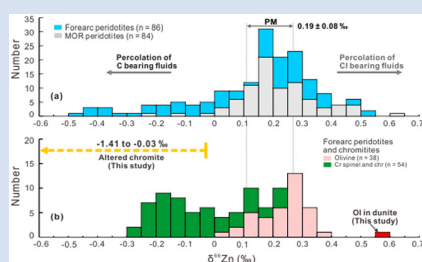


Altered chromite as hidden carrier of isotopically light Zn in forearc mantle

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Abstract



bearing fluids, which are expected to create strong Zn isotope fractionation between aqueous carbonate and other Zn species under certain conditions. Such altered chromites may substantially affect Zn isotopic variations in Cr-rich forearc mantle rocks.

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Introduction

Zinc (Zn) is a transition metal that exhibits lithophile, siderophile or chalcophile behaviour depending on geological processes (Moynier *et al.*, 2017). In hydrothermal solutions, Zn is highly mobile and significant Zn isotope fractionation occurs between different aqueous Zn species (chloride, sulfide, and carbonate) under certain fluid conditions (Fujii *et al.*, 2011, 2014), making Zn and its isotopes ($\delta^{66}\text{Zn}$) a useful tracer of fluid chemistry among different geological environments (Pons *et al.*, 2011; Debret *et al.*, 2018, 2021, 2024).

Several Zn isotope studies of oceanic peridotites/serpentinites from mid-ocean ridges and forearcs have documented a wide $\delta^{66}\text{Zn}$ range (−0.48 to +0.62 ‰; see summary in Fig. 1) relative to that of the primitive mantle (0.19 ± 0.08 ‰; Fang *et al.*, 2022). Such large isotopic variations cannot be ascribed to high temperature magmatic processes (Sossi *et al.*, 2018; Chen *et al.*, 2021), but are likely induced by widespread fluid-rock interactions associated with peridotite exhumation and serpentinisation (Pons *et al.*, 2011; Debret *et al.*, 2018, 2024). Within such solid + fluid systems, the changes in Zn isotopes in peridotites are primarily driven by mineral dissolution-reprecipitation reactions (Putnis, 2009), depending on the conditions and nature of fluids in different geological environments (Pons *et al.*, 2011; Debret *et al.*, 2018, 2024). In addition, it has been shown that spinel, as an accessory phase in upper mantle peridotites, hosts up to ~60 % of the bulk rock Zn budget (Fang *et al.*, 2022). This

implies that dissolution and precipitation of spinels during widespread fluid-rock interactions (Arai and Akizawa, 2014) may considerably affect Zn elemental and isotopic variations in oceanic peridotites (Debret *et al.*, 2024). Moreover, a recent study demonstrated that Cr spinels in oceanic peridotites display overall lighter $\delta^{66}\text{Zn}$ than that of Al spinels in cratonic peridotites, probably reflecting a chemical control on Zn isotope systematics of spinels (Yang *et al.*, 2021). Nonetheless, an assessment of Zn isotope systematics in Cr spinels and its potential effect on Zn isotopic variations in oceanic mantle rocks is lacking.

In this study, we present mineral chemistry, elemental mapping, and Zn isotopes of variably altered chromites from high Cr-type (spinel Cr# > 60) ophiolitic chromitites in Tibet, China. Our new data reveal significant $\delta^{66}\text{Zn}$ variability in chromite, including the lightest values recorded in peridotite minerals, and demonstrate that these significantly light isotopic signatures primarily stem from secondary Zn, Fe enriched phases precipitated during fluid mediated alteration. These findings highlight the pivotal role of chromite alteration in driving Zn elemental and isotopic variability in Cr-rich forearc mantle rocks.

Results

Eight chromitites and one chromite-rich dunite collected from several Tibetan ophiolites in China were studied in detail (Fig. S-1, Tables S-1–S-7; see Supplementary Information (SI)). The

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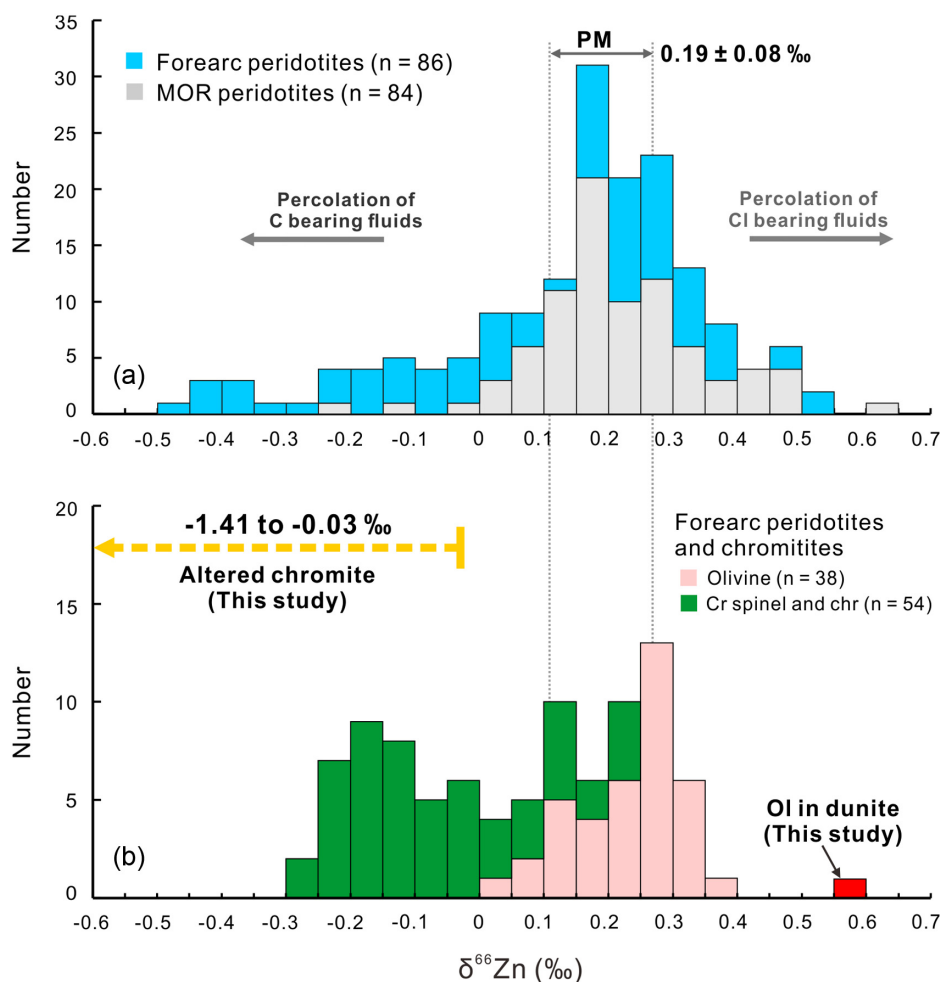


Figure 1 (a) A summary of Zn isotope data of oceanic peridotites/serpentinites (see [Supplementary Information](#) for data sources). (b) Zn isotopes of chromites from forearc ophiolitic peridotites and chromites presented in this and previous studies. The grey dashed lines mark the range of primitive mantle (PM; [Fang et al., 2022](#)).

chromitite samples display a variety of textures (Figs. S-2–S-4), with chromite modes ranging from 25 % to 97 % (Table S-1). Primary silicates (mostly olivines) in these samples have been completely replaced by secondary mineral assemblages consisting of chlorite ± serpentine ± dolomite ± sulfides (Table S-1, Fig. S-5). Chromites are variably replaced by secondary Fe enriched zones along the rims and fractures (Figs. S-4–S-8). The relative proportions of Fe enriched zones in mapped chromites in these samples vary from <5 % to ~60 % (Fig. S-9).

The core regions of chromites in these samples have low variability of Cr# [$100 \times \text{Cr}/(\text{Cr} + \text{Al})$] values (63.0–78.5), Mg# [$100 \times \text{Mg}/(\text{Mg} + \text{Fe}^{2+})$] values (60.8–75.8), and $\text{Fe}^{3+}/\text{Fe}_{\text{total}}$ ratios (0.14–0.29) (Figs. S-10, S-11). Minor elements (Zn, Mn) in the core regions also show restricted variations (Figs. 2, S-6–S-8), with Zn contents of 432–1058 µg/g (mostly <600 µg/g). In contrast, Fe enriched zones in these samples show systematically increased Cr# (up to ~100), decreased Mg# (down to ~0), as well as highly variable $\text{Fe}^{3+}/\text{Fe}_{\text{total}}$ ratios (0.03–0.67) and Zn contents (<200 to ~40,000 µg/g) compared with corresponding cores (Figs. S-10, S-11).

The chromitite samples are further subdivided into three groups, based on the extent of Zn enrichment in Fe enriched zones (Figs. 3, S-10 and S-11). In Group 1 samples, the Fe enriched zones are almost Fe^{2+} -rich (ferrous) chromites, with $\text{Fe}^{3+}/\text{Fe}_{\text{total}}$ ratios of 0.02–0.35 and Zn contents of 300–1445 µg/g (mostly <1000 µg/g). In Group 2 samples, the Fe enriched

zones are also ferrous chromites, with similar $\text{Fe}^{3+}/\text{Fe}_{\text{total}}$ ratios (0.05–0.34) but apparently higher Zn contents (427–4209 µg/g, mostly >1000 µg/g). In Group 3 samples, the Fe enriched zones are mixtures of ferrous chromites ($\text{Fe}^{3+}/\text{Fe}_{\text{total}} = 0.03$ –0.39, Zn = 610–39,060 µg/g), Fe^{3+} -rich (ferrian) chromites ($\text{Fe}^{3+}/\text{Fe}_{\text{total}} = 0.41$ –0.59, Zn = 786–15,090 µg/g), and Cr magnetites ($\text{Fe}^{3+}/\text{Fe}_{\text{total}} = 0.61$ –0.67, Zn = <200–2889 µg/g).

In terms of Zn isotopes, chromites from Group 1 samples have $\delta^{66}\text{Zn}$ of -0.15 ‰ to -0.03 ‰, while anomalously negative $\delta^{66}\text{Zn}$ are documented in chromites from Group 2 (-0.62 ‰ to -1.29 ‰) and Group 3 (-0.96 to -1.41 ‰) samples (Fig. 4). Given that chromites in these samples are variably altered, these isotopic values likely represent mixed signatures between primary cores and secondary Fe enriched zones. For comparison, both chromites and olivines in the chromite-rich dunite (21DZ02) are unaltered (Fig. S-4), with $\delta^{66}\text{Zn}$ of 0.57 ± 0.04 ‰ and -0.21 ± 0.02 ‰, respectively (Table S-7).

Discussion

Fluid-mediated alteration of chromite. Chromite alteration is common in ophiolitic peridotites and chromitites ([Gervilla et al., 2012](#); [Eslami et al., 2021](#); [Liu et al., 2023](#)), with textures and compositions of primary chromites can be modified to variable extents *via* mineral replacement (dissolution-precipitation)

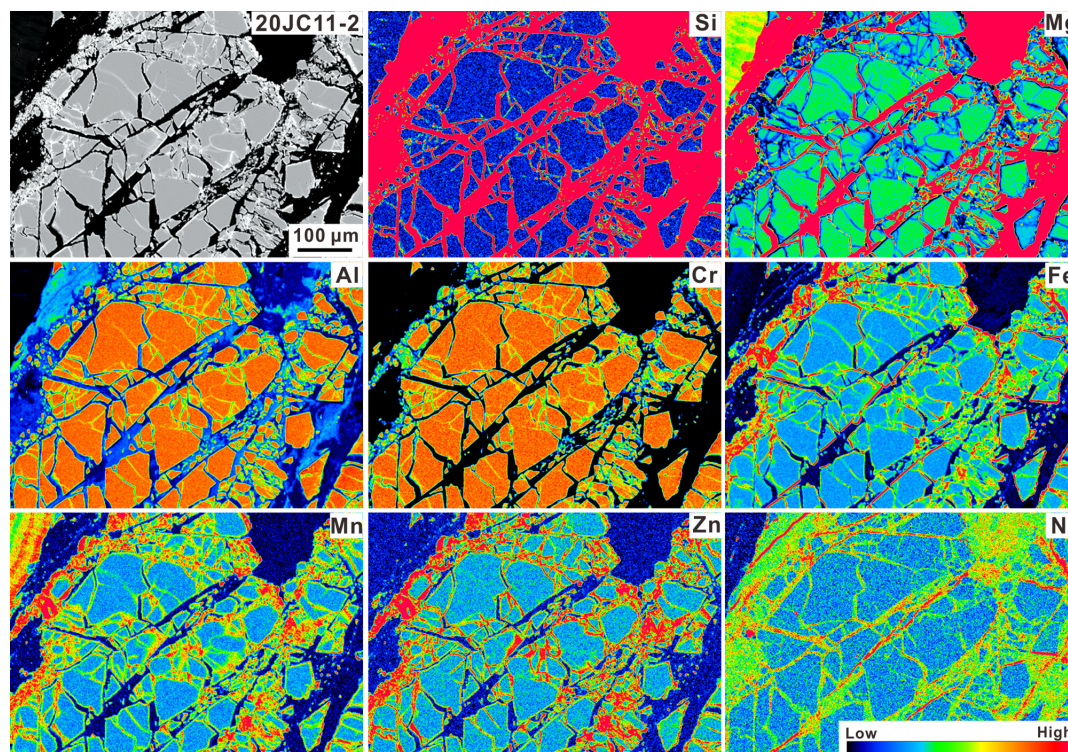


Figure 2 Elemental distribution maps of chromite from one heavily fragmented and altered sample (20JC11-2) that displays significantly light Zn isotopes.

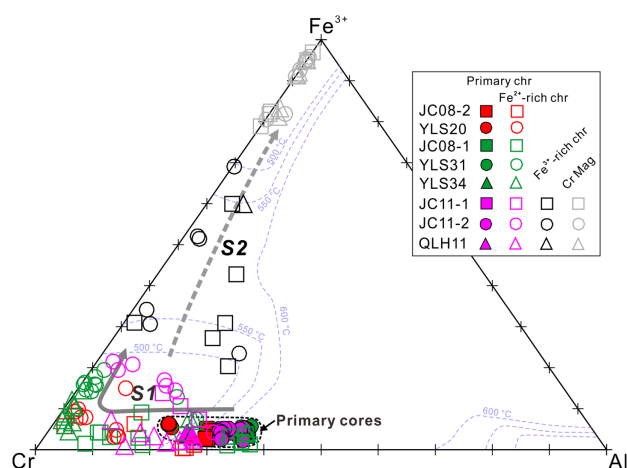


Figure 3 Ternary Cr-Fe³⁺-Al plot showing compositional variations of primary and altered chromites. S1 and S2 display different stages of chromite alteration. The isothermal lines (defined by olivine Fo = 90) demonstrate the phase relations within the spinel prism [(Fe,Mg)(Al,Cr,Fe³⁺)₂O₄] over the temperature range of 500–600 °C (cf. Sack and Ghiorso, 1991).

reactions in the presence of fluids (Putnis, 2009; Arai and Akizawa, 2014). For Group 1 and 2 samples, the secondary Fe enriched zones are almost ferrous chromites, characterised by progressive increase in Cr and Fe (mainly in Fe²⁺ form) coupled with decrease in Mg and Al compared with corresponding chromite cores. Notably, Fe enriched zones in some samples are closely associated with chlorite inclusions within chromite cores (Fig. S-5c), or develop porous textures with abundant chlorites filling the pores (Fig. S-5d,e). These features together with co-precipitation of interstitial chlorite $\pm$ serpentine suggest

reaction between primary chromite and co-existing olivine under H₂O saturated conditions, during which primary chromites were progressively dissolved along the rims and fractures and re-precipitated as porous Fe²⁺-rich chromites along with chlorite filling the pores (see SI; Gervilla *et al.*, 2012).

For Group 3 samples, the Fe enriched zones are composed of ferrous, ferrian chromites, and Cr magnetites, which occur as both single phase and mixed phases. In the former cases, the contacts between ferrous/ferrian chromites and primary cores are diffusive (Fig. S-5f), in contrast with the sharp contacts between Cr magnetites and primary cores (Fig. S-5i). In the latter cases, successive outward transitions from ferrous to ferrian chromite and Cr magnetite are documented (Fig. S-5g,h). These features, together with the progressive increase in Fe³⁺/Fe_{total} and decrease in Cr from ferrous to ferrian chromite and then to Cr magnetite (Fig. S-10), suggest a two-stage alteration process. The first stage involves the precipitation of ferrous chromites similar to those documented in Group 1 and 2 samples. In the second stage, the growth of intervening ferrian chromites and Cr magnetites likely resulted from inter-crystalline diffusion reaction between early formed, ferrous chromite and a Cr-poor, Fe³⁺-rich end member phase in presence of fluids (see SI; Prabhakar and Bhattacharya, 2013). The most likely candidate for this end member phase is magnetite, given the sharp contact between Cr magnetite and primary cores, and the continuous change in Cr and Fe³⁺ from ferrian chromite to Cr magnetite that trends toward pure magnetite.

The presence of ferrian chromites and Cr magnetites in Fe enriched zones in Group 3 samples suggests more advanced alteration, consistent with higher extents of fragmentation of chromites in these samples (Fig. S-4). Similar two-stage alteration of chromites has been widely documented in ophiolitic peridotites and chromitites, and is generally interpreted as the results of fluid-rock interaction in the course of their exhumation (Gervilla *et al.*, 2012; Eslami *et al.*, 2021). Several thermodynamic

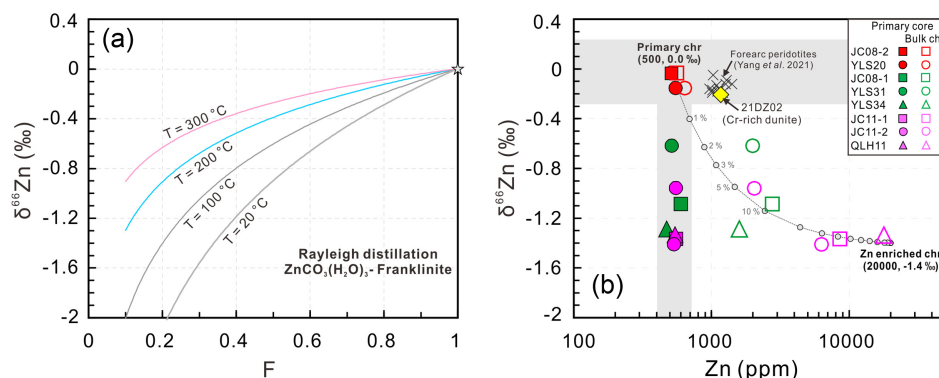


Figure 4 (a) Rayleigh distillation modelling results for $\delta^{66}\text{Zn}$ evolution of secondary Zn, Fe enriched chromites (e.g., franklinite) precipitated from a carbonate bearing fluid at different temperatures (see SI). $\delta^{66}\text{Zn}_{\text{initial}}$ was set to +0.00 ‰, similar to that documented in chromite from Group 1 samples. (b) Plot of $\delta^{66}\text{Zn}$ versus bulk Zn of chromites (hollow symbol) in the samples. Dashed line denotes mixing result between a primary chromite end member and a secondary Zn, Fe enriched chromite end member (see SI). Grey areas mark Zn range of primary chromites in studied samples and $\delta^{66}\text{Zn}$ range of primary chromites reported from forearc ophiolitic peridotites and chromites (−0.27 to +0.23 ‰; Yang *et al.*, 2021; Lian *et al.*, 2024; Su *et al.*, 2026).

models of chromite alteration under various temperature and $f\text{O}_2$ conditions further predicted that these first stage reactions take place under high temperature ranges (>450 to ~700 °C) with increasing $f\text{O}_2$, while the second stage reactions take place under lower temperatures with decreasing $f\text{O}_2$ (Eslami *et al.*, 2021; Liu *et al.*, 2023). These inferred conditions are supported by observed compositional variations in secondary phases formed during different stages. In the first stage, the high temperature is evidenced by these less chloritised ferrous chromites that display compositions approaching those of primary cores, while the increasing $f\text{O}_2$ is suggested by these heavily chloritised ferrous chromites which tend to contain more Fe^{3+} as the alteration reaction proceeds (Fig. 3). In the second stage, the precipitation of a magnetite end member along with serpentine ± talc ± Ni-Fe sulfides in the matrix (Figs. S-4–S-8) is consistent with the expected products of serpentinisation, which is favoured by low temperature (<400 °C) and reducing conditions (McCollom and Bach, 2009).

Zn enrichment and significantly light Zn isotopes in altered chromite. Chromites in the studied chromitites display a wide $\delta^{66}\text{Zn}$ range (−1.41 to −0.03 ‰), including the lightest value ever documented in terrestrial rocks and minerals. Given that chromites in these samples are variably altered, these $\delta^{66}\text{Zn}$ values most likely represent mixed signatures. Notably, chromites from Group 1 samples as well as the chromite-rich dunite show $\delta^{66}\text{Zn}$ (−0.21 to −0.03 ‰) similar to values reported for fresh chromites from forearc ophiolitic peridotites and chromitites (−0.27 to +0.23 ‰, Fig. S-12; Yang *et al.*, 2021; Lian *et al.*, 2024; Su *et al.*, 2026), while chromites from Group 2 and 3 samples feature highly negative $\delta^{66}\text{Zn}$ (−1.41 to −0.62 ‰). Elemental mapping and quantitative analysis further show that Zn is more evenly distributed in chromites from Group 1 samples, with limited Zn enrichment in secondary Fe enriched zones relative to corresponding cores (Figs. S-10, S-11). In contrast, Zn is significantly and heterogeneously enriched in secondary Fe enriched zones (up to 4.86 wt. %) relative to corresponding cores in chromites from Group 2 and 3 samples (Figs. S-10, S-11), linking these anomalously negative $\delta^{66}\text{Zn}$ with Zn, Fe enriched zones precipitated during fluid mediated alteration.

Theoretical studies have predicted significant Zn isotopic fractionation between different Zn species (e.g., chloride, sulfide, carbonate) in hydrothermal fluids under certain conditions (Fujii *et al.*, 2011, 2014). Notably, at high $p\text{CO}_2$ and pH conditions, isotopically heavy Zn is preferentially incorporated into aqueous

carbonates, resulting in negative $\delta^{66}\text{Zn}$ values in other co-existing Zn species in fluids (Fujii *et al.*, 2011). The documentation of dolomite + chlorite ± serpentine associations in the matrix provide direct evidence of carbonate bearing fluids involved in the alteration processes. This means secondary Zn, Fe enriched chromites precipitated from these fluids are expected to be enriched in isotopically light Zn. Simple Rayleigh distillation modelling between Zn bearing chromite (ZnFe_2O_4) and a fluid phase complexing Zn mainly as carbonate ($\text{ZnCO}_3(\text{H}_2\text{O})_3$), following the procedures detailed described by Debret *et al.* (2024), further show that the precipitated Zn bearing chromites can have highly negative $\delta^{66}\text{Zn}$ (see SI; Fig. 4a).

The inferred alteration effect on Zn isotopic variations in chromites is consistent with the results of binary mixing between a primary chromite end member and a secondary Zn, Fe enriched chromite end member (Fig. 4b). As documented above, chromites from Group 1 samples are little affected by alteration and largely preserve primary compositional signatures, while chromites from Group 2 and 3 samples are more intensely and heterogeneously modified (Fig. S-13). In this frame, Zn and $\delta^{66}\text{Zn}$ of the primary chromite end member were set to 500 $\mu\text{g/g}$ and 0.00 ‰, respectively, similar to those values documented from Group 1 samples. For the Zn enriched end member, Zn and $\delta^{66}\text{Zn}$ were set to 20,000 $\mu\text{g/g}$ and −1.41 ‰, respectively, covering those values documented from Group 2 and Group 3 samples. Then, the bulk Zn contents of chromite in each sample were recalculated using the Zn contents of Fe enriched zones and primary cores, and their relative proportions as estimated based on mapped chromite grains (Fig. S-9). It should be noted that the extent of Zn enrichment in Fe enriched zones is determined by both the nature of fluids (*i.e.* Zn budget in the system) and fluid/rock ratios (*i.e.* the extent of reaction). Despite the uncertainties for estimated bulk Zn in chromites, the results show that chromites from most Group 2 and 3 samples display coupled $\delta^{66}\text{Zn}$ -bulk Zn variations generally following the modelled mixing curve (Fig. 4b).

The strong similarity in the distribution pattern of Fe enriched zones in different sample groups suggests that Zn was mainly enriched during widespread fluid-rock reactions. The close association of these Zn, Fe enriched zones with the chlorite-serpentine matrix further suggests these enriched elements could be released from olivine (host peridotites) upon serpentinisation and redistributed during different stages of chromite alteration *via* dissolution-precipitation reactions (Arai and Akizawa, 2014). The

progressive Zn enrichment in Fe enriched zones from Group 1 to Group 3 samples is also consistent with an increasing extent of chromite alteration with decreasing temperatures and increasing fluid/rock ratios (Fig. S-13). Note that highly enriched Zn (up to several wt. %) has been documented in some ferrous and ferrian chromites from Group 3 samples, indicating some of the Zn could be externally added to the system by Zn-rich mineralising fluids.

Effect of chromite alteration on Zn isotope systematics in forearc mantle. High Cr-type (spinel Cr# > 60) ophiolitic chromitites primarily form at forearc settings, with slab-derived fluids playing a critical role in their genesis and subsequent modification (Su *et al.*, 2020, 2021). Available Zn isotope studies of forearc peridotites/serpentinites have documented a wide $\delta^{66}\text{Zn}$ range (−0.48 to +0.52 ‰; Fig. 1a), with non-equilibrium Zn isotope fractionation in these highly serpentinised rocks having been attributed to extensive fluid-rock reactions (Debret *et al.*, 2024). On the other hand, Zn isotope analyses of fresh Cr spinels/chromites from forearc ophiolitic peridotites and chromitites revealed a moderate range of $\delta^{66}\text{Zn}$ (−0.27 to +0.23 ‰; Fig. 1b), which covers the observed $\delta^{66}\text{Zn}$ variations in least affected chromites from Group 1 samples as well as the chromite-rich dunite (Fig. 4b). However, these anomalously negative $\delta^{66}\text{Zn}$ documented in intensely altered chromites from Group 2 and 3 samples are unexpected, suggesting Zn isotopes in chromites can be significantly changed during alteration.

Despite its low modal abundance (1–3 %) in most forearc peridotites, primary Cr spinels host a certain amount (up to 60 %) of the Zn budget in the peridotites (Yang *et al.*, 2021; Fang *et al.*, 2022). More importantly, primary chromites can be significantly modified by hydrothermal solutions (Arai and Akizawa, 2014), resulting in highly enriched Zn (up to several wt. %) in altered chromites (Singh and Singh, 2013; Gamal El Dien *et al.*, 2019). A wide range of geological and geophysical data have indicated extensive hydration in the forearc mantle, with estimated serpentinisation levels reaching 50 % in some forearcs (Hyndman and Peacock, 2003). These highlight the possibility that altered chromites serve as a potential sink for Zn and other compatible elements (*e.g.*, Mn) in the serpentinised forearc mantle (Gamal El Dien *et al.*, 2019). Moreover, at shallower depths, slab derived, carbonate bearing fluids are significantly transferred to forearc mantle (Kawamoto *et al.*, 2013; Debret *et al.*, 2018), which can dissolve more metals than pure H₂O and greatly affect the chemical evolution of the mantle wedge. In this frame, hydrothermally altered Cr spinels by carbonate bearing fluids may not only host a certain amount of Zn budget in the forearc mantle, but preferentially carry isotopically light Zn (Pons *et al.*, 2011; Fujii *et al.*, 2011), providing a plausible explanation for very negative $\delta^{66}\text{Zn}$ documented in some forearc serpentinites (Fig. 1). These forearc serpentinites may release isotopically light Zn in fluids to modify the mantle wedge, producing light Zn isotopic signatures observed in some arc-front lavas (Chen *et al.*, 2021). Note that many forearc serpentinites display a primitive mantle-like or much heavier $\delta^{66}\text{Zn}$ (Fig. 1), suggesting the effect of chromite alteration is highly dependent on the nature (*e.g.*, carbonate bearing) of fluids as well as the extent of alteration reactions.

Conclusions

This study reports significantly light Zn isotopes in variably altered chromites from ophiolitic chromitites. Textural and chemical data reveal heterogeneously enriched Zn in secondary Fe enriched zones developed along rims and fractures of chromites, pointing to different stages and extents of fluid mediated alteration. The involvement of carbonate bearing fluids is expected to result in anomalously negative $\delta^{66}\text{Zn}$ in secondary

Zn, Fe enriched chromites precipitated from the fluids, which facilitates Zn elemental and isotopic variability in bulk chromites. These findings highlight the role of this type of chromite alteration in affecting Zn isotopic variations in highly serpentinised forearc mantle.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2638>.



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Altered chromite as hidden carrier of isotopically light Zn in forearc mantle

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Supplementary Information

The Supplementary Information includes:

- Analytical Methods
- Geological Background and Sample Description
- Alteration Reactions of Chromite
- Rayleigh Distillation Modelling
- Tables S-1 to S-7
- Figures S-1 to S-13
- Supplementary Information References
- Additional References for Data Sources in Figure 1

Analytical Methods

Mineral elemental compositions

Major element compositions of minerals were measured using a JEOL JXA-8230 electron probe micro-analyzer at the Wuhan SampleSolution Analytical Technology Co., Ltd., China (WSSA). The operating conditions include 20 nA beam current, 15 kV accelerating voltage, 10 s counting times for peak, and 5 s counting times for upper and lower background per element. The spot sizes are 1 μm for most minerals and 3 μm for chlorite. The analytical standards used are natural minerals from international suppliers. Data were corrected online using a ZAF (atomic number, absorption, fluorescence) correction procedure. The analytical uncertainties are generally better than $\pm 2\%$. Mineral elemental distribution maps were acquired through surface scanning analysis using the same equipment, with 20 kV accelerating voltage and 200 nA probe current. The image pixel grid sizes are 1.5–6 * 1.5–6 μm , the pixel sizes are 350–600 * 300–480, and the dwell time is 20–30 ms.

Trace-element compositions of chromite were determined on thin sections using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the WSSA. Detailed operating conditions for the laser ablation system and the ICP-MS instrument, and data reduction strategy follow those described by Liu *et al.* (2008). Laser sampling was performed using a GeolasPro laser ablation system comprising a COMPexPro 102 ArF excimer laser (with a wavelength of 193 nm and a maximum energy of 200 mJ) and a MicroLas optical system. An Agilent 7700e ICP-MS instrument was used to acquire ion-signal intensities. Helium was applied as a carrier gas, and argon was used as the make-up gas that was mixed with the carrier gas via a T-connector before entering the ICP. The spot size and frequency of the laser were set to 44 μm and 5 Hz, respectively. Each analysis incorporated a background acquisition of ~ 20 s (gas blank) followed by 50 s data acquisition from the unknowns. Multiple reference materials (BHVO-2G, BCR-2G, BIR-1G, GSE-1G and NIST SRM610) were analyzed and the preferred values for the USGS reference glasses are from the GeoReM database (<http://georem.mpch-mainz.gwdg.de/>). Off-line selection and integration of background and analyte signals, and time-drift correction and quantitative calibration were performed by ICPMSDataCal (Liu *et al.*, 2008). The analytical accuracies (RD%) for elements in the reference materials discussed here is better than $\pm 10\%$.

Zinc isotopes

Zn isotopic analyses were conducted at Metallogenic Elements and Isotopes Laboratory in the Institute of Geology and Geophysics, Chinese Academy of Sciences, following the protocol described in Wang *et al.* (2023). Mineral separates of chromite and olivine from crushed rock samples were prepared. The chromites were preprocessed using a M6 Microwave Oven (PreeKem, China) with mixture of concentrated 4.5 mL HCl, 1.5 mL HNO₃, 3 mL HF and 1 mL HClO₄. And then followed the general routine like other minerals.

10–60 mg of mineral powders were dissolved using concentrated HNO₃ and HF acids in 7 mL PTFE Teflon square digestion vessels at 160 °C for 2 days. Following the evaporation of HNO₃ and HF, the samples were subsequently treated with HNO₃ and HCl (1:3) and refluxed at 80 °C for 1 day. Then the samples were dried down and re-dissolved in 1 mL concentrated HCl at 130 °C. Finally, the samples were dried down again and brought into solution in 8 M HCl (+ 0.001% H₂O₂) for chemical purification.

Zn was purified using a two-step ion exchange chromatography. The solution was loaded onto pre-conditioned 2 mL Bio-Rad AG-MP-1M resin first. Matrix elements were eluted in the next 37 mL of 8 M HCl, 18 mL of 2 M HCl and 2 mL of 0.5 M HNO₃ sequentially. Zn fraction was collected in 10 mL of 0.5 M HNO₃ afterwards, evaporated to dryness and then redissolved in 1 mL of 8 M HCl (+ 0.001% H₂O₂) which was loaded onto pre-conditioned 1 mL Bio-Rad AG-1-X8 resin for second purification. 9 mL of 8 M HCl and 10 mL of 0.4 M HCl were used to elute the matrix elements, and Zn fraction was collected in 7 mL of 0.5 M HNO₃, which was evaporated to dryness and diluted in 2% HNO₃ to 500 ng g⁻¹ for Zn isotopic measurements. The total procedure blank for Zn isotope analyses is < 5 ng.

Zn isotopic measurements were performed on Nu Sapphire CC-MC-ICP-MS at the conventional pathway in low resolution mode using the sample-standard bracketing method. Beams of ⁶⁴Zn, ⁶⁶Zn, ⁶⁷Zn and ⁶⁸Zn were collected in Faraday cups in static mode, with ⁶²Ni monitor to correct for interference ⁶⁴Ni on ⁶⁴Zn. Each analysis consisted of a block with 50 cycles of 3 s integration. A 30 s wash was performed in 2% HNO₃ between each standard and sample to

avoid cross contamination. Long-term reproducibility is 0.03‰ (2SD), evaluated by analyses of NIST SRM 3168a (Wang *et al.*, 2023). Zn isotope results $\delta^{66}\text{Zn}$ in this study were reported as the per mil deviation relative to standard JMC 3-0749L.

Geological Background and Sample Description

Ophiolites in Tibet (China) are mainly distributed along two major suture zones, the Bangong-Nujiang suture (BNS) in the north and the Yarlung-Zangbo suture (YZS) in the south (Fig. S-1). These ophiolites are widely interpreted as fragments of oceanic lithosphere generated at forearc settings of the Mesozoic Neo-Tethyan Ocean (e.g., Hebert *et al.*, 2012; Wang *et al.*, 2016; Qian *et al.*, 2020). Chromite mineralization is commonly observed in mantle sections of most ophiolites, but economically viable chromite deposits are only found in a few massifs such as Luobusa and Dongqiao.

Eight chromitite samples collected from several massifs, including Yilashan, Qielihu, and Jiangcuo, scattered across the central segment of the BNS were studied in detail (Fig. S-1). These massifs consist mainly of moderately to highly serpentized harzburgites, and in some areas, the harzburgites contain meter-sized dunite lenses or patches, often associated with chromitites (Girardeau *et al.*, 1984; Wu *et al.*, 2021; Xu *et al.*, 2023). In addition, one very fresh chromite-rich dunite sample collected from the Xigaze massif in the YZS was studied as a reference. The mantle section of this massif is dominated by quite fresh harzburgites, with centimetre-scale dunite lenses or patches are occasionally observed within the harzburgites (Zhang *et al.*, 2017).

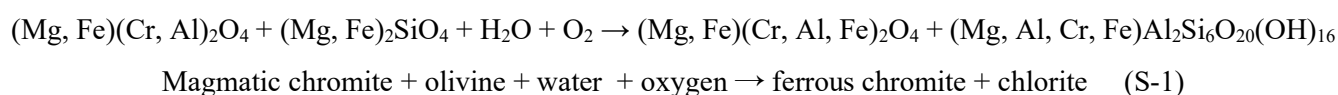
The studied chromitites display a variety of textures, including disseminated-veinlet, banded, and massive (Figs. S-2, S-3). Chromites in samples with disseminated-veinlet textures (20YLS-20, 20QLH11) commonly show sub-rounded to anhedral shapes, and may contain silicate inclusions in the interior regions (Fig. S-4b, j). These inclusions in sample 20YLS-20 are composed almost of sub-rounded chlorites (Figs. S-5c, S-6c), while those in sample 20QLH11 are composed of serpentine, chlorite and amphibolite (Figs. S-5m, S-8a). In contrast, chromites in these samples with semi-massive to massive textures (20JC08-1, 20JC08-2, 20YLS31, 20YLS34) commonly exhibit subhedral shapes (Fig. S-4c, d, k, m). Note that chromites in two other samples (20JC11-1, 20JC11-2) are highly fragmented, showing cataclastic textures (Fig. S-4e, g, i).

All the chromitite samples have undergone certain degrees of alteration, with primary silicates (mainly olivines) in the matrix as well as those hosted by chromites are completely replaced by secondary mineral assemblages consisting of chlorite±serpentine±dolomite±sulfides (Table S-1, Fig. S-4). On the other hand, chromite grains are mildly to heavily altered in different samples, mainly along grain margins and fractures (Fig. S-4). These altered regions in chromites are easily identified due to their overall higher reflectivity compared to unaltered cores under reflected light and backscattered-electron (BSE) imaging (Figs. S-5–S-8). In the chromite-rich dunite sample, both chromite and co-existing olivines are fresh, with some sub-rounded to anhedral chromites containing fresh olivine inclusions (Fig. S-6a).

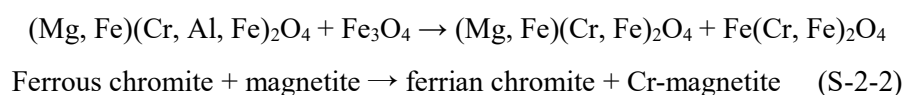
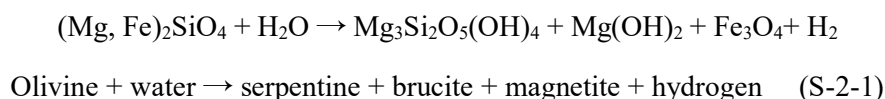
Alteration Reactions of Chromite

The alteration of chromite/Cr-spinels in ultramafic rocks have been widely recognized and studied since 1970s (e.g., Bliss and MacLean, 1975; Evans and Frost, 1975; Burkhard, 1993; Mellini *et al.*, 2005; Gervilla *et al.*, 2012; Barra *et al.*, 2014; Colás *et al.*, 2014; Eslami *et al.*, 2021; Roy *et al.*, 2022; Liu *et al.*, 2023). In recent years, detailed microtextural and mineralogical study of hydrothermally altered chromites as well as thermodynamic modelling consistently showed that the alteration of chromite involves a general two-stage process with increasing fluid/rock ratios and decreasing temperatures (e.g., Gervilla *et al.*, 2012; Barra *et al.*, 2014; Colás *et al.*, 2014; Eslami *et al.*, 2021; Roy *et al.*, 2022; Liu *et al.*, 2023).

Based on textures and chemical evolution of altered chromites in different sample groups, a similar two-stage alteration process was proposed. The first stage fluid-mediated metamorphic reactions take place under high temperatures between primary chromite and co-existing silicate minerals (mainly olivine), and the reactions were described by Eslami *et al.* (2021) and Liu *et al.* (2023):



The second stage reactions take place under lower temperatures corresponding to serpentinization of host peridotites, and the reactions were described by McCollom and Bach (2009) and Liu *et al.* (2023):



Rayleigh Distillation Modelling

The $\delta^{66}\text{Zn}$ evolution of secondary Zn-, Fe-enriched chromites presumably precipitated from a carbonate-bearing fluid was modelled using a simple Rayleigh distillation model, according to Debret *et al.* (2024) by using the following equations:

$$10^3 \ln(\alpha_{\text{fluid-mineral}}) = 10^3 \ln(\beta_{\text{fluid}}) - 10^3 \ln(\beta_{\text{mineral}})$$

$$\Delta^{66}\text{Zn}_{\text{final}} = (1000 + \delta^{66}\text{Zn}_{\text{initial}}) \times (F^{(\alpha-1)} - 1) + \delta^{66}\text{Zn}_{\text{initial}}$$

where α is the fractionation factor between the Zn-bearing chromite [Franklinite, ZnFe_2O_4] and a fluid phase complexing Zn mainly as $[\text{ZnCO}_3(\text{H}_2\text{O})_3]$ between 100 and 300°C. β_{fluid} and β_{mineral} represent the reduced partition function ratios for aqueous Zn carbonate $[\text{ZnCO}_3(\text{H}_2\text{O})_3]$ and precipitated Zn-bearing chromite $[\text{ZnFe}_2\text{O}_4]$, based on ab initio calculation results of Fujii *et al.* (2014) and Ducher *et al.* (2016), respectively. We restrict the discussion to Zn-bearing chromite precipitation when smithsonite (ZnCO_3) does not reach saturation. $\delta^{66}\text{Zn}_{\text{Initial}}$ was set to +0.00 ‰, similar to those documented in chromite from Group-1 samples. F is the fraction of Zn remaining in the fluid.

Supplementary Tables

Table S-1	A summary of texture and alteration characteristics of chromite in the studied chromitites and dunite.
Table S-2	Major and minor element compositions of primary chromites in the studied chromitites and dunite.
Table S-3	Major and minor element compositions of altered chromites in the studied chromitites.
Table S-4	Major element compositions of secondary silicates in the studied chromitites.
Table S-5	Major element compositions of secondary carbonates in the studied chromitites.
Table S-6	Major element compositions of secondary sulfides in the studied chromitites.
Table S-7	Zn isotopic compositions of chromite in the studied chromitites and dunite.

Tables S-1 to S-7 (xlsx) are available for download from the online version of this article at <https://doi.org/10.7185/geochemlet.2638>.

Supplementary Figures

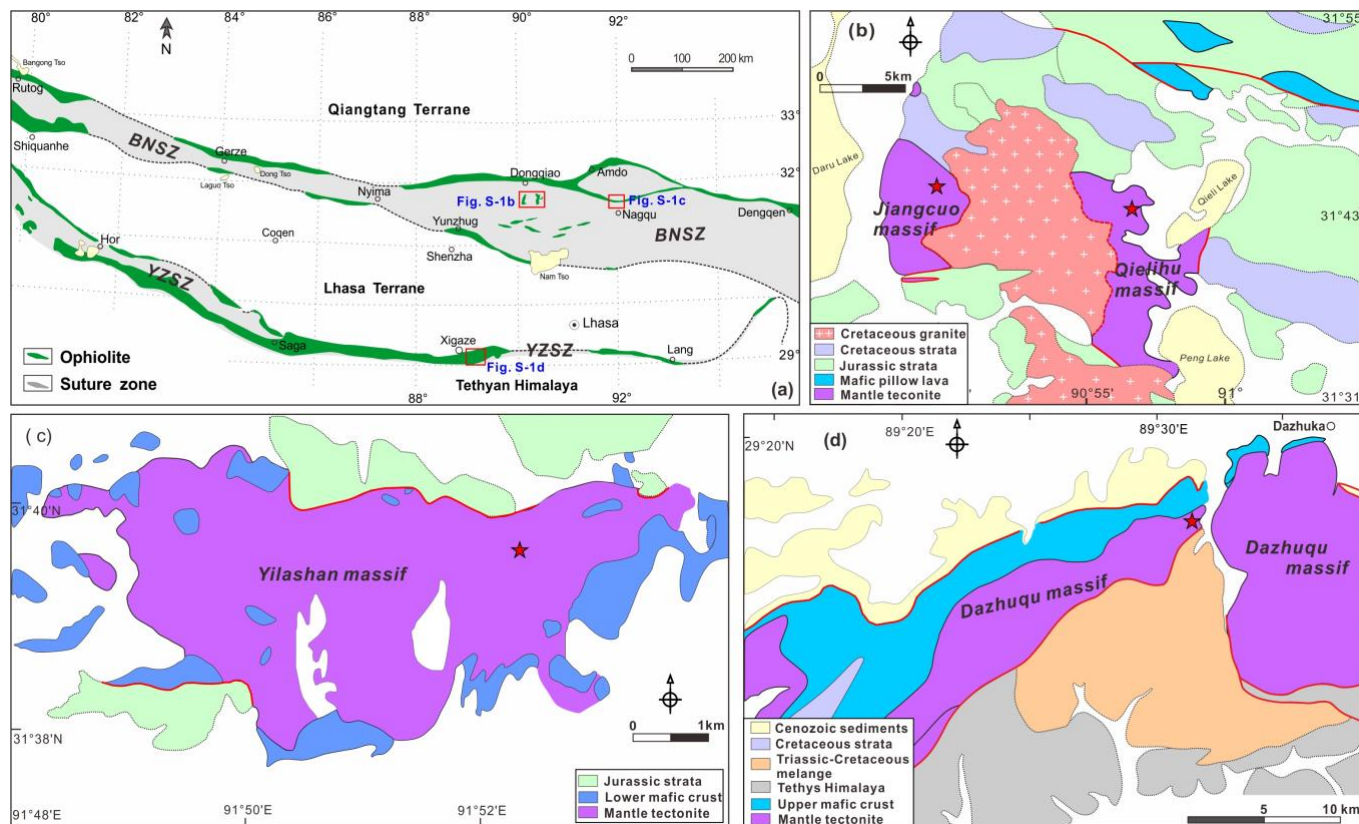


Figure S-1 (a) Tectonic framework of Tibet showing the distribution of ophiolites within two major suture zones (the Bangong-Nujiang and the Yarlung Zangbo sutures). (b-d) Simplified geological maps of the Qielihu (QLH), Jiangcuo (JC), Yilashan (YLS) and Dazhuqu (DZ) ophiolite massifs. Red stars mark the sampling locations in each massif.



Figure S-2 Hand specimen photographs of studied chromitite and chromite-rich dunite (21DZ02) samples.

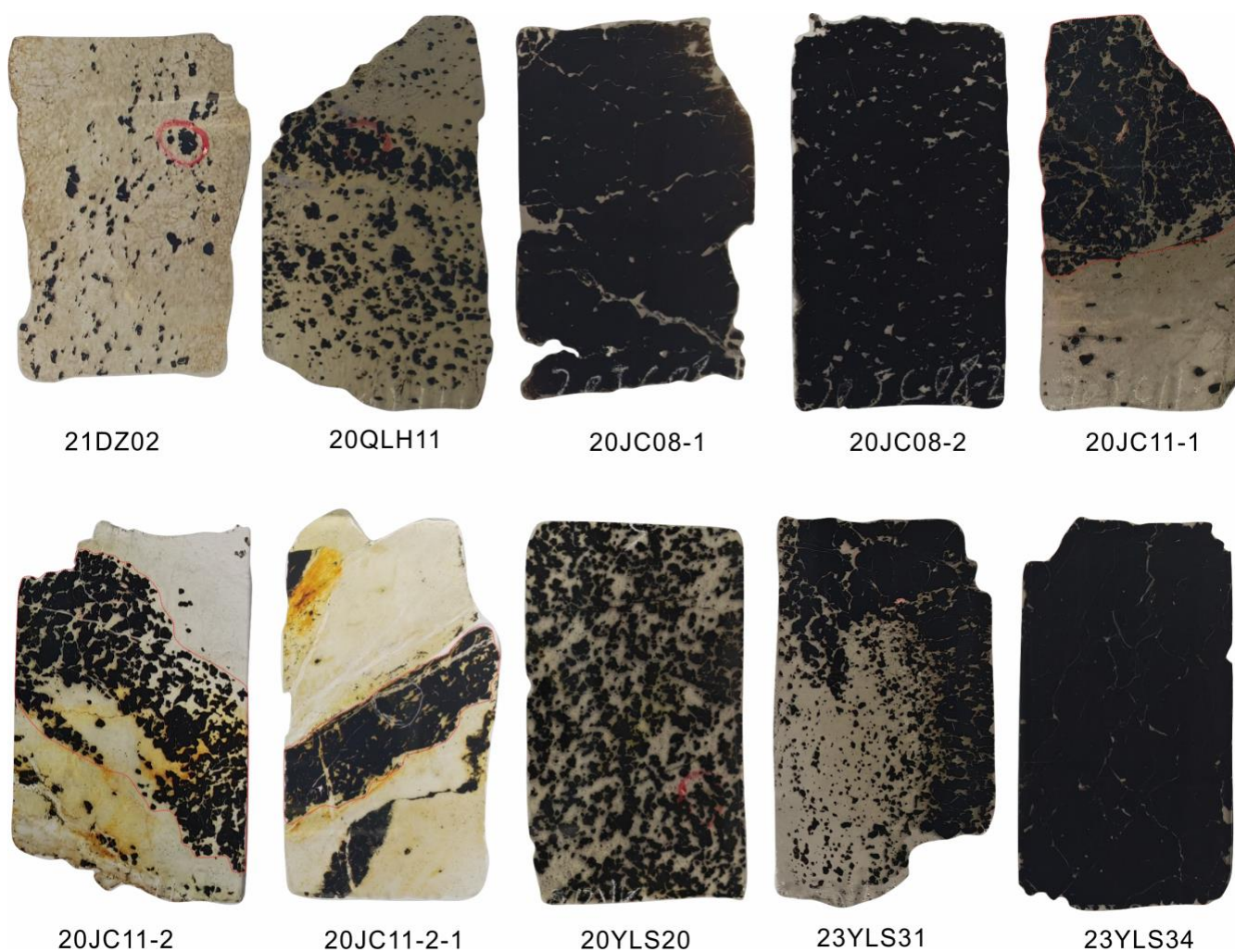


Figure S-3 Thin section photographs of studied chromitite and chromite-rich dunite (21DZ02) samples. Note that 20JC11-2 and 20JC11-2-1 were cut from the same rock sample.

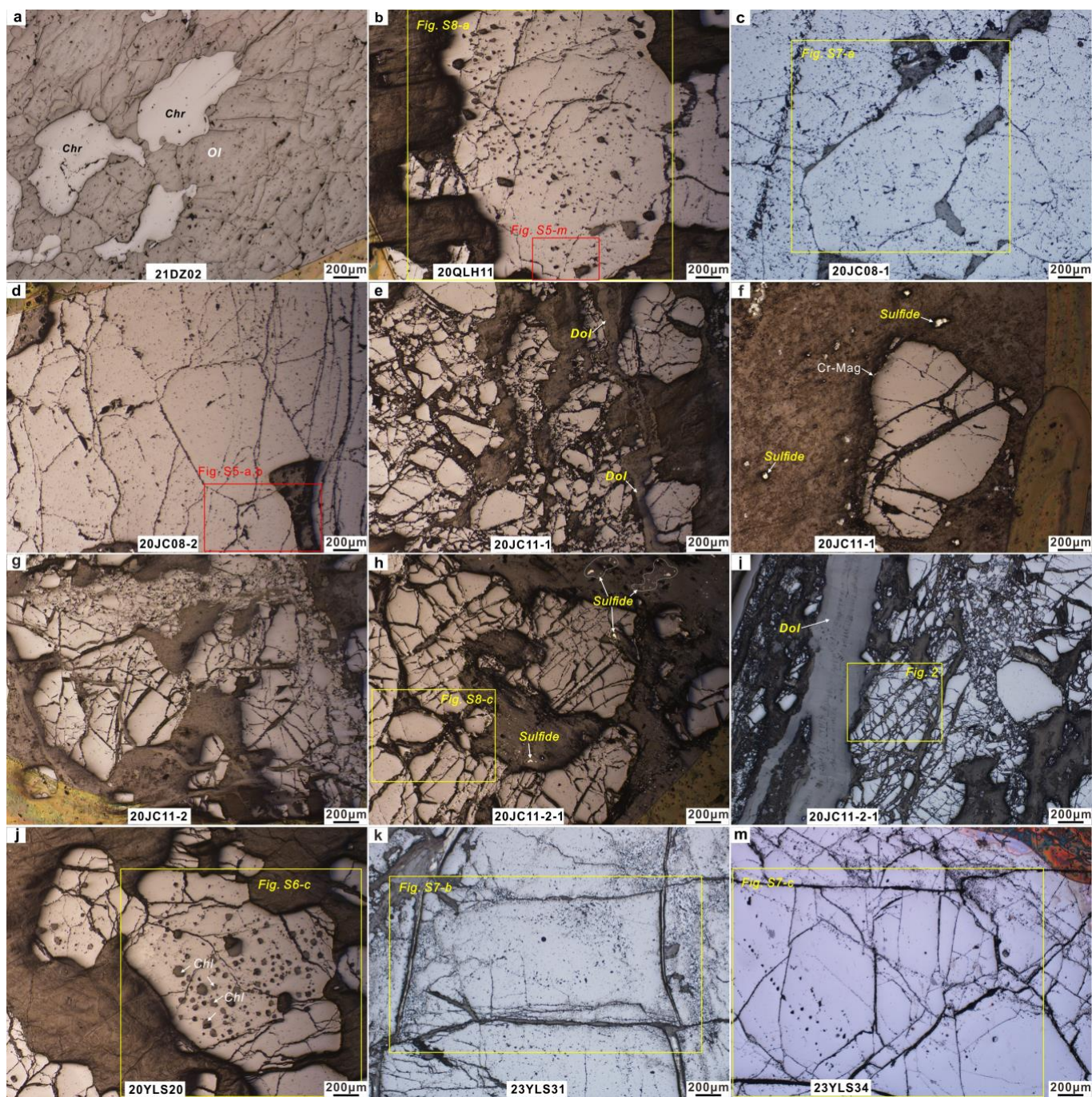


Figure S-4 Microstructures of chromite in the studied chromitite and chromite-rich dunite (21DZ02) samples. **(a)** Fresh Cr-spinel and olivine in the dunite. **(b)** Chromites in disseminated chromitites showing sub-rounded to anhedral shapes, with abundant inclusions (serpentine+chlorite+amphibole). **(c, d, k, m)** Chromites in massive chromitites exhibiting subhedral shapes, with interstitial chlorites. **(e-i)** Chromites in highly fragmented chromitite samples showing cataclastic textures. Note that dolomite veins and sulfides are observed within the serpentine matrix. **(j)** Chromites in disseminated chromitite showing sub-rounded to anhedral shapes, with abundant chlorite inclusions.

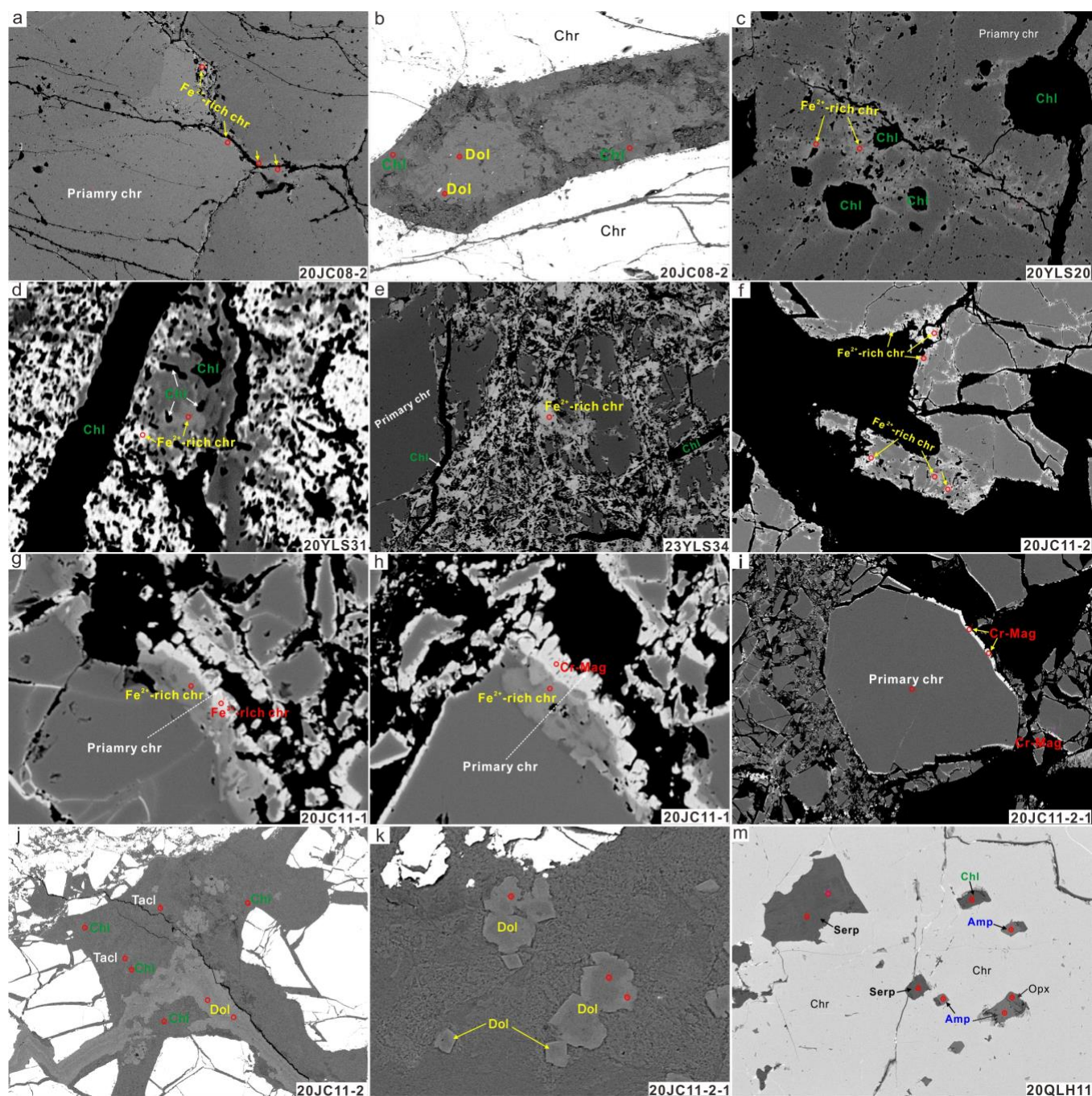


Figure S-5 BSE images showing various secondary phases in studied chromitites. **(a)** Minor Fe-enriched zones (ferrous chromite) formed along margins of primary chromite. **(b)** Secondary dolomite+chlorite formed between chromites. **(c)** Secondary chlorite inclusions and Fe-enriched zones (porous ferrous chromite+chlorite) formed within primary chromite. **(d-f)** Partially to completely replacement of primary chromites by secondary Fe-enriched zones (porous ferrous chromite+chlorite). **(g)** Primary chromite successively rimmed by ferrous chromite and then ferric chromite. **(h)** Primary chromite successively rimmed by ferrous chromite and then Cr-magnetite. **(i)** Primary chromite directly rimmed by Cr-magnetite, with a sharp contact. **(j)** Secondary dolomite+chlorite+taenite in the matrix. **(k)** Dolomite grains within the serpentine matrix. **(m)** Secondary serpentine+amphibole+chlorite inclusions within primary chromite.

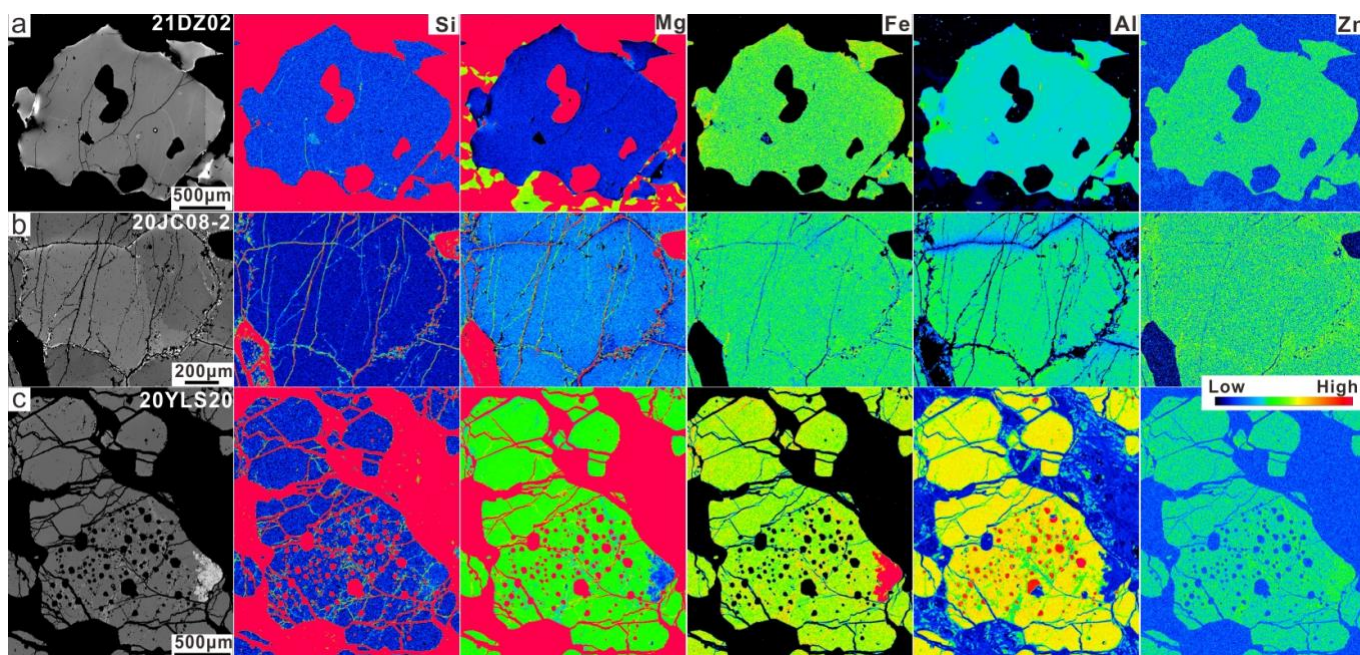


Figure S-6 Elemental distribution maps of selected chromite from Group-1 chromitites and chromite-rich dunite. Chromite in these samples are least affected by alteration, with limited Zn enrichment in secondary Fe-enriched zones (ferrous chromite) compared with corresponding core regions.

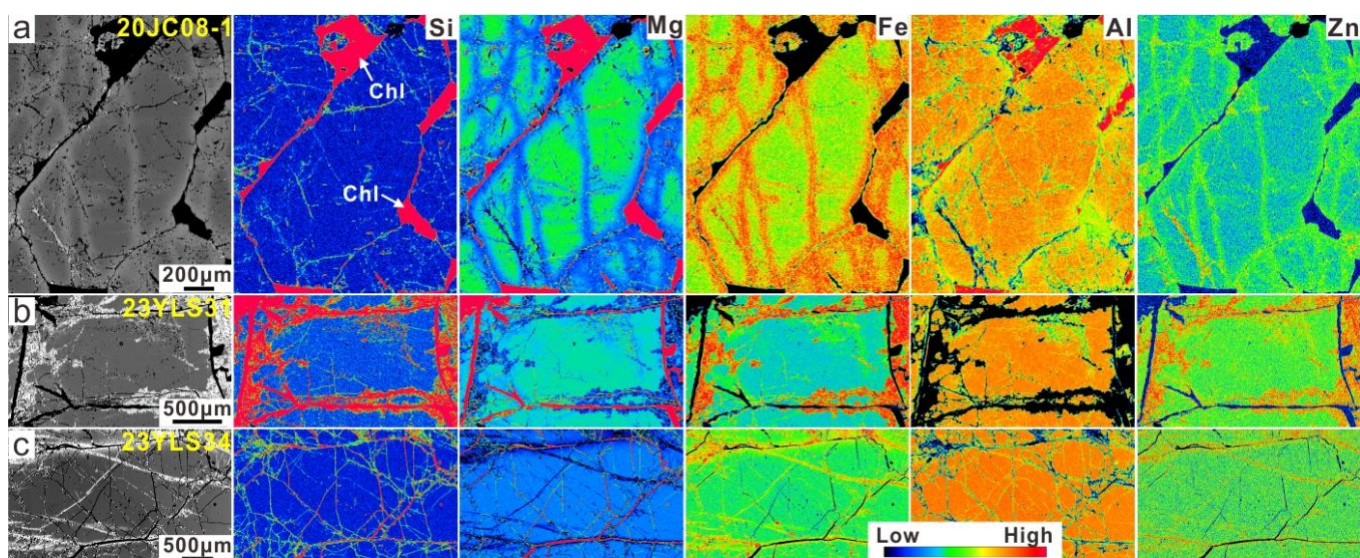


Figure S-7 Elemental distribution maps of chromite from Group-2 chromitites. Chromite in these samples are more heavily affected by alteration, with heterogeneously (moderately) enriched Zn in secondary Fe-enriched zones (ferrous chromite) compared with corresponding core regions.

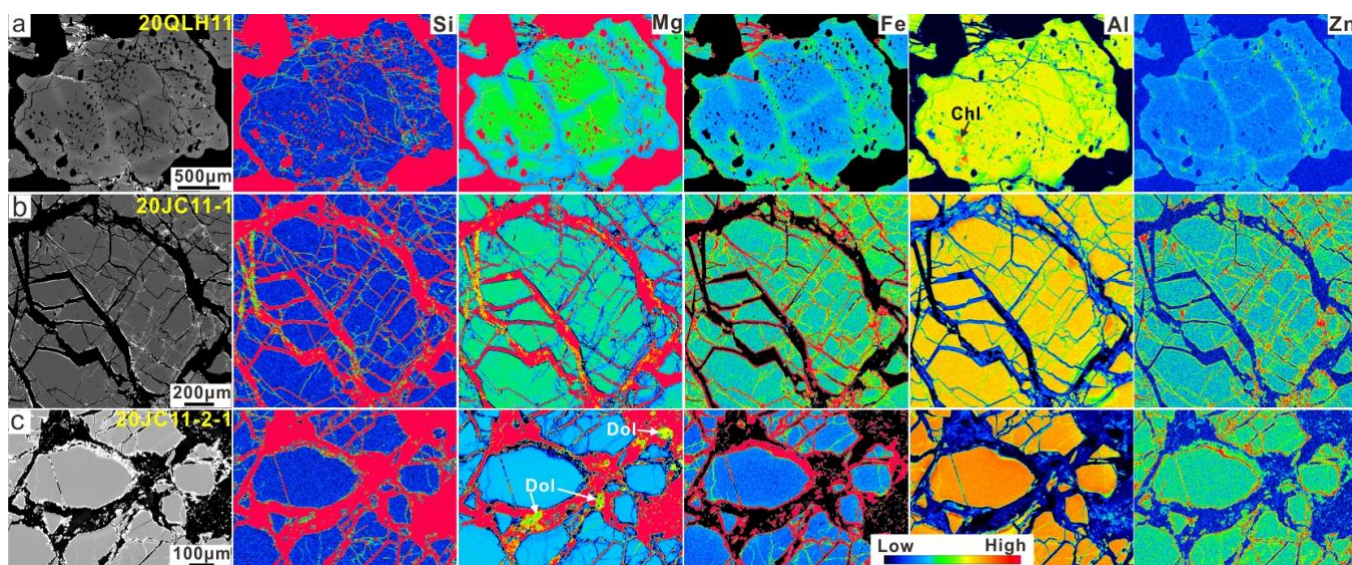


Figure S-8 Elemental distribution maps of chromite from Group-3 chromitites. Chromite in these samples are more heavily affected by alteration, with heterogeneously (highly) enriched Zn in secondary Fe-enriched zones (ferrous-, ferrian-chromite, and Cr-magnetite) compared with corresponding core regions.

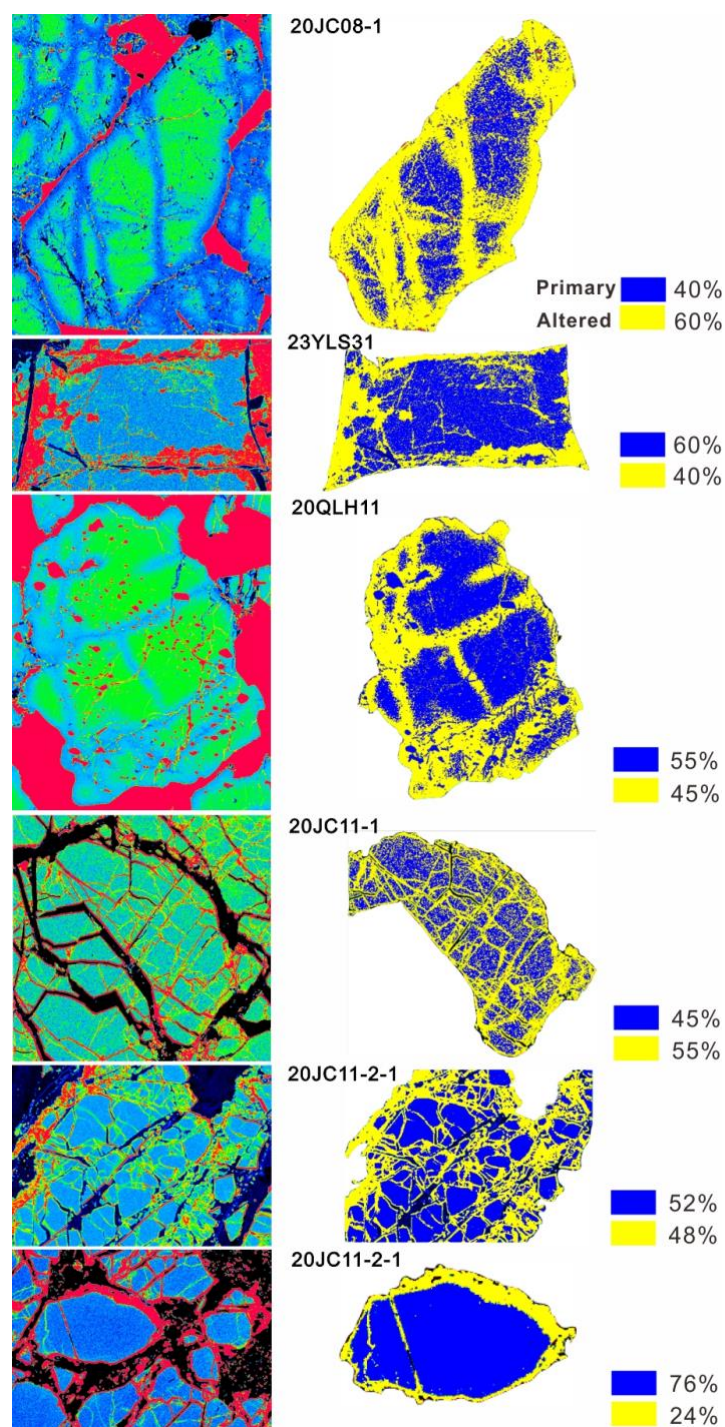


Figure S-9 Estimated relative area proportions (%) between altered Fe-enriched zones and primary cores for selected chromites from Group-2 and 3 samples. The relative area proportions were analysed using Image-Pro plus software, based on elemental distribution maps.

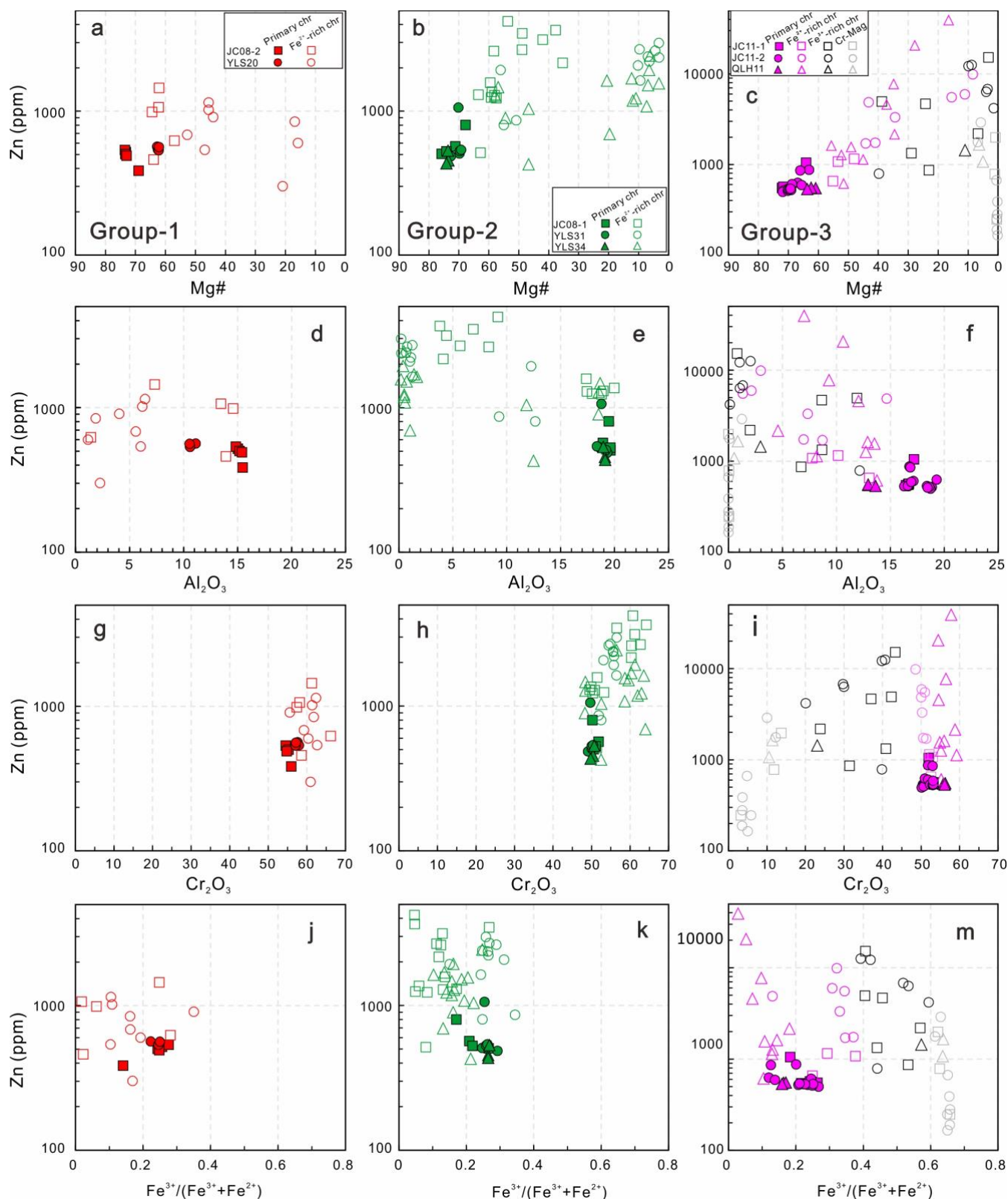


Figure S-10 Compositional variations of primary and altered chromites in different sample groups. (a-c) Zn versus Mg#; (d-f) Zn versus Al_2O_3 ; (g-i) Zn versus Cr_2O_3 ; (j-m) Zn versus $\text{Fe}^{3+}/(\text{Fe}^{3+}+\text{Fe}^{2+})$.

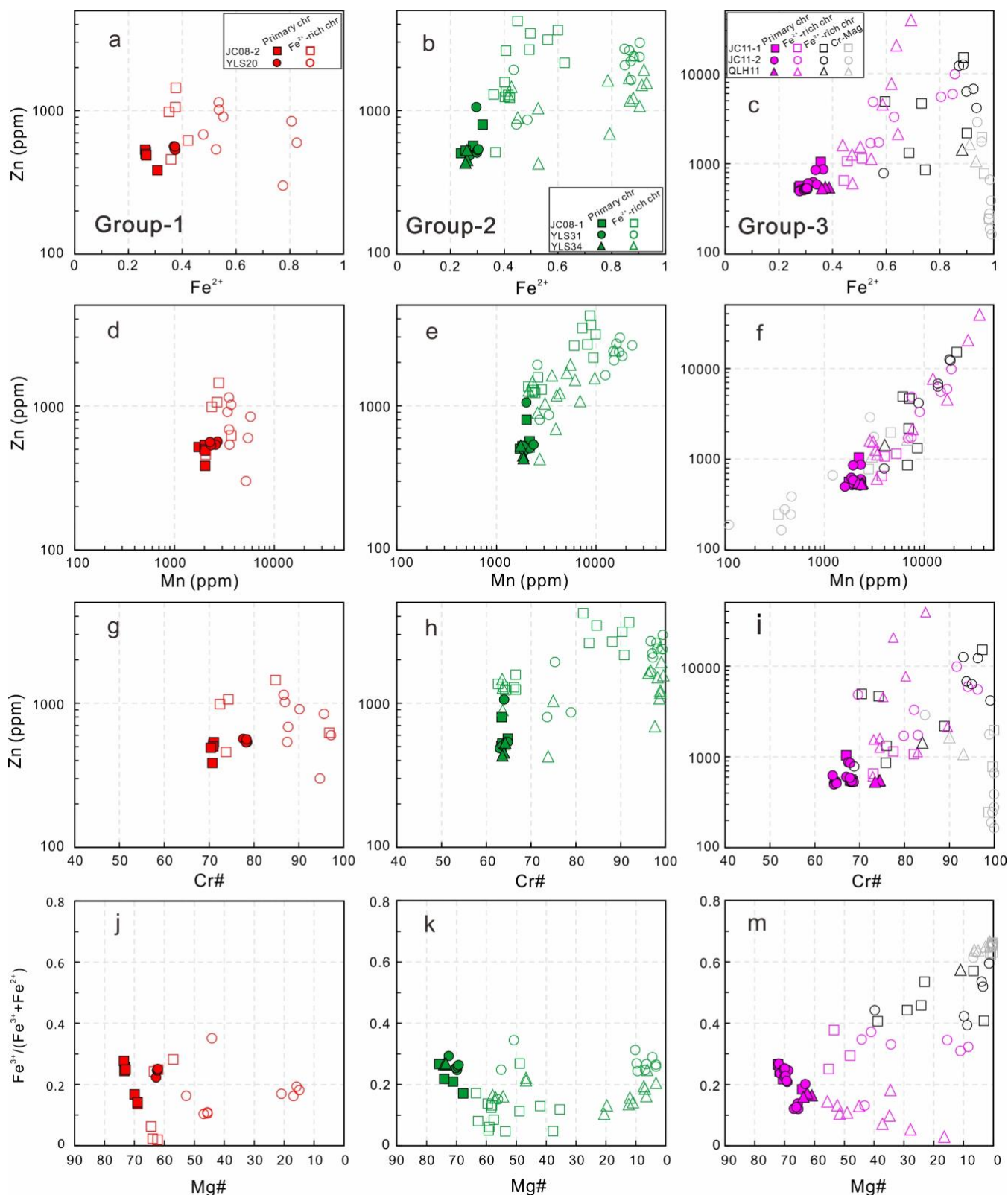


Figure S-11 Compositional variations of primary and altered chromites in different sample groups. (a-c) Zn versus Fe^{2+} ; (d-f) Zn versus Mn; (g-i) Zn versus Cr#; (j-m) $\text{Fe}^{3+}/(\text{Fe}^{3+}+\text{Fe}^{2+})$ versus Mg#.

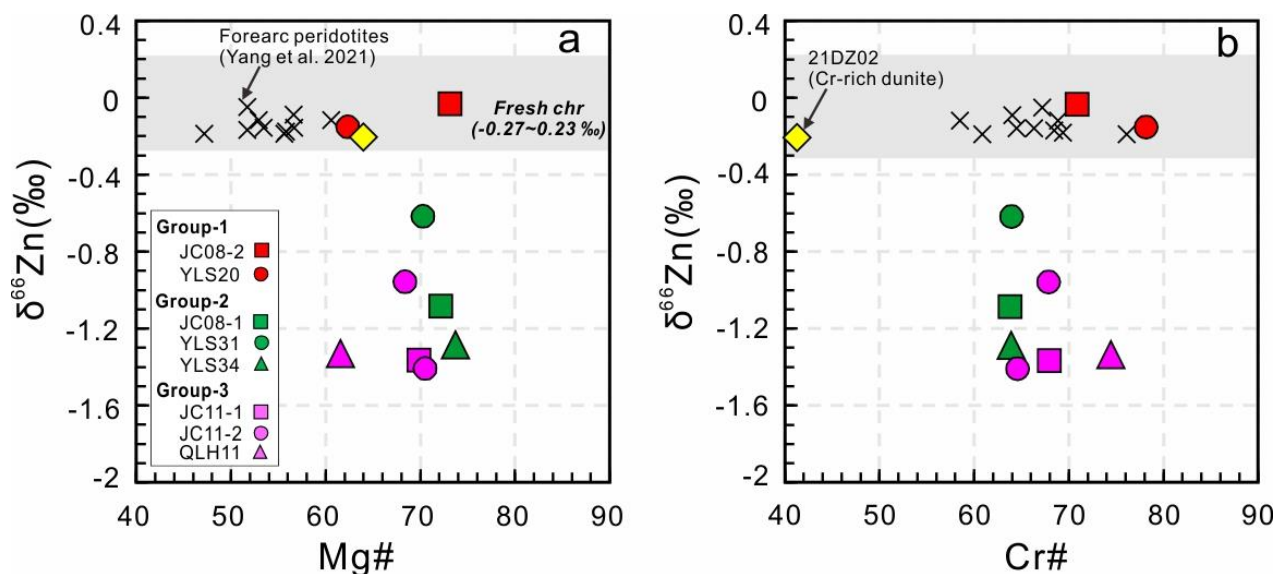


Figure S-12 $\delta^{66}\text{Zn}$ versus Mg# (a) and Cr# (b) of primary chromite from the studied chromitites and dunite. $\delta^{66}\text{Zn}$ range of fresh/primary chromites from forearc ophiolitic peridotites and chromitites (Yang *et al.*, 2021; Lian *et al.*, 2024; Su *et al.*, 2026) is also shown.

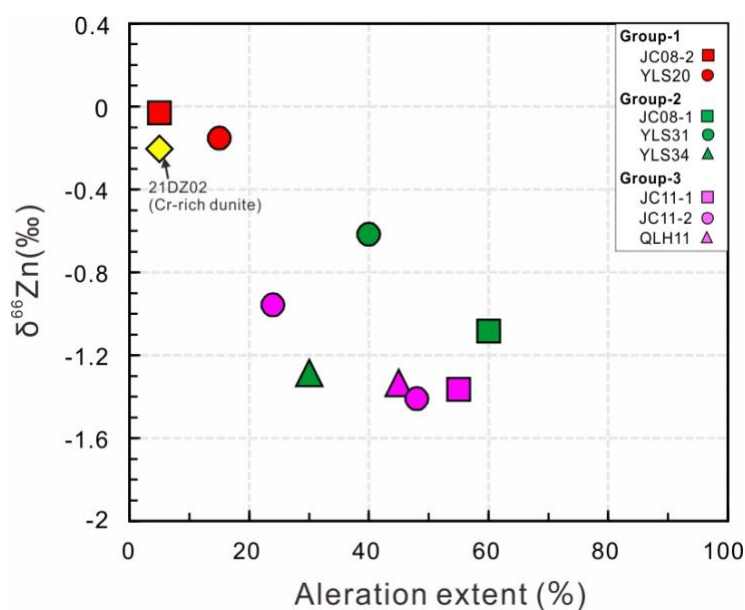


Figure S-13 $\delta^{66}\text{Zn}$ versus Alteration extents (%) of chromite for the studied chromitites and dunite. The estimated alteration extent of chromite in each sample (see Table S-1) was based on elemental distribution maps presented in Fig. S-9.

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