Supplementary information for:

Detection and Quantification of Adulteration in Krill Oil with Raman and Infrared Spectroscopic Methods

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Figure S1 : An overview of all oil samples used in this experiment. Abbreviation: KO = krill oil; PO = palm oil; O3C = ω-3 concentrates in ethyl ester; FO = fish oil

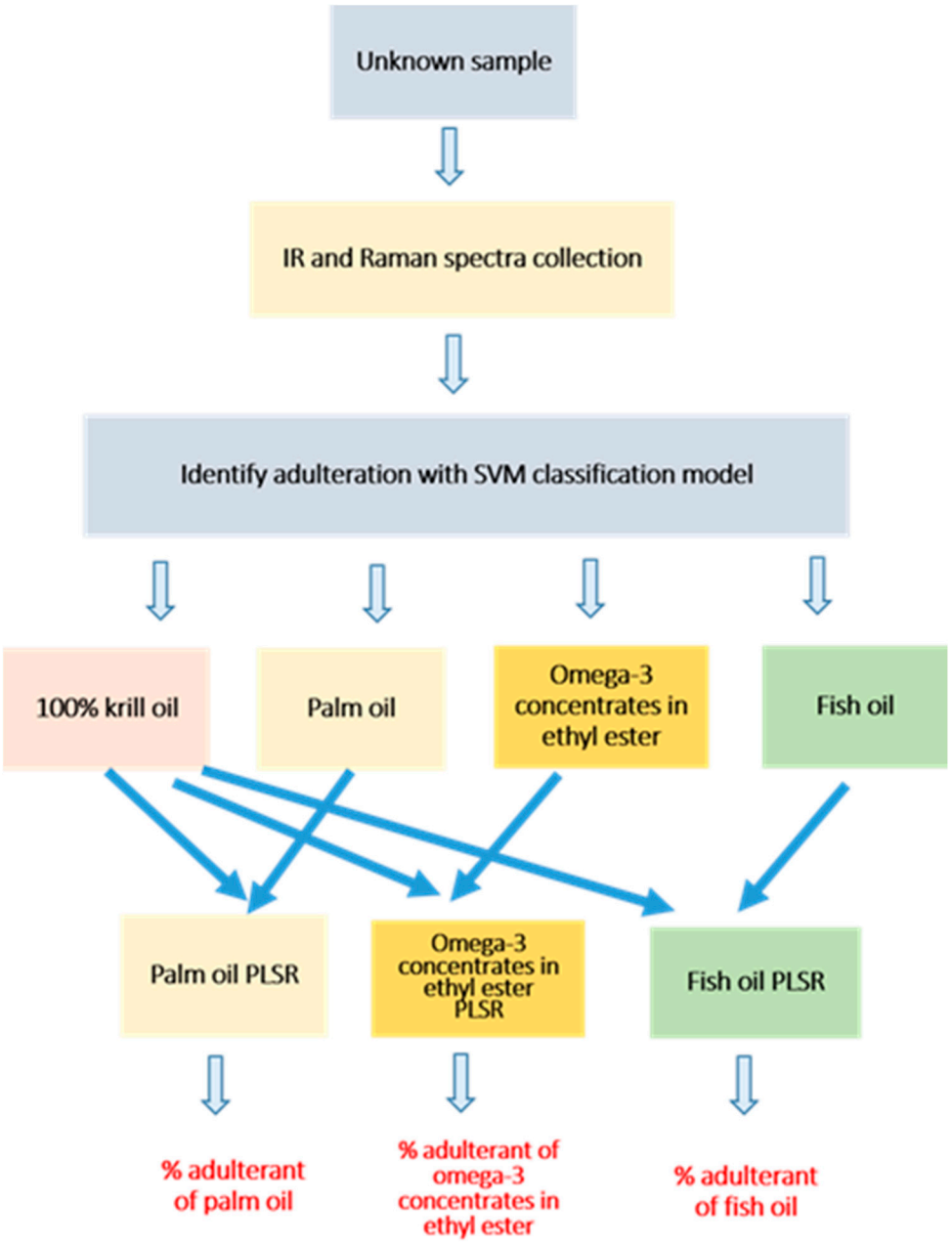

Figure S2 : Decision tree/workflow for evaluating unknown samples

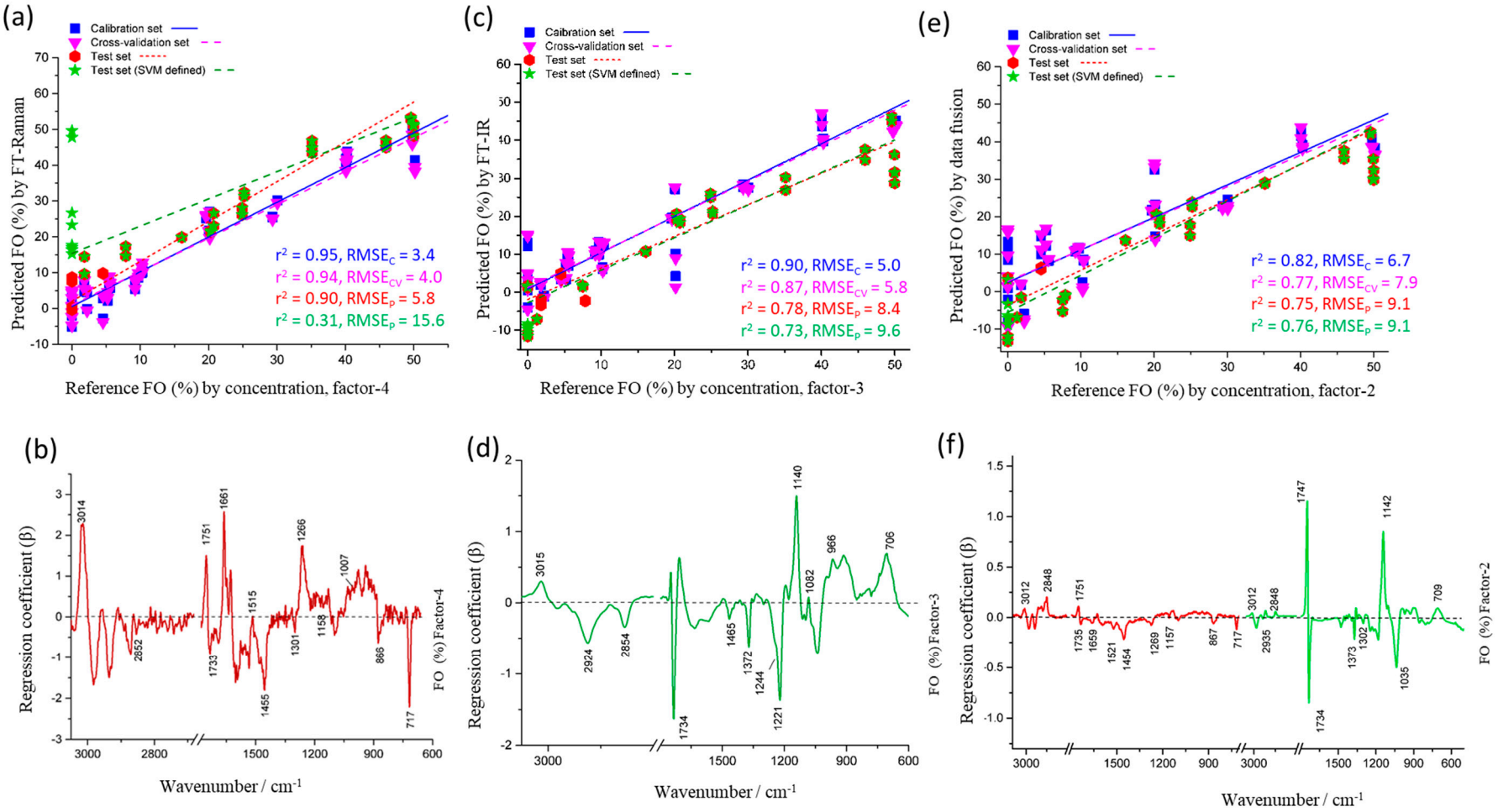

Figure S3 : PLSR calibration lines and regression coefficients for quantitative prediction of FO concentration in krill oil by Raman (a,b), IR (c,d) and low-level fused Raman plus IR data (e,f)

Table S1: A summary of the studied samples with sample name, country of origin, company, batch number and best before

Sample name	Country of origin	company	batch no.	best before
Antarctic krill oil (KO1)	New Zealand	SR	SRE-00453	07/2023
Red super krill (KO2)	New Zealand	Good health	P90L	03/2023
Go Krill oil (KO3)	New Zealand	Go healthy	JO270	01/2023
Krill oil (KO4)	New Zealand	PipingRock	B980E	08/2023
Neptune krill 1000 (KO5)	USA	NOW	1627BT	06/2023
Superior red krill oil (KO6)	New Zealand	Sanderson	B7629	5/2024
Fish oil 2000 concentrated (O3C)	New Zealand	Lighthouse	T8005	09/2021
Palm oil (PO)	New Zealand	Go native New Zealand	01216	12/2022
Fish oil (FO)	New Zealand	Health by habit	KO875	07/2023

Table S2: Summary of the sample mixtures used in this study. Samples are expressed as weight percentages (% w/w). Abbreviations: VO = valuable oil; KO1 to KO6 = Krill oil batch; PO = palm oil; O3C = ω-3 concentrates in ethyl ester; FO = Fish oil; M = model set; T = test set

Predominant oil type	Batch	VO (% w/w)	PO (% w/w)	O3C (% w/w)	FO (% w/w)	Dataset (M or T)
Krill oil samples	KO1	100.0	0.0	0.0	0.0	M
	KO1	97.8	0.0	0.0	2.2	M
	KO1	89.8	0.0	0.0	10.2	M
	KO1	70.0	0.0	0.0	30.0	M
	KO1	50.2	0.0	0.0	49.8	M
	KO1	94.9	0.0	5.1	0.0	M
	KO1	79.6	0.0	20.4	0.0	M
	KO1	97.8	2.2	0.0	0.0	M
	KO1	89.2	10.8	0.0	0.0	M
	KO1	69.6	30.4	0.0	0.0	M
	KO1	51.6	48.4	0.0	0.0	M
	KO1	60.0	0.0	40.0	0.0	M
	KO1	79.9	0.0	20.1	0.0	M
	KO2	100.0	0.0	0.0	0.0	M
	KO2	94.5	0.0	0.0	5.5	M
	KO2	94.4	0.0	0.0	5.6	M
	KO2	90.8	0.0	0.0	9.2	M
	KO2	90.3	0.0	0.0	9.7	M
	KO2	80.4	0.0	0.0	19.6	M
	KO2	59.7	0.0	0.0	40.3	M
	KO2	97.4	0.0	2.6	0.0	M
	KO2	89.8	0.0	10.2	0.0	M
	KO2	70.6	0.0	29.4	0.0	M
	KO2	50.1	0.0	49.9	0.0	M
	KO2	50.0	0.0	50.0	0.0	M
	KO2	94.7	5.3	0.0	0.0	M
	KO2	79.6	20.4	0.0	0.0	M
	KO2	75.8	24.2	0.0	0.0	M
	KO2	75.0	25.0	0.0	0.0	M
	KO2	60.7	39.3	0.0	0.0	M
	KO2	59.9	40.1	0.0	0.0	M
	KO4	100.0	0.0	0.0	0.0	M

	KO4	98.2	0.0	0.0	1.8	M
	KO4	89.6	0.0	0.0	10.4	M
	KO4	70.7	0.0	0.0	29.4	M
	KO4	49.8	0.0	0.0	50.2	M
	KO4	93.6	0.0	6.4	0.0	M
	KO4	80.1	0.0	19.9	0.0	M
	KO4	60.6	0.0	39.4	0.0	M
	KO4	98.0	2.0	0.0	0.0	M
	KO4	90.3	9.7	0.0	0.0	M
	KO4	70.4	29.6	0.0	0.0	M
	KO4	49.7	50.3	0.0	0.0	M
	KO6	100.0	0.0	0.0	0.0	M
	KO6	95.5	0.0	0.0	4.5	M
	KO6	79.9	0.0	0.0	20.1	M
	KO6	59.9	0.0	0.0	40.1	M
	KO6	98.0	0.0	2.0	0.0	M
	KO6	89.9	0.0	10.1	0.0	M
	KO6	70.9	0.0	29.1	0.0	M
	KO6	50.3	0.0	49.7	0.0	M
	KO6	94.4	5.6	0.0	0.0	M
	KO6	79.9	20.1	0.0	0.0	M
	KO6	60.1	39.9	0.0	0.0	M
	KO6	80.2	19.8	0.0	0.0	M
	KO6	79.8	0.0	0.0	20.2	M
	KO6	90.3	0.0	9.7	0.0	M
	KO6	94.7	5.3	0.0	0.0	M
	KO6	94.8	0.0	0.0	5.2	M
	KO6	99.1	0.0	0.9	0.0	M
	KO3	100.0	0.0	0.0	0.0	T
	KO3	95.5	0.0	0.0	4.5	T
	KO3	83.9	0.0	0.0	16.1	T
	KO3	79.7	0.0	0.0	20.3	T
	KO3	79.2	0.0	0.0	20.8	T
	KO3	74.8	0.0	0.0	25.2	T
	KO3	54.1	0.0	0.0	45.9	T
	KO3	50.0	0.0	0.0	50.0	T
	KO3	95.6	0.0	4.4	0.0	T
	KO3	90.8	0.0	9.2	0.0	T
	KO3	74.5	0.0	25.5	0.0	T
	KO3	70.1	0.0	29.9	0.0	T
	KO3	69.8	0.0	30.2	0.0	T

	KO3	64.7	0.0	35.3	0.0	T
	KO3	50.2	0.0	49.8	0.0	T
	KO3	84.5	15.5	0.0	0.0	T
	KO3	74.9	25.1	0.0	0.0	T
	KO3	54.1	45.9	0.0	0.0	T
	KO3	49.5	50.5	0.0	0.0	T
	KO3	50.0	0.0	0.0	50.0	T
	KO5	100.0	0.0	0.0	0.0	T
	KO5	98.2	0.0	0.0	1.8	T
	KO5	92.1	0.0	0.0	7.9	T
	KO5	75.1	0.0	0.0	24.9	T
	KO5	64.9	0.0	0.0	35.1	T
	KO5	50.4	0.0	0.0	49.6	T
	KO5	98.3	0.0	1.7	0.0	T
	KO5	85.0	0.0	15.0	0.0	T
	KO5	75.6	0.0	24.4	0.0	T
	KO5	55.3	0.0	44.7	0.0	T
	KO5	49.7	0.0	50.3	0.0	T
	KO5	98.5	1.5	0.0	0.0	T
	KO5	92.4	7.6	0.0	0.0	T
	KO5	75.2	24.9	0.0	0.0	T
	KO5	65.3	34.7	0.0	0.0	T
	KO5	49.8	50.2	0.0	0.0	T
	KO5	92.5	0.0	0.0	7.5	T
	KO5	98.8	0.0	0.0	1.2	T

Table S3: SVM Model performance for classification of pure (KO) and adulterants (PO, O3C and FO) using FT-Raman, FT-IR and Fused data

	FT-Raman	**Test set (76% accuracy)**			
Confusion matrix	Actual	1	2	3	4
Predicted		KO	PO	O3C	FO
1	KO	0	0	0	2
2	PO	4	18	0	4
3	O3C	0	0	17	0
4	FO	0	0	7	19
	FT-IR	**Test set (79% accuracy)**			
Confusion matrix	Actual	1	2	3	4
Predicted		KO	PO	O3C	FO
1	KO	2	4	2	4
2	PO	2	13	0	2
3	O3C	0	0	22	1
4	FO	0	0	0	18
	Fused data	**Test set (84% accuracy)**			
Confusion matrix	Actual	1	2	3	4
Predicted		KO	PO	O3C	FO
1	KO	4	2	3	4
2	PO	0	16	0	1
3	O3C	0	0	20	0
4	FO	0	0	1	19

Table S4: SVM model performance with accuracy, sensitivity and specificity test of pure (KO) and adulterants (PO, O3C and FO) using FT-Raman, FT-IR and Fused data

Technique	Training set		Test set								
	Accuracy (calibration)	Accuracy (cross validation)	Accuracy	Sensitivity to individual classes (%)				Specificity to individual classes (%)			
				KO	PO	O3C	FO	KO	PO	O3C	FO
FT-Raman	100	86	76	0	100	71	76	97	85	100	85
FT-IR	100	96	79	50	76	92	72	85	92	98	100
Fused data	100	92	84	100	89	83	79	86	98	100	98