The influence of landscape spatial configuration on nitrogen and phosphorus exports in agricultural catchments

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Abstract:	Nitrogen (N) and phosphorus (P) exports from rural landscapes can cause eutrophication of inland and coastal waters. Few studies have investigated the influence of the spatial configuration of nutrient sources – i.e. the spatial arrangement of agricultural fields in headwater catchments – on N and P exports. This study aimed to 1) assess the influence of the spatial configuration of nutrient sources on nitrate (NO_3^-) and total phosphorus (TP) exports at the catchment scale, and 2) investigate how relationships between landscape composition and landscape configuration vary depending on catchment size. We analysed NO_3^- and TP in 19 headwaters (1-14 km², Western France) every two weeks for 17 months. The headwater catchments had similar soil types, climate, and farming systems but differed in landscape composition (percentage agricultural land-use) and spatial configuration. We developed a landscape configuration index (LCI) describing the spatial configuration of nutrient sources as a function of their hydrological distance to streams and flow accumulation zones. We calibrated the LCI's two parameters to maximise the rank correlation with median concentrations of TP and NO_3^-. We found that NO_3^- exports were controlled by landscape composition, whereas TP exports were controlled by landscape configuration. For a given landscape composition, landscape spatial configuration was heterogeneous at small scales (<10 km2) but homogeneous at larger scales (>50 km2).

	The spatial configuration of nutrient sources influences TP but not NO3- exports. An ideal placement of mitigation options to limit TP export should consider both the hydrological distance to streams and flow accumulation zones.
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Title

The influence of landscape spatial configuration on nitrogen and phosphorus exports in agricultural catchments

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Abstract

Context

Nitrogen (N) and phosphorus (P) exports from rural landscapes can cause eutrophication of inland and coastal waters. Few studies have investigated the influence of the spatial configuration of nutrient sources – i.e. the spatial arrangement of agricultural fields in headwater catchments – on N and P exports.

Objectives

This study aimed to 1) assess the influence of the spatial configuration of nutrient sources on nitrate (NO_3^-) and total phosphorus (TP) exports at the catchment scale, and 2) investigate

how relationships between landscape composition and landscape configuration vary depending on catchment size.

Methods

We analysed NO_3^- and TP in 19 headwaters (1-14 km², Western France) every two weeks for 17 months. The headwater catchments had similar soil types, climate, and farming systems but differed in landscape composition (percentage agricultural land-use) and spatial configuration. We developed a landscape configuration index (LCI) describing the spatial configuration of nutrient sources as a function of their hydrological distance to streams and flow accumulation zones. We calibrated the LCI's two parameters to maximise the rank correlation with median concentrations of TP and NO_3^-.

Results

We found that NO_3^- exports were controlled by landscape composition, whereas TP exports were controlled by landscape configuration. For a given landscape composition, landscape spatial configuration was highly heterogeneous at small scales (<10 km²) but became homogeneous at larger scales (>50 km²).

Conclusions

The spatial configuration of nutrient sources influences TP but not NO_3^- exports. An ideal placement of mitigation options to limit TP export should consider both the hydrological distance to streams and flow accumulation zones

Keywords

Eutrophication; landscape pattern; landscape organisation; landscape composition; heterogeneity; scaling

1. Introduction

Excessive loads of nitrogen (N) and phosphorus (P) cause the eutrophication of marine and freshwater ecosystems (Dodds and Smith 2016; Le Moal et al. 2018), which threatens biodiversity and human activities (Steffen et al. 2015). In streams and rivers that drain agricultural landscapes, N and P originate mainly from fertilised agricultural fields. Water quality mitigation measures often result in reduced yields or increased production costs; hence, they decrease profitability for farmers (Withers et al. 2014). It is therefore necessary to locate mitigation options such as set-aside areas and buffer zones where they will be most effective (Cole et al. 2020). Mechanistic models (Casal et al. 2019; McDowell et al. 2014) and methods based on statistical analysis of landscape properties (Doody et al. 2012; Hashemi et al. 2018) have been used to optimise locations of mitigation practices.

Most N and P enter the hydrographic network in headwater streams (Dodds and Oakes 2008), which represent 90% of global stream length (Downing 2012) but are rarely monitored, except for research purposes (Bishop et al. 2008). At this scale, nutrient exports vary greatly in space (Abbott et al. 2018; Temnerud and Bishop 2005). While in large (>100 km^2) catchments, both N and P exports can be statistically related to proxies of agricultural pressure intensity, such as percentage of agricultural land-use or agricultural surplus (Dupas et al. 2015; Goyette et al. 2018), these relationships break down at the scale of headwater catchments (<20 km^2) (Bol et al. 2018; Burt and Pinay 2005). The reason why the relationship between landscape composition and nutrient exports is scale-dependent is unclear. Identifying the factor(s) responsible for this loss of relationship, and the characteristic spatial scale at which it occurs, would help guide water quality mitigation measures at the catchment scale. In this article, we explore the hypothesis that the spatial configuration of nutrient sources – i.e. the spatial arrangement/distribution/organisation of agricultural fields, hereafter called “landscape configuration”, –influences N and P exports at the headwater catchment scale. For a given region, landscape configuration can be highly heterogeneous among headwater catchments but homogeneous among larger catchments (Abbott et al. 2018; Temnerud and Bishop 2005). If verified, these two hypotheses could explain the breakdown of the relationship between agricultural pressure intensity and nutrient exports at the headwater catchment scale.

Several authors have reviewed the influence of landscape spatial configuration on nutrient loads at multiple scales (Lintern et al. 2018; Uuemaa et al. 2007). Metrics used in landscape ecology are based on the area/density/edge, shape, isolation, interspersion, and connectivity of patches, and they have been applied to predict water quality parameters at the catchment scale (Liu et al. 2020; Shi et al. 2013; Xiao et al. 2016). However, these approaches often rely on regressions of several landscape metrics and water quality parameters, which risks

over-fitting certain relationships and/or spurious correlations. Landscape spatial configuration can also be understood as the overlap and proximity of features such as streams, depressions, and flow accumulation areas to land-use patches (e.g. crops, forest fragments, urban fabric). In line with this concept, Peterson et al. (2011)) related parameters of stream ecological conditions to multiple spatially explicit landscape "topological" metrics and found that these spatially explicit methods clearly had more predictive power than landscape composition metrics. Staponites et al. (2019)), using similar metrics, suggested that the spatial organisation of nutrient sources (i.e. landscape configuration) influenced the transfer of reactive or labile water quality parameters (e.g. TP, NO_2^- and PO_4^{3-}), whereas percentages of land use (i.e. landscape composition) have more predictive power for more temporally stable water quality parameters (e.g. Ca^{2+}, NO_3^-, conductivity). These results are consistent with current knowledge on transfer pathways of nutrients in catchments, with deeper flowpaths for N species than for P species (Strohmenger et al. 2020), and explains why landscape features had less influence on NO_3^- than P (Thomas and Abbott 2018). These landscape metrics, however, are relatively rigid, as topological influences (e.g. hydraulic distance to stream, surface flow accumulation, slope), whether considered in the metrics or not, cannot be weighted. In addition, certain topological features vary over a much wider range of values than others, which may obscure other features when no weighting coefficient is included in the landscape metric. For example, flow accumulation (which can have large values) can overshadow the influence of slope (which varies over a narrow range of values) in an index that considers both without weighting them.

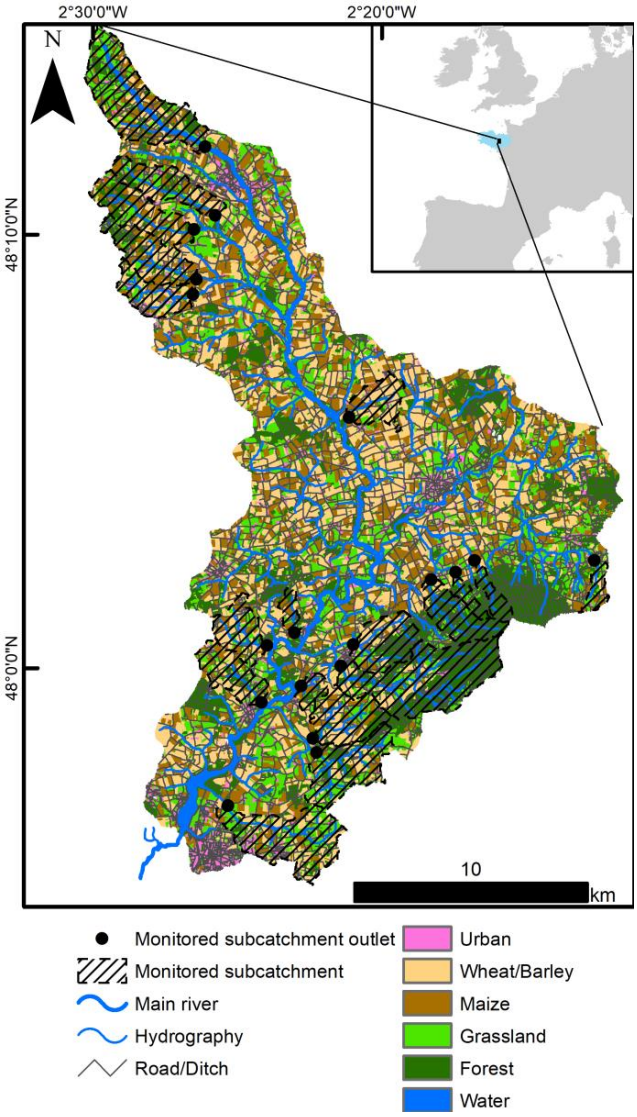
The idea that landscape configuration influences N and P loads is the basis for the concept of critical source area (CSA), i.e. the idea that a small percentage of the agricultural area (e.g. <20%) contributes disproportionately to the nutrients transferred to streams (e.g. >80%). CSAs are defined as the intersection of nutrient sources and hydrologically sensitive areas (Gburek and Sharpley 1998; Pionke et al. 2000). Initially defined at the sub-field scale, the concept of CSA was extended to larger scales; entire fields or subcatchments can also be CSAs (Buchanan et al. 2013b; Page et al. 2005; Reaney et al. 2019; Sharpley et al. 2011; Srinivasan and McDowell 2009). Substantial uncertainties remain, however, in their delineation at all scales (Doody et al. 2012). Validation at the sub-field scale can be based on observing erosion marks (Reaney et al. 2019) or tracers (Collins et al. 2012). However, studies that validate CSA delineation based on N or P concentrations in streams and rivers are rare and are based on only a few hydrologically contrasting headwater catchments (McDowell and Srinivasan 2009; Shore et al. 2014; Thomas et al. 2016a) or larger catchments for which land-use composition is already a good predictor (Giri et al. 2018).

To address the limits of “expert-based” delineation of CSAs and the rigidity of spatially explicit landscape metrics, we developed a stochastic, data-driven approach based on 30 synoptic samplings of 19 agricultural headwater catchments to answer two questions: i) Does landscape configuration influence N and P exports?; and ii) Does the location of CSAs depend on the hydrological distance of the nutrient source to the hydrological network, their overlap with flow accumulation areas, or both? We then investigated why some relationships between water quality parameters and landscape composition metrics break down below a certain catchment size. For this, we studied how the relationship between landscape composition and configuration varied as a function of catchment size in 500 randomly selected subcatchments in the study area.

2. Methods

2.1. Study site

The Yvel catchment is a 375 km^2 agricultural catchment of Strahler order 5 in Brittany, western France (Fig. 1). The Yvel River is the main tributary of a three million m^3 water reservoir that has been subjected to cyanobacteria blooms since the 1970s (ODEM 2012), for which N and P are deemed responsible (Shatwell and Köhler 2018).



143

144 **Fig. 1** Monitored subcatchments, land use in 2018, hydrography and roads in the Yvel
145 catchment. The inset shows the location of Brittany, France, in western Europe

146 The climate is temperate oceanic with mean annual precipitation (1998-2017) of 777 mm
147 (sd=132 mm), mean annual temperature of 11.7°C (sd=0.5°C), and mean annual runoff of
148 254 mm (sd=143 mm). The Yvel River's discharge is monitored for 300 km^2 of its 375 km^2,
149 and monthly mean discharge varies from 157 $L.s^{-1}$ in August to 5,600 $L.s^{-1}$ in February. The
150 hydrology is controlled by the dynamics of the shallow groundwater within unconsolidated
151 weathered material that caps impervious schist bedrock (Casquin et al. 2020). The land use
152 is dominated by arable fields (maize and winter cereals), which cover 54% of the catchment
153 (Fig. 1). Grasslands (21%, mainly leys in rotation), forests (18%), and urban areas (6%)
154 comprise the rest of the catchment area. Hedgerow density is 71 $m.ha^{-1}$. Soils in the
155 catchment are generally shallow (<100 cm), are well-drained in the upland part of the
156 hillslope, and are often hydromorphic in valley bottoms. The elevation varies from 33-297 m
157 above sea level. The centre of the catchment is the flattest area (most slopes <5%), with long

158 and regular hillslopes. In the north and south, the relief is more rugged, with shorter and
159 steeper slopes. The southeast portion of the watershed of the catchment is forested and has
160 the steepest slopes (5-15%).

161 **2.2. Subcatchment monitoring**

162 The monitoring strategy consisted of repeated synoptic sampling of 19 subcatchments (Fig.
163 1). The 19 subcatchments were selected based on Strahler order (1-2), size (0.8-12.6 km^2,
164 mean=5.1 km^2), absence of a wastewater treatment plant, and accessibility. Their
165 percentage of agricultural land-use ranged from 17-94% (mean=74%), with mean slopes
166 ranging from 2.7-6.6% (mean=4.9%). Together, these 19 subcatchments covered 28% of the
167 Yvel catchment's area. All 19 monitoring points were sampled approximately every two
168 weeks from April 2018 to July 2019 (30 dates in total). Samples were filtered *in situ*
169 immediately after sampling with cellulose acetate filters of 0.20 µm pore size for nitrate (NO_3^-)
170 analysis. All filters were rinsed in the laboratory with 20 ml of deionised water before use. An
171 unfiltered water sample was also collected to analyse TP. The samples were transported to
172 the laboratory in a cool box and then refrigerated at 4°C until analysis. TP was analysed
173 within 48 h of sampling, while NO_3^- was analysed within one week. TP was determined
174 colorimetrically via reaction with ammonium molybdate (Murphy and Riley 1962) after
175 digesting the samples in acidic potassium persulfate. The precision of TP measurements
176 was ±13 $\mu g.L^{-1}$, while that of NO_3^- concentrations, analysed by ionic chromatography
177 (Dionex, DX120), was ±4%. Hydrochemical data are available at
178 https://www.hydroshare.org/resource/7c7d7f6dd1f14450883ae1c243c3c28f/ (Dupas 2020).

179 **2.3. Landscape configuration index**

180 We developed the landscape configuration index (LCI) (Eq. 1) to test the hypothesis that the
181 hydrological proximity of agricultural areas to watercourses and their overlap with flow
182 accumulation areas influence nutrient exports at the headwater catchment scale. For each
183 monitored subcatchment, the index was calculated as follows:

184 $$LCI(a,b) = \frac{\frac{1}{n} \times \sum_{i=1}^{n} LU_i \times FAcc_i^a \times FLS_i^{-b}}{\frac{1}{N} \times \sum_{i=1}^{N} LU_i \times FAcc_i^a \times FLS_i^{-b}} \quad (Eq.\ 1)$$

185 where a and b are calibrated parameters, i=1..n are the pixels of a subcatchment; i=1..N are
186 the pixels of the entire catchment, $FAcc_i$ is the flow accumulation on pixel i (m^2), FLS_i (m) is
187 the distance along the surface flow line from pixel i to the stream/ditch, and LU_i equals 1 if
188 pixel i is a source of nutrients (i.e. an agricultural area), otherwise 0.

Fig. 2 details the calculations for two subcatchments with similar land-use composition, but whose LCI numerator varies by a factor of 2 when parameters (a, b) = (1.5, 1).

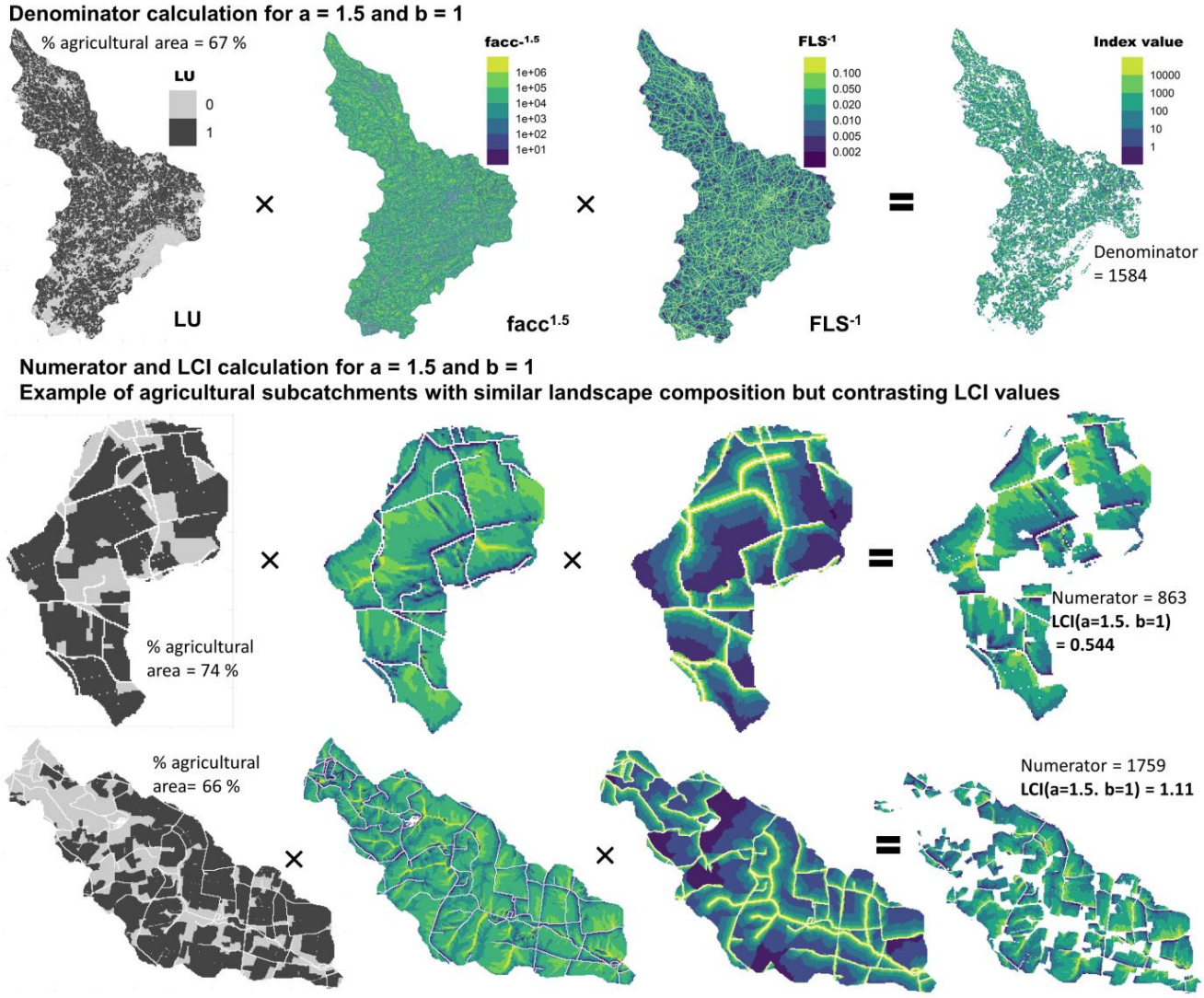


Fig. 2 Steps used to calculate the index given a=1.5 and b=1.0 for 2 of the 19 monitored subcatchments. Note the logarithmic colour scale

The denominator is a normalisation factor that corresponds to the mean value of the numerator for the entire Yvel catchment. When LCI > 1, a subcatchment's nutrient sources are located predominantly in flow accumulation zones and/or near streams. For large values of a or b, the LCI assigns high weights to a small percentage of the area. We restricted the ranges of parameters a and b so that a few pixels with the highest Facc and lowest FLS would not control the values of LCI. We explored the parameter space for pairs of (a, b) for which the highest 5% of cumulative LCI in the landscape did not exceed 95% of the overall cumulative LCI. When b=0, we varied a within the range 0-2, whereas when a=0, we varied b within the range 0-4.

By construction, when a and b equal 0 for a subcatchment, its LCI equals its percentage of agricultural land-use (i.e. landscape composition) divided by the percentage of agricultural land-use in the entire Yvel catchment. For other values of a and b, the LCI indicates the landscape configuration. High values of parameter a increase the weight of pixels in flow accumulation zones in the LCI, while high values of parameter b increase the weight of pixels in near-stream zones.

2.4. Spatial data sources and pre-processing

2.4.1. Hydrographic network

The hydrographic network consisted of both the “natural” stream network and the ditch network because 1) it is often difficult to distinguish a ditch from a rectified stream and 2) our synoptic sampling verified that most of the ditches were deep (> 1 m) and flowed in winter (high-flow season). Ditch networks are a shortcut between agricultural areas and the "natural" river network (Ahiablame et al. 2011; Buchanan et al. 2013a). Moreover, evidence suggests that ditches act as 1st order streams when considering nutrient spiralling and can retain and remobilise N and P (Dunne et al. 2007; Hill and Robinson 2012; Smith 2009). Thus, we considered them part of the hydrographic network. We used the road network as a proxy for the ditch network, as we observed that ditches bordered all roads in the study area. Streams (permanent and intermittent) and roads were extracted from vector data (accuracy of ca. 1.5 m) provided by the *Institut National de l'Information Géographique et Forestière* (IGN) at 1:25,000 scale. We converted these vector data to raster format and aligned them with the Digital Elevation Model (DEM) for later analysis.

2.4.2. Digital Elevation Model

The DEM, with a native resolution of 5 m (IGN 2018), was resampled to 10 m using cubic splines in ArcGIS 10.6. Filling was used to remove the depressions on hillslopes (Planchon and Darboux 2002), and the value "NA" was assigned to the pixels corresponding to roads and streams. We calculated flow accumulation and the hydrological distances to streams (i.e. following the surface flow paths) using the multiple flow direction algorithm (Qin et al. 2007). We chose this algorithm for its ability to generate realistic flow accumulation maps, unlike the D8 algorithm (O'Callaghan and Mark 1984).

2.4.3. Nutrient sources

We extracted the agricultural area from the national Land Parcel Identification System (Levavasseur et al. 2016). The data are provided as a vector dataset at the 1:5000 scale for each year since 2010 and contain field boundaries and a code that identifies the crop type. We used the 2018 dataset and verified the spatial accuracy of the agricultural area boundaries based on 50 cm orthophotos and the hydrography. We then rasterized this

dataset aligned with the DEM. We assigned the value 1 to agricultural areas and 0 to non-agricultural areas. We included riparian buffer strips in agricultural areas because they are recent and have been fertilised for years, and are a well-documented legacy source of nutrients in headwater catchments (Gu et al. 2018).

2.5. Optimisation of (a, b) parameters and interpretation

For each pair (a, b) within the previously defined domain of exploration, we calculated Spearman's rank correlation (Rs) between the LCI and median NO_3^- and TP concentrations of each subcatchment (Figure 3). Since several of the subcatchments were intermittent, we calculated the median concentrations for the 22 dates (out of 30) when at least 17 of the 19 subcatchments were flowing, so as not to bias calculation of the median. We focused on ranks rather than concentrations because several studies have shown that concentration estimates had high uncertainty when calculated with low-frequency data (e.g. Cassidy and Jordan 2011; Moatar et al. 2020), while the ranks of subcatchments could be predicted with high degree of confidence, as they are stable across flow conditions (Abbott et al. 2018; Dupas et al. 2019, Gu et al., 2020 - in review). The result interpretation was twofold:

- The spatial configuration of sources was considered to have an effect if Rs for at least one pair (a, b) ≠ (0, 0) was significantly higher than that for (a, b) = (0, 0).
- Optimal values of a and b were examined to assess the relative importance of hydrological distance to streams and flow accumulation on hillslopes.

We then calculated the ratio of LCI with optimal (a, b) to LCI with (a, b) = (0, 0) for 500 randomly selected subcatchments in the Yvel catchment, and analysed how the relationship between these two metrics evolved as catchment size increased.

3. Results

3.1. Comparison of land-use composition and configuration metrics as predictors of NO_3^- and TP concentrations in headwater catchments

Landscape composition (i.e. LCI with (a, b) = (0, 0)) predicted median NO_3^- concentrations well (Rs=0.84, $p<0.001$) but not those of TP (Rs=0.33, p=0.18). Varying (a, b) did not substantially improved the prediction of median NO_3^- concentration rank; thus, landscape composition predicted NO_3^- exports well at the headwater catchment scale, and considering landscape configuration did not improve the prediction (Fig. 3A). For NO_3^-, this result refutes our first hypothesis that the spatial configuration of nutrient sources influences nutrient exports.

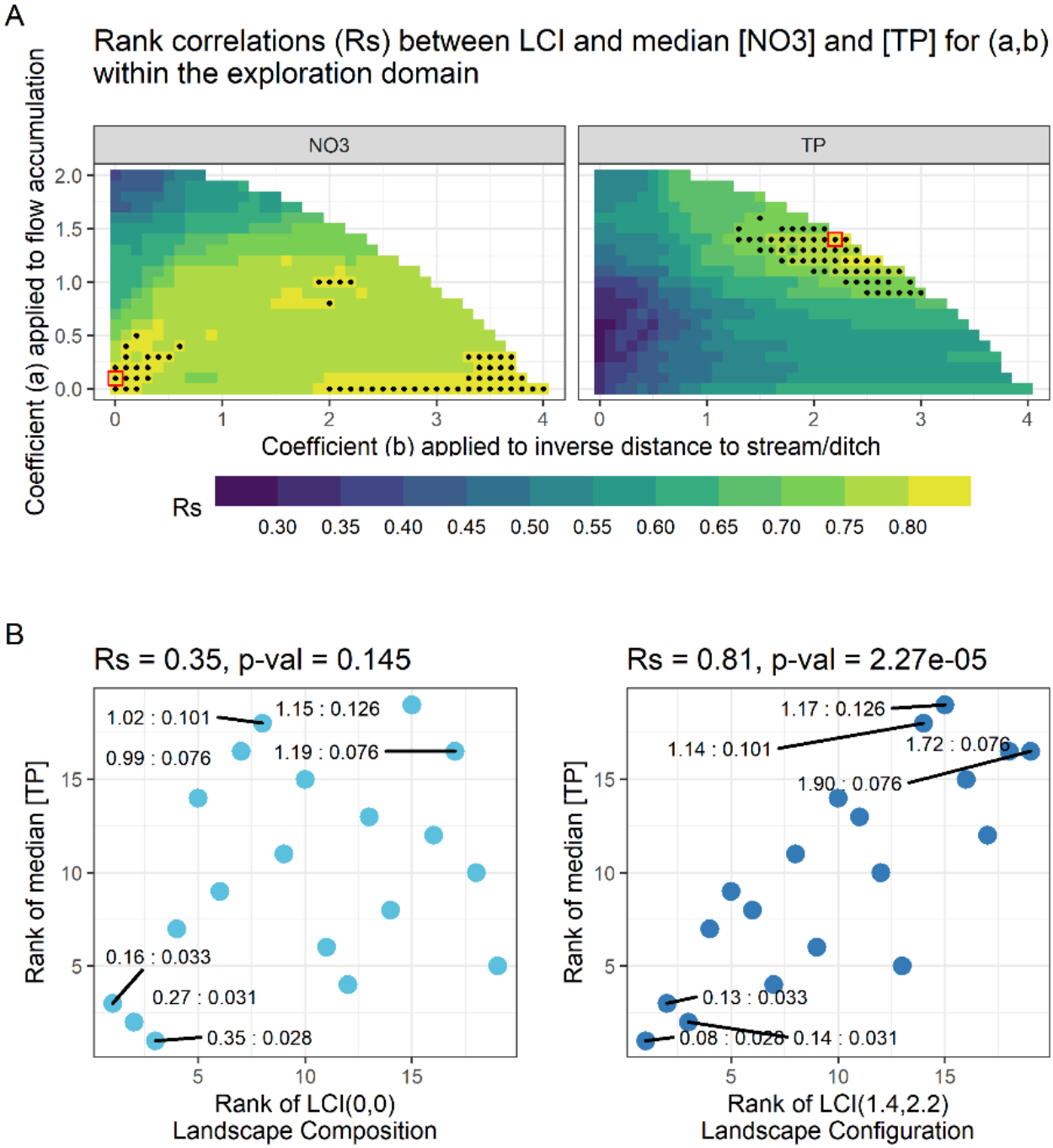


Fig. 3 A) Optimisation plan for parameters a and b used to predict median concentration of nitrate (NO_3^-) and total phosphorus (TP) in 19 headwater catchments. Rank correlations (Rs) not shown (blank) indicate (a, b) outside of the exploration domain. For each optimisation plan, black dots indicate 10% highest Rs, red square the best correlation B) Rs between median [TP] and 1) landscape composition and 2) optimised landscape configuration index (LCI). Annotations show LCI:median [TP]

279 The correlation between the percentage of agricultural land and median TP concentrations
280 ((a, b) = (0, 0)) was 0.35 ($p=0.145$) (Fig. 3B). This correlation improved as weights increased
281 for sources near streams (Rs increased as b increased) and for sources that overlapped
282 surface flow accumulation (Rs increased as a increased) (Fig. 3A). This confirms our first
283 hypothesis that landscape configuration influences P exports, and agricultural areas near
284 watercourses and that overlap surface flow accumulation areas result in larger P exports.
285 The optimum correlation was obtained for $a=1.4$ and $b=2.2$ ($Rs=0.81$, $p<0.001$), at the limit of
286 our field of exploration, which means that 5% of the area determined nearly 95% of the
287 index.

288 Using the same parameter exploration scheme, we optimised (a, b) for TP concentrations on
289 each sampling date (Fig. 4C). The LCI with $(a, b) \neq (0, 0)$ always predicted TP concentrations
290 better than landscape composition (Fig. 4A). The correlation between the optimised LCI and
291 TP concentrations differed significantly ($p<0.05$) from 0 for all but one sampling date, except
292 during the low-flow periods (Fig. 4B).

Fig. 4 Comparison of landscape composition and optimised landscape configuration index (LCI) as predictors of TP concentrations, for all sampling dates. (a) Spearman's rank correlation (Rs), (b) -log10-transformed p-values (dashed line indicates p=0.05), and (c) for optimised LCI, parameters a and b applied to flow accumulation and inverse distance to the stream/ditch (d). Relative weight of the top 5% of weighted pixels. Grey areas represent low-flow periods, when two or more sampled streams were dry

During the low-flow season (Aug-Dec 2018) and beginning of the rewetting season (Jul 2019), TP concentrations in the headwaters were not correlate with the optimised LCI ($p>>0.05$) (Fig. 4B). Note that 13 of the 19 streams were dry at the peak of the low-flow season (Fig. 1S). Outside the low-flow season, optimal values of parameters a and b were stable, varying little around a median value of 1.4 and 2.2, respectively. Therefore, the total weight of the top 5% of weighted area remained close to 95% (Fig. 4D), except during the low-flow season. The optimised LCI based on median TP concentrations, hereafter referred to as LCI-TP, appears to be a robust sensitivity index to determine TP CSAs across flow conditions.

3.2. Spatial aggregations of the LCI-TP

The data-driven optimization of the LCI parameters to predict the ranking of exports at the catchment scale yielded an index that assigned nearly all of the weights (95%) to small areas (5%). Most of these areas (Fig. 5A) were located near streams and ditches, but their width varied. The CSAs often extended ca. 100 m or more into agricultural areas, especially on long and convex hillslopes. Using LCI-TP values at the pixel level, we calculated $LCI\text{-}TP_{field}$ as the mean LCI-TP of each agricultural field. The $LCI\text{-}TP_{field}$ (Fig. 5B) revealed that most fields were weak sources of P (i.e. $LCI\text{-}TP_{field} << 1$), whereas a few fields were CSAs, as their $LCI\text{-}TP_{field}$ exceeded 10 and even 40. The associated histogram (Fig. 5C) followed a lognormal distribution, which confirms the high variability at the field scale (Q5-Q95=0.001-14.087, sd=39.05). When the LCI-TP was aggregated into 1 km^2 subcatchments ($LCI\text{-}TP_{1km2}$) (Fig. 5D), its variability decreased drastically (Fig. 5F, min-max=0.17-5.38, sd=0.49). The number of subcatchments with $LCI\text{-}TP_{1km2} > 1$ was approximately the same as the number of subcatchments with $LCI\text{-}TP_{1km2} < 1$ (Fig. 5F, median=0.92), but the distribution remained lognormal. Following the same pattern, the LCI-TP aggregated into 25 km^2 subcatchments ($LCI\text{-}TP_{25km2}$) had even lower variability (min-max=0.71-1.36): at this scale the distribution was symmetrical (apparently normal), and no subcatchment could be considered a CSA. The information at the subfield scale (Fig. 5A) and field scale (Fig. 5B), which is relevant for farmers and catchment managers, was generated for the entire study area (https://antoine-csqn.github.io/YV1.html).

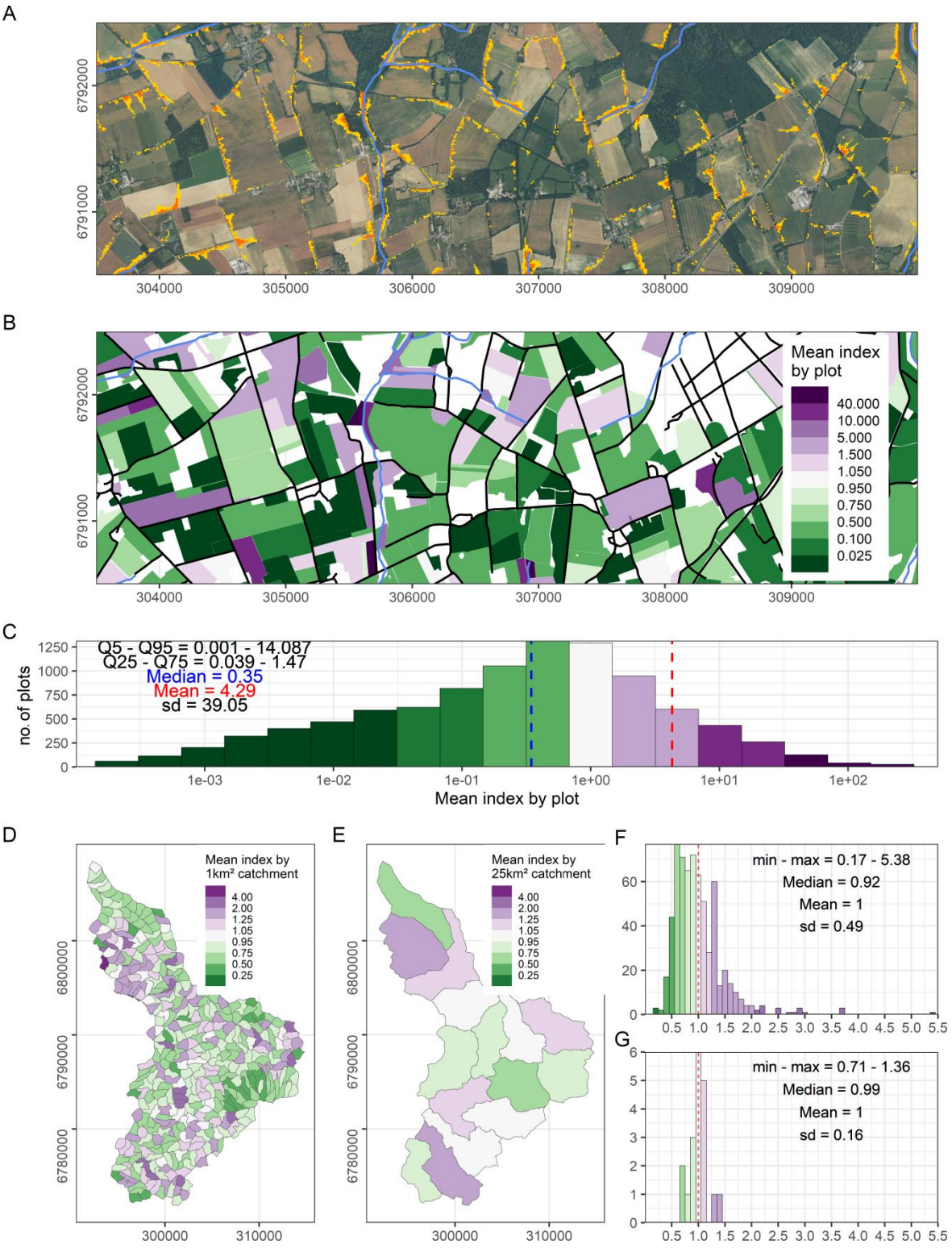


Fig. 5 A) Excerpt of the study area, ca. 6 km × 2 km: top 5% (yellow) and 1% (red) of weighted pixels according to LCI-TP, B) Mean LCI-TP aggregated at the field scale (LCI-TP $_{field}$), and C) histogram of associated values (note the log scale on the x-axis). D) LCI-TP for 1 km^2 subcatchments (LCI-TP$_{1km2}$) and E) 25 km^2 subcatchments (LCI-TP$_{25km2}$) and associated histograms (F and G, respectively)

3.3. Homogenisation of the LCI-TP with increasing catchment size

We delineated 500 subcatchments within the study area based on 500 points randomly generated over the hydrographic network. For each subcatchment, we calculated the LCI-TP (i.e. the LCI for a=1.4 and b=2.2), the landscape composition index (i.e. the LCI for a=b=0), and their ratio, which we examined as a function of subcatchment area (Fig. 6).

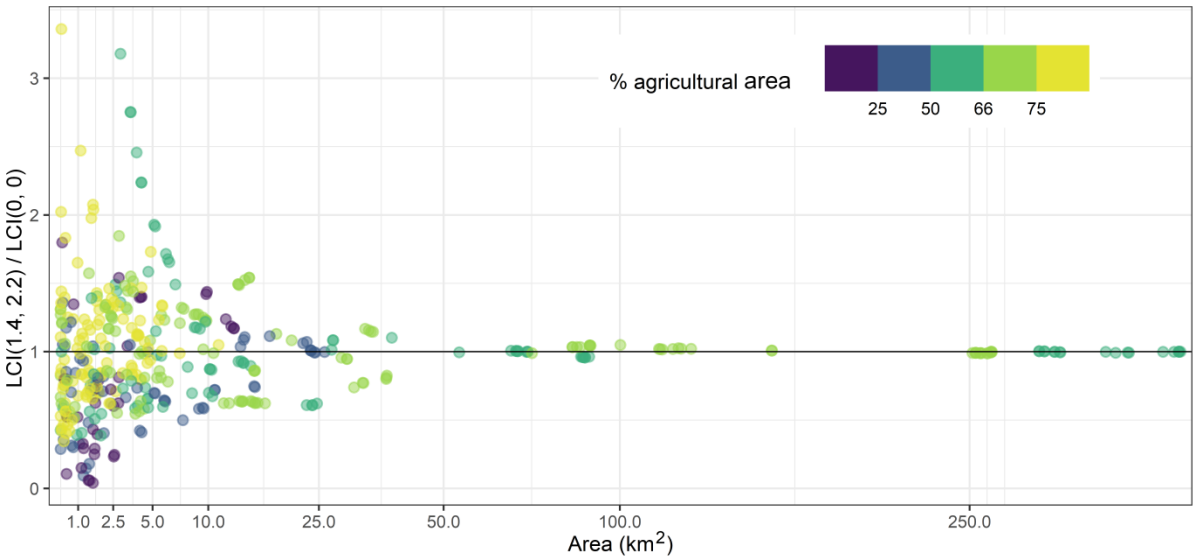


Fig. 6 Stabilisation of the ratio of the LCI-TP (landscape configuration index (LCI) for a=1.4 and b=2.2) to landscape composition index (LCI for a=b=0) as a function of area (square-root-transformed x-axis) of subcatchments in the study area (points)

This ratio varied greatly (0.02-3.37) for subcatchments smaller than 10 km^2, the typical size of 1st or 2nd order stream catchments but varied less (0.55-1.53) for subcatchments of 10-50 km^2. The high variability in this ratio, even for heavily farmed subcatchments, indicates that the landscape can still be optimised at this scale. For catchments larger than 50 km², which corresponds to 4th order rivers in the study area, the ratio converged to 1. The main implication is that for headwater catchments (<50 km^2) the correlation between landscape composition and configuration (as defined by the LCI-TP) is weak and non-significant (Fig. 2S), while for larger catchments the correlation is strong (R^2 near 1) (Fig. 2S).

4. Discussion

4.1. Spatial configuration of nutrient sources influences P but not N exports

Landscape composition is a strong predictor of median N concentrations, while landscape configuration as defined here, seems to have no influence (Fig. 2A). This result agrees with previous research that indicates a strong correlation between metrics of agricultural N pressure intensity, such as N surplus or N concentrations in streams (Dupas et al. 2015;

Kronvang et al. 2005), leaving little space for other controlling factors, such as landscape configuration. However, distributed process-based modelling of N fate in agricultural catchments has shown that landscape configuration can have a of second-order influence on N concentrations (Beaujouan et al. 2002; Casal et al. 2019; McDowell et al. 2014), which may not have been captured in our study. P concentrations are not related to landscape composition (Bol et al. 2018) or even P inputs (Dupas et al. 2015; Frei et al. 2020) in headwater catchments. Multiple factors influence P transfers to streams: soil type and P content (Djodjic et al. 2004), tile-drainage (King et al. 2015a; King et al. 2015b), small ponds and hillside storage reservoirs (Schmadel et al. 2019), in-hillslope depressions (Smith and Livingston 2013), leaks from septic tanks (Withers et al. 2011) and livestock buildings, bank erosion (Kronvang et al. 2012), and ditch-dredging management (Smith and Pappas 2007). While hedgerows have little influence on N transfers at the catchment scale (Benhamou et al. 2013; Thomas and Abbott 2018), it is difficult to quantify their combined effects on P transfers. Without considering these factors, our optimised LCI ranked the headwater catchments reasonably well according to their P exports (Fig. 2B), although the residuals may have been due to these other factors. We demonstrate that the spatial configuration of P sources in the landscape is critical to understand P transfer from land to streams in headwater catchments. The influence of the spatial configuration of nutrient sources on P but not N concentrations in agricultural headwater streams results in high variability in N:P ratios in these ecosystems and has many consequences on stream algae communities (Pringle 1990; Stelzer and Lamberti 2001).

The observation that landscape spatial organisation influences N and P transfers is not new. Nonetheless, our approach differs from previous studies that used single or multiple regression of several landscape metrics compiled in FRAGSTATS (McGarigal and Marks 1995) to predict nutrient exports at the catchment scale. Despite the large body of studies that use these landscape metrics (Bu et al. 2014; Lee et al. 2009; Ouyang et al. 2014; Zhang et al. 2019), no robust cause-effect relationships emerge, and many of them give conflicting results (Wang et al. 2020). Three reasons may explain why these studies, taken together, are not conclusive: i) the many pairwise or multiple regressions between water quality parameters and landscape metrics generate spurious correlations that are not generally discussed, ii) landscape metrics depend greatly on the resolution of GIS input data, and iii) the studies do not consider the topological dimension, which is fundamental for explaining hillslope-to-stream transfers and landscape configuration (Thomas et al. 2016b). Several approaches have integrated this topological control with the hypothesis that nutrient sources near streams (Sliva and Dudley Williams 2001; Yates et al. 2014) and/or that overlap flow accumulation areas (Peterson et al. 2011; Staponites et al. 2019) have a disproportionally

higher influence on nutrient exports than other areas. While these approaches still depend on the spatial resolution of GIS data, they are “hypothesis-driven” rather than purely “data-driven”, which decreases the risk of spurious correlations. A longstanding weakness of these approaches, however, is their rigidity due to the lack of calibrated parameters. For example, the HAiFLS index (Peterson et al. 2011) and Flow-A index (Staponites et al. 2019) included both the distance to streams and flow accumulation, but the latter dominates the index value by construction, because it varies more than the former, and neither index allows both factors to be weighted by calibrated coefficients. The stochastic approach in the LCI developed here is more flexible, and we show that both hydrological distance and flow accumulation influence P transfer.

4.2. Spatial variability and temporal stability of critical source areas

The strongest correlation between median NO_3^- concentrations and LCI was for (a, b) = (0, 0), which shows that each source (agricultural area) contributed the same, regardless of its distance to streams or overlap with flow accumulation areas. This confirms the need to consider the entire catchment to reduce N loads in agricultural catchments. For median P concentrations, the optimal was found for (a, b) = (1.4, 2.2), a value for which 5% of the agricultural area concentrated 95% of the weight assigned by our index at the 10 m-pixel resolution. These values are similar to those found by Thomas et al. (2016a)), who classified 1.6-3.4% of the catchment area (during median storm events) and 2.9-8.5% (during upper-quartile events) as prone to P transfer, based on a CSA model that also considered land use and topography as input variables. Summing the LCI-TP at the field scale indicates that the 20% of fields at highest risk represent 85% of the total weights, which are the fields on which mitigation measures should be prioritised. The distribution is asymmetrical, with 69% of fields having a mean $LCI\text{-}TP_{field}$ less than 1. This information at multiple scales can be a tool to maximise ecosystem services at the catchment scale, by reorganising landscapes to decrease P transfer without increasing the percentage of set-aside areas (Doody et al. 2016).

The shape and location of the sub-field CSAs overlap both the mandatory riparian buffer strips (5 m according to local application of the European Union Nitrate Directive (DREAL 2018)) and in-hillslope CSAs based on the Topographical Wetness Index. Because most buffer strips in the study area were installed recently, we included them in P sources as they were enriched in P before conversion (Dodd and Sharpley 2016; Gu et al. 2017; Gu et al. 2018; Jarvie et al. 2017; Roberts et al. 2012). The location and shape of the CSAs indicate the need for new shapes of buffer zones, with variable widths and locations along ditches as well. Because buffer strips are critical sources of nutrients at the headwater catchment scale, they require new management practices. Potential solutions include sowing species that can

capture more P (William M. Roberts 2019), mowing and exporting the residues each year (Fiorellino et al. 2017), and applying amendments that have high P-sorbing ability (Borno et al. 2018).

The temporal stability of the optimal parameters (a, b) for TP during the flow period is consistent with the spatial stability concept (Abbott et al. 2018), but is contrary to the concept of variable source areas (Collick et al. 2015; Dahlke et al. 2012). Our interpretation of this temporal stability is that even though the soil-to-stream connectivity varied temporally during the sampling period, the TP concentrations observed may reflect the remobilisation of sediments transferred during rare erosion events. Observing storm events that result in surface transfers requires frequent observations (Cassidy and Jordan 2011). During the low-flow dates, the correlations with LCI-TP were not significant: different sources and sinks likely dominate the influence of agricultural areas and their configuration, which predominates during the flow period. Leaks from septic tanks or animal buildings and desorption from sediments are the most likely sources of P during this ecologically sensitive period, while hydrological disconnection, uptake, and sedimentation can be sinks of P (Lannergård et al. 2020; Sandstrom et al. 2020).

4.3. Landscape homogenisation with increasing catchment size: consequences for management, monitoring, and modelling

When means were aggregated into 1 km^2 subcatchments, which is the typical size of a 1^{st} order catchment in the study area, the LCI-TP showed high spatial variability (Fig. 5D, 5F). As mentioned, this variability decreased as the aggregation size increased (Fig. 5E, 5G), and landscape composition was weakly correlated with landscape configuration for catchments smaller than 50 km^2 but strongly correlated for those larger than 50 km^2 (Fig. 6, Fig. 2S). These results explain, at least partially, why relationships between water quality parameters and landscape composition metrics break down below a certain catchment size (Bol et al. 2018). They also shed light on the long-standing difficulty in scaling from nutrient export models at the field scale to those at the catchment scale: the spatial configuration of the nutrient sources can be critical.

The variability in the LCI-TP in small subcatchments and its homogenisation in larger catchments is the expression of a degree of unstructured heterogeneity (i.e. randomness) of the spatial configuration of agricultural areas (Musolff et al. 2017). Some almost entirely agricultural subcatchments have a TP sensitivity index less than 1, while some mixed-land-use catchments have a TP sensitivity index greater than 1. The latter provides the opportunity to introduce structured heterogeneity, i.e. to reorganise agricultural activities spatially to reduce P transfers to streams (Musolff et al. 2017).

These results have implications for both modelling and monitoring. Semi-distributed models that simulate P exports from an entire catchment with simulation units smaller than 50 km^2 should include a coefficient to represent the spatial configuration of the agricultural areas. A distributed model should consider ditches (as they are the entry point of P into the hydrographic network) and, especially, the spatial variability in P sources. When monitoring subcatchments that are smaller than the homogenisation threshold and have similar agricultural land-use composition, the observed differences in P loads cannot be related directly to the agricultural practices or soil properties. The spatial configuration of agricultural areas appears to exert a major control on median P concentrations. This is particularly important for targeting measures to the most cost-effective fields (Doody et al. 2016) and could increase in importance due to the recent development of innovative financial tools to improve water quality, such as payment for ecosystem services schemes, whose obligation to achieve results is increasing (Hejnowicz et al. 2014). Our research provides a data-driven method to identify CSAs and thus the most cost-effective fields on which to implement mitigation measures.

The 50 km^2 landscape homogenisation threshold found in this study is similar to the 18-68 km^2 stream-concentration thresholds found by Abbott et al. (2018) for the Rance and Haut-Couesnon catchments, also located in western France. We assume that the optimised coefficients and homogenisation threshold would vary with the topo-climatic conditions and agricultural landscape characteristics. More research is needed to confirm this connection between landscape configuration and P loads in different environmental settings. Given the high heterogeneity of landscape configuration, this research calls for a spatially dense monitoring network of headwater catchments. We also recommend using the LCI to investigate the influence of landscape configuration on other water contaminants that are transferred mainly by surface flow paths, such as pesticides and faecal bacteria.

Declarations

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Conflicts of interest/Competing interests (include appropriate disclosures)

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval (include appropriate approvals or waivers)

Not applicable

Consent to participate (include appropriate statements)

Not applicable

Consent for publication (include appropriate statements)

All co-authors – A. Casquin, R. Dupas, S. Gu, E. Couic, G. Gruau and P. Durand – have approved the manuscript and agree with its submission to Landscape Ecology

Availability of data and material (data transparency)

This study is based on a recently published dataset entitled *Repeated synoptic sampling for water chemistry monitoring in the Yvel catchment, northwestern France* available at https://www.hydroshare.org/resource/7c7d7f6dd1f14450883ae1c243c3c28f/ (Dupas *et al.* 2020). It contains the hydrochemical data and the geographic information of the study site.

Code availability (software application or custom code)

The code (R script) will be made available along with the article upon acceptance on a github repository.

Authors' contributions

AC: Conceptualization, Methodology, Formal analysis, Visualization, Writing - original draft. RD: Conceptualization, Supervision, Funding acquisition. SG: Sample collection and analysis. EC: Sample collection and analysis. GG: Project administration, Funding acquisition. PD: Supervision. All authors contributed substantially to revisions.

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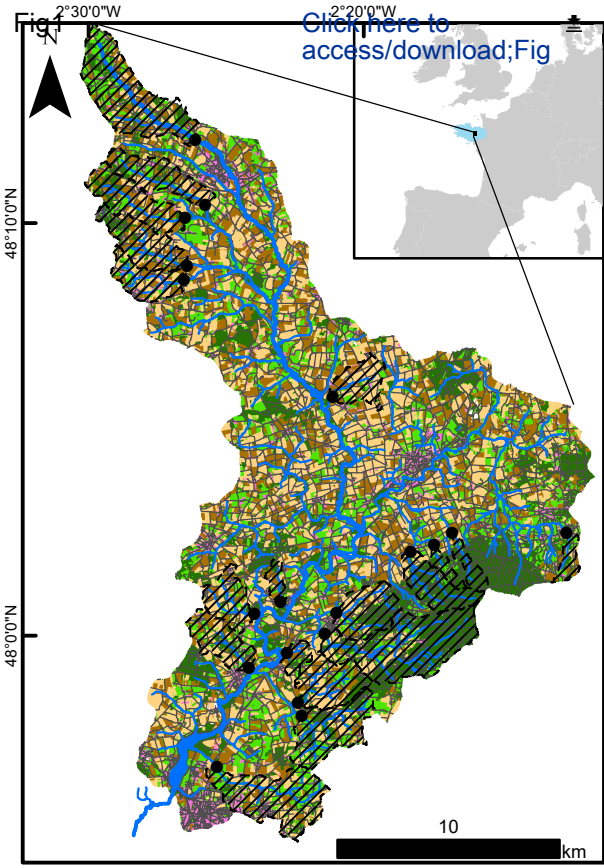
Fig 1



- Monitored subcatchment outlet
- Monitored subcatchment
- Main river
- Hydrography
- Road/Ditch
- Urban
- Wheat/Barley
- Maize
- Grassland
- Forest
- Water

Fig2

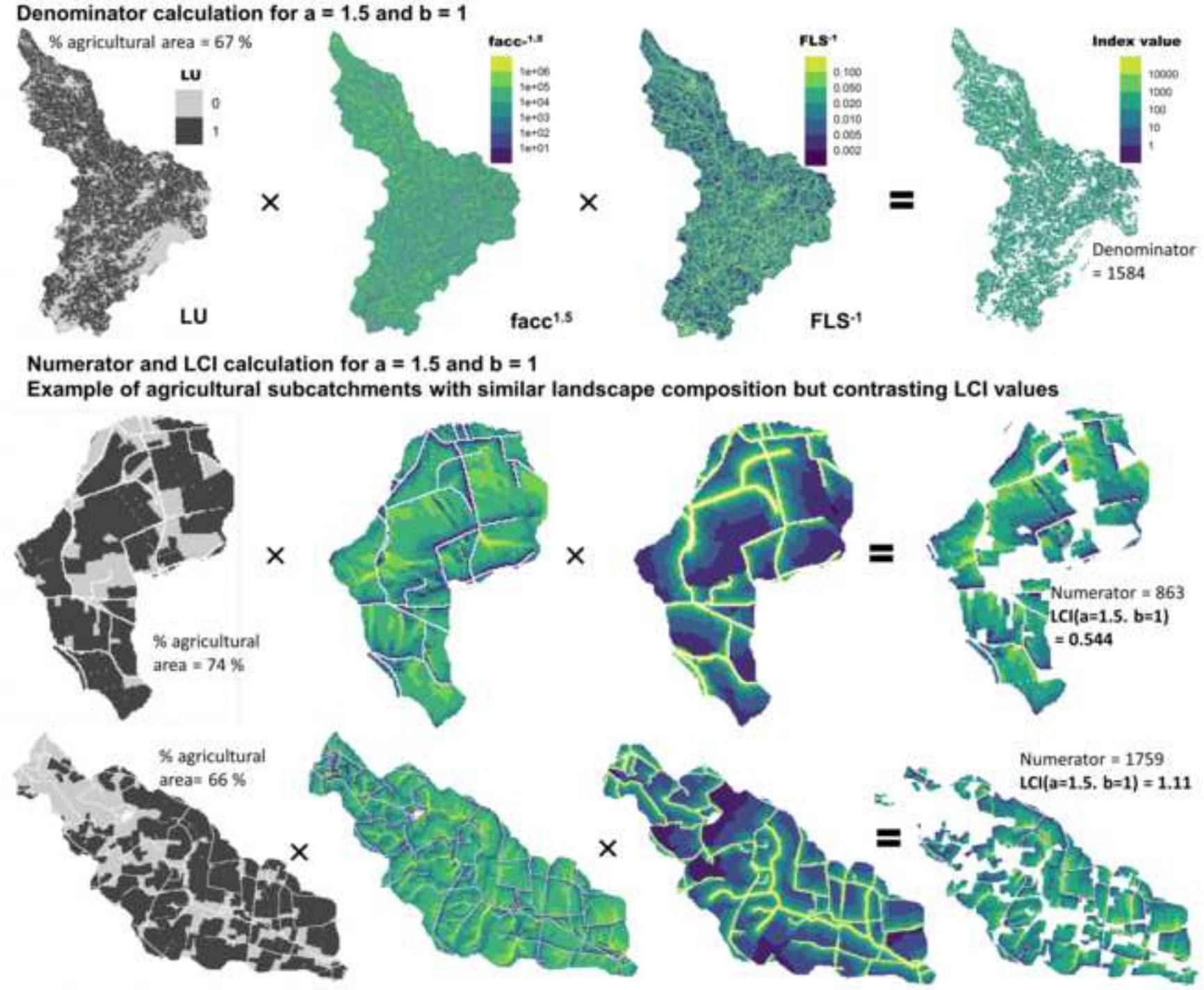
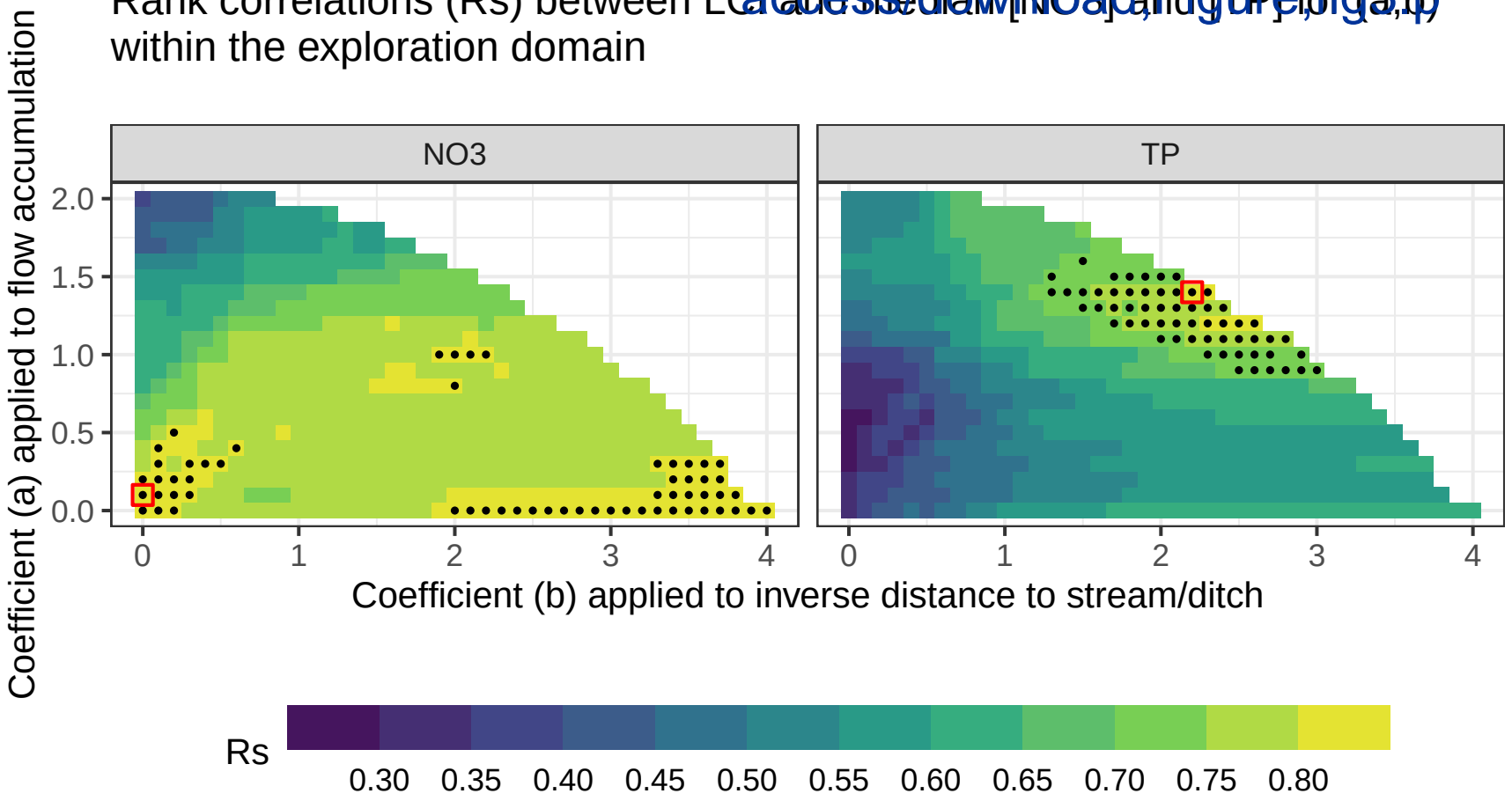


Fig3

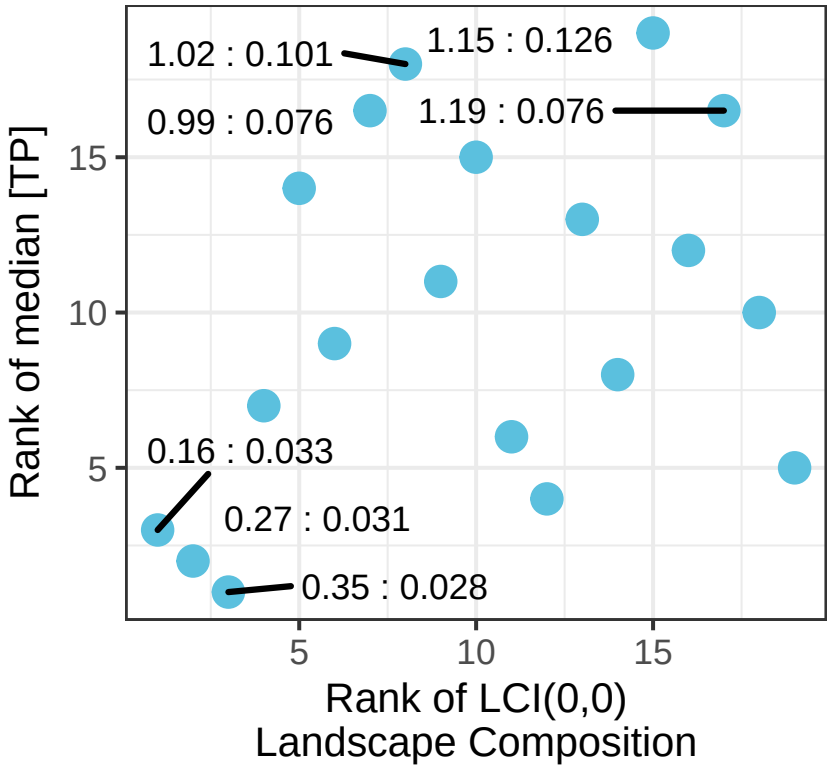
A

Rank correlations (Rs) between LCA at sediment [NO3] and TP [TP] for (a,b) within the exploration domain



B

Rs = 0.35, p−val = 0.145



C

Rs = 0.81, p−val = 2.27e−05

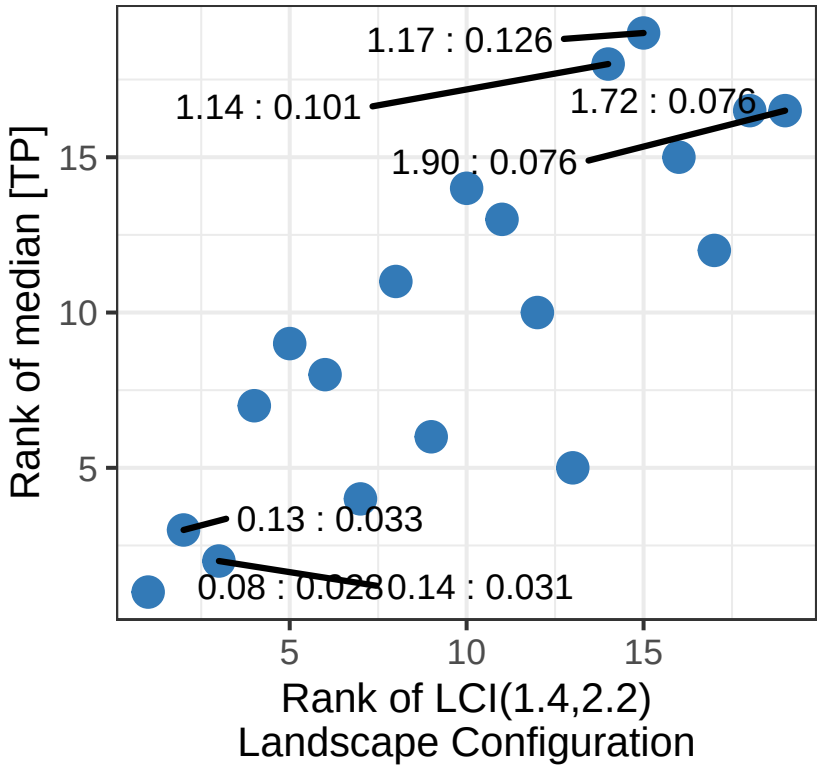


Fig4

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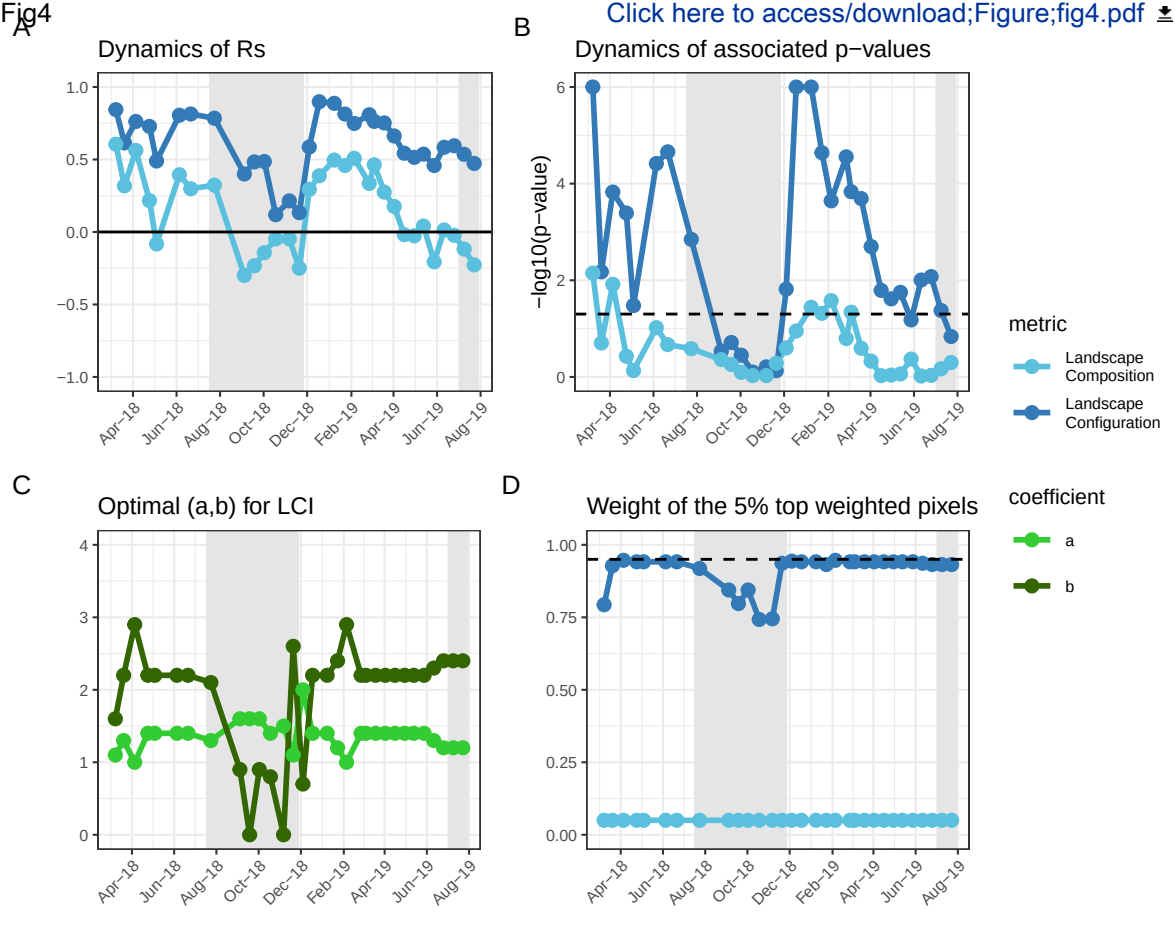


Fig5

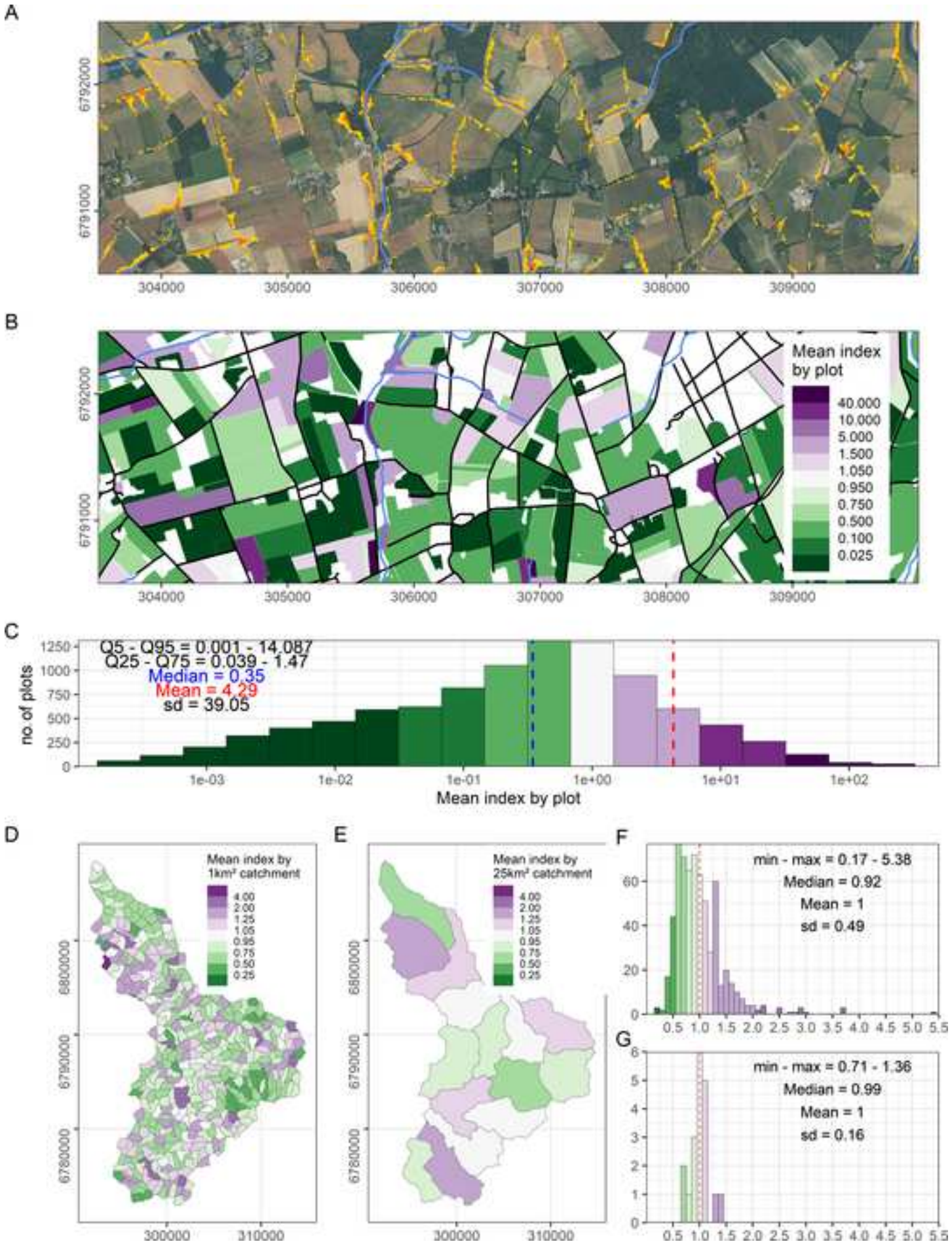
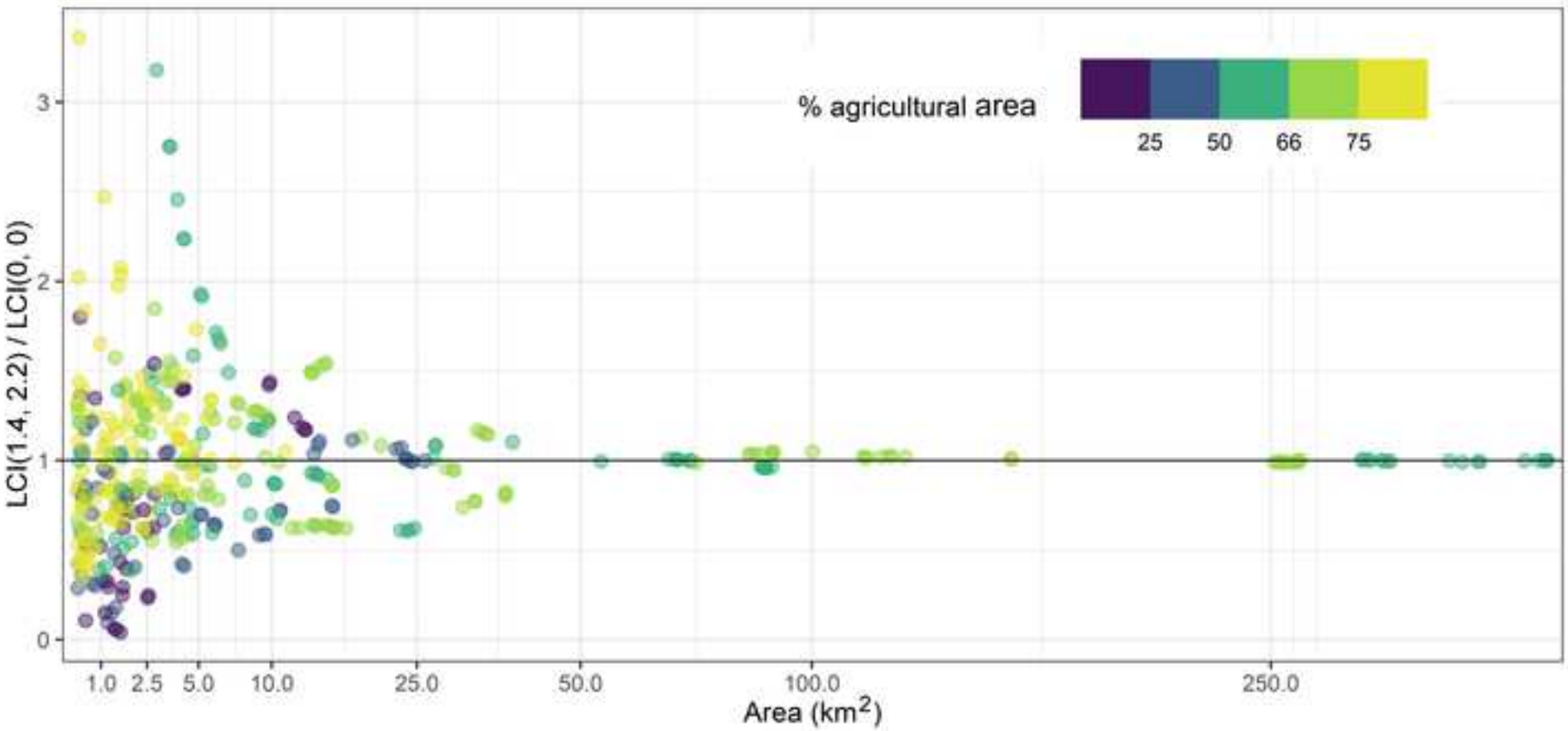


Fig6

Click here to access/download;Figure;FIg6.tif



Editor du Jour

Certificate of English-Language Proofreading

This certificate confirms that the manuscript entitled **"The influence of landscape spatial configuration on nitrogen and phosphorus exports in agricultural catchments"** by Antoine Casquin et al. was edited and proofread for correct English-language content by two native speakers of American English.

Michelle Corson

29 January 2021

Certificate no. 2021-04

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Subject: Submission of a manuscript for evaluation and publication in *Landscape Ecology*

Dear Editors,

Enclosed is our manuscript, "The influence of landscape spatial configuration on nitrogen and phosphorus exports in agricultural catchments" by Antoine Casquin et al., which we would like to submit for publication as a Research paper in *Landscape Ecology*.

This manuscript has not been previously published, in whole or in part, and is not under consideration by another journal. All co-authors – A. Casquin, R. Dupas, S. Gu, E. Couic, G. Gruau and P. Durand – have approved the manuscript and agree with its submission to *Landscape Ecology.* We had the English proofread by a professional proofreader ("Editor du Jour"; see enclosed English editing certificate).

The spatial variability in nutrient transfer from land to stream in seemingly similar headwater catchments is currently not fully understood, and upscaling results from the plot scale to the catchment scale remains difficult. Our study indicates that including the influence of the spatial configuration of nutrient sources in relation to the hydrographic network is crucial to explore this issue.

We assessed the influence of landscape spatial arrangement on nitrate (NO_3^-) and total phosphorus (TP) exports at the catchment scale. We developed a novel landscape configuration index that we optimised based on NO_3^- and TP concentrations in 19 headwater streams. Results show that the spatial configuration of nutrient sources and sinks in headwaters has a large influence on TP transfers but not NO_3^- transfers. We also reveal that the spatial configuration of sources is heterogeneous at small scales (<1 km^2) but homogeneous at larger scales (> 50 km^2), and we discuss the implications of these findings.

This study is based on a recently published dataset entitled *Repeated synoptic sampling for water chemistry monitoring in the Yvel catchment, northwestern France*. We plan to make the code (R script) available along with the article upon acceptance.

Thank you for considering our manuscript.

Sincerely,

Antoine Casquin