REPORT

Predicting the export and concentrations of organic carbon, nitrogen and phosphorus in boreal lakes by catchment characteristics and land use: A practical approach

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Received: 24 August 2015 / Revised: 31 March 2016 / Accepted: 27 April 2016 / Published online: 1 June 2016

Abstract The majority of C, N and P in boreal lakes are in organic form. Organically bound nutrients are released through biodegradation or photodegradation which affects the water quality, eutrophication and greenhouse gas emissions of lakes. We tested whether open land-use data combined with land-use-specific export coefficients can be used to predict total organic carbon (TOC), dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) loading and lake water concentrations. Using data from 12 lake catchments in eastern Finland, we found that land use and management of the catchment explained a substantial proportion of the variations in TOC ($r^2 = 0.78$), DON ($r^2 = 0.55$) and DOP ($r^2 = 0.80$) concentrations between lakes. The computation does not account for in-lake processes, which are reflected as mismatch between the predicted and observed concentrations. However, this simple practical approach is useful in ranking lakes according to their water quality. The results indicated that natural sources dominate TOC, DON and DOP exports; the background leachings accounted for 57–99 %, 48–96 % and 55–99 % of TOC, DON and DOP export, respectively. The proposed method has promise as a practical decision support tool for assessing the impacts of land use on water quality. The results showed that possibilities to control TOC, DON and DOP loading to surface waters are limited to catchments where the peatland proportion is low and anthropogenic sources significant.

Keywords Carbon · Land use · Organic nitrogen · Organic phosphorus · Specific export coefficient · Water quality

INTRODUCTION

The properties and land use of catchments greatly influence surface water quality and the biogeochemistry of aquatic

ecosystems (Allan 2004; Aitkenhead-Peterson et al. 2005; Kortelainen et al. 2006; Mattsson et al. 2009). In Finland and other boreal regions, where catchments land area is dominated by forests and peatlands, many lakes receive naturally high loads of dissolved organic matter (DOM), causing the colour of the water to turn brown (Kortelainen 1993; Sobek et al. 2007). DOM consists of a combination of organic molecules containing carbon and nutrients which can be seen as a correlation between total organic carbon (TOC), dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) (Mattsson et al. 2003; Kortelainen et al. 2006). The fact that TOC is a source of energy (Keskitalo and Eloranta 1999; Wiegner and Seitzinger 2001; Hanson et al. 2003) has many physical, chemical and biological implications for aquatic ecosystems (Keskitalo and Eloranta 1999; Solomon et al. 2015). When present in lakes, TOC affects transparency and thereby the temperature and thermal stratification of lake (Keskitalo and Eloranta 1999; Findlay and Sinsabaugh 2003) and the structure of aquatic food webs (Jansson et al. 2000; Kankaala et al. 2010). It also has impacts on the acid–base chemistry of surface waters (Buffam et al. 2008), C budgets of catchments (Kindler et al. 2011; Schelker et al. 2012), greenhouse gas fluxes (Cole et al. 2007), and the mobility, toxicity and bioavailability of organic pollutants and trace metals such as mercury (Porvari et al. 2003; Akkanen et al. 2004; Ravichandran 2004; Bergknut et al. 2011).

DON and DOP often constitute large and dominant fractions of the total N and P exports in temperate and boreal catchments (Hedin et al. 1995; Kortelainen et al. 2006; Pellerin et al. 2006; Stanley and Maxted 2008). According to a large Finnish lake survey consisting of 874 lakes, DON and DOP account for 92 and 85 % of total dissolved N and total dissolved P concentrations,

respectively (Rantakari et al. 2004). Part of the DON and DOP can be further mineralized, photochemically degraded or nitrified in watercourses (Brookshire et al. 2005; Stedmon et al. 2007) where they can cause eutrophication and increase harmful algal blooms.

TOC, DON and DOP exports are dependent on climatic and hydrologic factors (Sobek et al. 2007; Sarkkola et al. 2009), soil organic matter stocks (Sobek et al. 2007), land use (Pellerin et al. 2006; Mattsson et al. 2009; Kindler et al. 2011) and management such as forest cutting (Schelker et al. 2012), ditching (Joensuu et al. 2002), peat mining (Kløve 2001) and other agricultural practices (Worrall et al. 2012). In the boreal region lake water, C is mainly of allochthonous origin, i.e. derived externally from terrestrial sources (e.g. Fiebig et al. 1990; Jonsson et al. 2001), especially from peatlands (Kortelainen 1993; Dillon and Molot 1997). Typically, TOC, DON and DOP export loads increase with increasing peatland proportion in catchment areas (Dillon and Molot 1997; Aitkenhead-Peterson et al. 2005; Kortelainen et al. 2006). The naturally occurring background leaching of TOC is high in Finland because of the large proportion of peatlands in the Finnish landscape (Mattsson et al. 2003; Kortelainen et al. 2006). The annual background leaching from unmanaged peatland and forest dominated catchments have been reported to be 15–140 kg ha⁻¹ for TOC and 0.2–2.1 kg ha⁻¹ for DON, respectively (Mattsson et al. 2003; Kortelainen et al. 2006). Previous studies have mainly concentrated on quantifying background leaching (Mattsson et al. 2003; Kortelainen et al. 2006) or the effect of certain forest or agricultural management practices on TOC, DON and DOP export in small first order catchments (Joensuu et al. 2002; Nieminen et al. 2010; Puustinen et al. 2010; Schelker et al. 2012; Palviainen et al. 2014; Soinne et al. 2014), and the mutual significance of different TOC, DON and DOP loading

sources in catchments with mixed land use have so far received little attention.

Currently the national and international environmental regulations (e.g. the EU's Water Framework and Marine Strategy directives and HELCOM's Baltic Sea Action Plan) obligate monitoring, planning and control of water quality, which necessitates a practical computation method for assessing the impact of land use on water quality. In this study, we test whether easily available land-use data can be used in the prediction of lake water quality. The objectives of this study were to (1) estimate the export of TOC, DON and DOP from the terrestrial part of the catchment on the basis of geographical information systems (GIS), land-use and land-management data, (2) evaluate how well the background leaching and land use explain the observed TOC, DON and DOP concentrations in lakes, and (3) assess the relative importance of different export sources and consequently evaluate possibilities to control TOC, DON and DOP loadings.

MATERIALS AND METHODS

Study catchments and lakes

Twelve lakes in eastern Finland, whose water colours range from clear to brown, were selected for the study (Table 1; Fig. 1). The aim was to include lakes that (i) are loaded with terrestrial organic C, including peat mining, (ii) have naturally brown- or (iii) clear-water. The catchments of the lakes had variable land uses, which represented typical land-use patterns in Finland (Statistical Yearbook of Finland 2005; Finnish Statistical Yearbook of Forestry 2014). The boundaries of the third-order catchments of the lakes were derived from the Finnish Environmental Centre

Table 1 Characteristics of the study lakes

Lake	Surface area (ha)	Maximum depth (m)	Mean depth (m)	Volume (km ³)	Shoreline (km)	Elevation (m)
1 Hattujärvi	515	9.2	3.3	0.017	29	169
2 Harkkojärvi	437	10.3	3.5	0.015	26	150
3 Mekrijärvi	822	2.7	1.8	0.015	32	144
4 Koitere	16 367	46.5	6.7	1.098	424	143
5 Mujejärvi	351	21.0	4.5	0.016	20	197
6 Nuorajärvi	4024	12.0	2.3	0.093	206	145
7 Kuohattijärvi	1081	18.0	5.6	0.061	29	162
8 Ätäskö	1389	7.5	3.5	0.049	30	80
9 Pyhäjärvi	20 679	27.0	8.1	1.669	477	80
10 Kermajärvi	8557	56.1	10.1	0.866	453	80
11 Ylinen	369	35.0	12.0	0.044	17	120
12 Kuorinka	1292	31.6	10.5	0.136	22	87

Fig. 1 Location of the study lakes

(SYKE) database (<http://www.syke.fi/avointieto>). The mean annual air temperature (1980–2010) was 2–4 °C and the mean annual precipitation 600–700 mm in the study catchments (Pirinen et al. 2012). The mean annual regional runoff was 325 mm (SD 95 mm) during the study period 1999–2013. The areas of peatlands, forests, agricultural fields and surface waters, and built and peat mining area data were derived from the CORINE (CLC2006) land cover (25 × 25 m grid) database (Bossard et al. 2000) provided by the Finnish Environmental Centre (<http://www.syke.fi/avointieto>). Forests on mineral soils and peatlands accounted for 40–65 % and 3–43 % of the catchment area, respectively (Table 2). Forests were dominated by coniferous species, mainly Norway spruce (*Picea abies* Karsten) and Scots pine (*Pinus sylvestris* L.). The proportion of peatlands varied between 3 and 47 % and agricultural land between <1 and 17 % of the catchment area. The build-up

areas consisted of paved surfaces such as urban or industrial areas and road networks, and accounted only for <1–8 % of the catchments. The proportion of surface waters ranged from 8 to 32 % of the catchment areas. All the study catchments, except Pyhäjärvi, were sparsely populated and point-source loading was negligible. In the Pyhäjärvi catchment, the load from wastewater treatment plant of a village with 1500 habitants varied between 10 and 70 kg year⁻¹ for total P and 4400–11 100 kg year⁻¹ for total N (OIVA database; <http://www.syke.fi/avointieto>).

Computation of export loads

The Kustaa computation tool (Launiainen et al. 2014) was modified to compute the export of TOC, DON and DOP. The specific export approach (e.g. Kenttämies 2006) was used to compute the export of compound *k* as:

Table 2 The characteristics of study catchments. The land uses are shown both absolute and relative to total area

Catchment	Area (km ²)	Water (ha/%)	Peatland (ha/%)	Open peatlands (ha/%)	Forests on peatlands (ha/%)	Forests on mineral soil (ha/%)	Agricultural land (ha/%)	Built area (ha/%)	Peat mining areas (ha/%)
1 Hattujärvi	93.4	749.6/8.0	4394/47.0	398/4.3	3996/42.8	4145/44.4	11.3/0.12	40.6/0.43	0/0
2 Harkkojärvi	204.1	1761/8.6	8772/43.0	930.6/4.6	7841/38.4	9682/47.4	30.9/0.15	144.4/0.71	0/0
3 Mekrijärvi	995.3	11 153/11.2	42 183/42.4	12 236/12.3	29 947/30.1	39 783/40.0	2297/2.3	2363/2.4	648/0.65
4 Koitere	2074.5	26 788/12.9	81 689/39.4	16 399/7.9	65 300/31.5	96 432/46.5	842/0.41	1393/0.67	40.8/0.02
5 Mujejärvi	108.7	934/8.6	4246/39.0	391/3.6	3854/35.4	5575/51.3	47/0.4	55/0.5	0/0
6 Nuorajärvi	475.4	9709/20.4	16 641/35.0	4435/9.3	12 206/25.7	19 264/40.5	556/1.2	791/1.7	60/0.13
7 Kuohattijärvi	161.9	1388/8.6	5064/31.3	247/1.5	4818/29.8	8712/53.8	668/4.1	347/2.1	0/0
8 Ätäsö	150.5	1582/10.5	3715/24.7	296/2.0	3419/22.7	6851/45.5	2001/13.3	850/5.6	0/0
9 Pyhäjärvi	804.0	23 473/29.2	12 072/15.0	1119/1.4	10 953/13.6	35 675/44.4	5905/7.3	3198/3.98	0/0
10 Kermajärvi	1652.4	41 466/25.1	14 955/9.1	439/0.3	14 515/8.8	96 358/58.3	6957/4.2	5196/3.1	6.9/0.004
11 Ylinen	31.2	535/17.1	2407.7	7/0.2	233/7.5	2031/65	109/3.5	172/5.5	1.2/0.04
12 Kuorinka	41.5	1309/31.6	126/3.1	2/0.05	124/3.0	1667/40.2	706/17.0	340/8.2	0/0

$$E_{kt} = A_T B_k + A_l D_{kt} + \sum_{m=1}^{n_M} \sum_{k=1}^3 \sum_{t=2009}^{2013} A_{mt} S_{mkt}, \quad (1)$$

where E_{kt} is the total terrestrial export of compound k (TOC, DON or DOP) in year t (kg ha⁻¹ year⁻¹), A_T is the land area of the catchment, B_k is annual background leaching of k (kg ha⁻¹ year⁻¹) which would occur without catchment management, A_l is the lake area (ha), D_k is direct atmospheric deposition to the surface waters (kg ha⁻¹ year⁻¹), A_{mt} is area of land-use managements in year t (ha), S_{mk} is the specific export (kg ha⁻¹ year⁻¹) caused by different land-use and management practices, and n_M is the number of different land-use management methods. The specific export approach is widely used in estimating the impacts of different land-use operations on water quality and in managing lakes, rivers and coastal waters (Kenttämies 2006; Lepistö et al. 2006; Finér et al. 2010; Puustinen et al. 2010). The specific export values are derived from the literature and based on field experiments and catchment-scale studies (Table 3).

Background leaching of TOC (B_{TOC} , kg ha⁻¹ year⁻¹) and DON (B_{DON} , kg ha⁻¹ year⁻¹) were predicted from the areal share of peatlands (A_p , % of catchment area) by using nonlinear models (Eqs. 2 and 3) that we constructed based on the data of Kortelainen et al. (2006), which encompass data from 21 unmanaged Finnish headwater catchments and streams (Fig. 2). The coefficient of determination (R^2) for TOC was 0.78 and for DON 0.57.

$$B_{TOC} = 13.98 (A_p + 1)^{0.45} \quad (2)$$

$$B_{DON} = 0.41 (A_p + 1)^{0.30} \quad (3)$$

Background leaching for DOP (B_{DOP} , kg ha⁻¹ year⁻¹) was made proportional to background leaching of TOC as

$$B_{DOP} = B_{TOC}/697, \quad (4)$$

where the 697 is the mean lake water C:P ratio based on a survey of 874 lakes in Finland (Rantakari et al. 2004).

Land management

Annual forest regeneration areas (Table 4) were compiled from the most recent multi-source National Forest Inventory data (Tomppo et al. 2009), which was collected from the Natural Resources Institute Finland database (<http://kartta.metla.fi/index-en.html>). The stand age in 2011 was used to estimate the time of clear-cuttings, and for the period 2012–2013 the annual average of regeneration areas during 1999–2011 were used. Maintenance ditching areas were received from the Regional Centre for Economic Development, Transport and the Environment records (Table 4). The number of houses and cottages (Table 4) was obtained from the cadastral index map of the National Land Survey of

Table 3 Specific export values ($\text{kg ha}^{-1} \text{ year}^{-1}$) used in the study

Source	TOC	DON	DOP	References
Background leaching	Eq. 2	Eq. 3	Eq. 4	Rantakari et al. (2004) and Kortelainen et al. (2006)
Atmospheric deposition	10	0.60	0.01	Piirainen et al. (2004) and Lindroos et al. (2007)
Final cutting on mineral soils	10	0.40	0.02	Data from Kenttämies and Mattsson (2006), Schelker et al. (2012), and Palviainen et al. (2014)
Final cutting on peatlands	28	1.90	0.05	Lundin (1999) and Nieminen (2004)
Maintenance ditching	–15	0	0	Ahti et al. 1999, Joensuu et al. (2002), and Nieminen et al. (2010)
Peat mining areas	6–30	1.75	0.08	Kløve (2001) and Pöyry (2013)
Agriculture	40	0.80	0.06	Räty et al. (2014) and Soinne et al. (2014)
Livestock	7.3 ^a	0.50	0.10	HELCOM (2011)
Wastewater rural areas ^a	7.3 ^a	0.29	0.05	Hallanaro and Kujala-Räty (2011)

^a Unit kg/inhabitant/year

Table 4 Number of houses and summer cottages on the shores of the lakes, the numbers of livestock, the area of clear-cuttings in forests growing on mineral soil and peatlands and the area of maintenance ditching during 1999–2013

Lake	Houses	Summer cottages	Livestock	Clear-cuttings on mineral soils (ha)	Clear-cuttings on peatlands (ha)	Maintenance ditching (ha)
1 Hattujärvi	7	29	5	110	2	618
2 Harkkojärvi	35	53	14	320	8	790
3 Mekrijärvi	1842	609	1133	3242	99	4131
4 Koitere	405	409	175	3532	160	2282
5 Mujejärvi	15	14	22	150	12	770
6 Nuorajärvi	326	363	111	1058	39	1210
7 Kuohattjärvi	234	85	307	545	59	862
8 Ätäskö	574	110	921	672	38	781
9 Pyhäjärvi	1660	1633	2716	2962	158	1545
10 Kermajärvi	2938	2874	3200	7137	188	870
11 Ylinen	78	150	24	145	7	42
12 Kuorinka	223	146	463	118	2	0

Finland and one inhabitant per house/cottage was assumed. The amount of livestock was obtained directly from the agricultural secretaries of the municipalities (Table 4). The livestock consisted of cattle, beef cattle, pigs and sheep, and no poultry farms existed in the studied catchments.

Specific export of TOC in agriculture has been little studied. Despite being scarce, the existent studies indicate minor differences in TOC export between different agricultural management practices (Soinne et al. 2014; Räty et al. 2014), and thus the averages of the reported specific export values, and the total agricultural area in the catchment (from the Corine database) were used in the calculations. Export of N and P from agriculture is mainly in mineral form (Vuorenmaa et al. 2002), DON and DOP exports were estimated from TOC export by assuming similar C:N and C:P ratios as in background leaching. There are no studies about the specific export of TOC from livestock and it was estimated by using the Rural Areas Wastewater Act's minimum requirement (HELCOM

2011). DON and DOP exports from livestock and wastewater treatment plant were achieved by assuming that 1/3 of the total N and P export are in organic form (Vuorenmaa et al. 2002; Puustinen et al. 2010).

Validation of the model results

The computed total TOC, DON and DOP loads were indirectly validated against measured lake water concentrations. First, the lake water TOC, DON and DOP concentrations that would result from the estimated mean annual export loads were computed by dividing the mean annual export load by the mean annual runoff ($325 \pm 95 \text{ mm}$). Second, the lake water quality monitoring data for the years 1999–2013 were collected from the Finnish Environmental Centre OIVA database (<http://www.syke.fi/avointieto>), and arithmetic averages and standard deviations over the whole period (1999–2013)

Fig. 2 The relationship between the proportion of the catchment covered by peatland and the background leaching of total organic carbon (TOC), the background leaching of dissolved organic nitrogen (DON), and the carbon to nitrogen (C:N) ratio of background leaching (Data from Kortelainen et al. 2006)

were computed for all the lakes. All available TOC concentration data were used in the comparison, while only water quality samples where total and mineral fractions of N and P were measured at the same time were included in comparisons of DON and DOP concentrations. The DON and DOP concentrations were calculated by subtracting the concentration of the mineral forms from total N and P.

Finally, the measured and calculated concentrations were compared using linear regression metrics. In addition, the data were ranked according to the measured and modelled concentrations and Wilcoxon signed rank test was used to evaluate whether the computation could reproduce the rank of the measured lake TOC, DON and DOP concentrations. In this study, the computed concentrations were fully independent of the monitoring data used in the comparison, and no model calibration was done. Further, it is assumed that lake water TOC, DON and DOP concentrations are not significantly affected by the lake processes.

Water quality monitoring

The lake water quality monitoring was performed according to the national guidelines. Water samples for chemical analyses were taken 2–8 times per year at regular depth intervals from the surface to the bottom. The samples were analysed using accredited methods (SFS-EN 45001:1990, SFS-EN ISO/IEC 17025:2000, SFS-EN ISO/IEC 17025:2005) in the laboratories of Regional Environment Centres and Finnish Environmental Centre. The acidified TOC samples were bubbled with nitrogen to remove inorganic carbon (CO_2) and TOC concentrations determined using high-temperature oxidation followed by infrared gas measurements. Total N concentration was determined colorimetrically after oxidization with $\text{K}_2\text{S}_2\text{O}_8$, $\text{NH}_4\text{-N}$ was determined spectrophotometrically with

hypochlorite and phenol, and the sum of $\text{NO}_3\text{-N}$ and $\text{NO}_2\text{-N}$ by the cadmium method. Total P was analysed by the molybdenum blue method after digestion with $\text{K}_2\text{S}_2\text{O}_8$. $\text{PO}_4\text{-P}$ was measured by the molybdenum blue method.

RESULTS

The computed TOC export to lakes varied from 27 to 71 $\text{kg ha}^{-1} \text{ year}^{-1}$ of which the proportion of background leaching was 57–99 % (Fig. 3). The peatland proportion, ranging from 3 to 47 %, was the most important factor determining export and lake TOC concentrations (Figs. 3, 4). The results indicated that also the direct atmospheric deposition to the lake surface can be a significant source of TOC (2–16 %). Agriculture was the largest anthropogenic source of TOC (0–21 %), while in general the significance of forestry operations was small, only ≤ 1 % of the total TOC export. The modelled TOC concentrations corresponded well ($r^2 = 0.784$) with the measured TOC concentrations even though the model overestimated TOC concentrations (Fig. 4). The overestimation occurs especially in clear-water lakes with low observed TOC concentrations (and low peatland percentage). The residuals of the TOC concentration estimates were on average 4.4 mg L^{-1} . The Wilcoxon signed rank test showed that the ranked modelled concentrations corresponded the ranking of the measured results (Z-value -0.155 , $p = 0.877$).

DON export to lakes varied from 0.80 to 1.22 $\text{kg ha}^{-1} \text{ year}^{-1}$, and that of DOP from 0.04 to 0.10 $\text{kg ha}^{-1} \text{ year}^{-1}$ (Fig. 3). The major proportion of the DON (48–96 %) and DOP (55–99 %) export also originated from background leaching. In relation to land use, DON and DOP export behave in a similar manner as TOC; agriculture is the main anthropogenic source and forestry operations have only a minor effect on the export loads. Land use and catchment management explained substantial proportion of the

Fig. 3 Modelled total organic carbon (TOC), dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) exports for different land-use types in the study catchments

variation in DON ($r^2 = 0.55$) and DOP ($r^2 = 0.80$) concentration in the lakes (Fig. 4). The average residuals for DON and DOP concentration estimates were 44.0 and 12.1 $\mu\text{g L}^{-1}$, respectively. The model underestimated the variability of DON concentrations between the lakes. The

DOP concentrations were generally overestimated but the variability well predicted. The Wilcoxon signed rank test showed that the model was in good agreement with the ranking of the measured DON and DOP concentrations (Z -value < 0.001 , $p = 1.000$).

Fig. 4 Predicted and observed total organic carbon (TOC), dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) concentrations (mean $\pm$ SD) in the study lakes. The numbers of the lakes are presented in Tables 1, 2 and 4. The solid line is the regression line and the dashed line is the 1:1 line. White areas in the topmost figure indicate the criteria for the general water quality classification in Finland (Kortelainen 1993, Finnish Environment Institute SYKE 2005): excellent (water colour <50 mg Pt L⁻¹ corresponds to <8 mg TOC L⁻¹), good (water colour 50–100 mg Pt L⁻¹ corresponds to 8–12 mg TOC L⁻¹), satisfactory (water colour 100–150 mg Pt L⁻¹ corresponds to 12–17 mg TOC L⁻¹) and passable (water colour >150 mg Pt L⁻¹ corresponds to >17 mg TOC L⁻¹)

DISCUSSION

The results indicated that TOC and DOP export and resulting concentrations in boreal lakes can be predicted reasonably well by using simple readily available land-use data and specific export values. The DON predictions, in turn, were only satisfactory. The catchments with the highest peatland proportion had the highest TOC, DON and DOP export loads and modelled and measured

concentrations, which is a typical observation in boreal and temperate regions (Dillon and Molot 1997; Pellerin et al. 2004; Mattsson et al. 2005; Williams et al. 2005; Kortelainen et al. 2006). DOM exports are large because peatlands are in close hydrological contact with receiving watercourses, peat C storage is large, organic matter decomposition is incomplete due to the anaerobic conditions, and the peat contains few compounds that can retain C such as iron and aluminium (Domisch et al. 2000; Kalbitz et al. 2000; Freeman et al. 2001). In mineral soil sites, C that is leached from organic layer is effectively retained on iron- and aluminium-rich mineral soil layers even after disturbances such as clear-cutting and soil preparation (Pirainen et al. 2007) and eventually a large proportion of organic matter they contain are mineralized into CO₂. The amounts of TOC, DON and DOP in streams and lakes have been shown to be significantly related to soil type with higher TOC, DON and DOP export from peatland dominated catchments in comparison with forest dominated upland catchments (Willett et al. 2004; Kortelainen et al. 2006).

A lower export of TOC, DON and DOP from catchments where agriculture is the significant land-use form is consistent with other studies (Vuorenmaa et al. 2002; Stedmon et al. 2006; Mattsson et al. 2009). This can be because agricultural soils typically contain less organic material due to the annual harvesting of crops, and are relatively well-drained by ditches and subsurface-drains, which reduces the contact time between soil organic material and water decreasing the dissolution of organic matter (Mulholland 2003; Stedmon et al. 2006). On the regional scale this reflects the choice of land use: in agriculture mineral soils have been preferred over peatlands. Published data on exports of TOC from agricultural land is scarce, and more experimental data is needed to quantify the specific export values from agricultural areas.

The model overestimated the concentrations of TOC and DON in catchments (lakes 9, 10, 11 and 12) with a small peatland percentage (Fig. 4). The background leaching was modelled based on the data by Kortelainen et al. (2006), which showed nonlinear behaviour for low peatland percentages. Further analyses of TOC and DON data in Kortelainen et al. (2006) showed that the C:N ratio of DOM is lower in areas with low peatland percentage indicating high biodegradability (Fig. 2). This may lead to overestimation of TOC and DON concentrations, because the model does not take the biodegradation processes into account. In addition to low peatland percentage, the catchments of the lakes 9–12 also contained agricultural land (3.5–17 % of the area). The C:N ratio of leached organic matter is typically lower in agricultural than peatland dominated catchments (Mattsson et al. 2005), which may also increase the decomposition of organic

matter and result in lower TOC, DON and DOP concentrations. Furthermore, it has been found that TOC, DON and DOP concentrations and export decrease with increasing percentage of lakes in the catchment areas (Mattsson et al. 2005). The catchments of lakes 9–12 had high water percentage (17–32 % of the catchment area), and the overestimation of TOC, DON and DOP concentrations may be due to that part of the organic matter is retained in the upstream lakes (Mattsson et al. 2005). The clear-water lakes 9–12 were rather large and deeper than the other lakes (Table 1). In large and deep lakes, the catchment-derived organic matter is diluted in a large volume of water, typically resulting in lower TOC, DON and DOP concentrations compared with shallow lakes (Kortelainen 1993). Because of longer residence times, the degradation and sedimentation processes are likely to be more complete than in small and shallow lakes (Molot and Dillon 1996; von Wachenfeldt and Tranvik 2008). The model clearly underestimated DON concentrations in lake 3 (Mekrijärvi). The predicted values represent annual input of DON to the lake, whereas the observed higher concentrations may indicate the increase in DON concentrations over the longer period of time. In the Mekrijärvi catchment, there is long history of DON loading from ditching and peat mining. When ditching and peat mining first began (during the 1960–1970) water protection methods were not used or they were less efficient than nowadays. Furthermore, different peatland types have been found to produce different DON concentrations in runoff waters, and this fact can cause variations between predicted and observed concentrations in peatland dominated catchments (Joensuu et al. 2001; Kløve 2001; Joensuu et al. 2002).

The model over-predicted DOP concentrations in all the lakes by ca. $12 \mu\text{g L}^{-1}$ while well recovering the variability among the study. The over-prediction may due to the fact that background leaching, which dominated the export of DOP in all lakes (Fig. 3), is overestimated. The other reason for the overestimation can be that because P is typically the limiting nutrient in lakes it is efficiently taken up by phytoplankton, also in dissolved organic fraction (Li and Brett 2013). Furthermore, P efficiently reacts with iron, which is abundant in boreal lakes (Kortelainen et al. 2006), and subsequently is sedimented (Li and Brett 2013). These processes decrease P concentration in lake water and may cause a mismatch with the computed DOP which actually represents the export of DOP from the headwater catchments.

When comparing modelled and measured concentrations, it is important to be aware of the large variability in the measured concentrations (Fig. 4) that occurs between measuring points, sampling depth and sampling time. The modelled concentration describes the concentration of ‘bulk’ water entering the lake, which is assumed to be

completely mixed and unchanged in time. If the measured among-lake variability in concentrations (i.e. regression slopes in Fig. 4) are well predicted, this suggests that catchment characteristics and its land use dominate the water quality in the lakes over the internal lake processes. On the other hand, if the variability is not well captured, this can indicate either incorrect specific export values or the importance of internal lake processes in determining water quality.

The model succeeded in predicting the variability of TOC and DOP concentrations, and rank the lakes according to TOC, DON and DOP concentrations. This suggests that simple models such as the one developed here can be used for rough classification of lakes and further estimate the influence of land-use changes on water quality. All water quality classes were present among the study lakes (Fig. 4, Finnish Environment Institute SYKE 2005). The surface waters have been classified into five classes: excellent ($<8 \text{ mg TOC L}^{-1}$), good ($8\text{--}12 \text{ mg TOC L}^{-1}$) satisfactory ($12\text{--}17 \text{ mg TOC L}^{-1}$) and passable ($>17 \text{ mg TOC L}^{-1}$) for the EU’s Water Framework Directive purposes. Finnish lakes are typically coloured by high concentrations of TOC, and the percentage of lakes with TOC concentrations $>5 \text{ mg L}^{-1}$ in the whole country is 93 % (Kortelainen 1993).

Annual TOC, DON and DOP export to lakes were within the range reported in other boreal catchments (Mattsson et al. 2005; Kortelainen et al. 2006; Schelker et al. 2012). In Finland, the annual loads of terrestrial TOC, DON and DOP from catchments to watercourses have been reported to be $9\text{--}140 \text{ kg ha}^{-1}$, $0.2\text{--}3.2 \text{ kg ha}^{-1}$ and $0.01\text{--}0.14 \text{ kg ha}^{-1}$, respectively (Mattsson et al. 2005; Kortelainen et al. 2006). Also TOC ($3\text{--}21 \text{ mg L}^{-1}$), DON ($176\text{--}648 \mu\text{g L}^{-1}$) and DOP ($3\text{--}19 \mu\text{g L}^{-1}$) concentrations in the study lakes corresponded well with the average TOC (8 mg L^{-1}), DON ($397 \mu\text{g L}^{-1}$) and DOP ($11 \mu\text{g L}^{-1}$) concentrations, respectively, reported for Finnish lakes (Rantakari et al. 2004). The land-use patterns in our study catchments represented rather well the land use in Finland. The majority of catchments were covered by forests and peatlands (Table 2). In Finland, forests cover 78 % of the total area (Statistical Yearbook of Finland 2005). Two-thirds of the forests grow on upland soils and one-third on peat soils. Peatlands cover about one-third of the total land area, half of which has been drained for forestry. The proportions of agricultural, water and built-up areas in the study catchments were on average 5, 16 and 3 %, respectively. Agricultural land accounts for 8 % of the total area of Finland; surface waters cover about 10 %, and the rest 4 % of the total area are made up of built-up areas.

The fact that background leaching dominate TOC, DON and DOP export is consistent with the modelling study by Seitzinger et al. (2005), which indicated that $>80 \%$ of DON

and DOP export is from natural sources at the global scale. Our results imply that TOC, DON and DOP export is naturally low in boreal catchments where the proportion of peatlands is small. Consequently, the possibilities to reduce TOC, DON and DOP loads are restricted to areas where anthropogenic sources are significant. Forestry accounted only for a small proportion of TOC export which may due to the fact that while forest regeneration increases TOC export (Nieminen 2004; Schelker et al. 2012) maintenance ditching has the opposite effect (Joensuu et al. 2002; Nieminen et al. 2010). In the study catchments, the annual area of clear-cuttings during 1999–2013 varied from 0.11 to 0.46 % of the forest area which was a little less than the whole country average (0.81 %; Finnish Statistical Yearbook of Forestry 2014). Also the area of ditch network maintenance was smaller (varied from 0.23 to 1.52 %) than the country average 1.68 % (Finnish Statistical Yearbook of Forestry 2014). Even with the country average for areas of forestry operations, their contribution to TOC, DON and DOP loading in the study catchments would remain very small.

The specific export method is an easy tool to estimate the magnitude of terrestrial loading to watercourses and addressing the relative importance of different diffuse and point-sources. The advantage of the method is that it requires considerably less input information than the use of process-based models. Some uncertainties are, however, always related to the specific export coefficients and the area estimates of different land-use forms (McFarland and Hauck 2001; Johnes and Butterfield 2002; Nieminen et al. 2010). There are variabilities in the specific export values in the literature which reflects site specific variations in environmental conditions such as slope and soil texture (McFarland and Hauck 2001; Nieminen et al. 2010; Puustinen et al. 2010). Furthermore, the method does not take into account the retention processes such as the sedimentation of nutrients and suspended solids (Molot and Dillon 1996; von Wachenfeldt and Tranvik 2008) or mineralization and photodegradation of organic matter to inorganic forms of C, N and P in lakes (Tranvik and Bertilsson 2001; Lepistö et al. 2006; Schelker et al. 2014). The presented method uses open land-use and management data to upscale empirical export coefficients to a larger scale. The results suggest that the method can be used in rough classification of lakes in terms of their water quality. In the future, climate change, and to some extent also decreased acid deposition, may increase TOC export and concentrations in lake waters (Monteith et al. 2007; Sarkkola et al. 2009).

CONCLUSIONS

Open land-use data and specific export values were shown to predict the export of organic fractions of C, N and P to

boreal lakes reasonably well. The method provides a practical decision support tool for land-use management and assessment of water quality. The results indicated that the proportion of peatlands is the most important factor determining the exports of organic fractions of C, N and P in boreal catchments. In the catchments where the proportion of peatlands was low, agriculture was the largest anthropogenic source of TOC, DON and DOP. Consequently, the possibilities for controlling TOC, DON and DOP loadings are limited to catchments where peatland proportion is low and anthropogenic sources are significant.

Acknowledgments This study is a part of the TERLA project (Nos. 263429 and 292902) supported by the AKVA programme of the Academy of Finland. We thank agricultural secretaries of the municipalities, the staff of the regional Centre for Economic development, Transport and the Environment, and Ms. Anne Nylander from the Natural Resources Institute for help with data collection.

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