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Key Points:

- Compositional variations in potassic basalts are related to lithospheric thickness
- Melt-rock interaction occurred in the lithospheric mantle and phlogopite precipitated
- The magma lost K and Rb during melt-rock interaction

Supporting Information:

- Supporting Information S1
- Data set S1

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Lithospheric thickness controlled compositional variations in potassic basalts of Northeast China by melt-rock interactions

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Abstract Melt-rock interaction is a common mantle process; however, it remains unclear how this process affects the composition of potassic basalt. Here we present a case study to highlight the link between compositional variations in the potassic basalts and melt-rock interaction in cold lithosphere. Cenozoic potassic basalts in Northeast China are strongly enriched in incompatible elements and show EM1-type Sr–Nd–Pb isotopes, suggesting an enriched mantle source. These rocks show good correlations between $^{87}Sr/^{86}Sr$ and K_2O/Na_2O and Rb/Nb. Notably, these ratios decrease with increasing lithospheric thickness, which may reflect melt-lithosphere interaction. Phlogopite precipitated when potassic melts passed through the lithospheric mantle, and K and Rb contents of the residual melts decreased over time. The thicker the lithosphere, the greater the loss of K and Rb from the magma. Therefore, the compositions of potassic basalts were controlled by both their enriched sources and reactions with lithospheric mantle.

1. Melt-Rock Interaction in the Upper Mantle

Evidence of melt-rock interaction is ubiquitous in orogenic massifs [*Bodinier et al.*, 1990; *Malaviarachchi et al.*, 2010; *Marchesi et al.*, 2013], ophiolites [*Abily and Ceuleneer*, 2013; *Rampone et al.*, 2008; *Zhou et al.*, 1996] and mantle peridotite xenoliths [*Chen and Zhou*, 2005; *Kelemen et al.*, 1998; *Xu et al.*, 2008; *Zhang et al.*, 2001]. In orogenic massifs and ophiolites, melt-rock reactions commonly result in the consumption of orthopyroxene and the precipitation of olivine when a silica-poor melt migrates upward through peridotite [*Malaviarachchi et al.*, 2010; *Marchesi et al.*, 2013]. Such reactions can explain the genesis of dunite in orogenic massifs [*Bodinier et al.*, 1990] and in ophiolites [*Abily and Ceuleneer*, 2013; *Rampone et al.*, 2008]. However, when a silica-rich melt derived from recycled crustal material ascends into mantle peridotite, melt-rock interactions are characterized by the dissolution of olivine and the precipitation of orthopyroxene. Such reactions have been reported in experimental studies [*Lambart et al.*, 2012; *Mallik and Dasgupta*, 2012; *Mallik and Dasgupta*, 2013; *Mallik and Dasgupta*, 2014; *Mallik et al.*, 2015; *Yaxley and Green*, 1998] and account for the excess orthopyroxene in some harzburgites of the lithospheric mantle beneath ancient cratons [*Kelemen et al.*, 1992; *Kelemen et al.*, 1998; *Zhang et al.*, 2001].

Regardless of the type of reaction, melt-rock interaction not only results in the metasomatism of peridotite in the upper mantle but also modifies the initial composition of ascending melt. The possible genetic links between intraplate magmatism and melt-rock interaction in the mantle have been discussed previously [*Mallik and Dasgupta*, 2012; *Mallik and Dasgupta*, 2013; *Mallik and Dasgupta*, 2014; *Tang et al.*, 2006; *Xu et al.*, 2005; *Zeng et al.*, 2013]. *Xu et al.* [2005] proposed that interaction between asthenosphere-derived melt and lithospheric mantle can explain the genesis of the tholeiitic basalts from Datong, west North China Craton, which have higher SiO_2 and Cr, and lower Al_2O_3 and CaO contents than alkali basalts, and rare earth element (REE) patterns showing a kink at Gd that are thought to reflect the dissolution of orthopyroxene and the precipitation of olivine. *Mallik and Dasgupta* [2012] demonstrated experimentally that reaction between eclogite-derived low-degree andesitic melt and fertile peridotite can transform an initial siliceous melt to basanite, and the reacted melts have major element compositions similar to those of alkalic ocean island basalts (OIBs). Moreover, at a similar andesite:peridotite ratio but in the presence of small amount of dissolved CO_2, the derivative liquid becomes nephelinitic owing to the reactive crystallization to form orthopyroxene at the expense of olivine and clinopyroxene [*Mallik and Dasgupta*, 2013], and the degree of alkalinity or silica undersaturation of the reacted melts increases with increasing CO_2 content in reacting melt [*Mallik and Dasgupta*, 2014]. Recently, experiments proved that interactions between sediment-derived hydrous siliceous melts and peridotite/dunite can generate potassic to ultrapotassic melts [*Mallik et al*., 2015; *Pirard and Hermann*, 2015; *Pirard and Hermann*, 2014], although it is still generally accepted that such potassic/ultrapotassic rocks are derived from metasomatized lithospheric mantle [*Foley*, 1992; *Foley et al*., 1987; *Prelevic et al*., 2012; *Prelević et al*., 2008; *Condamine and Médard*, 2014]. However, no previous study has investigated the link between lithospheric thickness and the compositional variations in intraplate potassic (and ultrapotassic) rocks. In this study, we show that compositional variations in Cenozoic potassic basalts from Northeast China are related to the thickness of underlying lithosphere. We propose that this relation reflects melt-rock interaction between potassium-rich melt derived from recycled crust and depleted lithospheric mantle.

2. Cenozoic Potassic Volcanism in Northeast China

Cenozoic continental intraplate volcanic rocks are widespread in Northeast China [*Chen et al*., 2007; *Liu et al*., 2001], covering an area of ~50,000 km^2 (Figure 1b). These lavas occur mainly along the flanks of the Songliao Basin; e.g., the Greater Khingan Range to the west, the Lesser Khingan Range to the north, and the Changbai Mountains to the east [*Liu et al*., 2001]. Tectonically, the Greater Khingan Range and the Songliao Basin belong to the Xing'an-Mongolia Orogenic Belt (XMOB) (Figure 1a), the eastern part of the Paleozoic Central Asian Orogenic Belt, which formed during subduction and collision between the North China block to the south and the Siberian plate to the north [*Jahn et al*., 2000; *Sengör et al*., 1993]. Subduction of the paleo-Pacific plate resulted in a change in the tectonic regime beneath the XMOB from compression to extension during the late Mesozoic, which in turn resulted in asthenospheric upwelling, magma underplating, and the formation of rift basins [*Meng*, 2003; *Wu et al*., 2002; *Zorin*, 1999]. *P* wave and *S* wave receiver function analytical results suggest that the crust and the lithosphere beneath the Songliao Basin are obviously thinner than those beneath the two flanks of the basin [*Guo et al*., 2014; *Tao et al*., 2014; *Zhang et al*., 2014]. Analytical results from receiver function data suggest that the Songliao Basin has a relative thin crust (~31 km), while the Moho beneath the Greater Khingan Range can be as deep as 42 km [*Tao et al*., 2014]. Recent analysis of *S* wave receiver functions from a dense seismic array has revealed that the lithosphere thickness beneath the Songliao Basin is about 100–120 km, thinner than those beneath the Greater Khingan Range (140–160 km) and the Changbai mountain (120–140 km) [*Zhang et al*., 2014], which is consistent with previous analysis from Bouguer gravity data [*Ma*, 1987].

Five potassium-rich volcanic fields (Xiaogulihe, Nuominhe, Keluo, Wudalianchi, and Erkeshan; [*Zhang et al*., 1998]), as classic intracontinental monogenetic volcanic fields in the world, are located at the boundary between the northwestern margin of the Songliao Basin and the Greater Khingan Range (Figure 1b). These rocks cover an area of >3000 km^2 and can be subdivided into three main eruptive episodes: middle–late Miocene (16.5–7.0 Ma), late Pliocene to Pleistocene (2.3–0.13 Ma), and recent (up to 1721 A.D.) [*Zhang et al*., 1998]. The Nuominhe volcanic field is located in the northern Greater Khingan Range, where *S* wave receiver function images indicate a thick lithosphere (~150 km; Figure 1b) compared with that beneath the Keluo, Wudalianchi, and Erkeshan fields at the margin of the Songliao Basin [*Zhang et al*., 2014]. The Erkeshan volcanic field is the field closest to the center of the Songliao Basin, and it is underlain by the thinnest lithosphere. Toward the margin of the basin, the thickness of the underlying lithosphere increases from the Erkeshan field to the Keluo field, and the lithosphere beneath the Wudalianchi volcanic field has an intermediate thickness of ~120 km (Figure 1b) [*Ma*, 1987]. In summary, the relative thicknesses of lithosphere beneath the four potassic volcanic fields in Northeast China are as follows: Nuominhe $>$ Keluo $>$ Wudalianchi $>$ Erkeshan. The thickness of the lithosphere beneath Xiaogulihe is unknown due to a lack of detailed geophysical data.

3. Compositional Variations in Cenozoic Potassic Basalts of Northeast China

Cenozoic volcanic rocks from Northeast China are dominantly alkali basalts (Figure S1a in the supporting information) that can be divided into ultrapotassic series (e.g., Xiaogulihe; $K_2O/Na_2O > 2$), potassic series (e.g., Nuominhe, Keluo, Wudalianchi, and Erkeshan; $1 < K_2O/Na_2O < 2$), and sodic series (e.g., Halaha-Chaihe; $K_2O/Na_2O < 1$) (Figure S1b) [*Chu et al*., 2013; *Ho et al*., 2013; *Sun et al*., 2014; *Zhang et al*., 1995] according to the scheme of *Foley et al*. [1987]. Here we compiled geochemical data of potassic and ultrpotassic basalts from Northeast China (Data set S1). The potassic (and ultrapotassic) volcanic rocks have notably lower Al_2O_3 contents and higher K_2O contents (Figures S1c and S1d) for a given MgO content than does the sodic rocks from Northeast China [*Chu et al*., 2013; *Sun et al*., 2014]. In a primitive-mantle-normalized incompatible

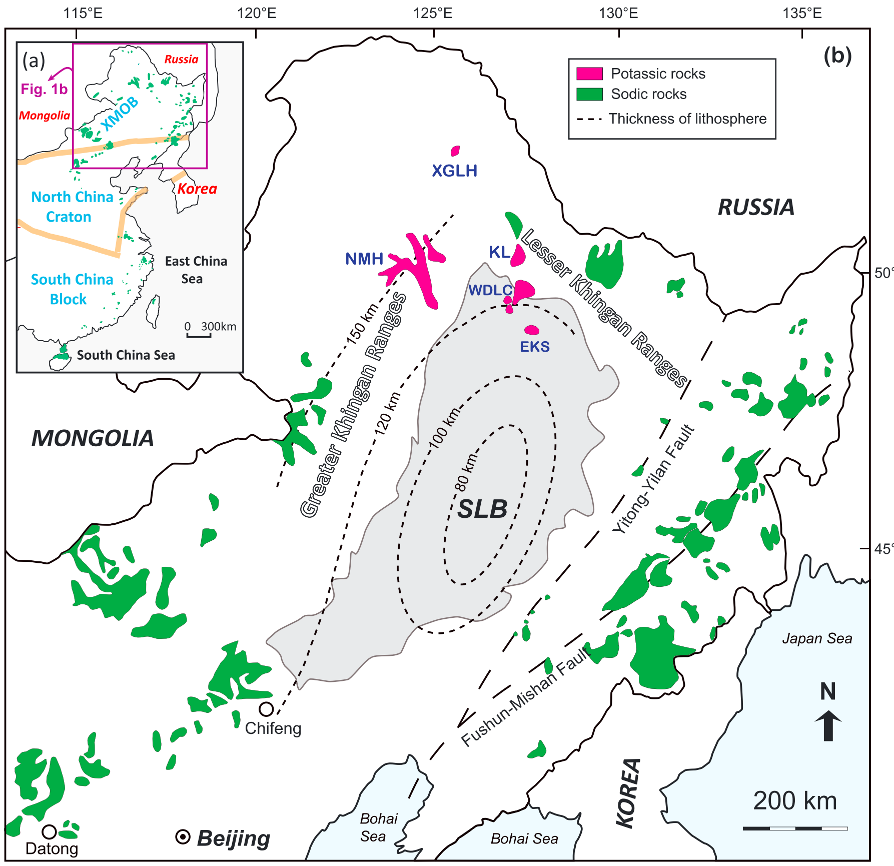

Figure 1. (a) Simplified geological map shows the spatial distribution of tectonic plates and Cenozoic volcanic rocks in eastern China. (b) Distribution of Cenozoic volcanic rocks in Northeast China [*Liu et al.*, 2001]. The contours represent lithospheric thickness beneath the Songliao Basin and Greater Khingan Range [*Ma*, 1987]. Abbreviations are as follows: XMOB, Xing'an-Mongolia Orogenic Belt; SLB, Songliao Basin; XGLH, Xiaogulihe; NMH, Nuominhe; KL, Keluo; WDLC, Wudalianchi; and EKS, Erkeshan.

element diagram (Figure S2), these potassic (and ultrapotassic) basalts are enriched in light rare earth elements and large ion lithophile elements but depleted in high field strength elements, with positive Ba, K, and negative Nb–Ta, Th–U, and Ti anomalies. They have extremely low radiogenic $^{206}Pb/^{204}Pb$ (16.44–17.23), moderate $^{87}Sr/^{86}Sr$ (0.70466–0.70572), and low ε_{Nd} (-6.3 to -0.8) toward the EM1 end-member (Figures 2a and S3) [*Chu et al.*, 2013; *Sun et al.*, 2014; *Zhao et al.*, 2014]. Thus, we refer to these potassic (and ultrapotassic) basalts from Northeast China as EM1-type volcanic rocks (Figure S1).

The potassic rocks show positive correlations of K_2O/Na_2O and Rb/Nb versus $^{87}Sr/^{86}Sr$ (Figure 2). For a given $^{87}Sr/^{86}Sr$ value, the Xiaogulihe ultrapotassic basalts have elevated K_2O/Na_2O and Rb/Nb ratios and deviate from the trends shown by potassic basalts. Furthermore, these compositional variations in potassic basalts show a direct correspondence to the thickness of the underlying lithosphere. For the basalts erupted over a transect from the Erkeshan to Nuominhe fields, the average Sr–Nd isotopic composition becomes less enriched, while MgO increases and K_2O/Na_2O and Rb/Nb decrease with increasing lithospheric thickness (Figure 3).

4. Melt-Rock Interaction and Compositional Evolution of Potassic Melts

Cenozoic potassic (and ultrapotassic) basalts in Northeast China show enriched incompatible trace element signatures (Figure S2) and EM1-type Sr–Nd–Pb isotopic compositions (Figure S3). It is still debated whether the sources of these basalts were located in the lithospheric mantle [*Chu et al.*, 2013; *Sun et al.*, 2014; *Sun et al.*, 2015; *Zhang et al.*, 1995; *Zou et al.*, 2003] or asthenospheric mantle [*Choi et al.*, 2006; *Kuritani et al.*, 2013] and whether the enriched components originated from recycled sediments [*Kuritani et al.*, 2013; *Sun et al.*, 2014; *Sun et al.*, 2015] or lower continental crust [*Chu et al.*, 2013; *Zhao et al.*, 2014]. However, previous

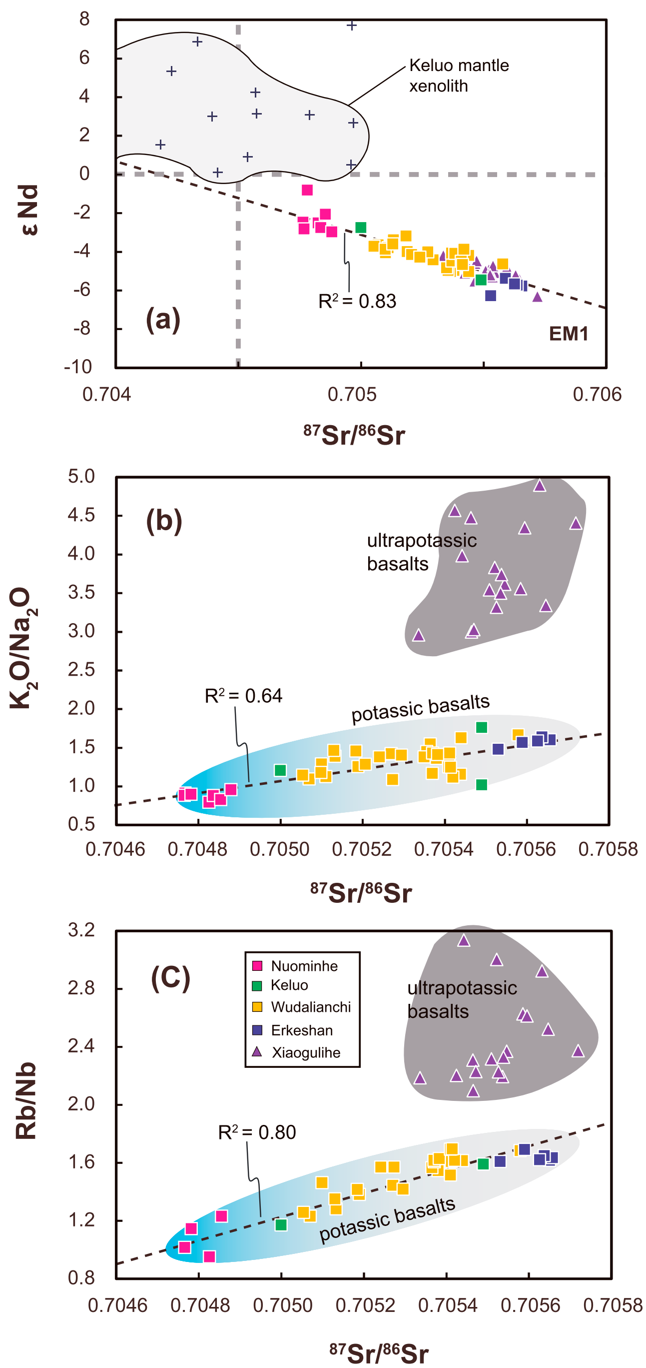

Figure 2. $^{87}Sr/^{86}Sr$ versus (a) ε_{Nd}, (b) K_2O/Na_2O, and (c) Rb/Nb for the Cenozoic potassic (ultrapotassic) rocks from the Northeast China. The Xiaogulihe ultrapotassic basalts display elevated K_2O/Na_2O and Rb/Nb at a given $^{87}Sr/^{86}Sr$. Literature data for Nuominhe basalts is from *Zhao et al.* [2014], Wudalianchi, Erkeshan, and Keluo potassic basalts is from *Basu et al.* [1991], *Chen et al.* [2007], *Chu et al.* [2013], *Fan and Hooper* [1991], *Hsu and Chen* [1998], *Kuritani et al.* [2013], *Liu et al.* [1994], *Zhang et al.* [1995], and *Zou et al.* [2003], and Xiaogulihe ultrapotassic basalts is from *Sun et al.* [2014]. The literature data of Keluo mantle xenoliths is from *Zhang et al.* [2011].

studies have not considered the relation between the geochemistry of the basalts and the thickness of underlying lithosphere. Thus, the aim of this paper is not to identify the origin of the enriched material but to clarify the process responsible for the compositional variations in the potassic basalts.

With increasing lithospheric thickness, the Sr–Nd isotopic composition becomes less enriched, MgO content increases, and K_2O/Na_2O and Rb/Nb decrease from the Erkeshan to Nuominhe potassic basalts, suggesting the influence of lithospheric mantle on basalt geochemistry (Figure 3). These observations indicate that the lithosphere influenced the potassic melts but were not their direct source. Until now, the "lid effect" has been invoked to explain the correlation between lithospheric thickness and compositional variations in ocean island basalts (OIBs) [*Humphreys and Niu*, 2009; *Niu et al.*, 2011] and in continental volcanic rocks [*Davies et al.*, 2015]. For convecting mantle-derived basalts, lavas that erupt onto thicker lithosphere should have a lower average degree of partial melting, thus containing higher $(La/Yb)_N$ and more enriched radiogenic Sr–Nd–Hf isotopic compositions (i.e., less diluted) than those erupted onto thinner lithosphere [*Niu et al.*, 2011]. However, the opposite is observed in the potassic basalts from Northeast China. The Nuominhe basalts, which have the thickest underlying lithosphere, have the least enriched radiogenic isotopic ratios (Figure 2a). Since Yb is compatible in garnet, whereas La is incompatible, the $(La/Yb)_N$ ratio will be elevated under a lower degree of melting corresponding to thicker lithospheric mantle. However, in the potassic basalts the average $(La/Yb)_N$ (Figure S4) decreases with increasing lithospheric thickness, which is opposite to the trend predicted by the lid effect.

Lithospheric mantle not only controls the final melting depth and pressure of upwelling mantle material [*Davies et al.*, 2015; *Humphreys and Niu*, 2009; *Niu et al.*, 2011] but also contaminates the ascending melt through melt-rock

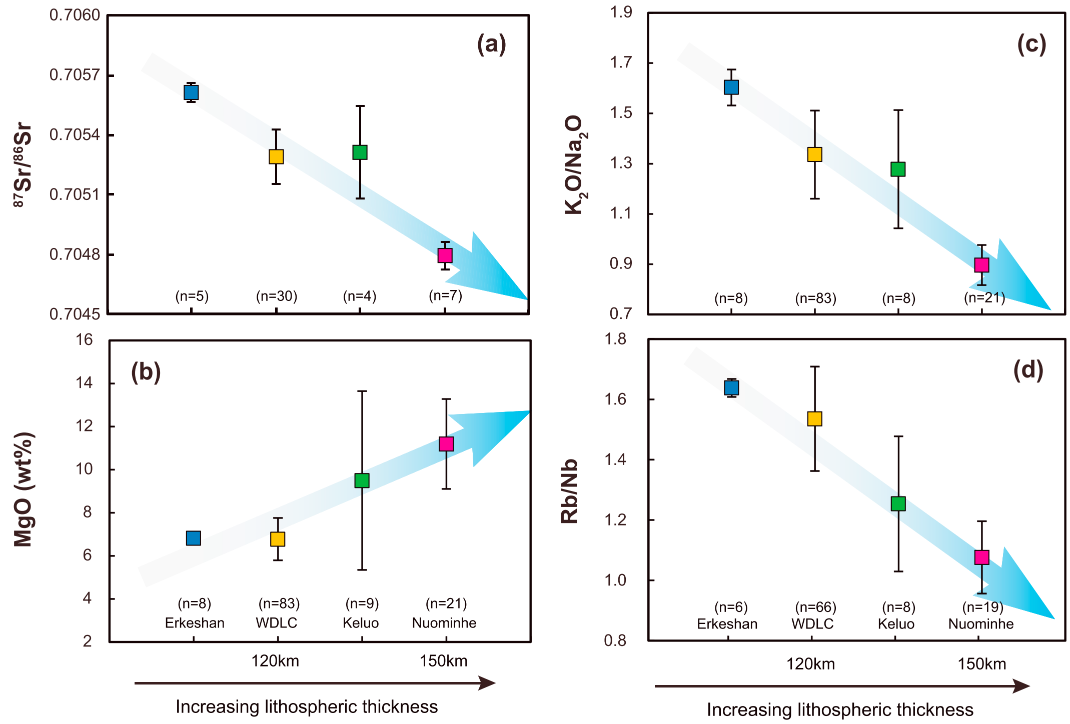

Figure 3. Average (a) $^{87}Sr/^{86}Sr$, (b) K_2O/Na_2O, and (d) Rb/Nb decrease, while (c) MgO increases with increasing lithospheric thickness for the Cenozoic potassic basalts from Northeast China. N represents the number of samples averaged for geochemical compositions and the error bars correspond to 1 standard error (1 SE) of the mean.

interaction due to compositional disequilibria [*Xu et al.*, 2005; *Tang et al.*, 2006]. In the experiments of *Mallik and Dasgupta* [2012], interaction between an initial andesitic melt and peridotite led to the consumption of olivine and precipitation of orthopyroxene and garnet in the peridotite, which increased the MgO content and decreased the SiO_2, Al_2O_3, and K_2O contents in the reacted melt. The compositional variations in the reacted melt depended mainly on the duration and extent of melt-rock reaction, which are related to the thickness of lithosphere through which the melt ascends (i.e., a thicker lithospheric mantle corresponds to a longer duration and greater extent of melt-rock interaction). Northeast China, as the eastern part of the Phaneozoic Central Asian Orogenic Belt, has a juvenile lithospheric mantle with depleted Sr–Nd isotopes (low $^{87}Sr/^{86}Sr$ ratio, 0.70364–0.70627; high ε_{Nd} values, 0.10–7.71; [*Zhang et al.*, 2011]) (Figure 2a). The effects of melt-rock interactions in the lithospheric mantle mean that basaltic melt erupted onto thicker lithosphere will have higher MgO contents and less enrichment in Sr–Nd isotopic compositions (descendant $^{87}Sr/^{86}Sr$ ratio and elevated ε_{Nd} value), which is consistent with the trends observed in the Cenozoic potassic basalts of Northeast China (Figures 3a and 3b).

The occurrence of melt-rock interactions in the lithospheric mantle beneath Northeast China is also evidenced by the mineralogical and petrological characteristics of mantle xenoliths in the potassic basalts. The occurrence of veined (Figure S5a) or intergranular (Figure S5b) phlogopite as the main metasomatic phase in the mantle xenoliths provides direct evidence for melt-rock reactions in the lithospheric mantle beneath Northeast China [*Zhang et al.*, 2011; *Sui et al.*, 2014]. Phlogopites in the Nuominhe garnet peridotite have high K_2O (8.42–10.14 wt %) and TiO_2 (5.41–7.79 wt %) contents [*Sui et al.*, 2014], indicating metasomatism by a potassium-rich melt. Because K and Rb are compatible in phlogopite but Na and Nb are incompatible, the precipitation of phlogopite in the peridotite would have reduced the K_2O/Na_2O and Rb/Nb ratios of the reacted melts. The precipitation of phlogopite means that a longer duration of melt-rock reaction under thicker lithosphere would consume more K and Rb and lead to lower K_2O/Na_2O and Rb/Nb ratios in the residual melts, which is consistent with the variations in elemental ratios of the potassic basalts in Northeast China (Figures 3c and 3d). Furthermore, the clinopyroxenes in the Keluo mantle xenoliths have sinusoidal-shaped chondrite-normalized REE patterns characterized by MREE enrichment [*Zhang et al.*, 2000], and the garnets in the Nuominhe harzburgite have anomalously high CaO contents (5%–7%) [*Sui et al.*, 2012], also indicating interaction between enriched siliceous melts and depleted mantle peridotite.

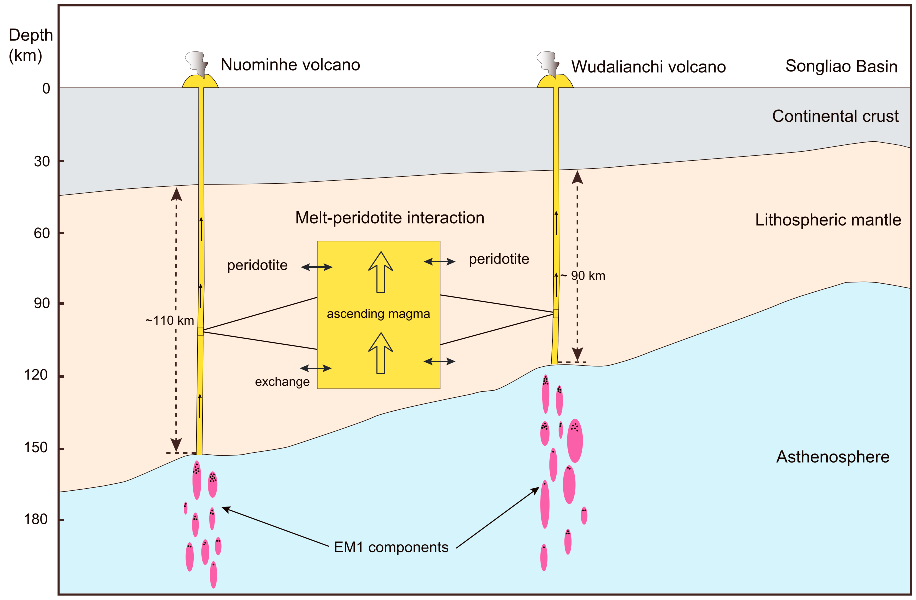

Figure 4. Simplified cartoon describing the formation process of Cenozoic potassic basalts from Northeast China. Primary melt characterized by EM1-type isotopic compositions was generated in the upmost asthenosphere. Melt-peridotite reaction occurred when this melt ascended through the lithospheric mantle before eruption. This reaction would consume olivine and precipitate orthopyroxene and phlogopite, leading to elevation of MgO contents and reduction of K_2O/Na_2O and Rb/Nb ratios of the reacted melts.

The Xiaogulihe ultrapotassic basalts deviate from the trends defined by the potassic basalts, with higher K_2O and lower Al_2O_3 contents for a given MgO content (Figures S1c and S1d), and higher K_2O/Na_2O and Rb/Nb for a given $^{87}Sr/^{86}Sr$ (Figures 2b and 2c). Such compositional deviations can be explained by compositional differences of their initial melts. The ultrapotassic basalts may have no K-rich phase, e.g., phengite in the source (dry recycled crustal material?) during low-degree melting, and generate a kind of ultrapotassic initial siliceous melts with higher K_2O content, and higher K_2O/Na_2O and Rb/Nb ratios than those of initial melts of potassic basalts. The estimated H_2O content (0.36–0.50 wt %) and H_2O/Ce ratio (~15) of the Xiaogulihe primary magma are clearly lower than those of melts from normal depleted mantle [*Chen et al*., 2015], which supports above proposal.

Figure 4 shows the formation process of the potassic basalts in Northeast China. Silica- and potassium-rich melts were generated near or below the lithosphere-asthenosphere boundary from EM1-type components (recycled ancient sediment or lower continental crust, or both). When these enriched melts ascended through the cold continental lithosphere, they reacted with the mantle peridotite owing to compositional disequilibrium. This process caused the consumption of olivine and the precipitation of orthopyroxene and garnet, which increased the MgO content and decreased the SiO_2 and Al_2O_3 contents of the reacted melt. Melt-rock reaction resulted in the formation of potassium-rich metasomatic minerals in mantle xenoliths, (e.g., phlogopite), which in turn lowered the K_2O/Na_2O and Rb/Nb ratios of the initial melt. The Nuominhe basalts, which erupted onto thicker lithosphere, experienced longer duration and greater extent of interaction with depleted mantle peridotite and have less enriched Sr–Nd isotopes, higher MgO contents, and lower K_2O/Na_2O and Rb/Nb ratios than the Wudalianchi potassic basalts that were erupted onto thinner lithosphere. This model leads to the question of why the basaltic melt records interaction with lithospheric mantle rather than with the continental crust when ascending. In explanation, melt-peridotite interaction increases MgO and decreases SiO_2 contents in melts and thereby reduces melt viscosity. Thus, the reacted melt would have ascended rapidly through the continental crust and therefore avoided significant crustal contamination.

5. Conclusions

We summarized geochemical data of Cenozoic potassic basalts from Northeast China to investigate how melt-rock interactions modified their composition and to explain the observed compositional variations. These basalts have EM1-type elemental and isotopic compositions and were probably derived from recycled sediments or lower continental crust in the convecting mantle. The ε_{Nd} values, and K_2O/Na_2O and Rb/Nb ratios of the potassic basalts show good correlations with $^{87}Sr/^{86}Sr$ values. Moreover, these elemental and isotopic ratios, together with the MgO contents, are related to the thickness of the underlying lithosphere. We attribute this relation to interactions between a silica- and potassium-rich melt and depleted lithospheric mantle. Such reactions can lead to the consumption of olivine and precipitation of orthopyroxene and phlogopite, which would increase the MgO content and decrease the K_2O/Na_2O and Rb/Nb ratios of the reacted melts.

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