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Contrasting seasonal patterns in particle aggregation and dissolved organic matter transformation in a sub-Arctic fjord

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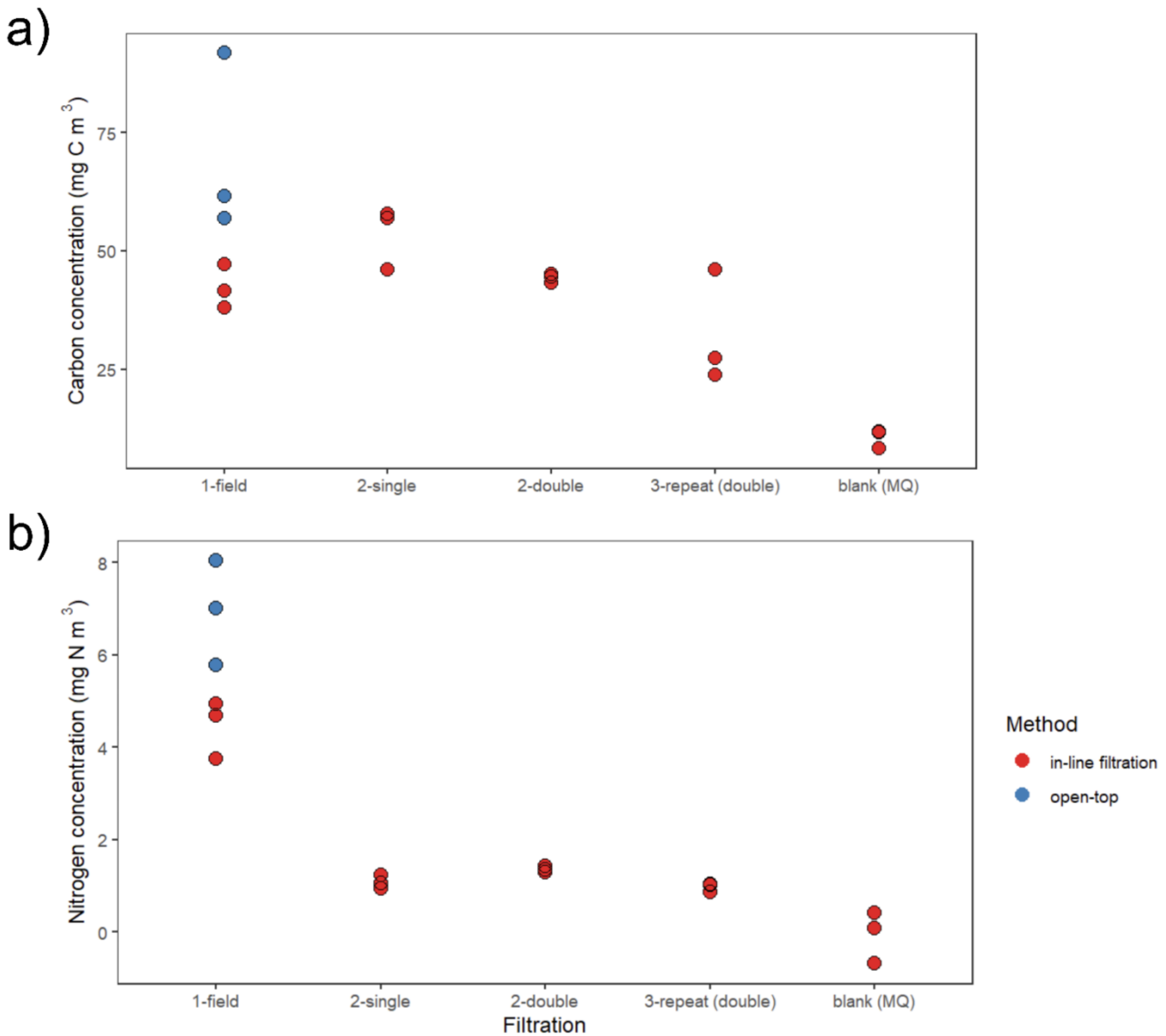

Figure S1: Results from the double- and single-layer filtration test in January. The effect of filtration through a single or a double layer of GF/F filters on particulate organic carbon (POC) and particulate nitrogen (PN) concentrations in filtered water was tested. The filtration steps are shown as numbers 1-3. POC concentrations in the field in January are shown on the leftmost side (1-field), followed by the concentrations measured on POC samples taken after filtration through a single GF/F filter (2-single; Whatman, 147mm diameter), and the concentrations measured after filtration through a double layer of filters (2-double). The collected filtrate from the double layer was filtered and measured once again (3-repeat). Blank POC concentrations of MilliQ are shown on the rightmost side.

Description of results and discussion: POC concentrations in the filtrate of any of the filtration methods did not reach blank (MilliQ) concentrations. POC concentrations gradually decreased from filtrations through a single filter, a double filter and a refiltration through the double filter; however, variation of the three replicates were large in single-filter filtrations and the repeated double-filter filtrations. We concluded that filtration through a double filter mainly decreased the variation of replicates and therefore we used double filters for the experiment from February onwards. In-line filtrations revealed lower POC concentrations compared to open-top filtrations; possibly because the risk of contamination is higher for open-top filtration. Another possibility for the discrepancy is that during open-top filtration, the water is poured into the funnel which can lead to additional aggregation, while during in-line filtration, the water was gently sucked through peristaltic tubing. Interestingly, POC concentrations of the filtrate from the filtration through the single filter were slightly higher compared to field values of POC measured after in-line filtration, in accordance with our results in the experiment of relatively higher initial (t0) POC concentrations of filtered water during winter compared to the other months. No further decrease of PN concentrations was measured in the filtrate for any filtration method; however, similar to POC, in-line filtration revealed lower field levels of PN compared to open-top filtration.

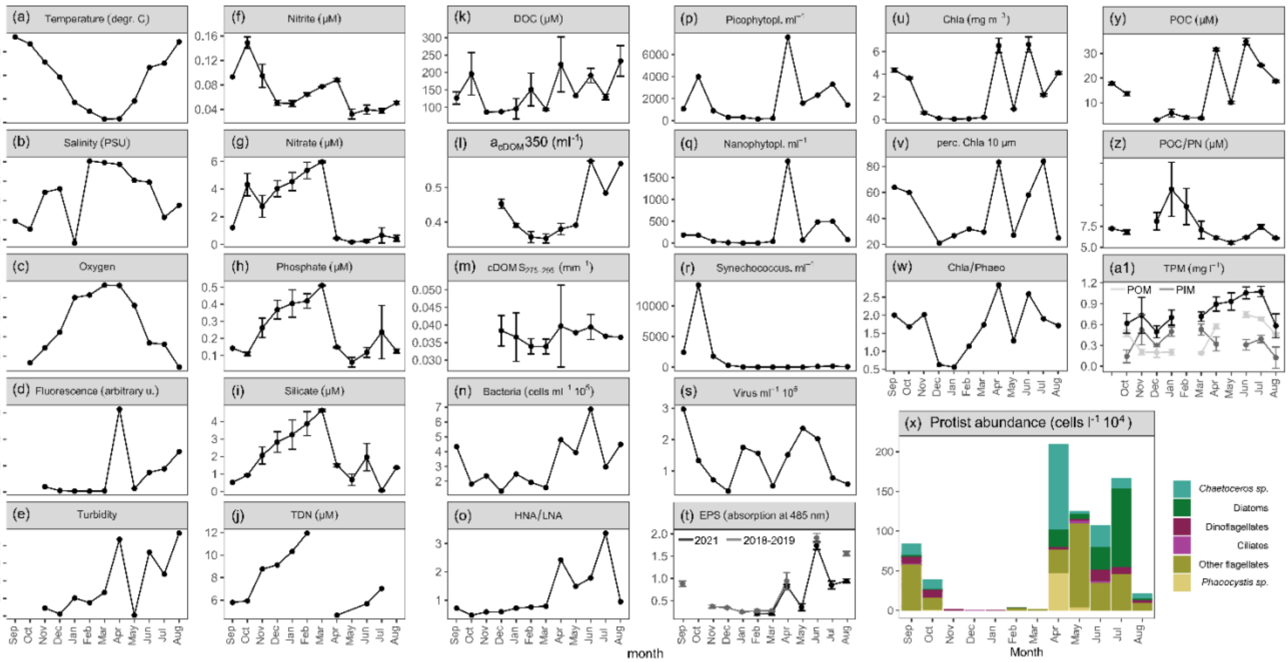

Figure S2: Mean values and standard deviations (if available; n= 2-3) of biogeochemical parameters measured on a monthly basis in Ramfjorden, Tromsø. Abbreviations are j) total dissolved nitrogen (TDN), k) dissolved organic carbon (DOC), l-m) colored dissolved organic matter (CDOM), o) ratio of high nucleic to low nucleic acid (HNA/LNA), p) Picophytoplankton (Picophytopl.), q) Nanophytoplankton (Nanophytopl.), t) extracellular polymeric substances (EPS), u) Chlorophyll-a (Chl-a), v) percentage of Chl-a > 10µM (perc. Chl-a 10µm), w) Chl-a to Phaeopigment ratio (Chl-a/Phaeo), y) particulate organic carbon (POC), z) particulate organic carbon to nitrogen ratio (POC/PN), a1) total particulate matter (TPM; black line), particulate inorganic matter (PIM; dark gray line), particulate organic matter (POM; light gray line). EPS was sampled in 2018 (dark gray line) and 2020-2021 (black line), respectively.

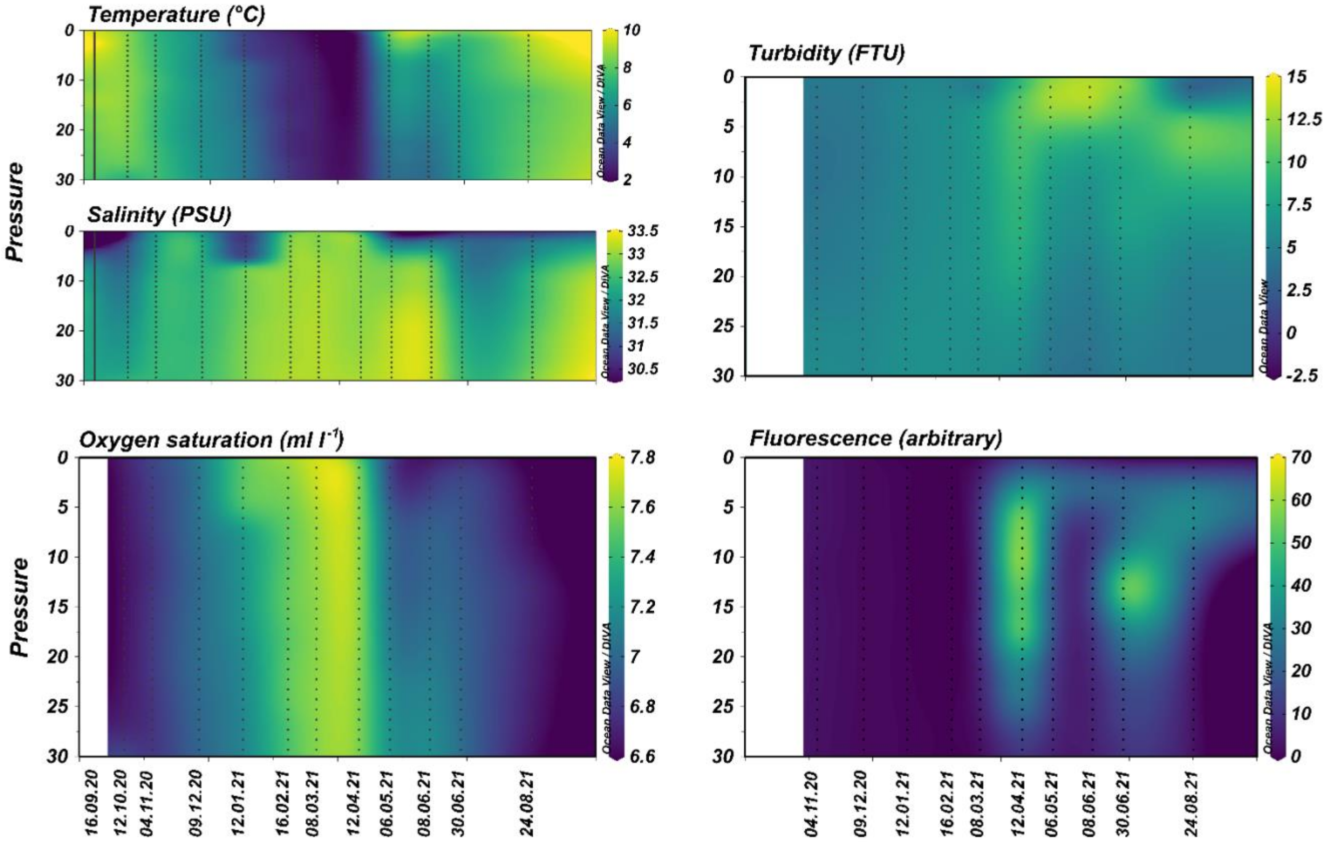

Figure S3: DIVA-interpolated monthly CTD measurements. Seasonal changes of temperature, salinity, oxygen saturation, turbidity and fluorescence between September 2020 and August 2021 of the upper 30m in the mouth of Ramfjorden, Tromsø. The dotted lines show sampling events.

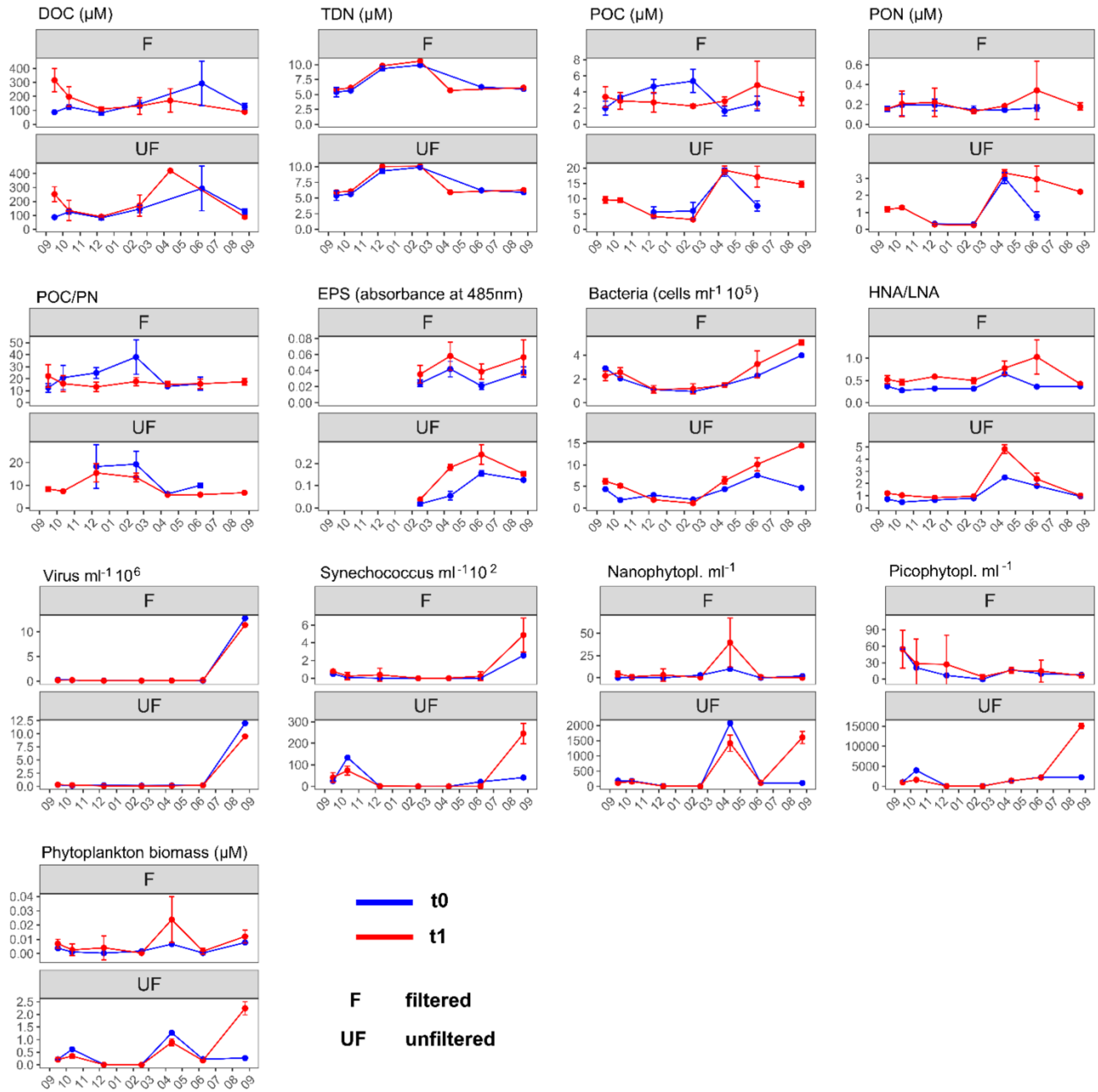

Figure S4: Absolute mean values and standard deviations (if applicable; n= 2 - 5) measured at the start of incubation (t0, blue line) and at the end of incubation (t1, red line) during each month. Dissolved organic carbon (DOC), total dissolved nitrogen (TDN), particulate organic carbon (POC) and particulate nitrogen (PN) concentrations; ratio of POC/PN; relative concentrations of extracellular polymeric substances (EPS; relative absorption at 485nm); high DNA (HNA) and low DNA (LNA) bacterial abundances, and relative bacterial activity (HNA/LNA ratio) for filtered (F) and unfiltered (UF) treatments.

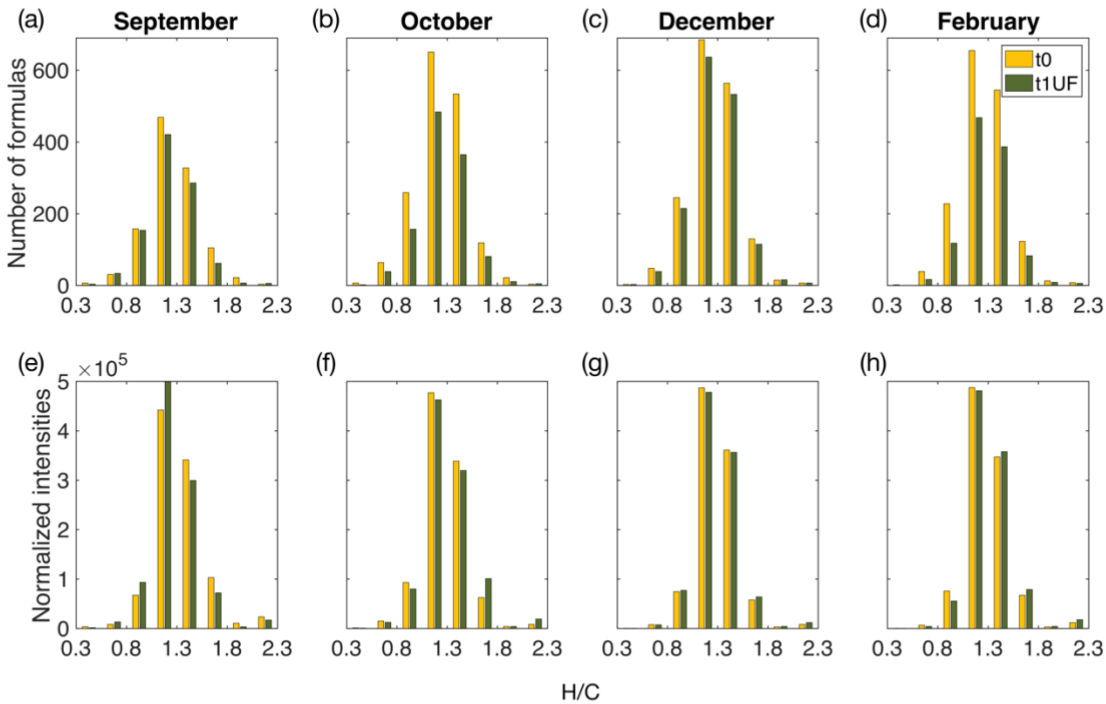

Figure S5: Histograms of all identified organic matter formulas and normalized intensities determined by high-resolution mass spectrometry during start (t0) and end (t1) of incubations of UF treatment water. Number of identified molecular formulas are plotted according to the hydrogen to carbon (H/C) atomic ratio for incubation experiments in a) September, b) October c) December and d) February. Normalized intensities (e-h) are plotted according to H/C ratios for respective months. The start of the incubation (t0) is shown in yellow and the end of the incubation (t1) in green for UF treatment.

UF treatment was used to test the effects of the seasonal biological community and particles, in the 0.7 – 90 µm size range, on DOM composition during the incubation time. UF treatment refers to unfiltered water for the duration of the 36 h incubation and was filtered immediately prior to collection at t1. More details of experiment set up can be found in the methods section. September shows a decrease in formulas and intensities for formulas with higher H/C ratios. Winter period (December and February) show a decrease in formulas in the low to mid H/C range (0.5 – 1.6). Results are similar to F treatment where the productive period had a decrease in formulas of higher hydrogen saturation. Winter period for both F and UF treatment shows a decrease in formulas from lower hydrogen saturation (< 1.6). October indicates a transition period with loss of formulas across H/C ratios in both treatments.

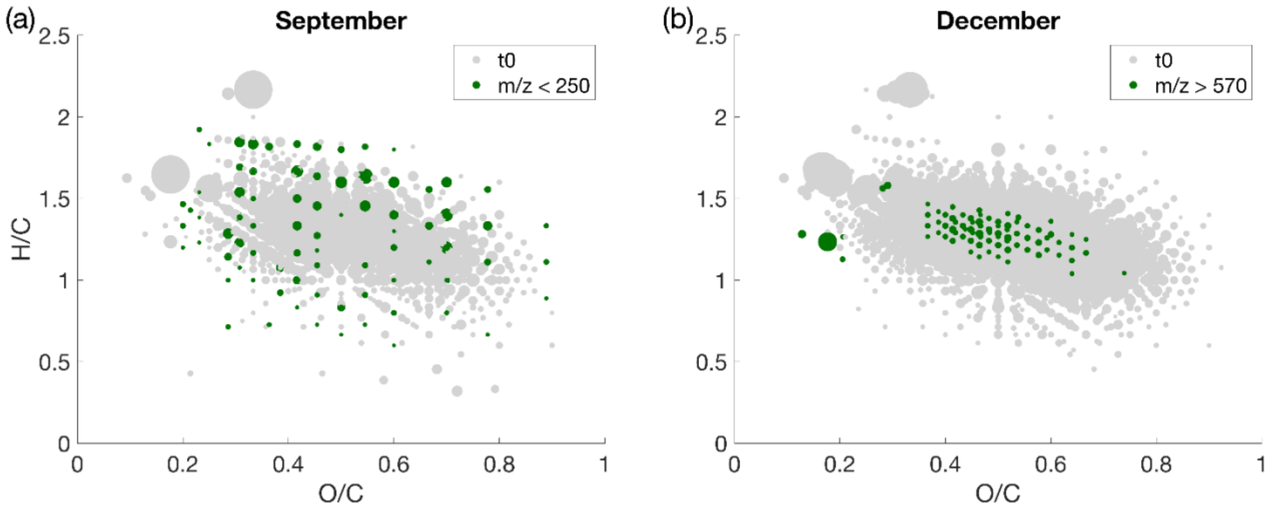

Figure S6: van Krevelen diagram of organic matter characterization of molecular weight groups at t0. Each point is an identified molecular formula (intensity shown as size of point) and plotted according to the oxygen to carbon (O/C) and hydrogen to carbon (H/C) atomic ratio of the molecular formula. The start of incubation (t0) is shown in grey for a) September and with molecular formulas less than 250 mass to charge (m/z) highlighted in green. The start of incubation (t0) for b) December are shown in grey and molecular formulas greater than 570 (m/z) are highlighted in green. Results show that low molecular weight formulas (< 250) in September have higher intensities at high H/C saturation. Molecular weight formulas > 570 m/z in December are in the middle O/C and H/C region of the van Krevelen diagram.

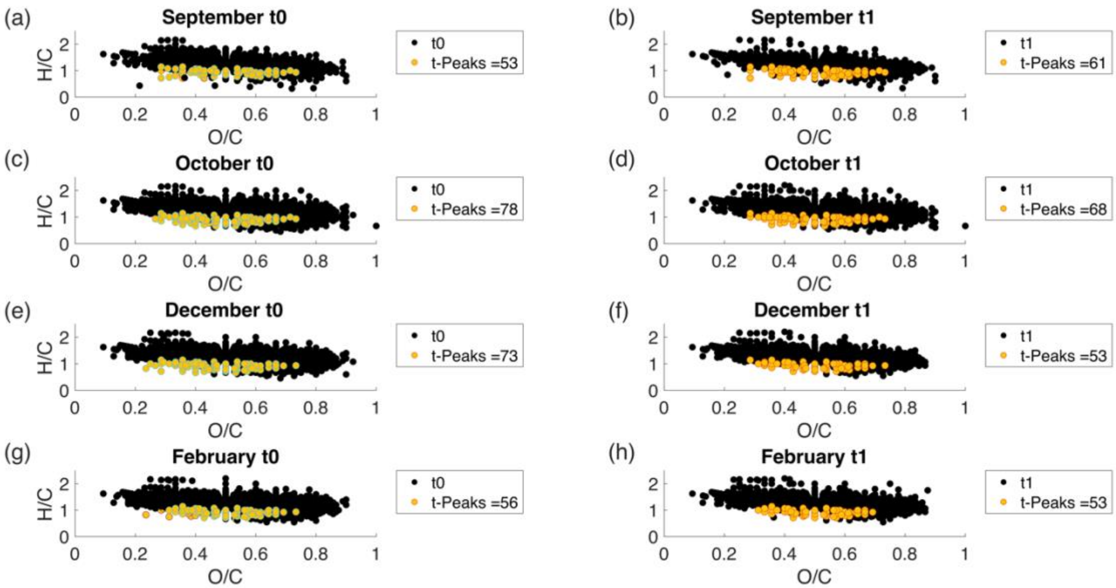

Figure S7: van Krevelen diagram of t-Peaks during experiment months. Each point is an identified molecular formula and plotted according to the oxygen to carbon (O/C) and hydrogen to carbon (H/C) atomic ratio of the molecular formula. start of incubation (t0) for each month are shown in black and t-Peak molecular formulas present are highlighted in orange (a,c,e,g). End of incubation (t1) (F treatment) for each month are shown in black and t-Peak molecular formulas matches highlighted are in orange (b,d,f,h). T-peak molecular formulas are obtained from Medeiros et al. (2016). Results show significant decrease of t-Peaks in February (t-test, $p = 0.04$, t-value = 2.96, df = 4) and October (t-test, $p = 0.04$, t-value = 3.02, df = 4)incubations and no significant change in t-peaks during September (t-test, $p = 0.08$, t-value = -2.31, df = 4) and December (t-test, $p = 0.09$, t-value = 2.21, df = 4) incubations

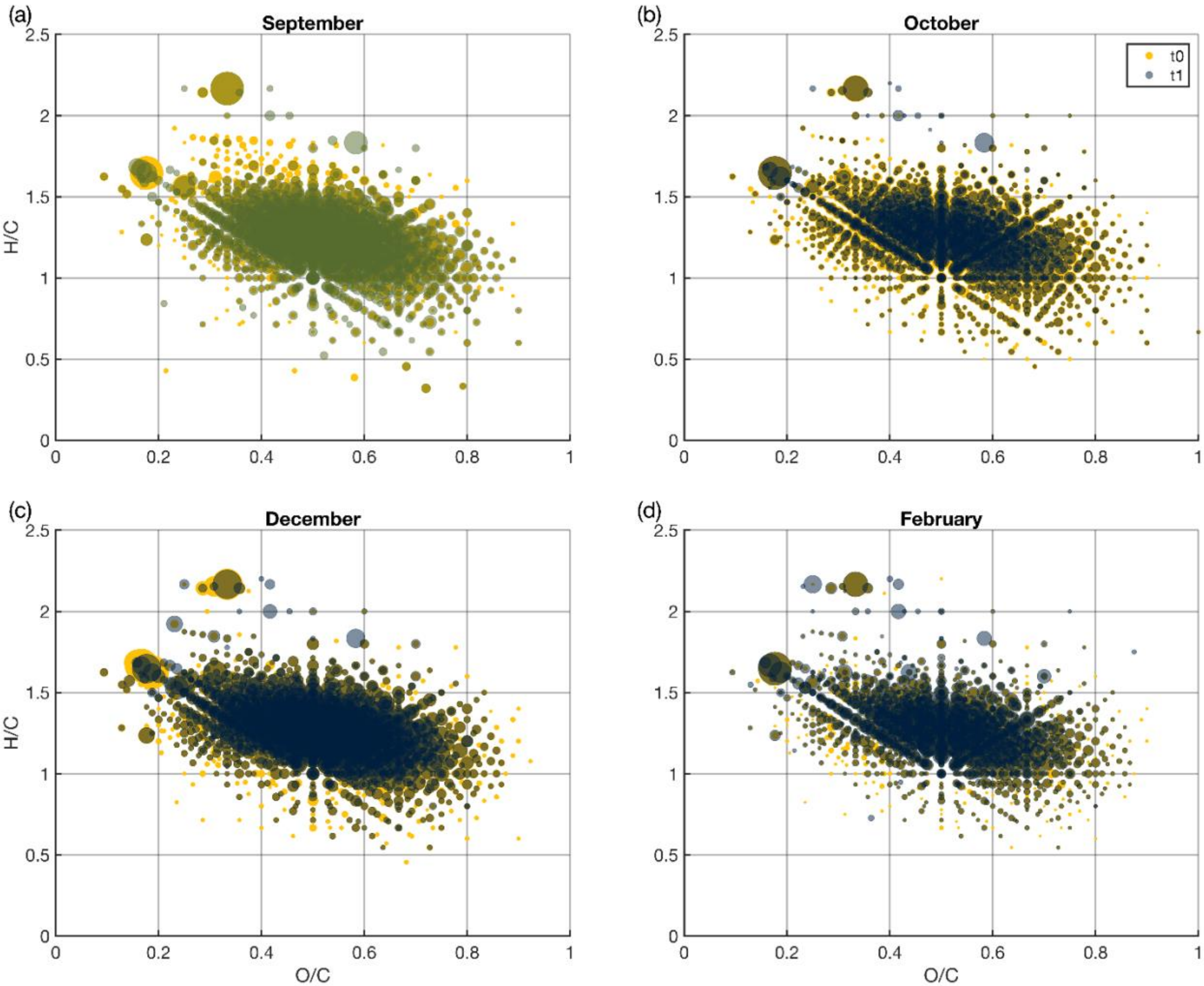

Figure S8: van Krevelen diagram of organic matter characterization determined by high-resolution mass spectrometry for start (t0) and end (t1) of (F treatment) incubations. Each point is an identified molecular formula (intensity shown as size of point) and plotted according to the oxygen to carbon (O/C) and hydrogen to carbon (H/C) atomic ratio of molecular formula for incubations in a) September, b) December and c) February. Molecular DOM composition at the start of the incubation (t0) is shown in yellow and at the end of the incubation (t1) in green or blue for F treatment.

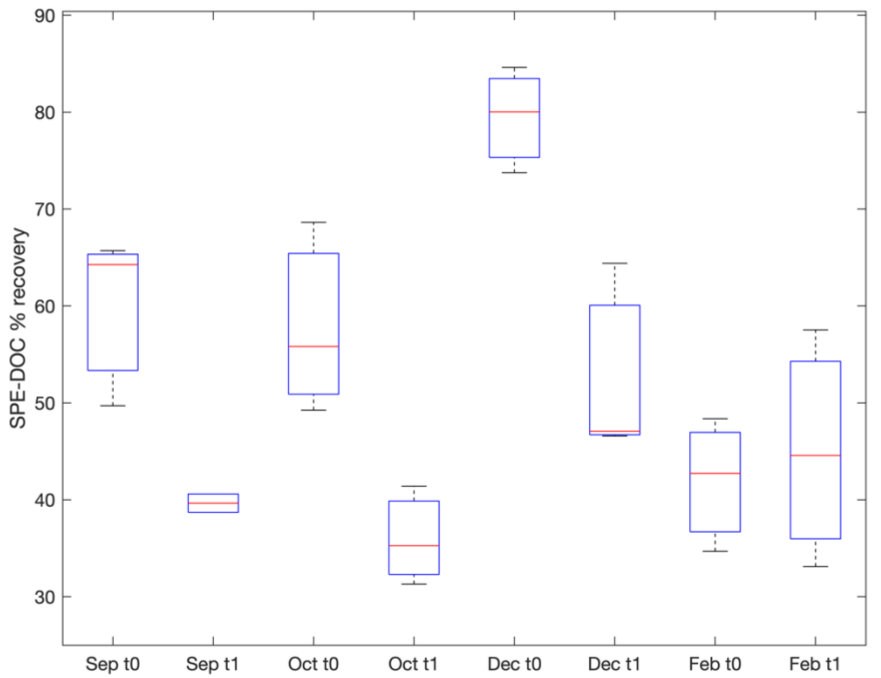

Figure S9: SPE-DOC % recovery for experiment at the start (t0) and end of incubations (t1) for September, October, December and February (n = 3). The highest SPE-DOC recovery is observed in December at the start of incubation and lowest recoveries at the end of incubations in September and October.

Table S1: POC concentrations and standard deviations (if applicable; n= 2 - 5) in µM measured in the field (in situ), at the start (t0) and at the end (t1) of incubation during each month. Fjord water collected from the field was filtered using a 0.7 µm GFF filter. For the incubation treatments, fjord water was sieved through a 90 µm mesh and either filtered (F, 0.7 µm GFF) or left unfiltered (UF).

Samling month	Field	t0 (F)	t1 (F)	t0 (UF)	t1 (UF)
September	17.82 ± 0.6	2 ± 0.89	3.41 ± 1.26	n.a.	9.65 ± 1.11
October	13.62 ± 0.85	3.35 ± 0.18	2.91 ± 1.03	n.a.	9.45 ± 0.76
December	3.08 ± 0.09	4.7 ± 0.89	2.72 ± 1.2	5.58 ± 1.78	4.21 ± 0.23
January	5.84 ± 1.58				
February	4.02 ± 0.74	5.36 ± 1.44	2.26 ± 0.21	6.05 ± 2.81	3.21 ± 0.29
March	3.88 ± 0.39				
April	31.58 ± 0.59	1.64 ± 0.59	2.88 ± 0.53	18.57 ± 1.34	19.3 ± 1.32
May	10.16 ± 0.5				
June	34.87 ± 1.33	2.59 ± 0.9	4.87 ± 2.96	7.65 ± 1.71	17.14 ± 3.43

July	25.21 ± 0.2				
August	18.8 ± 0.45	n.a.	n.a.	n.a.	n.a.

Table S2: High resolution mass spectrometry results of DOM variables showing intensity weighted means of hydrogen to carbon (H/C_{wa}), oxygen to carbon (O/C_{wa}), molecular weight (MW_{wa}) and modified aromaticity index ($AI_{mod\ wa}$). The weighted standard deviation (SD_w, Equation 3) and the standard error of the weighted mean (SEM, Equation 4) are computed for each sample using the DOM metrics associated with the molecular formulas within that sample. The variability in metric values among the many identified formulas is reflected in the standard deviations. The benefit of the large number of formulas for each treatment is the high certainty in the mean which is also shown by the low standard error of mean.

Treatment	H/C_{wa} (SD_w)(SEM)	O/C_{wa}(SD_w)(SEM)	MW_{wa}(SD_w)(SEM)	$AI_{mod\ wa}$(SD_w)(SEM)
Sep t0-1	1.31 (0.24) (0.008)	0.50 (0.13) (0.004)	360.35 (74) (2)	0.21 (0.12) (0.004)
Sep t0-2	1.30 (0.24) (0.008)	0.50 (0.14) (0.005)	366.39 (74) (3)	0.21 (0.12) (0.004)
Sep t0-3	1.33 (0.25) (0.008)	0.48 (0.14) (0.005)	357.46 (76) (3)	0.21 (0.12) (0.004)
Sep t1-1	1.25 (0.21) (0.007)	0.52 (0.12) (0.004)	365.09 (68) (2)	0.24 (0.12) (0.004)
Sep t1-2	1.27 (0.22) (0.007)	0.50 (0.13) (0.004)	364.16 (68) (2)	0.23 (0.12) (0.004)
Sep t1-3	1.27 (0.23) (0.007)	0.50 (0.15) (0.005)	359.22 (66) (2)	0.23 (0.12) (0.004)
Oct t0-1	1.27 (0.22) (0.005)	0.51 (0.14) (0.003)	364.70 (71) (2)	0.22 (0.12) (0.003)
Oct t0-2	1.26 (0.21) (0.006)	0.51 (0.13) (0.004)	363.85 (70) (2)	0.23 (0.12) (0.003)
Oct t0-3	1.26 (0.21) (0.005)	0.51 (0.13) (0.003)	365.04 (72) (2)	0.23 (0.12) (0.003)
Oct t1-1	1.27 (0.22) (0.006)	0.51 (0.14) (0.004)	361.95 (70) (2)	0.22 (0.12) (0.003)
Oct t1-2	1.30 (0.23) (0.007)	0.50 (0.15) (0.004)	358.08 (67) (2)	0.22 (0.12) (0.003)
Oct t1-3	1.28 (0.23) (0.006)	0.51 (0.14) (0.004)	360.13 (68) (2)	0.22 (0.12) (0.003)
Dec t0-1	1.28 (0.23) (0.006)	0.51 (0.14) (0.004)	360.13 (68) (2)	0.22 (0.12) (0.003)
Dec t0-2	1.28 (0.20) (0.005)	0.51 (0.13) (0.004)	368.64 (70) (2)	0.22 (0.11) (0.003)
Dec t0-3	1.28 (0.20) (0.005)	0.51 (0.13) (0.003)	368.70 (71) (2)	0.22 (0.11) (0.003)
Dec t1-1	1.27 (0.19) (0.005)	0.51 (0.13) (0.003)	372.14 (72) (2)	0.22 (0.11) (0.003)
Dec t1-2	1.29 (0.20) (0.006)	0.51 (0.13) (0.004)	366.95 (68) (2)	0.22 (0.11) (0.003)
Dec t1-3	1.30 (0.21) (0.006)	0.50 (0.14) (0.004)	366.74 (69) (2)	0.21 (0.11) (0.003)
Feb t0-1	1.29 (0.21) (0.006)	0.50 (0.14) (0.004)	365.64 (69) (2)	0.22 (0.11) (0.003)
Feb t0-2	1.28 (0.21) (0.005)	0.51 (0.14) (0.004)	365.38 (69) (2)	0.22 (0.11) (0.003)
Feb t0-3	1.28 (0.20) (0.005)	0.50 (0.13) (0.003)	366.26 (69) (2)	0.22 (0.11) (0.003)
Feb t1-1	1.28 (0.20) (0.006)	0.52 (0.14) (0.004)	366.52 (68) (2)	0.21 (0.11) (0.003)
Feb t1-2	1.30 (0.22) (0.006)	0.50 (0.13) (0.004)	363.38 (68) (2)	0.21 (0.11) (0.003)
Dec t1-3	1.31 (0.23) (0.007)	0.50 (0.14) (0.004)	363.08 (67) (2)	0.21 (0.11) (0.003)