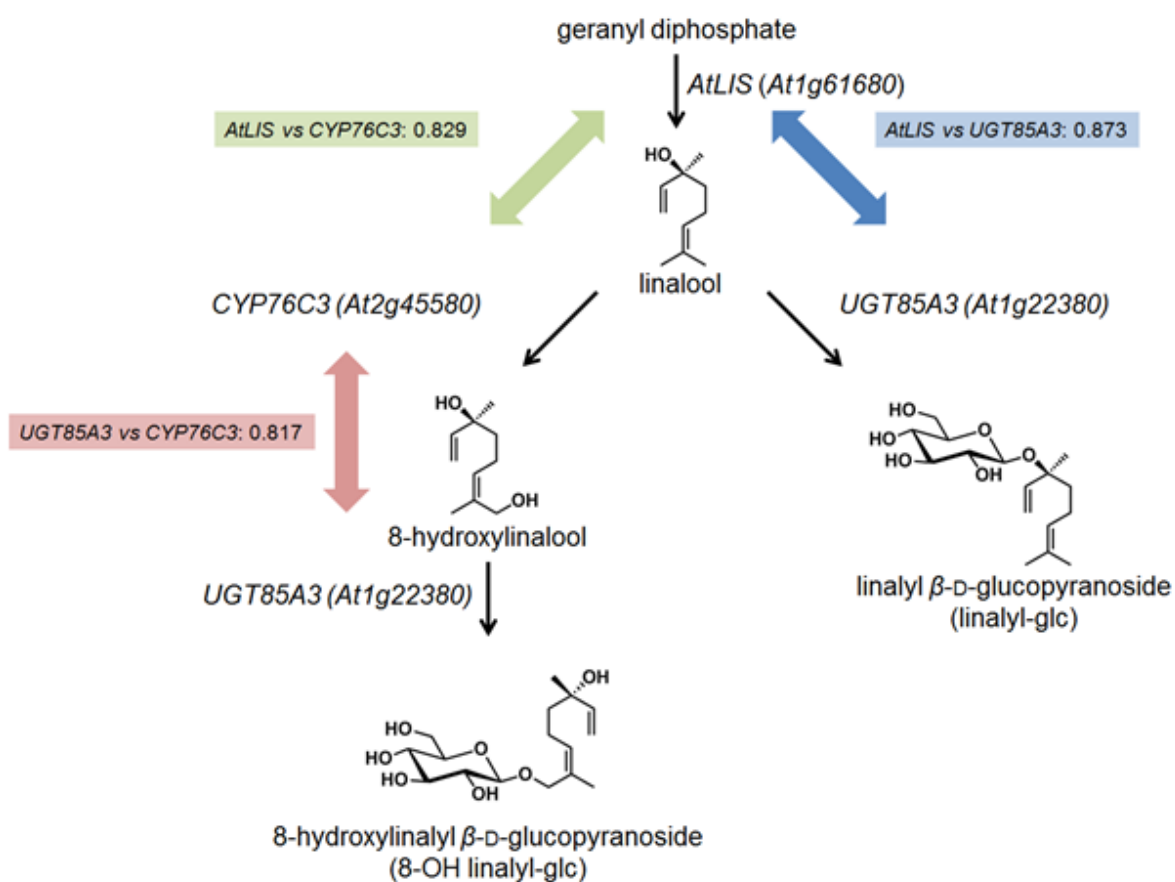
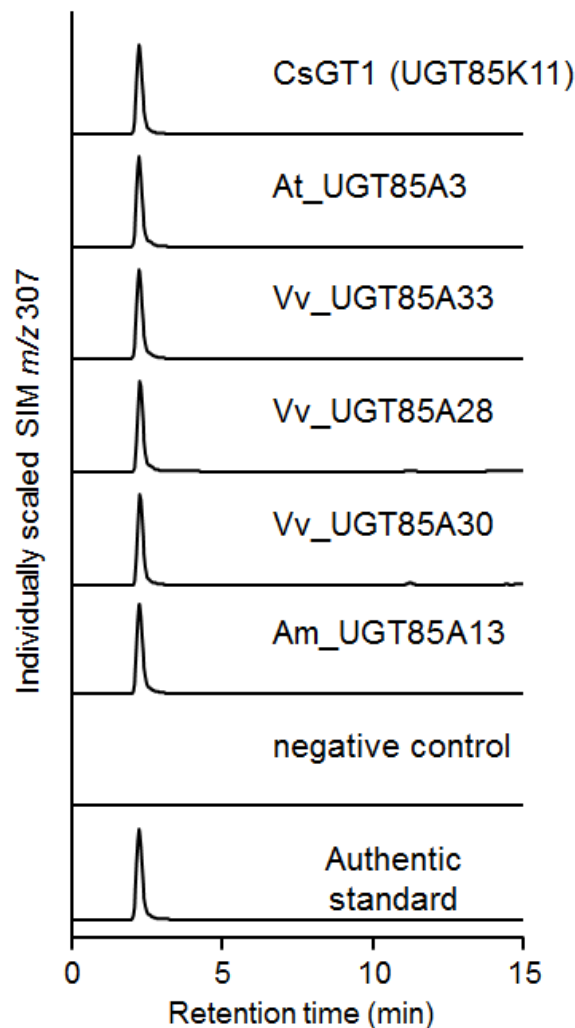
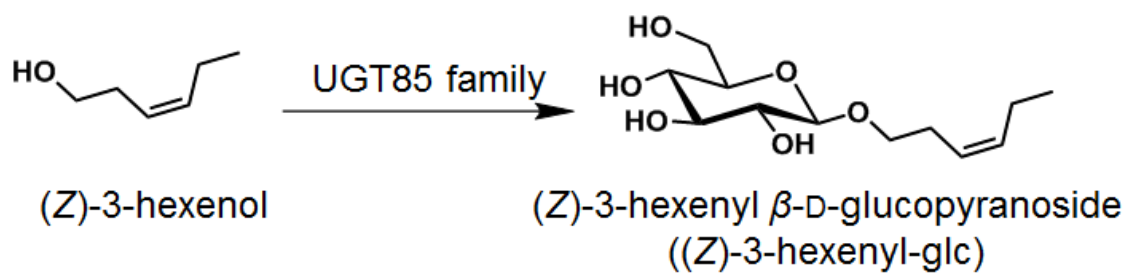


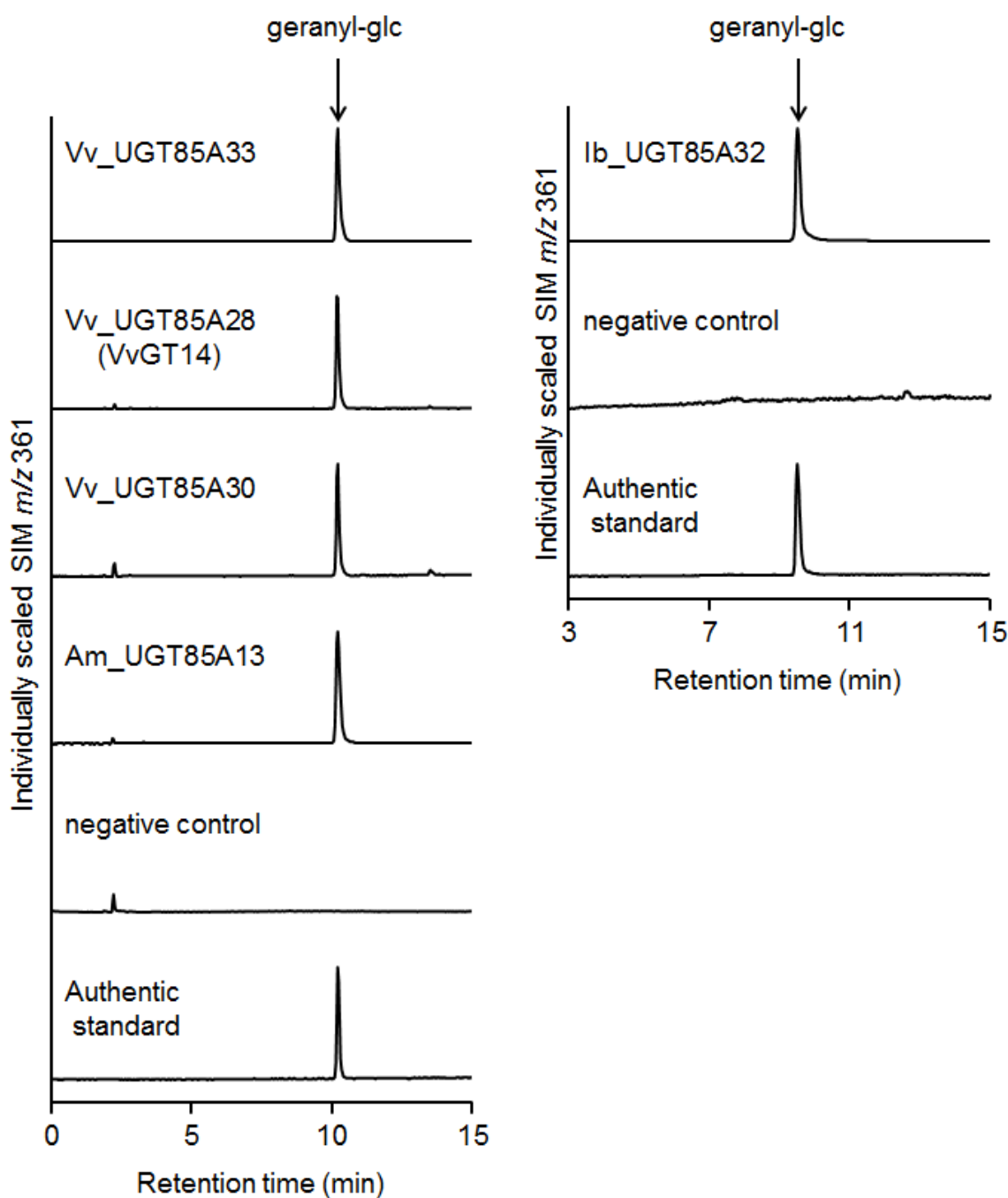
Supplemental Figures



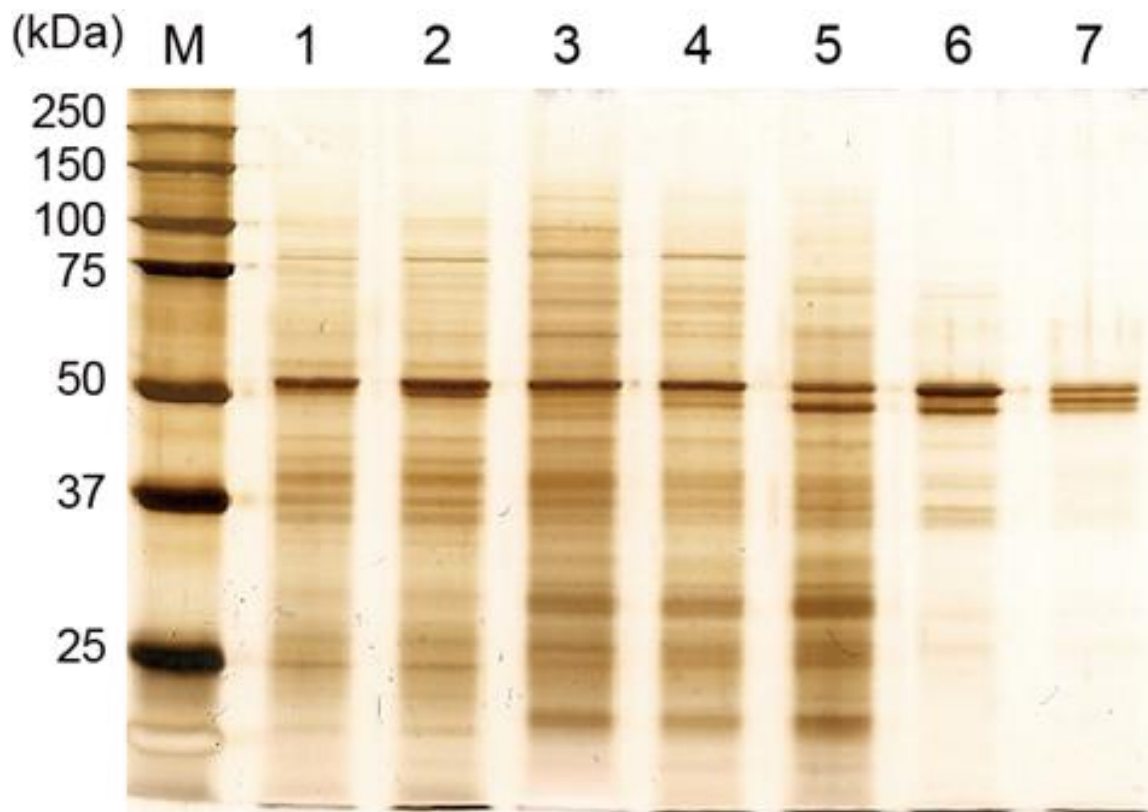
Supplemental Figure S1. Co-expression analysis of *AtLIS*, *CYP76Cs* and *UGT85A3* by ATTDII (Gene coexpression database, <http://atted.jp/>) with Ver. C4.1.



Supplemental Figure S2. Enzymatic activity of CsGT1 homologs for (Z)-3-hexenol. *In vitro* characterization of UGT85s toward (Z)-3-hexenol with tea plant (CsGT1; UGT85K11), Arabidopsis (At_UGT85A3), grapevine (*Vitis vinifera*: Vv_UGT85A33, Vv_UGT85A28, Vv_UGT85A30) and snapdragon (*Antirrhinum majus*: Am_UGT85A13). LC-MS analysis of the enzymatic product of CsGT1 (UGT85K11), At_UGT85A1, At_UGT85A3, Vv_UGT85A33, Vv_UGT85A28, Vv_UGT85A30 and Am_UGT85A13 compared to the authentic standard ((Z)-3-hexenol).



Supplemental Figure S3. Enzymatic activity of CsGT1 homologs for geraniol. *In vitro* characterization of UGT85As toward geraniol with grapevine (*Vitis vinifera*: Vv_UGT85A33, Vv_UGT85A28, Vv_UGT85A30), sweet potato (*Ipomoea batatas*: Ib_UGT85A32), snapdragon (*Antirrhinum majus*: Am_UGT85A13). LC-MS analysis of the enzymatic product of Vv_UGT85A33, Vv_UGT85A28, Vv_UGT85A30 and Am_UGT85A13 and Ib_UGT85A32 compared to the authentic standard (geranyl-glu).



Supplemental Figure S4. Purified enzymes catalyzing the second xylosyltransferase (CsGT2). SDS-PAGE analysis of each active fraction obtained from purification steps. Lane 1; crude enzyme, lane 2; ammonium sulfate, lane 3; HiTrap DEAE FF, lane 4; HiTrap Q FF, lane 5; Macro-prep Ceramic Hydroxyapatite Type III, lane 6; HiTrap Blue HP, lane 7; Mono Q 5/50 GL. SDS-PAGE was stained by a silver staining method (Sil-best stain one, Nacalai, Kyoto, Japan)

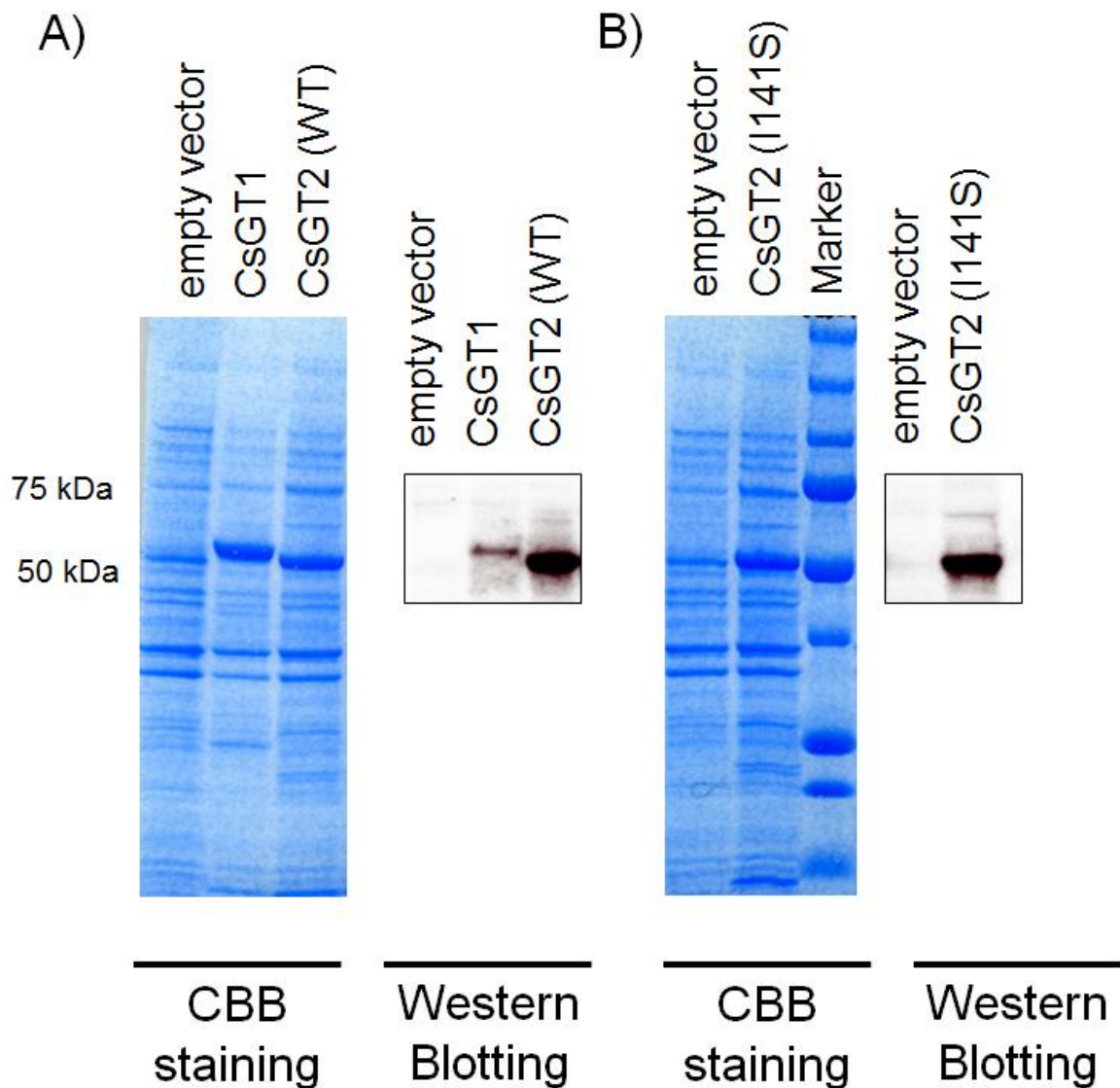
A)

Sequence	Charge	Observed <i>m/z</i>	<i>Mr</i> (Calc)
FPEVEKVELEEEALPK	+3	586.3224	1755.9294
GLVVEGWAPQAR	+2	641.852	1281.6829
EEIEEIAHGLELSMVNFIWVRFPEVEK	+4	840.4036	3357.6948

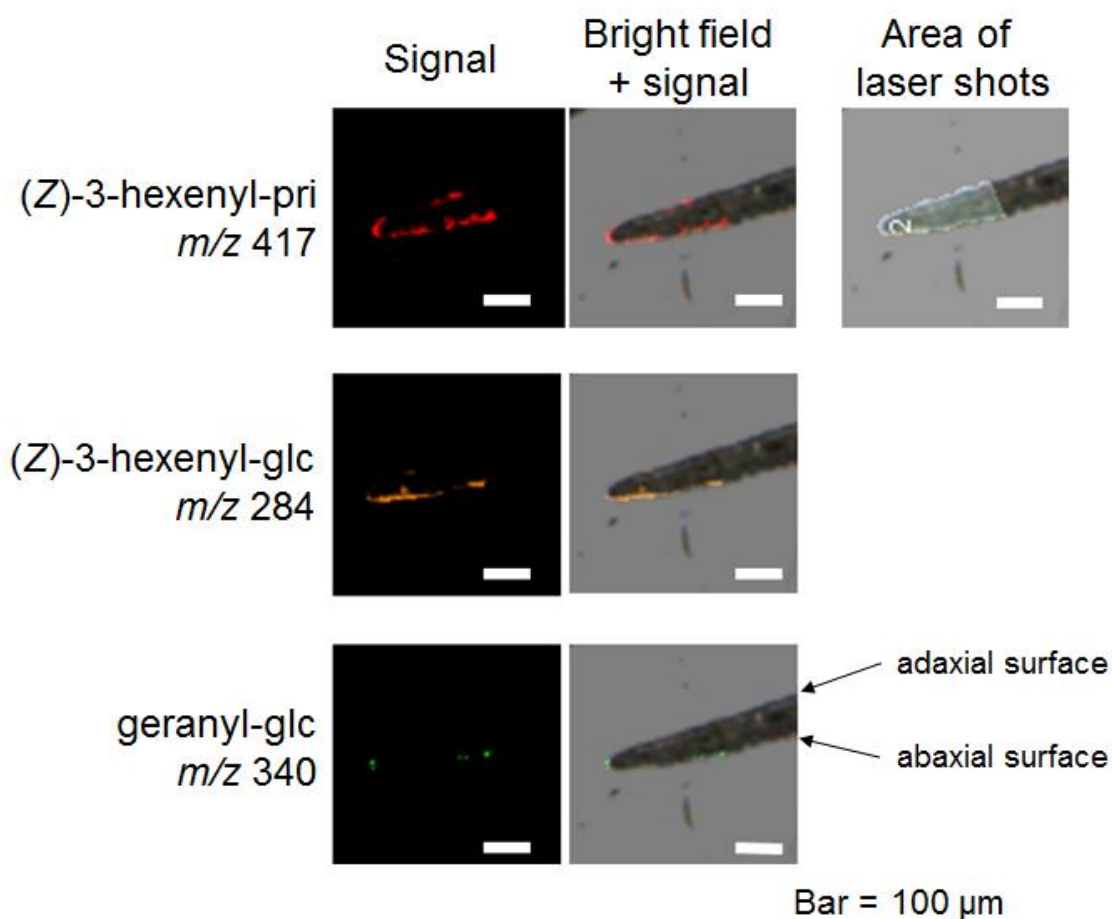
B)

1 ENEHSEIIQW LDNKGEYSTL FVSEFGSEYFM SK**EEIEEIAH** **GLELSMVNFI**
 51 **WVRFPEVEK** **VELEEALPKG** FIDRVGERGL **VVEGWAPQAR** ILTHSSTGGF
 101 VSHCEWNSVL ESLKFGVPMV AIPMQYEQPL NAKLVEEVGV AAEVNRDING
 151 RLNREEIAQV IRKVVVEKSG EDIRIKARIF GDKIRMGDE EIDEAVEVLL
 201 QLCKDVKLLK NSNFIEIFKW VEKVFYFYFI LFYWVVWYTN SLIRLYTSRR
 251 STNIVFDHVV LVLDLVALFF IYIYRFTIIG VIVSSFCSID TGGILNMV

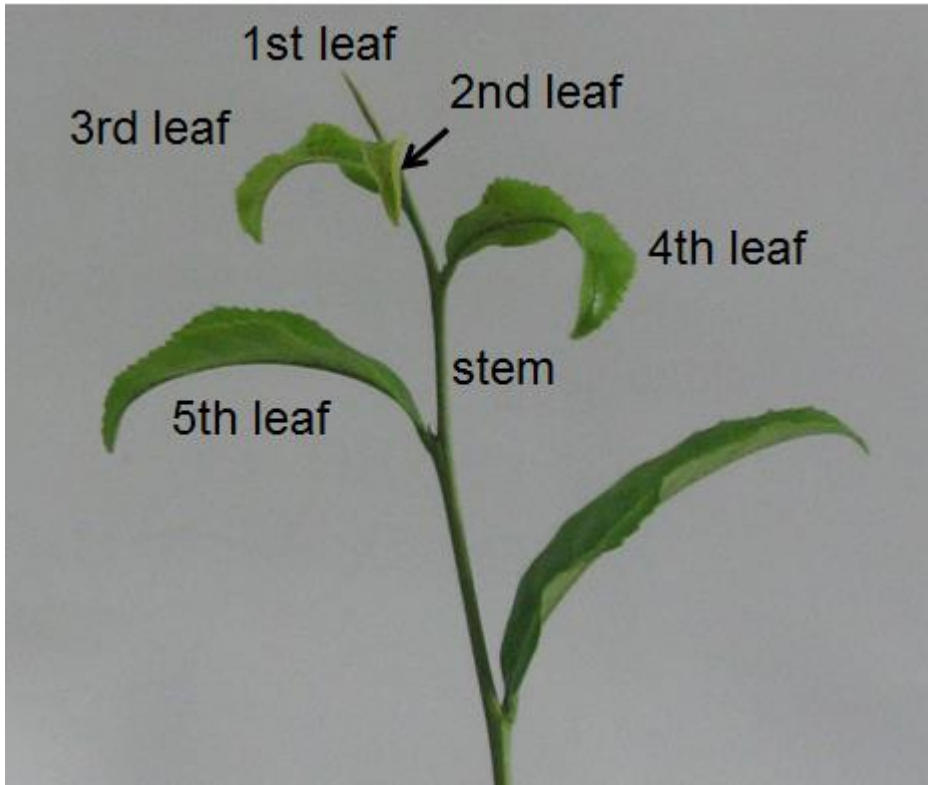
Supplemental Figure S5. Partial peptide sequence of the CsGT2. LC-MS/MS analysis of the partial peptide sequences of CsGT2 (UGT94P1) that purified from tea leaves. A) Three peptide sequences were exactly identified with tea EST database contig 13. B) Predicted amino acid sequence of tea EST database contig 134. Red letters show identical peptides with tea EST database and LC-MS/MS analytical results.



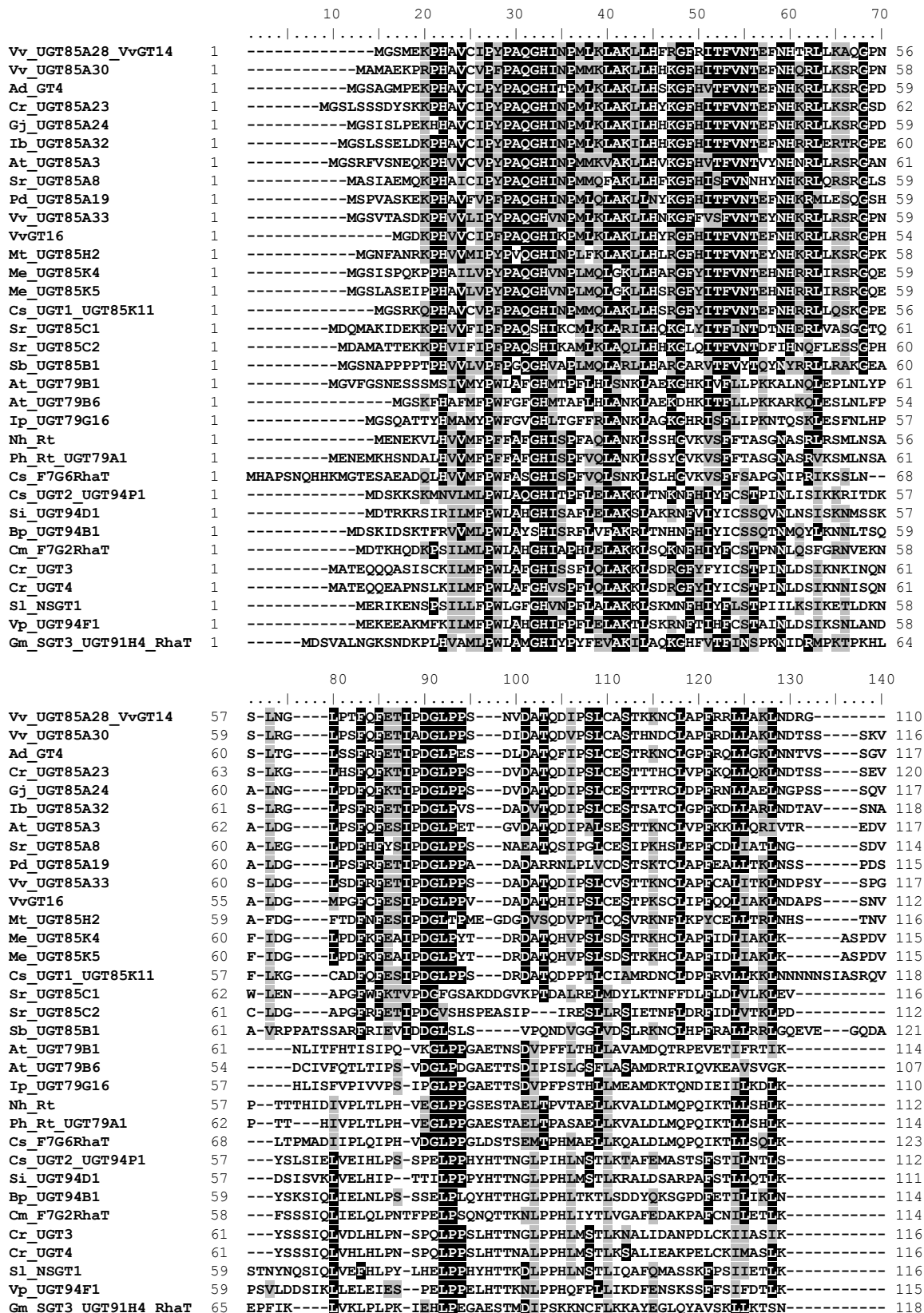
Supplemental Figure S6. Recombinant proteins of a series of CsGTs. Specific bands responsible of CsGT1 and CsGT2 (A), and CsGT2-I141S proteins (B) were detected around the size marker of 50kDa in SDS-PAGE, are roughly consistent with the estimated molecular size of CsGT1 (55.7 kDa) and CsGT2 (51.1 kDa), respectively. These specific bands were immunologically confirmed to be the recombinant CsGTs by Western blotting analysis.



Supplemental Figure S7. Imaging MS of young fresh leaves of *C.sinensis*. Specific signal (red; *m/z* 417) corresponding to (Z)-3-hexenyl-pri. Specific signal (orange; *m/z* 284) corresponding to (Z)-3-hexenyl-glc. Specific signal (green; *m/z* 340) corresponding to geranyl-glc.



Supplemental Figure S8. Harvesting individual tissues for quantification of endogenous aroma glycosides and quantitative real-time PCR (qRT-PCR) of CsGTs and β -PD. The young and mature developmental stages of leaves and stems were defined as follows: young leaves, which are the first, second, and third leaves (plucking part for high grade tea products); mature leaves, which are the fourth and fifth leaves; stem, which are green young non-lignified part between the 3rd leaf and 5th leaf; branch, which are brown lignified old part.



Supplemental Figure S9. Multiple sequence alignment of protein sequences of UGT-glucosyltransferase OG2 and OG8 family. The alignment was performed using ClustalW2.1.

	290	300	310	320	330	340	350																																															
Vv UGT85A28_VvGT14	245	P-IYTI	GPIQLLPDQI	THD----	SELKLGISN	LWKKEE	PECLK	WLD	SKEE	NSV	VVYV	NFGS	-ITV	MT	PQ	QI	307																																					
Vv UGT85A30	251	P-IYTI	GPIQLLNQMP	D----	NDLKSIE	SNLWKE	EPGCL	EWLDA	KEE	ESV	VVYV	NFGS	-VT	MT	PQ	QI	313																																					
Ad GT4	252	P-IYTI	GPIQLLNQI	KE----	ESLKMIG	SNLWKE	EPMC	LEWLN	SKEE	PKS	VVYV	NFGS	-IT	MT	PQ	QI	314																																					
Cr UGT85A23	255	P-IYTI	GPIQLLNQVDD	----	ESLKVLG	SNLWKE	EEPE	CLWLD	TKD	ENS	VVYV	NFGS	-IT	MT	ND	QI	317																																					
Gj UGT85A24	252	P-IYTI	GPIQLQKEVKD	----	ERLSVLG	SNLWKE	EEPE	CLD	WLD	SKD	ENS	VVYV	NFGS	-IT	MT	PQ	QI	314																																				
Ib UGT85A32	253	P-VYAV	GPIQLQTVKD	----	SNVRALAS	NLWKE	DTSC	LEWLD	TKD	ENS	VVYV	NFGS	-IT	MT	PD	QI	315																																					
At UGT85A3	252	P-VVPI	GPIHLLVNR	EIEE--	DSEIGRM	GSNLWKE	EETE	CLQ	WLN	TKSR	NS	VVYV	NFGS	-IT	MT	TA	QI	316																																				
Sr UGT85A8	249	Q-IYTI	GPIHMMQ-Q	YVDH--	DERLKH	IGSNLWKE	EDV	SCIN	WLD	TKD	ENS	VVYV	NFGS	-IT	MT	KE	QI	312																																				
Pd UGT85A19	250	P-IYSI	GPIQLPYSE	IPSE--	YNDLKA	IGSNLW	AE	TECL	NWLD	TKD	ENS	VVYV	NFGS	-IT	MT	NE	QI	314																																				
Vv UGT85A33	253	P-VYSI	GPIQHLVDQ	ISD----	DRLKSMG	SNLWKE	QTD	CLQ	WLD	SKEE	NS	VVYV	NFGS	-IT	MT	SQ	QI	315																																				
VvGT16	248	T-ICTV	GPIPLLNQI	PPD----	NSIESNL	WREETE	CLQ	WLN	SKD	ENS	VVYV	NFGS	-IT	MT	PE	QI	308																																					
Mt UGT85H2	251	S-IYPI	GPIPSLLKQ	TPQ----	IHQLD	SLSN	LWKE	DETE	CLD	WLE	SKEE	NS	VVYV	NFGS	-IT	MT	PE	QI	314																																			
Me UGT85K4	250	KNIY	TVGPFIL	LEKGI	PEI----	KSKAFRS	SLWKE	DLSC	LEWLD	KRE	ES	VVYV	NFGS	-VT	IT	NE	QI	313																																				
Me UGT85K5	250	KNIY	TVGPFIL	LEKGI	PEI----	KSKAFRS	SLWKE	DLSC	LEWLD	KRE	ES	VVYV	NFGS	-VT	IT	NE	QI	313																																				
Cs UGT1 UGT85K11	253	N-IYTI	GPIQLLNQV	VIDG----	EFKLNS	LWKE	DTK	CLQ	WLD	TKD	ENS	VVYV	NFGS	-IT	MT	DQ	QI	315																																				
Sr UGT85C1	252	N-VYTI	GPIQLLNK	ITQ	KETNN--	DSYSLN	WKEE	PE	VE	WLN	SKEE	NS	VVYV	NFGS	-L	AV	SL	QI	314																																			
Sr UGT85C2	246	H-IYTI	GPIQLLDQ	IP	EEKKQ	GITS	LWKE	ET	V	KEE	PE	CF	WLN	OS	SKEE	NS	VVYV	NFGS	-T	IT	NE	QI	313																															
Sb UGT85B1	257	P-IYTV	GPIAEV	IAS	SDSAS--	AGLAAM	D	IS	Q	ED	TR	CL	S	WLD	CR	P	A	S	VVYV	NFGS	-M	AV	MT	A	Q	321																												
At UGT79B1	242	KPVY	LTG----	PVLP	GSQP----	NQPSLD	P	Q	W	A	E	W	A	K	F	N	H	G	S	VV	F	C	A	F	G	S	-Q	P	V	N	K	I	D	Q	295																			
At UGT79B6	225	RKVL	LTG----	PML	PEPDN----	SKP-LE	D	Q	W	R	Q	W	L	S	K	F	D	E	G	S	V	I	C	A	F	G	S	-Q	I	L	L	E	K	D	Q	276																		
Ip UGT79G16	236	KPVLL	AG----	PAL	PVP----	SKSTME	Q	K	W	S	D	W	L	G	K	F	K	E	G	S	V	I	C	A	F	G	S	-E	C	T	L	R	K	D	K	F	Q	286																
Nh Rt	240	KPVLL	AG----	PV	-PDPP----	SGK-LE	E	K	W	D	A	W	L	N	K	F	E	A	G	T	V	I	C	S	F	G	S	-E	E	T	L	K	D	D	Q	I	K	290																
Ph Rt UGT79A1	242	KPV	FSNR----	TR	S-GPA----	SGK-LE	E	K	W	A	T	W	L	N	K	F	E	G	T	V	I	C	S	F	G	S	-E	E	T	L	T	D	D	Q	292																			
Cs F7G6RhaT	248	KPVLL	AG----	PL	VNPEPP----	SGE-LE	E	R	W	A	N	W	L	G	K	F	E	G	S	V	I	C	S	F	G	S	-E	E	T	L	T	D	D	Q	299																			
Cs UGT2 UGT94P1	227	KRVV	PVG----	PL	VQIDD----	DE-NE	H	S	E	I	I	Q	W	L	N	K	G	E	S	T	L	F	V	S	F	G	S	-E	Y	F	M	S	K	E	E	I	E	278																
Si UGT94D1	225	KRFV	PVG----	PL	VQVGC----	DMENE	G	N	D	I	E	W	L	D	G	R	R	S	A	V	F	S	S	F	G	S	-E	Y	F	L	S	N	E	E	I	E	277																	
Bp UGT94B1	223	KRV	PVG----	PL	VQEASL----	LQ-DD	H	I	W	I	M	K	W	L	D	K	K	E	S	S	V	V	F	V	F	G	S	-E	Y	L	S	N	E	I	E	274																		
Cm F7G2RhaT	228	NEI	PVG----	PL	IQEP----	TFKVD	D	T	K	I	M	D	W	L	S	O	K	E	E	R	S	V	V	A	S	F	G	S	-E	Y	F	P	S	T	D	E	I	H	278															
Cr UGT3	231	SKV	PV	CVIS	-LNN	NDQG----	QGNK	D	E	E	I	I	Q	W	L	K	K	S	H	R	S	V	V	F	S	F	G	S	-E	Y	F	L	N	Q	E	E	I	E	286															
Cr UGT4	235	RKM	PV	CLANS	PD	NNNHQE----	QSNK	D	G	D	E	L	I	Q	W	L	E	T	K	S	E	R	S	V	V	F	S	F	G	S	-E	Y	F	L	N	Q	E	E	I	E	291													
Sl NSGT1	233	KELI	P	GPI	I	REAM	IDEEE----	DWGT	I	Q	S	W	L	D	K	K	D	Q	L	S	C	V	V	V	S	F	G	S	-E	S	F	L	S	K	Q	E	I	E	285															
Vp UGT94F1	225	KQI	P	T	G	-----	LLIAN	-----	SDE	K	D	E	P	E	I	M	Q	W	L	D	E	K	S	E	R	S	T	V	V	I	S	F	G	S	-E	C	F	L	S	K	Q	E	I	E	274									
Gm SGT3 UGT91H4_RhaT	243	VV	VV	PV	G	L	P	P	S	M	Q	I	R	D	V	E	-----	EED	N	N	P	D	W	V	R	I	K	D	W	L	D	T	E	S	S	V	V	V	I	C	F	G	S	-E	L	K	L	S	Q	E	D	I	T	301

	360	370	380	390	400	410	420																																																				
Vv UGT85A28_VvGT14	308	EFAW	GLANS	NQ	S	F	L	W	I	R	P	D	L	V	S	G	----	ES	A	I	L	P	P	E	V	A	E	T	E	D	----	R	G	L	L	A	G	W	C	P	O	E	V	L	T	H	O	A	I	G	G	F	L	369					
Vv UGT85A30	314	EFAW	GLANS	NK	F	L	W	I	R	P	D	L	V	A	G	----	DA	A	I	L	P	A	D	E	V	A	Q	T	K	E	----	R	S	L	L	A	S	W	C	P	O	E	V	L	T	H	P	A	I	G	G	F	L	375					
Ad GT4	315	EFAW	GLANS	NQ	S	F	L	W	I	R	P	D	L	V	G	----	ES	A	V	L	P	P	E	V	A	V	T	K	E	----	R	G	M	L	A	S	W	A	P	O	E	V	L	T	H	S	S	V	G	G	F	L	376						
Cr UGT85A23	318	EFAW	GLANS	S	K	Q	F	L	W	I	R	P	D	L	I	S	G	----	ES	S	I	L	G	E	E	V	E	E	T	K	E	----	R	G	L	I	A	S	W	C	H	O	E	V	I	N	H	P	A	I	G	G	F	L	379				
Gj UGT85A24	315	EFAW	GLANS	S	K	Q	F	L	W	I	R	P	D	L	V	S	G	----	DS	A	I	L	P	P	E	L	E	E	T	K	D	----	R	G	L	L	A	S	W	C	P	O	E	V	L	T	H	S	P	A	I	G	G	F	L	376			
Ib UGT85A32	316	EFAW	GLANS	S	K	K	F	L	W	I	R	P	D	L	V	T	G	----	E	A	A	I	P	P	E	L	E	E	T	K	D	----	R	G	M	S	S	W	C	S	O	E	V	L	N	H	P	A	I	G	G	F	L	377					
At UGT85A3	317	EFAW	GLA	A	T	K	E	F	L	W	M	R	D	S	V	A	G	----	E	E	A	V	P	K	E	L	A	E	T	A	D	----	R	R	M	L	T	S	W	C	P	O	E	V	L	T	H	S	P	A	I	G	G	F	L	378			
Sr UGT85A8	313	EFG	W	GLANS	S	K	K	D	F	L	W	I	R	P	D	I	V	G	----	N	E	A	M	P	A	E	L	E	E	T	K	E	----	R	G	M	V	T	S	W	C	S	O	E	V	L	K	H	P	S	I	G	V	F	L	374			
Pd UGT85A19	315	EFS	W	GLANS	S	K	K	F	L	W	I	R	P	D	L	V	T	G	----	E	A	V	V	P	P	E	L	E	E	T	K	E	----	R	G	M	L	A	S	W	C	P	O	E	V	L	T	H	S	P	A	I	G	G	F	L	376		
Vv UGT85A33	316	EFAW	GLANS	S	N	K	F	L	W	I	R	P	D	L	V	G	----	DS	A	I	L	P	P	E	V	T	E	T	K	D	----	R	G	M	L	A	S	W	C	P	O	E	V	L	K	H	P	A	I	G	G	F	L	377					
VvGT16	309	EFAW	GLANS	S	H	K	F	L	W	I	R	P	D	L	V	G	----	DS	V	I	L	P	P	E	V	N	E	T	I	Q	----	R	G	L	M	A	G	W	C	P	O	E	V	L	N	H	P	S	V	G	G	F	L	370					
Mt UGT85H2	315	EFAW	GLANS	C	K	S	F	L	W	I	R	P	D	L	V	I	G	----	GS	V	I	F	S	S	E	T	N	E	I	A	D	----	R	G	L	I	A	S	W	C	P	O	D	R	V	L	N	H	P	S	I	G	G	F	L	376			
Me UGT85K4	314	EFAW	GLANS	S	K	H	F	L	W	I	R	P	D	V	V	M	G	----	ES	A	V	L	P	P	E	V	E	E	T	K	D	----	R	G	L	L	V	S	W	C	P	O	D	R	V	L	O	H	P	A	V	G	V	F	L	375			
Me UGT85K5	314	EFAW	GLANS	S	K	H	F	L	W	I	R	P	D	V	V	M	G	----	ES	A	V	L	P	P	E	V	E	A	L	K	D	----	R	G	L	L	V	S	W	C	P	O	D	R	V	L	O	H	P	A	V	G	V	F	L	375			
Cs UGT1 UGT85K11	316	EFAW	GLANS	S	K	H	F	L	W	I	R	P	D	I	V	M	G	----	DS	A	I	L	P	P	E	V	E	E	T	K	D	----	R	G	L	L	V	S	W	C	P	O	E	V	L	S	H	P	S	I	G	V	F	L	377				
Sr UGT85C1	315	EFG	W	GLV	N	S	N	H	F	L	W	I	R	N	L	I	D	G	----	K	P	A	V	M	P	O	E	L	K	E	A	M	N	E	----	K	G	F	V	G	S	W	C	S	O	E	V	L	N	H	P	A	V	G	G	F	L	376	
Sr UGT85C2	314	EFG	W	GLANS	N	H	F	L	W	I	R	S	N	L	V	I	G	----	EN	A	V	L	P	P	E	L	E	H	T	K	----	R	G	F	I	A	S	W	C	S	O	E	V	L	K	H	P	S	V	G	G	F	L	375					
Sb UGT85B1	322	EFA	L	G	L	S	C	G	S	P	F	L	W	K	R	D	V	V	E	G	----	E	E	V	L	P	E	A	L	D	E	V	A	R	----	G	R	L	V	V	P	W	C	P	O	A	A	V	L	K	H	A	A	V	G	L	E	V	384
At UGT79B1	296	E	L	C	L	E	S	T	G	F	P	L	V	A	I	K	P	S	G	V	S	----	T	V	E	A	L	P	E	G	R	V	Q	----	G	R	-G	V	V	F	G	W	I	Q	P	I	V	L	N	H	P	S	V	G	C	F	V	359	
At UGT79B6	277	E	L	C	L	E	M	E	L	T	G	L</																																															

