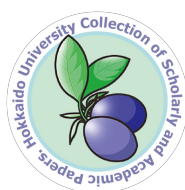




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Title	Tracing Sediment-Affected Water Masses From Coastal Areas to Offshore Oceans Using In Situ Sensing of Fluorescent Organic Matter
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Key Points:

- A portion of the fluorescent organic matter in the Kuroshio-Oyashio transition zone could be laterally transported from coastal areas
- High levels of fluorescent organic matter in subsurface layers in coastal regions are linked with sediment-derived nutrients
- Sensor-based observation of fluorescent organic matter can be used to effectively trace the export of coastal sediment-derived nutrients

Supporting Information:

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

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Tracing Sediment-Affected Water Masses From Coastal Areas to Offshore Oceans Using In Situ Sensing of Fluorescent Organic Matter

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Abstract The lateral transport of nutrient-rich water masses from coastal areas to offshore areas is an important factor controlling the primary production of phytoplankton in offshore regions. If the coastal low-salinity waters have high nutrient levels, lateral transport can be traced via salinity observations. However, if high level of nutrients in the waters are not coupled with low salinity, for example, input from coastal sediments, water transport cannot be traced via salinity observations. In this study, visible fluorescent organic matter (FOM_{vis}) was monitored by an in situ sensor as a tracer for sediment-affected water masses from coastal areas. The FOM_{vis} distribution in the upper 500 m water layer in offshore Japan along the 143°E transect was generally controlled by the distributions of subtropical water (i.e., Kuroshio water and Tsugaru warm current water) and subarctic water (i.e., Oyashio water). However, anomalously high levels of FOM_{vis} were observed at depths of 20–60 m at an observational station, which could not be explained by physical mixing. This likely originated from the lateral transport of water masses from coastal areas. In and around Sendai Bay, where observations were carried out as an example of the Japanese coast, high levels of FOM_{vis} were observed, accompanied by negative N*, an indicator of denitrification, and phosphate and silicic acid inputs from anoxic sediments. These observational results imply that FOM_{vis} observation via in situ sensors can be useful for tracing the input of nutrient-rich sediment-affected water masses from coastal areas to offshore oceans.

Plain Language Summary Phytoplankton activities in offshore oceans are often controlled by the supply of nutrients. Since the coastal oceans are generally characterized by high levels of nutrients, lateral transport of nutrients from the coastal ocean to the offshore ocean is likely an important nutrient source for phytoplankton, particularly in the boundary region. Rivers and sediments supply nutrients to coastal oceans. Nutrient transport accompanied by river water can be traced by salinity observations using conductivity sensors. However, the transport of sediment-derived nutrients is difficult to trace via sensor-based observations. The visible fluorescence of nonliving organic matter (traditionally defined as humic-like fluorescence) has been suggested to be useful for tracing river water as well as coastal sediment-affected water in marine environments. Here, we observed the simultaneous presence of sediment-derived nutrients and visible fluorescent organic matter, which was detected by a sensor in the subsurface layer in the coastal area, that is, in and around Sendai Bay, Japan. A relatively high level of fluorescent organic matter was also observed in the Kuroshio-Oyashio transition zone. These observational results indicate that fluorescent organic matter as detected by in situ sensors can be useful for tracing inputs of coastal sediment-derived nutrients to offshore oceans.

1. Introduction

The spatial pattern and magnitude of primary production of phytoplankton in the open oceans are controlled by several environmental factors, and macronutrients (N, P, and Si) and micronutrients (e.g., Fe) are among these factors (Moore et al., 2013). These nutrients are transported vertically from the subsurface layer (Gupta et al., 2022; Ledwell et al., 1993; Nishioka et al., 2020) and through atmospheric deposition (Jickells & Moore, 2015) or laterally from high-nutrient regions, such as coastal areas (Giraud et al., 2008; Ueno et al., 2023; Whitney et al., 2005). The composition (stoichiometry) of macro- and micronutrients is also an important factor in shaping primary production in the open oceans, particularly in high-nutrient low-chlorophyll regions (Browning

& Moore, 2023; Moore et al., 2013). The most important source of macro- or micronutrients is dependent on the season as well as the oceanic region (Jickells et al., 2017). Although the lateral transport of water masses from coastal areas is likely important as a source of nutrients around the boundary region between coastal areas and offshore oceans, model studies have suggested the importance of coastal nutrient supplies to the open oceans at basin scales (Giraud et al., 2008; Bernard et al., 2011; da Cunha & Buitenhuis, 2013).

Coastal areas are highly productive due to the input of nutrients from rivers, submarine groundwaters, sediments, the atmosphere, and upwelling from slopes (Bianchi, 2007; Cloern et al., 2014; Nakajima et al., 2021; Santos et al., 2021). Most of the organic matter produced by coastal primary producers is remineralized in seawater and sediment in coastal areas (Bianchi, 2007; Cloern et al., 2014). The nutrient flux from coastal sediment is comparable to or greater than river flux (Bianchi, 2007; Giraud et al., 2008). The nutrients transported from coastal areas to offshore oceans, which are linked with freshwater transport, can be traced via salinity (Sharples et al., 2017). However, the water masses with nutrients derived from coastal sediments often do not have low salinity but can be laterally transported to the offshore oceans by nearshore ocean currents and associated mesoscale eddies (Ueno et al., 2023). The transport of sediment-derived nutrients can be traced via chemical parameters such as N^* , a tracer for denitrification and nitrogen fixation (Gruber & Sarmiento, 1997; Yoshikawa et al., 2006), and radon and radium isotopes (Moore, 1996; Sugimoto et al., 2016). Although salinity can be determined easily and continuously by using a conductivity sensor, the determination of chemical parameters usually requires water sampling and chemical analysis. Therefore, the spatiotemporal resolution of chemical parameter distributions is considerably lower than that of salinity, making it difficult to assess the transport of nutrients from coastal sediments to offshore oceans.

The fluorescent fraction of dissolved organic matter (DOM) can be determined by fluorescence sensors (Coble et al., 1998; Conmy et al., 2014; Shigemitsu et al., 2020; Yamashita et al., 2015). The fluorescence signals are separated into two groups, namely, UVA fluorescence (290–370 nm emission wavelength; traditionally defined as protein-like fluorescence) and visible fluorescence (370–600 nm emission wavelength; traditionally defined as humic-like fluorescence) (Coble, 1996; Stedmon & Nelson, 2015). Since the levels of visible fluorescent DOM in river water are usually greater than those in seawater and sea-ice melt water, visible fluorescent DOM has been used as a tracer of freshwater (Chen & Gardner, 2004; Mizuno et al., 2018; Stedmon et al., 2021; Tanaka et al., 2016) and fresh submarine groundwater (Cho et al., 2021; Kim et al., 2018; Suryaputra et al., 2015) in coastal areas. In addition to terrestrial sources, visible fluorescent DOM is microbially produced in seawater and marine sediments (Chen et al., 2016; Goto et al., 2020; Shimotori et al., 2012; Skoog et al., 1996); thus, the levels of visible fluorescent DOM in sediment porewater are generally greater than those in overlying seawater (Burdige et al., 2004; Chen & Bada, 1990). Furthermore, high levels of visible fluorescent DOM in the water columns, which were likely derived from sediments, were observed in the Bering and Chukchi Seas (Hioki et al., 2014; Yamashita et al., 2019), the Baltic Sea (Kowalczyk et al., 2015; Reader et al., 2019), off Peru (Loginova et al., 2016), and the Izu-Ogasawara Trench (Shigemitsu et al., 2021). Visible fluorescent DOM is easily degraded by sunlight (Mopper et al., 1991; Omori et al., 2011; Timko et al., 2015) but is not susceptible to microbial degradation (Yamashita et al., 2020; Yamashita & Tanoue, 2008). These observations indicate that visible fluorescent DOM can be used as a tracer of sediment-affected water masses to trace sediment-derived nutrients in subsurface oceans.

In this study, we observed visible fluorescence by using an in situ fluorometer connected to a conductivity-temperature-depth (CTD) sensor during a cruise conducted along a south–north transect off Japan, including the Kuroshio-Oyashio transition zone (Figure 1). A near-shore path of Kuroshio and Oyashio could carry nutrients from coastal areas offshore. The entrainment of water masses in coastal areas into offshore oceans by mesoscale eddies related to the Kuroshio and Oyashio currents has been reported (Kasai et al., 2002; Long et al., 2019). To evaluate coastal sediments as a source of visible fluorescence and nutrients, we carried out an additional cruise to observe the spatial distributions of visible fluorescence and nutrients in and around Sendai Bay, Japan, as an example of the Japanese coast. Sendai Bay is the area between Oshika Peninsula and Cape Unoo, with a coastline of about 130 km, a bay mouth of about 68 km, and water depths shallower than 50 m for the most part (Figure 1). The near-bottom dissolved oxygen (DO) concentration of Sendai Bay is known to be highest in April and decreases until October (Ishikawa et al., 2023). Macronutrient inputs from the sediment to the water column were observed in summer and fall with decreasing DO concentration and increasing water temperature (Ishikawa et al., 2023). The visible fluorescence determined by in situ fluorometers was derived not only from the visible fluorescence of DOM but also from that of particulate organic matter, even though most visible

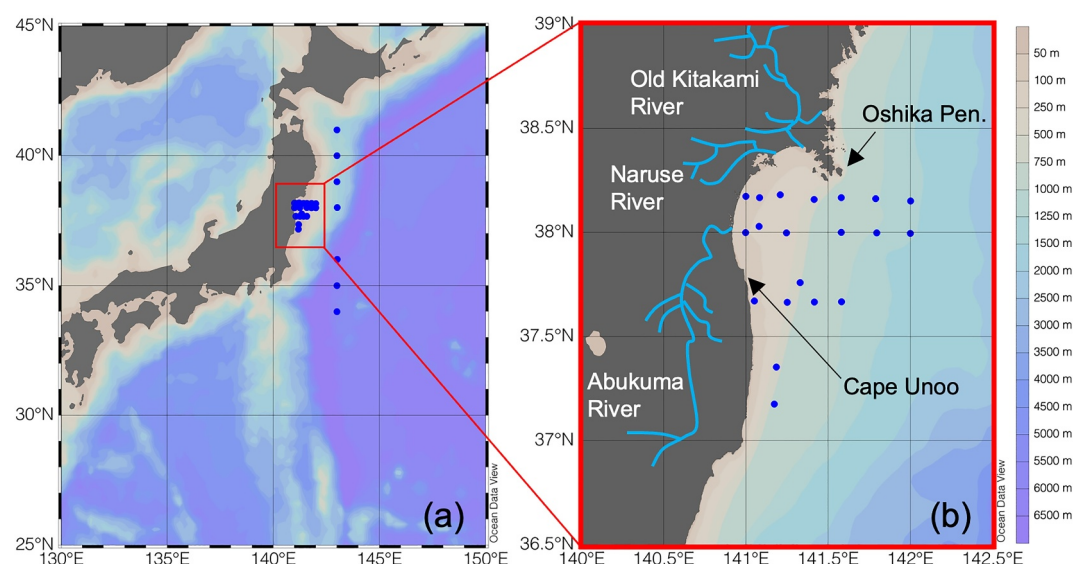


Figure 1. Map of the study area showing (a) all observation stations and (b) observation stations in and around Sendai Bay, Japan.

fluorescence was found to be derived from DOM in oceanic environments (Yamashita et al., 2021; Yamashita & Tanoue, 2009). Thus, we defined visible fluorescence as determined by an in situ fluorometer as fluorescent organic matter (FOM_{vis}) according to Yamashita et al. (2015).

2. Materials and Methods

2.1. Observations

Observations were conducted from the surface water to a depth of 1,500 m at a transect extending along 143°E, off Japan (Figure 1), during a cruise by the T/S *Oshoro-Maru* (C077, 14–20 September 2019). An in situ fluorometer to determine FOM_{vis} (ECO-FL(RT)D, Sea-Bird Scientific, Inc., WA, USA) was connected to a CTD unit (SBE 9plus, Sea-Bird Electronics, Inc., WA, USA). The excitation and emission wavelengths of the fluorometer were 370 and 460 nm, respectively. The fluorometer was attached to the frame of the CTD-Carousel water sampler system, and its optical head was facing downward. A DO sensor (SBE 43, Sea-Bird Electronics, Inc.) was also connected to the CTD. The DO concentration that was measured by the sensor was calibrated against the concentration determined by the Winkler method. The apparent oxygen utilization (AOU) was calculated as the difference between the solubility, which was calculated using the function of Weiss (1970), and the measured DO concentration. Seawater samples were collected with Niskin-X bottles equipped with a CTD-Carousel water sampler system from 12 depths ranging from 5 to 1,500 m at each station for FOM_{vis} and nutrient analyses. The samples for FOM_{vis} and nutrient analyses were collected without filtration in acid-washed spitz tubes and spitz tubes, respectively, and stored frozen until analysis in the laboratory on land.

Other observations were carried out in and around Sendai Bay, Japan (Figure 1), during a cruise by the R/V *Wakataka-maru* (WK2107, 30 July–5 August 2021). A fluorometer to measure FOM_{vis} (ECO-FL(RT)D) was also connected to a CTD unit (SBE 9plus) during the cruise. Seawater samples were collected with Niskin-X bottles equipped with a CTD-Carousel water sampler system from 2 to 6 depths ranging from 10 to 7 m above the bottom at each station for FOM_{vis} and nutrient analyses. Seawater samples were also collected from surface water using a bucket. The samples for FOM_{vis} and nutrient analyses were collected without filtration in acid-washed spitz tubes and spitz tubes, respectively, and stored frozen until analysis in the laboratory on land. Samples for salinity analysis with a salinometer were also collected by a bucket and Niskin-X bottles, and the salinity measured by CTD was corrected by that determined by the salinometer.

It should be noted that the oceanographic conditions, such as positions of Kuroshio Current, Oyashio Current, Tsushima Warm Current, and associated mesoscale eddies were different between the two cruises, because the

two cruises were conducted at different times (September 2019 vs. July–August 2021). Therefore, the observational results from two cruises were not directly linked.

2.2. Chemical Analyses

In the laboratory on land, the fluorescence intensity of FOM_{vis} was determined with a benchtop fluorometer (FluoroMax-4, Horiba) according to Yamashita et al. (2015). The frozen samples were thawed and left until they reached room temperature prior to fluorescence measurements. The emission spectrum from 400 to 500 nm taken at 1 nm intervals was obtained at 370 nm excitation. The slit width was set to 20 nm for both excitation and emission. The fluorescence spectrum was scanned with a 0.25 s integration time and acquired in the signal-to-reference mode with correction of the instrument-specific response (Sc/Rc mode). The fluorescence spectrum of freshly prepared Milli-Q water was subtracted from the sample spectra, and the fluorescence intensity was subsequently corrected to modified Raman units (RU_{370}) with the height of the Raman scattering at 370 nm excitation of the freshly prepared Milli-Q water (Yamashita et al., 2015). The fluorescence intensity at 460 nm was used for calibration of the in situ fluorometer.

The salinity and nutrient concentrations were measured using a salinometer (AUTOSAL8400B, Guildline Instruments) and an autoanalyzer (QuAatro2–HR, SEAL Analytical or AACS-III; Bran + Luebbe), respectively. N^* , a potential tracer of denitrification, was calculated according to Yoshikawa et al. (2006) as follows:

$$N^* = ([NO_3^-] + [NO_2^-] - 16[PO_4^{3-}] + 2.9) \times 0.87$$

2.3. Correction and Calibration of the In Situ Fluorometer

All the parameters obtained by the sensors were processed in the same way and were averaged over each pressure with SBE Data Processing-win32 (Sea-Bird Electronics, Inc.). The effect of temperature on FOM_{vis} was standardized according to Watras et al. (2011) as follows:

$$FOM_{vis,r} = FOM_{vis,m} / [1 + \rho(Tm - Tr)]$$

where subscripts r and m represent the reference and measured values, respectively. Tm and Tr are the in situ temperature and 20°C, respectively. For ρ , the temperature-specific coefficient of fluorescence ($^{\circ}C^{-1}$), -0.006 , was used according to Yamashita et al. (2015). The $FOM_{vis,r}$ was hereafter referred to FOM_{vis} . The data from the down-cast were used to obtain the vertical profile of each parameter, but water samples were collected from the up-cast. To calibrate the FOM_{vis} sensor for deep offshore ocean observations during the T/S *Oshoro-Maru* C077 cruise, the FOM_{vis} measured by the in situ fluorometer during the down-cast was compared with that collected during the up-cast and determined by a benchtop fluorometer to avoid the effect of pressure hysteresis in the in situ fluorometer (Shigemitsu et al., 2020). However, to calibrate the in situ sensor for shallow coastal observations during the R/V *Wakataka-maru* WK2107 cruise, both the FOM_{vis} measured by the in situ fluorometer and that determined by the benchtop fluorometer were obtained from the up-cast.

3. Results

3.1. Distributions of Potential Temperature, Salinity, AOU, and FOM_{vis} Along the 143°E Line

The potential temperature and salinity varied from 2.19 to 28.28 and 33.14 to 34.85, respectively, in the upper 500 m of the south–north transect along 143°E (Figure 2). Figure 3 shows the T–S diagram from the surface to 1,500 m of the transect. In the upper 500 m, the potential temperature and salinity in the southern region were generally greater than those in the northern region. These distributional patterns occurred because the Kuroshio and Oyashio water originated from the south and north of the study regions, respectively. On the one hand, the subsurface waters at the three southernmost stations were located in the Kuroshio water system according to the T–S diagram (Hanawa & Mitsudera, 1987). On the other hand, the subsurface waters at the three northernmost stations were characterized as Oyashio water system according to the T–S diagram (Hanawa & Mitsudera, 1987). Interestingly, the potential temperature and salinity in the upper 200 m of water at the northernmost station (41°N) were greater than those at stations located south of this station (39°N and 40°N) due to the distribution of the Tsugaru warm current at the northernmost station (Figures 2 and 3). Although the subsurface water at the 38°N

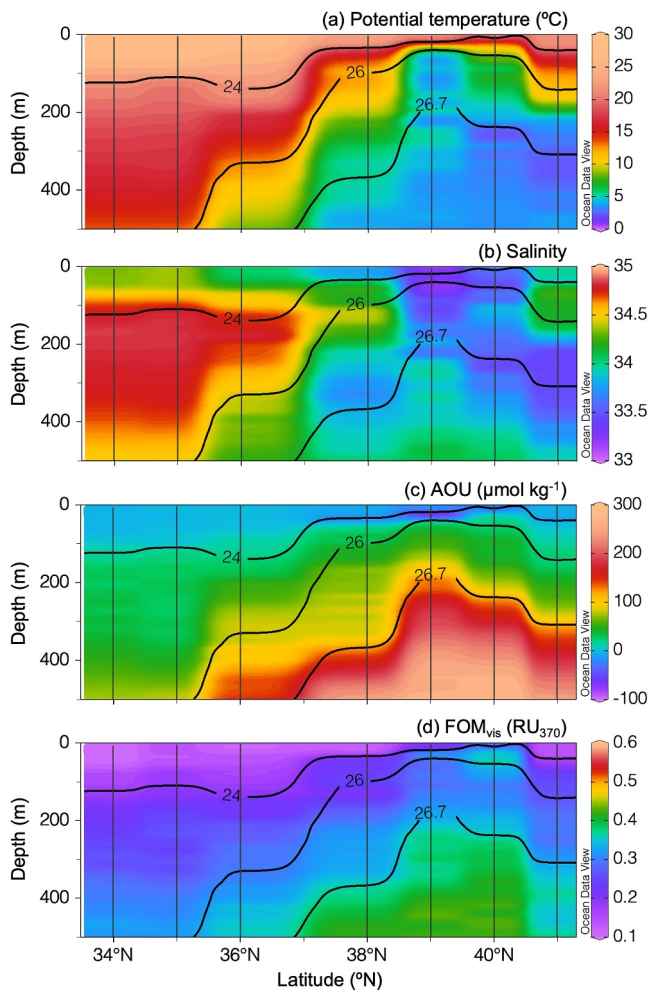


Figure 2. The south–north transect distributions of (a) potential temperature, (b) salinity, (c) AOU, and (d) FOM_{vis} as determined by sensors along a 143°E transect. The contour lines in each panel represent sigma-theta values of 24.0 (the boundary between the surface-layer water system and the subtropical water systems), 26.0 (the approximate boundary between the surface-layer water system and the Oyashio water system), and 26.7 (the major boundary between the cold lower-layer water system and other water masses). The depth of the surface mixing layer (defined as the temperature within 0.5°C of the surface water temperature) was 62 m at 34°N, 61 m at 35°N, 45 m at 36°N, 26 m at 38°N, 13 m at 39°N, 14 m at 40°N, and 31 m at 41°N.

Information S1). Extremely high levels of FOM_{vis} ($>0.6 RU_{370}$) were found in the subsurface layer at some stations (Figure 4c) but were not linked with changes in potential temperature or salinity (Figure 5). Figure 6 shows the relationships between salinity and FOM_{vis} with respect to the color of N^* using samples obtained by the CTD-Carousel water sampler system and the bucket. The high levels of FOM_{vis} can be separated into two groups. The first group was characterized by low-salinity water and positive N^* values, and the second group was associated with near zero and negative N^* values. The vertical profiles of the second group FOM_{vis} determined by the sensor showed the extremely high values near the bottom at two shallow stations in Sendai Bay and high values with a 10 m thick layer at a relatively deep station located outside Sendai Bay (Figure S2 in Supporting Information S1).

station was also characterized as a Tsugaru warm current system in the T-S diagram (Hanawa & Mitsudera, 1987), the water was likely a mixture of the Kuroshio water system and Oyashio water system.

The AOU was lowest in surface water and increased with depth in the upper 500 m of the water column (Figure 2c). Markedly negative AOU values were observed in the upper 40 and 20 m of the water column at 39°N and 40°N, respectively. At depths ranging from 100 to 500 m, the distributional pattern of AOU was opposite to those of potential temperature. AOU at the depths is high in the Oyashio water system but low in the Kuroshio water system and Tsugaru warm current system.

The FOM_{vis} level ranged from 0.07 to 0.46 RU_{370} and was generally lowest in surface water and increased with depth in the upper 500 m of the water column (Figure 2d). The FOM_{vis} level in surface water is generally related to potential temperature and salinity; that is, the FOM_{vis} level is low in high-temperature and high-salinity water and vice versa. However, interestingly, the FOM_{vis} level at 20–60 m at 40°N was greater than that at 39°N, even though the temperature and salinity of the water mass at 40°N were greater than those at 39°N. At depths ranging from 100 to 500 m, the distributional pattern of FOM_{vis} was similar to that of AOU; that is, a high level of FOM_{vis} was linked with high AOU.

3.2. Distributions of Potential Temperature, Salinity, N^* , and FOM_{vis} in and Around Sendai Bay, Japan

The vertical profiles of potential temperature, salinity, and FOM_{vis} in and around Sendai Bay are shown in Figure 4. The potential temperature ranged from 3.68 to 26.37°C and was highest at the surface and decreased with depth in and around the bay. The salinity ranged between 31.59 and 34.16, and the lowest salinity occurred at the surface. Low salinity (<33) in the surface waters (<10 m) was mainly observed at nearshore stations, where the bottom depth was approximately 20–30 m (Figure S1 in Supporting Information S1). The major water masses in the subsurface layer (20–150 m) were characterized as the Tsushima warm current system, and the water masses beneath the subsurface layer were characterized as the Oyashio water system (Figure 5; Hanawa & Mitsudera, 1987).

The FOM_{vis} concentration ranged from 0.14 to 1.21 RU_{370} and was generally lower in the surface waters than in the subsurface layer (Figure 4c). The FOM_{vis} levels in the surface waters at nearshore stations where the bottom depth was approximately 20–30 m were relatively high ($>0.4 RU_{370}$; Figure S1 in Supporting Information S1). High FOM_{vis} levels were linked with low-salinity water (Figure 5 and Figure S1 in Supporting

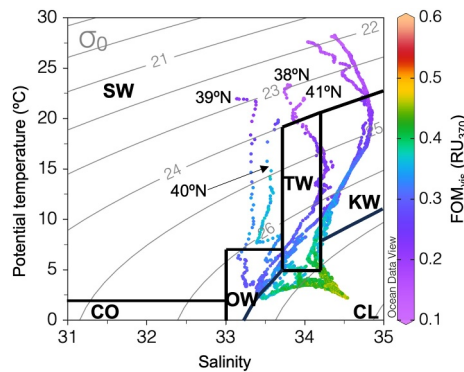


Figure 3. Potential temperature-salinity (T-S) diagram with FOM_{vis} along a $143^{\circ}E$ transect. All the parameters were determined by sensors. The gray lines in the diagram indicate the contour lines of σ_{θ} . The six water systems are classified according to Hanawa and Mitsudera (1987) as follows: TW: Tsugaru warm current water system; OW: Oyashio water system; KW: Kuroshio water system; CL: cold lower-layer water system; SW: surface-layer water system; CO: Coastal Oyashio water system.

4. Discussion

4.1. Factors Controlling the FOM_{vis} Level Along the $143^{\circ}E$ Line

The FOM_{vis} levels in the $143^{\circ}E$ line were generally low in the Kuroshio water and Tsugaru warm current systems, which originate from the subtropical region, but high in the Oyashio water system, which is a part of the western subarctic gyre. The levels of visible fluorescence of DOM and chromophoric DOM (CDOM) in the surface layer in the subtropical region are generally lower than those in the subarctic region due to extensive photodegradation and relatively long residence times (Nelson et al., 2010; Swan et al., 2009; Yamashita et al., 2013, 2017; Yamashita & Tanoue, 2009). Recently, visible fluorescence of DOM has been suggested to be a useful tracer for water masses in the Kuroshio-Oyashio confluence region because the ranges of visible fluorescence and salinity are different among Oyashio Water, Kuroshio Water, and North Pacific Intermediate Water (NPIW) (Wang et al., 2022, 2023). The NPIW is defined by a density (σ_{θ}) ranging from 26.6 to 27.4 or 27.5 (Yasuda, 2004; Yasuda et al., 2001) and is mostly overlapped by a cold-lower-layer water system in the T-S diagram (Figure 3).

The FOM_{vis} level in the NPIW was highest in the $143^{\circ}E$ transect (Figures 2 and 3). Thus, the general distributional pattern of FOM_{vis} in the upper 500 m except for the surface water along the $143^{\circ}E$ line is likely controlled by a physical mixing process. Although the levels of FOM_{vis} can change with local biogeochemical processes, that is, production by phytoplankton and heterotrophic prokaryotes (Fukuzaki et al., 2014; Goto et al., 2020; Rochelle-Newall & Fisher, 2002; Romera-Castillo et al., 2010) and photodegradation (Mopper et al., 1991; Omori et al., 2011; Timko et al., 2015), these processes seem to be minor factors controlling the levels of FOM_{vis} in the upper 500 m except in the surface water along the $143^{\circ}E$ line.

The distributional pattern of FOM_{vis} at depths of 20–60 m at $40^{\circ}N$ (Figure 2d) was characterized as a surface-layer water system (Figure 3); however, FOM_{vis} did not follow simple mixing patterns between low- FOM_{vis} subtropical waters (Kuroshio water and Tsugaru warm current water) and high- FOM_{vis} subarctic water (Oyashio water). Although the water mass at $40^{\circ}N$ exhibited more subtropical characteristics than that at $39^{\circ}N$, the FOM_{vis} level at depths of 20–60 m at $40^{\circ}N$ was greater than that at $39^{\circ}N$, indicating that the high FOM_{vis} level at depths of 20–60 m at $40^{\circ}N$ could not be explained by simple mixing or warming of Oyashio water. The other possibility for the water mass at $40^{\circ}N$ is the contribution of river water with high FOM_{vis} levels to the Tsugaru warm current water. However, low-density river water can mix directly with a lighter fraction of Tsugaru warm current water. Thus, the occurrence of local sources of FOM_{vis} , namely, local production and/or lateral transport of water masses

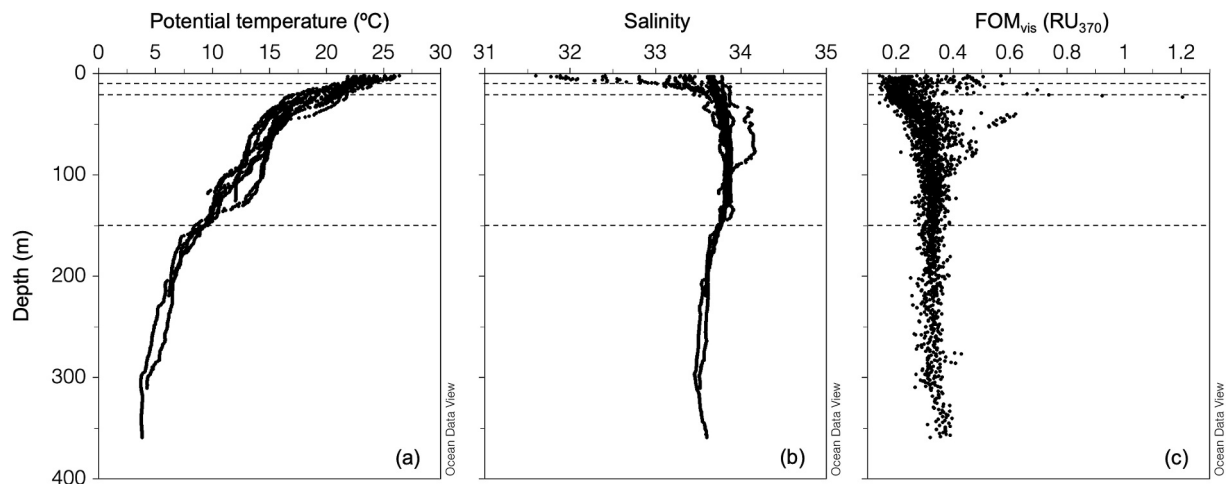


Figure 4. Vertical profiles of (a) potential temperature, (b) salinity, and (c) FOM_{vis} in and around Sendai Bay, Japan. All the parameters were determined by sensors. The dotted lines in each panel represent 10, 20, and 150 m, respectively.

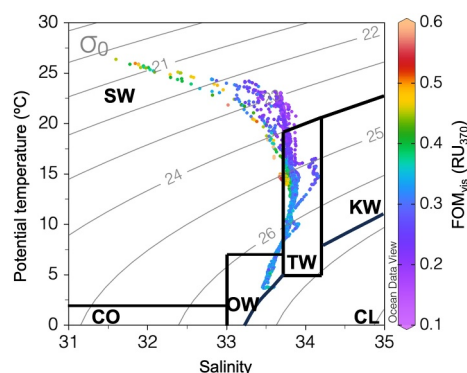


Figure 5. Potential temperature-salinity (T-S) diagram with FOM_{vis} in and around Sendai Bay, Japan. All the parameters were determined by sensors. The gray lines in the diagram indicate the contour lines of σ_θ . The six water systems are classified according to Hanawa and Mitsudera (1987) as follows. TW: Tsugaru warm current water system; OW: Oyashio water system; KW: Kuroshio water system; CL: cold lower-layer water system; SW: surface-layer water system; CO: Coastal Oyashio water system.

(Figure S3 in Supporting Information S1). The N^* values at 30 and 50 m depths at $40^\circ N$ (-4.8 and -5.3) were similar to those at $39^\circ N$, indicating that neither N^* nor FOM_{vis} at 30 and 50 m depths at $40^\circ N$ were controlled by the mixing of subtropical and subarctic waters and biological processes. The negative N^* is a signature of macronutrients influenced by denitrification in anoxic conditions, especially in anoxic sediments (Gruber & Sarmiento, 1997; Yoshikawa et al., 2006). Therefore, the potential temperature, salinity, N^* , and FOM_{vis} indicate that the water masses at depths of 20–60 m at $40^\circ N$ were likely influenced by water masses that were affected by anoxic sediments from coastal areas.

4.2. Factors Controlling the FOM_{vis} Level in and Around Sendai Bay

The FOM_{vis} levels at some stations in and around Sendai Bay (Figures 4c, 5, and 6), as an example of the Japanese coast, were considerably greater than those observed in the $143^\circ E$ line (Figures 2d and 3) and were likely derived from two local sources. The first source of high levels of FOM_{vis} was found in the surface waters at nearshore stations in Sendai Bay and was related to low-salinity water (Figures 5, 6 and Figure S1 in Supporting Information S1). A negative relationship was found between salinity and FOM_{vis} in this source and the high-salinity endmember (dotted line in Figure 6), indicating the dilution of riverine FOM_{vis} with river water and seawater mixing. The levels of visible fluorescence of DOM in river water are usually greater than those in seawater (Blough & Del Vecchio, 2002). In fact, several rivers (e.g., the Old Kitakami, Naruse, and Abukuma Rivers; Figure 1) flow into Sendai Bay. We collected FOM_{vis} samples from these rivers in the summer of 2022, and the FOM_{vis} levels were ~ 10 times greater than those in the seawater in Sendai Bay.

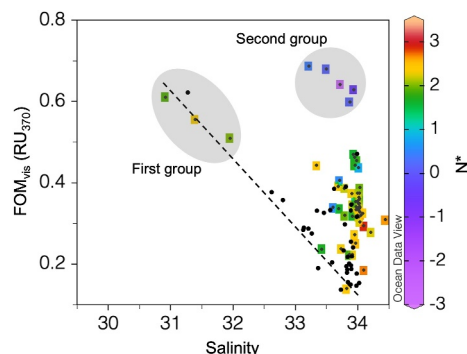


Figure 6. The relationship between salinity and FOM_{vis} with N^* in and around Sendai Bay, Japan. The data obtained by chemical analyses of water samples collected from buckets and Niskin bottles were used for the figure. The data for which N^* was below the detection limit are plotted with no color. The dotted line shows the dilution line of the riverine FOM_{vis} with the mixing of river water and seawater.

from coastal areas, which are not directly affected by river water, might be responsible for the high FOM_{vis} level at depths of 20–60 m at $40^\circ N$.

FOM_{vis} is produced by phytoplankton and heterotrophic prokaryotes. If FOM_{vis} is produced by phytoplankton during a bloom, an increase in FOM_{vis} accompanies an increase in DO concentration (a decrease in AOU) due to photosynthesis. The AOU at the 20–60 m depth at $40^\circ N$, below the surface mixing layer, was greater than that at $39^\circ N$, indicating that the high levels of FOM_{vis} at $40^\circ N$ were most likely not derived from phytoplankton. Postbloom microbial degradation may decrease the DO concentration (increase AOU) and result in the production of FOM_{vis} . Thus, it is possible that phytoplankton blooms and subsequent microbial degradation after the mixing of subtropical and subarctic waters cause high levels of FOM_{vis} at depths of 20–60 m at $40^\circ N$. This possibility can be confirmed with N^* because N^* in subtropical water is greater than that in subarctic water (Deutsch et al., 2001), and N^* essentially does not change with biological uptake (except in the N-fixation process) or microbial remineralization (Gruber & Sarmiento, 1997). At depths of 30 and 50 m, the N^* values were below the detection limit and were 0.0 and 0.5 in water masses with subtropical characteristics at $38^\circ N$ and $41^\circ N$ but were -3.1 and -4.1 in water masses with subarctic characteristics at $39^\circ N$

The second group of high FOM_{vis} values were found in the subsurface layer and could not be categorized by potential temperature or salinity (Figure 5 and Figure S2 in Supporting Information S1). This group can be characterized by high FOM_{vis} levels with high salinity and near zero and negative N^* (Figure 6). A negative N^* is the result of denitrification in anoxic environments (Gruber & Sarmiento, 1997). In Sendai Bay, it was observed that macronutrients were supplied from sediments in summer and fall due to low DO concentration and high water temperature (Ishikawa et al., 2023). Visible fluorescent DOM has been reported to transport from anoxic sediments to the overlaying water but not from the oxic sediments due to efficient trap via sorption onto iron oxyhydroxides (Skoog et al., 1996). The production of visible fluorescent DOM in anoxic sediments has also been reported (Chen et al., 2016). Thus, the source of FOM_{vis} in the second group with near zero and negative N^* was most likely derived from anoxic sediments in and around

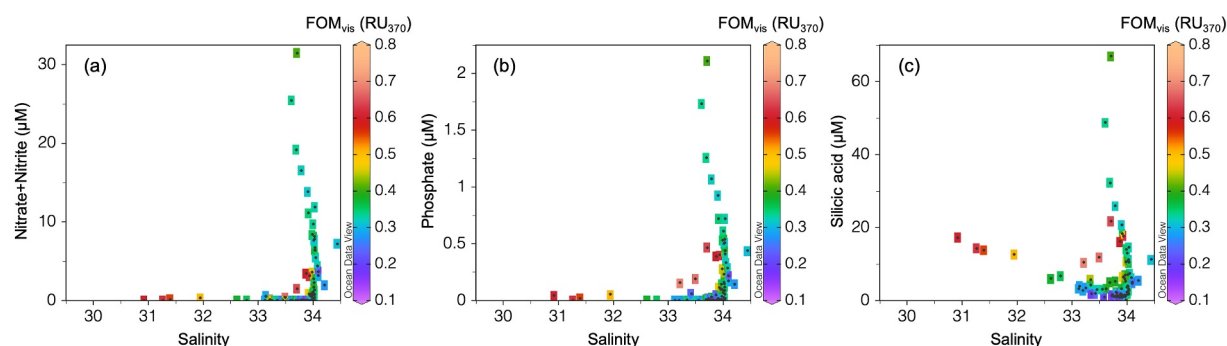


Figure 7. Relationships between salinity and (a) nitrate and nitrite concentration, (b) phosphate concentration, and (c) silicic acid concentration in and around Sendai Bay, Japan. The data obtained by chemical analyses of water samples collected from buckets and Niskin bottles were used for the figure.

Sendai Bay. In the second group, on the one hand, extremely high FOM_{vis} near the bottom at two stations were probably direct inputs of FOM_{vis} from the bottom sediments (Figure S2 in Supporting Information S1). On the other hand, high FOM_{vis} with a 10 m thick layer at a relatively deep station located outside Sendai Bay could be the result of lateral transport of sediment-derived FOM_{vis} from Sendai Bay.

4.3. FOM_{vis} as a Tracer for Sediment-Affected Water Mass From Coastal Areas

A link between high levels of visible fluorescent DOM as well as FOM_{vis} and negative N^* has recently been observed in dense shelf water in the Bering and Chukchi Seas and the Sea of Okhotsk, which was formed during sea-ice formation (Hioki et al., 2014; Yamashita et al., 2019; Yamashita & Nishioka, 2023). Sedimentary inputs of visible fluorescent DOM into water columns have also been suggested for other coastal regions (Kowalczyk et al., 2015; Reader et al., 2019). Using an in situ fluorometer, Loginova et al. (2016) observed FOM_{vis} input from sediment to the water column in a Peruvian coastal system. The sediment-derived visible fluorescent DOM has been reported to be laterally transported conservatively from the Sea of Okhotsk to the North Pacific with the circulation of intermediate water (Yamashita et al., 2020, 2021). The FOM_{vis} observations at the offshore ocean along 143°E and in and around Sendai Bay carried out in this study imply that FOM_{vis} -rich coastal subsurface water with anoxic sediment input is likely laterally transported to the offshore ocean.

The satellite data of sea surface temperature and CDOM (Figure S4 in Supporting Information S1) as well as the absolute dynamic topography and geostrophic current (Figure S5 in Supporting Information S1) indicate that the water mass with relatively high CDOM and low water temperature originated from the north, the coastal region of Hokkaido, was transported south by an anti-cyclonic circulation and stayed at 40°N, where the FOM_{vis} -rich subsurface water was observed in the 143°E line. Therefore, the source region of the FOM_{vis} -rich subsurface water was probably the coastal region of Hokkaido, Japan. The satellite data also showed that the FOM_{vis} -rich subsurface waters observed in and around Sendai Bay were likely transported to the offshore ocean (Figures S6 and S7 in Supporting Information S1). Mesoscale eddies are known to transport nutrient-rich water masses from coastal areas to offshore oceans (Ueno et al., 2023). These recent observational results, including those from this study, indicate that the visible fluorescent DOM and FOM_{vis} can be used to trace the transport of anoxic sediment-affected water masses from coastal areas to offshore oceans by nearshore ocean currents and associated mesoscale eddies.

Figure 7 shows the relationships between salinity and nutrient concentrations and FOM_{vis} levels in and around Sendai Bay. High levels of macronutrients (i.e., nitrate + nitrite, phosphate, and silicic acid) were observed in the subsurface layer in the offshore region (Figure 7 and Figure S8 in Supporting Information S1). Thus, in and around Sendai Bay, the upwelling of subsurface water from offshore regions is likely an important source of macronutrients (Nakajima et al., 2021). A negative relationship was observed between salinity and silicic acid but not between salinity and nitrate + nitrite or phosphate in waters with salinities less than 33. The riverine nitrate + nitrite and phosphate were likely consumed in low-salinity waters (<30), but a portion of the riverine silicic acid escaped biological consumption and was transported to high-salinity (>30) waters. Relatively high levels of phosphate and silicic acid with high levels of FOM_{vis} were observed in water masses with salinities ranging from 33 to 34, indicating that anoxic sediment is likely the third important source of phosphate, FOM_{vis}

and silicic acid due to the high sedimentation rate of organic matter and biogenic silica in and around Sendai Bay. Since such sediment-derived phosphate, silicic acid, and FOM_{vis} in the subsurface water in coastal regions, where photosynthesis and photodegradation of FOM_{vis} are less likely, can be transported to offshore oceans by near-shore ocean currents and associated mesoscale eddies, coastal sediment-derived nutrients can be traced via FOM_{vis} observations.

5. Conclusions

The links between sediment-derived nutrient signals and high levels of FOM_{vis} , which is affected by coastal anoxic sediment, were observed not only the Japanese coast (i.e., in and around Sendai Bay) but also in the Kuroshio-Oyashio transition zone. These observational results indicate that FOM_{vis} measurement by an in situ sensor can be a useful method for tracing the lateral transport of anoxic sediment-derived nutrients from coastal regions to offshore oceans. In situ sensors for FOM_{vis} observation can be installed on ship-based CTD systems (Coble et al., 1998; Shigemitsu et al., 2020; Stedmon et al., 2021; Yamashita et al., 2015, 2019) and also on other oceanographic observational tools, such as underwater pumping systems (Chen & Gardner, 2004; Coble et al., 1998), biogeochemical-argo floats (Organelli & Claustre, 2019; Xing et al., 2012, 2014), gliders (Hendry et al., 2021; Loginova et al., 2016), and buoys (Qu et al., 2020). The combination of FOM_{vis} observations by using in situ sensors, analysis of a tracer for sediment-water column exchange, and nutrient analyses can be used to quantitatively assess transport of sediment-derived nutrients from coastal regions to offshore oceans with high spatiotemporal resolution.

Data Availability Statement

All data used in this study are available at Hokkaido University Collection of Scholarly and Academic Papers (HUSCAP) via <http://hdl.handle.net/2115/91002> (Yamashita, 2024). The data set of absolute dynamic topography and geostrophic current were obtained from <https://doi.org/10.48670/moi-00148> (CMS, 2023). The data set of sea surface temperature and chromophoric dissolved organic matter were obtained from <https://gportal.jaxa.jp/gpr/notice/case/view/1092> (JAXA, 2021).

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