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Decadal shift of biogenic sinking particle flux in the western North Pacific subpolar region

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Abstract Time series of biogenic sinking particle flux in the western North Pacific subpolar region over two decades (1989–2008) revealed that the biogenic $CaCO_3$ (CC) flux has shown a significant decreasing trend of 2.7% $year^{-1}$ (annual average, -0.88 ± 0.13 mg m^2 day^{-1} $year^{-1}$) along with the decreasing particulate organic carbon (POC) flux of 0.7% $year^{-1}$, while the biogenic opal (OP) flux had no long-term trend. Comparing these results with the decreasing rate of satellite-derived surface CC with -0.7% $year^{-1}$, we concluded that three fourths of the decreasing trend of CC flux was derived from the strengthening of $CaCO_3$ dissolution through seawater column due to the weakening of water ventilation and the rest was from the decline of $CaCO_3$-shelled species, indicating the enhancement of the efficiency in oceanic sequestration of atmospheric CO_2 in the sea surface of this region due to the increase of OP/CC ratio.

1. Introduction

The biological fixation of CO_2 in the ocean is 45 Pg C $year^{-1}$, which is even now about 7 times larger than the amount of anthropogenic released CO_2 [e.g., *Falkowski et al.*, 1998]. Especially, the production of diatoms with an opal test plays an important role for the rapid and large-scale sinking of carbon to the ocean interior, which accounts for 40% of the global marine primary productivity, while the production of $CaCO_3$ as coccolithophorids and foraminifera induces an escape of CO_2 from sea to air [e.g., *Honda et al.*, 2002]. Thus, the ratio of opal to $CaCO_3$ influences the efficiency in oceanic sequestration of atmospheric CO_2. The recent climate change might have already caused the decadal decrease of opal/$CaCO_3$ ratio in sinking particles in the North Atlantic in the past several decades [*Deuser et al.*, 1995; *Antia et al.*, 2001]. However, since this region is not under silicate-rich condition with higher biological productivity, it is still unknown whether the oceanic change works as a positive or negative feedback against the global change. The North Pacific subpolar region where diatom species are dominant is an important area of oceanic sequestration of atmospheric CO_2 for the global carbon budget [*Honda*, 2003]. If the ocean stratification progresses with the climate change, there is a high probability that we can find evidence of the remarkable change in oceanic sinking particle derived from the climate change.

In this region, recent studies have already shown the increase of water temperature and the changes in hydrographic properties as deoxygenation, suggesting the significant weakening of the surface-deep water mixing derived from the climate change [e.g., *Watanabe et al.*, 2003]. It was pointed out that the possibility of a shift in the phytoplankton species from large size (as diatoms) to small size (as coccolithophorids and dinoflagellates) [*Ishida et al.*, 2009]. Furthermore, the progress of ocean acidification derived from anthropogenic CO_2 increasing has been also reported in the North Pacific [*Feely et al.*, 2008], which can cause the decrease in $CaCO_3$ production. In the western North Pacific subpolar region, *Wakita et al.* [2013] showed that the depth of the saturation state of $CaCO_3$ recently has shoaled to the bottom of surface mixed layer. However, it is still unclear how these biological changes work against the global change. In the western North Pacific subpolar region, we thus tried to clarify how the ratio of biogenic opal/$CaCO_3$ (OP/CC) changes by using a 20 year time series of sinking particle fluxes from 1989 to 2008 (Figure 1).

2. Sampling and Methods

We used time series of sinking particle flux collected by the sediment traps deployed during 1989–2008 at the ocean time series observation site, Station KNOT (44°N, 155°E) within the western subarctic gyre of the North Pacific [*Noriki et al.*, 1999; *Honda et al.*, 2002; *Otosaka and Noriki*, 2005; unpublished new data] (Figure 1). Eleven sediment traps (Nichiyu Co. Ltd., HX-10) were installed in the range from 770 to 1200 m with a sampling time

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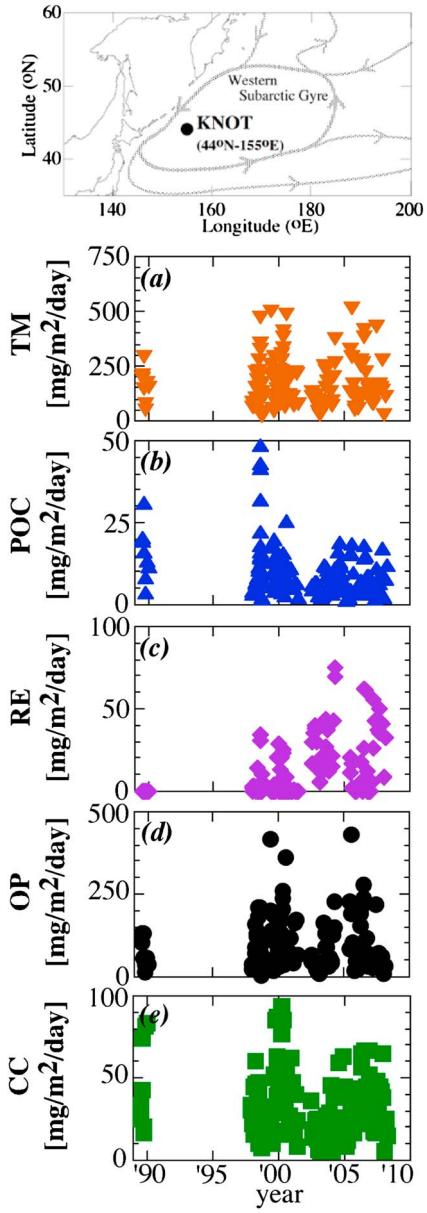

Figure 1. Sampling site for sinking particle fluxes in the western North Pacific subpolar region and the time series of (a) TM flux, (b) POC flux, (c) RE flux, (d) OP flux, and (e) CC flux from 1989 to 2008. Unit for all sinking particle fluxes was mg m^2 day^{-1}. General surface circulation patterns are cited from *Harrison et al.* [1999].

series interval of about 1 month. Each sampling collector was filled with a solution of 10% neutral formaldehyde diluted with filtered seawater, which was prepared to 39 psu by the addition of sodium chloride in order to prevent degradation of organic matter. Samples were kept in the dark at 4°C until analysis. The particulate materials in each sampling collector were filtered through preweighed 0.6 μm Nuclepore filters after removing "swimmers" and then freeze dried overnight. The total mass (TM) flux was estimated from the dry weights of particulate materials collected on the filters [*Shigemitsu et al.*, 2010]. As components of sinking particles, $CaCO_3$ (CC) and particulate organic carbon (POC) were measured by using the coulometric technique with acid addition and the CHN elemental technique [e.g., *Noriki et al.*, 1999] or inductively coupled plasma-atomic emission spectrometry [*Honda et al.*, 2002]. Biogenic organic matter (OM) was estimated by using POC divided by the average stoichiometric ratio of phytoplankton (POC/OM = 0.35) [*Redfield et al.*, 1965]. Biogenic opal ($SiO_2 \cdot 0.4H_2O$, OP) was estimated by using the leaching Si method [e.g., *Noriki et al.*, 1999]. Residue (RE) was defined as subtraction of OP, CC, and OM from TM. The precision of analysis for each component was within ± 5%. Since a vertical flux of the sinking particle changes with depth due to remineralization/dissolution, we here normalized these data to the sample at 1000 m depth according to the concept for reconstructing the vertical distributions of sinking particle at an arbitrary depth [*Honda et al.*, 2002] as follows:

$$CC_{(d)} = CC_{(obs)}(d/z)^{-0.38}, \quad (1)$$

$$POC_{(d)} = POC_{(obs)}(d/z)^{-0.63}, \quad (2)$$

$$OP_{(d)} = OP_{(obs)} \exp(-0.00003(d - z)), \quad (3)$$

where *z*, *d*, and obs are the sampling depth (m), the arbitrary depth (m), and the observed flux of each property. We here used 1000 m as *d*. The changes of sinking particle fluxes derived from the normalization were almost within our analytical errors. Furthermore, in order to complement the time series of sinking particle fluxes in the region, we used time series of satellite-derived chlorophyll *a* (Chl), water temperature (*T*), and CC composite data in the sea surface near Station KNOT (42–46°N, 153–157°E) from 1997 to 2012 [*Goddard Earth Sciences Data and Information Services Center*, 2013].

3. Results and Discussion

3.1. Long-Term Changes of Biogenic Sinking Particle Fluxes

Over the time series, the average values of TM flux, POC flux, RE flux, OP flux, and CC flux were 160 ± 31 mg m^{-2} day^{-1} (standard error), 8.4 ± 1.6 mg m^{-2} day^{-1}, 15 ± 3 mg m^{-2} day^{-1}, 88 ± 25 mg m^{-2} day^{-1}, and 33 ± 1 mg m^{-2} day^{-1}, respectively (Figure 1 and Table 1). The ratio of POC flux to the mean annual primary productivity (80–90 g C m^{-2} $year^{-1}$) [*Imai et al.*, 2002; *Honda et al.*, 2002] was estimated to be 3–5% as an export efficiency from the surface to the ocean interior, which was the highest in the Northern Hemisphere [*Honda et al.*, 2002]. We also found out that the average mole ratio of OP/CC was 4.0 ± 1.2 while both OP flux and CC flux had no long-term trends (Figures 1d and 1e), which was consistent with previous studies [*Honda*, 2003], indicating that the western North Pacific subpolar region is the silica-dominated ocean with a high productivity and it is a crucial ocean to

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Table 1. Long-Term Trends of Biogenic Particulate Organic Flux (POC), Opal Flux (OP), and $CaCO_3$ (CC) Flux Between 1000 m Depth and the Sea Surface in Station KNOT

	POC	OP	CC
In the sampling depth, 1000 m			
Annual mean sinking flux (mg m^{-2} day^{-1}) [a]	8.4 ± 1.6	88 ± 25	33 ± 1
Δ, Change of sinking flux (mg m^{-2} day^{-1} $year^{-1}$) [a]	−0.06 ± 0.02	0 [b]	−0.88 ± 0.13
D, Decreasing rate (% $year^{-1}$) [a]	−0.7 ± 0.2	0 [b]	−2.7 ± 0.4
In the sea surface			
D, Net decreasing rate (% $year^{-1}$) [c]	−1.1 ± 0.5 [d]	-	−0.7 ± 0.5

[a]Annual mean values based on the seasonal composite time series of sinking particle fluxes.
[b]No significant trend.
[c]Annual mean values based on the seasonal composite time series of satellite-derived data.
[d]We estimated *D* based on the time series of satellite-derived Chl data.

understand the oceanic response against the global change due to the ratio of OP/CC being more than 1 of the global mean value [*Honda*, 2003].

However, we found no significant long-term changes of all these particle fluxes due to the large seasonal scatters. In order to make sure whether the long-term changes of biogenic particle fluxes have occurred in this region, it is necessary to examine these fluxes in more detail. Generally, we can find the largest bloom of phytoplankton from March to May and the strong seasonal stratification of surface mixed layer from June to August in this region. From September to November, there is also the autumn bloom of phytoplankton [*Imai et al.*, 2002]. To elucidate the seasonal changes of biogenic particle fluxes, we tried to divide the time series of particle flux into four seasonal periods: winter (December–February), spring (March–May), summer (June–August), and autumn (September–November). In addition, since previous studies reported that many hydrographic properties in this region had shown long-term linear trends superimposed on the decadal oscillation [e.g., *Watanabe et al.*, 2008], we applied an equation of the Fourier sine expansion to the time series of seasonal composite data in order to supplement the time series of biogenic particle fluxes: $X = -a \cdot y + b + c \cdot \sin\{2\pi\ (y - d)/e\}$, where "*X*" refers to one of several properties; "*y*" is the calendar year; and "*a*," "*b*," "*c*," "*d*," and "*e*" are constants. That is, *X* = linear trend component (*L*) + oscillation component (*O*).

These seasonal composite data enabled us to clarify several critical changes in biogenic particle fluxes over the past two decades. TM, POC, and CC fluxes had decadal oscillation of 18.5 ± 1.6 years on average in both the wintertime and the summertime (Figure 2), which synchronized with the oscillations of dissolved oxygen and nutrients (18.6 years) during almost the same period around Station KNOT [e.g., *Watanabe et al.*, 2008]. What has caused the decadal oscillations of these particle fluxes? Taking notice of the interaction between the atmosphere and the ocean, we here focused on the North Pacific Index (NPI) as an anomaly of the sea level pressure in the wintertime of the North Pacific (30°N–65°N, 160°E–140°W) [*Minobe*, 2000] (Figures 2f–i). In both the wintertime and the summertime, the oscillations of TM, POC, and CC fluxes almost synchronized with that of NPI (the correlation coefficient (*r*) on wintertime average = 0.90 ± 0.02, *r* on summertime average = 0.67 ± 0.18), indicating that the oscillations of these particle fluxes were caused by the changes of sea surface and atmospheric conditions. When the sea surface pressure dropped, the reduction of nutrient supply from the ocean interior possibly occurred probably due to cold air breaks [*Cui and Senjyu*, 2010], which was consistent with the time series of sea surface nutrient in this region [*Whitney*, 2011]. Consequently, the primary productivity could decrease.

Considering these oscillations, we found the significant decreasing trends of CC flux in both the wintertime and the summertime (annual mean: −0.88 ± 0.13 mg m^{-2} day^{-1} $year^{-1}$, the decreasing rate (*D*) = −2.7 ± 0.4% $year^{-1}$) even though OP flux had no long-term trend (Figures 2d and 2e and Table 1), suggesting a possibility that coccolithophorids and/or foraminifera with $CaCO_3$ shells decreased while diatoms or radiolarian with opal shells were constant in the past two decades. In addition, POC flux in the wintertime had a decreasing trend of 0.25 mg m^{-2} day^{-1} $year^{-1}$ (annual mean: −0.06 ± 0.02 mg m^{-2} day^{-1} $year^{-1}$, *D* = −0.7 ± 0.2% $year^{-1}$), suggesting a long-term decline of primary productivity, at least in the wintertime (Figure 2b and Table 1). On the other hand, RE flux through all seasons had the increasing trend of 2.4 ± 0.6 mg m^{-2} day^{-1} $year^{-1}$ on average although there was no significant oscillation (Figure 2c). The increase of RE flux might be derived from the change of atmospheric dust deposition. Although the impact of volcanic ash increased in 2008 in the eastern North Pacific subpolar region [*Hamme et al.*, 2010], there was still no report in the western North Pacific.

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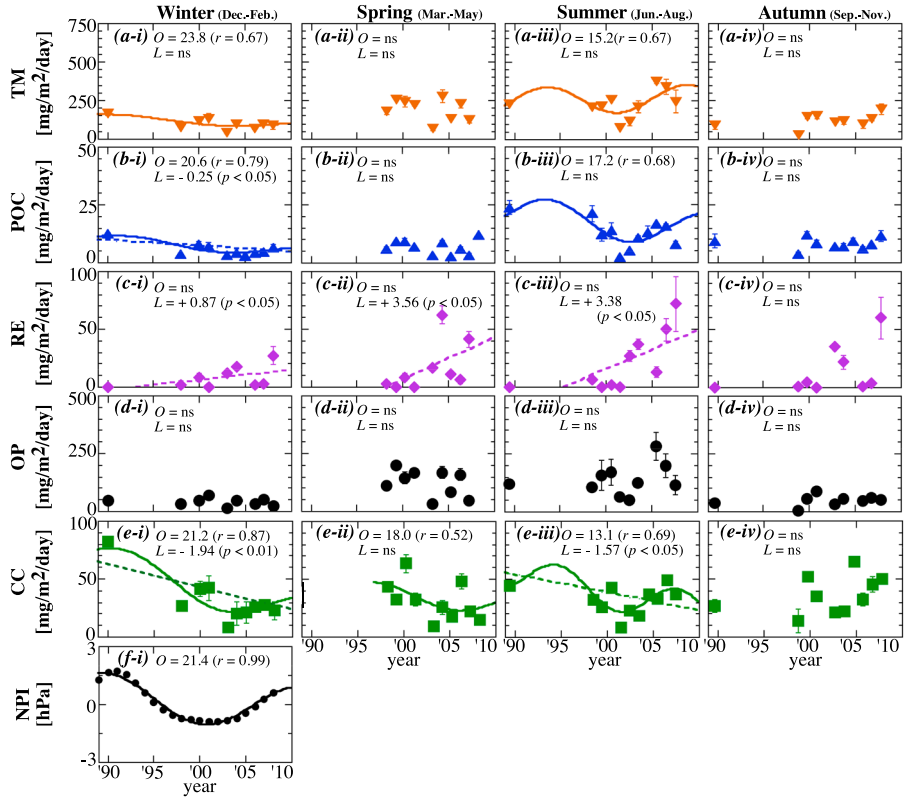

Figure 2. Seasonal composite time series of (a) TM flux, (b) POC flux, (c) RE flux, (d) OP flux, and (e) CC flux in the sinking particle along with (f) NPI. Unit for all sinking particle fluxes was mg m^{-2} day^{-1}. Solid and dashed lines indicate the significant fitted curves (*O*, year) and linear trends (*L*, mg m^{-2} day^{-1} $year^{-1}$) within the 95% confidence level with the correlation coefficient (*r*) for the fitted curve equation. "ns" means no significance. NPI (hPa) was shown as the 10–80 year band-pass filtered time series of NPI after 1900 based on http://climatedataguide.ucar.edu/sites/default/files/cas_data_files/asphilli/npindex_monthly_1.txt.

Since CC and POC fluxes significantly decreased over the past two decades while OP flux had no trends and RE flux increased, the mole ratio of OP/CC increased with annual mean of 0.11 ± 0.06 $year^{-1}$. This result was opposite to the long-term trend of mole ratio of OP/CC in the Atlantic Ocean (-0.01 to -0.04 $year^{-1}$ during the 1980s–1990s) [*Deuser et al.*, 1995; *Antia et al.*, 2001], suggesting a possibility of the recent acceleration in oceanic absorption of atmospheric carbon in the western North Pacific subpolar region.

3.2. Implication of Decreasing Trend of $CaCO_3$ Flux in the Sinking Particle

What led to the significant decreasing trend of CC flux despite no trend of OP flux in this region? To explain the decreasing trend of CC flux, we must consider the following two possibilities derived from the oceanic acidification [e.g., *Gehlen et al.*, 2007]: (i) the strengthening of $CaCO_3$ dissolution through seawater column and (ii) the decrease in productivity of $CaCO_3$-shelled species.

Using time series of satellite-derived CC, Chl, and *T* composite data in the sea surface around Station KNOT (42–46°N, 153–157°E) from 1997 to 2012 [*Goddard Earth Sciences Data and Information Services Center*, 2013], we found that the content of CC in the sea surface had *D* of $-0.7 \pm 0.5\%$ $year^{-1}$ (annual mean trend: -0.003 ± 0.002 μmol L^{-1} $year^{-1}$) significantly, which was almost equal to *D* of Chl with $-1.1 \pm 0.5\%$ $year^{-1}$ (annual mean trend: -0.006 ± 0.002 μg L^{-1} $year^{-1}$) along with the increasing trend of *T* (annual mean trend: -0.07 ± 0.04°C $year^{-1}$) (Figure 3 and Table 1). Thus, the decreasing productivity of $CaCO_3$-shelled species in the sea surface must cause most of the decrease in primary productivity in this region.

Provided that Chl is proportional to POC, *D* of POC at 1000 m depth corresponded with that in the surface. On the other hand, *D* of CC at 1000 m depth was about 4 times as large as that in the sea surface and the difference of *D* between the surface and 1000 m was -2.0% $year^{-1}$. What brought about the significant

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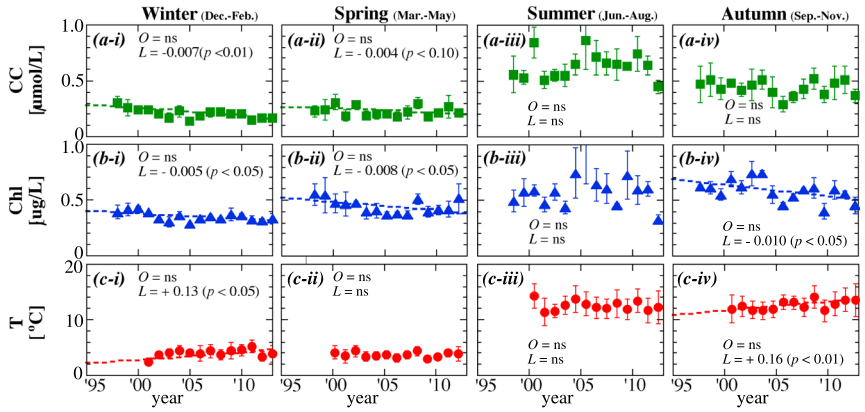

Figure 3. Seasonal composite time series of (a) CC ($\mu mol\,L^{-1}$), (b) Chl ($\mu g\,L^{-1}$) and (c) *T* (°C) in the surface near Station KNOT (42–46°N, 153–157°E) from 1997 to 2013 based on the time series of satellite observation [*Goddard Earth Sciences Data and Information Services Center*, 2013].

difference in *D* of CC between the two layers? Around Station KNOT, the strengthening of ocean stratification caused the decrease in dissolved oxygen of 0.7–1.8 $\mu mol\,kg^{-1}\,year^{-1}$ in the subsurface water [*Wakita et al.*, 2010; *Watanabe et al.*, 2008], leading to a potential that saturation state of $CaCO_3$ can be declined by 1.8% $year^{-1}$ due to the change in alkalinity. In the western North Pacific subpolar region, therefore, we concluded that (i) the strengthening of $CaCO_3$ dissolution through seawater column had contributed to 75% of the decreasing trend of CC flux, and (ii) the decrease in productivity of $CaCO_3$-shelled species contributed to 25% of the decreasing trend of CC flux.

Considering the contribution of decreasing productivity of $CaCO_3$-shelled species (25%) together with the long-term trend of OP/CC ($0.11 \pm 0.06\,year^{-1}$) in the biogenic particle flux, we found that the mole ratio of OP/CC had increased with annual mean of $0.03 \pm 0.02\,year^{-1}$ in the sea surface of the western North Pacific subpolar region. Consequently, the efficiency in oceanic sequestration of atmospheric CO_2 might be enhanced recently due to the increase of OP/CC ratio.

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