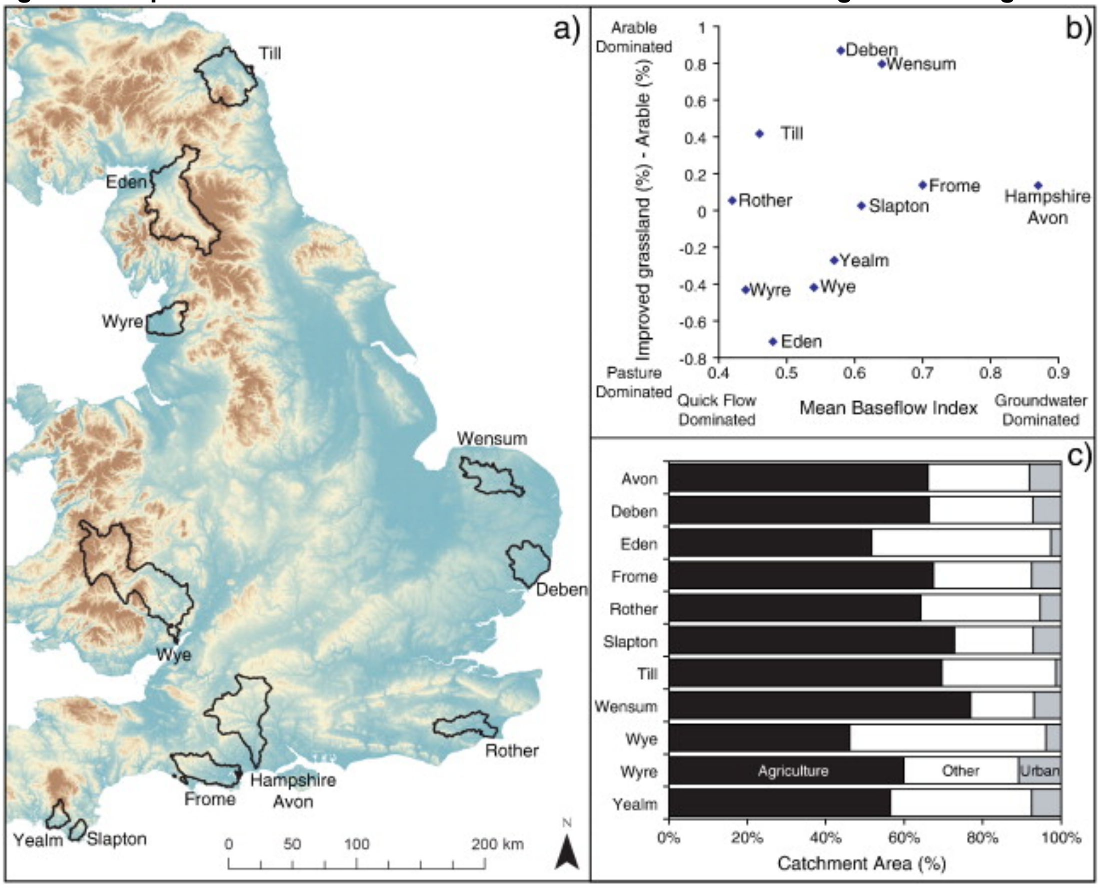


Figure 3: Model results for P and N in the (Hampshire) Avon. 3a shows the dotty plots, 3b the two-dimensional probability density functions, and 3c the risk weighting optimisation plots, showing the mean (solid line) and standard deviation (dashed line) of the weightings associated with correlations with GQA data greater than or equal to the associated correlation; the lines are bold where the risk weighting is significantly different from the null (no influence) case with 95 % confidence. Red horizontal lines show correlation values required for 95 % (dashes), and 99 % (solid) confidence in the correlation. In each section the top row shows results for P and the bottom row for N.

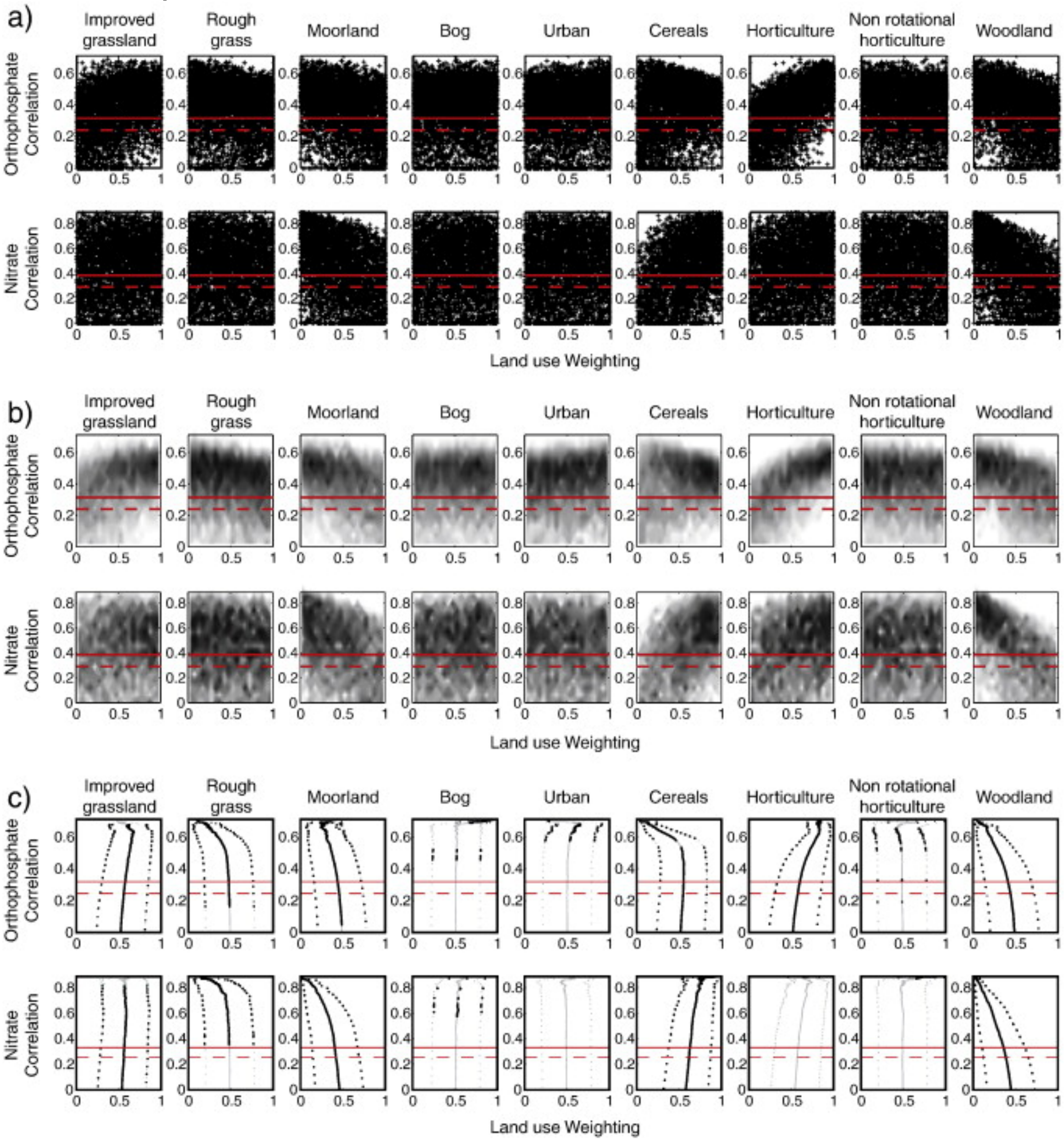


Figure 4: Scatter plots comparing the identifiability of the risk weighting assigned to each land cover with its percentage share of the catchment. Identifiability is quantified using the standard deviation of the optimum risk weighting for each land cover in each catchment. Plots are split to show results for: P (a and b) and N (c and d) for: high risk (a and c) and low risk (b and d) land covers.

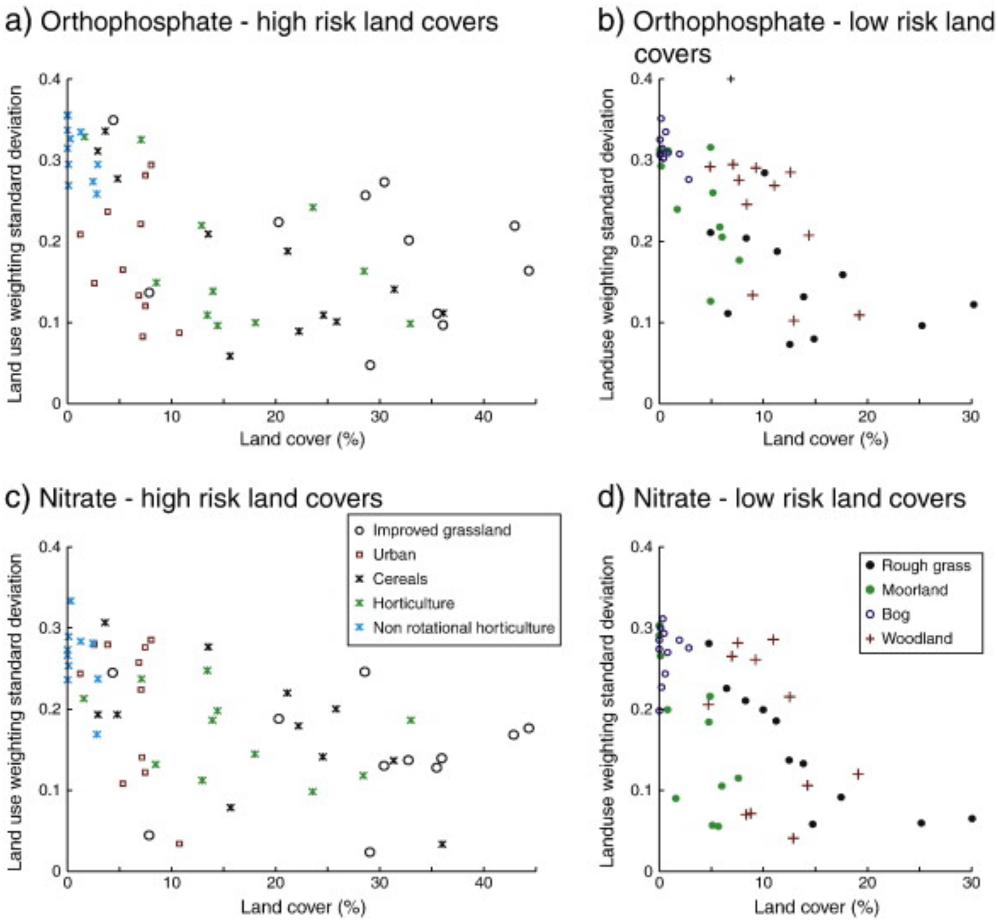
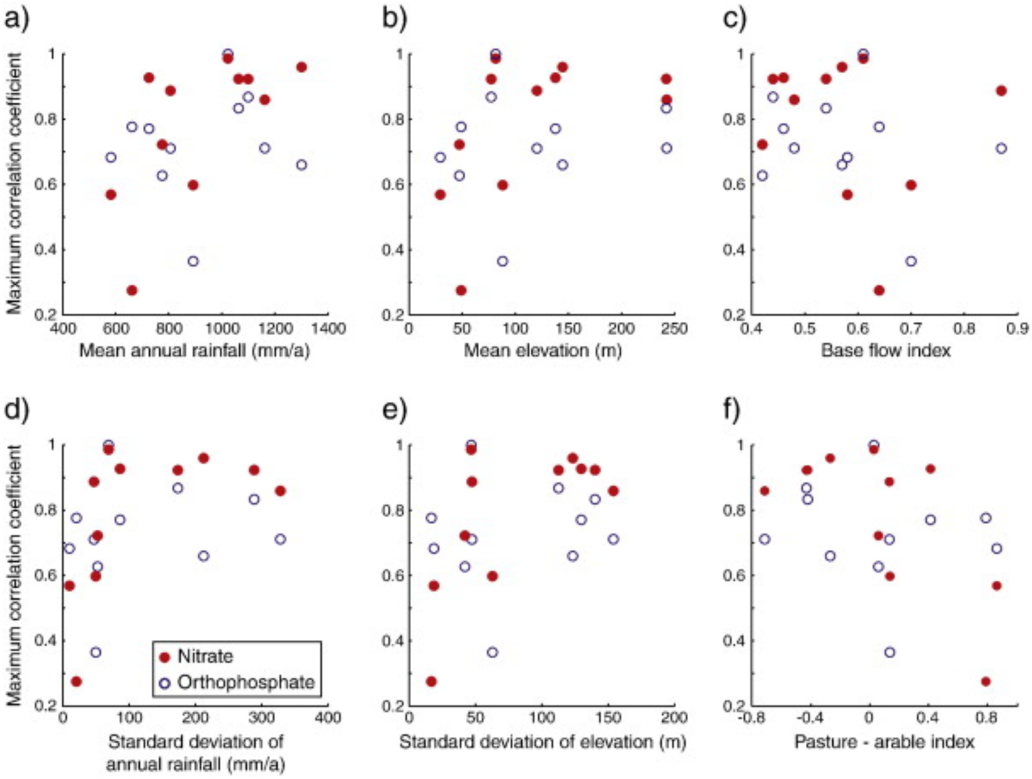


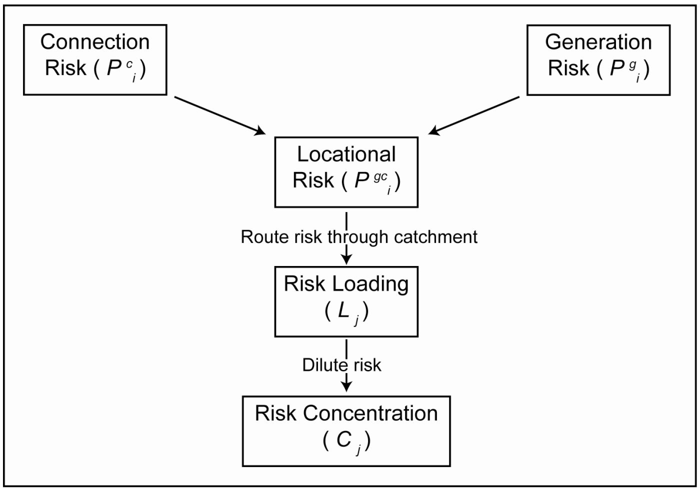
Figure 5: Scatter plots showing the relationship between SCIMAP performance and catchment characteristics. Model performance is quantified using the correlation coefficient of predicted risk against GQA observations for the optimum SCIMAP run. The catchment characteristics are quantified through a set of metrics to represent mean and variability in annual rainfall (a and d); mean and variability in elevation (b and e); hydrological regime using the baseflow index (c) and catchment land use from the pasture arable index (f).



Appendix 1: SCIMAP Model Structure

Formulation of the model requires: 1) determination of the generation risk (p^g_i); 2) determination of the delivery index, or connection probability (p^c_i) for that entrained material; 3) convolution of (1) and (2) to get the locational risk (p^{gc}_i); 4) routing of the locational risk to determine a risk loading (L_j); and 5) transformation of the risk loading to a risk concentration (C_j). An overview of the processing steps for the generation of the risk map is shown in Figure 6.

Figure 6: SCIMAP processing steps



Generation Risk

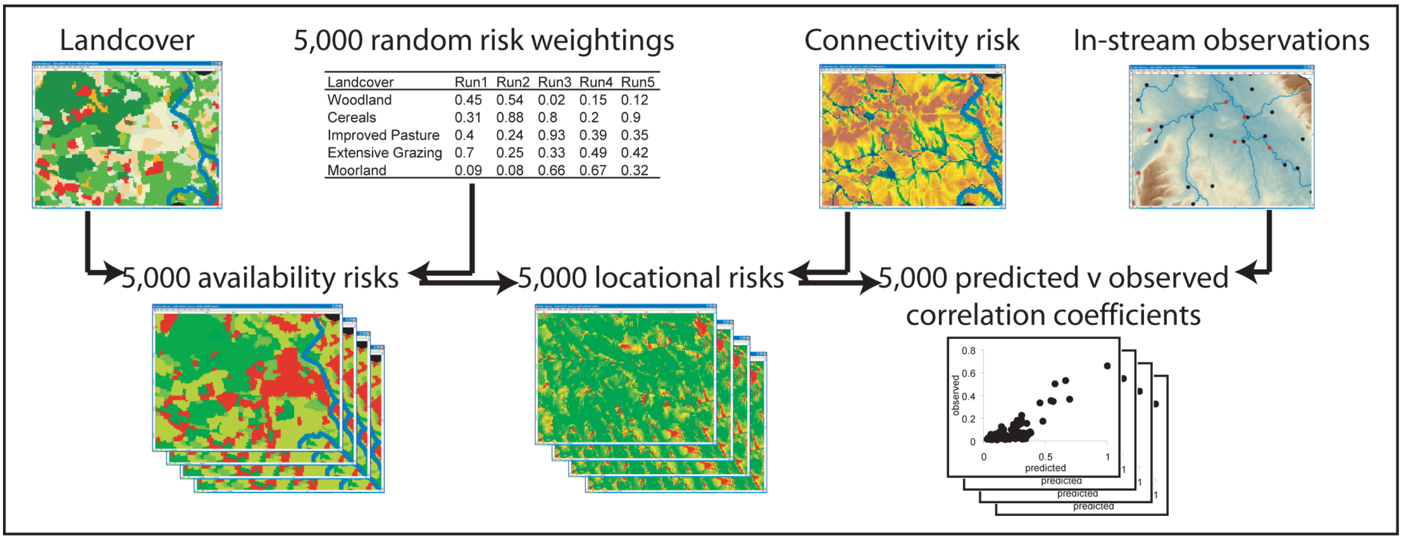
Generation risk (p^g_i) is defined as the likelihood that a location (i) in the catchment can *generate* (g) risk. Our treatment of generation risk depends on whether the generation requires physical entrainment. We assume that generation risk (p^g_i) for P and N does not require physical entrainment, i.e. that these nutrients can be dissolved in water, as a result the generation risk is solely a factor of the availability (p^e_i) of the nutrient at location i and we can equate p^g_i with p^e_i.

Equation 1

$$p_i^g = p_i^e$$

Availability (p^e_i) can be predicted using an inverse approach (Figure 7). We approach the inverse problem of unknown land cover risk weightings by using an uncertainty analysis (see below) to identify the values of p^e_i that best reproduce the spatial structure of distributed in-stream nutrient concentrations: i.e. we make no *a priori* assumptions about the p^e_i or its relationship with land cover. Instead we are matching SCIMAP predictions to in-stream nutrient observations in order to predict generation risk. There are two potential methods of achieving this: 1) an optimisation procedure based upon perturbation of the land use risk weightings to identify optimal set of risk values; or 2) a likelihood estimation procedure (e.g. Beven and Binley, 1992) in which we identify the range of plausible land use risk weighting values. Both of these methods are with respect to independent validation data. We chose the latter as we were interested in the extent to which the delivery index treatment, coupled with the risk accumulation and dilution, yielded values that were logical with respect to what we know about the relationship between landuse and nutrient availability. We run 5000 model simulations, randomly selecting values in the range $0 \leq p^e_i \leq 1$ for each land cover for each simulation, i.e. with no *a priori* likelihood of any one land cover having a particular value of p^e_i. We determine an objective function to assess model performance appropriate to the nature of the validation data available for each simulation. In this case the selected objective function is the correlation coefficient from the relationship between predicted risk and observed nutrient concentrations.

Figure 7: summary of the inverse modelling methodology



Connection Risk

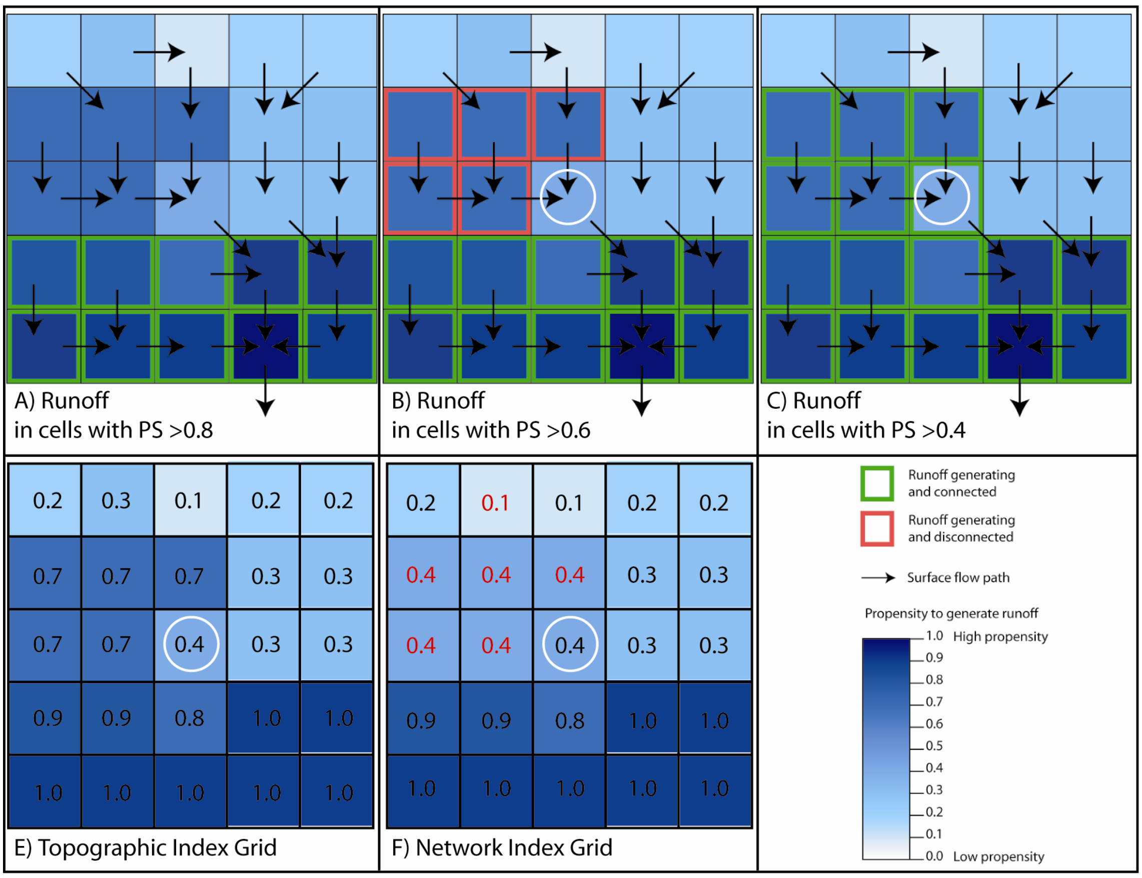
Our treatment of delivery or connection risk (p^c_i) has three primary assumptions. First, that rapid lateral surface or shallow subsurface flow is the dominant pathway for N and P delivery to the river network. The connectivity index represents disconnection associated with these processes provided that disconnection is controlled by a spatial distribution of soil moisture related to the condition where bedrock topography and surface topography have the same morphology. This is likely to be most valid for P, which is mostly sediment-bound (Walling et al., 1997; Withers et al., 1999), but also valid for some elements of the lateral flux of N, which can be transported in solution. Second, that the frequency and length of connected periods for each point in the landscape will be spatially structured, leading to a variable connection strength across the catchment. If we can find a reliable description of this spatial structure then we can use it to determine the likelihood that generated material is delivered to the drainage network. Third, that the topographic wetness index (Kirkby, 1975) can be used to describe propensity to saturation and therefore the balance between lateral flux and vertical flux at a point. Flow paths (from source to receiving water) where the soil columns are generally wetter *throughout* the flow path, are more likely to be able to flux water, and hence material, laterally (Lane *et al.*, 2009). If we can identify the point along the flow path where flux is most likely to be vertical, and quantify the extent to which that is the case, we have a measure of the likelihood of disconnectedness, the inverse of which is the propensity to connect. Here, we use the network index (Lane *et al.*, 2004*)* to determine this attribute: this is the lowest value of the topographic index along the dominant flow path between a location in the catchment and the river network. The topographic wetness index (TI) expresses the propensity to saturation as the ratio of the upslope area per unit contour length (A) draining through a point in the landscape and the tangent of the local slope (β), the latter assumed to represent the hydraulic gradient.

Equation 2

$$TI = \ln\left(\frac{A}{\tan\beta}\right)$$

Locations with a low value of the network index are assumed to have a particularly dry cell along their flow path (Lane *et al.*, 2004*)*, and hence are less likely to hydrologically connect, whether through shallow subsurface flow or surface flow. We map the network index onto the probability of connection (p^c_i) or delivery index using a distribution approach, scaling the network index between the 5th and 95th percentiles and assigning risk values of zero and one at either end of this distribution.

Figure 8: Schematic illustrating the Network index: Boxes A-C illustrate an example of cells on a hillslope generating runoff during a rainstorm. Early in the storm cells near the channel with a high propensity to saturation begin to generate runoff, these are connected to the river (green); later as more rain falls and the catchment becomes wetter a patch with a slightly lower propensity to saturation begins to generate runoff but these cells are not connected to the channel (red) because there is no continuous flow path of runoff generating cells connecting them with the channel. Finally the (white ringed) cell with a still lower propensity to saturation begins to generate runoff and at this point all the cells upslope of it that are generating runoff become connected to the channel (green). Boxes E and F show the differences between propensity to saturation as defined by the topographic index (E) and propensity to connection as defined by the network index (F). Note the red values in F which highlight cells where these two values differ, and the white ring which highlights the cell controlling connectivity in this case.



Lane *et al.* (2009) compared the information on the spatial patterns of hydrological connectivity revealed by continuous simulation using a physically based, distributed hydrological model with the network index. They found that significant spatial variability in both the propensity to connection within a time period, as well as the duration of that connectivity, can be explained using the network index. Although specifically formulated for surface overland flow, the analysis ought to apply equally for shallow subsurface flows and fluxes of material in terms of vertical versus lateral fluxes. They found that, locations with a higher Network Index are connected for longer, and the spatial signal of topographically induced wetness results in partial control of the dynamics of surface overland flow connectivity and potentially delivery.

Locational risk

We combine the generation and delivery risks to determine the locational risk of delivery of generated material to the drainage network ($p^{gc}{}_i$):

Equation 3

$$p_i^{gc} = p_i^g \cdot p_i^c$$

Routing, Accumulating and Dilution of locational risk

We route and accumulate the locational risk under the assumption that this is driven by the topographically-driven accumulating area: i.e. the risk at a point is the sum of all locational risks upstream of that point. This leads to the risk loading to a point in the drainage network (L_j) with j upslope contributing cells that will increase monotonically with distance down through the drainage network:

Equation 4

$$L_j = \sum_{i=1}^{j} p_i^g \cdot p_i^c$$

The risk loading takes no account of: 1) the propensity for dilution, where a high loading from a small upstream contributing area will have a more serious environmental effect than a high loading from a high upstream contributing area (e.g. Figure 2); or 2) loss of risk (e.g. due to deposition or chemical transformation). In this report, we assume that (2) is negligible. However, dilution is a critical property of drainage networks. The simplest way to deal with dilution is to scale the loading by the upslope contributing area to give a risk loading per unit area, akin to a concentration (C_j);

Equation 5

$$C_j = \frac{\sum_{i=1}^{j} p_i^g \cdot p_i^c}{\sum_{i=1}^{j} a_i \cdot r_i}$$

where: a_i is the cell size and r_i is the rainfall weighting factor. This equation takes account of possible rainfall variations between sub catchments and the propensity for such variation will increase with basin size. This is represented by weighting upslope contributing areas by the amount of upstream contributed precipitation, using temporal averages that reflect the time-integration of the study. However, such an analysis is complicated by the fact that spatial variability in precipitation should also result in spatial variability in connectivity. Hence, the predicted relative long term average wetness also utilises the rainfall weighting factor.

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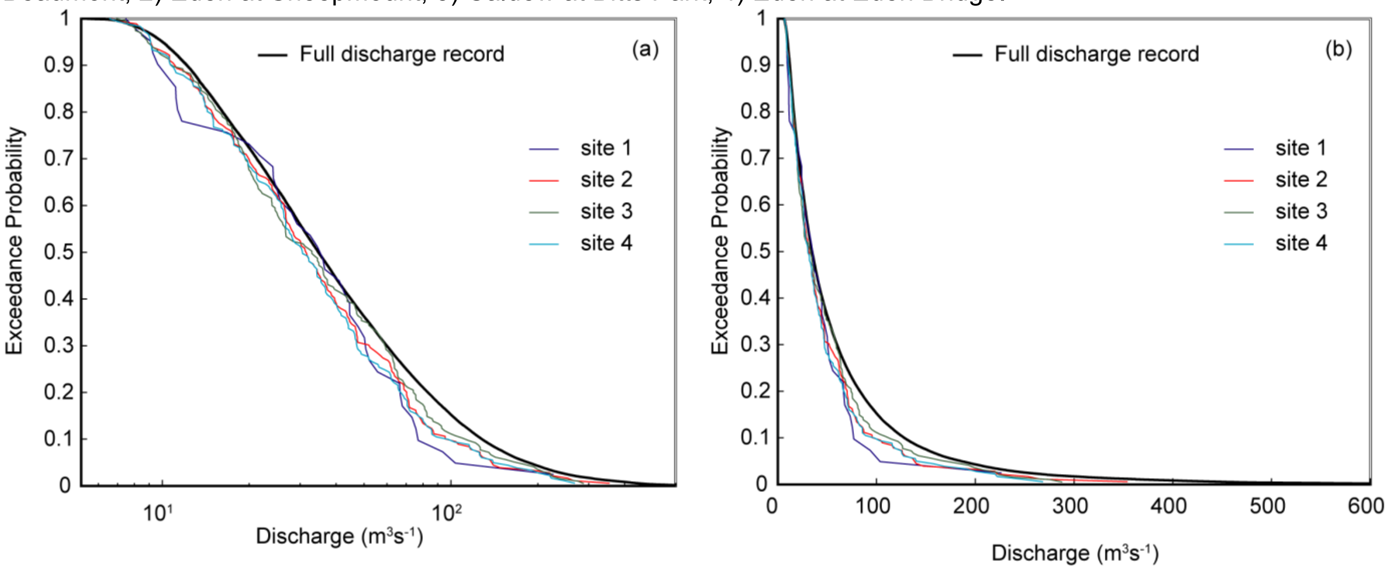
Appendix 3: Testing suitability of simple means to integrate monthly N and P concentrations

In this paper we use the mean of a set of concentration measurements as a time integrated measure of relative risk. Our approach aims to make use of the GQA data which are imperfect but are the best spatially distributed observations available. This approach has been used for the same environment agency datasets at other UK sites (Davies and Neal, 2007; Rothwell et al., 2010). It assumes that the monthly samples an adequately representative sample of instream nutrient concentrations to give the relative magnitudes of the mean concentrations. We tested this assumption for the Eden catchment where we had both concentration and discharge data. Our GQA data record was 15 years long from 1990 to 2005. The mean number of observations per site was 155 with a standard deviation of 38 observations. We compared the distribution of flows over which concentration samples were collected with the full distribution of flows to check for a low flow bias to our samples.

Figure 1 shows the results from our comparison of exceedance probabilities for only times that concentration samples were collected and for the full record. The exceedance probability curves (Figure 1) suggest that concentration measurements were taken across a reasonable distribution of high and low flows. On the basis of our results we suggest that the GQA data contain a large enough number of samples over a long enough period to sample the range of flow conditions so that they do not suffer a strong low flow bias. Flow weighting might improve the concentration estimates slightly but the number of sites that could then be used would be limited by the availability of discharge data, which is even more limited than concentration data in terms of spatial coverage. Since we are interested in the mean concentrations as a time integrated measure of relative risk slight improvements in mean concentrations are not worthwhile if they require significant reductions in spatial coverage.

Figure 9: Exceedance probabilities for discharge (on (a) a logarithmic scale and (b) a linear scale) for the Eden at Sheepmount and for the discharges at which orthophosphate samples were collected from sites 1) Eden at Beaumont; 2) Eden at Sheepmount; 3) Caldew at Bitts Park; 4) Eden at Eden Bridge.



Appendix 3: Identifying the number of simulations required to sample the parameter space

We have tested the influence of the number of simulations for the Hampshire Avon catchment by calculating the optimum risk weightings using from 50 to 5000 simulations at 50 simulation increments. Our results suggest that the means and standard deviations become stable at around 2500 simulations and optimized means do not change their relative ranking beyond 3000 simulations (Figure 10). The optimized mean weights vary by <0.08 (or <27% of their average standard deviation) between 4000 and 5000 simulations and standard deviations vary by <0.04 (or <20% of their average value). There is a considerable reduction in this variability if considering only the high or low risks (significantly different from the null case at 90% confidence). For these land covers the optimized means vary by <0.04 (or <16% of their average standard deviation) between 4000 and 5000 simulations. These results suggested that 3000 simulations might be adequate to produce stable estimates and that 5000 simulations would include considerable redundancy. One of the reasons for the relatively low number of simulations required to identify stable values may be the distribution of land covers in our catchments. In each catchment some land covers have little or no coverage, reducing the number of dimensions for the parameter space and therefore the number of required simulations.

Figure 10: optimised risk weightings calculated based on different numbers of simulations for the Hampshire Avon.