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Metasomatized Lithospheric Mantle beneath the Western Qinling, Central China: Insight into Carbonatite Melts in the Mantle

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ABSTRACT

Mantle xenoliths from the Western Qinling, central China, are dominated by lherzolites, which can be divided into four subgroups—namely, garnet-facies, coexisting spinel-garnet, spinel-facies, and carbonate-bearing ones. All these rocks display light rare earth element enrichment, positive Sr and Ba anomalies, carbonatite-like trace element patterns, and Sr-Nd-Pb isotopic mixing between depleted mantle and enriched mantle type II end members, consistent with the geochemical features resulting from carbonatite metasomatism. The garnet-facies lherzolites show high trace element concentrations but low La_N/Yb_N ratios, and they show high Sr and Pb isotopic ratios that are similar to those of carbonatites, suggesting that they were highly metasomatized. The spinel-facies group has the lowest trace element concentrations but higher La_N/Yb_N ratios than the garnet-facies group; their lowest Sr and Pb isotopic ratios are closer to those of the depleted mantle end member, implying low-degree metasomatism. Geochemical variation of the coexisting spinel-garnet sample lies between that of the garnet and spinel groups. The elevated and highly variable trace element concentrations and Sr-Pb isotopic values of the carbonate-bearing lherzolite group are most likely related to the modal content of carbonate minerals. Collectively, these geochemical features indicate a rising front of carbonatite metasomatism in the lithospheric mantle beneath the Western Qinling. Combining experimental and empirical data, the positive Pb, Y, and high-field strength element anomalies in the peridotites might be ascribed to the involvement of a subduction component in the carbonatite melts. On the basis of the data presented in this article, we propose a general model for carbonatite metasomatism in the lithospheric mantle to interpret the different signatures recorded in the garnet-facies peridotites (chemical imprint) and spinel-facies peridotites (occurrence of carbonate minerals), which has potential application to other regions that have undergone carbonatite metasomatism.

Online enhancements: appendix tables.

Introduction

One of the most remarkable physicochemical characteristics of carbonatite melts is their low viscosity and density (Treiman 1989; Genge et al. 1995; Dobson et al. 1996), which makes them more likely than silicate melts to infiltrate and spread upward in the mantle via grain boundary percolation, crack propagation, and/or thermal fracturing (Frezzotti et al. 2002; Rosatelli et al. 2007). Consequently, carbonatite metasomatism has been recognized as a widespread and important process in the subcontinental lithosphere (O'Reilly and Griffin 1988; Genge et al. 1995; Neumann et al. 2002). Such an upwelling carbonatite metasomatic front not only would have a marked influence on the composition and structure of the lithospheric mantle but also would potentially result in compositional heterogeneities (Rudnick et al. 1993; Wang and Gasparik 2000; Gasparik and Litvin 2002). It has been proposed that subduction-induced carbonate recycling could change the composition of the carbonatite

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melts, which consists of one of the major types of metasomatic agents in the lithospheric mantle (Rea and Ruff 1996; Plank and Langmuir 1998; Laurora et al. 2001; Walter et al. 2008). Hence, carbonatized mantle xenoliths under subduction settings have attracted considerable attention to investigate the process of carbonatite metasomatism in the lithospheric mantle, despite the rarity of such samples (Vidal et al. 1989; Maury et al. 1992; Szabo et al. 1995, 1996; Laurora et al. 2001).

The Qinling-Dabie Orogen, located in central China (fig. 1), is a tectonic suture of the subduction of the Tethyan Ocean and the subsequent collision between the North China Craton and the Yangtze Craton (Zhang et al. 2001*a*, 2002, 2009; Zhao et al. 2009; Dong et al. 2011). In the western part of the orogen, known as the Western Qinling, numerous mantle xenoliths are exposed that are considered to have experienced variable degrees of carbonatite metasomatism (Su et al. 2009, 2010*a*, 2010*b*, 2012*a*). These xenoliths thus offer us a good opportunity to trace the process of carbonatite metasomatism and the origin of carbonatite melts. In this article, we present whole-rock trace element and Sr-Nd-Pb isotope data on 11 peridotite samples collected from the Western Qinling, and we attempt to evaluate the influence of carbonatite metasomatism in the lithospheric mantle and address the question of whether the subduction process has made any significant contribution to the composition of the carbonatite melts.

Regional Geology and Samples

The Qinling-Dabie Mountains are tectonically sutured between the North China Craton and the Yangtze Craton (fig. 1*A*). This central orogenic belt was formed after the closure of the Paleo-Tethyan Ocean and the collision between the North China and Yangtze Cratons from the Paleozoic to the Mesozoic (Zhang et al. 2001*a*, 2002). Voluminous exhumation of subduction-related ultra-high-pressure rocks (i.e., eclogites) took place in the Dabie Mountains rather than in the Qinling Mountains (Zhang et al. 2001*a*; Li et al. 2009). However, numerous Cenozoic volcanic rocks (7–23 Ma) outcrop in the Tianshui-Lixian fault basin in the Western Qinling and are classified as kamafugites, a type of ultrapotassic rock that is associated with carbonatites (fig. 1*B*; Yu and Zhang 1998; Yu et al. 2003, 2004, 2005; Mo et al. 2006; Dong et al. 2008; Zhao et al. 2009; Tang et al. 2012). Mantle xenoliths are mainly entrained in the Haoti and Baiguan kamafugite cinder cones and have been studied extensively to decipher the nature and deep processes of the lithospheric mantle beneath the Western Qinling (Yu et al. 2001; Shi et al. 2003; Su et al. 2006, 2007, 2009, 2010*a*, 2010*b*, 2011*a*, 2011*b*, 2012*a*, 2012*b*).

The Cenozoic lithosphere beneath the Western Qinling is geothermally hotter than typical cratonic and oceanic environments, and its thickness is estimated as ca. 120 km with a garnet-spinel transition zone at a depth of ca. 74 km (Shi et al. 2003; Su et al. 2007, 2010*b*). The lithosphere in this region is compositionally stratified with a gradual decrease in fertility with depth, a feature that probably resulted from the varying degrees of partial melting and metasomatism (Su et al. 2009, 2010*a*, 2010*b*, 2011*b*). The carbonatite metasomatism in this region was easily recognized on the basis of the presence of carbonate veins and discrete grains, the major and trace element compositions of clinopyroxenes, and in situ Li isotopes of constitute minerals (Su et al. 2009, 2010*b*, 2012*a*, 2012*b*). All the xenoliths studied here are lherzolites, including eight samples collected from Haoti and three from Baiguan. The Haoti group is composed of four garnet-facies, two spinel-facies, and one coexisting spinel-garnet lherzolites, whereas all the Baiguan group samples are spinel-facies lherzolites and contain carbonate minerals that imply modal carbonatite metasomatism. The garnet lherzolites preserve breakdown garnets, most of which show relict cores and kelyphite rims consisting of spinel-clinopyroxene-orthopyroxene mineral assemblage (fig. 2*A*, 2*B*; Su et al. 2007). The coexisting spinel-garnet lherzolites are not commonly present in the Western Qinling peridotite suite and have discrete spinel and garnet grains in an individual sample, and spinels are also observed in kelyphite rims of garnets (fig. 2*C*, 2*D*). The spinel lherzolites display various mineral textures, including spongy texture of clinopyroxene and spinel, breakdown texture of orthopyroxene, deformed texture of olivine, and melt pocket with calcite occurrence (fig. 2*E*, 2*F*; Su et al. 2010*a*, 2010*b*, 2011*a*). These xenoliths could thus represent different depths of the mantle domains that have undergone variable degrees of metasomatism. Detailed petrological features of these samples have been described in our previous studies (Su et al. 2009, 2010*a*, 2010*b*, 2011*a*), along with their mineral compositions (Su et al. 2010*a*, 2011*a*).

Analytical Methods

The analyses of trace element abundances and Sr-Nd-Pb isotopes of all 11 samples in this study were carried out at the Analytical Laboratory of Beijing

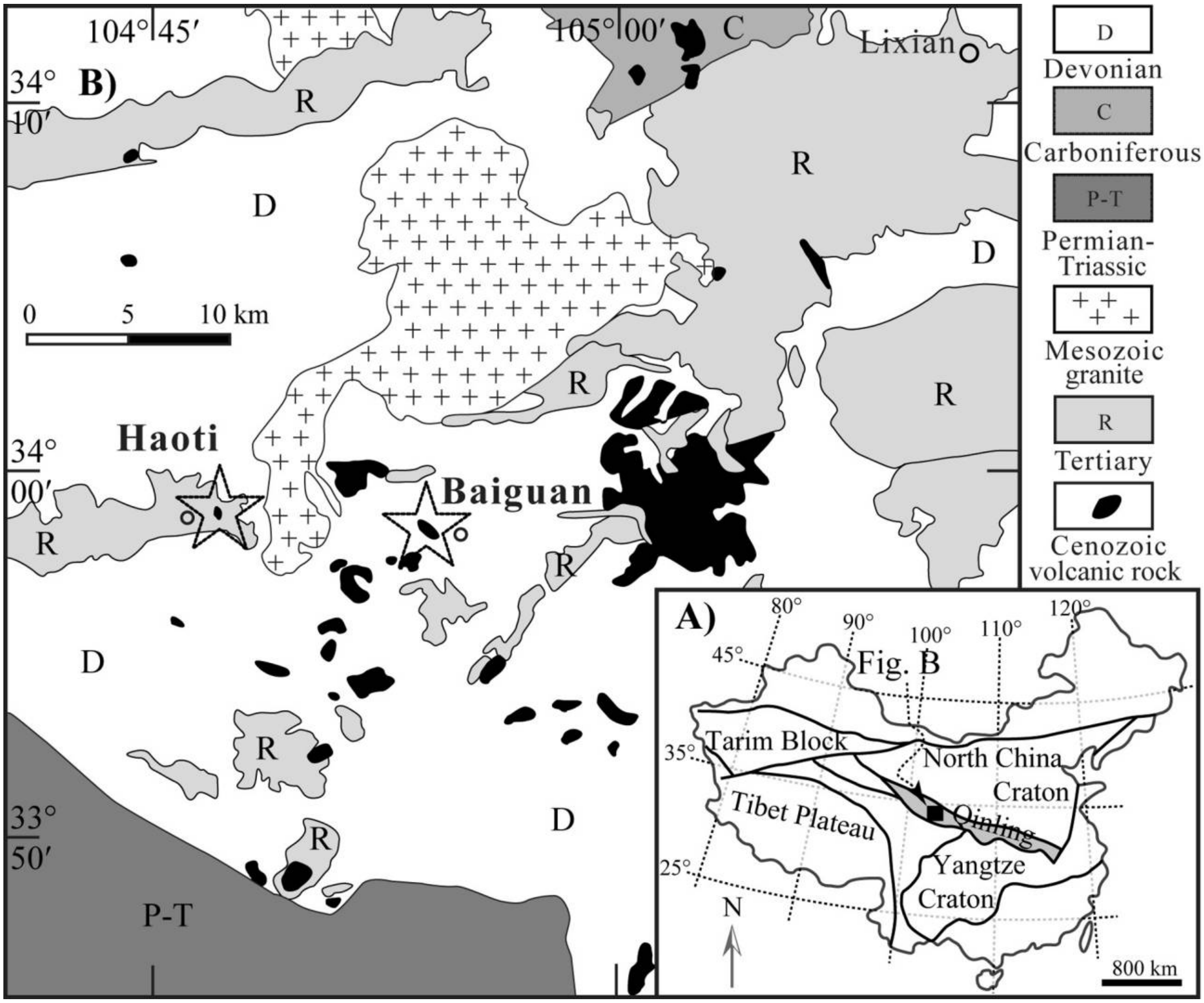

Figure 1. *A*, Generalized tectonic framework of China showing the major blocks and the location of the study area. B, Geological map showing the distribution of Cenozoic kamafugites in the Western Qinling and sample localities (Haoti and Baiguan; modified from Yu et al. 2004 and Dong et al. 2008).

Research Institute of Uranium Geology. Trace elements were determined by inductively coupled plasma mass spectrometry (ICP-MS). Samples were digested using a mixture of ultrapure HF and HNO_3 in Teflon bombs. Analytical procedures are described in detail in Chu et al. (2009). The precisions of the ICP-MS analyses were generally better than 5%.

Rb-Sr, Sm-Nd, and Pb isotopic analyses were performed on a VG-354 thermal ionization magnetic sector mass spectrometer. Procedures for chemical separation and isotopic analyses followed those of Zhang et al. (2001*b*). Mass fractionation corrections for Sr and Nd isotopic ratios were based on $^{86}Sr/^{88}Sr$ and $^{146}Nd/^{144}Nd$ values of 0.1194 and 0.7219, respectively. Uncertainties of Rb/Sr and Sm/Nd ratios were less than 2% and 0.5%, respectively.

Analytical Results

The Western Qinling xenoliths display large variations in trace element chemistry and show distinct chondrite-normalized rare earth element (REE) and primitive-mantle-normalized trace element patterns (table A1, available in the online edition or from the *Journal of Geology* office; fig. 3), in accordance with the different mineralogical compositions of different rock types. The REE concentrations show an overall decreasing trend from garnet lherzolites (REE: 10.3~36.8 ppm) through spinel-garnet lherzolite (REE: 22.5 ppm) to spinel lherzolites (REE: 6.11~13.8 ppm), whereas the La_N/Yb_N (the subscript N stands for chondrite normalized) ratios show an increase from 2.37 to 22.0 through 10.9 to 11.0~16.7. In particular, the car-

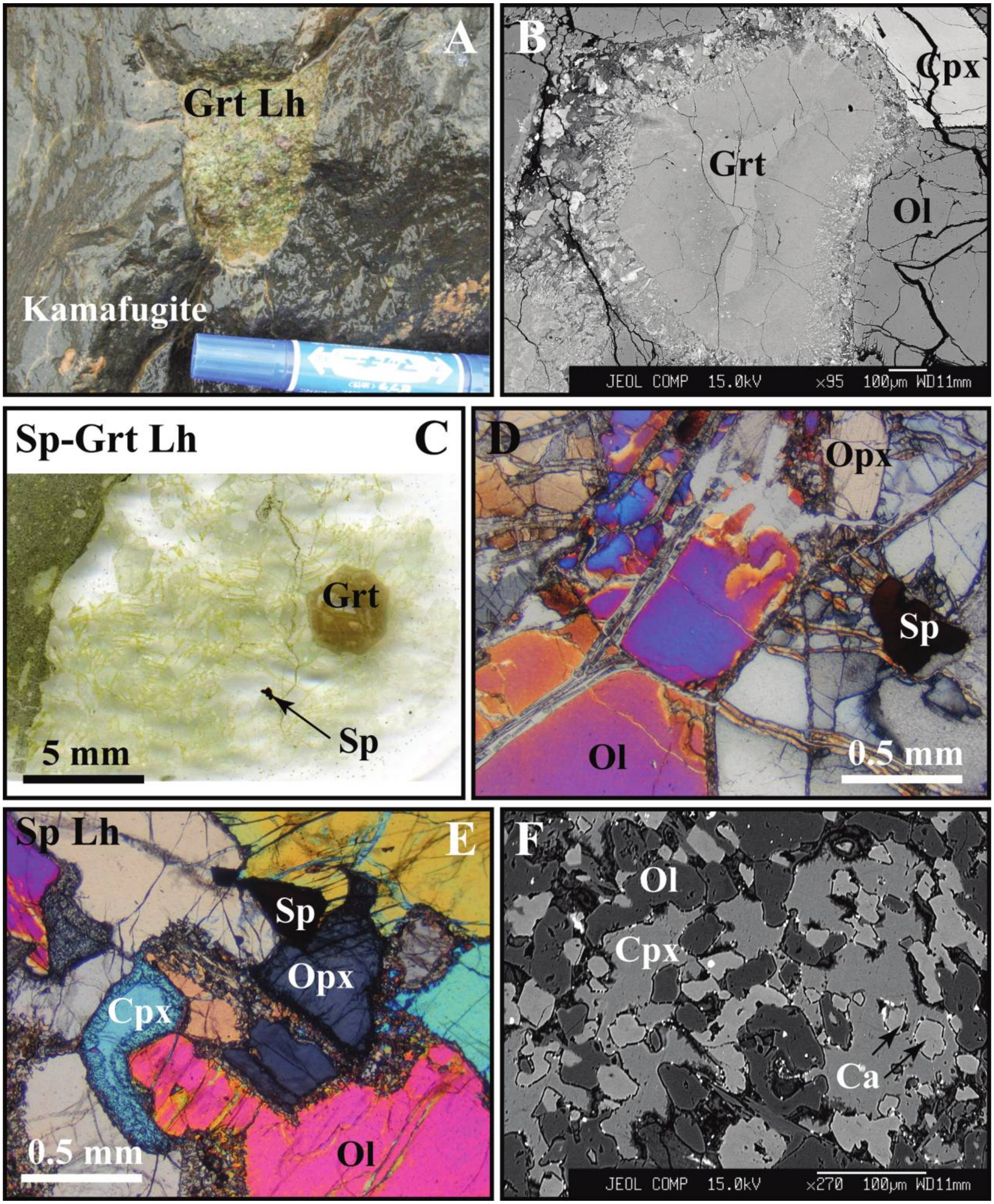

Figure 2. Petrophotographical features of the peridotite xenoliths from the Western Qinling. *A*, *B*, Occurrence of garnet lherzolite (Grt Lh) in the host kamafugite and the breakdown texture of garnet. *C*, *D*, Mineral assemblage and relationship in spinel-garnet lherzolite (Sp-Grt Lh). *E*, *F*, Typical textures of minerals and occurrence of calcite in spinel lherzolite (Sp Lh).

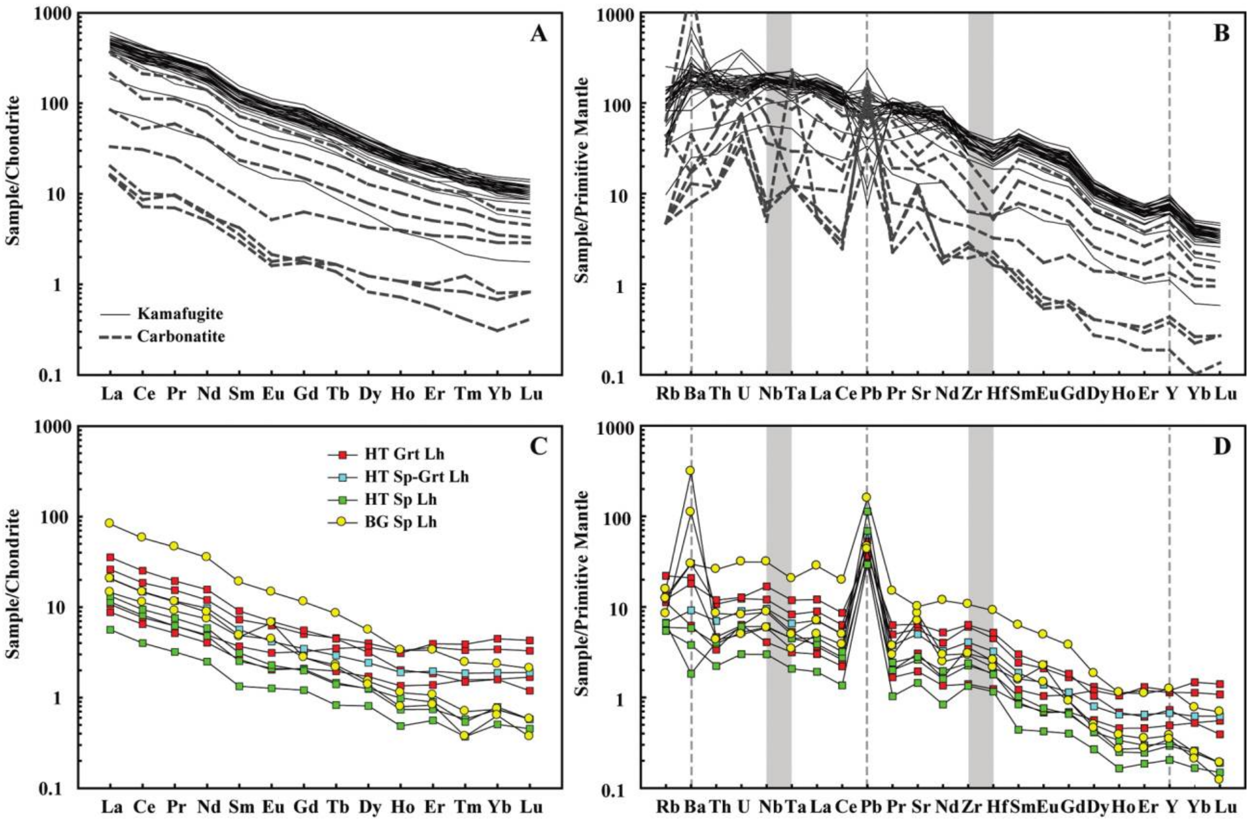

Figure 3. Rare earth element patterns and spider diagrams of the kamafugites, carbonatites, and peridotite xenoliths from the Haoti (HT) and Baiguan (BG) localities in the Western Qinling. Kamafugite data are compiled from Yu and Zhang (1998), Wang and Li (2003), Yu et al. (2004), and Dong et al. (2008). Carbonatite data are compiled from Yu et al. (2004). Chondrite-normalizing values are from Anders and Grevesse (1989), and primitive-mantle-normalizing values are from Sun and McDonough (1989). Grt Lh = garnet lherzolite, Sp-Grt Lh = spinel-garnet lherzolite, Sp Lh = spinel lherzolite.

bonate-bearing xenoliths from Baiguan possess higher REE concentrations (16.8–83.0 ppm) and markedly higher La_N/Yb_N ratios (19.3~35.0) than those of other groups (table A1). On the other hand, their trace element patterns are all characterized by light REE (LREE) enrichment, flat heavy REE (HREE) distribution (fig. 3), apparent positive Pb and Sr anomalies, slightly positive high-field strength elements (HFSE; i.e., Nb, Zr, and Hf), Gd and Y anomalies, and negative Th anomalies (fig. 3). Most importantly, the pronounced Ba-enriched features make these carbonate-bearing xenoliths remarkably distinctive from their counterparts (fig. 3).

The Sr-Nd-Pb isotopic results are presented in table A2 (available in the online edition or from the *Journal of Geology* office). All the peridotite xenoliths analyzed in this study show restricted Sr and Nd isotopic compositions ($^{87}Sr/^{86}Sr$: 0.704076~0.705578; $^{143}Nd/^{144}Nd$: 0.512697~0.512991) but possess large variations in Pb isotopic ratios ($^{206}Pb/^{204}Pb$: 16.142~18.847; $^{207}Pb/^{204}Pb$: 15.159~15.639; $^{208}Pb/^{204}Pb$: 36.107~38.928; table A2). Their Sr isotope values exhibit an increasing trend from spinel-facies to garnet-facies and carbonate-bearing peridotites, overlapping with the ranges of kamafugites and carbonatites (fig. 4*A*; Yu and Zhang 1998; Stoppa et al. 2003; Yu et al. 2004; Mo et al. 2006; Zhao et al. 2009). The most striking feature of the Western Qinling peridotites is that they have very low U/Pb ratios (table A1) and a marked linear correlation between Pb isotopic ratios. As shown in figure 4*B*, the spinel lherzolites have the lowest $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios, whereas the carbonate-bearing lherzolites have the highest values, intersecting the North Hemisphere reference line at the spinel-garnet lherzolite.

Discussion

Upwelling Carbonatite Metasomatism. For the carbonatite-peridotite system, Ba, Sr, and LREE are generally incompatible, whereas Sc, Y, HFSE (Hf

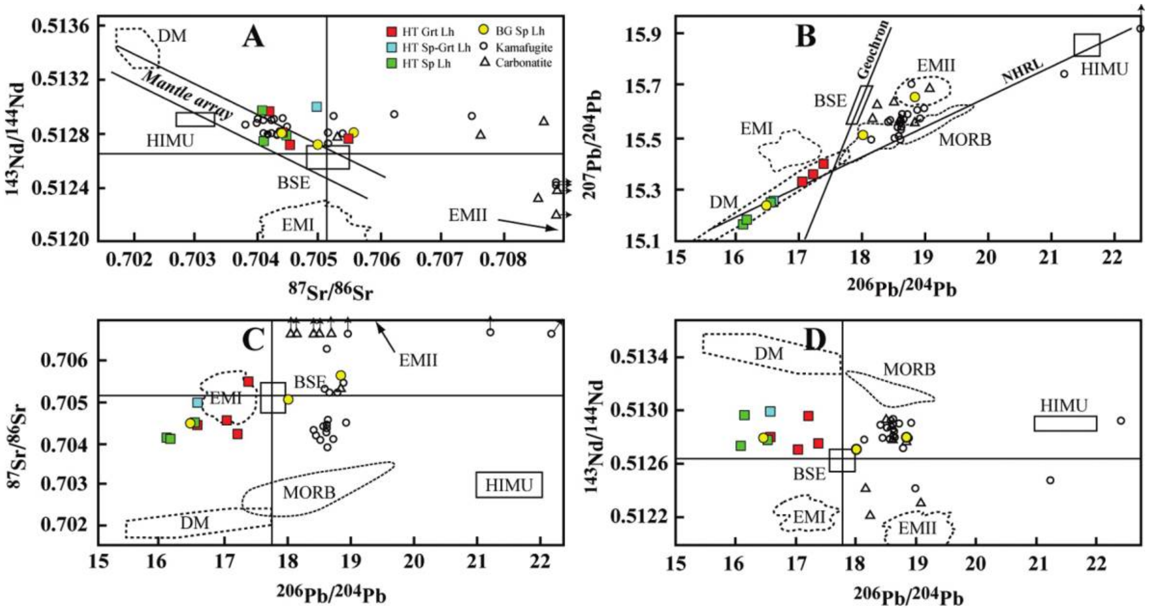

Figure 4. Sr-Nd-Pb isotope correlation diagrams of kamafugites, carbonatites, and peridotite xenoliths from the Haoti (HT) and Baiguan (BG) localities in the Western Qinling. Kamafugite data are compiled from Yu and Zhang (1998), Stoppa et al. (2003), Yu et al. (2004), and Mo et al. (2006). Carbonatite data are compiled from Yu et al. (2004). The mantle reservoirs of Zindler and Hart (1986) are plotted as follows: BSE = bulk silicate earth, DM = depleted mantle, EMI and EMII = enriched mantle, HIMU = mantle with high U/Pb ratio, MORB = mid-ocean ridge basalts. The Pb isotopic values of BSE and the Northern Hemisphere reference line (NHRL) are from Allegre et al. (1988). Grt Lh = garnet lherzolite, Sp-Grt Lh = spinel-garnet lherzolite, Sp Lh = spinel lherzolite.

and Ti), and HREE are relatively compatible (Dalou et al. 2009). The geochemical signature of deep carbonatitic melt is consistent with the flat HREE pattern of carbonatites and is usually characterized by Rb depletion and a high Zr/Hf ratio (Hoernle et al. 2002; Dalou et al. 2009). Carbonatite metasomatism would, therefore, result in a high La_N/Yb_N ratio and relative depletions of Ti and Hf with respect to REE and Ba (Yaxley et al. 1991; Rudnick et al. 1993; Laurora et al. 2001; Hoernle et al. 2002; Neumann et al. 2002).

The mantle xenoliths from the Western Qinling are characterized by LREE enrichment, flat HREE, high La_N/Yb_N and Zr/Hf ratios, high Nb and Ta concentrations, positive Sr, and remarkable Ba and slight Y anomalies (table A1; fig. 3), which clearly match the typical features described above. The overall shapes of the REE and trace element patterns of the xenoliths are nearly identical to those of the carbonatites from the Western Qinling, despite some subtle differences in elements such as U, Nb, and Ta (fig. 3). The Sr and Pb isotopic plots of these xenoliths demonstrate a mixing trend between depleted mantle and enriched mantle type II (EMII) end members (fig. 4*B*, 4*C*); a similar trend for the carbonatites have been documented in some studies (Yu and Zhang 1998; Yu et al. 2004; Dong et al. 2008). The above lines of evidence provide further support for our hypothesis that the lithospheric mantle beneath the Western Qinling has experienced extensive carbonatite metasomatism (Su et al. 2010*a*, 2010*b*).

Different subgroups of peridotites examined in this study display distinct geochemical features. The spinel-facies lherzolite group, which represents the shallow lithospheric mantle, has lower trace element concentrations but higher La_N/Yb_N ratios, slightly positive or even negative Ba anomalies (table A1; fig. 3), and more depleted Sr-Nd-Pb isotopic compositions that plot far away from the carbonatite field in figure 4, indicating low-degree or incipient metasomatism. The garnet lherzolite group, representing the deeper lithospheric mantle, has higher trace element concentrations than the spinel lherzolites and shows close similarities in trace element patterns and Sr-Nd-Pb isotopic chemistry to the Western Qinling carbonatites (figs. 3, 4), suggesting that they were metasomatized to a higher degree than the spinel lherzolites. The coexisting spinel-garnet sample occurring in the spinel-garnet transition zone (Su et al. 2007, 2010*b*) is geochemically intermediate between the overlying and underlying peridotites (figs. 3, 4). The relatively higher trace element concentrations and larger variations—in particular, Pb and Sr isotopes—of the carbonate-bearing lherzolites (table A1; fig. 4) are probably induced by the presence of variable amounts of carbonate minerals. Collectively, the geochemical data on the Western Qinling peridotites suggest an upwelling carbonatite metasomatism within the lithospheric mantle (fig. 5).

Subduction Components in Carbonatite Melts. As mentioned above, negative HFSE (Nb, Ta, Zr, Hf, and Ti) anomalies have been widely observed in carbonatite-metasomatized peridotites and their constituent clinopyroxenes (e.g., O'Reilly and Griffin 1988; Yaxley et al. 1991; Rudnick et al. 1993; Ionov 2001; Hoernle et al. 2002; Rosatelli et al. 2007), but this signature has also been debated by some investigators. On the basis of the experimental mineral-melt partition coefficients, Sweeney et al. (1992) pointed out that mantle affected by carbonatite would show markedly high concentrations of Na, Nb, Ta, and Sr. Furthermore, Veksler et al. (1998), Laurora et al. (2001), and Dalou et al. (2009) observed positive Nb, Ta, Zr, and Hf anomalies in some carbonatite-metasomatized peridotites. Interestingly, a similar phenomenon has also been recorded in clinopyroxenes (Su et al. 2010*b*, 2011*a*) and in whole-rock chemistry (fig. 3) of the Western Qinling peridotite xenoliths.

The inconsistencies mentioned above provide useful information to investigate the components of carbonatite melts. Comprehensive compilation and examination reveal that carbonatite melts could exhibit dramatically wide compositional variations (e.g., Harmer and Gittins 1998; Harmer et al. 1998; Laurora et al. 2001; Ying et al. 2004; Walter et al. 2008; Dalou et al. 2009), identical to the data on the trace elements and isotopes of the Western Qinling carbonatites (figs. 3, 4; Yu and Zhang 1998; Yu et al. 2004), which are generally attributed to the involvement of subducted materials. Many authors have considered the high ^{87}Sr and Pb isotopic values for carbonatites to be inherited from subduction-related melts and thus correlated the source of these carbonatites to mixtures between EMI mantle reservoir and HIMU (mantle with high U/Pb ratio) component or the EMII reservoir (Harmer et al. 1998; Downes et al. 2002; Yu et al. 2004; Rosatelli et al. 2007; Walter et al. 2008). An important process of recycling CO_2 into the deep mantle is the subduction of carbonate component within the basaltic crust of the subducted slabs and those in marine sediments (Zhang and Liou 1994; Gasparik and Litvin 2002; Santosh and Omori 2008*a*, 2008*b*; Maruyama et al. 2009). Furthermore, marine sediments are typically enriched in LREE, Gd, and Y (Zhang and Nozaki 1996; Frimmel 2009), and these enriched signatures are reflected in the carbonatite-metasomatized peridotites from the Western Qinling (fig. 3). Although the HREE and HFSE could be retained in minerals (such as garnet and rutile, respectively) during subduction and slab-dehydration processes (e.g., Walter et al. 2008), Laurora et al. (2001) suggested that, in addition to REE, slab-derived fluid/melt can also transport significant amounts of HFSE, since their solubility will increase at increasing pressure. Thus,

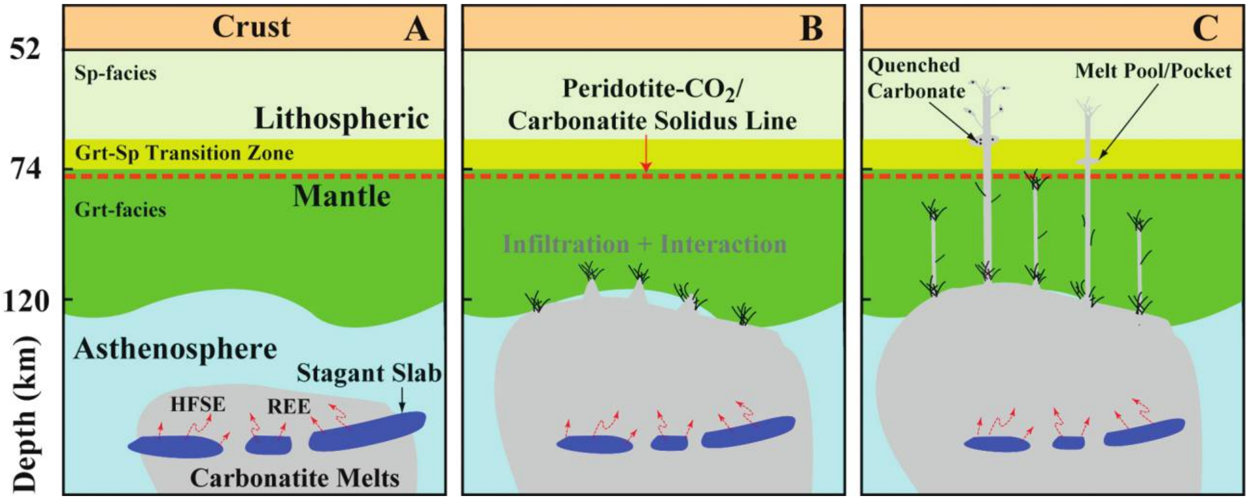

Figure 5. Schematic model for carbonatite metasomatism in the lithospheric mantle. The lithospheric structure is modified from Su et al. (2010*b*) on the basis of *PT* estimates of mantle xenolith from the Western Qinling. The peridotite-CO_2/carbonatite solidus line is after Harmer et al. (1998) and Harmer and Gittins (1998). See text for details. Grt = garnet, HFSE = high-field strength elements, REE = rare earth elements, Sp = spinel.

the HFSE anomalies observed in these xenoliths are considered to be either a slab-inherited feature or acquired during fluid/melt percolation (Gasparik and Litvin 2002). The chemical and carbon isotopic compositions of volatiles of the Western Qinling kamafugites investigated by Tang et al. (2012) reflect a recycled crustal component derived from the devolatilization of subducted oceanic plate or sedimentary rocks. They further inferred that the subducted materials could be related to the Paleo-Tethyan oceanic plate or the Northern China plate, with Yangtze plate subduction and collision under the system of India-Asia collision (Tang et al. 2012). Taking the geological setting into account, the subducted materials, which were exhumed in the Dabie Mountains, were most likely stagnated in the deep mantle beneath the Western Qinling. Although subduction and collision occurred in this region during the Paleozoic, the chemical imprint of subducted slab can possibly be preserved for billions of years (Walter et al. 2008), and these subducted materials could be partly incorporated into upward-migrating carbonatite melts that subsequently react with lithospheric mantle later (fig. 5).

Model for Carbonatite Metasomatism. The garnet-facies peridotites from the Western Qinling record strong signals of carbonatite metasomatism but do not contain any visible carbonate minerals. On the contrary, the spinel-facies samples, which have undergone only weak metasomatism, contain a considerable amount of carbonate veins and discrete grains (Su et al. 2009, 2010*a*, 2010*b*). Globally, carbonate minerals in mantle xenoliths reported to date have been found only in spinel-facies peridotites, such as in the case of In Teria in Algerian Sahara (Dautria et al. 1992), southern Patagonia (Gorring and Kay 2000; Laurora et al. 2001), Tenerife in the Canary Islands (Neumann et al. 2002), Mount Vulture in southern Italy (Rosatelli et al. 2007), and Fangcheng in northern China (Zhang et al. 2010). These uncommon records are probably correlated with the physicochemical properties of carbonatite melts and the related metasomatic processes.

Since carbonatite melts usually have low densities compared with their wall rock, they tend to migrate upward fast from deep mantle (Hunter and McKenzies 1989; Hammouda and Laporte 2000; Gasparik and Litvin 2002). Once the carbonatite melts encounter a stagnant slab, they would probably heat the overlying slab and then trap typical slab-released elements, such as Na, REE, and HFSE (fig. 5*A*; Veksler et al. 1998; Laurora et al. 2001; Walter et al. 2008; Dalou et al. 2009). When the melts rise up to the base of the rigid lithospheric mantle, they would infiltrate upward into the lithosphere because of their low viscosity and great wetting properties (Hunter and McKenzies 1989; Hammouda and Laporte 2000), and the contrasting chemical compositions would trigger subsequent interaction with the lithospheric mantle (fig. 5*B*). The resulting metasomatism, taking place over long periods of time, could transfer the chemical signatures of the carbonatite melts to the whole-scale shallow lithospheric mantle.

Experimental studies have constrained the solidus between carbonatite melt and wehrlite at a pressure of <2.5 GPa (Dalton and Wood 1993; Sweeney 1994). Harmer's study further constrained that the peridotite-CO_2 or carbonatite solidus is 75 ± 5 km at depth, where the carbonatite melts are likely to be retained (Harmer and Gittins 1998; Harmer et al. 1998). Although the upward migration of the carbonatite would continue because of the density contrast between the melt and the surrounding solid mantle (Gasparik and Litvin 2002), they should preferentially quench to carbonate crystals at shallow depth (≤75 km) to form melt pools or pockets (fig. 5*C*); thus, the shallow lithospheric mantle could be only weakly metasomatized. This inferred model is demonstrated by the observations in the Western Qinling. The spinel-garnet transition zone is located at ca. 74 km (Su et al. 2007, 2010*b*), and above this zone carbonate-bearing pockets and veins are widely observed in weakly metasomatized spinel-facies peridotites (Su et al. 2010*a*, 2010*b*).

Conclusions

Trace elemental and Sr-Nd-Pb isotopic data on the peridotite xenoliths from the Western Qinling in central China provide important constraints on the carbonatite metasomatism of the lithospheric mantle. The geochemical variations among the different peridotite subgroups reveal that the degree of metasomatism followed an upwelling front, with the garnet-facies lherzolite xenoliths, representing deeper lithospheric mantle, showing a high degree of metasomatism and the spinel-facies samples, from shallower levels of the lithospheric mantle, displaying low-degree-metasomatized features.

The positive HFSE and some typical trace element anomalies, together with the $^{87}Sr/^{86}Sr$ ratios and linear variations in Pb isotopes of the peridotite xenoliths, could be ascribed to the involvement of a subduction component in the carbonatite melts. Combining the comprehensive data from carbonatite melts, a model for carbonatite metasomatism is proposed to interpret the different signatures recorded in the garnet-facies peridotites (chemical imprint) and the spinel-facies peridotites (occurrence of carbonate minerals).

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