

Supplementary Materials: The following are available online at www.mdpi.com/article/10.3390/f12091260/s1, Figure S1: Analysis of the total ion current (TIC) in different quality control (QC) samples. The abscissa represents the retention time (min) of metabolite detection, and the ordinate represents the intensity (cps: count per second) of the ion current. Figure S2: Metabolites difference between leaves of H11 (green leaves) and H12 (red leaves). (a) Compared H12 with H11, most isoflavone and flavone metabolites were up-regulated based on relative quantitative contents. (b) The up-regulation of major flavanone, flavonol, anthocyanidin and proanthocyanidin metabolites. (c) The down-regulation of most amino acid derivatives and organic acids. The darker the color, the higher the metabolites expression level. Figure S3: Go classification of different expression genes in leaf transcriptomes of *E. ulmoides* among different samples. Figure S4: KEGG enrichment of down-regulated genes in H12 (red leaves) compared with H11 (green leaves) in *E. ulmoides*. Figure S5: Differential expression of the genes related to photosynthesis pathway. The expression level was based on FPKM value. The darker the color, the higher the gene expression level. Figure S6: Phylogeny of all EuMYBs and AtMYBs. Those MYBs involved in flavonoids biosynthesis were focused. Phylogenetic analysis followed the settings: the maximum likelihood method, the substitution model was Dayhoff, priors set to the Yule Model, the birth rate was 1.0, the mcmc chain length was 10,000,000, bootstrapping set to 1,000, and the TreeAnnotator program set the posterior probability limit to 1.0 with a burn in percentage of 90. The scale bar indicates the average number of amino acid substitutions per site. Figure S7: Phylogeny of all EubHLHs and AtbHLHs. Phylogenetic analysis followed the above settings. Figure S8. qRT-PCR analysis of the expression of candidate genes in *E. ulmoides* leaves. Table S1: Primer sequences of genes for RT-qPCR verification. Table S2: Candidate genes involved in chlorophyll metabolism pathway. Table S3: Candidate genes involved in photosynthesis pathway. Table S4. Candidate genes related to flavonoids biosynthesis pathway. Table S5: The DEGs of gene family compared H11 (green leaves) and H12 (red leaves). Table S6: The expression profile of EuMYBs and EubHLHs transcripts related to flavonoids synthesis. Table S7: The node genes of flavonol and anthocyanin biosynthesis.

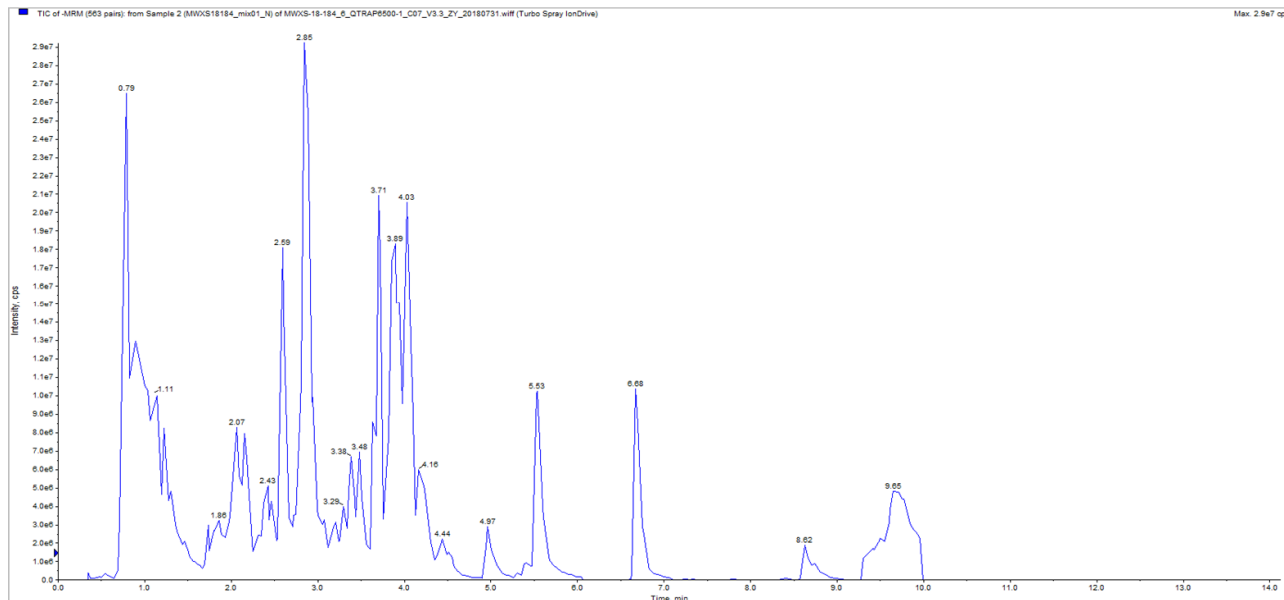
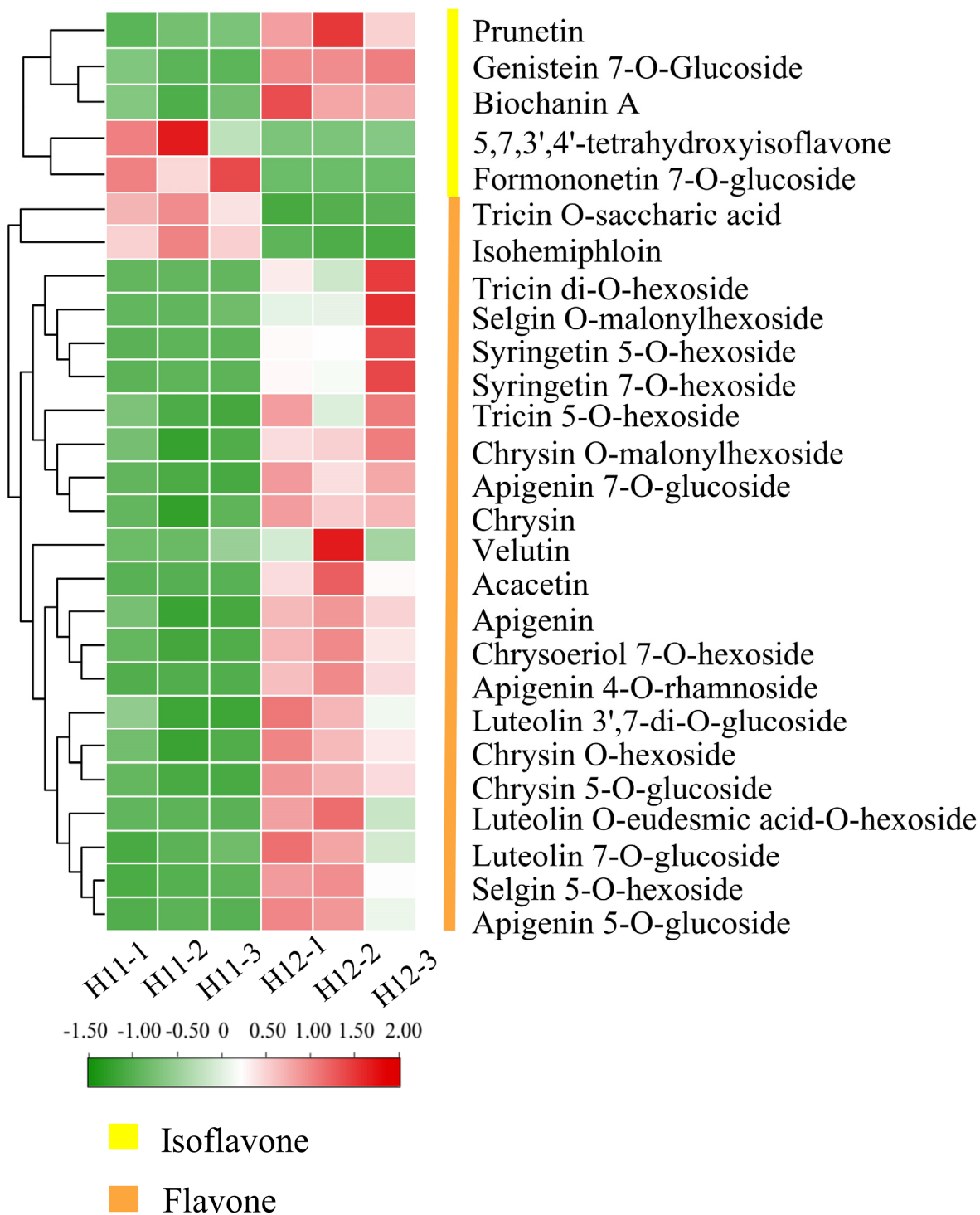
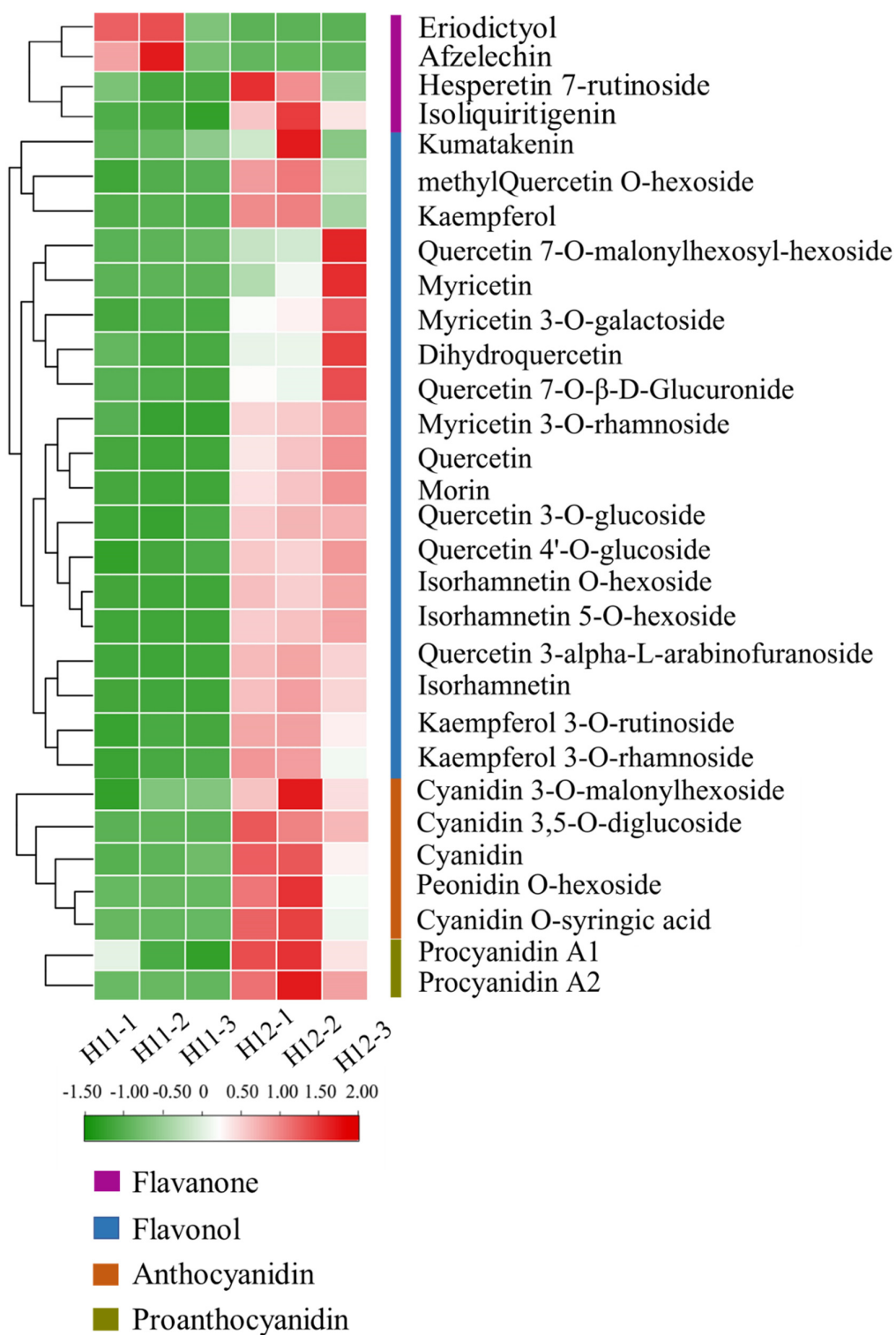


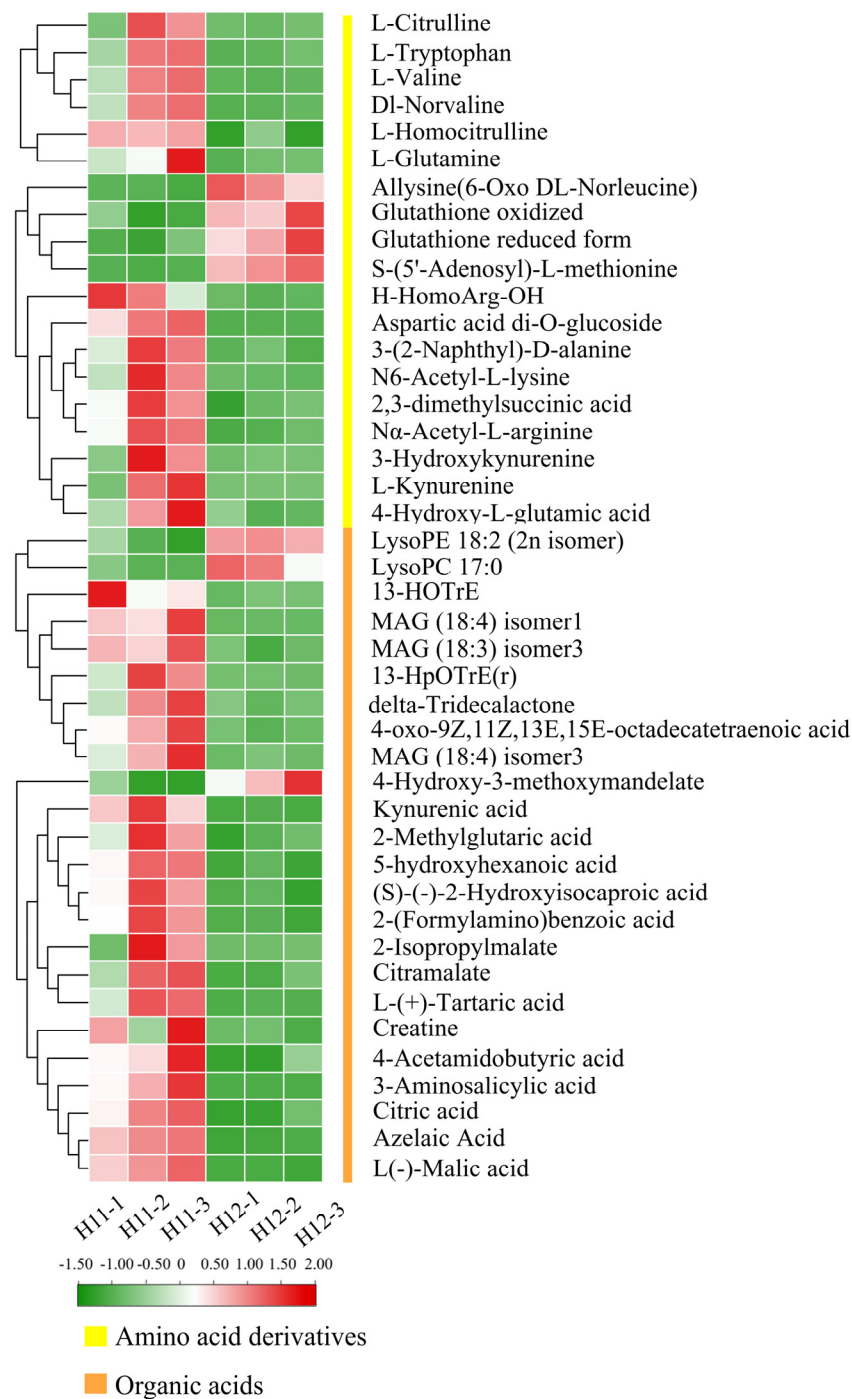
Figure S1. Analysis of the total ion current (TIC) in different quality control (QC) samples. The abscissa represents the retention time (min) of metabolite detection, and the ordinate represents the intensity (cps: count per second) of the ion current.



(a)



(b)



(c)

Figure S2. Metabolites difference between leaves of H11 (green leaves) and H12 (red leaves). (a) Compared H12 with H11, most isoflavone and flavone metabolites were up-regulated based on relative quantitative contents. (b) The up-regulation of major flavanone, flavonol, anthocyanidin and proanthocyanidin metabolites. (c) The down-regulation of most amino acid derivatives and organic acids. The darker the color, the higher the metabolites expression level.

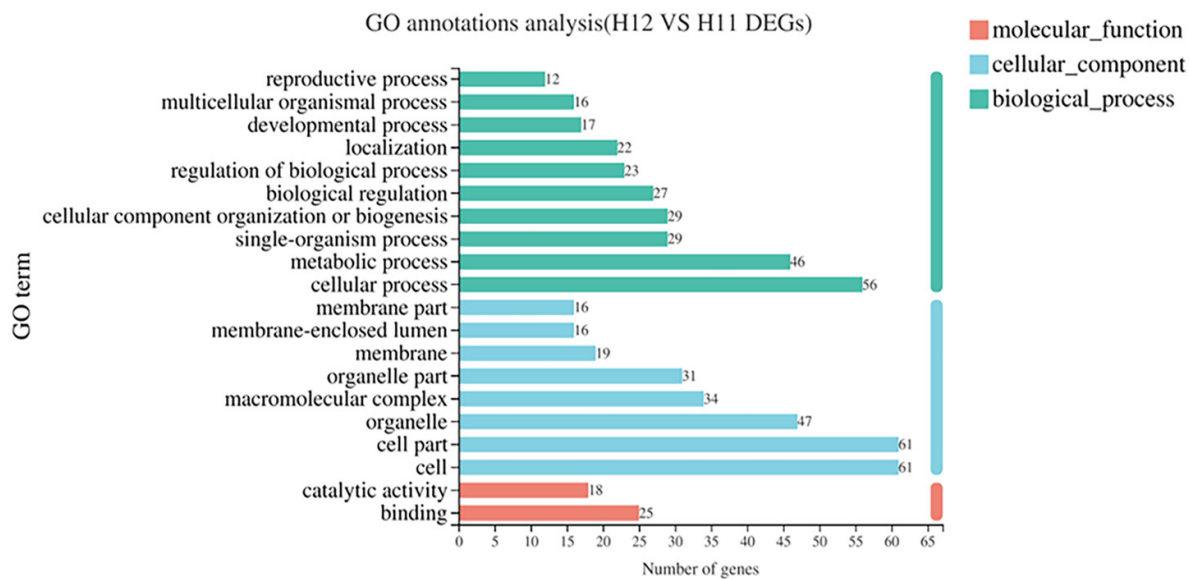


Figure S3. Go classification of different expression genes in in leaf transcriptomes of *E. ulmoides* among different samples.

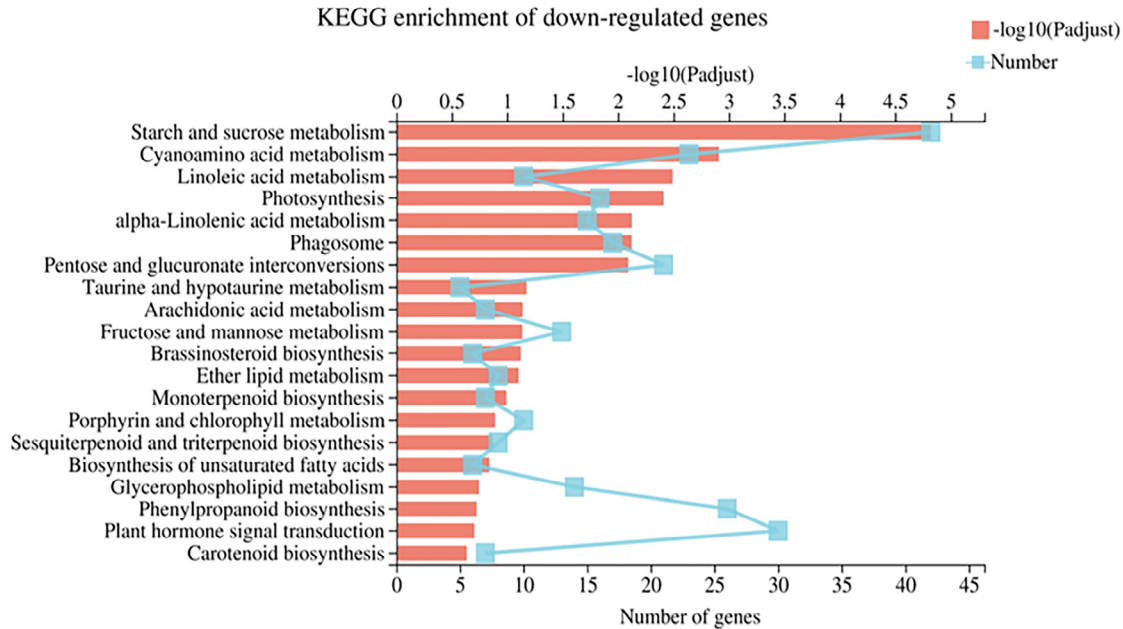


Figure S4. KEGG enrichment of down-regulated genes in H12 (red leaves) compared with H11 (green leaves) in *E. ulmoides*.

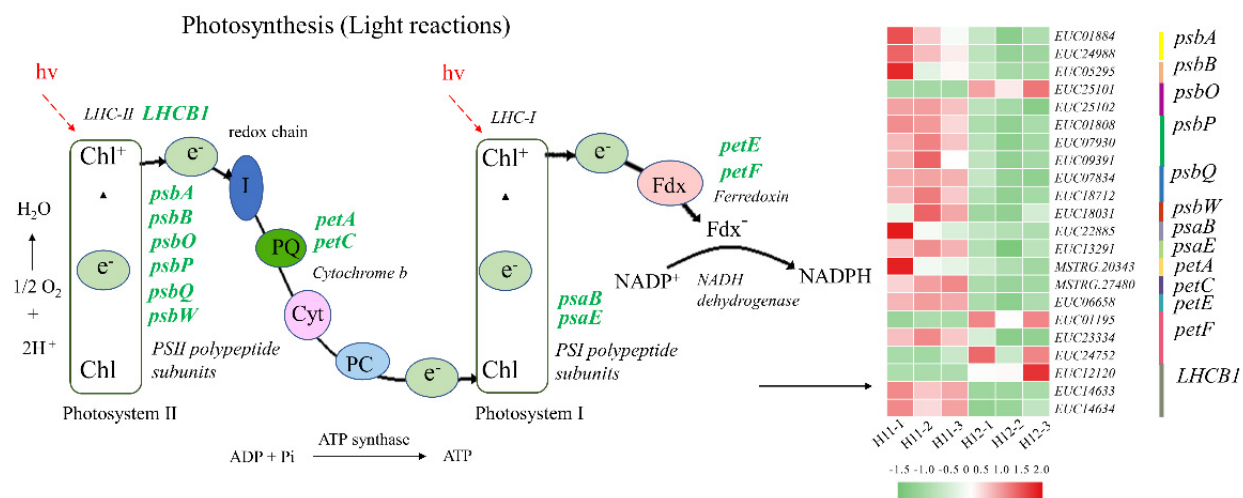


Figure S5. Differential expression of the genes related to photosynthesis pathway. The expression level was based on FPKM value. The darker the color, the higher the gene expression level.

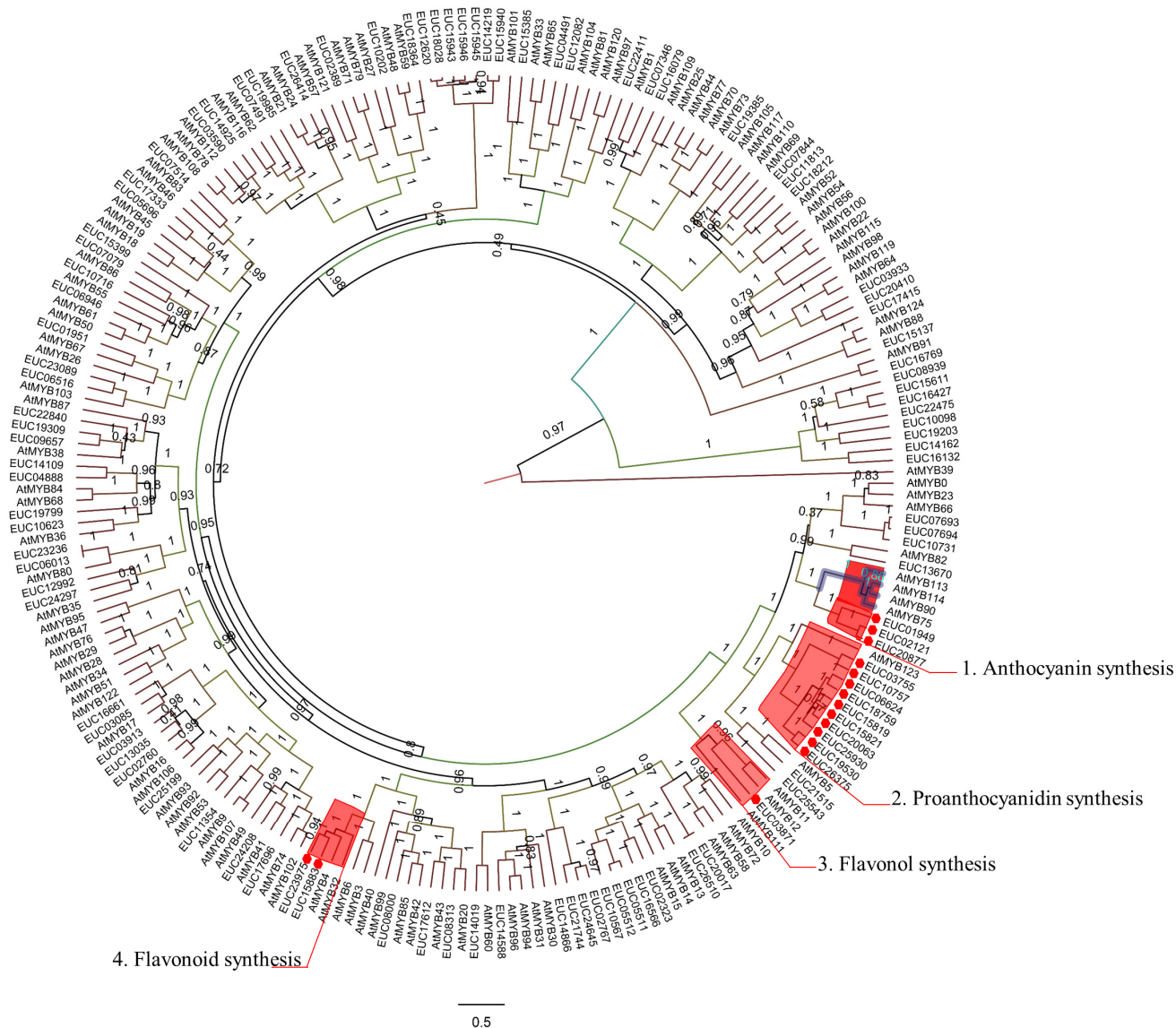


Figure S6. Phylogeny of all EuMYBs and AtMYBs. Those MYBs involved in flavonoids biosynthesis were focused. Phylogenetic analysis followed the settings: the maximum likelihood method, the substitution model was Dayhoff, priors set to the Yule Model, the birth rate was 1.0, the mcmc chain length was 10,000,000, bootstrapping set to 1,000, and the TreeAnnotator program set the posterior probability limit to 1.0 with a burn in percentage of 90. The scale bar indicates the average number of amino acid substitutions per site.

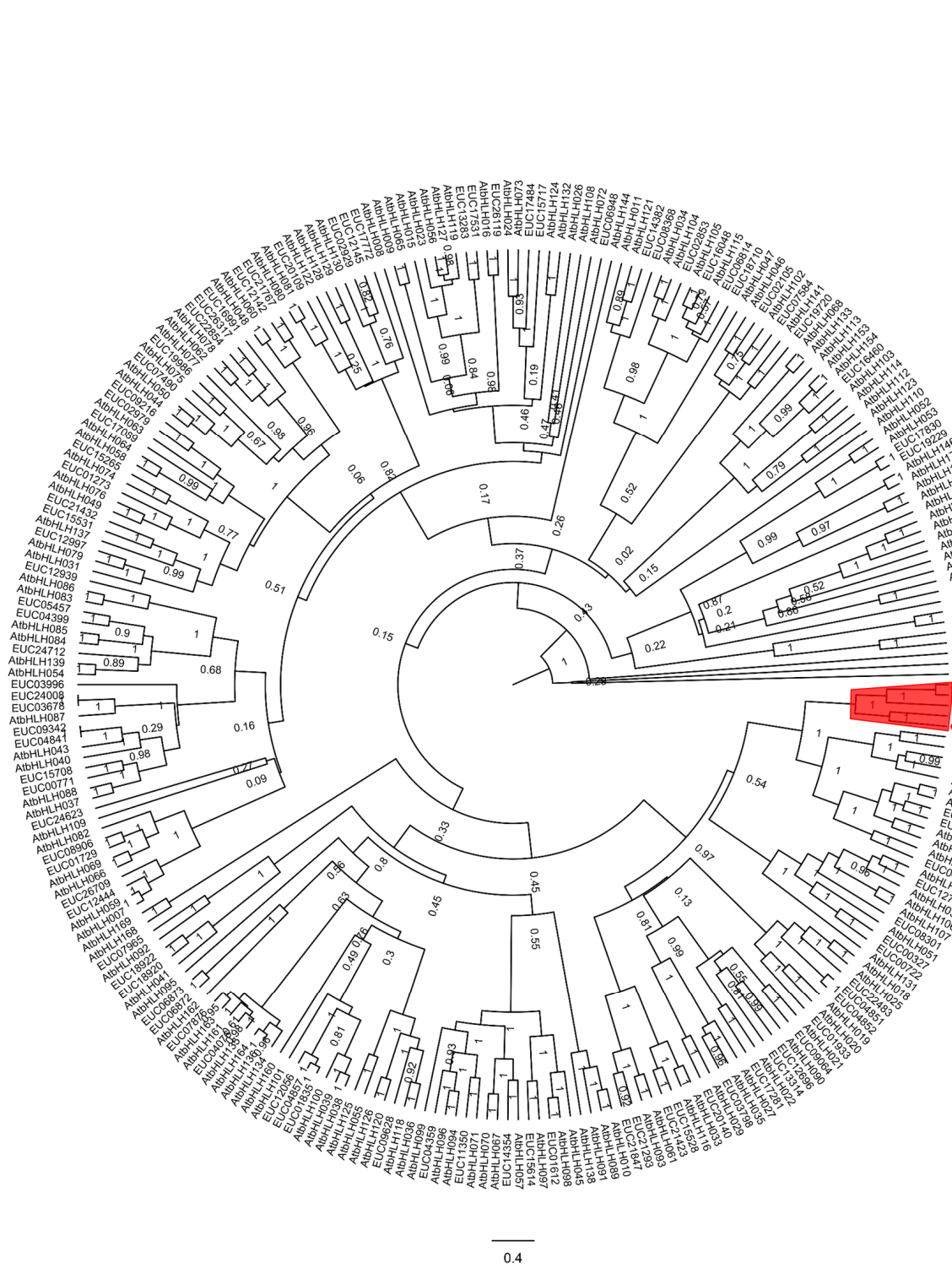


Figure S7. Phylogeny of all EubHLHs and AtbHLHs. Phylogenetic analysis followed the above settings.

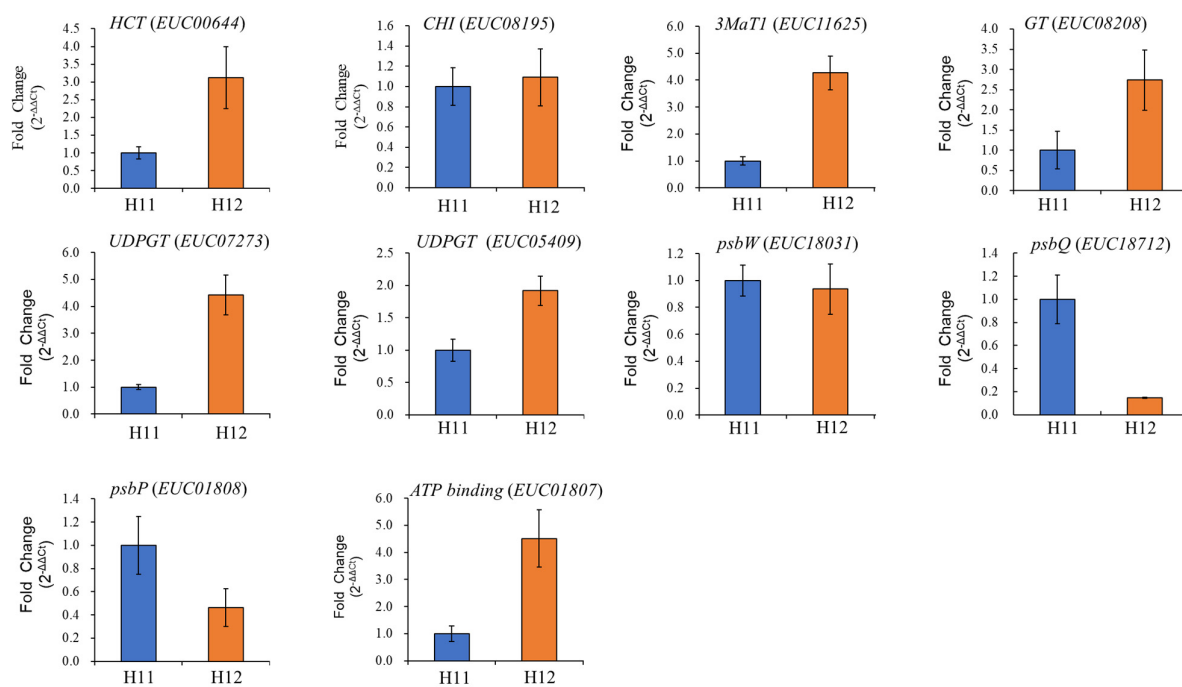


Figure S8. qRT-PCR analysis of the expression of candidate genes in *E. ulmoides* leaves.

Table S1. Primer sequences of genes for RT-qPCR verification

Gene name	Gene category	Forward primer	Reverse primer
<i>EUC05716</i>	<i>GAPDH</i>	ATGAAATCAAGGCCGCCATC	TCGGTATACCCCAAGATGCC
<i>EUC00644</i>	<i>HCT</i>	CCGGCATGACAGCAGACAGC	CGGACTCACTCGTTGCCATTACC
<i>EUC08195</i>	<i>CHI</i>	GCCACAAGGATCTCCGACTTACTG	CTCCGCCATTCTCCAACACCATC
<i>EUC11625</i>	<i>3MaT1</i>	GCAGCTAATGAGTCGTCTGAGTCG	GCCGTGGTGATTGGTGAAGCC
<i>EUC08208</i>	<i>UFOG6</i>	TTCGAGCTGGAATCACAGGCAATC	CAGCAGAGGTGGAGTTGTTGTCTG
<i>EUC07273</i>	<i>UDPGT</i>	CCTGAAGTGGCTCGACGATCAAC	TTGGTCAACGCCGAAGCATCC
<i>EUC05409</i>	<i>UDPGT</i>	GACCAGGTGACGAACGCCAAG	TTCTTCCACTTCAACGCCTTCTCC
<i>EUC18031</i>	<i>psbW</i>	TGGTGGACGAGAGGATGAGCAC	CAAGCCGAACACTCCGAACAGG
<i>EUC18712</i>	<i>psbQ</i>	AGCCACTGTCGCCAGCAGAG	CTTCGCTGAGATCACGGTGTGAG
<i>EUC01808</i>	<i>psbP</i>	CTTGACAAGGACAGCCGATGGAG	TGTAGAGCTTGCCACCATTACAG
<i>EUC01807</i>	<i>ATP binding</i>	CCCTGCCCAACATCAAGGTC	TGTCCTTGCCTCCCCAGAT

Table S2. Candidate genes involved in chlorophyll metabolism pathway.

Function	Gene	Enzyme	Total No.	DEG No.
ALA formation	<i>HemA</i>	Glutamyl-tRNA reductase	2	0
	<i>HemL</i>	Glutamate-1-semialdehyde 2,1-aminomutase	5	0
Proto IX formation	<i>HemB</i>	Porphobilinogen synthase	1	0
	<i>HemC</i>	Hydroxymethylbilane synthase	4	0
	<i>HemD</i>	Uroporphyrinogen-III synthase	1	0
	<i>HemE</i>	Uroporphyrinogen decarboxylase	2	1
	<i>HemF</i>	Coproporphyrinogen-III oxidase	2	1
	<i>HemY</i>	Oxygen-dependent protoporphyrinogen oxidase	1	0
Chlorophyll formation	<i>chlH</i>	Magnesium chelatase subunit H	1	1
	<i>chlM</i>	Magnesium protoporphyrin IX methyltransferase	1	0
	<i>chlE</i>	Magnesium-protoporphyrin IX monomethyl ester (oxidative) cyclase	1	0
	<i>DVR</i>	Divinyl chlorophyllide-a 8-vinyl-reductase	3	3
	<i>POR</i>	Protochlorophyllide reductase	4	3
	<i>chlG</i>	Chlorophyll synthase	6	3
Chlorophyll cycle	<i>CAO</i>	Chlorophyllide a oxygenase	2	0
	<i>CLH</i>	Chlorophyllase	5	0
	<i>NYC1</i>	Chlorophyll (ide) b reductase	4	0
	<i>HCAR</i>	7-Hydroxymethyl chlorophyll a reductase	1	0
Chlorophyll degradation	<i>PAO</i>	Pheophorbide a oxygenase	1	0
	<i>RCCR</i>	Red chlorophyll catabolite reductase	1	0

Table S3. Candidate genes involved in photosynthesis pathway.

Function	Gene	Enzyme	Total No.	DEG No.
photosystem II	<i>psbA</i>	photosystem II P680 reaction center D1 protein	4	2
	<i>psbB</i>	photosystem II CP47 chlorophyll apoprotein	4	1
	<i>psbO</i>	photosystem II oxygen-evolving enhancer protein 1	2	2
	<i>psbP</i>	photosystem II oxygen-evolving enhancer protein 2	4	3
	<i>psbQ</i>	photosystem II oxygen-evolving enhancer protein 3	4	2
	<i>psbW</i>	photosystem II PsbW protein	1	1
photosystem I	<i>psaB</i>	photosystem I P700 chlorophyll-a apoprotein A2	4	1
	<i>psaE</i>	photosystem I subunit IV	3	1
cytochrome	<i>petA</i>	apocytochrome f	1	1
b6/f complex	<i>petC</i>	cytochrome b6-f complex iron-sulfur subunit	3	1
photosynthetic electron transport	<i>petE</i>	plastocyanin	1	1
	<i>petF</i>	ferredoxin	6	3
Photosynthesis- antenna proteins	<i>LHCB1</i>	light-harvesting complex II chlorophyll a/b binding protein 1	7	3

Table S4. Candidate genes related to flavonoids biosynthesis pathway.

Function	Gene	Enzyme	Total No.	DEG No.
Phenylpropanoid synthesis pathway	<i>PAL</i>	Phenylalanine ammonia-lyase	6	2
	<i>4CL</i>	4-Coumarate: CoA ligase	5	0
Flavonoid synthesis pathway	<i>CHS</i>	Chalcone synthase	2	0
	<i>CHI</i>	Chalcone isomerase	1	1
	<i>F3H</i>	Flavanone 3-hydroxylase	1	0
	<i>F3'H</i>	Flavonoid 3'-hydroxylase	1	1
	<i>F3'5'H</i>	Flavonoid-3', 5' - hydroxylase	1	1
Anthocyanin synthesis pathway	<i>DFR</i>	Dihydroflavonol 4-reductase	13	11
	<i>ANS</i>	Anthocyanidin synthase	2	0
	<i>UGT</i>	UDP-glucose: anthocyanidin 3-O-glucosyltransferase	1	1
	<i>3MatI</i>	anthocyanin 3-O-glucoside-6'-O-malonyltransferase	1	1
	<i>3GGT</i>	anthocyanidin 3-O-glucoside 2"-O-glucosyltransferase	1	0
Flavonol synthesis	<i>FLS</i>	Flavonol synthase	2	0
Procyanidin	<i>LAR</i>	Leucoanthocyanin reductase	6	0
	<i>ANR</i>	Anthocyanidin reductase	2	0

Table S5. The DEGs of gene family compared H11 (green leaves) and H12 (red leaves).

TF family	H11_vs_H12		Total
	Up	Down	
MYB and MYB-related	9	6	15
bHLH	5	12	17
C2C2	14	13	27
NAC	9	7	16
B3 superfamily	8	5	13
HB	7	2	9
GRAS	4	5	9
AP2/ERF	12	8	20
bZIP	15	5	20

Table S6. The expression profile of EuMYBs and EubHLHs transcripts related to flavonoids synthesis.

Function	Transcript ID	Homologous sequence	H11_tpm	H12_tpm	log2FC (H12/H11)	Significant	Regulate
Anthocyanin biosynthesis	EUC02121	AtMYB75,	2.29	2.52	0.28	no	up
	EUC01949	AtMYB113,	0	0.04	0	no	no change
	EUC20877	AtMYB114	0.02	0.11	0	no	no change
Proanthocyanidin biosynthesis	EUC25930	AtMYB123	16.15	11.74	-0.35	no	down
	EUC03755		0.03	0	0	no	no change
	EUC10757		0.06	0	0	no	no change
	EUC06624		0.08	0	0	no	no change
	EUC18759		0.21	0	0	no	no change
	EUC20063		0.04	0.12	0	no	no change
	EUC26375		0	0	0	no	no change
Flavonol synthesis	EUC03871	AtMYB11	4.54	6.59	0.36	no	up
Flavonoid synthesis	EUC23975	AtMYB4	3.84	9.85	1.22	yes	up
	EUC15883		26.52	20.53	-0.34	no	down
Flavonoid synthesis	EUC06643	bHLH042/TT8	18.98	55.61	1.52	yes	up

Table S7. The node genes of flavonol and anthocyanin biosynthesis.

Gene	Gene Id	Significant	Regulate
<i>EuPAL1</i>	<i>EUC19549</i>	yes	up
<i>EuPAL2</i>	<i>MSTRG.7756</i>	yes	down
<i>EuCHI</i>	<i>EUC08195</i>	yes	up
<i>EuF3'5'H</i>	<i>EUC01284</i>	yes	down
<i>EuF3'H</i>	<i>EUC03526</i>	yes	up
<i>EuDFR1</i>	<i>EUC00272</i>	yes	up
<i>EuDFR2</i>	<i>MSTRG.13706</i>	yes	up
<i>EuDFR3</i>	<i>MSTRG.13719</i>	yes	up
<i>EuDFR4</i>	<i>MSTRG.15870</i>	yes	up
<i>EuDFR5</i>	<i>MSTRG.15871</i>	yes	up
<i>EuDFR6</i>	<i>MSTRG.24809</i>	yes	up
<i>EuDFR7</i>	<i>MSTRG.14354</i>	yes	up
<i>EuDFR8</i>	<i>MSTRG.17635</i>	yes	up
<i>EuDFR9</i>	<i>MSTRG.18190</i>	yes	up
<i>EuDFR10</i>	<i>MSTRG.18191</i>	yes	up
<i>EuDFR11</i>	<i>MSTRG.18192</i>	yes	up
<i>Eu3MaT1</i>	<i>EUC11625</i>	yes	up