**Supplementary Material**

The supplementary material includes 10 figures (Figs. S1-10) and 6 tables (Table S1-6).

**Table captions:**

**Table S1.** Crystal sizes and aspect ratios from 197 zircon grains for K-bentonites in Ordovician-Silurian transition across in nine sections in South China

**Table S2.** LA-ICP-SFMS zircon U-Pb isotopic and analysis of the K-bentonite samples B2, B14 and B18 in the Songmutang section of the Upper Yangtze region

**Table S3.** Zircon trace element data (in ppm) of the K-bentonite samples B2, B14 and B18 in the Songmutang section of the Upper Yangtze region

**Table S4.** Major element concentrations of K-bentonites in the Songmutang section of the Upper Yangtze region (wt%)

**Table S5.** Trace-element concentrations of K-bentonites in the Songmutang section of the Upper Yangtze region (ppm)

**Table S6.** Mineralogical and geochemical characteristics of K-bentonites from Songmutang section and global K-bentonites

**Figure captions:**

**Fig. S1.** Comparative X-ray diffraction patterns of Songmutang K-bentonites and those of South China elsewhere. Data source: YC0709 from [Hu *et al*. (2009)](#_ENREF_19); D006X2 from [Wang *et al*. (2015)](#_ENREF_56); PY1-V5 from [Ge *et al*. (2019)](#_ENREF_12).

**Fig. S2.** The relation of zircon crystal size and aspect ratio for K-bentonites from South China (A) and Th vs U concentrations of K-bentonite beds from Zhoujiaxi group in Songmutang section (B). Data source: Mayangzhai from [Luo *et al*. (2017b)](#_ENREF_37); Taoyuan from [Xie *et al.* (2012)](#_ENREF_64); Lunshan from [Yang *et al*. (2019)](#_ENREF_71); Anji from [Wang *et al*. (2015)](#_ENREF_56); Xinwen, Wuxing and Liangba from [Ge *et al*. (2019)](#_ENREF_12) ; Wangjiawan from [Hu *et al*. (2008)](#_ENREF_18).

**Fig. S3.** Comparison of chondrite-normalized REE diagrams for K-bentonite from O-S transition in South China with those of the worldwide elsewhere. The normalization values are from [Sun & McDonough (1989)](#_ENREF_51), Wangjiawan and Huanghuachang from [Hu *et al*. (2009)](#_ENREF_19); Yanzixiang and Liziping from [Xiong *et al*. (2019)](#_ENREF_66); Anji from [Wang *et al*. (2015)](#_ENREF_56); Mayangzhai from [Luo *et al*. (2017b)](#_ENREF_37); Lunshan from [Yang *et al*. (2019)](#_ENREF_71); Tianjiawan, Liangbai, Liangcun, Fenghuang and Wuxing date from [Ge *et al*. (2019)](#_ENREF_12); Subduction-related dacite from [Yi *et al*. (2014)](#_ENREF_74); Precordillera argentine from [Huff *et al*. (1998)](#_ENREF_23); Appalachian from [Bergström *et al*. (1998)](#_ENREF_6); Podolia from [Huff *et al*. (2000)](#_ENREF_21); Isles from [Huff *et al*. (1991)](#_ENREF_20).

**Fig. S4.** Comparison of primitive mantle-normalized multi-elemental patterns for K-bentonite from O-S transition in South China with those in the world elsewhere. The normalization values are from [Sun & McDonough (1989)](#_ENREF_51), Wangjiawan and Huanghuachang from [Hu *et al*. (2009)](#_ENREF_19); Yanzixiang and Liziping from [Xiong *et al*. (2019)](#_ENREF_66); Anji from [Wang *et al*. (2015)](#_ENREF_56); Mayangzhai from [Luo *et al*. (2017b)](#_ENREF_37); Lunshan from [Yang *et al*. (2019)](#_ENREF_71); Tianjiawan, Liangbai, Liangcun, Fenghuang and Wuxing from [Ge *et al*. (2019)](#_ENREF_12); Subduction-related dacite from [Yi *et al*. (2014)](#_ENREF_74); Precordillera argentine from [Huff *et al*. (1998)](#_ENREF_23); Appalachian from [Bergström *et al*. (1998)](#_ENREF_6); Podolia from [Huff *et al*. (2000)](#_ENREF_21); Isles from [Huff *et al*. (1991)](#_ENREF_20).

**Fig. S5.** Temporal-spatial distribution of volcanic events within the global scale during O-S transition from South China, North America, Norway, Sweden and Denmark. Isotopic geochronological dates obtained from K-bentonites in South China ([Hu *et al*. 2008](#_ENREF_18); [Xie *et al*. 2012](#_ENREF_64); [Luo *et al*. 2016](#_ENREF_37); [Xiong *et al*. 2017](#_ENREF_65); [Xiong *et al*. 2019](#_ENREF_66); [Yang et al., 2019](#_ENREF_71)); North America ([Tucker & McKerrow 1995](#_ENREF_53); [Min *et al*. 2001](#_ENREF_40); [Smith *et al*. 2011](#_ENREF_49); [Sell *et al*. 2013](#_ENREF_48)); Norway ([Sell *et al*. 2013](#_ENREF_48); [Svensen *et al*. 2015](#_ENREF_52); [Ballo *et al*. 2019](#_ENREF_4)), Sweden ([Tucker & McKerrow 1995](#_ENREF_53); [Min *et al*. 2001](#_ENREF_40); [Bergström *et al*. 2008](#_ENREF_7); [Bauert *et al*. 2014](#_ENREF_5)); Denmark ([Min *et al*. 2001](#_ENREF_40); [Sell *et al*. 2013](#_ENREF_48)); Estonia ([Sell *et al*. 2013](#_ENREF_48); [Bauert *et al*. 2014](#_ENREF_5)).

**Fig. S6.** Nb vs Y tectonic discriminant diagrams after [Pearce *et al*. (1984)](#_ENREF_42). (A) Comparison of K-bentonites from O-S transition in South China and Early Paleozoic granitoids in Qinling area; (B) Comparison of K-bentonites from O-S transition and Early Paleozoic granitoids in South China. Granitoids in Dabie from [Ma *et al*. (2004)](#_ENREF_38) and [Zhang *et al*. (2007)](#_ENREF_78); Granitoids in northern Qinling area from [Li *et al*. (2001)](#_ENREF_29); [Qin *et al*. (2015)](#_ENREF_45); [Wang *et al*. (2009)](#_ENREF_58); [Ren *et al*. (2008)](#_ENREF_47); [Wang *et* *al*. (2018)](#_ENREF_55); [Xu *et al*. (2018)](#_ENREF_68); [Yuan *et al*. (2017)](#_ENREF_76); [Yao *et al*. (2017)](#_ENREF_72); [Abdallsamed *et al*. (2017)](#_ENREF_1); Central Qinling from [Qin *et al*. (2014)](#_ENREF_44) and [Wang *et al*. (2017b)](#_ENREF_57); Eastern Qinling from [Li *et al*. (2019)](#_ENREF_28); Granitoids date in Tongbai from [Jiang *et al*. (2009)](#_ENREF_24); [Li *et al*. (2018)](#_ENREF_31); [Wang *et al*. (2017a)](#_ENREF_54). Granitoids date in eastern Guangxi from [Li *et al*. (2017)](#_ENREF_30); [Nong *et al*. (2017)](#_ENREF_41); [Zou *et al*. (2017)](#_ENREF_81); [Jiang *et al*. (2017)](#_ENREF_25); northeastern Guangxi from [Bai *et al*. (2015)](#_ENREF_3); [Cheng *et al.* (2016)](#_ENREF_11); [Chen *et al*. (2012)](#_ENREF_8); Wugongshan from [Zhong *et al*. (2016)](#_ENREF_79); [Lou *et al*. (2002)](#_ENREF_34); [Wu & Zhang (2003)](#_ENREF_61); [Xu *et al*. (2006)](#_ENREF_67); [Cheng *et al*. (2009)](#_ENREF_10); Yunkai area from [Peng *et al*. (2006)](#_ENREF_43); [Yu *et al*. (2018)](#_ENREF_75); [Yan *et al*. (2017)](#_ENREF_69); Wuyi area from [Liu *et al*. (2008)](#_ENREF_33); [Zeng *et al*. (2008)](#_ENREF_77); [Xia *et al*. (2014)](#_ENREF_62); [Wang *et al*. (2011)](#_ENREF_59); Southern Jiangxi-Northern Guangdong from [Meng *et al*. (2016)](#_ENREF_39); [Qin *et al*. (2018)](#_ENREF_46); [Luo *et al*. (2017a)](#_ENREF_35); K-bentonite from [Hu *et* *al*. (2009)](#_ENREF_19); [Su *et al*. (2009)](#_ENREF_50); [Yi *et al*. (2014)](#_ENREF_74); [Xiong *et al*. (2019)](#_ENREF_66); [Luo *et al*. (2017b)](#_ENREF_37). ORG, ocean ridge granites, VAG, volcanic arc granites, WPG, within-plate granites, Syn-COLG, syn-collision granites.

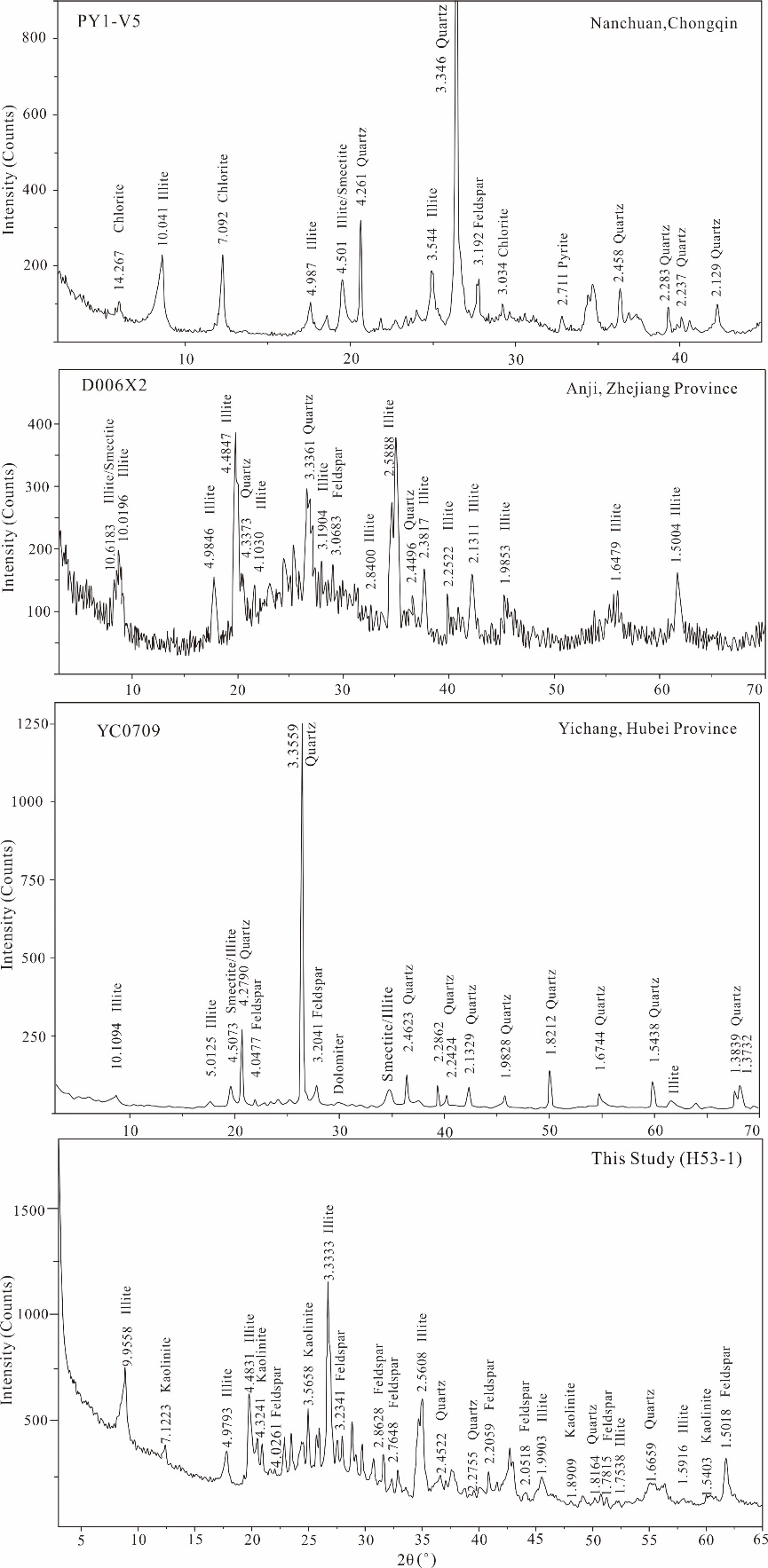
**Fig. S7.** The tectonic-lithofacies paleogeographic map show the development of a diachronous and progressive clastic wedge from the southeastern to the northwestern foreland basin in Central Hunan Province during Late Ordovician to Early Silurian (modified from [Xiao *et al*. 2017)](#_ENREF_63). (A) Tianmashan Fm, 5-10 cm thick sandstones display Ta-Tc-Td members of Bouma sequence in Daoxian area；(B) Tianmashan Fm, exhibits Bouma Tc and Td divisions in Daoxian area; (C) Zhoujiaxi Gp, thin-bedded facies of sandstones with Bouma Tc-Td-Te members; (D) Zhoujiaxi Gp, thin-bedded facies of siltstone shows are composed of amalgamated Td and Te division; (E) Xiaoheba Fm, the littoral-neritic facies of sandstone; (F) Huixingsao Fm, greyish green siltstones intercalated with mudstones; (G) Rongxi Fm, purple siltstone with mudstone intercalation; (H) Xiushan Fm, alternating sandstone and mudstone.

**Fig. S8.** Representative field photographs of unconformity interface in Central Hunan Province. (I) Unconformity between the Tianmashan and Tiaomajian Fm in Yongzhou area; (J) Unconformity between the Tianmashan and Tiaomajian Fm in Jiangyong area; (K) Unconformity between the Zhoujixi Gp and Tiaomajian Fm in Xinhua area; (L) Unconformity between the Zhoujixi Gp and Tiaomajian Fm in Dongkou area. The position of unconformity in Fig. S7 are included.

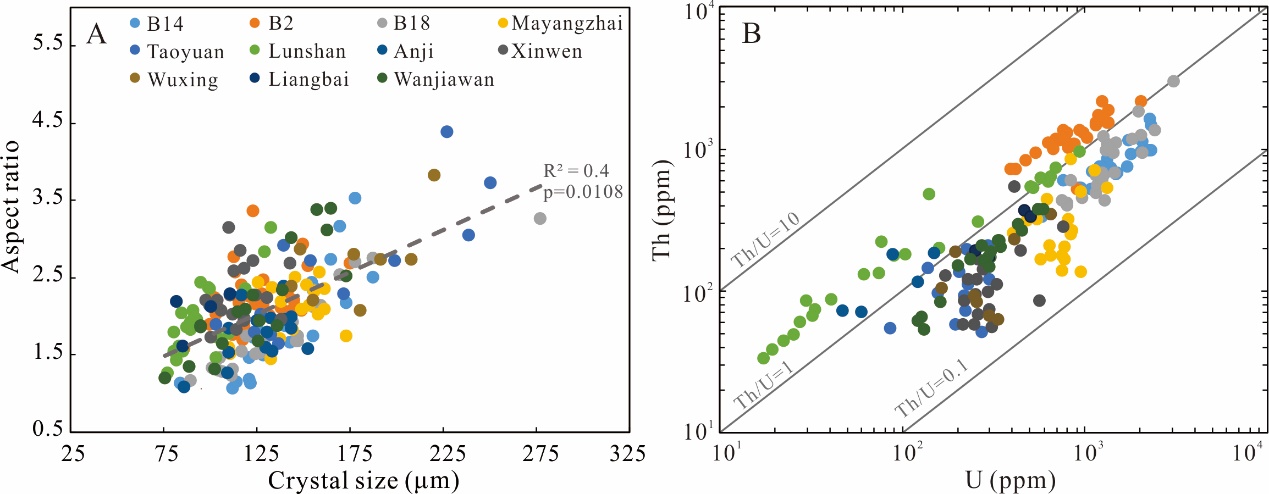
**Fig. S9.** Regional correlation of Late Ordovician to Early Silurian stratum from SE Cathaysia to the Yangtze region through Xuefeng mountain, South China. Orange areas denote sequence missing; dark and grey, black shales; yellow zones, K-bentonite beds; red zones, shallow marine red beds. Chronostratigraphic subdivision after International Commission on Stratigraphy 2018 (ICS); strata division and contrast mainly after [Chen *et al*. (2014)](#_ENREF_9) and [Su *et al*. (2009)](#_ENREF_50); K-bentonites chronological data from [Ge *et al*. (2019)](#_ENREF_12); [Hu *et al*. (2008)](#_ENREF_18) and this study.

**Fig. S10.** Simpliﬁed geological map of South China Sea and adjacent regions showing the possible Nanhai blocks with Precambrian basement. Some major faults and the location of blocks are modified after [Ye *et al*. (2018)](#_ENREF_73); [Yang *et al*. (2017)](#_ENREF_70); [Guo *et al*. (2016)](#_ENREF_14). The distribution of well and trawl from [Liu *et al*. (2012)](#_ENREF_32) and [Zhu *et al*. (2017)](#_ENREF_80).

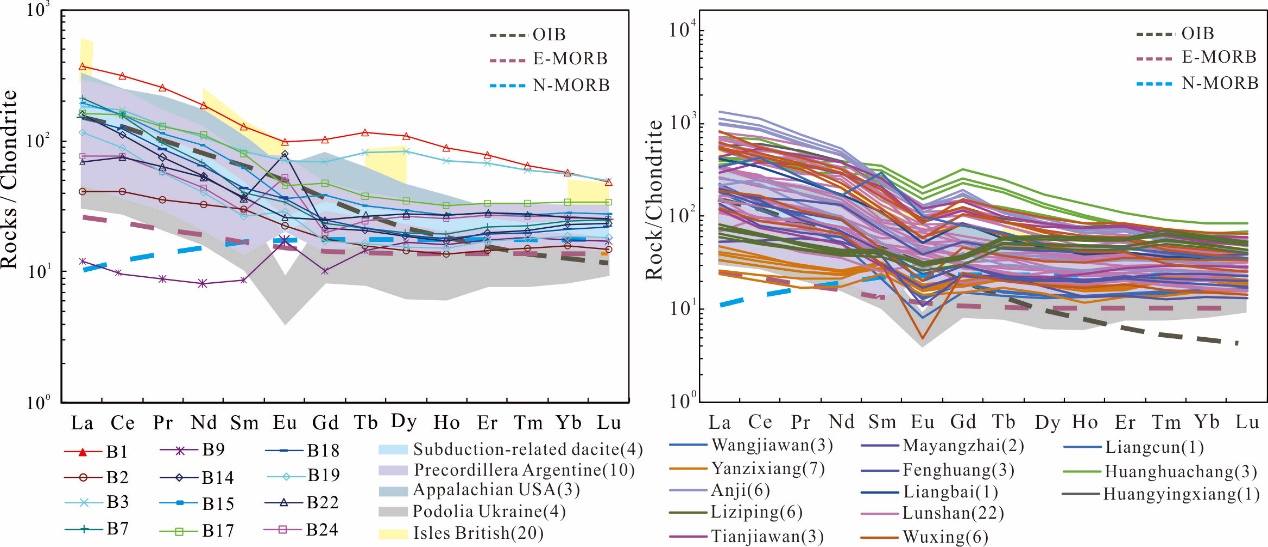
**Fig. S1**



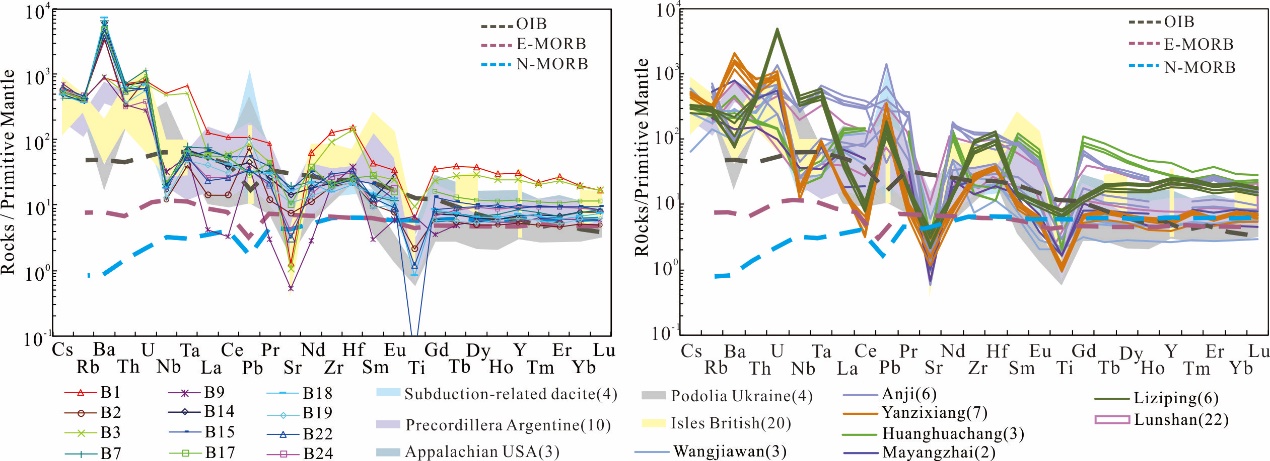
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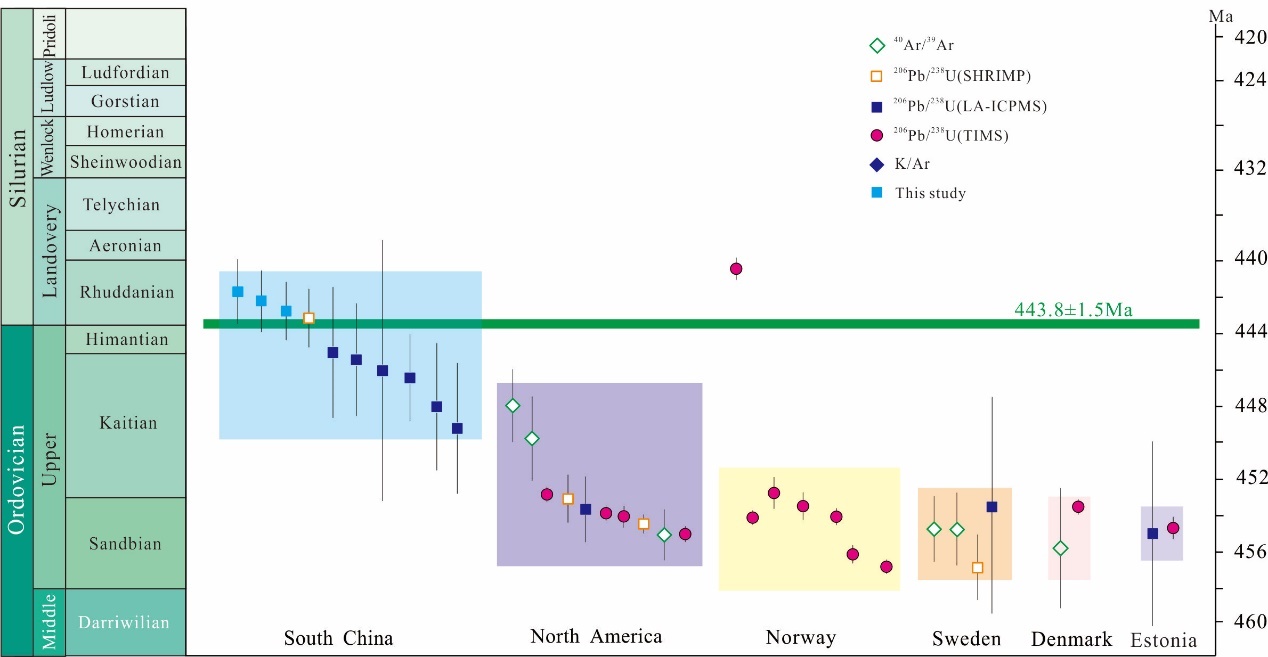
**Fig. S3**



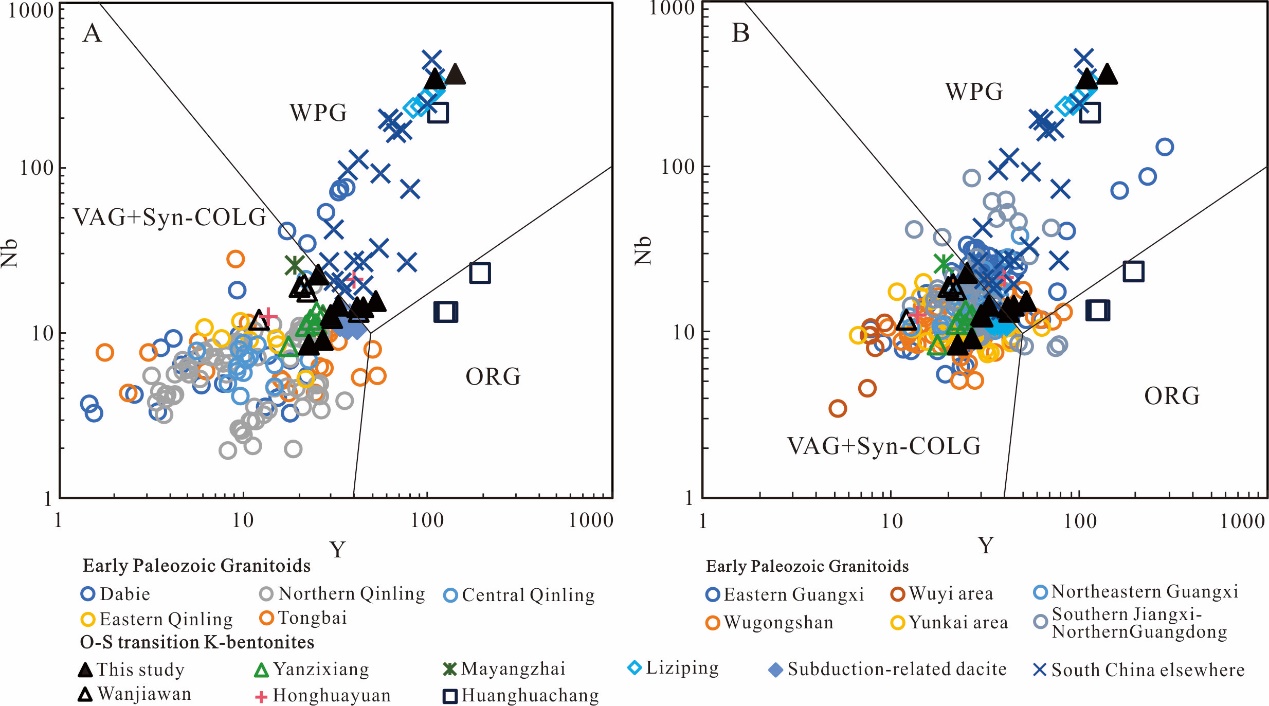
**Fig. S4**



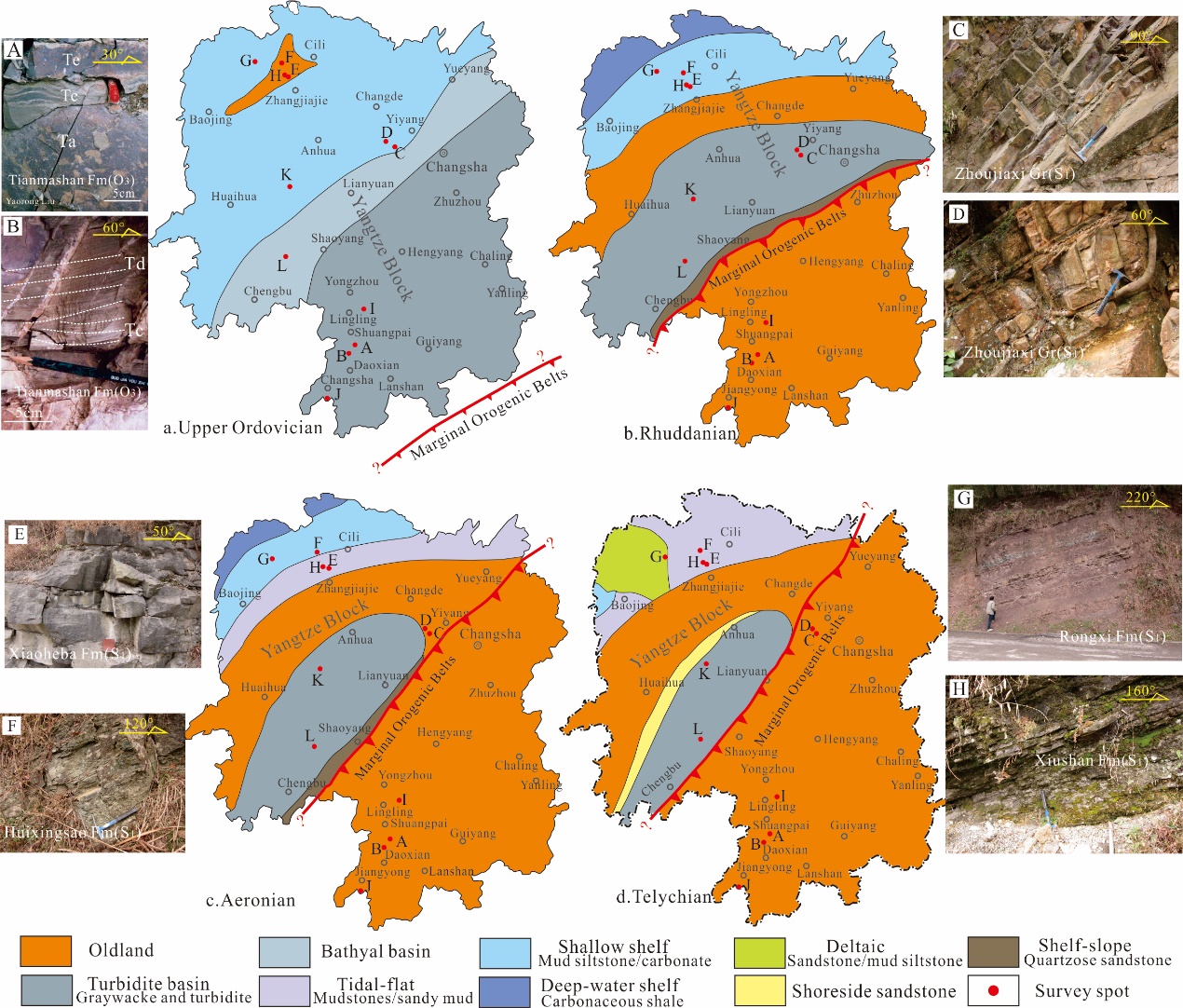
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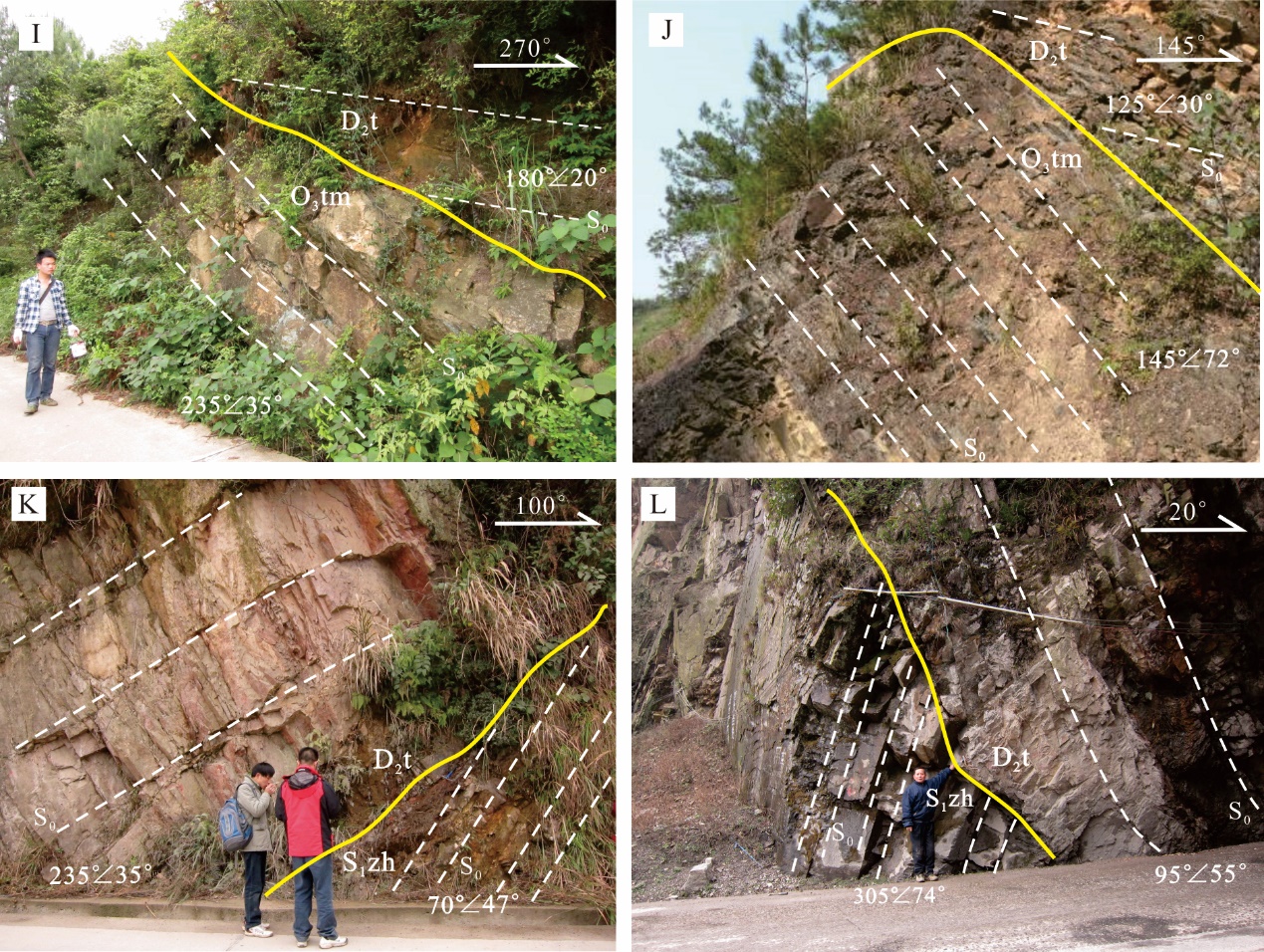
**Fig. S6**



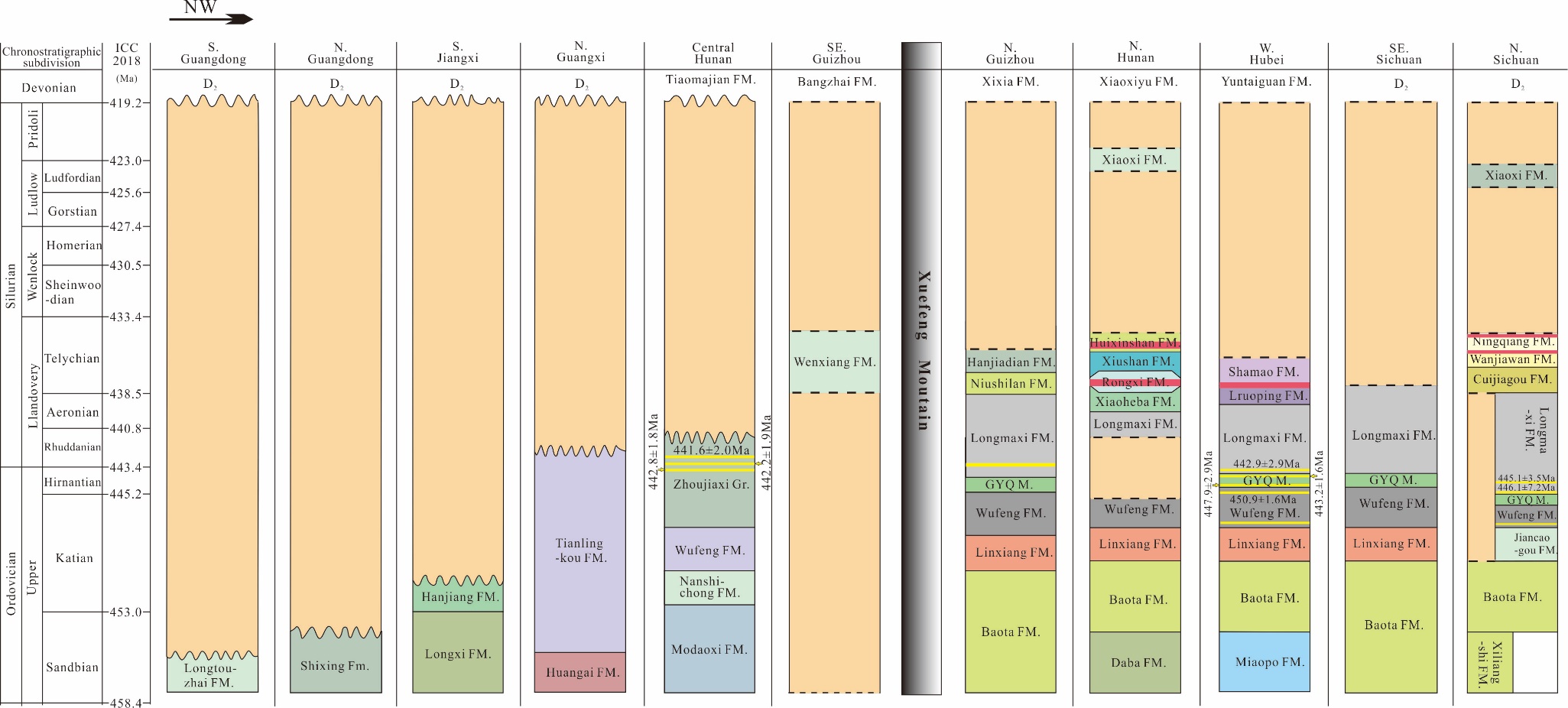
**Fig. S7**



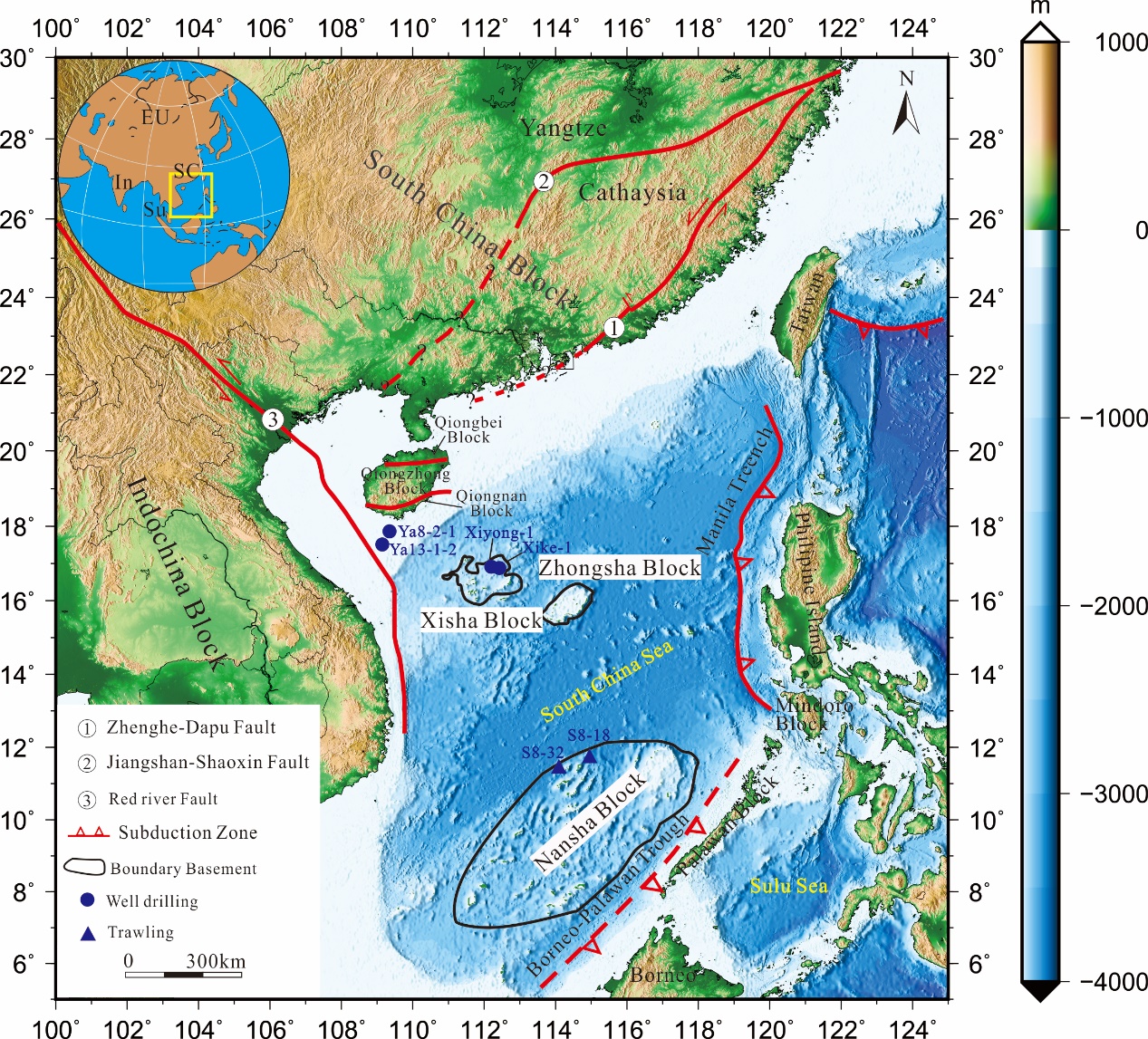
**Fig. S8**



**Fig. S9**

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**Fig. S10**



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