Review HESS-2015-69

Dear Editor of Hydrology and Earth System Sciences
Dr Frans van Geer

As explained in the response letters to the editor and reviewers, we have made major changes to the organization and logic of the article. Following the requirements of the special issue of HESS and taking into account the suggestions from you and the reviewers, we have endeavoured to take full advantage of the strengths of our work while removing or restructuring the weak aspects identified during the review process. In the revised version, we have focused on the strategies for monitoring stream phosphorus levels in contrasting climate-driven flow regimes, and land use intensity is now considered as a contrasting condition under each climatic/hydrologic scenario. We therefore place emphasis on the overall behavior of the catchments studied in relation to the measured temporal dynamics of phosphorus and on comparing the performance of alternative monitoring strategies under contrasting climate-driven flow and land use conditions.

The substantial changes in the manuscript include:

1) Reformulation of the title, objectives and expected results, in the line stated in the response letter, aiming at improving their rigour and coherence.
2) Revision of the structure of the Introduction, and a more precise and focused Material and Methods section.
3) Addition of relevant information as required by the reviewers, some of it as supplementary material (GIS maps on land use).
4) Improvement of general presentation of Results and reduction of the number of figures in this section.
5) Revision of the structure and reduction in length of the Discussion section, and removal of the previous subsections to achieve a more coherent piece of text.
6) Removal of the Conclusions section after presentation of main conclusions at the end of the Discussion section.

We believe that we have managed to consider most (if not all) of the suggestions made by the two reviewers and by the editor and are grateful for the many valuable comments, which have greatly improved our work.

We hope that your prestigious journal will consider accepting and publishing our new revised manuscript.

Looking forward to your response, kind regards,
Guillermo Goyenola
On behalf of all authors

~~Phosphorus dynamics in lowland streams as a response to climatic, hydrological and agricultural land use gradients~~

Monitoring strategies of stream phosphorus under contrasting climate-driven flow regimes

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Abstract

Climate and hydrology are relevant control factors ~~for~~ determining the timing and amount of nutrient losses from land to downstream aquatic systems, in particular of P from agricultural ~~fields to freshwaters. In this~~ lands. The main objective of the study~~, we evaluated the effect of agricultural intensification on the concentrations, dynamics and~~ was to evaluate the differences in P export ~~of phosphorus (P)~~patterns and the performance of alternative monitoring strategies in streams ~~in two~~under contrasting climate~~ and hydrological~~ -driven flow regimes~~ (~~. We compared a set of paired streams draining lowland micro-catchments under temperate climate and stable discharge conditions (Denmark) and ~~subtropical~~ under sub-tropical climate and flashy conditions (Uruguay). We applied two alternative nutrient sampling ~~programmes~~programs (high frequency composite sampling and low frequency instantaneous-grab sampling) and ~~three alternative methods to estimate exported P from the catchments. A source apportionment model was applied to evaluate the~~ estimated the contribution derived from point and diffuse sources ~~in all four catchments studied. Climatic and hydrological characteristics of catchments expressed as flow responsiveness (flashiness), exerted control on catchment and stream TP dynamics, having consequences that were more significant than the outcome of different TP monitoring and export estimation strategies. The impact of intensification of agriculture differed between the two contrasting climate zones. Intensification had a significant impact on subtropical climate with much higher total (~~fitting a source apportionment model. We expected to detect a pattern of higher total and particulate phosphorus export from diffuse sources in streams in Uruguay streams, mostly as a consequence of higher variability in flow regime (higher flashiness). Contrarily, we found a higher contribution of dissolved P in flashy streams. We did not find a notably poorer performance of the low frequency sampling program to estimate P exports in flashy streams compared to the less variable streams. We also found signs of interaction between climate/hydrology and land use intensity, in particular in the presence of point sources of P, leading to a bias towards underestimation of P in hydrologically stable streams and overestimation of P in flashy streams. Based on our findings, we suggest that the evaluation and use of more accurate monitoring methods, such as ~~high as 4436 µg P L^{-1}), particulate, dissolved and reactive soluble P concentrations and higher P export (as high as 5.20 kg P ha^{-1} year^{-1}).~~automatized flow-proportional water samplers and automatized bankside analysers, should be prioritized whenever is logistically possible. However, ~~we did not find~~ it seems particularly relevant in currently flashy systems and also in systems where climate change predictions suggest an ~~increased contribution of particulate P to total P as consequence of higher~~ increase in stream flashiness ~~and intensification of agriculture. The high P concentrations at low flow and predominance of dissolved P in subtropical streams actually exacerbate the environmental and sanitary risks associated with eutrophication. In the other hand, temperate intensively farmed stream had lower TP than extensively farmed stream. Our results suggest that the lack of environmental regulations of agricultural production has more severe consequences on water quality, than climatic and hydrological differences between the analysed catchments.~~.

1 Introduction

~~Land use is intensifying worldwide (Foley et al., 2005), particularly in many developing countries that are undergoing a process of agricultural expansion and intensification (Alexandratos and Bruinsma, 2012). It has been suggested that agriculture can promote regime shifts in freshwater ecosystem by directly and indirectly altering different components of the hydrological cycle (Gordon et al., 2007).In particular, the impacts of agriculture on water quality depend on several control factors such as cropping system, livestock type and density, use of fertilizers, tillage operations, among others (Moss, 2008; Sharpley et al., 1994). Moreover, climate, hydrology and soil types in the catchment are important for determining the timing and amount of nutrient losses from agricultural fields~~ The global demand for phosphorus for fertilizers is projected to increase in the coming decades, although the existing global reserves will likely be exhausted within this century (Cordell et al., 2009). Nowadays, the global flow of phosphorus runs from mines through farms to aquatic systems, contributing to eutrophication and triggering aquatic ecosystem degradation (Carpenter et al., 1998; Elser and Bennett, 2011). Thus, humanity faces the challenge of sustaining food production while reducing the associated environmental costs

The biogeochemical processes inside a catchment, which determine the loss of phosphorus from the land to aquatic systems, are mainly dependent of climatic and hydrological regimes (Bormann and Likens, 1967). Streams located in areas with short duration-high magnitude rainfall events may respond with frequent and rapid changes in discharge (to be “flashy” streams sensu Baker et al., 2004) and may potentially have a higher risk of diffuse transfer of nutrients from land to water ~~(Haygarth and Jarvis, 2002; Kronvang et al., 2007)~~(Cassidy and Jordan, 2011; Haygarth et al., 1999)~~.~~

~~Streams located in areas with short duration-high magnitude rainfall events may respond with frequent and rapid changes in discharge (to be “flashy” streams sensu Baker et al., 2004) and potentially have a higher risk of diffuse transfer of nutrients from soils to water (Cassidy and Jordan, 2011; Haygarth et al., 1999). Additionally, modifications in land use and management may lead to an increase or decrease in flashiness (Baker et al., 2004). Nutrient monitoring may, therefore, need to employ different sampling frequencies in stable or highly flashy streams, to~~

~~accurately capture and describe the temporal dynamics of nutrient concentrations in the aquatic ecosystems (Cassidy and Jordan, 2011).~~

~~Accurate estimation of phosphorus (P) dynamics and mass delivered to lotic systems is crucial to determine causal relationships with catchment activities, mass balances, temporal trends, environmental impacts and develop models (Kronvang and Bruhn, 1996). Irrespective of the hydrological nature of the catchments, nutrient monitoring programmes are frequently based on low cost/low frequency sampling at discrete intervals. Comparisons of monitoring and calculation methods to estimate exported P have been reported in a vast series of papers (e.g. Aulenbach and Hooper, 2006; Jordan and Cassidy, 2011; Kronvang and Bruhn, 1996; Preston et al., 1989; Raymond et al., 2014; Robertson and Roerish, 1999). Among others, monitoring strategies vary from low frequency instantaneous grab sampling and linear interpolation of concentrations between sampling days (henceforth LFS-LI; Kronvang and Bruhn, 1996) through rating curve estimations of nutrient loads (Stenback et al., 2011) and composite or serial sampling (Defew et al., 2013; Harmel et al., 2003), to near-continuous monitoring stations including bankside analysers (Cassidy and Jordan, 2011; Outram et al., 2014).~~

~~We conducted a comparative study of hydrology and the concentrations and export of different P forms in paired lowland micro-catchments (< 20 km^2) having different agricultural land use intensity and covering two distinct climate-hydrological conditions (temperate and stable discharge regime in Denmark and sub-tropical and flashy conditions in Uruguay). We applied two alternative nutrient sampling programmes (high frequency composite sampling and low frequency instantaneous-grab sampling) and three alternative methods to estimate exported P from the catchments. The main objective of the study was to evaluate the direction and magnitude of the effects of agricultural intensification on the concentrations, dynamics and export of P in streams under contrasting climate and hydrological regimes. Three questions were addressed: i) How do alternative P monitoring programmes capture the differences in climate-hydrology between the two kinds of land use? ii) Do streams in intensively farmed micro-catchments have higher P concentrations and exports than those in extensively farmed catchments, beyond climatic-hydrologic differences? and iii) Is there a synergistic interaction between agricultural intensification and climatic-hydrological variability in water P dynamics?~~

. The importance of understanding hydrology-driven variations in nutrient discharge increases in the current climate change scenario where strong hydrological changes are expected in many different parts of the world.

Irrespective of the hydrological nature of the catchments, nutrient monitoring programs are frequently based on low frequency sampling at discrete intervals. This kind of sampling strategy complemented by interpolation methods are prone to very high uncertainties due to typical under-representation of high discharge, short-duration events (Defew et al., 2013; Jones et al., 2012; Jordan and Cassidy, 2011; Stelzer and Likens, 2006). A priori, the risk of missing the key moments when phosphorus is delivered to the streams is higher in flashy streams than in hydrologically stable ones. An approach to advance the understanding of how different monitoring schemes capture catchment phosphorus processes in contrasting climate-driven flow regimes is to compare monitoring performances in catchments under different climatic conditions and under different conditions of nutrient inputs.

The main objectives of our study were to evaluate the differences in stream P export patterns and the performance of alternative monitoring strategies in contrasting climate-driven flow regimes. We expected to detect higher total and particulate P exports in streams located in sites with higher frequency of extreme rainfall events and higher stream hydrological variability (flashiness). We also expected poorer performance of low frequency sampling programs under such conditions.

2 Material and Methods

~~2.1. Catchment selection~~

2.1. ~~Based on~~ Design rationale: Selection of ~~our prior knowledge about the~~ case studies

We conducted a comparative study of concentrations and export of different ~~hydrologic regimes of temperate~~ P forms in two paired streams ~~(higher~~ under two distinct climatic-hydrological ~~stability~~conditions: temperate climate and stable discharge conditions (Denmark) and subtropical ~~streams (higher hydrological variability), four comparable low order streams~~

~~draining lowland~~climate and flashy conditions (Uruguay). In both countries, the topography of the selected areas can be described as gently rolling plains (mean slope < 5%). Two main causes explaining the differences in flashiness between Denmark and Uruguay are precipitation patterns (annual average precipitation, 745 mm since 1990 in Denmark and 1300 mm in Uruguay, according to the Danish Meteorological Institute (DMI, 2015) and the National Institute for Agriculture Research, INIA, Uruguay (Castaño et al., 2011), respectively, and depth of soils and derived water storage capacity. The long-term continuous monitoring data in Danish catchments and the existence of published works evaluating alternative sampling strategies (e.g. Kronvang and Bruhn, 1996) decided us to use the temperate streams as the reference systems in our comparison.

In each area, two lowland non-experimental micro-catchments ~~were selected in Denmark (Europe; Gudenå River basin) and Uruguay (South America; Santa Lucía Chico River basin). Two streams draining catchments with contrasting land uses~~(< 20 km^2) were selected ~~in each climate. Intensive agriculture~~as typical productive systems to represent extremes of land use intensity (hereafter LUI) in each area.

As higher LUI catchments, we selected catchments where intensive farming comprising more than 90% of the total land area, with arable cropping systems~~ was the predominant land use (more than 90% of total area) in one of the two catchment types~~, intensive use of fertilizers, and high density of livestock (Table 1). In the Uruguayan ~~intensive~~high-LUI catchment, the ~~agricultural~~farming production system was based on forage crops, no-till practices associated with intensive application of mineral fertilizers over the soil surface (Derpsch et al., 2010), dairy cattle feeding all year ~~around~~round in open fields, and no effluent treatment facilities on milking plants~~, while in~~. In Denmark, the ~~intensive~~high-LUI catchment included a ~~rotational~~rotation cropping system with winter cereals and confined pig farms with ~~effluent~~slurry storage facilities. ~~Moreover, the intensive catchment in Denmark mostly consisted of arable~~ In Denmark, most loamy agricultural fields are drained with subsurface tile drainage systems ~~(Grant et al., 1996)~~(Grant et al., 1996)~~.~~ , and the manure originating from farming activities is reutilized with a demand on a 75% reuse of N in slurry.

The ~~extensive land use~~lower-LUI catchments were chosen so as to ~~best~~ represent local more preserved conditions. The Uruguayan ~~extensive~~low-LUI catchment was dominated by the natural ~~grassland~~grasslands of the Pampa Biome ~~(Allaby, 2006)~~(Allaby, 2006)~~ as a basis of a~~ and sustained low density cattle production (70% of total area and below 1 head by hectare;

Table 1). ~~A~~In contrast, a mixture of deciduous and coniferous ~~trees~~forests dominated the Danish ~~extensive~~low-LUI catchment (Table 1).

The subtropical ~~intensive~~high-LUI catchment had 170 inhabitants and the ~~extensive~~low-LUI catchment only 20 inhabitants ~~(Instituto Nacional de Estadística, 2015). The~~(National Institute of Statistics, 2015). In the former~~ catchment featured~~, the sewage from only 10 households is treated in a facultative ~~lagoon that receives effluents from 10 households.~~pond. All other households in both subtropical catchments had leaking septic tanks. The point sources ~~for~~in the temperate catchments ~~corresponded~~were mainly ~~to~~ scattered dwellings without connection to sewage treatment plants. The temperate ~~intensive~~high-LUI stream received ~~the~~ stormwater outlets ~~pouring~~ from a small village~~, but their~~ whose sewage water is pumped to a treatment plant with tertiary treatment outside the catchment.

Maps including land use for each catchment were included as supplementary material. Henceforth, we will refer to the temperate- Danish ~~catchments~~streams as “~~TEMP~~STABLE” and the subtropical-Uruguayan ~~catchments~~streams as “~~SUBTR~~FLASHY”, while LUI categorization of the intensive and extensive production catchments ~~with intensive and extensive land use~~ will be referred to as “~~INT~~high-LUI” and ~~as “EXT~~“low-LUI”, respectively.

~~Measurements~~

2.2. Phosphorus monitoring

Similar gauging stations were established in all four micro-catchments. We applied two alternative nutrient sampling programs: low frequency instantaneous-grab sampling and high frequency composite sampling. Instantaneous-grab sampling of water was conducted fortnightly, and P exports were estimated by two daily step interpolation methods. High frequency composite sampling was undertaken using Glacier refrigerated automatic samplers (ISCO-Teledyne). The samplers collected an equal water volume every four hours, and the composite samples were also collected fortnightly. The final phosphorus concentration in the only sampler carboy thus represented a time-proportional average for the fortnightly sampling period. As the high frequency composite samples integrated more information (i.e. shorter time steps, with higher probability of capturing extreme events), we expected this method to provide better estimates of the 'true' exported P from the catchments. Based on this assumption, we evaluated the deviation relative to the estimation methods based on instantaneous-grab samples.

We analyzed the measurements and results from a two-year monitoring period ~~were analysed~~ (March 2010 to March 2012 in ~~TEMP, and~~STABLE; January 2011 to January 2013 in ~~SUBTR).~~FLASHY).

~~2.2. Field measurements~~

~~Similar gauging and monitoring stations were established in all four micro-catchments in order to register meteorological data, stream hydrology and phosphorus (P) concentrations.~~

2.3. Meteorological and hydrometric monitoring

In all catchments, CR10X data loggers (Campbell Scientific Ltd.) ~~were configured to register~~collected data every 10 minutes. In the ~~SUBTR~~FLASHY streams, we used CS450 Submersible Pressure Transducers (Campbell Scientific Ltd.) for water stage monitoring as well as HMP45C temperature probes ~~HMP45C~~ (Campbell Scientific Ltd.) and Rain-O-Matic Professional rainfall automatized gauges ~~Rain-O-Matic Professional~~ (Pronamic). In the ~~TEMP~~STABLE catchments, water level was registered with PDCR 1830 pressure sensors (Druck), while meteorological information was obtained from the Danish Meteorological Institute monitoring network based on a 10 x10 km grid.

~~Instantaneous periodic~~Periodic instantaneous flow measurements were ~~conducted during the project development period~~taken using a C2-OTT Kleinflügel, transferring data to software for the calculation of instantaneous discharge (VB-Vinge 3.0, Mølgaard Hydrometri). Non-linear stable regressions between stage and discharge at each monitoring station (rating curves) were fitted. ~~Rating curve was~~The rating curves were used ~~for calculation of~~to generate a 10 minutes ~~and daily~~ discharge ~~utilising~~data series utilizing the software HYMER (www.orbicon.com). For comparisons, discharge data is reported as area-specific runoff.

~~Two alternative water sampling strategies were implemented for nutrient analysis. High frequency composite sampling was undertaken using Glacier refrigerated automatic samplers (ISCO-Teledyne). The samplers were programmed to collect an equal water volume every four~~

~~hours, and the composite samples were collected following a fortnightly sampling programme. The final phosphorus concentration in the sampler carboy represents a time-proportional average for the complete sampling period. Complementary low frequency instantaneous grab sampling of water was conducted at every fortnightly visit to the stations.~~

~~2.3. Laboratory analysis~~

~~Sediments decanted in composite and grab sample bottles were resuspended before aliquots were taken~~

2.4. Phosphorus analysis

All instantaneous-grab and composite water samples were analyzed for total phosphorus (TP~~)~~ ~~analysis. Grab samples for~~), total dissolved phosphorus (TDP~~)~~), and particulate phosphorus (PP). In addition, also soluble reactive phosphorus (SRP) and soluble non-reactive phosphorus (NSRP) were estimated from instantaneous-grab samples.

Instantaneous-grab samples for TDP and SRP analysis were filtered ~~using~~through 0.~~45μm~~45 μm membranes. For TDP analysis, ~~TEMP~~ STABLE high frequency composite samples were filtered using 0.~~45μm~~45 μm pore size membranes, while ~~SUBTR~~ FLASHY-high frequency composite samples were filtered using Whatman GF/C (pore size 1.2~~.~~ μm). To detect possible ~~symptoms of~~ bias derived from the type of filter used, we performed a Kruskal-Wallis test ~~to evaluate statistical differences for~~on the proportional contribution of ~~dissolved forms~~TDP to TP ~~(TDP/TP)~~ between ~~SUBTR grab and composite samples. In this sense, no significant differences were found in the comparison between TDP/TP values estimated from~~ FLASHY instantaneous-grab and high frequency composite samples ~~for the SUBTR-INT stream (median for composite data = 86.7%; for grab data = 87.7%; H = 0.128, 1 degrees of freedom; p = 0.720)~~ and ~~for the SUBTR-EXT stream (median for composite data = 69.1%; for grab data = 69.7%; H = 1.580, 1 degrees of freedom; p = 0.209).~~found no significant differences. Consequently, we consider grab and composite TDP samples to be comparable. Particulate phosphorus (PP) was estimated as the difference between TP and TDP. Soluble non-reactive phosphorus (SNRP) was also calculated as the difference between TDP and SRP.

~~All the samples were determined as molybdate reactive P by equivalent spectrophotometric methods, preceded by strong oxidation in case of total forms. Samples from the Uruguayan streams were frozen at -20 °C and later analysed following the method proposed by Valderrama (1981). The water samples from the Danish streams were analysed within 48 hours following Danish standards (Danish-Standards-Association, 1985a, b). All P concentrations are expressed as micrograms of P element per litre (µg P L^{-1}).~~

All the samples were determined as molybdate reactive P by equivalent spectrophotometric methods, preceded by strong oxidation with a solution of potassium persulfate, boric acid, and sodium hydroxide following Valderrama (1981).

~~2.4.~~2.5. Data processing and analysis

~~2.4.1. Climate and hydrology~~

Climatic and runoff patterns were explored in order to investigate the main parameters relevant for P temporal dynamics. As a proxy of catchment water balance, the runoff ratio (i.e. annual percentage of rainfall water exported as runoff) was calculated ~~(Wu et al., 2013)~~(Wu et al., 2013). Additionally, to quantify ~~hydrological responsiveness,~~the variation in flow regime we calculated the Richards-Baker Index ~~(hereafter R-B Index; Baker et al., 2004)~~(hereafter R-B Index; Baker et al., 2004). The R-B Index allows for evaluation of the “flashiness~~”,~~,” or the annual ratio of absolute day-to-day fluctuations of streamflow relative to total flow~~, which is an important aspect of the hydrologic regime (Baker et al., 2004)~~(Baker et al., 2004)~~. Increasing its value with flashiness, the R-B Index ranges among 0 and infinity and assumes a value of 1 when the accumulated volume of daily oscillations has the same magnitude as the annually accumulated discharge. In the literature, maximum values reach 0.43 for mountain streams in Slovakia and Austria (Holko et al., 2011), 1.009 for Michigan streams (USA) (Fongers, 2012), and 1.32 for the catchments in the US Midwestern States (Baker et al., 2004). The baseflow index, considered as the deeper groundwater contribution to the stream flow, was estimated from daily hydrographs using the automatic routine proposed by Arnold et al. (1995).~~

~~2.4.2. Phosphorus dynamics~~

. Increasing its value with increasing flashiness, the R-B Index varies between 0 and infinity and assumes a value of 1 when the accumulated volume of daily oscillations has the same magnitude as the annually accumulated discharge. The relative contribution of baseflow to total stream flow was estimated from daily hydrographs using the automatic routine proposed by Arnold et al. (1995) as an indicator of the deeper groundwater contribution. Percent contribution of stormflow to total flow was estimated as complementary to the baseflow contribution (Table 2).

The statistical relationship between all phosphorus compounds ~~were~~ from instantaneous-grab samples was analysed by ~~means of~~ Spearman rank order correlation. The temporal dynamics of P forms were ~~presented~~followed for total P (TP), particulate P (PP), total dissolved P (TDP) and soluble reactive P (SRP~~)~~), as minimum (min), median (med) and maximum (max~~),~~) range and interquartile range (IQR~~) and were graphically presented in scatter and boxplots. As the parametric test assumptions generally failed to be accomplished even with transformed data, the~~). The statistical comparisons ~~were made~~of P temporal dynamics between the four streams were conducted using Kruskal-Wallis tests ~~(Zar, 2010)~~(Zar, 2010)~~. When differences in the median values among treatment groups were greater than would be expected by chance,~~, followed by a *post hoc* pairwise multiple comparison procedure ~~was applied to isolate the group or groups differing from the others (Dunn, 1964). Differences were considered statistically significant~~ when ~~the value of p was ≤ 0.05.~~appropriate (Dunn, 1964).

~~2.4.3. Modelling inputs from diffuse and point sources and estimation of export~~

Three different methods were used for the calculation of stream P ~~load calculations.~~export. The first method ~~(COMP)~~ was based on multiplying ~~fortnightly~~the TP and TDP concentrations obtained from the ~~time proportional~~high frequency fortnight composite samples by the accumulated discharge for the same time period ~~(Kronvang and Bruhn, 1996)~~(Kronvang and Bruhn, 1996). ~~During~~Missing data from the relatively short periods when the automatic samplers were not in operation (e.g. frozen in Denmark~~), missing data~~) were ~~regenerated~~re-generated through linear interpolation of concentrations ~~for the whole period as in previous works (Jones et al., 2012)~~(Jones et al., 2012).

Secondly, we calculated ~~daily~~exported TP and TDP ~~exported~~ from ~~linear~~ the low frequency instantaneous-grab data by two alternative methods of concentration interpolation ~~between the fortnightly grab samples to obtain daily concentrations (LFS-LI), which were subsequently multiplied by daily discharge~~: linear ~~(Kronvang and Bruhn, 1996)~~(Kronvang and Bruhn, 1996)~~.~~ ~~Lastly, concentration-discharge relationships (C-Q) were established based on grab samples for all four streams by applying the load apportionment model developed by Bowes et al. (2008). This alternative estimation strategy is based on the same low frequency data set as the LFS-LI. This simple modelling approach does not require GIS information on land use, catchment size, population or livestock density and may act as a valuable and versatile tool for catchment managers to determine/decide on suitable river mitigation options (Bowes et al., 2008).~~

~~The Bowes'~~ and concentration-discharge relationships (Bowes et al., 2008). Daily real and interpolated concentrations were subsequently multiplied by daily accumulated discharge to obtain daily export estimates.

Concentration-discharge relationships (C-Q) were established based on instantaneous-grab samples for all four streams by applying the load apportionment model developed by Bowes et al. (2008). This simple modelling approach does not require GIS information on land use, catchment size, population, or livestock density and may act as a valuable and versatile tool for catchment managers to determine suitable catchment mitigation options (Bowes et al., 2008). Several authors have found similar relationships and used them to characterize P dynamics (e.g. Meyer and Likens, 1979), calculate P sources (e.g. Bowes et al., 2008), or to calculate P transport (e.g. Kronvang and Bruhn, 1996).

The Bowes et al. (2008) method assumes that the load of phosphorus from point and diffuse sources can be modelled as a power-law function of the river volumetric flow rate (Equation 1). The total load of P at the sampling point is then a linear combination of the loads from diffuse and point source inputs, as shown in Eq. (1).

$$\mathrm{PC}= dso\_PC + pso\_PC = A.Q^{B-1} + C.Q^{D\text{-}1} \qquad (1)$$

where PC is phosphorus concentration, dso_PC is diffuse source originated PC, and pso_PC the point~~-~~-source originated PC~~,~~. Q is discharge (daily accumulated), ~~and~~while A, C, (proportionality constants) and B, D (exponents) are empirically determined parameters. ~~Parameters~~Parameter estimation was ~~made~~conducted by using a nonlinear generalized reduced gradient method to select values that ~~minimise~~minimize the residual sum of squares~~ in the Solver function in Microsoft EXCEL©.~~. Parameter B was constrained to values lower than 1 (dilution effect over point contributions) and D values greater than 1 (at no flow, the diffuse inputs tend to be zero and to increase with increasing flow). Each established C-Q relationship was used ~~as a model~~ for the calculation of daily mean concentrations and then multiplied by the daily discharge to achieve daily ~~and annual loads.~~exports. The proportional annual contribution from point sources and diffuse sources was also calculated with this method.

For ~~SUBTR~~FLASHY catchments, we estimated the maximum P contribution from human inhabitants based on the composition of household wastewater (i.e. urine, faeces, and greywater) and biodegradable solid waste per person and year based on ~~Vinnerås (2002)~~Vinnerås (2002). For ~~TEMP~~STABLE catchments, we~~ have~~ estimated the total annual load from scattered dwellings not connected to sewage treatment plants and stormwater outlets from ~~modelled results~~validated models ~~(Wiberg-Larsen et al., 2013)~~(Wiberg-Larsen et al., 2013).

~~As composite samples integrate more information (i.e. shorter time steps, higher probability of capturing extreme events) and do not force temporal variation to arbitrary dynamics (as linear on LFS-LI approximation), we consider this method to provide better estimates of the 'true' exported P from the catchments. Based on this assumption, we evaluated the uncertainty involved when utilising the two other load estimation methods: LFS-LI and C-Q. It should be noted that the concentrations obtained in composite samples are time weighted averages for each time lapse/each sample sequence, while effective exported P estimates are directly dependent on an instantaneous combination of concentration and discharge (flow-weighted averages).~~

~~2.4.4. Total, dissolved and particulate phosphorus~~

The relative contribution of ~~particulate (~~PP~~) and dissolved (TDP & SRP) P~~ to total exported P was ~~analysed for loads and concentrations in two ways. First, we calculated the percentage of the total amount of phosphorus exported in particulate forms for each hectare of catchment and~~

~~monitored year (COMP & LFS-LI methods). Then, we~~ estimated ~~the contribution of PP to TP as~~ based on data from low frequency instantaneous-grab, sampling and linear interpolation, high frequency composite sampling, and flow-weighted concentrations (FWC) estimated from high frequency composite samples on a monthly basis~~ as accumulated load divided by total flow for the considered period.~~. FWC estimation allows calculation of a flow-~~normalised~~normalized comparison of P concentrations between catchments.

3 Results

3.1. Climate and hydrology

The climate characteristics of the study period can be considered typical years for both Denmark and Uruguay. During the study period, minimum, mean, median, and maximum air temperatures were between 8 and 12 °C ~~higher~~lower in the ~~SUBTR~~temperate/STABLE catchments than in the ~~TEMP~~subtropical/FLASHY catchments (Fig. 1). The annual average temperature in the ~~TEMP~~temperate/STABLE catchments was around 8.8 °C and ranged between -7.0 to 20.4 °C. The corresponding figures for the ~~SUBTR~~subtropical/FLASHY catchments ~~was~~were around 17.5 °C and ranged between 3.7 to 32.2 °C.

In both climates, catchments ~~show~~showed similar intra-yearly distributed rain patterns, but with marked differences in frequency and intensity (Fig. 2). In the ~~TEMP~~STABLE catchments, it rained ~~for~~ almost 6 out of 10 days (58%), the rain frequency being nearly half in the ~~SUBTR~~FLASHY catchments (31%). Although there were more rainy days in the ~~TEMP~~STABLE catchments, the daily average amount of rainfall was lower (3.4 mm d^{-1}) than in ~~SUBTR~~FLASHY catchment where it amounted to 10.7 mm d^{-1}. Only one event of 50 mm d^{-1} was registered in the ~~TEMP~~STABLE catchments during the 2-year study period, while in the ~~SUBTR~~FLASHY catchments rainfall ~~was~~events > 50 mm d^{-1} ~~for~~occurred approximately 1.5% of the days, reaching extremes of > 100 mm d^{-1}. The annual rainfall was 1.44 times higher in the ~~SUBTR~~FLASHY than in the ~~TEMP~~STABLE catchments (Table 2).

Most of the water flowing in the FLASHY streams was exported during stormflow conditions (stormflow contribution > 60.8%), while in the STABLE streams water was exported during baseflow conditions (stormflow contribution < 36.4%; Table 2). The STABLE/low-LUI stream showed a very different hydrological behavior than the other three streams in that a very high percentage of the rainfall was discharged (> 62 %), with high minimum flows and low temporal variability (Fig. 2 and Table 2).

~~TEMP~~The Danish streams exhibited ~~a~~ stable ~~hydrologic regime at the annual (~~hydrological behaviour characterized by low inter-annual variability of total discharge~~)~~, and also low variability at daily scale (the R-B Index never reached values higher than 0.3; Table 2). In contrast, the ~~SUBTR~~Uruguayan streams could be classified as ~~flashy~~FLASHY systems, with an R-B index ranging ~~between~~ around 1 (0.9~~ and -~~-1.3~~ (Table 2). When comparing the hydrological regime of the streams within each climate, those~~; Table 2). The stream draining the ~~intensively farmed~~high-LUI catchments ~~had~~was the most flashy ~~characteristic~~ (Table 2).

~~Most of the water flowing in the TEMP streams was exported during baseflow conditions (64-88%), while in the SUBTR streams most was exported during stormflow conditions (29-39%; Table 2). The TEMP-EXT stream showed a very different hydrologic behaviour than the other three streams in that a very high percentage of the rainfall was discharged (64-67%), with high minimum flows and low variability (Fig. 2 and Table 2).~~

3.2. Phosphorus temporal dynamics

Total P concentrations were positively correlated with all other P fractions (PP, TDP, SRP, SNRP) in ~~the TEMP-INT~~STABLE and ~~SUBTR-INT~~FLASHY/high-LUI streams, whereas ~~for extensive~~in the low-LUI streams TP only showed a significant relationship with PP in ~~TEMP~~the STABLE and with TDP and PP in ~~SUBTR~~the FLASHY stream (Table 3). The relationships between TDP and SRP were ~~less strong~~weaker but significant ($p < 0.05$) in the ~~extensive~~low-LUI streams than in the ~~intensive~~high-LUI ones under both ~~climate~~climatic conditions (Table 3). The contributions of PP to TP were relatively similar ~~for~~in the ~~intensive~~low and ~~extensive agricultural land use~~high-LUI catchments in ~~TEMP~~STABLE, but in ~~SUBTR~~FLASHY the proportion of PP decreased with ~~the intensification~~declining intensity of ~~agriculture~~land use (Table 3). ~~Strongest~~ The strongest relationships between PP and TP were found in both ~~the TEMP and SUBTR-EXT~~STABLE streams and the FLASHY/low-LUI stream (Table 3). ~~By~~In contrast, in ~~SUBTR-INT~~FLASHY/high-LUI, TDP, and particularly SNRP, showed the strongest relationship with TP (Table 3). ~~Negatives~~Negative relationships were found

~~uniquely~~solely for ~~EXT~~low-LUI streams, between PP and TDP, ~~the first~~PP and SNRP for ~~TEMP~~the STABLE stream and between SNRP and SRP for ~~SUBTR~~the FLASHY stream (Table 3).

~~The median~~Median TP concentrations calculated for the four streams differed significantly (H = 107.~~805~~8; p ≤ 0.001), being ~~significantly~~pronouncedly higher in ~~SUBTR-INT~~the FLASHY/high-LUI stream than in any of the ~~other studied streams~~others (min = 271; med = 1.024; max = 4436 µg P L^{-1}; Fig. 3). All other paired comparisons of TP revealed no significant differences, except for the ~~TEMP~~STABLE streams where the TP concentration was significantly lower in the stream draining the ~~intensively farmed~~high-LUI catchment (median = 76 µg P L^{-1}) than in the ~~extensively farmed~~low-LUI catchment (med = 108 µg P L^{-1}; Fig. 3). No differences were registered between the ~~TEMP~~STABLE and ~~SUBTR-EXT~~FLASHY/low-LUI catchments (med = 100 µg P L^{-1}; Fig. 3~~; Fig. 3.~~).

A significant difference (H = 43.~~548~~6; p ≤ 0.001) in median PP concentrations was found between most streams (Fig. 3), with ~~higher~~highest and ~~lower~~lowest values being registered in the ~~SUBTR-INT~~FLASHY high-LUI and ~~SUBTR-EXT~~low-LUI streams (~~med~~median = 146 µg P L^{-1} and 25 µg P L^{-1}, respectively~~),~~) and intermediate values in the ~~TEMP~~STABLE streams (~~med~~median = 52 µg P L^{-1} in the ~~intensive~~high-LUI, and 80 µg P L^{-1} in the ~~extensive~~low-LUI stream). ~~The intensity of agricultural land use affected the PP~~ Particulate P concentrations ~~differently, being~~were highest in the ~~effect on the extensive~~STABLE low-LUI stream ~~in the TEMP climate,~~ and *vice versa* in the ~~SUBTR climate. The TEMP~~FLASHY streams. As expected, the STABLE streams exhibited lower temporal variation in PP than the ~~SUBTR~~FLASHY streams (IQR= 23-37 and 53-227 µg P L^{-1}, respectively).

Furthermore, a significant difference in median TDP concentrations ~~was traced between the streams (~~occurred (H = 133.~~298~~3; p ≤ 0.001; Fig. 3). *Post hoc* analysis revealed statistical ~~significance~~ equivalence only for the ~~TEMP~~STABLE streams ~~only (med~~(median = 28 µg P L^{-1} and 23 µg P L^{-1} for ~~extensive~~low and ~~intensive~~high-LUI streams). Intermediate TDP concentrations were found in ~~SUBTR-EXT~~FLASHY/low-LUI and the highest concentrations ~~were revealed~~appeared in the ~~SUBTR-INT~~FLASHY/ high-LUI stream (~~med~~median = 74 µg P L^{-1} and 756 µg P L^{-1} respectively; Fig. 3).

The median SRP concentrations also exhibited statistically significant differences between the streams (H = 141.~~157~~2; p ≤ 0.001; Fig. 3). SRP levels resembled TDP, with the lowest concentrations in the ~~TEMP~~STABLE streams (median: 2 µg P L^{-1} in both), intermediate levels in the ~~SUBTR-EXT~~FLASHY/low-LUI stream (median: 45 µg P L^{-1}~~)~~), and the highest levels in the ~~SUBTR-INT~~FLASHY/ high-LUI (median: 659 µg P L^{-1}; Fig. 3). SRP in the ~~TEMP~~STABLE streams never exceeded 23 µg P L^{-1}, and in the ~~SUBTR-EXT~~FLASHY/low-LUI stream it never exceeded 87 µg P L^{-1}. ~~By~~In contrast, the ~~SUBTR-INT~~FLASHY/high-LUI stream never had SRP concentrations lower than 219 µg P L^{-1} and SRP reached a maximum concentration of ~~1920~~1,920 µg P L^{-1} (Fig. 3).

3.3. Modelling phosphorus inputs from diffuse and point sources

~~A graphical exploration of concentration-discharge (C-Q) relationships for the SUBTR streams showed a typical pattern (Bowes et al., 2008),~~Graphical exploration of C-Q relationships for the FLASHY streams showed the typical pattern described by Bowes et al. (2008), with high TP concentrations at low discharges followed by steeply declining TP concentrations with increasing discharge (dilution associated with point source-originated P input~~)~~), and a less pronounced increase in concentrations at higher discharges (associated with diffuse source-originated P inputs; Fig. 4). The C-Q relationships for the two ~~TEMP~~STABLE streams did not show any dilution effect associated with point source inputs; therefore, the best fitting was obtained when considering only a diffuse input signal (Fig. ~~4; Table 4). The TEMP-INT C-Q relationship showed an outlier of a very high TP concentration that we could not explain.~~4; Table 4). The performance of the models evaluated with the Nash-Sutcliffe model efficiency coefficient was generally low ~~(Moriasi et al., 2007)~~(Moriasi et al., 2007), reaching a maximum value of 0.25 for the ~~TEMP-EXT~~STABLE/low-LUI stream (Table 4).

When considering the relationships established for point source-originated TP for the ~~SUBTR~~FLASHY streams, we found a higher exponent (B) in the C-Q relationships for the ~~intensive~~high-LUI catchment (Table 4). As a consequence, the decrease in TP with increasing flow (the dilution effect~~),~~) was less pronounced for the ~~intensive~~high than ~~extensive~~the low-LUI stream (at 1,000 L s^{-1} the ~~SUBTR-INT~~FLASHY/high-LUI catchment reached 85 µg P L^{-1}, while the ~~SUBTR-EXT~~FLASHY/low-LUI dropped to 5 µg P L^{-1}, Fig. 4).

Considering only diffuse sources of TP, we found a higher coefficient (C) but slightly lower exponents (D) in the C-Q relationships established for the ~~intensive~~high-LUI than for the

~~extensive~~low-LUI catchments under both climate conditions (Fig. 4 and Table 4). However, the ~~SUBTR-INT~~FLASHY/high-LUI stream always had higher TP concentrations from diffuse sources than the other streams (Fig. ~~4). All other C-Q relationships tested for TP, TDP or SRP produced poorer fits (results not shown).~~4).

3.4. Estimation of phosphorus export ~~and sources~~

Comparing TP export estimates based on the high frequency composite sampling, we found an underestimation pattern when applying the low frequency sampling/linear interpolation method for both STABLE catchments and an overestimation for the FLASHY catchments (Table 6). This bias was always higher for the low than for the high-LUI catchments (Table 6).

The TP and TDP export from the ~~SUBTR-INT~~FLASHY/high-LUI catchment was higher than in ~~comparison with~~ the other three catchments (Table 5). Moreover, for STABLE streams, the TP export was always higher from the ~~TEMP-EXT~~low-LUI than from the ~~TEMP-INT~~high-LUI catchment (Table 5).

~~When~~Also comparing ~~TP export estimates with the high frequency-composite sampling, we found a pattern of underestimation when applying the LFS-LI method for both TEMP catchments, and an overestimation for the SUBTR catchments (Fig. 5; Table 6). The deviation of exported TP calculated based on COMP sampling and from grab samples using the LFS-LI estimation method was always higher for the extensive than the intensive catchments independent of climate (Table 6). The comparison of exported TP based on COMP sampling and~~with the high frequency composite estimates, the ~~LFS-LI estimation showed that the difference was most pronounced for TP in the TEMP catchments, a pattern that could not be traced in SUBTR (Table 6).~~

~~The~~ C-Q relationships used to calculate exported TP ~~(compared with COMP estimates)~~ produced more accurate results than ~~LFS-LI~~the linear interpolation for the two ~~intensive~~high-LUI catchments, irrespective of ~~the~~ climatic region (~~Fig. 5 and~~ Table 6). The largest and disproportionate deviations of exported TP were obtained ~~by~~when applying the C-Q model to the ~~SUBTR-EXT~~FLASHY/low-LUI catchment compared to ~~COMP~~the high frequency composite sampling estimates (364-400%; ~~Fig. 5 and~~ Table 6).

The ~~field evidence showed a high~~ contribution of PP to exported P~~, percentages~~ was never ~~being~~ lower than 65% of the annual exported TP in the ~~TEMP~~STABLE catchments (Table 5). A contrasting pattern was recorded for ~~SUBTR~~FLASHY streams where the contribution of PP ~~to TP~~ never exceeded 48% of TP, reaching values as low as 13.6% (Table 5). This pattern ~~showing~~of a major contribution of PP in the ~~TEMP~~STABLE catchments ~~is~~ repeats itself in the estimations made with the low frequency instantaneous-grab~~,~~ samples, high frequency composite samples, and flow-weighted concentrations (Fig. ~~6). A~~ 5). We found a tendency ~~of~~to a higher, though rarely significant, dissolved P contribution in streams draining ~~intensively farmed~~high-LUI catchments ~~was found~~ (Fig. ~~6~~5).

The estimated contributions of TP from point sources and diffuse sources indicated that ~~the~~ most of the TP export from the ~~TEMP~~STABLE catchments ~~were derived~~came from diffuse sources~~,~~ as point source contribution from human sources only reached a maximum of 18% of the exported P (Table 5~~). Contrary,~~), but it was still too low to be detectable in ~~SUBTR~~our established C-Q relationships as point sources (Table 4). Contrarily, in the FLASHY catchments point sources dominated the P export~~ contributing,~~ always constituting more than 83% of the exported P, with human sources contributing < ~~10% and~~8%, dairy cattle being the most probable source of the remaining ~~delivered from livestocks~~P (Table 5).

4 Discussion

~~4.1. Importance of climate and hydrology for P dynamics and sources~~

~~A clear difference was found in hydrological regimes between the TEMP and SUBTR climates, being the TEMP more stable than the SUBTR streams, which were classified as flashy (higher R-B Index and lower contribution of baseflow to total flow). Consistently with the climatic characteristics of the catchments investigated, the TEMP catchments having a lower but more evenly distributed rainfall during the year, no extreme rainfall events (> 50 mm day^{-1}) and lower temperature (and therefore low evapotranspiration), and the SUBTR catchments exhibiting an opposite profile. The results of the present work provide insight into the hydrological future for Danish streams given the predicted changes to a more extreme, warmer and wetter climate, and probably flashier streams (Hanssen-Bauer et al., 2005).~~

~~The established point source and diffuse source relationships between TP concentrations and discharge clearly showed that climate and hydrology control TP concentrations in streams (Fig. 4), as expected. Several authors have found similar relationships and used them to characterise P dynamics (e.g. Meyer and Likens, 1979), calculate P sources (e.g. Bowes et al., 2008) or simply to calculate P transport (e.g. Kronvang and Bruhn, 1996).~~

~~According to C-Q relationships, the P dynamics of the streams and catchments studied differed in several ways. Firstly, only the SUBTR catchments showed strong impacts from point sources on stream TP concentrations, an effect that was much more pronounced in the intensive agricultural than in the extensive agricultural catchment. This pattern is consistent with the well-documented reduced influence of P from larger point sources in Europe due to improved wastewater treatment (European Environment Agency, 2005; Kronvang et al., 2005b). Secondly, as to diffuse source-generated TP in streams, the intensive catchments exhibited a more pronounced TP response at low flow in both climates, while the extensive catchments showed a more gradual but sustained increase in TP with increasing flow. When focusing on diffuse source inputs, the resultant P dynamics were similar for the two TEMP and the SUBTR-EXT catchments, while the SUBTR-INT catchment exhibited the highest TP concentrations at comparable flows (Fig. 4). The results agree with Jarvie et al. (2010) as catchments with intensive livestock farming had much higher stream water P loadings (derived from highly consistent diffuse source TP yields) than catchments with arable farming only.~~

~~4.2. Comparison of different phosphorus monitoring schemes~~

~~The performance of different monitoring frequencies and load estimation methods seems to reflect the hydrologic character or flashiness of the investigated streams. Low frequency sampling and interpolation methods are prone to very high uncertainties due to under-representation of high discharge, short-duration events (Defew et al., 2013; Jones et al., 2012; Jordan and Cassidy, 2011; Stelzer and Likens, 2006). However, we did not find a notorious poorer performance of LFS-LI in estimating P exports in flashy SUBTR compared to TEMP streams. On the other hand, the results suggest that climate and hydrological conditions generate a bias in load estimations towards underestimation in TEMP (hydrologically stable streams) and overestimation in SUBTR (flashy streams). This underlines the inadequacy of the method to depict stream dynamics, which is consistent with previous findings (e.g. Jones et al., 2012).~~

~~Furthermore, we detected considerable interaction between sampling frequency and concentration-discharge relationships, and similar to Richards and Holloway (1987), we conclude that loads of substances whose concentration increase during runoff periods (as the TEMP catchments with only diffuse P sources) will be underestimated, while loads of substances whose concentration decrease during runoff periods (as the SUBTR catchments with high contribution from point sources) will be overestimated.~~

~~Additionally, we found signs of interaction between climatic/hydrological and land use factors, such as lower deviation of LFS-LI estimates for TEMP-INT compared to TEMP-EXT catchments. Therefore, land use intensity seems also to be an control factor that has to be considered when planning a sampling strategy to capture P dynamics in streams.~~

~~The concentration-discharge (C-Q) relationships established for the streams appeared as relatively poor in all cases. However, the estimated annual TP export based on the C-Q method produced similar results to those of the LFS-LI method, except for the SUBTR-EXT stream where the C-Q method yielded strongly disproportionate overestimations of P export. The C-Q method also produced close-fitting results like the Comp method for the intensive catchments, irrespective of climate zone.~~

~~4.3. Importance of agricultural intensification on P forms, P dynamics and P sources~~

~~The impact of intensification of agriculture on stream P concentrations and P exports differed between the two contrasting climate zones. Intensification had a significant impact on P concentrations and P export in the SUBTR catchments, with much higher TP, PP, TDP and SRP concentrations and higher P export considering all sampling and load estimation methods considered. Estimated P losses from the SUBTR-INT catchment exceeded the maximum range of 35 comparable Nordic/Baltic micro-catchments studied by Kronvang et al. (2007).~~

~~The difference found between the TEMP-INT and TEMP-EXT catchments, with the extensive catchment showing higher concentrations of TP and PP, can possibly be explained by different processes, including higher bank erosion related to the year-round higher discharge (higher stream power; Laubel et al., 2003), discharge of anaerobic groundwater with high natural SRP concentrations (Kronvang et al., 2007) and deeper soil aeration promoted by tile draining and~~

~~resultant higher binding capacity for surplus P from agricultural production in the intensive catchment (Leinweber et al., 2002).~~

~~Even if the TEMP-EXT stream had similar TP concentrations to the TEMP-INT and SUBTR-EXT streams, the former had a higher TP export as a consequence of higher flow. However, all the calculated annual TP export values were within the range of estimations reported for comparable micro-catchments with grassland-agriculture production (5 km^2; 0.89 to 3.98 kg P ha^{-1} y^{-1}; Campbell et al., 2015), arable (9 and 11 km2; 0.12 to 0.83 kg P ha-1 y-1; Melland et al., 2012) in Ireland, and for Norwegian arable catchments (4.5 and 6.8 km2; 0.5 to 5.8 kg P ha-1 y-1; Kronvang et al., 2005a).~~

~~The contribution of TP from point sources was negligible in both TEMP catchments, but never contributed with less than 83% in the two SUBTR catchments irrespective of land use. These results may be due to an interaction between climate and differences in land use management between the investigated catchments. A higher contribution of dissolved P forms to TP is probably a result of the lack of local treatment of sewage from dairy facilities and is exacerbated by direct cattle access to the stream channel. The joint effect of the two factors can explain that the highest levels of TP, TDP, SRP and SNRP were found in the SUBTR-INT catchment, together with the highest relative importance of dissolved P forms. The direct contribution of phosphorus from local inhabitants was relatively low in SUBTR catchments (4 to 8% of exported P), and slightly higher in TEMP catchments (10 to 18%) but too low to be detectable in our established C-Q relationships as point sources. In the SUBTR-EXT catchment, increased rates of bank erosion, deterioration of buffer strips, direct deposition in water and mobilisation from sediments, likely promoted, among other factors, by cattle activity, could lead to the high proportion of dissolved P in the water (Laubel et al., 2003;Sheffield et al., 1997; Jarvie et al., 2010; Kronvang et al., 2012).~~

~~Despite the level of consistency in the results on the relative magnitude of point sources in the SUBTR catchments, the contribution of diffuse sources to exported P seems to be underestimated by the C-Q model. This is probably a consequence of the underrepresentation of the grab sampling programme during high discharge events (due to inaccessibility to the sites), aspect that would become more relevant as the flashier character increases. Higher frequency data on P concentrations should therefore be derived, perhaps using sequential samplers instead of composite samples; stratifying sample collection in function of the hydrologic status of the system (e.g. baseflow condition, stormflow condition), and changing~~

~~sampling frequency following a strategy of adaptive assessment (i.e. time paced monitoring programs complemented with storm sampling, thereby increasing the number of potentially diffuse-dominated data points at high flows). Whenever possible, flow-weighted sampling has to be implemented, through automatic samplers triggered by increases of flow (Rodríguez-Blanco et al., 2013), water level or rain events, or by means of (expensive) automatized bankside analysers (Campbell et al., 2015). Other strategies, such as passive sampling using flow-proportional samplers (Audet et al., 2014; Jordan et al., 2013) still remain to be evaluated for flashy-warm streams.~~

~~Assumptions of the C-Q model applied for source apportionment of P, such as that all point source inputs are continuous and all diffuse inputs are flow dependent, are probably not fulfilled for the analysed SUBTR catchments due to processes such as sediment mobilisation by cattle trampling and storm flow water running from cattle milking yards.~~

~~Both SUBTR catchments showed a contribution of P from point sources that was not detected in the two TEMP catchments (Fig. 4 and Table 4). This result is possibly linked to the high efficiency of effluent treatment facilities and productive regulations in TEMP catchments which are so far absent in the SUBTR catchments. Moreover, all the cattle have direct access to the stream channel in the SUBTR catchments, which is not the case in the TEMP catchments where fencing off the stream channels is mandatory. The impact on P by cattle accessing the stream channel can be noticeable, arising from trampling (Trimble and Mendel, 1995), bank erosion (Laubel et al., 2003) and direct pollution via excretion (James et al., 2007). Such impact is included in both the point source and diffuse source signal from established C-Q relationships.~~

~~One of the strongest differences between the TEMP and SUBTR catchments was that dissolved P forms dominated over particulate forms in SUBTR streams, irrespective of methods used. The high stream flashiness combined with intensification of agriculture was expected to increase the contribution of particulate P to the TP export. However, this was not the case as TDP dominated the TP export in both SUBTR streams (52-86%). The reason is probably related to farming practices in the SUBTR catchments where cattle have access to the stream channel and tertiary treatment of sewage is lacking. In agreement with previous works (Sharpley and Smith, 1994; Sharpley et al., 1996), we suggest that one of the factors potentially increasing TDP losses to streams is the dominant no-till practices associated with the application of fertilizers over the soil surface, a practice that has become absolutely dominant in Uruguay (Derpsch et al., 2010).~~

~~4.4. Implications to management~~

~~Our work confirms that intensive agricultural systems can be developed without detrimental effects on water quality in streams as long as agricultural production and its various environmental impacts are properly managed, as is the case in, for instance, Denmark (Kronvang et al., 2005a). In contrast, this study clearly proves that today's Uruguayan intensive agricultural production, associated with dairy production, has severe environmental implications for the quality of receiving surface waters where P concentration may markedly exceed biological limits values, contributing to the eutrophication process of downstream reservoirs, as seen elsewhere (Carpenter et al., 1998; Carpenter, 2008; Correll, 1998; Dodds and Welch, 2000; Jeppesen et al., 1999; Moss et al., 1996; Smith et al., 2006).~~

~~In Uruguay, a large part of the population has experienced eutrophication-induced events of bad flavour and odour of drinking water, and the Uruguayan Environment Ministry has therefore introduced an action plan for the protection of environmental quality of surface waters (DINAMA, 2013). The plan includes compulsory effluent treatment in dairy farms. Although it is not currently allowed, re-utilisation of manure as fertilizer could assist in establishing a more closed P cycle in agriculture, contributing both to reducing the extent of P inputs to the streams and to lowering the import and use of mineral fertilizers in catchments (Kronvang et al., 2005a).~~

~~Occurrence of the highest P concentrations during low-flow periods, predominance of total dissolved phosphorus (TDP) and highly reactive forms (SRP) in the SUBTR catchments exacerbate the environmental and sanitary risks associated with eutrophication, particularly when the values frequently reach and strongly exceed the allowed milligram of P per litre. According to the Uruguayan national water quality standards 25 µg P L^{-1} is the maximum allowed, and this will probably be raised to 100 µg P L^{-1} as a result of more than a decade-long negotiations (DINAMA, 2008). The comparatively high levels of P in the SUBTR-INT lead to the question whether nitrogen could become the limiting nutrient here. In this case, nitrogen-fixing primary producers might be favoured, involving environmental and health risks arising from the development of toxic cyanobacteria blooms when or where the residence time of the water becomes sufficiently high.~~

~~Even at low density, cattle activity in the SUBTR-EXT catchment might lead to predominance of dissolved P forms in the stream, reflecting strong point source influence, and dissolved P concentrations that exceeded the national standards in almost half of the grab samplings. To reduce dissolved P, some management plans include fencing off of cattle (Laubel et al., 2003; Sheffield et al., 1997) or ensuring availability of off-stream water sources, which has been shown to reduce the time spent by the cattle in the stream by 92% in Virginia, USA (Sheffield et al., 1997).~~

~~To achieve compliance with water quality targets, long-term efforts to reduce TP/TDP losses to surface waters should probably include other measures than just soil conservation centered on reduction of erosion (Decree 405, 2008). Although they are necessary for soil conservation and contribute to reduce P losses, in the Uruguayan reality probably does not guarantee water quality themselves. Strategies that allow to generate scientific based management actions which maximize agronomic productivity while reducing environmental concerns include the development of catchment models (e.g. SWAT; Gassman et al., 2007), establishment of P budgets (e.g. McKee and Eyre, 2000) and mass balances (e.g. Bowes and House, 2001), evaluation of P surplus (e.g. Wismer et al., 1985) and/or generation a soil P index (e.g. Andersen and Kronvang, 2006). One or more of these approaches should be implemented in Uruguay to ensure best-practice management actions.~~

~~5 Conclusions~~

~~This work comparing two intensive and two extensive agricultural farmed catchments across two climate zones (Danish temperate and Uruguayan subtropical) shows that climatic and hydrological characteristics of catchments expressed as flow responsiveness, exert clear control on catchment and stream TP dynamics, having consequences that are more significant than the outcome of different TP monitoring and export estimation strategies.~~

~~Our results suggest that the lack of environmental regulations of agricultural production has more severe consequences on water quality, than climatic and hydrological differences between the analysed catchments. These consequences includes high TP concentrations (as high as 4436 µg P L^{-1}), P exports (as high as 5.20 kg P.ha^{-1}.year^{-1}), and extremely high proportion of~~

~~dissolved P (as high as 86.4%), as in the case of subtropical intensively farmed catchment studied.~~

~~Investigations of the P dynamics in subtropical streams are at a starting point, and further research and high frequency sampling is needed to investigate and explain the important hydrological pathways linkages between fields and streams, and to achieve an improved knowledge about the magnitude of diffuse sources utilizing new innovative monitoring methods such as automatized P bankside analysers.~~

~~Finally, we believe that calibration of hydrological and phosphorus models on data from the four catchments is needed to allow for a more detailed interpretation of the processes behind the nutrient cycling and dynamics measured.~~

Our results show that climatic and hydrological variability affects the temporal dynamics of P in streams and that low frequency monitoring strategies may fail to adequately capture such dynamics.

As expected, we found strong concordance between climatic characteristics and stream hydrological regimes. Danish catchments have a lower rainfall that is more evenly distributed during the year, with rare or no extreme rainfall events (> 50 mm day^{-1}) and lower temperature (and therefore low evapotranspiration). These conditions resulted in stream flows characterized by higher stability (lower flashiness, R-B index = 0.1-0.3 in our study). In contrast, the Uruguayan streams exhibited a much higher contribution of stormflow to total flow and higher flashiness (R-B index = 0.9-1.3), reflecting the local climate and likely also soil conditions. As reference, reported data in the literature includes daily maximum values of R-B Index reaching 0.43 in mountain streams in Slovakia and Austria (Holko et al., 2011), 0.65 in 30 agricultural dominated catchments in seven Nordic and Baltic countries (Deelstra et al., 2014), 1.01 at 204 stations in Michigan temperate streams (USA) (Fongers, 2012), and 1.32 at 515 stations in catchments located in six US Midwestern States (Baker et al., 2004).

The calculated annual TP export values in all streams fell within the range reported in the literature for comparable micro-catchments, for instance streams with grassland-agriculture production in Ireland (0.89 to 3.98 kg P ha^{-1} y^{-1}; Campbell et al., 2015) Irish streams with catchment farming activities (0.12 to 0.83 kg P ha^{-1} y^{-1}; Melland et al., 2012), and Norwegian streams (0.5 to 5.8 kg P ha^{-1} y^{-1}; Kronvang et al., 2005a). The temporal variability of exported PP followed the hydrological variability in the streams (Fig. 2), but the amount of exported P and PP (the latter likely derived from diffuse sources) did not systematically increase with increasing variability. This was valid for both Uruguayan catchments where the highest and the lowest P loads were exported, and dissolved P forms always predominated over particulate forms. The pattern of P loads exported in relation to conditions of high hydrological variability was thus opposite to our *a priory* expectations (i.e. higher total and particulate P export from diffuse sources in connection with higher flashiness) (Table 5), likely due to other factors related to land use, less input of P with eroded stream bank material due to potential low content of P and maybe also less erosion (Kronvang et al., 2012) and particularly the presence/absence of point sources in the catchments.

Several factors may contribute to the predominance of dissolved P forms in the Uruguayan streams. The direct access of cattle to the stream channels is one of those reasons, being a practice that results in direct manure deposition in the water and trampling, and mobilization from stream bed sediments (James et al., 2007; Jarvie et al., 2010; Kronvang et al., 2012; Laubel et al., 2003; Sheffield et al., 1997; Trimble and Mendel, 1995). In the FLASHY/high-LUI catchment this contribution was exacerbated and further aggravated by the additional effects of the lack of slurry treatment in dairy facilities and the widespread no-till practices associated with the application of fertilizers over the soil surface (Sharpley and Smith, 1994; Sharpley et al., 1996). Some or all of these factors together may explain the high levels of TP, TDP, SRP, and SNRP, exceeding the maximum range found in 35 comparable Nordic⁄Baltic micro-catchments studied by Kronvang et al. (2007).

The contribution of TP from point sources was negligible in both STABLE catchment, but was always higher than 83% in the two FLASHY catchments irrespective of land use intensity. The magnitude of P point sources seems to have a much stronger influence on the hydrochemistry of the Uruguayan stream waters than do hydrological variability and flashiness *per se*. In contrast, the Danish pattern seems to be consistent with the well-documented reduced influence of P from point sources in northern Europe (European Environment Agency, 2005; Kronvang et al., 2005b). Apart from the climatic and hydrological differences between the analyzed catchments, the extremely high TP concentrations (as high as 4,436 µg P L^{-1}), P exports (as high as 5.20 kg P ha^{-1} $year^{-1}$), and the extremely high proportion of dissolved P (as high as 86.4%) estimated for the FLASHY/high-LUI catchment may have implications for the environmental regulations of farming production in Uruguay given the severe deterioration of water quality in our case study. Our results also provide insight into the future behavior of P in northern European temperate streams seen in the context of the predicted change in climate towards more extreme, warmer, and wetter conditions, probably giving flashier streams (Hanssen-Bauer et al., 2005).

The importance of understanding hydrology-driven variations in nutrient discharge will most likely increase in the near future. In our study, the performance of the different monitoring frequencies and P export estimation methods reflected the hydrological character or flashiness of the investigated streams. The performance of the low frequency sampling programs in estimating TP and TDP exports was low in comparison with the high frequency sampling program. Although the performance of the models evaluated with the Nash-Sutcliffe efficiency coefficient was generally low, the C-Q method did not have a comparatively poorer performance.

Although our study was limited to four representative catchments (due to logistic reasons), our results suggest that a clear interaction between climate/hydrology and land use intensity occurred. This was shown by a lower deviation in the exported P estimations of the low frequency sampling for the high-LUI than for the low-LUI catchments. However, the low frequency sampling and linear interpolation method performed equally poor for the FLASHY and the STABLE streams. Our results suggest that climate and hydrological conditions may promote/may yield a bias in P load estimations at low sampling frequency, with a tendency towards underestimation for the hydrologically STABLE streams and overestimation for the FLASHY streams (Table 6). These results are in accordance with the findings of Richards and Holloway (1987) that the loads of P from diffuse sources (whose water concentration increases with runoff) tend to be underestimated, while the loads of P from point sources (whose concentration decreases with runoff) tend to be overestimated. This underlines the potentially high inadequacy of low frequency sampling programs to properly depict stream dynamics, which is consistent with previous findings (e.g. Jones et al., 2012).

Research into P dynamics in subtropical streams is in its initial phase. The expected intensification of agricultural production in many regions of the world, such as in southern South America, highlights the need for appropriate stream monitoring programs. Accurate estimation of P temporal dynamics and exports can help explain the linkages between climate, hydrology, land use, and water quality. Based on our findings, we suggest that the evaluation and use of more accurate monitoring methods, such as automatized flow-proportional water samplers and automatized bankside analysers, should be prioritized whenever is logistically possible. However, it seems particularly relevant in currently flashy systems and also in systems where climate change predictions suggest an increase in stream flashiness.

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Table 1. Site coordinates (datum WGS84), name and catchment size, dominant soils and land use. Danish catchments are part of Gudenå River basin, and the Uruguayan streams are part of the Santa Lucía Chico River basin. Source: (*) World Reference Soil Database classification, European Commission and European Soil Bureau Network (2004); (#) SOTERLAC database, ISRIC Foundation, (www.isric.org). ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use.~~ intensity.

Id	**~~Stream name~~Name**	**Coordinates**	**Dominant soils**	**Land use (area %)**
~~TEMP-EXT~~STABLE low-LUI	Granslev 740 ha	56°17'2"N 9°53'51"E	Haplic Luvisols (*)	Forest (59); arable farming (29); pasture/meadow (7);
~~TEMP-INT~~STABLE high-LUI	Gelbæk 1180 ha	56°13'29"N 9°48'41"E	Gleyic Luvisols (*)	Arable farming (92); forest (2); urban (1); other (5)
~~SUBTR-EXT~~FLASHY low-LUI	Chal-Chal 1880 ha	33°49'31"S 56°16'55"W	Luvic Phaeozem & Eutric	Extensive pasture (~70); arable farming (~30)
~~SUBTR-INT~~FLASHY high-LUI	Pantanoso 840 ha	33°54'13"S 56°00'23"W	Eutric Regosols (#)	Arable farming and dairy farms (90); extensive pasture (7); urban (3)

Table 2. Yearly accumulated rainfall and runoff in mm (direct measures), runoff ratio (percentage of rainfall water exported as runoff~~)~~), and R-B Index (Richards-Baker Index) of flashiness for each monitored year. Stormflow contribution: Percent contribution of ~~base flow~~stormflow to total flow ~~(BF/TF)~~ was estimated for the complete data set~~. TEMP: temperate~~ (2 years). STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use~~ intensity.

	~~TEMP-EXT~~STABLE **low-LUI**	**~~TEMP-INT~~STABLE** **high-LUI**	**~~SUBTR-EXT~~FLASHY** **low-LUI**	**~~SUBTR-INT~~FLASHY** **high-LUI**
Accumulated rainfall 1st year	770 mm	778 mm	1030 mm	1196 mm
Accumulated rainfall 2nd year	756 mm	766 mm	1010 mm	1405 mm
Total accumulated runoff 1st year	515 mm	223 mm	170 mm	235 mm
Total accumulated runoff 2nd year	472 mm	198 mm	294 mm	255 mm
~~runoff~~Runoff ratio 1st year	66.9%	28.6%	16.5%	19.6%
~~runoff~~Runoff ratio 2nd year	62.4%	25.9%	29.1%	18.2%
R-B Index 1st year	0.1	0.3	1.0	1.3
R-B Index 2nd year	0.1	0.3	0.9	1.2
~~BF/TF~~Stormflow contribution (%)	~~88.2~~11.8%	~~63.6~~36.4%	~~39.2~~60.8%	~~29.4~~70.6%

Table 3. ~~Correlations~~Correlation matrices of total (TP), particulate (PP), total dissolved (TDP), soluble reactive (SRP), and soluble non-reactive (SNRP) phosphorus from instantaneous-grab samples. Numeric values represent Spearman rank order correlation and were included only when significant (p ≤ 0.05). ns: non-significant. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use~~ intensity.

	TP	**PP**	**TDP**	**SRP**	**SNRP**
TP	\	0.78	0.75	0.68	0.57
PP	0.81	\	ns	ns	ns
TDP	ns	-0.31	\	0.93	0.70
SRP	ns	ns	0.86	\	0.44
SNRP	ns	-0.34	0.56	ns	\

~~TEMP-~~

~~TEMP-EXT~~STABLE/low-LUI

	TP	**PP**	**TDP**	**SRP**	**SNRP**
TP	\	0.63	0.90	0.67	0.84
PP	0.80	\	ns	ns	0.41
TDP	0.58	ns	\	0.83	0.81
SRP	ns	ns	0.56	\	0.40
SNRP	ns	ns	0.41	-0.37	\

~~SUBTR-~~

~~SUBTR-EXT~~FLASHY/low-LUI

Table 4. Estimated parameters for the load apportionment model fitted for each stream. RSS: residual sum of squares. NSC: Nash-Sutcliffe model efficiency coefficient. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use~~ intensity.

Source	**Parameter**	**~~TEMP-EXT~~STABLE low-LUI**	**~~TEMP-INT~~STABLE high-LUI**	**~~SUBTR-EXT~~FLASHY low-LUI**	**~~SUBTR-INT~~FLASHY high-LUI**
Point	**A**	0	0	1915	2550
	B	-	-	0.140	0.501
Diffuse	**C**	7.145	20.677	3.658	399.000
	D	1.58	1.40	1.64	1.15
Global	**RSS (10^3)**	42.5	460	253	36362
	NSC	0.25	0.12	0.12	0.10

Table 5. Alternative estimations of annual total phosphorus (TP) and total dissolved phosphorus (TDP) exported from the four catchments expressed as kg P ha^{-1} year^{-1}. Estimation strategies: “COMP”: ~~Composite~~high frequency composite sampling~~ strategy~~. “LFS-LI”: Low frequency sampling and linear interpolation~~ strategy~~. “C-Q~~”~~”: low frequency sampling and concentration-discharge relationships ~~applying the load apportionment model.~~interpolation. % PP~~.~~: percentage TP exported in particulate form. “% hs” under COMP represents the percentage of annual exported P from human sources. “% ps” under ~~CQ~~C-Q represents the percentage of the total annual exported load from point sources *sensu* the model. ~~TEMP: temperate, SUBTR: subtropical. EXT: extensive or~~STABLE: low flashiness Danish streams; FLASHY: Uruguayan flashy streams; low and high-LUI: low and high land use intensity~~ land use; INT: intensive land use~~.

	~~TEMP-EXT~~	**~~TEMP-INT~~STABLE/low-LUI**			**~~SUBTR-EXT~~STABLE/high-LUI**			**~~SUBTR-INT~~FLASHY/low-LUI**			**FLASHY/high-LUI**		
		COMP	**LFS-LI**	**C-Q**	**COMP**	**LFS-LI**	**C-Q**	**COMP**	**LFS-LI**	**C-Q**	**COMP**	**LFS-LI**	**C-Q**
1st year	**TP**	1.09 (12.5% hs)	0.64	0.61 (0% ps)	0.34 (10.5% hs)	0.29	0.34 (0% ps)	0.13 (6.9% hs)	0.25	0.52 (89.7% ps)	2.28 (7.5% hs)	2.36	2.86 (83.1% ps)
	TDP	0.17	0.20	\	0.08	0.10	\	0.08	0.13	\	1.97	1.83	\
	% PP.	84.4	68.6	\	76.5	66.9	\	38.5	48	\	13.6	22.5	\
2nd year	**TP**	0.74 (17.7% hs)	0.47	0.54 (0% ps)	0.35 (10.4% hs)	0.25	0.33 (0% ps)	0.25 (3.6% hs)	0.27	0.91 (83,6% ps)	5.20 (4.2% hs)	5.76	5.19 (86.5% ps)
	TDP	0.10	0.11	\	0.07	0.06	\	0.14	0.21	\	4.07	4.7	\

% PP.	86.5	76.4	\	80	76.9	\	44	22.2	\	21.7	18.4	\

Table 6. The percentages ~~resumes~~summarizes the relative fit of alternative estimation methods in relation to the reference annually exported load estimated by the composite sampling programme (~~as reference).~~see Table 5 for references). 100% represents the same annual P exported in kg~~.~~ ha^{-1}~~.~~ year^{-1}. Values below 100% (~~italic) represents~~italics) represent underestimation (less than 90%) relative to the estimation of composite sampling. Values over 110% (bold) ~~represents~~represent overestimation relative to the estimation of composite sampling. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use~~ intensity.

		~~TEMP-EXT~~STABLE **low-LUI**		**~~TEMP-INT~~STABLE** **high-LUI**		**~~SUBTR-EXT~~FLASHY** **low-LUI**		**~~SUBTR-INT~~FLASHY** **high-LUI**	
		LFS-LI	**C-Q**	**LFS-LI**	**C-Q**	**LFS-LI**	**C-Q**	**LFS-LI**	**C-Q**
1st year	**TP**	*58.7%*	*56.0%*	*85.3%*	100.0%	**192.3%**	**400.0%**	103.5%	**125.4%**
	TDP	**117.6%**	\	**125.0%**	\	**162.5%**	\	92.9%	\
2nd year	**TP**	*63.5%*	*73.0%*	*71.4%*	94.3%	108.0%	**364.0%**	**110.8%**	99.8%
	TDP	110.0%	\	*85.7%*	\	**150.0%**	\	**115.5%**	\

Fig. 1. Left: Mean daily air temperature variation for ~~one~~a temperate (~~TEMP~~Danish) and ~~one~~a subtropical (~~SUBTR~~Uruguayan) catchment ~~for~~in 2011. Right: boxplots of the same data. The boundary of the box closest to zero indicates the 25th percentile, a line within the box marks the median, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentiles. Black dots display outliers.

Fig. 2. Accumulated daily rainfall and specific runoff hydrographs for the four monitored streams. For each variable a fixed scale was used to aid visual comparison. ~~Flashiness of subtropical streams is clear.~~ R-B Index (Richards-Baker Index) of flashiness.

Fig. 3. From top to bottom: temporal variation of total (TP), particulate (PP), total dissolved (TDP~~)~~), and soluble reactive phosphorus (SRP) concentrations from ~~gram~~grab samples for all the monitored catchments. Log$_{10}$ scale was selected on the vertical axe to improve ~~visualisation.~~vizualisation. The phosphorus concentration is always expressed as µg P L^{-1}. Right: boxplots are based on the same data. Letters A, B, and C are used ~~for~~to display statistical groups ~~following~~according *post hoc* paired comparison analysis. The boundary of the box closest to zero indicates the 25th percentile, a line within the box marks the median, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentiles. Black dots display outliers. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use.~~ intensity.

Fig. 4. Scatterplots of total phosphorus (TP) ~~concentration~~concentrations of instantaneous-grab samples relative to discharge from the four streams. The dots connected by lines ~~represents~~represent the predicted values according to the load apportionment model (see Table 4). For ~~temperate~~STABLE catchments, only the diffuse originated term of the model is included. Lowest graphs display the predicted TP concentration of all catchments from point (left) and diffuse sources (right) for a range of 0 to 1000 L s^{-1}. ~~Noted~~Note the Log_{10} scale for TP. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use.~~ intensity.

Fig. 5. ~~Accumulated fortnightly total phosphorus (TP) exported loads estimated low frequency instantaneous-grab sampling and linear interpolation (LFS-LI), concentration-discharge relationships by applying the load apportionment model (C-Q) and by high frequency-composite sampling (COMP). TEMP: temperate streams; SUBTR: subtropical streams; EXT: extensive land use; INT: intensive land use.~~

~~Fig. 6.~~ Contribution of particulate phosphorus (PP) to exported total phosphorus (TP). Left boxplots are based on ~~grab concentrations~~instantaneous-grab sampling and linear interpolation estimation, centre boxplots are based on composite data, and the right boxplot on flow-weighted concentrations. Letters A, B, and C are used ~~for~~to display statistical groups ~~following~~according *post hoc* paired comparison analysis. The boundary of the box closest to zero indicates the 25th percentile, a line within the box marks the median, and the boundary of the box farthest from zero indicates the 75th percentile. Whiskers above and below the box indicate the 90th and 10th percentiles. ~~TEMP: temperate~~STABLE: low flashiness Danish streams; ~~SUBTR: subtropical~~FLASHY: Uruguayan flashy streams; ~~EXT: extensive~~low and high-LUI: low and high land use~~; INT: intensive land use.~~ intensity.

Fig.1

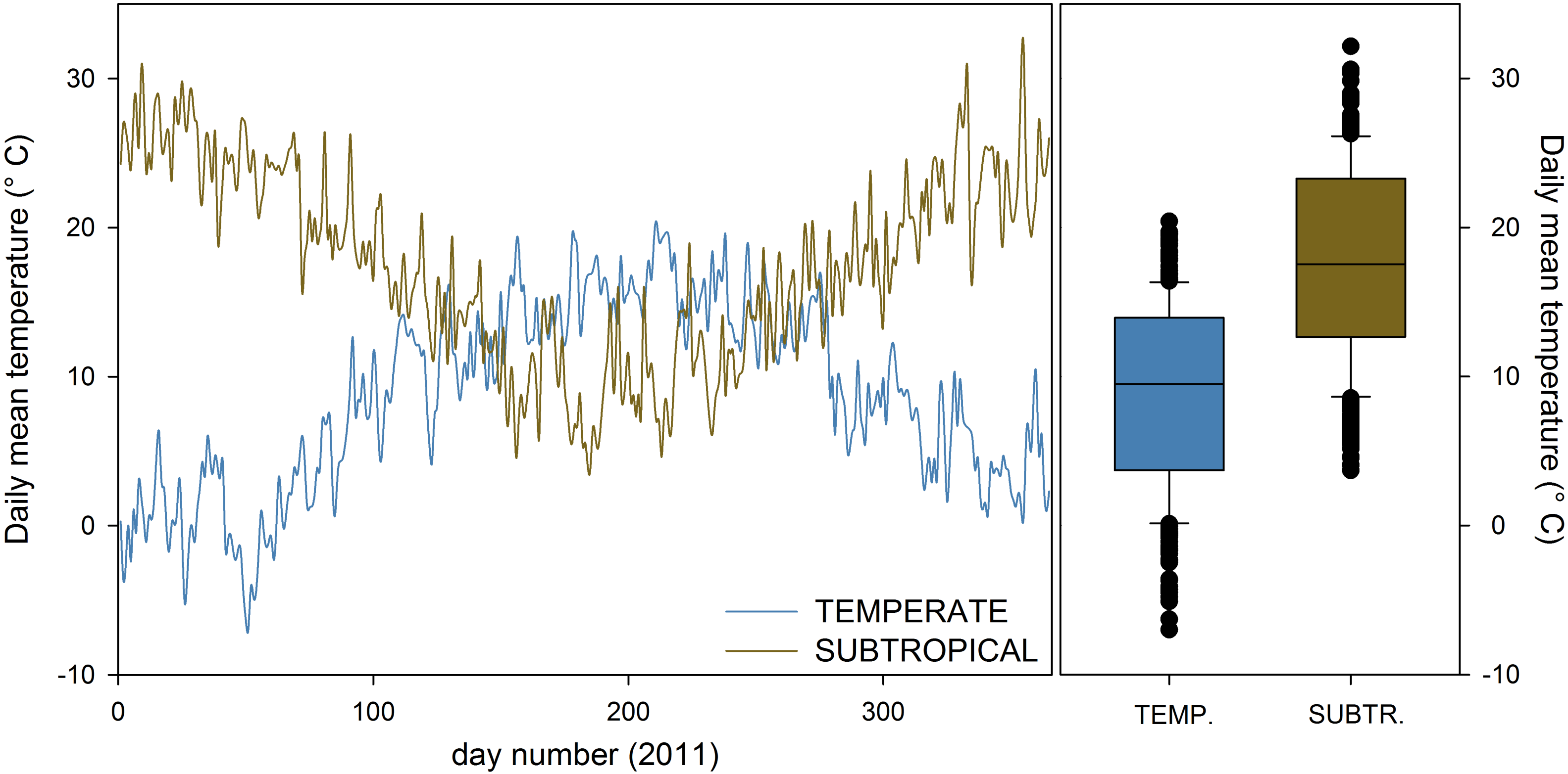

Daily mean temperature (° C)
30
20
10
0
-10
0
100
200
300
day number (2011)
TEMPERATE
SUBTROPICAL
TEMP.
SUBTR.
Daily mean temperature (° C)

Fig. 2

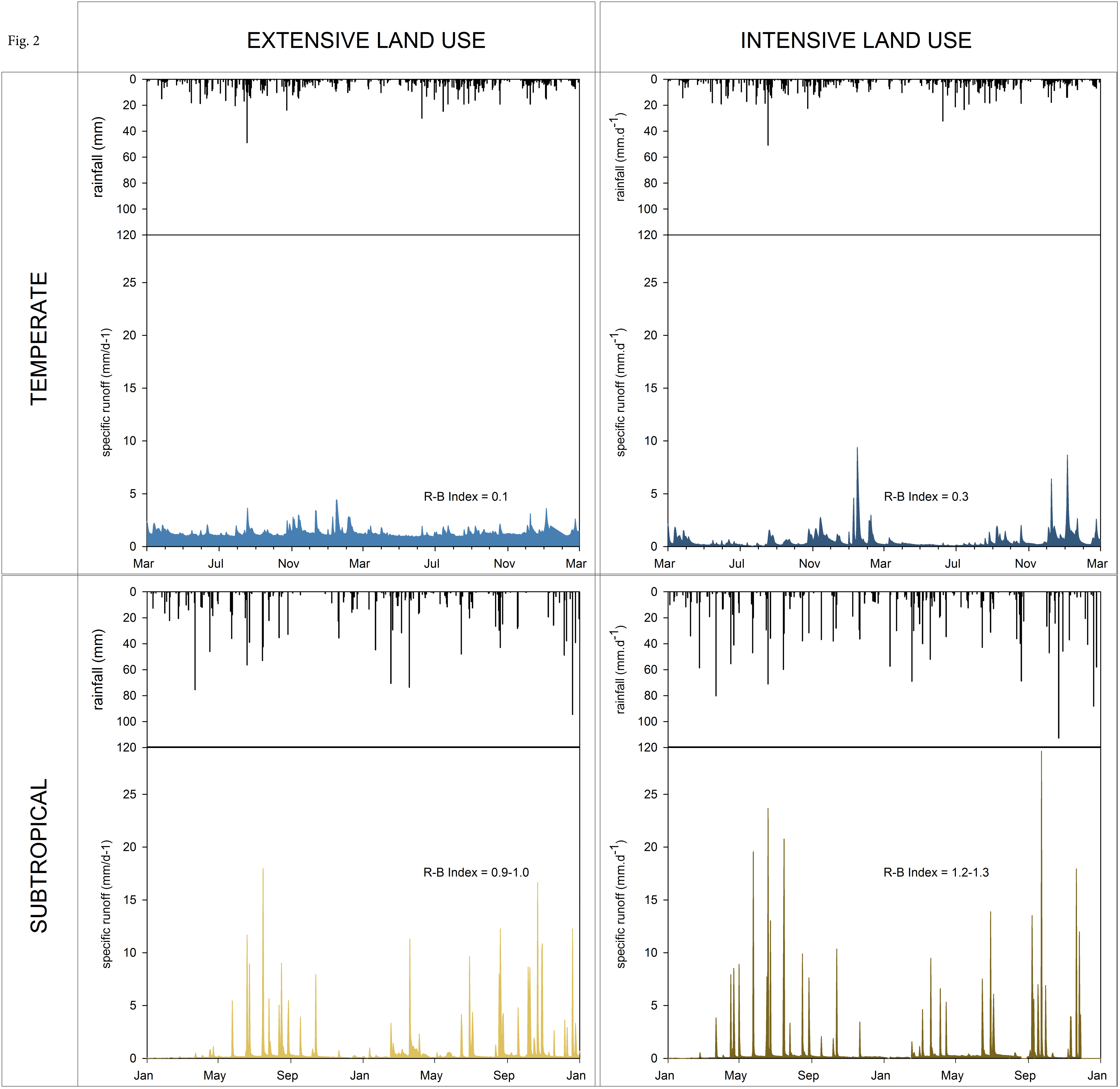
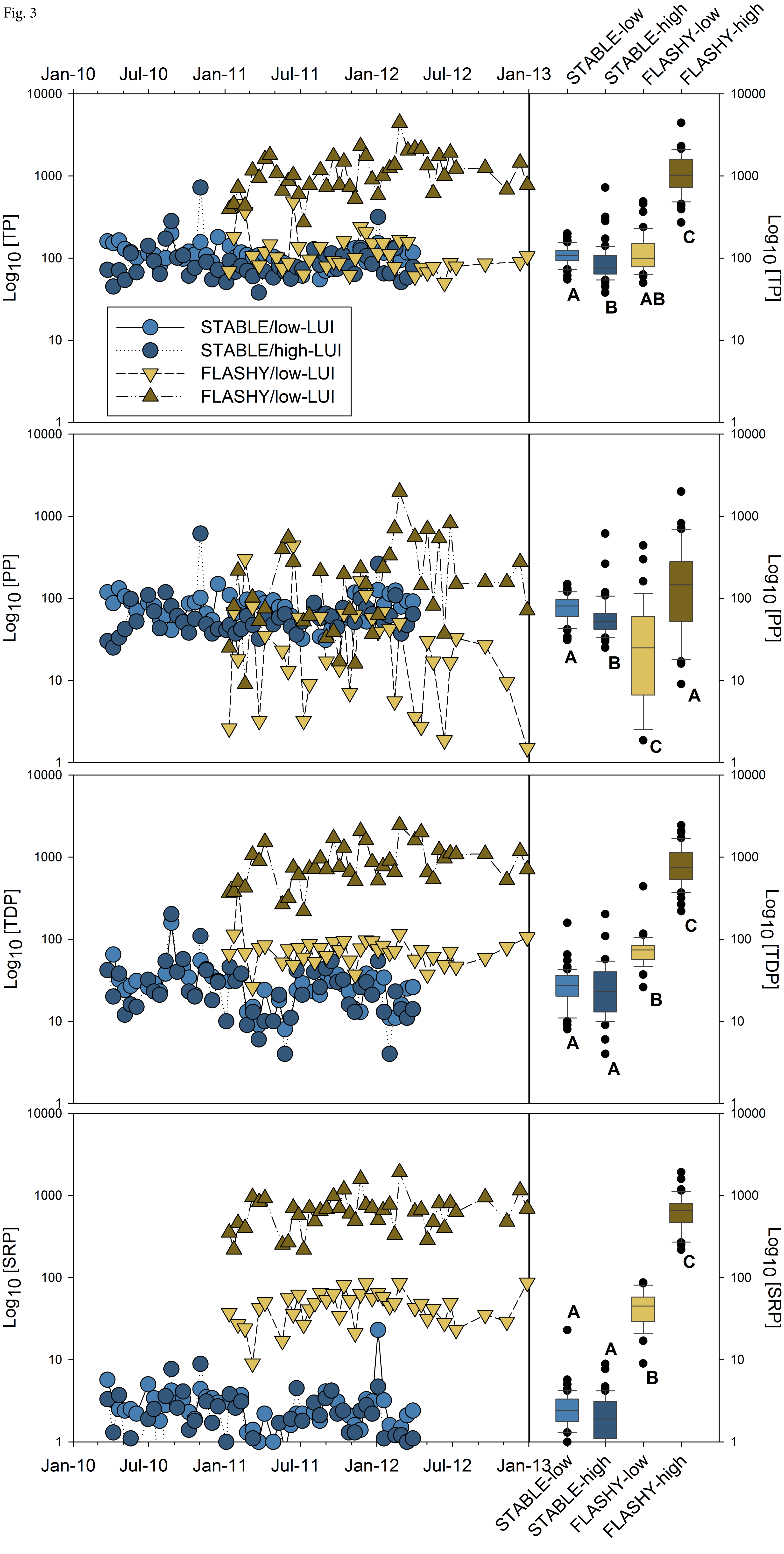

EXTENSIVE LAND USE
INTENSIVE LAND USE
TEMPERATE
SUBTROPICAL
rainfall (mm)
rainfall (mm.d-1)
specific runoff (mm/d-1)
specific runoff (mm.d-1)
0
20
40
60
80
100
120
25
20
15
10
5
0
R-B Index = 0.1
R-B Index = 0.3
R-B Index = 0.9-1.0
R-B Index = 1.2-1.3
Mar
Jul
Nov
Mar
Jul
Nov
Mar
Jan
May
Sep
Jan
May
Sep
Jan

Fig. 3



Jan-10
Jul-10
Jan-11
Jul-11
Jan-12
Jul-12
Jan-13
STABLE-low
STABLE-high
FLASHY-low
FLASHY-high
10000
1000
100
10
1
Log10 [TP]
Log10 [PP]
Log10 [TDP]
Log10 [SRP]
STABLE/low-LUI
STABLE/high-LUI
FLASHY/low-LUI
FLASHY/low-LUI
A
B
AB
C
A
B
C
A
A
A
B
C
A
A
B
C

Fig. 4

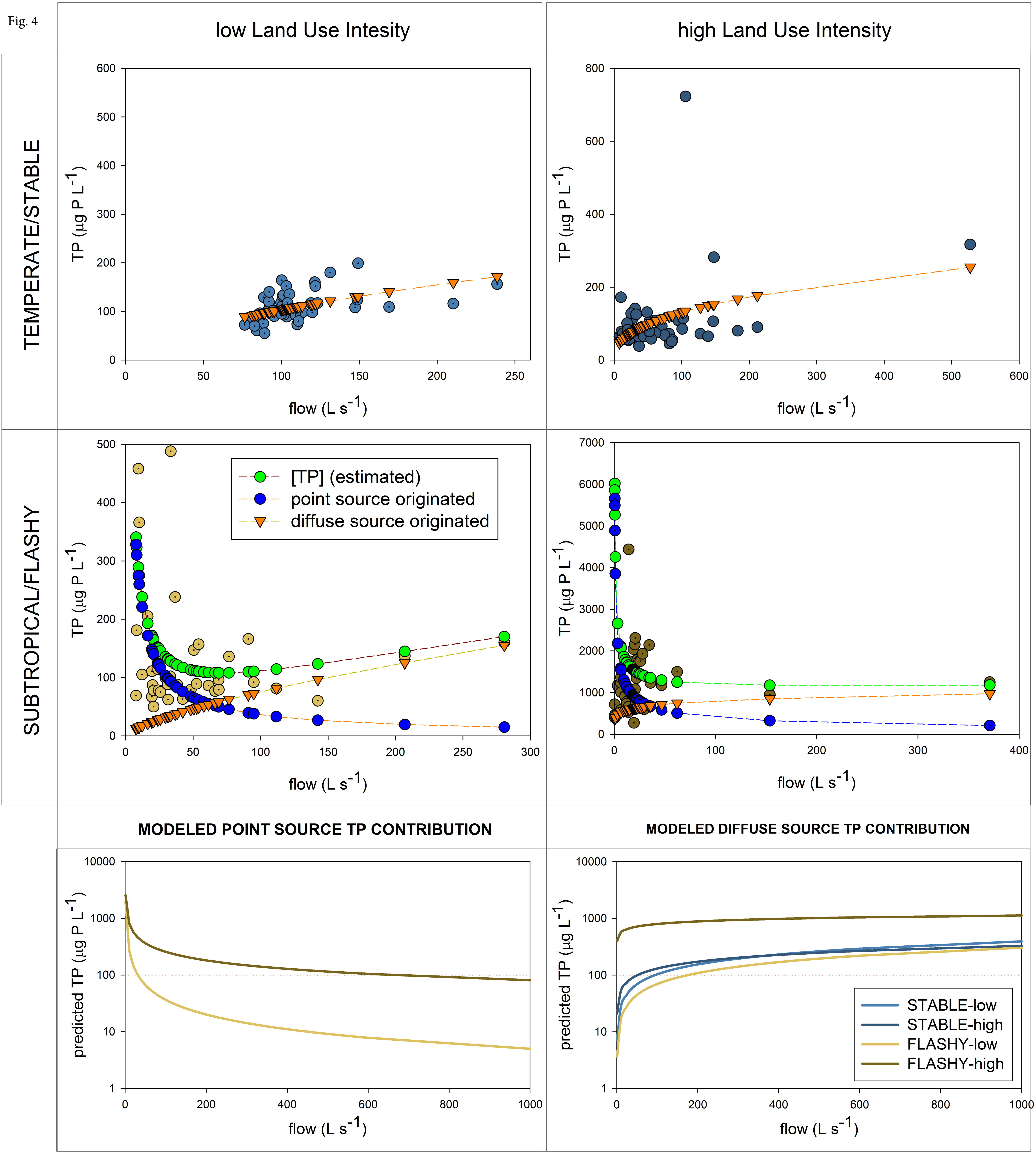
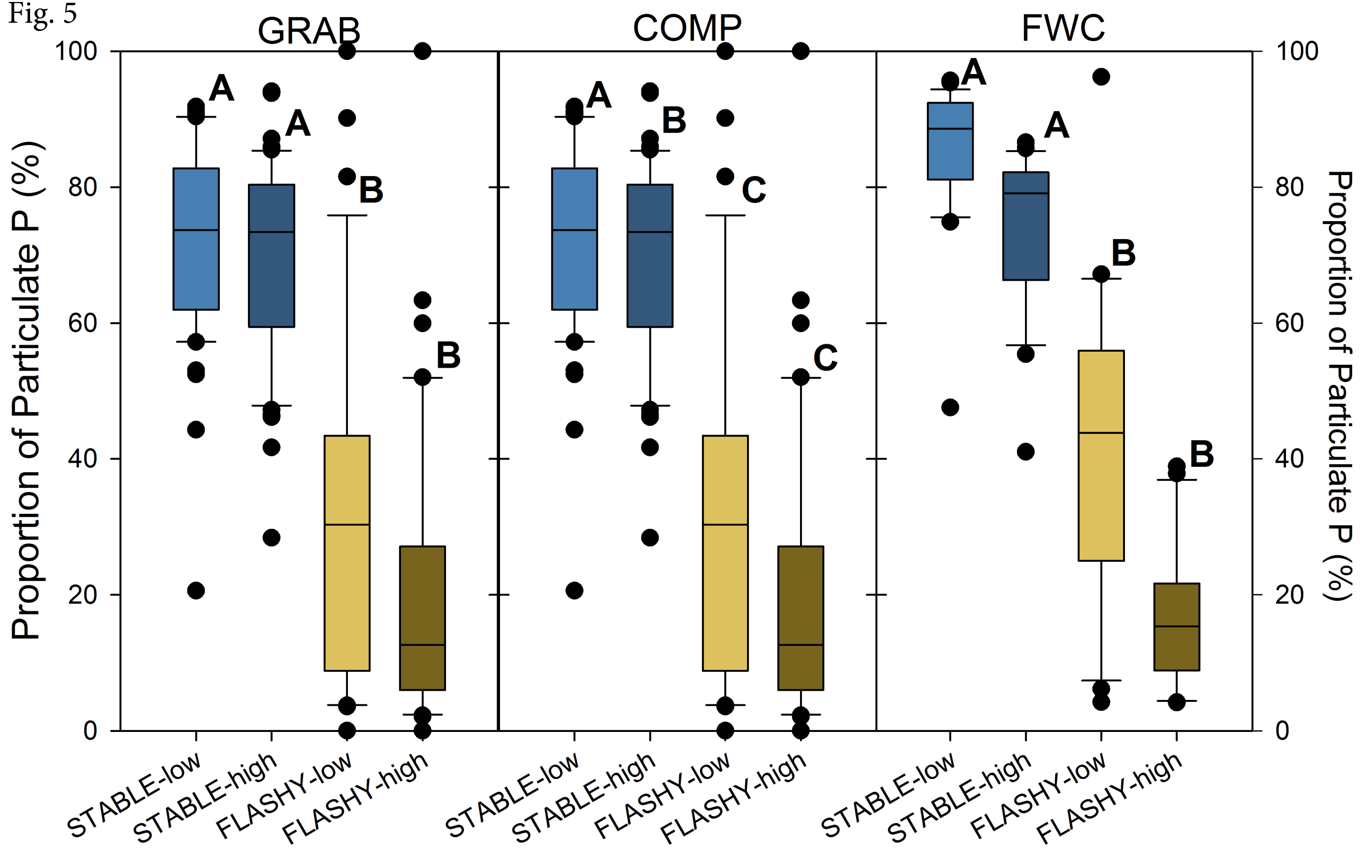
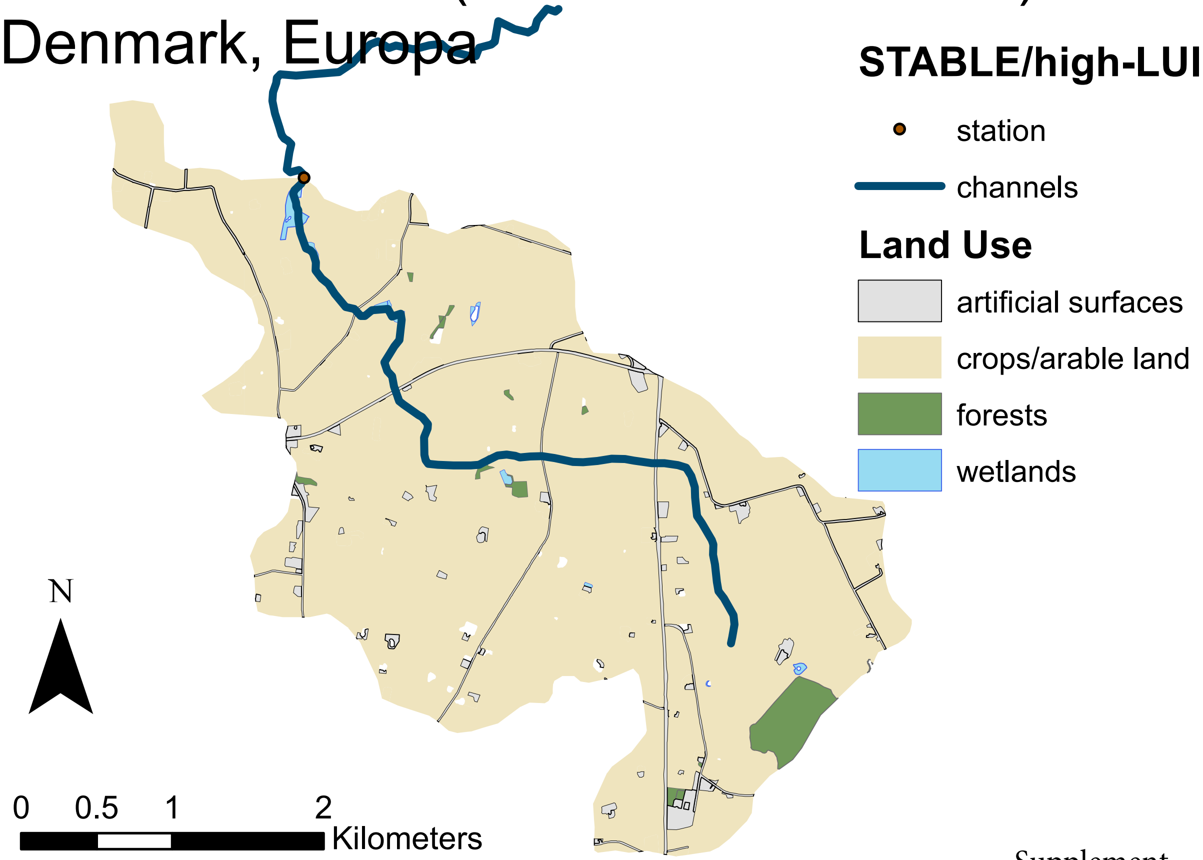

low Land Use Intesity
high Land Use Intensity
TEMPERATE/STABLE
SUBTROPICAL/FLASHY
TP (µg P L-1)
flow (L s-1)
0
50
100
150
200
250
300
400
500
600
700
800
1000
2000
3000
4000
5000
6000
7000
[TP] (estimated)
point source originated
diffuse source originated
MODELED POINT SOURCE TP CONTRIBUTION
MODELED DIFFUSE SOURCE TP CONTRIBUTION
predicted TP (µg P L-1)
predicted TP (µg L-1)
1
10
100
1000
10000
STABLE-low
STABLE-high
FLASHY-low
FLASHY-high

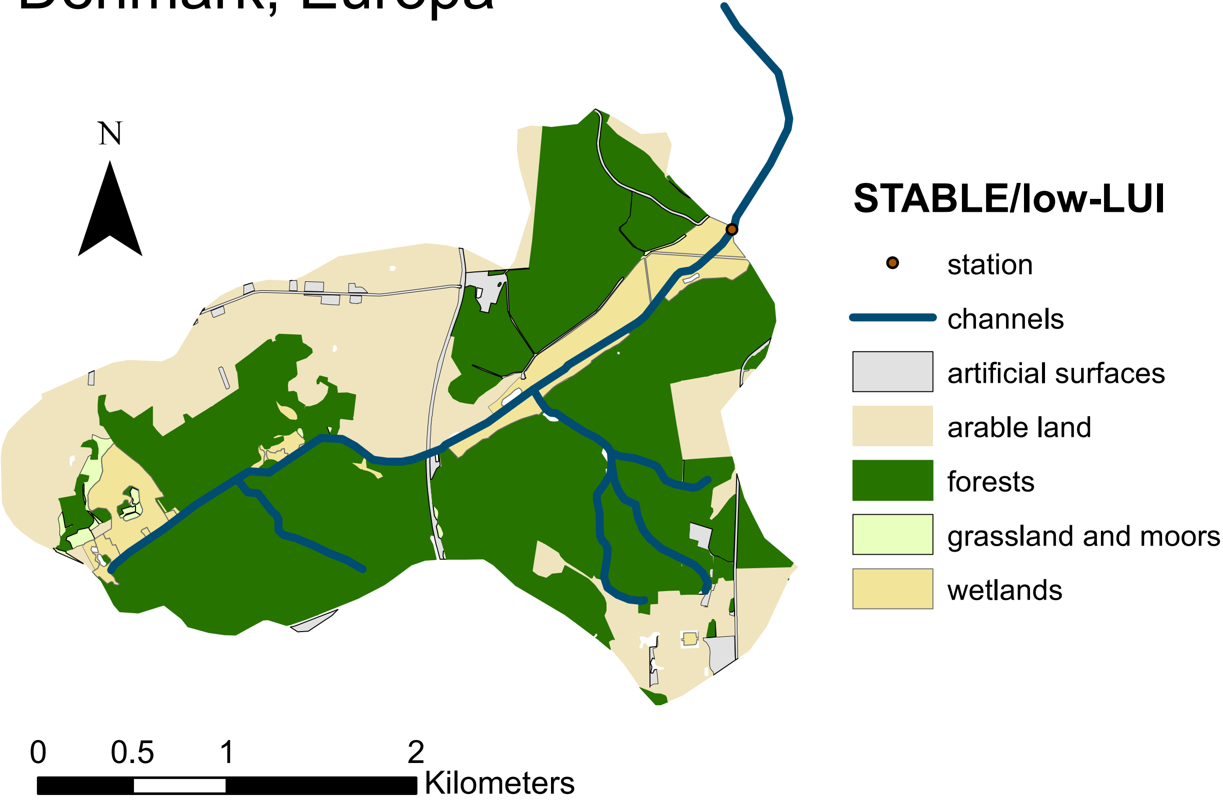
Fig. 5


GRAB
COMP
FWC
Proportion of Particulate P (%)
Proportion of Particulate P (%)
0
20
40
60
80
100
A
A
B
B
A
B
C
C
A
A
B
B
STABLE-low
STABLE-high
FLASHY-low
FLASHY-high

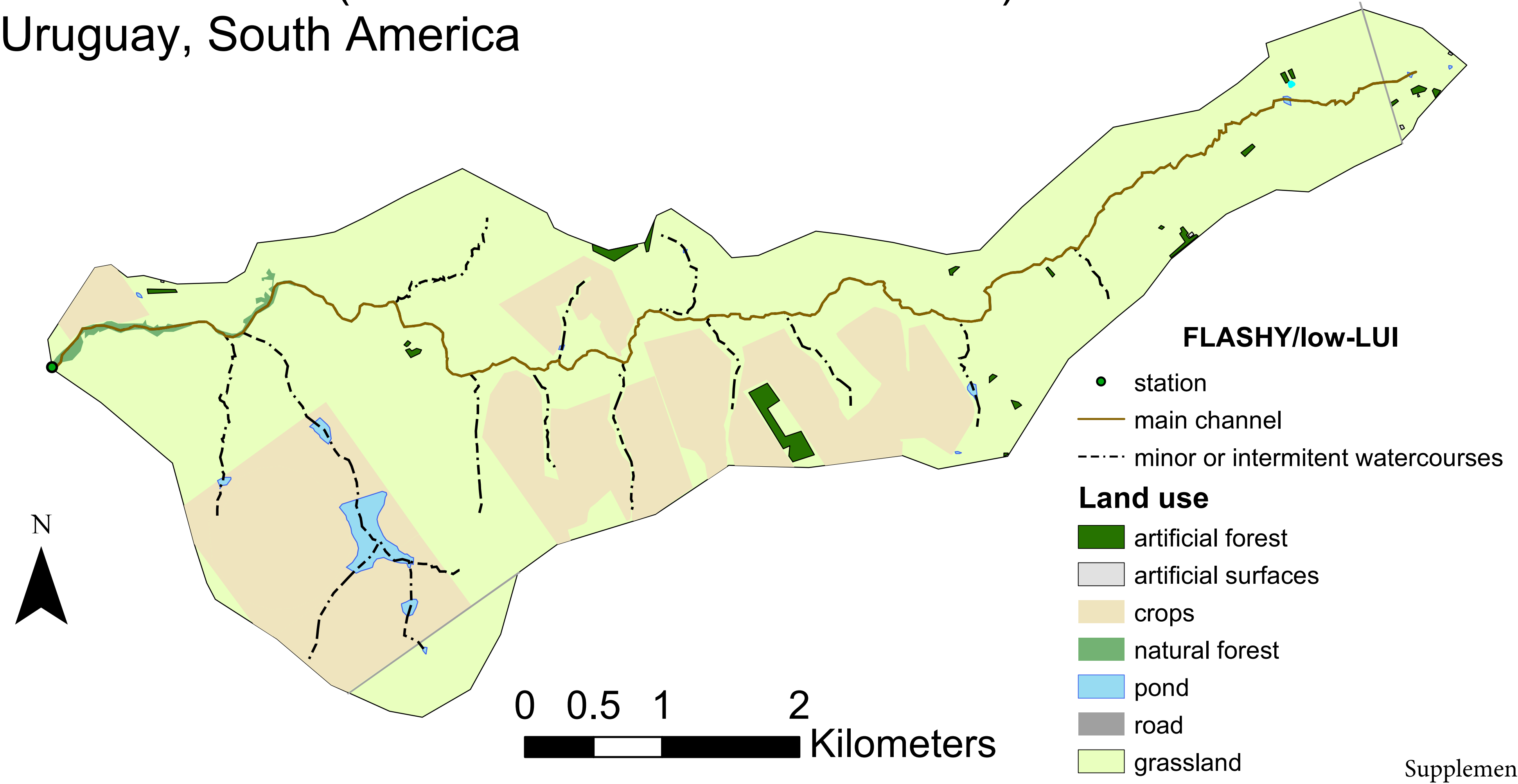
Gelbaek stream (Gudenå River Basin) Denmark, Europa

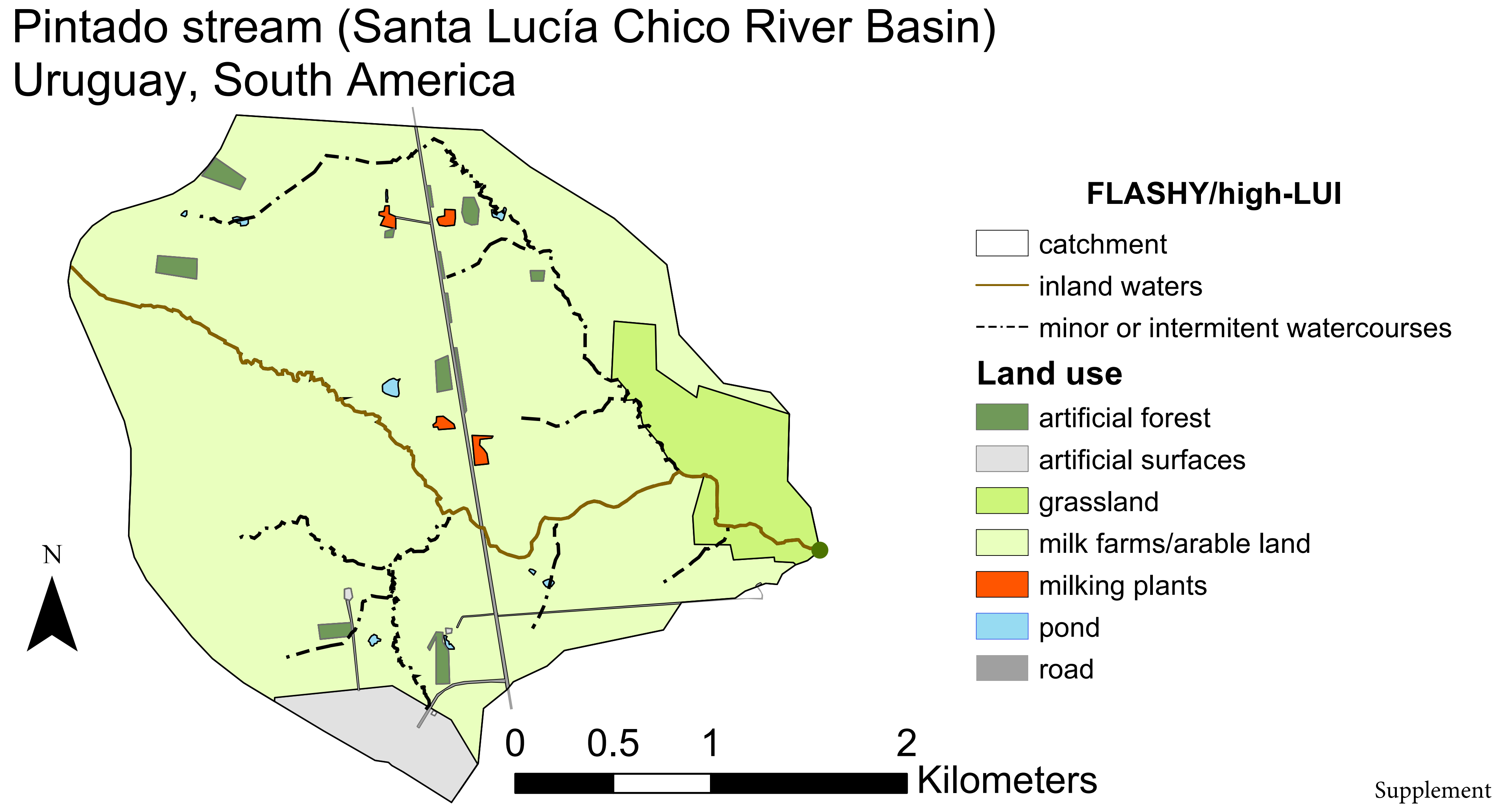


Granslev stream (Gudenå River Basin) Denmark, Europa



Pintado stream (Santa Lucía Chico River Basin)
Uruguay, South America




Pintado stream (Santa Lucía Chico River Basin)
Uruguay, South America
FLASHY/high-LUI
catchment
inland waters
minor or intermitent watercourses
Land use
artificial forest
artificial surfaces
grassland
milk farms/arable land
milking plants
pond
road
N
0
0.5
1
2
Kilometers

Supplement