Supplementary file S2. Spectral data

Supplementary S2.1. Reflectance spectra and Raman spectra for inorganic-based colors

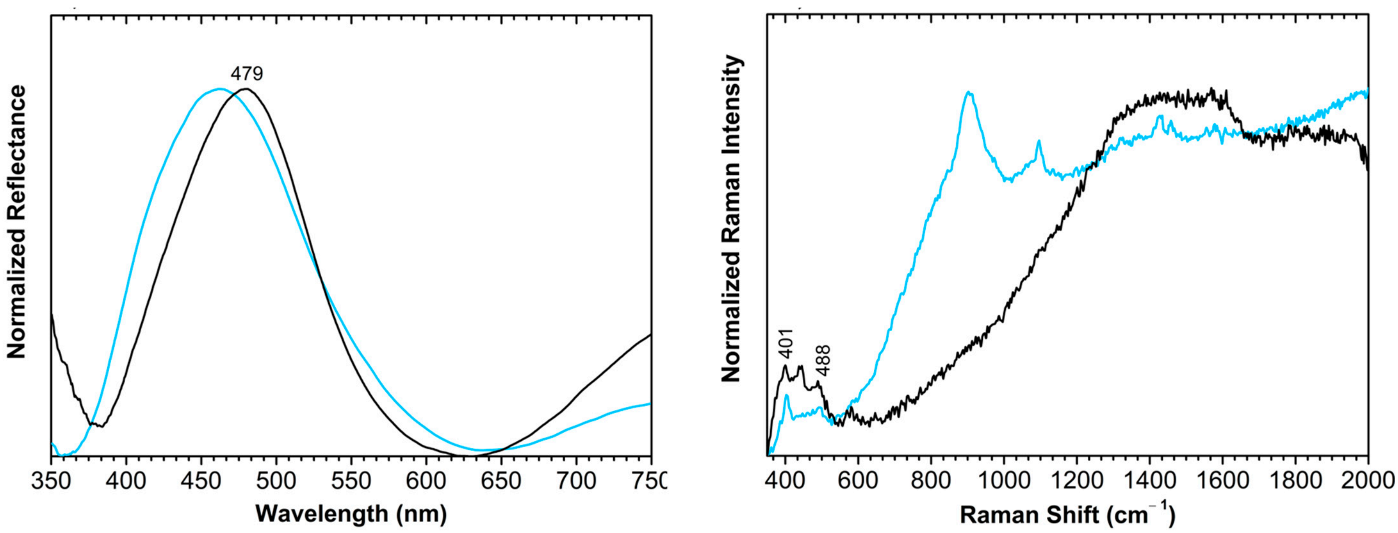

Figure S1. Reflectance and Raman spectra of azurite found in folio 212r of the *Rich codex* (black), compared with an azurite reference (blue). This blue color was identified in a restricted area, being not the main blue. While the reflectance spectrum indicates the presence of azurite, with Raman spectroscopy, its identification is less straightforward as the spectrum is affected by fluorescence, with only two characteristic bands being identified at 401 and 488 cm^{-1}.

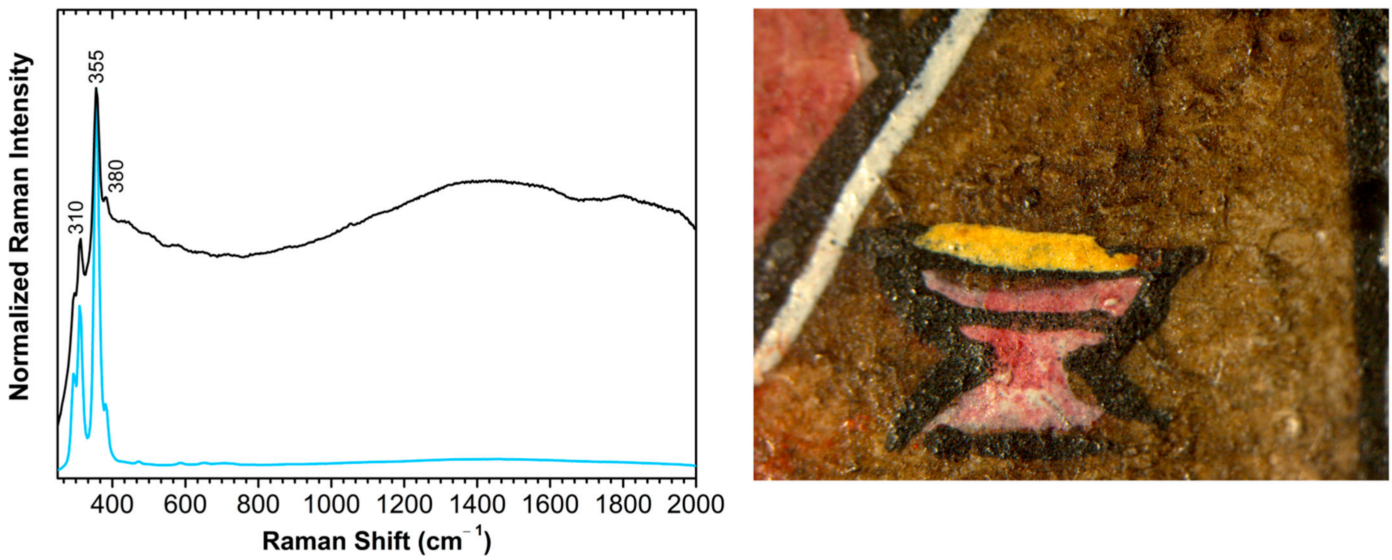

Figure S2. Raman spectra of orpiment found in folio 192r of *Rich codex* (black), compared with orpiment reference (blue). This pigment was applied pure only in this manuscript, in the detail of a paint bottle.

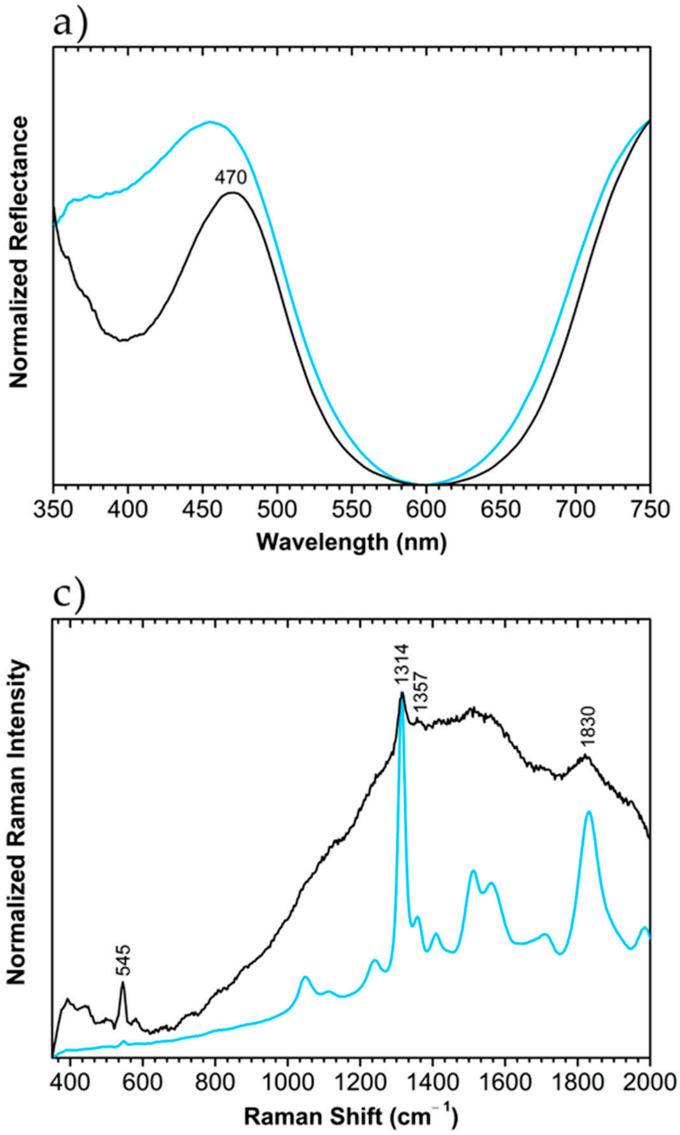

Figure S3. Spectra of lapis lazuli applied pure (black) compared with references (blue). Reflectance (a, b) and Raman (c, d) spectra of lapis lazuli in the *Rich codex* and *Musicians' codex*, respectively. Lapis Lazuli was applied as a pure pigment mostly in backgrounds.

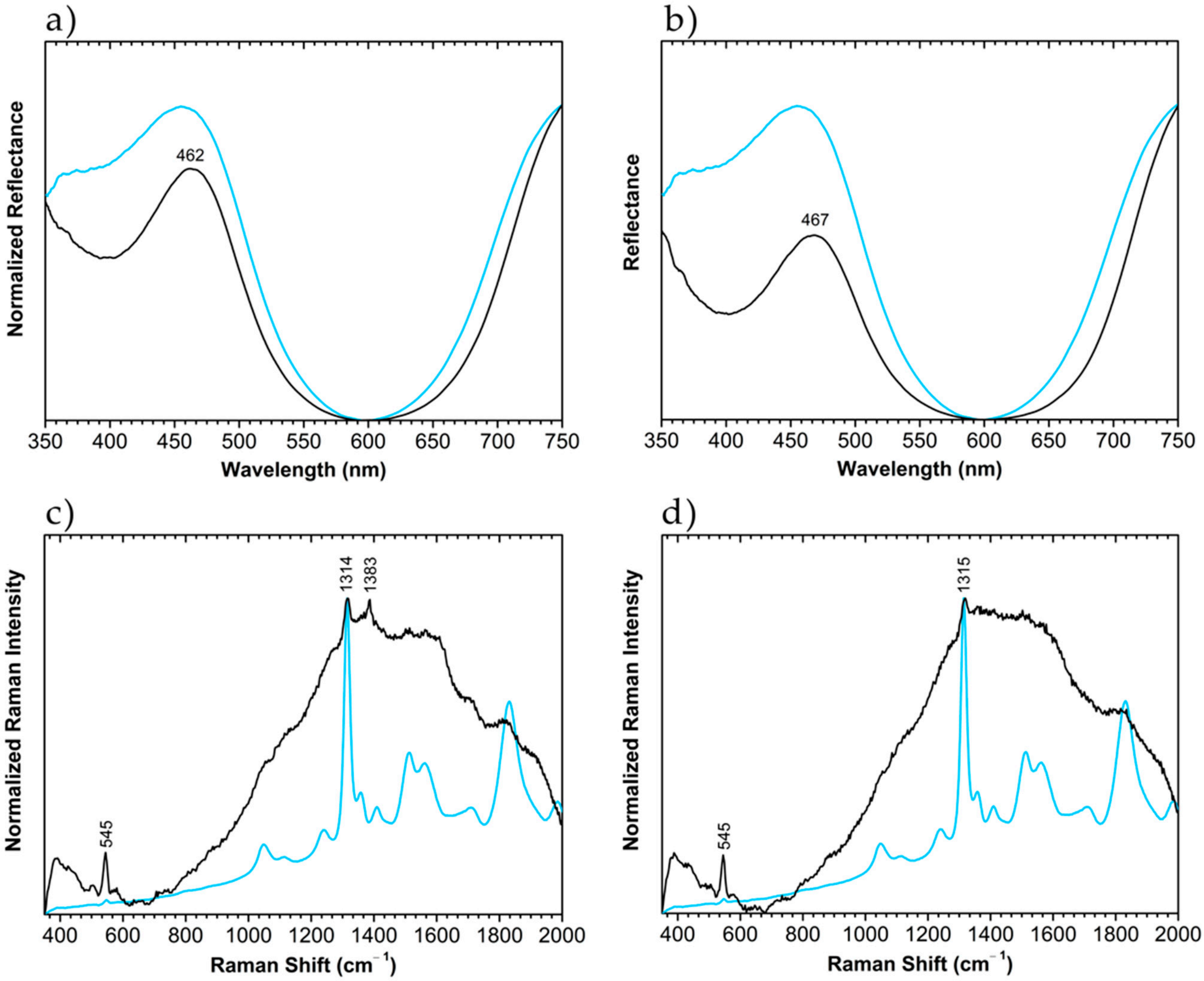

Figure S4. Spectra of lapis lazuli applied pure (black) compared with references (blue): Reflectance (a, b) and Raman (c, d) spectra of lapis lazuli in *Lapidary* and *Book of Games*, respectively. Lapis Lazuli was applied as a pure pigment mostly in backgrounds.

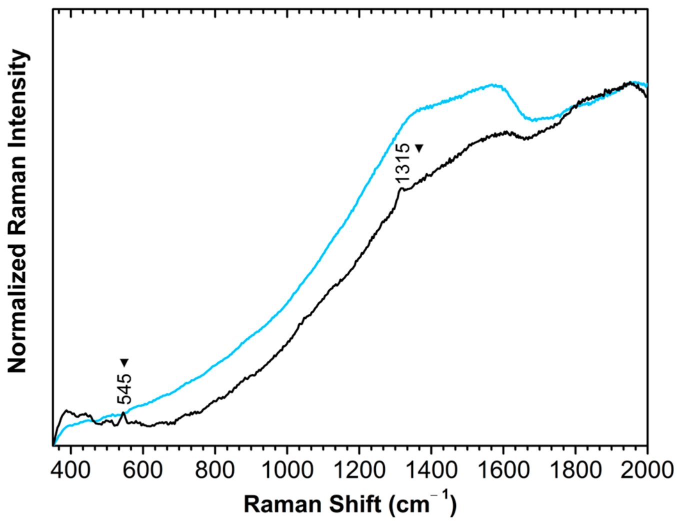

Figure S5. Raman spectrum of a mixture of lapis lazuli and carbon-based black in *Musicians' codex* (black) compared with the bone black reference (blue).

The analysis presented in Figure S5 is an example of a common question when analyzing small details: is carbon-based black within a mixture with lapis lazuli or is the signal coming from a nearby contouring line? The visual observation plays a fundamental role, as the blue appears to be darker than the paints with pure lapis lazuli signal, so the present paint is most probably a mixture. Nonetheless, these questions reinforce the need for a multianalytical approach and the importance of possible microanalysis.

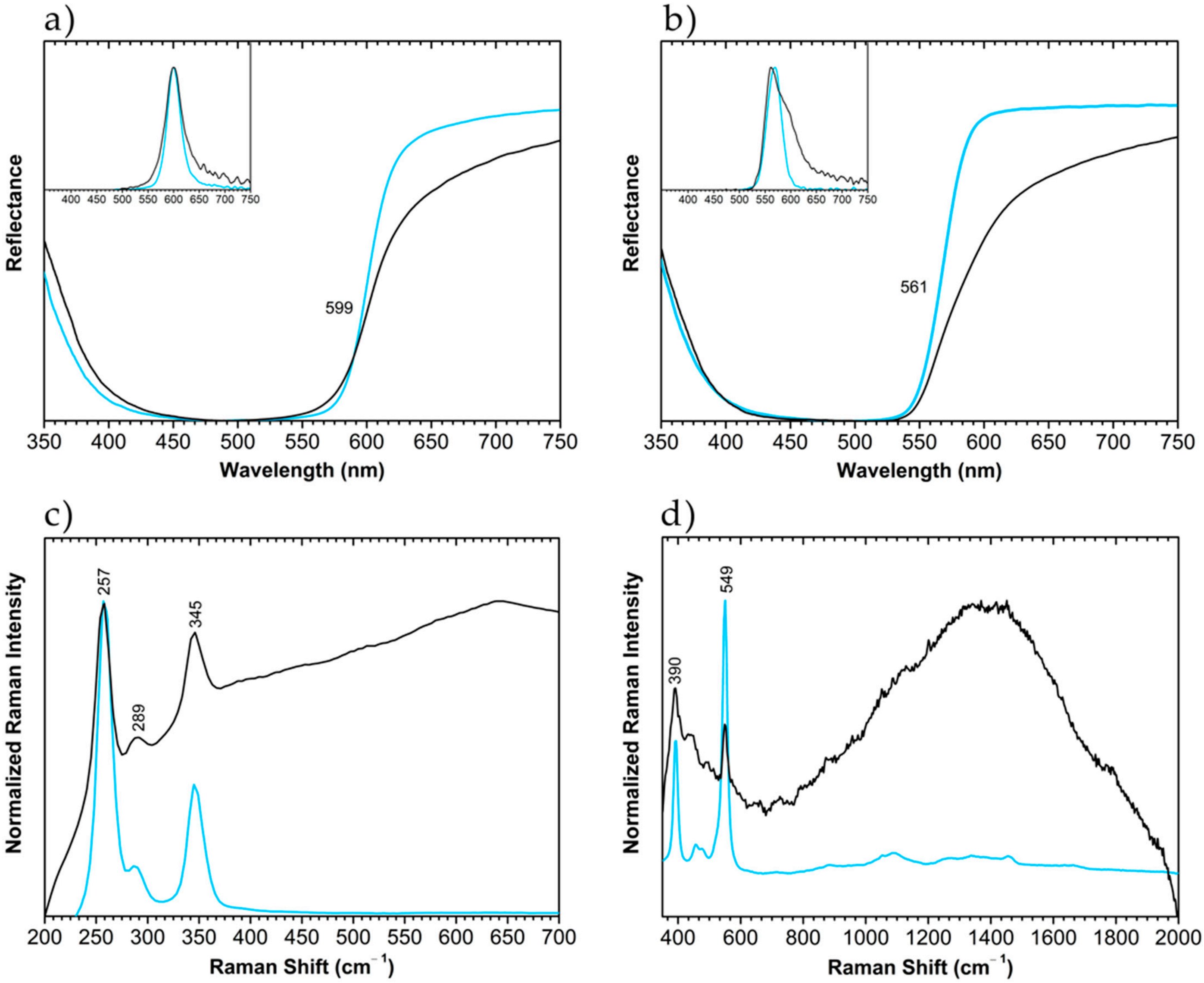

Figure S6. Spectra of red and orange paints (black), representative of the pigments found in the four manuscripts, compared with references (blue). Reflectance spectra and first derivative indicating (in the inset) vermilion (a) and red lead (b) in *Lapidary*. There is a close match between the historical paints and the references. This identification is confirmed with Raman analysis, where vermilion (c) is identified by its characteristic bands at 257, 289, and 345 cm^{-1} and red lead (d) by its characteristic bands at 390 and 549 cm^{-1}.

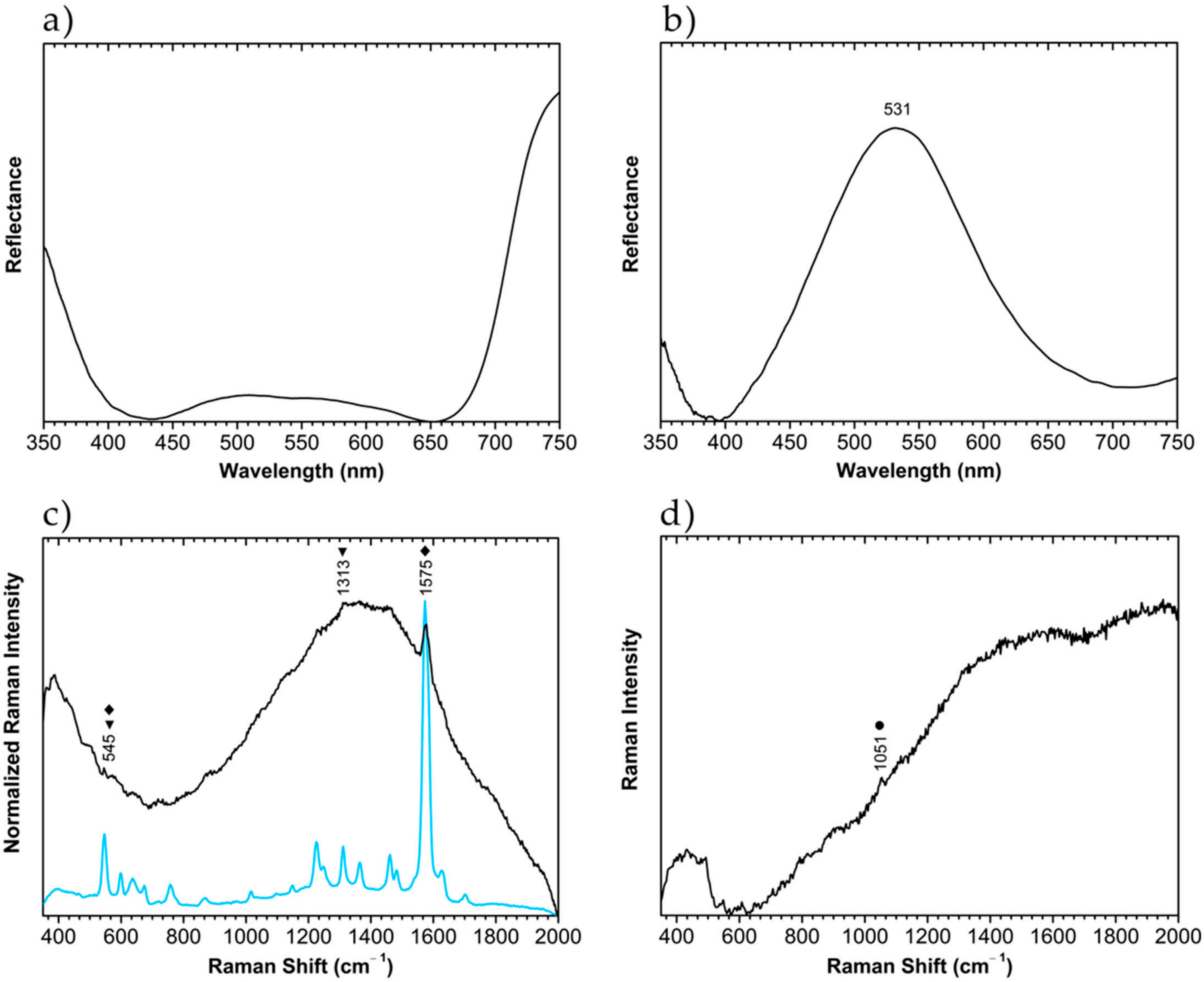

Figure S7. The spectra of two distinct greens in *Musicians' codex* (a, c) and *Lapidary* (b, d). Reflectance spectra indicate (a) the characteristic spectrum of indigo and (b) the presence of a green, probably of cuprous nature. The Raman spectrum in c) corroborates the presence of indigo, identifying also lapis lazuli. There was no signal in Raman spectroscopy for the green in *Lapidary*; however, in a similar paint of the same manuscript, it was possible to identify lead white in a mixture with the green. The main Raman bands are assigned to lapis lazuli (▼), indigo (♦), and white lead (●). *Vergaut* and bottle green are discussed in the main text.

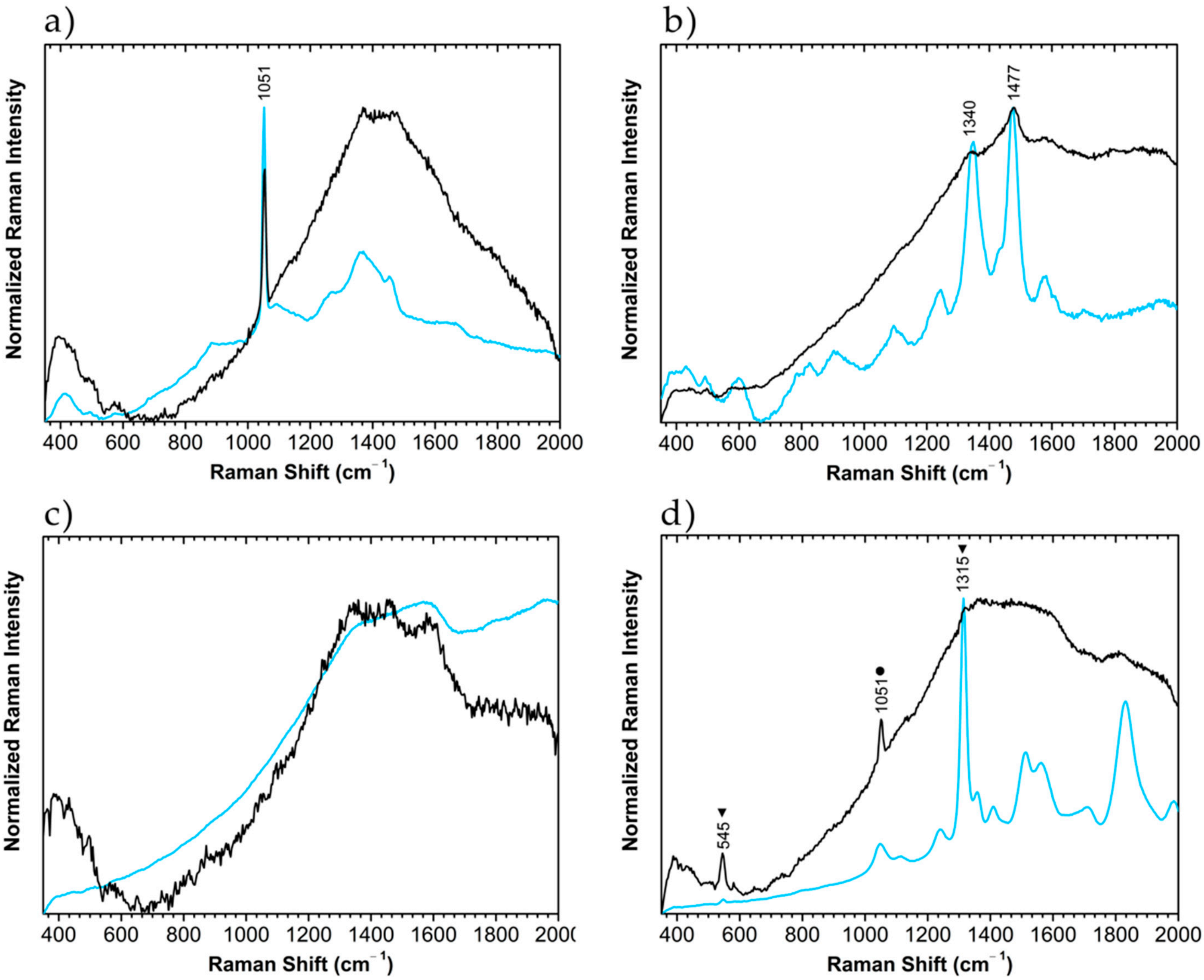

Figure S8. Raman spectra of white and black in particular objects (black) compared with references (blue) for (a) *Rich codex*, lead white paint found in the white lion of folio 192r; (b) *Lapidary*, black paint used to paint a bird of folio 49r, that corresponds to iron-gall ink; (c) *Lapidary*, carbon-based black used to paints scorpion in folio 63r; and d) *Lapidary*, blue found in little stone, resulting from the mixture of lapis lazuli and lead white. The main Raman bands detected are assigned to lapis lazuli (▼) and white lead (●).

Table S3. Details of the elements presented in the previous analysis (Figure S8), from left to right: the white lion of the *Rich codex*, the bird, the scorpion, and the stone of *Lapidary*, in folios 49r, 63r and 87r.

Supplementary S2.2. Reflectance data on colors based on dyes: pinks and purples

Fiber Optic Reflectance Spectroscopy (FORS) was used for the first screening to identify dyes, as it can be very indicative for brazilwood lake pigments. As is observed in Figure S9a, the anthraquinones of vegetable and animal origin are characterized by two bands, with the most intense being around 493 and 513 nm, respectively. The identification is straightforward compared to the unique apparent absorbance band of brazilwood at 555 nm. Even when comparing to dragoon's blood, the difference between both apparent absorbance maxima is clear.

The dyes used for this comparison are described in the bibliography and studies of medieval illuminated manuscripts. They are also the main red dyes described in medieval sources.

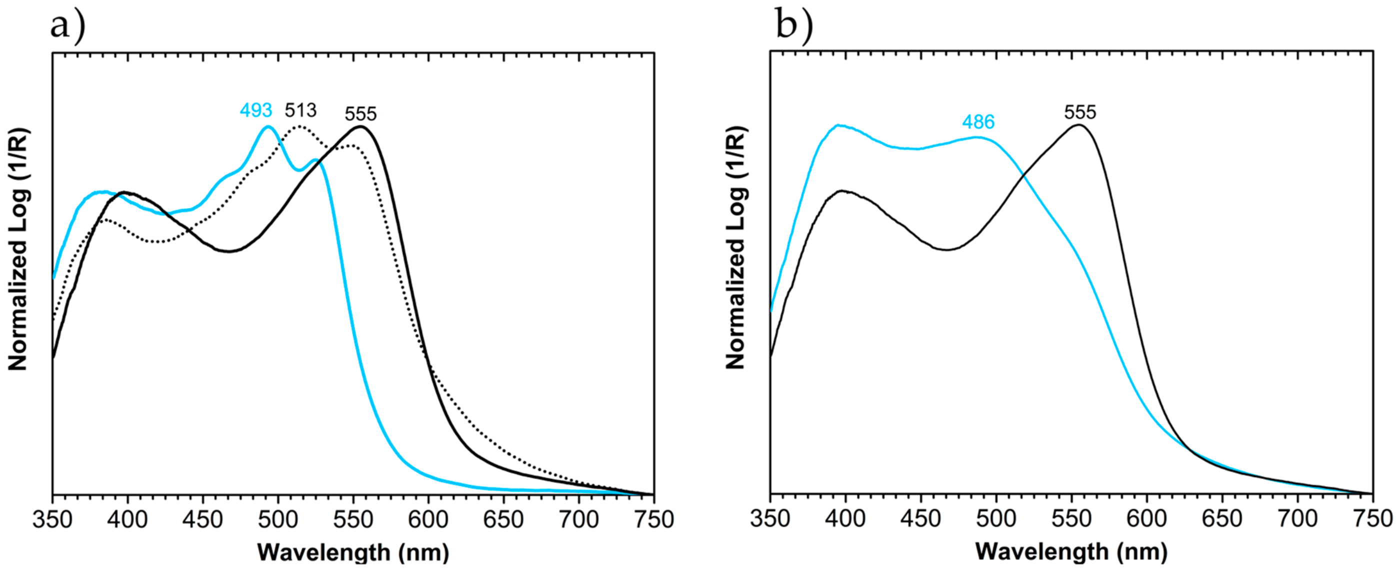

Figure S9. Apparent absorbance spectra of a) red dyes as pigments lakes, such as madder (blue) and lac dye (pointed); and b) dragons' blood (blue), a dye that is frequently described in medieval treatises as being used in illuminations compared to brazilwood (black).

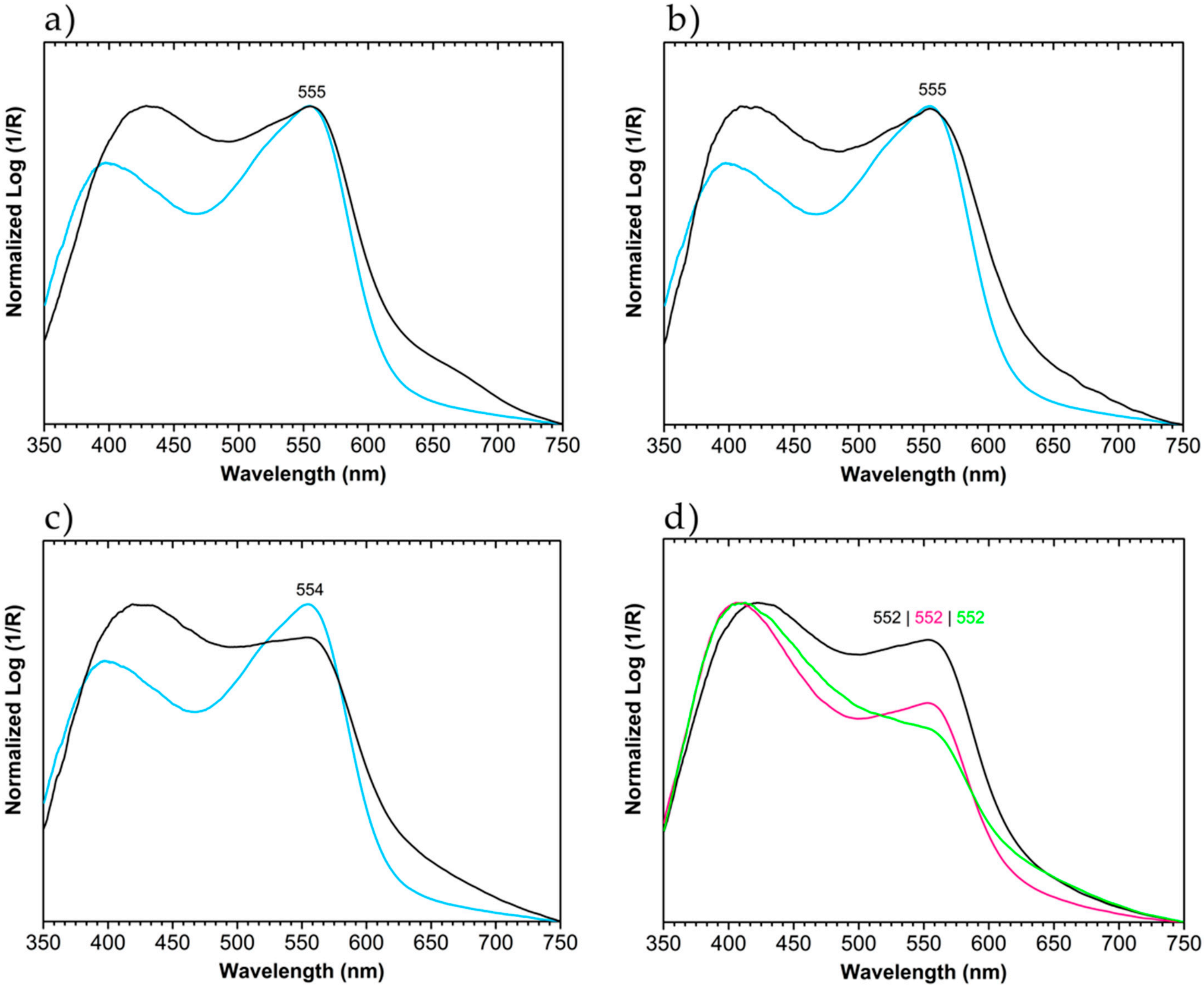

Figure S10. Apparent absorbance spectra of brazilwood pinks (black) in the manuscripts, compared with references (blue): (a) *Rich codex*, (b) *Book of Games*, (c) *Musicians' codex* and d) *Lapidary*. In *Lapidary*, the spectra are shifted, which can be indicative of the presence of additives such as lead white (black), calcium carbonate (pink), and gypsum (green).

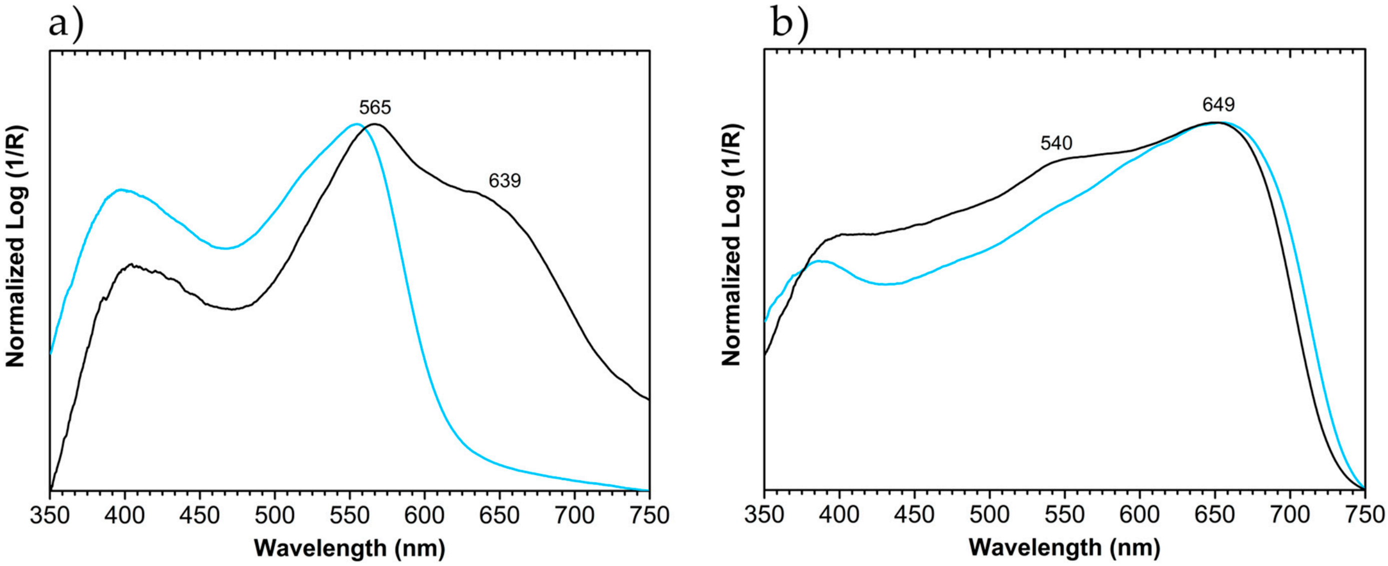

Figure S11. Apparent absorbance spectra of purple colors were found in the vestments of a) *Musicians' codex* and b) *Lapidary*. Both examples indicate the presence of brazilwood lake pigments, whose bands are shifted. The bands at 639 and 649 nm can represent the presence of indigo in the mixture, found in most purple mixtures and confirmed using Raman spectroscopy.