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<b>Citation</b>	<b>Canadian Journal Of Earth Sciences, 1995, v. 32 n. 7, p. 1046-1057</b>
<b>Issued Date</b>	<b>1995</b>
<b>URL</b>	<b><a href="http://hdl.handle.net/10722/42089">http://hdl.handle.net/10722/42089</a></b>
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# Multiple origins for mantle harzburgites: examples from the Lewis Hills, Bay of Islands ophiolite, Newfoundland

Stephen J. Edwards and John Malpas

**Abstract:** Harzburgite is a common rock type in ophiolites and alpine peridotites. It is considered to be typical of a residual mantle mineralogy, i.e., material left behind after periods of extensive mantle melting and melt–rock and fluid–rock interactions that produce a variety of basaltic melts. The processes by which these melts and residua are produced are complicated; therefore, to fully understand them, it is necessary to undertake detailed and integrated field, petrographic, and geochemical studies of large exposures of mantle material as part of the investigative process. Such a study in the Bay of Islands ophiolite exposed in the Lewis Hills of Newfoundland has enabled the identification of four major types of harzburgite, which represent examples of a complete spectrum of this rock type. Depleted, residual harzburgite and associated dunite, with positive-sloping rare earth element patterns, may develop U-shaped rare earth element patterns and a visible orthopyroxene enrichment by the introduction of a component of high-Mg, quartz-normative melt, or a hydrous fluid component with a high Si/Al ratio. Conversely, U-shaped rare earth element patterns and apparent orthopyroxene depletion may occur by the addition of low-Si/Al, hydrous fluid. Such enrichments and depletions of orthopyroxene by solution–precipitation reactions may result not only in the variety of harzburgite types, which on partial melting might produce a range of melt products, but also in fronts of harzburgite migrating through the mantle.

**Résumé :** L'harzburgite est un type de roche fréquent dans les ophiolites et les péridotites alpines. Elle est considérée comme étant typique du résidu minéralogique du manteau, c.-à-d., le matériel laissé par soustraction après les phases de fusion extensive et les interactions du magma et des roches avec les fluides engendrant une variété de magmas basaltiques. Les processus impliqués dans la production des magmas et des résidus réfractaires sont complexes; par conséquent, pour bien les comprendre, il est nécessaire d'entreprendre des études détaillées et intégrées de leur géochimie, pétrographie, et aussi sur le terrain dans les régions où affleure abondamment du matériel mantellique faisant partie de l'étude. Les travaux effectués sur l'ophiolite de Bay of Islands, exposée dans les collines Lewis de Terre-Neuve, ont permis de reconnaître quatre types principaux d'harzburgite, lesquels représentent un spectre complet de ce type de roche. Le résidu appauvri d'harzburgite et la dunite associée, avec leurs diagrammes de terres rares à pente positive, peuvent aussi développer des diagrammes de terres rares en forme de U, et un enrichissement en orthopyroxène aisé à reconnaître, causé par la venue d'un composant magmatique à quartz normatif riche en Mg, ou d'un composant de fluide hydraté avec un rapport Si/Al élevé. Réciproquement, les diagrammes de terres rares en forme de U et un appauvrissement apparent de l'orthopyroxène peuvent être une réponse à l'addition d'un fluide hydraté avec un rapport Si/Al faible. De tels enrichissements et appauvrissements en orthopyroxène causés par des réactions de dissolution–précipitation peuvent apparaître, non seulement dans les divers types d'harzburgites, qui par fusion partielle peuvent créer une série de produits magmatiques, mais aussi dans les zones frontales d'une harzburgite migrant au travers le manteau.

[Traduit par la rédaction]

Received August 5, 1994. Accepted February 22, 1995.

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## Introduction

Our present understanding of mantle mineralogy and the processes which generate basaltic melts in the mantle suggests that a significant portion of the mantle beneath oceanic crust, including that of island arcs, is harzburgite. Such is born out by the preponderance of harzburgite in the mantle sections of certain well-preserved ophiolite suites that are thought to represent ancient oceanic lithosphere. Indeed, in

some of these ophiolites (e.g., Troodos), the dominance of harzburgite has led to their classification as harzburgite-type, or, where present together with lherzolite (e.g., Bay of Islands), as lherzolite–harzburgite type (Boudier and Nicolas 1985). These mantle sections can be used to provide a regional picture of a variety of mantle processes, including those involved in the genesis of harzburgite.

Although for many years it has been argued that harzburgites in ophiolites represent depleted, refractory residues derived by partial melting and melt removal events in the mantle (e.g., Moores 1969; Himmelberg and Loney 1973; Malpas 1976), this simple relationship is complicated by the U-shaped rare earth element (REE) patterns of many refractory harzburgites and the light rare earth element (LREE)-depleted patterns of associated crustal sequence basaltic rocks (Suen et al. 1979; Pallister and Knight 1981). To preserve the concept of a simple parent–daughter relationship between residual mantle rocks and crustal rocks, Prinzhofer and Allègre (1985) proposed the sequential integrated disequilibrium melting model. In this model, melting is envisaged to occur under disequilibrium conditions in an upwelling mantle diapir. The rising peridotite melts sequentially through a range of stability fields, garnet, spinel, and plagioclase, with resultant enrichment of LREEs in the residue as garnet moved into the melt. This model, however, fails to explain the wide occurrence of peridotites, including many harzburgites, which exhibit relative LREE depletion.

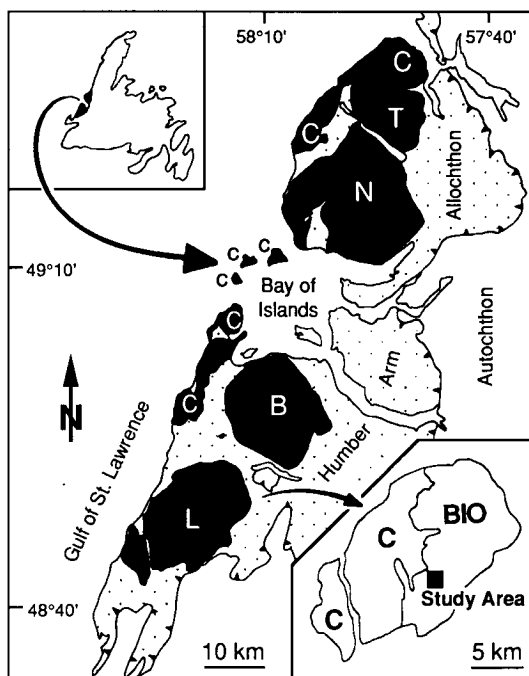
Indeed, it is apparent that there are harzburgites and harzburgites. Some harzburgites may simply be residual from partial melting, but many now appear to be products of partial melting and coupled or subsequent melt–peridotite interaction (Stosch and Seck 1980; Edwards 1990, 1991; Bodinier et al. 1991; Kelemen et al. 1992). These interactions may produce the LREE enrichment seen in many harzburgites, either by introduction of a LREE-enriched component into a LREE-depleted residue (Frey and Green 1974), or by ion exchange between melt or fluid and peridotite (Navon and Stolper 1987; Bodinier et al. 1990, 1991; Vasseur et al. 1991; Takazawa et al. 1992).

In this paper, we report results from an integrated field, petrographic, and geochemical study of mantle samples taken from the Springers Hill area of the Lewis Hills, Bay of Islands ophiolite, Newfoundland. This has permitted definition of four types of harzburgite as members of a harzburgite continuum. Interpretation of the genesis of these harzburgite types is based on field, petrographic, and mineral chemical data, which, in turn, are used to put whole-rock geochemical data, especially REE data, in context. Consequently, some of the processes invoked to explain REE patterns in harzburgite in the Springers Hill area are somewhat different from new models based more on geochemical modelling (e.g., Bodinier et al. 1991; Kelemen et al. 1992; Takazawa et al. 1992).

## Geological setting

The Bay of Islands ophiolite lies to the east of the Coastal Complex in the highest structural slice of the Humber Arm Allochthon in western Newfoundland (Fig. 1). The Coastal Complex is  $505 \pm 3$  Ma and dominated by arc-related ultramafic and mafic crustal rocks; the Bay of Islands ophiolite

**Fig. 1.** Generalized geological map of the Humber Arm Allochthon, western Newfoundland, Canada, after Smith (1958), Williams (1973), Karson (1979), and Williams and Cawood (1989). The Bay of Islands ophiolite comprises Table Mountain (T), North Arm Mountain (N), Blow Me Down Mountain (B), and the eastern part of the Lewis Hills (L) marked BIO. C, Coastal Complex. Barbs are on the upper thrust slice.



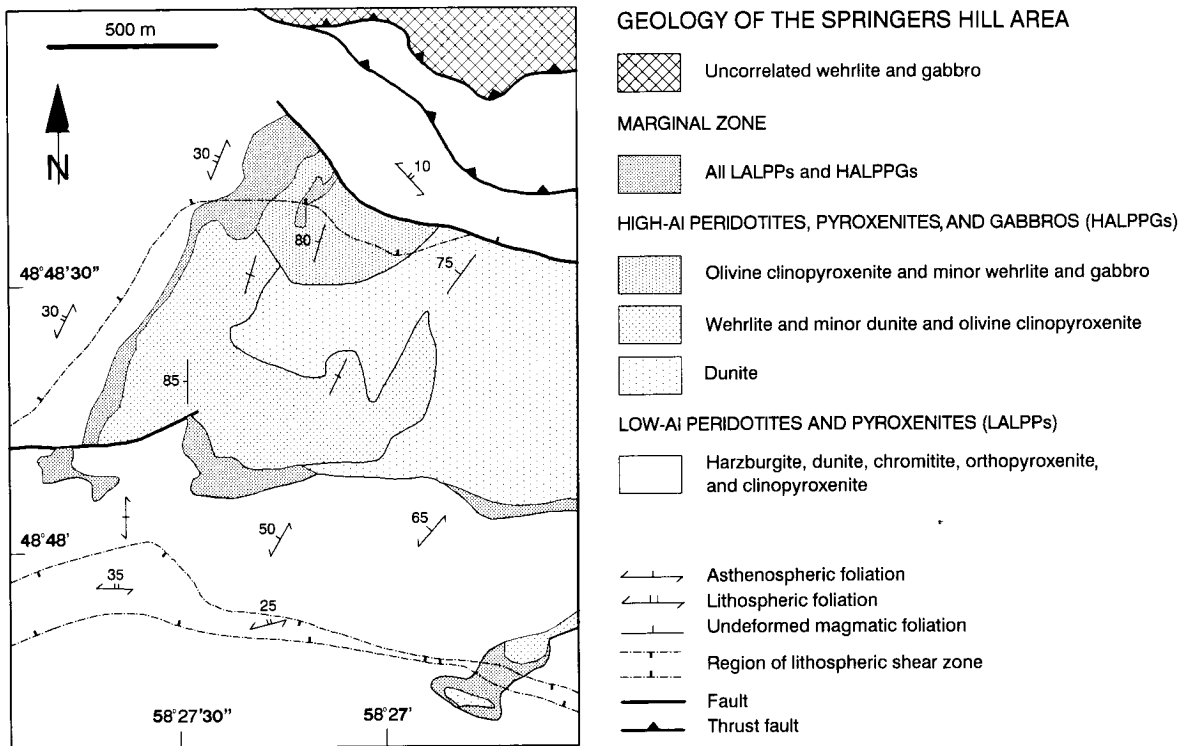
is  $484 \pm 5$  Ma and comprises suprasubduction-zone ultramafic and mafic mantle and crustal rocks (Jenner et al. 1991). The Bay of Islands ophiolite formed at a spreading centre and was later thrust onto the Coastal Complex along a fracture zone that originally separated the latter from the former (Casey and Dewey 1984; Cawood and Suhr 1992).

## Mantle section of the Bay of Islands ophiolite

The Bay of Islands ophiolite is a well-preserved, complete ophiolite that provides extensive exposure of mantle peridotites in four massifs: Table Mountain, North Arm Mountain, Blow Me Down Mountain, and Lewis Hills (Fig. 1). The lithology of the mantle section varies from north to south. The Table Mountain section comprises harzburgite, lherzolite, and dunite, which is in contrast to the highly refractory and depleted nature of the Lewis Hills section of mainly harzburgite and dunite. In all massifs, the peridotites record two distinct tectonic events (Suhr 1993). The earliest is a high-temperature – low-stress flow history relating to a phase of oceanic spreading (here referred to as asthenospheric deformation; temperature  $> 1000^\circ\text{C}$ ). This is variably overprinted by a lower temperature – high-stress flow history resulting from intraoceanic thrusting and obduction of the Bay of Islands ophiolite (here referred to as lithospheric deformation; temperature  $< 1000^\circ\text{C}$ ).

The mantle peridotites in the Lewis Hills preserve numerous magmatic (large intrusions, dykes, veins, and interstitial melt and fluid phases) and structural components of the

**Fig. 2.** Geological map of the Springers Hill area in the eastern part of the Lewis Hills. Residual and type I orthopyroxenite-bearing harzburgites are part of the low-Al peridotites and pyroxenites (LALPPs) formed in the asthenosphere. Type II orthopyroxenite-bearing and amphibole harzburgites, type II orthopyroxenites, and amphibole dunites are contemporaneous with the high-Al peridotites, pyroxenites, and gabbros (HALPPGs) and lithospheric deformation, and occur mainly in the marginal zone and lithospheric shear zones.



spreading and thrusting phases, as they developed at or close to the ridge during spreading and lay adjacent to the site of nucleation of intraoceanic thrusting during the initiation of obduction (Suhr 1992; Cawood and Suhr 1992). Nowhere are these events better exemplified than in the unique exposure in the Springers Hill area of the Lewis Hills.

### Springers Hill area

The Springers Hill area occupies the western extent of the Bay of Islands ophiolite in the Lewis Hills (Fig. 1). The section of ophiolitic mantle exposed in the area is harzburgite-type, in that a depleted, refractory harzburgite–dunite mixture hosts a large wehrlitic body (Dunsworth et al. 1986). More precisely, Edwards (1991) has divided the area into components that formed under asthenospheric and lithospheric conditions (Fig. 2). The asthenospheric component is represented by low-Al peridotites and pyroxenites (LALPPs), characterized by Cr-spinel with  $\text{Cr}\# = 66\text{--}90$  ( $\text{Cr}\# = 100\text{Cr}/(\text{Cr} + \text{Al})$ ). This component comprises harzburgite, which hosts dykes, veins, and irregular bodies of dunite and chromitite and dykes and veins of orthopyroxenite (here referred to as type I orthopyroxenite), and clinopyroxenite. The lithospheric component postdates the formation of the asthenospheric component and is represented by Ca-amphibole-bearing, high-Al dunites, wehrlites, clinopyroxenites, and gabbros (HALPPGs), characterized by spinel with  $\text{Cr}\# = 24\text{--}35$ . In the absence of faults, contacts between LALPPs and HALPPGs are normally defined by the marginal zone, which contains all LALPPs and HALPPGs

and abundant evidence for interaction of melt and fluid with LALPPs. Also in this zone and lithospheric shear zones in LALPPs are a second set of amphibole dunites (here specifically referred to as amphibole dunites) and orthopyroxenite veins (type II orthopyroxenites). These have compositions between LALPPs and HALPPGs, but both are contemporaneous with the HALPPGs, i.e., they belong to the lithospheric component.

The field relations, mineralogy, and geochemistry of the asthenospheric and lithospheric components led Edwards (1991) to propose two distinct episodes in the evolution of the Springers Hill area (Fig. 3). The asthenospheric component is considered to have formed in an upwelling zone of asthenosphere in a suprasubduction-zone mantle wedge. The lithospheric component is also considered to have formed in a suprasubduction-zone environment, perhaps during initial displacement of the Bay of Islands ophiolite, when LALPPs (high Si/Al) were metasomatized by LREE-depleted, low-Si/Al, fluid-bearing basaltic melts derived from the underthrust slab of oceanic lithosphere. The melt-dominated metasomatites are the HALPPGs, and the fluid-dominated metasomatites are amphibole dunites and type II orthopyroxenites.

### Harzburgite types

Olivine–orthopyroxene assemblages in the Springers Hill area cover a range from orthopyroxene-bearing dunites to orthopyroxenites. Harzburgites *sensu stricto* have a modal compo-

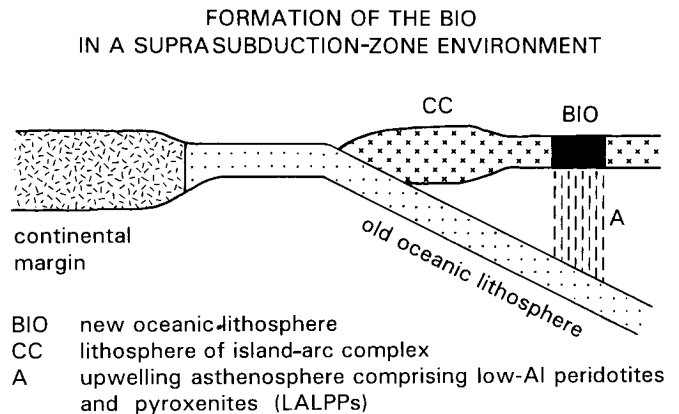
sition of 50–90% olivine, 10–50% orthopyroxene, <2% Cr-spinel, common, but modally insignificant, Ca-amphibole, and very rare clinopyroxene (Table 1). Although we stress that there is a continuum of harzburgite lithologies in the area, this is conveniently divided into four generic types: residual, type I orthopyroxenite bearing, type II orthopyroxenite bearing, and amphibole bearing. The diagnostic textural and mineralogical features of each type of harzburgite are summarized in Table 1 and Fig. 4.

Residual harzburgites, part of the asthenospheric component (LALPPs), are the oldest rock type in the area, host all other lithologies, and grade in places into low-Al dunite. They have a high-temperature porphyroclastic texture indicative of deformation during asthenospheric flow. In relatively low strain domains, the texture of spinel suggests partial melting and recrystallization of pyroxene (Fig. 4a). Minute traces of interstitial amphibole, which clearly postdate deformation, have been observed in the harzburgite matrix. Field relations suggest these interstitial phases represent a fluid-dominated component added to the harzburgite during formation of the HALPPGs.

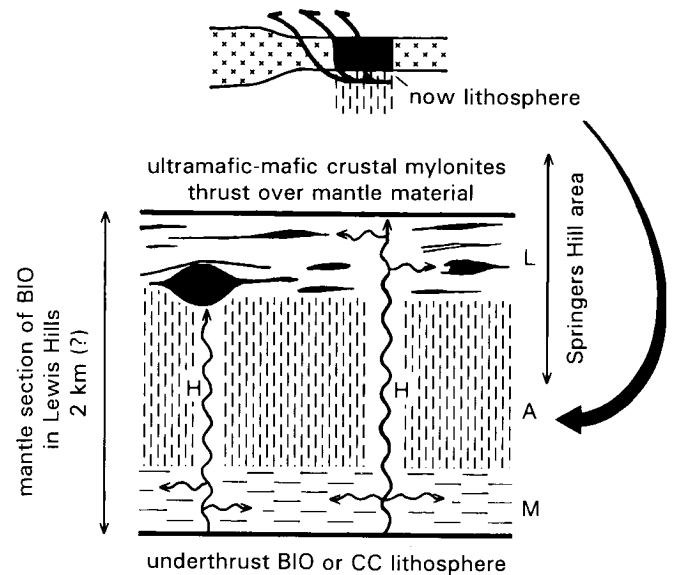
Type I orthopyroxenite-bearing harzburgites are part of the asthenospheric component, but postdate the residual harzburgites and associated dunites in which they are hosted. They are rich in orthopyroxene and are most obvious at margins or terminations of dykes of type I orthopyroxenite, or where such dykes are stretched, boudinaged, and highly disaggregated (Fig. 4b). It is rare to find type I orthopyroxenite hosted in anything but residual and type I orthopyroxenite-bearing harzburgites. Many of the petrographic features of the type I orthopyroxenite-bearing harzburgites are similar to those of the residual harzburgites, with the obvious exceptions of much higher concentrations of orthopyroxene and melt impregnation textures exhibited by orthopyroxene in zones of low strain (Fig. 4c). It has not been possible to estimate how much of the orthopyroxene enrichment in type I orthopyroxene-bearing harzburgite resulted from mechanical mixing of type I orthopyroxenite with residual harzburgite and associated dunite, or how much was due to precipitation of interstitial orthopyroxene from melt that impregnated the peridotite host.

Although type II orthopyroxenite-bearing and amphibole harzburgites are contemporaneous with the HALPPGs, many of their features are reminiscent of the LALPPs. These harzburgites are most common along, or are rooted in, reaction zones between LALPPs and HALPPGs and in lithospheric shear zones contemporaneous with the development of HALPPGs. Amphibole harzburgites clearly preserve features of residual and type I orthopyroxenite-bearing harzburgites, but orthopyroxene shows evidence of fluid-induced incongruent breakdown to olivine and minor spinel (Fig. 4d). Similarly, spinel apparently recrystallized in the presence of fluid and contains inclusions of edenitic amphibole and phlogopite (Fig. 4e). Where orthopyroxene is completely reacted out, amphibole harzburgite grades into amphibole dunite. Type II orthopyroxenite-bearing harzburgites occur where type II orthopyroxenite forms veins and networks in low-Al dunite (Fig. 4f), thereby preserving many features of the original low-Al dunite, although Cr-spinel associated with the orthopyroxenite is in places recrystallized. The type II orthopyroxenite-bearing harzburgites have been observed

**Fig. 3.** Unscaled schematic cross sections constructed from interpretations of Edwards (1991) and Cawood and Suhr (1992), which show formation of the Bay of Islands ophiolite (BIO) in a suprasubduction-zone environment, and displacement of the ophiolite by intraoceanic thrusting during the initial stages of obduction. Residual and type I orthopyroxenite-bearing harzburgites are part of A, whereas M, H, and L all formed contemporaneously with intraoceanic thrusting. The metasomatites in L are high-Al peridotites, pyroxenites, and gabbros (HALPPGs), amphibole dunites, type II orthopyroxenites, and type II orthopyroxenite-bearing and amphibole harzburgites. CC, coastal complex.



INITIAL DISPLACEMENT OF THE BIO



- M lithospheric ultramafic mylonites  
H hydrous, low-Si/Al basaltic melts originating in underthrust lithosphere  
A preserved asthenospheric component, which is part of the lithosphere at this stage  
L lithospheric mylonites containing isolated patches of component A and abundant, variably deformed metasomatites (black)

**Table 1.** Composition of types of harzburgite.

	Residual	Type I orthopyroxenite- bearing	Amphibole- bearing	Type II orthopyroxenite- bearing
Olivine <sup>a</sup>	84	54	90	63
Mg# <sup>b</sup>	91.4–91.9	91.2–91.3	91.0–91.5	91.4–91.7
Orthopyroxene	14	44	8	34
Mg#	91.7–92.0	91.7–92.2	91.7–91.9	91.7–92.0
Al <sub>2</sub> O <sub>3</sub> (wt. %)	0.82–0.85	0.32–0.88	0.50–1.40	0.55–0.83
Clinopyroxene	None	None	Trace	Trace
Cr-spinel	1–2	1–2	<1	<1
Cr# <sup>c</sup>	75.8–77.8	73.4–76.7	58.6–75.3	67.2–67.9
Ca-amphibole	Trace	None	<1 Tremolite, edenite	1–2 Tremolite, pargasite
Phlogopite	None	None	Trace	None
SiO <sub>2</sub> (wt. %)	38.30	40.00	35.20	40.70
Al <sub>2</sub> O <sub>3</sub>	0.18	0.36	0.23	0.29
Fe <sub>2</sub> O <sub>3</sub>	4.13	2.41	3.02	3.50
FeO	3.02	4.43	4.35	3.42
MnO	0.10	0.12	0.11	0.11
MgO	40.23	40.55	42.25	39.52
CaO	0.26	0.80	0.40	0.34
Na <sub>2</sub> O	0.02	0.02	0.02	0.14
P <sub>2</sub> O <sub>5</sub>	0.01	0.00	0.00	0.05
LOI	13.93	11.00	14.51	12.45
Total	100.18	99.69	100.09	100.52
Mg#	91.4	91.6	91.4	91.5
CaO/Al <sub>2</sub> O <sub>3</sub>	1.4	2.2	1.7	1.2
La (ppb)	3.2	4.1	21	1.7
Ce	8.5	7.8	44	3.2
Pr	1.3	0.78	5.5	(0.36)
Nd	7.5	5.1	24	(2.6)
Sm	(1.9)	(1.11)	5.9	(1.21)
Eu	1.0	0.51	2.5	(0.10)
Gd	3.7	2.9	7.2	(1.0)
Tb	0.61	0.55	1.55	(0.31)
Dy	4.5	5.2	10.6	3.1
Ho	1.03	1.40	2.0	0.68
Er	3.06	5.9	7.1	3.65
Tm	0.43	1.36	1.16	0.61
Yb	4.3	11.1	7.9	5.0
Lu	0.75	2.05	1.52	1.07

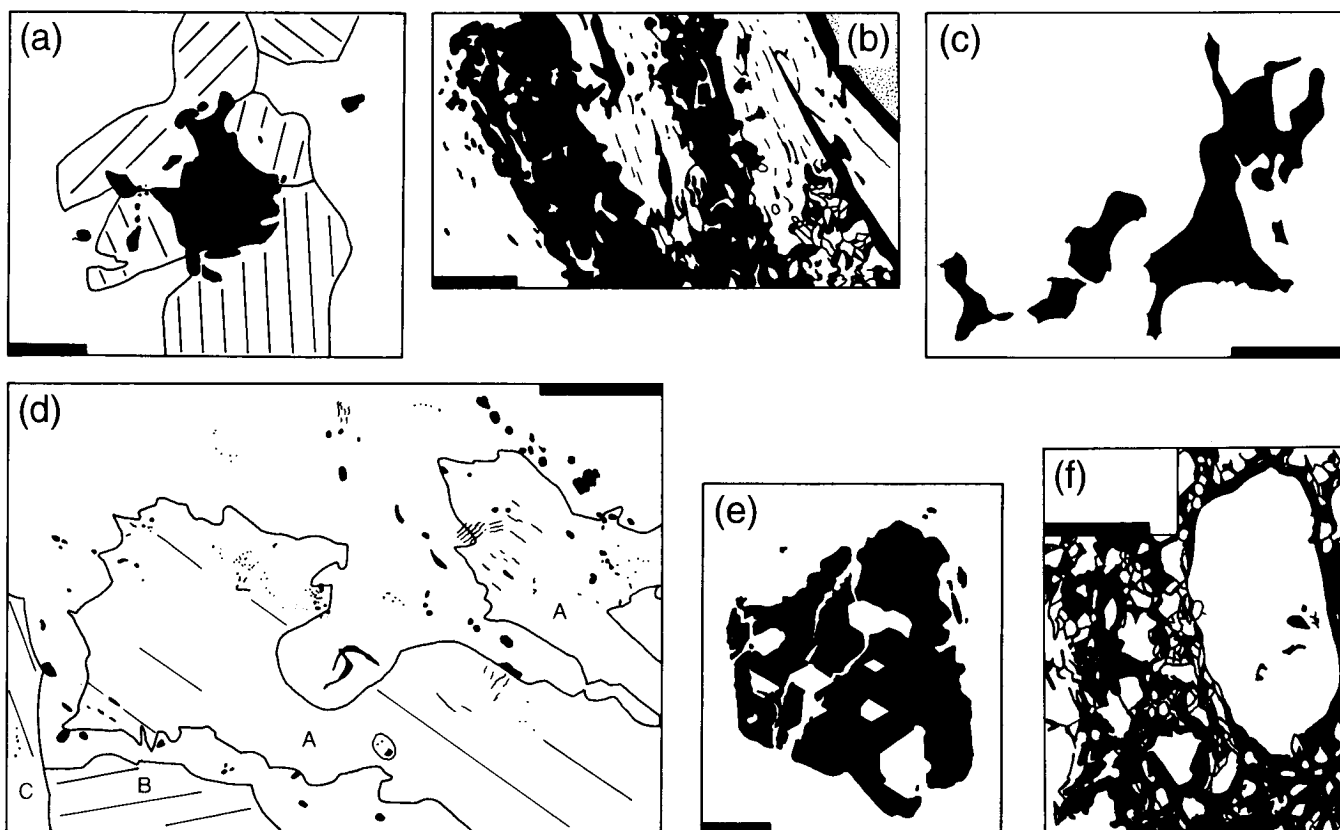
**Notes:** Mineral compositions are ranges of analyses of cores of grains. For whole-rock samples K<sub>2</sub>O = 0.00 wt.%, TiO<sub>2</sub> < 0.02 wt.%. LOI, loss on ignition. Concentrations of REEs are on an as-received basis and no correction has been made for LOI. Concentrations of REEs which either lie close to the limit of detection or may be affected by the blank are placed in parentheses.

<sup>a</sup>Modal compositions are in percent and are approximate because samples are heterogeneous and estimation was visual.

<sup>b</sup>Mg# = 100Mg/(Mg + Fe<sup>2+</sup>).

<sup>c</sup>Cr# = 100Cr/(Cr + Al).

**Fig. 4.** Diagnostic textures in types of harzburgite from the Springers Hill area. (a) Cr-spinel (black) in an aggregate of porphyroclastic–neoblastic grains of orthopyroxene (lines defining cleavage) hosted by serpentinized olivine (white) in residual harzburgite. The morphology of Cr-spinel suggests progressive melting-out of pyroxene and crystallization–recrystallization of Cr-spinel. Scale bar = 0.2 mm. (b) Type I orthopyroxenite-bearing harzburgite developed where partially disaggregated veins of type I orthopyroxenite (black), at the termination of a massive dyke of type I orthopyroxenite, are hosted in a low-Al dunitic matrix (white). Scale bar = 100 mm. (c) Melt impregnation texture exhibited by a single grain of orthopyroxene (black) in a matrix of serpentinized olivine (white) in type I orthopyroxenite-bearing harzburgite. Scale bar = 0.5 mm. (d) Disequilibrium texture resulting from fluid-induced incongruent breakdown of orthopyroxene (individual grains marked A, B, and C; straight lines defining cleavage) to olivine (white) and spinel (black blobs and dots) in amphibole harzburgite; symplectitic inclusions (short irregular lines) may be fluid inclusions. Scale bar = 1 mm. (e) Fluid-induced recrystallized grain of Cr-spinel (black) hosting inclusions of olivine, serpentine, magnetite, phlogopite, and Ca-amphibole in amphibole harzburgite. The grain boundary of spinel is relatively even where in contact with serpentinized olivine, and embayed where in contact with Ca-amphibole. Scale bar = 100  $\mu\text{m}$ . (f) Type II orthopyroxenite-bearing harzburgite resulting from web-like veining of low-Al dunite (white) by type II orthopyroxenite (black). The dunitic areas comprise single crystals or aggregates of olivine. Scale bar = 50 mm.



along contacts between low-Al dunite and amphibole dunite, and between low-Al dunite and amphibole harzburgite.

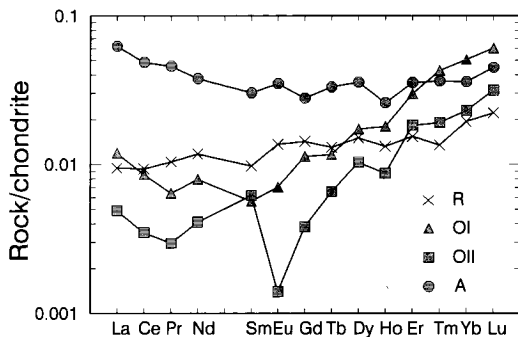
### Analytical techniques

Detailed information on analytical techniques is given by Edwards (1991), and an exhaustive documentation is inappropriate here, as the data in Table 1 are simply used to define the major diagnostic features of the harzburgite types. Silicates and spinels were analysed using JEOL 733 and JEOL JXA-50A electron microprobes, respectively. Major elements in whole-rock samples were determined by atomic absorption spectrophotometry. For mineral and whole-rock major element analyses, concentrations are accurate and in most cases precision is better than 2%.

The REE data need special consideration, as determina-

tion of reliable concentrations of these elements in whole-rock samples of depleted, refractory mantle peridotites is notoriously difficult because concentrations are so low. The problems are enhanced by the fact that concentrations in peridotite standards with similar levels of REEs are not known to better than  $\pm 10\%$  (Frey 1984). The REE data in Table 1 were obtained by inductively coupled plasma – mass spectrometry following acid digestion and preconcentration of sample. The United States Geological Survey standard PCC-1 was prepared and analysed along with the Springers Hill samples. It demonstrated that absolute concentrations should be considered minimum values, and although chondrite-normalized REE patterns are accurate, their absolute position is obviously a minimum. Precision of the pattern is better for LREEs than for heavy rare earth elements (HREEs). In this study, the general shape of the

**Fig. 5.** Chondrite-normalized REE patterns of residual (R), type I orthopyroxene-bearing (OI), type II orthopyroxene-bearing (OII), and amphibole (A) harzburgites. Data points are original concentrations with no correction made for LOI. Normalization values are those of Wakita et al. (1971).



chondrite-normalized patterns in Figs. 5 and 6 is the main consideration, and not the absolute values. Modelling using REE data has not been attempted.

### Serpentinization

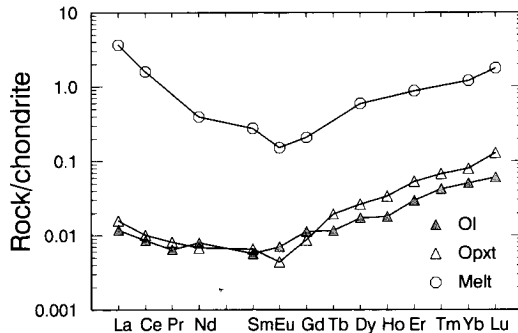
The high values for loss on ignition (LOI) in Table 1 reinforce petrographic evidence that the Springers Hill harzburgites have suffered similar degrees of extensive serpentinization of olivine. There is a positive correlation between the estimate of the original modal abundance of olivine and the value of LOI for the corresponding whole-rock sample (Table 1). In-depth discussion on the effect of serpentinization on the composition of the samples studied is beyond the scope of this paper, but initial assessment suggests modification occurred under nonconstant-volume conditions and was largely isochemical (Coleman and Keith 1971; Eckstrand 1975). In the case of estimates of modal proportions in Table 1, volume changes are relatively unimportant because only extreme variations in modal mineralogy are used in discussion. Mineral and whole-rock compositions are consistent with refractory mantle compositions reported in other studies, and Table 1 shows that Mg# (whole rock) is comparable with Mg# (olivine) ( $Mg\# = 100Mg/(Mg + Fe_{total}^{2+})$ ), which implies that Mg-Fe redistribution essentially did not occur.

The mobility of REEs during serpentinization remains a topic of debate, but Frey (1984) and Prinzhofer and Allègre (1985) present data suggesting immobility of REEs during this alteration process. In addition, Stosch and Seck (1980) demonstrated that REE patterns of whole-rock samples of mantle peridotite are controlled by the mineral assemblage of the sample, as a result of analysing acid-leached mineral separates. Based on these studies and evidence that serpentinization in the Springers Hill area was isochemical, the whole-rock REE patterns of Springers Hill harzburgites are adequately explained by their mineralogy which predated serpentinization in the area.

### Harzburgite formation

The identification of a spectrum of harzburgite types represented by the four lithological types outlined above forms the basis for further investigation of a complexity of

**Fig. 6.** Chondrite-normalized REE patterns of type I orthopyroxene-bearing harzburgite (OI), type I orthopyroxene (Opxt) (sample SJE-L268 from Edwards 1991), and melt estimated to have been in equilibrium with the type I orthopyroxene (Melt). The pattern for the melt is calculated using the Berthelot-Nernst equation, assuming that all crystalline products remained in chemical equilibrium with the melt and that no melt was trapped in the orthopyroxene (full details are given in Edwards 1991). The absolute position of the pattern is a minimum for reasons given in the text. Data points are original concentrations with no correction made for LOI. Normalization values are those of Wakita et al. (1971).



mantle processes. Field relations and textures of the various types must be considered, as must detailed aspects of their mineralogy and geochemistry; no one factor can be used in isolation. For example, the chemistry of Cr-spinel in all of the harzburgites restricts them to formation in a suprasubduction-zone environment, according to the criteria of Dick and Bullen (1984). However, there is likely a significant change from the asthenospheric conditions during which the residual and type I orthopyroxene-bearing harzburgites formed, to the lithospheric conditions when Type II orthopyroxene-bearing and amphibole harzburgites formed. Although the latter types may have formed synchronously with ophiolite obduction, the former clearly preceded this event (Fig. 3). Likewise, if one were to simply use the whole-rock major element criteria of Ishiwatari (1985), all the harzburgite types should be considered residual from partial melting of between 25% (type I orthopyroxene-bearing harzburgite) and 30% (residual harzburgite). We believe that field, petrographic, and additional geochemical evidence are inconsistent with this interpretation and point rather to multiple origins and thus a complexity of mantle processes.

### Residual harzburgite

The texture, mineralogy, mineral chemistry, and whole-rock major element chemistry of residual harzburgite suggest it is a refractory residue from partial melting beyond the clinopyroxene-out phase boundary (Dick and Bullen 1984), which equates to a total of 30% partial melting using the parameters of Ishiwatari (1985). However, this figure is expected to be an underestimate, as the model of Duncan and Green (1987) predicts that residual harzburgite containing spinel with Cr# of 76–78 (Table 1) formed from at least 43% total partial melting of upwelling mid-ocean ridge basaltic (MORB) pyrolite. The final melt extracted should have been a high-Mg quartz tholeiite formed at <30 km



depth (Duncan and Green 1987). These constraints agree with the prediction of Kostopoulos (1991) that 42% total partial melting of adiabatically rising MORB-pyrolite mantle is required to completely remove clinopyroxene from the resultant harzburgite residue.

The gently, positively sloping REE pattern of residual harzburgite in Fig. 5 is similar to those of some harzburgites from the Troodos (Kay and Senechal 1976), New Caledonia (Nicolas and Dupuy 1984), and Trinity (Brouxel and Lapierre 1988) ophiolites, and from the Ronda peridotite (Frey et al. 1985). However, this pattern is relatively rare and is inconsistent with simple models of extremely high degrees of partial melting. Equilibrium melting models, especially for fractional or incremental melting, predict a highly depleted LREE pattern for the residue (e.g., Johnson et al. 1990). The relatively flat REE pattern in residual harzburgite, therefore, is unlikely to result solely from partial melting, but from melt percolation associated with partial melting or from melt infiltration after partial melting. In the case of the former, Bodinier et al. (1991) proposed that the composition of massive refractory peridotites may be controlled by a single process involving partial melting and interaction of the extracted melt with the residue. In this model, the percolating melt suffers chromatographic fractionation (Navon and Stolper 1987; Vasseur et al. 1991), whereby LREE enrichment occurs over long percolation distances, but HREE concentrations are rapidly buffered by the refractory peridotite matrix. An alternative explanation involves partial melting and melt extraction to form a LREE-depleted residue, which later experienced REE enrichment, particularly of the light and middle REEs, by addition of a fluid-dominated component from a different melt source. This explanation is favoured for the residual harzburgite because it contains traces of undeformed amphibole resulting from fluid impregnation. The source of the introduced component is predicted to be the HALPPGs, and REE enrichment is expected to have occurred in the manner described later for amphibole harzburgites. Only a very small volume of the REE-enriched fluid component entered the harzburgite matrix, as evidenced by the small volume of Ca-amphibole seen in thin section and the absence of significant modification of the original texture, mineral chemistry, and whole-rock major element chemistry.

The residual harzburgites are, therefore, predicted to have a two-stage evolution. Their initial highly refractory residual character was produced in the asthenosphere by a high degree of partial melting and, finally, extraction of high-Mg quartz tholeiite (Duncan and Green 1987), predicted to have been LREE depleted. Subsequently, under lithospheric conditions during formation of the HALPPGs, they were impregnated by a small volume of REE-enriched fluid, which probably controls their REE pattern. As such, the asthenospheric component of the residual harzburgites may be part of a simple parent–daughter relationship between residual mantle harzburgites and basaltic crustal rocks, both with positive-sloping REE patterns exhibiting significant LREE depletion.

#### Type I orthopyroxenite-bearing harzburgite

Type I orthopyroxenite-bearing harzburgite is rich in orthopyroxene and has a U-shaped REE pattern (Fig. 5). Kelemen

et al. (1992) suggested that harzburgites with these characteristics are unlikely to be residual from simple partial melting of primitive mantle. Instead, they proposed that these harzburgites may form by pervasive melt–rock reaction, whereby peridotite experiences clinopyroxene dissolution, orthopyroxene and olivine formation, and LREE addition as melt mass decreases. The composition of the type I orthopyroxenite-bearing harzburgites also precludes an origin solely by partial melting, but their mode of formation is somewhat different from that proposed by Kelemen et al. (1992), as they are modelled as hybrids composed of residual harzburgite, with associated dunite, and introduced type I orthopyroxenite (Edwards 1990). Field and textural relations suggest that melt passed through residual harzburgite and associated dunite and, depending on the mode of migration and the volume, crystallized veins and dykes of type I orthopyroxenite and interstitial orthopyroxene in the peridotite matrix. Syn- or post-crystallization asthenospheric deformation thoroughly mixed this orthopyroxene component with the peridotite host. Original magmatic features are preserved in zones of low strain.

The component of introduced orthopyroxene is apparent in the field and in thin section, but not from mineral chemistry. The latter is expected, as the composition of minerals in residual harzburgite and type I orthopyroxenite are similar, with the exception of Cr-spinel, which has a higher Cr# in the latter (Edwards 1991). Most differences in mineral chemistry that did exist have likely been smoothed by reequilibration under both magmatic and subsolidus conditions. Nevertheless, the U-shaped REE pattern of type I orthopyroxenite-bearing harzburgite, similar to that of type I orthopyroxenite, is the most obvious chemical indication that orthopyroxene has been introduced (Fig. 6).

An important point to note is that the melt in equilibrium with type I orthopyroxenite, i.e., that which was added to residual harzburgite and associated dunite by melt impregnation and veining or mechanical mixing, was similar in composition to boninitic and other low-Ti, high-Mg, quartz-normative subduction-related melts with U-shaped REE patterns (Fig. 6). This implies that a component of this melt was added to peridotite (harzburgite or dunite) and imparted its mineralogical and geochemical signature to the resultant harzburgite. Hence, harzburgites like type I orthopyroxenite-bearing harzburgites may reflect the composition of the melt added to them, rather than the composition of the melt that was originally derived from them by partial melting.

#### Amphibole harzburgite

The major differences between amphibole harzburgite and those types already described are the broad range of Cr#s exhibited by Cr-spinel, the occurrence of Ca-amphibole and phlogopite, and the disequilibrium textures exhibited by both Cr-spinel and orthopyroxene. We conclude that these features imply that amphibole harzburgites originated as residual or type I orthopyroxenite-bearing harzburgites and later recrystallized and partially reequilibrated in the presence of fluid, under lithospheric conditions. Field relations suggest that the fluid source was the HALPPGs and, therefore, ultimately related to the low-Si/Al, fluid-bearing basaltic melt parental to the HALPPGs. On release from the HALPPGs, the fluid was H<sub>2</sub>O rich with a low Si/Al ratio

and aggressively assimilated orthopyroxene as a consequence of the ability of  $H_2O$  to lower  $SiO_2$  activity in fluid (Kushiro 1969; Sisson and Grove 1993). Where complete orthopyroxene dissolution was achieved, dunite or amphibole dunite was formed. This process is very similar to that described by Dungan and Avé Lallemant (1977) for the metasomatic transformation of harzburgite to dunite in the Canyon Mountain ophiolite in northeast Oregon.

To produce the U-shaped REE pattern characteristic of the amphibole harzburgite (Fig. 5), the fluid that reacted with residual and type I orthopyroxenite-bearing harzburgites must have been relatively LREE enriched. The initial fluid-bearing basaltic melt parental to the HALPPGs was LREE depleted, but through physical separation and fractionation of clinopyroxene and Ca-amphibole to form LREE-depleted HALPPGs, a LREE-enriched hydrous fluid entered the LALPPs (Edwards 1991). The LREEs would have been readily transported in this hydrous fluid (Eggler 1987), and this fluid interacted with the residual and type I orthopyroxenite-bearing harzburgite host to produce LREE enrichment. The mechanism of producing LREE enrichment in the fluid is very similar to that proposed by Arth and Barker (1976) for generating LREE enrichment in trondhjemitic-tonalitic melts.

#### Type II orthopyroxenite-bearing harzburgite

Type II orthopyroxenite-bearing harzburgites have many features characteristic of low-Al dunite and type II orthopyroxenite in olivine-rich and orthopyroxene-rich zones, respectively. Like amphibole harzburgites, they contain Cr-spinel exhibiting disequilibrium textures and hosting Ca-amphibole. As with type I orthopyroxenite-bearing harzburgites, type II orthopyroxenite-bearing harzburgites are too rich in orthopyroxene to be normal residues from partial melting of primitive upper mantle. The web-like texture exhibited by the type II orthopyroxenite veins also rules out a residual origin (Fig. 4f).

Based on field evidence and the proposed origin of amphibole harzburgites, type II orthopyroxenite-bearing harzburgites formed by interaction of low-Al dunite with hydrous fluid after this fluid had been involved in the formation of amphibole harzburgite or amphibole dunite. During formation of the latter rock types, the fluid would have developed a high Si/Al ratio by assimilation of orthopyroxene. During continuous reaction to form amphibole harzburgites and amphibole dunites, orthopyroxene dissolution would eventually have ceased as the fluid equilibrated with the host. Wherever the fluid subsequently interacted with low-Al dunite, reaction with olivine to produce orthopyroxene would have taken place until a new equilibrium had been attained. Because the dominant rock type in the LALPPs through which the fluid initially passed and interacted was harzburgite, the fluid would have progressively equilibrated with harzburgite as a result of reaction, and, subsequently, would have reacted with dunite to form orthopyroxene until the bulk composition of the olivine-orthopyroxene assemblage was harzburgitic. Such an origin for orthopyroxene is similar to the hydrothermal production of pyroxene modelled by Bowen and Tuttle (1949).

At this time the origin of the REE pattern in type II orthopyroxenite-bearing harzburgite is unclear, particularly

as modelling is not possible with the uncertainty in the extremely low concentrations of a number of light and middle REEs (Fig. 5). As with amphibole harzburgites, the fluid may have been responsible for LREE enrichment. Alternatively, coupled effects of orthopyroxene formation in equilibrium with harzburgite and chromatographic fractionation during fluid migration may have played a significant role. Further investigation is required to determine the dominant process(es).

#### REEs and spinel in amphibole harzburgite

The partitioning of REEs into spinel is generally considered to be negligible, as spinel-melt partition coefficients of  $<0.01$  for all REEs have been reported by Arndt and Lesher (1992) and calculated by Prinzhofer and Allègre (1985) from the data of Stosch (1982). However, amphibole harzburgite exhibits a U-shaped REE pattern and contains Cr-spinel with inclusions of Ca-amphibole and phlogopite, which formed by reaction of LREE-enriched fluid with the spinel. Without analysing the hydrous minerals directly it is difficult to predict their REE content, but other studies have shown that they often exhibit LREE-enriched patterns (Irving and Frey 1984). If this is the case, then, although spinel-melt partition coefficients are likely to have little effect on the REE pattern of amphibole harzburgite, the reaction and recrystallization of Cr-spinel in the presence of fluid promotes the formation of amphibole and phlogopite, which may control enrichment of LREEs in addition to Ti, Na, K, and  $H_2O$ . It may be possible to use a similar explanation for the initial enrichment of LREEs in type II orthopyroxenite-bearing harzburgite, which also contains Ca-amphibole-bearing Cr-spinel. Consequently, spinel-fluid interactions may play a fundamental role in concentrating REEs, Na, K, and Ti in highly refractory mantle peridotites.

#### Discussion

The harzburgite types identified in this work are best preserved in the mantle section of the Lewis Hills in zones of lowest strain. In addition, some harzburgites were effectively removed from the mantle environment soon after their formation by ophiolite obduction and, therefore, underwent little or no recrystallization or deformation at mantle temperatures. It is only because of this degree of preservation that it is possible to delineate a range of harzburgite types, with a range of characteristic features which allows for the recognition of their likely origins. If types I and II orthopyroxenite-bearing and amphibole harzburgites had been completely recrystallized at mantle temperatures, they would no doubt have exhibited field, petrographic, and mineral chemical features often described as residual in origin. In such a case, it would be difficult to explain the higher than predicted orthopyroxene concentrations and (or) the occurrence of Ca-amphibole and phlogopite. For the Springers Hill area, we have been able to place the field, petrographic, and mineral and whole-rock chemical data, which clearly show a more complex history than a simple residual origin, in a suitable context of harzburgite formation.

Having identified a range of different harzburgite types, it is interesting to speculate on how they might relate to melt compositions thought to be produced by mantle partial melt-

ing. For example, present models for the generation of boninitic melts in suprasubduction-zone environments require an initially depleted, refractory peridotite source, similar, perhaps, to the residual harzburgite before it experienced fluid impregnation, which resulted in the flat REE pattern. This is thought to undergo enrichment in large-ion lithophile elements, including LREEs, and Zr, Sr, Na, and possibly Si and Al, prior to or during the melting event (see summary by Crawford et al. 1989). Excluding residual harzburgite with a relatively flat REE pattern, we have shown that all other types of harzburgite exhibit whole-rock and (or) mineralogical evidence for such an enrichment in LREEs and Al (types I and II orthopyroxenite-bearing and amphibole harzburgites), Si (types I and II orthopyroxenite-bearing harzburgites), and Na and Ti (type II orthopyroxenite-bearing and amphibole harzburgites). Frey and Green (1974) suggested that the enriched component resides on grain boundaries in upper mantle peridotites, but field and petrographic data of our study demonstrate that it is more likely contained within the mineral phases of the type I orthopyroxenite component in type I orthopyroxenite-bearing harzburgites, or in hydrous phases formed by metasomatic reactions involving orthopyroxene and Cr-spinel in type II orthopyroxenite-bearing and amphibole harzburgites. It appears that types I and II orthopyroxenite-bearing and amphibole harzburgites have developed from the equivalent of LREE-depleted residual harzburgite and associated dunite to the composition required of a source for boninitic melts, albeit with large variations in depleted and refractory character. Ca-amphibole plays a key role in the genesis of type II orthopyroxenite-bearing and amphibole harzburgites, which supports the prediction of Pearce et al. (1992) that amphibole was significant in the genesis of boninites sampled on Ocean Drilling Program Leg 125 in the Bonin–Mariana forearc.

We are as yet unable to delineate the source of the original enrichment of the type I orthopyroxenite, which was subsequently passed to the type I orthopyroxenite-bearing harzburgites. However, in the case of the type II orthopyroxenite-bearing and amphibole harzburgites, this study and those of Edwards (1991) and Edwards and Malpas (1993) suggest that the enriching hydrous fluid came from fluid-bearing basaltic melt, which was produced by partial melting of underthrust, hydrated oceanic lithosphere during initial displacement of the Bay of Islands ophiolite (Fig. 3). This melt, and the fluid phase liberated from it, underwent complex physical and chemical evolution before reaching the appropriate enriched composition. Similarly, enriched components derived from altered, subducted oceanic lithosphere have been proposed by Kyser et al. (1986) and Dobson and O'Neil (1987).

The evolution of metasomatizing melts and fluids in the mantle is a direct consequence of physical and chemical changes imposed upon them as they progressively equilibrate with changing host rock compositions. Commonplace amongst these are likely to be repeated solution–precipitation reactions involving orthopyroxene. These may result in “fronts” of harzburgite that progressively move upward through the mantle or laterally along shear zones. Such a process may explain the association of dunite, harzburgite, and orthopyroxenite-veined harzburgite in the uppermost mantle sections of ophiolites. If such is the case, then there

are major implications for the origin and extent of dunitic transition zones in ophiolites and the oceanic lithosphere, and for the rate of chemical versus physical migration of harzburgite in the mantle, which have yet to be further investigated.

## Acknowledgments

P. Horan and P. King greatly assisted with acquisition of REE data. G. Andrews, G. Veinott, B. MacKay, and the Department of Mines and Energy (Newfoundland) assisted with whole-rock and mineral analyses. D. Groom and D. Williams helped with drafting diagrams. We especially thank G. Suhr and A. Szybinski for informative discussions and J.-L. Bodinier and M. Menzies for friendly and constructive reviews, all of which improved the manuscript. This work was supported by a Canadian Commonwealth Scholarship to Edwards and the 1984–1989 Canada–Newfoundland Mineral Development Agreement. Some aspects of this study were undertaken by the first author as part of his Ph.D. research at Memorial University of Newfoundland. The completion of this paper was made possible by a grant to Edwards from the Joseph Burr Tyrell Fund of the Geological Society of London.

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