Title: Impact of various microbial-fermented methods on the chemical profile of dark tea using a single raw tea material

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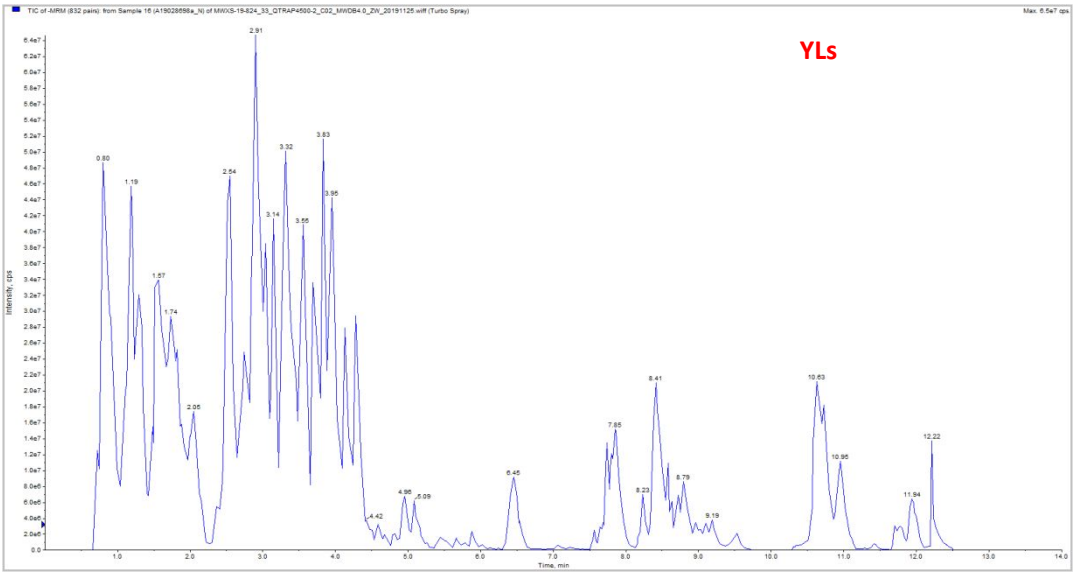
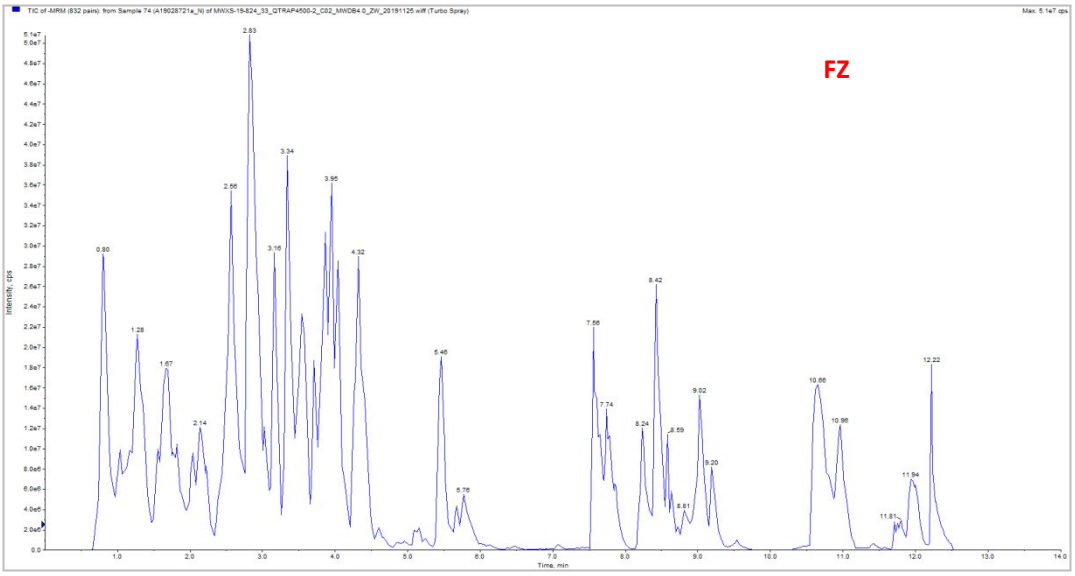
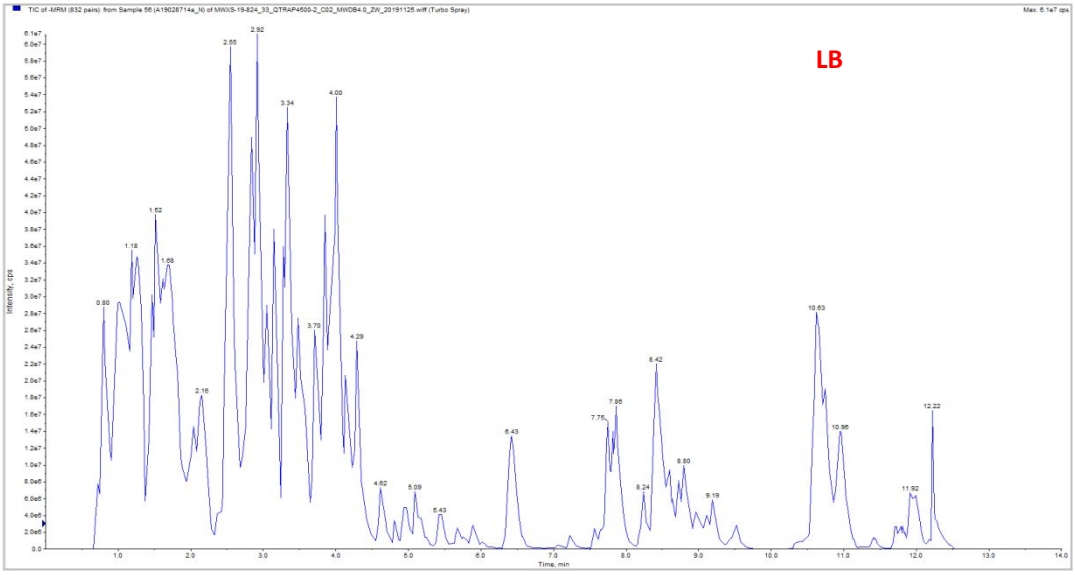

YLs
Intensity, cps
Time, min


FZ
Intensity, cps
Time, min


LB
Intensity, cps
Time, min

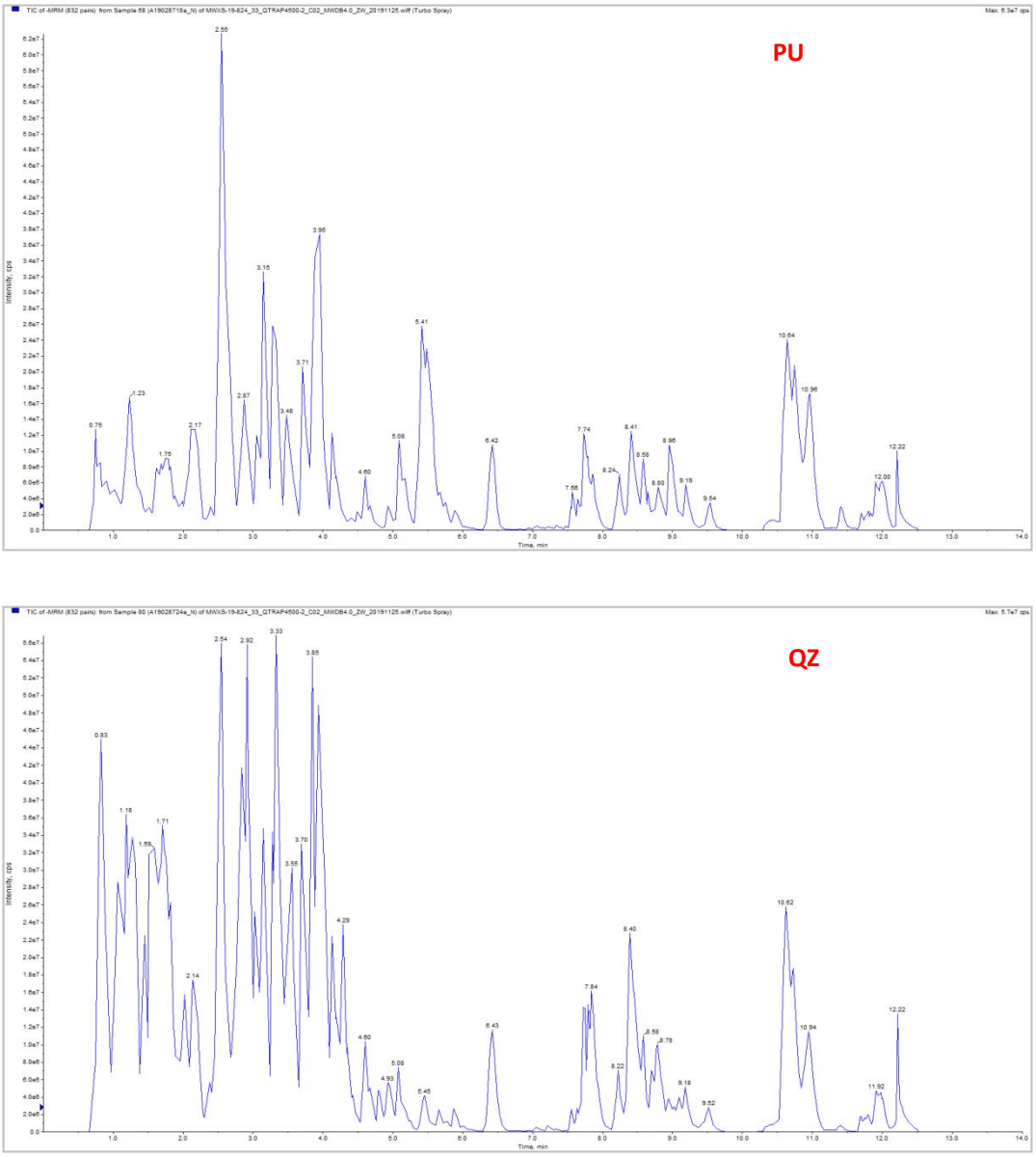
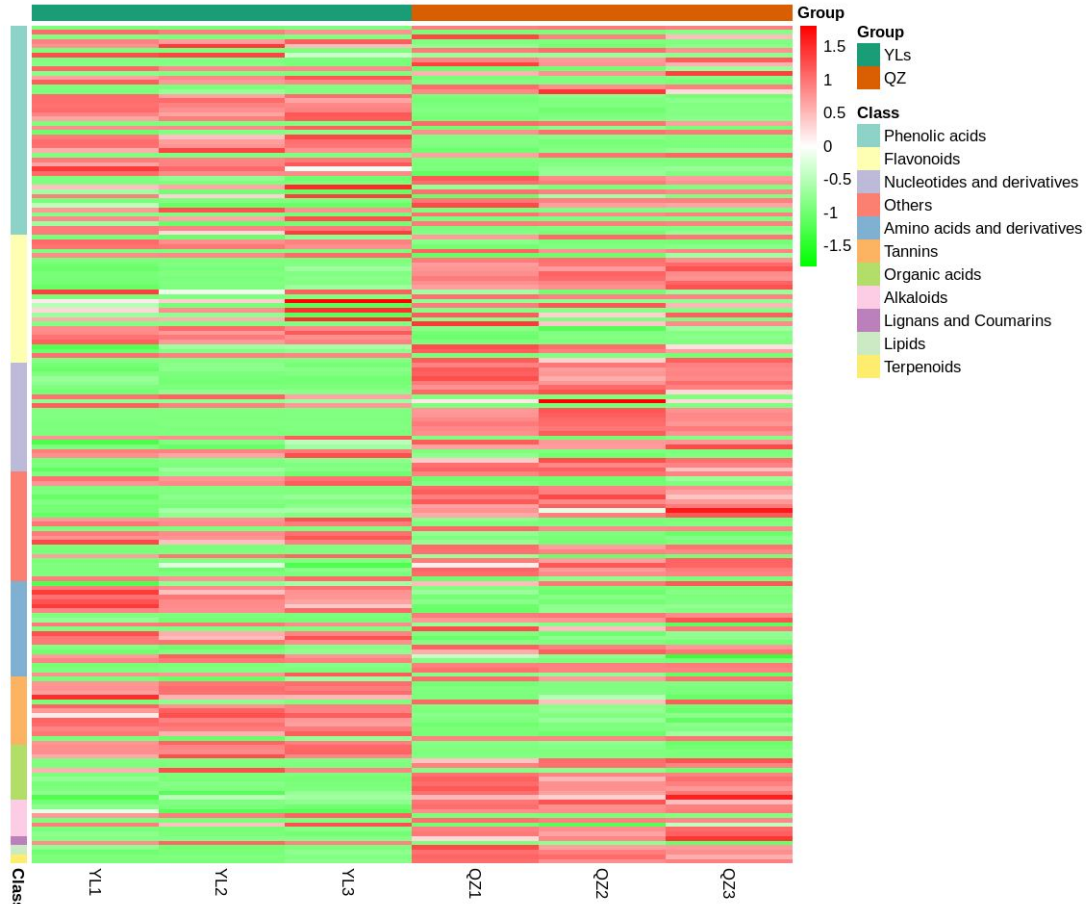
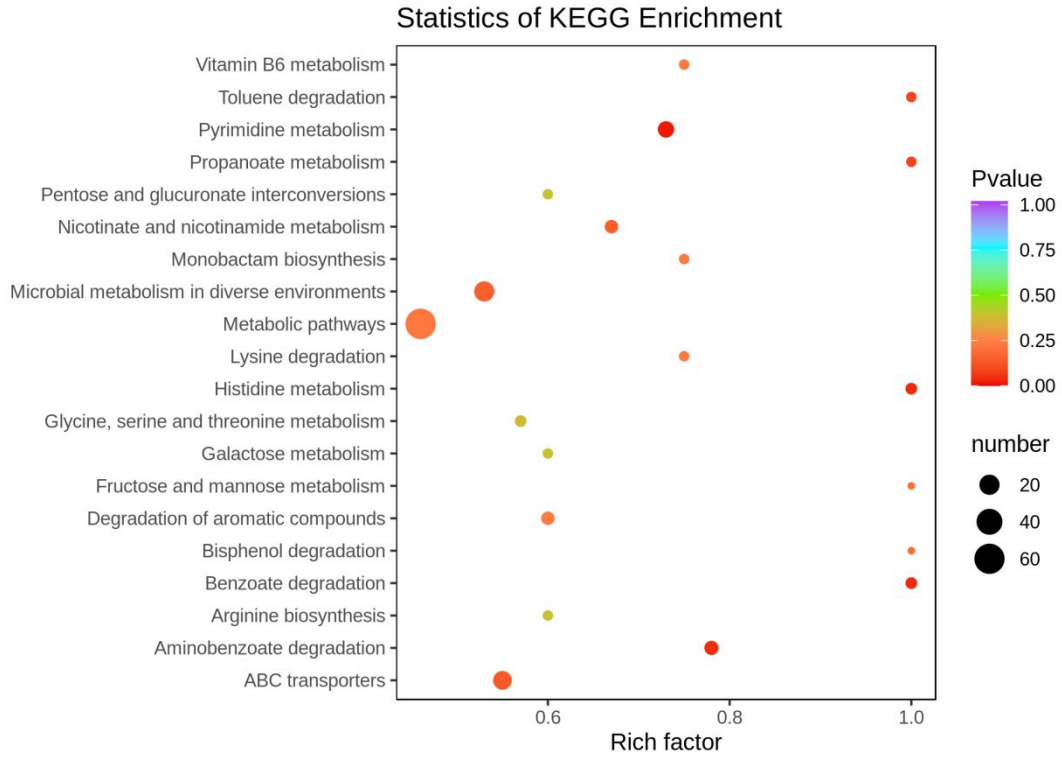


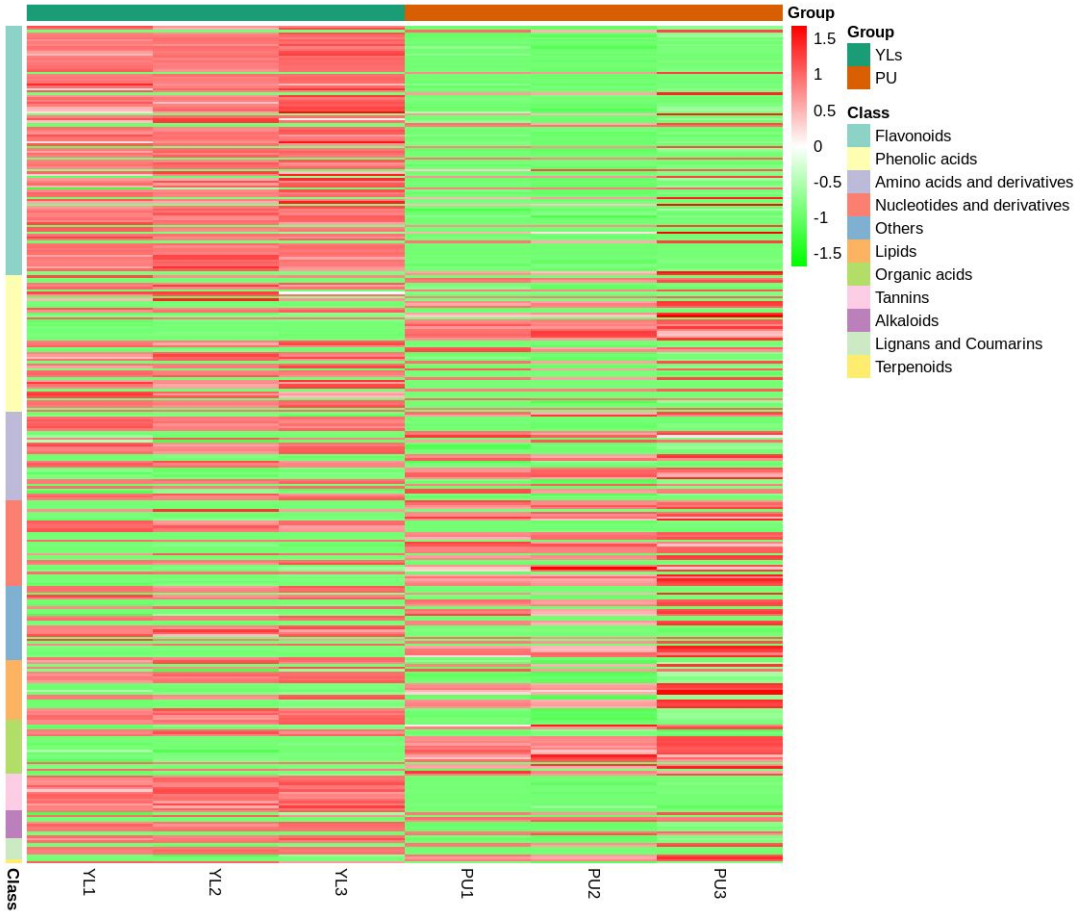
Figure S1. Typical total ion current chromatograms (TIC) of dark raw tea materials (YLs), Fuzhuan (FZ) tea, Liu-~~p~~bao (LB) tea, Pu-erh (PU) tea, and Qingzhuan (QZ) tea.

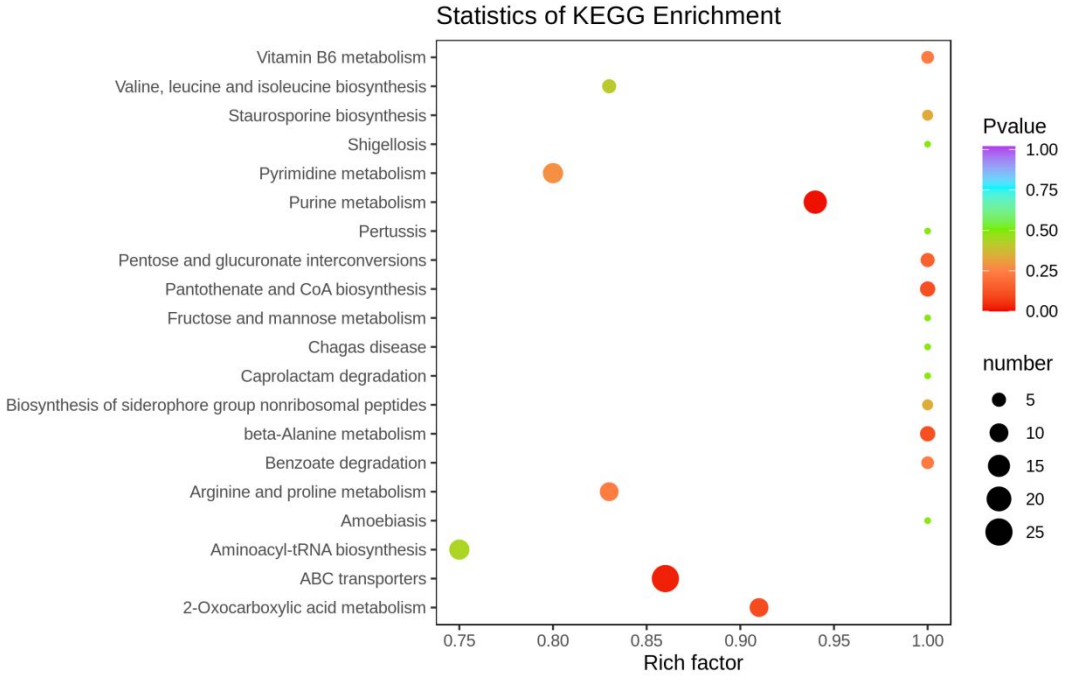
A


Group
1.5
1
0.5
0
-0.5
-1
-1.5
Group
YLs
QZ
Class
Phenolic acids
Flavonoids
Nucleotides and derivatives
Others
Amino acids and derivatives
Tannins
Organic acids
Alkaloids
Lignans and Coumarins
Lipids
Terpenoids
Class
YL1
YL2
YL3
QZ1
QZ2
QZ3

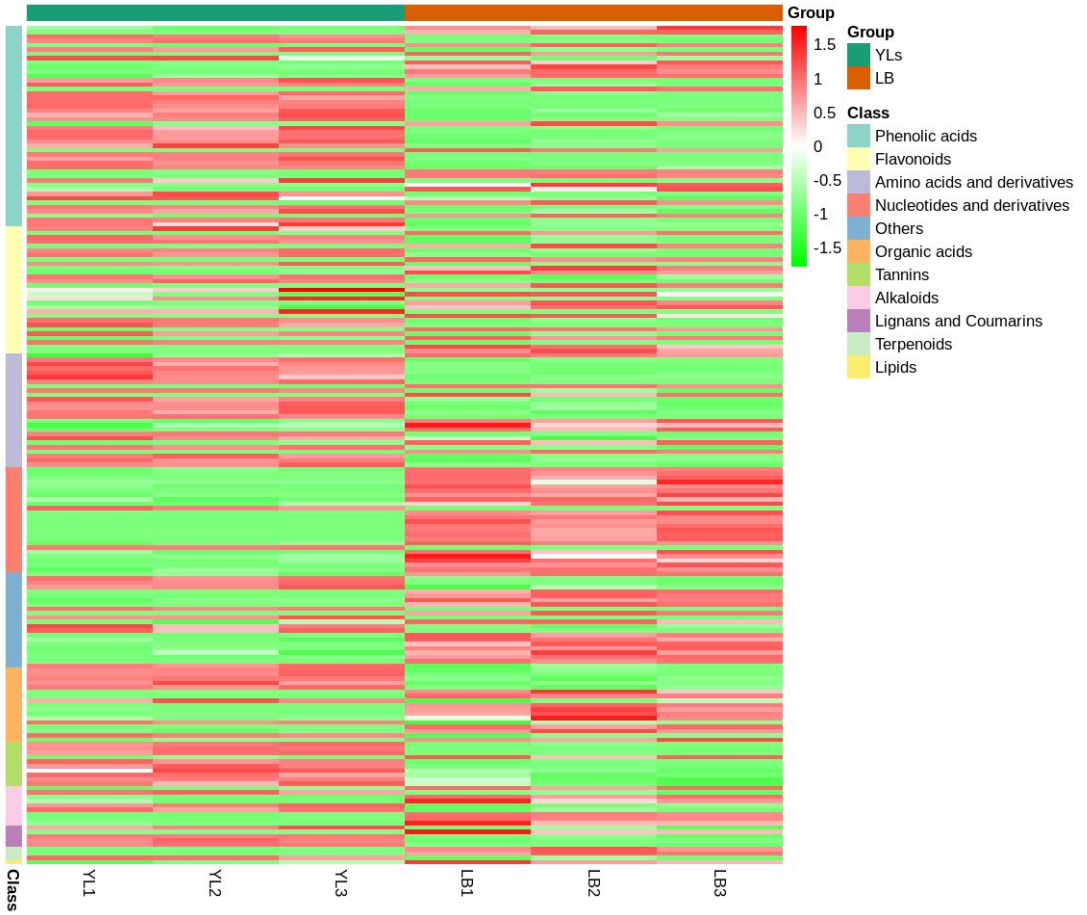

Statistics of KEGG Enrichment
Vitamin B6 metabolism
Toluene degradation
Pyrimidine metabolism
Propanoate metabolism
Pentose and glucuronate interconversions
Nicotinate and nicotinamide metabolism
Monobactam biosynthesis
Microbial metabolism in diverse environments
Metabolic pathways
Lysine degradation
Histidine metabolism
Glycine, serine and threonine metabolism
Galactose metabolism
Fructose and mannose metabolism
Degradation of aromatic compounds
Bisphenol degradation
Benzoate degradation
Arginine biosynthesis
Aminobenzoate degradation
ABC transporters
0.6
0.8
1.0
Rich factor
Pvalue
1.00
0.75
0.50
0.25
0.00
number
20
40
60

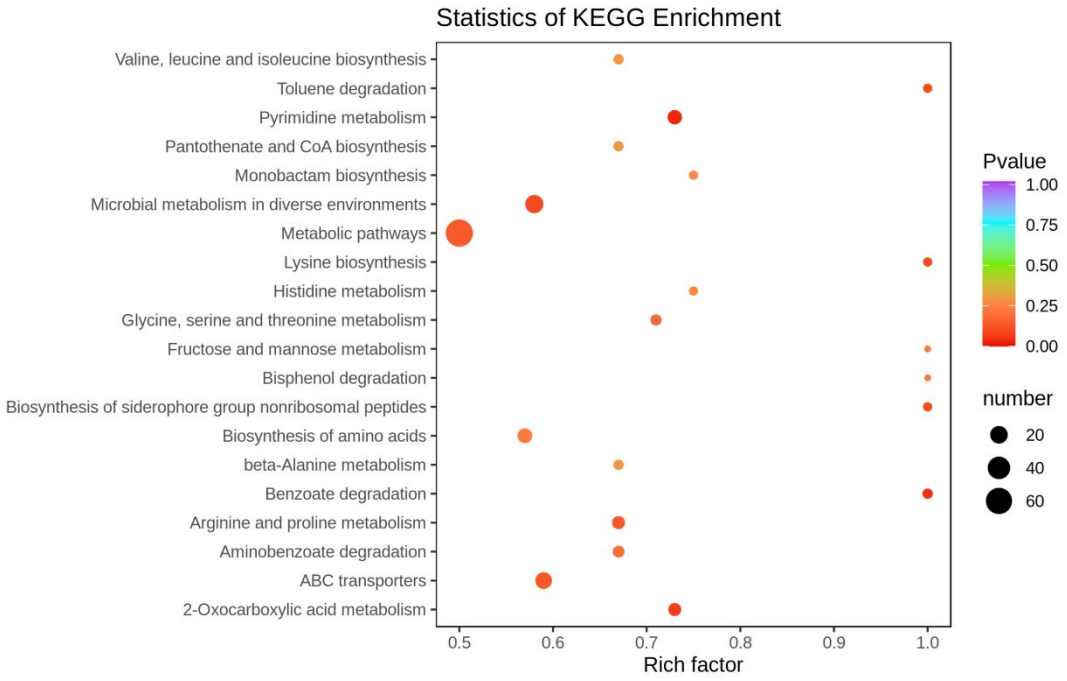
B


Group
1.5
1
0.5
0
-0.5
-1
-1.5
Group
YLs
PU
Class
Flavonoids
Phenolic acids
Amino acids and derivatives
Nucleotides and derivatives
Others
Lipids
Organic acids
Tannins
Alkaloids
Lignans and Coumarins
Terpenoids
Class
YL1
YL2
YL3
PU1
PU2
PU3


Statistics of KEGG Enrichment
Vitamin B6 metabolism
Valine, leucine and isoleucine biosynthesis
Staurosporine biosynthesis
Shigellosis
Pyrimidine metabolism
Purine metabolism
Pertussis
Pentose and glucuronate interconversions
Pantothenate and CoA biosynthesis
Fructose and mannose metabolism
Chagas disease
Caprolactam degradation
Biosynthesis of siderophore group nonribosomal peptides
beta-Alanine metabolism
Benzoate degradation
Arginine and proline metabolism
Amoebiasis
Aminoacyl-tRNA biosynthesis
ABC transporters
2-Oxocarboxylic acid metabolism
0.75
0.80
0.85
0.90
0.95
1.00
Rich factor
Pvalue
1.00
0.75
0.50
0.25
0.00
number
5
10
15
20
25

C


Group
1.5
1
0.5
0
-0.5
-1
-1.5
Group
YLs
LB
Class
Phenolic acids
Flavonoids
Amino acids and derivatives
Nucleotides and derivatives
Others
Organic acids
Tannins
Alkaloids
Lignans and Coumarins
Terpenoids
Lipids
Class
YL1
YL2
YL3
LB1
LB2
LB3


Statistics of KEGG Enrichment
Valine, leucine and isoleucine biosynthesis
Toluene degradation
Pyrimidine metabolism
Pantothenate and CoA biosynthesis
Monobactam biosynthesis
Microbial metabolism in diverse environments
Metabolic pathways
Lysine biosynthesis
Histidine metabolism
Glycine, serine and threonine metabolism
Fructose and mannose metabolism
Bisphenol degradation
Biosynthesis of siderophore group nonribosomal peptides
Biosynthesis of amino acids
beta-Alanine metabolism
Benzoate degradation
Arginine and proline metabolism
Aminobenzoate degradation
ABC transporters
2-Oxocarboxylic acid metabolism
0.5
0.6
0.7
0.8
0.9
1.0
Rich factor
Pvalue
1.00
0.75
0.50
0.25
0.00
number
20
40
60

D

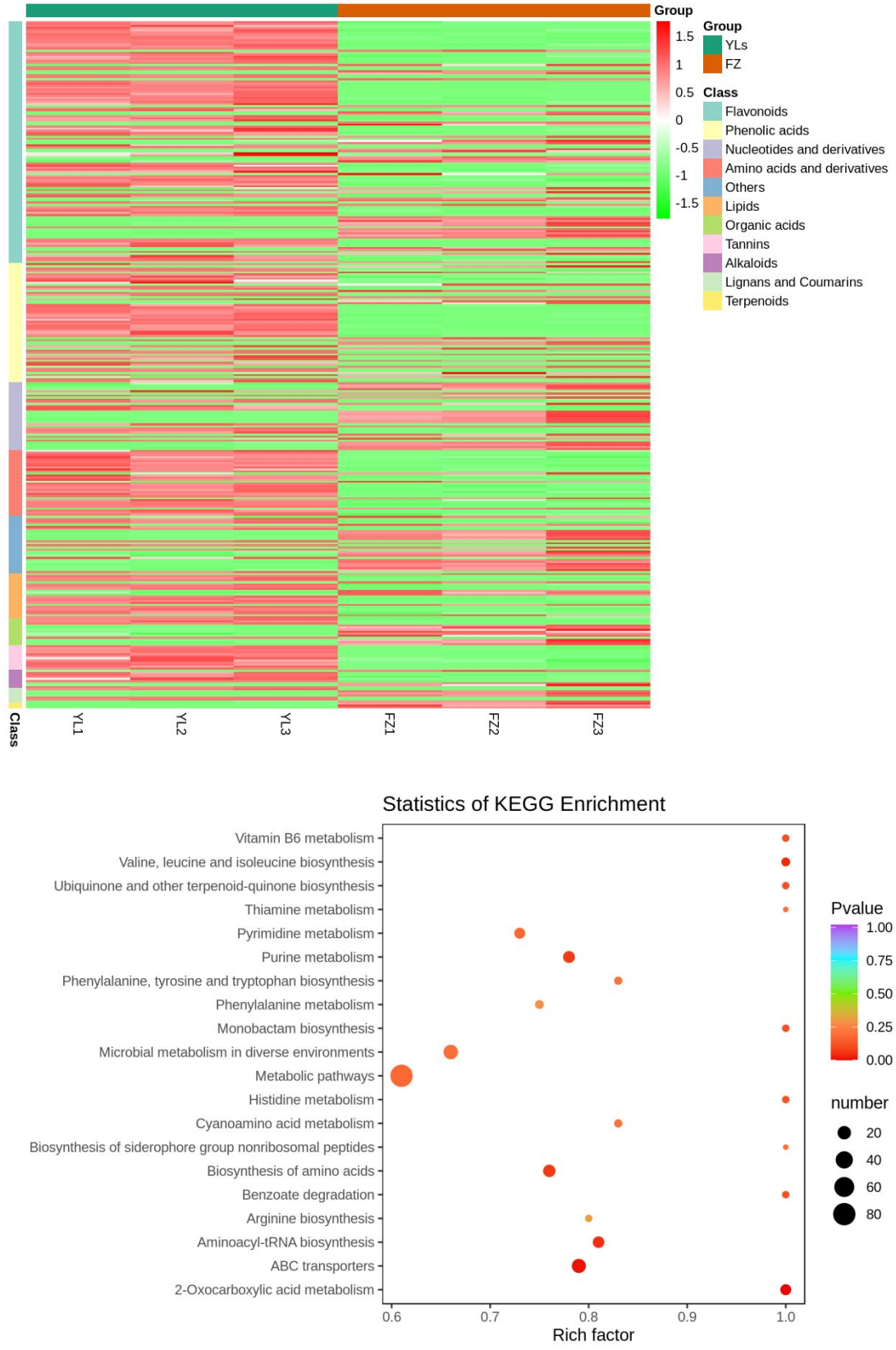
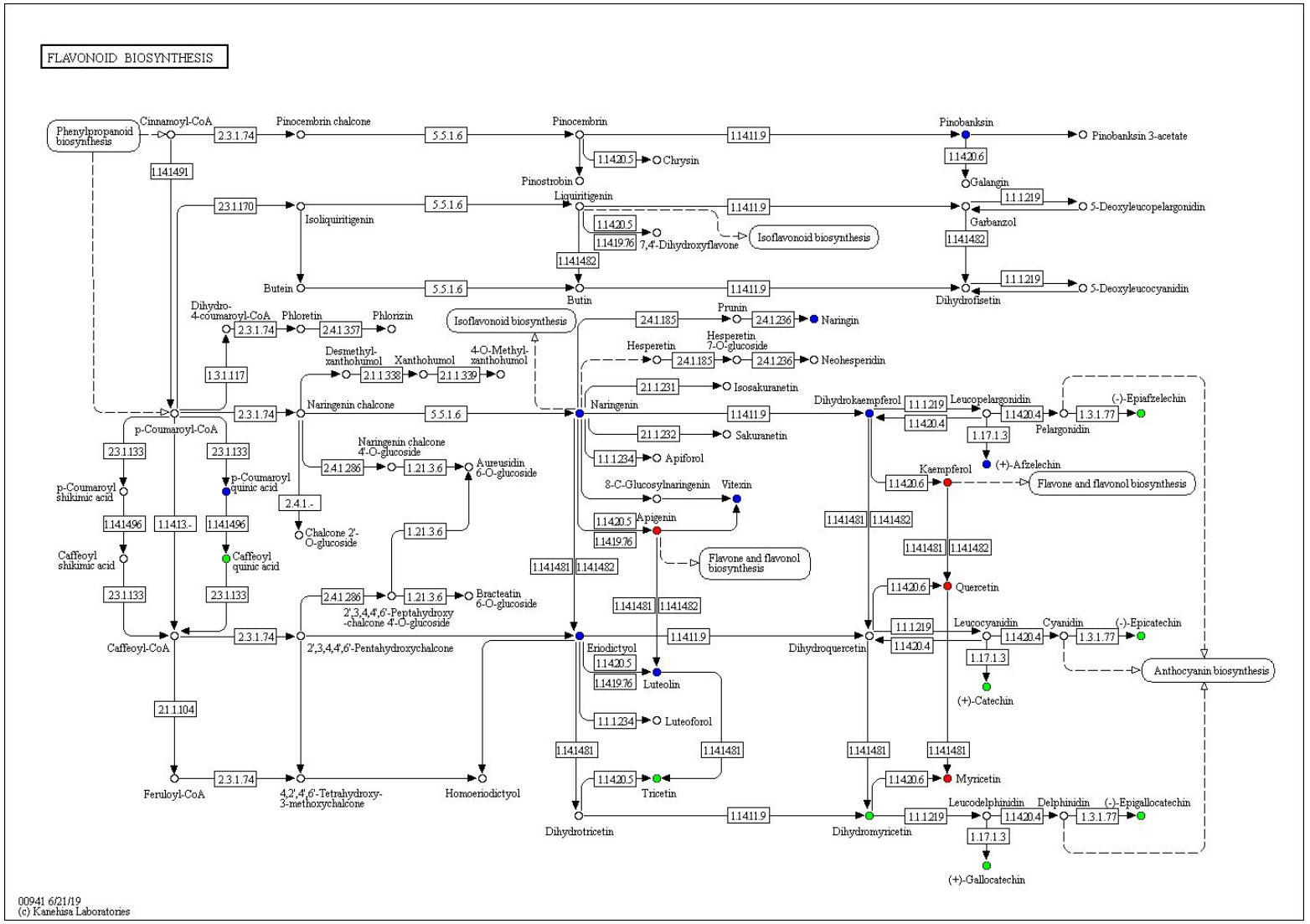
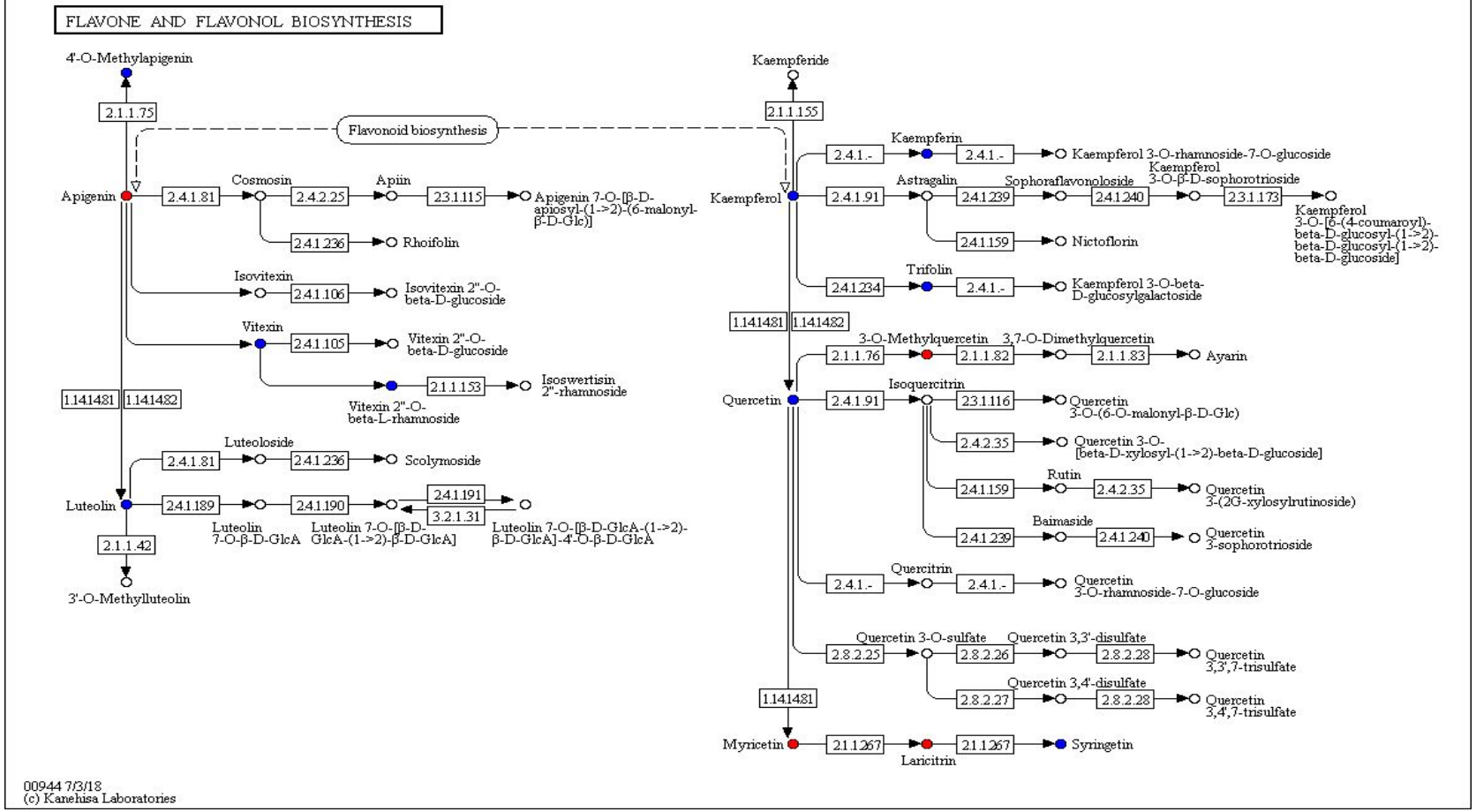


Figure S2. Heatmap of differential metabolites in CDTs compared with YLs respectively, and the KEGG-based enrichment of aforementioned metabolites. A, B, C, D, exhibited heatmap of differential metabolites in YLs_vs_QZ, YLs_vs_PU, YLs_vs_LB, and YLs_vs_FZ, and the responsive KEGG enrichment

A


FLAVONOID BIOSYNTHESIS
Phenylpropanoid biosynthesis
Cinnamoyl-CoA
2.3.1.74
Pinocembrin chalcone
5.5.1.6
Pinocembrin
1.14.11.9
Pinobanksin
Pinobanksin 3-acetate
1.14.20.6
1.14.20.5
Chrysin
1.14.14.91
Pinostrobin
Galangin
2.3.1.170
Isoliquiritigenin
Liquiritigenin
1.1.1.219
5-Deoxyleucopelargonidin
Garbanzol
1.14.19.76
7,4'-Dihydroxyflavone
Isoflavonoid biosynthesis
1.14.14.82
Butein
Butin
Dihydrofisetin
5-Deoxyleucocyanidin
Dihydro-4-coumaroyl-CoA
Phloretin
2.4.1.357
Phlorizin
Prunin
2.4.1.185
2.4.1.236
Naringin
Hesperetin
Hesperetin 7-O-glucoside
Neohesperidin
Desmethyl-xanthohumol
Xanthohumol
4-O-Methyl-xanthohumol
2.1.1.338
2.1.1.339
1.3.1.117
2.1.1.231
Isosakuranetin
Naringenin chalcone
Naringenin
Dihydrokaempferol
Leucopelargonidin
(-)-Epiafzelechin
Pelargonidin
p-Coumaroyl-CoA
2.1.1.232
Sakuranetin
1.17.1.3
2.4.1.286
1.21.3.6
Aureusidin 6-O-glucoside
Naringenin chalcone 4'-O-glucoside
1.1.1.234
Apiforol
Kaempferol
(+)-Afzelechin
p-Coumaroyl shikimic acid
p-Coumaroyl quinic acid
8-C-Glucosylnaringenin
Vitexin
Flavone and flavonol biosynthesis
2.4.1.-
Chalcone 2'-O-glucoside
Apigenin
Caffeoyl shikimic acid
Caffeoyl quinic acid
Flavone and flavonol biosynthesis
Quercetin
Bracteatin 6-O-glucoside
2',3,4,4',6'-Peptahydroxy-chalcone 4'-O-glucoside
Leucocyanidin
Cyanidin
(-)-Epicatechin
Caffeoyl-CoA
2',3,4,4',6'-Pentahydroxychalcone
Eriodictyol
Dihydroquercetin
Anthocyanin biosynthesis
Luteolin
(+)-Catechin
2.1.1.104
Luteoforol
Feruloyl-CoA
4,2',4',6'-Tetrahydroxy-3-methoxychalcone
Homoeriodictyol
Tricetin
Myricetin
Leucodelphinidin
Delphinidin
(-)-Epigallocatechin
Dihydrotricetin
Dihydromyricetin
(+)-Gallocatechin
00941 6/21/19
(c) Kanehisa Laboratories



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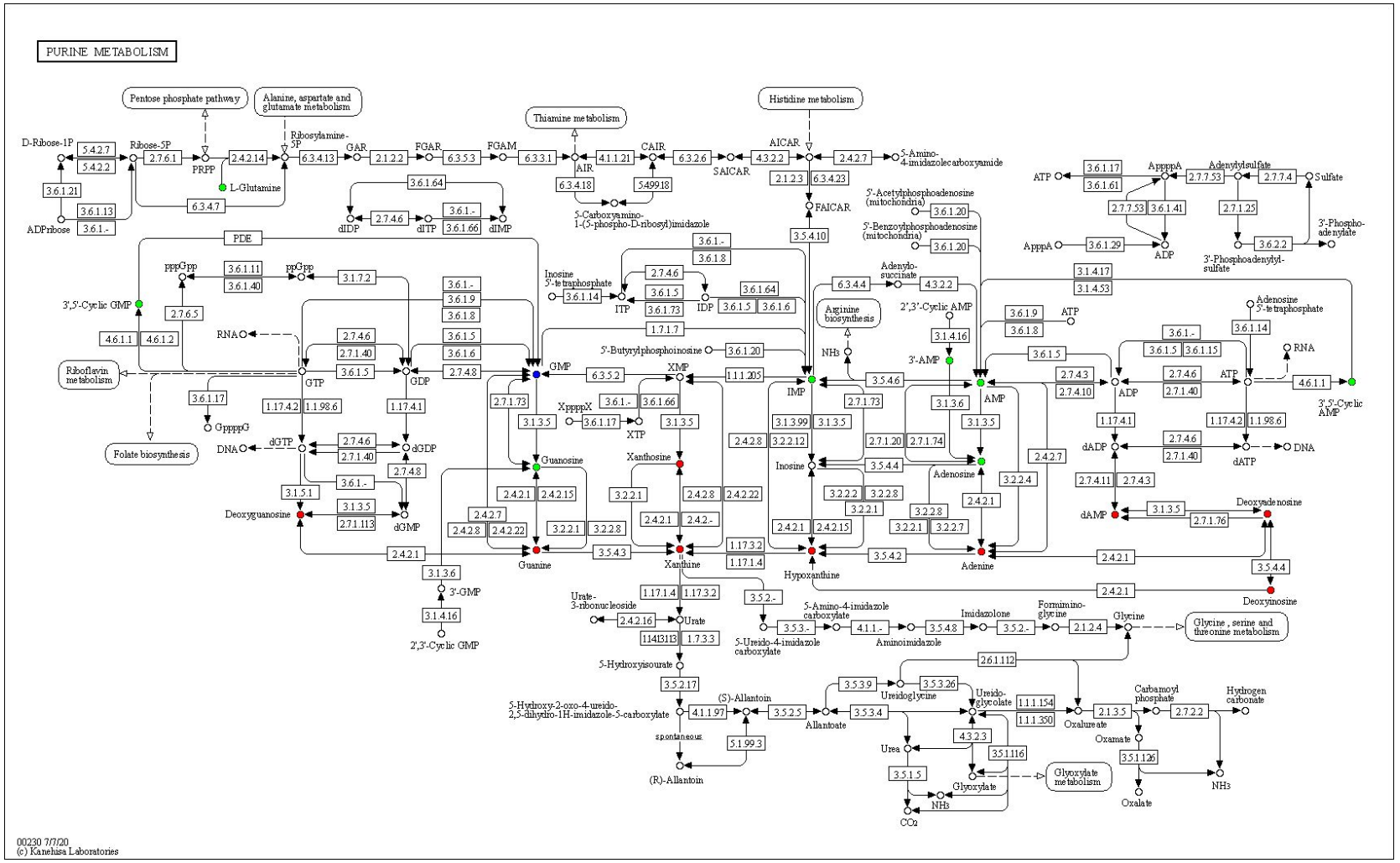
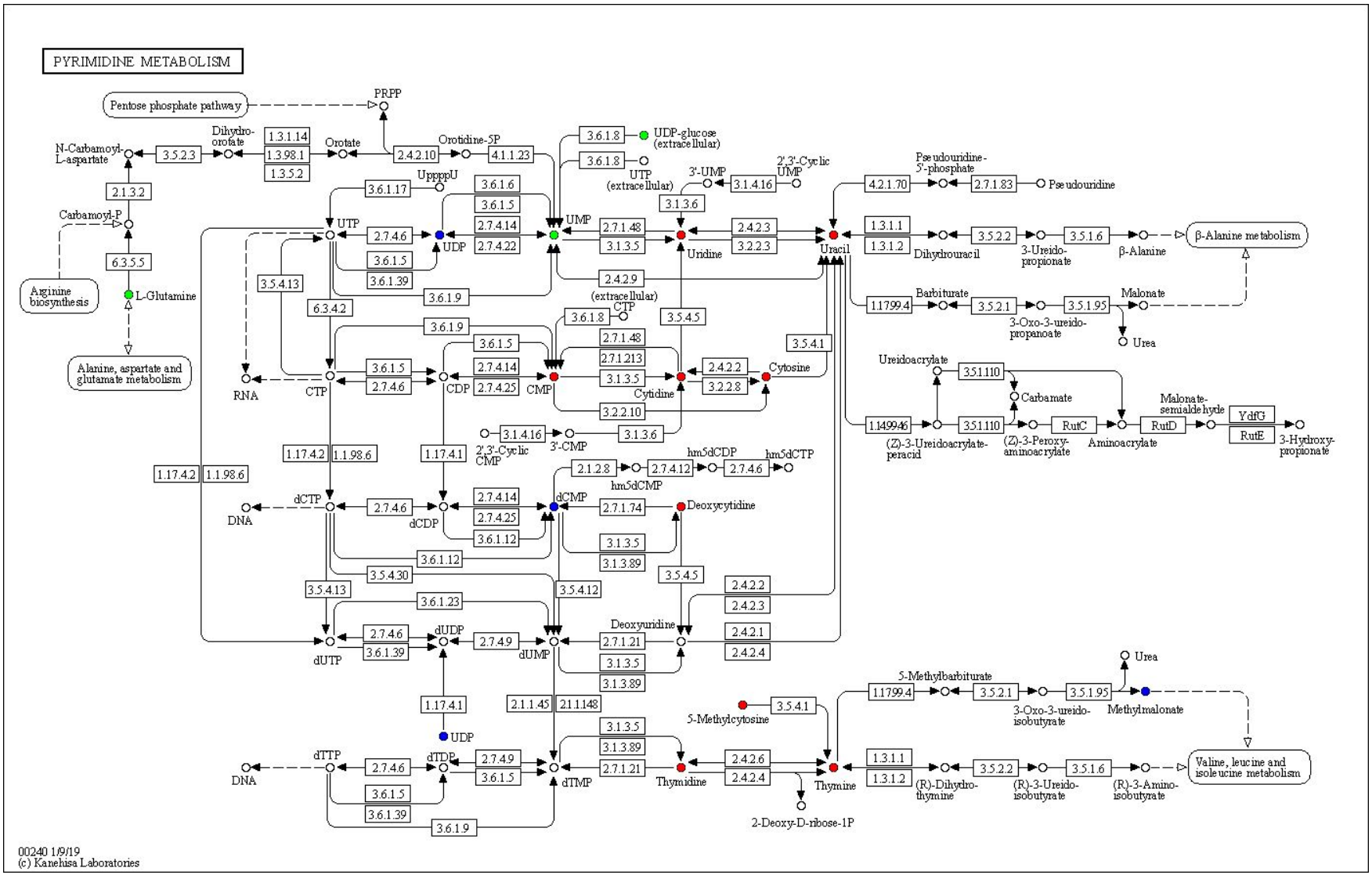
B


PURINE METABOLISM
Pentose phosphate pathway
Alanine, aspartate and glutamate metabolism
Thiamine metabolism
Histidine metabolism
D-Ribose-1P
Ribose-5P
PRPP
L-Glutamine
Ribosylamine-5P
GAR
FGAR
FGAM
AIR
CAIR
SAICAR
AICAR
5-Amino-4-imidazolecarboxyamide
FAICAR
5-Carboxyamino-1-(5-phospho-D-ribosyl)imidazole
ADPribose
dIDP
dITP
dIMP
PDE
pppGpp
ppGpp
3',5'-Cyclic GMP
RNA
Riboflavin metabolism
GTP
GDP
GMP
XMP
IMP
5'-Butyrylphosphoinosine
Inosine 5'-triphosphate
ITP
IDP
Arginine biosynthesis
Adenylo-succinate
2',3'-Cyclic AMP
3'-AMP
NH3
ATP
ADP
AMP
Adenosine 5'-triphosphate
3',5'-Cyclic AMP
GppppG
XpppX
XTP
dGTP
dGDP
dGMP
DNA
dATP
dADP
dAMP
Folate biosynthesis
Guanosine
Xanthosine
Inosine
Adenosine
Deoxyguanosine
Deoxyadenosine
Guanine
Xanthine
Hypoxanthine
Adenine
Deoxyinosine
3'-GMP
2',3'-Cyclic GMP
Urate-3-ribonucleoside
Urate
5-Hydroxyisourate
5-Hydroxy-2-oxo-4-ureido-2,5-dihydro-1H-imidazole-5-carboxylate
(S)-Allantoin
spontaneous
(R)-Allantoin
Allantoate
5-Amino-4-imidazole carboxylate
5-Ureido-4-imidazole carboxylate
Aminoimidazole
Imidazolone
Formimino-glycine
Glycine
Glycine , serine and threonine metabolism
Ureidoglycine
Ureido-glycolate
Oxalureate
Carbamoyl phosphate
Hydrogen carbonate
Urea
Oxamate
Oxalate
Glyoxylate
Glyoxylate metabolism
NH3
CO2
AppppA
Adenylylsulfate
Sulfate
3'-Phospho-adenylate
3'-Phosphoadenylyl-sulfate
5'-Acetylphosphoadenosine (mitochondria)
5'-Benzoylphosphoadenosine (mitochondria)
00230 7/7/20
(c) Kanehisa Laboratories


PYRIMIDINE METABOLISM
Pentose phosphate pathway
PRPP
N-Carbamoyl-L-aspartate
Dihydro-orotate
Orotate
Orotidine-5P
UDP-glucose (extracellular)
UTP (extracellular)
3'-UMP
2',3'-Cyclic UMP
Pseudouridine-5'-phosphate
Pseudouridine
UppppU
Carbamoyl-P
UTP
UDP
UMP
Uridine
Uracil
Arginine biosynthesis
L-Glutamine
Alanine, aspartate and glutamate metabolism
RNA
CTP
CDP
CMP
Cytidine
Cytosine
2',3'-Cyclic CMP
3'-CMP
hm5dCDP
hm5dCTP
hm5dCMP
dCTP
dCDP
dCMP
Deoxycytidine
DNA
dUTP
dUDP
dUMP
Deoxyuridine
dTTP
dTDP
dTMP
Thymidine
Thymine
5-Methylcytosine
2-Deoxy-D-ribose-1P
Dihydrouracil
3-Ureido-propionate
β-Alanine
β-Alanine metabolism
Barbiturate
3-Oxo-3-ureido-propanoate
Malonate
Urea
Ureidoacrylate
Carbamate
(Z)-3-Ureidoacrylate-peracid
(Z)-3-Peroxy-aminoacrylate
Aminoacrylate
Malonate-semialdehyde
3-Hydroxy-propionate
5-Methylbarbiturate
3-Oxo-3-ureido-isobutyrate
Methylmalonate
(R)-Dihydro-thymine
(R)-3-Ureido-isobutyrate
(R)-3-Amino-isobutyrate
Valine, leucine and isoleucine metabolism
00240 1/9/19
(c) Kanehisa Laboratories

C

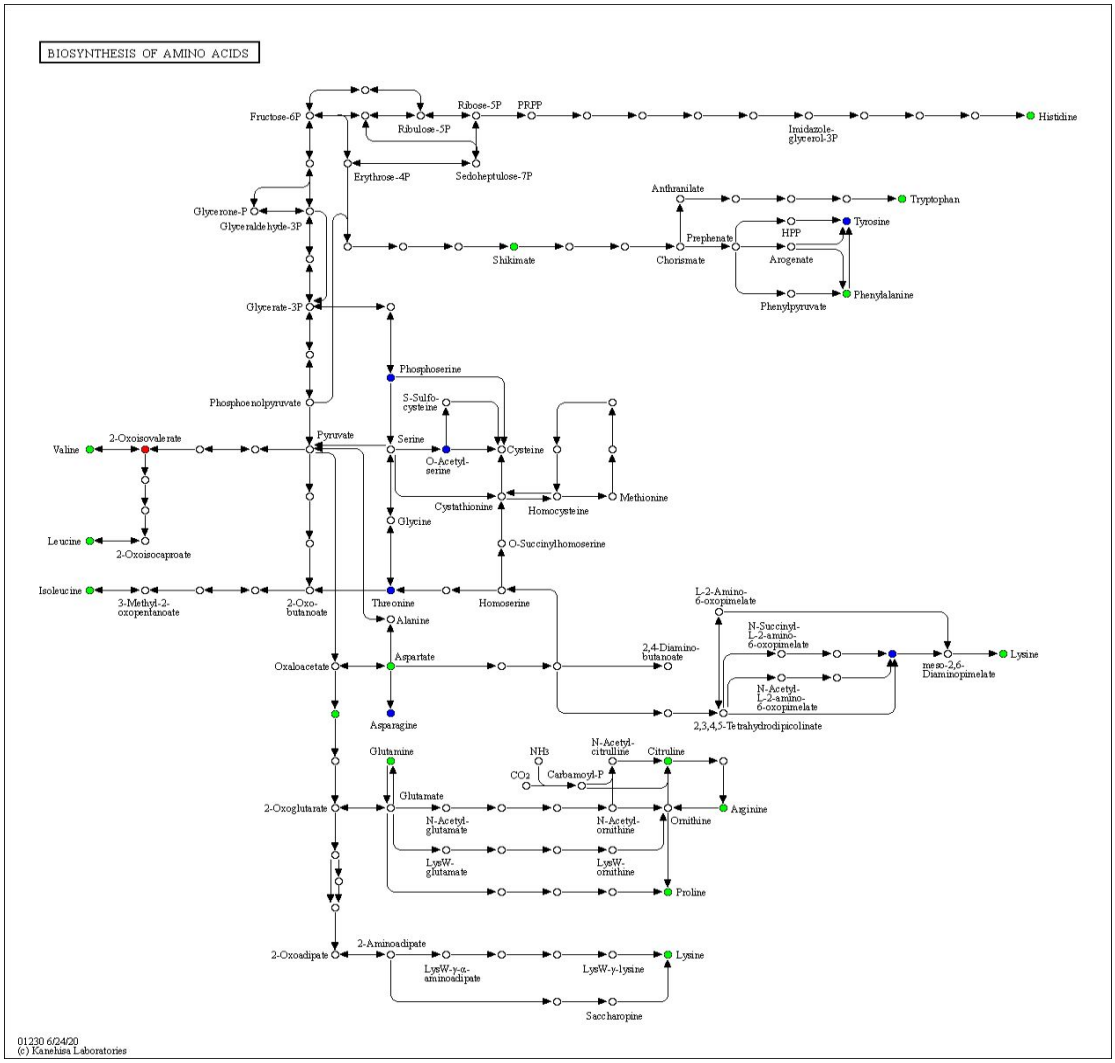


Figure S3. Visualization of differential regulated metabolites in different dark tea (CDTs) compared its raw materials (YLs). (A) Flavonoid biosynthesis pathway referring to KEGG plant and microbial library; (B) Purine and pyrimidine alkaloids metabolism pathway; and (C) Amino acid metabolism pathway.