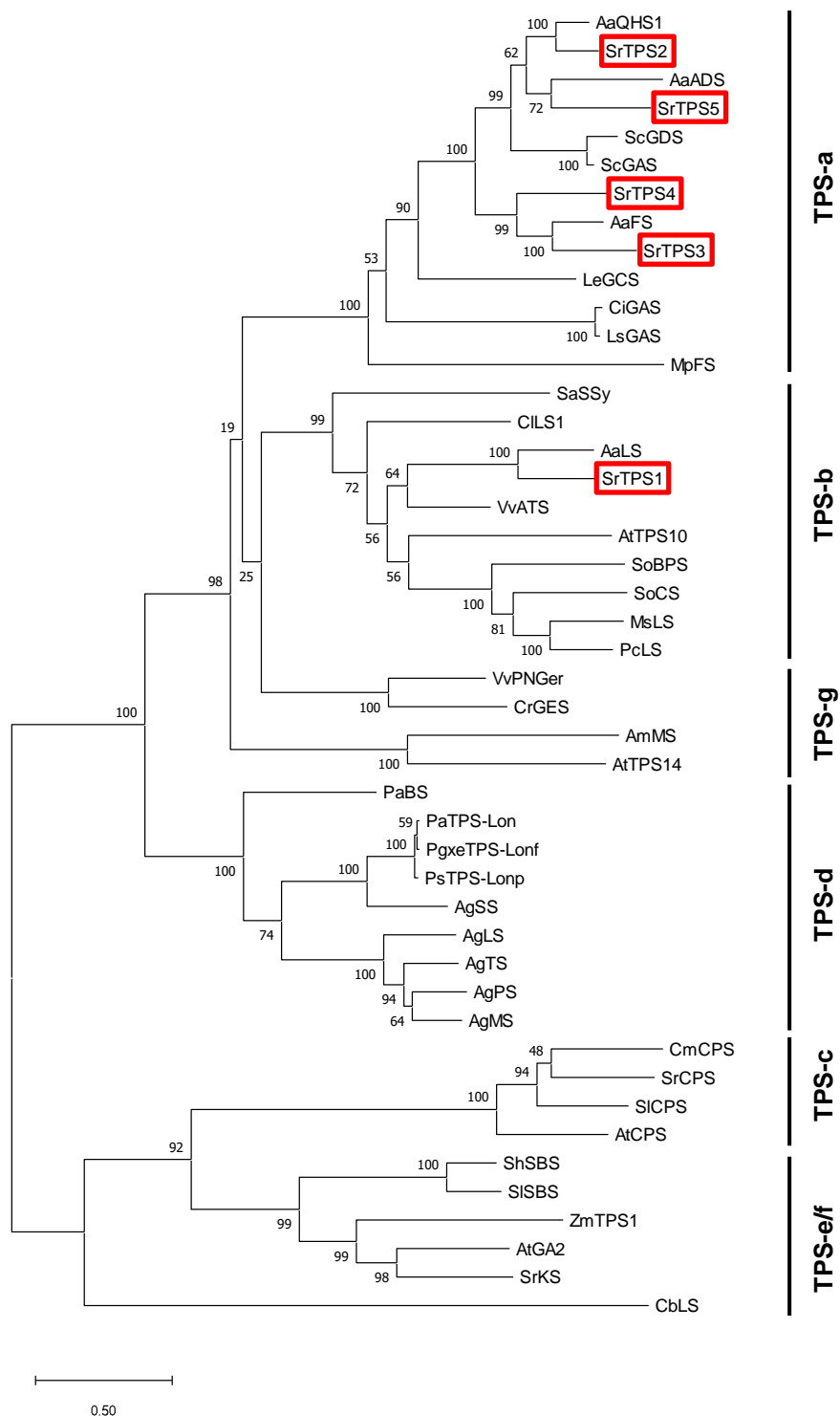


Supplementary Figure S1. Variation in quality and quantity among VOCs identified from Stevia tissues - leaves, flowers, stems and roots. The dotted regions are enlarged in Figure 1A. The arrows indicate the peak of internal standard camphor. TIC, Total ion chromatogram.



Supplementary Figure S2. Molecular phylogenetic analysis of SrTPSs. Multiple sequence alignment was carried out using CLUSTALW and the evolutionary history was inferred by using the maximum likelihood method in MEGA X. Accession numbers of proteins used in this phylogenetic analysis are provided in Table S2.

SrTPS1 1 MASIFFPCSLLFCKKTHVNVSPFNVCKPVNSQPKSIVVEPLVRRSANYQSSVWSYEHVQSLSIKYS GKDYAARADRLK
 SrTPS2 1 -----MSIKQEEVIRPNANFP PSVWGDEFLIYEETGEQGLDQORVEELK
 SrTPS5 1 -----MTSKQEAFRPRRSIS PSIWGDMFLIYEKK-AELGEDLKIVENLK
 SrTPS3 1 MGEL--PIN-----QFLNIAI-MSTLPISSAFFFSSSLPPVARTVTSH PSIWGDQFLTYDEK-EDFVIEKQIVVEELK
 SrTPS4 1 -----MS--LTSLVPNKTSFKQDVIRNSVTFHRSIWGDQFLSYDER-KDQACDQORAKELK

SrTPS1 81 EVVKTMIQKS--SVVEYPLSSIKLVDDLKRLGIAYHFKEEISDILEMLFYQTRDKWYTM-DINLRSLGFRLLRQHGYVPV
 SrTPS2 46 ENVRKDIVSS-LDVQVEHANLLKLIDAIQRLGIAYLFEEIKQALQHIYDTYGEHWKGE----NPSLWFRLLRQQGFYVS
 SrTPS5 44 EEVKKITVA-LKSPNEHTNLLKLIDAIQRLGISYFEEETIANLQKIYEAGDEWNGE-----SPSLWFRLLRQHGYVS
 SrTPS3 70 GEVRKMLMLK-PFEEPMQYVMVKVIDVQIRGLSYHFDGEIEDVLQHIYTTYGNWIDKNEKESVSLWFRLLRQQGFYVS
 SrTPS4 54 EKVRKELVIEGSKDPTQHIKLELIDVVRQLGVAYHFDEIEECLETRIYVMYGDKWINEDSLQSTSLWFRLLRQQGFYVS

SrTPS1	158	QDIFGNFKERIEINLNPCHOEDMIAILNLNLEASYSHSFEDERILDEARDLTKKYLKENLDGIN---	GSIASLVSHALEVPL
SrTPS2	121	SGIENFYKDENGSFKESLTRDVEGLLELYEATYLGVOGEFVLDEALVETRTHLSKIAKDLSEN-	SILSTHIQDALQOPF
SrTPS5	119	CDIFNKYKDEHGAFFKESLTNNVDEMELYDATCLVRGEVVLDEALEFTKTHLDDIAKDPHRTS-	STLSAHIQDALVTPF
SrTPS3	149	SETKHNKTEENGNEKFTLCNDVQGMALYEAAVMRVEGEQVLDEALEFTKTHLIAIAKDPSCD-	SLSRTHIQRALNPL
SrTPS4	134	NEIFYKYKNEDGNFLESMKDDIHGLLSLYEATHMRVEGEDILDEALEFTKSKLTNKNINHVCINDASLE	TOIHQALQOPL

SrTPS1 234 HWRVPRVEARWLEIVYEKRS DANPTLIE LAKLDFNMVQAIHVKDLKHASRWWRNISWDEKLTFFARDRLVECFLWTVGYSY
 SrTPS2 200 RKRLPRLEALRYIPFYQQQDSHNESLVE LAKLGFNLIQSLHRKELSOISRWWKRLDVPNNLSYARDRMVESYFWALGVYF
 SrTPS5 198 HRRTPRIEALS YIRFYEKQVSHNKSLLKAKLGFNLQSQHKSELNQS KWKWKIDVQKNMPPYARDRIVECYFWAFLGYS
 SrTPS3 227 RRTLPRLGAVGYIRFYQQQDPHNEVLL LAKLDFNLQSMHKKELSQLCKWKKDLGNRLPYVRDRDCEGYFWILAVYS
 SrTPS4 214 RKRLPRLEALRYIPFYQQQDSHNHDLTLTAKLDFNLQL ELHRKELSOISKWKNLDSNKLPLYVRDRIVEGYFWILAVY

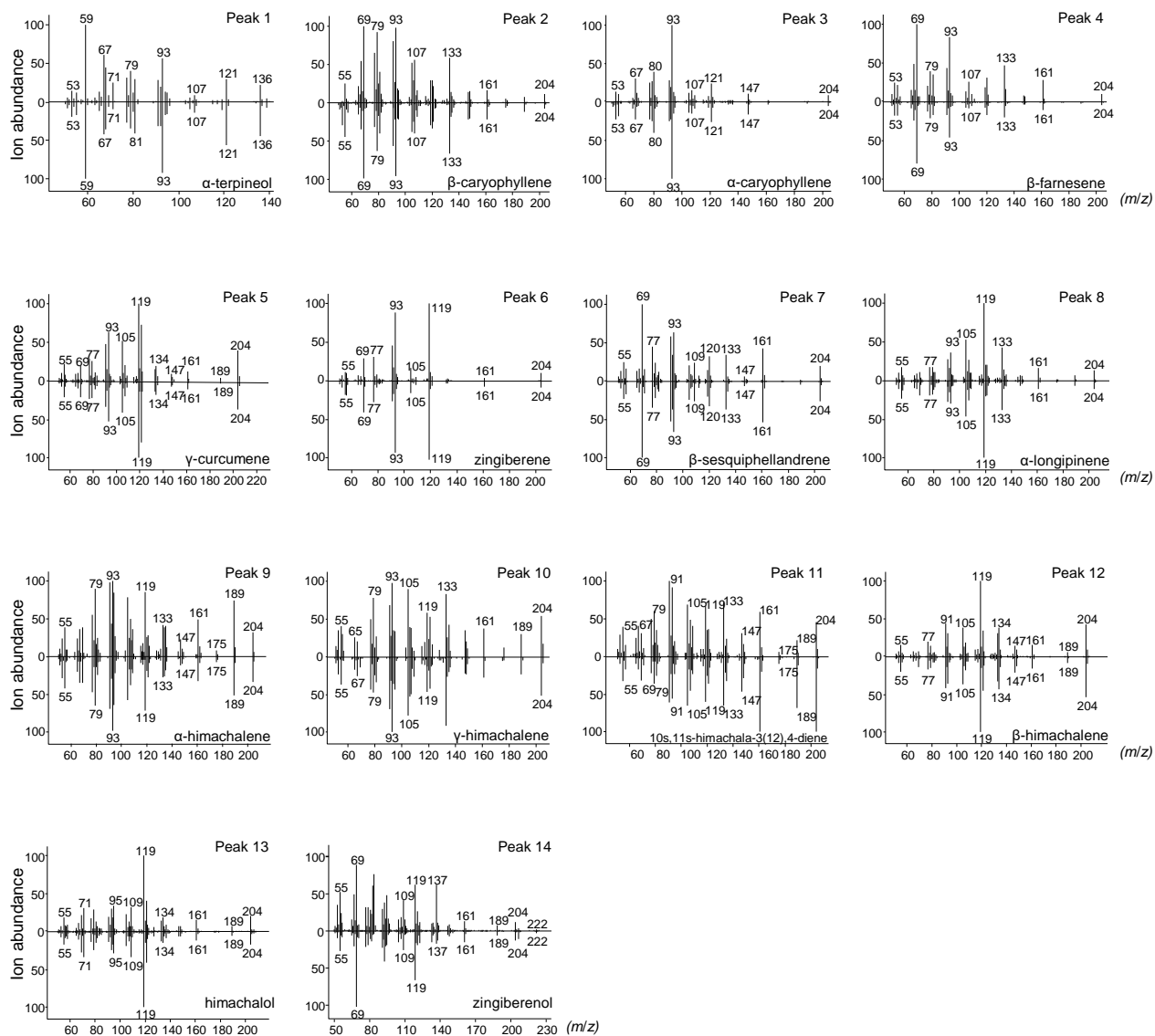
SrTPS1 314 VPRFSLGKRDITKVNAMITTTDDVFDVYGTLDLELEQFTDVISRWDINEVEELPDYMKICFLGYNTINIEIAYNTLINTRI
SrTPS2 280 EPKYSHARVFLAKIISMAAVLDDTYDVYGTYEELKIFTEAIQRWSTCIDVLPEYMKPIYQCLLDMYKEMEIEILGKEGKA
SrTPS5 278 EPKYSLARVFLAKIIQMATVVDVDTFDAYGTYEELQIFTEAVERWCITCLDGLPEYMKVLYQKLLLDLYGEMPEIMEMOEIT
SrTPS3 307 EPEHYTRMFLFELCTCMWLVLDDTVDNVYTYEEFEITFQVAVRWSTISCLDMLPEYMKLIYQCLLDNHDDMESLEKEGKA
SrTPS4 294 EPOHSESRIFLMKACNLVITLDDTYDNVYGTYEELIEFTKAVQTWSISCLDMLPEYMKPIYQCLLDVHKAEADLLLEKKGS

SrTPS1 394 LVLPLVKKAWALCKSYLQEARWYHNGHTPTLTLDYLNNACTITIGTPLTFMHIMISTSINSTOEIFQWHEETENIVHYTAL
 SrTPS2 360 HHLVYAKESMKETFRSYMMEAKWANEGYVPATDEHMAVSFVSSCYGMLTTTHCYLGMGDMVTDSEFKWALTNPPIVKAQSL
 SrTPS5 358 PLFNCSEKFMIFIAVKAMVMEANWVNEGHGPTIDEHTHTIAFATGGDLLTSSCYLGMGDMITNNAINWALTNPPLFKASSV
 SrTPS3 387 YQIHVKEAAKDFVRSGLLEAKWLKEGYKPTLDEYISNASTIGQHVIIARSYVGRGDMVTEFTFRWLATYVPLVLKSSSL
 SrTPS4 374 YRSYTKEMVKEYTRNLLTEAKWVNEKYIPTVEEHMAVTLVTCAYPMIVAKCYVHGDDSVTEDETYKWLSTYPPIVKASSL

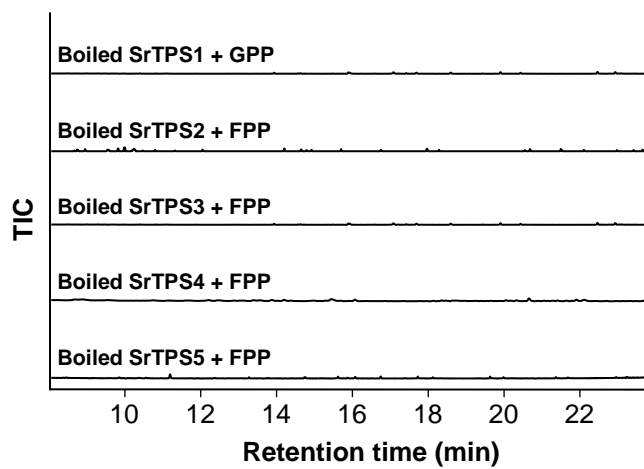
SrTPS1 474 IVRLADDLATSSDEMERGDIKPSIQCYMHENDATEDERRYIKDLIMNTWKKLNEERANAKSTFQQEFIDYAMDIVRMAQ
 SrTPS2 440 IARFMDDIFSQKEKERKNHVSSVESYKQYNVTEDEVHNVFNKKLEDAWKDLTRESLTCKNV-EMPLVIRVINLARVMD
 SrTPS5 438 IGRLLNDIVSYKKEQERHVPSPFYQCYMKNNHDVSEEVAINMVFREEEDVWKDINQDVLMLCKDI-PRPLIVVPIINYARTLY
 SrTPS3 467 IFRLLMDLVTHQREQERDHPSCIECYIKETGASKEAEVFFAKQVEDAWKVINRSLRPTDI-PFSLSPINIS-VIH
 SrTPS4 454 ILRLMDDIATHEEQERNHVASSIECYMKQYGVTEETHCVFSKLVEDNWKIINQESLRPTDV-PIPLLIPPINLARVCD

SrTPS1 554 FMYSGKG DGHGHPNVT-KSEVLSLLFNPIQGI
 SrTPS2 519 VLYKRKDNFTVEVGEELIGHIKSLIIHDNNI-
 SrTPS5 517 YLYKHNDNFSEVGEDIKDHIKSLFIHAMSII-
 SrTPS3 545 VFYGRGNDNYGHAGEEMIGYIKSLLVHPMVV-
 SrTPS4 533 VLYTRGDYYNHGAKCEMIDNIKALLVNPISV-

Supplementary Figure S3. Amino acid sequence alignment of SrTPSs. Deduced amino acid sequences of SrTPSs were aligned using CLUSTALW. The transit peptide sequence of SrTPS1 is underlined in red. The domains essential for TPS activity such as R(R)X₈W motif, the Asp-rich metal cofactor binding regions, DDXXD and NSE/DTE are highlighted. Conserved amino acids are based on 100% (black), 60% (dark gray) and 30% (light gray).



Supplementary Figure S4. The mass spectra of terpenoids described in Figures 4 and 5.



Supplementary Figure S5. *In vitro* enzyme assay using heat-inactivated recombinant proteins. TIC, total ion chromatogram.

