**Table S8.** The abundance and detailed information of each metabolite, and the correlation along with enhanced cropping years by Spearman’ correlation test.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Metabolites | CCY02 | CCY05 | CCY10 | Correlation with enhanced years | kingdom | super\_class | class | sub\_class |
| [2-hydroxy-5-(7-hydroxy-4-oxo-4H-chromen-2-yl)phenyl]oxidanesulfonic acid | 1.96  | 1.94  | 1.78  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | Flavones |
| 13-Hydroxy-9-methoxy-10-oxo-11-octadecenoic acid | 1.79  | 1.77  | 1.40  | DOWN | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Lineolic acids and derivatives |
| 3-[4-hydroxy-3-(4-hydroxy-3-methylbut-2-en-1-yl)phenyl]prop-2-enoic acid | 1.89  | 1.50  | 1.40  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Cinnamic acids and derivatives | Hydroxycinnamic acids and derivatives |
| 3-Methyl-3-butenyl apiosyl-(1->6)-glucoside | 3.48  | 3.48  | 3.36  | DOWN | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acyl glycosides |
| 4-Hydroxyandrostenedione glucuronide | 1.54  | 1.06  | 0.79  | DOWN | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Cholestane steroids |
| 4-Nitrophenol | 2.55  | 2.41  | 2.35  | DOWN | Organic compounds | Benzenoids | Phenols | Nitrophenols |
| 5-Heptyltetrahydro-2-oxo-3-furancarboxylic acid | 2.57  | 2.28  | 2.23  | DOWN | Organic compounds | Organoheterocyclic compounds | Lactones | Gamma butyrolactones |
| 6-allyl-8b-Carboxy-ergoline | 1.65  | 0.73  | 0.64  | DOWN | Organic compounds | Alkaloids and derivatives | Ergoline and derivatives | Lysergic acids and derivatives |
| 6''-Malonylcosmosiin | 2.92  | 2.66  | 2.63  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | Flavonoid glycosides |
| 6-Methoxyluteolin 7-glucuronide | 2.39  | 2.14  | 2.08  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | Flavonoid glycosides |
| 6-Methoxy-pyridine-3-carboxylic acid | 1.89  | 1.69  | 1.49  | DOWN | Organic compounds | Organoheterocyclic compounds | Pyridines and derivatives | Pyridinecarboxylic acids and derivatives |
| Alpha-CEHC | 2.99  | 2.97  | 2.47  | DOWN | Organic compounds | Organoheterocyclic compounds | Benzopyrans | 1-benzopyrans |
| Artonin Q | 2.06  | 1.60  | 1.46  | DOWN | Organic compounds | Organoheterocyclic compounds | Benzopyrans | 1-benzopyrans |
| Avenestergenin A1 | 2.44  | 2.37  | 2.19  | DOWN | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Triterpenoids |
| Capsicum annuum Fluorescent chlorophyll catabolite | 1.85  | 1.72  | 1.31  | DOWN | Organic compounds | Organoheterocyclic compounds | Tetrapyrroles and derivatives | Not Available |
| Clausarinol | 3.86  | 3.76  | 3.74  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Coumarins and derivatives | Pyranocoumarins |
| Corticrocin | 3.08  | 2.68  | 2.68  | DOWN | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acids and conjugates |
| Cyclopassifloic acid B | 3.02  | 2.92  | 2.53  | DOWN | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Cycloartanols and derivatives |
| Diisobutyl phthalate | 2.19  | 2.10  | 2.00  | DOWN | Organic compounds | Benzenoids | Benzene and substituted derivatives | Benzoic acids and derivatives |
| Elenaic acid | 1.52  | 1.29  | 0.94  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Dicarboxylic acids and derivatives |
| Formylfusarochromanone | 2.17  | 1.94  | 1.63  | DOWN | Organic compounds | Organoheterocyclic compounds | Benzopyrans | 1-benzopyrans |
| Furanofukinin | 2.93  | 2.79  | 2.59  | DOWN | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Sesquiterpenoids |
| Genipinic acid | 3.06  | 2.93  | 2.79  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Dicarboxylic acids and derivatives |
| Genistein 4'-rhamnoside | 1.00  | 0.93  | 0.38  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Isoflavonoids | Isoflavonoid O-glycosides |
| Glaucarubolone 15-O-beta-D-glucopyranoside | 2.52  | 2.41  | 2.30  | DOWN | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene lactones |
| Glutamylvaline | 2.59  | 2.56  | 2.30  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Hematoporphyrin IX | 1.86  | 1.78  | 1.51  | DOWN | Organic compounds | Organoheterocyclic compounds | Tetrapyrroles and derivatives | Porphyrins |
| Isorhamnetin 3-(6''-malonylglucoside) | 1.62  | 1.10  | 1.01  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | Flavonoid glycosides |
| Kamahine C | 2.49  | 2.15  | 1.99  | DOWN | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Ethers |
| L-alpha-Amino-5-oxo-2(5H)-isoxazolepropanoic acid | 1.97  | 1.73  | 1.39  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Methionyl-Proline | 1.57  | 1.53  | 1.31  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Methyl 4-pentenoate | 2.06  | 1.93  | 1.73  | DOWN | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acid esters |
| Monomenthyl succinate | 1.38  | 0.95  | 0.82  | DOWN | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Monoterpenoids |
| Mytilin A | 2.00  | 1.79  | 1.65  | DOWN | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| N-(4-aminobutyl)-3-(4-hydroxyphenyl)propanimidic acid | 1.77  | 1.58  | 1.44  | DOWN | Organic compounds | Benzenoids | Phenols | 1-hydroxy-2-unsubstituted benzenoids |
| Noradrenochrome | 1.79  | 1.70  | 1.61  | DOWN | Organic compounds | Organoheterocyclic compounds | Indoles and derivatives | Not Available |
| Pentigetide | 2.60  | 2.43  | 2.35  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Isoflavonoids | Hydroxyisoflavonoids |
| Phenylpyruvic acid | 1.48  | 1.12  | 1.04  | DOWN | Organic compounds | Benzenoids | Benzene and substituted derivatives | Phenylpyruvic acid derivatives |
| Sphinganine | 4.51  | 4.46  | 4.39  | DOWN | Organic compounds | Organic nitrogen compounds | Organonitrogen compounds | Amines |
| Thioguanosine 5'-diphosphate | 1.79  | 1.53  | 0.67  | DOWN | Organic compounds | Organoheterocyclic compounds | Benzazepines | Dibenzazepines |
| Tridecanol | 3.37  | 3.28  | 3.27  | DOWN | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty alcohols |
| Valtrate | 1.27  | 1.07  | 0.54  | DOWN | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Monoterpenoids |
| Wampetin | 2.27  | 2.01  | 1.90  | DOWN | Organic compounds | Phenylpropanoids and polyketides | Coumarins and derivatives | Furanocoumarins |
| (R)-2-Hydroxysterculic acid | 0.65  | 1.33  | 1.80  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty alcohols |
| (Z)-3-Oxo-2-(2-pentenyl)-1-cyclopenteneacetic acid | 0.54  | 1.14  | 2.25  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbonyl compounds |
| 1-Hydroxy-10-methylacridone | 0.80  | 1.68  | 1.71  | UP | Organic compounds | Organoheterocyclic compounds | Quinolines and derivatives | Benzoquinolines |
| 1-Hydroxy-1-phenyl-3-octadecanone | 1.94  | 2.34  | 2.74  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty alcohols |
| 1-Linoleoylglycerophosphocholine | 4.04  | 4.31  | 4.33  | UP | Organic compounds | Lipids and lipid-like molecules | Glycerophospholipids | Glycerophosphocholines |
| 2-Hydroxy-6-tridecylbenzoic acid | 2.48  | 2.67  | 2.98  | UP | Organic compounds | Benzenoids | Benzene and substituted derivatives | Benzoic acids and derivatives |
| 2-Hydroxyestradiol | 1.36  | 1.80  | 2.07  | UP | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Estrane steroids |
| 2-Nonenoic acid gamma-lactone | 1.03  | 1.53  | 1.61  | UP | Organic compounds | Organoheterocyclic compounds | Dihydrofurans | Furanones |
| 4-(3-hydroxybutyl)-2-methoxyphenol | 1.84  | 2.10  | 2.15  | UP | Organic compounds | Benzenoids | Phenols | Methoxyphenols |
| 4-Deoxyphysalolactone | 1.68  | 1.70  | 1.96  | UP | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Steroid lactones |
| 4-Ethyl-2-hydroxy-3-methyl-2-cyclopenten-1-one | 1.53  | 1.70  | 1.92  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbonyl compounds |
| 4-Ethyl-5-pentyloxazole | 0.65  | 0.73  | 1.67  | UP | Organic compounds | Organoheterocyclic compounds | Azoles | Oxazoles |
| 4-Hydroxy-5-(phenyl)-valeric acid-O-glucuronide | 2.01  | 2.11  | 2.13  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbohydrates and carbohydrate conjugates |
| 4-Hydroxyproline galactoside | 1.45  | 1.77  | 2.13  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acyl glycosides |
| 4-Hydroxyvalsartan | 1.29  | 1.35  | 2.07  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| 4'-O-Methylkanzonol W | 0.46  | 0.57  | 2.10  | UP | Organic compounds | Phenylpropanoids and polyketides | Isoflavonoids | Pyranoisoflavonoids |
| 5-Hexyltetrahydro-2-furanoctanoic acid | 1.76  | 2.27  | 2.33  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Eicosanoids |
| 5-Hydroxyindoleacetic acid | 1.26  | 1.52  | 1.83  | UP | Organic compounds | Organoheterocyclic compounds | Indoles and derivatives | Indolyl carboxylic acids and derivatives |
| 6alpha-Hydroxyphaseollin | 0.56  | 0.77  | 1.77  | UP | Organic compounds | Phenylpropanoids and polyketides | Isoflavonoids | Furanoisoflavonoids |
| 6-Ketomyristic acid | 1.77  | 2.09  | 2.33  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acids and conjugates |
| 8-Ocimenyl acetate | 0.18  | 0.57  | 2.50  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty alcohol esters |
| 9(S)-HPODE | 2.09  | 2.46  | 2.84  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Lineolic acids and derivatives |
| Abscisic alcohol 11-glucoside | 0.63  | 1.01  | 1.92  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene glycosides |
| Adenine | 1.68  | 2.03  | 2.43  | UP | Organic compounds | Organoheterocyclic compounds | Imidazopyrimidines | Purines and purine derivatives |
| Alanyltryptophan | 2.03  | 2.53  | 2.62  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Anhydrocinnzeylanol | 0.87  | 0.90  | 2.25  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene lactones |
| Arginyl-Histidine | 2.56  | 2.83  | 3.01  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Artabsinolide D | 0.86  | 1.24  | 1.58  | UP | Organic compounds | Organoheterocyclic compounds | Lactones | Gamma butyrolactones |
| Auberganol | 1.55  | 1.96  | 2.10  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Sesquiterpenoids |
| Avenoleic acid | 1.78  | 2.02  | 2.28  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Lineolic acids and derivatives |
| Bakkenolide B | 2.57  | 2.65  | 2.81  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene lactones |
| Capsianoside I | 0.44  | 0.49  | 2.31  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acyl glycosides |
| Celastrol | 2.54  | 2.76  | 2.83  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Triterpenoids |
| Choline | 2.38  | 2.45  | 2.74  | UP | Organic compounds | Organic nitrogen compounds | Organonitrogen compounds | Quaternary ammonium salts |
| Chrysaloin | 1.85  | 1.86  | 2.04  | UP | Organic compounds | Benzenoids | Anthracenes | Not Available |
| Cibaric acid | 0.94  | 1.27  | 1.70  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Lineolic acids and derivatives |
| Cinnamoside | 1.79  | 2.28  | 2.38  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene glycosides |
| Copalic acid | 1.89  | 2.40  | 2.72  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Diterpenoids |
| Creatinine | 0.00  | 0.00  | 0.00  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Cucurbitacin C | 1.64  | 2.14  | 2.25  | UP | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Cucurbitacins |
| Cytosine | 1.40  | 1.90  | 2.08  | UP | Organic compounds | Organoheterocyclic compounds | Diazines | Pyrimidines and pyrimidine derivatives |
| Deoxycytidine | 1.58  | 1.72  | 1.89  | UP | Organic compounds | Nucleosides, nucleotides, and analogues | Pyrimidine nucleosides | Pyrimidine 2'-deoxyribonucleosides |
| Deoxyinosine | 1.56  | 2.12  | 2.18  | UP | Organic compounds | Nucleosides, nucleotides, and analogues | Purine nucleosides | Purine 2'-deoxyribonucleosides |
| Diosbulbinoside F | 1.80  | 1.95  | 2.38  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Terpene glycosides |
| D-Pipecolic acid | 2.09  | 2.27  | 2.66  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Estradiol | 0.93  | 1.19  | 1.72  | UP | Organic compounds | Lipids and lipid-like molecules | Steroids and steroid derivatives | Estrane steroids |
| Gravacridonediol | 0.36  | 1.24  | 1.60  | UP | Organic compounds | Organoheterocyclic compounds | Quinolines and derivatives | Benzoquinolines |
| Guanine | 1.34  | 1.69  | 2.09  | UP | Organic compounds | Organoheterocyclic compounds | Imidazopyrimidines | Purines and purine derivatives |
| Histidinyl-Arginine | 1.55  | 2.14  | 2.34  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Hydroxyprolyl-Lysine | 1.37  | 1.41  | 1.62  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Hypoxanthine | 1.46  | 1.94  | 2.07  | UP | Organic compounds | Organoheterocyclic compounds | Imidazopyrimidines | Purines and purine derivatives |
| Indole-3-carboxylic acid | 1.43  | 1.69  | 1.69  | UP | Organic compounds | Organoheterocyclic compounds | Indoles and derivatives | Indolecarboxylic acids and derivatives |
| Isoliensinine | 0.85  | 0.96  | 2.14  | UP | Organic compounds | Organoheterocyclic compounds | Isoquinolines and derivatives | Benzylisoquinolines |
| Isoputreanine | 0.53  | 0.59  | 0.62  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Kukoamine B | 1.79  | 1.83  | 2.10  | UP | Organic compounds | Benzenoids | Phenols | Benzenediols |
| Leukotriene E3 | 2.02  | 2.19  | 2.37  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Eicosanoids |
| Linoleoyl Ethanolamide | 3.09  | 3.41  | 3.58  | UP | Organic compounds | Organic nitrogen compounds | Organonitrogen compounds | Amines |
| L-Phenylalanine | 2.45  | 2.87  | 3.12  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| L-trans-alpha-Amino-2-carboxycyclopropaneacetic acid | 2.40  | 2.55  | 2.79  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| L-Tryptophan | 1.65  | 2.01  | 2.15  | UP | Organic compounds | Organoheterocyclic compounds | Indoles and derivatives | Indolyl carboxylic acids and derivatives |
| Lucidenic acid M | 2.21  | 2.49  | 2.59  | UP | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | O-methylated flavonoids |
| LysoPC(15:0) | 3.61  | 3.77  | 3.86  | UP | Organic compounds | Lipids and lipid-like molecules | Glycerophospholipids | Glycerophosphocholines |
| LysoPC(18:1(11Z)) | 4.41  | 4.57  | 4.71  | UP | Organic compounds | Lipids and lipid-like molecules | Glycerophospholipids | Glycerophosphocholines |
| Mangalkanyl glucoside | 1.71  | 1.73  | 2.16  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbohydrates and carbohydrate conjugates |
| Methyl helianthenoate F glucoside | 1.17  | 1.43  | 2.01  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acyl glycosides |
| MG(0:0/18:1(9Z)/0:0) | 3.04  | 3.15  | 3.35  | UP | Organic compounds | Lipids and lipid-like molecules | Glycerolipids | Monoradylglycerols |
| MG(16:1(9Z)/0:0/0:0) | 3.48  | 3.54  | 4.05  | UP | Organic compounds | Lipids and lipid-like molecules | Glycerolipids | Monoradylglycerols |
| N-(1-Deoxy-b-D-fructopyranosyl) (R)C(S)S-alliin | 1.34  | 2.09  | 2.87  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| N-Benzoylanthranilic acid | 1.16  | 1.26  | 2.25  | UP | Organic compounds | Benzenoids | Benzene and substituted derivatives | Benzoic acids and derivatives |
| Niazirinin | 0.30  | 0.41  | 1.87  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbohydrates and carbohydrate conjugates |
| N-lactoyl-Isoeucine | 2.25  | 2.41  | 2.75  | UP | Organic compounds | Organic acids and derivatives | Keto acids and derivatives | Medium-chain keto acids and derivatives |
| Peonidin 3-rhamnoside | 0.38  | 1.54  | 1.62  | UP | Organic compounds | Phenylpropanoids and polyketides | Flavonoids | Flavonoid glycosides |
| Pimelic acid | 1.62  | 1.99  | 2.15  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acids and conjugates |
| Prostaglandin F3a | 1.92  | 2.16  | 2.43  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Eicosanoids |
| Prunasin | 1.19  | 2.04  | 2.07  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbohydrates and carbohydrate conjugates |
| Ricinoleic acid | 1.79  | 2.68  | 2.79  | UP | Organic compounds | Lipids and lipid-like molecules | Fatty Acyls | Fatty acids and conjugates |
| Sagittariol | 1.85  | 1.99  | 2.57  | UP | Organic compounds | Lipids and lipid-like molecules | Prenol lipids | Diterpenoids |
| Serylglutamic acid | 1.10  | 2.02  | 2.06  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Small bacteriocin | 1.28  | 2.12  | 2.28  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Valyl-Proline | 1.85  | 1.95  | 2.13  | UP | Organic compounds | Organic acids and derivatives | Carboxylic acids and derivatives | Amino acids, peptides, and analogues |
| Vanillin | 1.17  | 1.21  | 1.43  | UP | Organic compounds | Benzenoids | Phenols | Methoxyphenols |
| Xi-4-Hydroxy-4-methyl-2-cyclohexen-1-one | 0.73  | 0.95  | 1.28  | UP | Organic compounds | Organic oxygen compounds | Organooxygen compounds | Carbonyl compounds |
| Yuzu lactone | 0.59  | 1.20  | 2.71  | UP | Organic compounds | Phenylpropanoids and polyketides | Macrolides and analogues | Not Available |