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Dynamics and temporal changes in suspended sediment transport in northern Finland: transport of very fine particulate matter

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Quantification of suspended sediment yield from rivers is essential to determining how land cover and land use affect water quality in fluvial ecosystems and erosion rates in catchment basins. In this study we used long-term (1967–2011) river-runoff and sediment-concentration data to seek new insights into suspended sediment transport and its governing factors. We found that long-term suspended sediment concentrations and yield of very fine sediment (< 0.4–$1.2\ \mu$m) are dependent on: (i) temporal changes in biogeochemical processes in catchment soils and fluvial systems, and (ii) temporal variations in metal humate colloid concentrations, especially in peat-covered catchments. The results also showed that the proportion of very fine sediment varies seasonally and can significantly affect transport rates in boreal rivers. Thus national monitoring activities should include parameters describing the quality of suspended sediment (e.g. particle size range and loss-on-ignition) in order to provide information relevant to land management and ecological assessments in boreal rivers.

Introduction

Suspended sediment is one of the key water-quality parameters in boreal freshwaters, affecting river-bed quality, transport of metals, nutrients and pollutants, light conditions, and the structure of aquatic food webs (Heikkinen 1994, Keskitalo and Eloranta 1999). While several factors affect the dynamics of suspended sediment, changes in land-use patterns are often the main cause of altered suspended sediment concentrations and yield (Syvitski *et al*. 2005, Valkama *et al*. 2007, Stenberg *et al*. 2015). Most suspended sediment discharge typically occurs during seasonal flood events, resulting in high temporal variations in sediment transport (Walling and Fang 2003). Suspended sediment transport is also controlled by soil type, topography,

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geology, vegetation, and land-use patterns and intensity (Walling and Fang 2003, Västilä *et al.* 2016). Moreover, suspended sediment transport is affected by various peatland uses such as forestry and peat extraction, which can increase erosion in countries with large peatland areas at high latitudes and in tropics (Marttila and Kløve 2008, 2010, Tuukkanen *et al.* 2014).

The majority of suspended sediment studies to date have been performed in major global rivers (Walling and Fang 2003, Syvitski *et al.* 2005, Hassan *et al.* 2008), mountainous regions (Wulf *et al.* 2012), areas with intensive agriculture (Oeurng *et al.* 2010) and temperate and tropical climates (Dang *et al.* 2010, Wu *et al.* 2012). In all those cases, transport rates were generally higher than in boreal regions. Only a few suspended-sediment studies have been conducted in boreal regions (Lobbes *et al.* 2000, Holmes *et al.* 2002, Bobrovitskaya *et al.* 2003). The importance of particulate organic matter (POM) for fluvial fluxes was highlighted in a recent study in the UK (Worrall *et al.* 2014), but similar assessments in boreal streams are lacking. There is thus an obvious need for information about the factors controlling suspended sediment yield at temporal and spatial scales in boreal rivers. Although suspended sediment concentrations are often low in boreal lowland river systems, their ecosystems are vulnerable and can be adversely affected even by small increases in suspended-sediment concentration (Kemp *et al.* 2011, Jones *et al.* 2011). Land-use activities in boreal peatland watersheds can result in high proportions of POM in transported suspended sediment and can thus strongly affect the carbon balance in boreal rivers (Heikkinen 1990, Worrall *et al.* 2014). Very fine particulate sediment (< 1 μm diameter) can be of inorganic (i.e. mainly clay) or organic origin. Both POM and particulate organic carbon (POC) have been receiving increasing attention due to their potential effect on carbon budgets (Cole *et al.* 2007) and their influence on aquatic ecosystems (Tank *et al.* 2010). In humic boreal river catchments with high peatland coverage, particulate matter is mainly transported as ferric iron (Fe^{3+}) organic complexes (Heikkinen 1990, Pokrovsky and Schott 2002). The dominance of iron is due to the higher attraction to organic particulate matter of ferric iron compared with that of other trivalent complexing metals (e.g. Al), and due to ferric iron typically being the most abundant metal in the waters of humic boreal rivers and peatland-derived runoff (*see* review by Vuori 1995).

Transport of suspended sediment is governed by physical factors such as stream flow, transport capacity and sediment availability. Transport of very fine fractions of POM is also affected by coupled biogeochemical factors, which control e.g. transport of iron and organic matter. However, understanding of the mechanisms of organic fraction transport and its interactions with microbiological, chemical, geological and hydrological factors is still incomplete. The significance of trivalent metal hydroxides for the flocculation and aggregation of particulate and dissolved humic substances is widely acknowledged (Heikkinen 1990, Jonsson 1997, von Wachenfeldt and Tranvik 2008). However, factors governing the temporal yield of suspended sediments and the role of long-term catchment processes in particulate transport in boreal river systems remain poorly understood. The process which is often overlooked in sediment transport analysis is the formation and transport of POC from terrestrial or aquatic autochthonous sources, which also affects suspended sediment yield. Terrestrial matter inputs also affect organic suspended matter quality and bioavailability to downstream biological communities (Wiegner *et al.* 2005, Wipfli *et al.* 2007, Ekholm and Lehtoranta 2012).

The aims of the present study were to: (i) review patterns of suspended sediment export in boreal rivers; (ii) assess the effect of methodological procedures (i.e. the use of different filters) on observed suspended sediment concentrations; (iii) study the dynamics and transport of very fine suspended sediment fractions (i.e. 0.4 and 1.2 μm filter residues) in boreal rivers, with an emphasis on possible interactions between colloids formed from Fe, Al and total organic carbon (TOC) and their impact on transported yield; and (iv) assess potentially significant temporal and spatial differences in transport of very fine suspended sediment, especially in streams with high TOC concentrations.

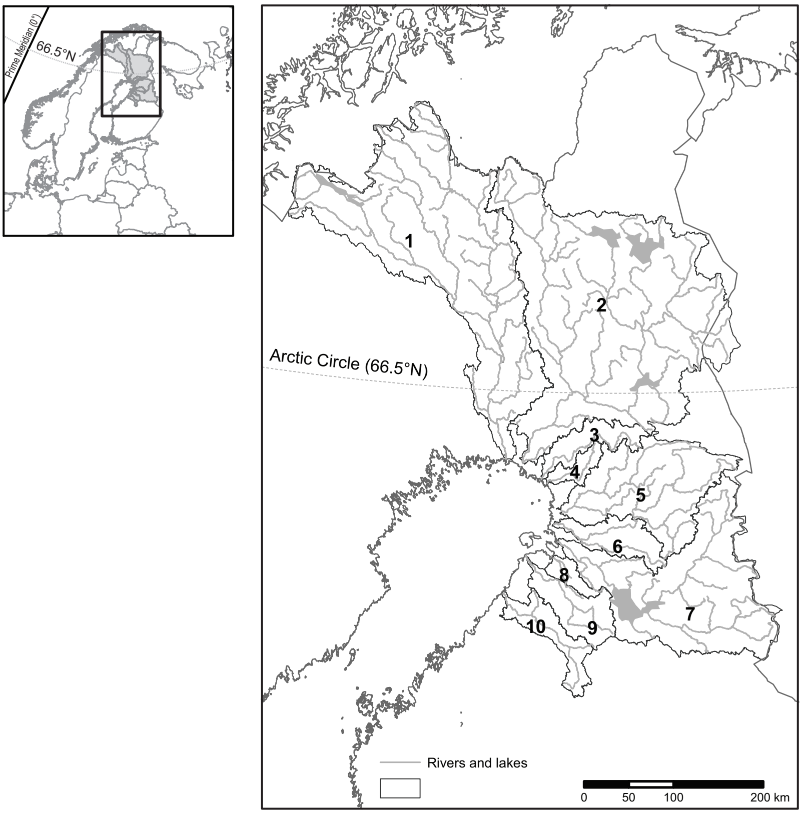

Fig. 1. Location of the 10 rivers and their catchments analysed in this study: 1 = Torniojoki, 2 = Kemijoki, 3 = Simojoki, 4 = Kuivajoki, 5 = Iijoki, 6 = Kiiminkijoki, 7 = Oulujoki,8 = Temmesjoki, 9 = Siikajoki, 10 = Pyhäjoki.

Material and methods

Sample collection and processing

Ten rivers that discharge into the Gulf of Bothnia along the Baltic northwestern coast of Finland were selected for the analysis (Fig. 1). The catchments of those rivers differ in size and land-use patterns (Table 1). Southern catchments are primarily flatland with agricultural dominance, whereas the northern ones typically contain small hills and higher forest cover. Glaciofluvial deposits (till, clay and peat formations) dominate in the soil of the catchments. Water quality in the rivers has been actively monitored since the 1960s, and all of them are included in the current national monitoring network in Finland. The monitoring data provide a good overview of suspended sediment transport, which has been measured using two filter fractions (0.4 μm and 1.2 μm, based on SFS3037) concurrently for over a decade in all studied rivers except the Temmesjoki (Table 1). The use of these two filter fractions enabled us to compare and evaluate the transport of very fine, suspended sediment (0.4–1.2 μm) in boreal river systems. Discharge data were taken from the OIVA database (Finnish Environmental administration).

Water samples were taken with a grab, according to the national sampling standards (Kettunen *et al*. 2008) as part of national monitoring of water quality of riverine inputs into the Baltic Sea. They were taken from the river outlets (Fig. 1) four times a year during 1962–1981. Between 1982 and 2011 the samples were taken 10–18 times, monthly outside the flood period and, for better representativeness, 1–2 per month during the spring flood. Water quality data were acquired from the OIVA database (Finnish Environmental administration).

Suspended solids were initially monitored using a 1.2 μm filter (Whatman GF/C, hereaf-

Table 1. Information on the 10 studied rivers and their catchments. *A* is the catchment area, *L* is the percentage of lakes in a catchment, MQ is the mean discharge, HQ is the high flow, 1.2 μm and 0.4 μm are suspended sediment concentrations measured with 1.2-μm and 0.4-μm filters, respectively; land use in 2006 was derived from CORINE (http://www.eea.europa.eu/publications/COR0-landcover).

River	*A* (km^2)	*L* (%)	MQ (m^3 s^{-1})	HQ (m^3 s^{-1})	Agriculture (%)	Forest (%)	Wetlands (%)	First obs. (1.2 μm)	First obs. (0.4 μm)	Min/mean/max conc. of 1.2 μm (mg l^{-1})	Min/mean/max conc. of 0.4 μm (mg l^{-1})	Annual sediment yield from 1.2 μm (t y^{-1}/t km^{-2} y^{-1})	Annual sediment yield from 0.4 μm (t y^{-1}/t km^{-2} y^{-1})
Tornionjoki	40131	4.63	423	3179	0.8	76.0	16.0	10.V.1967	11.V.1988	0.1/5/110	0.1/4/43	61418/1.53	61277/1.53
Kemijoki	51127	4.30	571	4557	0.75	77.6	16.3	10.V.1967	11.V.1988	0.2/3/39	0.1/3.6/51	63624/1.24	73773/1.44
Simojoki	3160	5.66	44.7	595	1.7	69.6	21.8	9.V.1967	1.VI.1988	0.1/4/129	0.1/4/50	6936/2.19	6990/2.21
Kuivajoki	1356	2.72	17.2	264	2.1	67.2	26.7	3.XI.1969	1.III.1994	0.4/5/24	1.4/10/62	3253/2.39	6311/4.65
Iijoki	14191	5.67	171	806	7.5	71.0	8.2	1.III.1971	1.III.1994	0.1/4/39	0.7/6/45	19733/1.39	27585/1.94
Kiiminkijoki	3814	2.97	42.8	609	2.4	68.4	24.1	4.III.1970	18.V.1988	0.3/5/60	0.8/9/40	7413/1.94	13940/3.65
Oulujoki	22841	11.47	263	848	2.6	72.7	10.8	1.III.1967	28.I.1988	0.2/4/110	0.5/6/72	26890/1.17	41287/1.81
Temmesjoki	1181	0.50	2.9	113	17.8	60.0	19.0	13.VIII.1969	11.V.2009	0.8/15/152	10/20/44	6353/5.38	6162/5.22
Siikajoki	4318	2.18	39.9	465	16.5	66.4	8.8	6.III.1967	7.I V.1994	0.3/13/350	2.4/19/100	21774/5.04	26683/6.18
Pyhäjoki	3724	5.16	29	267	16.1	66.8	8.7	6.III.1967	9.III.1988	0.5/9/61	0.7/15/66	10974/2.94	17417/4.67

ter $SS_{1.2}$) and from 1990 onwards also using a 0.4 μm filter (Nuclepore polycarbonate (PC) filters, hereafter $SS_{0.4}$) (*see* Table 1). Water samples were taken at the same time, placed in different sampling bottles and filtered separately. For the present analysis, we calculated the difference between the concentration on the 1.2 μm filter and that on the 0.4 μm filter (hereafter SS_{diff}). SS_{diff} represents the concentration of the particles between 0.4 and 1.2 μm in diameter. We hypothesized that the variation in SS_{diff} reflects the changes in formation of metal hydroxide organic colloids. Higher SS_{diff} values represent conditions in which physico-chemical flocculation by Fe, Al and DOC is more likely to take place. According to Pokrovsky and Schott (2002), the size of iron colloids in boreal fluvial systems of Russian Karelia varies between 0.8 μm and 100 kilodalton (kD). Therefore, we expected that metal colloids would mainly pass through $SS_{1.2}$, but would be partly trapped by $SS_{0.4}$. However, we acknowledge that estimation of SS_{diff} is non-proportional through time for variable particle sizes and variations in inorganic/organic proportions in transported suspended solids. Therefore, the objective of this study was to report relative differences between the 1.2 and 0.4 μm filters rather than provide estimates of absolute concentrations of very fine particulate matter.

The long-term national monitoring programme in Finland include analysis of total organic carbon (TOC: SFS-EN 1484:1997 (IC017), unfiltered), chemical oxygen demand (COD_{Mn}: SFS 3036:1981 (IC002), unfiltered), oxygen (O_2), water temperature (temp), pH, turbidity (FTU), water colour (colour), and electrical conductivity (EC). In addition, iron (Fe, not filtered) and aluminium (Al, not filtered) are analysed by inductively-coupled plasma optical emission spectroscopy (ICP-OES). Water samples are taken at the same time as suspended solids samples and all analyses are performed in the laboratory within 24 hours of sampling. The long-term national monitoring programme does not provide data on particle size or loss-on-ignition to reveal the POM fraction, or data from traditional ultra-filtration or dialysis techniques to isolate different particulate fractions. While this was a shortcoming for the present study, the available data set was still deemed suitable to examine long-term and temporal changes in transport of very fine suspended sediment.

We studied contributions of TOC, Al, Fe and sampling month to suspended sediment concentrations by using mixed models fitted for the variables $SS_{0.4}$ and $SS_{1.2}$. The mixed models (Eq. 1) accounted for individual river effects and let the variance of random errors vary with the river type. River types were defined as: large rivers (Oulujoki, Iijoki, Kemijoki and Tornionjoki), peatland-dominated rivers (Kiiminkijoki, Kuivajoki and Simojoki) and agriculture-dominated rivers (Pyhäjoki and Siikajoki).

$$\begin{aligned} SS_{0.4ij} &= \beta_0 + \beta_1 Fe_i + \beta_2 Al_i + \beta_3 TOC_i \\ &+ \beta_4 SS_{1.2i} + \beta_1 Month2_i + \ldots \\ &+ \beta_{15} Month12_i + u_j + \varepsilon_{ij} \end{aligned} \quad (1)$$

where $i = 1, \ldots, 1978$, and $j = 1, \ldots, 9$. In addition, $u \sim N(0,\sigma_u^2)$ and $\varepsilon \sim N(0,\sigma^2 \times \delta^2)$, where δ is related to the type of land use. The mixed model used for the variable $SS_{1.2}$ had the same form as Eq. 1, where $SS_{0.4}$ was used as a response variable instead of $SS_{1.2}$. To meet the assumptions of mixed models, normality of the variables $SS_{0.4}$, $SS_{1.2}$, Fe, Al and TOC was tested and those which did not meet the assumptions were log-transformed prior to fitting them into the model. A mixed model was run separately for SS_{diff} but the results were not statistically significant and are therefore not reported here. We included discharge (Q) variable into the original mixed models but as it did not explain sediment concentration it was omitted from the final model. We also tested the individual explanatory power of different variables for $SS_{0.4}$ and $SS_{1.2}$ concentrations by calculating the coefficients of determination (marginal R^2 and conditional R^2, Nakagawa and Schielzeth 2013) for mixed models with just a single fixed explanatory variable at a time. Marginal R^2 values represent the proportion of variance explained by the fixed part of a mixed model, while conditional R^2 values include the proportion of variance explained by the random factors and the fixed part, i.e. account for individual river effects. For the $SS_{0.4}$ data, we also tested the model by removing two outliers $SS_{0.4}$ = 0 mg l^{-1} from the data. All mixed model analyses were conducted using the *nlme* package (Pinheiro *et al*. 2014), and the *MuMIn* package (Nakagawa and Schielzeth 2013, Johnson 2014) of R (R Core Team 2014). Furthermore, by interpolating the measured concentrations linearly to estimate daily concentrations and multiplying those by daily flow, we were able to obtain annual and monthly sediment transport rates (RLOAD software, University of Linköping, Sweden). The authors note that concentration changes can be affected by major rain events that occur between sampling. We also tested the use of the conventional rating curve against discharge at sampling time (Walling 1997, Syvitski *et al*. 2000, Marttila and Kløve, 2010).

Results

Spatial, temporal and seasonal variations in suspended sediment transport

During the 44-year observation period (1967–2011), $SS_{1.2}$ and $SS_{0.4}$ concentrations in the studied rivers varied widely among years (Table 1 and Fig. 2). The highest concentrations in all rivers were recorded during spring and autumn flood periods. The interannual hydrological variability, i.e. flow magnitude and total runoff in the river basins, was large during the studied period and had obvious effects on suspended sediment transport and yield. Discharge controlled the overall general suspended sediment concentration and yield in all rivers studied, but its effect on $SS_{0.4}$ or SS_{diff} was not as pronounced (Figs. 3 and 4). Seasonally, suspended sediment did not always follow the patterns of runoff and displayed periodic fluctuations. A marked peak in the $SS_{0.4}$ fraction was observed during the wet period in 1998–1999, which was preceded by a particularly dry period between 1994 and 1997. For example in the Kiiminkijoki, mean SS_{diff} during 1994–1997 was 4.3 mg l^{-1}; whereas in 1998, SS_{diff} was 15.6 mg l^{-1}. Annual sediment yield in the 10 rivers ranged from 1.17 to 5.38 t km^{-2} y^{-1} for the 1.2 μm fraction and from 1.44 to 6.18 t km^{-2} y^{-1} for the 0.4 μm fraction and was highest during spring and autumn flood periods (Table 1). In almost all studied rivers, high sediment transport yield occurred in 1998–1999, with large differences between the yield of the 1.2 μm and 0.4 μm fractions.

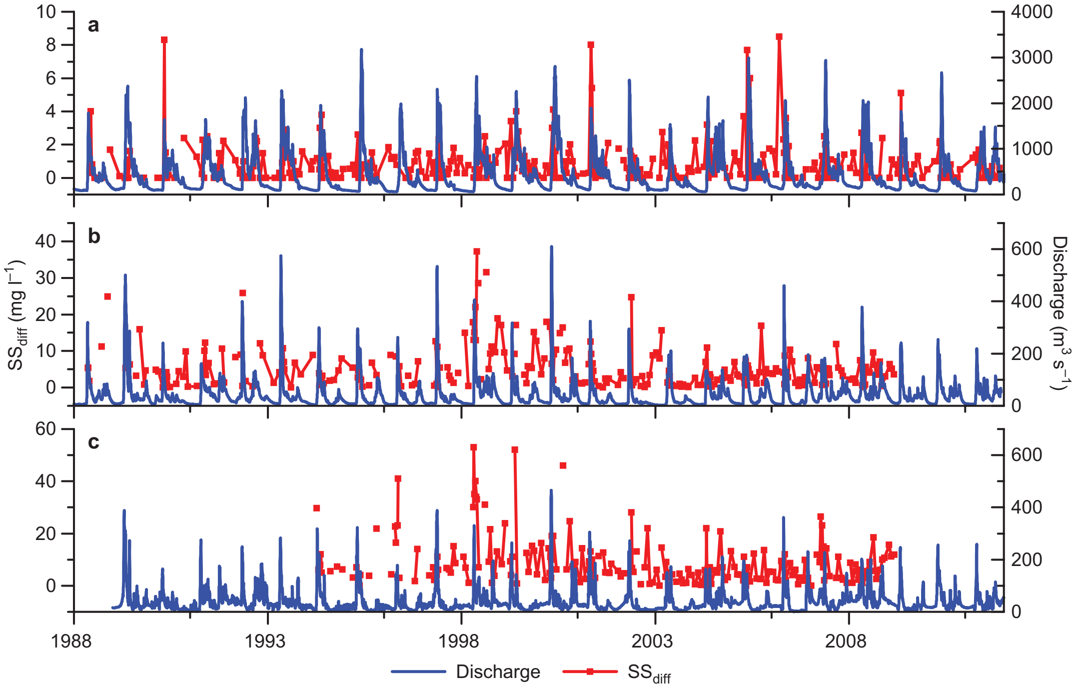

Fig. 2. Discharge and SS_{diff} (i.e. difference between $SS_{1.2}$ and $SS_{0.4}$) in (**a**) Tornionjoki, (**b**) Kiiminkijoki and (**c**) Siikajoki.

When the whole data set was used, the correlation between discharge and suspended sediment concentration was not statistically significant ($p > 0.05$), because the data included highly regulated rivers such as the Oulujoki. However, for individual sites there was a clear correlation between discharge and suspended sediment concentration. Power functions with $SS_{1.2}$ performed rather well for most rivers, but those with $SS_{0.4}$ and SS_{diff} showed high levels of variation (Figs. 3 and 4). In rivers with high concentrations of humic substances, such as the Kiiminkijoki and Kuivajoki, the correlation between discharge and suspended sediment was weak (Fig. 5), indicating the importance of other processes in suspended sediment transport.

Variations in the difference between $SS_{1.2}$ and $SS_{0.4}$

Comparisons of the difference (SS_{diff}) between $SS_{1.2}$ and $SS_{0.4}$ concentrations provided insights into differences in the timing and seasonality of very fine sediment transport in boreal rivers. Concentration differences were highest during moderate high flow or low flow periods. The patterns of change in the two suspended solid fractions exhibited clear differences in almost all rivers (Figs. 2 and 3). The greatest concentration differences occurred before spring snowmelt (e.g. in the Kiiminkijoki), during summer low flow conditions (all rivers) and during summer/autumn high flow periods (especially the Simojoki, Kuivajoki and Kiiminkijoki). In contrast to peatland-dominated rivers, in agriculture-dominated rivers (the Siikajoki and Temmesjoki) SS_{diff} was greatest during the spring flood. The greatest long-term concentration differences and the highest concentrations of Fe, Al and TOC occurred during the relatively wet period 1998–1999, which was preceded by a long dry period. This indicates a clear link between alteration of wet and dry seasons, more specifically formation of humate colloids during dry and their increased transport in wet seasons. During the wet period 1998–1999, the concentrations of $SS_{0.4}$ and SS_{diff} were especially high in the Simojoki, Iijoki, Kuivajoki and Kiiminkijoki.

Based on the random effects of individual rivers in the mixed model, the Siikajoki and Pyhäjoki had the highest impact on $SS_{0.4}$ con-

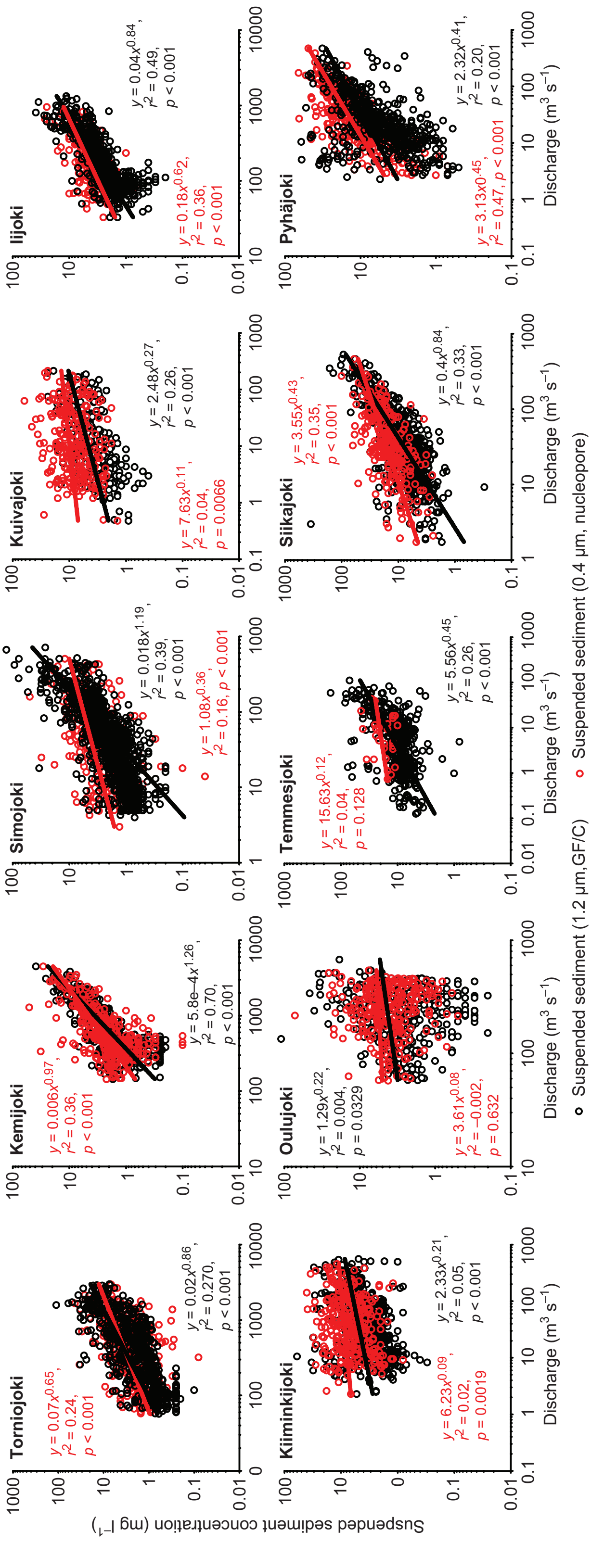

Fig. 3. Regression curves for suspended sediment (0.4 *μm* and 1.2 *μm* fractions) against discharge in the study rivers.

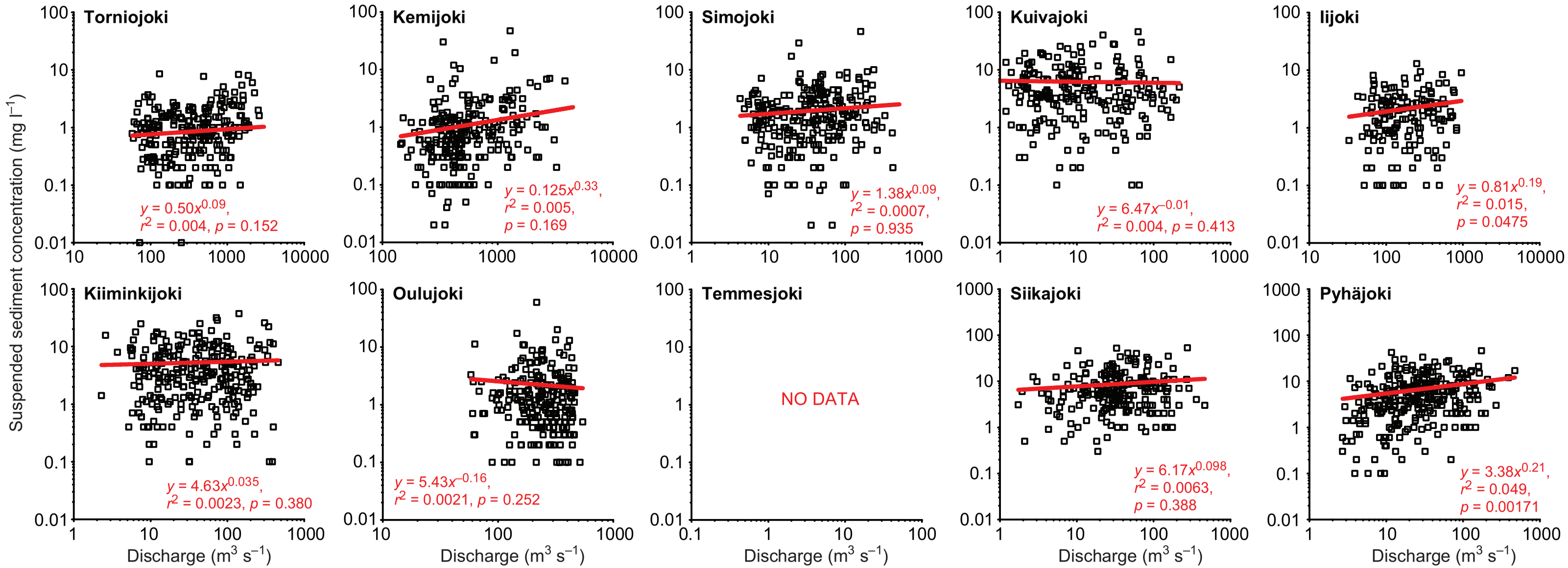

Fig. 4. Regression curves for SS_{diff} fractions against discharge in study rivers.

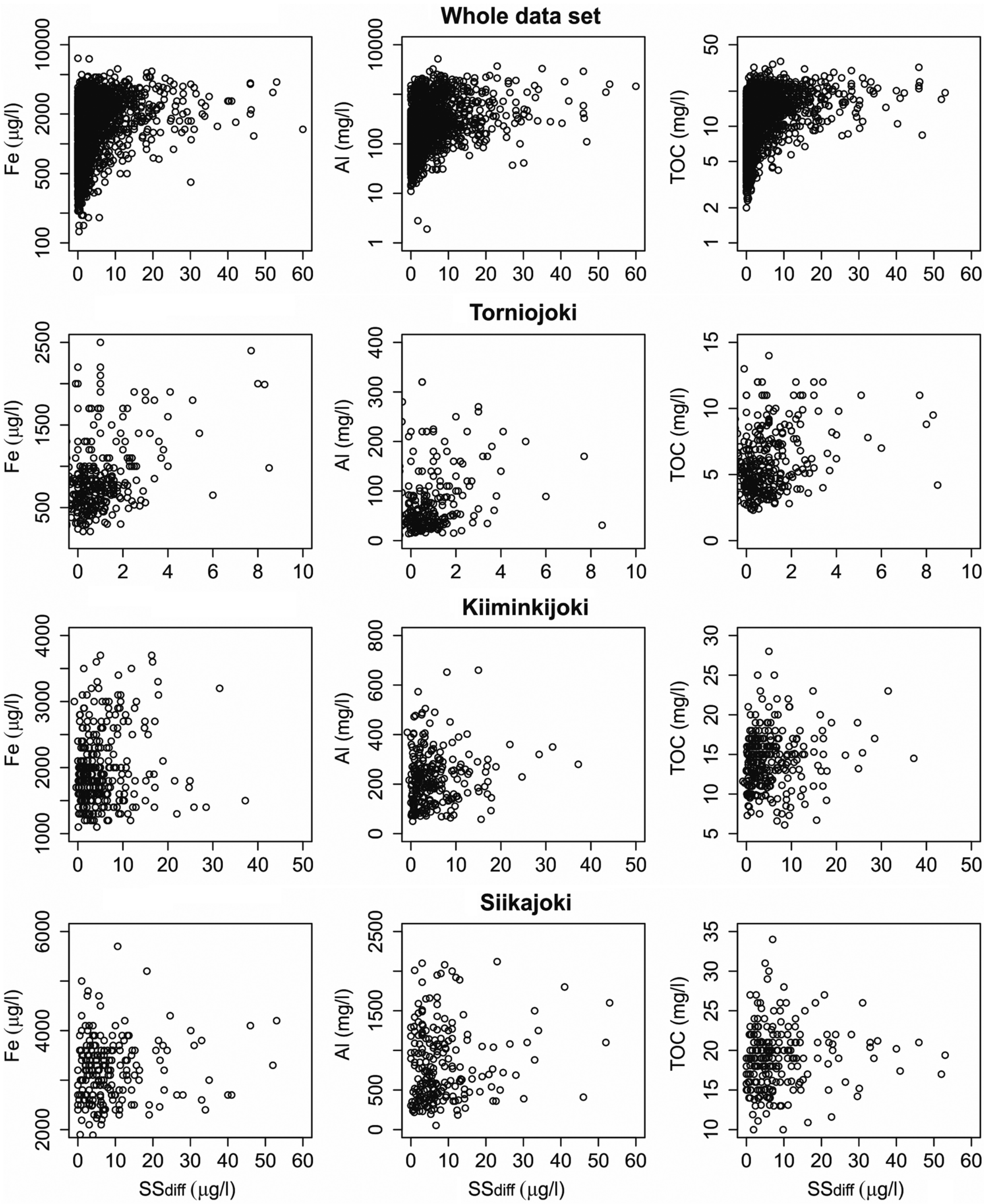

Fig. 5. Relationship between SS_{diff} and Fe, Al and TOC for the entire data set and for the Tornionjoki (large rivers), Kiiminkijoki (agriculture-dominated catchment) and Siikajoki (peatland-dominated catchment), which are chosen to represent different categories used in the study.

centrations. Among the 10 studied rivers, these two rivers were also characterized by the highest agricultural impact and the lowest wetland coverage (%) in their catchments (Table 1). The Temmesjoki, which was characterized by the highest agricultural impact and the lowest forest

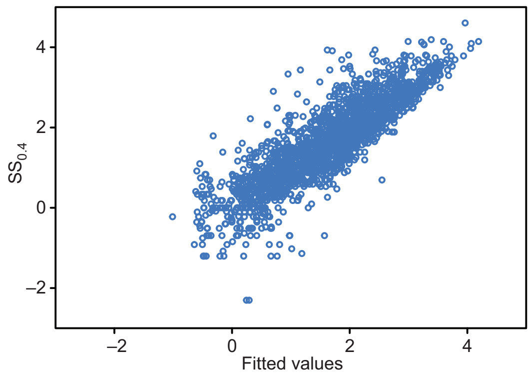

Fig. 6. Log-transformed real values of the response variable (mixed model for $SS_{0.4}$, all data included) plotted against the fitted values from the mixed model.

coverage (%) in its catchment, had to be omitted from the mixed model due to missing values.

In addition to the constant (Month1), statistically significant variables in the mixed model were Fe ($p < 0.001$) and $SS_{1.2}$ ($p < 0.001$) (Table 2). Seasonal variation in the mixed model was not statistically significant, as the other sampling months (Months2–12, $p > 0.2$) did not differ from Month1 (β_0). When the mixed model was tested without $SS_{1.2}$, Fe and Al and also TOC were statistically significant. When $SS_{1.2}$ was included in the model, only Fe ($p = 0.00$) was left as a statistically significant variable, but not Al ($p = 0.857$) or TOC ($p = 0.063$).

Based on the coefficients of determination (conditional R^2), the mixed models fitted the data relatively well (Table 2). Although the aim of the mixed models was to estimate the quality and not the quantity of different fine-sediment fractions ($SS_{0.4}$ and $SS_{1.2}$), even the mixed model for $SS_{0.4}$ (all data) with the lowest conditional R^2 value (0.40) fitted the data relatively well (Fig. 6). Based on the marginal and conditional coefficients of determination (R^2) calculated from mixed models fitted for $SS_{1.2}$ values with a single fixed explanatory variable, Al explained the variation in $SS_{1.2}$ concentration the best. The situation was less clear for $SS_{0.4}$, as none of the explanatory variables alone explained the results well. When two outliers (both $SS_{0.4} = 0$ mg l^{-1}) were removed from the data set, the marginal and conditional R^2 increased for most variables, but no single variable explained $SS_{0.4}$ particularly well by itself (Table 3).

Discussion

Knowledge on the spatial and temporal patterns of transport of different fractions of suspended sediment is essential for understanding the dynamics of suspended sediment transport by boreal rivers draining into the Baltic Sea. Our results suggest that suspended sediment in boreal rivers contains a large quantities (up to 50%) of very fine particulate matter (0.4–1.2 μm). The dynamics of this very fine particulate matter does not depend on flow conditions only, but rather on temporal changes and alternations in hydrological patterns and chemical accumulation of metal hydroxides with particulate and dissolved organic compounds (e.g. humic substances) in surface waters.

While the transport processes of fine suspended sediment followed river discharge patterns, the transport of very fine particulate matter was determined by other factors. Al was responsible for much of the variation in the mixed model for $SS_{1.2}$ concentrations, whereas in the case of the mixed models for $SS_{0.4}$ concentrations, none of the variables explained the variation particularly well. One possible catchment-scale process that could explain the observed differences is physico-chemical flocculation by Fe, Al and TOC, which results in the formation of metal hydroxide organic colloids. Fe and Al have been shown to be effective flocculants of organic matter (Lefebvre and Legube 1993, Libecki and Dziejowski 2008) and can control the transport of organic material (Pokrovsky and Schott 2002). However, findings on the relationship between discharge and Fe concentration are contradictory. Grieve (1984) and Knorr (2012) reported positive correlations between discharge and Fe, whereas Heikkinen (1990) reported negative correlations for a humus-rich river. A review by Vuori (1995) concluded that mobilisation and transport of Fe in rivers is largely controlled by dissolved and particulate organic complexing agents and that Fe dynamics are governed by the same factors that control the quality and quantity of organic material transport. This supports the findings by Heikkinen (1990), who found the highest proportion of the POC load in the Kiiminkijoki during spring floods and a clear increase in POC with discharge also in summer,

Table 2. Comparison of results for the $SS_{0.4}$ and $SS_{1.2}$ mixed models (*see* Eq. 1).

Fixed effect parameter	$SS_{1.2}$ all data			$SS_{0.4}$ all data			$SS_{0.4}$ outliers removed		
	Value (SD)	p	95%CI	Value (SD)	p	95% CI	Value (SD)	p	95%CI
β_0	–1.31 (0.41)	0.001	(–2.11,–5.11)	–1.45 (0.27)	0.000	(–2.00, –0.92)	–5.51 (0.24)	0.000	(–5.98, –5.04)
Fe	0.27 (0.06)	0.000	(0.15, 0.39)	0.30 (0.04)	0.000	(0.22, 0.38)	0.42 (0.04)	0.000	(0.35, 0.49)
Al	–0.01 (0.04)	0.887	(–0.08, 0.07)	0.05 (0.03)	0.054	(0.00, 0.11)	0.53 (0.03)	0.000	(0.49, 0.58)
TOC	0.15 (0.08)	0.070	(–0.01, 0.30)	0.03 (0.05)	0.588	(–0.07, 0.12)	0.16 (0.05)	0.001	(0.07, 0.25)
SS1.2	0.64 (0.04)	0.000	(0.57, 0.72)	0.58 (0.02)	0.000	(0.54, 0.63)	–	–	
Month2	–0.01 (0.11)	0.952	(–0.23, 0.21)	0.03 (0.08)	0.707	(–0.12, 0.18)	–0.24 (0.07)	0.001	(–0.38, –0.11)
Month3	0.05 (0.10)	0.635	(–0.16, 0.25)	0.02 (0.07)	0.780	(–0.12, 0.16)	–0.02 (0.07)	0.756	(–0.15, 0.11)
Month4	0.00 (0.10)	0.982	(–0.20, 0.19)	0.00 (0.07)	0.960	(–0.14, 0.13)	0.59 (0.06)	0.000	(0.47, 0.71)
Month5	0.05 (0.10)	0.630	(–0.14, 0.24)	0.00 (0.07)	0.959	(–0.12, 0.15)	0.85 (0.06)	0.000	(0.73, 0.96)
Month6	–0.09 (0.11)	0.429	(–0.30, 0.13)	–0.08 (0.07)	0.279	(–0.21, 0.06)	0.62 (0.06)	0.000	(0.50, 0.74)
Month7	–0.14 (0.12)	0.227	(–0.35, 0.11)	–0.09 (0.08)	0.260	(–0.24, 0.07)	0.64 (0.07)	0.000	(0.50, 0.78)
Month8	–0.12 (0.12)	0.295	(–0.31, 0.11)	–0.13 (0.07)	0.069	(–0.28, 0.01)	0.51 (0.07)	0.000	(0.38, 0.64)
Month9	–0.11 (0.10)	0.290	(–0.31, 0.09)	–0.09 (0.07)	0.200	(–0.23, 0.05)	0.46 (0.06)	0.000	(0.34, 0.59)
Month10	0.05 (0.10)	0.610	(–0.15, 0.26)	–0.04 (0.07)	0.553	(–0.18, 0.10)	0.56 (0.06)	0.000	(0.44, 0.69)
Month11	–0.05 (0.11)	0.670	(–0.26, 0.17)	–0.07 (0.07)	0.308	(–0.22, 0.07)	0.21 (0.07)	0.002	(0.08, 0.34)
Month12	0.06 (0.11)	0.568	(–0.16, 0.29)	–0.01 (0.08)	0.907	(–0.14, 0.16)	0.03 (0.07)	0.698	(–0.11, 0.17)
Random effects	σ^2			σ^2			σ^2		
u	0.04			0.04			0.09		
Random errors	$\sigma^2 \times \delta^2$			$\sigma^2 \times \delta^2$			$\sigma^2 \times \delta^2$		
Large rivers	1.08			0.26			0.27		
Peatland-dominated rivers	1.27			0.31			0.19		
Agriculture-dominated rivers	0.21			0.21			0.19		
Marginal R^2	0.38			0.68			0.73		
Conditional R^2	0.40			0.72			0.80		

during which drifting algae formed the majority of POC. This implies that physico-chemical factors primarily dominate colloidal dynamics and availability of fine suspended sediment whereas discharge is acting as transporting vector in boreal humus-rich rivers.

Even though our data set did not contain measurements of organic fractions or particle size (not included in the national monitoring programme), our results suggest that the remarkably large difference between $SS_{1.2}$ and $SS_{0.4}$ concentrations is mainly caused by transport of colloids. We base this assumption on the results from the mixed model analysis. Fe was the only other statistically significant variable when the variable $SS_{1.2}$ was included in the mixed model, (i.e. besides $SS_{1.2}$), that explained the concentration of $SS_{0.4}$. This indicates that Fe plays a role in the $SS_{0.4}$ transport. When $SS_{1.2}$ and Al were omitted from the mixed model, TOC and part of the term for seasonal variation (sampling months) became statistically significant. This suggests that $SS_{1.2}$ (and Al) include most of the information in TOC and seasonal variation.

Based on the marginal and conditional R^2 values (Table 3), Al seems to be responsible for most of the variation in $SS_{1.2}$ and $SS_{0.4}$ concentrations, although the effect was less pronounced for $SS_{0.4}$ concentrations. The $SS_{0.4}$ concentration may be more difficult to explain since measurements do not include information on the mineral component in suspended solids, which could explain some of the variation in $SS_{0.4}$ concentration. This is also indicated by the low R^2 values in both $SS_{0.4}$ mixed models for TOC. The marginal R^2 values indicated that Fe and Al could explain 43% and 52% of the variation, respectively. However, our results may also have been affected by the location of the sampling stations, which were situated in the lower reaches of large rivers and thus more affected by mineral soils with naturally higher Al and inorganic matter concentrations. This would explain why we observed Al playing a greater role than Fe in transporting $SS_{1.2}$ in the lower reaches of large rivers. In addition, a large proportion of the $SS_{0.4}$ fraction in the lower reaches possibly comprised inorganic matter. Also channel processes (erosion and deposition) can affect temporal variation of SS concentrations. These results are applicable to other similar boreal regions with high peat land-cover.

Therefore, our results confirm findings of previous studies, which highlight the importance of organic material and metals for suspended sediment transport in humus-rich waters (Sedell *et al*. 1978, Ingri *et al*. 2000, Pokrovsky and Schott 2002, Perez *et al*. 2011, Ilina *et al*. 2013). Due to the lack of direct measurements of particulate fractions in the official national monitoring data, we were unable to distinguish between, or rank, the relative importance of Al, Fe and organic colloids. However, our results show that even very minute particle-size differences (i.e. range 0.4–1.2 μm) can significantly affect measured suspended sediment concentrations in boreal rivers. While in theory, this result could partly be explained by the physical differences of the different filters, we know of no detailed laboratory that have assessed the degree filter physical properties on results. Previous studies have clearly demonstrated that the processes governing transport of metal hydroxides and organic matter are closely related (*see* also Tipping *et al*. 2002) and that iron colloids are typically less than 0.8 μm in size (Pokrovsky and

Table 3. Marginal and conditional coefficients of determination (R^2) calculated for mixed models with single fixed explanatory variable, i.e. the explanatory power of individual variables.

Mixed-model variable	$SS_{1.2}$ all data		$SS_{0.4}$ all data		$SS_{0.4}$ outliers removed	
	Marginal R^2	Conditional R^2	Marginal R^2	Conditional R^2	Marginal R^2	Conditional R^2
Al	0.68	0.73	0.29	0.33	0.52	0.56
Fe	0.42	0.57	0.26	0.36	0.43	0.58
TOC	0.43	0.51	0.21	0.28	0.28	0.41
$SS_{1.2}$	–	–	0.34	0.41	0.53	0.66
Month	0.29	0.59	0.08	0.36	0.13	0.56

Schott 2002). Therefore, we were able to obtain good estimates of the proportion of these very fine suspended sediment fractions (< 1.2 μm) in boreal river systems. However, we were unable to deduce from our data whether this difference is related to the increase in inorganic Fe and Al hydroxides or in POM. For peat-covered catchments in particular, our results indicate that the difference between $SS_{1.2}$ and $SS_{0.4}$ concentrations is predominantly attributable to natural organometallic complexes (Fe, Al) and TOC. The mixed model results did not show any clear and strong effects of different land use types, but based on the random effects of individual rivers in the mixed model agricultural area (%) has the greatest impact on $SS_{0.4}$ concentration. To fully determine the effect of land use, more detailed measurements of inorganic and organic proportions and direct particle-size measurements are needed. Previous studies have shown that the organic matter constitutes 2.1%–36% of river particulate matter in boreal regions (Marttila and Kløve 2015), highlighting the importance of POM for suspended particulate matter transport. The results of the present study indicate the potential importance of measuring POM content in suspended solids, which would provide better knowledge and understanding on POM transport in national monitoring programmes.

A clear increase in the transport of SS_{diff} concentrations was observed after dry–wet periods, which are often linked to increased leaching of DOC from catchments (Lepistö *et al*. 2008, Laudon *et al*. 2011). In general, dry periods lower the local groundwater level and expose soil layers to oxidation processes. Ensuing wet periods with elevated groundwater levels and associated increased surface runoff from TOC-rich topsoils then flush previously mobilised substances into stream networks (Laudon *et al*. 2011). In addition, wet periods can change soil profile redox conditions to reductive, thus increasing the flushing of dissolved ferrous iron (Fe^{2+}) (Sarkkola *et al*. 2013), which can later form colloids in redox boundaries by precipitation of Fe^{3+} (Pokrovsky *et al*. 2002). Thus the increased SS_{diff} concentration after dry seasons is most likely a result of organometallic colloid formation following increases in Al, TOC and Fe transport, indicating supply limitation of very fine sediment. The effect of particulate metal and humic substances on fine sediment transport was particularly pronounced in peatland rivers (e.g. the Kiiminkijoki, Kuivajoki, Simojoki) compared with larger rivers (e.g. the Tornionjoki and Kemijoki).

Measurements of suspended sediment concentrations as such are of little ecological value since they do not predict bioavailability. Further, data on suspended sediment concentrations is often sparse and display a high degree of temporal and spatial variability, and a poor overall relationship between sampling and discharge. If aquatic monitoring is intended to assess the potential ecological impacts of terrestrial land-use patterns, more attention should be paid to the actual quality of the allochthonous matter and the processes affecting it. There is clearly a need for in-depth studies specifically addressing ecological responses to changes in particulate matter quality and quantity, and the relationship to overall particle yield.

Conclusions

This study showed that very fine particulate matter POM plays a role in determining annual suspended sediment yield in peat-dominated catchments and that more focus should be on the quality of organic matter as it enables better evaluation of the possible sources of particulate matter. A comparison between $SS_{0.4}$ and $SS_{1.2}$ enables making rough, literature-based estimates of the suspended sediment constituents, but also suggests that national water-monitoring programmes could benefit from analysing the quality of suspended solids (e.g. loss-on-ignition and Fe). This study also showed that temporal variations in metal humate colloid concentrations can strongly affect suspended sediment yield, especially in peat-covered catchments. Thus, the very fine suspended sediment fractions need to be taken into account in load calculations and ecological assessments of boreal rivers. The overall importance of organic matter is known, but previous analyses have focused on the dissolved fraction of organic matter and not on the particulate fraction. Future studies should distinguish between the different sources of very fine suspended sediment and acknowledge the role of metal-hydroxide–humus colloids in their formation. This will enable assessment of their effect on particulate transport and, ultimately, their impacts on marine environments.

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