

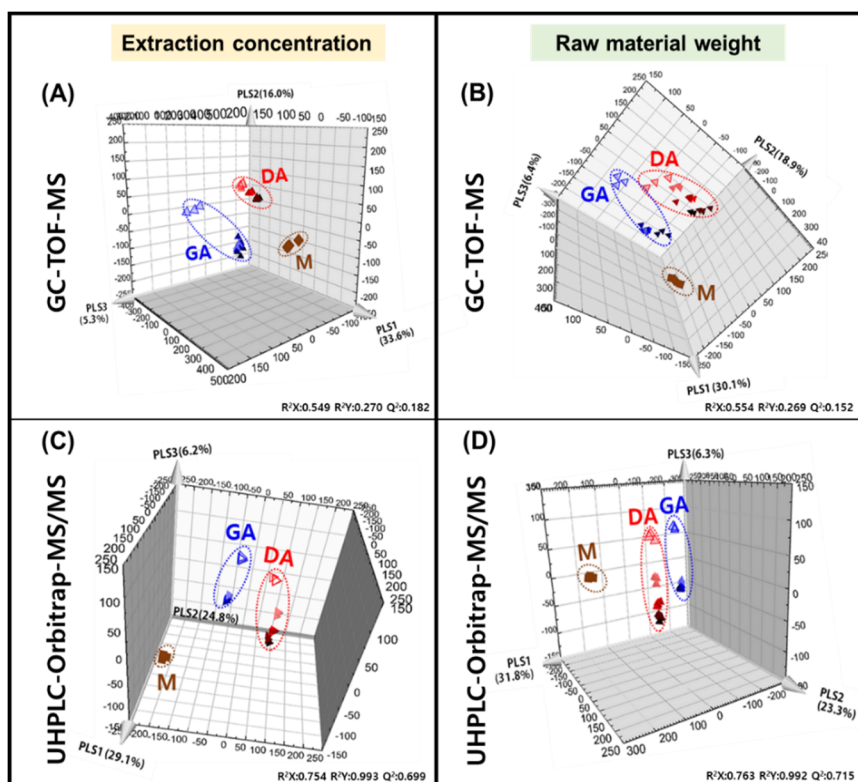


Figure S1. PLS-DA score plots based on concentrations derived from GC-TOF-MS (A) and UHPLC-Orbitrap-MS/MS (C). PLS-DA score plots normalized to raw material weight derived from GC-TOF-MS (B) and UHPLC-Orbitrap-MS/MS (D). Different processes symbolized as: *meju* (raw material: $\blacklozenge$), *doenjang* aging (0 d: $\blacktriangle$; 60 d: $\blacktriangle$; 90 d: $\blacktriangle$; 120 d: $\blacktriangle$; 360 d: $\blacktriangle$), *ganjang* aging (0 d: $\blacktriangle$; 60 d: $\blacktriangle$; 90 d: $\blacktriangle$; 120 d: $\blacktriangle$; 190 d: $\blacktriangle$).

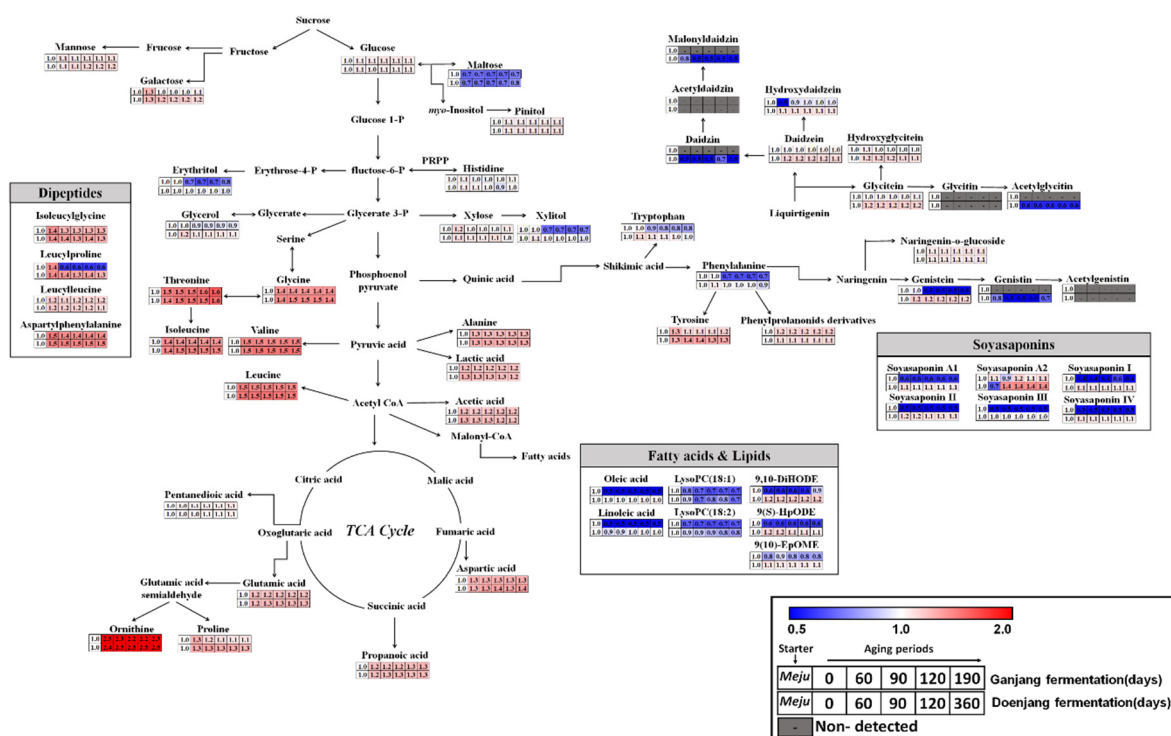


Figure. S2. Pathways of the relative levels of discriminant metabolites at different times of *doenjang* and *ganjang* as determined using the PLS-DA data sets based on raw material weight (VIP > 1.0) for GC-TOF-MS and UHPLC-Orbitrap-MS/MS analyses. The discriminant metabolites were further correlated with corresponding steps in the biosynthetic pathways adapted from the Kyoto Encyclopedia of Genes and Genomes database. The values indicate the log10-transformed fold changes based on the values of *meju*.

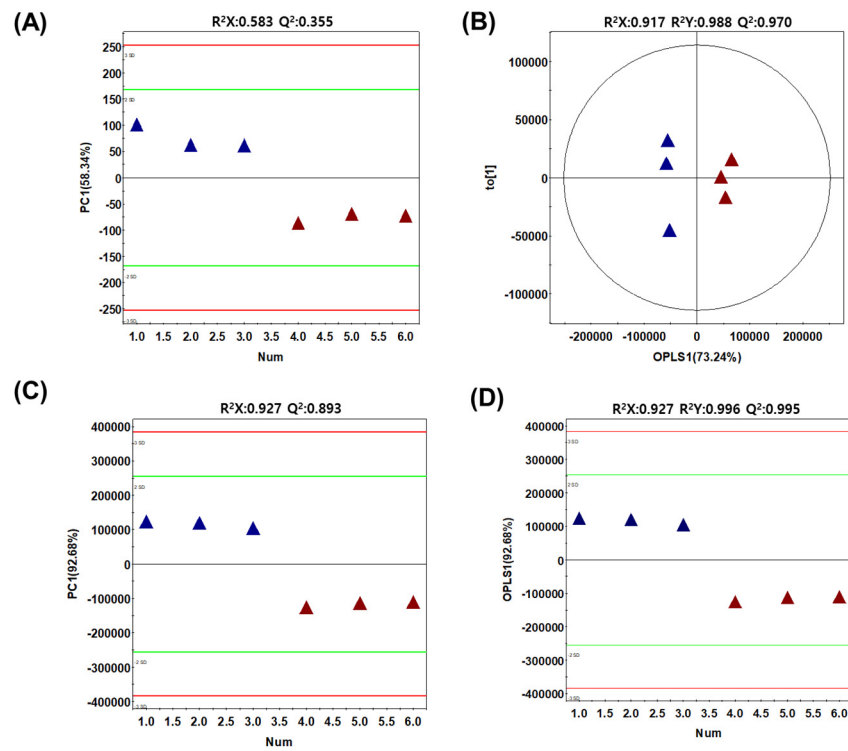


Figure. S3. PCA and OPLS-DA score plots derived from non-targeted metabolite profiling of *doenjang* and *ganjang* end products analyzed using GC-TOF-MS (A, B) and UHPLC-Orbitrap-MS/MS (C, D) (VIP > 1.5, $p < 0.05$). The score plot color codes indicate *doenjang* 360 d (▲) and *ganjang* 190 d (▲).

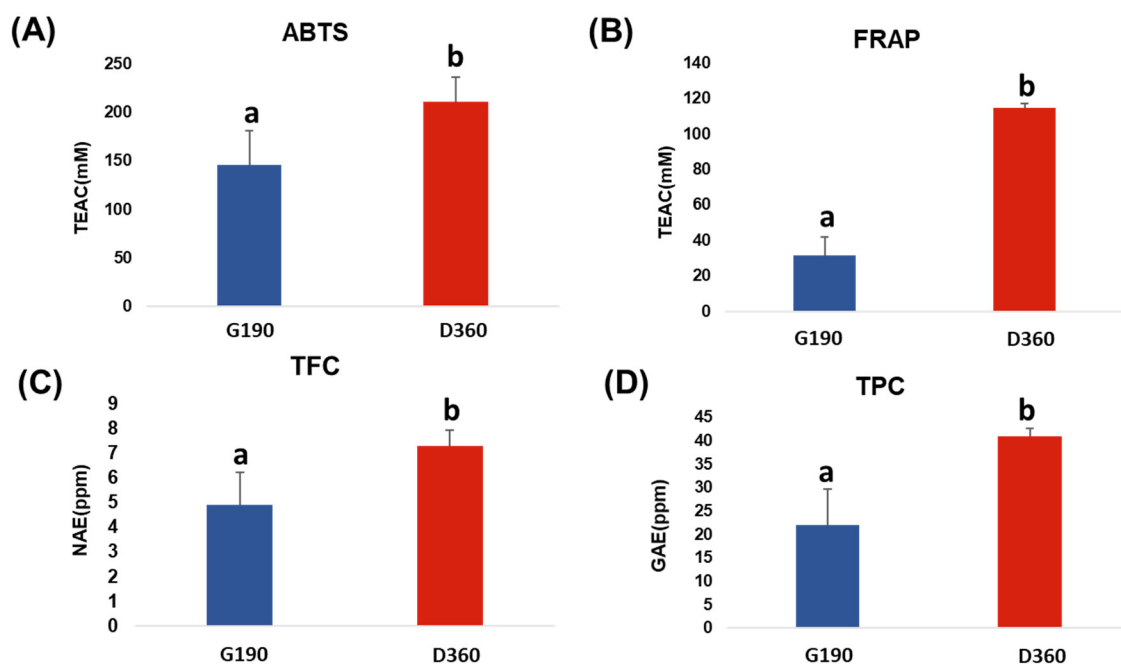


Figure. S4. Antioxidant activity analysis using ABTS (A), FRAP (B), TPC (C), and TFC (D) of *doenjang* (D360), and *ganjang* (G190) end-products. Different letters in the bar graph indicate significant differences as analyzed by ANOVA followed by Duncan's new multiple range test.

Table S1. Tentatively identified *meju* metabolites from different materials based on the GC-TOF-MS analysis.

NO.	Tentative identification ^a	GC-TOF-MS			
		RT(min) ^b	Identified ion (m/z)	Mass Fragment pattern	TMS ^c
	<u>Amino acids</u>				
1	Alanine	5.5	116	190,147,133,116,103,73,66	2
2	Valine	6.7	218	218,144,133,117,100,86,73,59	2
3	Leucine	7.2	102	158,138,102,75,73	2
4	Isoleucine	7.5	158	218,117,75,147,45,158,73	3
5	Proline	7.5	142	216,142,133,100,73,59	2
6	Glycine	7.6	174	248,188,174,158,133,117,100,86,73	3
7	Threonine	8.3	219	291,219,203,159,129,117,101,86	3
8	Aspartic acid	9.5	232	232,218,202,188,174	3
9	Glutamic acid	10.3	128	230,204,174,147,128,114,100,84,73	3
10	Ornithine	11.3	186	216,186,174,159,142,130,116,100,86	4
11	Tyrosine	12.6	100	147,133,100,73,59	3
12	Phenylalanine	10.4	218	218,192,177,160,147,120,100,91	2
13	Histidine	13.8	154	73,203,154,147,103,75,74,59	3
14	Tryptophan	14.3	202	348,231,202,174,130,95,73,45	3
	<u>Fatty acids</u>				
15	Linoleic acid	14.3	131	233,219,205,190,147	2
16	Oleic acid	14.2	117	313,201,117,75,73	1
	<u>Organic acids</u>				
17	Lactic acid	5.0	117	147,133,117,101,88,73,66	2
18	Acetic acid	5.1	66	177,147,133,117,103,81,73,66	2
19	Propanoic acid	6.1	147	233,218,177,147,130	2
20	Pentanedioic acid	9.9	129	147,129,116,103,101,85,75	3
	<u>Sugar&Sugar derivatives</u>				
21	Glycerol	7.3	117	147,133,117,103,89,73,59,55	3
22	Erythritol	9.4	217	307,277,217,189,147,103,73	4
23	Xylose	10.7	103	233,217,204,189,160	4
24	Xylitol	11.1	217	217,205,189,157,147	5
25	Maltose	16.7	361	174,130,100,86,73,59	8
26	Galactose	11.3	117	160,128,117,89,73,58	4
27	Mannose	12.3	319	160,147,129,117,103,89,73,59	5
28	Glucose	12.4	160	160,147,129,117,103,89	5
29	Pinitol	11.9	260	217,207,191,177,159	5
	<u>Non-Identifications</u>				
30	N.I 1	7.0	179	179,135,105,77,51	1
31	N.I 2	7.2	174	174,147,100,73,45,50	3
32	N.I 3	7.9	99	147,126,113,99,85,73,56	2
33	N.I 4	9.7	263	263,247,207,175,115,91	0
34	N.I 5	10.3	174	228,200,174,147,129,116,100,82,73	3
35	N.I 6	12.6	319	319,217,189,147,103,73,59	5
36	N.I 7	12.7	217	319,217,147,103,73,59	6

* The differential metabolites were selected using the VIP (>1.0) and *p*-values (<0.7) from the partial least squares-discriminant analysis model in Figure 2A. ^a: Retention time; ^c: TMS, trimethylsilyl; ^a: confirmed with the National Institutes of Standards and Technology (NIST) database and in-house libraries; STD, mass spectrum, consistent with that of the standard compounds.

Table S2. Tentatively identified *meju* metabolites from different materials based on the UHPLC-Orbitrap-MS/MS analysis.

NO.	Tentative identification	UHPLC-Orbitrap-MS/MS							
		RT(min) ^a	[M-H] ⁻	[M+H] ⁺	M.W. ^b	M.F. ^c [M-H] ⁺	Error (ppm)	MS fragmentation	ID
	<u>Dipeptides</u>								
37	Isoleucylglycine	1.32	187.1114	189.1224	188	C8 H17 O3 N2	-5.49	196,171,143>125,86	GNPS ^d
38	Leucylproline	1.94	227.1074	229.1534	228	C11 H21 O3 N2	-5.49	211,183,155,116>78,70	GNPS ^d
39	Leucylleucine	4.09	243.1745	245.1847	244	C12 H25 O3 N2	-5.10	235,227,199>69,57	GNPS ^d
40	Aspartylphenylalanine	2.36	279.1000	281.1144	280	C13 H17 O5 N2	-5.12	261>217,175,147,83	HMDB ^e
	<u>Biogenic amines</u>								
41	Phenylethylamine	1.44	-	122.0971	121	C8 H12 N	-5.70	119,112,105>79,76,68	STD
42	Histamine	0.65	-	112.0875	111	C5 H10 N3	-2.98	108,97,95>68	STD
43	Tryptamine	3.02	-	161.1080	160	C10 H12 N2	-7.36	-	STD
	<u>Phenylpropanoids</u>								
44	Phenylpropanoid derivatives	3.36	293.1187	295.1306	294	C19 H17 O3 [-]	1.34	275,221,187,>164,127(-)	HMDB ^e
	<u>Flavonoids</u>								
45	Malonyldaidzin	4.92	-	503.1198	502	C24 H23 O12	-1.93	457,344,253>224,209,181(-)	Ref [1]
46	Acetyldaidzin	5.26	457.1139	459.1275	458	C23 H23 O10	-2.27	441,255>227,199,137	Ref [1]
47	Acetylglucitin	5.30	487.1245	489.1379	488	C24 H25 O11	-2.57	-	Ref [1]
48	Acetylgenistin	5.71	473.1080	475.1223	474	C23 H23 O11	-2.42	457,271>243,215,153	Ref [1]
49	Daidzin	4.51	415.1034	417.1171	416	C21 H21 O9	-2.04	287,255>227,199,137	Ref [1]
50	Glycitin	4.61	445.1133	447.1274	446	C22 H23 O10	-2.60	429,285>270,229,167	Ref [1]
51	Genistin	4.92	431.0977	433.1140	432	C21 H21 O10	-1.99	433>271,253,243,215,153	Ref [1]
52	Daidzein	5.67	253.0524	255.0676	254	C15 H11 O4	4.50	255,227,199>181,170,157	Ref [1]
53	Glycitein	5.78	283.0644	285.0786	284	C16 H13 O5	-5.47	270,253,229,166>152,108	Ref [1]
54	Genistein	6.31	269.0500	271.0625	270	C15 H11 O5	4.70	253,243,158,152>147,130	Ref [1]
55	Hydroxydaidzein	4.89	269.0492	271.0600	270	C15 H11 O5	-5.20	271,253,243,225>152,132	Ref [2]
56	Hydroxyglycitein	5.05	299.0598	301.0692	300	C16 H13 O6	-4.83	301,286,245,213>167,139	Ref [2]
57	Naringenin-O-glucoside	3.62	433.1181	435.1332	434	C21H23O10	-4.28	433> 415, 343, 313> 285, 269	Ref [3]
	<u>Soyasaponins</u>								
58	Soyasaponin A1	5.08	1267.6111	1269.6049	1269	C59 H97 O29	-4.84	1107,945,747,615>351,203	Ref [4]
59	Soyasaponin A2	5.31	1105.5488	1107.5699	1107	C53 H87 O24	-6.24	945,813,729,597,579>403,363,253	Ref [4]
60	Soyasaponin I	6.99	941.5245	943.5392	943	C48 H77 O18P [-]	4.73	925,797,617,581,551>423,405,351	Ref [5]
61	Soyasaponin II	7.19	911.5071	913.5102	913	C47 H77 O17	-5.84	893,615,525,457>437,409,393,233(-)	Ref [5]
62	Soyasaponin III	7.22	795.4650	797.4760	797	C42 H69 O14	-6.71	777,759,613>467,413(-)	Ref [5]
63	Soyasaponin IV	7.32	765.4499	767.4536	766	C41 H67 O13	-5.30	615,457,409,357>295(-)	Ref [5]
64	Soyasapogenol A	9.75	-	457.3678 ^f	475	C30 H49 O3	6.26	439,421,403>393	STD
65	Soyasapogenol B	10.06	-	441.3697 ^f	458	C30 H49 O2	6.02	423,405>283,203,187	STD
	<u>Lipids & Fatty acids</u>								
66	LysoPC(18:2)	8.39	518.2715	520.3448	519	C26 H51 O7 N P	-5.83	520> 502> 443> 184	Ref [6]
67	LysoPC(18:0)	9.52	568.3691 ^g	524.3757	523	C26 H55 O7 N P	-5.41	524> 506, 447> 311, 184	Ref [6]
68	9,10-DiHODE	7.11	311.2258	313.2370	312	C18 H33 O4	-4.57	275,249>231,177,127	Ref [7]
69	9(S)-HpODE	7.60	311.2256	313.2359	312	C18 H33 O4	-5.66	275,249>231,177,97	Ref [7]
70	9(10)-EpOME	9.56	295.2313	297.2408	296	C18 H33 O3	-9.11	277,249>155,141,127	GNPS ^d
	<u>Non-identification</u>								
71	N.I. 7	10.27	390.3045	392.3195	391	-	-	391,374,346,261,243>241,212	-
72	N.I. 8	7.83	374.2517	376.2627	375	-	-	358,343,302,293>218,135,107	-
73	N.I. 9	10.47	378.3048	380.3195	379	-	-	379,362,334>243,212,170	-

* The differential metabolites were selected using the VIP (>1.0) and *p*-values (<0.7) from the partial least squares discriminant analysis model in Figure 3A. ^a: RT, retention time; ^b: M.W., molecular weight; ^c: M.F., molecular formula; ^d: GNPS, <https://gnps.ucsd.edu/>; ^e: HMDB, <https://hmdb.ca/>; ^f: [M-H₂O]⁺; ^g: [M-FA+H]⁺.

Table S3. Bacterial and fungal illumina data sets derived from *meju*, *doenjang*, and *ganjang* samples and their statistical diversity analysis.

Sample name	Time (day)	Bacteria ^a				Fungi ^a			
		High quality reads	OTUs	Chao1	Shannone-Weaver	High quality reads	OTUs	Chao1	Shannone-Weaver
<i>Meju</i>	–	9,773	33	36.9	3.3	127,781	15	15.6	0.1
	0	1,450	66	75.1	4.6	34,709	16	16	2.4
	60	14,243	69	98.0	4.2	43,934	18	18	2.8
<i>Doenjang</i>	90	11,645	69	90.5	4.3	36,376	13	13	1.4
	120	7,738	59	81.4	4.1	42,927	17	17	2.1
	360	944	37	37.9	3.6	50,337	28	28	2.7
	0	13,583	66	89.0	4.4	32,272	15	15	2.4
	60	4,866	58	67.5	4.0	95,935	17	16.2	1.2
<i>Ganjang</i>	90	5,421	50	56.5	4.0	37,723	29	29	2.4
	120	6,875	46	53.6	3.9	101,559	43	42.7	2.0
	190	9,174	44	58.9	3.8	85,438	32	32.1	2.0

Abbreviation: OTU, operational taxonomic unit.

^a The bacterial and fungal sequences in each *doenjang* samples were normalized to 944 and 32,272, respectively and diversity indices in each *doenjang* samples were calculated using the normalized sequences.

Supplementary references

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