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Carbonate metasomatism in the lithospheric mantle: Implications for cratonic destruction in North China

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Abstract The activity of melts and fluids may have played a key role in inducing the destruction of the eastern North China Craton in the early Cretaceous. Carbonate melts are important agents in mantle metasomatism and can significantly modify the physical and chemical properties of the subcontinental lithospheric mantle. Carbonate metasomatism can be identified by specific geochemical indices in clinopyroxene, such as high Ca/Al and low Ti/Eu ratios. This study presents the spatial and temporal variations of carbonate metasomatism in the lithospheric mantle beneath the eastern North China Craton. Three types of carbonate metasomatism are classified based on the geochemical compositions of clinopyroxene in mantle peridotites. Clinopyroxene formed by Type 1 carbonate metasomatism is characterized by very high Ca/Al ratios (15–70) and 87 Sr/ 86 Sr ratios (0.706–0.713). Clinopyroxene derived from Type 2 carbonate metasomatism shows relatively high Ca/Al ratios (5–18) and 87 Sr/ 86 Sr ratios (0.703–0.706). However, clinopyroxene resulting from Type 3 carbonate metasomatism has low Ca/Al ratios (5– 9) and ${}^{87}Sr$ ⁸⁶Sr ratios (0.702–0.704). Deep (garnet-bearing) and shallow (spinel-bearing) lithospheric mantle beneath the Sulu orogen and surrounding areas in the eastern North China Craton were affected by intense Type 1 carbonate metasomatism before the late Triassic. The deep subduction of the South China Block with its accompanying carbonate sediments was the trigger for Type 1 carbonate metasomatism, which reduced strength of the lithospheric mantle and provided a prerequisite for the destruction of the eastern North China Craton in the early Cretaceous. After the destruction of the eastern North China Craton, the ancient relict lithospheric mantle, represented by spinel harzburgite xenoliths hosted in the late Cretaceous to Cenozoic basalts, only recorded Type 2 carbonate metasomatism. This implies that the lithospheric mantle experienced the intense Type 1 carbonate metasomatism was completely destroyed and not preserved during decratonization. Spinel lherzolite xenoliths hosted in the late Cretaceous to Cenozoic basalts represent the young, fertile lithospheric mantle formed after the cratonic destruction and only a few samples record Type 2 and 3 carbonate metasomatisms. We suggest that carbonate melts derived from the subduction-modified asthenospheric mantle with variable proportions of recycled crustal material was responsible for the Type 2 and 3 carbonate metasomatisms. The carbonate metasomatism of the lithospheric mantle beneath the Jiaodong Peninsula and surrounding areas is very pervasive and is spatially consistent with the remarkable thinning of lithospheric mantle and giant gold deposits in this region. Therefore, we conclude that carbonate metasomatism in the lithospheric mantle played a crucial part in the modification, destruction and gold deposits in the eastern North China Craton.

Keywords Cratonic destruction, North China, Carbonate metasomatism, Ca/Al in clinopyroxene, Lithospheric mantle

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1. Introduction

Archean cratons generally have thick (>200 km) lithospheric mantle keels and are characterized by low densities and low surface heat flows. They are considered to be the most stable regions on Earth and have low levels of tectonism, mag-matism, mineralization and earthquakes [\(Sleep, 2003;](#page-15-0) [Carl](#page-13-0)[son et al., 2005;](#page-13-0) [Lee et al., 2011\)](#page-14-0). The North China Craton has some characteristics in common with other Archean cratons, such as the preservation of ancient crustal remnants as old as 3.8 Ga [\(Liu et al., 1992](#page-14-1)), a stable environment from 1.9–1.8 Ga to the Paleozoic [\(Zhao et al., 2001;](#page-17-0) [Zhao and](#page-17-1) [Zhai, 2013](#page-17-1)) and a thick lithospheric mantle keel (Gao et al, 2002; [Zhang H F et al., 2008\)](#page-17-2). However, in contrast to other Archean cratons, the North China Craton experienced intense reactiviation and modification in the Mesozoic ([Chen,](#page-13-1) [1956](#page-13-1); [Zhang et al., 2003;](#page-17-3) [Gao et al., 2004](#page-13-2); [Wu et al., 2005](#page-16-0)) as shown by widespread structural deformation, magmatism, the presence of fault-bounded basins and mineralization ([Li](#page-14-2) [et al., 2012](#page-14-2); [Meng, 2003](#page-15-1); [Zhu G et al., 2012](#page-18-0), [2015](#page-18-1); [Deng and](#page-13-3) [Wang, 2016](#page-13-3))—which lead to its destruction. Although lithospheric mantle thinning is common and the destruction of the Wyoming Craton in North America has been determined to take place in the Mesoproterozoic [\(Carlson et al., 2005;](#page-13-0) [Sleep, 2005](#page-15-2); [Lee et al., 2011](#page-14-0)), the Mesozoic destruction of the North China Craton is unique and thus has important implications for global continental dynamics [\(Zhu et al.,](#page-18-2) [2012a\)](#page-18-2).

The spatio-temporal distribution, mechanisms and dynamics of the cratonic destruction in North China have been determined by systematic observations of geology and geophysics as well as studies of experimentation and theoretical modelling ([Zhu et al., 2012a\)](#page-18-2). The peak time for the destruction of the North China Craton has been constrained to occur at 130–120 Ma in the early Cretaceous ([Zhu et al.,](#page-18-3) [2012b](#page-18-3)). The destruction basically occurred in the eastern part of the North China Craton and was mainly triggered by subduction of the Paleo-Pacific plate ([Zhu et al., 2012a\)](#page-18-2); the western part of the North China Craton was not modified ([Zhu et al., 2011\)](#page-18-4). The possible physicochemical mechanisms for the destruction of the eastern North China Craton include thermo-chemical erosion [\(Menzies et al., 1993;](#page-15-3) [Xu,](#page-16-1) [2001](#page-16-1); [Zheng et al., 2001](#page-17-4)), lithospheric delamination ([Gao et](#page-13-4) [al., 2002](#page-13-4), [2004,](#page-13-2) [2008](#page-13-5), [2009](#page-13-5); [Deng et al., 2004;](#page-13-6) [Wu et al.,](#page-16-0) [2005](#page-16-0); [Xu W L et al., 2013\)](#page-16-2), melt-peridotite interactions ([Liu](#page-15-4) [et al., 2005](#page-15-4), [2008,](#page-15-5) [2010](#page-15-6); [Zhang, 2005](#page-17-5), [2009](#page-17-6)) and hydration ([Niu, 2005\)](#page-15-7). These mechanisms may have occurred at different stages or in different regions. A common signature of these mechanisms is related to diverse melt/fluid-peridotite interactions. Therefore, it is of great importance to decipher the mechanism for the destruction of the eastern North China Craton, to recognize the origin, signature and spatio-temporal distribution of the melt/fluid activity in the lithospheric mantle.

Based on the water content of clinopyroxene phenocrysts in the Mesozoic Feixian basalt (125 Ma), [Xia et al. \(2013\)](#page-16-3) calculated a high water content for the lithospheric mantle source, which may have significantly reduced the viscosity of the lithospheric mantle and provided a prerequisite for the removal of the cratonic root in eastern North China. Numerical modeling suggests that perturbation of the strongly hydrous mantle transition zone, caused by slab flattening and stagnation, could trigger cratonic destruction and removal of the lithospheric root [\(Wang Z S et al., 2016\)](#page-16-4). [Zhang \(2009\)](#page-17-6) emphasized that melt-peridotite interactions are crucial to the destruction of cratonic lithospheric mantle. However, an orthopyroxene-rich zone may be formed in the reaction front of silicate melt-peridotite interactions under dry hydrostatic conditions [\(Rapp et al., 1999](#page-15-8); [Wang et al., 2010](#page-16-5); [Zhang J F et](#page-17-7) [al., 2012](#page-17-7); [Wang C G et al., 2013](#page-16-6)) and an amphibole-rich zone may be formed under hydrous conditions ([Gervasoni et al.,](#page-13-7) [2017\)](#page-13-7). Therefore, the silicate melt-peridotite interactions are sluggish, self-restrained processes, athough these interactions can be promoted by tectonic deformation [\(Zhang J F et](#page-17-7) [al., 2012](#page-17-7)).

Compared with silicate melts, carbonate melts have a lower density and lower viscosity, but a higher reactivity, and are considered to be an effective agent for mantle metasomatism ([Green and Wallace, 1988](#page-13-8); [Ionov et al., 1993b](#page-14-3); [Yaxley et al., 1998](#page-17-8); [Blundy and Dalton, 2000;](#page-13-9) [Hammouda](#page-14-4) [and Laporte, 2000;](#page-14-4) [Grassi and Schmidt, 2011](#page-13-10); [Sokol et al.,](#page-15-9) [2016;](#page-15-9) [Gervasoni et al., 2017\)](#page-13-7). The experimental results of [Wang C et al. \(2016\)](#page-16-7) indicate that carbonate melts derived from a subducted slab can significantly modify and destroy cratonic lithospheric mantle. Slab subduction may also carry carbonate sediments into the deep mantle, with only a limited amount of carbonate being released during decarbonation at shallow depths [\(Yaxley and Green, 1994;](#page-17-9) [Molina and Poli,](#page-15-10) [2000;](#page-15-10) [Kerrick and Connolly, 2001](#page-14-5); [Dasgupta et al., 2005](#page-13-11); [Liu](#page-15-11) [Y S et al., 2015;](#page-15-11) [Chen C F et al., 2016\)](#page-13-12). Therefore, large amounts of carbonate sediment remnants may be stored in the deep mantle or transformed into carbonate melts by partial melting ([Wallace and Green, 1988;](#page-16-8) [Dasgupta et al.,](#page-13-11) [2005\)](#page-13-11). Since the Paleozoic, the southward, northward and westward subductions of the Paleo-Asian plate, the Paleo-Tethyan plate and the Paleo-Pacific plate, respectively, would have resulted in multiple modifications of the North China Craton over long periods of time ([Windley et al.,](#page-16-9) [2010\)](#page-16-9). These subducted slabs may also have carried large amounts of carbonate sediments into the deep mantle [\(Liu Y](#page-15-11) [S et al., 2015;](#page-15-11) [Chen C F et al., 2016;](#page-13-12) [Zhang et al., 2017](#page-17-10)), inducing extensive carbonate metasomatism of the lithospheric mantle and providing a prerequisite for the destruction of the North China Craton. We provide here a systematic summary of the spatio-temporal variations of carbonate metasomatism in the lithospheric mantle beneath the eastern

North China Craton and try to establish a possible link between carbonate metasomatism and cratonic destruction.

2. Occurrence of lithospheric mantle samples in the eastern North China Craton

Numerous mantle peridotite xenoliths have been found in Paleozoic kimberlites and Jurassic-Quaternary volcanic rocks in the eastern North China Craton [\(Figure 1\)](#page-2-0). Some mantle-derived peridotite terranes also occur in the Triassic Sulu ultrahigh pressure (UHP) belt [\(Figure 1](#page-2-0)). These mantle peridotites can be used to constrain the evolution of the lithospheric mantle before and after the destruction of the eastern North China Craton [\(Zheng, 2009](#page-17-11)). The Paleozoic mantle peridotite xenoliths hosted in the Mengyin, Fuxian and Tieling kimberlites [\(Gao et al., 2002](#page-13-4); [Wu et al., 2006;](#page-16-10) [Zheng et al., 2006,](#page-17-12) [2007\)](#page-17-13) and the Triassic Sulu orogenic mantle peridotite terranes [\(Zheng et al., 2005b](#page-17-14), [2006;](#page-17-12) [Yuan et](#page-17-15) [al., 2007;](#page-17-15) [Zhang Z M et al., 2011;](#page-17-16) [Su et al., 2016\)](#page-15-12) have

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undergone strong serpentinization, which makes it difficult to determine their protoliths, although they are all ancient and refractory with Re-depletion Os model ages (T_{RD}) of 3.2–1.5 Ga ([Figure 2](#page-3-0)). The Jurassic Xinyang and early Cretaceous Tietonggou mantle peridotite xenoliths consist of refractory harzburgites and dunites [\(Zheng et al., 2005a](#page-17-17); [Xu](#page-16-11) [et al., 2008](#page-16-11)) and represent ancient lithospheric mantle before the destruction of the North China Craton. The 4 Ma Hebi mantle peridotite xenoliths are dominated by harzburgites with $T_{\rm RD}$ ages of 3.0–1.8 Ga and are also considered as representative of the ancient and refractory lithospheric mantle beneath the North China Craton ([Zheng et al., 2001](#page-17-4), [2007](#page-17-13); [Sun et al., 2012\)](#page-16-12).

The mantle peridotite xenoliths hosted in the late Cretaceous Fuxin (~100 Ma), Qindao Pishikou (86–78 Ma) and Junan (67 Ma) basalts, the Neogene Shanwang (18 Ma), Changle (19 Ma), Qixia (18–4 Ma) and Penglai (6–4 Ma) basalts and the Quaternary Huinan $(0.6 Ma)$, Kuandian $(0.6 Ma) and Nushan (0.7 Ma) basalts, which are formed$ after the destruction of the eastern North China Craton and

> Huinar $0.6 M$

Kundian

<0.6 Ma

Changchun

Tieling
470 Ma? Shenyang

Euvir

100 Ma

Xing-Meng Orogenic Belt

Zhangjiako

[Figure 1](#page-2-0) Distribution of mantle-derived peridotite xenoliths and terranes in the eastern North China Craton.

[Figure 2](#page-3-0) Cumulative probability plot of Re-depletion Os model ages (T_{RD}) of the Paleozoic-Cenozoic lithospheric mantle xenoliths and terranes in the eastern North China Craton. Data are from [Gao et al. \(2002\)](#page-13-4), [Wu et](#page-16-10) [al., \(2006\),](#page-16-10) [Yuan et al. \(2007\)](#page-17-15), Zheng et al. ([2007\)](#page-17-13), [Chu et al. \(2009\),](#page-13-15) [Yang](#page-17-18) [et al. \(2010\),](#page-17-18) [Sun et al. \(2012\)](#page-16-12) and [Su et al. \(2016\).](#page-15-12)

are dominated by fertile lherzolites with a few refractory harzburgites ([Figure 1](#page-2-0)). These lherzolites have much younger T_{RD} ages than the harzburgites ([Figure 2\)](#page-3-0). We therefore conclude that the lherzolites in the eastern North China Craton represent new, fertile lithospheric mantle and the harzburgite xenoliths represent ancient, refractory mantle. Mantle peridotite xenoliths hosted in Triassic (223 Ma) kimberlites and Neogene (6.6 Ma) basalts from North Korea have previously been suggested to represent new and fertile lithospheric mantle, athough no petrological classification was available ([Yang et al., 2010](#page-17-18)).

3. Geochemical indices of cryptic carbonate metasomatism in the lithospheric mantle

Mantle-derived peridotite xenoliths in volcanic rocks and terranes in orogenic belts provide direct samples of the lithospheric mantle, giving an important window with which to observe mantle metasomatism ([Bodinier et al., 2003](#page-13-13); [Pearson et al., 2003](#page-15-13)). Most mantle metasomatism is cryptic metasomatism describing changes in composition of preexisting minerals without the formation of new phase. Therefore, some special geochemical indices in minerals are useful to decipher cryptic mantle metasomatism of the lithospheric mantle. By combining elemental partitioning coefficients between carbonate melts and silicate minerals with the elemental signatures of carbonatites, [Rudnick et al.](#page-15-14) [\(1993\)](#page-15-14) suggested that high La/Yb, Nb/La, Ca/Al and Zr/Hf ratios and low Ti/Eu ratios are effective geochemical indices for tracing carbonate metasomatism in the mantle. [Klemme](#page-14-6) [et al. \(1995\)](#page-14-6) reported that Ti/Eu is more sensitive to carbo-nate metasomatism. [Coltorti et al. \(1999\)](#page-13-14) used $(La/Yb)_N$ and Ti/Eu ratios in clinopyroxene to distinguish carbonate metasomatism from silicate metasomatism ([Figure 3a](#page-3-1)). On the basis of published geochemical data at that time, [Coltorti et](#page-13-14)

[Figure 3](#page-3-1) Variations of (a) $(LaYb)$ _N with Ti/Eu (modified from [Coltorti et al. \(1999\)\)](#page-13-14) and (b) Ca/Al with Mg# in clinopyroxene from natural and experimental mantle peridotite. Experimental data for the partial melting of carbonate-free peridotite are from [Walter \(1998\),](#page-16-13) [Gaetani and Grove \(1998\),](#page-13-16) [Pickering-Witter and Johnston \(2000\),](#page-15-15) [Falloon et al. \(1999\)](#page-13-17), [Baker and Stolper \(1994\)](#page-12-0), [Wasylenki et al. \(2003\)](#page-16-14) and [Schwab and Johnston \(2001\)](#page-15-16) (only experiments with lherzolite and harzburgite residue were selected), the data for high-silicate melt-peridotite interactions are from [Wang et al. \(2010\)](#page-16-5) and [Yaxley and Green \(1998\)](#page-17-19) and the data for carbonate melt-peridotite interactions are from [Dalton and Wood \(1993\),](#page-13-18) [Brey et al. \(2008\),](#page-13-19) [Klemme et al. \(1995\),](#page-14-6) [Sokol et al. \(2016\)](#page-15-9) and [Gervasoni et al. \(2017\)](#page-13-7). The data for typical carbonate metasomatism in nature are from [Coltorti et al. \(1999\)](#page-13-14), [Yaxley et al. \(1998\)](#page-17-8) and [Neumann et al. \(2002\)](#page-15-17).

[al. \(1999\)](#page-13-14) suggested that the clinopyroxene that results from carbonate metasomatism is generally characterized by (La/ Yb _N>3–4 and Ti/Eu<1500, although these are not absolute values. [Figure 3](#page-3-1) shows that some clinopyroxenes resulting from typical carbonate metasomatism have Ti/Eu rstios close to 2000 and $(La/Yb)_N$ values \leq 3. We therefore suggest that the trends of $(La/Yb)_N$ and Ti/Eu in clinopyroxene may be used to distinguish carbonate metasomatism from silicate meta-somatism [\(Figure 3a](#page-3-1)). However, despite the rapid development of *in situ* analyses that could be used to apply this trace element index to trace carbonate metasomatism in the lithospheric mantle [\(Zheng et al., 2007;](#page-17-13) [Sun et al., 2012;](#page-16-12) Deng et al, 2017), there is only a limited amount of published data on the trace elemental contents of minerals from the North China Craton. This has hindered the systematic spatio-temporal assessment of carbonate metasomatism in the lithospheric mantle in this region.

Carbonate melts have very high Ca/Al ratios [\(Rudnick et](#page-15-14) [al., 1993](#page-15-14)) and both Ca and Al are stoichiometric elements of clinopyroxene. Thus, the Ca/Al ratio of clinopyroxene can be considered as an effective major elemental index to trace carbonate metasomatism in the lithospheric mantle. [Figure](#page-3-1) [3b](#page-3-1) shows the variation of the major element (Ca/Al and Mg#) composition of clinopyroxene in natural and experimental mantle peridotites. Clinopyroxene in lherzolite and harzburgite after partial melting ([Baker and Stolper, 1994;](#page-12-0) [Gaetani and Grove, 1998;](#page-13-16) [Walter, 1998](#page-16-13); [Falloon et al., 1999;](#page-13-17) [Pickering-Witter and Johnston, 2000;](#page-15-15) [Schwab and Johnston,](#page-15-16) [2001](#page-15-16); [Wasylenki et al., 2003\)](#page-16-14) and the interactions between experimental eclogite-derived high-silicate melts and peridotites [\(Wang et al., 2010](#page-16-5); [Yaxley and Green, 1998\)](#page-17-19) show the low Ca/Al ratios <5 and Mg# values <92 ([Figure 3b\)](#page-3-1). However, most of the Ca/Al ratios are >5 in clinopyroxene formed during experimental carbonate melt-peridotite interactions ([Figure 3b\)](#page-3-1) ([Dalton and Wood, 1993;](#page-13-18) [Klemme et](#page-14-6) [al., 1995](#page-14-6); [Brey et al., 2008;](#page-13-19) [Sokol et al., 2016;](#page-15-9) [Gervasoni et](#page-13-7) [al., 2017\)](#page-13-7). All the clinopyroxenes resulting from natural carbonate metasomatism show high Ca/Al ratios >5 [\(Figure](#page-3-1) $3b$). We therefore conclude that a Ca/Al ratio >5 is an important geochemical index with which to trace carbonate metasomatism in the lithospheric mantle. Major element contents, including Ca and Al, are commonly reported in mantle clinopyroxenes from the North China Craton. Clinopyroxene is also the main carrier of trace elements and has very high Sr and very low Rb contents, making it an ideal mineral for *in situ* Sr isotope analysis [\(Malarkey et al., 2011;](#page-15-18) [Tong et al., 2016](#page-16-15)). The very low Rb/Sr ratio means the neglected radiogenic Sr in clinopyroxene that can be used to record the initial Sr isotopic composition in order to constrain the metasomatic agent. We therefore used a comprehensive combination of major element (Ca/Al and Mg#), trace element (Ti/Eu and $(La/Yb)_N$) indices and Sr isotopic compositions of clinopyroxene (Figures 4–[6\)](#page-7-0) to study the

spatio-temporal variations of carbonate metasomatism in the lithospheric mantle beneath the eastern North China Craton.

4. Widespread carbonate metasomatism in the lithospheric mantle beneath the North China Craton

The existence of carbonate melt in the deep mantle is a prerequisite for carbonate metasomatism in the lithospheric mantle. Carbonate melt may be erupted at the surface as carbonatite or may be completely exhausted during interactions with peridotite. Proterozoic and Mesozoic carbonatites are exposed at more than ten locations in the North China Craton. The Meso-Proterozoic Bayan Obo (1.3 Ga), Fengzhen (1.8–1.7 Ga), Huaian (1.8 Ga) and Fangcheng (0.79 Ga) carbonatites have depleted Sr-Nd isotopic compositions $({}^{87}Sr/{}^{86}Sr_{(t)}=0.7028-0.7031$; $\varepsilon_{Nd(t)}=-1.2$ to -6.1). However, the Mesozoic Zhuolu (239 Ma), Songxian (209 Ma), Huairen (229 Ma), Linxian (132 Ma), Huayin (205 Ma), Luonan (221 Ma) and Laiwu (124 Ma) carbonatites show evolved Sr-Nd compositions (${}^{87}Sr/{}^{86}Sr_{(t)}=0.7043-0.7106$; $\varepsilon_{Nd(t)}=-2.1$ to −18.2) [\(Yan et al., 2007\)](#page-16-16), indicating that these carbonatites were derived from enriched mantle sources or recycled crust. [Kim et al. \(2016\)](#page-14-7) suggested that the evolved Sr-Nd isotopic compositions (${}^{87}Sr/{}^{86}Sr_{(t)}=0.703-0.706$; $\varepsilon_{Nd(t)}=-26$) of the Mesozoic Hongcheon carbonatite $(\sim 230$ Ma) in the South Korea is a result of an ancient enrichment event in the lithospheric mantle.

[Ying et al. \(2004\)](#page-17-20) proposed that the enriched signature of the Mesozoic (124 Ma) Laiwu-Zibo carbonatites is related to the subduction, with continental material, of the South China Block into the deep mantle beneath the North China Craton. [Chen C F et al. \(2016\)](#page-13-12) reported that the carbonatite intrusion in the late Tertiary Hannuoba basalt has the geochemical characteristics of carbonate sediments, which could be the result of subduction of the Paleo-Asian oceanic plate. [Zhang](#page-17-10) [et al. \(2017\)](#page-17-10) found the same carbonate melt from the Miocene basalts in the Dongbahao region (Inner Mongolia) along the northern margin of the North China Craton, where two-stage carbonate metasomatism related to the subduction of the Paleo-Asian oceanic plate was proposed for xenoliths of mantle peridotites [\(Chen et al., 2017;](#page-13-20) [Wu et al., 2017](#page-16-17)). The Dalihu carbonatite in the Central Asian Orogenic Belt (CAOB) has a similar trace element and oxygen isotopic $(\delta^{18}O_{\text{SMOW}}=20.7-21.5)$ composition to sedimentary carbonates and contains microscopic diamonds; this is the first direct evidence of sedimentary carbonate recycling during subduction of the Paleo-Asian oceanic plate [\(Liu Y S et al.,](#page-15-11) [2015\)](#page-15-11). Thus, the presence of widespread subduction-related carbonate melts suggests the possibility of intense carbonate metasomatism in the lithospheric mantle beneath the North China Craton.

[Figure 4](#page-5-0) (a) Petrological classification of harzburgite xenoliths and terranes, variations of the ((b), (e)) Ca/Al ratio with Mg# and ((c), (f)) the (La/Yb)_N with the Ti/Eu in clinopyroxene, and (d) variation of the Ca/Al of clinopyroxene with time in the eastern North China Craton. Except for the strongly serpentinized Mengyin and Xinyang peridotite xenoliths and the Sulu orogenic peridotite terrane, detailed mineral counting work has been carried out on the other harzburgites. The Kuandian data are from [Xu et al. \(2013b\);](#page-16-18) the Huinan data are from [Xu et al. \(2003b\);](#page-16-19) the Hebi data are from [Zheng et al. \(2001\);](#page-17-4) the Penglai data are from [Chu et al. \(2009\);](#page-13-15) the Qixia data are from [Zheng et al. \(1998\);](#page-17-4) the Pishikou data of the Qingdao are from [Zhang \(2009\);](#page-17-6) the Xinyang data are from [Zheng et al. \(2005a\);](#page-17-17) the Tietonggou data are from [Xu et al. \(2008\)](#page-16-11) and [Zhou et al. \(2013\);](#page-18-5) the Lijiatun date are from [Su et al. \(2016\)](#page-15-12); the Zhimafang, Xugou and Jiangzhuang data in the Donghai are from [Yang and Jahn \(2000\),](#page-17-21) Zheng et al. ([2005b,](#page-17-14) [2006\)](#page-17-12), [Yuan et al. \(2007\)](#page-17-15) and [Zhang Z M et al.](#page-17-16) [\(2011\)](#page-17-16). Data for the South Africa Craton are from [Boyd et al. \(2004\),](#page-13-21) [Franz et al. \(1996\),](#page-13-22) Gregoire ([2003,](#page-13-23) 2005), [Saltzer \(2001\),](#page-15-19) [Simon et al. \(2003\),](#page-15-20) [Stiefenhofer et al. \(1997\)](#page-15-21) and [Whitehead et al. \(2002\);](#page-16-20) data for the Greenland Craton are from [Bernstein et al. \(1998\)](#page-12-1), [Bizzarro and Stevenson \(2003\),](#page-13-24) [Hellebrand and Snow \(2003\)](#page-14-8) and [Holm and Prægel\(2006\)](#page-14-9); data for the Slave Craton are from [Aulbach et al. \(2004\),](#page-12-2) [Kopylova and Caro \(2004\),](#page-14-10) [McCammon](#page-15-22) [and Kopylova \(2004\)](#page-15-22), [Menzies et al. \(2004\),](#page-15-23) [van Achterbergh et al. \(2004\)](#page-16-21), [Kopylova et al. \(1999\),](#page-14-11) [MacKenzie and Canil \(1999\)](#page-15-24), [Schmidberger and Francis](#page-15-25) [\(1999\),](#page-15-25) [Shi et al. \(1998\)](#page-15-26), [Schmidberger et al. \(2003\),](#page-15-27) [Schmidberger and Francis \(2001\)](#page-15-28), [Peslier \(2002\)](#page-15-29) and [Griffin et al. \(1999\);](#page-14-12) data for the Siberian Craton are from Ionov et al. [\(1993a,](#page-14-13) [2005a,](#page-14-14) [2005b,](#page-14-15) [2005c,](#page-14-16) [2006](#page-14-17)), [Ionov and Hofmann \(1995\),](#page-14-18) [Griffin et al. \(1996\)](#page-14-19), [Boyd et al. \(1997\),](#page-13-25) [Litasov et al. \(2000\),](#page-14-20) [Ionov](#page-14-21) [\(2004\),](#page-14-21) [Griffin et al. \(2005\)](#page-14-22) and [Sharygin et al. \(2007\)](#page-15-30); data for the Australia Craton are from [Ghorbani and Middlemost \(2000\)](#page-13-26), [Bruce et al. \(2000\)](#page-13-27), [Powell et](#page-15-31) [al. \(2004\),](#page-15-31) [Roach \(2004\),](#page-15-32) [Woodland et al. \(2004\)](#page-16-22), Yaxley et al. ([1997,](#page-17-22) [1998](#page-17-8)), [Eggins et al. \(1998\),](#page-13-28) [Varela et al. \(1999\)](#page-16-23) and [Yaxley and Kamenetsky\(1999\);](#page-17-23) data for the Wyoming Craton are from [Downes \(2004\)](#page-13-29) and [Carter Hearn \(2004\);](#page-13-30) data for the Tanzania Craton are from [Dawson \(2002\)](#page-13-31) and [Lee et al. \(2000\).](#page-14-23)

[Figure 5](#page-6-0) ((a), (d)) Petrological classification of the late Cretaceous to Quaternary lherzolite/wehrlite xenoliths. ((b), (e)) Variations of the Ca/Al ratio with Mg# and ((c), (f)) the (La/Yb)_N ratio with the Ti/Eu in clinopyroxene of the eastern North China Craton. The Fuxin data are from [Zheng et al. \(2007\)](#page-17-13); the Qixia data are from [Zheng et al. \(1998\)](#page-17-4) and [Xia et al. \(2010\);](#page-16-24) the Changle data are from [Xiao et al. \(2010\)](#page-16-25) and [Deng et al. \(2017\)](#page-13-32); the Pishikou data of the Qingdao are from [Zhang \(2009\);](#page-17-6) the Junan data are from [Ying et al. \(2006\)](#page-17-25) and [Zhang \(2009\);](#page-17-6) the Penglai data are from [Chu et al. \(2009\)](#page-13-15) and [Xia et al.](#page-16-24) [\(2010\);](#page-16-24) the Kuandian data are from Xu R et al. [\(2013a,](#page-16-26) [2013b\)](#page-16-18); the Huinan data are from Xu et al. [\(2003a,](#page-16-27) 2003b); the Nushan data are from [Xu et al. \(1998\)](#page-16-28) and [Xu et al. \(2000\)](#page-16-29); the Shanwang data are from [Zheng et al. \(1998, 2006\)](#page-17-4) and [Chu et al. \(2009\)](#page-13-15); Data for the peridotite xenoliths in the North Korea are from [Yang et al. \(2010\)](#page-17-18).

The elemental and isotopic compositions of mantle-derived mafic and ultramafic rocks can be used to indirectly constrain their mantle sources [\(Hofmann, 1997](#page-14-24)). Mesozoic to Cenozoic basalts are widely distributed in the North China Craton [\(Liu et al., 2008;](#page-15-5) [Zheng et al., 2018](#page-18-6)). Based on a detailed elemental study, [Zeng et al. \(2010\)](#page-17-24) proposed that the mantle source of the Cenozoic alkaline basalts in the Shandong Province was dominated by carbonated peridotite. Melt inclusions in the olivine phenocrysts of alkaline basalts from Yangzhuang in the Shandong Province contain methane

[Figure 6](#page-7-0) ((a), (b)) Variations of the Ca/Al ratio with Sr isotopic composition in clinopyroxene of the peridotite xenoliths from the eastern North China Craton. ((c), (d)) Variations of Sr isotopic composition with the Ti/Eu and Ca/Al ratios of clinopyroxene in the Kuandian lherzolite xenolith. (e) Variations of Sr isotopic composition with the Ca/Al ratio in clinopyroxene of the Shanwang lherzolite xenolith. (f) Sr isotopic profile of clinopyroxene in the Changle Type 2 lherzolite xenolith. Data for the Donghai orogenic peridotite terrane are from [Yang and Jahn \(2000\);](#page-17-21) the Tietonggou data are from [Xu et al. \(2008\)](#page-16-11) and [Zhou et al. \(2013\)](#page-18-5); Hebi data are from [Sun et al. \(2012\)](#page-16-12); the Penglai data are from [Chu et al. \(2009\);](#page-13-15) the Huinan data are from Xu et al. (2003b); the Pishikou data of the Qingdao are from Zhang et al. (2009); the Changle data are from [Xiao et al. \(2010\)](#page-16-25) and [Deng et al. \(2017\)](#page-13-32); the Junan data are from Zhang (2009); the Kuandian data are from [Xu et al. \(2013a\);](#page-16-26) the Nushan data are from [Xu et al. \(2000\)](#page-16-29); the Shanwang data are from [Chu et al. \(2009\);](#page-13-15) Data for the North Korea are from [Yang et al. \(2010\).](#page-17-18)

 $(CH₄)$ and other hydrocarbons and some carbonaceous minerals such as graphite and carbonate, indicating a carbonated mantle source [\(Liu F et al., 2015\)](#page-14-25). Late Cretaceous to Cenozoic basalts in eastern China are characterized by light Mg isotope and heavy Zn isotope compositions, which has been suggested to be a result of the westward subduction of the Pacific oceanic plate and the recycling of carbonate sediments into the mantle [\(Yang W et al., 2012](#page-17-26); [Liu et al., 2016;](#page-14-26)

[Li et al., 2017](#page-14-27)).

Triassic low-Si alkaline rocks occur widely along the northern margin of the North China Craton and are considered to represent a low degree of partial melting of metasomatosed and enriched mantle [\(Yan et al., 2001](#page-16-30); [Niu et al.,](#page-15-33) [2012;](#page-15-33) [Yang J H et al., 2012](#page-17-27); [Zhang S H et al., 2012](#page-17-28)). These alkaline rocks show a crustal elemental and Sr-Nd isotopic signature that is different in comparism to the asthenospheric

mantle. Thus, the metasomatic melt/fluid is contributed to recycling of the crust ([Yan et al., 2002;](#page-16-31) [Zhu et al., 2017\)](#page-18-7). The presence of carbonate veins and intergranular calcite in the Fanshan alkaline complex suggest that the original magma contained certain amounts of $CO₂$ ([Yan et al., 2007;](#page-16-16) [Niu et](#page-15-33) [al., 2012](#page-15-33)). The very high Ca content in the early crystallized clinopyroxene is consistent with Ca enrichment in the original magma, which could be derived from a carbonated mantle source ([Niu et al., 2012](#page-15-33); [Hou et al., 2015\)](#page-14-28) resulting from subduction of the Paleo-Asian oceanic plate [\(Niu et al.,](#page-15-34) [2017](#page-15-34)).

Carbonate minerals such as calcite, dolomite and magnesite can be formed during carbonate metasomatism [\(Yaxley](#page-17-29) [et al., 1991;](#page-17-29) [Ionov et al., 1993b](#page-14-3), [1996](#page-14-29); [Rudnick et al., 1993\)](#page-15-14). These index minerals can therefore be used directly to identify carbonate metasomatism. Intergranular calcite grains have been found in peridotite xenoliths from the Changle basalts in the eastern North China Craton ([Xiao et](#page-16-25) [al., 2010](#page-16-25); [Deng et al., 2017](#page-13-32)) and the Datong basalts in the Trans-North China Orogen (TNCO) ([Wang C Y et al., 2016\)](#page-16-32). Carbonate melt inclusions hosted in olivine and clinopyroxene were also present in the Changle peridotite xenolith ([Deng et al., 2017\)](#page-13-32). Carbonate metasomatism is always accompanied by the formation of secondary clinopyroxene ([Ionov et al., 1993b;](#page-14-3) [Coltorti et al., 1999](#page-13-14); [Sun et al., 2012\)](#page-16-12), which is formed during the reaction of carbonate melts with orthopyroxene ([Dalton and Wood, 1993\)](#page-13-18). Wehrlites composed of olivine and clinopyroxene are therefore considered to be the end-product of carbonate metasomatism in the lithospheric mantle ([Yaxley et al., 1991](#page-17-29), [1998](#page-17-8); [Rudnick et al.,](#page-15-14) [1993](#page-15-14); [Neumann et al., 2002\)](#page-15-17). Wehrlite samples have been reported in Cenozoic Changle peridotite xenoliths [\(Xiao et](#page-16-25) [al., 2010](#page-16-25)) and early Cretaceous high-Mg diorites from the western Shandong Province ([Zhou et al., 2013](#page-18-5)) in the eastern North China Craton.

Orogenic mantle peridotite terranes in the Triassic Sulu orogenic belt are generally considered to be fragments of the lithospheric mantle of the North China Craton that have been carried to crustal depths during the exhumation of the deeply subducted South China Block [\(Zheng et al., 2006;](#page-17-12) [Zhang R Y](#page-17-30) [et al., 2008](#page-17-30), [Zheng Z M et al., 2011;](#page-17-16) [Su et al., 2016\)](#page-15-12). Magnesite and dolomite are common accessory minerals in the Donghai garnet-bearing mantle peridotite in the southern Sulu orogenic belt, in which ~225 Ma metamorphic zircon grains contain magnesite and $CO₂$ fluid inclusions [\(Zheng Z](#page-17-16) [M et al., 2011\)](#page-17-16). In the northern Sulu orogenic belt, the Lijiatun spinel-bearing mantle peridotite includes intergranular dolomite, in which (clinopyroxene+olivine) veins with wehrlite mineral assemblage crosscutting orthopyroxene porphyroblasts are considered as the direct evidence of car-bonate metasomatism ([Su et al., 2016](#page-15-12)). Based on the geochemical indices $(La/Yb)_N$ and Ti/Eu of clinopyroxene, [Zheng et al. \(2007\)](#page-17-13) suggested that the lithospheric mantle beneath the North China Craton experienced carbonate metasomatism in the Paleozoic and early Mesozoic, but silicate metasomatism in the late Mesozoic and Cenozoic. In this study, however, we showed that three different types of carbonate metasomatism with spatio-termporal variations can be identified in the Paleozoic to Cenozoic lithospheric mantle beneath the eastern North China Craton on the basis of major element (Ca/Al and Mg#) and trace element (Ti/Eu and $(La/Yb)_{N}$ indices and the Sr isotope composition of mantle clinopyroxene (Figures 4–[6\)](#page-7-0).

5. Temporal variations of carbonate metasomatism in the lithospheric mantle beneath the eastern North China Craton

Carbonate metasomatism characterized by the high Ca/Al, $Mg#$ and $(La/Yb)_N$ and low Ti/Eu ratios of clinopyroxene is recorded by harzburgite xenoliths in the volcanic rocks, orogenic mantle peridotites in the Triassic Sulu UHP belt and mantle peridotite xenoliths and xenocrysts in the Paleozoic kimberlites of the eastern North China Craton, although not in the Huinan harzburgite xenoliths ([Figure 4\)](#page-5-0). The high Ti/ Eu ratios in the clinopyroxenes of the Huinan harzburgite [\(Figure 4f\)](#page-5-0) are results of the interactions of basaltic melts and peridotites ([Xu et al., 2003a](#page-16-27), [2003b](#page-16-19)). However, the slightly high Ca/Al ratio of 4–8 in clinopyroxene is consistent with a carbonate overprint or a small amount of metasomatims by a volatile-rich melt [\(Xu et al., 2003a](#page-16-27)). This further indicates that a comprehensive consideration of the major (e.g., Ca/Al) and trace (e.g., Ti/Eu) element geochemical indices in clinopyroxene is effective in tracing carbonate metasomatism in the lithospheric mantle.

Clinopyroxene in the Triassic Sulu orogenic peridotites has a very high Ca/Al ratio (median 29), which is defined as Type 1 carbonate metasomatism, whereas the median Ca/Al ratios of clinopyroxene in the Paleozoic Mengyin peridotite xenoliths and the global cratonic lithospheric mantle are 17 and 10, respectively [\(Figure 4b\)](#page-5-0). Apart from the early Cretaceous Tietonggou harzburgites and wehrlites with records of Type 1 carbonate metasomatism, the Ca/Al ratio in the clinopyroxene of harzburgite xenoliths hosted in the Jurassic to Cenozoic volcanic rocks decreases to 5–18, indicative of Type 2 carbonate metasomatism ([Figure 4e](#page-5-0)). However, based on the geochemical signature of natural carbonatites ([Rud](#page-15-14)[nick et al., 1993\)](#page-15-14) and the elemental partitioning of experimental carbonate melts [\(Klemme et al., 1995\)](#page-14-6), mantlederived magma can inherit the geochemical indices of a carbonated mantle source, such as high Ca/Al and $(La/Yb)_N$ and low Ti/Eu ratios ([Zeng et al., 2010](#page-17-24)). The Ca/Al and (La/ Yb _N ratios are higher and the Ti/Eu ratio is lower in the Triassic mafic rocks of the Jiaodong Peninsula than those of the Cretaceous and Cenozoic mafic rocks of the eastern

[Figure 7](#page-9-0) (a) Variations of the Ca/Al ratio with the Ti/Eu ratio and (b) the (La/Yb) _N with the Ti/Eu ratio in Mesozoic-Cenozoic mafic rocks from the eastern North China Craton. Data for the eastern North China Craton are from [Dai et al. \(2016\),](#page-13-33) [Liu et al. \(2008\)](#page-15-5) and [Zeng et al. \(2010\).](#page-17-24) The mid-ocean ridge basalt (MORB) data are from [Gale et al. \(2013\)](#page-13-34); the oceanic island basalt (OIB) data are from <http://georoc.mpch-mainz.gwdg.de/georoc/>. Only MgO >7.5% data are used in this study.

North China Craton ([Figure 7\)](#page-9-0). Therefore, we conclude that carbonate metasomatism in the eastern North China Craton was transformed from Type 1 to Type 2 during the Triassic period ([Figure 4d\)](#page-5-0), which is consistent with Type 1 and Type 2 carbonate metasomatisms showing the striking $87\text{Sr}/86\text{Sr}$ composition of clinopyroxene of 0.706–0.713 and 0.703– 0.706, respectively ([Figure 6a\)](#page-7-0).

After the peak decratonization of the eastern North China Craton, the clinopyroxene of the 100 Ma Fuxin peridotite xenolith and some Tertiary Changle lherzolite xenoliths were characterized by the relatively high Ca/Al ratios (>5) , but low Mg# signatures. This is defined as Type 3 carbonate metasomatism [\(Figure 5b](#page-6-0)). Type 3 carbonate metasomatism also displays by medium Ti/Eu ratios $(\sim 1000-3000)$ and slightly high $(La/Yb)_{N}$ ratios in clinopyroxene and plots in the transitional domain between typical carbonate metasomatism and silicate metasomatism ([Figure 5c](#page-6-0)).

6. Spatial variations of carbonate metasomatism in the lithospheric mantle beneath the eastern North China Craton

Partial melting of the upwelling asthenosphere generally produces silicate-rich melts that induce silicate metasomatism and form juvenile lithospheric mantle during thinning of the lithospheric mantle ([Xu et al., 2003b](#page-16-19)). However, the juvenile lithospheric mantle in the eastern North China Craton is overprinted by carbonate metasomatism with obvious spatial variations. Lherzolite xenoliths with Type 2 and Type 3 carbonate metasomatism are mainly found in the Jiaodong Peninsula, whereas silicate metasomatism is seen far from the Jiaodong Peninsula in the eastern North China Craton. Clinopyroxenes in some lherzolite xenoliths from the Qixia, Penglai, Qingdao, Changle (Type 1 lherzolite) and Junan in the Jiaodong Peninsula and surrounding areas show element signatures of Type 2 carbonate metasomatism (high Ca/Al ratios and Mg#; low Ti/Eu ratios) [\(Figure 5b](#page-6-0) and c). These lherzolites show the same Sr isotopic composition as the harzburgites that experienced Type 2 carbonate metasomatism, with ${}^{87}Sr/{}^{86}Sr$ ratios of 0.703–0.705 and 0.703–0.706, respectively ([Figure 6b\)](#page-7-0). Type 3 carbonate metasomatism has only been recorded by a few lherzolite xenoliths ([Figure](#page-6-0) [5e](#page-6-0) and f) and the ${}^{87}Sr/{}^{86}Sr$ ratio shows a negative relationship with the Ca/Al in clinopyroxene for most lherzolite xenoliths [\(Figure 6e\)](#page-7-0) from the Shanwang, indicating that the carbonate metasomatic agent may have been derived from very depleted asthenospheric mantle $({}^{87}Sr/{}^{86}Sr< 0.7023)$. Such Type 3 carbonate metasomatism with a very low $87\text{Sr}/86\text{Sr}$ ratio has also been reported in the lherzolite xenoliths from the nearby Changle ([Deng et al., 2017\)](#page-13-32) (Figures 5b, [6b](#page-7-0) and f). However, our unpublished data show that the Qingdao harzburgite and lherzolite xenoliths altered by Type 3 carbonate metasomatism have a relatively high ${}^{87}Sr/{}^{86}Sr$ ratio of 0.7033–0.7040 in clinopyroxene with a Ca/Al ratio of $4.1-6.0$ ([Figure 6b](#page-7-0)). The Cenozoic lherzolite xenoliths from the Kuandian and Nushan far away from the Jiaodong Peninsula, mainly experienced silicate metasomatism, with very weak or absent carbonate metasomatism [\(Figure 5e](#page-6-0) and f). Most of the lherzolite xenoliths from the Kuandian show a clear positive relationship between the ${}^{87}Sr/{}^{86}Sr$ ratios (0.7028–0.7042) and Ti/Eu ratios in clinopyroxene [\(Figure 6c](#page-7-0)), which is consistent with silicate metasomatism ([Xu R et al., 2013a](#page-16-26)). However, a few lherzolite xenoliths have slightly higher ${}^{87}Sr/{}^{86}Sr$ (0.7041– 0.7055) and Ca/Al ratios ([Figure 6d\)](#page-7-0), very high Ti/Eu ratios (>4000) and no relationship between the Ti/Eu with $87\text{Sr}/86\text{Sr}$ ratios in clinopyroxene [\(Figure 6c\)](#page-7-0), indicating a low degree of metasomatism by a CO_2 -rich silicate melt ([Figure 6d](#page-7-0)).

Therefore, the juvenile lithospheric mantle underneath the Jiaodong Peninsula and surrounding areas has affected by intense carbonate metasomatism, which is spatially consistent with the remarkable lithosphere thinning, large volume of asthenosphere upwelling and giant gold deposits in this region.

7. Link of carbonate metasomatism to the destruction of the eastern North China Craton and associated giant gold deposits

A short-lived giant igneous events (135–110 Ma) ([Wu et al.,](#page-16-0) [2005](#page-16-0); [Zhu et al., 2012b;](#page-18-3) [Zhang et al., 2014](#page-17-31)), tectonic extension([Lin et al., 2013;](#page-14-30) [Zhu et al., 2015](#page-18-8)) and the formation of giant gold deposits [\(Yang et al., 2003](#page-16-33); [Li et al., 2012](#page-14-2); [Zhu](#page-18-8) [et al., 2015](#page-18-8); [Fan et al., 2016\)](#page-13-35) are well developed in the early Cretaceous of the eastern North China Craton. Based on two types of mafic igneous rocks from the Jiaodong Peninsula with contrasting geochemical compositions, [Dai et al. \(2016\)](#page-13-33) suggested that the nature of the mantle lithosphere in the eastern North China Craton changed from an ancient lithospheric mantle to a juvenile lithospheric mantle at \sim 121 Ma —that is the time of termination of the peak decratonization. As a result of the good spatio-temporal consistence between the destruction of the eastern North China Craton and the westward subduction of the Pacific plate, Paleo-Pacific subduction has been suggested as the main dynamic factor triggering the destruction of the eastern North China Craton and the formation of giant gold deposits ([Sun et al., 2007;](#page-16-34) [Zhu et al., 2011,](#page-18-4) [2012a,](#page-18-2) [2015](#page-18-8)).

The westward subduction of the Paleo-Pacific plate not only modified the North China Craton, but also had a significant impact on the architecture of the whole of eastern Asia ([Sun et al., 2007](#page-16-34); [Yin, 2010](#page-17-32); [Dong et al., 2015\)](#page-13-36). For example, the subduction of the Paleo-Pacific plate induced widespread Cretaceous magmatism ([Wang Y J et al., 2013](#page-16-6)) and extensional tectonism ([Lin et al., 2013](#page-14-30)) in the South China Block, which is a stable craton and lacks Jiaodong/ decratonic gold deposits ([Deng and Wang, 2016\)](#page-13-3). This suggests that the lithospheric mantle beneath the eastern North China Craton may have had unique characteristics before its destruction in the early Cretaceous. Carboniferousearly Permian (324–270 Ma) and late Permian-Triassic (262–236 Ma) alkaline rocks occur widely on the northern margin of the North China Craton as a result of the subduction of the Paleo-Asian oceanic plate during the late Paleozoic [\(Zhu et al., 2012b](#page-18-3)). The southeastern North China Craton was modified by the deep subduction of the South China Block and the formation of the late Triassic (225–210 Ma) post-collision magmatism ([Zhu et al., 2012b](#page-18-3)). The late Triassic peridotite xenoliths hosted by the ~223 Ma North Korea kimberlites are characterized by juvenile lithospheric mantle, which demonstrates that the eastern North China Craton was partially destroyed in the Triassic ([Yang et al.,](#page-17-18) [2010\)](#page-17-18).

Long-lived and multiple subduction-related modifications around the North China Craton significantly changed the physical and chemical properties of the lithosphere mantle [\(Windley et al., 2010\)](#page-16-9), providing a prerequisite for the final decratonization. The westward subduction of the Paleo-Pacific plate in the late Mesozoic simply provided a final driving force for the destruction of the eastern North China Craton. The subducted slab not only contributed the dehydrated water to reduce the strength of the upper lithospheric mantle [\(Niu, 2005;](#page-15-7) [Windley et al., 2010](#page-16-9); [Xia et al., 2013](#page-16-3)), but also triggered subduction-related crustal recycling and meltperidotite interactions in the lithospheric mantle [\(Liu et al.,](#page-15-4) [2005,](#page-15-4) [2010](#page-15-6)). The low viscosity and low density carbonate melt reacted easily with mantle peridotite and consumed orthopyroxene ([Russell et al., 2012;](#page-15-35) [Kamenetsky and Yaxley,](#page-14-31) [2015\)](#page-14-31), which modified the structure and composition of the lithospheric mantle. Experimental work by [Wang C et al.](#page-16-7) [\(2016\)](#page-16-7) showed that carbonate melt can very quickly percolate along grain boundaries in harzburgite to form a porridgelike texture, dispersing the cratonic lithospheric mantle.

Compared with the clinopyroxene of global cratonic peridotites, the clinopyroxene in the lithospheric mantle beneath the Sulu orogenic belt and surrounding areas in the eastern North China Craton is characterized by very high Ca/ Al ratios (up to 70) before the late Triassic ([Figure 4b\)](#page-5-0), which indicates strong Type 1 carbonate metasomatism. The lithospheric mantle therefore had a global unique characteristic before the destruction of the eastern North China Craton. The presence of the clinopyroxene (Ca/Al=47–63) and olivine veins with wehrlite mineral assemblages crosscutting the orthopyroxene porphyroblasts in the \sim 240 Ma Lijiatun orogenic peridotite is the direct petrological evidence for strong Type 1 carbonate metasomatism in the lithospheric mantle beneath the eastern North China Craton in the Triassic [\(Su et al., 2016\)](#page-15-12). In general, carbonate melt can exist in the deep mantle, such as the asthenospheric mantle beneath thick subcontinental lithospheric mantle. However, Type 1 carbonate metasomatism is present in both the Donghai garnet-bearing and the Lijiatun spinel-bearing orogenic peridotites, which implies that the deep and shallow lithospheric mantle beneath the eastern North China Craton was strongly modified by Type 1 carbonate metasomatism before the late Triassic. Such Type 1 carbonate metasomatism can also been identified in the ~130Ma Tietonggou peridotite xenolith before the peak $(\sim 125$ Ma) decratonization of the eastern North China Craton ([Figure 4b](#page-5-0) and d). However, Type 1 carbonate metasomatism can not be identified in all the peridotite xenoliths that were trapped after the destruction of the eastern North China Craton ([Figures 4d](#page-5-0), e and 5b, e). Therefore, we suggest that Type 1 carbonate

metasomatism is a prerequisite for the final destruction the eastern North China Craton under global mantle heating in the Cretaceous [\(Machetel and Humler, 2003\)](#page-15-36) and the mantle convection and destabilization induced by the subduction of the Paleo-Pacific plate ([Zhu et al., 2011\)](#page-18-4).

The Jiaodong Peninsula is one of the most important gold deposit provinces in China. It covers <1% of the land area, but accounts for >25% of the total proven gold reserves in China. The Jiaodong gold deposit province was formed within a few million years in the early Cretaceous (130–120 Ma) and was coevally with the peak destruction of the eastern North China Craton and spatially consistent with a pervasive carbonate metasomatism in the lithospheric mantle ([Figure 1\)](#page-2-0). Its ore-forming fluids were thought to have been largely derived from a cooling magma and/or from mantle degassing, which is distinct from typical orogenic gold deposits worldwide [\(Yang et al., 2003;](#page-16-33) [Li et al., 2012;](#page-14-2) [Zhu et](#page-18-8) [al., 2015;](#page-18-8) [Deng and Wang, 2016;](#page-13-3) [Fan et al., 2016](#page-13-35)). However, D-O-C-S stable isotopic results showed that the ore-forming fluids were mainly derived from the enriched lithospheric mantle ([Mao et al., 2008](#page-15-37)). The coeval lamprophyre dykes in the Jiaodong gold province, in which carbonate minerals are commonly observed ([Sun et al., 2001;](#page-15-38) [Guo et al., 2004;](#page-14-32) [Yang](#page-16-35) [et al., 2004](#page-16-35); [Ma et al., 2016](#page-15-39)), are characterized by C–O stable isotopic compositions typical of the upper mantle ([Sun et al.,](#page-15-38) [2001](#page-15-38)). Based on a detailed study of fluid inclusions in quartz veins, [Fan et al. \(2003\)](#page-13-37) showed that the $CO₂$ concentration gradually decreased from the early to late mineralizing stages. Gold in the mantle is controlled by sulfides ([Li and](#page-14-33) [Audétat, 2012](#page-14-33); [Lorand and Luguet, 2016;](#page-15-40) [Jenner, 2017\)](#page-14-34), which can be destabilized under the oxidizing conditions induced by carbonate melts derived from the crustal recycling. This process is thought to release gold- and sulfurrich fluid. $CO₂$ has the ability to buffer the pH of this fluid at a level where elevated concentrations of gold can be transported ([Phillips and Evans, 2004\)](#page-15-41). We therefore suggest that carbonate metasomatism in the lithospheric mantle may have played a crucial part in the activation, transport, enrichment and mineralization of gold in the eastern North China Craton.

8. Genesis of carbonate melts

Type 1 carbonate metasomatism is characterized by high 87 Sr/ 86 Sr ratios of 0.706–0.713 [\(Figure 6a](#page-7-0)). The metasomatic agent should be high Ca/Al ratios and $CO₂$ concentrations and low $SiO₂$ and $Al₂O₃$ concentrations, as seen in the carbonate melt formed from the partial melting of carbonated eclogite [\(Figure 8\)](#page-12-3). The Mesozoic (124 Ma) Laiwu-Zibo carbonatite in the Shandong Province with ⁸⁷Sr/⁸⁶Sr ratios of 0.7095–0.7106 is thought to have been derived from the partial melting of the South China Block and its accompanying carbonate sediments, which was subducted into the deep lithospheric mantle beneath the eastern North China Craton in the Triassic [\(Ying et al., 2004](#page-17-20)). The continental material of the South China Block should have been subducted into the mantle to depths >200 km ([Ye et al., 2000](#page-17-33)). The enriched mantle resulting from crust-mantle interactions during continental deep subduction is suggested to the source of the postcollisional mafic igneous rocks ([Zhao et al., 2013](#page-17-34)). The Triassic (210–201 Ma) and early Cretaceous (133–111 Ma) postcollisional mafic igneous rocks of the Jiaodong Peninsula involve recycled continental crust material and have evolved Sr isotopic compositions $(^{87}Sr)^{86}Sr=0.706-$ 0.714) [\(Dai et al., 2016\)](#page-13-33) consistent with the Sr isotopic composition $(^{87}Sr)^{86}Sr=0.706-0.713$ of clinopyroxenes affected by Type 1 carbonate metasomatism ([Figure 6a](#page-7-0)). The late Triassic mafic igneous rocks show high Ca/Al and $(La/Yb)_N$ ratios and low Ti/Eu ratios ([Figure 7](#page-9-0)) typical of carbonate metasomatism. Carbonate sediments were well developed along the northern margin of the South China Block from the Neoproterozoic to the Paleozoic [\(Zheng and](#page-17-35) [Hu, 2010\)](#page-17-35). Carbonated ultrahigh pressure eclogites also occur widely in the Sulu orogenic belt [\(Proyer et al., 2013;](#page-15-42) [Liu](#page-14-35) [P L et al., 2015](#page-14-35); [Chen Y X et al., 2016](#page-13-38)). We therefore propose that the deep subduction of the South China Block carried significant amounts of carbonate sediments into the lithospheric mantle beneath the North China Craton, inducing intense Type 1 carbonate metasomatism, which provided a prerequisite for the final destruction of the eastern North China Craton.

Combined with the positive relationship between Ca/Al and $CO₂$ and the negative relationship between Ca/Al and $SiO₂$ in the carbonate melt ([Figure 8b](#page-12-3) and c), the low Ca/Al ratio in clinopyroxene affected by Type 2 and Type 3 carbonate metasomatism ([Figures 4d](#page-5-0), e and 5b, e) implies that their metasomatic agents should have high $SiO₂$ and low $CO₂$ contents. This is consistent with clinopyroxenes of Type 3 carbonate metasomatism, which show the relatively high Ti/ Eu ratios ([Figure 5c](#page-6-0) and f) considered to be a result of silicate metasomatism. The low and variable Sr isotopic compositions $\binom{87}{5}$ r/ $\binom{86}{5}$ r=0.7022–0.7056) indicate that the carbonate melt of Type 2 and Type 3 metasomatism was mainly derived from the asthenospheric mantle, but also involves recycled crustal material in variable proportions. This is demonstrated by the Changle lherzolite xenoliths, in which high $87\text{Sr}/86\text{Sr}$ ratio (>0.7035) and low ⁸⁷Sr⁸⁶Sr ratio (< 0.7024) clinopyroxenes are suggested to record the carbonate metasomatism caused by recycled oceanic crust and upwelling of the asthenospheric mantle, respectively ([Deng et al., 2017\)](#page-13-32). The elemental and Mg-Zn stable isotopic compositions show that the Cenozoic basalts from asthenosphere involved a carbonate melt that resulted from crustal recycling in the eastern North China Craton ([Zeng et al., 2010;](#page-17-24) [Yang W et al., 2012](#page-17-26); [Liu et al., 2016;](#page-14-26) [Li et al., 2017](#page-14-27)). Therefore, Type 2 and Type 3 carbonate metasomatisms are considered as the modifica-

[Figure 8](#page-12-3) Variations of ((a), (b)) CO₂ with SiO₂ and Ca/Al, and ((c), (d)) Ca/Al with SiO₂ and Al₂O₃ in experimental melts as a result of the parital melting of carbonated eclogite and peridotite. Data for the partial melting of carbonated eclogite are from [Dasgupta et al. \(2005\),](#page-13-11) [Hammouda \(2003\)](#page-14-36) and [Yaxley and](#page-17-36) [Brey \(2004\)](#page-17-36), data for the partial melting of carbonated peridotite are from [Dasgupta et al. \(2013\),](#page-13-39) [Sokol et al. \(2016\),](#page-15-9) [Brey et al. \(2008\),](#page-13-19) [Klemme et al. \(1995\),](#page-14-6) [Gervasoni et al. \(2017\)](#page-13-7), [Dasgupta et al. \(2007\)](#page-13-40), [Kruk et al. \(2016\)](#page-14-37) and [Dalton and Wood \(1993\);](#page-13-18) the mid-ocean ridge basalt (MORB) data are from [Gale et al.](#page-13-34) [\(2013\);](#page-13-34) the oceanic island basalt (OIB) data are from <http://georoc.mpch-mainz.gwdg.de/georoc/>.

tion of the lithospheric mantle in the eastern North China Craton, which is caused by the carbonate melts derived from the asthenospheric mantle with variable proportions of the recycled crustal material.

9. Conclusions

(1) The geochemcial indices of major elements Ca/Al and Mg#, trace elements $(La/Yb)_N$ and Ti/Eu, and the Sr isotopic composition of clinopyroxene in peridotite can be used to trace carbonate metasomatism in the lithospheric mantle.

(2) Three types of carbonate metasomatism were identified in the lithospheric mantle beneath the eastern North China Craton. Type 1 carbonate metasomatism was a result of the interaction between carbonate melt derived from recycled continental crust and the lithospheric mantle before the late Triassic, which is suggested to have provided a prerequisite for the final destruction of the eastern North China Craton. Type 2 and Type 3 carbonate metasomatism that modified the lithospheric mantle in the eastern North China Craton was caused by carbonate melts derived from asthenospheric mantle with variable proportions of recycled crustal material.

(3) The strong carbonate metasomatism of the lithospheric mantle is spatially consistent with the formation of the Jiaodong giant gold deposits, indicating that carbonate metasomatism played a crucial part in the formation of the giant gold province in the eastern North China Craton.

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