



Primitive CaO-rich, silica-undersaturated melts in island arcs: Evidence for the involvement of clinopyroxene-rich lithologies in the petrogenesis of arc magmas

P. Schiano

Department des Sciences de la Terre (UMR 6524), Université Blaise-Pascal, 5 rue Kessler, 63038 Clermont-Ferrand cedex, France (schiano@opgc.univ-bpclermont.fr)
Division of Geological and Planetary Sciences (170-25), California Institute of Technology, Pasadena, California 91125

J. M. Eiler

Division of Geological and Planetary Sciences (170-25), California Institute of Technology, Pasadena, California 91125 (eiler@gps.caltech.edu)

I. D. Hutcheon

Analytical and Nuclear Chemistry Division, Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, California 94551 (hutcheon@llnl.gov)

E. M. Stolper

Division of Geological and Planetary Sciences (170-25), California Institute of Technology, Pasadena, California 91125 (ems@gps.caltech.edu)

[1] **Abstract:** On the basis of the study of olivine-hosted melt inclusions in a calc-alkaline basalt from Batan Island (Philippines) we define a distinctive type of primitive, nepheline-normative island arc magma characterized by unusually high CaO contents (up to 19.0 wt %) that cannot be simply explained by melting of the metasomatized peridotitic mantle wedge above subducting oceanic lithosphere. CaO-rich melt inclusions with these characteristics are preserved in Fo₈₅₋₉₀ olivine, and compositional variations among the inclusions are interpreted to reflect mixing between melts such as those found in the most CaO-rich inclusions (present in Fo₉₀ olivine) and melts similar to primitive “normal” island arc magmas (trapped in Fo₈₅ olivine). Compilation of primitive island arc magmas from the literature shows that whole rocks and olivine-hosted melt inclusions with CaO contents >13 wt % are found in many arc volcanoes from all over the world in addition to Batan. These inclusions occur in lavas ranging from CaO-rich ankaramites to basaltic andesites with low-CaO contents (i.e., <13 wt %). The globally occurring CaO-rich inclusions and whole rocks comprise a group that although defined on the basis of their CaO contents is compositionally distinctive when compared to island arc lavas that have lower CaO contents; for example, they have lower FeO at a given SiO₂ content than most arc lavas, and they are all nepheline normative, with normative nepheline contents positively correlated with CaO contents. Variations in CaO content and normative compositions of experimental partial melts of lherzolite related to changes of pressure, temperature, and source composition suggest that there are no conditions under which partial melting of peridotite can generate melts having CaO contents and other properties comparable to those observed for the primitive, CaO-rich arc-derived melts identified here. Although melting of peridotite at high pressure in the presence of CO₂ can produce CaO-rich, silica-poor liquids,

we consider it unlikely that this is responsible for producing the CaO-rich, silica-undersaturated melts considered in this study because there are significant differences in nearly all other compositional characteristics between the CaO-rich arc magmas and melts known or thought to be produced by melting of carbonated peridotite. Model major element compositions of partial melts of clinopyroxene-rich lithologies (mantle pyroxenites, lower crustal pyroxenites, and eclogites) calculated using the MELTS algorithm suggest that the most CaO-rich, nepheline-normative melt inclusions and whole rocks identified here could represent intermediate to high degree (~10–40 wt %) partial melts of pyroxenites at lower crustal to upper mantle pressures. Such a hypothesis is supported by the comparison between the trace element compositions of model pyroxenite sources of the Batan CaO-rich melt inclusions and naturally occurring pyroxenites. The most likely source of the primitive CaO-rich, silica-undersaturated arc melts identified here is lower crustal and shallow upper mantle pyroxene-rich cumulates from arc environments because these cumulates have CaO concentrations at the upper end of the range observed for mantle pyroxenites. They are therefore more likely to yield partial melts with the restricted range of remarkably high CaO contents of the most CaO-rich inclusions and whole rocks identified here. Moreover, these cumulates often contain amphibole, which would lower their solidus temperatures relative to the anhydrous pyroxenite equivalents to values more consistent with those expected in deep crustal or shallow subarc environments.

Keywords: Melt inclusions; arc lavas.

Index terms: Chemical evolution; geochemical cycles; major element composition.

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1. Introduction

[2]K Most island arc lavas are believed to be derived from magmas formed by partial melting of the mantle overlying subducting oceanic lithosphere, where melting is fluxed by metasomatic enrichment in water (and other elements) carried by aqueous fluids and/or silicate melts released from the subducted oceanic crust [e.g., *Nicholls and Ringwood, 1973*]. The mantle sources on which this coupled enrichment-melting process operates are thought to be peridotites similar to or somewhat more depleted than the sources of mid-ocean ridge basalts (MORBs) [*Tatsumi et al., 1986; Davidson, 1987; Nohda and Wasserburg, 1981; Ryerson and Watson, 1987*].

The trace element characteristics of arc lavas e.g., their high concentrations of large-ion lithophile elements (LILE) and their relatively low concentrations of high-field strength elements (HFSE)) and their major element compositions are influenced by the metasomatism, and high thermodynamic activities of H₂O strongly affect the compositions of primary magmas and their subsequent differentiation sequences [see *Stolper and Newman, 1994*, and references therein].

[3]K Although coupled metasomatism and fluxed melting of peridotites in the mantle wedge is believed to be the dominant process by which primary arc magmas form, some arc magmas have also been proposed to form by

melting of nonperidotitic sources: for example, partial melting of subducted crust [Defant and Drummond, 1990; Drummond and Defant, 1990; Kay, 1978], melting at deep crustal levels [Pichler and Zeil, 1972], and melting of an ocean island basalt-type source [Ito and Stern, 1985] have all been proposed. In this article, we document a distinctive type of primitive, nepheline-normative island arc magma characterized by unusually high CaO contents (up to 19 wt %) that cannot be simply reconciled with the standard model of arc petrogenesis described above. Although arc-derived samples with these characteristics have been previously reported [Metrich et al., 1999; Sisson and Bronto, 1998; DellaPasqua and Varne, 1997; Gioncada et al., 1998; Metrich and Clocchiatti, 1996; Foden and Varne, 1980; Foden, 1983; Kennedy et al., 1990; Carr and Rose, 1984; Thirwall and Graham, 1984; Shimizu and Arculus, 1975; Arculus, 1976], they have not been recognized as a coherent, globally occurring magma type or as distinct from CaO-rich magmas found in oceanic island [Tronnes, 1990], mid-ocean ridge [Kamenetsky et al., 1998; Sours-Page et al., 1999], and back arc [Kamenetsky et al., 1997] settings. After documenting the existence of this distinctive magma type, emphasizing in particular a well-defined group from Batan Island (northernmost Philippines, Luzon-Taiwan arc), but also showing that such magmas occur in arcs from around the world, we examine the compositions of CaO-rich, silica-undersaturated magmas in subduction zone settings with the following objectives in mind: (1) to provide a compilation of the occurrences and compositions of such magmas; (2) to examine their relationship to more typical magmas in subduction zone settings; (3) to estimate plausible chemical and mineralogical characteristics of their sources; and (4) to evaluate the implications of their existence for models of melt generation in subduction zones.

2. Definition of a Distinctive CaO-Rich, Silica-Undersaturated Magma Type From Subduction Zone Settings

2.1. CaO-Rich, Silica-Undersaturated Melt Inclusions in Olivine Phenocrysts in Lavas From Batan Island (Northernmost Philippines, Luzon-Taiwan Arc)

2.1.1. Sample description, analytical techniques, and results

[4] The geologic setting of Batan Island and the petrography of its principal rock types (basalts, basaltic andesites, and andesites) are described by Richard et al. [1986]. Plagioclase, olivine, and clinopyroxene dominate the phenocryst assemblages of the basalts, while basic and acid andesites contain phenocrysts of clinopyroxene, hornblende, plagioclase, and titanomagnetite. Sample B45 from Mount Iraya, the youngest (2.32 to <0.1 myr) volcano on Batan Island [Richard et al., 1986], is a high-MgO (10.8 wt %) calc-alkaline basalt [Metrich et al., 1999; Richard et al., 1986]. Compositions of primary melt inclusions in olivine phenocrysts (Fo_{90-75}) from this sample were determined on glasses produced after heating each inclusion up to its homogenization temperature (i.e., the temperature at which the inclusion contents (gas, glass, and crystals) homogenized visually to a uniform melt phase). Heating experiments were performed with a high-temperature, optical heating-stage apparatus [Sobolev et al., 1980] that allowed visual monitoring of individual melt inclusions during heating. Homogenization temperatures were $1220 \pm 20^\circ\text{C}$, and effective times of quenching were <1 s. Comparison between the compositions of homogenized and unheated glass inclusions trapped in olivine phenocrysts with similar forsterite content shows that the amount of host olivine needed to be added to the unheated glass inclusions to get the heated ones is relatively

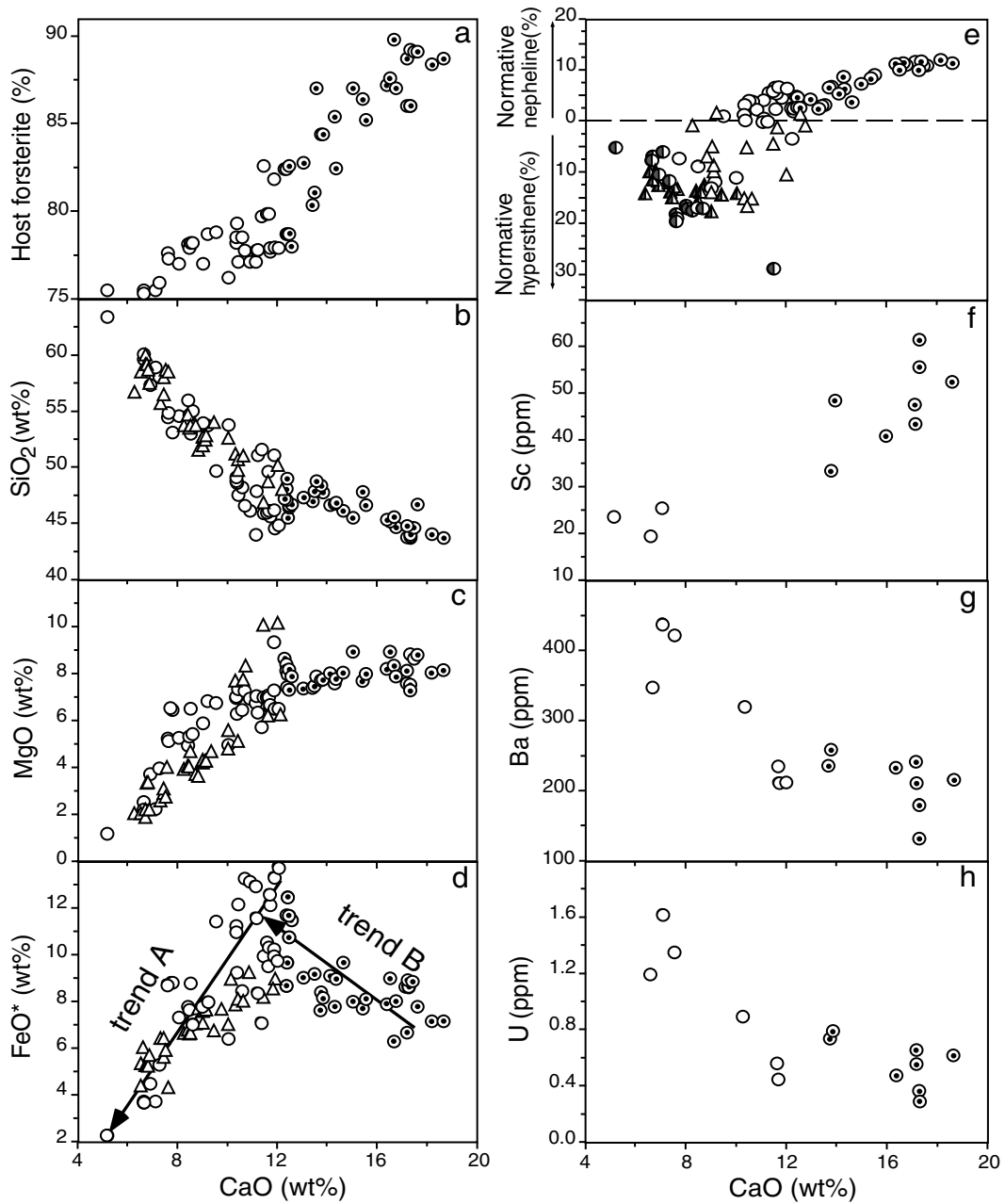
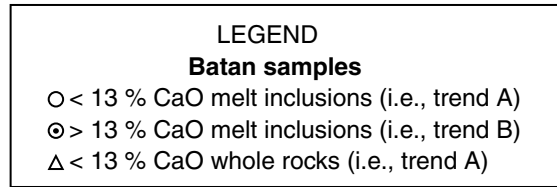
small ($\leq 15\%$ in weight). Consequently, the effect of the heating procedure on the concentrations of elements other than MgO (and to a lesser extent FeO and SiO₂) is relatively small, and therefore the main conclusions of the paper

would be unchanged if unheated inclusions were considered. It should be noted that this statement applies not only to the Batan samples but also to those in the compilation that will be presented later in this article.

Table 1. Representative Major and Trace Element Compositions of Homogenized Melt Inclusions in Olivine in a Calc-Alkaline Basalt (B45) From Batan Island (20°25'N–121°54'E, Luzon-Taiwan Arc)^a

	Sample Identification								
	B7-1	B7-2	B2-1	B4	B1-2	B1-1	B5-1	B8-1	B6-1
SiO ₂ , wt %	44.02	45.05	44.00	45.61	44.10	44.04	45.89	49.42	54.73
TiO ₂ , wt %	0.70	0.64	0.79	0.67	0.94	0.76	0.74	0.91	0.98
Al ₂ O ₃ , wt %	16.01	16.87	14.92	16.58	16.13	16.02	16.43	15.39	14.44
FeO*, wt %	7.31	6.80	8.86	8.02	8.82	8.85	12.27	11.11	8.84
MnO, wt %	0.12	0.10	0.15	0.14	0.14	0.15	0.20	0.18	0.19
MgO, wt %	8.30	8.27	8.96	8.36	7.67	7.72	6.71	7.17	5.63
CaO, wt %	18.54	17.08	17.20	16.27	17.20	17.06	11.57	10.22	7.48
Na ₂ O, wt %	2.58	2.56	2.43	2.53	2.29	2.60	2.75	3.05	3.29
K ₂ O, wt %	0.93	1.01	0.86	1.01	0.90	1.01	1.40	1.43	2.17
Cr ₂ O ₃ , wt %	0.05	0.04	0.05	0.08	0.04	0.03	0.08	0.08	0.03
SUM	98.57	98.42	98.20	99.26	98.23	98.24	98.02	98.95	97.77
HOST	Fo ₈₉	Fo ₈₉	Fo ₈₉	Fo ₈₇	Fo ₈₆	Fo ₈₆	Fo ₇₈	Fo ₇₉	Fo ₇₈
Ba, ppm	219	214	185	237	136	245	215	323	423
Th, ppm	2.12	2.18	1.57	2.32	1.04	2.87	2.06	3.74	6.75
U, ppm	0.63	0.57	0.38	0.49	0.31	0.67	0.39	0.91	1.37
Nb, ppm	2.20	2.26	4.40	2.35	2.09	2.83	2.68	4.00	6.17
Sr, ppm	537	539	498	490	398	571	536	663	561
Zr, ppm	56.1	60.7	52.6	58.6	47.0	66.9	62.7	90.2	124.7
Y, ppm	17.3	17.0	19.9	14.1	13.4	17.1	17.9	26.7	25.2
Sc, ppm	53	44	62	43	56	48	N.D.	N.D.	26
La, ppm	9.21	9.39	8.31	9.10	7.45	10.52	9.11	13.13	18.40
Ce, ppm	21.4	20.9	18.4	21.2	15.7	24.9	19.8	31.5	43.3
Nd, ppm	14.3	14.2	12.3	13.6	10.7	19.3	13.2	22.4	26.2
Sm, ppm	3.15	2.99	2.61	3.00	1.95	3.49	2.86	4.33	4.99
Eu, ppm	0.95	0.92	0.67	0.85	0.57	0.99	0.83	1.30	1.26
Dy, ppm	2.81	2.95	2.47	2.58	1.94	3.01	2.46	4.56	4.56
Er, ppm	2.09	2.01	1.85	1.82	1.21	2.07	1.93	3.42	3.32
Yb, ppm	2.34	2.26	2.26	2.09	1.39	2.31	2.25	3.77	3.37

^aMajor element compositions were measured with the Caltech 5-spectrometer JEOL 733 microprobe using an accelerating voltage of 15 Kv, a sample current of 10 nA and a defocused beam (size $\sim 20 \mu\text{m}$). Data were reduced using a modified ZAF procedure (CITZAF [Armstrong, 1988]). The reproducibility and accuracy of the Na measurements were tested on glass standards. The abundances of Sr, Y, Zr, Nb, Ba, rare earth elements, Th, and U were determined with the Lawrence Livermore National Laboratory (LLNL) ion microprobe, a modified Cameca IMS-3f instrument, using a 17 keV, ¹⁶O⁻ primary ion beam, focused into a $\sim 20 \mu\text{m}$ -diameter spot. Positive secondary ions were extracted and accelerated, nominally to 4500 V. A field aperture inserted in the sample image plane allowed only ions from the central 30- μm -diameter area of the imaged field to enter the mass spectrometer. Isobaric molecular interferences were minimized by energy filtering, using a 40-eV window and offsetting the accelerating voltage by 100 V from the voltage at which the energy distribution of ¹⁶O⁺ dropped to 10% of its maximum value. Trace element concentrations were determined from ⁴²Ca-normalized ion intensities using sensitivity factors established from analyses of a suite of silicate glass and mineral standards. Each analysis consisted of 8–10 cycles over the masses of interest. On the basis of analyses of NIST glass standards (NBS-610, -612, and -614, using the concentrations given by Pearce *et al.* [1997]), the accuracy varies between ± 3 and $\pm 10\%$ for Sr, Y, Zr, Ba, La, Ce, Nd, Er, and Yb, between ± 10 and $\pm 15\%$ for Nb, Th, Sm, and Dy, and between ± 15 and $\pm 25\%$ for U and Eu. Sc analyses were carried out at a mass resolving power of 4000 and zero energy offset, using a 2-nA ¹⁶O⁻ primary ion beam. The ⁴²Ca-normalized ion intensities were converted to concentrations using sensitivity factors established from analyses of NIST standard glass SRM 612.



[5] Homogenized inclusions were analyzed for major and minor elements using the electron microprobe at Caltech and for trace elements (Sc, Sr, Y, Zr, Nb, Ba, rare earth elements (REE), Th, and U) using the ion microprobe at the Lawrence Livermore National Laboratory (Table 1; see footnote for analytical procedures). The inclusions range in composition from nepheline-normative basalt to quartz-normative dacite; major element variations in these glasses are illustrated in Figure 1. The CaO and MgO contents decrease from 18.5 to 5.0 wt % and 9.5 to 1.3 wt %, respectively, as SiO₂ increases from 44.3 to 63.7 wt %. Variations in the compositions of melt inclusions are well correlated with the Mg# of their host olivines, with the most SiO₂-poor and CaO- and MgO-rich inclusions trapped in the most magnesian olivines (~Fo₉₀) and the most SiO₂-rich and CaO- and MgO-poor inclusions trapped in the least magnesian olivines (~Fo₇₅).

[6] Trace element compositions of olivine-hosted melt inclusions are also well correlated with major element composition: for example, Sc content decreases from 65 to 20 ppm and incompatible trace elements increase regularly as CaO decreases (Figures 1f–1h). The mid-ocean ridge basalt (MORB)-normalized trace element patterns of the inclusions (Figure 2a) are typical of arc magmas (i.e., enriched in LILE and depleted in HFSE and in high REE (HREE) relative to MORB; enriched in low REE (LREE) relative to HREE); these trace element features are thought to reflect melting of mantle

sources depleted with respect to the sources of MORB that have been metasomatized by a slab-derived, H₂O-rich component [Stolper and Newman, 1994]. Note that although the incompatible element concentration variations are correlated with major element composition, the La/Yb ratio does not increase as the CaO content decreases (Figure 2a).

[7] Although in most variation diagrams the melt inclusions and previously analyzed whole rocks from Batan [Richard *et al.*, 1986; Maury *et al.*, 1998; McDermott *et al.*, 1993] define a single trend, with the CaO-rich inclusions falling on a continuous extension of the trend defined by the inclusions with lower CaO contents, Figure 1d shows that this is not the case for FeO: that is, the CaO-rich inclusions define a population offset toward lower FeO contents from the trend of whole rocks and lower-CaO inclusions. For the purposes of later discussion, we refer to the trend defined by the Batan whole rocks and lower-CaO inclusions as trend A and the higher CaO, lower FeO trend as trend B. Note that inclusions from trends A and B do not occur in the same olivine phenocrysts. The distinction between trends A and B is also illustrated in Figure 3, in which the compositions of the melt inclusions and Batan whole rocks were first recalculated to equivalent CaO-MgO-Al₂O₃-SiO₂ (C-M-A-S) [O'Hara, 1968; *Basaltic Volcanism Study Project*, 1981] and then projected from alumina onto the S-C-M plane. Also shown on this diagram are experimentally determined trends for olivine + clin-

Figure 1. CaO variation diagrams comparing melt inclusions (circles) in olivine phenocrysts from B45 basalt from Batan Island (Luzon-Taiwan arc) (this study and *Metrich et al.* [1999]) with Batan whole rocks (triangles) [Richard *et al.*, 1986; Maury *et al.*, 1998; McDermott *et al.*, 1993]. Trend A (open symbols) comprises low-CaO melt inclusions and all Batan whole rocks and is interpreted as due to progressive fractionation of olivine + clinopyroxene + amphibole (see Figure 3); trend B (dotted circles) comprises the CaO-rich inclusions and is interpreted as due to magma mixing between liquids similar to the most CaO-rich inclusions in trend B (i.e., in the Fo₉₀ olivines) and melts near the most primitive end of trend A. Normative compositions are calculated assuming $Fe^{3+} = 0.15 \sum Fe$. Half-solid circles and triangles in Figure 1e are quartz normative samples.

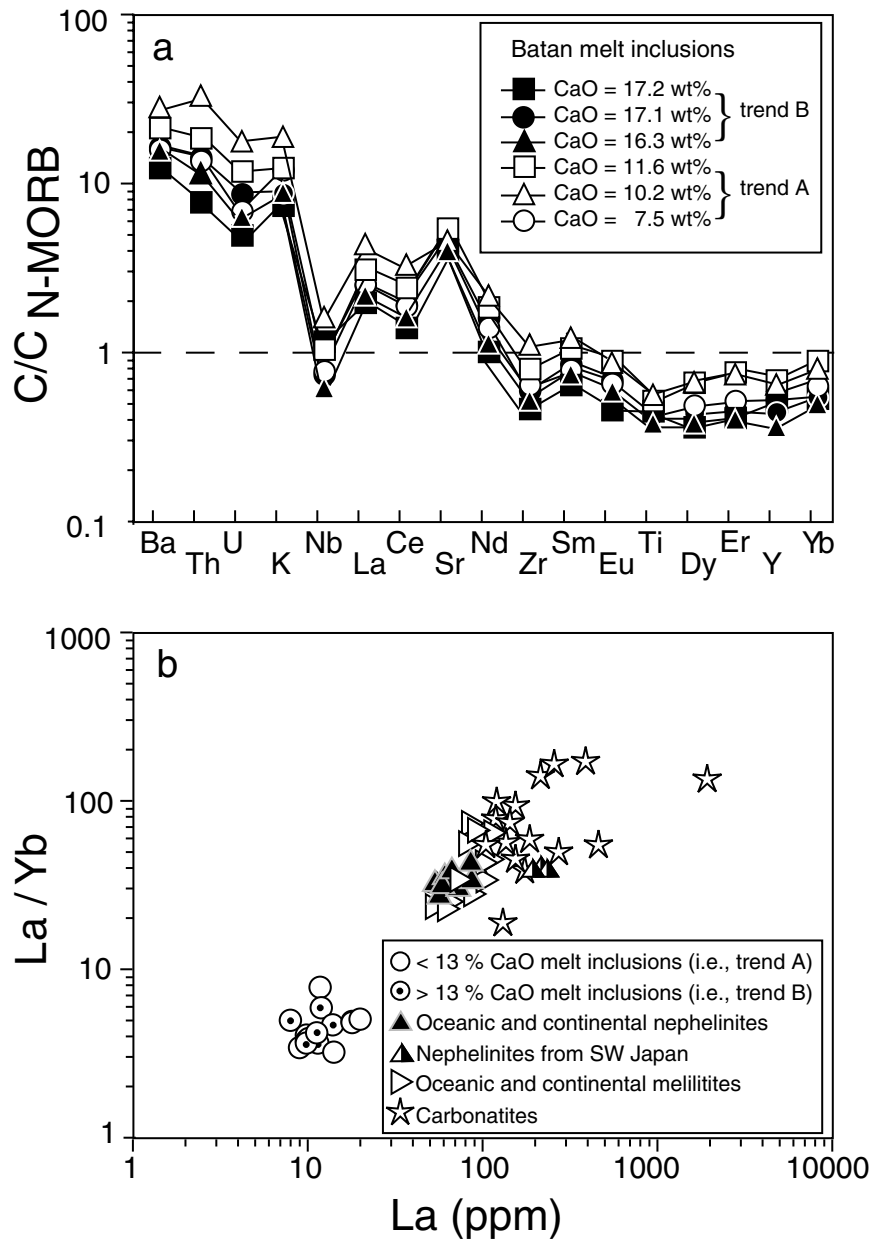


Figure 2. (a) Normal mid-ocean ridge basalt (NMORB)-normalized diagram showing trace element data for melt inclusions in olivine phenocrysts in B45 basalt from Batan Island. The order of elements is based on the incompatibility sequence during MORB petrogenesis [Hofmann, 1988]. Compositions are normalized to the average NMORB composition of Hofmann [1988]. (b) Plot of La/Yb ratio versus La concentration comparing data for Batan melt inclusions with data for primitive melilitites [Wilson *et al.*, 1995], nephelinites from oceanic and continental regions [Wedephol *et al.*, 1994; Hoernle and Schmincke, 1993], nephelinites from SW Japan [Tatsumi *et al.*, 1999], and carbonatites from Africa, Australia, Europe, and North and South America [Nelson *et al.*, 1988]. Symbols are given in the accompanying legend, and the distinction between trend A and trend B for the Batan samples is the same as in Figure 1.

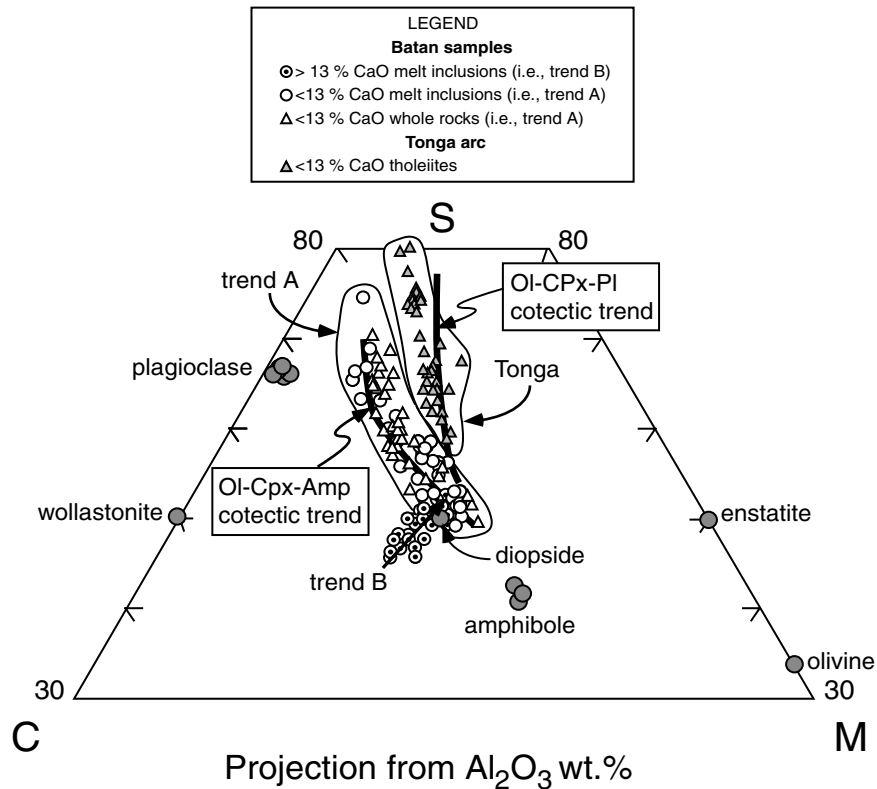


Figure 3. Compositions of Batan melt inclusions (circles) and whole rocks (triangles) recalculated to equivalent CaO-MgO-Al₂O₃-SiO₂ (C-M-A-S) and then projected from alumina onto the S-C-M plane. The distinction between trend A (open symbols) and trend B (dotted circles) for the Batan samples is the same as in Figure 1. Also shown on this diagram are experimentally determined trends for olivine-clinopyroxene-plagioclase-saturated melts at 1 atm and olivine-clinopyroxene-amphibole-saturated liquids at 2 kbar (H₂O-saturated) [Tormey *et al.*, 1987; Grove and Bryan, 1983; Walker *et al.*, 1979; Grove *et al.*, 1982; Cawthorn and O'Hara, 1976; Sisson and Grove, 1993] and the trend of whole rocks (shaded triangles) from the Tonga arc (SW Pacific), a typical tholeiitic arc series [Ewart *et al.*, 1973, 1977; Gill, 1981]. Plagioclase and amphibole compositions are from [Tormey *et al.*, 1987; Grove and Bryan, 1983; Walker *et al.*, 1979; Grove *et al.*, 1982; Cawthorn and O'Hara, 1976; Sisson and Grove, 1993]. Analyses have been recalculated as CMAS components by following a modified version [Basaltic Volcanism Study Project, 1981] of the procedure used by O'Hara [1968]. The procedure is as follows: C = (CaO - P₂O₅/3 + K₂O + 2Na₂O)56.08; A = (Al₂O₃ + Cr₂O₃ + Fe₂O₃ + Na₂O)101.96; M = (MgO + FeO + MnO)40.30; S = (SiO₂ - 2Na₂O - 4K₂O)60.08. All oxides are in molecular proportions.

opyroxene + plagioclase-saturated liquids at 1 atm and olivine + clinopyroxene + amphibole-saturated liquids at 2 kbar (H₂O saturated) [Tormey *et al.*, 1987; Grove and Bryan, 1983; Walker *et al.*, 1979; Grove *et al.*, 1982; Cawthorn and O'Hara, 1976; Sisson and Grove, 1993] and the trend of whole rocks from the

Tonga arc (SW Pacific), a typical tholeiitic arc series [Ewart *et al.*, 1973, 1977; Gill, 1981].

2.1.2. Compositional evolution of Batan melt inclusions

[8] The observation that the most CaO-rich and SiO₂-poor melts are trapped in the most for-

steritic olivine crystals (Figure 1a) suggests that the CaO-rich, silica-undersaturated melts are relatively primitive. In this section, we evaluate the origin of the compositional variation from this primitive end of the array of melt inclusion compositions to the CaO-poor, SiO₂-rich compositions contained in the least forsteritic olivines.

[9] One possible explanation for the relationship between melt inclusion composition and the Mg# of the host olivine is that compositional variations in melt inclusions represent a trend of fractional crystallization. However, although the correlated decreases in the Mg# of the host olivines and the MgO and CaO contents of the inclusions would be consistent with fractional crystallization of a clinopyroxene ± olivine-bearing assemblage, such a fractionating assemblage (assuming the clinopyroxene to be compositionally similar to the diopside, augite, or salite phenocrysts in Batan lavas [Richard *et al.*, 1986]) could not readily account for the decrease in normative nepheline with decreasing CaO content shown in Figure 1e (i.e., crystallization of such an assemblage from nepheline-normative liquids typically results in increasing rather than decreasing normative nepheline [Sack *et al.*, 1987]). Likewise, although fractional crystallization of assemblages dominated by nepheline-normative minerals such as amphibole, jadeite, or acmite could, in principle, decrease the normative nepheline contents of residual liquids, it would not decrease their CaO contents.

[10] Figures 1b–1d show that trend A (defined by melt inclusions) is typical of the compositional trend of whole rock analyses of lavas from Batan (shown as triangles in Figure 1). As indicated above, Figure 1d also demonstrates the important point that trend B, defined by the CaO-rich inclusions, is not simply an extension or continuation of the compositional trends of arc lavas from Batan;

this point is also made in Figure 3, where the CaO-poor melt inclusions and whole rocks defining trend A follow the olivine-clinopyroxene-amphibole cotectic but the trend B melt inclusions fall off this cotectic. The correspondence of trend A to the olivine-clinopyroxene-amphibole cotectic suggests that the compositional variations of Batan whole rocks could primarily reflect relatively low pressure crystallization of an amphibole-bearing assemblage from low-CaO, high-FeO parental melts comparable to those with ~12 wt % CaO trapped in Fo₈₅ olivine. This consistency of major element variations in Batan lavas with fractional crystallization of an amphibole-bearing assemblage agrees with the conclusions of several previous studies [Richard *et al.*, 1986; Maury *et al.*, 1988; McDermott *et al.*, 1993]. Note that the tholeiitic trend of whole rock lavas from the Tonga arc shown in Figure 3 contrasts with the Batan trend and suggests the Tonga trend is produced by low-pressure fractionation of an olivine-clinopyroxene-plagioclase assemblage. This hypothesis is supported by mass balance calculations [see Gill, 1981, page 272] that indicate that fractionation of an olivine-clinopyroxene-plagioclase assemblage can account quantitatively for major element variation within the Tonga volcanic suite.

[11] We thus conclude that trend A, comprising the low-CaO Batan melt inclusions and the Batan whole rock lavas, can be interpreted relatively simply in terms of progressive extraction of olivine + clinopyroxene + amphibole from a parental basaltic liquid with ~12 wt % CaO and 8–10 wt % MgO, but that trend B cannot be explained as readily by crystallization differentiation. Likewise, progressive partial melting of a peridotitic source is unlikely to account for trend B, since this would produce a positive correlation between CaO and FeO if the source contains residual clinopyroxene [e.g., Kinzler and Grove, 1992a, b; Baker and Stolper, 1994; Hirose and Kush-



Table 2. Representative Major Element Compositions of the CaO-Rich, Silica-Undersaturated Melt Inclusions and Whole Rocks in Island Arcs Identified in This Study^a

	Host	Arc	Volcano	Sample	SiO ₂	TiO ₂	Al ₂ O ₃	FeO*	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Cr ₂ O ₃	Sum	Reference
Inclusion	FO ₈₉	Java	Galunggung	J-1	45.00	1.20	19.20	6.04	0.09	4.39	18.70	3.07	0.53	0.14	N.D.	98.45	S&B (98)
Inclusion	FO ₈₈			F-1	43.90	1.28	18.40	7.53	0.09	4.99	18.40	3.49	0.66	0.18	N.D.	99.03	S&B (98)
Inclusion	FO ₈₄			I-E	44.20	1.30	19.20	8.60	0.10	4.10	17.70	2.70	0.42	0.12	N.D.	98.57	S&B (98)
Inclusion	FO ₉₄	Vanuatu	Merelava	31551	46.20	0.49	13.68	8.87	0.09	12.47	15.53	1.55	0.56	0.41	N.D.	99.98	D&V (97)
Inclusion	FO ₉₁			31551	45.88	0.46	14.07	8.87	0.02	12.18	15.89	1.59	0.52	0.37	N.D.	99.98	D&V (97)
Inclusion	FO ₉₁	E. Sunda	Rindjani	48002	43.39	1.13	15.35	9.36	0.05	11.77	15.24	2.43	0.98	0.16	N.D.	100.00	D&V (97)
Inclusion	FO ₉₀			48001	45.49	0.93	14.23	8.57	0.14	9.54	17.76	1.85	1.33	0.16	N.D.	100.13	D&V (97)
Lava				48001	48.32	0.69	10.53	9.05	0.17	14.02	14.38	1.50	0.90	0.15	N.D.	99.85	F (83)
Inclusion	FO ₉₀	Eolian	Stromboli	o15/b118	48.88	0.75	12.97	6.51	N.D.	8.16	13.78	1.85	1.59	0.32	N.D.	94.91	M&C (96)
Inclusion	FO ₉₀		Vulcano	IV88.1	44.46	0.55	11.69	7.82	0.15	8.50	15.58	2.63	3.67	0.43	0.05	95.65	G (98)
Inclusion	FO ₉₀			IV88.3.1	45.34	0.80	9.99	7.92	0.11	8.94	15.30	2.34	3.04	0.44	0.06	94.40	G (98)
Lava		N. Guinea	Lihir	L3	47.35	0.76	12.46	10.36	0.21	9.12	14.73	2.90	0.44	0.36	N.D.	98.85	K (90)
Lava				L6	47.24	0.90	12.69	10.37	0.21	7.47	13.12	2.15	2.23	0.49	N.D.	97.03	K (90)
Lava		Nicaragua	N.D.	CR-T	43.74	1.75	13.81	9.39	0.15	8.87	15.26	2.66	1.03	0.90	N.D.	97.70	C&R (84)
Lava		Lesser	Grenada	6155	46.93	0.97	16.16	9.39	0.17	7.57	14.53	1.82	1.01	0.20	N.D.	98.88	T&G (84)
Lava		Antilles		450	47.17	0.96	15.58	10.01	0.18	7.06	14.43	2.12	0.52	0.12	N.D.	98.31	T&G (84)
Lava				503	45.60	0.95	15.91	8.53	0.18	10.95	13.50	1.88	1.01	0.40	N.D.	99.04	S&A (75)
Inclusion	FO ₈₉	Luzon	Batan	B45/53	44.33	0.73	16.74	7.19	0.12	8.18	18.07	2.72	0.91	0.37	N.D.	99.91	M (98)
Inclusion	FO ₈₉			B45/43a	46.98	0.64	14.03	7.82	0.14	8.95	17.50	2.51	0.96	0.17	N.D.	100.14	M (98)
Inclusion	FO ₈₉			B45/43b	44.92	0.77	15.68	8.93	0.15	8.79	17.36	2.44	0.83	0.64	N.D.	101.05	M (98)

^aFeO* is total iron reported as FeO. N.D., not determined. References: S&B (98), *Sisson and Bronto* [1998]; D&V (97), *Della-Pasqua and Varne* [1997]; F (83), *Foden* [1983]; M&C (96), *Metrich and Clocchiatti* [1996]; G (98), *Gioncada et al.* [1996]; K (90), *Kennedy et al.* [1990]; C&R (84), *Carr and Rose* [1984]; T&G (84), *Thirwall and Graham* [1984]; S&A (75), *Shimizu and Arculus* [1975]; M (98), *Metrich et al.* [1998].

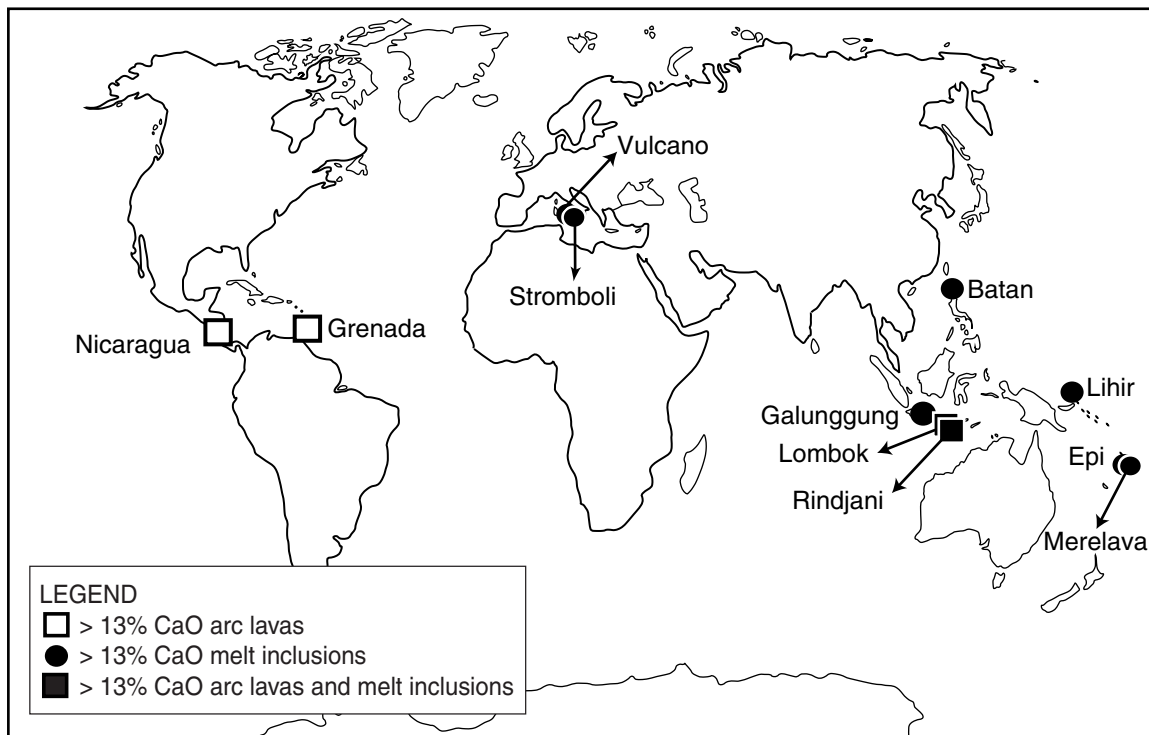


Figure 4. Map showing the locations of the CaO-rich, silica-undersaturated whole rocks and melt inclusions mentioned in the text. Solid circles indicate locations where CaO-rich inclusions have been found, open squares indicate locations where CaO-rich whole rocks have been found, and solid squares indicate locations where both have been found.

ivo, 1993], contrary to what is observed for trend B (Figure 1d). Moreover, as discussed below (see section 4.1), trend B probably cannot be explained by progressive melting of a pyroxene-rich lithology at a constant pressure, because current estimates suggest that progressive melting of pyroxenite results in liquids with increasing CaO and decreasing normative nepheline, contrary to the observed trend.

[12] Our preferred explanation for the compositional variation among the CaO-rich inclusions (i.e., those defining trend B) is that it represents a mixing trend between the most CaO-rich inclusions (i.e., those in the Fo₉₀ olivines) and melts near the most primitive end of the “normal” Batan trend that we have defined as trend

A. Figures 1 and 3 demonstrate that this can indeed explain the trend B. Although it is difficult to test this explanation for trend B using only the chemical compositions of melt inclusions, in principle, isotopic differences between inclusions along the compositional array or detailed petrographic investigation of whole rocks on this trend could be used to evaluate this hypothesis.

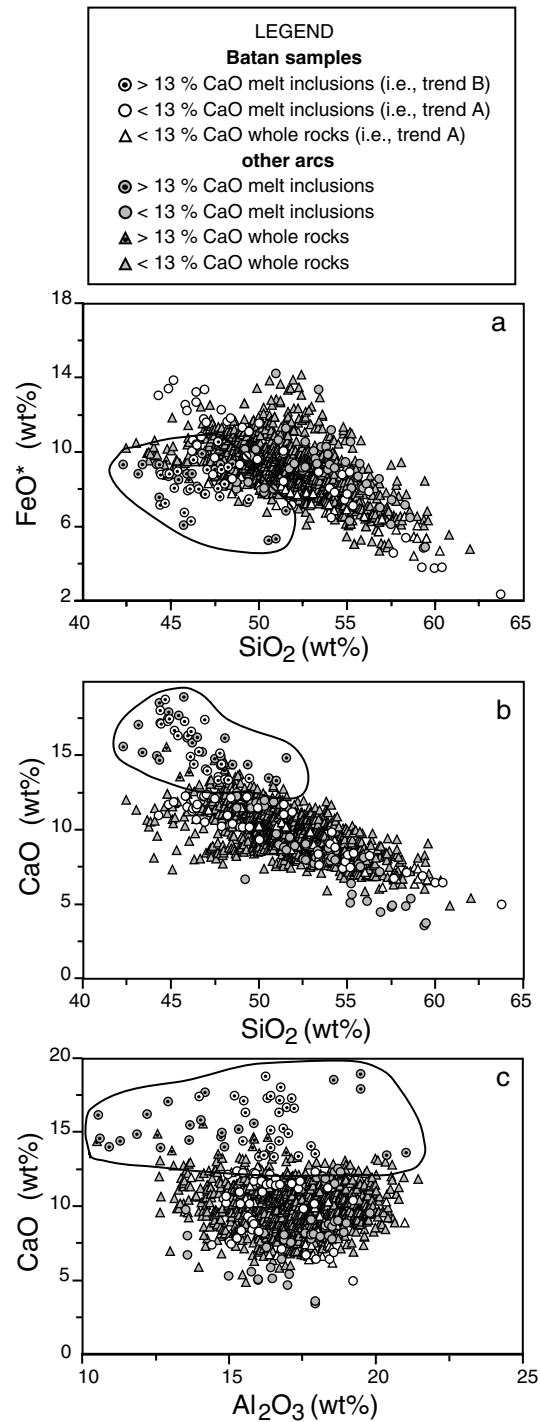
2.2. Compilation of the Occurrences and Compositions of CaO-Rich, Silica-Undersaturated Melts in Subduction Zone Settings

[13] We compiled from the literature the compositions of arc-related whole rock samples

and olivine-hosted melt inclusions having CaO contents >13 wt % (Table 2). In addition to the Batan inclusions (this study and *Metrich et al.* [1999]), whole rocks and melt inclusions with CaO contents >13 wt % are found in volcanoes from many convergent margins (Figure 4), including Galunggung (Sunda arc), Stromboli and Vulcano (Aeolian arc), Lihir (New Guinea), Nicaragua (Central America), Grenada (Lesser Antilles), Lombok (Sunda arc), Rindjani (Sunda arc), and Epi and Merelava (Vanuatu arc) [*Sisson and Bronto*, 1998; *Della-Pasqua and Varne*, 1997; *Gioncada et al.*, 1998; *Metrich and Clocchiatti*, 1996; *Foden and Varne*, 1980; *Foden*, 1983; *Kennedy et al.*, 1990; *Carr and Rose*, 1984; *Thirwall and Graham*, 1984; *Shimizu and Arculus*, 1975; *Arculus*, 1976].

[14] All the inclusions reported in Table 2 are hosted by Mg-rich olivine (Fo₈₄₋₉₄) in basalts, basaltic andesites, or ankaramites. As was done for the Batan samples reported here, they were homogenized by high-T experiments to reverse postentrapment crystallization of the host crystal [*Roedder*, 1984; *Schiano and Bourdon*, 1999], with the exception of inclusions from Galunggung [*Sisson and Bronto*, 1998], which were instead corrected by oli-

Figure 5. Variation diagrams comparing Batan CaO-rich melt inclusions (i.e., trend B) and CaO-rich melt inclusions and whole rocks from other arc environments (all of which have CaO > 13 wt %) with “normal” melt inclusions and whole rocks from Batan and other arcs (all of which have CaO contents < 13 wt %) (shaded triangles without dot, 1350 basalts with MgO > 4.5 wt % from 30 arcs, compiled by *Plank and Langmuir* [1988]. Shaded circles without dot, melt inclusions in olivine phenocrysts from *Gioncada et al.* [1998] and *Sisson and Layne* [1993]). Symbols are given in the accompanying legend. FeO* is total iron reported as FeO. Major element concentrations have been normalized to 100%.



vine addition (5–13% in weight [Sisson and Bronto, 1998]) to be in Fe-Mg equilibrium with the host olivine.

[15] The CaO-rich melt inclusions from arcs worldwide overlap the trend B inclusions from Batan in major element variation diagrams (Figure 5). They are also indistinguishable from CaO-rich whole rocks from arcs. On the other hand, the trend A inclusion and whole rocks from Batan overlap the compositional trend of CaO-poor whole rocks and melt inclusions from other arcs (Figure 5). Inspection of Figure 5 shows that as shown above on a local scale for the Batan samples, on a global scale these arc-derived CaO-rich compositions collectively form a group that although defined on the basis of their CaO contents is compositionally distinctive when compared to arc lavas and melt inclusions having lower CaO contents. The compositional characteristics of this group are the same as those of trend B melt inclusions from Batan described above: for example, CaO-rich inclusions and lavas are characterized by lower FeO concentrations at a given SiO₂ content than most arc lavas (Figure 5), and they are all nepheline normative (0.5–16.2% normative nepheline), with normative nepheline contents positively correlated with CaO contents (Figure 6). They thus differ from the CaO-rich compositions found as olivine-hosted melt inclusions and whole rocks in oceanic island [Tronnes, 1990], mid-ocean ridge [Kamenetsky *et al.*, 1998; Sours-Page *et al.*, 1999], and back arc [Kamenetsky *et al.*, 1997] settings, all of which are hypersthene normative (Figure 6). The CaO-rich inclusions and whole rocks also have highly variable CaO/Al₂O₃ ratios (0.8–1.4) and silica contents at the low end (44–48.5 wt %) of the distribution of arc lavas. The CaO-rich whole rocks are characterized by relatively high MgO (>6.5 wt %) contents, and although MgO contents of inclusions are sensitive to the correction for

postentrapment crystallization, homogenized CaO-rich inclusions are also MgO-rich (≥ 7.5 wt %). Note that although the whole rocks are clinopyroxene-phyric (diopside-augite [Foden and Varne, 1980; Foden, 1983; Kennedy *et al.*, 1990; Thirwall and Graham, 1984]), their elevated CaO contents and silica undersaturation cannot be attributed to accumulation of pyroxene alone because their accumulated clinopyroxenes are not nepheline normative [Foden and Varne, 1980; Foden, 1983; Kennedy *et al.*, 1990; Carr and Rose, 1984; Thirwall and Graham, 1984; Shimizu and Arculus, 1975; Arculus, 1976], and thus mixtures of hypersthene- or quartz-normative liquids and such clinopyroxenes would not be nepheline normative, regardless of their CaO contents. Similarly, dissolution by melt inclusions of clinopyroxenes with compositions in the range observed for phenocrysts in the host lavas, either prior to or during entrapment, cannot contribute to their nepheline-normative character.

[16] Nepheline-normative, high-CaO melt inclusions occur in lavas ranging from relatively rare ankaramites that are also CaO-rich and nepheline normative to more normal hypersthene- or quartz-normative basalts and basaltic andesites with relatively low-CaO (<13 wt %) contents. Compositional and isotopic differences between melts preserved in olivine phenocrysts and their host lavas have been observed previously and in a variety of settings [e.g., Sobolev and Shimizu, 1993; Gurenko and Chaussidon, 1995; Nielsen *et al.*, 1995; Clocchiatti *et al.*, 1998; Saal *et al.*, 1998; Shimizu *et al.*, 1998]. In these cases, melt inclusions have been interpreted as sampling a range of liquids (reflecting a range of source compositions, extents of melting, and/or melting processes) trapped before mixing and homogenization in magma chambers diluted or averaged them to produce the compositions of erupted lavas. In

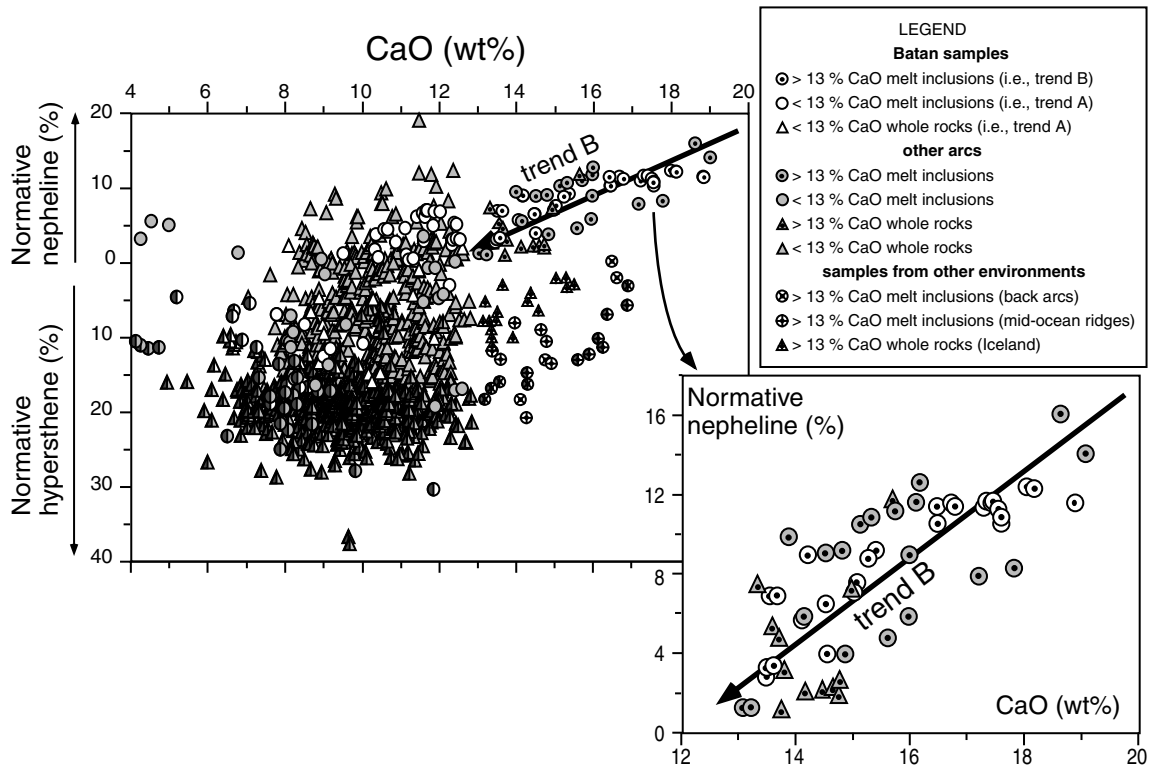


Figure 6. Plot of normative abundances of nepheline and hypersthene versus CaO concentrations comparing Batan CaO-rich melt inclusions (i.e., trend B) and CaO-rich melt inclusions and whole rocks from other arcs (all of which have CaO > 13 wt %) with “normal” melt inclusions and whole rocks from Batan and other arcs (all of which have CaO contents < 13 wt %) (same compilation as in Figure 5). Also shown in this diagram are olivine-hosted CaO-rich melt inclusions from back arc and mid-ocean ridge settings [Kamenetsky *et al.*, 1998; Sours-Page *et al.*, 1999; Kamenetsky *et al.*, 1997] and CaO-rich lavas from Iceland [Tronnes, 1990]. Symbols are given in the accompanying legend. Half-solid symbols indicate samples with normative quartz. Major element compositions have been normalized to 100%, and norms were calculated assuming $\text{Fe}^{3+} = 0.15 \sum \text{Fe}$.

this context, the occurrences of CaO-rich, nepheline-normative melt inclusions in “normal” host lavas and in environments in which whole rocks with comparable compositions have not been identified (Figure 4) demonstrate that the conditions required to produce melts of this sort are more common in subarc environments than might be apparent from the abundance of comparable whole rocks and that such melts contribute more widely to island arc magmatism than their

low abundance as erupted magmas might otherwise suggest.

3. Comparison of the Composition of CaO-Rich Melts With Experimental Melts of Iherzolite

[17] The chemical compositions of high-CaO melt inclusions and whole rocks are unusual relative to typical arc magmas and to basaltic



magmas generally, suggesting that production of such magmas requires something other than partial melting of lherzolitic sources. Experimental studies of peridotite melting constrain the range of possible chemical compositions of primary magmas formed over a range of pressures (1 atm to ~ 100 kbar), degrees of melting (between ~ 2 and 80%), and bulk compositions of the peridotite sources (plagioclase, spinel, and garnet-bearing peridotites) [Ito and Kennedy, 1967; Mysen and Kushiro, 1977; Jaques and Green, 1980; Hirose and Kushiro, 1993; Baker and Stolper, 1994; Kushiro, 1996; Walter, 1998; Kinzler and Grove, 1992a, b; Stolper, 1980; Takahashi and Kushiro, 1983; Fujii and Scarfe, 1985; Falloon and Green, 1987; Walter and Presnall, 1994; Baker et al., 1995; Kinzler, 1997; Wasylenki, 1998; Hirose and Kawamoto, 1995; Hirose, 1997a; Green, 1973; Kushiro, 1972; Gaetani and Grove, 1998]. Over this range of variables, CaO concentrations reported for partial melts of dry and hydrous peridotite range from 4 to 14 wt %. When experiments on melting of lherzolite in the presence of CO₂ are included, this range extends to CaO contents >20 wt % [e.g., Dalton and Presnall, 1998]. In this section, we briefly review how variations in CaO content (and normative nepheline versus hypersthene) of partial melts of lherzolites relate to changes of pressure, temperature, and source composition (including H₂O and CO₂ content).

[18] Under volatile-free conditions, melts of typical lherzolitic sources can under certain conditions become decreasingly hypersthene normative and even nepheline normative with increasing pressure [Green and Ringwood, 1967; O'Hara, 1968]; however, they also typically become poorer in CaO, reflecting the decreasing CaO content of clinopyroxene with increasing solidus temperature [e.g., Longhi, 1995]. As a result, the effects of

pressure alone are unlikely to be capable of extending upward the range of CaO contents of direct melts of lherzolites so as to account for the high CaO content of the magmas in which we are interested or its correlation with normative nepheline. At a given pressure, CaO concentrations (and CaO/Al₂O₃ ratios) in partial melts of lherzolites increase with increasing degree of melting from the solidus and reach a maximum value when clinopyroxene is exhausted from the residue, beyond which further melting leads to decreases in the CaO contents of the partial melts. The maximum CaO contents of liquids produced by volatile-poor experimental melting of fertile lherzolites (~ 13 wt % [Jaques and Green, 1980; Kinzler and Grove, 1992b; Hirose and Kushiro, 1993; Baker and Stolper, 1994; Kushiro, 1996; Walter, 1998]) thus correspond to the relatively high (up to 25%) degrees of melting typically required for clinopyroxene exhaustion. Although this maximum can increase to 14 wt % CaO at clinopyroxene-out for more depleted peridotites [Wasylenki, 1998] (reflecting the low Na₂O contents of the partial melts of depleted peridotites relative to comparable melt fractions of fertile peridotite [Wasylenki, 1998; Hirschmann et al., 1998]), it is still well below the CaO contents of the CaO-rich inclusions and lavas identified here. Moreover, liquids produced by the high degrees of melting required to exhaust clinopyroxene from typical lherzolitic sources are not nepheline normative.

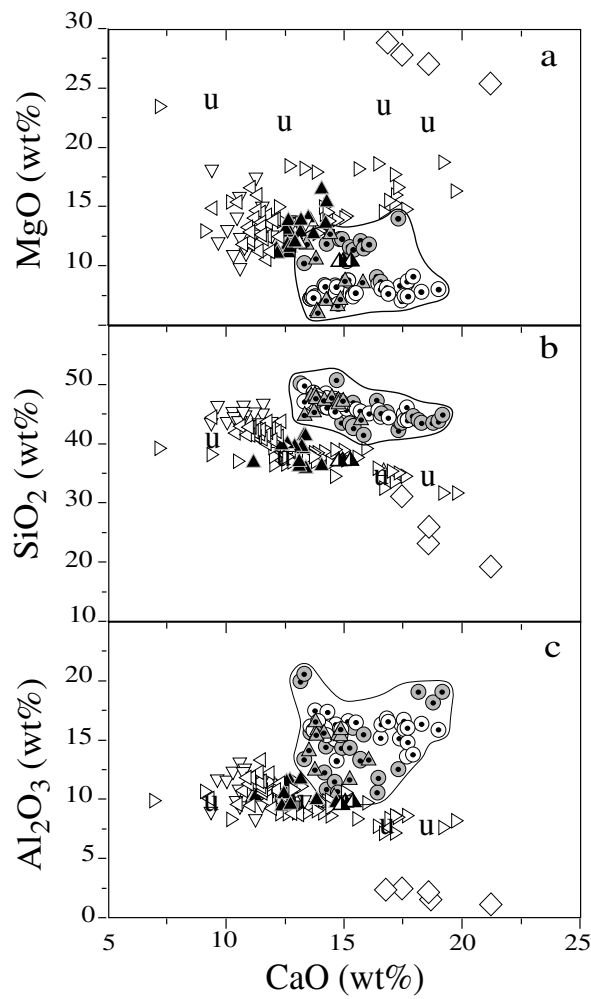
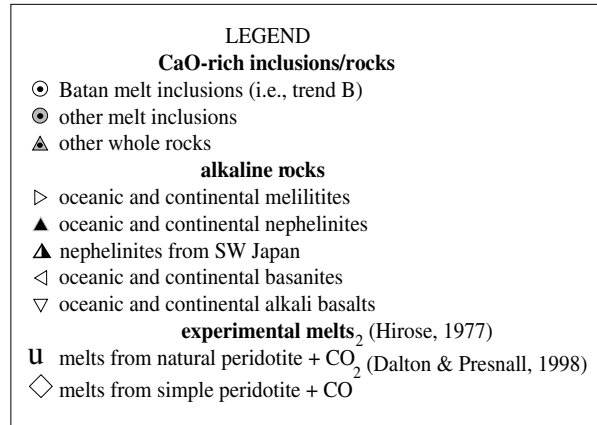
[19] Gaetani and Grove [1998] showed that increases in the H₂O content of partial melts of lherzolite at constant pressure and decreasing temperature are associated with decreases in the concentrations of SiO₂, FeO, and MgO and with increases in SiO₂/(MgO + FeO) ratios. Consequently, hydrous peridotite melts are SiO₂-rich when compared to anhydrous melts on a volatile-free basis. In addition, melting of lherzolite under hydrous conditions decreases



the CaO concentration of melts at a given melt fraction relative to dry melting [Hirose and Kawamoto, 1995; Hirose, 1997a]. Consequently, melting of water-bearing lherzolite is not a plausible explanation for the origin of the CaO-rich, silica-undersaturated melts identified here. This is consistent with the observation that CaO-rich, silica-undersaturated olivine-hosted melt inclusions in basalts from Galunggung have low H₂O contents (0.2–0.4 wt %) [Sisson and Bronto, 1998].

[20] With the exception of the Lesser Antilles, all the arcs where the CaO-rich melt inclusions and whole rocks have been identified have CO₂-rich sediment subducting [Plank and Langmuir, 1998]. Moreover, at pressures above 25 kbar, melting of lherzolite in the presence of CO₂ (or H₂O + CO₂, with X_{CO₂} > 0.5 [see Eggler, 1978]) produces SiO₂-poor liquids with high CaO contents (>20 wt %) and CaO/Al₂O₃ ratios [Dalton and Presnall, 1998; Eggler, 1978; Huang and Wyllie, 1974; Eggler, 1974; Adam, 1988; Hirose, 1997b]. Therefore relatively high pressure melting of carbonated peridotite could play a role in generating the CaO-rich, silica-undersaturated melts identified in this study [DellaPasqua and Varne, 1997]. However, we consider this unlikely based on the significant differences between the CaO-rich, silica-undersaturated arc magmas identified in this study and both experimental melts obtained at high pressures for natural and simple carbonated peridotite systems [Dalton and Presnall, 1998; Hirose, 1997b] and primitive melilitites, nephelinites, and related rocks from oceanic, continental, and arc regions [Alibert et al., 1983; Dupuy et al., 1989; Wedephol et al., 1994; Wilson et al., 1995; Maaloe et al., 1992; Hoernle and Schmincke, 1993; Clague and Frey, 1982; Cheng et al., 1993; Tatsumi et al., 1999], which are widely considered to be melts of carbonated peridotite [Eggler, 1978; Huang and Wyllie, 1974; Eggler, 1974; Adam, 1988; Hirose, 1997b; Brey and Green, 1975; Brey, 1978; Wallace and Green,

1988]. For example, the CaO-rich, silica-undersaturated arc magmas identified here have higher SiO₂ and Al₂O₃ and lower MgO contents at a given CaO content than do the experimental melts obtained at high pressures for natural and simple carbonated peridotite systems (Figure 7). They also show systematic and significant compositional differences from primitive alkali olivine basalts, basanites, olivine nephelinites, and melilitites: that is, although melilitites and nephelinites have CaO contents comparable to those of the inclusions and whole rocks we have studied, the SiO₂ contents of these highly alkalic lavas are typically 5–15 wt % lower than those of the CaO-rich arc magmas. In addition, melilitites and nephelinites (and carbonatites [Nelson et al., 1988]) are characterized by very high concentrations of highly incompatible elements and very high LREE/HREE ratios relative to melt inclusions from B45 sample from Batan (Figure 2b). Finally, CaO-rich, silica-undersaturated melt inclusions and basaltic melt inclusions have similar CO₂ contents (500–750 ppm) in Galunggung basalts [Sisson and Bronto, 1998]. Note also that although direct incorporation of carbonate rocks by the primitive arc magmas identified here can account for their elevated CaO contents, it cannot contribute to their nepheline-normative character. Moreover, as Sr and CaO partition strongly into marine carbonates, Sr should accompany any Ca contributed by marine carbonate sediments and carbonate-derived Sr is easily distinguished from mantle-derived Sr by its radiogenic seawater-derived isotopic composition. However, the CaO-rich and CaO-poor lavas from Lombok (Sunda arc), Rindjani (Sunda arc), and Lihir (New Guinea) have undistinguishable ⁸⁷Sr/⁸⁶Sr ratios [Foden and Varne, 1980; Kennedy et al., 1990] and in the case of Grenada (Lesser Antilles), the CaO-rich lavas have lower Sr isotopic ratios than the CaO-poor series [Hawkesworth et al., 1979; Thirwall and Graham, 1984]. We therefore conclude that carbonate contribution cannot be



reconciled with the composition of the CaO-rich melts identified here.

[21] In summary, we are aware of no conditions under which partial melting of peridotite generates melts having CaO contents and other properties comparable to those observed for CaO-rich melts considered in this study. In particular, only melting at high pressures in the presence of significant CO₂ appears capable of producing liquids with the required CaO contents, but such liquids are significantly different from the CaO-rich arc melts in nearly all other compositional characteristics. In section 4, we develop the hypothesis that the presence of CaO-rich, silica-undersaturated melts similar to the primitive end member of trend B (as defined by the Batan melt inclusions) in arc environments results from the partial melting of clinopyroxene-rich lithologies (e.g., eclogites and/or pyroxenites).

4. Possible Origin of the CaO-Rich, Silica-Undersaturated Arc Melts As Melts of Pyroxene-Rich Sources

[22] In recent years, there have been many suggestions that melting of mafic, pyroxene-rich lithologies may play a significant role in the petrogenesis of basaltic magmas [Allègre and Turcotte, 1986; Hirschmann and Stolper, 1996; Hauri, 1996; Sigmarsson et al., 1998]. Motivations for these suggestions include evidence for heterogeneous sources [Hanson,

1977; Wood, 1979; Zindler et al., 1984; Prinzhofer et al., 1989] (i.e., melting of a single peridotitic source cannot explain the compositional or isotopic variability of suites of related magmas), the fact that pyroxenites and eclogites are an observed component of mantle and deep, crustal rocks [see Hirschmann and Stolper, 1996, and reference therein], and the fact that as the plutonic equivalents of basaltic magmas, they are readily envisioned as a component at depth that can be introduced into other sources as a heterogeneity [e.g., Allègre and Turcotte, 1986]. Moreover, subduction of the oceanic crust introduces basaltic components into the mantle, and this provides a plausible mechanism for introducing mafic heterogeneities into largely ultramafic mantle sources. In the context of the high-CaO magmas identified here, an additional reason for thinking about pyroxene-rich source components is that such rocks can have very high CaO contents (see below), and thus high-degree melts of such sources (and perhaps low-degree melts as well) can have CaO contents much higher than melts of peridotite.

[23] In this section, we evaluate in more detail which, if any, naturally occurring pyroxene-rich lithologies are plausible sources of the distinctive magma types identified here. We examine the hypothesis that the primitive CaO-rich, silica-undersaturated magmas identified here reflect partial melting of mafic material by (1) comparison of the major ele-

Figure 7. CaO variation diagrams comparing data for Batan CaO-rich melt inclusions (trend B) and CaO-rich melt inclusions and whole rocks from other arc environments with liquids produced by experiments on natural (lherzolite + 2.5 wt % CO₂ at 30 kbar [Hirose, 1997b], solid diamond) and simple (lherzolite-saturated liquids in the CaO-MgO-Al₂O₃-SiO₂-CO₂ system at 60 kbar [Dalton and Presnall, 1998], open diamond) carbonated peridotites at high pressure and with primitive (FeO* < MgO in wt %) alkali olivine basalts, basanites, olivine nephelinites, and olivine melilitites from oceanic and continental regions [Alibert et al., 1983; Dupuy et al., 1989; Wedephol et al., 1994; Wilson et al., 1995; Maaloe et al., 1992; Hoernle and Schmincke, 1993; Clague and Frey, 1982; Cheng et al., 1993]. Also reported are nephelinites from SW Japan [Tatsumi et al., 1999]. Symbols are given in the accompanying legend.

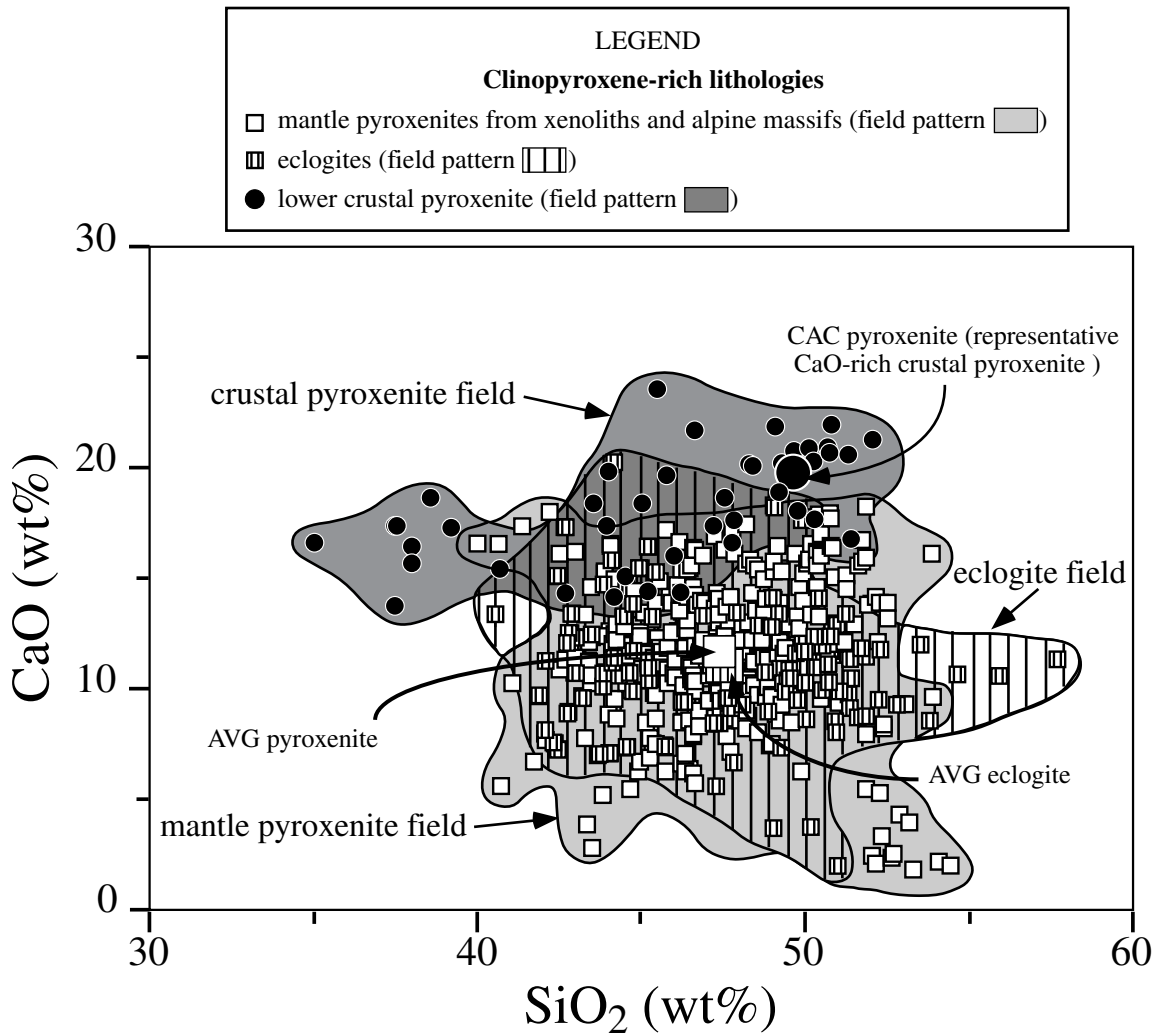
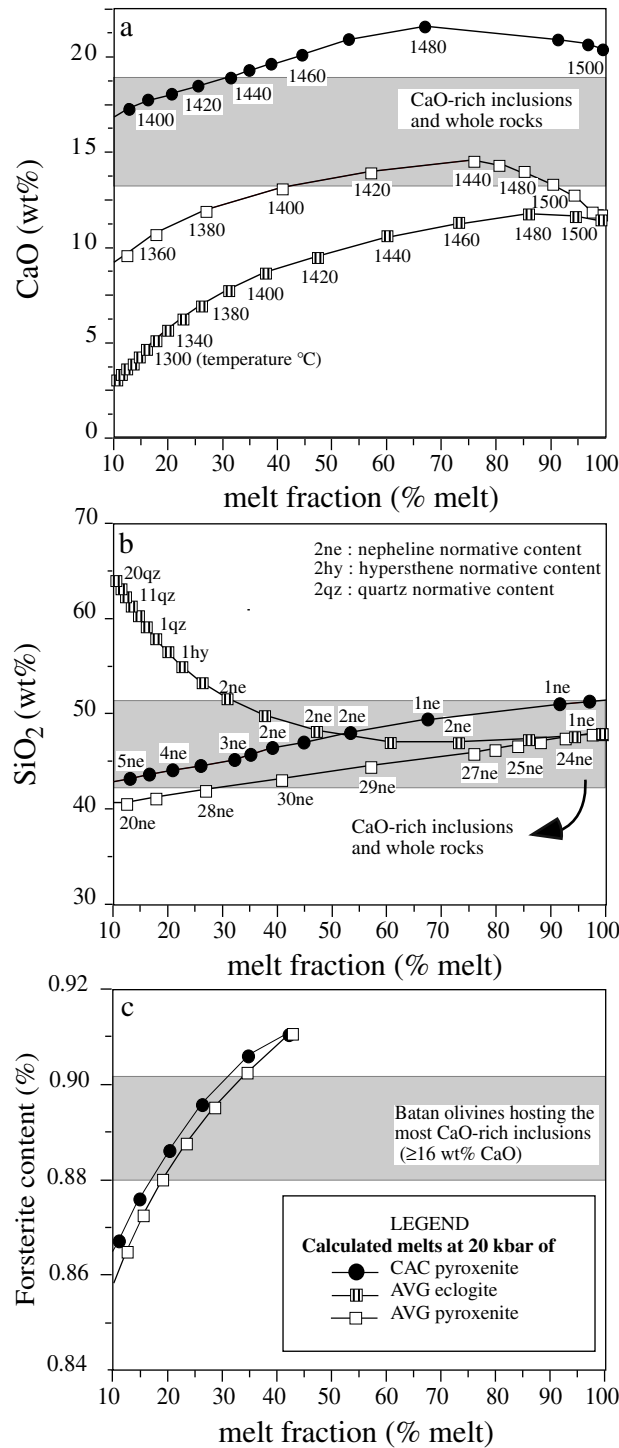


Figure 8. CaO versus SiO₂ concentrations for mantle pyroxenites (open squares) from xenoliths and alpine massifs (compilation by *Hirschmann and Stolper* [1996]), eclogites (dashed squares, compilations from *Hirschmann and Stolper* [1996] and R. L. Rudnick (personal communication, 1999)), and lower crustal pyroxenite cumulates (solid circles) from arc environments [*Irvine, 1973; Snoke et al., 1981; Wyllie, 1967; Rucknick and Noble, 1959; Findlay, 1969; James, 1971*]. Eclogites and pyroxenites were distinguished based on what they were named in the literature. Also shown are the compositions of the average pyroxenite (AVG pyroxenite, corresponding to the mean of the mantle pyroxenite population), average eclogite (AVG eclogite, corresponding to the mean of the eclogite population), and representative CaO-rich crustal pyroxenite (CAC pyroxenite, from Duke Island [*Irvine, 1973*]) used in the MELTS calculations (Figures 9 and 10).

ment compositions of the arc-derived CaO-rich inclusions and whole rocks with model compositions of partial melts of pyroxenite and eclogite calculated using the MELTS algorithm

[*Ghiorso, 1994; Ghiorso and Sack, 1995*] and (2) comparison of the trace element compositions of the arc-derived CaO-rich samples with the expected trace element compositions of





melts of model mantle peridotite, eclogite, and pyroxenite.

4.1 Model Major Element Compositions of Partial Melts of Pyroxenite and Eclogite

[24] Although MELTS is not likely to predict accurately the melting relations of pyroxenites, we, nevertheless, use it as a guide to what to expect because there is such a wide range of pyroxenite and eclogite compositions (Figure 8) and such a small number of melting experiments on such rocks. We acknowledge, however, that the results of these MELTS calculations are at best only semiquantitative at this point and that they must be interpreted cautiously. As shown in Figure 9, MELTS-calculated CaO contents of partial melts of average pyroxenite and eclogite at 20 kbar increase from values at the solidus lower than the concentration in the source to a maximum higher than the concentration in the source, then decrease to the concentration in the source when the liquidus is reached; the maximum in predicted CaO content occurs at very high degrees of melting. This predicted behavior is typical of the pyroxenite and eclogite bulk compositions shown in Figure 8, and it is also predicted for the most CaO-rich pyroxenites (e.g., the representative crustal pyroxenite composition from Duke Island shown in Figure 8). Note that the maximum CaO contents of liquids produced by melting of average mantle

pyroxenite and eclogite are, however, still below the CaO contents of the most CaO-rich inclusions and lavas identified in this study, and, in particular, they are lower than the CaO-rich end of trend B defined for the Batan samples. Therefore, according to MELTS, in order to generate melts with CaO contents as high as those required by the most CaO-rich inclusions and lavas at melt fractions of several tens of percent or less, the mafic source would have to be CaO-rich. An important aspect of the MELTS calculations is that even though the average pyroxenite and eclogite sources have similar CaO and SiO₂ contents (Figure 8), melts of average pyroxenite are predicted to have significantly higher CaO and lower SiO₂ contents than melts of average eclogite up to high degrees of melting; the predicted divergent behavior of SiO₂ at low melt fractions (i.e., to very high SiO₂ for low degree melts of the average eclogite and to very low SiO₂ for low degree melts of the average pyroxenite) is particularly striking in Figure 9b. It should also be noted that to the degree to which the pyroxenites are similar in compositions to basalts (or even picrites), they have higher Fe²⁺/Mg ratios than mantle peridotites, and thus the first liquids they produce on partial melting will have higher Fe²⁺/Mg ratios than liquids that could be produced by melting of peridotites. Therefore olivine that could coexist with the low degree melts of the pyroxenites must have much lower forsterite content than the typical

Figure 9. Concentrations of CaO and SiO₂ in batch partial melts of the anhydrous AVG pyroxenite (open squares), AVG eclogite (dashed squares), and CAC pyroxenite (solid circles), calculated as a function of melt fraction using the MELTS algorithm at 20 kbar [Ghiorso, 1994; Ghiorso and Sack, 1995]. Numbers on curves in Figure 9a refer to calculated temperatures (degrees Celsius) of melting (corrected for the overestimation of 100°C due to the overprediction of the stability of orthopyroxene relative to olivine, see Hirschmann *et al.* [1998]); numbers on curves in Figure 9b refer to nepheline, hypersthene, and quartz normative contents of the calculated melts. The ranges of CaO and SiO₂ contents for the CaO-rich inclusions and whole rocks defined in this study are shown for comparison. (c) Forsterite content of residual olivine of the MELTS-calculated partial melts of AVG pyroxenite and CAC pyroxenite versus melt fraction at 20 kbar. The range of forsterite contents for Batan olivines hosting the most CaO-rich inclusions (i.e., the end member of Batan trend B) is shown for comparison.

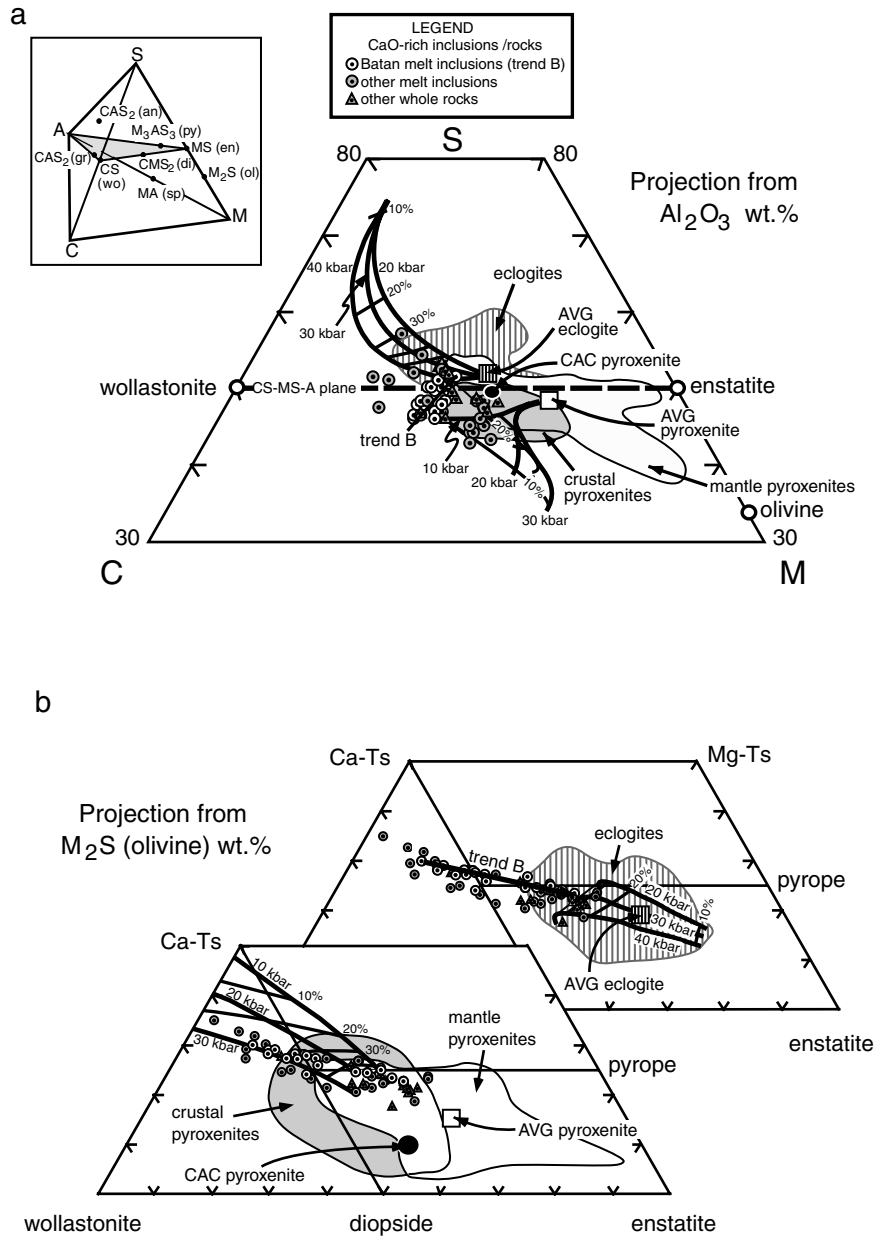


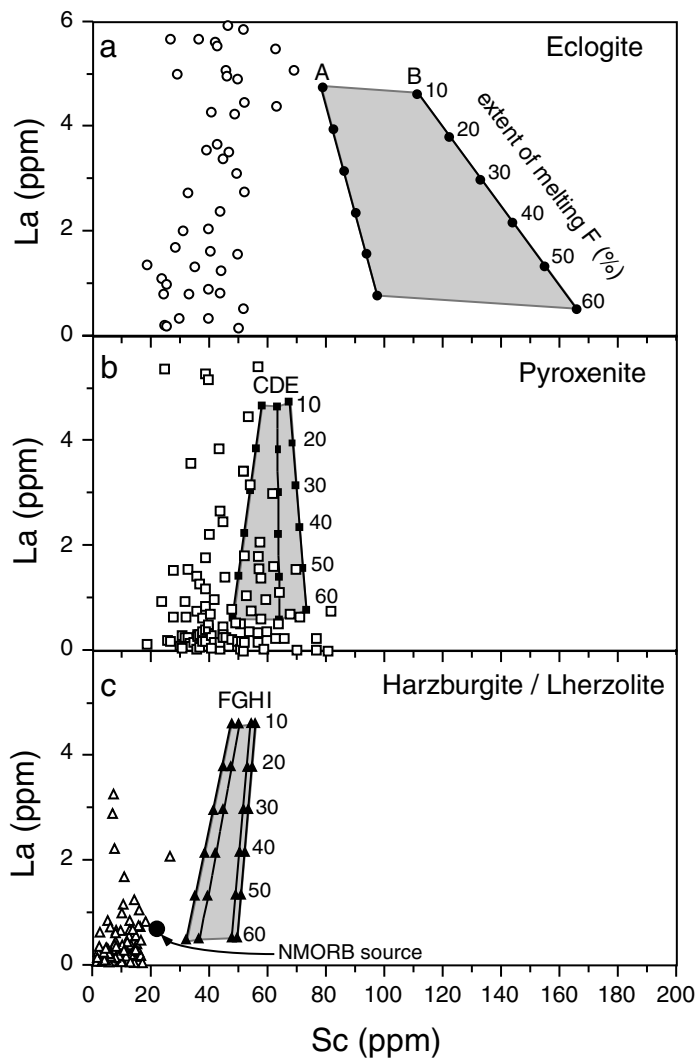
Figure 10. Compositions of CaO-rich melt inclusions from Batan (i.e., trend B) and CaO-rich melt inclusions and whole rocks from other arc environments recalculated to equivalent CaO-MgO-Al₂O₃-SiO₂ (C-M-A-S) and then projected (a) from alumina onto the S-C-M plane and (b) from M₂S (olivine) onto the CS-MS-A (wollastonite-enstatite-alumina) plane. Symbols are given in the accompanying legend. Melting grids calculated using MELTS for AVG pyroxenite (at 10, 20, and 30 kbar) and AVG eclogite (at 20, 30, and 40 kbar), and fields drawn around mantle pyroxenites, eclogites, and lower crustal pyroxenite cumulates (references in caption to Figure 8) are also shown. Figure 10a shows that according to MELTS the CS-MS-A plane is a thermal divide at 10–40 kbar, in agreement with *O'Hara* [1968]: that is, melting (or crystallization) paths of compositions on opposite sides of this plane diverge. (insert) Relationships of planes and projection points in the C-M-A-S system.



Fo₉₀ of mantle peridotites. However, the Fe²⁺/Mg ratio of the partial melts of the pyroxenite will decrease with increasing melt fraction, and thus if the melt fraction is high enough, the forsterite content of the olivine that could crystallize from the partial melts can be as magnesian as Fo₉₀ (Figure 9c).

[25] The compositions of partial melts of average eclogite and mantle pyroxenite calculated by MELTS and the compositions of natural CaO-rich, silica-undersaturated melt inclusions and whole rocks have been recast into the C-M-A-S system [O'Hara, 1968; *Basaltic Volcanism Study Project*, 1981] and then projected from Al₂O₃ onto the S-C-M plane (Figure 10a) and from M₂S (olivine) onto the CS-MS-A plane (Figure 10b). Figure 10a shows that the average pyroxenite and eclogite compositions are on opposite sides of the CS-MS-A plane. Since this plane is a thermal divide with respect to pyroxene and/or garnet crystallization [O'Hara, 1968], melting and crystallization paths of compositions on opposite sides of this plane diverge: Model partial melts of compositions below this plane (i.e., plotting on the silica-poor side of the wollastonite-enstatite join in Figure 10a, as does the average mantle pyroxenite and the representative CaO-rich crustal pyroxenite) produce silica-poor liquids at low degrees of melting, whereas compositions above this plane (i.e., plotting on the silica-rich side of wollastonite-enstatite join in Figure 10a, as does the average eclogite) produce silica-rich liquids at low degrees of melting. The fact that the average eclogite and pyroxenite compositions are on opposite sites of the CS-MS-A thermal divide thus explains the strongly contrasting trends in silica for these two compositions shown in Figure 9b. Note that because the CS-MS-A plane is a thermal divide, partial melts of pyroxene-rich lithologies are confined to one or the other side of this plane, so the fact that the CaO-rich, nepheline-normative melt inclusions (including those defining trend B

in Figure 1d) and whole rocks extend across this plane (Figure 10a) confirms our earlier assertion that Batan trend B is not consistent with progressive isobaric melting of pyroxenite, thus leading us to favor a magma mixing origin for this trend. The primitive (i.e., most CaO-rich and nepheline-normative) end of array of CaO-rich magmas plots below the thermal divide, and based on the MELTS calculations, liquids grossly similar to this end of the array could be generated by partial melting of the average mantle pyroxenite (Figures 9 and 10). However, although MELTS calculations based on the average pyroxenite composition can just barely produce partial melts with CaO contents as high as those in trend B (Figure 9a), natural pyroxenites vary substantially in composition, and partial melts similar in CaO content to natural CaO-rich melts considered in this study and in equilibrium with Mg-rich olivine (Figures 9a and 9c) could be generated by intermediate to high degrees of partial melting of a specific subset of natural CaO-rich pyroxenites (lower crustal pyroxenite cumulates such as the representative Duke Island pyroxenite; see section 4.3) at a few tens of kilobars. Moreover, MELTS calculations shows that the FeO contents in the partial melts of pyroxenites are strongly dependent on pressure; i.e., higher FeO melts are produced at higher pressure. Therefore the low FeO content of the CaO-rich, silica-undersaturated melt inclusions and whole rocks relative to "normal" island arc lavas (Figure 5) could reflect their relatively low pressure of formation. When recast into the C-M-A-S system and projected onto the planes of Figure 10, melts of the representative CaO-rich crustal pyroxenite composition shown in Figures 8 and 9 produce at a given pressure liquids similar to the end of the trend B array at lower melt fractions (10–30%) than the average mantle pyroxenite. It should also be noted that most eclogites plot on the silica-rich side of the thermal divide in Figure 10a and thus are less suitable than pyr-



- eclogitic residues
 - A: 85% cpx and 15 % gt
 - B: 25% cpx and 75 % gt
- pyroxenitic residues
 - C: 40% ol and 60 % cpx
 - D: 10% ol, 50% opx and 40% cpx
 - E: 85% cpx and 15% sp
- natural and synthetic lherzolite residues
 - F (KLB-1): 58% ol, 25% opx, 15% cpx and 2% sp
 - G (MM3 at 10 kbar): 50% ol, 30% opx, 17% cpx and 3% sp
 - H (MM3 at 36 kbar): 56.3% ol, 11.6% opx, 21.4% cpx and 10.7% gt
 - I (DMM1): 64% ol, 27% opx, 7% cpx and 2% sp



oxenites (which typically plot on the silica-poor side of the CS-MS-A plane) as potential sources of CaO-rich, nepheline-normative magmas.

4.2 Trace Element Compositions of Melts of Model Mantle Peridotite, Eclogite, and Pyroxenite

[26] The trace element compositions of liquids at the most CaO-rich, nepheline-normative end of trend B melt inclusions from Batan offer a potential test of the hypothesis that they can be produced by partial melting of pyroxenite. To do this, we have calculated the concentrations of Sc (a typical compatible trace element in clinopyroxene) and La (a typical incompatible trace element) required in the sources of the most CaO-rich melt inclusions from Batan Island, assuming batch melting of those sources, and either pyroxenitic, eclogitic, lherzolitic, or harzburgitic residual assemblages (regardless of major element evidence for or against these possible sources presented above). The measured Sc and La concentrations of the most

CaO-rich, nepheline-normative melts are 62 and 8.3 ppm (Table 1; the assumed mineral/melt partition coefficients and residual mineralogies are given in the caption of Figure 11). The calculated compositions of sources consistent with the Sc and La contents of the CaO-rich end of trend B and the Sc and La concentrations of natural mafic and ultramafic rocks [Bodinier *et al.*, 1987; Bodinier, 1988; Beard *et al.*, 1992; Griffin *et al.*, 1988; Gil Ibarra *et al.*, 1989; Irving, 1974; Kurat *et al.*, 1993; Pearson *et al.*, 1993; Suen and Frey, 1987; Taylor and Neal, 1989; Van Calsteren, 1978] are compared in Figure 11. As shown in Figures 11a and 11c, the source compositions required if eclogitic, lherzolitic, or harzburgitic assemblages are assumed are significantly higher in Sc than any naturally occurring mantle eclogites, lherzolites, or harzburgites. In contrast, as shown in Figure 11b, if a pyroxenitic residue is assumed, there is overlap between the required model source compositions and naturally occurring mantle pyroxenites for both Sc and La. These results are consistent with the hypothesis that the

Figure 11. Comparison of the calculated compositions of sources consistent with the Sc and La contents (62 and 8.3 ppm, Table 1) of the most CaO-rich melt inclusions from Batan Island (assuming variable degrees of batch melting of those sources, that the proportions of residual minerals do not change with the degree of melting, and either mafic or ultramafic residual assemblages) with the Sc and La concentrations of natural pyroxenites (open squares), eclogites (open circles), and fertile and depleted lherzolites (open triangles) rocks [Bodinier *et al.*, 1987; Bodinier, 1988; Beard *et al.*, 1992; Griffin *et al.*, 1988; Gil Ibarra *et al.*, 1989; Irving, 1974; Kurat *et al.*, 1993; Pearson *et al.*, 1993; Suen and Frey, 1987; Taylor and Neal, 1989; Van Calsteren, 1978]. Shaded regions represent the ranges of calculated source composition for (a) eclogitic, (b) pyroxenitic, and (c) lherzolitic or harzburgitic residual assemblages, assuming modal phase abundance (given in the accompanying legend) that spans most of the range known in mafic and ultramafic rocks (modal phase abundance for KLB-1 from Hirose and Kushiro [1993], MM3 at 10 kbar from Baker and Stolper [1994], MM3 at 36 kbar from M. Baker (personal communication), and DMM1 from Wasylenki [1998]); the lines in each diagram are the calculated trends of Sc and La concentrations required in these residual assemblages, assuming batch melting of those sources and the numbers along the lines indicate the degree of melting. The concentrations of Sc and La required in the sources were calculated using the general equation of batch melting $C_L = C_0/[D(1 - F) + F]$, where D is the bulk solid/melt partition coefficient weighted by the proportions of the residual solid phases, C_L and C_0 are the concentrations in the liquid and source, respectively, and F is the melt fraction. $D_{\text{solid/melt } i}$ values used in the calculations are for eclogitic, pyroxenitic, and lherzolitic residues assuming the modal phase abundance given in the accompanying legend. Individual mineral melt partition coefficients were compiled from the Geochemical Earth Reference Model (GERM, <http://www-ep.es.lnl.gov/germ>). Also shown for comparison is the composition of the source of average NMORB estimated by Hofmann [1988] based on a crust–upper mantle mass balance.



most CaO-rich, nepheline-normative melts in arc environments are produced by several tens of percent partial melting of pyroxenitic sources and they contribute to evidence against their derivation from melting peridotitic or eclogitic sources.

[27] In summary of this and the previous section, thermodynamic calculations of major element compositions and trace element modeling suggest that the most CaO-rich, nepheline-normative melt inclusions and whole rocks identified in this study are consistent with intermediate to high degree ($\sim 10\text{--}40$ wt %) partial melts of pyroxenites at lower crustal to upper mantle pressures (i.e., up to a few tens of kilobars). If this interpretation is correct, although they are rare as erupted magmas, the widespread geographic occurrence of such melts in arc environments as either melt inclusions or whole rocks would represent previously unrecognized evidence of a widespread contribution to arc magmas of these compositionally distinctive partial melts of mafic material.

4.3. Mantle Pyroxenites Versus Lower Crustal Pyroxenites

[28] MELTS calculations predict that temperatures required to generate CaO-rich melts from anhydrous mantle pyroxenite at 10–30 kbar would be very high ($\geq 1400^\circ\text{C}$) (Figure 9), a result also supported by preliminary experimental melting of model clinopyroxenites at 10 and 25 kbar [Hirschmann *et al.*, 1995; Hirschmann and Schiano, 1999]. Thermal models of arcs [Davies and Stevenson, 1992; Furukawa, 1993] predict that such high temperatures are present only at 100 km and deeper below the volcanic arc. Such temperatures are also much higher than the homogenization temperatures obtained for the CaO-rich, silica-undersaturated melt inclusions trapped in olivine (typically $1220^\circ\text{--}1260^\circ\text{C}$; see *Metrich et al.* [1999], *Della-Pasqua and*

Varne [1997], *Gioncada et al.* [1998], and this study), and although the homogenization temperature is formally only a minimum estimate of the entrapment temperature, these low temperatures are also consistent with equilibrium between trapped melt and the host [Roedder, 1984; Schiano and Bourdon, 1999; Lowestern, 1994; Student and Bodnar, 1996].

[29] As we emphasized above, although model melts of pyroxenite are predicted to have CaO concentrations higher than the bulk pyroxenite at high degrees of melting (Figure 9a), the remarkably high CaO contents of the primitive end member of Batan trend B (up to 18–20 wt %) and the existence of a coherent, well-defined high CaO (>14 wt %) group of melt inclusions and whole rocks based on this study would be most easily explained by source compositions characterized by CaO concentrations at the high end of the very large range of CaO contents found in natural pyroxenite. In other words, as shown in Figure 8, mantle-derived pyroxenites are chemically diverse, spanning a range in CaO contents from 18.5 wt % to <5 wt % [Hirschmann and Stolper, 1996], and MELTS calculations predict that partial melting of this range of pyroxenites in the upper mantle would generate mostly nepheline-normative melts with similarly variable CaO concentrations, in contrast with the relatively narrow compositional range of CaO-rich, silica-undersaturated melts identified here from eight arcs around the world. Therefore the characteristics of these CaO-rich melts, when combined with the preceding arguments in favor of their derivation from partial melting of pyroxene-rich lithologies on the silica-poor side of the CS-MS-A thermal divide, suggest that they preferentially sample an unusually CaO-rich subset of the full range of naturally occurring pyroxenites.

[30] Clinopyroxene-rich rocks are found in the lower crust beneath exhumed volcanic arcs and



as plutonic xenoliths in arc lavas [Murray, 1972; Irvine, 1973; Snoke *et al.*, 1981; Conrad and Kay, 1984; Kay and Kay, 1985; Ducea and Saleeby, 1996; DeBari and Coleman, 1989; DeBari *et al.*, 1987]. They occur as zoned ultramafic complexes [see Wyllie, 1967, and references therein], typified by the Duke Island ultramafic complex in the Alaskan orogenic belt [Irvine, 1973]. These zoned ultramafic complexes have been interpreted as cumulates crystallized from primitive island arc magmas that form layered complexes of hornblende-olivine pyroxenites and less abundant gabbros and diorites [e.g., Irvine, 1973; Snoke *et al.*, 1981; Conrad and Kay, 1984; Kay and Kay, 1985; Ducea and Saleeby, 1996; DeBari and Coleman, 1989; DeBari *et al.*, 1987]). The hornblende-olivine pyroxenites in such complexes are characterized by relatively high CaO concentrations (between 13 and 24 wt %), i.e., at the upper end of the range observed for mantle pyroxenites (Figure 8), so melting of these lower crustal lithologies at relatively low pressure to produce the CaO-rich arc magmas (see Figure 9) (rather than the highly variable mantle pyroxenites and eclogites) could thus provide a simple explanation for the restricted, relatively uniform composition of these CaO-rich (and FeO-poor) magmas worldwide. Furthermore, the common presence of amphibole in these pyroxenites would lower their solidus temperatures relative to the anhydrous pyroxenite equivalents we used in the MELTS calculations, therefore at least partially resolving the apparent discrepancy (described at the start of this section) between the inferred temperatures of the CaO-rich, silica-undersaturated melts and the high temperatures required by anhydrous MELTS calculations to produce partial melts of pyroxenites comparable to the trend B compositions. Given the known existence of such cumulates at lower crustal and shallow upper mantle depths beneath arcs [Ducea and Saleeby, 1996], their appropriateness as potential sources of CaO-rich, FeO-poor, ne-

pheline-normative melts, and their uniformly high CaO contents relative to mantle pyroxenites, partial melting of lithospheric cumulate pyroxenites rather than of mantle-derived pyroxenites seems to us to be a more plausible explanation for the origin of the distinctive CaO-rich melts considered in this study. The heat source for melting of these lower crustal lithologies could either be trend A arc magmas as they pass through the lower crust or hot ($\geq 1320^{\circ}\text{C}$ [see Sisson and Bronto, 1998]), adiabatically upwelled mantle that both produces water-poor arc magmas and heats the overlying lower crust [Sisson and Bronto, 1998].

5. Conclusions

[31] 1. Study of major and trace element compositions of olivine-hosted melt inclusions in a high-MgO calc-alkaline basalt from Batan Island (northernmost Philippines, Luzon-Taiwan arc) and of whole rocks (i.e., lavas) from Batan has led to the identification of two populations of magma: (1) A low-CaO (<13 wt %) group defined by melt inclusions in Fo₈₅₋₇₅ olivines and by Batan whole rocks is referred to as trend A. The compositional trend of these inclusions and whole rocks from Batan is similar to other arc-related lavas, and it is consistent with progressive extraction of olivine + clinopyroxene + amphibole from a parental basaltic liquid with 10 wt % MgO and 45 wt % SiO₂. (2) A CaO-rich (>13 wt %), nepheline-normative group of melt inclusions in Fo₉₀₋₈₅ olivines is defined as trend B. The compositional trend of these inclusions is interpreted in terms of mixing between the most CaO-rich, nepheline-normative inclusions (i.e., in Fo₉₀ olivine) and melts near the most primitive end of trend A defined by more typical island arc magmas.

[32] 2. Primitive island arc magmas characterized by very high CaO contents (up to 19.0 wt %) are present as olivine-hosted melt inclusions and whole rock samples from ten arc volcanoes

from all over the world, including Batan (Luzon-Taiwan arc), Galunggung (Sunda arc), Stromboli and Vulcano (Aeolian arc), Lihir (New Guinea), Nicaragua (Central America), Grenada (Lesser Antilles), Lombok (Sunda arc), Rindjani (Sunda arc), and Epi and Merelava (Vanuatu arc). These CaO-rich magmas define a group that is compositionally distinctive when compared to normal island arc lavas: Not only are they CaO-rich, but they are also nepheline normative and FeO-poor relative to previously recognized primitive arc magmas, and their normative nepheline contents are positively correlated with their CaO content.

[33] 3. The CaO contents of experimental partial melts of fertile or depleted peridotites under dry or hydrous conditions range up to 13–14 wt %: i.e., they are lower than the CaO contents of the CaO-rich, silica-undersaturated arc melts identified in this study. Although melting of peridotite at pressures of a few tens of kilobars in the presence of CO₂ can produce CaO-rich, silica-poor liquids, we consider it unlikely that this is responsible for producing CaO-rich arc-derived melts identified here based on their strong compositional contrasts with the compositions of primitive nephelinites and melilitites and with the compositions of experimental melts of carbonated peridotites.

[34] 4. Thermodynamic calculations of major element compositions (using the MELTS algorithm) and trace element modeling suggest that the most CaO-rich, FeO-poor, nepheline-normative melt inclusions and whole rocks identified in this study could represent intermediate to high degree (~10–40 wt %) partial melts of pyroxenites at lower crustal to upper mantle pressures (up to a few tens of kilobars). However, MELTS calculations predict that temperatures required to generate CaO-rich melts from anhydrous pyroxenite would be $\geq 1400^{\circ}\text{C}$, which is high relative to what is required for typical arc magmas. Also, the

remarkably high CaO contents of the most CaO-rich inclusions and whole rocks imply a source composition characterized by CaO concentrations at the high end of the range of natural pyroxenites.

[35] 5. Plausible sources of the CaO-rich, silica-undersaturated melts are lower crustal and shallow upper mantle pyroxene-rich cumulates from arc environments because such cumulates have CaO concentrations at the upper end of the range observed in mantle pyroxenites. Moreover, these cumulates often contain amphibole, which would lower their solidus temperatures to values more consistent with those expected in deep crustal or shallow subarc environments.

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