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MICRO-RAMAN SPECTROSCOPIC STUDY OF PIGMENTS IN WALL PAINTINGS, ICONS AND ILLUMINATED MANUSCRIPTS FROM REPUBLIC OF MACEDONIA

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The micro-Raman spectroscopic analysis of pigments in the Republic of Macedonia comprises identification of pigments on different supports: mortar, wood and paper. The analyzed wall paintings pigments cover a historic period dated from the late Roman time (4th to 6th century AD) until the mid of 19th century. Pigments used in icons, dating from 11th to the mid of 19th century were also analyzed. As for the pigments in manuscripts, only pigments in 16th century oriental manuscripts were analyzed so far.

The study of the pigments used in the red, white, black and blue paints showed that they were quite uniform with little changes through the centuries. The pigments for the yellow color were much more numerous, while green pigments showed a pronounced change over the time.

Red pigments used for the red color/paint were mainly minerals: cinnabar, hematite, or lead based red pigments that did not change through the ages. The palette of the **yellow pigments** started with mineral based pigments and grew richer than the red ones. It included either natural minerals, such as goethite, massicot, realgar or orpiment, but also manufactured pigments, such as lead-tin yellow type II, chrome yellow and chrome yellow deep, and even, Indian yellow. **White and black pigments** were mainly gypsum or calcite (for the white), and ivory black or lamp black (for the black). Lapis lazuli (ultramarine) was the most frequently identified **blue pigment** in our micro-Raman spectroscopic studies. This highly valued blue mineral was found in almost all wall paintings, except the late Roman frescoes and in almost all icons as well as in the 16th century oriental manuscripts. Besides this blue pigment, indigo and azurite were also used in the blue paint.

Green pigments used in paints, profoundly changed through the centuries and were the most interesting and, sometimes the most difficult pigments to identify. Generally, the nature of the green pigments could be related to the historic period of the artifact that they were taken from. The green pigment used in the Roman frescoes, found in the Roman cities of *Scupi* and *Stobi* (4th-6th century AD), was green earth. This earth mineral (whose main constituents are the minerals celadonite

and /or glauconite) was a widely used green pigment in the Roman times. However, according to the geologists, celadonite and glauconite have not been found on the territory of the Republic of Macedonia, suggesting that they have been imported during the Roman times. This could probably be the reason why in the medieval time, green earth was rarely found as a pigment applied on Byzantine wall paintings and icons. Instead, in these art works, the green color of the paint was obtained by mixing the blue pigment (such as lapis lazuli, azurite or Prussian blue) with yellow pigment (goethite or chrome yellow deep). The richest palette of green pigments was detected in the oriental illuminated manuscripts (that were copied in Iznik), where copper minerals, malachite, bronchanitite and atacamite, or the mixture of indigo and orpiment (vergaüt) were detected.