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General characterization of Antarctic micrometeorites collected by the 39th Japanese Antarctic Research Expedition: Consortium studies of JARE AMMs (III)

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Abstract: From November 1998 to January 1999, the 39th Japanese Antarctic Research Expedition (JARE-39) undertook Japanese first large-scale collection of Antarctic micrometeorites (AMMs), with sizes larger than 10 μm, at the Meteorite Ice Field around the Yamato Mountains in Antarctica (at three different locations, for a total of 24 collection sites). The number of collected AMMs larger than 40 μm is estimated to be about 5000. Here we present the general characterization (*i.e.*, micro-morphology and surface chemical composition using SEM/EDS) of ~810 AMMs chosen from 5 of the 24 sites. Additionally, the mineral composition of 61 out of 810 AMMs was determined by Synchrotron X-ray radiation. Preliminary results on mineralogical and chemical compositions show similarities with that of previous studies, even though a pronounced alteration of some AMMs is noticed. A correla-

tion is found between the Mg/Si ratio at the sample's surfaces of unmelted AMMs and the age of snow/ice in which the AMMs are embedded.

1. Introduction

Micrometeorites with sizes smaller than 1 mm are the dominant contributor of extraterrestrial material accreted onto the Earth. Since the first successful collection of micrometeorites in the blue ice lakes of Greenland (Maurette *et al.*, 1986, 1987), Antarctic micrometeorites (AMMs) have been also recovered from various locations in Antarctica, such as the blue ice fields of Cap-Prudhomme (Maurette *et al.*, 1991, 1994), the Astrolabe glacier (Gounelle *et al.*, 1999), the till from the Dominion Range and other locations in the Beardmore Glacier/Walcott Neve region in the Transantarctic Moutains (Koeberl and Hagen, 1989), the South Polar water well at the Scott-Amundsen Station (Taylor *et al.*, 1998), and an ice water tank at the Dome Fuji Station on the top of the Queen Maud Land (Nakamura *et al.*, 1999; Noguchi *et al.*, 2000).

AMMs collected by the Japanese Antarctic Research Expedition (JARE) have been studied by Japanese consortia, since Yano and Noguchi (1998) first demonstrated AMM sample processing and initial analysis techniques. Nakamura *et al.* (1999) and Noguchi *et al.* (2000) then successfully performed the initial characterization of, and established the curation system for, the "Dome Fuji collection", consisting of about 230 AMMs from an ice water tank at the Dome Fuji Station on the top of the Queen Maud Land. Following these works, this is therefore the third consortium study on AMMs in Japan, which is organized by AMM researchers from 13 institutes in Japan (Table 1). Detailed studies on AMMs collected by JARE have been published by Japanese AMM researchers: bulk composition of individual AMMs using synchrotron X-radiation by Nakai *et al.* (2000), mineralogy using TEM by Noguchi and Nakamura (2000), noble gas isotopic study by stepped pyrolysis (Nakamura and Takaoka, 2000) and by laser extraction of individual AMMs (Osawa *et al.*, 2000a), infrared microspectroscopic analyses (Osawa *et al.*, 2000b), oxygen isotopes (Hiyagon *et al.*, 2000), instrumental neutron activation analysis (Fukuoka *et al.*, 2000), and U-Pb analysis using SIMS

Table 1. Organization of the consortium study on JARE-39 AMMs.

Institution	Picking	SEM/EDS	remarks	Investigators
Hiroshima Univ.	○		SIMS, EPMA, X-ray microscopy	Terada K.*, Mori T.
Kyushu Univ. Rissho Univ.	○	○	EPMA, Gandolfi, Noble gas INAA	Nakamura T.*, Yada T., Nozaki W. Takaoka N Fukuoka T.*
NIPR@ Gakushuin Univ.	○	○	Curation of AMMs Web Catalog	Kojima H.* Murakami T.*
Dokkyo Univ.	○		X-ray microscopy	Nogami K.*, Ohmori R.
Ibaraki Univ. Tokyo Univ. Fish	○	○	EPMA, TEM AMS	Noguchi T.*,Matsumoto N., Kamata J. Ohashi H.*
ISAS#	○	○	EPMA	Yano H.*
Tokyo Univ.	○		SIMS	Hiyagon H.*, Mizutani S.
Tokyo Univ. Osaka Univ.	○		Noble gas, Infrard spec. X-ray CT	Nagao K.*, Osawa T. Tsuchiyama A.*
Tokyo Sci. Univ.	○		Synchrotron XRF	Nakai I.*, Sasaki M., Itabashi M., Setoyanagi T.

NIPR@; National Institute of Polar Research
ISAS#; Institute of Space and Astronautical Science
* arc Principal Investigators of each institution

Table 2. Overview of JARE-39 collection.

Collection area	Collection site	Heating duration for melting (hr; min)	Weight of ice (tons)	Weight of 40-100 μm Size Fraction* (g)	Weight of 100-238 μm Size Fraction* (g)	Weight of >238 μm Size Fraction* (g)	Comments
South of Minami-Yamato Nunataks	#1	4h45m	0.99	0.01134	0.01162	0.00811	Pre.
	#2	5h20m	1.11	0.01089	0.00418	0.00561	Req.(unbiased)
	#3	4h30m	0.94	0.00365	0.00479	0.00534	Flux,Cons.Req.(cataloged)
Kuwagata Nutataks	#1	10h	2.09	0.00750	0.01104	0.02162	Pre.
	#2	4h25m	0.92	0.00523	0.00209	0.00402	Flux, Cons.
	#3	9h55m	2.07	0.00439	0.00519	0.0121	Req.(unbiased)
	#4	5h25m	1.13	0.00328	0.00455	0.008	Req.(unbiased)
	#5	12h55m	2.7	0.00442	0.00545	0.00402	Req.(unbiased)
	#6	5h55m	1.24	0.00659	0.00699	0.00994	Req.(unbiased)
	#7	12h45m	2.66	0.00619	0.00763	0.01383	Pre.
	#8	6h	1.25	0.00656	0.00837	0.00317	Pre.
	#9	9h45m	2.04	0.00760	0.02013	0.0101	Pre.
	#10	6h20m	1.32	0.00396	0.00676	0.00559	Req.(unbiased)
	#11	9h15m	1.93	0.00242	0.00305	0.0027	Flux, Cons.
JARE IV Nunataks	#1	9h45m	2.04	0.06804	0.08564	0.04762	Pre.
	#2	7h15m	1.52	0.01545	0.02493	0.02659	Pre.
	#3	4h40m	0.98	0.00592	0.00792	0.00999	Pre.
	#4	6h20m	1.32	0.02243	0.00524	0.00373	Req.(unbiased)
	#5	6h50m	1.43	0.00654	0.00871	0.00834	Req.(cataloged)
	#6	5h15m	1.1	0.02849	0.01629	0.01845	Pre.
	#7	6h30m	1.36	0.00438	0.00747	0.00367	Req.(unbiased)
	#8	7h	1.46	0.00303	0.00331	0.00836	Req.(unbiased)
	#9	5h30m	1.15	0.00513	0.00228	0.00296	Flux, Cons.
	#10	5h	1.04	0.00186	0.00281	0.00139	Flux, Cons.
Total(24 sites)		171h20m	35.79	0.24533	0.26646	0.24527	

"Pre." stands for samples for future analysis
"Req." stands for samples for allocation at this time
"Flux" stands for samples studied for estimation of AMM flux
"Cons" stands for sample partially analyzed by the consortium for preliminary studies
* The weight of all kind of filtered particles, including terrestrial particles and artificial contaminants as well as micrometeorites

(Terada *et al.*, 2000).

From November 1998 to January 1999, JARE-39 undertook Japanese first large-scale and systematic collection of AMMs at three different areas in the Meteorite Ice Field around the Yamato Mountains in Antarctica; South of Minami-Yamato Nunataks, Kuwagata Nunatak, and JARE IV Nunataks (Yada and Kojima, 2000). The important objectives of this study are: (i) the general characterization of collected AMM samples as well as assessment of terrestrial and artificial contamination, (ii) judging from the collected AMMs the quality and efficiency of sample collection techniques from polar ice fields, and (iii) developments of handling and instrumental techniques in laboratories. Point (i) is an indispensable information obtained from this mission, and can be used for flux analysis on AMMs prior to initial analyses of elemental and other properties of the individual AMMs within the consortium. Point (ii) also provides important information for judging the best sample sites and whether samples at each site were artificially biased or not. Such information has been used to feed back to the AMM collection methods for JARE-41, which is currently being undertaken. Point (iii) provides new insights into the sample processing and initial analysis of fine grained extraterrestrial materials for future work such as the ISAS's MUSES-C sample return mission from a near-Earth asteroid (Fujiwara *et al.*, 1999).

For these reasons, all the samples were divided into three different groups: (1) samples from 5 different sites were used for flux counting (See, Yada, 2001), and for initial investigation within this consortium study, (2) samples from 10 other sites are held as "Research Samples" which are internationally available to qualified investigators, and (3) samples from the last 9 sites will be kept for future work, as summarized in Table 2. The collection sites used for this consortium study are site #3 of south of Minami-Yamato Nunataks, sites #2 & #11 of Kuwagata Nunataks, and sites #9 & #10 of JARE IV Nunataks (hereafter we will call them, MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10, respectively). In this study, we have performed the general characterization of those JARE-39 AMMs from 5 sites by nondestructive methods: size distribution, surface chemical composition, and mineral composition.

2. Samples and methods

2.1. *Collection method*

We will briefly describe here the collection method used by JARE-39: a radiator filled with warm (40–50°C) liquid was laid on bare ice surface, melted the ice, and finally formed a ~1 m-deep pond in about 6 hours. After pulling up the radiator from the pond, the melted water was pumped through a Teflon tube to the first filter holder. It was then filtered by four grades of stainless-steel sieves, and consequently fine particles were separated into four size fractions in situ (10–40 μm, 40–100 μm, 100–238 μm, and $>$238 μm). In order to maximize the particle collection, the pump swept the water at the bottom of the pond and pumped up most of the particles at the bottom. The duration of the heating at each collection site ranged from 5 to 14 hours, as the efficiency of heating and melting at each collection site was different. Only 200 liters of melt ice water (~10% of the total amount) in each pond could be pumped and filtered, as it began to freeze about one hour after removing the radiator. A total of 36 tons of bare ice at 24 different sites was melted (see Table 2). All samples were kept frozen during shipping back to Japan. The collection procedures are described in more detail by Yada and Kojima (2000).

2.2. *Flow chart*

First, all AMM candidates, which are black, irregularly shaped and/or rounded particles, were hand-picked under an optical stereo-microscope. These were then analyzed, without being polished or coated, by a scanning electron microscope equipped with an energy dispersive spectrometer (SEM/EDS). The AMMs among them were identified by their surface structure and chondritic composition, whose validation as a criterion for an extraterrestrial origin was first proven by Olinger *et al.* (1990). These procedures were based on the previous studies of precipitated micro particles collected from sediments in the Dome Fuji water tank (Nakamura *et al.*, 1999; Noguchi *et al.*, 2000). More than 800 AMMs larger than 40 μm were identified in this way from five sites (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10). This indicates that about 5000 AMMs in total must have been collected by JARE-39. Details of the AMMs from five sites are summarized in Table 3. More detailed information on individual AMMs has been cataloged on the web site (http: //dust.cc.gakushuin.ac.jp/dust). On the other hand, it is very difficult to identify AMMs in the size fraction 10–40 μm and $>$238 μm, and only 17 AMMs in the size range 10–40 μm were used for this study.

Next, the Japanese AMM consortium member investigated the same 75 unmelted AMM samples (17 AMMs in the size range of 10–40 μm, 49 AMMs in the size range of 40–100 μm and 9 AMMs in the range of 100–238 μm) using various analytical techniques. Mineral compositions of 61 of the 75 AMMs were investigated by a Gandolfi X-ray camera using synchrotron radiation, and 5 of them were further investigated by Transmission Electron Microscopy (TEM). 21 AMMs of the 61 were each split into 2 fragments individually. One fragment of each AMM was analyzed by infrared micro-spectroscopy and the other fragment was analyzed by Electron Probe Micro Analyzer (EMPA). In addition, 35 whole AMMs where also analyzed by EMPA. After this, the isotopic studies were carried out by Secondary Ion Mass Spectrometer (SIMS). 16 AMMs were analysed for Rare Earth Elements (REEs), U, Th, and Pb, and 15 AMMs for oxygen isotopes, respectively. Trace element compositions of 9 AMMs in the range 100–238 μm were determined by Instrumental Neutron Activation Analysis (INAA). All of these results will be presented elsewhere by other consortium members.

In this study, we present the results of size distribution, surface chemical composition, and the mineral composition as the general characterization of JARE-39 AMMs by nondestructive methods.

Table 3. Species of AMMs larger than 40 μm.

	Unmelted AMMs	Stony spherules	Glassy spherules	Iron spherules	Total
MY-3	101	21	33	24	179
Kuwa-2	134	37	42	59	272
Kuwa-11	172	42	32	10	256
JARE4-9	18	1	1	2	22
JARE4-10	30	27	22	4	83
Total	455	128	130	99	812

2.3. Analytical methods

The observation of surface structure and the major element compositions of sample surfaces (without polishing or conductive coating) were investigated by the scanning electron microscopes with energy dispersive spectrometers at Kyushu University (JEOL JSM-5800LV), Ibaraki University (JEOL JSM-5600LV), and National Institute of Polar Research (JEOL JSM-5900LV), which were operated at an electron accelerating voltage of 20 kV and emission current of 1 nA.

In order to investigate the mineral composition of individual AMMs from Kuwa-11 and MY-3, nondestructive techniques, X-ray diffraction analysis was performed using a Gandolfi camera with a monochromated synchrotron X-ray microbeam of 2.16 Å wavelength at the beam line 3A in the Photon Factory Institute of Material Structure

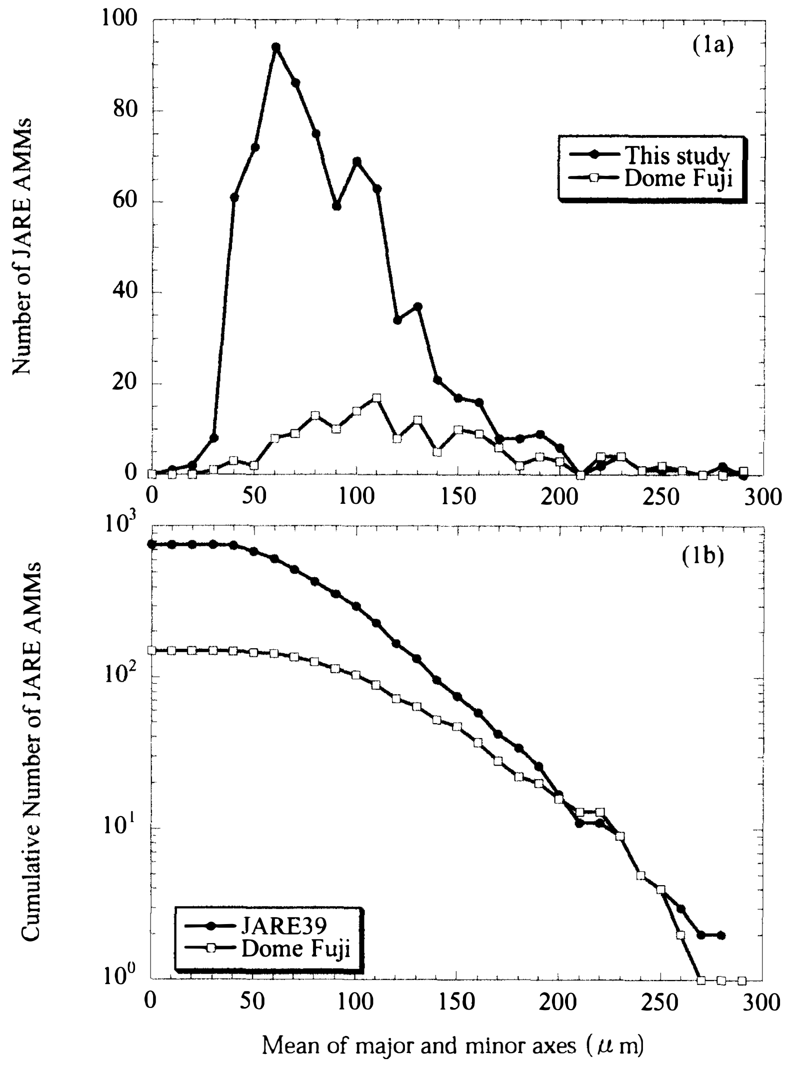

Fig. 1. Size frequency distributions of JARE-39 AMMs larger than 40 μm, collected from 5 collection sites (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10). The X-axis shows the mean values of major axis and minor axis of individual AMMs, and the Y-axes represent the size frequency distribution in the top diagram (1a) and the cumulative distribution in the bottom diagram (1b), respectively.

Science, High Energy Accelerator Research Organization, Tsukuba, Japan. (Analytical procedures are described in more detail in Nakamura *et al.*, 2001).

3. Results

3.1. Size-frequency distribution

Size frequency distributions of the AMMs and cosmic spherules from the size fraction larger than 40μm were investigated based on the SEM observations. Figure 1 shows the size frequency distributions and the cumulative size distributions of AMMs recovered by JARE-39 (this study) compared with those from Dome Fuji AMMs collected by JARE-37. The X-axis is the mean value of the major and minor axes of individual AMMs. The slope of the JARE-39 AMMs in cumulative distributions is

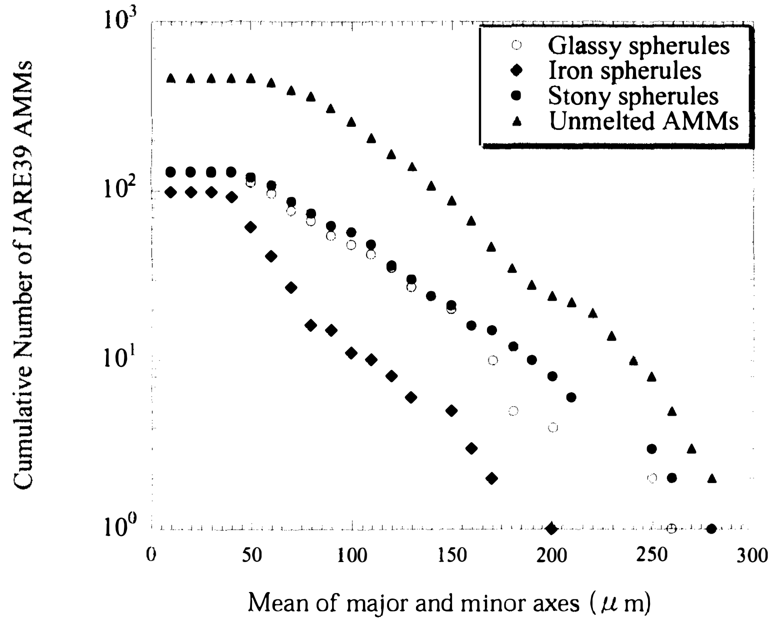

Fig. 2. Size frequency distribution of spherules and unmelted AMMs larger than 40μm, collected from 5 collection sites (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10). The X-axis shows the mean values of major axis and minor axis of individual AMMs, and the Y-axis shows the cumulative numbers. The species of AMMs and spherules were identified by SEM/EDS analyses.

Table 4. The tail slopes of cumulative size distributions.

		unmelted AMMs	Glassy spherules	Stony spherules	Iron spherules
	40 - 100 μm	-0.6*	-1.1*	-1.0*	-2.4*
This study	> 100 μm	-4.4*	-4.1*	-3.6*	-3.5*
Murrell et al.(1983)	> 200 μm			-3.83	-
Kyte (1983)	> 200 μm			-3.4±0.7	-5.9±3.0
Taylor et al.(2000)#	> 200 μm			-3.5±0.7	-4.4±2.2
Taylor et al.(2000)@	> 200 μm			-5.2±0.5	-2.4±1.2

* The uncertainties of these values are about ±10%
# DSS Box Core sample
@SPWW sample

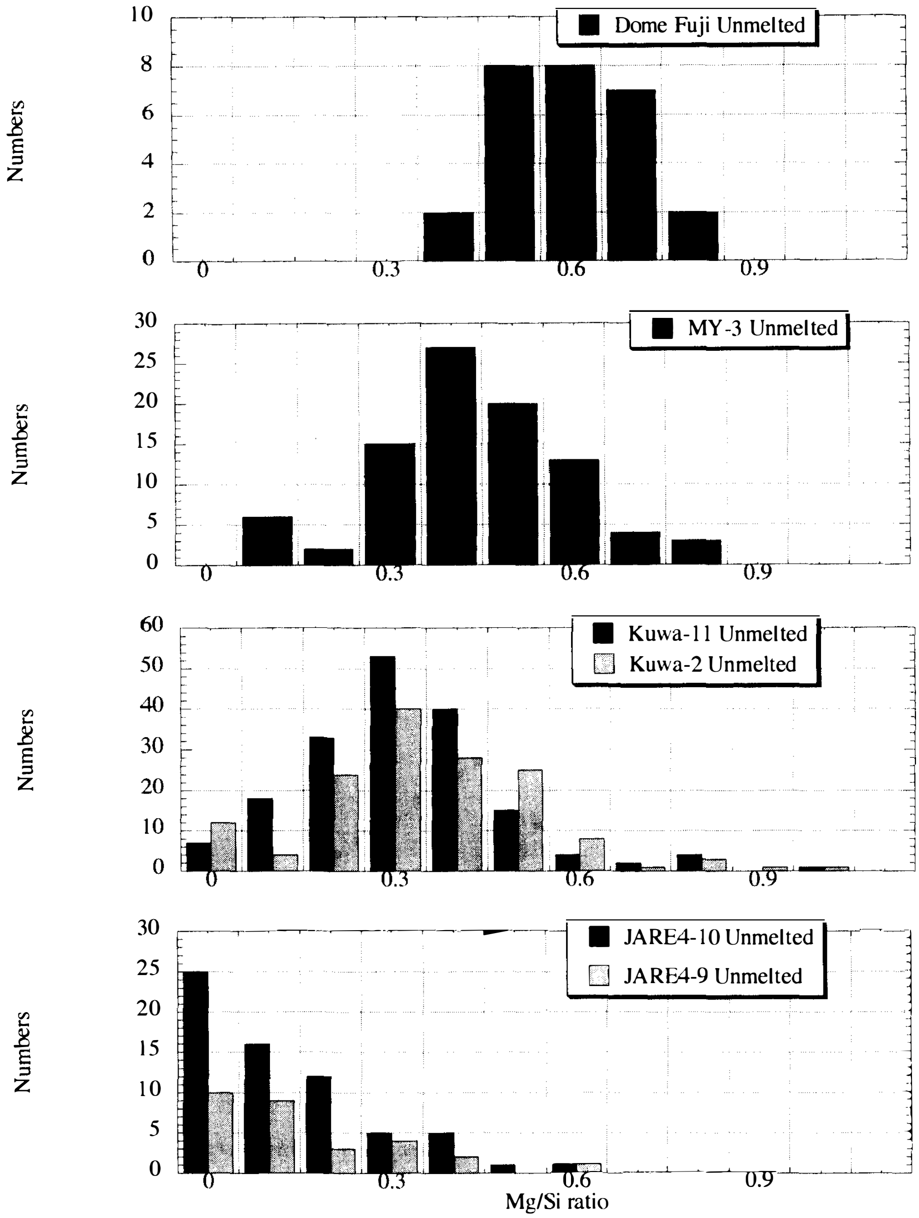

Fig. 3. Distribution of the Mg/Si ratios of peak heights in X-ray spectra measured by SEM/EDS at the surface of unmelted AMMs of the JARE-39 samples (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10) and Dome Fuji AMMs.

apparently steeper than that of the Dome Fuji collection, indicating that AMMs in the smaller size range (especially less than 100μm) were more efficiently collected by this project. The cumulative size distributions of unmelted AMMs and three kinds of cosmic spherules larger than 40μm are shown in Fig. 2, where the abscissa represents the mean of the minor and major axes used as a measure of the particle's sizes. The distributions of glassy and stony spherules are similar, but different from that of iron (type I) spherules (especially, smaller than 100μm). The tail slope of the cumulative size distribution is partially well fitted by an equation of $N\ (>d)=d^{\alpha}$, where d is a diameter of AMMs and $N\ (>d)$ is a cumulative number larger than d. The tail slopes

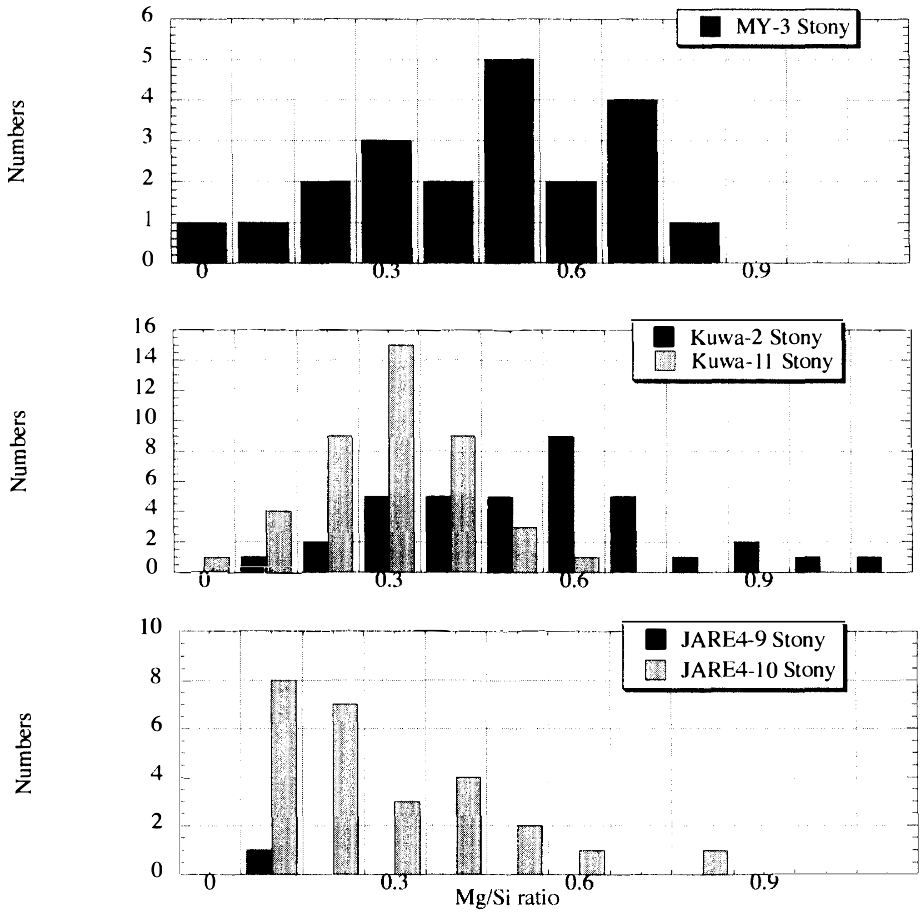

Fig. 4. Distribution of the Mg/Si ratios of peak heights in X-ray spectra measured by SEM/EDS at the surface of stony spherules of the JARE-39 samples (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10).

(α) of these distributions are summarized in Table 4.

3.2. Chemical composition of the sample's surfaces

Almost all AMMs collected from different locations showed the following compositional features in the X-ray energy spectra: (1) large peaks of Mg, Si and Fe, (2) small or no peaks of Al and Ca, (3) very small or no peaks of Cr, Ni and S, which are variable. These features are very similar to previous works of EUROMET samples (Maurette *et al.*, 1991; Kurat *et al.*, 1994; Engrand and Maurette, 1998) and Dome Fuji sample (Nakamura *et al.*, 1999; Noguchi *et al.*, 2000). The distributions of Mg/Si ratios of peak heights in X-ray spectra measured by EDS for unmelted AMMs, stony spherules and glassy spherules are shown in Fig. 3, Fig. 4 and Fig. 5, respectively. It is noticed that the distribution of the Mg/Si ratios of the spherules exhibit a larger variation range than that of unmelted AMMs, for which the reproducibility of Mg/Si measurements is less than a few percent. For unmelted AMMs, the average Mg/Si for the Dome Fuji collection has the highest value of about 0.64 ± 0.11 (1σ), that of MY-3 is 0.48 ± 0.16, averages for Kuwa-2 and Kuwa-11 are similar (0.40 ± 0.17 and 0.43 ± 0.20, respectively), and those for JARE4-9 and JARE4-10 are comparable (0.22 ± 0.23

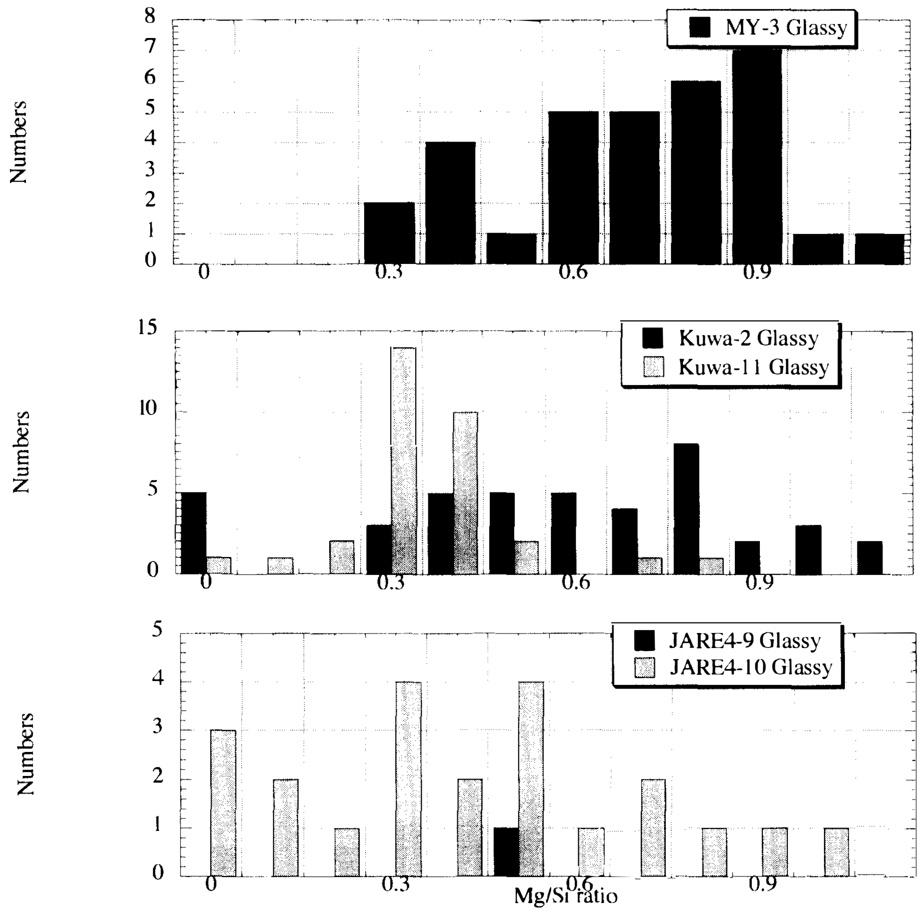

Fig. 5. Distribution of the Mg/Si ratios of peak heights in X-ray spectra measured by SEM/EDS at the surface of glassy spherules of the JARE-39 samples (MY-3, Kuwa-2, Kuwa-11, JARE4-9 and JARE4-10).

and 0.19 ± 0.14, respectively). In comparison, The Mg/Si ratios measured on the surface of CM chondrite matrix investigated at the same conditions is 0.74 ± 0.17.

The Ca/Si and Fe/Si distributions of the spherules and unmelted AMMs are also investigated individually, but unlike Mg/Si there is a large variation range for each collection site and no systematic difference between sites. No significant correlation between the Mg/Si, Fe/Si and Ca/Si ratios was found. The Ca/Si and Fe/Si distributions of unmelted JARE-39 AMMs are shown in Fig. 6 and Fig. 7.

3.3. Mineralogical compositions

Typical X-ray diffraction patterns of these AMMs are shown in Fig. 8, illustrating (a) olivine-rich, (b) pyroxene-rich, and (c) olivine-pyroxene samples. 87%, 100%, 51% and 57% of those AMMs contained olivine, magnetite, low-Ca pyroxene, and Fe sulfides (mainly troilite), respectively. Our results indicate that 58 of the 61 AMMs (95% of them) consist entirely of a combination of four kinds of anhydrous minerals such as olivine, low-Ca pyroxene, magnesiowüstite, and Fe-sulfides. The remaining 3 AMMs (5% of the totals) contain phyllosilicates. One of them consists of smectite and magnetite, and the other two consist of serpentine and magnetite. Although the

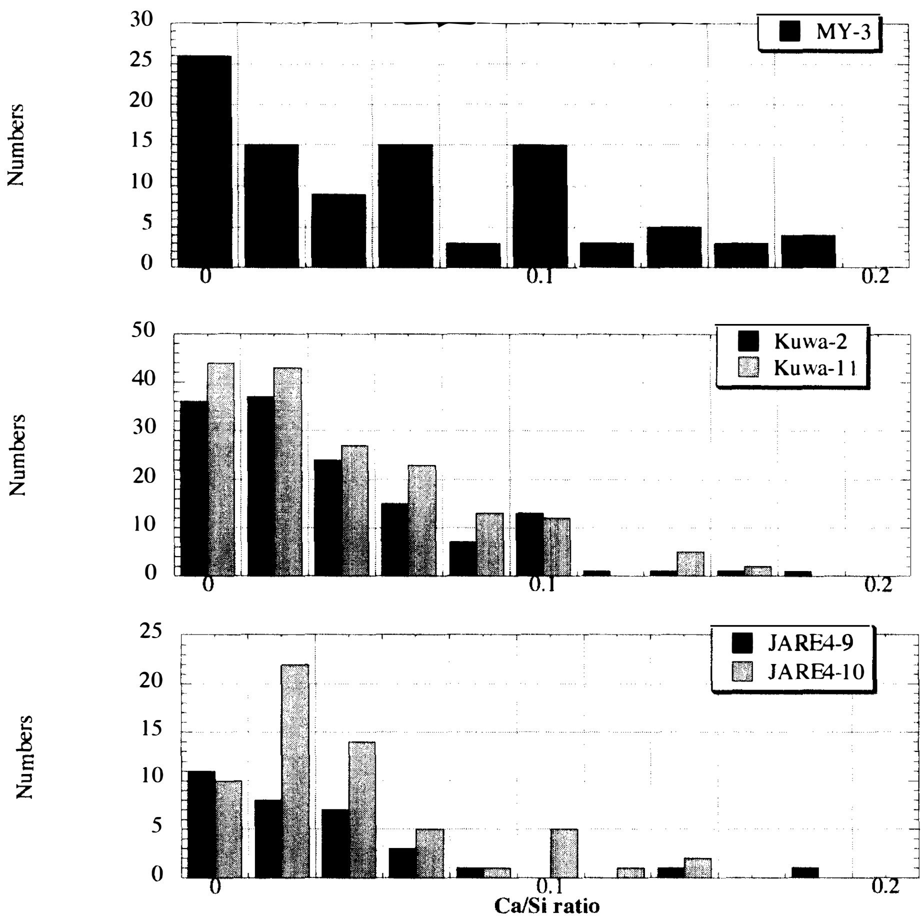

Fig. 6. Distribution of the Ca/Si ratios of peak heights in X-ray spectra measured by SEM/EDS at the surface of the unmelted AMMs.

existence of these minerals in AMM were pointed out by Kurat *et al.* (1994), Klöck and Stadermann (1994), Maurette *et al.* (1996), Engrand and Maurette (1998), it is found in this study that such a relative abundance of mineral assemblages is very similar to those of the Dome Fuji collection and EUROMET samples (Nakamura *et al.*, 2001). Moreover, jarosites [$KFe_3(SO_4)_2(OH)_6$] were also found in 5% (=1 AMM over 24) of the MY-3 samples and in 43% (=16 AMMs over 37) of the Kuwa-11 samples, yet they have not been reported from either the Dome Fuji or EUROMET collections. These results are summarized in Table 5.

4. Discussion

4.1. Size distribution of AMMs

As shown in Fig. 1, the distributions of Dome Fuji and JARE-39 AMM collections are quite different (especially below 100 μm). Since the Dome Fuji collection was made by recovering the precipitated particles at the bottom of a water tank used for human activity, only about 0.3 wt% of fine particles were AMMs (Nakamura *et al.*, 1999). The remaining terrestrial materials are mainly composed of spherules formed during welding and deposits of other human activity. On the other hand, the fine extraterres-

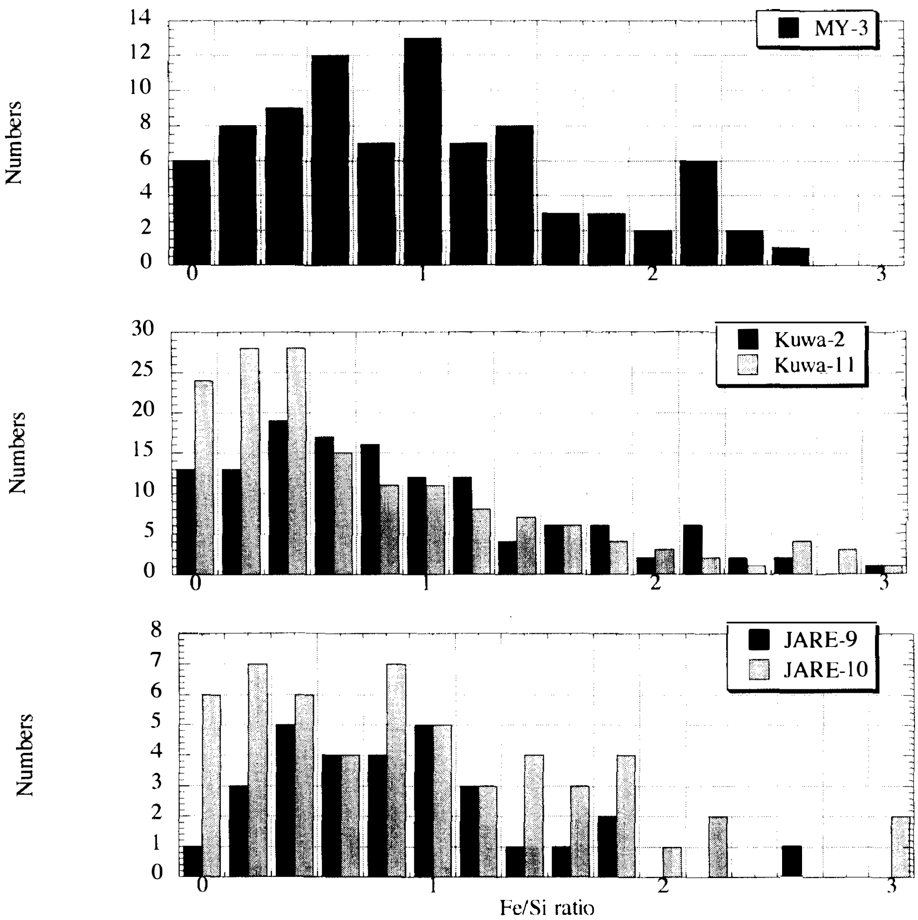

Fig. 7. Distribution of the Fe/Si ratios of peak heights in X-ray spectra measured by SEM/EDS at the surface of the unmelted AMMs.

trial particles recovered from the 5 sites presently studied represent roughly a few percent of the material collected by JARE-39. Such a high concentration of AMMs with regard to terrestrial grains would diminish the probability of overlooking the small extraterrestrial particles during hand-picking.

Recently, the size distribution of Antarctic spherules was discussed by Taylor *et al.* (1998, 2000). As shown in Table 4, the features of the cumulative distributions of stony and iron spherules in the range $>100\mu m$ are compatible with those found by previous studies of Murrel *et al.* (1980), Kyte (1983) and Taylor *et al.* (2000) in the range $>200\mu m$, except for the case of the stony spherule distribution found in the SPWW sample by Taylor *et al.* (2000). In this study, it is also confirmed that the slopes of all kinds of spherules are similar in the range $100\mu m$. The cumulative distributions of glassy and stony spherules are flat in the 10–40μm range. This tendency suggests a preferential loss of small particles of such kinds of spherules, as discussed by Taylor *et al.* (2000).

The distribution of unmelted AMMs show that the slope below $100\mu m$ of the distribution of unmelted AMMs are flatter (slope of -0.6) than those of spherules. The JARE-39 samples were recovered from the bottom of the pond by pumping and filtering only 10% of the melt ice water. It is thought that irregularly-shaped AMMs

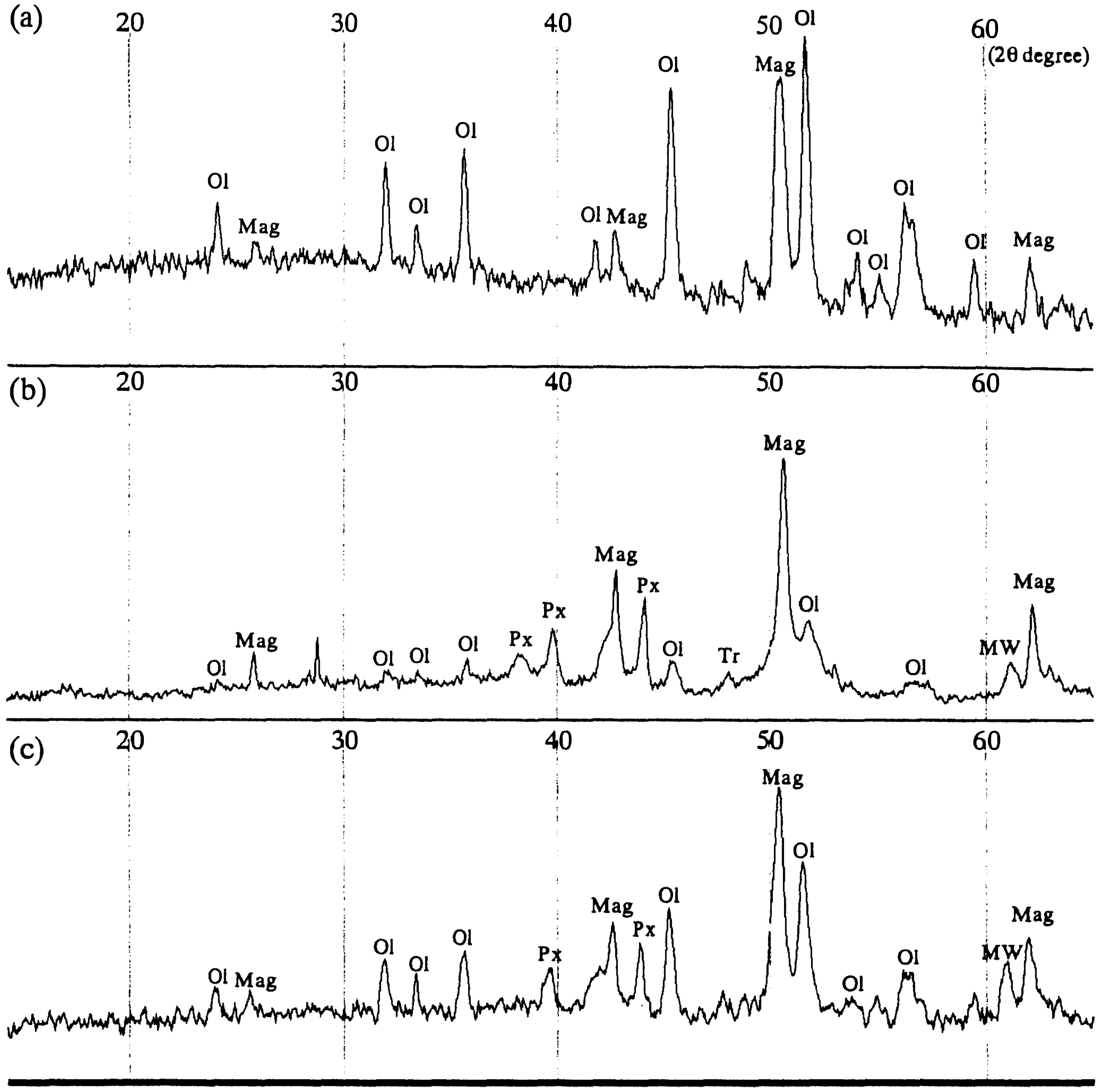

Fig. 8. Typical X-ray diffraction patterns of AMMs. (a) Olivine-rich, (b) pyroxene-rich, and (c) olivine-pyroxene samples. Abbreviations: Ol=olivine, Px=low-Ca pyroxene, Mag=magnetite, Tr=troilite, and MW=magnetiowüstite.

Table 5. Mineral species JARE-39 AMMs in comparison with those collected at other locations.

	This study		Dome Fuji & EUROMET samples#	
AMMs containing olivine	53/61	(87%)	53/56	(95%)
AMMs containing magnetite	61/61	(100%)	53/56	(95%)
AMMs containing low-Ca pyroxene	31/61	(51%)	36/56	(64%)
AMMs containing Fe-sulfides	35/61	(57%)	32/56	(57%)
AMMs containing phyllosilicate	3/61	(5%)	3/56	(5%)
AMMs containing magnesiowustite*	25/61	(41%)	22/56	(39%)
AMMs containing jarosite	17/61	(28%)	0/56	(0%)

*magnesiowustite with variable Fe/Mg ratio
#Nakamura et al. (2001)

experience greater friction with the melt water and thus take longer to settle to the bottom of the pond than spherules, and therefore there is a possibility that irregular AMMs might have been less efficiently collected than the spherules. This could be corroborated by the observation of the different slopes in the cumulative distributions of Dome Fuji and JARE-39 AMMs (Fig. 1). Selection effects during handpicking under an optical stereomicroscope also are possible because small unmelted AMMs are harder to recognize than the spherules, even though all candidates were picked up and investigated by SEM/EDS. Overlooking during identification of AMMs candidates by using SEM/EDS is also possible, because severely altered AMMs might show the negligible small peaks (discuss them in Section 4.2). Another possibility is that unmelted micrometeorites are more fragile than spherules so they tend to be broken into fine particle smaller than 10μm during a long duration embedded in the ice or during the filtration procedure. In order to diminish these probable selection effects, the collection method of the current JARE-41 project has been modified such that all the melted water undergoes filtration.

4.2. Alteration on sample surface of MM in the ice

The average elemental ratio of Mg/Si of unmelted AMMs collected from 5 different sites shows different values from each other. The average values of AMMs from JARE 4–9 and JARE4–10 sites, which were the closest to the Yamato Mountains (about 5 km away), have the lower values of 0.22 ± 0.23 and 0.19 ± 0.14, respectively (1σ). Those of Kuwa-2 and Kuwa-11 samples, which were about 44 km away from the mountain, have values of 0.40 ± 0.17 and 0.43 ± 0.20 respectively. The MY-3 samples, which were the furthest from the mountain in this expedition (about 77 km), have the highest value of 0.48 ± 0.16. We believe that this is not an experimental artifact due to a shielding effect of the Mg X-ray by various thickness of the magnetite rim generated at the surface of the AMMs during atmospheric entry, as there is no correlation between the Mg/Si and Fe/Si ratio. These results mean that the variations of the average values of Mg/Si measured at the AMMs surface apparently correlate with the distance between the ice field from which they where collected and the Yamato Mountains.

According to the surface flow modeling of ice in the Yamato bare ice field (Azuma *et al.*, 1985), the greater the distance from mountains and nunataks, the younger the bare ice becomes. The ice core which is closest to the JARE IV Nunataks (JARE4–9 and JARE4–10) has the oldest age, 50000–60000 years. The δ^{18}O variation in air bubbles trapped inside the ice core samples close to the Kuwagata Nunataks was well investigated by Nakagawa *et al.* (private commun. 2000), indicating that the age of the ice core close to the surface is 31000–32000 years. The age of the ice core South of Minami-Yamato (MY-3) has not been published, but the age of ice/snow in which the Dome Fuji AMMs were embedded is very young (at most 100 years, Nakamura *et al.*, 1999). The correlation between the age of ice/snow and the mean values of Mg/Si ratio is shown in the Fig. 9, which is well fitted by a linear relationship of $\mathrm{Mg/Si}=0.658-6.99\times10^{-6}\times\mathrm{Age}$ ($R=0.995$). Here, assuming that the rate of Mg depletion at the sample surface should be proportional to the Mg concentration at the same surface (in other words, $-\mathrm{dMg}/\mathrm{d}t\propto\mathrm{Mg}$), this negative correlation could be well described arithmetically by an exponential equation of $\mathrm{Mg/Si}=0.72\times\exp(-0.000021\times\mathrm{Age})$ ($R=$

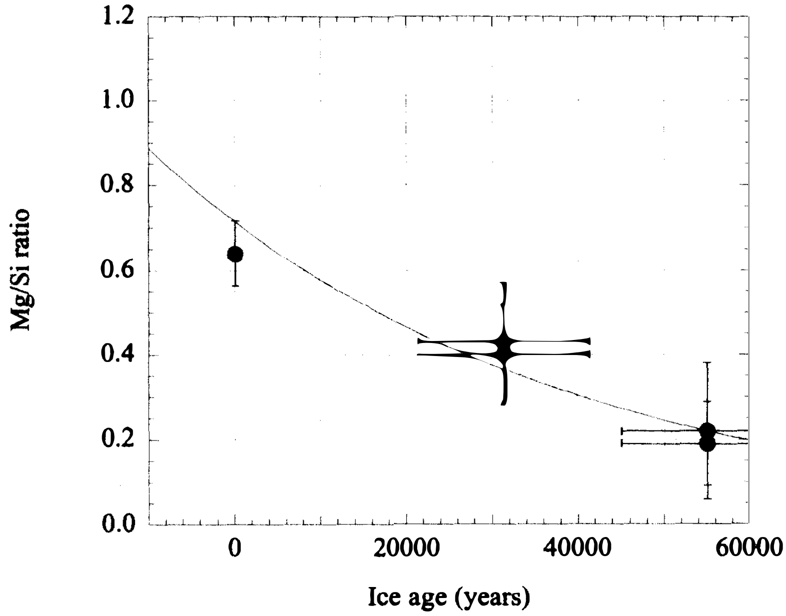

Fig. 9. Correlation diagram between the Mg/Si ratios measured by EDS at the surface of JARE-39 unmelted Antarctic micrometeorites (AMMs) and the age of the ice/snow which the AMMs were embedded. The error bars of the Mg/Si ratios are calculated standard deviations (1 σ).

0.987), where it should be noted that the normalization value of 0.72 (at Age=0) is very similar to the 0.74±0.17 for CM chondrite.

The abundance of jarosite [$KFe_3(SO_4)_2(OH)_6$], which is considered to be a by-product mineral resulting from aqueous alteration of sulfide minerals while in the ice, also correlates with the distance from the Yamato Mountains, which is equivalent to the age of ice/snow. Indeed, jarosite is observed in ~43% of Kuwa-11 samples (about 44 km away, 31000–32000 years), in ~5% of MY-3 samples (about 77 km away), and is not significantly found in the Dome Fuji AMMs (at most 100 years). For the first time, such a correlation between the variation of the chemical/mineralogical compositions of AMMs and the embedded duration time in the ice/snow is, confirmed by a systematic collection of AMM at different sites by the JARE-39 team.

The depletion of Ca, Na, Se, S, Mg, and Mn and the enrichments of Au and As in AMMs have been discussed elsewhere (Maurette *et al.*, 1991, 1992; Klöck *et al.*, 1992; Koeberl *et al.*, 1992; Kurat *et al.*, 1992a), although there has been no consensus regarding the cause of these effects (preterrestrial or terrestrial). For example, it was pointed out that the comparison of the compositions of unmelted AMMs with cosmic spherules shows that depletions in Ca and Ni can be preterrestrial (Maurette *et al.*, 1992). Based on the bulk composition of the EUROMET AMMs in the size range 50–400 μm using INAA, Kurat *et al.* (1992b) suggested that the Mg and Ca depletions could be due to terrestrial leaching of dolomite and calcite respectively, both of which are common phases in CM and CI chondrites, and missing in AMMs. Our finding of a negative correlation between the Mg/Si ratio and the age of the embedded ice strongly supports "terrestrial leaching". However, the fact that relict of dolomite and other "strange" minerals even in the least altered Dome Fuji sample have not been found in the AMMs by Gandolfi X-ray Camera analyses indicates that the Mg depletion process on the AMM's surface differs from that of bulk Mg depletion explained by carbonate dissolution, and maybe results in alteration of more major minerals such as olivine and pyroxene. In this case, the correlation between the crystalline quality of these minerals on the surface and the ice/snow ages could be confirmed by observation of the textures of individual AMMs. Using Transmission Electron Microscopy (TEM), Yano and Noguchi (1998) have already pointed out that ferroan olivine grains in some AMMs include planar defects parallels to (001) plane, which are formed under acidic aqueous conditions. For a better understanding of alteration processes inside/outside of AMMs while in the ice/snow, detailed mineralogical studies and quantitative observations of more mobile elements in polished section of individual AMMs will be required. This is however beyond the scope of this present consortium study based on the non-destructive analyses.

5. Summary and future prospects

The results of the general characterizations of JARE-39 AMM carried out by this consortium are as follows.

(1) AMMs larger than about 40 μm were successfully recovered by the collection methods described in details by Yada and Kojima (2000). Irregular AMMs smaller than that (especially in the range of 10–40 μm) could not be efficiently collected due to unknown selection effects.

(2) A correlation between the Mg/Si ratio and the ages of snow/ice embedding the AMMs was discovered, which suggests that the Mg depletion results from "terrestrial leaching". Assuming that the original major element compositions of these AMMs are very similar and comparable to that of CM chondrite matrix, these results strongly indicate that the Dome Fuji collection, which shows almost the same Mg/Si ratio as fragments of CM chondrite matrix, contains the freshest (*i.e.*, least altered) samples, and is the best sample collection site of the four locations (South of Minami-Yamato Nunataks, Kuwagata Nunataks, JARE IV Nunataks and Dome Fuji Station) where JARE worked. This finding suggests that the age of ice/snow, which are still unknown, could be estimated by the measurement of surface composition of AMMs, which are embedded in them. It may also provide new insights into glaciology.

(3) The abundance of jarosite [$KFe_3(SO_4)_2(OH)_6$], which is considered to be a by-product mineral resulting from aqueous alteration of sulfide minerals while in the ice, also correlates with the age of ice/snow.

These results should be taken into account in the current/future projects of AMM collection by JARE.

With regard to the "Research Samples", some of the cataloged AMMs from the three locations and unbiased samples collected at ten other locations will become internationally available for qualified investigators upon submission of research proposals to the NIPR AMM curator (Prof. H. Kojima). More details about sample request procedures will be announced on the Japanese AMM web site (http: //dust.cc.gakushuin.ac.jp/dust/).

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