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## REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

Dear Authors,

This is the manuscript dealing with chromite deposits from the Zambales ophiolite, Philippines. The authors provided mineral compositional data as well as bulk Os isotope geochemistry for the peridotite and chromitite samples collected from both Coto and Acoje blocks of the Zambales ophiolite and compared them with ophiolitic chromitites worldwide.

Briefly, the authors suggest that the high-Al and high-Cr chromite deposits occurred after MORB-like magmatism, and during the tholeiitic and boninitic-like magmatism, respectively, produced by the subsequent flux melting associated with subduction initiation. They also prove that the proto-forearc mantle shows increased contribution of asthenosphere but decreased slab-derived component from high-Al to high-Cr chromitites formation, considering the more radiogenic Os isotopic composition of high-Al chromitites compared to the high-Cr chromitites due to the asthenospheric upwelling in the context of slab rollback. Especially the high-Al chromitites have long been considered to have been produced in Mid-Ocean-Ridge or back-arc-basin environments. However, the authors perfectly explain and model the high-Al chromitites formation at the earliest stage of subduction initiation. The proposed model is well-supported by the data the authors provided. Therefore, this paper, for the first time, makes it possible to clarify the genetic relationships among different types of chromitites and their associated peridotites. Relevant findings will greatly improve our knowledge of the origins of podiform chromitites, ophiolites and subduction initiation.

Although the manuscript is very well-written and organized, I provide below some moderate comments which, I believe, help authors to make the manuscript better in science.

L67: Provide a brief reason explaining why high-Al chromitites are unlikely products of typical MORB-like magmatism.

L106: Any criteria for defining cpx-poor lherzolite?

L114: A linguistic problem here. The sentence should probably be “dunite veins with high-Cr chromite grains were found cross-cutting harzburgites that were modified by parental magmas of high-Al chromitites” based on the diagrams of Fig. 4. Although the high TiO<sub>2</sub> contents of chromite grains in the harzburgite support that the harzburgite must have been modified by tholeiitic magmas (parental magmas of high-Al chromitites), it is better to offer more details here or in the figure caption to make the process easier to be understood by readers from other unrelated fields.

L155: I suggest that the authors compile more data regarding this line and add one figure (e.g., TiO<sub>2</sub> vs. Cr#) in the supplementary file to support the large TiO<sub>2</sub> variation of chromite in high-Al chromitites worldwide, which will make the statement here more persuasive.

L106: How to define low-Ti tholeiitic magmas? Based on the illustration in Figure 5A, is it better to use “depleted tholeiitic” or “ultra-depleted tholeiitic” instead?

L191: “appears” or “appeared”?

L207-208: Rephrase the sentence “at 45-43 Ma, significantly less than the half-life of <sup>187</sup>Re”. It reads a little strange.

L219-220: Although it is common sense that high-Cr chromitites always have higher Os concentrations than high-Al chromitites, it would be better to compile relevant data from previous publications and add

one figure (e.g., Os vs. Cr#) in the supplementary file.

L242: According to the work of Liu et al. (2018, *Geology*) on peridotites from the New Caledonia ophiolite, Os may not be effectively mobilized from subducted slabs at the depth of forearc mantle. My own experience is that the conclusion seems fine when the studied samples were peridotites, but it cannot explain why ophiolitic chromitites from worldwide sometimes show supra-chondritic Os isotopic features, which indicate that slab-derived Os must have been involved in the mantle sources of proto-forearc magmatism. By using the two-stage model of early hydration by slab-derived fluids and later refertilization by asthenospheric melts, this study well resolved such an inconsistency. The postulation also well matches the results of petrological observation and could be a highlight of this study, which should be but was not presented very well in this section. I strongly suggest that the authors emphasize such a finding with more lines in the main text.

L276: Reaction with all kinds of mafic melts can elevate the contents of Al<sub>2</sub>O<sub>3</sub> and REE of clinopyroxene (cpx) grains in peridotites. It is more important to point out if the cpx grains are more depleted in LREE than in HREE, which would support if the reactant melts were originated from asthenospheric mantle or not.

L327: Point out why it is impossible to define the formation temperature of melts. For example, the exact water contents in the mantle sources of magmas are difficult to be determined.

L339: I agree that we have no effective methods at the moment for constraining the exact formation temperatures of different magmas, and using the results of experimental and melt inclusion studies would offer us good references on this problem. In general, it is fine to claim that there is a heating period from the formation of high-Al chromitites to high-Cr ones, but I would suggest ending the paragraph with the general conclusion first before applying the conclusion to the chromite deposits of this study. This is because the data on temperatures mentioned in the paragraph were compiled from previous publications rather than the authors' own findings.

L350: Use the plural form of "concentration".

L353: It is better to clarify here that olivine is the earliest crystallization phase from Mg-rich magmas, and their captured melt inclusions preserve the primary compositions of magmas.

L381: It should be pointed out that the Ti-rich high-Al and the Ti-poor high-Al chromitites were from the Coto block.

L412: Add "ophiolitic blocks" before "meanwhile".

L436: It is great to propose that high-Cr chromitites were possibly the products of high-Ca boninitic magmatism, but more information can be mentioned here for the low-Ca boninitic magmatism, for the sake of better understanding the whole evolutionary process of subduction initiation.

L417-418: Use the plural form of "pathway" and the single form of "ophiolite". More details can be offered here to explain the atypical cases.

L961: I think the figure caption can be simplified. The main text (L327-341) regarding Fig. 8 reads much clearer than this figure caption.

Fig. 5b caption: Ibrahim et al. 2009  $\diamond$  Uysal et al. 2009.

Fig. 5b: Turkey  $\diamond$  Turkey.

Also, what do the circles and triangles for in Fig. 5b? Please explain on figure or in the caption. In this diagram, the TiO<sub>2</sub>\_melt composition for high-Al chromitites was calculated using the regression line produced by melt inclusion-host chromite data from MORB; therefore, parental melt TiO<sub>2</sub> composition of these chromitites would have been re-calculated (?) using the regression line of arc proposed in Rollinson (2008) to compare the data with your own as you propose that the high-Al chromitites are the

product of subduction initiation, not MORB. I would recommend also checking the caption for Fig. 5b, and explaining all the details in the caption.

Reviewer #2 (Remarks to the Author):

The manuscript titled "Chromite Mineralization Variabilities Arising from Chemo-Thermal Evolution in Mantle During Subduction Initiation" by Zhang and coauthors explores the elemental and Os isotopic compositions of different chromitites within the Zambales ophiolite and compares them with other ophiolitic chromitites worldwide, particularly with ophiolites that host both high-Cr and high-Al chromitites. This paper investigates the processes involved in the formation of chromitites during subduction initiation, contributing to an ongoing scientific debate. However, notable concerns warrant attention before considering this manuscript for publication in Nature Communications.

Firstly, there is a substantial contradiction regarding the processes occurring during subduction initiation. The paper proposes that radiogenic Os components in mantle sources of high-Al chromitites may originate from sinking slabs during subduction initiation, challenging the traditional view of continuously intensified slab dehydration during this process. However, existing knowledge suggests minimal fluid contribution from the subducting plate during the formation of high-Al chromitites, such as those akin to fore-arc basalts (FAB), as documented by Ishizuka et al. (2014) and Whattam and Stern (2011). The authors must address this inconsistency in light of the well-established magmatic evolution associated with subduction initiation, including FAB, boninites, and Island Arc Tholeiites (IAT).

Ishizuka, O., Tani, K., & Reagan, M.K. (2014). Izu-Bonin-Mariana forearc crust as a modern ophiolite analogue. *Elements*, 10, 115-120. DOI: 10.2113/gselements.10.2.115.

Whattam, S.A., Stern, R.J., 2011. The 'subduction initiation rule': a key for linking ophiolites, intra-oceanic forearcs, and subduction initiation. *Contributions to Mineralogy and Petrology* 162, 1031-1045. DOI 10.1007/s00410-011-0638-z.

Additionally, the interpretation that decreasing  $^{187}\text{Os}/^{188}\text{Os}$  ratios from high-Al chromitites to high-Cr chromitites can be explained solely by modification of asthenospheric melts requires further substantiation. The authors should provide more evidence and discuss other factors influencing the Os isotopic composition of chromitites, including mantle source heterogeneity, the degree of partial melting, extent of melt-rock interactions, and local sulfur fugacity, as suggested by numerous references (e.g., Hattori and Hart 1991; Burton et al. 1999; Brandon et al. 2000; Meibom et al. 2002; Walker et al. 2002; Büchl et al. 2004a, 2004b; Malitch 2004; and others listed in Gervilla et al. 2005).

Brandon, A.D., Snow, J.E., Walker, R.J., Morgan, J.W., Mock, T.D. (2000)  $^{190}\text{Pt}$ – $^{186}\text{Os}$  and  $^{187}\text{Re}$ – $^{187}\text{Os}$  systematics of abyssal peridotites. *Earth Planet Sci Lett* 177:319–335

Büchl, A., Brüggemann, G., Batanova, V.G. (2004a) Formation of podiform chromitite deposits: implications from PGE abundances and Os isotopic compositions of chromites from the Troodos complex, Cyprus. *Chem Geol* 208:217–232.

Büchl, A., Brüggemann, G., Batanova, V.G., Hofmann, A. (2004b) Os mobilization during melt percolation. The evolution of Os isotope heterogeneities in the mantle sequence of the Troodos ophiolite, Cyprus.

Geochim Cosmochim Acta 68:3397–3404

Burton, K., Schiano, P., Birck, J.-L., Allegre, C.J. (1999) Osmium isotope disequilibrium between mantle minerals in a spinel-lherzolite. *Earth Planet Sci Lett* 172:311–322

Gervilla, F., Proenza, J.A., Frei, R., González-Jiménez, J.M., Garrido, C.J., Melgarejo, J. C., Meibom, A., Díaz-Martínez, R., Lavaut, W. (2005) Distribution of platinum-group elements and Os isotopes in chromite ores from Mayarí-Baracoa Ophiolitic Belt (eastern Cuba). *Contrib. to Mineral. Petrol.* 150, 589–607.

Hattori, K., Hart, S.R. (1991) Osmium-isotope ratios of platinum group minerals associated with ultramafic intrusions: Os-isotope evolution of the oceanic mantle. *Earth Planet Sci Lett* 107:499–514

Meibom, A., Sleep, N.H., Chamberlain, C.P., Coleman, R.G., Frei, R., Hren, M.T., Wooden, J.L. (2002) Re–Os isotopic evidence for long lived heterogeneity and equilibration processes in the Earth's upper mantle. *Nature* 419:705–708

Walker, R.J., Prichard, H.M., Ishiwatari, A., Pimentel, M. (2002) The osmium isotopic composition of the convecting upper mantle deduced from ophiolite chromites. *Geochim Cosmochim Acta* 66:329–345

The speculation surrounding the presence of three types of peridotites (cpx-poor Hz1, cpx-rich Hz2, and cpx-poor Hz3) based on limited evidence needs further substantiation. The authors should provide additional supporting data for the presence of these peridotite types and the transformation from Hz1 to Hz2.

The temperature variations during different types of chromite mineralization are inconsistent with previously published data, such as those related to Forearc Basalt (FAB) temperatures, as discussed by Whattam et al. (2020, AM). The authors should reconcile this inconsistency with existing data.

The sections titled "Origins of different types of chromite deposits" and "Geodynamic implication" lack sufficient support from the data presented. For instance, the statement regarding a "climax stage of slab impact in the proto-forearc mantle" requires clarification, especially in light of the minimal fluid contribution from the subducting plate during subduction initiation, as documented in Ishizuka et al. (2014) and Whattam and Stern (2011).

In conclusion, this manuscript by Zhang and colleagues requires substantial revisions and additional supporting evidence before it can be considered for publication in *Nature Communications*. The proposed model must be rigorously tested against the existing body of knowledge related to subduction initiation and chromitite formation. It is my sincere hope that these comments will aid the authors in improving the quality and robustness of their work.

Minor suggestions/corrections

INTRODUCTION

- Lines 31-34: the sentence is unclear

- Line 46: delete "extraction"

- Line 48: Cr# is not referred as "wt%", correct the statement. In addition, with Cr#=7 the mineral probably cannot be classified as chromite

- Line 50: include the reviews on chromitites by González-Jiménez et al.

González-Jiménez, J. M., Griffin, W. L., Gervilla, F., Proenza, J. A., O'Reilly, S. Y., & Pearson, N. J. (2014).

Chromitites in ophiolites: How, where, when, why? Part I. A review and new ideas on the origin and significance of platinum-group minerals. *Lithos*, 189, 127-139.

González-Jiménez, J. M., Griffin, W. L., Proenza, J. A., Gervilla, F., O'Reilly, S. Y., Akbulut, M., ... & Arai, S. (2014). Chromitites in ophiolites: How, where, when, why? Part II. The crystallization of chromitites. *Lithos*, 189, 140-158.

- Line 54: correct the reference to Xiong et al.
- Lines 55-59: include the references to support this statement
- Line 57: Figure 1 shows the range of Cr# in chromitites and associated rocks. It does not include the origin of the peridotites or the presence of diopsidite veins. Change the sentence, the reference to the figure, or change the figure.
- Line 58: add "formation of" before "high-Cr chromite mineralization"
- Lines 59-60: include references for this statement
- Line 66: indicate examples of this type of subduction-initiation ophiolites
- Line 89: the reference to Fig. 2B-D in this sentence is not pertinent
- Line 101: the abbreviation of Cpx should be capitalized (in the whole text)
- Lines 113-116: the sentence is not clear, indicate that this is an interpretation from previous works. The presence of high-Cr chromite in dunites in the ophiolitic sequence is typical, hence it is not that straightforward that it indicates that high-Cr chromitites are younger than the high-Al ones
- Line 116: figure 4 is not clear and maybe not needed. Explain the figure (including the different pictures and diagrams) or change it

#### RESULTS AND DISCUSSION

- Line 130: it is better to use the term "ophiolitic chromitites" instead of "podiform chromitites"
- Lines 153-155: include the used equations to estimate the melts in equilibrium
- Line 159: indicate the composition of boninite used to compare and the reference
- Line 161: indicate the compositions of FAB to low-Ti tholeiites used to compare and the references
- Line 161: figure 5C should have been cited before 5D, figure 5C is not cited in the text, correct or remove the figure
- Line 181: explain the model that has been carried out to obtain these values
- Line 254: indicate the reference for the Os isotopic values for abyssal peridotites
- Line 257: for the chromitites in Mugla, Turkey, it is not that clear that the high-Al chromitites have much higher Os isotopic ratios
- Lines 263-264: add reference(s) for the statement
- Lines 288-289: add reference(s) for the statement
- Lines 387-389: the authors should indicate the chromitites they are referring to
- Line 412: the word "meanwhile" does not make sense in the sentence
- Lines 430: authors do not need to invoke slab roll-back to generate boninitic-like melts

#### METHODS

- Line 452: indicate how many samples of each and the coordinates
- Line 995: indicate the meaning of "LAB"

#### FIGURES

- Line 958: correct the reference
- Figure 1: Medellín is in Colombia, not Columbia
- Figure 5: in B correct the country "Turkey", C is not cited in the text
- Figure 6: the orange color in the legend does not match the yellow in the used symbol in A.

- Figure 7: the figure would be easier to understand if the authors used the same scale for all the chromitites and differentiate those classified as high-Cr and high-Al with different colors

Reviewer #3 (Remarks to the Author):

Comments:

This manuscript reports high-Al to high-Cr chromitites from classical Zambales ophiolite, Philippines. The elemental and Os isotopic compositions were presented to discuss parental magmas of chromitites and the modification from upwelling deep asthenospheric materials and sinking slab. Finally, mineralization conditions and stages of different types of chromitites in the Zambales proto-forearc mantle were well illustrated. This ms generally well-written, well-organized, and clearly argued. Below list major and minor comments, which are hopefully useful during revision.

Major comments:

1-- For discussion part 1, maybe it is more clearly to discuss the parental magmas of chromitites from the Acojie and Coto blocks, separately, since the various types of chromitites may likely to make readers confused. A schematic diagram could be useful to show how FMM experiences 5-10% or 10-20% melting to mantle sources and then experiencing divers degrees of melting to parental magmas of various types of chromitites from the Acojie and Coto blocks, separately.

2--You should make it clear when say high-Al chromitites, since both Acojie and Coto has them. This equivocal expression is all through your manuscript. The related statements should be clarified in revision.

3-- Peridotites in Acojie and Coto also have chromites, how their Cr# and Ti values correlate to that of chromitites? Especially in Acojie, chromitites were rounded by dunites as showed in Fig.3. what is the relationship between dunites and chromitites?

Specific comments:

1. In figure 3. I do not understand the meaning of ordinate axis form 8Km to 1Km. Does this mean the depth of samples used? If so, please make it clear which rock type (chromitites, dunites, or cpx-poor harzburgites) are these chromites and clinopyroxenes from.

2. No scale in figure 4.

3. In figure 5B, what is the hollow triangle represent?

4. In figure 7. It is hard for me to gain the point that high-Al chromitites always show higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than high-Cr chromitites, especially for Mugla Turkey and Masquad Oman.

5. In Figure 6. The color is not uniformed for Cpx-poor harzburgite between samples and legend.

line 114: dunites and harzburgites are adjacent, but no dunite vein could be seen in Figure 4. Please give

a more appropriate figure to show cutting relationship.

Line 180: Both Acojie and Coto blocks have High-Al chromitites, while showing different Cr# number ranges. Should you discuss their parental magmas respectively?

Line 180: As you said above, the parental magmas of high-Al and high-Cr chromitites were formed by melting of peridotites under relatively low (S-saturated) and high (S-undersaturated) degrees, respectively. Why the parental magmas of the high-Cr chromitites and Ti-rich high-Al chromitites were generated from the similar mantle sources which had experienced ~ 5-10% melting of fertile MORB mantle (FMM)?

Line 190: the Ti-rich high-Al chromitites you discussed throughout this paragraph are only from the Acojie block, or both Acojie and Coto? If only from the Acojie, how to interpret parental magmas of Ti-rich high-Al chromitites from the Coto block?

Line 192: The Ti-poor high-Al chromitites were exclusively found in the Coto block, along with Ti-rich high-Al chromitites. Which process makes the variety of their Ti values?

Line 195: The tradition view is that the Cr# numbers could indicate the melting degrees of mantle sources. While it seems that Ti is crucial when evaluating the melting degree in your discussion. So how to balance the two indicators?

Line 256: Your point of view here is just suitable for the case of the Acojie block, where high-Al chromitites have more Os isotopic ratios than that of high-Cr chromitites, but not the case of the Coto block where most high-Al chromitites show Os isotopic ratios comparable to abyssal peridotites, not elevated.

Line 259: in figure 6A, the highest Os isotopic ratios are from high-Al chromitites in the Acojie block, not the Coto block. Does it mean intensified slab dehydration during subduction initiation?

Line 279: Both high-Al chromitites and high-Cr ones in the Coto block were altered by asthenospheric melts. Which the former has elevated  $^{187}\text{Os}/^{188}\text{Os}$  ratios?

Line 369: Most high-Al chromitites in the Coto block show  $^{187}\text{Os}/^{188}\text{Os}$  ratios equal to abyssal peridotites, while only few ones have more radiogenic Os isotopes. How to explain the selectivity?

Line 381: "the Ti-rich high-Al chromitites must have formed before the Ti-poor ones." Which block are these high-Al chromitites from, Acojie or Coto? Please make it clear.

Line 404: As showed in figure 6, high-Al chromitites from the Acojie block have more elevated  $^{187}\text{Os}/^{188}\text{Os}$  ratios than these from the Coto block. The effects of slab components on the proto-forearc mantle should be stronger on Coto than Acojie, since the former was altered earlier. How to explain?

Line 414: More radiogenic Os isotopic compositions were brought by slab components, since upwelling



of asthenosphere could not bring supra-chondritic Os isotopic ratios. (See L-244)

Line 416: The re-upwelling process of asthenosphere not only replenished fertile components (e.g., Ca, Al and Ti) (Line 397-398)...since that, why the fertilized mantle could produce high-Cr parental magmas while depleted mantle could not?

## REVIEWER COMMENTS

### Reviewer #1 (Remarks to the Author):

#### Dear Editor and Authors,

This is the manuscript dealing with chromite deposits from the Zambales ophiolite, Philippines. The authors provided mineral compositional data as well as bulk Os isotope geochemistry for the peridotite and chromitite samples collected from both Coto and Acoje blocks of the Zambales ophiolite and compared them with ophiolitic chromitites worldwide.

Briefly, the authors suggest that the high-Al and high-Cr chromite deposits occurred after MORB-like magmatism, and during the tholeiitic and boninitic-like magmatism, respectively, produced by the subsequent flux melting associated with subduction initiation. They also prove that the proto-forearc mantle shows increased contribution of asthenosphere but decreased slab-derived component from high-Al to high-Cr chromitites formation, considering the more radiogenic Os isotopic composition of high-Al chromitites compared to the high-Cr chromitites due to the asthenospheric upwelling in the context of slab rollback. Especially the high-Al chromitites have long been considered to have been produced in Mid-Ocean-Ridge or back-arc-basin environments. However, the authors perfectly explain and model the high-Al chromitites formation at the earliest stage of subduction initiation. The proposed model is well-supported by the data the authors provided. Therefore, this paper, for the first time, makes it possible to clarify the genetic relationships among different types of chromitites and their associated peridotites. Relevant findings will greatly improve our knowledge of the origins of podiform chromitites, ophiolites and subduction initiation.

*Although the manuscript is very well-written and organized, I provide below some moderate comments which, I believe, help authors to make the manuscript better in science.*

**Our reply:** Many thanks for reviewing our paper. Your comments and suggestions are very helpful for improving this paper. We have replied all of them carefully and made revisions in the main text accordingly. Please see them one by one below. Many thanks again for your time and effort to our work.

*L67: Provide a brief reason explaining why high-Al chromitites are unlikely products of typical MORB-like magmatism.*

**Our reply:** Many thanks for the comment. Chromite grains in the high-Al chromitites mostly have lower TiO<sub>2</sub> contents than those hosted in MORB-like magmas, including average MORB, back-arc basin basalts (BABB) and forearc basalts (FAB), indicating the more depleted nature of their parental magma sources than FMM. That is why we claimed high-Al chromitites are unlikely products of typical MORB-like magmatism. We have added the reason into the line accordingly. Please have a check there.

*L106: Any criteria for defining cpx-poor lherzolite?*

**Our reply:** Many thanks for the question. Cpx-rich harzburgites sometimes transition

to lherzolite due to the heterogeneous distribution of Cpx, but the modal% of Cpx in such lherzolites are not that high (usually slightly > 5%) as in fertile MORB mantle. That is why we called them as Cpx-poor lherzolites. We also checked the definition from previous publications and found [lherzolites with 5-10 modal% of Cpx can be called Cpx-poor lherzolites](#). Please see the reference below. We also have added such a criteria in the line.

Petrogenetic evolution of orogenic lherzolite massifs in the central and western Pyrenees

J Fabriès, JP Lorand, JL Bodinier - Tectonophysics, 1998 - Elsevier

... by the garnet pyroxenites and granular lherzolites. Compared to the single decompression and cooling step identified in the EP lherzolite massifs, the CWP ... Cpx-poor spinel lherzolites ...

[Redacted]

*L114: A linguistic problem here. The sentence should probably be “dunite veins with high-Cr chromite grains were found cross-cutting harzburgites that were modified by parental magmas of high-Al chromitites” based on the diagrams of Fig. 4. Although the high TiO<sub>2</sub> contents of chromite grains in the harzburgite support that the harzburgite must have been modified by tholeiitic magmas (parental magmas of high-Al chromitites), it is better to offer more details here or in the figure caption to make the process easier to be understood by readers from other unrelated fields.*

**Our reply:** Many thanks for the reminder. We also realized this problem and have revised the sentence accordingly. Moreover, [relevant information and Fig.4 have been moved to the supplementary materials, based on the suggestions of other reviewers](#). Please have a check there.

*L155: I suggest that the authors compile more data regarding this line and add one figure (e.g., TiO<sub>2</sub> vs. Cr#) in the supplementary file to support the large TiO<sub>2</sub> variation of chromite in high-Al chromitites worldwide, which will make the statement here more persuasive.*

**Our reply:** Many thanks for the suggestion. We have made the compilation and put it in the Supplementary Fig-2A already. Please have a check there.

*L106: How to define low-Ti tholeiitic magmas? Based on the illustration in Figure 5A, is it better to use “depleted tholeiitic” or “ultra-depleted tholeiitic” instead?*

**Our reply:** Many thanks for the suggestion. In the subduction-related context, low-Ti tholeiitic magmas belong to a special group of magmas between MORB-like and boninites, and they were called as different names in publications, e.g., transitional lavas, transitional FAB, depleted FAB, IAT, depleted tholeiitic magmas (Reagan et al., 2010; Morishita, et al., 2011; Golowin et al., 2017; Shervais et al., 2019). As a result, [such magmas have lower TiO<sub>2</sub> contents than MORB-like magmas](#) (FAB and BBAB). We checked previous publications and found many papers did not mention the exact definition (TiO<sub>2</sub> content) for low-Ti tholeiitic magmas. However, the paper Bèbien et al. (2000, Ophiolite) [mentioned a boundary \(upper limit\) value 0.75 wt.% for low-Ti magmas](#). Considering that most FAB have more than 0.8 wt.% TiO<sub>2</sub>, we think that such a value is appropriate for the definition of low-Ti tholeiitic magmas. Actually,

the phrase low-Ti tholeiitic was used more popularly than depleted tholeiitic in literatures. To make a compromise, we explained that low-Ti tholeiitic magmas were called different names in literatures in the 2nd paragraph of the Introduction. Please have a check there.

Albanian ophiolites. I-Magmatic and metamorphic processes associated with the initiation of a subduction

J Bébien, A Dimo-Lahitte, P Vergély... - Ofioliti, 2000 - ofioliti.it

... The volcanic component consists of Tirich tholeiitic basalts. The eastern (E... low-Ti and the very low-Ti groups on the TiO<sub>2</sub> versus FeO\*/MgO diagram (Fig. 3a). Both low-Ti and very low-Ti ...

*L191: “appears” or “appeared”?*

**Our reply:** Many thanks for the suggestion. We have re-written the sentence already. Please have a check there.

*L207-208: Rephrase the sentence “at 45-43 Ma, significantly less than the half-life of 187 Re”. It reads a little strange.*

**Our reply:** Many thanks for the problem. We have revised the sentence as “All the chromitites overall have low Re/Os ratios (between 0.001 and 1) and were formed at ~ 45-43 Ma. Compared with the significant long half-life of <sup>187</sup>Re (<sup>187</sup>Re→<sup>187</sup>Os, ~ 4.3 Gyrs), the Os isotopic systems of the chromitites could not have changed too much after the mineralization”. Please have a checked there.

*L219-220: Although it is common sense that high-Cr chromitites always have higher Os concentrations than high-Al chromitites, it would be better to compile relevant data from previous publications and add one figure (e.g., Os vs. Cr#) in the supplementary file.*

**Our reply:** Many thanks for the suggestion. We have did the compilation accordingly and put it in the Supplementary Fig-2B already. Please have a check there.

*L242: According to the work of Liu et al. (2018, Geology) on peridotites from the New Caledonia ophiolite, Os may not be effectively mobilized from subducted slabs at the depth of forearc mantle. My own experience is that the conclusion seems fine when the studied samples were peridotites, but it cannot explain why ophiolitic chromitites from worldwide sometimes show supra-chondritic Os isotopic features, which indicate that slab-derived Os must have been involved in the mantle sources of proto-forearc magmatism. By using the two-stage model of early hydration by slab-derived fluids and later refertilization by asthenospheric melts, this study well resolved such an inconsistency. The postulation also well matches the results of petrological observation and could be a highlight of this study, which should be but was not presented very well in this section. I strongly suggest that the authors emphasize such a finding with more lines in the main text.*

**Our reply:** Many thanks for the comment. Yes, we quite agree with what you said above. After being modified by asthenospheric melts, proto-forearc mantle peridotites would take on abyssal-like Os isotopic compositions, even though their proto-lithos showed obviously high <sup>187</sup>Os/<sup>188</sup>Os ratios. The paper of Liu et al. (2018) did not take the refertilization process into consideration. That is why they claimed that Os may

not be effectively mobilized from subducted slabs at the depth of forearc mantle. Actually, we discussed such information in 2nd and 3rd paragraphs of the Section “Effects of refertilization on Os isotopes of the proto-forearc mantle”. In particularly, we mentioned at the end of that section that “slab-derived Os still existed in parental magma sources of all the chromitites, regardless of whether their  $^{187}\text{Os}/^{188}\text{Os}$  ratios are nominally abyssal-like or not”. Please have a check there.

*L276: Reaction with all kinds of mafic melts can elevate the contents of  $\text{Al}_2\text{O}_3$  and REE of clinopyroxene (Cpx) grains in peridotites. It is more important to point out if the Cpx grains are more depleted in LREE than in HREE, which would support if the reactant melts were originated from asthenospheric mantle or not.*

**Our reply:** Many thanks for the comment. we quite agree with what you said. In order to better revise the sentence, we have changed the content as “imposed higher  $\text{Al}_2\text{O}_3$  contents and abyssal-like REE patterns on the clinopyroxene”. Please have a check there.

*L327: Point out why it is impossible to define the formation temperature of melts. For example, the exact water contents in the mantle sources of magmas are difficult to be determined.*

**Our reply:** Many thanks for the suggestion. we quite agree with your point above and have added the reason here. Now the sentence has become as “Chromite and olivine are both nominally anhydrous phases. The water contents in the parental magma sources of chromitites are thus difficult to determine, making it impossible to constrain the formation temperatures of the parental magmas precisely”. Please have a check.

*L339: I agree that we have no effective methods at the moment for constraining the exact formation temperatures of different magmas, and using the results of experimental and melt inclusion studies would offer us good references on this problem. In general, it is fine to claim that there is a heating period from the formation of high-Al chromitites to high-Cr ones, but I would suggest ending the paragraph with the general conclusion first before applying the conclusion to the chromite deposits of this study. This is because the data on temperatures mentioned in the paragraph were compiled from previous publications rather than the authors' own findings.*

**Our reply:** Many thanks for the comments. We quite agree with your points here. Based on your instruction, we have revised the sentence as “Consequently, both the experimental and melt inclusion studies suggest parental magmas of high-Cr chromitites were generated under high temperature conditions than those of high-Al chromitites, also supporting presence of a heating period from the formation of high-Al chromitites to that of high-Cr chromitites in ophiolites”. Please have a check there.

*L350: Use the plural form of “concentration”.*

**Our reply:** Many thanks for the reminder. We have made the revision already. Please have a check there.

*L353: It is better to clarify here that olivine is the earliest crystallization phase from Mg-rich magmas, and their captured melt inclusions preserve the primary compositions of magmas.*

**Our reply:** Many thanks for the constructive suggestions. We have added relevant information according to your instruction. Please have a check there.

*L381: It should be pointed out that the Ti-rich high-Al and the Ti-poor high-Al chromitites were from the Coto block.*

**Our reply:** Many thanks for the reminder. We also realize this problem and have revised the sentence accordingly. Please have a check there.

*L412: Add “ophiolitic blocks” before “meanwhile”.*

**Our reply:** Many thanks for the suggestion. We have added the two words into the line already. Please have a check there.

*L436: It is great to propose that high-Cr chromitites were possibly the products of high-Ca boninitic magmatism, but more information can be mentioned here for the low-Ca boninitic magmatism, for the sake of better understanding the whole evolutionary process of subduction initiation.*

**Our reply:** Many thanks for the suggestion. We emphasize the time sequence of the high-Ca boninitic magmatism and low-Ca boninitic magmatism, so that the temporal variation of the mantle fertility could be better understood now.

*L417-418: Use the plural form of “pathway” and the single form of “ophiolite”. More details can be offered here to explain the atypical cases.*

**Our reply:** Many thanks for the corrections. We have revised the forms of the two words already. Moreover, we also added more details to explain the atypical case, which was supposed by us but may not happen exactly.

*L961: I think the figure caption can be simplified. The main text (L327-341) regarding Fig. 8 reads much clearer than this figure caption.*

**Our reply:** Many thanks for the suggestion. We have tried to simplify the caption in the revised version. Please have a check there, and hope you are fine with the revised version.

*Fig. 5b caption: Ibrahim et al. 2009, Uysal et al. 2009.*

**Our reply:** Many thanks for the reminder. Sorry for the mistake. The format of citation has been changed according to the requirement, so we do not need to mention the author name anymore.

*Fig. 5b: Turkey, Turkey.*

**Our reply:** Many thanks for the reminder. Sorry for the mistake. We have deleted relevant content already, since we have compiled more data in the supplementary materials. Please check it in the Fig. 5b.

*Also, what do the circles and triangles for in Fig. 5b? Please explain on figure or in the caption. In this diagram, the TiO<sub>2</sub>\_melt composition for high-Al chromitites was calculated using the regression line produced by melt inclusion-host chromite data from MORB; therefore, parental melt TiO<sub>2</sub> composition of these chromitites would have been re-calculated using the regression line of arc proposed in Rollinson (2008) to compare the data with your own as you propose that the high-Al chromitites are the product of subduction initiation, not MORB. I would recommend also checking the caption for Fig. 5b, and explaining all the details in the caption.*

**Our reply:** Many thanks for the suggestion. Please see our answers below.

The grey circles, red triangles, and green triangles represent the high-Al chromitites from the Coto block, the high-Al chromitites from the Acoje block, and the high-Cr chromitites from the Acoje block, respectively. The legends has been given in the Fig. 5a.

According to the JP paper of Kemenetsky, the two regression lines were obtained based on the Al contents of chromite and their parental magmas, regardless if the chromite grains were from certain geological backgrounds. In the original diagram (left below), we can see that the data of high-Al chromite from BAB and OIB settings can also be plotted on the regression line for those high-Al chromite from MORB. Similarly, the data of low-Al (high-Cr) chromite from the LIP and BAB settings fall on the regression line for those high-Cr chromite from arc lavas. If we checked the paper of Rollinson, we can find that he plotted two data of BABB background into his diagram (right below). One is for high-Al chromite, the other for high-Cr chromite. Just as we mentioned above, the two data fall onto the regression lines for MORB and arc-related settings, respectively. *As a result, the Cr# of chromite seems to be the criteria for us to choose which regression formula to use for the calculation.* Of course, using the formulas for arc-related setting would generate even lower TiO<sub>2</sub> contents for the high-Al chromitites. Consequently, *using either formulas will not change our conclusion that the parental magmas of high-Al chromitites have lower TiO<sub>2</sub> contents than those typical MORB-like magmas.*

[Redacted]

Many thanks again for the suggestion. Moreover, we also found some problem with the Fig.5B (Fig. 4B in the new version) and have revised the diagram during the revision. Please have a check there.

**Reviewer #2 (Remarks to the Author):**

*The manuscript titled "Chromite Mineralization Variabilities Arising from*

*Chemo-Thermal Evolution in Mantle During Subduction Initiation" by Zhang and coauthors explores the elemental and Os isotopic compositions of different chromitites within the Zambales ophiolite and compares them with other ophiolitic chromitites worldwide, particularly with ophiolites that host both high-Cr and high-Al chromitites. This paper investigates the processes involved in the formation of chromitites during subduction initiation, contributing to an ongoing scientific debate. However, notable concerns warrant attention before considering this manuscript for publication in Nature Communications.*

**Our reply:** Many thanks for reviewing the paper and the constructive comments and suggestions. After reading your words below, we realize that some points of our work need to be further clarified. We have considered them carefully in the revised version, Please also check our answers below.\*\*\*\*\*To ensure that we can explain everything clearly, we divided several of your questions into some minor ones and replied with many details, so it will be time-consuming to read all of the replies. We are sorry for possibly taking you much time during the next-round review. Really hope you would be inclusive and considerate on this issue.\*\*\*\*\*Many thanks again for your time and efforts to our paper, and hope you would be fine with our replies.

**(HOPE YOU WILL NOT IGNORE THE COLORED BLUE LINES ABOVE)**

**Q2.1:** *Firstly, there is a substantial contradiction regarding the processes occurring during subduction initiation. The paper proposes that radiogenic Os components in mantle sources of high-Al chromitites may originate from sinking slabs during subduction initiation, challenging the traditional view of continuously intensified slab dehydration during this process. However, existing knowledge suggests minimal fluid contribution from the subducting plate during the formation of high-Al chromitites, such as those akin to fore-arc basalts (FAB), as documented by Ishizuka et al. (2014) and Whattam and Stern (2011). The authors must address this inconsistency in light of the well-established magmatic evolution associated with subduction initiation, including FAB, boninites, and Island Arc Tholeiites (IAT).*

*Ishizuka, O., Tani, K., Reagan, M.K. (2014). Izu-Bonin-Mariana forearc crust as a modern ophiolite analogue. Elements 10, 115-120. DOI: 10.2113/gselements.10.2.115. Whattam, S.A., Stern, R.J., 2011. The 'subduction initiation rule': a key for linking ophiolites, intra-oceanic forearcs, and subduction initiation. Contributions to Mineralogy and Petrology 162, 1031-1045. DOI 10.1007/s00410-011-0638-z.*

**Our reply:** Many thanks for the question. I like the works of Ishizuka et al. and Whattam & Stern very much and quite agree with their ideas on the genesis of FAB. However, we realize that we didn't clarify some points.

First, we think that the parental magmas of high-Al chromitites are not normal FAB but akin to those transitional FAB-boninitic magmas, the mantle sources of which indeed had slab-derived fluids involved based on isotopic studies as well as our other postulations.

Secondly, although we mentioned that our study challenges the traditional view of continuously intensified slab dehydration during this process (we found such narration may be inappropriate and revised), we did not deny the intensity of slab dehydration became stronger from the FAB magmatism to the transitional



FAB-boninitic ones. Since high-Al chromitites (products of transitional magmas) overall have higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than high-Cr chromitites (products of high-Ca boninitic magmas), what we wanted to say was that **the contribution of slab components** in the proto-forearc mantle decreased from the transitional magmatism to high-Ca boninitic magmatism (we only emphasized such a short period rather than the whole course of subduction initiation). Moreover, we realized the statement “challenging the traditional view of continuously intensified slab dehydration during this process” may not be appropriate, so we have deleted relevant sentences from the main text in the revised version. We also explained the meaning of the phrase “slab impact” [ $O_{\text{slab}}/(O_{\text{slab}}+O_{\text{asthenosphere}})$  rather than  $O_{\text{slab}}$  itself] in the revised version of our manuscript (L293, pls also see our reply to the Q2.5), because both slab dehydration and upwelling of asthenosphere would inject new components into the lithospheric mantle below proto-forearcs. As a result, it is better to **emphasize the relative contribution of slab rather than the absolute intensity of slab dehydration**. We also explained such information again in the reply to the Q2.5, which read similar to this question.

Sorry for the mis-understanding caused. In order to better clarify why high-Al chromitites were not products of normal FAB magmatism, we divided this question into four minor ones. Please see our explanations below.

**Q2.1.1 What compositions do the parental magma of high-Al chromitites have? (unlikely normal FAB, but are more depleted ones. Another points for your reference; MORB-like lavas are more widely distributed than boninitic ones. If MORB-like melts are the parental magmas of high-Al chromitites, there would be abundant high-Al chromite deposits than high-Cr ones. Actually, but the real case is just the opposite)**

Chromite grains in high-Al chromitites worldwide overall have distinctly lower  $\text{TiO}_2$  contents than those hosted in all kinds of MORB-like lavas, including BABB and FAB (MORB-like tholeiitic basalt). You can clearly see the compositional ranges of chromite from typical MORB, BABB and FAB in the new Fig. 4A of our manuscript (all featured by higher  $\text{TiO}_2$ ; modified from Pearce et al., 2000). The results of such a comparison suggest that the parental magmas of our high-Al chromitites could not be normal FAB but should have more depleted compositions. Moreover, our calculations show the parental magmas of high-Al chromitites from worldwide had  $< 0.7$  wt.%  $\text{TiO}_2$  contents, overall lower than the reported  $\text{TiO}_2$  contents of typical FAB (Reagan et al., 2010; Shervais et al., 2019). These comparisons clearly tell that **the  $\text{TiO}_2$  contents of normal FAB are still higher than the anticipated  $\text{TiO}_2$  contents of the parental magmas of high-Al chromitites (generally  $> 0.8$  wt.% vs.  $< 0.7$  wt.%).**

[Redacted]

**Fig. 2.1.1** diagrams showing the TiO<sub>2</sub> range of different lavas from Morishita et al. (2011; *Geology*) and Shervais et al. (2019), respectively. The blue areas in the diagrams mark the calculated TiO<sub>2</sub> range for the parental magma of high-Al chromitites, overall lower than those of normal MORB and FAB but overlapping those of transitional FAB and depleted FAB (D-FAB).

According to the past abundant studies, the compositions of magmas generated during subduction initiation varying from FAB to boninitic ones. Besides the two types of magmas, there are also some magmas overall with tholeiitic but transitional compositions, e.g., with intermediate TiO<sub>2</sub> and HREE contents between those of FAB and boninites. These transitional magmas have a certain compositional span, and we found that their erupted counterparts were called as transitional lavas, transitional FAB, depleted FAB, depleted tholeiitic lavas and others in different papers, with different genetic explanations (Reagan et al., 2010; Morishita, et al., 2011; Shervais et al., 2019), but they have identifiably different compositions to normal FAB.

Given the required lower TiO<sub>2</sub> contents of the parental magmas of high-Al chromitites than normal FAB, it is more reasonable to claim that the parental magmas have transitional FAB-boninitic compositions, and in our paper, we used the term “low-Ti tholeiitic” to describe their compositions based on their overall tholeiitic but more depleted features than FAB (MORB-like tholeiitic), which would make the compositions more easily to be imagined. Based on the TiO<sub>2</sub> contents of chromite in high-Al chromitites, the parental magmas should be of different depletion degrees, so Ti-rich and Ti-poor high-Al chromitites could be generated. Even so, from our own understanding, it is not easy to define a contrasting border for FAB and transitional FAB-boninitic magmas, so it is quite understandable why the studies of Morishita et al. and Shervais et al. used the names “transitional FAB” and “depleted FAB”. Anyway, these papers are very inspiring, and we like their work very much.

### **Q2.1.2 Why did previous studies not attribute formation of high-Al chromitites to FAB magmatism? (*insufficient comparisons and research gaps*)**

We think there are at least three reasons, and please see them below one by one.

First, previous studies realized that the parental magmas of high-Al chromitites must have lower TiO<sub>2</sub> contents than normal MORB, and incidentally, FAB overall have lower TiO<sub>2</sub> contents than MORB. However, they did not realize normal FAB still have higher TiO<sub>2</sub> contents than the parental magmas of high-Al chromitites, so the two items were simply linked together, but without further considering that how low TiO<sub>2</sub> contents the parental magmas of high-Al chromitites exactly had (e.g., in Morishita et al. 2011, *Geology*; pls see the marked lines below). Especially for those Ti-poor chromitites, their parental magmas (TiO<sub>2</sub> down to 0.2 wt.%) could not be originated from fertile MORB mantle directly.

[Redacted]

**Fig. 2.1.2** All the three photos were cut from Morishita et al. (2011, Geology). The blue area in the data diagram marks the calculated TiO<sub>2</sub> range for the parental magmas of high-Al chromitites.

Secondly, transitional magmas generated during subduction initiation were also classified as FAB in many papers, just as what had done in *Morishita et al. (2011)* and *Shervais et al. (2019)*, in which they were called transitional FAB and depleted FAB, respectively. However, if you check their compositions, you will find it better to call them transitional ones, with intermediate TiO<sub>2</sub> and HREE contents (pls see the data in Fig. 2.1.1 and Fig. 2.1.3). Specifically, *Morishita et al. (2011)* was (probably) the first paper mentioning the linkage between high-Al chromitites and FAB, but we believe that Prof. Morishita must also have realized the transitional compositional features for the parental magmas of high-Al chromitites, based on at least two places of narrations in his paper (*Pls see the Fig. 2.1.2 above; 2011, Geology*). One is that “some FABs show a transition in chemistry from FAB to boninites”. The other is that “medium-Cr# spinel-bearing dunite was caused by interaction with a melt transitional between MORB-like and boninitic melts, probably due to an increasing contribution of slab-derived fluids in an island arc setting”, the idea of which is generally consistent with ours for high-Al chromitites (pls note medium-Cr# spinel is used synonymously to high-Al chromite in literature for us working on chromite deposits).

[Redacted]

**Fig. 2.1.3** Comparisons of REE patterns between E-FAB, N-FAB, D-FAB, transitional lavas and boninitic lavas. It can be seen that D-FAB has intermediate HREE compositions between N-FAB and boninitic ones, just like how transitional lavas show in the right diagram (Pls see the circle).

Although Ti-poor chromite nodules or slivers had also been found in dunites below slow-spreading MOR settings, e.g., SWIR, [these Ti-poor chromite grains were explained to be finally equilibrated with ultra-depleted melts rather than MORB-like melts](#) (Morishta et al., 2007; pls see the marked area below in Fig. 2.1.4). As a result, we believe that the problem regarding the parental magmas of high-Al chromitites (should be more depleted than FAB) would gradually be clarified with more studies in the future.

[Redacted]

**Fig. 2.1.4** Ti-poor spinel found in the spinel micropods from the Southwest Indian Ocean Ridge.

Again, we would like to say that it is not easy to define a clear border for the FAB and transitional magmas, so it is quite understandable why the early studies of Morishta and Shervais used the names “transitional FAB” and “depleted FAB”. [If people still like to consider the parental magmas of high-Al chromitites as FAB, that is fine, but the “FAB” should be transitional FAB or depleted FAB with depleted tholeiitic compositions rather than normal FAB with MORB-like compositions.](#)

Thirdly, it has to be recognized that [researchers on forearc lavas of the IBM systems and those on ophiolitic chromitites are not the same group of people](#). We are often not 100% fully familiar with the work of each other, so [some gaps exist between the two groups](#) regarding issues of mantle magmatism and relevant processes. As a result, more exchange are highly needed to enhance our mutual understanding, and I would take opportunities to have more interactions with different groups in the future.

**Q2.1.3 Why were slab fluids so necessary in forming transitional FAB-boninitic magmas (helping to lower the solidus)? How to prove their presence? (Enriched Os-Sr-Nd-Pb-Hf isotopic ratios than FAB)**

FABs have been accepted to be generated via decompressional melting of fertile asthenospheric mantle below proto-forearc spreading centers, just as if how MORB were generated. [After extraction of FAB, the residual mantle sources will become harzburgitic in compositions.](#) Depending on melting degrees having happened in the mantle sources of FAB (10-20%, more or less), the residual mantle would have moderately or highly refractory (equally used as depleted in many papers, but actually not the same) compositions, probably equivalent to Cpx-poor lherzolites and

Cpx-poor harzburgites, respectively, from the lithological speaking. What would happen to the proto-forearc mantle then? For one thing, **if the geothermal gradient do not increase, no further melting would happen** to the residual harzburgitic mantle sources anymore. For another, **harzburgitization in the proto-forearc mantle means the lithospheric mantle would be thickened** after the FAB formation (pls see in the Fig. 2.1.5 and explanations in the next paragraph below), possibly making up the proto-form of sub-arc mantle, if the mature arc could be finally established.

Asthenosphere is the main source of heat flux for the overlying lithosphere, and the geothermal gradient of lithospheric mantle depends on the position (depth) of the lithosphere-asthenosphere boundary (LAB). **The shallower the LAB is, the hotter the lithospheric mantle is** (Pls see the diagram of geothermal gradient below). From the FAB magmatism to transitional ones, the mantle sources of magmas changed from fertile asthenospheric mantle to refractory lithospheric ones (not thick). Accordingly, **the LAB would have fallen down to deeper levels during the transitional magmatism. As a result, the geothermal gradient during the transitional magmatism could not be higher than that during the FAB magmatism**, although it cannot be determined exactly how large the gradient was decreased.

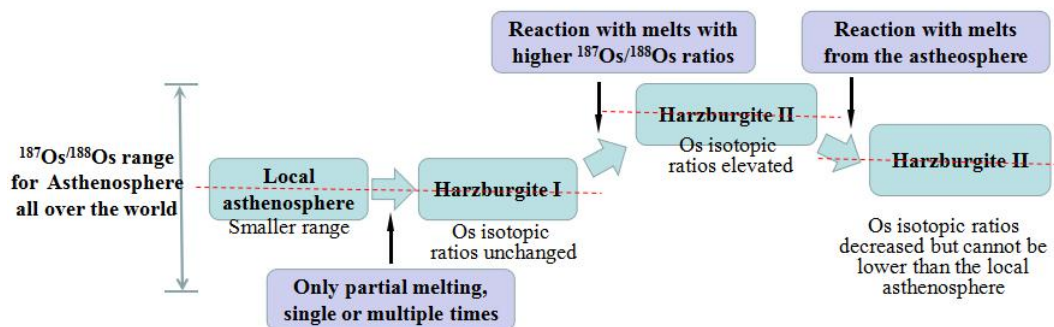
[Redacted]

**Fig. 2.1.5** Variation of geothermal gradients in lithospheric mantle with the LAB depth. The steep grey line defines the geotherm of the adiabatic asthenospheric mantle, while those gentle-sloped ones represent the geotherms in lithospheric mantle. The depths of the yellow dots mark the LAB. Overall, the shallower the LAB is, the hotter the lithospheric mantle is.

As we mentioned in the main text (L360-366 in the new version), **with the combined effects of harzburgitization and lowered geothermal gradient**, a second stage of partial melting became difficult in the proto-forearc mantle, and **addition of slab fluids would be highly necessary** in order to initiate further melting in the mantle. This explains why some high-Al chromitites have high  $^{187}\text{Os}/^{188}\text{Os}$  ratios.

Actually, **the mantle sources of all our chromitites, including both the high-Al and high-Cr ones, had contributions of slab fluids**, even though their  $^{187}\text{Os}/^{188}\text{Os}$  ratios fall into the range of global abyssal peridotites. Such a conclusion was obtained based on comparison with the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of harzburgites analyzed in our study. As we discussed in L290-300 (before revision), the origins of harzburgites in ophiolitic

mantle sequences can all be traced back to decompressional melting of asthenosphere, despite their melt modification histories. Bearing in mind that the process of partial melting alone cannot fractionate Os isotopes, harzburgites from the mantle sequence of some ophiolites should have indistinguishable Os isotopic ratios to the local asthenosphere, as long as only partial melting (single or multiple times) happened to the harzburgites. However, the fact is that harzburgites in the mantle sequences of ophiolites had been modified by parental magmas of different chromitites, all of which had higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the harzburgites, so **the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the proto-liths of the harzburgites should be even lower than those of the harzburgites themselves, not to mention to be compared with the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the parental magmas of our chromitites and their mantle sources.** Specific to the case of Zambales ophiolite, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of local asthenosphere (not the wider range compiled for global abyssal peridotites) below the Zambales proto-forearc could not be higher than those of our analyzed harzburgites, all below 0.1275 and smaller than the ratios of all the Zambales chromitites. Please see how the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the harzburgites changed during different processes in the flowchart below (note the relative positions of the dashed red lines).



Apart from the supra-chondritic Os isotopic features of high-Al chromitites, **transitional magmas/lavas also show more enriched Sr-Nd-Pb-Hf isotopic components than FAB**, overall comparable to those of high-Si boninites and arc lavas (Reagan et al., 2010, G3; pls see the diagrams in Fig. 2.1.5 below), supporting presence of slab fluids in the mantle sources of transitional magmas. Please see more details of the isotopic features of different lavas from the IBM forearc (Reagan et al., 2010, G3). In addition to all the postulations above, **the transitional period between anhydrous FAB and hydrous LSB (or called high-Ca boninites) must be affected by slab fluids.**

[Redacted]

*Fig. 2.1.6 Sr-Nd-Hf-Pb isotopic features of FAB (black dots and yellow squares outside the red circles), boninites (red diamonds), arc lavas (blue triangles) and transitional lavas (yellow squares in the red circles). All diagrams above were from Reagan et al. (2010, G3). Please note that the boninites shown in the diagrams above are high-Si boninites based on their compositions. This kind of boninites are thought to be formed in geodynamic setting more similar to arc lavas and considered as the start of real subduction. Please see the diagram below from Reagan et al. (2019, EPSL, we will not name it here).*

[Redacted]

**Q2.1.4 Why are transitional magmas more capable in forming chromitites than MORB-like FAB? (re-melting of residues left by FAB would release more Cr into magmas)**

Chromium is a compatible element in the mantle and are highly compatible in chromite (Cr-spinel) and clinopyroxene particularly. Generally, FABs and all other

magmas with MORB affinities were formed under moderate partial melting degrees, still with chromite and Cpx retained in their residual mantle. Consequently, FAB and other magmas with MORB affinities only have several hundreds of ppm Cr, overall lower than the saturation of Cr level about 1400 ppm. By contrast, **re-melting of a harzburgitic mantle left by MORB-like melt extraction would be more conducive for enrichment of Cr in the generated magmas, e.g., the transitional and boninitic magmas**, such that chromitites of economic significance could be more easily formed by such magmas. This is easy to be understood and can also be partly supported by the compositions of melt inclusions hosted in olivine from lavas. We have mentioned such information in the L340-347 (new revision).

For your better understanding, I would like to show you here a very persuasive phenomenon. As we all know, **anhydrous MORB-like magmatism are widespread in the world, in settings of MOR, BAB and infant subduction zones, but high-Al chromite deposits are very few**. Boninitic-like magmatism are spatially very restricted, but we have found much more high-Cr chromite deposits than high-Al ones. Even for the largest high-Al chromite deposit in the world, the Coto deposit we studied in this paper, its reserve is only comparable to a medium-sized high-Cr chromite deposit in the world. Anyway, **if people insist that high-Al chromite deposits were formed by typical MORB-like magmatism, the practical question needs to be answered first why there are much fewer high-Al chromite deposits than high-Cr ones in the world**. With more and more work carried in the future, I believe more and more people will accept that high-Al chromitites were products of low-Ti tholeiitic (transitional) magmatism.

**To be concluded**, the parental magmas of high-Al chromitites are for sure not typical MORB-like FAB but are more depleted low-Ti tholeiitic magmas with transitional FAB-boninitic compositions (*pls see at L143-146*), and their mantle sources were harzburgitic residue left by FAB extraction and hard to be further molten without additions of slab fluids (*pls see at L358-366*). Presence of slab fluids in their mantle sources can be supported by the high  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the high-Al chromitites in our study (*pls see at L219-232 and L290-300*), as well as enriched Sr-Nd-Pb-Hf isotopic features reported from previous studies. The higher concentrations of Cr in transitional magmas than FAB are better for the oversaturation of chromite from magmas (*pls see at L350-353*).

Although we have mentioned such information in different parts of the main text (pls see the places we noted above), we did further revisions in the new version in order to emphasize our new findings, including:

**L162-165:** Re-emphasized “the parental magmas of high-Al chromitites are not typical MORB-like magmas but are more depleted low-Ti tholeiitic ones”.

**L364 and 371:** Re-emphasized “typical MORB-like magmas do not have enough Cr for the oversaturation of chromite, but low-Ti tholeiitic ones have”. The exact reason can be found in the related two paragraphs”.

***Q2.2*** *Additionally, the interpretation that decreasing  $^{187}\text{Os}/^{188}\text{Os}$  ratios from high-Al chromitites to high-Cr chromitites can be explained solely by modification of asthenospheric melts requires further substantiation. The authors should provide more evidence and discuss other factors influencing the Os isotopic composition of chromitites, including mantle source heterogeneity, the degree of partial melting, extent of melt-rock interactions, and local sulfur fugacity, as suggested by numerous*



references (e.g., Hattori and Hart 1991; Burton et al. 1999; Brandon et al. 2000; Meibom et al. 2002; Walker et al. 2002; Büchl et al. 2004a, 2004b; Malitch 2004; and others listed in Gervilla et al. 2005).

Brandon, A.D., Snow, J.E., Walker, R.J., Morgan, J.W., Mock, T.D. (2000) 190Pt–186Os and 187Re–187Os systematics of abyssal peridotites. *Earth Planet Sci Lett* 177:319–335

Büchl, A., Brüggemann, G., Batanova, V.G. (2004a) Formation of podiform chromitite deposits: implications from PGE abundances and Os isotopic compositions of chromites from the Troodos complex, Cyprus. *Chem Geol* 208:217–232.

Büchl, A., Brüggemann, G., Batanova, V.G., Hofmann, A. (2004b) Os mobilization during melt percolation. The evolution of Os isotope heterogeneities in the mantle sequence of the Troodos ophiolite, Cyprus. *Geochim Cosmochim Acta* 68:3397–3404

Burton, K., Schiano, P., Birck, J.-L., Allegre, C.J. (1999) Osmium isotope disequilibrium between mantle minerals in a spinel-lherzolite. *Earth Planet Sci Lett* 172:311–322

Gervilla, F., Proenza, J.A., Frei, R., González-Jiménez, J.M., Garrido, C.J., Melgarejo, J. C., Meibom, A., Díaz-Martínez, R., Lavaut, W. (2005) Distribution of platinum-group elements and Os isotopes in chromite ores from Mayarí-Baracoa Ophiolitic Belt (eastern Cuba). *Contrib. to Mineral. Petrol.* 150, 589–607.

Hattori, K., Hart, S.R. (1991) Osmium-isotope ratios of platinum group minerals associated with ultramafic intrusions: Os-isotope evolution of the oceanic mantle. *Earth Planet Sci Lett* 107:499–514

Meibom, A., Sleep, N.H., Chamberlain, C.P., Coleman, R.G., Frei, R., Hren, M.T., Wooden, J.L. (2002) Re–Os isotopic evidence for long lived heterogeneity and equilibration processes in the Earth's upper mantle. *Nature* 419:705–708

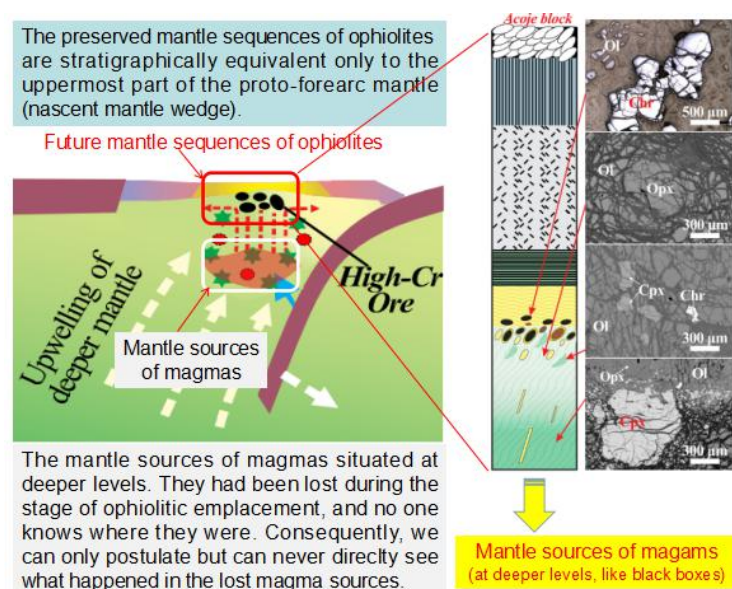
Walker, R.J., Prichard, H.M., Ishiwatari, A., Pimentel, M. (2002) The osmium isotopic composition of the convecting upper mantle deduced from ophiolite chromites. *Geochim Cosmochim Acta* 66: 329–345.

**Our reply:** Many thanks for the detailed and constructive comments. We quite agree with you that the Os isotopic compositions of mantle peridotites were controlled by the varied factors you pointed out above, which, we know very well, were originally mentioned in *Gervilla et al. (2005, CMP)*. Moreover, the recommended papers are also very helpful for understanding important processes regarding the origin of ophiolitic chromitites and their parental magmas.

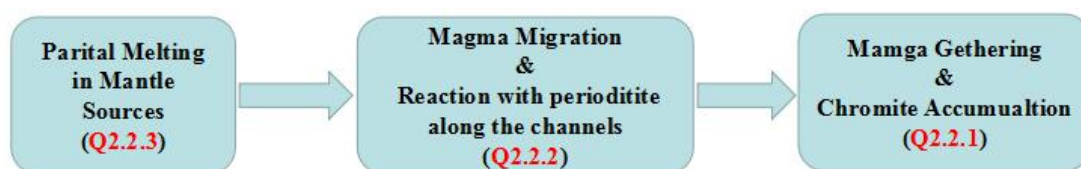
Generally, ophiolitic chromitites were generated in magma chambers or melt channels. In either case, the formation environments of chromitites were dominated by melts. As the Mg-rich parental magmas migrated through the upper mantle, they would react with mantle harzburgites, dissolving pyroxene in the harzburgites and converting the harzburgites into dunites. As a result, **Os in the parental magmas of our chromitites were basically sourced from two places. One was the mantle sources of the parental magmas; the other was the peridotites that the magmas passed by and reacted with.** Just as you reminded, the mantle sources of the parental magmas could be affected by a varieties of processes.

However, it is noted that the parental magma sources of all chromitites were stratigraphically situated deeper than the kept mantle sequences of the ophiolite (pls see the diagram below), and **they had detached from the current mantle sequences during emplacement** (they were possibly still in the mantle), so it is impossible to see what happened in the magma sources directly, and we cannot carry out our study in

the way how some of the recommended papers had done. Luckily, we can simplify the modification processes as binary mixing processes in black boxes, and the final  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the modified mantle regimes should fall in the intermediate range between their original ratios and those of the reactant melts, no matter how complex the modification processes were. Therefore, addition of low- $^{187}\text{Os}/^{188}\text{Os}$  components would be the only way of lowering the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the proto-forearc mantle. We have added such information into the revised version L241-250.



In order to answer this question more comprehensively, we further divided this question into five minor ones. Please see our explanations and diagram below. However, we would like to show first below what processes the parental magmas of chromitites had experienced before the accumulation of chromitites.



**Q2.2.1 Did Os isotopes fractionate between the crystallized chromitites and their parental magmas? (No; pls notice that the crystallization process of chromite can be considered as the moment of chromitites formation, and the parental magmas had experienced migration and accompanied melt-rock reaction, partial melting of their mantle sources, and other processes mentioned below)**

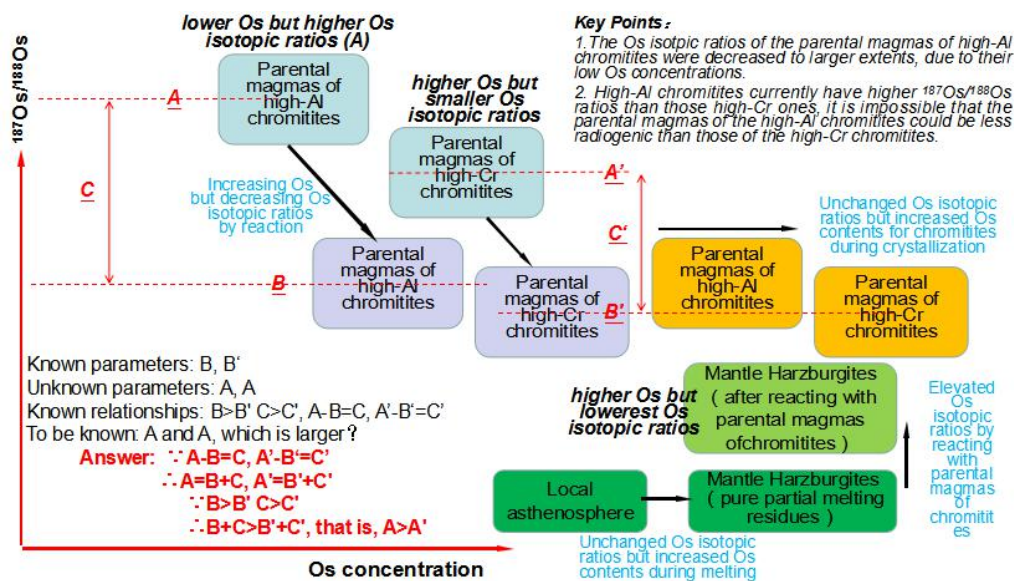
As we mentioned above, chromite and other minerals (e.g., olivine and PGM) in chromitites were crystallized from their parental magmas under melt-dominated conditions. Even though some grains may have been captured from surrounding rocks, their compositions must have been completely modified to be equilibrated with the magmas. As a result, **the compositions of chromitites were theoretically well buffered by their parental magmas.** Just like other traditional radiogenic isotopes (Sr-Nd-Pb), it is a common sense that **Os isotopes do not fractionate between the solid phases and their parental magmas.** Therefore, chromitites, as a whole, should have same Os isotopic compositions to their parental magmas at the moment of crystallization.

As was also commonly recognized, **chromitites always have very low Re/Os ratios**, and their  $^{187}\text{Os}/^{188}\text{Os}$  ratios cannot be largely changed after long-term radiogenic decay, not to mention that the age of the Zambales ophiolite is only 43-45 Ma. Accordingly, it is reasonable to claim that **the current  $^{187}\text{Os}/^{188}\text{Os}$  ratios of our chromitites should be overall same to their parental magmas**. Relevant discussions can be found in the revised paragraph L206-219.

**Q2.2.2 Did the Os isotopes of magmas change during their migration ? (Yes, but the general variation trend of  $^{187}\text{Os}/^{188}\text{Os}$  could not be changed; pls also notice that the migration process happened before the formation of chromitites)**

During the upward migration of the parental magmas of chromitites in the melt channels or when they were injected into magma chambers, the magmas often reacted with the pyroxene-bearing harzburgites (the dominant rocks in the mantle sequence). This is because the Mg-rich parental magmas of chromitites are under-saturated in Si and would dissolve pyroxenes in peridotites. Such reaction processes would not only convert the pyroxene-bearing peridotites to pyroxene-poor dunites but also modify the Os isotopic compositions of parental magmas of chromitites. Given that **chromitites overall have higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than harzburgites in the Zambales ophiolite**, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of their parental magmas can only be decreased after the reaction.

Osmium is a mildly compatible element in the mantle. Because parental magmas of high-Cr chromitites were generated at higher degrees of partial melting than those of high-Al chromitites, they theoretically have higher Os concentrations than those of high-Al chromitites. This is also a well recognized fact in our research field (pls see our newly added Os-Cr# diagram in the supplementary Fig. 2-B). From the perspective of reactant harzburgites, their Os concentrations and isotopic ratios overlap, no matter if they are Cpx-rich or not. Therefore, when reaction took place between the parental magmas of chromitites and harzburgites, **the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the parental magmas of high-Al chromitites, due to their lower Os concentrations, should have been modified (decreased) to larger extent than those of the parental magmas of high-Cr chromitites**. Given that high-Al chromitites currently have higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than those high-Cr ones, it is impossible that the parental magmas of the high-Al chromitites could be less radiogenic than those of the high-Cr chromitites.



We have illustrated how the Os isotopic compositions of the parental magmas of

different chromitites were modified in the the above diagram. Please see the details there, and hope you are fine with the explanation. [Relevant discussions can be found in the revised paragraph L220-233](#). Many thanks.

**[Q2.2.3 Isotopic changes during partial melting? \(No; the process happened before the magma migration and accompanied reactions along the way\)](#)**

It should be clarified first that the parental magma sources of all chromitites had been detached from the exposed mantle sequences during ophiolitic emplacement (they were probably still kept in the mantle). As a consequence, [it is impossible for us to recover what exactly happened in the parental magma sources \(what we cannot see\) in the way how we studied mantle xenolithos \(what we can see\)](#), because we do not have direct samples for carrying out the work. Instead, we have to take the geochemical compositions of the chromitites as important tools for relevant studies. [If Os isotopes cannot be fractionated during partial melting, the Os isotopic compositions of chromitites will be able to be used as the proxies for those of their parental magma sources and the magma sources.](#)

Although different mineral phases in cold samples may have different Os isotopic compositions sometimes, it is accepted that [such disequilibrium can be homogenized under conditions of high temperature, e.g., during certain degrees of partial melting and metasomatic melt-peridotite reaction, as were mentioned in your recommended papers \*Burton et al. \(1999, EPSL\)\* and \*Meibom et al. \(2002, Nature\)\*](#). This is because [such high-T conditions can effectively enhance the diffusivity and transportation of Os in minerals](#) and facilitate the homogenization of Os isotopic compositions in the rocks. This is particular true that, different from silicates, sulfides would be mostly liquefied during partial melting, so sulfides would quickly reach equilibration with other phases, melts or minerals, under high temperature conditions, especially during partial melting. Although disequilibrated features possibly still exist in some well enclosed Os-bearing grains (e.g., sulfide) in silicate, the amounts of such enclosed grains are overall very low (e.g., [Burton et al., 1999, EPSL](#)), and the whole-rock Os isotopic compositions of peridotites were mainly dominated those of interstitial sulfide and silicate themselves. Please see more details in the paragraphs below from [Burton et al. \(1999, EPSL\)](#)

[Redacted]

Up to now, we have not found in published papers that Os isotopes could be fractionated during partial melting. This can also be easily understood. To put it more directly, if Os isotopes can be fractionated during partial melting, all the previously established theories regarding Os isotopes could not stand anymore.

[Redacted]

**Q2.2.4 What kind of pre-melting processes could decrease the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of mantle? (The only practical way is to add exotic low- $^{187}\text{Os}/^{188}\text{Os}$  materials; pls note that *the processes mentioned below happened before the parental magmas of our chromitites were generated*, and the mantle regimes mentioned below will refer to the mantle sources of the parental magmas of our chromitites rather than at shallower levels where the parental magmas migrated upward. By contrast, *the processes shown in Q2.2.1-2.2.3 happened after the parental magmas were generated* and during their upward migration processes. So pls do not mix the two together)**

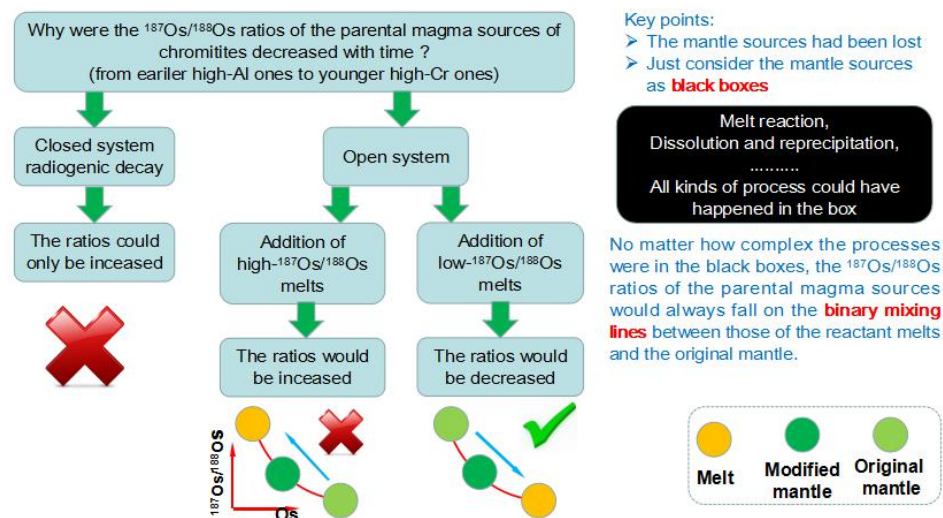
Before answering this question, we would like to reiterate that the parental magma sources of our chromitites had been lost before the final emplacement of the Zambales ophiolites. Therefore, we can only postulate what happened in the magma sources based on the elemental and isotopic compositions of the chromitites. Again, we quite agree that some of the processes you pointed could modify the Os isotopic compositions of the mantle sources, but unfortunately, the currently mantle sequences of the Zambales blocks do not include peridotites from such deep levels, so we have no direct sample but can only use other genetically relevant samples (e.g., chromitites in our study) to restore these processes. Even so, please see our explanations below how the factors of mantle source heterogeneity, degree of partial melting, extent of melt-rock interactions, and local sulfur fugacity affected the Os isotopic compositions of mantle rocks. We know well these factors were mentioned in Gervilla et al. (2005, CMP), but please note that the factor “local sulfur fugacity” mentioned in that paper was used to discuss the crystallization environments of PGM and sulfide rather than in the parental magma sources of chromitites.

Now, let's see the case of “mantle heterogeneity” first. For geochemists, “mantle heterogeneity” generally refers to spatial variation of chemical compositions in same

**temporal framework.** However, the topic we discussed is that high-Al chromitites were produced earlier than high-Cr chromitites in ophiolites, so **it is better to attribute the Os isotopic difference to “temporal variation” rather than “spatial heterogeneity”.** Beside the chromitites of the Zambales ophiolite, it is a common feature that high-Al chromitites overall show higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than their coexisting high-Cr chromitites in each ophiolitic block across the world, indicating that the difference was a prevalent feature rather than an occasional one.

As was answered to the Q2.2.3, **partial melting** cannot fractionate Os isotopes between magmas and their mantle sources. This is the precondition allowing to use the Os isotopic ratios of chromitites as proxies for those of their parental magma sources. Given the different Os isotopic ratios of high-Al and high-Cr chromitites, the precondition means that the parental magma sources of the two types of chromitites should often have inconsistent Os isotopic compositions, and that some certain processes must have happened in the proto-forearc mantle from the high-Al chromite mineralization to the high-Cr chromite mineralization.

So how to explain the decreasing  $^{87}\text{Os}/^{188}\text{Os}$  ratios? Why did it occur? Firstly, **the decreasing  $^{187}\text{Os}/^{188}\text{Os}$  ratio cannot be products of radiogenic decay of  $^{187}\text{Re}$  in the closed systems,** because radiogenic decay of parent bodies could only elevate the isotopic ratio of their daughter bodies, not to mention that the two types of chromitites were generated in a short duration. Thus, the possibility of close-system evolution can be ruled out, and **extra open-system processes are required in the mantle,** involving **input-output of melts and melt-rock reaction** under varying conditions (*again, pls note that the processes mentioned here happened in the deeper mantle level where the parental magmas of chromitites would be generated in the near future, and the melts mentioned here were not the parental magmas of our chromitites either, so the spatial and temporal frameworks used here are totally different from those mentioned in Q2.2.2-2.2.3*). However, **it is impossible to recover related details anymore because the mantle sources had been detached** from the current ophiolitic mantle sequences during emplacement. To postulate what happened to the lost parental magma sources of our chromitites under open-system conditions, **we have nothing to do but only to consider them as black boxes.**



Bearing such an assumption (black boxes) in mind, no matter how complex the melt-rock reaction processes were, **the compositions of the parental magma sources of**

our chromitites would always fall on the binary mixing line between those of the reactant melts and the original mantle regime prior to reaction in the diagrams of Os isotopes (Pls see the diagram above). Such binary-system mixing model is frequently used and easily understood. Thus, in order to draw supra-chondritic  $^{187}\text{Os}/^{188}\text{Os}$  ratios back to sub-chondritic levels, addition of melts with low sub-chondritic Os isotopic values was the only way. Please see the newly added lines L262-270.

Regarding the point of sulfur fugacity, we checked how such a parameter was mentioned in Gervilla et al. (2005, CMP), which is very classical and popular paper. Actually, contents regarding sulfur fugacity in the paper were all on the crystallization process of PGM in chromitites, without anything on the mantle sources. The original sentence in the paper is that “Cr#, PGE abundances, and bulk Os isotopic composition of chromitites are determined by the combined effects of mantle source heterogeneity, the degree of partial melting, the extent of melt-rock interactions, and the local sulfur fugacity”. However, local sulfur fugacity mainly control the species of PGM and their crystallization from magmas, so as to affect the types of PGM phases in chromitites and whole-rock PGE concentrations of chromitites. As we mentioned early to this question, crystallization cannot fractionate the Os isotopic compositions between minerals and magmas, so variation of sulfur fugacity can only control which types of PGM formed from the parental magmas of chromitites but could not change the Os isotopic compositions of chromitites at all. Although melt-rock reaction may change the sulfur fugacity in the parental magma sources of chromitites (this was not mentioned in Gervilla’s paper), the final Os isotopic compositions would definitely fall on the mixing lines between those of the original mantle peridotites and reactant melts based on the binary mixing theory, no matter how complicated the reaction processes were (remember the black-box model). As a result, it seems that the role of sulfur fugacity matters in controlling the Os isotopic compositions of our chromitites, although it may have affected their PGE concentrations.

[Redacted]

**Q2.2.5 Where were the required low- $^{187}\text{Os}/^{188}\text{Os}$  melts from? (Asthenosphere). Any geological evidence? (Yes, presence of Cpx-harzburgites, refertilized products)**

As we mentioned in the main text (L219-220, new revision), asthenospheric

upwelling and slab dehydration were the two dominant processes that could modify the elemental and isotopic features of proto-forearc mantle during subduction initiation. On one hand, slab-derived components are featured by highly radiogenic Os isotopic features, so addition of slab materials can only help to maintain the high Os isotopic ratios of the mantle sources rather than decreasing the ratios. Therefore, this possibility can be ruled out. By contrast, [asthenosphere always has lower Os isotopic ratios than slab components, so modification by asthenospheric melts could be the potential way of lowering the  \$^{187}\text{Os}/^{188}\text{Os}\$  ratios of slab-modified mantle](#). That is, there were more contributions of asthenospheric melts in the proto-forearc mantle at the formation stage of high-Cr chromitites than that of the high-Al chromitites.

How to prove the postulation (increasing contribution of asthenosphere) above from other perspectives? Apart from inducing abyssal-like Os isotopic features, [addition of asthenospheric melts would also cause refertilization in the lithospheric mantle via replenishing fusible elements \(e.g., Ca and Ti\)](#).

On one hand, there are Cpx-rich harzburgites (including Cpx-poor lherzolites) in the Acoje mantle sequence. These Cpx-rich harzburgites make up the host rocks of chromitites and dunites in the mantle sequence, so they were formed earlier than the chromitites (please see more details about their genesis in our answers to Q2.3). Specially, [clinopyroxene grains in the Cpx-rich harzburgites mostly have finger-like protrusions](#) (look like octopus with contractile arms) that stretched into the interstitial spaces. Such textures support the outward growth history of the clinopyroxene with aid of interstitial melts. Moreover, [these clinopyroxene, especially those in the Cpx-rich harzburgites from bottom part of the mantle sequence have comparable compositions \(Ca, Al, Ti and REE\) to those in the abyssal peridotites](#), indicating the asthenospheric affinities of the refertilizing melts (pls see more details from the two classical cases below Fig. 2.2.1).



[Redacted]

**Fig. 2.2.1** Textural and compositional features of Cpx grains from Cpx-rich harzburgites, which is called as Cpx-bearing harzburgites for the Luobusa ophiolite. The two BSE images show the irregular outlines of Cpx and their alignment in the rocks, supporting their out-ward growth histories from interstitial melts. The four data diagrams below show the compositions of Cpx from Cpx-rich harzburgites (S-1,S-2,S-3,S-5, LBS-05, LH-08, LH-05). It can be seen that Cpx from Cpx-rich harzburgites have compositions generally comparable to the abyssal peridotites, especially those in the Cpx-rich harzburgites (some are lherzolites) from the bottom parts of the mantle sequence (e.g., S-1, S-2, LH-08, LH-05). In case that they were notably modified, their compositions would move to more depleted directions (lower Al and HREE).

On the other, the Ti contents of chromite in high-Cr chromitites are overall no lower than those of chromite in the high-Al chromitites, reflecting that the parental magma sources of the high-Cr chromitites were overall less depleted than those of the high-Al chromitites (to be exactly, comparable to those of Ti-rich high-Al chromitite, but more fertile than those of Ti-poor high-Al chromitites). Moreover, [addition of asthenospheric melts into the overlying lithospheric mantle means active asthenospheric upwelling, which will bring about elevated geothermal gradients](#) (Please see more details in our answers to Q2.4). This would facilitate higher degrees of melting and generate boninitic-like magmas. Please see two aspects in the revised version L400-405.

**Combined all the information and postulations above together**, it is clear that the required low-<sup>187</sup>Os/<sup>188</sup>Os components must be originated from asthenospheric mantle. However, relevant details cannot be retrieved now, because it is impossible to get direct samples from the parental magma sources of chromitites, which had been lost during ophiolitic emplacement. What we can do at this stage is only to consider the processes in the mantle sources as black boxes. Whatever processes happened to the mantle sources, their final Os isotopic compositions would definitely fall on the mixing lines between those of the original mantle peridotites and reactant melts based on the binary mixing model. [Again, we emphasized relevant contents in the revised manuscript L261-270.](#) Hope you are fine with our explanations above. Many thanks.

[Redacted]

**Q2.3** The speculation surrounding the presence of three types of peridotites (Cpx-poor Hz1, Cpx-rich Hz2, and Cpx-poor Hz3) based on limited evidence needs further substantiation. The authors should provide additional supporting data for the presence of these peridotite types and the transformation from Hz1 to Hz2.

**Our reply:** Many thanks for the question. Actually, we have solid evidence to support the multi-stage transformation of peridotites in the mantle sequences, and such transformation history has been well recognized in previous studies, including our owns (e.g., Zhang et al., 2016, 2019, 2020a, 2020b).

Overall, our postulations were mainly obtained based on petrographic textures and the mutual spatial relationships among different types of peridotites, **which, we want to emphasize here, are the absolute preconditions for interpreting the geneses of mantle rocks.** This is because peridotites that experienced different processes but have similar mineral proportions can be featured by similar chemical compositions, from the perspectives of bulk rocks and each mineral phase. Take a

piece of Cpx-poor lherzolite for example. Based on chemical compositions alone, it is usually (we are not saying always here) difficult to know if it was left after low degrees of partial melting of the FMM, or if it was refertilized by MORB-like melts from harzburgites. However, if it has refertilized genesis, there must be special petrographic features that are absent in the pure partial melting residues, e.g., protrusions of Cpx mentioned in our study showing the outward growth history of the grains from interstitial melts. As a consequence, if we simply treated all peridotites in the ophiolitic mantle sequences as partial melting residues, that would be quite wrong. The discussions in some papers can support my statement above (Pls see the classical one on the lherz massif).

The Lherz spinel lherzolite: refertilized rather than pristine mantle  
V Le Roux, JL Bodinier, A Tommasi, O Alard... - Earth and Planetary ..., 2007 - Elsevier

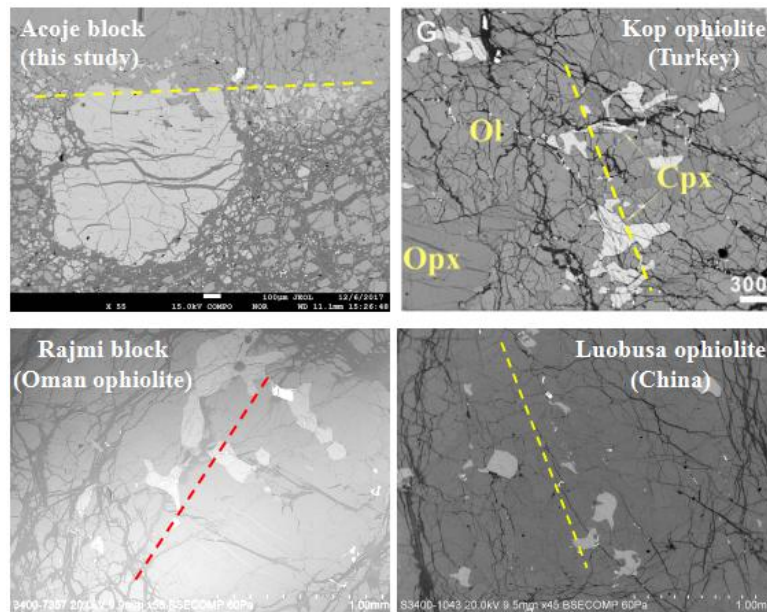
[Redacted]

Anyway, I am confident of my conclusions about the multi-stage transformation of peridotites, which were obtained based on unequivocal petrographic features and spatial relationships among different lithologies. To better answer this question, we divide it into two minor questions. Please see our explanations below.

### **Q2.3.1 Any evidence to support the transformation from the Cpx-poor Hz1 to Cpx-rich Hz2 (Prevalent protrusions of Cpx and presence Cpx veins)?**

The textures of minerals are important features for restoring the formation processes of rocks. In the Cpx-rich harzburgites, clinopyroxene grains are mostly featured by finger-like protrusions (like octopus with contractile arms) that stretched into the interstitial spaces. Sometimes, the anhedral clinopyroxene grains even make up clinopyroxenite veins. Such textural features support the outward growth history of the clinopyroxene with the aid of infiltrating melts. Generally, clinopyroxene grains in the Cpx-rich harzburgites have comparable major element compositions and REE compositions to those grains in the abyssal peridotites, indicating the asthenospheric affinities of the refertilizing melts. Since the Cpx-rich harzburgites were refertilized products rather than pure partial melting residues, there must be reactant proto-lithos for the newly-formed Cpx-rich harzburgites, and we think it is better to call them as Cpx-poor harzburgites, given that they had fewer Cpx grains. **Anyway, if something itself was a modified product, there must be a proto-type for it.**

To better convince you the refertilization origin of the Cpx-rich Hz2, we found more photos from ophiolites world. Please see them in the Fig. 2.3.1 below.



**Fig. 2.3.1** BSE photos of Cpx-rich harzburgites (even Cpx-poor lherzolites) from different classical ophiolites in the world. The second brightest mineral grains are clinopyroxene. They can be seen to have figure-like protrusions and make up lineation, indicating melt infiltration in the rocks along the directions of dashed lines.

**Q2.3.2 Any evidence to support the transformation from the Cpx-rich Hz2 to Cpx-poor Hz3?** (Yes, it is common sense for researchers on podiform chromitites and ophiolitic peridotites. Note that the cpx-poor Hz3 we mentioned here can be seen in the field, but the ever existence of the cpx-poor Hz 1 was postulated based on the textures of cpx-rich harzburgites. Pls refer to the Q2.3.1 for the answer.)

Actually, it has become a common sense for researchers on ophiolites and podiform chromitites that Cpx-poor harzburgites in ophiolitic mantle sequences were modified products of Cpx-rich harzburgites by Mg-rich and SiO<sub>2</sub>-undersaturated melts, which were able to dissolve pyroxene in the harzburgites and converted them into Cpx-poor harzburgites, dunites and even chromitites under conditions of increasing melt/rock ratios. The Cpx-poor harzburgites that we see nowadays in ophiolites were all formed via the way above (remember that Cpx-poor Hz1 had been replaced by Cpx-rich Hz2). Although it is often not easy to take all these different lithologies in small photos, we did have some wonderful composite samples from other ophiolites (e.g., the Luobusa ophiolite, Tibet; the Fig.2.3.2 below) to support our idea, and the compositions of mineral phases (especially chromite) show systematic variation from the Cpx-rich harzburgites to the Cpx-poor harzburgites, dunites and chromitites (Zhou et al., 1996, JP).

[Redacted]

A chromitite-dunite vein hosted in Cpx-rich harzburgite body from the Tibetan Luobusa ophiolite. The coarse grains on the surface are pyroxenes. Some of them are Cpx grains but cannot be easily recognized on outcrops and hand-species. Generally, peridotites with abundant pyroxene are probably Cpx-rich harzburgites, and this is our empirical approach to tell which are Cpx-rich harzburgites and which are Cpx-poor harzburgites in the field. Just an important reminder here, peridotites in ophiolites look quite different from xenoliths entrapped by lavas.

[Redacted]

**Fig. 2.3.2** A composite sample of Cpx-rich harzburgite, Cpx-poor harzburgite (only a narrow slice, the transitional zone between the LBS-04 and LBS-05), dunite and chromitite from the Tibetan Luobusa ophiolite. It records a reaction history between the Cpx-rich harzburgites and the parental magma of the chromitites. In the data diagram, the data of clinopyroxene grains from the Cpx-rich harzburgites were plotted into the green dashed line, overlapping the range of clinopyroxene in the abyssal peridotites, while grains from the dunite and chromitite parts show lower  $Al_2O_3$  contents. The diagrams below show the systematic compositional variations of chromite from the chromitite zone (left) to the Cpx-rich harzburgite zone (right) for your reference.

[Redacted]

Zhou, M.F., Robinson, P.T., Malpas, J., & Li, Z. (1996). Podiform chromitites in the Luobusa ophiolite (southern Tibet): implications for melt-rock interaction and chromite segregation in the upper mantle. *Journal of Petrology*, 37(1), 3-21.

Although we didn't collect such composite samples from the mantle sequence of the Acoje block, we have evidence to support the transformation history from Cpx-rich harzburgites to Cpx-poor harzburgite in the mantle sequence. Like what we observed in the classical composition sample from the Luobusa ophiolite, [We would like to handle this problem from two perspectives: \(1\) do the two types of harzburgites exist together; \(2\) are there any compositional evidence?](#)

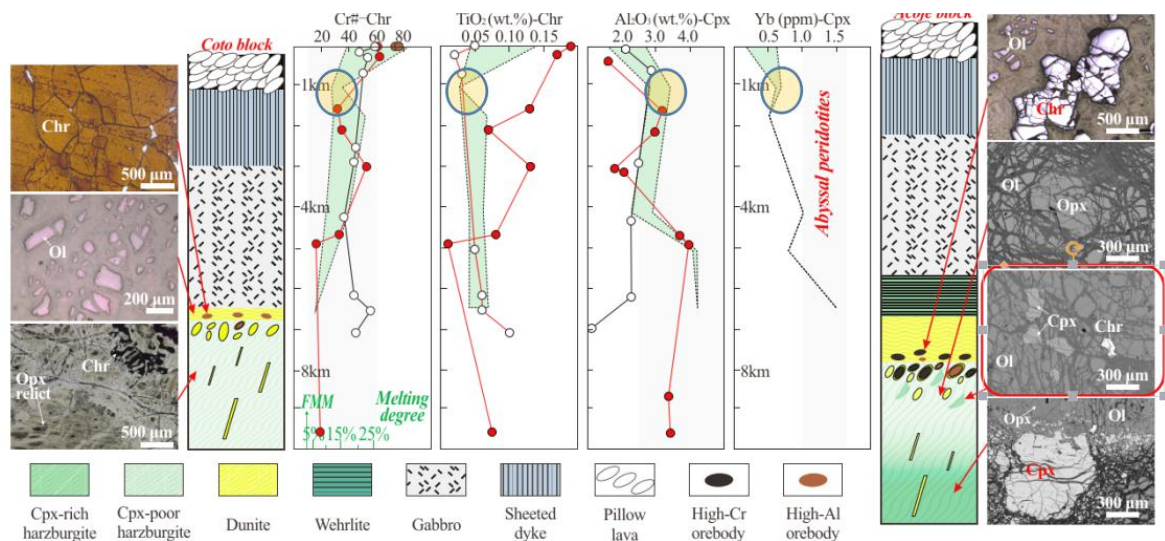
First, let's see if the two types of harzburgites co-exist. [In the mantle sequence of the Acoje block, Cpx-rich harzburgites are distributed mainly at the bottom parts, but they were also found sometimes to be spatially mixed with Cpx-poor harzburgites in the uppermost sequences.](#) For example, the rocks found at the Stop 4 shown in the geological map (Fig. 2C) were Cpx-rich harzburgites (pls see their photomicrographs in the Fig. 3 of our manuscript or the marked one in the Fig. 2.3.3 below), but peridotites surrounding them are overall Cpx-poor harzburgites according to our field and petrographic observations (more details can be seen in Zhang et al., 2021, Lithos). Even at the mantle-MTZ boundary (e.g., the stop 5), both Cpx-rich harzburgites and Cpx-poor harzburgites can be found to be mixed together (Zhang et al., 2021, Lithos). [These observations strongly support the wide distributions of Cpx-rich harzburgites in the mantle sequence of the Acoje block, and we mentioned such observations in the Section "Geological Background" \(peridotites of the Acoje block vary from Cpx-poor harzburgites with enclosed dunite and Cpx-rich harzburgite \(3-5 modal% Cpx\) lens at the topmost mantle sequence\).](#) According to our experiences, such a feature has also been found in other classical ophiolites that host high-Cr chromite deposit, e.g., the Luobusa and Kop ophiolite.

Zhang, P.F., Zhou, M.F., Yumul Jr, G.P., & Wang, C.Y. (2021). Geodynamic setting of high-Cr chromite mineralization in nascent subduction zones: Li isotopic and REE constraints from the Zambales ophiolite, Philippines. *Lithos*, 384, 105975.

To further convince you that cpx-poor harzburgites and cpx-rich harzburgites are mixed in the uppermost part of the Acoje mantle sequence, [we would also like to show you some \*\*compositional evidence\*\*.](#) In the Fig. 3 of our manuscript, it can be

seen that **clinopyroxene grains** overall become more and more depleted in  $\text{Al}_2\text{O}_3$  and Yb from bottom cpx-rich harzburgites upward to those cpx-poor harzburgites and dunites in the mantle sequence of the Acoje block (pls see it below). Generally, **two reasons account for such variations**. First, reaction with Mg-rich melts, playing the same role as partial melting, would not only make peridotites more refractory (converting them to cpx-poor harzburgites and even dunites) but also extract fusible and incompatible elements from the peridotites, and the extents of modifications depend on melt/rock ratios. Secondly, migration of melt below spreading centers usually changed from pervasive to channelized flows and finally merged into the magma chambers of the MTZ, just like the roots of trees changing from the capillary root system to the trunk), so the melt/rock ratio in the mantle sequences of ophiolites generally increased from bottom to the MTZ. As a result, peridotites in the mantle sequences of ophiolites generally become more and more refractory from bottom upward and finally vary into dunites in the MTZ. The explanations above are common senses for us researchers on ophiolitic chromite deposits.

Despite such general variations, **it can be found that clinopyroxene grains in harzburgites at the stop 4 show less depleted compositions** (e.g., higher  $\text{Al}_2\text{O}_3$  and Yb contents) than those in the harzburgites from both deeper and shallower levels. Given that the clinopyroxene grains in the cpx-rich harzburgites overall have less depleted compositions than those in the cpx-poor harzburgites, the variation shown at the stop 4 should be caused by **presence of cpx-rich harzburgites in the large-scale background of cpx-poor harzburgites**. Moreover, even at the mantle-MTZ boundary (0 km, e.g., stop 5), clinopyroxene grains show large variation, e.g., their  $\text{Al}_2\text{O}_3$  contents varying from 1.5 to 2.7 wt.%. Based on our records, the highest  $\text{Al}_2\text{O}_3$  contents were obtained for clinopyroxenes found in the cpx-rich harzburgites. However, it is noted that these cpx-rich harzburgites in the uppermost part of the mantle sequence were also modified by Mg-rich harzburgites, but the melt/rock ratios in these cpx-rich harzburgites were not as high as those in the cpx-poor harzburgites, such that more pyroxene (including both Cpx and Opx) were preserved in them. As a result, **both our field observation and compositional features support presence of cpx-rich harzburgites in the cpx-poor harzburgites**, and the two types of harzburgites do exist together in the uppermost mantle sequence of the Acoje block (that is why we drew some patches of cpx-rich harzburgites along with chromite pods in the uppermost mantle sequence of the Acoje block; pls see this feature in the right stratigraphic column).



**Fig. 2.3.3** Compositional anomalies of minerals in harzburgites collected from the Stop 4, as were marked by the shaded brown circles. It can be seen that clinopyroxene grains from the Stop 4 have more enriched compositions than those from shallower and deeper levels. The BSE image marked by red rectangle show the petrographic feature of Cpx-rich harzburgites from the Stop 4, which is stratigraphically located almost at the uppermost part of the mantle sequence.

Then, are there any compositional evidence from **the perspective of chromite** to support the transformation from the Cpx-rich HZ2 to the Cpx-poor HZ3. Overall, **chromite grains in the Cpx-poor harzburgites have higher Cr# and TiO<sub>2</sub> contents than those in the Cpx-rich harzburgites** (see the variation trends in Fig.2.3.3 above). Since Ti is an incompatible element, the increasing TiO<sub>2</sub> contents with Cr#s (taken as a proxy of partial melting degree) could not be caused by partial melting. Instead, such increasing trend can only be explained by using **melt-peridotite reaction, which would elevate both the Cr# and TiO<sub>2</sub> contents of chromite with increasing melt/peridotite ratios**. Especially, when the melt/peridotite ratio reached high enough, chromite in the peridotites would have similar Cr# and TiO<sub>2</sub> contents to those in chromitites or chromite-bearing dunites. This is a **common sense for us** researchers on podiform chromite deposits and peridotites of ophiolites. Combined with the spatial relationship between the Cpx-poor and Cpx-rich harzburgites and also the compositional variation trend between them, it is reasonable to claim that the **Cpx-poor harzburgites were converted from the Cpx-rich harzburgites via melt-rock reactions**, and the Cpx-rich harzburgites probably once occupied the whole mantle sequence of the ophiolite according to their wide distribution. More details about the effects of melt-rock reactions can be found in *Pearce et al. (2000)*. We also list his diagram and our diagram above (**Fig. 2.3.4**). Please check the details below.

[Redacted]

**Fig. 2.3.4** The effect of melt-rock reaction (e.g., red arrow in the left diagram) on the compositions of chromite in harzburgites. Due to the high concentrations of Ti in melts, reaction with melts would overall elevate the TiO<sub>2</sub> contents of chromite in the harzburgites, and the elevated extents depend on the melt/rock ratios.

[Redacted]

To better show different reaction histories that happened to peridotites in the mantle sequences of many ophiolites, we used the diagram Fig. 2.3.5 below to give a summary, from Cpx-poor harzburgite I (A) to Cpx-rich harzburgite II via refertilization (B) and finally to Cpx-poor harzburgites, dunites and chromitites via



melt-peridotite reactions (C and D) (from *Zhang et al., 2020, OGR*).

[Redacted]

**Fig. 2.3.5** Lithological transitions from the earliest cpx-poor harzburgite (I) to cpx-rich harzburgite (II) and finally to cpx-poor harzburgite (III), dunite and chromitite under conditions of increasing melt/rock ratios. The cpx-rich harzburgites (II) were refertilized products by asthenospheric melts, and clinopyroxene grains in them have finger-like protrusions and are featured by compositions (e.g., major element and REE) similar to those in the abyssal peridotites, as were shown in the Fig. 2.3.1, Fig. 2.3.2 and Fig. 2.2.1. **The cpx-poor harzburgite (I) had totally disappeared (replaced by the cpx-rich harzburgites II), and their presence was postulated based on our petrographic observation of cpx-rich harzburgites (II). What we can exactly see in ophiolitic mantle sequences nowadays are the cpx-poor harzburgites (III) rather than the cpx-poor harzburgites (I).** Please note that the outlines of cpx and opx should actually be irregular in real cases.

Actually, we showed the main evidence mentioned above in our manuscript, but in simplified versions. Please see them below.

**L89-100:** **Emphasized the coexistence of Cpx-rich and Cpx-poor harzburgites.** “Peridotites of the Acoje block vary from Cpx-poor harzburgites with enclosed dunite and Cpx-rich harzburgite (3-5 modal% Cpx) lens at the topmost mantle sequence to Cpx-rich harzburgites and Cpx-poor lherzolites (5-10 modal% Cpx; hereinafter grouped as Cpx-rich harzburgites) at the bottom mantle sequence” and “The Cpx-poor harzburgites and dunites in the mantle sequences of both blocks were modified from less depleted peridotites by Mg-rich melts (e.g., parental magmas of different chromitites) under increasing melt/rock ratio conditions (*Zhang et al., 2020b, 2021*)”

**L259-268:** **Use the petrographic features of Cpx to support the refertilized origin of the Cpx-rich harzburgites (changed from CP-HZ I; melt impregnation means the grains experienced growth history from interstitial melts).** “Clinopyroxene grains in the Cpx-rich harzburgites show signs of melt impregnation and even make up diopsidite veins (*Fig. 3*). Combined with the compositions of clinopyroxene and chromite in peridotites, the Cpx-rich harzburgites cannot be residues of low-moderate degrees of melting but are refertilized products of previous Cpx-poor harzburgites (CP-Hz I, not those in the current mantle sequence) by asthenospheric melts”

**L278-285:** **Use the observed spatial relationship and compositional variation between the Cpx-rich harzburgites and Cpx-poor harzburgites (pls note that they are not the CP-HZ I, which had been refertilized into Cpx-rich harzburgites already) to tell the**

transformation from the former to the latter. “In the uppermost Acoje mantle sequence, Cpx-rich harzburgite bodies occur as enclosed lens in the Cpx-poor harzburgites at places, and chromite shows positively correlated TiO<sub>2</sub> and Cr# from the Cpx-rich to Cpx-poor harzburgites, a trend consistent with the result of melt-peridotite reaction but contradictory to that of mantle melting. These features suggest that the Cpx-rich harzburgites, refertilized products of the early CP-Hz I, later reacted with Mg-rich magmas and formed new Cpx-poor harzburgites”.

**Q2.4** *The temperature variations during different types of chromite mineralization are inconsistent with previously published data, such as those related to Forearc Basalt (FAB) temperatures, as discussed by Whattam et al. (2020, AM). The authors should reconcile this inconsistency with existing data.*

**Our reply:** Many thanks for the comment and the mentioned paper of Whattam et al. (2020). As we told previously, Whattam’s works are really great, and we learned a lot from his papers in the past.

Before answering this question, we want to confirm at first if you were asking FAB should be generated under higher temperatures than boninitic-like ones according to Whattam et al. (2020). If yes, **such a problem was probably aroused because you think that high-Al chromitites were generated by FAB (this is how all your main concerns were caused)**, so that you wanted to compare the formation temperatures of FAB and boninitic magmatism. However, the problem is that **high-Al chromitites (at least the majority of them) were actually not generated by typical FAB magmatism but by more depleted ones** (pls see our explanations to the Q2.1). Therefore, it is not our aim to compare the formation temperatures of FAB and boninitic magmas directly, and it is not easy to make the comparisons for them either.

It has become a common sense that magmatism during subduction initiation generally varies from FAB to transitional type and then to boninitic with time. **To understand how the geothermal gradient in the proto-forearc mantle varies all through subduction initiation, we need conduct two sets of comparisons:** (1) between the parental magmas of high-Al chromitites (low-Ti tholeiitic, transitional FAB-boninitic) and the formation temperatures of the parental magmas of high-Cr chromitites (boninitic-like); and (2) between the formation temperatures of FAB and the formation temperatures of the parental magmas of high-Al chromitites, separately.

In order to better reply to this comment and clarify some important points, we divide it into several minor questions. Please see our explanations below.

**Q2.4.1 Were parental magmas of high-Al chromitites (depleted tholeiitic) formed under lower temperature conditions than those of high-Cr chromitites (boninitic-like)? (Yes, not only a common sense but also supported by experimental and geological work. Please see our explanation in the last two sections of the MS)**

The answer to this question can be simply reasoned with our general knowledge. This is because, compared with magmas of tholeiitic series, boninitic-like magmas are products of higher degrees of melting and have higher MgO contents, indicative of higher temperatures of melting in the mantle. **Although most studies like saying that the mantle sources of boninitic-like magmas are refractory and hydrous in compositions, having such mantle sources alone can only generate high-Mg magmas of arc affinities rather than exactly boninitic-like magmas if the geothermal gradients were not high enough.** To put it more understandable and straightforward, nobody would doubt that picritic and komatiitic magmas, both of which are featured by high

MgO contents, could be formed under same temperature conditions to average mafic ones. Particularly, we found a diagram below regarding the formation temperatures of different magmas from *Grove and Parman (2004)* for your reference.

[Redacted]

Now, let's see what happened in our work. As we discussed in the L312-316 (before revision), the primary compositions of mantle-derived magmas depend on the mantle source compositions and fluid contents in the sources. On one hand, our calculations show that the parental magma sources of Ti-rich high-Al chromitites and high-Cr chromitites share similar fertility. On the other, the overall higher Os isotopic ratios of high-Al chromitites mean more involvement of slab fluids in the mantle sources than in those of high-Cr chromitites. Under similar conditions of melting temperature and mantle source compositions, presence of more fluids in the mantle sources obviously means more Mg-rich magmas would be generated. If this is the real case, the parental magmas of high-Al chromitites would be more Mg-rich than those of the high-Cr chromitites. However, such a assumption is contradictory to the fact that the parental magmas of high-Cr chromitites had more Mg-rich boninitic-like compositions. That is to say, the parental magmas of high-Al chromitites must have been generated under lower temperature conditions than those of high-Cr chromitites, and there must be a heating period in the proto-forearc mantle from the mineralization stage of high-Al chromite to that of high-Cr chromite.

Do we have any evidence to support presence of a heating period in the proto-forearc mantle? Yes, we do have. Theoretically, the heating processes could only be caused via upwelling of asthenosphere, the process of which works just as if fire is lighted below a pot and is the main source of heat flux in the mantle (except there are mantle plumes rising up). Although we cannot see how the heating process happened, there must be some geochemical responses left. Upwelling of asthenosphere would have caused certain degrees of refertilization and replenishment of fusible elements (e.g., Ti, Al and Ca) in the overlying lithospheric mantle. From viewpoints of mantle petrologists, refertilization of lithospheric mantle below

spreading centers or rifting systems means upwelling of asthenosphere and enhancing contributions of fertile components in the lithospheric mantle. As a result, presence of refertilized peridotites by asthenospheric melts can be considered as an indirect evidence of heating process in the lithospheric mantle.

Specifically in the mantle sequence of [the Acoje block](#), [the Cpx-rich harzburgites and Cpx-poor lherzolites have been proven as refertilized products of asthenospheric melts](#), revealed by the outward growth textures and MORB-like affinities of the newly formed clinopyroxene (please see more details in our replies to the Q2.2 and Q2.3). Importantly, these refertilized peridotites were formed prior to the high-Cr chromite mineralization based the spatial relationships among different lithologies, suggesting that asthenospheric mantle had upwelled before the mineralization. Moreover, [taking the North China Craton as an extreme example \(we also mentioned this case in the discussion\)](#), this craton had thick lithospheric root up to 200 km deep in Paleozoic, but the lithospheric mantle of its east part underwent intensive refertilization during Meso-Cenozoic and large volumes of magma eruption due to the strong upwelling of asthenosphere, where a regional strike-slip system was developed. Consequently, the originally thick cold harzburgitic lithospheric mantle there was finally completely replaced by thin hot fertile lherzolitic mantle nowadays (only tens of km thick now). By contrast, the cool refractory lithospheric mantle of its western part is still being well preserved due to feeble asthenospheric upwelling (*pls see the three small columns, from west to east, at the lower right part of the diagram below*).

[Redacted]

Having these background information, we are confident that there was a heating period before the high-Cr chromite mineralization. Even so, we would like to

specially emphasize that the parental magma sources of high-Cr chromitites overall had moderately refractory compositions rather than FMM-like fertile ones, because the pre-mineralization refertilization was not that intensive and cannot be compared with what happened below the North China Craton at all.

**Q2.4.2 Were the parental magmas of high-Al chromitites (depleted tholeiitic) formed under lower temperature conditions than FAB? (Yes, the LAB fell down with the harzburgitization of the uppermost mantle and caused lower geothermal gradients. We have mentioned these information in L353-358)**

This issue can only be qualitatively evaluated. As we have mentioned above and in the discussions, except the special case of mantle plume, asthenosphere is the main source of heat for lithospheric mantle, and the geothermal gradient of lithospheric mantle depends on the position (depth) of the lithosphere-asthenosphere boundary (LAB). **The shallower the LAB is, the hotter the lithospheric mantle is** (pls see the critical dots in yellow colors in the diagram below; the depths of these dots mark the LAB). On one hand, FAB was formed via decompressional melting of fertile asthenospheric mantle below spreading centers, just as if how MORB were generated. On the other, the parental magmas of high-Al chromitites with transitional FAB-boninitic compositions were derived from harzburgitic mantle sources left after extraction of the FAB. That is to say, **from the FAB magmatism to the transitional ones, the mantle sources of magmas changed from fertile asthenospheric mantle to refractory harzburgitic ones.** Thus, the lithospheric mantle below proto-forearc spreading centers must have been thickened after the FAB magmatism (making up the proto-form of subarc mantle), and **the LAB would accordingly have fallen down to deeper levels during the transitional magmatism. As a result, the geothermal gradient during the transitional magmatism could not be higher than that during the FAB magmatism,** although it cannot be determined how large the gradient was decreased.

[Redacted]

**Fig. 2.4.1** Variation of geothermal gradients in lithospheric mantle with the LAB depth. The steep grey line defines the geotherm of the adiabatic asthenospheric mantle, while those gentle-sloped ones represent the geotherms in lithospheric mantle. The depths of the yellow dots mark the LAB. Overall, the shallower the LAB is, the hotter the lithospheric mantle is.

[Redacted]

**Q2.4.3 Were the parental magmas of high-Cr chromitites (boninitic-like) formed under lower temperature conditions than FAB? (It is difficult to make the comparison, but luckily, this is not our focus)**

Before answering the questions, we would like to reiterate again that it is not our aim to compare the formation temperatures of FAB and boninitic magmas directly, and it is not easy to make comparisons for them, from both the quantitative and qualitative perspectives. This is because the fertility and fluid contents in their respective mantle sources are different from each other. Moreover, the formation temperatures of boninitic-like magmas themselves would vary from case to case. Based on melt-inclusion studies, some boninitic melts were even generated under temperature nearly 1500 °C.

What we can tell as general is that both the FAB and the parental magmas of high-Cr chromitites were formed at higher temperature conditions than the parental magmas of high-Al chromitites. That is why we think the formation period of high-Al chromitites marked the trough period of heat flux in the proto-forearc mantle. Moreover, the boninitic-like parental magmas of high-Cr chromitites were possibly generated at temperature conditions closer to the FAB (or slightly lower than the FAB based on the LAB position, but it is hard to determine the exact difference, pls also see the diagrams below).

[Redacted]

(Note that transitional FAB-boninitic magmas were formed in similar ways to magmas developed in mature arcs, but the two are not exactly the same. We just used the diagram for a reference)

**Q2.4.4 How to reconcile the inconsistent data of temperatures? (the calculated temperatures for evolved products based elemental equilibration cannot represent the formation temperatures of magmas themselves, which should be much higher)**

Actually, mantle-derived lavas erupted on the surface cannot be completely primary, even though they are featured by high Mg# (> 60). This is because fractional crystallization was unavoidable as the magmas migrated upward, especially in magma chambers. Even as for lavas with boninitic compositions, which people traditionally think primary due to their high MgO contents and Mg#, fractional crystallization of olivine and chromite must have happened at depth as well. Otherwise, there will be no olivine and chromite phenocrysts in boninitic lavas. When pyroxene started to be generated from boninitic magmas, evolution must have happened to the magmas, and the temperatures of the magmas must have decreased to certain degrees. That is, the

crystallization temperatures of pyroxenes cannot represent the formation temperatures of magmas, which should be higher. For your reference, we list below the calculated formation temperatures of FAB by Shervais et al. (2019) (left diagram below) may reach 1450 °C by using the method of Lee et al. (2009), higher than the crystallization temperatures 1142-1190 °C for clinopyroxene in FAB.

[Redacted]

Specifically, ophiolitic chromitites are mainly composed of chromite and olivine, with accessory clinopyroxene filling the interstitial spaces (some plagioclase for high-Al chromitites). Such an mineral association means that effective precipitation of chromite and olivine must have happened before the compositions of their parental magmas evolved to the stability field of clinopyroxene, which means that the magma temperatures had been obviously decreased then and could not represent the original magma temperatures.

Although olivine and chromite are the first two phases crystallized from Mg-rich magmas, it is the same that the obtained results based on thermobarometers between them can only represent the lower limits for their crystallization temperatures, rather than the formation temperatures of their parental magmas themselves. Especially, it should be emphasized that presence of water in magmas delays the crystallization of olivine and chromite, which is a common sense and can be easily testified by using the software of MELT. As a result, any calculated crystallization temperatures based on the compositions of pyroxene and evolved magmas cannot represent the formation temperatures of the primary magmas, not to mention that pyroxenes was not the first phase to crystallize from parental magmas of chromitites.

**Finally**, we would like to say thanks again for the comments, which enhanced our understanding of the meanings of different calculated temperatures. That is, any calculated crystallization temperatures based on the compositions of pyroxene and evolved magmas should be lower than the formation temperatures of the primary magmas. Anyway, our comparisons show that high-Al chromite mineralization and related low-Ti tholeiitic magmatism took place at the trough period of geothermal gradients during subduction imitation, lower than those during the FAB and (high-Ca)

boninitic magmas. Again and importantly, it is not our aim to compare the formation temperatures of FAB and boninitic magmas directly, because high-Al chromitite swere not generated by FAB magmatism but by more depleted ones. It is not easy to make comparisons for them either. Combined with the comments and suggestions of other reviewers, we have added more lines in the related section (Pls see below).

[Redacted]

Please also check in the main text the information regarding how fluctuation of LAB depth affected the geothermal gradient, as we have explained in detail above.

[Redacted]

*Q2.5 The sections titled "Origins of different types of chromite deposits" and "Geodynamic implication" lack sufficient support from the data presented. For instance, the statement regarding a "climax stage of slab impact in the*



*proto-forearc mantle" requires clarification, especially in light of the minimal fluid contribution from the subducting plate during subduction initiation, as documented in Ishizuka et al. (2014) and Whattam and Stern (2011).*

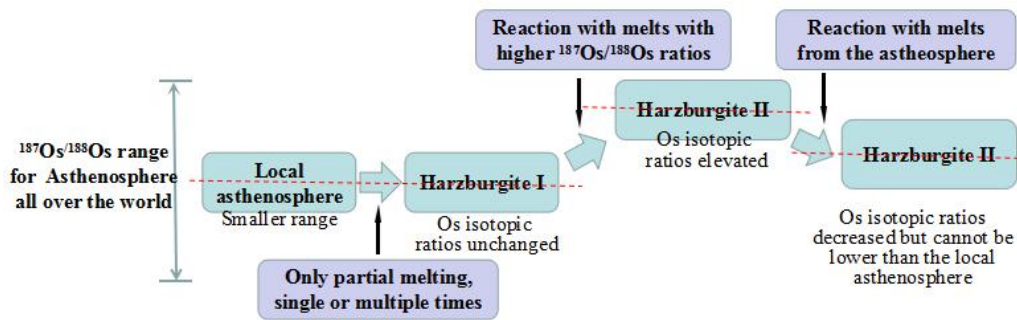
**Our reply:** Many thanks for the questions first. Sorry that we did not have these points clarified, so some misunderstanding were caused. Again, we would like to re-state that the parental magmas of high-Al chromitites have depleted low-Ti tholeiitic compositions, transitional compositions between FAB and boninitic ones, rather than typical FAB. This was how the problem came. Remarkable dehydration did happen at the stage of low-Ti tholeiitic magmatism. In order to better answer this question, we divided it into two minor ones. In particular, we explain the meaning of “slab impact” in the second minor question. Please see our explanations below.

**Q2.5.1 Were there slab contributions in the parental magma sources of all the chromitites? (*Yes, supported by their Os isotopic compositions*)**

As we emphasized previously, the main reason that you asked this question is you think the parental magmas of high-Al chromitites were normal FAB magmas. According to our reasoning in Q2.1, however, their parental magmas must be more depleted ones. You can call them “depleted FAB”, “transitional FAB”, “depleted tholeiitic basalts” and anyone matching their compositions, but they are indeed more depleted than the exactly normal FAB. As we replied to the Q2.1.3, from the FAB magmatism to transitional ones, the mantle sources of magmas changed from fertile asthenospheric mantle to refractory lithospheric ones (not that thick), and the geothermal gradient during the transitional magmatism also decreased due to the falling down of the LAB. With the combined effects of harzburgitization and lowered geothermal gradient, a second stage of partial melting became difficult in the proto-forearc mantle, and addition of slab fluids would be highly necessary in order to initiate further melting in the mantle. This explains why some high-Al chromitites have high  $^{187}\text{Os}/^{188}\text{Os}$  ratios.

As we mentioned in the main text (L306-311 before the revision) and our reply to the Q2.1.3 (we will repeat the reply here for the convenience of your check), the mantle sources of all our chromitites, including both the high-Al and high-Cr ones, had been modified by slab fluids, even though their  $^{187}\text{Os}/^{188}\text{Os}$  ratios fall into the range of global abyssal peridotites. Such a conclusion was obtained based on comparison with the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of harzburgites analyzed in our study. As we discussed in the L275-300 (new version), the origins of harzburgites in ophiolitic mantle sequences can be initially traced back to decompressional melting of asthenosphere, despite their melt modification histories. Bearing in mind that the process of partial melting alone cannot fractionate Os isotopes, harzburgites from the mantle sequence of some an ophiolites should have indistinguishable Os isotopic ratios to the local asthenosphere, as long as only partial melting (single or multiple times) happened to the harzburgites. However, the fact is that harzburgites in the mantle sequences of ophiolites had been modified by parental magmas of chromitites, all of which had higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the harzburgites, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the photo-liths of the harzburgites should be even lower than those of the

harzburgites. Specific to the case of the Zambales ophiolite, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the local asthenosphere below the Zambales proto-forearc before the formation of chromitites could not be higher than those of the harzburgites we analyzed, all below 0.1275 and smaller than the ratios of all the Zambales chromitites. Consequently, the mantle sources of all chromitites must have contributions of slab fluids, even though their  $^{187}\text{Os}/^{188}\text{Os}$  ratios fall into the range of abyssal peridotites. Please see how the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the harzburgites changed during different processes in the flowchart below (note the relative positions of the dashed red lines).



**To put it simple**, the range of abyssal-like  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios was established based on a large dataset from the global, but the  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratio of asthenosphere below some a place unlikely have such large variation but more likely fall in a narrow range of its own. To judge if there are slab contributions in the magma sources, we need to compare their Os isotopic ratios with their associated harzburgites rather than simply with the range of the abyssal peridotites.

**Q2.5.2 Why did mineralization of high-Al chromite overall mark climax stage of slab impact in the proto-forearc mantle during subduction initiation? (*They have the highest  $^{187}\text{Os}/^{188}\text{Os}$  ratios*)**

To answer this questions, we need to know the contributions of slab components in the proto-forearc mantle during each stage of magmatism, and the Os isotopic ratios of magmatic products are the index of our judgement. Our study involves three stages of magmatism, including the earliest stage of FAB magmatism, a second stage of high-Al chromite mineralization (transitional magmatism), and a third stage of high-Cr chromite mineralization (high-Ca boninitic magmatism). Please note that the framework of our study did not include the stage of low-Ca (high-Si) boninitic magmatism, which happened after the high-Cr chromite mineralization and marked the start of real subduction according to previous studies, as was mentioned in papers. Although we mentioned some information about low-Ca boninitic magmatism, such a stage did not account for chromite mineralization (pls see our reply to the Q2.4.4 regrading the sequence of crystallization and the subtitle), so this stage is beyond the focuses of this study (diversities of chromite mineralization).

On on hand, **let's see the stage of FAB magmatism that happened before the high-Al chromitite formation**. As you emphasized over and over again, **only tiny amounts of slab fluids were involved in the mantle during FAB magmatism, just like what happened in the MORB mantle**. We quite agree with this idea and have no problem with it at all. That is, the FAB magmatism happened under relatively fluid-poor conditions (although there were indeed some). Actually, this is also the

precondition for our conclusion. Based on the genesis of FAB and  $^{187}\text{Os}/^{188}\text{Os}$  data of harzburgites (initially residues of FAB but later modified by melts, having the lowest  $^{187}\text{Os}/^{188}\text{Os}$  ratios) in our study, the products of FAB magmatism likely have  $^{187}\text{Os}/^{188}\text{Os}$  ratios no more than 0.1275, overall lower than ratios of both the high-Al and high-Cr chromitites.

On the other hand, let's see the two stages of chromite mineralization. According to the Os isotopic distributions, the highest  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios of chromitites were obtained for high-Al chromitites. Such a feature happened in each ophiolite that have coexisting high-Al and high-Cr chromitites, supporting that it is possibly a prevalent feature for ophiolitic chromitites. If we consider the  $^{187}\text{Os}/^{188}\text{Os}$  ratios as the proxy of slab contributions in the mantle sources of magmas, the contributions of slab components in the parental magma sources of the high-Al chromitites must be larger than those in the parental magma sources of the high-Cr chromitites.

By using the term “impact”, we are saying the proportions of slab contributions rather than their absolute values in the mantle sources, of course, from the perspective of Os isotopes. As we mentioned before, additions of slab fluids played an important role in causing partial melting in the parental magma sources of high-Al chromitites, mainly because of the lower geothermal gradient during the mineralization. However, due to the effect of asthenospheric upwelling, there were contributions not only from slab components but also from asthenospheric components. That is to say, the effects of slab components would be diluted with the presence of asthenospheric components, and dilution effects would be more and more intensive with the continuous of asthenospheric upwelling. As a result, “slab impact” used in our study means the ratio  $[\text{Os}_{\text{slab}}/(\text{Os}_{\text{slab}}+\text{Os}_{\text{asthenosphere}})]$  but not the absolute amount of slab-derived Os added to the proto-forearc mantle. Thus, at least three points can be inferred: (1) without notable asthenospheric component added, the impact of slab-derived Os outweighed; the lower  $^{187}\text{Os}/^{188}\text{Os}$  ratios of high-Cr chromitites does not necessarily mean fewer slab-derived Os were added into their parental magma sources; and (3) having the highest  $^{187}\text{Os}/^{188}\text{Os}$  ratios only means notable slab dehydration but does not necessarily mean the most intense period of slab dehydration. Please see the diagram below for the illustration.



probably the most noticeable during the stage of high-Al chromite mineralization.”

*In conclusion, this manuscript by Zhang and colleagues requires substantial revisions and additional supporting evidence before it can be considered for publication in Nature Communications. The proposed model must be rigorously tested against the existing body of knowledge related to subduction initiation and chromitite formation. It is my sincere hope that these comments will aid the authors in improving the quality and robustness of their work.*

## Minor suggestions/corrections

### INTRODUCTION

- Lines 31-34: the sentence is unclear

**Our reply:** Many thanks for the question. We have re-written the abstract due to the required word limit. Please see the new version there.

- Line 46: delete “extraction”

**Our reply:** Many thanks for the suggestion. We have deleted “extraction” from the sentence in the revised version. Please have a check there.

-Line 48: Cr# is not referred as “wt%”, correct the statement. In addition, with Cr#=7 the mineral probably cannot be classified as chromite.

**Our reply:** Many thanks for pointing out the mistake. We have deleted the “wt.%”. Yes, you are quite right. The boundary Cr# value between spinel and chromite is actually 50. However, it has become our tradition to call both spinel and chromite collectively as chromite in podiform chromite deposit-related studies. That is, those with < 50 Cr# are usually called high-Al chromite, those with > 50 Cr# called high-Cr chromite. As for chromite with Cr# < 10, the host rocks is actually a piece of chromite ore from the Muğla ophiolite, Turkey (please see the reference below). It is not an piece of average peridotites. Of course, it is fine to call it as spinel orerock, but we do not have such a tradition. What we did are just following the traditions and not the first one doing so.

Discovery of extremely high-Al podiform chromitites from the Lycian (Marmaris) ophiolite, SW Turkey: Implications for chromitite genesis  
Y Xu, S Chen, O Parlak, S Arai, C Dönmez, J Hong - Ore Geology Reviews, 2020 - Elsevier  
... high-Cr chromitites were crystallised, suggesting a boninitic affinity. The extremely high-Al chromitites possibly formed through faint interactions between intruding high-Al magma and ...

Even for spinel in harzburgites, some like calling them as magnesio-chromite, while others like calling them as chrome spinel (Cr-spinel), regardless of their exact Cr# (< 50 or not), so the definitions of chromite and spinel have been blurred now, at least in ophiolite-related studies (pls see the references below). Hope you would be fine with our explanation.

Composition and textures of chromite and platinum-group minerals in chromitites of the western ophiolitic belt from Pampean Ranges of Córdoba, Argentina  
JA Proenza, F Zaccarini, M Escayola, C Cávana... - Ore Geology ..., 2008 - Elsevier  
... of a metamorphosed suite of harzburgites and subordinated impregnated ... accessory chromites in metamorphosed dunites and harzburgites are systematically altered to ferrian chromite ...

Podiform chromitite-bearing ultrabasic rocks from the Braganca massif, northern Portugal: Fragments of island arc mantle?

JC Bridges, HM Prichard, CA Meireles - Geological Magazine, 1995 - cambridge.org

... harzburgite is used in this paper to avoid confusion with the separate orthopyroxene-free dunite layers within which chromite ... Accessory chromite grains in the Braganca harzburgite ...

-Line 50: include the reviews on chromitites by González-Jiménez et al.

González-Jiménez, J. M., Griffin, W. L., Gervilla, F., Proenza, J. A., O'Reilly, S. Y., & Pearson, N. J. (2014). *Chromitites in ophiolites: How, where, when, why? Part I. A review and new ideas on the origin and significance of platinum-group minerals. Lithos, 189, 127-139.*

González-Jiménez, J. M., Griffin, W. L., Proenza, J. A., Gervilla, F., O'Reilly, S. Y., Akbulut, M., ... & Arai, S. (2014). *Chromitites in ophiolites: How, where, when, why? Part II. The crystallization of chromitites. Lithos, 189, 140-158.*

**Our reply:** Many thanks for the suggestion. These two references are very impressive, and actually, we are also familiar with the two papers. Due to the limitation of total number of papers in the reference list, we decided incorporated the second paper into the revised version, because the main text of the second paper is more suitable here. Please have a check.

- Line 54: correct the reference to Xiong et al.

**Our reply:** Many thanks for the reminder. This is because we have two guys sharing the same family name Xiong, but with different given names. However, this problem will not exist in the revised version, because the format of citation have been changed. Please see the change in the revised version of our manuscript.

- Lines 55-59: include the references to support this statement

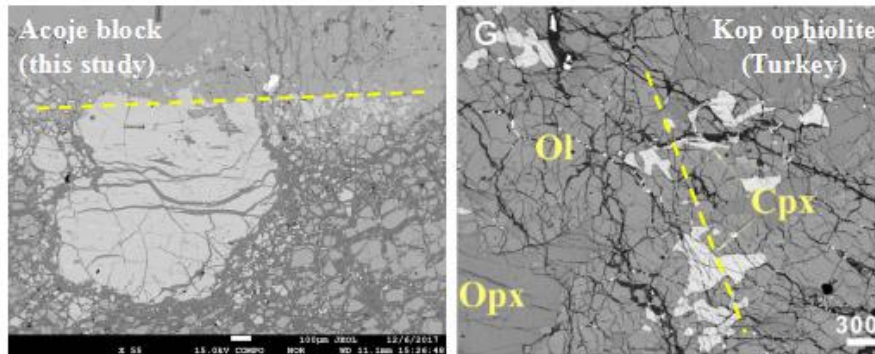
**Our reply:** Many thanks for the suggestion. This statement was not directly cited from published papers. Instead, it was found by ourselves after years of work and based on compilation of cases from worldwide, which were shown in the Fig. 1 already. **All the cited cases had also been shown in the Fig.1 and its caption.** Since the journal has limitation for overall length and numbers of references, it is better not put them in the main text. That is why we used the “**Fig. 1 and references therein**” instead. Even so, **we have to move all the citations to the supplementary files (Supplementary References)** due to the length limit required by the journal. As a result, we were not deliberate to do so. Hope you can understand this situation.

- Line 57: Figure 1 shows the range of Cr# in chromitites and associated rocks. It does not include the origin of the peridotites or the presence of diopsidite veins. Change the sentence, the reference to the figure, or change the figure.

**Our reply:** Many thanks. We quite agree with what you said. Just as you told, it is better to extend the contents here based on the contents of the diagram. **We have changed the sentence as “ophiolites which host high-Cr chromitites mostly contain lherzolites and Cpx-rich harzburgites in their mantle sequences”,** although the term “abyssal peridotite” itself generally refers to “fertile peridotites” in the context of ophiolite-related studies.

Moreover, we wanted to say that Cpx-rich harzburgites (3-5 modal%) cannot be

distinguished from Cpx-poor lherzolites (5-10 modal% Cpx) sometimes, especially when Cpx grains are heterogeneously distributed in a piece of peridotite. In this case, some part of the peridotite may look like Cpx-rich harzburgite, while other parts may look like lherzolite or even wehrlite and pyroxenite. Such heterogeneous distribution of minerals can be explained as the result of melt refertilization along some micro-scale infiltration channels of melts. As we mentioned for times, the refertilizing melts had asthenospheric origin.



- Line 58: add “formation of” before “high-Cr chromite mineralization”.

**Our reply:** Many thanks for the suggestion. However, the term “mineralization” itself means formation of high-Cr chromite deposits, so it’s better not to have the two word used together. As per your suggestion, we have revised the sentence as “ during the formation period of high-Cr chromitites. Hope you are fine with such a change.

- Lines 59-60: include references for this statement.

**Our reply:** Many thanks for the suggestion. This statement was not directly cited from published papers. Instead, it was found by ourselves after years of work and based on compilation of cases from worldwide, which were shown in the Fig. 1 already. All the cited cases had also been shown in the Fig.1 and its caption. Since the journal has limitation for overall length and numbers of references, it is better not put them in the main text. That is why we used the “[Fig. 1 and references therein](#)” instead. Even so, we have to move all the citations to the [supplementary files \(Supplementary References\)](#) due to the length limit required by the journal. As a result, we were not deliberate to do so. Hope you can understand this situation.

- Line 66: indicate examples of this type of subduction-initiation ophiolites

**Our reply:** Many thanks for the suggestion. It is really helpful. We have added the cases of the Troodos (Cyprus), Semail (Oman), and Zambales (Philippines) ophiolites here. Please have a check.

- Line 89: the reference to Fig. 2B-D in this sentence is not pertinent

**Our reply:** Many thanks for the reminder. After reading this question, we also think it is better to make some adjustment. Now, the positions of Fig. 2C and 2D have been transposed in the new diagram in order to keep the normal sequence for their first appearances in the main text. Please see the changes in the revised version.

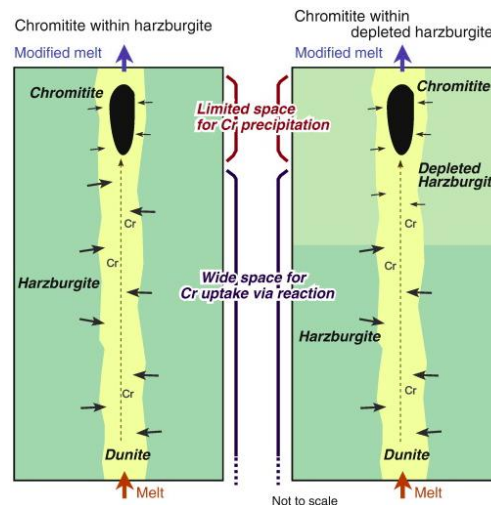
- Line 101: the abbreviation of Cpx should be capitalized (in the whole text).

**Our reply:** Many thanks for the reminder. We have made relevant changes across the whole main text. Please have a check in the revised version.

- Lines 113-116: the sentence is not clear, indicate that this is an interpretation from

previous works. The presence of high-Cr chromite in dunites in the ophiolitic sequence is typical, hence it is not that straightforward that it indicates that high-Cr chromitites are younger than the high-Al ones.

**Our reply:** Many thanks for the question. As we mentioned before, it is a common sense that dunites in the mantle sequences of ophiolites were generated by reaction between harzburgites and Mg-rich magmas (pls see the diagram below from *Arai and Miura, 2016, Lithos*). Such a process not only dissolves pyroxene in the harzburgites but also can elevate the TiO<sub>2</sub> contents of chromite. As a result, the dunite part in the diagram must have formed later than the harzburgite part. This is very clear and can be easily understood. However, it is more important to prove that the harzburgite part in this sample was particular modified product by the parental magmas of high-Al chromitites or similar melts. Please see our detailed explanation below.



#### Formation and modification of chromitites in the mantle

S Arai, M Miura - *Lithos*, 2016 - Elsevier

... podiform chromitites, but also changes our understanding of the whole picture of podiform chromitite ... for hydrothermal modification/formation chromite/chromitite in the mantle, which is a .

Q(a) Was the harzburgite part partial melting residue or modified products of melts? (yes, definitely melting residue)

Actually, if you look at the compositions of chromite in the harzburgites, you will find that these chromite grains are all high-Al ones (Cr-spinel) and have TiO<sub>2</sub> contents around 0.13 wt.%. Comparatively, residual chromite left by partial melting, especially in cpx-free harzburgites (surely > 20% partial melting), should have very low TiO<sub>2</sub> contents (mostly < 0.05 and even close to zero), due to the incompatibility of Ti during partial melting. As a result, the high TiO<sub>2</sub> contents of chromite in the harzburgite must be caused by melt-rock reaction (what kind of melt? Pls see in Q(b)), which elevated the TiO<sub>2</sub> contents of chromite beyond the level of partial melting residues, as we mentioned above.



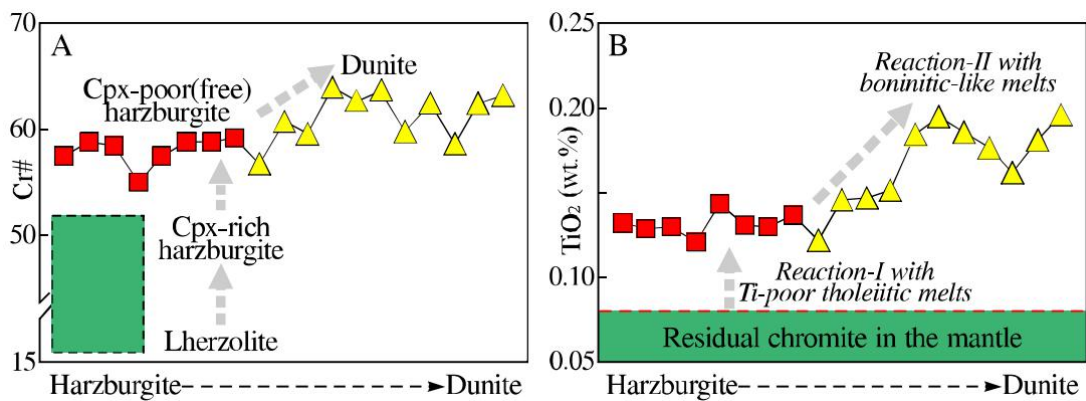
[Redacted]

Effect of melt-rock reaction (e.g., red arrow in the left diagram) on the compositions of chromite in harzburgites. Due to the high concentrations of Ti in melts, reaction with melts would overall elevate the  $\text{TiO}_2$  contents of chromite in the harzburgites, and the elevated extents depend on the melt/rock ratios.

Q(b) Which kinds of melts elevated the  $\text{TiO}_2$  contents of chromite in the harzburgite? parental magmas of high-Al chromitites or those of high-Cr ones? (*the early one*).

Based on the Cr# of chromite in the dunite part, the reactant melt generated the dunitites should have boninitic-like compositions (not that typical, more affinitive to basaltic boninite). During the reaction, the reactant melts diffused from magma bodies into surrounding peridotites, with the melt/rock ratios decreasing gradually. Generally, the higher melt/rock ratio was (mainly in the dunite part), the more these values were elevated. In zones where the melt/rock ratios were very low (mainly in the preserved harzburgite part), the Cr# and  $\text{TiO}_2$  values would not be elevated and keep unchanged. Although reaction with Mg-rich melts would elevate the Cr# and  $\text{TiO}_2$  contents of chromite in the harzburgites to certain degrees at the same time, increases of both the values during one single reaction event should be successive rather than in an abrupt way, due to the successively decreasing melts/rock ratios from magma bodies to host peridotites.

In the two provided diagrams (below), elevation of both the Cr# and  $\text{TiO}_2$  values were mainly found for the first four or five chromite grains in the dunite part, but those in the harzburgite part have stable Cr# and  $\text{TiO}_2$  values, without notable increase trends of both values. Such features suggest that the chromite grains in the harzburgite part were not remarkably affected by the dunite-forming reaction but another different one. Based on the Cr# of chromite in the cpx-free harzburgites, the reactant melts involved in the harzburgites had composition similar to the parental magmas of high-Al chromitites.



**To be concluded**, the composite sample first reacted with low-Ti tholeiitic melts (R-I), resulting in cpx-free harzburgites and the elevated Cr# (nearly 60) and TiO<sub>2</sub> (~0.13 wt.%) in it. Later, the cpx-free harzburgites reacted with boninitic-like melts (R-II) and was partly transformed as dunites. The Cr# and TiO<sub>2</sub> contents of chromite were further elevated again during the R-II. In particular, **the contact zone between dunite and harzburgite was featured by not that high melt/rock ratios** (the value gradually increased from harzburgite to dunite), so the Cr# and TiO<sub>2</sub> contents of chromite (the first four or five chromite grains in the dunite) were only moderately elevated.

Finally, **we just want to use the example and explanation above to illustrate the temporal sequence of different magmatism, which has actually been well recognized in the framework of subduction initiation**. More detailed explanations can be found in our paper published on Lithos in 2020. Moreover, **we have moved this part to the supplementary file (Supplementary Fig-1)**.

Coexistence of high-Al and high-Cr chromite orebodies in the Acoje block of the Zambales ophiolite, Philippines: Evidence for subduction initiation

PF Zhang, MF Zhou, GP Yumul Jr - Ore Geology Reviews, 2020 - Elsevier

- Line 116: figure 4 is not clear and maybe not needed. Explain the figure (including the different pictures and diagrams) or change it.

**Our reply:** Many thanks for the suggestion. As we replied to your last question above, **we just wanted to use the example to illustrate the temporal sequence of different magmatism (from FAB to low-Ti tholeiitic and finally to boninitic ones with time), which has actually been well recognized in the framework of subduction initiation**. As a result, we quite agree with you that we may not need this figure. It is better to put them in the supplementary materials if anyone want to read it. We have done this according to your suggestion. Many thanks.

## RESULTS AND DISCUSSION

- Line 130: it is better to use the term “ophiolitic chromitites” instead of “podiform chromitites”

**Our reply:** Many thanks for the suggestions. We have made the revision accordingly. Please have a check there.

- Lines 153-155: include the used equations to estimate the melts in equilibrium.

**Our reply:** Many thanks for the suggestion. We have added the equations into this part. Please have a check there.

- Line 159: indicate the composition of boninite used to compare and the reference.

**Our reply:** Many thanks for the suggestion. The overall TiO<sub>2</sub> contents of boninites are less than 0.5 wt.% according to many publications, e.g., Ishizuka et al. (2020, JP) and Perez et al. (2018, Solid Earth). In particular, the paper of Perez reported the compositions of boninitic lavas from the Zambales ophiolite. We have added such a values and relevant papers into the line. Please have a check there.

- Line 161: indicate the compositions of FAB to low-Ti tholeiites used to compare and the references.

**Our reply:** Many thanks for the suggestion. Again, we would like to state that the parental magmas of high-Al chromitites do not have typical FAB (MORB-like tholeiitic) compositions (mostly, TiO<sub>2</sub> > 0.8 wt.%; some, low to 0.6 wt.%). Instead, they show more affinities to depleted FAB (overall TiO<sub>2</sub>, 0.2-0.8 wt.%), which is also called transitional FAB, transitional FAB-boninitic lavas, transitional lavas, low-Ti tholeiites and etc. in different literature. Please see our reply to the Q 2.1.1.

To better compare the parental magma compositions of high-Al chromitites with different lavas, we added the TiO<sub>2</sub> range of FAB (normal FAB), low-Ti tholeiites (including depleted FAB), boninitic lavas into the Fig. 5B (Fig. 4B in the new version). The data of FAB, low-Ti tholeiites, and boninitic lavas (including LSB and HBS) were from Reagan et al. (2010, G3); Morishta et al. (2011; Geology), Shervais et al. (2019, G3) and Portnyagin et al. (2009, EPSL); and Ishizuka et al. (2020, JP) and Perez et al. (2018, Solid Earth). respectively.

Moreover, we have changed the sentence as “the parental magmas of high-Al chromitites are found, not as traditional thought, akin to typical MORB-like magmas (mostly > 0.8 wt.% TiO<sub>2</sub>), but show obvious affinity with more depleted low-Ti tholeiitic magmas (0.2-0.8 wt.% TiO<sub>2</sub>; Fig. 5B)”, which will help to emphasize our new idea here.

- Line 161: figure 5C should have been cited before 5D, figure 5C is not cited in the text, correct or remove the figure.

**Our reply:** Many thanks for pointing out the problem. Sorry for the mistake. We have removed Fig. 5D from this line and added Fig. 5C into the next paragraph, e.g., by saying that “the parental magmas of some Ti-poor high-Al chromitites even have less TiO<sub>2</sub> than those of the high-Cr chromitites (Fig. 5B-C)”. Please have a check there.

- Line 181: explain the model that has been carried out to obtain these values.

**Our reply:** Many thanks for the question. Because of the length limit, we cannot put too much details about the calculation in the main text, but relevant details have been presented in the [Supplementary Note-2](#). In the revised version, we have pointed out that such information can be found in the Fig. 5D and the supplementary materials.

- Line 254: indicate the reference for the Os isotopic values for abyssal peridotites.

**Our reply:** Many thanks for the suggestion. The range of abyssal peridotites was compiled by Liu et al. (2018) based on previous studies, e.g., Brandon et al. (2000);

Harvey et al.(2006) and also his own work on the abyssal peridotites from the Gakkal ridge, Arctic Ocean (2008, Nature). Considering the limitation requirement for the total amount of papers in the references. We only added Liu et al. (2018) into the reference list. Moreover, this paragraph has been largely revised, and the paper Liu et al. (2018) were cited in the next paragraph.

Limited recycling of crustal osmium in forearc mantle during slab dehydration  
CZ Liu, Y Xu, FY Wu - Geology, 2018 - pubs.geoscienceworld.org

... Here we report Os isotopic compositions of peridotites from New Caledonia (southwest Pacific Ocean) ophiolites, which represent relics of a forearc mantle. Some New Caledonia ...

- Line 257: for the chromitites in Mugla, Turkey, it is not that clear that the high-Al chromitites have much higher Os isotopic ratios.

**Our reply:** Many thanks for the comment. Yes, the case of Muğla chromitites is a little different. Some high-Cr chromitites also show higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the range of abyssal peridotites. However, we can still find a negatively variation trend from the high-Al chromitites to the high-Cr ones. It was possibly because we put the data into a narrow column, making you feel that the variation trend is weak. **However, the trend indeed exists and was also mentioned by the authors themselves** (Uysal et al., 2009, CMP). Please see the original diagrams from the paper below.

[Redacted]

*(Please note that Uysal also plotted two duplicate samples in the diagram above, so they have 6 datapoints for the high-Al chromitites)*

Petrology of Al-and Cr-rich ophiolitic chromitites from the Muğla, SW Turkey: implications from composition of chromite, solid inclusions of platinum-group mineral ...

I Uysal, M Tarkian, MB Sadiklar, F Zaccarini... - ... to Mineralogy and ..., 2009 - Springer

- Lines 263-264: add reference(s) for the statement

**Our reply:** Many thanks for the suggestion. It has been widely recognized that slab components have higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the uncontaminated asthenosphere (also called as “convecting mantle”, “depleted mantle”, “MORB mantle”, depending

on the cases studied; please see the phrases in the paper of *Anderson (1995)* below). Detailed comparison can be seen in the diagram below from the *Saha et al. (2005, EPSL)*.

Lithosphere, asthenosphere, and perisphere

DL Anderson - Reviews of Geophysics, 1995 - Wiley Online Library

... the depleted mantle (usually called the "convecting mantle," "asthenosphere" or "upper mantle") not ... part of the sublithospheric shallow mantle and is constantly refreshed by recycling. It ...

Slab devolatilization and Os and Pb mobility in the mantle wedge of the Kamchatka arc

A Saha, AR Basu, SB Jacobsen, RJ Poreda... - Earth and Planetary ..., 2005 - Elsevier

[Redacted]

In order to better support our words, we have added the paper of Saha above and the other two classical ones as the references. In particular, one of them is working on podiform chromite deposits from the famous Troodos ophiolite. Please have a check.

The osmium isotopic composition of convecting upper mantle deduced from ophiolite chromites

RJ Walker, HM Prichard, A Ishiwatari... - ... et Cosmochimica Acta, 2002 - Elsevier

Formation of podiform chromitite deposits: implications from PGE abundances and Os isotopic compositions of chromites from the Troodos complex, Cyprus

A Büchl, G Brügmann, VG Batanova - Chemical Geology, 2004 - Elsevier

- Lines 288-289: add reference(s) for the statement

**Our reply:** Many thanks for the suggestion. Generally, formation of harzburgites from lherzolites requires consumption of pyroxenes (Opx and Cpx), which can be achieved via two basic ways or the combined effects of both. The first one is via partial melting, because pyroxene grains are more fusible than olivine, peridotites would become richer and richer in olivine and could even be converted to dunites at high enough partial melting degrees. The other way is more prevalent for peridotites

in the uppermost mantle sequence, via reaction with Mg-rich but Si-undersaturated melts. Such melts would dissolve pyroxene and precipitate olivine at the same time, making the modal% of olivine higher and higher. Of course, the combined effects of both could also convert lherzolites to harzburgites.

Sorry that we simply thought the statement was a common sense. In the revised version, we have added two classical papers about the origins of harzburgites in the reference list. Please see them below.

Formation of harzburgite by pervasive melt/rock reaction in the upper mantle  
PB Kelemen, HJB Dick, JE Quick - Nature, 1992 - nature.com

Multiple origins for mantle harzburgites: examples from the Lewis Hills, Bay of Islands ophiolite, Newfoundland  
SJ Edwards, J Malpas - Canadian Journal of Earth Sciences, 1995 - cdnsiencepub.com

- Lines 387-389: the authors should indicate the chromitites they are referring to

**Our reply:** Many thanks for the reminder. You clearly pointed out what we wanted to express. Sorry that we did not realize the problem. We have revised the subject of the sentence as “the compositional variation of high-Al chromitites in other ophiolites”. Please have a check there.

- Line 412: the word “meanwhile” does not make sense in the sentence

**Our reply:** Many thanks for the reminder. We have deleted the word from the line. Please have a check here.

-Lines 430: authors do not need to invoke slab roll-back to generate boninitic-like melts.

**Our reply:** Many thanks for the question. We quite agree that generation of boninitic magmas does not always require slab rollback during subduction initiation, but that depends on which types of boninitic magmas were generated. We were not saying that all of them were formed in the context of slab rollback. It is more reasonable to say that slab rollback in the context of our study was inductive for generation of boninitic magmatism, at least some of them, just as if how we did here (*slab rollback —high temperature —boninitic-like magmas*). Therefore, slab rollback is overall a helpful but not always a necessary condition for forming boninitic-like magmas. In order to better answer your question, we present more details below. Please have a look.

*Q(a): Has anybody reported before that some (not all) boninitic magmas were formed in the context of slab rollback? (Yes, specially some high-Ca boninitic magmas)*

Boninites can be further classified into low-Si and high-Si varieties, which are generally equivalent to high-Ca boninite and low-Ca boninite (although not exactly), respectively, from the perspective of compositions. Overall, the geodynamic settings for these different magmatism are quite different. For instance, in the subduction initiation model, formation of high-Si magmas were thought to roughly mark the beginning of real subduction and presence of embryonic arc, whereas low-Si boninitic magmas was still developed via flux melting but during spreading stage of subduction initiation, called the late stage of proto-forearc spreading (pls see the diagrams below from Regean et al, 2019, EPSL). Therefore, even from the theory of subduction initiation itself, some boninitic magmatism (be more exactly, should be high-Ca boninitic) can be developed at spreading setting, which is driven by slab rollback

below proto-forearcs. Particularly, we pointed out in the subsequent sentence that “such a geodynamic scenario was proposed by some to account for high-Ca boninitic magmatism”. Therefore, our postulation was somehow reasonable.

**Forearc ages reveal extensive short-lived and rapid seafloor spreading following subduction initiation**

[MK Reagan, DE Heaton, MD Schmitz, JA Pearce ... - Earth and Planetary ..., 2019 - Elsevier](#)

[Redacted]

*Q(b): Do we have any geological evidence to support that some boninitic magmatism could be developed in the context of spreading? (Of course, sheeted dykes have boninitic compositions sometimes)*

An ophiolite is generally composed of different sequences, but the sequence of sheeted dykes is the most representative feature to support that the ophiolite formed in spreading setting. As a result, if the sheeted dykes have boninitic-like compositions or chemical affinities, we can claim that at least some boninitic magmas were developed in spreading settings, and we do have such samples.

One of the most prominent cases is the sheeted dykes of Troodos ophiolite. Some sheeted dykes have boninitic compositions. Please see the narration from Ribeiro et al. (2023, CG) below.

[Redacted]

Here is another example reported by Dilek and Thy (2009, Lithos): the sheeted dykes of the Kizlidag ophiolite, Turkey, have boninitic compositions.

[Redacted]

In the Zambales ophiolite, some pillow lavas and dykes from northern part of the ophiolite (e.g., the Barlo district of the Acoje block) were found to have boninitic-like compositions. Please see their compositions below.

[Redacted]

*Q(c): Why does slab rollback facilitate generation of boninitic-like magmatism?*

Although the process of slab rollback itself feels simple, it controls development of proto-forearc spreading, extent of mantle upwelling and subsequent fluctuation of geothermal gradient below proto-forearcs. To be more specific, slab rollback spares spaces for the upwelling of hotter asthenosphere below lithospheric mantle, such that the lithosphere-asthenosphere boundary would move upward, with the geothermal gradient being elevated at the same time. However, it is also noted that the rate of slab rollback and proto-forearc spreading was not that fast because the lithospheric mantle was only refertilized to be cpx-rich harzburgites or cpx-poor lherzolites rather than fertile lherzolites. Relevant messages have been mentioned in our reply to the Q2.4. Please see more details there and also in the two diagrams below.



[Redacted]

*(The left diagram shows the formation temperature of different magmas. The right one illustrates the co-variation between geothermal gradient (grey lines) and position of the LAB (yellow dots))*

By contrast, the movement of slabs in mature subduction zones takes place in a down-dipping way; upwelling of asthenospheric mantle is weak, so the geothermal gradients in mature subduction zones are generally lower than those below spreading settings. As we explained in our reply to Q2.4, boninitic-like and other Mg-rich magmas (e.g., picritic) require higher geothermal gradients. This reasonably explains why boninitic-like lavas were mainly formed during subduction initiation or the initial stage of subduction, when the mantle geothermal gradients were still high enough to produce such Mg-rich magmas.

Despite the explanation above, we quite agree with your point that generation of boninitic magmas does not always require slab rollback during subduction initiation, and it depends on which types of boninitic magmas were generated. In our study, we think generation of high-Cr chromitites were linked to high-Ca boninitic magmatism, which were induced in the spreading context sometimes. Again, slab rollback is not always a necessary condition for forming boninitic-like magmas, but the process would help to elevate the geothermal gradient in the proto-forearc mantle, facilitating generating of Mg-rich magmas. In order to be more cautious for this sentence, we have revised it as “facilitating higher degrees of mantle melting and generation of more Mg-rich magmas (e.g., boninitic-like ones)”.

## **METHODS**

- Line 452: indicate how many samples of each and the coordinates

**Our reply:** Many thanks for the question. We used 18 chromitites and 10 harzburgites, which have been mentioned in the method. We added their coordinate in the table (Supplementary Table-1)

- Line 995: indicate the meaning of “LAB”

**Our reply:** Many thanks for the reminder. Sorry for not pointing out its full name. LAB is short for lithosphere-asthenosphere boundary and is a famous boundary in the inner earth. We have added this abbreviation into the main text and also its full name here. Please have a check.

## FIGURES

- Line 958: correct the reference

**Our reply:** Many thanks for the reminder. As we replied before, there are two guys sharing the same family name “Xiong” in the references, but their given names are different. However, this problem will not exist in the revised version, because the format of citation will be changed.

- Figure 1: Medellín is in Colombia, not Columbia

**Our reply:** Many thanks for the reminder. Sorry for the mistake. We have made the revision accordingly. Please have a check there.

- Figure 5: in B correct the country “Turkey”, C is not cited in the text

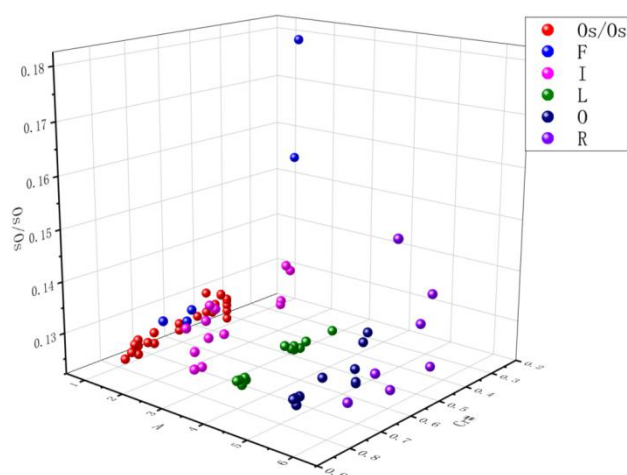
**Our reply:** Many thanks for the reminder. Sorry for the mistake, but we have removed the two cases from the diagram and compiled more data in the Supplementary Fig.2.

- Figure 6: the orange color in the legend does not match the yellow in the used symbol in A.

**Our reply:** Many thanks for the reminder. Sorry for the mistake. We have revised the color of the symbols already. Please have a check there.

- Figure 7: the figure would be easier to understand if the authors used the same scale for all the chromitites and differentiate those classified as high-Cr and high-Al with different colors.

**Our reply:** Many thanks for the suggestions. Actually, we tried to draw the figure as you told. However, the isotopic ranges of the Dongpo and Masquod chromitites are much larger than the other ones. **If we put them in the same scale, the decreasing trend of chromitites from other ophiolites cannot be well displayed, but the trends do exist based on their own scales** (note that a range 0.120-0.135 is not small for  $^{187}\text{Os}/^{188}\text{Os}$  ratios). Below is an old version of this diagram previously drew by us, which include all the data together but looks not good. Hope you can be understandable on this issue.



As for the other suggestion, we quite agree to use different colors to differentiate

the high-Cr and high-Al chromitites. However, please also highly note that the value 60 is only a reference boundary value of Cr# for classifying the high-Al and high-Cr chromitites. This is only a tradition, but there are still other ways of classification. For example, those with Cr# between 60-70 are sometimes called as intermediate ones, which means they are transitional from high-Al to high-Cr ones. Therefore, please only consider the value 60 as a reference boundary. For those chromite with Cr# around 60, it is better to consider them as transitional ones rather than exactly high-Al or high-Cr ones. Especially considering high-Al chromitites formed by transitional FAB-boninitic magmatism, such a viewpoint is reasonable. Take the case of the Zedang-Luobusa chromitites for example, chromite grains in some of the chromitites have Cr#s slightly higher than 60, but they are plotted close to the data points of high-Al chromitites, far away from those of the high-Cr chromitites, In this case, we also consider such chromitites more like as high-Al types rather than high-Cr ones. We have specified such information in the Figure caption and used different colors to tell apart the two groups of chromitites.

Again, many thanks for the suggestions. Please also check our revisions in the new version of our manuscript.

### Reviewer #3 (Remarks to the Author):

#### Comments:

This manuscript reports high-Al to high-Cr chromitites from classical Zambales ophiolite, Philippines. The elemental and Os isotopic compositions were presented to discuss parental magmas of chromitites and the modification from upwelling deep asthenospheric materials and sinking slab. Finally, mineralization conditions and stages of different types of chromitites in the Zambales proto-forearc mantle were well illustrated. This ms generally well-written, well-organized, and clearly argued. Below list major and minor comments, which are hopefully useful during revision.

**Our reply:** Many thanks again for your time and effort to our work. Your comments and suggestions are very helpful for improving our paper. We have replied all of them carefully and made revisions accordingly. Please see them one by one below.

#### Major comments:

*1-- For discussion part 1, maybe it is more clearly to discuss the parental magmas of chromitites from the Acoje and Coto blocks, separately, since the various types of chromitites may likely to make readers confused. A schematic diagram could be useful to show how FMM experiences 5-10% or 10-20% melting to mantle sources and then experiencing divers degrees of melting to parental magmas of various types of chromitites from the Acoje and Coto blocks, separately.*

**Our reply:** Many thanks for the suggestions. After reading the points above, we also find the possibility that readers may be confused by the interpretations of different chromitites. Therefore, it is quite necessary to make some changes here.

Overall, the calculated compositions for the parental magmas of chromitites depend on the compositions of chromite grains in chromitites. **As long as the compositions of chromite is fixed, their parental magma compositions would be fixed, no matter where the chromitites were collected from.** In the new version, we have separated the cases of high-Cr chromitites, Ti-rich high-Al chromitites, and Ti-poor high-chromitites, so that future readers will easily understand how their parental magmas were produced.

In the previous version of the manuscript, we mentioned that “the parental magmas of the high-Cr chromitites and Ti-rich high-Al chromitites were generated via ~15-25% and ~5-15% flux melting respectively of similar mantle sources, which themselves had experienced ~5-10% melting of fertile MORB mantle”, and that “the parental magmas of the Ti-poor high-Al chromitites were formed by ~5-15% flux melting of mantle sources that had undergone ~10-20% melt extraction from the FMM”. However, **we found such narrations are incorrect during the revision.** This is because the mantle sources of these magmas had experienced complicated melt modification event. For example, the parental magma sources of our high-Cr chromitites had been refertilized before the magmas were generated. As a result, **we think it better to say that their mantle sources had compositions equivalent to melting residues of the FMM after some partial melting degrees, rather than the melting residues exactly.** Moreover, we have depicted the formation processes of the parental

magmas of different chromitites in the final cartoon. You can see the variations of mantle fertility and geothermal gradients in the diagrams.

Many thanks for the question. Hope you are fine with our answers above. Please also see our revisions below.

168 (Fig. 4D; Supplementary Note-2), it appears that parental magmas of the high-Cr  
 169 chromitites were generated via ~ 15-25% flux melting of moderately depleted mantle  
 170 sources that themselves had compositions equivalent to ~ 5-10% melting residues of  
 171 fertile MORB mantle (FMM) (Fig. 4D). The parental magmas of the Ti-rich high-Al  
 172 chromitites from both blocks originated from sources with similar compositions to the  
 173 high-Cr ones but were generated via further 5-15% flux melting. However, parental  
 174 magmas of the Ti-poor high-Al chromitites were formed by 5-15% flux melting of  
 175 mantle sources that had compositions equivalent to 10-20% melt residues from the  
 176 FMM (Fig. 4D). The results above indicate that the parental magmas of all our

*2--You should make it clear when say high-Al chromitites, since both Acoje and Coto has them. This equivocal expression is all through your manuscript. The related statements should be clarified in revision.*

**Our reply:** Many thanks for the reminder. This suggestion is very helpful for making our contents clear. We also found such a problem and have clarified where the high-Al chromitites came from in the revised version when it is necessary. Please have a check in the new version of our manuscript.

*3-- Peridotites in Acoje and Coto also have chromites, how their Cr# and Ti values correlate to that of chromitites? Expecially in Acoje, chromitites were rounded by dunites as showed in Fig.3. what is the relationship between dunites and chromitites?*

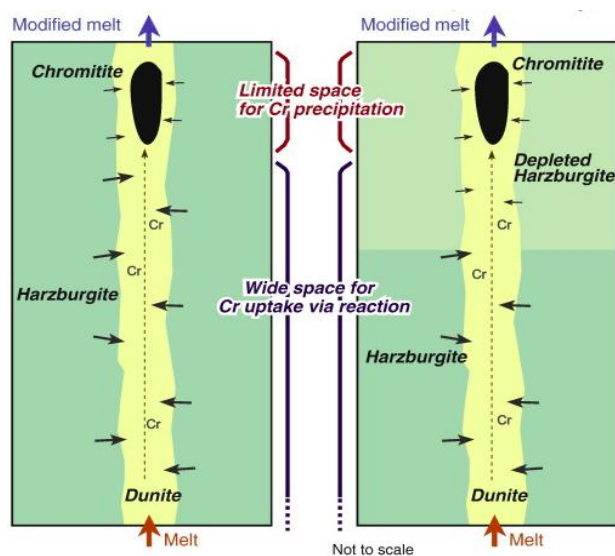
**Our reply:** Many thanks for the questions above. We quite understand your question above. Actually, many people are confused by the concepts of chromite, chromitites, chromite ores and chromite orebody. We will make simple explanations here.

Chromite	Spinel-group mineral; exist in varying mantle rocks; mostly as the accessory phase in peridotites; spinel (Cr#<50) is also called as chromite (magnesiochromite or high-Al chromite) for some researcher.
Chromitite	The name for rocks made of chromite and olivine
Chromite orerock	Generally refer to those chromitites from orebodies.
Chromite orebody	Mixture of chromitites and dunites; defining where can be explored; in the form of lenticular or irregular shapes.

For better answering your question, we have divided the question into three minor ones. Please see our replies step by step.

### Q3.1 how were chromitites and dunites generated in ophiolitic mantle sequences?

Chromitites are thought to be cumulate products of chromite and olivine from their parental magmas in melt-dominated environments, e.g., magma channels or chambers. Dunites can be either cumulate products of Mg-rich magmas (e.g., parental magmas of chromitites) or reaction products between harzburgites and Mg-rich magmas under different melt/rock ratio conditions. If the melt/rock ratio is high enough, the effects of melt-harzburgite reaction would be the same to that of crystal accumulation, with each mineral phases chemically equilibrated with the magmas. If the melt/rock ratio is not too high, the reaction may not be able to transform harzburgites to dunites, but it could make the harzburgites more depleted (please see the cartoon below). Meanwhile, the compositions of minerals in the harzburgites would also be modified, e.g., the Cr# and TiO<sub>2</sub> of chromite (please see more in the reply to Q3.3)



### Q3.2 Why are chromitites generally surrounded or accompanied by dunites?

Based on the formation processes of chromitites and dunites, it is easy to know that the two lithologies are genetically related. First, let's see the case when chromitites were produced in melt channels. The parental magmas of ophiolitic chromitites were all Mg-rich and Si-undersaturated. As such magmas migrated upward along channels in the harzburgitic (or even lherzolitic) mantle sequence, reaction avoidably happened between the magmas and the harzburgitic wall rocks, which would account for the formation of dunites and depleted harzburgites (pls see the cartoon above). Secondly, let's see the case when chromitites were formed in magma chambers or pods (usually in the MTZ). Chromite and olivine are the first two phases to be crystallized from Mg-rich magmas. Specially, when chromitites formed, chromite were the first phase to precipitate from the magma, followed by precipitation of more abundant olivine grains (at least several times larger than chromite; this is because Cr is only a minor elements in the magmas). Such a crystallization sequence would result in inter-layered chromitites and dunites. In both cases above, it is seen that formation of chromitites were accompanied by formation of dunites. That is why

ophiolitic chromitites are always associated with dunites. To a large scale, chromitite bodies are generally all enclosed in dunite bodies in the forms of pods, so ophiolitic chromite deposits are also called as podiform chromite deposits in literature.

For more information, you can refer to the papers shown below.

Formation of podiform chromitites by melt/rock interaction in the upper mantle

[MF Zhou](#), [PT Robinson](#), [WJ Bai](#) - *Mineralium Deposita*, 1994 - Springer

... We suggest podiform chromitites can form in much the same manner except that the melt composition is changed by melt/rock reaction rather than by magma mixing. In this model, ...

Podiform chromitites of the Tari-Misaka ultramafic complex, southwestern Japan, as mantle-melt interaction products

[S Arai](#), [H Yurimoto](#) - *Economic Geology*, 1994 - [pubs.geoscienceworld.org](#)

... a chromitite pod coated by a dunitic envelope and are collectively enclosed by clinopyroxene-bearing harzburgite. Chromitite ... in spinels of the enveloped chromitite than in those ...

Iron isotopic fractionation and origin of chromitites in the paleo-Moho transition zone of the Kop ophiolite, NE Turkey

[PF Zhang](#), [MF Zhou](#), [BX Su](#), [LUysal](#), [PT Robinson](#)... - *Lithos*, 2017 - Elsevier

... The paleo-Moho transition zone (MTZ) of the Kop ophiolite in NE Turkey is mainly composed ... melts with different  $\delta^{56}\text{Fe}$  values, accounting for the Fe isotopic variations of the Kop MTZ. ...

*Q3.3 how are the Cr# and Ti values of chromite in harzburgites correlate to that of chromitites?*

As we mentioned above, harzburgites had been in the mantle sequence no later than the formation of chromitites, but the compositions of harzburgites could be modified if the parental magmas of chromitites infiltrated into the surrounding harzburgites. Depending on the compositions of reactant melts, the variation trend of Cr# and  $\text{TiO}_2$  will be different. For example, reaction with boninitic melts would elevate the Cr#s and  $\text{TiO}_2$  contents of chromite at the same time. On the other hand, depending on the melt/rock ratios, the Cr# and  $\text{TiO}_2$  values of chromite would deviate from their original compositions to different degrees. For example, reaction under higher melt/rock ratio condition would make the compositions of chromite in the harzburgites closer to those of chromite in chromitites. Please see the two diagrams below for more variation trends.

[Redacted]

(The left diagram shows the compositional variation trend of chromite in harzburgites via reaction with different melts, after Pearce et al. 2000; CMP; the right diagram shows the compositional variation trend of chromite in harzburgites, dunites and chromitites toward two different directions; Zhang et al., 2020; OGR)

Before the end, we would like to emphasize that the genesis of chromite in mantle harzburgites (residual or modified products of melts) are quite different from that of chromite in chromitites (cumulate products). The geneses of harzburgites were not the main focus of this study, and our conclusions can be obtained without too many discussions on the harzburgites.

### Specific comments:

1. In figure 3. I do not understand the meaning of ordinate axis form 8Km to 1Km. Does this mean the depth of samples used? If so, please make it clear which rock type (chromitites, dunites, or Cpx-poor harzburgites) are these chromites and clinopyroxenes from.

**Our reply:** Many thanks for the question, and sorry for not making these issue clear. Yes, as you told above, the depths 1-8 km mean the distances to the MTZ-mantle borders (petrological Moho; the solid lines between the harzburgite and dunite zones) in Fig. 2C-D. To be more proper, they mark the stratigraphic depths of the sampling locations in the mantle sequences. For example, the stratigraphic depths of both the location 5 and 6 are zero, because both of them are just located on the MTZ-mantle border of the Acoje block. We have made the change already in the caption.

As we mentioned in the geological background, the mantle sequence of the Coto block comprises mainly Cpx-poor harzburgites, and there is no obvious variation of mineral proportions from top to bottom of its mantle sequence; whereas peridotites of the Acoje block vary from Cpx-poor harzburgites with enclosed dunite and Cpx-rich harzburgite (3-5 modal% Cpx) lens at the topmost mantle sequence to Cpx-rich harzburgites and Cpx-poor lherzolites at the bottom mantle sequence. Actually, the Fig.3 was specially drawn to illustrate the stratified mantle sequences of the Zambales ophiolite. and such stratified features can be understood from two mutually related perspectives, petrologically or compositionally. To put it simpler, the petrological variation in the mantle sequence can be revealed by compositional variation (rocks or minerals), and vice versa. However, it is impossible to show all the petrographic features in a small diagram. Instead, it is more realistic to show the compositional variation of minerals in the columns. This is because minerals in the Cpx-rich harzburgites and Cpx-poor lherzolites have higher affinities to abyssal peridotites and those in the Cpx-poor harzburgites, so we can roughly judge if the harzburgites are Cpx-rich or Cpx-poor varieties based on the mineral compositions. Of course, minerals in the dunites and chromitites would have much more depleted compositions, showing obvious affinities to boninitic and low-Ti tholeiitic magmas.

Among all minerals in peridotites, chromite and clinopyroxene are the most two sensitive phases to melt events, so their compositional variations can be best used for making up the diagrams. In the four columns, the data used were from two sources. One is from Evans and Hawkins (1989) that presented the data of both both blocks, the other from our own that presented the data of the Acoje block. We can see the variation trends (overall shapes) defined by our data and those of Evans and Hawkins (1989) are comparable. However, we could not tell the exact rock type for the red and white dots. This is because the paper Evans and Hawkins (1989), where the data of



the red and white dots are from, did not tell the exact rock type of each sample, although we can make general judgement based on the compositions of chromite and clinopyroxene, as we told in the second paragraphs.

In our work, we found that the middle and upper parts of the Acoje mantle sequence are composed of different harzburgites and dunites. In particular, Cpx-rich harzburgites, Cpx-poor harzburgites and dunites were found coexisting at uppermost part of the Acoje mantle sequence, accounting for the large compositional variation of chromite and clinopyroxene at this level (shaded green zones, from our own data, wider green zones mean presence of different peridotites). Accordingly, *it is better not to arrange some a rock type to define the lithology at a certain stratigraphic level in the mantle sequence.* Actually, we have shown in the Fig.2C-D how different peridotites are distributed in the mantle generally, and you can refer to the grey bars (Cpx-rich harzburgites) and white bars (Cpx-poor harzburgites) shown in diagrams.

Many thanks again for the suggestions. We have added more information in the figure caption, which would make the diagram better understood. Please have a check there.

*2. No scale in figure 4.*

**Our reply:** Many thanks for the reminder. We have added scale bars to the photos. Moreover, the figure has been moved to supplementary Fig-1 according to the suggestion of the Reviewer 2#. Please have a check there.

*3. In figure 5B, what is the hollow triangle represent?*

**Our reply:** Many thanks for the question. They are chromite grains hosted in the Lau basin basalts. Lau basin is a newly formed back-arc basin in the West Pacific. We just put them here for comparison so that you will know where chromite in BABB would be plotted in the diagram. We have clarified the legends already.

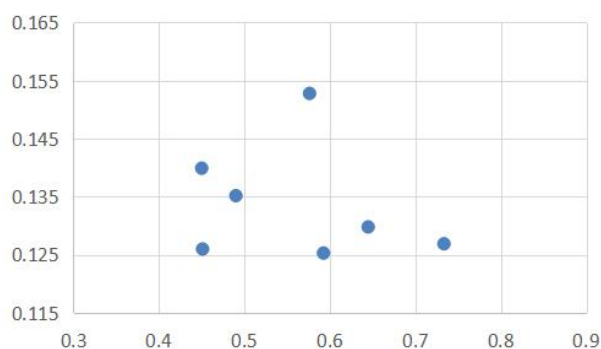
*4. In figure 7. It is hard for me to gain the point that high-Al chromitites always show higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than high-Cr chromitites, especially for Mugla Turkey and Masquad Oman.*

**Our reply:** Many thanks for the comment. Although the cases of Muğla and Masquad seem a little different in our diagram, *it was possibly because we put the data into a narrow column, making you feel that the variation trend is weak.* If we enlarge the X axials, the overall negative variation trends would be clearer. Please see the upper and lower diagrams below for the Muğla and Masquad chromitites, respectively. I think the trends would possibly be much better if there were more data for these ophiolites.

Especially, the trend for the Muğla samples indeed exists and was also mentioned by the authors themselves (*Uysal et al., 2009, CMP*). Please see the original diagrams from the paper below.

[Redacted]

*(Please note that Uysal also plotted two duplicate samples in the diagram above, so they have 6 datapoints for the high-Al chromitites)*



*5. In Figure 6. The color is not uniformed for Cpx-poor harzburgite between samples and legend.*

**Our reply:** Many thanks for the reminder, and sorry for the mistake. We have made the corrections already in the new version. Please have a check.

*Line 114: dunites and harzburgites are adjacent, but no dunite vein could be seen in Figure 4. Please give a more appropriate figure to show cutting relationship.*

**Our reply:** Many thanks for the question. It has been a common sense that dunites in ophiolitic mantle sequences were produced via melt-harzburgite reactions. The melts overall have Mg-rich and Si-undersaturated compositions, so that they can transform harzburgites into dunite by dissolving Si-saturated pyroxenes. As a result, dunites formed later than harzburgites in the mantle sequences of ophiolites.

[Redacted]

Unfortunately, I encountered heavy rain caused by a typhoon during the excursion. My sampling area that day was in a deserted open pit, with lateritic muds on outcrops everywhere. In order to better observe the feature of our samples, I had no choice but smashed the rocks to get some clean surfaces. Meanwhile, I needed to keep the clean surfaces dry at the same time, so that I could know if there were pyroxene grains in the rocks. This was important for me to tell if the rocks were harzburgites or dunites. After having the rock smashed and seeing a clear border between the dunite part and the harzburgite part. I quickly photoed what I saw and broked the sample into a smaller piece so as to take it away. You know, it was raining then, and I needed to ensure my sampling work forward. Hope you can be considerate

on this issue.



As we told in the first paragraph above, the dunite part resulted from reaction between harzburgite and Mg-rich melt. We can clearly see the compositional variation trends of chromite grains caused by the reaction, e.g., elevated Cr# and TiO<sub>2</sub> contents crossing the lithological border. Anyway, as a common sense for us working on the ophiolitic mantle sequences, I am confident that the dunite part was formed later than the harzburgite part, not to mention that it is not rare to see dunite veins in ophiolitic harzburgites. In addition, variation from the FAB to low-Ti tholeiitic (transitional) and finally to boninitic has been a well known feature for subduction initiation, and such a variation trend does not need to be verified by the information of such a sample. We just wanted to use the it to illustrate that the mantle sequence also preserve the

record of such variation trend of magmatism.

However, with the reminder of your question, and as was also suggested by other reviewers, we have moved relevant information to supplementary materials. Please see more details there. Many thanks again for the question.

*Line 180: Both Acoje and Coto blocks have high-Al chromitites, while showing different Cr# number ranges. Should you discuss their parental magmas respectively?*

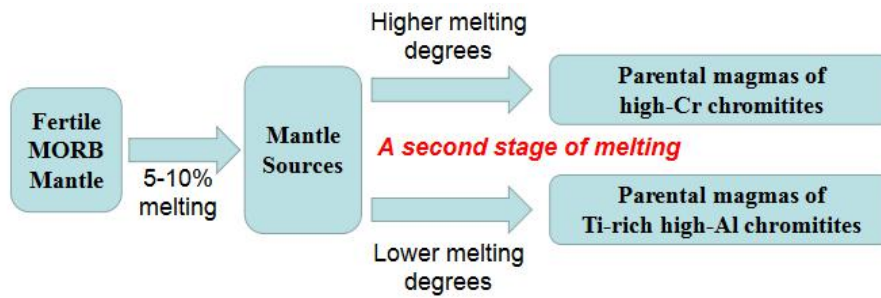
**Our reply:** Many thanks for the question. As we mentioned in our reply to your first question, the compositions postulated for the parental magmas of chromitites overall depend on the compositions of chromite in chromitites. As long as the compositions of chromite is fixed, their parental magma compositions would be fixed, no matter where the chromitites were collected from. Obviously, it is unlikely to get two sets of magma compositions if we only have one set of chromite composition. Actually, both the Cr#s and TiO<sub>2</sub> contents of chromite from the Acoje high-Al chromitites fall in the ranges of the Coto Ti-rich high-Al chromitites. Therefore, it is normal to claim that their parental magmas had similar compositions. However, their origins were different from each other based on the further discussion, which we cannot put too much information here, because the main theme of the paragraph is to recover the parental magma compositions of different chromitites (we have show in the new version how the parental magma compositions of high-Cr chromitites, Ti-rich high-Al chromitites and Ti-poor high-Al chromitites, separately). Please see our discussions in the third paragraph of the section “Origins of different types of chromite deposits”.

*Line 180: As you said above, the parental magmas of high-Al and high-Cr chromitites were formed by melting of peridotites under relatively low (S-saturated) and high (S-undersaturated) degrees, respectively. Why the parental magmas of the high-Cr chromitites and Ti-rich high-Al chromitites were generated from the similar mantle sources which had experienced ~ 5-10% melting of fertile MORB mantle (FMM)?*

**Our reply:** Many thanks for the question. Sorry for the misunderstanding caused.

Actually, the points we wanted to show here is that the parental magmas of both the high-Cr chromitites and Ti-rich high-Al chromitites share similar mantle sources from the perspective of chemical composition, which were overall equivalent to ~ 5-10% melting residues of the FMM. What we were mentioning here was the mantle sources before magmas were produced rather than the residual mantle after magmas were extracted. I think the misunderstanding was caused in this way.

Having such a fixed mantle sources first, if we want the parental magmas of high-Cr chromitites to be generated, the required partial melting degree would be high. Instead, if the aimed magmas are the parental magmas of high-Al chromitites, then lower melting degrees would be fine.



Please note that we used “had experienced” (past perfect tense) in the subordinate clause that describe the state of the “mantle sources”. This means 5-10% melting had happened first and formed the mantle sources of different magmas rather than the magmas themselves. However, we found such narrations are incorrect during the revision. This is because the mantle sources of these magmas had experienced complicated melt modification event. For example, the parental magma sources of our high-Cr chromitites had been refertilized before the magmas were generated. As a result, we think it better to say that their mantle sources had compositions equivalent to melting residues of the FMM after some partial melting degrees, rather than the melting residues exactly. Please see the revised lines shown below.

168 (Fig. 4D; Supplementary Note-2), it appears that parental magmas of the high-Cr  
 169 chromitites were generated via ~ 15-25% flux melting of moderately depleted mantle  
 170 sources that themselves had compositions equivalent to ~ 5-10% melting residues of  
 171 fertile MORB mantle (FMM) (Fig. 4D). The parental magmas of the Ti-rich high-Al  
 172 chromitites from both blocks originated from sources with similar compositions to the  
 173 high-Cr ones but were generated via further 5-15% flux melting. However, parental  
 174 magmas of the Ti-poor high-Al chromitites were formed by 5-15% flux melting of  
 175 mantle sources that had compositions equivalent to 10-20% melt residues from the  
 176 FMM (Fig. 4D). The results above indicate that the parental magmas of all our

Many thanks again for the question. Hope we clarified the issue with the explanations above.

*Line 190: the Ti-rich high-Al chromitites you discussed throughout this paragraph are only from the Acoje block, or both Acoje and Coto? If only form the Acoje, how to interpret parental magmas of Ti-rich high-Al chromitites from the Coto block?*

*Line 192: The Ti-poor high-Al chromitites were exclusively found in the Coto block, along with Ti-rich high-Al chromitites. Which process makes the variety of their Ti values?*

**Our reply:** Many thanks for the question. We found that the two questions above are mutually related. We will reply to them together. Please see our explanations below.

The main theme of this paragraph was to evaluate fertilities of the parental magma sources of different chromitites. From the perspective of compositions, the Ti-rich high-Al chromitites from both blocks are roughly similar to each other, so they were treated in the same way during the calculations and discussions. Just wondering if you were confused by the sentence that “it is hard to determine the formation sequence of the Ti-rich and Ti-poor high-Al chromitites in the Coto block”. Actually, we claimed that the Ti-rich high-Al chromitites in the Coto block formed probably earlier than the Ti-poor variety later in the L377-386 (new version). However, we could not talk too much here, because this is not the focus of this paragraph.

As we have mentioned, the Ti contents of chromite in our chromitites depend on the the fertilities of their parental magma sources. According to our calculated results, the parental magmas of Ti-rich and Ti-poor high-Al chromitites were originated from moderately depleted and highly depleted mantle sources, respectively. Depending on the formation order of the Ti-rich and Ti-poor high-Al chromitites, the stories behind will be quite different.

If the Ti-rich high-Al chromitites formed earlier than the Ti-poor ones, the variation trend would mean poorer and poorer fertility in the mantle, which can be realized with progressive melting in the mantle. By contrast, if the Ti-rich high-Al chromitites formed later than the Ti-poor ones, the variation trend would mean increasing fertility in the mantle sources, which can be achieved via refertilization. As a result, the key criterion to determine the formation order of the two types of chromitites lies in if their parental magma sources experienced refertilization event or not in the Coto block (pls note the refertilization event in the Acoje block does not count).

As we mentioned in the geologic background, the mantle sequence of the Coto block is dominated by Cpx-poor harzburgites, and no evidence supports the mantle sequence had been refertilized. This is different from the mantle sequence of the Acoje block, which has lherzolites (bottom) and Cpx-rich harzburgites (middle and upper levels) of refertilized origins. Consequently, we postulate that the Ti-rich high-Al chromitites formed earlier than the Ti-poor ones in the Coto block, because a reverse formation order would mean presence of certain amounts of Cpx-rich harzburgites and even lherzolites in the mantle sequence, just as if what we see in the Acoje mantle sequence. However, we did not see such features at all. Actually, we discussed the formation order of Ti-rich and Ti-poor high-Al chromitites already in the L371-389 (original version). Please see relevant information there. Different from the case of the Coto block, all the chromitites found in the Acoje block were formed in the context of asthenospheric upwelling, and their parental magma sources were all moderately depleted harzburgites. That is why we did not see Ti-poor chromitites in

the Acoje block.

Moreover, we have revised the sentence “it is hard to determine the formation sequence of the Ti-rich and Ti-poor high-Al chromitites in the Coto block” as “Although the formation sequence of the Ti-rich and Ti-poor high-Al chromitites in the Coto block need to be further clarified in specific geological context” (L178-180, new version), so that less confusion would be generate. Please see more details in our revised version of that paragraph. Many thanks again for the question.

178 Ti-poor high-Al chromitites. Although the formation sequence of Ti-rich and Ti-poor  
179 high-Al chromitites in the Coto block need to be further clarified in specific  
180 geological context, the Zambales proto-forearc mantle had surely become increasingly  
181 fertile as the mineralization proceeded from the Ti-poor high-Al chromitites in the  
182 early Coto block to the Ti-rich high-Al chromitites and high-Cr chromitites in the  
183 younger Acoje block, regardless if the Ti-rich high-Al chromitites formed earlier or  
184 than the Ti-poor ones in Coto block. Such a result differs from the conventional view

377 According to melting-related calculations, the parental magma sources of the Ti-rich  
378 high-Al chromitites and Ti-poor ones, had moderately depleted and highly depleted  
379 compositions, respectively (Fig. 4D). Since no evidence supports that the mantle  
380 sequence of the Coto block had been refertilized, the Ti-rich high-Al chromitites in

381 the Coto block must have formed earlier than the Ti-poor ones. To use a  
382 reversal formation order of the two types of high-Al chromitites would require similar  
383 refertilization process in the Coto block as in the Acoje block, producing new  
384 Cpx-rich harzburgites and restoring Ti content in the Coto mantle sequence. As such,  
385 variation of high-Al chromitites from the Ti-rich to Ti-poor varieties in the Coto block  
386 probably reflects increasingly depleted magma sources (S2-S3 in Fig. 4D). Because

*Line195: The tradition view is that the Cr# numbers could indicate the melting degrees of mantle sources. While it seems that Ti is crucial when evaluating the melting degree in your discussion. So how to balance the two indicators?*

**Our reply:** Many thanks for the question. Yes, we quite agree that both Cr# and TiO<sub>2</sub> contents of chromite can be used for evaluating partial melting degrees in the mantle, but the way how they can be applied are very different. Please see our explanations below step by step.

**When can the Cr# of chromite be used for the evaluation?** I have no problem with that Cr# of chromite can be used as an indicator of melting degrees, but **the precondition is that the chromite (Cr-spinel) used in relevant discussions should be those hosted in mantle lherzolites or harzburgites of strictly melting residual origins.** If the peridotites had once been modified by melts, the Cr#s of chromite will also be changed and not suitable to be as the indicator of melting degrees anymore.

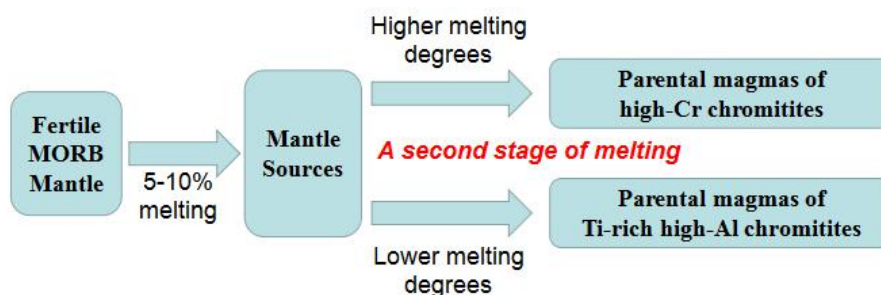
**Why didn't we use Cr#s of chromite for the evaluation?** This is because the chromite grains used in the discussions were from chromitites, which were cumulate products of their parental magmas. It is highly noted that chromite residues left in the magma sources would have different Cr#s and compositions from those newly crystallized ones from the magmas, even if both of them were chemically equilibrated with the same magmas. This is because the P-T-fO<sub>2</sub> parameters all changed as the magmas migrated upward to the shallow levels, and such changes would result in chromite with different compositions. Generally, newly crystallized chromite have higher Cr#s than the residual ones. Consequently, the Cr#s of chromite in chromitites and other cumulate rocks can only be partly used for evaluating the melting degrees of their parental magma sources, comparatively but not strictly. So from our viewpoint, never use the Cr# of chromite from chromitites (and dunites) to judge the partial melting degrees required for their formation. Even though some values can be obtained, the values can only be taken for references but are actually not that meaningful at all.

**Why did we use Ti for the evaluation?** Different from other multi-valence, its valence state in chromite and most mineral phase are stable. For one thing, the TiO<sub>2</sub> contents of chromite have been used for long to constrain the compositions of their parental magmas. For another, the parameters of Ti in mantle rocks (e.g., concentrations and partition coefficients) have been well obtained, so that we can use melting models to confine the TiO<sub>2</sub> contents partitioned into the generated magmas under conditions of different mantle sources and partial melting degrees. Accordingly, comparisons can be made between the calculated TiO<sub>2</sub> contents of magmas via the two methods above, in order to constrain relevant melting processes and compositions of their mantle sources. This is how we did in our study.

To be concluded, regarding on how to evaluate partial melting degrees, if the selected indicator is the Cr#s of chromite, the studied objects should be the host peridotites of the chromite, and the peridotites themselves should have residual origins rather than being cumulate products or those obviously modified by melts (e.g.,



highly refractory harzburgites and dunites). However, if the studied objects are chromitites, please remember never to say how many degrees of melting they underwent, because they are cumulate products from magmas rather than residual ones of partial melting. What we can do is to evaluate two sets of partial melting degrees for their parental magma sources from two perspectives: (1) how many degrees of partial melting did the mantle sources had experienced before the parental magmas of the cumulates were generated (a first stage of melting; e.g., 5-10% in the below diagram); (2) How many degrees of melting is further needed for the mantle sources to generate the parental magmas of the cumulates (a second stage of melting).

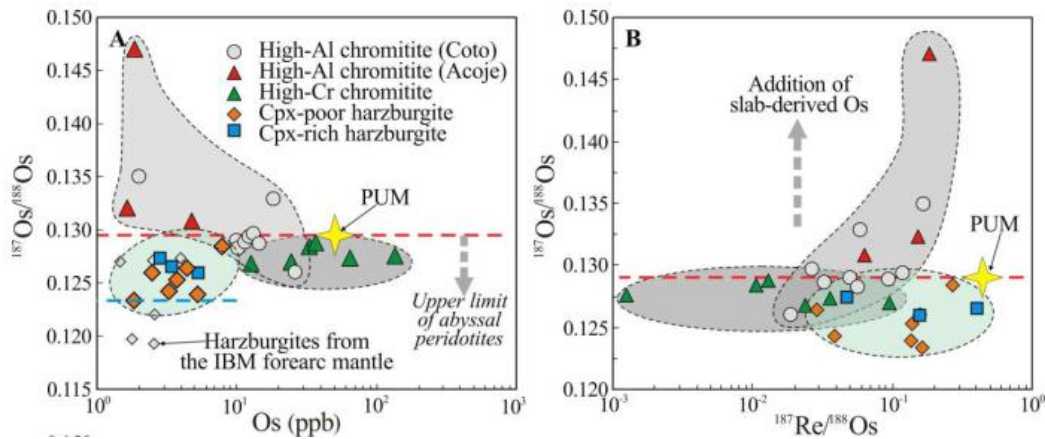


All in all, what we wanted to say above is that the  $\text{TiO}_2$  contents of chromite from chromitites are more useful than their Cr#s for constraining the states of their parental magma sources and the required degrees of partial melting. Hope you are fine with our explanation above.

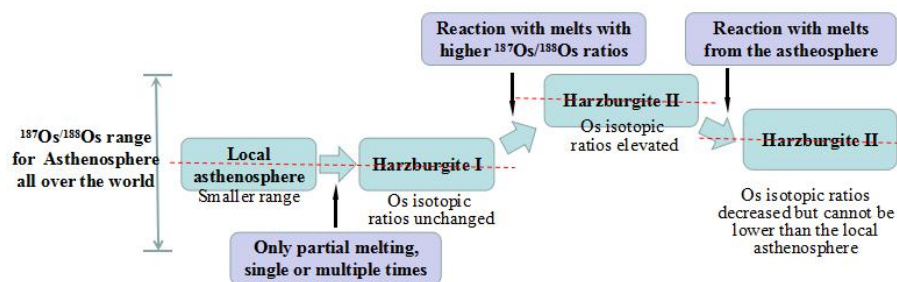
*Line 256: Your point of view here is just suitable for the case of the Acoje block, where high-Al chromitites have more Os isotopic ratios than that of high-Cr chromitites, but not the case of the Coto block where most high-Al chromitites show Os isotopic ratios comparable to abyssal peridotites, not elevated.*

**Our reply:** Many thanks for the question. I can understand your point here very well. Although most of the high-Al chromitites have Os isotopic ratios falling in the range of abyssal peridotites, we can still find that the Coto high-Al chromitites overall have identifiably higher Os isotopic ratios than the high-Cr chromitites from the Acoje block, not to mention that several samples do have  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios beyond the range of abyssal peridotites.

Moreover, we want to emphasize that having abyssal-like Os isotopic compositions does not necessarily mean there is no slab components in the studied mantle. Please see our explanation below. As we mentioned in the main text (L295-300, new version; L306-311 before the revision) and our reply to the Q2.1.3 and Q2.5.1 (Question of the 2<sup>nd</sup> reviewer; we would like to repeat the reply here for your convenience), the mantle sources of all our chromitites, including both the high-Al and high-Cr ones, had been modified by slab fluids, even though their  $^{187}\text{Os}/^{188}\text{Os}$  ratios fall in the range of abyssal peridotites. Such a conclusion was obtained based on comparison with the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of harzburgites analyzed in our study.



As we discussed in the L275-300 (new revision), the origins of harzburgites in ophiolitic mantle sequences can be initially traced back to decompressional melting of asthenosphere, despite their melt modification histories. Bearing in mind that the process of partial melting alone cannot fractionate Os isotopes, harzburgites from the mantle sequence of some an ophiolites should have indistinguishable Os isotopic ratios to the local asthenosphere, as long as only partial melting (single or multiple times) happened to the harzburgites. However, the fact is that harzburgites in the mantle sequences of ophiolites had been modified by parental magmas of chromitites, all of which had higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the harzburgites, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the photo-liths of the harzburgites should be even lower than those of the harzburgites. Specific to the case of the Zambales ophiolite, the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the local asthenosphere below the Zambales proto-forearc before the formation of chromitites could not be higher than those of the harzburgites we analyzed, all below 0.1275 and smaller than the ratios of all the Zambales chromitites. Consequently, the mantle sources of all chromitites must have contributions of slab fluids, even though their  $^{187}\text{Os}/^{188}\text{Os}$  ratios fall into the range of abyssal peridotites. Please see how the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of the harzburgites changed during different processes in the flowchart below (note the relative positions of the dashed red lines).



To put it simple, the range of abyssal-like  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios was established based on a large dataset from the global, but the  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratio of asthenosphere below some a place unlikely have such large variation but more likely fall in a narrow range of its own. To judge if there are slab contributions in the magma sources, we need to compare their Os isotopic ratios with their associated harzburgites rather than simply with the range of the abyssal peridotites.

*Line 259: in figure 6A, the highest Os isotopic ratios are from high-Al chromitites in*

*the Acoje block, not the Coto block. Does it mean intensified slab dehydration during subduction initiation?*

**Our reply:** Many thanks for the question. To be realistic, the elevated  $^{187}\text{Os}/^{188}\text{Os}$  ratios would definitely mean addition of slab components in the parental magmas, but it is not easy to constrain the comparative intensities of slab dehydration during the two stages of chromite mineralization you mentioned above. Based on our data, **it is more appropriate to say that the parental magmas of the high-Al chromitites, both the Ti-rich and Ti-poor varieties, were overall more obviously modified by the slab fluids than those of the high-Cr chromitites**, but it is difficult to tell which period of high-Al chromite mineralization was featured by more intensive slab dehydration, not to mention that the  $^{187}\text{Os}/^{188}\text{Os}$  ratios of high-Al chromitites from the two blocks largely overlap each other. Please see more explanation below.

Interaction between the proto-forearc mantle and subsiding slabs during subduction initiation is complicated, and the compositions of slab-released fluids that were added into the proto-forearc mantle would vary from time to time. Firstly, even for the same piece of slab, different batches of fluids released from it had different compositions, elemental and isotopically (especially under varying temperature conditions). Secondly, slab rollback is the fundamental driving force for development of all the geological processes during subduction initiation. With slab rollback going on, the proto-forearc spreading center moves toward the trend. Specifically in our study, the spreading center moved from the Coto region to the Acoje region, so the dehydrating part of the rollback slab below the Coto proto-forearc spreading center should be different from that below the Acoje spreading center. Consequently, **the fluids added into the Acoje proto-forearc mantle probably had different Os isotopic compositions from those added into the Coto proto-forearc mantle, so it is hard to claim which period was featured by more intensive dehydration exactly.**

Considering the information above, we would like to re-state that the largely overlapped  $^{187}\text{Os}/^{188}\text{Os}$  ratios of high-Al chromitites from the two blocks mean their parental magma sources were overall modified to similar degrees, although some abnormal cases would exist. Even so, we prefer not to deliberately over-read some an abnormal data too much, which would generate unrealistic conclusion in many cases. Based on our model, it is quite possible that formation of the Acoje Ti-rich high-Al chromitites happened in a more intensive period of slab dehydration, as you mentioned, because the geothermal gradient has been elevated slightly in the context of asthenospheric upwelling. However, our data do not unequivocally support this postulation, and **it is more cautious to say that formation of high-Al chromitites overall mark a climax period of slab impact in the proto-forearc mantle compared with the period of high-Cr chromite mineralization (and also the MORB-like FAB stage).** Such a conclusion is a progress for subduction initiation-related studies.

Many thanks again for your question. We also want to emphasize here again that it is more appropriate to say that the parental magmas of the high-Al chromitites, both the Ti-rich and Ti-poor varieties, were overall modified more obviously by the slab fluids than those of the high-Cr chromitites, but **it is difficult to tell which period of**

high-Al chromite mineralization was featured by more intensive slab dehydration. Moreover, we would like to re-state that it is better to consider the data of each type of sample as a whole and interpret them from the average perspective, but never over-read each data too much. This is because the physico-chemical conditions in the parental magma sources of each chromitites may be different, **it would be misleading by simply assuming all of them were genetically related and that their parental magma sources the share same physico-chemical** (e.g., fertility, temperature, fluid contents,  $fO_2$ ) conditions. So we need to allow presence of differences in each type of samples. Our conclusions are better to be reached based on the overall variation trend, in a more cautious way. Hope you would be fine with our explanation above.

*Line 279: Both high-Al chromitites and high-Cr ones in the Coto bock were altered by asthenospheric melts. Which the former has elevated  $^{187}Os/^{188}Os$  ratios?*

**Our reply:** Many thanks for the question. Just want to confirm with you first if you were referring to the chromitite from the Acoje block, since we do not have high-Cr chromitites in the Coto block.

As we mentioned in the L375-376 and L405-407 (new version), asthenospheric upwelling happened before both types of chromite mineralization in the Acoje block (Ti-rich high-Al and high-Cr types), which can be evidenced by both geological observation and the calculated results of fertility for their mantle sources (evaluated via  $TiO_2$  in the mantle). Since high-Cr chromitites were formed later than high-Al ones, the proto-forearc mantle would theoretically be more sufficiently modified by asthenospheric components during the mineralization stage of high-Cr chromite than that of high-Al chromite. Such a process is easy to understand. The longer the modification was, the more the effects of slab fluids in the proto-forearc mantle would be diluted. That is why we told “the effects of slab components on the proto-forearc mantle remained very strong during the early stage of asthenosphere”.

In order to make the sentence better understood, we have revised the sentence as “**the impact of slab components on the proto-forearc mantle remained very strong and had not yet been largely diluted by the replenished asthenospheric components during the early stage of asthenospheric re-upwelling**”. Please have more relevant details in the last two portions of the main text.

407 radiogenic Os isotopic compositions than the high-Cr chromitites. This suggests that  
408 the impact of slab components on the proto-forearc mantle remained very strong and  
409 had not yet been largely diluted by the replenished asthenospheric components during  
410 the early stage of asthenospheric re-upwelling. Further given minimal presence of slab  
411 fluids in the FAB sources<sup>6,9</sup>, the slab impact in the proto-forearc mantle was probably  
412 the most noticeable during the stage of high-Al chromite mineralization. In spite of

*Line 369: Most high-Al chromitites in the Coto block show  $^{187}\text{Os}/^{188}\text{Os}$  ratios equal to abyssal peridotites, while only few ones have more radiogenic Os isotopes. How to explain the selectivity?*

**Our reply:** Many thanks for the question. As we explained to the Q.L256 and the comments to the second reviewer, having abyssal-like Os isotopic compositions does not necessarily mean there is no slab components in the studied mantle. The range of abyssal-like  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratios was established based on a large dataset from the global, but the  $^{187}\text{Os}/^{188}\text{Os}$  isotopic ratio of asthenosphere below some a place unlikely have such large variation but more likely fall in a range of its own. **To judge if there are slab contributions in the magma sources, we need to compare their Os isotopic ratios with their associated harzburgites rather than simply with the range of the abyssal peridotites.** Please see more details to our reply to the Q.L256, and hope you are fine with our explanation here.

*Line 381: “the Ti-rich high-Al chromitites must have formed before the Ti-poor ones.” Which block are these high-Al chromitites from, Acoje or Coto? Please make it clear.*

**Our reply:** Many thanks for the reminder. The high-Al chromitites mentioned in this sentence refer to those from the Coto block, which have both Ti-rich and Ti-poor high-Al chromitites at the same time. After reading your question, we also realize that we did not make it clear here. Sorry for the confusion caused. In the new version of our manuscript, we have revised the sentence as “the Ti-rich high-Al chromitites in the Coto block must have formed before the Ti-poor ones” and added more details to support the statement.

377 According to melting-related calculations, the parental magma sources of the Ti-rich  
378 high-Al chromitites and Ti-poor ones, had moderately depleted and highly depleted  
379 compositions, respectively (Fig. 4D). Since no evidence supports that the mantle  
380 sequence of the Coto block had been refertilized, the Ti-rich high-Al chromitites in  
381 the Coto block must have formed earlier than the Ti-poor ones. This is because a  
382 reversal formation order of the two types of high-Al chromitites would require similar  
383 refertilization process in the Coto block as in the Acoje block, producing new

*Line 404: As showed in figure 6, high-Al chromitites from the Acoje block have more elevated  $^{187}\text{Os}/^{188}\text{Os}$  ratios than these from the Coto block. The effects of slab components on the proto-forearc mantle should be stronger on Coto than Acoje, since the former was altered earlier. How to explain?*

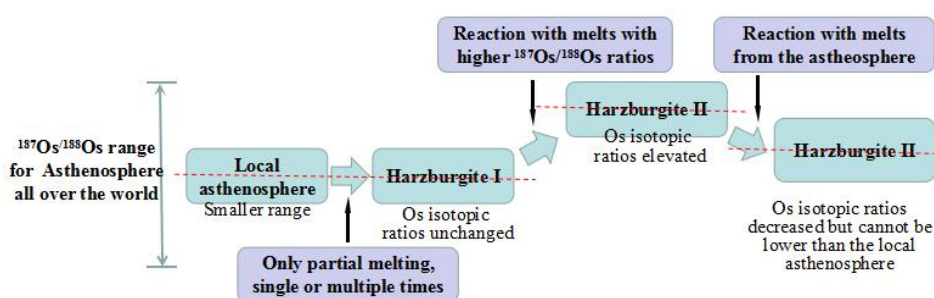
**Our reply:** Many thanks for the question. As we replied to Q.L259, the  $^{187}\text{Os}/^{188}\text{Os}$

ratios of high-Al chromitites from the two blocks largely overlapped, indicative of similar slab impact in most of their parental magma sources, although some extraordinary cases existed. Based on the data of our own and compiled ones, what we can claim for sure is that the parental magma sources of the high-Al chromitites, both the Ti-rich and Ti-poor varieties, were overall more obviously modified by the slab fluids than those of the later high-Cr chromitites, and presence of such slab components was critical for producing the tholeiitic-like magmas from depleted mantle sources under relatively low temperature conditions (pls see our explanation in L358-363). It is difficult to tell which period of high-Al chromite mineralization was featured by more intensive slab dehydration. Anyway, we prefer to emphasize the overall variation trend in our studies, which is more reliable than some one or two abnormalities. Otherwise, we may reach some misleading conclusions. Hope you would be fine with our explanation here. Many thanks.

*Line 414: More radiogenic Os isotopic compositions were brought by slab components, since upwelling of asthenosphere could not bring supra-chondritic Os isotopic ratios. (See L-244)*

**Our reply:** Many thanks for the comment. Actually, **what we explained here is the same to your point above, so we are confused where the problem is**, or is there any misunderstanding caused here?

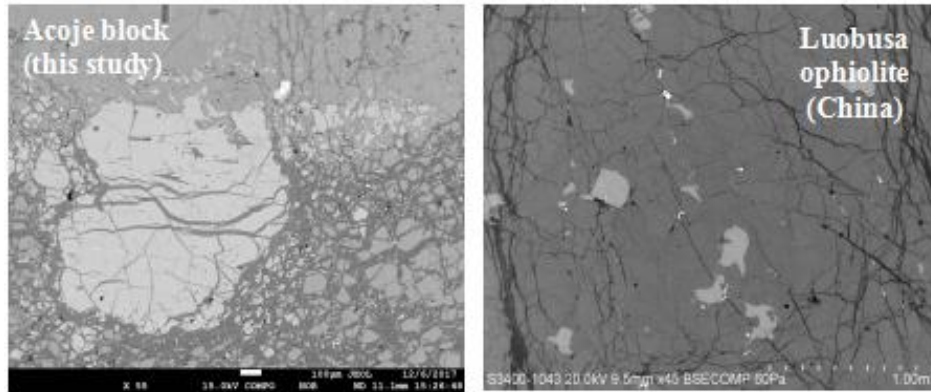
As we repeated in the main text, there is generally a common variation trend that high-Al chromitites have higher  $^{187}\text{Os}/^{188}\text{Os}$  ratios than the high-Cr ones in ophiolites that meanwhile host high-Al and high-Cr chromitites, indicating less slab impact in the parental magma sources of high-Cr chromitites than in those of the high-Al chromitites. Since addition of asthenospheric components was the only way to draw the Os isotopic ratios back to lower levels in the context of subduction initiation, that is why we claim that “upwelling of asthenosphere happened in the proto-forearc before the high-Cr chromite mineralization.



Moreover, the conclusion above obtained based on Os isotopes can also be supported by petrographic and compositional features. This is because **addition of asthenospheric melts would also cause refertilization and replenishment of fusible elements (e.g., Ca and Ti) in the modified lithospheric mantle.**

On one hand, there are Cpx-rich harzburgites (+lherzolites) in the Acoje mantle sequence. These Cpx-rich harzburgites make up the host rocks of chromitites and dunites in the mantle sequence, so they were formed before the chromitites (please see more details about their genesis in our answers to Q2.3). Specially, **clinopyroxene**

grains in the Cpx-rich harzburgites mostly have finger-like protrusions (look like octopus with contractile arms) that stretched into the interstitial spaces. Such textures support the outward growth history of the clinopyroxene with aid of interstitial melts. Moreover, these clinopyroxene, especially those in the Cpx-rich harzburgites from bottom part of the mantle sequence have comparable compositions (Ca, Al, Ti and REE) to those in the abyssal peridotites, indicating the asthenospheric affinities of the refertilizing melts (pls see more details from the two classical cases below Fig. 2.2.1).



[Redacted]

*Textural and compositional features of cpx grains from cpx-rich harzburgites. The two BSE images show the irregular outlines of cpx and their alignment in the rocks, supporting their out-ward growth histories from interstitial melts. The four data diagrams below show the compositions of cpx from cpx-rich harzburgites (S-1,S-2,S-3,S-5). It can be seen that cpx from cpx-rich harzburgites have compositions generally comparable to the abyssal peridotites. In case that they were notably modified, their compositions would move to more depleted directions (lower Al and HREE).*

On the other, the Ti contents of chromite in high-Cr chromitites are overall no lower than those of chromite in the high-Al chromitites, reflecting that the parental magma sources of the high-Cr chromitites were overall less depleted than those of the high-Al chromitites (to be exactly, comparable to those of Ti-rich high-Al chromitite, but more fertile than those of Ti-poor high-Al chromitites). Moreover, addition of asthenospheric melts into the overlying lithospheric mantle means active asthenospheric upwelling, which will bring about elevated geothermal gradients (Please please see more details in our answers to Q2.4). This would facilitate higher degrees of melting and generate boninitic-like magmas.

Again, our content in the L414 is agreement with what you told above. Many thanks for your point here.

*Line 416: The re-upwelling process of asthenosphere not only replenished fertile*

*components (e.g., Ca, Al and Ti) (Line 397-398)...since that, why the fertilized mantle could produce high-Cr parental magmas while depleted mantle could not?*

**Our reply:** Many thanks for the question. Sorry for any misunderstanding caused.

From our viewpoints, both moderately depleted and highly depleted mantle sources can generate boninitic magmas, and both of them can also produce less magnesian tholeiitic magmas. This is a matter of melting degree, and the controlling factor on this issue (in our study) lies in the geothermal gradient during the partial melting. If the geothermal gradient is high enough, the magmas are boninitic or arc picritic ones. Otherwise, only tholeiitic magmas can be produced, just like the case of the parental magmas of high-Al chromitites. Apparently, highly depleted sources will generate more Mg-rich and Cr-rich magmas than moderately depleted ones under the same partial melting degrees.

Moreover, we would like to re-state that, despite the refertilization process in the mantle below the Acoje proto-forearc spreading center, the lithospheric mantle was not converted back to fertile MORB mantle (fertile lherzolites) at all, quite different from the case that happened below the North China Craton. This is because the spreading rate was not that developed as below normal mid-ocean ridges.

Many thanks again for the question, and hope we have clarified this issue with the explanation above.

417 In spite of the refertilization event, it is highly noted that the parental magmas of all

418 the Acoje chromitites were originated from more depleted mantle sources than the

419 FMM (Fig. 4D). This suggests that the fertility of the proto-forearc mantle was not

420 recovered to the level of FMM anymore after the MORB-like FAB magmatism.

421 Accordingly, the extent of asthenospheric upwelling and rate of slab rollback during

422 high-Cr chromite mineralization were not as remarkable as those during the FAB

423 magmatism, even though both processes were accelerated after the period of Ti-poor

424 high-Cr chromite mineralization.



## REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

I have reviewed the revised version of the manuscript submitted by Zhang et al., which is a substantial improvement over the original submission. The authors have diligently addressed the concerns raised in the previous review, and their efforts are commendable. The detailed "response to reviewers' comments" letter provided by the authors showcases their commitment to enhancing the quality of their work.

**Improvements in Clarity and Structure:** The revised manuscript demonstrates significant improvements in clarity and organization compared to the previous version. The authors have restructured the content to enhance readability, making it easier for readers to follow the flow of the argument.

**Addressing Reviewers' Comments:** I appreciate the authors' thorough responses to the reviewers' comments. They have addressed each concern raised in the previous review comprehensively, providing explanations and revisions where necessary. This demonstrates their dedication to refining their research based on constructive feedback.

**Minor Points:**

There are only a few minor corrections that I would suggest, primarily related to grammar and formatting. These can easily be addressed during the proofreading stage before final acceptance. Here are minor suggestions:

- 1- Line 97-10: I would suggest rephrasing the statement to make it more clear. They mean probably "by interaction of Mg-rich melt".
- 2- Line 247, 352: I would delete "regime".
- 3- L353: Please replace "inclined to produce" with "prone to yield"?
- 4- Fig. 2 caption: I would replace "diagram" with "panel". Also, L25; add space before "(major)".
- 5- Fig. 4 caption (L81): udmt to udtm
- 6- Fig. 8: Please show LAB on panel B.

**Recommendation:**

Based on the substantial improvements made in the revised manuscript and the authors' responsiveness to reviewers' feedback, I recommend accepting the paper for publication in "Nature Communications". The paper is well-written, technically sound, and relevant to the scope of the journal.

Kind regards,

Reviewer #2 (Remarks to the Author):

The authors have made substantial progress in addressing the concerns raised in the initial reviews, answering exhaustively every point and making the appropriate changes in the manuscript. However, I would have appreciated that the answer would be clearer and more concise, avoiding repetition, and eliminating irrelevant sub-sections and figures. Reviewing such an extensive response is extremely time-consuming. There are also some points that are not completely resolved, but their interpretations are generally supported by the presented data. Find below some detailed comments:

- One of the main issues of the first manuscript was the relation of the high-Al chromitites with FAB. However, they have strongly clarified that, in this case, the studied high-Al chromitites are related to transitional tholeiitic melts. However, I would like to remark that the mantle that undergoes melting in the fore-arc region is already depleted, hence, it should not be considered a fertile mantle, but a depleted mantle (see Workman & Hart, 2005). This should be addressed in the manuscript.
- Their model does not explain how from the different lithologies they can form the chromitites. But still this is a major issue that cannot be resolved from the mass balance point of view.
- The description of the lithologies has improved. However, they show evidence from other works and other ophiolites to support their Cpx-poor Hz1, Cpx-rich Hz2, and Cpx-poor Hz3 lithologies. It would be more interesting for the work that they show these differences in the samples that they have studied. In the letter they give a lot of information regarding this topic, but they use images from other ophiolites and it is not clear if this differentiation arises from their observations or not.
- Regarding the temperature of transitional magmas vs boninitic magmas, the figure they are including from Grove and Parman (2004) would not apply, since it does not considerate fore-arc basalts (it is previous to its description) or transitional magmas. Besides, the water content in boninites is a feature that needs consideration. Besides, the authors should provide appropriate references to support that boninites forming high-Cr chromitites had higher temperatures than the transitional magmas forming high-Al chromitites. There are many statements in the letter that are not supported by appropriate references.

Overall, the manuscript shows marked improvement and comprehensively addresses many of the previous comments. I recommend the acceptance of the manuscript for publication after minor revision.

Reviewer #3 (Remarks to the Author):

I am very satisfied with the revisions to the manuscript and the author's answers to the questions raised, and do not have any further suggestions for revisions.

## **Replies To NCOMMS-23-40610A**

### **Reviewer #1 (Remarks to the Author):**

*I have reviewed the revised version of the manuscript submitted by Zhang et al., which is a substantial improvement over the original submission. The authors have diligently addressed the concerns raised in the previous review, and their efforts are commendable. The detailed "response to reviewers' comments" letter provided by the authors showcases their commitment to enhancing the quality of their work.*

*Improvements in Clarity and Structure: The revised manuscript demonstrates significant improvements in clarity and organization compared to the previous version. The authors have restructured the content to enhance readability, making it easier for readers to follow the flow of the argument.*

*Addressing Reviewers' Comments: I appreciate the authors' thorough responses to the reviewers' comments. They have addressed each concern raised in the previous review comprehensively, providing explanations and revisions where necessary. This demonstrates their dedication to refining their research based on constructive feedback.*

**Our reply:** Many thanks for reviewing the paper again. The following suggestions and instructions are really great for improving the contents of relevant lines in the main text. We have made revisions accordingly. Please check them below and in the revised main text.

#### **Minor Points:**

*There are only a few minor corrections that I would suggest, primarily related to grammar and formatting. These can easily be addressed during the proofreading stage before final acceptance. Here are minor suggestions:*

*1- Line 97-10: I would suggest rephrasing the statement to make it more clear. They mean probably "by interaction of Mg-rich melt".*

**Our reply:** Many thanks. Based on your suggestions and the overall meaning here, we changed the sentence as "modified from less depleted peridotites by interaction with Mg-rich melts". Please have a check here.

*2- Line 247, 352: I would delete "regime".*

**Our reply:** Many thanks for the suggestion. We quite agree with you here and have deleted the words already.

*3- L353: Please replace "inclined to produce" with "prone to yield"?*

**Our reply:** Many thanks. Your suggested phrases are much better than ours. We have made the revision accordingly.

*4- Fig. 2 caption: I would replace "diagram" with "panel". Also, L25; add space before "(major)".*

**Our reply:** Many thanks. We quite agree with you on this point, and sorry for the mistake. We have made the revisions accordingly.

*5- Fig. 4 caption (L81): udm to udtm*

**Our reply:** Many thanks for pointing the mistake. Sorry for that. We have made the

revision accordingly.

*6- Fig. 8: Please show LAB on panel B.*

**Our reply:** Many thanks for the instruction. Lithosphere (L) and asthenosphere (A) in the diagrams were shown in yellow and green, respectively. The four cartoons are full of explanations, and it is not suitable to add more notations anymore. Based on your suggestion, we have explained in the panel B where the LAB (the boundary between the green and yellow parts) is . Please see the revision there.

*Recommendation:*

*Based on the substantial improvements made in the revised manuscript and the authors' responsiveness to reviewers' feedback, I recommend accepting the paper for publication in "Nature Communications". The paper is well-written, technically sound, and relevant to the scope of the journal.*

*Kind regards,*

#### **Reviewer #2 (Remarks to the Author):**

*The authors have made substantial progress in addressing the concerns raised in the initial reviews, answering exhaustively every point and making the appropriate changes in the manuscript. However, I would have appreciated that the answer would be clearer and more concise, avoiding repetition, and eliminating irrelevant sub-sections and figures. Reviewing such an extensive response is extremely time-consuming. There are also some points that are not completely resolved, but their interpretations are generally supported by the presented data. Find below some detailed comments:*

**Our reply:** Many thanks for your understanding and reviewing the paper again. We really hoped to clarify each point mentioned by you last time. Sorry that it took you long time to finish reading the replies. Your kindness and consideration are highly appreciated. Also, we found the following suggestions and instructions are great. They are really helpful for further improving the contents of the paper. We have replied to all them one by one and made revisions accordingly. Please check them below and in the revised main text.

*Q1. One of the main issues of the first manuscript was the relation of the high-Al chromitites with FAB. However, they have strongly clarified that, in this case, the studied high-Al chromitites are related to transitional tholeiitic melts. However, I would like to remark that the mantle that undergoes melting in the fore-arc region is already depleted, hence, it should not be considered a fertile mantle, but a depleted mantle (see Workman & Hart, 2005). This should be addressed in the manuscript.*

**Our reply:** Many thanks for the comments. We quite agree with you that all the upper mantle had undergone partial melting already, even for the mantle sources of MORB. Sorry that we did not make relevant points clear enough. [What we wanted to tell was that the mantle sources of transitional tholeiitic magmas \(10-20% depletion of the](#)

[FMM](#)) were more depleted than MORB sources (2-3% depletion of the PUM, as told [in the recommended paper](#)). Please see our explanations step by step.

### *1.1 What is the difference between DMM and FMM? (generally, the same item)*

MORB were originated from asthenosphere, and asthenosphere is MORB source mantle. Asthenosphere is the residue of the primitive upper mantle (PUM) after melt extraction during crust-mantle separation (2-3% partial melting residues, pls also see in your recommended paper), so it is (slightly) depleted in incompatible elements and radiogenic isotopes. However, 2-3% melting of the PUM could not change the major (and minor) element compositions of the mantle too much, so asthenospheric mantle overall has similar major elemental compositions to the PUM, and it is still dominated by fertile lherzolites. As a result, [asthenosphere has dual geochemical characteristics, fertile in major elements but meanwhile isotopically depleted](#). It is highly noted that the [antonyms of “depleted” and “fertile” are “enriched” and “refractory”, respectively](#), but many people mixed them together nowadays. Anyway, a depleted mantle can be fertile at the same time. Please see the reference shown below.

[Redacted]

In literature, both DMM and FMM can be used to describe the MORB source mantle (or asthenosphere). In particular, [DMM was named from the perspective of isotopes \(and also incompatible trace elements\)](#), referring to the more depleted radiogenic isotopic features (including Sr, Nd, Hf, Os and etc.) than the PUM. By contrast, [FMM was named from the perspective of major \(and minor\) element compositions that can affect magma production](#). As a result, people use DMM when they are talking about isotopic issues, but FMM is used when they are talking about elemental issues, especially for discussions of partial melting.

### *1.2 Was MORB-like magmatism developed in the Zambales ophiolite? (Yes)*

Previous studies have proved that MORB-like FAB products were indeed found in the Coto block of the ophiolite, and such MORB-like FAB magmatism varied into IAT-like (in both the Coto and Acoje blocks) and boninitic ones (in the Acoje block only) with time, well comparable to the magmatism sequence developed in the IBM forearc system. This is a well-recognized reality and has been discovered for long, e.g., by Geary et al. (1989) and Perez et al. (2018). Julian Pearce mentioned in his paper (1992) that the Acoje block of the ophiolite can be the best analogy for the Izu-Bonin outer-arc high, while the Coto block is a possible candidate for crust created just

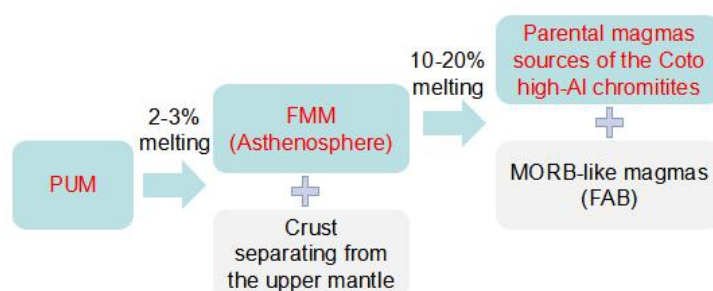
before or at the start of subduction. As a result, the evolutionary history of the Zambales ophiolite makes it one of the most classical cases of subduction initiation, and fertile MORB mantle (FMM) or mantle sources of similar fertility indeed existed below the Zambales proto-forearc during the earliest stage of subduction initiation, before the transitional magmatism occurred.

Geary, E.E., Kay, R.W., Reynolds, J.C. & Kay, S.M. *Geochemistry of mafic rocks from the Coto Block, Zambales ophiolite, Philippines: trace element evidence for two stages of crustal growth. Tectonophysics* **168**, 43-63 (1989).

Pearce, J.A. et al. *Boninite and harzburgite from Leg 125 (Bonin-Mariana forearc): A case study of magma genesis during the initial stages of subduction. In Proceedings of the ocean drilling program, Scientific Results 125*, 623-659 (1992).

Perez, A., Umino, S., Yumul Jr, G.P. & Ishizuka, O. *Boninite and boninite-series volcanics in northern Zambales ophiolite: doubly vergent subduction initiation along Philippine Sea plate margins. Solid Earth* **9**, 713-733 (2018).

Further combined with the calculated results that the parental magma sources of the Coto high-Al chromitites have equivalent compositions to 10-20 partial melting residues of the FMM, the Zambales proto-forearc mantle should be more fertile than the parental magma sources of high-Al chromitites at the starting period of subduction initiation. The former is equivalent to 2-3% (or so) melting residues of the PUM, the latter equivalent to 10-20% melting residues of FMM or 12-22% melting residues of PUM.



In order to better emphasize the idea that the parental magma sources of high-Al chromitites (10-20 depletion of the FMM) were more depleted than the sources of MORB (FMM, asthenospheric mantle, 2-3% depletion of the PUM), we added more information in the L338-343 (new version). Please see the changes as below. Also many thanks again for the comment and instruction on this issue.

337 **Origins of different types of chromite deposits**

338 Our results show that the parental magmas of high-Al chromitites in the Coto  
 339 block originated from mantle sources that were chemically equivalent to ~ 10-20%  
 340 melting residues of the FMM. This earlier 10-20% melting event was possibly linked  
 341 to extraction of MORB-like FAB magmas from the FMM (asthenosphere) below the  
 342 Zambales proto-forearc, which happened at the beginning of subduction initiation in  
 343 the region (S1 in Fig. 4D; Fig. 8A), present as lavas and dykes in the Coto block<sup>15</sup>.

*Q2. Their model does not explain how from the different lithologies they can form the chromitites. But still this is a major issue that cannot be resolved from the mass balance point of view.*

**Our reply:** Many thanks for the comment. We can understand your concern very well, and sorry that we did not make this point clear. The mechanism of Cr concentration from magmas is another issue. Actually, the third reviewer asked this question last time. We can make some simple explanations below.

Chromium (Cr) is only a minor element in Mg-rich magmas, but it is the major element in chromite ores. **To generate one portion of chromitite, tens to hundreds times of volumes of magmas were needed**, depending on the grades of the orerocks. This means that large amounts of magmas had passed through the melt channels or magma chambers already before a tiny piece of chromitite was finally generated. **This is why we said that chromitites were generated in melt-dominated environments**. Such a mass balance-based point is easy to understand and very clear for researchers on chromitites. Our research group has studied ophiolitic chromitites for 30 years, so we are quite confident on this issue. We have taken these above information for granted. Sorry that we did not realized it is necessary to make some explanations in our paper.

Due to the length limitation required by the journal, we cannot provide too much details on the topic you mentioned above. Moreover, the main focus of this paper is to clarify how, when and why different ore-forming magmas were generated during the evolution course of subduction initiation. In order to let future readers know that formation of chromitites need large volumes of magmas as you reminded, we made the following revisions accordingly. Many thanks again for your kind reminder and instruction here. Hope you would be fine with our explanations and revision.

114 **Mantle fertility during different periods of chromite mineralization**

115 Mantle-derived magmas generally have < 4000 ppm Cr, but the Cr<sub>2</sub>O<sub>3</sub> contents  
116 of chromitites reach dozens of wt.%<sup>12</sup>. Such contrasting Cr contents suggest that the  
117 parental magmas of chromitites must be dozens to hundreds of times larger than the  
118 chromitites themselves<sup>21</sup>. Therefore, ophiolitic chromitites are commonly thought to  
119 be generated via accumulation of chromite and olivine under magma-dominated  
120 environments, e.g., in melt channels or magma chambers<sup>21,22</sup>, and their compositions

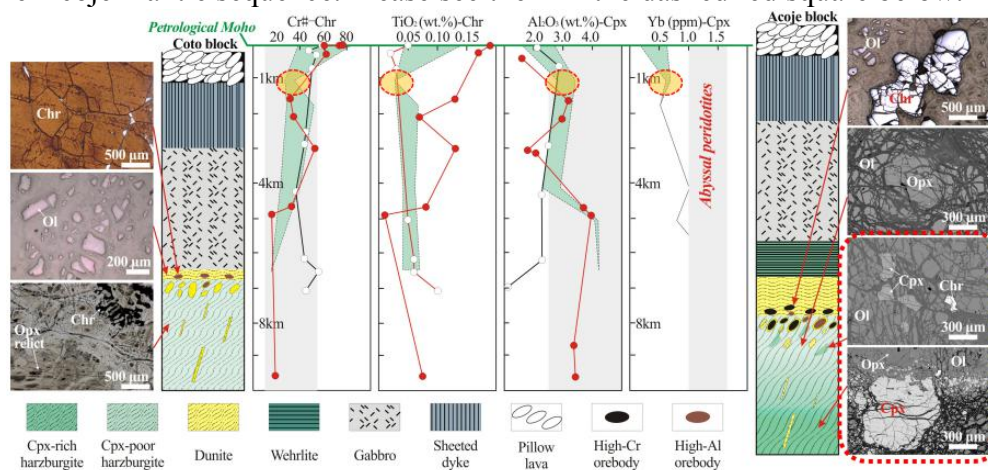
*Q3. The description of the lithologies has improved. However, they show evidence from other works and other ophiolites to support their Cpx-poor Hz1, Cpx-rich Hz2, and Cpx-poor Hz3 lithologies. It would be more interesting for the work that they*

show these differences in the samples that they have studied. In the letter they give a lot of information regarding this topic, but they use images from other ophiolites and it is not clear if this differentiation arises from their observations or not.

**Our reply:** Many thanks for the comment. Sorry for my redundant explanations last time that may have confused you. Actually, we offered geological and compositional evidence of our own in our last response letter. The figures and evidence from other ophiolites were used to better support that our conclusions can be widely applied. We would like to give some brief explanations again below.

### 3.1 Transformation from Cpx-poor Hz-1 to Cpx-rich Hz-2

In the current Acoje mantle sequence, we cannot see Cpx-poor harzburgites of the first generation (Cpx-poor Hz-1) anymore. This is because all of them had been converted to Cpx-rich harzburgites (Cpx-rich Hz-2), which dominate bottom mantle sequence of the Acoje block and can also be found in the middle and uppermost parts of the sequence. As we explained last time based on the textural and compositional features, the Cpx-rich Hz-2 were refertilized products of asthenospheric melts, so there must be proto-lithos for the Cpx-rich Hz-2, and these proto-lithos must have less modal% of Cpx than the Cpx-rich Hz-2. This is how the Cpx-poor Hz-1 came from. In addition, we showed in our paper the photos of Cpx-rich Hz-2 from different levels of the Acoje mantle sequence. Please see them in the dashed red square below.



Note: The two BSE images marked by the dashed red square (on the right) are for Cpx-rich Hz-2 found at the bottom and uppermost mantle sequence of the Acoje block, respectively. The four brown circles marked in the composition columns show the more fertile enriched compositional features of the Cpx-rich Hz-2 found in the uppermost part of the Acoje mantle sequence.

### 3.2 Transformation from Cpx-rich Hz-2 to Cpx-poor Hz-3

Let's see the transformation from the Cpx-rich Hz-2 to the Cpx-poor Hz-3, both of which can be found in the current mantle sequence. The uppermost part of the mantle sequence experienced intense melt-peridotite reaction due to the high melt/rock ratios at this level.

**Cpx-rich harzburgite+Mg-rich Melts→Cpx-poor harzburgite/dunites+less Mg-rich melts**

This is why Cpx-poor harzburgites and dunites dominate this level. On the other hand, Cpx-rich Hz-2 can also be found mixing with Cpx-poor harzburgites in the uppermost mantle sequence. E.g., the peridotites found at the Stop 4 (Fig. 2D) were Cpx-rich harzburgites, but peridotites surrounding them are Cpx-poor harzburgites based

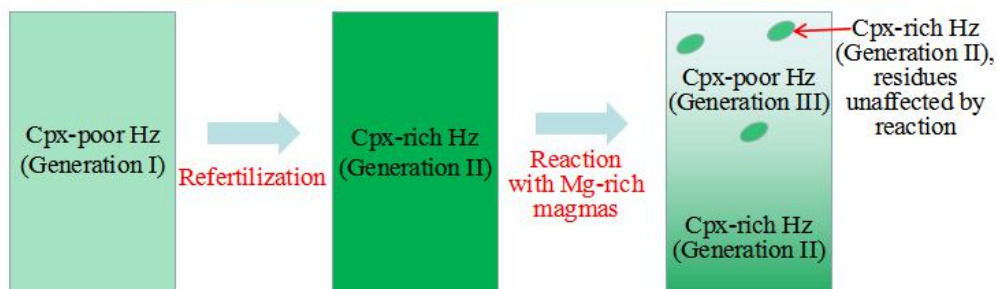


on our field and petrographic observations. Even at the mantle-MTZ boundary (e.g., Stop 5), both Cpx-rich harzburgites and Cpx-poor harzburgites can be found to be mixed together. Such spatial relationships mean the Cpx-rich harzburgitic bodies in the uppermost mantle sequence were residues of melt-peridotites reaction, or they were not strongly affected by the reaction. The compositional variation trend below also support that the Cpx-poor harzburgites and dunites were reaction products between the Cpx-rich Hz-2 and Mg-rich magmas, e.g., boninitic ones (please see the variation trend toward IBM boninites).

[Redacted]

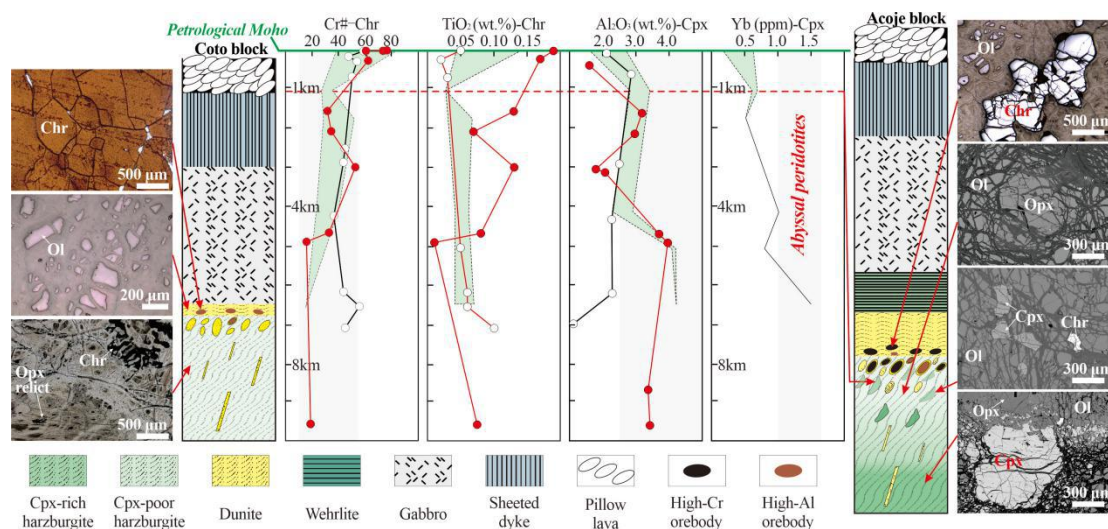
Cpx-poor Hz-1	<ul style="list-style-type: none"> <li>➤ Disappeared and possibly the partial melting residues of MORB-like FAB.</li> <li>➤ Already transformed to Cpx-rich harzburgite (Cpx-rich Hz-2).</li> <li>➤ Their ever presence can only be postulated from the textures of Cpx-rich Hz-2, especially based on the outward growth history of Cpx.</li> </ul>
Cpx-rich Hz-2	<ul style="list-style-type: none"> <li>➤ Transformed from the Cpx-poor Hz-1.</li> <li>➤ Once occupied the whole Acoje mantle sequence.</li> <li>➤ Now still dominate the bottom mantle sequence.</li> <li>➤ Can also be found as enclosed bodies in Cpx-poor harzburgite (Cpx-poor Hz-3) in the uppermost mantle sequence. (pls see the diagram below)</li> </ul>
Cpx-poor Hz-3	<ul style="list-style-type: none"> <li>➤ Compositions of minerals (e.g., high TiO<sub>2</sub> contents of chromite) and their spatial relationship to the Cpx-rich Hz-2 do not support they were partial melting residues.</li> <li>➤ They were transformed products from the Cpx-rich Hz-2 via reaction with Mg-rich magmas (pls see the compositional diagram above).</li> <li>➤ The enclosed Cpx-rich Hz-2 in them are reaction residues.</li> </ul>
Dunite Chromitite	<ul style="list-style-type: none"> <li>➤ Generated at the same time to the Cpx-poor Hz-3.</li> </ul>

**Temporal Lithological Variation and Stratification in the Acoje Mantle Sequence**



It is noted that the uppermost mantle sequence was the place where magmas merged and even formed magma chambers (imagine the case below spreading centers), so the melt/rock ratios at this level were higher than at the bottom part, and the degrees of melt modification at the uppermost mantle were thus the most remarkable. This explains why Cpx-poor Hz-3 were only found at the uppermost mantle sequence. In places where melt/rock ratios were very low, Cpx-rich Hz-2 could be preserved as enclosed bodies. This is why we found Cpx-rich Hz bodies in the Cpx-poor Hz background.

In order to better remind future readers that there are indeed enclosed bodies of Cpx-rich harzburgites (Cpx-rich Hz-2) in the Cpx-poor Hz-3 at the uppermost Acoje mantle sequence, we specially draw in the Fig.3 a dashed red line at the corresponding level and connected the line to the enclosed Cpx-rich harzburgites in the lithological column of the Acoje block. Please see it in the revised Fig.3 below. Moreover, we also have more detailed field photos in the Supplementary Information. Please also see there for more information.



*4. Regarding the temperature of transitional magmas vs boninitic magmas, the figure they are including from Grove and Parman (2004) would not apply, since it does not considerate fore-arc basalts (it is previous to its description) or transitional magmas. Besides, the water content in boninites is a feature that needs consideration. Besides, the authors should provide appropriate references to support that boninites forming high-Cr chromitites had higher temperatures than the transitional magmas forming high-Al chromitites. There are many statements in the letter that are not supported by appropriate references.*

**Our reply:** Many thanks for the comment. Sorry for not making relevant point clear. Actually, our explanations on the temperature variation last time were based on other factors, e.g., LAB depth, geothermal gradients and mantle source compositions. [The paper Grove and Parman \(2004\) was not cited in our paper](#), and the paper was just used in the response letter to support our ideas and for better answering your previous question. Please see more of our replies on this below.

*Q4.1 Why did we use the paper Grove and Parman (2004) for the comparison?*

As we know and reiterated before, [forearc basalts have MORB-like compositions and share similar origin to MORB](#), both of which were generated via decompressional melting of asthenosphere at shallow depth below spreading centers. As a result, it is reasonable to postulate that the formation temperatures of forearc basalts and MORB are similar to each other. Actually, we can consider the MORB to a broader meaning as MORB-like magmas.

As for the transitional magmas, they were also called depleted tholeiites, low-Ti tholeiites, depleted FAB, and transitional FAB. The compositions of [these transitional magmas are similar to island-arc tholeiites](#) (IAT, e.g., [Dilek and Thy, 2009, Lithos](#)).

Island arc tholeiite to boninitic melt evolution of the Cretaceous Kizildag (Turkey) ophiolite: Model for multi-stage early arc-forearc magmatism in Tethyan subduction factories

Yildirim Dilek <sup>a,\*</sup>, Peter Thy <sup>b</sup>

[Such IAT-like magmas can be developed not only at the transitional FAB-boninitic period but also at the early stage of real subduction when arc is established](#) (e.g., [Viruete et al., 2006, Lithos](#)). This is because the sub-arc mantle during the early stage of real subduction was also featured by more depleted compositions and lower geothermal gradient than FMM (sources for MORB-like magmas).

Please also see the following diagram from [Ishizuka et al. \(2014, Element\)](#), which also shows presence of arc tholeiites after boninitic magmatism in the lava sequence.

[Redacted]

Such similarity [enable us to compare the origins of the transitional magmas with those of arc tholeiitic basalts developed in arcs](#). That is why we used the diagram of [Grove and Parman \(2004\)](#) last time for illustration, but we did cite that paper in our work.

*Q4.2 The author should provide appropriate references to support that boninites forming high-Cr chromitites had higher temperatures than the transitional magmas forming high-Al chromitites.*

Actually, we used the results of melt inclusions and also experimental works in our main text to support what you mentioned above. Unfortunately, we did not mention this in the last response letter, so you may not realize relevant discussions.

As we know, melt inclusions in crystals were captured melt droplets during the crystallization of their crystal host, so the compositions of melt inclusions could be considered equal to those of the parental magmas of the crystal hosts. As a result, the homogenization temperatures of melt inclusions in high-Al chromite and high-Cr chromites could be used for the understanding the question mentioned by you. The melt inclusions in high-Al chromite can be homogenized completely at  $\sim 1300$  °C, whereas those in high-Cr chromite and associated olivine phenocrysts from picritic and boninites generally require higher homogenization temperatures, even up to over  $1400$ °C. Such comparisons mean that the parental magmas of high-Cr chromite were formed under higher temperature conditions than those of high-Al ones. Moreover, the results of experimental studies can also support this. We have incorporated such information in the Fig. 7, especially in Fig. 7B where the information of melt inclusions are included.

In order to emphasized the importance of melt inclusion works cited in the paper, we added more information about the principles of melt inclusion studies. Please see our information about melt inclusions and the newly added lines.

331 (Fig. 7B). Melt inclusions in chromite grains are generally interpreted as trapped  
332 parental melts of the chromite hosts. Given that chromite is the first crystallized phase  
333 from the parental magmas of chromitites<sup>3,21</sup>, the homogeneization temperatures of  
334 melt inclusions in chromite from chromitites could be approximately taken as the  
335 liquidus temperatures of their parental magmas<sup>49</sup>. According to previous studies, The  
336 melt inclusions in high-Al chromite can be homogenized completely at  $\sim 1300$  °C<sup>50,51</sup>,  
337 whereas those in high-Cr chromite and associated olivine phenocrysts from picritic  
338 and boninites generally require higher homogenization temperatures, even up to  
339  $1400$  °C<sup>49,52</sup> (Fig. 7B). Consequently, both experimental and melt inclusion studies  
340 suggest the parental magmas of high-Cr chromitites generally formed under higher  
341 temperature conditions than those of high-Al ones, also supporting presence of a

*Overall, the manuscript shows marked improvement and comprehensively addresses many of the previous comments. I recommend the acceptance of the manuscript for publication after minor revision.*

**Reviewer #3 (Remarks to the Author):**

*I am very satisfied with the revisions to the manuscript and the author's answers to the questions raised, and do not have any further suggestions for revisions.*

**Our reply:** Many thanks for reviewing the paper again and your encouragement to us. Your time, effort and consideration are all highly appreciated. All our best wishes to you.