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Carbon cycling in the deep Mariana Trench in the western north Pacific Ocean: Insights from radiocarbon proxy data

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ABSTRACT

The Mariana Trench in the western North Pacific (NP) contains the deepest (11,000 m) point in the ocean and thus remains the least explored environment. Carbon cycling in the deep Mariana Trench is largely unknown and remains a curious mystery. Here, we report radiocarbon (Δ^{14} C) and stable carbon isotope (δ^{13} C) compositions of dissolved inorganic and organic carbon (DIC and DOC) and sinking particulate organic carbon (POC) for samples collected at 2000 m, 4000 m and 6000 m from the Mariana Trench in the western NP. DOC had much lower Δ^{14} C values (-189% to -538%) than DIC (38% to -236%) and POC (11%–38%) at the same depths, indicating the different cycling processes and time scales of different carbon pools. The 14 C ages of DOC were approximately 1700 years and 4400 years older than those of DIC in the surface (2 m) and deep (10,000 m) waters of the Mariana Trench. Both DOC and DIC Δ^{14} C values at hadal depths (>6000 m) in the trench are comparable to values at other sites in the deep NP, suggesting that water movement and mixing at hadal depths in the Mariana Trench are active and follow the deep-water circulation patterns of the NP. POC fluxes at the three deep depths showed large variations, but their modern Δ^{14} C values indicate that the effective biological pump that drives fresh surface-produced particles to sink rapidly to deep depths in the trench could provide essential food sources for microbial and benthic communities living in an extremely harsh environment of the Mariana Trench.

1. Introduction

Carbon cycling at hadal depths (>6000 m) in the ocean, mostly in trenches, could be an important process linked to the global carbon cycle, but due to the sampling difficulties in the Challenger Deep, it remains the least studied environment, and little information is available. The Mariana Trench, located in the western equatorial North Pacific (NP), with a water depth of \sim 11,000 m, is known as the deepest ocean trench on Earth. In recent years, as deep-sea sampling techniques have advanced, several studies have investigated the biosphere and biogeochemical processes at hadal depths in the Mariana Trench (Nunoura et al., 2015; Guo et al., 2018; Luo et al., 2018; Tian et al., 2018; Liu et al., 2019; Hand et al., 2020). Studies have shown that the

hadal environment harbors distinct microbial planktonic communities (Nunoura et al., 2015; Tarn et al., 2016; Liu et al., 2019). Using an in situ autonomous microprofiling system, Glud et al. (2013) reported that the rates of biological consumption of oxygen in the Challenger Deep of the Mariana Trench were even higher than those at a nearby 6000-m-deep site. They concluded that the elevated deposition of organic matter (OM) in the Challenger Deep of the Mariana Trench maintains intensified microbial activity at extreme pressures in the bottom of the trench (Glud et al., 2013; Luo et al., 2018). A recent study by Wang et al. (2019) reported that modern bomb ¹⁴C was detected in amphipod tissues collected at 11,000 m depth in the Mariana Trench, indicating a food supply from the newly produced surface OM. The results of these studies suggest that there must be an active and rapid supply of sinking

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particulate organic carbon (POC) from the surface to the hadal ocean to support the life of microbial communities living in extreme environments (Nunoura et al., 2015; Guo et al., 2018; Tian et al., 2018; Liu et al., 2019; Hand et al., 2020). However, no direct measurement of POC fluxes has been reported for the Mariana Trench.

Radiocarbon, a unique tracer of global carbon cycle dynamics, has been revealed as the most useful proxy of the different carbon pools, e.g., dissolved inorganic and organic carbon (DIC, DOC) and POC, in the ocean and has revealed the most accurate and credible information on the transformation and cycling time scales of each carbon pool in the ocean (Druffel et al., 1992, 2008; McNichol et al., 2000; Key et al., 2002; Hwang et al., 2010). Studies have shown that the average ¹⁴C age of DOC is 4000 years in the deep Atlantic and 6000 years in the deep Pacific (Williams and Druffel, 1987; Bauer et al., 1992; Druffel et al., 1992, 2019), which are much older than the average ¹⁴C ages of DIC in the deep water of the two oceans (600 years and 2000 years, respectively) (McNichol et al., 2000; Key et al., 2002; Druffel et al., 2008), indicating that DOC is cycled on much longer time scales than DIC in the deep ocean. In contrast, POC is cycled on much shorter time scales in the water column. Δ^{14} C measurements of POC have revealed that recently produced surface OM particles can sink rapidly (weeks to months) to the deep interior of the ocean, indicating the efficiency of the biological pump and particle sinking dynamics in the deep ocean (Druffel et al., 1992; Kim et al., 2017; Honda et al., 2000; Armstrong et al., 2002; Hwang et al., 2010).

Here, we present the first data set of radiocarbon and stable carbon isotopes measured for DIC, DOC and POC in samples collected from the Mariana Trench, in combination with the measurement of the concentrations of DIC and DOC and the fluxes of POC in the water column. As part of our efforts to explore the challenging depths of the ocean, these results provide the first information of carbon cycling dynamics at hadal depths in the Mariana Trench in the western NP.

2. Methods

2.1. Sample collection

Water samples were collected at site MT-1 (11,000 m, 11.3641°N, 142.9066°E) in the Mariana Trench in the western NP onboard R/V Dongfanghong-2 in December 2015 and February–March 2017 (Fig. 1). Water samples for DOC and DIC measurements were collected using 12 L Niskin bottles deployed on a rosette with a conductivity-temperaturedepth (CTD) device. Due to limited ship time in December 2015, water samples were collected at six depths (2, 1,000, 1,760, 3,700, 5,370, and 8730 m) only. After collection, water for DOC was transferred directly into a 1 L precombusted (at 550 °C for 4 h) glass bottle after the bottle was rinsed three times with in situ seawater. The water was filtered immediately onboard through precombusted (550 °C for 4 h) Whatman GF/F filters (0.7 µM pore size), acidified with super-highpurity 85% H_3PO_4 (Aladdin®) to pH=2 and preserved frozen until chemical analysis. Water for DIC was collected in precombusted 100 mL glass bottles with fine ground-glass stoppers using precleaned silicone tubing connected directly to the Niskin bottle. After adding 50 µL saturated HgCl₂ solution, the bottles were capped tightly with greasecoated, ground-glass stoppers and secured with rubber bands to ensure a gas-tight seal (McNichol et al., 1994; Ge et al., 2016). DIC samples were kept in the dark at room temperature for 2-3 weeks before

Sinking POC samples were collected using time series sediment traps (McLane Research Laboratories model PARFLUX Mark78H-21) deployed in a series connection at three depths (2000 m, 4000 m, and 6000 m) on bottom-tethered hydrographic moorings at station MT-1 in the Mariana Trench. The sediment traps were deployed for twelve continuous months from January to December 2016 and were recovered during the February–March 2017 cruise. Sample cups were filled with seawater and treated with 0.3% mercuric chloride as a preservative of

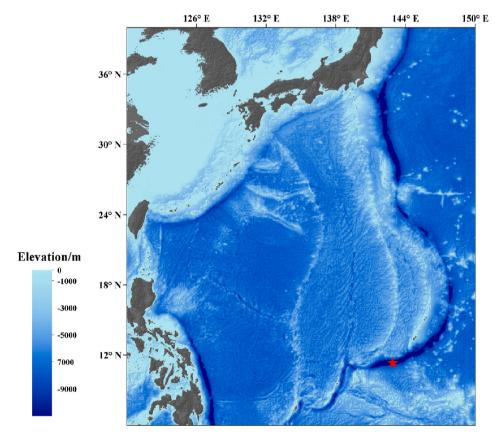


Fig. 1. Map showing the study area and sampling site (MT-1, 11.3641^o N, 142.9066^o E) in the Mariana Trench in the North Pacific during the 2015 and 2016 cruises.

POC (Hwang et al., 2015). The twelve sample cups were programmed to rotate every 30 days to collect over the 12-month period, but only nine cups from January to September were successfully collected, and the device failed for the last three months of collection. After recovery, sample cups were stored at 4 $^{\circ}$ C until further analysis. Along with the sediment trap deployment, single-point acoustic current meters (RCM Seaguard) were also mounted on the subsurface mooring at 1510 m, 3640 m, and 5770 m for continuous current measurements.

2.2. DOC, DIC and POC analyses

Concentrations of DOC were analyzed by the high-temperature catalytic oxidation (HTCO) method using a Shimadzu TOC-L analyzer. Potassium hydrogen phthalate (KHP) dissolved in high-purity Milli-Q water was used as the DOC standard. The quality assessment for DOC measurements was checked against reference low-carbon water and deep-sea water (supplied by Hansell Organic Biogeochemistry Laboratory at the University of Miami, USA). The instrumental blank was subtracted using high-purity Milli-Q water. The average blank of the DOC measurement was $\leq 3 \mu M$, and the analytic precision of triplicate injections was $\pm 4\%$. DIC concentrations were measured using a Shimadzu TOC-L analyzer in total inorganic carbon (IC) mode. Sodium carbonate and sodium bicarbonate dissolved in Milli-Q water were used as DIC standards, and the concentration values were checked against DIC reference deep-sea water for quality assessment (supplied by Dr. Dickson at Scripps Institution of Oceanography). The analytical precision was <3%. Sinking particles collected in sediment traps were filtered using a precombusted GF/F filter and were then freeze-dried. Total sinking mass fluxes (mg m⁻² d⁻¹) were determined based on the total particle weight (mg dry wt.) divided by the trap open area (m²) and trap deployment time (day). For POC measurement, freeze-dried particles were acidified with 10% HCl for 6 h, rinsed with Milli-Q water twice and dried again at 50 °C in an oven. The concentration of POC was then measured using a Thermo Flash 2000 CHN elemental analyzer with a precision of $\pm 2\%$. The flux of POC (mg C m⁻² d⁻¹) was then calculated based on the total mass flux (mg m $^{-2}$ d $^{-1}$) and POC content (% dry wt.) of the particles collected each month.

2.3. Carbon isotope analysis

Values of Δ^{14} C and δ^{13} C were measured for DIC, DOC and POC. For DIC, samples were first extracted as gaseous CO2 using our modified method (Ge et al., 2016) based on McNichol et al. (1994). Briefly, 50 ml seawater was injected into a pre-evacuated 100 ml borosilicate glass reactor with stripping probes in a N2-filled glove bag. After injecting 1.0 ml 85% H₃PO₄, the reactor with acidified seawater was placed in a hot water bath (70 °C) for 40 min, and the DIC (in CO2 form) was extracted on a vacuum line with an extraction efficiency >96% and low blank values (<3 μg C) (Ge et al., 2016). Purified CO₂ was collected cryogenically, measured and sealed in 6 mm OD Pyrex tubes for isotope analysis. For carbon isotope measurement of DOC, acidified water samples (500-600 ml) were first UV oxidized in a series of custom-made 100 ml quartz reaction tubes using a 1.2 kW medium-pressure mercury arc UV lamp (Hanovia Co.) for 5 h. Following UV irradiation, the CO2 generated from UV oxidation of DOC was purged with ultra-high-purity (UHP) helium gas through the vacuum line and collected cryogenically. The purified CO₂ was flame-sealed inside 6 mm OD Pyrex tubes for ¹³C and ¹⁴C analyses. The oxidation efficiency and blanks associated with the UV oxidation of DOC were tested using Milli-Q high-purity water and a DOC standard (oxalic acid) solution, yielding a high oxidation efficiency (\sim 95%) and relatively low (3–4 μg C) DOC. For POC isotope analysis, acidified sinking particles were oxidized in 9 mm OD quartz tubes (precombusted at 850 °C for 2 h) with CuO and Ag wire added and combusted at 850 °C for 2 h (Druffel et al., 1992). The resultant CO₂ was collected cryogenically on a vacuum line and flame-sealed in a 6 mm OD glass tube for further isotopic analysis.

The purified CO₂ was graphitized, and ¹⁴C was measured by accelerator mass spectrometry (AMS) at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility at Woods Hole Oceanographic Institution (WHOI). The Δ^{14} C results were reported as fraction modern (McNichol et al., 1994), and the conventional radiocarbon ages (years before present) were calculated using the Libby half-life as described by Stuiver and Polach (1977). The errors in Δ^{14} C measurements based on the duplicate sample analyses were \pm 3–8% for POC, \pm 6–18‰ for DOC and \pm 3–4‰ for DIC. A small split of CO $_2$ from DOC and POC oxidation was analyzed for δ^{13} C using a Thermo 253-Plus dual-inlet isotope ratio mass spectrometer (IRMS) in the Qingdao National Laboratory for Marine Science and Technology (QNLM) at the Center for Isotope Geochemistry and Geochronology (CIGG) in Qingdao, China. DIC δ^{13} C was analyzed using a Thermo Delta V IRMS coupled with a Thermo Gasbench II system at CIGG. The δ^{13} C values are reported in % relative to the VPDB standard for DIC and relative to IAEA standards (IAEA-CH3 Cellulose and IAEA-600 Caffeine) for DOC, and the δ^{13} C analytic precision was ± 0.2 %.

3. Results

The concentrations and carbon isotope values ($\delta^{13}C$ and $\Delta^{14}C$) of DIC and DOC are summarized in Table 1, and the results for the total fluxes of sinking particles and POC are summarized in Table 2. The hydrodynamic properties of the water column (temperature and salinity) of the sampling site in the Mariana Trench can be referred in Tian et al. (2018).

3.1. DIC and DOC concentrations

The DIC concentrations were low in the upper 50 m of the water column (1785–1880 $\mu mol/kg$), increased rapidly to 1000 m and then remained relatively constant (2100–2200 $\mu mol/kg$) from 1000 m to 8730 m (Fig. 2a). The depth profiles of DOC showed a trend opposite to that of DIC, i.e., higher at the surface (58–87 μM), decreasing rapidly to \sim 40 μM at 1000 m and then remaining relatively constant in the deep depth (Fig. 2b). DOC concentrations in the surface water (2 m) were significantly higher (87 μM) in February 2017 than in December 2015 (58 μM). The average DOC concentration (39 \pm 4 μM) in the deep (>2000 m) Mariana Trench water column was comparable, within statistical uncertainty, to the values recently reported for the deep NP Ocean (35–42 μM , Carlson, 2018; Druffel et al., 2019).

3.2. DIC and DOC $\delta^{13}C$ and $\Delta^{14}C$ values

The δ^{13} C values of DIC ranged from -0.05% to 0.62%, with an average of -0.29% (Table 1). The depth profiles of DIC δ^{13} C showed a relatively constant distribution in the water column (Fig. 2c). In contrast, the depth profiles of DIC Δ^{14} C showed high value (38‰) at the surface and decreased very rapidly down to -236% at ~ 1500 m depth and remained relatively constant below to 8730 m (Fig. 2d). The corresponding 14 C ages of DIC was modern in the surface and 2100 years BP (before present) in the deep waters (>1500 m) of the Mariana Trench.

The δ^{13} C values of DOC ranged from -21.2% to -22.9%, with an average value of -22.0% (Table 1). The depth profiles of DOC δ^{13} C were also relatively constant through the water column (Fig. 2c). The DOC Δ^{14} C values ranged from -189% at the surface to -538% in the deep layer, and its depth profiles, similar to those of DIC, decreased rapidly from the surface down to 1500 m and then remained relatively constant from 1500–10,000 m (Fig. 2d). The 14 C ages of DOC, however, were much older than those of DIC. The 14 C ages of DOC were \sim 1700 years at the surface and \sim 6100 years at depths of 8000–10,000 m, approximately 1700 years older at the surface and 4400 years older at great depth than the 14 C ages of DIC in the Mariana Trench site.

Table 1 Concentration and δ^{13} C and Δ^{14} C values measured in DIC and DOC collected in the Mariana Trench during 2015 and 2016.

Depth (m)	DIC				DOC					
	μmol/kg	δ ¹³ C (%)	Δ ¹⁴ C (%)	Ag(yr BP)e	μΜ	δ ¹³ C (%)	Δ ¹⁴ C (%)	Ag(yr BP)e		
MT-1 (11.3641°N, 142.9066°E)										
2015.12										
2	1785	0.37	38	Modern	58	-21.7	-195 ± 6	1800		
1000	2236	-0.48	-186	1650	41	-21.2	-487	5362		
1760	2181	-0.70	-231	2060	43	-22.1	-501 ± 10	5584		
3700	2141	-0.79	-220	1945	41	-21.5	-511	5747		
5370	2169	-0.32	-203	1745	42	-21.7	-517 ± 18	5846		
8730	2178	-0.42	-201	1734	38	-22.2	-527	6014		
2017.2.										
2	1880	0.62	36	Modern	87	-21.5	-189 ± 12	1683		
53	1981	0.33	23.	Modern	82	-22.4	-282	2660		
100	2074	-0.43	17	Modern	79	-22.1	-276 ± 11	2595		
150	2121	0.16	18	Modern	58	-21.7	-368	3687		
200	2170	-0.53	1.1	Modern	57	-22.2	-364	3687		
500	2185	-0.44	-101	798	48	-21.6	-390 ± 9	3976		
1000	2210	-0.44	-179	1514	40	-22.1	-469	5080		
1500	2180	-0.54	-222	1950	42	-22.6	-512	5758		
2000	2182	-0.92	-236	2100	40	-22.4	-517 ± 16	5844		
3000	2165	-0.55	-234	2079	39	-22.1	-532	6100		
4000	2170	-0.05	-218	1910	43	-21.7	-524	5963		
6050	2146	-0.30	-202	1740	41	-22.4	-529 ± 7	6048		
8000	2138	-0.11	-201	1734	38	-22.6	-538	6180		
10000	n.d.				40	-22.9	-535 ± 14	6150		

Note: The DOC Δ^{14} C values with errors represent duplicate measurements.

Table 2 Total mass and POC fluxes, and δ^{13} C and Δ^{14} C values of POC measured for sediment trap samples collected in the Marian Trench during 2016.

Month	Mass Flux (mg/m²/d)			POC Flux (mgC/m ² /d)		POC-8 ¹³ C (%)			<u>POC-Δ</u> ¹⁴ <u>C (%)</u>			
	2000m	4000m	6000m	2000m	4000m	6000m	2000m	4000m	6000m	2000m	4000m	6000m
MT-1 (11.36	641°N, 142.9	066°E)										
January	7.9	10.1	83.5	0.13	0.02	1.77	-22.0	-17.5	-22.3	35 ± 3	28 ± 4	26 ± 2
February	32.0	25.7	22.9	0.91	0.36	0.82	-22.0	-20.4	-22.2	29	28	22
March	47.3	27.3	21.3	1.83	0.42	0.49	-21.4	-18.4	-22.0	32 ± 5	35 ± 3	22 ± 6
April	18.6	9.1	26.9	0.66	0.05	0.66	-21.4	-18.0	-22.3	36	23	26
May	24.3	11.5	28.3	1.04	0.20	0.95	-21.3	-20.0	-21.8	33 ± 6	38 ± 4	23 ± 7
June	32.0	8.7	24.0	1.20	0.10	0.79	-20.7	-18.0	-22.1	34	29	21
July	16.9	2.3	24.7	0.41	0.02	0.57	-22.0	-20.2	-22.0	37	11	24
August	19.9	24.0	29.5	0.63	0.19	0.56	-21.1	-18.7	-22.3	38 ± 5	26 ± 8	20 ± 6
September	12.0	6.1	21.1	0.43	0.03	0.44	-21.4	-20.0	-22.0	13	25	21
Mean	23.4 \pm	13.9 \pm	31.4 \pm	$\textbf{0.80}~\pm$	$\textbf{0.15}\ \pm$	$\textbf{0.78}\ \pm$	$\mathbf{-21.5} \pm$	$-19.0\ \pm$	$\mathbf{-22.1}\ \pm$	$\textbf{32}\pm \textbf{7}$	$\textbf{27}\pm \textbf{7}$	$\textbf{23}\pm 2$
	11.3	8.7	18.6	0.48	0.14	0.38	0.4	1.1	0.2			

Note: The DOC $\Delta^{14}C$ values with errors represent duplicate measurements.

3.3. POC flux and $\delta^{13}C$ and $\Delta^{14}C$ values

The total mass fluxes of sinking materials collected at 2000 m, 4000 m and 6000 m in the Mariana Trench for nine continuous months in 2016 varied widely, ranging from 2.3 to 83.5 mg m $^{-2}$ d $^{-1}$, with average values of 23.4 \pm 11.3 mg m $^{-2}$ d $^{-1}$ at 2000 m, 13.9 \pm 8.7 mg m $^{-2}$ d $^{-1}$ at 4000 m and 31.4 \pm 18.6 mg m $^{-2}$ d $^{-1}$ at 6000 m (Table 2). Interestingly, the total mass flux at 6000 m was actually higher than those at 2000 m and 4000 m except in February and March (Fig. 3a). The flux of POC was much lower than the total mass flux, ranging from 0.02 to 1.83 mg C m $^{-2}$ d $^{-1}$ with averages of 0.80 \pm 0.48, 0.15 \pm 0.14 and 0.78 \pm 0.38 mg C m $^{-2}$ d $^{-1}$ at 2,000, 4000 and 6000 m, respectively (Table 2 and Fig. 3b). The flux of POC at 2000 m was higher in March and June and lower in January (Fig. 3b). In summary, the fluxes of POC at the 6000 m depth were higher in all nine months than those at 4000 m and even higher than those at 2000 m in January and July.

The δ^{13} C values of POC ranged from -17.5% to -22.3% (Table 2). The values showed some variations across the months at 4000 m depth but were quite constant at 2000 m and 6000 m during the nine-month period (Fig. 3c). The POC flux was lower in all months at 4000 m, but the POC δ^{13} C values measured at 4000 m were all higher than those at

2000 m and 6000 m. The average POC δ^{13} C value was $-21.5\pm0.4\%$ at 2000 m, $-19.0\pm1.1\%$ at 4000 m and $-22.1\pm0.2\%$ at 6000 m, respectively. The Δ^{14} C values of POC ranged from 11% to 38%, all with modern 14 C ages collected at the three depths of the Mariana Trench throughout the nine months (Fig. 3d). The average POC Δ^{14} C value was $32\pm7\%$ at 2000 m, $27\pm7\%$ at 4000 m and $23\pm2\%$ at 6000 m, being slightly higher at 2000 m than at the other two deep depths.

4. Discussion

4.1. Comparison of carbon isotopes between DIC and DOC

The Δ^{14} C values measured in seawater DIC provide useful information on water mass movement and penetration of anthropogenic CO₂ in the ocean (McNichol et al., 2000; Key et al., 2002; Druffel et al., 2008). A comparison of our DIC Δ^{14} C profiles at the Mariana Trench site with previously measured DIC Δ^{14} C profiles in the NP (WOCE P10 line, Sta. 47, 11.158°N, 149.331°E, 1993, Key et al., 2002) shows high similarity down to a depth of 6000 m (Fig. 2d) except in the upper 200 m layer. The lower DIC Δ^{14} C values in the surface water reflect the decrease in the dissolved atmospheric bomb 14 CO₂ signal from approximately 100‰

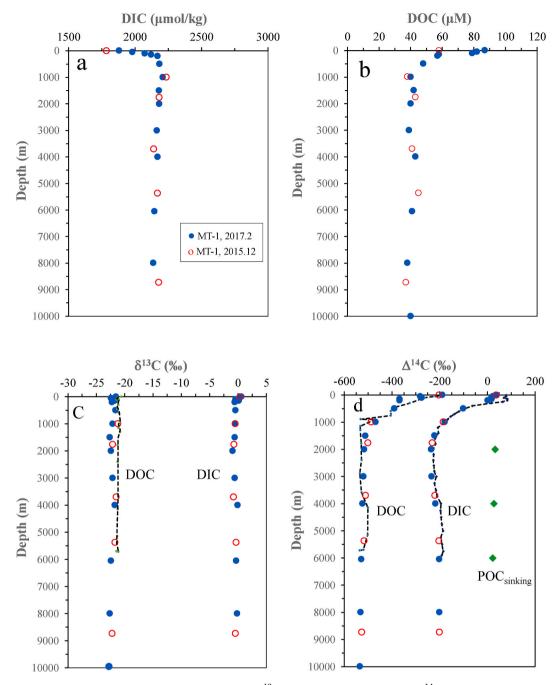


Fig. 2. Depth profiles of (a) DIC concentration, (b) DOC concentration, (c) $\delta^{13}C$ values of DIC and DOC and (d) $\Delta^{14}C$ values of DIC, DOC and sinking POC in the Mariana Trench. The dashed lines for DOC are plotted based on the isotope data reported by Druffel et al. (1992) for DOC in the central North Pacific Ocean (NP, $31^{\circ}00'N$, $159^{\circ}00'W$) for comparison. The dashed line for DIC is based on the $\Delta^{14}C$ values of DIC collected at nearby station 47 of line P10 (11.158°N, 149.331°E) during the World Ocean Circulation Experiment (WOCE) (1993/10/23) (Key et al., 2002) for comparison.

during the WOCE cruise in 1993 (Key et al., 2002) to ~50‰ in 2010 globally (Hua et al., 2013) and ~30‰ in the Northern Hemisphere in 2018 (Zigah et al., 2017; Gao et al., 2019), consistent with the $\Delta^{14}C$ values (~36‰) we measured for DIC. In the Mariana Trench, the bomb ^{14}C signal has been well mixed in the upper 200 m of water (all modern) and carried down to a depth of at least 1500 m by water mixing (Key et al., 2002; Kumamoto et al., 2013; Kaizer et al., 2020). Based on the DIC $\Delta^{14}C$ profile in the Mariana Trench, we observed a $\Delta^{14}C$ minimum (–235‰) depth at 2000–3000 m (Table 1, Fig. 2d). The average DIC $\Delta^{14}C$ value at 4000–8000 m was –207‰. This 28‰ difference is not due to our ^{14}C analytical uncertainty (1 $\sigma=\pm3.0$ ‰). Druffel et al. (2008) reported the same DIC $\Delta^{14}C$ profile trend in a North Central Pacific site (NCP, 31°N, 159°W), showing that the average DIC $\Delta^{14}C$ value

(–239‰) from depths of 1800–3600 m was significantly lower than the average value (–216‰) from depths of 4200–5770 m, a 23‰ difference. In their study, Masiello et al. (1998) reported a similar pattern in the NP and explained that the older DIC at the 2500 m depth than in the deepest water is due to the southward return flow of Pacific deep water, which has been isolated from the atmosphere longer than Pacific bottom water and thus has lower DIC $\Delta 14C$ values. Roussenov et al. (2004) performed an in-depth examination of the DIC $\Delta^{14}C$ distribution in the deep Pacific using two different models: a one-and-a-half-layer model for the bottom water and an isopycnal circulation model. They explained that the distribution of $\Delta^{14}C$ in the deep water of the Pacific is controlled by two regimes: a relatively fast horizontal transport of young $\Delta^{14}C$ in the bottom water and a much slower vertical balance between advection

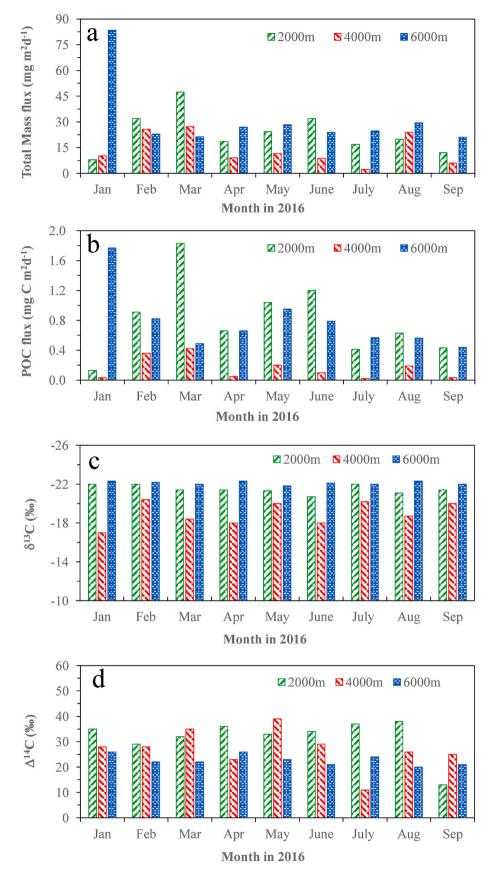


Fig. 3. Plots of (a) total mass flux, (b) POC flux, (c) POC δ^{13} C values and (d) POC Δ^{14} C values at the three depths (2000 m, 4000 m and 6000 m) collected during nine months in 2016 in the Mariana Trench.

diffusion and ^{14}C decay. Decreasing diapycnal mixing could lead to a greater northward penetration of bottom water with a relatively younger $\Delta^{14}\text{C}$ age into the NP (Roussenov et al., 2004). It is also possible that some of the bomb ^{14}C has penetrated the bottom waters of the Pacific during ocean circulation (Druffel et al., 2008). Down to hadal depths (>6000 m) in the Mariana Trench, the DIC $\Delta^{14}\text{C}$ value remained constant with that at 6000 m, suggesting that water mass circulation in the deep Mariana Trench is active and that the residence time of the water mass follows a pattern and time scale similar to those of the deep NP water circulation. No excessively old water mass remains at hadal depths in the Mariana Trench.

Unlike the DIC Δ^{14} C values, those of DOC in the ocean are controlled not only by water mass movement but also by biological processes to a large extent (Ogawa et al., 2001; Jiao et al., 2010; Carlson and Hansell, 2015; Zigah et al., 2017; Shen and Benner, 2019). A comparison of our DOC Δ^{14} C profiles at the Mariana Trench site with DOC Δ^{14} C profiles in the North Central Pacific (NCP, 31°00'N, 159°00'W) measured by Druffel et al. (1992) shows that they agree well down to 5700 m in the water column (Fig. 2d), and our values also agree with the recent DOC Δ^{14} C results reported for the NP (Zigah et al., 2017; Druffel et al., 2019). Down to hadal depths (>6000 m) in the Mariana Trench, the DOC concentration (40 μ M), DOC Δ^{14} C value (-535%) and DOC 14 C age (6150 years BP) together suggest that no excessive old high concentrations of DOC have accumulated in the deep trench. This is consistent with the conclusion derived from the DIC Δ^{14} C profiles indicating that water movement is active in the deep Mariana Trench and the water mass has similar concentrations and isotopic signatures of DOC as water in the deep NP at depths of 5000–6000 m, as reported in previous studies (Druffel et al., 1992, 2019; Zigah et al., 2017).

Bomb 14 C is also present in the DOC pool and penetrates down to approximately 1000 m depth in the Mariana Trench (Fig. 2d). A comparison of the DOC and DIC Δ^{14} C profiles indicates that the 14 C ages of DOC are \sim 1800 years older in the surface water and 4000 years older in the deep water than the 14 C ages of DIC in the Mariana Trench. These 14 C age differences between DOC and DIC are consistent with the results of other sites in the deep NP (Druffel et al., 1992, 2008; Key et al., 2002; Zigah et al., 2017, 2019; Ding et al., 2018), indicating that DOC below 1000 m in the water column is refractory in nature and cycled on much longer time scales than DIC in the ocean (Hansell et al., 2009; Jiao et al., 2010; Zigah et al., 2017; Druffel et al., 2019). A good linear correlation ($r^2=0.89$) exists between Δ^{14} C values of DOC and DIC in the Mariana Trench, as shown in Fig. 4, suggesting that both the distribution and cycling of DOC and DIC in the deep water of the trench are quite homogenous and are largely controlled by water movement and

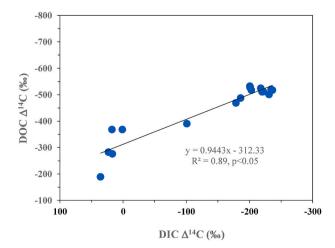


Fig. 4. Plot of DOC $\Delta^{14}C$ values vs. DIC $\Delta^{14}C$ values for water samples collected in the Mariana Trench during the 2015 and 2016 cruises. The line is a linear regression fit to the data.

hydrodynamic mixing, as found in other oceans (Beaupre and Aluwihare, 2010; Beaupre et al., 2020; Ding et al., 2020).

4.2. Rapid and temporal variability of POC flux

The fluxes of total sinking particles collected for the nine months in 2016 in the Mariana Trench were generally low. Even at 2000 m depth, the average flux of particles was 23.4 \pm 11.3 mg m⁻² d⁻¹, which is significantly lower than the global average biogenic particle flux of 68 mg m⁻² d⁻¹ at the mesopelagic/bathypelagic boundary of 2000 m (Honjo et al., 2008; Hwang et al., 2015). The POC fluxes were even lower, averaging 0.80 mg C m $^{-2}$ d $^{-1}$, 0.15 mg C m $^{-2}$ d $^{-1}$ and 0.78 mg C m⁻² d⁻¹ at depths of 2000 m, 4000 m and 6000 m, respectively. These POC values accounted for only a very small fraction (3.4%, 1.1% and 2.5%) of the total particle fluxes at each depth. In our study, we determined that lithogenic material, opal and particulate inorganic carbon (PIC) accounted for most of the total particle fluxes of our samples in the Mariana Trench (Xiao et al., 2020). A total of 24 different minerals, including silicates, carbonates, sulfates and phosphate, were identified, and the mineral abundances in the sinking particles were consistent with the total mass fluxes at each depth. The volcanic materials likely eroded from surrounding ridges, and oceanic basins contributed most of the minerals (Xiao et al., 2020). It seems that the dominant minerals apparently lowered the POC contents in the sinking particles we

The POC fluxes we measured in the Mariana Trench deep water were significantly lower than those reported in the ocean. This could also be largely due to the low primary production (PP) of the site. PP was calculated based on satellite chlorophyll data (http://sites.science.orego nstate.edu/ocean.productivity/index.php) using a vertically generalized production model (VGPM) (Behrenfeld and Falkowski, 1997), and the average PP was approximately 160 ± 14 mg C m⁻² d⁻¹ in spring in the Mariana Trench in 2016. As evidence, the total nitrogen and phosphate concentrations we measured in the surface water (100 m) during the two cruises were also quite low (<5 µM), which limited PP at the Mariana Trench site (Tian et al., 2018). The PP of the Mariana Trench site is significantly lower than the annual mean PP of 303 mg C m⁻² d⁻¹ at a western NP station (S1, 30⁰ N, 145⁰ E) reported by Matsumoto et al. (2016). In their recent study, Sukigara et al. (2019) reported that the POC fluxes at 100 m and 200 m at station S1 (30°N, 145°E) ranged from 13.8 to 59.2 and from 16.1 to 36.9 mg C m^{-2} d^{-1} , respectively, accounting for 4.6-19.4% of the PP. In contrast, the mean POC fluxes we measured at 2000 m, 4000 m and 6000 m in the Mariana Trench only accounted for 0.5%, 0.1% and 0.5%, respectively, of the surface PP, much lower than the POC fluxes at 100 m and 200 m measured at station S1 (Sukigara et al., 2019). Armstrong et al. (2002) reported that POC fluxes in the deep ocean could vary significantly. Along a transect at 140°W in the NP, POC fluxes of \sim 5.0 mg C m $^{-2}$ d $^{-1}$ were measured at 5°N from 2000-4000 m, while POC fluxes decreased to 0.5-1.0 mg C m⁻² d⁻¹ at 9°N in the same depth range. In general, studies have demonstrated that the depth profiles of POC fluxes in the ocean typically show an exponential decrease from the surface to the deep-water column (Lee et al., 1998, 2009; Honjo et al., 1999; Wakeham et al., 2000; Hwang et al., 2015), implying that most remineralization of POC occurs in the upper 1000 m water depth and that only a very small fraction of surface POC can pass 1000 m and reach the ocean floor (Martin et al., 1987; Armstrong et al., 2002). In the Mariana Trench, however, we found that the overall POC fluxes at the three deep depths were well correlated with the monthly total mass fluxes, as shown in Fig. 5, suggesting that the total mass flux, such as that of clay minerals, is an important factor that causes POC to sink to deep depths and perhaps even hadal depths (Armstrong et al., 2002), as suggested in some recent studies (Glud et al., 2013; Luo et al., 2018; Tian et al., 2018; Wang et al., 2019). However, since we deployed our upper trap at 2000 m depth far below the euphotic zone, we were not able to evaluate the sink dynamics of POC in the upper water layer of the Mariana Trench. These

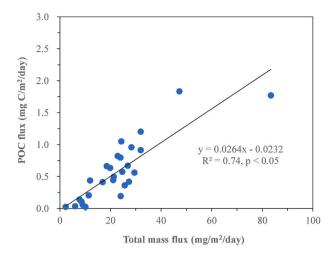


Fig. 5. Plot of the monthly POC flux vs. monthly total mass flux at the three depths in the Mariana Trench in 2016. The solid line is a linear regression fit to the data.

speculations therefore need to be further studied in the future.

Despite the low POC fluxes measured at great depth in the Mariana Trench, the high POC Δ^{14} C values and their modern 14 C ages indicate that the surface marine-derived recently fixed POC sank rapidly to hadal depths, perhaps in days or weeks. This is an important finding for active carbon cycling in the deep Mariana Trench. Modern Δ^{14} C values (118‰, 66‰) have also been reported for sinking POC collected at depths of 4200-5100 m in the NCP (31°00'N, 159°00'W) and the Sargasso Sea (31°50′N, 63°30′W) (Druffel et al., 1992). As discussed above, the rapid sinking of fresh surface POC to deep depths in the trench, even in small amounts, is an important process providing essential carbon flow for microbial and biological communities adapted to the extreme high-pressure environment (Ohara et al., 2012; Glud et al., 2013; Nunoura et al., 2015; Tarn et al., 2016; Okumura et al., 2016; Tian et al., 2018). A recent study by Wang et al. (2019) reported that modern bomb $^{14}\mathrm{C}$ (10–65‰) was detected in amphipod muscle tissues collected in three Pacific Ocean trenches, including the Mariana Trench, indicating the dietary preference of amphipods for fresh and labile POC that sinks from the surface, consistent with our POC Δ^{14} C results measured in the deep Mariana Trench.

4.3. Reasons for the unexpected vertical mass and POC fluxes

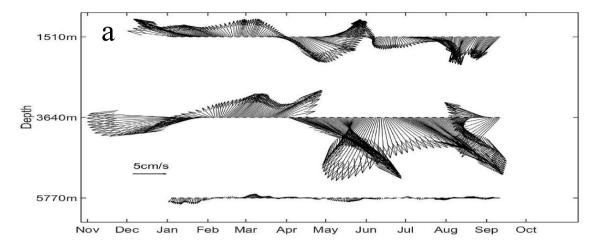
As shown in Fig. 3a and b, we observed large spatial and temporal variations in total particle and POC fluxes in the deep water of the Mariana Trench and especially higher fluxes at 6000 m depth. These flux variations, however, cannot be simply explained by seasonal variations in PP. In early spring (February and March) and early summer (May and June), the relatively high POC fluxes measured at the 2000 m depth could be related to the high PP seasons in the NP (Fujiki et al., 2014; Matsumoto et al., 2016). At the 6000 m depth, however, the POC fluxes were higher than those at the 4000 m depth and even higher than those at the 2000 m depth in January, July and September. These exceptionally higher fluxes of POC at 6000 m thus did not follow the typical profiles of an exponential decrease in POC flux in the water column of the ocean (Martin et al., 1987; Armstrong et al., 2002; Lee et al., 2009). We expect that lateral transport could play an important role in affecting the observed flux variation at different depths. Providing strong evidence for such lateral transport, the water movements measured at the deep-water depths during trap deployment in the Mariana Trench clearly indicate that the horizontal current at 3640 m depth was notably more energetic than those at depths of 1510 m and 5770 m (Fig. 6a). We calculated the monthly average flow rates and flow direction for the three depths and plotted in Fig. 6b, and plotted the monthly total mass

flux vs. the average flow rate (Fig. 6c) and POC flux vs. the average flow rate (Fig. 6d). A general negative linear correlation was observed for both total mass flux ($R^2 = 0.34$, p < 0.01) and POC flux ($R^2 = 0.37$, p < 0.01) vs. the flow rates. The total mass fluxes and POC fluxes at the 6000 m were clearly correlated to the very lower flow rates compared with those at the 1510 m and 5770 m in the Mariana Trench. We believe that the horizontal current measured at 3640 m could extend to 4000 m and had a significant influence on sinking particles. The force generated by horizontal flow could remove some particles, thus reducing the vertical total mass and POC fluxes at this depth. Down at a depth of 6000 m with relatively calm water movement, the V-shaped terrain structure of the trench could play an important role acting as the top open end of a funnel trap, resulting in accumulated POC fluxes in the deep trench. As a similar case, Shin et al. (2002) reported that the total mass flux measured at 4200 m (677 mg m $^{-2}$ d $^{-1}$) was 8 times higher than that measured at 1000 m (82 mg m $^{-2}$ d $^{-1}$) in the Japan Trench in the western NP, affected largely by lateral transport. We propose deploying traps at >6000 m depth in the Mariana Trench to further verify this hypothesis.

The modern ¹⁴C ages of POC collected in the deep-water depths of the Mariana Trench are somewhat surprising if horizontal water movement played an important role in the observed POC fluxes. The horizontal current can carry resuspended sediment from the surrounding area and contribute to aged POC (Nakatsuka et al., 1997; Honda et al., 2000; Hwang et al., 2008). Hwang et al. (2010) examined the correlations between the Δ^{14} C values of POC and the Al contents of sinking particles collected by sediment traps in different oceans, including the northeastern Pacific, Okinawa Trough, and Canada deep basin. They found a strong negative relationship with high Al contents and more depleted Δ^{14} C values of POC. In addition, they calculated that overall, approximately 35% of the sinking POC in the deep water (>1000 m) of the oceans was derived from resuspended sediment that contributed aged POC, and the average Δ^{14} C value of POC was $-87~\pm$ 63‰. Nakatsuka et al. (1997) also reported more depleted Δ^{14} C values (-147 \pm 60% and -281 \pm 82%) for sinking POC collected at 4789 m and 8789 m, respectively, in the Japan Trench, indicating a significant influence of resuspended sediment carried by water currents. Our results clearly showed differences for sinking POC, indicating that all modern ¹⁴C ages of POC sinking to the deep Mariana Trench had a very limited influence compared with aged POC of resuspended sediment. The modern bomb ¹⁴C value (10–65‰) detected in amphipod muscle tissues collected at the bottom of the Mariana Trench also provides direct evidence supporting our POC results. This exceptional phenomenon could suggest that in the deep Mariana Trench, even though the horizontal water currents could have a significant influence on the POC fluxes, the lithologic minerals transported by the currents are mainly from volcanic sources (Xiao et al., 2020), which could be free in POC.

4.4. Implication for carbon cycling in the Mariana Trench

The carbon isotope results we obtained for DIC, DOC and POC in the Mariana Trench provide the first data set to evaluate the carbon cycling in this deepest trench in the ocean. Our data suggest that the carbon cycle at the deep and hadal depths in this trench is active and dynamic due to the well-mixed water masses in the trench. The water exchange time at hadal depths in the Mariana Trench is likely on the same order of magnitude as that in the deep NP water mass. No significant accumulation or extra aging of DIC and DOC at hadal depths was observed in the trench. The biological pump seems efficient and drives surface fresh POC to sink rapidly to the deep trench, and lateral movement of water currents plays important roles affecting the vertical fluxes of both total mass and POC. We propose that the rapid sinking of POC could reach hadal depths through the V-shaped trench, especially when the north-flowing current reaches the northern slope of the trench, which could result in more rapid sinking of particles. We believe that this rapid sinking of very labile POC to the deep trench could be an important process of carbon cycling that could provide essential food supplies to support organisms



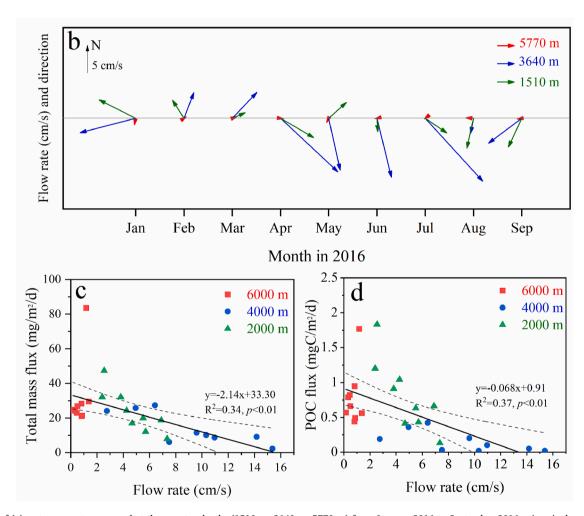


Fig. 6. Plots of (a) water currents measured at three water depths (1510 m, 3640 m, 5770 m) from January 2016 to September 2016 using single-point acoustic current meters; (b) calculated monthly average flow rates and flow directions in 2016 at 1510 m, 3640 m and 5770 m; (c) correlation between the monthly total mass fluxes and the average flow rates; and (d) correlation between the monthly POC fluxes and average flow rates in 2016 at 2000 m, 4000 m and 6000 m in the Mariana Trench. The solid lines (Fig. c,d) are linear regression fit to the data, and the dashed lines mark the 95% confidence interval.

living in the extreme hadal environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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