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Geochemistry of serpentinized peridotites from the Indus suture ophiolite in Swat, NW Pakistan

MOHAMMAD ARIF[1] & CHARLIE J. MOON[2]
[1]Department of Geology, University of Peshawar, Pakistan
[2]Department of Geology, University of Leicester, Leicester LE1 7RH, UK

ABSTRACT: *The Swat valley ophiolite predominantly consists of ultramafic rocks, which occur as lensoidal masses along the Main Mantle Thrust (MMT) between the Kohistan island arc and Indian plate. On the basis of their petrographic characteristics and modal mineralogy (abundant serpentine + subordinate amounts of bastitised orthopyroxene + variable proportions of olivine + accessory amounts of clinopyroxene + trace to accessory amounts of partly altered chrome spinel), most of the ultramafic rocks can be classified as spinel-, clinopyroxene-bearing harzburgite. Consistent with these mineralogical features, the mentioned rocks contain low to very low amounts of basaltic (Al_2O_3 and TiO_2), low concentration of the moderately incompatible (V, Sc, Zn and Ga) and high levels of moderately compatible to highly compatible components (Fe, Mn, Cr, Co, Ni and Mg). These major, minor and trace element geochemical characteristics suggest that the studied ultramafic rocks largely represent a residue of the upper mantle left after partial melting. A detailed comparison with residual rocks from elsewhere in the world shows that, prior to emplacement during the Cretaceous-Tertiary collision between the Indian plate and the Kohistan arc along the MMT, the source region of the studied rocks had suffered an intermediate degree (15 to 30 %) of partial melting.*

INTRODUCTION

Because of (1) analogy between oceanic dredged rocks and ophiolite samples and (2) a strong resemblance between the structure of ophiolite complexes and that proposed on geophysical grounds for the oceanic lithosphere, ophiolites are widely considered to be slices of oceanic crust and subjacent upper mantle (Cann, 1970). Therefore, ophilites are the only suitable source of direct information for knowing about the character and composition of the old oceanic lithosphere.

Good exposures of ultramafic rocks and other diagnostic lithological types with well-preseved oceanic features that characterise most of the world's known ophiolite sequences, occur in the area between the Mingora town and Lilauni village of Swat, NW Pakistan (Fig. 1). These rocks lie along the well-defined suture zone, i.e. the Main Mantle Thrust (MMT), which marks the collision of the Indian plate with the Kohistan island arc (Tahirkheli et al., 1979; Coward et al., 1986; Treloar et al., 1989). This important tectonic setting and ophiolitic character add to the petrologic significance of these rocks. Most of the petrogrpahic and mineralogical details of these rocks, particularly the ultramafic ones, are available in the form of both published and unpublished data (e.g., Chaudhry & Ashraf, 1986; Ashraf et al., 1989; Arif & Jan, 1993; Arif & Moon, 1994; 1996). Similarly, whole-rock analytical details regarding some of the altered equivalents of the ultramafic and mafic rocks have also been published (Barbieri et al., 1994; Arif & Moon, 1999). Similar details about the relatively less altered ultramafic rocks are, however, not available. Based on major, minor and trace element data, the current study aims at discussing geochemical characteriztion and petrogenesis of the serpentitnized peridotites from the Swat valley ophiolite.

REGIONAL TECTONICS AND GENERAL GEOLOGY

Three different types of rather loosely defined and tentatively demarcated melanges, i.e. blueschist melange, greenschsit melange and the ophiolitic melange, occur in the area between Mingora and Lilaunai (Kazmi et al., 1984). The lithological composition and local pesence of relict pillow structures strongly suggest that they are the remnants of the oceanic lithosphere between the Indian plate and the Kohistan island arc (Lawrence et al., 1989).

As a result of its continuous northward movement, the Indian plate collided with the rocks of the Kohistan island arc along the Main Mantle Thrust (MMT) in northern Pakistan. This collision, which most probably took place during the Early Tertiary (Treloar & Rex, 1990; DiPietro & Lawrence, 1991), and its associated processes of subduction and obduction are responsible for the metamorphism of rocks constituting the suture-associated melanges and their emplacement into present position.

The exposure of the ophiolitic melange is in the form of isolated lensoidal masses that are distributed within and along the frontal rocks of the Indian plate. The rocks constituting the ophiolitic melange include ultramafic and mafic plutonic rocks, different types of lavas and sedimentary rocks as well as plagiogranites and albitites. Like the blueschist and greeschist melanges, rocks of the ophiolitic melange are variably altered and metamorphosed.

Most of the ophiolite occurrences predominantly consist of ultramafic rocks. Although harzburgites enclosing small pods of Cr rich chromitite are by far the most abundant ultramafic rocks, minor amounts of dunite, lherzolite and websterite also occur in some of the ophiolite lenses. As multiple phases of pervasive alteration by fluids of differing composition have affected the rocks, their distinction in the field and, therefore, mapping as separate lithological units is not possible. These factors are also responsible for obscuring and obliterating most of the original textural features of the ultramafic rocks. Therefore, it is extremely difficult to distinguish these rocks into cumulate and tectonite (residual) varieties.

Hydartion leading to serpentinization is the most widespread process of alteration that has affected the studied ultramafic rocks. At least two episodes of serentinization are distinguishable. Besides, carbonate-alteration has also been involved in the mineralogical transformation of these rocks. A variable degree of alteration by this process has resulted in the formation of a variety of magnesite-rich assemblages, which host deposits of emerald that are being mined at different places in the area (Fig. 1). The occurrence of magensite veins that cut across serpentine matrix in some of the samples clearly shows that the carbonate-alteration commenced after serpentinization (Arif, 1994).

The occurrence of the gabbroic and volcanic rocks is rather rare and restricted to only a few of the ophiolite lenses (Fig. 1). Ranging in composition from basic to intermediate, most of the lavas display deformed pillow structures, however, amygdaloidal varieties are also present. Epidote, amphibole and chlorite are abundant, and plagioclase and pyroxene are correspondingly subordinate in both the gabbroic and volcanic rocks. This suggests that they, like the associated ultramafic rocks, are extensively altered and metamorphosed under the conditions of upper greenschist to lower amphibolite facies.

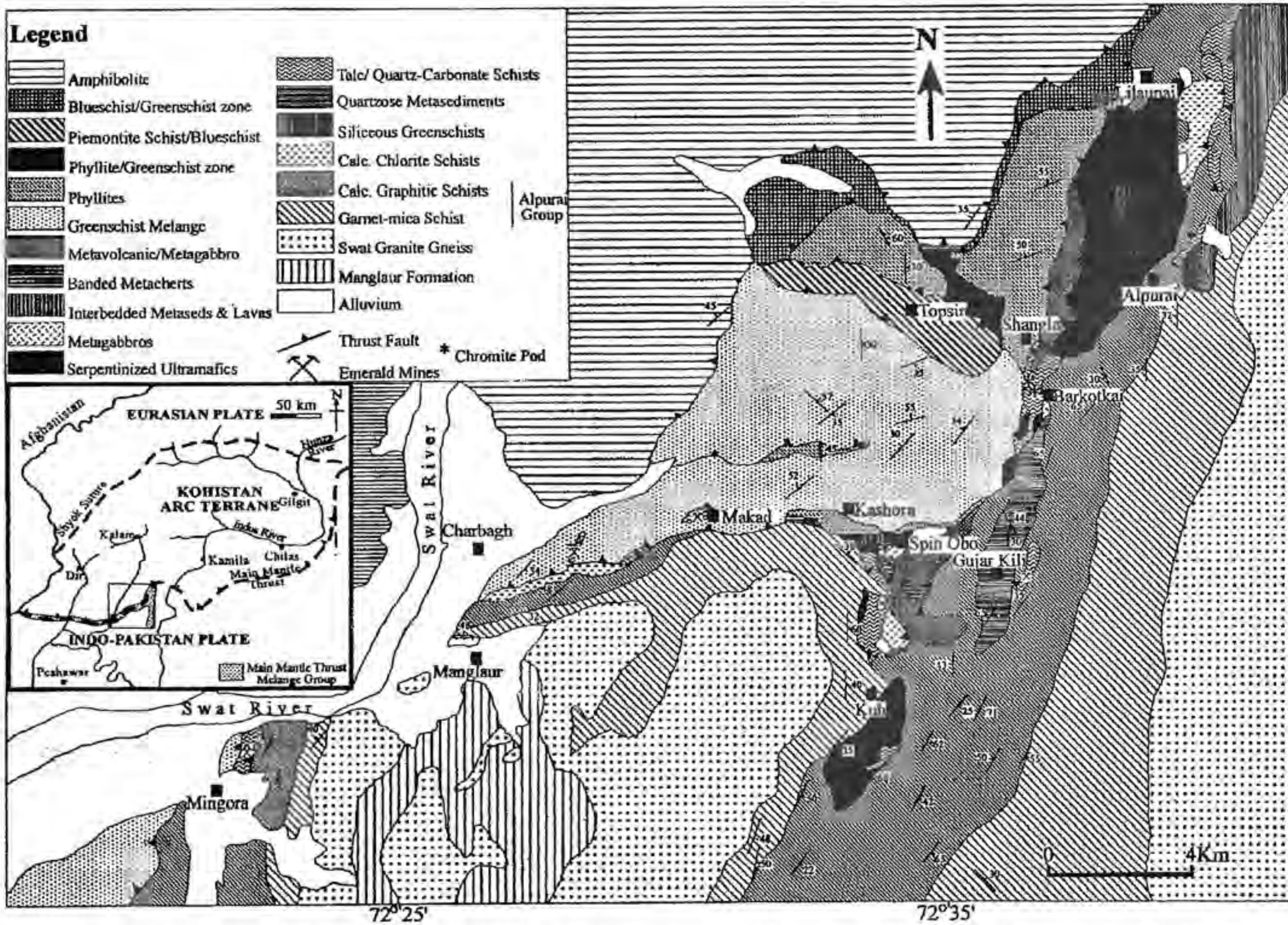

Fig. 1. Geological map of the Mingora-Lilaunai area, Swat, northwestern Pakistan (modified after Kazmi et al., 1984). The inset map shows general location of the study area.

The occurrence of ophiolitic sedimentary rocks is also rare. Only one of the ophiolite bodies contains a major exposure of such rocks (Fig. 1). The sedimentary lithologies include pelagic, calcareous and cherty varieties. The cherts are banded and locally maganiferous. Furthermore, some of the pelagic sediments are manganiferous.

SAMPLES AND ANALYTICAL TECHNIQUES

Eighty samples were collected from almost all the occurrences ,of the serpentinized ultramafic rocks in the area. After detailed mineralogical studies, 76 samples were selected for major, minor and trace element analyses. The major element analyses were performed on fusion beads (glass discs) whereas powder pellets were used to determine the contents of the minor and trace elements. The fusion beads were analysed on a Philips 1400 X-ray spectrometer and ARL 8420^{+} spectrometer; each equipped with a rhodium anode X-ray tube. The powder pellets were analysed on a Philips PW 1400/10 XRF spectrometer equipped with either a 3 kW rhodium anode tube or a tungsten anode tube. A set of international and internal standards was alternately run with each batch of samples to monitor and quantify the precision and accuracy of the instrument. The analytical results demonstrate a high degree of machine accuracy and precision (i.e. greater than 2 % at the 98 % confidence level) for all the major oxides as well as most of the minor and trace elements.

RESULTS

Petrography

As mentioned earlier, the ultramafic rocks of the Swat valley ophiolite are invariably serpentinized to varying degrees. As a result, most of them consist of abundant fine-grained serpentine probably after olivine, subordinate amounts of bastite pseudomorphs after orthopyroxene, and accessory amounts of clinopyroxene and partly to completely altered chrome spinel. Such a modal composition suggests that, prior to serpentinization, the majority of the studied rocks were spinel-, clinopyroxene-bearing harzburgites. In addition to the original magmatic clinopyroxene, some of the rocks also contain this phase as very fine prismatic crystals. Besides, variable amounts of fresh olivine occur as discrete grains in association and perfect textural equilibrium with serpentine as well as patches and veins within bastite. These peculiar modes of occurrence and diagnostic form coupled with the chemistry of both the olivine, i.e. low NiO and high MnO contents relative to Mg/ $(Mg+Fe^{2+})$ that ranges up to 0.985, and prismatic cliopyroxene [Mg/ $(Mg+Fe^{2+})$ ~ 0.98] suggest their formation during a prograde metamorphic process (Arif & Moon, 1996). Furthermore, the total lack of pseudomorphic (e.g., hourglass) textures in, and non-pseudomorphic character of, the serpentine itself suggest its development or re-equilibration under relatively high-temperature conditions.

Whole-rock chemistry

Representative chemical analyses of the rocks under investigation are listed in Table 1. Based on wt % Al_2O_3, the studied peridotitic rocks can be divided into five main groups: (1) Al_2O_3 <0.5; (2) Al_2O_3 >0.5 to <1.00; (3) Al_2O_3 >1.0 to <1.5; (4) Al_2O_3 >1.5 to <2.0; and (5) Al_2O_3 >2.0. Almost all the studied rocks fall into these categories. The purpose of this classification is to present maximum amount of the analytical data with greater clarity and, therefore, to facilitate geochemical characterisation and comparison.

The chemical characteristics of the studied rocks are compared with those of primitive mantle (Hartmann & Wedepohl,

1993) (Table 2). For this purpose, the average concentrations of the petrologically important components of each of the groups are normalized to the corresponding values of the primitive mantle. To avoid overcrowding and maintain clarity, the average normalized values of the different groups are plotted separately on different diagrams in Fig. 2. For further geochemical characterisation of the rocks under consideration, the primitive mantle-normalized patterns of rocks representing apparently similar geologic and tectonic settings from elsewhere in the world (Table 2) are also shown in Fig. 2.

TABLE 1. REPRESENTATIVE WHOLE-ROCK CHEMICAL ANALYSES OF THE INVESTIGATED PERIDOTITES

Sample	B25-III	B29-I	B31-III	L1-IV	L2-IV	L8-II	L19-III	Sp21-IV	Sp23-V	Sp25-III
Weight percent										
SiO_2	45.89	41.80	43.98	44.79	43.46	44.87	43.98	47.52	46.36	43.02
TiO_2	0.01	0.01	0.01	0.02	0.02	0.01	0.02	0.02	0.03	0.02
Al_2O_3	1.31	0.15	1.03	1.68	1.51	0.68	1.46	1.81	2.99	1.02
FeO*	5.26	8.05	6.20	6.60	7.74	6.51	8.08	7.46	7.64	6.70
MnO	0.11	0.12	0.20	0.08	0.11	0.11	0.14	0.08	0.12	0.12
MgO	45.85	47.35	44.67	46.25	44.48	46.34	43.93	40.60	41.61	47.69
CaO	0.56	0.46	2.43	0.13	1.13	0.83	0.95	1.45	0.15	0.14
Na_2O	0.29	<0.02	0.11	0.15	0.38	<0.02	0.28	0.26	0.16	0.17
P_2O_5	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.01
Total	99.17	97.83	98.64	99.63	98.72	99.25	98.71	99.13	98.96	98.77
LOI	10.66	5.17	5.67	9.17	7.15	9.19	8.08	12.27	11.26	6.94
Mg #	94.0	91.3	90.5	92.6	91.1	92.7	90.6	90.7	90.7	92.7
Parts per million										
Cr	2111	2163	2512	2306	2410	2038	2137	1814	1984	1837
Ni	1982	2308	2295	2028	2053	2194	2206	2357	2304	2366
Co	62	87	83	57	76	63	82	72	73	80
V	29	18	33	29	39	27	37	37	39	20
Zn	28	24	34	22	25	21	31	46	44	20
Ti	66	60	78	138	102	62	90	108	204	120
Sc	5	0	10	7	7	7	9	9	4	5
Ga	<2	2	<2	3	3	2	3	3	5	3

Samples designated as B, L and Sp represent respectively the bodies of ultramafic rocks in the Barkotkai, Lilaunai-Alpurai and Spin Obo-Kuh areas (Fig. 1). The roman numbers refer to the rock groups distinguished on the basis of Al_2O_3.

All the analyses were carried out by XRF spectrometry. The oxide compositions were determined on volatile-free basis using fusion beads; analyses for the trace elements were performed on powder pellets.

*Total iron recalculated as FeO

LOI = weight loss on ignition (%)

Mg # = 100 x Mg/(Mg + ΣFe)

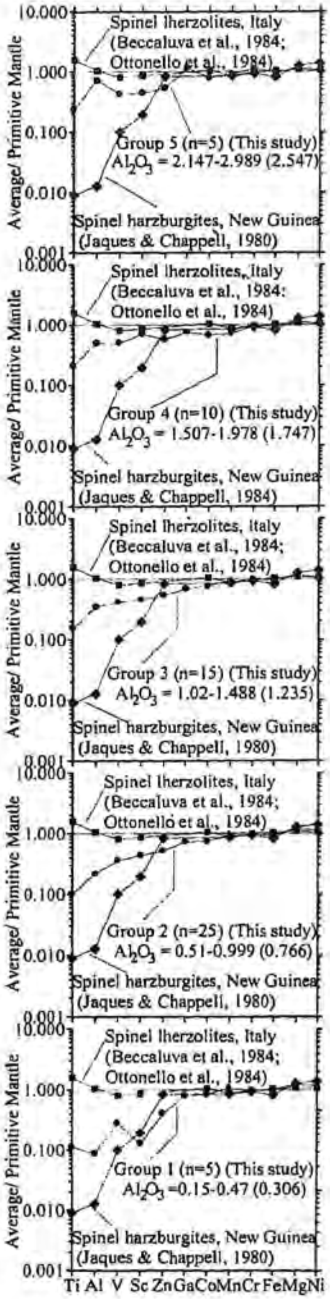

Fig. 2. Primitive mantle-normalized geochemical patterns of the petrologically important elements in the rocks studied (circles) and their comparison with such patterns for rocks of similar tectonic setting from northern Apennine (squares) and Papuan (diamonds) ophiolite belts. The values used for normalization and comparison are listed in Table 2.

TABLE 2. CHEMICAL COMPOSITION OF THE SAMPLES USED FOR NORMALIZATION AND COMPARISON

Sample	(1)	(2)	(3)
	Weight percent		
TiO_2	0.12	0.20	<0.002
Al_2O_3	3.53	3.69	0.05
Fe_2O_3	9.17	9.45	7.54
MnO	0.14	0.12	0.12
MgO	37.99	41.15	48.11
	Parts per million		
Ti	743	1175	7
Cr	2500	2442	2305
Ni	1990	2112	2758
Co	104	109	n. a.
V	85	68	8.5
Zn	51	------	43
Sc	16.7	14	3.3
Ga	3.4	------	n. a.

(1) Estimated composition of the primitive mantle (Table 2 in Hartmann and Wedepohl, 1993).

(2) Average composition of ultramafic tectonites (spinel lherzolites) from the northern Apennine ophiolites based on the data of five least serpentinized samples (Li8, Li9, 14R, 15R and 16R) representing both the internal and external Ligurides (Table 1 in Beccaluva et al., 1984, and Table 2 in Ottonello et al., 1984b).

(3) Average composition of ultramafic tectonites (spinel harzburgites) from the Papuan ultramafic belt, New Guinea (mean of the first four analyses listed in Table 3; Jaques and Chappell, 1980).

n.a. = not analysed

DISCUSSION AND CONCLUSIONS

Effects of serpentinization

Virtually all the studied samples are serpentinized to varying degrees. Therefore, a knowledge of the nature of serpentinization (whether isochemical or metasomatic) is essential before the bulk rock compositions could be used for any geochemical characterization and/or petrogenetic discussion. Although being largely the result of hydration (see, for example, Komor et al., 1985), serpentinization may involve addition and/or removal of some non-volatile components also. For example, the mass balance calculations of bulk chemistry by Labotka and Albee (1979) show that serpentinization may lead to the addition of SiO_2. Besides, the evidence for rodingitization (the by-product of serpentinization; Coleman, 1977) from many areas of serpentinized ultramafic rocks suggests that loss of CaO may accompany the process of serpentinization.

The following features indicate that SiO_2 was probably added to the studied rocks during their serpentinization:

(1) Mineralogical studies, as summarised above, show that the majority of the rocks are most probably harzburgites and as such originally had an olivine: pyroxene ratio >1, and that serpentinization was largely at the expense of olivine. Under such circumstances, MgO is released (because the MgO: SiO_2 of olivine is more than that of serpentine) and, therefore, a Mg-rich, SiO_2-free phase (either brucite, or magnesite if CO_2 is available) is expected to accompany the formation of serpentine. But as almost all the rocks under discussion are free of such a phase, the implication is that SiO_2 was added to the rocks due to which all the original olivine was converted to serpentine thereby eliminating the necessity of brucite/ magnesite formation.

(2) The SiO_2 content of some of the rocks exceeds that of a typical peridotite (~ 45 wt %) (Table 1).

(3) The SiO_2 content of the studied rocks shows a positive correlation with the degree of serpentinization (indicated by the loss on ignition values) (Table 1).

Most of the other elements were probably not affected by serpentinization because the concentration of none of them shows any relationship with the values of loss on ignition. However, the very low concentration of CaO in many of the rocks and the local occurrence of rodingites in the area suggest that some of this oxide might have been lost during serpentinization. That is why both CaO and SiO_2 are excluded from the list of components used for the geochemical characterization of the rocks under discussion.

Geochemical characterization

Geological and tectonic setting, massive character (absence of layering), modal mineralogy and an overall mineral-chemical as well as geochemical homogeneity (the individual bodies almost entirely consist of spinel-, clinopyroxene-bearing harzburgites), all show that most of the studied ultramafic rocks could be residual in origin. That is why their geochemical features are compared with, and discussed in the light of, those of the primitive mantle and the variably depleted peridotites (ultramafic tectonites) from elsewhere (Table 2; Fig. 2).

As also supported by experimental work (Mysen & Kushiro, 1977; Jaques & Green, 1980), the modal mineralogy, phase, and therefore whole rock, chemistry of residual peridotites vary systematically with the degree of partial melting (Dick, 1977; Dick et al., 1984; Michael & Bonatti, 1985). Thus peridotites containing a relatively high proportion of Al- rich clinopyroxene (with significant amounts of Na and Ti) associated with Cr-poor spinel are regarded to be only slightly depleted, i.e. they represent mantle which has undergone a low degree (<15 %) of partial melting. The Ligurian spinel lherzolites (Beccaluva et al., 1984; Ottonello et al., 1984a, b) are one of the examples of such peridotites. On the other hand, peridotites having Al-poor, low to very low modal clinopyroxene, and accessory amounts of Cr-rich spinel are believed to be highly depleted, i.e. they represent mantle, which has suffered a high degree (>30 %) of partial melting. The Papuan spinel harzburgites, New Guinea (England & Davies, 1973; Jaques & Chappell, 1980) are the typical examples of such highly depleted peridotites. As the mineralogical and geochemical characteristics of peridotites from the Yakuno ophiolite, south-west Japan are intermediate between the Ligurian lherzolites and Papuan harzburgites, they are considered as representative of upper mantle which was moderately depleted, i.e. had experienced an intermediate degree (15-30 %) of partial melting (Ishiwatari, 1985).

The studied rocks contain low to very low amounts of such incompatible elements as TiO_2 and Al_2O_3. Concentrations of V, Sc, Zn and Ga are also low (Table 1; Fig. 2). On the other hand, the content of highly and moderately compatible elements (Ni, Mg, Cr, Co, Fe and Mn) is more or less uniformly high. Such geochemical characteristics are typical of residual peridotites and, therefore, can best be explained only by invoking the phenomenon of partial melting (Loubet & Allègre, 1979; 1982; Jaques & Chappell, 1980; Ottonello et al., 1984a, b; Frey et al., 1985; Hartmann & Wedepohl, 1993).

The average concentrations of the highly compatible (Mg and Ni) and moderately compatible (e.g. Cr, Co and Fe) elements in virtually all the groups of the studied samples are only slightly different from those in the primitive mantle and residual peridotites from other areas (Fig. 2). However, the abundance of the moderately incompatible (basaltic) components (Ti, Al, Sc, V, Zn and Ga) in virtually all of the studied rocks is markedly low relative to that in the upper mantle. Due to very high variability, the concentrations of Ti and Al differ significantly even from one member to another within the individual groups. However, none of the samples has the abundance of any of these elements equal or even comparable to that in the primitive mantle. The relatively incompatible element parts of the average geochemical patterns of the studied peridotites are intermediate between that exhibited by the least depleted (Ligurian spinel lherzolite) and highly depleted tectonites (Papuan harzburgite). This suggests that the studied peridotites represent residual upper mantle that had suffered an intermediate degree (>15 to <30 %) of partial melting.

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