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# Solar Wind Noble Gases in Micrometeorites

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## 1. Introduction

Most extraterrestrial materials discovered on the Earth have no solar wind noble gases. In fact, only four types of extraterrestrial materials contain noble gases attributed to the solar wind or its fractionated component: gas-rich meteorites, lunar materials collected by the Apollo missions, asteroid samples returned from Itokawa by the Hayabusa mission, and micrometeorites. Except for micrometeorites, all of these have a specific history of solar wind irradiation on the surface of their parent bodies. On the other hand, solar wind noble gases in micrometeorites are implanted during orbital evolution in interplanetary space. Micrometeorites have a different origin and irradiation history from the other three materials and from typical meteorites, meaning that these tiny particles that fell on the Earth can provide us valuable information about the activity of the solar system. Of all the analytical methods in planetary science, noble gas analysis of extraterrestrial materials is one of the most useful, because the analysis can reveal not only their origin and age but also their history of irradiation by galactic and solar cosmic rays and solar wind. In particular, the most reliable positive proof of an extraterrestrial origin for micrometeorites is the solar wind noble gases. In this chapter, solar wind noble gases trapped in micrometeorites are reviewed.

### 1.1 Nomenclature of extraterrestrial dust

First, the nomenclature of extraterrestrial dust must be explained because the peculiar technical terms in the field of planetary science are perplexing for researchers belonging to different scientific fields. The main terms for extraterrestrial dust are *micrometeorite*, *interplanetary dust particle (IDP)*, *cosmic spherule*, and *cosmic dust*. *Micrometeorite* can indicate all types of extraterrestrial dust collected on the Earth, but is mainly used to indicate extraterrestrial dust collected in polar regions. *IDPs* are very small dust particles (<30  $\mu\text{m}$  in diameter) collected in the stratosphere by airplane and are often called *stratospheric dust particles* or *Brownlee particles*. *Cosmic spherules* are small spherical particles recovered from deep-sea sediment, polar regions, and sedimentary rocks. Their spherical shape is due to severe heating during atmospheric entry. Tiny spherical particles found in sedimentary rocks are generally called *microspherules*, *microkrystite*, or *microtektites*. *Unmelted micrometeorites* indicates micrometeorites other than the cosmic spherules, whose shape is irregular. *Cosmic dust* indicates all types of extraterrestrial dust, including intergalactic dust, interstellar dust, interplanetary dust, and circumplanetary dust. *Extraterrestrial dust* is

another versatile term synonymous with *cosmic dust*, but it is not as widely used as *cosmic dust*.

*Micrometeorite* is thought to be the best term representing extraterrestrial dust in this chapter for a few reasons. First, the cosmic dusts with solar wind noble gases reviewed here are not intergalactic dusts or interstellar dusts. Second, the Antarctic micrometeorites that are the main target of this paper are not IDPs. Therefore, the word *micrometeorite* adequately represents all types of cosmic dust that contain solar wind noble gases.

## 1.2 Collection of micrometeorites

It was already suspected in the Middle Ages that a large number of dusty objects existed in interplanetary space. Zodiacal light is a faint glow that extends away from the sun in the ecliptic plane of the sky, visible to the naked eye in the western sky shortly after sunset or in the eastern sky shortly before sunrise. Already in 1683, Giovanni Domenico Cassini presented the correct explanation of this prominent light phenomenon visible to the human eye. Its spectrum indicates it to be sunlight scattered by interplanetary dust orbiting the sun. It is called “counter-glow” or “Gegenschein” in German (Yamakoshi, 1994). The zodiacal light contributes about a third of the total light in the sky on a moonless night. The sky is, however, seldom dark enough for the entire band of zodiacal light to be seen. Micrometeorites in interplanetary space, contributors to the zodiacal light, are constantly produced by asteroid collisions and liberated from the sublimating icy surfaces of comets. Since the radiation pressure of the sun is sufficient to blow submicron grains (beta meteoroids) out of the solar system, only larger grains (20–200  $\mu\text{m}$ ) contribute to the zodiacal light. Poynting-Robertson drag causes larger grains to depart from Keplerian orbits and to spiral slowly toward the sun.

Micrometeorites are the main contributors of extraterrestrial material accreted on the Earth. The accretion rate of cosmic dust particles has been estimated by various means so far, and the values calculated in those reports are different. There is, however, no difference in the conclusion that micrometeorites are the primary extraterrestrial deposit on Earth. Published reports estimating the accretion rate of extraterrestrial matter are well summarized in an appendix table of Peucker-Ehrenbrink (1996). For example, Love and Brownlee (1993) determined the mass flux and size distribution of micrometeoroids in the critical submillimeter size range by measuring hypervelocity impact craters found on the space-facing end of the gravity-gradient-stabilized Long Duration Exposure Facility (LDEF) satellite. A small-particle mass accretion rate of  $40,000 \pm 20,000$  tons/yr was obtained. In another estimate, a Japanese micrometeorite research group carefully picked up Antarctic micrometeorites and accurately counted their numbers, yielding accretion rates of 5,600–10,400 tons/yr (Yada et al., 2001).

Although such a large amount of micrometeorites is continuously supplied to the Earth, micrometeorites have been collected in places where extraterrestrial particles are concentrated and/or terrestrial dust is rare, such as the deep sea, the stratosphere, and polar regions. It is very difficult to discover micrometeorites in inhabitable areas that are contaminated by artificial and terrestrial dusts. Since E. Nishibori collected micrometeorites in Antarctica in 1957–1958 (Nishibori and Ishizaki, 1959), a large number of micrometeorites have been recovered from the Antarctic and Greenland ice sheets and northern Canada

(Theil and Schmidt, 1961; Shima and Yabuki, 1968; Maurette et al., 1986, 1987, 1991; Koeberl and Hagen, 1989; Cresswell and Herd, 1992; Taylor et al., 1997, 1998; Nakamura et al., 1999; Yada and Kojima, 2000; Iwata and Imae, 2002; Rochette et al., 2008; Carole et al., 2011). Antarctic micrometeorites (AMMs) have larger sizes (50–300  $\mu\text{m}$ ) than the IDPs captured in the stratosphere (<30  $\mu\text{m}$ ). Since most of the mass accreted by the Earth is contained in larger particles (50–400  $\mu\text{m}$ ) (Kortenkamp and Dermott, 1998), AMMs represent the interplanetary dust population well.

## 2. Solar wind noble gases in deep-sea sediment

Isotopic noble gas study on micrometeorites was difficult for a long time because of terrestrial contamination and the small sizes of micrometeorites. Measurements on single cosmic particles had to wait for great improvement of analytical devices. Therefore, the first noble gas study on micrometeorites was a measurement on deep-sea sediments in which micrometeorites were concentrated. The first noble gas isotopic study on deep-sea sediments was performed by Merrihue (1964). Magnetic and nonmagnetic separates of modern red clays from the Pacific Ocean were analyzed using a glass extraction and purification system, and excess  $^3\text{He}$  and  $^{21}\text{Ne}$  were discovered. The reported  $^3\text{He}/^4\text{He}$  ratios (shown as  $^4\text{He}/^3\text{He}$  in Merrihue's paper) are clearly higher than that of the terrestrial atmosphere, and a relatively high  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio ( $11.0 \pm 1.0$ ) is reported in the 1000°C step of the magnetic separate.  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios lower than that of the atmosphere in the 1000°C and 1400°C steps of the magnetic separate ( $268 \pm 7$  and  $172 \pm 8$ ) were clearly detected, indicating the presence of extraterrestrial materials. This excellent research for the first time presented overwhelming evidence that extraterrestrial materials with extraterrestrial noble gases had accumulated in the deep-sea sediments. Nine years later, Krylov et al. (1973) reported He isotopic compositions of fifteen oceanic oozes recovered from various regions of the Pacific and Atlantic oceans and the iceberg-melting region of Greenland, which were analyzed by researchers in the Soviet Union. The isotopic ratios for Pacific red clays are tens or a hundred times that found in the various crustal rocks. On the other hand, Atlantic red clays have low  $^3\text{He}/^4\text{He}$  ratios of  $2\text{--}3 \times 10^{-6}$  and no  $^3\text{He}$  anomaly was found in the Greenland samples. They believed that the likely source for the elevated  $^3\text{He}$  content in the Pacific Ocean sediments is cosmic rather than the hypothetical  $^3\text{He}$  from the mantle in the clays. The idea was confirmed by studying nitric-acid-treated ooze, which had the same order of  $^3\text{He}/^4\text{He}$  ratios as untreated ooze. Indeed, the high  $^3\text{He}/^4\text{He}$  ratios found in the red clays should be attributed to micrometeorites.

After these two reports, research in the field stagnated for a long time, and these important researches were forgotten completely. Japanese researchers, however, renewed study in the field in the 1980s. Ozima et al. (1984) measured thirty-nine sediments from twelve different sites, ten sites from the western to central Pacific and two sites from the Atlantic Ocean. They found  $^3\text{He}/^4\text{He}$  ratios higher than  $5 \times 10^{-5}$  for six sites and concluded that the very high  $^3\text{He}/^4\text{He}$  ratios in the sediments reflected the input of extraterrestrial materials. Amari and Ozima (1985) subsequently reported a He anomaly in deep-sea sediments, and they rediscovered that the carrier of exotic He was concentrated in magnetic fractions, which was consistent with the result of Merrihue's analysis. Since most terrestrial particles are nonmagnetic, magnetic cosmic dusts are concentrated in magnetic separates. They concluded that the ferromagnetic separates are essentially magnetite using thermomagnetic

analyses. They also performed a stepwise degassing experiment, which suggested that He is trapped fairly tightly. Amari and Ozima (1988) analyzed magnetic fractions separated from four deep-sea sediments from the Pacific Ocean. Notably, the study presented Ne and Ar isotopic compositions of the sediments. In all the samples, the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios were constant ( $11.6 \pm 0.6$ ) in most temperature steps. This result should now be interpreted as being caused by a mixing of solar wind (SW) and implantation-fractionated solar wind (IFSW) components, although they concluded that the Ne was from a unique component.  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios lower than that of the atmosphere, 296, were evidently detected in high-temperature fractions of all samples, indicating the existence of extraterrestrial Ar. They concluded from the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios and thermal release patterns of He that the extraterrestrial noble gases are implanted solar flare particles.

Fukumoto et al. (1986) determined elemental abundances and isotopic compositions of noble gases in separates and acid-leached residues of deep-sea sediments collected on a cruise of R/V Hakureimaru, Geological Survey of Japan. A  $^3\text{He}/^4\text{He}$  ratio of  $(2.73 \pm 0.06) \times 10^{-4}$  was detected for the magnetic separate B2M. Nitric acid treatment did not affect the isotopic ratio, and the  $^3\text{He}/^4\text{He}$  ratio of the leached sample B2M-1 is  $(2.74 \pm 0.08) \times 10^{-4}$ , suggesting that the acid did not attack the carrier of the high  $^3\text{He}/^4\text{He}$  ratio. Ne isotopic compositions show that the extraterrestrial materials in the sediments were affected by SW component rather than cosmic-ray spallation. Extraterrestrial Ar was detected in the acid-leached residue B1-3, whose  $^{40}\text{Ar}/^{36}\text{Ar}$  was  $194.3 \pm 52.2$ . Matsuda et al. (1990) carried out stepwise extraction analyses for the magnetic separate and 3M-HCl-leached residues of the same sample used by Fukumoto et al. (1986). Extraterrestrial He and Ne were observed in most temperature steps of all samples. The magnetic separate lost about 75% of its  $^3\text{He}$  without a drastic change in its isotopic ratios when it was dissolved in 3M HCl at room temperature for two days, and a sample more severely etched for six days had similar elemental and isotopic compositions of He and Ne to those of the two-day-etched sample, indicating that the extraterrestrial He and Ne should be concentrated in fine particles and/or on the surface of the magnetic grains. These studies performed by Japanese institutes clarified that extraterrestrial materials with solar-derived He and Ne are concentrated in deep-sea sediments and that the most plausible candidate for the carrier of the extraterrestrial noble gas is micrometeorites accreted on the Earth.

Reported  $^3\text{He}/^4\text{He}$  ratios are summarized in Fig. 1. There are some differences in the isotopic ratios among the reports, and the ratio gradually increased with the year of the study, with the exception of the data from Merrihue (1964), reflecting the improvement in sample separation. Very high  $^3\text{He}/^4\text{He}$  ratios were consistently detected in the study by Matsuda et al. (1990) because of their use of magnetic separation (they analyzed only 0.53% by weight of the dry sediment) and acid leaching. Such physical and chemical separations concentrated the extraterrestrial materials that exist in deep-sea sediment. The  $^3\text{He}/^4\text{He}$  ratios reported by Merrihue (1964) are clearly too high, and the true values should be lower than the reported ratios. Ne isotopic compositions are also summarized in Fig. 2. The plots are distributed between the values of the SW and IFSW, indicating that the extraterrestrial materials in deep-sea sediments do not have chondritic noble gas compositions and that cosmogenic Ne is not dominant. The remarkably low  $^{21}\text{Ne}/^{22}\text{Ne}$  ratios detected in the magnetic separates from deep-sea sediments are clearly consistent with the isotopic compositions of individual micrometeorites.

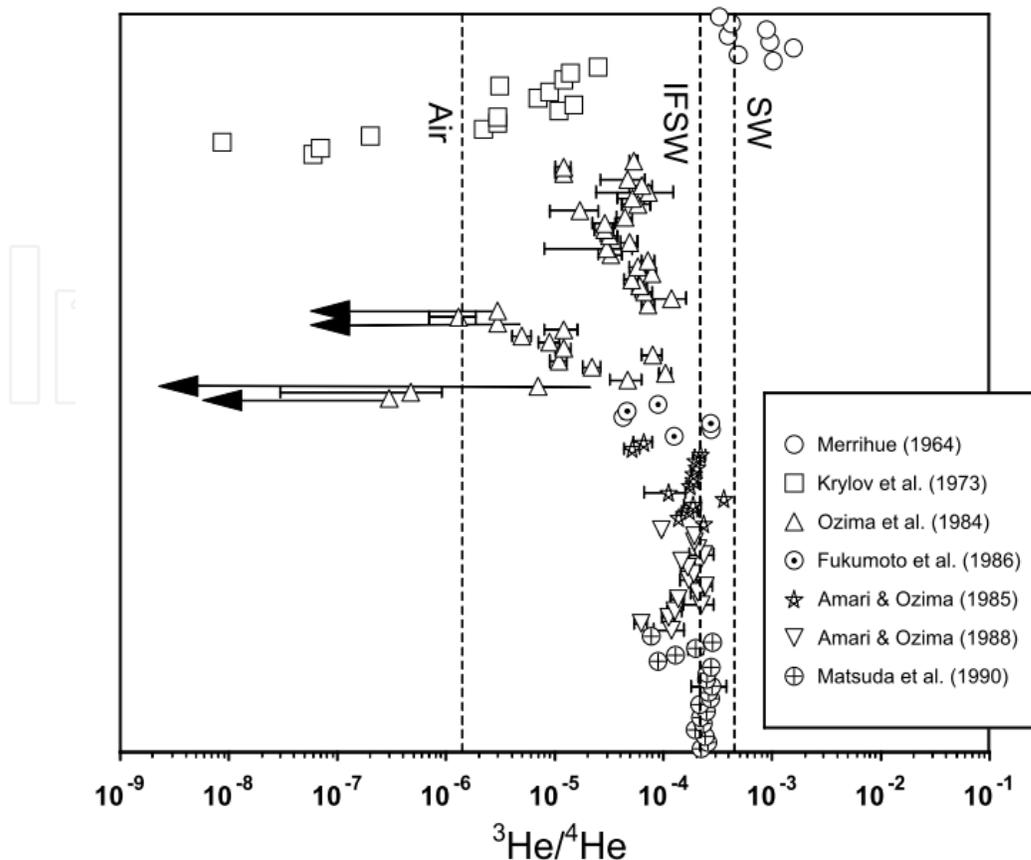


Fig. 1. Reported  $^3\text{He}/^4\text{He}$  ratios of deep-sea sediments. Dotted lines show the isotopic ratios of the terrestrial atmosphere at  $1.4 \times 10^{-6}$ , implantation-fractionated solar wind (IFSW) at  $2.17 \times 10^{-4}$  (Benkert et al., 1993), and solar wind (SW) at  $4.53 \times 10^{-4}$  (Heber et al., 2008).

### 3. Solar wind noble gases detected in individual unmelted micrometeorites

Since noble gas isotope analysis for a single micrometeorite is very difficult because of the extremely small amount of noble gases in a particle, a mass spectrometer with high sensitivity and low background is required to determine accurate isotopic ratios of noble gases released from individual micrometeorites. The first attempt to measure single micrometeorites from deep Pacific Ocean sediments was made by Nier et al. (1987, 1990). They measured He and Ne in deep Pacific particles collected directly from the ocean floor with a 300 kg towed magnetic sled. The samples used were bulk magnetic fines that passed through a  $100 \mu\text{m}$  sieve (they called them "deep Pacific magnetic fines") and individual particles larger than  $100 \mu\text{m}$  in diameter. The individual particles were irregular, and their elemental composition, mineralogy, and texture were consistent with those of meteoritic materials. They measured thirty-five magnetic fines and six individual particles and suggested the possibility that there could be several types of extraterrestrial particles present in the magnetic fines. The most significant result in the paper was the extremely high He isotopic ratios observed in the  $1600^\circ\text{C}$  steps of the magnetic fines and individual particles. They attributed the exotic noble gas compositions to solar flare particles.

IDPs collected from the stratosphere have provided valuable information on extraterrestrial noble gases trapped in cosmic dust particles. The first report concerning noble gas

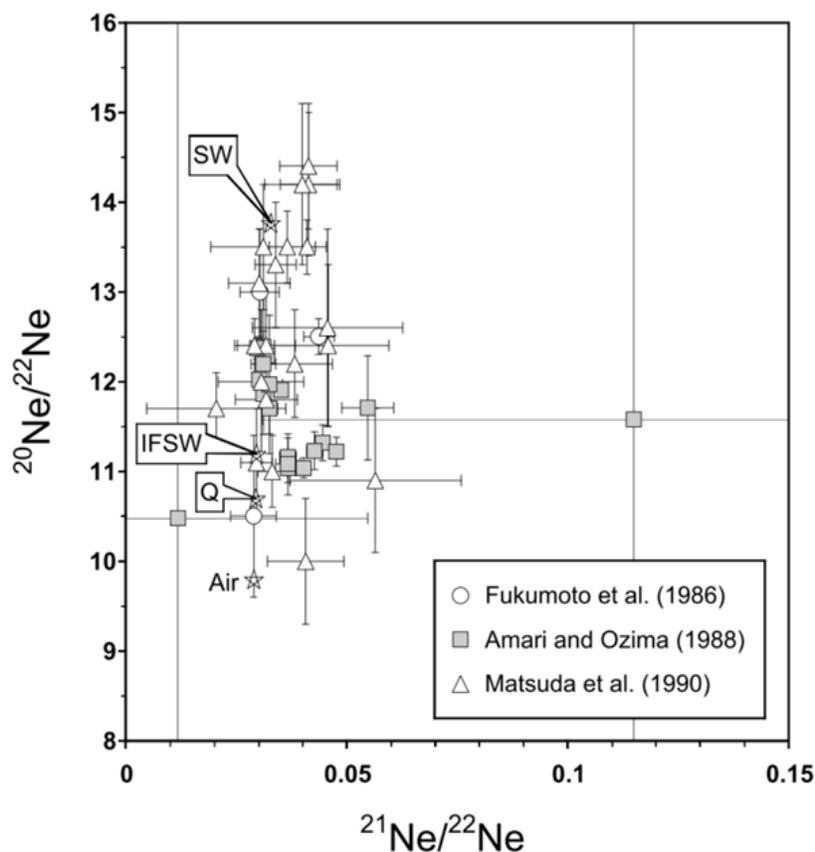


Fig. 2. Three-isotope plot of Ne for deep-sea sediments. SW and IFSW data are from Heber et al. (2008) and Benkert et al. (1993), respectively.

compositions of IDPs is that by Rajan et al. (1977). They detected very high concentrations of  $^4\text{He}$  ranging from 0.002 to 0.25  $\text{cm}^3$  STP/g in ten stratospheric particles collected by NASA U-2 aircraft and asserted that the particles were extraterrestrial and that some or all of them were exposed to solar wind for at least 10–100 years. Hudson et al. (1981) selected thirteen chondritic stratospheric particles and measured Ne, Ar, Kr, and Xe by stepwise heating at 1400°C, 1500°C, and 1600°C. The  $^{20}\text{Ne}/^{36}\text{Ar}$  ratio in the particles is  $9 \pm 3$ , indicating the presence of solar-type light noble gas. On the other hand, the  $^{132}\text{Xe}$  concentration of  $\sim 10^{-7}$   $\text{cm}^3$  STP/g and the heavy noble gas elemental pattern suggested a substantial contribution from planetary sources. This is the only report on Kr and Xe in extraterrestrial dusts before Osawa et al. (2000).

The first noble gas measurement for individual IDPs was performed by Nier and Schlutter (1989). They measured He and Ne isotopic compositions for sixteen individual stratospheric particles. The samples were wrapped in a small piece of previously degassed Ta foil, and noble gases were extracted by heating, which was accomplished by passing an electric current directly through the foil. Except for one sample, the IDPs had  $^3\text{He}/^4\text{He}$  ratios of  $1.5\text{--}4.3 \times 10^{-4}$ . The average of the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio was  $12.0 \pm 0.5$ . In the next stage, they performed stepwise heating for fragments from twenty individual particles to clarify the origin of the particles using the release pattern of  $^4\text{He}$  (Nier and Schlutter, 1992). Twelve of the IDP fragments contained an appreciable amount of  $^4\text{He}$ , 50% of which was released by the time the particles were heated to approximately 630°C. Four IDP fragments contained appreciably less  $^4\text{He}$ , and this was released at a higher temperature. The remaining four

fragments had too little  $^4\text{He}$  to permit a determination. This result suggested that the parent IDPs of the twelve particles that contained an appreciable amount of  $^4\text{He}$  suffered very little heating in their descent and are likely of asteroidal origin, although one cannot rule out the possibility that at least some of them had a cometary origin and entered the Earth's atmosphere at a grazing angle. Nier and Schlutter later performed pulse-heating sequences for twenty-four individual IDPs to learn about the thermal history of the particles and distinguish between IDPs of asteroidal and cometary origin. In this investigation, fifteen of twenty-four particles had  $^3\text{He}/^4\text{He}$  ratios above  $10^{-3}$ , and the highest value,  $2 \times 10^{-2}$ , was found in L2011 D7. They had no explanation for this anomaly.

Kehm et al. (1998a) performed combined trace element and light noble gas measurements on fourteen IDPs from the L2036 stratospheric collector using a laser gas-extraction system and a synchrotron X-ray microprobe. The Ne isotopic compositions in these IDPs were dominated by implanted solar components including SW and IFSW Ne. The Ar isotopic compositions of six large IDPs ( $>25 \mu\text{m}$  in their longest dimension) demonstrated enrichment in solar components. Low  $^4\text{He}$  contents were observed in five particles that exhibited Zn depletion, indicating severe heating and volatile loss during atmospheric entry. Kehm et al. (1998b) later performed trace element and noble gas measurements on ten large IDPs ( $\sim 20 \mu\text{m}$ ). They suggested preferential He loss during atmospheric entry heating in this study. Kehm et al. (1999) performed noble gas measurements on JJ-91 IDPs and presented major differences between the result of their measurements and the data of Nier and Schlutter (1993). Kehm et al. (1999) did not detect an anomalously high  $^3\text{He}/^4\text{He}$  ratio in a fragment of 2011 cluster 11, in which a very high  $^3\text{He}/^4\text{He}$  ratio was detected by Nier and Schlutter (1993). However, the reasons for the differences were not clear. Kehm et al. (2002) measured noble gases in 32 individual IDPs, and the  $^4\text{He}$ ,  $^{20}\text{Ne}$ , and  $^{36}\text{Ar}$  contents were determined for 31 IDPs. The noble gas elemental compositions were consistent with the presence of fractionated solar wind, but the isotopic compositions were unknown.

Ne isotopic compositions of individual unmelted micrometeorites collected from seasonal lakes on the Greenland ice sheet were reported by Olinger et al. (1990). The extraterrestrial origin of the particles was confirmed by the isotopic data. Maurette et al. (1991) reported Ne isotopic compositions of unmelted and partially melted micrometeorites recovered from Antarctic blue ice. Stuart et al. (1999) measured He isotopes in forty-five putative micrometeorites in the size range of  $50\text{--}400 \mu\text{m}$  recovered from Antarctic ice. They determined the He isotopic compositions of twenty-six particles. Pepin et al. (2000, 2001) reported He, Ne, and Ar isotopic ratios for many IDPs and discussed the extremely high  $^3\text{He}$  concentration found in some large cluster particles by Nier and Schlutter (1993). They proposed several possibilities to explain the overabundance of  $^3\text{He}$ . The noble gas research group at the University of Tokyo reported isotopic compositions of noble gases including Ar, Kr, and Xe for individual unmelted AMMs using a highly established mass spectrometer with a laser gas extraction system (Osawa and Nagao, 2002a, 2002b; Osawa et al., 2000, 2001, 2003). These studies clarified that many micrometeorites contain not only extraterrestrial He and Ne but also extraterrestrial Ar. It is, however, very difficult to detect extraterrestrial Kr and Xe because the concentrations of heavy noble gases are extremely low and the effect of adsorbed terrestrial atmosphere cannot be ignored. Osawa and Nagao (2003) and Osawa et al. (2010) reported noble gas compositions of individual cosmic spherules recovered from Antarctica, and about 40% of the cosmic spherules preserved extraterrestrial noble gases, although their noble gas concentrations were very low due to severe heating.

### 3.1 He isotopic ratios of micrometeorites

Compiled He isotope data for unmelted AMMs and IDPs are depicted in Fig. 3. The data on IDPs with strikingly high  $^3\text{He}/^4\text{He}$  ratios reported by Nier and Schlutter (1993) are excluded here. The  $^3\text{He}/^4\text{He}$  ratios in the AMMs and IDPs are plotted against the concentrations of  $^4\text{He}$  in this figure. The range of  $^4\text{He}$  concentrations extends from  $10^{-6}$  to  $10^2 \text{ cm}^3 \text{ STP/g}$ , which may reflect the degree of entry heating for each AMM and IDP. The  $^3\text{He}/^4\text{He}$  ratios of most AMMs are distributed between those of SW and IFSW value, showing the presence of SW He, but there is no significant correlation between the isotopic ratios and  $^4\text{He}$  concentration. Since the SW noble gas is thought to become saturated in the surface layer of a small particle in interplanetary space within about a few decades (e.g., Hudson et al., 1981), solar-wind-derived He is implanted in the surface of AMMs and IDPs. It is, however, notable that the isotopic ratios are not clustered around the SW value, and more than half of the particles have  $^3\text{He}/^4\text{He}$  ratios lower than that of SW. This is due to isotopic fractionation during solar wind ion implantation and the loss of the surface layer of the particles during atmospheric entry. The surface layers of the micrometeorites were preferentially heated and ablated by flash heating (e.g., Love and Brownlee, 1991). However, the SW He in the micrometeorites had not been completely extracted by the heating, and the remaining solar-wind-derived He proves the extraterrestrial origin of the AMMs and IDPs.

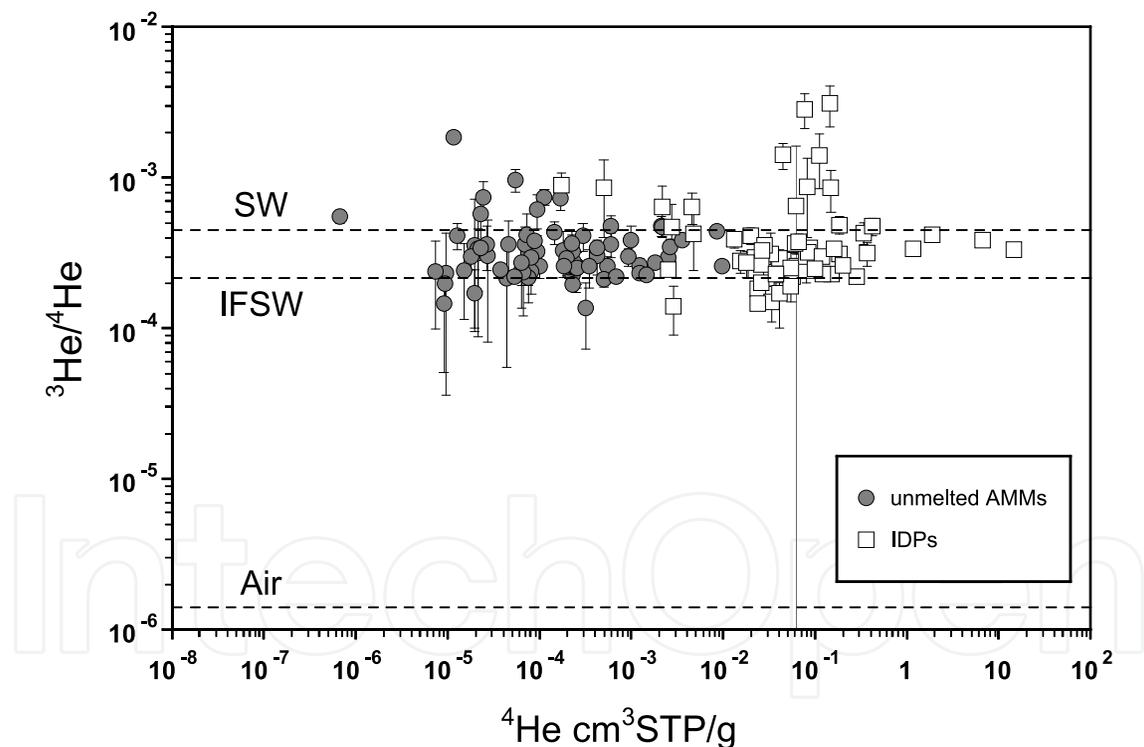


Fig. 3.  $^4\text{He}$  concentration and  $^3\text{He}/^4\text{He}$  ratio of unmelted AMMs and IDPs. IDP data are from Nier and Schlutter (1990, 1992) and Pepin et al. (2000, 2011). Unmelted AMM data are from Stuart et al. (1999), Osawa and Nagao (2002b), and Osawa et al. (2003).

The very large difference in  $^4\text{He}$  concentration between AMMs and IDPs is remarkable; IDPs have a much higher concentration of  $^4\text{He}$  than do AMMs, but the  $^3\text{He}/^4\text{He}$  ratio of most IDPs falls in a similar range to that of AMMs. The large difference in  $^4\text{He}$  concentration is mainly caused by the size range;  $^4\text{He}$  concentrations in cosmic dust particles correlate with

their grain sizes (Stuart et al., 1999). IDPs are smaller than AMMs and have a higher surface area/volume ratio than do AMMs. Since the mechanism of accumulation of SW noble gases in micrometeorites is ion implantation, the concentration of SW noble gases depends on surface area. A high surface area/volume ratio thus causes a high noble gas concentration. A secondary reason for the high He concentration of IDPs is the lower heating temperature; IDPs can escape severe heating because of their low weight and density. He loss in AMMs occurs in response to the thermal decomposition of phyllosilicates and diffusive loss and bubble rupture during atmospheric entry, rather than melting (Stuart et al., 1999). Aqueous alteration in the Antarctic snow can be another possible cause of He loss in AMMs. For example, jarosite  $[\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6]$ , a by-product mineral resulting from aqueous alteration of sulfide minerals, is observed in ~43% of the AMMs collected from 30,000 year old glacial ice (Terada et al., 2001), and these AMMs have lower He concentrations than AMMs collected from fresh snow, indicating He loss due to aqueous alteration (Osawa and Nagao, 2002). Osawa et al. (2003) reported that jarosite-bearing AMMs have relatively low concentrations of  $^4\text{He}$ , suggesting loss of He during long-term storage in ice. However, since jarosite is not often found in AMMs, aqueous alteration in ice is not the main cause of the low He concentration of AMMs.

Although the He isotopic ratios of most AMMs and IDPs simply reflect solar-derived He, it is not possible to completely deny the contributions of other components such as planetary He and cosmogenic  $^3\text{He}$ , an additional component found in some IDPs. In addition, isotopic fractionation during entry deceleration heating should be taken into consideration. Some AMMs and IDPs have higher  $^3\text{He}/^4\text{He}$  ratios than that of SW. These probably reflect cosmogenic  $^3\text{He}$  because the  $^3\text{He}/^4\text{He}$  ratio of cosmogenic He is very high, about 0.2. Since cosmogenic  $^3\text{He}$  is more strongly retained in a micrometeorite than SW He, which exists mostly in the surface layer because of the low energy of solar wind, the  $^3\text{He}/^4\text{He}$  ratio is elevated by the preferential loss of solar-wind-derived He. If cosmogenic  $^3\text{He}$  does not exist in the AMMs, the  $^3\text{He}/^4\text{He}$  ratio will approach the ratio of IFSW after the loss of the surface layer of the micrometeorites (Grimberg et al., 2008). The cosmogenic  $^3\text{He}$  concentrations of some unmelted AMMs with relatively high  $^3\text{He}/^4\text{He}$  ratios are much lower than those of IDPs with high concentrations of cosmogenic  $^3\text{He}$  of over  $5 \times 10^{-6} \text{ cm}^3 \text{ STP/g}$  (Pepin et al., 2001). Strikingly high  $^3\text{He}/^4\text{He}$  ratios, possibly due to some unknown reservoir, were reported for some IDPs (Nier and Schlutter, 1993; Pepin et al., 2000). For example, the IDP L2011D7 has a low  $^4\text{He}$  content ( $3.4 \times 10^{-12} \text{ cm}^3 \text{ STP}$ ) and an unusually high  $^3\text{He}/^4\text{He}$  ratio ( $(2.0 \pm 0.3) \times 10^{-2}$ ; Nier and Schlutter, 1993). Kehm et al. (1999), however, did not detect such anomalously high  $^3\text{He}/^4\text{He}$  ratios in individual IDP grains separated from the same cluster IDP L2011. In their measurement, nine of eleven IDPs had high He content ( $0.7\text{--}7 \times 10^{-10} \text{ cm}^3 \text{ STP}$ ) and low  $^3\text{He}/^4\text{He}$  ratios; the He compositions correspond to those of typical IDPs shown in Fig. 3. The high  $^3\text{He}/^4\text{He}$  ratios found in the enigmatic IDPs are thus very problematic. If the large overabundance of  $^3\text{He}$  is to be attributed to cosmogenic  $^3\text{He}$ , extremely long periods of cosmic-ray irradiation time are required (Pepin et al., 2001). It is noted that the lack of Ne isotopic data obstructs the interpretation of the problem of excess  $^3\text{He}$  in IDPs. Even if the enigmatic IDPs are excluded in this discussion, the excess  $^3\text{He}$  concentrations of AMMs are clearly low compared to those of IDPs. Since the low concentration of cosmogenic  $^3\text{He}$  presumably indicates preferential loss of He due to severe entry heating,  $^3\text{He}$  exposure ages of AMMs are not reliable, in contrast to those of IDPs.

The geometric average of  $^3\text{He}/^4\text{He}$  ratios of AMMs,  $3.10 \times 10^{-4}$ , is slightly lower than that of IDPs,  $3.55 \times 10^{-4}$ , which may also reflect the difference in the degree of surface loss or heating during atmospheric entry. This result is consistent with the large difference in He concentration between the two micrometeorite series. Note that a geometric average is more suitable for evaluating the representative He isotopic ratio of micrometeorite samples than an arithmetic mean because the distributions of  $^3\text{He}/^4\text{He}$  ratios of AMMs and IDPs are evidently not normal distributions. In conclusion, unmelted AMMs and IDPs preserve extraterrestrial He derived from energetic implantation of solar wind, but the effects of gas loss and fractionation cannot be ignored. SW He trapped in micrometeorites found on the Earth does not, therefore, represent pure solar wind.

It is extremely difficult to detect solar wind noble gases in the totally melted cosmic spherules because most volatiles have been depleted by harsh heating during atmospheric entry. It is, however, surprising that extraterrestrial He, Ne, and Ar still remain in some cosmic spherules (Osawa and Nagao, 2003; Osawa et al., 2010). Fig. 4 shows the  $^4\text{He}$  contents and the  $^3\text{He}/^4\text{He}$  ratios of unmelted AMMs and cosmic spherules. Since only 29 of 130 spherules preserved detectable amounts of  $^3\text{He}$ , the  $^4\text{He}$  contents of the spherules presented in Fig. 4 do not reflect the distribution of the noble gas contents of all spherules. Even the  $^4\text{He}$  contents of the gas-rich cosmic spherules shown in the figure are much lower than those of unmelted AMMs. All of the gas-rich cosmic spherules have  $^3\text{He}/^4\text{He}$  ratios higher than that of terrestrial air within one sigma error, proving their extraterrestrial origin. Furthermore, many spherules have He isotopic ratios close to that of SW, as do the unmelted micrometeorites, indicating that the spherules have preserved solar-derived He in

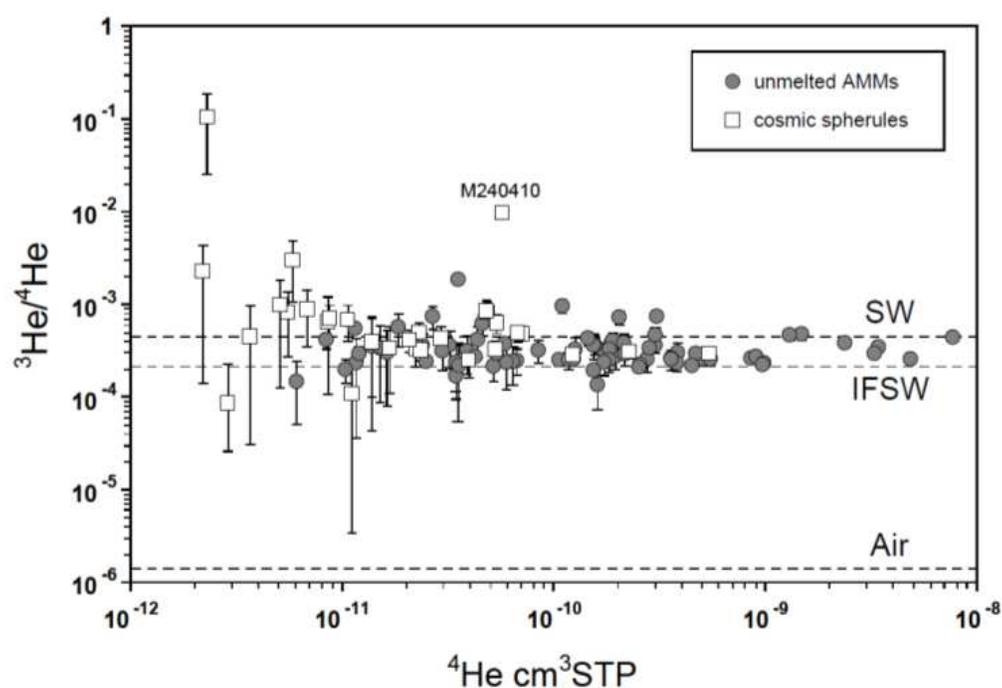


Fig. 4. Relationship between  $^4\text{He}$  content and  $^3\text{He}/^4\text{He}$  ratio of unmelted AMMs and cosmic spherules. Unmelted AMM data are from Stuart et al. (1999), Osawa and Nagao (2002b), and Osawa et al. (2003). Cosmic spherule data are from Osawa and Nagao (2003) and Osawa et al. (2010).

spite of their severe heating. This result implies that the spherules are small particles in interplanetary space and not fragments of meteorites fallen to the Earth, as solar-gas-rich meteorites are quite rare. Osawa et al. (2010) discovered an exotic cosmic spherule, M240410, which has an extraordinarily high  $^3\text{He}/^4\text{He}$  ratio  $(9.7 \pm 1.1) \times 10^{-3}$  and high  $^3\text{He}$  content  $(5.53 \times 10^{-13} \text{ cm}^3\text{STP})$  that resulted from cosmogenic production of  $^3\text{He}$ . Such high isotopic ratios have not been found in unmelted micrometeorites, indicating that this specific spherule may have an exceptional history. The highest  $^3\text{He}/^4\text{He}$  ratio reported to date in an unmelted micrometeorite is  $(1.843 \pm 0.050) \times 10^{-3}$  (Stuart et al., 1999), which is much lower than that of M240410.

### 3.2 Ne isotopic ratios of micrometeorites

Ne isotope data on micrometeorites can provide information on solar wind, fractionated solar wind, and cosmogenic nuclides. These three components can be separated using a diagram because Ne has three stable isotopes in contrast with He, which has only two. The Ne isotopic composition is thus useful for separating SW components from cosmogenic nuclides, but the Ne concentration of micrometeorites is much lower than the He concentration.

Fig. 5 displays Ne isotopic compositions of unmelted micrometeorites, IDPs, and cosmic spherules. It is remarkable that most micrometeorite data are clustered around the IFSW value and show no cosmogenic  $^{21}\text{Ne}$  within the error limit, indicating short exposure ages. Several micrometeorites have  $^{21}\text{Ne}/^{22}\text{Ne}$  ratios higher than that of SW; for example, two exceptional Dome Fuji AMMs have long cosmic-ray exposure (CRE) ages ( $>100$  Myr). However, most micrometeorites have exposure ages shorter than 1 Myr (Osawa and Nagao, 2002a). An enigmatic cosmic spherule, M240410, has an extremely high concentration of cosmogenic  $^{21}\text{Ne}$  and was calculated to have a very long CRE age of 393 Myr when  $4\pi$  exposure to galactic and solar cosmic rays was taken into consideration, indicating that the source of the particle may have been an Edgeworth-Kuiper belt object (Osawa et al., 2010). The Ne isotopic compositions of several unmelted micrometeorites are close to, or above, the SW  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of 13.77 (Heber et al., 2008). These are Greenland micrometeorite compositions reported by Olinger et al. (1990), and the high  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios are due to the overestimation of  $\text{CO}_2^{++}$  interference. Hence, the SW-like Ne compositions detected in some micrometeorites do not indicate the presence of unfractionated solar wind, and the solar-derived Ne in all types of micrometeorites is partially depleted and fractionated.

The effect of partial loss of Ne can be observed in a trend in the  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio. The average  $^{20}\text{Ne}/^{22}\text{Ne}$  ratios of IDPs, unmelted micrometeorites, and cosmic spherules are 11.92, 11.39, and 10.57, respectively; the difference in the isotopic ratios among the three micrometeorite groups may reflect the degree of atmospheric entry heating. The smaller IDPs ( $\sim 20 \mu\text{m}$ ) experienced lower entry temperatures compared to the larger micrometeorites ( $\sim 100 \mu\text{m}$ ) because the maximum temperature during the trajectory depends on particle radius (e.g., Rizk et al., 1991). The average  $^{20}\text{Ne}/^{22}\text{Ne}$  ratio of cosmic spherules is lower than the IFSW ratio, 11.3, reflecting contamination by the terrestrial atmosphere. Although noble gases in cosmic spherules are considerably depleted by severe flash heating, some spherules preserved solar-wind-derived He and Ne, suggesting that the cosmic spherules have been exposed to solar wind and/or solar flares before atmospheric entry and that they are not simple atmospheric entry ablation fragments of meteorites.

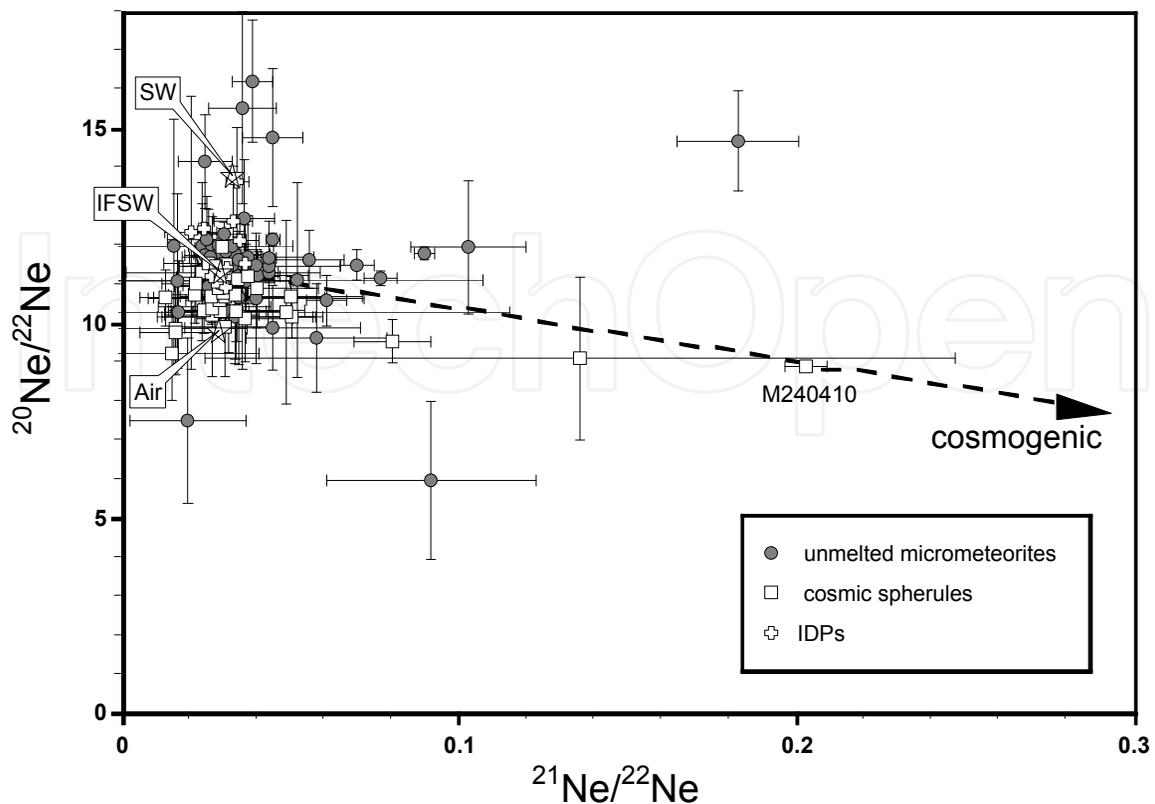


Fig. 5. Three-isotope plot of Ne for unmelted AMMs, cosmic spherules, and IDPs. Unmelted AMM data are from Olinger et al. (1990), Osawa and Nagao (2002b), and Osawa et al. (2003). Cosmic spherule data are from Osawa and Nagao (2003) and Osawa et al. (2010). IDP data are from Pepin et al. (2000). An arrow shows the direction of cosmogenic Ne.

### 3.3 Ar isotopic ratios of micrometeorites

Ar isotopic compositions of individual micrometeorites were reported only by two groups, at Washington University and the University of Tokyo (Kehm et al., 1998a; Osawa and Nagao, 2002a, 2002b, 2003; Osawa et al., 2000, 2001, 2003, 2010). Merrihue (1964) reported a low  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio ( $172 \pm 5$  in the  $1400^\circ\text{C}$  fraction) in a magnetic separate of Pacific red clay and suggested that it contains meteoritic material, but that the data do not correspond to those of a single micrometeorite. Since Ar has three stable isotopes, as does Ne, the Ar isotopic compositions of micrometeorites can clarify the contributions of more than two components. A three-isotope plot of Ar for unmelted AMMs and cosmic spherules is presented in Fig. 6. IDP data from Kehm et al. (1998a) are not plotted in this diagram because of the lack of raw data. All unmelted micrometeorites with detectable amounts of Ar have  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios lower than that of the terrestrial atmosphere, 296, confirming their classification as extraterrestrial because terrestrial materials with  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios lower than that of terrestrial air are very few. Although the Ar isotopic compositions of cosmic spherules have large uncertainties due to the very low Ar concentrations, the  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios of many spherules are lower than the atmospheric value. This indicates that extraterrestrial Ar is detectable for these samples because significant gas loss and terrestrial contamination do not overwhelm the extraterrestrial Ar completely.

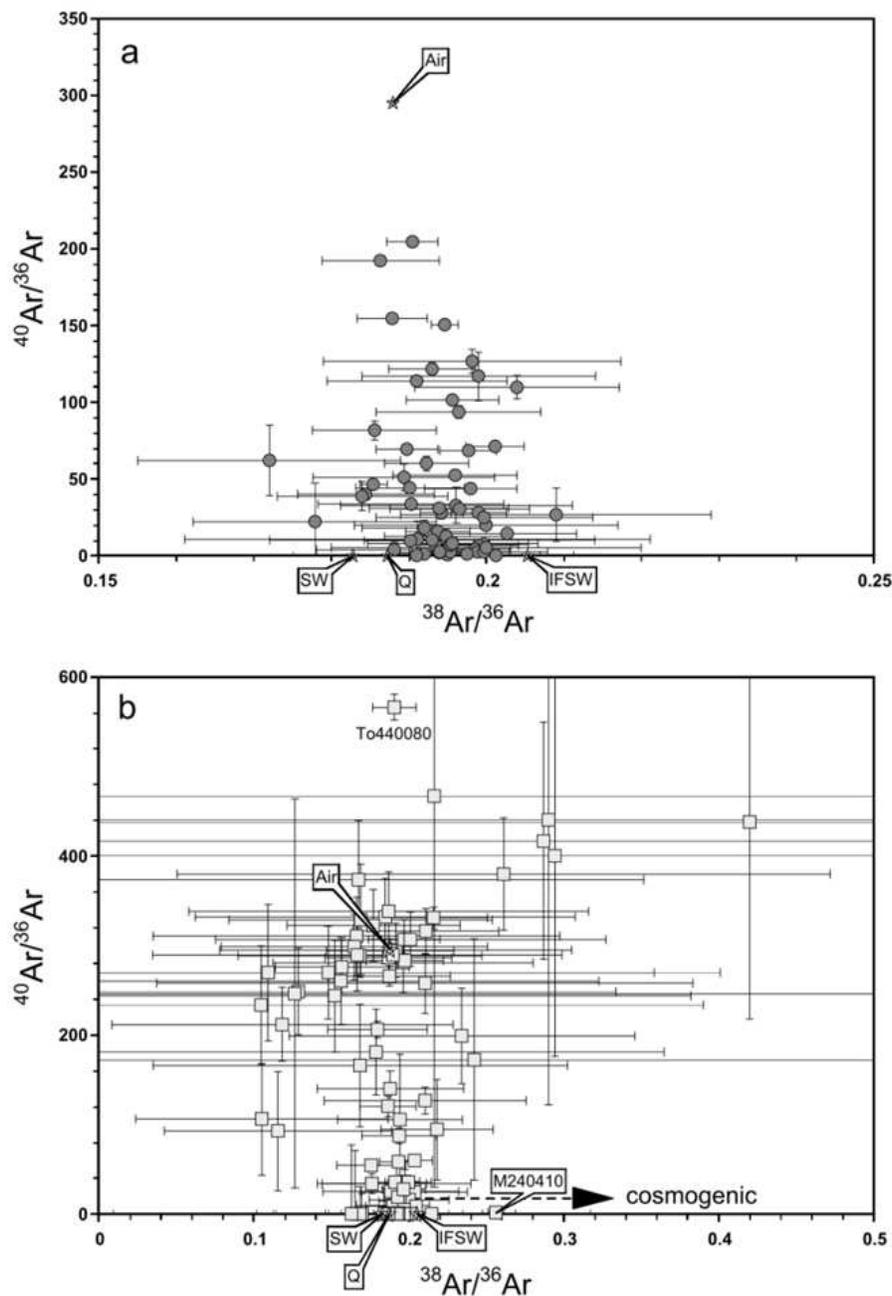


Fig. 6. Ar isotopic compositions of (a) unmelted AMMs and (b) cosmic spherules. Unmelted AMM data are from Osawa and Nagao (2002b) and Osawa et al. (2003). Cosmic spherule data are from Osawa and Nagao (2003) and Osawa et al. (2010).

Cosmic-ray-produced spallogenic  $^{38}\text{Ar}$  was detected only in spherule M240410, which has a detectable amount of cosmogenic  $^3\text{He}$  and  $^{21}\text{Ne}$ . The concentration of cosmogenic  $^{38}\text{Ar}$  of the spherule is  $4.8 \times 10^{-8} \text{ cm}^3 \text{ STP/g}$ , and the CRE age calculated with  $2\pi$  irradiation is 382.1 Myr (Osawa et al., 2010). All micrometeorites other than this exceptional spherule have no cosmogenic  $^{38}\text{Ar}$ , even the unmelted AMMs with relatively high  $^{21}\text{Ne}/^{22}\text{Ne}$  ratios, presumably due to the lower rate of cosmic-ray production of  $^{38}\text{Ar}$  than that of  $^{21}\text{Ne}$  (Eugster, 1988).

The Ar isotopic composition of unmelted AMMs is composed of three components: terrestrial atmosphere, IFSW, and a component of primordial trapped Ar, such as the Q component (Osawa and Nagao, 2002b). Q-noble gas is the main component of heavy noble gases in primitive chondrites hosted by the phase Q, which is an oxidizable phase of a residue of treatment with hydrochloric acid and hydrofluoric acid (e.g., Lewis et al. 1975; Ott et al., 1981; Huss et al., 1996).  $^{38}\text{Ar}/^{36}\text{Ar}$  ratios that are relatively high compared to the SW value are observed in unmelted AMMs, and the average  $^{38}\text{Ar}/^{36}\text{Ar}$  ratio of 0.193 is higher than the Q-Ar value of 0.187 (Busemann et al., 2000). This indicates the presence of IFSW Ar, in agreement with the IFSW-like Ne composition shown in Fig. 5. The contribution of unfractionated SW component is small, and fractionated absorbed air need not be considered. In contrast with the cases for He and Ne, the contribution of the primordial trapped Ar component is detectable.

About 40% of cosmic spherules and most unmelted AMMs preserved detectable amounts of extraterrestrial Ar but were affected by atmospheric contamination; most  $^{40}\text{Ar}$  in the micrometeorites was dominantly derived from the terrestrial atmosphere. It is not obvious that there exists radiogenic  $^{40}\text{Ar}$  produced in situ because  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios higher than those of the Q or solar components can be explained by atmospheric contamination (Osawa and Nagao, 2002b), and the concentrations of potassium in AMMs are low (Nakamura et al., 1999; Kurat et al., 1994). The enigmatic spherule To440080, however, has an exceptionally high  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio ( $566.3 \pm 14.8$ ), in spite of the presence of IFSW-like Ne. The high isotopic ratio is clearly due to radiogenic  $^{40}\text{Ar}$ . This spherule has a high  $^{36}\text{Ar}$  concentration ( $6.5 \times 10^{-7} \text{ cm}^3 \text{ STP/g}$ ) in spite of its high  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio, although meteorites with such high  $^{36}\text{Ar}$  concentrations generally have lower  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios than this spherule. An IFSW  $^{36}\text{Ar}$  contribution of approximately 50% is calculated from the concentration of  $^{20}\text{Ne}$ , if a  $^{20}\text{Ne}/^{36}\text{Ar}$  ratio of 47 is adopted as the IFSW ratio (Murer et al., 1997). If this estimation is correct, the original  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio of this spherule was over 1000, and this spherule undoubtedly originated in a different type of the parent body than did the other micrometeorites.

The contributions of the three Ar components (air, Q, and IFSW) in unmelted AMMs can be estimated using a simple mixing model. In this estimation, all of the  $^{40}\text{Ar}$  is assumed to be atmospheric because the  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios of the IFSW and Q components are inaccurate but assumed to be very low. Atmospheric  $^{36}\text{Ar}$  and  $^{38}\text{Ar}$  are thus probably overestimated, but they contribute only 5% and 4% of the total Ar, respectively. The contribution of the Q component is comparable to that of IFSW component, and the average contributions of  $^{36}\text{Ar}$  and  $^{38}\text{Ar}$  of the Q component are found to be 45% and 47% of the total Ar, respectively (Osawa et al., 2002). Since  $^{38}\text{Ar}/^{36}\text{Ar}$  ratios of cosmic spherules have large uncertainties, as shown in Fig. 6(b), it is difficult to differentiate the contribution of IFSW Ar from that of primordial trapped Ar in individual spherules. The contribution from the Q component may be comparable with that from IFSW component, as it is in the case of unmelted AMMs, since there is no sign that the original noble gas compositions of the cosmic spherules (other than To440080 and M240410) are different from those of the unmelted AMMs. In conclusion, the low  $^{40}\text{Ar}/^{36}\text{Ar}$  ratios of micrometeorites are not only due to solar wind irradiation.

### 3.4 Kr and Xe in micrometeorites

Since Kr and Xe concentrations of single micrometeorites are extremely low, their isotopic compositions cannot be determined accurately, and Kr and Xe isotopic ratios of micrometeorites typically have uncertainties larger than 20% (Osawa and Nagao, 2002b).

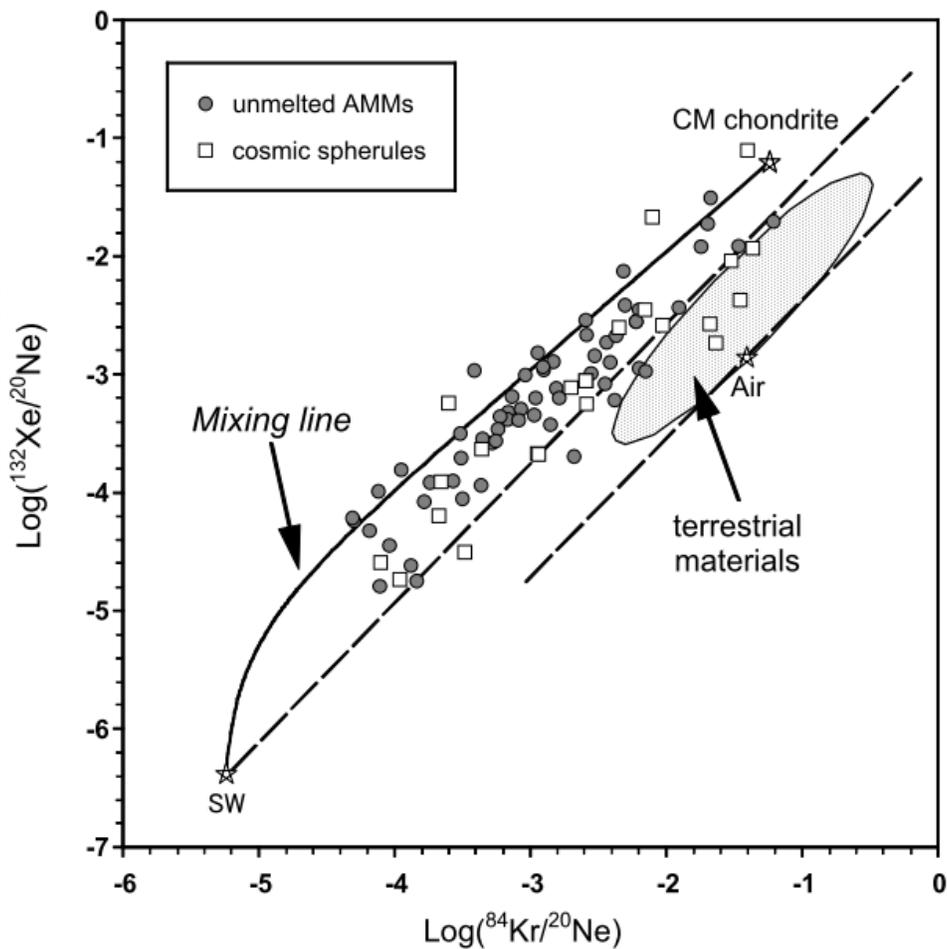


Fig. 7.  $^{84}\text{Kr}/^{20}\text{Ne}$  ratios versus  $^{132}\text{Xe}/^{20}\text{Ne}$  ratios on logarithmic scale. Dotted lines show theoretical fractionation lines of terrestrial air and SW component established by mass-dependent Rayleigh distillation. A solid line shows a mixing of SW and CM chondrite compositions. Air is from Ozima and Podosek (2002). SW data is represented by the 71501 low-temperature regime in Becker et al. (1989). CM2 chondrite is represented by Belgica-7904 (Nagao et al., 1984). Unmelted AMM data are from Osawa and Nagao (2002b) and Osawa et al. (2003). Cosmic spherule data are from Osawa and Nagao (2003) and Osawa et al. (2010).

In addition, Kr and Xe have no large isotopic anomalies, in contrast with the cases for light noble gases. Indeed the mean values of the Kr and Xe isotopic ratios of micrometeorites are identical within the error to the atmospheric values (Osawa et al., 2000; Osawa and Nagao, 2002a, 2002b). Although micrometeorites may preserve solar-derived Kr and Xe, the isotopic compositions of Kr and Xe are useless to identify the solar component. Even the rocky grains of the asteroid Itokawa recovered by the Hayabusa spacecraft have no Kr and Xe attributable to solar wind, although terrestrial contamination of the samples is very low (Nagao et al., 2011).

The noble gas elemental composition including  $^{84}\text{Kr}$  and  $^{132}\text{Xe}$  is, however, useful for identifying the sources of heavy noble gases. The relative abundances of  $^{20}\text{Ne}$ ,  $^{84}\text{Kr}$ , and  $^{132}\text{Xe}$  are depicted in Fig. 7 on a logarithmic scale. All terrestrial materials are distributed below the theoretical mass fractionation line of SW noble gases because the abundance of

terrestrial Xe is low, having been selectively depleted by unknown causes (the so-called “missing Xe”). Extraterrestrial materials can thus be distinguished using the diagram. Most of the unmelted AMM data points do not overlap the area representing terrestrial materials, indicating an extraterrestrial origin of the unmelted AMMs. Most of the unmelted AMMs are distributed above the mass fractionation line of SW noble gases. On the other hand, a few cosmic spherules are plotted in the area representing terrestrial materials, indicating contamination by terrestrial atmosphere.

The solid line shows mixing between SW and the primordial trapped component represented by the noble gas composition of a CM2 chondrite, Belgica-7904 (Nagao et al., 1984)). The noble gas composition of Belgica-7904 mainly reflects the Q component for Kr and Xe and the HL component for Ne. HL gas is a primitive component trapped in presolar diamonds. SW data is substituted for IFSW data in the diagram, under the assumption that there is no difference between IFSW and SW value since the noble gas elemental abundance of IFSW component is unclear. Most unmelted AMMs are distributed between the SW-CM2 chondrite mixing line and the mass fractionation line of SW noble gases. The figure clearly shows that both the primordial trapped component and the SW component are preserved in the micrometeorites. The noble gas compositions of the micrometeorites are thus explained by mixing of three components: a primordial trapped component, SW, and terrestrial contamination. The contribution of each component can be roughly estimated using the simple mixing model. If unfractionated air is assumed in the calculation, the average contributions of atmospheric  $^{84}\text{Kr}$  and  $^{132}\text{Xe}$  are 1.5% and 2% of the total Kr and Xe, respectively. These values are, however, not accurate because air adsorbed on the surface of micrometeorites should be fractionated and its noble gas elemental ratios cannot be determined accurately (Osawa and Nagao, 2002b). If the elemental compositions of adsorption-fractionated air are arbitrarily set to be  $^{84}\text{Kr}/^{20}\text{Ne} = 0.1$  and  $^{132}\text{Xe}/^{20}\text{Ne} = 0.0043$ , the mean contribution of the fractionated air is only 0.6% of the total  $^{84}\text{Kr}$  and  $^{132}\text{Xe}$ . 99% of  $^{132}\text{Xe}$  and 95% of  $^{84}\text{Kr}$  in micrometeorites is due to the primordial trapped component, and the contribution of SW component for Kr and Xe is very low (Osawa et al., 2003). This estimation implies that it is almost impossible to identify the SW Kr and Xe from the isotopic compositions of Kr and Xe.

#### 4. Conclusion

Development of noble gas mass spectrometers has enabled the analysis of single micrometeorites, and noble gas isotopic research has revealed that most micrometeorites collected on the Earth preserved detectable amounts of SW-derived He, Ne, and Ar. However, Kr and Xe are dominated by the primordial component, and solar-derived Xe is almost negligible. The anomalously high  $^3\text{He}/^4\text{He}$  ratio and solar-wind-like Ne isotopic composition observed in deep-sea sediments are caused by abundant micrometeorites accumulated on the bottom of the ocean. SW noble gases in micrometeorites were energetically implanted into the surface of micrometeorites in interplanetary space during orbital evolution, but they were partially depleted and fractionated by atmospheric entry heating. Noble gases in cosmic spherules were considerably depleted by harsh heating. The short CRE ages of most micrometeorites inferred from the lack of cosmogenic  $^{21}\text{Ne}$  and  $^{38}\text{Ar}$  show that the duration of solar wind exposure is less than 1 Myr. Since the terrestrial ages of IDPs and AMMs recovered from fresh Antarctic snow are very low, the trapped SW noble gases in these micrometeorites reflect the composition of recent solar wind.

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This book consists of a selection of original papers of the leading scientists in the fields of Space and Planetary Physics, Solar and Space Plasma Physics with important contributions to the theory, modeling and experimental techniques of the solar wind exploration. Its purpose is to provide the means for interested readers to become familiar with the current knowledge of the solar wind formation and elemental composition, the interplanetary dynamical evolution and acceleration of the charged plasma particles, and the guiding magnetic field that connects to the magnetospheric field lines and adjusts the effects of the solar wind on Earth. I am convinced that most of the research scientists actively working in these fields will find in this book many new and interesting ideas.

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