Studies on *Origanum vulgare* L. as a traditional dye plant in Latvia

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- University of Latvia research project no. ZD2015/AZ85 'The territory of Latvia as a zone of contact between different cultural spaces, religions, and political, social and economic interests from prehistory up to the present day'
- L`ORÉAL Baltic Fellowship for Women in Science 2018 awarded project 'Determination of pigments used in the traditional dyeing processes of textiles using chromatography'

LATVIJAS UNIVERSITATE
ANNO 1919
UNIVERSITY OF LATVIA

INSTITUTE FOR ENVIRONMENTAL SOLUTIONS

Origanum vulgare L.





Sourses for the Reseach:

- Latvian folklore and linguistic evidences;
- Ethnographic descriptions;
- Reconstruction of dyeing methods;
- Chemical analysis of mass spectrometry, determining the content of pigments in:
  - *Origanum vulgare* L. dyeing solution,
  - samples of textiles dyed nowadays.

Origanum vulgare L.

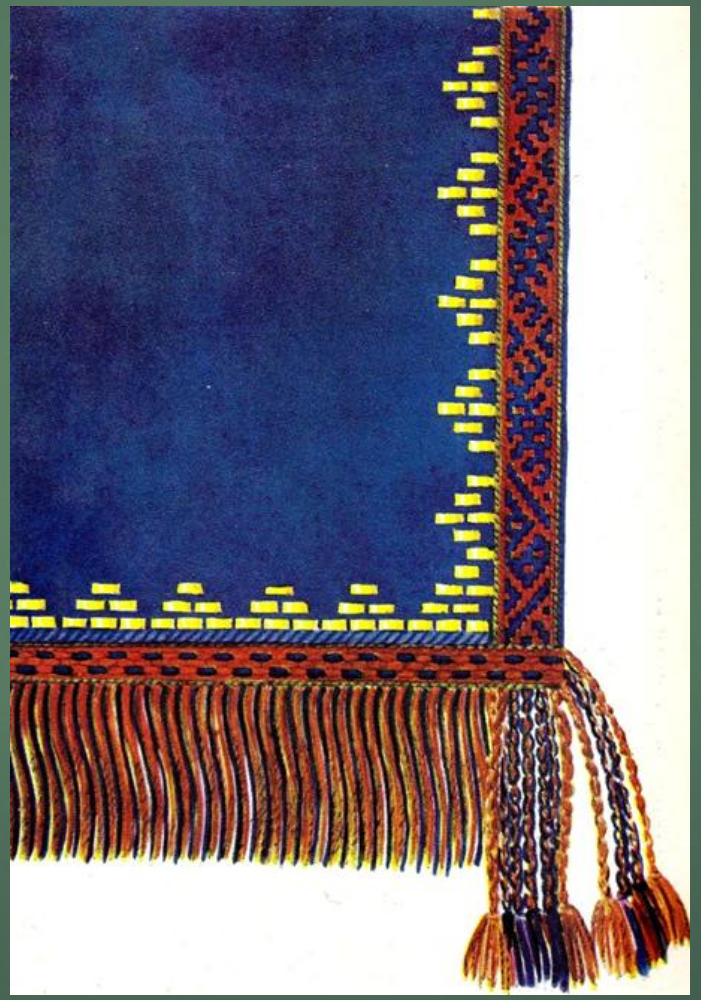
- In Latvian literary language: parastā **raudene**; (from Latvian ‘ruds‘ – redish, ginger, rusty)
- in folk tradition (some of used names):
  - **sarkanes** (from Latvian ‘sarkans’ – red)
  - **dzīpari** (old Latvian name for fine, coloured yarn, usually used for embroidery)
  - **sarkanā rauda** (the red ‘rauda’, when ‘rauda’=‘ruds’ - redish/ginger/ rusty)

Dye plants, mentioned in Latvian folk songs:

- ‘mēles’ (krāsu mēle - Woad *Isatis tinctoria* L.) for dark blue (Latvian = mēļš) colour;
- ‘madaras’ (madaras – Bedstrows *Galium* L.) roots, for red colour;
- ‘alkšņi’ (alksņi – Alders *Alnus* L.) for dark or black colour;
- ‘bērzs’ (bērzs – birch Betula sp.) leaves, without mention colour;
- ‘dzeltes’ (‘dzeltens’ – yellow) (staipekņi – *Licopodium* sp.) without mention colour;
- **‘sarkane’ (parastā raudene – *Oreganum vulgare* L.) for red colour**

Reconstructions of Baltic tribe's (Latgalian) dress of the13th century







Dyeing recipes in ethnographic descriptions

- In total are known 26 descriptions, where is mentioned dyeing with *Origanum vulgare* L.;
- They are fixed from 1876 till 1942;
- 10 of them are unpublished sources - expedition records;
- 16 are publications of ethnographic material, 7 of which are first-time information publications



The colours, got from *Origanum vulgaris* L. in dyeing with heating (without fermentation)



Information about dyeing

- Dyeing recipes or indications of the dyeing process with *Origanum vulgare* L. in the ethnographic materials are available in 8 records;
- Fermentation is mentioned in 4 cases;
- Apple (*Malus* spp.) leaves are mentioned together with *Oreganum vulgare* L. in 4 recipes;
- Colours mentioned in the descriptions are: **red**, **dark red**, **bluish ('cold') red** and **brown**.

Dyeing recepieces in ethnographic descriptions:

- **1876** (published 1885)

The red yarn was dyed in 'sarkanes' (flower species) and apple leaves. Apple leaves were gathered, especially those with red stems, and 'sarkanes' - before blossoming. Then they were both finely chopped and moistened with lukewarm water, pressed the liquid out and put them on a cloth in the sun until they became reddish. Then placed them in a pot, poured on warm water, added yeast, and fermented for a day or more; then put in a well-washed white woollen yarn and fermented for another three days. After that, he poured the fermented liquid with all the yarn in a pot and cooked for a long time. Then took the yarn out, shook out parts of the plants and dried the yarn in the sun.

(P.Blaus (manuscript))

Dyeing with *Origanum vulgare* L. and *Malus* spp. with fermentation

EXPERIMENT









Dyeing with *Origanum vulgare* L. and *Malus* spp. with fermentation

EXPERIMENT







Dyeing with *Origanum vulgare* L. and *Malus* spp. with fermentation

EXPERIMENT



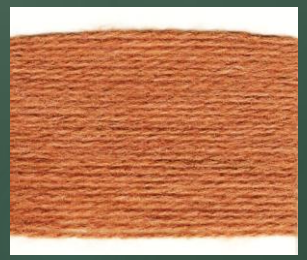








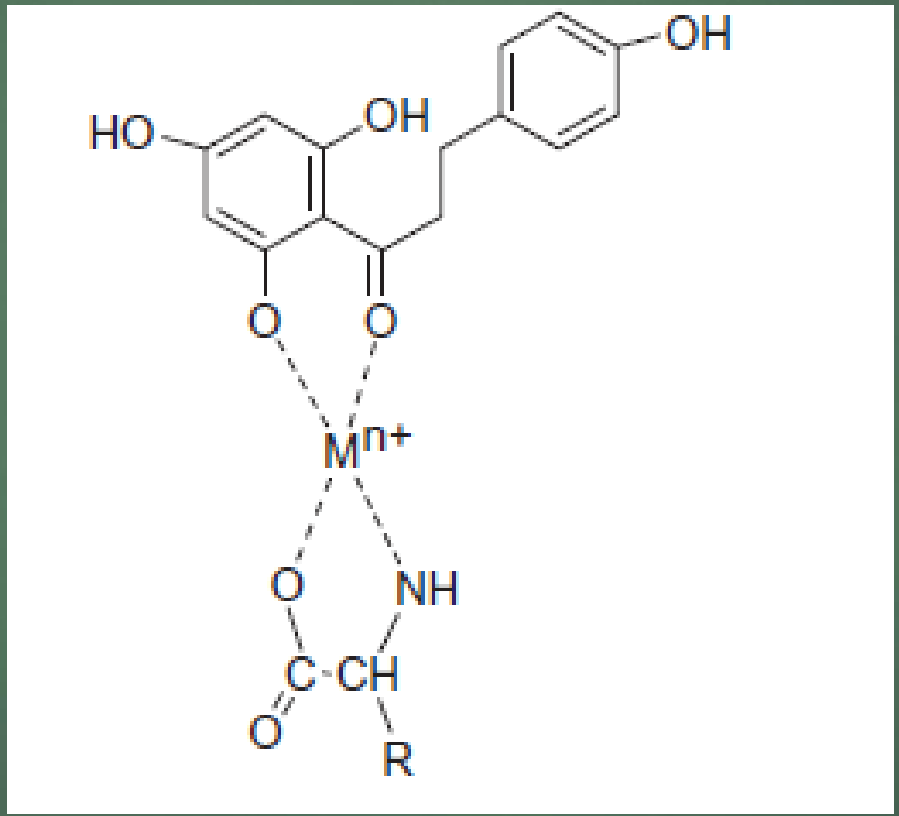
Mordanted with alumn and tartar



Without mordant

Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

EXTRACTION



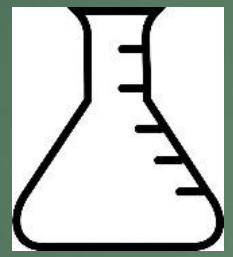
Proposed structure of metal complex that may occur between main colorant of apple leaves extract and wool.

F. ESER ET AL. JOURNAL OF NATURAL FIBERS, Volume 13, 2016 –Issue 3

Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

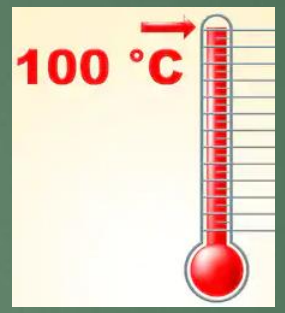
EXTRACTION





400 µL
HCl :MeOH:H_2O
(2:1:1)



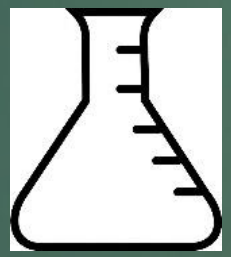


Heat at 100 C
for 10 min

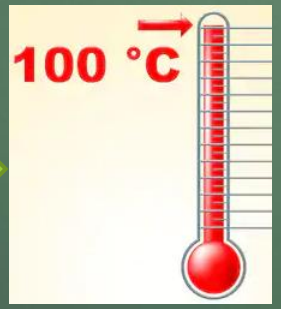


Centrifugate at
10000 rpm
for 5min

N_2

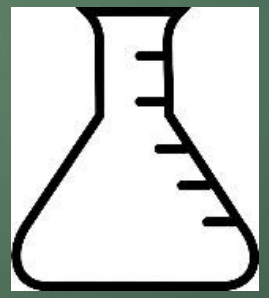


400 µL
MeOH:DMSO (1:1)



Heat at 100 C
for 5 min

N_2



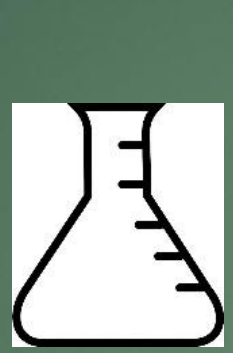
150 µL
MeOH



Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

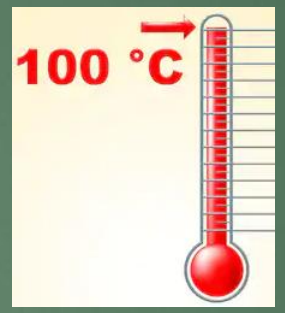
EXTRACTION





400 µL
HCl :MeOH:H_2O
(2:1:1)





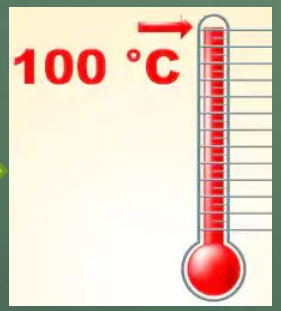
Heat at 100 C
for 10 min



Centrifugate at
10000 rpm
for 5min

N_2

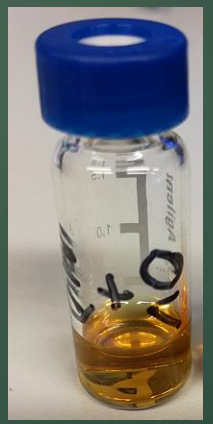
400 µL
MeOH:DMSO (1:1)



Heat at 100 C
for 5 min

N_2

150 µL
MeOH



Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

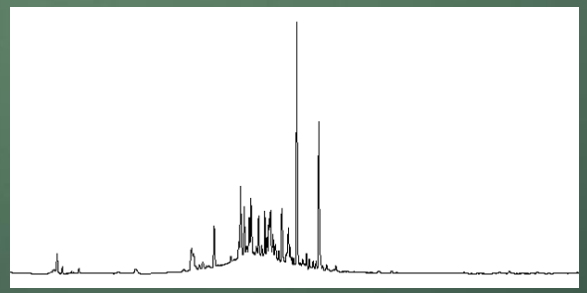
EXPERIMENTAL



Agilent 1290

Column: Zorbax Extend C18 RRHD 1.8 µm (3.0 x 150 mm)







Agilent 6230 TOF

Mobile Phase A: 0.5% FA in water
Mobile Phase B: ACN
Flow rate: 0.3 mL/min
Injection volume: 20 µL
Column temperature: 60 °C
DAD detection: 220, 280, 320, 340 and 520 nm

Ionization: (ESI+)
Mass range: m/z 75 - 1500

Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.





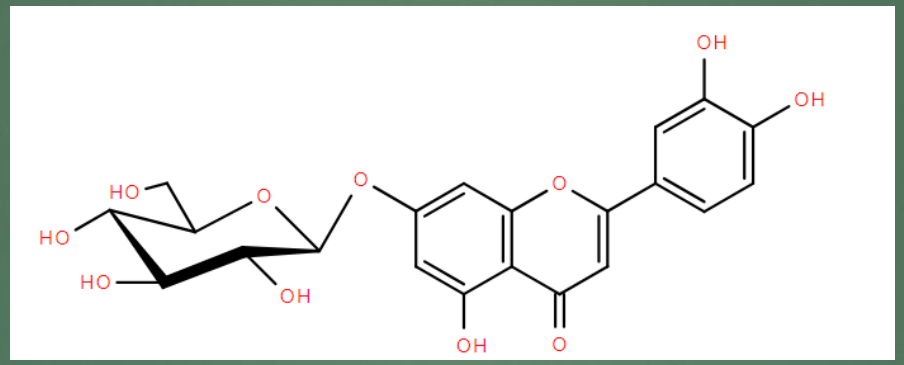
RESULTS

Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

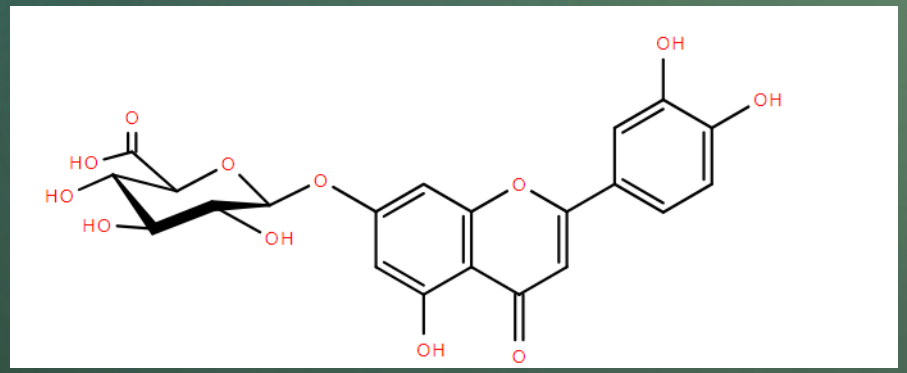
RESULTS

Luteolin type

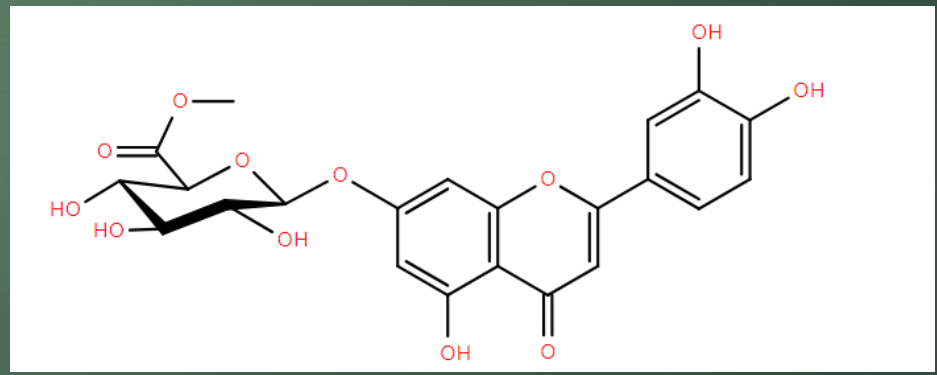
Luteolin 7-glucoside
m/z 449



Luteolin 7-O-glucuronide
m/z 463



Luteolin 7-methylglucuronide
m/z 477

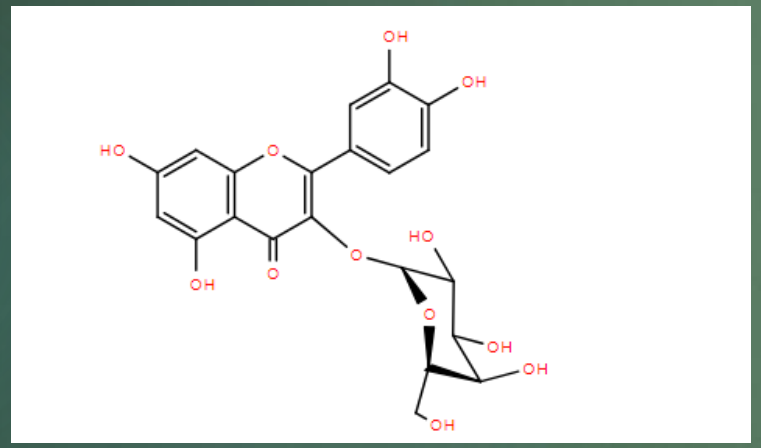


Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

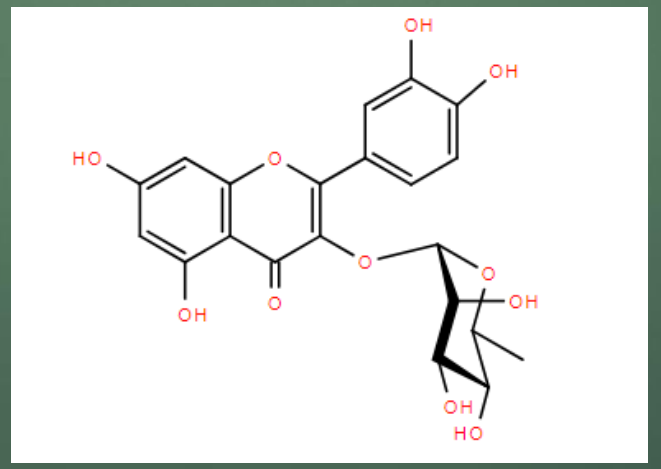
RESULTS

Quercetin type

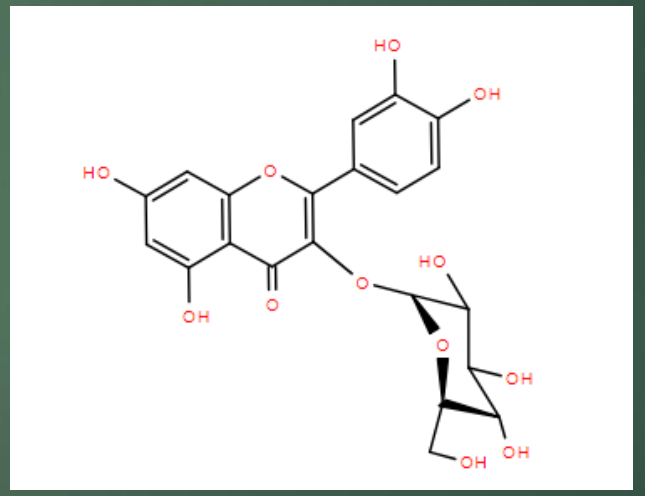
Quercetin-3-galactoside
m/z 465



Quercetin-3-rhamnoside
m/z 449



Quercetin-3-glucoside
m/z 465

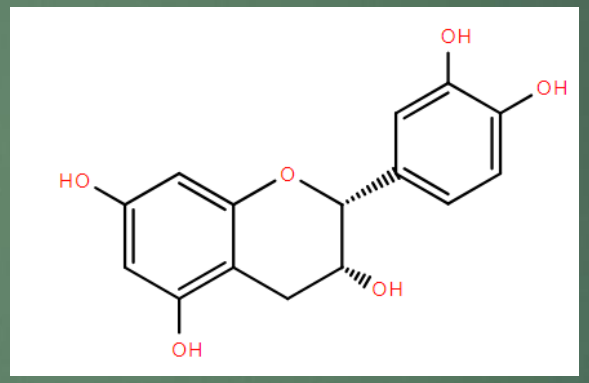


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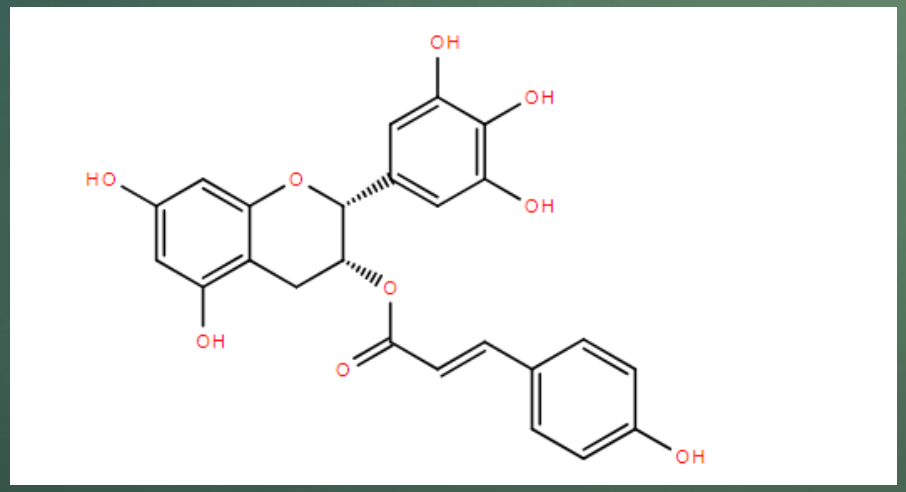
RESULTS

Flavan-3-ols

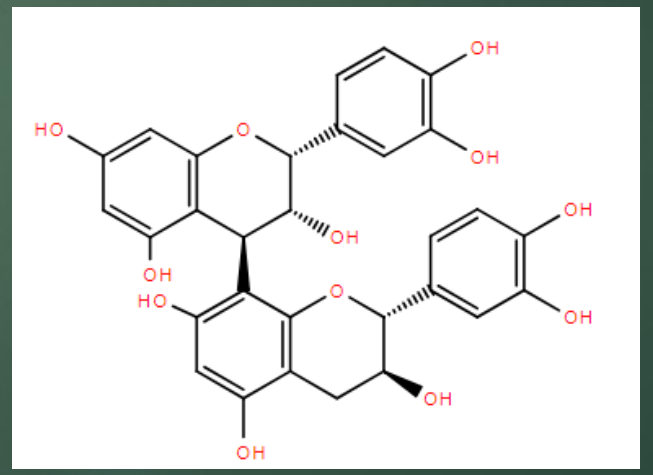
Epicatechin
m/z 291



Epigallocatechin 3-O-p-coumarate
m/z 453



Procyanidin B1
m/z 579



Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.

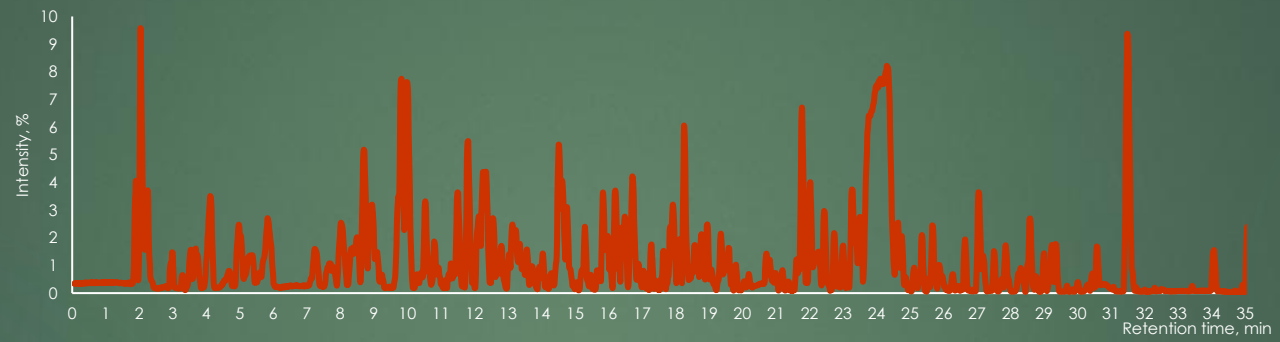
RESULTS



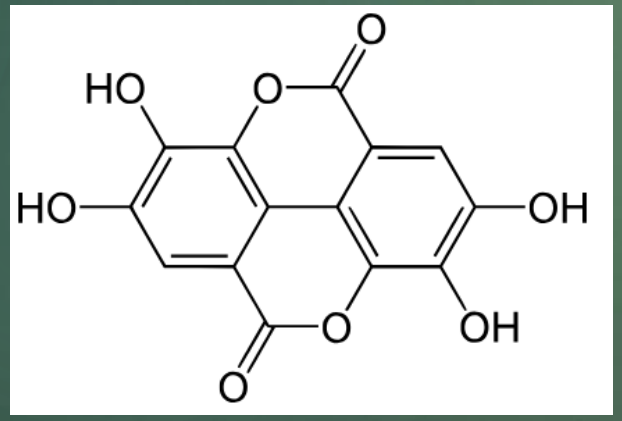
Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.



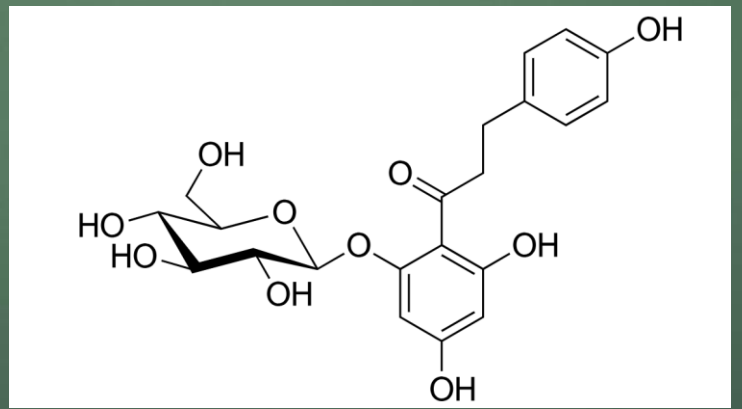
RESULTS



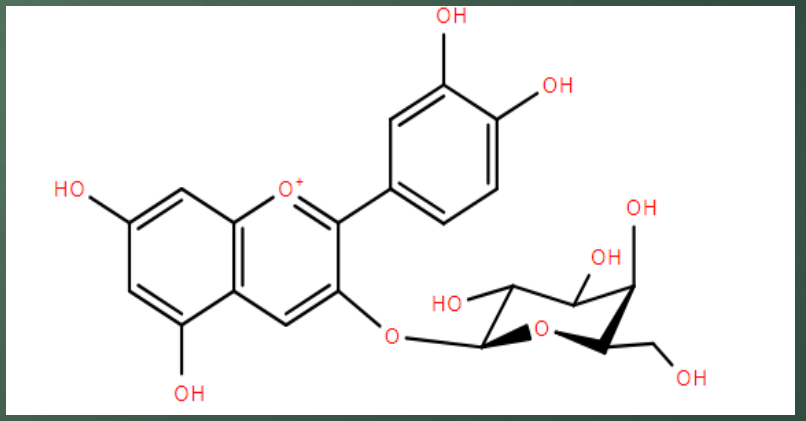
Ellagic acid
m/z 303



Phloridzin
m/z 437



Cyanidin 3-O-galactoside
m/z 449

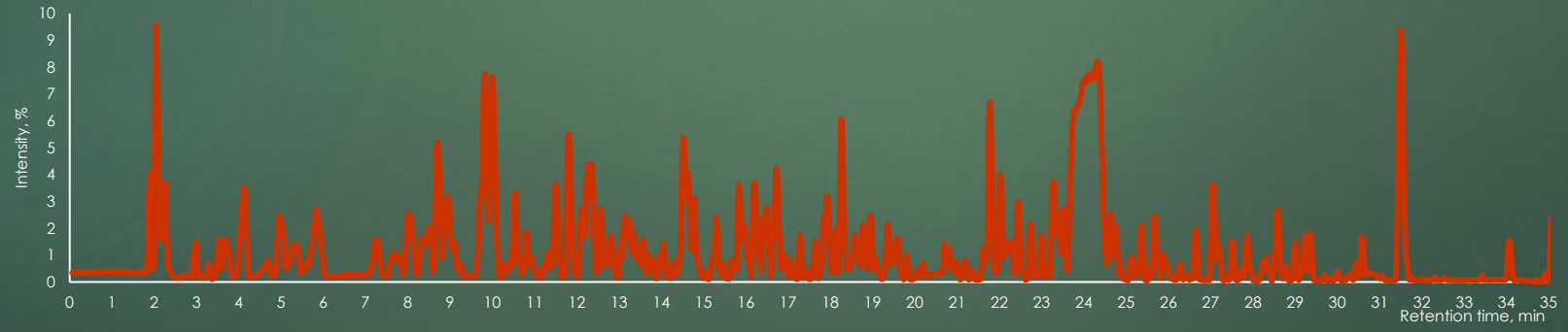


Chromatographic Investigation of Wool Dyed with *Origanum vulgare* L. and *Malus* spp.



CONCLUSIONS

- Extraction using hydrochloric acid/methanol and liquid chromatography coupled with high resolution mass spectrometry facilitated the effective separation and identification of pigments presented in the wool dyed with *Origanum vulgare* L. and *Malus* spp.
- Separated compounds indicate the presence of yellow, brown and some red pigments in wool extract.





Thank You for your attention!