Supplementary

Analytical Strategies for LC-MS Based Untargeted and Targeted Metabolomics Approaches Reveal the Entomological Origins of Honey

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Abstract

A comprehensive LC-MS-based metabolomics approach was developed to discriminate honey harvested from *Apis mellifera ligustica* Spinola (*A. mellifera*) and *Apis cerana cerana* Fabricius (*A. cerana*). Based on an untargeted strategy, ultrahigh-performance liquid chromatography electrospray ionization quadrupole orbitrap high-resolution mass spectrometry (UPLC Q-Orbitrap) was combined with chemometrics techniques to screen and identify tentative markers from *A. mellifera* and *A. cerana* honey. In targeted metabolomics analysis, a sensitive method of solid-phase extraction followed by ultrahigh-performance liquid chromatography coupled with triple quadrupole tandem mass spectrometry (UPLC-MS/MS) was established for quantifying the three markers, and the results showed that 3-amino-2-naphthoic acid and methyl indole-3-acetate could be considered markers of *A. cerana* honey, as they were present in higher amounts in *A. cerana* honey than in *A. mellifera* honey, whereas kynurenic acid was determined to be a marker of *A. mellifera* honey. This work highlights critical information for the authentication of *A. cerana* and *A. mellifera* honey.

Key words: honey, *A. mellifera* and *A. cerana*, untargeted and targeted discrimination, UPLC Q-Orbitrap, UPLC-MS/MS

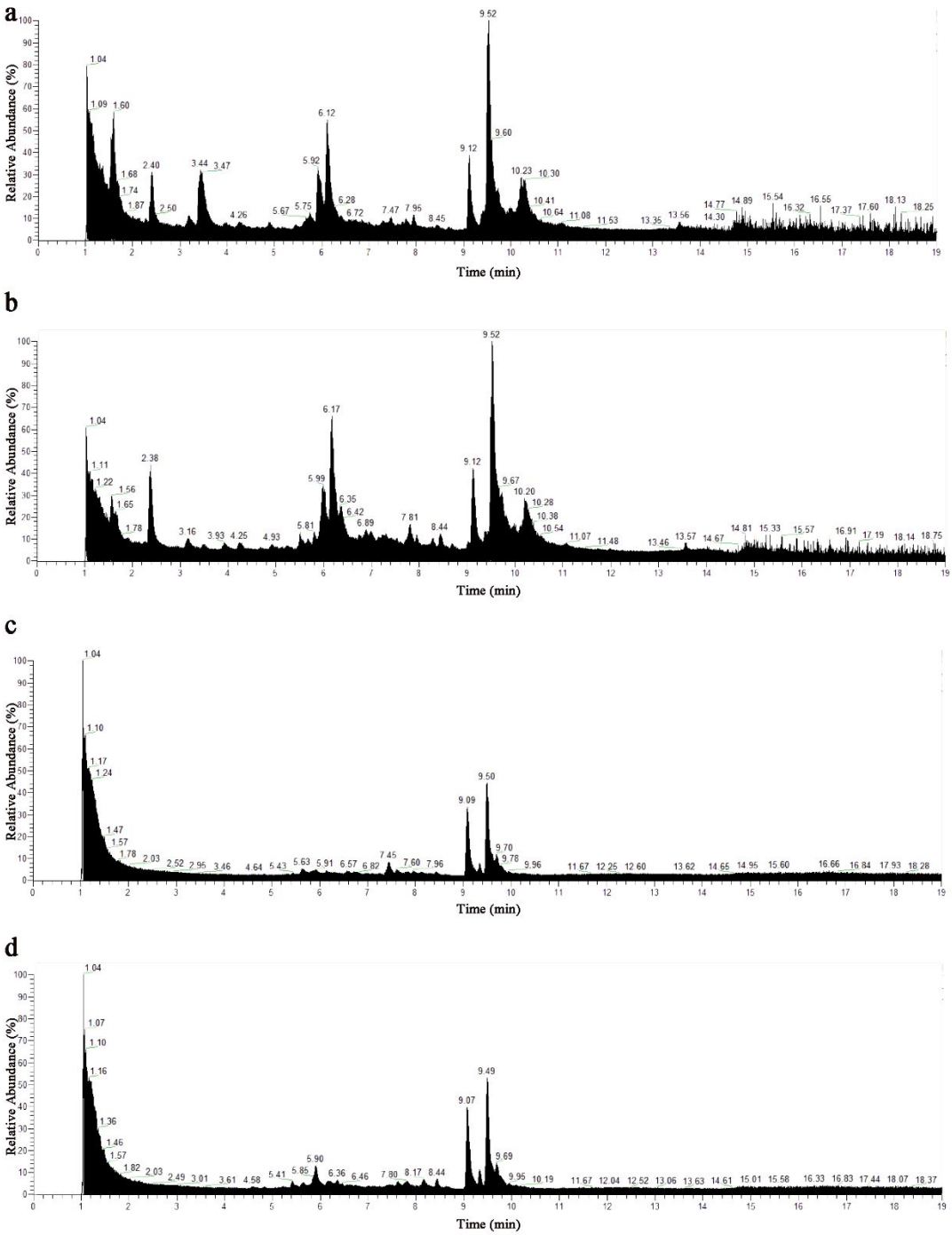

Figure S1: The TIC of honey for data acquisition by UHPLC/ESI Q-Orbitrap. a: The TIC of *Apis cerana cerana* Fabricius honey in positive ion; b: The TIC of *Apis mellifera ligustica* Spinola honey in positive ion; c: The TIC of *Apis cerana cerana* Fabricius honey in nagative ion; d: The TIC of *Apis mellifera ligustica* Spinola honey in nagative ion.

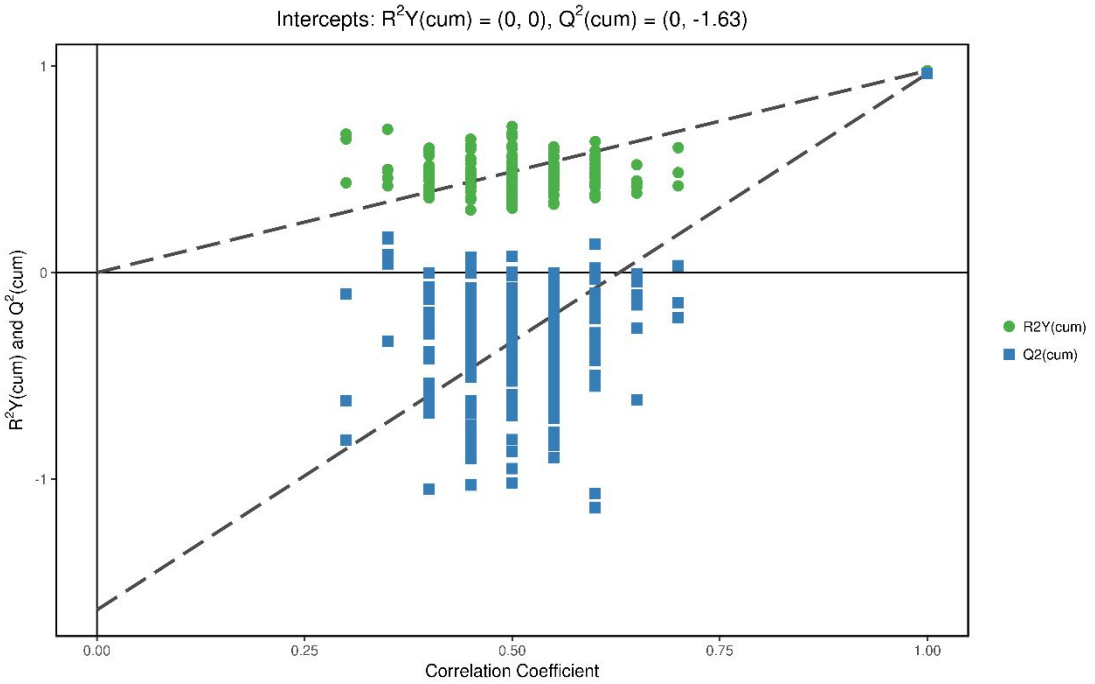

Figure S2: The validation for OPLS-DA model by 200 permutations.

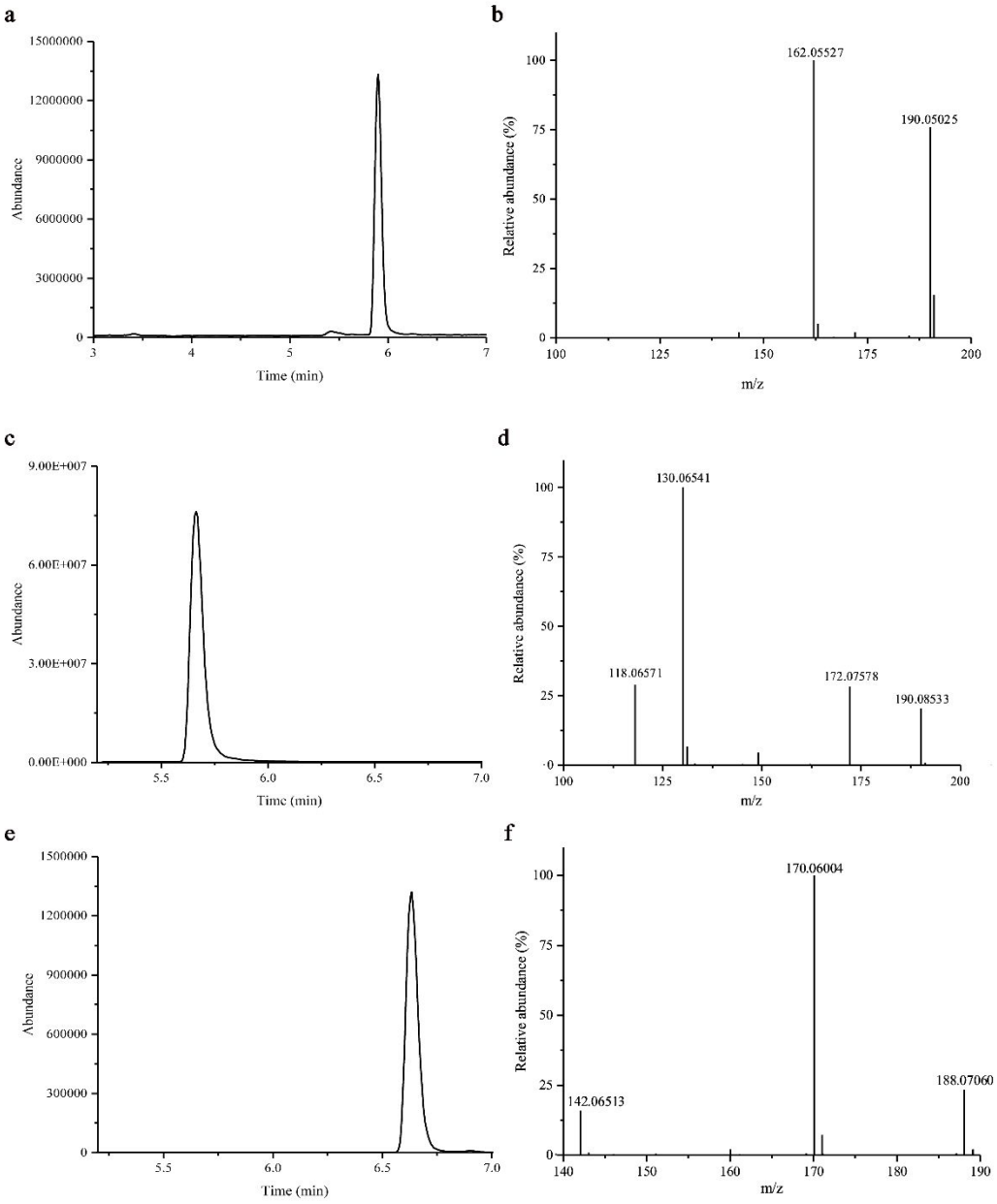

Figure S3: The extracted ion chromatogram and MS^2 spectra of standard substances of three markers. a: Extracted ion chromatogram of kynurenic acid; b: MS^2 spectra of kynurenic acid; c: Extracted ion chromatogram of methyl indole-3-acetate; d: MS^2 spectra of methyl indole-3-acetate; e: Extracted ion chromatogram of 3-amino-2-naphthoic acid; f: MS^2 spectra of 3-amino-2-naphthoic acid.

Table S1 The information of lychee honey samples.

Sample	Nectar plants	Entomological origins	Province	Geographical regions	Geographical coordinates	
LZ-01	lychee	*Apis cerana*	Guangdong	Southern China	22°60N	113°94E
LZ-02	lychee	*Apis cerana*	Guangdong	Southern China	22°60N	113°94E
LZ-03	lychee	*Apis cerana*	Guangdong	Southern China	22°62N	113°98E
LZ-04	lychee	*Apis cerana*	Guangdong	Southern China	22°62N	113°98E
LZ-05	lychee	*Apis cerana*	Guangdong	Southern China	22°51N	113°91E
LZ-06	lychee	*Apis cerana*	Guangdong	Southern China	22°51N	113°91E
LZ-07	lychee	*Apis cerana*	Guangdong	Southern China	22°51N	113°91E
LZ-08	lychee	*Apis cerana*	Guangdong	Southern China	22°90N	113°88E
LZ-09	lychee	*Apis cerana*	Guangdong	Southern China	22°94N	113°87E
LZ-10	lychee	*Apis cerana*	Guangdong	Southern China	22°98N	113°99E
LZ-11	lychee	*Apis cerana*	Guangdong	Southern China	22°98N	113°99E
LZ-12	lychee	*Apis cerana*	Guangdong	Southern China	22°88N	113°78E
LZ-13	lychee	*Apis cerana*	Guangdong	Southern China	22°98N	113°99E
LZ-14	lychee	*Apis cerana*	Guangdong	Southern China	21°85N	110°96E
LZ-15	lychee	*Apis cerana*	Guangdong	Southern China	21°85N	110°96E
LZ-16	lychee	*Apis cerana*	Guangdong	Southern China	21°85N	110°96E
LZ-17	lychee	*Apis cerana*	Guangdong	Southern China	21°85N	110°96E
LZ-18	lychee	*Apis cerana*	Guangdong	Southern China	21°85N	110°96E
LZ-19	lychee	*Apis cerana*	Guangdong	Southern China	21°73N	111°38E
LZ-20	lychee	*Apis cerana*	Guangdong	Southern China	21°73N	111°38E
LZ-21	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-22	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-23	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E

LZ-24	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-25	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-26	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-27	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-28	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-29	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-30	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-31	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-32	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°94E
LZ-33	lychee	*Apis melifera*	Guangdong	Southern China	22°60N	113°98E
LZ-34	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-35	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-36	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-37	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-38	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-39	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E
LZ-40	lychee	*Apis melifera*	Guangdong	Southern China	22°62N	113°98E

Table S2 The detailed information of *A. cerana* honey and *A. mellifera* honey samples.

Sample	Nectar plants	Entomological origins	Province	Geographical regions	Geographical coordinates	
BH-16	wildflower	*Apis cerana*	Anhui	Eastern China	30°43N	116°27E
LZ-56	lychee	*Apis melifera*	Fujian	Southern China	24°89N	118°61E
BH-01	wildflower	*Apis cerana*	Gansu	Northwestern China	33°45N	106°27E
BH-02	wildflower	*Apis cerana*	Gansu	Northwestern China	33°99N	105°55E
BH-03	wildflower	*Apis cerana*	Gansu	Northwestern China	34°19N	105°18E
BH-04	wildflower	*Apis cerana*	Gansu	Northwestern China	33°78N	106°31E
BH-05	wildflower	*Apis cerana*	Gansu	Northwestern China	33°29N	104°87E
BH-06	wildflower	*Apis cerana*	Gansu	Northwestern China	33°91N	106°31E
BH-07	wildflower	*Apis cerana*	Gansu	Northwestern China	33°50N	105°144E
BH-08	wildflower	*Apis cerana*	Gansu	Northwestern China	33°85N	105°68E
BH-09	wildflower	*Apis cerana*	Gansu	Northwestern China	33°01N	105°77E
BH-10	wildflower	*Apis cerana*	Gansu	Northwestern China	34°03N	104°36E
BH-11	wildflower	*Apis cerana*	Gansu	Northwestern China	32°77N	105°35E
BH-12	wildflower	*Apis cerana*	Gansu	Northwestern China	33°89N	104°70E
BH-13	wildflower	*Apis cerana*	Gansu	Northwestern China	33°63N	106°22E
BH-14	wildflower	*Apis cerana*	Gansu	Northwestern China	33°84N	105°60E
BH-15	wildflower	*Apis cerana*	Gansu	Northwestern China	33°49N	105°02E
BH-17	wildflower	*Apis cerana*	Gansu	Northwestern China	34°07N	106°29E
BH-18	wildflower	*Apis cerana*	Gansu	Northwestern China	33°97N	105°87E
BH-19	wildflower	*Apis cerana*	Gansu	Northwestern China	34°18N	106°50E
WJ-6	sapium	*Apis cerana*	Guangdong	Southern China	23°54N	114°39E
LZ-42	longan	*Apis melifera*	Guangdong	Southern China	23°54N	114°39E
LZ-51	longan	*Apis melifera*	Guangdong	Southern China	23°66N	114°35E
LZ-55	longan	*Apis melifera*	Guangdong	Southern China	23°66N	114°36E

WJ-3	sapium	*Apis melifera*	Guangdong	Southern China	23°66N	114°35E
LZ-60	lychee	*Apis cerana*	Guangxi	Southern China	22°82N	108°37E
LZ-61	lychee	*Apis cerana*	Guangxi	Southern China	22°82N	108°37E
LZ-62	lychee	*Apis cerana*	Guangxi	Southern China	22°82N	108°37E
LZ-59	lychee	*Apis cerana*	Guangxi	Southern China	22°82N	108°37E
LZ-57	lychee	*Apis melifera*	Guangxi	Southern China	22°82N	108°37E
LZ-58	lychee	*Apis melifera*	Guangxi	Southern China	22°82N	108°37E
LZ-89	longan	*Apis melifera*	Guangxi	Southern China	22°82N	108°37E
WJ-2	sapium	*Apis melifera*	Guangxi	Southern China	22°82N	108°37E
TS01	rape	*Apis melifera*	Hebei	Northern China	40°01N	118°01E
TS02	Acacia	*Apis melifera*	Hebei	Northern China	40°01N	118°01E
TS03	chaste	*Apis melifera*	Hebei	Northern China	40°01N	118°01E
TS04	jujube	*Apis melifera*	Hebei	Northern China	40°01N	118°03E
TS12	chaste	*Apis melifera*	Hebei	Northern China	40°22N	118°21E
TS22	jujube	*Apis melifera*	Hebei	Northern China	39°69N	118°2E
TS27	Acacia	*Apis melifera*	Hebei	Northern China	40°77N	118°17E
TS28	jujube	*Apis melifera*	Hebei	Northern China	40°77N	118°17E
TS31	jujube	*Apis melifera*	Hebei	Northern China	40°83N	118°05E
TS78	chaste	*Apis melifera*	Hebei	Northern China	40°83N	117°85E
TS80	chaste	*Apis melifera*	Hebei	Northern China	39°76N	119°22E
HLJ101	linden	*Apis melifera*	Heilongjiang	Northeast China	45°73N	128°19E
HLJ105	linden	*Apis melifera*	Heilongjiang	Northeast China	45°81N	128°22E
HLJ111	linden	*Apis melifera*	Heilongjiang	Northeast China	45°84N	128°84E
HLJ115	linden	*Apis melifera*	Heilongjiang	Northeast China	45°60N	128°99E
HLJ125	linden	*Apis melifera*	Heilongjiang	Northeast China	46°08N	128°66E
HLJ138	linden	*Apis melifera*	Heilongjiang	Northeast China	46°25N	128°58E

HLJ149	linden	*Apis melifera*	Heilongjiang	Northeast China	46°59N	129°26E
HN01	wildflower	*Apis cerana*	Henan	Central China	33°03N	112°13E
HN02	wildflower	*Apis cerana*	Henan	Central China	33°26N	113°01E
HN05	wildflower	*Apis cerana*	Henan	Central China	32°69N	112°08E
HN09	wildflower	*Apis cerana*	Henan	Central China	33°31N	111°48E
HN10	wildflower	*Apis cerana*	Henan	Central China	33°26N	113°01E
HN11	wildflower	*Apis cerana*	Henan	Central China	32°68N	112°81E
HN12	wildflower	*Apis cerana*	Henan	Central China	33°22N	112°53E
HN15	wildflower	*Apis cerana*	Henan	Central China	34°05N	111°07E
FX2017-023	Acacia	*Apis melifera*	Henan	Central China	34°75N	113°63E
HN17	jujube	*Apis melifera*	Henan	Central China	34°05N	111°07E
ES01	wildflower	*Apis cerana*	Hubei	Central China	31°05N	110°33E
ES03	wildflower	*Apis cerana*	Hubei	Central China	31°31N	110°64E
ES05	wildflower	*Apis cerana*	Hubei	Central China	31°31N	110°68E
ES06	wildflower	*Apis cerana*	Hubei	Central China	30°49N	111°17E
ES08	wildflower	*Apis cerana*	Hubei	Central China	30°11N	111°25E
ES09	wildflower	*Apis cerana*	Hubei	Central China	29°73N	109°50E
ES10	wildflower	*Apis cerana*	Hubei	Central China	30°69N	109°76E
ES12	wildflower	*Apis cerana*	Hubei	Central China	31°20N	110°23E
ES14	wildflower	*Apis cerana*	Hubei	Central China	30°43N	108°69E
ES18	wildflower	*Apis cerana*	Hubei	Central China	31°31N	110°38E
ES21	wildflower	*Apis cerana*	Hubei	Central China	31°05N	110°33E
ES24	wildflower	*Apis cerana*	Hubei	Central China	30°49N	111°17E
ES25	wildflower	*Apis cerana*	Hubei	Central China	30°50N	111°17E
ES26	wildflower	*Apis cerana*	Hubei	Central China	29°88N	110°03E
ES27	wildflower	*Apis cerana*	Hubei	Central China	30°30N	108°97E

ES32	wildflower	*Apis cerana*	Hubei	Central China	31°31N	110°68E
ES38	wildflower	*Apis cerana*	Hubei	Central China	31°35N	110°64E
ES39	wildflower	*Apis cerana*	Hubei	Central China	30°50N	111°16E
ES40	wildflower	*Apis cerana*	Hubei	Central China	30°43N	111°22E
ES42	wildflower	*Apis cerana*	Hubei	Central China	30°43N	111°22E
ES44	wildflower	*Apis cerana*	Hubei	Central China	30°36N	111°33E
ES45	wildflower	*Apis cerana*	Hubei	Central China	30°22N	111°22E
ES46	wildflower	*Apis cerana*	Hubei	Central China	30°17N	111°08E
ES50	wildflower	*Apis cerana*	Hubei	Central China	30°03N	110°44E
ES52	wildflower	*Apis cerana*	Hubei	Central China	30°00N	110°25E
ES53	wildflower	*Apis cerana*	Hubei	Central China	29°88N	110°03E
ES56	wildflower	*Apis cerana*	Hubei	Central China	29°82N	109°54E
ES57	wildflower	*Apis cerana*	Hubei	Central China	29°82N	109°54E
ES58	wildflower	*Apis cerana*	Hubei	Central China	29°73N	109°5E
ES59	wildflower	*Apis cerana*	Hubei	Central China	30°10N	109°45E
ES60	wildflower	*Apis cerana*	Hubei	Central China	30°12N	109°4E
ES61	wildflower	*Apis cerana*	Hubei	Central China	30°70N	109°78E
ES65	wildflower	*Apis cerana*	Hubei	Central China	31°20N	110°21E
ES66	wildflower	*Apis cerana*	Hubei	Central China	30°60N	110E
ES67	wildflower	*Apis cerana*	Hubei	Central China	30°60N	110°08E
ES15	Acacia	*Apis melifera*	Hubei	Central China	31°06N	110°34E
ES17	Acacia	*Apis melifera*	Hubei	Central China	31°05N	110°34E
ES19	Acacia	*Apis melifera*	Hubei	Central China	30°69N	109°76E
ES28	chaste	*Apis melifera*	Hubei	Central China	31°31N	110°68E
HuB-YC-17	rape	*Apis melifera*	Hubei	Central China	29°73N	112°32E
HuB-YC-19	rape	*Apis melifera*	Hubei	Central China	29°84N	112°48E

HuB-YC-45	rape	*Apis melifera*	Hubei	Central China	31°21N	112°48E
HuB-YC-47	rape	*Apis melifera*	Hubei	Central China	31°20N	112°44E
WJ-5	sapium	*Apis melifera*	Hunan	Central China	27°52N	112°63E
KH01	sunflower	*Apis melifera*	Inner Mongolia	Northern China	44°13N	123°33E
HuB-YC-15	rape	*Apis melifera*	Jiangsu	Eastern China	33°35N	120°15E
WJ-4	sapium	*Apis melifera*	Jiangxi	Eastern China	28°68N	115°89E
JL51	linden	*Apis melifera*	Jilin	Northeast China	43°78N	126°64E
JL64	linden	*Apis melifera*	Jilin	Northeast China	44°09N	127°08E
JL74	linden	*Apis melifera*	Jilin	Northeast China	44°25N	127°08E
FX2017-011	Acacia	*Apis melifera*	Liaoning	Northeast China	41°72N	123°45E
LN01	Acacia	*Apis melifera*	Liaoning	Northeast China	40°73N	120°5E
LN17	Acacia	*Apis melifera*	Liaoning	Northeast China	40°89N	120°19E
LN21	chaste	*Apis melifera*	Liaoning	Northeast China	40°73N	120°42E
LN43	chaste	*Apis melifera*	Liaoning	Northeast China	40°72N	119°95E
TS64	Acacia	*Apis melifera*	Liaoning	Northeast China	41°21N	121°07E
TS75	jujube	*Apis melifera*	Liaoning	Northeast China	41°21N	121°07E
TS94	chaste	*Apis melifera*	Liaoning	Northeast China	41°29N	120°85E
TS99	chaste	*Apis melifera*	Liaoning	Northeast China	41°23N	120°35E
KH05	sunflower	*Apis melifera*	Ningxia	Northwestern China	37°79N	107°41E
QM01	buckwheat	*Apis melifera*	Ningxia	Northwestern China	37°79N	107°41E
QH01	rape	*Apis melifera*	Qinghai	Northwestern China	35°46N	101°13EE
QH13	rape	*Apis melifera*	Qinghai	Northwestern China	36°58N	101°54E
QH19	rape	*Apis melifera*	Qinghai	Northwestern China	37°8N	100°34E
SX28	jujube	*Apis cerana*	Shaanxi	Northwestern China	36°08N	110°27E
KH03	sunflower	*Apis melifera*	Shaanxi	Northwestern China	37°45N	107°32E
KH09	sunflower	*Apis melifera*	Shaanxi	Northwestern China	37°97N	109°29E

KH13	sunflower	*Apis melifera*	Shaanxi	Northwestern China	37°60N	107°61E
KH19	sunflower	*Apis melifera*	Shaanxi	Northwestern China	37°50N	108°26E
QM03	buckwheat	*Apis melifera*	Shaanxi	Northwestern China	37°67N	107°83E
QM06	buckwheat	*Apis melifera*	Shaanxi	Northwestern China	37°97N	109°29E
QM07	buckwheat	*Apis melifera*	Shaanxi	Northwestern China	37°58N	108°57E
QM10	buckwheat	*Apis melifera*	Shaanxi	Northwestern China	37°6N	107°61E
SX01	jujube	*Apis melifera*	Shaanxi	Northwestern China	36°87N	110°36E
SX13	jujube	*Apis melifera*	Shaanxi	Northwestern China	36°55N	109°82E
SX17	jujube	*Apis melifera*	Shaanxi	Northwestern China	37°03N	110°14E
ZH-04	jujube	*Apis melifera*	Shaanxi	Northwestern China	38°04N	110°5E
ZH-09	jujube	*Apis melifera*	Shaanxi	Northwestern China	36°88N	110°2E
FX2017-018	Acacia	*Apis melifera*	Shandong	Eastern China	36°65N	117°12E
w2018-199	wildflower	*Apis cerana*	Shanxi	Northern China	38°42N	112°73E
w2018-269	wildflower	*Apis cerana*	Shanxi	Northern China	38°42N	112°73E
k18-208	Acacia	*Apis melifera*	Shanxi	Northern China	38°42N	112°72E
shanxi-1	Acacia	*Apis melifera*	Shanxi	Northern China	37°95N	110°99E
JC18001	wildflower	*Apis cerana*	Sichuan	Southwestern China	29°68N	105°18E
JC18003	rape	*Apis cerana*	Sichuan	Southwestern China	29°62N	105°12E
JC18037	citrus	*Apis cerana*	Sichuan	Southwestern China	31°01N	103°99E
JC18009	rape	*Apis melifera*	Sichuan	Southwestern China	29°47N	104°91E
JC18016	rape	*Apis melifera*	Sichuan	Southwestern China	31°23N	104°52E
JC18029	rape	*Apis melifera*	Sichuan	Southwestern China	30°42N	103°47E
JC18043	citrus	*Apis melifera*	Sichuan	Southwestern China	30°93N	104°04E
JC18057	citrus	*Apis melifera*	Sichuan	Southwestern China	31°54N	104°86E
JC18064	citrus	*Apis melifera*	Sichuan	Southwestern China	29°15N	103°92E
JC18069	citrus	*Apis melifera*	Sichuan	Southwestern China	29°49N	103°6E

JC18075	citrus	*Apis melifera*	Sichuan	Southwestern China	30°02N	103°52E
JC18086	citrus	*Apis melifera*	Sichuan	Southwestern China	31°78N	104°75E
JC18101	citrus	*Apis melifera*	Sichuan	Southwestern China	30°05N	103°62E
WJ-1	sapium	*Apis melifera*	Yunnan	Southwestern China	25°72N	101°33E
ZJ13	wildflower	*Apis cerana*	Zhejiang	Eastern China	28°74N	118°62E
ZJ15	wildflower	*Apis cerana*	Zhejiang	Eastern China	28°83N	118°68E

Table S3 The sequence of the samples, QC samples and blank samples.

Sample Type	Sample	Position	Injection volume (μL)
Blank	CK_0	GA1	5
Blank	CK_1	GA1	5
Blank	CK_2	GA1	5
QC	QC_1	GA2	5
QC	QC_2	GA2	5
QC	QC_3	GA2	5
QC	QC_4	GA2	5
QC	QC_5	GA2	5
QC	QC_6	GA2	5
Unknown	Z_6	GA3	5
Unknown	Y_6	GA4	5
Unknown	Y_16	GA5	5
Unknown	Z_9	GA6	5
Unknown	Z_15	GA7	5
Unknown	Y_4	GA8	5
Unknown	Z_3	GB1	5
Unknown	Z_19	GB2	5
Unknown	Y_12	GB3	5
Unknown	Z_18	GB4	5
QC	QC_7	GA2	5
QC	QC_8	GA2	5
Blank	CK_3	GA1	5
Unknown	Y_9	GB5	5
Unknown	Z_13	GB6	5

Unknown	Y_18	GB7	5
Unknown	Y_19	GB8	5
Unknown	Z_4	GC1	5
Unknown	Z_11	GC2	5
Unknown	Y_13	GC3	5
Unknown	Z_20	GC4	5
Unknown	Z_10	GC5	5
Unknown	Y_15	GC6	5
QC	QC_9	GA2	5
QC	QC_10	GA2	5
Blank	CK_4	GA1	5
Unknown	Y_10	GC7	5
Unknown	Y_20	GC8	5
Unknown	Z_12	GD1	5
Unknown	Y_2	GD2	5
Unknown	Z_17	GD3	5
Unknown	Y_7	GD4	5
Unknown	Z_16	GD5	5
Unknown	Y_1	GD6	5
Unknown	Z_7	GD7	5
Unknown	Y_8	GD8	5
QC	QC_11	GA2	5
QC	QC_12	GA2	5
Blank	CK_5	GA1	5
Unknown	Z_14	GE1	5
Unknown	Y_5	GE2	5

Unknown	Z_2	GE3	5
Unknown	Y_14	GE4	5
Unknown	Y_17	GE5	5
Unknown	Z_1	GE6	5
Unknown	Z_8	GE7	5
Unknown	Y_11	GE8	5
Unknown	Z_5	BD1	5
Unknown	Y_3	BD2	5
QC	QC_13	GA2	5
QC	QC_14	GA2	5
Blank	CK_6	GA1	5

Y represented *Apis mellifera ligustica* Spinola honey; Z represented *Apis cerana cerana* Fabricius honey; CK represented blank samples.

Table S4 The information of all potential differential metabolites.

Id	Retention Time (min)	m/z	Mean Y	Mean Z	VIP	p-Value	Fold Change
1	6.882	187.06352	2.28423E-05	7.41954E-07	1.507799679	0.001012745	30.78671997
2	1.886	180.12654	7.6237E-07	0.00056364	1.503005452	0.018633782	0.001352582
3	5.669	189.07918	8.04822E-05	3.45488E-06	1.504000719	0.001087698	23.29525153
4	6.743	283.10584	3.78481E-05	1.13834E-06	1.510952297	0.001191566	33.24837499
5	7.993	217.13161	1.86673E-06	9.14495E-05	1.515197453	0.038650637	0.020412688
6	2.771	331.16342	8.78175E-07	9.81748E-05	1.518490382	0.020178666	0.008945016
7	8.395	478.11182	3.00374E-07	1.22716E-05	1.519644634	0.00059684	0.024477116
8	5.148	272.11622	1.55134E-05	4.20726E-07	1.527834325	0.000236135	36.87283798
9	2.948	572.30369	2.22389E-05	5.29101E-07	1.532375483	0.000623538	42.03151998
10	4.435	262.09566	1.07161E-05	3.30136E-07	1.534655582	0.000205436	32.45962653
11	2.129	460.21688	2.10473E-05	4.03122E-07	1.536859772	0.000229669	52.21083923
12	6.618	301.16802	1.55415E-05	5.74208E-07	1.54272168	0.00226055	27.06601318
13	3.99	230.10578	0.000331439	7.12005E-06	1.543591453	0.000353285	46.55007696
14	7.89	302.04287	1.50446E-06	3.76558E-05	1.546161025	6.86302E-05	0.039953068
15	3.022	132.06894	9.88337E-06	4.66607E-07	1.54738416	0.000145716	21.18135321
16	7.247	252.08995	8.23776E-06	3.9084E-07	1.552097512	0.000106077	21.07707576
17	4.962	372.19005	3.0507E-05	1.43049E-06	1.556818453	1.18988E-05	21.32632719
18	4.695	586.34476	2.29736E-05	5.12994E-07	1.561909516	0.002954607	44.78335837
19	2.724	309.67726	1.96177E-05	6.27834E-07	1.562899248	0.000530767	31.24662569
20	9.181	262.0743	1.10495E-05	2.52214E-07	1.567724302	0.000202829	43.80989368
21	5.608	145.08933	9.7061E-06	3.87527E-07	1.568875416	0.004251502	25.04627056
22	5.824	283.10582	0.000218082	4.63095E-06	1.569588531	0.001179586	47.09233584

23	7.788	313.11636	0.000177081	3.24864E-06	1.569961245	0.000513	54.50939177
24	7.474	245.10538	5.19815E-07	1.20325E-05	1.583356405	0.001890774	0.043200987
25	8.482	91.0428	8.81295E-06	4.22693E-07	1.584580772	0.000348276	20.84953502
26	6.78	358.1744	3.83594E-07	1.60743E-05	1.585663023	0.005475963	0.023863832
27	2.121	265.09532	0.000169377	5.1206E-06	1.586886658	0.000202794	33.0775659
28	9.611	470.20552	3.33241E-05	1.65823E-06	1.587347263	0.000120967	20.09624065
29	8.473	119.03748	0.000148159	2.79426E-06	1.589313591	0.000179895	53.02254141
30	5.529	573.34934	6.30428E-06	2.6715E-07	1.592098009	0.000213477	23.59828642
31	2.168	473.29667	2.2383E-05	8.79772E-07	1.596662264	0.001208573	25.441871
32	10.603	119.03723	0.000209647	6.07441E-06	1.597796354	0.000899218	34.51309692
33	8.627	280.12144	1.51952E-05	3.93044E-07	1.60166865	0.000102487	38.66038178
34	6.891	301.16801	1.89016E-05	3.90669E-07	1.602216009	0.002851902	48.38257898
35	4.966	147.06859	0.000131157	5.19391E-06	1.602488364	2.67245E-05	25.25204739
36	3.194	216.0902	0.000218346	5.32714E-06	1.603463339	0.000121818	40.9875037
37	7.904	464.09613	1.04389E-06	5.85589E-05	1.605989746	0.00023384	0.017826403
38	8.486	209.11672	0.000141977	6.75537E-06	1.613587944	1.12635E-05	21.01687985
39	7.826	241.14674	2.1487E-05	5.47615E-07	1.618642017	0.00237087	39.23743022
40	2.234	447.17445	1.41009E-05	5.90884E-07	1.621022965	0.000145532	23.86402822
41	7.661	133.05296	0.000138161	9.98305E-06	1.627208721	0.000116475	20.83956516
42	2.669	492.1749	9.42189E-06	3.87894E-07	1.630219546	0.000219411	24.28986072
43	3.223	700.38767	8.90337E-05	4.39013E-07	1.632588121	0.016355725	202.8042761
44	3.167	172.10025	1.33745E-05	5.08769E-07	1.637505231	0.000319324	26.28788357
45	2.741	273.18026	0.000113307	3.56885E-06	1.64081016	4.07714E-05	31.74882766
46	6.757	301.16805	4.19776E-05	5.93059E-07	1.647108894	0.001788298	70.78153724
47	10.72	343.1416	6.2085E-05	1.91307E-06	1.650147618	3.81777E-05	32.4530609

48	7.489	241.14678	2.86754E-05	4.18243E-07	1.654417394	0.000678114	68.56167804
49	2.678	342.12174	4.05185E-05	1.75812E-06	1.660198733	9.54685E-06	23.04647269
50	5.991	161.04782	2.0719E-05	0.000537752	1.675941858	0.00156495	0.038528864
51	5.99	189.04276	0.000176674	0.00545062	1.677413171	0.000574784	0.032413554
52	2.745	457.30173	0.000540769	1.28439E-05	1.683175124	5.58799E-05	42.10308766
53	8.474	341.1265	4.47444E-05	1.32264E-06	1.69320494	6.67064E-06	33.82969369
54	8.881	456.19001	0.000140329	1.97261E-06	1.698892975	1.21816E-05	71.13860194
55	7.066	339.18965	2.30793E-06	5.05068E-05	1.714596057	1.36202E-09	0.045695413
56	6.559	289.13146	1.88639E-05	8.94154E-07	1.721458408	9.80109E-05	21.09694725
57	2.735	386.26446	3.26674E-05	5.89622E-07	1.725554654	3.15883E-05	55.40403432
58	8.474	456.19002	0.000139333	2.15118E-06	1.727977461	9.83323E-06	64.77037675
59	7.025	289.13164	3.75111E-05	1.46203E-06	1.730806937	6.32364E-05	25.6567824
60	3.493	767.39231	8.80422E-07	3.01214E-05	1.733179807	1.64571E-05	0.029229116
61	3.378	222.64511	2.57093E-05	8.22865E-07	1.745605472	2.23345E-06	31.24361205
62	5.374	663.37105	1.56998E-05	3.08925E-07	1.746852012	5.95524E-07	50.82066943
63	3.391	445.29016	2.63446E-05	9.47059E-07	1.754907794	9.62765E-07	27.81725837
64	5.062	486.31698	1.02639E-06	8.12301E-05	1.759126762	9.6217E-12	0.012635539
65	7.64	393.22984	2.38652E-05	1.04966E-06	1.759236316	2.3004E-08	22.73612809
66	5.89	227.13213	2.09438E-05	7.33196E-07	1.760806146	1.22042E-06	28.56513845
67	6.571	243.15846	2.69701E-06	7.83914E-05	1.762552077	3.17552E-09	0.034404386
68	7.651	786.45998	2.35151E-05	8.96685E-07	1.765487929	1.77287E-08	26.22447145
69	6.577	486.31653	2.72979E-06	7.90619E-05	1.767073355	2.00088E-09	0.034527274
70	6.979	747.42863	0.000119719	2.38951E-06	1.770356556	5.20529E-08	50.10187466
71	6.038	383.19892	1.6012E-05	4.96448E-07	1.774684302	1.82959E-07	32.25312647
72	3.606	761.46557	5.53196E-05	9.35836E-07	1.776041252	7.67831E-07	59.11247252

73	2.021	305.49568	2.10807E-05	8.82175E-07	1.777408259	4.51202E-05	23.8963239
74	6.672	514.32298	1.67998E-05	7.22794E-07	1.777756206	1.75655E-07	23.24291724
75	6.27	415.14806	4.19007E-05	1.73561E-06	1.778317935	5.89871E-05	24.14175533
76	6.125	534.29166	8.15293E-07	1.70582E-05	1.781902971	6.39556E-08	0.047794856
77	2.014	916.48743	1.96737E-05	8.76892E-07	1.783749497	2.37333E-05	22.43567537
78	1.32	561.31289	1.24931E-06	3.23153E-05	1.783906103	2.27148E-08	0.038660016
79	1.284	280.65643	1.09875E-06	3.29761E-05	1.785337858	2.09253E-08	0.033319772
80	2.156	433.25439	3.07783E-05	6.42677E-07	1.786449484	6.77073E-05	47.8908439
81	5.23	599.37626	1.68435E-06	0.000127116	1.789901579	5.53439E-05	0.013250464
82	7.719	461.18398	5.38895E-07	2.46065E-05	1.790294764	1.39812E-10	0.021900557
83	7.529	446.30563	5.93049E-05	1.5491E-06	1.791861573	1.30637E-06	38.28352975
84	2.165	216.62709	3.04989E-05	5.42254E-07	1.793645837	5.49291E-05	56.24461121
85	3.758	437.19168	2.58051E-05	6.95598E-07	1.794991199	1.71406E-10	37.09773438
86	6.061	515.30711	3.30981E-06	7.3573E-05	1.800666779	1.2861E-10	0.044986724
87	7.536	892.61164	5.23688E-05	1.40904E-06	1.802528761	2.33328E-08	37.16638544
88	5.172	287.19594	5.29471E-07	3.82807E-05	1.804444719	2.67873E-07	0.013831269
89	6.601	372.23749	3.33677E-05	1.24336E-06	1.806694651	2.68039E-11	26.83669924
90	7.875	1150.65037	5.35622E-07	3.35998E-05	1.807056551	1.79796E-06	0.0159412
91	6.675	460.25379	7.94238E-05	2.04032E-06	1.812768863	1.21185E-11	38.92718683
92	7.518	378.8785	8.15678E-05	3.51901E-07	1.814327161	9.23008E-06	231.7921196
93	6.594	445.25404	9.86014E-07	5.79901E-05	1.816951917	9.25012E-11	0.017003156
94	1.492	537.21926	7.38825E-07	3.07279E-05	1.817850247	3.29831E-08	0.024044062
95	1.409	279.16228	0.000145707	2.14878E-06	1.820995062	1.51536E-07	67.80916092
96	7.525	1136.6351	0.000160688	2.03314E-06	1.822073893	3.93904E-07	79.03448481
97	6.573	962.55526	2.2641E-05	8.72245E-07	1.824233631	4.93169E-10	25.95719822

98	2.276	853.40523	2.46168E-05	5.39855E-07	1.825499703	3.07937E-08	45.59888519
99	5.275	461.21271	4.01445E-05	1.61914E-06	1.82709681	4.28698E-12	24.7936743
100	6.564	574.32647	9.59356E-07	2.71897E-05	1.827243528	8.03879E-07	0.035283846
101	6.074	549.32795	2.19195E-05	6.03141E-07	1.828745319	4.83532E-08	36.34221816
102	7.251	739.43521	6.79189E-07	2.8924E-05	1.828785275	6.61961E-10	0.023481839
103	1.356	482.16906	4.27133E-05	1.33783E-06	1.82942027	8.94141E-08	31.9274037
104	7.58	1073.52976	3.66502E-05	6.15766E-07	1.829871646	1.83697E-08	59.51960088
105	7.388	818.46567	5.68322E-07	1.98712E-05	1.83145139	6.23147E-10	0.028600211
106	5.769	448.21732	1.72679E-05	5.83214E-07	1.833209802	2.19658E-11	29.60820668
107	5.969	941.53057	4.29815E-07	6.33879E-05	1.833822773	6.27333E-09	0.006780714
108	5.392	535.30105	1.46171E-05	2.67105E-07	1.834315535	1.98719E-07	54.72429731
109	5.174	471.31726	7.96012E-07	0.000157332	1.836617179	3.29185E-07	0.005059446
110	1.596	570.31316	0.000234755	1.30959E-06	1.837269027	4.68953E-08	179.257504
111	8.238	400.2687	2.77592E-05	5.94636E-07	1.838652869	1.46801E-10	46.68269825
112	2.692	600.36006	4.89496E-05	8.24759E-07	1.838791914	7.4426E-13	59.35013529
113	5.785	323.14841	2.12776E-05	9.88943E-07	1.840390273	7.80314E-11	21.51552609
114	2.749	588.32375	3.07306E-05	5.24378E-07	1.841586396	4.64322E-09	58.60384025
115	2.475	472.30133	3.96071E-05	1.13199E-06	1.844201532	1.40761E-09	34.98891016
116	6.946	415.89644	2.05877E-05	4.51619E-07	1.844757135	5.27899E-10	45.58643039
117	2.05	672.35618	3.24448E-05	4.53518E-07	1.846713343	2.16888E-08	71.5403372
118	3.562	605.33906	5.30586E-07	8.43633E-05	1.847535433	1.24272E-08	0.006289296
119	7.445	761.44423	5.22072E-07	9.5906E-05	1.847978603	1.71268E-10	0.005443584
120	2.088	503.27076	4.44872E-07	2.6321E-05	1.84954262	5.6295E-11	0.016901812
121	3.55	302.66985	5.98755E-07	8.70373E-05	1.849990667	7.85681E-09	0.006879296
122	4.539	713.41906	5.41023E-07	0.00012973	1.851549628	1.50509E-09	0.004170378

123	4.551	356.70952	5.2668E-07	0.000133807	1.851743853	6.68611E-10	0.003936109
124	3.632	534.31703	3.83313E-07	0.000116468	1.853187623	1.22376E-08	0.003291145
125	7.044	407.24242	2.89296E-05	5.14079E-07	1.853617969	3.64168E-09	56.27471538
126	5.261	614.37569	3.00315E-07	4.23956E-05	1.855024312	4.94273E-14	0.007083647
127	2.409	483.24461	3.50581E-05	6.13358E-07	1.857035389	6.66097E-14	57.15760123

Y represented *Apis mellifera ligustica* Spinola honey; Z represented *Apis cerana cerana* Fabricius honey