

Carbonate Coatings on Plant Twigs Found from a Travertine-Depositing Small Stream, Dijon, France

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Abstract: A number of coated grains (spherical to elongated ones in shape) were collected from a small stream, Dijon, France. They were characterized by typical concentric lamination surrounding broken twigs, and were thus identified as concentric precipitation on plant twigs. Within carbonate coatings of the plant twigs, two morphological groups including eukaryotic green algae (*Vaucheria* sp.) and cyanobacteria (*Scytonema* sp. and *Rivularia* sp.) were detected, which form carbonate crystals that are surrounding their filaments. The filaments could have triggered carbonate precipitation by photosynthetic removal of CO₂ causing the increase of alkalinity of the water, and by supporting their sheaths as nucleation sites. Such encrusted twigs were found from 70 meters downstream, in which temperature and pH were measured as 11.1°C and 8.18, respectively. These water chemistries (11.1°C and pH 8.18), with the aid of microbial photosynthesis, were likely to provide a suitable condition for carbonate precipitation on the twigs.

Keywords: carbonates, coated grains, cyanobacteria, encrusted twigs, travertine

Introduction

In modern environments, calcareous minerals are actively depositing in fluvial systems, particularly dominant in rivers or lakes of European countries including France, Germany, Italy and Croatia [see Ford and Pedley (1996) for a list of 22 European countries]. This is mainly because surface waters of the fluvial systems of those countries are highly supersaturated with respect to calcium carbonates, consequently forming extensive carbonate deposits termed as travertine or tufa (e.g., Chafetz and Folk, 1984; Srdoc et al., 1985; Riding, 1991; Pentecost, 2005; Golubic et al., 2008). Depositional condition of such precipitates is governed mainly by physico-chemical factors (e.g., temperature and pH) of the

water bodies, which are in turn closely related with the amount of carbon dioxide in water system (Pedley, 1990; Pentecost, 2005). In addition, metabolic activities of photosynthetic organisms living in the water also play a major role in the formation of such carbonate deposits, because photosynthesis removes CO₂ from the water, increasing pH of the water favorable for carbonate precipitation (Merz, 1992). This phenomenon is more easily identified in a running water system where chemical gradients change dynamically downstream with distance. Under such a highly saturated running water system, carbonate deposits occur as coated grains, crusts on hard substrates, and encrustations on plant twigs submerged in water, all of which form travertine or tufa deposits [see Pentecost (2005) for review].

Carbonate coatings on plant twigs are quite common precipitates, forming spherical to elongated structures in travertine-depositing running water system. Although diverse organisms (e.g., mosses, green algae and cyanobacteria) have been known to be involved in the formation of such coatings (Freytet and Verrecchia, 1998), controversies have existed for years over the relative importance of inorganic and

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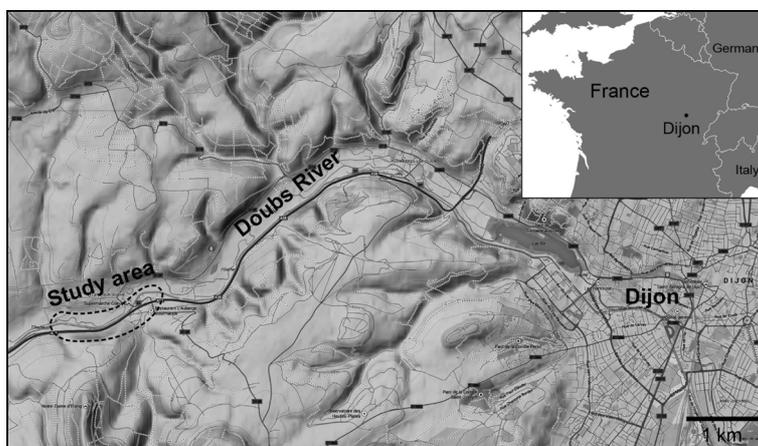


Fig. 1. Locality map of the study area. Encrusted twigs were collected from the small stream (marked as a dashed circle), Dijon, France.

organic processes in their formation (e.g., Folk et al., 1985; Chafetz et al., 1991). Here in this paper, we introduce carbonate coatings on plant twigs collected from modern tufa depositing environments, and relate the observed mineral formation with physico-chemical properties of the source water. We also focus several fundamental questions concerning dynamics of carbonate growth and microbial role in the formation of encrusted plant twigs.

Sampling Locality, Materials and Methods

A number of small streams along Doubs river in the area of Antheuil, west of Dijon, France exist along the mountain hills, which provide the dominant source of water to Bourgogne Canal flowing northwest (Fig. 1). The streams are fed by ground water issued from underground cave. After being issued from the cave, the stream water (a few decimeters deep, 1-10 m wide, and <5 degree) flows downslope (Fig. 2). As the stream becomes wider, a series of pools and dam-like barriers of travertine are developed, producing many micro-terrace formation along the creek (Fig. 2B). Three major carbonate deposits in this stream were detected: thin crusts on hard substrates (Fig. 2C), coated grains (spherical to slightly elongated structures, Fig. 2D, F), and encrustations on plant twigs (Fig. 2F-

H). Crusts are so soft and thin that they could be collected by slipping a knife blade underneath and gently lifting them (Fig. 2C). Coated grains are more frequently encountered on the bottom of micro-terrace pools, which is covered predominantly by transported pebbly sands and recently formed carbonate particles (Fig. 2F). Such grains are often fused together forming small carbonate barriers (Fig. 2D). Most of encrusted twigs are found in marginal pools and along the stream margin where water flows slowly (Fig. 2E-H).

Samples of carbonate coatings on plant twigs and coated grains (particularly elongated ones) were collected along the stream, from 70 to 195 m downstream systematically. Samples of the encrusted twig were cut perpendicular to long axis of twigs, and petrographic thin sections were prepared for light microscopy. For microbiological analysis, encrusted twigs were also collected separately with fixation in 3% formaldehyde in stream water and stored in a refrigerator. The organisms incorporated within the carbonate coatings were extracted after dissolving carbonate minerals with acetic acid under the dissecting microscope. Some organisms extracted were stained with alcian blue and ruthenium red to visualize the structures of extracellular polysaccharide sheaths, and biological slides were prepared for light microscopic examination.

In order to document physico-chemical conditions

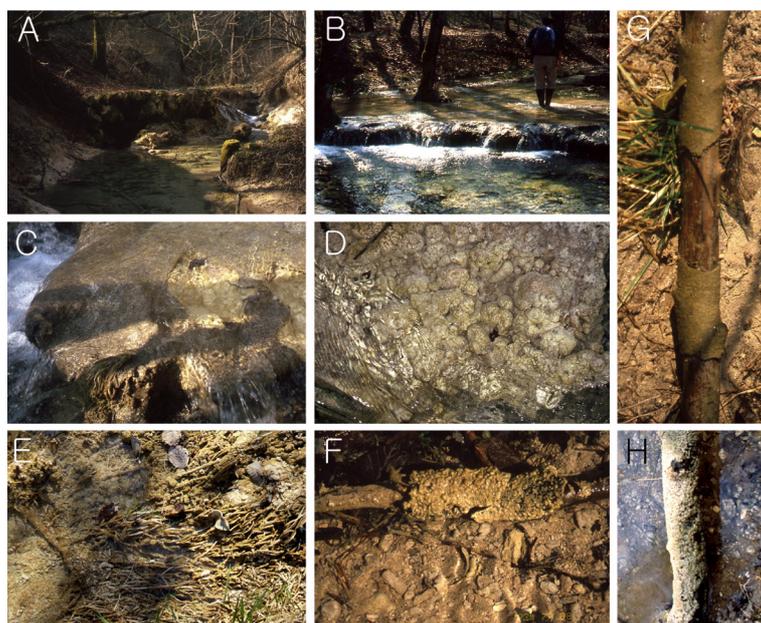


Fig. 2. Photographs of diverse carbonates found from the small stream. (A) No precipitates were detected from the source water to 60 m downstream. (B) Dam-like barriers developed across the stream. (C) Thin crusts of soft carbonates covers the surface of travertine dams. (D) Bumpy surface structures of travertines developed on rocky substrates. (E, F) Both encrusted twigs and elongated coated grains were found in marginal pools. (G, H) Details of encrusted twigs.

for carbonate precipitation in this running water system, water chemistry (temperature and pH) was measured in situ using micro-electrodes along the stream. A series of measurements (5 meters interval) downstream from the source waters were repeated for each sampling period to establish the temporal variability of the measured parameters. Water samples were also collected for measurements of calcium and bicarbonate concentrations, and alkalinity after filtering in the field (for the detailed method, see Franson, 1976).

Result

Water chemistry

Ground water issues from a carbonate cave to surface, from which the stream starts to flow downstream. As the process continues, the stream becomes wider and water flow is subdivided, often forming small dam-like barriers across the stream (Fig. 2B). Both temperature and pH were measured at every 5 meters

from the starting point (0 m) to 230 m where a small village is located (Fig. 3). It was expected that the stream water changes in its chemistry as the stream flows because temperature of stream water tends to increase, and because carbonate minerals were observed to precipitate below 60 m from the starting point of the stream. Stream waters are saturated in terms of calcium carbonate with great amount of Ca^{+2} and HCO^{-3} , mainly because the waters are issuing out of the Jurassic basement limestones. Thus comparison of water chemistries alone from the measuring places would provide valuable information about chemical changes responsible for carbonate precipitation on plant twigs, and such measurements can also be compared with the results obtained in the analysis of carbonate precipitates.

At starting place of the stream, the source water was cold with temperature of 10.8°C and pH was slightly alkalic with a value of 7.52 (Fig. 3). As water flows downstream, the temperature of stream water was increased more or less constantly, up to 12.3°C .

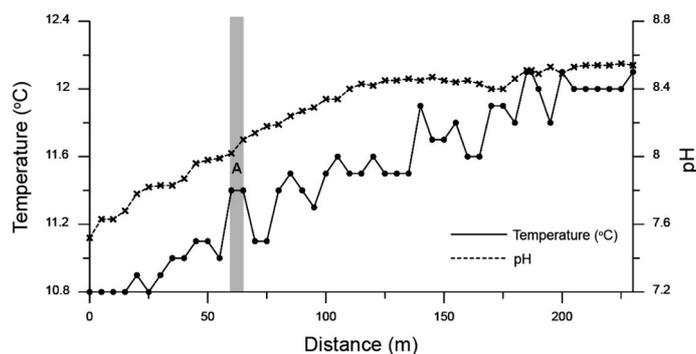


Fig. 3. Diagram of temperature (solid line) and pH (dashed line) measured in a small stream. Note that both temperature and pH increased with distance. A in grey box represents a first place where encrusted twigs were found.

This increase is due to continuous contact of stream waters with ambient atmosphere of about 20°C, and due to increase of exposure time to sunlight of stream waters. Sudden decreases of temperature were often observed in some places (e.g., at interval from 70 to 75 m, Fig. 3). The places were characterized by dense shade of trees, which was likely to lower the temperature of stream waters.

A value of pH was also increased constantly downstream from 7.54 to 8.55 (Fig. 3). Such an increase of pH is clearly related with CO₂ loss of stream waters, because loss of CO₂ in water column tends to lower concentration of hydrogen ions (H⁺) that causes alkalinity of the water (Chen et al., 2004; Anzalone et al., 2007). Three factors, in this stream system, may be responsible for the loss of CO₂ downstream: degassing of CO₂ through agitation of running water, biological uptake of CO₂ through photosynthesis, and lowering of CO₂ solubility due to temperature increase.

Carbonate coatings on plant twigs

Carbonate precipitation including coated grains, crusts and encrusted twigs was observed to initiate from 60 m below the source waters (Fig. 3, grey box). Characteristic travertine deposits, below this level (60 m), were found along the waterfall built by dam-like terrace where thin crusts with living mosses covered surface of dams (Fig. 2B). These dam-like structures are most easily encountered carbonates in

travertine-depositing system in European countries (D'argenio et al., 1995; Ford and Pedley, 1996; Golubic et al., 2008). From 80 m below, stream becomes wider and deeper, forming a series of marginal pools.

In such marginal pools where water runs slowly, leaves and broken twigs (0.2 to 1.0 cm wide, 4.0 to 10.0 cm long) were observed to be encrusted thinly by white carbonate minerals (Fig. 2E). In small pools below 80 m, almost all the twigs submerged in the water were thickly encrusted, and thus encrusted twigs characterized by concentric carbonate coatings were easily found (Fig. 2F, G, and H). Most of the encrusted twigs were deposited individually in the bottom of the marginal pool like clastic sediments. Top portion of the encrusted twigs deposited in floor showed largely brown, while bottom was white in color. In addition, thickness of encrusted carbonates differed at both sides: thick carbonate minerals in upper parts while thin in lower parts attached to the bottom. Such differences between two parts, in terms of color and thickness of carbonate minerals, seem to be closely related with absence or presence of living microbes responsible for the carbonate production (e.g., Golubic et al., 2008). No such differences, however, were observed in carbonate coatings of vertically erected stems, which is probably due to water and light availability from all directions.

Spherical (oncoids) to elongated coated grains (0.5 to 1 cm wide and 1 to 5 cm long) were also commonly

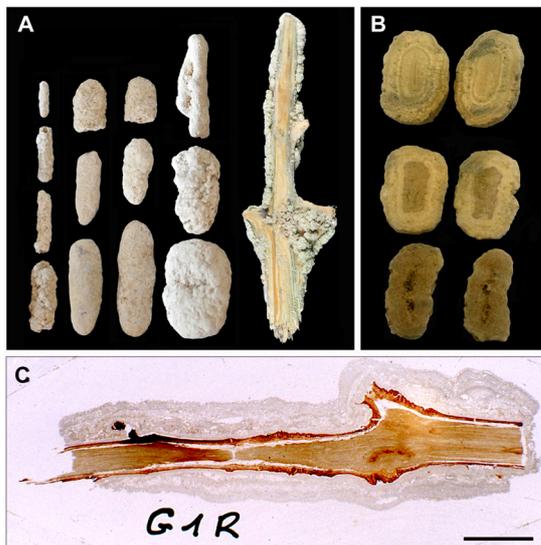


Fig. 4. Encrusted twigs and diverse coated grains. (A) Spherical to elongated coated grains found in the bottom of marginal pools. Original plant materials are present inside the structure, as clearly seen in right sample. (B) Oncoid slabs sectioned perpendicular to long axis, showing central nucleus and concentric lamination. (C) Thin section of an encrusted twig shows carbonate coatings on a broken twigs. Scale bar in (C) represents 1 cm for (A) and (B), 0.5 cm for (C).

found in the bottom of marginal pools and shallow channels developed below 90 m downstream (Fig. 2F and Fig. 4). Some of spherical structures (Fig. 4B) are normal oncoids showing concentric lamination with a central nucleus, while others or elongated structures (Fig. 4A) differ from normal oncoids in presence of broken twigs (a right sample in Fig. 4A and Fig. 4C) in the central parts. This is clearly evidenced when viewed in cross section of the elongated structures, from which original plant tissues coated by thick carbonates were remained intact (Fig. 4C).

It is likely that the elongated structures formed initially by carbonate precipitation around broken plant twigs that were originally floating in the water column. As precipitation on twigs proceeded, they became bigger and heavier, and sank finally to the bottom of the marginal pool or in the water channels. After the encrusted twigs were deposited in the bottom, more carbonate became precipitated in the middle parts then at the edge of the broken twigs, finally giving an

appearance of slightly elongated structures to the encrusted twigs.

Organisms involved in the formation of encrusted twigs

Diverse organisms including aquatic mosses, diatoms, green algae and cyanobacteria were identified to be involved in the formation of carbonate minerals in this stream. Like in travertine-depositing systems of European countries (Freytet and Verrecchia, 1998; Plenkovic-Moraj et al., 2002), aquatic mosses were the most dominant organisms in the formation of carbonate crust along the waterfall (Fig. 2B, C). They grew in waterfalls and rapids where they trapped and precipitated porous carbonate rocks. Others including diatom and green algae were also observed within the carbonate minerals, but it is unclear whether they were involved in the production of carbonate minerals, or they were simple inhabitants on the crust. Within carbonate coatings on plant twigs, two morphological groups, eukaryotic green algae (*Vaucheria* sp.) and cyanobacteria (*Scytonema* sp. and *Rivularia* sp.) were detected, forming carbonate crystals surrounding their filaments.

Filamentous cyanobacteria were found as predominant organisms in thin sections of carbonate coatings. Tightly spaced and/or diverging filaments were surrounded by micritic grains that outline the original gelatinous sheath of the filamentous cyanobacteria (Fig. 5). Carbonate minerals initiated to nucleate on sheath's exterior surfaces along the filaments, finally forming calcareous tubes (Fig. 5C, D, E). In order to escape burial, thereafter, cellular trichome glided out of the tubes and formed new sheaths (Seong-Joo et al., 2000; Golubic et al., 2008). Empty tubes now filled with white calcareous spar are vestiges of cyanobacterial filaments (Fig. 5C), which are also seen in cross section as empty sparitic circles (Fig. 5D). SEM images of surface of encrusted twigs document more evidently the vestiges of the filaments in which carbonate crystals grew epitaxially outward from the filaments (Fig. 5E).

Constructed by dense, vertically erected and slightly diverging calcareous tunnels, the encrusted twigs show

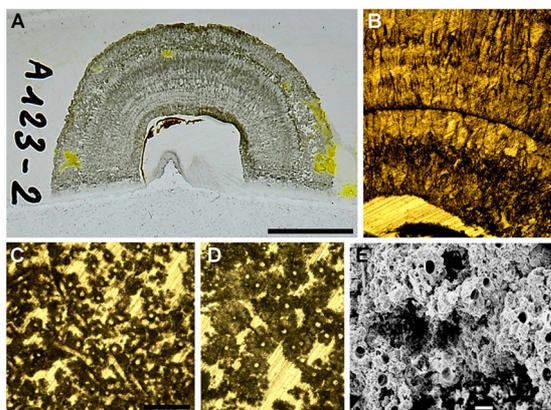


Fig. 5. Carbonate structures of encrusted twigs. (A, B) Profile section of encrusted twigs shows the growth direction of carbonate minerals, in form of upward diverging bushes. (C, D) Upper part of the encrusted twigs characterized by abundance of filamentous tubes. Original filaments are now represented by sparite-filled tubes (C), circles (D), or empty tubules (E). Scale bar in (A) is 1cm, scale bar in (C) is 200 μm for (B, C, D), and scale bar in (E) is 20 μm .

zonal changes of mineral density and porosity (Fig. 5A). The zonation is characterized by an initial zone (Fig. 5B) of radiating and compacted calcite crystals followed by a porous zone (Fig. 5C, D) with a great number of filaments associated. Such a zonation may reflect seasonal changes in environmental conditions for microbial growth and calcification (Emeis et al., 1987; Merz-Preiss and Riding, 1999; Golubic et al., 2008).

For example, photosynthetic filaments under favorable conditions (generally in summer) may have thrived, and thus lowered CO_2 level in water column, which caused rapid precipitation along the filaments. During the winter season, however, carbonate precipitation was very slow as a result of restricted microbial growth. Such a cyclic growth is known to create zonation pattern in carbonate deposits such as travertine and stromatolite (Golubic et al., 2000). In this study, however, it is more likely that the zonations were resulted from diagenetic processes that modified the texture of the deposit and obliterated the tubular traces left by filaments. This interpretation may be supported by the fact that the lower parts (close to the

center) of the coatings are characterized by compacted carbonate crystals with a few remains of filaments (Fig. 5B), while algal remains increase in number toward the outer parts of the coatings (Fig. 5C, D).

Discussion

Encrusted twigs were found from 60 m downstream, which was characterized by temperature of 11.4°C and pH of 8.1. Below this level, not only submerged twigs but also most hard grounds were observed to be highly calcified. Based on such a simple observation, it may be concluded that this chemical condition (11.4°C and pH 8.1) is suitable for carbonate precipitation on plant twigs. Indeed, the value of pH 8.1 seems a critical point that initiated carbonate precipitation in this stream system. However, if we consider CO_2 removal by photosynthetic filaments associated in the encrusted twigs, carbonate precipitation without aid of photosynthesis may have initiated from the level below 70 m. This is because photosynthetic removal of CO_2 from the water lowers concentration of hydrogen ions (H^+), and thus provides more favorable condition for carbonate precipitation (Chen et al., 2004; Anzalone et al., 2007). This interpretation, however, does not confirm biological role in the formation of encrusted twigs. This is mainly because micro-environments, favorable condition generated by photosynthesis, tend to be diluted quickly in a running water system. This may explain the phenomenon that broken twigs in standing water were more thickly calcified than those in running water.

Proponents for biological roles have insisted that two factors play a critical role in carbonate precipitation: bicarbonate uptake during photosynthesis and suitability of sheath for calcite nucleation (Pentecost and Bauld, 1988; Merz, 1992; Schultze-Lam and Beveridge, 1994; Ferris et al., 1997). Cyanobacteria, indeed, has long been known to serve as heterogeneous nucleation templates for calcite precipitation (e.g., Merz, 1992; Schultze-Lam and Beveridge, 1994). On the other hand, chemical models have not been ruled out either, or even considered alternative possibilities (Chafetz

and Folk, 1984; Pentecost, 1978; Lorah and Herman, 1988). The chemical model has argued that 1) calcifying cyanobacteria are restricted to waters that are highly supersaturated with respect to carbonate, and that 2) in such environments almost all substrates, even copper substrates (Chafetz et al., 1991), are highly calcified. Accordingly, supporters of chemical models have insisted that precipitation caused predominantly by chemical factors leads to sheath encrustation, whereby filaments only provide a suitable site for crystal nucleation (Pentecost, 1978; Dennen and Diecchio, 1984).

It is true, in study area, that highly calcified twigs were found more in stagnant pools than in shallow channels of running water. It is, thus, likely that photosynthetic CO₂ removal is almost negligible in this running water streams, as suggested by some researchers (Chafetz and Folk, 1984; Pentecost, 1978; Lorah and Herman, 1988). In addition, no species-specific calcification (different crystals around different calcifiers such as mosses and cyanobacteria, see more in Merz and Zankl, 1993) was not observed in the encrusted twigs. However, a positive correlation between the presence of cyanobacteria and carbonate coatings on twigs may support biogenic interpretation of the encrusted twigs, even though we do not decide clearly, at this moment, degree of biogenicity of the encrusted twigs in this fast-flowing environment.

Summary

A variety of coated grains (spherical to elongated structures) are present in travertine-depositing running water systems, particularly in European countries. We collected those grains from a small stream of Dijon, France, and documented that they (particularly elongated structures) were carbonate coatings on plant twigs. Original plant twigs were detected from inside of many elongated structures and filamentous cyanobacteria were also preserved within the carbonate coatings. Cyanobacterial filaments were vertically erected to the surface of the carbonate coatings. Empty tubular structures, now filled with sparitic calcite, were

vestiges of cyanobacterial sheaths left after gliding of trichomes. Filamentous sheaths seems to have provided suitable sites to nucleate carbonate crystals, and thus a great number of tiny micrite grains were lined along the surface of cyanobacterial sheath.

These encrusted twigs were found from 60 m downstream, and below this level, not only submerged twigs but also most hard ground were observed to be highly calcified. Water chemistry of 11.4°C and pH 8.1, measured at the first places where encrusted twigs were observed, seems a critical point that initiated carbonate precipitation in this stream system. However, if we accept negligible effect of CO₂ removal by microbial photosynthesis in this running water system, it is premature to decide the degree of biogenicity in the formation of the encrusted twigs.

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