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New biocolourant database for the quality control of natural colourants and products containing them

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Abstract

A new digital biocolourant database in under development in BioColour project (BioColour – bio-based dyes and pigments for colour palette) funded by the Strategic Research Council at the Academy of Finland. The database supports biocolourant companies and offers quality tools also to authorities, laboratories, researchers, and all those working in the field of natural colourants and naturally coloured products. It strengthens their traceability, the production and helps to detect adulterations and to maintain high quality. To enhance the prerequisites for large-scale natural colourant production, and new applications, the database considers the entire production chain. The correct identification and thorough knowledge of the raw material is the core. Only the data from the studies with the securely identified biological source are included providing scientific pre-reviewed information about natural dye sources, their geographical origin, colourant and other secondary metabolite composition and their chemical characteristics. The database consists of two parts. The part A of the taxonomically structured database integrates information from taxonomy, botany, chemistry, and, to a lesser extent also genetics, about the dye sources, combining the strengths of each of these areas. Using a tool, based on the traditional chemotaxonomical principles, one can make queries and comparisons e.g. about dye sources, colourant molecules and their characters. The organisms, i.e. dye sources, are connected taxonomically to the part B. Using a tool, one can get information about visual colour (e.g. CIELab-values) measured from the materials after different natural dyeing processes. The traditional approaches are complemented by modern data science approaches, as a part of the prementioned data is utilized by data mining and machine learning methods to support in the quality analyses of biocolourants.

Keywords: natural dyes, natural colourants, quality, authenticity, traceability, chemotaxonomy

INTRODUCTION

Natural products, including natural colourants and products containing them, are especially vulnerable to quality problems and adulterations (e.g. Saxena and Raja, 2014). There are several reasons for such vulnerabilities. One of them is in the identification of raw material in the start of the production chain. When using natural wild materials, oppositely to a common cultivar with the known genetic background and cultivation characteristics, a source material from the wild has a local genetic and phenotypic (Linhart and Grant, 1996;

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Jaakola and Hohtola, 2010) variation, because wild plant populations are mixtures of genotypes. However, despite this infraspecific variation, the species are generally confidentially identified and characterized. Some species may be more difficult to identify e.g. due to their tendency to hybridize with genetically close relative growing in the same area, or those species have high phenotypic plasticity (e.g. *Salix* species). These factors may add risk to unintentional misidentifications leading to adulterations, in addition to consciously deliberate adulterations to achieve economic gain. Adulterations with counterfeit ingredients are commonly met in the market of natural products (Applequist & Miller 2013; Ma et al. 2013) concerning also natural colourants (e.g. Cardon, 2010; Saxena and Raja, 2014). The way of deliberate adulterations are various, common adulterations include e.g. addition of cheaper raw material, mislabeling of botanical origin (e.g. Cordella et al. 2002) and colour enhancement or masking inferior quality by adding nonauthentic substance (e.g. artificial colourant) (Moore et al. 2012). The detection of adulterations is an important part of the quality work and to protect natural colourants against adulterations, the principles and methods of authenticity and quality analyses are presented in plentiful papers (e.g. Cordella et al., 2002; Cardon, 2010; Primetta, 2014; Saxena and Raja, 2014).

To respond to the needs for the authenticity studies e.g. for the correct identification of raw material, quality development (e.g. better reproducibility) and analyses, a new digital biocolourant database is under development (https://biocolour.fi).[b] To enhance the prerequisites for large-scale production for new, or "newly found" dye sources and applications, the database provides scientific information for the identification of dye sources, characterization, and quality analyses of dye source materials, biocolourants and naturally dyed products. The correct identification of the raw material is fundamental. Classical botanical and traditional macromorphological assessment techniques represent the basis of identification and quality assessment upstream of other techniques (chemistry, genetics). It is the suite of botanical and chemical techniques that provides the great confidence for ensuring the identity and good quality of natural ingredient and product (Simmler et al., 2015; Upton et al.,2020). To start enlarged application of natural colourants a certain level of repeatability and quality level is required (Bechtold and Mussak, 2010; Primetta, 2014; Saxena and Raja, 2014). This is possible to achieve when a thorough knowledge and analyses of the relevant characteristics of the raw material are available in order to adjust the product to a fixed standard (Bechtold and Mussak, 2010; Saxena and Raja, 2014). Database brings knowledge how to protect the dye production field from fraudulent actions and to help to detect adulterations and fraudulent imitations. The aim is to support companies in the production, development, and the use of biocolourants, to offer scientific information and tools to authorities, laboratories, researchers, and all those who are working in the field of natural raw materials and naturally coloured products, especially in the quality, origin, and authenticity related questions. Database aims to aid researchers of history, archaeology, and conservation science (Cardon, 2010) bringing data about dye and other characteristic molecules typical for certain genera and species used for dyeing. It brings information about the characteristics of dye molecules, their detection and identification, as well some degradation products together with an information about the geographical origin of natural dye sources.

[b] The database will be published at the end of the year 2024. The official name will be informed during the same month.

The whole spectrum of colour can be obtained from plant, fungal and animal taxa, of which the plants are the major sources (Saxena and Raja, 2014). Natural colourants are the products of secondary metabolism of plants or animals and very diverse in terms of chemical structure. Secondary metabolites are broadly divided into two groups: nitrogen-containing molecules (alkaloids) and nitrogen-deficient molecules (terpenoids, phenolics). Phenolics can be divided into phenolic acids, tannins, and flavonoids (e.g. anthocyanins, flavonols) (Porter, 1989; Tanaka et al., 2008; Patra et al., 2013, Räisänen et al., 2016). A large part of secondary metabolites belongs to the group of phenolic compounds. During the evolution, the phenolic compounds have become progressively enriched to provide specific adaptations for different plant families (Boudet, 2007). The biosynthesis of phenolic compounds is tissue specific and developmentally regulated in different plant organs, providing a specific pattern of metabolites, characteristic for each species (chemical phenotype) (Pichersky and Gang, 2000; Vogt, 2010, Miller et al., 2011). Thus, each species displays a distinct, qualitative phenolic profile, which means the presence or absence of certain compound(s) (Macheix et al., 1990; Primetta, 2014). Quantitative variation (e.g. relative proportions) is caused by external factors, such as light intensity, temperature, and processes (Macheix et al., 1990; Jaakola and Hohtola, 2010) and there is a variation in the response between species and within individual flavonoid groups (Jaakola and Hohtola, 2010). Natural colourants exist in each species as mixtures, and most generally, they are used as mixtures. This can bring challenges to their characterization and quality control. On the other hand, this characteristic mixture can bring answers about its quality, biological source, and authenticity and sometimes about geographical origin (e. g. Primetta et al., 2013; Barbosa et al., 2020). When processing natural raw material, it has been found, that similar processes affect similar ways on the contents of phenolic compounds (e. g. Primetta, 2014). Phenolic compounds have been used for the authentication of berry and fruit cultivars, such as grapes (*Vitis* spp.) and their corresponding wines (e.g. Pomar et al., 2005; Radovanocic et al., 2010), botanical supplements, extracts (Cassinese et al., 2007; Madrigal-Carballo et al., 2009), fruit juices (e.g. Cautela et al., 2008), geographical origin of wines (e.g. Anastasiadi et al., 2009; de Andrade et al., 2013), juices (Guo et al., 2013) and spices (Barbosa et al., 2020).

Taxonomy deals with identifying, describing, and categorizing organisms from species to higher taxa (Winston, 2018). Colour compounds are secondary metabolites and can also contribute much to taxonomy and systematics (e.g. Vickery and Vickery, 1981) and thus serve as tools to quality and authenticity evaluation. Chemotaxonomy has been traditionally distinguished plants and other organisms according to the differences and similarities in their biochemical composition (e.g. Iwashina, 2000; Reynolds, 2007). Secondary metabolite profiling, including phenolic profiles, complements the taxonomic and genomic authentication of plant species (e.g. Waterman, 2007).

Database integrates information from botany, taxonomy, chemistry, and, also, but to a lesser extent, knowledge about the genetic background of dye sources. Each of the prementioned areas has its own strengths and weaknesses, what comes to quality analysis and authentication (Simmler et al., 2015; Simmler et al., 2016; Parveen et al., 2016). For example, DNA-based identification cannot differentiate parts of the sample, but their chemical composition is different. Sometimes DNA can be of poor quality or degraded (Gryson, 2010; Parveen et al., 2016), in those cases chemical analyses of characteristic profile of phytochemicals is recommended. (e.g. Ma et al., 2013). In chemical analyses, it can be sometimes found, for example. that differences are not sufficient between closely related species with the molecules studied (Pawar et al., 2017; Primetta, 2014).

MATERIALS AND METHODS

Only the published peer-reviewed studies in which an expert identifies the biological source are included in the database. The dyes sources will include the genera of historically important natural colourant sources such as common and less common species or cultivars used or known to have good dyeing potential. The database includes natural colourant sources from the wild, cultivation, and from industrial side-streams. The plant part(s) used will be reported and the date of the collecting time and a location if it has been informed in the publication.

The focus in part A (chemotaxonomy) of the database in on gathering and organising data from the scientific studies which have been used liquid chromatography coupled with MS-spectrometry or NMR. Part B of the database uses the techniques which describes how the colour looks like (e.g. in textiles) providing e.g. RGB- and CIELab-values obtained from the natural dyes and materials.

The use of multiple techniques is necessary in order to achieve the highest possible certainty level. The use of multiple markers in the authentication adds another layer of complexity and further cost to combatting fraudulent practices.

The database has taxonomical basis and data focuses at the first stage on the secondary metabolites, of them especially phenolic compounds, and then gradually botanical/mycological and genetic data and information will be added to complement. Later, also photos and microscopical pictures will be added. The strength of colourant profiles is that they give specific information for authenticity analyses, and simultaneously can work as quality meter in the process, especially when nonspecific methods are not sufficient.

RESULTS AND DISCUSSION

The database provides scientific information about natural dye sources and products, more specifically about their dye composition and the chemical characteristics of the molecules. In addition, for example, certain analytical information is included, and the geographical origin of the dye source and/or information about the voucher specimen(s) if exist in the publication. At first, the majority of the taxa is of botanical origin.

The dye compounds may have different names, as also dye sources, the database provides information about synonyms and other possible names if those have been used in the scientific studies of the related publications.

The taxonomically structured database is composed of two main parts (Figure 1). Part A provides information about organisms, and their chemical compounds, especially colourant molecules and their identification. It provides information and tools based on the traditional taxonomical and chemotaxonomical principles. Using a novel type of tool, the user can make queries and comparisons about dye sources, colourant molecules and their characters. At first, the information will be focused on the first stages of the production cycle, but gradually will proceed at the naturally coloured products themselves too. The products are first simple processed powders and liquids, naturally dyed textiles, and later also products from other fields such as from the food industry (e.g. spices), many of them are used as dyes too.

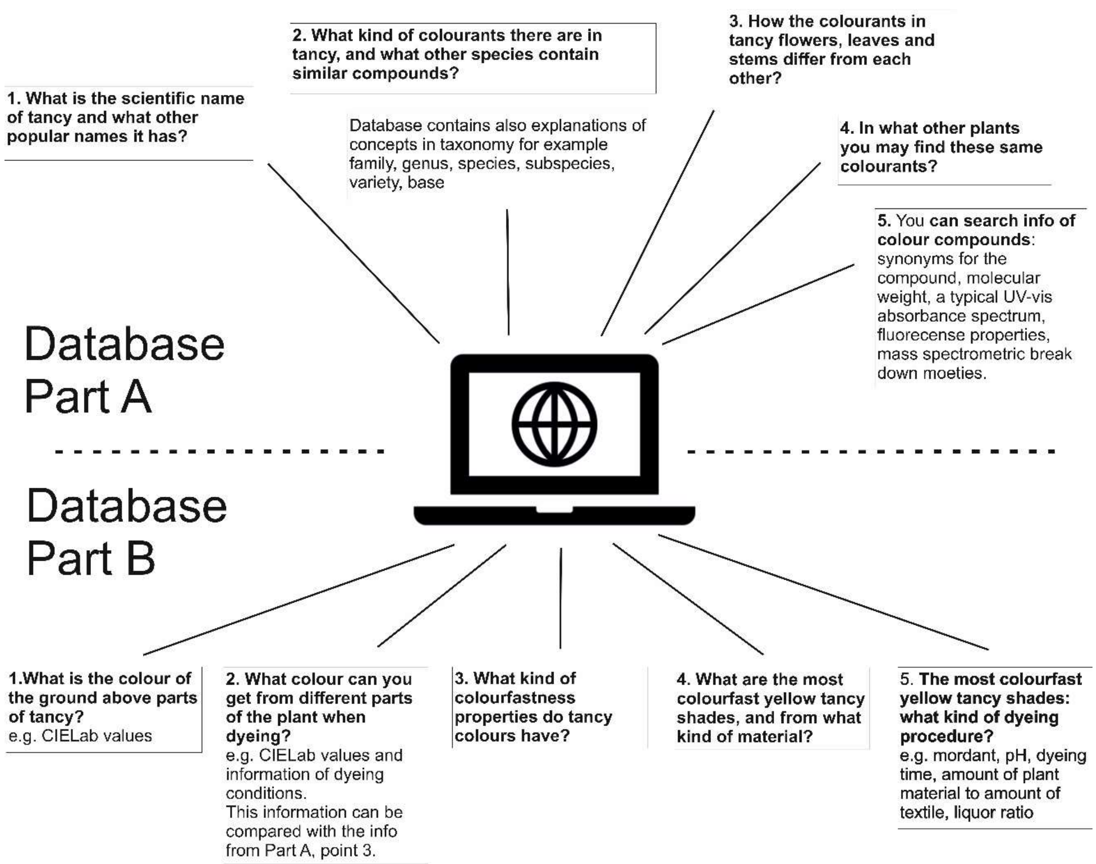

Figure 1. Structure of the biocolours' database. Tancy (*Tanacetum vulgare*) has been used as an example for the search options.

Through taxonomical basic structure, the biological sources, organisms, are connected to part B. This part gives information about visual colour (e.g. RGB-values, CIELab-values) measured from the various materials after certain dyeing processes which are described (e.g. mordant used, information about dye path). At the later phases of the database development, the pictures will available. The aim is to develop a user-friendly tool so that the information needed would be found quickly and easily. At the later phase, the traditional approaches are complemented in a module by modern data science approaches, as a part of the data is utilized by data mining and machine learning methods to support in the quality analyses of biocolourants. The database also provides information about authentication methods, adulterants, and adulteration cases.

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Literature cited

de Andrade, R.H.S., do Nascimento, L.S., Pereira, G.E., Hallwass, F., and Paim, A.P.S. (2013). Anthocyanin composition of Brazilian red wines and use of HPLC-UV–Vis associated to chemometrics to distinguish wines from different regions. Microchem. J. 110, 256–262.

Applequist, W. and Miller, J.S. (2013). Selection and authentication of botanical materials for the development of analytical methods. Anal. Bioanal. Chem. 405, 4419–4428. https://doi.org/10.1007/s00216-012-6595-1

Barbosa, S., Campmajó, G., Saurina, J., Puignou, L., and Núñez, O. (2020). Determination of phenolic compounds in paprika by ultrahigh performance liquid chromatography-tandem mass spectrometry: application to product designation of origin authentication by chemometrics. J. Agric. Food Chem. 68, 591–602. https://doi.org/10.1021/acs.jafc.9b06054

Bechtold, R., and Mussak, R., eds. (2010) Handbook of natural colorants. Wiley Series in Renewable Resources. (West Sussex, UK: A John Wiley and Sons, Ltd.), pp. 412.

Boudet, A.M. (2007). Evolution and current status of research in phenolic compounds. Phytochem. 68, 2722–2735. https://doi.org/10.1016/j.phytochem.2007.06.012

Cautela, D., Laratta, B., Santelli, F., Trifirò, A., Servillo, L., and Castaldo, D. (2008). Estimating bergamot juice adulteration of lemon juice by high-performance liquid chromatography (HPLC) analysis of flavanone glycosides. J. Agric. Food Chem. 56, 5407–5414. https://doi.org/10.1021/jf8006823

Cardon, D. (2007). Natural dyes. Sources, tradition, technology, and science. (London: Archetype Publications Ltd.), pp. 804.

Cardon, D. (2010). Natural dyes, our global heritage of colours. Textile Society of America 12th Biennial Symposium, Lincoln, Nebraska Textile Society of America.

Cassinese, C., de Combarieu, E., Falzoni, M., Fuzzati, N., Pace, R., and Sardone, N. (2007). New liquid chromatography method with ultraviolet detection for analysis of anthocyanins and anthocyanidins in *Vaccinium myrtillus* fruit dry extracts and commercial preparations. J. AOAC Int. 90, 911–919.

Cooper-Driver, G. A., and Bhattacharya, M. (1998). Role of phenolics in plant evolution. Phytochem. 49, 1165–1174. https://doi.org/10.1002/chin.199910249

Cordella, C., Moussa, I., Martel, A.-C., Sbirrazzuoli, N., and Lizzani-Cuvelier, L. (2002). Recent developments in food characterization and adulteration detection: technique-oriented perspectives. J. Agric. Food Chem. 50, 1751–1764. https://doi.org/10.1021/jf011096z

Gryson N. (2010). Effect of food processing on plant DNA degradation and PCR-based GMO analysis: a review. Analytical and Bioanalytical Chemistry, 396, 2003–2022. https://doi.org/10.1007/s00216-009-3343-2

Guo, J., Yue, T., Yuan, Y., and Wang, Y. (2013). Chemometric classification of apple juices according to variety and geographical origin based on polyphenolic profiles. J. Agric. Food Chem. 61, 6949–6963. https://doi.org/10.1021/jf4011774

Iwashina, T. (2000). The structure and distribution of the flavonoids in plants. J. Plant Res. 113, 287–299. https://doi.org/10.1007/PL00013940

Jaakola, L., and Hohtola, A. (2010). Effect of latitude on flavonoid biosynthesis in plants. Plant, Cell & Environ. 33, 1239–1247. https://doi.org/10.1111/j.1365-3040.2010.02154.x

Linhart, Y.B., and Grant, M.C. (1996). Evolutionary significance of local genetic differentiation in plants. Annu. Rev. Ecol. Syst. 27, 237–277. https://doi.org/10.1146/annurev.ecolsys.27.1.237

Ma, Y.-C., Chen, S.-L., Thibault, M.E., and Ma, J. (2013). Enhancing quality control of botanical medicine in the 21st century from the perspective of industry: the use of chemical profiling and DNA barcoding to ensure accurate identity. HerbalGram 58–67.

Macheix, J.-J., Fleuriet, A., and Billot, J. (1990) Fruit phenolics. (Boca Raton, Florida: CRC Press), p. 390.

Madrigal-Carballo, S., Rodriguez G., Krueger, C.G., Dreher, M., and Reed, J.D. (2009). Pomegranate *(Punica granatum)* supplements: authenticity, antioxidant, and polyphenol composition. J. Funct. Foods 1, 324–329. https://doi.org/10.1016/j.jff.2009.02.005

Miller, R., Owens, S.J., and Rørslett, B. (2011). Plants and colour: Flowers and pollination. Opt. Laser. Technol. 43, 282–294. https://doi.org/10.1016/j.optlastec.2008.12.018

Moore, J. C., Spink, J., & Lipp, M. (2012). Development and application of a database of food ingredient fraud and economically motivated adulteration from 1980 to 2010. J. Food Sci. 77, R118–R126. https://doi.org/10.1111/j.1750-3841.2012.02657.x

Parveen, I., Gafner, S., Techen, N., Murch, S. J., and Khan, I. A. (2016). DNA barcoding for the identification of botanicals in herbal medicine and dietary supplements: strengths and limitations. Planta Med. 82, 1225–1235. https://doi.org/10.1055/s-0042-111208

Patra, B., Schluttenhofer, C., Wu, Y., Pattanaik, S., and Yuan, L. (2013). Transcriptional regulation of secondary metabolite biosynthesis in plants. Biochim. Biophys. Acta, 1829, 1236–1247. https://doi.org/10.1016/j.bbagrm.2013.09.006

Pawar, R. S., Handy, S. M., Cheng, R., Shyong, N., & Grundel, E. (2017). Assessment of the authenticity of herbal dietary supplements: comparison of chemical and DNA barcoding methods. Planta Med. 83, 921–936. https://doi.org/10.1055/s-0043-107881

Pichersky, E., and Gang, D. R. (2000). Genetics and biochemistry of secondary metabolites in plants: an evolutionary perspective. Trends Plant Sci. 5, 439–445. https://doi.org/10.1016/s1360-1385(00)01741-6

Pomar, F., Novo, M., and Masa, A. (2005). Varietal differences among the anthocyanin profiles of 50 red table grape cultivars studied by high performance liquid chromatography. J. Chromatogr. A, 1094, 34–41. https://doi.org/10.1016/j.chroma.2005.07.096

Porter, L. J. (1989) Tannins. 11. In: Plant phenolics. Vol 1. Harborne, J. B., ed. (New York and London: Academic Press), pp. 608.

Primetta, A. K., Jaakola, L., Ayaz, F. A., Inceer, H., and Riihinen, K. R. (2013). Anthocyanin fingerprinting for authenticity studies of bilberry (*Vaccinium myrtillus* L.). Food Control 30, 662–667. https://doi.org/10.1016/j.foodcont.2012.09.009.

Primetta, A. (2014). Phenolic compounds in the berries of the selected *Vaccinium* species. The potential for authenticity analyses. Publications of the University of Eastern Finland. Dissertations in Forestry and Natural Sciences, 134. pp. 104 + append. https://erepo.uef.fi/handle/123456789/12896?locale-attribute=en

Radovanović, B. C., Radovanović, A. N., and Souquet, J. M. (2010). Phenolic profile and free radical-scavenging activity of Cabernet Sauvignon wines of different geographical origins from the Balkan region. J. Sci. Food Agric. 90, 2455–2461. https://doi.org/10.1002/jsfa.4106

Räisänen, R., Primetta, A., and Niinimäki, K. (2016) Dyes from nature. (London: Archetype Publications Ltd.), pp. 284.

Saxena, S., and Raja, A.S.M. (2014). Natural dyes: sources, chemistry, application, and sustainability issues. In: Muthu, S.S., ed. Roadmap to sustainable textiles and clothing technology. Textile Science and Clothing Technology. (Singapore: Springer), 37–80. https://doi.org/10.1007/978-981-287-065-0_2

Simmler, C., Chen, S. N., Anderson, J., Lankin, D. C., Phansalkar, R., Krause, E., Dietz, B., Bolton, J. L., Nikolic, D., van Breemen, R. B., and Pauli, G. F. (2015). Botanical integrity: the importance of the integration of chemical, biological, and botanical analyses, and the role of DNA barcoding. HerbalGram, 106, 58–60.

Simmler, C., Chen, S. N., Anderson, J., Lankin, D. C., Phansalkar, R., Krause, E., Dietz, B., Bolton, J. L., Nikolic, D., van Breemen, R. B., and Pauli, G. F. (2016). Botanical integrity: part 2: traditional and modern analytical approaches. HerbalGram, 109, 60–64.

Tanaka, Y., Sasaki, N., and Ohmiya, A. (2008). Biosynthesis of plant pigments. anthocyanins, betalains and carotenoids. Plant J. 54, 733–749. https://doi.org/10.1111/j.1365-313X.2008.03447.x

Upton, R., David, B., Gafner, S., and Glasl, S. (2020). Botanical ingredient identification and quality assessment: strengths and limitations of analytical techniques. Phytochem. Rev. 19, 1157–1177. https://doi.org/10.1007/s11101-019-09625-z

Vickery M.L., and Vickery, B. (1981) Secondary plant metabolism. (London: The Macmillan Press Ltd.), pp. 335.

Vogt T. (2010). Phenylpropanoid biosynthesis. Mol. Plant, 3, 2–20. https://doi.org/10.1093/mp/ssp106

Winston, J. E. (2018). Twenty-first century biological nomenclature — the enduring power of names. Integr. Comp. Biol., 58, 1122–1131. https://doi.org/10.1093/icb/icy060