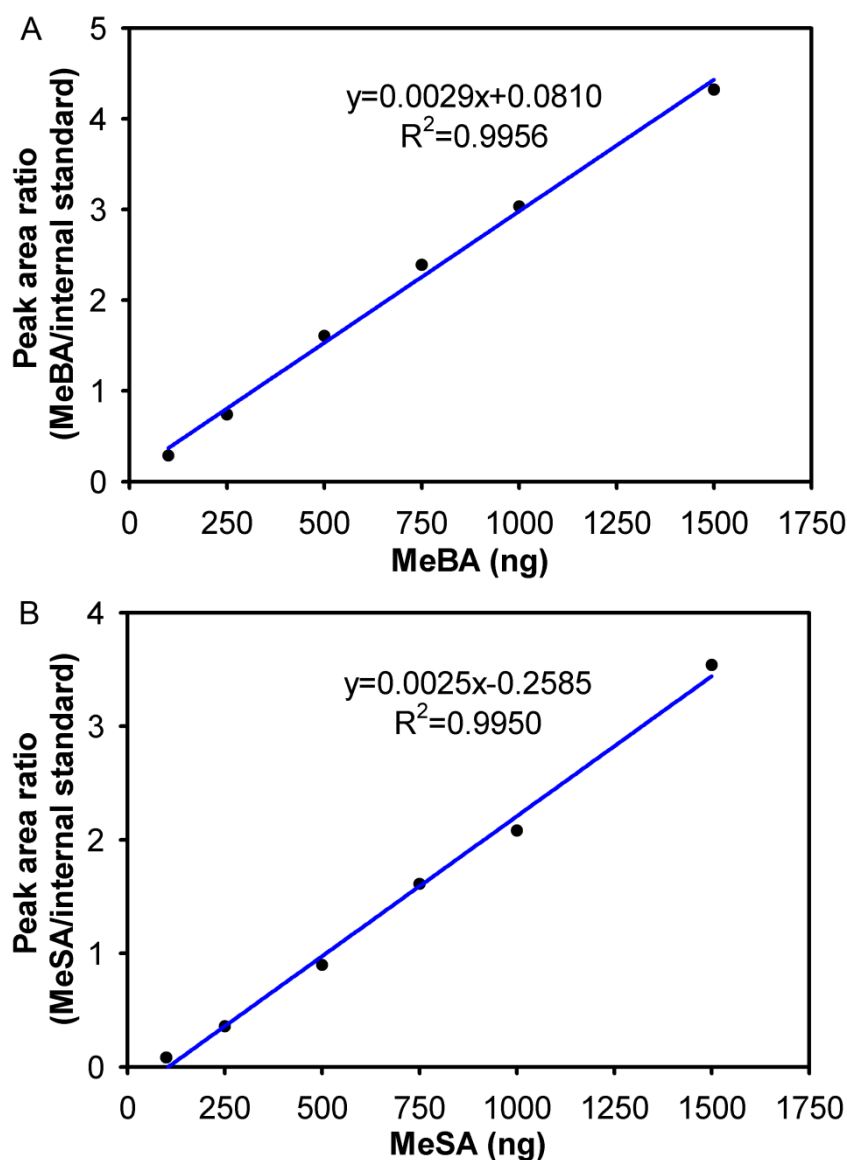
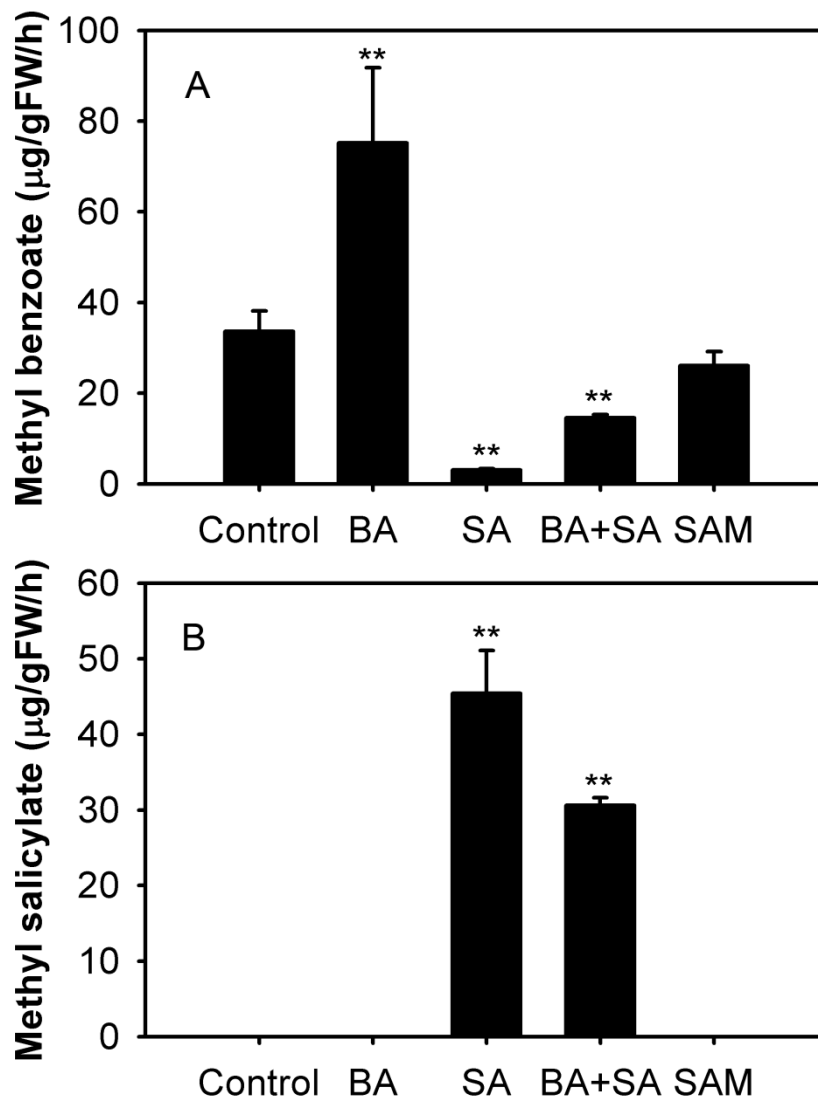


**Supplementary Figure 1.** Representative photographs of different floral organs (A) and flowers at different floral developmental stages (B) in *H. coronarium*. SS: styles and stigmas; A: anthers; F: filaments; L: labella; LP: lateral petals; Se: sepals; B: bracts; Le: leaves. Some small floral tissues are indicated by an arrow.



**Supplementary Figure 2.** Standard curves of methyl benzoate (A) and methyl salicylate (B). The curves were generated by three repeats. The x-axis stands for the content of methyl benzoate or methyl salicylate used for solid-phase microextraction (SPME) and GC-MS detection. The y-axis represents the peak area ratio of methyl benzoate or methyl salicylate to internal standard (IS). Ethyl benzoate (210 ng) was used as IS for methyl benzoate quantification, while methyl benzoate (297.5 ng) as IS for methyl salicylate quantification. Using MS scanning technique of selected ion monitor (SIM), two groups of ions were used to monitor methylated product and internal standard in each sample, respectively. Methyl benzoate was monitored by a group of ions with mass/charge ratio ( $m/z$ ) 77, 105 and 136, ethyl benzoate by a group of ions with  $m/z$  77, 105 and 150, and methyl salicylate by a group of ions with  $m/z$  92, 120 and 152. The scanning dwell time for each ion was set to 100 ms.



**Supplementary Figure 3.** Emission of floral methyl benzoate (A) and methyl salicylate (B) after infiltration of substrates BA, SA and SAM into petals in *H. coronarium*. Error bars indicate standard deviation of three biological replicates. Asterisks represent significant differences between treatments and controls by Student's *t*-test (\*\* indicates a significant difference at the level of  $P < 0.01$ ).

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HcBSMT1 : -----MLPNTTIMEGEMSLKVEHALHMVGGTGETSYVANSRLQEKATHQTKSLLAIAVEETYKALH-----SDQ : 64
HcBSMT2 : -----MGIKVEQALHMVGGSGGETSYATNSRLQEKATYRTRKFLVLAATTEEMKYGLL-----PEH : 53
OsBSMT1 : -----MKVEQDLHMSRCDGETSYAANSRLQEKAILKTRILLHKAVEEAHASLSGLSRAPAGGK : 58
PhBSMT1 : -----MEVEVLEHMNGGNGDSSYANNSILVQOKVILMTKFEITEQAIDLYS-SLFP-----ET : 51
AtBSMT1 : MDPREINTIPSLRYDDDKCDEYAFVKALCMSGGDCANSYSANSRLQKVLVSMAKFVLRNTEEMMNLDFP-----TY : 74
CbSAMT : -----MDVROVLEHMKKGAGENSYAMNSFIQRQVISITKEITEAATALYSGDVTV-----TR : 52

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HcBSMT1 : MVVVDLGCSSGENTLIMLSHVLSVATKLP-RRMEL-----OFFINDLPGNDFNCFQSLIEGKKRTEGEIEGDLIVPY : 137
HcBSMT2 : MVVVDLGCSSGENTLIVVSOVLDIIVELR-RRMEMKKPLEVQFFINDLPGNDFNIVFOQLDKKKNVEESKGELLV : 132
OsBSMT1 : MVVADLGCSSGENTLIVSEVLSAVANRSSCDHKSSILVADVQFFINDLPGNDFNIVFOQLDELKKAEMEF-GKAL : 137
PhBSMT1 : ICIADLGCSLGANTLIVVSOVQVKEKER-KKHGFK-SPEFYFHNDLPGNDFNIVFOQLGAEQEDLRKHI-GESFG : 128
AtBSMT1 : IKVAELGCSSGENSELAIIEIINTINVLC--QHVNKNSPEIDCCINDLPEDFNITFEKVPFENKEIMITN----K : 148
CbSAMT : IAIADLGCSSGENALFAVELIKTVEELR-KKMGRENSEPYQIFINDLPGNDFNATFRSL-PIENDV-----G : 122

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HcBSMT1 : VAGVAGSYFGRLEFRASVLELHSSFCIMWLSQVEEGKIEHGVPLNKGNITWETSLAKVEKAYQOFQKDFSTFL : 217
HcBSMT2 : VVGAGSYFGRLEFPCASVHEFFHSSYCLMWSQVEEELNDQGVSLNKGNITWETSSQVEKAYRQYKRDFTFLS : 212
OsBSMT1 : IAGVAGSYFGRLEFRDPSVLELHSSYCLMWRKSVEDKIAS--GEVINAGNMYTWTTPPSVVKIYQRFQKDF : 215
PhBSMT1 : FSGVAGSYFTRLEFESKLEFVYSSYSLMWSQVENGTEN-----NKGNITWARTSPLSVIKAYYKQYED : 202
AtBSMT1 : VYCAFGSYFGRLEFRNSLLELHSSYALHSLKVEKLEN-----NKGNLYTSSSPQSAKAYLQKDFDTMFL : 222
CbSAMT : IINGVAGSYFGRLEFRNTLLEFHHSSYSLMWSQVEIGLES-----NKGNITWANTCQPSVNLAYYQKQF : 196

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HcBSMT1 : TELNVGCGMVLIVFVGRRRK-TPGNDFVHYGLLIGELNSMVLEGTIQEEKVDTFNLEPTYGASFEVESIHN : 296
HcBSMT2 : TELNIGCGMMLIFLGRRRK-TPGHGDLCHWRLLAEADNSMVLEETIPEEKVSTFNLEPTYGPSLEEVKSIH : 291
OsBSMT1 : DELVSGCGMVLIFELGRKNR-DVLRGEVSY YGLLAQALQSLVQECRVEEKLDSFNLEPTYSVDEVKA : 294
PhBSMT1 : BELMKGCGMVLILLGRESE-DPTSKECCYTWELLAMLNKLVBEGLIKKEKVDAFNIPQYTPSPA : 281
AtBSMT1 : EELVSNCGMVLIFIGRNTLNDPLYRDCCHFWTLNNSRDVLFEGLVSEKLDFAFNMPRYDENVOELKEV : 302
CbSAMT : CEVVPCCGMVLILLGRRSE-DRASTECCLLWQLLAMALNQVSEGLIPEEKMDKENIPQYTPSPTE : 275

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HcBSMT1 : RAEIFESSDPFDSSKDDIYAIPNYMRSKNNVANYIRAVVEPLIVHCFQTVILDDLEARYAHLVSKHLLK : 376
HcBSMT2 : RVEIFESSDPFDSSGDDSFDLSNYTKSAKNVADCIIRAVVEPLIVHCFQDVILLDFETRYAQNVLKHLL : 371
OsBSMT1 : HICLFESNDPQDDSDDDVATLDSVRSQVNVARCIIRAVVEPLIHARFGRCTVDDLEDMYARNVAQHE : 374
PhBSMT1 : RLETSRLVHNNASNNEKNGG-----YNSRRCMRAVAEPLLVSHFDKELMDIVFHYEEIVSDCM : 351
AtBSMT1 : ELESHGFDLGHYYEEDDFEAG-----RNEANGIRAVSEPMLLAHFGEELIDTLEDYAYEVTQHANC : 374
CbSAMT : HLEASEIYSSCTKDGDDGGGSVE---EEGYNVRCMRAVAEPLLDHFCEALIEDVHRYKLLIIRMSKE : 352

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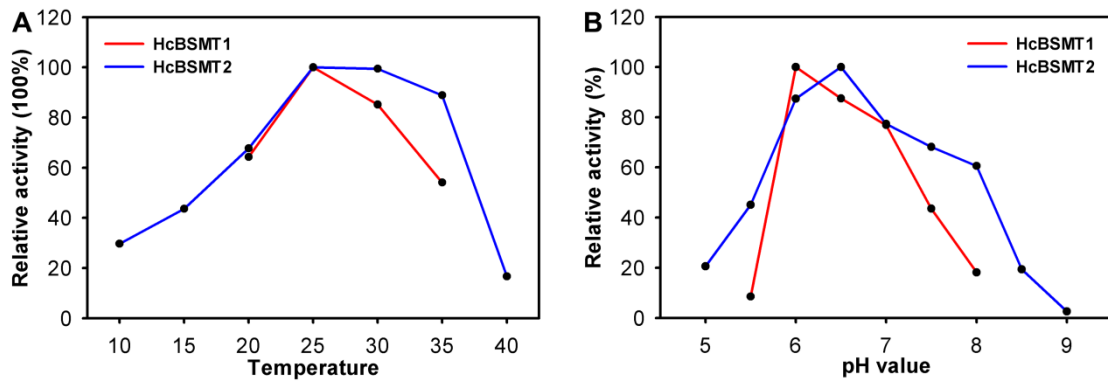
&

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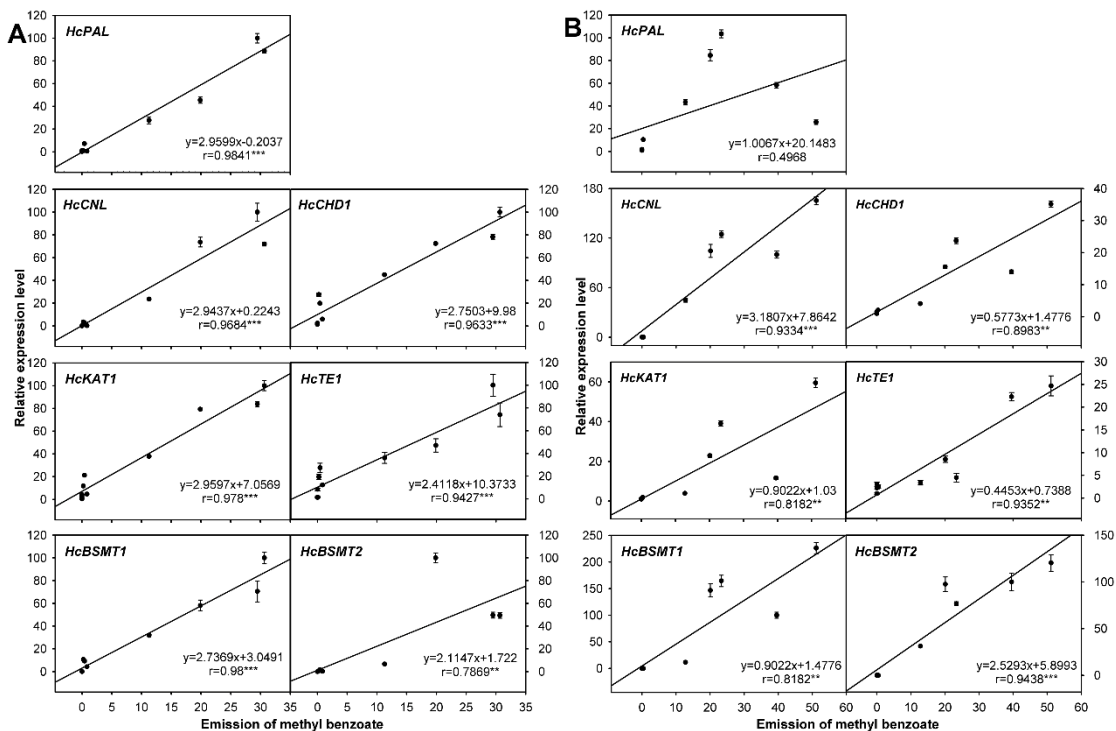
HcBSMT1 : ALKKKA- : 382
HcBSMT2 : ALKKRV- : 377
OsBSMT1 : SLKARR- : 380
PhBSMT1 : SLTKIN- : 357
AtBSMT1 : SLTKK-- : 379
CbSAMT : SLIRKSD : 359

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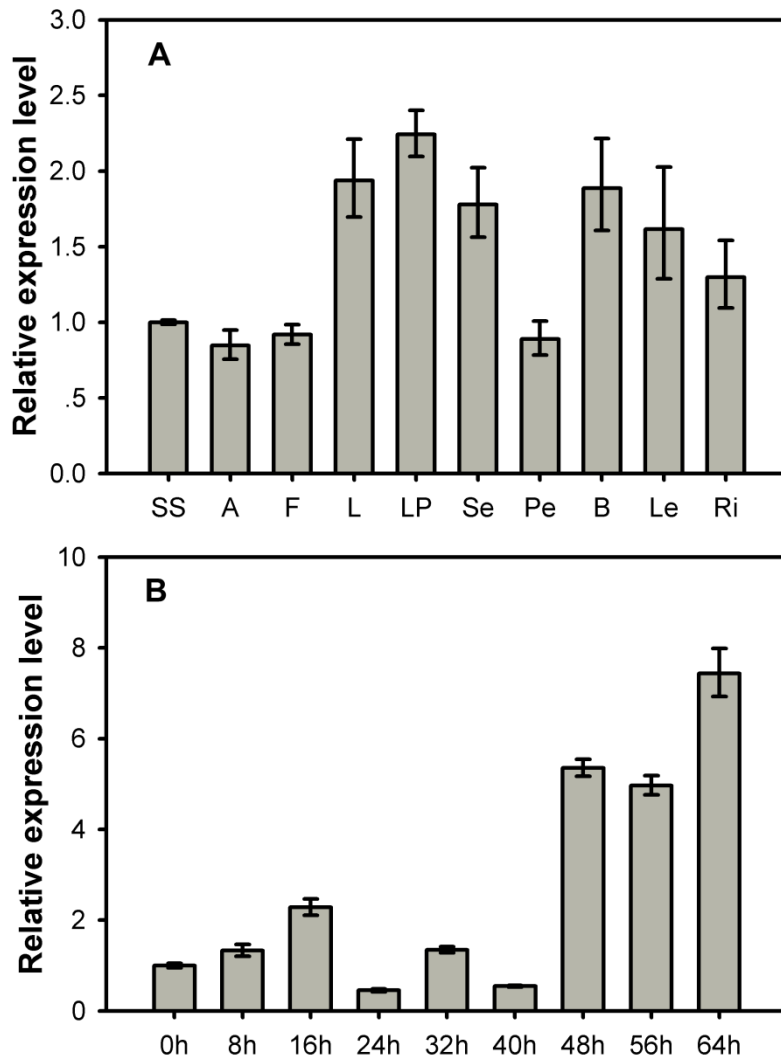
**Supplementary Figure 4.** Alignment of amino acid sequences of HcBSMTs with representative benzenoid carboxyl methyltransferases. The alignment was performed by ClustalX and shaded with GeneDoc. Amino acid residues shaded in black, gray and light gray represent 100, 80 and 60 % conserved identity, respectively. Dashes indicate gaps inserted for optimal alignment. Residues with “#” below indicate SAM binding sites. The amino acids with “&” below indicate the acid substrate binding sites, which are identified from the crystal structure of CbSAMT (Zubieta et al., 2003).



**Supplementary Figure 5.** Effect of temperature (A) and pH value (B) on HcBSMT1/2 activity with SAM and BA as substrates. The highest activity with a given reaction condition was set to 100%. All values shown in this figure are averages of three independent measurements. Optimum reaction temperature was determined at different temperature levels (10-40 °C). The optimum pH for HcBSMTs activity was assessed using two buffer systems. Reactions were carried out in 50 mM Bis-Tris propane buffer with pH ranging from 6.5 to 9.0 and in 50 mM 2-(N-morpholino)ethanesulfonic acid (MES) buffer with pH ranging from 5.0 to 6.5.



**Supplementary Figure 6.** Correlation analysis of methyl benzoate emission and related biosynthetic genes expression in different tissues (A) and during petal development (B). \*\* indicates a significant correlation at the level of  $P < 0.01$ . \*\*\* indicates a highly significant correlation at the level of  $P < 0.001$ .



**Supplementary Figure 7.** Expression analysis of *HcBALDI*. (A) *HcBALDI* gene expression quantity in different tissues. *HcACT* gene was used as an endogenous control. SS: styles and stigmas; A: anthers; F: filaments; L: labella; LP: lateral petals; Se: sepals; Pe: pedicels; B: bracts; Le: leaves; Ri: rhizomes. (B) *HcBALDI* gene expression quantity in petals at different floral developmental stages. *HcRPS* gene was used as an endogenous control. Error bars indicate the calculated maximum and minimum expression quantity of three replicates.

**Supplementary Table 1.** Analysis of putative genes related to benzenoid biosynthesis.

Gene	Gene ID	Description	Full-length (bp)	ORF (bp)	Gene expression quantity (RPKM)		
					D1	D4	D6
<i>HcPAL</i>	<b>comp46957_c0</b>	<b>Phenylalanine ammonialyase</b>	<b>2,449</b>	<b>2,169</b>	<b>222.79</b>	<b>1,166.32</b>	<b>325.79</b>
<i>HcCTS1</i>	comp39483_c0	COMATOSE	4,983	4,056	10.69	27.49	22.93
<i>HcCTS2</i>	comp43213_c0	COMATOSE	4,657	3,987	9.63	5.32	4.22
<i>HcCNL</i>	<b>comp18339_c0</b>	<b>Cinnamoyl-CoA ligase</b>	<b>2,290</b>	<b>1,752</b>	<b>0</b>	<b>479.54</b>	<b>364.13</b>
<i>HcCHD1</i>	<b>comp17550_c0</b>	<b>Cinnamoyl-CoA hydratase/dehydrogenase</b>	<b>2,783</b>	<b>2,175</b>	<b>315.10</b>	<b>668.73</b>	<b>756.00</b>
<i>HcCHD2</i>	comp25044_c1	Cinnamoyl-CoA hydratase/dehydrogenase	2,456	2,175	0.62	32.53	2.07
<i>HcCHD3</i>	comp43947_c0	Cinnamoyl-CoA hydratase/dehydrogenase	2,667	2,169	58.08	50.06	55.32
<i>HcKAT1</i>	<b>comp37768_c0</b>	<b>3-Ketoacyl CoA thiolase</b>	<b>1,831</b>	<b>1,377</b>	<b>236.35</b>	<b>619.74</b>	<b>1,082.13</b>
<i>HcKAT2</i>	comp39625_c0	3-Ketoacyl CoA thiolase	1,591	1,305	24.02	39.77	10.29
<i>HcBALD1</i>	comp17566_c0	Benzaldehyde dehydrogenase	1,927	1,629	86.96	54.40	78.17
<i>HcBALD2</i>	comp25915_c0	Benzaldehyde dehydrogenase	1,801	1,617	45.85	0.29	0
<i>HcTE1</i>	<b>comp46336_c0</b>	<b>Thioesterase</b>	<b>1,002</b>	<b>492</b>	<b>123.00</b>	<b>469.45</b>	<b>306.83</b>
<i>HcTE2</i>	comp41950_c0	Thioesterase	768	486	31.56	2.69	0.56
<i>HcBCMT1</i>	<b>comp37774_c0</b>	<b>Benzenoid carboxyl methyltransferase</b>	-	-	<b>0.65</b>	<b>7,896.98</b>	<b>7,382.24</b>
<i>HcBSMT1</i>	<b>comp37774_c0</b>	<b>Benzoic acid/salicylic acid methyltransferase</b>	<b>1,498</b>	<b>1,146</b>	-	-	-
<i>HcBSMT2</i>	<b>comp37774_c0</b>	<b>Benzoic acid/salicylic acid methyltransferase</b>	<b>1,666</b>	<b>1,131</b>	-	-	-
<i>HcBCMT2</i>	comp36217_c0	Benzenoid carboxyl methyltransferase	1,251	1,134	5.04	0.44	4.22
<i>HcBCMT3</i>	comp48707_c0	Benzenoid carboxyl methyltransferase	1,416	1,095	0.96	5.52	6.63

D1, D4 and D6 represent three flower developmental stages (squaring stage, blooming stage and senescence stage, respectively). RPKM: reads per kilobase of exon model per million mapped reads. Genes cloned in this study were in bold. Two different transcripts (named as *HcBSMT1* and *HcBSMT2*) were cloned under *HcBCMT1* unigene.



**Supplementary Table 2.** Primer sequences used in this study.

<b>Gene</b>	<b>Forward/reverse primer sequence (5'-3')</b>	<b>Purpose</b>
<i>HcPAL</i>	TCTGCTCATTCTTCGTCGTCC CATAGTTCGGATTTTCATTGGACAC	gene isolation
<i>HcCNL</i>	GAAAACCGAGCATGGTGGTCAA GACTCACGAATTATGCACGTGATG	gene isolation
<i>HcCHD1</i>	TGGAATTGGATTGGCGGAGATC GTTCCAAAGGACGAGCAAGATG	gene isolation
<i>HcKAT1</i>	CGTGAACGCTTCTTCTTGGTTC TTGCATTTCCGAGGATTGATGTAT	gene isolation
<i>HcTE</i>	TAACCTGCGTGCCGAAGATTGA CTAGATCGCTGACTGATTTCTTG	gene isolation
<i>HcBSMT1</i>	TTCTTGCCCTTCATTCATTGAG GAACAAAAGTGTTCTGAGCTTG	gene isolation
<i>HcBSMT2</i>	CCCGTAGTTCATGCTCCAT GTTTCAATTTACGTCCACATCAGC	gene isolation
<i>HcBSMT1</i>	<i>EcoRI</i> -GAATTCATGCTCCCTAACACGACAATC <i>NotI</i> -GCGGCCGCAAGCTTTCTTCTTCAAGGCGAAG	bacterial expression
<i>HcBSMT2</i>	<i>EcoRI</i> -GAATTCATGGGTTTGAAGGTGGAGCA <i>NotI</i> -GCGGCCGCATACTCTTTTCTTCAAGGCAATGAC	bacterial expression
<i>EGFP</i>	<i>SacI</i> -GAGCTCATGGTGAAGGGCGAGGAG <i>KpnI</i> -GGTACCTTACTTGTACAGCTCGTCCATG	transient expression
<i>HcBSMT1</i>	<i>SacI</i> -GAGCTCATGCTCCCTAACACGACAATC <i>KpnI</i> -GGTACCTTAAGCTTTCTTCTTCAAGGCG	transient expression
<i>HcBSMT2</i>	<i>SacI</i> -GAGCTCATGGGTTTGAAGGTGGAGCA <i>KpnI</i> -GGTACCTTATACTCTTTTCTTCAAGGCAATG	transient expression
<i>HcPAL</i>	CTCATGTTCCGCCAATTCTC TTATGCTGCTCCGCACTCT	real-time PCR
<i>HcCNL</i>	CTCACTGCCGCAGCAACATG CTCAGCAGGAACTTCTGGATC	real-time PCR
<i>HcCHD1</i>	AACTTATCAGAATCCCAGCAGTC CATCCTTGATGGCGATGAACAG	real-time PCR
<i>HcKAT1</i>	CCAGTCCCACAACCTTCTTATCC GCCGTAGAGTTTGTTCATGTG	real-time PCR
<i>HcTE</i>	CGCACAAGGATTACGAAGACAC GCATCAACGACGAGCAAGGTT	real-time PCR
<i>HcBSMT1</i>	TATTTGCGGATATGCACATCTTG CATGTAGATATGCATACTAATAATCACC	real-time PCR
<i>HcBSMT2</i>	AGGAGAAAGCCAATCACACC GGCATGATTCAGTTTGACAAG	real-time PCR
<i>HcACT</i>	GTATGTTGCTATTCAGGCTGTCC GAAGAATGGCATGAGGTAGAGC	real-time PCR
<i>HcRPS</i>	TTAGTAGCATCGGCTGCAATAAG CTCTTTTGGGAAGACGGTTGAG	real-time PCR