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ORIGINAL ARTICLE

Contribution of Asian dust to soils in Southeast China estimated with Nd and Pb isotopic compositions

Jianwu Li[1] · Zhaoliang Song[1,4] · Lukas Van Zwieten[3] · Li Ruan[1,2] · Feili Li[5]

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Abstract Asian dust has been identified in subtropical soils of China. Neodymium (Nd) and lead (Pb) elemental and isotopic geochemistry of soils in Southeast China were used to assess the significance of local versus extraneous sources. The $\varepsilon_{Nd}(0)$ values were close to the parent rocks (+ 2.9) in the young soils (NSJ); while their values were always negative (~ -3.7) in the old soils (OMJ), implying dust deposition. The young soils contained less Asian dust contribution (< 8 %) while the old soils contained more than 50 % Asian dust contribution. This implied that dust contributions were gradually increasing with the soil development stage and Asian dust input had become the principal Nd source for the old soils in this area. On the other hand, Pb excesses and low $^{207}Pb/^{206}Pb$ values ($\sim$ 0.8503) in near-surface soils indicated a significant anthropogenic Pb addition onto surface soils. The close relationship between the Pb content and isotopic ratios in the soil profile indicated that the excessive lead in the surface soil was exogenous. These results suggested that Asian dust made up a significant fraction in the old soils, but that local sources (i.e., basalt and anthropogenic) were not trivial in Southeast China.

Keywords Aeolian dust · Soil genesis · Rural soils · Basalt

✉ Jianwu Li
jameslee@zafu.edu.cn

✉ Li Ruan
ruanli@tricaas.com

1 Key Laboratory of Soil Contamination Bioremediation of Zhejiang Province, Zhejiang A & F University, Hangzhou 311300, China

2 National Center for Tea Plant Improvement, Tea Research Institute, Chinese Academy of Agricultural Sciences, Hangzhou 310008, China

3 New South Wales Department of Primary Industries, 1243 Bruxner Highway, Wollongbar, NSW 2477, Australia

4 Institute of the Surface-Earth System Science Research, Tianjin University, Tianjin 300072, China

5 College of Environment, Zhejiang University of Technology, Hangzhou 310014, China

1 Introduction

As an important participant in the global biogeochemical cycle, soils receive material transport from the atmosphere (Brantley et al. 2007; Gross et al. 2015). Atmospheric deposition plays an important role in the formation and evolution of global soil (Li et al. 2013a, b; Zeng et al. 2015; Zhao et al. 2018). For instance, Saharan dust has a huge impact on the soil along the Mediterranean coast and throughout Europe; the annual deposition of Asian dust can be as high as 1128.22 kg m^{-2} in the arid area of northwest China and even formed the famous Loess Plateau (An 2000). The main atmospheric deposition source areas in the world bring up to 800–2000 $\times$ 10^9 kg of dust reduction per year (Lawrence and Neff 2009). The mineral aerosols and marine sediments in the Northwest Pacific mainly come from Asian dust (Zhang and Huang 1992). Atmospheric deposition is crucial for biogeochemical cycles globally and is an important source of nutrients for maintaining terrestrial ecosystems (Okin et al. 2004). Thus, the input of atmospheric deposition has a profound impact on the formation and evolution of global soils (Rea 1994; Takanori et al. 2004; Yang et al. 2016; Gross et al. 2016).

Neodymium (Nd) and lead (Pb) isotopes are powerful fingerprints in tracing soil provenance (Borg and Banner 1996; Grousset and Biscaye 2005; Albarede et al. 2012).

Nd exists in nature as a mixture of ^{143}Nd and ^{144}Nd isotopes. With the improvement of neodymium isotope geochemistry theory and the improvement of modern analysis and testing technology, Nd isotope source tracing has been widely used (Chadwick et al. 1999; Grousset and Biscaye 2005). This is because the difference between Sm–Nd isotopic composition is directly related to the source property, and this difference does not change with the weathering, transportation, and precipitation process of the source materials (Kurtz et al. 2001; Sun 2005). The Nd isotopic composition characteristics of any extraneous deposits record the provenance attributes of the source area. Therefore, Nd isotopes have been used to identify the presence of Aeolian dust in soils (Hyeong et al. 2011; Li et al. 2013a, b). The Nd isotopic ratios have also been used to improve understanding of interrelated aspects of dust content, soil composition and texture, and soil hydrology (Reynolds et al. 2006; Hyeong et al. 2011; Shi et al. 2018). In addition, lead (Pb) is a toxic element that can cause neurological, digestive, and reproductive diseases. Human beings are exposed to lead through natural and natural and anthropogenic ways (Kamenov and Gulson 2014). The U decays over geological time to produce the daughter products ^{207}Pb, ^{206}Pb, and ^{208}Pb, respectively, with one Pb stable isotope (^{204}Pb). A comparison of ^{207}Pb/^{206}Pb ratios is a traditional method for tracing provenance. The ^{207}Pb/^{206}Pb ratios from anthropogenic are higher than natural sources (Monastra et al. 2004). So the Pb content and isotope ratios have been used as a proxy of anthropogenic Pb addition in soils (Teutsch et al. 2001; Li et al. 2011), which can make up for the shortage of Nd in tracing anthropogenic sources. Ferrat et al. (2012) reported Pb atmospheric deposition rates and isotopic trends in Asian dust during the last 9.5 kyr on the eastern Qinghai–Tibetan Plateau. The use of Pb isotopic signatures can assist in the identification of Pb sources (Emmanuel and Erel 2002; Cheng and Hu 2010). Pb isotopic composition of urban soils showed some information related to Pb origins in soils (Ettler et al. 2004).

In brief, Nd and Pb isotopes are good tracers for estimating the contributions of atmospheric deposition and anthropogenic addition in soils (Chen et al. 2005; Kamenov and Gulson 2014). Zhejiang Province is located in the subtropical monsoon area in the southeast of China. Due to the impact of Asian continental high (An 2000), it is mainly affected by atmospheric deposition from the center of the North Asian continent in winter. So the atmospheric deposition has an important impact on the soil in this area (Huang et al. 2011). In addition, owing to locating the most developed area of the Yangtze River Delta in China, rapid economic development had also led to a lot of anthropogenic pollution. Does the natural atmospheric deposition or anthropogenic addition have a crucial influence on soils in Southeast China? Thier impacts and contributions still have not been resolved.

Soils derived from basalts in Southeast China provide an excellent opportunity to study the role of natural or anthropogenic sources in soil development. The soils in this study develop on geologically homogeneous basalt regolith and have distinctly different Nd and Pb isotopic ratios in comparison to continental dust (Vitousek et al. 1999; Zou et al. 2000; Kurtz et al. 2001). The study of soil provenance and evolution characteristics in this area has important theoretical and practical significance for understanding the occurrence and evolution of soil, the contribution of atmospheric source, and the pollution status of the surface environment. Therefore, we measured the Nd and Pb concentration and the stable Nd and Pb isotope compositions of soils, bedrock, aiming to (i) investigate the Nd and Pb elemental and isotopic geochemistry of soils derived from basalts in Southeast China; (ii) identify the potential natural or anthropogenic sources in soils; (iii) make clear the impacts and contributions of Asian dust and anthropogenic sources by quantifying their contributions.

2 Materials and methods

The study area is located in Xinchang-Shengzhou Basin, Southeast China, between $120°2'$ E$-121°0'$ E and $29°1'$ N$-29°5'$ N (Fig. 1). It belongs to the southern fringe of northern subtropics (Xiong and Li 1987) and has a mean annual air temperature of 16.6 °C and mean annual precipitation of 1500 mm of which nearly 70 % falls during the wet season (April–September). The soils support plants that are dominated by *Machilus thunbergii* and *Camellia* sp. Soils in this region are most commonly derived from in situ weathering of basalt (Zou et al. 2000). Two soil profiles (i.e. NSJ and OMJ profiles) were located in the rural area of Sanjie and Mengjiatang in Zhejiang province, respectively. They were selected on primary shield volcano surfaces, so surface runoff and groundwater appear not to contribute much extraneous material to soils. And the interruption is not observed in the intergradation from basalt to soils. The ages of their basalt bedrock in NSJ site and OMJ were 15 ka and 1600 ka, respectively, which were dated by K–Ar chronology. The NSJ and OMJ soils were classified as Primosols and Ferrosols (Chinese Soil Taxonomy (CST) 2001), or Entisols and Ultisol according to USDA Soil Taxonomy (Soil Survey Staff 2014), respectively. The parent rock is fresh tholeiitic basalt, which was collected beneath the sampling sites. Soil samples were taken from bottom to top according to the genetic horizon.

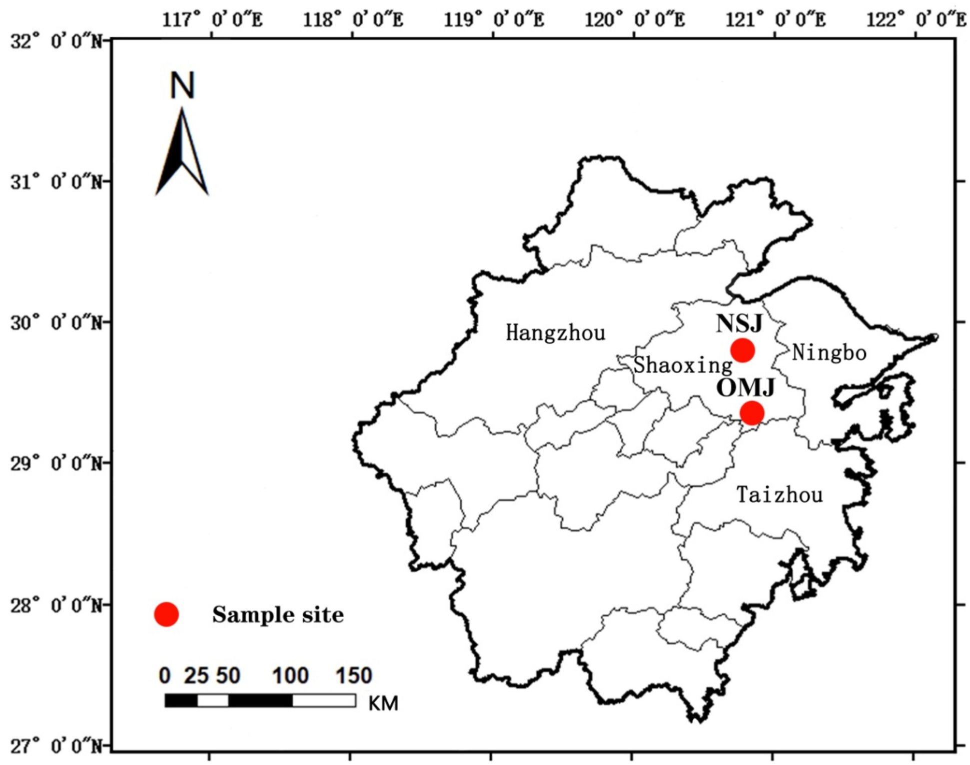

Fig. 1 The location of sampling sites

The collected soil samples were dried under natural ventilation, milled, and sieved through a 2 mm sieve for future use. General physical and chemical properties (such as pH values, soil organic matter (SOM), and soil bulk density) are determined by conventional analytical methods (Zhang and Gong 2012). For the Nd and Pb concentration determination, the soil samples were digested with an acid solution (5 ml 65 % HNO_3, 30 % HCl and 40 % HF; v/v). Then were assayed using an inductively coupled plasma mass spectrometer (ICP-MS) at the Institute of Geochemistry, Academy of Chinese Science. The standard reference materials were GSR-3 and GXR-6. Analytical uncertainties were $<$ 5 %.

The Nd and Pb isotopes were measured by inductively coupled plasma-mass spectroscopy (ICP-MS) at the University of Science and Technology of China, and the methodology of the Nd and Pb isotopes were described in Li et al. (2013a, b) and Walraven et al. (2014), with an uncertainty of 0.04 and 0.05 % for Nd and Pb isotopes measurement, respectively. The reagent blank was also measured and blank subtraction was done for the final intensity of each isotopic Nd and Pb in the sample. The $^{143}Nd/^{144}Nd$ ratio was normalized using $^{146}Nd/^{144}Nd = 0.7219$. $\varepsilon_{Nd}(0) = ((^{143}Nd/^{144}Nd_{Measured})/(^{143}Nd/^{144}Nd_{CHUR}) - 1) \times 10^4$, is the normalized present-day isotopic composition, in which the Chondritic Uniform Reservoir (CHUR) value is 0.512638.

The mass fraction of dust-derived Nd (f_{dust}^{Nd}) was calculated to estimate the contributions of dust to the soils in Southeast China by using the Nd isotope mass balance. Assuming the Nd isotope ratios of each sample is the mixture of basalts ($\varepsilon_{Nd} = +2.9$; this study) and Asian dust end members ($\varepsilon_{Nd} = -10.4$; Kurtz et al. 2001; Li et al. 2013a, b), the f_{dust}^{Nd} value was calculated using the equation (Chadwick et al. 1999):

$$f_{dust}^{Nd} = \frac{\varepsilon_{Nd}^{soil} - \varepsilon_{Nd}^{basalt}}{\varepsilon_{Nd}^{dust} - \varepsilon_{Nd}^{basalt}} \quad (1)$$

where the ε_{Nd}^{soil}, ε_{Nd}^{dust} and $\varepsilon_{Nd}^{basalt}$ are the ε_{Nd} values of soils, dust, and basalts, respectively.

3 Results

Some selected physicochemical characteristics of soils in Southeast China are shown in Table 1. The pH values range from 5.8 to 6.2 in soil samples (Table 1). The pH values in the A horizon is lower compared with the C horizon in the NSJ and OMJ profiles. The bulk density was lower in A horizon (0.98–0.99 g cm^{-3}) than in the C horizon (1.07–1.12 g cm^{-3}) for each soil profile. The soil organic matter (SOM) shows a decreasing trend with depth, with maximum values up to 41.3 g kg^{-1}, and 37.9 g kg^{-1} in the A horizon of the NSJ and OMJ profiles, respectively.

Table 1 Selected physicochemical characteristics of soil profiles

Soil profiles	Age (ka)	Depth (cm)	Horizon	pH	Bulk density (Mg m^{-3})	SOM (g kg^{-1})
NSJ	15	0–15	A	5.96	0.99	41.3
		15–40	AC	5.88	1.05	23.4
		40–70	C	6.12	1.07	8.9
OMJ	1600	0–15	A	5.89	0.98	37.9
		15–40	B1	5.78	1.01	28.6
		40–70	B2	5.83	1.06	27.2
		70–90	BC	6.02	1.09	18.5
		90–135	C	6.15	1.12	7.3

Nd and Pb concentration of soils and parent rock (i.e. basalt) was shown in Table 2. Nd concentrations of soil samples ranged from 27.0 to 38.5 mg kg^{-1} and 29.5 to 51.2 mg kg^{-1} in the NSJ and OMJ profiles, respectively. Nd concentrations of soils in B horizon were higher than those of A and C horizons for the two profiles (Table 2). On the other hand, Pb concentrations of soils in A horizons were extremely high compared to the fresh basalt (3.9 mg kg^{-1}). On the surface of soil profiles, Pb concentrations can reach up to 15.3 and 17.8 mg kg^{-1} in the NSJ and OMJ profiles, respectively. Furthermore, Pb concentration of soils showed a rapidly increasing trend from C horizon to A horizon, showing remarkable enrichment of Pb content for soils of Southeast China in near-surface horizons.

Nd and Pb isotope ratios of soils in Southeast China were shown in Table 2. The $^{143}Nd/^{144}Nd$ ratios of soil samples were from 0.512732 to 0.512786 and 0.512449 to 0.512745 for NSJ and OMJ profile, respectively. The $^{143}Nd/^{144}Nd$ ratios (0.512745) of soils in A horizon for OMJ profile were far from those values of their parent rock (Table 2). The significantly low $^{143}Nd/^{144}Nd$ ratios in OMJ profile were close to values of continental dust (Jahn et al. 2001; Sun 2005), implying likely influence of Nd from continental dust (Kurtz et al. 2001; Li et al. 2013a, b). On the other hand, the $^{207}Pb/^{206}Pb$ and $^{208}Pb/^{206}Pb$ ratio of soil samples ranged from 0.8375 to 0.8503 and 2.0906 to 2.1071, respectively (Table 2). The $^{207}Pb/^{206}Pb$ values for soils also showed a decreasing trend with depth. It was worth noting that the $^{207}Pb/^{206}Pb$ ratios of soil samples in the top horizons (0–20 cm) were significantly high

Table 2 Concentrations and isotopic ratios of Nd and Pb in soil samples for the NSJ and OMJ profiles

Soil profiles	Depth (cm)	Nd (mg kg^{-1})	Pb (mg kg^{-1})	$^{143}Nd/^{144}Nd$	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$
NSJ	0–5	31.8	15.3	0.512732	2.0991	0.8503
	5–10	32.8	14.3	0.512758	2.0992	0.8501
	10–15	30.3	9.8	0.512739	2.0992	0.8497
	15–20	34.1	8.1	0.512760	2.1010	0.8479
	20–30	38.5	7.3	0.512775	2.0969	0.8453
	30–40	37.8	7.0	0.512782	2.0961	0.8438
	40–50	29.1	6.4	0.512779	2.0946	0.8428
	50–70	27.0	5.0	0.512786	2.0906	0.8412
OMJ	0–5	31.4	17.8	0.512449	2.1032	0.8478
	5–10	36.5	17.1	0.512494	2.1031	0.8478
	10–20	42.6	13.4	0.512481	2.1024	0.8471
	20–30	44.0	11.9	0.512546	2.0992	0.8464
	30–40	47.3	11.6	0.512560	2.1020	0.8447
	40–50	42.8	10.5	0.512582	2.0983	0.8440
	50–60	36.7	9.9	0.512612	2.0988	0.8439
	60–75	51.2	9.1	0.512616	2.0990	0.8437
	75–90	42.9	9.2	0.512602	2.0991	0.8440
	90–105	37.8	7.0	0.512675	2.1067	0.8413
	105–120	36.9	7.1	0.512725	2.1071	0.8410
	120–135	29.5	5.4	0.512745	2.1069	0.8375
Fresh basalt		23.1	3.9	0.512787	2.0760	0.8361

(0.8541), with a value that far from those value of their parent rock, implying anthropogenic Pb input in research areas (Erel et al. 1997; Duzgoren-Aydin et al. 2004).

4 Discussion

4.1 Nd isotopic characterization of the potential sources

Besides the parent rock, long-range transport of Asian dust is another important contributor to soils (Chadwick et al. 1999; Lehmkuhl et al. 2014). On the one hand, the $\varepsilon_{Nd}(0)$ values of soils in Xinchang–Shengzhou Basin showed a trend with depth, with negative values near the surface and positive values similar to the bedrock at depth (Fig. 2). Because soils were developed from in situ weathering of basalt (see the "Materials and methods" section), all potential Nd source changes display signatures of parent rock (i.e. basalt) (Table 2). In the subsoil near the bedrock (> 100 cm), the $\varepsilon_{Nd}(0)$ value of soil samples was positive. This is due to the fact that the weathering of in situ weathering soil profile gradually strengthened from bottom to top, and the closer to the subsoil, the stronger the inheritance characteristics of parent rock provenance. While in the near-surface layer, due to the strong weathering and leaching in the tropical area, the material inheritance from the parent rock in the soil is obviously weakened; and owing to the continuous replenishment and accumulation of dust deposition, the $\varepsilon_{Nd}(0)$ value of the near-surface soil is significantly reduced, with the negative values, which is closer to the $\varepsilon_{Nd}(0)$ value of Asian dust (− 10.44; Fig. 2). On the other hand, the $\varepsilon_{Nd}(0)$ values of soils were from − 3.7 to + 2.1 for the old soils of OMJ (1600 ka), while those values of the young soils (NSJ, 18 ka) were positive (> + 1.8). This result showed that the dust impact was likely to be much less than at OMJ because the Nd values are more similar to the parent rock (basalt) for NSJ. And this result was also in line with the assumption that the soil's residence time of NSJ is shorter than that of OMJ, because there had not been enough time at NSJ for large amounts of dust to accumulate.

Mass fractions of dust-derived Nd (f_{dust}^{Nd}) were calculated according to equal (1). For the young soils (NSJ), the f_{dust}^{Nd} values of all soil samples were low (< 8 %), implying less influence of Asian dust (Fig. 3). But for the old soils (OMJ), the f_{dust}^{Nd} values in soil samples of deeper horizons were low, indicating the dominance of lava-derived Nd; while the f_{dust}^{Nd} values for soils in A horizon were significantly high (∼ 49 %; Fig. 3), implying great contributions of Asian dust to the soils.

The deserts and loess in northern and northwestern China are some of the largest sources of global atmospheric dust (Merrill et al. 1994). Asian dust is transported globally

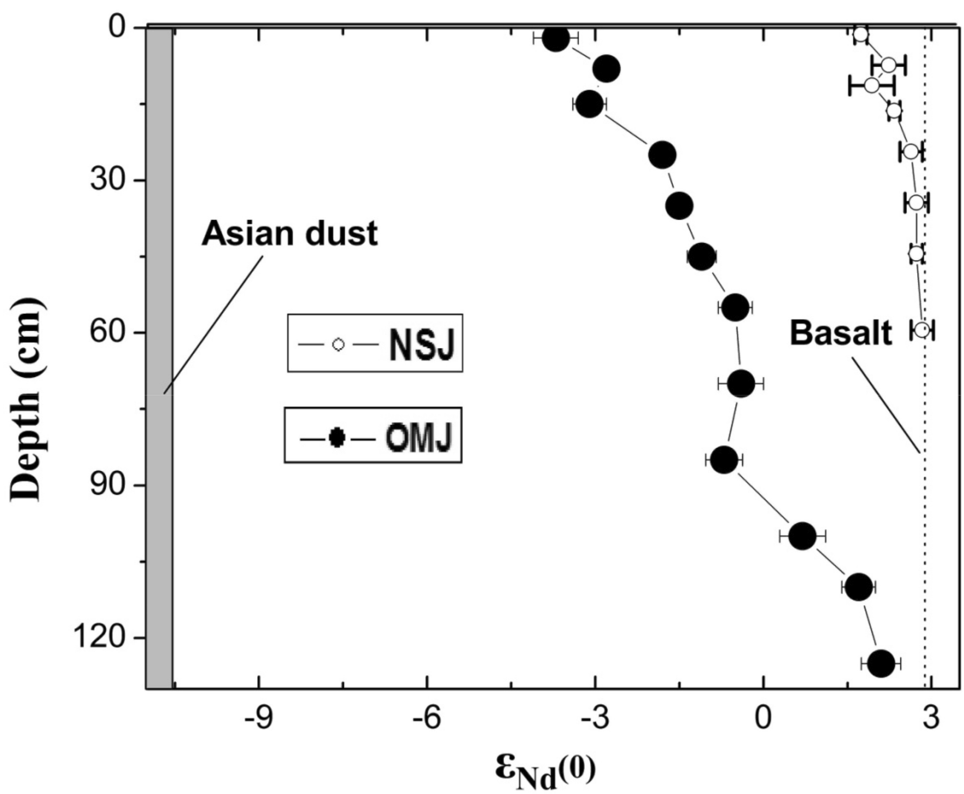

Fig. 2 The values of $\varepsilon_{Nd}(0)$ for soils in Southeast China. The dotted line and gray solid rectangle represent the $\varepsilon_{Nd}(0)$ values of basaltic bedrock and Asian dust, respectively

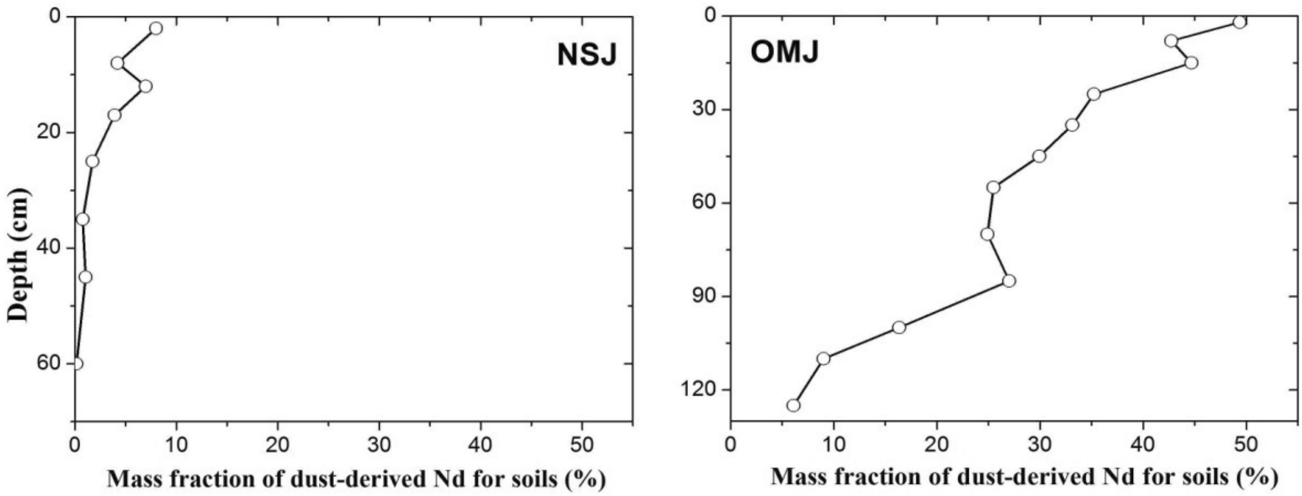

Fig. 3 Mass fraction of anthropogenic Nd for soils

and has been found in aerosols and archives in regions as far removed as Hawaii (Kurtz et al. 2001) and marine sediments (Rea 1994). Soil profiles under study were located in Southeast China, close to the source of the Asian continental dust which may contribute large amounts of mineral material to most areas of China (An 2000; Sun 2005; Jiao et al. 2018). Furthermore, the loess-like Quaternary red clay and the Xiashu Loess were found in South China, indicating the occurrence of heavy dust deposition (Li et al. 2001; Xiong et al. 2002). The Xiashu Loess was located in Zhenjiang of Southeast China, which was believed to be Asian dust in origin (Li et al. 2001). The loess-like Quaternary red clay showed the occurrence of Asian dust deposition between 29° N and 31° N in subtropical China (Hu et al. 2010). Li et al. (2013a, b) also reported marked Aeolian characteristics of the red clays in the Jinhua–Quzhou Basin, Southeast China by grain size evidence. Our study area was near the areas of red clays and the Xiashu Loess (Fig. 1). Similarly, the old soils should partly originate from distant dust sources in Northwest China and share the same source provenance with the loess in the Chinese Loess Plateau (Li et al. 2001; Xiong et al. 2002). In brief, our results showed that the weathering of basalt was the principal Nd source for the young soils (NSJ) and Asian dust should have less influence on the soils; but for the old soils (OMJ), besides the parent rock, Asian dust was the main Nd source of in south China.

4.2 Pb isotopic characterization of the potential sources

Based on the identification of the natural source for soils by Nd isotopes, lead isotope was used as another tracer of anthropogenic input in soils, because it can make up for the shortage of Nd isotopes in tracing anthropogenic sources (Ferrat et al. 2012). The $^{207}Pb/^{206}Pb$ ratios of natural Pb in basalt (0.836; this study) and Asia dust (Jones et al. 2000) were approximate, so the natural source can be regarded as a whole. We took the natural and anthropogenic Pb sources as a two end-member mode. The $^{207}Pb/^{206}Pb$ ratios of soil samples in the C horizon (0.837) were close to the ratio of natural Pb sources (Table 2). Besides natural sources, anthropogenic Pb sources may contribute remarkably to the soils. Pb concentrations of the surface soils (17.8 mg kg^{-1}) were several times higher than the parent bedrocks (3.9 mg kg^{-1}; Table 2). The enrichment of surface soils Pb concentrations indicated anthropogenic Pb contributions to soils (Monastra et al. 2004).

Cheng and Hu (2010) reviewed the types of Anthropogenic Pb sources in China and had given their $^{207}Pb/^{206}Pb$ ratios. Chinese coal had an average $^{207}Pb/^{206}Pb$ ratio of 0.845, whilst $^{207}Pb/^{206}Pb$ ratio of the leaded petroleum ranged up to 0.900 (Erel et al. 1997). So the higher $^{207}Pb/^{206}Pb$ values in surface soils might have partially resulted from coal burning or petroleum usage. Another possible source with low $^{207}Pb/^{206}Pb$ ratios was Pb ores. Zhang et al. (2007) reported that the $^{207}Pb/^{206}Pb$ ratios of Chinese Pb ores from south China ranged from 0.840 to 0.865, which were very close to those of topsoils in NSJ and OMJ profiles (Fig. 4). Meanwhile, ancient mining and smelting activities were begun around 1600 B.C. (Peng et al. 1999). Long ores mining history was likely to have an important influence on the exogenous input to topsoils (Mukai et al. 2001). Thus, Pb isotope ratios of topsoils in our study is close to that of Pb ores in south China, particularly that of Jiangsu–Zhejiang region, which is neighboring with Shaoxing. This indicates that top soils in our study were more or less affected by the surrounding areas in terms of Pb contamination. Although surface soils were

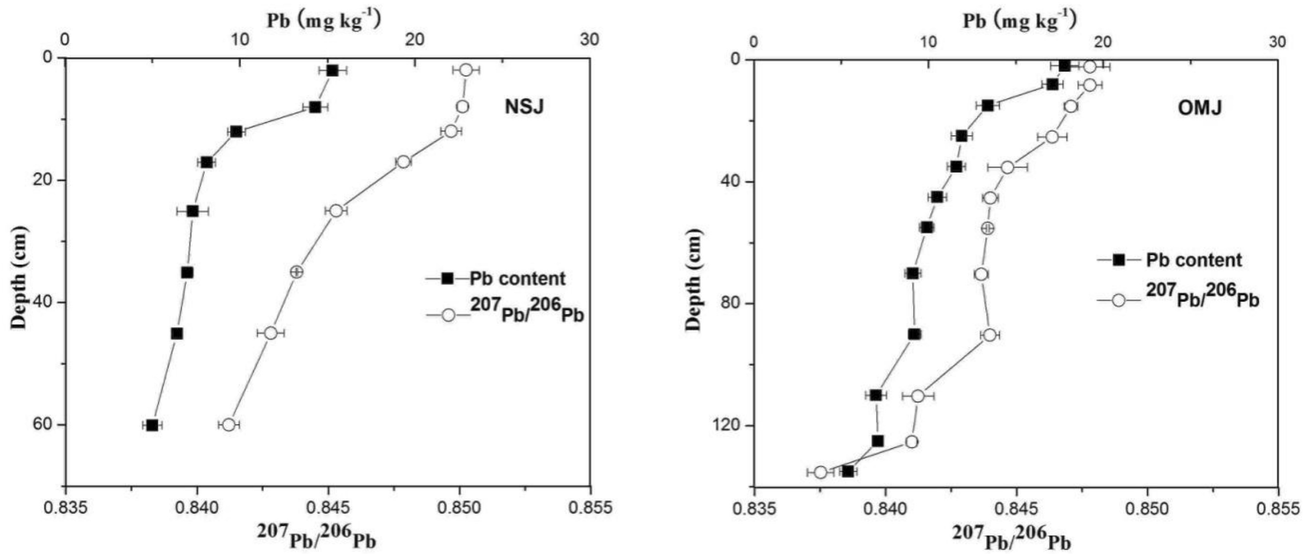

Fig. 4 Pb content and $^{206}Pb/^{207}Pb$ ratios for soils

found normally contaminated by Anthropogenic Pb from coal burning or petroleum combustion or Pb ores mining, we still need more data to identify the different Anthropogenic Pb sources and quantify the contribution of the sources discussed above in the future. In addition, as shown in Table 2, the $^{207}Pb/^{206}Pb$ ratios were from 0.841 to 0.850 and 0.838 to 0.848 for the NSJ and OMJ soils, respectively. This suggested that Pb sources with a higher $^{207}Pb/^{206}Pb$ value should have been mixed in the soils (Fig. 4). It is noteworthy, the close correlation between Pb concentration and $^{207}Pb/^{206}Pb$ ratios in soil profiles suggested that the excess lead in the surface soils was exogenous. High $^{207}Pb/^{206}Pb$ ratios were consistent with an anthropogenic source for the exogenous Pb.

5 Conclusions

Besides the parent rock, Asian dust and exogenous anthropogenic sources have been identified in subtropical soils of China. The $\varepsilon_{Nd}(0)$ values were close to the parent rocks in the young soils; while the $\varepsilon_{Nd}(0)$ values are always negative in the old soils, which is significantly lower than the parent rocks, implying involvement of dust deposition. The f_{dust}^{Nd} values showed the contributions of Asian dust. The old soils (OMJ) contain 10–50% of Asian dust, and the young soils (NSJ) contain less Asian dust (< 8%) and more material released by the in situ weathering of basalt.

Due to the Pb isotopic ratios of basalt (parent rock) and Asia dust was very approximate, we can take them as the same end-member, that is to say, natural sources. We took the natural and anthropogenic Pb sources as a two end-member mode. The extremely high Pb concentrations compared to the fresh basalt indicated remarkable Pb enrichment for soils in near-surface soils, implying exogenous inputs of Pb. The $^{207}Pb/^{206}Pb$ ratios of the deep soils profiles were closer to basalt, implying influence of parent rocks; while the noticeably high $^{207}Pb/^{206}Pb$ ratios of top soils (< 30 cm) indicated external anthropogenic Pb sources. High $^{207}Pb/^{206}Pb$ ratios and the corresponding high Pb contents of top soils showed that besides the weathering of their parent rock, external anthropogenic Pb sources may exist. The results of the present study suggest that Asian dust makes up a significant fraction in the old soils, but that local sources (i.e., basalt and anthropogenic) are not trivial.

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Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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