

Geophysical Research Letters[•]

RESEARCH LETTER

10.1029/2021GL093968

Key Points:

- Significant upper ocean (0–850 m) declining trends in nitrate and phosphate are identified in the Northwest Pacific Ocean
- Maximum declining trends of nutrients and increase in temperature are identified at mid-depths (300–500 m)
- The Northwest Pacific is shifting toward N-limitation in contrast to previously inferred P-limitation

Supporting Information:

Supporting Information may be found in the online version of this article.

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Citation:

Kim, J. H., Kim, G., Cho, H.-M., Nam, S. H., Hwang, J., Park, S., et al. (2022). Decline in the nutrient inventories of the upper subtropical Northwest Pacific Ocean. *Geophysical Research Letters*, 49, e2021GL093968. https://doi. org/10.1029/2021GL093968

Received 3 MAY 2021 Accepted 28 APR 2022

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Decline in the Nutrient Inventories of the Upper Subtropical Northwest Pacific Ocean

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Abstract The surface nutrient concentrations of the Northwest Pacific Ocean have shown distinct changes over the past four decades, which have been variably attributed to changes in anthropogenic nitrogen inputs or stratification. Here, we present a new data compilation that demonstrates a long-term decline in the surface nutrient concentrations caused by a significant decrease (~25%) in the nutrient inventories of the permanent thermocline (100–850 m) rather than by enhanced stratification. This subsurface nutrient decrease is likely due to the northward expansion of high-temperature, low-nutrient subtropical gyre waters associated with climate change and a regional physical regime shift. Furthermore, a declining trend in the N:P ratio pointing toward greater N-limitation is observed in this research, in contrast to current assumptions inferring P-limitation by anthropogenic nitrogen inputs. Our results imply that these shifts may influence new production as well as carbon sequestration in this region.

Plain Language Summary In the Northwest Pacific Ocean, contrasting descriptions of surface nutrient trends have been reported, with different mechanisms such as anthropogenic inputs and climate change. In this study, we show that the nutrients have declined over the past four decades in the surface layer of the Northwest Pacific Ocean as a result of a significant decrease in the nutrient inventories of the subsurface waters. This decline in nutrient concentrations is likely associated with regional climate change. The results show that the nutrient biogeochemistry of the region is shifting toward N reduction and limitation, which may influence new production and carbon sequestrations in this region.

1. Introduction

Over the past few decades, declining surface nutrient concentrations have been directly observed and indirectly inferred through chlorophyll observations in many regions of the oceans, especially in the subtropical oceans (Behrenfeld et al., 2006; Boyce et al., 2010; Polovina et al., 2008; Yasunaka et al., 2016). Many studies have suggested a physical obstruction of the upward flux of nutrients associated with stratification and mixed layer shoaling as the cause of the surface nutrient decline (Behrenfeld et al., 2006; Boyce et al., 2010; Doney, 2006; Yasunaka et al., 2016). However, recent observational studies have shown that the mixed layer has been deepened at a rate of several meters per decade (Sallée et al., 2021; Somavilla et al., 2017), while the stratification estimated by the density contrast across the mixed layer base has been increased during the past 60 years at varying global mean rates of 1%–9% per decade (Li et al., 2020; Sallée et al., 2021; Yamaguchi & Suga, 2019). Moreover, the lack of correlation between the local stratification and the surface nutrient changes has been reported previously (Dave & Lozier, 2010, 2013; Lozier et al., 2011). Thus, the exact mechanisms causing the recent changes in surface nutrient concentrations are still unclear.

In the Northwest Pacific Ocean, some long-term observational studies report decreases in surface nutrient concentrations from the 1960s to the 2010s and attribute these changes to surface stratification (Aoyama et al., 2008; Watanabe et al., 2005; Yasunaka et al., 2016). However, other studies report increases in the surface nutrient concentrations, especially nitrogen, in the Northwest Pacific from the 1980s, as a result of anthropogenic pollution from the Asian continent (Kim et al., 2011, 2014). The Kuroshio Current, which contains one of the lowest concentrations of nutrients in the world's major currents with less than 1 μ M nitrate, flows through the region as the western boundary current of the North Pacific subtropical gyre (Figure 1a). The Kuroshio Current brings these low-nutrient waters northward and eastward into the North Pacific (Miller et al., 2004), as well as the



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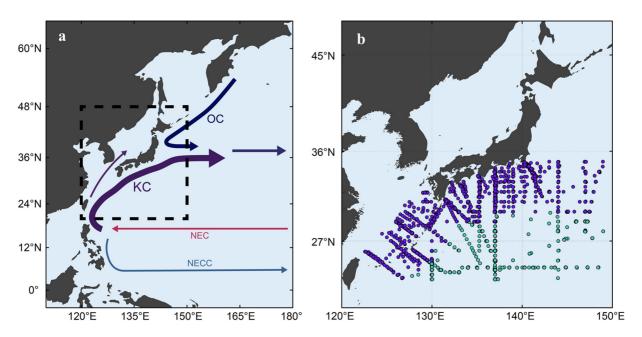


Figure 1. Currents and data collection sites in the Northwest Pacific Ocean. (a) A map of the Northwest Pacific Ocean including the major currents and the study area (black dashed box). The Kuroshio Current (KC) flows through the study region, originating from the North Equatorial Current (NEC) that bifurcates into the KC and the North Equatorial Counter Current (NECC). The Oyashio Current (OC) flows above the KC. (b) A map of data collection sites (n = 9,992) in the study regions: the Kuroshio mainstream (purple points) and the inner gyre (green points). The two study regions were divided based on the surface geostrophic current (stronger in the Kuroshio mainstream) and decadal trends of temperature (more rapid increase in the Kuroshio mainstream).

marginal seas of the region (Y. K. Cho & Kim, 2000; Ichikawa & Beardsley, 2002), including the East China Sea, the Yellow Sea, the South China Sea, and the East/Japan Sea. Changes in nutrient conditions of the Northwest Pacific Ocean and the Kuroshio Current can be expected to have implications for the entire Pacific Ocean system. Thus, there is a need to clarify mechanisms resulting in the surface nutrient trends.

In previous studies, the inventory of nutrients available in the subsurface layer has been largely ignored despite its importance as the primary source of nutrients to the surface. Accordingly, this study investigates nutrient trends in the upper 850 m of the Northwest Pacific Ocean to present the explanation for the changes in the nutrients in both the surface and subsurface layers of the Kuroshio system.

2. Materials and Methods

We compiled and analyzed observational data of the Kuroshio system in the Northwest Pacific Ocean from 1980 to 2017 (Figure 1) to study long-term changes in the observed nutrient concentrations of the system. The nutrient concentrations and temperature data archived by the Japan Meteorological Agency (JMA: http://www. data.jma.go.jp/gmd/kaiyou/db/vessel_obs/data-report/html/ship/ship_e.php) were utilized in this study. Quality control for dissolved inorganic nitrogen (N), dissolved inorganic phosphorus (P), and temperature data in the Northwest Pacific was based on the methods of T. W. Kim et al. (2011) and Kodama et al. (2016) (Text S1 in Supporting Information S1). The study region was partitioned into the Kuroshio mainstream and the inner subtropical gyre (Figure 1b) based on the surface geostrophic current averaged from satellite altimetry-derived sea surface height data over the period from 1993 to 2017 (https://resources.marine.copernicus.eu/product-detail/SEALEVEL_GLO_PHY_L4_MY_008_047/INFORMATION) and the decadal trends of water temperature data (Text S1 and Figure S4a in Supporting Information S1).

For spatial analysis, the quality-controlled data were binned into 10 depth layers (Table S1 in Supporting Information S1). At each depth layer, monthly averages of each parameter were calculated in 1° by 1° grids followed by 3° by 3° grids. For temporal analysis, the 3° by 3° grid data were further averaged annually from 1980 to 2017. The annual means of each parameter for the entire study region were used to obtain the 5-year averages starting from 1980 to 2017. Then, a linear regression was applied to the 5-year means in each study region to determine the 37-year trends of phosphate, nitrate, and temperature at each depth layer (Figure 2).



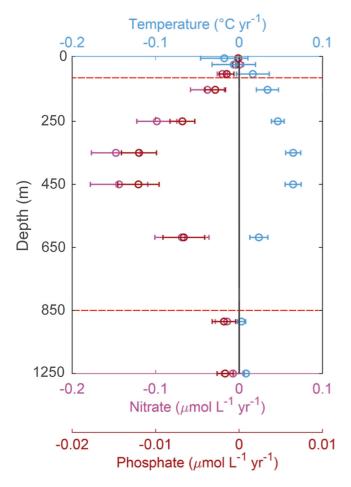


Figure 2. Vertical profiles of 37-year trends of temperature (blue), nitrate (pink), and phosphate (maroon) at different depth layers in the upper 1,500 m of the Kuroshio mainstream waters. The boundaries of the surface, subsurface, and deep ocean layers are marked by red dashed lines. The greatest change in nutrients and temperature is observed in the subsurface layer (300–500 m).

We also examined the linear temporal trends of the nutrient concentrations, temperature, N:P ratio, and excess N concentration (N*) over the entire 37-year period for both the surface (0–100 m) and subsurface (100–850 m) layers, down to the bottom of the permanent thermocline (Figures 3 and 4). The N* was calculated with the equation, N* = N- $R_{N:P} \times P$, where $R_{N:P}$ is the Redfield ratio (Gruber & Sarmiento, 1997; T. W. Kim et al., 2011). For the linear trend analysis, the quality-controlled observations were averaged to seasonal bins at monthly resolution (Figure 3). The seasonal bins include spring (March–May), summer (June–August), autumn (September–November), and winter (December–February). Finally, we estimated the changes in the inventories in two different vertical layers (the surface layer (0–100 m) and the subsurface layer (100–850 m)) by integrating the nutrient concentrations in the corresponding layers for the 5-year intervals between the periods 1980–1984 and 2015–2017 (Figure S1 in Supporting Information S1).

3. Results and Discussion

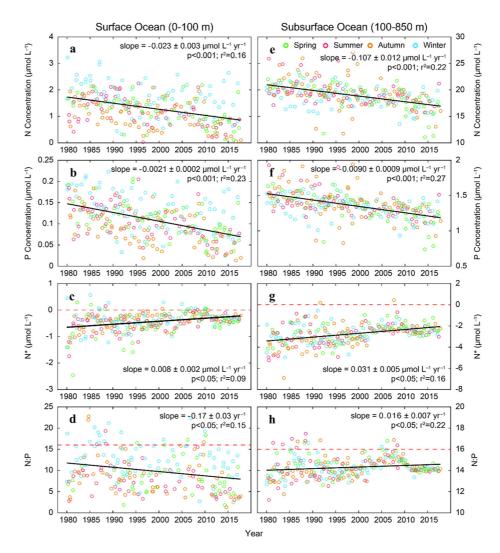
3.1. Nutrient Trends in the Kuroshio Mainstream

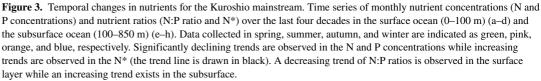
The averaged vertical profiles of long-term trends in nitrate, phosphate, and temperature of the Kuroshio mainstream are shown in Figure 2. A significant decreasing trend in nutrients is observed throughout almost the entire water column from 0 to 1,500 m, with a maximum rate of decrease at 300–500 m. This decreasing trend in nutrient concentrations is closely mirrored by the increases in temperature, except at the sea surface where the temperature shows a slight decreasing (although not statistically significant) trend. The maximum rate of warming occurs at 300–500 m, which is coincident with the most rapid decline in the nutrient concentrations. These trends result in a structural change in the vertical profiles of the nutrients and temperature (Figures S1 and S3a in Supporting Information S1). The averaged vertical profiles of the nutrient concentrations show a persistent increase in concavity (initially concave downward) from the 1980s to the 2010s, which translates to a significant decrease in the nutrient inventory of the subsurface ocean (particularly at 300–500 m; Figure S1 in Supporting Information S1).

In the surface ocean (0–100 m), a significant decline in the nutrient concentrations (p < 0.001) was identified through a linear regression (Figures 3a

and 3b) at rates of $-0.023 \pm 0.003 \mu \text{mol N L}^{-1} \text{ yr}^{-1}$ and $-0.0021 \pm 0.0002 \mu \text{mol P L}^{-1} \text{ yr}^{-1}$. As a result of these trends, the nutrient concentrations in the surface layer have been nearly halved over the four decades: the average concentrations of N and P in 1980 were ~1.7 and ~0.17 μ M, respectively, while those in 2017 were ~1.1 and ~0.09 μ M. The 37-year decrease in the surface N and P inventories in the Kuroshio mainstream was approximately 112 \pm 11 g N m⁻² and 10.9 \pm 0.8 g P m⁻² (approximately 71% and 66% reduction), respectively, estimated by integrating the change in the 5-yearly depth profiles in the layer 0–100 m (Figure S1 in Supporting Information S1). The nutrient concentration declined in all seasons at significant (p < 0.001) rates. In detail, we observed the declines of $-0.024 \pm 0.006 \,\mu\text{mol N L}^{-1} \,\text{yr}^{-1}$ and $-0.0021 \pm 0.0004 \,\mu\text{mol P L}^{-1} \,\text{yr}^{-1}$ in summer, and $-0.021 \pm 0.005 \,\mu\text{mol N L}^{-1} \,\text{yr}^{-1}$ and $-0.0018 \pm 0.0004 \,\mu\text{mol P L}^{-1} \,\text{yr}^{-1}$ in winter. These seasonally consistent trends imply that the decline in surface nutrient concentrations is continuing throughout the year regardless of the seasonal cycle.

In the subsurface layer (100–850 m) of the Kuroshio Current, a decrease in the nutrient concentrations over the 37 years was also evident (Figures 3e and 3f). The subsurface nutrient concentration declined slightly (p < 0.001) with slopes of $-0.107 \pm 0.012 \mu mol N L^{-1} yr^{-1}$ and $-0.0090 \pm 0.0009 \mu mol P L^{-1} yr^{-1}$. However, the estimated 37-year decreases in the subsurface nutrient inventories based on the averaged vertical profiles were $4,650 \pm 400 \text{ g N m}^{-2}$ and $381 \pm 30 \text{ g P m}^{-2}$ (approximately 23% and 26% reduction, respectively). Despite the smaller reduction in terms of percentage, the absolute decrease of the inventory in the subsurface is more than 30-fold of the surface nutrient inventory decrease. In the layer deeper than 850 m, we confirmed that there was





no significant change in temperature and nutrient concentrations from 1980 to 2012 for all seasons based on the linear regression (Figure S2 in Supporting Information S1).

The reduced nutrient reservoir of the subsurface may have a significant influence on new production, especially in the subtropical waters of the Northwest Pacific, as the nutrients in the euphotic zone are mainly replenished from the subsurface reservoir by convective mixing (Sukigara et al., 2011).

3.2. Potential Mechanism of Nutrient Decline

Like numerous previous studies (Behrenfeld et al., 2006; Kodama et al., 2016; Watanabe et al., 2005), and in contrast to the findings of T. W. Kim et al. (2011), our results show a significant decline in the surface ocean nutrient concentrations in the Northwest Pacific Ocean over the past 40 years. The surface nutrient declines in the Northwest Pacific have generally been tied to stratification as a result of surface warming. The surface stratification mechanism, however, is not supported by a statistically significant correlation between interannual variability in stratification within the upper ocean and ocean chlorophyll (highly correlated to nutrient concentrations;



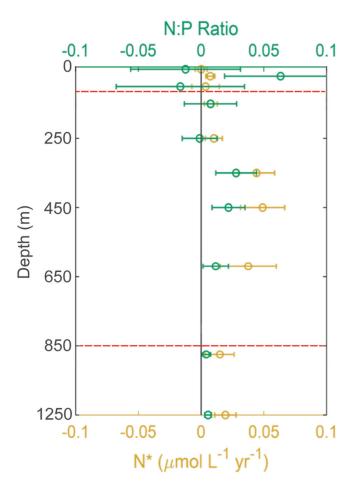


Figure 4. Vertical profiles of 37-year trends of N:P ratios (green) and N* (yellow) at different depth layers in the upper 1,500 m of the Kuroshio mainstream waters. The boundaries of the surface, subsurface, and deep ocean layers are marked by red dotted lines. There were higher increase rates of both N* and N:P ratios in the subsurface (~400 m).

Dave & Lozier, 2010, 2013; Lozier et al., 2011). Instead, according to our results, the observed warming trends with the maximum warming rates at 300–500 m would result in a decreased stratification and enhanced vertical mixing near the surface layer unless temperature changes are salinity compensated (Figure 2). Furthermore, there is no evidence of statistically significant trends in the upper ocean stratification or mixed layer depth shoaling in the North Pacific (Dave & Lozier, 2010; Somavilla et al., 2017). Therefore, our results suggest an alternative mechanism that a decline in the subsurface nutrient reservoir is large enough to cause a decrease in the surface nutrient concentrations regardless of an increase or decrease in vertical water mixing near the surface (further discussed below).

To investigate the cause of the decline in the subsurface nutrient concentrations, the spatial extent of the trends and the alterations in the stability of the water column were examined (Figure S3–S5 in Supporting Information S1). The increase in temperature is especially pronounced in the Kuroshio mainstream region (Figure 2 and S3a in Supporting Information S1), whereas the vertical profiles of temperature in the inner gyre region and the Hawaii Ocean Time-series site display no significant change over the study period (Figure S3b and S3c in Supporting Information S1). These results, coupled with the statistically significant correlation between each of the nutrient concentrations and temperature (Figure S4 in Supporting Information S1), suggest that northward expansion of the subtropic gyre could have caused the decrease in the nutrient concentrations as the high-temperature, low-nutrient waters are advected to a greater degree into the Northwest Pacific by the Kuroshio Current. This northward expansion of the North Pacific subtropical gyre is consistent with previous findings (Polovina et al., 2008; Saenko et al., 2005; Zhang et al., 2014).

The vertical water stability of the Kuroshio mainstream was examined by calculating the vertical temperature gradient for every 100 m depth layer as an index for thermal stratification (Figure S5 in Supporting Information S1). The pronounced winter-spring difference in water temperature is limited to the upper 200 m in the recent decade (2000–2010, lines with squares in Figure S5a in Supporting Information S1) whereas it expanded down to 600 m in the 1980s (1980–1990, lines with circles in Figure S5a in Supporting Information S1). This implies a change in winter-spring transition in the recent

decade, which is consistent with strengthened spring-winter entrainment (particularly at depths between 200 and 600 m) and the reemergence process suggested by recent studies (S. Y. Kim et al., 2020; Kwon et al., 2010; Pak et al., 2014, 2019). The structural change is consistent with the physical regime shift in the Kuroshio region, which occurred between 1985 and 1990 (S. Y. Kim et al., 2020; Kwon et al., 2010; Pak et al., 2014, 2019). Due to this regime shift, thermal conditions in the upper ~ 300 m of the Northwest Pacific Ocean have shifted from being primarily controlled by the East Asian winter monsoon (atmospheric forcing) to being controlled by the spring-initiated reemergence process (ocean dynamics; S. Y. Kim et al., 2020; Kwon et al., 2010; Pak et al., 2014, 2019). Among the layers below 200 m, the temporal mean and the decadal increase of the vertical temperature gradient reach the highest values at 400-500 m from 1980 to 2017 (Figure S5b in Supporting Information S1). The observation coincides with the depth of the maximum nutrient concentration decrease and the temperature increase (around 400 m; Figures S1 and S3 in Supporting Information S1), although the trend is insignificant or less clear for the other depth layers. The regime-shift-associated change in the water column structure might have resulted in the enhanced vertical mixing in the upper ~ 300 m and could explain the increasing (decreasing) trend of temperature (nutrients) peaking at around 300-500 m. The increased stability of the water column or enhanced thermal stratification implying weakened vertical mixing in the layers below 500 m accounts for the slower (lower) warming (nutrient declining) rates than in the waters above (Figure 2).

3.3. Effects of Nutrient Decline on the Upper Ocean Biogeochemistry

The decline in the nutrient concentrations of the upper ocean is also likely to impact the fraction of anthropogenic nutrients in the ocean. As natural concentrations of nutrients decrease, anthropogenic N input from the atmosphere (Kim et al., 2011, 2014) and the continental shelves (H. M. Cho et al., 2019) will comprise an increasing fraction of the nutrient inventory in the upper ocean. The N* can be used as an index to estimate the increase in the anthropogenic fraction of N (Kim et al., 2011, 2014). A significant increase in the N* is observed (p < 0.05) in both the surface ($0.008 \pm 0.002 \mu \text{mol L}^{-1} \text{ yr}^{-1}$; $r^2 = 0.09$) and the subsurface layer ($0.031 \pm 0.005 \mu \text{mol L}^{-1} \text{ yr}^{-1}$; $r^2 = 0.16$) where the N* is approaching 0 (Figures 3c and 3g). The increase of N* in the upper Northwest Pacific Ocean can be explained by an increased anthropogenic nitrogen input from the East Asian continent (I. N. Kim et al., 2014) and/or nitrogen fixation (Gruber & Sarmiento, 1997). I. N. Kim et al. (2014) suggested that N₂ fixation does not add substantially to the long-term increase of the N* in this region based on the higher increase rate of N* in the surface layer relative to the subsurface. However, we cannot exclude N₂ fixation as a source of N* in this study because the highest rate of N* increase was observed in the subsurface layer (Figure 4).

Theoretically, the increased N* should cause a shift toward P-limitation by increasing N:P ratios as anticipated by prior reports (Kim et al., 2011, 2014). In contrast to the prediction, the measured N:P ratios demonstrated a decreasing trend in the surface layer and hence, a shift toward N-limitation (Figure 3d). Under the N-limited condition, the N:P ratio in the surface water is dependent on the proportion of the nutrient drawdown by sinking organic matter at a near Redfield ratio (Anderson & Sarmiento, 1994), as well as the N* increase, against the seawater nutrient inventory. The analogous trends of decrease in the nutrient concentrations and the N:P ratios in the surface layer (Figure 3) suggest that the effect of nutrient drawdown is larger than the N* increase under a nutrient decline condition.

Conversely, the subsurface N:P ratios increased while the N and P concentrations decreased (Figures 3e, 3f, and 3h). The increased N:P ratios could be due to an increased effect of remineralization of sinking organic matter with a near Redfield ratio under the nutrient decline condition as shown in the East Sea (Japan Sea) of the Northwest Pacific Ocean, the upstream of the Kuroshio Current (T. H. Kim & Kim, 2013). In the East Sea, the N:P ratios peaked in the subsurface, approaching 16, while the N:P ratios of the surface and deep ocean were lower than 5 and 13, respectively (T. H. Kim & Kim, 2013). According to T. H. Kim & Kim (2013), the significant correlation ($r^2 = 0.66$) between the AOU (apparent oxygen utilization) and the N:P ratios indicates the Redfield-like remineralization of sinking organic matter in the subsurface. The subsurface layer of our study region remains N-limited despite the increase of N:P ratios toward 16 because the original N:P ratios in the subsurface and the nutrients sunk from the surface have remained lower than 16 (Figure 3).

4. Conclusions

The Northwest Pacific Ocean has entered a mode of nutrient decline in the Kuroshio Current system. The reduced subsurface nutrient reservoir is likely causing the decline of the surface nutrient concentrations. We highlight the possibility that surface nutrient decline can occur independently of the stratification, as the nutrient concentration in the subsurface water becomes insufficient to replenish the surface nutrient stock. The nutrient concentration decrease in the subsurface can be attributed to the northward expansion of high-temperature, low-nutrient oligotrophic gyre waters into the region, combined with the enhanced stratification of the permanent thermocline associated with a regional climate regime shift starting in ~1990. This reduction in the nutrients of the Northwest Pacific Ocean may have a considerable influence on new production and carbon export. Furthermore, in contrast to the recent predictions that the region is shifting toward P-limitation (Kim et al., 2011, 2014), the Northwest Pacific is becoming more N-limited as the nutrients supplied from the subsurface waters diminish. These conditions may lead to further ecological shifts toward severely N-limited conditions.

Data Availability Statement

The nutrient concentration and temperature data were obtained from the JMA (http://www.data.jma.go.jp/gmd/kaiyou/db/vessel_obs/data-report/html/ship/ship_e.php). The mean climatological sea surface height data of the Northwest Pacific Ocean were produced by Ssalto/Ducas and distributed by Aviso+ (https://www.aviso.altime-try.fr/en/data/products/sea-surface-height-products/global/gridded-sea-level-anomalies-mean-and-climatology).

Acknowledgments

This research was supported by the

project titled "Deep Water Circulation

and Material Cycling in the East Sea

(20160400)", which is funded by the

and the National Research Foundation

(NRF-2018R1A2B3001147).

Ministry of Oceans and Fisheries, Korea,

(NRF) funded by the Korean government

The temperature data at Station ALOHA were obtained from the Hawaii Ocean Time-series observations (https:// hahana.soest.hawaii.edu/FTP/hot/ctd/). The altimeter satellite gridded sea surface heights data were obtained from the Copernicus Marine Service (https://doi.org/10.48670/moi-00148).

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