

## Unveiling the Phytochemical Nature of Acorns: The Relevance of Dehusking

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**Table S1**

Individual p-values of the Kruskal–Wallis test for the different phenolic contents and antioxidant activity between the different manually dehusked acorn species from Gerês. Significance differences were established at  $p < 0.05$ .

<b>Parameter</b>	<b>QP x QR</b>	<b>QR x QI</b>	<b>QP x QI</b>
<b><i>Phenolics</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	n.s.	n.s.	n.s.
Total	p<0.05	p<0.05	p<0.05
<b><i>Flavonoids</i></b>			
Free	p<0.05	p<0.05	n.s.
Bound	n.s.	n.s.	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>Hydrolysable tannins</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	-	-	-
Total	p<0.05	p<0.05	p<0.05
<b><i>Condensed tannins</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	-	-	-
Total	p<0.05	p<0.05	n.s.
<b><i>Gallic acid</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	p<0.05	p<0.05	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>Ellagic acid</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	p<0.05	p<0.05	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>ABTS</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	p<0.05	p<0.05	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>DPPH</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	n.s.	p<0.05	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>ORAC</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	p<0.05	n.s.	p<0.05
Total	p<0.05	p<0.05	p<0.05
<b><i>FRAP</i></b>			
Free	p<0.05	p<0.05	p<0.05
Bound	p<0.05	p<0.05	p<0.05
Total	p<0.05	n.s.	p<0.05

QP: *Quercus pyrenaica*; QR: *Quercus robur*; QI: *Quercus ilex*; n.s.: not significant ( $p > 0.05$ ).

**Table S2**

Individual p-values of the Kruskal–Wallis test for the different phenolic contents and antioxidant activity between the different manually dehusked acorn species from Gerês. Significance differences were established at  $p < 0.05$ .

Parameter	<i>Q. pyrenaeca</i>		<i>Q. robur</i>		<i>Q. ilex</i>	
	M x T	M x D	M x T	M x D	M x T	M x D
<b>Phenolics</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>Flavonoids</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>Hydrolysable tannins</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Bound	-	-	-	-	-	-
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
<b>Condensed tannins</b>						
Free	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Bound	-	-	-	-	-	-
Total	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
<b>Gallic acid</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>Ellagic acid</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>ABTS</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	n.s.	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>DPPH</b>						
Free	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b>ORAC</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	n.s.
<b>FRAP</b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	n.s.	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$

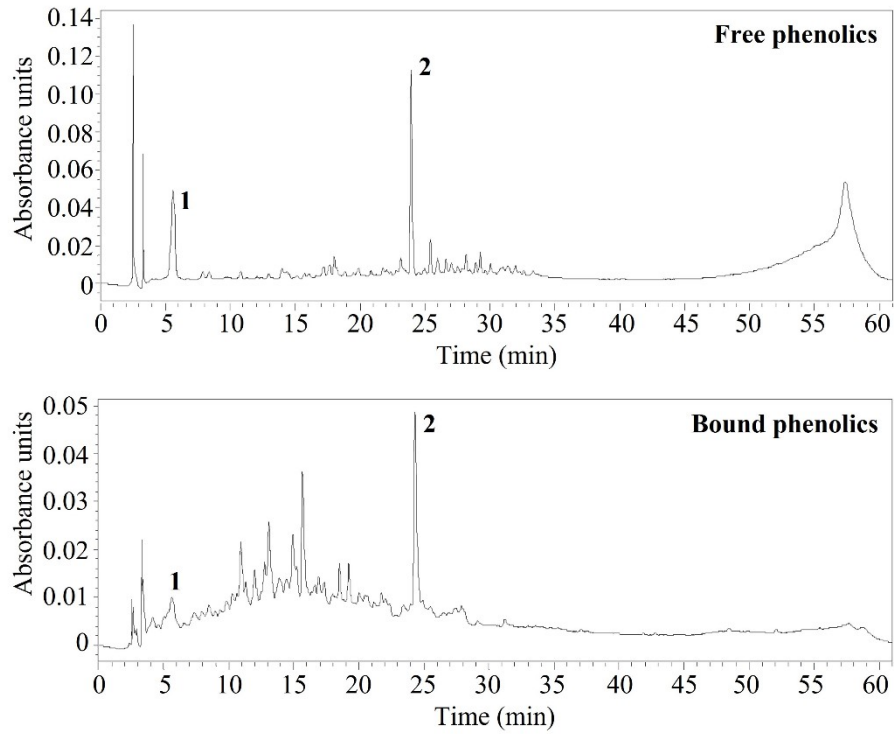
M: Manual dehusking; T: Thermal dehusking; D: Dehusking by drying; n.s.: not significant ( $p > 0.05$ ).

**Table S3**

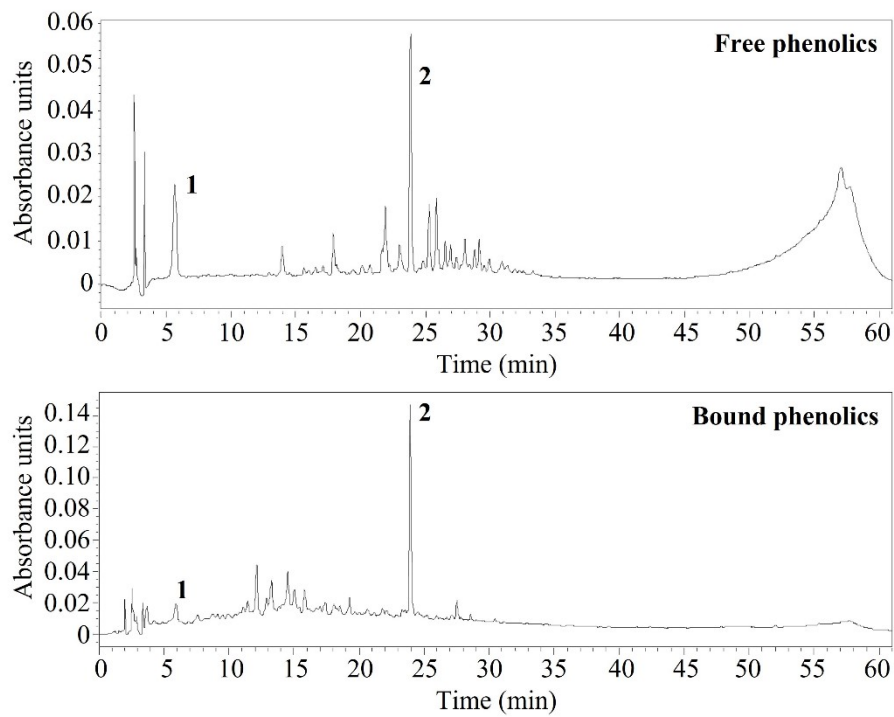
Individual p-values of the Kruskal–Wallis test for the different phenolic contents and antioxidant activity between the different dehulled by drying acorn species harvested in Gerês and the commercial acorns from Alentejo. Significance differences were established at  $p < 0.05$ .

<i>Parameter</i>	<i>QP x C</i>	<i>QR x C</i>	<i>QI x C</i>	<i>QP x QR</i>	<i>QR x QI</i>	<i>QP x QI</i>
<b><i>Phenolics</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>Flavonoids</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	n.s.
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
<b><i>Hydrolysable tannins</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	-	-	-			
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>Condensed tannins</i></b>						
Free	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	-	-	-			
Total	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>Gallic acid</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$
<b><i>Ellagic acid</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>ABTS</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>DPPH</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
<b><i>ORAC</i></b>						
Free	$p < 0.05$	n.s.	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	n.s.
Total	$p < 0.05$	$p < 0.05$	n.s.	n.s.	$p < 0.05$	$p < 0.05$
<b><i>FRAP</i></b>						
Free	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Bound	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$
Total	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$	$p < 0.05$

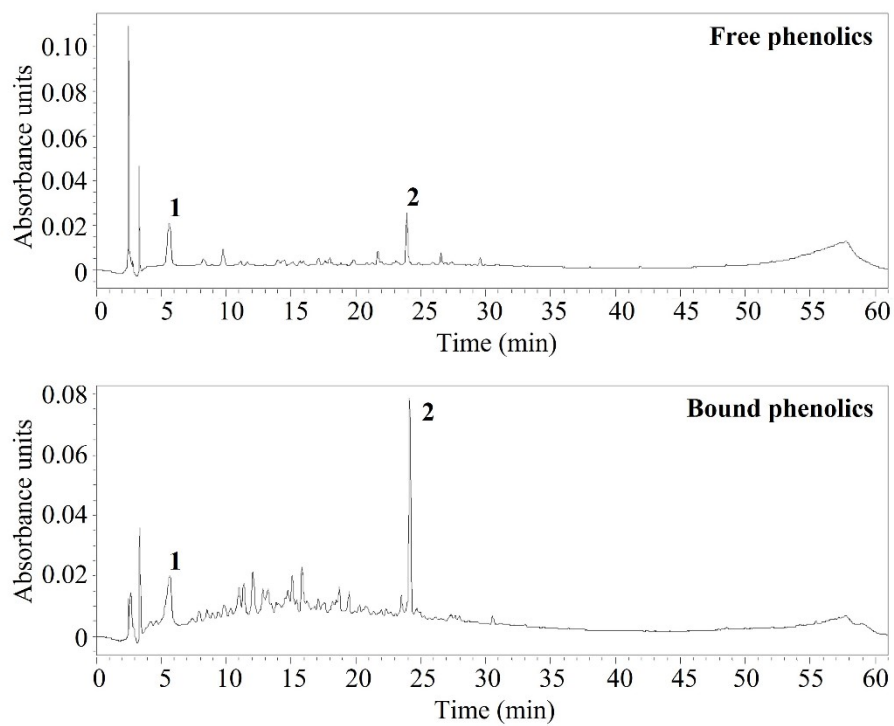
C: Commercial acorns; QP: *Quercus pyrenaica*; QR: *Quercus robur*; QI: *Quercus ilex*; n.s.: not significant ( $p > 0.05$ ).



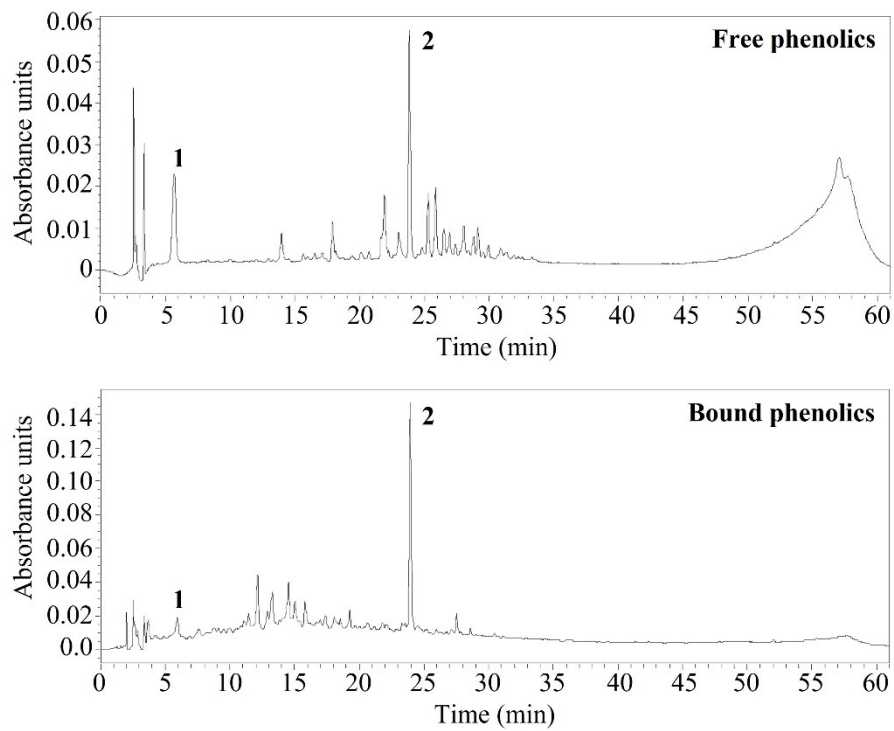
**Fig. S1:** Free and bound phenolic profile at 280 nm of the *Q. pyrenaica* acorns manually dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



**Fig. S2:** Free and bound phenolic profile at 280 nm of the *Q. pyrenaica* acorns thermally dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.

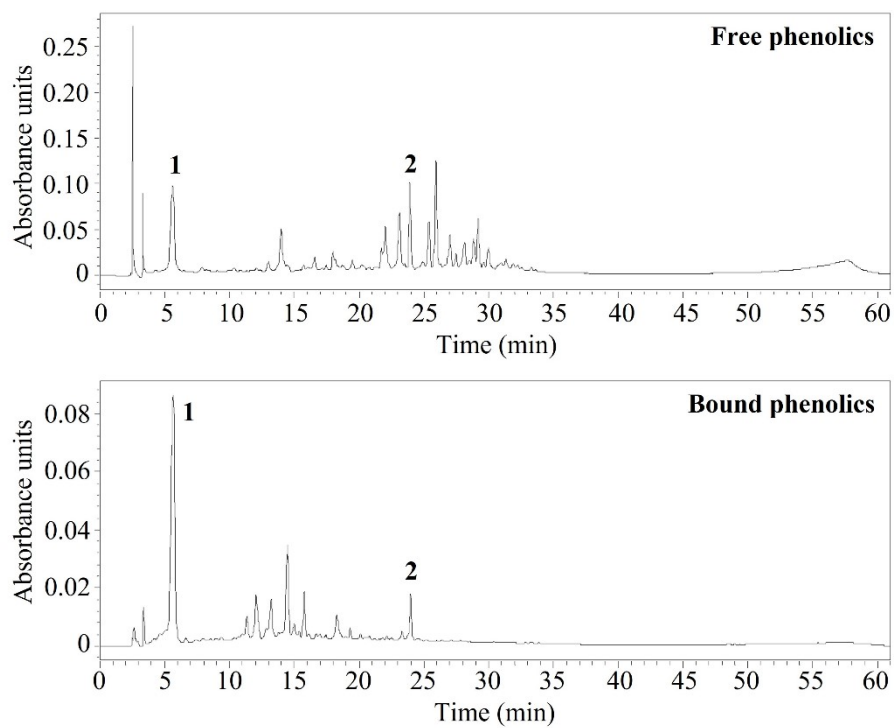


**Fig. S3:** Free and bound phenolic profile at 280 nm of the *Q. pyrenaica* acorns dehusked by drying. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.

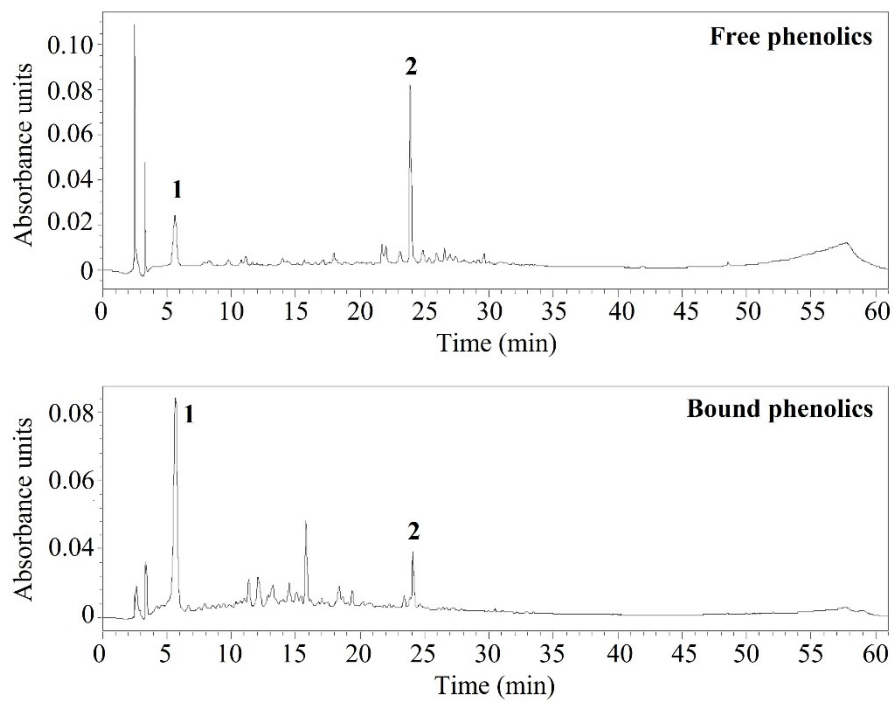


**Fig. S4:** Free and bound phenolic profile at 280 nm of the *Q. robur* acorns manually dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.

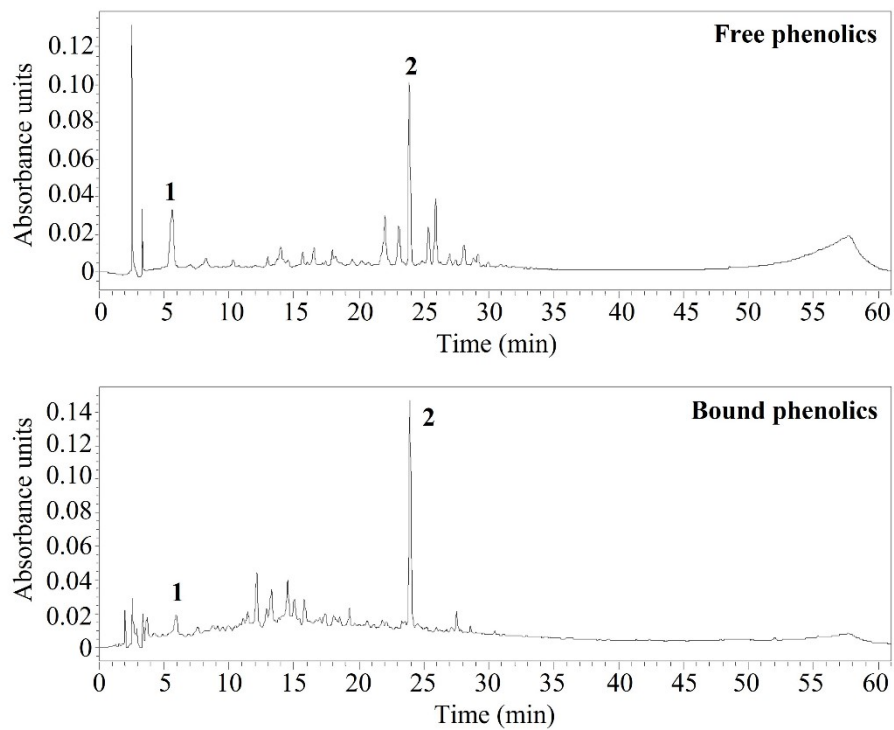




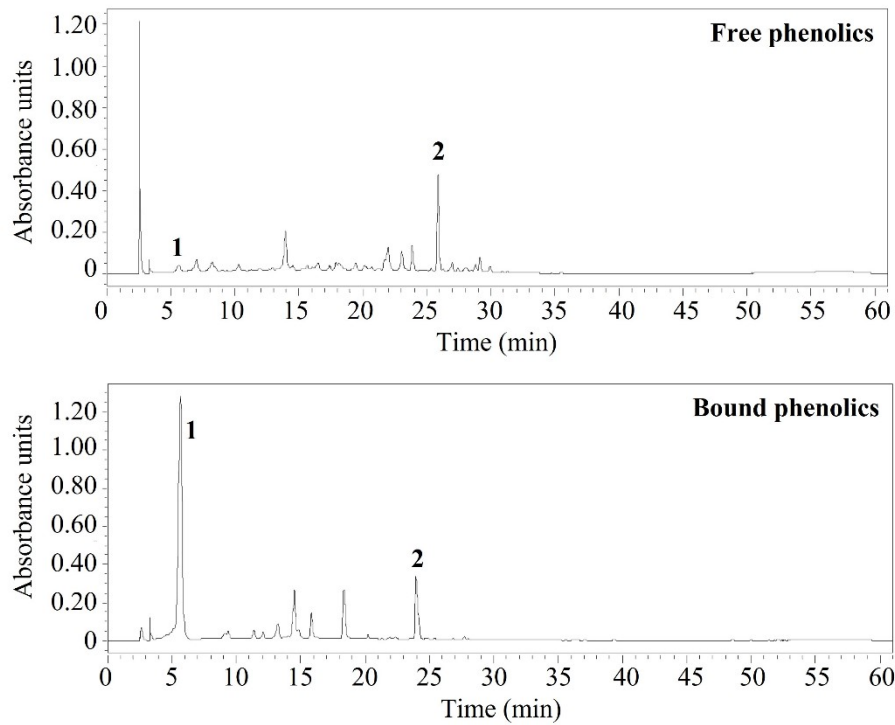
**Fig. S5:** Free and bound phenolic profile at 280 nm of the *Q. robur* acorns thermally dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



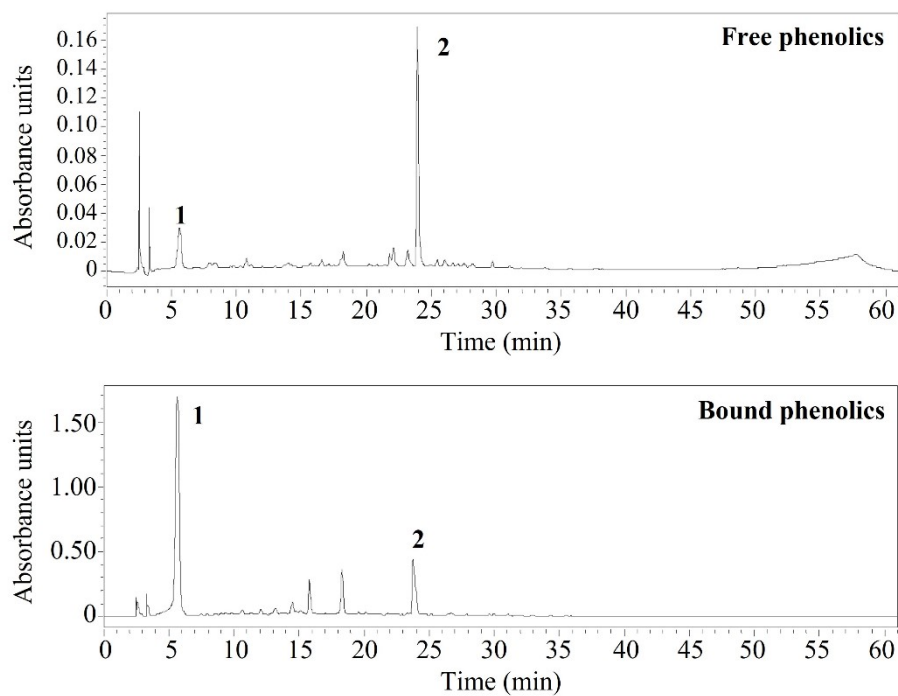
**Fig. S6:** Free and bound phenolic profile at 280 nm of the *Q. robur* acorns dehusked by drying. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



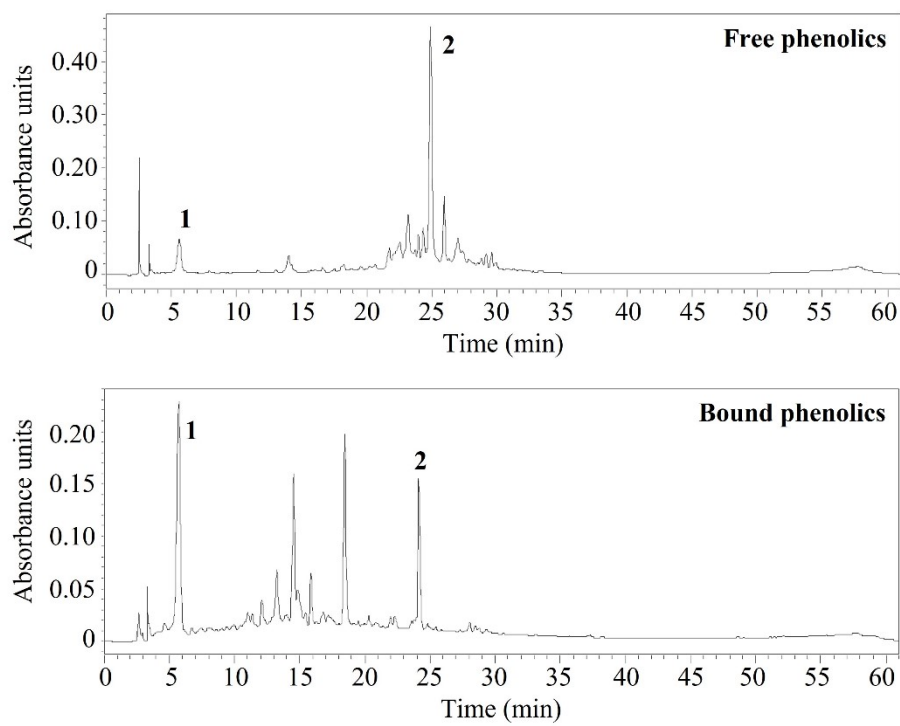
**Fig. S7:** Free and bound phenolic profile at 280 nm of the *Q. ilex* acorns manually dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



**Fig. S8:** Free and bound phenolic profile at 280 nm of the *Q. ilex* acorns thermally dehusked. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



**Fig. S9:** Free and bound phenolic profile at 280 nm of the *Q. ilex* acorns dehusked by drying. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.



**Fig. S10:** Free and bound phenolic profile at 280 nm of the commercial acorns. Peaks 1 and 2 correspond to gallic and ellagic acid, respectively.