ARE STABLE ISOTOPIC TRENDS IN AMPHIBOLITE TO GRANULITE FACIES TRANSITIONS METAMORPHIC OR DIAGENETIC?—AN ANSWER FOR THE ARENDAL AREA (BAMBLE SECTOR, SOUTHEASTERN NORWAY) FROM MID-PROTEROZOIC CARBON-BEARING ROCKS

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ABSTRACT. The isotopic \( \delta^{13}C \) and \( \delta^{18}O \) compositions of primary (marbles) and secondary (veins) calcites and primary (fahlbands and metapelites) and secondary (veins) graphites sampled across the amphibolite to granulite facies transition in the Bamble Sector (southeastern Norway), have been investigated. The primary calcites display a regional trend in a \( \delta^{13}C/\delta^{18}O \)-plot which is the reverse of the expected trend. The granulate grade primary calcites essentially preserved most of their initial sedimentary compositions, whereas the amphibolite grade calcites have increasingly lower \( \delta^{13}C \) values. The \( \delta^{18}O \) compositions of amphibolite grade primary calcites and of some vein types appear to be buffered by the O-reservoir of the country rock. The \( \delta^{13}C \) of graphite of different types is specific for a certain lithology but not spatially related to the metamorphic zoning.

Reaction progress calculations and Rayleigh fractionation modeling prove that prograde metamorphic decarbonation reactions alone cannot be held responsible for the isotopic trend in the marbles. The reversed isotopic trend in the marbles themselves combined with the lack of any trend in the primary graphites is not compatible with pervasive flushing with a homogeneous, mantle derived \( \text{CO}_2 \)-rich fluid at granulate facies metamorphism. The marble trend and the constant \( \delta^{13}C \) composition of primary graphite throughout the metamorphic zoning are best explained by low grade processes that did affect the stable isotopic compositions of the marbles but left the graphites unaffected. The processes may even occur under diagenetic conditions. Later amphibolite-granulite facies metamorphism preserved the trend despite the production of calcisilicates. Subsequent retrograde hydrothermal action produced graphite-quartz veins and several types of calcite veins, without resetting stable isotope systematics on a regional scale.

INTRODUCTION

Especially under granulate facies conditions, the isotopic and chemical composition of the sparse initial fluid is buffered and controlled by the excess amount of rock and mineral reactions taking place (Sheppard,
Fluids are commonly present in sedimentary rocks during early metamorphism. They may either represent formation fluids captured during deposition, fluids generated internally and controlled by proceeding devolatilization reactions, hydrothermally introduced fluids, or any combination of these (Rice and Ferry, 1982; Rumble, 1982). Synmetamorphically crystallizing magmas contribute not only to their own fluid budget but also to that of the country rock in their immediate vicinity (Touret, 1971b, 1985; Frost, Frost, and Touret, 1989), whereas anatectic magmas may negatively influence the fluid budget, that is, extract fluid from the surrounding rocks (Fyfe, 1973).

The role of fluids in the transition from upper amphibolite to granulite facies metamorphic conditions is a crucial but still unsolved question, as well as their composition and way of transport. Fluid inclusions may contain direct information on fluid composition (Touret and Hartel, 1990), although post-peak metamorphic recrystallization of quartz during uplift and cooling may affect original densities and composition by several independent processes (Hollister, 1990; Bakker, 1992; Bakker and Jansen, 1990, 1991). Coexisting minerals may yield indirect information on the composition of the fluid present during metamorphism. Stable isotope systematics of carbonaceous rocks have been demonstrated to act as sensitive monitors of metamorphic fluids under amphibolite facies conditions (Rye and others, 1976). In some cases, however, carbonaceous rocks, which enjoyed granulite facies metamorphism, preserved their pre-metamorphic isotope characteristics, for example in Rogaland, southwestern Norway (Bol and others, 1990), the Adirondacks, northeastern United States of America (Valley and O'Neil, 1984), and the Madukkarai area in Tamil Nadu, India (Srikantappa and Valley, 1992).

**GEOLOGICAL SETTING**

The Bamble Sector of the Baltic Shield (fig. 1) is a classic amphibolite to granulite facies transition zone (Bugge, 1940, 1943). High grade metamorphism occurred during both the Gothian (Kongsbergian, 1.7-1.5 Ga) and the Sveconorwegian (Grenvillian, 1.25-0.9 Ga) orogenies and resulted in a metamorphic gradient ranging from upper amphibolite facies to LILE-depleted granulite facies (Touret, 1971a; Field, Drury, and Cooper, 1980; Smalley and others, 1983). The transition zone is featured by a well developed isograd sequence (from northeast to southwest): muscovite-out in quartzites, cordierite-in in metapelites, orthopyroxene-in in amphibolites, orthopyroxene-in in acidic gneisses, allanite- and titanite-out in gneisses, metapelites, and amphibolites (Nijland and Maijer, 1993). These isograds are accompanied by a change in the pleochroic colors of hornblende, increased recrystallization of sillimanite, and cross cut by a possible kyanite-out isograd in metapelites and cordierite-orthoamphibole rocks (Nijland and Maijer, 1993). Peak metamorphic conditions range from $752 \pm 34^\circ C$, $7.1 \pm 0.4$ kb in the amphibolite facies area to about $836 \pm 49^\circ C$, $7.7 \pm 0.3$ kb in the core of the
Fig. 1. Map of the Arendal area, Bamble Sector, Norway, with sample locations and the zones of Smalley and others (1983). Dot = type 1-2 calcite, circle = type 3-7 calcite; upright crosses = type 1-2 graphite, oblique crosses = type 3-4 graphite.

granulite facies area (Nijland and Majer, 1993). Metamorphism is characterized by a clockwise prograde P-T path (Visser and Senior, 1990), followed by near isobaric cooling (Touret and Olsen, 1985; Visser and Senior, 1990; Nijland, Jånsen, and Majer, 1993). The metamorphic
complex is made up of intrusive rocks and a supracrustal suite composed of subordinate marbles, calcisilicate rocks, and skarns among graphiticgneisses, quartzites, quartz-rich banded gneisses, amphibolites, andminor lenses of cordierite-orthoamphibole rocks. For a more elaborate
description of the lithologies and structures, the reader is referred to

Investigated lithologies.—The skarns in the vicinity of Arendal (fig. 1)
are associated with extensive magnetite mineralizations mined from
medieval times until the 1960’s. The skarns occur intercalated with
marbles and charno-enderbite gneisses in a narrow discontinuous band
following the strike in the coastal strip between Arendal and Grimstad
(Bugge, 1940, 1943; Starmer, 1985b). Mineralogy of these skarns has
been described by Barth (1925) and Bugge (1951, 1954). A second band
of marbles and calcisilicate rocks is located between Ubergsmoen and the
Nidelva Quartzite Complex (Nijland and others, 1993).

Earlier work on stable isotope systematics in the Bamble Sector
includes δ13C and δ18O studies on CO2 of fluid inclusions, quartz, and
graphite (Andreae, 1974; Hoefs, and Touret, 1975; Pineau and others,
carbonates from the area. In this study, concordant metasedimentary
marbles and discordant carbonate veins as well as some quartz segrega-
tions and graphites from gneisses have been analyzed for their carbon
and oxygen stable isotopic composition. The purpose was (1) to establish
the presence and extent of control of any metamorphic fluids, and, if
present, (2A) to understand the effect on amphibolite-granulite transi-
tion, or, if not present, (2B) to explain the origin of the observed stable
isotope systematics.

PETROGRAPHY

Carbonate samples.—The samples may be divided into primary
(marbles and skarns) and secondary (vein) carbonates. Carbonate veins
range in width from microscopic size up to 30 cm and usually cut across
the layering. Seven different types of calcites in the carbonaceous rocks
can be distinguished on the basis of mineralogy, distribution of silicate
minerals, grain size, shape, and orientation of the rock body. They are
summarized in table 1, and their corresponding mineralogical composi-
tions in table 2. All thin sections have been stained with alizarin-red in
order to check for the presence of dolomite.

Types 1 and 2 contain primary calcite, originally deposited during
the early Gothian and recrystallized during subsequent Gothian and
Sveconorwegian metamorphism. At Lofstad (loc. D1), marbles are inter-
calated with enderbitic gneisses and amphibolites. Samples MB 034 and
MB 034a were taken respectively from the rim and core of the same
marble body. The zone C exposure at Barbudalen (loc. C1) consists of
skarns, marbles, and calcisilicates and is locally very rich in magnetite. In
zone B, the exposure at Skoletjern (loc. B1) consists of gneissic metasedi-
ments with thin intercalated marble bands. At locality A1, small massivé
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Mineral abbreviations according to Kretz (1983), plus Dat = datolite, Sul = sulfides. Sizes in millimeters.

calcite bodies border the Blengsvatn gabbro, which intruded the Nidelva Quartzite Complex. Fragments of gabbro float in the carbonate; no reaction rims are present. The marble at locality A4 (sample TN 35) occurs in a sequence of sillimanite-bearing metapelites, quartzites, amphibolites, and calcisilicate gneisses, which grade into the Selås banded gneisses (Tourret, 1966). Sample CM 687 (loc. A2) was taken from the metasedimentary sequence hosting the old Ettedalen Pb-Zn-Ag ore deposit (Naik, 1975; Krijgsman, ms). This occurrence is near the Porsgrunn-Kristiansand Fault which separates the Bamble Sector and the Telemark Basement Gneiss Complex.

The total carbonate-content of the outcrops at localities D1, C1, and A1 amounts to about 10 percent. The outcrop at the B1 locality contains less than 1 percent total carbonate. The fresh roadcuts all display tight to isoclinal folds and boudinage, implying intense deformation. Marbles are half to 2 m thick and concordant to the layering of the surrounding rocks. Major constituents of the type 1 and type 2 carbonates (marbles) are calcite, dolomite, diopsidic to hedenbergitic clinopyroxene, eastonite-rich phlogopite, olivine, scapolite, spinel, titanite, vesuvianite (MB 021) with accessory amphibole, magnetite, and rutile. Clinopyroxene is occasionally altered to amphibole, and amphibole and mica partly to chlorite and pumpellyite. In contrast to this, the olivine is extremely fresh. Dolomite is present in samples from locality D1 and in marble from locality A4 but never occurs adjacent to or in the vicinity of quartz. Marbles from locality D1 contain up to four different types of dolomite exsolutions. Silicate minerals are usually concentrated in bands that may represent original sedimentary layering.

Type 3 calcite originates from veins directly related to the primary carbonates. These veins were sampled from all localities described above. The veins consist of calcite, garnet, clinopyroxene, scapolite, and quartz. Accessories are apatite, sulfides, titanite, and tourmaline. An apatite vein, rich in Ti-minerals and with minor calcite (sample MB RØN), occurs at locality A1.
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</tbody>
</table>

Mineral modi are calculated after pointcounting 1000 points per sample and are accurate to 0.1 percent. All other modi were estimated from hand-specimen and are approximations. All abbreviations used for minerals are according to Kretz (1983). Further abbreviations mean: No = sample number, Tp = type of calcite (compare table 1), Zn = metamorphic zone, Loc = location in figure 1, x = not measured, Amph = clinopyroxene, Opq = opaque minerals, s = serpentinized olivine, bir = birefringent.
Type 4 veins are directly related to the primary carbonates, but their mineral assemblage (axinite, datolite, et cetera) indicates retrograde forming conditions. These veins were sampled only at locality D1 but have also been described from locality C1 (Bugge, 1940). They typically consist of calcite, clinzoisite, epidote, low-albite, prehnite, axinite, datolite, and zeolites (Bugge, 1954).

Type 5 veins occur in amphibolites and small discordant metagabbro bodies which intruded during the early stages of the Sveconorwegian Orogeny (De Haas, Verschure, and Maijer, 1993). The veins were sampled from an amphibolite near Flaten (loc. A5), from the Arendal Gabbro (loc. C3; De Haas, Verschure, and Maijer, 1993), and from the Vestre Dale Gabbro (loc. A8; De Haas and others, 1992). Type 5a veins cut across the metagabbro/amphibolite and are composed of idiomorphic scapolite, biotite, pyrrhotite, ilmenite, and interstitial calcite. Type 5b veins consist of plain calcite. Sample MB 208 contains two different calcites: The older calcite MB 208o occurs as a lining directly along the fracture walls and forms large idiomorphic grains up to 3 cm across. The younger calcite MB 208y forms a 1 cm thick finer grained (<3 mm) encrustation on the older calcite.

Type 6 and 7 veins postdate ductile deformation in the area and were formed under lowest grade metamorphic conditions, which were either late Sveconorwegian or younger. Both types occur throughout the entire area and are not restricted or directly related to some specific lithology. Type 7 veins have the same mineral assemblage as small cavities in late basic dikes which are generally considered to be related to the Oslo Rift.

Retrogradation occurs in all exposures. Thin sections reveal millimeter to centimeter-sized alteration trails. Amphibole, calcite, chlorite, clinzoisite, epidote, as well as kaolinitization of K-feldspar and scapolitization of plagioclase are commonly observed, together with accessory grains of opaque minerals and tourmaline.

**Graphite samples.**—Graphite has been sampled from various lithologies throughout the whole area. They may be divided in four types (table 3). Primary graphite constitutes the occasionally microfolded foliation of the rocks together with biotite and occurs in two types. Type I occurs in so-called fahlbands (Gammon, 1966), which are sulfide-rich, quartzitic banded gneisses. Type II occurs in garnet and biotite-bearing metapelitic

<table>
<thead>
<tr>
<th>Type</th>
<th>Rocktype</th>
<th>Size</th>
<th>Texture</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
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<td>I</td>
<td>fahlband</td>
<td>fine</td>
<td>parallel with micas</td>
<td>primary</td>
</tr>
<tr>
<td>II</td>
<td>metapelite</td>
<td>fine</td>
<td>parallel with micas</td>
<td>primary, new overgrowths</td>
</tr>
<tr>
<td>III</td>
<td>vein</td>
<td>coarse</td>
<td>random in feldspars</td>
<td>segregations in fahlbands</td>
</tr>
<tr>
<td>IV</td>
<td>vein</td>
<td>coarse</td>
<td>random in quartz</td>
<td>hydrothermal</td>
</tr>
</tbody>
</table>
gneisses, usually with sillimanite and accessory sulfides. This graphite may have thin rims of syntactic newly grown graphite flakes (compare Kwiecińska, 1980; Duke and Rumble, 1986). Secondary graphites (types III and IV) occur in veins and segregations: Type III graphite occurs in veins that form irregular feldspar-rich segregations with accessory garnet and sulfides in the fahler bands (type I); type IV occurs in late hydrothermal quartz lenses or in thin veinlets with some accessory clinopyroxene (sample MB 192).

Quartz samples.—All samples except one were collected at Tromøy from orthopyroxene-bearing neosomes in enderbitic gneisses by Van den Kerkhof, Touret, and Kreulen (1994), who studied the fluid inclusions. One sample (FK 31d) was collected from a discordant quartz vein at Tromøy.

Analytical methods

Carbonate samples.—Marbles and carbonate veins were microsampled by taking 4 mm diameter cores with a watercooled diamond drill. Veins not in exact alignment with the drill could be sampled only partly with this technique. Some of the interstitial carbonate, carbonate from larger veins, and carbonate with intergrown sulfide-mineral grains were prepared for analysis by chiselling manually. Sulfide mineral-bearing samples were purified by handpicking in order to avoid any possible effect of these minerals on the CO₂-extraction. Carbonate was milled to about 0.1 mm grainsize in a tungsten carbide ball-mill. Before the next sample, the mill was cleaned by milling with quartz, washing with acetone, and airdrying.

The CO₂-gas was extracted in a number of batches of about 10 samples each, including a number of duplicates and an internal standard, following the method of McCrea (1950). Attack of the dolomite present in some samples was avoided by careful control of reaction time and temperature (Sharma and Clayton, 1965; Al-Aasm, Taylor, and South, 1990). Gas pressures resulting from the reaction were measured directly on the vacuum-line and recalculated to weight percentages of calcite.

A 100 percent calcitic marble from Naxos, Greece with a δ¹³C of 2.07 ± 0.05 permil and a δ¹⁸O of 23.67 ± 0.10 permil was used as an internal standard for the carbonate stable isotope extractions. The average δ¹³C and δ¹⁸O composition of this standard as measured with the analytical procedure in this study is 2.06 ± 0.04 permil and 23.67 ± 0.08 permil (n = 13), respectively.

Graphite samples.—Selected graphite-rich samples were prepared for measurement by pulverization in an agate vibratory disc-mill and subsequently washed with hydrochloric acid to remove possible traces of carbonate. Whenever possible, graphite was handpicked from the hand-specimen and separately prepared for measurement. Graphite was combusted with tank-oxygen to CO₂ in the presence of CuO at 900°C according to the method of Craig (1953), with some minor adaptations. The total graphite content of the samples was recalculated from the
resulting CO₂ pressure. A graphite-quartzite from Naxos with a δ¹³C of 26.76 permil and a C-content of 0.52 percent was used as an internal standard with the graphite combustions.

Both calcite and graphite isotopic ratios have been measured with a VG ISOGAS SIRA 24 mass spectrometer at the Department of Geochemistry, Utrecht University. ¹³C is normalized to the Cretaceous Pee Dee Belemnite Standard (PDB), and ¹⁸O to Standard Mean Ocean Water (SMOW). Values are presented in permil relative to these standards. Reproducibility of the measurements was better than ± 0.1 permil for both δ¹³C and δ¹⁸O from calcite, better than ± 0.2 permil for δ¹³C from graphite, and within 3 and 0.03 wt percent for calcite and graphite, respectively.

Quartz samples.—Quartz δ¹⁸O measurements were performed at Kline Geology Laboratory, Yale University. Oxygen was extracted from the quartz by reaction with BrF₅ and subsequently converted to CO₂ by reaction with a hot graphite rod in presence of Pt (Clayton and Mayeda, 1963). More detailed information about analytical procedures can be found in Schiffries and Rye (1989). The isotopic composition of the CO₂
Table 4

B: Stable isotope $\delta^{13}$C (PDB) and $\delta^{18}$O (SMOW) compositions in permil of secondary type 3-7 calcites from the area around Arenal

<table>
<thead>
<tr>
<th>Loc</th>
<th>No</th>
<th>Tp</th>
<th>Cr</th>
<th>%Cal</th>
<th>$\delta^{13}$C</th>
<th>$\delta^{18}$O</th>
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<tbody>
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<td>052</td>
<td>4</td>
<td>I</td>
<td>6.4</td>
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<td>13.2</td>
</tr>
<tr>
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<td>052</td>
<td>4</td>
<td>II</td>
<td>4.7</td>
<td>-4.3</td>
<td>12.4</td>
</tr>
<tr>
<td></td>
<td>052</td>
<td>4</td>
<td>III</td>
<td>3.9</td>
<td>-5.8</td>
<td>12.9</td>
</tr>
<tr>
<td></td>
<td>052</td>
<td>4</td>
<td>IV</td>
<td>18.0</td>
<td>-4.8</td>
<td>12.9</td>
</tr>
<tr>
<td></td>
<td>066</td>
<td>3a</td>
<td>I</td>
<td>2.7</td>
<td>-2.7</td>
<td>15.6</td>
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<tr>
<td></td>
<td>066</td>
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<td>I</td>
<td>4.1</td>
<td>-1.3</td>
<td>14.1</td>
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<tr>
<td></td>
<td>058</td>
<td>3a</td>
<td>I</td>
<td>4.1</td>
<td>-2.7</td>
<td>14.2</td>
</tr>
<tr>
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<td>060</td>
<td>3a</td>
<td>I</td>
<td>30.2</td>
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<td>060</td>
<td>3a</td>
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<td>III</td>
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<td>C1</td>
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<td>3a</td>
<td>I</td>
<td>4.7</td>
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<td>3a</td>
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<td>6.6</td>
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<td>3a</td>
<td>I</td>
<td>2.6</td>
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<td>071</td>
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<tr>
<td></td>
<td>080</td>
<td>5a</td>
<td>x</td>
<td>-</td>
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Loc = location in figure 1, No = sample number, Cr = core number, Tp = carbonate type (compare table 1), %Cal = wt% calcite, x = sample purified by handpicking. For further explanation, see text.

gas was measured using a Finnigan MAT 251 EM mass spectrometer equipped with an automatic gas handling system. All analyses were checked against the NBS 28 standard and are reported relative to SMOW. The average of these measurements was 9.68 ± 0.08 permil (n = 4).

RESULTS

Primary calcite (types 1-2).—The results of the isotopic analyses are summarized in table 4A and graphically presented in figure 2A. The $\delta^{13}$C and $\delta^{18}$O isotopic compositions of the marbles range from −1.3 to −11.4 permil and from 18.2 to 10.3 permil, respectively. Marbles from the highest grade zone D are high in $\delta^{13}$C and $\delta^{18}$O. The core of a single marble body (MB 034a) is heavier in carbon than its rim (MB 034), while the amount of calcite minerals remains perfectly constant (compare table 2A). A thick marble is heavier in carbon than a thin one (compare samples MB 054, MB 055, and MB 057; table 4A). The $\delta^{13}$C of zone C marbles partly coincide with that of zone D, but $\delta^{18}$O is about 3 permil lower. Zone B marbles are even lower in $\delta^{13}$C but show $\delta^{18}$O values...
Fig 2(A) $\delta^{13}C$ versus $\delta^{18}O$ plot for primary carbonates (type 1-2), arranged to metamorphic grade. Triangles: zone D; squares: zone C; stars: zone B; circles: zone A. Shaded area indicates non-metamorphic Proterozoic carbonates after Schidlowski, Eichmann and Junge (1975).

(B) $\delta^{13}C$ versus $\delta^{18}O$ plot for vein carbonates (type 3-7), arranged to metamorphic grade. Triangles: zone D; squares: zone C; stars: zone B; circles: zone A.
(C) $\delta^{13}C$ versus $\delta^{18}O$ plot for vein carbonates (type 3-7), arranged to type of calcite after table 1.

comparable to zone C marbles. The marbles of amphibolite facies zone A are the lowest in $\delta^{13}C$ but are approx 1.5 permil higher in $\delta^{18}O$ than those from zones B and C. Three exceptions occur: (1) MB 182 is lower in $\delta^{18}O$ than the other marbles of location D1; (2) CM 687 is a few permil higher in $\delta^{13}C$ than other zone A marbles; and (3) TN 35 of locality A4 is higher in both $\delta^{13}C$ and $\delta^{18}O$ than all other type 1 calcites, even those of zone D. The marbles generally become isotopically lighter in carbon and, to a lesser degree, in oxygen with lower metamorphic grade.

Secondary calcite (types 3-7).—The results of the isotopic analyses of vein calcites are summarized in table 4B and graphically presented in figure 2B (ordered to metamorphic zoning) and 2C (ordered to type of calcite). Type 3a and type 4 veins of locality D1 display a larger scatter in $\delta^{13}C$ and $\delta^{18}O$ than the corresponding marbles and are located directly under/around those marbles (compare fig. 2A, B, and C). Type 4 veins of locality D1 are somewhat lighter in carbon than type 3a veins. Type 3a veins from locality C1 are confined to a small region in figure 2B and C directly under the corresponding marbles. Two type 3a veins (MB 082 and TG 63) have much heavier oxygen isotopes than all other 3a veins. However, the same holds true for their respective associated marbles. Veins of types 5a, b, 6, and 7 which do not have a close spatial relationship to marbles and/or calcisilicates scatter widely over the diagram. They do not seem to correlate with the metamorphic grade of the host rocks. Type
Table 5

<table>
<thead>
<tr>
<th>Loc</th>
<th>No</th>
<th>Tp</th>
<th>%Grp</th>
<th>δ^{13}C</th>
<th>Loc</th>
<th>No</th>
<th>Tp</th>
<th>%Grp</th>
<th>δ^{13}C</th>
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<td>AS 2292</td>
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<td>TN 571x</td>
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<td>Ai</td>
<td>AV 73</td>
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<td>Ab</td>
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<td>Ad</td>
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<td>9.75</td>
<td>-15.3</td>
</tr>
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<td>AV 113</td>
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<td>0.21</td>
<td>-21.4</td>
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</tr>
</tbody>
</table>

Loc = location in figure 1, No = sample number, Tp = graphite type (compare table 3), %Grp = wt% graphite, x = sample purified by handpicking. For further explanation, see text.

5b veins seem more or less restricted to the lower right quarter of figure 2B and C, except sample TN 441.

Graphite samples.—The results are summarized in table 5 and graphically presented in figure 3. Graphites from fahlsbands (type I) range in δ^{13}C isotopic composition between −21.4 and −28.3 permil. The isotopic range of graphites from metapelitic gneisses (type II) is smaller: from −20.2 to −23.9 permil. Graphite types III and IV from veins have the highest isotopic values. They range in isotopic composition from −15.3 to −20.2 permil.

Quartz samples.—The quartz segregations in enderbite gneisses have δ^{18}O values between 8.1 and 10.8 permil, while the discordant quartz vein (FK 31d) gives 7.8 permil.

**DISCUSSION**

Isotopic evolution of the marbles.—At metamorphic temperatures, decarbonation reactions (Shieh and Taylor, 1969; Ferry, 1991) tend to lower the initially high δ^{18}O and δ^{13}C ratios of originally sedimentary carbonates. Exchange reactions with infiltrating fluids (Sheppard, 1986) may either raise or lower the original isotopic composition of the carbonate sediments. Isotopic trends as displayed by the Bamble marbles (fig. 2) have been observed in many contact metamorphic aureoles (Lattanzi, Rye, and Rice, 1980; Nabelek and others, 1984; Bol and others, 1990). In such aureoles, isotopic values usually decrease with increasing metamorphic gradient (Valley, 1986; Ferry, 1991; Nabelek, 1991). In regional metamorphic terranes, marbles often show decreasing isotopic values toward their contacts with siliceous rocks, due to infiltration of carbon dioxide-bearing aqueous fluids (Rye and others, 1976; Baker and others, 1989) or the expulsion of reaction-generated CO₂-rich fluids (Valley, 1986; Ferry, 1991).
Fig. 3. $\delta^{13}C$ versus type of graphite and other carbon bearing rocks for comparison. 1-4: graphite types (table 3); M: primary carbonates (type 1-2); CV: vein carbonates (type 3-7); A(−Opx): amphibolite facies graphites from Andrea (1974); A(+Opx): granulite facies graphites from Andrea (1974); P: late carbonates from Pineau and others (1981); AVG: average for each group.

Type I and 2 calcites in the Bamble Sector show decreasing $\delta^{18}O$ and $\delta^{13}C$ values with decreasing metamorphic gradient (fig. 2). The calcites from the inner part of the individual marbles cluster close around a distinct isotopic trend. Calcites from the rim of the marbles usually show a small shift toward lighter isotopic compositions (samples MB 034 and MB 034a in fig. 2). As there is no increase in decarbonation toward the boundary of the marble with the more siliceous metapelites of the country rock (that is, the calc-silicate minerals are homogeneously distributed throughout the marble), such a shift toward a lower $\delta^{13}C$ must indicate an increasing interaction with fluids characterized by somewhat lower $\delta^{18}O$ and much lower $\delta^{13}C$ values. This implies that at the time of that particular fluid interaction, the marble bodies were comparatively impermeable to fluids, that is, represented aquitards (Thompson and Connolly, 1990; Ferry, 1991).

A regional isotopic trend might be caused by variable progressive decarbonation. Prograde decarbonation liberates CO$_2$ and may consume water if so required by reaction stoichiometry and will consequently cause high CO$_2$/H$_2$O ratios in the coexisting fluid. These ratios commonly increase gradually with the metamorphic gradient. Touret (1971b, 1972) observed such a shift in the composition of fluid inclusions in
quartzites and in quartz veins in gneisses in the Bambale Sector. Very few
CO$_2$-rich inclusions have, however, been found near or in formerly
carbonate-rich rocks (Touret, 1985, 1986). Since forsterite, diopside,
phlogopite, and tremolitic amphibole are commonly found, the follow-
ing reactions must have proceeded in the Bambale marbles and consumed
dolomite:

1. $\text{Dol} + 2 \text{Qtz} \rightleftharpoons \text{Di} + 2\text{CO}_2$
2. $3\text{Dol} + \text{Kfs} + \text{H}_2\text{O} \rightleftharpoons \text{Phl} + 3\text{Cal} + 3\text{CO}_2$
3. $13\text{Dol} + 2\text{Kfs} \rightleftharpoons 6\text{Fo} + 13\text{Cal} + \text{Spl} + 13\text{CO}_2 + \text{K}_2\text{O}$
4. $5\text{Dol} + 8\text{Qtz} + \text{H}_2\text{O} \rightleftharpoons \text{Tr} + 3\text{Cal} + 7\text{CO}_2$
5. $\text{Dol} + \text{Crn} \rightleftharpoons \text{Spl} + \text{Cal} + \text{CO}_2$

Corundum was not found in any thin section but may have been
present for example, in MB 182, which has too much spinel to be
explained only by reaction (4). The corundum may have been replaced
by spinel according to reaction (6). This early spinel then served as a
nucleus for later spinel that formed from reaction (4). Dolomite is still a
major constituent in most marbles from locality D1 and some other
marbles (table 2). This indicates the presence of excess dolomite with
respect to quartz and K-feldspar, obstructing further reaction. The two
exceptions to the cluster of isotopic values for locality D1 have almost no
dolomite (MB 034 and MB 034a: 0.4 percent) or no dolomite at all (MB
182). In contrast, the marble from locality A4 (TN 35) with its higher
isotopic values than marbles from locality D1 still contains dolomite.

Production and subsequent escape of carbon dioxide during pro-
grade metamorphism will result in a fractionation of oxygen and carbon
isotopes. The resulting shifts in isotopic composition of the calcites may
be described in terms of the combination of two endmember processes:
(1) batch devolatilization in which the fluid produced escapes at once
after reequilibration with the rock, and (2) Rayleigh devolatilization in
which the fluid is expunged from the system immediately after produc-
tion, without any reequilibration (Valley, 1986). Rayleigh and batch-
de carbonation are physically speaking endmember processes; in most
natural settings, decarbonation will take place by a process intermediate
to these extremes. For small amounts of devolatilization (that is, high
$\f$-values), the difference between these processes is small. For high
decarbonation levels ($\f(\text{C}) < 0.2$), Rayleigh devolatilization will produce
a significantly larger shift in $\delta^{13}\text{C}$ (Valley, 1986; Ferry, 1991).

To establish the contribution of prograding metamorphic reactions
and related CO$_2$-production on the isotopic composition of the Precam-
brian marbles, the measured isotopic compositions of type 1 and 2
calcites have been recalculated to initial values (fig. 4) using reaction
progress analysis after Rice and Ferry (1982). The recalibration proce-
dure was identical to that outlined by Bol and others (1990). A field for
non-metamorphic Proterozoic carbonate sediments (Schidlowski, Eich-
mann, and Junge, 1975; Veizer and Hoefs, 1976; see also Anderson and
Arthur, 1983) has also been indicated. It should be kept in mind that
these carbonates may have interacted with meteoric water after the
Fig. 4. $\delta^{13}$C versus $\delta^{18}$O plot with calculated initial isotopic compositions of primary carbonates corrected for decarbonation, assuming a Raleigh model. Open figure = measured composition; solid figure = calculated initial composition. Shaded area indicates non-metamorphic Proterozoic carbonates after Schidlowski, Eichmann, and Junge (1975).

Proterozoic, whereas the Bamble marbles were metamorphosed during the Mid-Proterozoic. Nevertheless, marbles from granulate facies localities D1 and (partly) C1 coincide with the field of non-metamorphosed Proterozoic carbonates. Marbles from localities in zones B and A plot increasingly farther away from this field, with ever decreasing $\delta^{13}$C values toward lower metamorphic grade.
The initial isotopic compositions of the marbles inferred from the reaction-progress calculations with total carbonate (Rice and Ferry, 1982) still form a trend comparable in magnitude to a metamorphic “calcsilicate-producing-reaction” trend. Furthermore, the spread in calculated initial compositions is greater than those measured. In general, type 1-2 calcites from zones D-B display Rayleigh-type decarbonation as indicated by the length and dip of the lines connecting measured δ\(^{13}\)C to δ\(^{18}\)O compositions with calculated initial compositions (fig. 4). A group of zone D marbles enjoyed batch-decarbonation instead, and for some samples the decarbonation process was intermediate between these extremes. This illustrates once more the very local control of metamorphic reactions.

Assuming that all primary carbonates had an initial isotopic composition within the box of non-metamorphic Proterozoic carbonates, figure 4 shows that decarbonation cannot cause the measured shift in isotopic compositions. The trend displayed by the investigated marbles remains after correction of the isotopic values with the volatilization model. In addition to this, no systematic relationship can be established between the average amount of reaction produced calcisilicates (and hence CO\(_2\)) and the observed isotopic composition, since the mineral modes are not related to metamorphic zoning. The observed trend in isotopic compositions is apparently caused by processes other than prograde decarbonation alone.

The variation in isotopic composition of the marbles may be considered as the result of open system fluid-calcite exchange with either pure CO\(_2\) fluid or with aqueous fluids containing only a few percent of CO\(_2\). To discriminate between these two possibilities we used the method described by Rye and Bradbury (1988). A δ\(^{13}\)C of -5.0 permil (Kyser, 1986) is assumed for the CO\(_2\) of the fluid used in modelling, as the carbonic fluids present in the granulite facies rocks of the Bamble Sector are supposed to have been derived ultimately from the mantle (Touret, 1971b, 1985, 1986; Hoefs and Touret, 1975; Frost, Frost, and Touret, 1989). Due to the relatively large amount of oxygen in the surrounding siliceous country rocks, the δ\(^{18}\)O of the model fluid is considered to be buffered by and in equilibrium with quartz having an average δ\(^{18}\)O of 10 permil. Even sample TN 35 with its highest δ\(^{13}\)C and δ\(^{18}\)O values enjoyed decarbonation as witnessed by the calcisilicates. We therefore take the Rayleigh-corrected isotopic composition of this sample as the minimum initial value for all type 1 and 2 Bamble calcites.

The marbles and the respective associated veins of localities D1, C1, B1 define a convex curve in the δ\(^{13}\)C versus δ\(^{18}\)O diagram (fig. 2A). This implies that if the trend results from fluid-rock interaction during metamorphism, this trend typically points to a water-rich fluid (Rye and Bradbury, 1988). This is supported by the fact that fluid inclusions in the marbles are composed mainly of water (Touret, 1986) and by the occurrence of vesuvianite in the peak metamorphic mineral assemblage of the skarns at locality C1, which also indicates high H\(_2\)O/CO\(_2\) ratios (Valley and others, 1985).
However, the outermost margins of the marbles show small shifts with respect to the isotopic values of their cores. The isotopic values of the margins are very close to those of type 3 and 4 associated calcite veins. It is unlikely that these shifts are caused by deformation assisted processes, as the contacts between the marbles and the neighboring rocks are neither sheared nor more deformed than the surrounding lithologies themselves. No petrographic evidence exists for a higher degree of alteration of the margins with respect to the cores of the marbles (see also table 2). Therefore, the shift in δ\textsuperscript{13}C (and to a lesser degree in δ\textsuperscript{18}O) of the rims as compared to the cores may indicate local isotopic exchange with an external CO\textsubscript{2}-bearing aqueous fluid during metamorphism. If we assume that this fluid was homogeneous and in equilibrium with the highest average calcite (δ\textsuperscript{13}C = -11 permil and δ\textsuperscript{18}O = 10.5 permil), it is possible to model the CO\textsubscript{2}/H\textsubscript{2}O ratio of the fluid and the degree of exchange within the individual marbles. The ratios of the amounts of initial and decarbonation CO\textsubscript{2} in the vein calcites can also be calculated. The model after Rye and Bradbury (1988) is principally independent of temperature. Their curves show that within the specified limitations, the calcites may indeed have been crystallized by exchange with water-rich fluids. However, the amount of water may be overestimated since the large silicate reservoir hosting the marbles may have provided additional oxygen to the marbles. As no additional carbon may be derived from this reservoir, additional oxygen must be attributed to the influence of water. This suggests ostensibly lower CO\textsubscript{2}/H\textsubscript{2}O ratios.

The isotopic trend displayed by the Bamble marbles is reversed to the one commonly observed marble in amphibolite to granulite facies transition zones (Lofoten-Vesteralen, northern Norway; Baker and Fallback, 1988), which is coupled to increasing metamorphic grade. The isotopic trend of type 1 and 2 calcites is remarkably similar to the one displayed by chlorite zone vein and wall rock carbonates of the Waterville Limestone, Maine (Rumble and others, 1991). These authors found no conclusive arguments to exclude either a diagenetic or infiltrative metamorphic origin for this trend but favored the latter.

A reversed isotopic trend similar to the one defined by the Bamble marbles was found in Proterozoic granulite facies marbles around the anorthositic Rogaland Igneous Complex, southwestern Norway (Bol and others, 1990). This carbonate trend from Rogaland is supported by the trend in δ\textsuperscript{18}O values of coexisting phlogopite and diopside. This indicates that the trend must result from pre-metamorphic or prograde interaction and not of syn- to post-metamorphic reactions and evidently not of retrogradation (Sauter, 1983), despite Caledonian burial metamorphism which partially reset K-Ar and Rb-Sr isotopic systems (Verschure and others, 1980). Bol and others (1990) showed that the reversed trend neither related to infiltration of metamorphic fluids nor to magmatic fluids derived from the anorthosites. They attribute the observed isotopic shift to low temperature exchange during early metamorphic or diagenetic processes. The heaviest part of both the Bamble and Rogaland
trends are strikingly similar to the lightest part of the trend displayed by depositional to prograde diagenetically altered dolomites from the Precambrian Beck Spring Dolomite, California (Tucker, 1982).

Summarizing, the reversed marble trend resulted from exchange with a water-rich, CO$_2$-bearing fluid with relatively light isotopic compositions, with diminishing effect toward higher metamorphic grade. This raises the following questions: (1) What was the relative timing of marble-fluid interaction with respect to metamorphism?, and (2) Did retrogradation affect the isotopic composition of the calcites?

The oxygen isotope system.—The δ$^{13}$C and δ$^{18}$O marble trend (fig. 2A) is generally followed by the arrangement of the isotopic values of type 3 veins (fig. 2C). The associated veins (types 3-4) generally cut across the marble fabric or that of other nearby rocks. In zones B, C, and D the average isotopic compositions of the associated veins are lower than those of the corresponding marbles. The spread in isotopic composition is larger for the vein calcites than for the respective associated marbles, especially so for δ$^{13}$C. It is evident that the isotopic composition of the vein forming fluids was influenced by the buffering action of the nearby rocks, not only in oxygen, but typically also in carbon. The δ$^{18}$O values of vein calcites with a close spatial relationship to marbles are restricted to a compositional band between 10 and 12 permil. The δ$^{18}$O values of marble calcites of the C1 locality and some of the D1 locality also plot in this band. The vast amount of oxygen in the country rocks (compared to the small vein calcite O-reservoir) obviously controlled their oxygen isotopic composition.

Quartz from segregations in enderbritic gneisses on Tromøy contains CO$_2$-rich fluid inclusions, which may have been trapped from the segregation-forming fluid. The δ$^{18}$O isotopic composition of quartz in orthopyroxene-bearing segregations in enderbitic gneisses from Tromøy may serve to identify the δ$^{18}$O isotopic composition of the metamorphic fluid and consequently that of the rocks passed by the segregation-forming fluid. The quartz from the segregations has δ$^{18}$O values between 8.1 and 10.8 permil, while a discordant quartz vein yielded 7.8 permil. The range of δ$^{18}$O values of the vein calcites (9.5-10.8 permil) is consistent with the highest values (9.3-10.8 permil) for the quartzes, taking equilibrium fractionation between calcite and quartz at relevant temperatures into account. Pineau and others (1981) reported δ$^{18}$O values of 10.1 and 10.3 permil for quartz from enderbitic gneisses on Tromøy. In zone D and very probably in the entire Arendal area, quartz from veins and the surrounding rocks have comparable δ$^{18}$O isotopic compositions. Hence, the δ$^{18}$O isotopic composition of the vein calcites can be said to be controlled by the country rock.

The carbon isotope system.—The question remains—to what extent was the δ$^{13}$C isotopic composition of the metamorphic or infiltrating fluid controlled by the country rocks. If the amount of carbon in the fluid greatly exceeds that of the rock system, then its isotopic composition would remain fixed, and the precipitated calcites would have a uniform
composition. Such may happen if the fluid is sufficiently carbon rich at a low fluid/rock ratio or with a low-carbon fluid when the fluid/rock ratio is high. In all other cases, the isotopic composition of precipitated calcites will be buffered by the original sedimentary phases graphite and carbonate in the country rock. The buffering capacity of graphite in the area is much larger than that of calcite simply because (1) graphite is far more abundant and because (2) of its much higher content in C than calcite (100 percent compared to 12 percent by weight). The average amount of graphite in silicic metasedimentary granulites and gneisses is 0.2 wt percent, and that of carbonate 0.03 wt percent (Andreae, 1974). The average amount of intergranular CO₂ is less than 0.02 wt percent, as established from fluid inclusions in quartz (Pineau and others, 1981). Silicic metasedimentary rocks are ubiquitous in the Bamble Sector, whereas marbles are rare and constitute less than 0.1 percent by volume of the lithologies. Considering the molecular weights of graphite, calcite, and carbon dioxide, the overall carbon isotope buffering potential offered by graphite exceeds that of calcite by far. Relatively ineffective buffering capacity of graphite is at least partly counterbalanced by its relative abundance. The local action of the marbles as a CO₂-reservoir is illustrated by the fact that the isotopic composition of calcite in associated (type 3-4) veins is directly related to that of the marbles.

Pineau and others (1981) presented isotope analyses on carbonate traces originating from the alteration of feldspars in high grade gneisses. Their δ¹³C values for the same area range from −10.6 to −4.3 permil, which is similar to our vein calcites. The higher values coincide with our range of −5.8 to −3.3 permil of type 4 veins whose mineralogy (axinite, datolite, et cetera) indicates a low grade genesis. Their corresponding δ¹⁸O values range from 10.7 to 24.5 permil. Of these, the lower values coincide with the composition of calcites from associated veins (types 3-4), whereas the higher values are comparable with the δ¹⁸O values of our unassociated veins of types 6 and 7 and some of type 5. From the combined data, it may be concluded that the δ¹³C of these vein calcites is inherited from metamorphic carbonates, whereas the variable δ¹⁸O reflects an increasing influx of meteoric water toward higher values (Sheppard, 1986). The vast majority of the marbles from localities D1 and C1 fall outside the range of typically retrograde δ¹³C values of unassociated veins, indicating that only minor retrogradation took place.

Retrograde δ¹⁸O fractionation temperatures of 365°C between secondary amphibole and calcite were calculated for four orthogneisses (Pineau and others, 1981). This implies that the secondary calcite would have been in equilibrium with CO₂ having a δ¹³C between −8.4 and −1.3 permil and with graphite of δ¹³C between −20.4 and −13.4 permil (Chacko and others, 1991). The empirical calibration of Dunn and Valley (1992) yields graphite values of −23.3 and −13.0 permil.

The graphite C-reservoir.—The δ¹³C values of disseminated graphite in fahlbands and metapelites from zone A vary between −28.3 and −20.2 permil (table 5). They provide isotopic evidence for a biogenic origin of
the graphite as has already been argued by Field and Starmer (1982) from field observations. The average δ^{13}C of type II graphites from metapelites is evidently lower than those of the faahband gneisses (type I). It emphasizes the lack of communication that may occur between the two lithologies in one outcrop.

It might be argued that the isotopic buffering potential of graphite is ineffective due to slow exchange kinetics between graphite and CO₂, and that the only contribution of graphite would have been in a chemical way by means of oxidation to CO₂ or reduction to CH₄, which was then expelled and added to the fluid. It has been argued that metamorphism in the Bambale Sector occurred under relatively oxidizing conditions (Cameron, 1989), which is supported by the fact that fluid inclusion evidence for the presence of CH₄ is scarce (Touret, 1985), and whole rock Fe^{3+}/Fe^{Tot} determinations (Field, ms; Beeson, ms) as well as Fe^{3+}/Fe^{Tot} estimated for biotites and amphiboles (Nijland, 1993).

Investigations by Kreulen and Van Beek (1983) and Dunn and Valley (1992) have made clear that isotopic equilibrium between calcite and graphite via CO₂ is attained at temperatures above about 620°C. The inferred metamorphic temperatures of about 750°C for the amphibolite facies to about 840°C for the granulate facies (Nijland and Maijer, 1993) in the Arendal area fulfill the temperature prerequisite for equilibrium, that is, slow kinetics are ruled out for not attaining equilibrium. A metamorphic fluid in equilibrium with graphites from types I and II may have contained CO₂ with δ^{13}C values of −18 to −14 permil at 700°C, which is far too low to be in carbon isotopic equilibrium with the marbles and veins (Chacko and others, 1991). Thus, fluid exchange between the marbles, faahband gneisses, and metapelites was negligible or small.

If the regional isotopic trend of the type 1-2 marble calcites reflects a variation due to fluid infiltration at granulate conditions, then graphites of types I and II would be expected to illustrate a δ^{13}C trend aligned to but with lower values than the calcite trend. This is obviously not the case. Andreac (1974) measured δ^{13}C values of graphite from both the amphibolite and granulate facies zones. Combined with our analyses, these data do not show any isotopic trend on a regional scale for graphite. Type I and II graphites maintain their specifically low δ^{13}C values throughout the metamorphic zoning.

Furthermore, graphite is still a common accessory phase in many metasediments (Andreae, 1974; Pineau and others, 1981; Field and Starmer, 1982). If large amounts of graphite were oxidized and the resulting CO₂ expelled to the fluid, then the buffering capacity of the graphite was apparently larger than the oxidizing capacity of the fluid. Moreover, in order to explain the observed trend in the primary calcites, the amount of graphite should be substantially lower in the amphibolite facies zones that in the granulate facies terrain. If so, the then stronger oxidation in the amphibolite facies should have produced more of ¹³C-depleted CO₂ after re-equilibration, resulting in a lower δ^{13}C calcite
as observed. Instead, the graphite content of amphibolite facies zones is higher than that of the granulite facies zone (Field and Starmer, 1982).

The relatively long (that is, 350 Ma) burial of the Bamble Sector under high grade conditions (Visser and Senior, 1990) and the relatively widespread abundance of graphite may have at least partly compensated the slow exchange kinetics. Taking all the above into account, it seems hard to argue that the amphibolite to granulite facies transition originates from substantial infiltration of an externally derived fluid or from flushing by decarbonation derived CO₂ out of zone D.

The average δ¹³C of 19 amphibolite facies graphites is −20.9 permil; the average of 19 granulite facies graphites is −21.5 permil (Andreae, 1974; this study). Granulite facies graphites are thus lighter and have more biogenic compositions than their amphibolite facies counterparts. If we arbitrarily take a δ¹³C of −22 permil as the upper limit for an unchanged biogenic isotopic composition, the lighter δ¹³C values for the graphite imply closed rock systems with respect to CO₂. Apparently, closed system behavior occurred at both sides of the orthopyroxene isograds. The average amount of graphite in amphibolite facies samples is 0.23 wt percent, whereas it is only 0.19 wt percent in the granulite facies samples. This might suggest that more graphite was consumed by local buffering or degassing reactions under granulite than under amphibolite facies conditions. Despite the higher amounts of graphite in the amphibolite facies gneisses, their average δ¹³C values are shifted by 1 permil toward the isotopic composition of hydrothermally precipitated graphite (types III and IV; see below) encountered in quartz veins. This could be explained by (1) a larger amount of fluid in the open rock systems of the amphibolite facies terrain than in the granulite facies zones or (2) heavier CO₂ in the amphibolite facies fluids. The second option obviously is in conflict with the reversed isotopic trend defined by type 1-4 calcites from marbles and associated veins.

The large size of the idiomorphic graphite flakes together with their random orientation, not intergrown with mica, provides evidence for a hydrothermal origin for five quartz veins (type III) and some migmatic neosomes (type IV) from zone A. The isotopic composition of these graphites (−20.2 to −15.3 permil) consequently provides direct information on the carbon isotopic composition of the fluids from which they precipitated. The δ¹³C values of vein (type III-IV) and disseminated graphites (type I-II) are clearly different. This may indicate that (1) no isotopic exchange took place between graphite and CO₂-bearing fluid due to kinetic inhibitions, or (2) the fluids moving through the veins were isotopically different from those passing through or resident in the gneisses, and that the carbon isotopic systems of both rock systems were separated. For type IV graphites, which occur in hydrothermal quartz bodies, isotopic isolation of option (1) seems reasonable. In contrast, type III graphites occur in migmatic neosomes. As the precipitation of graphite from granitic melts is uncommon, this graphite may be refrac-
tory. In this case, their original isotopic signature has been changed and consequently will be due to option (2). The isotopic signature of the carbon dioxide precipitated from the veins may find its origin in mixing of small quantities of decarbonation-derived CO\textsubscript{2} with aqueous fluids containing externally derived CO\textsubscript{2}. The spread in δ\textsuperscript{13}C of the vein graphites makes clear that either the vein forming fluid was inhomogeneous or consisted of a number of different pulses that cannot be distinguished on vein-structural basis.

The range of δ\textsuperscript{13}C values of the fluid as inferred from the graphite data at 700°C does not agree with the range of δ\textsuperscript{13}C values of type 5-7 calcites from the unassociated veins. However, at a retrograde temperature of 400°C, the graphites would be in equilibrium with CO\textsubscript{2} with δ\textsuperscript{13}C between −11 and −7 permil, and calcites precipitated from this CO\textsubscript{2} would have δ\textsuperscript{13}C values between −9 and −4 permil. Most of our vein calcites (types 3-7) also show values in this range. Therefore, it may be concluded that most vein calcites represent retrograde phenomena in equilibrium with hydrothermally precipitated graphites. Some of the hydrothermal graphites occur in neosomes, so the conclusion above requires that these neosomes acted as fluid pathways during the retrograde history. It is truly a pity that graphite and calcite were not encountered in the same vein, but some graphites were sampled in the direct vicinity of calcite-bearing veins. If these are considered as graphite-calcite pairs, they have Δ values of about −12 to −7 permil (sample locations MB: A2-A5, Aa-Ae; for graphite values, see table 5; for calcite values, see table 4B), that point to equilibration temperatures of 225° to 425°C, using the fractionation curve of Chacko and others (1991), which is well below peak-metamorphic temperatures (Lamb, Smalley, and Field, 1986; Nijland and Maijer, 1993). This temperature also compares well to the retrograde temperatures of 365°C of Pineau and others (1981) for pairs of calcite and secondary amphibole from orthogneisses.

Implications for amphibolite to granulite facies transition.—The isotopic investigations on marbles, calcite veins, quartz segregations, disseminated graphite, and hydrothermal graphite veins indicate a general basence or at least a low abundance of metamorphic fluids during granulate facies metamorphism. The regional shift in isotopic composition of the marbles, the lack of isotopic communication between calcite and disseminated graphite, and the very limited isotopic interaction between veins and country rock are all expressions of this general deficiency of metamorphic fluids, particularly CO\textsubscript{2}-rich fluids, in the amphibolite to granulite facies transition zone in the Bamble Sector. Complex intergrowths like symplectites and coronas of several hydrous and even anhydrous minerals occur in various rocks throughout the Bamble Sector. Halogen distributions among high-grade metamorphic minerals in amphibolites illustrate extremely local, internally controlled rock systems (Nijland, Jansen, and Maijer, 1993). The small scale heterogeneities in element distributions in many mineral assemblages provide evidence that mineral reactions were controlled by solid state diffusion.
rather than by fluid controlled diffusion (Tracy and McLellan, 1985). Due to the fluid deficiency, the disseminated graphite and marble calcites resisted chemically and isotopically the granulite facies metamorphism. Only in spatially related veins did sufficient water-rich fluid control the isotopic exchange between calcite or graphite and CO$_2$ of the fluid. Most of these veins contain minerals like epidote, prehnite, chlorite, axinite, et cetera (table 2) and consequently represent retrograde metamorphic conditions.

The question remains why the occurrence of CO$_2$-rich fluid inclusions narrowly coincides with the orthopyroxene-in isograd for basic rocks in the amphibolite to granulite facies transition zone in the Bamble Sector (Touret, 1971, 1972). Several authors demonstrated the retrograde entrapment of CO$_2$-rich fluids (for example Lamb, Valley, and Brown, 1987; Lamb, Brown, and Valley, 1991). Morrison and Valley (1991) draw attention to the fact that charnockitic gneisses owe their typical color to the retrograde development of chlorite (Howie, 1967). This led these authors to suggest that the alteration of orthopyroxene to chlorite and feldspars to sericite could locally transform small amounts of mixed H$_2$O-CO$_2$ fluid into a CO$_2$-rich fluid as chlorite and sericite forming reactions proceed, consuming H$_2$O and precipitating calcite and CO$_2$-rich fluid inclusions. A similar process to explain the CO$_2$-dominated fluid inclusions, involving reaction of fluid and orthopyroxene to hornblende and the inclusions, has been proposed by Konnerup-Madsen (1979). These hypotheses may at least partly explain the enrichment in carbon dioxide of the inclusions in granulite facies rocks but not the origin of the CO$_2$ involved.

Pineau and others (1981) and Van de Kerkhof, Touret, and Kreulen (1994) measured $\delta^{13}$C values of CO$_2$ of fluid inclusions from quartz veins. Commonly, such bulk analyses of fluid inclusions represent mixtures of several generations of inclusions. Nevertheless, the carbon isotopic composition might be inherited from trapped high grade CO$_2$ present at peak metamorphic conditions, assuming no exchange with later fluids occurred. Small particles of calcite occurring in some trails of fluid inclusions may affect enormously the results of their isotopic analysis by thermal decrepitation (Van den Kerkhof, personal communication); therefore, only crushing experiments give reliable results. The $\delta^{13}$C values of the CO$_2$ from fluid inclusions derived from these experiments vary between $-6.5$ and $-5.3$ permil. This corresponds with the $\delta^{13}$C values of the retrograde vein calcites, which range from $-9$ to $-4$ permil. The compositional range is inconsistent with the calcite compositions of localities A3 and D1 as well as with the composition of graphite over the entire area. The fluid inclusions only reflect the composition of local, mainly retrograde, channelized fluids that were not able to exchange freely with the surrounding rocks. As demonstrated by several authors (Lamb, Valley, and Brown, 1987; Lamb, Brown, and Valley 1991; Morrison and Valley, 1991), a high density of (part of) the fluid inclusions does not disprove a retrograde entrapment of those inclusions. According
to Bakker and Jansen (1991), lower density (retrograde) fluid inclusions may contain old (high grade) CO₂.

**Origin of the marble trend: Early Gothian diagenesis?**.—The fact that the disseminated graphites display a fairly uniform composition not related to metamorphic zoning whereas the primary marble calcites define a reversed isotopic trend may stress the difference in their premetamorphic composition. The graphite composition originated from sedimentary, biogenic precursors, and its δ¹³C composition is inherited therefrom. Sedimentary calcites are as a rule relatively heavy in oxygen and carbon isotopes and uniform in composition for a specific depositional environment (Veizer and Hoefs, 1976). However, our primary calcites (type 1-2) are not heavy in ¹⁸O. Thus, the isotopic compositions and the invoked trend displayed by such primary calcites from the Arendal area were set by a low grade process that did affect the calcites but left the graphites untouched. Such a low grade process under diagenetic or anchizonal conditions typically has a large isotopic fractionation between calcite and fluid (Irwin, Curtis, and Coleman, 1977). Despite low reaction and diffusion rates under such conditions, a large isotopic shift may be produced (Hudson, 1977; Dickson and Coleman, 1980).

It is striking that the initially dolomitic marbles exposed in both the Bamble and Rogaland granulite facies terranes of southern Norway display reversed isotopic carbon and oxygen trends; it has been independently argued that these trends predate peak metamorphism and were most probably diagenetic. The reversed Bamble and Rogaland isotopic trends do not correlate chronologically with the metamorphic trend from lower toward higher grade metamorphism. The trends seem to be proportional to the inland distance from a pre-Caledonian coast line. The distance may correlate with a burial depth during Early Gothian diagenesis or incipient metamorphism. For the Bamble Sector, the few sedimentary structures preserved in the area show the same constant upward younging direction of the supracrustal suite (T.G. Nijland and C. Maier, unpublished data; P. Padget, personal communication). This implies that zone A amphibolite facies marbles must have occupied a lower stratigraphic position than the zone D granulite facies marbles. As mentioned earlier, the uppermost part of the Bamble trend perfectly forms an extension of the lowermost side of the trend displayed by the Proterozoic Beck Spring dolostones studied by Tucker (1982). The reversed isotopic trend being an extension supports the idea of having been invoked by diagenetic or anchizonal metamorphic processes as described by Irwin, Curtis, and Coleman (1977) and Dickson and Coleman (1980).

**SUMMARY AND CONCLUSIONS**

1. The trend in type 1-2 primary carbonate combined with the lack of a similar trend in primary type 1-2 graphite is incompatible with pervasive flushing at high metamorphic grade with a CO₂-rich fluid of mantle origin. In such case, all initial sedimentary isotopic compositions would have been smoothened to a more or less uniform value. Due to
fluid deficiency, disseminated graphite and marble calcites chemically and isotopically persevered high grade metamorphism.

2. The $\delta^{13}C$ composition of type 1-2 primary carbonate can be coupled directly to the metamorphic zoning of the Bamble Sector but is falsely correlated with it in both time and space (reversed). The $\delta^{13}C$ composition of type 3-7 secondary carbonates and of type I-IV graphites cannot be related to this zoning but can instead be imputed to the respective hosting lithologies.

3. Decarbonation reactions alone, consuming original dolomite and producing calcisilicates, cannot be responsible for the observed trend in the primary carbonates. Remodelling of the type 1-2 carbonates assuming Rayleigh fractionation shifts the isotopic composition of these primary carbonates only a few permil toward a "Proterozoic nonmetamorphic sedimentary carbonate" composition. This implies that the isotopic trend has to be pre-decarbonation reaction.

4. Metamorphic fluid-rock interaction is very limited in space. Only veins of type 3-4 with a close spatial relationship to marbles of type 1-2 have a clearly marble derived isotopic $\delta^{13}C$ composition. Mineral reactions were solid state diffusion controlled rather than fluid diffusion controlled, as supported by data on element distributions.

5. The isotopic $\delta^{18}O$ composition of secondary type 3-7 carbonate is buffered by the overwhelming amount of oxygen provided by the country rock. With the exception of most type 5b veins that are very late (post ductile deformation) and two of type 3a plus one of type 7 that are weathered (MB 082, TG 63, MB 197), all carbonates have a $\delta^{18}O$ composition coinciding with that of Tromøy quartz lenses.

6. Most type 1-2 marble carbonates from zones D and C have not only preserved their initial premetamorphic sedimentary isotopic values throughout high grade metamorphism by their refractory behavior but also during subsequent retrogradation. The confinement of retrograde alteration in the marbles to small trails underscores the relative impermeability of the marbles as compared to the surrounding lithologies and the channelization of the altering fluids.

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REFERENCES


Andreæ, M. O., 1974, Chemical and stable isotope composition of the high grade metamorphic rocks from the Arendal Area, S. Norway: Contributions to Mineralogy and Petrology, v. 47, p. 300–316.


Barth, T. F. W., 1925, On contact minerals from Precambrian limestones in southern Norway: Norsk Geologisk Tidsskrift, v. 8, p. 93–114.


