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Microbial Processing of Sediment-Derived Dissolved Organic Matter: Implications for Its Subsequent Biogeochemical Cycling in Overlying Seawater

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Key Points:
- Coastal sediment pore water contains a large fraction of labile dissolved organic matter (DOM)
- Resuspension of coastal sediments would weaken their role as a net sink of carbon
- Labile sediment pore water DOM can be transformed into long-lived recalcitrant DOM by successive microbial communities in overlying seawater

Supporting Information:
- Supporting Information

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Abstract Coastal sediments contain a large amount of dissolved organic matter (DOM), which can be mobilized into the overlying water by natural and anthropogenic activities. The bioavailability and subsequent biogeochemical effects of this sediment-derived DOM are unclear. To investigate those, we collected a sediment pore-water DOM (SDOM) sample and its overlying seawater to conduct a bioassay experiment, which allowed tracking of both short-term and long-term microbial processes in the context of DOM transformations. Short-term incubation results show that the SDOM extract supported the growth of specific taxa. The microbial community composition changed dramatically and an approximately 50% of SDOM-derived carbon was consumed within the first 2 days. Viruses likely played a role in promoting bacterial community succession, further enhancing transformation of this SDOM. Long-term incubation results show that labile DOM was gradually consumed, while approximately 16% of the initial SDOM appeared to be recalcitrant to microbial utilization and remained at the end (after 110 days) of the incubation experiment. Despite the short-term drastic changes in microbial community composition, a highly diverse microbial community is similar to the control at the end of the incubation. It is suggested here that resuspension of coastal sediments weakens their role as a net sink of carbon, with most of the mobilized SDOM transformed by successive microbial communities in the overlying seawater and the remaining recalcitrant organic material becoming part of the long-lived DOM pool. Thus, the bioavailability of the coastal SDOM might influence the carbon budget in coastal oceans.

1. Introduction

Approximately 80% of marine sedimentary organic matter is buried in the shelf and slope areas (Hedges & Keil, 1995; Mahmoudi et al., 2017). Recycling of organic material in marine sediments is an important component of biogeochemical cycles because these sediments are recognized to be critical for long-term carbon sequestration (Arndt et al., 2013; Orsi et al., 2018). Much of the sediment pore water characterized as dissolved organic matter (DOM) originates from deposited marine particulate organic matter, which is produced in surface water and sinks to the sediment via the biological pump (Burdige & Komada, 2015; Rossel et al., 2016). A conceptual model of sediment DOM (SDOM) cycling shows that processes mediated by bacteria/archaea and exoenzymes may catalyze the transformation of high molecular weight organic molecules into low molecular weight (LMW) DOM (Arnosti, 2004; Burdige & Komada, 2015).

In a bulk sense, the mostly LMW of the SDOM is assumed to be recalcitrant (Burdige & Komada, 2015). This assumption is consistent with water column studies, which have shown that LMW DOM represents a more stable and diagenetically altered fraction of the total DOM (Benner & Amon, 2015; Walker et al., 2016) and that recalcitrant carboxyl-rich alicyclic molecules (Hertkorn et al., 2006) dominate the nuclear magnetic resonance spectra of sediment pore water DOM samples (Fox et al., 2018). However, molecular composition of DOM in sediment pore water is also characterized by abundant compounds such as unsaturated aliphatic lipids, fatty acids, and peptides, which are indicative of recent phytodetritus deposition and could be easily utilized by heterotrophic microorganisms (Rossel et al., 2016). A recent study shows that labile organic molecules in surface continental shelf sediments can be preserved due to hypoxic or anoxic conditions (Jessen et al., 2017). Thus, the intrinsic bioavailability of SDOM in the overlying seawater remains unclear but may have to do with oxidation of SDOM in oxygenated seawater. Still, much of the SDOM remains

uncharacterized at the compound/class or molecular levels; this also hinders our understanding of the bioavailability of sediment organic matter. In addition, the dissolved organic carbon (DOC) concentration of coastal sediment pore water has been reported to be up to an order of magnitude higher than in the overlying water (Burdige & Gardner, 1998). Understanding the intrinsic bioavailability and subsequent biogeochemical effects of SDOM in overlying seawater is important because mobilization of SDOM commonly occurs in coastal and estuarine areas through diffusion and resuspension by natural and anthropogenic activities (e.g., tidal currents, wind energy and storms, fishing, dredging, and the rotation of ships' propellers) (Burdige & Komada, 2015; Eggleton & Thomas, 2004; Reisinger et al., 2017; Superville et al., 2014; Zuo et al., 2016).

Although the carbon cycle of the coastal ocean is an important component of the global carbon budget, the diverse sources and sinks of carbon and their complex interactions in these systems are not well understood (Bauer et al., 2013) and that is particular true during resuspension events and it is known that disturbance strongly impacts the microbial diversity and production in coastal sediments (Galand et al., 2016). If SDOM contains labile organic matter, mobilization of which into the overlying seawater, where it mixes with the existing DOM, should stimulate additional microbial processes that would mainly impact remineralization of this SDOM. However, understanding these processes requires more in situ observations and experimental tests. In this study, SDOM was extracted from sediments in a transitional zone where riverine and oceanic systems combine and used for an incubation experiment with the overlying seawater. This approach allowed tracking of the microbial processes and organic matter transformations at the molecular level, providing novel insights into the overlooked biogeochemical effects and unclear fate of SDOM in overlying seawater.

2. Materials and Methods

2.1. Preparation of SDOM Extract

Twenty-three surface sediment samples (5 cm) were collected from the Xiamen coastal area (supporting information Figure S1) using a Kajak sediment core sampler (5 cm in diameter and 50 cm in length; KC-Denmark, Denmark) on 7 January 2016. For each sample, the top 1 cm of sediment was discarded, and the lower 4 cm was kept cold and in the dark for DOM extraction. Approximately 10 kg of sediment were added to 5 L of prechilled (4 °C) Milli-Q water in batches and shaken in the dark at 4 °C for 1 hr. The sediment-water mixture was centrifuged at 10,000 × g and 4 °C for 10 min, after which the supernatant was sequentially filtered through precombusted (480 °C for 5 hr) GF-75 glass fiber filters (0.3 μm, Advantec) and prerinsed (Milli-Q water) 0.2-μm polytetrafluoroethylene membranes (Millipore). The filtrate was acidified to pH = 2 using 30% HCl (p.a. grade, Merck). Solid-phase extraction of DOM was performed using standard methods (Dittmar et al., 2008). Briefly, 1-g styrene divinyl benzene copolymer cartridges (Bond Elut PPL, Agilent) were rinsed, activated with methanol (HPLC grade, Merck) and rinsed with Milli-Q water (pH = 2). In duplicate, the filtrates were passed by gravity though the cartridges, which were subsequently extensively rinsed with 0.1% (v:v) formic acid (HPLC grade, Sigma-Aldrich) water and completely dried before elution with HPLC-grade methanol. The methanol was then completely evaporated under ultrapure N_2. The extraction efficiency of the SDOM is approximately 70%.

2.2. Experimental Setup for SDOM Incubation

Prior to sediment sampling, a seawater sample (18.2 °C) was collected using a Niskin sampler (5 × 2.5 L) at the depth of 9 m, about 0.2 m above the sediment. The incubation experiment was conducted in 10-L polycarbonate carboys that were sequentially prewashed with 1 M HCl, Milli-Q water, and the designated seawater. Each carboy was then filled with 8-L seawater that was prefiltered through 3.0-μm polycarbonate filters (Millipore) to remove large particles and grazers while maintaining the free-living prokaryotic community and viral particles. The SDOM extract was then mixed into the seawater-filled carboys as SDOM treatments, while the no addition carboys were made as controls. In triplicate, both the control and SDOM treatments were conducted in the dark and at a conditioned temperature (18–20 °C) for 110 days.

2.3. Determining Bacterial and Viral Abundances

Water samples (2 ml) in triplicate were collected at intervals of the incubation experiment in cryovials and fixed with 1% glutaraldehyde (final concentration), flash-frozen in liquid nitrogen for 10 min and then stored

at −80°C until analyzed. At the end of the experiment, samples were stained with SYBR Green-I (Life Technologies). Bacterial and viral abundances were measured using a Becton Dickinson Accuri C6 flow cytometer and a Beckman Coulter Epics Altra II flow cytometer, respectively (Marie et al., 1997; Vaulot et al., 1989).

2.4. Organic Carbon Concentration Analysis

In triplicate, samples for total organic carbon (TOC) analyses were collected with glass pipettes into 40-ml glass vials, while water samples were filtered through precombusted GF-75 glass fiber filters (0.3 μm) for DOC samples. Both types of organic carbon samples were acidified to pH = 2 with H_3PO_4 and stored at −20 °C until analyzed. Organic carbon was then measured using the high temperature combustion method with a Shimadzu TOC-VCPH TOC analyzer and Milli-Q water for system blank subtraction (Callahan et al., 2004). Reference deep seawater (provided by the Hansell Organic Biogeochemistry Laboratory at the University of Miami, USA) served as an additional control.

2.5. DNA Extraction, PCR, Sequencing, and Bacterial Community Analysis

Water samples for DNA analysis were filtered through 0.2-μm pore size polycarbonate filters (47 mm, Millipore). Microbial genomic DNA was extracted from the polycarbonate membranes using the Power Soil DNA Isolation kit (MoBio Laboratories) according to the manufacturer's instructions. The 16S rRNA gene sequences were amplified by polymerase chain reaction (PCR) as described elsewhere (Herlemann et al., 2011). Phusion High-Fidelity PCR Master Mix (NEB) was used to amplify the V3+V4 hypervariable region of the 16S rRNA gene from microbial genomic DNA with the primer pair 341F (5′-CCTAYGGGRBGCASCAG-3′) and 806R (5′-GGACTACNNGGGTATCTAAT-3′) that also contained sample-specific barcodes. Equimolar amounts of PCR amplicons from different samples were mixed and purified using the QIAquick Gel Extraction kit (Qiagen). Sequencing libraries were generated using the NEB Next Ultra DNA Library Prep Kit for Illumina and sequenced on the Illumina MiSeq platform. Approximately 300-bp paired-end reads from the original DNA fragments were generated and merged using the FLASH software (Magoč & Salzberg, 2011). Sequences that contained more than one ambiguous nucleotide that did not have a complete barcode and primer at one end or that were shorter than 200 bp after removal of the barcode and primer sequences were eliminated. Chimeric sequences were also identified and removed. After quality filtering, denoising, and removal of potential chimeras, sequences were used to study the total bacterial community compositions. Sequence clustering was performed using the Uparse software (V7.0.1001) with a similarity cutoff of 97%, and data were clustered into operational taxonomic units (OTUs) (Zhang et al., 2016) The most abundant sequence in each cluster was selected to be the representative sequence. Sequence data were deposited in the NCBI Sequence Read Archive: SRP098780.

A principal coordinate analysis (PCoA) was used to determine the dissimilarity of samples to each other based on Bray-Curtis similarities with PAST software (ver. 3.13) (Hammer et al., 2001). Bray-Curtis dissimilarities were calculated based on relative abundance matrices of OTUs for communities.

2.6. Fourier Transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR-MS) Analysis of the Extracted DOM

DOM was solid-phase extracted as described by Dittmar et al. (2008). Here, 500-mg styrene divinyl benzene copolymer cartridges (Bond Elut PPL, Agilent) were activated with HPLC grade methanol and rinsed with acidified Milli-Q water (pH = 2). An aliquot of 500-ml seawater sample was filtered through a precombusted GF-75 glass fiber filter and then passed by gravity though the 500-mg PPL cartridge, which was subsequently extensively rinsed with 0.1% (v:v) aqueous formic acid solution and completely dried before elution with HPLC grade methanol. The DOM extracts were adjusted to yield an approximate DOC concentration of 25 mM and analyzed using a Bruker Apex Ultra FT-ICR mass spectrometer equipped with a 9.4-T superconducting magnet. Sample solutions were infused via an Apollo II electrospray ion source (ESI) at 180 μL/hr with a syringe pump. Typical operating conditions for negative ESI were as follows: spray shield voltage 3.5 kV, capillary column introduce voltage 4 kV, and capillary column end voltage −320 V. The ion transformation parameter for the quadrupole (Q1) was optimized at m/z 300. The mass range was set to m/z 200–800. The 2-M word size was selected for the time domain signal acquisition. A total of 128 time domain signals was added together to enhance the signal-to-noise ratio and dynamic range. The FT-ICR mass spectrometer was calibrated using a known homologous series of the Suwannee River natural organic matter sample (obtained

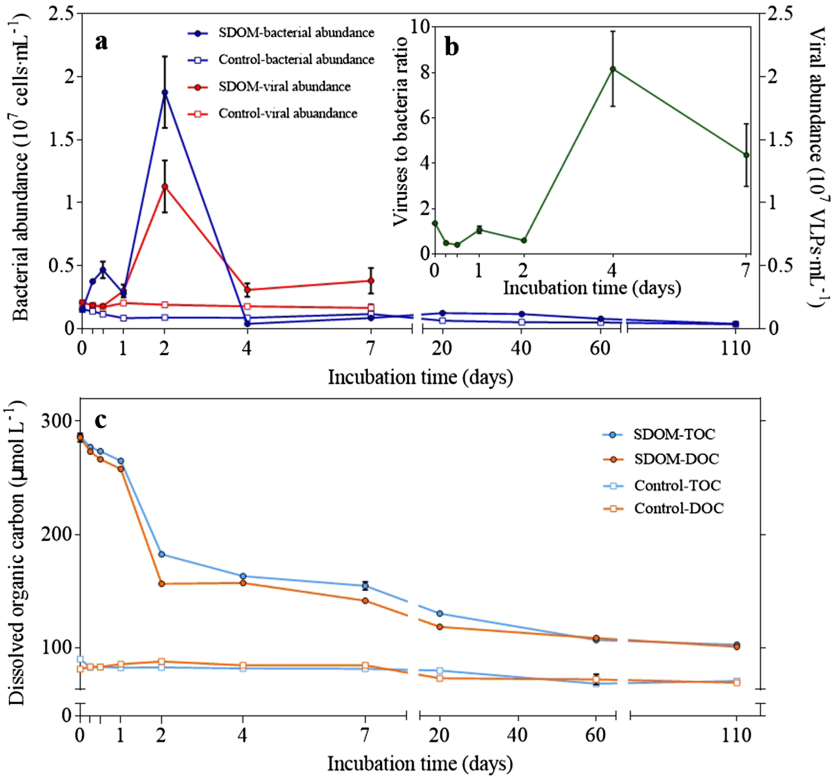

Figure 1. Utilization of SDOM by seawater microbial assemblages during the 110-day incubation. (a) Variation in bacterial and viral abundances. (b) Virus to bacteria ratios during the first 7-day incubation period. (c) Variation in total organic carbon (TOC) and dissolved organic carbon (DOC) concentrations over the course of the experiment. The experiment was conducted in triplicate; some values of error bars for the TOC and DOC data are too small and hidden within the symbols.

from International Humic Substances Society, USA), which contained a relatively high abundance of oxygen-containing compounds. These calibrant formulas also matched with seawater DOM. Molecular formulas were assigned with the calibrated mass data using an in-house software (Li et al., 2019). All assigned formulas had to meet the following basic chemical criteria: (1) the number of H atoms cannot exceed 2C + N + 2; (2) the sum of H and N atoms must be even (the "nitrogen rule"); and (3) the number of N or O atoms cannot exceed the number of C atoms (Flerus et al., 2012; Koch et al., 2007; Kujawinski & Behn, 2006). Mass lists of m/z values with signal-to-noise ratio of ≥6 and mass accuracy <1 ppm were discarded.

2.7. Correlation Analysis Between Microbial Community and DOM Composition

Associations between the bacterial OTUs and molecular weight of DOM were examined via Mantel's test (using vegan package in R). Mantel's testing was conducted between the relative abundance of the 29 most abundant OTUs at the family level (Bray-Curtis distance) and the DOM compound series (CHO, CHNO and CHOS) with their molecular weight (from 200 to 600 Da) within the first 7 days of control and SDOM incubations. The results were plotted as a heatmap using R (for windows 3.3.1), with different colors representing Spearman's rank correlation coefficients.

3. Results and Discussion

3.1. Microbial Utilization of SDOM Extract

Bacterial abundance and organic carbon concentrations (total and dissolved) were monitored to determine the bioavailability of the mobilized SDOM (Figure 1). Two apparent bacterial abundance changes were observed during the periods of 0 hr to 1 day and 1 day to 4 days after the SDOM addition (Figure 1a). The initial TOC and DOC concentrations are 89.8 ± 0.2 μmol C L^{-1} and 81.0 ± 0.9

Table 1
Weighted Averages of the Oxygen to Carbon Ratios (O/C_a), Hydrogen to Carbon Ratios (H/C_a), Double-Bound Equivalent (DBE_a), Modified Aromatic Index (AI_a), and Molecular Weight ($Mass_a$) Were Calculated From the Thousands Formulas of Each Sample (Control and SDOM Treatments at 0 hr and 110 Days) Measured by the Ultrahigh Resolution Mass Spectrometry

Sample	O/C_a	H/C_a	DBE_a	AI_a	$Mass_a$
Control-0 hr	0.417 ± 0.001	1.273 ± 0.001	8.997 ± 0.058	0.269 ± 0.001	438.959 ± 0.120
SDOM-0 hr	0.421 ± 0.007	1.284 ± 0.001	8.764 ± 0.033	0.264 ± 0.004	426.730 ± 0.046
Control-110 d	0.428 ± 0.002	1.253 ± 0.001	9.321 ± 0.044	0.276 ± 0.001	446.343 ± 2.328
SDOM-110 d	0.445 ± 0.004	1.241 ± 0.001	9.498 ± 0.017	0.279 ± 0.008	455.346 ± 3.166

μmol C L^{-1} in control treatments, and 286.6 ± 0.5 μmol C L^{-1} and 285.5 ± 3.7 μmol C L^{-1} in SDOM treatments (Figure 1c and supporting information Table S1). The rapid increase in bacterial abundance (Figure 1a) along with a sharp decline in DOC concentration (from 285.5 ± 3.7 μmol C L^{-1} to 156.3 ± 1.8 μmol C L^{-1}) (Figure 1c) within the first 2 days of the SDOM addition indicates that a large fraction (near half) of the SDOM was rapidly utilized or incorporated into the bacterial biomass. Both the TOC and DOC concentrations in control and SDOM treatments became relatively invariant after 60 days of incubation (Figure 1c and supporting information Table S1). At the end of the 110-day incubation, TOC and DOC concentrations are 69.3 ± 2.6 μmol C L^{-1} and 68.8 ± 1.6 μmol C L^{-1} in control treatments, and 102.6 ± 1.8 μmol C L^{-1} and 100.8 ± 0.7 μmol C L^{-1} in SDOM treatments. Comparison of DOC concentrations in the control and SDOM treatments shows that approximate 32 μmol C L^{-1} of SDOM (~16% of the added SDOM extract) remained, indicating the microbial resistance of residual SDOM even after the long incubation period.

Via the FT-ICR-MS analysis, we were able to describe the molecular composition changes of the SDOM. DOM mass spectra of the control and SDOM treatments at 0-hr and 110-day samples are shown in supporting information Figures S2 and S3 from one batch as examples. Masses were assigned to the solid-phase extracted DOM from the incubation experiments. A modified aromaticity index (AI) and double-bound equivalent (DBE) were calculated for each assigned formula according to Koch and Dittmar (2006). Weighted averages of the molecular weight ($Mass_a$), hydrogen to carbon ratios (H/C_a), oxygen to carbon ratios (O/C_a), AI_a and DBE_a were calculated for the control and SDOM treatments at 0 hr and 110 days to evaluate the overall characteristics of the DOM samples. We note that isotopologues were excluded from these calculations. Table 1 shows the H/C_a ratio of the initial SDOM treatments (1.284 ± 0.001) is significantly higher than that of the initial controls (1.273 ± 0.001) (t test, $p < 0.01$), whereas the $Mass_a$ and DBE_a values are found relatively low in the SDOM treatments (426.730 ± 0.046 and 8.764 ± 0.033 for $Mass_a$ and DBE_a, respectively) than the control treatments at 0 hr (438.959 ± 0.120 and 8.997 ± 0.058 for $Mass_a$ and DBE_a, respectively) (t test, $p < 0.01$ for $Mass_a$ and $p < 0.05$ for DBE_a), indicating an overall relatively high saturated of the SDOM extract. Molecules with higher H/C ratios have been found to accompany relatively high $\Delta^{14}C$ values, indicating that the relatively saturated organic compounds could be younger (Flerus et al., 2012), which might be preferentially utilized by microbes. In our study, Figure 2 shows a suite of molecules ($n = 1167$, with relatively high H/C and O/C ratios) decreased in normalized peak intensities during the 0-hr to 1-day incubation period in the SDOM treatments, whereas the other suite of molecules ($n = 1125$, with relatively low H/C and O/C ratios) decreased in normalized peak intensities during the 1-day to 2-day incubation. This indicates that heterotrophic microbes preferentially utilized the fraction of SDOM that is relatively saturated, some of which could represent aliphatic-like or saturated fatty acid-like compounds according to the definition of Šantl-Temkiv et al. (2013) and Medeiros et al. (2015). It should be emphasized here that FT-ICR-MS does not give any structural information beyond that what can be inferred from atomic ratios of each assigned molecular formula. Previous work by Rossel et al. (2016) shows that some degraded phytodetrital products, such as peptides, unsaturated aliphatics, and saturated fatty acids, can be prevalent in the surface sediment pore water. Similar to our findings, a recent study in freshwater found that aliphatic/lipid-like compounds in lake pore water DOM samples decreased in abundance after a 40-day incubation (Valle et al., 2018). Some other organic compounds, such as amino acids, carbohydrates, and peptides, might be prevalent in the sediments (Arndt et al., 2013;

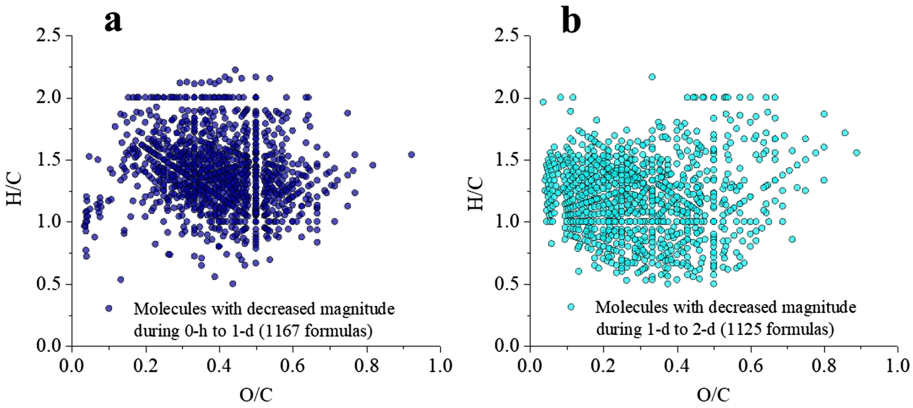

Figure 2. Detection of molecules metabolized during the early SDOM incubation experiment. Molecules with decreased peak intensities (normalized) during the (a) 0-hr to 1-day (navy) and (b) 1-day to 2-day (cyan) incubations are shown as van Krevelen diagram.

Niggemann et al., 2017) and are commonly labile for heterotrophic bacteria, mobilization of which could result in their large consumption. But these organic compounds are not detectable in negative mode ESI and our solid-phase extraction technique (Hertkorn et al., 2013; Osterholz et al., 2016).

However, the SDOM were not completely degraded. The residue SDOM might contain rather stable molecules from SDOM (Fox et al., 2018) and recalcitrant DOM (RDOM) produced after microbial processing (Gruber et al., 2006; Jiao et al., 2010; Ogawa et al., 2001). An overall significant decrease of H/C_a and increase of $Mass_a$, AI_a and DBE_a values were found in the SDOM 110-day treatments (1.241 ± 0.001, 455.346 ± 3.166, 0.279 ± 0.008 and 9.498 ± 0.017 for H/C_a, $Mass_a$, AI_a, and DBE_a, respectively) compared to the SDOM 0-hr treatment (1.284 ± 0.001, 426.730 ± 0.046, 0.264 ± 0.004, and 8.764 ± 0.033 for H/C_a, $Mass_a$, AI_a, and DBE_a, respectively) (*t* test, $p < 0.001$ for H/C_a, $Mass_a$, and DBE_a, $p < 0.05$ for AI_a) (Table 1). This again indicates that heterotrophic bacteria preferentially utilized more saturated compounds, and produced organic molecules that have more unsaturated structures. Previous studies found that decreasing DOM $\Delta^{14}C$ values go along with a shift in the molecular composition with increasing average Mass and DBE, but H/C ratios decrease when DOM sampling is conducted from surface to the deeper ocean (Flerus et al., 2012; Lechtenfeld et al., 2014; Li et al., 2019; Medeiros et al., 2015). In our incubation study, the transformation of SDOM during the 110 days (decreasing H/C_a from 1.284 ± 0.001 to 1.241 ± 0.001, increasing $Mass_a$ from 426.730 ± 0.046 to 455.346 ± 3.166 and DBE_a from 8.764 ± 0.033 to 9.498 ± 0.017) is somewhat similar to the trends observed in the water column DOM and changes observed from surface to deeper ocean waters (mentioned above), where has a relatively high content of RDOM that resist microbial degradation (Barber, 1968; Hansell, 2013; Hedges, 1992; Hertkorn et al., 2013; Hertkorn et al., 2006).

Excitation emission matrix fluorescence and parallel factor analysis were used in combination (supporting information Method) to evaluate variation of fluorescent DOM (FDOM) components in the control and SDOM treatments during the 110-day incubation. Three fluorescent components were analyzed (supporting information Figure S4). Comparison to previous studies indicated that C1 exhibited fluorescence peak typical of protein-like (tyrosine-like) component, and the C2 and C3 are often referred to as humic-like components (Catalá et al., 2015; Guo et al., 2014; Jørgensen et al., 2011; Zheng et al., 2019). Trends of these components were monitored during the 110-day incubation and shown in supporting information Figure S4. Specifically, the component C1 was decreasing, while the two large Stoke's shift peaks (C2 and C3) were increasing during the incubation. The component C1 has been often associated with more labile organic matter, while C2 and C3 seem to be indicative of relatively recalcitrant DOM in natural environments (Catalá et al., 2015; Tanaka et al., 2014). The C2 and C3 have also been observed to be associated with sulfate reduction (Luek et al., 2017) and degradation of picocyanobacteria-derived DOM (Zhao et al., 2017). These results from FDOM supports the process that the microbes in the overlying seawater utilized the labile materials and produced rather recalcitrant DOM, which then accumulates during the incubation experiments.

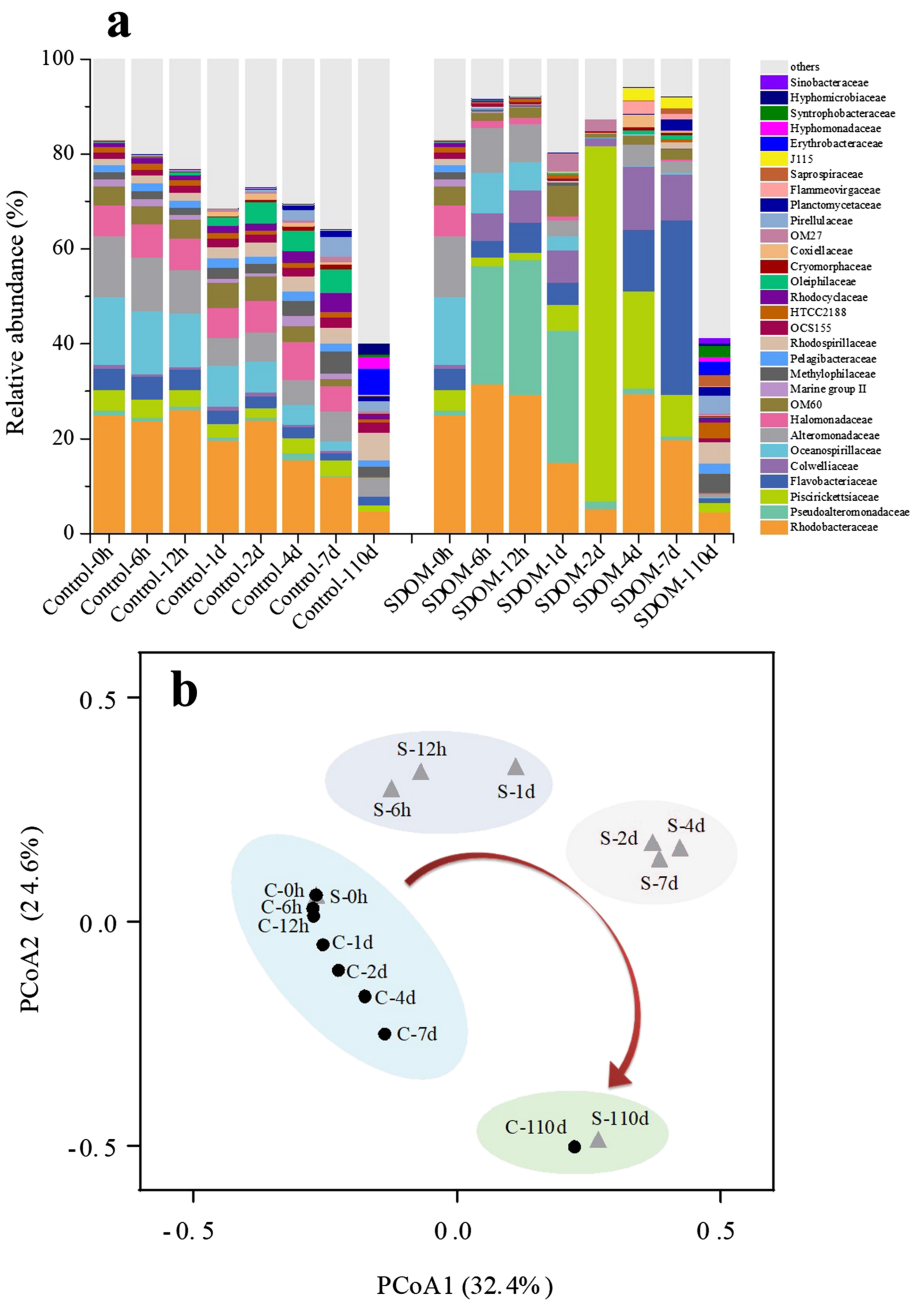

Figure 3. Microbial community succession during the 110-day incubation. (a) The relative abundance of each microbial family is expressed as the percentage of total sequences obtained in the treatment. (b) Principal coordinate analysis (PCoA) of the heterotrophic bacterial communities of control (C-, in dots) and SDOM (S-, in triangles) incubations using relative abundance of OTUs based on Bray-Curtis dissimilarities, similar bacterial communities are shown by colored ellipses.

3.2. Microbial Community Succession in the Incubation

To understand the changes in the bacterial community structure that occurred during the incubation, relative proportions of the OTUs to the total number of sequencing reads were calculated for each sample. The bacteria in the control samples were dominated by the class Gammaproteobacteria within the phylum Proteobacteria (supporting information Figure S5), and addition of the SDOM extract made further increase of their relative proportion over the incubation. Communities were relatively invariant in controls but fluctuated over the first 7 days of the SDOM treatments (Figure 3). Specifically, at the family level (Figure 3a),

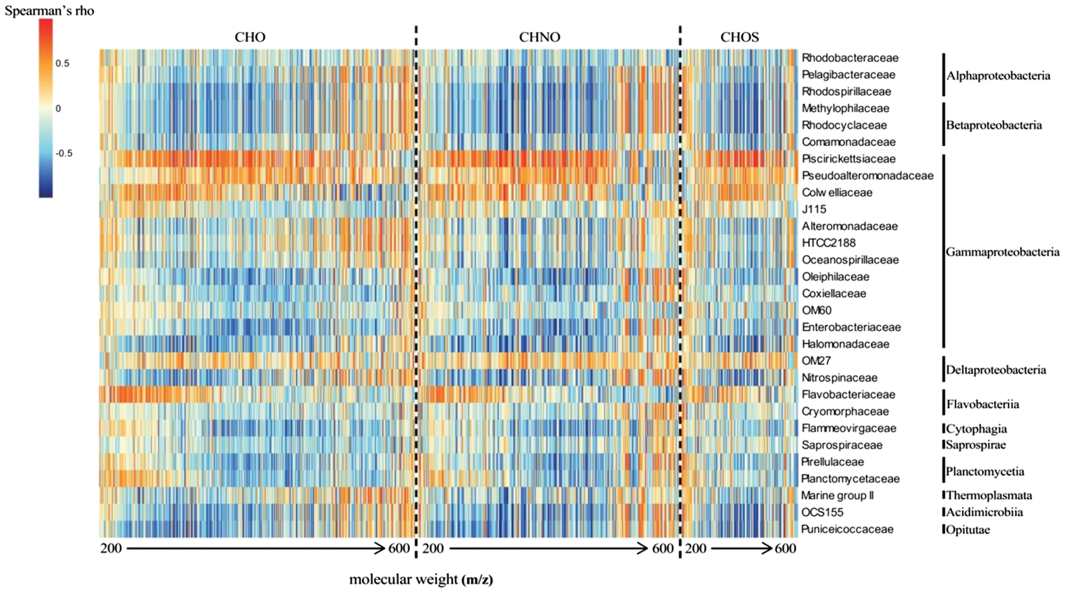

Figure 4. Correlation between OTU abundance and DOM characteristics. The heatmap shows the Spearman's rank correlation coefficients (Spearman's rho) from the correlation analyses between the relative abundance of the 29 most abundant OTUs (at the family level) and the DOM compound series with their molecular weight (m/z from 200 to 600) over the first 7 days of the control and SDOM incubations. A high correlation coefficient in red indicates a strong positive correlation between the relative abundance of the OTUs and DOM.

Pseudoalteromonadaceae within the Gammaproteobacteria was the major family that responded to SDOM addition during the first day incubation. *Piscirickettsiaceae* within the Gammaproteobacteria subsequently dominated the community at the 2-day incubation period. Gradually, *Flavobacteriaceae* within the Flavobacteria replaced *Piscirickettsiaceae* as the dominant family by 7 days of incubation. The changes in the bacterial community during the early phase of the incubation are also reflected in the PCoA results (Figure 3b). Active Bacterial members, such as the *Gammaproteobacteria*, *Flavobacteriia*, some *Alphaproteobacteria* and others, can grow rapidly becoming abundant under suitable condition (Fuhrman et al., 2015), which is the addition of labile SDOM in current experiment. Specifically, the most labile fraction of SDOM extract might be responsible for altering the bacterial community composition, supporting the quick growth of communities. The strong correlation between specific taxa and DOM within the molecular weight of 200–600 Da during the first 7 days (Mantel's test; $P < 0.001$, $r = 0.9947$, Figure 4), combined with the result of Figure 3 indicates that different microbial taxa were active in transforming different molecules of the SDOM. Finally, the taxonomic composition of the bacterial community and the PCoA results both show diversified and similar bacterial community composition in the 110-day control and SDOM treatments (Figure 3). Marine microbial communities are dynamic but also resilient. It has been proposed that behaviors of microbes are broadly predictable, which means that the altered community can be maintained back to steady after a disturbance (Fuhrman et al., 2015; Teeling et al., 2016).

The viruses and DOM are major top-down and bottom-up factors controlling microbial composition and diversity (Liu et al., 2017; Suttle, 2007), together affecting microbial community succession. In this study, both the control and SDOM treatments were prefiltered through 3.0-μm filters to remove large particles and grazers. Likely, no grazing effect, sloppy feeding, and dissolution of particulate organic matter existed in the experiment. The difference between TOC and DOC could then represent the major cellular carbon. Sharp decline in bacterial abundance (from 1.88×10^7 cells ml^{-1} to 3.78×10^5 cells ml^{-1}) and a decrease of TOC concentration (from 184.4 ± 1.8 μmol C L^{-1} to 163.0 ± 0.5 μmol C L^{-1}) (Figure 1) were found in the SDOM treatments from 2 days to 4 days, but the DOC concentration remained relatively stable during this period (156.3.0 ± 1.8 μmol C L^{-1} and 157.0 ± 2.2 μmol C L^{-1} for 2 days and 4 days, respectively). These results indicate that some cellular organic carbon was

transformed to DOC or microbially utilized. In addition, the sharp decline in bacterial abundance along with a concomitant increase in the viruses to bacteria ratio (Figure 1b) was found during the incubation period of 2 days to 4 days in the SDOM treatments, indicating that the release of cellular carbon probably occurred via viral lysis or cell death. Previous studies have demonstrated that the virus-mediated lysate is mostly labile and can support new bacterial communities (Haaber & Middelboe, 2009; Lønborg et al., 2013; Middelboe & Jorgensen, 2006; Sheik et al., 2014). The virus-mediated transformation of cellular organic carbon to dissolved forms, which is known as the "viral shunt," influencing biogeochemical cycles in marine ecosystems (Brussaard et al., 2008; Malits et al., 2014; Suttle, 2005). Considering the shifts in bacterial community composition, and the viral behavior within the first 7 days of this incubation, we suggest that viral-bacterial dynamics would play a role in promoting bacterial community succession, which subsequently propels DOM transformation. Combined this with the molecular composition changes of the SDOM, our experiment demonstrated that successive processing of DOM by microbes is essential for the transformation of labile organics into RDOM (Jiao et al., 2018; Lechtenfeld et al., 2014; Osterholz et al., 2015).

3.3. Implications for the Resuspension of SDOM Into the Overlying Seawater

Our experimental results show that dynamics of organic carbon concentrations, DOM composition, and bacterial community structure were coupled. The response of the microbial community, for example, the *Pseudoalteromonadaceae*- and *Piscirickettsiaceae*-dominated communities likely contribute to the major SDOM degradation in the beginning, resulted in consumption of a large fraction of the SDOM. These results indicate that most of the partially cycled SDOM by bacteria and/or archaea in the sediments (Burdige & Gardner, 1998; Burdige & Komada, 2015; Orsi et al., 2018; Valle et al., 2018) is labile though it can be preserved in hypoxic sediment cores (Jessen et al., 2017). Since resuspension of coastal sediments would introduce DOM into seawater, if SDOM contains more labile molecules, which can be largely respired by microbes though a small fraction of RDOM is produced as well. In such case, resuspension of coastal sediments would weaken their role as a net sink of carbon. Therefore, the bioavailability of SDOM would directly influence the gross carbon budget in coastal areas.

The long-term incubation shows the almost depleted of labile SDOM (Figure 1c) and the accumulation of relatively refractory DOM by FDOM analysis (supporting information Figure S4). With the FT-ICR-MS analysis, the overall properties of the residual SDOM indicate that the SDOM was microbially modified to a higher unsaturated state and likely containing more aromatic rings in molecules, these properties of residue SDOM are similar to the properties of deep-sea RDOM (Catalá et al., 2015; Flerus et al., 2012; Hertkorn et al., 2013; Lechtenfeld et al., 2014; Li et al., 2019; Medeiros et al., 2015). The chemical changes of DOM properties (Table 1) combined with the succession of microbial community (Figure 3b) observed in our study might reflect the generation of RDOM via the microbial carbon pump (Jiao et al., 2010), which proposes that the microbial successive processing on labile DOM generates long-lived RDOM in the water column (Jiao et al., 2018; Jiao et al., 2014). Our SDOM incubation experiment suggests that the microbial mediated RDOM may contribute to the autochthonous recalcitrant carbon in coastal environments (Asmala et al., 2018), which could be subjected to further cycling through biotic and abiotic processes in the ocean. In estuaries and coastal waters, processes have been identified to affect the cycling of RDOM, such as tidal variation, salinity gradient, terrestrial inputs, photochemical degradation, storms, sediment remobilization, changing microbial assemblage, and others (Bauer et al., 2013; Lønborg & Álvarez-Salgado, 2012). Future studies are urged to study effects of physical and biochemical processes on the carbon budget at the sediment-water interface.

4. Conclusions

This bioassay study demonstrates that coastal SDOM contains a large fraction of labile organic compounds that can be quickly utilized by microbes in the overlying seawater. In addition, by analyzing chemical composition and microbial community, successive processing of DOM is suggested vital in transforming labile SDOM to a higher recalcitrant state. The remained SDOM after the 110-day incubation was recalcitrant to microbial utilization. This recalcitrant organic material would become part of the long-lived DOM pool and can be subjected to further cycling through further biogeochemical processes in the ocean. Overall,

the bioavailability of coastal SDOM would directly influence the carbon budget at sediment-water interface, since we found that resuspension of labile SDOM into overlying seawater may weaken the role of sediment as a net sink of carbon.

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