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Contributions of terrestrial organic carbon to northern lake sediments

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Abstract

Sediments of northern lakes sequester large amounts of organic carbon (OC), but direct evidence of the relative importance of their sources is lacking. We used stable isotope ratios of nonexchangeable hydrogen (δ2H) in topsoil, algae, and surface sediments in order to measure the relative contribution of terrestrial OC to the sediments of lakes from boreal and arctic regions. Catchment properties and light explain trends in OC sources in sediments across lakes. However, differences between mountainous arctic and lowland boreal lakes suggest that particulate and dissolved OC are the predominant sources of sediment terrestrial OC in the arctic and boreal lakes, respectively.

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Author Contribution Statement: CG and JK came up with the research question and designed the field survey. CG conducted the field survey. CG, KK, and CC designed the sample treatment and CG and CC conducted the sample preparation and laboratory work. CC, CG, and SF contributed with sediment chemical analyses. MR conducted and YO made possible the isotope analyses. CG conducted the statistical analyses and AA conducted the analyses of the mixing models analyses with the Monte Carlo approach. CG wrote the paper with contributions from all authors.

Data Availability Statement: Data are available in the Figshare repository at https://figshare.com/s/16c45acf8ee91b71a11c%20

Additional Supporting Information may be found in the online version of this article.

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Quantifying the lateral transfer and fate of organic matter moving between land and water is critical for understanding landscape carbon (C) cycling. Lakes of the northern hemisphere bury significant amounts of organic carbon (OC), and the stocks of OC are comparable to peatlands, soils, and terrestrial biomass (Molot and Dillon 1996; Kortelainen et al. 2004). Sediment OC originates from internal primary production (autochthonous) as well as from terrestrial primary production (allochthonous) imported from the drainage area. Based on budgeting techniques, models and statistical methods, the catchment-derived inputs of dissolved and particulate OC have been used to attribute significant terrestrial contribution to lake sediments (Molot and Dillon 1996; Stallard 1998; Kortelainen et al. 2004). Burial of allochthonous OC in lake sediments is of great interest but the mechanisms are not fully understood (Cole 2013). This requires direct quantitative estimates of the origin of OC stored in sediments, which are currently lacking.

The burial of OC in northern lakes has increased over the last century (Heathcote et al. 2015). Both climate and land use has been shown to have major impact on burial rates (Anderson et al. 2013, 2014; Lundin et al. 2015). However, the exact mechanism causing changes in burial rates is difficult to isolate due to the lack of quantitative OC source separation. Hence, uncertainties in estimating OC sources of lake sedimentary OC pools limit our understanding of the drivers and mechanisms of OC burial, the feedback to environmental changes and the magnitude of the role of lakes in the overall landscape C budget.

The fraction of allochthonous OC buried in lake sediments (i.e., sediment allochthony; SA) depends on the supply and burial efficiency of allochthonous and autochthonous OC. However, the patterns of SA in surface sediments may differ, depending on the extent of the mineralization and protection of OC. Multiple lines of evidence suggest that mineralization of allochthonous OC in lake sediments is strongly constrained, with limited degree of post-depositional degradation (Bastviken et al. 2003; Gudasz et al. 2012; Chmiel et al. 2015). Hence, if we assume a limited loss of allochthonous OC, then SA is largely a result of the supply of OC sources, the extent of OC sorptive mineral protection, and the degree of mineralization of autochthonous OC. While the protective sorption to mineral surfaces was not related to OC burial efficiency (Sobek et al. 2009), the relative importance of this mechanism may be masked by the lack of OC source (allochthonous vs. autochthonous) and pathway separation (erosional vs. flocculated allochthonous OC input). Increased supply of allochthonous OC originating in dissolved forms in northern lakes (von Wachenfeldt et al. 2008), along with constrained autochthonous production by e.g., reduced light and nutrient availability (Ask et al. 2009b; Seekell et al. 2015a) are likely to result in high SA. Hence, at the end of the spectrum, a brownwater lake can be expected to exhibit high SA. In contrast, clear-water lakes are expected to primarily source autochthonous OC, thereby decreasing SA in surface sediments (Ask et al. 2009a). Although we can expect a similarly low SA in naturally eutrophic lakes due to increased autochthonous production, the ponds in agricultural landscapes may receive significant amounts of allochthonous OC derived from extensive soil erosion (Downing et al. 2008), which may elevate the SA in both surface and deep sediments.

Stable isotope ratios of C ($\delta^{13}C$), have been used to trace source contribution in bulk particulate OC (POC; Vonk et al. 2014). However, overlapping $\delta^{13}C$ signatures of the sources hamper the identification of fractional contribution (e.g., Cloern et al. 2002). Most recently, the stable isotope ratios of nonexchangeable hydrogen ($\delta^2H_n$) has been used to discriminate between allochthonous and autochthonous sources in bulk organic matter and food webs (Wilkinson et al. 2013; Karlsson et al. 2015). One of the main advantages in using $\delta^2H_n$ compared to $\delta^{13}C$ is the larger separation between the end-members (Wilkinson et al. 2013; Karlsson et al. 2015). However, measurements of $\delta^2H_n$ in bulk sediments are currently lacking, which have long been hampered because of interference from the mineral matrix. A recently developed technique of soil demineralization has been successfully used to measure $\delta^2H_n$ in bulk soils (Ruppenthal et al. 2013), and may be similarly applied to lake sediments.

We surveyed lakes across the arctic and boreal zone in Sweden, in order to quantify the relative contribution of allochthonous matter to surface sediment OC pool (i.e., allochthony; SA). We hypothesized that increasing allochthonous OC inputs (i.e., increasing color and dissolved OC concentration) have higher SA, and that lakes with potentially higher contribution of autochthonous OC such as eutrophic and clear-water lakes have lower SA.

**Materials and methods**

**Study site and sampling**

We investigated 14 lakes in the arctic and boreal regions of Sweden (Fig. 1). The sites of the arctic region were located in the oroarctic tundra (Virtanen et al. 2015) and Nordic mountain birch forest-tundra ecotone (Wielgolaski 2005), while the North and South forest sites belonged to the boreal region (Kerstin et al. 2008). Lake types spanned from clear-water to humic and oligotrophic to eutrophic (see also Tables 1, 2 and Supporting Information). Sediment cores were taken within the deepest area of lakes with an UWITEC gravity corer. Bulk sediment of the upper 0–1 and 0–5 cm layers was sampled and pooled from five sediment cores per lake. Filamentous algae were collected from submerged rocks and branches. The topsoil (upper 5 cm) was sampled at three locations of each catchment close to the lake shoreline. We compiled published and new data on water chemistry (see Table 1).
The absorption coefficient at 420 nm ($a_{420}$) was calculated using Beer–Lambert relation: $a_{420} = \ln(10) \cdot A_{420}/d$, where $A_{420}$ is absorbance at 420 nm and $d$ is cuvette thickness ($\text{m}^{-1}$). Where data were not available, the light attenuation coefficient ($k_d$) was modeled based on the relationship between $a_{420}$ and $k_d$ in a different set of 18 arctic and boreal lakes (see Supporting Information). The percentage of surface light transmitted to the sediment surface ($I_z$) was calculated following:

$$I_z = e^{-k_d \cdot D_s} / C_1 \cdot 100,$$

where $D_s$ is sampling depth (Table 1). The mean light irradiance ($I_m$), a dimensionless estimate of the variation in light climate between lakes, was calculated following:

$$I_m = \frac{1 - e^{-k_d \cdot z_m}}{k_d \cdot z_m},$$

where $z_m$ is lake mean depth. We calculated the benthic primary production (PP) at the sampling depth in the arctic lakes (Ask et al. 2009a), based on lake-specific relationships between depth and PP (see Supporting Information). The benthic PP (i.e., gross PP) was measured from change in dissolved inorganic carbon during 24 h light and dark incubation. The pelagic PP was measured over 4 h at multiple depths using the $^{14}$C incorporation method following Ask et al. (2009b) and converted to daily rates using the ratio of PAR during the incubation period to whole day irradiance. Whole-lake area-weighted pelagic PP was calculated based on lake bathymetry. This likely corresponds to net PP.

All samples were frozen at $-20^\circ\text{C}$ and then freeze-dried. Soil samples collected from three locations were pooled and then sieved through a series of metal sieves down to 2 mm in order to remove the fine roots. Roots were handpicked and removed from the soil samples. All samples were ground in an agate ball mill prior to analysis. To demineralize sediment and soil samples we treated 0.5 g of samples with 40 mL HF (20% vol) and 40 mL HCl 0.1M for 14 h, followed by washing three times with 18.18 M nano pure water (Ruppenthal et al. 2013). After the treatment, the solution containing the solubilized organic matter was re-captured following solid phase extraction using a 1 g Agilent Bond Elut PPL cartridge, which was eluted with methanol and acetone. The rinsed out solution was dried under $N_2$ gas flow, re-solubilized with nano pure water and then added back to the demineralized soils and sediment before freeze-drying and isotopic and elemental analyses.

The sediment water content was determined by drying (36 h at 60°C). The acid treated bulk sediment samples were analyzed for the concentration of C and N with an Elemental Analyzer (EA) (vario EL III, Elementar Analysensysteme, Hanau, Germany). We determined $\delta^{2}H_{in}$ values via steam equilibration of the samples, following the method detailed in Ruppenthal et al. (2013). The $\delta^{2}H$ values were determined with a Vario PyroCube EA (Elementar Analysensysteme, Hanau, Germany) coupled to an isotope-ratio mass spectrometer (IRMS) (Isoprime, GV Instruments, Manchester, United Kingdom).

![Fig. 1. Map of the sampling sites in Sweden and biogeographic regions.](image)
Table 1. Original and literature data used in the analysis. Lake area (LA), drainage area (DA), drainage ratio (catchment : lake area, DR), mean depth ($z_{\text{mean}}$), maximum depth ($z_{\text{max}}$), sampling depth ($D_s$), benthic primary production at the sampling site (PP_b), lake primary production (benthic + pelagic, PP), absorption coefficient at 420 nm ($a_{420}$), dissolved organic carbon (DOC), total phosphorus (TP), total nitrogen (TN), diffuse attenuation coefficient ($k_d$), percentage of surface light transmitted at the sediment surface ($I_z$) and mean light irradiance ($I_m$) in the 14 lakes investigated in this study.

| Lake                  | LA (ha) | DA (ha) | DR      | $z_{\text{mean}}$ (m) | $z_{\text{max}}$ (m) | $D_s$ (m) | PP_b (mg C m$^{-2}$ d$^{-1}$) | PP (mg C m$^{-2}$ d$^{-1}$) | $a_{420}$ (m$^{-1}$) | DOC (mg L$^{-1}$) | TP (mg L$^{-1}$) | TN (mg L$^{-1}$) | $k_d$ (m$^{-1}$) | $I_z$ (%) | $I_m$ (m$^{-1}$) |
|-----------------------|---------|---------|---------|-----------------------|-----------------------|----------|-----------------------------|-----------------------------|---------------------|----------------|----------------|----------------|----------------|---------------|-----------|----------------|
| Sourojävri            | 17.42   | 124.0   | 7.12    | 4.7*                  | 15.8*                 | 14.5     | 3.93*                       | 26.48*                      | 0.3                 | 1.5*           | 7.3*           | 0.080*         | 0.32*         | 0.9           | 0.52      |
| Knivsjön              | 10.85   | 149.0   | 13.73   | 4.5*                  | 10.7*                 | 11.0     | 38.52*                      | 50.79*                      | 1.0                 | 2.4*           | 14.7*          | 0.089*         | 0.41*         | 1.1           | 0.45      |
| Vuorejaure            | 3.50    | 32.0    | 9.14    | 2.8*                  | 8.5*                  | 6.0      | 46.48*                      | 52.95*                      | 1.8                 | 2.8*           | 14.0*          | 0.135*         | 0.44*         | 7.1           | 0.57      |
| Solbacka              | 3.61    | 7.71    | 2.13    | 1.8                   | 5.6                   | 4.0      | 237.16*                     | 261.15*                     | 1.1                 | 9.4            | 17.1          | 0.365          | 0.18*         | 15.7          | 0.85      |
| Almberga              | 5.48    | 30.36   | 5.54    | 3.2*                  | 6.0*                  | 6.0      | 58.49*                      | 69.35*                      | 1.7                 | 4.0*           | 11.2*          | 0.178*         | 0.51*         | 4.8           | 0.49      |
| Övre Björntjämn       | 4.84    | 324.90  | 67.07   | 4.0*                  | 8.0*                  | 7.0      | NA                          | 19.74**                     | 26.1                | 21.1†          | 29.3†          | 0.494†         | 3.50†         | 0.07          |          |
| Lillsjölden           | 0.79    | 29.20   | 36.86   | 2.8†                  | 5.2                   | 4.5      | NA                          | 22.22**                     | 22.4                | 17.0†          | 26.3†          | 0.469†         | 3.20†         | 0.11          |          |
| Strupltjärn           | 3.11    | 83.20   | 26.71   | 3.5†                  | 5.8                   | 5.0      | NA                          | 84.58**                     | 33.5                | 22.1†          | 27.5†          | 0.488†         | 3.55†         | 0.08          |          |
| Stortjärn             | 3.90    | 86.60   | 22.21   | 2.7†                  | 6.7                   | 6.5      | NA                          | 19.87**                     | 32.0                | 21.9†          | 18.8†          | 0.484†         | 4.15†         | 0.09          |          |
| Lilla Sångaren        | 23.8‡   | 238.0‡  | 10.0‡   | 6.6‡                  | 18.4‡                 | 17.0     | NA                          | 2.9‡                        | 6.50‡               | 11.4‡           | 0.467‡         | 0.68          | 0.22‡         |          |
| Svarttjärn            | 0.70‡   | 113.40‡ | 162.0‡  | 3.6‡                  | 6.7‡                  | 6.0      | NA                          | 22.2‡                       | 28.0‡               | 15.1‡           | 0.743‡         | 3.03          | 0.09          |          |
| Strandsjöen           | 123.0¶  | 2167.4¶ | 17.62   | 1.7¶                  | 4.0¶                  | 2.9      | NA                          | 5.5¶                        | 20.8‡               | 41.3‡           | NA             | 0.99‡          | 5.7‡          | 0.48          |          |
| Valloxen              | 279.0¶  | 3019.5¶ | 10.82   | 3.8¶                  | 9.1¶                  | 6.0      | NA                          | 2.5¶                        | 17.5¶               | 46.7¶           | 1.09¶          | 0.63‡         | 2.3§          | 0.38      |
| Fälaren               | 214.0¶  | 2218.2¶ | 10.37   | 1.5¶                  | 2.8¶                  | 2.6      | NA                          | 10.7¶                       | 34.3¶               | 20.5¶           | 1.105¶         | 1.62          | 1.5‡          | 0.37      |

Data from: * Ask et al. (2009b); † Karlsson et al. (2015); § Gudasz et al. (2012); ¶ Chmiel et al. (2015); | http://vattenweb.smhi.se/svanwebb/; || http://vattenweb.smhi.se/mode-larea/; # François Guillemette, Université du Québec à Trois-Rivières, seasonal average, seasonal average; ** Anne Deininger, Umeå University, personal communication; †† arctic lakes; Unless labeled, data original to this study.
Mixing model and statistical analyses

We applied a two-source isotope mass-balance mixing model in order to determine SA:

\[
SA = \frac{(\text{sediment } \delta^2\text{H}_n - \text{autochthonous } \delta^2\text{H}_n)}{(\text{allochthonous } \delta^2\text{H}_n - \text{autochthonous } \delta^2\text{H}_n)}
\]

We used topsoil and periphyton OC as allochthonous and autochthonous end-members, respectively. Periphyton is suitable as autochthonous end-member, given the similar photosynthetic fractionation factors between algae and lake water in periphyton and phytoplankton (Karlsson et al. 2012, 2015). Both allochthonous and autochthonous end-members were aggregated for lakes within a region (i.e., tundra, Nordic mountain birch forest-tundra ecotone, boreal North and boreal South) for subsequent data analyses. To account for the variability of the end-members and the measurements uncertainties in SA estimates, we used a Monte Carlo approach (Andersson 2011; Sheesley et al. 2011). The end-members distributions were represented by normal distributions using empirically determined means and standard deviations. The Monte Carlo approach effectively samples the probability distribution of the fractional source contributions, allowing us to estimate statistical parameters, e.g., mean, median, and standard deviation. The calculations were run with 100,000 iterations using MATLAB version 2014b (Mathworks). A detailed flowchart for the Monte Carlo methodology is described in Andersson (2011). Briefly, random numbers were drawn from the end-member distributions to calculate an estimated fractional contribution that fits with the observations for each iteration. The large number of iterations allows effective sampling of end-member distributions. The calculated mean and median SA as well as the associated uncertainty (standard deviation, 2.5% and 97.5% percentiles) can be found in Supporting Information. For our analyses we used mean SA.

A one-sample t-test was used to test the difference between SA in bulk 1 cm and 5 cm layers. Analyses were conducted in GenStat V17 software. Regression and robust regression analyses were carried out in JMP 12.0, (SAS Institute) and R version 3.3.1 (R Core Team 2016). Data were log-transformed, to satisfy assumptions of homoscedasticity and normality of residuals. We used linear multivariate model by means of partial least-squares projections to latent structures (PLS; Eriksson et al. 2006), to identify relevant variables and their magnitudes of influence in explaining variance in SA (see also Supporting Information).

Results

The \( \delta^2\text{H}_n \) of the algae (\( M = -255, SD = 32_{\%} \)) and near-lake topsoil (\( M = -145, SD = 7_{\%} \)) end-members were on average separated by 110_{\%} (range between 92_{\%} and 128_{\%} across regions; Fig. 2a). The \( \delta^2\text{H}_n \) of algae showed larger variation both within and between regions compared to the \( \delta^2\text{H}_n \) of soils (Fig. 2a). On average, the isotopic composition of bulk surface 1 cm and 5 cm sediment was similar, i.e., \( M = -178, SD = 10_{\%} \) and \( M = -178, SD = 11_{\%} \), respectively.

There was no difference in mean SA between bulk 0–1 cm and 0–5 cm sediments, mean difference of \( -0.00177, (95\% CI, -0.0292, 0.0257) \); \( t(13) = -0.14, p = 0.89 \). We base our further analyses on the 0–5 cm sediments only. The SA

Table 2. Sediment characteristics in sampled lakes. Weight-% water content (WW), % sediment mineral matrix (SMM), % carbon (C), atomic ratio of carbon to nitrogen (C/N). Data original to this study.

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<th>Lake</th>
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*Analyses based on HF-HCl treated sediment.
ranged between 0.46 and 0.80 (Fig. 2b) across lakes (M = 0.66, SD = 0.095). While the SA was somewhat similar in the arctic (M = 0.68, SD = 0.144) and boreal lakes (M = 0.66, SD = 0.065), DOC concentration was markedly lower in the arctic (M = 4.0, SD = 3.1) compared to boreal lakes (M = 21.0, SD = 7.6).

SA was surprisingly high in clear-water arctic Sourojávrí, Knivsjón, and Vuorejaure (0.80, 0.79, and 0.72), which was similar to contrasting boreal humic lakes such as Övre Björntjärn, Svarttjärn, Stortjärn, and Struptjärn (0.76, 0.76, 0.68, and 0.67). Moreover, the values of the lowest observed SA in clear-water arctic (Almberga, 0.46) and eutrophic boreal lakes (Valloxen, 0.57) were unexpectedly large. There were no clear trends with single variables that could alone explain the pattern in SA across all lakes. However, there were distinct trends in SA with in the arctic and boreal regions when analyzed separately. SA increased with increasing proxies of allochthonous OC (Fig. 3a), such as drainage ratio in both arctic (R² = 0.85, p < 0.05) and boreal (R² = 0.68, p < 0.001) lakes. However, SA decreased with increasing percentage of sediment mineral content (p < 0.01) in boreal lakes, while it increased in the arctic lakes (R² = 0.92, p < 0.001) lakes (Fig. 3a). The proxies of autochthonous OC showed that SA decreased with mean light irradiance in both arctic (R² = 0.69, p < 0.05) and boreal (R² = 0.57, p < 0.05) lakes (Fig. 3b). Similarly, SA decreased with increasing PP in arctic (PP, R² = 0.93, p < 0.05) lakes, but it was not significant (R² = 0.07, p = 0.73) in four boreal lakes where data was available (Table 1).

A PLS analysis conducted with additional variables, proxies of allochthonous and autochthonous input, supports the distinct patterns in SA between the regions. In the arctic lakes PLS analysis extracted two significant components (Fig. 4a) that explained 93% of the variance in SA (R²Y = 0.93). The PLS model has high predictive power (Q²Y = 0.72), but higher background correlation (R²Yvalidated = 0.35). The PLS analysis in boreal lakes (Fig. 4b) extracted one significant component from the data matrix, which explained 84% of the variance in SA (R²Y = 0.84, R²Yvalidated = 0.55) and with good predictive power (Q²Y = 0.68).

These models demonstrate that in addition to the significant effects of drainage ratio, sediment mineral content, light, and PP on SA outlined above, there were differences between arctic and boreal lakes. Hence, the sampling depth was moderately important and positively correlated with SA in the arctic, but not important in the boreal model. DOC was moderately important in both models, but negatively correlated with SA in the arctic, while positively correlated in the boreal model. The k₃ and a₄₂₀ were important and positively correlated with SA only in the boreal model and not important in the arctic model. Total nitrogen was moderately important and negatively correlated with SA only in the arctic lakes. Total phosphorus was not important in any of the models.

**Discussion**

This study provides direct evidence for the relative importance of allochthonous vs. autochthonous OC in sediments.
Fig. 3. Sediment allochthony (SA) as a function of allochthonous (a) and autochthonous (b) OC proxies in the arctic (squares) and boreal lakes (circles). Relationships between: (a) drainage ratio (catchment: lake area, DR) and sediment mineral matrix (SMM); (b) arctic lake mean light irradiance (Im) vs. benthic + pelagic lake primary production (squares, PP) and benthic PP (triangles). The triangles show the benthic PP at the sampling site in the arctic lakes. The solid line describes the relationship between SA and DR (arctic $R^2 = 0.85$, $p < 0.05$ and boreal $R^2 = 0.68$, $p < 0.01$), SMM (arctic $R^2 = 0.92$, $p < 0.01$ and boreal $p < 0.0001$), Im (arctic $R^2 = 0.68$, $p < 0.05$ and boreal $R^2 = 0.57$, $p < 0.05$) and between SA and benthic + pelagic PP (arctic $R^2 = 0.93$, $p < 0.05$). Both axes are represented on a log-scale. *Marks a robust linear fit.

Fig. 4. Loadings plots of the PLS regression analysis of sediment allochthony in (a) arctic and (b) boreal lakes. For the boreal model, the PLS component 2 was not significant, but was included to make the separation along the component 1 visible. The graph shows how the Y-variables (red squares) correlate with X-variables (circles), as well as the correlation structure of the Xs and Ys. The X variables are classified according to their VIP scores (variable influence on projection) such as: highly influential (black circles), moderately influential (gray circles), and less influential (white circles). The plot should be read by drawing an imaginary line from a Y-variable through the origin and across the plot, followed by projecting orthogonally each of the X-variables on this line. Thus, the X-variables along this line situated far away from the origin of the plot (on the positive or negative side) are highly correlated with Y and are the most influential for the model. Variables close to the origin of the plot are poor predictors of the Y-variables. The X-variables situated closer to each other and near Y-variables are positively correlated to them and those situated on the opposite side are negatively correlated. Data was log-transformed prior to analyses.
across various lake types. Our results support the suggestion of the prevalence of allochthonous OC in northern lake sediments (Molot and Dillon 1996; Kortelainen et al. 2004). Differences in catchment and lake properties point to a shift in the importance of particulate relative to dissolved allochthonous OC input in the arctic compared to boreal lakes in Sweden.

The long-term buried OC is of ultimate interest. Our study addressed SA in bulk surface 0–1 cm and 0–5 cm sediments, years to decades old (e.g., Chmiel et al. 2015). Based on the strong constraint on allochthonous compared to autochthonous OC mineralization (Bastviken et al. 2003; Gudasz et al. 2012), we expected increased SA in the 0–5 cm compared to 0–1 cm layer. However, we did not find any significant difference. This may indicate that the differences in SA within these sediment depth intervals were not large enough and erased by homogenizing the layers, or that the supply of the allochthonous OC vs. autochthonous OC has changed over time.

We found a mismatch between the high SA in the arctic lake (Sourrajärv and Knivsån) and C/N ratio, a well-known proxy of OC sources in lake sediments (e.g., Meyers 1994). The C/N ratio of the surface sediments (see Table 2) was consistently closer to that indicative of autochthonous OC (C/N between 4 and 10) than of terrestrial sources (C/N > 20), when SA was 0.8. This mismatch between C/N ratios of sediments and that of the OC sources can likely be attributed to enrichment of sediment mineral particles with organic nitrogen during diagenesis. Preferential association of amino acids to clay and silt sediment particles (Hedges and Hare 1987; Tanoue and Handa 1979), and as a consequence their increased protection (Keil et al. 1994) may result in nitrogen enrichment relative to carbon. It is likely that a similar mechanism may have occurred in our lakes and in particular in the silt and clay rich arctic lake sediments (e.g., Sourorjärv, Knivsån, and Vuorejaure, Table 2).

The results of this study suggest that SA is controlled differently in the mountainous arctic compared to the lowland boreal lakes. Drainage ratio was an important predictor in both regions (Figs. 3, 4a,b). However, in boreal lakes the variation in allochthonous DOC input was likely the main factor controlling sediment SA. We found a strong positive correlation between SA and water-color, light attenuation and DOC (Fig. 4b), i.e., factors all related to high supply of allochthonous DOC. It is likely that terrestrial DOC input resulted in elevated SA both directly by providing a source of C to the sediments through DOC flocculation (von Wachenfeldt and Tranvik 2008), as well as indirectly by increasing the light attenuation and thus reducing lake PP (Ask et al. 2009b; Seekell et al. 2015a). This is supported by the strong negative correlation between SA and mean light irradiance (Figs. 3b, 4b). With the exception of Strandsjön, Vallo xen, and Fälaren, none of the boreal lakes had relative light levels at the sampled site sediment surface above 1% (Table 1), which limited the contribution of benthic PP to sediment C. Although light is the main controlling factor for PP in boreal lakes, availability of nutrients also limits pelagic PP. However, we did not find a clear negative relationship between nutrients and SA. The, potential nutrient impact was likely obscured by the negative effects of allochthonous DOC on lake PP (Ask et al. 2009b; Seekell et al. 2015a).

The SA in clear-water arctic lakes was unexpectedly high (Fig. 2b), i.e., comparable to most humic boreal lakes (e.g., Övre Björntjärn). This raises the question of the origin of the allochthonous OC in the high latitude lakes. The primary production in shallow, clear-water arctic lakes is generally dominated by benthic algae (Table 1, Ask et al. 2009a). Hence, we found a strong negative correlation of SA with PP (Figs. 3b, 4a). Changes in light limiting conditions at the sediment surface in these lakes were primarily driven by changes in depth and less by water-color and light attenuation. Based on the calculated percentage of light transmitted to the sediment surface at the sampling sites, it is only in Sourorjärv and Knivsån that light was potentially limiting photosynthesis (< 1% of surface level; Table 1). Thus, the relatively high SA in the arctic lakes cannot be explained by relatively low autochthonous OC input. Unlike boreal lakes, the arctic lakes have low catchment productivity which yields low DOC export and low DOC concentration in lakes (Jansson et al. 2008). Instead, we suggest that the supply of allochthonous POC derived from soil erosion is a relatively more important source for sediment OC in the arctic lakes. The SA and mineral content of sediments were strongly positively correlated (Fig. 3a) and soil mineral content is a known important determinant of the OC stored in soils (Torn et al. 1997). Depending on the precipitation regimes (Klumind et al. 2009) and contributions from nivose aeolian processes (Fahnestock et al. 2000; Bullard et al. 2016), lakes in the arctic areas of high relief may receive significant contributions of soil POC. Hence, a higher potential for increased allochthonous POC relative to allochthonous DOC input, may suggest a divergent predominant source pathway of allochthonous OC in arctic compared to boreal lakes.

A critical question is to what degree the results on SA based on data at the deep location reflect whole lake conditions. Depth integrated measurements of sediment metabolism (Karlsson et al. 2008; Ask et al. 2012) and δ13C (Karlsson et al. 2008, 2009) suggests a strong depth gradient of OC sources in arctic clear-water lakes, but less so in humic lakes. Accordingly, depth emerged as a relatively important variable of SA in arctic but not in boreal lakes. Thus, the lack of strong depth effect in boreal humic lakes indicated that SA likely reflected to a large degree the whole lake SA. However, the decrease in benthic PP with depth in the arctic clear-water lakes (Ask et al. 2009b), suggest that basin-scale estimates of SA are needed for reliable assessments of the relative importance of sediment OC sources in these lakes.

Differences in drainage basin size and topography likely played a role explaining the trends in soil erosional input (Fig. 3a). While drainage ratio was an important variable (Fig. 4a,b), there was no obvious pattern of drainage ratio in Sweden (Seekell et al. 2014). However, Seekell et al. (2014) described two distinct biogeochemical regions, which broadly separated the
mountainous arctic and lowland boreal lakes by a threshold DOC concentration of about 5 mg L\(^{-1}\) (Seekell et al. 2015b). While median SA in the arctic and boreal lakes were similar (about 0.7), median DOC concentration was seven times lower in the arctic compared to boreal lakes. Thus, the transition of the importance of particulate relative to dissolved OC input may follow the regional pattern of DOC in lakes.

Tracing OC sources in lake sediments using \(\delta^13\)C is a novel tool, which provided new insights of lake sediment OC source contribution. Future studies would benefit by including the quantification of the supply of allochthonous and autochthonous OC sources, which would improve our mechanistic understanding of the OC burial in lakes and response to environmental change.

**References**


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