CHINESE JOURNAL OF GEOCHEMISTRY Vol. 26 No. 1 2007

DOI: 10.1007/s11631-007-0028-4

A study on the environmental conditions of the microsparite (Molar-tooth) carbonates

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Received December 14, 2005; accepted January 16, 2006

Abstract Along with the progress in research on the Precambrian, Molar-tooth carbonates (simplified as MT, or microsparite carbonates or MT structure) which were formed in the Middle-Late Proterozoic have become a hot subject recently. The Proterozoic Molar-tooth (MT) carbonate rocks refer to those Meso- to Neoproterozoic (1600–650 Ma) carbonates with MT structure, i.e., a series of peculiar, ptygmatically folded and spar-filled cracks in fine-grained carbonates of Precambrian age, located in the environment of mid- to inner ramp and shallow platform. MTS, like a bridge connecting the inorganic world with the organic one, are closely related to the evolution of paleo-oceans, atmosphere and biosphere. Their development and/or recession are/is related to the origin of life and the abruption of sedimentary geochemistry events of marine carbonates. By using modern instruments and testing methods adequately, the contents of oxides in sandstones were measured and the REE distribution pattern curves were established; an accurate value of isotopic ratio of $^{87}Sr/^{86}Sr$ was obtained, that is, the age of MT formation is about 750–900 Ma; C and O isotopes of some fresh micrite limestone samples were analyzed; the energy spectrum analysis revealed that the MT consists mainly of microspar calcite, while as for its chemical composition, the matrix shows outstanding peaks of Ca, Mg, Al, Si, and K. The geochemical indicators proved that Neoproterozoic MT carbonates in the Jilin-Liaoning region were developed at the margin of a stable continent, in the torrid zone where the paleo-temperature was about 50℃, the seawater had normal salinity when MT was formed during the Wanlong period in southern Jilin and during the Yingchengzi and Xingmincun periods in eastern Liaoning. The sedimentary environment was located in the inner ramp. In summary, it is of great importance to understand the origin of MT, ascertain the paleo-climate and paleo-environment characteristics, constrain the age and the stratigraphic division and comparison of the Proterozoic so as to study the geochemical characteristics of MT carbonates and their formation environment.

Key words Molar tooth; microsparite carbonate; geochemistry; Jilin-Liaoning region; Neoproterozoic

1 Introduction

MT carbonates have been found in the strata of over 40 regions in Canada, U.S.A., Europe, Africa, Australia, Siberia, China, etc. (James et al., 1998), including China's Hunjiang area in Jilin, Benxi, Fuzhou, and Jinzhou in Liaoning, Yanshan of Beijing, as well as Shandong, Henan, Anhui and northern Jiangsu provinces since it was first used by Bauerman (1885) to describe carbonates "resembling the marking on the molar tooth of an elephant" in the Belt supergroup near the Canada-USA border in western Montana. To compile and confirm the literature from more than 40 formations in 26 localities around the world, MT is temporally restricted (James et al., 1998). MT is rarely recognized or exists in the Achaean and Early Proterozoic or Phanerozoic strata. There are only 2 examples from the Cretaceous (Rossetti, 2000) and Cambrian (Qiao Xiufu and Gao Linzhi, 1999), which convinced what had been found is not MT but a marker of Middle-Late Proterozoic (1600–650 Ma). Most of the MT structures were developed in the shallow water carbonates of Meso-to-Neoproterozoic age. The MT declined abruptly during the Varanger/Marinoan glaciation before 600 Ma, which is thought that MT was restricted to the Late Precambrian (Shields, 1999,

Table 1. Distribution of MT carbonates in the Jilin-Liaoning region

Section	No.of Micro-cycles	Percentage of total cycles (%)
Wanlong Fm., Erdaojiang	19	19.59
Wanlong Fm., Laoning	44	18.41
Nanguanling Fm., Huagou, Dalian	42	37.50
Yingchengzi Fm., Qipanmou	69	36.70
Xingmincun Fm., Jinshitan	30	61.22

2002; Smith, 1968). Further evidence may extrand the temporal range of Molar-tooth structure, but our update viewpoint is that this fabric structure is the characteristic feature of Meso-Early Neopro- terozoic (1700–750 Ma) carbonates, most rare in older and younger strata. In this study area the MT-bearing Neoproterozoic successions in eastern Liaoning Peninsula and southern Jillin Province were involved. The MT-rich formation is listed in Table 1. Research on MT is of important significance both in theory and application, which will help to solve some problems encountered in Precambrian research. This study attempts to shed light on the forming conditions of MT from the geochemical points of view.

2 The tectonic setting of formation of the Molar-tooth carbonates

The contents of oxides in sandstones and the REE distribution pattern curves indicate that the Neoproterozoic MT carbonates in the Jilin-Liaoning region were developed at the margin of a stable continent.

2.1 The relationship between the contents of oxides in sandstones and the tectonic environment

By comparing the statistics data on the chemical composition of 23 clastic samples (the data from the Liaoning Regional Geological Survey Team) with the standard of Bhatia (Liu Xiufeng, 1991), all the parameters for the samples fall in the range of the margin of the passive continental environment. This shows the study area is located at the margin of a passive continent during the Neoproterozoic.

2.2 The relationship between REE and the tectonic environment

The features of the REE in the strata at the margin of the passive continent are: (1) LREE enrichment; (2) smooth HREE distribution patterns; (3) ubiquitous negative Eu anomaly. The features of the REE in the strata at the edge of the active continent are: 1) varying LREE enrichment degree; 2) medium HREE abundance; and 3) ubiquitous negative Eu anomaly, varying from 0.6 to 1.0 (Zhao Zhenhua, 1997). In region of Dalian (in the eastern part of Liaoning), no matter the samples are carbonates, sandstones or shales, their REE features LREE enrichment and negative Eu anomaly, indicating that they underwent medium-strong REE fractionation, though the HREE fractionation is weak, which is coincident with the REE characteristics of sediments at the margin of the passive continent (Fig. 1).

The tectonic setting of the MT carbonates in the Jilin-Liaoning region is very similar to that of other places in the world where MT is developed, such as the shallow ramp of the Neoproterozoic in the Flinders Ranges, Australia; the stable cratonic basin in western Africa, the Russian platform and the northern continent of eastern Europe evolved as a paleo-craton during the Neoproterozoic. As far as the MT-bearing strata in northern America and Canada, for example the Belt Supergroup, were considered as shallow-sea cratonic basin on the edge of passive continent (Pratt, 1998; James et al., 1998; Meng Xianhua et al., 1995).

3 The specific time range of MT carbonates in the Jilin-Liaoning region

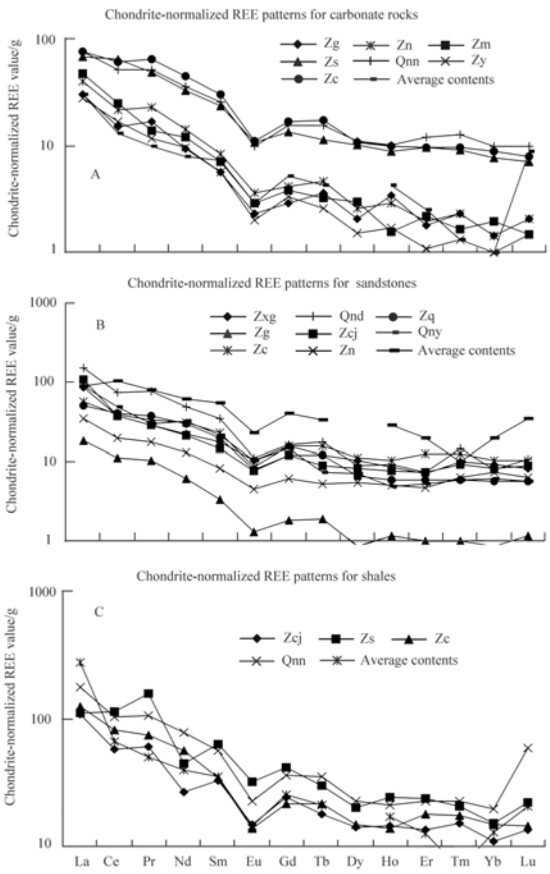

Fig. 1. Contents of lanthanum and chondrite-normalized REE patterns (A, B and C).

As far as the study on the Proterozoic stratigraphy in China is concerned, a large age difference from several decades of million years to several hundreds of million years (Ma) would exist for the Proterozoic strata if different age determination methods were used. For example, for the same Proterozoic strata, in the 1970s and 1980s, the glauconite ages obtained were less than 500 Ma and even reached 400 Ma, this warp might be caused by the paulopost reworking of glauconite (Meng Xianghua and Ge Ming, 2005; Yu Chunshui and Gu Lianxing, 2000). And then in the 1990s, better Pb/Pb isochron ages were obtained than the previous ages. By this method, the age of the Xingmincun Fm. and Wanlong Fm. of Neoproterozoic era is 600–700 Ma or 670±50 Ma. And recently, comparing the values measured by the $^{87}Sr/^{86}Sr$ with the values in isotopic age curves presented by Graham shields (Fig. 2), we find that most of the values are 700 Ma, or more than 700 Ma or 800 Ma. So the temporal distribution of the Molar-tooth carbonates could vary from 650 Ma to 700–800 Ma. Using Sr and C isotope to measure the geological age is a new method in recent years. Generally, the ratio of Sr in the global sea is considered to be invariable during any geological time, but this ratio is not same in different period; the C isotopic ratio mainly reflects the pure burial speed of organisms with time goes by. In recent years, geologists in the world measured lots of Precambrian carbonate samples and a relative credible Sr and C isotopic change curve of late Precambrian seawater has been set up (Asmerom et al., 1991; Kaufman and Knoll, 1995; Braiser et al., 1996; Montanez et al., 1996; Yang Jiedong et al., 2000; Shields, 1999, 2002), which established the basis for global Precambrian stratigraphy correlation. The $^{87}Sr/^{86}Sr$ isotopic ratio is obtained accurately to estimate stratigraphic age, e.g. the age of Sino-Korean plate may be earlier than that now due to Sr isotope. Yang Jiedong et al. (2000, 2001) and Zheng Wenwu et al. (2004) made a Sr and C isotope comparison study between the southern Liaoning and the northern Jiangsu and Anhui using the Sr and C isotope change curve diagram in the seawater. We studied the Sr and C isotope of Wanlong Formation in the southern Jilin and compared the age value of the Wanlong Formation, the Badaojiang Formation and the Qinggouzi Formation with the Sr and C isotopic age curve from Shields (1999). The result shows that the ages of the Wanlong Formation, the Badaojiang Formation and the Qinggouzi Formation are up to 700–850 Ma. Therefore, we could

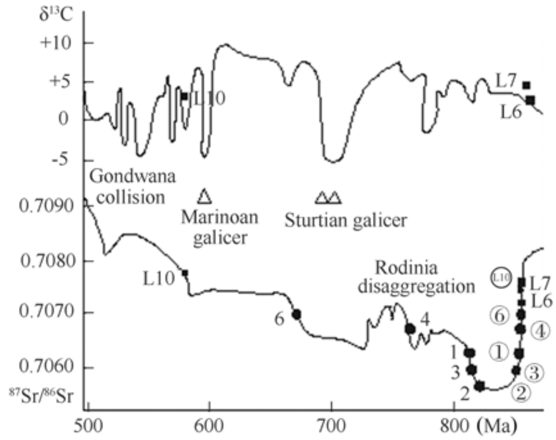

Fig. 2. Evolution map of Sr and C isotope in the sea water of Cambrian to Neoproterozoic (the initial map from Shields, 1999). ■ shows Sr and C isotopic data from southern Liaoning Province (Zheng Wenwu et al., 2004) and ● represents points of fall of Neoproterozoic in the southern Jilin Province; △ represents the samples from southern Liaoning; circles with numbers are the codes of the second fall points of all samples.

Table 2. Tectonic environments and chemical compositions of sandstones in eastern Liaoning

Oxide and related parameter	Oceanic island arc	Continental island arc	Active continental margin	Passive continental margin	Fuzhou of Dalian
SiO_2	58.83	70.69	73.86	81.95	88.9
TiO_2	1.06	0.64	0.46	0.49	0.27
Al_2O_3	17.11	14.04	12.89	8.41	4.83
Fe_2O_3	1.95	1.43	1.30	1.32	1.34
FeO	5.52	3.05	1.58	1.76	0.88
MnO	0.15	0.10	0.10	0.05	0.04
MgO	3.65	1.97	1.23	1.39	0.29
CaO	5.83	2.68	2.48	1.89	0.56
Na_2O	4.10	3.12	2.77	1.07	0.29
K_2O	1.60	1.89	2.90	1.71	1.49
P_2O_5	0.26	0.16	0.09	0.12	0.05
TFe_2O_3+MgO	11.73	6.79	4.63	2.89	1.63
Al_2O_3/SiO_2	0.29	0.20	0.18	0.10	0.05
K_2O/Na_2O	0.39	0.61	0.99	1.60	5.14
$Al_2O_3/(CaO+Na_2O)$	1.72	2.42	2.56	4.15	5.68

Table 3. The paleo-temperature and paleo-salinity when MT carbonates were formed

Code	Section	Lithology	$\delta^{13}C_{PDB}$ (‰)	$\delta^{18}O_{PDB}$ (‰)	*Z*	*T* (℃)
KQ20030221	Wanlong Fm., Dayangcha	Micritic limestone（matrix）	3.30	-6.80	130.67	52.51
KQ20030222	Wanlong Fm., Dayangcha	MT limestone（microsparite）	2.80	-5.30	130.40	43.21
KQ20030223	Wanlong Fm., Erdaojiang	Micritic limestone（matrix）	3.20	-8.20	129.77	61.85
KQ20030226	Wanlong Fm., Erdaojiang	MT limestone（microsparite）	3.40	-6.50	131.03	50.60
KQ20030225	Wanlong Fm., Erdaojiang	MT limestone（microsparite）	3.60	-7.10	131.14	54.46
L-6	Yingchengzi Fm., Qipanmou	Micritic limestone	4.74	-6.80	133.63	52.48

Note: Sample L-6 from Zheng Wenwu et al. (2004), and other samples measured by National Experimental Test Center of the Chinese Academy of Geological Sciences.

deduce that MT carbonates of Jilin formed during 750–900 Ma.

4 The paleo-temperature at the time of formation of MT carbonates

We selected some fresh micrite limestone samples, with no diagenetic vein and no recrystallization, to analyze their C and O isotopes (including MT and its matrix). It was discovered that it was a torrid zone and the paleo-temperature was about 50℃ (Table 2), the seawater had normal salinity when MT was formed during the Wanlong period in southern Jilin, and during the Yingchengzi and Xingmincun periods in eastern Liaoning (Table 3).

In general, most of the researchers use some experimental indices to confirm whether the C isotopic composition of oceanic carbonate sediments varies greatly or not after they are buried, in consideration that the C isotopic composition may change during a series of post-depositional processes (Kaufman and Knoll, 1995). The sample is invaluable if the O isotopic value is less than -10‰ (PDB) (Zhou Chuanming, 1997). Oxygen isotopic values are up to -10% (PDB) in this work, so the result is credible in Table 3. The value of Z shows the paleo-salinity significance of MT formation. Carbon isotopes are difficult to exchange and are stable when the old carbonates are formed. The value of Z is determined mainly by $\delta^{13}C$ and it can be expressed as the following equation:

$$Z=2.048\ (\delta^{13}C+50)+0.498\ (\delta^{18}O+50)$$
(Keith and Weber, 1964)

where $\delta^{13}C$, $\delta^{18}O$—PDB standard; Z>120 for marine limestone; Z<120 for fresh water limestone.

The result showed that the seawater had a normal salinity when MT carbonates were formed.

Zheng Wenwu (1994) thought that the paleo-temperature was about 40℃ when the Huainan Group was formed. The formation of the Huainan Group took palce just at the time the MT structure developed very well, illuminating MT was formed in the torrid zone condition. The paleo-temperature and paleo-salinity varied from one period to another in the Meso-Neoproterozoic in the Yanshan Basin (Zhu Shixing et al., 1994). The southern Jilin platform was also located in northern hemisphere at the lower latitude, a large amount of algae occurred in the torrid zone during the Wanlong and Badaojiang periods (Bi Shouye et al., 1995); at the same time, strata in eastern Liaoning Peninsula were located in the lower-latitude area, too (Meng Xianghua and Ge Ming, 2005).

5 Compositional characteristics of MT-bearing limestones and their sedimentary environment

5.1 Energy spectrum analysis

The energy spectrum analysis indicated that MT is composed mainly of microspar calcite. CaO is absolutely dominant and its lowest content is not less than 97%, with minor Sn (Table 4). One or two samples contain small amounts of SbO, MgO in the Wanlong and Nanguanling formations and samples from the Wanlong Formation of Laoning and the Yingchengzi Formation in Dalian contain a small

Table 4. Composition of MT revealed by energy spectrum analysis in the study area

Section	MgO	SiO_2	CaO	FeO	SnO	SbO	No. of samples
Wanlong Fm., Erdaojiang			98.25		2.07	3.42	13
Wanlong Fm., Laoning	0.79	1.76	97.59	1.1	1.64	3.57	14
Nanguanling Fm., Huagou, Dalian	0.8		98.76		1.73	2.87	12
Yingchengzi Fm., Qipanmou			99.2	1.13	1.71		14
Xingmincun Fm., Jinshitan			99.32		1.87		11

amount of FeO. It is worth noting that the chemical composition of the MT is relatively complex. In addition to calcite, it also contains SiO_2, MgO, FeO, SnO, etc (Table 5). In combination with microscopic observation, it was found that collapse occurred when sediments were deposited. Both the matrix and MT were not solid when the collapse occurred. As a result, the material of the matrix found its way into MT bands. This phenomenon can be observed very often in the lower part of the Wanlong Formation of the Laoning section. In general, Molar-tooth structure limestone differs significantly in composition and texture from the matrix, which is composed mainly of impure micritic calcite (normally with a lot of mud and silty particles, while Molar-tooth is composed of pure microspar calcite with even texture (the microspar calcite is measured at about 0.01 mm in diameter). According to the energy spectrum analysis of the matrix and MT samples, the matrix shows outstanding peaks of Ca, M, Al, Si and K, while the MT is marked only by the peak of Ca.

The matrix changes regularly in chemical composition along with the seawater from shallow to deep from the Wanlong Formation to the Xingmincun Formation, for instance the contents of Ca vary from low to high and then decline again, and those of Si and Mg vary from high to low and then increase again. In summary, the contents of terrigenous components, such as SiO_2 and Al_2O_3 are higher, the contents of FeO are also high in the Wanlong Formation in southern Jilin, indicating that the forming environment of MT is close to the provenance, and the seawater was relative deep when the MT was deposited. A lot of graded bedding and the great-type storm hammoeky cross bedding also implies that the MT was developed in the mid-deep ramp under the control of storms. Compared with the MT of Jilin, the contents of terrigenous components of sediments are low, those of pyrite are low, too, and the sedimentary structure is mainly small-type hammoeky cross bedding. All this shows that the sediments are far away from the provenance and lie in the shallow-middle ramp where many sediments were deposited in the fine weather. The sea gradually changed from open to semi-closed and completely closed, e.g., both the Yingchengzi and Xingmincun formations are located in the semi-cutoff environment. Nevertheless, calcites account for 97% of the chemical components of the MT in Jilin or Liaoning.

5.2 Electron microprobe analysis

For further studying the compositional features of MT carbonates, we selected calcisiltite samples from the Yingchengzi Formation and bearing-MT marl samples from the Xingmincun Formation. By comparing the MT microspar with the marl matrix, no Mn or K was found, the contents of Mg, Al and Si greatly decreased in the MT; similar situation is noticed for other components. The chemical compositions are: 200×10^{-6} Sr, 600×10^{-6}–1200×10^{-6} Fe, 200×10^{-6} Mn, 200×10^{-6}–600×10^{-6} S, 1900×10^{-6}–7600×10^{-6} Mg, Al and Si for the samples from the Xingmincun Formation. The samples from the Yingchengzi Formation may be influenced by post-diagenesis because they were exposed and leached for a long period of time so that there is an insignificant difference between MT and the calcisiltic matrix. This still reflected the medium condition at the beginning of sediment deposition, for instance the ratio of Mg over Ca is low, it contains a part of clay minerals and has a small amount of pyrite, which shows the rocks were formed in a reducing environment to some extent.

Bourrouilh-LE Jan et al. (2002)[1] and Demange Michel et al. (2003)[2] examined some samples from the Neoproterozoic strata in the Jinshitan Section in Dalian and the Erdaojiang Section in Jilin. Our results are consistent with theirs.

Our study shows that the MT was formed in a specific geochemical environment. The composition of the MT in the Yinchengzi Formation is close to that in the Helena Formation in the Belt supergroup, Canada, but is not so similar to that in the Xingmincun Formation, which implicates that the environment where the MT was formed is not invariable for ever. That is, the MT formation needs a stable environment. The Earth evolution is stable, before the continent was of disaggregation; the place in which MT was developed is located at the low latitude in terms of the paleo-geomegnetism data; the environment is fit for the flourishing growth of algae and acritarch florae. All these created good conditions for the development of MT carbonates.

6 The formation conditions of MT reflected by biomarkers

Biomarker analysis showed that organisms in the MT carbonates deposited during the Wanlong and Xingmincun periods are very abundant, such as prokaryotic aerobic and anaerobic bacteria, blue

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1 Bourrouilh-LE Jan F.G. et al. (2002) Electron microprobe analysis of Molar Tooth structure from Dalian, Liaoning Province, China. In *New Research of UNESCO/IUGS/IGCP447 Beijing Meeting* (eds. Meng Xianghua et al.) [C]. pp.26.

2 Demange Michel et al. (2003) Geochemical cartography of Molar-tooth structures from Precambrian of Jilin Province, North China. In *New Research of Microsparite (MT) Carbonates and Evolution of the Earth in the Prote- rozoic* (ed. Secretariat of IGCP447) [C]. pp.9.

Table 5. Composition of the matrix revealed by energy spectrum analysis in the study area

Section	Lithology	Facies	Na_2O	MgO	Al_2O_3	SiO_2	SO_2	K_2O	CaO	Ti_2O	FeO	SnO	No. of samples
Wanlong Fm., Erdaojiang	MT-calcisiltite	Mid-deep ramp	0.82	8.23	10.15	46.91	0.70	3.46	24.58	0.80	3.80	1.86	7
Wanlong Fm., Laoling	MT-micritic limestone	Middle ramp	0.85	1.59	5.62	34.90		1.81	51.95	0.88	2.16	1.93	9
Nanguanling Fm., Huagou	MT-micritic limestone	Shallow ramp		1.01	2.38	8.53		1.54	81.55		1.06	2.91	7
Yingchengzi Fm., Qipanmou	Calcisiltite-bearing MT	Shallow ramp		0.80	1.91	5.83		1.04	87.12		1.64	2.56	9
Xingmincun Fm. , Jinshitan	MT- marl	Middle ramp		11.57	3.08	16.91	0.81	1.31	63.84		1.44	2.22	9

Note: Analyzed by National Experimental Test Center of the Chinese Academy of Geological Sciences.

Table 6. Biomarker distribution in Neoproterozoia microcalcsparite (MT) carbonates from the Jilin-Laoning area

Code	Normal alkane	Main peak of *n*-alkane	Pr/Ph	CPI	Acyclic isoprenoid	Main peak of acyclic soprenoid	Alkylcyclohexane	Alkylmethyl-cyclohexane
1	nC_{15}-C_{36}	nC_{18}, nC_{24}	0.94	1.12	iC_{16}, iC_{18}-iC_{36}	iC_{19}, iC_{27}	C_{15}-C_{33}	C_{15}-C_{32}
2	nC_{14}-C_{35}	nC_{17}, nC_{25}	1.13	1.12	iC_{16}, iC_{18}-iC_{36}	iC_{19}, iC_{27}	C_{15}-C_{33}	C_{14}-C_{33}
3	nC_{15}-C_{34}	nC_{18}, nC_{23}	0.77	1.14	iC_{16}, iC_{18}-iC_{36}	iC_{19} , iC_{27}	C_{16}-C_{31}	C_{15}-C_{33}
4	nC_{15}-C_{34}	nC_{17}, nC_{24}	0.99	1.13	iC_{16}, iC_{18}-iC_{36}	iC_{19} , iC_{27}	C_{15}-C_{32}	C_{15}-C_{31}

bacteria and phytoplankton as well as eukaryotict algae (green algae, red algae) and fungi (Table 6). This is consistent with the results of author's study on MT sedimentology, i.e., the Pr/Ph ratios of 0.77–0.99 in the Wanlong Formation indicated a normal sea environment of poor reduction, while the Pr/Ph ratios of 0.94–1.13 in the Xingmincun Formation showed an oxidizing normal sea environment of poor reduction.

It may be concluded that MT is the result of bacterial and algal activities, which is a kind of organisms between aerobic and anaerobic bacteria, being reproduced well in normal or saline seawater under weak redox conditions. It made calcite deposited quickly into microcalcsparite under the influence of some mechanism (Kuang Hongwei et al., 2004).

In summary, it is not only of great importance to understand the origin of MT, but also to ascertain the paleo-climate and paleo-environment characteristics and constraining the age and the stratigraphy division and comparison of the Proterozoic so as to study the geochemical characteristics of MT carbonates and their formation environment.

Acknowledgements This Project was funded by the International Geological Comparative Program 447 (IGCP447) (No. SC/GEO/546/447), the National Natural Science Foundation of China (Grant No. 40172043) and the Natural Science Developmental Fund of the Yangtze University.

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