



Temporal variation of particulate organic carbon flux at the mouth of Tokyo Bay

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Abstract

A sediment trap experiment was conducted at a depth of 750 m at the mouth of Tokyo Bay to clarify the quantity and transport process of particles from the bay to the open ocean. The high total mass flux ($8.7 \pm 4.5 \text{ g m}^{-2} \text{ d}^{-1}$) suggests that the particles not only originate in the surface layer right above the trap, but are also focused in Uraga Channel and discharged into the bay mouth. The organic carbon and nitrogen isotope ratios ($\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}$) of the trapped particles were like those of the surface sediment in the bay, that is, a mixture of particles in rivers and suspended particles in the surface layer of the bay. Compared with the results of the experiment conducted in 1995–2002, the average total mass flux was reduced by 70% and organic carbon content was reduced by 50%. The $\delta^{13}\text{C}_{\text{org}}$ values of trapped particles were also lower than those observed in the previous experiment, indicating a lower contribution from surface-suspended particles with high $\delta^{13}\text{C}_{\text{org}}$ values in the bay. These results could partly reflect a decrease of the concentration of the suspended particulate carbon in the bay by half over 20 years. Another factor contributing to the decrease of the flux at the bay mouth would be that the intrusion of Kuroshio coastal water into the bay, which pushes particles out to the bay mouth, has not occurred in recent years.

Keywords Tokyo Bay · Sediment trap · Particulate organic carbon · Particulate nitrogen · Isotope ratios · Material transport

1 Introduction

Accounting for approximately 10% of the total ocean area, the coastal area is a biologically active area where 20–30% of the primary production of the entire ocean occurs (Wolast 1991). Significantly higher production in coastal areas is supported by the influx of terrestrial or human-derived materials that contain high concentrations of nutrients and other substances necessary for primary production. The inflowing materials are used for photosynthesis in coastal areas; these become particulate matter, and settle on the seafloor. Subsequently, they are decomposed and returned by wind

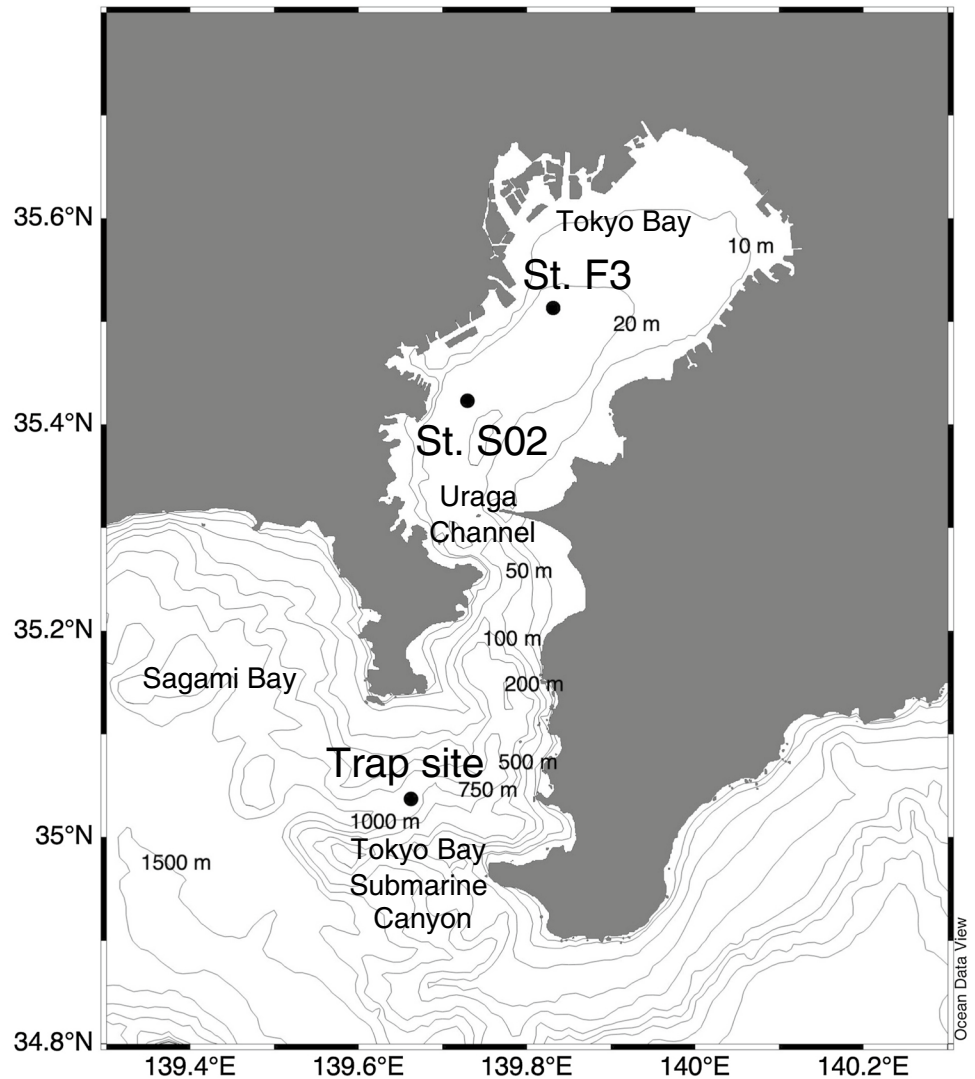
and tidal mixing to the surface, where they are once more used for photosynthesis. Material regeneration cycles are more rapid in coastal areas than in the open ocean (Bauer et al. 2013). Most of the inflow material flowed into the open ocean. Dissolved materials and suspended particulate matter flow out of the interaction between coastal and open ocean waters; however, particles that settle near the seafloor are thought to be gradually transported to deeper layers via the continental shelf slope by repeated resuspension owing to tides and residual currents in coastal areas. This process of transporting particles from coastal areas to the deep ocean via the bottom layer is considered to play a role in carbon sequestration in the ocean. The recent increase in human activity is thought to have considerably altered the coastal environment, affecting primary production and ecosystems. This material transport process is expected to change in response to environmental changes.

Tokyo Bay (Fig. 1), which is surrounded by some of the most densely populated cities and industrial areas in Japan, is one of the most eutrophic coastal regions worldwide (Ishimaru 1991). Eutrophication in Tokyo Bay peaked in the early 1970s with the highest nutrient concentrations

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Fig. 1 Map of Tokyo Bay



(Furota and Nomura et al. 2011). The nutrient concentration in the bay has been decreasing since the 1990s owing to improvements in water supply, sewage systems, and sewage treatment technology (Kubo et al. 2019). However, red tides and hypoxic water masses, common in eutrophic coastal areas, have been frequently reported (Tokyo Metropolitan Government Bureau of Environmental Protection 2020). Hamana et al. (submitted in this issue) suggested that compared with those around 2000, the primary productivity and photosynthetic activity off Haneda in Tokyo Bay increased in 2018–2021. The number of reported red tide cases in Tokyo Bay has not decreased despite the decrease in nutrient loading, because nutrients are regenerated by the decomposition of organic matter deposited and used for photosynthesis by phytoplankton (Kajiyama 2019). Meanwhile, nutrient concentrations in Tokyo Bay have been decreasing since the 2000s (Kubo et al. 2019). Ando et al. (2021) also indicated that the area of hypoxic water mass decreased from 30% in 2007 to less than 15% in 2015, and this trend was

prominent on the east side of Tokyo Bay. These suggest that sediments containing organic matter in the bay are decreasing as they were decomposed and discharged into the open ocean as nutrient.

Some of the organic matter that flows into or is produced in Tokyo Bay is deposited in the bay, and some is discharged into the open ocean with seawater. The bottom layer of the Uruga Channel in the southern part of Tokyo Bay is a steep continental shelf slope toward Sagami Bay, and is called the Tokyo Bay Submarine Canyon (Fig. 1). As the suspended particles in the bottom layer of the bay approach the slope, some gradually sink to deeper layers along the slope. Yanagi et al. (1992) showed that this process is driven mainly by tides and named it the “tidal pump.” In contrast, in the middle and bottom layers (deeper than 30 m depth) of the Uruga Channel, when the Kuroshio Current approaches or leaves the bay, the Kuroshio coastal water penetrates the same density layer in Tokyo Bay and pushes out the surface and bottom waters from the bay (Yanagi et al. 1989; Hinata

et al. 2000, 2001). At this time, high temperature, low salinity, and high turbidity bay water are observed in the offshore surface layer (Uraga Channel), while low temperature and high turbidity bay water are observed in the bottom layer, suggesting that suspended particles and sediments flow out of the bay with water. A sediment trap experiment conducted in the late 1990s and the early 2000s at the mouth of Tokyo Bay demonstrated an average total mass flux of $30 \text{ g m}^{-2} \text{ d}^{-1}$ and average organic carbon flux of $1.3 \text{ g m}^{-2} \text{ d}^{-1}$ (Sukigara and Saino 2005, 2007). These values are tens to hundreds of times higher than fluxes observed at the same depth in other areas, suggesting that the quantity of particles discharged from Tokyo Bay to the middle and deep oceans is large. Tokyo Bay is probably a coastal area with a mechanism for efficiently discharging particulate organic matter produced in the bay into the deep open ocean.

This study aimed to clarify the transport process of particulate matter from Tokyo Bay to the open ocean. Particles collected by a sediment trap experiment at the mouth of Tokyo Bay since 2018 were investigated for their records by analyzing the organic carbon and nitrogen isotope ratios of the particles. By comparing the results with those of previous studies, we discuss the effect of the recent decrease in organic matter storage in Tokyo Bay owing to a decrease in nutrient concentrations on the transport of particulate organic carbon from the bay to the middle and deep layers in the open ocean.

2 Materials and methods

2.1 Sediment trap experiment

A sediment trap (SMD26S–6000, Nichiyu Giken Kogyo, Japan) with a collection area of 0.0079 m^2 was deployed at the mouth of Tokyo Bay ($35^\circ 02.20 \text{ N}$, $139^\circ 38.91 \text{ E}$, and water depth of 850 m, Fig. 1) from December 2018 to March 2021. The trap mooring system was deployed and retrieved by the training vessel (T/V) *Seiyo Maru* of the Tokyo University of Marine Science and Technology. Trap sampling was interrupted for three months, from July to September 2020, owing to COVID-19 infection control on the observation ship. The trap was placed 100 m above the seabed to avoid resuspended sediments from entering. The trap had 26 collecting cups (volume of 250 mL) rotating every seven days. Each cup was filled with filtered seawater containing 10% neutral formaldehyde to prevent the biological decomposition of organic matter. After recovering the sediment trap, the particles were gently mixed and a portion of the particles (10 mL) was sampled for microscopic observation. In the laboratory, sediment trap samples were divided into three parts: half for major component analysis, a quarter for organic

analysis, and a quarter for archiving. This sediment trap experiment was conducted with exactly the same settings as the previous experiment (Sukigara and Saino 2005, 2007; Takahashi and Noriki 2007).

After the collection of sediment trap, zooplankton in the trap samples were removed under microscope as “swimmer.” Then, the samples were filtered with membrane filters (Cytiva Life Sciences, USA or Millipore, USA) with a pore size of $0.6 \mu\text{m}$, which was previously weighed and washed with pure water after filtration. The particles with the filters were dried at 40°C , and the weight was measured after sufficient cooling to calculate the total mass flux (TMF).

2.2 Collection of suspended particles

Suspended particles were collected in July, August, and December 2019 and January 2020 at two stations (F3, $35^\circ 30.42 \text{ N}$, $139^\circ 49.48 \text{ E}$, and water depth of 26 m; S02, $35^\circ 25.42 \text{ N}$, $139^\circ 43.81 \text{ E}$, and water depth of 35 m; Fig. 1) at depths of 5 m from the surface and 3 m from the bottom in Tokyo Bay. Water samples of 2 to 6 L were filtered using pre-combusted GF/F Whatman 47 mm glass fiber filters (Cytiva Life Sciences, USA) and washed with pure water. The particles with the filters were stored in a freezer until analysis.

2.3 Analysis of major components of particulate matter

Sinking particles collected on the membrane filter were rinsed with deionized water during filtration and dried under vacuum at 60°C . The following analytical methods are the same as those used in the previous study (Takahashi and Noriki 2007). After the weight measurement of the dried samples to calculate the total mass flux, the particles were crushed and sealed in a plastic container. An aliquot ($\sim 60 \text{ mg}$) of the powdered samples was decomposed with a mixed acid solution in a Teflon-sealed vessel, and the concentration of silicon (Si) in the decomposed solution was measured by molybdenum yellow colorimetry (Noriki et al. 1980). The decomposed solution was heated and evaporated, the dried product was dissolved in nitric acid, and the concentrations of aluminum (Al) and calcium (Ca) were measured by inductively coupled plasma (ICP) mass spectrometry (Agilent 7700, Agilent Technologies, Inc., USA). Concentrations of lithogenic matter and biogenic carbonate were calculated using Eqs. (1 and 2), assuming that the Al and Ca concentrations of the lithogenic material were 8.2% and 0.89% (Takahashi and Noriki 2007), respectively.

$$\text{Lithogenic matter content} = [\text{Al}] / 8.23 \times 100 \quad (1)$$

$$\text{Biogenic carbonate content} = ([\text{Ca}] - 0.89 / 8.23 \times [\text{Al}]) \times 100 / 40 \quad (2)$$

where [Al] and [Ca] are the concentrations of Al and Ca in the trapped particles, respectively.

The loss-of-ignition method was used to determine the organic matter content. In this method, samples were heated in a muffle furnace at 500 °C for 24 h. When an adequate number of the samples were obtained, sample analysis was conducted in duplicate, and the precision was within 5%. As reference materials of elemental composition, JG-1a (granodiorite) and JLK-1 (lake sediment) from National Institute of Advances Industrial Science and Technology, Japan were analyzed. The concentrations obtained using this method coincided with certified values within 6%.

Concentrations of biological silicate (opal) were calculated assuming that the difference between the concentrations of total Si and the lithogenic Si is the biogenic Si concentration, and the relative water content in the biogenic opal was 10% (i.e., biogenic opal = $\text{SiO}_2 \cdot 0.4\text{H}_2\text{O}$) (Mortlock and Froelich 1989). Lithogenic Si was calculated based on the Al concentration as lithogenic Si:Al = 58.8:8.23 (Takahashi and Noriki 2007).

2.4 Analysis of organic components of particulate matter

The dried trapped particles were removed from the filter and homogenized by grinding in an agate mortar and pestle to measure the organic components. To analyze the organic carbon content and isotope ratio, approximately 20 mg of the dried samples were fumigated with hydrochloric acid for 12 h to remove particulate carbonate and dried in a vacuum oven. The organic carbon and nitrogen contents of trapped particles, particulate organic carbon and nitrogen concentrations of suspended particles, and isotopic compositions ($\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$) of trapped and suspended particles were determined using a continuous flow isotope mass spectrometer (Delta PLUS, Thermo Fisher Scientific, USA) fitted with an elemental analyzer (NC-2000, CE Instruments, UK) with a ConFlo II (Thermo Fisher Scientific, USA). The analytical precision based on the replicate analyses of $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ was $\pm 0.2\text{‰}$. For the analysis of organic components, we used different machines (Finnigan MAT 252, Thermo Fisher Scientific, USA and another NC-2000, CE Instruments, UK) in the previous study (Sukigara and Saino 2005), but used the same standards to confirm isotope ratios and masses of organic carbon and nitrogen.

3 Results

3.1 Trapped particles

The values of particulate TMF collected at the mouth of Tokyo Bay varied from $3.0 \text{ g m}^{-2} \text{ d}^{-1}$ (October 2020)

to $29.3 \text{ g m}^{-2} \text{ d}^{-1}$ (August 2019), with a mean value of $8.7 \pm 4.5 \text{ g m}^{-2} \text{ d}^{-1}$ (Fig. 2a). The temporal variation in TMF tended to be high (mean value of $9.3 \text{ g m}^{-2} \text{ d}^{-1}$) in autumn and winter (September to February) and low (mean value of $7.6 \text{ g m}^{-2} \text{ d}^{-1}$) in spring and summer (March to August). As for the four major components of trapped particles, more than 80% were lithogenic matter, and the next largest component was organic matter ($10.5 \pm 1.1\%$) (Table 1). The mean and standard deviation for opal and calcium carbonate were $9.5 \pm 2.7\%$ and $5.8 \pm 0.8\%$, respectively.

The organic carbon particle flux, which was calculated by multiplying the TMF and organic carbon content, varied from $90 \text{ mg C m}^{-2} \text{ d}^{-1}$ (April 2019) to $660 \text{ mg C m}^{-2} \text{ d}^{-1}$ (August 2019), with a mean value of $230 \pm 110 \text{ mg C m}^{-2} \text{ d}^{-1}$. The temporal variation in the organic carbon flux was similar to that of TMF (Fig. 2b). Particulate nitrogen flux varied from $12 \text{ mg N m}^{-2} \text{ d}^{-1}$ (April 2019) to $105 \text{ mg N m}^{-2} \text{ d}^{-1}$ (August 2019), with a mean value of $32 \pm 16 \text{ mg N m}^{-2} \text{ d}^{-1}$ (not shown).

The organic carbon content of trapped particles varied from 2.3% (August 2019) to 3.8% (November 2020), with a mean value of $2.7 \pm 0.3\%$ (Fig. 2c). The nitrogen content of trapped particles varied from 0.31% (February 2020) to 0.61% (January 2021) with a mean value of $0.38 \pm 0.05\%$ (not shown).

The values of $\delta^{13}\text{C}_{\text{org}}$ of the trapped particles varied from -24.7‰ (July 2020) to -21.6‰ (July 2019). The weighted average of $\delta^{13}\text{C}_{\text{org}}$ over the observation period was -22.4‰ (Fig. 2d); from December 2018 to September 2019, the values were approximately -22.5‰ ; however, from October 2019 to June 2020, they continued to decrease to about -24.5‰ . By the end of September 2020, it rose to -22.0‰ , but then continued to decline until February 2021.

The values of $\delta^{15}\text{N}$ of trapped particles varied from 5.1‰ (April 2019) to 7.7‰ (January 2021), with a weighted average of 5.6‰ over the observation period (Fig. 2e). The $\delta^{15}\text{N}$ showed a relatively distinct seasonal variation throughout the observation period, with a low value ($\sim 5.0\text{‰}$) from February to April, followed by an increase for 1–2 months, high values ($\sim 6.3\text{‰}$) from summer to autumn, and a decrease from approximately November.

3.2 Suspended particles

The organic carbon and nitrogen concentration of suspended particles in the surface layer in Tokyo Bay were high in the summer ($1.07 \pm 0.53 \text{ mg C L}^{-1}$, $0.21 \pm 0.10 \text{ mg N L}^{-1}$) and low in the winter ($0.23 \pm 0.07 \text{ mg C L}^{-1}$, $0.05 \pm 0.01 \text{ mg N L}^{-1}$) (Table 2).

The $\delta^{13}\text{C}_{\text{org}}$ values of suspended particles in the surface layer of the bay varied from -22.9‰ (January 2020, S02) to -16.7‰ (July 2019, F3) (Fig. 3). Low $\delta^{13}\text{C}_{\text{org}}$ values ($< -21\text{‰}$) were observed in January 2020. The $\delta^{15}\text{N}$ value

Fig. 2 Time series records of flux and parameters collected by a sediment trap of the mouth of Tokyo Bay; **a** total mass flux, **b** organic carbon flux, **c** organic carbon content, **d** organic carbon isotope ratio ($\delta^{13}\text{C}_{\text{org}}$), and **e** nitrogen isotope ratio ($\delta^{15}\text{N}$)

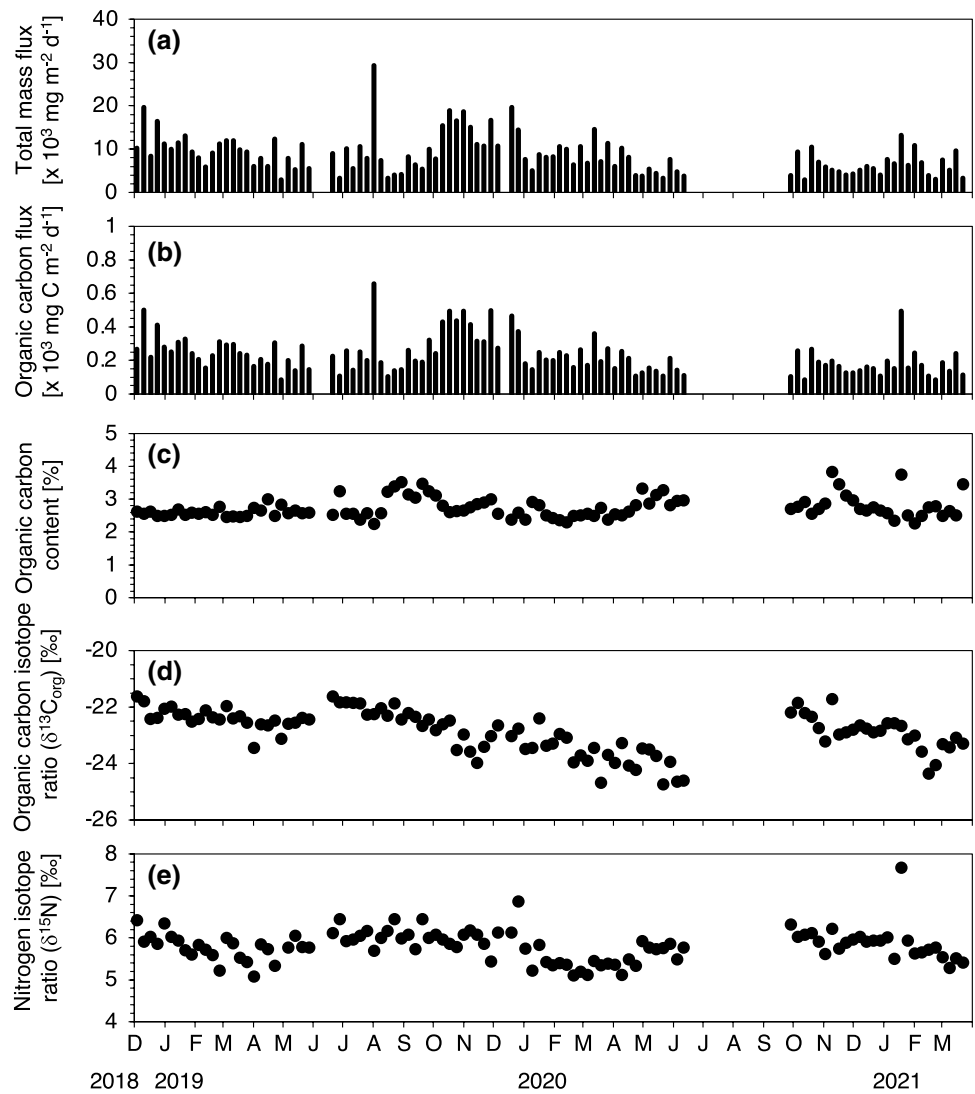


Table 1 Means of contents of major components in trapped particles collected at the mouth of Tokyo Bay

Sinking particles	Major components (%)			
	Lithogenic matter	CaCO ₃	Opal	Organic matter
Mean (<i>n</i> = 52)	80.2 ± 3.6	5.8 ± 0.8	9.5 ± 2.6	10.5 ± 1.1
(Min–max)	(73.7–89.0)	(0.9–6.9)	(4.8–15.2)	(7.6–12.7)

Table 2 Means of particulate organic carbon and nitrogen concentrations in suspended particles in the surface layer in Tokyo Bay

Suspended particles	POC concentration (mg C L ⁻¹)	PN concentration (mg C L ⁻¹)
Summer (July and August)		
Mean in 2001 (<i>n</i> = 4)	1.97 ± 0.42	0.35 ± 0.16
Mean in 2019 (<i>n</i> = 4)	1.07 ± 0.53	0.21 ± 0.10
Winter (December and January)		
Mean in 2000–2001 (<i>n</i> = 4)	0.50 ± 0.06	0.10 ± 0.02
Mean in 2019–2020 (<i>n</i> = 4)	0.23 ± 0.07	0.05 ± 0.01

of the suspended particles in the surface layer varied from 5.7‰ (January 2020, F3) to 13.8‰ (December 2019, F3) (Fig. 3). $\delta^{15}\text{N}$ values of approximately 10 were observed in July 2019 and January 2020 at S02.

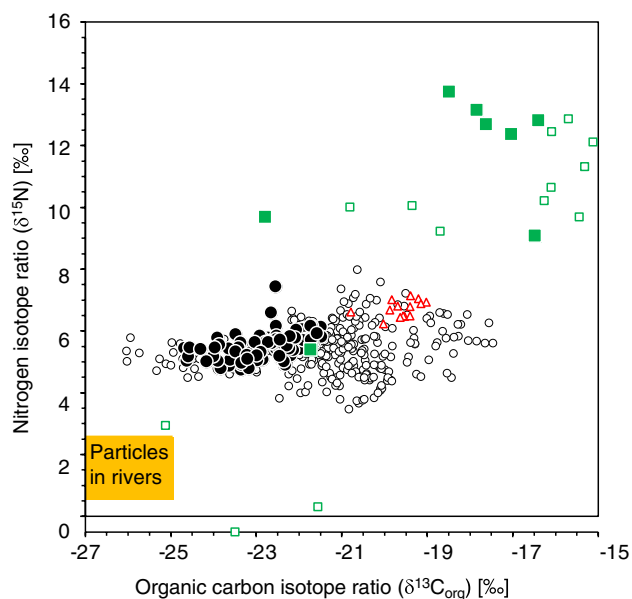


Fig. 3 Comparison of organic carbon and nitrogen isotope ratios ($\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$). Open circles: results of trapped particles at the mouth of Tokyo Bay during 1995 to 2002 (Sukigara and Saino 2005), closed circles (this study): results of trapped particles at the mouth of Tokyo Bay during 2018 to 2021, green open squares: results of suspended particle in the surface layer in the Tokyo Bay during 2000 to 2001, green closed squares (this study): results of suspended particle in the surface layer in the Tokyo Bay during 2019 to 2020, red open triangles: results of surface layer of sediment in the Tokyo Bay during 2000–2001, orange box: the $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ variation range of particles in rivers ($\delta^{13}\text{C}_{\text{org}}$: -27 to -25‰ , $\delta^{15}\text{N}$: 1 to 3 , Ogawa et al. 1994; Kubo and Kanda 2017)

4 Discussion

4.1 Origins and transport processes of trapped particles at the mouth of Tokyo Bay

The TMF of trapped particles collected by the sediment trap at the mouth of Tokyo Bay was substantially higher than that of the trap experiments at similar depths in other areas (Table 3). Reported TMF in the sediment trap experiments conducted in the seas around Japan were $33.7 \pm 45.5 \text{ mg m}^{-2} \text{ d}^{-1}$ at 500 m depth in the western North Pacific subtropical gyre (Mino et al. 2020), $114 \pm 140 \text{ mg m}^{-2} \text{ d}^{-1}$ at 500 m depth in the western North Pacific subarctic gyre (Mino et al. 2016), and $150\text{--}450 \text{ mg m}^{-2} \text{ d}^{-1}$ at 1000 m depth in the Japan Sea (Otsuka et al. 2004), and were one several hundredths to one several tenths of the TMF at 750 m depth at the mouth of Tokyo bay observed in this study ($8700 \pm 4500 \text{ mg m}^{-2} \text{ d}^{-1}$). Sediment trap experiments conducted in highly productive seas (e.g., the Arabian Sea, the Cariaco Basin, the Gulf of California, and East Alboan Gyre; Honjo et al. 1999; Thunell 1998; Thunell et al. 2000; Müller-Karger et al.

2004; Sanchez-Vidal et al. 2004), relatively high carbon fluxes ($10\text{--}40 \text{ mg C m}^{-2} \text{ d}^{-1}$) at depths of 500–1000 m were reported, which were one-tenth of those at the mouth of Tokyo Bay. Large TMFs have been reported in the sediment trap experiments at similar depths and distances from the seabed at the edge of the continental shelf as this study (Hung et al. 1999; Giraudeau et al. 2000). In the southern East China Sea, larger TMFs were observed but carbon fluxes were similar to those in this study.

Generally, the magnitude of organic carbon flux at a certain depth depends on the primary productivity in the euphotic layer. There are no reports on the productivity at the mouth of Tokyo Bay, but the primary productivity, $14\text{--}163 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ($168\text{--}1956 \text{ mg C m}^{-2} \text{ d}^{-1}$) (Hashimoto et al. 2005) was reported in the central part of Sagami Bay adjacent to Tokyo Bay (Fig. 1). To estimate the carbon flux this productivity would provide, the equation proposed by Pace et al. (1987), which calculates particulate organic carbon (POC) flux at any given depth from the primary productivity, was used:

$$\text{POC flux } (z) = 3.523 \times z^{-0.734} \times \text{primary productivity} \quad (3)$$

where z is the depth. The POC flux from the surface layer at a depth of 750 m at the mouth of Tokyo Bay was estimated $4.6\text{--}53.5 \text{ mg C m}^{-2} \text{ d}^{-1}$. This estimated value is lower than the observed values ($230 \pm 110 \text{ mg C m}^{-2} \text{ d}^{-1}$), indicating that the trapped particle originate from not only the primary productivity in the euphotic layer at the bay mouth but also other sources, and the contribution of allochthonous particles is significant.

One possible location supplying a large quantity of organic matter to the bottom layer at the bay mouth is Tokyo Bay, which has high primary production ($900\text{--}16,400 \text{ mg C m}^{-2} \text{ d}^{-1}$; Yamaguchi 1993; Hamana et al. submitted this issue). The POC flux at a depth of 20 m in the inner bay of Tokyo Bay observed by a sediment trap experiment was reported to be $750 \text{ mg C m}^{-2} \text{ d}^{-1}$ (Sasaki et al. 1989). In this study, we estimated the POC flux at a depth of 750 m using Eq. (4), assuming that the POC flux decreases with depth according to Martin's model: (Martin et al. 1987),

$$\text{POC flux } (z) = \text{POC flux } (z_0) \times (z/z_0)^{-b} \quad (4)$$

where z and z_0 ($= 100 \text{ m}$) are the depth, POC flux (z), and POC flux (z_0) area POC fluxes at depths of z and z_0 , respectively, and exponential b is a dimensionless scaling factor that indicates the magnitude of flux attenuation with depth. It is known that b varies with location and season (e.g., 0.5 in the Northwest Pacific subarctic gyre to 1.6 in the North Atlantic subtropical gyre, Lamborg et al. 2008; Marsay et al. 2015). Using this equation and the POC flux at 20 m in

Table 3 Observed total mass fluxes and organic carbon fluxes collected by sediment trap experiments

Region	Latitude (+°N, −°S)	Longitude (+°E, −°W)	Period	Depth of trap (m)	Total mass flux (mg m ^{−2} d ^{−1})	Organic carbon flux (mg C m ^{−2} d ^{−1})	References
Seas around Japan							
Japan Sea	+41.24	+132.35	2000–2002	1000	150–450		Otosaka et al. (2004)
Western North Pacific subarctic gyre (K2)	+47.0	+160.0	2010–2014	500	114 ± 140	6.8 ± 7.5	Mino et al. (2016)
Western North Pacific subtropical gyre (S1)	+30.0	+145.0	2010–2014	500	33.7 ± 45.5	3.5 ± 3.7	Mino et al. (2020)
Seas with high organic carbon fluxes							
Arabian sea	+17.4	+58.8	1994–1995	914		17.2	Honjo et al. (1999)
Cariaco	+10.5	−64.7	1995–2005	840		37.3	Thunell et al. (2000), Mul- ler-Karger et al. (2004)
North Central Pacific	+46.8	+162.1	1985–1986	700		33.7	Tsunogai and Noriki (1991)
Gulf of California	+27.9	−111.7	1990–1997	500		18.9	Thunell (1998)
East Alboan Gyre	+36.3	−1.5	1997–1998	645		26.4	Sanchez Vidal et al. (2004)
Continental margins							
Southern East China Sea	+25	+122	1994–1995	900 ^a	11,890–15,730	200–242	Hung et al. (1999)
Shelf edge off Walvis Bay	−23	+12.98	1989–1999	495 ^b	143–376	10.6–40.6	Giraudeau et al. (2000)
The mouth of Tokyo Bay	+35.04	+139.66	2018–2021	750 ^a	8700 ± 4500	230 ± 110	This study

^a100 m above the bottom^b50 m above the bottom

Tokyo Bay to calculate the POC flux at 750 m, the value was 2–123 mg C m^{−2} d^{−1} (using attenuation factors *b* of 0.5, 1.6). The higher observed POC fluxes in the bay mouth than the estimated values may be due to the convergence of inner-bay particles (whether of suspended particles or sediments origin) in the barrow Uruga Channel, which is the only outlet, as they were transported into the bay mouth.

The organic carbon and nitrogen contents of the surface sediment previously reported at Station F3 in Tokyo Bay were 3.4 ± 0.3% and 0.45 ± 0.04% (Sukigara and Saino, 2005), which are similar to the trapped particles at the bay mouth in this study (organic carbon: 2.7%, Fig. 2c, nitrogen: 0.38%). The mean values of δ¹³C_{org} and δ¹⁵N of the surface sediment (δ¹³C_{org}: −19.8‰, δ¹⁵N: 7.0‰) are between the values of suspended particles in the surface layer in Tokyo Bay (weighted means of monthly observation from September 2000 to August 2001, −17.1‰ and δ¹⁵N: 10.4‰) and reported values of particles in the rivers (δ¹³C_{org}: −27 to −25‰, δ¹⁵N: 1 to 3‰, Ogawa et al. 1994; Kubo and Kanda 2017) (Fig. 3). The difference in isotope ratios between particles in rivers and in the surface layer of the bay reflects the environment in which the organic matter is produced. Organic matter of terrestrial origin, which can easily utilize carbon dioxide (CO₂) and nutrients, has low isotope ratios, and those in the ocean, where CO₂ and nutrient concentrations vary greatly by photosynthesis, also vary with the concentration of substrates (Rau et al. 1989, 1991; François et al. 1993). Hence, the organic matter on the surface

sediment is considered a mixture of organic matter of land and sea surface origin. The mean values of δ¹³C_{org} and δ¹⁵N of particles collected at the bay mouth, both previously reported (δ¹³C_{org}: −21.3‰, δ¹⁵N: 5.7‰, Sukigara and Saino 2005) and measured in this study (δ¹³C_{org}: −22.4‰, δ¹⁵N: 5.6‰), were lower than those of the surface sediment in the bay (δ¹³C_{org}: −19.8‰, δ¹⁵N: 7.0‰). This could be owing to the decomposition of organic matter during the transport of particles from the bay to the bottom layer at the bay mouth (Saino 1993; Lehmann et al. 2002). These results indicate that organic matter in the trapped particles at the bay mouth, like the surface sediment in the bay, originated from particles in the river, which were produced on land, and also from suspended particles, which were produced in the surface layer of the bay. Particles on the surface sediment in the bay repeatedly deposited and resuspended in the bay and discharged to its mouth.

The variation ranges of δ¹³C_{org} and δ¹⁵N of trapped particles at the bay mouth were larger than those on the surface sediment in the bay (Figs. 2e, f and 3). This may be owing to the excessive addition of suspended particles in the surface layer of the bay with large seasonal variations in δ¹³C_{org} and δ¹⁵N (Sukigara and Saino 2005; Kubo and Kanda 2017) that sunk directly to the bottom of the bay mouth without mixing with the seabed sediment in the bay. During the summer, active photosynthesis in the surface layer of the bay reduces CO₂ and nutrient concentrations, raises the values of δ¹³C of CO₂ and δ¹⁵N of nitrogenous nutrients, and increases the

values of $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ of suspended organic matter in the surface layer. In winter, due to the depression of the photosynthesis rate and the supply of substrates for photosynthesis (CO_2 by cooling and nutrients by vertical mixing), the values of $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ of suspended organic matter in the surface layer become low. A portion of the suspended particles sink in the bay and enter the surface sediment, while another portion slowly sinks and discharges out of the bay. Even if the isotopic ratio of surface-floating particles has large seasonal variation, the particles trapped in the bay mouth may exhibit small seasonal variation owing to dilution with sediments with little seasonal variation. The existence of intact diatom cells, which are often observed in the bay, in the trapped particles by microscopic analysis supports that the trapped particles contain relatively fresh organic matter produced in the bay.

4.2 Comparisons with an experiment 20 years ago and factors of changes

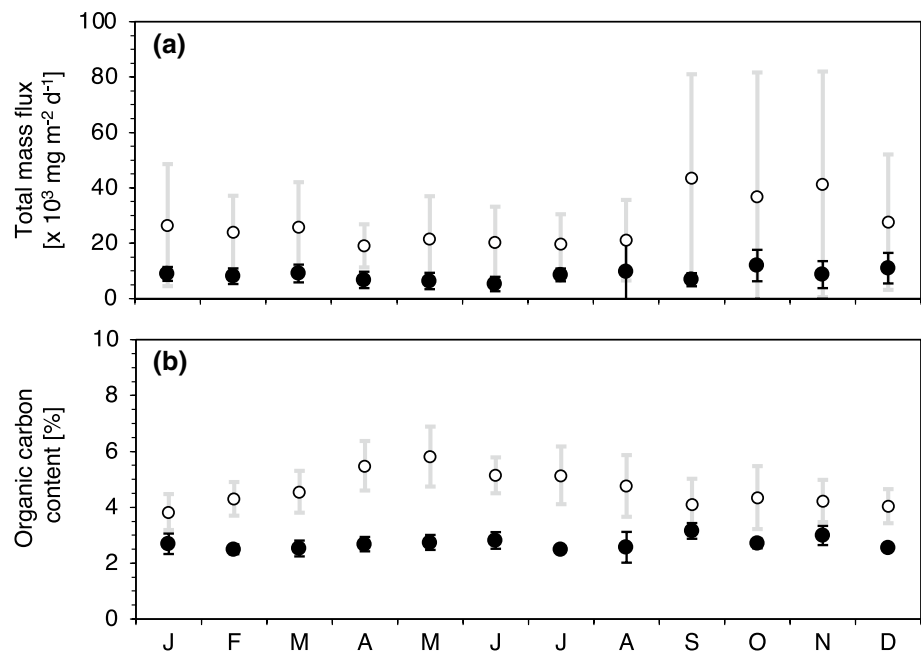
The fluxes and chemical compositions of trapped particles at the mouth of Tokyo Bay since 2018 were compared with those of a similar experiment conducted 20 years ago (Table 4). According to the comparison, the mean value of TMF in recent years was less than one-third of that of 20 years ago. The TMF range of variation in the past experiment was 3.3 to 226.7 $\text{g m}^{-2} \text{d}^{-1}$, but the recent maximum was one order of magnitude lower (29.3 $\text{g m}^{-2} \text{d}^{-1}$). A comparison of the mean values of TMF for each month in the two experimental periods shown in Fig. 4a revealed that the difference was large from September to November. In particular, high TMFs in the past experiment were observed from September to December 1999 and from December 2000 to January 2001. This is thought to be owing to the approach of the Kuroshio Current axis to Tokyo Bay, causing the intrusion of the Kuroshio coastal water near the bay mouth into the middle layer in the bay and discharging the bay water, suspended particles on the surface, and resuspended sediment from the bay to its mouth (Sukigara and Saino 2007). The approach and separation of the Kuroshio Current axis is one of the factors causing particles in the bay to discharge to the open ocean regardless of the season. Hinata et al. (2000, 2001) showed that the intrusion of Kuroshio coastal water into the bay also occurred during the winter and summer of 1998. Discharge events may have occurred frequently during the experimental period. In this study, however, the position of the Kuroshio current axis has hardly moved because of the large meander of the Kuroshio since 2017. This change in the Kuroshio Current axis is thought to be one of the factors reducing the particle flux at the bay mouth throughout the observation period since 2018.

Comparing the chemical composition of trapped particles (Table 4), the fraction of lithogenic materials and biogenic

Table 4 Comparison of flux and parameters of sinking particles between the previous (1995–2002) and present (2018–2021) experiments

Sinking particles	Total mass flux ($\text{mg m}^{-2} \text{d}^{-1}$)	Lithogenic matter (%)	CaCO_3 (%)	Opal (%)	Organic matter (%)	Organic carbon flux ($\text{mg C m}^{-2} \text{d}^{-1}$)	Organic carbon content	$\delta^{13}\text{C}_{\text{org}}$ (‰)	Nitrogen content	$\delta^{15}\text{N}$ (‰)
1995–2002 (Sukigara and Saino 2005; Takahashi and Noriki 2007)										
Mean	28,000 ± 26,500	66.8 ± 9.8	2.7 ± 1.9	15.0 ± 2.0	15.0 ± 2.0	1300 ± 1100	4.8 ± 1.0	− 21.3	0.57 ± 0.16	5.7
(Min–max)	(3300–226,700)					(300–8300)	(2.8–8.0)	(− 26.1 to − 17.8)	(0.26–1.13)	(3.9–8.2)
2018–2021 (this study)										
Mean	8700 ± 4500	80.2 ± 3.6	5.8 ± 0.8	9.5 ± 2.6	10.5 ± 1.1	230 ± 110	2.7 ± 0.3	− 22.4	0.38 ± 0.05	5.6
(Min–max)	(3300–29,300)	(73.7–89.0)	(0.9–6.9)	(4.8–15.2)	(7.6–12.7)	(90–660)	(2.3–3.8)	(− 24.7 to − 21.6)	(0.31–0.61)	(5.1–7.7)

Fig. 4 Monthly means of **a** total mass flux and **b** organic carbon content of trapped particles collected by a sediment trap at the mouth of Tokyo Bay. Open circles: results during 1995 to 2002, closed circles: results during 2018 to 2021. Lines from each symbol mean standard deviation



carbonate increased (lithogenic material: 67% to 80%, and biogenic carbonate: 2.7% to 5.8%) while the fractions of opal and organic matter decreased significantly (opal: 15.0% to 9.5%, organic matter: 15.0% to 10.5%). It should be noted that, while the fraction of lithogenic materials increased, the flux decreased. In Tokyo Bay, diatoms have been observed in abundance in all seasons (Nomura 1998), suggesting that the origin of opals in trapped particles is diatoms in the bay. The decreases in opal and organic matter contents indicate that the discharge of material produced in the bay to its mouth was greatly reduced. The organic carbon fluxes also decreased from a mean value of $1300 \text{ mg m}^{-2} \text{ d}^{-1}$ in the previous experiment to $230 \text{ mg m}^{-2} \text{ d}^{-1}$ at this time. Comparing the monthly means of organic carbon content showed that it has decreased in all months in recent years, and the seasonal variation has become smaller (Fig. 4b).

The detailed organic compositions of the previous and present studies were compared to investigate the factors responsible for the decrease in the organic carbon flux and contents of trapped particles. Comparing the $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ of the past and recent trapped particles in Fig. 3 and Table 4 shows that the recent data had a lower $\delta^{13}\text{C}_{\text{org}}$ and a smaller range of variation in $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ than the past data. Because the $\delta^{13}\text{C}_{\text{org}}$ of suspended particles in the bay has not changed much between the past and recent years, it can be inferred that the reason for the lower $\delta^{13}\text{C}_{\text{org}}$ of trapped particles in recent years is that the contribution of particles in the surface of the bay has decreased. The reduction in the variation range of $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ in the recent period also indicated that compared with the past period, the discharge amount of suspended organic matter in the bay decreased. A plausible explanation for the decreasing

contribution of particles originating in the surface layer of the bay to the trapped particles in the bottom layer of the bay mouth is that the concentration of suspended particles in the bay has been decreasing. During the previous experimental period (2000 to 2001), the POC concentration in the surface layer of the bay was 2.0 mg C L^{-1} in the summer and 0.5 mg C L^{-1} in the winter; however, recent results were reduced by half (Table 2). A similar change was observed in the PN concentration. The decrease in the amount of particulate organic matter in the bay may have altered the ratio of terrestrial to marine organic matter and reduced the carbon flux of the trapped particles at the bay mouth. Kubo et al. (2019) showed that nutrient concentrations in Tokyo Bay have decreased due to the regulation of anthropogenic nutrient discharge, which has been a cause of eutrophication in Tokyo Bay. Ando et al. (2021) reported that the area anoxic water masses has been decreasing in late summer in Tokyo Bay. These findings suggest a decrease in the ability of particle production in the bay. A decrease in the organic carbon and nitrogen concentrations of suspended particles in the bay would reflect the change of environment in Tokyo Bay.

5 Conclusion

The TMF and organic carbon fluxes of the trapped particles collected in the bottom layer of the mouth of Tokyo Bay were tens to hundreds of times higher than those at the same depth in the open ocean. These high fluxes indicate that the particles in Tokyo Bay are focused on the narrow Uruga Channel and discharge to the bay mouth. Analysis of the organic components of the trapped particles revealed

that they originated from the surface sediment in the bay, which is a mixture of particles from the river and suspended particles in the surface layer of the bay. The seasonal variations observed in $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ of trapped particles may reflect the large seasonal variation in suspended particles in the surface layer of the inner bay.

Comparing the results of this study (2018–2021) and the previous study (1995–2002) demonstrated substantial differences in fluxes: compared with 20 years ago, the mean value of TMF was reduced by 70%, and that of organic carbon content was reduced by 50%. Temporal changes in the chemical composition of the trapped particles showed a decrease in the fraction of biogenic materials (organic matter and opal) and an increase in the fraction of lithogenic materials. Compared with previous years, the $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ analyses of trapped particles revealed a decrease in the discharge amount of organic matter originating in the surface layer of the bay. The fact that POC and PN concentrations in the surface layer in the bay have been halved in recent years supports a decrease in organic matter discharged to the bay mouth. Another possible factor reducing the transport of particles in the bay to the bay mouth is the Kuroshio Current meandering. The Kuroshio Current Large Meander, which has been continuing since 2017, has stagnated the Kuroshio axis around the mouth of Tokyo Bay. This is likely to reduce the number of events in which Kuroshio coastal water enters the bay and pushed particles in the bay to the bay mouth. This weakening of the particle transport process that causes large outflows of particles from the bay to the bay mouth, regardless of the season, would have contributed to the decrease of particle fluxes at the bay mouth as well as the decrease in the particle concentration in the bay. However, to clarify the influence of the Kuroshio Current, conducting a similar trap experiment after meandering has ended will be necessary.

Coastal areas facing large cities, such as Tokyo Bay, have high primary productivity owing to the inflow of anthropogenic materials. The steep slope at the bay mouth efficiently transports particles from the bay to the middle and deep layers of the open ocean. This may supply a large quantity of organic matter to the deep layers at Sagami Bay and the Tokyo Bay Submarine Canyon and support the ecosystem there. However, the decrease in the amount of organic matter transported, as revealed in this study, may be beginning to impact the benthic ecosystem. Furthermore, a decrease in the transported particulate organic carbon from the coastal zone to the deep ocean would also reduce the efficiency of carbon sequestration. In the future, when organic matter and nutrient discharges are regulated to improve water quality in coastal areas, the amount of carbon transported from the coastal region to the open ocean mesopelagic layer will decrease, which may affect carbon absorption into the ocean. To monitor changes in ecosystems and material transport

in coastal regions, coastal observations such as those conducted by the T/V *Seiyo Maru* are required worldwide in the future.

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