

Article

A new occurrence of terrestrial native iron in the earth's surface: The Ilia thermogenic travertine case, northwestern Euboea, Greece

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Abstract: Native iron has been identified in an active thermogenic travertine deposit, located at Ilia area (Euboea Island, Greece). The deposit is forming around a hot spring, which is part of a large active metallogenetic hydrothermal system depositing ore-bearing travertines. The native iron occurs in two shapes: nodules with diameter 0.4 and 0.45 cm and angular grains, with length up to tens of μm . The travertine laminae around the spherical/ovoid nodules grow smoothly around the nodules; the angular grains are trapped inside the pores of the travertine. Their mineral-chemistry is ultra-pure, containing, other than Fe, only Mn (0.34–0.38 wt%) and Ni (≤ 0.05 wt%). After evaluating all the possible environments where native iron has been reported up today, we support the hypothesis that the native iron at Ilia is remobilized from ophiolitic rocks, originated initially from reducing fluids, which deposited it in the past during serpentinization processes and now it is remobilized and transferred from the hydrothermal fluid and syngenetically enclosed as clastic grains within the active thermogenic travertine. The native iron mineral-chemistry, morphology and the presence of other mineral phase at the same thermogenic travertine, which have been documented in ophiolitic rocks at which native iron is present, support this hypothesis.

Keywords: Native iron; hydrothermal system; thermogenic travertine; serpentinization processes

1. Introduction

The occurrence of native elements as authigenic mineral phases in nature is an extremely rare phenomenon. Examples of elements present in native (zero-valence) state include among others, Al [1,2], Si [3], Ni [4], Cu and Ti [5]. Native iron is unstable under the oxygen fugacity conditions prevailing earth's crust and mantle, with a few exceptions [6].

Iron is one of the most abundant elements in the Universe and in our planet. It is the 6th most abundant chemical element in the Universe and the 4th in earth's crust, which it is made up of about 5% iron [7]. With the until now available data, the inner core of the Earth is believed to be a solid iron-nickel alloy. Iron belongs to the eighth group of the periodic table, with atomic number 26 and occurs in a wide range of oxidation states, -2 to $+6$, with $+2$ and $+3$ to be the most common. It is a very

reactive element and oxidizes very easily. It is strongly magnetic and melts at 1535 °C [8]. Iron is found in many minerals as main or trace compound. Most of it, is found combined with oxygen forming Fe-oxide minerals, such as hematite (α -Fe₂O₃) and magnetite (Fe₃O₄).

Iron could occur as native element in reducing environments, but is highly reactive in the presence of oxygen and water. For that reason, most of the native iron on earth is extraterrestrial (meteoritic iron) and it mainly comes from iron-nickel meteorites, which are preserved by the vacuum of space prior to falling on earth. Metallic meteorites are composed mainly of Fe-Ni alloys like taenite and kamacite and contain small quantities of other impurities such as cobalt, manganese, tin, chromium, sulfur, carbon, chlorine, copper and phosphorus.

However, there are some specific earth environments around the world where terrestrial native iron (telluric) can be found, mainly in igneous rocks [9]. More specifically, terrestrial native iron has been found in basalts, ultramafic members of ophiolitic complexes, kimberlite-diamonds and in hydrothermal systems. Except from serpentinization, in all other cases the terrestrial native iron is a product of a reduced deep mantle environment.

Regarding the presence of native iron in basalts, the most well-known and documented occurrences are from the basalt at Ovifak, Disko Island, Greenland [10-13], from the Bühl basalt, Kessel, Germany [14,15] and from the Khungtukun intrusion, Maimecha-Kotui magmatic province in northern Siberia [16-22]. The most prevailing theory, based on geochemistry of highly siderophile elements and Re-Os isotopic systematics, favours the formation of such occurrences by interaction of basaltic magma with carbon-rich sediments which effect in the generation of low oxygen fugacities (fO₂) during magmatic crystallization, resulting in the precipitation of native-iron, (e.g. [23] and references therein).

Additionally, native iron has been found in ophiolitic rocks such as serpentinized dunites, peridotites, serpentinite and chromitites. The best known and documented occurrences are from the Muskox, Canada [24], the Mina do Abessedo, Portugal [25], the Shaman, Baikal [26], Kachkanar, Ural [27], the Vetrenii belt, Baltic shield [28], the Josephine, U.S.A. [29], the Sanbagawa belt, central Japan [30], the Maqсад ophiolite, Oman [31], the Balangero, Italy [32], the Luobusa ophiolite, southern Tibet [33,34,3], the Ray-Iz ophiolite, Polar Urals [35] and from the Skyros island, Greece [36].

Native iron has been identified in kimberlites and kimberlite diamonds, emphasizing its usual reduced deep mantle origin [37]. Occurrence have been reported from the Yakutia kimberlite [38], the Sloan kimberlitic diamond [39], from the Rio Soriso in association with low-Ni pyrrhotite [40] and the Pandrea-1 pipe in association with ferropericase and chrome spinel [41], both from the Juina area, Brazil, and from the placer Kankan in Guinea [42].

In geothermal systems, the presence of native iron has been reported only from the Kuril Islands, Russia. Spherical nodules of native iron have been found in a deep geothermal drill [43,44] and [45] reported the occurrence of native iron in active fumaroles at the Kudryavy volcano, Iturup Island (group of Kuril Islands).

The native iron on earth is actually not totally pure, but usually contains noticeable percentage of some other elements, mainly Ni (2.44 – 0.03 wt.% Ni; [46]). The iron-nickel alloy grains have been described to have either an extra-terrestrial [47] or terrestrial-mantle origin [33]. In addition to nickel, the terrestrial iron occurrences also include other elements such as As, Co, Cu, Mn (see [30,32,37,35,48]) and the extra-terrestrial occurrences may incorporate elements such as Cr, Co, Mn, Ni, Si (see [33]).

This paper reports for the first time on the presence of an exceptional rare occurrence of ultra-pure native iron (Ni ≤ 0.05 wt.%) from Fe-As-rich thermogenic travertines in Ilia area, Euboea Island, Greece. The aim of this study is to describe the composition and identify the origin of native iron within the Ilia travertines, on the basis of multi-analytical mineralogical and geochemical studies and comparisons with other occurrences of native iron elsewhere.

2. Geological setting

Ilia area is located in the northwestern part of Euboea Island, Greece (Figure 1). The study area is located at the western extremity of the North Anatolian Fault and in a back-arc position with respect to the South Aegean Active Volcanic Arc [48-50]. It is one of the most neo-tectonically active areas in Greece dominated by extensional tectonics similarly to the rest of the Aegean Sea.

The northwestern Euboea island comprises of a lower series of Permian–Triassic volcanoclastic rocks overlying a pre-middle to middle Carboniferous metamorphic basement, which are in turn overlain by middle Triassic shallow marine clastic and carbonate rocks intercalated with volcanic rocks best developed at the southeast part of Aedipsos town [51,52]. On top of this sequence, Jurassic limestones and Late Jurassic– Early Cretaceous ophiolites occur (mainly metabasites and serpentinites, 51-53 Figure 1). The Palaeozoic and Mesozoic sequences were folded or imbricated as the result of two main tectonic events (Alpine and Eo-Alpine). In the broad area of northern Euboea, lignite layers have been identified as intercalations within Neogene–Lower Pleistocene lake sediments [54].

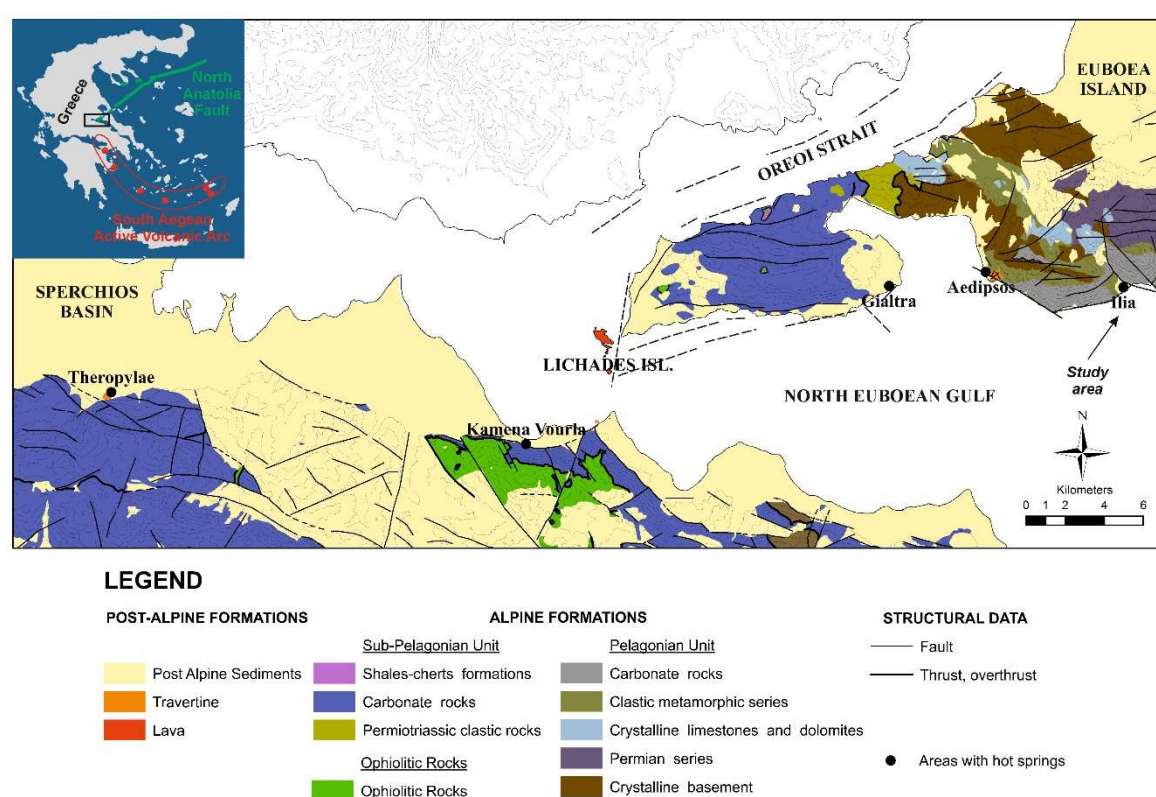


Figure 1. Simplified geological map of Northern Euboea island and neighboring area of Sperchios (mainland; modified from [63]).

The central part of the Euboean Gulf is occupied by the Plio-Pleistocene volcanic center of Lichades Islands [55,48], and comprises 0.5 Ma (K-Ar, [56]) trachyandesitic lava flows. Magma emplacement took place along major tectonic structures in the area. [57] and [58] showed that there is a magma chamber at depths 7-8 km, under the North Euboean Gulf. [59] based on Sr-Nd-Pb isotopic data, related this volcanic center with the large volcanic belt that developed north of the Pelagonian–Attic–Cycladic–Menderes massifs, encompassing a 35 Ma timespan which is widespread over a large area from NW Greece–Macedonia to the Aegean–western Anatolia. According to the above authors, the Lichades volcanic products are orogenic in character and partially contemporaneous with the south Aegean active volcanic arc, but with different geochemical features, related to distinct magma sources (e.g. lithospheric mantle wedge and a depleted asthenospheric mantle wedge north and south of the Pelagonian–Attic–Cycladic–Menderes massifs respectively).

At the northwestern part of Euboea Island and the neighboring area of Sperchios (mainland), several hot springs occur (Fig. 1), considered to be part of a single hydrothermal system, which is controlled by active tectonics, and supplied with heat by the deep magma chamber [60]. Euboea's hot springs are characterized by a mixture of seawater and deep magmatic fluid with only limited meteoric water contribution [60]. Another salient feature of them, is the dominance of thermogenic travertine deposition [62-64]. [60] described active metallogenetic processes at the system leading to ore-bearing travertines including Ilia's travertine.

In Ilia area, an artesian borehole cross-cutting metamorphic rocks [65], deposit iron and arsenic rich travertine on surface ([63, 60], Figure 2A-C). Ilia is located on a marked bend of the marginal fault system, where the average strike changes from SE-NW to E-W (or ENE-WSW). That deep fault system is being used for the circulation of the hydrothermal fluid.

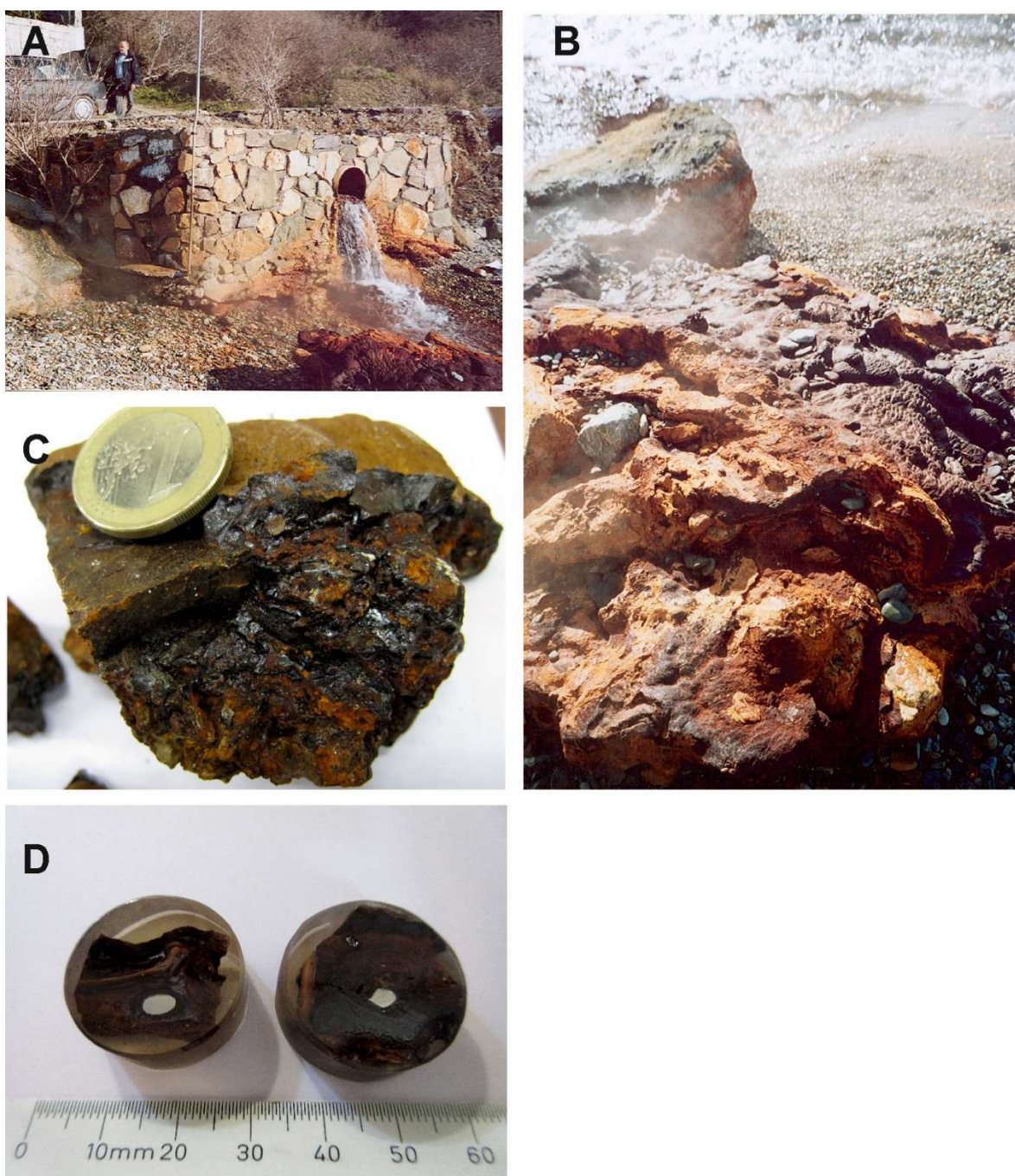


Figure 2. Iron-rich thermogenic travertines from Ilia: (A) Ilia hot spring. The Fe-rich thermogenic travertine deposits at the point where the pipe from the borehole vents the hot water. Local authorities remove the depositions, in order to avoid clogging of the spring; (B) Laminated Fe-rich

travertine from Ilia; (C) Cross section at Fe-rich botryoid laminated travertine; (D) Two polished sections including two different iron nodules and the hosting travertine.

3. Materials and Methods

Two polished sections including metallic grains and the host travertine were firstly studied under stereomicroscope and optical microscope. SEM-EDS analysis was carried out using a Jeol JSM 5600 SEM instrument, equipped with an Oxford ISIS 300 micro analytical device, at the National and Kapodistrian University of Athens. At the Natural History Museum of London further analyses and mapping was performed with a JEOL 5900LV, equipped with an Energy Dispersive X-ray (EDX) and a Wavelength Dispersive X-ray (WDX). Electron microprobe analyses (EMPA) were carried out using the Jeol JXA 8200 Superprobe WD/ED combined microanalyzer at the University of Lausanne. Operating conditions were accelerating voltage of 15 kV, beam current of 20 nA, and beam diameter of 1 μm . Counting times of 20 s on peak and 10 s on background on both sides of the peak were used for all elements. Overlap and matrix corrections were carried out following the established protocol and software in the laboratory.

In addition, spot XRD analysis was done with a Micro-X-ray diffraction, with an Inel Curved Position Sensitive Detector (PSD) and a high brightness X-ray source, at the Natural History Museum of London.

4. Results

Several metallic grains of native iron were identified in the iron-rich thermogenic travertine deposit from Ilia area, northwestern Euboea Island (Greece). The biggest grains of native iron are two spherical nodules visible even with naked eye and whose diameters are 0.4 and 0.45 cm respectively (Fig. 2d, 3a-c). Smaller grains, with angular shape (from few μm up to tens of μm , Fig. 3D) were also identified and studied.

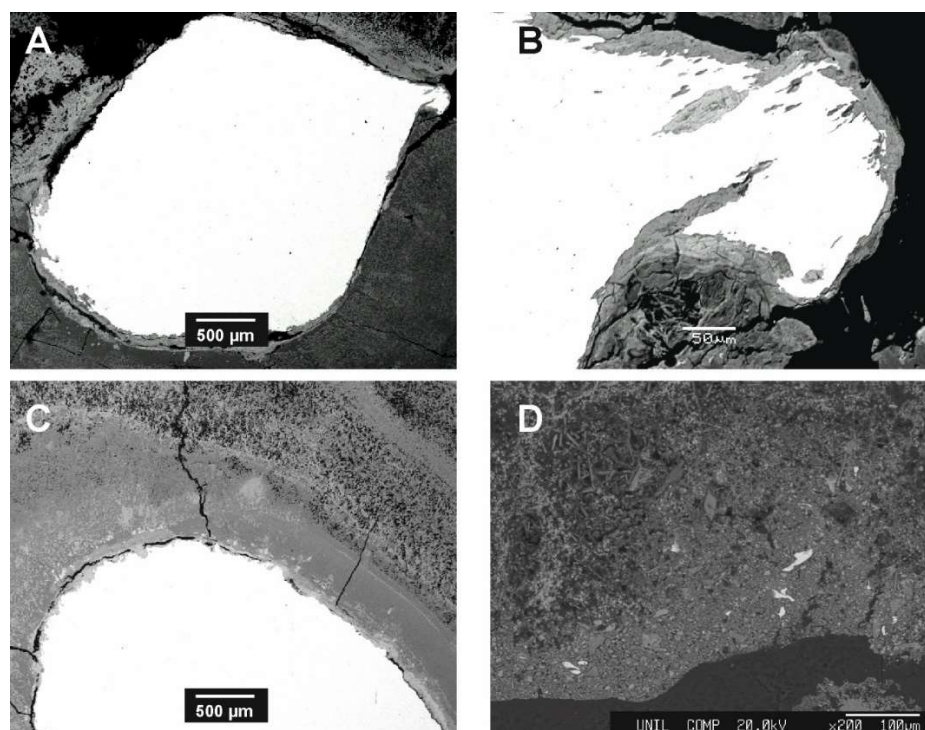


Figure 3. SEM photomicrographs: (a) Native iron nodule; (b) Oxidations were identified only at the external surface of the grains, as a result of their exposure to atmospheric conditions and water during their transfer and until their entrapment inside the travertine; (c) The travertine laminae around the nodules are not interrupted, but were developed around the nodules, suggesting that the nodules were placed simultaneously with the creation of the travertine; (d) Several smaller angular pieces of native iron were found inside the travertine (from few μm up to tens of μm).

The travertine laminae around the two spherical/ovoid nodules are not interrupted, instead they are developed smoothly around them (Fig. 3c), suggesting that the nodules were placed simultaneously with the deposition of the travertine. The smaller grains (up to few μm) are located inside the pores of the travertine (Fig. 2d) in common to other metallic mineral phases like pyrite, arsenopyrite, galena, sphalerite which are considered to be products of mechanical transport from deeper levels through the hydrothermal system [60].

The two biggest nodules of native iron are almost spherical with smooth outlines, steel gray in color, with metallic luster under the stereomicroscope and white, isotropic and with very high reflectivity under the ore microscope. Spot XRD analysis was performed to one of the nodules. The XRD pattern showed diffraction lines attributable to $\alpha\text{-Fe}$ (JCPDS File No. 6-696; Fig. 4).

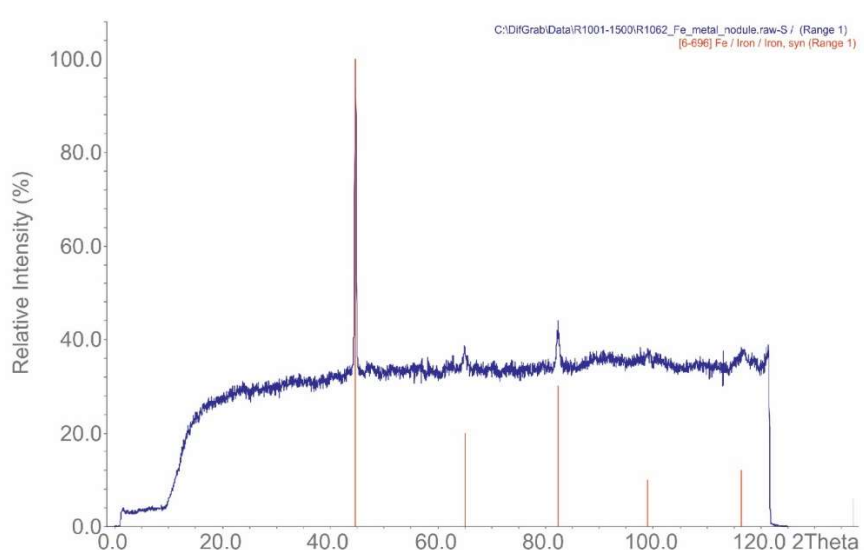


Figure 4. XRD pattern from spot XRD analysis performed to the center of one of the nodules. Diffraction lines attributable to $\alpha\text{-Fe}$ (JCPDS File No. 6-696).

Analyses with SEM-EDS, -WDX and EMPA performed to both nodules and to several smaller angular grains (Table 1) indicate a high purity of the grains. Except Fe only a very low participation of Mn (0.34-0.38 wt%) and Ni (≤ 0.05 wt%) was found. Microprobe analytical profiles from cores to the rims of both nodules (Table 1), as well as element mapping, revealed their chemical homogeneity (Figure 5). No chemical zonation or differentiation was found. Oxidation at the external surface of the grains was most probably the result of their exposure to atmospheric conditions and water during their transfer and until their entrapment inside the travertine (Fig. 5).

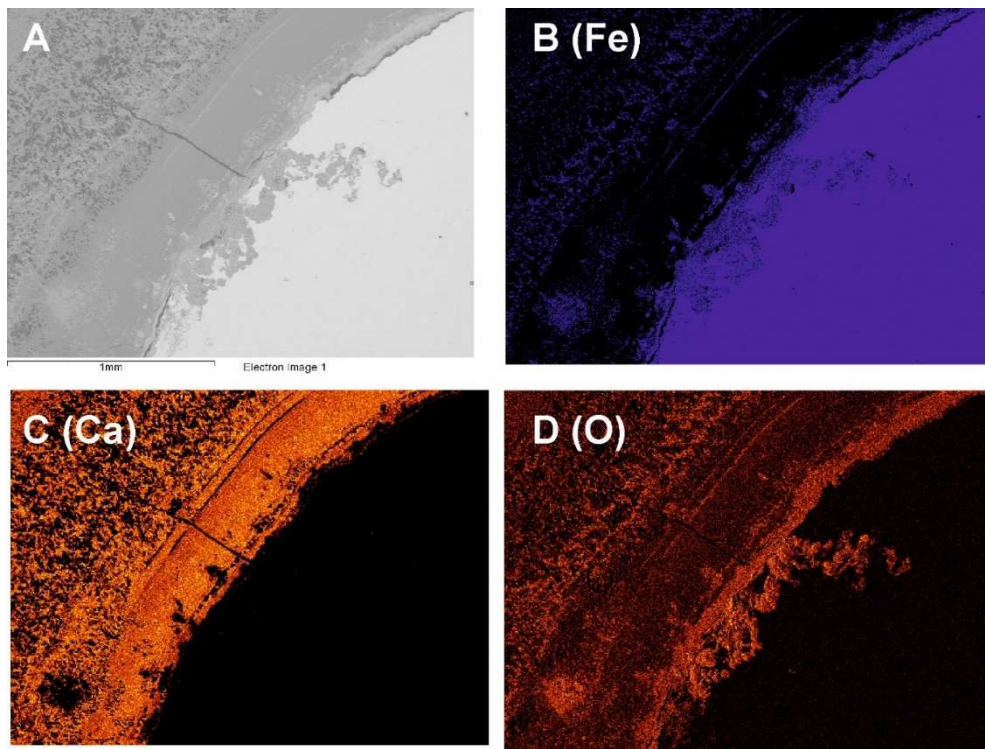


Figure 5. (a) back-scattered electron image (BSEI) of the contact between iron nodule and the Fe-rich laminated travertine; (b-d) false color BSEI results of mapping, displaying the distribution of Fe (purple), C (orange) and O (red).

Table 1. Representative EMPA microanalysis of native iron (in element wt%).

	1	2	3	4	5	6
Fe	101.18	101.04	98.45	98.66	98.79	98.67
S	bd	bd	bd	bd	bd	bd
Co	bd	bd	na	na	na	na
Ni	0.05	0.05	na	na	na	na
Cu	bd	bd	bd	bd	bd	bd
Zn	bd	bd	bd	bd	bd	bd
As	bd	bd	na	na	na	na
Se	bd	bd	bd	bd	bd	bd
Ag	bd	bd	bd	bd	bd	bd
Sb	bd	bd	na	na	na	na
Te	bd	bd	na	na	na	na
Au	bd	bd	na	na	na	na
Pb	bd	bd	na	na	na	na
Bi	bd	bd	na	na	na	na
Mn	na	na	0.38	0.35	0.38	0.34
Cd	na	na	bd	bd	bd	bd
Ga	na	na	bd	bd	bd	bd
In	na	na	bd	bd	bd	bd
Total	101.23	101.09	98.84	99.01	99.17	99.01

bd = below detection limit, na = not analysed.

5. Discussion

Ilia's hot spring is part of a large geothermal system, with hot springs manifestations in northwestern Euboea Island and the neighboring part of the mainland in eastern Central Greece, i.e. Sperchios area [60], recognized as having one of the highest geothermal gradients in Greece, after the

South Aegean Active Volcanic Arc [66]. The hydrothermal system is controlled by active tectonics, and supplied with heat by a 7-8 km deep magma chamber, with surface manifestation, the Plio-Pleistocene trachyandesitic volcanic center of Lichades. The hydrothermal fluid is a mixture of seawater and deep magmatic fluid with only limited meteoric water contribution in Euboea part [60]. Geologically, the broad area of northwestern Euboea also includes Late Jurassic- Early Cretaceous ophiolitic rocks, mainly metabasites and serpentinites ([51-53]; Fig. 1) and Neogene-Lower Pleistocene lignite lake sediments layers [54], which may be of importance for the genesis of the native iron occurrences (see below).

The hot spring of Ilia is enriched in iron (5-13.3 mg/L) compared to the other hot springs of the hydrothermal system [60]. The spring displays a water temperature in the range from 61 to 63.7 °C and pH from 5.9 to 6.4 [60] and is degassing mainly CO₂ [67]. [60] have presented several geochemical and mineralogical evidence supporting the magmatic contribution to the hydrothermal system.

Accordingly, Ilia's thermogenic travertine has extreme enrichment in iron (up to 28.8 %), arsenic (up to 1.8 %) and REE compared to the others thermogenic travertines around the world [62,60]. The main mineral phases are an amorphous hydrous ferric oxyhydroxide phase, identified as ferrihydrite, aragonite and calcite [63,60], creating a laminated to banded iron-rich travertine by biotic and abiotic processes (Fig. 2c; [61]). Except from native iron, previous studies [62,60] have identified and documented the presence of several mineral phases including sulfides such as pyrite, arsenopyrite, galena, chalcopyrite, sphalerite and stibnite, native elements such as Pb and Ni, alloys such as Ni-Co-Cu-Sn, Au±Cu-Ag and other mineral phases such as awaruite, fluorite and REE-bearing phases, which are syngenetically enclosed as clastic grains within the pores of travertine.

The shape and mineral chemistry of the studied native iron grains suggest that they did not formed in situ, but they are products of a deeper environment and they were transferred by the hydrothermal fluid, as [60] suggested for most of the metallic minerals enclosed in the travertines of the system.

The question here is, what is the origin of that ultra-pure native iron grains of Ilia? We examine here the following scenarios for the native iron formation: (a) meteoritic, (b) anthropogenic contamination, (c) kimberlite- and diamond-related, (d) magmatic immiscibility in basaltic rocks (e) magmatic-hydrothermal formation in active geothermal fields (f) in chromitites and ophiolitic rocks (e.g. serpentinization).

(a) There is no evidence for a meteorite fall in the area and most importantly the very low Ni concentration (≤ 0.05 wt. %) in Ilia's native iron, lead to the exclusion of the scenario of extraterrestrial origin.

(b) Anthropogenic contamination is always a possibility, but we strongly believe this is not the case for Ilia. The two main nodules with diameter 0.4 and 0.45 cm were identified during the cutting process of the samples. A series of several samples were created from the same rock formation and several additional from other thermogenic travertines, but no such grains were identified in other samples from Ilia's travertines and other travertines from other locations in the system. All the samples were processed in exactly the same way. In addition, a variety of other native metals (Pb, Ni, Cu) and alloys (Ni-Co-Cu-Sn, Au±Cu-Ag) were found in other samples from Ilia and travertines from other locations, belonging to the Euboea-Sperchios hydrothermal system [62,60] demonstrating the occurrence of in-situ metal formation.

(c) Native iron has been reported in kimberlites and diamonds. [68] suggested that local and transient reducing conditions microenvironment results from the production of hydrogen when diamond grows from a methane-dominated fluid [69]. But, the absence of kimberlites and diamond in the greater area, their incompatibility with the geological setting of the area, exclude that scenario also. Additionally, [37] showed that the kimberlite- and diamond-related native iron grains, present complex and variable compositions including Cr (1.13–2.37 wt.% Cr₂O₃), as well as Ni (0.28–0.40 wt.% NiO), Mg (0.16–0.94 wt.% MgO), Mn (0.17– 0.47 wt.% MnO), Ti (0.02–0.12 wt.% TiO₂) and Al (0.09–0.27 wt.% Al₂O₃).

(d) Some of the most well-known and documented occurrences of native iron are in basalts and more specifically in three basalt fields around the world (Bühl basalt, Kessel, Germany, Disko Island, Greenland and Khungtukun intrusion, Maimecha-Kotui magmatic province in northern Siberian, Russia). In all cases, the occurrences of reduced Fe have been attributed to basalt magmas intrusion into carbon-bearing sedimentary rocks (graphite, coal, shale, [70,19,48]). According to [21] the creation of native iron is due to magmatic immiscibility (i.e. unmixing of melt phases), which begins when a melt reaches a composition where its Gibbs free energy is higher than that of paired melts whose compositions equal the precursor homogeneous melt. Native iron in Khungtukun (Russia) occurs as disseminated individual spherical nodules throughout the basaltic rocks, having ovoid and spherical shaped inclusions in the silicate minerals. According to [21] that specific textural i.e. drop- and bleb-like shape is an evidence which strongly indicates that native Fe crystallized from a metallic liquid at the temperature of the basaltic liquid, which is below the solidus of pure iron (1538 °C). Carbon, provided by assimilated host local coal-bearing sedimentary rocks, played the role of the main fluxing and reducing agent reducing the magma-derived iron and depressed the liquids temperature. In agreement to [21], also [48] suggested that primary native Fe is a rare crystallizing phase from terrestrial basaltic magmas, requiring highly reducing conditions in order to form. Reducing conditions in basaltic magmas can be achieved through assimilation of carbonaceous crustal material, such as coal beds or carbonaceous black shales, which leads to formation of an immiscible, molten, C-rich, native Fe alloy liquid in all known occurrences of native iron in basalts. Also, if this liquid also contains sufficient sulfur, it can undergo further division into conjugate Fe–C-rich and Fe–S-rich immiscible melts that can effectively scavenge highly siderophile elements such as Au, Ni and Cu. It should be emphasized here that similar elements are also found in Ilia travertine as transferred grains from deep-seated sources [60].

Also, the lignite layers found in the area [54] could play the role of the fluxing and reducing agent of the magma-derived iron and depressed the liquids temperature. However, available data (no presence of basaltic rocks) so far, do not support a hypothesis of formation of the Ilia native iron blebs by magma immiscibility at depth, that it is later transport via hydrothermal processes to the surface.

Additionally, electron microprobe analyses of native iron from basalt at Ovifak, Disko Island, Greenland, Bühl basalt, Kessel, Germany and Khungtukun intrusion, Maimecha-Kotui magmatic province in northern Siberia revealed the presence of Co (0.03–0.7 wt.%), Ni (0.2–2.5 wt.%) and Cu (0.02–0.65 wt.%) [48]. This strongly contrasts to the composition of Ilia native iron grains that contain only Mn (0.34–0.38 wt.%) and Ni up to 0.05 wt.% and concentrations of Cu and Co below detection limit, weakening even further the scenario of iron blebs by magma immiscibility, in our case.

(e) In active geothermal systems, native iron has only been found in Kuril Islands, Russia in two environments: Firstly, at 1.5 km depth within a geothermal drillhole, just above the magmatic chamber, where spherical nodules of native iron are interpreted to have been formed by injection into metasomatites of a “dry” recovered fluid having a temperature > 500–600°C and originated from a depth ≥1.5–2km (zone of hydrothermal circulation, [43,44]). Secondly, in the system of the Kuril Islands (volcano Kudryavy, Iturup Island) native iron grains were also reported at surface active fumaroles [45]. [45] suggested that the native iron was formed at great depth under very low fO_2 conditions, where Fe is present in native state (Fe^0), and came to the surface as a result of gas transport. In both, cases the native iron grains were products of a deep origin and were transferred by the hydrothermal fluid (inc. gas and liquid phases).

(f) Finally, native iron has been documented in ophiolitic rocks like serpentinitized dunites, peridotites, serpentinite and in associated chromitites, in several cases such as Josephine, U.S.A [29], Sanbagawa belt, central Japan [30], Maqсад ophiolite, Oman [31], Balangero area, Italy [32], Luobusa ophiolite, southern Tibet [33,34,3], Ray-Iz ophiolite, Polar Urals [35] and the neighboring to study area, Skyros island, Greece [36].

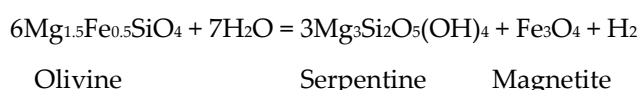
In Luobusa ophiolite, southern Tibet, native iron occurs in two forms in the chromitites, as small round nodular intergrowths and as anhedral masses of acicular crystals. The composition of these grains is close to 100% Fe, although some have minute amounts of Mn (0.91 to 1.82 wt.%) and

Si (0.17 to 0.65 wt.%) [33]. This closely resembles the native iron grains of Ilia which are also present in two shapes (round nodules and angular grains) and their mineral composition also includes Mn (0.34 to 0.38 wt. %), but very low Ni (≤ 0.05 wt.%). [33] suggest that Fe–Ni and Fe–Co alloys and native Fe and Ni in the Luobusa chromitites are secondary minerals formed by alteration of PGE-bearing sulfides. [3] based on the shape and composition of the inclusions in the native Fe concluded that most probably, they represent immiscible silicate liquids that underwent partial crystallization upon cooling. They proposed that it is unlikely all of the native elements and alloys in the Luobusa chromitites could have formed by secondary processes. However, it is equally unlikely that they are primary minerals in the sense of having crystallized from the melt that formed the chromitites. The chromitites must have formed under hydrous, oxidizing conditions. Therefore, [3] argue that the native elements, PGE alloys and ultra-high pressure minerals, such as diamond, moissanite are xenocrysts derived from deep-mantle sources that were incorporated into the chromitites during crystallization.

In many cases, the presence of native iron in ophiolitic rocks has been associated with the serpentinization processes e.g. at Muskox intrusion, Nunavut-Canada where native iron, native copper, awaruite, wairauite and magnetite were documented in serpentinized lizardite [24]; at Josephine, Oregon-U.S.A. where native iron was documented in late, low temperature serpentine veins, cutting serpentinized harzburgite [29]; at Kachkanarskii massif, Russia, where native iron, nickel-iron alloy, wairauite and awaruite were documented in ore-olivinites [27]; and at Balangero, Italy where native iron with minor amounts of Ni, As, Co and sometimes Cu was documented in serpentinite [32].

[30] documented native iron in the serpentine vein of the second stage of serpentinization in dunites, from the ultramafic masses in the Sanbagawa belt, central Japan. In that case, the native iron has ovoidal and irregular shapes and is surrounded by magnetite. The native iron from the Sanbagawa belt, incorporates Ni (0.26–2.03 wt.%) and Co (0.26–2.78 wt.%) and is deficient in Cu and S.

[30] argue that the formation of native metals in serpentinites is not fully explained. At the same time, they suggested that during the serpentinization, the iron is released from olivine leading to the creation of magnetite. The serpentinization of olivine also produces hydrogen gas as indicated by the following reaction [24]:



In turn, the hydrogen gas could reduce magnetite to native iron and sulfides such as pentlandite and chalcopyrite to awaruite, native copper etc. Serpentinization process is a rather low temperature process where the upper stability of serpentine is ~ 450 °C [71], but it is uncertain if dissociation of magnetite is effected under such temperatures.

Ophiolitic rocks are present in the northwestern Euboea Island, as well as in neighboring part of the mainland in eastern Central Greece, i.e. Sperchios area, where the hydrothermal system also extends. In Northern Euboea, the ophiolites include mainly metabasites and serpentinites [52–54], however little is known about their mineralogy and origin. Although, the outcrops are very limited in Euboea island, [72] and [73] studying the soils and groundwaters, respectively, have identified their geochemical signature in great areas of northwestern Euboea. In the neighboring island of Skyros, [36] has identified native iron at ophiolite associated chromites. The ophiolite of Skyros includes also serpentinized harzburgites, gabbroic rocks, dolerites, tholeiitic basaltic lavas, rodingites, as well as ophealcites [74,75].

Based on all the above-mentioned data and all the possible scenarios evaluation, we suggest here a scenario, where the Ilia's native iron originates from deep ophiolite rocks and/ or chromites possibly associated with serpentinization processes. The native iron grains were remobilized and transferred by the hydrothermal fluid and were syngenetically enclosed as clastic grains within the thermogenic travertine.

6. Conclusions

The Ilia's hot spring represents a terrestrial active mineralizing hydrothermal system associated with ore-bearing travertines, located at the western extremity of the Northern Anatolia Fault and at the back-arc position of South Aegean Active Volcanic Arc.

Within the Ilia iron-rich thermogenic travertines, a new occurrence of ultra-pure native iron grains (with only up to 0.38 wt.% Mn and ≤ 0.05 wt.% Ni) accompany previously described metal-bearing minerals such as sulfides, native elements (Pb and Ni, alloys such as Ni-Co-Cu-Sn, Au±Cu-Ag), awaruite and REE-bearing phases. In common to the above metallic phases it is here suggested that also native iron was probably a product from a deeper environment that was remobilized and transferred by the hydrothermal fluids to the surface.

Regarding the origin of the native iron grains, after evaluating all the possible environments which native iron could be form and based on the until now available geological data of the study area, we suggest as the most possible scenario that the native iron is remobilized from ophiolitic rocks and chromites of the area, originated initially from reducing fluids which deposit it in the past during a serpentinization processes and later transferred from the hydrothermal fluid and syngenetically enclosed as clastic grains within the pores of travertine. We base our scenario on native iron mineral chemistry, morphological characteristic and the presence of other mineral phase at the same thermogenic travertine, such as other native elements like Pb and Ni, alloys like Ni-Co-Cu-Sn, and other mineral phases like awaruite, which have been documented in ophiolitic rocks elsewhere.

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