

## Supplementary Materials:

**Table S1.** The list of primers used in this study.

Gene name	Species (GenBank)	Primer sequences (5'→3')	Usage
<i>LaOMT1</i>	<i>L. aurea</i>	P1: 5'-ATGATGGGTGCTAGCCAAGATGATTATG-3' P2: 5'-TTATCAATAAAGACGTCGACAAATAGTCA-3'	<i>LaOMT1</i> ORF amplification
<i>LaOMT1</i>	<i>L. aurea</i>	P3: 5'-CAGCCATATGATGGGTGCTAGCCAAGATGATTATG-3' P4: 5'-CGCACACTCGAGTCATAAAAGACGTCGACAAATAGTCAGTC-3'	<i>LaOMT1</i> protein expressions in <i>E. coli</i>
<i>LaOMT1</i>	<i>L. aurea</i>	P5: CAGCTCTAGAATGGGTGCTAGCCAAGATGATTATG-3' P6: CGCAGGATCCATAAAGACGTCGACAAATAGTCAGTC-3'	Subcellular localization
<i>LaOMT1</i>	<i>L. aurea</i>	P7: 5'-CAACACGCTCTGGTTCGGA-3' P8: 5'-CGACTGAGACTTGGATATC-3'	qRT-PCR analysis
<i>LaTIP41</i>	<i>L. aurea</i>	P9: 5'-GCAACCATCCAAAGTTAACTGCT-3' P10: 5'-AATGTGCAAGCAGGGCTAGTAA-3'	Reference gene for qRT-PCR analysis
<i>LaEXP1</i>	<i>L. aurea</i>	P11: 5'-ATTGAAACAAACCTACACCGCAA-3' P12: 5'-GCTGTAAGAATGCTAACCGTTCA-3'	Reference gene for qRT-PCR analysis
<i>AtPEX7</i>	Arabidopsis (At1g29260)	P13: 5'-CGCTTCTAGAATGCCGGTGTCAAAGCTCC-3' P14: 5'-AATCCCCGGGACTGGCTCTAGGATCCATCCC-3'	Subcellular localization for marker protein
<i>AtGAPC1</i>	Arabidopsis (At3g04120)	P15: 5'-CGCTTCTAGAATGGCTGACAAGAAGATTAG-3' P16: 5'-AATCCCCGGGGCCTTGACATGTGGACGAT-3'	Subcellular localization for marker protein
<i>AtAra6</i>	Arabidopsis (At3g54840)	P17: 5'-CGCTTCTAGAATGGGATGTGCTTCTCTTT-3' P18: 5'-AATCCCCGGGTGACGAAGGAGCAGGACGAGGT-3'	Subcellular localization for marker protein

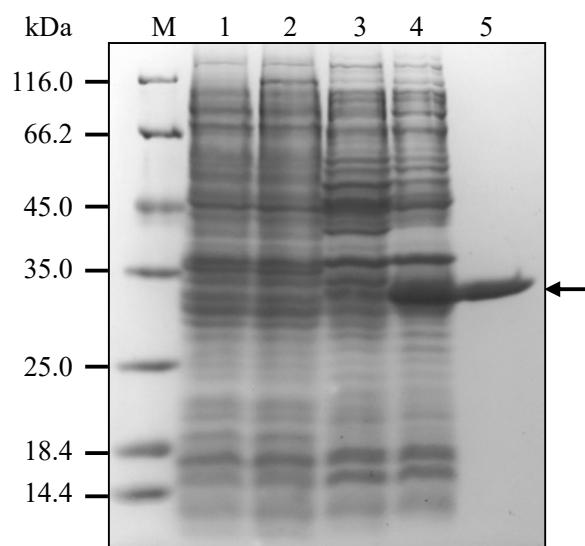
**Table S2.** GenBank accession numbers of OMT proteins in Fig. 1

Protein names	Species	Accession number	Class type
LaOMT	<i>Lycoris aurea</i>		I
AtCOMT	<i>Arabidopsis thaliana</i>	NP_200227.1 (At5g54160)	II
AtCCoAOMT7	<i>Arabidopsis thaliana</i>	NP_567739.1 (At4g26220)	I
Cr16OMT	<i>Catharanthus roseus</i>	ABR20103.1	II
CrF4OMT	<i>Catharanthus roseus</i>	AAR02420.1	II
CiOMT2	<i>Carapichea ipecacuanha</i>	BAI79244.1	II
CaCOMT	<i>Chrysosplenium americanum</i>	AAA86982.1	II
Cj4OMT	<i>Coptis japonica</i>	BAB08005.1	II
Cj6OMT	<i>Coptis japonica</i>	BAB08004.1	II
CjCoOMT	<i>Coptis japonica</i>	BAC22084.1	II
HsOMT	<i>Homo sapiens</i>	NP_000745.1	I
HvF7OMT	<i>Hordeum vulgare subsp. vulgare</i>	CAA54616.1	II
Ms7OMT	<i>Medicago sativa</i>	AAC49927.1	II
MsCCoAOMT	<i>Medicago sativa</i>	AAC28973.1	I
MxSafC	<i>Myxococcus xanthus</i>	AAC44130.1	I
NpN4OMT	<i>Narcissus sp. aff. Pseudonarcissus</i>	AIL54541.1	I
NtCOMT	<i>Nicotiana tabacum</i>	CAA50561.1	II
ObEOMT1	<i>Ocimum basilicum</i>	AAL30424.1	II
OsCOMT	<i>Oryza sativa</i>	Os08g0157500	II
Ps7OMT	<i>Papaver somniferum</i>	ACN88562.1	II
Ps4OMT1	<i>Papaver somniferum</i>	AAP45313.1	II
Ps4OMT2	<i>Papaver somniferum</i>	AAP45314.1	II
PtCCoAOMT	<i>Populus tremuloides</i>	AAA80651.1	I
PdOMT	<i>Prunus dulcis</i>	CAA11131.1	II
PfOMT	<i>Mesembryanthemum crystallinum</i>	AY14552.1	II
PpOMT	<i>Pyrus pyrifolia</i>	BAA86059.1	II
RnOMT	<i>Rattus norvegicus</i>	NP_036663.1	I
SlAnthOMT	<i>Solanum lycopersicum</i>	NP_001289828.1	I
SbCCoAOMT	<i>Sorghum bicolor</i>	XP_002436550	I
SynOMT	<i>Synechocystis sp. Strain PCC 6803</i>	WP_010873795.1	I
TtCOMT	<i>Thalictrum tuberosum</i>	AAD29842.1	II
Tf4OMT	<i>Thalictrum flavum subsp. glaucum</i>	AAU20768.1	II
VpCOMT	<i>Vanilla planifolia</i>	AAS64572.1	II
VpOMT5	<i>Vanilla planifolia</i>	ADZ76154.1	I
VpOMT4	<i>Vanilla planifolia</i>	ADZ76153.1	I
VvCCoAOMT	<i>Vitis vinifera</i>	CAA90969.1	I
VvFAOMT	<i>Vitis vinifera</i>	C7AE94.1	I
ZmCOMT	<i>Zea mays</i>	AAB03364.1	II
ZvCOMT	<i>Zinnia violacea</i>	AAA86718.1	II
ZvCCoAOMT	<i>Zinnia violacea</i>	AAA59389.1	I

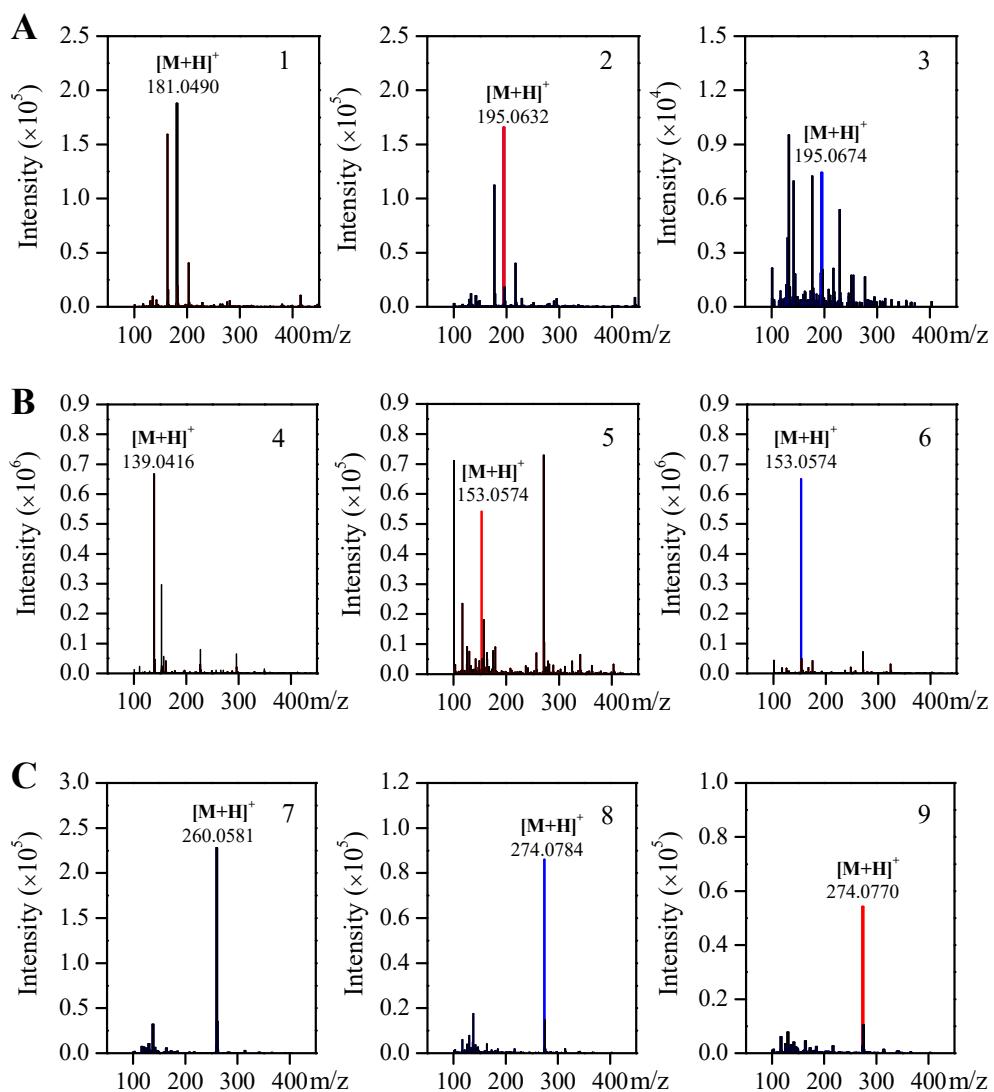
**Figure S1.** Nucleotide and predicted protein sequence of LaOMT1.

1 ATGGGTGCTAGCCAAGATGATTATGCACATAATCCACAAGAATATTTGCATAGTGAAGAT  
1 M G A S Q D D Y A L I H K N I L H S E D  
61 CTTCTTAAGTACATATTGGAGACTAGTGTTCACCCAAGAGAGCATGAACAGCTCAAGGGG  
21 L L K Y I L E T S V Y P R E H E Q L K G  
121 TTGAGGGAGGTGACTGAGAACATGAATGGAGTACGGCGCTTGTGCAGCCGATGAAGGA  
41 L R E V T E K H E W S T A L V A A D E G  
181 TTATTTCTTCTATGTTAAAGCTCATGAATGCCAAGAGAACATTGAGATTGGTGTAA  
61 L F L S M L L K L M N A K R T I E I G V  
241 TACACCGGTTATTCTCTGCTCACAAACCGCTTGGCTTACCAAGATGGAAAGATAACG  
81 Y T G Y S L L T T A L A L P E D G K I T  
301 GCAATTGACGTCAACAAGTCCTACTTGAGATAGGACTGCCATTATTCAAGAAAGCAGGA  
101 A I D V N K S Y F E I G L P F I Q K A G  
361 GTTGAGCATAAGATCAATTTCATTGAATCAGAACGCACTTCCTGTTGATCATATGCTT  
121 V E H K I N F I E S E A L P V L D H M L  
421 CAAGAGATGAAGGAAGAACCTCTACGACTTGCAATTGTCATGCAGACAAACCAAAC  
141 Q E M K E E D L Y D F A F V D A D K P N  
481 TATGCTAATTACACGAGCGATTAGTGAAGCTTGTCAAGGGTTGGAGGAGCAATCGTCTAC  
161 Y A N Y H E R L V K L V R V G G A I V Y  
541 GACAACACGCTCTGGTCCGAACTGTAGCATTCCAGAATATCCAGGCCTCATCCAGAA  
181 D N T L W F G T V A F P E Y P G L H P E  
601 GAGGAGGAGTGTAGGGTCTTTCAAGAACCTGAATAAGCTCTGGCAGCTGATCCCCGT  
201 E E E C R V S F R N L N K L L A A D P R  
661 GTCGAGATATCCAAGTCTCAGTCGGCAGGGACTGACTATTGTCGACGTCTTATTGA  
221 V E I S Q V S V G D G L T I C R R L Y \*

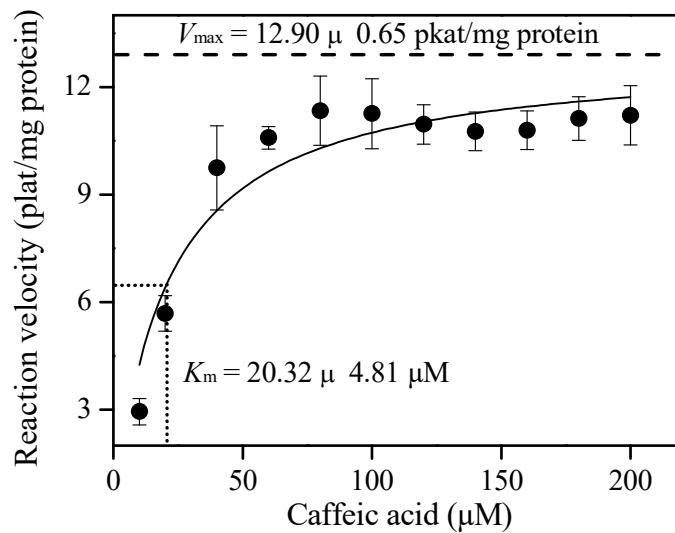
**Figure S2.** Expression and purification of N-terminal 6 × histidine-tagged LaOMT1 protein. *E. coli* BL21 (DE3) harbouring pET-28a or pET28a-LaOMT1 was incubated with or without 1 mM IPTG (isopropyl- $\beta$ -d-thiogalactopyranoside) at 30 °C for 16 h. Protein samples were separated on a 10% SDS-PAGE followed by Coomassie blue staining. M, molecular mass standards; lane 1, total proteins in 20- $\mu$ l aliquots from *E. coli* BL21 culture harboring pET28a; lane 2, total proteins in 20- $\mu$ l aliquots from *E. coli* BL21 culture harboring pET28a induced by IPTG; lane 3, total proteins in 20- $\mu$ l aliquots from *E. coli* BL21 culture harboring pET28a-LaOMT1; lane 4, total proteins in 20- $\mu$ l aliquots from *E. coli* BL21 culture harboring pET28a-LaOMT1 induced by IPTG; lane 5, 10  $\mu$ g recombinant LaCOMT1 protein purified by affinity chromatography. Arrow indicates the position of the fusion protein of LaOMT1.



**Figure S3.** Mass spectrometry (MS) of new compounds produced by LaOMT1 enzyme in catalyzing the substrate caffeic acid (A), 3,4-dihydroxybenzaldehyde (B), and noberlladine (C). 1, substrate caffeic acid; 2, ferulic acid produced by LaOMT1; 3, isoferulic acid produced by LaOMT1; 4, substrate 3,4-dihydroxybenzaldehyde; 5, isovanillin produced by LaOMT1; 6, vanillin produced by LaOMT1; 7, substrate norbelladine; 8, 4'-*O*-methylnorbelladine; 9, 3'-*O*-methylnorbelladine.



**Figure S4.** Measurement of the  $K_m$  and  $V_{max}$  of LaOMT1 for substrate caffeic acid.



**Figure S5.** Measurement of the  $K_m$  and  $V_{max}$  of LaOMT1 for substrate 3,4-dihydroxybenzaldehyde.

