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Constraints from strontium and neodymium isotopic ratios and trace elements on the sources of the sediments in Lake Huguang Maar

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ABSTRACT

The sediments in Lake Huguang Maar in coastal South China were previously thought to originate mainly from wind-blown dust transported from North China, such that the lake sediments recorded the varying strength of the Asian winter monsoon. An alternative explanation was that the local pyroclastic rocks supplied the lake sediments, but the actual contributions from the different sources remained unclear. Geochemical analyses including $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ and trace elements support the local pyroclastic rock as the dominant source: <22% of the total Sr in the lake sediments and ~17% of the Nd arises from the distant source. Nb/Ta and Zr/Hf for the lake sediments are identical to those for the local rock but differ from the ratios for the wind-blown dust, and chondrite-normalized rare earth element patterns for the lake sediments are similar to those for the local rock and soil, but differ from those for the distant source. The sediments in Lake Huguang Maar are probably input into the lake through runoff and thus controlled by the hydrology of the lake. Wind-blown dust transported by the Asian winter monsoon from arid North China is only a minor contribution to the sediments.

Introduction

The Asian monsoon is an important component of the global climate system. Investigation of the Asian monsoon variation has long been one of the major goals for paleoclimatic studies, and reconstruction of the paleo-Asian monsoon based on the loess-paleosol series in the Loess Plateau in North China has been informative (Liu, 1985; Liu and Ding, 1998; An, 2000). Nevertheless, these investigations suffer from uncertainties in the dating and the low deposition rate of aeolian sediments. Thus the loess has only been useful for studying climatic and environmental changes on multi-centennial or longer time scales. However, increasing evidence indicates significant variations of the Asian monsoon system on shorter decadal to interannual time scales (Charles et al., 1997; Kumar et al., 1999). It is necessary to understand these variations and their controlling mechanisms.

Although speleothem stable oxygen isotope ($\delta^{18}O$) records from monsoonal China (Wang et al., 2001; Yuan et al., 2004; Dykoski et al., 2005; Hu et al., 2008; Zhou et al., 2008a) provide an opportunity to explore the Asian summer monsoon variation on short time scales, similar reconstruction of the winter monsoon has proven less satisfactory. Recently, however, Yancheva et al. (2007a) provided such a record for the last 16,000 yr based on the Ti content of the sediments in Lake Huguang Maar in coastal South China. They suggested that the lake "receives a minimal quantity of material by runoff" and "acts as a natural sediment trap for dust delivered to the site by the northerly winds of the winter monsoon," and thus the Ti content of the sediments in the lake should reflect the strength of the winter monsoon. This suggestion was disputed by Zhou et al. (2007), who suggested that a local source might contribute more sediment and Ti in Lake Huguang Maar than wind-blown dust from remote North China. They suggested that the variation of the Ti content of the sediments might be related to the paleohydrology of the lake. This alternative interpretation was based on the abundance of coarse grains in the lake sediments, and on sedimentological evidence of a high flux of lithogenic materials to the lake (Wang et al., 2000). Neither Yancheva et al. (2007a) nor Zhou et al. (2007) presented detailed geochemical evidence such as $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios and rare earth element (REE) spectra, which are commonly used in sediment-source tracing (Cullers et al., 1988; Gallet et al., 1996; Jahn et al., 2001; Sun, 2005). Sr and Nd isotopic ratios and REE patterns of wind-blown dust and basaltic rock should contrast strongly, providing a useful test of the competing hypotheses (e.g., Dia et al., 2006; Kurtz et al., 2001). In addition, the sedimentological evidence

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Table 1
Grain size distribution (%) of the sediment samples collected from Lake Huguang Maar and comparison with typical wind-blown dusts from the Loess Plateau in North China.

Sample ID	Sand	Silt			Clay		Φ_{50} (μm)	M_z (μm)	Sediment type
	>63 μm	63–30 μm	30–16 μm	16–4 μm	4–2 μm	<2 μm			
HGY-4	15.51	19.13	25.04	25.25	6.52	8.55	19.6	18.6	Lake sediments
HGY-6	1.60	14.09	27.93	36.01	9.34	12.10	12.2	9.5	
HGY-7	7.61	21.18	26.73	28.35	6.87	9.27	15.2	13.7	
HGY-8	9.64	25.02	26.22	25.90	5.37	7.84	19.6	15.6	
HGY-9	53.65	12.05	14.83	12.71	2.66	4.08	136	100	
HGY-10	21.84	17.63	25.57	22.73	4.90	7.33	22.9	22.9	
L1-1	2.92	22.65	28.34	31.72	6.81	7.56	15.1	13.5	Aeolian sediments
L1-8	0.68	16.61	26.43	34.83	10.49	10.96	11.4	10.2	
L15-1	1.62	19.98	27.73	34.34	7.91	8.42	13.5	12.0	
L15-2	1.52	19.22	28.09	35.78	7.68	7.72	13.5	12.3	

Note. Φ_{50} is median grain size and M_z is mean grain size.

provided by Zhou et al. (2007) cannot determine whether wind-blown dust from North China has made a significant contribution to the sediments of Lake Huguang Maar, which may be resolved with some isotopic ratios such as $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ (Faure and Mensing, 2005). This is important not only for interpretation of the paleoclimatic and paleoenvironmental proxies archived in this lake, but also for modern economic and social development in coastal South China because a lithogenic flux to the lake as high as suggested by Yancheva et al. (2007a) is comparable with the modern mineral dust flux monitored on the Loess Plateau (Sun et al., 2003), and implies that coastal South China may be frequently affected by dust storm as was witnessed in North China in past decades (Chen et al., 2003).

Therefore, more geochemical investigation is needed to determine whether wind-blown dust from arid North China is an important source for the sediments deposited in Lake Huguang Maar. This is the main purpose of the present study. We focus on the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements of the sediments of Lake Huguang Maar, which give a strong support to the argument that the sediments in Lake Huguang Maar were provided mainly by local pyroclastic rock (Zhou et al., 2007) rather than by wind-blown dust transported by the Asian winter monsoon from remote arid North China (Yancheva et al., 2007a,b).

Geological setting

Yancheva et al. (2007a) provided a detailed description of the geological setting, which is summarized here. Lake Huguang Maar was formed in a volcanic crater situated in the north part of the Leizhou

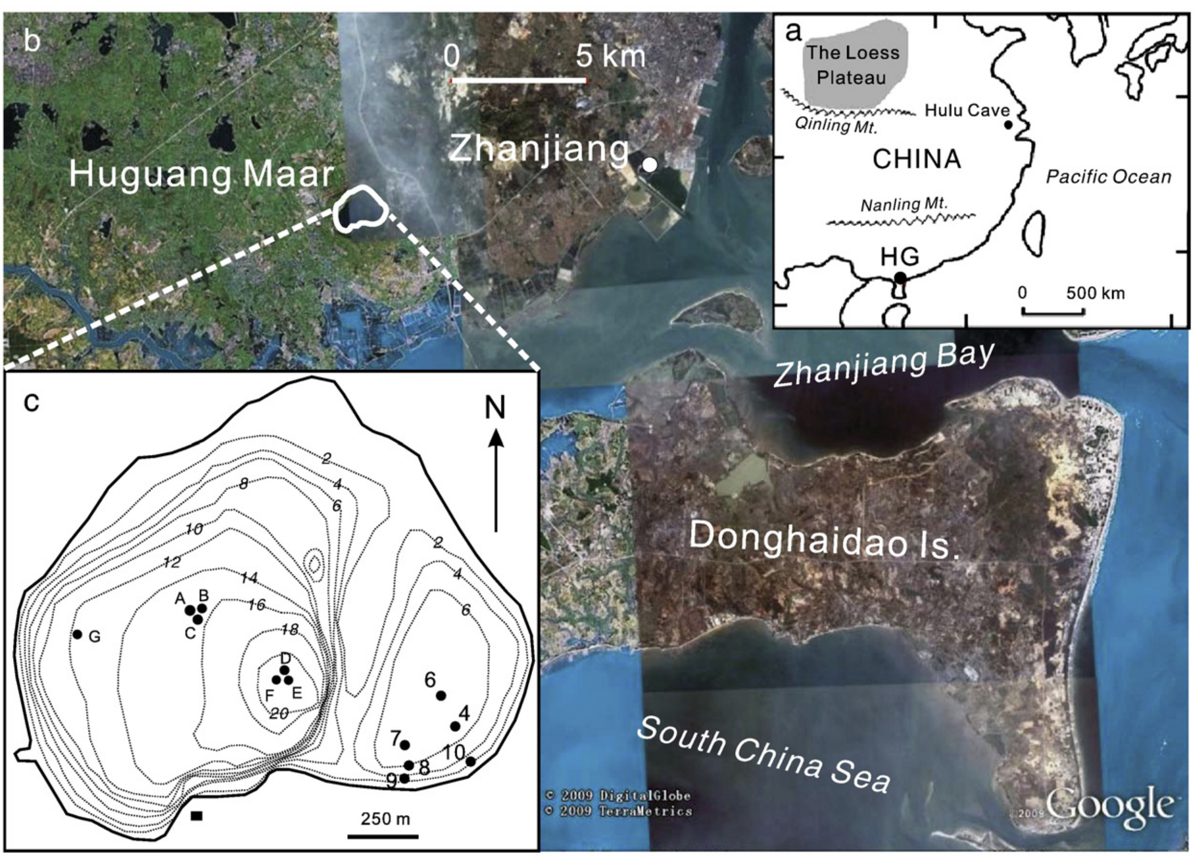

Figure 1. (a) Locations of Lake Huguang Maar (HG), Hulu Cave and the Loess Plateau in China. (b) A snapshot from Google Earth showing a part of coastal South China in which Lake Huguang Maar is located. (c) An enlargement of Lake Huguang Maar with isobath. A to G in the west part of the lake indicate the seven cores studied by Yancheva et al. (2007a). In the east part of the lake, the numbers 4, 6–10 near black solid circles indicate the sediment samples HGY-4, HGY-6 to 10, respectively. The black square indicates the naturally exposed profile of pyroclastic rock along the southwest margin of the lake. Three pyroclastic rock samples, SSB, SSM and SST were collected from this profile and the soil sample HGY-11 was collected in the woodland near this profile.

Peninsula in coastal South China (Fig. 1a). This volcanic crater may have formed early in the late Pleistocene (Feng, 1992; Huang et al., 1993). Lake Huguang Maar is small, with a surface area of only 2.25 km^2. Its catchment area is likewise small: 3.2 km^2. A south–north shoal, which may emerge during dry season, divides this small lake into two parts (Fig. 1c; Yancheva et al., 2007a). The catchment of the lake is densely vegetated at present. There is a large amount of pyroclastic rock surrounding the lake. Pyroclastic rock is commonly found in Quaternary volcanic deposits in the Leizhou Peninsula and Hainan Island and consists mainly of olivine, pyroxene, plagioclase, quartz, and volcanic glass (Huang et al., 1993). However, mineral analysis on the pyroclastic rock in the catchment of Lake Huguang Maar has not been reported before and it was complemented in this study (see the section "Mineralogy").

Coastal South China experiences a typical sub-tropical summer monsoon climate at present, with a high annual mean temperature

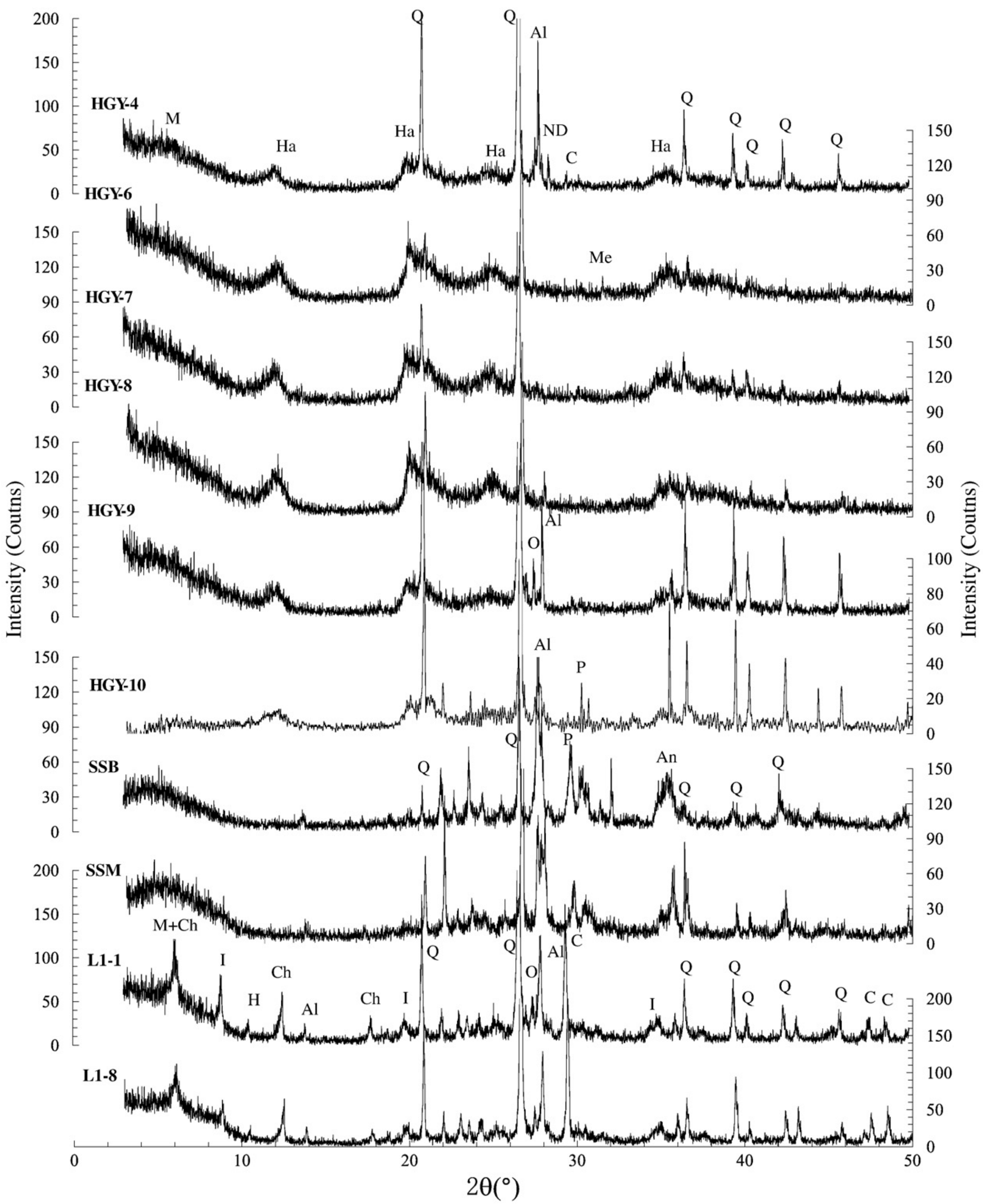

Figure 2. X-ray diffraction patterns of the bulk sediments recovered from Lake Huguang Maar and comparison with those of local pyroclastic rock and wind-blown dust from North China. M-Montmorillonite; Ch—Chlorite; I—illite; Ha—Halloysite; Q—Quartz; Al—Albite; An—Anorthite; P—Pyroxene; H—Hornblende; C—Calcite; O—Orthoclase; Me—Melilite; ND—Not determined.

Table 2
Minerals of the sediments of Lake Huguang Maar and comparison with those of local pyroclastic rock and wind-blown dust from North China.

	Wind-blown dust		Lake sediments						Pyroclastic rock	
	L1-1	L1-8	HGY-4	HGY-6	HGY-7	HGY-8	HGY-9	HGY-10	SSB	SSM
Quartz	23.1	25.0	42.1	15.2	30.4	13.9	60.7	63.0	7.8	18.8
Halloysite			21.6	80.8	69.6	80.6	13.4	17.3		
Orthoclase	14.4	21.2					9.6			
Albite	17.6	17.1	16.0			5.5	16.3	17.1		
Montmorillonite	7.5	4.0	13.4							
Illite	16.1	7.1								
Chlorite	10.7	12.3								
Calcite	7.6	11.4	4.0							
Hornblende	3.0	2.0								
Anorthite									48.5	34.7
Pyroxene								2.0	16.5	11.8
Melilite				4.0						
Not determined			2.8							

(AMT) and heavy annual mean precipitation (AMP) (The AMT and AMP at Zhanjiang are ~23°C and ~2100 mm (1951–1980), respectively. Data source: http://www.naturalresources.csdb.cn/index.asp). The majority of the precipitation (>90%) falls during the summer half year (from May to October) and much of the summer precipitation falls in strong convective weather or typhoon activity (Woo et al., 1997).

Sample description and analytical methods

In this study, the sediments in Lake Huguang Maar and local pyroclastic rock and soils were sampled and analyzed for $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements. A total of ten samples were collected, including six sediment samples retrieved from the eastern part of the lake, three pyroclastic rock samples obtained from a naturally exposed profile with a height of ~30 m along the southwest margin, and one soil sample collected in the woodland near this profile (Fig. 1c). Three pyroclastic rock samples were sampled at the top, middle and bottom part of the exposed profile, respectively, in an effort to represent an average of the pyroclastic rock. The sediments were recovered using a tube of bamboo with an inner diameter of ~8 cm. Sampling depth was ~20 cm below the lake floor, suggesting that the sediments recovered correspond roughly to the last 200 yr based on the sedimentation rate estimated by Yancheva et al. (2007a). These sediment samples were collected along two transects at different distances from the lake margin (Fig. 1c). This sampling approach was adopted in order to check (1) whether the grain size of the sediments decreases with distance, which is a feature of lacustrine sediments input through runoff (Nanjing Institute of Geography and Limnology et al., 1989), and (2) whether the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements (including REEs) of the sediments vary systematically with grain size. The two samples retrieved from water depths of 3.5 and 2 m, respectively, and close to the margin (HGY-9 and HGY-10), are very coarse and contain a large amount of sand, while the two samples farthest from the margin along each transect, HGY-6 and HGY-7, are much finer (see the section "Grain size"). Plant remains are present in HGY-7 and in the soil sample (HGY-11).

Grain size analyses were performed following international standard procedure (ISO 13320-1: 1999, Particle size analysis — Laser diffraction methods — Part 1: General principles, MOD). Minerals of the bulk sediments as well as local pyroclastic rock were analyzed at Guangzhou Institute of Geochemistry using a Bruker D8 Advance X-ray diffractometer (XRD) after the samples were ground to powders smaller than 200 mesh. Geochemical analyses (including the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements) on the sediments, pyroclastic rock and soil were carried out at the Earth Dynamic System Research Center (EDSRC), National Cheng-Kung University using the following procedure after apparent plant remains were removed. All these samples were dried at room temperature before they were ground to powders

Table 3
The $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios of the sediments, soil and pyroclastic rock at Lake Huguang Maar in coastal South China and comparison with those of wind-blown dust deposited in the Loess Plateau in North China.

Sample name	$^{87}Sr/^{86}Sr$ (± error, 2σ)	$^{143}Nd/^{144}Nd$ (± error, 2σ)	Sr (ppm)	Nd (ppm)	εNd	Note
HGY-4	0.707157 ± 13	0.512590 ± 9	77	17.8	−0.9	Lake sediments
HGY-6	0.706880 ± 9	0.512713 ± 4	66	26.8	1.5	
HGY-7	0.707241 ± 12	0.512698 ± 4	70	23.5	1.2	
HGY-8	0.707834 ± 13	0.512703 ± 5	60	24.8	1.3	
HGY-9	0.706425 ± 14	0.512642 ± 7	59	12.3	0.1	
HGY-10	0.705964 ± 12	0.512630 ± 5	117	15.5	−0.1	
average	0.7069	0.51266	75	20.1	0.5	
HGY-11	0.704909 ± 12	0.512712 ± 6	189	18.9	1.5	Soil
SSB	0.704120 ± 11	0.512822 ± 4	500	16.8	3.6	Pyroclastic rock
SSM	0.704441 ± 12	0.512741 ± 4	441	17.1	2.0	
SST	0.704364 ± 13	0.512746 ± 5	413	16.4	2.1	
Average	0.7043	0.51277	451	16.7	2.6	
Luochuan[a]	0.7163	0.51214	162	29.1	−9.7	Aeolian sediments in the Loess Plateau, average for each site
Xining[b]	0.7146	0.51212	273	25.7	−10.1	
Xifeng[b]	0.7157	0.51213	180	27.5	−9.9	
Jixian[b]	0.7161	0.51209	176	26.6	−10.6	
Jingchuan[c]	0.7246	0.51208	103	21.7	−10.9	

[a] Gallet et al. (1996).
[b] Jahn et al. (2001).
[c] Sun (2005).

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smaller than 200 mesh. The ground samples were dried again in an oven overnight at ~90°C. Then ca. 25 mg of each sample was used for $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratio and trace-element analyses. The powders were dissolved with concentrated HNO_3 and HF (with a mixing ratio of ca. 1:2) on hot plate (Xu and Marcantonio, 2004) after organic materials was removed with H_2O_2. The majority of the solution was used for Sr and Nd isotopic analyses which were conducted on a Finnigan Triton thermal ionization mass spectrometer after Sr and Nd were separated following standard cation exchange column procedures. Reference materials SRM 987 and La Jolla Nd standard were used for quality control, respectively. SRM 987 run during the same period yielded a mean $^{87}Sr/^{86}Sr$ ratio of 0.710217 ± 14 (2σ) while the La Jolla Nd standard run during the same period yielded a mean $^{143}Nd/^{144}Nd$ ratio of 0.510837 ± 6 (2σ). ε_{Nd} was calculated relative to $^{143}Nd/^{144}Nd$ CHUR = 0.512638. A small aliquot of the dissolved powder was used for trace-element analysis. Trace elements were measured with an ICP-MS with a dilution factor of ca. 5000. BIR-2, BCR-2, BHVO-2, AGV-1, GSP-2 and G2 were used for external calibration. All concentrations are reported relative to sample mass before organic materials were removed.

For comparison, four loess samples collected at Lantian in the south of the Loess Plateau were measured for grain size and two of them were analyzed for minerals. These samples were analyzed along with those collected at Lake Huguang Maar using the same approaches. The bulk sediments rather than the lithogenic materials in the lake sediments were used for geochemical analysis because of the following reasons: (1) to make it clear whether the sediments in Lake Huguang Maar are derived mainly from local pyroclastic rock as was suggested by Zhou et al. (2007); (2) chemical weathering in coastal South China is intensive because of high temperature and heavy precipitation (Woo et al., 1997) and almost all the Sr and much of the Nd are mobilized from parent rock under strong chemical weathering (Ma et al., 2007), and (3) particularly mobilization of different elements or isotopes may differ significantly owing to various activation energy of different minerals (Li et al., 2007). Strong chemical weathering probably change the chemical composition of local pyroclastic rock. Therefore, it is possible that the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements (including REEs) of the lithogenic materials in the lake sediments may not be identical to those of local pyroclastic rock even if the sediments in Lake Huguang Maar were provided by local pyroclastic rock.

Results

Grain size

The grain size distributions of the sediment samples collected from Lake Huguang Maar are listed in Table 1 and are compared with that of typical wind-blown dust. It's clear that wind-blown dust deposited in North China has a relatively uniform grain size distribution, i.e. usually <3% sand, 78–83% silt, and 15–21% of clay. The sediments recovered from Lake Huguang Maar, however, vary significantly in grain size. For example, HGY-9 contains as much as 54% sand, while HGY-6 contains only 1.6% sand. In general, samples collected close to margin (such as HGY-9 and HGY-10) are coarser and contain more sand relative to those obtained far away from margin (such as HGY-6 and HGY-7). This is consistent with the suggestion that the sediments in Lake Huguang Maar might have been input through runoff (Zhou et al., 2007). HGY-6 and HGY-7 have a grain size distribution comparable with that of wind-blown dust (Table 1).

Mineralogy

The results of XRD analysis are shown in Figure 2 and Table 2. Minerals contained in the pyroclastic rock include quartz, anorthite, pyroxene and some amorphous phases while the lake sediments are dominated by quartz and halloysite and some albite is presented as well. Quartz and halloysite in the lake sediments are reversely correlated with each other (Table 2). Generally, coarser samples have higher quartz contents and lower halloysite contents (Tables 1 and 2). Wind-blown dust deposited in North China consists of quartz, K-feldspars, albite, montmorillonite, illite, chlorite, calcite and hornblende.

$^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios

The Sr and Nd isotopic ratios are listed in Table 3. The $^{87}Sr/^{86}Sr$ ratios for local pyroclastic rock range from 0.70436 to 0.70444, slightly lower than the ratio of 0.70491 for the soil sample HGY-11. All these ratios, however, are apparently lower than those for the lake sediments which range from 0.70596 to 0.70783 (Table 3). The $^{143}Nd/^{144}Nd$ ratios for local pyroclastic rock fall between 0.51274 and 0.51282. Unlike the $^{87}Sr/^{86}Sr$ ratios, they are a little higher than the value of 0.51271 for the soil sample. They are also higher than the $^{143}Nd/^{144}Nd$ ratios for the lake sediments which range from 0.51263 to 0.51271 (Table 3).

The $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios for local pyroclastic rock are in line with the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios for Cenozoic basalts in East China (Liu et al., 1995) (Fig. 3). Although the two coarse sediment samples (HGY-9 and HGY-10) have relatively low $^{87}Sr/^{86}Sr$ ratios (Table 1 and 3), both $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios of the lake sediments do not show a systematic variation with grain size (Fig. 4a, b).

Trace elements

Concentrations of trace elements of the lake sediments, local pyroclastic rock and soil are listed in Table 4. For comparison, average concentrations of these elements in wind-blown dust deposited in North China, which were reported by Gallet et al. (1996), are also listed (Table 4).

Nb and Ta, the two most refractory and immobile elements under supergene environment (Kurtz et al., 2000), have relatively uniform contents in the pyroclastic rock (e.g., Nb ranges from 22.4 to 25.0 ppm

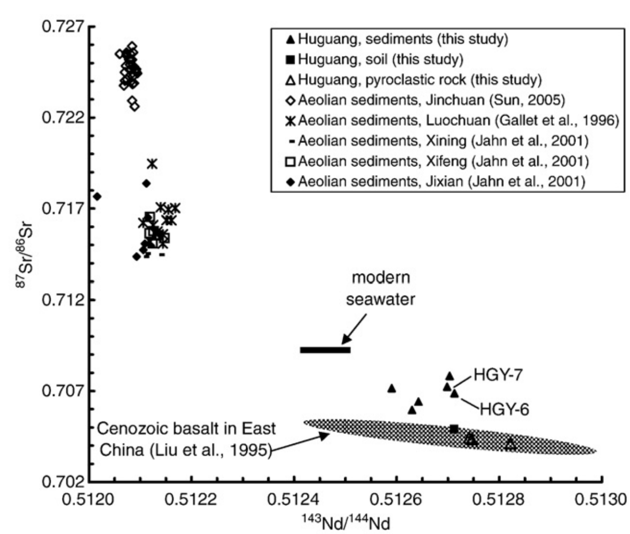

Figure 3. Plot of $^{87}Sr/^{86}Sr$ vs. $^{143}Nd/^{144}Nd$ ratios of the sediments, soil and pyroclastic rock at Lake Huguang Maar in coastal South China and wind-blown dust in the Loess Plateau in North China (Gallet et al., 1996; Jahn et al., 2001; Sun, 2005). Also shown include the Cenozoic basalt in East China (the grey ellipse) (Liu et al., 1995) and modern seawater (the horizontal bar). For modern seawater, its $^{87}Sr/^{86}Sr$ ratio refers to Burke et al. (1982) while the $^{143}Nd/^{144}Nd$ ratio to Ling et al. (1997). Note that the points for Jingchuan represent only the fine fraction (<20 μm) of aeolian sediments (Sun, 2005) and deviate significantly from the rest wind-blown dust samples from the Loess Plateau (Gallet et al., 1996; Jahn et al., 2001). The sediments of Lake Huguang Maar locate between the aeolian dust from arid North China and the pyroclastic rock surrounding the lake, but much closer to the latter. HGY-6 and HGY-7 indicate the two sediment samples having a grain size distribution comparable with that of wind-blown dust form North China.

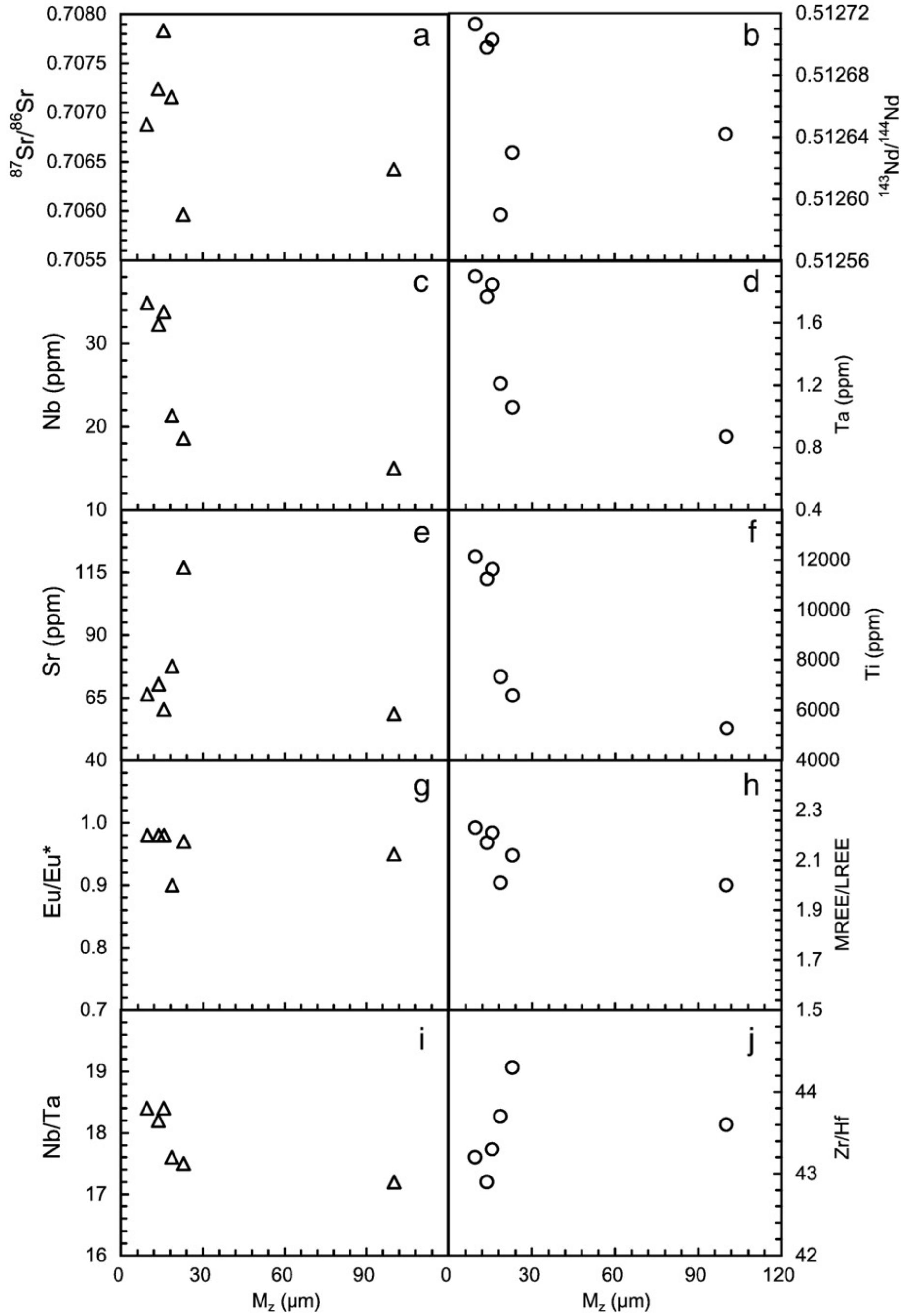

Figure 4. Plots of some geochemical indices vs. mean grain size (M_z) of the sediments in Lake Huguang Maar. (a) $^{87}Sr/^{86}Sr$-M_z; (b) $^{143}Nd/^{144}Nd$-M_z; (c) Nb-M_z; (d) Ta-M_z; (e) Sr-M_z; (f) Ti-M_z; (g) Eu/Eu*-M_z; (h) MREE/HREE-M_z; (i) Nb/Ta-M_z; (j) Zr/Hf-M_z.

and Ta from 1.25 to 1.33 ppm) but vary significantly in the lake sediments (Nb from 15.0 to 34.9 ppm and Ta from 0.87 to 1.90 ppm). These contents show a weak negative correlation with grain size of the lake sediments (Figs. 4c, d). Sr content of the pyroclastic rock ranges from 413 to 500 ppm, much higher than those of the lake sediments which are usually below 80 ppm (Table 4) and are not correlated with grain size (Fig. 4e). In the cores studied by Yancheva et al. (2007a), a large amount of calcareous gyttja is found at depth greater than ~4.5 m (corresponding to ~3.8 ka) (Mingram et al., 2004) and may act as a sink for Sr input into the lake.

In recent years, it was found that Lake Huguang Maar lost its water through leakage (http://news.sina.com.cn/c/2007-03-19/1444114-45918s.shtml). This may explain the low Sr content of the lake sediments (Table 4). Ti in the lake sediments, used by Yancheva et al. (2007a,b) as a proxy for the Asian winter monsoon, ranges from 5300 to 11200 ppm and, like Nb and Ta, displays a weak negative correlation with grain size (Fig. 4f). On average, contents of REEs in the lake sediments are lower than in wind-blown dust from North China except Eu which is a little higher in the lake sediments than in wind-blown dust (Table 4).

Most of the trace elements in the lake sediments display strong correlations with quartz and halloysite with higher trace elements corresponding to less quartz and more halloysite (Fig. 5).

Discussion

Kennedy et al. (1998) suggested that, in Hawii, soluble Sr in local rainfall was dominated by sea-salt aerosols. Huguang Maar is close to the coastline (Fig. 1b). Therefore, it is reasonable to expect a

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Table 4
Concentration (ppm) of trace elements in the pyroclastic rock, soil and lake sediments collected at Lake Huguang Maar and comparison with wind-blown dust deposited in North China.

	Pyroclastic rock			Soil	Lake sediments						Pyroclastic rock, ave.	Lake sediments, ave.	Loess, ave.	Paleosol, ave.
	SSB	SSM	SST	HGY-11	HGY-10	HGY-9	HGY-8	HGY-7	HGY-6	HGY-4				
Cs	0.65	1.11	1.04	1.05	1.04	0.8	1.45	1.6	1.73	1.21	0.93	1.30	7.36	8.01
Rb	27.5	35.1	33.3	20.5	17.1	13.3	19.4	21.2	21	20.2	31.9	18.7	97	85
Ba	181	259	241	269	220	159	220	286	293	227	227	234	456	458
Th	3.12	3.79	3.56	4.41	3.90	3.30	6.58	5.85	6.37	5.31	3.50	5.20	12.2	11.9
U	0.66	0.79	0.76	0.88	0.79	0.64	1.10	1.12	1.15	1.06	0.74	0.97	2.76	2.24
Nb	25.0	23.4	22.4	24.7	18.6	15.0	33.8	32.3	34.9	21.3	23.6	26.0	10.9	11.9
Ta	1.33	1.31	1.25	1.32	1.06	0.87	1.84	1.77	1.90	1.21	1.3	1.44	0.86	0.94
La	17.7	18.5	17.5	20.4	16.5	13.2	26.0	26.4	30.1	19.4	17.9	21.9	32.3	35.5
Ce	34.1	35.6	33.7	40.9	32.9	26.2	49.4	48.0	55.6	38.9	34.4	41.8	64.6	57.6
Pb	12.1	15.6	15.0	10.5	21.0	6.6	20.3	26.5	19.1	19.6	14.2	18.8	21	21
Pr	3.99	4.14	3.94	4.59	3.77	3.02	5.99	5.65	6.47	4.40	4.02	4.89	8.16	9.05
Sr	500	441	413	189	117	59	60	70	66	77	451	75	197	137
Nd	16.8	17.1	16.4	18.9	15.5	12.3	24.8	23.5	26.8	17.8	16.7	20.1	28.1	32
Zr	119	111	107	119	93	78	159	158	167	110	113	128	189	177
Hf	2.66	2.50	2.39	2.67	2.10	1.79	3.67	3.68	3.87	2.52	2.52	2.94	5.00	4.79
Sm	3.96	3.97	3.80	4.43	3.63	2.82	5.80	5.49	6.20	4.07	3.91	4.67	5.70	6.62
Eu	1.30	1.25	1.19	1.39	1.11	0.85	1.80	1.69	1.92	1.16	1.24	1.42	1.12	1.35
Ti	8901	8161	7767	8710	6591	5277	11630	11245	12135	7335	8276	9035	4359	4864
Gd	3.69	3.71	3.54	4.18	3.35	2.63	5.32	5.04	5.74	3.77	3.65	4.31	5.11	6.18
Tb	0.60	0.61	0.59	0.69	0.55	0.43	0.89	0.85	0.96	0.63	0.60	0.72	0.79	0.95
Dy	3.37	3.36	3.24	3.86	3.08	2.42	4.86	4.66	5.25	3.48	3.32	3.96	4.57	5.53
Y	17.6	18.1	17.3	21.1	16.7	13.1	26.2	24.4	28.2	19.0	17.7	21.2	29.17	34.23
Ho	0.63	0.64	0.61	0.74	0.58	0.47	0.93	0.88	1.00	0.68	0.63	0.76	0.93	1.12
Er	1.61	1.65	1.58	1.93	1.53	1.22	2.38	2.29	2.57	1.79	1.61	1.96	2.61	3.09
Tm	0.24	0.25	0.24	0.29	0.23	0.20	0.35	0.34	0.37	0.27	0.24	0.29	0.43	0.50
Yb	1.34	1.39	1.34	1.62	1.31	1.09	1.99	1.92	2.12	1.55	1.36	1.66	2.70	3.09
Lu	0.19	0.19	0.19	0.23	0.18	0.15	0.28	0.27	0.30	0.22	0.19	0.23	0.41	0.46
Nb/Ta	18.8	17.9	17.9	18.7	17.5	17.2	18.4	18.2	18.4	17.6	18.2	18.1	12.7	12.7
Zr/Hf	44.7	44.4	44.8	44.6	44.3	43.6	43.3	42.9	43.2	43.7	44.8	43.5	37.8	37.0
Ti/Nb	356	348	347	353	354	351	344	348	348	344	351	348	400	409
Ti/Nd	530	477	474	461	425	429	469	479	453	412	496	450	155	152
Eu/Eu*	1.03	0.99	0.99	0.98	0.97	0.95	0.98	0.98	0.98	0.90	1.00	0.96	0.63	0.64
LREE/MREE	3.25	3.53	3.51	3.50	3.55	3.71	3.45	3.73	3.74	3.99	3.44	3.68	6.88	6.27
MREE/HREE	2.27	2.20	2.19	2.14	2.12	2.00	2.21	2.17	2.23	2.01	2.22	2.14	1.56	1.64

Eu/Eu* = $Eu_n/\mathrm{sqrt}(Sm_n{*}Gd_n)$; LREE/MREE = La_n/Eu_n; MREE/HREE = Gd_n/Yb_n: where n indicates normalized to chondrite (Anders and Grevesse, 1989).

significant amount of Sr transported from nearby South China Sea to Huguang Maar and incorporated into authigenic materials in the lake (such as secondary minerals).

If the sediments in Lake Huguang Maar derived mainly from wind-blown dust from North China, with a Sr content between 100 and 300 ppm (Gallet et al., 1996; Jahn et al., 2001), Sr in the lake sediments would have two possible sources, wind-blown dust and nearby seawater. Either of the two sources, or their combination, however, cannot explain the $^{87}Sr/^{86}Sr$ ratios of the sediments in Lake Huguang Maar. The mean $^{87}Sr/^{86}Sr$ ratio of modern seawater is about 0.7092 (Burke et al., 1982; Hodell, 1994) and aeolian sediments deposited at Luochuan, Xining, Xifeng and Jixian in the Loess Plateau in North China fall between 0.714 and 0.719 (Gallet et al., 1996; Jahn et al., 2001). Sun (2005) analyzed the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ ratios of the fine fraction (<20 μm) of the aeolian sediments deposited at Jingchuan in the Loess Plateau after carbonate was removed and found that the $^{87}Sr/^{86}Sr$ ratios since the Pleistocene ranged from 0.7226 to 0.7259. The $^{87}Sr/^{86}Sr$ ratios of both modern seawater from nearby South China Sea and wind-blown dust from remote arid North China are distinctly higher than those of the sediments in Lake Huguang Maar, which fall between 0.7060 and 0.7078 (Table 3). Therefore, another Sr source which is much less radiogenic than modern seawater and wind-blown dust must have contributed a significant amount of Sr to Lake Huguang Maar.

The pyroclastic rock surrounding Lake Huguang Maar has a high Sr content (410–500 ppm, Table 4) and low $^{87}Sr/^{86}Sr$ ratios (0.7041–0.7045; Table 3). It is the most appropriate candidate for a third Sr source, especially considering the high temperature and heavy precipitation in coastal South China (Woo et al., 1997) and the steep slope surrounding the lake (Mingram et al., 2004) which should promote weathering of the rocks and transport of water and sediment to the lake. This possibility is supported by the $^{143}Nd/^{144}Nd$ ratios and trace elements of the sediments in Lake Huguang Maar.

The $^{143}Nd/^{144}Nd$ ratios of the sediments in Lake Huguang Maar are close to those of local pyroclastic rock, but much higher than the published $^{143}Nd/^{144}Nd$ ratios of wind-blown dust from North China (Fig. 3; Table 3; Gallet et al., 1996; Jahn et al., 2001; Sun, 2005). Unlike Sr, sources of Nd in the lake sediments should not be complicated by a seawater end-member because Nd concentration in seawater is low (Alibo and Nozaki, 1999) and input of Nd from seawater should be negligible. It is evident that wind-blown dust from North China alone cannot explain the $^{143}Nd/^{144}Nd$ ratios of the sediments in Lake Huguang Maar (Fig. 3; Table 3). A large proportion of the Nd in the lake should come from local pyroclastic rock.

Nb, Ta, Zr, Hf and Ti are refractory elements in basalt (Nesbitt and Wilson, 1992). Although Nb and Ta vary significantly in content in the lake sediments (Table 4), which may be due to the dilution effect of some authigenic materials, plotting Nb vs. Ta indicates that all the samples from Huguang Maar distribute almost exactly along a straight line (Fig. 6a). Wind-blown dust deposited in the Loess Plateau in North China also distributes roughly along a line on this plot (Gallet et al., 1996; Jahn et al., 2001), but deviates significantly from the trend line of the samples from Lake Huguang Maar (Fig. 6a). Plotting Zr and Hf gives a similar picture (Fig. 6b). These variation diagrams suggest a closer association of the sediments in Lake Huguang Maar with local pyroclastic rock than with wind-blown dust from North China.

Like $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios, the rare earth elements (REEs), a group of elements which are similar in atomic structure and chemical properties, are often used for identifying the provenance of sediments (Cullers et al., 1988; McLennan, 1989; Zhou

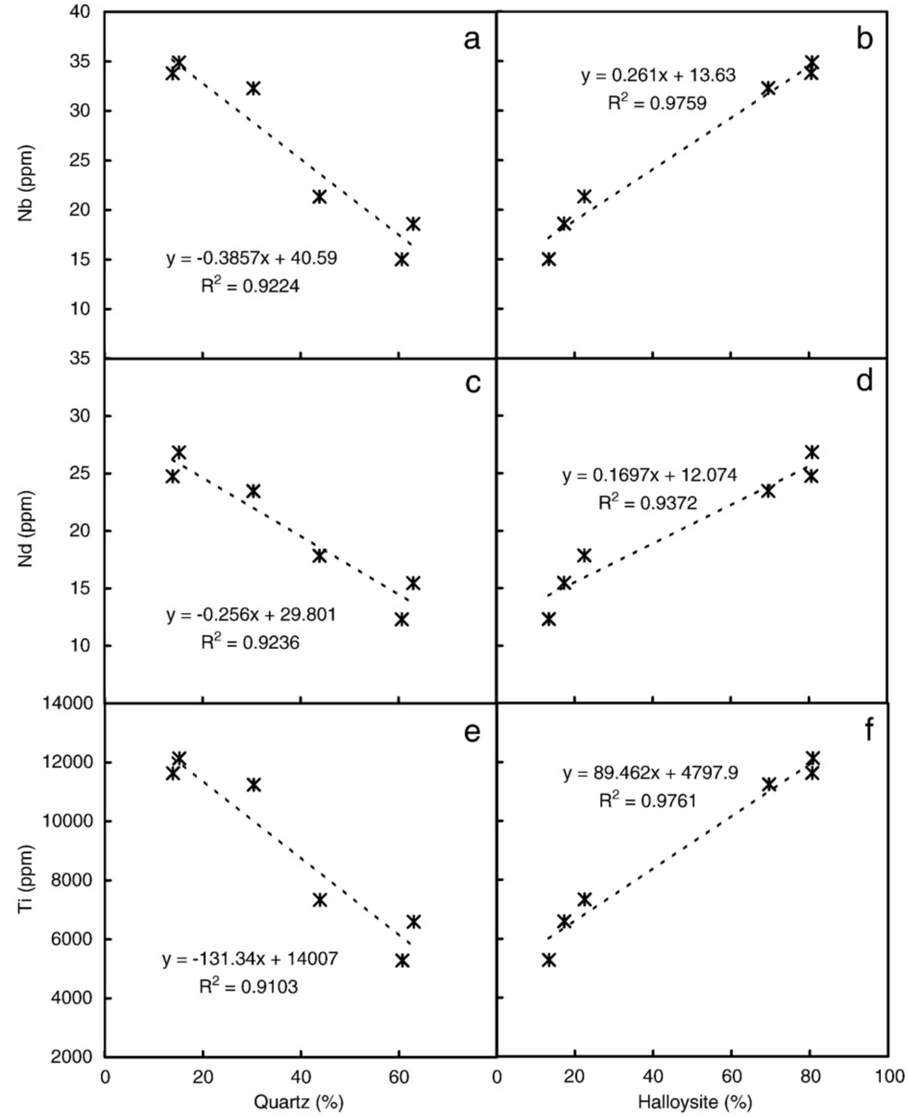

Figure 5. Correlations of Nb, Nd and Ti with quartz and halloysite in the sediments of Lake Huguang Maar. (a) Nb-quartz; (b) Nb-halloysite; (c) Nd-quartz; (d) Nd-halloysite; (e) Ti-quartz; (f) Ti-halloysite.

et al., 2008b). The REEs of the sediments in Lake Huguang Maar, local pyroclastic rock and soil were normalized to a chondrite standard (Fig. 7a) as was usually done for wind-blown dust deposited in the Loess Plateau (Gallet et al., 1996). The REE contents of chondrite published by Anders and Grevesse (1989) were used for this normalization. For comparison, the chondrite-normalized REE pat-

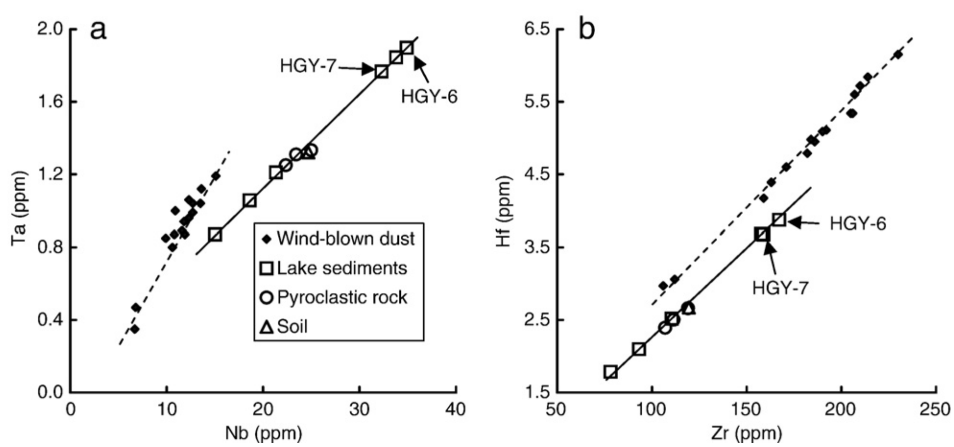

Figure 6. Plots of Nb vs. Ta (a) and Zr vs. Hf (b) for the lake sediments, pyroclastic rock and soil collected at Lake Huguang Maar and wind-blown dust from North China. HGY-6 and HGY-7, the two samples having a grain size distribution comparable with that of wind-blown dust form North China, locate on the trend line defined by the samples collected at Lake Huguang Maar, not on the line defined by wind-blown dust from North China.

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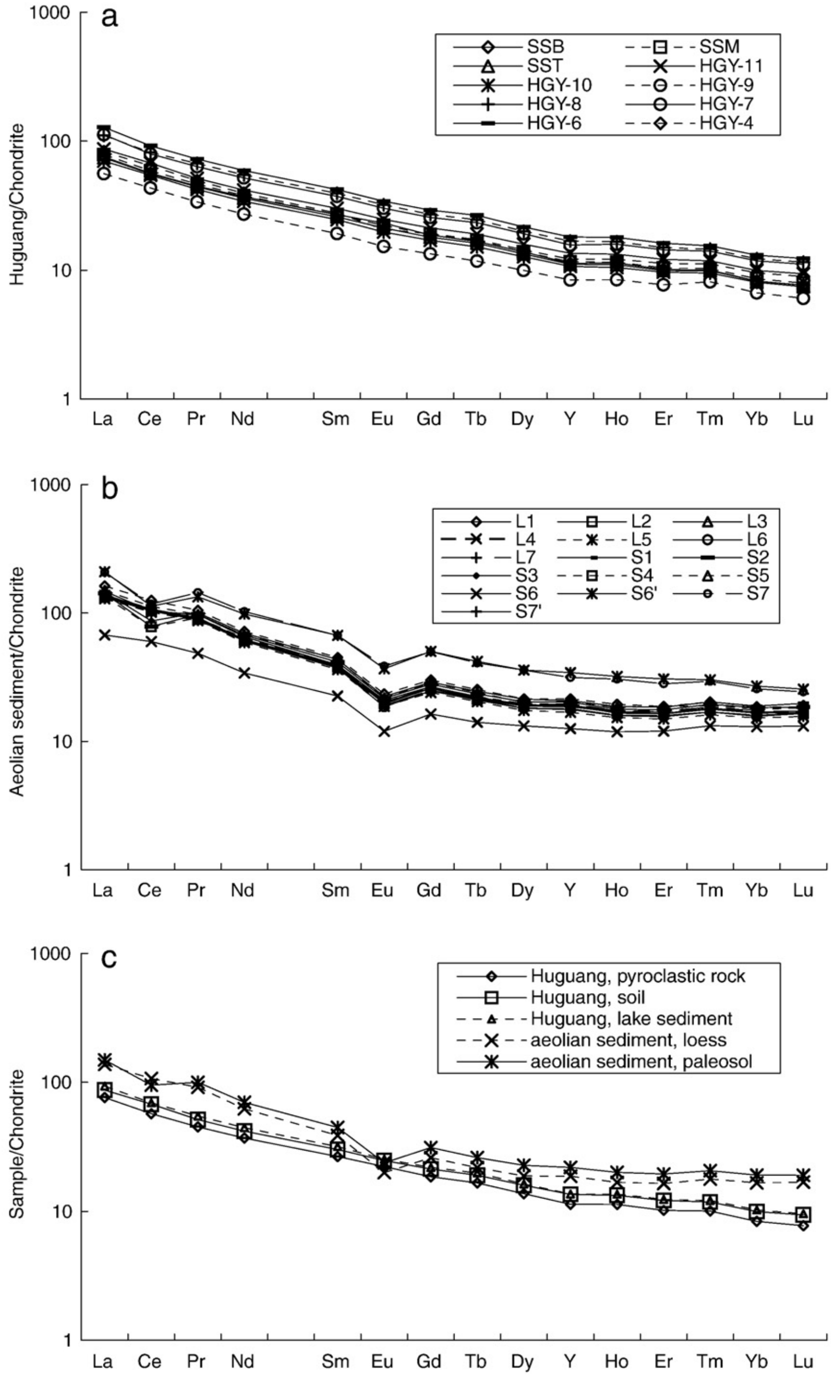

Figure 7. The chondrite-normalized REY patterns for (a) the pyroclastic rock, soil and sediments at Lake Huguang Maar in coastal South China, (b) the aeolian sediments in the Loess Plateau in North China (L: loess; S: paleosol) (Gallet et al., 1996), and (c) a comparison of their averages.

terns for wind-blown dust deposited in the Loess Plateau (Gallet et al., 1996) are also illustrated (Fig. 7b). It is clear that the REE patterns for the lake sediments are parallel with those for local pyroclastic rock and soil, but are differ from those for wind-blown dust deposited in the Loess Plateau. For example, the notable negative europium anomaly for wind-blown dust is not observed in the REE patterns for the lake sediments, pyroclastic rock and soil. In addition, wind-blown dust appears to display steeper light REE and flatter heavy REE relative to the samples collected at Lake Huguang Maar (Fig. 7c).

In summary, all the geochemical indices including $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios and trace elements (including REEs) suggest that the sediments in Lake Huguang Maar are more closely related to local pyroclastic rock than wind-blown dust from North China. This is supported by minerals of the lake sediments which contain a large amount of halloysite (Fig. 2 and Table 2) probably derived from strong chemical weathering of anorthite and pyroxene in local pyroclastic rock under high temperature and heavy precipitation, whereas the mineral assemblages, especially the clay minerals of the lake sediments are differed significantly from the wind-blown dust from North China (Fig. 2 and Table 2).

It may be argued that although the bulk sediments in Lake Huguang Maar display a geochemistry much different from that of wind-blown dust from North China, the silt-size fraction in the lake sediments may still be dominated by wind-blown dust. However, this is inconsistent with the following evidence. (1) Although the sediment samples vary significantly in grain size (Table 1), their

$^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios, REE patterns and ratios of refractory elements are not correlated with grain size (Fig. 4a, b, g–j). (2) The $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios, REE patterns and ratios of refractory elements for all the sediment samples, including HGY-6 and HGY-7 which have a grain size distribution comparable with wind-blown dust (Table 1), are close to those for local pyroclastic rock and remarkably differ from those of wind-blown dust from North China (Fig. 3, 6, 7; Table 3, 4).

In the following sections, we will try to quantify the contribution of wind-blown dust from North China to the sediments in Lake Huguang Maar using $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios. As discussed above, $^{87}Sr/^{86}Sr$ ratios of the sediments in Lake Huguang Maar may be controlled by three end members, local pyroclastic rock, wind-blown dust from North China, and nearby seawater. The relative contribution of Sr from wind-blown dust is estimated by the following equation:

$$\left(^{87}Sr/^{86}Sr\right)_{ls} = \left(^{87}Sr/^{86}Sr\right)_{sw} {}^*C_{sw} + \left(^{87}Sr/^{86}Sr\right)_{wd} {}^*C_{wd} + \left(^{87}Sr/^{86}Sr\right)_{pr} {}^*C_{pr} \quad (1)$$

where $(^{87}Sr/^{86}Sr)_x$ and C_x represent the Sr isotopic ratio and relative contribution (percentage) from source X, ls denotes lake sediments, pr denotes pyroclastic rock, wd denotes wind-blown dust and sw denotes seawater. Note that:

$$C_{sw} + C_{wd} + C_{pr} = 1 \quad (2)$$

Based on Eqs. (1) and (2), the following equation can be derived,

$$C_{sw} = (C_{wd}{}^*\left(\left(^{87}Sr/^{86}Sr\right)_{wd} - \left(^{87}Sr/^{86}Sr\right)_{pr}\right) - \left(\left(^{87}Sr/^{86}Sr\right)_{ls} - \left(^{87}Sr/^{86}Sr\right)_{pr}\right)) / \left(\left(^{87}Sr/^{86}Sr\right)_{pr} - \left(^{87}Sr/^{86}Sr\right)_{sw}\right). \quad (3)$$

Because $C_{sw} \geq 0$ and $(^{87}Sr/^{86}Sr)_{pr} - (^{87}Sr/^{86}Sr)_{sw} < 0$ (Table 1; Burke et al., 1982), thus,

$$C_{wd} \leq \left(\left(^{87}Sr/^{86}Sr\right)_{ls} - \left(^{87}Sr/^{86}Sr\right)_{pr}\right) / \left(\left(^{87}Sr/^{86}Sr\right)_{wd} - \left(^{87}Sr/^{86}Sr\right)_{pr}\right) \quad (4)$$

Using the average $^{87}Sr/^{86}Sr$ ratios of the sediments in Lake Huguang Maar, pyroclastic rock (Table 1) and wind-blown dust from North China (Table 3; Gallet et al., 1996; Jahn et al., 2001), and in Eq. (4), C_{wd} is calculated to be less than 22%. It reaches the highest value of ~22% when $C_{sw} = 0$. This means that if there was no Sr input from nearby seawater, Sr contribution from wind-blown dust would account for only about a quarter of that from local pyroclastic rock. Both C_{wd} and the ratio of C_{wd}/C_{pr} would decrease with an increase in C_{sw}. In fact, as we suggested previously, Sr input from nearby seawater should be important for Lake Huguang Maar. C_{wd} should be significantly less than 22% and the C_{wd}/C_{pr} ratio less than 1/4. Therefore, although Sr concentrations in precipitation at Lake Huguang Maar and in nearby seawater are not clear at present, local pyroclastic rock seems to be more important than wind-blown dust from North China for Sr deposited in the lake.

Sr is one of the most mobile elements under strong chemical weathering and in particular, as there is Sr loss through leakage, $^{87}Sr/^{86}Sr$ ratio may not be able to trace the sediments, especially the lithogenic materials in Lake Huguang Maar. The $^{143}Nd/^{144}Nd$ ratios of the lake sediments could be used to double check the contribution of wind-blown dust to the sediments in Lake Huguang Maar.

The Nd in Lake Huguang Maar has two possible sources, wind-blown dust and local pyroclastic rock. The $^{143}Nd/^{144}Nd$ ratios of the lake sediments suggest that only ~17% of the Nd comes from wind-blown dust from North China, which is in accordance with the $^{87}Sr/^{86}Sr$ ratios of the lake sediments. This agrees also with the flux of the lithogenic materials to Lake Huguang Maar (Yancheva et al., 2007a) and model-derived dust deposition rates in coastal South China (Ginoux et al., 2001; Mahowald et al., 1999). Modern dust deposition rates in coastal South China derived from model simulations range from <0.2 to 0.5 mg cm^{-2} yr^{-1} (Ginoux et al., 2001; Mahowald et al., 1999) and the flux of the lithogenic materials estimated by Yancheva et al. (2007a) is 5–10 mg cm^{-2} yr^{-1} in Holocene. Considering that sediment focusing in Maar lake may be measured up to fivefold (Schettler et al., 2006), a ratio of ~17% is reasonable for dust-derived sediments in Lake Huguang Maar.

According to the average Sr and Nd contents of local pyroclastic rock and wind-blown dust from North China (Table 4), a mixture consisting of 17% dust and 83% pyroclastic rock would have a Sr content of 408 ppm and a Nd content of 18.7 ppm. The Sr contents of the lake sediments are much less than 408 ppm (Table 4), which may be due to water leakage in the lake as indicated in recent years. The average Nd content of the lake sediments, 20.1 ppm (Table 4) is consistent with 18.7 ppm. However, this is coincidental because the lake sediments derived from significantly altered pyroclastic rock and may have a large amount of authigenic materials. 17% of dust and 83% of pyroclastic rock would lead to a $^{87}Sr/^{86}Sr$ ratio of 0.70624 in the mixture, which is lower than the average $^{87}Sr/^{86}Sr$ ratio 0.7069 in the lake sediments (Table 3). Assuming that seawater from nearby South China Sea has a $^{87}Sr/^{86}Sr$ ratio of 0.7092 as reported for modern seawater (Burke et al., 1982; Hodell, 1994), it is estimated that on average, about 23% of the total Sr in the bulk sediments of Lake Huguang Maar is from seawater, meanwhile ~13% and ~64% of the Sr are derived from wind-blown dust and pyroclastic rock, respectively.

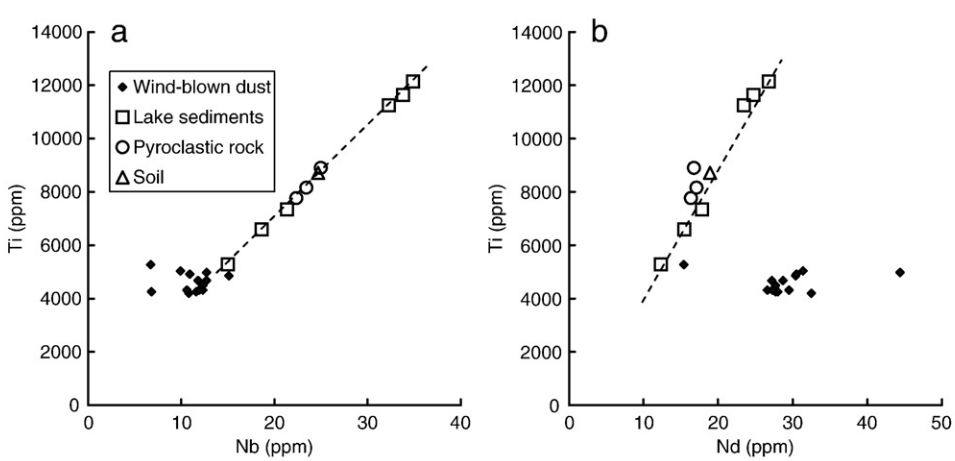

Figure 8. Plots of Ti vs. Nb (a) and Nd (b) for the lake sediments, pyroclastic rock and soil collected at Lake Huguang Maar and wind-blown dust from North China.

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In conclusion, although (1) the lake sediments analyzed are not from the same cores studied by Yancheva et al. (2007a), and (2) wind-blown dust has been identified in soils globally, even in places far away from dust source regions (Simonson, 1995), the sediments in Lake Huguang Maar, a place not on a main dust trajectory pathway (Ginoux et al., 2001), should be provided mainly by local pyroclastic rock. Wind-blown dust from arid North China, which may be transported by the Asian winter monsoon (Yancheva et al., 2007a,b), should have a much lower contribution.

Although the major source of the sediments of Lake Huguang Maar is geochemically identified as local pyroclastic rock, we argue further that Ti in the lake sediments should also come mainly from the pyroclastic rock because of the following lines of evidence: (1) Ti in basalt in coastal South China appears to be more easily removed under strong chemical weathering (Ma et al., 2007) and the ratio of Ti/Nd in local pyroclastic rock is much higher than in wind-blown dust from North China (Gallet et al., 1996; Table 4). Given that local pyroclastic rock provided ~83% Nd in the lake sediments, a great amount of Ti should have been transported from this source into the lake. (2) The Ti/Nd ratio of the lake sediments ranges from 410 to 480, overlapping with that of pyroclastic rock (475–531) but remarkably higher than that of loess sediments deposited in the Loess Plateau in North China (ranging from 129 to 172 and averaging 156, calculated using data from Gallet et al., 1996 and Jahn et al., 2001; Table 4). (3) Plots of Ti vs. Nb or Nd indicate that all the samples collected at Lake Huguang Maar distribute along a straight line and deviate significantly from wind-blown dust from North China (Fig. 8), suggesting that Ti, Nb and Nd in the lake sediments originate mainly from the same source of local pyroclastic rock. (4) Ti in the lake sediments display a strong positive correlation with halloysite (Fig. 5f), suggesting that Ti may be associated with halloysite derived from anorthite and pyroxene in local pyroclastic rock.

Therefore, the sediments and Ti in Lake Huguang Maar are dominated by local pyroclastic rock and are controlled by the hydrology of the lake (Zhou et al., 2007). However, wind-blown dust transported by the Asian winter monsoon from arid North China may make a minor contribution to the sediments in Lake Huguang Maar. Although it seems unlikely that variations of Ti in the lake sediments (Yancheva et al., 2007a) were caused largely by changes in the Asian winter monsoon, it is possible that wind-blown dust has contributed to the long-term Ti variations observed in Lake Huguang Maar (Yancheva et al., 2007a). Quantification of this contribution, however, needs more studies on the cores recovered by Yancheva et al. (2007a). Furthermore, how to correlate other proxies adopted by Yancheva et al. (2007a,b), especially the redox-sensitive proxies such as S-ratio and total organic carbon with past climate and environment is not straightforward and warrants further investigation. Monitoring of the lake system may be an appropriate way to solve these issues in the future.

Conclusions

$^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic ratios, REE pattern and ratios of refractory trace elements indicate that the sediments collected from Lake Huguang Maar are geochemically much different from wind-blown dust deposited in the Loess Plateau in North China (Gallet et al., 1996; Jahn et al., 2001; Sun, 2005). Instead, they are closely similar to local pyroclastic rock (Table 3 and 4; Fig. 3, 6–8). $^{87}Sr/^{86}Sr$ ratio indicates that less than 22% of the Sr in the bulk sediments of Lake Huguang Maar comes from wind-blown dust while $^{143}Nd/^{144}Nd$ ratio indicates ~17% Nd contribution from this source. The majority of the sediments and Ti in Lake Huguang Maar were provided by local pyroclastic rock, probably input into the lake through surface runoff. These observations are in line with the sedimentological evidence (Wang et al., 2000; Zhou et al., 2007) and minerals in the lake sediments (Table 2). Wind-blown dust transported by the Asian winter monsoon from arid North China is only a minor source for the sediments in Lake Huguang Maar and may have contributed to the long-term Ti variations observed (Yancheva et al., 2007a). Quantification of this contribution, however, warrants further investigation in the future.

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