

**Supplementary Table S1.** Classification and identification of differential metabolites in Zikui (ZK) and N61 tea plants.

Type	Number	Percentage
All	446	100
Phenolic acids	73	16.368
Amino Acid And Its Derivatives	54	12.108
<b>Flavonoid</b>	<b>34</b>	<b>7.623</b>
Nucleotide and its derivates	30	6.726
Organic acids	29	6.502
Flavonols	27	6.054
Alcohols and polyols	23	5.157
Free fatty acids	23	5.157
Flavonoid carbonoside	19	4.26
flavanols	15	3.363
LPC	14	3.139
Others	14	3.139
<b>Proanthocyanidins</b>	<b>14</b>	<b>3.139</b>
Alkaloids	11	2.466
Lignans	8	1.794
Dihydro flavonoid	7	1.57

Glycerol ester	7	1.57
<b>Anthocyanins</b>	<b>6</b>	<b>1.345</b>
Dihydroflavonol	5	1.121
LPE	5	1.121
Isoflavones	4	0.897
Phenolamine	4	0.897
vitamin	4	0.897
Coumarins	3	0.673
Sphingolipids	3	0.673
Chalcone	2	0.448
PC	2	0.448
Plumerane	2	0.448
Orange Flavonoids	1	0.224
Steroid	1	0.224
Tannin	1	0.224
Xanthone	1	0.224

---

**Supplementary Table S2.** Anthocyanin and proanthocyanidin metabolites content in Zikui (ZK) and N61 tea plants in 15-d-old leaves.

Index	15 d N61	15 d ZK	log2_FC(15 d ZK / 15 d N61)	Compounds	Class II
mws0997	3.36E+05	3.96E+07	6.88	Petunidin 3- <i>O</i> -glucoside	Anthocyanins
mws1048	1.48E+06	1.58E+07	3.42	Cyanidin3- <i>O</i> -galactoside	Anthocyanins
pmb0550	1.58E+06	1.44E+07	3.19	Cyanidin 3- <i>O</i> -glucoside	Anthocyanins
pmp000004	6.38E+05	2.83E+06	2.15	Jaceosidin	Anthocyanins
mws0171	6.37E+06	8.18E+06	0.36	Cyanin chloride	Anthocyanins
mws0169	3.41E+06	3.18E+06	-0.10	Cyanidin 3–rutinoside	Anthocyanins
mws0836	8.54E+05	7.51E+05	-0.18	Procyanidin B1	Proanthocyanidins
pmb0837	1.33E+05	8.67E+04	-0.62	Procyanidin A3	Proanthocyanidins
pmn001647	6.80E+05	4.07E+05	-0.74	procyanidin C2	Proanthocyanidins
pmb2947	1.45E+06	8.18E+05	-0.82	Catechin-catechin-catechin	Proanthocyanidins
pmp000280	2.17E+06	1.20E+06	-0.86	procyanidin C-2	Proanthocyanidins
pme0436	3.26E+07	1.71E+07	-0.93	Procyanidin B3	Proanthocyanidins
pmp000275	5.99E+04	3.14E+04	-0.93	gambiriin A-1	Proanthocyanidins
pmp000274	1.21E+07	5.59E+06	-1.11	procyanidin B-3	Proanthocyanidins
pmn001646	5.15E+05	1.83E+05	-1.49	procyanidin C1	Proanthocyanidins
mws1293	5.71E+06	1.47E+06	-1.95	Theaflavin	Proanthocyanidins

pmp000273	1.55E+05	2.57E+04	-2.59	gambiridin B-3	Proanthocyanidins
mws1295	2.19E+06	2.99E+05	-2.87	Theaflavin-3'-gallate	Proanthocyanidins
mws1294	2.82E+06	3.79E+05	-2.89	Theaflavin-3-gallate	Proanthocyanidins
mws1296	5.02E+06	4.31E+05	-3.54	Theaflavin-3-3'-digallate	Proanthocyanidins

Key metabolites are enclosed in red boxes.

**Supplementary Table S3.** Anthocyanin and proanthocyanidin metabolite content in Zikui (ZK) tea plant at 15 d and 45 d.

Index	15 d ZK	45 d ZK	log2_FC(15 d ZK /45 d ZK)	Compounds	Class II
mws1293	1.47E+06	2.12E+04	6.12	Theaflavin	Proanthocyanidins
mws1294	3.79E+05	7.66E+03	5.63	Theaflavin-3-gallate	Proanthocyanidins
mws1295	2.99E+05	8.90E+03	5.07	Theaflavin-3'-gallate	Proanthocyanidins
mws1296	4.31E+05	1.58E+04	4.77	Theaflavin-3-3'-digallate	Proanthocyanidins
mws1048	1.58E+07	3.20E+06	2.30	Cyanidin 3- <i>O</i> -galactoside	Anthocyanins
pmb0550	1.44E+07	3.32E+06	2.12	Cyanidin 3- <i>O</i> -glucoside	Anthocyanins
mws0997	3.96E+07	1.14E+07	1.80	Petunidin 3- <i>O</i> -glucoside	Anthocyanins
pmp000004	2.83E+06	1.28E+06	1.14	Jaceosidin	Anthocyanins
pmb2947	8.18E+05	5.89E+05	0.47	Catechin-catechin-catechin	Proanthocyanidins
pmb0837	8.67E+04	6.67E+04	0.38	Procyanidin A3	Proanthocyanidins
mws0836	7.51E+05	7.34E+05	0.03	Procyanidin B1	Proanthocyanidins
pmp000280	1.20E+06	1.18E+06	0.02	procyanidin C-2	Proanthocyanidins
pmp000274	5.59E+06	5.69E+06	-0.03	procyanidin B-3	Proanthocyanidins
pmn001646	1.83E+05	2.35E+05	-0.36	procyanidin C1	Proanthocyanidins
pme0436	1.71E+07	2.46E+07	-0.52	Procyanidin B3	Proanthocyanidins
pmn001647	4.07E+05	7.24E+05	-0.83	procyanidin C2	Proanthocyanidins

pmp000275	3.14E+04	7.52E+04	-1.26	gambiriin A-1	Proanthocyanidins
pmp000273	2.57E+04	6.85E+04	-1.41	gambiriin B-3	Proanthocyanidins
mws0169	3.18E+06	9.00E+06	-1.50	Cyanidin 3-rutinoside	Anthocyanins
mws0171	8.18E+06	2.41E+07	-1.56	Cyanin chloride	Anthocyanins

---

Key metabolites are enclosed in red boxes.

**Supplementary Table S4.** Base information statistics.

Sample	Raw Data (bp)	Clean Data (bp)	AF_Q20 (%)	AF_Q30 (%)	AF_GC (%)
15 d ZK-1	7760782200	7728994925	98.05	94.30	47.53
15 d ZK-2	8149985700	8113066866	96.19	90.25	47.17
15 d ZK-3	7440591000	7408970072	98.03	94.29	47.34
45 d ZK-1	8391961800	8366095572	98.09	94.36	47.70
45 d ZK-2	7894026900	7867425490	98.03	94.20	47.62
45 d ZK-3	9351836400	9322501197	97.83	93.75	47.36
15 d N61-1	9682011900	9642763026	97.79	93.68	46.98
15 d N61-2	9845860500	9805718151	98.08	94.36	47.95
15 d N61-3	10474228800	10438291339	97.95	94.09	46.68
45 d N61-1	10721295300	10692850135	98.03	94.24	48.07
45 d N61-2	9989629800	9960436284	98.05	94.33	48.00
45 d N61-3	10472289300	10443240899	97.78	93.70	48.01

**Supplementary Table S5.** Pearson correlation coefficient between positively correlated metabolites and genes.

Gene Name	Gene ID	mws0997		mws1048		pmb0550		Annotation
		PPC	<i>P</i> -value	PPC	<i>P</i> -value	PPC	<i>P</i> -value	
<i>CsLIMYBLX4</i>	MSTRG.47184	0.90	5.20E-05	0.99	8.35E-10	0.98	6.21E-08	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X4 [ <i>Oryza brachyantha</i> ]
<i>CsMYB90</i>	MSTRG.14584	0.80	1.79E-03	0.96	8.93E-07	0.92	2.18E-05	anthocyanin 1 [ <i>Camellia sinensis</i> ]
<i>CsLIMYBL1</i>	MSTRG.15849	0.91	3.47E-05	0.91	3.23E-05	0.93	1.58E-05	PREDICTED: L10-interacting MYB domain-containing protein-like [ <i>Nicotiana attenuata</i> ]
<i>CsLIMYBL2</i>	MSTRG.32788	0.85	4.29E-04	0.92	2.83E-05	0.91	5.03E-05	PREDICTED: L10-interacting MYB domain-containing protein-like, partial [ <i>Setaria italica</i> ]
<i>CsMYB4</i>	TEA_029924	0.83	8.87E-04	0.93	1.27E-05	0.91	3.11E-05	PREDICTED: myb-related protein Myb4 [ <i>Theobroma cacao</i> ]
<i>CsBHLH018</i>	MSTRG.29737	0.83	7.47E-04	0.96	9.91E-07	0.93	9.96E-06	transcription factor BHLH018, partial [ <i>Vaccinium corymbosum</i> ]
<i>CsBHLH51</i>	TEA_000397	0.71	9.97E-03	0.87	1.95E-04	0.86	3.91E-04	PREDICTED: transcription factor bHLH51 [ <i>Vitis vinifera</i> ]
<i>CsBHLH49</i>	MSTRG.19098	0.74	5.81E-03	0.82	1.21E-03	0.81	1.51E-03	PREDICTED: transcription factor bHLH49 isoform X2 [ <i>Vitis vinifera</i> ]
<i>CsLIMYBLX2</i>	MSTRG.45518	0.79	2.23E-03	0.82	1.16E-03	0.81	1.50E-03	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X2 [ <i>Glycine max</i> ]
<i>CsANS2</i>	MSTRG.17031	0.67	1.82E-02	0.84	7.18E-04	0.79	2.46E-03	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsANS3</i>	TEA_014163	0.60	4.10E-02	0.78	2.98E-03	0.75	5.33E-03	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsF3'H</i>	TEA_014906	0.71	1.36E-02	0.83	7.86E-04	0.81	1.51E-03	flavonoid 3'-hydroxylase 1 [ <i>Camellia sinensis</i> ]
<i>CsF3'HI</i>	TEA_009006	0.61	1.99E-01	0.81	5.01E-02	0.78	6.96E-02	flavonoid 3'-hydroxylase 1 [ <i>Camellia sinensis</i> ]

PPC : Pearson correlation coefficient. mws0997, mws1048 and pmb0550 respectively represent the positively correlated metabolites petunidin 3-*O*-glucose, cyanidin 3-*O*-galactoside, and cyanidin 3-*O*-glucoside.

**Supplementary Table S6.** Pearson correlation coefficient between negatively correlated metabolites and genes.

Gene Name	Gene ID	mws1293		mws1295		mws1296		Annotation
		PPC	<i>P</i> -value	PPC	<i>P</i> -value	PPC	<i>P</i> -value	
<i>CsLIMYBX3</i>	TEA_001647	0.93	1.53E-05	0.96	5.66E-07	0.94	4.37E-06	L10-interacting MYB domain-containing protein-like isoform X3 [ <i>Ananas comosus</i> ]
<i>CsBHLH25</i>	TEA_014434	0.92	1.97E-05	0.93	8.65E-06	0.88	1.44E-04	PREDICTED: transcription factor bHLH18 [ <i>Theobroma cacao</i> ]
<i>CsPPO4</i>	TEA_004892	0.90	6.36E-05	0.91	3.39E-05	0.85	4.24E-04	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO5</i>	TEA_024006	0.94	6.83E-06	0.94	7.80E-06	0.89	1.23E-04	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO6</i>	TEA_022444	0.95	1.69E-06	0.97	2.51E-07	0.94	6.20E-06	polyphenoloxidase [ <i>Camellia nitidissima</i> ]

PPC : Pearson correlation coefficient. mws1293, mws1295, and mws1296 represent negatively correlated metabolites theaflavin, theaflavin-3'-gallate, and theaflavin-3, 3'-digallate, respectively.

**Supplementary Table S7.** Expression levels of genes with strong correlations with key metabolites in Zikui (ZK) and N61 tea plants at 15 d.

Gene Name	Gene ID	15 d N61 (mean-FPKM)	15 d ZK (mean-FPKM)	log <sub>2</sub> FC (15 d ZK/15 d N61)	P-value	Regulated	Annotation
<i>CsLIMYBLX4</i>	MSTRG.47184	0.01	16.81	10.30	5.81E-34	up	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X4 [ <i>Oryza brachyantha</i> ]
<i>CsMYB90</i>	MSTRG.14584	1.97	34.47	4.13	3.2E-113	up	anthocyanin 1 [ <i>Camellia sinensis</i> ]
<i>CsLIMYBL1</i>	MSTRG.15849	0.52	6.14	3.55	1.85E-32	up	PREDICTED: L10-interacting MYB domain-containing protein-like [ <i>Nicotiana attenuata</i> ]
<i>CsLIMYBL2</i>	MSTRG.32788	5.62	17.52	1.64	2.4E-35	up	PREDICTED: L10-interacting MYB domain-containing protein-like, partial [ <i>Setaria italica</i> ]
<i>CsMYB4</i>	TEA_029924	2.18	6.35	1.54	1.62E-12	up	PREDICTED: myb-related protein Myb4 [ <i>Theobroma cacao</i> ]
<i>CsBHLH018</i>	MSTRG.29737	0.00	2.11	11.05	3.04E-13	up	transcription factor BHLH018, partial [ <i>Vaccinium corymbosum</i> ]
<i>CsBHLH51</i>	TEA_000397	1.78	5.89	1.72	6.45E-09	up	PREDICTED: transcription factor bHLH51 [ <i>Vitis vinifera</i> ]
<i>CsBHLH49</i>	MSTRG.19098	1.57	3.14	1.00	9.39E-06	up	PREDICTED: transcription factor bHLH49 isoform X2 [ <i>Vitis vinifera</i> ]
<i>CsLIMYBLX2</i>	MSTRG.45518	0.59	3.78	2.68	8.47E-17	up	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X2 [ <i>Glycine max</i> ]
<i>CsANS2</i>	MSTRG.17031	103.66	238.45	1.20	4.85E-22	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsANS3</i>	TEA_014163	10.84	18.50	0.77	1.13E-14	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsF3'H</i>	TEA_014906	0.51	1.05	1.04	0.00463	up	flavonoid 3'-hydroxylase 1 [ <i>Camellia sinensis</i> ]

<i>CsF3'H1</i>	TEA_009006	74.11	116.83	0.66	1.34E-12	up	flavonoid 3'-hydroxylase 3 [ <i>Camellia sinensis</i> ]
<i>CsLIMYBX3</i>	TEA_001647	13.13	0.03	-8.77	2.88E-22	down	L10-interacting MYB domain-containing protein-like isoform X3 [ <i>Ananas comosus</i> ]
<i>CsBHLH25</i>	TEA_014434	8.14	2.49	-1.71	7.58E-15	down	PREDICTED: transcription factor bHLH18 [ <i>Theobroma cacao</i> ]
<i>CsPPO4</i>	TEA_004892	42.29	5.01	-3.08	6.84E-65	down	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO5</i>	TEA_024006	12.37	2.94	-2.07	5.27E-15	down	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO6</i>	TEA_022444	14.79	1.64	-3.17	7.99E-17	down	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
-	MSTRG.29079	0.03	3.57	6.74	6.13E-07	up	-
-	MSTRG.17415	0.54	5.15	3.25	1.17E-05	up	-
-	MSTRG.35246	3.03	8.56	1.50	5.02E-09	up	-
-	TEA_003607	7.63	32.39	2.09	4.91E-14	up	-
-	MSTRG.27856	0.24	5.65	4.56	1.09E-16	up	-
-	MSTRG.6132	0.06	4.20	6.21	1.19E-26	up	-
-	MSTRG.30086	0.08	1.28	4.06	0.007107	up	-
-	MSTRG.31110	0.00	5.55	12.44	2.33E-17	up	-
-	MSTRG.2094	0.36	3.00	3.05	2.28E-07	up	-
-	MSTRG.12458	0.08	1.30	3.97	6.1E-05	up	-
-	MSTRG.33921	2.26	0.78	-1.54	2.55E-04	down	-
-	MSTRG.29824	0.94	0.08	-3.62	1.32E-02	down	-

-	MSTRG.14480	1.79	0.60	-1.57	1.34E-02	down	-
-	MSTRG.1185	2.65	0.51	-2.38	1.78E-03	down	-
-	MSTRG.38417	2.40	1.18	-1.03	2.50E-02	down	-
-	MSTRG.4356	5.79	2.05	-1.50	1.46E-07	down	-
-	MSTRG.46725	7.71	0.82	-3.24	9.07E-25	down	-
-	MSTRG.45508	0.97	0.42	-1.22	1.60E-01	down	-
-	TEA_019718	0.39	0.06	-2.63	5.69E-03	down	-

---

**Supplementary Table S8.** Expression levels of genes strongly related to key metabolites in Zikui (ZK) at 15 d and 45 d.

Gene Name	Gene ID	15 d ZK (mean-FPKM)	45 d ZK (mean-FPKM)	log <sub>2</sub> FC(15 d ZK/45 d ZK)	P-value	Regulated	Annotation
<i>CsLIMYBLX4</i>	MSTRG.47184	16.81	3.10	2.44	1.19E-15	up	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X4 [ <i>Oryza brachyantha</i> ]
<i>CsMYB90</i>	MSTRG.14584	34.47	0.16	7.75	2.81E-53	up	anthocyanin 1 [ <i>Camellia sinensis</i> ]
<i>CsLIMYBL1</i>	MSTRG.15849	6.14	3.32	0.89	9.46E-03	up	PREDICTED: L10-interacting MYB domain-containing protein-like [ <i>Nicotiana attenuata</i> ]
<i>CsLIMYBL2</i>	MSTRG.32788	17.52	10.46	0.74	4.81E-03	up	PREDICTED: L10-interacting MYB domain-containing protein-like, partial [ <i>Setaria italica</i> ]
<i>CsMYB4</i>	TEA_029924	6.35	0.69	3.21	2.55E-22	up	PREDICTED: myb-related protein Myb4 [ <i>Theobroma cacao</i> ]
<i>CsBHLH018</i>	MSTRG.29737	2.11	0.50	2.08	4.42E-05	up	transcription factor BHLH018, partial [ <i>Vaccinium corymbosum</i> ]
<i>CsBHLH51</i>	TEA_000397	5.89	0.52	3.51	7.33E-10	up	PREDICTED: transcription factor bHLH51 [ <i>Vitis vinifera</i> ]
<i>CsBHLH49</i>	MSTRG.19098	3.14	2.02	0.64	1.52E-01	up	PREDICTED: transcription factor bHLH49 isoform X2 [ <i>Vitis vinifera</i> ]
<i>CsLIMYBLX2</i>	MSTRG.45518	3.78	1.96	0.95	4.51E-03	up	PREDICTED: L10-interacting MYB domain-containing protein-like isoform X2 [ <i>Glycine max</i> ]
<i>CsANS2</i>	MSTRG.17031	238.45	97.25	1.29	1.83E-03	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsANS3</i>	TEA_014163	18.50	0.95	4.28	6.07E-62	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsF3'H</i>	TEA_014906	1.05	0.64	0.71	2.51E-01	up	flavonoid 3'-hydroxylase 1 [ <i>Camellia sinensis</i> ]
<i>CsF3'H1</i>	TEA_009006	116.83	28.42	2.04	3.76E-18	up	flavonoid 3'-hydroxylase 3 [ <i>Camellia sinensis</i> ]

<i>CsLIMYBX3</i>	TEA_001647	0.03	0.01	1.58	7.61E-01	up	L10-interacting MYB domain-containing protein-like isoform X3 [ <i>Ananas comosus</i> ]
<i>CsBHLH25</i>	TEA_014434	2.49	1.96	0.34	7.68E-01	up	PREDICTED: transcription factor bHLH18 [ <i>Theobroma cacao</i> ]
<i>CsPPO4</i>	TEA_004892	5.01	2.64	0.92	5.91E-03	up	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO5</i>	TEA_024006	2.94	0.09	4.98	5.43E-19	up	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
<i>CsPPO6</i>	TEA_022444	1.64	0.00	10.68	7.84E-06	up	polyphenoloxidase [ <i>Camellia nitidissima</i> ]
-	MSTRG.29079	3.57	0.32	3.50	2.34E03	up	-
-	MSTRG.17415	5.15	2.59	0.99	1.44E01	up	-
-	MSTRG.35246	8.56	3.70	1.21	9.01E03	up	-
-	TEA_003607	32.39	0.21	7.25	2.20E09	up	-
-	MSTRG.27856	5.65	2.30	1.30	2.79E02	up	-
-	MSTRG.6132	4.20	0.12	5.09	5.49E11	up	-
-	MSTRG.30086	1.28	0.09	3.83	3.50E02	up	-
	MSTRG.31110	5.55	3.03	0.87	2.46E02	up	-
-	MSTRG.2094	3.00	0.91	1.72	1.53E02	up	-
-	MSTRG.12458	1.30	0.00	10.35	3.06E05	up	-
-	MSTRG.33921	0.78	0.26	1.56	5.44E02	up	-
-	MSTRG.29824	0.08	0.00	6.26	5.64E01	up	-
-	MSTRG.14480	0.60	0.23	1.39	2.72E01	up	-
-	MSTRG.1185	0.51	0.00	8.99	1.47E02	up	-

-	MSTRG.38417	1.18	0.17	2.76	2.89E03	up	-
-	MSTRG.4356	2.05	0.57	1.85	1.78E02	up	-
-	MSTRG.46725	0.82	0.06	3.69	3.84E04	up	-
-	MSTRG.45508	0.42	0.00	8.70	3.74E03	up	-
-	TEA_019718	0.06	0.00	5.98	1.50E01	up	-

---

**Supplementary Table S9.** Pearson correlation coefficient between positively correlated metabolites and unannotated genes.

Gene Name	Gene ID	mws0997		mws1048		pmb0550		Annotation
		PPC	P-value	PPC	P-value	PPC	P-value	
-	MSTRG.29079	0.90	8.29E-05	0.91	4.01E-05	0.91	4.05E-05	-
-	MSTRG.17415	0.90	8.39E-05	0.87	2.60E-04	0.88	1.50E-04	-
-	MSTRG.35246	0.89	9.13E-05	0.88	1.39E-04	0.90	6.75E-05	-
-	TEA_003607	0.89	1.19E-04	0.90	5.51E-05	0.91	3.11E-05	-
-	MSTRG.27856	0.89	1.26E-04	0.86	3.65E-04	0.89	1.16E-04	-
-	MSTRG.6132	0.87	2.07E-04	0.87	2.40E-04	0.87	2.33E-04	-
-	MSTRG.30086	0.90	7.02E-05	0.92	2.08E-05	0.95	3.11E-06	-
-	MSTRG.31110	0.86	3.19E-04	0.91	4.05E-05	0.89	8.62E-05	-
-	MSTRG.2094	0.86	3.73E-04	0.84	5.65E-04	0.84	5.76E-04	-
-	MSTRG.12458	0.84	5.97E-04	0.89	1.28E-04	0.87	2.44E-04	-

PPC : Pearson correlation coefficient. mws0997, mws1048 and pmb0550 respectively represent the positively correlated metabolites petunidin 3-*O*-glucose, cyanidin 3-*O*-galactoside, and cyanidin 3-*O*-glucoside.

**Supplementary Table S10.** Pearson correlation coefficient between negatively correlated metabolites and unannotated genes.

Gene Name	Gene ID	mws1293		mws1295		mws1296		Annotation
		PPC	<i>P</i> -value	PPC	<i>P</i> -value	PPC	<i>P</i> -value	
-	MSTRG.33921	0.98	1.09E-08	0.97	7.80E-08	0.94	4.71E-06	-
-	MSTRG.29824	0.98	3.55E-08	0.99	2.21E-09	0.97	2.02E-07	-
-	MSTRG.14480	0.97	9.61E-08	0.97	3.40E-07	0.94	4.80E-06	-
-	MSTRG.1185	0.97	2.22E-07	0.97	7.66E-08	0.97	3.22E-07	-
-	MSTRG.38417	0.97	3.30E-07	0.93	1.03E-05	0.87	2.23E-04	-
-	MSTRG.4356	0.96	4.27E-07	0.96	5.04E-07	0.91	3.34E-05	-
-	MSTRG.46725	0.96	5.40E-07	0.98	4.68E-08	0.93	1.23E-05	-
-	MSTRG.45508	0.96	6.00E-07	0.92	1.74E-05	0.86	3.60E-04	-
-	TEA_019718	0.96	7.84E-07	0.98	2.00E-08	0.96	7.45E-07	-

PPC : Pearson correlation coefficient. mws1293, mws1295, and mws1296 represent negatively correlated metabolites theaflavin, theaflavin-3'-gallate, and theaflavin-3, 3'-digallate, respectively.

**Supplementary Table S11.** Expression levels of structural genes involved in anthocyanin biosynthesis in Zikui (ZK) at 15 d and 45 d.

Gene Name	Gene ID	15 d ZK (mean-FPKM)	45 d ZK (mean-FPKM)	log <sub>2</sub> FC (15 d ZK /45 d ZK)	P-value	Regulated	Annotation
<i>CsPAL1</i>	TEA_003071	35.12	15.47	1.18	3.70E-04	up	phenylalanine ammonia-lyase [ <i>Populus szechuanica</i> ]
<i>CsPAL2</i>	TEA_020823	348.34	73.18	2.25	4.22E-07	up	phenylalanine ammonia-lyase [ <i>Camellia japonica</i> ]
<i>CsPAL3</i>	TEA_022055	101.93	18.54	2.46	3.69E-12	up	phenylalanine ammonia-lyase [ <i>Camellia sinensis</i> ]
<i>CsPAL4</i>	MSTRG.12851	66.50	17.34	1.94	5.20E-06	up	phenylalanine ammonia-lyase [ <i>Camellia sinensis</i> ]
<i>CsC4H1</i>	TEA_013313	551.85	191.38	1.53	2.60E-08	up	cinnamate 4-hydroxylase [ <i>Osmanthus fragrans</i> ]
<i>CsC4H2</i>	MSTRG.6357	351.34	67.81	2.37	1.00E-11	up	cinnamic acid 4-hydroxylase [ <i>Camellia chekiangoleosa</i> ]
<i>CsC4H3</i>	MSTRG.6358	5.34	1.52	1.82	1.41E-04	up	cinnamic acid 4-hydroxylase [ <i>Camellia chekiangoleosa</i> ]
<i>Cs4CL1</i>	TEA_023253	62.54	8.77	2.83	2.42E-32	up	PREDICTED: 4-coumarate--CoA ligase 2-like [ <i>Jatropha curcas</i> ]
<i>Cs4CL2</i>	MSTRG.2903	247.05	41.56	2.57	1.74E-16	up	4-coumaroyl CoA ligase [ <i>Camellia sinensis</i> ]
<i>Cs4CL3</i>	MSTRG.33696	2.11	0.00	11.05	7.37E-03	up	4-coumarate--CoA ligase 2 [ <i>Glycine soja</i> ]
<i>CsF3'H2</i>	TEA_006169	379.61	151.49	1.33	2.42E-03	up	flavonoid 3'-hydroxylase 2 [ <i>Camellia sinensis</i> ]
<i>CsF3'H3</i>	MSTRG.6382	21.65	0.69	4.98	2.01E-24	up	flavanone 3-hydroxylase, partial [ <i>Camellia japonica</i> ]
<i>CsF3'5'H</i>	TEA_011933	1239.64	428.30	1.53	1.21E-11	up	flavonoid 3',5'-hydroxylase [ <i>Camellia sinensis</i> var. <i>sinensis</i> ]
<i>CsANS1</i>	TEA_014156	1.15	0.05	4.43	1.05E-04	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsANS2</i>	MSTRG.17031	238.45	97.25	1.29	1.83E-03	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]
<i>CsANS3</i>	TEA_014163	18.50	0.95	4.28	6.07E-62	up	anthocyanidin synthase [ <i>Camellia sinensis</i> ]

<i>CsUGT</i>	TEA_001487	70.11	1.94	5.17	3.22E-41	up	UDP-glycosyltransferase 78A15 [ <i>Camellia sinensis</i> ]
--------------	------------	-------	------	------	----------	----	--

---

**Supplementary Table S12.** Primer sequences used in the study.

Gene Name	Gene ID	Forward Primer (5'→3')	Reverse Primer (5'→3')	Purpose
<i>TUBA3</i>	TEA_013193	TTAGGACAGGGACTTACCGC	TCCAAAAGCACAGCCACATC	qRT-PCR
<i>CsLIMYBLX4</i>	MSTRG.47184	ACCCGCAAGACCCACTAATC	GCCGTCAGAACCACCCAG	qRT-PCR
<i>CsMYB90</i>	MSTRG.14584	GGTCGTTGATTGCGGGTAGA	GGTAGTGGAAATGGCTCGCT	qRT-PCR
<i>CsBHLH51</i>	TEA_000397	TCGCATAAAAAGCAAGAGGCA	ACTGAGAACTAATCGCTGGAGG	qRT-PCR
<i>CsANS2</i>	MSTRG.17031	CTGCCCTCACCTTCATCCTC	CGCCACGAAATCCTCACTT	qRT-PCR
<i>CSF3'H1</i>	TEA_009006	GGTTAGCAACAGCAGCCATC	TGGGGGAGAGAGCCGATTAG	qRT-PCR
<i>CsLIMYBX3</i>	TEA_001647	ATTTGGTGGTGGTTCGTCCT	TGAGGTTTGTGTTGAAGTTGCT	qRT-PCR
<i>CsBHLH25</i>	TEA_014434	CACTGTCGTTGCTCAGATGG	CACACTTCCTCCGCTCCTG	qRT-PCR
<i>CsPPO4</i>	TEA_004892	ACCACCATTCCCACCCATTC	AGCCTCCTAAGCCAATAAGCA	qRT-PCR
<i>CsMYB90</i>	MSTRG.14584	ATGGACAGTGTTTGCTGTG	TCATTCATCACCTAACAG	Gene cloning

**Supplementary Figure legends:**

**Supplementary Figure S1:** Kyoto Encyclopedia of Genes and Genomes (KEGG) enrichment of differential metabolites in Zikui (ZK) and N61 tea plants at 15 d.

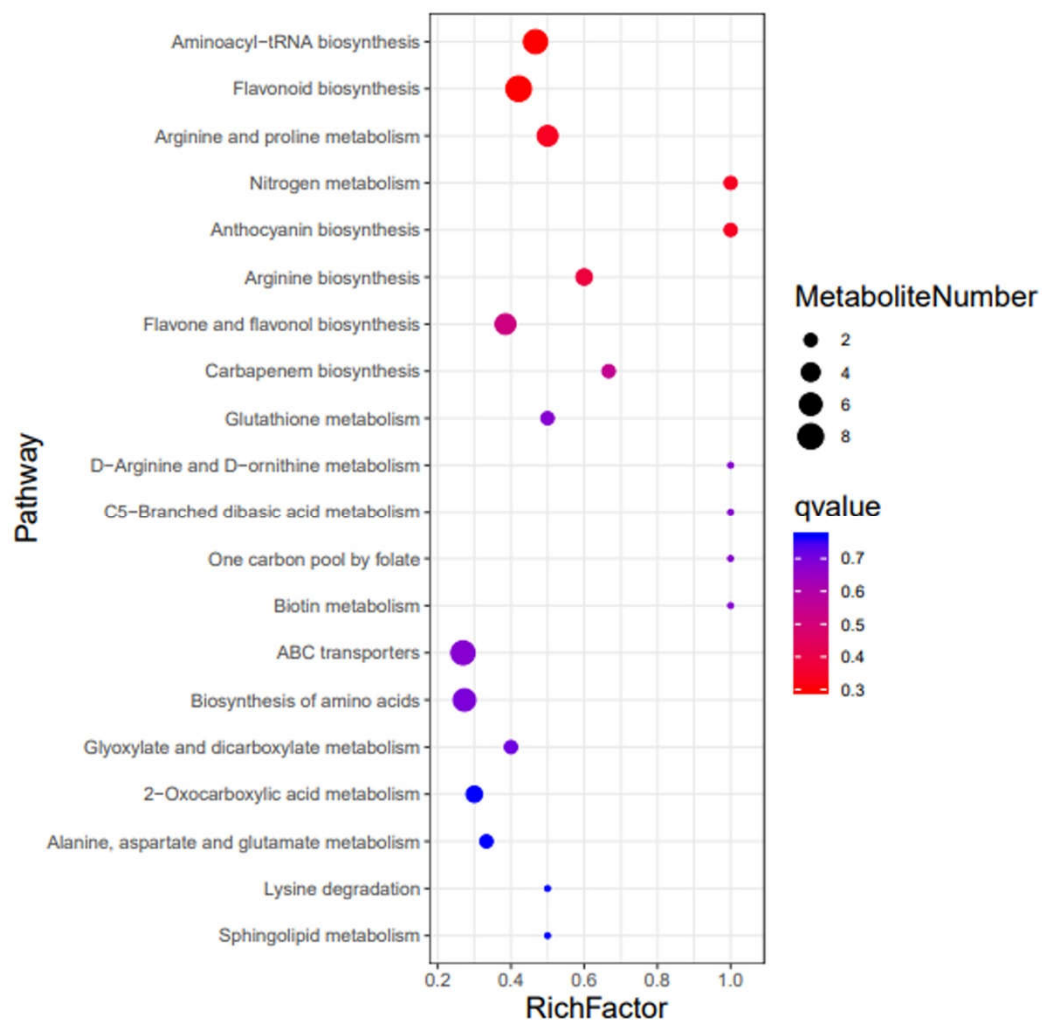
**Supplementary Figure S2:** Gene Ontology (GO) enrichment of differentially expressed genes of Zikui (ZK) and N61 tea plants at 15 d and 45 d. (A) GO enrichment of 15 d N61 vs. 15 d ZK; (B) GO enrichment of 15 d ZK vs. 45 d ZK.

**Supplementary Figure S3:** KEGG enrichment of differentially expressed genes of Zikui (ZK) and N61 tea plants at 15 d and 45 d. The y-coordinate represents the pathway item, and the y-coordinate represents the rich factor. Dots represent the number of genes with significant differential expression, the size of the dot represents the number of genes with significant differential expression, where larger dots indicate more genes with significantly different expression. Dots in different colors represent different q-values.

**Supplementary Figure S4:** Expression level of *CsMYB90* in wild-type (WT) and transgenic *Nicotiana tabacum* 'K326' (n = 3 independent biological samples).

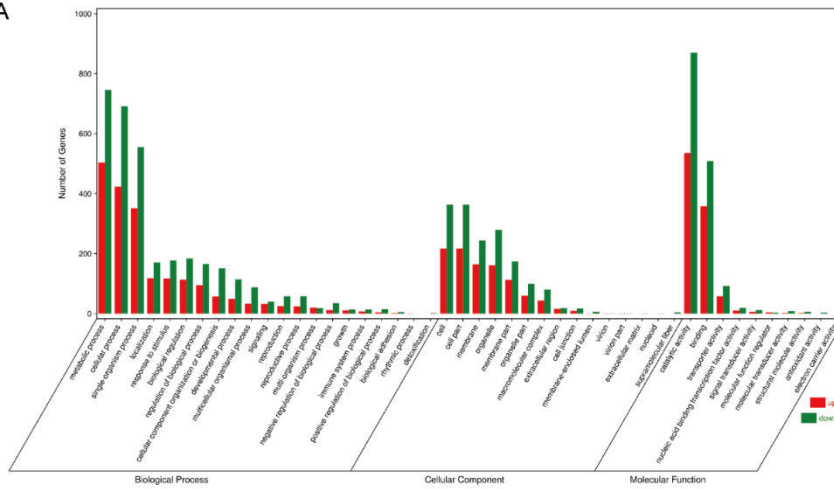
**Supplementary Figure S5:** Metabolomic data of Zikui (ZK) and N61 leaves. (A) PCA (Principal component analysis) clustering based on Zikui (ZK) and N61 metabolomic data; (B) HCA (hierarchical cluster analysis) clustering based on Zikui (ZK) and N61 metabolomic data.

Supplementary Figure S1

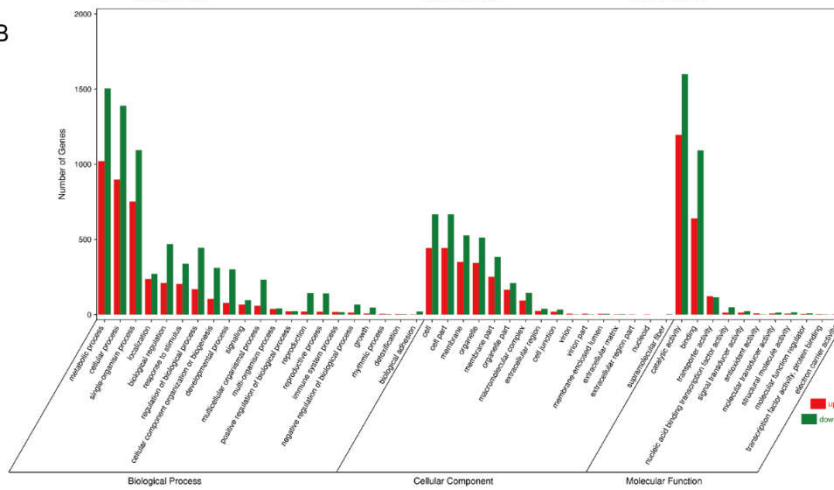


### Supplementary Figure S2

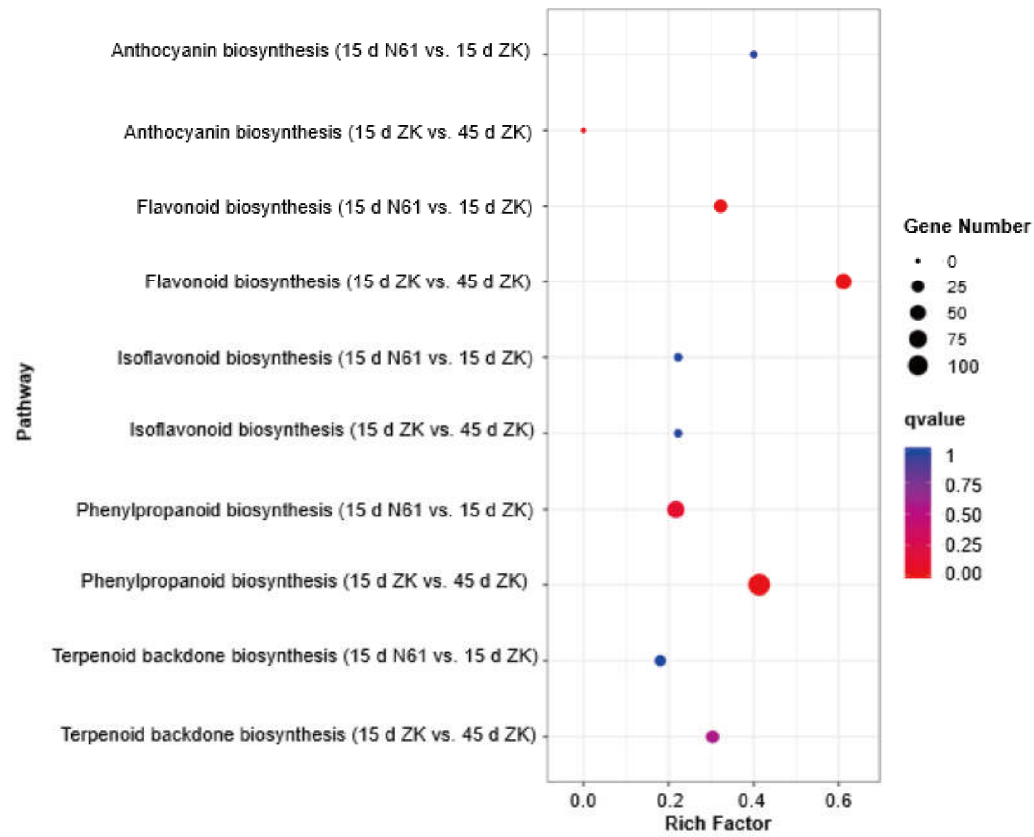
A



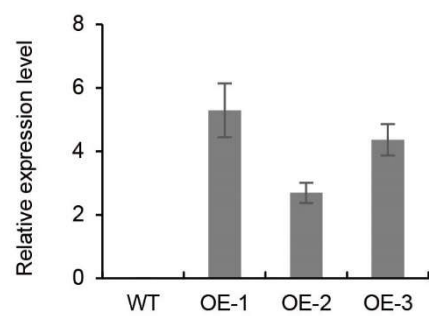
B



Supplementary Figure S3



**Supplementary Figure S4**



Supplementary Figure S5:

