

Figure S1. Leaf metabolite OPLS-DA score plot at different developmental stages.

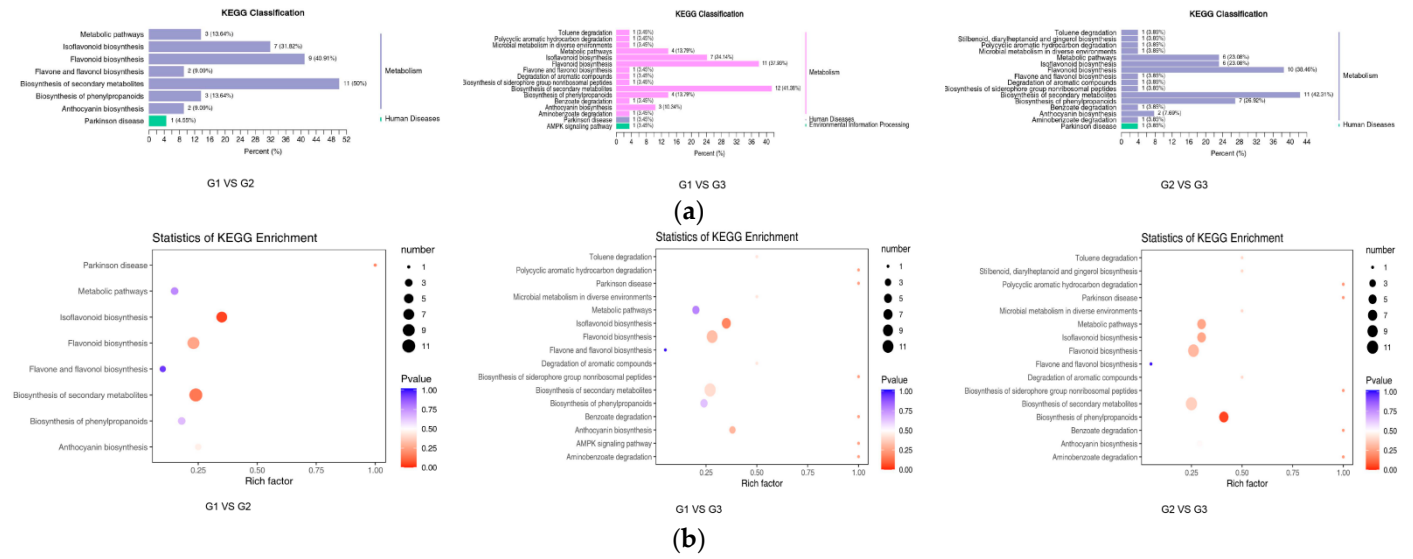
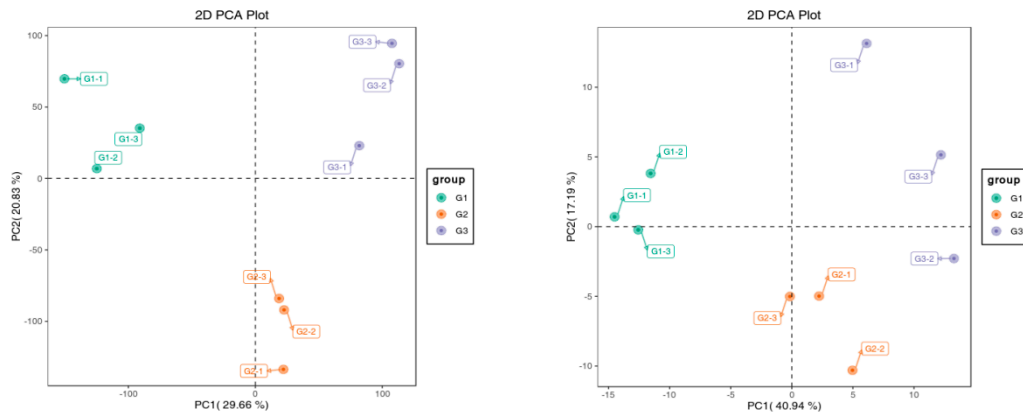


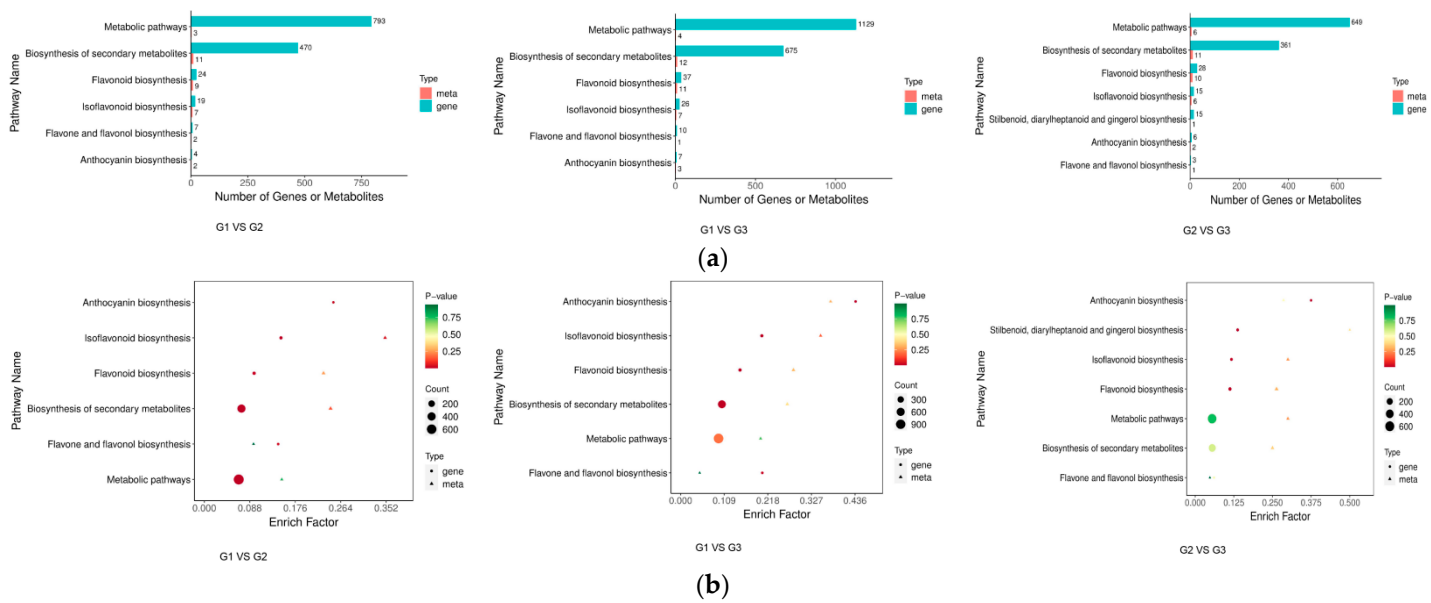
Figure S2. KEGG map of differential metabolites: (a) Differential metabolite KEGG classification chart; (b) Differential metabolite KEGG enrichment plot.



(a)

(b)

**Figure S3.** PCA analysis: (a) Metabolome PCA analysis diagram ; (b) Transcriptome PCA analysis diagram. The horizontal coordinate table represents principal component 1, the vertical coordinate represents principal component 2, and the dots of different colors represent samples of different groups.



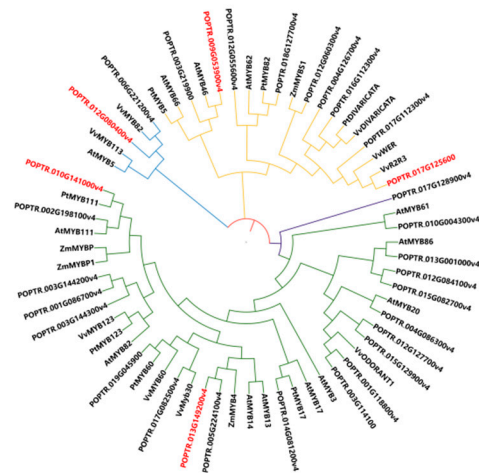
**Figure S4.** Association analysis based on KEGG: (a) KEGG enrichment analysis bar chart; (b) KEGG enrichment bubble diagram.

(a)

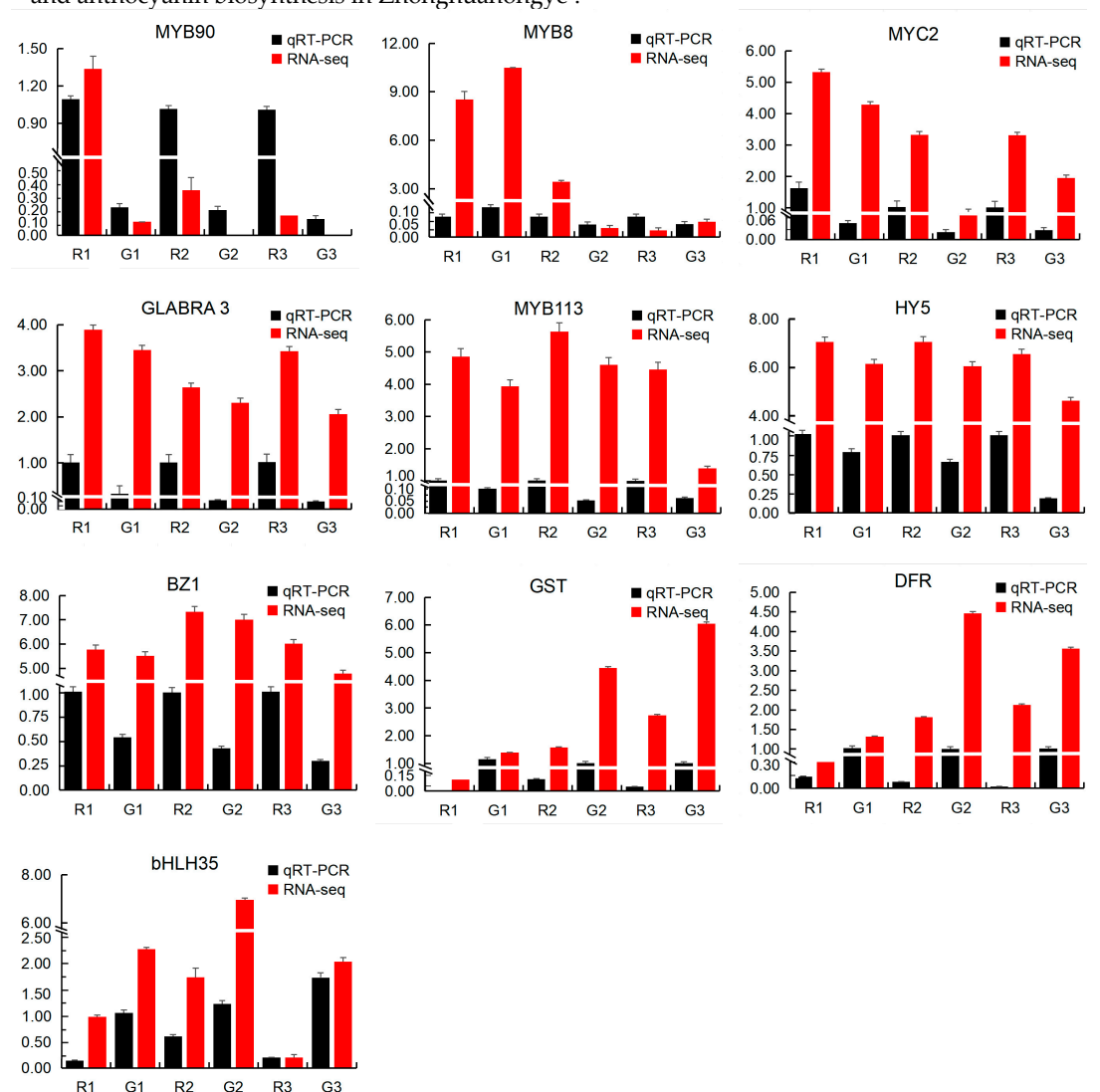
(b)

**Figure S5.** Correlation network diagram.

Note: Metabolites are marked in green squares and genes in red circles. The solid line represents the positive correlation and the dashed line represents the negative correlation.



**Figure S6.** Phylogenetic tree of the MYB transcription factor that related to regulation of flavonoids and anthocyanin biosynthesis in ‘Zhonghuahongye’.



**Figure S7.** Real-time fluorescence quantitative results.

**Table S1.** Primers used in qRT-PCR.

| Gene Name | Gene ID | Forward primer | Reverse primer |
|-----------|---------|----------------|----------------|
|-----------|---------|----------------|----------------|

|  |                    |                          |                           |
|--|--------------------|--------------------------|---------------------------|
| bHLH35   | POPTR_018G141700v4 | TAAGCCAACCATCTCTTCC      | ACTTATATAACAACTATATGGGCTG |
| HY5  | POPTR_018G029500v4 | TAATGAGAGAAACATGATGCAC   | AAATTTCAATAAAGGGGTCCA     |
| MYB90  | POPTR_017G125800v4 | CTGCTATCTGGTTTGCCTC      | TTAATTGTGAACATGCTGCCTA    |
| MYB8   | POPTR_018G049600v4 | CCGTGTGATTTCTTCAACA      | ACTCTTTCTGATTGTGCAT       |
| MYC2   | POPTR_002G176900v4 | TGAGTAAAACCAGCAAATAGCAG  | TAAAAGCTAATGGAACAGGCAAC   |
| GLABRA 3   | POPTR_001G103600v4 | CACTACAGAGAAATGGACCCGAT  | TTTCTAGTCCATAGCTTGCCCAGA  |
| MYB113   | POPTR_017G125600v4 | TGACTATGCTGCTACTACCG     | TAATTTAATAAGTTGCGCTATGGG  |
| DFR<br>(putative<br>anthocyanidin<br>reductase)<br>BZ1<br>(anthocyanidin<br>3-O-glucosyltransferase 7)<br>GST<br>(probable<br>glutathione<br>S-transferase parC) | POPTR_003G138400v4 | AGATTGCGTTAGGAATTCTACCAC | GAGTCTTCAACAGCCAATACCAG   |
|  | POPTR_013G143900v4 | ATAGATATAGCCTTCCTACCAG   | GCCACAGAATTAGATTTCTCCC    |
|  | POPTR_011G113125v4 | TTAGAGTTTTAGGTTTCGGGTAT  | AACCCGAAACCCTAAACCAC      |

**Table S2.** Significant correlation between differential metabolites and differential genes regulating anthocyanin synthesis of 'Zhonghuahongye'.

| Metabolites                                   | Gene ID            | Correlation |
|---|--------------------|-------------|
| Naringenin 7-O-glucoside<br>(Prunin)(pme0371) | POPTR_005G028400   | -           |
|   | POPTR_008G024900   | -           |
|   | POPTR_001G113100v4 | -           |
|   | POPTR_010G056300v4 | +           |
| Dihydromyricetin(pme2898)                     | POPTR_010G056400   | -           |
|   | POPTR_001G304800v4 | -           |
|   | POPTR_005G028400   | +           |
| Hesperetin(pme2319)                           | POPTR_005G028100   | +           |
|   | POPTR_019G001400   | +           |
|   | POPTR_019G001200v4 | +           |

Note: -: negative correlated; +: positive correlated.

**Table S3.** Enzymes bioanabolically associated with anthocyanins of 'Zhonghuahongye'.

| Full name                                     | Abbreviation |
|---|--------------|
| UDP-glucose:flavonoid 3-O-glucosyltransferase | 3GT          |
| 4-Coumarate:CoA ligase                        | 4CL          |
| Anthocyanin synthase                          | ANS          |
| Cinnamate 4-Hydroxylase                       | C4H          |
| Chalcone isomerase                            | CHI          |
| Chalcone reductase                            | CHR          |
| Chalcone synthase                             | CHS          |
| Dihydroflavonol 4-reductase                   | DFR          |

|   |        |
|---|--------|
| Flavonoid-3',5'-hydroxylase               | F3'5'H |
| Flavonoid-3'-hydroxylase                  | F3'H   |
| Flavanone 3-hydroxylase                   | F3H    |
| Flavonol synthase                         | FLS    |
| Ferredoxin-NADP+ oxidoreductase           | FNR    |
| Glycosyl transferases                     | GT     |
| Isoflavone synthase                       | IFS    |
| Leucoanthocyanidin dioxygenase            | LDOX   |
| O -methyltransferase                      | OMT    |
| Phenylalaninammo—Nialyase                 | PAL    |
| UDP-glucose flavonoid glycosyltransferase | UGFT   |

**Table S4.** Differentially expressed genes.

|                    |                    |                    |
|--------------------|--------------------|--------------------|
| POPTR_006G126800v4 | POPTR_010G223900v4 | POPTR_010G224200   |
| POPTR_001G036900v4 | POPTR_003G188500   | POPTR_004G082000v4 |
| POPTR_004G102000v4 | POPTR_006G036200v4 | POPTR_006G169700   |
| POPTR_001G051500   | POPTR_002G141400v3 | POPTR_009G128800   |
| POPTR_012G138800v4 | POPTR_005G028100   | POPTR_005G028400   |
| POPTR_005G028500v3 | POPTR_019G001200v4 | POPTR_019G001400   |
| POPTR_001G371900   | POPTR_002G125400v4 | POPTR_009G133300v4 |
| POPTR_001G113100v4 | POPTR_001G165200   | POPTR_019G078400v4 |
| POPTR_019G087300v4 | POPTR_019G108900v4 | POPTR_011G047800   |
| POPTR_001G113100v4 | POPTR_003G119100v4 | POPTR_006G101100v4 |
| POPTR_016G117100v4 | POPTR_001G304800v4 | POPTR_004G050500v4 |
| POPTR_008G136600v3 | POPTR_011G059500v4 | POPTR_012G006400v4 |
| POPTR_013G121300v4 | POPTR_013G122900   | POPTR_013G129500v4 |
| POPTR_019G093000v4 | POPTR_019G093200v3 | POPTR_001G113100v4 |
| POPTR_003G119100v4 | POPTR_006G101100v4 | POPTR_008G069300v4 |
| POPTR_016G117100v4 | POPTR_003G155200v3 | POPTR_003G155300v4 |
| POPTR_006G219600v4 | POPTR_001G152500v4 | POPTR_003G155300v4 |
| POPTR_005G028200v4 | POPTR_006G034100v3 | POPTR_008G069300v4 |
| POPTR_011G124300   | POPTR_001G023800   | POPTR_001G214600v4 |
| POPTR_006G195300v4 | POPTR_016G060700   | POPTR_010G056300v4 |
| POPTR_010G056400   | POPTR_011G124268v4 | POPTR_013G027000v4 |
| POPTR_001G113100v4 | POPTR_005G028400   | POPTR_008G024900   |
| POPTR_009G128800   | POPTR_010G056300v4 | POPTR_009G069100   |
| POPTR_001G333400   | POPTR_001G304800v4 | POPTR_010G056400   |
| POPTR_004G139700v4 | POPTR_002G125400v4 | POPTR_010G125400v3 |
| POPTR_001G113100v4 | POPTR_003G119100v4 | POPTR_006G069600v4 |
| POPTR_019G061700v4 | POPTR_019G071200v3 | POPTR_008G116500v4 |
| POPTR_013G121400   | POPTR_013G122100v4 |                    |