Supplement of Biogeosciences, 15, 457–470, 2018
https://doi.org/10.5194/bg-15-457-2018-supplement

Biogeosciences
Open Access

Supplement of

Quality transformation of dissolved organic carbon during water transit through lakes: contrasting controls by photochemical and biological processes

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Text S1. Supplemental methods

Discharge measurements

Daily discharge for the whole study periods 2012-2014 was extracted from hourly measurements performed for five of the study lakes, either at the outlet or at the inlet. Inlet measurements were re-scaled through multiplication by the ratio of ‘outlet catchment area’ : ‘inlet catchment area’. Water height loggers of model WT-HR 100 (Trutrack Inc., New Zealand) were placed at reaches with well-defined banks, and discharge was calculated using established rating curves based on the salt dilution method (number of observations per stream: 24-32, normalized root mean square error: 0.13-0.29). In the four lakes with the smallest catchments (Lapptjärn, Mångstrettjärn, Nästjärnen and Fisklösan), we considered inlet runoff to be too small to be assessed from water height. These lakes had no continuously flowing inlets and diffuse water flow may dominate during parts of the year. For these lakes, we assumed that specific discharge was identical to that of the lake Övre Björntjärnen, and thus we rescaled the discharge from Övre Björntjärnen according to the catchment size of each of these four lakes. The assumption of similar specific runoff can be justified by the proximity of the lakes (see Table S1), the similar catchment slope gradient (ca 10%) and composition of bedrock (paleoproterozoic granitoids etc.), soils (peat and podzol soils on glacial till) and vegetation (coniferous forest and peatlands).

Prior to 2012, discharge for all sampled lakes was approximated from specific discharge from the stream Kallkällsbäcken in the Krycklan catchment (50 km northeast of Övre Björntjärnen), where stream water levels have been recorded continuously since 1980 using a pressure transducer and a 90° V-notch weir housed in a heated shed (Laudon et al. 2013). Discharge measured at the Övre Björntjärnen inlet, 1996-1998 (Jansson et al. 2001), indicated that the mean specific discharge (approximately 10 L s-1 km-2) was not significantly different from that recorded in the Krycklan catchment over the same period (Köhler et al. 2008). Moreover, manually registered water levels at the inlet of Övre Björntjärnen on a total of 18 dates, 2007-2009, demonstrated a strong correlation with the water level in the Krycklan stream ($R^2 = 0.85$, $n = 18$, $p < 0.01$).

References to supplemental methods Text S1

Jansson, M., A. K. Bergström, S. Drakare, and P. Blomqvist. 2001. Nutrient limitation of bacterioplankton and phytoplankton in humic lakes in northern Sweden. Freshwat. Biol. 46: 653-666.

Köhler, S. J., I. Buffam, H. Laudon, and K. H. Bishop. 2008. Climate's control of intra-annual and interannual variability of total organic carbon concentration and flux in two contrasting boreal landscape elements. J. Geophys. Res. Biogeosciences 113: G03012.

Laudon, H., I. Taberman, A. Agren, M. Futter, M. Ottosson-Lofvenius, and K. Bishop. 2013. The Krycklan Catchment Study-A flagship infrastructure for hydrology, biogeochemistry, and climate research in the boreal landscape. Water Resour. Res. 49: 7154-7158.

Table S1. Statistical details for linear regressions of dissolved organic carbon (DOC) properties as functions of water transit time (yrs). The regressions are used in Fig. 2 and partly in Fig. 3 of the main paper. Full data plots are shown in Fig. S5. Columns from left to right depict the name of lake, water layer ('Epi' for epilimnion and 'hypo' for hypolimnion), response (y) variable, slope ± standard error, y-axis intercept, number of observations (n), explained variance (R^2) and 2-tailed significance of the slope (p).

Site name	Layer	y	Slope ± SE	Intercept	n	R^2	p
Fisklösan	Epi	$a_{254} : a_{365}$	0.42 ± 0.17	4.15	38	0.14	0.019
Nästjärnen	Epi	$a_{254} : a_{365}$	0.45 ± 0.13	3.10	40	0.23	0.002
Mångstrettjärn	Epi	$a_{254} : a_{365}$	0.11 ± 0.06	3.97	41	0.08	0.081
Lapptjärn	Epi	$a_{254} : a_{365}$	0.20 ± 0.08	3.98	41	0.14	0.015
Lillsjöliden	Epi	$a_{254} : a_{365}$	-0.08 ± 0.19	4.11	35	0.00	0.696
Nedre Björntjärnen	Epi	$a_{254} : a_{365}$	-0.11 ± 0.09	4.05	59	0.03	0.220
Struptjärnen	Epi	$a_{254} : a_{365}$	-0.22 ± 0.10	4.10	38	0.14	0.027
Övre Björntjärnen	Epi	$a_{254} : a_{365}$	-0.41 ± 0.14	4.11	72	0.11	0.004
Stortjärnen	Epi	$a_{254} : a_{365}$	-0.50 ± 0.07	4.02	31	0.44	0.000
Fisklösan	Epi	a_{420} : DOC	-0.06 ± 0.04	0.38	38	0.07	0.113
Nästjärnen	Epi	a_{420} : DOC	-0.08 ± 0.03	0.60	40	0.19	0.005
Mångstrettjärn	Epi	a_{420} : DOC	-0.02 ± 0.02	0.51	41	0.02	0.368
Lapptjärn	Epi	a_{420} : DOC	-0.04 ± 0.04	0.51	41	0.03	0.315
Lillsjöliden	Epi	a_{420} : DOC	-0.03 ± 0.06	0.50	35	0.00	0.662
Nedre Björntjärnen	Epi	a_{420} : DOC	0.08 ± 0.05	0.55	59	0.05	0.089
Struptjärnen	Epi	a_{420} : DOC	-0.02 ± 0.06	0.56	38	0.00	0.716
Övre Björntjärnen	Epi	a_{420} : DOC	0.05 ± 0.08	0.54	72	0.01	0.542
Stortjärnen	Epi	a_{420} : DOC	0.15 ± 0.05	0.57	31	0.26	0.004
Fisklösan	Epi	a_{420} (m^{-1})	-0.64 ± 0.28	3.07	38	0.12	0.028
Nästjärnen	Epi	a_{420} (m^{-1})	-0.69 ± 0.23	4.85	40	0.19	0.004
Mångstrettjärn	Epi	a_{420} (m^{-1})	-0.80 ± 0.33	6.70	41	0.13	0.021
Lapptjärn	Epi	a_{420} (m^{-1})	-1.80 ± 0.57	7.54	41	0.20	0.003
Lillsjöliden	Epi	a_{420} (m^{-1})	1.61 ± 1.88	7.30	35	0.01	0.395
Nedre Björntjärnen	Epi	a_{420} (m^{-1})	2.00 ± 1.22	10.08	59	0.05	0.106
Struptjärnen	Epi	a_{420} (m^{-1})	2.75 ± 2.47	10.81	38	0.04	0.273
Övre Björntjärnen	Epi	a_{420} (m^{-1})	3.04 ± 2.74	11.32	72	0.02	0.270
Stortjärnen	Epi	a_{420} (m^{-1})	4.91 ± 1.04	11.27	31	0.27	0.000
Fisklösan	Epi	DOC (mg L^{-1})	-0.48 ± 0.56	8.18	38	0.02	0.389
Nästjärnen	Epi	DOC (mg L^{-1})	-0.36 ± 0.27	8.76	40	0.04	0.192
Mångstrettjärn	Epi	DOC (mg L^{-1})	-1.22 ± 0.33	13.23	41	0.26	0.001
Lapptjärn	Epi	DOC (mg L^{-1})	-2.68 ± 0.56	14.86	41	0.37	0.000
Lillsjöliden	Epi	DOC (mg L^{-1})	1.88 ± 5.13	15.31	35	0.00	0.717
Nedre Björntjärnen	Epi	DOC (mg L^{-1})	1.87 ± 3.00	18.31	59	0.01	0.536
Struptjärnen	Epi	DOC (mg L^{-1})	4.71 ± 4.63	19.85	38	0.03	0.317
Övre Björntjärnen	Epi	DOC (mg L^{-1})	2.55 ± 6.55	21.39	72	0.00	0.698
Stortjärnen	Epi	DOC (mg L^{-1})	4.77 ± 2.11	18.97	31	0.15	0.031

Fisklösan	Hypo	$a_{254} : a_{365}$	0.11 ± 0.21	4.13	28	0.01	0.624
Nästjärnen	Hypo	$a_{254} : a_{365}$	0.04 ± 0.09	3.68	30	0.01	0.675
Mångstrettjärn	Hypo	$a_{254} : a_{365}$	-0.04 ± 0.10	3.84	31	0.00	0.732
Lapptjärn	Hypo	$a_{254} : a_{365}$	-0.01 ± 0.15	3.85	31	0.00	0.926
Lillsjöliden	Hypo	$a_{254} : a_{365}$	0.05 ± 0.20	3.85	33	0.00	0.799
Nedre Björntjärnen	Hypo	$a_{254} : a_{365}$	-0.23 ± 0.13	4.06	29	0.10	0.087
Struptjärnen	Hypo	$a_{254} : a_{365}$	-0.01 ± 0.17	3.83	29	0.00	0.934
Övre Björntjärnen	Hypo	$a_{254} : a_{365}$	-0.26 ± 0.12	4.02	45	0.10	0.031
Stortjärnen	Hypo	$a_{254} : a_{365}$	-0.62 ± 0.10	4.07	29	0.58	0.000
Fisklösan	Hypo	a_{420} : DOC	0.02 ± 0.06	0.39	28	0.00	0.776
Nästjärnen	Hypo	a_{420} : DOC	0.03 ± 0.04	0.46	30	0.01	0.538
Mångstrettjärn	Hypo	a_{420} : DOC	0.11 ± 0.05	0.40	31	0.14	0.037
Lapptjärn	Hypo	a_{420} : DOC	0.09 ± 0.07	0.54	31	0.05	0.249
Lillsjöliden	Hypo	a_{420} : DOC	-0.06 ± 0.07	0.56	33	0.02	0.424
Nedre Björntjärnen	Hypo	a_{420} : DOC	0.19 ± 0.10	0.49	29	0.11	0.072
Struptjärnen	Hypo	a_{420} : DOC	-0.02 ± 0.07	0.62	29	0.00	0.776
Övre Björntjärnen	Hypo	a_{420} : DOC	0.17 ± 0.05	0.50	45	0.20	0.002
Stortjärnen	Hypo	a_{420} : DOC	0.20 ± 0.05	0.54	29	0.42	0.000
Fisklösan	Hypo	a_{420} (m^{-1})	-0.16 ± 0.53	3.26	28	0.00	0.770
Nästjärnen	Hypo	a_{420} (m^{-1})	-0.42 ± 0.56	7.11	30	0.02	0.453
Mångstrettjärn	Hypo	a_{420} (m^{-1})	0.4 ± 0.97	7.48	31	0.01	0.679
Lapptjärn	Hypo	a_{420} (m^{-1})	-0.21 ± 1.51	8.89	31	0.00	0.889
Lillsjöliden	Hypo	a_{420} (m^{-1})	-1.04 ± 0.84	9.79	33	0.05	0.227
Nedre Björntjärnen	Hypo	a_{420} (m^{-1})	3.33 ± 1.37	9.67	29	0.18	0.022
Struptjärnen	Hypo	a_{420} (m^{-1})	3.12 ± 3.25	13.98	29	0.03	0.345
Övre Björntjärnen	Hypo	a_{420} (m^{-1})	3.83 ± 1.12	10.81	45	0.21	0.001
Stortjärnen	Hypo	a_{420} (m^{-1})	17.7 ± 2.57	5.40	29	0.64	0.000
Fisklösan	Hypo	DOC (mg L^{-1})	-0.16 ± 0.53	8.25	28	0.10	0.110
Nästjärnen	Hypo	DOC (mg L^{-1})	-0.42 ± 0.56	13.95	30	0.06	0.205
Mångstrettjärn	Hypo	DOC (mg L^{-1})	0.4 ± 0.97	16.74	31	0.13	0.044
Lapptjärn	Hypo	DOC (mg L^{-1})	-0.21 ± 1.51	15.67	31	0.14	0.075
Lillsjöliden	Hypo	DOC (mg L^{-1})	-1.04 ± 0.84	17.28	33	0.00	0.831
Nedre Björntjärnen	Hypo	DOC (mg L^{-1})	3.33 ± 1.37	19.40	29	0.00	0.747
Struptjärnen	Hypo	DOC (mg L^{-1})	3.12 ± 3.25	22.55	29	0.08	0.132
Övre Björntjärnen	Hypo	DOC (mg L^{-1})	3.83 ± 1.12	21.60	45	0.00	0.888
Stortjärnen	Hypo	DOC (mg L^{-1})	17.7 ± 2.57	12.19	29	0.56	0.000

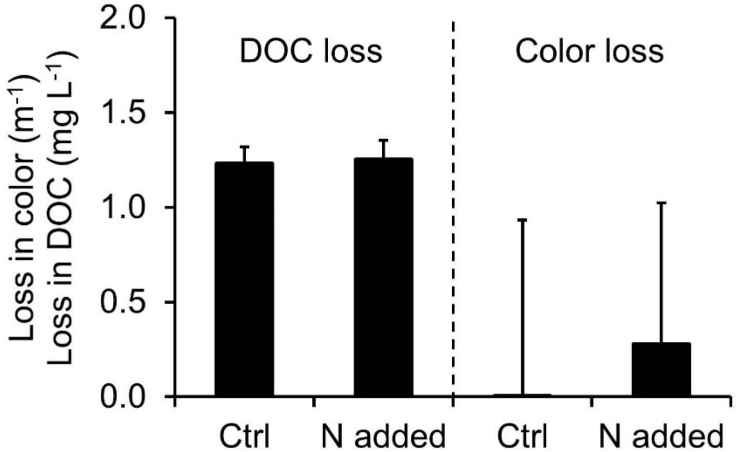

Figure S1. Losses in dissolved organic carbon (DOC) and in color (absorbance at 420 nm) during 2-week dark bioassays in 20 °C performed on ambient and nitrogen-amended water from Nedre Björntjärnen (four replicate dates representing different seasons). If N was limiting the DOC processing rates *in situ*, there should have been an impact by N addition also in these short-term laboratory experiments. Bars and error bars show mean + standard error of four dates prior to the whole-lake nutrient manipulation (see methods in main paper for more information).

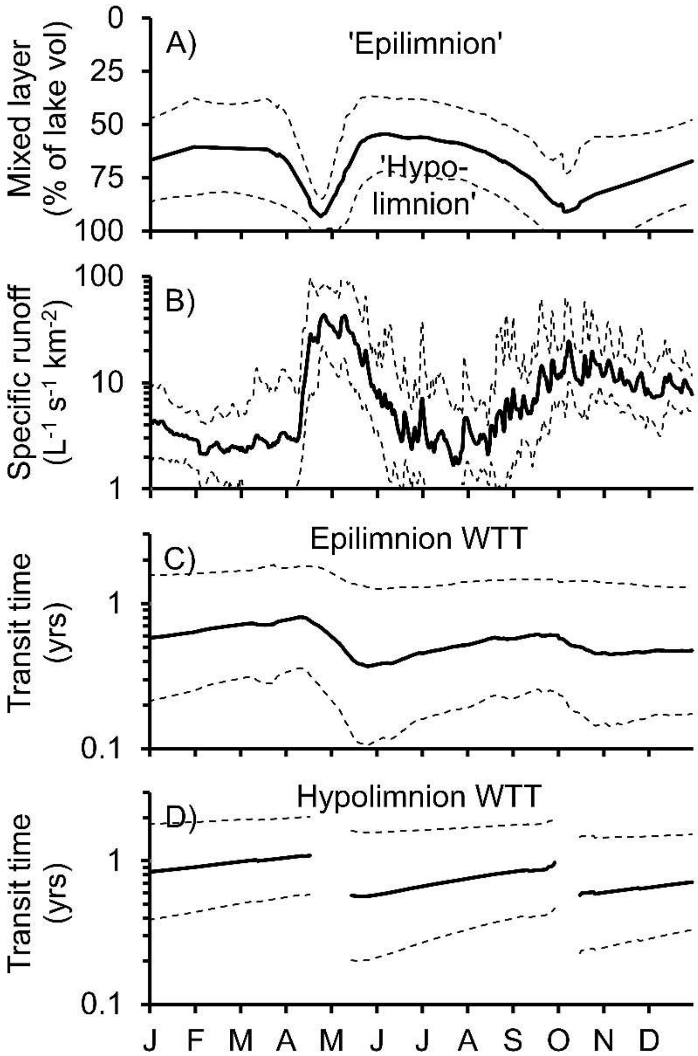

Figure S2. Means (solid lines) ± 1 *SD* (dotted lines) of hydrological variables describing (A) lake mixing, (B) catchment discharge and (C-D) water transit time (WTT) as functions of time of year (month of year). Note that B-D show mean and *SD* derived from the log-normal distribution of y-axis values. The variables were assessed separately for each lake and study year, but for illustrative purposes the figure shows the average of 38 linearly interpolated curves obtained from 9 lakes in northern Sweden during 3-7 years of measurement per lake. The hypolimnetic WTT (in D) is only shown for periods during which the hypolimnion was typically present.

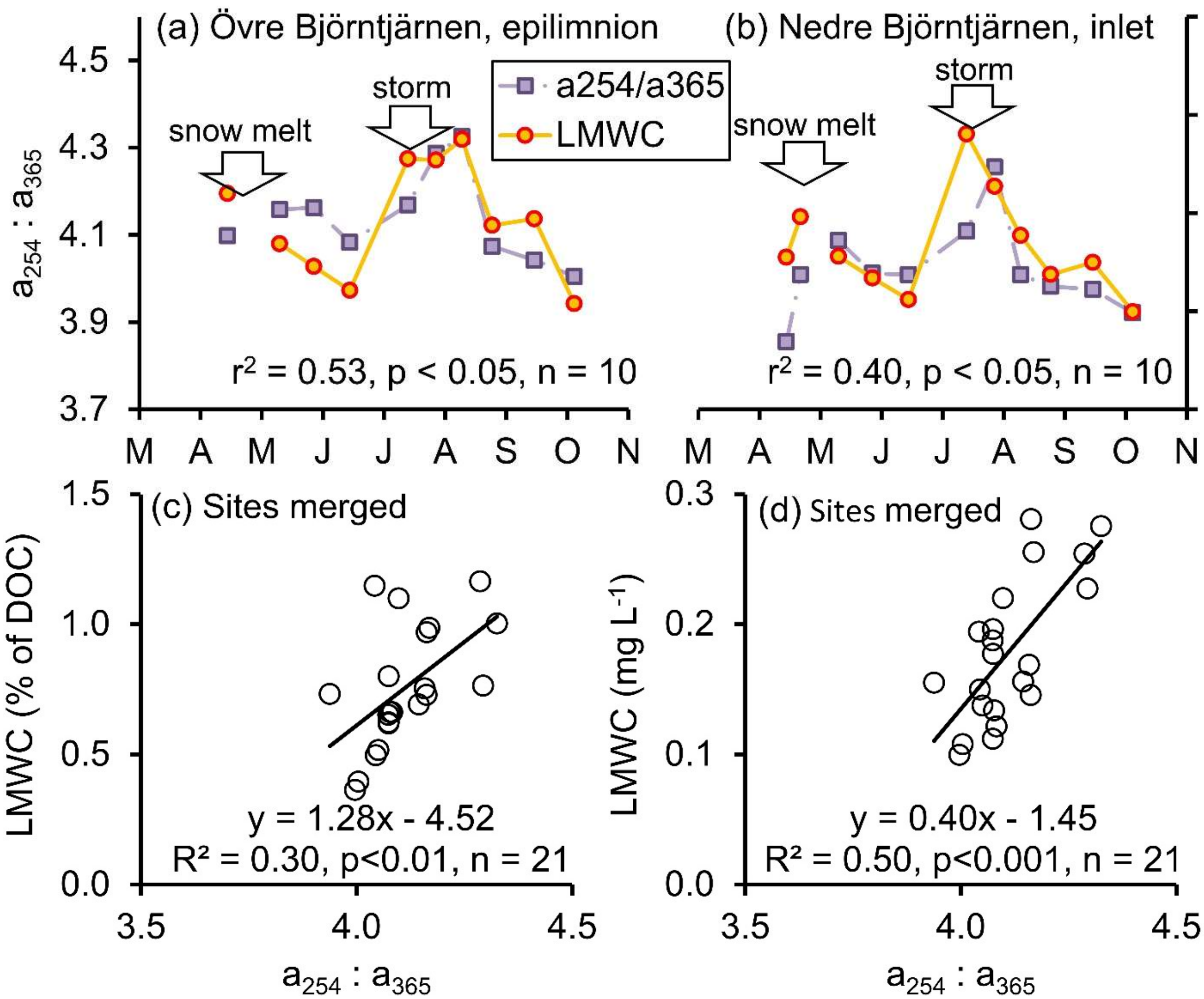

Figure S3. Comparisons between absorbance ratio a_{254} : a_{365} and the measured concentrations of low molecular weight carbon compounds (LMWC). (a-b) Timeline plots for a_{254} : a_{365} and absolute LMWC concentrations shown for two sites in the Björntjärnarna catchment (a and b, respectively) from April to October 2009. Significant correlations between the two variables are indicated by the statistics below each curve pair. (c-d) Linear relationships between a_{254} : a_{365} and (c) relative or (d) absolute LMWC concentrations in the pooled dataset of all observations. The LMWC represents a sum of 39 of the most common organic acids, free amino acids and simple carbohydrates. Note that panels a-b share the same y axes.

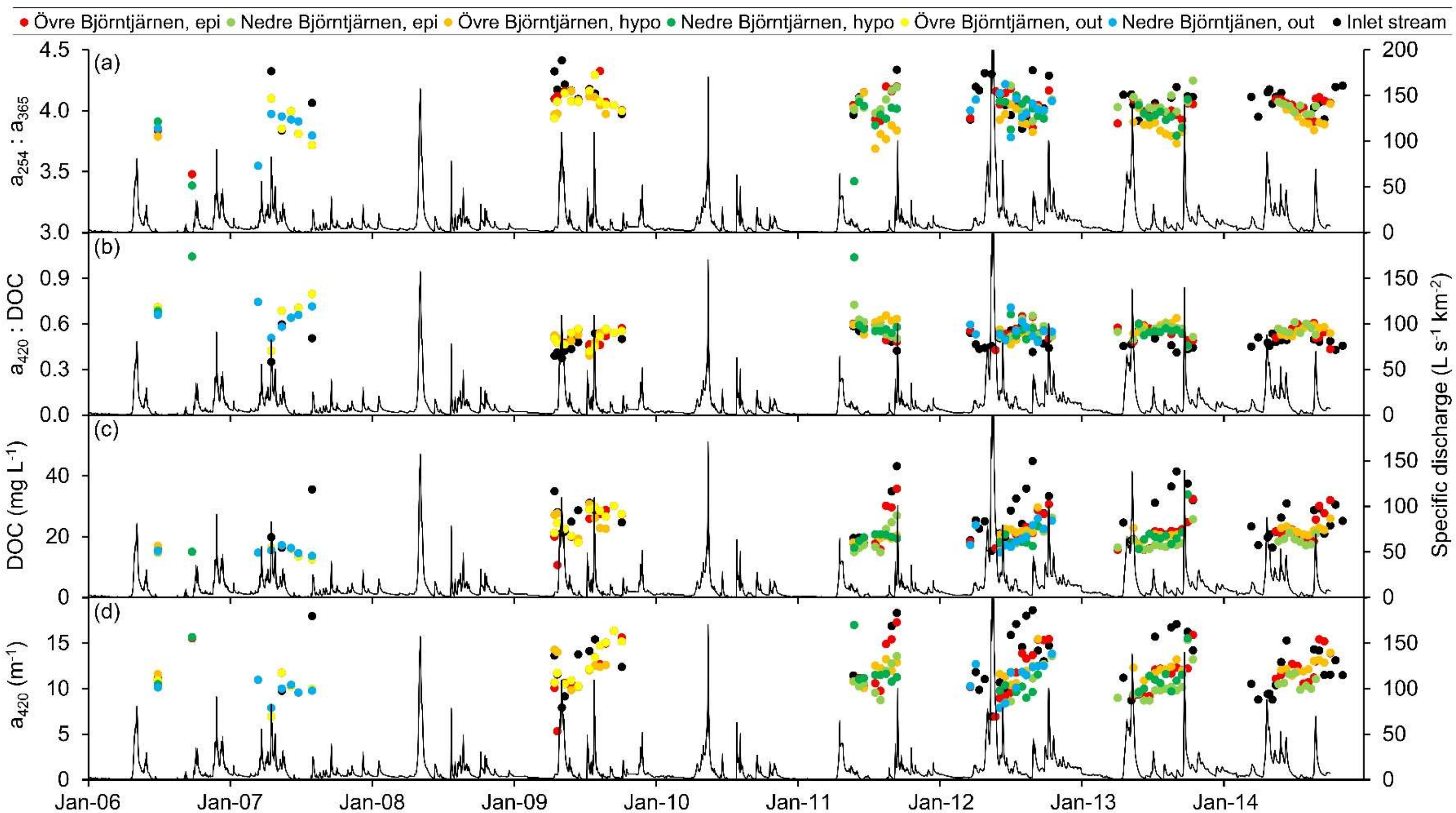

Figure S4. Discharge (line, right y axis) and dissolved organic carbon properties (symbols, left y axis) plotted over Gregorian calendar time for the Björntjärnarna catchment in northern Sweden. The primary y-axis variables are (A) ratio between the absorbance at the wavelengths of 254 nm and 365 nm, (B) ratio between absorbance at 420 nm and DOC, (C) dissolved organic carbon and (D) absorbance at 420 nm (n = 260, study years 2006-2014).

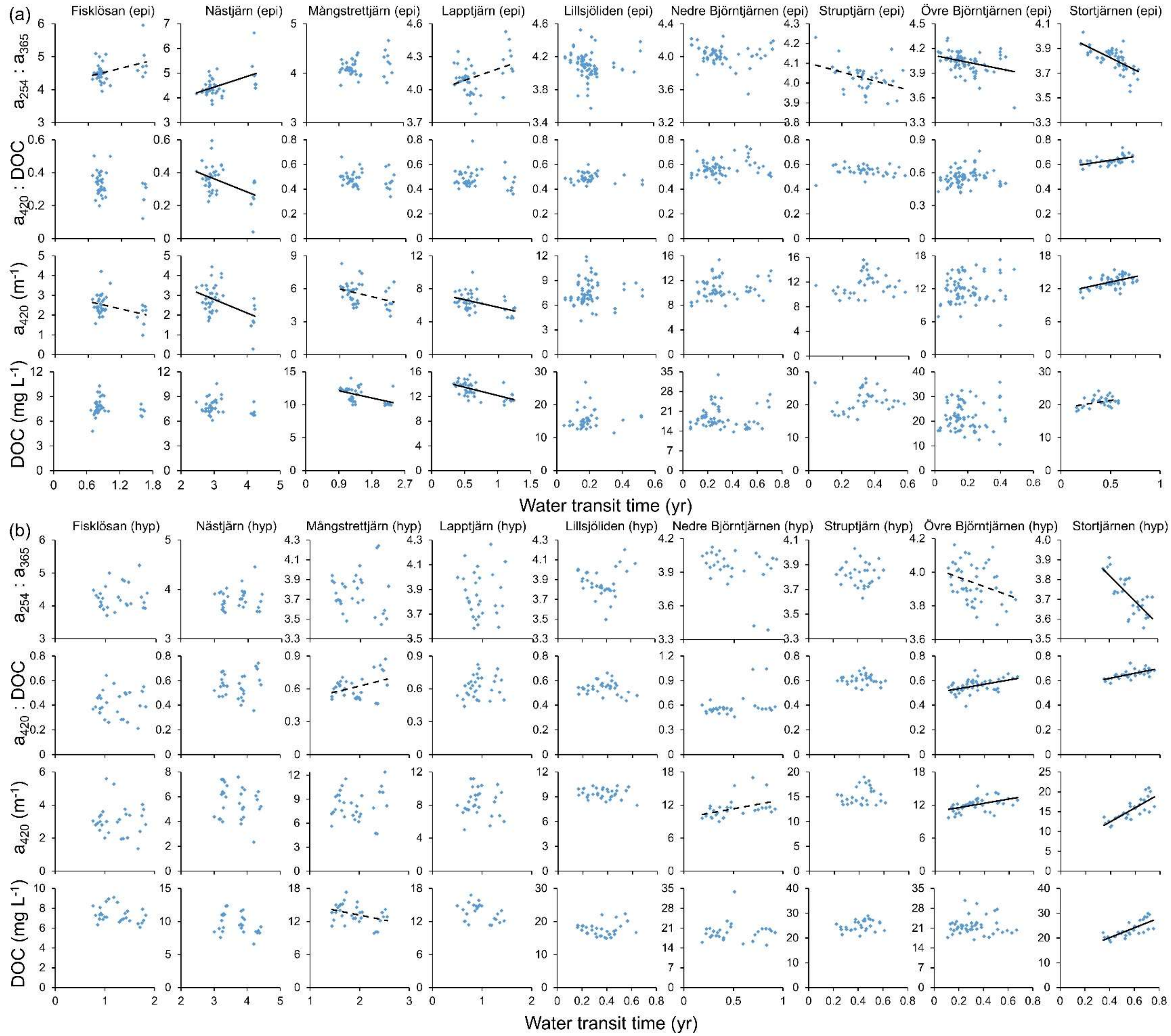
(a)
Fisklösan (epi)
Nästjärn (epi)
Mångstrettjärn (epi)
Lapptjärn (epi)
Lillsjöliden (epi)
Nedre Björntjärnen (epi)
Struptjärn (epi)
Övre Björntjärnen (epi)
Stortjärnen (epi)
a_{254} : a_{365}
a_{420} : DOC
a_{420} (m^{-1})
DOC (mg L^{-1})
Water transit time (yr)
(b)
Fisklösan (hyp)
Nästjärn (hyp)
Mångstrettjärn (hyp)
Lapptjärn (hyp)
Lillsjöliden (hyp)
Nedre Björntjärnen (hyp)
Struptjärn (hyp)
Övre Björntjärnen (hyp)
Stortjärnen (hyp)
a_{254} : a_{365}
a_{420} : DOC
a_{420} (m^{-1})
DOC (mg L^{-1})
Water transit time (yr)

Figure S5 (previous page). Raw data plots for linear regressions* detailed in Table S1 and used in Figs. 2-3 of the main paper. (a) The upper half of figure shows epilimnetic data ('epi' sites) and (b) the lower half shows hypolimnetic data ('hyp' sites).
*Significance: solid line, $p < 0.01$; dashed line, $p < 0.05$. See table S1 for details.