

Supplementary Information

Diversities of chromite mineralization induced by chemo-thermal variations of mantle during subduction initiation

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Supplementary Information List:

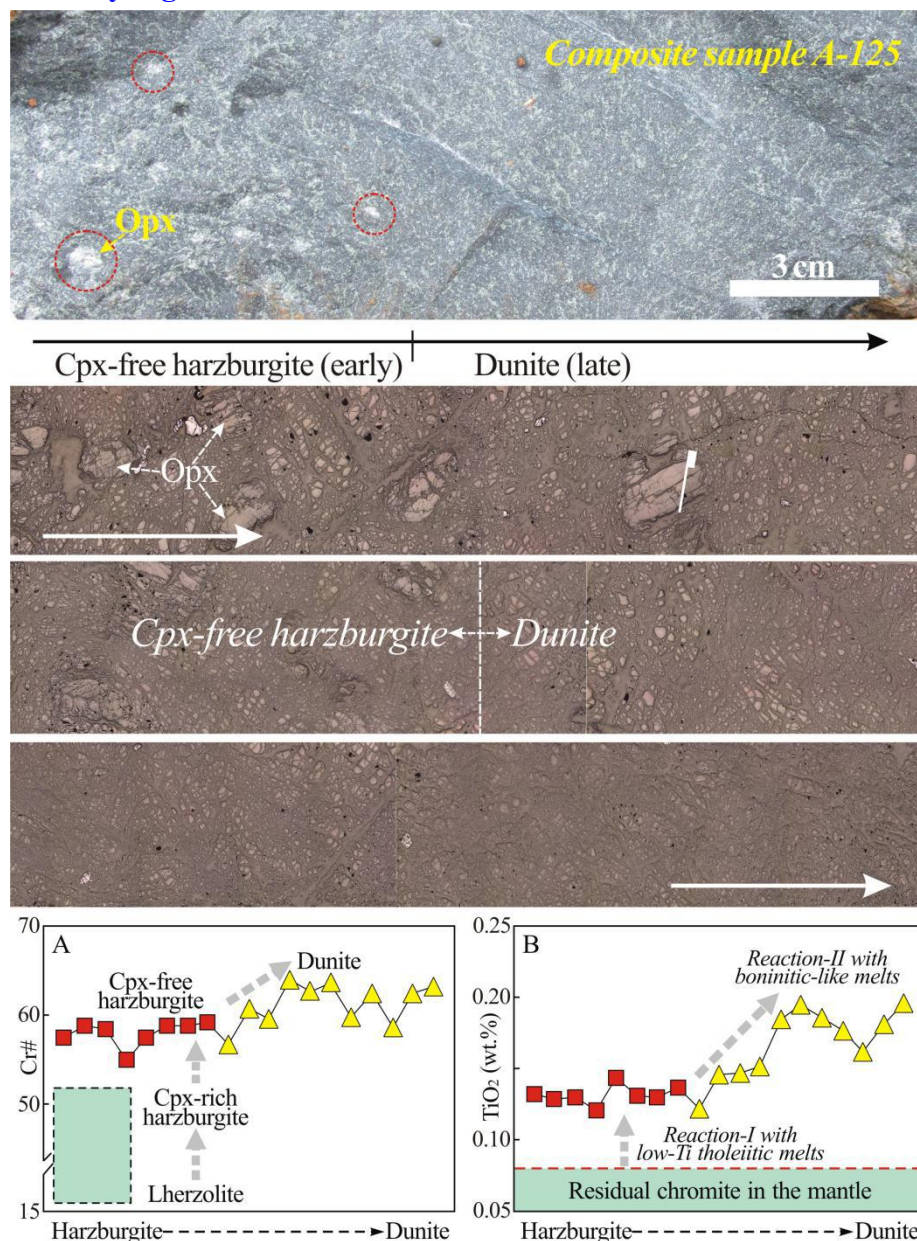
- **Supplementary Information to the Fig-1.**
- **Supplementary Fig-1:** Petrological and compositional variations in a composite sample of harzburgite and dunite from mantle sequence of the Acoje block.
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Supplementary Information to the Fig-1.

Papers cited for each ophiolitic block in the Fig.1 can be found as below and in the [Supplementary References](#):

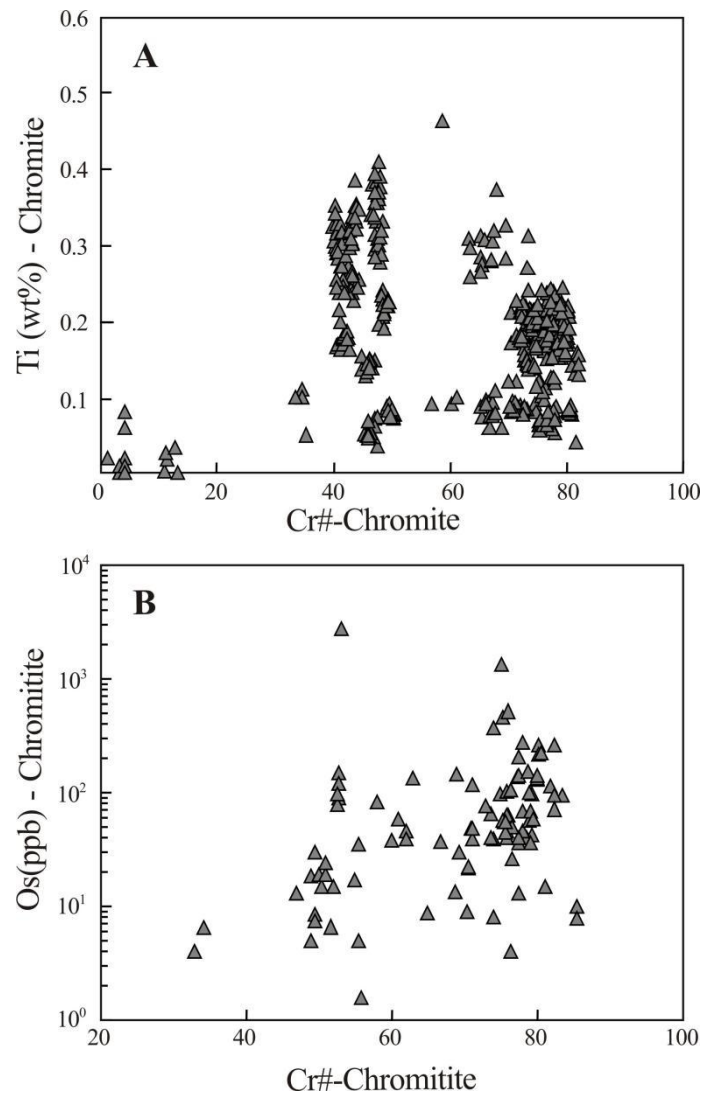
Sangun, Matsumoto et al. show¹; **Tari-Misaka**, Arai & Yurimoto show²; **Sartohay**, Zhou et al. show³; **Hegenshan**, Robinson et al. show⁴; **Moa-Baracoa**, Proenza et al. show⁵, Marchesi et al. show⁶, Rui et al. show⁷; **Coto**, Zhang et al. show⁸; **Cabangan**, Yumul & Dimalanta show⁹; **Medellin**, Hernández-González et al. show¹⁰; **Outokumpu**, Liipo et al. show¹¹; **Mayari-Crystal**, Proenza et al. show⁵, Marchesi et al. show⁶; **Acoje**, Zhang et al. show¹²; **Cyprus**, Greenbaum show¹³, Batanova & Sobolev show¹⁴; **Kop**, Zhang et al. show¹⁵; **Lycian**, Uysal et al. show^{16,17}, Xu et al. show¹⁸; **Fizh**, Takazawa et al. show¹⁹, Rollinson et al. show²⁰; **Tiebaghi**, Moutte show²¹, Ulrich et al. show²²; **Kempirsai**, Melcher et al. show²³, Savelieva et al. show²⁴; **Kraka**, Saveliev & Fedoseev show²⁵, Garuti et al. show²⁶; **Purang**, Gong et al. show²⁷, Xiong et al. show²⁸; **Dongbo**, Xiong et al. show²⁹; **Zedang-Luobusa**, Xiong et al. show³⁰, Zhang et al. show³¹.

Supplementary Fig-1.



Supplementary Fig-1. Petrological and compositional variations in a composite sample of Cpx-free harzburgite and dunite from mantle sequence of the Acoje block. The hand-specimen and photo-micrographs above show decreasing modal% of orthopyroxene (Opx) from the Cpx-free harzburgite to the dunite parts, marked by the black and white arrows. (A): Variation of Cr# of chromite from the Cpx-free harzburgite to dunite. (B): Variation of TiO₂ contents of chromite from the Cpx-free harzburgite to dunite. Based on the Cr# and TiO₂ contents of chromite, the harzburgite part with high-Al chromite in the composite sample was not pure melting residue of FMM but had been modified by melt with low-Ti tholeiitic affinity (Reaction-I), whereas the dunite with high-Cr chromite was formed via reaction between Cpx-free harzburgite and boninitic-like melts (Reaction-II). The reactions elevated the TiO₂ contents of chromite twice from extremely low level of partial melting residue to ~ 0.2 wt.%. Such spatial relationship supports that low-Ti tholeiitic magmatism happened earlier than boninitic-like ones in the Acoje block, consistent with the framework of subduction initiation. The data of chromite are from Zhang et al. show¹².

Supplementary Fig-2.



Supplementary Fig-2: Plots of TiO₂ vs. Cr# of chromite (A) and bulk-rock Os vs. Cr# of chromite (B) in ophiolitic chromitites worldwide. The data used in the panel A are from Zhang et al. Show¹⁵, Xu et al. Show¹⁸, Xiong et al. show^{29,33}, Zhou et al. show³², Liu et al. show³⁴, and Su et al., show³⁵. The data used in the panel B are from Uysal et al. show¹⁶, Xiong et al. Show^{28,29,37,38}, and Gervilla et al. show³⁶.

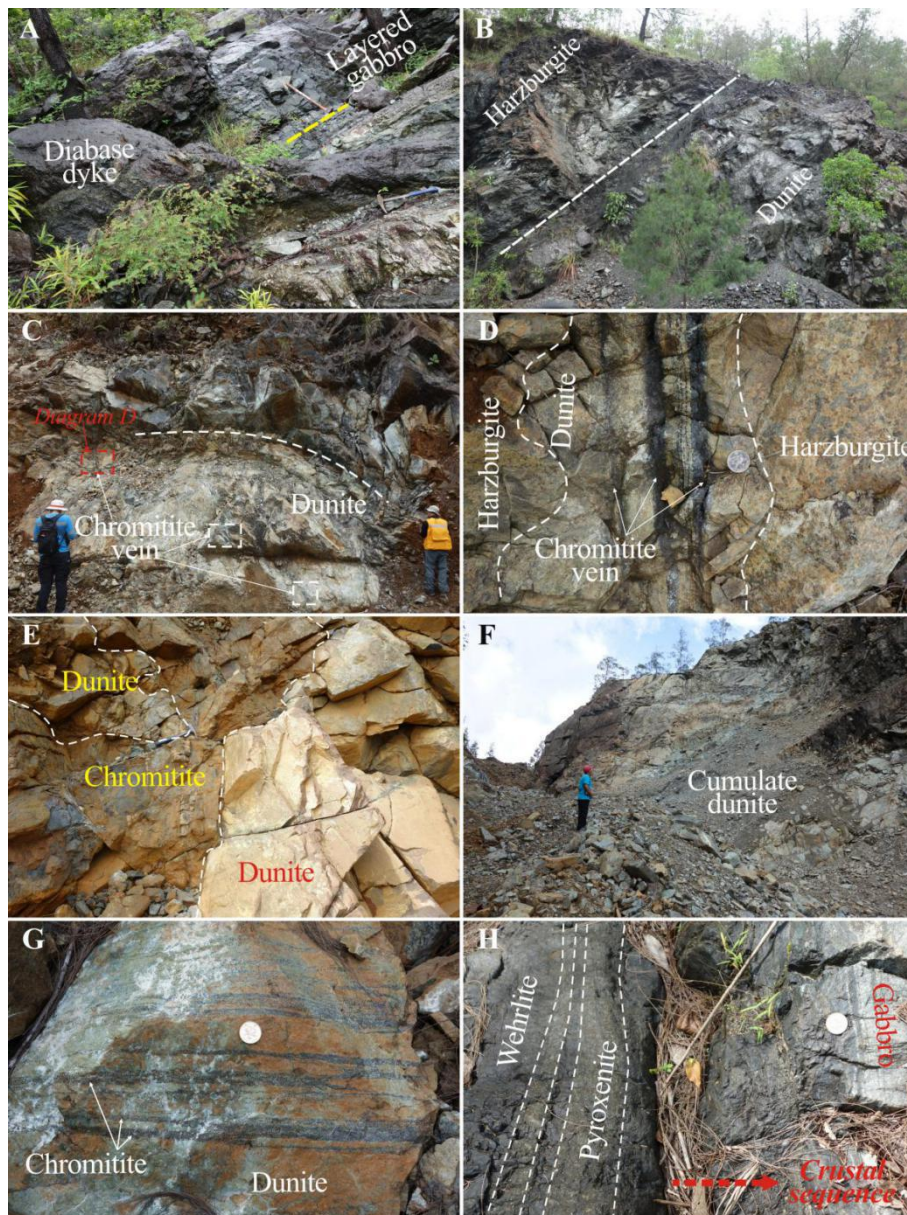
Supplementary Note-1: Geological background of the Zambales ophiolite

The Zambales ophiolite, situated in northwest Luzon Island (Fig. 2A-B), ~ 130 km long and 4500 km² big in area³⁹. The ophiolite consists from north to south of the Masinloc, Cabangan, and San Antonio massifs (Fig. 2B). The Masinloc massif is the biggest and further divided into the Acoje block in the north and the Coto block in the south^{40,41} (Fig. 2B), which host world-class high-Cr and high-Al chromite deposits, respectively (25 Mt vs 6.34 Mt)⁴². Systematic chronological studies documented that the magmatism in the Acoje block took place later than in the Coto block (44-43 vs 45 Ma)^{43,44}, but the two blocks were revealed as adjacent parts of the same proto-forearc lithosphere with transitional compositions and generated in an evolving subduction initiation system^{8,43,47}. The Coto block records the stage of proto-forearc spreading before boninitic magmatism, including an early stage of FAB magmatism and a later stage of arc tholeiitic magmatism, whereas the Acoje block exhibits magmatism varying from arc tholeiitic to boninitic types^{43,45-48}.

The Coto block contains four stratigraphic sequences: harzburgitic mantle, dunitic MTZ, mafic cumulates, and a volcanic-hypabyssal suite^{40,43,48,49} (Supplementary Fig. 3A-B; Fig. 2C). The mantle sequence of the block is mainly made of Cpx-poor harzburgites with few clinopyroxene, and there is no obvious variation of mineral proportions is observed from top to bottom of the mantle sequence^{8,40,49} (Fig. 3). There is no evidence suggesting that the mantle sequence of the block was ever refertilized either. Clinopyroxene and chromite in the harzburgites at different stratigraphic levels show limited chemical variations and have compositions similar to their counterparts in the Acoje Cpx-poor harzburgites⁴⁹ (Fig. 3). The MTZ is mainly made of serpentinized dunites and host chromitite pods near the mantle-MTZ border^{8,50} (Fig. 2C and 3). Chromite in the ores share similar Cr#s to those high-Al ones in the Acoje block but have large TiO₂ variation (< 0.07-0.28%)^{8,32} (Fig. 3 and 5). Geochemical studies suggest the Coto block successively experienced FAB and IAT magmatism in a proto-forearc setting^{43,45}. However, development of the Coto block expired after the IAT magmatism and high-Al chromite mineralization, leaving no obvious record of boninitic magmatism in the block.

The Acoje block is also divided into four sequences from west to east: mantle, Moho Transition zone (MTZ), mafic cumulates and hypabyssal-volcanic rocks^{45,52} (Fig. 2D). The uppermost part of the mantle sequence mainly comprises of Cpx-poor harzburgites that contain dunite and Cpx-rich harzburgite bodies^{12,40,49,51} (Fig. 2D and 3). From top to bottom of the mantle sequence, there is an increasing trend of pyroxene modal%, which can be judged from the pyroxene relicts on the weathered surfaces of peridotites (Supplementary Fig. 3C-D), and the Cpx-poor harzburgite vary gradually into Cpx-rich harzburgites and Cpx-poor lherzolites (grouped as Cpx-rich harzburgites in this study)^{12,49,51} (Fig. 3). Clinopyroxene grains in the Cpx-rich harzburgites show extended growth into the interstitial spaces and are characterized by prevalent figure-like protrusions, forming clinopyroxenite veins at times^{40,51} (Fig. 3). The compositions of both chromite and clinopyroxene grains in the harzburgites vary from those with MORB-like at the bottom to more boninitic-like affinities at the top of the mantle sequence^{12,49,51} (Fig. 3). The MTZ is mainly made up of dunites^{40,52}

(Fig. 2D and 3), and the chromite deposits are all distributed along the mantle-MTZ boundary^{12,49,52} (Fig. 2D and 3). Apart from some high-Al chromitites¹² (Sampling location 5 in Fig. 2D), the majority of chromitites in the block belong to the high-Cr variety and are everywhere spatially associated with dunites (Fig. 3; Supplementary Fig. 3E-G). The MTZ dunites vary stratigraphically upward into layered wehrlites and pyroxenites near the MTZ-crust boundary (Fig. 3; Supplementary Fig. 3H). The lithological distribution, petrographic features and chemical variations above have been ascribed to the combined effects of an early event of refertilization by asthenospheric components and later modification of the refertilized peridotites by Mg-rich melts related to different types of chromite mineralization^{12,51}. The former event took place in the whole mantle sequence and converted all peridotites to Cpx-rich harzburgites, whereas the latter happened only at the uppermost mantle sequence, consuming pyroxene in the previously refertilized mantle and resulting in Cpx-poor harzburgites, dunites and chromitites under varying melt/rock ratios in both the upper mantle and MTZ⁵¹.



Supplementary Fig-3. Outcrops of different lithologies and their spatial relationship in the Coto (A-B) and Acoje blocks (C-H). (A): Layered gabbro and diabase dyke at the lowermost crustal sequence of the Coto block. The yellow mark the occurrence of the gabbro layer. (B): Harzburgite and dunite at the uppermost mantle sequence of the Coto block. (C): Contact between harzburgite and dunite bodies at top of the Acoje mantle sequence (near the Stop-4 in [Fig.2D](#)). Chromitite veins (~ 3-10 cm wide) can be found in the dunite body. (D): Close-up image showing detailed spatial relationships among chromitite vein, dunite and harzburgite (country rock) at top of the Acoje mantle sequence (near the Stop-6 in [Fig.2D](#)). The zones of chromitite and dunite were reaction products between the harzburgite and parental magmas of the chromitites. (E): Co-existing chromitite and dunite bodies (Stop-7 in [Fig.2D](#)). (F): Dunite body of cumulative origin in the Acoje MTZ (Near the Stop-6 in [Fig.2C](#)). (G): Interlayered dunite and chromitite found in the dunite body showed the panel F, supporting their cumulative origin. (H): Layered wehrlites, pyroxene and gabbros at the topmost part of the MTZ. Presence of gabbros marks the bottom level of the Acoje crustal sequence.

Supplementary Note-2: Fractional melting model and related parameters used in the Fig. 4D.

The TiO₂ content of fertile MORB mantle (FMM) used for the modeling is set to be 0.18% according to Pearce et al. show⁵³. The modes (X_i) of olivine, orthopyroxene, clinopyroxene and spinel grains in the FMM are set to be 58.9%, 23.5%, 14.7% and 2.80%, respectively, as were used in Jerram et al. show⁵⁴ for fertile spinel peridotites. The melting modes (p_i) of olivine, orthopyroxene, clinopyroxene and spinel used in the forward modeling are set to be -0.29%, 0.46%, 0.71% and 0.11% under anhydrous conditions according to Jerram et al. show⁵⁴. The values are set as -0.205%, 0.505%, 0.63% and 0.07% under hydrous conditions by averaging relevant data compiled in Gaetane & Grove show⁵⁵. Melting of the FMM is supposed to happen under anhydrous condition, whereas melting of the mantle residues (MR) after 10%, 15% and 20% melting degrees are supposed to happen under hydrous conditions. The partition coefficients (k) of Ti between melt and olivine, orthopyroxene, clinopyroxene and spinel are 0.015, 0.15, 0.3 and 0.07, respectively, according to Kelemen et al. show⁵⁶.

The melting equations for obtaining the TiO₂ contents of melts (Cl) and solid residues (Cs) during fractional melting are listed as below:

$$Cl = (Co/F) * [1 - (1 - PF/Do)(1/P)]$$

$$Cs = [Co / (1-F)] * [(1 - PF/Do)(1/P)]$$

$$Do = Xo(ol) * k(ol) + Xo(opx) * k(opx) + Xo(cpx) * k(cpx) + Xo(sp) * k(sp)$$

$$P = p(ol) * k(ol) + p(opx) * k(opx) + p(cpx) * k(cpx) + p(sp) * k(sp)$$

Where Co , Xo , Do , F and P represent the Ti contents in the starting peridotites (not necessarily FMM), the mode of each phase in the starting peridotites, initial bulk-rock distribution coefficients of Ti, partial melting degree, and bulk reaction coefficient of Ti, respectively. When some a phase is used out during the calculation, the overall modes of all phase will be re-normalized to 100% for further processing.

Supplementary Table-1 Major element compositions of chromite in chromitites and Re-Os concentrations and isotopes of chromitites and harzburgites.

Sample	GPS Coordinates and Location	Cr#	TiO ₂ (wt.%)	Re(ppb)	2SE	Os (ppb)	2SE	¹⁸⁷ Os/ ¹⁸⁸ Os	2SE	¹⁸⁷ Re/ ¹⁸⁸ Os	2SE	
<i>Acoje chromitites</i>												
AC-01*	15°43'13"N, 120°03'22"E; Stop-7	73.4	0.21	0.118	0.028	64.828	0.126	0.12734	0.00015	0.036	0.003	
AC-05*	15°43'13"N, 120°03'22"E; Stop-7	71.9	0.18	0.486	0.043	24.588	0.028	0.12700	0.00024	0.095	0.008	
AC-19	15°43'13"N, 120°03'22"E; Stop-7	74.8	0.21	0.063	0.038	12.707	0.014	0.12683	0.00022	0.024	0.014	
AC-38	15°43'13"N, 120°03'22"E; Stop-7	74.8	0.19	0.073	0.026	32.955	0.073	0.12844	0.00016	0.011	0.004	
AC-33	15°43'13"N, 120°03'22"E; Stop-7	74.0	0.19	0.099	0.027	36.781	0.118	0.12881	0.00021	0.013	0.004	
AC-57	15°42'0.9"N, 120°03'23"E; Stop-6	73.2	0.19	0.036	0.027	135.165	1.084	0.12760	0.00024	0.001	0.001	
AC-42	15°42'36"N, 120°03'21"E; Stop-5	46.2	0.15	0.052	0.027	1.659	0.001	0.13205	0.00047	0.151	0.078	
AC-45	15°42'36"N, 120°03'21"E; Stop-5	46.8	0.15	0.071	0.050	1.862	0.001	0.14710	0.00045	0.185	0.131	
AC-46	15°42'36"N, 120°03'21"E; Stop-5	46.6	0.15	0.063	0.025	4.778	0.004	0.13085	0.00038	0.063	0.026	
<i>Coto chromitites</i>												
ZB-03*	120°05'33"E, 15°34'46"N; Open pit	48.3	0.22	0.301	0.009	12.245	0.029	0.12942	0.00022	0.118	0.004	
ZB-07*	120°05'33"E, 15°34'46"N; Open pit	50.9	0.08	0.157	0.009	18.378	0.087	0.13294	0.00024	0.059	0.004	
ZB-10*	120°05'33"E, 15°34'46"N; Open pit	42.7	0.14	0.120	0.008	10.239	0.165	0.12829	0.00052	0.057	0.004	
ZB-12*	120°05'33"E, 15°34'46"N; Open pit	45.7	0.11	0.224	0.015	11.509	0.089	0.12888	0.00048	0.094	0.006	
ZB-14*	120°05'33"E, 15°34'46"N; Open pit	41.9	0.18	0.067	0.007	1.949	0.005	0.13489	0.00036	0.166	0.018	
C-26	120°05'0.5"E, 15°34'21"N; Open pit	47.3	0.06	0.101	0.006	26.158	0.216	0.12605	0.00027	0.019	0.001	
C-31	120°05'0.5"E, 15°34'21"N; Open pit	46.6	0.07	0.073	0.006	13.153	0.043	0.12971	0.00021	0.027	0.002	
DEC-01	120°05'0.5"E, 15°34'21"N; Open pit	47.3	0.03	0.104	0.008	10.007	0.037	0.12904	0.00028	0.050	0.004	
DEC-02	120°05'0.5"E, 15°34'21"N; Open pit	47.2	0.05	0.098	0.008	14.497	0.046	0.12869	0.00025	0.032	0.003	

Note: the Cr# and TiO₂ contents of chromite in the chromitites with and without * are from [Zhou et al. show¹⁴](#) and [Zhang et al. show^{8,12}](#), respectively.

Supplementary Table-1 Continued

Sample	GPS Coordinates and Location	Re (ppb)	2SE	Os (ppb)	2SE	$^{187}\text{Os}/^{188}\text{Os}$	2SE	$^{187}\text{Re}/^{188}\text{Os}$	2SE
<i>Cpx-rich harzburgites</i>									
A-75	15°41'49"N, 120°02'58"E; Stop-4	0.172	0.012	2.843	0.016	0.12736	0.00017	0.045	0.009
A-78	15°41'49"N, 120°02'58"E; Stop-4	0.290	0.012	3.456	0.015	0.12654	0.00021	0.404	0.016
A-154	15°41'31"N, 120°02'44"E; Stop-3	0.173	0.008	5.325	0.064	0.12599	0.00020	0.156	0.007
<i>Cpx-poor harzburgites</i>									
C-14	15°34'20"N, 120°05'50"E; Open pit	0.081	0.002	2.491	0.016	0.12600	0.00025	0.156	0.005
C-15	15°34'20"N, 120°05'50"E; Open pit	0.027	0.005	4.426	0.039	0.12647	0.00016	0.029	0.006
C-20	15°34'20"N, 120°05'50"E; Open pit	0.063	0.005	1.848	0.011	0.12338	0.00020	0.165	0.013
C-21	15°34'20"N, 120°05'50"E; Open pit	0.149	0.009	5.275	0.039	0.12391	0.00022	0.136	0.008
C-22	15°34'20"N, 120°05'50"E; Open pit	0.063	0.013	3.292	0.014	0.12428	0.00014	0.039	0.007
A-120	15°42'36"N, 120°03'21"E; Stop-5	0.442	0.013	7.866	0.120	0.12846	0.00022	0.271	0.009
A-124	15°42'36"N, 120°03'21"E; Stop-5	0.108	0.005	3.733	0.035	0.12532	0.00016	0.139	0.006
(BIR-1)-01		0.683	0.012	0.342	0.000	0.13419	0.00025	9.628	0.166
(BIR-1)-02		0.693	0.013	0.332	0.001	0.13406	0.00023	10.063	0.185
(BIR-1)-03		0.684	0.011	0.352	0.000	0.13403	0.00022	9.357	0.146
BLK-01		0.9	1.2	1.083	0.020	0.15155	0.00324		
BLK-02		0.8	1.2	0.908	0.046	0.16892	0.00218		
BLK-03		1.7	0.9	0.362	0.010	0.19252	0.00210		

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