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A study on cosmic dust particles in Antarctic ice, snow and non-Antarctic region and their origins*

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Abstract A large number of cosmic dust particles, micrometeorites and volcanic dust bands have been found and collected in Antarctic ice, snow and glacial sediments, especially in meteorite concentrated regions. Extraterrestrial spherules also have been discovered from the stratosphere and deep-sea sediments. On the basis of their distributive characteristics the cosmic dust particles are classified into interplanetary dust particles and interstellar dust particles. According to their origins cosmic dust particles can be divided into cometary origin particles, asteroidal origin particles, ablation particles from meteorites and interstellar origin particles. The criteria for identifying cosmic dust particles have been established and the origins of cosmic dust particles are also discussed in this paper.

Key words Antarctica, cosmic dust particles, interplanetary dust particles, interstellar dust particles.

1 Introduction

Since the first magnetic spherules in deep-sea sediments was discovered about 100 years ago (Murray and Renard, 1891), a number of dust particles have been collected from different terrestrial environments, most notably in deep-sea sediment, stratosphere, strata, sea water and area of high latitude. With the development of microquantitatively analytical technology of micrograins, and the study on the microtexture and chemical composition, the extraterrestrial origin of these dust particles was revealed. Recently, the detection of dust particles has been extended from the space near Earth to cosmic space, such as, the comprehensive studies of cometary flybys or remote observations, thermal emission from interplanetary dust cloud. More recently, new in-situ provided by the Long Duration Exposure Facility, the space station Mir, the Hilten spacecraft and the Galileo space probe. The question of interrelation between interstellar or circumstellar dust and interplanetary dust received much attention.

According to presented data a distinction can be made between dust particles in in-

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terstellar medium and the large amount of dust particles of solar system origin which are mainly derived from comets and from collisions in the asteroid belt and named extraterrestrial dust or cosmic dust. The particle lifetime in solar system is controlled by the Poynting-Robertson effect, the ratio of solar gravitational forces to radiation pressure, collisions among particles, and gravitational effect, principally from Jupiter and Saturn (Taylor, 1992). Thus, dust is lost from the interplanetary medium on timescales of 10^5 years due to Poynting-Robertson effect. As a result, there have must been constant fresh injection. Additionally, the concentration of dust particles in the stratosphere is enhenced by a factor of 10^6 over than in space due to the braking effect of the atmosphere (Taylor, 1992).

According to their origin and source region cosmic dusts can be divided into primitive and molten or differentiated groups. Primitive or undifferentiated dust particles include interplanetary dust particles, interstellar dust particles, cometary dust particles, and micrometeorites. The molten differentiated dust particles are mainly ablated from iron or stony meteorites during the meteorite's entry into the atmosphere. The interplanetary dust particles collected from atmosphere may provide the information of asteroids and comets in the solar system. The cosmic spherules collected in different terrestrial environments are classified into three groups: iron spherules, silicate spherules and glassy spherules, which are mainly products of meteoritic ablation. Recent years, some cosmic dust and volanic ash have been discovered in antarctic ice, snow, ice core and glacial sediment. In this paper we review the investigated results of these dust particles and their origins compared with dust particles collected in non-antarctic environment.

2 General characteristics of cosmic spherules

Table 1 shows cosmic dusts in ice, snow, glacial sediment, ice core and volcanic dust bands in the blue ice fields in Antarctica. Table 2 shows cosmic dusts in the terrestrial atmosphere, lunar surface and deep-sea sediment.

3 Evidences and identifications of extraterrestrial origin

Most of stratospheric particles that have been analyzed are compositionally analogous to CI or CM chondrites through the study of particles of extraterrestrial origin collected from a variety of environments. They mostly have high amounts of carbon, being enriched over CI abundances by a factor of 2～5. In comparison with solar abundances, the C/Si ratios of Halley dust particles, interplanetary dust particles, CI, CM and L chondrites are 0. 5, 0. 15, 0. 06, 0. 03 and 0. 001 respectively. Therefore, the interplanetary dust particles (IDPs) are closer in composition to solar than chondritic (Taylor, 1992). Studies of petrology, mineralogy, mid-infrared absorption spectra, Raman spectra and isotopic properties of IDPs show that IDPs appear to be less altered samples of nebular and prenebular materials than those found in meteorites. Most IDPs originate from comets and a part of dust particles consist of presolar interstellar-cloud material. IDPs exhibit both similarities and differences as compared with meteorites, but the properties of IDPs

Table 1. Cosmic dust particles in the ice, snow, glacial sediment, ice core and the volcanic dust bands in the blue ice fields in Antarctica

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
Glacial sediment	Beardmore glacial region, Trans-antarctic Mountains. Sediment from the so-called "Meteorite Moraine" close to Lewis Cliff ice field (125 ~ 500 μm in diameter, 1~4000 spherules per 100 g sediment)	Most spherules have a smooth surface. Some spherules display a characteristic brickwork pattern similar to some deep-sea sphere and Tunguska spherules. The brickwork pattern results from aligned olivine and magnetite on the surface. Large euhedral olivine crystals are intergrown with fine-grained dendritic magnetite. Glassy Fe-rich matrix is present. Matrix contains very fine-grained dendritic magnetite and rare accessory wustite. Most spherules have the same mineral compositions: forsterite crystals, glassy Fe-rich and Mg-poor matrix, and magnetite. Most olivine crystals are zoned, being Fe-rich at the rim and Fe-poor in the center. The rare earth element in some spherules has a flat pattern, which is characteristic of undifferentiated chondritic materials. They are enriched in Ir and other siderophile elements(with Ir contents ranging from 10 to >1000 ppb). Si, Mg, Al and Fe are very close to the chondritic abundance ratios. The REEs in individual spheres show a flat chondrite normalized pattern. Siderophile elements such as Ir or Os are enriched in some spherules compared to terrestrial materials. The high abundance of the Antarctic spherules in the glacial sediment is an important factor. Their abundance ranges up to several thousands of spherules per 100 g of bulk sediment. It indicates that a very efficient mechanism has been active in concentrating these spherules from glacial ice into glacial sediment. Glacial action and wind erosion-deposition may contribute to the concentration of the spherules.	Silicates and glassy spherules	Spherules ablated from chondrites or molten micrometeorites; REE pattern and Al/Si (wt%)-Ca/Si (wt%) plot	Koeberl and Hagen, 1989; Hagen *et al.*, 1990
Glacial ice	Meteorite ice field, Yamato Mountains	Black spherical microparticles, glassy microparticles and other irregular-shaped particles were collected from the glacial ice. The annual accretion rate of black spherical microparticles on the whole earth is estimated at 2.9×10^4 t/a to 5.0×10^4 t/a. (assuming that the accumulation rate of snow in the Yamato Mountains is 20 g/a and that the specific gravity of microparticles is 5 g/cm^3. These results agree with the annual fall rate of 3.0×10^4 t/a of cosmic dust calculated from the observation on Antarctic ice near Syowa Station. The irregular-shaped microparticle size is generally smaller than 100 μm, but some particles are a few hundred μm in diameter. Mineralogical composition is magnetite and goethite. Fe was the main component taking up about 60~65%.	Black spheric micro particles, glassy micropaticles and irregularshaped particles. Several kinds of silicate minerals, clay minerals and metal oxides are recognized. The sizes of them are less than 100 μm	Extraterrestrial origin	Yabuki *et al.*, 1976
Bare ice, surface snow and ice core	Allan Hills meteorite collection field (76° 40'S, 159° 20'E); ice core (at depth of 32 to 33.5 cm) and the surface snow at Mizuho Station (70° 42'S, 44°20'E)	CTS(Ca-Ti-rich type): Three of eight spherules from the Mizuho ice core have Ca-Ti rich composition similar to perovskite. The chondrite normalized REE abundance pattern shows that positive anomaly of Sm and Nd excess(~10 and 3 times respectively) those of the REE pattern for terrestrial perovskite. The Cr abundance is about 30 times as high as that of C1 chondrite and about 3000 times as high as that of terrestrial perovskite. The Fe abundance is times	CTS/FCN were found together with typical spherules supposed to be metero ablation debris(OSS/ OIS); OSS/ OIS generally found in	Extraterrestrial origin, mineralogy, trace element abundance and REE pattern	Tazawa, 1987

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
		lower than 10, relative to that of C1 chondrite. They are mineralogically composed of perovskite and wustite. FCN (Fe-Cr-Ni-rich type) from ice cores has a Fe-Cr-Ni-rich composition similar to oxidized stainless steel, but it has considerable amounts of Ca, Ti and light REEs which pattern is similar to that for CTS, but it is lowered by a factor of 30. It is mineralogically composed of magnetites (probably also chromite and trevorite). OSS (ordinary stone/silicate type) from ice cores are mafic (Mg-Fe-rich) silicate spherules composed of magnetite, wustite and olivine. OIS (ordinary iron/magnetite type) are Fe-rich spherules composed of magnetite and wustite. CAS (chondritic, Au-S-undepleted type) from Allan Hills bare ice have chemical composition similar to chondrite. They are rich in Fe, Ni and S, and poor in Mg, as compared with Allende C3 chondrte. CAS spherules are mineralogically composed of olivine and magnetite.	deep-sea sediment are mostly metero ablation debris; CAS are compositionally similar to chondrites.		
Air	Antarctica	Iron cosmic dust (10～30 μm in diameter) which is chemically composed of Fe, 50～70%; Ni, 1～15%; Mn, 0～0.5%; SiO_2, 5～10% and small amount of Cr, Zn and Cu; silicate cosmic dust (SiO_2, 60～70%; Fe, 1～10% and small amount of Al and Ti).			Shima *et al.*, 1972
Snow	Antarctica	Cosmic dust (>10 μm in diameter); The annual fall rate is estimated at (1.2～1.8) × 10^5 t/a.			
Ice	McMurdo Station and Syowa Station	Iron spherules (Fe, 50～80%; Ni, 0.1～20%; Mn, 0～1% and SiO_2 0～5%) containing Cu, Zn and Cr; silicate spherules (SiO_2, 50～70%; Fe, 0～5%) containing Ti and Al.			
Ice and snow	Greenland	Cosmic dust (>5 μm). The annual fall rate is estimated at 2 × 10^5 t/a～9.1 × 10^5 t/a.			
	Antarctica and Green land	The mineral composition of 250 micrometeorites have been studied. Olivine and low-calcium pyroxenes with crystals larger than 5 μm have been analyzed. Nearly 30% of the olivines have <5% Fa, the majority of crystals have 5 to 30% Fa, with a broad peak near 20～32% Fa and a few crystals have 50% Fa, such a distribution is similar to that of C-chondrites with a peak corresponding to H (Fa_{17-20}) and LL (Fa_{23-32}) groups of the ordinary chondrites. About 50% of the pyroxenes have <5% Fs (composition typical in carbonaceous chondrites), while the other 50% have 26% Fs with a peak corresponding to H (Fs_{15-18}) and L-LL (Fs_{19-24}) groups of the ordinary chondrites. Most of micrometeorites are highly unequilibrated materials which are similar to carbonaceous chondrites compositionally, but they are low in Ni and S and have different oxygen isotopic composition (forsterite, $\delta^{18}O = -2.9 \pm 0.6$, $\delta^{17}O = -2.5 \pm 1.0$; olivine, $\delta^{18}O = -2.0 \pm 1.4$, $\delta^{17}O = -2.7 \pm 1.4$).			Christophe *et al.*, 1992

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
Dust bands of volcanic origin	Lewis Cliff/Beardmore glacial area, Yamato Mountains and Allan Hills (three major meteorite accumulation areas)	Blue ice field in Atarctica, as a stranding zone of Antarctic meteorites, records numerous events since it formed. One of the most important structures on the ice fields are the dust bands which is mostly of volcanic origin and consists of small volcanic glass shards and lithic fragments. The magnificant ash layers are deposited at a distances of several thousands of kilometers from the volcanic source area. The dust layers have been found in ice cores from different stations and blue ice fields. Several sources are possible. The most important ones would be (1) material scraped from subglacial bedrock debris by the movement of the glacier; (2) volcanic material deposited on the ice; (3) cosmic particles falling as micrometeorites or meteorite ablation spherules, and (4) continental and marine dust and aerosols transported by wind. The grain size of Lewis Cliff dust ranges from about 2 to 25 μm, or mostly 2～10 μm in diameter. They are much smaller than dust particles in Allan Hills or Yamato Mountains, which are usually 10～100 μm in diameter. Lewis Cliff tephra consist predominantly of brown to black glass shards. Crystal fragment such as feldspar plagioclase, and olivine are also present as minor components. The glass consists of basanite, K-trachyte, and peralkaline K-trachyte, respectively. Some of these dust samples were found to contain iridium in concentation up to 7.5 ppb. Ir is in positive correlation with Se, both being rich in As, Sb and other volcanogenic elements. The Ir enrichment shows a surface effect and probably caused by condensation of vapor phase. It suggests that the Ir in the K/T boundary clay is not of cosmic origin, but may have originated from the mantle materials and may have been brought over during extensive volcanic eruptions triggered by impact events. Iridium enrichments have been found in volcanic exhalation and aerosols from the Kilanea volcano, Hawaii. Other volatiles and typical volcanogenic elements (like Se, Hg Cd, Au, As, Sb and the halogens) are also enriched in the aerosols. The Ir may be mobilized in form of a volatile fluorine complex together with some of other elements. Large grains collected from Lewis Cliff have been identified to be sedimentary materials, probably of the Beacon Supergroup formation. Volcanic ash in the Allan Hills is derived from the McMurdo volcanic group. The glass shards have trachybasaltic composition, and crystal fragments of plagio clase, feldspar, olivine. Vocanic materials collected from Yamato Mountains have a tholeiitic andesite composition.	Trachyte (subglacial bedrock), basanite, peralkaline K-trachyte	The dust bands a the blue ice field are mostly of volcanic origin and may be correlated with individual Cenozoic volcanoes in Antactica and the sub-Antarctic regions. Volcanic eruptions dispersed large quantities of volcanic dust over Antarctica, and fell on snow accumulation area, and was incorported in the ice, transported to ablation zones. The dust layers constitute an isochronous layer that is of great value for the study of the ice fields and probably for dating.	Koeberl, 1990

Table 2. Cosmic dust in the terrestrial atmosphere, lunar surface and deep-sea sediment

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
Stratosphere	U-2 plane, NASA, USA	Collected 5~50 μm particles, including both interplanetary dust and meteor ablation debris, have been classified into five groups: chondritic(60%), FSN(30%), mafic silicates(6%), Fe-Ni(3%) and others(1%). Chondritic particles are either aggregates(90%) of particles 100 μm or ablation debris (10%). Their elemental abundances are within a factor of 2 of chondritic meteorites for Fe, Mg, Si, C, S, Ca and Ni. Carbon ranges from 2~15% in these particles. The minerals of these chondritic aggregates include hydrated silicates, olivine, magnetite and pyrrhotite. The chondritic ablation particles are spherules or spheroidals, being once molten. Minerals include olivine and magnetite. The particles are rich in Fe, Ni and S. Sulfur content varies greatly as a result of deficient sulfur relative to stoichiometric FeS. FSN particles are sometimes intimately associated with the chondritic aggregates. The minerals include pyrrhotite, pentlandite, magnetite and wustite. Mafic silicates consist of olivine, pyroxene, magnetite and pyrrhotite. Fe-Ni particles are mostly spheurles, containing taenite, magnetite and wustite. Other particles contain a few chondritic silicate particles that have numerous Fe-Ni mounds on their surface. Most of stratospheric particles are compositionally analogous to CI or CM chondrites, but they have higher amounts of carbon, being enriched over CI abundances by a factor of 2~5. The C/Si ratios, in comparison with solar abundances, of Halley dust particles, interplanetary dust particles, CI, CM and L chondrites, are 0.5, 0.15, 0.06, 0.03 and 0.001 respectively. Thus, the interplanetary dust particles is compositionally closer to solar than the chondritic. There are two dominant groups among stratospheric particles. One group is referred to chondritic porous(CP) which are porous on a micrometeor scale. The other is smooth on a micrometeor scale and are distinguished as chondritic smooth(CS). The CP particles are perhaps samples of cometary dust. It is unlikely that they are derived from asteroids for their fragile nature. The CS particles are mostly hydrated silicates. The CP particles are mainly pyroxene while olivine particles are rare. The hydrated phase are serpentine. CS and CP particles range from 4 to 40 μm(mostly 6~15 μm) in size. It is likely that there are presolar grains among interplanetary dust particles. The best evidence for it is the very high D/H ratio with enrichment up to 2500 per mil, suggesting that they are interstellar materials.	Interplanetary dust particles, chondritic ablation dust particles	FSN and Fe-Ni spherules are chondritic ablation debris. Chondritic aggregates, mafic silicates and FSN are a kind of special ablation debris or undifferentiated inter planetary dust particles	Blanchard and Kyte, 1978
Atmosphere	Japan aeroplane and baloons at high altitudes 10~25 km	Cosmic dust(10~40 μm); iron spherules(Fe, 65~80%, Ni, 1.0~8.5%, Mn, 0.5~5%); silicate cosmic dust(SiO_2, 60~70%, Fe, 1~10%) other cosmic dust containing Ca, Al, Cu and Ti. The annual falls rate of cosmic dust is estimated at 10^6~10^7 t/a.	Black spherules, glassy spherules	Products of ablated iron meteorites, silicate spherules similar to microtektite.	Yabuki *et al.*, 1969

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
Lunar	Fine-grained material of lunar surface No. 1008489 and No. 12070	Irregular-shaped spherules contain ilmenite, metal Fe, troilite, glassy materials, pyroxene feld-spar and olivine. The diameter of metal spherules is 20～100 μm(Fe,85～95%, Ni,2～10%). Mineralogical composition is mostly of taenite,kamacite and metal Fe, but without wustite.	Meteoritic materials in lunar soil	Residual materials on the lunar surface shocked by meteorites	
Air	Norikura Cosmic-ray Observatory, Tokyo Univ.	Black spherules and some glassy spherules (10 ～ 70 μm). 517 spherules were collected. Large ilmenite spherules (400 μm) and glassy magnetite spherules were found. The annual fall rate of black spherules is calculated to be (3. 4～4. 0)$\times 10^7$ t/a.	Black spherules	Products of ablated iron meteorites	Shima *et al.*, 1972
Deep-sea sediment (depth >4500 m)	Hawaii Island	Magnetic spherules, whose sizes range from 16 μm to 1440 μm, have been classified into three categories: 1) rough, brittle and black spherules with specific gravities lower than 4. 0; 2) dull and black spherules with smooth surfaces, whose specific gravities are lower than 5. 0; and 3) shiny metallic grey or shiny black spherules with occasionally small vesicular cavities. These spherules have higher specific gravities(≥5). The ratios of Co/Fe, Ir/Fe and Au/Fe are similar to the thermally degenerated compositions of metal phase of ordinary chondrites and iron meteorites. In the largest sized spherules(approximately>50 μm) Fe content was determined to be approximately 68～70% in weight. This means that these spherules were entirely oxidized and changed into hematite (Fe_2O_3; Fe = 69. 9%) or magnetite (Fe_3O_4; Fe = 72. 4%). Co/Fe, Ir/Fe and Au/Fe ratios are calculated to be (4. 5～11. 4)$\times 10^{-3}$, (1. 7～31)$\times 10^{-6}$ and (1. 4～6. 3)$\times 10^{-6}$, respectively.	Black magnetic spherules	Many metal phases from the Bruderheim (L6) with these ratios as follows Co/Fe, (1. 3～8. 6)$\times 10^{-3}$; Ir/Fe, (0. 4～13. 2)$\times 10^{-6}$, Au/Fe, (0～6. 4)$\times 10^{-6}$; in iron meteorites, Co/Fe, (2. 9～12)$\times 10^{-3}$, Au/Fe, (0. 26～2. 6)$\times 10^{-6}$; bulk ratios of Co/Fe, Ir/Fe and Au/Fe in metal phase of ordinary chondrites are (6. 6～10. 2)$\times 10^{-3}$, (5. 3～10. 2)$\times 10^{-6}$, (1. 7～2. 6)$\times 10^{-6}$ respectively. after the melting and evaporating of iron meteorite in the vacuum and following the decrease of residual mass of the sample, Co/Fe and Ir/Fe gradually increased while Au/Fe ratio rapidly decreased.	Yamakoshi, 1985
	North Pacific Ocean	Iron spherules with golden Fe-Ni core and silicate spherules with golden Ni-Fe inclusion: Their grain-size was generally 100～200 μm and they have strong magnetic properties	Magnetic spherules	Products of ablated iron meteorites	Peng, *et al.*, 1985

Occurrence	Location	Structure, mineralogy and chemical composition	Type of spherules	Origin and indicator	References
Pleistocene sediments	Alberta, Canada	A large number of Ni-Fe alloy spherules, containing Ni from 27.63 up to ~100% in weight. Most spherules are Ni-Fe metals, a few have iron oxide rims and some have iron-rich dendrites in the main metal phase. The dominant mineral phase is taenite. The spherules appear to be similar in composition, morphology and size of Ni-Fe cores of cosmic spherules found in deep-sea sediments and polar ice. It is estimated that the accretion rate of cosmic Fe spherules to Earth was $\sim 8.9 \times 10^6$ t/a.	Magnetic spherules	The Ni-Fe alloy spherules represent the ejected Ni-Fe cores of cosmic spherules. The formation of these Ni-Fe alloy spherules must have involved melting, fractionation and ejection. A model for generation of Ni-Fe alloy spherules was proposed. A molten Fe-Ni metal spherule ($Ni < 10wt\%$) ablates from iron or iron-rich stony meteorites → Fe in the Fe-Ni metal begins to be oxidized and the nickel-rich metal core ($Ni > 10wt\%$) forms due to the atmospheric oxidation → with increasing oxidation, Ni continues to be concentrated in the shrinking Ni-Fe metal core ($Ni \gg 10$ wt%) and then Fe-Ni core may move off center or protrude from the spherule's surface.	Dong *et al.*, 1993
Primitive C chondrites, CR-chondrites	interstellar	Interstellar SiC, microdiamond, amorphous carbon, graphite and hydrocarbons from Orgueil, Murchison, CR and Semarkona primitive chondrites are characterized by large enrichments in ^{13}C, ^{14}N, ^{29}Si, ^{30}Si, $^{46,47,49,50}Ti$. Oxygen and Mg isotope anomalies in Orgueil corundum grain suggested that they are interstellar oxide grains. Corundum has an extreme ^{17}O excess. Red giant stars are enriched in ^{17}O with $^{17}O/^{18}O \geqslant 1$, Suggesting that these stars are likely source of the interstellar corundum. The source of these isotopic signature can be explained by the survival of presolar organic materials in these meteorites.	Interstellar grains, Carbon, Star, Red Giant Star explosion. Products of supernova	Several models have been proposed, including chemical vapor deposition; interstellar shock and UV-annealing of small graphite particles and high-energy particles produced from U and Th by irradiation, highly isotopic anomalies of some isotopes.	

do not fit into the framework established for meteorites, partly because of their tiny sizes ($\sim 10^{-3}$cm in diameter). The laboratory study of IDPs is in its infancy. Diagnostic marks of extraterrestrial origin of IDPs are as follows: (1) Iron spherules have high abundances of siderophile elements. Silicate spherules have chondritic compositions and the REE abundances. Chondritic particles generally are defined as those which have relative abundances of Mg, Al, S, Ca, Fe and Ni, measured with respect to Si, within a factor of 3 of those found in C-type meteorites. Some particles with strong depletions or enrichments of specific elements have been classified as chondritic by different investigators. Other particles consist primarily of Fe, Ni and S (FSN). Terrestrial particles from volcanic eruption are common. In addition, the correlation diagram between Al/Fe and Ca/Si (wt%) ratios, Co/Fe, Ir/Fe, Au/Fe ratios in metal phase, and Co/Fe, Au/Fe ratios in iron meteorites are compared with those of extraterrestrial origin dust particles. Fig. 1 shows correlation diagram between Al, Ca, and Si in extraterrestrial spherules as compared to chondrites. Fig. 2 shows chondrite-normalized REE plot for four individual spherules. All spherules have a flat and undifferentiated chondritic pattern (Koeberl and Hagen, 1989). The neutron activation analysis of silicate spherules from oceanic sediment indicates that among trace elements the rare earth elements provide effective tool to investigate the extraterrestrial silicate samples (Nagasawa *et al.*, 1980). In the terrestrial igneous processes REEs behave similarly with regular variations parallel to atomic number with an exception of Eu which shows anomalous behavior under reducing condtion. During cosmochemical processes evaporation of REE could take place under low-pressure, high temperature condition to produce fractionation accompanied by anomalies of Ce, Yb and possibly of Tm, in addition to that of Eu. The extraterrestrial materials have cosmo-

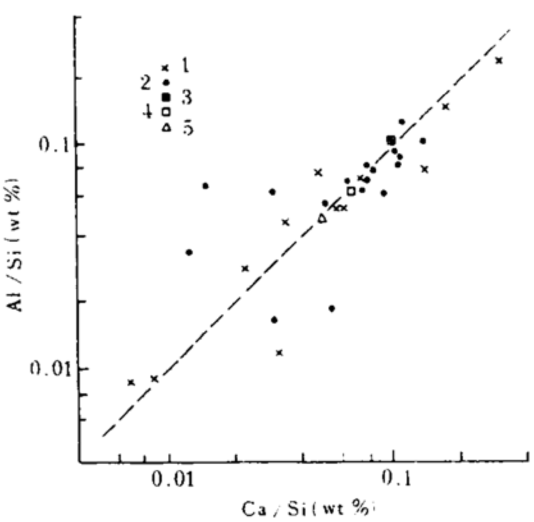

Fig. 1. Correlation diagram between Al, Ca and Si in extraterresial spherules as compared to chondrites. 1. Antarctic spherules; 2. Deep-sea spherules; 3. C1-chondrites; 4. Ordinary chondrites; 5. Enstatite chondrites.

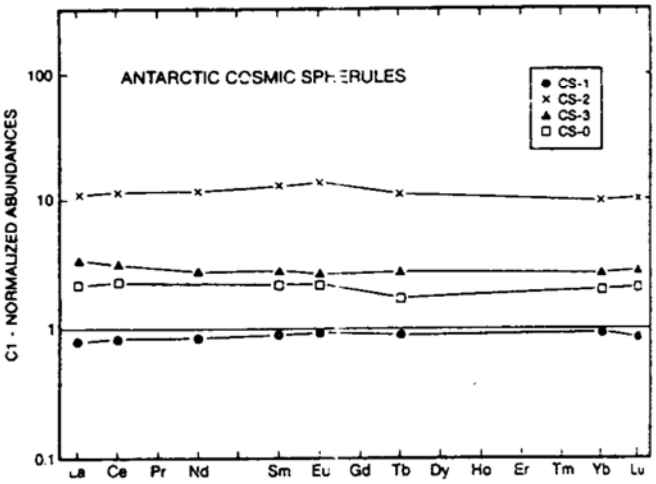

Fig. 2. Chondrite-nomalized REE plot for four individual spherules from the Transantarctic Mountains, Antarctica.

chemically fractionated chemical composition. Nagasawa *et al.* (1980) suggested the criteria for identification of cosmic spherules are: (a) similarity of REE abundance pattern to those in meteorites; (b) anomalies of Ce, (Eu), Tm and/or Yb, as a high temperature condensate in a primitive solar nebula or in a supernova, or an evaporation residue of meteoritic material or cosmic dust; (c) Irregular REE pattern indicating an extrasolar origin.

(2) Chondritic particles contain large concentrations of He similar to those found in lunar soil samples that have been exposed to the solar wind. High ^{4}He content ($>11.0\times10^{-11}$cm^3) are found among particles characterized by their porous texture and occurrence of unequilibrated mineral phases. These particles contain abundant glass, preserve solar flare tracks, and have chondritic Zn abundances. Brownlee *et al.* (1993) suggested that the measured He release profiles of step-heated IDPs provide a quantitative determination of maximum temperature experienced by an individual IDP during entry into atmosphere. Using the straightforward atmospheric entry model for dust, the temperature can be used to infer the entry velocity for particles with known mass and density. These techniques can distinguish the difference between typical asteroidal dust that enters the atmosphere at velocity near 12 km/s and typical cometary dust that enters the atmosphere at velocity above 15 km/s. For 10 μm IDPs with similar densities the temperature difference between comet dust and asteroid dust is in the range of 200～300 C or more. Evidence from the degree of entry heating and solar flare track densities suggested that a large fraction of the silicate IDPs recovered from the stratosphere are derived from main belt asteroidal parent bodies. In addition, IDPs enriched in Ne and Ar have been found.

(3) The determination of hydrogen isotopes indicates some IDPs have a high D/H ratios with enrichments up to 2500/cm^3. Such high values are relatively uncommon in solar system materials, but common in molecular clouds, apparently due to ion-molecule reactions (Taylor, 1992). Similar enrichments in meteorites such as Semarkona suggested the survival of interstellar materials. Hydrogen-isotopic measurements demonstrated the existence of large D enrichments in some particles, providing further evidence for their interplanetary origin.

(4) The high densities of fossil nuclear tracks in the silicate minerals within some particles were observed. These tracks are produced by the irradiation of the particles in space by heavy nuclear from solar flares (Fleischer *et al.*, 1975).

(5) Chondritic IDPs are dominated by either anhydrous minerals or layer-lattice silicates. The anhydrous group can be further divided into pyroxene and olivine the silicates minerals provide the best match for the observed intrared 10 μm spectral features. Anhydrous IDPs appear to be composed of single mineral grains, tar balls, a carbonaceous phase and glass. Olivine compositions vary in the form of $Fo_{40\sim100}$ and the grains occur (～0.05 to 1 μm in diameter) in the form of anhedral grain, euhedral crystal, platelets and spherical crystals. Pyroxenes include enstatite, hypersthene, fassaite, diopside and augite compositions (Bradley *et al.*, 1988). Other less common single mineral constituents of IDPs include magnetite, Fe-Ni alloy (kamacite), Fe-Ni carbides and chromite. Tar balls consist of aggregates of extremely small ($<$ 0.01～0.05 μm) round-

ed crystals embedded in a carbonaceous matrix and their overall compositions are chondritic, and possibly they are products of an accretional event prior to their incorporation into IDPs. Kamacite and olivine are the dominant minerals, together with minor amounts of Fe-rich sulfides. Carbonaceous material is widespread as a disordered material. Glass is also present in anhydrous IDPs and usually contains embedded olivine, pyroxene or Fe-rich sulfide grains. The layer-lattice silicates have low porosities and consist of pyroxene, olivine and Mg- and Fe-rich carbonates. These layer silicates have been identified as smectite and serpentine. Christofferson and Buseck(1986) identified a suite of refractory phase including Mg-Al spinel, anorthite and perovskite, a tetragonal Fe sulfide, high Sn grains, Al-, Si-, Ti- and Bi- oxides, SiC, Fe phosphide, Fe-Cr sulfide, FeOOH, and $BaSO_4$. Smectite is the dominant layer-lattice silicate in IDPs, whereas it is serpentine in CM carbonaceous chondrites(Barber, 1985).

(6) The study of the mid-infrared spectra of IDPs indicates that almost all the chondritic IDPs in the 2.5 to 25 μm ($4000\sim400$ cm^{-1}) spectral region have a dominant absorption feature at ~10 μm(1000 cm^{-1}). The position and shape of this band varies from particle to particle, but three broad grouping are evident. These spectral groups have been labeled olivine, pyroxenes and layer-lattice silicates. The observed relative abundance of olivine, pyroxenes and layer-lattice silicte particles is roughly 1 : 1 : 2 respectively(Fig. 3). The spectra of IDPs in layer-lattice silicate group have additional bands at 3.0, 3.4, 6.0, 6.8, 7.9 and 12.5 μm(3300, 2940, 1670, 1470, 1270 and 800 cm^{-1}, respectively). A weak band near 11.4 μm(880 cm^{-1}) is also seen in some spectra. The bands centered at 3.0 and 6.0 μm are characteristic of terrestrial layer-lattice silicates and are attributed to absorbed water. The 6.8 μm absorption band is due to CO_3. The carbonate identification is further demontrated by the presence of the 11.4 μm CO_3. The 7.9, 12.5 and possibly the 3.4 μm bands are characteristic of materials containing hydrocarbon functional groups. Unfortunately, the silicon oil used to collect the IDPs also exhibits features at the same locations, and the bands may be due to contamination. In spite of differences in the mineral structures, infrared spectra of IDPs in the layer-lattice silicate infrared groups are similar to those of CM carbonaceous chondrites.

(7) Ion-probe D/H measurements have now been made on 31 chondritic IDPs. A total of 13 of them have fragments with $\delta D > 100$ ‰ and 3 of them have parts with $\delta D \geqslant 1000$ ‰. FSN type exhibited a δD value of 173 ‰ suggestive of an extraterrestial origin (Bradley *et al.*, 1988).

As mentioned above, IDPs represent new classes of meteoritic material. Because of orbital decay driven by non-gravitational effects and lack of high ram pressures during atmospheric entry, the IDP population in the Earth's atmosphere contains samples of all asteroids and short period comets(Brownlee, 1993). Although most recovered IDPs are similar to carbonaceous chondrites which have several properties that distinguish them from existing meteorite groups. Typical IDPs have C/Si abundances ~ 3×CI values. The high C abundances of IDPs are beyond the provenance of carbonaceous chondrites and it is suggested that they are from the samples of C-rich matter such as that measued in Comet Halley. The volatile elements Ga, Ge, Cu, Se, Zn and Br are enhanced in most IDPs by a factor of about 3. Zn content reversely correlates with the degree of atmo-

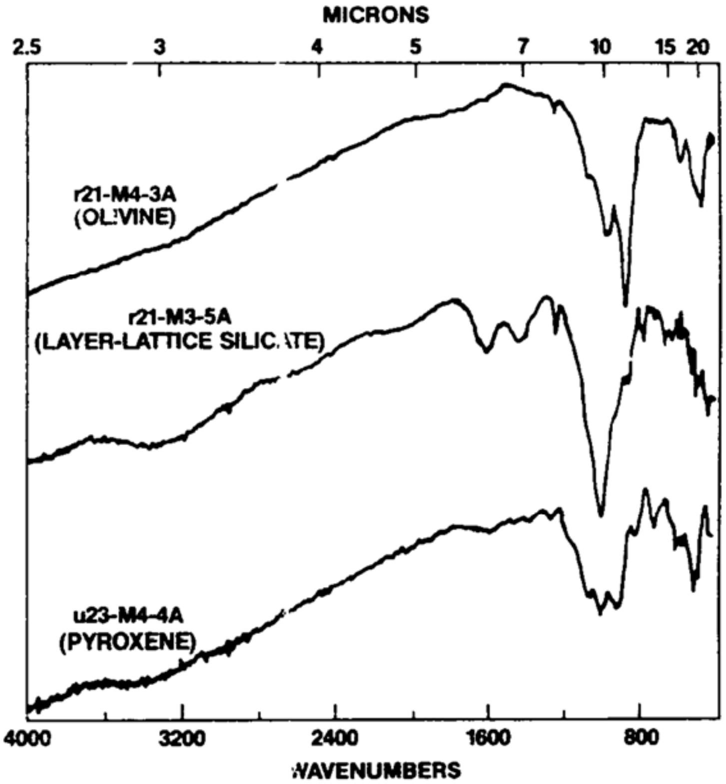

Fig. 3. Representative infrared transmission spectra of interplanetary dust particles(Sandford and Walker, 1985).

spheric entry heating. Indicators used are volatile(Zn, Ge, Ga and S) depletion, magnetite rim formation, infrared changes, layer silicate alteration, track erasure, carbon graphitization and the He loss. On the basis of He release curve about 20 % of the 10 μm IDPs collected are cometary and that >50 % are low velocity particles from main belt asteroids. This results agree with infrared astronomical satellite analysis implying that > 40 % of small particles at 1 AU are derived from asteroids.

4 Interstellar grains in primitive meteorites

Recent years, interstellar grains or presolar grains (such as SiC, microdiamond, graphite, TiC, corundum and organic matter) preserved in Orgueil and Murchison primitive carbonaceous chondrites have been found. Their interstellar origin is proven by highly anomalous isotopic ratios. They are stable only under highly reducing condition (C/O>1) and apparently are "stardust" formed in stellar atmosphere(Anders and Zinner, 1993). Microdiamonds with medium size about 1 nm are most aboundant(about 400 ~1800 ppm). They contain anomalous noble gases including Xe-HL, which is derived from supernova. Silicon carbide(0. 2~10 μm in size and ~6 ppm in abundance) apparently comes mainly from red giant carbon(AGB) stars of 1~3 solar masses. Some grains appear to be $\geqslant 10^9$ a older than the solar system. Graphite spherules, 0. 8~7 μm in size and <2 ppm in abundance contain highly anomalous C and noble gases, and large amounts of fossil ^{26}Mg. They seem to come from at least three sources, probably AGB stars, novae, and Wolf-Rayet stars. As we know, the elements are continually synthesized in stars, ejected into the interstellar medium, and then cycled back into next generation of stars, where the process is repeated. Thus isotopic composition of the elements vary in time and space and serve as marks of exotic material. In the early 1960s, Rowe and Kuroda (1965) reported that some meteorites had enrichments of the Xe isotopes 129 and 131～136 from decay of the extinct radionuclides $^{129}I(t_{1/2}=16\ Ma)$ and $^{244}Pu(t_{1/2}=82\ Ma)$. Apparently some young material synthesized only prior to about 200 Ma had been added to the early solar system. During the 1970s, relict of interstellar material was about 4% enrichment of ^{16}O in the most refractory phases of primitive meteorites, calcium-aluminum-rich inclusions (CAI). Other isotopic anomalies were found for Mg, Si, Ca, Ti, etc. The magnitude of these anomalies generally is much smaller than expected in fresh stardust and implies that the grains were severely reprocessed and diluted in the solar nebula, leaving only an isotopic fingerprint. The primitive interstellar grains were the fine- grained, low-temperature matrix. Four C-bearing grain types have been discovered since 1987, and all are chemically quite resistant, permitting them to be isolated by dissolving meteorites in acids (Tang and Anders, 1988). A fifth type, Al_2O_3 is resistant, but not C-bearing. Other candidates have been found (e. g. Si_3N_4), but their interstellar origin has not yet been established. The exotic noble gases have been found in meteorites (Anders and Zinner, 1993). They are as follows: (1) Xe-HL is enriched in isotopes ^{124}Xe and ^{136}Xe. Their host phase is diamond (C_δ) with grain size of 0.001 μm, and abundance of 400 ppm in C2 chondrites. The $^{12}C/^{13}C$ and $^{14}N/^{15}N$ ratios which are relative solar ratios are 1.04 and 1.52 respectively; (2) Xe-S is enriched in isotopes ^{130}Xe. Their host is silicon carbide (SiC) or C_β with grain size of 0.03～10 μm and abundance of 7 ppm. The $^{12}C/^{13}C$ and $^{14}N/^{15}N$ ratio are 0.02～28 and 0.05～60 respectively; (3) Ne-E(L) is enriched in isotopes ^{22}Ne. Their host phase is graphite (C_α) with grains size of 0.08～7 μm and abundance of <2 ppm. The $^{12}C/^{13}C$ and $^{14}N/^{15}N$ ratios are 0.02～80 and 0.15～1.8 respectively; (4) Ne-E(L) is enriched in isotopes ^{22}Ne. Their host phase is SiC or Cε with grain size of 0.03～10 μm and abundance of 7 ppm. The $^{12}C/^{13}C$ and $^{14}N/^{15}N$ ratios are 0.02～60 and 0.05～60 respectively. Diamond is enriched in heavy and light isotopes and is therefore called Xe-HL. Xe-S has a characteristic of the s-process (neutron capture on a slow time scale). It is believed to take place in red giants. Ne-E(L) has somewhat higher $^{20}Ne/^{22}Ne$ ratios. Ne-E(L) has low $^{20}Ne/^{22}Ne$ ratios and is nearly mono-isotopic ^{22}Ne. Titanium carbide has been found only as inclusions within graphite spherules and SiC crystals (Anders and Zinner, 1993). Only two interstellar corundum grains have been identified so far. They have large $^{26}Al/^{27}Al$ ratios, large excesses in ^{17}O, and smaller deficits in ^{18}O (Huss *et al.*, 1993a, 1993b). Up to now, presolar grains have been investigated in a considerable number of primitive meteorites. The diamond/silicon carbide ratio is not constant (Fig. 4). Russell *et al.* (1992) pionted out that the enstatite chondrite Indarch appears to be particularly enriched in SiC as compared to its diamond content, whereas the CV3s are relatively poor in SiC. The abundance of SiC in CV3 chondrites seems to depend strongly on the oxidation state. The highly oxidized Allende has much less SiC than the more reduced Vigarano. This differences imply either heterogeneity in the solar nebula or different destruction mechanisms for the two components. The abundance of both diamond and silicon carbide in primitive meteorites decrease with increasing petrologic type. The nitrogen content of the diamond varies considerably in a way which might be dependent on petrologic type. Interstellar grains, which have been found in primitive meteorites, provide a new and valuable source of astronomical information, complementing traditional methods of astronomy and astrophysics. They also provide important information of chemical processes in both stellar atmosphere and the interstellar medium.

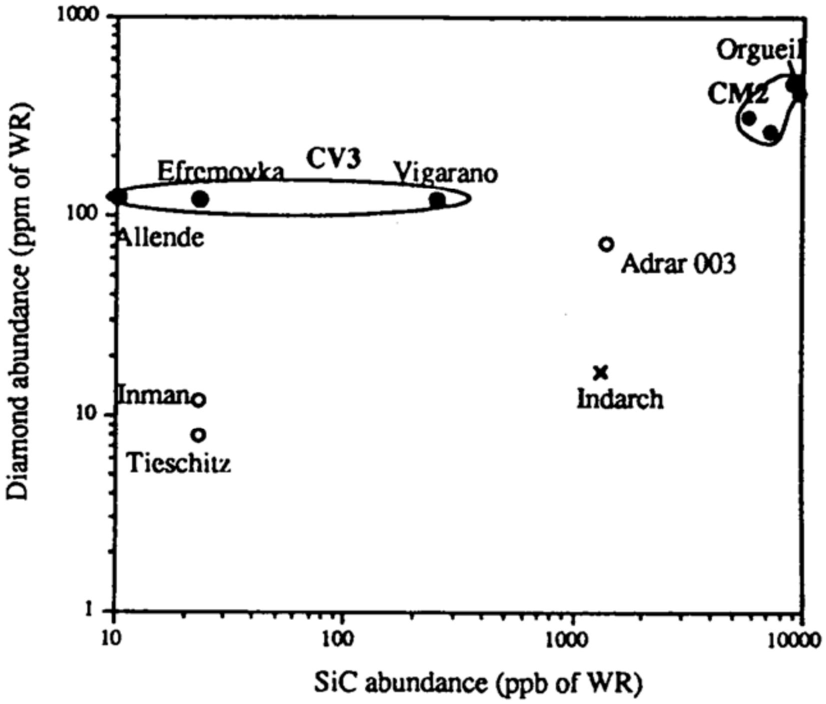

Fig. 4. Diamond/silicate carbide ratio. The enstatite chondrite Indarch appears to be particularly enriched in SiC as compared to its diamond content, whereas the CV3s are relatively SiC poor.

5 Discussion

Interplanetary dust particles are responsible for zodiacal light and gegenschein, the Infrared Astronomical Satellite(IRAs) dust bands, meteors and impact pits on lunar rock surfaces. The IDPs have also been detected by space probes and they have been shown to persist out to at least 18 AU(Humes, 1980). Dohnanyi(1978) calculated the Poynting-Robertson effect and particle-particle collisions limit ~10 μm dust particle in size to lifetimes of ~10^4 a in the inner solar system. Thus, the dust cloud presently observed in the solar system is not primordial, but must either be a transient phenomenon or be continuously replenished by one or more sources. Whipple(1967) calculated that a total production rate of ~8 t/s is required to maintain the present cloud. Although there are a number of possible sources for the IDPs, the major ones are comets, asteroids, Sun, planets with their rings and moons, and interstellar medium. Comet dust must be an important constituent of interplanetary dust complex, for example, comet Kohoutek was observed to lose ~2×10^{13} g of solid material inside 2 AU(Ney,1982) of which approximately one half is expected to remain within the solar system. Since the total mass of the zodiacal dust cloud is ~10^{19} g, it should take 10000 comet passages like that of comet Kohoutek to replenish the cloud. Therefore,the passages of one such comet evey five years would provide enough dust. Some calculations suggeted that even a single comet(comet Encke) was situated in a unique short-period orbit. The cometary origin is supported by in-situ measurements made by spacecrafts. The orbital parameters of dust near 1 AU as determined by experiments on Pioneer 8,10 and 11 are best fit by cometary sources(Leinert *et al.* ,1983) . Some meteor showers are also associated with the orbits of some well-known comets. For example, the Orionid and Aquarid meteoroid streams are associated with Halleys comet, the Taurids with comet Encke. New information about comet dust has been gathered by the several spacecrafts that made flyby of comet Halley. These probes found that Halley was producing particles with mass at least as low as 10^{-16} g each(Vaisberg *et al.* , 1986) and was injecting approximately 3×10^{6} g/s of dust into the interplanetary medium. Many of grains had roughly chondritic elemental abundances, but some particles consisted of O,Mg,Si and Fe, and these are probably individual silicate mineral grains. Other particles were rich in H,C,N and O, suggesting an organic composition. Various mixtures of different compositional types(H,C,O,N; H,C,N;H,C,O;H,C) were also found. The carbon and nitrogen abundances in the Halley material appear to be much higher than in any of chondrites. The C/Mg and N/Mg in Halley dust are enhanced relative to CI by factors 6 to 12 and 6 to 8 respectively. These particles are dominated by C,H,O and N(Brownlee,1990).

Asteroids are also the sources of interplanetary dust particles. The strongest evidence for it comes from the results of the Infrared Asteronomical Satellite(IRAs) which indicated band of dust running continuously around the solar system(Low *et al.* ,1984). The bands have color temperature of 165 to 200 K, which places them at the position of the main asteroid belt and suggests that dust is derived from asteroid-asteroid collisions (Sykes and Greenberg,1986).

The study of solar-flare tracks may make it possible to determine the fractions which come from asteroids and comets. The density of tracks produced in IDPs depends on its space-exposure time, its distance from the Sun and the flux of track-producing solar-flare nuclei. All of these factors are function of the orbit of a dust particles. Particle from an asteroidal source should have an narrow range of non-zero track densities at 1 AU, while cometary particles should display a wide range of track densities that extend to lower values. This is because asteroidal particles will undergo space exposure before arriving at 1 AU in near circular orbits. Comets produce dust in highly elliptical orbits that originally cross the Earth's orbit.

The evolutional model from the interstellar to the interplanetay medium is as follows (Greenberg,1991): interstellar medium → dense clouds → protoplanetary nebular dust → aggregation → $20\text{ K}<T_0<100\text{ K}$, comets formation, preservation of all volátiles(ice, oganic refractories, amorphous anhydous silicate), production of comet dust (zodiacal particles, IDPs and meteors) → $100\text{ K}<T_0<500\text{ K}$, formation of asteroids and asteroidal dust particle (zodiacal particles, IDPs and meteors), preservation of H_2O and other volatiles at low T_0, only silicates and less volatile fraction of organic refractories, and perhaps recondensation and reprocesses of ice, crystalline silicates(hydrated and anhydrous) → meteorites(C1, C2, C3). The organic matter decreases and silicate crystallinity increases with the aggregation temperature T_0. In addition, collisions between asteroids are also contributed to zodiacal particles, IDPs and meteors.

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