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# Helium isotopes of seawater in adjacent sea of Nansei Islands, Southwest Japan

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We have measured helium isotopic ratios of 43 water samples with various depths collected in Eastern East China Sea and Northwestern Philippine Sea, adjacent region of Nansei Islands, Southwestern Japan. The $^3He/^4He$ ratios vary significantly from 0.994 $R_{atm}$ to 1.242 $R_{atm}$ where $R_{atm}$ is the atmospheric ratio of $1.39 \times 10^{-6}$. Mid-depth (750–1500 m) samples in this study would be affected by the source of subduction-type mantle helium in the Okinawa Trough. Noble gas abundances (neon, argon, krypton and xenon) were measured in 24 water samples with various depths collected in Northwestern Pacific Ocean and Northwestern Philippine Sea (Nansei Trench). Neon abundances show slight excess relative to air saturated seawater at ambient temperature and salinity. This may be due to either air bubble effect or contamination during the sampling. When these effects are corrected using the neon anomaly, heavier noble gas abundances (argon, krypton and xenon) of samples with the temperature higher than 5°C (shallower than 500 m) agree well with those of calculated air saturated seawater, while the lower temperature samples (deeper than 500 m) indicate anomaly of –7% to +10%.

Keywords: $^3He/^4He$ ratio, mantle helium, heavy noble gas, Okinawa Trough, seawater

## INTRODUCTION

The $^3He/^4He$ ratios of mantle derived samples such as mid-ocean ridge basalts and volcanic gases in island arcs show high values of about $1 \times 10^{-5}$, while those of granitic rocks and continental natural gases are low with the ratio of about $1 \times 10^{-7}$ (Lupton, 1983; Mamyrin and Tolstikhin, 1984; Sano and Wakita, 1985). The $^3He/^4He$ ratio is one of the most sensitive and conservative tracer in chemical oceanography (e.g., Jenkins *et al*., 1972; Craig *et al*., 1975; Sano *et al*., 1995) because of the primordial signature, rapid mobility and chemical inertness of the isotopes. Even though more than 1,700 $^3He/^4He$ measurements were carried out in the three main oceans, helium isotope data in the neighboring waters of the Japanese Islands are very sparse in the literature (Östlund *et al*., 1987; Igarashi *et al*., 1987; Ishibashi *et al*., 1988; Sano *et al*., 1989). Recently Sano *et al*. (2004) have reported the $^3He/^4He$ ratios of Pacific water samples with various depths collected in adjacent region of Honshu, Japan. Mid-depth (750–1500 m) profile of $^3He/^4He$ ratios at Northwestern Pacific Ocean (Off Joban) was significantly different from that at Northern Philippine Sea (Nankai Trough), suggesting that these waters were separated by a topographic barrier of Izu-Ogasawara Ridge. Higher $^3He/^4He$ ratio at the Nankai Trough was attributable to the hypothetical source of the subduction-type mantle helium in the Okinawa Trough. In order to verify the source of the mantle helium, we study the $^3He/^4He$ ratios of water from Eastern East China Sea and Northwestern Philippine Sea, adjacent region of Nansei Islands, Southwestern Japan.

Noble gases (helium, neon, argon, krypton and xenon) can be used as conservative tracers to constrain possible origin and trajectory of deep seawaters. The dissolved gas abundances in the sea would reflect the temperature of the open sea surface where deep water formed from the surface waters, since the varying concentrations of noble gases are controlled by their respective solubility equilibrated with air. However, there are only three data set of noble gas abundances (from helium to xenon) in deep sea water in literatures (Bieri *et al*., 1964; Mazor *et al*., 1964; Igarashi *et al*., 1987). We report here preliminary data of heavier noble gas abundances in Northwestern Pacific Ocean and Northwestern Philippine Sea.

## EXPERIMENTAL METHOD

Seawater sampling was conducted on the KT-02-14 cruise of the Research Vessel, Tansei Maru of Ocean Research Institute, the University of Tokyo (October 1–9, 2002) in the adjacent region of Nansei Islands, Southwestern Japan, those are ST1 (East of East China Sea; adjacent Kyushu Island), ST7 (East of East China Sea; northeast of Okinawa Trough), ST9 (Northwestern Philippine Sea; Nansei Trench), ST10 (Northwestern

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*Table 1. Location and bottom depth of sampling stations together with sampling depth of adjacent sea of Nansei Islands*

| Station | Location | Bottom depth (m) | Sampling depth (m) |
|---|---|---|---|
| ST1 | 32°00′ N, 129°20′ E | 740 | 10, 100, 200, 400, 500 |
| ST7 | 27°20′ N, 126°50′ E | 1560 | 10, 100, 200, 400, 600, 800, 1000, 1250, 1500 |
| ST9 | 24°00′ N, 127°00′ E | 6490 | 10, 50, 500, 1000, 1500, 2000, 3000, 3500, 4000, 4500, 5000 5500, 6000 |
| ST10 | 23°30′ N, 123°10′ E | 3990 | 10, 50, 250, 500, 750, 1000, 1500, 2000, 2500, 3000 |
| ST11 | 25°20′ N, 124°00′ E | 2050 | 100, 300, 500, 750, 1000, 1250, 1500, 2000 |

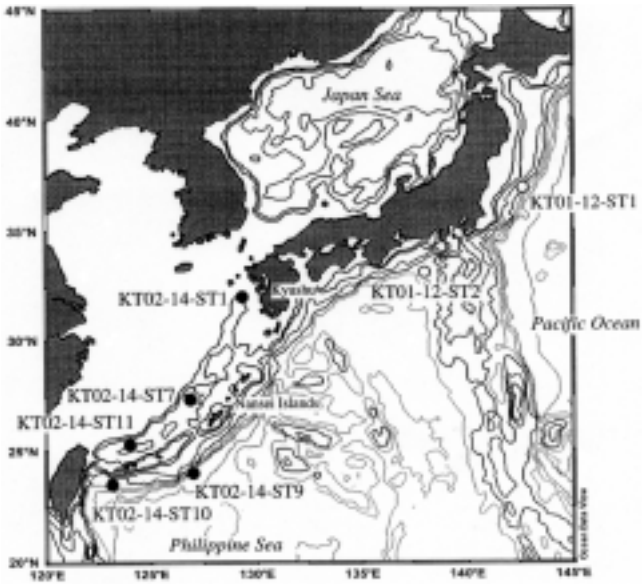


*Fig. 1. Sampling sites (KT02-14-ST1, ST7, ST9, ST10 and ST11) of seawaters from adjacent sea of Nansei Islands, Southwest Japan (closed circles) together with those of KT01-12 cruise (open circles). Position of active seafloor hydrothermal systems in the Okinawa Trough was also plotted (star). This map was drawn by the Ocean Data View software (http://www.awi-bremerhaven.de/GEO/ODV).*

Philippine Sea; south of Iriomote Island) and ST11 (East of East China Sea; southwest of Okinawa Trough). The sampling points are shown in Fig. 1. Samples for helium isotope measurements were collected from 4 depths at ST1, and from 8–12 depths at ST7, ST9, ST10 and ST11 by using a CTD carousel system equipped with 10-liter Niskin bottles. Details are listed in Table 1. Seawater was transferred without exposure to atmosphere from the Niskin bottles into about 30 $cm^3$ copper-tubing containers for storage (Sano *et al.*, 1989). For heavier noble gas abundance measurements, we have collected seawater samples in about 8 $cm^3$ copper-tubing containers at ST9 of the KT-02-14 cruise and ST1 of the KT-01-12 cruise (August 3–9, 2001), located Northwestern Pacific Ocean east of Japan (Off Joban; 37°00′ N, 142°40′ E).

At the laboratory the 30 $cm^3$ copper container was connected to a stainless steel high vacuum line and dissolved gases of seawaters were extracted in vacuo. Helium in the exsolved gases was purified using hot titanium-zirconium getters and charcoal traps held at liquid nitrogen temperature. Helium was separated from neon by using a cryogenic charcoal trap held at 40 K, since the neon may introduce a small error in the $^3He/^4He$ ratio of the terrestrial samples (Rison and Craig, 1983; Sano and Wakita, 1988). The $^3He/^4He$ ratios were measured on a conventional noble gas mass spectrometer (VG5400, MicroMass Co.) which was moved from Laboratory for Earthquake Chemistry, the University of Tokyo and reinstalled at Ocean Research Institute. Ion beams of $^3He$ and $^4He$ were measured by a double collector system. A resolving power of ~550 at 1% of the peak height was used for the complete separation of the $^3He^+$ beam from those of $H_3^+$ and $HD^+$. The observed $^3He/^4He$ ratios of samples were calibrated against atmospheric helium collected in August 2000 in Higashi Hiroshima, Japan. Experimental error of $^3He/^4He$ ratio is 1~2% ($1\sigma$) estimated by repeated measurements of standard air whose amount is equivalent to those of samples. Also to check the accuracy of the analytical system, we analyzed air equilibrated seawater samples taken from a water reservoir in a room controlled thermostatically. Observed data agree well with the reported values in a literature within the experimental error. The helium blank level (the same experimental procedure without the sample) was less than 5% of the samples, which hardly affected the calibrated ratios.

Noble gas abundances (neon, argon, krypton and xenon) of samples collected at ST9 of the KT-02-14 cruise and ST1 of the KT-01-12 cruise were measured by a quadrupole-based mass spectrometric system. After known amounts of isotopic spikes ($^{22}Ne$, $^{36}Ar$, $^{86}Kr$ and $^{124}Xe$) were introduced into the preparation vacuum line, dissolved gases were extracted from seawater and well mixed with the spikes by ultra-sonic vibration. Noble gases were purified using three stage hot Ti getters and separated by two activated charcoal traps held at low temperature (liquid nitrogen and dry-ice ethanol). Abundances

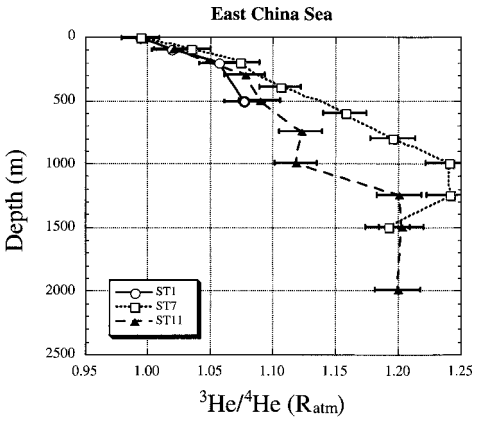


*Fig. 2. Helium isotopic profiles of Eastern East China Sea (KT02-14-ST1, ST7 and ST11). There is a significant discrepancy at mid-depth from 500 m to 1250 m between ST7 and ST11. Error assigned to the symbol is one sigma.*

of noble gases were measured by a relatively inexpensive quadrupole mass spectrometer (QMG422, Pfeiffer Vacuum Co.) based on the isotope dilution technique. Experimental details will be given elsewhere (Sano and Takahata, 2004).

## RESULTS AND DISCUSSION

### *Helium isotopes in Eastern East China Sea*

Observed $^3He/^4He$ ratios are listed in Table 2 by $R_{atm}$ notation where $R_{atm}$ is the atmospheric ratio of $1.39 \times 10^{-6}$ (Lupton, 1983). Temperature and salinity are also listed in Table 2. The $^3He/^4He$ ratios of Eastern East China Sea (ST1, ST7 and ST11) are ranging from 0.994 $R_{atm}$ to 1.242 $R_{atm}$. Figure 2 shows the helium isotope profiles at ST1, ST7 and ST11. It is well known that the mid-depth of all over the western North Pacific water is affected by the mantle helium with a high $^3He/^4He$ ratio probably derived from East Pacific Rise (Östlund *et al.*, 1987; Lupton, 1995; Sano *et al.*, 1995, 2004; Pacific Marine Environmental Laboratory, 2003). Observed data with excess $^3He$ relative to the air at mid-depth are similar to those of North Pacific water. The excess may be due to the mantle helium derived from East Pacific Rise, about 12,000 km distant from the sampling sites, suggesting that helium isotopic ratio is the most sensitive and conservative tracer in chemical oceanography.

More precisely the ST7 profile is identical to ST1 and ST11 from the surface to the 500 m deep. In contrast there is a significant difference of data from 600 m to 1250 m deep. This suggests that East China Sea at ST7 is well mixed with those of ST1 and ST11 above 500 m and they are probably separated or independent beneath 500 m. However in the region between ST7 and ST11, there is no topographic high which blocks the water mass of up to 500 m in the Okinawa Trough. It is difficult to explain the difference of 10% excess $^3He$ at 1000 m deep by radioactive decay of natural tritium (Craig and Clarke, 1970). The excess is possibly due to the subduction-type mantle helium (Sano and Wakita, 1985) with a high $^3He/$

*Table 2. Potential temperature, salinity and helium isotope ratios of Eastern East China Sea and Northwestern Philippine Sea water*

| Station | Pressure (db) | Temperature (°C) | Salinity | $^3He/^4He$ ($R_{atm}$) |
|---|---|---|---|---|
| ST1 | 9.4 | 26.076 | 33.873 | 0.994 |
| | 101 | 20.651 | 34.555 | 1.019 |
| | 200 | 14.017 | 34.557 | 1.057 |
| | 400 | 8.561 | 34.376 | — |
| | 500 | 7.375 | 34.375 | 1.077 |
| ST7 | 10.1 | 27.601 | 34.403 | 0.995 |
| | 100 | 22.605 | 34.842 | 1.035 |
| | 200 | 18.688 | 34.789 | 1.074 |
| | 400 | 13.289 | 34.501 | 1.107 |
| | 599 | 7.723 | 34.275 | 1.168 |
| | 799 | 5.482 | 34.375 | 1.206 |
| | 1001 | 4.357 | 34.415 | 1.241 |
| | 1250 | 3.860 | 34.446 | 1.242 |
| | 1504 | 3.686 | 34.456 | 1.164 |
| ST9 | 9.7 | 28.161 | 34.506 | 0.995 |
| | 49.8 | 28.064 | 34.492 | 0.997 |
| | 501 | 11.267 | 34.341 | 1.084 |
| | 999 | 4.025 | 34.350 | 1.146 |
| | 1500 | 2.609 | 34.564 | — |
| | 1999 | 1.894 | 34.624 | 1.226 |
| | 3001 | 1.371 | 34.670 | 1.182 |
| | 3489 | 1.318 | 34.679 | 1.163 |
| | 3945 | 1.265 | 34.683 | 1.136 |
| | 4510 | 1.218 | 34.691 | 1.120 |
| | 5039 | 1.206 | 34.689 | 1.114 |
| | 5557 | 1.208 | 34.689 | 1.108 |
| | 6081 | 1.210 | 34.686 | 1.102 |
| ST10 | 11.2 | 27.919 | 34.338 | 1.004 |
| | 50.1 | 27.478 | 34.548 | 1.011 |
| | 252 | 17.069 | 34.745 | 1.048 |
| | 502 | 9.579 | 34.280 | 1.068 |
| | 751 | 5.458 | 34.303 | 1.166 |
| | 1001 | 3.819 | 34.436 | 1.182 |
| | 1501 | 2.413 | 34.576 | 1.228 |
| | 2001 | 1.833 | 34.621 | 1.235 |
| | 2502 | 1.489 | 34.661 | 1.216 |
| | 3005 | 1.315 | 34.673 | 1.193 |
| ST11 | 101 | 22.917 | 34.849 | 1.021 |
| | 301 | 15.332 | 34.649 | 1.078 |
| | 501 | 10.066 | 34.359 | 1.090 |
| | 751 | 5.708 | 34.396 | 1.123 |
| | 1001 | 4.284 | 34.437 | 1.119 |
| | 1250 | 3.870 | 34.462 | 1.201 |
| | 1502 | 3.725 | 34.466 | 1.203 |
| | 2000 | 3.581 | 34.453 | 1.200 |

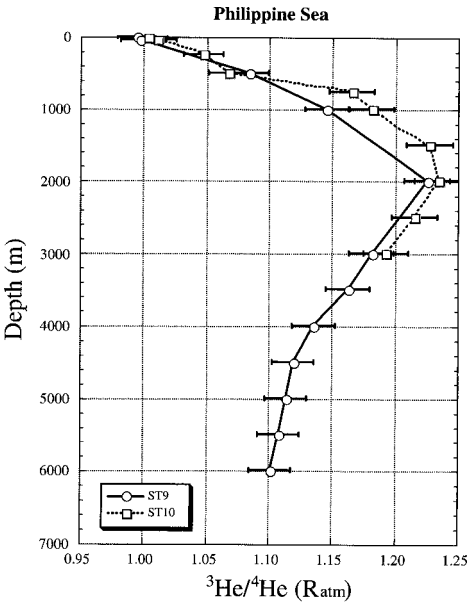


*Fig. 3. Helium isotopic profiles of Northwestern Philippine Sea (KT02-14-ST9 and ST10). Error assigned to the symbol is one sigma.*

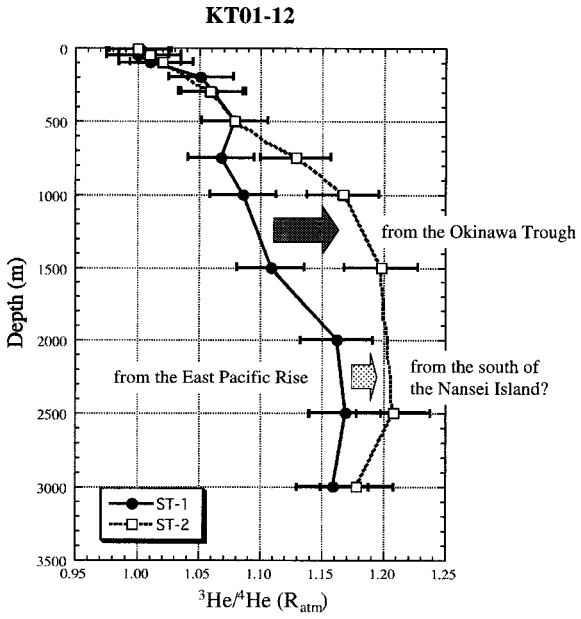


*Fig. 4. Helium isotopic profiles of Off Joban (KT01-12-ST1) and Nankai Trough (ST2) (Sano et al., 2004). There is a significant discrepancy at mid-depth from 750 m to 2000 m. Hydrothermal activity in the Okinawa Trough can be candidate to explain the difference in mid-depth water. Error assigned to the symbol is one sigma.*

$^4$He ratio originated from the Okinawa Trough. Ishibashi *et al.* (1988) found a distinct hydrothermal plume in the Iheya Graben very close to ST7. The maximum $^3He/^4He$ ratio in the plume is found at similar depth (about 1300 m) though it is higher than that at ST7. Ishibashi *et al.* (1995) observed that significant mantle helium with the $^3He/^4He$ ratio of 3~6 $R_{atm}$, which is a typical subduction-type helium, is currently releasing from two high-temperature hydrothermal sites, the JADE and CLAM sites in the Okinawa Trough. There are two very active hydrothermal fields at Izena cauldron (including JADE site) and Iheya North in the mid-Okinawa Trough. Both of them is closer to ST7 than ST11 and the water depth of them is about 1000~1400 m deep. It is well known that the maximum excess $^3$He was discovered at about 200 m above the volcano in the East Pacific Rise (Lupton and Craig, 1981). Taking account of different depth of hydrothermal activities between in the Okinawa Trough and in the East Pacific Rise, maximum excess $^3$He may appear in the depth of several hundreds meters above the hydrothermal fields (about 800~1400 m) if it is originated from there. This is consistent with the fact that larger excess $^3$He is observed at ST7 than ST11 in the depth from 600 m to 1250 m. Thus the $^3He/^4He$ difference may be attributable to the additional source of mantle helium in the Okinawa Trough.

*Helium isotopes in Northwestern Philippine Sea*

The $^3He/^4He$ ratios of Northwestern Philippine Sea (ST9 and ST10) vary from 0.995 $R_{atm}$ to 1.235 $R_{atm}$, which is comparable to those of Eastern East China Sea. Figure 3 shows the helium isotope profiles at ST9 and ST10. Maximum $^3$He excess relative to the air is observed at about 2000 m deep, which is different from that of Eastern East China Sea at 1000~1250 m deep (see Fig. 2). The helium profiles of Northwestern Philippine Sea are rather similar to the profile found at Nankai Trough, Northern Philippine Sea south of Honshu Island (KT01-12-ST2 of Fig. 4). This would be due to the location of sampling sites; Northwestern Philippine Sea is far from the source of mantle helium in the Okinawa Trough compared to Eastern East China Sea.

Sano *et al.* (2004) reported that there is a difference of $^3He/^4He$ ratio in mid-depth (from 750 m to 1500 m) of Pacific water between Off Joban located east of North Japan (KT01-12-ST1 of Fig. 1) and Nankai Trough (KT01-12-ST2), suggesting that these waters were separated by a topographic barrier of Izu-Ogasawara Ridge (Fig. 1). All the 20% excess $^3$He observed at Nankai Trough may not be explained by the helium plume originated from the East Pacific Rise, since the maximum excess is constrained to about 15% in western North Pacific (Lupton, 1995). Additional source of mantle helium (additional 5% excess) at about 2000~2500 m deep should be considered in Nankai Trough. Sano *et al.* (2004) suggested that hydrothermal activity in the Okinawa Trough can be a candidate to explain the difference of

*Table 3. Noble gas abundances of sea water from Northwestern Pacific Ocean (Off Joban, KT01-12-ST1) and Northwestern Philippine Sea (Nansei Trench, KT02-14-ST9)*

| Pressure (db) | Ne ($\times 10^{-7}$ cm$^3$STP/g) | Ar ($\times 10^{-4}$ cm$^3$STP/g) | Kr ($\times 10^{-8}$ cm$^3$STP/g) | Xe ($\times 10^{-9}$ cm$^3$STP/g) |
|---|---|---|---|---|
| KT01-12-ST1 | | | | |
| 4.8 | 1.43 | 2.25 | 4.86 | 6.30 |
| 49.4 | 1.57 | 2.63 | 5.92 | 8.04 |
| 99.7 | 1.67 | 2.90 | 6.76 | 9.30 |
| 200 | 1.69 | 2.99 | 7.00 | 10.0 |
| 299 | 1.69 | 3.29 | 7.86 | 11.5 |
| 501 | 1.89 | 3.53 | 8.43 | 12.4 |
| 749 | 1.88 | 3.83 | 9.40 | 13.6 |
| 1001 | 1.71 | 3.51 | 8.34 | 12.0 |
| 1500 | 1.80 | 3.89 | 9.35 | 13.0 |
| 2002 | 1.82 | 3.89 | 9.91 | 13.6 |
| 2502 | 1.72 | 3.84 | 9.56 | 14.0 |
| 2999 | 1.74 | 3.79 | 9.46 | 13.8 |
| KT02-14-ST9 | | | | |
| 9.7 | 1.51 | 2.21 | 4.73 | 6.08 |
| 49.8 | 1.54 | 2.23 | 4.82 | 6.14 |
| 501 | 1.71 | 3.03 | 7.09 | 9.94 |
| 999 | 1.85 | 3.60 | 9.10 | 13.4 |
| 1999 | 1.73 | 3.50 | 8.32 | 12.5 |
| 3001 | 1.82 | 3.69 | 9.61 | 14.6 |
| 3489 | 1.90 | 3.79 | 9.56 | 14.2 |
| 3945 | 1.74 | 3.67 | 8.88 | 13.2 |
| 4510 | 1.70 | 3.38 | 8.37 | 13.3 |
| 5039 | 1.85 | 3.93 | 9.71 | 14.3 |
| 5557 | 1.75 | 3.64 | 8.85 | 13.9 |
| 6081 | 1.93 | 3.99 | 9.65 | 14.3 |

*Error of abundance is about 4% (2$\sigma$).*

$^3$He/$^4$He ratio in mid-depth of Pacific water (Fig. 4). This is the case since Eastern East China Sea (Okinawa Trough) show significant excess $^3$He at 1000–1250 m deep.

The additional 5% excess at about 2000~2500 m deep found at Nankai Trough should be explained by the other source than the Okinawa Trough. Fujio *et al*. (1992) reported that abyssal current of 2000 m deep in Philippine Sea is northeastward along the Okinawa-Amami Islands and the major flow goes further northeast and arrives at Nankai Trough. This flow was also shown in Reid's (1997) steric height map at 2000 dbar and 2500 dbar. This water mass may also incorporate hydrothermal plume of the Okinawa Trough at 1000–1250 m deep which run through the Kerama Gap. It is necessary to study the reason of additional 5% excess at about 2000~2500 m deep by intensive sampling and analysis of deep Pacific water in Off Shikoku and Kyushu Islands as well as deep Philippine Sea.

It is noted that the all 1000–2000 m deep water of Eastern East China Sea and Northwestern Philippine Sea show excess $^3$He of up to 25%. This suggests that the mid-depth of all over the western North Pacific water is probably affected by the mantle helium with a high $^3$He/$^4$He ratio. This phenomena was early expected by Lupton (1995) and significantly different from those of the Atlantic and Indian Ocean where excess $^3$He is relatively small (Jenkins *et al*., 1972; Jenkins and Clarke, 1976).

*Noble gas abundances*

Table 3 lists preliminary data of noble gas abundances in seawater at ST9 (Nansei Trench; Northwestern Philippine Sea) of the KT-02-14 cruise and ST1 (Off Joban; Northwestern Pacific Ocean) of the KT-01-12 cruise. Errors of abundances are about 2% at one sigma obtained by repeated measurements of standard air. Neon abundances vary from $1.43 \times 10^{-7}$ cm$^3$STP/g to $1.93 \times 10^{-7}$ cm$^3$STP/g. Generally deeper samples show larger concentrations, which is simply due to higher solubility with the lower temperature. Taking into account the temperature and salinity, we have calculated theoretical abundance of neon in these samples based on the solubility data in NaCl brine reported by Smith and Kennedy (1983) as follows:

$$\ln\beta = A_1 + A_2/Z + A_3 \ln Z - S[B_1 + B_2/Z + B_3 \ln Z]$$

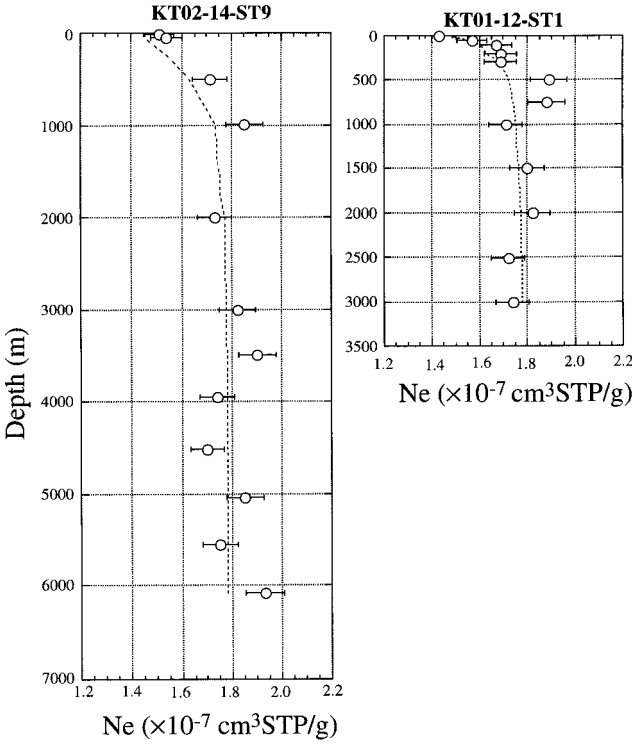


*Fig. 5. Neon abundance profiles of Northwestern Philippine Sea (KT02-14-ST9) and Northwestern Pacific Ocean (KT01-12-ST1) together with a curve of calculated solubility by the ambient temperature and salinity. Error assigned to the symbol is two sigma.*

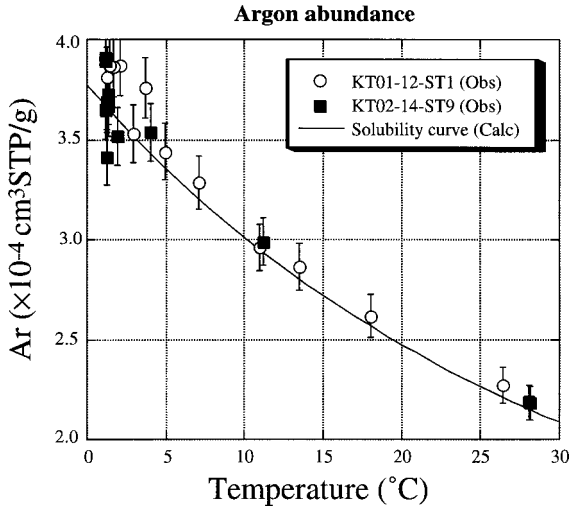


*Fig. 6. A correlation between temperature and corrected argon abundance of Northwestern Philippine Sea (KT02-14-ST9) and Northwestern Pacific Ocean (KT01-12-ST1) together with a curve of calculated solubility. Error assigned to the symbol is two sigma.*

where $Z$ equals $T/100$, $A_i$ and $B_i$ are constant, and $\beta$ and $S$ are bunsen coefficient and salinity, respectively. Figure 4 shows depth profiles of neon abundances at Nansei Trench and Off Joban together with the calculated values. Discrepancies of measured data from theoretical ones vary from –4.5% to +9.6%. There is no relationship between the depth and neon anomaly. Taking into account the experimental error, neon depletion of down to –4.5% may not be significant. On the other hand, neon excess up to +9.6% could not be explained by the error.

Craig and Weiss (1971) reported that discrepancy of noble gas abundance in seawater from the calculated solubility are due to following three effects; (1) Pressure effect: variation of atmospheric pressure and humidity make noble gas partial pressure in gas phase change, which would lead to the variation of noble gas abundance in water. (2) Temperature effect: rapid variation of atmospheric temperature may alter the solubility. For example seawater is in equilibrium condition with atmosphere at some temperature and then isolated from the air. If immediately after the isolation, water temperature changes significantly by some reason, noble gas abundance can not be explained by the ambient temperature. (3) Bubble effect: when the air bubble is entrapped in seawater at the surface, noble gas abundance can not show the equilibrium solubility value. In this case noble gas abundance ratios are constrained by the air composition. Based on the neon abundance data, Craig and Weiss (1971) suggested that the bubble effect is the most important factor in Pacific surface, and Atlantic surface and deep waters. Observed neon excess at Nansei Trench and Off Joban water would be attributable to the bubble effect. On the other hand, there is a possibility of air contamination by small bubbles in a copper tube when we collected samples. It is difficult to distinguish the bubble effect from the air contamination, since both components have atmospheric noble gas compositions. Therefore we claim these abundances as preliminary data.

The bubble effect and/or air contamination of heavier noble gases (argon, krypton and xenon) can be corrected using neon abundances as follows:

$$[\mathrm{NG}]_{\mathrm{cor}} = [\mathrm{NG}]_{\mathrm{obs}} - ([\mathrm{Ne}]_{\mathrm{obs}} - [\mathrm{Ne}]_{\mathrm{cal}}) \times A_{\mathrm{NG}}/A_{\mathrm{Ne}}$$

where $[\mathrm{NG}]_{\mathrm{cor}}$, $[\mathrm{NG}]_{\mathrm{obs}}$, $[\mathrm{Ne}]_{\mathrm{obs}}$ and $[\mathrm{Ne}]_{\mathrm{cal}}$ denote corrected and observed noble gas abundance, observed and calculated neon abundance, respectively. $A_{\mathrm{NG}}$ and $A_{\mathrm{Ne}}$ are concentrations of noble gas and neon in the air.

Observed argon abundances vary from $2.21 \times 10^{-4}$ to $3.99 \times 10^{-4}$ $\mathrm{cm^3STP/g}$, while corrected values range from $2.18 \times 10^{-4}$ to $3.91 \times 10^{-4}$ $\mathrm{cm^3STP/g}$. There is not a large difference between observed and corrected ones. Figure 5 shows a correlation diagram between the ambient temperature and corrected argon abundance together with the

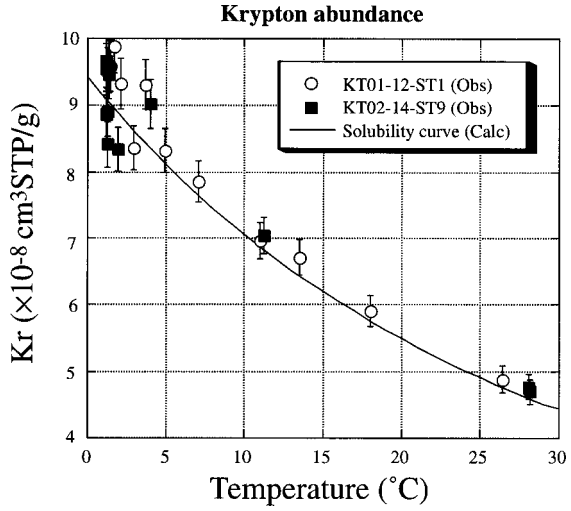


*Fig. 7. A correlation between temperature and corrected krypton abundance of Northwestern Philippine Sea (KT02-14-ST9) and Northwestern Pacific Ocean (KT01-12-ST1) together with a curve of calculated solubility. Error assigned to the symbol is two sigma.*

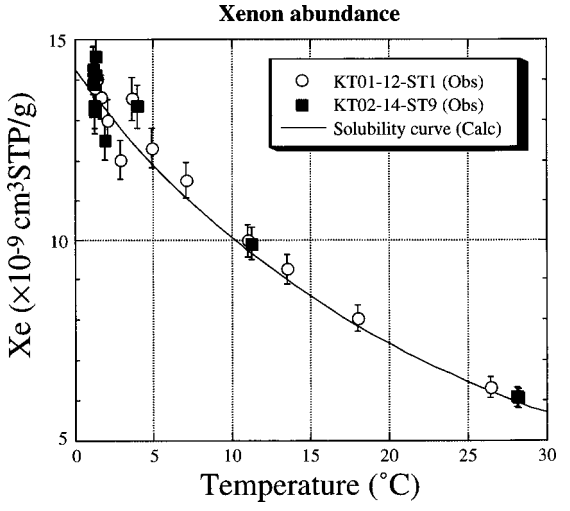


*Fig. 8. A correlation between temperature and corrected xenon abundance of Northwestern Philippine Sea (KT02-14-ST9) and Northwestern Pacific Ocean (KT01-12-ST1) together with a curve of calculated solubility. Error assigned to the symbol is two sigma.*

calculated solubility. Argon concentrations of samples with higher temperature than 5°C, which are coresponding to samples shallower than 500 m, agree well with or slightly above those of calculated ones. This implies the mixing between surface water and intermediate water which has isolated from the atmosphere. On the other hand, some of the samples lower than 5°C (deeper than 500 m) show significant argon anomaly from –6.9% to +8.1%. The negative anomaly may be due to either pressure effect or temperature effect as suggested by Craig and Weiss (1971), while the positive anomaly can only be attributable to pressure effect, since it is difficult for samples of 2~3°C to make a positive anomaly with further lower temperature. However the +8% higher atmospheric pressure may not occur so frequently. There is a possibility of experimental artifact in sample collection and measurement.

Figure 6 indicates a correlation diagram between the ambient temperature and corrected krypton abundance together with the calculated solubility. Krypton concentrations of samples with higher temperature than 5°C are consistent with or slightly above those of calculated ones, while some of those lower than 5°C do not agree with theoretical ones, which is similar to the trend of argon anomaly. The krypton discrepancies vary from –6.5% to +9.8%. Figure 7 shows a correlation diagram between the ambient temperature and corrected xenon abundance together with the calculated solubility. Again xenon concentrations of samples with higher than 5°C are consistent with or slightly above those of calculated ones, while some of those lower than 5°C do not agree with theoretical ones, where the xenon anomaly ranges from –6.7% to +7.9%. There are positive correlation between Ar and Kr anomaly with a correlation coefficient of 0.850 and that between Kr and Xe anomaly with the coefficient of 0.749. Therefore krypton and xenon anomalies should be attributable to the same way of argon. It is rather plausible to explain negative anomaly, which is about –6% depletion, by the temperature effect. On the other hand, we can not give a constraint on the mechanism of positive anomaly up to 10%. Further data of heavy noble gas abundance are required to explain the excess argon, krypton and xenon in future.

## SUMMARY

We have investigated helium isotopic ratios of twenty one Eastern East China Sea water and twenty two Northwestern Philippine Sea water collected in adjacent region of Nansei Islands, Southwest Japan. Mid-depth profiles of $^3He/^4He$ ratios at Eastern East China Sea may be attributable to the mantle helium derived from some hydrothermal activity in the Okinawa Trough. The $^3He/^4He$ profiles of Northwestern Philippine Sea agree well with that of Northern Philippine Sea (Nankai Trough) and is different from that of Northwestern Pacific Ocean (Off Joban), which may be due to a topographic barrier of Izu-Ogasawara Ridge. Noble gas abundances of twenty four samples in Northwestern Pacific Ocean and Northwestern Philippine Sea were measured. There is a small excess of neon concentrations, probably due to air bubble effect and/or air contamination. Even though the excess

is corrected using the neon content, heavier noble gas (argon, krypton and xenon) of some samples with temperature lower than 5°C show anomaly from –7% to +10%. Reason of these anomaly is not well understood.

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