



²¹⁰Po and ²¹⁰Pb as Tracers of Particle Cycling and Export in the Western Arctic Ocean

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The distribution and vertical fluxes of particulate organic carbon and other key elements in the Arctic Ocean are primarily governed by the spatial and seasonal changes in primary productivity, areal extent of ice cover, and lateral exchange between the shelves and interior basins. The Arctic Ocean has undergone rapid increase in primary productivity and drastic decrease in the areal extent of seasonal sea ice in the last two decades. These changes can greatly influence the biological pump as well as associated carbon export and key element fluxes. Here, we report the export of particulate organic and inorganic carbon, particulate nitrogen and biogenic silica using ²¹⁰Po and ²¹⁰Pb as tracers for the seasonal vertical fluxes. Samples were collected as a part of US GEOTRACES Arctic transect from western Arctic Basin in 2015. The total activities of ²¹⁰Po and ²¹⁰Pb in the upper 300 m water column ranged from 0.46 to 16.6 dpm 100L⁻¹ and 1.17 to 32.5 dpm 100L⁻¹, respectively. The ²¹⁰Pb and 210 Po fluxes varied between 5.04–6.20 dpm m⁻² d⁻¹ and 8.26–21.02 dpm m⁻² d⁻¹, respectively. The corresponding particulate organic carbon (POC) and particulate nitrogen (PN) fluxes ranged between 0.75–7.43 mg C m⁻² d⁻¹ and 0.08–0.78 mg N $m^{-2} d^{-1}$, respectively, with highest fluxes observed in the northern ice-covered stations. The particulate inorganic carbon (PIC) and biogenic silica (bSi) fluxes were extremely low ranging from 0 to 0.14 mg C m⁻² d⁻¹ and 0.14 to 2.88 mg Si m⁻² d⁻¹, respectively, at all stations suggesting absence of ballast elements in facilitating the biological pump. The variability in POC fluxes with depth suggest prominent influence of lateral transport to downward fluxes across the region. The results provide a better understanding of the spatial variability in the vertical fluxes POC, PN, bSi, and PIC in the western Arctic which is currently undergoing dramatic changes.

Keywords: export production, POC flux, particle export, Arctic Ocean, organic matter export

INTRODUCTION

The Arctic Ocean covers an area of 9.6×10^6 km² (Serreze et al., 2006), which corresponds to 5% of the world oceans by surface area, with 1.5% by volume (Meybeck and Ragu, 1997; Guay and Falkner, 1998) indicating that it is a relatively shallow ocean with about 50% of the total Arctic Ocean surface area as continental shelf area (Jakobsson et al., 2004). Approximately 10% of the world's rivers discharge flow into the Arctic Ocean making it one of the most dynamic systems, similar

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to estuarine systems (Aagaard and Carmack, 1989; Meybeck and Ragu, 1997; Guay and Falkner, 1998; Dickson et al., 2007). The catchment area of these rivers is estimated to store about 30% of the world's soil carbon (Hugelius et al., 2014; Schuur et al., 2015). Thus, recent climate changes can have profound impacts on the Arctic region resulting in increased export of organic carbon, and associated trace elements and sediments to the Arctic Ocean (Rachold et al., 2000; Moran et al., 2005; Jorgenson et al., 2006; Schuur et al., 2008; Lannuzel et al., 2020; and reference therein).

The input of freshwater from Arctic rivers into the Arctic Ocean has increased significantly during recent decades (Peterson et al., 2002; Ahmed et al., 2020). This increase in freshwater discharge, coupled with increased coastal erosion, permafrost thaw and sea ice melting suggest that the riverine input of carbon, nutrients, and metals will continue to increase in the future. The highly productive shallow shelf serves as a source of organic carbon and nutrients to the central Arctic Ocean through the two major currents systems, Transpolar Drift (TPD), and Beaufort Gyre (e.g., Wheeler et al., 1997; Krishnamurthy et al., 2001; Klunder et al., 2012; Charette et al., 2020). The Arctic Ocean has also undergone more than 20% increase in primary productivity in the last two decades due to decrease in the areal extent of sea ice cover (Arrigo and van Dijken, 2011). The sea ice in this region serves as a platform for retaining atmospherically delivered particle-reactive species, as well as a vehicle for transport of organic and inorganic species incorporated into sea ice formed in the coastal areas (e.g., Krishnamurthy et al., 2001; Tovar-Sánchez et al., 2010). Thus, changes in freshwater input and extent of sea-ice melting can greatly influence the biological pump and particle scavenging in this region.

The present research focuses on the Western Arctic Canada Basin where the vertical fluxes of biogenic materials are reported to be lowest compared to the global ocean, with only 1–2% of the new production reaching the deep basin (Honjo et al., 2010; Hwang et al., 2015). However, expected changes in freshwater discharge will affect the lateral transport of particles into the interior basin (Fahl and Nöthig, 2007; Hwang et al., 2008; Honjo et al., 2010). Water mass exchange between the highly productive shelf and interior basin has been reflected in the observed shelf scavenging signal into the interior basin (Moore and Smith, 1986; Smith et al., 2003). The biogeochemistry in the western Arctic Basin can thus be significantly altered by the influx of shelf-derived materials, and their subsequent transport by TPD (Kipp et al., 2018).

A better understanding of particle cycling and their export to deeper ocean is crucial considering the rapid changes occurring on multiple fronts in this region. The naturally occurring radioisotope pairs from ²³⁸U decay series such as ²³⁴Th – ²³⁸U, ²¹⁰Pb – ²²⁶Ra, and ²¹⁰Po – ²¹⁰Pb have been widely used as tracers of particulate matter in the water column, especially in the estimation of the export flux and cycling of particulate matter. Most of the ²²⁶Ra ($t_{1/2} = 1602$ y) in the oceanic water column in deep ocean basins are derived from bottom sediment via diffusion (Cochran, 1992). ²¹⁰Pb (T_{1/2} = 22 y) is produced from *in situ* decay of its grandparent ²²⁶Ra (which has highest concentration near bottom water) present in the water column as well as direct atmospheric deposition (highest concentration at air-sea

interface). ²¹⁰Po is derived from decay of ²¹⁰Pb in the water column. The ²¹⁰Po/²¹⁰Pb activity ratio in atmospheric deposition is generally < 0.1 (Baskaran, 2011), and thus acts as a minor source of ²¹⁰Po in the surface ocean. Both the ²¹⁰Pb and ²¹⁰Po are highly particle reactive, with partition coefficient (K_d) values of 10⁴-10⁶ and 10⁵-10⁹, respectively (Baskaran and Santschi, 2002; Su et al., 2017; Tang et al., 2017; Bam et al., 2020) and hence are scavenged by both suspended and sinking particulate matter. Higher partition coefficient values represent higher affinity of elements to be adsorb/attach to particles. A deficiency of ²¹⁰Po relative to ²¹⁰Pb is often observed in the upper few hundred meters of the water column due to higher scavenging efficiency of Po (Bacon et al., 1976; Nozaki et al., 1976). Po has stronger affinity for biogenic particulate matter and thus preferential removal of ²¹⁰Po takes place in the upper water column. While ²¹⁰Pb is only adsorbed onto particle surfaces, ²¹⁰Po is also assimilated into phytoplankton cells (Bacon et al., 1976; Nozaki et al., 1976; Fisher et al., 1983). The preferential adsorption and accumulation of ²¹⁰Po in organic matter is well documented in both lab culture and field studies (Fisher et al., 1983; Stewart and Fisher, 2003a,b). Similarly, preferential adsorption of ²¹⁰Pb compared to ²¹⁰Po onto silicious frustules had been previously reported (Friedrich and Rugters van der Loeff, 2002; Lin et al., 2021). The ²²⁶Ra - ²¹⁰Pb and ²¹⁰Pb - ²¹⁰Po pairs can thus be used to trace particle transport processes and quantify chemical scavenging and particle removal rates in the upper ocean for time scales of months to years.

In this article, we utilize these two isotope pairs to investigate the spatial variability in fluxes of particulate organic carbon (POC), particulate inorganic carbon (PIC), particulate nitrogen (PN), and biogenic silica (bSi) across the open water to permanently ice-covered region of the western Arctic Ocean. The spatial and seasonal changes in sea-ice cover, primary productivity, riverine input of freshwater, sediment resuspension, physical processes, and strong halocline play important roles in the distribution of particles, nutrients, and trace elements in the Arctic Ocean (Rachold et al., 2004; Lepore et al., 2009; Chen et al., 2012; Jeandel et al., 2015). Here, we report export fluxes of POC (using POC/²¹⁰Po ratio), PN (using PN/²¹⁰Po ratio), bSi (using bSi/²¹⁰Pb ratio), and PIC (using PIC/²¹⁰Pb ratio) from upper 250 m of the water column.

MATERIALS AND METHODS

Study Area

Arctic water comprises of several distinct water masses which include the high salinity and low temperature Pacific water coming through the Bering Strait, freshwater from river discharge, and high salinity and high temperature Atlantic water coming through Fram Strait (Moore and Smith, 1986; Rutgers Van Der Loeff et al., 1995; Hu et al., 2014). The largest freshwater inventory is found in the upper 300 m of the Canadian Basin (Yamamoto-Kawai et al., 2008; Rabe et al., 2011). Pacific water contributes to the surface and halocline waters of the Canadian Basin (Bauch et al., 1995). Pacific water of winter origin tends to enter the interior Arctic below the upper mixed layer because of their higher salinity and creating the lower halocline water masses (Weingartner et al., 1998; Zhong et al., 2019). Warmer Pacific and colder Atlantic water masses make the system highly stratified and lead to formation of two distinct halocline; the upper and the lower halocline (Smith et al., 2003; Lepore et al., 2009; Zhong et al., 2019). Halocline waters from the shelf seas are the key source of high nutrients in the Arctic basin (Aagaard et al., 1981; Moore and Smith, 1986; Fripiat et al., 2018; Granger et al., 2018).

The upper halocline is mainly confined to the Canadian Basin and is characterized by high nutrients and low dissolved oxygen, whereas the lower halocline is characterized by lower nutrients and higher dissolved oxygen (Jensen et al., 2019, 2020). The compositions of these water masses are greatly altered during their residence on the shelves due to ice formation and melting, primary production, and exchanges with the atmosphere and the seafloor (Rutgers Van Der Loeff et al., 1995).

Sample Collection

Samples were collected during the US Arctic GEOTRACES (GN01) cruise onboard US Coast Guard Cutter Healy (HLY1502) from August 9th to October 11th, 2015. Water samples for dissolved and particulate ²¹⁰Po and ²¹⁰Pb, particulate organic carbon (POC), particulate inorganic carbon (PIC), particulate nitrogen (PN), and biogenic silica (bSi) were collected at Station 30, Station 43, Station 48, and Station 56 (Figure 1). Station 30 and 43 are permanently ice covered, station 48 is seasonally ice covered and station 56 is predominantly open water located on the continental slope (Figure 1). The station 30, 43, and 48 are within the area of TPD and Beaufort Gyre whereas the station 56 is influenced by Pacific Water inflow and Beaufort Gvre. About 20-L water samples were collected at various depths using a 30-L Niskin bottles. For dissolved ²¹⁰Po and 210 Pb, water samples were filtered through a 0.45 μ m cartridge filter within 1-2 h after collection and transferred to acidcleaned cubitainers. The dissolved samples were adjusted to pH 1-2 by adding 6 M HCl within less than 2-3 h after collection and stored onboard. Particulate samples were collected using battery-operated submersible pumps (McLane Research Laboratories, Inc., Falmouth, MA, United States). The pump deployment consisted of a vertical array of pumps at depths coinciding with dissolved samples. For the particulate matter, large-volume water samples (400-600 L) were filtered at a flow rate of \sim 6 L min⁻¹ through acid-washed 150 mm pre-filters (51 µm polycarbonate screen) and then onto pre-acid-washed, precombusted 1 µm nominal, 150 mm diameter Quartz Microfiber Filter (QMA) (Whatman, Kent, United Kingdom) to capture suspended particles. The particles captured on 1 and 51 μ m filters are referred to as small and large particles, respectively.

Dissolved and particulate ²¹⁰Po and ²¹⁰Pb analysis were carried out in the lab following procedures outlined in Bam et al. (2020). Plating of Po was done by spontaneous electrodeposition onto silver planchets, following methods described in Geotraces Cookbook (2017) and Bam et al. (2020). The average time between sample collection and initial plating of ²¹⁰Po for dissolved and particulate samples was approximately 1.5–2 months. The final Po-Pb data was appropriately corrected for decay and ingrowth between the sampling and first plating along with other radiometric decay correction (Baskaran et al., 2013; Bam and Maiti, 2021). Data on POC, PN, PIC, bSi, and $\delta^{13}C_{POC}$ data were obtained from BCO-DMO website and details on sampling and analytical methods are reported in Xiang and Lam, 2020 and (Lam, 2020)¹. The $\delta^{13}C$ -DIC data were also obtained from BCO-DMO website (Quay, 2019)² and sampling and analytical methods are reported in Ko and Quay (2020). ²²⁶Ra activities were also obtained from BCO-DMO website (Charette and Moore, 2020)³ and the details for sampling and analytical methods were reported in Kipp et al. (2019). Temperature, fluorescence, dissolved oxygen, and salinity in-situ data were collected using the shipboard CTD at each station and are reported in BCO-DMO website (Cutter et al., 2019)⁴.

Pb and Po Export Models

The major source of ²¹⁰Pb in seawater is from *in-situ* decay of ²²⁶Ra and atmospheric deposition. The rate of change of ²¹⁰Pb activity can be expressed as follows (Moore and Smith, 1986; Friedrich and Rugters van der Loeff, 2002; Smith et al., 2003):

$$\frac{\partial A_{Pb}}{\partial t} = (A_{Ra} - A_{Pb})\lambda_{Pb} + I_{Pb} - F_{Pb} - V \tag{1}$$

where, A_{Ra} and A_{Pb} are the activities of dissolved ²²⁶Ra (dpm L⁻¹) and total ²¹⁰Pb (dpm L⁻¹), λ_{Pb} is decay constant (d⁻¹) of ²¹⁰Pb, I_{Pb} is the atmospheric depositional input flux of ²¹⁰Pb assumed to be 0.17 dpm cm⁻¹ y⁻¹, based on the mean value (range 0.12–0.22 dpm cm⁻¹ y⁻¹) of previous estimates by Baskaran (2011), F_{Po} is the export flux of ²¹⁰Pb (dpm m⁻² d⁻¹) on to sinking particles and V is the sum of advective and diffusive fluxes of ²¹⁰Pb activities (dpm m⁻² d⁻¹).

Similarly, the major source of ²¹⁰Po in the water column is from the in-situ decay of its longer-lived grandparent ²¹⁰Pb. ²¹⁰Po can also be added via atmospheric deposition from the decay of ²¹⁰Pb within the atmosphere, but this activity usually represents < 10% of the ²¹⁰Pb activity because of the short atmospheric residence time of ²¹⁰Pb-containing aerosols (Harada et al., 1989; Kim et al., 2005; Baskaran, 2011). The rate of change of ²¹⁰Po due to export can be expressed as follows: (e.g., Bacon et al., 1976; Moore and Smith, 1986):

$$\frac{\partial APo}{\partial t} = (A_{Pb} - A_{Po})\lambda_{Po} + I_{Po} - F_{Po} - V$$
(2)

where, I_{Po} is the atmospheric depositional input flux of ²¹⁰Po (dpm cm⁻¹ y⁻¹) which is extremely low for this region (Baskaran, 2011), A_{Pb} is the total ²¹⁰Pb activity in the water column (dpm L⁻¹), A_{Po} is the total ²¹⁰Po activity (dpm L⁻¹), λ_{Po} is the decay constant (d⁻¹), F_{Po} is the export flux of ²¹⁰Po (dpm cm⁻¹ y⁻¹) on sinking (large) particle, and V is the sum of advective and diffusive fluxes of ²¹⁰Po activities (dpm cm⁻¹ y⁻¹).

The export fluxes of ²¹⁰Pb can be estimated using a 1-D steady state model (i.e., $\partial APb/\partial t = 0$ total ²¹⁰Pb remain constant),

¹https://www.bco-dmo.org/dataset/807340

²https://www.bco-dmo.org/dataset/751211

³https://www.bco-dmo.org/dataset/718440

⁴https://www.bco-dmo.org/dataset/651599/data



²¹⁰Po and ²¹⁰Pb were collected onboard US Coast Guard Cutter *Healy* (HLY1502) from August 9th to October 11th, 2015. The arrows represent the m circulation pattern of the surface layer water mass. The map was created using Ocean Data View 5.2.0.

which assumes negligible advective and diffusive fluxes (V = 0) as follows:

$$F_{Pb} = \int_0^z \lambda_{Pb} \left(A_{Ra} - A_{Pb} \right) dz + I_{Pb} \tag{3}$$

where, F_{Pb} is the ²¹⁰Pb flux, A_{Ra} is the total activity of ²²⁶Ra in the water column, A_{Pb} is the total activity of ²¹⁰Pb in the water column, λ_{Pb} is the ²¹⁰Pb decay constant, I_{Pb} is the atmospheric depositional input flux of ²¹⁰Pb, and *z* is the integrated thickness of the water column layer. Similarly, the export fluxes of ²¹⁰Po can be estimated using a 1-D steady state model (i.e., $\partial APo/\partial t = 0$, total ²¹⁰Po remain constant), which assumes negligible advective and diffusive fluxes (V = 0) and negligible ²¹⁰Po atmospheric flux (considering the fact that ²¹⁰Po/²¹⁰Pb activity in atmospheric deposition is < 0.1) as follows:

$$F_{Po} = \int_0^z \lambda_{Po} \left(A_{Pb} - A_{Po} \right) dz \tag{4}$$

where, F_{Po} is the ²¹⁰Po flux, A_{Pb} is the total activity of ²¹⁰Pb in the water column, A_{Po} is the total activity of ²¹⁰Po in

the water column, λ_{Po} is the ²¹⁰Po decay constant, and *z* is the integrated thickness of the water column layer. However, western Arctic Ocean can be influenced by shelf interaction which could overestimate the ²¹⁰Po and ²¹⁰Pb flux due to ²¹⁰Po and ²¹⁰Pb deficit in shelf water. Previous studies have shown lateral input and transport of materials in the western Arctic from the continental shelf areas (Lepore et al., 2009; Kipp et al., 2018; Rutgers Van Der Loeff et al., 2018). The melting ice can also contribute to water column ²¹⁰Pb and ²¹⁰Po inventory, but previous studies have suggested melting of all sea-ice present in the Arctic will only contribute to 10% of the ²¹⁰Pb inventory (Masqué et al., 2007; Roca-Martí et al., 2016). Moreover, such contribution from ice is likely to have ²¹⁰Po-²¹⁰Pb in equilibrium and will have negligible impact on ²¹⁰Po flux estimates in the Arctic.

The ²¹⁰Po has higher particle affinity for biogenic material such as POC and PN whereas the ²¹⁰Pb has higher affinity for species such as PIC and bSi (Fisher et al., 1983; Friedrich and Rugters van der Loeff, 2002). Thus, ²¹⁰Po fluxes can be used to estimate the sinking fluxes of POC and PN and ²¹⁰Pb fluxes can be used to estimate the sinking fluxes of PIC and bSi. The POC flux can be calculated as follows.

$$F_{POC} = F_{210Po} \left[\frac{POC}{APo} \right]_{particles}$$
(5)

where, F_{POC} is the flux of POC, F_{210Po} is the flux of 210 Po, and A_{Po} is the activity of 210 Po in the particulate phase and POC is the POC concentration in the particles. The PN, PIC, and bSi fluxes were also calculated in a similar method as the POC flux using equation 5.

Estimation of export of particulate phases in the water column, using equations 3 and 4 involve the following assumptions: (i) the atmospheric input of ²¹⁰Pb and ²¹⁰Po in the surface ocean is at steady state on time scale that we integrate for the estimation of POC fluxes. Previous studies from this region have shown the atmospheric input in the Arctic (60-80 °N), to be extremely low ranging between 0.12 and 0.22 dpm $cm^{-2} y^{-1}$ (Baskaran, 2011); (ii) the elemental to radionuclide ratios measured in particles collected by in situ pumps are representative of sinking particles. In this study, the elemental ratios in both the small (1-51 um) and large (>51 um)particles were measured, we used both the ratios in large and small particles to estimate the export fluxes. However, we primarily focused on the larger particle export similar to earlier studies (Stewart et al., 2007; Maiti et al., 2016; Roca-Martí et al., 2016); (iii) the one-time sampling in the summer is representative for much longer time scales of months to years over which ²¹⁰Po and ²¹⁰Pb fluxes integrates. This region is characterized by phytoplankton bloom at the onset of summer which could result in non-steady condition. However, our sampling was carried out in late summer and the longer halflife of 210 Po (t_{1/2} = 138 d) results in flux integrated over timescales of months, making it less sensitive to short-term non-steady state events.

RESULTS

Hydrological Parameters

The national snow and ice data center sea ice index suggests that permanent sea ice (multi-year) is present throughout the year at stations 30 and 43, station 48 is covered with sea ice most of the time during the year whereas station 56 is rarely covered with ice. The mixed layer, defined as the depth where density increased from its surface value by 20% of the difference surface and 100 m, varied between 40 and 50 m in all four stations (Shaw et al., 2009). The top 50-75 m water column is strongly stratified as shown by the temperature and salinity transect (Figures 2A,B). The stations 30 and 43 had maximum chlorophyll-a concentration in the upper 10 m of the water column, whereas stations 48 and 56 had maximum chlorophyll-a concentrations at 50-60 m (Figure 2C). The presence of surface maximum chlorophyll concentration in stations 30 and station 43 could be attributed to melting of sea ice (sea ice present during the sampling) with high concentrations of algae in sea ice. The dissolved oxygen (DO) varied between 7.02 and 12.2 mg L^{-1} , with station 30 having higher DO in the mixed layer (Supplementary Figure 1). In the upper 500 m water column, the silicate, nitrate, and phosphate concentrations varied between 0 and 50 μ mol kg⁻¹, 0 and 20 μ mol kg⁻¹ and 0 to 2.5 μ mol kg⁻¹, respectively, with significantly higher concentrations between 100 and 250 m (Figure 3). The station 30 had the lowest nutrient concentration which increased from North to Southward. The nutrient maximum layer showed a similar pattern to temperature and salinity profiles corresponding to the distinct water masses. The spatial distribution of nutrient in the Arctic water column is mainly influenced by the water masses specially the warm Pacific Water (Jensen et al., 2019, 2020).

Distribution of ²²⁶Ra, ²¹⁰Pb, and ²¹⁰Po in the Water Column

The ²²⁶Ra activity in the upper 300 m water column ranged from 7.53 ± 0.28 to 14.5 ± 0.5 dpm $100L^{-1}$ with lower ²²⁶Ra activities at stations 30 and 43 compared to stations 48 and 56 (Figure 4). Elevated ²²⁶Ra activities were observed at depths of 75-100 m (station 43), 65-190 m (station 48), and 82-237 m (station 56) (Figure 4). The total ²¹⁰Po and ²¹⁰Pb activities (dissolved and particulate) in the upper 300 m water column varied between 0.47 ± 0.06 to 8.2 ± 1.2 dpm $100L^{-1}$ and 1.18 ± 0.09 to 11.0 ± 1.0 dpm $100L^{-1}$, respectively (Figure 4). There is deficiency of ²¹⁰Po with respect to ²¹⁰Pb throughout most of the upper 500 m water column, with exceptions at 225 m for station 43 and at 56 m for station 56, where ²¹⁰Po activities and ²¹⁰Pb were similar within uncertainties. The particulate ²¹⁰Po and ²¹⁰Pb activities in the large particles varied between 0.016 \pm 0.001 to 2.32 \pm 0.04 dpm 100L^{-1} and 0.054 \pm 0.004 to 2.50 \pm 0.06 dpm 100L⁻¹, respectively, with stations 30 and 43 having lower particulate ²¹⁰Po activities (Figure 5). The total particulate (large + small) activities of ²¹⁰Po and ²¹⁰Pb followed a similar trend (Supplementary Figure 2) with an exception at station 56 where the ²¹⁰Po activities were higher at 25 and 56 m depth and similar at 86 m depth. The ²¹⁰Pb and ²¹⁰Po data for dissolved and



particulate samples are archived on BCO-DMO website⁵, ⁶ and ⁷ (Baskaran and Krupp, 2020; Maiti and Bam, 2020a,b). The total ²¹⁰Pb/²²⁶Ra activity ratio in the mixed layer (50 m)

The total ²¹⁰Pb/²²⁶Ra activity ratio in the mixed layer (50 m) varied from 0.12 to 1.1 showing a progressively decreasing trend toward north. The total activity ratio of ²¹⁰Po/²¹⁰Pb ranged from

⁵https://www.bco-dmo.org/dataset/808151

0.19 to 1.11 and was <0.75 for 90% of the sample in the upper 300 m water column, indicating high particle scavenging intensity and low remineralization rate of organic matter.

Distribution of Particulate C, N, and bSi in the Water Column

The POC and PN concentrations ranged from 0.002 \pm 0.001 to 0.158 \pm 0.002 $\mu mol~L^{-1}$ and below detection limit to 43.9 \pm 0.2

⁶https://www.bco-dmo.org/dataset/808502

⁷https://www.bco-dmo.org/dataset/794064



nmol L⁻¹, respectively (**Supplementary Figure 3**). The PIC and bSi concentration ranged from below detection limit to $0.005 \pm 0.001 \ \mu$ mol L⁻¹ and 0.78 ± 0.29 to 85.5 ± 0.3 nmol L⁻¹, respectively (**Supplementary Figure 3**). The POC content was higher for station 30 compared to other stations, whereas PIC content was higher for station 43 and 48 compared to station 30. The PN and bSi concentrations were lowest for station 56, which also had lower PIC concentrations. In general, POC, PIC,

PN, and bSi concentration decreased significantly with depth in the upper 100 m.

Radionuclide and Elemental Fluxes

Water column ²¹⁰Pb and ²¹⁰Po fluxes were calculated according to eqn 3 and 4, respectively, at the base of the euphotic zone (100 m) as well as at 50 m and 100 m below the base of the euphotic zone (150 and 200 m water depth). The ²¹⁰Pb fluxes ranged



between 5.04 \pm 0.92 and 6.20 \pm 0.92 dpm m⁻², d⁻¹ with highest 210 Pb flux at station 48 and lowest at station 56 (**Figure 6A**). The 210 Pb fluxes for all the stations were similar within uncertainties. The 210 Po flux ranged between 8.26 \pm 0.89 and 21.0 \pm 1.9 dpm m⁻² d⁻¹ with highest 210 Po flux observed at station 56 and lowest at station 30 (**Figure 6B**).

In order to translate these water column fluxes of 210 Po and 210 Pb to elemental fluxes, corresponding depth distribution of POC/ 210 Po, PN/ 210 Po, PIC/ 210 Pb, and bSi/ 210 Pb ratio at each station is needed (**Figure 7**). The POC/ 210 Po ratio ranged between 1.04 \pm 0.41 and 257 \pm 11 μ mol dpm⁻¹ (**Figure 7A**),

PN/²¹⁰Po ranged from 0.11 ± 0.06 to $35.2 \pm 1.5 \,\mu$ mol dpm⁻¹ (**Figure 7B**). PIC/²¹⁰Pb ranged from 0.13 ± 0.03 to $2.12 \pm 0.63 \,\mu$ mol dpm⁻¹ (**Figure 7C**) and bSi/²¹⁰Pb ranged from $0.19 \pm 0.07 \,$ to $18.1 \pm 0.9 \,\mu$ mol dpm⁻¹ (**Figure 7D**). On average, station 30 had higher ratio of POC/²¹⁰Po and PN/²¹⁰Po and station 43 had higher bSi/²¹⁰Pb ratio than other stations. These values are comparable to the previously reported water column activities from the region (He et al., 2015; Roca-Martí et al., 2016).

The POC, PN, PIC, and bSi export fluxes were calculated according to eqn 5 at the base of euphotic zone (100 m) as well as at 150 and 200 m depth horizons to estimate the production



export and flux attenuation with depth. The POC flux varied between 1.59 \pm 0.16 and 7.43 \pm 0.41 mg C m $^{-2}$ d $^{-1}$ at 100 m (Figure 8A). Similarly, it varied between 0.75 \pm 0.07 and 7.23 \pm 0.41 mg C m $^{-2}$ d $^{-1}$ at 150 m and 1.1 \pm 0.10 to 4.86 \pm 0.40 mg C m $^{-2}$ d $^{-1}$ at 200 m (Figure 8A). The POC flux was highest at station 43 and lowest at station 56. The PN flux ranged from 0.18 \pm 0.02 to 0.78 \pm 0.04 mg N m $^{-2}$ d $^{-1}$ at 100 and 200 m, ranged from 0.08 \pm 0.008 to 0.65 \pm 0.04 mg N m $^{-2}$ d $^{-1}$ (Figure 8B). The flux was higher at the ice stations compared to open water stations with decreasing trend from North to

South. The spatial distribution trend of POC and PN fluxes were similar. The POC and PN fluxes estimated using POC/Po and PN/Po ratio in 1–51 um size fraction were in general found higher by up to a factor 2 compared to those estimated using the POC/²¹⁰Po ratio in the >51 um size fraction (**Supplementary Tables 1, 2**). This observation is similar to what other studies have reported and can be attributed to the fact that not all suspended particles in 1–51 μ m sink fast enough to contribute to the net sinking fluxes (Buesseler et al., 2006; Tang and Stewart, 2019; Bam and Maiti, 2021). The flux estimates using POC/²¹⁰Po ratio on smaller size fraction particles tend to overestimate when



compared with sediment traps, as a significant portion of this pool can be slow sinking particles whose contribution to net flux is reduced by remineralization in the water column. Thus, we suggest that the flux estimate is probably lower bound by the estimates from large particle ratio and upper bound by estimates from small particle ratio.

The PIC fluxes in all stations were low and varied from below detection limit to 0.042 \pm 0.007 mg C $m^{-2}~d^{-1}$ at 100 m, 0.010 \pm 0.001 to 0.145 \pm 0.023 at 150 m, and 0 to $0.059 \pm 0.009 \text{ mg C m}^{-2} \text{ d}^{-1}$ at 200 m depth (Figure 8C). Overall, station 48 had the highest and station 56 had the lowest PIC flux (Figure 8C). The bSi flux ranged from 0.67 \pm 0.12 to 1.54 ± 0.26 mg Si m⁻² d⁻¹ at the euphotic zone (**Figure 8D**). The bSi flux ranged from 0.14 \pm 0.02 to 2.88 \pm 0.46 and 0.14 \pm 0.02 to 1.25 ± 0.19 at 150 m and 200 m depths, respectively (**Figure 8D**). Station 43 had the highest bSi flux, followed by stations 30, 48, and 56. The spatial distribution trend of bSi and PIC fluxes were similar expect for station 56, where extremely low PIC fluxes were observed at 100 m and 200 m. PIC and bSi fluxes were also estimated by utilizing Po fluxes (Supplementary Tables 3, 4) which were higher than ²¹⁰Pb estimates. The ²¹⁰Po based fluxes represent the fluxes integrated over shorter time scale whereas the ²¹⁰Pb based fluxes represent the fluxes integrated over a longer time scale. The major reasons for difference in PIC and bSi fluxed based on ²¹⁰Pb and ²¹⁰Po is discussed in the section below.

DISCUSSION

Water Mass Structure and ²²⁶Ra, ²¹⁰Pb, and ²¹⁰Po Distribution

The hydrological characters and elemental distribution in the Western Arctic are mainly determined by water masses with unique thermohaline macronutrient distribution (Aagaard et al., 1985; Aagaard and Carmack, 1989; Rudels, 2015; Carmack et al., 2016). The basin wide water column section of temperature and salinity showed the mixed layer was limited to upper \sim 50 m. During the sampling transect, four major water masses were encountered – the surface polar mixed layer (PML) (0–50 m), upper halocline layer (UHL) (50–150 m), lower halocline layer (LHL) (150–400 m), and Makarov/Amundsen basin single halocline (50–300 m) (Jensen et al., 2019).

The ²²⁶Ra activities are similar in the mixed layers across all the sampling stations and showed a general decrease in activities with depth, similar to earlier results (Moore and Smith, 1986; and other articles). At certain discrete depths, ²²⁶Ra activities were elevated and these depths also correspond to nutrient maximum in the halocline (Figures 3, 4). The general pattern of increase in ²²⁶Ra activities as a function of increase in the silicate concentration were found to be similar to the previously reported relationship showing – Δ^{226} Ra/ Δ Si = 0.1 dpm/ 100 μ mol (Broecker et al., 1976; Moore and Smith, 1986). Previous studies have also reported such concurrent increase in phosphate, silicate, and ²²⁶Ra in the Arctic basin which were attributed to the halocline water (Kinney et al., 1970; Moore et al., 1983; Moore and Smith, 1986). The elevated ²²⁶Ra activities observed between 100 and 200 m at station 56 is likely related to the shelf-modified Pacific inflow of water, whereas the elevated ²²⁶Ra activities observed between 100-200 m and 75-125 m at station 43 and station 48, respectively, are probably associated with Chukchi winter water (CWW), remnant winter water (RWW), and meteoric cold water (MCW) associated with ice melt that has been carried by the TPD (Kipp et al., 2018). The higher nutrients concentrations and ²²⁶Ra activities at this depth likely represent the different water masses and circulation pattern compared to the surface and below the halocline. ²²⁶Ra activities in the upper 100 m were found to be consistently higher than that of total ²¹⁰Pb activities (210 Pb/ 226 Ra < 1) which is due to the low 210 Pb





atmospheric input of 0.12–0.22 dpm cm⁻² y⁻¹ in the Arctic (Baskaran, 2011). In most oceans, the contribution of ²¹⁰Pb from atmospheric fallout to the upper ~500 m is higher and thus, the ²¹⁰Pb/²²⁶Ra activity ratio is usually > 1.0 in surface waters (Bacon et al., 1976; Cochran et al., 1983; Rigaud et al., 2015; Niedermiller and Baskaran, 2019).

The total activities of ²¹⁰Po and ²¹⁰Pb in the water column showed a general decrease in activity with depth (**Figure 4**). In the Arctic Ocean beside atmospheric deposition, additional surface input of ²¹⁰Pb is possible during the melting of sea ice which can trap atmospheric flux of ²¹⁰Pb (Masqué et al., 2007; Chen et al., 2012). The ²¹⁰Pb input from sea-ice melting is probably not significant, as previous estimates indicate that melting all of sea ice in the Arctic can only contribute up to 10% of the ²¹⁰Pb inventory in the region (Smith et al., 2003; Masqué et al., 2007; Roca-Martí et al., 2016). However, release of ice-rafted sediment (IRS) from sea ice melt can release a large amount of ²¹⁰Pb, as its concentration in IRS has been reported to be 1–2 orders of magnitude higher than those in the Arctic benthic sediments (Baskaran, 2005). The largest ²¹⁰Po – ²¹⁰Pb disequilibria were observed in the surface layers of most stations, which is expected due to increased biological scavenging of ²¹⁰Po with respect to ²¹⁰Pb in the euphotic layer as well the atmospheric deposition which has ²¹⁰Po/²¹⁰Pb AR of < 0.1. However, permanently sea ice covered station 30 was an exception where we did not observe a ²¹⁰Po-²¹⁰Pb disequilibrium in the surface layer (**Figure 4**). The



particulate ²¹⁰Pb activities were also higher in the surface layer which could be attributed to release of particles from melting of sea ice at partially ice-covered stations 43 and 48 and lateral input from the adjacent area for station 56. In the Arctic, particle Po/Pb ratio are mostly < 1 in this study which is consistent with other results from the Arctic but contrast with ratios > 1 reported in other oceanic settings. This is probably due to (i) lower atmospheric input of ²¹⁰Pb in the Arctic and (ii) release of the lithogenic particulate materials during the melting of sea-ice, which contributes higher proportion of ²¹⁰Pb in the water column resulting in Po/Pb ratio < 1. Thus sea-ice dynamics plays an important role in the distribution of particulate ²¹⁰Po-²¹⁰Pb in the Arctic Ocean.

The low activities of $^{210}\rm{Pb}$ and $^{210}\rm{Po}$ and high activities of $^{226}\rm{Ra}$ observed in the nutrient rich water mass indicate

scavenging of ²¹⁰Po and ²¹⁰Pb by particles and lateral transport of shelf-derived ²²⁶Ra (Baskaran et al., 2021). The relative increase in the ²¹⁰Po and ²¹⁰Pb activities below 150 m depth compared to halocline at stations 30 and 43 likely indicate lateral transport of particulate material by the Transpolar drift and Beaufort Gyre. The ²¹⁰Pb and ²²⁶Ra distribution appear to be predominantly regulated by the shelf-basin interactions and the halocline water circulation in this region. The particulate and total ²¹⁰Po and ²¹⁰Pb activities in this study were similar to previously reported activities in this region (Smith et al., 2003; He et al., 2015; Roca-Martí et al., 2018). The activity ratios of ²¹⁰Po/²¹⁰Pb and ²¹⁰Pb/²²⁶Ra showed the preferential scavenging of ²¹⁰Po and ²¹⁰Pb, respectively, throughout the upper 300 m of the water column for all the stations. Further, the spatial pattern in ²¹⁰Po/²¹⁰Pb and ²¹⁰Pb/²²⁶Ra ratios demonstrate low scavenging rates at higher latitudes in the Arctic basin.

²¹⁰Pb and ²¹⁰Po Fluxes

The export fluxes of ²¹⁰Pb at the base of the euphotic zone, were found to be similar for stations 30, 43 and 48 whereas station 56 had lower ²¹⁰Pb flux (**Figure 6A**). The ²¹⁰Pb fluxes increased with depth at all stations. The sharp increase in the ²¹⁰Pb fluxes between 100 and 250 m at stations 48 and 56 can be attributed to the lateral input of particulate matter from the inflow of Pacific water and the Beaufort Gyre. Previous sediment trap studies in the interior Canadian Basin have reported significant contribution of lateral input to the vertical flux (Honjo et al., 2010). We utilized previously reported water column activities of ²²⁶Ra and ²¹⁰Pb (Smith et al., 2003) to estimate ²¹⁰Pb fluxes for three stations in proximity of station 48 and 56. The estimated fluxes for these three stations ranged from 5.32 to 5.48 dpm $m^{-2} d^{-1}$ and 5.94 to 6.38 dpm $m^{-2} d^{-1}$ at 100 m and 200 m, respectively, which is similar to our current estimates (4.99-6.18 $dpm m^{-2} d^{-1}$).

There are spatial differences in ²¹⁰Po fluxes at the four stations, which did not follow any latitudinal trend (Figure 6B). Station 56 in the Canadian basin had the highest ²¹⁰Po flux which is expected due to higher particle scavenging in the slope area compared to interior stations. The ²¹⁰Po fluxes varied between $8.26-13.76\ 7.23\ dpm\ m^{-2}\ d^{-1}$ and $12.54-16.89\ dpm\ m^{-2}\ d^{-1}$ at 100 and 150 m, respectively (Figure 6B), which are higher than previously reported ²¹⁰Po fluxes in the Arctic Ocean. Roca-Martí et al. (2016) reported ²¹⁰Po fluxes of 7.2 dpm m⁻² d⁻¹ and 3.7 dpm m⁻² d⁻¹ at 150 m for two stations in the central Arctic which are in proximity of our stations 30 and 43. The higher observed fluxes at stations 30 and 43 is likely due to the sea ice dynamics. Roca-Martí et al. (2016) sampled during the record sea-ice minimum with mostly single year ice whereas during our sampling in 2015 multi-year sea-ice was observed. The particulate ²¹⁰Po and ²¹⁰Pb were found to be orders of magnitude higher during our sampling, pointing to the possible increased input of sediment from melting of multi-year sea-ice which could result in higher ²¹⁰Po fluxes. The ²¹⁰Po fluxes estimated using previously reported data (Smith et al., 2003) ranged between 3.95-6.69 dpm $m^{-2} d^{-1}$ and 5.29–7.42 dpm $m^{-2} d^{-1}$ at 100 m and 200 m depth, respectively. These stations were in proximity to our stations 48 and 56 and the fluxes are lower than our current estimates (8.26-21.0 dpm m⁻² d⁻¹). ²¹⁰Po fluxes in this region is overall lower compared to other regions of the world ocean (Murray et al., 2005; Wei et al., 2011; Anand et al., 2018; Bam and Maiti, 2021). In the future, the ²¹⁰Po and ²¹⁰Pb fluxes in the Arctic might change with changing Arctic climatic conditions such as rapid ice melting, increased ice algae growth and enhanced transport of shelf sediments but our current comparison of fluxes with data collected over two decades back (Smith et al., 2003) showed no significant differences. However, it must be noted that large spatiotemporal variability in this region can result in export fluxes varying by a factor of 7 (Roca-Martí et al., 2016). Thus, it will be difficult to identify any recent changes unequivocally in a single study, unless time series measurements are carried out at a particular site for several years.

Distribution of Elemental and Radionuclide Ratios

The higher concentration of POC and PN in surface water at station 30 (Supplementary Figure 3) could be attributed to the supply of POC and PN from melting of the ice (Yu et al., 2012). In the central Arctic, there is no direct external input of POC (Xiang and Lam, 2020) even though the TPD influences the distribution of trace metal and dissolve organic matter (Kipp et al., 2018; Charette et al., 2020). Overall, the surface water in the top 20 m had the highest concentration of POC, PIC, PN and bSi. PIC concentrations were low throughout this region and was below detection limit at station 56 (Supplementary Figure 3). The coccolithophores are the major source of PIC in the ocean and the lower concentration of PIC indicates the absence of coccolithophores in the Arctic waters (Honjo et al., 2010, Xiang and Lam, 2020 and references therein). The total bSi concentration were relatively higher in the shelf/slope region (station 48 and 56) compared to interior station (30 and 43) (Supplementary Figure 3), which is attributed to the diatoms. In the Arctic Ocean, diatoms are major phytoplankton community responsible for > 45% of the total primary production (Coupel et al., 2012; Balch et al., 2014).

The ratios of POC/210Po, PN/210Po in the water column decreased with depth following a general power law (Figures 7A,B) as previously observed by number of studies (Maiti et al., 2008; Tang and Stewart, 2019; Bam and Maiti, 2021). This is expected because of the remineralization of particulate organic matter during settling through the water column and preferential association of 210 Po with POC (R² = 0.65) and PN ($R^2 = 0.54$). The PIC/²¹⁰Pb and bSi/²¹⁰Pb ratios did not show such well-defined power law attenuation with depth (Figures 7C,D). This is because both bSi and PIC will solubilize with depth, with the solubility of bSi being much faster than PIC in the water column (Cappellen et al., 2002; Rickert et al., 2002). For all four ratios, maximum variability is observed in the upper 100 m layer, which is due to high biological activity in the euphotic zone. The shelf station 56 had the lowest POC/210Po ratio while station 30 had the highest POC/210Po ratios. This gradual poleward increase in POC/210Po ratio could be due to change in the phytoplankton communities as the station transited from open ocean to permanently sea-ice covered. In the western Arctic Ocean, the coccolithophores and foraminifera are nearly absent thus resulting in low PIC/210Pb ratio (Coupel et al., 2012, 2015; Xiang and Lam, 2020). On the other hand, the bSi/²¹⁰Pb ratios in the water column were relatively high compared to other oceans due to the abundance of diatoms in the western Arctic (Lalande et al., 2009; Boetius, 2013).

Elemental Fluxes From the Euphotic Zone

In the Arctic, the export production is high during the early summer. The sampling for this study was carried out from August – October which represents the end of high productive season. The 138 days half-life of ²¹⁰Po results in fluxes integrated over seasonal timescales. Thus, the ²¹⁰Po flux reported here probably reflects average export for the entire summer season.

In the central Arctic, the export fluxes are highest during June-August and ice algae contributes significantly to the export fluxes (Fahl and Nöthig, 2007; Lalande et al., 2009). The ²¹⁰Po based POC flux estimates at 100 m depth varied widely with higher values in the northern ice-covered stations and lowest in the Canada basin (station 56) (Figure 8A). A previous study showed that the POC exports are greater in areas with sea-ice coverage than in open water (Roca-Martí et al., 2016). Similarly, the studies from the central Arctic using different method (²³⁴Th-²³⁸U) showed POC fluxes to be less than 60 mg C m⁻² d⁻¹ (Moran et al., 1997; Baskaran et al., 2003; Roca-Martí et al., 2016). In contrast, He et al. (2015) reported ²¹⁰Po based POC export fluxes to vary between 2.7 and 31 mg C m⁻² d⁻¹ with fluxes decreasing northward. A comprehensive analysis of reported POC flux in the published literature from the Arctic Ocean shows that POC export fluxes vary between 0.23 and 216 mg C m⁻² d⁻¹ (Table 1) which are difficult to compare given the inherent differences in various flux estimation methods (Maiti et al., 2016; Anand et al., 2018). For example, the POC flux estimated using ²³⁴Th-²³⁸U methods are higher than ²¹⁰Po-²¹⁰Pb and traps in the compiled literature (Table 1). The highest fluxes are reported from the marginal seas where biological productivity is expected to be higher while the lowest fluxes are in the central Arctic. Our study indicates that the POC export at the pole is higher than other parts of the deep basin pointing to the importance of sea-ice algae in driving POC fluxes in ice covered region. Ice algae and diatoms are the major component influencing the export fluxes of production in the Arctic Ocean (Lalande et al., 2009; Boetius, 2013).

With increasing freshwater input and associated terrestrial organic matter in the Arctic, it is crucial to understand the source of the POC contributing to this downward flux. The δ^{13} C

composition of POC indicate predominantly marine origin of
POC and PN at all the four stations (Figure 9A : Xiang and Lam.
2020). The δ^{13} C-DIC in the water column can provide relative
estimate of primary productivity (Ko and Quay 2020) while
δ^{13} C POC in large particle can help understand the source of
POC contributing to fluxes (Griffith et al. 2012: Tolosa et al.
2013: Xiang and Lam 2020) The higher δ^{13} C-DIC values are
associated with enhanced biological productivity in the water
column (Ko and Quay 2020). The value of λ^{13} C-DIC values
ranged from -0.04 to 1.47% with station 48 having higher
values (Figure 9B) indicating higher productivity. The difference
in the λ^{13} C-DIC for the stations suggest the difference in the
magnitude of primary productivity among stations λ^{13} C-DIC
in the top 100 m is usually depleted compared to the surface
due to untake of CO ₂ and incorporation of lighter 12 C by the
nbytoplankton into organic matter (Bauch et al. 2015). The top
100 m is biological active as reflected by the higher fluorescence
content (Figure 2C). Thus S^{13} C DIC water column distribution
suggests that station 48 might have higher primary productivity
followed by station 42 and then station 20. The S ¹³ C DOC.
values were greatly deploted ($20.8 \pm 5.7\%$ to $24.45 \pm 0.4\%$)
values were greatly depicted (-50.8 \pm 5.7% to -24.45 \pm 0.4%),
(Views and Law 2020). Figure 0.4). The S^{13}_{13} C DOC values are
(Alang and Lam, 2020; Figure 9A). The of C POC values are
Similar to values reported by Brown et al. (2014) in the central
Canada Basin. In general, the phytoplankton in open ocean show
3^{10} C values ranging -25 to -18% , nowever, the 3^{10} C POC in
polar systems pelagic and sea ice can exhibit -20 to $-34.\%$
(Sallon et al., 2011; Pineault et al., 2013; Brown et al., 2014).
Further, the highly depleted δ^{13} C POC _{large} might also reflect the
slow growth rates of the phytoplankton (Fry, 1996; Griffith et al.,
2012; Tolosa et al., 2013), and temperature effect (Rau et al.,
1989). This suggests that POC in the upper water column is

TABLE 1 Published data on POC fluxes in the Arctic Ocean.							
Sampling	Sampling	POC flux	Export	Method	References	Note	
Location	Year	mg C m ⁻² d ⁻¹	Depth (m)				
Amundsen Gulf Shelf	1988	14.4	118	Trap	O'Brien et al., 2006	Sea-ice	
Mackenzie Est.	1988	23.8	145	Trap	O'Brien et al., 2006	Near Estuary	
Tuktoyaktuk Shelf	1988	4.2	125	Trap	O'Brien et al., 2006	Shelf Edge	
Laptev Sea	1995–1996	6–30	150	Trap	Lalande et al., 2009		
Lomansov Ridge	1996	3.2	150	Trap	Fahl and Nöthig, 2007	Sea-ice	
Bering Sea	1999	118–164	100	Th-U	Chen et al., 2003		
Fram Strait	2012	21.6-216	100	Th-U	Le Moigne et al., 2015		
Chukchi Rasie	1996	1.61	120	Trap	Honjo et al., 2010	Sea-ice	
Chukchi Shelf	2010	2.7–31	150	Po-Pb	He et al., 2015		
Chukchi Sea	2002	0.37–34.8	150-200	Th-U	Moran et al., 2005	Shelf-slope	
Canada Basin	1996	0.23	200	Trap	Honjo et al., 2010	Sea-ice	
Canada Basin	1998	67–78	100	Th-U	Baskaran et al., 2003		
Canada Basin	1999	12	100	Th-U	Chen et al., 2003		
Canada Basin	2000	31.2	100	Th-U	Trimble and Baskaran, 2005		
Central Arctic	2012	1.2-57.6	150	Po-Pb	Roca-Martí et al., 2016		
Central Arctic	1994	3.6-84	30	Th-U	Moran et al., 1997		
Central Arctic	2012	24–84	150	Th-U	Roca-Martí et al., 2016		
Western Arctic	2003	28.8–243	50	Th-U	Yu et al., 2010		
Western Arctic	2015	0.75-7.4	150	Po-Pb	This study	Basin Wide	



supported by sea ice and not from terrestrial sources, despite the strong riverine input of POC at the coastal and shelf region, which continues to increase due to increasing river discharge and coastal erosion (Rachold et al., 2000, 2004; Krishnamurthy et al., 2001).

The ²¹⁰Po and ²¹⁰Pb fluxes can both be used to estimate the bSi and PIC fluxes. However, there are some fundamental differences on which tracer better represents the fluxes of inorganic material such as bSi and PIC. The key differences are due to (i) the bioaccumulation and adsorption of ²¹⁰Po and ²¹⁰Pb in the phytoplankton and particles and (ii) the flux integration timescale. Previous studies have reported differences in the bioaccumulation of ²¹⁰Po and ²¹⁰Pb in phytoplankton (Fisher et al., 1983; Stewart and Fisher, 2003a,b). ²¹⁰Po is incorporated inside the cells of phytoplankton while the ²¹⁰Pb is mostly bound to cell walls (Fisher et al., 1983; Friedrich and Rugters van der Loeff, 2002). A recent study by Lin et al. (2021) showed the difference in binding capacity of ²¹⁰Po and ²¹⁰Pb in diatoms and coccolithophore. Therefore, it is expected that ²¹⁰Pb would be a better tracer for inorganic components whereas ²¹⁰Po would better reflect the organic component. Using the ²¹⁰Po, ²¹⁰Pb and ²³⁴Th, Friedrich and Rugters van der Loeff (2002) showed that ²¹⁰Po and ²³⁴Th fluxes were more useful to estimate POC and ²¹⁰Pb to estimate bSi. For this study, bSi and PIC fluxes are estimated from ²¹⁰Pb fluxes although bSi and PIC fluxes based on ²¹⁰Po fluxes are also included in supplementary data (Supplementary Tables 3, 4). ²¹⁰Pb fluxes represent longer time scale for the export compared to ²¹⁰Po based fluxes which represent much shorter seasonal scale. Thus, a direct comparison of ²¹⁰Pb based and ²¹⁰Po based fluxes is

not appropriate but in general ²¹⁰Po based fluxes tend to be higher than ²¹⁰Pb fluxes. The bSi and PIC fluxes showed a similar trend with highest fluxes at station 43 followed by stations 30, 48 and 56 (Figures 8C,D). Based on the low PIC and bSi fluxes at station 30, we attribute the POC flux at station 30 is mostly contributed by the sea algae. The PIC fluxes were extremely low because the primary productivity in this region is currently driven by picoplankton, diatoms, and green algae such as prasinophytes (Lalande et al., 2009; Boetius, 2013; Metfies et al., 2016). However, the eco-regime shifts to coccolithophore, and diatoms blooms are expected (Forest et al., 2008; Honjo et al., 2008) which could increase the PIC and bSi fluxes in future. Due to the ballast effect, the increase in the PIC and bSi fluxes could potentially result in increase of POC and PN fluxes. Biominerals or lithogenic particles increase the density of sinking, particles because of their high density and higher specific gravity, thus increasing the sinking rates and POC export fluxes known as ballast effect.

The primary productivity in the Arctic has been increasing over the last couple of decades (Arrigo and van Dijken, 2011), however, this enhanced productivity might not necessarily translate to larger POC exports fluxes due to changing Arctic environmental conditions as new conditions favor phytoplankton community structure which is based on the smaller cells (Li et al., 2009). With the changes in the phytoplankton community structure due to the changes in environmental conditions such as ice melting, nutrient input and temperature, the magnitude of export flux will be impacted. If the smaller cell phytoplankton community is favored, the export flux might decrease while the larger phytoplankton community could lead to increase in flux. Thus, the changing climatic and environmental conditions could directly or indirectly impact not only the marine carbon cycle but also the trace elements and nutrients distribution in the Arctic Ocean.

Particle Flux Attenuation and Lateral Input in Mesopelagic Zone

The POC fluxes at 150 m and 200 m depth were estimated to understand the particle attenuation and the remineralization below the euphotic zone. At station 30, the POC and PN fluxes increased by factor of 2 at 150 m and subsequently decreased to 15% at 200 m depth with respect to the fluxes measured at 100 m (Figure 8A). This increase in the POC and PN fluxes at 150 m could be attributed to lateral transport by the RWW and MCW. The particle attenuation and remineralization were higher at station 43, 48 and 56 which is expected due to high nutrient rich water. At station 56, POC and PN fluxes were 50% lower at 150 m than at 100 m (Figures 8A,B). However, there was 45% increase in POC and PN fluxes between 150 and 200 m which suggests contribution from lateral transport by inflow of Pacific water. The lower halocline layer with Pacific nutrient rich water is located between 150 and 400 m depth which contributed to the lateral transport of organic matter. The bSi flux at 150 m increase at station 30 and 43 whereas it decreased for stations 48 and 56 compared to the flux estimates at 100 m depth (Figure 8D). High concentration of bSi were observed at station 43 and other nearby stations which could possibly be due to sporadic sinking of ice diatoms such as Melsoria arctica (Fahl and Nöthig, 2007; Lalande et al., 2014). Station 43 is covered with the sea ice during most of the time. The RWW and MCW cold water from TPD and the edge of the high nutrient water with sea ice on the top makes this station one of the most dynamics and interesting in terms of biogeochemical interaction. These combined physical and hydrological characteristics probably enhanced the export fluxes.

Export Efficiency

The export efficiency or e-ratio is defined as the ratio of export flux and net primary productivity (NPP). The mean NPP reported were 265 mg C m^{-2} d⁻¹ in the Beaufort Basin, 195 mg C m⁻² d⁻¹ in Chukchi Basin (Arrigo and van Dijken, 2011) and 12-84 mg C m⁻² d⁻¹ in the central Arctic Ocean (Fernandez-Mendez et al., 2015). The average annual export efficiency (>30%) is high in the Arctic Waters (Henson et al., 2015). Since the NPP data during our sampling time was not available, the export efficiency was estimated using the average NPP values for sea-ice covered and open water areas from Fernandez-Mendez et al. (2015),. The export efficiency ranged from 5 to > 50% with lowest export efficiency in the slope (5–15%) and highest in the sea-ice covered stations (12-50%). Previous studies have reported export efficiency > 30% in the Eurasian Basin (Gustafsson and Andersson, 2012), 26% in the Canada Basin (Chen et al., 2003), and >30% in the central Arctic (Roca-Martí et al., 2016) which are within the range of this study. The export efficiencies estimated in the Arctic Ocean are higher compared to other oceans (Buesseler et al., 1992, 1995, 1998; Bacon et al., 1996; Murray et al., 1996; Buesseler and Boyd, 2009; Haskell et al., 2013; Anand et al., 2018; Laws and Maiti, 2019; Bam and Maiti, 2021) probably due to lower metabolic rates associated with colder temperature. The lower export efficiency in the slope (station 56) and central Canada basin (station 48) compared to ice-covered interior stations 43 and 30 suggest higher recycling of the particles in the slope and central Canada basin which could be related to relatively higher microbial respiration at these stations compared to colder ice-covered stations.

CONCLUSION

We estimated the export flux of POC, PN, PIC, and bSi based on the ²¹⁰Po and ²¹⁰Pb fluxes obtained during the 2015 US Arctic GEOTRACES cruise in the western Arctic Ocean. Overall, the vertical fluxes of the four key elements remain extremely low throughout the upper 200 m of the ocean. The low fluxes of bSi and PIC indicate that ballast particle fluxes which represent an important mechanism of POC transport to deep ocean are orders of magnitude lower than other regions of the global ocean, similar to what has been shown using deep sediment trap studies (Honjo et al., 2010). The overall low vertical fluxes of POC and prominent influence of lateral transport presented in this study concurs with previous work which suggested that the biological pump is currently inefficient in the cryopelagic Canada Basin. Thus, in this region a lateral POC pump from the shelf/slope region might be playing a more dominant role in transporting POC to the deeper basin.

Our study showed that the particulate organic matter in the western Arctic basin is predominantly the result of in-situ biological activities supported by biological pump. Thus, any changes to the in-situ primary production in the western region will influence the export efficiency and overall carbon cycling. In this study, the production export showed an increasing tend toward north with station 43 having the highest export fluxes. The projected melting of sea ice in future can result in large changes to the top Polar Mixed layer which can potentially transform the biological pump in this region (Macdonald et al., 2002). Thus, it is pertinent that carbon cycling and accompanying biogeochemical responses in the rapidly changing Arctic be closely monitored as it can have far reaching global impact in the future.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/**Supplementary Material**, further inquiries can be directed to the corresponding authors.

AUTHOR CONTRIBUTIONS

WB: data curation, formal analysis, methodology, validation, writing-original daft, review, and editing. KM: conceptualization, funding acquisition, methodology, supervision, validation, writing-review, and editing. MB: conceptualization, funding acquisition, methodology, writing-review, and editing.

All authors contributed to the article and approved the submitted version.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fmars. 2021.697444/full#supplementary-material

Supplementary Figure 1 | Water column CTD profiles showing Salinity [%], Fluorescence (0- 5 VDC volts), and Dissolved Oxygen [mg L⁻¹] for all the sampling station. The fluorescence has been magnified 100 times to show on the same x-scale. The top X-axis represents the salinity and fluorescence, and the bottom X-axis represents the dissolved oxygen.

Supplementary Figure 2 | The activity depth profile of particulate (large and small) ²¹⁰Po (open circle) and ²¹⁰Pb (filled circle) for the sampling station in the upper 300 m.

Supplementary Figure 3 | POC, PIC, PN, and bSi concentration on the large particle samples for upper 300 m water column depth.

Supplementary Table 1 | POC fluxes in large and small particle fraction sizes using $^{210}{\rm Po}$ flux.

Supplementary Table 2 | PN fluxes in large and small particles fraction sizes using ²¹⁰Po flux.

Supplementary Table 3 | PIC flux estimated based on the ²¹⁰Pb and ²¹⁰Po fluxes in large particles.

Supplementary Table 4 | bSi flux estimated based on the ²¹⁰Pb and ²¹⁰Po fluxes in large particles.

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