

Supplementary materials

S1. The equations of sensitivity (SE), specificity (SP), classification accuracy and area under curve (AUC)

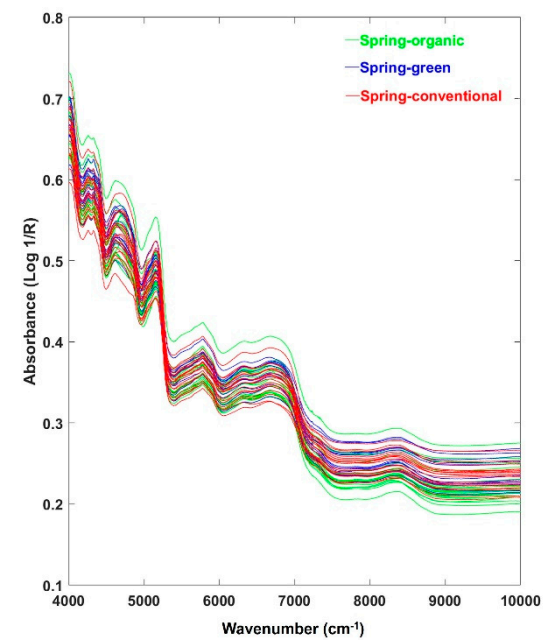
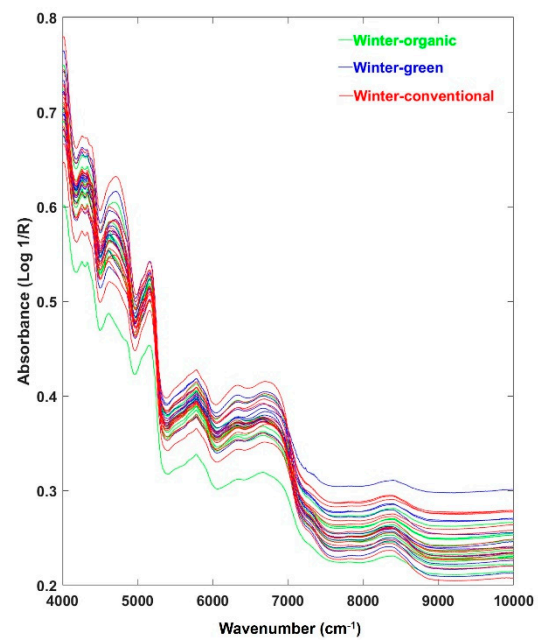
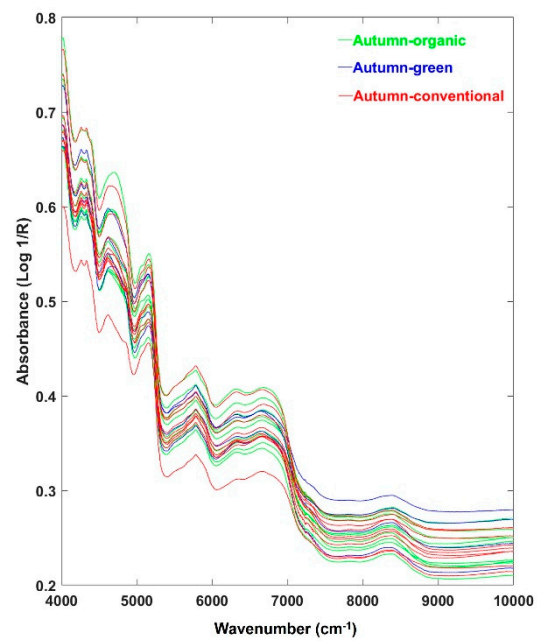
$$SE (\%) = \frac{TP}{TP + FN} \times 100$$

$$SP (\%) = \frac{TN}{TN + FP} \times 100$$

$$Accuracy (\%) = 1 - \frac{FP + FN}{TP + FP + TN + FN} \times 100$$

Where TP: true positive (positive samples correctly classified), TN: true negative (negative sample correctly classified), FP: false positive (positive samples incorrectly classified), FN: false negative (negative samples incorrectly classified).

Area under curve (AUC) is a receiver operating characteristic curve, which is plotted on the vertical axis of sensitivity (true positive rate) and abscissa of false positive rate (1- specificity) according to a series of different dichotomies (cut-off values or determination thresholds). The closer the AUC is to 1.0, the higher the authenticity of the method.



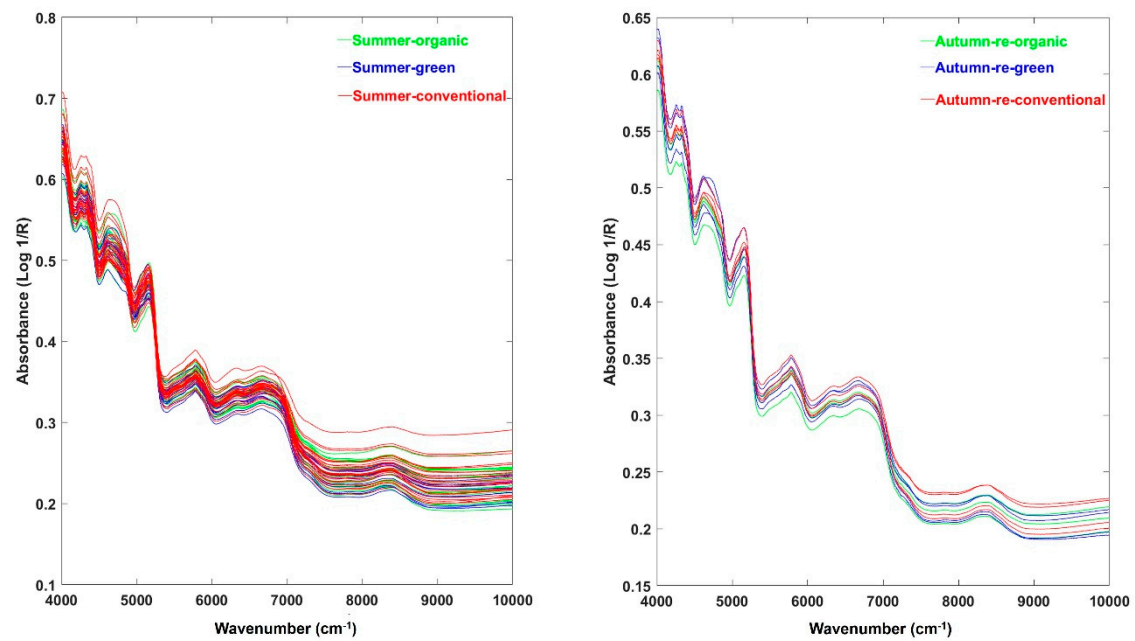


Figure S1. BC raw NIR spectra under different seasons

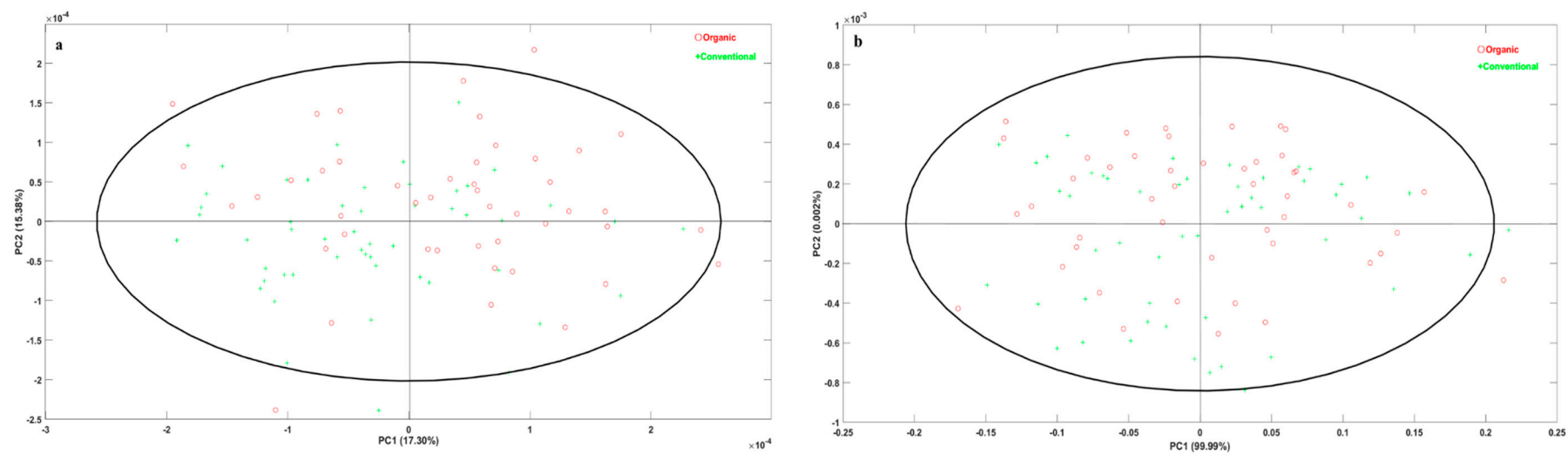


Figure S2. The first two principal component score plots of PLS-DA for different *BC* cultivation methods using NIR (a) and IRMS-NIR (b).

Table S1. PLS-DA models of BC different cultivation methods using IRMS

Cultivation methods	CV	Calibration set					Validation set accuracy (%)
	optLVs	Accuracy (%)	SE (%)	SP (%)	AUC (%)	accuracy (%)	
ORG <i>vs.</i> CON	2	80.45	63.64	88.89	78.68	77.55 (76/98)	75.76 (25/33)
ORG <i>vs.</i> GRE	4	61.27	88.00	43.33	70.28	71.25 (57/80)	51.85 (14/27)
GRE <i>vs.</i> CON	3	65.83	90.74	43.33	73.23	73.81 (62/84)	53.57 (15/28)

ORG: organic; GRE: green; CON: conventional; OptLVs: the optimal number of latent variables; SE: sensitivity; SP: specificity; AUC: classification error rate and area under curve.

Table S2. Comparison of parameters in the NIRS-PLS-DA models with NSD preprocessing method for different BCs cultivation methods.

Cultivation methods	Models	CV	Calibration set					Validation set
		optLVs	Accuracy (%)	SE (%)	SP (%)	AUC (%)	Accuracy (%)	Accuracy (%)
ORG <i>vs.</i> CON	PLS-DA	11	81.79	85.42	90.00	87.83	87.76	78.79
	NSD(3,3,1) ^a -PLS-DA	9	76.53	86.96	88.46	87.71	87.76	63.64
	NSD(3,3,2)-PLS-DA	10	73.47	100	98.08	98.94	98.98	72.73
	NSD(5,5,1)-PLS-DA	9	78.57	89.36	90.20	89.76	89.80	69.70
	NSD(5,5,2)-PLS-DA	11	70.41	91.30	92.31	91.81	91.84	81.82
	NSD(7,7,1)-PLS-DA	12	76.53	87.50	96.00	92.17	91.84	72.73
	NSD(7,7,2)-PLS-DA	22	69.39	100	100	100	100	75.76
	NSD(9,9,1)-PLS-DA	15	73.47	89.80	91.84	90.83	90.82	72.73
	NSD(9,9,2)-PLS-DA	27	69.39	100	100	100	100	66.67
ORG <i>vs.</i> GRE	PLS-DA	26	50.00	100	100	100	100	62.96
	NSD(3,3,1)-PLS-DA	16	58.75	100	100	100	100	59.26
	NSD(3,3,2)-PLS-DA	14	51.25	100	100	100	100	55.56
	NSD(5,5,1)-PLS-DA	15	51.25	96.00	96.67	95.75	96.25	66.67
	NSD(5,5,2)-PLS-DA	7	46.25	91.30	70.59	83.24	82.50	66.67
	NSD(7,7,1)-PLS-DA	24	51.25	100	100	100	100	66.67
	NSD(7,7,2)-PLS-DA	16	56.25	95.83	90.63	93.71	93.75	59.26
	NSD(9,9,1)-PLS-DA	24	58.75	100	100	100	100	66.67

GRE <i>vs.</i> CON	NSD(9,9,2)-PLS-DA	28	58.75	100	100	100	100	70.37
	PLS-DA	20	53.57	100	90.91	97.22	96.43	71.43
	NSD(3,3,1)-PLS-DA	14	67.86	97.96	94.29	96.53	96.43	57.14
	NSD(3,3,2)-PLS-DA	5	64.29	86.79	80.65	83.29	57.14	57.14
	NSD(5,5,1)-PLS-DA	19	59.52	100	97.14	99.00	98.81	57.14
	NSD(5,5,2)-PLS-DA	22	57.14	100	100	100	100	60.71
	NSD(7,7,1)-PLS-DA	25	58.33	100	100	100	100	60.71
	NSD(7,7,2)-PLS-DA	10	69.05	86.00	76.47	81.55	53.57	53.57
	NSD(9,9,1)-PLS-DA	23	57.14	100	100	100	100	67.86
	NSD(9,9,2)-PLS-DA	16	58.37	81.25	95.83	90.66	90.00	59.37

Table S3. The $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of different *BC* cultivation methods divided as the calibration and validation sets of the optimal IRMS-NIRS-PLS-DA models

Combination	Cultivation method	Variables	Calibration set				Validation set			
			No. of samples	Mean \pm SD	Range		No. of samples	Mean \pm SD	Range	
ORG <i>vs.</i> CON	ORG	$\delta^{13}\text{C}$	46	-29.05 \pm 1.32	-31.17— -25.90		17	-29.51 \pm 1.44	-32.09— -26.99	
		$\delta^{15}\text{N}$		+11.24 \pm 6.17	-1.28— +24.15			+8.50 \pm 6.03	-1.16— +23.13	
		$\delta^2\text{H}$		-79.05 \pm 9.32	-100.52— -58.55			-84.28 \pm 9.89	-101.88— -66.37	
		$\delta^{18}\text{O}$		+21.51 \pm 2.81	+15.41— +27.45			+21.19 \pm 3.05	+14.14— +26.53	
	CON	$\delta^{13}\text{C}$	52	-28.63 \pm 1.54	-31.73— -25.10		16	-29.26 \pm 1.17	-30.69— -27.44	
		$\delta^{15}\text{N}$		+4.01 \pm 5.78	-2.68— +24.44			+2.17 \pm 3.19	-3.20— +9.12	
		$\delta^2\text{H}$		-79.94 \pm 12.11	-109.90— -61.93			-81.32 \pm 9.55	-98.57— -68.93	
		$\delta^{18}\text{O}$		+20.63 \pm 2.39	+14.26— +25.70			+21.06 \pm 1.98	+17.37— +24.44	
ORG <i>vs.</i> GRE	ORG	$\delta^{13}\text{C}$	50	-29.06 \pm 1.40	-32.09— -25.90		13	-29.62 \pm 1.10	-31.44— -27.99	
		$\delta^{15}\text{N}$		+9.88 \pm 6.38	-1.28— +24.15			+12.87 \pm 4.98	+7.14— +20.63	
		$\delta^2\text{H}$		-80.67 \pm 10.06	-101.88— -58.55			-79.65 \pm 8.38	-91.84— -69.03	
		$\delta^{18}\text{O}$		+21.24 \pm 3.04	+14.14— +27.45			+22.12 \pm 1.91	+18.40— +24.89	
	GRE	$\delta^{13}\text{C}$	30	-29.07 \pm 1.30	-31.22— -26.42		14	-29.22 \pm 0.79	-30.18— -27.63	
		$\delta^{15}\text{N}$		+6.14 \pm 5.32	-1.33— +19.47			+8.01 \pm 4.69	+0.54— +15.22	
		$\delta^2\text{H}$		-78.54 \pm 10.76	-103.83— -59.22			-78.75 \pm 6.86	-89.51— -69.88	

GRE vs. CON	GRE	$\delta^{18}\text{O}$		+21.23±2.23	+17.01—+25.54		+21.68±1.07	+20.44—+23.33
		$\delta^{13}\text{C}$	32	-29.03±1.28	-31.22— -26.42	12	-29.36±0.72	-30.51— -28.24
		$\delta^{15}\text{N}$		+7.18±5.52	-1.33—+19.47		+5.55±3.97	+0.72—+13.95
		$\delta^2\text{H}$		-77.60±10.03	-103.83— -59.22		-81.29±8.12	-94.02— -70.90
	CON	$\delta^{18}\text{O}$		+21.47±2.15	+17.01—+25.54		+21.12±1.22	+18.92—+23.11
		$\delta^{13}\text{C}$	52	-28.75±1.57	-31.73— -25.10	16	-28.86±1.16	-30.69— -27.23
		$\delta^{15}\text{N}$		+4.21±5.81	-3.20—+24.44		+1.53±2.48	-1.84—+6.16
		$\delta^2\text{H}$		-80.43±12.29	-109.90— -61.93		-79.75±8.78	-92.56— -64.99
		$\delta^{18}\text{O}$		+20.64±2.44	+14.26—+25.70		+21.04±1.75	+17.37—+24.44

ORG: organic; GRE: green; CON: conventional; SD: Standard deviation.

Table S4. Comparison of parameters in the IRMS-NIRS-PLS-DA models with NSD preprocessing method for different BCs cultivation methods.

Combination	Models	CV	Calibration set					Validation set
		optLVs	Accuracy (%)	SE (%)	SP (%)	AUC (%)	Accuracy (%)	Accuracy (%)
ORG <i>vs.</i> CON	PLS-DA	12	35.71	80.43	86.54	83.71	83.67	87.88
	NSD(3,3,1) ^a -PLS-DA	12	71.43	88.37	94.55	91.96	91.84	78.79
	NSD(3,3,2)-PLS-DA	8	64.29	86.96	90.38	88.78	88.78	78.79
	NSD(5,5,1)-PLS-DA	13	73.47	86.05	94.55	91.08	90.82	78.79
	NSD(5,5,2)-PLS-DA	13	73.47	89.13	90.38	89.76	89.80	87.88
	NSD(7,7,1)-PLS-DA	13	69.39	86.05	92.73	89.86	89.80	81.82
	NSD(7,7,2)-PLS-DA	14	67.35	86.96	90.38	88.78	88.78	78.79
	NSD(9,9,1)-PLS-DA	12	71.43	90.70	83.64	86.62	86.73	84.85
	NSD(9,9,2)-PLS-DA	16	68.37	86.96	90.38	88.78	88.78	78.79
ORG <i>vs.</i> GRE	PLS-DA	25	51.25	97.87	100	98.53	98.75	81.48
	NSD(3,3,1)-PLS-DA	10	52.50	86.00	73.33	80.09	81.25	88.89
	NSD(3,3,2)-PLS-DA	16	48.75	100	100	100	100	88.89
	NSD(5,5,1)-PLS-DA	10	48.75	84.00	66.67	76.10	77.50	85.19
	NSD(5,5,2)-PLS-DA	15	52.50	94.00	100	95.45	96.25	85.19
	NSD(7,7,1)-PLS-DA	11	55.00	84.00	66.67	76.10	77.50	81.48
	NSD(7,7,2)-PLS-DA	9	60.00	90.00	70.00	82.05	82.50	81.48
	NSD(9,9,1)-PLS-DA	11	56.25	84.00	63.33	74.81	76.25	81.48

GRE <i>vs.</i> CON	NSD(9,9,2)-PLS-DA	9	55.00	88.00	66.67	79.20	80.00	85.19
	PLS-DA	20	57.14	96.16	81.25	91.07	90.48	75.00
	NSD(3,3,1)-PLS-DA	26	63.10	100	100	100	100	71.43
	NSD(3,3,2)-PLS-DA	11	51.19	98.11	96.77	97.44	97.62	57.14
	NSD(5,5,1)-PLS-DA	14	63.10	96.15	93.75	94.95	95.24	63.10
	NSD(5,5,2)-PLS-DA	12	58.33	90.57	83.87	87.22	88.10	58.33
	NSD(7,7,1)-PLS-DA	14	67.86	94.34	87.10	91.30	91.67	64.29
	NSD(7,7,2)-PLS-DA	13	60.71	88.68	70.97	81.25	82.14	71.43
	NSD(9,9,1)-PLS-DA	6	67.86	87.04	66.67	78.27	79.76	64.29
	NSD(9,9,2)-PLS-DA	11	60.71	81.13	70.97	75.72	77.38	67.86

^a The parameters (s, g, n) in NSD were defined by: s, the number of data in one segment; g, the number of data in one gap; n, 1 or 2 is the first derivative or second derivative.

ORG: organic; GRE: green; CON: conventional; OptLVs: the optimal number of latent variables; SE: sensitivity; SP: specificity; AUC: classification error rate and area under curve.