Riverine particulate C and N generated at the permafrost thaw front: case study of western Siberian rivers across a 1700-km latitudinal transect

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Key words: nutrient, particulate, suspended, landscape, bog, lake, forest, thaw, Siberia

Submitted to *Biogeosciences, after 2nd revision, October 2018*

Abstract

In contrast to numerous studies on the dynamics of dissolved (< 0.45 µm) elements in permafrost-affected high latitude rivers, very little is known of the behavior of river suspended (> 0.45 µm) matter (RSM) in these regions. In order to test the effect of climate, permafrost and physio-geographical landscape parameters (bogs, forest and lake coverage of the watershed) on RSM and particulate C, N and P concentration in river water, we sampled 33 small and medium size rivers (10 – 100,000 km² watershed) along a 1700 km N - S transect including both permafrost-affected and permafrost-free zones of the Western Siberian Lowland (WSL). The concentration of C and N in RSM decreased with the increase in river watershed size, illustrating *i*) the importance of organic debris in small rivers which drain peatlands and *ii*) the role of mineral matter from bank abrasion in larger rivers. The presence of lakes in the watershed increased C and N but decreased P concentrations in the RSM. The C:N ratio in the RSM reflected the source from deep rather than surface soil horizon, similar to that of other Arctic rivers. This suggests the export of peat and mineral particles through suprapermafrost flow occurring at the base of the active layer. There was a maximum of both particulate C and N concentration and export fluxes at the beginning of permafrost appearance, in the sporadic and discontinuous zone (62-64°N). This presumably reflected the organic matter mobilization from newly thawed organic horizons in soils at the active latitudinal thawing front. The results suggest that a northward shift of permafrost boundaries and an increase in active layer thickness may increase particulate C and N export by WSL rivers to the Arctic Ocean by a factor of 2, while P export may remain unchanged. In contrast, within a long-term climate warming scenario, the disappearance of permafrost in the north, the drainage of lakes and transformation of bogs to forest may decrease C and N concentration in RSM by 2 to 3 times.

1. Introduction

High-latitude rivers are most vulnerable to on-going climate change via altering their hydrological regime (Bring et al., 2016) and widespread permafrost thaw that stimulates nutrient release (Vonk et al., 2015). For carbon (C), the particulate fraction (POC) contributes substantially to the total organic C export from the continent to the ocean (Schlesinger and Melack, 1981; Lal, 2003; Ludwig and Probst, 1996; Galy et al., 2015; Li et al., 2017l; Coppola et al., 2018); a two-fold increase of Arctic rivers POC fluxes by 2100 is predicted (Gordeev and Kravchishina, 2009). Although the reasons for strong variations of POC in freshwaters are not yet fully understood (Tiang et al., 2015; Lee et al., 2015; Yang et al., 2016), the temperature (Hilton, 2016) and runoff (Goni et al., 2015) combined with local storm events (Jeong et al., 2012; Wiegner et al., 2009) are widely recognized as the most important driving factors. This may be especially true for northern aquatic systems, being highly sensitive to flood events, due to shallow water paths and short transit time in watersheds.

Of special interest to POC of the Arctic rivers is that, if soil organic C escapes degradation during river transport and thus buried in marine sediments, it can contribute to a geological carbon dioxide sink (e.g., Hilton et al., 2015). Further, potentially increased transport of P and N may significantly change primary productivity in riverine ecosystems (Wrona et al. 2016; McClelland et al. 2007), thereby impeding rigorous predictions of climate change impact on Arctic terrestrial-aquatic ecosystems. Despite significant efforts in characterizing the fluxes, chemistry, and origin of particulate organic matter (POM) in large Arctic Rivers (Lobbes et al., 2000; Dittmar and Kattner, 2003; Unger et al., 2005; Guo et al., 2004, Guo and Macdonald, 2006; Gladyshev et al., 2015; Emmerton et al., 2008; McClelland et al., 2016; Gareis and Lesack, 2017), these studies do not allow for assessment of mechanisms of POM generation in the watershed. In particular, the role of size of the river watershed and its landscape (physio-geographical) parameters is still poorly known. Thus, although detailed studies of particulate nutrients in small Arctic rivers helped to constrain seasonal features of export fluxes (Cai et al., 2008; Dornblaser and Striegl, 2007; Lamoureux and Lafreniére, 2014; McClelland et al., 2014), the key environmental driving factors of particulate nutrient concentration and stoichiometry in Arctic rivers—permafrost coverage and lakes and forest proportion on the watershed—remain poorly resolved.

In this regard, large continental plains such as the western Siberia Lowland (WSL), which contains sizeable reservoirs of frozen and thawed organic carbon, N, P and inorganic nutrients (Sheng et al. 2004; Stepanova et al., 2015; Raudina et al., 2017), may be especially useful in assessing environmental control on particulate nutrient transport to the Arctic Ocean. A vast amount of frozen peat in this region can strongly affect the coastal Arctic system in the event of permafrost thaw and enhanced RSM export from the watersheds. Due to the high homogeneity of the WSL landscape, lithology, and topography, one can use the natural north-south gradient of the permafrost zone distribution to assess the direct impact of permafrost conditions on river water chemistry.

Detailed studies of the dissolved fraction of WSL river waters demonstrated several typical features occurring over a sizeable gradient of climate and permafrost. In pioneering works of Frey and co-workers it was shown that southern permafrost-free regions export 3 to 4 times greater amounts of dissolved C, N and P (Frey and Smith, 2005; Frey et al., 2007a, b; Frey and McClelland, 2009) and that wetlands exert a significant positive effect on carbon and nutrient concentration in small rivers (Frey et al., 2007a; Frey and McClelland, 2009). Although the majority of these features were confirmed by a more recent study of dissolved carbon and nutrients in WSL rivers over main hydrological seasons (Pokrovsky et al., 2015 and Vorobyev et al., 2017, respectively), an assessment of particulate load transport in WSL rivers has not yet been performed and the mechanisms controlling particulate C, N and P mobilization from WSL soils to the Arctic Ocean remain unknown.

To improve current understanding of magnitude and seasonality of riverine particulate nutrient export, we quantified concentrations of C and macro- (N, P) nutrients across a vast latitudinal gradient (1700 km) with special emphasis on the permafrost-bearing zone during three main hydrological regimes: 1) the peak of spring flood (early June 2016), 2) the summer base flow (August 2016), and 3) the autumn high flow before the ice (October 2016). We aimed at characterizing the effect of latitude, permafrost coverage and fundamental landscape features (proportion of bogs, lakes and forest in the watershed) as well as the size of the river itself on particulate C, N and P concentration and the relative fraction of particulate versus total (particulate + dissolved) nutrient transport. We further used acquired knowledge to infer the mechanisms of particulate nutrient mobilization from soils to rivers and applied these mechanisms to predict change in particulate nutrient concentration under climate warming, landscape evolution and progressive permafrost thaw in the largest frozen peatland province in the world.

2. Study Site and Methods

The rivers were sampled in the Western Siberia Lowland (WSL), a huge (> 2 million km²), peatland and forest zone situated in the taiga forest, forest-tundra and tundra zone. The position of biomes follows the decrease of mean annual air temperature (MAAT) from -0.5°C in the south to -9.5°C in the north (Trofimova and Balybina, 2014). The annual precipitation increases from 550 mm at the latitude of Tomsk to 650-700 mm at Nojabrsk and further decreases to 600 mm at the lower reaches of the Taz River. The annual river runoff gradually increases northward, from 160-220 mm y^{-1} in the permafrost-free region to 280-320 mm y^{-1} in the Pur and Taz river basins located in the discontinuous to continuous permafrost zone (Nikitin and Zemtsov, 1986). The permafrost distribution also follows the latitudinal gradient of MAAT and changes from absent, isolated and sporadic in the south to discontinuous and continuous in the north (Baulin et al., 1967). The peat was actively forming since the beginning of the Holocene until freezing of bogs in sub-Boreal period (9-4.5 thousands y.a.). After that, the rate of peat formation in bog areas has decreased (Peregon et al., 2007; Batuev et al., 2015). The main mineral substrates underlying frozen peat layers of the WSL are quaternary clays, sands, and alevrolites (Klinova et al., 2012; Nazarov, 2007). The mineral substrates are quite similar across the WSL and were subjected to strong influence of aeolian processes in the beginning of the Holocene (Velichko et al., 2011). The vegetation of polygonal, mound, and ridge-hollow bogs is essentially oligotrophic and dominated by dwarf shrubs, lichens and mosses. The forest of southern part of the WSL are dominated by Siberian fir, Siberian spruce, Siberian pine, Scots pine, birch and small-leafed Linden. Further details of WSL physio-geographical settings, peat and lithological description of the territory are provided elsewhere (Kremenetski et al., 2003; Stepanova et al., 2015; Pokrovsky et al., 2015; Raudina et al., 2017). For each biome (taiga, forest-tundra and tundra) several rivers with different watershed sizes were chosen and the sampling campaign was performed along a latitudinal transect following previous strategies for WSL river dissolved load (Pokrovsky et al., 2015, 2016; Vorobyev et al., 2017).

Altogether, we sampled 33 rivers that belong to watersheds of Ob, Pur and Taz including these large rivers as well (**Fig. 1**). The landscape parameters of sampled catchments were determined by digitizing available soil, vegetation, lithological and geocryological maps (**Table S1** and Vorobyev et al., 2017). There was no covariation between river size and other landscape parameters including permafrost coverage. Sampling was performed during three main hydrological seasons: 1) spring flood (17 May – 15 June 2016), 2) summer baseflow (1 – 29 August 2016), and 3) autumn baseflow before ice (24 September – 13 October 2016). Note that the most interesting period—in terms of soil connection to the rivers—occurred in late autumn when the active layer depth was at its maximum. This period has not been covered in previous studies of dissolved WSL river load. The reason of sampling both summer and autumn period is to test the role of connectivity between soil fluids and the rivers. In fact, the main factor controlling elemental behavior during accelerating permafrost thaw and release of dissolved and particulate C and nutrients to surrounding aquatic landscapes is the connectivity between soils and rivers or lakes, which occurs via water and solute transport along the permafrost table ("suprapermafrost flow"). The suprapermafrost (shallow subsurface) water occurs in the active layer, typically at the border between the thawed and frozen part of the soil profile (Woo, 2012). In the frozen peatbogs of WSL, the active (unfrozen) layer thickens (ALT) is maximal at the end of unfrozen season, which is typically end of September - beginning of October (Raudina et al., 2018).

The sampling strategy consisted of moving from south to north in spring and autumn over a 2-3 week period, following the natural change of seasons. This allowed us to sample all rivers of the transect at approximately the same time after ice off and before ice on. The year 2016 was normal for western Siberia in terms of spring, summer and autumn precipitation but the temperature was 4 and 2.7 °C higher than normal spring and summer, respectively, and not different from the average T in autumn (Rosgidromet, 2017). For assessing inter-annual variations in RSM concentrations, we analyzed the RSM samples collected in WSL rivers across the same transect during a previous campaign in the spring of 2014 and 2015 and the summer and autumn of 2014 and 2015.

Large water samples were collected from the middle of the river at 0.5 m depth in pre-cleaned polypropylene jars (30 to 50 L) and were allowed to decantate over 2-3 days. The water of the bottom layer of the barrels (approx. 30% of the initial volume) was centrifuged on-site for 20 min at 3500 rpm using 50-mL Nalgene tubes; sediment was frozen at -18°C and freeze-dried later in the laboratory. In addition to decantation and centrifugation, RSM was collected via direct filtration of large volumes (20 to 30 L) of river water with an Inox (AISI 304) Teflon® PTFE-coated filtration unit (Fisher Bioblock) equipped with 142 mm acetate cellulose Sartorius membranes (0.45 µm) and operated at 5-7 bars. An average flow rate of 1-2 L h^{-1} was created by a peristaltic pump (MasterFlex B/T) with Teflon tubing. For determination of total concentration of suspended material, smaller volumes of freshly collected river water (1-2 L) were filtered on-site (at the river bank or in the boat) with pre-weighted acetate cellulose filters (47 mm, 0.45 µm) and Nalgene 250-mL polystyrene filtration units using a Mityvac® manual vacuum pump.

There was reasonably good agreement, typically within 10%, between the concentration of RSM collected in large barrels via decantation followed by centrifugation, a direct high-pressure filtration using 142-mm membranes and vacuum filtration using Nalgene 250-mL unit. The agreement was better than ±10% for large rivers in summer and autumn when the mineral component dominated the RSM. The difference between two methods was between 10 and 20% for small organic-rich rivers containing peat and plant debris especially in spring.

The C and N concentration in RSM collected from large-volume separation procedure was measured using catalytic combustion with Cu-O at 900°C with an uncertainty of ≤ 0.5% using Thermo Flash 2000 CN Analyzer at Tomsk University. The samples were analyzed before and after 1:1 HCl treatment to distinguish between total and inorganic C; however the ratio of $C_{organic}$: $C_{carbonate}$ in RSM was always above 20 and the contribution of carbonate C to total C in the RSM was equal in average 0.3±0.3% (2 s.d., *n* = 30). In addition to RSM, we compared total and HCl-treated C analysis in peat soil column (organic part and 3 separate mineral horizons) sampled from the middle part of river transect. The $C_{carbonate}$ share was below 2 % of total C content for both the mineral and organic part of soil columns. The analyses we performed could not distinguish mineral N linked to clays (NH_4^+ cation) and organic N in the RSM. For P, the RSM samples were subjected to full acid leaching in a clean room following ICP-MS (Agilent 7500 ce) analyses using methods for C_{org}-rich natural samples described by Stepanova et al. (2015). Water samples for DOC and total dissolved phosphorus (P_{tot}) were filtered on-site through 0.45 µm acetate cellulose filters (Millipore, Sartorius) and analyzed following methods previously described by Pokrovsky et al. (2015, 2016).

A regression analysis was used to quantify the relationship between C, N and P concentration in RSM and the % of permafrost, wetlands, lake and forest coverage of the watershed as well as the surface area of the watershed ($S_{watershed}$). In order to assess a general impact of the permafrost on RSM nutrient concentration we separated all sampled rivers into five categories according to the permafrost distribution on their watersheds: 1) permafrost-free (south of 61°N), 2) isolated (61 to 63.5°N); 3) sporadic (63.5 to 65°N); 4) discontinuous (65 to 66°N), and 5) continuous permafrost zones (north of 66°N). The non-parametric statistics were used because, based on Shapiro-Wilk test of the normality of variables, the data on C, N, P concentration in RSM and the % of element in suspended form were not normally distributed. For these reasons, we used the median, 1st and 3rd quartiles to trace dependence of nutrient concentration to the type of permafrost distribution. The differences in suspended C, N and P concentration between different seasons and between each two adjacent permafrost zones were tested using a Mann-Whitney U test for a paired data set with significance level at 0.05. For unpaired data, a non-parametric H-criterion Kruskal-Wallis test was performed for all watershed sizes and all permafrost zones.

3. Results

3.1. C, N and P concentrations in RSM and their link to seasons and watershed size

Mean bulk RSM concentration in the WSL river waters did not depend on the season of open-water period of the year and was equal to 7.1±3.9, 8.1±4.1, and 7.0±3.7 mg L^{-1} in spring, summer and autumn, respectively (**Table 1**). The RSM concentrations weakly depended on the size of the watersheds ($S_{watershed}$) with a negative relationship in autumn ($R^2 = 0.33$, $p < 0.05$, Fig. **S1 A**). Further, the RSM concentration increased with permafrost coverage and latitude ($R^2 = 0.56$ and 0.41), although this was visible only in autumn (**Fig. S1 B, C, Table S2**). The sporadic permafrost zone exhibited the highest RSM concentration in summer (**Fig. S1 D**). Finally, there was no correlation ($p > 0.05$) between lake, bog or forest coverage and the RSM concentration ($R^2 < 0.2$, see also **Table S2**). For RSM concentration, statistically significant difference between different permafrost zones, notably between permafrost-free and permafrost-bearing regions, were evidenced in summer and autumn using Kruskal-Wallis and Mann-Whitney tests (**Table S3**).

The concentrations of C, N and P in WSL rivers averaged over 3 seasons were equal to 15.3±9.7%, 1.2±0.9%, and 0.49±0.42% in mass of RSM (1.05±0.805, 0.083±0.066, and 0.035±0.036 mg L^{-1} in the riverwater). The watershed size sizably affected the C concentration: there was a power-law decrease of C with the size of watershed (R^2 = 0.28, 0.47, and 0.25 in spring, summer and autumn, respectively **Fig. 2A)** but there was no relationship with the N and P concentrations in RSM ($R^2 < 0.2$, **Fig. 2 B, C**). Generally, a 2 to 3-fold decrease in C_{org}, from ca. 20-30% in rivers with $S_{watershed}$ < 100 km² to C_{org} = 5-10% in rivers with $S_{watershed}$ > 10,000 km² was observed. The C:N ratio of RSM was independent on the watershed size in spring but decreased 2-3 times with $S_{watershed}$ increase ($R^2 = 0.4$) in summer and autumn (**Fig. 2D**).

The inter-annual variations of suspended nutrient concentration in WSL rivers were of secondary order importance when compared to season and watershed size control. We did not find any inter-annual differences (at $p < 0.05$) in RSM concentration and P concentration in RSM collected in June and August in 2014, 2015, and 2016 for the same 8 rivers (Agan, Trom'egan, Pyakopur, Aivasedapur, Purpe, Yamsovery, Pur and Taz, **Table S1**)

3.2. Role of permafrost distribution and landscape parameters for C, N, and P concentration and fraction of particulate nutrients

There was a local maximum of C and N concentration in isolated and sporadic permafrost zone (**Fig. 3 A, B, D, E**), which was not seen for P (**Fig. 4 C, F**). Overall, the differences in C and N concentrations in RSM among different permafrost zones were significant as verified by the non-parametric Kruckal-Wallis H-test ($0.005 < p < 0.05$), while the difference in P concentration between permafrost zones was not significant ($p > 0.05$, see **Table S3 C, D**). Specifically, the C demonstrated a maximum concentration (significant at $p < 0.02$ during all three seasons) at 62-64°N (**Fig. S2 A**). The latitude *per se* did not impact N and P concentration in RSM (**Fig. S2 B, C)**. However, significant differences between adjacent permafrost zones were evidenced by C and N in summer and autumn (**Table S3 D**).

The landscape parameters of the watershed (bogs, lakes and forest coverage) sizably affected ($p < 0.05$) suspended C and N. Bogs and lakes in the watershed increased the concentration of C and N in RSM whereas forest generally decreased C in RSM (**Fig. 4 A-B-C for C, and Fig. S3 A-B-C for N**). This increase in C and N % with bogs and lakes coverage and a decrease with forest presence was mostly visible in summer and autumn. The increase in lake coverage of the watershed led to a decrease in P concentration in RSM in summer and autumn ($R^2 = 0.31$ and 0.22, respectively, **Fig. S3 D-E-F)** that was especially visible in autumn in the permafrost-free zone ($R = -0.88$, **Table S2**). During this period, the P concentration in RSM positively correlated with the presence of forest in the permafrost zone ($R = 0.60$, **Table S2**).

The Mann Whitney U-test for the impact of watershed parameters demonstrated significant differences in C and N concentration (all seasons) and P concentration (summer baseflow) between watersheds having < 10% and > 10% lake coverage, **Table S3-E**. The differences were also observed among watershed with < 50% and > 50% of bogs for C (all seasons) and N (summer and autumn), **Table S3-F**. Finally, the forest coverage (< 30% and > 30%) exhibited significant effect on C and N (all seasons) and P (autumn baseflow), **Table S3-G**.

The share of particulate carbon versus total (dissolved + particulate C) did not demonstrate any significant dependence on $S_{watershed}$, bogs, forest and permafrost proportions on the watershed ($R^2 < 0.3$, not shown). However, there was a localized maximum of particulate carbon fraction around 64°N within the isolated to sporadic permafrost zone (**Fig. 5 A** and **C**). The presence of lakes sizably increased the particulate over total transport of C in rivers ($R^2 = 0.52$ and 0.32 in spring and summer, respectively, **Fig. 5 B)**. The share of particulate phosphorus versus total ranged from 10 to 90%. It did not demonstrate any link to size of river watershed, % of forest and bogs, and type of permafrost distribution (not shown).

3.3.C, N, P and RSM export fluxes by WSL rivers

Based on available hydrological data, we calculated open water-period fluxes of C, N and P in WSL rivers. This analysis takes into account the spatial and temporal variability of river discharge, performed using hydrological approaches elaborated for the dissolved (< 0.45 µm) fraction of the river water (Pokrovsky et al., 2015, 2016). The seasonal fluxes of C, N, P and RSM export by WSL rivers were calculated separately for spring (May and June), summer (July, August and September) and autumn period (September-October) for each 2° - wide latitudinal belt of the full WSL territory, following the approach developed for dissolved C and major and trace elements in the river water (**Fig. S4**). These 3 seasons of open-water period represent by far the largest contribution to overall annual element and RSM yield, following the results for other Arctic rivers (McClelland et al., 2016). Thus, 6 ice-covered months (November to April) represent only 12% of annual POC export flux by the Ob River. Based on results of 3 main seasons, an open-water period export fluxes of C, N, P and RSM were calculated (**Fig. 6)**. There is a clear maximum of C and N export fluxes at the beginning of permafrost appearance, in isolated to sporadic permafrost zone. The obtained particulate C and N yields are comparable with other Siberian rivers. For two largest WSL rivers, Pur and Taz, we found May to October export fluxes of 69 and 80 kg C km^2 y^{-1} which is lower than the annual POC yield of the Ob River (191 kg C km^{-2} y^{-1}) but similar to that of the Yenisey River (103 kg C km^{-2} y^{-1}), McClelland et al. (2016).

4. Discussion

4.1. Concentrations of C, N and P in the RSM and impact of the watershed size

The RSM values in WSL rivers (2 to 18 mg/L) are similar to other boreal rivers of low runoff which drain peatlands such as Severnaya Dvina (2.3 to 16 mg/L; Pokrovsky et al., 2010) but lower than the Ob River itself (around 30 mg/L; Gebhardt et al., 2004) and other big rivers of the Kara Sea basin (average 22 mg/L; Gordeev et al., 1996). The POC values of the WSL rivers (0.5 to 3.0 mg/L POC) are consistent with recent data on WSL river transects sampled in 2015 (Vorobyev et al., 2017) and are in agreement with those of the Ob-Taz River confluence measured in June (1.3 mg/L; Gebhardt et al., 2004), the Ob River at Salekhard in May through October (0.8 to 2.4 mg POC/L; Le Fouest et al., 2013), the low reaches of the Ob River (1.2 to 2.4 mg POC/L; McClelland et al., 2016), the mean multi-annual values of POC in subarctic rivers of Northern Eurasia draining peatlands (3.2, 0.3, 0.9 mg POC/L for S. Dvina, Pechora and Ob as compiled in Gordeev et al., 1996) and the Lena River basin (0.5 mg/L; Kutscher et al., 2015).

However, the C_{org} concentrations in RSM of WSL rivers (5 to 40%), notably in small and medium size (< 10,000-100,000 km²) ones, are an order of magnitude higher than those in other world rivers which drain mineral substrates (typically 1% C_{org} in RSM; Meybeck, 1993) and significantly higher than the values of the Siberian rivers (2.3, 3.6, 5.8, 3.0% for Ob, Yenisey, Lena and Kolyma, respectively; Gordeev and Kravchishina, 2009). For example, typical concentration of C_{org} in RSM of large ($S_{watershed}$ > 100,000 km²) Central Siberian rivers that drain larch forest is only 0.4 to 0.5 % (Pokrovsky et al., 2005). The C_{org} concentration in the RSM of Severnaya Dvina River (which has sizeable proportion of bogs and lakes within its watershed compared to WSL rivers) is 2.7±0.7% in May and 4.8±1.1% in August (Savenko et al., 2004). The N_{org} content in RSM ranges from 0.3 to 1.8 % (0.05 to 0.2 mg particulate N_{tot}/L) which is much higher than that in sedimentary rocks (0.05 to 0.06 %; Houlton et al., 2018) but is comparable with the value reported for the freshwater part of Ob river estuary (0.16 mg N/L; Gebhardt et al., 2004), the Ob River at Salekhard in May to October (0.1 to 0.3 mg PON/L; Le Fouest et al., 2013), the Yukon River (0.14±0.09 mg particulate N/L; Guo and MacDonald, 2006), and small rivers of the North slope of Alaska (0.05 to 0.6 mg PON/L; McClelland et al., 2014).

High concentrations of C (and N) in the RSM of WSL rivers may stem from the organic nature of soils that prevail on river watersheds. The Histosols, one of the dominant soil groups of WSL, are capable of providing a sizeable amount of organic particles given the higher susceptibility of peat to physical disintegration compared to mineral soils. The enrichment of the river water in C-rich particles may occur at both the river bank (especially in small rivers flowing through the wetlands) and within the extensive floodplains via remobilization of organic-rich sediments during high flow periods.

The concentration of C and N in RSM decreased with increase in $S_{watershed}$, thereby illustrating the importance of organic particles in small rivers draining peatlands and the role of mineral matter from bank abrasion in larger rivers. The impact of watershed size is more significant for C than for N. Presumably this is because N is more affected by autochthonous processes and that particulate N may partly be generated from phytoplankton and macrophytes in the river. Small rivers ($S_{watershed}$ < 100-1000 km^2) exhibited the largest scatter in particulate C, N (and P) concentrations. This is probably due to multiple sources of POM and the very short transit time in the watershed that results in fast responses of river particulate load to minor variations in surface hydrology including high sensitivity to local storm events.

The decrease of C:N in the RSM from small to large rivers likely reflected a shift in main origin of suspended matter, from peat in small rivers to more lithogenic (deep soil) in large rivers. This was mostly visible in summer and autumn; in spring the rivers exhibited a very homogeneous C:N signature which may be linked to a dominant source of RSM from bank abrasion and sediment transport as well as deposition within the riparian zone. In fact, the flood plain of the Ob river and other rivers of the WSL extend more than 10 times the width of the main channel (Vorobyev et al., 2015). Note that the C:N ratio in large rivers (>100,000 km²) approaches that of average sedimentary rocks (8.1; Houlton et al., 2018). In this regard, highly homogeneous C:N ratios in particulate load of Arctic rivers (7 to 18 for Mackenzie, Yukon, Kolyma, Lena, Yenisey and Ob regardless of season; McClelland et al., 2016) are interpreted as the mixture of deep soil sources where C:N < 10 (Schädel et al., 2014) and upper organic-rich horizons of soils with elevated C:N (Gentsch et al., 2015). The Ob River demonstrates the youngest POC of all Arctic Rivers (-203 to -220 ‰ $\Delta^{14}C$; McClelland et al., 2016) which certainly indicates a relatively fresh (ca. 1,000-2,000 years old) origin of particulate carbon that is presumably from intermediate peat horizons.

We believe that variations in C:N in RSM reflect different sources of organic material feeding the river depending on seasons and latitudes. A compilation of C:N ratios in peat and mineral horizons as well as in thermokarst lake sediments for four main sites of latitudinal transect considered in this study is given in **Fig. S5 of Supplement**. The range of C:N values in RSM rivers (10 to 20) is closer to that in sediments of thermokarst lakes (20 to 30). Note that the resuspension of sediments may be an important source of water column POC (Yang et al., 2016). The minerotrophic bogs, which are mostly linked to rivers via hydrological networks, have a C:N ratio in upper peat horizons ranging from 24 to 28. In mineral soils of the region, the C:N range is between 10 and 15 regardless of latitude, from the tundra situated Taz River riparian zone to the taiga situated middle channel of the Ob River. For upper organic horizons the C:N is always higher than the bottom mineral horizons. The old alluvial deposits of the Pyakopur River (discontinuous permafrost zone) had only 0.2% of POC with C:N equal to 6. Overall, there is an enrichment in N relative to C in the course of water transport of organic and organo-mineral solid particles from soils and riparian deposits to the river water.

Another important observation following from the consistently low C:N ratios of RSM across rivers of various size and climatic zones is that the flocculation and aggregation of riverine DOM in lothic waters of Siberian lowlands may be quite low. Further, the absence of significant relationship between the lake proportion at the watershed and the C:N ratio implies negligible impact of DOM coagulation due to photolysis (von Wachenfeldt et al. 2008, von Wachenfeldt and Tranvik, 2008) or bacterial activity (von Wachenfeldt et al. 2009), with subsequent transformation of coagulation products (Kortelainen et al. 2006b, 2013) as it is known in European humic lakes. Note that, because the range of C:N in RSM of WSL is far from that reported for DOM in soil solution of boreal taiga (ca. 100, Ilina et al. 2014; 40 to 80, Dymov et al. 2013) and humic (peatland) lakes (> 50, Chupakov et al., 2017), the coagulation of DOM from soil waters producing particles in the rivers is also unlikely.

4.2. A maximum of C and N in the isolated/sporadic permafrost zone and the impact of river watershed characteristics

Complementary to previous results on dissolved (< 0.45 µm) C and N concentrations in WSL rivers acquired by Frey et al. (2007a) and Vorobyev et al. (2017) that demonstrated weak or no impact of permafrost on DOC and DON, the particulate C and N were affected by the presence of permafrost in summer and autumn but not affected by its presence in spring. Moreover, during freshet the permafrost distribution did not influence the bulk RSM concentration in WSL rivers. This strongly implies that the delivery of RSM in rivers, and its chemical composition, are tightly linked to the thickness of the active layer and limited by transport of soil particles over the suprapermafrost flow to the river channel. This thickness is highest in September at the end of the active season. In agreement with this, the C and N demonstrated a maximum concentration and export fluxes at 62-64°N, in the sporadic to isolated permafrost zone, that was most visible during summer and autumn (**Fig. 3 A, B and 6 A, B**). This latitudinal belt can be considered as a large-scale thawing front for the frozen peat which corresponds to the southern boundary of permafrost persistence. It is important to note that that WSL rivers exhibit maximum CO_2 emission fluxes at the sporadic to isolated permafrost belt (Serikova et al., 2018), which could be linked to strong processing of POC and PON in the water column of WSL rivers. Interestingly, that rate of POC biodegradation, leading to potential CO_2 emissions, sizably exceeds that of DOC in boreal humic waters (Attermeyer et al., 2018). Furthermore, a maximum percentage of particulate C over total C (suspended + dissolved) was also in the isolated and sporadic permafrost zones in spring; this maximum shifted to the sporadic permafrost zone in summer and moved northward to the discontinuous permafrost zone in autumn (**Fig. 5 C)**. We believe that this corresponds to a progressive increase in the thickness of the active layer which controls the degree of peat and mineral particles leaching from the soil profile to the river. The thickness of this layer increases from spring to autumn and more importantly it moves northward during this period (Trofimova and Balybina, 2014). Enhanced mobilization of nutrients at the "hot spot" of permafrost thaw in frozen peat landscapes was recently demonstrated on a local scale in western Siberia (Loiko et al., 2017).

The impact of watershed characteristics on particulate C and N was clearly pronounced with increased C and N concentration in RSM where there were increased bog and lake proportions and decreased C and N concentration where there was increasing forest coverage. The stronger impact of lakes compared to bogs on C concentration in RSM suggests that the generation of C-rich particles occurs more efficiently in large water bodies than in stagnant shallow water bodies. Given the very short transit time of water from the surrounding peat to the lakes via suprapermafrost flow (Ala-aho et al., 2018a, b; Raudina et al., 2018), the allochthonous chromophoric DOM-rich material from peat soil water that arrives to the lakes may be subjected to fast degradation and coagulation such as that shown in Scandinavian lakes (Kortelainen et al., 2006b; von Wachenfeldt and Tranvik, 2008). Second, the peat abrasion at the border of the thermokarst lakes and thaw ponds, which are highly abundant in the territory (Polishchuk et al., 2017, 2018), occurs due to wave erosion and thermo-abrasion (Shirokova et al., 2013; Manasypov et al., 2015). Physical disintegration of peat at the lake coast likely generates a large amount of suspended organic-rich material that can be exported to hydrological networks during, for example, lake drainage or through already existing connecting channels (Kirpotin et al., 2008, 2011). Note that the maximal lake coverage of the WSL territory is in the 63°N to 64°N latitudinal belt (Polishchuk et al., 2017) where maximum C and N concentration and RSM export fluxes also occur. Because the majority of thermokarst lakes are isolated water bodies without inlet and outlet, this connectivity is achieved via water movement along the permafrost table in the thawed active layer (Raudina et al., 2018) in the form of so-called suprapermafrost flow between peat bogs, lakes, and rivers.

Finally, for particulate P, neither its concentration nor the particulate fraction were affected by permafrost distribution, probably due to the various processes of biological uptake and mineral precipitation controlling P removal both in soil profile and in the river water. For example, lakes and bogs retained particulate P, similar to that of dissolved P, which is in agreement with global assessments (Bouwman et al., 2013), P behavior in European northern wetlands and lakes (Lidman et al., 2014), and recent results on dissolved P in the WSL rivers (Vorobyev et al., 2017).

4.3. Mechanisms of RSM generation and prospective for climate warming in western Siberia

A framework of particulate C, N and P generation in WSL rivers across the permafrost gradient is shown in **Fig. 7**. We suggest that the concentration and export fluxes of suspended particles depends on both the supply and losses in the catchments. The sources of suspended particles in WSL rivers include: (*i*) vegetation litter which is washed by surficial flow to the river, especially in spring; (*ii*) surface (peat) soil horizons, which are also most active in spring, especially in the north; (*iii*) deep peat and mineral horizons which provide the particles via bank abrasion in spring and via suprapermafrost flow in summer and autumn, (*iv*) lake coastal abrasion due to wave erosion, and finally, (*v*) autochthonous organic debris of macrophytes, periphyton and phytoplankton, whose contribution is maximal in summer and autumn. A non-steady-state physical erosion of peat soils in WSL provides maximum particulate nutrients within the most fragile zone of actively thawing permafrost between 62 and 64°N of the sporadic to isolated permafrost zone. The maximal thickness of the active layer progressively moves north during the active season thereby leading to maximal export of particulate C, N, and P at the thawing front. However, we also suggest that part of the differences in mobilized particulates is masked by retention in recipient waters. The transit time of water and particles in the southern WSL rivers is much longer than that in northern rivers (Ala-aho et al., 2018a, b) hence the biological uptake mechanisms (Attermeyer et al., 2018) together with physio-chemical processes such as photo-degradation of POC (Mayer et al., 2006; Riggsbee et al., 2008) or cryocoagulation (Pokrovsky et al., 2018) have sufficient time to act on suspended matter of soil and shallow subsurface waters and to remove the nutrients from the river water as well. In rivers of the continuous permafrost zone, a relatively small stock of nutrient-rich particles within the soil profile and on soil surface (as plant litter) is largely compensated for by a more rapid flushing and shorter travel time through soils and rivers and also lower microbial and phytoplankton activity. As a result, the zone of sporadic to isolated permafrost exhibits both maximal release of soil particles and minimal uptake by in-stream processes. Further to the north, shallow unfrozen peat depth and low biomass cannot supply sufficiently high suspended nutrients and the particulate transport of C and N decreases. In contrast, for P, opposite gradients in supply versus in stream removal may cancel out the net effect of temperature and permafrost on suspended P in the river water.

Based on these results we can speculate on the conditions following warming and permafrost thaw. The lakes drainage and bogs colonization by forest is very common scenario of landscape evolution in Western Siberia under on-going climate warming (Kirpotin et al., 2009; 2011). Scenarios of thermokarst lake evolution under climate warming and permafrost thaw in western Siberia include 1) draining of large thermokarst lakes into hydrological network, which is especially pronounced in discontinuous permafrost zone (Smith et al., 2005; Polishchuk et al.,

2014) and 2) appearance of new depressions, subsidences and small thaw ponds (< 100-1000 m²), which is evidenced across all permafrost zones of this region (Shirokova et al., 2013; Bryksina and Polishchuk, 2015). In terms of landscape change, the area of hollows and subsidences will increase and the coverage of palsa by mounds and polygons will decrease (Moskalenko, 2012; Pastukhov and Kaverin, 2016; Pastukhov et al., 2016). On a short-term prospective (10-50 years), assuming a soil temperature rise of 0.15 to 0.3 degree per 10 years in WSL (Pavlov et al., 2009; Anisimov et al., 2012), the northern part of the WSL (discontinuous and continuous permafrost zones) will transform into sporadic and isolated permafrost zones (Anisimov and Reneva, 2006). This will lead to increase in C and N concentrations in RSM, C and N particulate export yield of the watershed, and overall increase in particulate versus dissolved transport of C and P. Given the contemporary maximum of C and N at the permafrost thawing front, this increase may be two-fold. However, on a longer prospective (50-100 years), even the continuous permafrost zone may disappear (Romanovsky et al., 2008; Nadyozhina et al., 2008) and this will decrease the particulate C and N concentration in the northern rivers and, consequently, their export to the coastal zone of the Kara Sea. Judging from the actual difference in nutrient concentrations and fluxes among adjusting permafrost zones, this decrease may be around a factor of 2 to 3. Furthermore, on the same long-term prospective, the drainage of lakes and disappearance of bogs due to colonization of northern palsa by forests (Anisimov et al., 2011; Anisimov and Sherstiukov, 2016; Kirpotin et al., 2008, 2009, 2011) should lead to a further decrease in particulate nutrient load of WSL rivers.

Conclusions

Relatively low bulk RSM concentration in WSL rivers stems from low runoff in this flat peatland province of boreal and subarctic zone. High concentrations of C and N in the RSM of WSL rivers reflect the essentially organic nature of soils across the WSL. At the isolated/sporadic permafrost zone, we observed a maximum concentration of C and N in the RSM, maximal fraction of particulate OC relative to total (dissolved + particulate), and maximal export fluxes. This suggests the enhanced generation of C, N-rich RSM at the thawing front of permafrost, where thickness of the active layer is maximal. The C and N concentrations in particulate load of WSL rivers decrease with forest coverage of the watershed and increase with the proportion of lakes and bogs; however, the bulk concentration of RSM did not depend on landscape parameters of the watersheds. This implies generation of C, N-rich particles via coastal peat abrasion and sediment resuspension rather than photo- and bio-coagulation of DOM in lentic surface waters which are hydrologically connected to rivers. Indeed, the consistently low C:N ratios of RSM suggest low importance of flocculation/aggregation of DOM in WSL inland waters. To model a northward permafrost boundary and forest line shifting with increase in air and soil temperature we used a substituting space for time scenario of climate warming in the WSL that was well developed for the dissolved fraction of C and nutrients. From a short-term climate warming prospective, the effect of a northward shift of permafrost boundary may produce about a two-fold increase in particulate C and N concentration and export fluxes in rivers of the discontinuous and continuous permafrost zones, and thus may enhance the delivery of these nutrients by the most northern WSL rivers to the Arctic Ocean. On a long-term prospective, the disappearance of permafrost in the northern part of WSL will decrease the concentrations and export of these nutrients to their current level. The P is unlikely to be significantly affected by permafrost change. Moreover, within a long-term climate warming scenario, the drainage of lakes and transformation of bogs to forest may decrease nutrient concentration in RSM and corresponding export flux to the Arctic Ocean.

Acknowledgements:

This work was mainly supported by RSCF No 18-17-00237 “Mechanisms of hydrochemical runoff of the Ob river...” (analyses, modeling). Partial support from RFBR project № 18-35-00563\18, Ministry of Education and Science of the Russian Federation № 6.7515.2017/9.1, and VR (the Swedish Research Council) grant no. 325-2014-6898 is also acknowledged.

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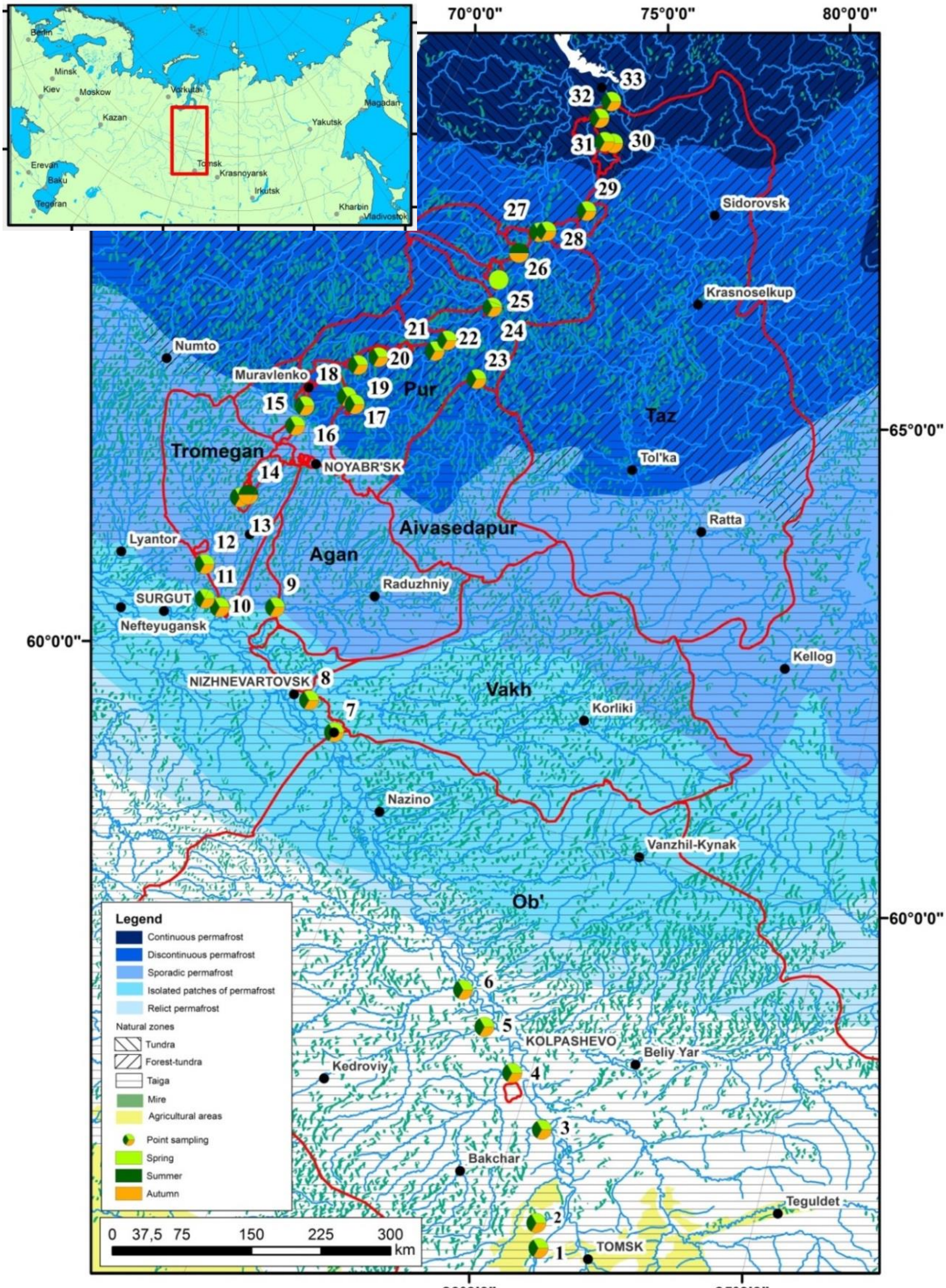

Fig. 1. Sampling sites and physio-geographical context of WSL territory investigated in this work. The sampling numbers are explained in Table S1.

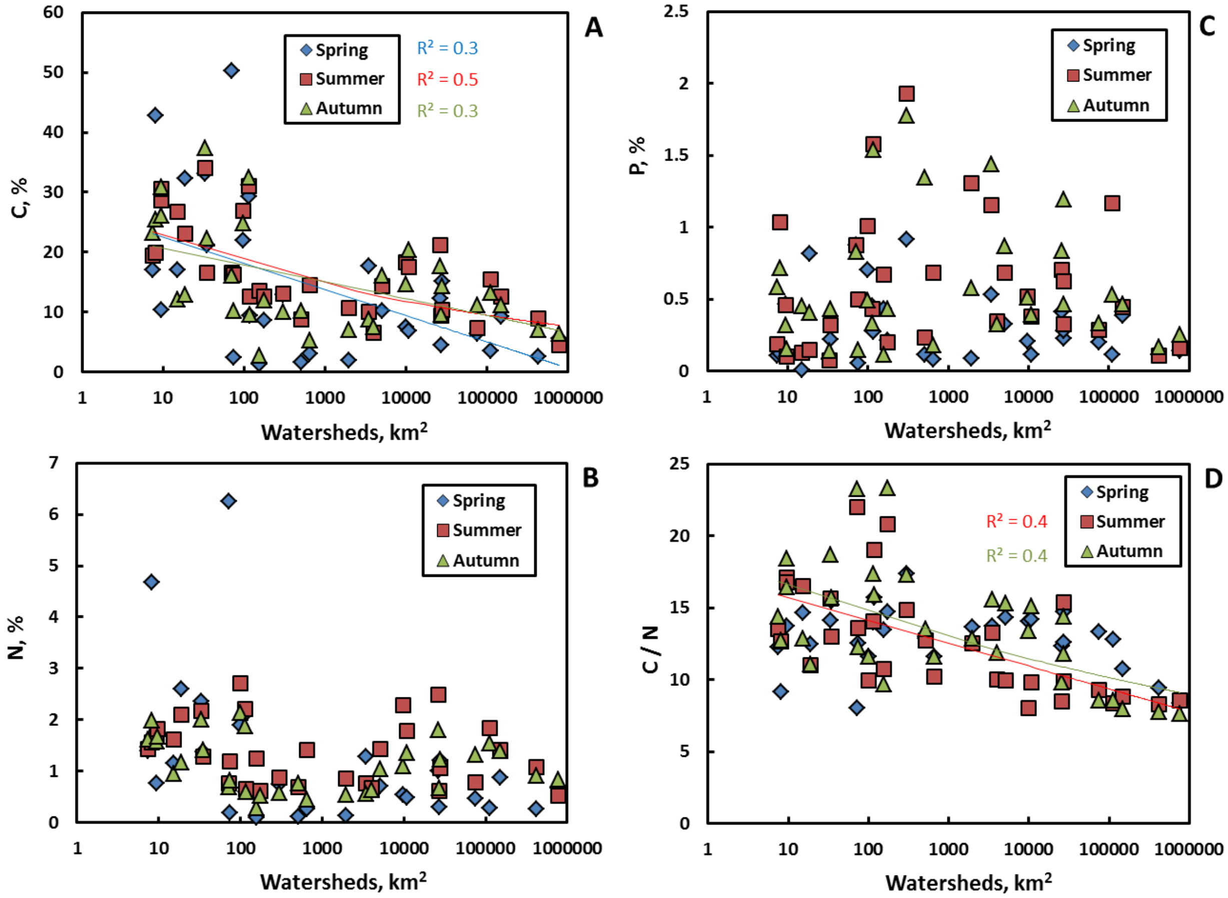

Fig. 2. Particulate (> 0.45 µm) C (**A**), N (**B**), P (**C**) concentration in the RMS (%) and C: N ratio (**D**) in RSM as a function of river watershed size. The solid lines represent a power law fitting of the data with regression coefficients shown for each season in corresponding panels. Only the curves with $R^2 > 0.3$ are depicted.

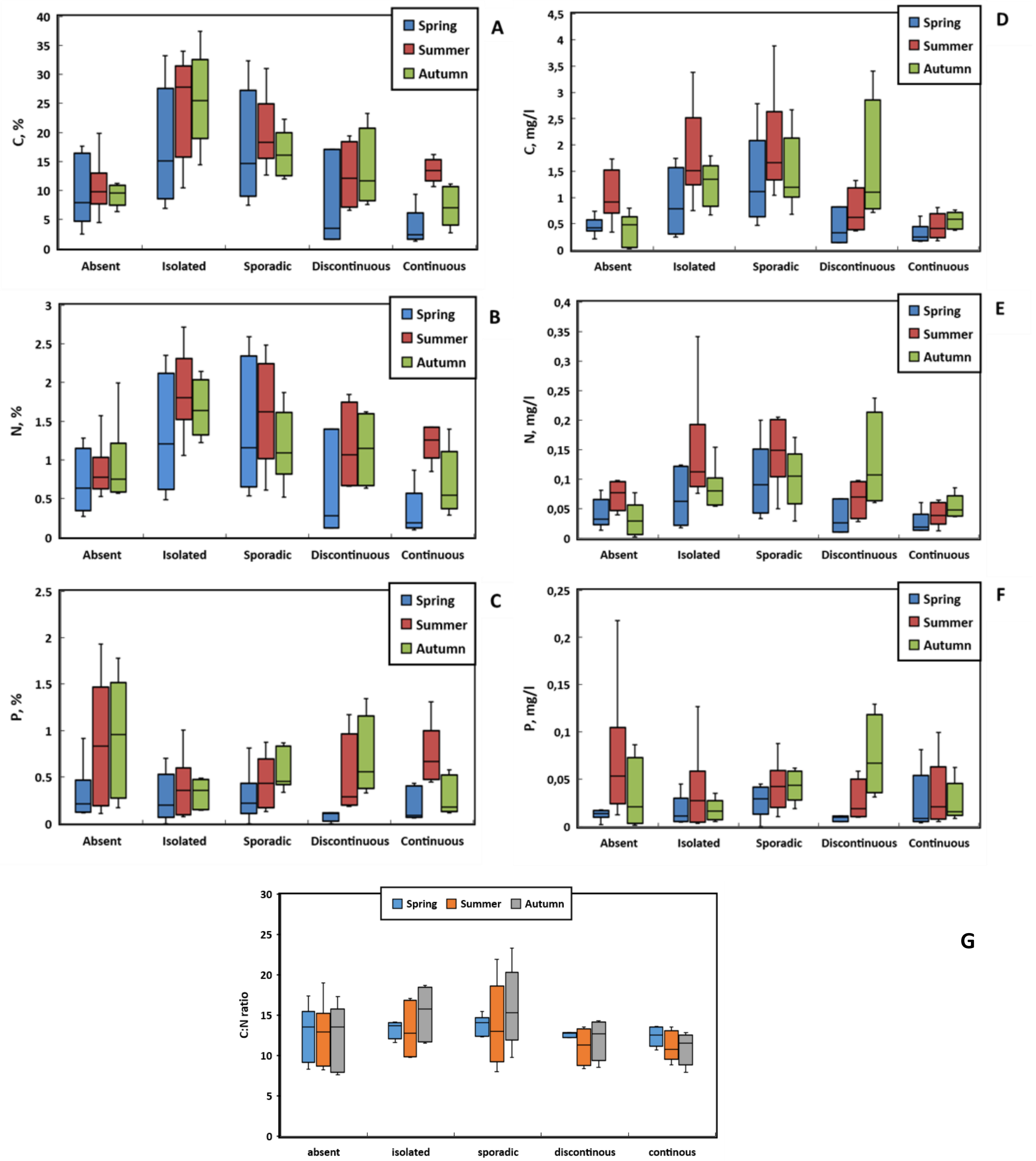

Fig. 3. Box plot of first and third quartiles (25 and 75%) of C (**A**), N (**B**) and P (**C**) concentration in RSM (%) in five permafrost zones over three seasons. The C, N and P concentrations in the river water are shown in panels **D**, **E** and **F**, respectively, and a C:N ratio is shown in **G.**

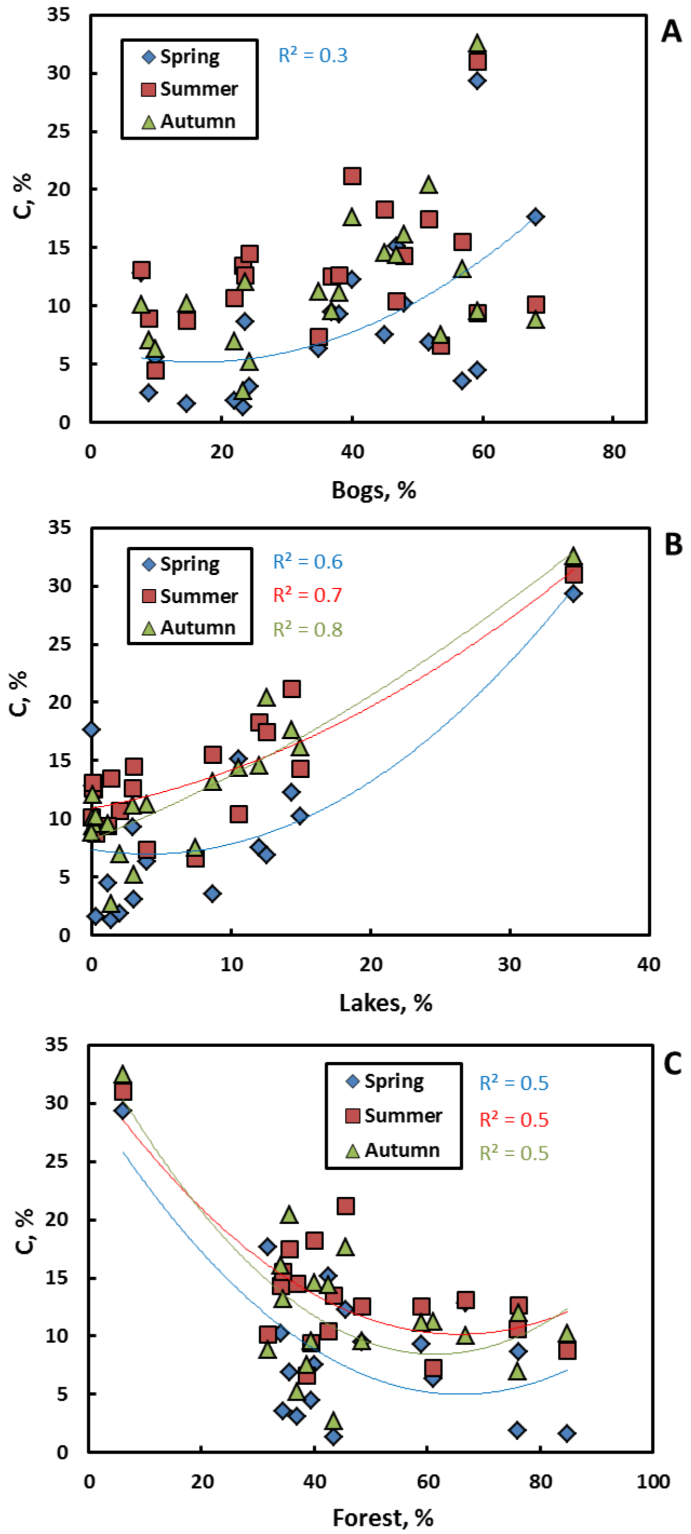

Fig. 4. The dependence of C concentration in RSM (%) on the coverage of watershed by bogs (**A**), lakes (**B**) and forest (**C**). The solid lines represent 2nd degree polynomial fitting of the data with regression coefficients shown for each season in corresponding panels.

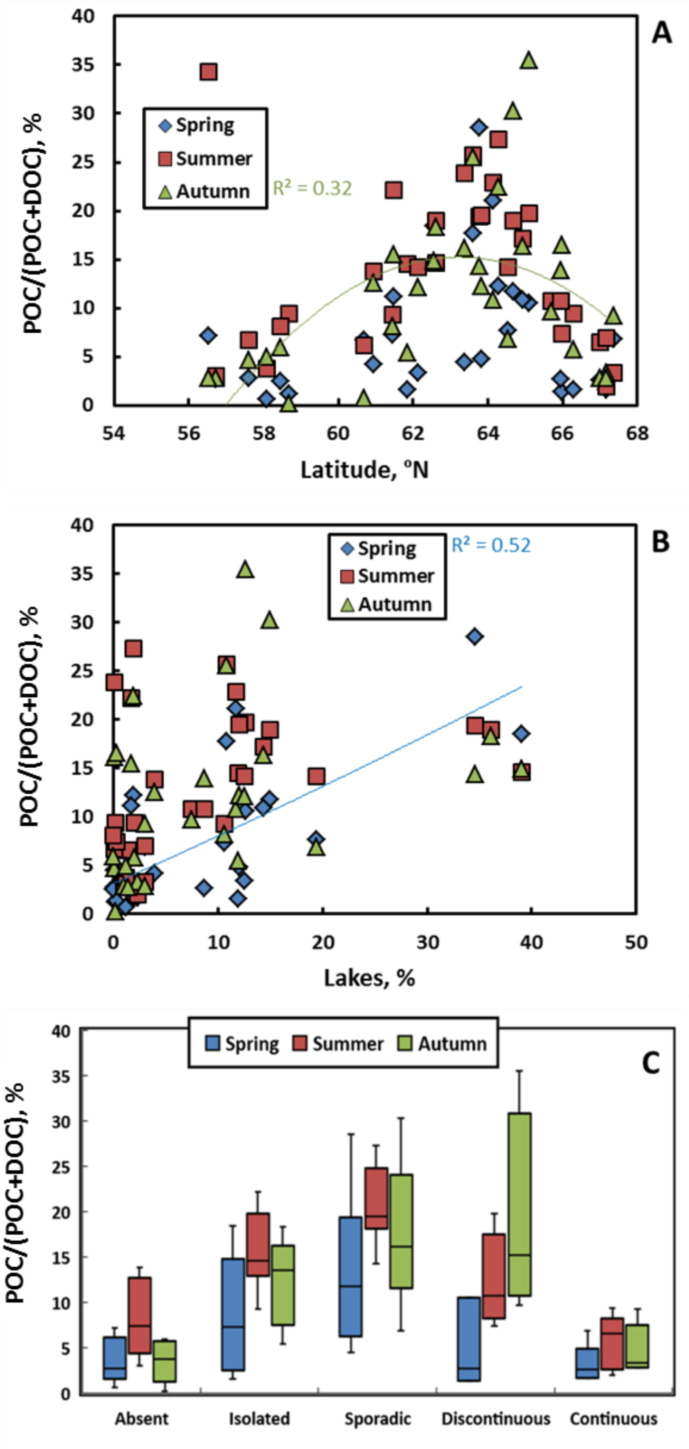

Fig. 5. Fraction of particulate OC of total (dissolved + particulate) form plotted as a function of latitude (**A**), lake fraction on the watershed (**B**) and a box plot of fractions for 5 permafrost zones (**C**). The solid lines in A and B represent 2nd degree polynomial (A, autumn) and linear (B, spring) fitting of the data with regression coefficients equal to 0.32 and 0.52, respectively.

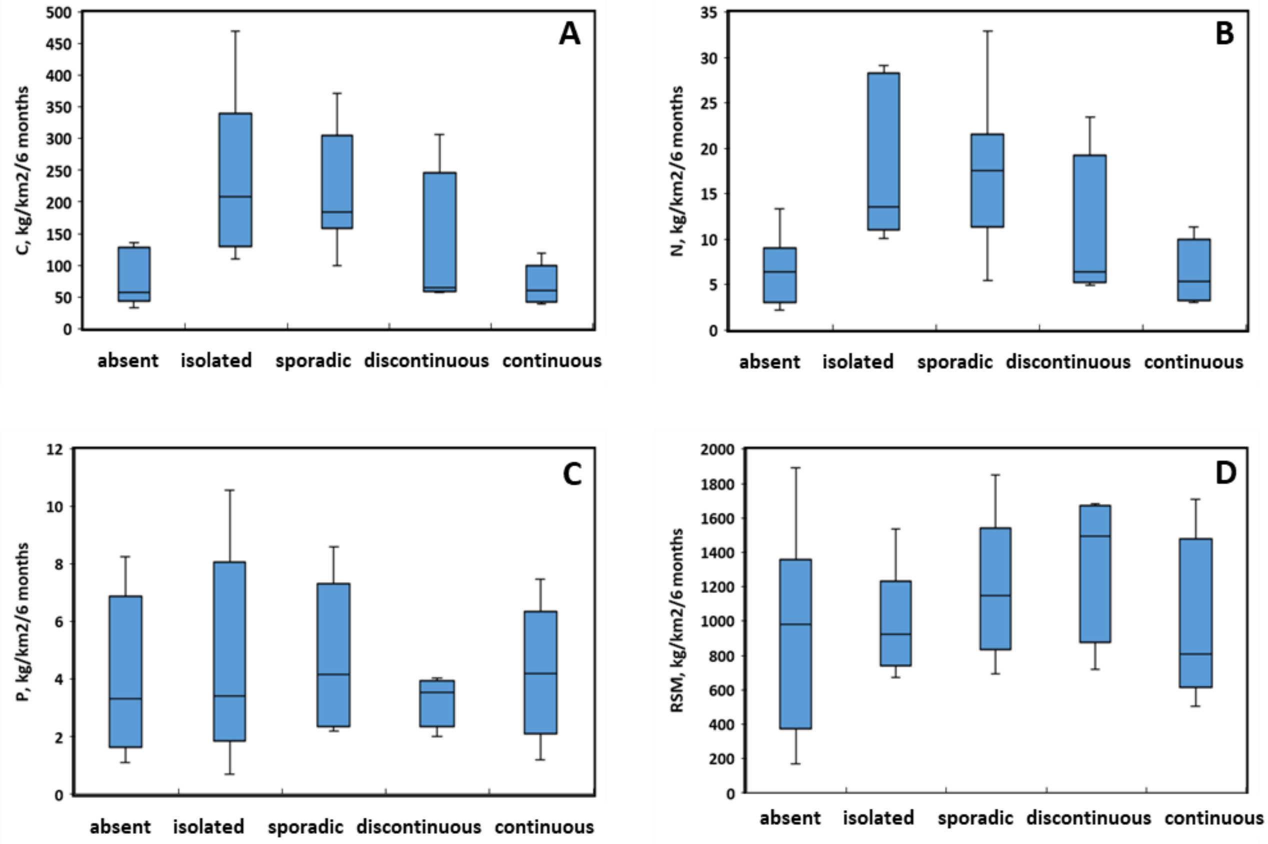

Fig. 6. Total open-water seasons (May to October) fluxes of particulate C (**A**), N (**B**), P (**C**) and suspended matter (**D**) in 5 permafrost-free and 4 distinct permafrost zones of WSL (box plots of 1st and 3rd quartiles). There is a clear maximum of C and N export at the beginning of permafrost appearance, in isolated to sporadic permafrost zone.

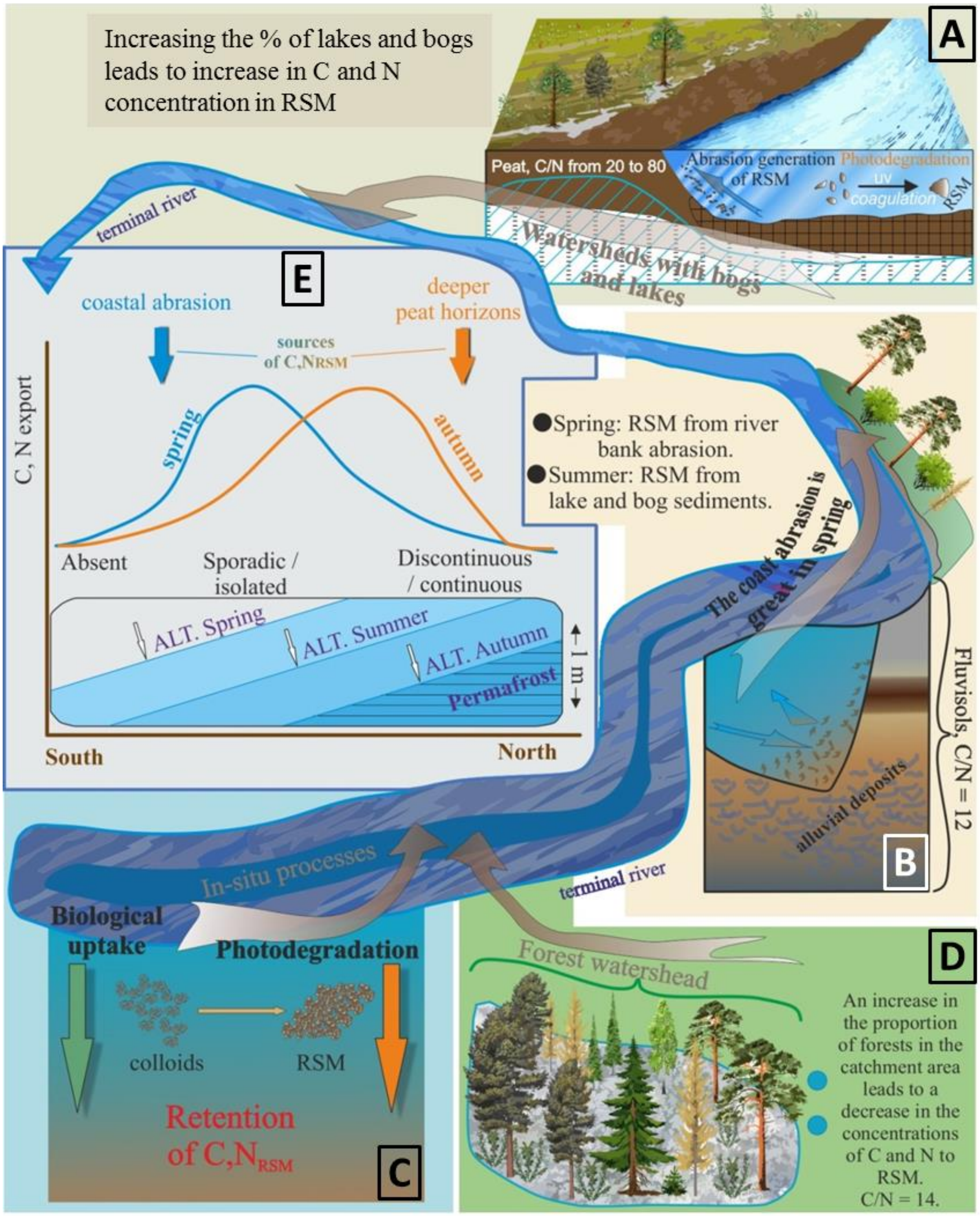

Fig. 7. A cartoon of spatial and temporal partitioning of particulate nutrients in WSL rivers across the permafrost gradient. The panels **A**, **B**, **C** and **D** represent from main sources (**A**, lakes and bogs in summer and **B**, alluvial deposits in spring) and sinks (**C**, photo-and bio-degradation) and **D**, uptake by taiga forest) of particulate nutrients in WSL rivers. The panel **E** depicts the spatial gradient of C and N in RSM occurring in spring (blue line) and autumn (red line). A non steady-state physical erosion of peat soils in WSL provides the maximum of particulate nutrients within the zone of most "fragile", actively thawing permafrost. The maximal thickness of active layer progressively moves to the north during the active season thus leading to the maximal removal of particulate C, N, and P at the thawing front.

Table 1. Mean (± SD) values of RSM, C, N, P concentration (mass %) and relative proportion of suspended C and P overall total concentration for 5 permafrost zones and 3 seasons across the WSL transect.

Season	Variable	Permafrost				
		Absent	Isolated	Sporadic	Discontinuous	Continuous
Spring	RSM, mg/l	6.2±4.9	4.9±1.5	7.2±3.0	7.7±2.5	10.2±4.9
	C, %	12.7±13.0	17.5±6.5	21±14	7.4±8.5	3.6±3.2
	N, %	1.4±1.5	1.3±0.8	1.8±1.8	0.6±0.7	0.3±0.3
	P, %	0.32±0.28	0.33±0.26	0.30±0.25	0.11±0.004	0.21±0.18
	% C_{RSM} of total C	3.5±2.4	8.4±6.7	13.2±7.9	4.9±5.0	3.1±2.2
	% P_{RSM} of total P	30.0±21.5	59.2±18.7	55.6±21.9	40.2±36.2	44.5±30.4
Summer	RSM, mg/l	10.0±4.6	7.5±2.9	10.2±3.7	5.8±1.5	3.6±2.5
	C, %	10.7±4.6	24.7±8.9	20.0±6.0	12.6±5.9	13.5±2.1
	N, %	0.9±0.3	1.9±0.6	1.6±0.7	1.2±0.6	1.2±0.2
	P, %	0.86±0.68	0.39±0.34	0.45±0.27	0.48±0.46	0.72±0.34
	% C_{RSM} of total C	10.7±10.1	15.6±4.4	21.0±4.2	12.2±5.3	5.6±3.0
	% P_{RSM} of P total	57.0±25.2	53.5±21.8	67.9±17.8	55.1±28.7	32.6±18.7
Autumn	RSM, mg/l	3.4±2.4	5.1±1.4	8.7±3.3	10.7±2.6	8.9±3.4
	C, %	11.0±6.0	25.7±8.0	17.4±6.5	13.6±6.9	7.3±3.5
	N, %	0.9±0.5	1.7±0.4	1.2±0.5	1.1±0.5	0.7±0.4
	P, %	0.93±0.64	0.33±0.15	0.57±0.21	0.70±0.45	0.30±0.21
	% C_{RSM} of total C	4.35±3.9	12.4±4.8	17.2±7.5	18.9±11.4	4.8±2.8
	% P_{RSM} of P total	42.8±32.7	71.9±9.9	82.8±11.4	76.9±14.0	40.8±8.6