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Natural and anthropogenic sources of atmospheric dust at a remote forest area in Guizhou karst region, southwest China

Yang Tang[1], Guilin Han[2,3]*, Fushan Li[1] & Qixin Wu[1,4]

[1] Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China
[2] School of Water Resources and Environment, China University of Geosciences (Beijing), Beijing 100083, China
[3] Present address: School of Water Resources and Environment, China University of Geosciences (Beijing), Beijing 100083, China
[4] Key Laboratory of Karst Environment and Geohazard Prevention, Ministry of Education, Guizhou University, Guiyang 550003, China
* Correspondence: hanguilin@cugb.edu.cn

Abstract: The abundance and distribution patterns of Rare Earth Elements (REE) in atmospheric dust in a typical karst forest area of the Maolan National Nature Reserve Park in Southwest China were determined during the period from May 2009 to January 2011. Total REE concentrations (ΣREE) recorded moderate seasonal variation, ranging from 31.0 mg kg^{-1} to 88.2 mg kg^{-1}, with an average of 53.5 mg kg^{-1}, similar to the ΣREE of local topsoil. The values of ΣREE were negatively correlated with the corresponding dust deposition fluxes, which may be due to different rates of deposition as a function of particulate size. PAAS-normalized REE patterns of the dust showed slightly positive Ce anomalies and enrichment in light REE. Most dust samples had similar PAAS-normalized REE patterns to local topsoil, whereas others were similar to the anthropogenic input material from urban areas.

Keywords: Rare earth element, Atmospheric dust, Karst, Anthropogenic input

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As part of the global biogeochemical cycle, atmospheric dust is crucial to surface ecosystems, because it provides nutrient elements (Kennedy *et al.* 1998; Soderberg & Compton 2007). The transportation distance of atmospheric dust ranges from hundreds of meters to thousands of kilometers (Swap *et al.* 1992; Takemura *et al.* 2002; Grousset *et al.* 2003), and dust from different sources mix with each other during transport (Lawrence & Neff 2009).

Rare Earth Elements (REE) are useful indicators of processes that affect biogeochemical cycles because of their unique chemical and physical characteristics relative to other elements (Taylor & McLennan 1985; Greaves *et al.* 1994; Tyler 2004; Salaün *et al.* 2011). Normalized REE patterns of dust can be changed during weathering or smelting, but not during transportation or deposition processes (De Oliveira & Imbernon 1998; Laveuf *et al.* 2008; Gueguen *et al.* 2012). Therefore, REE patterns have been used for tracing the sources of atmospheric aerosols and particulates in many studies (Yadav & Rajamani 2004; Moreno *et al.* 2006; Wu *et al.* 2009). In urban areas, airborne particulates are most likely of anthropogenic sources, which are typically enriched in light REE, as observed in the airborne particulate matter (APM) of the Netherlands (Wang *et al.* 2000), Wuhan (Lu *et al.* 2007), Mexico City (Moreno *et al.* 2008), and Tokyo (Suzuki *et al.* 2011). However, REE patterns in dust of Rome, Beijing (Tang *et al.* 2013), and Guiyang show that urban soils can preserve the original REE patterns and anthropogenic contributions.

REE patterns also can be used for tracing long-range air transportation. In peat sediments of southeast Florida, the REE patterns suggested a contribution of Sahara Desert material to dust (Kamenov *et al.* 2009). Ice cores from Mt. Everest recorded large seasonal variations of REE concentrations and patterns, showing a mixture of aerosols from western arid regions such as the Thar Desert (Yadav & Rajamani 2004), West Asia, and the Sahara Desert (Zhang *et al.* 2009).

In this study, we used the REE patterns of atmospheric dust samples from a remote forest area of Guizhou karst region to decipher the sources and deposition characteristics of this atmospheric dust, and explore their effects on biogeochemical cycles of karst ecosystem.

Experimental

Study area

The Maolan National Nature Reserve Park (25°09′20″N to 25°20′50″N; 107°52′10″E to 108°05′40″E) is located in the southeastern region of Guizhou Province, Southwest China (Fig. 1). It is famous for its dense, virgin evergreen forests on the peak of karst areas. The park covers an area of 200 km^2, consisting mostly of mountains of jagged carbonate rocks with 90% forest cover. The climate of this region is subtropical, monsoonal, and humid, with a mean annual temperature of 17°C and annual precipitation of 1750 mm (Han *et al.* 2015).

Dust samples were collected at Banzhai village (Fig. 1). This small village is located on a hill in the core area of Maolan park, and far from any town. Local residents are traditional farmers including the Yao and have minimal modern amenities. The prevailing wind direction in this region is northwesterly in winter, and southeasterly in summer (Fig. 1). The sampling strategy was designed to collect representative data from the whole area encompassing different weather conditions.

Sampling methods

Atmospheric dust was collected in a vessel made of polypropylene (Ganor *et al.* 2003), with size of 30 × 50 cm. The vessel was fixed to steel shelves in an isolated house in the village of Banzhai (25°13′884″N, 108°01′031″E), Maolan National Nature Reserve Park. Purified glycol solution (20%) was added to the vessel as a collection medium, as this depresses algal and microbe growth. In

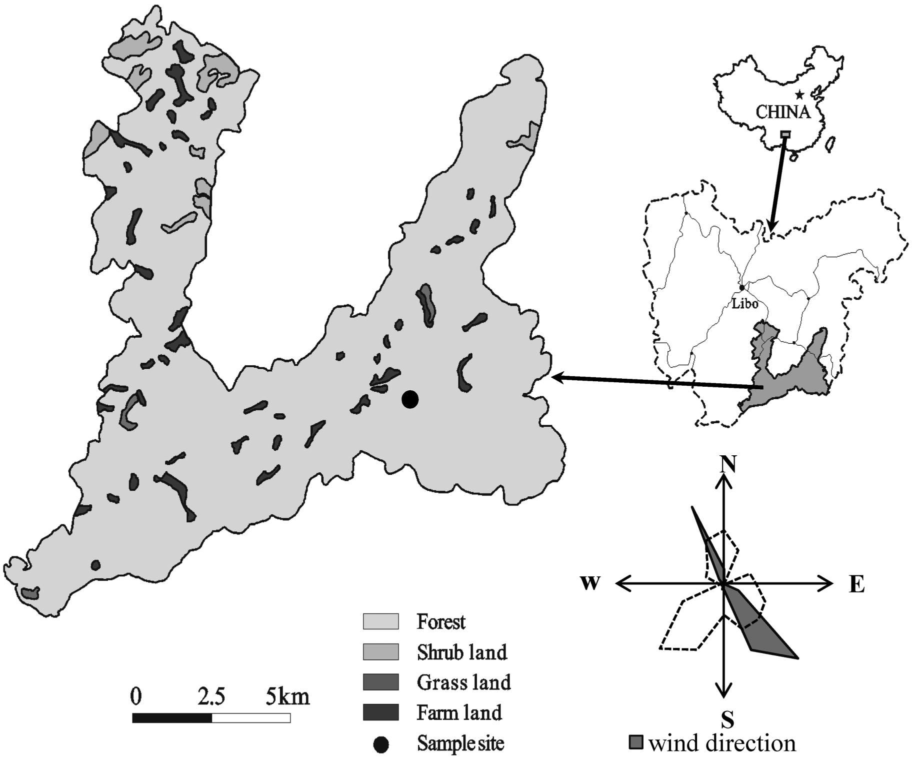

Fig. 1. Geography map and land use of Maolan Nature National Reserve Park showing sample sites.

the meantime, the topsoil near the sampling site was collected and analyzed to compare with the dust.

The collector was covered during rainy periods, and was exposed at all other times. Glycol solution was added as necessary to keep the bottom of the bucket submerged. Dust samples were gathered every two months, from May 2009 to January 2011. The solid and liquid mixture was transferred to a clean glass vessel, and then transported to the lab for processing. Impurities such as leaves and insects were manually removed by forceps. The mixture was dried in an evaporating dish at 80°C.

Analytical methods

Atmospheric dust and topsoil samples were ground to powder in an agate mortar, and dried in an oven at 105°C for 3 h before digesting. The digestion methods are summarized by Yang *et al.* (2007) and Roy & Smykatz-Kloss (2007).

About 100 mg of sample was added to a PFA jar (Savillex, US) and reacted with 1 ml HF and 3 ml HNO_3. The jar was heated on a hot plate at 140°C for 7 days to further break down the silicate, fluoride, and carbon compounds. The sample was then re-dissolved in 1 ml HF and 3 ml HNO_3, using the same treatment in step 1 and repeated until the solution was clear. After the samples are completely digested, 2 ml HNO_3 (1:1) was added twice to break up the fluoride compounds and then dried on a hot plate. The resulting residue was digested using 2% HNO_3 in a 100-ml volumetric flask.

The digestion was performed in the Ultra-Clean Lab of the Institute of Geochemistry, Chinese Academy of Sciences. The REE concentrations of the digested samples were analyzed using ICP-MS (ELEMENT, Finnigan-MAT, USA). Precision was controlled by adding an indium internal standard with calibration as described as Qi *et al.* (2000).

Results and discussion

The REE concentrations of dust are given in Table 1. The post-Achaean Australian average shale (PAAS) data from McLennan (1989) were used to normalize the REE data.

REE concentrations of dust

Total REE concentrations (ΣREE) of the Maolan atmospheric dust samples vary moderately, ranging from 31.0 mg kg^{-1} to 88.2 mg kg^{-1}, with an average of 53.5 mg kg^{-1}. These concentrations are similar to the ΣREE of local topsoil (57.8 mg kg^{-1}), but much less than average ΣREE of atmospheric dust from Guiyang was 93.9 mg kg^{-1}. Seasonal ΣREE variations of Maolan dust such as in December 2009 and February 2010 may reflect the drought event of southern China during the winter 2009 and spring 2010.

The ΣREE concentrations were negatively correlated with dust depositional fluxes (Fig. 2), with dust collected during lower deposition fluxes having higher ΣREE concentrations. In general, lower dust depositional fluxes are associated with dust particulates that are smaller at the same site. Some studies suggest that REE are enriched in fine particulates in geological materials (Castillo *et al.* 2008), which could explain the negative correlation between ΣREE concentrations and dust fluxes.

Normalized REE patterns and dust sources

In modern petroleum refining, light REEs are used as catalysts. Thus, the La/Sm ratio might be a practicable indicator of anthropogenic inputs (Suzuki *et al.* 2011). The La/Sm ratios of Maolan dust was 4.20–7.01, higher than the corresponding top soil at 4.01, but close to that of atmospheric dust from Guiyang (5.42), as well as the course (>11 μm) airborne particulate matter from Tokyo (6.5 ± 2.3, Suzuki *et al.* 2011). The La/Sm ratios of the atmospheric dust in Maolan are similar to those of the typical urban dust, which is consistent with dust from Maolan having a component from industrial sources.

The REE concentrations of atmospheric dust were normalized to PAAS (McLennan 1989) and the local topsoil, respectively. The PAAS-normalized patterns of REE of atmospheric dust from Maolan (Fig. 3) recorded slightly positive Ce anomalies (Ce/Ce* = 0.94 –1.25), but no obvious Eu anomalies except for dust of July 2010 (Eu/Eu* = 1.14). Local topsoil had slightly positive Ce anomalies and negative Eu anomalies (Ce/Ce* = 1.24; Eu/Eu* = 0.90).

Table 1. *Concentration of rare earth elements (mg kg^{-1}) in Maolan (ML) atmospheric dust, topsoil, and post-Archean Australian average shale (PAAS) normalized REE values*

Sample	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	ΣREE	Ce/Ce*	Eu/Eu*
PAAS	38.2	79.6	8.83	33.9	5.55	1.08	4.66	0.77	4.68	0.99	2.85	0.41	2.82	0.43			
ML0905	8.89	17.59	1.86	7.00	1.40	0.31	1.33	0.21	1.24	0.26	0.73	0.10	0.72	0.10	41.8	1.00	1.05
ML0908	6.53	14.68	1.43	5.30	1.02	0.21	1.09	0.16	0.90	0.19	0.53	0.08	0.53	0.07	32.7	1.11	0.94
ML0910	9.06	22.98	1.98	7.46	1.49	0.30	1.42	0.22	1.32	0.29	0.78	0.12	0.80	0.11	48.3	1.25	0.96
ML0912	16.77	33.12	3.29	12.11	2.39	0.50	2.34	0.36	2.03	0.46	1.25	0.18	1.29	0.18	76.3	1.03	0.98
ML1002	19.34	37.46	3.95	14.56	2.89	0.61	2.99	0.44	2.37	0.52	1.34	0.20	1.28	0.19	88.2	0.99	0.98
ML1005	8.55	17.88	2.02	7.67	1.62	0.37	1.66	0.26	1.61	0.35	0.99	0.15	1.05	0.16	44.4	0.99	1.04
ML1007	11.42	21.68	2.50	9.31	2.01	0.48	1.95	0.33	2.17	0.44	1.41	0.23	1.58	0.23	55.7	0.94	1.14
ML1009	12.95	26.73	2.70	9.68	1.93	0.42	2.02	0.30	1.71	0.39	1.03	0.14	0.97	0.14	61.1	1.04	1.01
ML1011	5.74	13.42	1.34	5.10	1.09	0.24	1.08	0.17	1.07	0.24	0.67	0.10	0.68	0.10	31.0	1.12	1.06
ML1101	9.10	22.99	2.50	9.70	2.17	0.50	2.24	0.37	2.16	0.50	1.38	0.20	1.44	0.22	55.5	1.11	1.06
ML-Soil	8.78	24.74	2.39	9.54	2.19	0.43	2.23	0.38	2.50	0.58	1.69	0.26	1.80	0.27	57.8	1.24	0.90
Blank	0.02	0.03	0.01	0.02	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD			

BD, Below Detectable Limit.

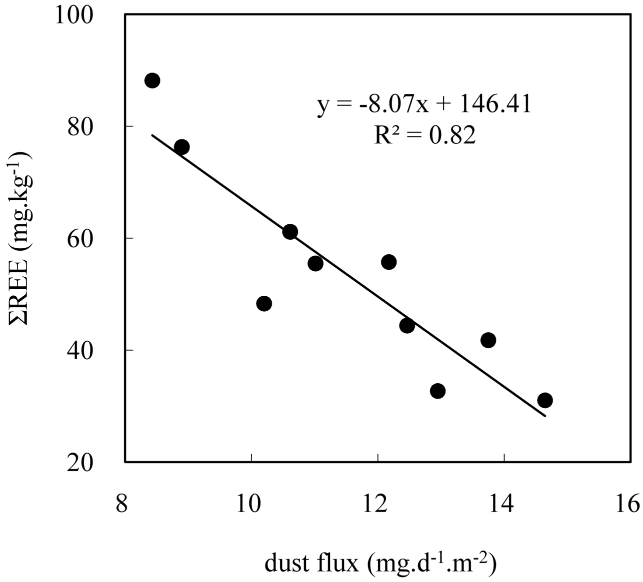

Fig. 2. Negative correlation between dust flux and ΣREE concentrations from Maolan National Natural Reserve Park.

When normalized to the local topsoil, most of the atmospheric dust samples had negative Ce anomalies and positive Eu anomalies. The compositions of REE minerals in the bauxite belt of this karst region indicate that the Eu anomaly could change subtly during weathering (Li *et al.* 2013).

The REE patterns of atmospheric dust from Maolan showed variations with different seasons. PAAS-normalized REE patterns of some dust samples were similar to that of the local topsoil, whereas other dust samples were MREE (Sm to Dy) enriched, in accord with what was observed in the urban dust in Beijing (Tang *et al.* 2013) and Guiyang. The REE of atmospheric dust normalized to local topsoil (Fig. 4) showed the LREE (La to Nd) were more enriched in dust than topsoil. In urban areas, anthropogenic particulates are probably enriched in LREE (Lu *et al.* 2007; Suzuki *et al.* 2011), as was observed in some of the atmospheric dust samples from Maolan. However, most dust samples from Maolan had different REE patterns from Guiyang and Beijing (Fig. 5), with the Moalan samples having lower La_N and La_N/Yb_N values similar to that of local soil, suggesting that the dust of Maolan is from natural sources. The relationship between La_N/Sm_N and Gd_N/Yb_N (Fig. 6), were similar to samples from Maolan and Guiyang, suggesting that dust of both areas are controlled by carbonates of the

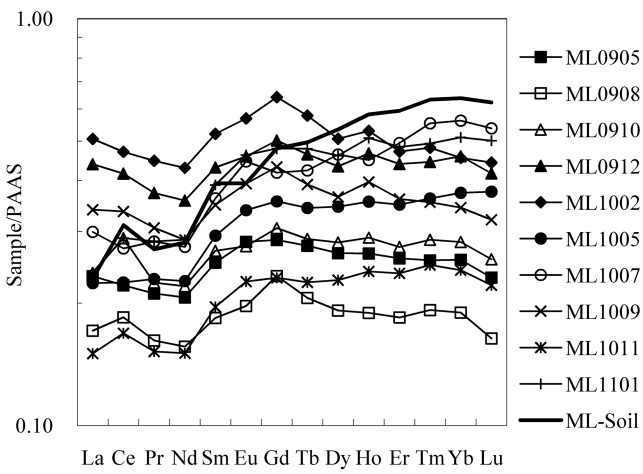

Fig. 3. PAAS-normalized REE patterns of the atmospheric dust and local topsoil.

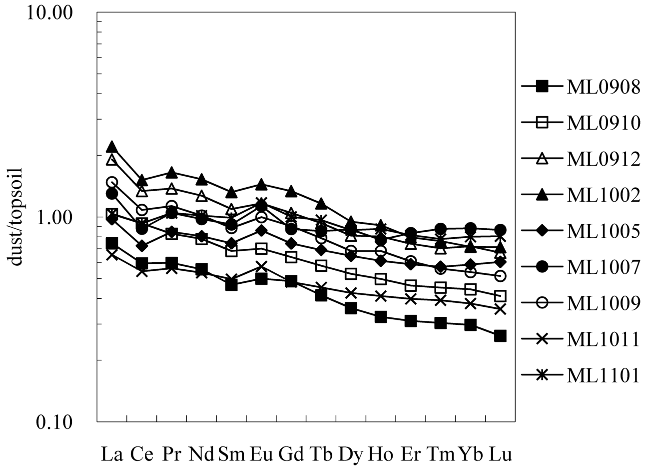

Fig. 4. Local topsoil-normalized REE diagrams of the atmospheric dust.

Karst region. Only a few samples had values similar to those of Beijing and Tokyo, indicating that these dust samples might have an anthropogenic input.

Triangular plots of La, Ce, and Sm have been used to distinguish airborne particulates from different end members (Moreno *et al.* 2008; 2010; Suzuki *et al.* 2011). Figure 7 showed that the atmospheric dust in Maolan were not inclined to any axis but overlapped with the topsoil, and most of the atmospheric particulates, except the fine particulates of Tokyo, were enriched in La and Ce (Suzuki *et al.* 2011). These relations were consistent with the Maolan dust being primarily from natural sources and urban areas.

REE enrichment factors (EF)

To better understand the sources of REE in atmospheric dust from Maolan, average REE concentrations were normalized to the corresponding local topsoil (EF values), using: $EF_d = (C_{ed}/CAl_d)/(C_{es}/CAl_s)$ where C_{ed} and CAl_d are concentrations of the element x and Al in dust and C_{es} and CAl_s are those in the local topsoil. If the EF values are close to 1, it indicates that local sources dominant. If values >5, it indicates that non-local or anthropogenic sources are

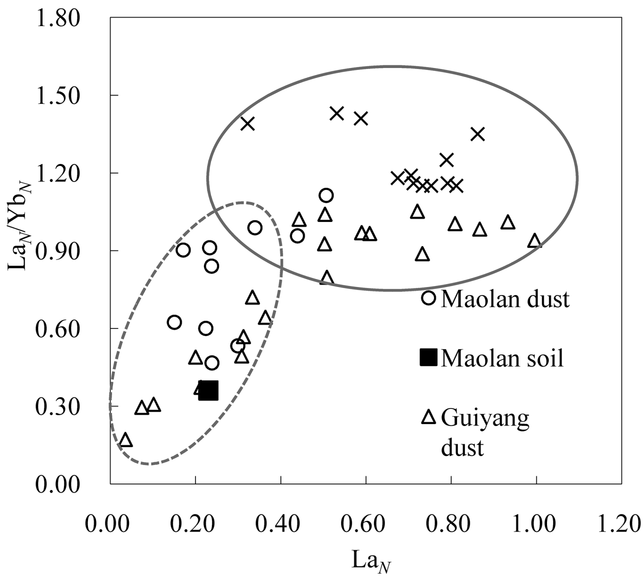

Fig. 5. Relationship between La_N and La_N/Yb_N in dust and topsoil of Maolan and dust of Beijing and Guiyang. Dust data from Beijing is from Tang *et al.* (2013), Guiyang is from Tang (pers. comm.).

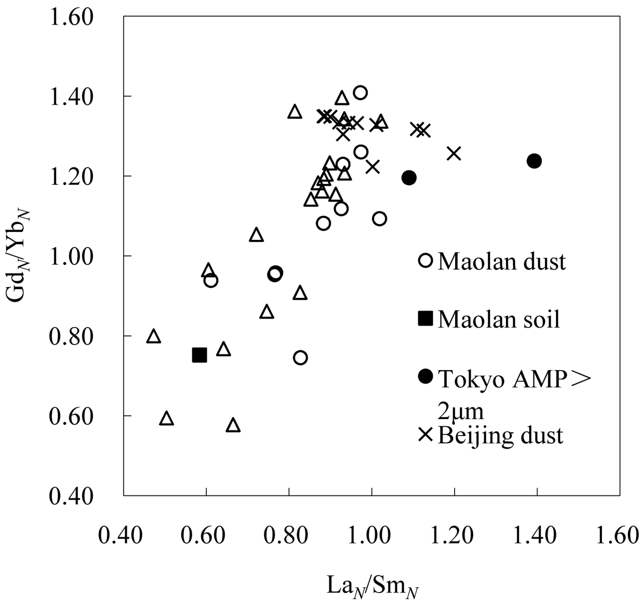

Fig. 6. Relationship between PAAS-normalized La_N/Sm_N and Gd_N/Yb_N in atmospheric dust and topsoil from Maolan National Natural Reserve Park. Data sources: APM of Tokyo (Suzuki *et al.* 2011); dust from Beijing (Tang *et al.* 2013); dust from Guiyang (Tang, pers. comm.).

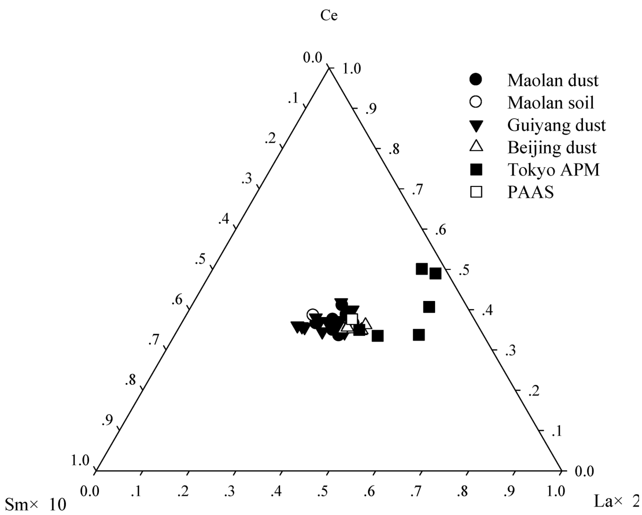

Fig. 7. Three component La-Ce-Sm plot for atmospheric dust and topsoil from Maolan National Natural Reserve Park. Data sources: PAAS (Taylor & McLennan 1985); Guiyang dust (Tang, pers. comm.); Beijing dust (Tang *et al.* 2013); Tokyo APM (Suzuki *et al.* 2011).

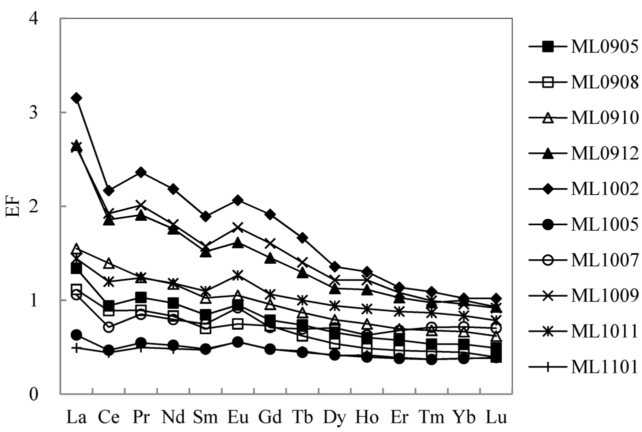

Fig. 8. Enrichment factors (EF) of dust normalized to the local topsoil.

considerable (Krachler *et al.* 2003; Sudheer & Rengarajan 2012). The EF values of REE in the dust in Maolan were similar in different seasons (Fig. 8), with EF values of the LREE being higher than the HREE. The EF values of REE in the dust at the village of Banzhai were between 0.37 and 3.15, suggesting that the dust might have a component of anthropogenic sources (Winchester *et al.* 1981).

Conclusions

The ΣREE of atmospheric dust in Maolan National Natural Reserve Park were close to the corresponding local topsoil, suggesting a significant link between them. The negative correlation between ΣREE and the corresponding dust depositional fluxes is mainly due to preference of coarse dust deposition, with lower REE concentrations than fine dust. These results demonstrated that seasonal variations of ΣREE are more likely influenced by monsoon and extreme weather conditions such as drought.

PAAS normalized REE patterns of dust from Maolan National Natural Reserve Park were enriched in LREE or MREE and depleted in HREE, being similar to the patterns of local topsoil and urban dust. The La_N, Yb_N values and La_N/Sm_N, Gd_N/Yb_N ratios indicate that the Maolan dust is mainly of local provenance mixed with anthropogenic source transported by airborne from urban areas. Most EF values of the Maolan dust were between 1 and 10, with EF values of LREE being greater than those of HREE, suggesting a mixture of natural and anthropogenic inputs.

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