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Intensive mixing along an island chain controls oceanic biogeochemical cycles

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[1] The subarctic Pacific is a high-nutrient low-chlorophyll (HNLC) region in which phytoplankton growth is broadly limited by iron (Fe) availability. However, even with Fe limitation, the western subarctic Pacific (WSP) has significant phytoplankton growth and greater seasonal variability in lower trophic levels than the eastern subarctic Pacific. Therefore, differences in Fe supply must explain the west-to-east decrease in seasonal phytoplankton growth. The Fe flux to the euphotic zone in the WSP occurs at a “moderate” value, in that it is significantly higher than its value on the eastern side, yet it is not sufficient enough to cause widespread macronutrient depletion, that is, HNLC status is maintained. Although we recognize several Fe supply processes in the WSP, the mechanisms that account for this moderate value of Fe supply have not previously been explained. Here we demonstrate the pivotal role of tidal mixing in the Kuril Islands chain (KIC) for determining the moderate value. A basin-scale meridional Fe section shows that Fe derived from sediments in the Sea of Okhotsk is discharged through the KIC into the intermediate water masses (~800 m) of the western North Pacific. The redistribution of this Fe-rich intermediate water by intensive mixing as it crosses the KIC is the predominant process determining the ratio of micronutrient (Fe) to macronutrients (e.g., nitrate) in subsurface waters. This ratio can quantitatively explain the differences in surface macronutrient consumption between the western and eastern subarctic, as well as the general formation and biogeochemistry of HNLC waters of the subarctic North Pacific.

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1. Introduction

[2] Along with sunlight, the nutrient content of surface water determines the global distribution of organic production in the ocean, thus controls cycles of biogenic elements such as carbon and nitrogen. The processes that form nutrient-rich surface water in the high-nutrient low-chlorophyll (HNLC)

region are particularly important for understanding global ocean biogeochemical cycles.

[3] The subarctic Pacific is a HNLC region where phytoplankton growth is broadly limited by iron (Fe) availability [Tsuda *et al.*, 2003; Boyd *et al.*, 2004]. Two gyres, the western subarctic gyre and the Alaskan gyre, dominate this region. Previous studies have compared the hydrographic properties of these two gyres. Harrison *et al.* [1999, 2004] found that despite their general similarities such as Fe deficiency, there are crucial differences between the two gyres, especially in lower trophic levels. They showed that there is stronger seasonality with greater phytoplankton growth and biomass in the western subarctic gyre than in the Alaskan gyre. For example, although chlorophyll concentrations are low in both gyres, the chlorophyll concentration in the western subarctic gyre (0.5–1.5 mg/m³ at station KNOT; 44°N, 155°E) is approximately twice as great as in the Alaskan gyre (0.3–0.5 mg/m³ at station P (OSP); 50°N, 145°W). In addition, the western subarctic Pacific (WSP) has the most effective biological pCO₂ drawdown and biological pump in the world ocean [Takahashi *et al.*, 2002; Buesseler *et al.*, 2007]. Phytoplankton growth is broadly limited by Fe, yet significant phytoplankton growth occurs simultaneously across the WSP. This seems to be contradictory, and Fe

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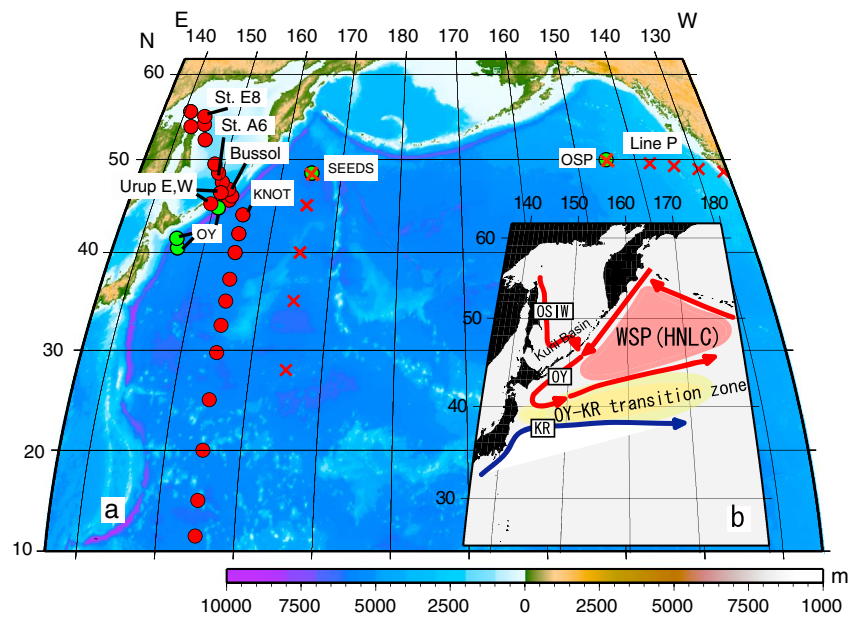


Figure 1. (a) Map showing sampling sites in the North Pacific and marginal seas examined in this study. Red circles indicate vertical section sampling observation sites for measurement of dissolved Fe, nitrate + nitrite, and hydrographic data in this study. Green circles indicate sites with previously reported data for dissolved Fe and nutrients in the Oyashio region (OY) [Nishioka *et al.*, 2007, 2011], oceanic site in the western subarctic Pacific, stations KNOT (44°N, 155°E), SEEDS (48°30'N, 165°E) [Nishioka *et al.*, 2007, 2011], and oceanic site in the eastern subarctic Pacific, Ocean Station Papa (OSP, 50°N, 145°W) [Nishioka *et al.*, 2001; Martin *et al.*, 1989]. Red crosses indicate sampling sites of previously published vertical sections along 165°E in the western Pacific [Nishioka *et al.*, 2007] and along line P in the eastern subarctic Pacific [Nishioka *et al.*, 2001]. (b) Schematic drawing of the water structure in the western North Pacific and the Sea of Okhotsk. OY: Oyashio region; WSP: western subarctic Pacific; KR: Kuroshio; OSIW: Okhotsk Sea Intermediate Water.

supply processes in the North Pacific must disentangle this apparent contradiction to explain the formation of HNLC water. A key element in explanations of HNLC water formation and biogeochemical cycles in this region is the concept of a “moderate” amount of Fe supply. Differences in Fe supply across the North Pacific can explain the eastward decrease in ocean productivity in the subarctic Pacific. Therefore, the Fe flux in the WSP should have a moderate value that is higher than that of the eastern side, but not so high as to cause widespread macronutrient depletion in surface waters (i.e., HNLC conditions are maintained).

[4] Mineral dust containing Fe from Asian deserts is known to be an important source of Fe in the North Pacific [e.g., Duce and Tindale, 1991]. Recently, attention has been drawn to the offshore transport of Fe-rich coastal water [Harrison *et al.*, 2004]. Subsequent studies [Lam *et al.*, 2006; Nishioka *et al.*, 2007; Lam and Bishop, 2008] have reported that resuspended sedimentary Fe from the continental shelf of marginal seas and the continental margin are important sources of Fe for the subarctic Pacific. Furthermore, eddy transport from the continental margin is considered to be an important process that supplies Fe to the open ocean [e.g., Johnson *et al.*, 2005; Lippiatt *et al.*, 2011]. However, the mechanisms controlling a moderate Fe supply value have not been explained in terms of these processes. Therefore, we do not yet quantitatively understand the simultaneous occurrence of significant phytoplankton growth and HNLC water formation, especially in the WSP.

[5] Large-scale transect observations covering the entire water column provide a comprehensive picture of water properties controlling the transport and biogeochemical cycling of elements in seawater [Sohrin and Bruland, 2011]. To understand the various phenomena affecting Fe distributions, sources, and cycles in the ocean basin, vertical section observations of dissolved Fe were planned as part of the International Polar Year GEOTRACES program (<http://www.geotraces.org/>). In this paper, we report basin-scale section profiles of dissolved Fe in the Sea of Okhotsk, including the Kuril Straits, and along the 155°E meridian in the western North Pacific Ocean from the subarctic to near the equator (11°30'N) (Figure 1a). The resulting composite profile reveals important processes that control the Fe distribution in this region and enables us to discuss the role of these processes in establishing the HNLC region of the North Pacific (Figure 1b).

2. Materials and Methods

[6] Observations in the Sea of Okhotsk were carried out during R/V *Professor Khromov* cruise Kh06 from August to September 2006 (stations indicated in Figure 1a) by performing a transect across the basin. A part of this cruise was coordinated by IPY-GEOTRACES. A meridional section in the North Pacific along 155°E was performed during R/V *Hakuho-Maru* cruise KH08-2 from August to September 2008 (stations indicated in Figure 1a). To characterize vertical

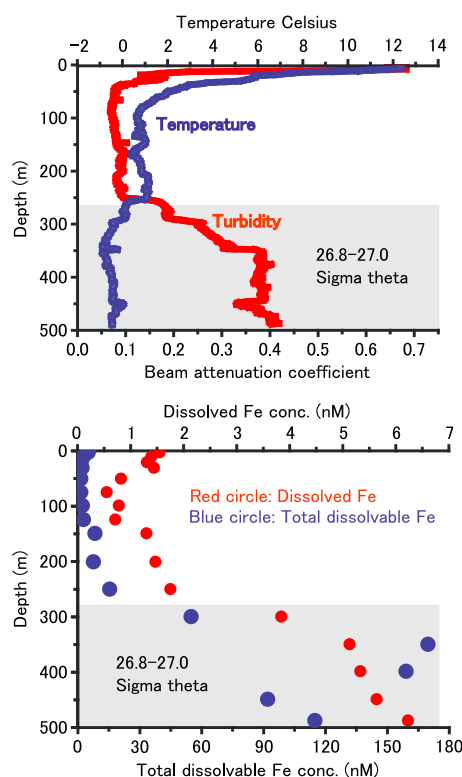


Figure 2. Vertical profiles of turbidity, temperature, total dissolvable Fe, and dissolved Fe in the northwestern continental shelf in the Sea of Okhotsk (station E8 in Figure 1a). Gray shading indicates DSW density of 26.8–27.0 σ_θ .

profiles of the Fe concentration, seawater samples and hydrographic data (including transmittance) were collected using a clean conductivity-temperature-depth-carousel multiple sampler system (SBE-911plus and SBE-32 water sampler, Sea Bird Electronics, Inc., C-star transmittance sensor, Wet Labs, Inc.), which housed 12 acid-cleaned Teflon-coated 10 or 12 L Niskin-X bottles, connected to armored cable. Subsampling was done with 0.22 μm Durapore filters (Millipak 100, Millipore Corp.) connected to the Niskin-X spigot, and the filtrate was collected in acid-cleaned 125 mL low-density polyethylene bottles (Nalgene Co., Ltd) under gravity pressure. The filtrate and unfiltered samples were acidified with distilled HCl in the onboard clean room. The filtrate samples from cruise KH08-2 were maintained at pH $\sim$ 2 for approximately 24 h at room temperature, and then the pH was adjusted to 3.2 immediately before analysis on board. The filtrate samples from cruise Kh06 were maintained at pH $\sim$ 2 for more than 1 month at room temperature, and then pH was adjusted to 3.2 immediately before analysis onshore. The unfiltered samples from both cruises were maintained at pH $\sim$ 2 for more than 6 months at room temperature, and then, the pH was adjusted to 3.2 immediately before analysis onshore. After this, the dissolved Fe (leachable Fe in 0.22 μm filtrate) and the total dissolvable Fe (dissolved plus leachable particulate Fe in unfiltered sample at pH $\sim$ 2) were analyzed using a flow injection analysis chemiluminescence detection system [Obata *et al.*, 1993]. All sample treatments were performed in a laminar flow bench in a clean-air laboratory.

[7] Our dissolved Fe measurement method and reference seawater were quality controlled using reference standard

seawaters from the SAFe (Sampling and Analysis of Iron) cruise [Johnson *et al.*, 2007] (distributed by the University of California Santa Cruz for an intercomparison study). SAFe reference standard seawater S containing 0.090 ± 0.007 nM Fe (avg $\pm$ 1 SD) and seawater D2 containing 0.90 ± 0.02 nM Fe (www.geotraces.org; November 2011) were measured at 0.10 ± 0.010 nM Fe ($n=3$) and 0.99 ± 0.023 nM Fe ($n=3$), respectively, by our method.

[8] Nitrate + nitrite concentrations were also measured in water samples collected from the same stations, using a Bran-Luebbe autoanalyzer (TRACCS 800).

3. Results and Discussion

3.1. Direct Observation of the Fe Source Water in the Sea of Okhotsk

[9] In the Sea of Okhotsk, sea ice forming on the northwestern continental shelf in winter produces a large volume of cold brine water which contributes to the formation of dense shelf water (DSW: 26.8–27.0 σ_θ) [Kitani, 1973; Martin *et al.*, 1998; Gladyshev *et al.*, 2003]. Nakatsuka *et al.* [2002] found that the DSW on the northwestern continental shelf and intermediate water was characterized by very high turbidity and very low temperatures and noted that the DSW contained resuspended sediment. We conducted direct observations of the Fe distribution in the sea ice formation area in September 2006. The vertical profiles of dissolved Fe and total dissolvable Fe concentrations in the northwestern continental shelf area (station E8 in Figures 1a and 2) show an extremely high anomaly (dissolved Fe > 5 nM and total dissolvable Fe > 150 nM) in the vicinity of the bottom sediments. This Fe-rich water is clearly consistent with water that has high turbidity and very low temperature (Figure 2), indicating that large amounts of dissolved and particulate Fe are introduced to the DSW by resuspension of sediment. The DSW tends to penetrate the upper part (250–450 m depth) of the Okhotsk Sea Intermediate Water (OSIW). The OSIW flows southward along the east coast of Sakhalin Island [Itoh *et al.*, 2003; Fukamachi *et al.*, 2004; Yamamoto-Kawai *et al.*, 2004] and discharges into the WSP through the Kuril Straits, mainly through Bussol' Strait, the deepest channel of the Kuril Islands. The vertical section of dissolved Fe (Figure 3b) delineates the transport of a substantial amount of Fe in the intermediate water from the northwestern continental shelf to the Sea of Okhotsk, and through Bussol' Strait to the WSP, as indicated by Nishioka *et al.* [2007].

[10] The section data also suggest the presence of another Fe source around the Kuril Straits. Judging from the Fe data at station E8, there is insufficient dissolved Fe originating from the northwestern continental shelf to account for the very high integrated dissolved Fe concentrations in the water column at Bussol' Strait. Previous studies have reported that particles of volcanic origin (this might be derived from the Kuril/Kamchatka margins) are Fe sources to the WSP [Weber *et al.*, 1996; Ootosaka *et al.*, 2004; Lam and Bishop, 2008; Lam *et al.*, 2012]. Although the solubility of Fe from volcanic particles is not fully understood, it is likely that supplementary Fe input by volcanogenic minerals from the Kuril/Kamchatka volcanic margin influences our section profile around the Kuril Straits.

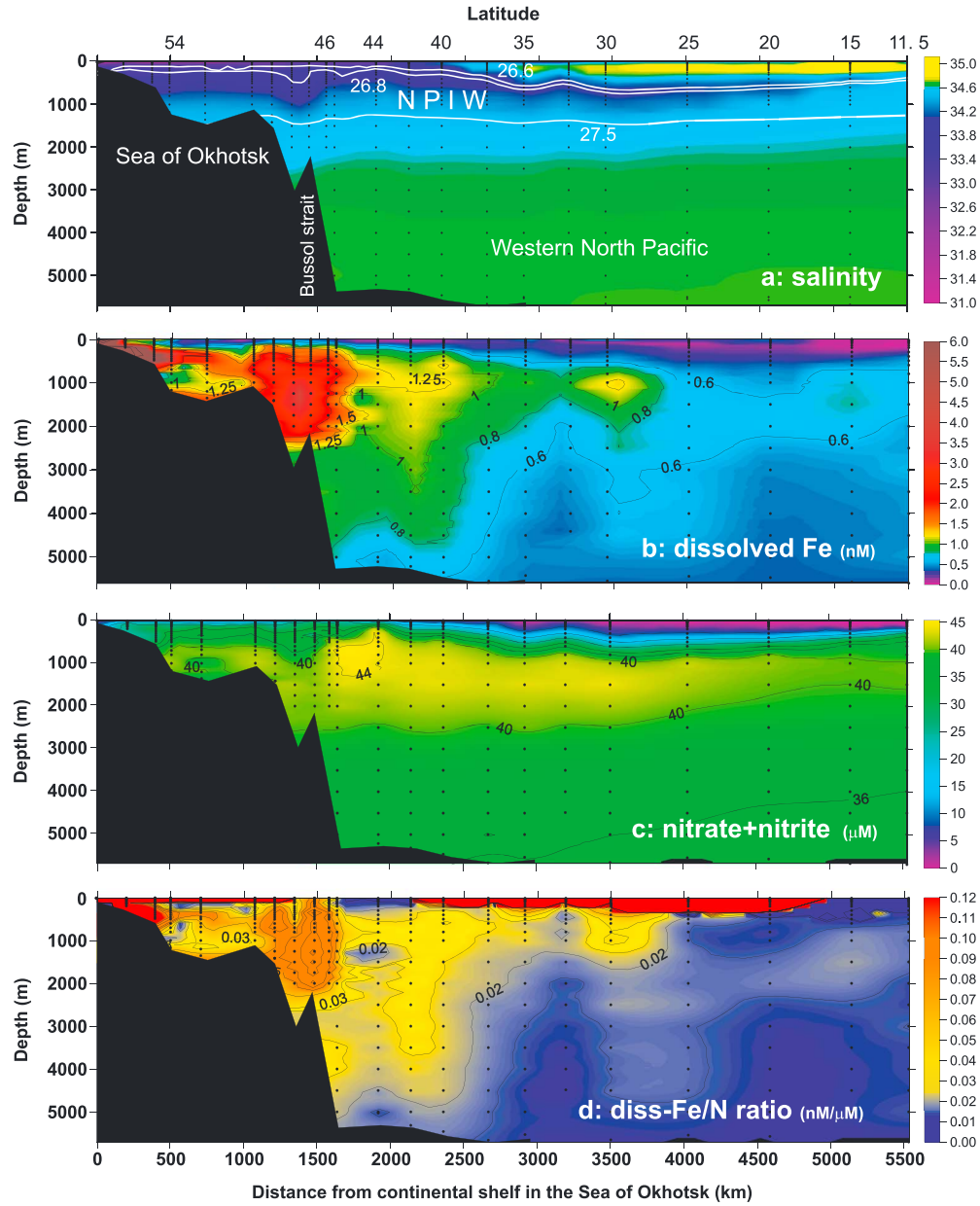


Figure 3. Basin-scale vertical sections profiles starting in the Sea of Okhotsk, passing through Bussol' Strait in the Kuril Islands, then heading south along 155°E from the subarctic, through the subtropics to near 11°30'N. (a) Salinity, (b) dissolved Fe, (c) nitrate + nitrite, and (d) diss-Fe/N ratio. The white numbers and lines in Figure 3a indicate the 26.6, 26.8, and 27.5 σ_θ isopycnal surfaces.

[11] Previous studies have shown that the strong vertical tidal mixing that reaches down to the OSIW occurs around the Kuril Straits [Yamamoto-Kawai *et al.*, 2004; Nakamura and Awaji, 2004]. Direct observations of turbulence reported by Itoh *et al.* [2010, 2011] and Yagi and Yasuda [2012] document strong vertical mixing at the straits due to interaction between the complicated topography and strong diurnal tidal currents, and the diffusivity there is 4 orders of magnitude higher (maximum diffusivity over $1 \times 10^{-1} \text{ m}^2 \text{ s}^{-1}$) than in the open ocean. The diapycnal mixing around the straits strongly affects the temperature, salinity, and dissolved oxygen properties from the surface to the deep layer [Ono *et al.*, 2007] and strongly influences the formation of North Pacific

Intermediate Water (NPIW) [Talley, 1991; Wong *et al.*, 1998; Nakamura and Awaji, 2004].

[12] The material flux in water flowing through the Kuril Islands chain (KIC) is an important issue. We estimated the vertical flux of dissolved Fe and nitrate + nitrite from the subsurface to the surface at Bussol' Strait using the equations

$$\begin{aligned} \text{dissolved Fe flux} &= K_p \times (d\text{Fe}/dz), \\ \text{nitrate + nitrite flux} &= K_p \times (dN/dz), \end{aligned}$$

where K_p is the vertical diffusivity and $d\text{Fe}/dz$ and dN/dz are the vertical gradients of dissolved Fe and nitrate + nitrite concentrations, respectively. Our measured vertical profiles of

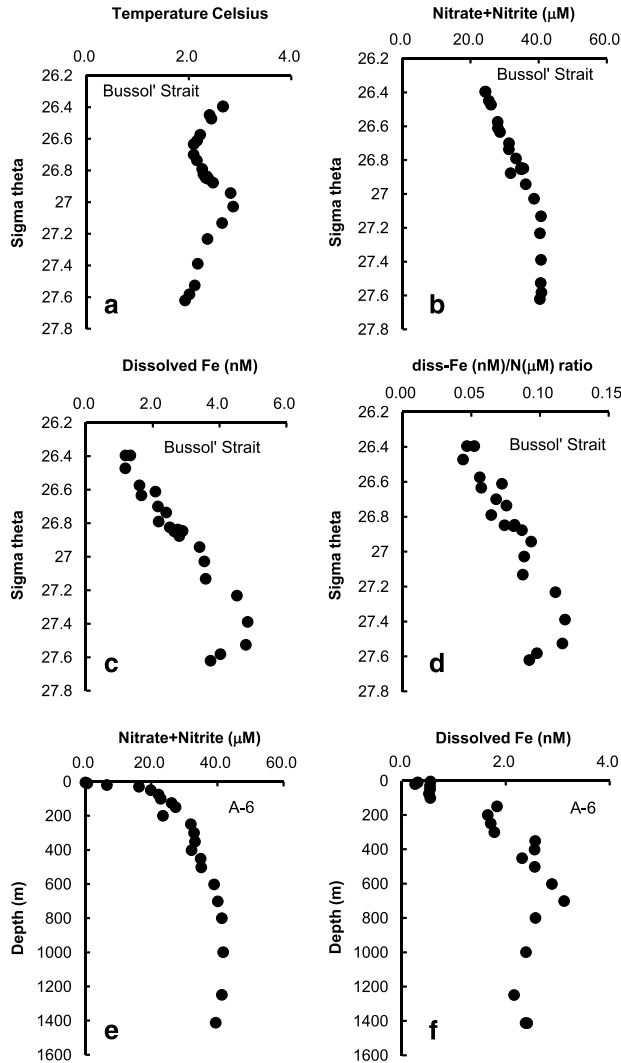


Figure 4. Vertical profiles of (a) temperature, (b) nitrate + nitrite, (c) dissolved Fe, and (d) diss-Fe/N ratio versus density at the Bussol' Strait. Vertical profiles of (e) nitrate + nitrite and (f) dissolved Fe versus depth at the station A-6 (see Figure 1a).

dissolved Fe and nitrate + nitrite in Bussol' Strait (Figures 4b and 4c) clearly display the influence of strong mixing in their disrupted gradients. Therefore, these gradients are not suitable for estimating material flux from intermediate to surface waters. Instead, we used the vertical profile obtained at station A6 (location is in Figure 1a, profiles in Figures 4e and 4f) in the Kuril Basin to approximate the state of the water before the mixing process. The vertical gradients of dissolved Fe (dFe/dz) and nitrate + nitrite (dN/dz) at station A6 are $0.0052 \mu\text{mol}/\text{m}^4$ and $0.073 \text{ mmol}/\text{m}^4$, respectively. Combining these gradients with 1 day average vertical diffusivity for depths of 100–500 m in Bussol' Strait reported by Yagi and Yasuda [2012] ($K_p = 1 \times 10^{-3} \text{ m}^2 \text{ s}^{-1}$), the estimated fluxes are $0.45 \mu\text{mol}/\text{m}^2 \text{ d}^{-1}$ for dissolved Fe and $6.3 \text{ mmol}/\text{m}^2 \text{ d}^{-1}$ for nitrate + nitrite. These fluxes are 2 orders of magnitude greater than that estimated in the nearby open ocean [Nishioka et al., 2007], indicating strong upward vertical transport around the Kuril Straits.

[13] The vertical sections of dissolved Fe (Figure 3b) and the ratio of dissolved Fe (in nM) to nitrate + nitrite (in μM), diss-Fe/N, (Figure 3d) indicate the impact of strong vertical mixing in the Kuril Straits. Dissolved Fe profiles in Bussol' Strait show that waters with a wide range of densities in the upper water column have high dissolved Fe concentrations ($>1 \text{ nM}$; Figures 3b and 4c), indicating that mixing in the Kuril Straits redistributes the Fe-rich OSIW and the Fe input from volcanogenic minerals from the Kuril/Kamchatka margin (see section 3.4). The mixing process helps to establish the chemical properties of the mixed water, including its high diss-Fe/N ratio (Figures 3d and 4d). Subsequently, these chemical properties broadly influence the subarctic, as well as the subtropical Pacific through NPIW formation (Figures 3b, 3d, and 5).

3.2. Fe Transport Into the North Pacific Intermediate Water

[14] NPIW is characterized by a salinity minimum at a potential density of approximately $26.8 \sigma_\theta$, and the core water of NPIW affects waters distributed over the subarctic and subtropical North Pacific [Ueno and Yasuda, 2000; You, 2003; Mitsudera et al., 2004; Masujima and Yasuda, 2009]. A tongue of high Fe concentration (with a high diss-Fe/N ratio) and low salinity extends southward at intermediate water depths (26.6 – $27.5 \sigma_\theta$) along the 155°E meridional section in the western North Pacific (Figures 3a, 3b, and 3d). These data indicate that the Fe-rich water mass is associated with NPIW formation and is transported from the subarctic region to the subtropical region (Figure 3b), with Fe decoupled from the nitrogen (regeneration) cycle (Figure 3c).

[15] Figure 5 shows the meridional section along 155°E with a previously published parallel meridional section along 165°E [Nishioka et al., 2007] and line P, which is the section from the south Vancouver Island to Ocean Station P (OSP) in the eastern subarctic Pacific (ESP) [Nishioka et al., 2001]. Both meridional sections display high dissolved Fe concentrations from near the surface to water as deep as $\sim 4000 \text{ m}$ in the subarctic area. This suggests that the Fe-rich mixed water from the Kuril Straits influences a broad range of depths over a wide area of the WSP. Although the line P section only reaches 1000 m in the ESP, a comparison of the Fe section in the WSP and line P indicates that the influence of the western Fe-rich intermediate water does not reach the ESP intermediate layer. Additionally, the full-depth vertical profile of dissolved Fe at station P reported by Martin et al. [1989] showed concentrations in deep water (below 1000 m) that were lower ($\sim 0.7 \text{ nM}$) than those in the WSP in our study ($\sim 1 \text{ nM}$). The difference in dissolved Fe concentrations between the WSP and the ESP is likely determined by the location of the Fe source region, the direction of the influence of the strong western boundary current, the efficiency of the remineralization of biogenic particles, and the scavenging effect during the long-range lateral transportation within the western subarctic and Alaskan gyres. To learn more details about the spatial variation of the west-east distribution and the controlling processes of Fe in the entire subarctic Pacific, more latitudinal transect data with higher resolution will need to be acquired.

[16] We observed cores of high Fe in intermediate water in the sections at 30°N , 155°E (Figure 3b) that corresponded oceanographically to one observed at 35°N , 165°E [Nishioka

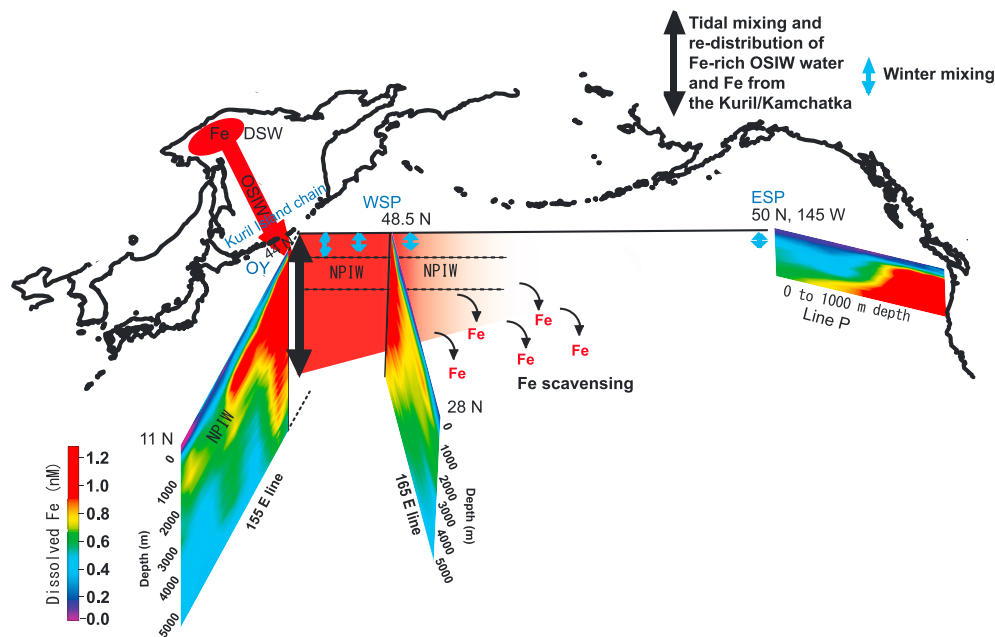


Figure 5. Illustration of the role of marginal sea and tidal mixing in the oceanic dissolved Fe cycle in the subarctic North Pacific, with section profiles of dissolved Fe measured in this study and profiles from previous studies (Line P and 165°E line in Figure 1a [Nishioka *et al.*, 2001, 2007]). Fe-rich intermediate water is indicated by the red arrow in the Sea of Okhotsk. Diapycnal mixing is indicated by the black arrow at the Kuril Straits. The mixing strongly influences the depth of the NPIW and around the depth of it (red shading in the subarctic Pacific). Winter deep vertical water mixing is indicated by blue arrows.

et al., 2007]. As Nishioka *et al.* [2007] described, the positions of these cores are consistent with the eastward flow of the Oyashio (OY), which is subducted below the warm Kuroshio (KR) extension water as estimated by numerical modeling [Mitsudera *et al.*, 2004]. Therefore, the high Fe core in intermediate water is influenced by the OY water pathway and suggests that Fe is a tracer of subarctic water in the subtropical region.

[17] The data from these two sections enable us to estimate the efficiency of eastward transport of dissolved Fe by the subarctic intermediate water and the intruded OY-originated water below the warm KR extension water. We evaluated the horizontal decay of dissolved Fe concentrations during eastward transport by the subarctic intermediate water from the OY region (40°30'N, 146°E) through KNOT station (44°N, 155°E) to SEEDS (48°30'N, 165°E) (see Figure 1) and during eastward flow of the intruded OY-originated water, from 30°N, 155°E to 35°N, 165°E. The scale length for dissolved Fe transport, defined as the distance over which the concentration drops to 1/e of the initial value, was estimated by plotting $\ln[Fe]$ from the dissolved Fe peak in the water column as a function of distance, as was done by Johnson *et al.* [1997], although we were limited to two or three data points. The slope of the decay of dissolved Fe in the subarctic intermediate water (-0.0003 km^{-1}) corresponds to a length scale of $\sim 3333 \text{ km}$, and that of dissolved Fe in the intruded OY-originated water (-0.0004 km^{-1}) corresponds to a length scale of $\sim 2500 \text{ km}$. This result means that the dissolved Fe was reduced to 59% during the subarctic water transportation from OY to SEEDS and to 65% during the transport of OY-originated water from 30°N, 155°E to 35°N, 165°E. These distances are greater than those observed in the Arctic

Ocean continental shelves, in the Nansen Basin ($\sim 260 \text{ km}$) by Klunder *et al.* [2012], but shorter than the distance ($\sim 5000 \text{ km}$) calculated by Johnson *et al.* [1997] from central California to the central Pacific within thermocline waters.

3.3. Fe in the Deep Water of the Subarctic Pacific

[18] In the section profile, water below 4000 m and south of 35°N has a low dissolved Fe concentration ($<0.5 \text{ nM}$) (Figure 3b) and a low diss-Fe/N ratio (Figure 3d). The position of this water is consistent with a branch of the Lower Circumpolar Deep Water (LCDW) [Kawabe and Fujio, 2010]. Our data reveal that the chemical properties of the transported LCDW are different from those of the North Pacific subarctic water, which includes the Fe supplied from the subpolar marginal sea and continental margin. The low dissolved Fe and low diss-Fe/N ratio may reflect a lower Fe input in the LCDW source area (the Southern Ocean) and removal of Fe during its transport to the North Pacific. These data also imply that the processes supplying Fe in the North Pacific are essential in determining the chemical properties of the upper subarctic water.

3.4. Transport of Fe From the Mesopelagic Layer to the Surface

[19] In the subarctic Pacific, physical processes that transport macronutrients and micronutrients from the mesopelagic layer to the surface have not been clearly identified. Sarmiento *et al.* [2004] used the combined distributions of silicic acid and nitrate to trace the main nutrient return path from deep water to above the thermocline (approximately $26.8 \sigma_\theta$ in the subarctic Pacific) and posited the existence of a return process in the northwest corner of the Pacific

Table 1. Quantitative Evaluation for Nitrate+Nitrite Drawdown From Winter to Summer, Residual Nitrate+Nitrite in Summer, Winter Mixed Layer Depth, diss-Fe/N Ratio Below the Winter Mixed Layer Depth in the Western Subarctic Pacific and the Eastern Subarctic Pacific

	Kuril Strait	Western Subarctic Pacific (Oyashio Region)	Western Subarctic Pacific (Oceanic Region)	Eastern Subarctic Pacific (OSP)
Nitrate + nitrite drawdown from winter to summer (μM) ^a		20 to 22	15 to 20	7 to 10
Residual nitrate + nitrite in summer (μM) ^a		0 ~	5	8
Winter mixed layer depth (m) ^b		100 to 200	~ 100	~ 80
diss-Fe/N (nM/ μM) ratio in intermediate water ^c	0.057 ± 0.022 ($n=72$)	0.033 ± 0.013 ($n=34$)	0.022 ± 0.006 ($n=20$)	0.009 ± 0.003 ($n=31$)
Number of data profiles for diss-Fe/N ratio	Four profiles (two Bussols, Urup East, Urup West from this study)	Five profiles (four profiles from Nishioka <i>et al.</i> [2007, 2011] and one from this study)	Three profiles (SEEDS, KNOT from Nishioka <i>et al.</i> [2007] and KNOT from this study)	Five profiles (four OSP from Nishioka <i>et al.</i> [2001] and one OSP from Martin <i>et al.</i> [1989])

^aData are referred from Tsurushima *et al.* [2002], Whitney and Freeland [1999], and Whitney [2011].

^bData are referred from Nishioka *et al.* [2001, 2007, 2011].

^cData for Oyashio region are from reference Nishioka *et al.* [2007, 2011] and this study, data for the western subarctic Pacific oceanic region are from reference Nishioka *et al.* [2007] and this study, and data for OSP are from reference Nishioka *et al.* [2001] and Martin *et al.* [1989]. All numbers are mean $\pm$ 1 SD values of diss-Fe/N ratio which is calculated by the data from subsurface gradient below winter mixed layer depth.

where there is enhanced vertical mixing, perhaps driven by tidal mixing at the KIC. Our data support their proposed mechanism. Low surface temperatures (2–3°C) were observed in Bussol' Strait (Figure 4a), and the vertical profiles of dissolved Fe and nitrate + nitrite in the surface to subsurface water showed atypically high concentrations (Figures 4b and 4c). This resulted in a high diss-Fe/N ratio (approximately 0.05) in water shallower than 26.6 σ_θ isopycnal surface (Figure 4d). Strong diapycnal tidal mixing at the Kuril Straits redistributes these Fe- and nutrient-rich intermediate water across a wide range of densities (Figures 3b, 4b, and 4c), as discussed in section 3.1 [Ono *et al.*, 2007; Itoh *et al.*, 2010, 2011; Yagi and Yasuda, 2012]. The section profiles (Figures 3b and 3d) and vertical profiles at Bussol' Strait (Figures 4a–4d) indicate that strong vertical tidal mixing in the Kuril Straits transports Fe and nutrients from deep water to water shallower than 26.6 σ_θ isopycnal surface and determines the diss-Fe/N ratio (Figure 4d). The Fe-rich water that is mixed at the Kuril Straits influences subsurface water over a broad depth range in the WSP and throughout the subarctic via NPIW transport (Figures 3b, 3d, and 5).

[20] Table 1 lists the diss-Fe/N ratios from the bottom of the winter mixed layer to the intermediate layer in the subarctic Pacific (~26.5–27.0 σ_θ in the WSP and ~26.0–27.0 σ_θ in the ESP). In the WSP, Talley [1991] reported that winter surface mixing is sufficiently deep to reach intermediate water, and dense water of 26.6–26.7 σ_θ appears in the winter at the surface around the Kuril Islands and the OY and OY-KR transition region. Additionally, Nishioka *et al.* [2011] found that intermediate water, which has a high diss-Fe/N ratio, directly outcrops in the OY and the OY-KR transition region. A similar process likely occurs in the oceanic region during winter mixing. Figure 6 shows a snapshot of the winter vertical profiles of temperature (Figures 6a–1, 6b–1, and 6c–1), nitrate + nitrite concentration (Figures 6a–2, 6b–2, and 6c–2), dissolved Fe concentration (Figures 6a–3, 6b–3, and 6c–3), and diss-Fe/N ratio (Figures 6a–4, 6b–4, and 6c–4) in the OY region, the WSP (station KNOT), and the ESP (OSP). The vertical profiles of the diss-Fe/N ratio (Figures 6a–4, 6b–4, and 6c–4) above and below the thermocline (identified from temperature profile, Figures 6a–1, 6b–1, and 6c–1) show

similar values throughout the water column, indicating that the chemical property of the water below the thermocline is lifted directly into the surface mixed layer instead of being transported by eddy diffusion (see below). Additionally, the observed diss-Fe/N ratios in the winter surface and intermediate waters in the OY (Figure 6a–4) and the WSP (Figure 6b–4) are >3 to 2 times higher than that of the ESP (Figure 6c–4 and Table 1). Nishioka *et al.* [2007] estimated the ratio of the Fe and nitrate + nitrite gradients ((dFe/dz)/(dN/dz)) in the OY region, the WSP, and the ESP to be 0.044, 0.052, and 0.004, respectively. If the same global mean coefficient of eddy diffusivity ($5 \text{ m}^2 \text{ d}^{-1} = 5.8 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$) is assumed for all regions, the resulting diss-Fe/N ratio in the surface WSP would be 10 times that of the ESP, which is inconsistent with the observations (Figures 6a–4, 6b–4, and 6c–4). Thus, if vertical eddy diffusion is the dominant process for transporting Fe and nutrients from intermediate water to the surface layer, then the similar diss-Fe/N ratio above and below the thermocline and the relative relationship of the winter surface diss-Fe/N ratio among the three regions (the OY, the WSP, and the ESP) cannot be explained. Therefore, the upward transport of materials from the intermediate water to the surface by tidal mixing at the Kuril Straits and entrainment by winter deep vertical water mixing [Talley, 1991; Suga *et al.*, 2004] must be important mechanisms of Fe and nutrient supply in the subarctic region.

3.5. Moderate Value Determined by Mixing Along an Island Chain

[21] The chemical properties of subsurface water, which are controlled by transportation of Fe-rich OSIW, Fe input in the Kuril Islands region, and diapycnal mixing in the Kuril Straits, influence the biogeochemical processes in the subarctic North Pacific. Previous studies have shown that the seasonal amplitude of $p\text{CO}_2$ in surface water is higher in the WSP than the ESP and that the biological drawdown effect on $p\text{CO}_2$ is especially intense at the edge of the western subarctic gyre in the OY and the OY-KR transition zone (Figure 1b) [Takahashi *et al.*, 2002]. The annual amplitude in biogeochemical processes was compared in the WSP and the ESP based on observations from two time series stations.

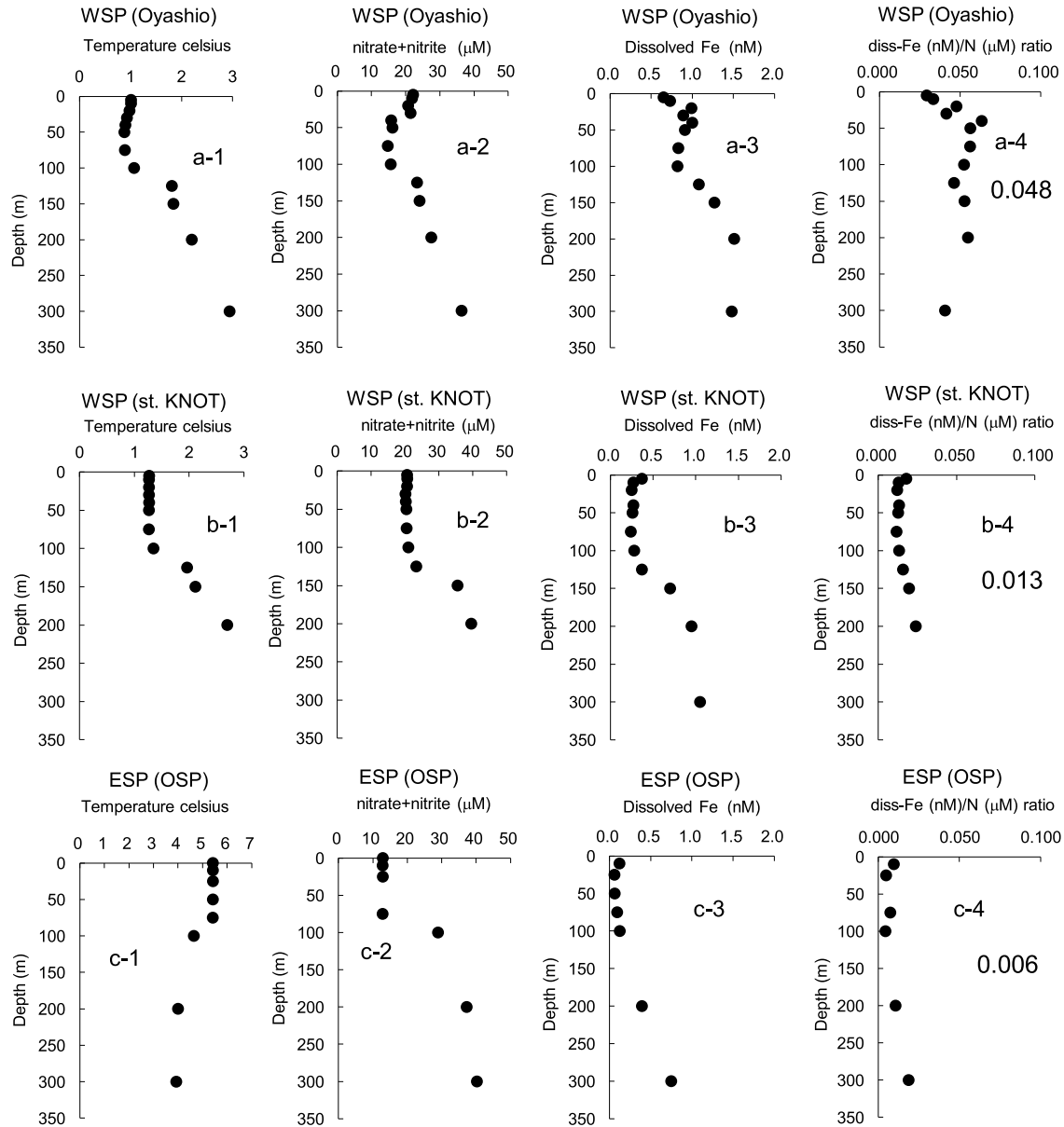


Figure 6. Winter vertical profiles of (1) temperature, (2) nitrate + nitrite concentration, (3) dissolved Fe concentration, and (4) diss-Fe/N ratio (a) in the OY (February 2003 at OY in Figure 1a), (b) in the WSP (March 2003 at station KNOT in Figure 1a), and (c) in the ESP (February 1999 at station P in Figure 1a). Numbers in Figures 6a–4, 6b–4, and 6c–4 indicate the average value of the diss-Fe/N ratio in the surface mixed layer. Data for the WSP and ESP are from Nishioka *et al.* [2001, 2007].

The seasonal nitrate drawdown from spring to summer in the WSP ($\sim 15\text{--}20\ \mu\text{M}$) is consistently twice that of the ESP ($7\text{--}10\ \mu\text{M}$), and substantial residual nitrate concentrations remained during summer in both regions ($\sim 5\ \mu\text{M}$ in the WSP, $\sim 8\ \mu\text{M}$ in the ESP) [Whitney and Freeland, 1999; Tsurushima *et al.*, 2002; Whitney, 2011] (Table 1). Observations in the OY, at the edge of the WSP, have documented nitrate consumption 3 times as high ($>20\ \mu\text{M}$) as that in the ESP (Table 1) and have shown that both nitrate and Fe become limiting nutrients after the spring bloom season [Nishioka *et al.*, 2011]. To explain the difference in seasonal amplitude in these two regions of the subarctic Pacific (stronger phytoplankton growth and greater nitrate consumption in the WSP than the ESP), the Fe supply must be higher in the west than the

east, but not sufficient enough to cause nutrient depletion. That is, the Fe supply into the WSP should have a moderate value which still results in HNLC condition.

[22] Previous studies have shown that high mineral dust concentrations are frequently observed in spring, and the flux of dust containing Fe over the WSP is an order of magnitude greater than in the ESP [Duce and Tindale, 1991; Mahowald *et al.*, 2005; Luo *et al.*, 2003]. Also, eddy transport from the continental margin is an important mechanism supplying Fe to the open ocean [e.g., Johnson *et al.*, 2005; Lippiatt *et al.*, 2011]. However, these processes would cause spatiotemporal heterogeneity in the Fe supply, and it is difficult to explain how such intermittent sources could result in a moderate Fe supply each year.

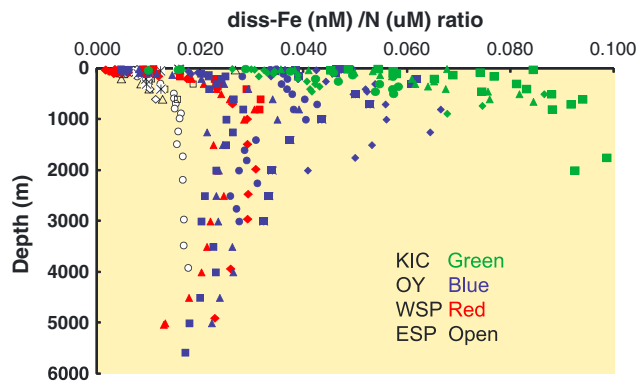


Figure 7. Comparison of vertical profiles of the diss-Fe/N ratio in the Kuril Islands chain (KIC) (green symbols; four profiles at Bussol, Urup E, and Urup W in Figure 1a), the OY region (blue symbols; five profiles at OY in Figure 1a), the WSP (red symbols; three profiles at KNOT and SEEDS in Figure 1a), and the ESP (open symbols; five profiles at OSP in Figure 1a).

[23] Only Fe transport from the Sea of Okhotsk into intermediate waters of the subarctic Pacific can explain the moderate value observed over much of the western North Pacific. A substantially high diss-Fe/N ratio of 0.022 ± 0.006 ($\text{avg} \pm 1 \text{ SD}$) (Table 1) in the NPIW of the WSP was observed along our survey section. The level of diss-Fe/N ratio in the WSP and the OY intermediate waters ranges from 3 (OY) to 2 times (WSP) higher than that of the intermediate layer in the ESP (Table 1). High diss-Fe/N ratios indicate that intermediate waters in the OY and WSP regions have a great potential to stimulate phytoplankton growth through high Fe availability, allowing higher utilization of macronutrients when intermediate water are supplied to the surface and maintain surface diss-Fe/N ratios as discussed in section 3.4. Concurrently, the diss-Fe/N ratios in the OY, the WSP, and the ESP can explain the relative relationship of macronutrient consumption in these three regions and the value in the WSP ranges to the moderate value that does not cause nutrient depletion (Table 1). Oceanic phytoplankton require a diss-Fe/N ratio of 0.026 (using the dominant diatom *Pseudo-nitzschia* spp. in the subarctic Pacific, minimum requirement of $4 \mu\text{M}$ Fe/mol C in Fe-limited conditions, calculated with the Redfield ratio) [Marchetti et al., 2006]. This value, combined with the intermediate diss-Fe/N supply ratio (Table 1), can quantitatively explain the residual nitrate in the WSP and the ESP during summer, and it describes the WSP as mildly Fe limited and the ESP as strongly Fe limited. According to Whitney [2011], the drawdowns of phosphate in the OY region, the WSP, and the ESP are 1.07, 0.8, and $0.54 \mu\text{M}$, respectively. The west-east gradient of diss-Fe/P ratios is smaller than the gradient in nitrate. This contrast might reflect the ambient diatom physiology, as Fe limitation causes a decline in the N:P uptake ratio [Price, 2005].

[24] Figure 7 compares vertical profiles of diss-Fe/N ratios for the Kuril Straits, the OY region, the WSP, and the ESP. The order of diss-Fe/N ratios in these four regions can be explained by their distance from the original Fe source and the physical dynamics of currents with loss of Fe during transport eastward (Figures 5 and 7). These results suggest

that the diss-Fe/N ratio in the OY and the moderate Fe value of the WSP are controlled by Fe discharge from the marginal sea and tidal mixing around the KIC (Figures 3b and 5).

4. Conclusion

[25] The existence of a subpolar marginal sea that connects with the North Pacific through the KIC is essential, combined with the strong diapycnal mixing in the KIC, for controlling the moderate level of Fe supply and the seasonal amplitude of the biogeochemical cycle in the subarctic Pacific. This natural Fe enrichment system simultaneously explains one of the largest biological pumps in the world [Buesseler et al., 2007] and the formation of the HNLC region in the North Pacific [Tsuda et al., 2003; Boyd et al., 2004].

[26] In the oceanic region, the present export of organic matter is large enough to deplete the nutrients in the waters above the thermocline within less than 60 years [Sarmiento and Gruber, 2006]. The process returning nutrients from the deep ocean to the thermocline is thus critical for maintaining the biological productivity and formation of the HNLC region. However, the physical processes transporting macronutrients and micronutrient from the mesopelagic layer to the surface have not been clearly identified. Our results provide observational evidence that strong tidal mixing in the KIC at the margin of the Pacific Ocean plays a pivotal role in transporting Fe and nutrients from deep water to the surface.

[27] Recent weakening of overturning in the NPIW has been reported [Nakanowatari et al., 2007]. Additionally, researchers have suggested a link between bi decadal variations in the NPIW and the 18.6 year modulation of diurnal tidal mixing [Watanabe et al., 2003; Yasuda et al., 2006]. Clarifying what causes these long-term changes of oceanic physical and biogeochemical conditions is an essential step for predicting future global climate changes. Our findings contribute to the understanding of the mechanisms controlling the biogeochemical cycles in the North Pacific.

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