Anthocyanins

The identification of anthocyanins was performed by comparing the retention times, the UV-Vis and MS^n data of each peak with those of the pure standards (when commercially available) run under the same chromatographic conditions.

The anthocyanins identified in grape samples were principally the mono-hexosides of five anthocyanidins (e.g. delphinidin, cyanidin, petunidin, peonidin and malvidin) and their corresponding derivatives, including acetyl, coumaroyl and caffeoyl esters, as also reported by previous studies (Table 1S).[1] In the MS analysis anthocyanins gave the $[M^+-2H]^-$ ion and the water adduct $[M^+-2H+H_2O]^-$ ion. The difference between the MS^2 main fragment and $[M^+-2H]^-$ ion allowed the determination of the sugar molecular weight. Furthermore, MS^3 scan provided the specific fragmentation pattern of the aglycone.

Malvidin-3-*O*-glucoside standard (n. **18**) is associated with the peak having a retention time of approximately 16 minutes (not shown). The mass spectrum at this retention time showed a $[M^+-2H]^-$ ion at *m/z* 491 and a water adduct $[M^+-2H+H_2O]^-$ ion at *m/z* 509. An additional peak with *m/z* 537 was detected; since the difference with $[M^+-2H]^-$ ion of malvidin-3-glucoside (*m/z* 491) was 46 amu (atomic mass unit), this was attributed to a formate adduct $[M^+-2H+HCOOH]^-$ ion.

Six different peaks (n. **2-7**) were attributed to malvidin conjugates considering that: 1) a fragment with *m/z* 329, corresponding to the malvidin aglycone, was present in their MS^2 spectra, and 2) an absorbance maximum was observed at 520 nm in their UV spectra. Compounds **2-3** showed the $[M^+-2H]^-$ ion at *m/z* 653. The difference between the $[M^+-2H]^-$ ion and the fragment ion corresponding to the aglycone (*m/z* 329) was 324 amu. When some compounds had the same molecular weight and frammentation pattern, the tentative identification was performed on the basis of their chromatographic behaviour (retention times).

Some anthocyanins were characteristic of specific varieties, such as delphinidin-acetyl-hexoside (n. **12**), petunidin-acetyl-hexoside (n. **20**) and petunidin-caffeoyl-hexoside (n. **19**).

Table 1S. Identification of 25 anthocyanins in red grape samples by HPLC-DAD-ESI-MS^n

n.	Time (min)	λ max (nm)	MW	MS (m/z)	MS^2 (m/z)	MS^3 (m/z)	Tentative identification
1	16.17	520	493	491 $[M-2H]^-$ 509 $[M-2H+H_2O]^-$ 537 $[M-2H+HCOOH]^-$	329	299, 314	Malvidin-3-*O*-glucoside*
2	27.33	520	655	653 $[M-2H]^-$ 671 $[M-2H+H_2O]^-$	329	299, 314	Malvidin-caffeoyl-hexoside
3	31.11	520	655	653 $[M-2H]^-$ 671 $[M-2H+H_2O]^-$	329	299, 314	Malvidin-caffeoyl-hexoside
4	30.89	520	639	637 $[M-2H]^-$ 655 $[M-2H+H_2O]^-$	329	299, 314	Malvidin-cumaroyl-hexoside
5	34.51	520	639	637 $[M-2H]^-$ 655 $[M-2H+H_2O]^-$	329	299, 314	Malvidin-cumaroyl-hexoside
6	23.11	520	535	533 $[M-2H]^-$	329	299, 314	Malvidin-acetyl-hexoside

				551 $[M-2H+H_2O]^-$			

Table 1S. Identification of 25 anthocyanins in red grape samples by HPLC-DAD-ESI-MS^n (*continue*)

n.	Time (min)	λ max (nm)	MW	MS (m/z)	MS^2 (m/z)	MS^3 (m/z)	Tentative identification
7	27.73	520	535	533 $[M-2H]^-$ 551 $[M-2H+H_2O]^-$	329	299, 314	Malvidin-acetyl-hexoside
8	9.84	520	465	463 $[M-2H]^-$ 481 $[M-2H+H_2O]^-$	301	257, 149, 239	Delphinidin-3-*O*-glucoside*
9	25.86	520	611	609 $[M-2H]^-$ 627 $[M-2H+H_2O]^-$	301	257, 149, 239	Delphinidin-coumaroyl-hexoside
10	17.60	520	627	625 $[M-2H]^-$	301	257, 149, 239	Delphinidin-caffeoyl-hexoside
11	23.20	520	627	625 $[M-2H]^-$	301	257, 149, 239	Delphinidin-caffeoyl-hexoside
12	17.79	520	507	505 $[M-2H]^-$ 523 $[M-2H+H_2O]^-$	301	257, 149, 239	Delphinidin-acetyl-hexoside
13	12.25	520	449	447 $[M-2H]^-$ 465 $[M-2H+H_2O]^-$	285	241	Cyanidin-3-*O*-glucoside*
14	27.82	520	595	593 $[M-2H]^-$ 611 $[M-2H+H_2O]^-$	285	241	Cyanidin-coumaroyl-hexoside
15	30.92	520	595	593 $[M-2H]^-$ 611 $[M-2H+H_2O]^-$	285	241	Cyanidin-coumaroyl-hexoside
16	13.55	520	479	477 $[M-2H]^-$ 495 $[M-2H+H_2O]^-$	315	271/273, 247, 229, 256	Petunidin-3-*O*-glucoside*
17	28.41	520	625	623 $[M-2H]^-$ 641 $[M-2H+H_2O]^-$	315	271/273, 247, 229, 256	Petunidin-coumaroyl hexoside
18	30.94	520	641	639 $[M-2H]^-$ 657 $[M-2H+H_2O]^-$	315	271/273, 247, 229, 256	Petunidin-caffeoyl-hexoside
19	34.43	520	641	639 $[M-2H]^-$ 657 $[M-2H+H_2O]^-$	315	271/273, 247, 229, 256	Petunidin-caffeoyl-hexoside
20	20.38	520	521	519 $[M-2H]^-$ 537 $[M-2H+H_2O]^-$	315	271/273, 247, 229, 256	Petunidin-acetyl-hexoside
21	15.59	520	463	461 $[M-2H]^-$ 479 $[M-2H+H_2O]^-$	299	-	Peonidin-3-*O*-glucoside*
22	30.95	520	609	607 $[M-2H]^-$ 625 $[M-2H+H_2O]^-$	299	-	Peonidin-coumaroyl- hexoside
23	35.17	520	609	607 $[M-2H]^-$ 625 $[M-2H+H_2O]^-$	299	-	Peonidin-coumaroyl- hexoside
24	27.12	520	625	623 $[M-2H]^-$ 641 $[M-2H+H_2O]^-$	299	-	Peonidin-caffeoyl-hexoside
25	23.03	520	505	503 $[M-2H]^-$ 521 $[M-2H+H_2O]^-$	299	-	Peonidin-acetyl-hexoside

* These compounds were confirmed with a standard commercially available

Flavan-3-ols and procyanidins

Table 2S reports the flavan-3-ols identified in grape samples. The monomers epicatechin (EC), epigallocatechin-3-gallate (EGCG), Epicatechin gallate (ECG), and epigallocatechin (EGC) (n. **27-30**) in grape samples were investigated by comparison with the commercial standards.

Two peaks showed the $[M-H]^-$ ion at *m/z* 289 with retention time of aproximately 16 (n. **26**) and 20 minutes (n. **27**). Peak n. **27** was identified as (-)-epicatechin for comparison with the commercial standard. Therefore, considering the MS^2 and MS^3 pattern, the compound n. **26** was associated to (+)-catechin (C).

Catechin or epicatechin fragment ion at *m/z* 245 was produced by the loss of a CH_2OH group as described by Pérez-Magariño;[2] the mechanism of fragment ions *m/z* 179 and 205 production has been previously described in the literature.[3,4]

Regarding the dimers of flavan-3-ols, the only commercial standard used was procyanidin B_2 (EC and C dimer). This compound (n. **34**) was identified both in wine and in table grape varieties. Other peaks characterized by the same molecular weight and fragmentation pattern (n. **31-34**) were detected in different samples. These peaks presented the $[M-H]^-$ ion *m/z* 577 and showed a considerable fragmentation with the elimination of 152 amu (Figure 1) (characteristic fragmentation pathway by retro Diels-Alder reaction),[5] and were identified as (epi)catechin dimers. Compounds **35-38** showed $[M-H]^-$ ion at *m/z* 729 and their fragmentation pattern was similar to that of procyanidin B_2. Considering the molecular weight, they were attributed to different proanthocyanidin dimer-gallate. Compounds **39-43**, characterized by different retention times (8.24, 15.66, 17.34, 18.14 and 21.96 minutes) showed $[M-H]^-$ ion at *m/z* 865 and were identified as proanthocyanidin trimers. The sequence was identified as (epi)catechin trimers. The structural analysis of proanthocyanidins was consistent with previously reported data for *V. vinifera* L. grape varieties.[6]

Table 2S. Identification of flavan-3-ols in grape samples by HPLC-DAD-ESI-MS^n

n.	Time (min)	λ max (nm)	MW	MS (m/z)	MS^2 (m/z)	MS^3 (m/z)	Tentative identification
26	16.35	279	290	289 $[M-H]^-$	**245**, 205, 179	161, 203	C (catechin)
27	20.10	278	290	289 $[M-H]^-$	**245**, 205, 179	161, 203	EC (epicatechin)*
28	15.07	270	306	305 $[M-H]^-$	**179**, 219	137	EGC (epigallocatechin)*
29	27.76	276	442	441 $[M-H]^-$	**289**, 169, 331, 305	245, 205	ECG (epicatechin gallate)*
30	21.18	273	458	457 $[M-H]^-$	**169**, 305, 331	125	EGCG (epigallocatechin gallate)*
31	14.91	279	578	577 $[M-H]^-$	289, **407**, 425, 451	285, 297	Proanthocyanidin dimer
32	15.81	279	578	577 $[M-H]^-$	289, **407**, 425, 451	285, 297	Proanthocyanidin dimer
33	18.48	279	578	577 $[M-H]^-$	289, **407**, 425, 451	285, 297	Proanthocyanidin dimer
34	19.04	279	578	577 $[M-H]^-$	289, **407**, 425, 451	285, 297	**Procyanidin B2** dimer *
35	21.47	274	730	729 $[M-H]^-$	407, 559, **577**	407, 289, 203	Proanthocyanidin dimer (-gallate)
36	23.59	274	730	729 $[M-H]^-$	**407,** 559, 577	285, 297	Proanthocyanidin dimer (-gallate)
37	28.64	274	730	729 $[M-H]^-$	**407**, 559, 577	285, 297	Proanthocyanidin dimer (-gallate)
38	33.05	274	730	729 $[M-H]^-$	**407**, 559, 577	285, 297	Proanthocyanidin dimer (-gallate)
39	8.24	279	866	865 $[M-H]^-$	**695**, 451, 577, 407	525, 407, 451, 543	Proanthocyanidin trimer

40	15.66	279	866	865 [M-H]$^-$	**695**, 451, 577, 407	525, 407, 451, 543	Proanthocyanidin trimer
41	17.34	279	866	865 [M-H]$^-$	**695**, 451, 577, 407	525, 407, 451, 543	Proanthocyanidin trimer
42	18.14	279	866	865 [M-H]$^-$	**695,** 451, 577, 407	525, 407, 451, 543	Proanthocyanidin trimer
43	21.96	279	866	865 [M-H]$^-$	**695**, 451, 577, 407	525, 407, 451, 543	Proanthocyanidin trimer

* These compounds were confirmed with a standard commercially available

In bold the ions used for MS^3 fragmentation

Flavonols

The following commercial standards were used for identifying the most abundant flavonols: quercetin (n. **44**), quercetin-3-*O*-galactoside (n. **45**), quercetin-3-*O*-glucoside (n. **46**), quercetin-3-*O*-glucuronide (n. **47**), quercetin-3-*O*-rutinoside (n. **48**), kaempferol (n. **49**) and kaempferol-3-*O*-glucoside (n. **51**). Generally in the MS analysis flavonols gave the $[M^+-H]^-$ ion. The difference between the MS^2 main fragment and $[M^+-H]^-$ ion allowed the determination of the sugar molecular weight. Furthermore, MS^3 scan provided the specific fragmentation pattern of the aglycone. When necessary, flavonols were tentatively identified comparing the fragmentation pattern with UV spectra at 360 nm, corresponding to the maximum absorbance of these compounds.[7]

In the kaempferol aglycone MS^2 spectrum, the precursor ion remained the most abundant, even increasing the collision energy.

Table 3S reports the tentative identification of flavonols in grape samples.

Different conjugates of flavonols were recognised: galactosides and glucosides were identified after the hexose loss (162 amu); glucuronide and rutinoside were detected after glucuronic acid (176 amu) and rutinose (308 amu) loss, respectively.

Several samples showed two abundant peaks at approximately 18 and 19 minutes, respectively, with *m/z* 447 (n. **55-56**). Both of them showed a fragment at *m/z* 401: the difference of 46 amu is characteristic of a formate adduct, as reported above. The further fragmentation showed a peak at *m/z* 269: the difference of 132 amu was characteristic of a pentose derivative. Considering the molecular weight of this fragment, the peaks with *m/z* 447 were tentatively identified as two different trihydroxyflavone-pentoside. The aglycone compounds could be identified as apigenin (5,7,4'-trihydroxyflavone), baicalein (5,6,7-trihydroxyflavone), norwogonin (5,7,8-trihydroxyflavone) or galangin (3,5,7-trihydroxyflavone).

Compounds n. **59** and n. **60** showed a $[M-H]^-$ ion with *m/z* 479 and a maximum absorbance at 318 nm, with a retention time of approximately 23 and 24 minutes, respectively. Both of them showed a fragment *m/z* 317 and, considering the difference of 162 amu, a hexose moiety neutral loss was supposed. The aglycone was tentatively identified as myricetin and the two peaks were associated to different hexose derivatives. The similarity of the chromatographic behaviour with other compounds (e.g. quercetin-3-*O*-

galactoside and quercetin-3-*O*-glucoside) suggested that they probably could be identified as myricetin-galactoside and myricetin-glucoside.

Peak **61** (with a retention time of 24 minutes) had *m/z* 509 and a maximum absorbance at 343 nm. A fragment at *m/z* 329 was also observed. Assuming that the peak at *m/z* 509 was a water adduct and the [M-H]- ion had *m/z* 491, the peak was attributed to a dimethylquercetin-hexoside.

Table 3S. Identification of flavonols in grape samples by HPLC-DAD-ESI-MS^n

n.	Time (min)	λ max (nm)	MW	MS (m/z)	MS^2 (m/z)	MS^3 (m/z)	Tentative identification
44	37.85	254, 369	302	301 $[M-H]^-$	**179**,151	151	Quercetin*
45	27.88	255, 353	464	463 $[M-H]^-$	301	179, 151	Quercetin-3-*O*-glalctoside*
46	28.32	255, 353	464	463 $[M-H]^-$	301	179, 151	Quercetin-3-*O*-glucoside*
47	34.23	255, 353	478	477 $[M-H]^-$	301	179, 151	Quercetin-3-*O*-glucuronide*
48	27.06	256, 353	610	609 $[M-H]^-$	301	179, 151	Quercetin-3-*O*-rutinoside*
49	40.15	264, 367	286	285 $[M-H]^-$	-	-	Kaempferol*
50	30.50	265, 346	448	447 $[M-H]^-$	285	-	Kaempferol-galactoside
51	31.67	265, 346	448	447 $[M-H]^-$	285	-	Kaempferol-3-*O*-glucoside*
52	35.48	265, 346	462	461 $[M-H]^-$	285	-	Kaempferol-glucuronide
53	30.22	279	594	593 $[M-H]^-$	285	-	Kaempferol-rutinoside
54	32.41	365, 350	478	477 $[M-H]^-$	315	285, 271	Isorhamnetin-hexoside
55	18.10	280	402	447 $[M-H+HCOOH]^-$	**401**, 269	269	Trihydroxyflavone-riboside
56	19.34	280	402	447 $[M-H+HCOOH]^-$	**401**, 269	269	Trihydroxyflavone-riboside
57	36.01	280	402	477 $[M-H+HCOOH]^-$	431, **269**	87, 207, 225	Trihydroxyflavone-hexoside
58	32.34	343	508	507 $[M-H]^-$	345	301, 273	Syringetin-hexoside
59	23.59	318, 279	480	479 $[M-H]^-$	299, **317**	-	Myricetin-hexoside
60	24.35	318, 279	480	479 $[M-H]^-$	299, **317**	271, 279	Myricetin-hexoside
61	24.31	343	492	509 $[M-H+H_2O]^-$	329, 347, 441	299	Dimethylquercetin-hexoside

* These compounds were confirmed with a standard commercially available

In bold the ions used for MS^3 fragmentation

Stilbenes and phenolic acids

Table 4S lists the stilbenes and organic acids tentatively identified in the samples under study. Resveratrol was present in two isomeric forms, *trans* and *cis*. These two isomers showed the same mass and fragmentation pattern, but they could be distinguished by their maximum absorbance and retention times.[8] *Trans*-resveratrol presented a maximum absorbance at 307 nm and the retention time was approximately 36 minutes (n. **62**); *cis*-resveratrol showed a maximum absorbance at 284 and a retention time of approximately 38 minutes (n. **63**) (not shown).

Table 4S. Identification of stilbenes and organic acids in grape samples by HPLC-DAD-ESI-MS^n

n.	Time (min)	λ max (nm)	MW	MS (m/z)	MS^2 (m/z)	MS^3 (m/z)	Tentative identification
62	36.14	307	228	227 $[M-H]^-$	**185**, 159	-	*trans*-Resveratrol *
63	38.33	284	228	227 $[M-H]^-$	**185**, 159	-	*cis*-Resveratrol*
64	33.13	280	390	389 $[M-H]^-$	227	185, 159	*cis*-Piceid
65	20.02	326, 242	312	311 $[M-H]^-$	**243**, 179, 149	175	Caftaric acid*°
66	24.58	312	296	295 $[M-H]^-$	**163**	119	*trans*-coutaric acid
67	25.77	286	296	295 $[M-H]^-$	**149,** 163	-	*cis*-coutaric acid
68	27.41	322	194	193 $[M-H]^-$	134, **149**, 178	-	Ferulic acid*
69	18.65	325	354	353 $[M-H]^-$	191	-	Chlorogenic acid*
70	18.78	323	180	179 $[M-H]^-$	135	-	Caffeic acid*

* These compounds were confirmed with a standard commercially available; °Caftaric acid was identified in samples from the HPLC-DAD chromatogram instead of MS analysis.

In bold the ions used for MS^3 fragmentation

Some dimers, trimers and tetramers of stilbenes were also identified on the basis of literature data: peak n. **65** was attributed to a resveratrol trimer (probably α-viniferin).[9] Compounds n. **66**, **67** and **68** were tentatively identified as three different resveratrol trimer, and one of them could be attributed to the mostly known "*trans*-ε-viniferin", according to data reported by Becker and co-workers.[10] Due to its molecular weight, compound n. **69** was attributed to a resveratrol tetramer (vitisin).[11]

The presence of organic acids (n. **70**, **73**, **74**, **75**) was also investigated. Standard caftaric acid (n. **70**) showed an incomplete ionization: so that, although it was observed in UV analysis, the peak was not well detectable in MS chromatogram.

Considering the fragmentation pattern of compounds n. **71**-**72**, they were tentatively identified as *trans* and *cis*-coutaric acid, as also described by Cantos et al.[6] According to the study reported by Mozetič,[12] compound n. **71** was identified as the *trans* isomer (λ max at 312 nm), while the compound n. **72** as the *cis* isomer (λ max at 286 nm).

Among organic acids, caftaric acid showed a difficult fragmentation in LC-MS analysis, and it was identified taking into account its UV spectrum (325 nm).

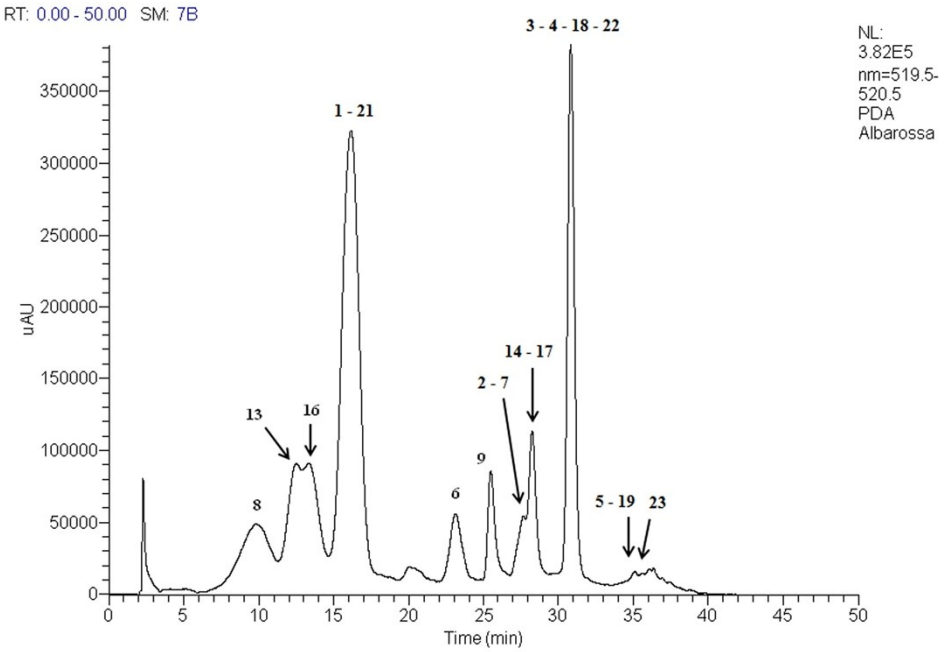

Figure 1S- Anthocyanin chromatographic profile (DAD at 520 nm) of Albarossa grape. Legend for numbers above peaks: 8-Delphinidin-3-O-glucoside; 13- Cyanidin-3-O-glucoside; 16- Petunidin-3-O-glucoside; 1- Malvidin-3-O-glucoside; 21- Peonidin-3-O-glucoside; 6-Malvidin-acetyl-hexoside; 9-Delphinidin-coumaroyl-hexoside; 2-Malvidin-caffeoyl-hexoside; 7- Malvidin-acetyl-hexoside; 14- Cyanidin-coumaroyl-hexoside; 17- Petunidin-coumaroyl hexoside; 3- Malvidin-caffeoyl-hexoside; 4- Malvidin-cumaroyl-hexoside; 18- Petunidin-caffeoyl-hexoside; 22- Peonidin-coumaroyl- hexoside; 5-Malvidin-cumaroyl-hexoside; 19-Petunidin-caffeoyl-hexoside; 23- Peonidin-coumaroyl- hexoside

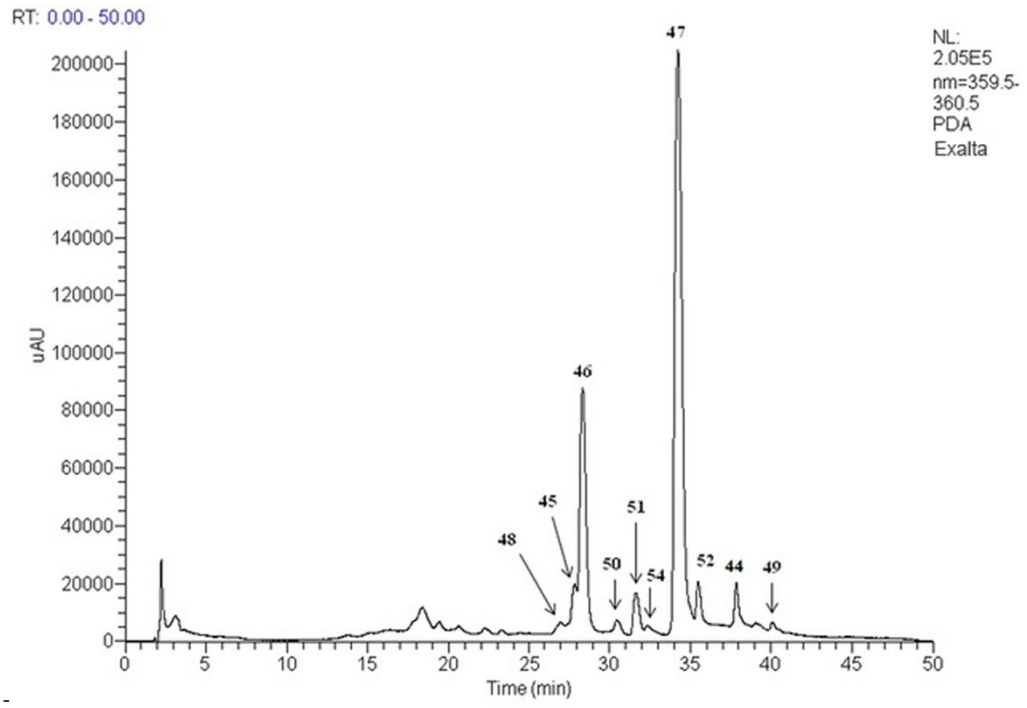

Figure 2S- Flavonol chromatographic profile (DAD at 360 nm) of Exalta grape. Legend for numbers above peaks: 48 Quercetin-3-O-rutinoside; 45 Quercetin-3-O-glalctoside; 46 Quercetin-3-O-glucoside; 50 Kaempferol-galactoside; 51 Kaempferol-3-O-glucoside; 54 Isorhamnetin-hexoside; 47 Quercetin-3-O-glucuronide; 52 Kaempferol-glucuronide; 44 Quercetin; 49 Kaempferol.

References

1 W. M. Stöggl, C. W. Huck and G. K. Bonn, Structural elucidation of catechin and epicatechin in sorrel leaf extracts using liquid-chromatography coupled to diode array-, fluorescence- ,and mass spectrometric detection, *J. Sep. Sci.*, , DOI:10.1002/jssc.200301694.

2 S. Pérez-Magariño, I. Revilla, M. L. González-Sanjosé and S. Beltrán, Various applications of liquid chromatography-mass spectrometry to the analysis of phenolic compounds, *J. Chromatogr. A*, 1999, **847**, 75–81.

3 W. M. Stöggl, C. W. Huck and G. K. Bonn, Structural elucidation of catechin and epicatechin in sorrel leaf extracts using liquid-chromatography coupled to diode array-, fluorescence- ,and mass spectrometric detection, *J. Sep. Sci.*, 2004, **27**, 524–528.

4 M. N. Bravo, S. Silva, A. V. Coelho, L. V. Boas and M. R. Bronze, Analysis of phenolic compounds in Muscatel wines produced in Portugal, *Anal. Chim. Acta*, 2006, **563**, 84–92.

5 S. De Pascual-Teresa and J. C. Rivas-Gonzalo, *Application of LC-MS for the identification of polyphenols. Methods in Polyphenol Analysis*, *Application of LC-MS for the identification of polyphenols. Methods in Polyphenol Analysis*, The Royal Society of Chemistry, Cambridge, U.K., 2003.

6 E. Cantos, J. C. Espín and F. A. Tomás-Barberán, Varietal differences among the polyphenol profiles of seven table grape cultivars studied by LC-DAD-MS-MS, *J. Agric. Food Chem.*, 2002, **50**, 5691–5696.

7 A. Schieber, P. Keller, P. Streker, I. Klaiber and R. Carle, Detection of isorhamnetin glycosides in extracts of apples (Malus domestica cv. 'Brettacher') by HPLC-PDA and HPLC-APCI-MS/MS, *Phytochem. Anal.*, 2002, **13**, 87–94.

8 L. Camont, C. H. Cottart, Y. Rhayem, V. Nivet-Antoine, R. Djelidi, F. Collin, J. L. Beaudeux and D. Bonnefont-Rousselot, Simple spectrophotometric assessment of the trans-/cis-resveratrol ratio in aqueous solutions, *Anal. Chim. Acta*, 2009, **634**, 121–128.

9 F. Mattivi, U. Vrhovsek, G. Malacarne, D. Masuero, L. Zulini, M. Stefanini, C. Mose, R. Velasco and G. Guella, Profiling of resveratrol oligomers, important stress metabolites, accumulating in the leaves of hybrid Vitis vinifera (Merzling ?? Teroldego) genotypes infected with Plasmopara viticola, *J. Agric. Food Chem.*, 2011, **59**, 5364–5375.

10 L. Becker, V. Carré, A. Poutaraud, D. Merdinoglu and P. Chaimbault, MALDI mass spectrometry imaging for the simultaneous location of resveratrol, pterostilbene and viniferins on grapevine leaves, *Molecules*, 2014, **19**, 10587–

10600.

11 R. Flamini, M. De Rosso, F. De Marchi, A. Dalla Vedova, A. Panighel, M. Gardiman, I. Maoz and L. Bavaresco, An innovative approach to grape metabolomics: Stilbene profiling by suspect screening analysis, *Metabolomics*, 2013, **9**, 1243–1253.

12 B. Mozetič, I. Tomažič, A. Škvarč and P. Trebše, Determination of polyphenols in white grape berries cv. Rebula, *Acta Chim. Slov.*, 2006, **53**, 58–64.