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Bulk Chemical Composition of the Ningqiang Carbonaceous Chondrite: An Issue of Classification

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Abstract: The Ningqiang meteorite is a fall carbonaceous chondrite, containing various Ca-, Al-rich inclusions that usually escaped from secondary events such as high-temperature heating and low-temperature alteration. However, it has not yet been classified into any known chemical group. In order to address this issue, 41 elements of the bulk Ningqiang meteorite were analyzed using inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atom emission spectrometry (ICP-AES) in this study. The Allende (CV3) carbonaceous chondrite and the Jilin (H5) ordinary chondrite were also measured as references, and our analyses are consistent with the previous results. Rare earth and other refractory lithophile elements are depleted in Ningqiang relative to both Allende and mean CK chondrites. In addition, the REE pattern of Ningqiang is nearly flat, while that of Allende shows slight enrichment of LREE relative to HREE. Siderophile elements of Ningqiang are close to those of mean CK chondrites, but lower than those of Allende. Our new analyses indicate that Ningqiang cannot be classified into any known group of carbonaceous chondrites, consistent with previous reports.

Key words: meteorite, carbonaceous chondrite, classification, bulk composition, ICP-MS

1 Introduction

Carbonaceous chondrites are the most primitive rocks of the solar system, composed of various components, such as Ca-, Al-rich inclusions (hereafter CAIs), silicate spherules (referred to as chondrules), opaque mineral assemblages, and fine-grained matrix, which formed at different processes during the evolution of the solar nebula. In addition, some important but trace components also survived in carbonaceous chondrites, including various presolar grains (Zinner, 1998) and organic compounds (Cooper et al., 2001; Kminek et al., 2002). Most carbonaceous chondrites were found long time after fall (up to a million years for those found in the Antarctica), hence experienced various degrees of terrestrial weathering. The Ningqiang carbonaceous chondrite fell in Ningqiang County, Shanxi Province, in 1983. Four pieces of the stones with a total mass of 4.61 kg were collected immediately after fall, therefore the samples were suffered little weathering.

Previous studies of Ningqiang revealed unique properties in comparison with other carbonaceous chondrite. It contains various petrographic types of CAIs similar to those in the Allende (CV3) carbonaceous chondrite, but most CAIs in the former show much lower degree of alteration than those in the latter (Lin and Kimura, 2003). Recently, the short-lived ^{36}Cl was first reported in a CAI in Ningqiang (Lin et al., 2005). However, the classification of Ningqiang is a long-standing controversial issue. It was firstly classified as an anomalous CV (an-CV) chondrite (Rubin et al., 1988), and later referred to as the first but anomalous CK3 (an-CK3) (Kallemeyn et al., 1991). Further study of its bulk chemistry revealed that Ningqiang did not fit into any known group of carbonaceous chondrites (Kallemeyn, 1996). It should be noticed that the discrepancy is mainly related to the bulk chemistry. On the other hand, Ningqiang shows a close relationship with the CV group in petrography (Rubin et al., 1988; Weisberg et al., 1996;

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Kimura et al., 1997) and thermal luminescence (TL) (Benoit et al., 1995).

Bulk chemical compositions are basic properties of meteorites, and serve as criteria for classification of meteorites. Most bulk data were acquired by neutron activation analysis (NAA) that required samples and standards to be irradiated in nuclear reactors. Furthermore, some elements (e.g. platinum group elements, except for Ir) have low nuclear reaction sections, hence low sensitivity. Recently, inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma atomic emission spectrometer (ICP-AES) have been significantly improved, with application to analyses of meteorites (Shinotsuka et al., 1995; Liu et al., 1998; Barrat et al., 2000; Kehm et al., 2003). The ICP-MS and ICP-AES would be powerful to measure bulk compositions of a large number of meteorites newly collected in the Grove Mountains, Antarctica (Dai et al., 2004; Lu et al., 2004). This paper reports the bulk composition of Ningqiang using ICP-MS and ICP-AES, and discusses its classification based on the new data. In addition, it should be pointed out that the Ningqiang sample used in this study is from a large amount of powder (~10 g) of the meteorite, hence more representative than previous analyses. This is important because that distribution of CAIs in Ningqiang is highly heterogeneous, which are major carriers of refractory elements in carbonaceous chondrites.

2 Samples and Experiments

2.1 Samples and sample preparation

A sample of 50 mg out of ~10 g of Ningqiang powder was used in this study. The powder (<350 mesh) was prepared in previous study (Shi et al., 1992), and it have been eluted with dichloromethane to extract organic compounds. About 1 gram of small pieces of the Jilin meteorite were cleaned several times in 18 MΩ Millipore water using ultrasonic cleaning. After dried, the pieces were ground using an agate mortar, and the powder was passed through 200 mesh. The Allende sample is powder prepared from split-8 (labeled as Allende-1), and it was used as an international reference material. We used each 50 mg of the Allende and Jilin samples.

Ten standards were selected as reference materials in this study. The standards from the National Research Center for Certified Reference Materials of China (GBW) are GSR-1 (granite), GSR-15 (plagio-amphibolite) and GSR-17 (kimberlite); those from the U.S. Geological Survey (USGS) are W-2 (diabase) and AGV-1 (andesite); those from the Canadian Certified Reference Materials Project (CCRMP) are UM-2 (ultramafic rock), UMT-1 (ultramafic ore tailings), UB-N (serpentinite) and MRG-1 (gabbro); those from the Council for Mineral Technology, South Africa, National Bureau of Standards (MINTEK) are SARM-4 (norite). 50 mg of each standard was used in this study.

The samples and standards were placed in Teflon vessels, and then they were digested using 0.3 mL of concentrated HF and 0.6 mL of 7 M HNO_3 with five drops of concentrated $HClO_4$. The sealed vessels were heated at 120°C for 3 days, and were vibrated in an ultrasonic water-bath for 30 min per 12 h. Then the vessels were sealed in stainless steel bombs, and were heated at 190°C for 24 h in an oven. After completely digested, the solutions were evaporated to dryness on a hotplate. Finally, the residues were re-dissolved with 3 mL of 4M HNO_3. All solutions of the samples and standards were diluted to 3 % HNO_3 for ICP-AES and ICP-MS measurements. The chemical procedures were carried out in an ultra-clean room.

The super-pure HNO_3 and HF in this work are obtained by double-distilling analytical-reagent (AR) grade HNO_3 and HF under sub-boiling conditions, respectively. The $HClO_4$ was 12 M redistilled product of Germany Sigma-Aldrich. The ultra-pure water (18 MΩ) was obtained from a Millipore water purification system.

2.2 ICP-AES and ICP-MS

Major elements were measured using the VISTA-PRO ICP-AES in the Key Laboratory of Isotope Geochronology and Geochemistry, Guangzhou Institute of Geochemistry. The X and Y positions of the torch were adjusted using 5 ppm Mn solution. Another multi-element solution (5 ppm for each element) was used to optimize the ICP-AES instrument. The sample and standard solutions were added with 1 ppm of Lu as an internal standard.

Minor and trace elements were determined using the Perkin-Elmer Sciex ELAN 6000 ICP-MS in the same laboratory. The instrument was optimized using a multi-elemental solution (10 ppb for each element), and it was conditioned to give sufficient signal normally more than 3×10^5 counts per second (cps) for 10 ppb Rh and less than 2% oxide yield. We used 10 ppb Rh as the internal standard. Details on interference corrections and the analyzing procedure were described by Liu et al. (1998).

2.3 Results

Forty-one elements of Ningqiang, Allende and Jilin were analyzed in this study, and the results are summarized in Table 1. Major elements of Al, Ti, Ca, Mg, Fe, Mn, Na and P were determined by ICP-AES, and the other minor and trace elements by ICP-MS. Relative standard deviations of the counting are within 10 %. Our analyses of most major and minor elements (except for K, V and Co) and REE (except for Dy and Lu) of Allende are consistent with the

Table 1 Bulk chemical compositions of Ningqiang, Allende and Jilin (in ppm except for noted)

	Ningqiang		Allende			Jilin		
	mean[a]	RSD (%)	mean[a]	RSD (%)	ref[b]	mean	RSD (%)	ref[c]
Al (%)	1.45	5	1.75	3	1.74	1.01	3	1.10
Ti	810	4	886	2	900	562	2	780
Ca (%)	1.60	5	1.85	2	1.84	1.20	2	1.35
Mg (%)	16.00	4	15.20	2	14.80	13.52	2	13.91
Fe (%)	22.40	5	23.60	2	23.60	31.25	2	28.20
Mn	1462	5	1450	2	1471	2232	2	2246
Na	3700	6	3509	2	3413	5856	3	6306
P	1028	5	1037	5	1048	972	3	1135
K	229.	3	202.	3	249	655.	5	912.
Sc	9.63	2	10.15	2	11	6.59	7	
V	71	3	72.36	2	92	49.67	3	
Cr	3944	2	3674	2	3625	3489	4	2736
Co	619	2	700	2	600	1125	4	820
Ni (%)	1.33	1	1.52	3	1.42	2.27	4	1.71
Cu	91	2	99	3	119	92	4	190
Ge	14.61	4	15.66	3	17.60	14.73	4	
Rb	1.18	6	1.12	4	1.20	2.33	2	2.26
Sr	11.08	3	12.62	3	12.00	9.03	3	
Y	2.87	3	2.86	2	3.10	1.92	2	
Zr	7.33	2	7.23	3	9.00	4.75	3	
Nb	0.49	5	0.52	3	0.53	0.36	3	
Cs	0.08	8	0.083	8	0.090	0.072	6	0.0090
Ba	5.06	1	5.19	2	4.00	3.96	4	
Hf	0.21	4	0.19	9	0.21	0.13	6	
Pb	1.09	4	1.00	4	1.39	0.16	8	
Th	0.065	4	0.065	6	0.063	0.051	7	
U	0.014	9	0.014	14	0.016	0.016	10	
La	0.45	3	0.52	3	0.52	0.29	6	0.34
Ce	1.15	3	1.33	3	1.33	0.77	5	0.89
Pr	0.18	6	0.21	4	0.21	0.11	6	0.14
Nd	0.90	4	1.01	5	0.99	0.55	6	0.65
Sm	0.28	6	0.31	8	0.34	0.18	5	0.21
Eu	0.11	8	0.11	7	0.11	0.068	6	0.078
Gd	0.39	7	0.40	8	0.42	0.24	7	0.30
Tb	0.068	3	0.071	6	0.078	0.046	6	0.054
Dy	0.44	4	0.48	4	0.43	0.30	6	0.36
Ho	0.10	5	0.10	5	0.10	0.067	4	0.081
Er	0.30	8	0.28	4	0.29	0.20	5	0.23
Tm	0.052	6	0.054	6	0.064	0.031	10	0.036
Yb	0.31	7	0.31	5	0.30	0.19	4	0.24
Lu	0.048	8	0.045	6	0.050	0.030	10	0.038

Note: [a] This work; [b] The recommended data (Jarosewich et al., 1987); [c] REE data after Liu et al. (1998), and the others after Wang et al. (1993).

recommended data within 10%, and those of the other elements (except for Pb) are less than 20%. The analyses of siderophile elements (e.g. Fe, Co, Ni) of Jilin are significantly higher than the previous analysis (Wang et al. 1993). (see Table 1). However, this discrepancy could be related to heterogeneous distribution of large grains of metallic Fe-Ni in the equilibrated ordinary chondrite.

The REE abundances of Ningqiang, Allende and Jilin are normalized to their Mg values and CI chondrites, and the results are plotted in Fig. 1. The Mg-normalization is to eliminate effects of variable abundances of metallic Fe-Ni among meteorites. Only a few data of REE of Ningqiang were reported (Rubin et al. 1988), which are slightly lower than our results (Fig. 1). This difference could be related to heterogeneous distribution of CAIs in this meteorite, and Rubin et al. (1988) used only a small amount of the sample (~0.6 g). Our analyses of REEs of the bulk Ningqiang sample (1.20$\times$ CI) are distinguishably lower than those of Allende (1.33 $\times$ CI) and the mean values of CK chondrites (1.28 $\times$ CI). It is also noticed that the REE pattern of Ningqiang is nearly flat (La/Lu = 0.97 $\times$ CI), consistent with the previous analyses (Rubin et al., 1988). In contrast, Allende shows a slight enrichment of LREE relative to HREE (La/Lu = 1.19$\times$CI, Fig. 1). Our analyses of Ningqiang and Allende show small anomalies of Tm that was not found in Jilin (Fig. 1).

CI-normalized abundances of siderophile elements (Fe, Ni, Co, Cu, Ge) of Ningqiang show a decreasing trend towards more volatile (Fig. 2a). Allende has the same pattern, but with higher abundances of siderophile elements in comparison with Ningqiang. The mean CK group was plotted between Ningqiang and Allende. The Jilin ordinary chondrite has a significantly sharper decreasing pattern in comparison with those of the carbonaceous chondrites. It contains much higher Fe, Ni and Co than Ningqiang, Allende and the mean CK-group, but the abundances of Cu and Ge are nearly the same between them, although higher abundance of Cu was reported in Jilin (Table 1).

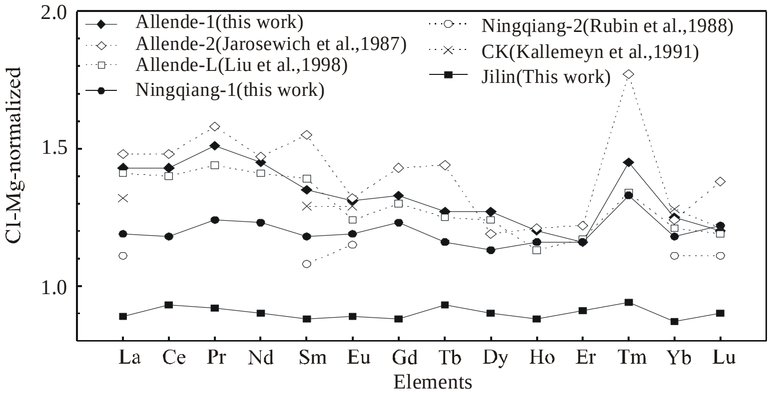

Fig. 1. REE patterns of Ningqiang, Allende and Jilin, normalized to their Mg concentrations and CI chondrites.
Ningqiang shows a low and flat REE pattern, different from those of Allende and mean CK-chondrites. The analyses of Jilin are also plotted for comparison.

Figure 2b demonstrates CI-Mg-normalized lithophile element patterns of Ningqiang in comparison with other meteorites. Most refractory lithophile elements are lower in Ningqiang than in Allende and mean CK chondrites. Other elements less refractory than Mg are almost undistinguishable among them. The Jilin ordinary chondrite is depleted in refractory lithophile elements, but enriched in alkali elements (Rb, K, Na) and Mn relative to Ningqiang, Allende and mean CK chondrites.

3 Discussions

3.1 Analysis accuracy of ICP-MS and ICP-AES

In this study, eight major and minor elements, i.e. Al, Ti, Ca, Mg, Fe, Mn, Na and P, were measured using ICP-AES. Their relative standard deviations of counting are lower than 6 % (Table 1). In order to assess accuracy of our analyses, deviations of our analyses of Allende relative to the recommended values were plotted in Fig. 3a, with comparison of other measurement data (Wakita and Schmitt, 1970; Hirota et al., 2002; Oura et al., 2002). Our data are well consistent with the recommended values (Jarosewich et al., 1987). In contrast, some of the other measurements (e.g. Ca, Fe) show significant differences from the recommended values.

Figure 3b plots deviations of 31 elements, including REE and other trace elements, of Allende determined in this work using ICP-MS relative to the recommended values, demonstrating agreement of our analyses with the latter. Our analyses of most major and minor elements (except for K, V and Co) and REE (except for Dy and Lu) are consistent with the recommended data within 10%, and those of the other elements (except for Pb) are less than 20%. The significant discrepancies of K, V and Pb could not be related to interferences of some compounds (e.g. ^{51}V by $^{35}Cl^{16}O$), because that the analyses are lower than the recommended values.

Our analyses of REE of Allende show a smoother pattern in comparison with the recommended data (Fig. 1), probably suggesting more representative of the former than the latter. Especially, the recommended data have a large positive anomaly of Tm, which is difficult to be explained. REEs of Allende and other CV chondrites are predominantly enriched in CAIs. MacPherson et al. (1988) summarized 5 types of REE patterns of CAIs, and only Group II shows an anomaly of Tm that is less enriched than LREE. Liu et al. (1998) reported concentrations of REEs of Allende, which are well consistent with ours (Fig. 1). In addition, the accuracy of our analyses can also be assessed by the rather flat REE pattern of Jilin (Fig. 1).

3.2 Classification of Ningqiang

Classification of Ningqiang is a controversial issue. Rubin et al. (1988) first analyzed the bulk composition of Ningqiang, and noted its relatively similar abundances of common and moderately volatile lithophiles and

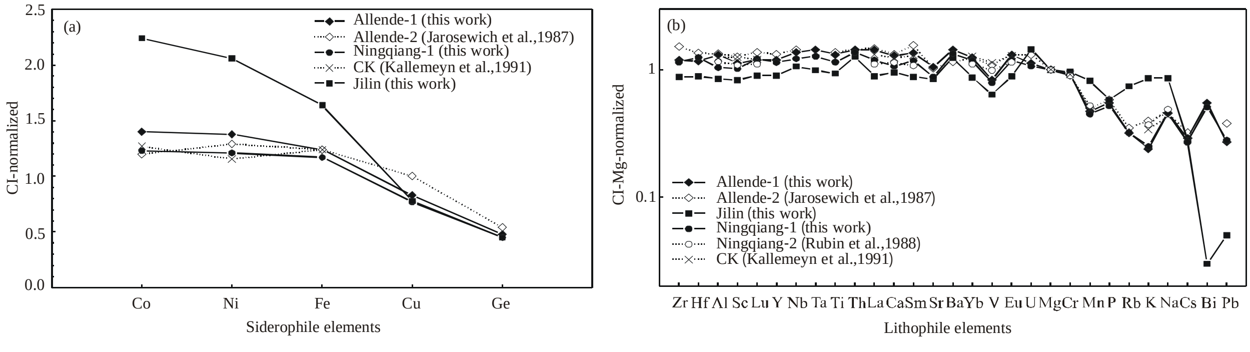

Fig. 2. Bulk composition of Ningqiang in comparison with Allende, mean CK chondrites and Jilin.
(a) CI-normalized siderophile element patterns. Elements range from left to right in order of increasing volatile; (b) CI-Mg-normalized lithophile element patterns, being relatively flat for refractory elements and decreasing toward more volatile elements.

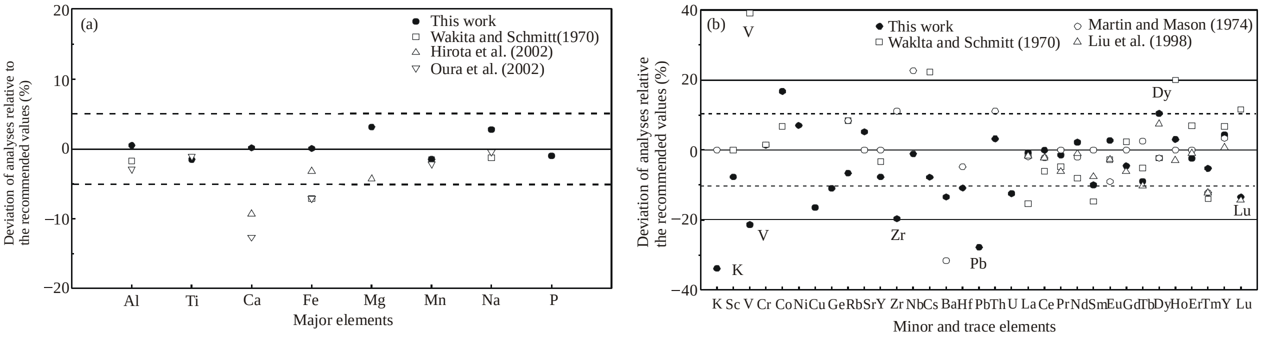

Fig. 3. Comparison of our analyses of Allende with literature data.

(a) Major and minor elements determined in this work using ICP-AES are well consistent with the recommended values (Jarosewich et al., 1987), in comparison with the significantly lower values of Ca and Fe reported by Hirota et al. (2002) and Oura et al. (2002); (b) Deviations of our analyses of 31 minor and trace elements relative to the recommended values. The analyses by other groups are also plotted for comparison.

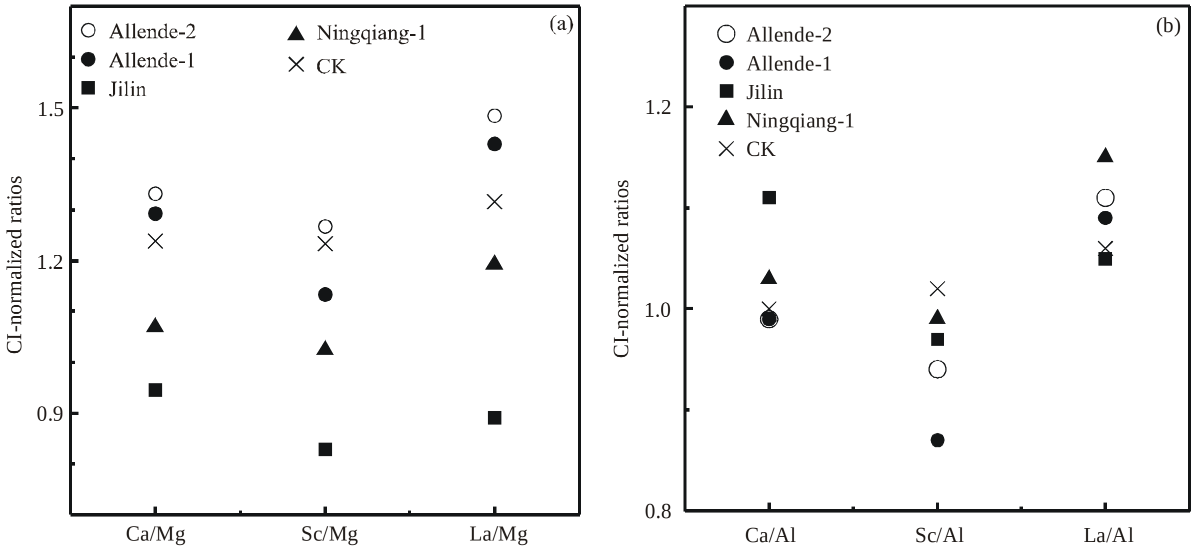

Fig. 4. Refractory lithophiles elements of Ningqiang in comparison with Allende, mean CK chondrites and Jilin.

(a) CI-normalized ratios of elements to Mg; (b) CI-normalized ratios of elements to Al.

siderophile with those of mean CV chondrites. In addition, Lin and Kimura (1998) found significantly lower abundance of CAIs in Ningqiang than in CV chondrites. Hence, Ningqiang was classified as an anomaly CV chondrite. Kallemeyn et al. (1991) included Ningqiang in the CK group mainly based on its closer petrography with the latter than with the CV group. They classified Ningqiang as an anomaly CK chondrite, because its bulk composition was distinct from typical CK chondrites. Further study of the bulk composition of Ningqiang (Kallemeyn, 1996) suggested that it could not be classified into any known chemical group of carbonaceous chondrites.

Although Kallemeyn et al. (1991) reported that the petrographic properties of Ningqiang are similar with CK chondrites, other previous studies demonstrated that Ningqiang was closer to Allende and other oxidized CV3 chondrites based on its large and abundant chondrules, high magnetite/metal ratio, oxygen isotopic composition (Weisberg et al., 1996; Kimura et al., 1997), and the induced thermoluminescence (TL) properties (Guimon et al., 1995). The abundance of CAIs is lower in Ningqiang (Rubin et al., 1988; Lin and Kimura, 2003) than in typical CV chondrites. However, petrographic types of CAIs commonly found in Allende were reported in Ningqiang too (Lin and Kimura, 2003). Consistent with previous studies, our analysis of the bulk Ningqiang sample is distinctly different from that of Allende (Figs. 1 and 2). The REE abundances of Ningqiang ($1.19\times$CI) are significantly lower than those of Allende ($1.35\times$ CI). Furthermore, Ningqiang shows a relatively flat REE pattern (La/Lu = $0.98\times$CI), distinct from the enrichment of LREE relative to HREE of Allende (La/Lu = $1.19\times$ CI, Fig. 1). The lower abundances of REEs of Ningqiang than those of Allende and mean CK chondrites are consistent with its relatively lower abundance of CAIs (~2.5 vol%) (Lin and Kimura, 2003), in comparison with Allende (CAIs ~9.4 vol%) (McSween, 1977) and mean CK chondrites (CAIs ~4 vol

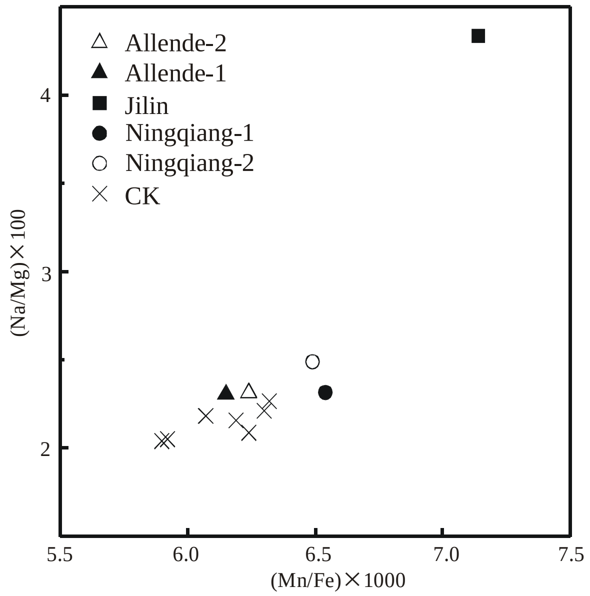

Fig. 5. Plots of moderately volatile element ratios of Mn/Fe vs. Na/Mg of Ningqiang, in comparison with Allende, mean CK chondrites and Jilin.
Filled symbols are this work; others are from Jarosewich et al. 1987 (Allende), Rubin et al. 1988 (Ningqiang) and Kallemeyn et al. 1991 (mean CK chondrites).

%) (Wang et al., 2005). However, this possibility cannot explain the different REE patterns of Ningqiang and Allende. The REE pattern of Ningqiang is closer to mean CK chondrites than to Allende, but their differences are still significant (Fig. 1).

Other refractory lithophile elements are also lower in Ningqiang than in Allende and mean CK chondrites (Fig. 2b), consistent with REEs. Figure 4a plots CI-normalized Ca/Mg, Sc/Mg and La/Mg ratios of Ningqiang in comparison with Allende, mean CK chondrites and Jilin. It is noticed that the ratios of Ningqiang are lower than those of Allende and mean CK chondrites. In addition, relative ratios of refractory lithophile elements, e.g. Ca/Al, Sc/Al and La/Al, confirm the differences between Ningqiang and Allende (Fig. 4b). The lower abundances of refractory lithophile elements of Ningqiang could be related to its depletion of CAIs.

Siderophile elements of Ningqiang are slightly lower than those of Allende, and are nearly the same as mean CK chondrites (Table 1, Fig. 2a). However, the ratios of Fe/Ni, Co/Ni, Cu/Ni and Ge/Ni of Ningqiang are slightly higher than those of Allende, but lower than CK (only available for Fe/Ni and Co/Ni). Figure 5 shows moderately volatile element ratios of Ningqiang, in comparison with Allende, mean CK chondrites and Jilin. The Mn/Fe ratio is higher in Ningqiang than in Allende and CK chondrites, while the Na/Mg ratios of them are nearly the same.

4 Conclusions

We analyzed meteoritic materials using ICP-AES for major and minor elements and using ICP-MS for trace elements. Our analyses of the Allende standard powder are consistent with the recommended values within 10% for major, minor and most trace elements, and within 20% for other trace ones. Only K, V and Pb have larger discrepancies, which could not be related to interferences of some compound ions because of their lower values. Furthermore, we determined a smoother REE pattern of Allende than the recommended data, suggesting that our analyses may be more representative than the latter. This is confirmed by consistence of our analyses with the previous result of Liu et al. (1998).

Forty-one elements of the bulk Ningqiang sample were analyzed using ICP-AES and ICP-MS, in order to assess its classification. The sample used in this work was an aliquot from a large mass of the Ningqiang powder, hence our analyses are more representative than previous results. This is important because most of refractory lithophile and siderophile elements are enriched in CAIs that distribute highly heterogeneous in the meteorite. The ratios of refractory lithophile elements to Mg are lower in Ningqiang (1.19× CI) than in Allende and CK chondrites (1.35 and 1.25× CI, respectively). These differences could be related to the lower abundance of CAIs in Ningqiang than those of CV and CK chondrites. However, Ningqiang shows a relatively flat REE pattern, different from the enrichment of LREE relative to HREE of Allende. Furthermore, Ningqiang contains lower siderophile elements than Allende, but close to mean CK chondrites. In addition, the ratio of Mn/Fe of Ningqiang is higher than those of Allende and CK chondrites. Although Ningqiang shows a close petrography to CV chondrites, its bulk composition is distinctly different from the latter. The Ningqiang meteorite cannot either be included in the CK group. We suggest listing Ningqiang as an ungrouped carbonaceous chondrite.

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