

# Discovery of Paleozoic Fe-Mg carpholite in Motalafjella, Svalbard Caledonides: A milestone for subduction-zone gradients

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## ABSTRACT

**Paleozoic blueschist facies rocks are relatively scarce on Earth due to warmer geothermal gradients at that time and/or later reequilibration. Ferro-magnesiocarpholite (Fe-Mg carpholite), the typical low-temperature blueschist facies index mineral in metapelites, was discovered 30 yr ago and is known only in Tethyan belts metamorphosed <80 m.y. ago. Herein we report the discovery of Paleozoic Fe-Mg carpholite in the ca. 470 Ma blueschists of Motalafjella, Svalbard Caledonides, the oldest known occurrence on Earth. The carpholite-bearing rocks reached pressure-temperature (*P-T*) conditions of 15–16 kbar and 380–400 °C and followed a nearly isothermal exhumation path. In the cooling Earth perspective, these *P-T* estimates for Motalafjella blueschists demonstrate the existence of cold subduction-zone gradients (~7 °C/km) from the middle Paleozoic onward.**

**Keywords:** high-pressure–low-temperature metamorphism, blueschists, caledonides, carpholite, Svalbard.

## INTRODUCTION

Blueschist outcrops can be considered as relicts of former subduction zones and thus mark old convergent margins. Remnants of Paleozoic blueschist facies rocks are notoriously rare compared to Mesozoic–Cenozoic blueschists (de Roever, 1956; Ernst, 1972), and only three Precambrian localities are known (Liou et al., 1990). The scarcity of older blueschists may be ascribed to a progressive cooling of the Earth, with higher geothermal gradients during Proterozoic time (22 and 15 °C/km at the end of the Middle Proterozoic and Late Proterozoic, respectively; Grambling, 1981; Maruyama and Liou, 1998), or to a mere preservation problem due to subsequent recrystallization and overprint of high-pressure–low-temperature (HP-LT) metamorphism.

Ferro-magnesiocarpholite (Fe-Mg carpholite) is a hydrated inosilicate with the formula (Fe, Mg)Si<sub>2</sub>Al<sub>2</sub>O<sub>6</sub>(OH)<sub>4</sub> (same as lawsonite substituting Fe-Mg for Ca), stable at temperature, *T* < 400–450 °C, and pressure, *P* > 7–8 kbar (Chopin and Schreyer, 1983; Vidal et al., 1992) (Table 1). Fe-Mg carpholite is a typical index mineral in low-temperature blueschist facies metapelites, and its importance has been increasingly recognized in recent petrogenetic grids (e.g., Holland and Powell, 1998; Wei and Powell, 2004). Its stability field is even more diagnostic of HP-LT conditions than the lawsonite–glaucofan stability field for metabasites, which extends toward lower pressure conditions.

Fe-Mg carpholite has until now been found

exclusively in Tethyan metapelites and only reported for rocks metamorphosed after 80 Ma (Table 1), supporting the view that formation of the mineral has been restricted to cold Mesozoic–Cenozoic subduction zones (B. Goffé, 2000, personal commun.). In this contribution we report the existence of Fe-Mg carpholite in Ordovician blueschists from Motalafjella, Svalbard Caledonides (e.g., Ohta et al., 1986), which is the oldest occurrence on Earth, and discuss the implications for subduction-zone gradients.

## GEOLOGIC SETTING

Motalafjella blueschists belong to the Vestgötabreen Metamorphic Complex of the west-

central basement of Spitsbergen (Fig. 1) and are the only blueschists known in Svalbard. These HP-LT rocks were metamorphosed during the Caledonian orogeny, and later refolded during the Tertiary orogeny (e.g., Horsfield, 1972), when Svalbard separated from Greenland (Harland, 1965). The Vestgötabreen Metamorphic Complex consists of two structural units separated by a refolded thrust contact (Ohta et al., 1986). The structurally highest unit consists of blueschist and eclogite facies metabasite lenses in a garnet-chloritoid-epidote-phengite matrix schist. Hirajima et al. (1988) estimated the *P-T* conditions as 580–640 °C and 18–24 kbar. The Fe-Mg-carpholite-bearing lowermost unit consists of phyllites and calc-schists with subordinate amounts of serpentinite, metabasalt, and metacarbonate boudins (Fig. 1). The lower unit resembles a strongly deformed metasedimentary sequence (as for the Alpine Schistes Lustrés) rather than a tectonic mélange with blocks or knockers (such as the Franciscan terranes). The presence of lawsonite, pumpellyite, and sodic amphibole points to HP-LT metamorphic conditions for the lower unit (Hirajima et al., 1984). The lack of major internal contact suggests that *P-T* conditions for the whole lower unit were broadly comparable.

The upper and lower units are unconform-

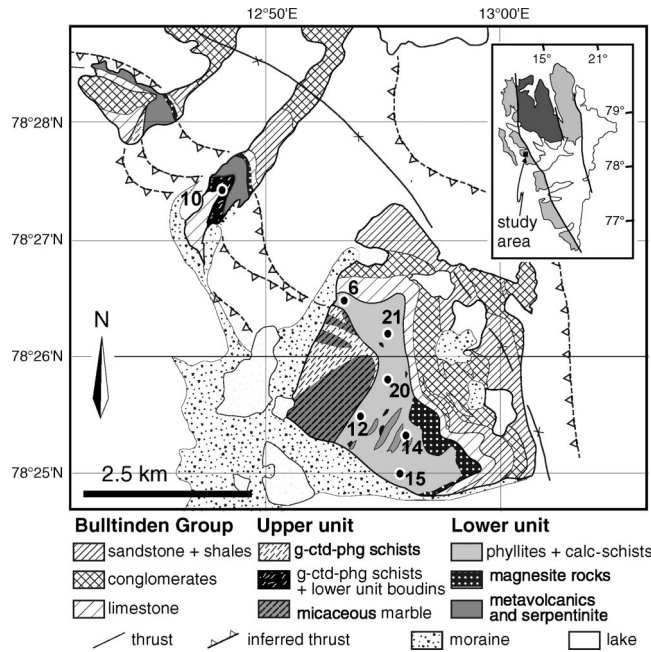
TABLE 1. FERRO-MAGNESIOCARPHOLITE (FMCAR) LOCALITIES WORLDWIDE

| Locality                | X <sub>Mg</sub><br>FMcar | KFMASH<br>Min. (+Phg) | <i>P-T</i> conditions<br>(kbar/°C) | Pr.<br>age | M. age<br>(Ma) |
|-------------------------|--------------------------|-----------------------|------------------------------------|------------|----------------|
| <b>Neotethyan realm</b> |                          |                       |                                    |            |                |
| W. Alps (Vanoise)       | 0.5–0.7                  | Chl-Prl-Ctd           | 1–12/300–380                       | PT         | 45–35          |
| W. Alps (S. Lustrés)    | 0.3–0.5                  | Chl-Ctd               | 12–15/350–400                      | K          | 55–45          |
| W. Alps (Liguria)       | 0.2–0.7                  | Chl-Prl-Ctd           | 7–10/300–400                       | J<         | –              |
| W. Alps (Corsica)       | 0.2                      | Chl                   | 11/400                             | K          | 55             |
| Swiss Alps              | 0.5–0.7                  | Chl                   | 11–14/350–380                      | K          | 60–40          |
| Crete–Peloponnese       | 0.3–0.8                  | Chl-Ctd-Sud           | 8–17/350–450                       | CT         | ca 20          |
| Calabria (Italy)        | 0.5–0.7                  | Chl-Ctd               | 10–12/340–380                      | K?         | ca 35          |
| Tuscany (Italy)         | 0.3–0.7                  | Chl-Prl-Ctd           | 8/350                              | T=         | 30–25          |
| Betics (Spain)          | 0.3–0.9                  | Chl-Ctd-Prl-Ky        | 8–12/300–480                       | PT         | 40–25          |
| Rif (Morocco)           | 0.5–0.9                  | Chl-Sud-Ctd           | c. 14/400                          | P          | 40–25          |
| Oman                    | 0.7–0.7                  | Chl-Ctd               | 8–9/350                            | P          | 80             |
| Turkey                  | 0.7–0.9                  | Chl-Ctd-Ky            | 10–14/440–480                      | T>         | E?             |
| Indonesia               | 0.2–0.4                  | Chl                   | c. 15/350?                         | J?         | KE             |
| New Caledonia           | 0.3–0.9                  | Chl-Prl               | 7/300                              | KE         | 50–30          |
| <b>Other</b>            |                          |                       |                                    |            |                |
| Spitsbergen             | 0.6–0.7                  | Chl-Sud-Ctd           | 15–16/380–400                      | Cm?        | >460           |

Note: FMcar compositions (X<sub>Mg</sub> taken as Mg/[Fe+Mg]), critical mineral associations, *P-T* conditions, and age of protolith (Pr) and metamorphism (M) are indicated (references can be obtained on request). C—Carboniferous; Cm—Cambrian; E—Eocene; J—Jurassic; K—Cretaceous; KFMASH—K<sub>2</sub>O-FeO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-H<sub>2</sub>O; P—Permian; T—Trias; W. Alps—Western Alps (< and = stand for Lower and Middle, respectively).

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**Figure 1.** Simplified geological map of Motalafjella terrane (after Ohta et al., 1986) outlining structural stack made of (from top to bottom) upper metamorphic unit, lower metamorphic unit, and unconformable series (Bulltinden Group overturned). Numbers refer to samples studied in text (Table 2). Inset: Location of study area in Spitsbergen, showing basement rocks (light gray) and large Devonian basin (dark gray). g-ctd-phg—garnet-chloritoid-phengite.



ably overlain by fossiliferous Upper Ordovician to Lower Silurian flysch, shale, and conglomerate deposits of the Bulltinden group (Ohta et al., 1986) (Fig. 1), thus giving minimum age constraints for the blueschists. Consistently, postmetamorphic cooling ages on phengite from the upper unit are 470–460 Ma with Rb/Sr and K/Ar methods (Dallmeyer et al., 1989). According to the latest palinspastic reconstructions (Torsvik et al., 2001) that place Svalbard north of Greenland until Permian time, Motalafjella blueschists formed dur-

ing the early convergence stages between the Spitsbergen-Greenland margin and Baltica (Harland, 1965).

### MINERAL DESCRIPTION AND P-T PATH

#### Ferro-magnesiocarpholite Occurrences

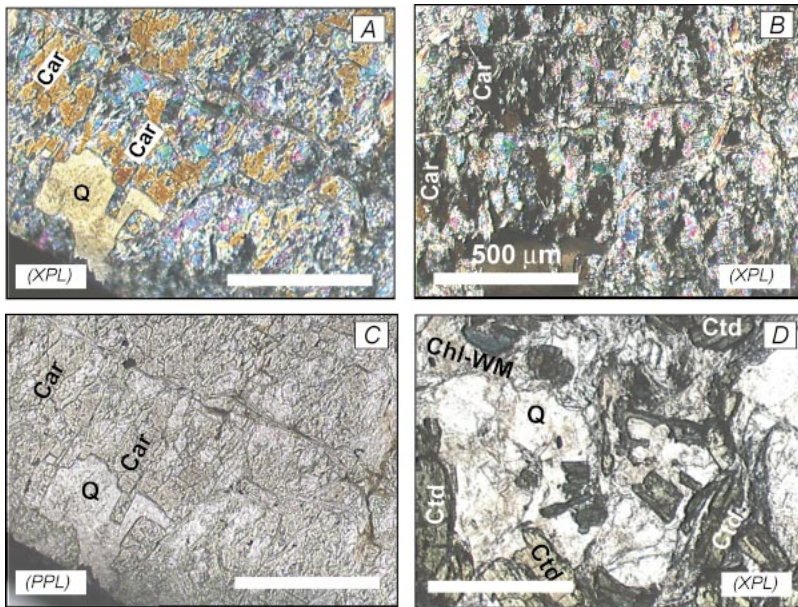
Our sampling of Motalafjella's lower unit focused on aluminous metapelites and calc-schists (Fig. 1). Relicts of Fe-Mg carpholite were found in sample 21Ba (Fig. 2) and pseudomorphs were found in sample 14. Fe-Mg

carpholite relicts appear as a nonpleochroic, second-order birefringent, straight extinction relict phase. Chloritoid (Ctd) appears either as postkinematic porphyroblasts, sometimes pseudomorphed by aggregated chlorite (Chl samples 20c and 21Ba), or as oriented crystals with sheared twin lamellae (samples 10 and 20b). Paragonite (Pg) is a common accessory mineral together with apatite. Sudoite (Sud), the typical low-*P* ditrioctahedral chlorite in carpholite (Car) bearing rocks (e.g., Vidal et al., 1992), occurs as late disoriented crystals in two samples (Table 2). Lawsonite was found in most calc-schists.

These observations suggest that, after crossing the divariant reaction  $\text{Car} = \text{Ctd} + \text{quartz} (\text{Q}) + \text{H}_2\text{O}$ , the samples reequilibrated with the Ctd-Chl phengite (Phg)  $\pm$  Pg paragenesis and were later transformed partly to Chl-Phg or Sud-Chl-Phg assemblages. These parageneses are comparable with those found in Tethyan Cenozoic blueschists (Table 1).

### Mineral Compositions

Representative electron probe analyses of Fe-Mg carpholite (Cameca SX 50; 15 kV, 10 nA beam conditions; WDS mode) point to a Mg-rich composition with  $X_{\text{Mg}} = 0.66$  (Table 3). Maximum  $X_{\text{Mg}}$  values for chloritoid reach 0.3 (Table 2). Phengite mostly plots along the  $(\text{Fe}, \text{Mg})^{\text{IV}}\text{Si}^{\text{IV}}\text{-Al}^{\text{VI}}\text{Al}^{\text{IV}}$  tschermak substitution line (celadonite-muscovite joint) (Fig. 3A), but also displays a minor yet significant pyrophyllite content. The celadonite content is minimum for the latest phengite generations. Chlorite shows strong variations not only in the paragonite stability field. Maximum *T* conditions estimated from TWEEQU calculations agree well with the Raman spectroscopy  $T_{\text{max}}$  estimate as well as with temperatures derived from the Chl-Ctd geothermometer (Fig. 3B). Furthermore, calculated maximum temperatures are close to the location of the divariant reaction  $\text{Car} = \text{Ctd} + \text{Q} + \text{H}_2\text{O}$  for  $X_{\text{Mg}} = 0.7$  (Fig. 3B), which is consistent with the preservation of Mg-rich carpholite as a relict phase in sample 21Ba. Such temperatures agree with those of comparable Fe-Mg carpholite compositions elsewhere (Table 1) and the retrograde *P-T* conditions are consistent with the occurrence of sudoite ( $P < 7$  kbar,  $T < 450$  °C; Vidal et al., 1992). The *P-T* estimates determined with TWEEQU point to an isothermal (or slightly cooling) decompression *P-T* path, a result in line with the occurrence of small and constant interlayer deficiencies in phengite (Agard et al., 2001a), and with the good preservation of lawsonite in both metabasites and calc-schists. The absence of heating on decompression advocates for synorogenic exhumation processes (e.g., Ernst, 1988; Trotet et al., 2001).



**Figure 2.** Sample 21Ba. A, B, and C: Photomicrographs showing Fe-Mg carpholite (Car) relicts. Note Fe-Mg Car's straight extinction in B and two cleavages at right angle in C. D: Characteristic aspect of chloritoid crystals (Ctd) in more recrystallized domains of thin section. PPL—plane polarized light; XPL—cross-polar light; Q—quartz; Chl-WM—chlorite-white mica.

TABLE 2. PARAGENESES OF THE STUDIED SAMPLES

| Sample | Phg                    | Chl                   | Car  | Ctd  | Other min.           |
|--------|------------------------|-----------------------|------|------|----------------------|
| 10     | 3.55/0.55<br>3.33/0.56 | 2.9/0.25<br>2.55/0.25 |      | 0.26 | Pg-Q-Cc-Rt-Mt-Ap-T   |
| 12     | 3.43/0.57<br>3.32/0.4  | 2.85/0.4<br>2.75/0.4  |      | r?   | Pg-Ab-Q-Rt-Mt-Ap-T   |
| 14     | 3.39/0.55<br>3.12/0.48 | 2.7/0.2               | p    | 0.08 | Q-Rt-Mt-Ap-T         |
| 15     | 3.09/0.25              | 2.7/0.44              |      |      | Q-Mt-Ap              |
| 20b    | 3.47/0.7<br>3.2/0.5    | 2.85/0.4<br>2.75/0.4  |      | r    | Q-Cc-Rt-Ap           |
| 20c    | 3.39/0.35              | 2.7/0.35              |      | 0.26 | Pg-Q-Cc-Rt-Mt-Ap     |
| 21Ba   | 3.22/0.5<br>3.1/0.3    | 0.77                  | 0.68 | 0.27 | Pg-Sud-Q-Cc-Rt-Mt-Ap |
| 21Bc   | 3.15/0.45              | 0.68                  |      | 0.22 | Sud-Q-Rt-Mt-Ap       |

Note: The characteristic compositions of the main minerals are indicated. Mineral abbreviations: Ab—albite; Ap—apatite; Car—(Fe, Mg) carpholite; Cc—calcite; Chl—chlorite; Ctd—chloritoid; Mt—magnetite; Phg—phengite; Pg—paragonite; Q—quartz; Rt—rutile; Sue—susoite; T—tourmaline. Others: p—pseudomorph; r—replaced by aggregated chlorite. Values given for Phg and Chl:  $Si^{4+}/X_{Mg} = Mg/(Mg+Fe)$ . Only  $X_{Mg}$  is given for Car and Ctd.

**Relative Scarcity of Fe-Mg Carpholite**

Fe and Fe-Mg carpholite varieties were only discovered 50 and 30 yr ago, respectively (de Roever, 1951; Goffé et al., 1973), despite abundant occurrences in, for example, the Alps and Turkey. The question arises as to why this important index mineral is relatively scarce despite its simple structural formula (Table 1). For a long time, Fe-Mg carpholite was probably confused with other mineral species such as tremolite. Its scarcity, compared to glaucophane, which is the equivalent index mineral in mafic rocks, is partly accounted for by its much narrower stability field with regard to both pressure and temperature (e.g., Evans, 1990; Wei and Powell, 2004). Bulk chemistry probably also plays a key role, since Fe-Mg carpholite has not been found in metasediments similar to circum-Pacific metagraywackes (type B of Maruyama et al., 1996). The high Ca content of metagraywackes is clearly detrimental to the formation of carpholite because most of the protolith's aluminum will be tschermak substitution, but also in di-trioctahedral substitution toward sudoite (mixed analyses between chlorite and sudoite are shown in Fig. 3A).

TABLE 3. MICROPROBE ANALYSES OF FE-MG CARPHOLITE

| Sample                         | 21Ba  | 21Ba  | 21Ba  | 21Ba  | 21Ba  |
|--------------------------------|-------|-------|-------|-------|-------|
| Analysis                       | ad7   | ad8   | af45  | af46  | af49  |
| SiO <sub>2</sub>               | 38.65 | 37.04 | 39.96 | 38.55 | 37.41 |
| TiO <sub>2</sub>               | 0.04  | 0.35  | 0.10  | 0.12  | 0.13  |
| Al <sub>2</sub> O <sub>3</sub> | 32.72 | 31.63 | 31.96 | 32.10 | 31.57 |
| FeO                            | 7.23  | 7.50  | 6.68  | 7.43  | 8.42  |
| MnO                            | 0.64  | 0.56  | 0.55  | 0.61  | 0.47  |
| MgO                            | 8.74  | 7.79  | 9.55  | 8.94  | 9.74  |
| Σ (wt%)                        | 88.04 | 84.95 | 88.80 | 87.79 | 87.76 |

| Structural formula (calculated with 8 oxygens) |      |      |      |      |      |
|--|------|------|------|------|------|
| Si   | 2.00 | 1.99 | 2.04 | 2.00 | 1.96 |
| Ti   | 0.00 | 0.01 | 0.00 | 0.00 | 0.01 |
| Al   | 1.99 | 2.00 | 1.92 | 1.96 | 1.95 |
| Fe <sub>tot</sub>                              | 0.31 | 0.34 | 0.28 | 0.32 | 0.37 |
| Mn   | 0.03 | 0.03 | 0.02 | 0.03 | 0.02 |
| Mg   | 0.67 | 0.62 | 0.73 | 0.69 | 0.76 |
| X <sub>Mg</sub>                                | 0.66 | 0.63 | 0.70 | 0.66 | 0.66 |

Note: Σ (wt%): sum in weight percent (other elements <0.05 wt%).

**P-T Calculations**

P-T estimates were obtained on the basis of textural equilibria using the TWEEQU program (Berman, 1991) and following Vidal and Parra (2000). Berman's June 92 thermodynamic database was complemented by thermodynamic data for chlorite and white mica end members (Parra et al., 2002; Vidal et al., 2001, 2005). One T<sub>max</sub> estimate was also obtained from the Raman spectrum of carbon-rich sample 6, using the geothermometer of Beyssac et al. (2002). This thermometer is based on the progressive yet irreversible increase of the organization of the carbonaceous matter toward graphite with temperature. THERMOCALC software (Holland and Powell, 1990, 1998) and the Chl-Ctd geothermometer from Vidal et al. (1999) were also used.

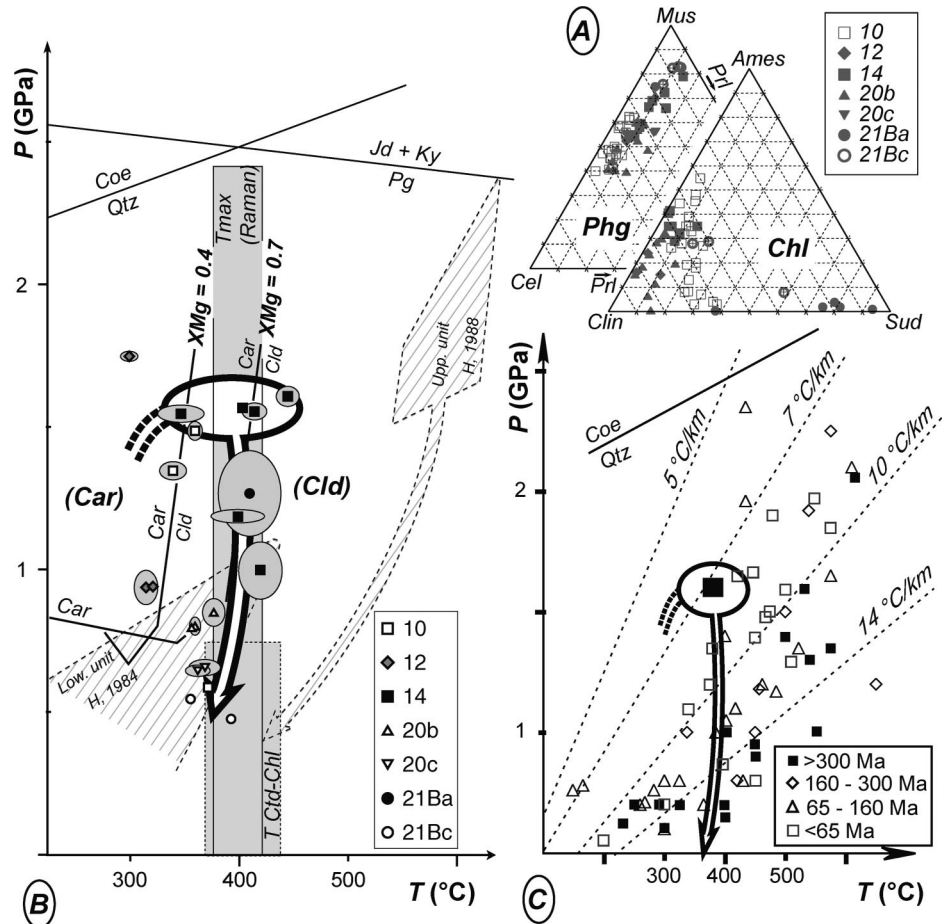


Figure 3. A: Range of chemical variations within phengite (Phg) and chlorite (Chl) in studied samples (see Fig. 1). Ames—amesite; Cel—celadonite; Clin—clinocllore; Mus—muscovite; Prl—pyrophyllite; Sud—sudoite. B: Pressure-temperature (P-T) estimates for Motalafjella samples as determined from TWEEQU software, Raman spectroscopy, and Ctd-Chl geothermometry. Ellipses correspond to P-T uncertainties. Higher P-T conditions are obtained than those previously inferred for lower unit (H, 1984, H, 1988—Hirajima et al., 1984, 1988). Steep black divariant lines separate carpholite (Car; to the left) and chloritoid (Cld; to right) stability fields; X<sub>Mg</sub> values given here refer to theoretical Mg/(Mg+Fe) ratio of Fe-Mg carpholite (after Vidal et al., 1992). C: Comparison of Motalafjella blueschists P-T estimates with those of other Phanerozoic blueschists worldwide grouped by age (data after Maruyama et al., 1996; Okay, 2002; Agard et al., 2001b). Note high P-T regime of Motalafjella blueschists compared to other old (>300 Ma) Paleozoic blueschists. Despite cooling trend with time observed throughout Phanerozoic, our results show that cold (~7 °C/km) subduction-zone gradients existed 470 m.y. ago.

The *P-T* results (Fig. 3B) point to much higher *P* low-grade blueschist facies conditions than those inferred by Hirajima et al. (1984) for the lower unit. Maximum burial conditions cluster around 15–16 kbar for *T* ~380–400 °C. Partial *P-T* paths can be obtained for samples 10, 12, and 14. In contrast, samples 20b, 20c, and 21Bc yield equilibrium conditions only for the retrograde (greenschist facies) part of the *P-T* path. Considering that the whole lower unit probably evolved as a single body, a composite *P-T* path can be established using these data. Raman spectroscopy of sample 6 gave a  $T_{\max}$  of  $378 \pm 6$  °C (eight spectra). THERMOCALC results yielded slightly higher *P-T* conditions for the Car-Ctd-bearing sample (21Ba) at  $19.5 \pm 2$  kbar and  $457 \pm 23$  °C.

## DISCUSSION

### P-T Constraints for the Lower Unit

These *P-T* estimates of 380–400 °C and 15–16 kbar are within or at slightly higher *T* than the carpholite stability field (Fig. 3B) (Vidal et al., 1992), and well within that taken up by lawsonite during the early prograde evolution at lower *P* conditions. The Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> ratio, which is much higher for meta-graywackes than for Alpine (or Spitsbergen) metapelites, may also be significant.

### Milestone for Subduction-Zone Gradients

The deduced thermal gradients for the lower unit, ~6–8 °C/km, are somewhat colder than the 13 °C/km inferred by Hirajima et al. (1988) for the upper unit. Both gradients are within the low *P-T* range given by Chopin and Schreyer (1983) for carpholite-chloritoid occurrences and show a value similar to estimates for cold Cenozoic subduction zones (e.g., 8 °C/km for the Western Alps; Agard et al., 2001b; 5–6 °C/km for Turkey; Okay, 2002).

Motalafjella blueschists represent the coldest *P-T* gradient estimated for Paleozoic blueschists (Fig. 3C) and the oldest cold subduction-zone geotherm documented so far (6–8 °C/km with respect to the 11–14 °C/km of Aksu, China; Liou et al., 1990, 1996). In the cooling Earth perspective (e.g., de Roever, 1956; Ernst, 1972; Grambling, 1981), a crucial change in thermal regimes was proposed to have taken place between 750 and 540 Ma by Maruyama and Liou (1998) (Fig. 3C), based on ultrahigh-*P* occurrences worldwide. Quantitative constraints for blueschists were nevertheless lacking. The results presented here demonstrate that cold, analogous to present-day, subduction gradients were well established 470 m.y. ago.

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