

Distribution of trace elements in spinel and garnet peridotites

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Abstract The distribution of trace elements in the upper mantle has been discussed on the basis of the trace element abundances in bulk rocks and constituent minerals of two spinel and garnet facies peridotite xenoliths in alkali basalts from eastern China. The data presented are consistent with the suggestion that highly incompatible elements (Rb, Ba, Th, U, Sr, Nb, Ta) mainly reside in intergranular components, and to a lesser extent in fluid inclusions in minerals. The LILE composition in olivine and orthopyroxene can be seriously affected by the presence of fluid inclusions. Consequently the subsolidus partitioning of the LILE cannot be used to infer the olivine-melt and orthopyroxene-melt partition coefficients for these elements. There is a significant difference in $(\text{Opx/Cpx})_{\text{HREE}}$ ratios for spinel and garnet peridotites, suggesting a P-T control on equilibrium partition coefficients.

Keywords: trace elements, distribution, peridotite xenolith, upper mantle, ICP-MS.

Knowledge about the trace element abundance in peridotites and their distribution in constituent minerals is pivotal to understanding the mantle composition and for successful geochemical modeling. There is a consensus concerning the distribution of rare earth elements (REE) in the "dry" upper mantle. It has been concluded that clinopyroxene and garnet are the main hosts of REE, whereas olivine and orthopyroxene do not show significant contributions to the bulk budget of REE^[1-5]. However, considerable debate still remains as to the distribution of highly incompatible elements (e.g. Rb, Ba, U, Th, Nb, Ta) in the mantle. Some authors suggest that these elements are mainly concentrated in intergranular components and in mineral fluid inclusions^[6-8]. In contrast, others found that the budget of highly incompatible elements can be quantitatively accounted for in mineral phases notably including amphibole and its breakdown products^[9]. These authors argued for no significant concentration of these elements in grain boundaries or in fluid inclusions. Obviously the solution of this dilemma requires more high quality trace element data on peridotites and their minerals. In addition, previous studies concentrated heavily on spinel facies peridotites, whereas there is no systematic investigation on garnet peridotites which are believed to come from a deeper portion of the upper mantle^[2]. We thus analyzed two spinel and garnet peridotite xenoliths and their silicate minerals from eastern China using an inductively coupled plasma-mass spectrometry (ICP-MS) to obtain the concentration of 24 trace elements (REE, Rb, Ba, U, Th, Nb, Ta, Sr, Hf, Zr, Ti). These data will be used (1) to investigate the contribution of different minerals or other components to the whole

rock trace element budget; (2) to study subsolidus element partitioning and then to estimate the mineral-melt partition coefficients under mantle conditions.

1 Samples

The samples were collected from Damaping in Hebei Province (X-23) and Mingxi in Fujian Province (S-6). Detailed isotopic studies have already been performed on these samples^[10]. X-23 is a spinel lherzolite whose equilibrium temperature is 1 031 °C. S-6 is a garnet peridotite with an equilibrium temperature of 1 150 °C and a pressure of 2.3 GPa. The modal compositions which were calculated on the basis of bulk rock and mineral major element chemistry are listed in table 1.

Table 1 Major and trace element abundances of spinel and garnet peridotites from E China and calculated modal proportions

	Spinel lherzolite X-23					Garnet lherzolite S-6					
	bulk rock	Opx	Cpx	Ol	Sp	bulk rock	Cpx	Opx	Ol	Gt	Sp
SiO ₂	44.10	55.55	54.43	41.10	0.53	44.20	52.29	54.81	41.50	42.70	0.07
TiO ₂	0.02	0.09	0.22	0.04	0.09	0.16	0.50	0.16	0.00	0.14	0.29
Al ₂ O ₃	2.40	2.83	4.58	0.00	51.75	4.09	6.44	4.92	0.06	23.33	50.09
Cr ₂ O ₃		0.42	1.33	0.01	17.94	0.44	1.21	0.86	0.07	1.62	18.27
FeO	7.75	5.40	2.40	8.84	9.43	7.62	3.16	6.07	9.71	7.00	12.31
MnO	0.11	0.15	0.00	0.36	0.39	0.14	0.05	0.19	0.11	0.31	0.15
NiO	0.00	0.00	0.00	0.42	0.45	0.06	0.06	0.10	0.06	0.00	0.10
MgO	42.04	33.11	15.80	48.72	19.08	37.30	16.18	32.06	49.13	21.28	19.22
CaO	1.83	0.44	19.86	0.06	0.03	3.74	18.46	1.05	0.07	5.06	0.03
Na ₂ O	0.24	0.00	1.68	0.00	0.00	0.35	1.68	0.18	0.02	0.01	0.00
K ₂ O	0.03	0.00	0.00	0.10	0.07	0.01	0.05				0.00
P ₂ O ₅	0.02	0.16	0.22	0.29	0.00	0.04					
H ₂ O ⁺	0.98					0.31					
Total	99.52	99.25	100.52	99.94	99.76	98.91	100.08	100.40	100.73	101.45	100.53
Mode (%) ^{a)}		20	9.1	69.5	1.5		10.2	12.6	60.2	16.8	<0.2
Trace element contents (10 ⁻⁶)											
Rb		0.106	0.038	0.05		0.344	0.437	0.139	0.058	0.063	
Ba	0.672	0.455	0.422	0.36		2.52	1.95	0.531	1.11	0.488	
Th	0.012	0.005	0.076	0.002		0.058	0.181	0.007	0.006	0.011	
U	0.016	0.002	0.016	0.001		0.067	0.03	0.002		0.01	
Nb	0.199	0.067	0.595	0.008		0.559	1.355	0.112	0.013	0.196	
Ta	0.008	0.002	0.053	0.001		0.035	0.142	0.009	0.001	0.012	
La	0.183	0.028	1.141	0.004		0.346	1.995	0.153	0.003	0.019	
Ce	0.329	0.048	3.082	0.009		0.797	5.613	0.549	0.006	0.128	
Pr	0.06	0.007	0.624	0.001		0.156	1.095	0.128	0.001	0.056	
Sr	6.399	0.295	46.57	0.088		19.33	77.36	6.941	0.183	0.295	
Nd	0.279	0.029	3.375	0.004		0.815	5.539	0.729	0.004	0.609	
Sm	0.09	0.01	1.197	0		0.32	1.634	0.243	0.001	0.744	
Zr	1.606	0.87	17.902	0.003		10.501	25.041	4.806	0.093	36.315	
Hf	0.023	0.003	0.57			0.243	0.931	0.178		0.665	
Eu	0.037	0.006	0.49			0.152	0.594	0.09		0.48	
Ti	259.9	473.4	1846.0	15.7		929.955	3217.8	1613.0	48.5	1561.8	
Gd	0.14	0.029	1.843	0.001		0.645	1.867	0.295	0.002	2.481	
Tb	0.027	0.008	0.35			0.141	0.273	0.046		0.642	
Dy	0.191	0.078	2.436	0.002		1.077	1.368	0.263	0.003	5.34	
Ho	0.045	0.024	0.543	0.001		0.262	0.226	0.048	0.001	1.344	
Er	0.141	0.099	1.61	0.003		0.8	0.522	0.12	0.003	4.321	
Tm	0.022	0.021	0.234	0.001		0.124	0.058	0.016	0.001	0.681	
Yb	0.152	0.177	1.476	0.011		0.855	0.318	0.091	0.005	4.691	
Lu	0.024	0.032	0.213	0.003		0.133	0.041	0.013	0.001	0.727	

a) The mode of sample S-6 is after ref. [11].

2 Analytical methods

The bulk major element compositions were obtained using a conventional wet chemistry method at the Guangzhou Institute of Geochemistry. The mineral chemistry analyses were performed with a Joel microprobe at the China University of Geosciences (Beijing). The rocks and minerals were also analyzed to measure the amount of 24 trace elements using ICP-MS at Guangzhou. The analytical procedure follows that described by Liu et al.^[12] The cpx separates were hand picked under a binocular microscope. They were washed with hot 6 mol/L HCl for 30 min and in a supersonic bath for 30 min, and then washed with MILLIQ pure water six times before acid digestion. The leachates were collected for estimation of the trace element composition in grain boundary components. The precision and accuracy were checked against the isotopic dilution data^[10] and are estimated to be between 5% and 10%.

3 Analytical results

3.1 Spinel lherzolite (X-23)

In the primitive mantle-normalized spiderdiagram, clinopyroxene displays a flat pattern (fig. 1(b)). Only Ba and Rb show somewhat strong depletion. There are weak negative anomalies of high field strength elements (HFSE: Nb, Ta, Hf, Zr). The REE contents in orthopyroxene are about 1/10 to 1/70 of those in clinopyroxene. There is a positively sloped distribution for elements

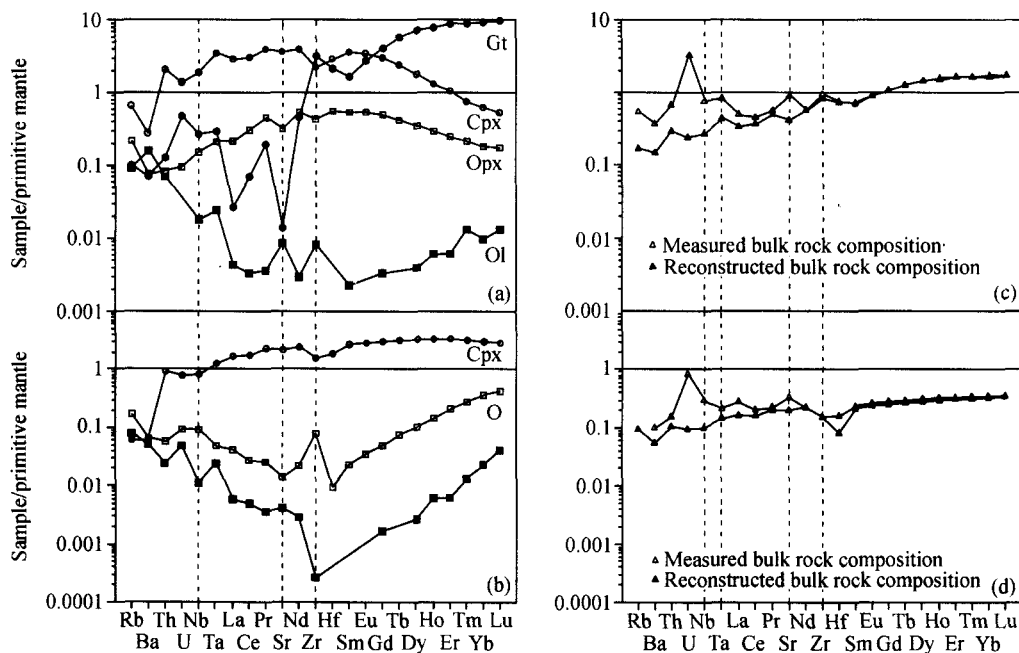


Fig. 1. Primitive mantle-normalized distribution patterns of trace elements in peridotites and minerals: (a) S-6 garnet lherzolite; (b) X-23 spinel lherzolite; comparison of calculated and measured bulk rock trace element compositions: (c) S-6 garnet lherzolite; (d) X-23 spinel lherzolite. The primitive mantle data is after ref. [13]. Ol = olivine; Cpx = clinopyroxene; Opx = orthopyroxene, Gt = garnet. ●, Gt; ○, Cpx; □, Opx; ■, Ol².

from Lu to Sm, whereas the elements more incompatible than Nd display a negatively sloped distribution. Among the silicate minerals analyzed, olivine has the lowest trace element concentration, which represents less than 1/1 000 of that in clinopyroxene. Similar to orthopyroxene, olivine shows a V-type distribution pattern (fig. 1(b)).

3.2 Garnet lherzolite (S-6)

The clinopyroxene in this sample shows an enrichment of the medium REE (MREE) relative to the heavy REE (HREE) and highly incompatible elements, with negative anomalies of HFSE. The orthopyroxene also displays a convex upward pattern but at a lower level. Unlike the spinel lherzolite, the orthopyroxenes in the garnet lherzolite have a negative Zr anomaly but with no anomalies of HFSE. Despite its very low concentration of trace elements, the olivine displays an enrichment of highly incompatible elements. The garnet is enriched in HREE but is depleted in LREE. There are marked positive HFSE anomalies and a negative Sr anomaly in garnet (fig. 1(a)).

4 Discussion

4.1 Mass balance calculation and trace element distribution in peridotites

The whole rock trace element composition of a peridotite can be reconstructed using modal proportions and trace element compositions in minerals. The comparison of this calculated value with the measured data can yield information concerning the trace element distribution in peridotites. It should be indicated that spinel was not considered during the calculation because of its extremely low concentration of trace elements and its low percentage in peridotites (<2%, table 1). The reconstructed whole rock HREE and MREE compositions of sample S-6 fit well with the measured values (fig. 1(c)), suggesting that these elements mainly reside in silicate minerals. However, the calculated values are slightly greater than the measured values in the case of sample X-23 (fig. 1(d)). This unrealistic phenomenon may result from the slightly higher systematic error of the ICP-MS analyses for the samples with relatively low trace element concentrations and the errors associated with the modal composition estimation. There is a marked difference between the calculated and measured concentrations of the highly incompatible elements. The reconstructed values are always lower than the measured ones, implying the residence of highly incompatible elements in the agents other than the silicate minerals.

In sample X-23, the REE in clinopyroxene accounts for 75%—95% of the whole rock REE abundance. This is the reason why the REE composition in clinopyroxene can be taken as a representative of the whole rock composition. Orthopyroxene contains <5% of the whole rock LREE contents. However, the contribution of orthopyroxene to the bulk rock budget increases from 5% to 25% for the elements from the MREE to the HREE (fig. 2). It is noted that the contribution of orthopyroxene to the whole rock Ti and Zr budget (30% and 10%, respectively) is markedly higher than other adjacent elements. It follows that the negative Ti and Zr anomalies in clinopyroxene do not necessarily reflect the characteristics in the bulk rocks, because of the compensation effect of positive Zr and Ti anomalies in orthopyroxene^[4,5] (fig. 2). On the other hand, the con-

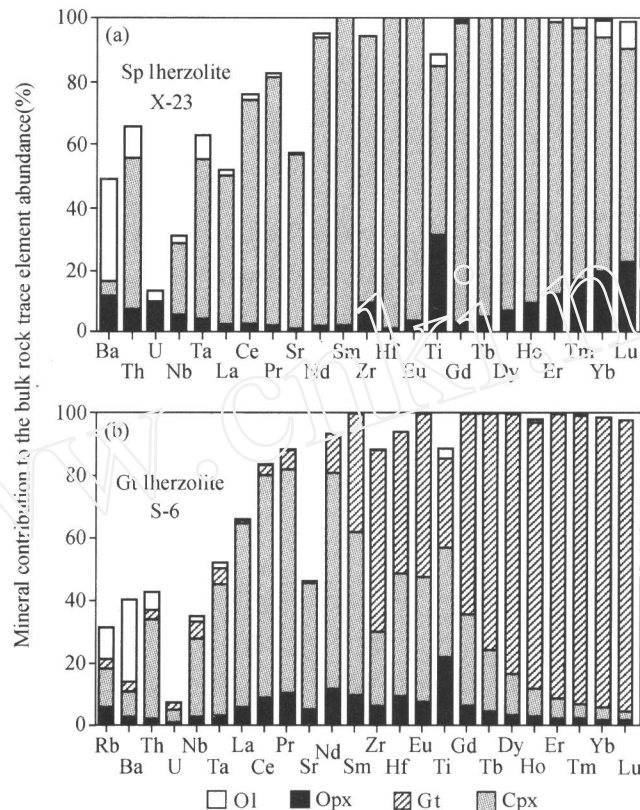


Fig. 2. Diagram illustrating the mineral contributions to the bulk rock trace element abundance of peridotites. (a) X-23; (b) S-6.

tribution of orthopyroxene for some highly incompatible elements (e.g. Ba) exceeds 10%. The role of olivine in whole rock trace element budget is commonly minor, except that it may account for 30% of Ba, that may be due to fluid inclusions in this mineral.

In sample S-6, the clinopyroxene and garnet are the dominant hosts of the REE (fig. 2(b)). Garnet can account for >90% of the HREE and clinopyroxene for >70% of the LREE of the whole rock budget. The contributions of orthopyroxene and olivine are less than 10% for most incompatible elements. The exception is for Ti (20%). Unlike the orthopyroxene in the spinel lherzolite, the orthopyroxene in the garnet lherzolite constitutes about 10% of the whole rock abundance for the LREE and MREE, which is significantly higher than for other highly and moderately incompatible elements (fig. 2).

In summary, more than 95% of the MREE and HREE reside in silicate minerals in dry peridotites. For the highly incompatible elements and HFSE, the total contribution of minerals to the whole rock budget is always less than 100%. This is particularly true for U (10%), Ba (<50%) and Sr (<60%). Is this due to the fact that spinel was not considered in the reconstruction? It is shown that spinel has a crystal structure notoriously unfavorable to the LILE. This is confirmed by some published analyses on spinel, which indicate that the trace element concentration in spinel is

only 1/1 000 of that in clinopyroxene^[8]. It is therefore reasonable to infer that the spinel is not the principle host for incompatible elements.

The experiments show that *Opx/melt* and *Ol/melt* partition coefficients for Rb and Ba are on the order of 0.000 1 and 0.001^[14], suggesting that olivine and orthopyroxene commonly have $<0.1 \times 10^{-6}$ Rb and/or Ba. However, the analyses of olivine and orthopyroxene show anomalously high Rb and Ba contents which exceed the reasonable range predicted by experiments (table 1), and are even higher than those in clinopyroxene (fig. 3). On the other hand, the *Ol/Cpx* and *Opx/Cpx* ratios for the elements more incompatible than Nd increase with increasing elemental incompatibility. More importantly, the *Ol/Cpx* and *Opx/Cpx* ratios in the spinel lherzolite show almost identical variation patterns (fig. 3), strongly suggesting that the distribution of highly incompatible elements in olivine and orthopyroxene is controlled by a similar mechanism. The most likely candidate for this is fluid inclusion. In fact, the trace element compositions of minerals obtained by the ICP-MS include those from minerals themselves and those from fluid inclusions.

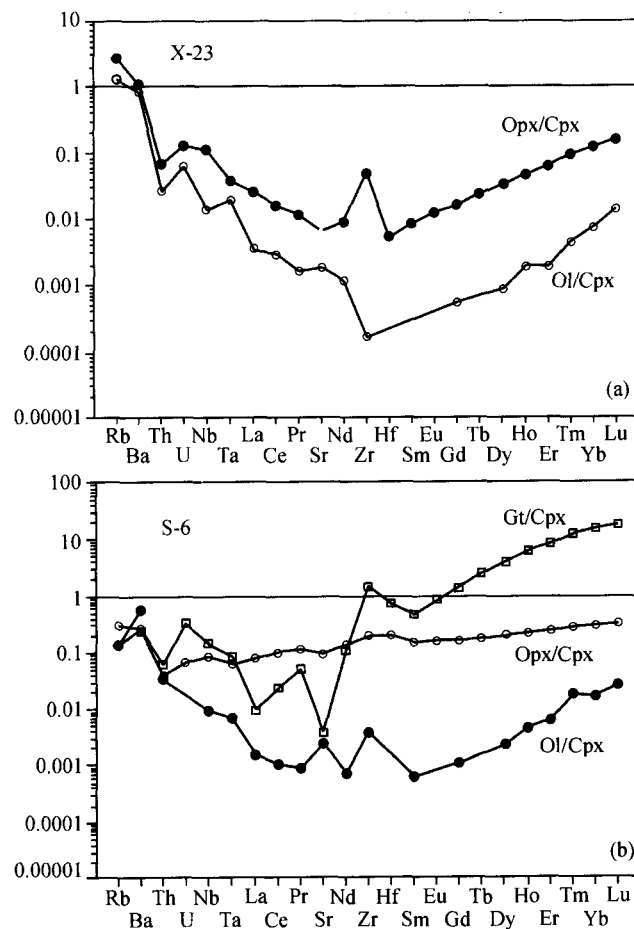


Fig. 3. Mineral-mineral partition coefficients. (a) X-23; (b) S-6.

The effect of fluid inclusion on mineral compositions varies depending on different minerals and different elements. Since it is often enriched in LREE and is depleted in HREE^[7], the fluid inclusion affects the composition of the HREE in minerals to a lesser degree than that of the LREE. The effect of inclusions on olivine and orthopyroxene which have very low trace element contents is the most important, particularly as the LILE contents in these minerals are predominated by those in the fluid inclusions. By contrast, owing to its high concentration of trace elements, clinopyroxene is practically unaffected by fluid inclusions.

Although it dominates in the LILE compositions of olivine and orthopyroxene, the fluid inclusion only plays a minor role in the whole rock budget of most trace elements except for a few highly incompatible elements (Ba) (fig. 3). From the above discussion, it is believed that most of the LILE appear to be concentrated at grain boundaries. This has been confirmed by the analyses of intergranular materials^[15]. Bodinier et al.^[8] have even identified Ti-oxides and phlogopite in the thin reaction layers coating the surface of spinel grains. Due to the extremely high affinity of Nb, Ta and Ba in these minerals, even a very small amount of these minerals can yield important influence on the distribution of these elements in peridotites^[8].

Given these considerations, the data presented in this paper are consistent with the suggestion that highly incompatible elements mainly reside in intergranular components and to a lesser extent in fluid inclusion^[6-8]. Our results call into question the general importance of amphibole and its breakdown products in distribution of highly incompatible elements^[9]. The samples used by Eggins et al.^[9] contain melt pockets. At least two points suggest that these samples are not ordinary peridotites and thus cannot be used as a representative of mantle peridotites. (1) The peridotite xenoliths containing melt pockets is not common and are only found in less than 1% of xenoliths^[16]; (2) the formation of melt pockets is generally related to pervasion of volatile-rich fluids. Consequently the samples with melt pockets commonly contain abundant fluid inclusions^[16]. However, fluid inclusion is not present in the samples of Eggins et al.^[9]

4.2 Determination of mineral-melt partition coefficients

There are already many reliable and internally consistent partition coefficients of trace elements between clinopyroxene and melt ($D^{Cpx/L}$)^[14]. It is therefore possible to estimate the D for other minerals using mineral-mineral partition data and $D^{Cpx/L}$. These data can be used as an important complementary data in addition to the experimentally determined data, and in some cases these data are even superior to the experimental data^[14, 16, 17]. This is because experiments are commonly carried out at unreasonably high temperature (1 400°C), that largely exceeds the magma crystallization condition. Moreover, crystals in near-liquidus experiments may be small in size and strongly zoned, which may cause serious problems in analyses and data interpretation. Despite these advantages, it will be shown below that the reliability of $D^{mineral/L}$ estimated from natural peridotite samples is also limited by various factors. Consequently this kind of data set should be used with caution.

The calculated and experimental $D^{O/L}$ are reasonably consistent for the moderately incompatible elements, but they show significant differences for the elements more incompatible than Nd (fig. 4). As discussed previously, the LILE concentrations in olivine obtained by the ICP-MS mainly reflect those of fluid inclusions, it is obvious that calculated $D^{O/L}$ for the LILE has no meaning. In contrast, the effect of fluid inclusions on garnet is not significant so that the calculated $D^{G/L}$ should be of significance. This inference is supported by the consistency between calculated and experimental $D^{G/L}$ (fig. 4(c)). Nevertheless, the calculated $(D^{G/L})_{LREE}$ are lower than the experimental ones. This cannot be due to the effect of fluid inclusions on garnet (e.g. for Ba and Rb) because it would result rather in a higher calculated $(D^{G/L})_{LREE}$ relative to the experimental values. Zack et al.^[14] attributed this to the low equilibrium temperature (700°C) compared with the experimental condition (1400°C). It is noted that the $D^{G/L}$ data obtained by Zack et al. according to the garnet pyroxenites are very similar to those obtained in this paper according to the garnet peridotite (fig. 4). Since the latter has a higher equilibrium temperature (1150 °C) than the former (930 °C), it seems that the difference between calculated and experimental values cannot be fully accounted for by the temperature effect.

$D^{Opx/L}$ calculated according to the garnet peridotite is also consistent with the experimental data. However, $(Opx/Cpx)_{HREE}$ in the spinel peridotite are markedly lower than those in the garnet peridotite (fig. 4). Limited Opx/Cpx data for garnet peridotites in literature also reveal similar features. This cannot be due to the effect of fluid inclusion largely because the distribution of the MREE and HREE can be nicely accounted for by the crystal structure control theory (fig. 4(b))^[17]. Since the spinel and garnet peridotites are equilibrated at different P-T conditions, it is possible that the variation of D is P-T dependent. Further research in this direction will certainly help to understand the relationship between them.

5 Conclusion

1) There are at least three distinct components which contribute to the trace element budget of dry peridotites: (a) silicate minerals, particularly clinopyroxene and garnet. For example, 100% of the HREE, 60%—90% of the LREE and some incompatible elements such as Sr, Zr, Hf; (b) intergranular components in which Ba, Th, U, Nb, Ta, Sr contents represent 20%—60% of the bulk rock budget; (c) fluid inclusions which may contain some LILE. For example, Rb and Ba in inclusions may represent 20% of the bulk rock abundance of these elements. However, the role of fluid inclusions for other trace elements in the whole rock budget is negligible. Since the origin of intergranular components is unclear, the whole rock LILE composition alone cannot be used to discuss mantle processes. Otherwise it may result in biased inferences.

2) The distribution of the LILE between mineral phases cannot be used to infer the mineral-melt coefficients due to the ubiquitous presence of fluid inclusions in peridotitic minerals. It

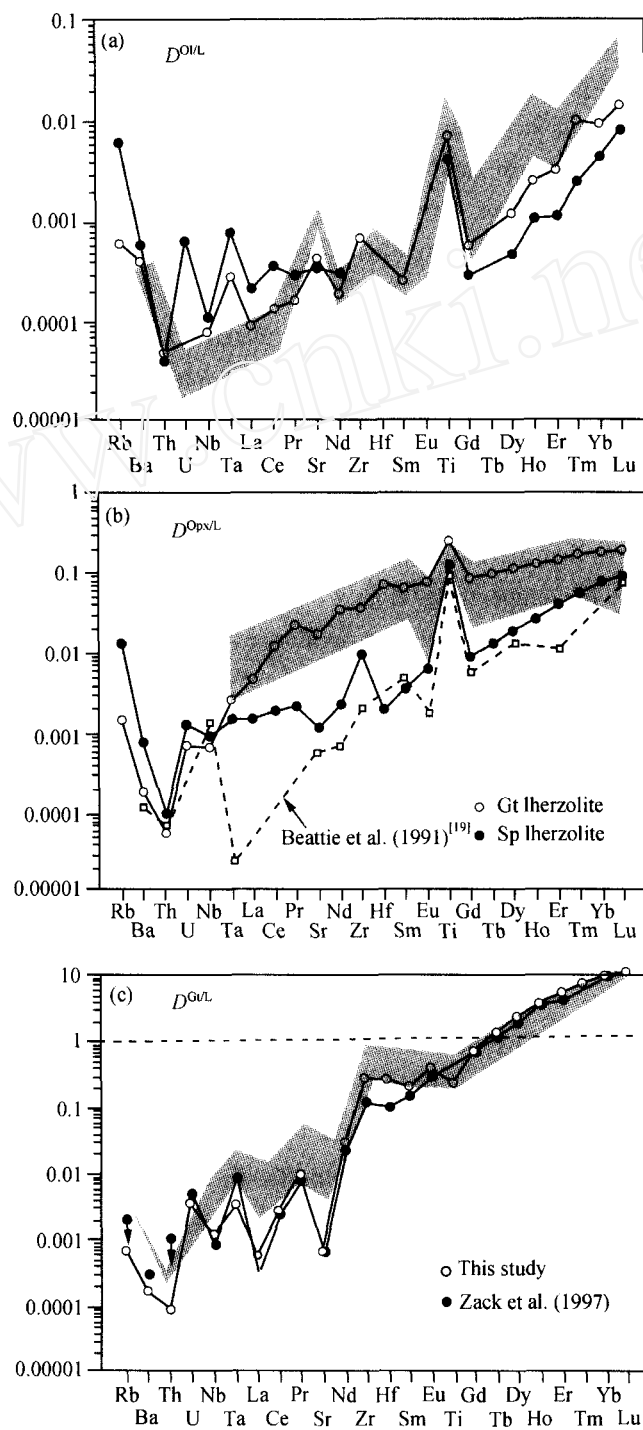


Fig. 4. Mineral-melt partition coefficients of trace elements estimated from mineral-mineral partition and $D^{Cp/L}$ ^[18]. (a) $D^{O/L}$; (b) $D^{Op/L}$; (c) $D^{G/L}$. The shaded area is the range of experimentally determined partition coefficients^[14].

is demonstrated that the LILE composition of olivine and orthopyroxene can be seriously affected by the presence of fluid inclusions, therefore the calculated $(D^{\text{Opx/L}})_{\text{LILE}}$ and $(D^{\text{Oli/L}})_{\text{LILE}}$ do not have any significance. Nevertheless, this method can be applied to the minerals (e.g. Gt) with relatively high trace element concentrations, because they are practically unaffected by fluid inclusions. There is a significant difference in $(\text{Opx/Cpx})_{\text{HREE}}$ ratio for spinel and garnet peridotites, suggesting a critical control of P-T condition on equilibrium coefficients, but the details of the mechanism deserve further investigation.

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