Table S1. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the red onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.468	$C_4H_{10}NO_3S^+$	152.0366	152.0381	9.9
2	Propiin	2.942	$C_6H_{14}NO_3S^+$	180.0681	180.0694	7.2
3	γ-Glutamyl-S-methyl cysteine	3.465	$C_9H_{17}N_2O_5S^+$	265.0851	265.0858	2.6
4	Isoalliin	2.890	$C_6H_{12}NO_3S^+$	178.0529	178.0538	5.1
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	3.803	$C_{11}H_{19}N_2O_6S^+$	307.0946	307.0964	5.9
6	γ-Glutamyl-S-propyl cysteine sulfoxide	4.006	$C_{11}H_{21}N_2O_6S^+$	309.1130	309.1120	3.2
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	3.580	$C_{14}H_{24}N_3O_8S^+$	394.1268	394.1284	4.1
8	S-(1-Propenyl) cysteine	3.465	$C_6H_{12}NO_2S^+$	162.0580	162.0589	5.5
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	3.790	$C_{20}H_{34}N_3O_{13}S^+$	556.1804	556.1812	1.4
10	γ–Glutamyl-S-(2-carboxypropyl) cysteine	3.940	$C_{12}H_{21}N_2O_7S^+$	337.1050	337.1069	5.9
11	S-(2-carboxypropyl) cysteine	3.940	$C_7H_{14}NO_4S^+$	208.0641	208.0643	1.0
11	γ-Glutamyl-S-propyl cysteine	4.006	$C_{11}H_{21}N_2O_5S^+$	293.1157	293.1171	4.8
12	γ-Glutamyl-S-(1-propenyl) cysteine	5.359	$C_{11}H_{19}N_2O_5S^+$	291.1004	291.1015	3.8
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	7.880	$C_{13}H_{22}N_3O_6S_2^+$	380.0938	380.0950	3.2
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	8.960	$C_{13}H_{24}N_3O_6S_2^+$	382.1081	382.1107	6.8

Table S2. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the shallot sample.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.519	$C_4H_{10}NO_3S^+$	152.0372	152.0381	5.9
2	Propiin	2.908	$C_6H_{14}NO_3S^+$	180.0694	180.0694	0.0
3	γ-Glutamyl-S-methyl cysteine	-	$C_9H_{17}N_2O_5S^+$	-	265.0858	-
4	Isoalliin	2.908	$C_6H_{12}NO_3S^+$	178.0542	178.0538	2.25
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	-	$C_{11}H_{19}N_2O_6S^+$	-	307.0964	-
6	γ-Glutamyl-S-propyl cysteine sulfoxide	-	$C_{11}H_{21}N_2O_6S^+$	-	309.1120	-
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	-	$C_{14}H_{24}N_3O_8S^+$	-	394.1284	-
8	S-(1-Propenyl) cysteine	3.635	$C_6H_{12}NO_2S^+$	162.0589	162.0589	0.0
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	-	$C_{20}H_{34}N_3O_{13}S^+$	-	556.1812	-
10	S-(2-carboxypropyl) cysteine	-	$C_7H_{14}NO_4S^+$	-	208.0643	-
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	-	$C_{12}H_{21}N_2O_7S^+$	-	337.1069	-
12	γ-Glutamyl-S-propyl cysteine	3.973	$C_{11}H_{21}N_2O_5S^+$	293.1161	293.1171	3.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	-	$C_{11}H_{19}N_2O_5S^+$	-	291.1015	-
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	-	$C_{13}H_{22}N_3O_6S_2^+$	-	380.0950	-
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	-	$C_{13}H_{24}N_3O_6S_2^+$	-	382.1107	-

Table S3. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the Red Label onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.535	$C_4H_{10}NO_3S^+$	152.0372	152.0381	5.9
2	Propiin	2.941	$C_6H_{14}NO_3S^+$	180.0694	180.0694	0.9
3	γ-Glutamyl-S-methyl cysteine	-	$C_9H_{17}N_2O_5S^+$	-	265.0858	-
4	Isoalliin	2.941	$C_6H_{12}NO_3S^+$	178.0531	178.0538	3.93
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	-	$C_{11}H_{19}N_2O_6S^+$	-	307.0964	-
6	γ-Glutamyl-S-propyl cysteine sulfoxide	-	$C_{11}H_{21}N_2O_6S^+$	-	309.1120	-
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	-	$C_{14}H_{24}N_3O_8S^+$	-	394.1284	-
8	S-(1-Propenyl) cysteine	3.33	$C_6H_{12}NO_2S^+$	162.0589	162.0589	0.0
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	-	$C_{20}H_{34}N_3O_{13}S^+$	-	556.1812	-
10	S-(2-carboxypropyl) cysteine	-	$C_7H_{14}NO_4S^+$	-	208.0643	-
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	-	$C_{12}H_{21}N_2O_7S^+$	-	337.1069	-
12	γ-Glutamyl-S-propyl cysteine	3.904	$C_{11}H_{21}N_2O_5S^+$	293.1175	293.1171	2.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	-	$C_{11}H_{19}N_2O_5S^+$	-	291.1015	-
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	-	$C_{13}H_{22}N_3O_6S_2^+$	-	380.0950	-
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	-	$C_{13}H_{24}N_3O_6S_2^+$	-	382.1107	-

Table S4. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the yellow onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.366	$C_4H_{10}NO_3S^+$	152.0372	152.0381	5.9
2	Propiin	2.992	$C_6H_{14}NO_3S^+$	180.0694	180.0694	0.0
3	γ-Glutamyl-S-methyl cysteine	3.431	$C_9H_{17}N_2O_5S^+$	265.0862	265.0858	1.5
4	γ-Glutamyl-S-propyl cysteine sulfoxide	3.533	$C_{11}H_{21}N_2O_6S^+$	309.1131	309.1120	3.6
5	Isoalliin	2.924	$C_6H_{12}NO_3S^+$	178.0542	178.0538	2.2
6	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	3.499	$C_{11}H_{19}N_2O_6S^+$	307.0967	307.0964	1.0
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	3.530	$C_{14}H_{24}N_3O_8S^+$	394.1297	394.1284	3.3
8	S-(1-Propenyl) cysteine	3.533	$C_6H_{12}NO_2S^+$	162.0589	162.0589	0.0
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	3.620	$C_{20}H_{34}N_3O_{13}S^+$	556.1819	556.1812	1.3
10	S-(2-carboxypropyl) cysteine	3.680	$C_7H_{14}NO_4S^+$	208.0639	208.0643	1.9
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	3.680	$C_{12}H_{21}N_2O_7S^+$	337.1080	337.1069	2.7
12	γ-Glutamyl-S-propyl cysteine	4.344	$C_{11}H_{21}N_2O_5S^+$	293.1175	293.1171	1.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	5.223	$C_{11}H_{19}N_2O_5S^+$	291.1021	291.1015	2.1
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	8.270	$C_{13}H_{22}N_3O_6S_2^+$	380.0949	380.0950	0.3
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	9.095	$C_{13}H_{24}N_3O_6S_2^+$	382.1122	382.1107	6.8

Table S5. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the white onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.501	$C_4H_{10}NO_3S^+$	152.0372	152.0381	5.9
2	Propiin	2.873	$C_6H_{14}NO_3S^+$	180.0694	180.0694	0.0
3	γ-Glutamyl-S-methyl cysteine	-	$C_9H_{17}N_2O_5S^+$	-	265.0858	-
4	Isoalliin	2.873	$C_6H_{12}NO_3S^+$	178.0541	178.0538	1.7
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	-	$C_{11}H_{19}N_2O_6S^+$	-	307.0964	-
6	γ-Glutamyl-S-propyl cysteine sulfoxide	-	$C_{11}H_{21}N_2O_6S^+$	-	309.1120	-
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	-	$C_{14}H_{24}N_3O_8S^+$	-	394.1284	-
8	S-(1-Propenyl) cysteine	3.279	$C_6H_{12}NO_2S^+$	162.0588	162.0589	0.6
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	-	$C_{20}H_{34}N_3O_{13}S^+$	-	556.1812	-
10	S-(2-carboxypropyl) cysteine	-	$C_7H_{14}NO_4S^+$	-	208.0643	-
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	3.670	$C_{12}H_{21}N_2O_7S^+$	337.1080	337.1069	2.7
12	γ-Glutamyl-S-propyl cysteine	3.769	$C_{11}H_{21}N_2O_5S^+$	293.1175	293.1171	1.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	-	$C_{11}H_{19}N_2O_5S^+$	-	291.1015	-
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	-	$C_{13}H_{22}N_3O_6S_2^+$	-	380.0950	-
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	-	$C_{13}H_{24}N_3O_6S_2^+$	-	382.1107	-

Table S6. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the sweet onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m/z*	Calculated *m/z*	Absolute error δ (ppm)
1	Methiin	2.333	$C_4H_{10}NO_3S^+$	152.0391	152.0381	6.6
2	Propiin	2.908	$C_6H_{14}NO_3S^+$	180.0694	180.0694	0.0
3	γ-Glutamyl-S-methyl cysteine	3.381	$C_9H_{17}N_2O_5S^+$	265.0863	265.0858	1.9
4	Isoalliin	2.874	$C_6H_{12}NO_3S^+$	178.0541	178.0538	1.7
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	3.466	$C_{11}H_{19}N_2O_6S^+$	307.0981	307.0964	5.5
6	γ-Glutamyl-S-propyl cysteine sulfoxide	3.466	$C_{11}H_{21}N_2O_6S^+$	309.1131	309.1120	3.6
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	3.480	$C_{14}H_{24}N_3O_8S^+$	394.1312	394.1284	7.1
8	S-(1-Propenyl) cysteine	3.490	$C_6H_{12}NO_2S^+$	162.0588	162.0589	0.6
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	3.580	$C_{20}H_{34}N_3O_{13}S^+$	556.1816	556.1812	0.7
10	S-(2-carboxypropyl) cysteine	3.650	$C_7H_{14}NO_4S^+$	208.0651	208.0643	3.8
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	3.670	$C_{12}H_{21}N_2O_7S^+$	337.1080	337.1069	2.7
11	γ-Glutamyl-S-propyl cysteine	4.277	$C_{11}H_{21}N_2O_5S^+$	293.1175	293.1171	1.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	5.004	$C_{11}H_{19}N_2O_5S^+$	291.1021	291.1015	2.1
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	8.250	$C_{13}H_{22}N_3O_6S_2^+$	380.0948	380.0950	0.5
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	9.095	$C_{13}H_{24}N_3O_6S_2^+$	382.1122	382.1107	3.9

Table S7. LC-ESI-Q-IMS-qToF-based identification of organosulfur compounds in the spring onion variety.

N°	Compounds	Retention time (min)	Ion formulae	Measured *m*/*z*	Calculated *m*/*z*	Absolute error δ (ppm)
1	Methiin	2.348	$C_4H_{10}NO_3S^+$	152.0372	152.0381	5.9
2	Propiin	2.976	$C_6H_{14}NO_3S^+$	180.0704	180.0694	5.6
3	γ-Glutamyl-S-methyl cysteine	3.398	$C_9H_{17}N_2O_5S^+$	265.0876	265.0858	6.8
4	Isoalliin	2.874	$C_6H_{12}NO_3S^+$	178.0552	178.0538	7.9
5	γ-Glutamyl-S-(1-propenyl) cysteine sulfoxide	3.466	$C_{11}H_{19}N_2O_6S^+$	307.0981	307.0964	5.5
6	γ-Glutamyl-S-propyl cysteine sulfoxide	3.466	$C_{11}H_{21}N_2O_6S^+$	309.1131	309.1120	3.6
7	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine	3.500	$C_{14}H_{24}N_3O_8S^+$	394.1297	394.1284	7.1
8	S-(1-Propenyl) cysteine	3.466	$C_6H_{12}NO_2S^+$	162.0598	162.0589	5.6
9	γ–Glutamyl-S-(2-carboxypropyl) cysteine glycine hexoside	3.600	$C_{20}H_{34}N_3O_{13}S^+$	556.1854	556.1812	7.6
10	S-(2-carboxypropyl) cysteine	3.650	$C_7H_{14}NO_4S^+$	208.0662	208.0643	9.1
11	γ–Glutamyl-S-(2-carboxypropyl) cysteine	3.670	$C_{12}H_{21}N_2O_7S^+$	337.1080	337.1069	2.7
12	γ-Glutamyl-S-propyl cysteine	4.261	$C_{11}H_{21}N_2O_5S^+$	293.1175	293.1171	1.4
13	γ-Glutamyl-S-(1-propenyl) cysteine	5.055	$C_{11}H_{19}N_2O_5S^+$	291.1035	291.1015	6.9
14	γ–Glutamyl-S-(S-1-propenyl)cysteine glycine	8.300	$C_{13}H_{22}N_3O_6S_2^+$	380.0979	380.0950	7.6
15	γ–Glutamyl-S-(S-propyl)cysteine-glycine	9.214	$C_{13}H_{24}N_3O_6S_2^+$	382.1122	382.1107	3.9

Table S8. Absolute mean area values quantified by LC-HRMS and GC-MS analytical techniques.

	Area absolute							
	Shallot	Purple	Red	Red Label	Spring	Sweet	White	Yellow
Met	2048.749	3617.318	3400.083	2681.590	3666.606	3056.313	2140.777	1814.675
Pro	1689.944	799.816	789.451	1112.536	328.514	436.295	1417.851	246.441
yGProCS	nd[1]	5811.794	278.748	nd	2806.665	2850.491	nd	3383.035
Iso	16731.977	5553.301	12387.222	10739.161	8497.926	6566.911	19017.010	4379.725
yGIsoCS	nd	75587.063	4984.265	0	70169.563	40753.418	nd	53135.734
yGMetC	nd	17158.963	5586.231	nd	19068.830	15621.348	nd	16331.191
yGIsoC	nd	21204.995	24387.422	nd	29215.514	20676.902	nd	26622.516
yGProC	170.332	2472.227	1305.380	366.667	2082.985	2757.401	524.715	2390.797
yGProCGly	nd	1613.392	768.341	nd	1244.316	1013.294	nd	961.281
yGIsoCGly	nd	1283.961	429.337	nd	786.078	631.501	nd	300.683
IsoC	829.857	121.238	1034.786	659.691	190.608	62.081	30.676	89.417
yG(2-carboxy)CGly	nd	60653.109	6140.687	nd	66115.773	56743.746	nd	57235.965
yG(2-carboxy)CGlyHex	nd	336.493	886.996	nd	641.535	379.290	nd	524.650
yG(2-carboxy)C	nd	147.545	858.152	nd	251.701	405.571	nd	112.320
(2-carboxy)C	nd	157.991	141.818	nd	169.247	303.680	nd	195.546
Meth	147904.281	201211.272	77050.691	101518.221	61318.503	95576.384	65470.381	39865.637
di-me Su	470511.160	398542.198	125977.970	215202.024	195806.252	417212.411	211556.172	123368.870
1-proth	496411.186	462376.228	152577.124	226416.526	152042.192	186313.369	406687.770	197955.787
SO2	396560.293	291885.822	292701.775	53775.771	0	202618.587	545197.823	281431.667
Met-thi	504652.825	632116.752	390431.569	431641.197	562534.880	345281.915	1020587.990	384281.940
di-me di-Su	634626.123	371626.365	211831.028	301544.499	241478.635	349392.504	142008.337	138591.793
2-met-thiph	67786.284	11076.491	34235.506	51654.014	10970.575	11498.931	26129.342	9298.048
3-met-thiph	100745.761	48096.440	74335.472	88264.454	38759.781	34494.615	71793.187	38332.314
2,5-met-thiph	30612.501	23851.065	19771.485	26980.585	36206.265	29487.044	27968.502	26568.892
3,4-met-thiph	213303.759	98619.189	109775.984	135638.639	114195.776	76796.191	162297.192	92961.742
Allyl(iso)sulf	48321.364	15513.494	16573.315	17158.370	5198.5093	33806.022	24201.800	10091.912

thi-pro	33831.983	69180.994	78399.683	78688.005	21715.993	50087.414	110123.683	41114.912
ox pro	65013.306	1039701.580	477319.847	246962.629	704789.189	933693.931	637416.849	1457999.570
me pro di-Su	204291.927	173392.084	44701.422	73744.374	55225.993	78423.266	99336.0164	51764.296
2,4-met-thiph	3276364.080	2813082.950	2229023.670	2539020.240	3023381.10	2080018.170	3121947.38	2572619.290
ZMeth(iso)disulf	299113.779	346990.278	184867.460	246615.665	341796.803	268435.527	193018.026	201672.389
EMeth(iso)disulf	796100.905	955595.362	513312.782	688933.916	930800.200	779947.957	588010.925	498780.256
di-me tri-Su	855159.470	572503.084	408652.797	480981.638	486616.480	517957.391	302155.609	310570.611

[1]nd = Not detected.

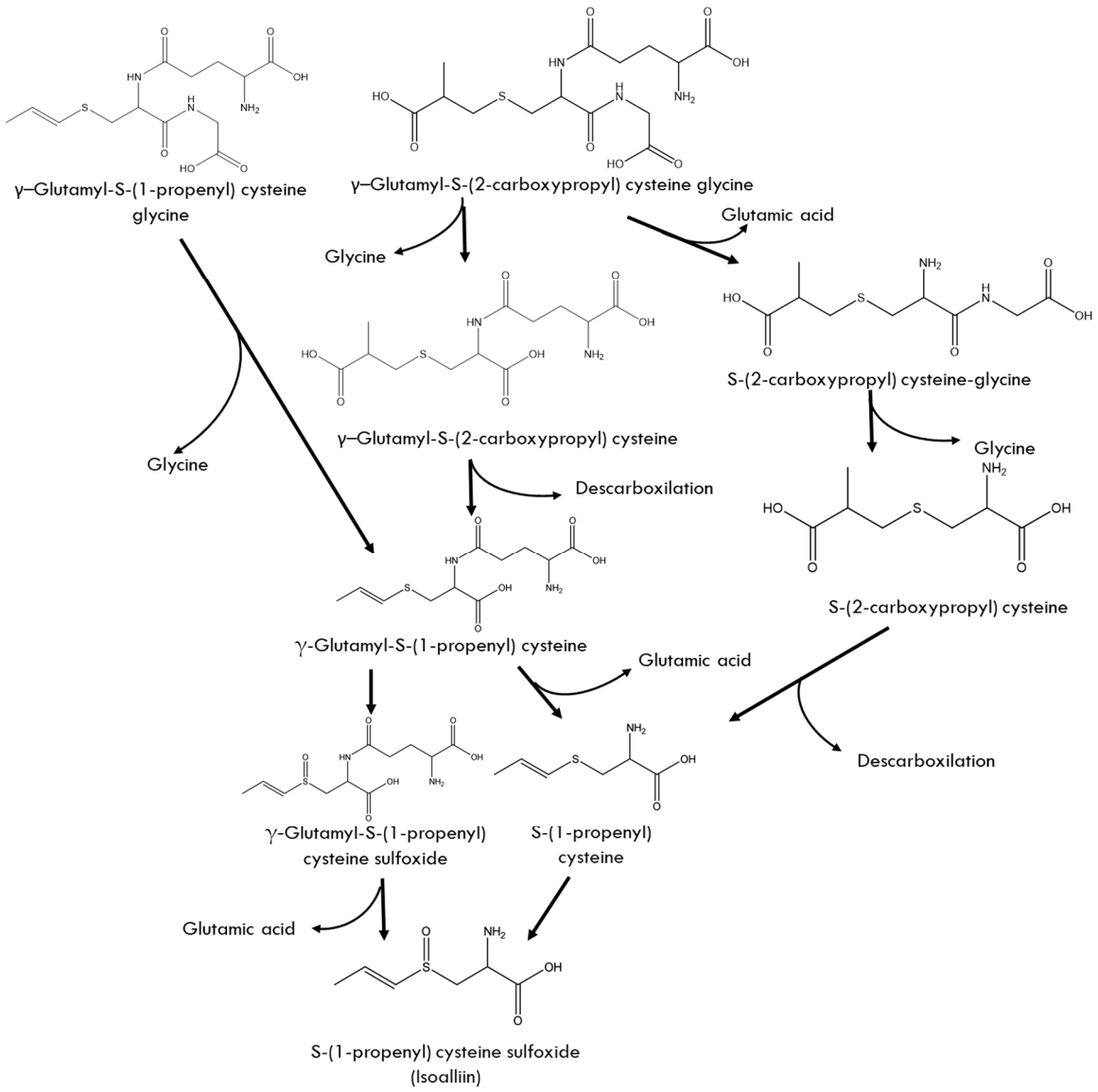


Figure S1. Metabolic pathways for the synthesis of isoalliin.