Key Points:
- Low-Mg series (LMS) arc igneous rocks originate from fluid-/sediment melt-modified mantle wedge, followed by crystal fractionation
- High-Mg series (HMS) arc igneous rocks likely originate from recycled crustal materials as mélange diapirs rising into the mantle wedge
- Continental crust is formed by extracting LMS from the mantle followed by re-melting of recycled LMS in the mantle wedge to generate HMS

Supporting Information:
Supporting Information may be found in the online version of this article.

Correspondence to:
D.-C. Zhu, dchengzhu@163.com

1 State Key Laboratory of Geological Processes and Mineral Resources, and School of Earth Science and Resources, China University of Geosciences, Beijing, China. 2 School of Earth, Atmosphere & Environment, Monash University, Melbourne, VIC, Australia. 3 Geosciences Department, University of Texas at Dallas, Richardson, TX, USA

Abstract
The compositional similarity between high-Mg andesite-dacite from accretionary orogens and bulk continental crust (CC) provides an opportunity to unravel the CC formation paradox. Compositional data from a global compilation of Quaternary magmatic arcs indicate the presence of low-Mg series (LMS) and high-Mg series (HMS). The LMS show trends of crystal fractionation and can be subdivided into high Ba/Th and high La/Sm groups, which likely originate from fluid- and sediment melt-modified mantle wedge, respectively. In contrast, the HMS have variably mixed compositions (e.g., high Mg#, Ba/Th, and La/Sm) and can be explained by partial melting of mélange diapirs rising into the mantle wedge, which are mixtures of subducted sediment, eroded arc crust or CC, buoyant oceanic crust, and peridotite. We, therefore, propose a two-step process for creating CC involving extraction of LMS from the mantle followed by re-melting of recycled LMS in the mantle to generate HMS and thus CC.

Plain Language Summary
Continental crust (CC) is extracted from the mantle primarily by subduction-related magmatism in accretionary orogens. However, igneous rocks sourced from the mantle have low SiO2 (basalt), whereas average CC has high SiO2 (andesite) and high MgO. To resolve this paradox, we analyzed Quaternary subduction-related igneous rocks worldwide and find that they can be divided into low-Mg series (LMS) and high-Mg series (HMS). We compare existing compositional data for LMS and HMS and find that LMS are dominated by basalt whereas HMS are dominated by andesite and similar to the average CC. Petrogenetic analyses reveal that LMS likely originate from the mantle and represent primitive CC, whereas HMS likely originate from recycled crustal materials in the mantle and represent mature CC. We propose a two-step process for creating CC, involving extraction of LMS from the mantle followed by re-melting of recycled LMS to generate HMS and thus CC. Our results provide a new solution to the CC formation paradox.

1. Introduction
Continental crust (CC) provides the long-term record of origin and evolution of Earth's lithosphere, atmosphere, hydrosphere, and biosphere (Cawood et al., 2013). On the modern Earth, and since the commencement of plate tectonics, the CC is interpreted to be extracted primarily by subduction-related magmatism from the mantle (Rudnick, 1995) in accretionary orogens (Cawood et al., 2009). However, this raises a well-known paradox of CC formation that mantle-derived melts are generally basaltic and differ from CC, which has an overall andesitic-dacitic composition (Figure 1a; Rudnick & Gao, 2014). Resolving this discrepancy has proven to be a challenge, and three models are generally invoked: (a) delamination of mafic/ultramafic rock of lower crust into the mantle (Figures 1b–1e; Herzberg et al., 1983; Jagoutz & Kelemen, 2015; Lee & Anderson, 2015; Ringwood & Green, 1966); (b) reamination of subducted felsic rock to the base of the arc (Figures 1d–1e; Hacker et al., 2011, 2015; Hess, 1962; Kelemen & Behn, 2016); and (c) partial melting of basaltic oceanic crust (Gazel et al., 2015, 2019; Green & Ringwood, 1968) or eroded arc crust (Gómez-Tuena, Mori, & Straub, 2018; Straub et al., 2015) in the mantle. All three processes may occur, but whether any mechanism dominates CC formation is unclear.

Arc igneous rocks are divisible into low-Mg series (LMS) (e.g., Lee & Bachmann, 2014) and high-Mg series (HMS) (e.g., Gómez-Tuena, Mori, & Straub, 2018) based on Mg# (molar Mg/(Mg + Fe)) of andesite to dacite. Interestingly, both mafic lower CC and felsic upper CC are estimated to have high Mg# values (Kelemen & Behn, 2016; Rudnick & Gao, 2014), so that CC is akin to HMS in composition. Hence, unraveling the HMS generation must play a key role in understanding CC formation by plate tectonic processes. The three mechanisms...
for transforming basaltic arc crust into high-Mg andesitic CC invoke different processes of HMS genesis, including (a) crystal fractionation of primary basaltic or andesitic magma with cumulate or arclogite foundering into the mantle (Jagoutz & Kelemen, 2015) (Figures 1b–1c), (b) mixing between magmas derived from the mantle and relaminated felsic rocks (Kelemen & Behn, 2016) (Figures 1d–1e), and (c) melting of basaltic rocks with involvement of mantle peridotite in the source (Castro et al., 2010; Marschall & Schumacher, 2012; Nielsen & Marschall, 2017) or during magma ascent (Kelemen, 1995) (Figures 1f–1g).

Here, we compare existing compositional and geodynamic data for LMS and HMS from Quaternary accretionary orogens worldwide. We find that LMS likely originate from the metasomatized mantle and then evolve by crystal fractionation, whereas HMS are most likely derived from mélange-diapiric melting in the mantle wedge. We thus propose a two-step magmatic process (named here as the two-step mélange-diapir model; Figures 1f–1g), developed from the basaltic re-melting model, to explain CC formation. The two-step mélange-diapir model involves (a) extracting basalt from the mantle to form primitive arc crust (LMS), and (b) extracting HMS from mélange diapirs, which originate from recycled primitive arc crust in step 1, to create mature CC. Our results provide a new solution to the paradox of CC formation by determining HMS genesis in global magmatic arcs.

2. Data Compilation and Filtering

Geochemical data were compiled from the GEOROC database (http://georoc.mpch-mainz.gwdg.de/georoc/) for both oceanic arc segments of the Izu, Bonin, Mariana, Tonga, Kermadec, Lesser Antilles, W. Aleutian, N. Kamchatka, and SE Central America, and continental arc segments of the Sunda, Andes Southern Volcanic Zone (SVZ), Andes Central Volcanic Zone (CVZ), and Trans-Mexican Volcanic Belt (TMVB) (Figure S1 in Supporting Information S1). We filtered the data set based on the assumption that coeval rocks with similar
compositions in a limited region are genetically related or similar. Therefore, only Quaternary rocks whose geodynamic setting is clear and that are limited to selected arc segments instead of the whole arc were compiled. Petrologically, fore-arc samples (e.g., boninite), samples with obvious cumulate origins, and alkaline samples, which were generated in special regions of a magmatic arc (e.g., above slab windows) or in the retro-arc, were excluded. We did not discard data based on previous controversies regarding petrogenetic interpretations. Details for distributions and features of individual arcs and data filtering are given in Text S1 and Figures S1−S7 in Supporting Information S1.

3. Classification and Petrogenesis of LMS and HMS

The compiled data set reveals that igneous rocks from different arcs are divisible into LMS and HMS with distinct Mg# for andesite to dacite, but with overlapping Mg# for basalt and rhyolite (Figures 2a−2c). Similar features are also observed in MgO, Cr, and Ni contents (Figure S2 in Supporting Information S1). At SiO$_2 = 55$−65 wt%, Mg# = $0.099\text{SiO}_2^2 - 12.878\text{SiO}_2 + 462.24$ is used as the boundary between LMS and HMS. The Mg# values of most (~92%) LMS samples are lower than 53 and 43 at SiO$_2 = 55$ and 65 wt%, respectively, whereas HMS contain higher Mg# than this, although there is some (~20%) overlap. In fact, LMS and HMS exhibit systematic compositional differences. For example, LMS are dominated by basalt and characterized by either high Ba/Th and Sr/Th, or high K$_2$O and La/Sm, but not both; whereas HMS are dominated by andesite and characterized by high K$_2$O, La/Sm, and Ba/Th, together (Figures 2d−2f, Figures S3–S4 in Supporting Information S1). More details are discussed below. The formation of arc igneous rocks must involve various magmatic sources and processes. However, the compositional similarity of igneous rocks from different arcs in the same group indicates that some mechanisms dominate. Below we evaluate the first-order mechanism in generating LMS and HMS by comparing analyses.

3.1. Generating LMS by Partial Melting of Fluid or Melt-Modified Mantle Wedge Followed by Crystal Fractionation

A metasomatized-mantle model has been widely accepted to account for generating arc igneous rocks (R. J. Stern, 2002; Wilson, 1989). This model suggests that rising of slab-released fluid into the mantle wedge can trigger mantle melting to generate primary magma, which evolves further to produce basalt to rhyolite (Lee & Bachmann, 2014). The LMS can be explained by such processes because LMS are dominated by basalt (Figure 2c) and compositional variations are consistent with characteristics of crystal-melt fractionation. For example, as SiO$_2$ increases, Mg# values (Figure 3a) and TiO$_2$ contents (Figure 3b) of individual arcs exhibit trends identical to fractional-crystallization paths obtained from experiments (e.g., Blatter et al., 2013; Nandedkar et al., 2014). The trace-element (e.g., Sr/Nd, Hf/Nd or Hf/Sm) and Sr-Nd isotopic compositions of LMS (e.g., the Tonga arc) can be explained by both a metasomatized-mantle (Handley et al., 2011; Wu et al., 2020) and mélangé (Nielsen & Marschall, 2017) source. For the metasomatized-mantle model, results of Sr-Nd elemental and isotopic modeling indicate high-degree melting of sediments (up to 30%), accompanied with strong amphibole but limited plagioclase fractionation from arc magmas (Nielsen & Marschall, 2017). We prefer the metasomatized-mantle model based on two reasons. First, to generate LMS basaltic magma, extensive incorporation of silica-rich mélangé is less likely, which is evidenced by calculations of sediment contribution (Nielsen & Marschall, 2017). Therefore, slab dehydration remains a requirement to trigger mantle melting. Second, a new model is not required when the classic model works and its many aspects have been well studied (e.g., Grove et al., 2012).

Previous studies have proposed that slab dehydration prevails at all P-T conditions (<5 GPa), whereas sediment melting dominates at high P-T conditions, crossing the wet solidus of pelite with the presence of slab-derived fluids (Grove et al., 2012; Hermann & Spandler, 2008; Labanieh et al., 2012). The LMS data set favors above inferences by the presence of two distinct groups: the fluid-modified and sediment melt-modified groups. The fluid-modified group shows high fluid proxies of Sr/Th and Ba/Th (Elliott, 2003; Turner et al., 1996), low sediment-melt proxies of Th/La and La/Sm (Labanieh et al., 2012; Plank, 2005) (Figure 2d), low K$_2$O, and high $\varepsilon_{\text{Nd}}(t)$ values (Figure S3–S5 in Supporting Information S1). Sediment contribution is very little in this group (<0.5% for the Tonga arc; Nielsen & Marschall, 2017) so that fluid can control trace elemental signatures of arc igneous rocks. In contrast, the sediment melt-modified group has high La/Sm and low Ba/Th (Figure 2d), moderate K$_2$O, and markedly low $\varepsilon_{\text{Nd}}(t)$ values (Figure S3–S5 in Supporting Information S1). Sediment contribution is relatively high in this group (<5% for the Lesser Antilles arc; Nielsen & Marschall, 2017) so sedimentary
signatures are significant due to much higher elemental abundances in sediment melt (Hermann & Rubatto, 2009) than in the fluid (Kessel et al., 2005) and mantle wedge (Salters & Stracke, 2004).

3.2. Generating HMS by Melting of Recycled Crustal Materials as Mélange Diapirs in the Mantle Wedge

The genesis of high-Mg andesite-dacite, which are the majority in HMS, however, is controversial under the metasomatized-mantle framework. (a) Intra-crust processes are less likely to be the dominant mechanism.

Figure 2. Classification diagrams for low-Mg series (LMS) and high-Mg series (HMS). (a–b) Plots of Mg# versus SiO2. (d–e) Plots of Ba/Th versus La/Sm. Panels (c and f) Bivariate kernel density estimates and histograms enclosing 75% and 100% data of each group, respectively. Data used are listed at https://doi.org/10.17605/OSF.IO/QM49Y.
Figure 3. Plots of Mg# versus SiO$_2$ (a), TiO$_2$ versus SiO$_2$ (b–c), and Al$_2$O$_3$ versus SiO$_2$ (d). Experimental fractional-crystallization paths of primary basalt are from Alonso Perez (2007), Ulmer et al. (2018), and those compiled in Wang et al. (2018). Data of experimental mélangé melts are compiled in Codillo et al. (2018). Intrusive rocks from the Famatinian (Walker et al., 2015) and Kohistan (Jagoutz, 2010) arc sections are shown for comparison. (e) Boxplots for comparison of Mg#, K$_2$O, Ba/Th, La/Sm, and La/Yb, and whole-rock $^{143}$Nd/$^{144}$Nd of andesite (SiO$_2$ = 55–65 wt.%) from low-Mg series (LMS) and high-Mg series (HMS) arcs. Data of mélangé (Marschall & Schumacher, 2012), average bulk continental crust (CC) (Rudnick & Gao, 2014), and Pacific N-MORB (Gale et al., 2013) are shown for comparison. N-MORB = normal mid-ocean ridge basalt.
Experimental results imply that crystal fractionation of basaltic magma at varying pressure (0.2–1.5 GPa), H₂O (1.6–10 wt%), and oxygen fugacity (NNO to NNO+2) (Figures 3a–3c; Alonso Perez, 2007; Blatter et al., 2013; Nandedkar et al., 2014; Ulmer et al., 2018) would generate LMS. Natural rocks with robust field and petrology evidence for basalt fractional crystallization at pressures of 0.6–1.5 GPa from two classic arc sections (Jagoutz, 2010; Walker et al., 2015) are also LMS (Figure 3a). Thus, generating HMS by crystal fractionation is challenged. Basaltic-felsic magma mixing (e.g., Beier et al., 2017; Streck et al., 2007) would result in linear trends on Harker diagrams, which is inconsistent with features of our data set (Figure 3d). Significant contamination of crustal rocks, generally isotopically enriched, into the primary magma is also excluded based on limited variations of whole-rock f_k(t) values with increasing SiO₂ for individual arcs (Figure S5 in Supporting Information S1). (b) Slab melting followed by melt-peridotite interaction can produce high-Mg andesite with high La/Yb because of residual garnet in the deep source. The varying La/Yb ratios of andesite (Figure 3e) argue against a common slab origin for all HMS arcs (Defant & Drummond, 1990). In addition, slab melting requires hot slab surfaces (Peacock et al., 1994), which are also less common. (c) Proposed mantle sources of hydrous herzolite (Grove & Till, 2019) or harzburgite (Wood & Turner, 2009) are not supported by our data set. Melt derived from the metasomatized mantle for hundreds of kilometers along the arc strike have either high Ba/Th or high La/Sm but not both as indicated by LMS (Figure 2f). However, HMS have both high Ba/Th and La/Sm (Figure 2f). Mixing between high La/Sm and high Ba/Th magmas requires a large magma chamber in the lower crust so that magmas derived from distinct mantle sources can mix. However, compositional variations along the strike of the SVZ arc indicate that magma chambers in the lower crust are very small (Hildreth & Mooibath, 1988). A hybridized pyroxenite/peridotite source is inferred to be the source of high-Mg andesite-dacite with high-Ni olivine in the TMVB (Straub et al., 2011). However, forming such a source by metasomatism requires abundant slab melts (>15%) (Straub et al., 2014) derived from the cold slab surface, which is difficult.

Alternatively, numerous studies proposed that mélanges, which are formed along the subduction channel and consist of mixtures from both the subducting and overriding lithosphere, could rise as diapirs into the mantle wedge as a source for arc magmas (Castro et al., 2013; Nielsen & Marshall, 2017). Multiple observations imply the existence of mélange diapirs, including P-T paths of ultrahigh-pressure metamorphic rocks (Chatterjee & Jagoutz, 2015; Little et al., 2011), P-wave scattering from some obstacles in the mantle wedge (Lin et al., 2021), the presence of inherited zircons in arc lavas, which were not acquired by crustal assimilation (Gómez-Tuena, Cavazos-Tovar et al., 2018), and numerical modeling results, which suggest that mélange would rise as individual, tubular plumes, enabling continued mantle corner flow (Gerya & Yuen, 2003; Marschall & Schumacher, 2012).

We favor a mélange-diapir source for HMS generation because HMS compositional signatures can be well explained by experiment results, theoretical predictions, and geodynamic settings. First, HMS from individual arcs show compositional diversity and can be produced by mélange-melting experiments (Figures 3a–3d; Castro et al., 2010, 2013; Codillo et al., 2018) because of mélange heterogeneities. High Mg# values (37–91; Figure 3a) of mélange melts are caused by the involvement of peridotite. This interpretation is consistent with the calculation result implying that adding minor peridotite to low-Mg andesite-dacite melt can significantly elevate the Mg# (Figure 1g). A mélange source composed of fluid-fluxed peridotite and sediment can produce both high Ba/Th and La/Sm signatures. Second, HMS from different arcs show systematic compositional variations. For example, as average Mg# values increase for andesite from different HMS arcs, there are systematic decreases in average K₂O, La/Sm, and La/Yb, and increases in average Ba/Th and ¹⁴⁴Nd/¹⁴⁴Nd (Figure 3e and Figure S6 in Supporting Information S1). Such variations can be successfully explained by varying proportions of crustal (e.g., sediment) and mantle materials in the source as La/Sm (Labanieh et al., 2012) and La/Yb (Straub et al., 2014) are proxies of sediment/crustal rocks in the deep mantle wedge. This interpretation is also evidenced by mélange-melting experiments, which reveal that increasing the proportion of sediment in the starting material can increase K₂O and decrease Mg# of the melt (Codillo et al., 2018). Third, subduction-zone parameters provide further support (Figure S1 in Supporting Information S1). Rate, angle, and H₂O content of subducting slab (Hayes et al., 2018; van Keken et al., 2011) and crustal nature and thickness of overriding plate (Profeta et al., 2015) vary from arc to arc and are indistinguishable between LMS and HMS arcs. A marked feature of HMS arcs compared with LMS arcs is that large volumes of crustal materials are subducting, including eroded forearc crust in the Andes CVZ (Goss et al., 2013; Stern, 1991) and TMVB (Parolarri et al., 2018), the Emperor seamount in the N. Kamchatka (Nishizawa et al., 2017), and the Cocos Ridge in the SE Central America (Gazel et al., 2015, 2019). Thermodynamic models reveal that subduction of large volumes of low-density material favors mélange diapirism (Klein & Behn, 2021).
4. Implications for Continental Crust Formation

4.1. Comparing LMS and HMS With CC Compositions

Overall, igneous rocks (both LMS and HMS) from accretionary orogens and CC share similar compositions, including calc-alkaline signatures (except the fluid-modified group), enrichment in incompatible elements, and depletion in Nb, Ta, Zr, and Hf compared with Rb, Sr, Ba, and U (Figures 4a−4c; R. J. Stern, 2002). Specifically, the CC is broadly divided into the lower, middle, and upper layers, all thought to have high Mg# (Figure 2c), resulting in slowly decreasing Mg# from mafic lower crust to felsic upper crust (Hacker et al., 2015; Rudnick & Gao, 2014), akin to HMS. Furthermore, our compiled data indicate that HMS compositions vary slightly from arc to arc but fall within the estimated range of bulk CC for almost all major and trace elements (e.g., the TMVB; Figure 4a). The HMS arcs as a whole are similar in composition to CC as indicated by K₂O, Mg#, and La/Yb (Figures 4d and 4g). The sediment melt-modified group (e.g., the Andes SVZ; Figure 4b) in LMS is also similar to CC, except for Mg#, SiO₂ (Figure 2c), K₂O (Figure 4c), and La/Yb (Figure 4g). In contrast, the LMS fluid-modified group (e.g., the Izu arc) have lower incompatible trace elements, plotting outside of the estimated CC range (Figures 4c and 4f). Hence, we conclude that HMS closely resemble CC whereas LMS differ from CC.

4.2. A Two-Step Mélange-Diapir Model for CC Formation

In the plate tectonic regime, CC is extracted from the mantle primarily by subduction-related magmatism (Rudnick, 1995). However, mantle-derived melts are dominated by basalt and likely evolve further to form LMS, requiring additional processes to transform LMS arc crust to high-Mg CC. Delamination and relamination are two popular models for this transformation (Figure 1) but are not preferred because they require large mass fluxes (Hacker et al., 2015; Jagoutz & Kelemen, 2015). Furthermore, although delamination and relamination likely
occur in some arcs, they might not dominate CC formation because andesite-dacite generated by these models through crystal fractionation or magma mixing (Figure 1) are LMS rather than HMS based on our petrogenetic interpretation.

Here, we propose a magmatic mechanism (Figure 5) based on HMS petrogenesis, the similarity between HMS and CC, and inspiration from Ringwood (1974), which suggests that extracting basalt from the mantle at mid-ocean ridges followed by oceanic-crustal melting in subduction zones plays a key role in CC formation. Our model can be described as (a) extraction of basalt from the mantle, largely at arcs generating LMS, but also in oceans forming oceanic crust, (b) subduction of LMS arc crust, as well as pre-existing CC and buoyant oceanic crust (including ridges, seamounts, and plateaus), and reworking this mixture into mélangé diapirs that rise and melt in the mantle wedge to generate HMS. Both LMS and HMS can be generated by arc magmatism and destroyed by subduction and subduction erosion (Stern, 2020; Straub et al., 2020). But remelting of recycled LMS and HMS can only generate HMS, resulting in high preservation potential for HMS during long-term subduction. Such an interpretation is consistent with the data set of igneous rocks from ancient accretionary orogens, which are dominated by HMS (Figure S7 in Supporting Information S1). Consequently, the CC is essentially HMS. Considerable evidence implies that abundant Archean CC (60%–80% of present CC volume) was formed by non-plate tectonic processes, and then was recycled into the mantle or reworked within the crust through plate tectonics, resulting in minor preservation (<15% of present volume) (Cawood et al., 2018; Dhuime et al., 2018). Our proposal for generating HMS provides a long-term mechanism for transforming Archean CC into present-day high-Mg andesitic CC by remelting recycled Archean CC in the mantle wedge.

Figure 5. Cartoon showing petrogenesis of low-Mg series (LMS) and high-Mg series (HMS), and formation mechanism of continental crust (CC). Data for MORB and oceanic plateau (Caribbean-Colombian) are from Gale et al. (2013) and GEOROC, respectively, and are listed at https://doi.org/10.17605/OSF.IO/QM49Y. BCC = bulk continental crust; MORB = mid-ocean ridge basalt. Bivariate kernel density estimates include 100% of compiled data.
Acknowledgments
The authors thank editorial handling by Steven Jacobsen and constructive comments from Horst Marschall, an anonymous reviewer, and Christopher Spencer on an earlier version. This research was financially co-supported by the Chinese National Natural Science Foundation (grant nos 42121002, 91755207, 41872006, and 41902047), Second Tibetan Plateau Scientific Expedition and Research Program (STEP) (grant 2019QZKK0702), 111 project (B180408), Australian Research Council (grant FL160100168), and China Postdoctoral Science Foundation (grant 2021M690141). This is UTD Geoscience contribution #1691 and CUGB petrogen-chemical contribution PGC2015-0091.

References


