Helium isotopes in South Pacific deep seawater

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We have measured 3He/4He and 4He/20Ne ratios and 4He concentrations in 19 South Pacific water samples with various depths from station SA-6 located in the Solomon Sea (6°38' S, 153°38' E) and SA-7 in the central Coral Sea Basin (14°16' S, 154°19' E) on the KH-92-4 cruise of the Research Vessel, Hakuho Maru of the University of Tokyo. The 3He/4He and 4He/20Ne ratios vary significantly from 0.988 R atm to 1.276 R atm and from 0.254 to 0.292, respectively. An extensive plume of water enriched in 3He has been discovered at a depth of about 2000 meter. Taking 3He/4He data of the Geosecs and the South Tow expeditions, 25% excess 3He plume can be traced over 5000 kilometers to the west of the East Pacific Rise. Observed 25% excess 3He plume at SA-6 and 7 may be independent from the ridge, which suggests that there is another source of the mantle derived helium in the area studied.

INTRODUCTION

Helium-3 is the most important tracer in many fields of Earth sciences such as volcanology (Kamensky et al., 1976; Sano et al., 1991), geothermal research (Torgersen and Jenkins, 1982; Sano and Wakita, 1988a), natural gas study (Wakita and Sano, 1983; Poreda et al., 1988), limnology (Clarke et al., 1977; Torgersen et al., 1977) and oceanography (Jenkins et al., 1972; Craig et al., 1975). This is based on the primordial signature, rapid mobility and chemical inertness of the isotope (Ozima and Podosek, 1983; Mamyrin and Tolstikhin, 1984). Taking oceanographical results, a striking intensity and lateral extent of excess 3He was discovered in the deep Pacific Ocean at latitude 15°S on the East Pacific Rise (Lupton and Craig, 1981). This plume shaped 3He anomaly was originated from the volcanic activity of the ridge and spreading westwards at the depth of the crest. Since then more than 1700 3He/4He measurements were carried out in the three main oceans (Ostlund et al., 1987). It is well documented that excess 3He in the Atlantic Ocean is smaller than in the Pacific.

In this work, we study the 3He/4He ratios of South Pacific water samples in the site of the Solomon Sea (SA-6) and the central Coral Sea Basin (SA-7) in order to verify the western end of 3He anomaly observed at 15°S on the East Pacific Rise. We also discuss other sources of the mantle helium than the mid-ocean ridge based on the 3He/4He-20Ne/4He relationship.

EXPERIMENTAL METHOD

Seawater samples were collected on the KH-92-4 cruise of the Research Vessel, Hakuho Maru of the University of Tokyo (Sep. 10–Oct. 27, 1992) in the South Pacific Ocean using Niskin hydrographic bottles, and transferred without exposure to atmosphere into 30 cm³ copper-tubing containers for storage (Sano et al., 1989a). The sampling sites are shown in Fig. 1 together with Geosecs station 251 and 263. At the laboratory the dissolved gases, including helium and neon, were extracted and purified in a stainless steel high vacuum line. The 3He/4He ratios were measured on a conventional noble gas mass spectrometer (VG5400, VG Isotopes). The observed 3He/4He...
ratios of samples were calibrated against atmospheric helium collected in August 1992 in Chiba, Japan. Helium was not separated from neon in this work, which may introduce a small error in the apparent $^3\text{He}/^4\text{He}$ ratio in the seawater samples (Rison and Craig, 1983; Sano and Wakita, 1988b). Sano and Wakita (1988b) reported the relation between the $^3\text{He}/^4\text{He}$ ratio of air standard and co-existing neon content. There was a negative correlation between the $^3\text{He}/^4\text{He}$ and $^{20}\text{Ne}/^{40}\text{He}$ ratios. Since the $^{20}\text{Ne}/^{40}\text{He}$ ratio of seawater is rather similar to that of air, the possible interference from neon to the helium isotope measurement may be compensated (Sano et al., 1993). In order to check the interference, we have measured air saturated distilled water at $20^\circ\text{C}$ and surface sea water samples collected in the South Pacific Ocean.

After $^3\text{He}/^4\text{He}$ measurements, $^4\text{He}/^{20}\text{Ne}$ ratios were analyzed using the same mass spectrometer by adjusting the magnet current. At the same time the $^{20}\text{Ne}/^{22}\text{Ne}$ ratios were measured in order to check interference from doubly charged ion of $^{40}\text{Ar}$ with $^{20}\text{Ne}$. The error of $^4\text{He}/^{20}\text{Ne}$ ratios were estimated to be about 5% based on the reproducibility of standard air measurements. The concentration of $^4\text{He}$ was calculated from the beam intensity of $^4\text{He}$ by a Faraday cup. The content measurement has about 10% error (Sano et al., 1993).

RESULTS AND DISCUSSION

Observed $^4\text{He}$ contents and $^3\text{He}/^4\text{He}$ ratios of three distilled water are listed in Table 1 together
He-isotopes in deep seawater with reference data. The $^{3}$He/$^{4}$He ratios are shown in $R_{\text{atm}}$ unit where $R_{\text{atm}}$ is atmospheric ratio of $1.39 \times 10^{-6}$. Discrepancy of the $^{3}$He/$^{4}$He ratio from the reference is less than 0.35% for distilled water, when we take into account the average value. The precision for the $^{3}$He/$^{4}$He measurements, based on the reproducibility of air standard is about 0.7% (1σ) during that time. Thus the discrepancy is smaller than the precision of measurements and ambiguity due to the interference of neon to helium isotope measurement is negligibly small. The small interference is partly due to the “tuning” of the mass spectrometer (Lupton and Craig, 1987) and is also due to similarity of helium/neon ratios between air and seawater samples (Sano and Wakita, 1988b), which may compensate the interference. Lupton and Craig (1987) reported that the helium-neon interference is less than 1% of observed $^{3}$He/$^{4}$He ratio based on the comparison between helium isotope analysis with Ne and without Ne of 90 selected Atlantic and Pacific seawater sample. Their results are consistent with the present work. Since the determination of $^{3}$He/$^{4}$He ratio for the seawater samples includes measurement errors for both the sample itself and the air standard, we take a total precision of 1% (1 sigma) for the $^{3}$He/$^{4}$He ratio measurement.

Observed $^{4}$He contents, and $^{3}$He/$^{4}$He and $^{4}$He/$^{20}$Ne ratios of 19 South Pacific water samples are listed in Table 1. Errors of $^{4}$He/$^{20}$Ne ratio and $^{4}$He abundance are 5% and 10% at 1 sigma, respectively (Sano et al., 1993). The $^{4}$He concentrations vary from $3.78 \times 10^{-8}$ ccSTP/g to $4.38 \times 10^{-8}$ ccSTP/g and are significantly smaller than that of air saturated water at 20°C ($4.6 \times 10^{-8}$ ccSTP/g). This difference is well explained by the salting out effect where helium is some 20% less soluble in sea water than in fresh water (Weiss, 1971; Ozima and Podosek, 1983). The $^{4}$He contents are consistent with those of Geosecs expedition (Ostlund et al., 1987). The $^{3}$He/$^{4}$He and $^{4}$He/$^{20}$Ne ratios are ranging from 0.988 $R_{\text{atm}}$ to 1.276 $R_{\text{atm}}$ and from 0.254 to 0.292, respectively. The $^{3}$He/$^{4}$He ratios of surface sea water samples agree well with those of literature (Ostlund et al., 1987). There is a slight positive correlation between the $^{3}$He/$^{4}$He and $^{4}$He/$^{20}$Ne ratios except for the surface water. This suggests two component mixing between air or air saturated water and mantle components with high $^{3}$He/$^{4}$He ratio, which will be discussed more precisely in the following section.

$^{3}$He/$^{4}$He ratios of South Pacific deep seawater

Figure 2 shows excess $^{3}$He depth profiles at the site SA-6 and SA-7 in South Pacific. There is an apparent excess $^{3}$He of more than 25% at the depth of about 2000 m of sites SA-6 and SA-7, which is comparable with those observed in the South Tow expedition (Lupton and Craig, 1981). A small

<table>
<thead>
<tr>
<th>Depth (m)</th>
<th>$^{4}$He (x 10^{-8} ccSTP/g)</th>
<th>$^{3}$He/$^{4}$He (R_{\text{atm}})</th>
<th>$^{4}$He/$^{20}$Ne (R_{\text{atm}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>4.72</td>
<td>0.9743</td>
<td>0.286</td>
</tr>
<tr>
<td>No. 2</td>
<td>4.85</td>
<td>0.9924</td>
<td>0.270</td>
</tr>
<tr>
<td>No. 3</td>
<td>4.94</td>
<td>0.9865</td>
<td>0.254</td>
</tr>
<tr>
<td>Average</td>
<td>4.84</td>
<td>0.9844</td>
<td>0.266</td>
</tr>
<tr>
<td>Reference</td>
<td>4.6 (a)</td>
<td>0.9837 (b)</td>
<td>0.270</td>
</tr>
<tr>
<td>Station SA-6 (6^\circ 38 ' S, 153^\circ 38 ' E)</td>
<td></td>
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<tr>
<td>10</td>
<td>3.92</td>
<td>0.988</td>
<td>0.292</td>
</tr>
<tr>
<td>384</td>
<td>3.84</td>
<td>1.062</td>
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<tr>
<td>571</td>
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<td>1.041</td>
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<tr>
<td>952</td>
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<td>1.223</td>
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<tr>
<td>1744</td>
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<td>5224</td>
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<tr>
<td>Station SA-7 (14^\circ 16 ' S, 154^\circ 19 ' E)</td>
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<tr>
<td>10</td>
<td>3.78</td>
<td>0.996</td>
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<tr>
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<tr>
<td>4072</td>
<td>3.97</td>
<td>1.127</td>
<td>0.262</td>
</tr>
</tbody>
</table>

(a) From Ozima and Podosek (1983).
(b) From Benson and Krause (1980).
(c) Error of $^{4}$He abundance is 10% at 1 sigma.
(d) Error of $^{3}$He/$^{4}$He ratio is 1% at 1 sigma.
(e) Error of $^{4}$He/$^{20}$Ne ratio is 5% at 1 sigma.
peak of excess $^3$He observed at the depth of 384 m of SA-6 may be due to the decay of tritium produced by nuclear experiments (Jenkins et al., 1972).

It is well documented by the South Tow expedition (Lupton and Craig, 1981) that South Pacific deep seawater has an extensive plume of water with excess $^3$He due to an injection of mantle derived helium at the spreading center axis on the East Pacific Rise. The $^3$He plume can be traced over 2000 km to the west. Figure 3 shows contour map of $\delta(^3$He) in section view over the East Pacific Rise at 15°S by the South Tow expedition (Lupton and Craig, 1981) and its extension to the west based on the present data and sites 251 and 263 of the Geosecs Pacific expedition (Ostlund et al., 1987). Geosecs Pacific Expedition data revealed that the $^3$He plume is ob-

Fig. 2. Excess $^3$He profiles at sites SA-7 and SA-6 in the South Pacific Ocean. The $^3$He/$^4$He ratios of sites SA-7 and SA-6 are calibrated against atmospheric helium collected in Chiba, Japan in August 1992. Bars indicate experimental error margin (1σ).

Fig. 3. Contours of $\delta(^3$He) in section view over the East Pacific Rise at 15°S (Lupton and Craig, 1981) and their western extension based on the present and Geosecs data (Ostlund et al., 1987). Note that the 25% excess $^3$He plume derived from the East Pacific Rise may be intermitted at a place between the site 251 and 263.
served at the site 5000 km to the west of the ridge where maximum excess $^3$He is found approximately 24% at the depth of about 2400 m (Ostlund et al., 1987). It is noted that the $^3$He residence time is about 4000 years in deep ocean (Lupton and Craig, 1981) and deep seawater has been isolated from the present atmosphere for at least 1500 years based on the carbon-14 measurement (Ostlund et al., 1987). Since the vertical movement of helium in the ocean is significantly smaller than that of horizontal way by the eddy diffusion, the $^3$He plume may be preserved in some 5000 km distant place from the spreading center axis on the East Pacific Rise. Thus $^3$He is a really good tracer in physical oceanography.

There are more than 25% $\delta(^3\text{He})$ of seawater samples in the present study. These samples were collected at the depth between 1500 m and 2300 m. As shown in Fig. 3, there is no 25% $\delta(^3\text{He})$ at the site 251 of the Geosecs expedition (Ostlund et al., 1987). Therefore the 25% excess $^3$He plume derived from the East Pacific Rise may be intermitted at a place between the site 251 and 263. This may be explained by a topographic high by New Herbrides and Solomon Islands (Fig. 3) and by a fairly strong boundary current which flows just east of New Zealand (Wooster and Volkman, 1960). The 27.6% excess observed at the station SA-7 may be attributable to other source than the mid ocean ridge, even though the discrepancy of the present and Geosecs data may be due to experimental artifact such as difference of standard air helium for the calibration (Sano et al., 1989b). However this is not the case since data of surface seawater samples agree well with those of Ostlund et al. (1987).

There are many Quaternary volcanoes close to the station SA-6 related to the New Britain arc-trench system (Both et al., 1986). Some of them are submarine volcanoes in the New Ireland Basin and hydrothermal activity is observed in a seismically active area south of the Lihir Island (Herzig et al., 1994). In addition significant hydrothermal activities were discovered in several places such as the Manus Basin (Both et al., 1986; Gamo et al., 1993), the North Fiji Basin (Ishibashi et al., 1994) and the Woodlark Basin (Binns et al., 1993; Trull et al., 1990). The $\delta(^3\text{He})$ values over 25% observed in this work may be derived from these local magmatic activities and the identification of the source will be discussed in the later section based on the $^{3\text{He}}/^{4\text{He}}$ ratios.

Source of mantle helium in South Pacific deep seawater

In order to estimate the local source of mantle helium, we have studied the relationship between $^{3\text{He}}/^{4\text{He}}$ and $^{20\text{Ne}}/^{4\text{He}}$ ratios (Fig. 4). There is a negative correlation between the $^{3\text{He}}/^{4\text{He}}$ and $^{20\text{Ne}}/^{4\text{He}}$ ratios of the deep seawater samples with a correlation coefficient of $-0.734$. Surface seawater samples do not match the trend because they are well explained by air saturated seawater at the temperature of higher than 25°C. This temperature may be related with the ambient temperature of atmosphere at the sampling site. In addition the 384 m sample of SA-6 is not taken into account because it may be affected by the decay of tritium. Least-squares fitting yields a straight line described by:

$$^{20\text{Ne}}/^{4\text{He}} = (4.70 \pm 0.24) - (0.589 \pm 0.146) \times ^{3\text{He}}/^{4\text{He}} \times 10^6$$

where errors assigned to the values are one sigma. The $^{3\text{He}}/^{4\text{He}}$ and $^{20\text{Ne}}/^{4\text{He}}$ ratios are given in the absolute ratios. The trend is well explained by a mixing between the air saturated seawater at about 0°C and mantle helium with the $^{3\text{He}}/^{4\text{He}}$ ratio of $5.74 \pm 1.45 \text{R atm}$ (at the $^{20\text{Ne}}/^{4\text{He}}$ ratio = 0). The estimated $^{3\text{He}}/^{4\text{He}}$ ratio is somehow smaller than those of $8 \pm 1 \text{R atm}$ of mid-ocean ridge basalt glasses (Lupton, 1983), but agrees well with those of $6-8 \text{R atm}$ of island arc volcanic gases (Sano and Wakita, 1985; Poreda and Craig, 1989). Therefore the source of $\delta(^3\text{He})$ values over 25% may be due to subduction-type helium, even though we can not exclude experimental artifact because the difference between the present and Geosecs data is small.

As is shown in the former section, there are four potential sources of mantle helium in the re-
Fig. 4. Correlation diagram between $^{3}$He/$^{4}$He and $^{20}$Ne/$^{4}$He ratios of the South Pacific seawater samples. Bars indicate experimental error margin (1σ). Air saturated seawater at 0°C and 25°C are also shown.

CONCLUSIONS

An extensive plume of water enriched in $^{3}$He has been discovered at a depth of about 2000 meter at the station SA-6 (the Solomon Sea) and SA-7 (the central Coral Sea Basin). Taking $^{3}$He/$^{4}$He data of the Geosecs and the South Tow expeditions,
20% excess $^3$He plume can be traced over 9000 kilometers to the west of the East Pacific Rise at latitude 15°S. Based on the negative correlation between $^3$He/$^4$He and $^{20}$Ne/$^4$He ratios, we have estimated the local source of mantle helium with the $^3$He/$^4$He ratio of $(5.74 \pm 1.45) \text{ Ratm}$, which is consistent with subduction-type helium signature and may be due to the hydrothermal activity of the Woodlark Basin including submarine volcanoes and the spreading center.

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REFERENCES


