

Trend detection in water-quality and load time-series from agricultural catchments of Yläneenjoki and Pyhäjoki, SW Finland

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Long-term water quality (WQ) and load trends were studied in the catchments of Yläneenjoki and Pyhäjoki in SW Finland, where agricultural water protection measures have been implemented since the mid-1990s. A univariate Mann-Kendall (MK) trend test, a multivariate Mann-Kendall (MMK) trend test, and a multivariate Mann-Kendall trend test applied to the WQ data flow-normalized by a semi-parametric model (FN-MMK) were used. The results of all methods were similar when the nutrient concentration data were used, but they differed when using the nutrient load data. The FN-MMK test was intended to detect trends caused by anthropogenic impact. In the clay-soil-dominated catchment of Yläneenjoki there were increasing trends in the concentrations and loads of total nitrogen, nitrate-nitrogen, dissolved reactive phosphorus, and a decreasing trend in suspended solid concentrations. However, no increasing or decreasing trends were detected for the majority of the concentrations or loads in sand-soil-dominated catchment of Pyhäjoki. This suggests different responses to comparable environmental and anthropogenic pressures in these two river basins.

Introduction

Deterioration of freshwater quality, particularly in areas with intensive agricultural practices, required large public and private funding to implement legislative framework and water protection measures in several countries (Volk *et al.* 2009). Climate and land-use changes are the most important stressors affecting freshwater quantity and quality. Simultaneous changes in both stressors during the last decades make it difficult to distinguish their relative effects on stream flow and water quality, respectively

(Tomer and Schilling 2009). Long-term water quality (WQ) data are important for detection of temporal and spatial changes in the condition of aquatic environments, for inference of factors affecting it and for effectiveness assessment of water protection measures (Hirsch *et al.* 2010). In Finland, water quality monitoring started in the early 1960s, mainly for lakes (Ekholm and Mitikka 2006) and some small pilot catchments (Rekolainen *et al.* 1991), and it has increased since 2000 to meet the requirements of the EU water directives (Niemi *et al.* 2001). Consequently, long-term data sets are

available for several sites. However, water quality monitoring data are usually highly variable: they are affected by human activities, and hydrological and weather conditions before and during the sampling as well as by sample treatment (Rode and Suhr 2007). Long-term water quality data are usually non-normally distributed and non-stationary (sudden changes), show seasonal cycle, contain missing values and temporal autocorrelation and have censored values (i.e. values below the detection limits) (Hirsch *et al.* 1982). Such characteristics induce high uncertainty in trend detection and interpretation and restrict the use of parametric methods in time-series trend analysis.

The non-parametric Mann-Kendall (MK) method (Yu *et al.* 1993) has been widely used to detect trends in environmental data. This method is based on ranks of the observations rather than their measured values, hence it is less sensitive to extreme values and not affected by the data distribution (Hamed 2008). The MK method assumes a monotonic trend and is affected by the length of the analysed period (Yu *et al.* 1993). However, the power of the MK test increases when the sample size is large (Chebana *et al.* 2013). Hirsch and Slack (1984) extended the MK trend test into a multivariate MK (MMK) test for data having seasonality, each season being an independent variable. Based on the MMK test, a further development of trend analysis for multivariate data sets, with a semi-parametric regression model, was introduced by Wahlin and Grimvall (2008). The semi-parametric regression model aims to fit a trend surface function to multiple time-series data to detect the presence of a common trend and remove randomness. By including covariates in the model — e.g. water flow — the model reduces the variability induced by the changes in flow (flow-normalization) and detects trends due to other causes than regular hydrologic fluctuations (Wahlin 2008). Other regression-based and statistical learning techniques (i.e. neural network) are also used to normalize water quality data with respect to a covariate. However, the suitability of these methods depends on the relationship between the response variable and the covariate (Libiseller 2004).

In this study, we examined long-term trends of nutrient concentrations and loads in two

catchments located in SW Finland. Univariate and multivariate trend analyses were applied to raw and flow-normalized WQ data to gain insight into the WQ response to agricultural water protection measures. The specific objectives of our study were: (1) to assess the capability of univariate and multivariate time-series analyses to detect trends in river water quality, and (2) to detect and compare long-term trends in river water quality and loads between two different agricultural catchments.

Material and methods

Study area

The study catchments, Yläneenjoki (231 km²) and Pyhäjoki (74 km²), are the most important inflows to Säkylän Pyhäjärvi (Fig. 1). In the Yläneenjoki catchment, agricultural areas cover 28%, built-up areas 4%, forests 64% and mire and peat areas 3%, while in the Pyhäjoki catchment, agricultural areas cover 24%, built-up areas 4% and forests 71%. In these catchments, agriculture together with animal husbandry, accounts approximately for 55% and 39% of the total nutrient loading to Säkylän Pyhäjärvi, respectively (Bärlund and Kirkkala 2008). In the Yläneenjoki catchment, most of the agricultural areas in the river valleys are located on clay soils, while in the Pyhäjoki catchment, agricultural areas are mostly located on sandy soils. In both catchments, the majority of agricultural fields are sub-drained (e.g. Tattari *et al.* 2009). Since 1995, nearly all farmers in the catchment have committed themselves to the European Union's (EU) agri-environmental programme to implement basic water protection measures. In addition, more intensive catchment management practices such as buffer zones, sedimentation ponds and wetlands have been introduced (Ventelä *et al.* 2011). Different methods of soil erosion control have been implemented, e.g. minimum tillage. In the Yläneenjoki catchment, arable lands, decreasing tillage or non-tillage methods increased from 17% to 39% in 2000–2010 (i.e. ploughing decreased from 83% to 61%) as a result of the national MYTVAS follow-up by agri-environmental programme

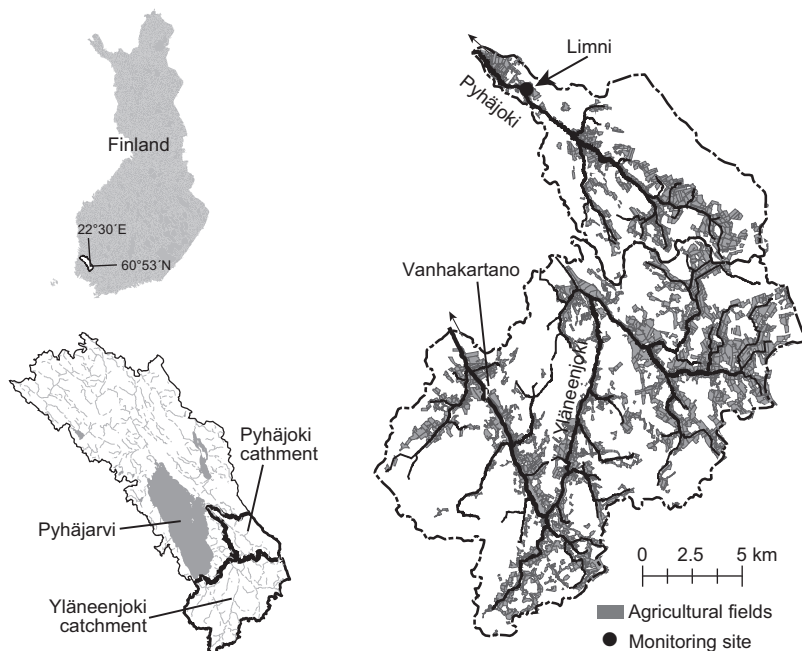


Fig. 1. Location of the study catchments in SW Finland.

(Aakkula and Leppänen 2014). In addition, three lime-sand filters have been established in the Yläneenjoki area, to test their nutrient-removal performance (Kirkkala *et al.* 2012).

The long-term average annual precipitation in the study area is 630 mm, of which approximately 11% is snow (Tattari *et al.* 2009). The average water flows (1980–2013) in the Yläneenjoki and Pyhäjoki are $1.97 \text{ m}^3 \text{ s}^{-1}$ and $0.69 \text{ m}^3 \text{ s}^{-1}$, respectively. The base flow indices (BFI, Institute of Hydrology 1980) in 1980–2013 were 0.37 and 0.53 in Yläneenjoki and Pyhäjoki, respectively. A typical pattern of a hydrological year consists of two periods of high flow (April and October–December) and two periods of low flow (May–September and January–March) (Ventelä *et al.* 2011), but very high flows may also occur during mild winters (Koskiahio *et al.* 2010).

Water quality data

The OIVA service of the Finnish Environment Institute (SYKE) (www.ymparisto.fi/oiva [in Finnish]) was used to obtain long-term water discharge (Q) and water quality (WQ) data. The following WQ variables or nutrients were

selected: total phosphorus (P_{tot}), dissolved reactive phosphorus (DRP), total nitrogen (N_{tot}), ammonium-nitrogen ($\text{NH}_4\text{-N}$) and nitrate-nitrogen ($\text{NO}_3\text{-N}$) concentrations. We also estimated the concentrations of organic nitrogen (ON) by subtracting inorganic N from N_{tot} ($N_{\text{tot}} - \text{NH}_4\text{-N} - \text{NO}_3\text{-N}$), and particulate phosphorus (PP) by subtracting DRP concentration from P_{tot} . For the Yläneenjoki, N_{tot} and P_{tot} data were available for a period of 34 years (1980–2013), while $\text{NO}_3\text{-N}$, DRP, ON, PP and total suspended solids (TSS) data were available for the period 1991–2013. For the Pyhäjoki, consistent data for all the selected WQ variables were available from 1995 to 2013. In both rivers, the sampling was carried out mainly during the high-flow periods: once a week in spring and autumn, every second week in summer, and once a month in winter. Daily flow data for both rivers have been available since the early 1970s.

Time-series analysis

First, long-term trend analysis was carried out by using a univariate Mann-Kendall (MK) test and Sen's slope estimator to find patterns (Yu *et al.* 1993) in annual averages of nutrient concentra-

tions and total annual nutrient loads. Second, a multivariate MK (MMK) test (Hirsch *et al.* 1982) was applied to monthly measured time series of nutrient concentrations and loads. A semi-parametric model (Wahlin and Grimvall 2008) was used to analyse trend patterns in WQ time series of multiple months and to flow-normalize WQ data. Third, the MMK test was applied to flow-normalized data of nutrient concentrations and loads (FN-MMK). In addition, to study the trends in time series of individual months, a univariate MK trend test was applied to the flow-normalized data of nutrient concentrations and loads.

In a semi-parametric model, a regression trend surface function is fitted to time-series vectors, $\mathbf{y}_t = (\mathbf{y}_t^{(1)}, \dots, \mathbf{y}_t^{(m)})^T$, $t = 1, \dots, n$. The n interval assumed for data observations define the m dimension of time series vectors. The model aims to decompose the observed data into: (i) a deterministic surface component, (ii) a regression component with the impact of the covariates, and (iii) random errors (Wahlin 2008). For the seasonal data, the model assumes the following general form:

$$\mathbf{y}_t^{(j)} = \boldsymbol{\alpha}_t^{(j)} + \sum_{i=1}^p \mathbf{b}_i^{(j)} \left[\mathbf{x}_{it}^{(j)} - E(\mathbf{x}_{it}^{(j)}) \right] + \boldsymbol{\varepsilon}_t^{(j)}, \quad (1)$$

$$j = 1, \dots, m, t = 1, \dots, n$$

where $\mathbf{y}_t^{(j)}$ is the observed response for the t th month of the j th year; $\boldsymbol{\alpha}_t^{(j)}$ is the sequence of intercept vectors $\boldsymbol{\alpha}_t = (\boldsymbol{\alpha}_t^{(1)}, \dots, \boldsymbol{\alpha}_t^{(m)})^T$, $t = 1, \dots, n$ representing a deterministic temporal trend; \mathbf{x}_{it} is the matrix of p covariate vectors; E denotes when the covariates are equal to their expectations; $\boldsymbol{\beta}_i^{(j)}$ is the matrix of time-independent coefficients of regression; $\boldsymbol{\varepsilon}_t^{(j)}$ ($j = 1, \dots, m, t = 1, \dots, n$) is the error term.

The model parameters were estimated using a penalized least-squares technique on the intercepts, which is determined by cross-validation or customized smoother. The smoothing over time (years) was similar for all variants, but the smoothing across vector components differ. The algorithm to perform the semiparametric model is implemented in an MS Excel-based package *Multitrend* (Wahlin 2008). Different types of smoothing factors are implemented in the package, depending on the nature of time series. For

the monthly data, when there was a relationship between the observations for adjacent months, a sequential smoothing approach was used (Wahlin 2008). The degree of uncertainty was analysed by resampling the model residuals by a bootstrap approach (Wahlin and Grimvall 2008).

Then, the data normalization with respect to covariate was obtained by the expression:

$$\tilde{\mathbf{y}}_t^{(j)} = \mathbf{y}_t^{(j)} + \sum_{i=1}^p \hat{\boldsymbol{\beta}}_i^{(j)} (\mathbf{x}_{it}^{(j)} - \bar{\mathbf{x}}_i^{(j)}), \quad (2)$$

$$j = 1, \dots, m, t = 1, \dots, n$$

where $\hat{\boldsymbol{\beta}}_i^{(j)}$ is the regression coefficient for the j th component of the i th covariate. A detailed mathematical description of the methods is given in Wahlin (2008).

Nutrient loads were estimated by smoothing and interpolating the measured concentration data with a non-parametric locally-weighted scatterplot smoothing function (*LOWESS*). Then, monthly and annual loads were calculated by summing up products of daily concentrations and flow values (Hussian *et al.* 2004).

A multivariate Mann-Kendall (MKK) test was performed with the *Kendall* package (McLeod 2015) in R (R Core team 2015).

Results

General statistics

The water-quality variables were non-normally distributed (Kolmogorov-Smirnov one-sample test), and they differed significantly between Yläneenjoki and Pyhäjoki (Table 1). In both rivers, TSS concentrations were strongly correlated with flow ($r = 0.79$ and 0.70 , respectively). The concentrations of N_{tot} and $\text{NO}_3\text{-N}$ in both rivers, and ON in Pyhäjoki, correlated moderately with flow (Table 2). Concentrations of PP and P_{tot} in Pyhäjoki, and of PP in Yläneenjoki correlated weakly with flow. DRP concentration did not correlate with flow in both rivers, and no correlations were found between ON and P_{tot} concentrations and flow in Yläneenjoki (Table 2). However, all estimated nutrient loads were very strongly correlated with monthly flow in both rivers ($r > 0.90$) (Table 2).

The correlation strength between nutrient concentrations and flow varied among the months (see Appendix). Overall, TSS correlated strongly with flow in both rivers in most of the months. Also N_{tot} , NO_3 and ON correlated strongly with flow in Pyhäjoki in most of the months, except in April when the correlations were moderate. In Yläneenjoki, N_{tot} , NO_3 and ON correlated moderately with flow during most of the months, with exception of March–May when NO_3 did not correlate with flow. No correlation between DRP and flow was found in any of the rivers in most of the months. Furthermore, in Pyhäjoki, P_{tot} and PP did not correlate with flow in January, February and April, and the existing correlations in other months varied from strong (September) to moderate (the rest of the months). Similarly, in Yläneenjoki, no correlation between P_{tot} and flow, and between PP and flow, was found in January–March and August–September, and the

correlations between these variables during other months were mostly low.

Trend analysis

In general, all the trend analysis methods produced the same results when the nutrient concentration data were used; the results, however, differed in case of the nutrient load data, particularly for Yläneenjoki (Tables 3 and 4). Trends were studied by smoothing the measured and flow-normalized monthly nutrient concentration and load data with the semi-parametric model (Figs. 2 and 3). The flow-normalization performed well for all nutrient loads and their inter-annual variations were reduced (Fig. 3), but it was less effective for most of the nutrient concentrations. The flow-normalized concentrations and the raw data

Table 1. Statistical parameters of the water quality variable concentrations (mg l^{-1}) and flow ($\text{m}^3 \text{s}^{-1}$) and a comparison of the means (Wilcoxon's test).

	Yläneenjoki			Pyhäjoki			Wilcoxon's test	
	Period	<i>n</i>	Mean \pm SD	Period	<i>n</i>	Mean \pm SD	<i>Z</i>	<i>p</i>
N_{tot}	1980–2013	935	1.96 \pm 1.07	1995–2013	512	1.69 \pm 1.15	-17.418	< 0.001
$\text{NO}_3\text{-N}$	1991–2013	601	1.23 \pm 1.07	1995–2013	471	1.08 \pm 1.03	-16.839	< 0.001
ON	1991–2013	524	0.75 \pm 0.37	1995–2013	427	0.57 \pm 0.29	-4.072	< 0.001
P_{tot}	1980–2013	950	0.11 \pm 0.05	1995–2013	513	0.07 \pm 0.04	-17.353	< 0.001
DRP	1991–2013	633	0.02 \pm 0.02	1995–2013	419	0.02 \pm 0.01	-20.208	< 0.001
PP	1991–2013	618	0.09 \pm 0.05	1995–2013	419	0.05 \pm 0.03	-10.388	< 0.001
TSS	1991–2013	703	37.08 \pm 31.66	1995–2013	503	16.66 \pm 16.94	-16.959	< 0.001
Flow	1980–2013	12202	1.97 \pm 3.32	1980–2013	12464	0.69 \pm 0.87	-62.390	< 0.001

Table 2. Spearman's correlations (r_s) between daily nutrient concentrations (mg l^{-1}) and flow ($\text{m}^3 \text{s}^{-1}$), as well as between nutrient loads (t month^{-1}) and flow ($\text{m}^3 \text{month}^{-1}$); values set in boldface indicate existing correlations.

	Yläneenjoki						Pyhäjoki					
	Concentrations			Loads			Concentrations			Loads		
	r_s	<i>p</i>	<i>n</i>	r_s	<i>p</i>	<i>n</i>	r_s	<i>p</i>	<i>n</i>	r_s	<i>p</i>	<i>n</i>
N_{tot}	0.56	< 0.001	935	0.95	< 0.001	408	0.67	< 0.001	512	0.96	< 0.001	208
$\text{NO}_3\text{-N}$	0.60	< 0.001	601	0.95	< 0.001	276	0.64	< 0.001	471	0.92	< 0.001	208
ON	0.10	0.046	524	0.98	< 0.001	276	0.61	< 0.001	427	0.96	< 0.001	200
P_{tot}	0.17	< 0.001	950	0.98	< 0.001	408	0.30	< 0.001	513	0.95	< 0.001	208
DRP	0	0.941	633	0.97	< 0.001	276	-0.06	0.202	418	0.90	< 0.001	192
PP	0.28	< 0.001	618	0.97	< 0.001	276	0.42	< 0.001	419	0.94	< 0.001	192
TSS	0.79	< 0.001	703	0.97	< 0.001	276	0.70	< 0.001	503	0.93	< 0.001	207

Table 3. Trends in nutrient concentrations and loads in Yläneenjoki. MK = univariate Man-Kendall test, MMK = multivariate Man-Kendall test, and FN-MMK = flow-normalized data by semi-parametric regression model followed by multivariate Man-Kendall test; τ = Mann Kendall tau, n = the number of observations. Trend analysis for N_{tot} and P_{tot} was carried out for the years 1980–2013 and for the rest of water-quality variables for 1991–2013. p values set in boldface indicate significant trends.

	Nutrient concentrations (mg l ⁻¹)												Nutrient loads (t yr ⁻¹)												
	MK				MKK				FN-MMK				MK				MMK				FN-MMK				
	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	
N_{tot}	0.42	< 0.001	33	0.15	< 0.001	370	0.24	< 0.001	0.24	< 0.001	370	0.19	0.109	33	-0.03	0.436	370	0.28	< 0.001	0.28	< 0.001	370	0.18	< 0.001	257
$NO_3\text{-N}$	0.17	0.267	22	0.10	0.052	257	0.18	< 0.001	0.18	< 0.001	257	0.08	0.634	22	-0.05	0.293	257	0.18	< 0.001	0.18	< 0.001	257	0.07	0.093	243
ON	-0.07	0.673	22	0.03	0.551	243	0.07	0.114	0.07	0.114	243	-0.07	0.673	22	-0.10	0.050	243	0.07	0.093	0.07	0.093	243	0.07	0.054	372
P_{tot}	0.11	0.374	33	-0.01	0.734	372	0.07	0.056	0.07	0.056	372	-0.12	0.328	33	-0.11	0.002	372	0.07	0.054	0.07	0.054	372	0.31	< 0.001	250
DRP	0.59	< 0.001	22	0.37	< 0.001	250	0.39	< 0.001	0.39	< 0.001	250	0.12	0.428	22	-0.05	0.228	250	0.31	< 0.001	0.31	< 0.001	250	0.02	0.583	248
PP	0	0.980	22	-0.06	0.171	248	0	0.924	0	0.924	248	-0.12	0.428	22	-0.14	0.001	248	0.02	0.583	0.02	0.583	248	-0.07	0.097	260
TSS	-0.39	0.009	22	-0.27	< 0.001	260	-0.24	< 0.001	-0.24	< 0.001	260	-0.28	0.065	22	-0.18	< 0.001	260	-0.07	0.097	-0.07	0.097	260	-0.14	0.583	248

Table 4. Trends in nutrient concentrations and loads in Pynhájoki. Trend analysis was carried out for the years 1995–2013. For symbol explanations see Table 2.

	Nutrient concentrations (mg l ⁻¹)												Nutrient loads (t yr ⁻¹)												
	MK				MKK				FN-MMK				MK				MMK				FN-MMK				
	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	τ	ρ	n	p	
N_{tot}	0.12	0.495	19	0.03	0.541	198	0.07	0.209	0.07	0.209	198	0.17	0.327	19	0.02	0.733	198	-0.01	0.890	-0.01	0.890	198	0.01	0.805	203
$NO_3\text{-N}$	0.14	0.448	19	0.03	0.613	203	0.03	0.549	0.03	0.549	203	0.17	0.327	19	0.02	0.715	203	0.01	0.805	0.01	0.805	203	-0.02	0.758	190
ON	-0.03	0.880	19	0.05	0.395	190	0	0.971	0	0.971	190	0.10	0.576	19	0.03	0.562	190	-0.02	0.758	-0.02	0.758	190	0	0.991	198
P_{tot}	0.09	0.624	19	0.01	0.805	198	0.03	0.613	0.03	0.613	198	0.09	0.624	19	0	0.991	198	-0.03	0.534	-0.03	0.534	198	0.11	0.050	187
DRP	0.26	0.149	19	0.03	0.627	187	0.08	0.138	0.08	0.138	187	0.10	0.576	19	0.03	0.627	187	0.11	0.050	0.11	0.050	187	0.08	0.167	187
PP	-0.07	0.705	19	-0.01	0.908	187	-0.02	0.720	-0.02	0.720	187	0.09	0.624	19	0	0.939	187	0.08	0.167	0.08	0.167	187	-0.14	0.010	202
TSS	-0.22	0.225	19	-0.12	0.050	202	-0.13	0.010	-0.13	0.010	202	0.10	0.576	19	-0.06	0.251	202	-0.14	0.010	-0.14	0.010	202	-0.07	0.097	260

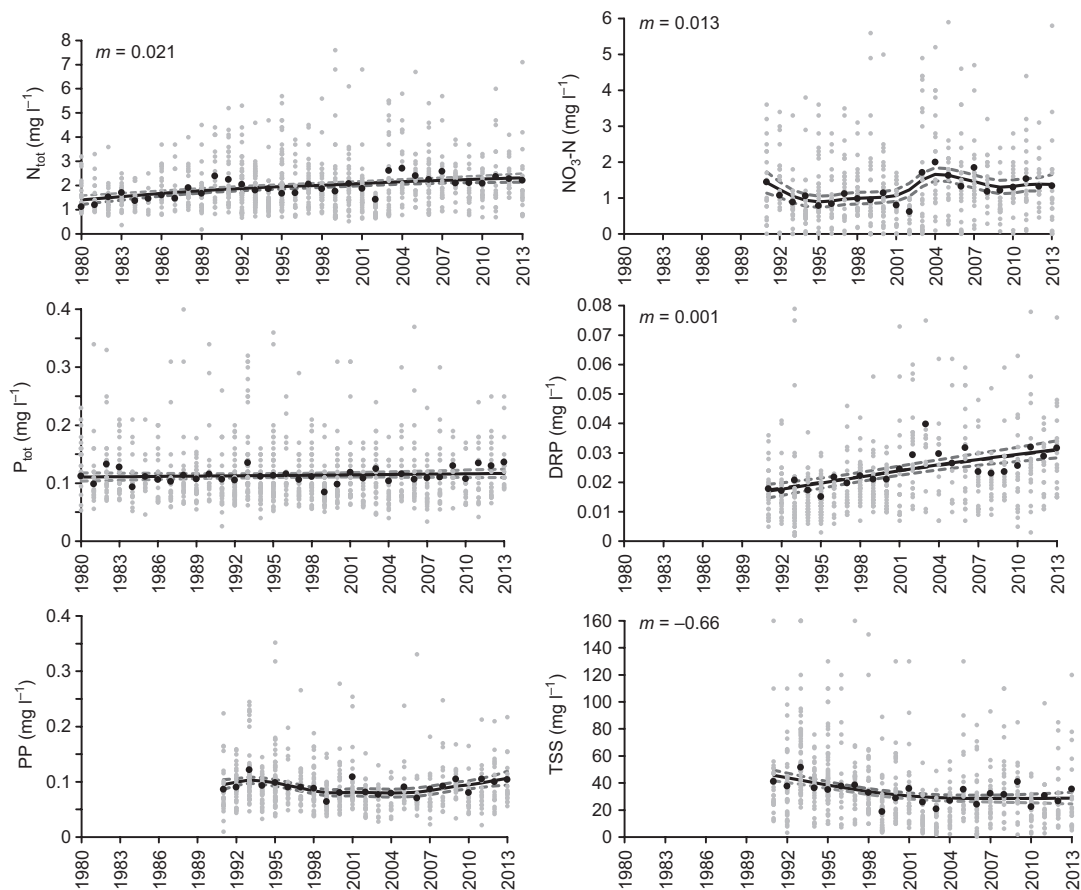


Fig. 2. Trends in the flow-normalized nutrient concentrations (mg l^{-1}) in Yläneenjoki. m denotes the rate of change per unit for significant trends as evaluated by the multivariate Mann-Kendall test. Solid black lines are the semi-parametric regression trends and dashed lines are the 95% confidence limits. Grey dots represent the measured concentrations and black dots are the flow-normalized annual average concentrations.

produced almost similar results. In Yläneenjoki, a significant increasing trend was detected for N_{tot} and DRP concentrations with all three trend tests (Table 3). Additionally, a significant increasing trend in $\text{NO}_3\text{-N}$ was detected with the FN-MMK method. The trends in TSS concentrations detected with all three tests were significant and decreasing. Also significant decreasing trends were found in loads of P_{tot} , PP and TSS using the MMK method, and by applying the FN-MMK trend test, significant increasing trends were found in N_{tot} , $\text{NO}_3\text{-N}$ and DRP (Table 3). Furthermore, a decreasing trend in the river flow was found by using the MMK test ($\tau = -0.13$, $p = 0.001$; see Fig. 4). In Pyhäjoki, significant decreasing trends were

found only in TSS concentration and load with the FN-MMK trend test. No trends were detected in the other WQ variables, loads nor flows with any of the used trend analysis methods (Table 4).

Trend analysis of monthly time-series of nutrient concentrations and loads in Yläneenjoki, was carried out using the univariate MK test and flow-normalized concentration and load data. Long-term significant increasing trends in N_{tot} concentrations were detected in January, May, July, November and December (Table 5). There were also significant increasing trends in the $\text{NO}_3\text{-N}$ concentrations in May–August, and in the DRP concentrations in most parts of the year, except in February, May and December (Table 5). The trends in TSS concentrations were

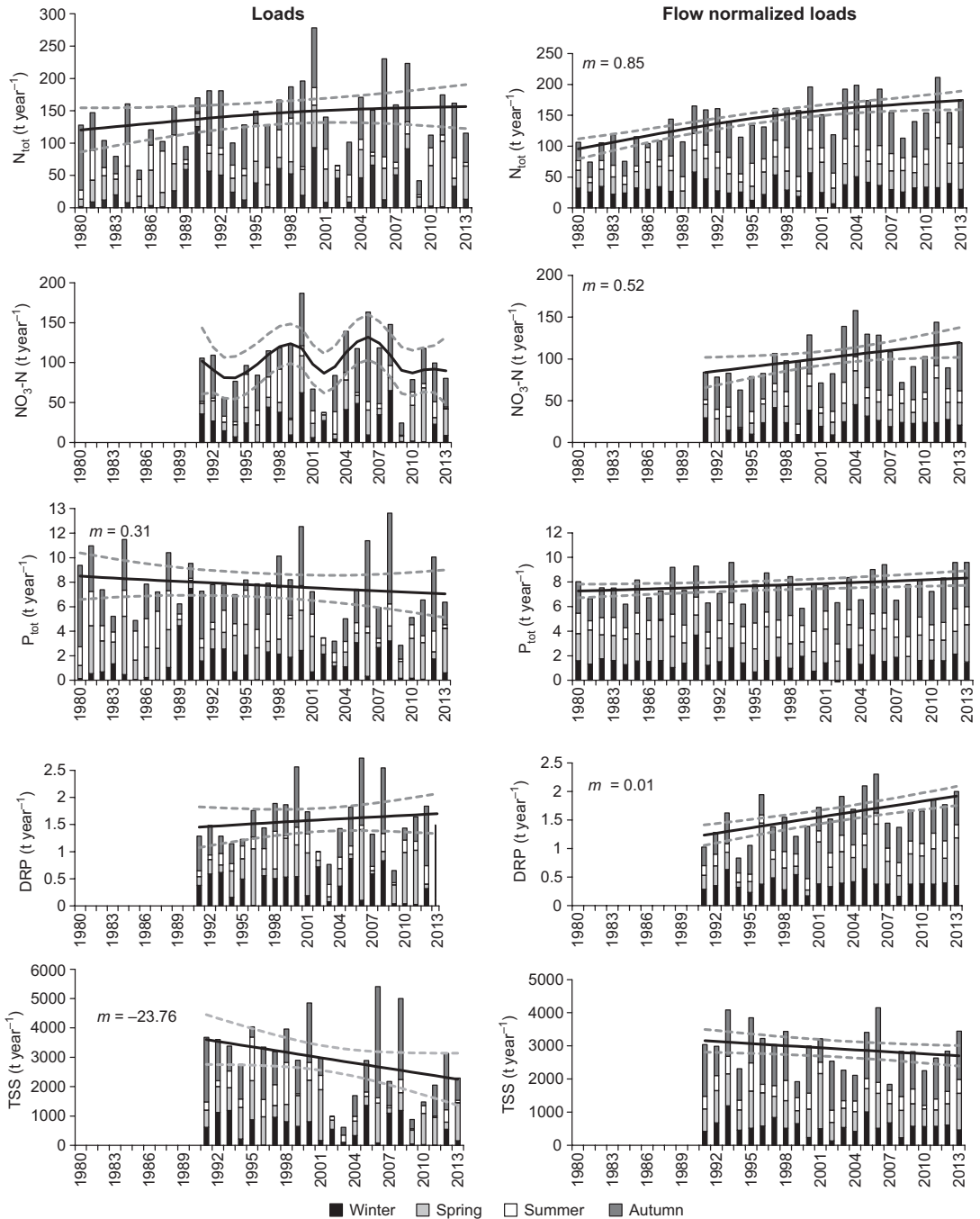


Fig. 3. Trends in the nutrient loads in the Yläneenjoki. *m* denotes the rate of change per unit for significant trends as evaluated by the multivariate Mann-Kendall test. Solid black lines are the annual trends and dashed grey lines the 95% confidence limits.

significant and decreasing in February, May, July and August (Table 5). The trends in the monthly time-series of loads of N_{tot} , NO_3-N , DRP and TSS were similar to the trends in their

monthly concentrations (Table 6). There were however discrepancies in some months. The trends in the ON, P_{tot} and PP concentrations and loads were significant in some months, but no

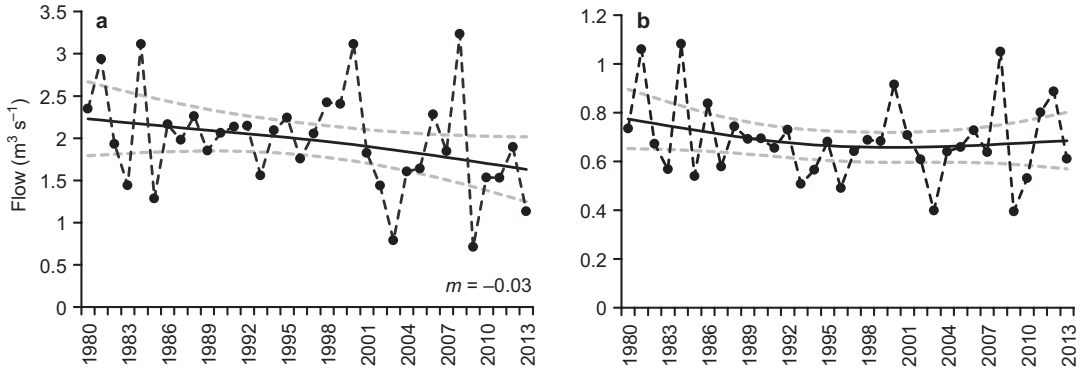


Fig. 4. Trends in the flow in the studied rivers. m denotes the rate of change per unit for significant trends as evaluated by the multivariate Mann-Kendall test. Dashed black lines with dots represents the annual average flow, solid black lines the semi-parametric regression trends and dashed grey lines the associated 95% confidence limits.

significant annual trends were detected in those variables. Overall, in Yläneenjoki, the average N_{tot} concentrations increased by 13%, from 1.87 mg l⁻¹ in 1980–2000 to 2.13 mg l⁻¹ in 2001–2013. Similarly, the average NO₃-N concentrations increased by 16%, from 1.14 mg l⁻¹ in 1991–2000 to 1.32 mg l⁻¹ in 2001–2013. Also, the average DRP concentrations increased by 52%, from 0.02 mg l⁻¹ in 1991–2000 to 0.03 mg l⁻¹ in 2001–2013. Further, the concentrations of TSS decreased by 33%, from 43.4 mg l⁻¹ in 1991–2000 to 29.0 mg l⁻¹ in 2001–2013, and also the average flow decreased by 21%, from 2.15 m³ s⁻¹ in 1980–2000 to 1.70 m³ s⁻¹ in 2001–2013.

Discussion

Detection of long-term water-quality trends is confounded by simultaneous influence of several factors (Hirsch *et al.* 2010). Climatic and hydrological variability provide a challenge, as typically periods with high and low concentrations and loads of nutrients are combined with extremely dry or wet years. For reliable trend detection a good long-term data record is required, because the trend can be buried in high data variability (Burt *et al.* 2008). In this study, we used the 19- to 34-year-long WQ time-series, which seemed to provide an adequate insight into water quality changes in the studied rivers. However, in spite of the long-term data we used in our analysis, some degree

of uncertainty in the detected trends might be expected due to sampling representativeness and inaccuracies in nutrient load estimations (Wang *et al.* 2001, Johnes 2007). In the study area, samples were taken more frequently during high- than during low-flow periods. Although this approach improved the nutrient-load estimation accuracy more than a regular fixed interval sampling throughout the year (Rekolainen *et al.* 1991), it still missed some important, random-flow events and associated nutrient exports. Several studies revealed that few extreme-flow periods export substantially large amounts (> 50%) of nutrients from agricultural areas (Pionke *et al.* 1996, Royer *et al.* 2006). Additionally, during a storm event, most of the water quality variables change in different directions and have different degrees of hysteresis (House and Warwick 1998, Bowes *et al.* 2005), depending on the prevailing sampling conditions. When all these variations are not captured in WQ sampling, substantial over- or underestimations of nutrient loadings can be expected. For the same Yläneenjoki, Koskiahio *et al.* (2010) found an underestimation of 1.5 fold for the TSS and 1.3 fold for P_{tot} loads, when they compared nutrient load estimates based on infrequent sampling data from normal monitoring with estimates based on sub-hourly automatic sensor data. Unfortunately, sensor-based data are currently available only for few WQ variables and for very short periods, and they are not free of errors (Kirchner *et al.* 2004). Therefore, their use for long-term trend

Table 5. Trends in flow-normalized nutrient concentration monthly time series (mg l⁻¹) in Yläneenjoki as evaluated by the univariate Mann-Kendall trend test; τ = Mann-Kendall tau. Trend analysis for N_{tot} and P_{tot} was carried out for the years 1980–2013 and for the rest of WQ variables for 1991–2013.

	N_{tot}			$\text{NO}_3\text{-N}$			ON			P_{tot}			DRP			PP			TSS		
	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n
Jan	0.29	0.037	27	0.09	0.581	20	0.18	0.33	20	0.22	0.109	28	0.35	0.042	19	0.20	0.234	19	-0.08	0.627	20
Feb	0.22	0.082	31	0.05	0.778	22	0.47	0.018	15	0.02	0.865	31	0.37	0.060	15	0.05	0.827	14	-0.37	0.015	23
Mar	0.07	0.588	33	0.18	0.263	21	-0.03	0.871	20	-0.05	0.685	32	0.33	0.037	21	0.18	0.263	21	-0.15	0.338	22
Apr	0.22	0.070	34	0.17	0.267	23	0.10	0.526	23	-0.13	0.273	34	0.30	0.035	23	-0.02	0.916	23	-0.26	0.091	23
May	0.61	< 0.001	34	0.51	0.001	21	0.16	0.319	21	-0.16	0.202	34	0.32	0.050	23	-0.15	0.342	23	-0.42	0.005	23
Jun	0.18	0.150	33	0.42	0.006	23	-0.06	0.735	22	-0.25	0.039	33	0.40	0.008	23	-0.36	0.017	23	-0.27	0.080	22
Jul	0.30	0.020	30	0.34	0.032	21	-0.06	0.970	19	0.12	0.353	30	0.52	0.002	19	-0.10	0.576	19	-0.38	0.020	21
Aug	0.05	0.707	28	-0.01	0.967	17	0.10	0.592	17	0.16	0.244	28	0.41	0.023	17	-0.03	0.902	17	-0.34	0.048	17
Sep	0.07	0.586	29	-0.13	0.456	20	-0.05	0.780	19	0.19	0.134	30	0.59	< 0.001	21	-0.10	0.566	21	-0.28	0.085	21
Oct	0.20	0.103	34	-0.06	0.735	22	0.16	0.319	21	0.28	0.021	34	0.44	0.004	23	0.00	1.000	23	-0.30	0.051	23
Nov	0.32	0.017	29	0.24	0.113	23	0.06	0.711	23	0.43	0.001	29	0.42	0.005	23	0.17	0.267	23	-0.10	0.526	23
Dec	0.27	0.047	28	0.19	0.205	23	0.00	0.890	23	0.22	0.095	29	0.27	0.080	22	0.19	0.236	22	0.04	0.822	22

Table 6. Trends in flow-normalized nutrient loads (t month⁻¹) and flow (m³ s⁻¹) in Yläneenjoki as evaluated by the univariate Mann-Kendall trend test. Trend analysis for N_{tot} and P_{tot} was carried out for the years 1980–2013 and for the rest of WQ variables for 1991–2013. For explanations of other symbols see Table 5.

	N_{tot}			$\text{NO}_3\text{-N}$			ON			P_{tot}			DRP			PP			TSS		
	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n	τ	ρ	n
Jan	0.22	0.066	34	0.17	0.267	23	0.24	0.113	23	-0.05	0.722	34	0.08	0.597	23	0.14	0.369	23	0.02	0.916	23
Feb	0.07	0.563	34	-0.04	0.792	23	0.15	0.316	23	-0.02	0.859	34	0.20	0.187	23	0.01	0.958	23	-0.34	0.023	23
Mar	0.06	0.609	33	0.29	0.057	23	0.24	0.113	23	0.00	1.000	34	0.12	0.460	23	0.12	0.460	23	0.07	0.673	23
Apr	0.29	0.018	34	0.28	0.064	23	0.19	0.205	23	-0.12	0.328	34	0.41	0.007	23	-0.16	0.291	23	-0.16	0.291	23
May	0.50	< 0.001	34	0.34	0.027	23	-0.13	0.398	23	-0.02	0.882	34	0.37	0.015	23	0.02	0.916	23	-0.13	0.398	23
Jun	0.36	0.003	34	0.21	0.169	23	0.03	0.958	23	-0.06	0.614	34	0.34	0.023	23	-0.23	0.139	23	-0.10	0.526	23
July	0.30	0.014	34	0.31	0.039	23	-0.03	0.874	23	0.15	0.224	34	0.27	0.073	23	0.03	0.874	23	-0.02	0.916	23
Aug	0.13	0.286	34	0.15	0.316	23	-0.07	0.673	23	0.04	0.767	34	0.27	0.073	23	-0.24	0.113	23	-0.16	0.291	23
Sep	0.41	0.001	34	0.18	0.245	23	0.27	0.073	23	0.11	0.374	34	0.56	< 0.001	23	0.10	0.526	23	-0.04	0.833	23
Oct	0.29	0.018	34	-0.06	0.712	23	0.14	0.369	23	0.26	0.035	34	0.49	0.001	23	0.16	0.291	23	-0.07	0.673	23
Nov	0.40	0.001	34	0.12	0.428	23	0.00	1.000	23	0.32	0.009	34	0.30	0.051	23	0.04	0.833	23	-0.19	0.224	23
Dec	0.40	0.002	34	0.21	0.170	23	0.17	0.267	23	0.21	0.085	34	0.25	0.101	23	0.30	0.045	23	0.26	0.091	23

analyses is limited. On the other hand, the use of nutrient-concentration or nutrient-load data in trend analysis depends on the purpose of the study. To study the river water-quality condition or effects of water protection measures, trend analysis using nutrient-concentration data are preferred, whereas trend analysis using nutrient loads is useful to study effect on downstream waterbodies (Hirsch *et al.* 2010). Furthermore, although concentrations and loads are related, they can have different patterns and trends (Hirsch *et al.* 2010).

In the north, climatic variability strongly affects stream flow and nutrient losses. In the Yläneenjoki, substantial inter-annual variations in nutrient loads have been found, particularly in winter, due to the occurrences of mild winters with intensive rainfall (Ventelä *et al.* 2011). Generally, inter-annual nutrient-loss variability is affected more by climate than agriculture (Stålnacke and Grimvall 2001). Therefore, to detect the signs of change in river WQ linked to agriculture, it is necessary to reduce the variability caused by climatic fluctuations (normalization). Usually river discharge data are used to normalize WQ data (flow-normalization, Medalie *et al.* 2012). Most of the regression-based flow-normalization methods usually assume that seasonal cycles and relationships between flow and nutrient concentration are constant over time, and the WQ trend is defined by change in the intercept, but Hirsch *et al.* (2010) showed that these relationships change with seasons and periods. In our data, the correlation strengths changed among the months for several WQ variables. Only a semi-parametric regression model (Wahlin 2008) and weighted regressions on time, discharge and season (WRTDS, Hirsch *et al.* 2010) took into account this fact. The semi-parametric regression model of normalization of environmental data has certain advantages compared with other methods: (1) It is very flexible and allows changes in the intercept and slope in the fitted function, (2) abrupt changes are controlled by a smoothing factor, (3) it allows missing values, (4) other covariate than discharge can be included, and (5) it has already been tested in several areas (Hussian *et al.* 2004, Sålacke and Grimvall 2001, Wahlin and Grimvall 2008).

Also, a two-step procedure, data normalization followed by a trend test, is convenient in cases when the relationship between concentration and flow varies among seasons and periods (Libiseller 2004) as in our case.

In general, all three trend tests we used detected similar trends in concentration data for most of the studied variables, but the results differed when the load data were used. The univariate MK method produced similar results as MMK and FN-MKK methods when annual average concentration data were used; the only exception being $\text{NO}_3\text{-N}$ for which a significant trend was detected with the FN-MKK method only. The MK method differed from the other methods by not detecting trends in annual loads of any studied variables. The WQ data are usually skewed, contain outliers and vary seasonally, which can substantially affect the representativeness of annual means (Hirsch *et al.* 1982), and therefore affect trend detection. A major uncertainty is involved in trend detection based on annual loads due to accuracy of estimation of annual loads as discussed above. The MMK and FN-MKK methods gave similar results when applied to the nutrient concentration data, but they differed in trend detection in the nutrient load data. This may be due to more effective flow-normalization of the nutrient-load than nutrient-concentration data. Other climatic covariates may also be taken into account to further normalize the nutrient concentration data, i.e. rainfall, even though the response of water chemistry to rainfall events is highly non-linear and strongly affected by the antecedent conditions (Libiseller 2004). An advantage of the MMK test is the full use monthly data providing a unique statistics for multiple time-series data (months) used. It also controls the overall significance level and avoids false trend detection (Manly 2005 as cited in Chebana *et al.* 2013). However, it does not provide statistics for individual time-series data and their contribution to the overall trend. Consequently, to study the trend of multiple time-series data thoroughly, both univariate and multivariate tests would be recommendable (Chebana *et al.* 2013).

There might be several causes for the detected WQ trends in the studied catchments, but there are also some signs of a possible link to current

agricultural land management. Firstly, there are some differences between the studied river catchments. In the Yläneenjoki catchment, most of the catchment is on clay soils with a lower base-flow index (0.37), indicating that higher proportion of flow in the river is formed by surface and near-surface flow pathways, especially during storm events. By contrast, in the Pyhäjoki catchment, most of the catchment area is on sandy soil having a higher base-flow index (0.53), suggesting that a higher proportion of the flow in the river is from groundwater sources. Several studies report differences in nutrient concentrations among catchments with contrasting base-flow indices (Jordan *et al.* 1997, Johnes 2007, Tesoriero *et al.* 2009). Secondly, the spatial variability of most of the water quality variables in the tributaries of the studied rivers was largely explained by the fraction of catchment in agricultural use (Gonzales-Inca *et al.* 2015). Therefore, most of the WQ trends, such as the decreasing trend in TSS, and also increasing trend in concentrations and loads of dissolved reactive phosphorus (DRP) in the Yläneenjoki, might be linked to the current effort of agricultural soil erosion reduction. Decreased tillage or no-tillage methods increased in the Yläneenjoki catchment from 17% to 39% between 2000 and 2010 (Aakkula and Leppänen 2014). Several other studies carried out in Finland found that the mobilization of DRP increases in areas with reduced tillage depth or no-till (Koskiahho 2002, Uusitalo *et al.* 2007), and in vegetation buffer zones (Uusi-Kämppe and Jauhiainen 2010). Plant residues release easily degradable form of P into the soil surface and accumulation of the organic matter in no-tillage soils may reduce the soil P retention due to the competition between organic anions and DRP for the same clay sorption sites (Muukkonen *et al.* 2009). Also surface-applied P fertilizer in no-tillage soil can increase bio-available P at the soil surface (Logan 1982). All these processes may contribute to high DRP concentrations in soil solution, which will be transported by the runoff into the streams. The decreasing trend in suspended solid is usually linked to the reduction of P_{tot} and PP in agricultural areas (Udeigwe *et al.* 2007). In our study area, in spite of a decrease in the TSS concentration, no clear long-term trend was identified in the P_{tot} or PP concentration.

Although in the Yläneenjoki decreasing trends in the P_{tot} , and PP loads were found using the MMK method, they were not significant when the load data were flow-normalized. It might be possible that these trends were just linked to the decreasing flow in Yläneenjoki.

The increasing trends in the concentrations and loads of N_{tot} and NO_3-N , despite the reduction in fertilizer application, may also be linked to the current agricultural management. In the study area, cereal cultivation and managed grasses make up around 90% of the agricultural area. In cereals, approximately 50% of the total plant N is located in below-ground biomass (Hatch *et al.* 2002) and crop residues can be enriched by N fertilizer and they decompose faster than other organic matter in soils (Kirchmann *et al.* 2002). Furthermore, it has been shown that after the harvest the organic-matter-derived N is predominant (~90%) in the soil, and the rest might correspond to applied inorganic nitrogen fertilizer (Bergström 1987, MacDonald *et al.* 1989, Kirchmann *et al.* 2002). Consequently, mineralization and nitrification of crop residues can contribute substantially to N leaching. The increasing trend in the N_{tot} concentrations and loads during autumn months and January may be related to these phenomena and N mineralization might be extended further into autumn due to warm and humid soil conditions in mild winters in changing climate, as it was suggested also in earlier studies (Roberts 1987, Mattikalli 1996, Groffman *et al.* 2001). The increasing trend in NO_3-N from May to July may reflect the high nitrification rate occurring in spring and summer (Kirchmann *et al.* 2002), when the snowmelt-wetted soil aeration improves.

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Appendix. Spearman correlations (r_s) between nutrient concentrations (mg l^{-1}) and flow ($\text{m}^3 \text{s}^{-1}$) in each month; correlation coefficients of existing correlations are set in boldface.

	N_{tot}		NO_3		ON		DRP		P_{tot}		PP		TSS	
	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p	r_s	p
Yläneenjoki														
Jan	0.48	0.002	0.47	0.012	0.56	0.002	0.08	0.693	0.23	0.158	0.36	0.055	0.70	<0.001
Feb	0.45	0.002	0.33	0.096	0.19	0.431	0.06	0.787	0.18	0.225	0.33	0.146	0.51	0.004
Mar	0.45	<0.001	0.07	0.637	0.42	0.007	-0.11	0.466	0.16	0.125	0.27	0.068	0.64	<0.001
Apr	0.15	0.055	-0.07	0.276	0.22	0.041	0.29	0.002	0.29	<0.001	0.30	0.002	0.40	<0.001
May	0.20	0.009	0.12	0.205	0.37	0.001	0.33	0.001	0.38	<0.001	0.43	<0.001	0.64	<0.001
Jun	0.75	<0.001	0.70	<0.001	0.29	0.067	-0.04	0.784	0.39	<0.001	0.57	<0.001	0.79	<0.001
Jul	0.37	0.004	0.59	<0.001	0.43	-0.13	0.27	0.094	0.40	0.002	0.43	0.006	0.71	<0.001
Aug	0.59	<0.001	0.76	<0.001	0.28	0.194	0.08	0.670	0.31	0.089	0.31	0.089	0.70	<0.001
Sep	0.77	<0.001	0.85	<0.001	0.41	0.59	-0.26	0.108	-0.01	0.926	0.47	0.002	0.82	<0.001
Oct	0.58	<0.001	0.72	<0.001	0.60	0.64	0.08	0.566	0.29	0.005	0.33	0.013	0.80	<0.001
Nov	0.51	<0.001	0.53	<0.001	0.58	0.57	0.03	0.828	0.56	<0.001	0.53	<0.001	0.67	<0.001
XII	0.49	0.001	0.23	0.174	0.35	0.65	-0.17	0.342	0.40	0.009	0.54	0.001	0.66	<0.001
Pyhäjoki														
Jan	0.87	<0.001	0.81	<0.001	0.19	0.61	-0.16	0.531	0.10	0.688	0.30	0.226	0.57	0.011
Feb	0.84	<0.001	0.67	0.001	0.22	0.91	<0.001	0.418	0.10	0.655	0.48	0.111	0.64	0.002
Mar	0.87	<0.001	0.69	<0.001	0.46	0.88	<0.001	0.342	0.54	<0.001	0.69	0.000	0.84	<0.001
Apr	0.41	<0.001	0.26	0.011	0.94	0.52	<0.001	0.28	0.38	<0.001	0.43	0.000	0.51	<0.001
May	0.85	<0.001	0.83	<0.001	0.72	0.39	<0.001	-0.40	-0.16	0.126	-0.08	0.458	0.53	<0.001
Jun	0.87	<0.001	0.84	<0.001	0.34	0.29	0.104	0.784	0.46	0.001	0.50	0.000	0.79	<0.001
Jul	0.64	<0.001	0.62	<0.001	0.34	0.57	0.001	0.326	0.24	0.164	0.41	0.048	0.72	<0.001
Aug	0.79	<0.001	0.66	0.002	0.19	0.68	0.002	0.276	0.80	<0.001	0.78	0.000	0.72	<0.001
Sep	0.77	<0.001	0.73	<0.001	0.34	0.83	<0.001	0.610	0.60	<0.001	0.64	0.000	0.63	<0.001
Nov	0.80	<0.001	0.81	<0.001	0.50	0.89	<0.001	0.166	0.62	<0.001	0.52	0.000	0.68	<0.001
Dec	0.59	0.003	0.47	0.025	0.23	0.74	<0.001	0.628	0.46	0.028	0.57	0.006	0.69	<0.001