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Chemical indexes of Paleoproterozoic sedimentary rocks from Liaohe Group, North China Craton: Implications for paleoclimate and provenance

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Abstract: Paleoproterozoic supracrustal materials have been widely identified in North China Craton, such as the Liaohe-, Hutuo- and Lyuliang groups. The Liaohe Group in the eastern part of the North China Craton is dominated by deformed and metamorphosed sedimentary and volcanic successions. Compared with that of the coeval volcanic rocks, geochemistry of the sedimentary rocks from the Liaohe Group has rarely been studied in detail, which can possibly provide information on the paleoclimate and provenance. The authors analyzed the whole-rock and detrital zircon geochemistry of sedimentary rocks from the Liaohe Group against a uniform process and proposed their different ways of paleoweathering of the lower and upper formations. That is to say, although the lower and upper formations within the Liaohe Group might be derived from the similar source composition in a tectonically active setting, the paleoclimate that they experienced was not exactly the same. The predominant derivations are the Paleoproterozoic granitoids and basalts within the Liaohe Group, with minor input of Archean continental crust. This study highlights the contributions of the Paleoproterozoic mafic sources, which has been generally overlooked in the previous researches.

Keywords: Liaohe Group; Paleoproterozoic; chemical index; paleoclimate; provenance

0 Introduction

In the last decade, many approaches have been applied to decipher the paleoclimate and provenance of sedimentary rocks, while the majority would not work if the sedimentary rocks have undergone extensive metamorphism and deformation. Fortunately, the geochemical and geochronological methods are increasingly served as robust tools to reveal the source weathering conditions, provenance and even tectonic

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setting of the clastic rocks, despite influence of the early tectono-thermal events (Li *et al.*, 2015). For example, the alkali and alkaline earth elements would be relatively depleted, but Al_2O_3 and TiO_2 would be enriched after intense chemical weathering of clastic rocks, thus such chemical indexes can be used to trace the weathering condition or source composition (McLennan *et al.*, 1993; Nesbitt & Young, 1989). In addition, the detrital zircon geochronology has attracted considerable attention as the zircon U-Th-Pb isotopic system could be closed even during high-grade metamorphism (Nebel *et al.*, 2014). Thus, the age spectrum of detrital zircons can provide constraints to the deposition age and provenance (Luo *et al.*, 2008). The metamorphosed Paleoproterozoic sedimentary rocks are extensively exposed in Liaodong Peninsula (Zhai & Santosh, 2013), which is an ideal target to deliberate paleoclimate and provenance through a combination study of the aforementioned methods. Compared with the coeval volcanic rocks, the geochemical characteristics of the sedimentary rocks have rarely been studied in detail, especially an induction and comparative study of the lower and upper formations (Li & Chen, 2014; Wang *et al.*, 2011, 2017). In this contribution, we compile geochemistry of the detrital zircons and sedimentary rocks of the Liaohe Group and propose different paleoweathering histories of the lower and upper formations, although they were both derived from similar source compositions under an active tectonic setting.

1 Geological settings

The North China Craton is the oldest and largest craton in China, which is bounded by the Central Asian orogenic belt and Qinling–Dabie–Su–Lu ultra-high-pressure metamorphic belt (Zhai & Santosh, 2013). Liaodong Peninsula, northeastern margin of the craton, contains Archean to Paleoproterozoic basement (Fig. 1). The Archean granitoid gneisses and supracrustal rocks are exposed in northern and south-

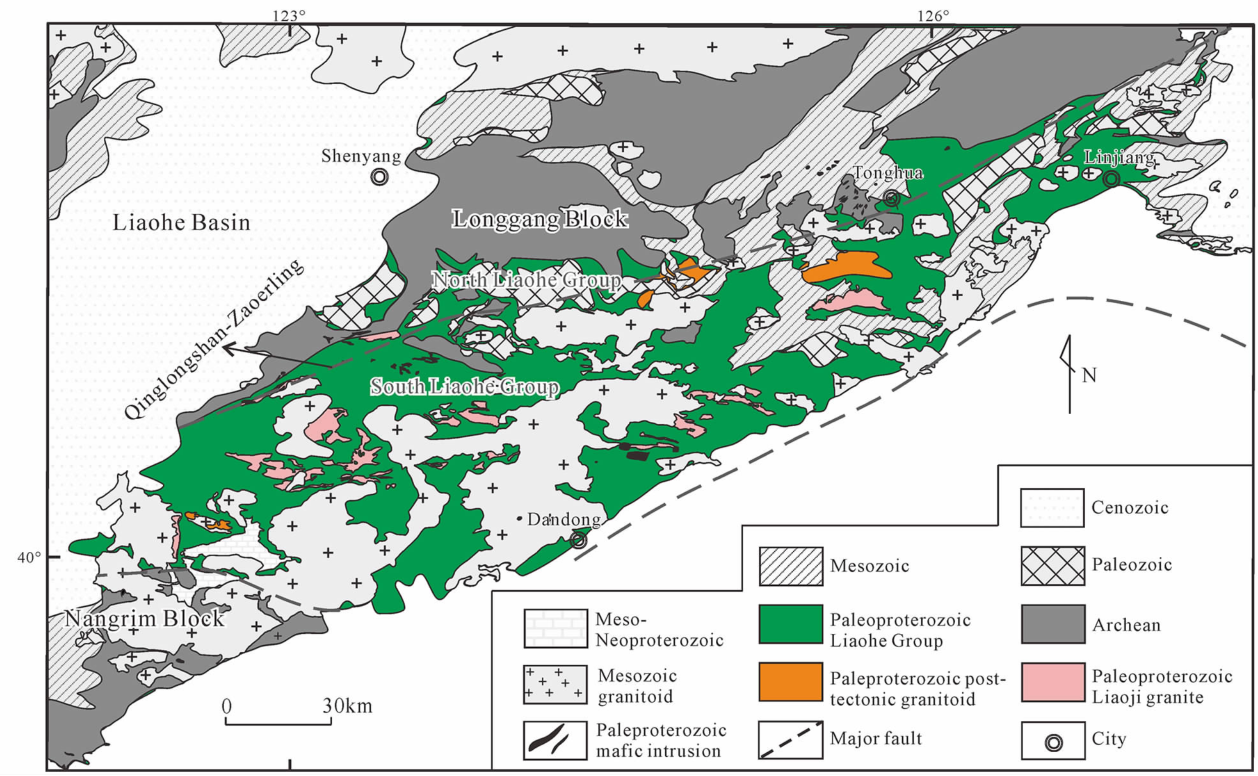


Fig. 1 Geological map of the Archean-Paleoproterozoic basement and post-Paleoproterozoic cover on Liaodong Peninsula (modified after Li *et al.*, 2017, 2019)

ern Liaodong Peninsula, viz. Longgang and Nangrim blocks (Lu *et al.*, 2004). Between the two blocks lies the Jiao–Liao–Ji belt in the central part of the Liaodong Peninsula (Li & Chen, 2014; Zhai & Santosh, 2013) (Fig. 1). The deformed Paleoproterozoic basement with amphibolite-facies metamorphism in the Jiao–Liao–Ji belt unconformably overlies the Archean basement, which consists of volcano-sedimentary successions with minor amounts of granitic and mafic intrusions (Li & Zhao, 2007; Li & Chen, 2014; Li *et al.*, 2015; Lu *et al.*, 2006; Luo *et al.*, 2008) (Fig. 1). The volcano-sedimentary successions are named as Liaohe Group in Liaodong Peninsula, and the lithotectonic equivalents are the Laoling- and Ji'an groups in southern Jilin Province and the Fenzishan- and Jingshan groups in Shandong Peninsula (Luo *et al.*, 2008; Meng *et al.*, 2014). Previous studies suggested that the Liaohe Group was divided into five formations, viz. the lowermost Langzishan Formation (an arkose-rich sedimentary sequence), Li'eryu- and Gaojiayu formations (a lower volcanic-rich sedimentary sequence), and Dashiqiao- and Gaixian formations (a central carbonate-rich and an upper pelitic sequence of sediments), respectively (Li *et al.*, 2015) (Fig. 2). The lower Li'eryu- and Gaojiayu formations comprise boron-bearing volcano-sedimentary rocks metamorphosed to fine grained felsic gneisses, amphibolite and mica quartz schists. Overlying the Gaojiayu Formation is the Dashiqiao Formation, which is composed dominantly of dolomitic marbles intercalated with minor carbonaceous slates and mica schist. The uppermost Gaixian Formation consists of phyllites, andalusite-cordierite mica schists, staurolite mica schists and sillimanite mica schists, with minor quartzites and marbles (Li *et al.*, 2017, 2019) (Fig. 2). Subsequently, the Archean to Paleoproterozoic basement in Liaodong Peninsula was covered by thick Meso-Neoproterozoic and Paleozoic to Cenozoic sediment sequences, both with unconformable contacts (Li *et al.*, 2017). In this paper, we use the lower (Li'eryu-and Gaojiayu formations) and the upper formations (Dashiqiao- and Gaixian formations) for comparison instead of using a single formation.

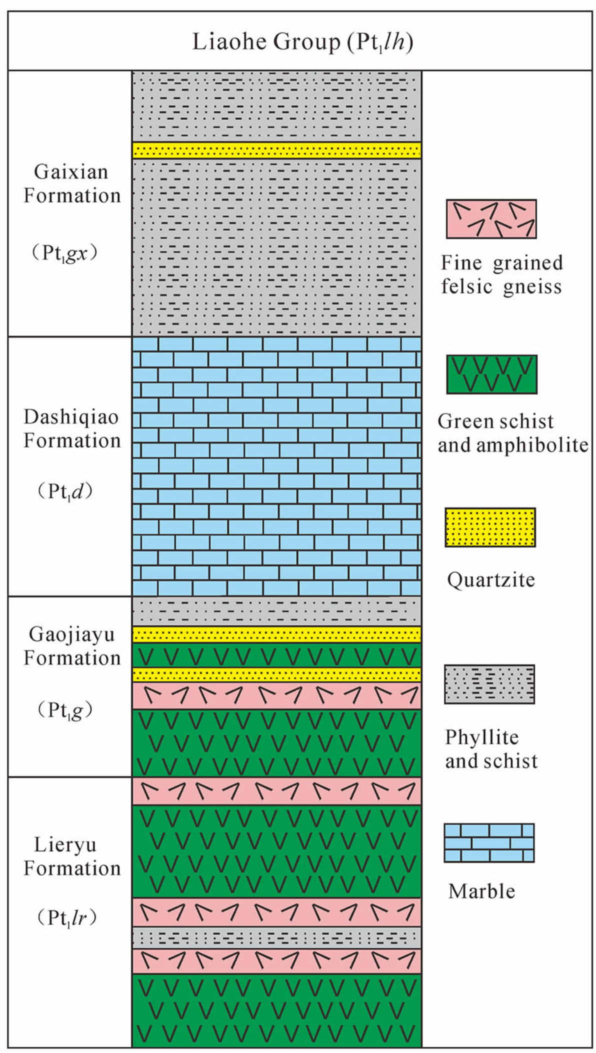


Fig. 2 Lithostratigraphic units of Liaohe Group (after Li *et al.*, 2019; Luo *et al.*, 2008)

2 Same deposition age of lower and upper formations within Liaohe Group

Previous studies indicated that the Liaohe Group was formed at *ca.* 2 300–1 800 Ma, according to their relationship with the country rocks in lithostratigraphy, K-Ar, Ar-Ar and Sm-Nd dating for the mineral and whole-rock, and conventional multi-grain zircon U-Pb methods. Firstly, the tectonic contacts are recognized between the Liaohe Group and country rocks, indicating a high state of uncertainty about the field-based geochronology. For example, a faulted contact between the Paleoproterozoic granitic intrusions and the lower formations within the Liaohe Group was observed during our field investigation (Li *et al.*, 2019). Secondly, the K-Ar and Ar-Ar systems would be disturbed during regional metamorphism, which also cannot yield reliable constraints on ages. Thirdly, the narrow range of Sm/Nd ratios (Li *et al.*, 2015), combined with uncertainties surrounding whether the samples have a cogenetic origin, which means that Sm-Nd whole-rock isochron ages are similarly unreliable. Fourthly, recent geochronological studies have shown that zircons from the Liaohe Group have complex age patterns, such as igneous and metamorphic zircons (Luo *et al.*, 2008), indicating that the multi-grain zircon U-Pb dating results might be geologically meaningless.

In the past decade, zircon U-Pb isotopic dating using the SHRIMP, SIMS, and LA-ICP-MS methods has provided firm constraints on geochronology of the sedimentary rocks within the Liaohe Group (Li *et al.*, 2015, 2019). For example, Li *et al.* (2019) recognized large scale of *ca.* 2.2 Ga volcanism in the lower formations (Li & Chen, 2014). The youngest detrital zircons identified from the lower strata have an age of 2.005 Ga, which can constrain the maximum deposition age of the lower formations (Luo *et al.*, 2008). The age differences between the volcanism and sedimentation in the lower formations suggest a diachronous stratigraphic unit. The minimum ages of the detrital zircons from the upper formations indicate that the deposition age was younger than 1.981 Ga (Li *et al.*, 2015). Taken the regional metamorphism at 1.93 Ga into consideration (Xie *et al.*, 2011), the sedimentary rocks of the lower and upper formations were deposited at 2.01–1.93 Ga and 1.98–1.93 Ga, respectively (Fig. 3). These ages show slight differences within the limit of error, thus supporting the same deposition age of the lower and upper formations within the Liaohe Group. It should be highlighted that the volcanic ages of the lower formations are earlier than the deposition age of the Liaohe Group, thus the volcanic might be an important provenance of the sedimentary rocks. Analyses with less than 5% discordance and more than 5% have both been shown in Fig. 3. Although discordant analyses add some increased scatter to the binned frequency histograms, this has little impact on our interpretations of detrital zircons populations because all comparisons are based on cluster of ages rather than individual ages (Fig. 3). Both analyses with <5% and >5% discordance ensure that inaccurate individual grain ages do not bias contrasts or correlations between age populations in different samples and possible source regions.

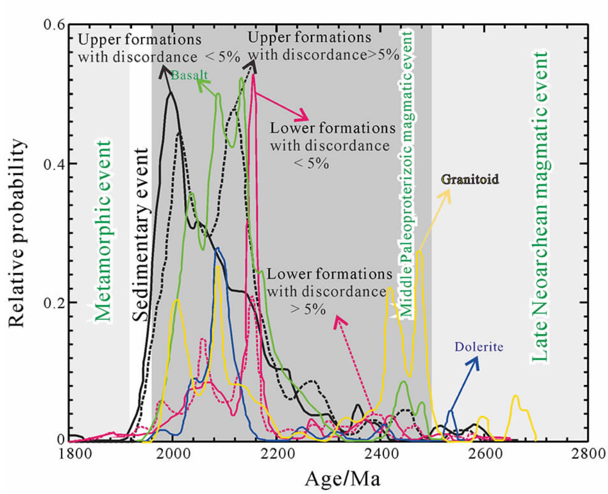


Fig. 3 Probability plot of detrital zircon U-Pb ages of upper and lower formations within Liaohe Group (according to Li *et al.*, 2015; Luo *et al.*, 2008)

3 Different ways and degrees of paleo-weathering of lower and upper formations

3.1 Classification

The protoliths of the metamorphosed clastic rocks of the upper formations are shale and greywacke, while those of the lower formations are shale, greywacke, Fe-bearing shale and Fe-bearing sandstone (Herron, 1988) (Fig. 4a). The Fe contents of the lower formations are relatively enriched, which is consistent with borate and Pb-Zn-Au deposits hosted in the Li'eryu Formation (Zhai & Santosh, 2013). According to the lg(Na_2O/K_2O) – lg(SiO_2/Al_2O_3) diagram (Blatt *et al.*, 1980), most are tightly clustered in the graywacke and litharenite fields (Fig. 4b). The Na_2O/K_2O ratios of the lower formations are higher than those of the upper formations, possibly due to late potassium metasomatism. They are mainly metamorphosed to the fine-grained gneiss and mica schist with plenty of muscovite, biotite and sericite, which further confirms a sedimentary origin (Li *et al.*, 2015, 2019; Luo *et al.*, 2008).

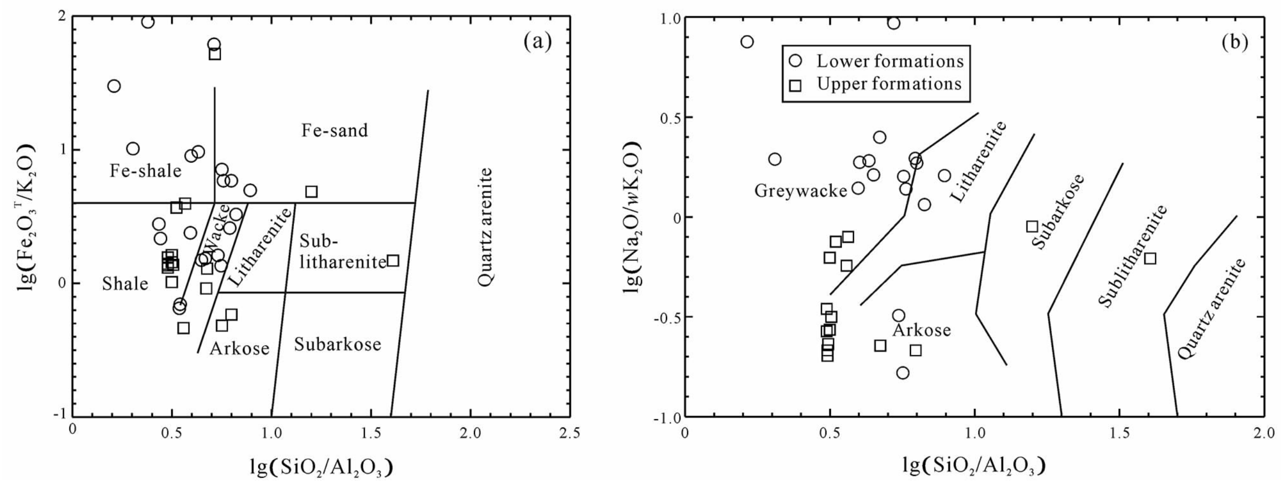


Fig. 4 lg($Fe_2O_3^T/K_2O$) – lg(SiO_2/Al_2O_3) (a) and lg(Na_2O/K_2O) – lg(SiO_2/Al_2O_3) (b) geochemical classification diagrams (after Blatt *et al.*, 1980; Herron, 1988) **for upper and lower formations within Liaohe Group**

3.2 Weathering

Chemical Index of Alteration (CIA = Al_2O_3/(Al_2O_3 + CaO^* + Na_2O + K_2O) × 100 in molecular proportion, where CaO^* represents the CaO associated with the silicate fraction) can quantitatively measure the intensity of paleoweathering in the source area (Nesbitt & Young, 1989). It essentially represents the abundance of alumina relative to other major ca tions, thereby defining the compositional maturity of the sedimentary rock. For example, the CIA values of the fresh igneous rocks are <50, whereas the intense weathering ones can generate CIA values up to 100 (Li *et al.*, 2015). The CIA values of the lower and upper formations approximately plot in the intermediate weathering field, indicating high erosion rates and short transport distances that generated poorly sorted and rapidly deposited sediments (Camiré *et al.*, 1993) (Fig. 5), which should be deposited in a tectonically active setting. This is also evidenced by the negative Eu anomalies of the Liaohe Group in the chondrite-normalized REE patterns (Li *et al.*, 2019).

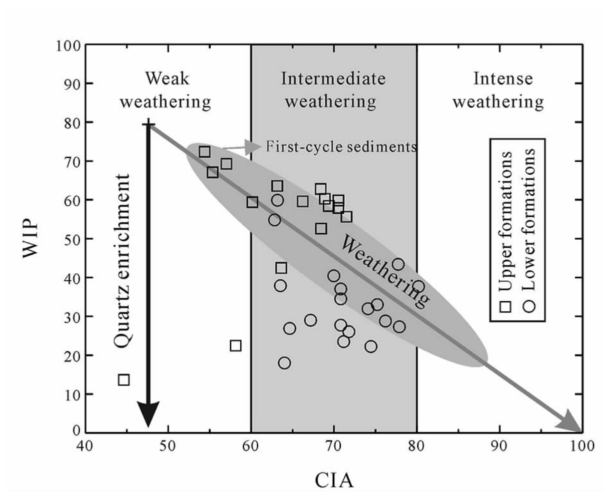
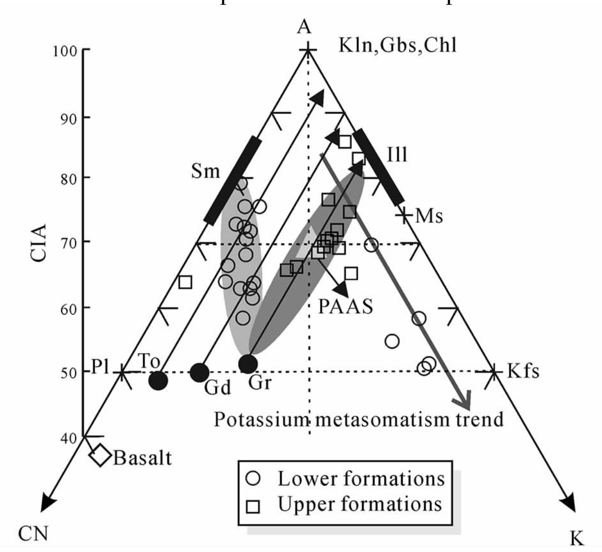


Fig. 5 WIP-CIA discrimination diagram for upper and lower formations within Liaohe Group (Garzanti *et al.*, 2013)

The relatively low CIA values of the upper formations are probably related to a relatively cold and arid climate with weak chemical weathering, while the lower formations with the high CIA values may be formed in warm and semi-humid climatic conditions. Ternary

$Al_2O_3-(CaO+Na_2O)-K_2O$ (A–CN–K) diagram can also provide insights into the weathering history of sources (Nesbitt & Young, 1989). Without regard to potassium metasomatism parallel to the A–K line, the sedimentary rocks of the lower formations define a trend towards smectite, while those of the upper formations plot near the ideal weathering trend parallel to the A–CN line towards illite surrounding the PAAS (post-Archean Australian average shales) (Taylor & McLennan, 1985, 1995) (Fig. 6). The individual trends of the smectite and illite suggest that their paleoclimates were not exactly the same. Formation of smectite requires a warm climate with rainy seasons separated by a dry season with intense evaporation, leading to soil solutions with high pH and concentration in silica and basic cations (Velde, 1995). Such chemical conditions are favored by mafic dykes/sills (Meng *et al.*, 2014; Wang *et al.*, 2011, 2017), and smectite might be derived from the basalts from the lower formations (Li & Chen, 2014). However, illite, a residual mineral produced by physical disaggregation of micaceous metamorphic rocks or feldspar sericitization, indicates relatively dry conditions (Velde, 1995). The results based on the A–CN–K diagram well match the aforementioned inference from CIA values of the lower and upper formations.

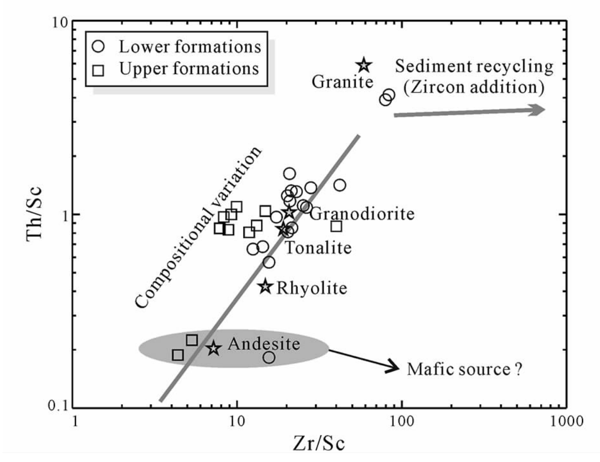


Arrowheads show the predicted weathering trends. Chl, chlorite; Gbs, gibbsite; Ill, illite; Kfs, potassic feldspar; Ms, muscovite; Sm, smectite.

Fig. 6 A–CN–K ($Al_2O_3-[CaO^*+Na_2O]-K_2O$) diagram for sedimentary rocks from upper and lower formations within Liaohe Group

3.3 Recycling

Th/Sc and Zr/Sc ratios are effective chemical indexes to reflect the amount of sedimentary sorting and recycling (Asiedu *et al.*, 2000; Garzanti *et al.*, 2013; McLennan *et al.*, 1993). The mafic rocks are enriched in Sc and depleted in Th relative to the silicic, but the Th/Sc ratio does not vary significantly during sedimentary recycling (Cullers, 1994). In contrast, the Zr/Sc ratios will increase considerably during sediment recycling (McLennan, 1989). Thus, first-cycle sediments show a simple positive correlation between Th/Sc and Zr/Sc, whereas recycled sediments usually show Zr/Sc increasing more rapidly than Th/Sc (Fig. 7). The majority define a trend with a strongly positive correlation between the Th/Sc and Zr/Sc ratios (Fig. 7), which indicates that geochemical variation was dominated by the composition of the sources but not sediment recycling (Cullers, 1994). It is also evidenced by the negative correlation in the WIP-CIA diagram ($WIP=100\times(CaO^*/0.7+2Na_2O/0.35+2K_2O/0.25+MgO/0.9)$ in molecular proportion; Parker,



Compositions of andesite, rhyolite, tonalite, granodiorite, and granite are after Condie (1993).

Fig. 7 lg(Th/Sc)-lg(Zr/Sc) discrimination diagram for sedimentary recycling (McLennan *et al.*, 1993)

1970) (Fig. 5). The WIP values of the Liaohe Group are hardly affected by quartz dilution and mainly controlled by degrees of weathering parallel to the first-cycle trend (Garzanti *et al*., 2013) (Fig. 5). In sum, sedimentary rocks of the Liaohe Group underwent no or minor sedimentary recycling.

4 Similar provenances of lower and upper formations

4.1 Age of provenance

As shown in Fig. 3, the sedimentary rocks of the Liaohe Group are derived from a mixed provenance, viz. Paleoproterozoic and Archean crustal materials. The Paleoproterozoic detrital zircons are well consistent with the age of volcanic rocks from the lower formations within the Liaohe Group and the Paleoproterozoic granitoid intrusions (Li & Zhao, 2007; Li & Chen, 2014; Li *et al*., 2015, 2017, 2019; Wang *et al*., 2011, 2017). The Archean detrital zircons are response to the Paleoarchean to Neoarchean gneissic granitoids and greenstone belts (Lu *et al*., 2004, 2006; Zhai & Santosh, 2013).

4.2 Composition of provenance

In the A–CN–K diagram, the weathering trends defined by sedimentary rocks can be projected backwards to the feldspar join to determine the source composition (Nesbitt & Young, 1989). In spite of the individual weathering trends of the lower and upper formations, they consistently project backwards to the granite join, indicating similar sources to granitic compositions (Fig. 6). However, it should be noted that the possibility of the contributions from basaltic sources cannot be ruled out. In the Th/Sc-Zr/Sc diagram, some sedimentary rocks have relatively low Th/Sc ratios, indicating input of intermediate-mafic source materials (Fedo *et al*., 1995) (Fig. 7).

4.3 Link between age and composition of provenance

In retrospect, there still exist a "missing link" between the source age and composition. Conspicuously, the geochemistry of the zircons can offer a prime opportunity to solve the issue. Based on the statistical analyses, Belousova *et al*. (2002) proposed that the geochemistry of the detrital zircons can be an indicator of source rock type. CART (classification and regression trees) was applied to solve this issue. That is, an unknown zircon run through the tree and ends up in a terminal node, which has been assigned a rock type defined by the majority of the zircons in the node. The proportion of zircons from the defined class gives an approximate estimate of the probability that an unknown zircon falling into this node has been correctly classified. For example, if the zircon has $601 \times 10^{-6} > Lu > 20.71 \times 10^{-6}$ and $Hf < 0.8\%$, it can be classified as one derived from basalt. However, if the zircon has $0.62 \times 10^{-6} < Lu < 20.71 \times 10^{-6}$ and $Hf < 0.62\%$, it can be classified as derived from syenite. In addition, CART shows a very high correlation between true and predicted classes in many cases (Belousova *et al*., 2002). The geochemistry of zircons within the Liaohe Group reveals that Paleoproterozoic sources consist not only granitoids but also mafic rocks (Fig. 3). Although the Paleoproterozoic mafic dyke/sills and basalts were extensively exposed in the Liaodong Peninsula (Li & Chen, 2014; Meng *et al*., 2014; Wang *et al*., 2011, 2017), the contributions of the mafic sources to the sedimentary rocks have not drawn much attention. Taken the high probability of the mafic sources (>50%) (Fig. 3), the quantity of the mafic dyke/sills and basalts during the Paleoproterozoic should be greater in extent than that exposed today. The Archean continental crust acts as a secondary derivation and probability of Archean gneissic granitoids and greenstone belts is consistent with their individual volumes (Zhai & Santosh, 2013) (Fig. 3).

5 Conclusions

The above geochemistry of the detrital zircons and sedimentary rocks within the Liaohe Group lead us to draw the following conclusions:

The protoliths of the metamorphosed clastic rocks of the upper formations in the Liaohe Group are shale and greywacke, while those of the lower formations are shale, greywacke, Fe-bearing shale and Fe-bearing sandstone. The Liaohe Group is mainly controlled by source composition and underwent intermediate weathering. The lower formations may be formed in warm and semi-humid climatic conditions, but the upper formations record relatively dry conditions. The sedimentary rocks were derived from a mixed provenance dominated by the Paleoproterozoic granitoids and basalts within the Liaohe Group, with minor input of Archean continental crust. The contributions of the Paleoproterozoic mafic sources were underestimated in the previous researches.

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