Senior Thesis

Description, Chemical Composition, and Origin of six Microscopic Spherules collected from the Meteorite Moraine in Antarctica; an SEM study

by
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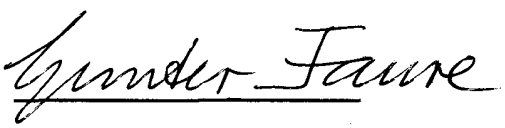
Approved by:



Dr. Gunter Faure

Abstract

Microscopic spherules (100-1000 micrometers) have been found in glacial moraine deposits in Antarctica. Six spherules from the Meteorite Moraine of the Lewis Cliff ice tongue were analyzed using a scanning electron microscope equipped with an energy dispersive X-ray analyzer. Both photomicrographs and chemical analyses were obtained. Calibration factors, based on four mineral standards, were used to convert peak-to-background ratios of most of the elements (Ti, Fe, Mn, Ca, Mg, Na, K, and Cr) to percent concentrations. The concentrations of Si, Al, and O were determined by the software used by the analyzer.

The results of the analyses were compared both to average chemical compositions of CI chondrites and bulk continental crust. All of the spherules were found to be chemically similar to CI chondrites. From this I concluded that the sperules have an extraterrestrial origin and that they are most likely debris from the ablation of meteorites traveling through the atmosphere of the earth.

Introduction

Microscopic extraterrestrial spherules have been found in many environments, including deep-sea sediment and meltwater lakes on the ice sheet of Greenland. Most notably, they have been found in Antarctica because this environment allows the best preservation of the spherules because the East Antarctic Ice Sheet contains no liquid water.

Spherules found in deep-sea sediments have been described by Blanchard et al. (1980). They showed that the spherules have a similar chemical composition to "laboratory-created ablation debris" and debris collected from the stratosphere; and concluded that the sperules formed from ablation of carbonaceous chondrites as they entered the atmosphere of the earth.

Hagen et al. (1989) described spherules they had found in glacial sediment in Antarctica. They concluded that the spherules were micrometeorites or ablation debris of larger meteorites.

This report describes six spherules recovered from glacial sediment in the Meteorite Moraine of the Lewis Cliff ice tongue in Antarctica and presents their chemical compositions in order to determine their origin.

Description of the Spherules

Photomicrographs of the spherules (Plates 1-9) were taken on a scanning electron microscope (see Methods section) using an accelerating voltage of 20,000 electron volts. The working distance was maximized to attain a high depth of field and, in some instances, a stage tilt was used to obtain more desirable photographic angles.

Six spherules from the Meteorite Moraine at the Lewis Cliff ice tongue, collected by John Schutt, range in size from 350 to 650 micrometers (Faure, 1994, personal communication). Spherules MM-1-A and D are nonmagnetic, whereas spherules MM-1-B, C. E, and F are magnetic indicating the presence of magnetite. The textures of the spherules range from smooth and glassy to irregular and crystalline. Spherules MM-1-C (Plates 4 and 5) and MM-1-E (Plates 7 and 8) exhibit a "brickwork" structure which was previously described by Blanchard et al. (1980) and Hagen et al. (1989). The crystals in the brickwork structure are most likely olivine and magnetite (Blanchard et al., 1980). Spherule MM-1-A (Plates 1 and 2) shows remnants of a possible "fusion crust" that has since flaked off (Hagen et al., 1989). Some of the spherules have dimples that may be "cupules", which are features left by the loss of dense material during the formation of the spherules (Hagen et al., 1989). The irregular surfaces of some of the spherules may be the result of the loss of the "fusion crust" or erosion.

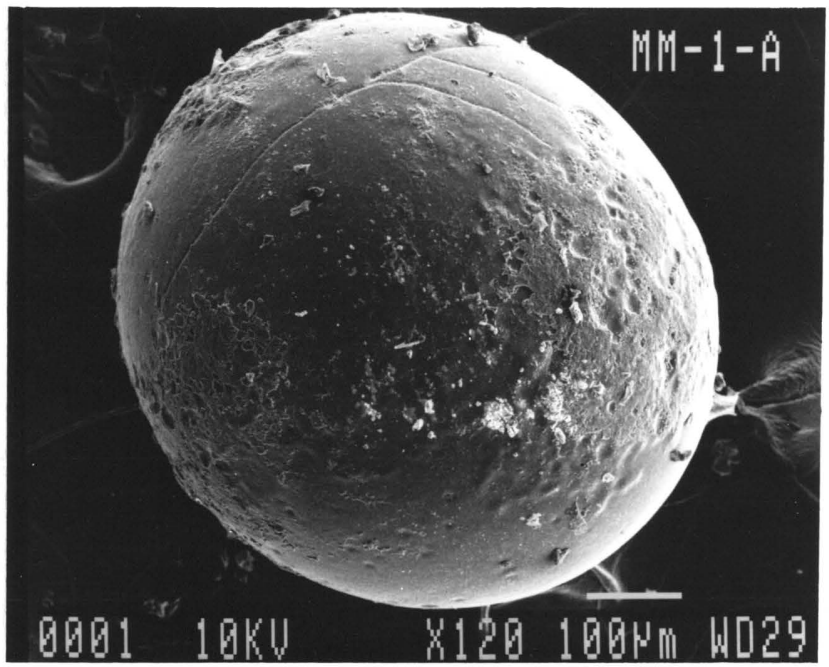


Plate 1.

Spherule MM-1-A showing glassy surface. Remnants of a "fusion crust"(?) still remain. (magnification 120x, marker bar is 100 micrometers)

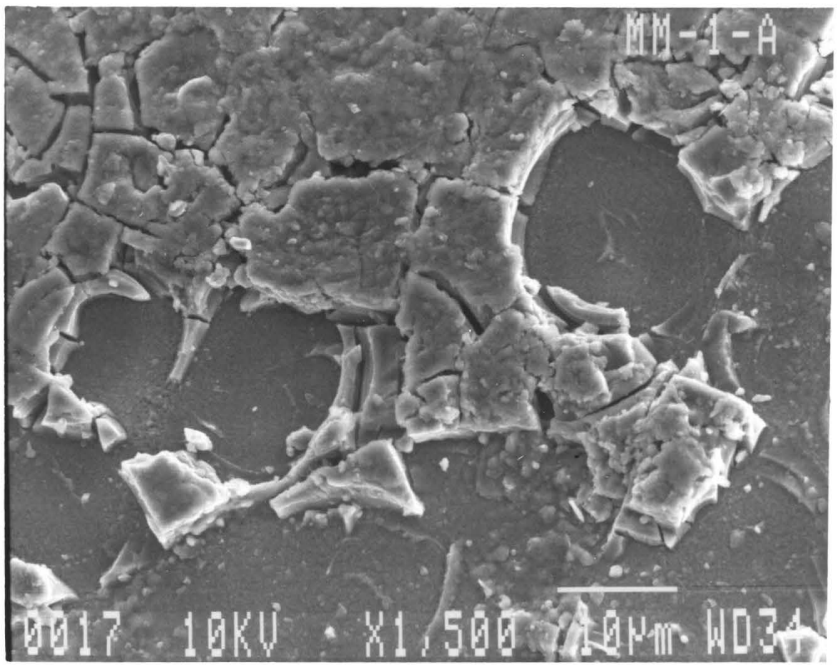


Plate 2.

Close-up of "fusion crust"(?) on spherule MM-1-A. Crustal material displays polygonal cracking. (magnification 1500x, marker bar is 10 micrometers)

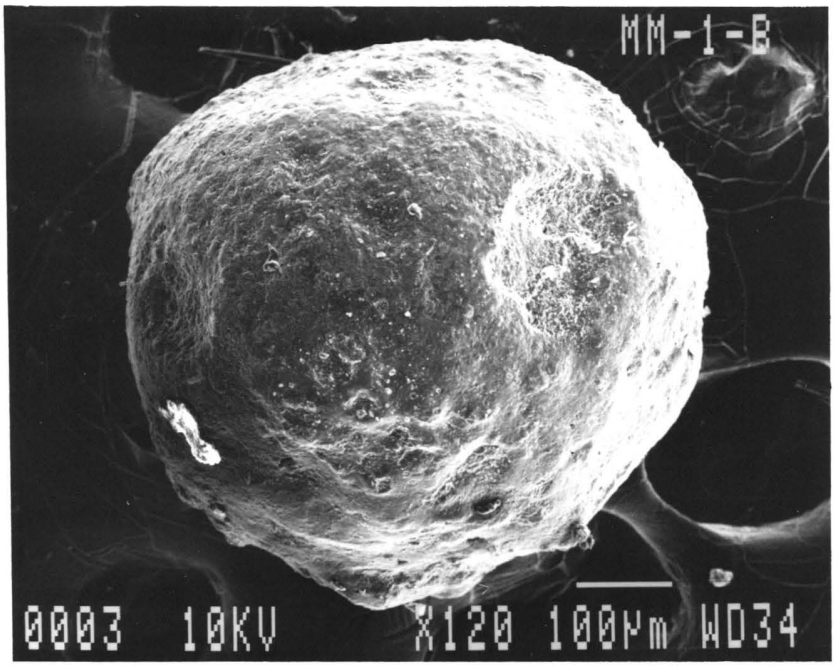


Plate 3.

Spherule MM-1-B has a very irregular surface and contains a dimple. (magnification 120x, marker bar is 100 micrometers)

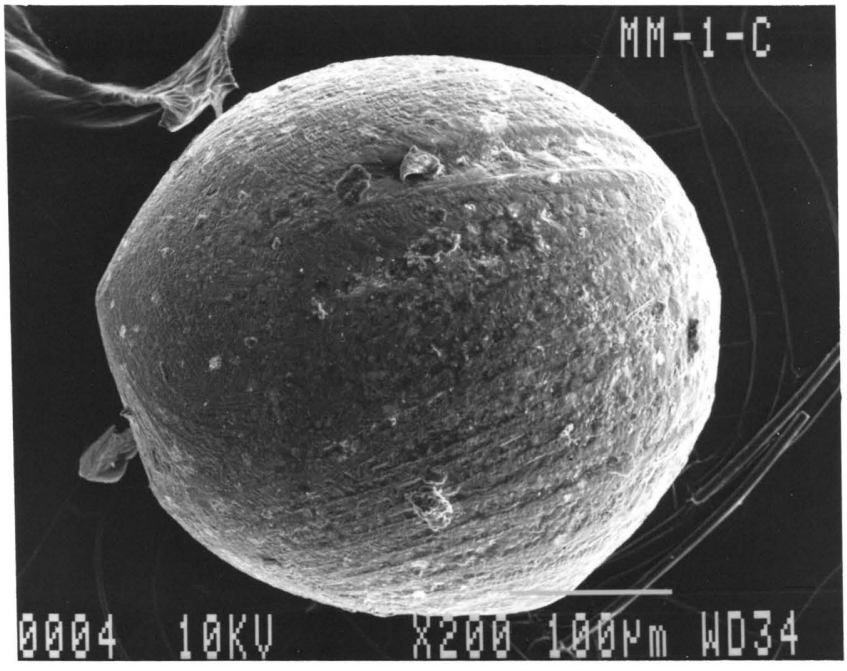


Plate 4.

Spherule MM-1-C displays "brickwork" structure. (magnification 200x, marker bar is 100 micrometers)

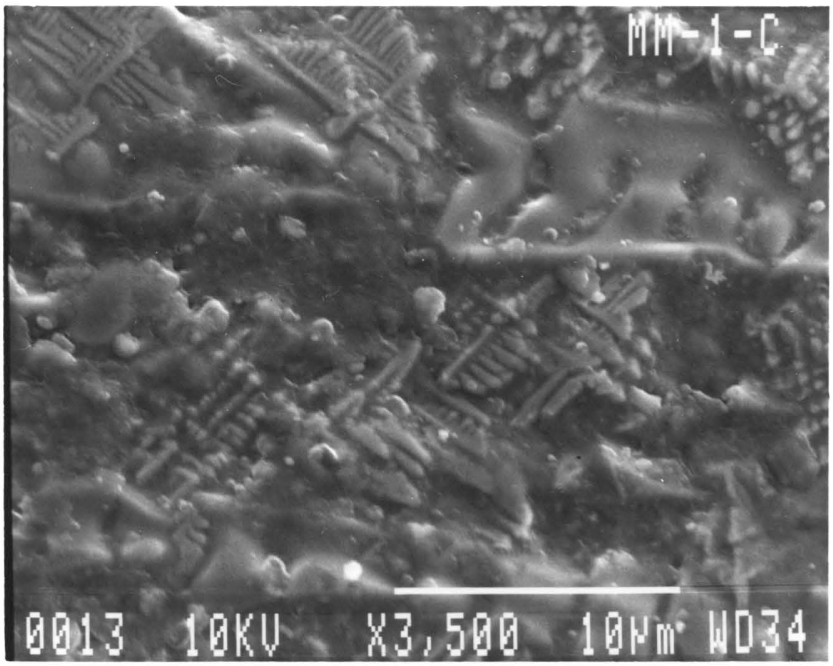


Plate 5.

Close-up of "brickwork" structure of spherule MM-1-C showing dendritic crystals of olivine and magnetite in a glassy matrix. (magnification 3500x, marker bar is 10 micrometers)

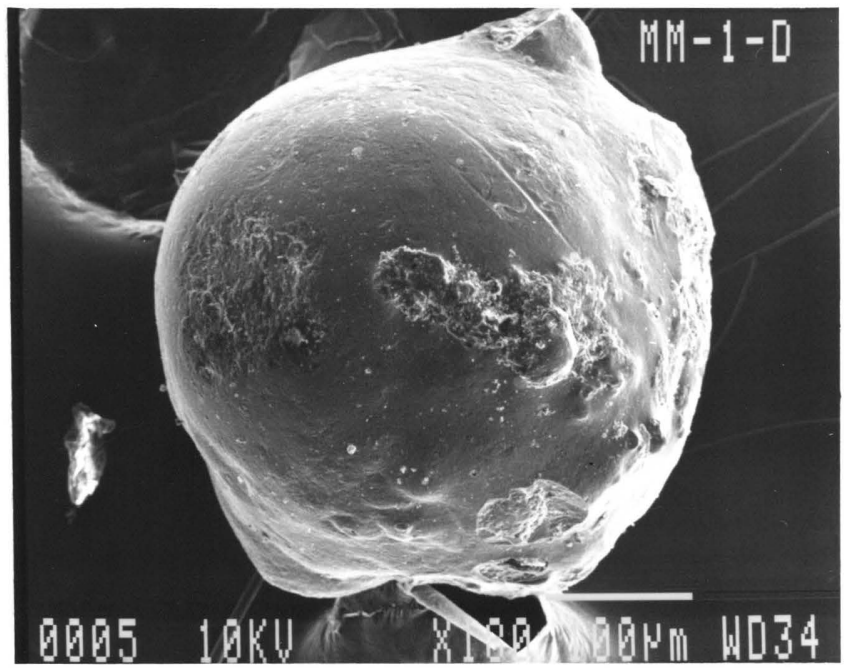


Plate 6.

Spherule MM-1-D has an irregular outline and a "lumpy" surface. The lumps may be olivine crystals or failed "cupules". (magnification 180x, marker bar is 100 micrometers)

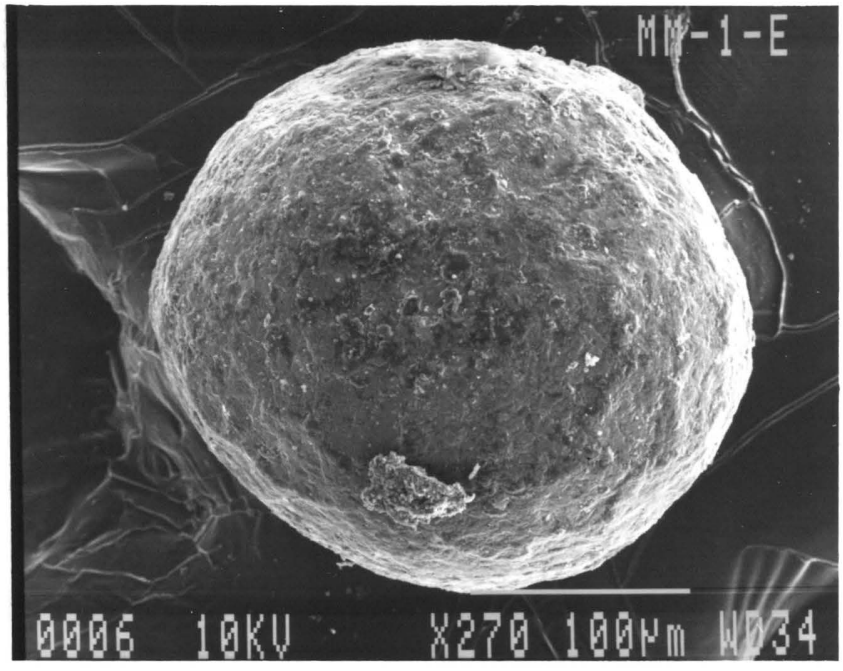


Plate 7.

Spherule MM-1-E has an irregular surface which may be a consequence of erosion or loss of "fusion crust". (magnification 270x, marker bar is 100 micrometers)

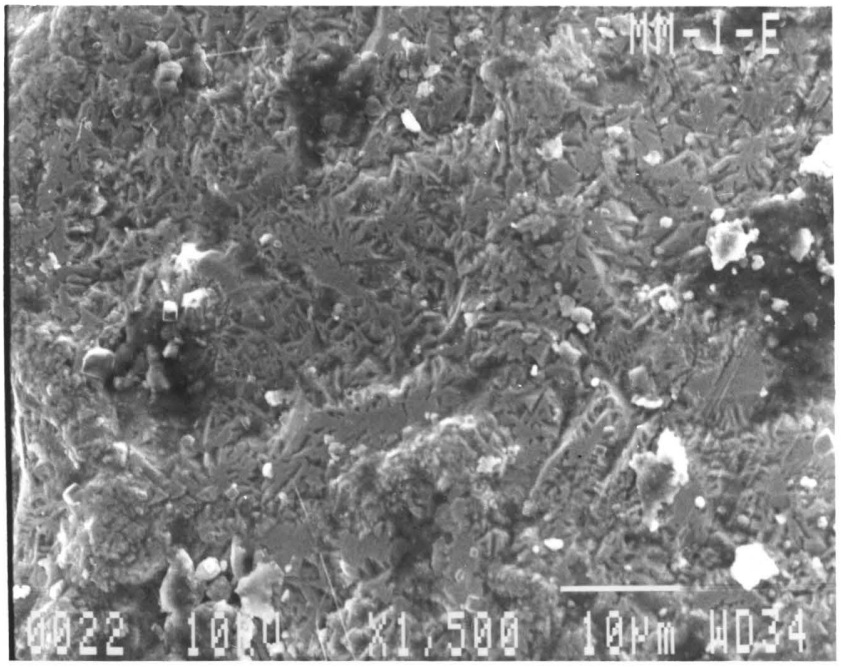


Plate 8.

Close-up of spherule MM-1-E showing crystals of olivine and magnetite in a glassy matrix. (magnification 1500x, marker bar is 10 micrometers)

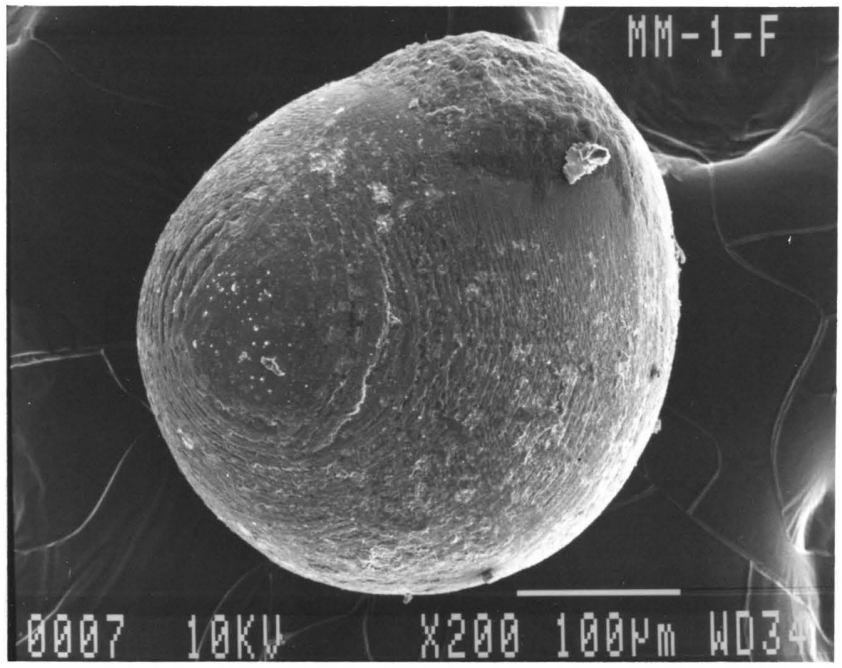


Plate 9.

Spherule MM-1-F displays "brickwork" structure. (magnification 200x, marker bar is 100 micrometers)

Methods

Instrument

Chemical analyses of the spherules pictured in Plates 1 to 9 were obtained using the JEOL JSM-820 scanning electron microscope equipped with a Link Analytical eXL energy dispersive X-ray analyzer (EDX). The operating conditions included a working distance of 39 mm, an accelerating voltage of 20,000 electron volts and a vacuum in the sample chamber of 10^{-5} Torr. In addition, the chemical analyses were carried out with a lithium-drifted silicon scintillation crystal, a probe current of 1.05 nA, and a collection time of 100 seconds. Stage tilt was not used during analyses and the beryllium window, which protects the detector, remained open to allow unhindered access of the x-rays to the detector.

The analyzer software utilized the ZAF-PB corrections to calculate the concentrations of the elements chosen. The 'Z' correction takes into account that the intensity of X-rays decreases with increasing atomic number of the elements being analyzed. The 'A' correction compensates for absorption effects--when X-rays generated deep within an atom are absorbed by the electrons of the outer shells or when X-rays emitted by light elements are absorbed by heavier elements. The 'F' correction adjusts for fluorescence--when energetic X-rays emitted from heavy elements cause X-rays to be generated from lighter elements. PB is the part of the software that utilizes the ratio of peak area to background and compares it to known ratios of selected elements that are stored in the memory of the computer (Mitchell, personal communication).

Standards

Four mineral standards, obtained from Dr. M. Barton, Director of the Microprobe Laboratory of the Department of Geological Sciences, were selected because they contained the elements of interest at a range of concentrations: chromium augite, pyrope, omphacite, and hornblende. The elemental concentrations of these standards had been determined using wet chemical methods and are listed in Table 1 based on information in Geostandards Newsletter, Vol. 4, No.1, April 1980. The concentrations of the oxides in Table 1 were recalculated to the elemental form because the analyses of the spherules are presented in this form.

Sample Preparation

The spherules were placed on a carbon stub using double-sided carbon tape. Both the spherules and the standards were coated with a thin layer of carbon (60-100 Ångstroms) and were then placed under vacuum to outgas them prior to analysis.

Analysis of the Standards

A quantitative chemical analysis was performed on each of the four standards using the machine settings described above. The electron beam was focused on each standard at five different places. This allowed for five separate analyses for each standard. The data from these analyses are summarized in Table 2.

Table 1
Chemical composition of standards in the Harvard Block expressed in % by wt. of oxides

	Cr-Augite	Pyrope[1]	Omphacite[2]	Hornblende[3]
SiO2	50.35	41.46	55.42	40.37
Al2O3	8.01	23.73	8.89	14.9
Fe2O3	1.04	0	1.35	3.3
FeO	3.76	10.68	3.41	7.95
MgO	17.28	18.51	11.57	12.8
CaO	17.26	5.17	13.75	10.3
Na2O	0.84	0	5	2.6
K2O	0	0	0.15	2.05
TiO2	0.51	0.47	0.37	4.72
MnO	0.12	0.28	0.1	0.09
Cr2O3	0.85	0	0	0
H2O	0	0	0.02	0.94
Sum	100.02	100.3	100.03	100.02

1. Pyrope, Kakanui, New Zealand (USNM 143968)
2. Omphacite, Roberts Victor Mine, So. Africa (USNM 110607)
3. Hornblende, Kakanui, New Zealand (USNM 143965)

Table 2. Summary of the Analyses of the Standards

1. Cr-Augite

Element	Actual, %	P/B, avg.	ZAF corr.% Normalized	Difference, %
Si	23.54	4.369	22.86	-2.89
Al	4.28	0.681	4.35	1.64
Fe	3.65	0.964	3.31	-9.32
Mg	10.42	1.471	12.29	17.95
Ca	12.34	3.472	16.06	30.15
Na	0.62	0.076	0.35	-43.55
Ti	0.31	0.073	0.27	-12.9
Mn	0.09	0.032	0.11	22.22
Cr	0.44	0.168	0.57	29.55
K	0	0	0	0
O	44.31	1.264	39.83	-10.11
Total	100		100	

2. Omphacite

Element	Actual, %	P/B, avg.	ZAF corr.% Normalized	Difference, %
Si	25.91	4.906	25.54	-1.43
Al	4.75	0.753	4.79	0.84
Fe	3.59	0.935	3.2	-10.86
Mg	6.98	0.978	8.18	17.19
Ca	9.83	2.773	12.73	29.5
Na	3.71	0.42	1.92	-48.25
Ti	0.22	0.063	0.23	4.55
Mn	0.08	0.017	0.06	-25
Cr	0	0	0	0
K	0.12	0.031	0.11	-8.33
O	44.81	1.378	43.25	-3.48
Total	100		100.01	

3. Pyrope

Element	Actual, %	P/B, avg.	ZAF corr.% Normalized	Difference, %
Si	19.33	3.844	18.58	-3.88
Al	12.65	2.131	12.69	0.32
Fe	8.28	2.216	7.1	-14.25
Mg	11.13	1.609	12.47	12.04
Ca	3.68	1.048	4.49	22.01
Na	0	0	0	0
Ti	0.28	0.071	0.24	-14.29
Mn	0.22	0.066	0.22	0
Cr	0	0	0	0
K	0	0	0	0
O	44.43	1.493	44.21	-0.5
Total	100		100	

4. Hornblende

Element	Actual, %	P/B, avg.	ZAF corr.% Normalized	Difference, %
Si	18.88	3.444	17.6	-6.78
Al	7.96	1.243	7.8	-2.01
Fe	8.49	2.124	7.15	-15.78
Mg	7.72	1.048	8.53	10.49
Ca	7.36	1.981	8.93	21.33
Na	1.93	0.214	0.96	-50.26
Ti	2.83	0.784	2.78	-1.77
Mn	0.07	0.013	0.05	-28.57
Cr	0	0	0	0
K	1.7	0.448	1.52	-10.59
O	43.06	1.441	44.66	3.72
Total	100		99.98	

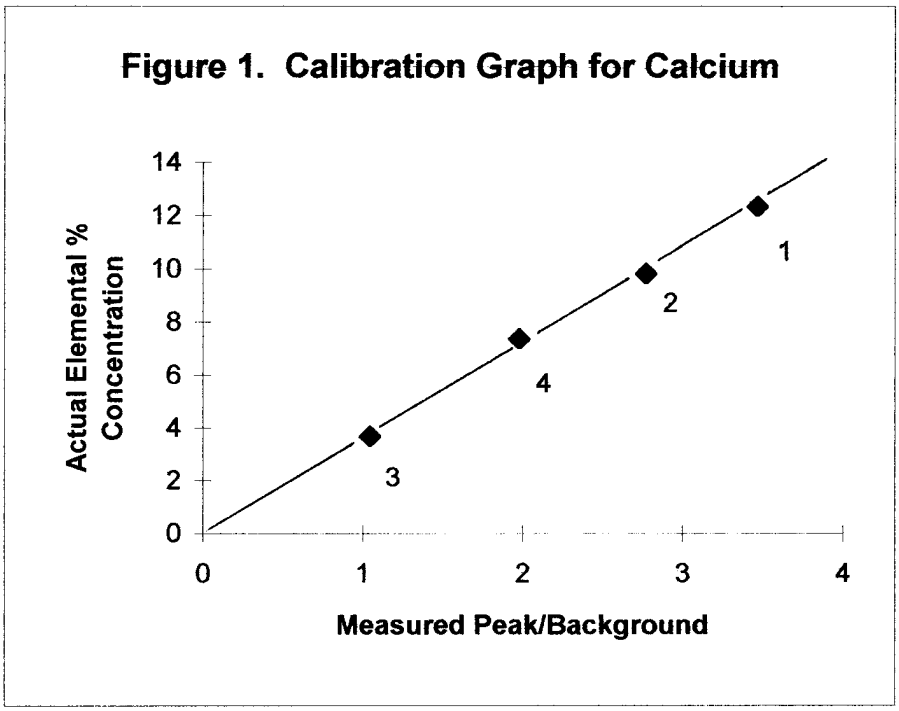
Calibration

The ZAF corrected concentrations of the standards in Table 2 were normalized to 100%, and the differences between the known concentrations and the measured concentrations were calculated by subtracting the known concentrations from the measured concentrations and by expressing the difference as a percent of the known concentrations. The results demonstrate that the measured concentrations of aluminum, silicon, and oxygen agree within five percent with the known concentrations; however, the measured elemental concentrations of the other elements differ by up to 50% from the known concentrations. For this reason, calibration curves were plotted based on the measured concentrations of calcium, iron, and magnesium in the standards versus the measured peak to background ratios (Figures 1,2,3). These graphs illustrate the fact that the data points fit straight lines that include the origin in each case. Therefore, the slopes of the calibration lines of all elements can be used to convert the measured peak to background ratios of unknowns into the corresponding concentrations of the elements without relying on the ZAF-PB corrections (calculations of the slopes and errors are given in Table 3.). Errors calculated from the slopes can be incorporated with the errors from the peak to background ratios by using the least-squares method.

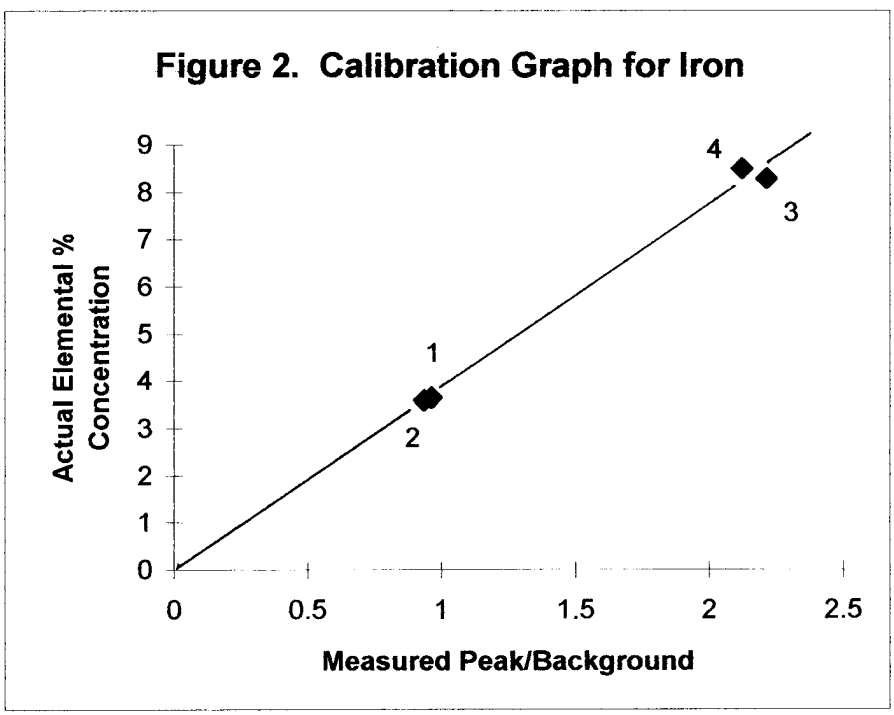
Analysis of the Spherules

A quantitative chemical analysis was carried out on each of the six spherules using the same machine settings described above. Three separate analyses

were obtained from each spherule. The elements analyzed were Si, Al, Ti, Fe, Mn, Ca, Mg, Na, K, Cr, and O. The ZAF-PB concentrations were used for Si, Al, and O; whereas the calibration factors in Table 3 were used to calculate the concentrations ot the other elements from the average peak to background ratios. The final elemental concentrations are given in Tables 4 to 6. Raw data from the analyses are presented in Tables 7-12 in the Appendix.



1. Cr-Augite; 2. Omphacite; 3. Pyrope; 4. Hornblende



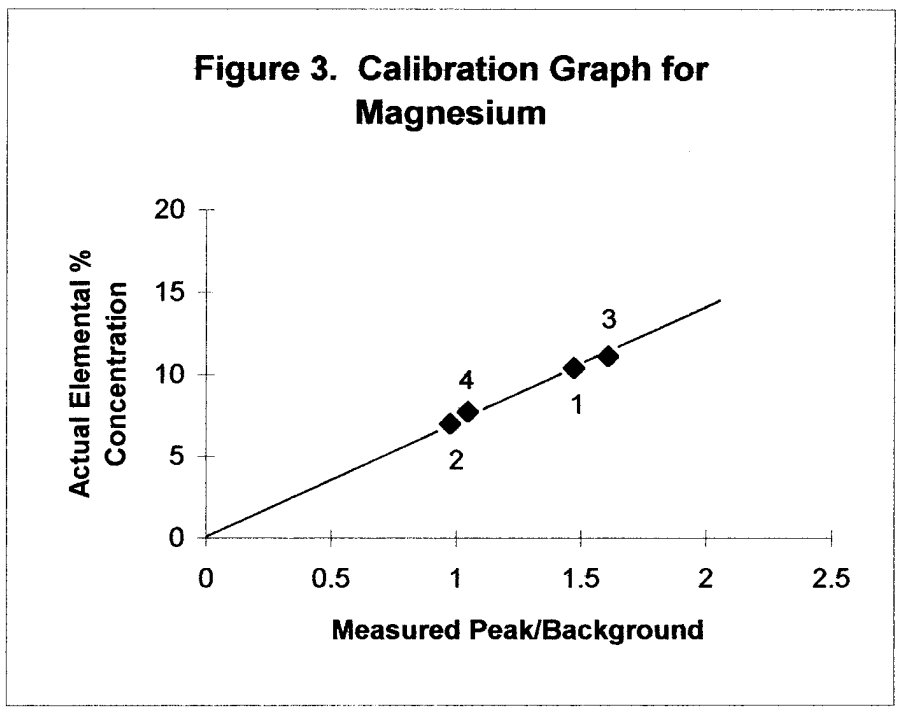


Figure 3. Calibration Graph for Magnesium
Actual Elemental % Concentration
20
15
10
5
0
0
0.5
1
1.5
2
2.5
Measured Peak/Background
1
2
3
4

Table 3. Calculations of Calibration Factors

	Magnesium			Sodium			Calcium			Potassium		
Standard	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**
1.	1.471	10.42	7.0836	0.076	0.62	8.16	3.472	12.34	3.55			
2.	0.978	6.98	7.137				2.773	9.83	3.54	0.039	0.12	3.08
3.	1.609	11.13	6.9173	0.42	3.71	8.83	1.048	3.68	3.51			
4.	1.048	7.72	7.3664	0.214	1.93	9.02	1.981	7.36	3.72	0.448	1.7	3.79
Avg.Slope=			7.13			8.67			3.58			3.44
Error(σ_{n-1})=			0.19			0.45			0.09			0.5

	Iron			Titanium			Manganese			Chromium		
Standard	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**	**X**	**Y**	**Slope**
1.	0.964	3.65	3.79	0.073	0.31	4.25				0.168	0.44	2.62
2.	0.935	3.59	3.84	0.063	0.22	3.49						
3.	2.216	8.28	3.74	0.071	0.28	3.94	0.066	0.22	3.33			
4.	2.124	8.49	4	0.784	2.83	3.61						
Avg.Slope=			3.84			3.82			3.33			2.62
Error(σ_{n-1})=			0.11			0.34			0			0

X = Measured Peak/Background; Y = Actual Elemental % Concentration
1. Cr-Augite; 2. Omphacite; 3. Pyrope; 4. Hornblende
The Avg. Slope was used as the Calibration Factor

Results

The results of the analyses are presented in Tables 4-6. Spherules MM-1-A, C, and D have higher concentrations of silicon than iron, while spherules MM-1-B, E, and F contain more iron than silicon. In spherule MM-1-C, magnesium has the highest concentration and in MM-1-D aluminum is dominant. Oxygen concentrations in the spherules range from 45.54 to 54.00 percent.

To determine the origin of the spherules, each spherule was compared to terrestrial and extraterrestrial material. The elemental concentrations of the spherules were divided by the elemental concentrations of the bulk continental crust and CI chondrites (Taylor and McClennan, 1985). If the ratio is less than 1.0, then the spherule is depleted in that element with respect to the bulk continental crust or CI chondrites. If the ratio is greater than 1.0, then the spherule is enriched. Therefore, this ratio is referred to as the enrichment factor.

In Figures 4-9, the log of the enrichment factors is plotted versus the atomic number of each element included in this study. The figures also list the enrichment factors for each element, and the averages with standard deviations. If the average and standard deviation of the enrichment factors relative to CI chondrites are lower than the average enrichment factors relative to the continental crust then the spherule is more similar in composition to CI chondrites. The graphs clearly show that spherules MM-1-B, C, E, and F are chemically similar to CI chondrites which is evidence for an extraterrestrial origin. The graphs for spherules MM-1-A and D are not as clear, but they too are more similar to CI chondrites than to the bulk continental crust.

Table 4. Results of the Analyses of the Spherules

		MM-1-A		
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	23.94	0.71	24.64	0.73
Al	3.16	0.13	3.25	0.13
Ti	0.11	0.05	0.11	0.05
Fe	7.58	0.31	7.8	0.32
Mn	0.23	0.06	0.24	0.06
Ca	1.19	0.06	1.22	0.06
Mg	15.82	0.59	16.28	0.61
Na	0.34	0.06	0.35	0.06
K	0.3	0.06	0.31	0.06
Cr	0.02	0.01	0.02	0.01
O	44.48	1.18	45.78	1.21
Total	97.17		100	

		MM-1-B		
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	11.57	0.28	13.25	0.32
Al	4.04	0.12	4.63	0.14
Ti	0.11	0.04	0.13	0.05
Fe	24	0.94	27.48	1.08
Mn	0.17	0.05	0.19	0.06
Ca	2.24	0.08	2.56	0.09
Mg	1.97	0.08	2.26	0.09
Na	0.29	0.05	0.33	0.06
K	0.08	0.03	0.09	0.03
Cr	0.09	0.08	0.1	0.09
O	42.78	0.95	48.98	1.09
Total	87.34		100	

Table 5. Results of the Analyses of the Spherules

	MM-1-C			
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	17.74	0.28	19.83	0.31
Al	0.57	0.12	0.64	0.13
Ti	0.07	0.04	0.08	0.05
Fe	10.7	0.94	11.96	1.05
Mn	0.07	0.05	0.08	0.06
Ca	0.57	0.08	0.64	0.09
Mg	18.12	0.08	20.26	0.09
Na	0.19	0.05	0.21	0.06
K	0.04	0.03	0.04	0.03
Cr	0.18	0.08	0.2	0.09
O	41.19	0.95	46.05	1.06
Total	89.44		99.99	

	MM-1-D			
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	19.25	0.65	20.36	0.69
Al	24.18	0.75	25.58	0.79
Ti	0.36	0.06	0.38	0.06
Fe	5	0.21	5.29	0.22
Mn	0.06	0.05	0.06	0.05
Ca	1.26	0.06	1.33	0.06
Mg	0.24	0.04	0.25	0.04
Na	0.23	0.04	0.24	0.04
K	0.9	0.14	0.95	0.15
Cr	0	0	0	0
O	43.05	0.98	45.54	1.04
Total	94.53		99.98	

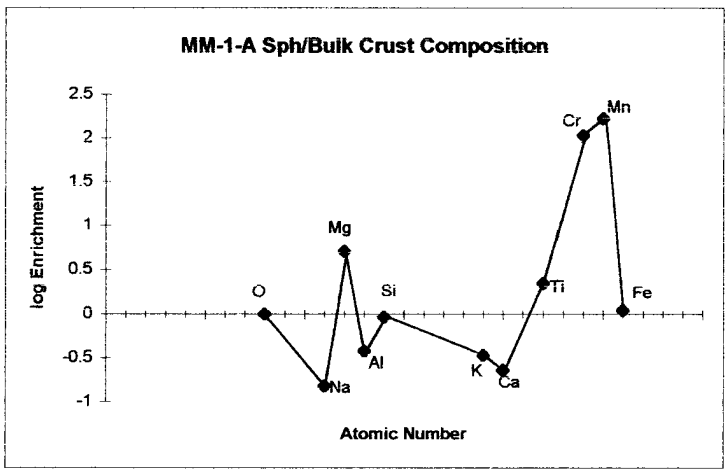
Table 6. Results of the Analyses of the Spherules

		MM-1-E		
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	1.6	0.06	2.06	0.08
Al	1.83	0.07	2.36	0.09
Ti	0.1	0.04	0.13	0.05
Fe	31.14	1.21	40.16	1.56
Mn	0.03	0.02	0.04	0.03
Ca	0.38	0.03	0.49	0.04
Mg	0.24	0.04	0.31	0.05
Na	0.26	0.05	0.34	0.07
K	0.08	0.03	0.1	0.04
Cr	0.01	0.01	0.01	0.01
O	41.87	0.97	54	1.25
Total	77.54		100	

		MM-1-F		
element	% Concentration	Error, %	% Concentration, Normalized	Error, % Normalized
Si	16.01	0.36	18.5	0.42
Al	1.44	0.08	1.66	0.09
Ti	0.03	0.03	0.03	0.03
Fe	19.17	0.76	22.15	0.88
Mn	0.1	0.05	0.12	0.06
Ca	0.44	0.04	0.51	0.05
Mg	8.42	0.28	9.73	0.32
Na	0.17	0.04	0.2	0.05
K	0.02	0.02	0.02	0.02
Cr	0.03	0.03	0.03	0.03
O	40.7	0.88	47.04	1.02
Total	86.53		99.99	

MM-1-A

element	Sph/B.C.	Sph/Cl
Si	0.92	1.54
Al	0.38	2.5
Ti	2.2	1.57
Fe	1.1	0.28
Mn	171.43	81.63
Ca	0.23	0.9
Mg	5.09	0.14
Na	0.15	0.48
K	0.34	3.88
Cr	108.11	5.03
O	1	1.19
Average	26.45	9.01
σ_{n-1}	57.81	24.13



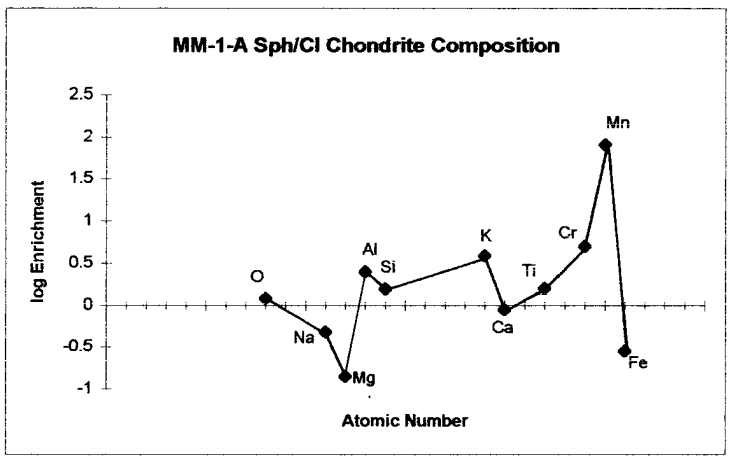
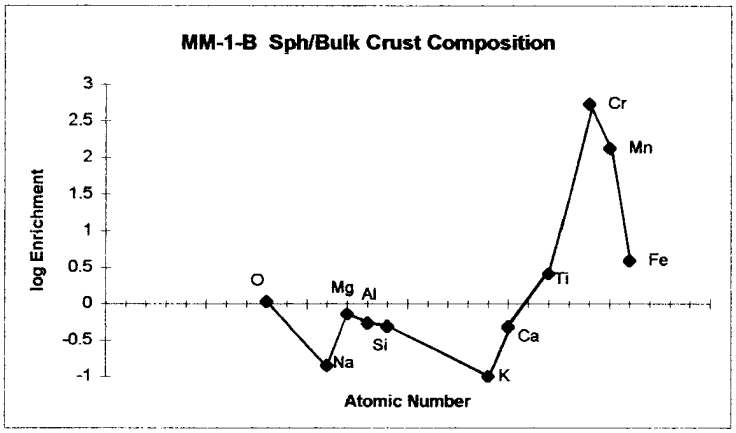


Figure 4. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and CI Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/CI is the enrichment factor of the spherule to CI Chondrites

MM-1-B		
element	Sph/B.C.	Sph/Cl
Si	0.49	0.83
Al	0.54	3.56
Ti	2.6	1.86
Fe	3.89	0.99
Mn	135.71	64.63
Ca	0.48	1.9
Mg	0.71	0.16
Na	0.14	0.45
K	0.1	1.13
Cr	540.54	25.16
O	1.07	1.28
Average	62.39	9.27
σ_{n-1}	163.65	19.72



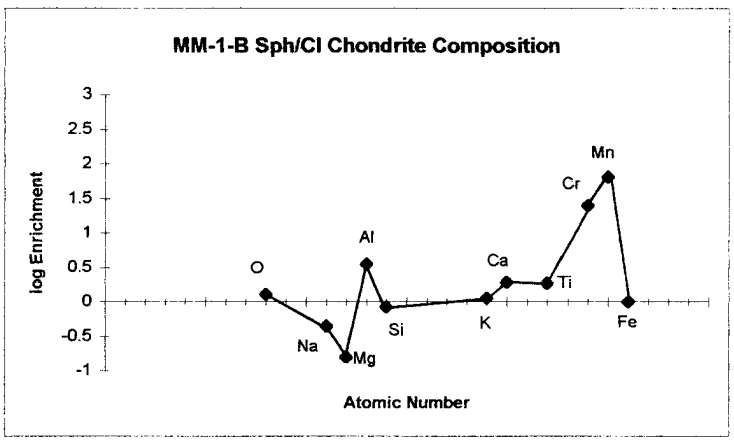
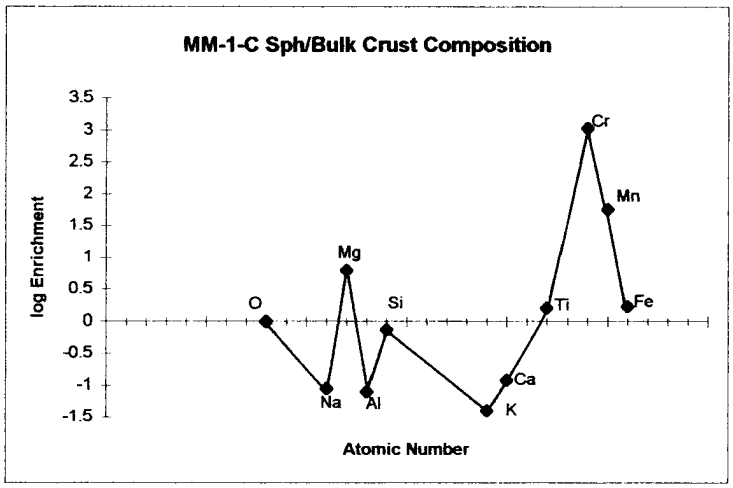


Figure 5. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and Cl Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/Cl is the enrichment factor of the spherule to Cl Chondrites

MM-1-C		
element	Sph/B.C.	Sph/Cl
Si	0.74	1.24
Al	0.08	0.49
Ti	1.6	1.14
Fe	1.69	0.43
Mn	57.14	27.21
Ca	0.12	0.47
Mg	6.33	1.42
Na	0.09	0.29
K	0.04	0.5
Cr	1081.08	50.31
O	1	1.2
Average	104.54	7.7
σ_{n-1}	324.32	16.21



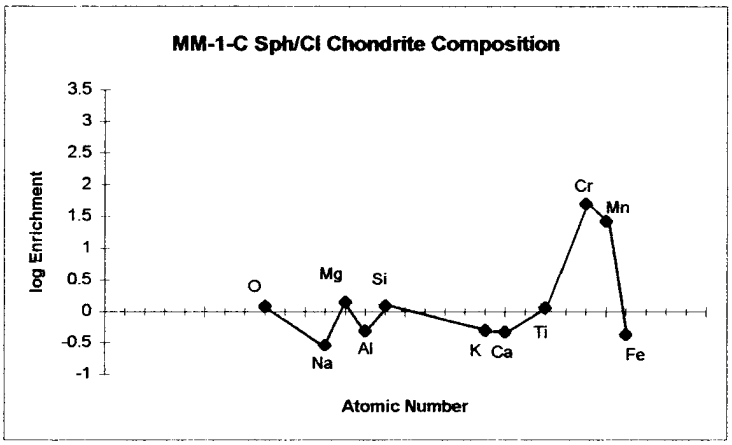
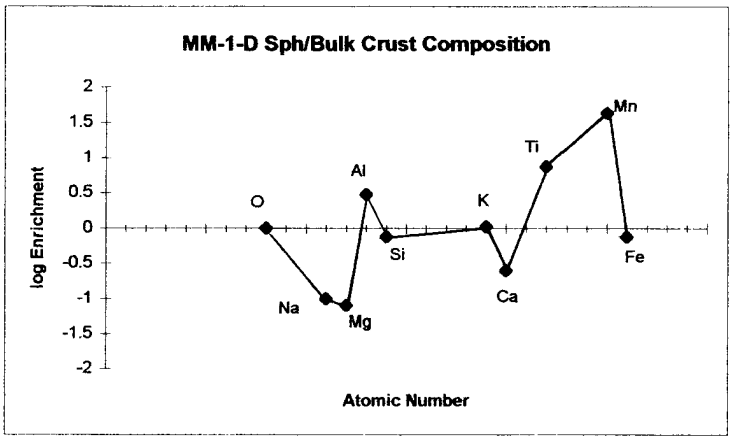


Figure 6. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and CI Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/CI is the enrichment factor of the spherule to CI Chondrites

	MM-1-D	
element	Sph/B.C.	Sph/Cl
Si	0.76	1.27
Al	3.01	19.68
Ti	7.6	5.43
Fe	0.75	0.19
Mn	42.86	20.41
Ca	0.25	0.99
Mg	0.08	0.02
Na	0.1	0.33
K	1.04	11.88
Cr	0	0
O	0.99	1.19
Average	5.22	5.58
σ_{n-1}	12.68	7.98



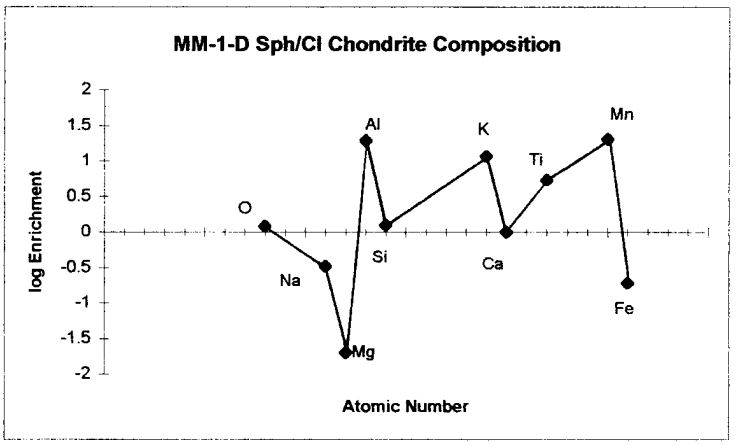
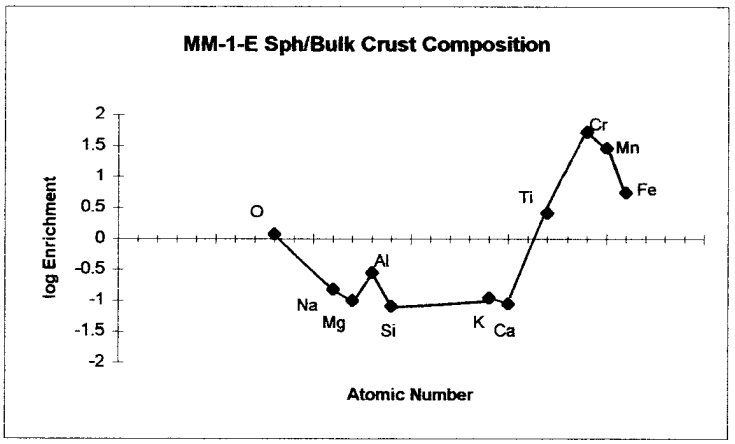


Figure 7. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and Cl Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/Cl is the enrichment factor of the spherule to Cl Chondrites

MM-1-E		
element	Sph/B.C.	Sph/Cl
Si	0.08	0.13
Al	0.28	1.82
Ti	2.6	1.86
Fe	5.67	1.44
Mn	28.57	13.61
Ca	0.09	0.36
Mg	0.1	0.02
Na	0.15	0.47
K	0.11	1.25
Cr	54.05	2.52
O	1.18	1.41
Average	8.44	2.26
σ_{n-1}	17.3	3.85



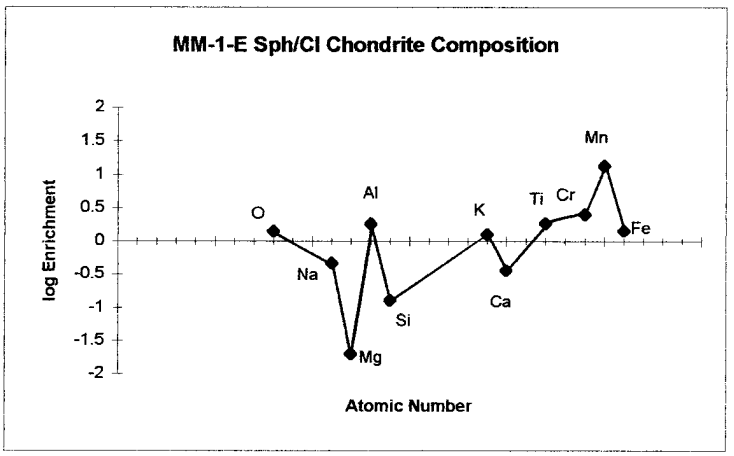
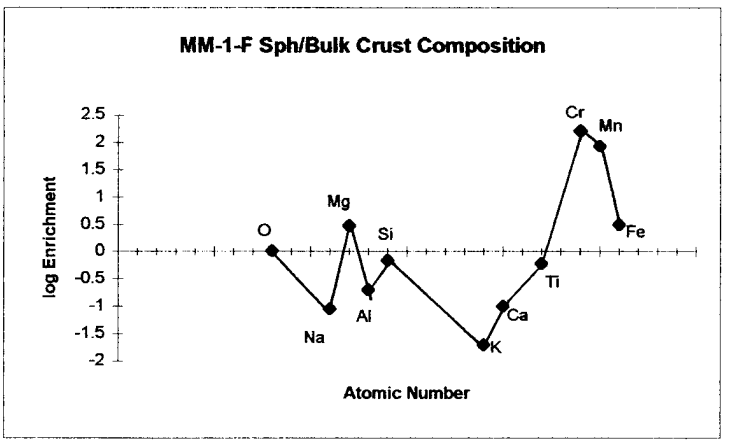


Figure 8. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and Cl Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/Cl is the enrichment factor of the spherule to Cl Chondrites

MM-1-F		
element	Sph/B.C.	Sph/Cl
Si	0.69	1.16
Al	0.2	1.28
Ti	0.6	0.43
Fe	3.13	0.8
Mn	85.71	40.82
Ca	0.1	0.38
Mg	3.04	0.68
Na	0.09	0.27
K	0.02	0.25
Cr	162.16	7.55
O	1.02	1.22
Average	23.34	4.99
σ_{n-1}	52.6	12.07



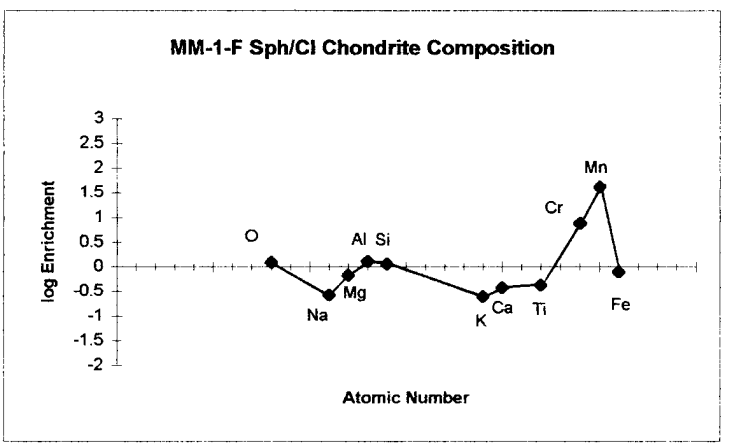


Figure 9. Algebraic and Graphical Comparisons of Spherule to Bulk Continental Crust and CI Chondrites

Sph/B.C. is the enrichment factor of the spherule to the Bulk Continental Crust
Sph/Cl is the enrichment factor of the spherule to CI Chondrites

Origin of the Spherules

These spherules most likely formed during the ablation of meteorites traveling at high speed through the earth's atmosphere. They are well preserved in Antarctica due to the absence of liquid water. The spherules were found in a moraine which suggests that they were once embedded in the ice sheet and then traveled with the ice until they were released from the ice in the zone of ablation of the glacier (Faure, 1995, personal communication). The spherules are then carried by the wind and are trapped by the moraine in which they were found.

Summary of Conclusions

The six spherules described in this report were determined to have an extraterrestrial origin based on the evidence that they are more closely related to carbonaceous chondrites than to bulk continental crust. It was also determined that by using calibration factors, an SEM equipped with EDX can provide a reliable microanalysis of these spherules.

Acknowledgments

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Appendix

Table 7. Raw Data from Analysis of Spherule MM-1-A

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	3.726	0.109	24.16	0.71	3.887	0.123	25.31	0.8	3.444	0.096	22.34	0.62
Al	0.259	0.014	2.05	0.11	0.251	0.014	1.99	0.11	0.687	0.023	5.45	0.18
Ti	0.028	0.013	0.13	0.06	0.021	0.013	0.09	0.06	0.041	0.013	0.19	0.06
Fe	2.113	0.059	9.06	0.25	2.146	0.061	9.24	0.26	1.663	0.047	7.13	0.2
Mn	0.062	0.018	0.27	0.08	0.091	0.019	0.41	0.08	0.053	0.017	0.24	0.08
Ca	0.354	0.015	2.03	0.08	0.359	0.015	2.07	0.08	0.283	0.014	1.62	0.08
Mg	2.434	0.066	25.32	0.68	2.602	0.076	27.2	0.8	1.624	0.037	16.88	0.38
Na	0.02	0.006	0.12	0.04	0.03	0.007	0.17	0.04	0.067	0.005	0.38	0.03
K	0.013	0.01	0.05	0.04	0.009	0.01	0.04	0.04	0.239	0.012	1.04	0.05
Cr	0.004	0.015	0.02	0.06	-0.003	0.015	-0.01	0.06	0.024	0.015	0.1	0.06
O	1.165	0.032	46.36	1.29	1.154	0.034	46.1	1.36	1.031	0.022	40.98	0.88
Total			109.57				112.61				96.35	

Table 8. Raw Data from Analysis of Spherule MM-1-B

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	2.847	0.071	21.75	0.54	1.46	0.03	11.72	0.24	0.137	0.006	1.23	0.06
Al	0.685	0.019	6.41	0.17	0.387	0.012	3.81	0.12	0.173	0.006	1.91	0.07
Ti	0.059	0.011	0.31	0.06	0.031	0.011	0.17	0.06	0	0.01	0	0.06
Fe	4.166	0.101	20.98	0.51	6.055	0.161	32.18	0.86	8.53	0.222	51.07	1.33
Mn	0.051	0.015	0.27	0.08	0.012	0.014	0.06	0.08	0.012	0.013	0.07	0.08
Ca	1.314	0.027	8.82	0.18	0.514	0.015	3.62	0.1	0.045	0.009	0.35	0.07
Mg	0.188	0.007	2.28	0.08	0.62	0.014	7.9	0.17	0.023	0.005	0.33	0.07
Na	0.035	0.005	0.23	0.03	0.029	0.005	0.2	0.04	0.037	0.005	0.29	0.04
K	0.038	0.009	0.19	0.04	0.016	0.008	0.09	0.04	0.011	0.007	0.07	0.04
Cr	0.011	0.013	0.05	0.06	0.037	0.013	0.18	0.06	0.032	0.012	0.16	0.06
O	0.918	0.021	42.69	0.96	0.858	0.02	42.54	0.97	0.764	0.016	43.12	0.93
Total			103.98				102.47				98.6	

Table 9. Raw Data from Analysis of Spherule MM-1-C

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	2.843	0.069	18.77	0.46	2.641	0.064	18	0.43	2.491	0.058	16.45	0.38
Al	0.045	0.008	0.36	0.07	0.129	0.009	1.07	0.08	0.035	0.007	0.28	0.06
Ti	0.019	0.012	0.09	0.05	0.004	0.013	0.02	0.06	-0.006	0.012	-0.03	0.06
Fe	2.83	0.072	12.39	0.32	2.942	0.077	13.29	0.35	2.59	0.068	11.34	0.3
Mn	0.004	0.015	0.02	0.07	0.027	0.016	0.12	0.07	0.029	0.016	0.13	0.07
Ca	0.106	0.011	0.62	0.06	0.304	0.014	1.83	0.08	0.068	0.011	0.4	0.06
Mg	2.944	0.079	31.26	0.83	2.134	0.051	23.32	0.56	2.552	0.061	27.1	0.65
Na	0.021	0.006	0.12	0.04	0.025	0.006	0.15	0.04	0.019	0.006	0.11	0.03
K	-0.008	0.009	-0.03	0.04	0.01	0.009	0.05	0.04	0.015	0.009	0.07	0.04
Cr	0.014	0.014	0.06	0.06	0.005	0.014	0.02	0.06	0.017	0.015	0.07	0.06
O	1.078	0.024	43.91	1	0.947	0.022	39.73	0.93	0.979	0.02	39.92	0.83
Total			107.57				97.6				95.84	

Table 10. Raw Data from Analysis of Spherule MM-1-D

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	3.09	0.103	20.06	0.67	3.038	0.099	19.89	0.65	2.958	0.104	17.81	0.63
Al	2.966	0.092	23.93	0.74	2.859	0.087	23.26	0.7	3.372	0.109	25.36	0.82
Ti	0.118	0.013	0.53	0.06	0.14	0.014	0.64	0.06	0.028	0.013	0.12	0.06
Fe	1.679	0.046	7.22	0.2	1.788	0.049	7.76	0.21	0.435	0.026	1.74	0.11
Mn	0.02	0.015	0.09	0.07	0.018	0.015	0.08	0.07	-0.014	0.016	-0.06	0.07
Ca	0.436	0.015	2.51	0.09	0.471	0.016	2.73	0.09	0.152	0.013	0.81	0.07
Mg	0.037	0.006	0.38	0.06	0.04	0.006	0.42	0.06	0.025	0.006	0.24	0.06
Na	0.031	0.004	0.18	0.02	0.032	0.004	0.19	0.02	0.019	0.004	0.1	0.02
K	0.267	0.012	1.17	0.05	0.222	0.012	0.98	0.05	0.295	0.014	1.2	0.06
Cr	0.002	0.014	0.01	0.06	-0.007	0.013	-0.03	0.06	-0.013	0.016	-0.05	0.06
O	1.115	0.024	44.34	0.97	1.088	0.025	43.64	1.01	1.113	0.026	41.16	0.95
Total			100.42				99.56				88.43	

Table 11. Raw Data from Analysis of Spherule MM-1-E

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	0.342	0.008	2.96	0.07	0.088	0.006	0.8	0.06	0.115	0.006	1.05	0.06
Al	0.219	0.007	2.32	0.07	0.111	0.006	1.24	0.07	0.102	0.006	1.93	0.08
Ti	0.043	0.01	0.25	0.06	0.017	0.011	0.1	0.06	0.019	0.011	0.13	0.06
Fe	7.055	0.171	40.65	0.98	8.253	0.22	50.07	1.34	9.023	0.23	54.74	1.58
Mn	-0.007	0.013	-0.04	0.07	0.023	0.014	0.14	0.09	-0.009	0.013	-0.06	0.08
Ca	0.234	0.01	1.77	0.08	0.05	0.009	0.4	0.07	0.035	0.009	0.27	0.07
Mg	0.056	0.005	0.77	0.07	0.02	0.005	0.29	0.08	0.024	0.006	0.34	0.08
Na	0.06	0.004	0.45	0.03	0.01	0.005	0.08	0.04	0.02	0.005	0.16	0.04
K	0.05	0.008	0.29	0.05	0.009	0.008	0.05	0.05	0.014	0.008	0.08	0.05
Cr	0.005	0.012	0.03	0.06	-0.004	0.012	-0.02	0.06	0.013	0.012	0.07	0.06
O	0.741	0.015	39.95	0.83	0.713	0.017	40.72	0.95	0.79	0.02	44.94	1.13
Total			89.4				93.87				103.65	

Table 12. Raw Data from Analysis of Spherule MM-1-F MM-1-F

	Analysis 1				Analysis 2				Analysis 3			
element	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error	P/B	P/B error	ZAF,%	ZAF % error
Si	1.962	0.043	14.63	0.32	2.246	0.052	16.87	0.39	2.179	0.047	16.52	0.36
Al	0.146	0.009	1.33	0.08	0.177	0.009	1.63	0.08	0.146	0.008	1.35	0.08
Ti	0.013	0.012	0.06	0.06	0.002	0.011	0.01	0.06	0.013	0.011	0.07	0.06
Fe	4.74	0.126	23.44	0.62	5.073	0.133	25.27	0.66	5.16	0.134	25.93	0.67
Mn	0.027	0.015	0.14	0.08	0.042	0.016	0.22	0.08	0.023	0.015	0.12	0.08
Ca	0.101	0.011	0.66	0.07	0.15	0.011	0.99	0.08	0.118	0.01	0.78	0.07
Mg	1.341	0.029	15.94	0.34	1.157	0.025	13.83	0.29	1.048	0.021	12.63	0.25
Na	0.022	0.005	0.14	0.04	0.018	0.006	0.12	0.04	0.02	0.005	0.14	0.04
K	0.01	0.009	0.05	0.05	0.011	0.009	0.06	0.05	0.001	0.008	0	0.04
Cr	-0.002	0.014	-0.01	0.06	0.014	0.014	0.06	0.06	0.018	0.013	0.08	0.06
O	0.847	0.018	39.05	0.83	0.903	0.02	41.88	0.94	0.879	0.019	41.18	0.88
Total			95.43				100.94				98.8	