Nutrient loads from agricultural and forested areas in Finland from 1 1981 up to 2010 - Can the efficiency of undertaken water protection 2 measures seen? 3

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- 14
- 15 Key words: Long-term monitoring, nutrient loading, trend analyses, water quality, mitigation measures

16 Abstract

17 Long-term data from a network of intensively monitored research catchments in Finland was analyzed. We

- 18 studied temporal (1981-2010) and spatial variability in nitrogen (N) and phosphorus (P, from 1987) losses, both 19 from agricultural and forestry land. Based on trend analysis, total nitrogen (TN) concentrations increased in two
- 20 of the four agricultural sites and in most of the forested sites. In agricultural catchments, the total phosphorus
- (TP) trends were decreasing in two of the four catchments studied. Dissolved P (DRP) concentrations increased
- 21 22 in two catchments and decreased in one. The increase in DRP concentration can be a result of reducing erosion
- 23 by increased non-plough cultivation and direct sowing. In forested catchments, the TP trends in 1987-2011 were
- significantly decreasing in three of the six catchments, while DRP concentrations decreased significantly in all
- 24 25 sites. At the same time, P fertilization in Finnish forests has decreased significantly, thus contributing to these
- 26 changes. The mean annual specific loss for agricultural land was on average 15.5 kg ha⁻¹yr⁻¹ for N and 1.1 kg ha⁻
- 27 ¹yr⁻¹ for P. In the national scale, total TN loading from agriculture varied between 34,000-37,000 t yr⁻¹ and total
- 28 P loading 2,400-2,700 t yr⁻¹. These new load estimates are of the same order than those reported earlier,
- 29 emphasizing the need for more efforts with wide-ranging and carefully targeted implementation of water
- 30 protection measures.

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31 Introduction

- 32 Non-point source loading, predominantly from agriculture, is recognised as a major source of anthropogenic
- 33 nutrient loading to watercourses throughout the world (e.g. Collins et al. 2016; Rocha et al. 2015; Heathwaite et
- al. 1993; Sharpley et al. 1994; Foy and Withers 1995) and also in Nordic and Baltic countries (e.g. Kronvang et
- al. 1995; Stålnacke 1996; Lääne et al. 2002). In Finland, the major share of nutrient loading originates from
- 36 agriculture (Rekolainen et al. 1997; Vuorenmaa et al. 2002). Although the area under cultivation in Finland is
- 37 relatively small (7.4%), there are many river basins where it may exceed 30% (Räike et al. 2003). Fields are
- 38 concentrated in southern and western Finland. Forests also contribute to diffuse pollution: they cover 78% of the
- total land area in Finland, of which forested peat lands cover one third. Forestry in Finland has been estimated
- 40 to contribute on average 9% of the total N export, with dominance towards eastern and northern parts of the
- 41 country: from 2-15% in the southern-mid-western Finland basins to 10-30% in the large northern basins
- 42 (Lepistö et al. 2006). Forestry loading occurs from a mosaic of numerous treatment areas (cuttings, forest
- 43 drainage), where the time period of the impact varies from site to site providing high uncertainties to load
- 44 estimates. Besides forestry, natural background loading is also important in the north.
- 45

46 Climate change induced mild winters have become more common in the last couple of decades (e.g. IPCC
47 2013). Such mild winters – with occasional, heavy rainfall events – transmute the traditional flow patterns with

- 48 predictable spring and autumn floods and low flow in midwinter into more unpredictable distributions of flow.
- 49 In Finland, Korhonen and Kuusisto (2010) have detected clear trends in many seasonal flow time series, with
- 50 increases in winter and spring mean flow together with earlier timing of the spring peak. An unwanted effect of
- 51 these trends is the increased loading to surface waters due to the increased erosion and leaching of nutrients
- from unfrozen soil with thin or no snow cover (e.g. Koskiaho et al. 2010).
- 53

54 Small representative catchments, experimental plots, or river basins loaded mainly by non-point sources provide 55 a framework to assess and monitor the non-point source nutrient loading. However, the transport of nutrients 56 from diffuse sources is strongly influenced by a complex combination of temporal and spatial factors, such as 57 fluctuating climatic and hydrological conditions, geomorphological characteristics, crop cycles and land-use 58 practices (Vuorenmaa 2002; Kyllmar et al. 2006, 2014; Santos et al. 2015; Pacheco and Sanches Fernandes

- **59** 2016).
- During 1981-1995, in these same small catchments that are studied here, the agricultural loading level averaged
- 61 15 kg ha⁻¹ yr⁻¹ for nitrogen (N) and 1.1 kg ha⁻¹ yr⁻¹ for phosphorus, with no clear trends in total losses and
- 62 concentrations. For forested catchments, the loading averaged 1.9 kg ha⁻¹ yr⁻¹ for nitrogen and 0.09 kg ha⁻¹ yr⁻¹
- 63 for P (Vuorenmaa et. al 2002). In 35 small agricultural catchments in Nordic and Baltic countries, loading
- 64 ranged from 5 to 75 kg ha⁻¹ yr⁻¹ for nitrogen in 1994-1997, with significant within-country and inter-annual
- 65 variation. The main explanations for this variability were runoff, fertilizer use (especially the amount of
- 66 manure), soil type and erosion, but there was poor correlation between nitrogen losses and surpluses (Vagstad et
- 67 al. 2004).
- 68

- 69 The Finnish Agri-Environmental Programme FAEP, launched in 1995, forms the most important policy
- 70 instrument for controlling agricultural nutrient loading. Since 2002, FAEP has covered about 93% of the
- 71 agricultural area (Ministry of Agriculture and Forestry 2004; Aakkula and Leppänen 2014). Due to reduced
- 72 fertilization limits of FAEP and increased fertilizer prices, national level N and P field balances have decreased
- 73 35% and 60%, respectively, in the period 1995-2013 (Aakkula and Leppänen 2014). Increased non-plough
- 74 cultivation and direct sowing have taken place in almost two thirds of earlier autumn-ploughed field areas (Farm
- 75 <u>Structure Survey / Agricultural Census 2010 Farmland management</u>). Additionally, 8,600 hectares of buffer
- 76 zones and about 500 constructed wetlands and sedimentation ponds have been established after 1995 according
- to FAEP (Berninger et al. 2012).
- 78
- The first studies made in the agricultural catchments after implementation of WFD indicated limited or no
 reduction of nutrient loads (Räike et al. 2003; Granlund et al. 2005). Later on, in the national MYTVAS followup study based on nutrient load monitoring in the agriculture-dominated river basins, the phosphorus load per
 hectare of cropland has nevertheless decreased in each programme period; being about 80% (0.72 kg ha⁻¹) in the
 third period (2007-2013) of the level of the first period (1995-1999) (0.90 kg ha⁻¹). However, the nitrogen load
 on waterways from agriculture continued to grow during the second programme period (2000-2006). In the third
 programme period (2007-2013), the nitrogen load per hectare of cropland (12.9 kg ha⁻¹) had decreased
- somewhat, i.e. 7% from the level of the first period (13.9 kg ha⁻¹) (Aakkula and Leppänen 2014).
- 87

88 Most of the agri-environmental practices and measures are established on a field basis, but benefits may be

- 89 realized in freshwaters lower in the river basin, or in coastal waters. For these reasons, there was a high demand
- 90 to revisit the long-term data of small catchments, including both old published and new unpublished data. The
- 91 small catchments are the missing link between field and river basin scales on planning agri-environment
- 92 programmes for WFD, on developing loading models and on the practical implementation of agri-environmental
- 93 measures in order to improve the state of watercourses.
- 94
- 95 The aim of this study was to evaluate 30-year long-term data (1981-2010 for nitrogen, 1987-2010 for
- 96 phosphorus) from 12 small catchments in Finland. We compared the period without agri-environment measures
- 97 before 1995 to the period with measures in order to look if impacts on loading levels can be seen. Further, we
- 98 compared loading levels in agricultural and forested catchments, annual and monthly trends in nutrient
- 99 concentrations and flow factors affecting changes in the catchments, together with factors affecting annual and
- 100 year-to-year variability and variation between different catchments. In addition, continuous water quality data
- 101 was used in one catchment to evaluate the accuracy of loading estimates for the period 2010-2013.

102 Materials and Methods

- 103
- 104 Characteristics of small research catchments105
- 106 The national network of 37 small catchments was established in 1957, while monitoring of runoff water quality107 started in 34 of these catchments in 1962 (Vuorenmaa et al. 2002). Geological conditions in Finland, such as

108 rather thin soil layers and impermeable bedrock, support the reliable estimation of water balance in these 109 catchments. The catchments have no lakes and their earlier main purpose was to obtain hydrological information 110 on the soil and climate conditions typical for Finland.

111

112 The catchments were divided into three classes: i) agricultural catchments (agricultural land area 39-100%), ii)

113 mixed catchments (agricultural land area 16-21%) and iii) forested catchments (agricultural land area <5%).

114 Haapajyrä is a special case within agricultural catchments, as it consists mainly of acid sulphate soils. Acid

sulphate soils contain high amounts of sulphidic acids and the phosphorus loads are found to be lower and nitrogen loads higher compared to other agricultural catchments (Rekolainen 1989). A total of four agricultural

117 catchments, two mixed and six forested, were included in this study; together they provide a good geographical

- 118 coverage in different parts of Finland (Fig. 1, Table 1).
- 119

120 Fig. 1. Location of studied research catchments and the share of arable land.

121

The catchment areas vary between 0.12 km² and 21.70 km² (Table 1). The highest percentage of agricultural land is in Hovi (100%), while Teeressuonoja, Huhtisuonoja and Vähä-Askanjoki are completely covered by forests and forested peatlands (Table 1). Most of the mineral soil in the forested catchments is non-graded moraine, and the coverage of peat soils is typically high: 13-50% in forested catchments with the exception of Laanioja. Most arable land in agricultural catchments is located on graded soils, with high proportions of silt and clay (Table 1).

128

Agricultural and mixed catchments130

Fields in Löytäneenoja, Savijoki and Haapajyrä are located on flat areas with average slopes under 0.8%, whereas the fields in Ruunapuro, Hovi and Latosuonoja are relatively steep for Finnish agricultural fields (Table 2). All crops in the catchment of Hovi consist of spring cereal, whereas in Löytäneenoja both spring cereals (68%) and root crops (27%) are cultivated. In addition to spring cereal (70%), some autumn cereals (17%) and root crops (0.9%) are cultivated in Savijoki (Table 2). Considerable amounts of livestock occur in Savijoki (livestock unit LSU=238) and Haapajyrä (LSU=119), but in the other catchments the LSUs are negligible (Table 2).

138

Table 1. Area (km²), percentage of agricultural land, and shares of graded soils, peat soils and moraines (%) in
 the studied catchments. Numbers of catchments refer to Fig. 1.

141

Table 2. Average slope (%) of agricultural fields and proportion (%) of different crops and livestock units (LSU)in the agricultural and mixed catchments.

144

145 The tillage practices in the Hovi catchment vary from year to year: autumn ploughing has decreased from 100% 146 in 1981-87 to 0-30% in 2005-2010 (Table 3). Before 1987, the autumnal ploughing, which leaves the soil prone 147 to erosion and nutrient losses, was the most common agricultural practice in Finland. At present, the greening of

- 148 the fields has become much more common, e.g. during 2014 less than 30% of the field area was ploughed in the
- 149 autumn or cultivated with winter grains. The most dominating practices nowadays are reduced tillage methods,
- 150 and permanent grass cover is also in use. During 1991-1994 there was a national obligation to leave part of the
- 151 field area fallow, of which two thirds was implemented as green fallow. In addition, the use of mineral fertilisers
- 152 in agriculture has decreased drastically in Finland from the early 1990s to the present day (Fig. 2).
- 153
- 154 Fig. 2. Forest fertilisation area and the use of mineral N and P fertilisers in agricultural land in Finland, both 155 during 1975-2011.
- 156

157 Table 3. The used tillage practice (1 to 4) and the corresponding share of the plant coverage areas (% of the total 158 arable area) during winter season (approximately Oct.-Apr.) in the Hovi research basin during 1981-2010.

159

160 **Forested catchments** 161

162 All of the forested small catchments represent managed forest land, where numerous year-to-year forestry 163 practices have occurred (Vuorenmaa et al. 2002). The most important forestry practices in 1960-1990 (drainage, 164 clear-cuttings, soil tillage, fertilising) in Huhtisuonoja, Kesselinpuro and Vähä-Askanjoki have been described 165 by Kortelainen and Saukkonen (1998).

166

167 Forest drainage works had been conducted in 48% of Huhtisuonoja catchment, 30% of Kesselinpuro, 18% of 168 Myllypuro and 8% of Vähä-Askanjoki, respectively, before 1990 (Saukkonen and Kortelainen 1995). After that, 169 in the 1990s and 2000s, more focus has been on supplementary drainage works (Metla Statistical Yearbooks), 170 but detailed areas in most of these catchments after 1990 are not known. At present, the areas under forestry 171 actions are much smaller than before 1990. In the Teeressuonoja catchment, there have been many clear-cuttings 172 between 1985-2001. Meanwhile, the Huhtisuonoja catchment was extensively fertilised during 1985-1986 with 173 P-fertilisers (70% of the total area of peatland i.e. 156 ha, where 32.7 P kg ha⁻¹ was applied) (Pietiläinen and 174 Rekolainen 1991; Vuorenmaa et al. 2002). The forest fertilisation area in Finland was highest in the mid-1970s 175 (250,000 ha year⁻¹), then decreased close to zero in the early 1990s, where after it has gradually risen but not to 176 high levels (see Fig. 2). Although some data is available on practices from single basins, no database with a 177 continuous survey of either forestry or agricultural practices unfortunately exists for all these research 178 catchments.

179

180 Monitoring of runoff and water quality 181

182

Runoff measurements, sampling and chemical analyses 183

184 The runoff in all of the 12 study catchments was measured continuously by overflow weirs with water stage 185 recorder, and stored to the hydrological database by the Finnish Environment Institute (SYKE) (Linjama 2012).

- 186 Monitoring of water quality was mostly based on manual, grab water samples. Sampling was concentrated on
- 187 spring and autumn high flow periods. Automatic flow-weighted water quality sampling (ISCO) has also been
- 188 utilised in five of the catchments (Vuorenmaa et al. 2002). The number of automatic samples has varied between

30 and 50 per year. On average for the whole monitoring period, sampling frequency has been quite high:
annual samples have varied between 23-58 in agricultural catchments and between 12-42 in forested
catchments, respectively.

192

193 In this study, total phosphorus (TP), dissolved reactive phosphorus (DRP), total nitrogen (TN) and nitrate 194 nitrogen (NO₃-N) concentrations were used for trends and loss estimates. Water quality variables were analyzed 195 in the laboratories of SYKE or Regional Environment Centers, with methods based on international standards 196 (Näykki et al. 2013). The DRP concentration was determined with a polycarbonate (0.4 μ m) filtration and by 197 colorimetric spectrometer. Before 1986, analyzing was done without filtration. Therefore, the DRP data is only 198 used from 1986 onwards. For the TP analysis, phosphorus was first digested with peroxodisulphate and then 199 determined by the molybdenum blue method (Helcom 2004). The TN was digested with peroxodisulphate and 200 determined with a spectrometer. The NO₃-N was analyzed by Cd-reduction to nitrite followed by colorimetric 201 determination. Turbidity was determined with a nephelometric method. The suspended sediment was first 202 filtered with polycarbonate (0.4 μ m) and then analyzed with gravimetric drying in 105 °C.

203 Continuous water quality measurements at Savijoki 204

205 Continuous water quality measurements were carried out in Savijoki, using the S::can Nitrolyser probe (S::can 206 Measuring Systems). The NO₃-N and turbidity were measured with a 5 mm measuring path length with a 207 measurement range of 0.3-70 mg l⁻¹ for NO₃-N and 5-1,400 FTU (Formazin Turbidity Unit) for turbidity 208 (Linjama et al. 2009). The accuracy and measuring range for NO₃-N is dependent on turbidity. The whole scale 209 can be measured in clear water, but if turbidity increases to near the upper limit of the scale, NO₃-N cannot be 210 measured. If turbidity increases to 250 FTU, an NO₃-N concentration of < 5 mg/l can still be measured (Linjama 211 et al. 2009). The probe was calibrated for the local conditions with utilisation of comparable readings from 212 laboratory analysis.

213

The measuring path and windows of the probe were cleaned with compressed air before each measurement. Once a month the probe was taken out and cleaned manually and twice a year checked and calibrated with distilled water (Linjama et al. 2009). Water level was measured with a pressure probe (Keller AG) with a 1 mm resolution. Discharge was determined according to a weir-specific stage-discharge curve from the water level measurements (Linjama et al. 2009).

219

220 Calculation methods

221

222 Nutrient loads

223

Slightly differing methods, such as interpolation method (Rekolainen 1989; Vuorenmaa et al. 2002) and the monthly mean method (Helcom 2014), have been utilised when estimating nutrient losses from Finnish catchments. The periodic method was chosen to be used in this study as it has been found to have the highest general reliability (RMSE) for estimation of TN load (Kauppila and Koskiaho 2003). The periodic method was utilised for monthly and annual calculations of nutrient losses. In this method, the nutrient fluxes are first calculated for each day by using the daily discharge measurements. The daily concentrations are calculated
based on the temporal midpoints of the observation days as illustrated in Fig. 3. The annual nutrient flux is
calculated with the following equation:

(1)

232
233
234
$$L_a = \sum_{i=1}^{365} c(t_i) \cdot Q(t_i)$$

235

236 where L_a is the annual nutrient flux 237 $c(t_i)$ is the concentration of the day i

- 238 $Q(t_i)$ is the mean discharge of the day i
- 239

Fig. 3. Schematic approach to the calculation of the concentrations over the studied period by using the periodicmethod.

242

In order to eliminate high year-to-year variation of runoff in load estimates, the annual flow-weighted concentrations were calculated by dividing the total annual load by the total annual runoff. Nutrient loads for 2010-2012 at Savijoki were also calculated according to the water quality samples measured in the laboratory and according to the continuous monitoring data.

247

248Trend analysis249

250 The non-parametric Mann-Kendall test was used for detection of monotonic trends in nutrient concentration and 251 runoff time series. Trend analysis was done for the monthly means of TP, DRP, TN and NO₃-N concentrations 252 as well as for runoff for each small catchment. For this, we used the Multitest application (a VBA macro 253 running in MS Excel) developed in Linköping University, Sweden. The basic principle of the Mann-Kendall 254 trend test is to examine the signs of pairwise differences in the examined time series so that all previous 255 observation values are subtracted from every observation value. If the sum of signs (test statistic) is zero there is 256 no trend, if positive, the trend is increasing and if negative, the trend is decreasing. The statistical significance 257 (*p*-value) of the trend depends on how much the test statistic differs from zero.

258

The Multitest application is able to calculate the trend and its significance not only over the whole time series, but also for each month separately according to principles presented by Hirsch and Slack (1984). This feature is particularly useful in Finnish conditions, where seasonal variation of flow is high. Indeed, in terms of total annual loading, the concentrations during the months with high flow are much more significant than those observed during the months with low flow.

264

The trend analysis period was 1981-2011 for the concentrations and 1981-2010 for runoff. However, for P concentrations the trends were analysed for periods beginning in 1987 due to a fundamental change in laboratory procedure of P analyses. Statistical significance of the trends was divided as follows: strongly

268 269 270	significant (***, p<0.001), intermediately significant (**, $0.001) and weakly significant (*, 0.01).$
271 272	Continuous water quality monitoring
273	Continuous water quality and discharge measurements of the Savijoki catchment were utilised to compare
274	traditional nutrient loading estimates, e.g. those calculated by the periodic method from the data collected by
275	grab sampling, to the hourly data collected automatically. The measured concentrations for turbidity and NO ₃ -N
276	were calibrated with the laboratory data. In total, 139 data pairs were utilised in order to form the calibration
277	equations. The calibration equations and R^2 values are shown in Table 4. In addition, the conversion equations
278	were also calculated for TP load with on-line measured turbidity data and TN load with on-line measured NO3-
279	N data.
280	
281	Table 4. Calibration equations for turbidity (FTU) and nitrate nitrogen (NO ₃ -N, mg l ⁻¹), and conversion
282	equations for total phosphorus (TP, $\mu g l^{-1}$) and total nitrogen (TN, mg l ⁻¹).
283	
284 285	Results
286	Precipitation and variability in runoff and nutrient concentrations between catchments
287 288	During 1981-2010 the average annual precipitation sums decreased slightly (2-7%) in all the other catchments
289	except for two northern areas. Myllynuro and Laanjoja, where the increases in annual average precipitation were
290	9% and 5%, respectively (Table 5). As a result of lesser rainfall, the annual average runoff typically decreased
291	by 4-7% in agricultural and forested catchments and by 11% in mixed catchments during the latter period (1995-
292	2010).
293	
294	Table 5. Average annual precipitation (mm) during 1981-1994 and 1995-2010 and the change (%) in
295	precipitation.
296	
297	In Fig. 4, the boxplots of annual average runoff (1981-2010) for all 12 catchments are presented. On the whole,
298	runoff and its variability seem to be rather similar for all land use types. The median runoff varied from 7.0 to
299	13.8 l s ⁻¹ km ⁻¹ . Slightly higher runoff values were observed in the northern forested catchments of Laanioja (13.8
300	l s ⁻¹ km ⁻¹), Vähä-Askanjoki (12.8 l s ⁻¹ km ⁻¹) and Myllypuro (11.8 l s ⁻¹ km ⁻¹). In the southern to south-western
301	agricultural catchments, median runoff varied from 7.0 to 12.8 l s ⁻¹ km ⁻¹ . There was high variability between the
302	years.
303	
304	Fig. 4. Box-plot distributions of annual runoff during 1981-2010 in all 12 small catchments. The catchments (x-
305	axis) are arranged with decreasing share of agricultural area from left to right.
306	

- 307 At the same time, large differences were observed in average TN concentrations in 1981-2010 (Fig. 5) between
- 308 these 12 catchments. The highest median values were measured in agricultural catchments, namely Hovi (4,380
- μ g l⁻¹), Löytäneenoja (4,190 µg l⁻¹) and Haapajyrä (6,320 µg l⁻¹). In the agricultural Savijoki catchment, the
- 310 median concentration was somewhat lower $(2,310 \ \mu g \ l^{-1})$ though slightly higher than in the mixed catchments.
- 311 In forested catchments, the median N concentrations were much lower (range 119-968 μ g l⁻¹). The year-to-year
- 312 variability in nitrogen concentrations was much higher for agricultural than forested catchments. The
- 313 exceptionally high concentrations were measured during the years 2003 (Savijoki), 2004 (Latosuonoja) and
- 314 2006 (Teeressuonoja), i.e. not in the years when the highest flows were measured.
- 315
- 316 Fig. 5. Box-plot distributions of average annual nitrogen concentration during 1981-2010 in all 12 small
- catchments. The catchments (x-axis) are arranged with decreasing share of agricultural area from left to right.

Total P boxplot pattern (Fig. 6) differed somewhat from the above TN pattern. For instance, the variability in annual average TP in 1987-2010 was considerably higher in the 100% agricultural Hovi catchment than in the other agriculture-dominated catchments. The variability was especially low in the forested catchments. The median TP concentration in the agricultural Hovi catchment was 290 μ g l⁻¹, and in the agricultural Löytäneenoja and Savijoki catchments 170 μ g l⁻¹. In the Haapajyrä catchment, which consists mainly of acid sulphate soils with high P retention, the median TP concentration was much lower, 79 μ g l⁻¹. In the forested catchments, the median TP concentration varied between 4-44 μ g l⁻¹.

- 326
- Fig. 6. Box-plot distributions of average annual phosphorus concentration during 1987-2010 in all 12 small
 catchments. The catchments (x-axis) are arranged with decreasing share of agricultural area (from left to right).
- The coefficients of variation for average annual runoff and concentrations of TN, NO₃-N, org-N, TP and DRP
 were considerably higher in the agricultural catchments (average 0.36) than in the forested catchments (average
 0.23). Generally, the coefficients of variation were under 0.40, but higher values were observed in agricultural
 catchments and particularly in NO₃-N, TP and DRP concentrations. In general, the coefficients increased in the
- latter period apart from org-N, which remained at the same level, and DRP, which slightly decreased. No
- 335 significant differences were observed between the different periods (Table 6).
- 336
- Table 6. Coefficients of variation for annual average concentration values during 1981-1994 (for TP and DRP:
 1987-1994) and 1995-2010 in the three groups of the studied catchments.
- 339
- 340 Long-term nutrient loads in 1981-2010
- 342 Nitrogen loads in the agricultural and mixed catchments
- 343

341

- 344 The TN loads were highest in the agricultural catchments, with an average annual load of 15.6 kg ha⁻¹ yr⁻¹ for
- $345 \qquad \text{Hovi, 13.2 ha}^{-1} \text{ yr}^{-1} \text{ for Löytäneenoja, 16.1 kg ha}^{-1} \text{ yr}^{-1} \text{ for Haapajyrä and 8.2 kg ha}^{-1} \text{ yr}^{-1} \text{ for Savijoki. These}$
- 346 values include load from all types of land use within the catchment. Of these catchments, only Hovi represents a

- 347 totally agricultural area. In spite of this, the annual mean load of Hovi was lower than in the Löytäneenoja catchment where the agricultural area covers 69%. The maximum annual loading varied from 13.6 kg ha⁻¹ yr⁻¹ 348 349 (2004, Savijoki) to 33.3 kg ha⁻¹ yr⁻¹ (2000, Hovi). Nitrate-N accounted for 57-72% of TN in these agricultural 350 catchments. Flow-weighted TN concentration seemed to increase in the Löytäneenoja and Haapajyrä catchments 351 (Fig. 7). 352 353 Fig. 7. Annual loads and flow-weighted concentration of TN and TP in the agricultural catchments. 354 In the mixed catchments, the average annual TN load was 3.6 kg ha⁻¹ yr⁻¹ (Ruunapuro) and 6.4 kg ha⁻¹ yr⁻¹ 355 356 (Latosuonoja). The flow-weighted TN concentration seemed to increase in the Latosuonoja basin, but the 357 variability between the years was very high (Fig. 8). 358 359 Fig. 8. Annual loads and flow-weighted concentration of TN and TP in the mixed catchments.

361

360 Phosphorus loads in the agricultural catchments

362 The TP loads were also highest in the agricultural catchments as expected, with average annual loads of 1.34 kg

363 ha⁻¹ yr⁻¹ for Hovi, 0.46 kg ha⁻¹ yr⁻¹ for Löytäneenoja, 0.23 kg ha⁻¹ yr⁻¹ for Haapajyrä and 0.62 kg ha⁻¹ yr⁻¹ for

Savijoki. The maximum estimated annual TP load was 2.75 kg ha⁻¹ yr⁻¹ (Hovi, 2000). As contrary to TN, no 364

365 long-term changes in flow-weighted TP concentration could be seen, but year-to-year variability was extremely

366 high at the Hovi catchment. The share of DRP of the TP load was highest (27.5%) in the Löytäneenoja

367 catchment, while at Haapajyrä it was only 5.9% (Fig. 7).

368

369 Nitrogen loads in the forested catchments

370

371 Fairly low annual average TN loading values were obtained in the forested catchments (1.8 kg ha⁻¹ yr⁻¹), while 372 14% of the TN loads, on average, were obtained in the agricultural catchments (13.1 kg ha⁻¹ yr⁻¹). In the forested 373 catchments, the average annual TN load varied from 0.6 to 3.0 kg ha⁻¹ yr⁻¹. The average annual TN loading was 374 lowest in the northern catchments, which was less than the often-used average TN background loading estimate 375 for Finland (1.4 kg ha⁻¹ yr⁻¹, Mattsson et al. 2003). Flow-weighted TN concentrations seemed to increase in the 376 Teeressuonoja and Huhtisuonoja catchments (Figs. 7 and 9). Nitrate-nitrogen accounted for only 3.6% of TN in 377 Myllypuro and up to 48% in Teeressuonoja. The average share (NO₃-N/TN) for all forested catchments was

- 378 18%, which was much less than in the agricultural catchments. This means that most of the TN is leached in
- 379 organic form in these forested catchments (Fig. 9).
- 380

381 Fig. 9. Annual loads and flow-weighted concentration of TN and TP in the forested catchments.

382 3.2.4. Phosphorus loads in the forested catchments

383

384 The annual average TP loads in the forested catchments were also at the modest level (0.07 kg ha⁻¹ yr⁻¹), which 385 is on average only 11% of the TP loads in the agricultural catchments (0.65 kg ha⁻¹ yr⁻¹). The TP loads varied

from 0.02 to 0.11 kg ha⁻¹yr⁻¹. Dissolved phosphorus (DRP) accounted for 26% of TP on average, which is of
the same order as in the agricultural Löytäneenoja catchment, but much higher than in all the other agricultural
catchments. Flow-weighted TP concentrations and loads seemed to decrease in the Vähä-Askanjoki and
Huhtisuonoja catchments (Fig. 9).

390

391 Averaged TN, NO₃-N, TP and DRP loads during the periods 1981-1994 and 1995-2010 are given in Table 7 for 392 different catchment types. In 1995 Finland joined the EU and started to implement agri-environmental water 393 protection measures of the FAEP. This means that the first period of 1981-1994 is a reference period when no 394 measures were taken. In agricultural catchments, nitrogen loads, both TN and NO₃-N, increased from the first to 395 the second period by 10% and 29%, respectively. However, a small reduction can be seen in TP loads (9%), 396 whereas DRP loads have increased (33%). The situation in the mixed catchments is ambivalent: in Ruunapuro 397 the water protection measures have had a positive effect while in Latosuonoja nitrogen loads (both TN and NO3-398 N) and TP loads have increased. In the forested catchments, the reduction of both TP and DRP loading can be 399 seen in most catchments. TN loads have decreased in Kesselinpuro, Myllypuro and Vähä-Askanjoki, while in

- 400 Teeressuonoja and Huhtisuonoja they have increased.
- 401

402 Table 7. Total nitrogen (TN), nitrate-nitrogen (NO₃-N), total phosphorus (TP) and dissolved reactive

phosphorus (DRP) loads (kg ha⁻¹ yr⁻¹) during 1981-1994 (1987-1994 for phosphorus) and 1995-2010 in the three
 groups of the studied catchments.

405 406

406 Loads related to percentage of arable land in catchments 407

We found a connection between the percentage of arable land in a catchment and average nutrient load from it
(Fig. 10). Here, the Haapajyrä catchment, which is the only one on acid sulphate soils, was excluded from the
analysis due to its specific character (high P retention in acid sulphate soils) discussed earlier by Rekolainen
(1989) and Vuorenmaa et al. (2002).

412

Fig. 10. Relationships between percentage of arable land in 11 study catchments and average TN, NO₃-N, TPand DRP loads. 95 % confidence limits are also shown.

415

416 Percentage of arable land explained 90% of the variability of TP losses and 95% of the TN losses, respectively.

From these equations (Fig. 10), we estimated the average background losses, including also forestry and other
sources, as follows: for TP 0.04 kg ha⁻¹yr⁻¹, for DRP 0.02 kg ha⁻¹yr⁻¹, for TN 1.85 kg ha⁻¹yr⁻¹ and for NO₃-N 0.65
kg ha⁻¹yr⁻¹.

420

421 The specific agricultural loads can be assessed from the equations in Fig. 10 (when the share of arable land is set

422 to 100%). Then the agricultural loads are 1.1 kg ha⁻¹yr⁻¹ for TP, 0.18 kg ha⁻¹yr⁻¹ for DRP, 15.5 kg ha⁻¹yr⁻¹ for TN

423 and 11.1 kg ha⁻¹yr⁻¹ for NO₃-N. These specific agricultural loads were extrapolated to average agricultural

424 nutrient loads in surface waters on a national scale. Here, the specific loading values for 1981-1995 are taken

from Vuorenmaa et al. (2002), and the following years are based on this study. As shown in Table 8, the

426 agricultural land area in 1996-2000 is smaller than before, mainly due to the fact that many small farms went out

- 427 of business after Finland joined the EU in 1995. Recently, the area of used agricultural land has risen again due
- 428 to the increased renting of agricultural land, but even at present, the total area is not at the same level that it was
- 429 in 1981-1994. The range in total TN loading to surface waters varies between 34,000-37,000 t yr⁻¹ (Table 8) and
- 430 between 2,400-2,700 t yr⁻¹ for TP, being lowest in 1996-2000 when agricultural land area was smallest. The
- 431 estimated specific load value used in calculations increased by 0.5 kg ha⁻¹ yr⁻¹ for TN for the latter half (from
- 432 1996 onwards) and remained the same for TP.
- 433

Table 8. Total area (1000 ha) of agricultural land and specific (kg ha⁻¹ yr⁻¹) and total (t yr⁻¹) agricultural nutrient
loads to surface waters in Finland in six consecutive five-year periods.

436

437 Trend analysis 438

439 Statistically significant decreasing runoff trends in 1981-2010 were found in the Löytäneenoja (21), Ruunapuro 440 (71) and Laanioja (121) catchments. In Ruunapuro, the decrease occurred mostly in autumn, while in Laanioja 441 the downward trend was due to the strongly declined winter runoff (Table 9). Due to a couple of wet years 442 experienced in Finland in the early 1980s, the runoff trends starting from 1981 were generally declining. An 443 exception to this was the Myllypuro (103) catchment, where the winter-time runoff has clearly increased 444 although the total trend is not statistically significant (Table 9).

445

Table 9. Trends of runoff in the studied catchments during 1981-2010. Dark grey cells denote upward and light
grey cells downward trend with *** = strong, ** = intermediate or * = weak statistical significance. Empty cell
and light
and light</

449

450 As for TP concentrations, the trends in 1987-2011 were mostly decreasing (Table 10). The strongest downward

- trends were found in the eastern and forested catchments of Huhtisuonoja (44) and Kesselinpuro (51), both of
- 452 which showed total P decline particularly in spring, with Huhtisuonoja also showing a decline in autumn.
- 453 Statistically significant downward total P trends were also found in the northern Vähä-Askanjoki (114)

454 catchment and in the agricultural Savijoki (22) and Haapajyrä (81) catchments. In terms of DRP concentrations,

- the trends in non-agricultural catchments were by and large in line with those of TP, but the statistical
- 456 significance was generally stronger (Table 10). Except for three agricultural catchments and one mixed
- 457 catchment (Ruunapuro 71), all DRP trends were significantly decreasing, mostly in forested catchments.
- 458 Significantly upward DRP trends were found in the agricultural Hovi (11) and Löytäneenoja (21) catchments,
- both of which had increasing trends occurring in autumn (Table 11).
- 460
- 461 Table 10. Trends of total phosphorus concentrations in the studied catchments during 1987-2011. Dark grey
- 462 cells denote upward and light grey cells downward trend with *** = strong, ** = intermediate or * = weak
- 463 statistical significance. Empty cell = no statistically significant trend.
- 464

- 465 Table 11. Trends of dissolved reactive phosphorus concentrations in the studied catchments during 1987-2011.
- Dark grey cells denote upward and light grey cells downward trend with *** = strong, ** = intermediate or * =
- 467 weak statistical significance. Empty cell = no statistically significant trend.
- 468
- 469 Total N concentrations in 1981-2011 showed predominantly increasing trends (Table 12). In agricultural
- 470 catchments there was an intermediately significant upward trend in Löytäneenoja (21) and a strongly significant
- 471 upward trend in Haapajyrä (81). The forested Huhtisuonoja (44) catchment also showed a strongly increasing
- trend of total N concentration. The only statistically significant decreasing total N trend was detected in the
- 473 northern Vähä-Askanjoki (114) site (Table 12).
- 474
- Table 12. Trends of total nitrogen concentrations in the studied catchments during 1981-2011. Dark grey cells
 denote upward and light grey cells downward trend with *** = strong, ** = intermediate or * = weak statistical
 significance. Empty cell = no statistically significant trend.
- 478

479 In terms of NO₃-N concentrations, the trend in Haapajyrä (81) was strongly increasing. Also in the southern

- 480 Löytäneenoja (21) and Teeressuonoja (14) catchments intermediately significant upward trends of NO₃-N
- 481 concentrations were found. Strongly significant decreasing NO₃-N trends were detected in the northernmost
- Vähä-Askanjoki (114) and Laanioja (121) catchments. Seasonally, the upward NO₃-N concentration trends
 seemed to be mostly due to the increases during the period from September to December. Meanwhile, in the
 downward NO₃-N trends detected in Vähä-Askanjoki and Laanioja, both spring and autumn periods played a
 role (Table 13).
- 486

Table 13. Trends of NO₃- nitrogen concentrations in the studied catchments during 1981-2011. Dark grey cells
denote upward and light grey cells downward trend with *** = strong, ** = intermediate or * = weak statistical
significance. Empty cell = no statistically significant trend.

490

491 Continuous water quality monitoring – the Savijoki case 492

- 493 The produced calibration equations (Table 4) were used to convert raw turbidity and NO₃-N concentration data
- into TP and TN concentrations. First, the calibrated automatic turbidity data was compared with the
- 495 corresponding laboratory measurements (Fig. 11). Even though the annual dynamics in turbidity was well-
- 496 captured with the on-line automatic sensor, there was more difference between measured and calculated data
- 497 during 2012 and especially during 2013 than in the beginning of the monitoring period. If a 10 per cent
- 498 deviation (< | 6.7 | FTU) from the mean concentration (2010-2013) is used as a criterion for "good data," 62
- per cent of the data passed the test. If the criterion is set to 20 per cent (< | 13.4 | FTU), 83 per cent of the data
- 500 could be classified as good. For NO₃-N, the visual fit between measured and calculated data was even better
- 501 than in the case of turbidity, but here the 10 per cent criterion ($< |0.138| mg l^{-1}$) gave only 40 per cent "good
- data," whereas the 20 per cent criterion gave 67 per cent "good data."
- 503

- Fig. 11. Hourly time series of the calibrated turbidity measured by optical sensors (NTU) together with turbidity
 analysed from water samples. The difference between measured and calculated values is shown with open
 circles.
- 507

508 Monthly TN and TP loads calculated with the periodic method and with the continuous water quality

- 509 measurements (1 hour frequency) are presented in Fig. 12. Most loading occurred either during the spring
- 510 snowmelt period or during late autumn, with substantial variability between the years. For both nutrients, the
- 511 greatest difference was observed during the high flow peaks. In 2010, the TN peak load was underestimated
- 512 with the Period method (PeriodMeth), while in 2011 and 2012, the peak load was overestimated with it. The
- 513 Continuous measurements -method (ContinMeth) is assumed to give the most correct load estimate. For TP
- 514 loads, the difference was greatest in 2010. The average annual loads for TN and TP are presented in Table 14.515
- 516 Fig.12. Monthly TN and TP loads in the Savijoki catchment in 2010–2012 as calculated on the bases of (i)
- automatic monitoring and (ii) daily flow and water sampling data (load calculated with the periodic method).
- Table 14. Annual average loadings (kg ha⁻¹yr⁻¹) for total nitrogen (TN) and total phosphorus (TP) in the Savijoki
 catchment as calculated with period method (PeriodMeth) and with 1-hour continuous water quality data
 (ContinMeth).
- 522

526

523 Discussion 524

525 Agricultural and mixed catchments

High differences were observed in nutrient concentrations between the agricultural catchments, explained
mostly by varying share of arable land within the catchments. Other contributing factors include soil types,
different agricultural practices, the amount of use and efficiency of water protection measures, and hydrometeorological variability. In mixed catchments the nutrient concentrations were lower than in the agricultural
catchments but higher than in the forested catchments.

532

533 In Finland, the major share of nutrient loading has originated from agriculture for decades (Rekolainen et al.

- 534 1997; Vuorenmaa et al. 2002), and the common pattern remains the same. The average TN load in agriculture-
- dominated catchments was 13.3 kg ha⁻¹yr⁻¹, i.e. 9 times higher than the average background N loss value of 1.40
- 536 kg ha⁻¹yr⁻¹. Respectively, the average P load in the four agricultural catchments here was 0.66 kg ha⁻¹yr⁻¹, i.e. on
- 537 average 12 times higher than the most often used background P loss value for Finland (0.054 kg ha⁻¹yr⁻¹ based
- 538 on 21 natural-state catchments, Mattsson et al. 2003). Spatially, a mosaic of a large number of crops, several
- 539 different agricultural practices and water protection measures are used in agriculture. Temporally, fertilisation
- 540 and ploughing are common practices almost every year, with year-to-year variability.
- 541
- 542 Detected changes based on trend analysis and load time-series 543

- Statistically significant decreasing runoff trends in 1981-2010 were found in Löytäneenoja, which could partly
 explain the increase in total N and NO₃-N concentrations in the summer period, but not during autumn or
 wintertime.
- 547

548 There was no increase in TP concentrations. The increase in DRP concentrations in some areas can be a result of 549 the increased non-plough cultivation and direct sowing, which have taken place in almost two thirds of the 550 earlier autumn-ploughed fields. Gonzales-Inca et al. (2016) found increasing trends in concentrations and loads 551 of dissolved reactive phosphorus (DRP) and decreasing trends in TSS in the agricultural Yläneenjoki river 552 basin: these trends might be linked to the current effort of agricultural soil erosion reduction. Decreased tillage 553 or no-tillage methods increased in the Yläneenjoki catchment from 17% to 39% of arable land area between 554 2000 and 2010 (Aakkula and Leppänen 2014). Several other studies carried out in Finland found that the 555 mobilisation of DRP increases in areas with reduced tillage depth or no-till (Koskiaho et al. 2002; Uusitalo et al. 556 2007), and in vegetated buffer zones (Uusi-Kämppä and Jauhiainen 2010). It is also notable that the average 557 fertilisation rates have decreased from 40 kg P ha⁻¹ to 8 kg P ha⁻¹ (see Fig. 2) and, in addition, farmers have put 558 many other water protection measures, e.g. constructed wetlands and controlled drainage, into operation. 559 Nevertheless, TP concentration decreased in only half of the catchments.

560

561 In Estonia, Iital et al. (2013) investigated temporal dynamics in N concentrations and losses in small catchments 562 for the period 1992-2011. The studied areas represented various agricultural production areas. The Mann-563 Kendall trend test revealed one (of eight) statistically significant upward trend in TN concentration. A less 564 significant upward trend in TN was also noted in two other streams. None of the streams showed a statistically 565 significant downward trend in nitrogen concentration, correspondingly as in our study. Iital et al. (2013) also 566 conclude that a proper understanding of nutrient loss processes at the small catchment scale requires updated 567 data on actual agricultural practices. In southern Europe environmental land use conflicts within catchments 568 have recently been recognized as major cause of soil and nutrient losses to rivers (Pacheco et al. 2014; Valle 569 Junior et al. 2014).

570

571 Kyllmar et al. (2006) investigated 27 small agricultural catchments in Sweden for 9-14-year study periods

572 starting in the late 1980s. Significant downward trends were revealed for NO₃-N, DRP and particulate P (PP) for

573 seven, eight and three catchments (out of 24 catchments), respectively, whereas upward trends were revealed for

574 NO₃-N and PP for only one and two catchments, respectively. The downward trend for NO₃-N was explained by

- 575 lesser amounts of manure applied, and a change in season for manure application and crop distribution, i.e. more
- 576 winter-sown crops and fallow. Significant downward trends were also detected for DRP. Kyllmar et al. (2006)
- 577 also concluded that the measures undertaken so far show in general a better response for NO₃-N and DRP, as
- 578 compared to PP. They also argued that a more thorough evaluation of agricultural data is needed to fully explain
- trends and non-trends in time series. However, in that Swedish study, the measures seem to respond better as
- 580 downward trends in NO₃-N and DRP than in Finland. In Finland, more erosion control measures have been

581 implemented.

582

- 583 In Denmark, the trends have been somewhat different. Windolf et al. (2012) conclude that the TN load to ten
- estuaries (catchments covering 35% of the Danish land area) has decreased by 39% during the period 1990-
- 585 2009. They also report an 18-55% reduction for the mean flow-weighted TN concentrations in inlet to estuaries.
- 586 Targeted and catchment-specific measures are the most cost-efficient way to achieve good chemical quality in
- 587 estuaries (Windolf et al. 2012). In Finland, the implementation of measures has not been well-targeted and
- 588 widely applied so far, which might explain rather limited improvements in water quality. On the other hand, the
- 589 concentration levels have been and are evidently higher in Danish water bodies than in the Finnish ones (see
- 590 EEA Wise database), which gives more possibilities for load reductions in Denmark than in Finland.
- 591

592 Efficiency of water protection measures 593

The efficiency of the water protection measures in agriculture is generally assessed at the field plot scale and the measures are then ranked according to different practices and techniques (e.g. Puustinen et al. 2007; Uusi-Kämppä et al. 1998; Ekholm et al. 2011; Salomon and Sundberg 2012). This is normally a good starting point for implementing the measures. However, from the perspective of the river basin scale, the scaling issues should be taken into account: what assortment and volume of measures will give noticeable impacts even at the

- 599 outlet of the river basin?
- 600

601 Some notable and wide-ranging changes took place in Finnish agriculture after Finland joined the EU in 1995. 602 The decrease in the use of fertilisers (Fig. 2) has led to reduced balances of nitrogen and phosphorus, the 603 reduction being 35% and 60%, respectively, from the earlier levels (Aakkula & Leppänen, 2014). The autumnal 604 ploughed area has declined by nearly one third from the earlier levels. Autumnal ploughing has been replaced 605 by reduced tillage, wintertime stubble or direct sowing. In total, the measures practicable at each field plot -i.e.606 in very large scales - will therefore be potentially efficient in nutrient reduction efforts. At the Hovi catchment 607 of this study, the tillage practices vary from year to year, but the major change was clear: autumn ploughing has 608 decreased from 100% in 1981-1987 to 0-30% in 2005-2010, replaced by reduced tillage methods. This may 609 have had a stabilising effect on nutrient loading: the nutrient concentrations have been stable (not increasing), 610 but for DRP there is indication of increasing concentration trends, particularly in autumn (Table 9) – probably 611 due to the increased reduced tillage practices.

612

613 The question remains why there are only a few significant downward trends especially for nitrogen at the

- 614 catchment scale despite all the efforts made. It is obvious that the number of efficient measures, in particular for
- 615 nitrogen, is still too modest. Factors related to climate change probably also contribute to another direction (e.g.
- Blombäck et al. 2012; Børgesen and Olesen 2011; Hägg et al. 2014). In Finland, the most wide-ranging
- 617 measure, i.e. wintertime vegetation cover, seems to work for PP, but there are signs of mobilisation, and parallel
- 618 increases in DRP concentrations and loads (e.g. Uusitalo et al. 2007; Gonzales-Inca et al. 2016).
- 619

Due to these aspects, we cannot say that the implemented water protection measures have been successful
enough to cut down the agricultural loading, even though the efficiency of many measures is shown at the field
plot level. There are some positive signs of decreases in total P loading, but not for N loading.

623

624 Forested catchments 625

626 High variability was observed in nutrient concentrations between the forested catchments, explained by i)

627 different intensity of forestry practices (drainage and supplementary drainage, cuttings, fertilisations etc.) over

the years, ii) by natural characteristics, and iii) by hydrometeorological variation. The most obvious changeshave been due to the changes in forestry, with some examples given below.

630

631 Forest loads - average levels and as compared with background loads 632

The average TN load here, 1.75 kg ha⁻¹yr⁻¹, was about the same as an earlier estimate for the average TN load
for managed forests, 1.83 kg ha⁻¹yr⁻¹ by Kortelainen & Saukkonen (1998). The average TP load, 0.07 kg ha⁻¹yr⁻¹,
was clearly lower (30%) than the earlier estimate of 0.10 kg ha⁻¹yr⁻¹ by Kortelainen & Saukkonen (1998). This
means that TP loading from managed forests seems to have decreased in large areas, most probably due to
decreased forest fertilisation, discussed more in detail below.

Average TP load in the six managed forested catchments was on average 1.3 times higher than the most often

640 used background P loss value for Finland, 0.054 kg ha⁻¹yr⁻¹ (Mattsson et al. 2003). Respectively, average N load

 $641 \qquad \text{was } 1.75 \text{ kg ha}^{-1} \text{yr}^{-1}, \text{ i.e. } 1.3 \text{ times higher than the average background N loss value of } 1.40 \text{ kg ha}^{-1} \text{yr}^{-1}. \text{ This}$

- 642 means that forest management increases loading from forests by one third, on average. Spatially, the mosaic of a
- huge number of treatment areas is typical in forestry. Temporally, treatment areas vary from year to year, and
- 644 the impacts of a single treatment may last from a couple of years to >10 years.
- 645

638

646 Detected changes - based on trend analysis and load time-series 647

648 The TN concentrations increased in most of the forested sites. They increased significantly in Huhtisuonoja and

649 Myllypuro with no changes in NO₃-N, which means that dissolved organic N concentrations (DON) are

650 increasing. In these catchments the percentage of organic peat soils is relatively high, i.e. 45% and 27%,

respectively. This DON increase seems to be related to increasing trends of winter flows (increasing trends inflow from November to January) particularly in Myllypuro. In Huhtisuonoja there is an increasing, but not

flow from November to January) particularly in Myllypuro. In Huhtisuonoja there is an increasing, but notstatistically significant, trend in annual flow in 1987-2010. Correspondingly, increasing trends in winter flow

- 654 were detected in the northern, forest and peatland -dominated river basin, together with increasing trends in
- TON (total organic N) concentrations in autumn mid-winter (Lepistö et al, 2008).
- 656

Practically all of the organic peat soils of Huhtisuonoja and Myllypuro catchments had been drained for forestry
in 1960-1992 (Kortelainen and Saukkonen 1998). According to Keinänen (2013), only 2.1% of Huhtisuonoja
and 5% of Myllypuro remained undrained in 2008.

660

661 The delivery of terrestrial DON to aquatic systems depends on production/decomposition rates, solubility and

the availability of hydrological transport. The leaching of organic matter, particularly DOC but also DON,

typically increases strongly with peatland proportion. Pellerin et al. (2004) found that 79% of the variance in

664 DON concentrations in streams and rivers in the northeastern U.S. was explained by the percentage of wetlands

- in a catchment. According to a recent study in Finnish watersheds, wetlands were found to play an important
 role: most of the TOC and TON were transported during the high flow following the spring snowmelt and
 during autumn rainfall (Mattsson et al. 2015).
- First time drainage works were ending in Finland in the early 1990s (Forest statistical yearbook). Supplementary
 drainage (ditch maintenance) areas increased all through the 1990s and 2000s on a national scale, and also in the
- 671 research catchments of Huhtisuonoja and Myllypuro, but the catchment-scale data of these areas is unfortunately
- 672 not available. Increasing winter flows in these drained, forested catchments most obviously increase leaching
- 673 losses of organic matter, detected here as increasing trends in DON. In many of these forested catchments
- studied by Keinänen (2013), particularly in Huhtisuonoja, the clearly increased concentrations of TOC havebeen detected.
- 676

677 On the other hand, in the southern Teeressuonoja site, an increase was detected in both total N and NO₃-N

678 concentrations. The increase in NO₃-N relates probably to increases in nitrification in catchment soils due to

679 several performed clear-cuttings in 1985-2001 during our monitoring period. It has been stated that considerable

nitrification in coniferous forests is possible after clear-cutting (e.g. Tamm et al. 1974), when plant nutrient

uptake is decreased and pH is increased. Together, clear-cuttings cover a total of 28% of the Teeressuonoja

- 682 catchment (Keinänen 2013).
- 683

For TP concentrations in forested catchments, the trends in 1987-2011 were mostly decreasing, and significantly
so in three of the six catchments, while dissolved DRP concentrations decreased significantly in all forested
catchments. At the same time P fertilisation in Finnish forests decreased significantly. The average fertilised
forest areas in Finland dropped to one tenth: from 70,000 ha/year in the late 1980s (1986-1990) to 7,000 ha/year
in the early 1990s (1991-1995), remaining relatively low during the late 1990s and 2000s (Fig. 2) (Metla Forest
Statistical Yearbook 2014).

690

P fertilisation plays a significant role with impacts on P losses: the application history of P fertilisation was detected as the most important predictor for the spatial variation in total P leaching from 22 small, managed and forested catchments in Finland (Kortelainen and Saukkonen, 1998). Fertiliser P along with potassium (K) is applied to peatland forests to increase their productivity. Leaching is typically highest in the first and second year after fertilization, and P concentrations in the outflow water may remain at a higher level than before fertilising for several years, even for over a decade (Ahtiainen and Huttunen 1999; Joensuu et al. 2001; Väänänen 2008).

698

This decrease of P fertilisation is very probably the main reason for decreased P concentration trends (total P
and DRP) in most of the forested sites. At Huhtisuonoja, specifically, extensive P fertilisation of 70% of the
total area of peatlands of the catchment took place in 1985-1986, just before the monitoring period (Pietiläinen
and Rekolainen 1991). This can be clearly seen in high detected P concentration levels of 80-90 μg l⁻¹ in 1987-

- 703 1989 (Fig. 9), with gradually decreasing concentrations and losses after that. Both total P and DRP
- 704 concentrations had statistically significant decreasing trends (Tables 9, 10).
- 705

4.3 Agricultural load on a national scale707

708 Strong connections were found between the percentage of arable land in a catchment and the average nutrient

- load from it (Fig. 10). The percentage of arable land explained 90% of the variability of TP losses and 95% of
- 710 the TN losses, respectively. The specific agricultural loads were extrapolated to average agricultural nutrient 711 loads on a national scale. The range in estimated total TN loading varies between 34 000-37 000 t y^{-1} (Table 8)
- and between 2 400-2 700 t y⁻¹ for TP. The agricultural land area in 2000-2010 was smaller than before (1981-
- 713 1995), which was mainly due to the decrease in the number of small farms. This study suggests that the specific
- nutrient loading is a bit higher for TN than earlier (Vuorenmaa et al. 2002) but is at the same level for TP. The
- new load estimates are of the same order than those reported earlier and variation is mostly due to changes in
- 716 agricultural area.

717 **5.** Conclusions 718

Total N concentrations increased in two of the four agricultural sites in 1981-2010. They increased significantly
in Löytäneenoja and Haapajyrä with concomitant change in NO₃-N. Statistically significant decreasing runoff
trends in 1981-2010 were found in three of the twelve sites and only one increasing trend was found in
Myllypuro. The TN concentrations increased in most of the forested sites. Total N concentrations increased
significantly in two forested sites with no changes in NO₃-N, which means that organic N concentrations (DON)
are increasing. Increased winter temperatures seem to contribute to increasing mineralization and increasing
organic-N losses, and play a role in N cycles of both agricultural and forested catchments.

726

727 For TP concentrations, the trends in 1987-2010 were decreasing in two of the four agricultural catchments, 728 while DRP concentrations decreased in one of the four catchments and increased in two others. There was no 729 increase in TP concentrations. The increase in DRP concentration may be a result of the increased non-plough 730 cultivation and direct sowing which have taken place in almost two thirds of earlier autumn-ploughed field 731 areas. For TP concentrations in forested catchments, the trends in 1987-2011 were mostly decreasing, and 732 significantly so in three of the six catchments, while DRP concentrations decreased significantly in all forested 733 catchments. At the same time P fertilization areas in Finnish forests decreased significantly, which has 734 obviously contributed to decreasing concentrations and loads.

735

Why can only a few significant downward trends be detected, especially for nitrogen, at the agricultural catchment scale despite all the efforts made? It is obvious that the extent of efficient measures, in particular for nitrogen, is still too modest. In Finland, the most wide-ranging measure, i.e. wintertime vegetation cover, seems to work for PP, but there are signs of parallel DRP load increases. More efforts with wide-ranging and carefully targeted implementation of water protection measures are needed in order to get visible impacts on the catchment scale.

742

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