

Particulate organic carbon export fluxes on Chukchi Shelf, western Arctic Ocean, derived from $^{210}\text{Po}/^{210}\text{Pb}$ disequilibrium*

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Abstract Fluxes of particulate organic carbon (POC) were derived from $^{210}\text{Po}/^{210}\text{Pb}$ disequilibrium during the 4th Chinese National Arctic Research Expedition (CHINARE-4) from July 1 to September 28, 2010. Average residence times of particulate ^{210}Po in the euphotic zone were -16.00 a to 1.54 a, which are higher than those of dissolved ^{210}Po (-6.89 a to -0.70 a). Great excesses of dissolved ^{210}Po were observed at all stations, with an average $^{210}\text{Po}/^{210}\text{Pb}$ ratio of 1.91 ± 0.20 , resulting from ^{210}Pb atmospheric deposition after sea ice melt. POC fluxes from the euphotic zone were estimated by two methods (E and B) in the irreversible scavenging model. Estimated POC fluxes were 945–126 mmol C/(m²·a) and 1 848–109 mmol C/(m²·a) by methods E and B, respectively, both decreasing from low to high latitude. The results are comparable to previous works for the same region, indicating efficient biological pumping in the Chukchi Sea. The results can improve understanding of the carbon cycle in the western Arctic Ocean.

Keyword: particulate organic carbon (POC) flux; $^{210}\text{Po}/^{210}\text{Pb}$ disequilibrium; Chukchi Shelf

1 INTRODUCTION

About 30% of anthropogenic CO₂ is taken up by the ocean, especially the Arctic and Southern oceans, owing to high biological production in the large ocean margin areas and low temperatures (Cai et al., 2010). The Chukchi Shelf is one of the most productive continental shelves in the Arctic Ocean and is important in carbon sequestration (Woodgate et al., 2005; Yu, 2010). The magnitude of atmospheric CO₂ absorbed in these areas has been of concern to oceanographers worldwide (Baskaran et al., 1995; Anderson et al., 1998; Pipco et al., 2002; Bates et al., 2006; Liu et al., 2007; Else et al., 2008).

As an interface for exchange of CO₂ between the surface ocean and interior ocean, the euphotic zone plays a key role in the growth, removal and cycling of biomass. Fluxes within the euphotic zone of carbon, nutrients and other associated elements involved in

biogeochemical cycles are very important in the study of global CO₂ (Yang et al., 2004; He et al., 2008). Radioactive isotopes, especially natural particle-reactive radionuclides (e.g., ^{234}Th and ^{210}Po) provide another possible means for quantifying export flux of particulate organic carbon (POC) from the surface ocean at various time scales, because of their specific half-lives (Verdeny et al., 2009; Stewart et al., 2011).

Both ^{210}Po and ^{210}Pb are progeny of the ^{238}U decay chain, and are naturally occurring particle-reactive radionuclides. ^{210}Pb ($t_{1/2}=22.3$ a) is produced in situ by decay of its longer-lived grandparent ^{226}Ra and from atmospheric deposition, whereas ^{210}Po is supplied

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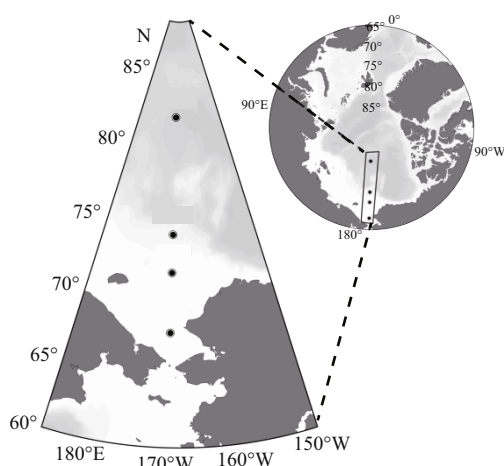


Fig.1 Study area and sampling stations

almost exclusively in situ by the decay of its grandparent ^{210}Pb , with a minor additional source from atmospheric deposition (Baskaran, 2011). Although ^{210}Pb and ^{210}Po are particle-reactive radionuclides, the preferential uptake of ^{210}Po by plankton and its transfer in the food chain can be used to quantify organic carbon fluxes (Carvalho, 2011; Kim et al., 2012). Furthermore, export on a timescale of several months can be derived by $^{210}\text{Po}/^{210}\text{Pb}$ disequilibrium, longer than the several weeks of $^{234}\text{Th}/^{238}\text{U}$ disequilibrium (Friedrich et al., 2002).

In this study, we estimated ^{210}Po -derived export fluxes of POC from the euphotic zone at different stations during the 4th Chinese National Arctic Research Expedition (CHINARE-4).

2 MATERIAL AND METHOD

2.1 Location and sample collection

During CHINARE-4 from July 1 to September 20, 2010, a conductivity, temperature and depth (CTD) rosette (SEABIRD 911/17) was used to collect 10-L seawater samples at various depths in the upper 0–100 m water column at four stations in the Arctic Ocean, for analysis of ^{210}Po and ^{210}Pb . At stations R03 and R09, sampling was done during July 21–24, whereas that at R12 and R20 was during August 24–29. A map of the study area and stations is provided in Fig.1.

One liter of each 10-L seawater sample was filtered through a pre-combusted (450°C, 2 h) 0.45- μm QMA membrane immediately after sampling, and then particles and the filter were sealed inside tin foil and stored frozen for subsequent POC analysis. The remaining nine liters of seawater from each sample

were filtered through a 0.45- μm membrane filter (120 mm diameter), to obtain dissolved samples and particulate samples. After filtration, dissolved samples were acidified to pH 1 with concentrated HCl. All samples were stored until laboratory analysis of POC, ^{210}Po and ^{210}Pb .

2.2 Radionuclide analysis

For dissolved and particulate ^{210}Po and ^{210}Pb , the analytical methods used followed the modifications of Nozaki (1986) and Masque et al. (2002). Briefly, filter samples were digested using sequential aqua regia, HNO_3 and HCl, with HClO_4 and HF, and the dissolved material was eventually taken up in 1.5 mol/L HCl. During digestion, samples were spiked with ^{209}Po as a yield tracer and stable lead as a recovery tracer. The dissolved ^{210}Po was co-precipitated with $\text{Fe}(\text{OH})_3$ and redissolved with 1 mol/L HCl. Po was plated onto nickel discs using stirring hot plates and counted on a low background alpha spectrometer (Canberra 7200-08; CT, USA). The plating solution was then replated to remove any residual ^{210}Po and ^{209}Po , then re-spiked with ^{209}Po and retained for at least 6 months for ingrowth of ^{210}Po to determine ^{210}Pb . Pb yields were determined through measurement of stable Pb by inductively coupled plasma mass spectrometry (ICP-MS), and recovery was generally between 80% and 90%. The ^{210}Po and ^{210}Pb activities were decay-corrected to the time of sampling according to Fleer and Bacon (1984).

3 RESULT AND DISCUSSION

3.1 Distribution of particulate and dissolved ^{210}Po and ^{210}Pb

The activity of ^{210}Po and ^{210}Pb in different phases and the activity ratio $^{210}\text{Po}/^{210}\text{Pb}_{\text{A.R}}$ are given in Table 1. Vertical profiles of $^{210}\text{Po}/^{210}\text{Pb}_{\text{A.R}}$ in different water columns are shown in Fig.2. The average dissolved ^{210}Pb of various stations shows an increasing trend from 1.12 to 1.78 Bq/m^3 northward from July to August, which may have resulted from atmospheric deposition of ^{210}Pb during ice melt. The average particulate ^{210}Pb (2.22 Bq/m^3) was higher than the dissolved ^{210}Pb (1.49 Bq/m^3), indicating intense particle scavenging of ^{210}Pb (Bacon et al., 1976; Shimmield et al., 1995).

The vertical profile of POC in different water columns and relationship between POC content and particulate Po are shown in Fig.3. The average content of POC decreased northward. However, the highest

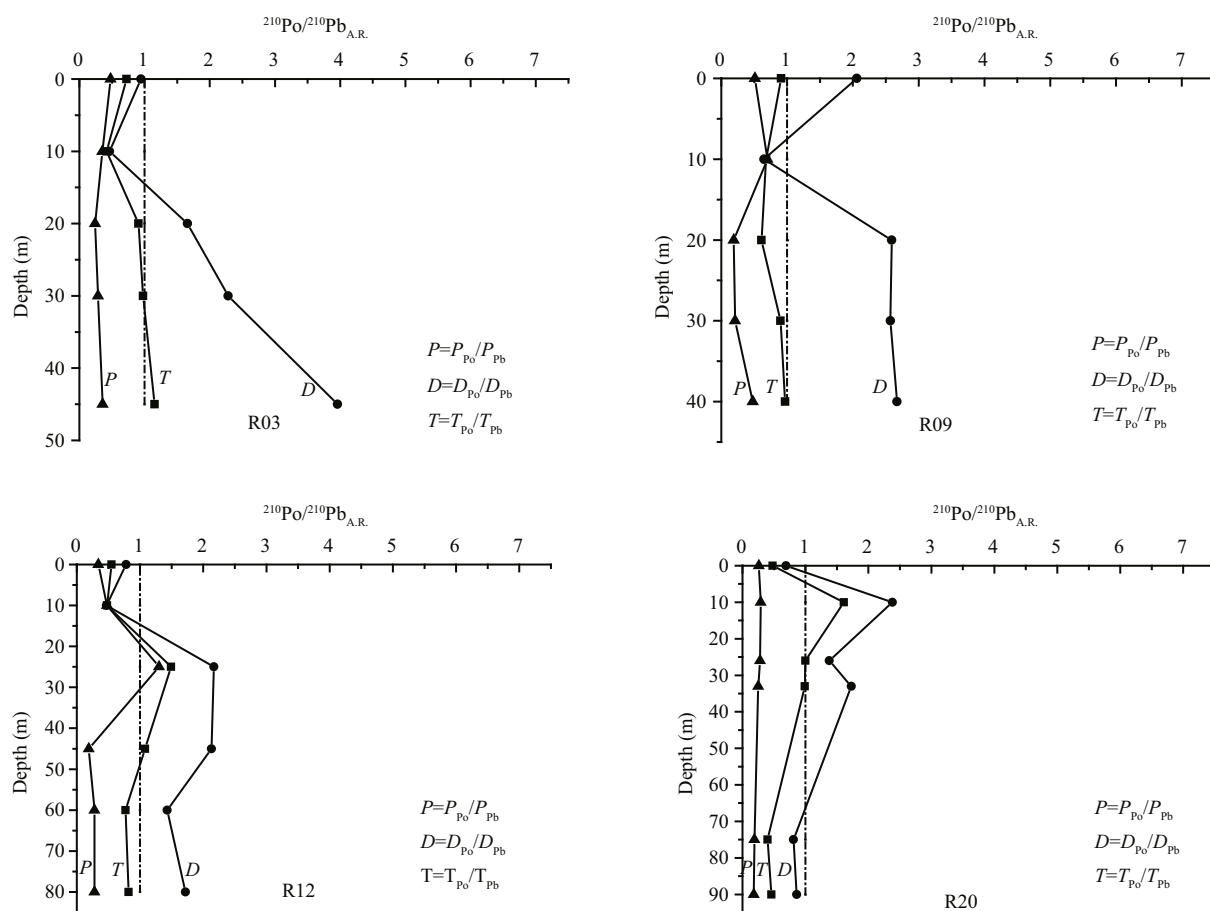


Fig.2 Vertical profiles of ^{210}Po , ^{210}Pb and POC in different water columns

P , D , and T represent particulate, dissolved, and total ^{210}Po or ^{210}Pb , respectively. $^{210}\text{Po}/^{210}\text{Pb}_{\text{A.R.}}$ is activity ratio of ^{210}Po to ^{210}Pb

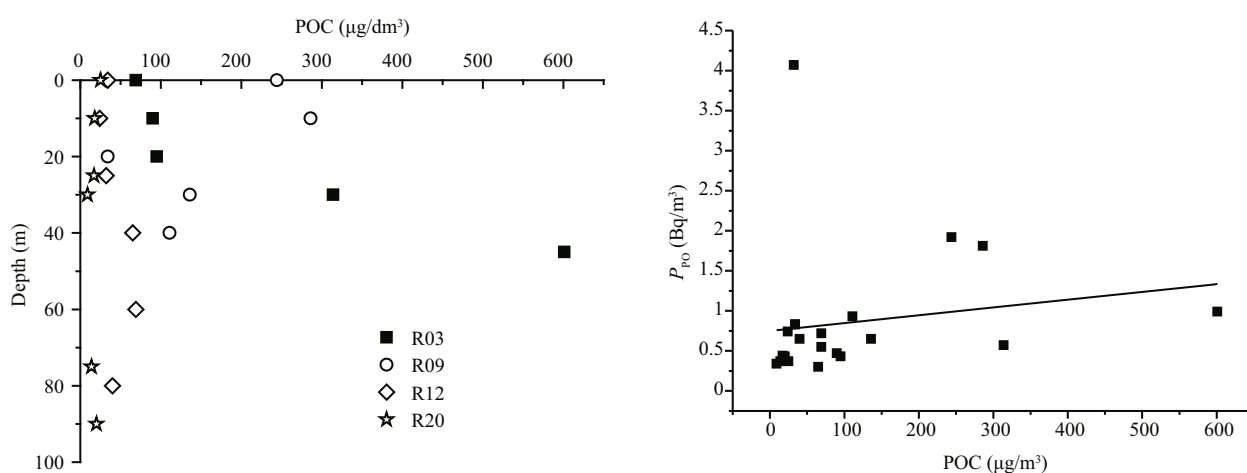


Fig.3 Vertical profile of POC in different water columns and relationship between POC content and particulate Po

value for surface water appeared at R09, perhaps caused by ice melt, which resulted in biomass bloom from the bottom ice (Yu et al., 2012). Positive correlation ($R^2=0.027$) between POC content and particulate Po confirms that Po may be used as a tracer of POC export.

3.2 Total Po and Pb, residence time of ^{210}Po , water-column disequilibrium and flux

The half-life of ^{210}Po constrains the seasonal time scale of disequilibrium of $^{210}\text{Po}/^{210}\text{Pb}$ for quantifying the export fluxes of POC. Boundary scavenging,

Table 1 POC content, ^{210}Po and ^{210}Pb activity, and Po/Pb ratio in water column samples

Station No.	Latitude	Longitude	Depth (m)	POC	D_{Po}	P_{Po}	T_{Po}	D_{Pb}	P_{Pb}	T_{Pb}	$^{210}\text{Po}/^{210}\text{Pb}_{\text{A.R.}}$		
				($\mu\text{g}/\text{dm}^3$)	(Bq/ m^3)						D	P	T
R03	68°N	169°W	0	69	1.19±0.20	0.55±0.05	1.73±0.14	1.26±0.13	1.14±0.16	2.40±0.23	0.94±0.10	0.48±0.05	0.72±0.07
			10	90	0.88±0.12	0.47±0.05	1.35±0.15	1.91±0.19	1.33±0.14	3.24±0.30	0.46±0.05	0.35±0.04	0.42±0.04
			20	95	2.64±0.30	0.43±0.05	3.08±0.35	1.59±0.17	1.80±0.16	3.40±0.33	1.66±0.12	0.24±0.02	0.91±0.10
			30	314	2.45±0.25	0.57±0.06	3.02±0.33	1.07±0.09	2.02±0.19	3.09±0.30	2.28±0.23	0.28±0.03	0.98±0.10
			45	601	3.13±0.35	0.99±0.10	4.12±0.40	0.79±0.80	2.78±0.26	3.57±0.37	3.96±0.40	0.36±0.03	1.15±0.12
R09	72°N	169°W	0	244	2.68±0.22	1.92±0.21	4.59±0.50	1.30±0.12	3.73±0.38	5.03±0.52	2.06±0.20	0.51±0.05	0.91±0.10
			10	286	0.97±0.79	1.81±0.17	2.78±0.31	1.49±0.16	2.57±0.25	4.06±0.40	0.65±0.06	0.70±0.07	0.68±0.07
			20	34	2.47±0.28	0.83±0.10	3.30±0.31	0.95±0.11	4.46±0.46	5.41±0.53	2.59±0.26	0.19±0.02	0.61±0.06
			30	136	3.36±0.28	0.65±0.10	4.00±0.38	1.31±0.11	3.13±0.31	4.44±0.46	2.57±0.26	0.21±0.02	0.90±0.09
			40	111	1.53±0.10	0.93±0.11	2.46±0.25	0.57±0.06	1.95±0.20	2.52±0.20	2.67±0.26	0.48±0.04	0.97±0.10
R12	74°N	169°W	0	34	1.72±0.16	0.83±0.08	2.55±0.26	2.19±0.25	2.44±0.26	4.63±0.43	0.78±0.08	0.34±0.03	0.55±0.05
			10	24	1.33±0.10	0.74±0.08	2.07±0.26	2.79±0.30	1.55±0.13	4.34±0.43	0.47±0.46	0.48±0.05	0.48±0.05
			25	32	1.94±0.26	4.07±0.45	6.01±0.68	0.89±0.12	3.14±0.29	4.03±0.41	2.17±0.22	1.30±0.13	1.49±0.15
			40	65	2.81±0.22	0.30±0.05	3.11±0.31	1.31±0.16	1.55±0.16	2.87±0.28	2.13±0.21	0.19±0.02	1.08±0.12
			60	69	2.72±0.30	0.72±0.08	3.44±0.38	1.90±0.18	2.56±0.30	4.46±0.44	1.43±0.14	0.28±0.03	0.77±0.09
R20	82°N	169°W	80	40	2.39±0.26	0.65±0.07	3.05±0.29	1.39±0.16	2.35±0.23	3.74±0.38	1.72±0.17	0.28±0.03	0.82±0.08
			0	25	1.06±0.10	0.37±0.04	1.43±0.12	1.54±0.15	1.43±0.16	2.97±0.30	0.69±0.07	0.26±0.03	0.48±0.05
			10	18	5.29±0.56	0.38±0.04	5.67±0.62	2.22±0.26	1.30±0.13	3.52±0.35	2.38±0.23	0.29±0.03	1.61±0.16
			25	17	4.05±0.38	0.44±0.04	4.49±0.45	2.93±0.30	1.55±0.16	4.48±0.45	1.38±0.16	0.28±0.02	1.00±0.11
			30	9	2.27±0.29	0.34±0.05	2.61±0.27	1.32±0.14	1.33±0.13	2.65±0.23	1.73±0.16	0.25±0.02	0.99±0.10
			75	14	0.85±0.11	0.37±0.04	1.22±0.12	1.06±0.10	2.00±0.20	3.06±0.29	0.81±0.10	0.19±0.02	0.40±0.04
			90	20	1.39±0.14	0.43±0.04	1.82±0.17	1.62±0.15	2.33±0.23	3.96±0.41	0.86±0.10	0.18±0.02	0.46±0.04

Notes: D_{Po} is the activity of dissolved ^{210}Po , P_{Po} the activity of particulate ^{210}Po , T_{Po} the activity of total ^{210}Po , D_{Pb} the activity of dissolved ^{210}Pb , P_{Pb} the activity of particulate of ^{210}Pb , and T_{Pb} the activity of total ^{210}Pb . $^{210}\text{Po}/^{210}\text{Pb}_{\text{A.R.}}$ is the activity ratio of ^{210}Po to ^{210}Pb .

Table 2 Inventory of particulate and dissolved ^{210}Po and ^{210}Pb in euphotic zone, and residence time and export fluxes of particulate and dissolved ^{210}Po

Station	Euphotic layer (m)	I_{DPo}	I_{PPo}	I_{DPb}	I_{PPb}	J_{Po}	P_{Po}	τ_{DPo}	τ_{PPo}
		(Bq/ m^2)				(Bq/($\text{m}^2\cdot\text{a}$))		(a)	
R03	30	53.70	15.15	43.73	47.18	-18.20	40.26	-2.95	0.38
R09	30	71.10	39.08	37.88	104.18	-60.63	58.18	-1.17	0.67
R12	40	78.00	59.40	71.80	86.80	-11.32	38.69	-6.89	1.54
R20	75	202.80	28.50	136.05	114.15	-121.82	34.49	-1.66	0.83

Notes: I_{DPo} , I_{PPo} , I_{DPb} and I_{PPb} are inventories of dissolved ^{210}Po , particulate ^{210}Po , dissolved ^{210}Pb and particulate ^{210}Pb above the euphotic zone, respectively; J_{Po} is the scavenging flux of dissolved ^{210}Po ; P_{Po} is export flux of particulate ^{210}Po ; τ_{DPo} and τ_{PPo} are residence times of particulate and dissolved ^{210}Po , respectively.

especially advective processes, is important for ^{210}Pb , owing to its half-life of 22.3 years (Friedrich et al., 2002; Kim et al., 2012). The spatially comparable activity of ^{210}Po and ^{210}Pb further constrains the fluxes because of advective processes, which are minor relative to vertical fluxes.

An irreversible, steady state model of ^{210}Po was built without advective and diffusive items and atmospheric deposition. To estimate the rates of ^{210}Po and ^{210}Pb scavenged by suspended particles and their removal via sinking particles, the following equations

were used for the disequilibrium between ^{210}Po and ^{210}Pb in seawater (Bacon et al., 1976):

$$J = \lambda_{\text{Po}} (I_{\text{DPb}} - I_{\text{DPo}}), \quad (1)$$

$$P = \lambda_{\text{Po}} (I_{\text{PPb}} - I_{\text{PPo}}) + J, \quad (2)$$

where J is the scavenging flux of dissolved ^{210}Po ; λ_{Po} is the decay constant (0.005/d) of ^{210}Po ; I_{DPo} and I_{DPb} are the inventory of dissolved ^{210}Po and ^{210}Pb , respectively; P is the export flux of particulate ^{210}Po ; I_{PPo} and I_{PPb} are the inventory of particulate ^{210}Po and ^{210}Pb . The residence times of particulate and dissolved ^{210}Po are given by

$$\tau_{DPo} = I_{DPo} / J, \quad (3)$$

$$\tau_{PPo} = I_{PPo} / P. \quad (4)$$

Based on Eqs. 1–4, J , P , τ_{DPo} , and τ_{PPo} are presented in Table 2. This shows that mean residence times of particulate and dissolved ^{210}Po in the euphotic zone of different stations were almost all negative, which may be caused by regeneration of particles (He et al., 2008). However, the residence time of particulate ^{210}Po was higher than that of dissolved ^{210}Po , indicating rapid biogeochemical cycling in the study area (Rutgers van der Loeff et al., 1995).

3.3 Distribution of particulate organic carbon concentrations

Concentrations of POC in the water column are given in Table 1. POC concentration was 9–601 $\mu\text{g C/L}$ in the study area, with average 106 $\mu\text{g C/L}$. The highest concentration of 601 $\mu\text{g C/L}$ were found at R03 during the first sampling period, this decreased northwards and offshore during the second sampling period, consistent with the distribution of chlorophyll-*a* (Yu et al., 2012). POC concentrations in surface water were lower than in deeper water, owing to sediment resuspension (Yu et al., 2012).

3.4 POC export fluxes from euphotic zone

The distribution of POC export fluxes in the Arctic Ocean has significant temporal and regional variability (Yu et al., 2012). Export fluxes of particulate ^{210}Po and its residence time in the euphotic zone were calculated in the irreversible scavenging model (Yang, 2005). Furthermore, POC export fluxes were calculated with the ratio of POC content to activity of ^{210}Po , per the equation of Buesseler et al. (1992) and Yang (2005):

$$B_{\text{POC-flux}} = f_{\text{POC}} = f_{\text{Po}} \times r, \quad (5)$$

where f_{Po} is the export flux of particulate ^{210}Po and r is the ratio of content of POC to activity of ^{210}Po at the bottom of the euphotic zone. With this equation, we hypothesized that the carriers of ^{210}Po and POC were the same particle but not that they have similar geochemical behaviors. Apparently, the former hypothesis was easily satisfied in the study area. The export fluxes and the residence times of ^{210}Po are listed in Table 2, and the POC fluxes (Method B, marked with $B_{\text{POC-flux}}$) are given in Table 3. At station R03, the residence time of ^{210}Po was the shortest, only 0.38 a, and the POC export flux was the highest, at 1 848 $\text{mmol C}/(\text{m}^2 \cdot \text{a})$. The latter may have been caused by the upwelling in the area (Yu, 2010).

Another equation to calculate POC export flux at steady state was developed by Eppley (1979), which

assumes the residence time of ^{210}Po and POC as

$$E_{\text{POC-flux}} = f_{\text{POC}} = f_{\text{POCinv}} / \tau_{\text{PPo}}, \quad (6)$$

where the f_{POCinv} is export flux of the POC inventory in the euphotic zone. The results (Method E, marked by $E_{\text{POC-flux}}$) derived from Eq. 6 are listed in Table 3. Compared with the export fluxes derived from method B, the results calculated by different methods are almost equal (within 40%; He et al., 2008). Considering errors for the activities of ^{210}Po and content of POC, we assume that the results derived by the above two methods are consistent.

Average POC export fluxes 945 $\text{mmol C}/(\text{m}^2 \cdot \text{a})$ (Method E) and 1 848 $\text{mmol C}/(\text{m}^2 \cdot \text{a})$ (Method B) in the euphotic zone at R03 were much higher than those at R20, 126 $\text{mmol C}/(\text{m}^2 \cdot \text{a})$ (Method E) and 109 $\text{mmol C}/(\text{m}^2 \cdot \text{a})$ (Method B). This shows a decreasing trend northward, comparable to prior work in the same region (Yu et al., 2012). Compared with other reported POC export fluxes (Table 3) in certain areas of the world ocean based on various approaches, the fluxes on the Chukchi Shelf in summer were very high. It is concluded that there is high export production on the Chukchi Shelf in summer. Because available light limits marine primary production at high latitude, this production is increased by sea ice melt during summer. Seawater in the Chukchi Sea is derived from the Bering Sea, with substantial nutrients to support primary production. Consequently, the Chukchi Shelf has a significant role as CO_2 sink during summer.

4 CONCLUSION

In the present study, average dissolved ^{210}Pb at different stations had an increasing trend northward from 1.12 to 1.78 Bq/m^3 during July and August, and average particulate ^{210}Pb 2.22 Bq/m^3 was higher than dissolved ^{210}Pb , at 1.49 Bq/m^3 . Residence time of particulate ^{210}Po was greater than that of dissolved ^{210}Po , indicating rapid biogeochemical cycling in the study area. The highest concentration of POC (601 $\mu\text{g C/L}$) was at station R03 during the first sampling period, which decreased northward during the second sampling period, consistent with the distribution of chlorophyll-*a*. Average POC export fluxes in the euphotic zone were derived by two methods with an irreversible scavenging model. These fluxes had a decreasing trend northward and were comparable to prior work in the study area. This suggests an efficient biological pump in the Arctic Ocean and confirms relatively high new production in that ocean during summer, indicative of a significant role as CO_2 sink. The results may expand knowledge of the carbon cycle in the western Arctic Ocean.

Table 3 Inventory and POC export fluxes in euphotic zone, and summary of POC export fluxes in areas of the world ocean

Station	Time	Euphotic layer (m)	POC inventory	POC/ P_{Po}	$E_{POC-flux}$	$B_{POC-flux}$	Reference
			(mmol/m ²)	(mmol/Bq)	(mmol C/(m ² ·a))		
R03	July, 2010	0–30	355.70	45.91	945±350	1 848±848	This work
R09	July, 2010	0–30	437.50	17.44	651±514	1 015±015	
R12	Aug, 2010	0–40	129.17	18.06	84±4.	699±990	
R20	Aug, 2010	0–75	103.75	3.15	126±26	109±09	
	Prydz Bay (March, 2008)				12 045–108 551		Ma et al., 2014
	Prydz Bay (Jan.–Feb., 2006)				19 000–67 525	10 767–95 776	He et al., 2008
	Chukchi Shelf (July–Sep., 2008)				3 029–28 908		Yu, 2010
	Chukchi Shelf (Aug.–Sep., 1994)				13 870		Moran et al., 1997
	Chukchi Continental (Aug.–Sep., 1994)				292–14 235		Moran et al., 2005
	Chukchi Continental (July–Sep., 2008)				657–912.5		Yu, 2010
	Chukchi Continental (Aug.–Sep., 1994)				1 533–20 878		Trimble and Baskaran, 2005
	Canada Basin (July–Sep., 1999)				365		Chen et al., 2003
	Canada Basin (Aug., 2000)				511		Trimble and Baskaran, 2005
	Western Arctic (1998)				2 701–4 635.5		Baskaran et al., 2003
	Subarctic northwest of North Pacific Ocean (May 1996, Feb. 1997, Aug. 1996)				1 022–2 591		Charette et al., 1999
	Weddle Sea (Spring bloom of 1992)				7 300–14 600		Rutgers Van Der Loeff et al., 1997

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