JOURNAL OF PETROLOGY | VOLUME 42 | NUMBER 9 | PAGES 1773–1779 | 2001

Comment on 'New Constraints on the *P–T* Evolution of the Alpe Arami Garnet Peridotite Body (Central Alps, Switzerland)' by Paquin & Altherr (2001)

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RECEIVED FEBRUARY 26, 2001; REVISED TYPESCRIPT ACCEPTED MAY 18, 2001

The 'Alpe Arami controversy' seems destined not to to end as more and more new analytical data become available. Recently, Paquin & Altherr (2001; hereafter P&A) presented an excellent set of major- and trace-element zoning profiles in garnets and in various generations of pyroxenes from the Alpe Arami peridotite body. The results led the authors to conclude that the peridotite experienced peak metamorphic conditions of 1180°C and 5·9 GPa. These values are in fairly good agreement with the high *P–T* estimates of Brenker & Brey (1997), but are at variance with the more recent estimates of Nimis & Trommsdorff (2001; hereafter N&T) of 840°C and 3·2 GPa. Although the thermobarometric estimates of P&A are much lower than postulated by Dobrzhinetskaya *et al.* (1996) and Green *et al.* (1997), P&A still invoke an ultra-high-pressure Alpine (35–43 Ma) metamorphism for Alpe Arami which does not fit the prograde, high-pressure Alpine metamorphic sequence of the Adula–Cima Lunga nappe, to which other ultramafic, mafic, ophicarbonate and pelitic rocks of this unit comply (see N&T and references therein). Overall, P&A's arguments in favour of high-*T* conditions are well grounded. However, we will show that (1) N&T's and P&A's temperature estimates are not in conflict with each other and reflect two distinct stages of equilibration characterized by different thermal conditions; (2) the low-*T* stage corresponds to N&T's estimates of 840°C and 3·2 GPa and is representative of the *P–T* recorded by the Alpe Arami peridotite *during Alpine subduction metamorphism* dated at 43 Ma (Gebauer *et al.*, 1992; Becker, 1993; Gebauer, 1996); (3) a short-lived thermal event at $T \geq 1100$°C, probably related to magmatic intrusion at 35 Ma (see Gebauer, 1996), caused complete re-equilibration of garnet Fe–Mg and Ni, but left pyroxene Ca, Cr, Al core contents virtually unaltered; (4) no unequivocal barometric estimate can be given for this high-*T* stage on the basis of available data, although pressures not very dissimilar from those achieved at the peak of subduction metamorphism are the most likely.

PAQUIN AND ALTHERR'S RESULTS

P&A carefully selected the compositions of the cores of the large, first-generation porphyroclasts (Ib according to P&A's classification) to estimate peak metamorphic conditions. P&A's arguments for an ultra-high-pressure metamorphism are mainly based on the high temperatures (~1100–1200°C) estimated studying Fe–Mg exchange equilibria between garnet (Grt), olivine (Ol) and pyroxenes (O'Neill & Wood, 1979; Harley, 1984; Krogh Ravna, 2000), Co and Ni partitioning between clinopyroxene (Cpx) and orthopyroxene (Opx) (Seitz *et al.*, 1999), and Ni partitioning between Ol and Grt (Canil, 1999). When coupled with the Opx–Grt barometer of

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Brey & Köhler (1990), these temperatures yield a pressure of ∼6 GPa. Temperatures based on pyroxene 'solvus' relations (Brey & Köhler, 1990) and on partitioning of other trace elements between Cpx Ib and Opx Ib yield much lower temperatures (∼700–900°C). P&A conclude that whereas Grt achieved equilibrium with both Cpx and Opx for Fe, Mg, Ni, Co, Ca and Al, the two pyroxenes Ib, which are never found in contact with each other, attained equilibration only for relatively fast-diffusing elements, such as Co and Ni, but did not equilibrate in terms of Ca, V, Ti, Sc and Cr partitioning. Similar arguments are used to establish a rapid formation of pyroxene neoblasts (II) at high *T*, shortly after peak metamorphic conditions.

COMMENTS

Ca partitioning between Opx and Cpx

P&A highlight the existence of plateaux in the zoning profiles of Fe, Mg, Ni, Co and Al in the cores of pyroxene porphyroclasts and suggest that these reflect equilibration of Cpx and Opx with Ol and Grt at some *P* and *T*. They also compare the Ca contents of pyroxene Ib porphyroclasts and show that, despite their fairly flat profiles, they do not match the Ca_{Opx}/Ca_{Cpx} systematics determined experimentally by Brey *et al.* (1990) on lherzolitic compositions at various *P* and *T*. They conclude that Cpx and Opx porphyroclast cores are not in equilibrium in terms of Ca partitioning, thus precluding the use of pyroxene solvus relations as a thermometer. However, their analysis is too simplified, as it does not take into account the role of other elements, which may have a strong influence on the partitioning of Ca. In particular, Na competes with Ca in Cpx M2 sites and thus has a major effect on the solubility of Ca. The ratio $Ca/(1 - Na)$ is therefore a much more robust indicator of temperature than is Ca alone (Bertrand & Mercier, 1985; Brey & Köhler, 1990). This issue is of crucial importance for Alpe Arami Cpx cores, which are characterized by high Na contents (0·134–0·151 a.p.f.u.) compared with Brey *et al.*'s experimental Cpx (<0·09 a.p.f.u.). Figure 1 shows that, when corrected for the effect of Na, the partitioning of Ca between Alpe Arami pyroxenes is consistent with the experiments and suggestive of $T \leq 900$°C. It should be noted, however, that the $Ca/(1 - Na)$ ratio is also slightly composition dependent and its use as a thermobarometer is not straightforward.

A more robust approach, already tackled by N&T, would have been to compare the temperatures yielded by the pyroxene solvus thermometer of Brey & Köhler (1990) or Taylor (1998), with those yielded by the Ca-in-Opx thermometer of Brey & Köhler (1990). The two thermometers provide us with two independent evaluations of the closure temperature of the Ca–Mg exchange, which rules pyroxene solvus relations; the former essentially depends on the composition of the Cpx, the latter on that of the Opx. The temperatures given by the two thermometers diverge by <40°C at $P = 3{\cdot}2$ GPa (Fig. 2). There is no indication of disequilibrium and therefore no reason to dismiss the low Cpx–Opx temperatures estimated by N&T and confirmed by a large number of samples, including those studied by P&A, Ernst (1978), Evans & Trommsdorff (1978) and Brenker & Brey (1997). The discrepancy between T_{solvus} and $T_{Grt-Opx,Cpx,Ol}$ requires a different explanation.

Al in Opx and Cr in Cpx

P&A rightly note that 'Al contents in the cores of pyroxene Ib porphyroclasts are nearly constant and similar to those of pyroxene Ia inclusions in garnet', suggesting 'equilibration with garnet at some *P* and *T*'. Therefore they deem the Opx–Grt Al-exchange barometer suitable for an evaluation of *P*. However, P&A do not take into due consideration the fact that also Cr exhibits flat profiles in both Cpx Ib porphyroclasts and Cpx Ia inclusions in garnet and that the contents of this element are virtually identical in the two textural types (see their fig. 5). Given the very low diffusivity of Cr, the flat profiles also suggest complete equilibration of Cpx with Grt, the other main Cr reservoir, at some *P* and *T*. The Cr-in-Cpx barometer of Nimis & Taylor (2000), which is based on Cr partitioning between Cpx and Grt, is therefore also suitable for an assessment of *P*. Both the Opx–Grt and the Cr-in-Cpx barometers are virtually insensitive to the composition of the garnet. In particular, the Cr-in-Cpx barometer has been widely tested on mantle xenoliths and diamond inclusions against the graphite–diamond curve (Nimis & Taylor, 2000). The Taylor (1998) version of the Opx–Grt barometer is preferred here to the more popular Brey & Köhler (1990) one because it yields estimates almost identical to those yielded by the Cr-in-Cpx barometer when applied to well-equilibrated peridotites (Nimis & Taylor, 2000). The Opx–Grt and Cr-in-Cpx barometers show a very good agreement when applied to P&A pyroxenes assuming a nominal temperature equal to T_{solvus}, but they diverge substantially at higher temperatures (Fig. 2). The use of the Brey & Köhler (1990) version of the Opx–Grt barometer would even enhance this divergence.

Al zoning in pyroxenes

P&A interpret the approximately M-shaped Al zoning profile in Opx Ib porphyroclasts as a result of decompression followed by cooling. They also assert that

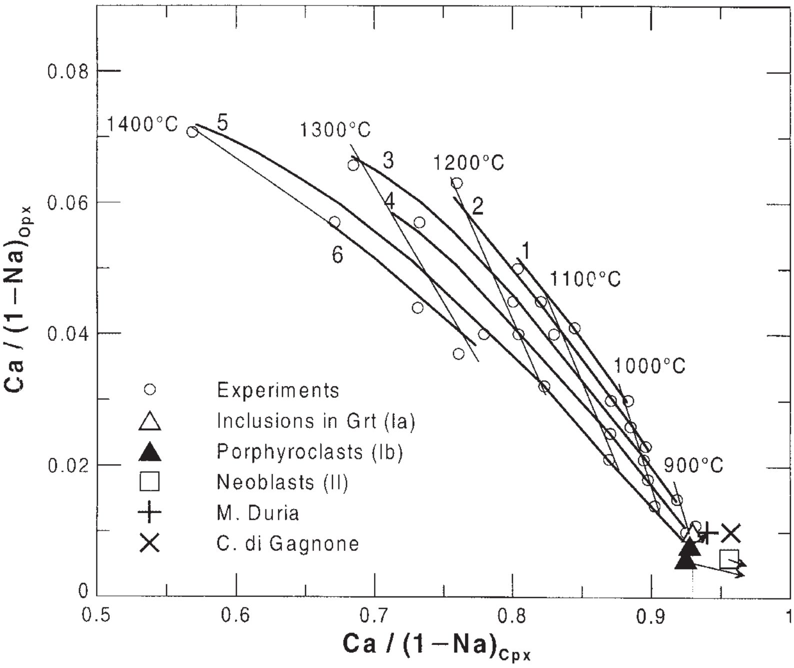

Fig. 1. Ca partitioning corrected for Na between clinopyroxenes and orthopyroxenes in garnet peridotites from Alpe Arami (Ia, Ib and II; Paquin & Altherr, 2001) and other Central Alps occurrences (Nimis & Trommsdorff, 2001), and in experiments on lherzolite compositions (Brey *et al.*, 1990). Core–rim trends are shown where significant as arrows. Thick and thin lines are respectively approximate isobars (GPa) and isotherms based on the experiments. It should be noted that, because of compositional factors, the use of Ca/(1 − Na) as a thermobarometer is not straightforward.

Al zonation in Cpx Ib porphyroclasts is characterized by 'continuously decreasing Al concentrations from core to rim, suggesting somewhat different *P*–*T* evolution' and therefore supporting their 'interpretation that both pyroxenes Ib were not in chemical equilibrium with respect to Ca and Al'. However, Na in Cpx Ib similarly shows a rimward decrease (see P&A, fig. 5), suggesting that Al content in Cpx has at least in part been controlled by jadeite-consuming reactions, rather than exclusively by Cpx–Grt equilibria. Similar trends are common in partially retrogressed garnet peridotites (e.g. Evans & Trommsdorff, 1978; Medaris, 1984; Nimis & Morten, 2000) and can be explained in terms of net transfer reactions of the type jadeite + 2diopside + (3/2)enstatite + H_2O → edenite, related to kelyphite and amphibole neoblast formation. Therefore, differences in the Al profiles in Cpx and Opx may simply reflect the higher sensitivity of Al_{Opx} to *P*–*T* variations during the garnet-stage and the higher sensitivity of $(Na,Al)_{Cpx}$ to retrogressive, post-garnet reactions. Relatively flat Na plateaux are anyway still preserved in the cores of the Cpx Ia and Ib, suggesting maintenance of the original equilibrium compositions. It is worth noting that for Cpx Ia inclusions in garnets, which were better preserved from hydration reactions, no kelyphite is typically observed at the Grt–Cpx Ia interface and they show an M-shaped profile that mimics that of Opx. The close similarity between the two profiles becomes apparent when figs 3 and 5 of P&A are reset to the same vertical scale. The apparently more complex zoning profiles in Cpx II neoblasts simply reflect the greater sensitivity of smaller Cpx grains to amphibole-producing reactions. [It is worth noting that jadeite loss has little effect on *P* calculations using the Cr-in-Cpx barometer of Nimis & Taylor (2000) and on *T* calculations using pyroxene solvus thermometers. Therefore their application to Cpx II neoblasts remains warranted.] There is therefore little reason to assign Cpx and Opx different *P*–*T* histories based on Al relations. It is also worth noting here that the initial rimward increase of Al in Opx (and in Cpx Ia) can also be interpreted as a result of isobaric heating, rather than decompression. The relative implications will be discussed below.

The high-*T* stage

The strongest argument put forward by P&A to support their high-*T* scenario is the finding of Mg- and Ni-rich cores in the garnets (see also Brenker & Brey, 1997). Assuming these cores are in equilibrium with olivine and pyroxenes, their composition implies high temperatures (~1100–1200°C; Fig. 2) and the observed rimward decrease of Mg and Ni indicates rapid cooling (see Paquin

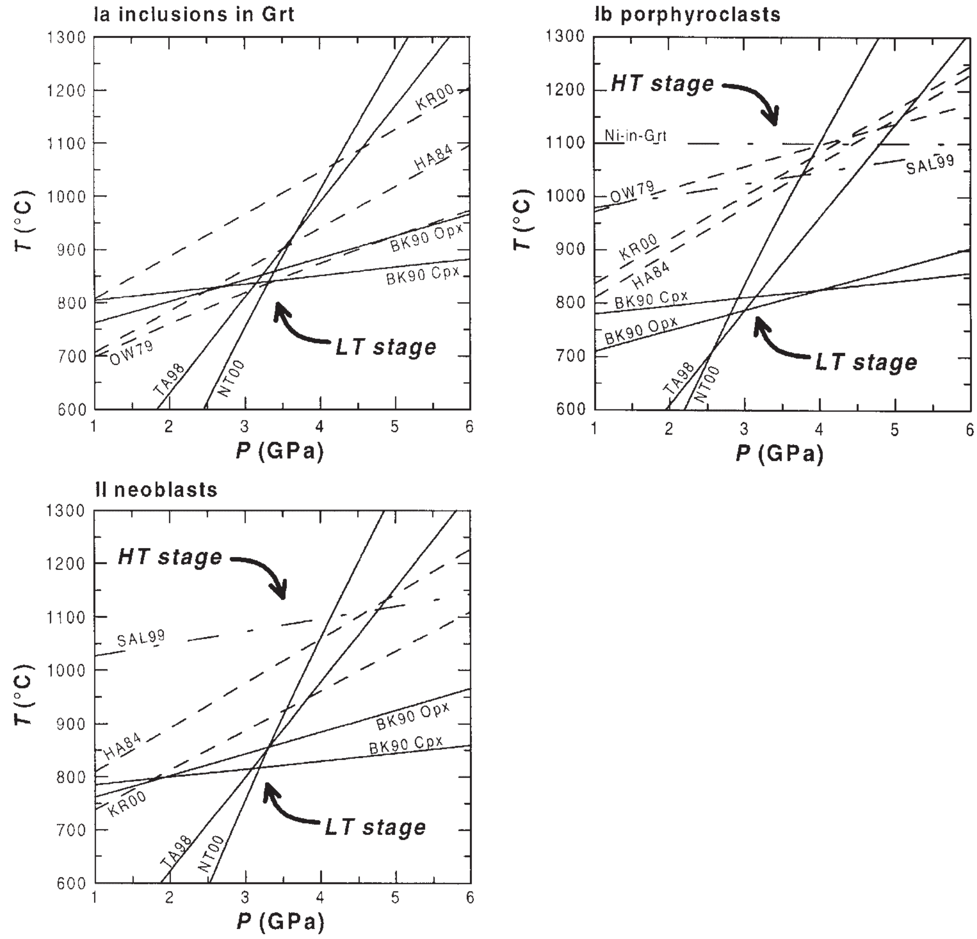

Fig. 2. Thermobarometry of P&A samples. P and T are calculated using mineral core compositions extracted from P&A's zoning profiles. Continuous lines refer to slow-diffusing element equilibria (Ca–Mg, Cr, Al); dashed lines refer to fast-diffusing element equilibria (Fe–Mg, Ni, Co). The low-T stage is ascribed to high-P metamorphism during Alpine subduction at 43 Ma. The high-T stage is ascribed to a short-lived event related to magmatic intrusion at 35 Ma. Accurate barometry of the high-T stage is not practicable because the thermometers intersect at low angle and because of probable partial re-equilibration under cooling, but the most consistent temperatures are obtained for pressures not very dissimilar from those attained during the low-T stage. Uncertainties for thermometers and barometers are probably within 50–70°C and 0·25 GPa, respectively. NT00, Nimis & Taylor (2000; Cr-in-Cpx); TA98, Taylor (1998; Opx–Grt; Al); BK90 Cpx, Brey & Köhler (1990; Cpx–Opx; Ca–Mg); BK90 Opx, Brey & Köhler (1990; Ca-in-Opx); HA84, Harley (1984; Opx–Grt; Fe–Mg); KR00, Krogh Ravna (2000; Cpx–Grt; Fe–Mg); SAL99, Seitz *et al.* (1999; Cpx–Opx; Co); Ni-in-Grt, Canil (1999; Ol–Grt; Ni); OW79, O'Neill & Wood (1979; Ol–Grt; Fe–Mg). The original HA84 thermometer has been empirically corrected to make up for its inconsistency at very low and very high T (see Brey & Köhler, 1990; Nimis & Trommsdorff, 2001).

& Altherr, 2000). Using the expression $h = \sqrt{(D.t)}$, based on Fick's second law, and assuming a diffusion coefficient (D) of $\sim 10^{-14}$ cm^2/s for the Fe–Mg couple in Grt at 1150°C and 3 GPa (Chakraborty & Ganguly, 1991), the estimated time required to completely equilibrate a 3·5 mm garnet porphyroblast would be <0·1 m.y. The diffusivity of Ni in Grt is comparable with that of the Fe–Mg couple (Griffin *et al.*, 1996). The diffusivities of the Ca–Mg couple in Cpx and of Al in both pyroxenes are two to three orders of magnitude lower and those of Cr can be even less (Sautter *et al.*, 1988; Smith & Barron, 1991; Dimanov & Sautter, 2000). A relatively short-lived thermal pulse could thus cause complete re-equilibration of the garnets in terms of Fe–Mg and Ni exchanges, although leaving the composition of the pyroxene cores virtually unaffected. Therefore, the high $T_{\text{Grt–Opx,Cpx,Ol}}$ calculated by P&A do not necessarily contradict the low-T estimates indicated by pyroxene solvus relations. Rather, the two temperatures may reflect distinct stages in the metamorphic history of the Alpe Arami peridotite.

Opx–Grt $T_{\text{Fe–Mg}}$ for Opx Ia inclusions in garnets are slightly lower than those calculated for the Opx Ib porphyroclasts (Fig. 2). This difference may reflect the greater susceptibility of small Opx inclusions to partial re-equilibration under cooling after the above thermal pulse. The apparent lack of Fe–Mg re-equilibration in

Cpx Ia inclusions after the thermal pulse is consistent with the much lower Fe–Mg diffusivity in Cpx relative to that in Opx and Grt (Dimanov & Sautter, 2000). The relatively low Cpx–Grt T_{Fe-Mg} obtained for Cpx II neoblasts probably reflects incomplete Grt–Cpx II equilibration during the high-T stage.

Because the solubilities of Al in Opx and Cpx coexisting with Grt, and of Ca in Opx coexisting with Cpx, increase significantly with rise in temperature, the short-lived thermal event is also the most probable source of the approximately M-shaped zoning profiles of Al in both Opx and Cpx Ia and of Ca in Opx (see P&A, figs. 3 and 5). Although less distinct, Cpx consistently exhibits complementary W-shaped Ca profiles. Furthermore, P&A admit they have no convincing explanation for the peculiar W-shaped Cr profiles in Grt (see P&A, fig. 2). These can be related to reactions of the type (2/3)uvarovite + (1/3)knorringite + (2/3)grossular + (1/3)pyrope → 2 $CaCrAlSiO_6$ + 2 diopside (see Nimis & Taylor, 2000), which reflect either decompression or a rise in T, and are therefore also consistent with a thermal pulse followed by cooling. Significantly, a complementary profile is exhibited by Cpx Ia inclusions, which show a tenuous, yet appreciable, Cr maximum ~100 mm from the grain edges.

Incidentally, we recall that P&A reiterate the use of Ca/Cr relations in garnet made by Brenker & Brey (1997) to constrain P–T estimates. This issue had already been discussed extensively by N&T, who demonstrated the inconsistency of the thermobarometric formulations of Brenker & Brey on the basis of experimental data on lherzolitic compositions. This point needs no further discussion.

Opx–Cpx trace element equilibria

The Opx–Cpx thermometer of Seitz *et al.* (1999) based on Co partitioning between Opx and Cpx yields high temperatures (1000–1100°C) when applied to P&A's pyroxenes. These temperatures are similar to those yielded by Fe–Mg and Ni exchange thermometry and are in line with the relatively high expected diffusivity of Co (see P&A). As for other trace elements, the Seitz *et al.* Sc, V and Cr thermometers yield very low temperatures (<750°C at 3·2 GPa). We agree with P&A that some degree of disequilibrium may have occurred between the pyroxenes at peak metamorphic conditions for these slow-diffusing elements. We just add that uncertainties in the determinations of trace element abundances, especially in Opx, may also have constituted a significant source of error. Finally, the finding of Li enrichments in Alpe Arami Cpx (Paquin *et al.*, 2000) cannot be invoked as a proof for general disequilibrium between the pyroxenes, as Li-enriched Cpx in cryptically metasomatized peridotites may well maintain equilibrium major-element compositions (Seitz & Woodland, 2000).

CONCLUSIONS

Compositional data and zoning patterns in pyroxenes offer no valid reason to consider early-generation Cpx and Opx cores out of equilibrium, at least in terms of Ca–Mg and Al partitioning. Then the low temperatures estimated on the basis of pyroxene solvus relations pose a challenging problem to the high temperatures calculated from Fe–Mg and Ni thermometry of Grt–Ol and Grt–pyroxene pairs. We are thus left with three possibilities: (1) all Grt-based temperatures are wrong; (2) pyroxene solvus-based temperatures are wrong; (3) Grt-based and pyroxene solvus-based temperatures reflect compositional relations that were produced at different stages and under different P–T conditions.

The first hypothesis was strongly favoured by us in N&T. In that paper we suggested that garnet core composition could in part have been controlled by local equilibria with pre-existing spinel grains, hence precluding the use of garnet cores to calculate P–T conditions. The high sensitivity of some thermometers to errors in the determination of equilibrium compositions, including those derived from analytical uncertainties, and propagation of errors on P estimates, would have produced the anomalously high T estimates. None the less, the remarkable agreement between the Ol–Grt, Opx–Grt and Cpx–Grt T_{Ni} and T_{Fe-Mg} estimated by P&A and, particularly, the shapes of Mg, Fe and Ca zoning profiles in garnets, which do not support the hypothesis of growth zoning on spinel sites, weaken the arguments originally put forward by N&T.

The second hypothesis was contended by P&A, but their arguments have been shown here to be inconclusive and, in part, incorrect. In any case, even rejecting solvus-based temperatures, the poor accord between the two independent Opx–Grt and Cpx–Grt barometers at high T renders P&A's barometric data for peak metamorphic conditions unreliable and suggests that they can have been significantly overestimated (Fig. 2). Accurate barometry of the high-T stage is not anyway practicable, both because the thermometers involved intersect at a low angle and because of probable partial garnet and pyroxene Fe–Mg, Ni and Co re-equilibration upon cooling. If P&A's pressure estimates were too high, their Grt–(Ol, Cpx, Opx) T_{Mg-Fe} calculated at $P = 6$ GPa may also be excessive. Using the nominally P-independent Ni-in-Grt thermometer, minimum temperatures of ~1100°C can be estimated for the high-T stage.

We believe the third hypothesis to be the most rational, as it reconciles both the high- and the low-temperature estimates obtained for different chemical equilibria. Following the same line of reasoning as used by P&A, the origin of the discrepancy in thermometric data must lie in the different diffusivities of the various cations exchanged. As Ca–Mg, Cr and Al in pyroxenes diffuse much more *slowly* than Fe–Mg and Ni in garnets, the low-T conditions recorded by pyroxene Ca–Mg equilibria must *pre-date* the thermal event responsible for the high Fe–Mg- and Ni-exchange temperatures. For the same reason, the barometric data extracted from pyroxene–garnet Cr and Al partitioning data should not be attributed to the high-T stage, but to the earlier low-T stage. This is corroborated by the good agreement between pressure estimates obtained using the Cpx–Grt and Opx–Grt barometers at T given by pyroxene solvus thermometry for all pyroxene generations (Ia, Ib, II). The corresponding P–T conditions of $\sim$800°C and $\sim$3 GPa (Fig. 2) fit the Alpine, prograde metamorphic sequence of the Adula–Cima Lunga unit and can therefore be assumed as representative of the peak metamorphic conditions reached by the Alpe Arami peridotite *during Alpine subduction* dated at 43 Ma by Gebauer (1996) on metamorphic zircons in Grt-pyroxenites, eclogites and Grt-peridotites. These moderate P–T conditions are also in line with the fact that unique early features are shared by the Alpe Arami and Cima di Gagnone garnet peridotites, the latter pointing to an upper pressure limit around 3 GPa and temperatures around 750°C (Nimis & Trommsdorff, 2001). These features are ilmenite rods and palisades (Risold *et al.*, 2001) and identical, metamorphic crystal preferred orientations with [100] perpendicular to foliation in first-generation olivine at both localities (Frese *et al.*, 2001). The short-lived thermal event at $T \geq$ 1100°C can be related to magmatic intrusions, which have been dated at 35 Ma by Gebauer (1996) on magmatic zircons in Grt-pyroxenites, eclogites and Grt-peridotites. [Gebauer (1996) proposed an origin of the magmatic zircons from melts produced by decompression-driven partial melting of the Alpe Arami peridotite. An exotic origin of the melts seems, however, more consistent with the thermobarometric data.] Accurate barometry of this high-T phase is not anyway practicable on the basis of available data, because of lack of high-T pyroxene–garnet equilibrium pairs. Most likely, pressure conditions were not very dissimilar from those recorded at the peak of the subduction metamorphism (Fig. 2). As a working hypothesis, we suggest a causal correlation between the heating event at 35 Ma and the magma-driven Li metasomatism described by Paquin *et al.* (2000).

ACKNOWLEDGEMENTS

We are grateful to Jens Paquin for continuing and stimulating discussion and for supplying a preprint version of the P&A paper. Paolo Nimis gratefully acknowledges financial support of MURST ex 60%, 'Progetti di Ricerca Giovani Ricercatori' (Padova) and C.N.R. 'Centro di Studio per la Geodinamica Alpina' (Padova).

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