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COMPOSITIONS AND MINERALOGIES OF UNMELTED POLAR MICROMETEORITES: SIMILARITIES AND DIFFERENCES WITH IDPs AND METEORITES

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Abstract: Micrometeorites from 60 to 1200 μm in diameter constitute ~80% of the continuous, planet-wide accretion of meteoritic material onto the Earth. Although interplanetary dust particles (IDPs) < 60 μm are collected from the Earth's stratosphere, and particles > 1 cm are sampled as meteorites, the 60 to 1200 μm size range has, thus far, only been sampled by the recovery of meteoritic particles from polar ices and lakes and from the sea.

Polar micrometeorites have major element abundances consistent with chondritic meteorites and IDPs, except that Ni and S are strongly depleted. Synchrotron X-Ray Fluorescence trace element abundance measurements of twelve Antarctic and four Greenland micrometeorites from 50 to 150 μm show enrichments by 2 to 5 times over CI abundances for Cu, Zn, and Ga, however Ni, Ge, and Se are depleted relative to CI. This abundance pattern differs significantly from both chondritic meteorites and IDPs, including IDPs heated on atmospheric entry. There is no correlation of element enrichment or depletion with nebula condensation temperature.

Olivine, pyroxene, magnetite, chromite, spinel, and glass, which commonly occur in IDPs and meteorites, were identified in these particles by analytical transmission electron microscopy. However, layer-lattice silicates, also common in IDPs, CI and CM carbonaceous meteorites, were not identified in any of these polar micrometeorites. Textural relationships and mineral compositions suggest these polar micrometeorites were strongly heated during atmospheric entry.

The chemical compositions and mineralogies of the polar micrometeorites show similarities to both the IDPs and the chondritic meteorites, but they are not identical to either type of material.

1. Introduction

1.1. *General properties*

The analysis of meteorites and Interplanetary Dust Particles (IDPs) has provided useful constraints on the origin and evolution of the solar system. The recovery of micrometeorites from the polar regions provides an opportunity to study extraterrestrial material in a size range never before examined in unmelted form. These polar micrometeorites, which sample for the first time unmelted material near the peak of the

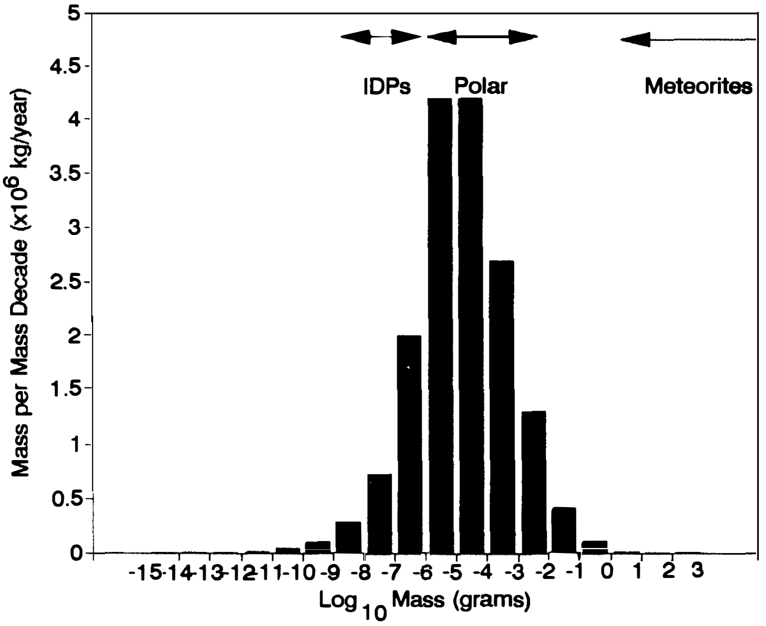

Fig. 1. Mass frequency distribution of interplanetary matter entering the Earth's atmosphere derived from visual and radar meteor and satellite dust detector measurements (data from HUGHES, *1978). There is a strong peak in the flux for particles from* 10^{-7} *to* 10^{-3} *grams (~60 to 1200 μm in diameter). The mass range of the recovered polar micrometeorites (Polar) corresponds to the peak in the interplanetary flux at Earth. The Interplanetary Dust Particles (IDPs) recovered from the Earth's stratosphere as well as the recovered meteorites (Meteorites) sample interplanetary matter which arrives at Earth in much lower abundances than that sampled by the polar micrometeorites.*

mass-flux curve (see Fig. 1), may provide new constraints on the origin and evolution of the solar system.

Micrometeorites up to several hundred microns in diameter have been recovered from polar lakes or ices in Greenland (MAURETTE *et al.*, 1987), Antarctica (MAURETTE *et al.*, 1991), and northern Canada (CRESSWELL and HERD, 1992). The recovered particles have survived atmospheric entry in a variety of conditions ranging from irregularly shaped fragments showing minimal physical evidence of heating, through particles exhibiting a scoriated texture indicating significant heating, and finally spherules which completely melted during atmospheric entry (SUTTON *et al.*, 1992).

Although smaller micrometeorites, up to ~60 μm in diameter, are recovered from the stratosphere of the Earth by NASA sampling aircraft (*e.g.*, review by BROWNLEE, 1985), micrometeorites >60 μm in size settle so rapidly that their concentration in the stratosphere is too low to allow efficient aircraft collection. Particles >60 μm in diameter are important to the understanding of the meteoritic flux at the Earth because the accreted meteoritic mass peaks sharply in the mass range from 10^{-7} to 10^{-3} grams, corresponding to a size range from about 60 to 1200 μm in diameter. As shown in Fig. 1, approximately 80% of the continuous, planet-wide accretion of meteoritic mass onto the Earth occurs in the mass range from 10^{-7} to 10^{-3} grams (HUGHES, 1978). Thus particles in the 60 to 1200 μm size range, unsampled by either the stratospheric IDP collections or the recovered meteorites but abundant in the polar micrometeorite collections, constitute the large majority of the meteoritic mass accreted on a continuous, planet-wide basis by the Earth.

The presence of radiogenic nuclei (RAISBECK and YIOU, 1989; NISHIIZUMI *et al.*, 1991) and spallogenic ^{21}Ne (OLINGER *et al.*, 1990) confirm an extraterrestrial origin for many of the cosmic spherules from the sea and the polar micrometeorites. In addition, the radiogenic nuclei confirm that most of these particles were irradiated in space in roughly the size and shape we see them now, indicating that most of the polar micrometeorites are not ablation products or fragmentation debris from much larger meteors (RAISBECK and YIOU, 1989; NISHIIZUMI *et al.*, 1991).

MAURETTE *et al.* (1991) report that the major element compositions and mineralogies of fifty-one irregular shaped micrometeorites from the Antarctic ices are similar to both the fine-grained matrix of primitive carbonaceous chondrites and to the 5 to 60 μm interplanetary dust particles (IDPs) collected from the Earth's stratosphere, but that the polar micrometeorites are not identical to either type of material. Both Ni and S are highly depleted relative to their CI meteorite abundances in the polar micrometeorites (MAURETTE *et al.*, 1991). They suggest that the large micrometeorites (50 μm to 1200 μm) recovered from the polar ices may be a new population of solar system material, mineralogically and chemically distinct from both the smaller stratospheric IDPs and the larger (> 1 cm) meteorites (MAURETTE *et al.*, 1991). ALEXANDER *et al.* (1992) indicate that 20 Antarctic micrometeorites, ranging from 50 to 110 μm in size, they studied are not simply larger versions of the IDPs because they do not show the heterogeneous enrichments of D/H measured in about one-third of the IDPs examined and because of the absence of a hydrated silicate signature in the infrared transmission spectra of their particles.

The abundances of volatile trace elements have previously proven useful in establishing genetic links among or distinguishing between the types of meteorites, and in identifying the IDPs recovered from the Earth's stratosphere as compositionally distinct from the chondritic meteorites (FLYNN and SUTTON, 1991b, 1992b). If the polar micrometeorites are mineralogically, chemically, and isotopically different from both the larger meteorites and the smaller IDPs, then the bulk of the meteoritic material accreted by the Earth may be compositionally and mineralogically distinct from the well characterized meteorites and the IDPs, and may provide new constraints on early solar system processes.

1.2. Sources

The survival of the polar micrometeorites on atmospheric entry provides information on their parent bodies. The peak temperature reached on atmospheric entry increases as the particle size, density, and entry velocity increase. Computer simulations show interplanetary dust particles > 75 μm in diameter survive atmospheric entry without melting only for entry velocities near earth escape velocity, the lower limit of the entry velocity distribution (FLYNN, 1990, 1992; LOVE and BROWNLEE, 1991). Such low entry velocities are characteristic of particles derived from main-belt asteroidal parent bodies but not from cometary parent bodies (FLYNN, 1989b). This suggests the unmelted polar micrometeorites are derived from main-belt asteroidal parent bodies (FLYNN, 1990, 1992; LOVE and BROWNLEE, 1991).

The space exposure ages of the polar micrometeorites inferred from the abundances of ^{21}Ne (OLINGER *et al.*, 1991) and radiogenic nuclei (RAISBECK and YIOU, 1989; NISHIIZUMI *et al.*, 1991) are consistent with the time required for the orbits of main-belt asteroidal particles to evolve into Earth-intersecting orbits. In addition, NISHIIZUMI *et al.* (1991) have inferred that the space exposures must have taken place in the inner solar system, eliminating particles in the highly elliptical orbits characteristic of cometary materials. Thus, the inferred space exposure ages support a main-belt asteroidal origin for the polar micrometeorites.

The computer simulations indicate that, for the distributions of atmospheric entry velocities measured for radar and visual meteors (HUGHES, 1978), most micrometeorites $> 100\,\mu$m in diameter vaporize on atmospheric entry (FLYNN and MCKAY, 1990; LOVE and BROWNLEE, 1991). The detection of radar meteor trails produced by particles as small as 10^{-6} g (HUGHES, 1978), about $120\,\mu$m in diameter, confirms that many of these particles are vaporized. Thus the large polar micrometeorites sample only the lowest portion of the velocity distribution of the interplanetary dust particles $> 100\,\mu$m in size incident on the Earth. These recovered particles represent a highly-biased sample of the total interplanetary dust population in this size range (FLYNN, 1990a). The extent to which this low-velocity sample of the 50 to $1200\,\mu$m interplanetary dust is representative of the composition and mineralogy of all the interplanetary dust in this size range can only be assessed by comparison of the polar micrometeorites with higher velocity particles in the same size range. Since these higher velocity particles are destroyed on atmospheric entry, this comparison will require either particle collection in space or analysis of the ion trails produced on vaporization. Such comparisons may be possible using orbital collection techniques as proposed for the Space Station Cosmic Dust Collection Facility.

2. Samples and Experimental Techniques

2.1. Samples

To examine the similarities and differences between the polar micrometeorites and both the IDPs and the chondritic meteorites, sixteen irregularly shaped polar micrometeorites were selected for analysis. Because of their irregular shapes these particles are not believed to have melted on atmospheric entry, and may, thus, better preserve the record of their pre-atmospheric chemical compositions and mineralogies than do the melted, spherical micrometeorites recovered from the polar ices and the sea floor.

Four of the micrometeorites studied were extracted from the cryoconite, a black dust collected from the bottom of the summer lakes which form in the melt zone of the Greenland ice cap (MAURETTE *et al.*, 1986, 1987). Most of this lake sediment consists of blue-green algae attached to mineral grains (MAURETTE *et al.*, 1986) but both melted spheres and irregularly shaped particles have also been recovered. Four irregular particles, ranging from 60 to $150\,\mu$m in size, from the MAURETTE *et al.* (1987) collection were selected for analysis.

Twelve of the micrometeorites are from a collection effort in which ~100 t of blue ice near Cap Prudhomme, Antarctica was melted using pressurized jets of water at

~80°C. The resulting water passed through three stainless steel filters (with 50, 100, and 400 μm openings), yielding a micrometeorite-rich dust (MAURETTE *et al.*, 1991). About 10 g of dust > 50 μm in size, including about 7500 irregular, friable particles and about 1500 melted spheres was recovered by Maurette *et al.* (1991). Twelve particles from the BI 54 collection were selected for analysis. They range from 50 to 150 μm in diameter.

2.2. *Element analyses*

Each particle was analyzed by Synchrotron X-Ray Fluorescence (SXRF) for trace element contents. SXRF is a non-destructive element analysis technique with a high sensitivity for trace elements between Fe and Br (SUTTON and FLYNN, 1988). These analyses were performed under the same experimental conditions previously employed to analyze smaller stratospheric IDPs (described in FLYNN and SUTTON, 1992a). The SXRF analyses provide element to Fe ratios for Ni, Cu, Zn, Ga, Ge, Se, Br, and Pb in each particle.

SXRF analyses were performed on the four Greenland micrometeorites after they were embedded in epoxy mounts following polishing for mineralogical analyses. The Br contents of the Greenland particles are uncertain because this epoxy contained Br. X-ray scattering in the epoxy also increased the background, precluding determination of the Se abundances in the four Greenland particles. Because of the possibility of elemental contamination during polishing, an improved experimental procedure was developed for subsequent measurements. The twelve Antarctic micrometeorites were analyzed by SXRF prior to epoxy mounting. These particles were mounted on a Kapton film using a small drop of silicone oil, a technique which has previously been shown to minimize background and contribute no significant peaks for the elements studied (SUTTON and FLYNN, 1988).

The SXRF X-ray beam was collimated by passing through a 8 μm diameter pinhole. This beam struck the sample at a 45° angle. The analyzed volume was about 10^4 cubic micrometers, or a few percent of the typical particle size. To test for element homogeneity, two fragments of one Antarctic micrometeorite (B1 #7) were analyzed separately. The abundances of Fe, Ni, Zn, and Pb in, one Greenland micrometeorite were mapped by stepping the 8 μm X-ray beam across the sample in 5 μm steps in a two-dimensional grid pattern, providing about 400 separate analyses of this sample.

The Fe abundances were measured on polished surfaces of the micrometeorites by electron microprobe, using a 20 μm defocused beam at 15 kV and 15 nA. These data were reduced using the Bence-Albee data reduction procedure. The Fe content of each particle was combined with the individual element to Fe ratios determined by SXRF to provide element concentrations. One Antarctic micrometeorite (B1 #2) was lost prior to the electron microprobe analysis. An Fe concentration of 18.5 %, the CI meteorite value, was assumed for this particle in order to determine concentrations from the element to Fe ratios measured by SXRF. The small fragment of micrometeorite B1 #7 has not been analyzed by electron microprobe. An Fe content of 27.5 %, equal to the Fe content of the large fragment of this particle, was assumed for the small fragment of B1 #7.

2.3. *Mineralogical identifications*

Analytical Transmission Electron Microscopy (ATEM) was employed to determine the mineralogies of these particles. Fragments from each micrometeorite were imbedded in epoxy and sectioned with a diamond knife using a Cambridge ultramicrotome. Section thicknesses range from 60 to 80 nm. Mineral analyses were performed with a Phillips CM20 Analytical Transmission Electron Microscope at 200 kV equipped with a Noran X-ray dispersive system (Ge detector). Electron probe diameters were on the order of 20 to 50 nm, depending on the size of the crystal to be analyzed. The Cliff-Lorimer technique was used for data reduction (CLIFF and LORIMER, 1975). Ultramicrotome thin sections of terrestrial and meteorite materials were produced to serve as thin film standards. The analytical uncertainties are $<5\%$ for the major elements, as determined by analyses of the mineral standards.

Fourteen of these polar micrometeorites have been studied by ATEM. Olivine, pyroxene, chromite, spinel, and magnetite were identified by composition, and glass was also present in some particles. Three of the particles were so fine-grained that mineral identifications were not possible.

3. Chemical Composition

3.1. *Compositions*

The element abundances determined for each of the polar micrometeorites are given in Table 1. One standard deviation errors due to counting statistics ranged from 0.1 to 15 %. Systematic errors are between 5 and 15 %. Thus, the overall uncertainties in the element concentrations reported in Table 1 range from 5 %, for Ni which is the most abundant element, to 20 %, for Ga, Ge, and Se which are least abundant. The two particles not analyzed by electron microprobe, Bl #2 and Bl #7 Sm, also have uncertainties because their Fe contents were assumed to be 18.5 % and 27.5 % respectively. The Fe contents in the other Antarctic particles range from 14.8 to 32.8 %, except for Bl #19 which is dominated by a single large pyroxene. The element concentrations in these two particles are uncertain to about 50 %.

Most elements agree to within ±20 % between the two fragments of Bl #7 (see Table 1), suggesting chemical homogeneity in this particle.

Further evidence of chemical homogeneity within individual particles comes from mapping the element distributions in Greenland particle #16. The Ni/Fe, Zn/Fe, and Pb/Fe ratios were relatively constant in the 400 individual analyses across the surface of this sample, indicating chemical homogeneity on the 10 μm scale within this particle.

However, the particles are generally distinct from each other in chemical composition, with each trace element showing more than an order-of-magnitude variation among the set of particles. This is consistent with the wide range of major element (Fe/Si, Mg/Si, Al/Si, and Ca/Si) abundances reported by MAURETTE *et al.* (1991) for similar Antarctic particles.

The wide variety of individual particle compositions makes it difficult to compare, on a particle by particle basis, individual polar micrometeorites with the IDPs or the various types of carbonaceous chondrite meteorites. To reconstruct the composition of the parent body (or parent bodies) of the polar micrometeorites, the compositions of

Table 1. Chemical contents and mineralogies of polar micrometeorites.

Particle	Fe %	Ni %	Cu ppm	Zn ppm	Ga ppm	Ge ppm	Se ppm	Br ppm	Pb ppm	Minerals[#]	Fa Olivine
Greenland											
#16	13.2	0.10	190	160	11	3	nd[@]	<3400[$]	1200	fg	—
#20	6.8	<0.002	5	8	<0.7	<0.5	nd	<940[$]	20	P, O, G	14–22
#21	5.8	0.03	39	50	1	4	nd	<3000[$]	190	fg, M	—
#22	11.9	0.14	100	80	17	6	nd	<1800[$]	1300	P, O, G, C, M	12–30
Avg. Green	9.4	0.07	84	75	7	3	—	<2280[$]	680		
Antarctic (BI 54 Sample)											
B1 #2	18.5[*]	0.21	690	2900	96	45	17	30	130	not analyzed	
B1 #4	26.9	0.03	780	2200	100	4	36	30	230	O, P, M, S	14–55
B1 #5	21.2	0.52	170	920	7	23	10	11	100	O, P, G, C, M, S	30–50
B1 #7 Lg[&]	27.5	0.43	670	870	13	5	5	3	210	O, P, M, G, S	44–50
B1 #7 Sm[&]	27.5[++]	0.46	510	600	15	3	4	6	210	not analyzed	
B1 #8	27.3	0.19	350	550	6	5	<2	4	150	O, P, M, G	30–68
B1 #17	21.4	0.25	630	2300	100	20	14	50	900	O, M, S	40–50
B1 #19	8.2	0.30	110	370	1	4	4	12	40	Enstatite, Metal	—
B1 #20	25.9	0.06	240	1100	20	42	9	21	130	fg	—
B1 #22	19.1	0.48	290	570	2	4	22	16	50	O, P, G	36–55[+]
B3 #1	14.8	0.11	3400	1500	28	36	5	48	440	O, M, C	55–67
B3 #2	32.8	0.14	1100	1700	35	5	13	52	890	O	8–56[***]
B3 #3	31.0	0.61	400	440	9	12	15	57	40	not analyzed	
Avg. Ant.	22.5	0.28	710	1220	33	16	12	26	270		
Average	19.5	0.23	560	950	27	13	12	26[**]	365		

[*] Fe assumed to be 18.5%, the CI value, in this particle not analyzed by electron microprobe.

[++] Fe assumed to be 27.5%, the value measured by electron microprobe in the large fragment of this particle.

[#] Identified by Analytical Transmission Electron Microscopy: O = olivine, P = pyroxene, C = chromite, M = magnetite, S = spinel, G = glass, fg = fine grained (< 20 nm) mineralogy not determined.

[@] nd = not detected.

[$] Br contents of Greenland micrometeorites include a contribution from the epoxy.

[&] B1 #7 broke into 2 fragments, denoted Lg and Sm, which were analyzed by SXRF separately.

[+] Large olivine grains were Fa 36–55, smaller olivine grains in glass were Fa 7–11.

[**] Average excludes Br in Greenland particles because of possible epoxy contribution.

[***] Large grains Fa 8 to 40, small grains Fa 21–56.

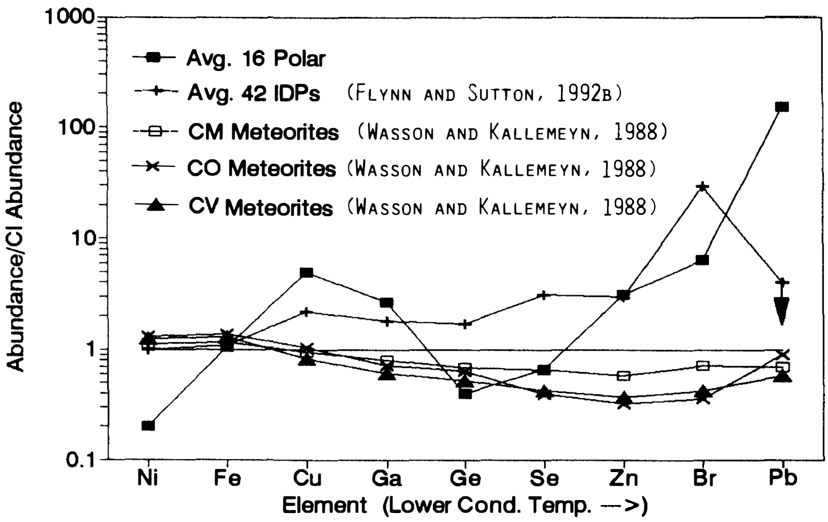

Fig. 2. Average CI normalized trace element abundances in the 16 polar micrometeorites, 42 IDPs, CI, CM, CO and CV carbonaceous chondrite meteorites. Elements are ordered with decreasing nebula condensation temperature (from Wasson*, 1985, except Br from* Fegley *and* Lewis*, 1979). Though the IDP and meteorite data show clear correlations of enrichment or depletion with nebula condensation temperature, the polar micrometeorite data shows no clear trend. The Pb content of IDPs has not been measured but the upper limit is < 10 ppm.*

the individual particles were averaged (see Table 1), as has been done previously for the IDPs (Flynn and Sutton, 1991b, 1992b). The CI-normalized average composition of the 16 polar micrometeorites is plotted in Fig. 2 and compared with the average compositions of 42 stratospheric IDPs and the CI, CM, CV, and CO carbonaceous chondrite meteorites.

3.2. *Volatile element contents*

The major differences in composition between the types of carbonaceous chondrites occur for those volatile elements with the lowest nebula condensation temperatures (see Fig. 2). Each of the volatile elements Ga, Ge, Se, Zn, and Br has a progressively lower abundance in moving from CI to CM to CV-CO chondrites. In addition Ga, Ge, Se, and Zn show progressively lower abundances correlated with lower nebula condensation temperatures within each chondrite group.

The polar micrometeorites are enriched above the CI meteorite concentrations in the volatiles Cu, Zn, Ga, Br, and Pb. These enrichments, except Pb, are comparable to the enrichments measured in forty-two 5 to 35 μm diameter chondritic IDPs collected from the stratosphere (Flynn and Sutton, 1992b). However the polar micrometeorites are depleted relative to the CI composition in Ge and Se, both of which are enriched relative to CI in the IDPs (Flynn and Sutton, 1992b). The Ge and Se contents of the polar micrometeorites compare with those in CM, CV and CO meteorites.

Since there is no obvious correlation of element enrichment or depletion in the polar micrometeorites with nebula condensation temperature, though clear correlations are seen for this set of elements in both the IDPs and the CM, CV, and CO carbonaceous chondrites (see Fig. 2), the average element abundance pattern measured in the polar micrometeorites seems not to result from a simple nebula condensation effect.

The enrichments above the CI abundances measured for Cu, Zn, Ga, Br, and Pb in these polar micrometeorites are not derivable from CI starting material by thermal alteration, as might result from atmospheric entry heating, since heating would decrease the volatile content relative to the more refractory elements. In addition, the element abundance pattern in the polar micrometeorites does not match the one reported for heated stratospheric IDPs, which show large depletions in Zn relative to Zn abundance in CI meteorites (Flynn and Sutton, 1992a).

Although the average element abundance pattern measured in these polar micrometeorites does not match the bulk pattern of either the IDPs or any type of carbonaceous chondrite, the shape of the pattern matches quite well with one previously reported by Ireland *et al.* (1990) for a < 1 μm size separate of the CV3 meteorite Allende as shown in Fig. 3. This Allende separate is dominated by olivine, but the grain size is so small that Ireland *et al.* (1990) were unable to identify individual minor phases. They suggest the unusual pattern in the < 1 μm Allende separate may result from the addition of disaggregated fine-grained calcium aluminum-rich inclusion material to Allende matrix material, explaining the increased abundances of refractory and volatile elements, and the loss of sulfides, explaining the Ni, Co, and Se depletion. (Ireland *et al.*, 1990). Similarly, the trace element abundance pattern of the polar micrometeorites might result from the alteration of material similar in composition to the IDPs or chondritic meteorite matrix by the loss of Ni, Ge, and Se-bearing sulfides and in the case of chondritic matrix the addition of a volatile-rich component.

We cannot rule out alteration of the element abundance pattern by either element addition through contamination by trace species from the polar ices or element loss through dissolution of water soluble phases while the micrometeorites resided in the water, snow or ice. Contamination may also occur if aerosol particles become attached

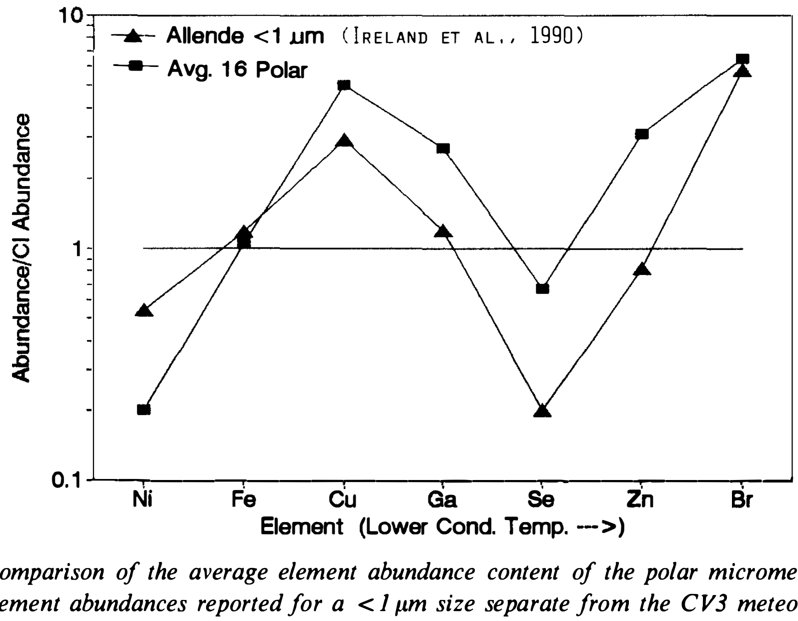

Fig. 3 Comparison of the average element abundance content of the polar micrometeorites with element abundances reported for a < 1 μm size separate from the CV3 meteorite Allende. Germanium and Pb have been excluded from this diagram since Ireland *et al. (1990) did not measure these elements in the Allende separate.*

to the surfaces of the micrometeorites.

Although the concentrations of most of these trace elements are low in polar ices, there is direct evidence for contamination of Antarctic meteorites by halogens (LANGENAUER and KRAHENBUHL, 1992) and terrestrial Pb (MISAWA *et al.*, 1992). The concentrations of major elements have been measured in a variety of snow and ice samples recovered from the Antarctic. One study showed $Al < 40$ ppb, $Na < 700$ ppb, $K < 16$ ppb, $Ca < 44$ ppb, and $Cl < 1400$ ppb in all snow samples from James Ross Island analyzed by ARISTAIN *et al.* (1982). These elements are assumed to be added to the snow and ice by precipitation of atmospheric aerosols (ARISTAIN *et al.*, 1982). Direct measurement of the compositions of aerosols collected in the Antarctic indicates the aerosol concentrations of the elements measured in the micrometeorites are generally much lower than the Al or Ca concentration. CUNNINGHAM and ZOLLER (1988) measured $Cu/Al = 0.1$, $Zn/Al = 0.05$, $Se/Al = 0.01$, and $Br/Al = 1$ in Antarctic aerosols sampled in 1975 and 1976, suggesting the concentrations of most of these elements in Antarctic ice are only a few ppb. Nonetheless, bulk contamination of small samples with long residence times in the ice seems possible. LANGENAUER and KRAHENBUHL (1992) have reported depth profiles of halogens in two Antarctic H-5 chondrites, showing surface contamination of F, Cl, Br, and I extending several millimeters into the interior.

Element loss can occur when water soluble phases dissolve. KALLEMEYN *et al.* (1991) have suggested that the depletions of siderophile and chalcophile elements observed in some weathered Antarctic CK meteorites may result from the dissolution of sulfide phases such as pentlandite. Although there is no direct evidence for element loss in the polar micrometeorites, they would be expected to be more susceptible to bulk chemical alterations by this mechanism than the larger Antarctic meteorites because the surface area to volume ratio of the micrometeorites is much greater than for the meteorites.

3.3. Ni content

The polar micrometeorites are significantly depleted in Ni, which is present at approximately the CI concentration in the IDPs and the carbonaceous chondrites. The pattern of element depletions (low Ni, Ge, and Se relative to CI) is suggestive of metal or sulfide loss.

Nickel is a major element in chondritic materials, present at the 1.1 % level. Previous major element analyses of larger sets of polar micrometeorites (MAURETTE *et al.*, 1987, 1991) and meteoritic deep sea spheres (BROWNLEE, 1985) have also reported this Ni depletion relative to CI, as well as a S depletion.

BROWNLEE (1985) has suggested an inertial separation mechanism, in which a metallic nugget separates from the less dense silicate liquid as the particle melts on atmospheric entry, to explain the low Ni content of melted meteoritic spheres collected from the ocean floor. However, melted micrometeorites $\sim 10\,\mu m$ in diameter, recovered from the Earth's stratosphere, show approximately chondritic Ni contents, but significant volatile depletions (FLYNN *et al.*, 1992b). This suggests that, at least for these smaller particles, melting during atmospheric entry does not give rise to Ni loss. The observation of substantial Ni-depletions in polar micrometeorites with irregular shapes, high volatile contents, and unequilibrated mineral compositions (see Section 4.3), all indicative that these particles were not melted on atmospheric entry, suggests some mechanism other than inertial separation is responsible for the low Ni contents of the polar micrometeorites.

The bulk Fe content of the eleven Antarctic particles averages 1.2 × CI (see Table 2). This indicates any Fe-metal or Fe-sulfide loss from the Antarctic particles we examined is likely to have been small. The four Greenland particles, on the other hand, all show Fe depletions from CI, with the average Fe content being only 0.5 × CI. Thus Fe-metal or Fe-sulfide loss is a possibility in the Greenland set of particles.

Alternatively, the present Ni abundance may represent the pre-atmospheric composition of the particles. PRESPER and PALME (1991) have suggested that chondrules, which have low Ni contents, may be the precursors to some melted cosmic spherules. The high volatile contents of all but one of the sixteen polar micrometeorites analyzed in this study are inconsistent with chondrule-like starting material for these particles. We cannot exclude the possibility that some of the melted spheres, not analyzed in this study, are remelted chondrules.

The low Ni content of the polar micrometeorites may indicate terrestrial alteration, such as the loss of Ni-bearing sulfides during residence in the polar ices. The highly weathered CK meteorites including Maralinga, Cook 003 and some CK meteorites from Antarctica are depleted in Ni relative to the unweathered CK meteorite Karoonda (KALLEMEYN *et al.*, 1991; GEIGER *et al.*, 1992). KALLEMEYN *et al.* (1991) suggest the exceptional sensitivity of Ni in CK meteorites to weathering indicates the Ni in these meteorites is mainly concentrated in a soluble phase such as pentlandite, which is removed by weathering. The depletion of Ni in the polar micrometeorites might, similarly, be a result of sulfide loss. The S content of polar micrometeorites is known to be low (MAURETTE *et al.*, 1991), consistent with sulfide loss. Sulfide loss might also explain the low Ge and Se contents of the recovered polar micrometeorites.

3.4. *Zinc depletions*

Atmospheric entry deceleration results in the heating of IDPs to temperatures which depend on the particle size, density, entry velocity, entry angle, and emissivity (see *e.g.*, FLYNN, 1989a). The low S contents of some IDPs recovered from the stratosphere have been attributed to S loss during atmospheric entry heating (see SCHRAMM *et al.*, 1989 for discussion). FLYNN (1989b) suggested that Zn loss might be a useful indicator of significant atmospheric entry heating, since the Zn loss temperature reported for chondritic meteorites was ~600°C (IKRAMUDDIN *et al.*, 1977). FLYNN and SUTTON (1992a) later identified a set of stratospheric IDPs which were chondritic in major and trace elements except for an order-of-magnitude Zn depletion. FLYNN *et al.* (1992a) and THOMAS *et al.* (1992) have observed correlations between these Zn-depletions and the presence of magnetite rims, produced during atmospheric entry heating, in the IDPs. Thus, in the stratospheric IDPs, Zn depletions seem to be a good indicator of significant particle heating on atmospheric entry.

Atmospheric entry heating calculations indicate that the typical 100 μm interplanetary dust particle should be heated about 500 K higher than the typical 10 μm interplanetary dust particle on Earth atmospheric entry (FLYNN and MCKAY, 1990). Thus we expected to find a significant number of Zn-depleted polar micrometeorites, at least comparable to the ~22% (4 of 18) of stratospheric IDPs found to have significant Zn depletions (FLYNN and SUTTON, 1992a). The heated textures observed by ATEM in many of these particles (see Section 4.3) seem to confirm that many of the polar micrometeorites experienced significant atmospheric entry heating.

Only one of the polar micrometeorites analyzed in this study, #20 from Greenland, shows a substantial depletion in Zn. If the Zn content of these particles has not been altered by contamination during residence in the polar ices, the lack of Zn depletions suggests the abundances we measured for the other volatile elements have not been altered significantly by atmospheric entry heating since order-of-magnitude Zn depletions are observed in the stratospheric particles before any of the other volatile elements we measure by SXRF show significant deviations from CI abundances (FLYNN and SUTTON, 1992a).

The absence of Zn depletions in all but one of these polar micrometeorites is a puzzle. One possibility is that Zn was depleted on atmospheric entry, but that Zn has subsequently been added to these polar micrometeorites by interactions with the polar ices. Alternatively, the Zn in the polar micrometeorites could be present in a different mineral phase (or phases) than in the smaller stratospheric IDPs, and might thus be lost from these particles at a significantly different temperature than from the IDPs.

3.5. *Pb content*

Each of the sixteen polar micrometeorites analyzed contained a very high concentration of Pb, ranging from 8 to 240 times the CI abundance. The high Pb content of the Greenland and Antarctic micrometeorites is distinct from both the stratospheric IDPs and the chondritic meteorites.

The Pb content of the particles may be altered by interactions with the polar ice/water during their long residence times or by the process of extraction from the ice. This led us to suggest that a significant fraction of the Pb might be contamination from the ice/water (FLYNN *et al.*, 1991). However, the Pb distribution in Greenland micrometeorite #16 was mapped by scanning the SXRF X-ray beam across the sample in a two dimensional grid pattern. The 8 μm diameter X-ray beam was stepped across the sample in 5 μm increments, dividing the sample into about 400 individual spots. The Pb/Fe ratio was approximately constant across the section of the ~60 μm particle, with no evidence for Pb concentrations near the particle surface. If the Pb is contamination it appears to be relatively uniformly distributed throughout the micrometeorite.

The uniformity of the Pb concentration does not, however, rule out contamination during residence in the Antarctic ice. Contamination by substantial abundances of terrestrial Pb is common in interior samples of meteorites recovered from the Antarctic (MISAWA *et al.*, 1992). Although stepwise acid leaching can eliminate the Pb contamination in meteorites this procedure may also extract indigenous elements (MISAWA *et al.*, 1992). No acid leaching was performed on the polar micrometeorites in this study in order to avoid alteration of the abundances of the other elements measured. Because of the known Pb contamination of the larger Antarctic meteorites, the Pb contents measured on the polar micrometeorites are likely to include some contamination.

3.6. *Differences between Greenland and Antarctic particles*

Although the sample set is small, the Antarctic micrometeorites appear to have substantially higher concentrations, by factors of 2 to 16, of the volatile elements Cu, Zn, Ga, Ge, and Se than do the Greenland micrometeorites. Zinc, for example, is more abundant in each of the 12 Antarctic micrometeorites than in the most Zn-rich of the Greenland micrometeorites. The average Zn content of the set of Antarctic micrometeorites analyzed is 16 times that of the set of Greenland micrometeorites.

The average element abundance patterns in the Greenland and Antarctic micrometeorites are quite similar to each other in shape (see Fig. 4). However the Greenland and Antarctic micrometeorites are distinct from each other in that for all these elements except Br and Pb the abundances are significantly lower in the Greenland particles than in the Antarctic particles. The true Br abundance in the Greenland particles is not known because of the high Br content of the epoxy in which those particles were analyzed.

The similarities in the patterns between the Greenland and the Antarctic micrometeorites are particularly significant because their recovery procedures differed dramatically. The Greenland particles were extracted from cryoconite, a deposit at the bottom of Blue Ice lakes (Maurette *et al.*, 1986, 1987). The Antarctic micrometeorites, on the other hand, were extracted by melting the frozen ice and filtering the resulting water. Since the two recovery procedures were so different, it seems likely that the unusual element abundance patterns were not produced by contamination or leaching during the particle collection procedures.

If the small differences measured between the two samples prove to be real, they may reflect time variations in the composition of the micrometeorite flux at Earth. Maurette *et al.* (1987) estimate that the Greenland micrometeorites fell within the past 3000 years. The Antarctic micrometeorites are believed to have fallen to earth

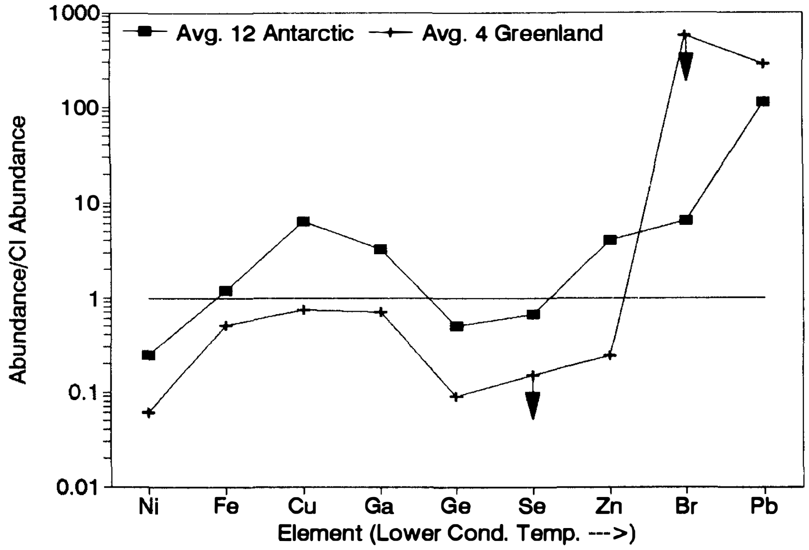

Fig. 4 Comparison of the average composition of the Greenland micrometeorites with the Antarctic micrometeorites shows similar enrichment and depletion patterns in both groups, though the Antarctic micrometeorites have higher volatile contents than the Greenland particles analyzed in this study.

more recently. MAURETTE (personal communication, 1992) estimates that 75 % of the Antarctic micrometeorites he collected were recent falls, within the past 25 years, while the remaining 25 % fell within the last 1000 years. A time variation in the composition of the micrometeorite flux was previously suggested by TAYLOR and BROWNLEE (1991) to explain major element compositional differences in melted meteoritic spheres of a similar size range.

3.7. Comparison with IDPs and meteorites

The low Ni content of the polar micrometeorites distinguishes them from both the chondritic IDPs and all types of unaltered chondritic meteorites.

The polar micrometeorites are enriched in the volatile elements Cu, Zn, Ga, and Br to approximately the levels previously reported by FLYNN and SUTTON (1992b) for the chondritic IDPs. This distinguishes the polar micrometeorites from even the most volatile-rich meteorites, the CI carbonaceous chondrites.

The Ge and Se contents of the polar micrometeorites are depleted relative to CI, falling in the range of the CM2, CO3 and CV3 meteorites. These Ge and Se contents are almost an order-of-magnitude lower than the average levels reported for the IDPs (FLYNN and SUTTON, 1992b). The high Zn content suggests that the polar micrometeorites could not be derived from IDP-like starting material which was thermally altered by atmospheric entry heating, because Zn is observed to be depleted more easily by entry heating of IDPs than are Ge and Se (FLYNN and SUTTON, 1992a). The loss of soluble sulfide phases during residence in the polar ice and water may explain the low contents of Ni, Ge, and Se.

The present chemical evidence is compatible with the polar micrometeorites being a new type of extraterrestrial material, but the possibility of terrestrial alteration of the element abundance pattern has not been ruled out.

4. Mineralogy

4.1. Layer-lattice silicates

Layer-lattice silicates are a major mineral phase in ~37 % of the IDPs recovered from the Earth's stratosphere (SCHRAMM *et al.*, 1989) and in the CI and CM carbonaceous chondrite meteorites. None of the thirteen polar micrometeorites analyzed by ATEM contained recognizable hydrated phases (see Table 1), such as the serpentines and smectites which occur frequently in IDPs. Three of the thirteen particles, however, are so fine-grained (grains$\ll$20 nm) that specific mineralogical identifications were not possible, so we cannot exclude the possibility that fine-grained particles or fine-grained areas in some particles might be relics of thermally altered hydrated phases

In a mineralogical study of 41 porous Antarctic micrometeorites BECKERLING *et al.* (1992) found textural similarities in several of the particles to the CI and CM meteorites (*e.g.*, magnetite framboids and sulfides) but reported no identification of hydrated phases. ALEXANDER *et al.* (1992) have examined 20 Antarctic micrometeorites, in the 50 to 110 μm size range, by infrared absorption. None of these infrared transmission spectra were dominated by the layer-lattice silicate absorption feature (ALEXANDER *et al.*, 1992), although this feature dominates the infrared transmission spectra of about 50 % of the IDPs studied by the same technique. Thus layer-lattice silicates seem to be much rarer in the polar micrometeorites than in either the IDPs or the CI and CM carbonaceous chondrites.

The absence of layer-lattice silicates may indicate that the polar micrometeorites experienced a different history, without the aqueous alteration that characterizes the CI and CM carbonaceous chondrites and a large fraction of the IDPs. Alternatively, the polar micrometeorites may have experienced significant thermal alteration during atmospheric entry. SANDFORD and BRADLEY (1990) have shown that layer silicates in IDPs are altered by pulse heating, as can be experienced on atmospheric entry, at temperatures near 600°C. Entry heating modeling by FLYNN and McKAY (1990) indicates that ~99 % of the 100 μm diameter interplanetary dust encountering the Earth are heated above 600°C on atmospheric entry.

4.2. *Anhydrous minerals*

Mineral assemblages in the coarse grained particles (grains 30 to 200 nm) consist of euhedral crystals of high-Ca and low-Ca pyroxenes, olivines, chromites, spinels, magnetites, and glass. The individual phases identified in each particle are indicated in Table 1.

Two olivine populations have been identified so far in these particles: low-Fe olivines (Fa 7 to 25), which usually occur as small crystals (< 100 nm) embedded in glass, and high-Fe olivines with Fe/Mg ratios from Fa 30 to 60. The Fe content of the individually analyzed olivine crystals in eight fine-grained Antarctic particles are shown in Fig. 5.

Mineral grains in Greenland particles #20 and #22 are approximately 100 nm and consist of olivines, pyroxenes, chromites, and magnetites embedded in glass.

Magnetite-rich crusts were observed on the Greenland micrometeorites (ROBIN *et al.*, 1990) and in natural and artificially produced meteorite fusion crusts (BROWNLEE *et al.*, 1975). Studies of stratospheric IDPs have shown that one mineralogical indicator of atmospheric entry heating is the development of a magnetite rim on the particle (KELLER *et al.*, 1992). In addition, in pulse heating experiments we observe that magnetites form within 25 s by heating layer-lattice silicates to 1100°C. This 25 s interval is comparable to the duration of the thermal spike experienced by a 100 μm micrometeorite on atmospheric entry (FLYNN, 1989a; LOVE and BROWNLEE, 1991). Two of the Greenland micrometeorites and six of the Antarctic micrometeorites in this study contain magnetite (see Table 1) which, along with some of the olivine, may have been formed during entry heating.

However some particles contain unequilibrated olivines. For example particle B3 #2 contains μm-sized olivines of Fa 8 and ranging from Fa 29 to Fa 40 (see Table 1 and Fig. 5). The core and rim compositions of the zoned grain in particle B3 #2 are shown in Fig. 5. Olivines < 100 nm in size in B3 #2 have compositions ranging up to Fa 70. Particle B1 #4 contains enstatite, high-Ca pyroxene, pyroxene high in Fe (Fs 20) and olivines from Fa 14 to 55 in contact with each other. This indicates these particles were not heated to high enough temperatures or for long enough times for their minerals to equilibrate.

Several particles consist of high-Fe olivines set in a glassy matrix. Olivines and

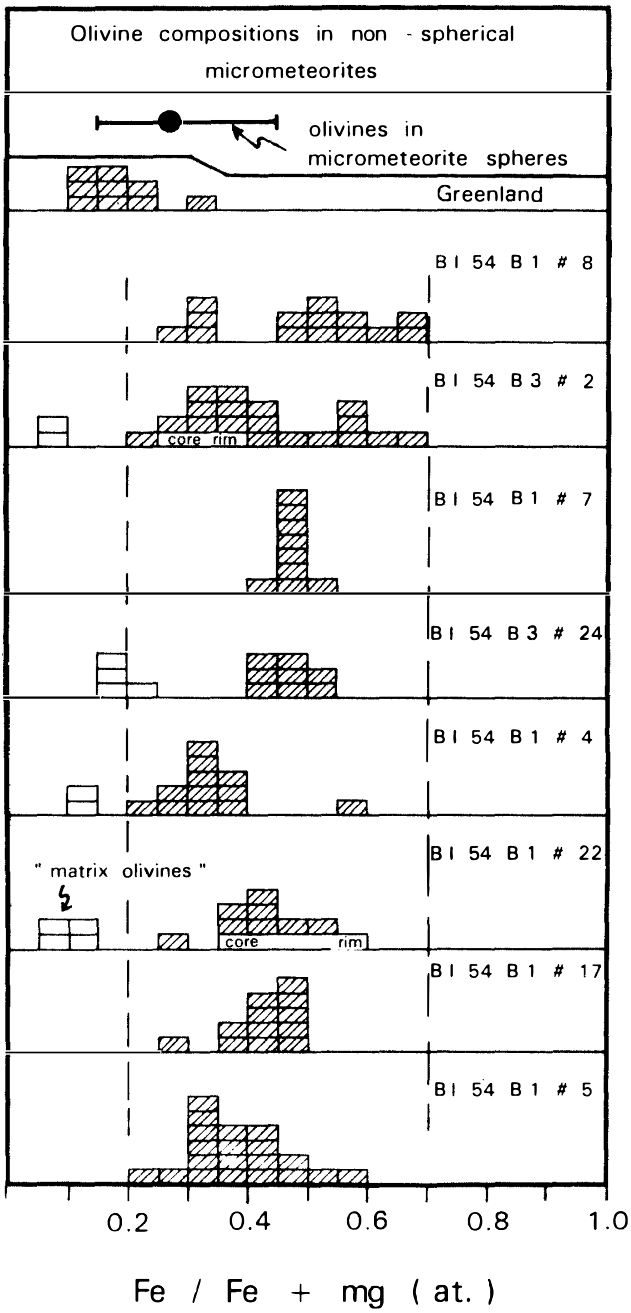

Fig. 5 *Histogram of the Fa contents of the olivines in 8 fine-grained, probably unmelted, micrometeorites from Antarctica. Compositions of small (approximately 100 nm) euhedral olivines are shown as hatched boxes. Olivines imbedded in glass are indicated as unshaded boxes and are labeled "matrix olivines." In two cases μm-sized olivines were found and the compositions of their cores and rims are indicated. For comparison, the range of compositions of olivines (excluding relic olivines) in melted micrometeorites is indicated and compositions of individual 100 nm olivines in two unmelted Greenland particles are shown. Except for the "matrix olivines" the range of olivine compositions in the unmelted Antarctic particles is between Fa 20 and Fa 67. The olivines in the two unmelted Greenland particles are less Fe-rich. Olivines in the melted micrometeorites are generally lower in Fe than olivines in the unmelted Antarctic particles. One particle, B3 #24, which has not been analyzed by SXRF is shown.*

magnetites often contain minor amounts of Ni (ranging from 0.2 wt%, the detection limit, up to 1 wt%), and these particles have textures indicating high temperature metamorphism. The Ni contents of olivines in primitive carbonaceous chondrites are usually only a few ppm. The presence of Ni in olivines is known from studies on CK meteorites to be an indicator of thermometamorphic processes (GEIGER *et al.*, 1992). We suspect these particles were heated significantly on atmospheric entry, and that the glass which formed during this short thermal event was quenched and had no time to recrystallize.

4.3. *Porosity*

The polar micrometeorites appear in thin section to be relatively compact objects, similar to the matrix of carbonaceous chondrites. Many of the smaller (5 to 30 μm) chondritic micrometeorites collected from the stratosphere of the Earth, when thin sectioned and examined in the Transmission Electron Microscope, exhibit large voids (MACKINNON *et al.*, 1987). FLYNN and SUTTON (1991a) report that the densities of 13 of 23 stratospheric IDPs fall in the range from 0.3 to 1.1 g/cm^3, also suggestive of large voids in their interiors. The absence of these large voids distinguishes the polar micrometeorites from many of the anhydrous, porous IDPs which constitute about 45% of the stratospheric IDP collection (SCHRAMM *et al.*, 1989). Highly porous micrometeorites have not yet been identified among the particles collected in Greenland and Antarctica. This could indicate that the volatile-rich, porous IDPs have higher geocentric velocities than the more compact particles, resulting in the larger porous IDPs melting or vaporizing on atmospheric entry. Alternatively, larger porous IDPs may fragment into smaller particles by the dynamic pressure experienced on atmospheric entry, or they may be more easily broken down during residence in or collection from the polar ices.

4.4. *Comparison with IDPs and meteorites*

The fine-grained portions of CI and CM2 carbonaceous chondrite meteorites consist predominantly of hydrated phases. The absence of layer-lattice silicates in the samples examined distinguishes the polar micrometeorites from the hydrated IDPs and hydrated carbonaceous chondrite matrices.

Matrices of the CO3 and CV3 carbonaceous chondrite meteorites are generally dominated by high-Fe olivines in the μm size range. SCOTT *et al.* (1988) indicate that olivines in the CO3 meteorites range from Fa 30 to Fa 60, while olivines in most of the CV3 meteorites range from Fa 40 to Fa 60. Within the CV3 group there is a monotonic increase in the homogeneity of the matrix olivine composition from Kaba to Allende suggestive of progressive thermal metamorphism acting on an initially heterogeneous olivine population (SCOTT *et al.*, 1988). Kaba and, to a lesser extent, Mokoia contain some low-Fe olivines, while the olivines in Vigarano, Grosnaja, and Allende are more equilibrated clustering around Fa 50 (SCOTT *et al.*, 1988). The low-Fe olivines found in several of the polar micrometeorites (see Fig. 5), thus, distinguish these particles from the equilibrated CV3 meteorites like Vigarano, Grosnaja, and Allende.

Though the polar micrometeorites appear to be texturally and compositionally distinct from primitive carbonaceous chondrite meteorites, we cannot, at present, exclude the possibility that they represent thermally altered hydrated carbonaceous chondrite matrix material.

The polar micrometeorites studied are texturally unlike the primitive anhydrous IDPs but show some similarities to heated IDPs with respect to their grain sizes, porosity, mineral assemblages and olivine compositions.

5. Conclusions

The average composition of the polar micrometeorites is enriched above CI meteorite concentrations for the volatiles Cu, Zn, Ga, and Br by amounts comparable to the enrichments measured in the 5 to 30 μm chondritic IDPs collected from the Earth's stratosphere. However the polar micrometeorites are depleted relative to CI in Ge and Se, both of which are enriched relative to the CI meteorite concentrations in the IDPs. The polar micrometeorites are also depleted in Ni, which is present at approximately the CI meteorite concentration in the IDPs. In accord with earlier major element measurements by MAURETTE *et al.* (1991) we find similarities in the trace element contents between the polar micrometeorites and both the IDPs and the carbonaceous chondrite meteorites, but the polar micrometeorites are not identical in chemical composition to either type of material. The present chemical data are consistent with these large micrometeorites being a new type of solar system material.

Because aqueous alterations giving rise to both the addition and the removal of various elements (particularly the addition of Pb and the removal of Ni-bearing sulfides) from the polar micrometeorites are possible, convincing identification of the polar micrometeorites as a new and distinct type of solar system material may have to wait until the recovery of large micrometeorites either from the stratosphere or in space (*e.g.*, by the proposed Cosmic Dust Collection Facility on the Space Station).

The cause of the large Ni depletion measured in almost all polar micrometeorites (avg. Ni/Fe = $0.2 \times \mathrm{CI}$ for this set of particles) remains unexplained. Stratospheric particles, even those with low Zn abundances suggestive of significant heating on atmospheric entry, have Ni abundances consistent with the CI concentration (FLYNN and SUTTON, 1992a). Thus Ni loss during atmospheric entry seems an unlikely explanation. The low Ni content may reflect the pre-atmospheric composition, but the loss of Ni-bearing sulfides due to terrestrial weathering is also possible. The trace element composition of the polar micrometeorites is consistent with a starting material having an IDP composition from which Ni, Ge, and Se have been removed, perhaps by the weathering of sulfides.

The mineral assemblages of the polar micrometeorites are different from the primitive (CI and CM) carbonaceous chondrite matrices which consist mainly of hydrated phases such as smectite, serpentine, and tochilinite, and from the ~37% of the stratospheric IDPs dominated by layer-lattice silicates. Thermal alteration of hydrated phases during atmospheric entry heating have not been well studied, and these alterations might explain the observed differences in mineral assemblages between hydrated meteorite matrices and polar micrometeorites. Laboratory simulations of these effects are required to determine if the polar micrometeorites might be alteration products from IDP or chondritic meteorite starting material.

The range of compositions and mineralogies of irregularly shaped polar micrometeorites indicates that main-belt asteroids contribute a diverse variety of particles to the interplanetary dust cloud.

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