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1   # Source of highly potassic basalts in northeast China: evidence

2   # from Re-Os, Sr-Nd-Hf isotopes and PGE geochemistry

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Abstract

23

24   The origin of the very young (0.5 Ma to 1721 AD) Wudalianchi-Erkeshan

25   (WDLC and EKS) highly potassic basalts in northeast (NE) China has been the

26   subject of considerable debate. In this study, we present new major-, trace- and

27   platinum group element (PGE) data together with Re-Os, Lu-Hf, and Sr-Nd isotopic

28   analyses for these potassic basalts in order to further constrain their source. It has been

29   shown that the WDLC and EKS basalts have unradiogenic to only moderately

30   radiogenic osmium isotope ratios with $^{187}Os/^{188}Os$ ranging from 0.1187 to ~0.17, and

31   only slight PPGE (Pt and Pd) enrichments relative to IPGE (Os, Ir and Ru). The

32   positive correlations between $^{187}Os/^{188}Os$ and 1/Os suggests that these basalts have

33   probably experienced about 2 ~ 8% of lower continental crust addition during magma

34   ascent. Moreover, the incorporation of sub-continental lithospheric mantle

35   (SCLM)-derived, primary sulphides and / or PGE micro-alloys during magma ascent

36   seems likely in some basalts, which preserve particularly unradiogenic Os

37   compositions. The complex interplay of signatures derived from crustal and SCLM

38   contamination means that the Os isotope systematics of the basalts do not

39   unequivocally fingerprint the source of the WDLC and EKS basalts.

40   The strong enrichments of light rare earth elements (LREE) and large ion

41   lithophile elements (LILE), high-K, EM1-like Sr-Nd-Hf isotopic characteristics, and

42   particularly strong fractionation of the heavy REE (($Sm/Yb)_N$ = 9.7 ± 0.6), suggest

43   that the WDLC and EKS basalts mainly originated from phlogopite-bearing

44   garnet-peridotite in the SCLM. Combined with low Ce/Pb ratios (~9.4-13.5), the most

likely source of WDLC and EKS basalts is SCLM that has been metasomatized by delaminated old, lower continental crust. This is different from previous hypotheses that implicate a dominantly asthenospheric source with a contribution from delaminated ancient SCLM or subducted oceanic crust with sediments, or a SCLM source that has been metasomatized by melts derived from deep asthenosphere during the Proterozoic.

1. Introduction

Cenozoic volcanic rocks, predominantly alkaline basalts, are widely distributed in Northeastern China (NE China) (Fig. 1), and form an important part of the West Pacific volcanic zone (e.g., Zhang et al., 1991, 1995, 1998, 2000; Zou et al., 2000, 2003; Liu et al., 2001; Choi et al., 2006; Chen et al., 2007; Yan and Zhao, 2008). However, their petrogenesis remains equivocal, despite numerous detailed prior investigations of the area. Interpreting the significance of the composition of Cenozoic alkaline basalts from NE China is further complicated by the complex and varying structure for the mantle beneath this region, as suggested by earlier studies (e.g., Zhang et al., 2000; Zou et al., 2000, 2003; Choi et al., 2006; Chen et al., 2007; Yan and Zhao, 2008; Zhang et al., 2011).

Among the Cenozoic basalts in NE China, the three potassic volcanic areas of

67   Wudalianchi (WDLC), Erkeshan (EKS) and Keluo (KL), together comprise the

68   Wudalianchi-Erkeshan-Keluo (WEK) volcanic field. This field is located in the

69   Xing'an Mongolian Orogenic Belt (XMOB) at the boundary between the

70   northwestern margin of the Songliao Basin and the Great Xing'an Ranges, in the

71   western Heilongjiang Province, NE China (Fig. 1). The geochemical features of the

72   WDLC, EKS and KL basalts (e.g., Zhang et al., 1991, 1995, 1998; Zou et al., 2003)

73   are similar to those of EM1-derived basalts (Zindler and Hart, 1986) and dramatically

74   different from those of Cenozoic basalts found elsewhere in eastern China (e.g., Zhou

75   and Armstrong, 1982; Peng et al., 1986; Song et al., 1990; Zhi et al., 1990; Basu et al.,

76   1991; Fan and Hooper, 1991; Liu et al., 1994; Chen et al., 2007).

77   Although the peculiar composition of the WEK potassic basalts in NE China has

78   been well documented, the origin of intraplate volcanism in this region is still the

79   subject of debate, resulting in several competing theories as to their origin: (i) Based

80   upon their Sr-Nd-Pb isotopic and trace elemental fingerprint, which resembles an

81   EM1 mantle component, it has been suggested that the WEK potassic basalts were

82   produced by small-degree (5 ~ 7 %) melting of ancient SCLM, which has

83   subsequently been overprinted by Proterozoic metasomatism after its formation

84   (Zhang et al., 1995, 1998, 2000). A phlogopite-bearing garnet peridotite source within

85   the 120 km thick lithosphere at this locality has been suggested as a probable source

86   (Zou et al., 2003); (ii) Based upon the similarities in incompatible trace element

87   concentrations with OIB, Choi et al. (2006) argued that the WEK basalts originated in

88   the shallow asthenosphere which contains fragments of delaminated SCLM from the

89   eastern North China Craton (NCC). The likely presence of "plum-pudding" mantle

90   (e.g., Phipps Morgan and Morgan, 1999) beneath NE China (Flower et al., 1998) and

91   basaltic Sr-Nd-Pb-Hf isotopic characteristics resembling a DMM-EM1 array have

92   been cited as strong evidence against a SCLM origin for the WEK basalts (Choi et al.,

93   2006); (iii) Mixing FOZO (Focal Zone; Hart et al., 1992) and LoMu (low U/Pb;

94   Douglass et al., 1999) mantle sources, instead of an EM1-like SCLM mantle source,

95   has been advocated by Chen et al. (2007); (iv) More recently, Kuritani et al. (2013)

96   argued that the WDLC and EKS basalts originated from a mantle transition zone

97   which has been metasomatized by K-rich sediment-derived fluids ~1.5 Ga ago,

98   through the stagnation of an ancient slab.

99   In addition, although wholesale contamination of the WDLC and EKS basalts

100   by the upper crust can largely be discounted, based on trace elemental and Sr-Nd-Pb

101   isotopic results (Zhang et al., 1995, 1998; Zou et al., 2003), previous studies have not

102   been able to unequivocally exclude the possible influence of less evolved lower crust

103   in the petrogenesis of the WDLC and EKS basalts (Zhang et al., 1995, 1998; Zou et

104   al., 2003). Unlike the lithophile-element based Sr-Nd-Pb isotope systems, Os isotopes

105   can be particularly sensitive to crustal contamination which generates isotope ratios

106   that are distinct from those of mantle sources (e.g., Chesley and Ruiz, 1998; McBride

107   et al., 2001; Chesley et al., 2004; Jung et al., 2011). Basaltic melt, contaminated

108   during its ascent through old continental crust, should develop a radiogenic Os

109   isotopic signature. In contrast, basalts originated from an enriched old lithospheric

110   mantle source would retain unradiogenic to only moderately radiogenic Os isotopic

111 compositions (e.g., McBride et al., 2001; Chesley et al., 2004; Rudnick and Walker,

112 2009). Therefore, Re-Os isotopes, together with PGE elemental abundances can

113 provide important constraints on the relative involvement of crustal materials in the

114 petrogenesis of the very young WDLC and EKS basalts.

115 Accordingly, this study presents new major-, trace- and platinum group element

116 (PGE) abundances together with Os, Hf and Sr-Nd isotopic data, for the WDLC and

117 EKS potassic basalts to further constrain their origin and the nature of the underlying

118 SCLM. Since the nearby Keluo basalts contain abundant mantle xenocrysts, they have

119 not been investigated for Re-Os isotopes and PGE geochemistry here. Based upon

120 new analyses, it seems likely that the WDLC and EKS basalts were mainly generated

121 in SCLM which has been metasomatized by recycled old lower continental crust

122 materials. Although crustal contamination during magma ascent has not materially

123 affected the elemental abundances of incompatible, lithophile elements and the

124 Sr-Nd-Hf isotope systems, evidence for low degrees of lower crustal contamination is

125 preserved in some basalts with particularly low Os abundances.

126

127 **2. Geological Background and sample descriptions**

128 The XMOB, part of the Central Asian Orogenic Belt (CAOB), is a composite

129 fold belt more than 1500 km wide, formed during a long period (Paleozoic to early

130 Mesozoic) of subduction and collision between the Siberian Craton in the north and

131 the NCC in the south (Sengör et al., 1993). The potassic basalts of the WEK field,

132 located in XMOB (Fig. 1), are distributed over an area of >1400 km^2. Among them,

133   the WDLC and EKS potassic basalts were erupted from about 0.5 Ma ago onwards

134   (Zhang et al., 1995; Liu et al., 2001). Two of the Wudalianchi cones, Laoheishan and

135   Huoshaoshan, erupted during 1719-1721 AD, according to the Qing Dynasty Records

136   (Zou et al., 2003). The potassic rocks consist mainly of olivine leucitite and leucite

137   basanite.

138   The samples studied here were collected from lava flows related to the

139   eruptions of Laoheishan (LHS) (1721 AD), Huoshaoshan (HSS) (1721 AD) and

140   Yaoquanshan (YQ) (0.25 Ma, Zhang et al., 1995) from the Wudalianchi volcanic area,

141   and lava flows related to the Erkeshan eruption (0.56Ma, Liu et al., 2001) from the

142   Kedong (KD) volcanic area (Fig.1). The sample locations (latitude and longitude) are

143   listed in Table 1. The detailed descriptions of the samples and petrography of some

144   representative samples are shown in the electronic supplement.

145

146   **3. Analytical methods**

147   The rock samples were first sawn into chips, and surface contamination was

148   completely removed during this step. The samples were then further split into small

149   chips using a hammer wrapped in soft cloth. Small chips devoid of surface alteration

150   and mantle or/and crustal xenocrysts, were chosen and cleaned with 10% hydrochloric

151   acid in an ultrasonic bath for about half an hour. After ultrasonification, the chips were

152   cleaned three times with deionized water. Subsequently, selected rock chips were

153   crushed using an alumina ceramic jaw crusher, and then were ground into a fine

154   powder in agate shatter-boxes. This produced 20-30g of powder for each sample.

155

3.1. Elemental analysis

Major element data for whole-rock samples were obtained by X-ray fluorescence spectrometry (XRF) on fused glass disks using an Axios-Minerals instrument at the Institute of Geology and Geophysics, Chinese Academy of Sciences (IGGCAS), following the procedures of Chu et al. (2009). Precision is 1-3% RSD for elements present at >1 wt%, and about 10% RSD for elements present at <1 wt%. A Chinese basalt reference material, GSR-3, was analyzed during the same period, and the values determined are well within the range of consensus values (GeoREM, http://georem.mpch-mainz.gwdg.de/) (Table 1).

Trace element concentrations, including the rare earth elements (REE), were determined by inductively coupled plasma mass spectrometry (ICP-MS) using an Agilent 7500a system at IGGCAS, following the procedures described in Chu et al. (2009). Basalt reference materials BHVO-2 and BCR-2 were measured to monitor the accuracy of the analytical procedure, and the results are in good agreement with reference values (GeoREM, http://georem.mpch-mainz.gwdg.de/) (Table 1). Precision is generally better than 3% for most elements based on replicate analyses of several samples.

Sulfur concentrations were determined at the National Research Center for Geoanalysis, Chinese Academy of Geological Sciences, using a high-frequency infrared absorption spectrometer (HIR-944B, Wuxi High-speed Analyzer Co., Ltd., China), following the procedures described in Chu et al. (2009). The detection limit

177   for S was about 50 ppm.

178

179   *3.2. Sr-Nd-Hf isotope analyses*

180   Strontium, Nd, and Hf isotope compositions were determined at the State Key

181   Laboratory of Lithospheric Evolution, IGGCAS. A combined chemical procedure for

182   Lu-Hf, Rb-Sr, and Sm-Nd isotopic analyses from one sample digestion was used, as

183   described in detail by Yang et al. (2010). The Rb-Sr and Sm-Nd isotopic analyses

184   were conducted using an Isoprobe-T thermal ionization mass spectrometer made by

185   Isotopx Company (formerly GV instruments). Measured $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$

186   ratios were corrected for mass-fractionation using $^{86}Sr/^{88}Sr = 0.1194$ and $^{146}Nd/^{144}Nd$

187    $= 0.7219$, respectively. During the period of data collection, the measured values for

188   the NBS-987 Sr standard and the JNdi-1 Nd standard were $^{87}Sr/^{86}Sr = 0.710245 \pm 16$

189   (2 SD, n = 8) and $^{143}Nd/^{144}Nd = 0.512117 \pm 10$ (2 SD, n = 8), respectively. Lutetium

190   and Hf were measured using a Thermo-Fisher Neptune multicollector ICP-MS system

191   (Yang et al., 2010). Hafnium isotopic ratios were normalized to $^{179}Hf/^{177}Hf = 0.7325$

192   and $^{176}Lu/^{175}Lu$ isotopic ratios were normalized using Yb isotopic ratios. During the

193   analytical session, an Alfa Hf standard was measured 9 times and the average value of

194    $^{176}Hf/^{177}Hf$ was 0.282181 ± 5 (2 SD). The USGS basalt reference materials BCR-2

195   and BHVO-2 were measured for Rb-Sr, Sm-Nd and Lu-Hf isotopic composition to

196   monitor the accuracy of the analytical procedures; the results are in excellent

197   agreement with the reported reference values (GeoREM,

198   http://georem.mpch-mainz.gwdg.de/) (Table 2). The procedural blanks were about 40

199   pg for Rb, 300 pg for Sr, 20 pg for Sm, 60 pg for Nd, 10 pg for Lu and 40 pg for Hf,

200   respectively.

201

202   *3.3. Re-Os and PGE analyses*

203   Re-Os isotopic compositions and PGE abundances were determined at the State

204   Key Laboratory of Lithospheric Evolution, IGGCAS, following procedures similar to

205   those described by Pearson and Woodland (2000).

206   In brief, about 2 g or 4 g of finely ground sample powder (200-400 mesh), and

207   appropriate amounts of a mixed ^{187}Re-^{190}Os spike and a mixed ^{191}Ir-^{99}Ru-^{194}Pt-^{105}Pd

208   spike were weighed into a clean, dry, chilled Pyrex$^{®}$ borosilicate glass Carius tube.

209   For 2 g samples, 3 mL of purified concentrated HCl and 6 mL of purified

210   concentrated HNO$_3$ were added to the tube, and then the tube was sealed (Shirey and

211   Walker, 1995); for 4 g samples, the amounts of these acids were scaled up accordingly.

212   The Carius tubes were put in a steel pressure vessel containing water to prevent the

213   explosion of the Carius tubes, as described in detail by Qi et al. (2007). The digestions

214   were performed at about 250 $^{\circ}$C for 48-72 h in an oven. After opening the tubes, Os

215   was extracted from the aqua regia solution into CCl$_4$ (Cohen and Waters, 1996) and

216   then back-extracted into 4 mL of HBr; for 4 g samples, the amounts of CCl$_4$ and HBr

217   used were also scaled up. The Os samples were further purified via microdistillation

218   (Birck et al., 1997). The total procedural Os blank was 0.2-1 pg with a ^{187}Os/^{188}Os of

219   about 0.16. The blank Os contribution on total Os was less than 1% for all samples

220   and the blank correction was therefore negligible.

221   Rhenium, Ir, Ru, Pt and Pd remaining in aqua regia were separated from the

222   matrix and purified first by anion exchange chromatography using 2 mL of AG 1 × 8,

223   100 - 200 mesh resin. The Re and Ru fraction was further purified using a small anion

224   exchange column packed with 0.2 mL resin (AG 1 × 8, 100-200 mesh), whereas the

225   Ir-Pt and Pd fractions were further purified to remove Zr and Hf using Eichrom®-LN

226   columns. Total procedural blank was about 2 - 5 pg for Re, 2 pg for Ir, and 15 pg for

227   Ru, Pt and Pd. The maximum blank corrections were < 10% for low-Re samples.

228   Osmium isotopic compositions were measured at IGGCAS on an Isoprobe-T

229   mass spectrometer operated in negative ion mode (Creaser et al., 1991; Volkening et

230   al., 1991). Purified Os was loaded onto platinum filaments with $Ba(OH)_2$ as an ion

231   emitter and measured as OsO_3^-. Most samples were measured using a single ETP

232   multiplier in peak-jumping mode, while some samples with relatively high Os

233   concentrations were run in static mode on Faraday cups. The measured Os isotopic

234   ratios were corrected for mass fractionation using $^{192}Os/^{188}Os$ = 3.08271 (Shirey and

235   Walker, 1998). The in-run precisions for Os isotopic measurements were better than

236   0.2% (2 RSD) for all the samples. During the period of measurement of our samples,

237   the $^{187}Os/^{188}Os$ ratio of the Johnson-Matthey standard of University of Maryland

238   (UMD) was 0.11378 ± 4 (2 SD, n = 20) on nano-gram sized loads measured with

239   Faraday cups and 0.1138 ± 4 (2 SD, n = 13) on 3.5-175 pg sized loads measured with

240   the electron multiplier.

241   Measurements of $^{185}Re/^{187}Re$, $^{191}Ir/^{193}Ir$, $^{194}Pt/^{196}Pt$, $^{105}Pd/^{106}Pd$ and $^{99}Ru/^{101}Ru$

242   were conducted at IGGCAS using a Thermo-Fisher Neptune Multi-Collector ICP-MS

243 system with an electron multiplier in peak-jumping mode or using Faraday cups in

244 static mode, according to the measured signal intensity. The interferences of HfO (on

245 Ir and Pt), ZrO (on Pd) were confirmed to be negligible by scanning on ^{90}Zr and ^{180}Hf

246 peaks before sample measurement. Mass fractionation was corrected using Re, Ir, Ru,

247 Pt, and Pd standards that were interspersed with the samples. Some Ru samples were

248 run on the Isoprobe-T mass spectrometer in negative ion mode after a further

249 micro-distillation purification step, following the procedures described by Becker and

250 Walker (2003). In-run precisions for ^{185}Re/^{187}Re, ^{191}Ir/^{193}Ir, ^{194}Pt/^{196}Pt, ^{105}Pd/^{106}Pd, and

251 ^{99}Ru/^{101}Ru were typically 0.1 - 0.5% (2 RSD).

252   The USGS basalt standard BHVO-2 was measured to monitor the reliability of

253 the whole procedure. The Re, Os, Ir, Ru, Pt, and Pd concentrations and Os isotopic

254 ratio are in agreement with the reported values (Meisel and Moser, 2004; Shinotsuka

255 and Suzuki，2007) (Table 3).

256

257 **4. Results**

258 *4.1. Major and Trace elements*

259   Bulk-rock major and trace element compositions are presented in Table 1.

260 Consistent with previously reported results (Zhang et al., 1995), the WDLC and EKS

261 basalts have high K_2O abundances ranging from 4.50 wt% to 6.09 wt% and K_2O/NaO

262 ranging from 1.09 to 1.67. The SiO_2 abundances range from 49.0 to 53.5 wt%. The

263 MgO, CaO and Al_2O_3 abundances range from 4.45 to 9.47 wt%, 5.09 wt% to 7.44

264 wt% and 12.2 wt% to 14.4 wt%, respectively. These basalts have relatively low $Mg^{\#}$

265   values $(100Mg^{2+}/(Mg^{2+}+Fe^{2+}))$, falling between 61.9 and 70.6, except for sample YQ-4,

266   having $Mg^{\#}$ value of 55.3. The loss on ignition (LOI) values range from -0.2 to 0.1

267   wt%, consistent with the generally low degrees of alteration of these samples.

268   Negative LOI values of some samples are probably due to the oxidation of FeO to

269   Fe_2O_3 being more significant than loss of volatiles. Consistent with previous reports

270   (Zhang et al., 1995; Zou et al., 2003; Chen et al., 2007), the WDLC and EKS basalts

271   have relatively low CaO and particularly low Al_2O_3 at a given MgO content compared

272   to the basalts from elsewhere in NE China (Fig. 2).

273   The WDLC and EKS basalts have high rare earth element (REE) and large ion

274   lithophile element (LILE) concentrations. On a chondrite normalized REE diagram

275   (Fig. 3a), the WDLC and EKS basalts show uniform REE characteristics with strong

276   LREE enrichment $((La/Yb)_N = 51.0 \pm 2.5)$. The heavy REEs are also strongly

277   fractionated with $(Sm/Yb)_N = 9.7 \pm 0.6$. These samples also have uniform primitive

278   mantle-normalized trace element patterns (Fig. 3b), with strong LILE enrichment and

279   pronounced enrichment of Pb and K, slight enrichment of Zr and Hf, and significant

280   depletion of U, Th, Nb and Ta. Ce/Pb ratios of these basalts show slight variations

281   (10.6 ± 1.3), and are much lower than the values of ocean island basalt (OIB; 25 ± 5,

282   Hofmann et al, 1986). The La/Nb ratios of most basalts are greater than 1.2. Ba/Nb

283   ratios range from 23 to 29.

284   Most WDLC and EKS basalts have low S contents, ranging from lower than 50

285   ppm to 100 ppm, except for a single LHS sample, LHS-6, having a S content of 140

286   ppm. These concentrations are much lower than those of mid-ocean ridge basalts (>

287   1000 ppm, e.g., Gannoun et al., 2007).

288

289   *4.2. Sr-Nd-Hf isotopes*

290   The Sr and Nd isotopic compositions of the WDLC and EKS basalts are listed

291   in Table 2. The basalt samples have relatively high $^{87}Sr/^{86}Sr$ ratios varying between

292   0.7051 and 0.7057. The $^{143}Nd/^{144}Nd$ ratios range from 0.5123 to 0.5125. The Sr-Nd

293   isotopes of the WDLC and EKS basalts are more evolved (i.e., Sr isotopic

294   compositions are more radiogenic and Nd isotopic compositions are less radiogenic)

295   than those of mantle xenoliths carried in nearby Keluo basalts (Zhang et al., 2000;

296   Zhang YL et al., 2011) and Cenozoic basalts elsewhere in NE China (e.g., Chen et al.,

297   2007; Yan and Zhao, 2008) (Fig. 4a). The $T_{DM}(Nd)$ model ages of these basalts range

298   from 918 to 1057 Ma. Consistent with previously reported results (Zhang et al., 1995;

299   Zou et al., 2003), $^{87}Sr/^{86}Sr$ correlates negatively with $^{143}Nd/^{144}Nd$, demonstrating

300   Sr-Nd isotope systematics similar to EM1 (Zindler and Hart, 1986) (Fig. 4a).

301   The Lu-Hf isotopic compositions of the WDLC and EKS basalts are also listed

302   in Table 2. The Lu/Hf ratios range from 0.0018 to 0.0030, which are far lower than the

303   value for modern chondrite (0.0332, Blichert-Toft and Albarède, 1997). The

304    $^{176}Hf/^{177}Hf$ ratios vary from 0.2825 to 0.2827 (Table 2; Fig. 4b). In term of Hf

305   isotopes, the WDLC and EKS basalts are also more evolved (i.e., have less radiogenic

306   Hf isotopic compositions) than the mantle xenoliths they host (Zhang et al., 2011)

307   (Fig. 4b). Similar to Nd model ages, these basalts give uniform $T_{DM}(Hf)$ model ages

308   ranging from 879 to 1076 Ma. On the Hf-Nd isotopic correlation diagram, no obvious

309   Hf-Nd decoupling is seen when compared to MORB or OIB (Fig. 4b).

310

311   *4.3. Re-Os isotopes*

312   Re-Os isotope data for the WDLC and EKS basalts are given in Table 3. In

313   general, compared with Cenozoic basalts from other intraplate localities such as

314   Columbia River, Hannuoba, South-Eastern (SE) Australia and Central Europe

315   (Chesley and Ruiz, 1998; McBride et al., 2001; Jiang and Zhi, 2010; Jung et al., 2011),

316   the WDLC and EKS basalts have higher Os and lower Re contents (Fig. 5a). Most

317   samples have Re concentrations less than 0.1 ppb. The only exception is one analysis

318   of sample LHS-6, giving Re concentrations of 0.895 ppb. The WDLC and EKS

319   samples have Os concentrations ranging from 0.029 ppb to greater than 0.2 ppb. One

320   analysis of sample LHS-6 gives extremely high Os concentration (1.55 ppb).

321   The ^{187}Os/^{188}Os values of most samples range from 0.13 to 0.17, which

322   overlaps with the ^{187}Os/^{188}Os ranges of OIB basalts summarized in Shirey and Walker

323   (1998) and Harvey et al. (2011), and MORB glass (e.g., Alard et al., 2005). The only

324   exception is the high Os concentration sample LHS-6. Two analyses of this sample

325   yield low ^{187}Os/^{188}Os values of 0.1187 and 0.1245, respectively. As shown in Fig. 5b,

326   compared with the Cenozoic continental basalts from Hannuoba, SE Australia and

327   Central Europe (Chesley and Ruiz, 1998; McBride et al., 2001; Jiang and Zhi, 2010;

328   Jung et al., 2011), the WDLC and EKS potassic basalts generally have much lower

329   age-corrected ^{187}Os/^{188}Os ratios.

330   Replicate analyses of some WDLC and EKS basalts (Table 3) not only rarely

331   gave reproducible Re and Os concentrations, but also failed to reproduce $^{187}Os/^{188}Os$

332   ratios. Especially, replicate analyses of sample LHS-6 gave highly heterogeneous Re

333   and Os concentrations, and Os isotopic ratio. The higher $^{187}Os/^{188}Os$ ratio of sample

334   LHS-6R relative to LHS-6 corresponds with a lower Os concentration.

335

336   *4.4. Platinum-group elements (PGE)*

337   PGE data for the WDLC and EKS basalts are presented in Table 3. In general,

338   the WDLC and EKS basalts have low PGE abundances, ranging from 0.0244 to 0.282

339   ppb for Ir, 0.0525 to 0.364 ppb for Ru, 0.240 to 0.971 ppb for Pt, and 0.0571 to 0.295

340   ppb for Pd. Similar to the Re and Os concentration measurements, the PGE

341   abundances in replicate analyses of some samples also vary considerably, far beyond

342   analytical uncertainties (Table 3). Despite this, as shown in Fig. 6, the

343   chondrite-normalized PGE patterns (and hence PGE element ratios) are generally

344   reproducible. The WDLC and EKS basalts show only slight PPGE enrichment relative

345   to IPGE $((Pd/Ir)_N = 1.9 \pm 1.1)$ (Table 3). Exceptionally, consistent with its

346   un-radiogenic Os, sample LHS-6 has relatively high Ir and Os concentrations relative

347   to Pt and Pd (Fig. 6). Additionally, sample LHS-6 and LHS-6R (particularly LHS-6)

348   have elevated Os/Ru and Os/Ir ratios compared to those of the other samples.

349

350   **5. Discussion**

351   *5.1. PGE and Os isotope heterogeneity in the WDLC and EKS basalts*

352   Mantle sulphides and refractory PGE-rich alloys have parts-per-million (ppm)

353   to weight percentage (wt %) concentrations of PGE (e.g., Lorand and Alard, 2001;

354 Walker et al., 2002; Pearson et al., 2004; Lorand et al., 2010, 2013), making them

355 prime candidates for a source of a possible "nugget effect". For example, a 20 µg

356 mantle sulphide with an Os concentration of 20 ppm (e.g., Alard et al., 2000, 2002;

357 Pearson et al., 2002; Harvey et al., 2006, 2010, 2011; Lorand et al., 2013) contains

358 twice as much Os as 2 g of basalt with an Os concentration of 100 ppt. Therefore, the

359 heterogeneous distribution of a component that contains anomalously high Os (+ PGE)

360 abundances throughout the sampled rock powder could account for the observed

361 heterogeneities in replicate basalt analyses (Table 3). Although no discrete sulphide or

362 PGE alloy grains have been observed in the WDLC and EKS basalts, a "nugget

363 effect" due to incorporation of such mantle phases during the ascent of basalt through

364 the SCLM should be considered. Indeed, the inclusions enclosed in xenocrystic

365 material in basaltic lavas have been previously suggested as a mechanism to account

366 for unradiogenic $^{187}Os/^{188}Os$ and elevated Os concentrations (e.g., Widom et al., 1999;

367 Jackson and Shirey, 2011).

368   Although analysed samples of the WDLC and EKS basalts were hand-picked to

369 avoid obvious xenolith debris, microprobe analyses of olivine "phenocrysts" showed

370 that minor proportions of the olivines are mantle xenocrysts (i.e., a few olivine grains

371 from samples LHS-4 and KD-4 have Fo > 90, CaO < 0.10 wt % (Table S1), cf.,

372 Sobolev et al., 2007; Jackson and Shirey, 2011). Any sulphides hosted within

373 xenocrystic grains (e.g., Burton et al., 1999) would not have the opportunity to

374 become dispersed within a larger volume of melt. If its host was not recognized as

375 xenocrystic at the time of basalt preparation, this could readily account for the

376   elevated Os (+ IPGE) abundance and unradiogenic ^{187}Os/^{188}Os in some WDLC and

377   EKS samples, especially for LHS-6. Moreover, the WDLC and EKS basalts with

378   relatively high Ni and S content have the highest Os concentrations and least

379   radiogenic Os isotopic compositions (Fig. 7a-d), also consistent with the incorporation

380   of sulphide. Since it is possible that refractory alloys could be mechanically

381   transported in an ascending melt (e.g., Ballhaus et al., 2006), the heterogeneous

382   distribution of refractory alloys may also be possible to cause the "nugget effect" in

383   WDLC and EKS samples. This is supported by the relatively low Pt and Pd

384   concentrations relative to Ir and Ru in these samples compared to OIB and MORB

385   (especially LHS-6) (Fig. 6). In addition, Cr-spinel xenocrysts, of probable mantle

386   origin, are observed in sample LHS-4 (Fig. S1b). Although spinel itself is more

387   Os-rich than silicate mantle phases (Burton et al., 1999; Harvey et al., 2010, 2011), it

388   is unlikely to account for a significant proportion of mantle-derived PGE itself.

389   Consequently, it appears that sulphides or PGE alloys enclosed in xenocrysts are the

390   most likely candidates for the heterogeneity for PGE concentrations and Os isotopic

391   compositions in the WDLC and EKS basalt.

392   Although the incorporation of moderately radiogenic, interstitial metasomatic

393   sulphides from the SCLM also seems to be a plausible source of heterogeneity, in all

394   likelihood those sulphides would be dissolved by a S-undersaturated melt (Reisberg et

395   al., 2005; Sen et al., 2011) and its PGE budget homogenized within a relatively large

396   volume of melt compared to the volume of sulphide dissolved. It seems that this

397   process would not account for the PGE heterogeneity between different powder splits

398   of the same sample.

399

400   *5. 2. Crustal contamination*

401   Since basaltic magmas have high concentrations of lithophile elements,

402   Sr-Nd-Pb-Hf isotopes and lithophile elements are not particularly sensitive to minor

403   crustal contamination. This will be especially true for the LILE- and LREE- rich

404   WDLC and EKS basalts. However, even minor amounts of crustal contamination

405   have the potential to significantly alter the Os isotopic composition of a basaltic

406   magma with low Os concentrations (cf., Lassiter and Luhr, 2001; McBride et al., 2001;

407   Xu et al., 2007; Qi et al., 2008; Jung et al., 2011). It is possible that some of the low

408   Os WDLC and EKS basalts have been affected by crustal contamination during

409   magma ascent, in particular, at degrees that are not easily detected using lithophile

410   elements. Therefore, the $^{187}Os/^{188}Os$ and PGE systematics of the WDLC and EKS

411   basalts potentially allow this small degree of crustal contamination to be quantified.

412   As argued above, it is possible that some WDLC and EKS basalts have

413   incorporated some primary sulphides (sulphides enclosed in silicate minerals, i.e.,

414   Os-rich, Re-poor, evolving in low-Re environment and preserving unradiogenic

415   osmium isotope ratios) or PGE alloys of SCLM origin during magma ascent.

416   Moreover, most of these basalts have relatively low MgO contents (Table 1) and are

417   thus unlikely to be primary melts, suggesting that Os concentrations in the WDLC and

418   EKS basalts were also likely to be strongly affected by differentiation. Consequently,

419   it is likely that the basaltic magmas arrived in the crust with a range of Os

420   concentrations. As a result, it is difficult to constrain the degree of crustal

421   contamination by assimilation–fractional crystallization (AFC) modeling (DePaolo,

422   1981). Nevertheless, as shown in Fig. 8, there is a significant covariation between

423    $^{187}Os/^{188}Os$ and 1/Os ($R^2 = 0.47$), demonstrating that it seems likely that the WDLC

424   and EKS basalts have been affected by minor crustal contamination during magma

425   ascent. An Os isotope ratio c. 0.128 is considered to be representative of the parental

426   basaltic magma as suggested by previous studies (e.g., McBride et al., 2001; Xu et al.,

427   2007; Jiang and Zhi, 2010; Jung et al., 2011). Therefore, we assume the parental

428   magma of the WDLC and EKS basalts to have roughly a $^{187}Os/^{188}Os$ of c. 0.128, but

429   with varying Os concentrations (about 0.03 ~ 1.6 ppb, based on samples analyzed in

430   this study, Table 3) when they arrive in the lower crust. Consequently, if an Os

431   isotope ratio of 0.8 and Os concentration of 49 ppt is taken as being representative of

432   the lower crust (Saal et al., 1998; Chesley et al., 2002), the degree of crustal

433   contamination observed in the WDLC and EKS basalts can be modeled by simple

434   binary mixing. It is found that the addition of about only 3.5% of lower crust can

435   replicate the observed covariation between $^{187}Os/^{188}Os$ and 1/Os ($R^2 = 0.47$; Fig. 8)

436   fairly well. Nonetheless, as shown in Fig. 8, it is possible that some samples,

437   especially from different volcanoes, have experienced different degrees of crustal

438   contamination ranging from 2% to 8%. The rough correlation between the Pd/Ir and

439    $^{187}Os/^{188}Os$ (Fig. 8) ($R^2 = 0.39$) is also consistent with minor crustal contamination in

440   some basaltic melts, since the samples with high Pd/Ir ratios are a result of low Ir

441   contents, not high Pd contents.

442   Nevertheless, there remains the possibility that some samples with radiogenic

443 Os isotopes, which plot above the line for 3.5% of crustal contamination, result from

444 preferential melting of interstitial sulphides (sulphides occurred along boundary of

445 mineral grains, Re-rich, Os-poor, evolving in high-Re environment and having

446 variable and radiogenic osmium isotope ratios) in a metasomatized mantle source

447 (Alard et al., 2005; Schulte et al., 2009; Harvey et al., 2011) and / or the addition of

448 minor SCLM-derived interstitial sulphides during magma ascent (e.g., Sen et al.,

449 2011). This is particularly true for the YQ samples, as reflected by their radiogenic Os

450 but relatively high Os concentrations. However, we still prefer that the YQ samples

451 have experienced a larger degree of crustal contamination since their Pd contents are

452 low. In contrast, the samples which plot below the line for 3.5% crustal contamination

453 are probably due to the incorporation of SCLM-primary sulphides.

454   Addition of minor amounts (2 ~ 8%) of lower crust would not have much effect

455 on incompatible elements and Sr-Nd-Pb-Hf isotopic ratios for the LILE- and

456 LREE-rich WDLC and EKS basalts. For example, if we assume the local lower crust

457 having Nd concentration of 20 ppm (Rudnick and Gao, 2003) and $^{143}Nd/^{144}Nd$ of

458 0.51265 (Wu et al., 2003), 8% contamination of lower crust only shift the $^{143}Nd/^{144}Nd$

459 of the WDLC and EKS basalts of about 0.000007. Similarly, incorporation of primary

460 mantle sulphides or the addition of metasomatized sulphides would also have

461 negligible effect on the lithophile element systems and Sr-Nd-Pb-Hf isotopes. As a

462 result, no correlation exists between Os isotopic compositions and incompatible trace

463 element ratios such as Ce/Pb and La/Nb, and Sr or Nd or Hf isotopic compositions

464   (not shown); indeed, there are very limited variations in Sr-Nd-Hf isotopes and Ce/Pb

465   and La/Nb ratios, especially for YQ samples (Tables 1 and 2).

466   *5.3. Source of the WDLC and EKS basalts —SCLM vs. asthenosphere*

467   It is highly debated that the EM1-like features of the Cenozoic basalts in NE

468   China originated from either the SCLM or the asthenospheric mantle (e.g., Basu et al.,

469   1991; Song et al., 1990; Zhang et al., 1995; Zou et al., 2003; Choi et al., 2006; Xu et

470   al., 2012; Kuritani et al., 2013). Based on the highly potassic, LREE and

471   LILE-enriched trace elemental characteristics, the strong fractionation of the heavy

472   REE (HREE), low Al_2O_3 content at a given MgO, significant excesses in ^{230}Th, and

473   Sr-Nd-Pb isotopic characteristics, Zhang et al. (1995) and Zou et al. (2003) suggested

474   that the WDLC and EKS basalts were likely produced by small-degree melting of

475   phlogopite-bearing garnet peridotite in metasomatized SCLM. The presence of garnet

476   constrains the depth of the magma source to 80–120 km (Zou et al., 2003). However,

477   several studies have suggested that delaminated cratonic lithosphere from eastern

478   China may exist locally in the shallow asthenosphere beneath NE China (e.g., Xu et

479   al., 2005; Choi et al., 2006) and that basalts from this region may preserve evidence

480   for re-melting of this reservoir. They argued that the asthenospheric mantle beneath

481   eastern Asian is characterized by two distinct, large-scale domains — one a mixture of

482   DMM and EM1 components, and the other a mixture of DMM and EM2. More

483   recently, some authors (Xu et al., 2012; Kuritani et al., 2013) argued that the Cenozoic

484   basalts in NE China originated from an asthenospheric mantle source containing

485   recycled oceanic crust materials. They suggest that the subducted Pacific slab (Huang

486 and Zhao, 2006) and / or the stagnant ancient Pacific slab (Kuritani et al., 2011) in the

487 mantle transition zone have contributed to the Cenozoic magmatism in NE China.

488 This is reasonable as the re-melting of upwelling asthenospheric mantle containing

489 recycled oceanic or lower continental crust has widely been invoked to account for the

490 evolved Sr-Nd-Hf isotopic composition of intraplate basalts (e.g., Sobolev et al., 2007;

491 Chen et al., 2009; Zhang et al., 2009; Xu et al., 2012).

492 As discussed above, crustal contamination has not played an important role in

493 the formation of the WDLC and EKS basalts. Therefore, consistent with previous

494 studies (Zhang et al., 1995; Zou et al., 2003), data obtained in this study, including

495 low Al_2O_3 content at a given MgO (Fig. 2a), high K content, LREE and

496 LILE-enriched trace elemental characteristics (Fig. 3a, b), particularly strong

497 fractionation of the heavy REE $((Sm/Yb)_N = 9.7 \pm 0.6)$, EM1-like Sr-Nd-Hf isotopic

498 characteristics (Fig. 4a, b), all suggest that the WDLC and EKS basalts mainly

499 originated from a phlogopite-bearing garnet peridotite in the SCLM. The presence of

500 phlogopite and rutile in some Keluo and WDLC xenoliths (Zhang et al., 2000; Zhang

501 YL et al., 2011) also supports that the SCLM beneath WEK area has been

502 metasomatized by K-rich silicate melt.

503 In the model involving delaminated ancient SCLM from eastern China that

504 exists locally in the asthenosphere beneath NE China (Choi et al., 2006), amphibole

505 and phlogopite will breakdown at high pressure (> 3 GPa) and are thus not considered

506 to be stable in the asthenospheric mantle (Class and Goldstein, 1997). Moreover, a

507 delaminated ancient SCLM from eastern China is also unlikely to be the source

508   material for the EM1-like signature because the delaminated Archaean SCLM beneath

509   eastern China does not have an EM1-like composition (e.g., Chu et al., 2009 and

510   references therein). Therefore, it seems plausible that phlogopite-bearing

511   metasomatized SCLM rather than an asthenospheric source containing fragments of

512   delaminated ancient cratonic mantle may provide a source for the WDLC and EKS

513   high-K basalts.

514   In the model that the potassic- and EM1-like natures of the basalts originated

515   from the mantle transition zone, which has been metasomatized by K-rich

516   sediment-derived fluids ~1.5 Ga ago through the stagnation of an ancient slab and

517   further recent hydration by the stagnation of the subducted Pacific slab (Kuritani et al.,

518   2013), it is difficult to envision such a process because of the great distance (>2000

519   km) between the WEK provinces and the active Pacific subduction zone (Japan Arc)

520   (Zhang et al., 1998; Zou et al., 2003) in western NE China. Significant ^{230}Th

521   enrichments in WDLC basalts also suggest that the source rocks were not

522   metasomatized by fluids released during recent (< 350 ka) subduction of Pacific

523   sediments (Zou et al., 2003). Furthermore, it has long been recognized that LILEs,

524   such as Ba, Rb, and Sr are fluid-mobile and have elevated concentrations in sub-arc

525   environments, while fluid-immobile elements such as Nb and Ta, are largely

526   unaffected by contributions of fluids from the subducting slab (Arculus and Powell,

527   1986). A mantle source which has been metasomatized by a subduction-related fluid

528   would be enriched in radiogenic Os (Brandon et al., 1996; Widom et al., 2003). Due

529   to the effect of preferential melting of metasomatic components for Os isotopes (e.g.,

530   Sen et al., 2011; Harvey et al., 2011), melting of such a mantle source would produce

531   melts with slightly increasing $^{187}Os/^{188}Os$ along with increasing Ba/Nb (Chesley et al.,

532   2002). Such a trend is absent in the WDLC and EKS basalts (Fig. 10). In contrast,

533   some samples especially YQ samples with high $^{187}Os/^{188}Os$ have relatively low Ba/Nb

534   ratios. In fact, the WDLC and EKS basalts have very low Ba/Nb ratios with limited

535   variability (23-29).

536   Consequently, it seems most likely that the WDLC and EKS mainly originated

537   from low degree melting of a SCLM source metasomatized by an EM1-like signature,

538   rather than an EM1 signature asthenospheric mantle source.

539   In terms of Os isotopes, the WDLC and EKS basalts generally have

540   unradiogenic to moderately radiogenic Os isotopic compositions ($^{187}Os/^{188}Os$ =

541   0.1187 ~ 0.17). In principle these relatively OIB-like Os isotopic values are not

542   inconsistent with an asthenospheric origin (Liu et al., 2008; Harvey et al. 2011).

543   However, as discussed before, the measured Os isotope ratios cannot be attributed

544   solely to the mantle reservoir from which the basalts were produced. Most of the

545     $^{187}Os/^{188}Os$ values have been elevated by up to 2 ~ 8% crustal contamination and / or

546   the effects of incorporating metasomatic sulphide en route to the surface, while the

547   most unradiogenic values may be deceptively low because of the incorporation of

548   xenocryst-hosted primary sulphide. Similarly, the relative flat PGE patterns ($(Pd/Ir)_N$

549   = 1.9 ± 1.1) compared to MORB and OIB (Fig. 6) could indicate the contribution of a

550   SCLM source for the WDLC and EKS basalts and, like Os isotope ratios, can also be

551   interpreted in a number of ways. Therefore, notwithstanding the clear utility of Os

552 isotopes and PGE in fingerprinting the sources of basalts that have not experienced

553 complex histories (cf., Shirey and Walker, 1998; Day, 2013), these tools do not

554 unequivocally distinguish between an asthenospheric and a lithospheric source for the

555 WDLC and EKS basalts.

556

557 *5.4. Nature of the SCLM underlying the WEK area*

558 As discussed above, the WDLC and EKS basalts mainly originated from the

559 underlying SCLM. Despite this, as the WDLC and EKS basalts have more evolved

560 Sr-Nd-Hf isotopic signatures than those of mantle xenoliths carried in nearby Keluo

561 highly potassic basalts (Zhang et al., 2011) (Fig. 5a, b), the role of peridotite with the

562 composition similar to that of Keluo xenoliths can be largely discounted in the

563 petrogenesis of the WDLC and EKS basalts. Moreover, since the Keluo xenoliths

564 mainly comprise spinel lherzolites and harzburgites with minor dunites and wehrlites,

565 they should have been located in the SCLM above the source of the WDLC and EKS

566 basalts (garnet peridotites). Therefore, it is possible that the lithospheric mantle

567 underlying the WEK area is layered, at least in terms of Sr-Nd-Hf isotopes, i.e., the

568 lower part of lithospheric mantle (the potential source of the WDLC and EKS basalts)

569 has more radiogenic Sr and less radiogenic Nd and Hf than the upper part of

570 lithospheric mantle (represented by the Keluo mantle xenoliths), despite their similar

571 Nd and Hf model ages (Zhang et al., 2000) (Table 2).

572 The crustal growth in XMOB during the Phanerozoic is significant, as reflected

573 by the radiogenic Nd-Hf isotopic compositions of granites in the area (e.g., Wu et al.,

574   2003; Zhang et al., 2010), while the potassic rocks originating from the underlying

575   SCLM have EM1-like Sr-Nd-Hf isotopic signatures as demonstrated above. This

576   suggests that the SCLM beneath the WEK area is decoupled from the overlying crust.

577   Previous studies argued that the SCLM beneath the Phanerozoic XMOB is ancient

578   and have been metasomatised extensively by SiO_2-undersaturated potassic magma

579   that was likely generated from low-degree partial melting of deep asthenosphere

580   during the Proterozoic (Zhang et al., 1995, 2000; Zou et al., 2003; Zhang YL et al.,

581   2011).

582   EM1-like signatures of the WDLC and EKS basalts cannot be owed to

583   metasomtism by melts derived from subducted oceanic crust related to the closure of

584   the Paleo-Asian Ocean in the Paleozoic and Paleo-Pacific Ocean during the Mesozoic.

585   Their isotopic compositions fall outsides the range for those of oceanic basalts and

586   MORB that are believed to be derived primarily from the asthenospheric mantle.

587   Especially, the very unradiogenic Pb isotopic signatures of the potassic rocks

588   ($^{206}Pb/^{204}Pb$=16.3-17.3), which require a long-term residence (>2 Ga) of low U/Pb in

589   the mantle source, indicate that the Pb isotopic signatures of the WEK basalts cannot

590   be attributed to modifications by Phanerozoic subduction events in the region (Zhang

591   et al., 1998). Moreover, the partial melting of peridotite cannot cause a large U/Pb

592   fractionation and, therefore, the melt generated from low-degree partial melting of

593   asthenosphere cannot have a significantly different U/Pb ratio from that of the

594   depleted mantle (Kuritani et al., 2013). Therefore, it is also difficult to derive the very

595   unradiogenic Pb isotopic signatures of the potassic rocks ($^{206}Pb/^{204}Pb$=16.3-17.3) with

596   the model presented by Zhang et al. (1995) and Zou et al. (2003).

597   As lower continental crust is known to be depleted in U relative to Pb (e.g.,

598   Rudnick and Gao, 2003), given enough time it will develop unradiogenic Pb isotopic

599   compositons. Consequently, based on low Ce/Pb (~9.4-13.5) and EM1-like Sr-Nd-Hf

600   isotopic signatures, in conjugation with low Pb isotope ratios (^{206}Pb/^{204}Pb = 16.61 to

601   17.06) as reported by Zhang et al. (1995) and Zou et al. (2003), different from the

602   previous studies (Zhang et al., 1995, 1998; Zou et al., 2003; Kuritani et al., 2013), we

603   suggest that the mantle source of WDLC and EKS basalts have been metasomatised

604   by recycled old lower continental crust materials (i.e., an EM1-like signature). It is

605   likely that some pyroxenite would be formed in the SCLM if mantle peridotite was

606   metasomatised by melt containing a contribution from lower crustal materials (e.g.,

607   Xu, 2002). Some pyroxenite xenoliths were found in WEK basalts (Zhang et al.,

608   2000), consistent with such an explanation. This is also supported by the relatively

609   low CaO abundance at a given MgO (Fig. 2b) for the WDLC and EKS basalts, as

610   melts of pyroxenite are generally low in CaO owing to the dominant effect of residual

611   clinopyroxene (e.g., Herzberg, 2011).

612   It is likely that the present lower crust in XMOB is significantly different from

613   earlier crust at this locality. As there are probably some Precambrian fragments

614   present in NE China (Wu et al., 2003; Meng et al., 2010), it is likely that the original

615   crust in WEK area was old. It is possible that the original lower crust together with

616   SCLM foundered probably during lithospheric thinning in eastern China that started

617   in the Mesozoic. The delamination of the old lower crust is probably related to the

618 multiple subduction events in this region. Subsequently, the newly-formed SCLM

619 beneath the WEK area was metasomatised by melt contributed from the delaminated

620 lower crust. The whole SCLM beneath the WEK area was extensively and

621 contemporaneously metasomtised as reflected by the similar Nd (Hf) model age

622 between the mantle xenoliths (Zhang et al., 2000; Zhang YL et al., 2011) and the

623 potassic basalts (Table 2) in this area. It is possible that the lower part of the SCLM

624 have been affected more strongly than the upper part of the SCLM. This

625 metasomatism leads to the SCLM underlying the WEK area having more radiogenic

626 Sr and less radiogenic Nd and Hf isotopic compositions than the overlying

627 Phanerozoic crust.

628   Conclusively, consistent with previous studies (Zhang et al., 1995, 1998; Zou et

629 al., 2003), we suggest that the WDLC and EKS basalts originated from

630 phlogopite-bearing garnet peridotite within the SCLM. During low degree of the

631 partial melting of the SCLM, metasomatic components such as phlogopites are easily

632 melted and their high-K characteristics exported into the WDLC and EKS basalts

633 (Zou et al., 2003). In contrast, garnet is mainly retained in the mantle source. This

634 leads to significant ^{230}Th excess in the WDLC and EKS basalts (Zou et al., 2003).

635 Similarly, hafnium isotopes could uniquely trace contributions from the SCLM if

636 melts are derived from garnet-bearing SCLM (Jung et al., 2011). Given sufficient time,

637 the strong increase of Lu/Hf relative to Sm/Nd in mantle with significant garnet would

638 result in trends that deviate from the crust-mantle Hf-Nd isotope array, towards

639 radiogenic ^{176}Hf/^{177}Hf at a given ^{143}Nd/^{144}Nd (e.g., Salters and Zindler, 1995; Bizimis

640 et al., 2007; Chen et al., 2009). However, the WDLC and EKS potassic lavas have

641 very low Lu/Hf ratios (Table 2) and do not show Hf-Nd isotope decoupling on Hf-Nd

642 correlation diagrams (Fig. 4b). This also indicates a low degree melting of the

643 phlogopite-bearing garnet peridotites, and consequently the Lu-Hf isotopic signature

644 of garnet in the SCLM has not been exported into the resulting basaltic magma.

645

646 **6. Concluding remarks**

647 　　(1) The WDLC and EKS highly potassic basalts show uniform REE and trace

648 elemental characteristics and relatively homogeneous Sr-Nd-Hf isotopic compositions,

649 but heterogeneous Os isotopic compositions ranging from 0.1187 to ~0.17. It is

650 possible that some samples with radiogenic Os isotopic compositions may have been

651 contaminated by minor amounts of lower crustal materials, while other samples, with

652 un-radiogenic Os isotopic compositions, may have incorporated small amounts of

653 mantle materials such as xenocryst-hosted primary sulphides and / or PGE

654 micro-alloys during magma ascent through the overlying SCLM. Both the correlation

655 between the $^{187}Os/^{188}Os$ and 1/Os, and the positive correlation between $^{187}Os/^{188}Os$

656 and Pd/Ir are consistent with these scenarios. In contrast, the minor crustal

657 contamination and incorporation of sulphides in the SCLM have an insignificant

658 influence on the lithophile elements and Sr-Nd-Hf isotopic compositions.

659 　　(2) The strong enrichments of LREE and LILE, and EM1 like Sr-Nd-Hf

660 isotopic characteristics suggest that the WDLC and EKS highly potassic basalts

661 mainly originated from metasomatized SCLM. Combined with the WDLC and EKS

662   basalts having Ce/Pb ratios (~9.4-13.5) much lower than typical OIB and MORB, and

663   low $^{206}Pb/^{204}Pb$ ratios, it seems likely that the source of WDLC and EKS basalts is

664   SCLM that has been metasomatized by delaminated old lower continental crust

665   materials. Furthermore, since the WDLC and EKS basalts have more radiogenic Sr

666   and less radiogenic Nd-Hf isotopic compositions than the SCLM xenoliths carried in

667   nearby Keluo highly potassic basalts, the lithospheric mantle underlying the

668   Wudalianchi-Erkeshan-Keluo (WEK) area is likely layered, at least in terms of

669   Sr-Nd-Hf isotopes.

670

681

682   **Figure captions**

683

684 **Fig. 1** Sketch map of Northeast (NE) China (modified after Chen et al., 2007), in

685 which the Cenozoic basalts, Great Xing'an Ranges, Songliao Basin and the locations

686 of the Wudalianchi, Erkeshan and Keluo volcanic rocks are shown. Dashed line is the

687 approximate boundary between the North China Craton (NCC) and Xing'an

688 Mongolian Orogenic Belt (XMOB); Abbreviations of volcanic fields are as follows:

689 WDLC (Wudalianchi), KD (Kedong), KL (Keluo) and HNB (Hannuoba).

690

691 **Fig. 2** CaO vs MgO (a) and Al_2O_3 vs MgO (b) for WDLC and EKS basalts. Solid

692 circles: this study; Open circles: WDLC and EKS data from Zou et al. (2003) and

693 Chen et al. (2007); Crosses: data for basalts from NE China elsewhere (Chen et al.,

694 2007; Yan and Zhao, 2008).

695

696 **Fig. 3** Chondrite-normalized rare earth element patterns (a) and primitive

697 mantle-normalized trace element patterns (b) for the WDLC and EKS basalts.

698 Chondrite normalizing values are from Masuda et al. (1973) divided by 1.2; Data for

699 OIB, N-MORB and primitive mantle are from McDonough and Sun (1995).

700

701 **Fig. 4** Sr-Nd (a) and Hf-Nd (b) isotope diagrams for WDLC and EKS basalts. In (a):

702 literature data for WLDC and EKS basalts are from Peng et al. (1986), Basu et al.

703 (1991), Zhang et al. (1995, 1998) and Zou et al. (2003); Field for lower continental

704 crust (LCC) in WDLC and EKS area is based on Wu et al. (2003). Field for Cenozoic

705 basalts in eastern China is based on Chen et al. (2007) and Yan and Zhao (2008);

706 MORB, OIB, BSE, EM1 and EM2 end members are from Zindler and Hart (1986). In

707 (b): MORB and OIB fields are based on Salters and White (1998); the bulk Earth

708 $^{176}Hf/^{177}Hf$ value (BSE) and the Mantle-Crust array are based on Blichert-Toft and

709 Albarède (1997).

710

711 **Fig. 5** Re vs. Os (a) and Os concentration vs. $^{187}Os/^{188}Os$ (b) variation diagrams. Data

712 sources: Central European (Jung et al., 2011); HNB, Hannuoba (Jiang and Zhi, 2010);

713 SE Australia (McBride et al., 2001); Columbia River (Chesley and Ruiz, 1998); Keluo

714 mantle xenoliths (Zhang et al., 2011); MORB and OIB fields are from Shirey and

715 Walker (1998): OIB TH, ocean island basalt tholeiites; OIB AB, ocean island alkali

716 basalts. In (b): Samples with Os > 0.8 ppb are not plotted for clarity; $^{187}Os/^{188}Os$ ratios

717 are all age-corrected values.

718

719 **Fig. 6** Chondrite-normalized PGE patterns for WDLC and EKS basalts. Chondrite

720 values are from McDonough and Sun (1995). Primitive mantle (PM) values are based

721 on Becker et al. (2006). Data for OIB-Canary Island alkali basalts with calculated

722 parental melt of 12 wt% MgO, and MORB are based on Day (2013) and references

723 therein. LCC (lower continental crust) values are based on Rudnick and Gao (2003).

724

725 **Fig. 7** Diagrams of Os concentration vs. Ni content (a), Os concentration vs. S content

726 (b), $^{187}Os/^{188}Os$ vs. Ni content (c) and $^{187}Os/^{188}Os$ vs. S content (d) for WDLC and

727 EKS basalts.

728

Fig. 8 ^{187}Os/^{188}Os vs. 1/Os. The solid lines represent binary mixing lines modeled as follows: (1) basaltic magma arrived in the crust with variable Os concentrations (about 0.03 ~ 1.6 ppb, based on samples analyzed in this study) but similar Os isotopic compositions (we assume ^{187}Os/^{188}Os = 0.128); (2) addition of 2%, 3.5% and 8% of lower crust (^{187}Os/^{188}Os = 0.8 and Os concentration = 49 ppt, Saal et al., 1998) to the basaltic magma. The line for addition of 3.5% of lower crust is consistent with the regression line (the dashed line, R^2 = 0.47) for data points from all the volcanoes. The arrows show the trends for addition of lower continental crust (LCC), secondary sulphides and primary sulphides.

738

Fig. 9 ^{187}Os/^{188}Os vs. (Pd/Ir)$_N$ for WDLC and EKS basalts. The solid line denotes the regression line for all the data points except for LHS-6R. Symbols and arrows as in Fig. 8. The Os isotopic composition and (Pd/Ir)$_N$ of LCC are about 0.8 (Saal et al., 1998) and 46 (Rudnick and Gao, 2003), repectively.

743

Fig. 10 ^{187}Os/^{188}Os vs. Ba/Nb for WDLC and EKS basalts. Symbols as in Fig. 8. Trends for contamination of mantle source of basalts by subduction-related fluids, fields of Ba/Nb and ^{187}Os/^{188}Os for the subduction fluids and the lower crust based are based on Chesley et al. (2002). Numbers along trend lines represent percentage of fluid required to influence the mantle source. Large numbers along trend lines represent the Os in ppt for subduction-related fluids used in the two-component

750 mixing model (the mantle source of WDLC and EKS basalts is assumed to be 1000

751 ppt). The real trend should possibly toward more radiogenic Os along with increasing

752 Bb/Nb ratios due to the effect of preferential melting of metasomatic components in

753 mantle source for Os isotopes (e.g., Harvey et al., 2011).

754

755 **Supplementary materials:**

756 Detailed descriptions for WDLC and EKS samples;

757 Petrography of representative samples for WDLC and EKS basalts (Figure S1);

758 EPMA data for olivines in WDLC and EKS basalts (Table S1).

759

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