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Article

# Ophiolite Association in the Rhodope Massif as indicator of the paleogeographical setting

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**Abstract:** All ophiolite associations mark epochs of active tectonic movements, which lead to significant petrological processes and modification of the relief of the Earth's crust. Here we present a geological-petrographical characterization of one ophiolitic associations composed of: a) serpentinites; b) amphibolites-metamorphosed volcanic rocks and tuffs; c) metagabbros and metagabbrodiabases, placed among the Proterozoic metamorphic complex in the Rhodope Massif of Bulgaria on the Balkan Peninsula, South-Eastern Europe. The goal is to clarify the paleogeographical and geological setting during its creation. The methods of lithostratigraphic profiling and correlations on the database of geological field mapping were used, supplemented by microscopic, geochemical and isotopic studies of numerous rock samples. The summarized results confirm a certain stratigraphic level of the Ophiolite Association among the metamorphic complex and a complicated and protracted heterogenetic development, which is typical for the ophiolite associations created in eras of closing oceans, opposite movement of tectonic plates, subduction-obduction environment with appearance of autochthonous Neoproterozoic magmatism. Obducted fragments of serpentinites mark an old erosional continental surface, subsequently covered by transgressively deposited pelitic-carbonate sediments. The general conclusion of our study confirms the concept that the metamorphic complex of the Rhodope Massif represents a unified stratigraphic system consisting of two petrographic groups of different ages, with which we oppose the idea of a trust construction, launched by a group of geologists.

**Keywords:** ophiolites; serpentinites; obduction; magmatism; Neoproterozoic; Rhodope Massif; Bulgaria

## 1. Introguction

The article offers a systematization and analysis of geological data for an Ophiolite Association from the Rhodope Massif, Bulgaria, South-Eastern Europe. The goal is to achieve new knowledge about the paleogeographic situation and dynamic processes during a period of the Neoproterozoic—700–550 Ma. The Ophiolite Association was chosen as the basis of the study because the creation and development of such formations mark epochs of active tectonic and petrological processes.

Interest in ophiolitic associations called "green rocks" began as early as the 18th and 19th centuries, but the first generalization was the so-called "Steinmann trinity", where three main rock types are distinguished: serpentinites, volcanics and gabbro/diabases [1]. The evolution of the ophiolite concept passed through conflicting views until it reached the Penrose Conference Definition in 1972, presenting ophiolites as a pseudostratigraphic sequence [2]. Coleman's [3] original obduction model was a successful solution for the implantation of serpentinite fragments on continental margins. Other researchers develop the concept in petrological and tectonic aspects [4,5].

The Ophiolites are distinctive and highly informative rock associations regarding in terms of plate tectonics. Then the relief of the ocean floor and the continental surface changes, new portions of igneous rocks appear, which renew the rock composition the old metamorphic terrains of Earth's crust. They considered to have been formed in different tectonic setting, marking epochs of oceanic closure and countermovement of the plates in which a suprasubduction setting is created upon their collision.

The Rhodope Ophiolite Association is a good example that reveals many moments of such development. The knowledge we have of this rock association has been acquired after many years of field and laboratory research on the spatial distribution and geological place in the structures of the Rhodope Massif. This allows us to determine with a fairly high degree of certainty its stratigraphic position in the metamorphic complex and the main petrographic, mineralogical and geochemical characteristics.

## 2. Materials and methods

In the Rhodope Massif is widespread a Neoproterozoic continental ophiolite association, composed of metamorphosed basic volcanites, metagabro and serpentinites, located among the metamorphic complex of the Rhodope Mountains. The serpentinites have been known since the beginning of the last century. The conditional geological mapping at a scale of 1:25,000 of the Rhodope Massif was carried out according to the lithostratigraphic method in 1948–1962 years, supplemented with stratigraphic correlations between different areas of the Rhodope Massif and thematic field research. The study was accompanied by laboratory microscopic observations and geochemical sampling. The results were summarized in a Geological Map of Bulgaria on a scale of 1:100,000, with descriptive Notes attached to it provided a complete picture of the distribution and the main petrographic characteristics of the serpentinites and amphibolites as well relationships between stratigraphic units and serpentinite bodies. Systematic geochemical study began with the works of Zhelyazkova-Panayotova [6] on the serpentinite from the Eastern Rhodopes.

The complex field and petrographic studies on the geological and stratigraphic position of ophiolites and their metamorphic changes led to the concept of a uniform heterogeneous Ophiolite Association undergoing regional metamorphism [7–11]. The finding of eclogites in metamorphic rocks posed the problem of high pressure metamorphism in Rhodope Massif [12–15]. A curious problem is the genesis of gabbronorites with a corona-structure [16–18]. Geochemical studies have revealed valuable mineralization of platinum and native gold in the serpentinites as well as provided material for interpretation on the geodynamic area of ophiolite creation [19–23]. The finding of microdiamonds included in garnet, sharpen attention to regional problems of construction and metamorphism in the Rhodope Massif [24–26]. One important achievement in knowledge of ophiolites was the determination of the absolute age of metabasites and serpentinites [27–30]. The determination of the Archaean age of the serpentinites is of key importance for the initial moments of the development of the ophiolites [31].

## 3. Geological setting

The Rhodope Massif is situated in the central part of the Balkan Peninsula—Southeast Europe on the territory of South Bulgaria and North Greece. The metamorphic basement of the Rhodope Massif is built of high-grade Precambrian metamorphic rocks divided into two complexes, named: Prarhodopian and Rhodopian Supergroups of different age and petrographic composition [32,33].

An updated version of lithostratigraphic division, based on additional field research, petrographic correlations and analyzes of the lithostratigraphic units affirm the existence of two complexes of different lithology and age: Prarhodopian and Rhodopian Groups (**Figure 1**) [34].

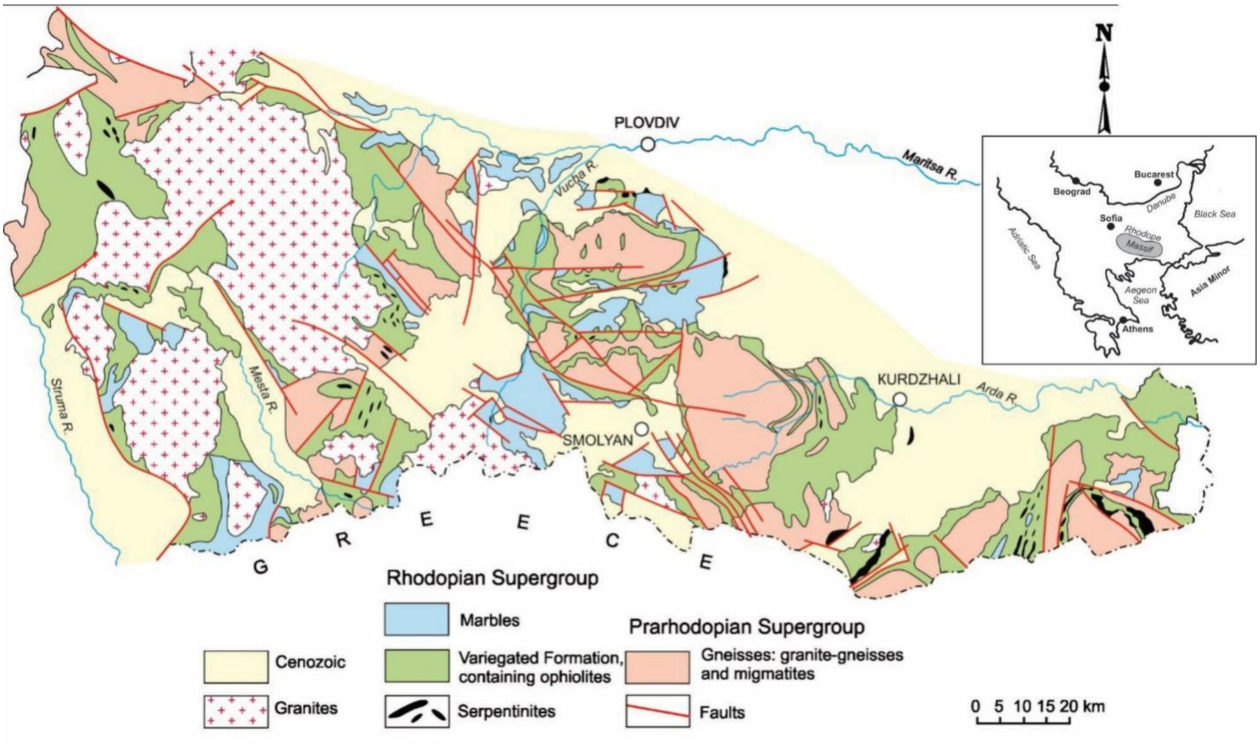


**Figure 1.** Geological map of the metamorphic complex of the Rhodope Massif.

The lower Prarhodopian Group (PRG) shows features of an ancient infracrustal continental complex, which may have been a fragment from some supercontinent. It consists of biotite and leptite gneisses with the packets of migmatic and granite-gneisses represented into three lithostratigraphic units up to top: Boykovo Formation, Bachkovo Formation and Punovo Formation. The absence of marbles is a specific feature of this group. The PRG builds up the core of anticlines and dome structures.

The upper Rhodopian Group (RG) is a well stratified supracrustal variegated complex that has been transgressively deposited on the Prarhodopian one. It is represented by metamorphosed volcanogenic-sedimentary rocks: amphibolites, eclogites, garnet-lherzolites, schists, quartzites, marbles, serpentinites, grouped in three parts, up to top: Lukovitsa Variegated Formation, Dobrostan Marble Formation and Belashtitsa Calc-silicate Formation. The Ophiolite Association is represented in the Lukovitsa Variegated Formation, where its rocks alternate with metamorphosed pelitic-carbonate sediments.

The Prarhodopian and Rhodopian Groups were subjected to folding at least twice. In the general structural plan, the diapiric raised domes and linear positive fold structures are clearly outlined by layers of the Rhodopian group. The spaces between them are occupied by deeply sunk subvertical, inclined or lying synclines, filled by the rocks of the Variegated Formation with ophiolites and marbles. Regardless of the folding deformations the ophiolites preserve their position in the crystalline complex and serve as basic stratigraphic marker. Sutures and deep tectonic zones, marked by ophiolites or discordant serpentinite wedges, are not found anywhere in the Rhodope Massif. The ideas of some authors presenting the Rhodopian Massif as Alpine nappe complex remained unproved by geological facts [35,36]. Later the author's views evolved with the recognition of "crustal-scale duplex terranes with different lithologies, deformation and metamorphic histories" [37].

## 4. The Ophiolite Association

### 4.1. Spread and stratigraphic position of the Ophiolite Association

The Ophiolite Association occupies a clearly defined stratigraphic position in the lower levels of the Variegated Lukovitsa Formation of the Rhodopian Group [34]. The association has an uneven area in the Rhodope Massif (**Figure 2**). It is more widespread in the Eastern Rhodopes. The largest serpentinite massifs as elongated bodies, lenses or megabudins in size from meters to 10–13 km in lentgth are located in the Eastern Rhodopes: Bela Reka dom-anticline, Avren syncline and Drandovo horst. The Ophiolite Association has awide and characteristic development in the Western Rhodopes—Gotse Delchev district. It covered large areas with orthoamphibolites among which bodies of metagabbro and serpentinites are revealed. In the Central Rhodopes and Pirin Mountains the serpentinites have a limited presence. A series of irregular serpentinite bodies are located also on the northern edge of the Rhodopes. Isolated small lenticular bodies are often found among the rocks of the Variegated Formations, in association with amphibolites and schists.

The serpentinite bodies are placed concordantly between the lower layers of the Lukovitsa Variegated Formation often directly on the gneiss sole of the Prarhodopian Group. Thus, the serpentinites mark the erosion level on the gneiss PRG complex and become a stratigraphic bench mark. They are covered or included by amphibolites and, less frequently, by schists and marbles. Discordant serpentinite wedges crossing metamorphic layers are not observed anywhere.

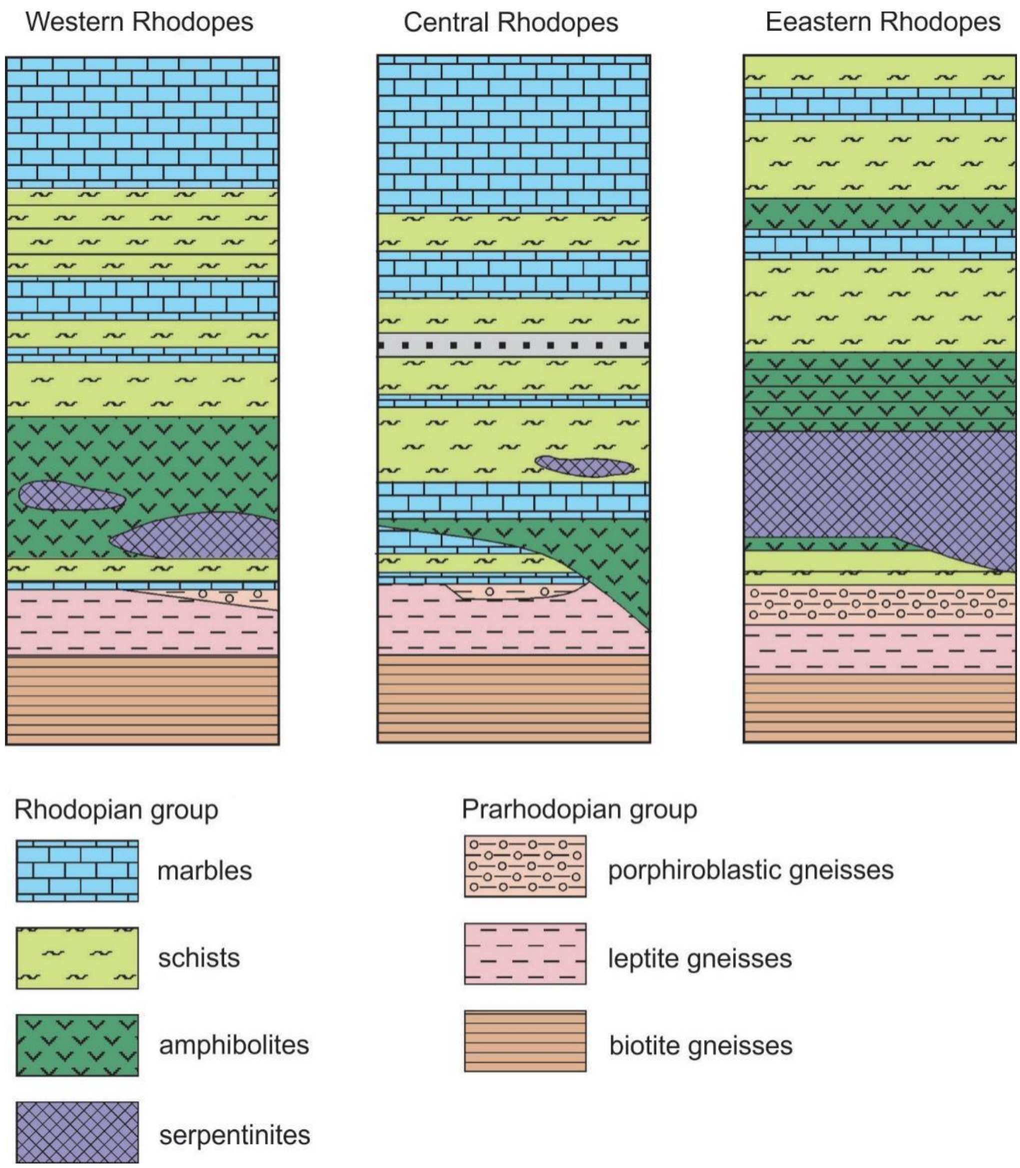


**Figure 2.** Stratigraphic columns of the Lukovitsa Formation in the Western, Central and Eastern Rhodopes.

## 4.2. Composition of the Ophiolite Association

The Ophiolite Association consists of: a) serpentinites; b) amphibolites (metamorphosed low potassium-high magnesium tholeiites and their tuffs); c) subintrusive bodies and dykes of metagabbros and metagabbrodiabases.

### 4.2.1. Serpentinites

The serpentinites are composed of lysardite, chrysotile and antigorite, rare relics of olivine (forsterite type), pyroxene and chromite. Lizardite is preserved inside large bodies. It fills the cells of the lattice microstructure characteristic of serpentinites, where together with chrysotile it forms a semi-isotropic, cryptocrystalline, vaguely fibrous mass, which often shows sectoral darkening. The cells are outlined by multi-layered chrysotile "cords", where the mineral builds cylindrical-fibrous individuals located perpendicular to the cell boundaries. Powdered to fine-grained magnetite, arranged in rows among the chrysotile, emphasizes the mesh structure. Larger chrysotile crystals fill cracks in serpentinite. Antigorite develops mainly on the peripheral parts of the bodies and in fault zones. The small thin bodies are composed entirely by fine scaly antigorite which orientation coincides with the general stratification and schistosity of the host rocks, evidence of its synmetamorphic crystallization.

The serpentinites from the Golyamo Kamenyne group—Eastern Rhodopes correspond to dunites and harzburgites, with the subordinate participation of lerzolites, pyroxenites, rhodingites and gondites developed mainly on the periphery of some bodies [6]. The rocks are ore-bearing. Relics of primary minerals-magnochromite in dunite zones, and chrompicotite in harzburgite zones are preserved. Sulfide copper-nickel and platinum mineralizations have also been found by Zhelyazkova-Panayotova [38], 1989. Chromites form nests and small bodies. They are classified into four groups: a) partially altered chromite; b) porous chromite; c) homogeneous chromite; d) zonal chromite.

Chromite deposits, forming about 200 bodies have been found also in the Dobromir serpentinite as well as mineralizations of native copper, gold, pyrhotite, nickel sulfides and elements from the platinum group [39]. Dobromirtsi serpentinites are enriched in Os, Ir and especially Ru and depleted in Pt and Pd [40]. During serpentinization and regional metamorphism, mobilization of some components occurs: enrichment of iron in the peripheral areas of the grains of chromium spinel [41], redistribution and recrystallization of native gold together with magnetite and tremolite. The chromites in chemistry are systematized in two groups: a) enriched with Os, Ir, Ru and b) enriched in platinoids, latter a third group with limited chromicity and increased magnesium has been distinguished [39-40]. The chemistry of primary magmatic chromite changes under the influence of metamorphic fluids. It is believed that the ultrabasic magma is fractionated by island arc toleite in archaic times—3000 Ma which underlined the continental crust, assimilated and later reworked.

Cr-Ni magnetite agregates present at the contacts of ultrabasic bodies with marbles, in the Central Rhodopes, Ardino region [42].

A number of ores have been also identified in the serpentinires from Western Rhodopes, Satovcha district as: magnetite, chromite, pentlandite, laurite, sulfarsenides and others the platinum group Os, Jr, Ru, Rh, Pt, Pd, as well as Au [43].

The chemical composition of serpentinites in their current form after Bazylev et al. [44], corresponds to dunites and peridotites with a magnesian coefficient M/F = 6 to 9. As in serpentinization the magnesian ratio increases the probability of extensive dunite involvement decreases. During serpentinization, silicate minerals such as olivine and pyroxene are highly altered, some of the calcium and iron content is extracted from the water, and the magnesian ratio increases. So, it must be assumed that the dunites were the smaller part of the serpentinite protolith where peridotites predominated.

The oldest ages according to U-Pb dating of zircons from the chromitites of the Dobromirtsi serpentinite massif indicate Paleoproterozoic era 2257 ± 80 Ma and 1952 ± 82 Ma, which is the age of the oceanic plate in the ancient ocean, from which the serpentinite fragments have been torn off [31].

The complex of petrographic, mineralogical, geochemical and isotopic data clearly indicates that the serpentinites are hydrated derivatives of peridotites from an old Archaean-Paleoproterozoic oceanic mantle plate.

### 4.2.2. Amphibolites

The amphibolites are widely distributed as layers of different thickness (0.5-15-20 m), alternating with amphibole-schists, amphibole-biotite or muscovite-biotite schists, gneisso-schists, carbonate schists and marbles. They are composed of amphibole (tschermakite-hastingsite) and plagioclase (andesine to bytownite) and with volatile and variable amounts of quartz, biotite, garnet, epidote, pyroxene, titanite, rutile, magnetite, ilmenite. Most of the amphibolites have clear foliation, but there are often those with a more massive texture. The rocks are fine, medium or coarse-grained, with a granoblastic structure. In terms of chemical composition, amphibolites correspond most often to high-magnesium toleites and less often to picrites.

In terms of chemistry, amphibolites correspond to gabbro group and according to basic toleic volcanics. Amphibolites from the Lukovitsa Formation, according to their content of trace and RRF elements, show a certain affinity for island-arc basic volcanics from the continental margins [45].

Amphibolites which are not affected by migmatization generally correspond to low-potassium toleite basalts, locally enriched in titanium (TiO 2%–4%) and iron (FeO 14%–18%), and to a lesser extent to basaltic and peridotite comatites. Variations in the main components are relatively limited. Zakariadze et al. [46] divided the amphibolites from the Eastern Rhodopes into three groups in the TiO vs. MgO. Examining the distribution of RRE the authors conclude that the high titanium amphibolites refer to the basalts of the mid-ocean ridges, while the low titanium ones are compared with those of the island arcs. According to petrochemical coefficients authors identified three groups of compositions: oceanic rift tholeiites, intraplate basalts and island arc tholeiites. Amphibolites from the Lukovitsa Formation, according to their content of trace and RRF elements, show a certain affinity for island-arc basic volcanics from the continental margins [45]. The amphibolites in Western Rhodopes correspond to toleite and high-magnesium toleiite basalts, with increased content of Fe, Ti, Al. Individual thin layers composed of actinolite schists, with high concentrations of magnesium, chromium and nickel, bring them closer to comatiites. The geochemical characteristics of individual samples of amphibolites from the vicinity of the villages Satovcha, Pletena, Oreshe and Kochan show affinity for island arc toleite and calcium-alkaline basalts and andesites [47].

Contemporary data of Bonev et al. [22] for the rare and trace elements in amphibolites indicate increased contents of Zr, Nb, Y, Ni, Cr interpreted as indicating a high degree of fractionation from a primitive mantle magma. According to the authors, the high-titanium group of metabasites has an affinity to the toleite magmas of the Mid-Ocean Ridges and partly of the Inland Ocean Plates while the low-titanium ones approach the island arc toleites.

Such a division by petrochemical calculations is difficult to find in accordance with the geological setting. The content of trace and RRE elements varies depending on the variations of the main components. There is a direct dependence in the contents of Fe and V, as well as between Mg and Cr, Ni, Co. The migmatization of amphibolites also increases the contents of Si, Al, alk, Ti, Sr and decreases that of Mg, Fe, Cr, Ni, Co.

The comparative analysis on the chemical character of the metamorphosed basic magmatites—amphibolites and gabbro shows their common belonging mainly to the group of basalts and a smaller part of them to the picrite basalts respectively to the normal and magnesium toleites. The great diversity of the chemical composition of the amphibolites, however, cannot be considered fully adequate to the primary composition of the magma, which does not give us the right to draw precise conclusions about the geodynamic zone of their creation. In our opinion, the great geochemical diversity in the amphibolites, where both basic and ultrabasic composition signatures are combined, is more reminiscent of contamination than of fractional magmatic processes. This circumstance supports an idea of suprasubduction rather than an island arc setting.

#### 4.2.3. Metagabbros

The metagabbros form isolated small bodies associated with the amphibolites. Rare dykes of massive amphibolites cross biotite and leptite gneisses of the Prarhodopian Groupe in Eastern, Central and Western Rhodopes. They are thin from 10–20 cm to 1 m, straight or deformed. Dykes of massive amphibolites cross also serpentinites. At the contacts between them the serpentinites become dehydrated and veins of elongated prisms of chrysotile appear. This is thought to be a reaction between the hot magma dyke and the serpentine.

A subintrusive body (300 × 700 m) crosses with intrusive contacts the leptite gneisses of the Bachkovo Formation in the Northern Rhodope anticline—Central Rhodopes and includes xenoliths from the gneisses [9]. It is related to the horizon of epidote amphibolites from the Lukovitsa Formation. The body is built of metagabbro-diabases with massive texture, relict gabbroophitic microstructure and mineral composition: tschermakite amphibole, garnet, andesine, epidote, zoisite, quartz, ilmenite and rutile. The chemical composition corresponds to high-aluminum diabases and gabbro. The body is evidence of the autochthony of the basic magmatism. The higher magnesium character of the metagabbro with respect to amphibolites is illustrated in all petrochemical diagrams. On the diagram in the parameters $SiO_2$ vs. ($Na_2O$ + $K_2O$) gabbro falls in the low alkaline part of the basalt and picrobasalts.

The lack of certainty in the petrochemical definitions of the type of magma and the geodynamic zone of formation, again shows the complex relationship between the genesis of ophiolites and the numerous factors influencing,

The absolute age of the metamorphous basic protolith is determined by U-Pb dating on zircon as Neoproterozoic—610 Ma in eclogites from Central Rhogopes; 678–572 Ma—metagabbro Bubino and 566 Ma—metagabbro Bela Reka [27–30]. These dates coincide with the time of ocean closure preceding the amalgamation of the Gondwana supercontinent.

## 5. Formation of the Ophiolite Association

The overall geological, petrological and geochemical characteristics of the Rhodope Ophiolite Association testify to a complex and long-lasting process of formation. We attempt to trace the development of of the association based on several points of reference:

a) serpentinites are hydrated derivatives of Archaean-Paleoproterozoic mantle peridotites. According to Deschamps et al. [48] a high degree of serpentinization—85%–95% in lizardite and chrysotile is only possible in the ocean basins, where on an ultrabasic ocean plate a layer of clay-like serpentinites up to several kilometers thick is developed;

b) the constant stratigraphic level of serpentinite bodies on a gneiss base on an eroded continental surface indicates tectonic processes of plate movement, the presence of a subduction zone and the transfer of serpentinite fragments on the continent by the mechanism of obduction;

c) Neoproterozoic basic volcanic rocks cover the serpentinite fragments, alternating with the metasediments of the Lukovitsa Variegatet Formation,

We propose a possible scenario for the formation of the Ophiolite Association, which unites in a logical scheme all known geological and theoretical arguments (**Figure 3**):

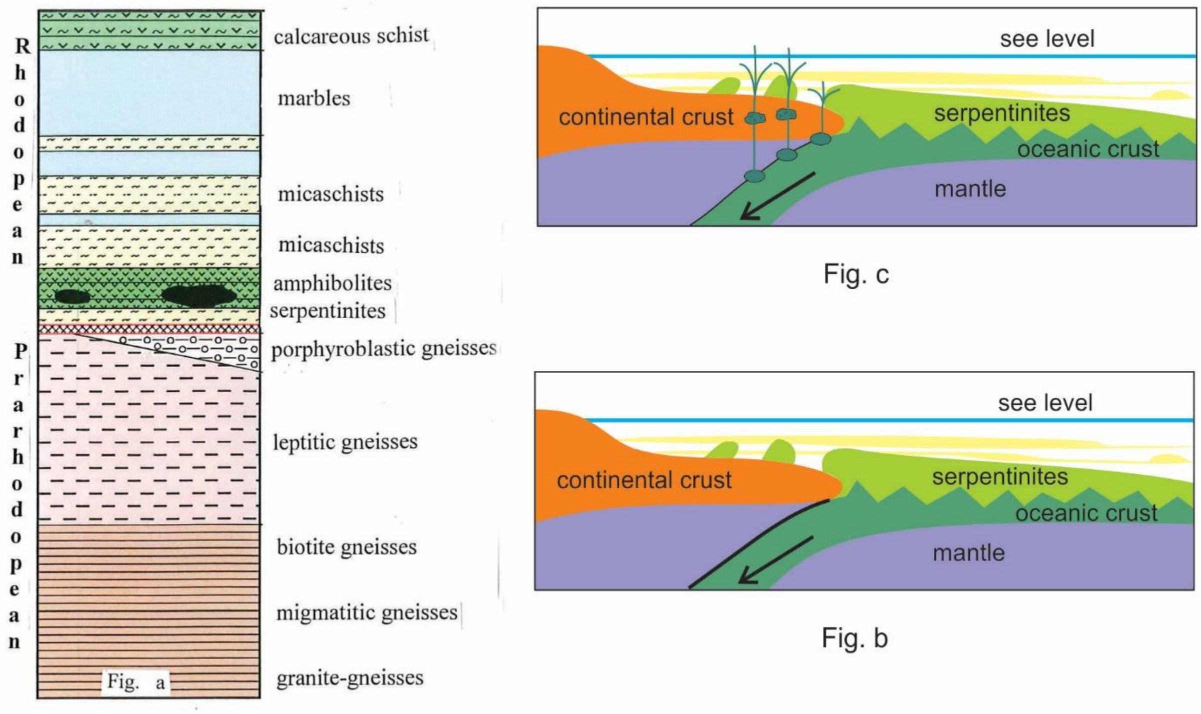


**Figure 3.** Simplified drawing of suprasubducting zone. **(a)** stratigraphic column of the metamorphic complex of the Rhodope Massif. Serpentinite bodies mark an erosional surface and the boundary between the Prarhodope and Rhodope groups. **(b)** subduction of the oceanic plate under the continental one and obduction of serpentinite fragments; **(c)** formation of melt and autochthonous magmatism.

During the Neoproterozoic, a situation of basin closure was created, which caused counter movement and collision between oceanic and continental plates. A microcontinent, the prototype of the Rhodope Massif, built from the gneisses of the Prarhodope Group, collides with an Archean-Paleoproterozoic oceanic mantle plate that is covered by clay-like soft serpentinites. A suprasubduction zone was developed at their convergent boundaries. The serpentinite fragments were scraped off from the serpentinite cover of the oceanic plate by the principle of the grater and obduced on the erosion plane of gneiss continental crust (**Figure 3**). As a result of the strong friction on the contact surface between the huge continental and oceanic plates and the resulting high temperature and pressure at certain depths, foci of molten gneiss and ophiolite rocks appeared. The melt penetrated into the gneisses through channels. Along the way it builds subintrusive bodies and dykes and covered the serpentinites as lavas and tuffs, together with pelitic-carbonate sediments. The location of the large serpentinite massifs is a known indication of proximity to the coastline of the microcontinent where the serpentinite fragments were obducted [49].

The formation of the Rhodope Ophiolite Association had taken place in three stages: a) static—serpentinization of the oceanic ultrabasic plate; b) dynamic—ocean closure, plate tectonic movement and obduction of serpentinite fragments, scraped from the hydrated coat of the sliding ultrabasic plate; c) constructive—autochthonous subintrusive magmatism and volcanism including and covering serpentinite bodies. This determines the heterogeneous nature of formation of the Ophiolite Association—a combination of rock members appearing in different places, times and geological setting.

## 6. Metamorphism of the ophiolites

Three main types of changes are distinguished on the metamorphic complexes in the Rhodope Massif: a) regional metamorphism; b) local high pressure metamorphism (HPM); c) metasomatism. They differ in their spatial, temporal and thermodynamic features and develop in distinctly diverse geological settings.

### 6.1. Regional metamorphism

Regional metamorphism as a broad spatial and comprehensive recrystallization where geothermal gradient and lithostatic (confining) pressure control the TP conditions of crystallization. All ophiolite rocks underwent a regional metamorphism of amphibolite facies: basic volcanic rocks were recrystallized into amphibolites, subintrusive ones—into metagabbros or metadiabases. The large serpentinite bodies were only peripherally metamorphosed in antigorite, talc-chlorite and chlorite-actinolite-tremolite schists, while in the inner parts they retained the lizardite-chrysotile aggregate in mesh cells. Anthophyllite mineralizations are localized mainly in cracks inside the serpentinites and less frequently in their contacts, forming anthophyllite-asbestos cores that were once exploited. In the veins, anthophyllite is associated with talc, tremolite, magnesite, and in rare cases with dolomite. The background regional metamorphism of the rocks is in amphibolite facies: $T$ = 480 °C–560 °C, P = 0.5–0.7 GPa. The preservation of lizardite-chrysotile indicate that the temperature of the general regional metamorphism never exceeded 600 °C. Otherwise, all serpentinites would have become pyroxenites. The latter, being in the relatively dry continental crust, would never be serpentinized again.

### 6.2. High pressure metamorphism (HPM)

High pressure metamorphism (HPM) posses completely opposite characteristics. They appear locally only within the range of shear zones of friction, formed as a consequence of seismotectonic events. While the regional metamorphism is a prolonged state of certain conditions, the HPM is a short living event. Earthquake events cause movement and friction between rock blocks and bedrock layers. The temperature and pressure rapidly rise to high values, causing recrystallization or melting of the zone's wall rocks. The main factor in this metamorphism is friction, which is why we call it *geotribometamorphism.* In petrology HPM is also known as eclogitization—sensu lato which affects different rock varieties, manifested in new mineral paragenesess depending of the chemical composition of the host rock. Typical eclogites, consisting of garnet, omphacie and rutile occur on a basic substrate among amphibolites, while garnet-lherzolites of pyrope, enstatite, olivine, spinel, augite, diopside are formed on serpentinites [50]. Calcifieres are found among marbles as thin (0.5–3 mm) layers, composed of fine-grained: garnet, scapolite, diopside, zoisite, spinel, calcite, dolomite, phlogopite, plagioclase, titanite, quartz. HPM in metapellites are represented by kyanite and phengite schists in some cases with microdiamond-bearing garnet [25,26].

#### 6.2.1. Eclogites

Eclogites are typical representatives of high-baric metamorphic rocks, the genesis of which is still a subject of discussion. Eclogite bodies are found in all metamorphic terrains of South Bulgaria—the Rhodopes, Verila Mt., Sredna Gora Mt. and Ograzhden Mt. [10,12–15,51–55]. Everywhere eclogites are included in formations, analogous in rock composition to the Lukovitsa Variegated Formation. The eclogites associate with the amphibolites and form among them concordant thin layers and lenses up to 10–20 cm as well as rarely compact layers with a thickness of 1–1.5 m. The eclogites often appear on the contact with mica-poor leptite and aplitoid gneisses in geological setting indicating an old friction zone. Eclogites are encountered also in cracks of 2–5 cm, intersecting gabbronorites, which is the most convincing evidence for their formation in friction zones [55]. The crystallization temperatures are most often within the range of 580 °C–680 °C at pressures of 1–1.6 to more seldom 2 GPa. Temperature of 800 °C–1100 °C and pressure of 2–4 GPa are recorded for the coesite and microdiamond containing eclogites [25,54]. All eclogites are affected by alterations [56]. The omphazite is replaced by symplectites of quartz, albite and diopside, garnet—by amphibole, which ultimately leads to complete replacement of eclogites by amphibolites. A characteristic eclogite deposit is known near the village of Kazak, Ivaylovgrad region, Eastern Rhodopes [10,57,58]. There, the eclogites show a layered structure due to the alternation of about 10 cm thin layers of coarse-grained and fine-grained garnet varieties deformed in small folds. All features of the eclogites indicate that they are not exotic bodies, but are an integral part of the Lukovitsa Variegated Formation formed in situ along mobile zones on lithological contacts and shear zones of friction.

#### 6.2.2. Banded eclogizited serpentinites

A rare case of rhythmic banded eclogitization is observed on a serpentinite body south of Avren village, Krumovgrad district—Eastern Rhodopes [50]. The body is a part of the Lukovitsa Variegated Formation which is related with the Kimi complex in North Greece where microdiamonds in garnet are found [24]. The body's peripheral parts (30–40 m) are affected by eclogitization. Bands of garnet lherzolites (1–20 mm), which are parallel to the contact, alternate with strips of unchanged serpentinite. The lherzolite bands close to the contact are more frequent and consist of pyrope-garnet diopside, enstatite, olivine and spinel, crystallized under conditions of *T* = 560 °C–820 °C, *P* = 8–15 kbar. Towards the interior of the serpentinite body the bands become rarer, do not contain garnet and gradually disappear. Obviously eclogitization is associated with the contact between the serpentinites and host gneisses, which has been most probably a previous paleoseismotectonic zone.

#### 6.2.3. Calcifires

Calcifiers are observed in many places among marbles as thin (0.5–3 mm) layers, composed of fine-grained: garnet, scapolite, diopside, olivine, spinel, calcite, dolomite, phlogopite, plagioclase, titanite, quartz. The calculated crystallization conditions are: *T* = 745 °C–770 °C, *P* = 0.5–1 GPa [58].

#### 6.2.4. Kyanite and fengite schists

Kyanite and fengite schists are found also in zones of interlaminar sliding. Microdiamond-containing garnet porphyroblasts in gneiss schists of the Variegated Formation are encountered in the Chepelare region of the Central Rhodopes; *T* = 700 °C–800 °C, *P* = 3.5–4.6 GPa [25,26].

High-thermobaric rocks are also an important indicator of the setting when they occur in friction zones. Since friction zones are seismic zones, this means that HPM rocks document paleoseismic events and the age of the eclogites fixes the age of the seismic events in our earth’s past.

### 6.3. Metasomatism

Metasomatism is a process of bulk chemical change in which deep derivatives from anathectic and granitoid magmas, as pegmatite-aplite veins, penetrate the regional metamorphic rocks, enriching them with Si, Al, K, Na. Metasomatic pegmatite-aplite pulses have occurred repeatedly during Proterozoic and Phanerozoic times of granitic magmatism, respectively. Ophiolites are strongly affected by metasomatism due to the contrasting chemistry between them and pegmatite-aplites resulting in hybrid rocks such as metasomatic gabbroides [56] and gabbro-norites with a corona-structure are created [18,59]. The genesis of the gabbro-norites with corona-structure is still a matter of debate. In our opinion, the most plausible version is the metasomatic one. It is likely that small serpentinite bodies, included in the amphibolite layer, reacted with the quartz-feldspar mineral composition of the surrounding migmatization environment. However, the metasomatic version is supported by findings of serpentinite inclusions in pegmatite veins in Ograzhden metamorphic rocks that show the same corona structure during recrystallization [59].

## 7. Discusion

The Rhodope Ophiolite Association is a highly informative formation in the Rhodope Massif. It is presented in a lot of instructive outcrops that allow us to consider and resolve some controversial questions regarding stratigraphy, tectonics, high thermobaric tribometamorphism and metasomatic hybridization.

One of the most hotly debated issues is the construction of the metamorphic complex in the Rhodope Massif and the place of ophiolites in it. To the established concept introduced in 1963 by Vergilov et al. [32], which presents the metamorphic complex as a single unified stratigraphic system, Bürg et al. [35–37] opposes a new version considering the metamorphic complex as a system of discordant plates "pile of thrust", based on observed somewhere mylonitized gneisses and microstructures that showed shear of sence deformation. The factual database of the two concepts is unmatched in weight and importance. The stratigraphic concept is substantiated by a huge database of geological mapping of the entire massif at a scale of 1:25,000, shown in numerous geological maps, stratigraphic profiles and correlation analyses of different parts of the terrain, which unequivocally confirm a normal stratigraphic sequence, underwent a general plastic deformation resulting in folded structures [32]. The opponents have so far not presented graphic material where the thrust plates in

question, being actual physical entities, must be delineated with clear boundaries and areal coverage. Then such material could be accepted as evidence for existence of the mentioned thrusts. In recent years, the rhetoric about the thrusts has evolved and there is already talk of undefined allochthons, shown on tectonic sketches of various shapes [60–62] In fact, the authors continue Bürg's idea about the tectonic structure of the Rhodope Massif. However, the arguments advanced by them do not correspond to the established geological facts about the relationships between the lithological units, which unequivocally show a lithostratigraphic sequence and normal rather than tectonic contacts.

The stable stratigraphic position of the Ophiolite Association also confirms the concept of a single stratigraphic sequence of the metamorphic complex and refutes the ideas of thrusting [35–37]. The Ophiolites have a definite and permanent stratigraphic position in the metamorphic complex and do not delineate suture zones anywhere. Also, meso- and microdeformations in the metamorphic rocks, marked as shear in cense criteria, cannot be accepted as evidence of Alpine nappe complex, because similar deformation microstructures can also appear in every fold structures.

The well preserved and uniform for the whole massif stratigraphic sequence and the dominating fold structure disprove the existence of thrust structures and suture zones in the outcropped part of the metamorphic basement on Bulgarian territory.

The statement that eclogites and ophiolites "are found in various units of crustal-scale duplex structure" and at the same time "these rocks delineate a suture zone" between two units as Burg [37] considers is factually unconfirmed and contradictory. The Ophiolites have a definite and permanent stratigraphic position in the metamorphic complex and do not delineate suture zones anywhere. Eclogites themselves form in friction seismic zones at high temperatures and pressures. In Rhodope Mountain, they mark old paleoseismic zones, among the amphibolites of the Ophiolite Association Lukovitsa Formation Ophiolite Association of the Rhodope Group and therefore they can by no means be accepted as markers of tectonic boundaries of thrust plate. We also believe that the Paleoproterozoic age definitions of the Dobromirtsi serpentinites can be applied to all serpentinites, due to their constant stratigraphic and geological position. This disproves the notion of their different age of implantation in the Rhodope metamorphic complex [63].

Determining the geochemical nature of ophiolites as well as the geodynamic zone of their formation directly from chemical analyzes is, in our opinion, an incorrect approach that carries the risk of errors. Very often highly hydrated ophiolites are not called by their current rock name "serpentinites", but the indefinite "ultramafites", "ultrabazites" or even simply "dunites" and "peridotites", calculated by the ratio of the components. It ignores the well-known fact that serpentinization extracts calcium and iron, which greatly increases the magnesianity and directs the interpretation to dunites. However, replacing the name of serpentinite, which is a magnesian clay formed in a water basin, with the name of an igneous rock - dunite or peridotite, radically changes the interpretation of the composition, genesis and development of an ophiolite association.

Analogous mistakes are made when interpreting the primary nature of amphibolites and metagabbras. When it comes to a continental Ophiolite Association created in a suprasubduction setting, such as the Rhodope ophiolites considered here, the composition of the autochthonous magmatism is to varying degrees contaminated. In the frictional surface of the subduction zone, both the ultrabasic rocks of the subducting oceanic mantle plate and the continental gneisses melt. Therefore, middle basic rocks sometimes bear geochemical signatures of a mantle signature. Very often, researchers overrely on geochemical data and, recalculating them using various formulas, coefficients and diagrams without comparing them to the specific geological situation, arrive at contradictory and unrealistic genetic interpretations.

An eloquent example of a geochemical study of three small metagabbro bodies in the Eastern Rhodopes is presented by Haydoutov et al. [21]. Petrochemical work is filled with a great diligence. Numerous recalculations of chemical analyzes and diagrams by different methods were made, as many ratios as possible between the components were deduced and compared with examples from recent basic volcanics from various distant regions of the planet, however, in the complete absence of consideration of the geological position and stratigraphic location of the studied samples. In the end, the authors came to the controversial conclusion that the rocks are boninites and island arc tholeiites, which together with metasediments form an ensimatic island arc. But at the same time the ultramafic rocks (as they call the serpentinites) were serpentinised in a supra-subduction zone (???) and may have had a genetic connection to the aforementioned ensimatic island arc. The mentioned article is an example of perfectly conducted geochemical calculations, which, however, without a solid geological base and terminological accuracy lose their interpretative value.

## 8. Conclusion

- The Rhodope Ophiolite Association is a continental heterogeneous serpentinite-basite supra-subduction rock formation. It is part of the Precambrian metamorphic complex of the Rhodope Massif on the Balkan Peninsula, Southeastern Europe. The association occupies the lower stratigraphic levels of the Lukovitsa Formation of the Rhodope Group of the metamorphic complex;
- The Ophiolite Association unites allochthonous serpentinites and autochthonous basic magmatic rocks, which are formed in different places, time and geological setting over a long period of time. The beginning of its creation takes place in a deep-sea ocean environment where a Paleoproterozoic mantle ultrabasic plate is subjected to prolonged serpentinization;

- Serpentinite fragments were obducted on the erosional surface of an old continent. This event marks an epoch of tectonic activity of ocean closure, countermovement and collision of continental and oceanic plates and formation of a subduction zone along the convergent boundaries of the two plates;
- Friction in the subduction zone produced autochthonous basic subintrusive and volcanic magmatism during the Neoproterozoic (700–550 Ma) concurrent with transgression and deposition of pelitic-carbonate sediments. The uneven distribution of the large serpentinite bodies gives an indication of their distance from the shoreline;
- The well-preserved stratigraphic sequence mainly the fold tectonic structure and the absence of regional thrusts, testifies that the subsequent development of the metamorphic terrain was relatively tectonically calm with a probable softer relief. Widespread regional amphibolite facies metamorphism, with no apparent zonation, also supports the view of a relatively more relaxed tectonic regime;
- It is assumed that deep slides in the roots of the Rhodope Massif provoked the formation of magmatic centers and granodiorite batholiths. Their derivatives such as pegmatite-aplite veins penetrate the higher levels and metasomatize the rocks. The metasomatic changes are clearly visible in the ophiolites, due to the contrasting chemical composition between them and the pegmatite-aplite granite derivatives;
- The metamorphic complex during the Precambrian and Phanerozoic was repeatedly cut by seismic zones of strong friction between rock blocks and layers marked by high-thermobaric rocks: eclogites, garnet lherzolites, calcifiers, kyanite and fengite schists;
- The Rhodope Massif was included in the Caledonian-Hercynian and Alpine mobile belts, but it has relatively well preserved its primary lithostratigraphic sequence, which is an indication of its consolidation. Only during the Paleogene did significant disintegrations occur and it was divided into three parts: Western, Central and Eastern Rhodopes.

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