1 Colocalization features for classification of tumors using desorption electrospray ionization mass spectrometry imaging

Paolo Inglese*, Gonçalo Correia◊, Pamela Pruski◊, Robert C Glen, Zoltan Takats*

Computational and Systems Medicine, Department of Surgery and Cancer, Faculty of Medicine, Imperial College London, London SW7 2AZ, UK (UK)

◊Contributed equally

*Correspondence to: p.inglese14@imperial.ac.uk, z.takats@imperial.ac.uk

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1 SUPPLEMENTARY METHODS

1.1 Co-localization features extraction

Let $\boldsymbol{I}_t$ be a set of mass spectral intensities associated with a tissue section denoted by the subscript t. Given the spatial dimensions n_x and n_y of t in number of pixels, and the *m/z* space dimension n_{mz}, then $\boldsymbol{I}_t$ can be defined as a 3-dimensional matrix

$$\boldsymbol{I}_t = (y_{ijk}), \quad i \in \{1,...,n_x\}, j \in \{1,..., n_y\}, k \in \{1,..., n_{mz}\} \tag{1}$$

where $y_{ijk} \in \mathbb{R}_+$ represents the k-th spectral peak intensity in the spatial location (i,j). Using this definition, the 2-dimensional sections of $\boldsymbol{I}_t$ corresponding to a fixed value of k, denoted $y_{..k}$, can be interpreted as images representing of the spatial distribution of the k-th spectral peak.

Since $\boldsymbol{I}_t$ may contain spatial regions unrelated with the sample-of-interest, the subset of pixels $ROI_t = (i,j) \subseteq \{1,...,n_x\} \times \{1,...,n_y\}$ associated with the tissue section are extracted by applying the 'kmeans2' method from the SPUTNIK package for R (Inglese, Correia, *et al.*, 2018) (Supplementary Table S3). The tissue-related set of spectral intensities are denoted as

$$\boldsymbol{I}_t^{ROI} = (y_{ijk}), \quad (i,j) \in ROI_t, k \in \{1,..., n_{mz}\} \tag{2}$$

A co-localization features vector of $\boldsymbol{I}_t$ is therefore defined as the s-tuple

$$\boldsymbol{f}_t = \left(f_1^{(t)}, f_2^{(t)}, f_3^{(t)},..., f_s^{(t)}\right) \tag{3}$$

consisting of the Spearman's correlations between all the pairs of vectorized image pixels $y_{i'j'a}, y_{i'j'b} \subset \boldsymbol{I}_t^{ROI}$,

$$\begin{gathered} f_p^{(t)} = Spearman(Vectorize(y_{i'j'a}), Vectorize(y_{i'j'b})), \\ p \in \{1, ...,s\}, \ a, b \in \{1,...,n_{mz}\}, \\ i'j' \in \mathcal{R} \subseteq ROI_t \end{gathered} \tag{4}$$

such that $a > b$ (or, equivalently, $a < b$). *Vectorize* is a function that concatenates the 2-dimensional spectral peak intensities column-wise, and $\mathcal{R}$ represents a set of randomly sampled ROI pixels, with $|\mathcal{R}| = N_{pix}$.

Spearman is the Spearman's rank correlation, defined as the Pearson's correlation between the ranked variables

$$Spearman(\boldsymbol{x},\boldsymbol{y}) = \frac{\mathrm{cov}(rank(\boldsymbol{x}), rank(\boldsymbol{y}))}{\mathrm{st.dev}(rank(\boldsymbol{x})) \times \mathrm{st.dev}(rank(\boldsymbol{y}))} \quad (5)$$

The correlations were selected to avoid duplicated values, due to its symmetricity under the exchange of the *m/z* indices.

In this way, the vector $\boldsymbol{f}_t$ consists of $s = n_{mz} \times (n_{mz} - 1)/2$ elements belonging to the interval $[-1,1]$. The asymptotic *t* approximation (Hollander and Wolfe, 1999) is applied to determine the significance of each Spearman's correlation $f_p^{(t)}$. The image correlations associated with a Benjamini-Hochberg corrected p-value (number of tests equal to s) larger than 0.05 are set equal to 0.

1.2 Cross-validation scheme

Before fitting the supervised models, the co-localization features are extracted using a randomly sampled number of pixels N_{pix} belonging to the tissue-related ROI.

The 48 MS images belonging to the *cross-validation set* are split into 10 disjoint subsets of almost equal size (their cardinalities must sum to 48) that will represent the validation sets of the 10 rounds of the cross-validation. The co-localization features and the tissue labels of the MS images that do not belong to the k-th validation set represent the training set used to train a PLS-DA model. Therefore, the fitted model is used to predict the tissue class of the MS images belonging to the k-th validation set. The performance metrics are calculated comparing the true and predicted tissue labels after each round of the cross-

validation. At the end of the procedure, the performances are summarized by the average values of the metrics calculated over the 10 rounds. Before fitting the PLS-DA model, all the constant co-localization features in the training set are removed from both training and validation set.

The cross-validation is repeated with a PLS-DA model fitted using a varying number of components in the range of 2 to 10.

The number of the pixels N_{pix} used to extract the co-localization features vary from 100 to 1000 in steps of 100 pixels.

To estimate the effect of the randomized selection of the tissue-related pixels used for the extraction of the co-localization features and the split of the 48 MS images into training and validation set, the entire procedure is repeated 500 times.

The final performances of the PLS-MODELS are summarized by averaging the metrics values of the repeated cross validations for a fixed number of PLS-DA components and number of pixels N_{pix} used to extract the co-localization features.

The optimal number of PLS-DA components K^* and the optimal number of sampled pixels N_{pix}^* are determined by

$$\underset{\substack{K \in \{2,\ldots,20\} \\ N_{pixels} \in \{100,200,\ldots,1000\}}}{\arg\max} \left(\frac{1}{500} \sum_{i=1}^{500} \{ accuracy_i^{(10\text{ - fold})}(N_{pix},K) \} \right) \qquad (6)$$

where $accuracy_i^{(10\text{ - fold})}(N_{pix},K)$ represents the summary accuracy value of the i-th repetition of the 10-fold cross-validation using N_{pix} pixels to extract the co-localization features, and K PLS-DA components.

1.3 External test performance

A PLS-DA model with K^* components, fitted on the co-localization features extracted from N_{pix}^* (Equation 5) of all the 48 MS images belonging to the *cross-validation set*, is used to predict the labels of the external *test set* MS images.

The procedure is repeated 500 times, with different randomly sampled N_{pix}^* pixels.

The final performance metrics are summarized by averaging their values over 500 repetitions.

1.4 Classification using spectral intensity features

In order to compare the performances of the co-localization features with the standard spectral peak intensities, an analogous classification scheme (Supplementary Methods 1.2) was applied to the pixel mass spectral vectors.

The mass spectral intensities vectors associated with the same randomly sampled pixels used to calculate the co-localization features of the training MS images were used as training features for a PLS-DA model.

The performances of the models were measured comparing the true and predicted tissue labels of the test MS images pixels spectra. After the rounds of the 10-fold cross-validation, the performance metrics were summarized by averaging over the 10 rounds.

The entire procedure followed the identical scheme used with the co-localization features.

1.5 Co-localization features variance estimation

Let $\boldsymbol{I}_t$ be the generic MS image belonging to the *cross-validation set*, with $t \in \{1, 2, ..., 48\}$. Let $\boldsymbol{f}_t^{(r)} = \left(f_1^{(t)(r)}, f_2^{(t)(r)}, ..., f_s^{(t)(r)}\right)$ represent the its co-localization feature vector (Equation 3) extracted in the repetition $r \in \{1, 2, ..., 500\}$.

The variance of the co-localization vector estimated over the 500 repetitions is the vector of the variances of the co-localization features

$$Var(\boldsymbol{f}_t) = \left(Var\left(f_1^{(t)(.)}\right), Var\left(f_2^{(t)(.)}\right), \ldots, Var\left(f_s^{(t)(.)}\right)\right) \quad (7)$$

The median of the components of $Var(\boldsymbol{f}_t)$ and its median absolute deviation (MAD) are reported in Figure 2 of the main text.

2 SUPPLEMENTARY DATA

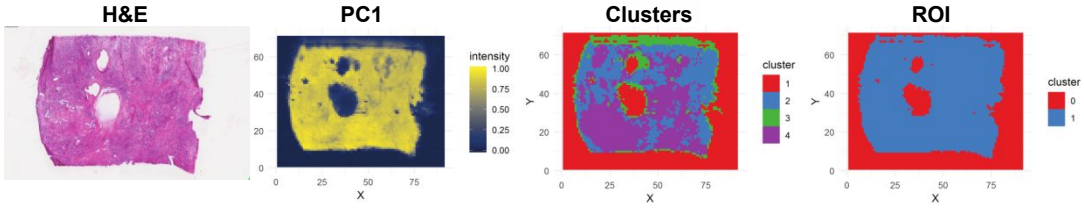


Figure S 1 – Example of tissue-related ROI. K-Means clustering with a number of clusters equal to 4 is applied to the MSI intensity matrix. The clusters that are not localized in the off-tissue area (corners used as reference) are merged into the final tissue-related ROI. A comparison with the first principal component scores (PC1) image and the optical image of the H&E stained tissue shows the close correspondence between the ROI and the area occupied by the tissue.

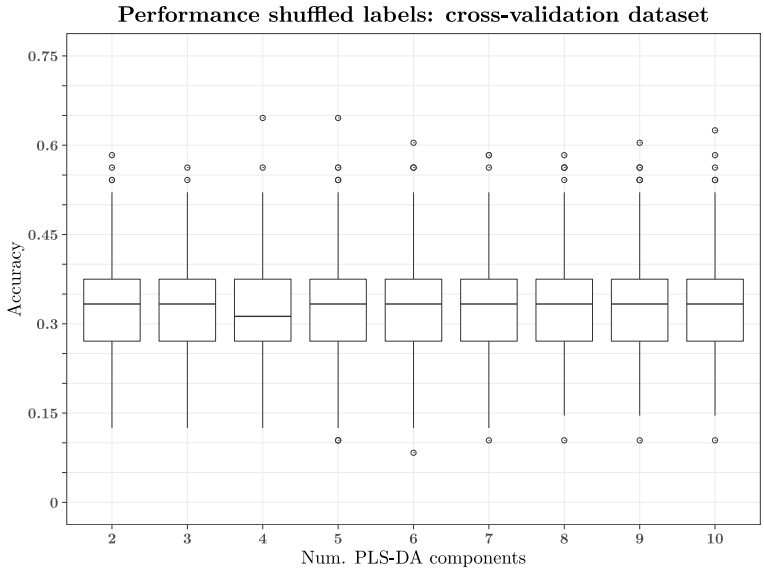


Figure S 2 – Performances of the PLS-DA models with shuffled tissue labels. The bar plots represent the accuracy values of the 500 repetitions using the optimal number of randomly sampled pixels equal to 900, and PLS-DA components varying from 2 to 10. The accuracy values confirm that the observed performances in the original PLS-DA model are not due to random chance.

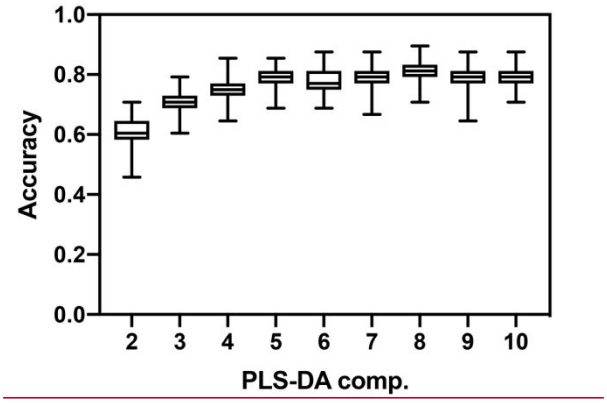


Figure S 3 - Performances of the PLS-DA models on the cross-validation set, using the mean peak intensities as features.

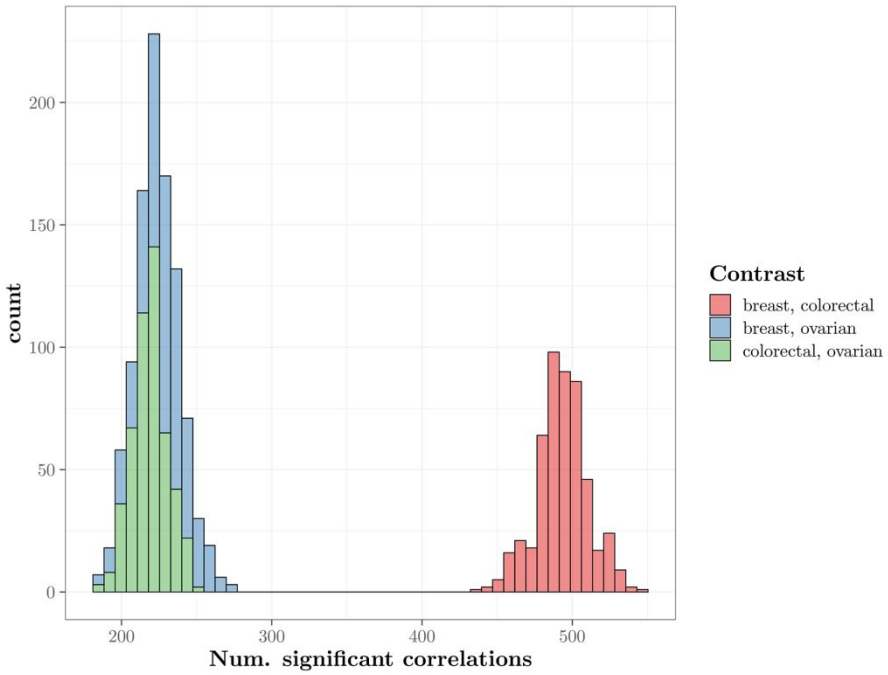
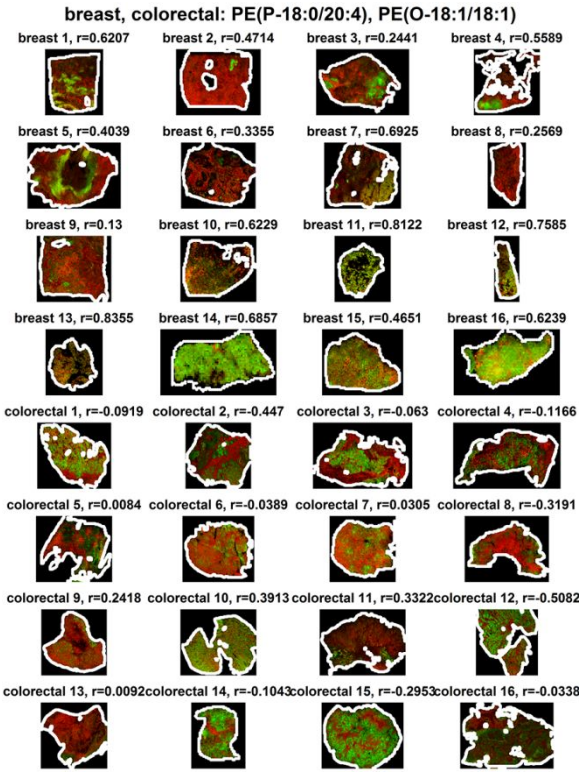


Figure S 4 - Histogram representing the number of significant correlations for the three contrasts, in the 500 repetitions.



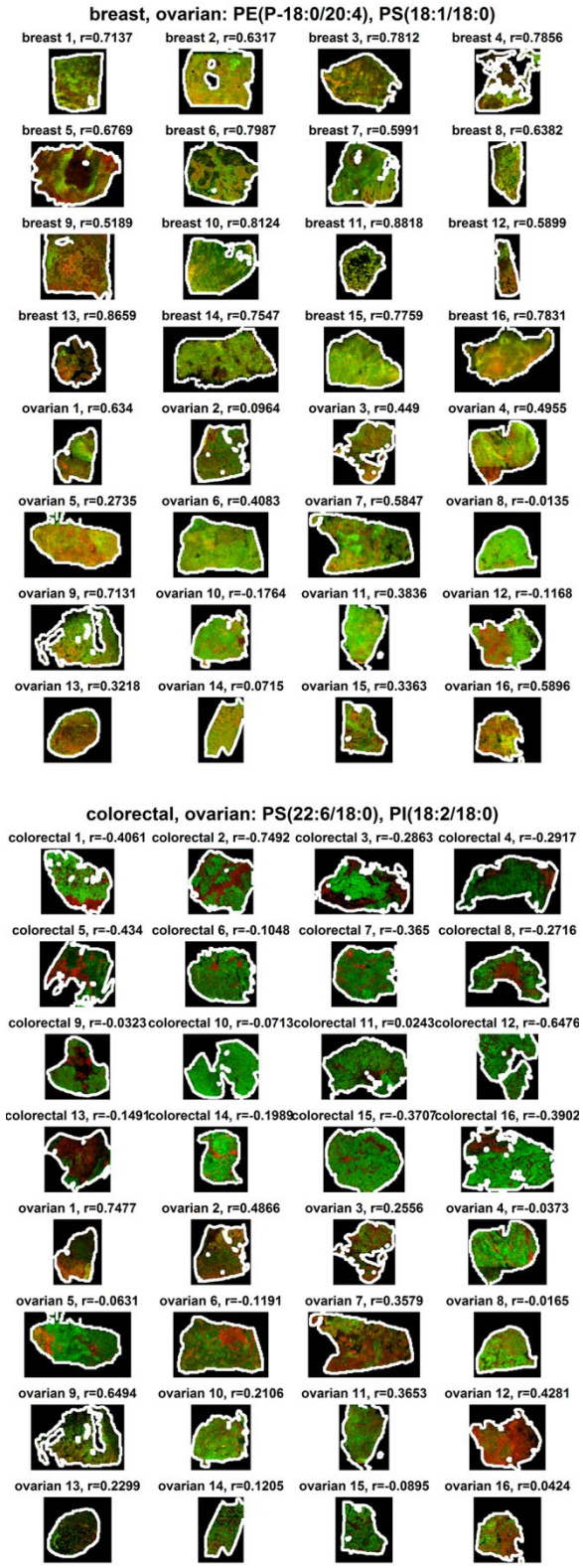
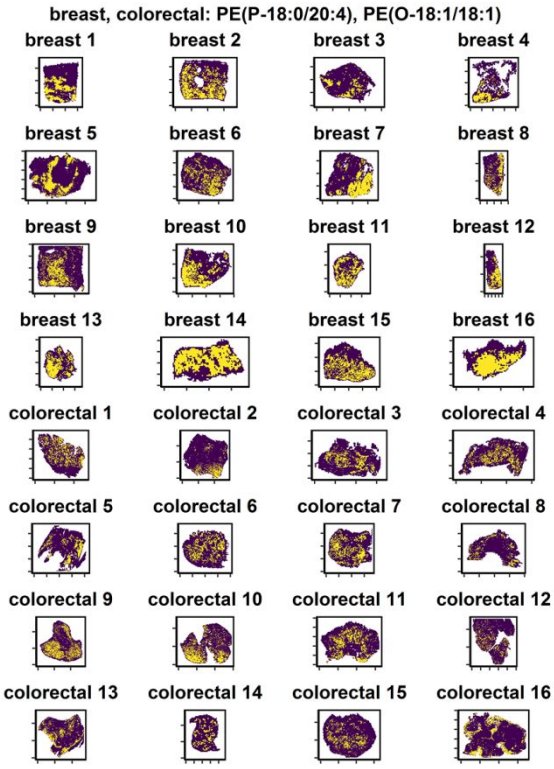


Figure S 5 – Spatial distribution of the spectral peaks associated with the most significantly different co-localization feature in the three contrasts. The relative abundances, scaled in the interval [0, 1], are represented as the red and green channels of an RGB image. The value of the correlation is reported on top of each image.


breast, colorectal: PE(P-18:0/20:4), PE(O-18:1/18:1)
breast 1
breast 2
breast 3
breast 4
breast 5
breast 6
breast 7
breast 8
breast 9
breast 10
breast 11
breast 12
breast 13
breast 14
breast 15
breast 16
colorectal 1
colorectal 2
colorectal 3
colorectal 4
colorectal 5
colorectal 6
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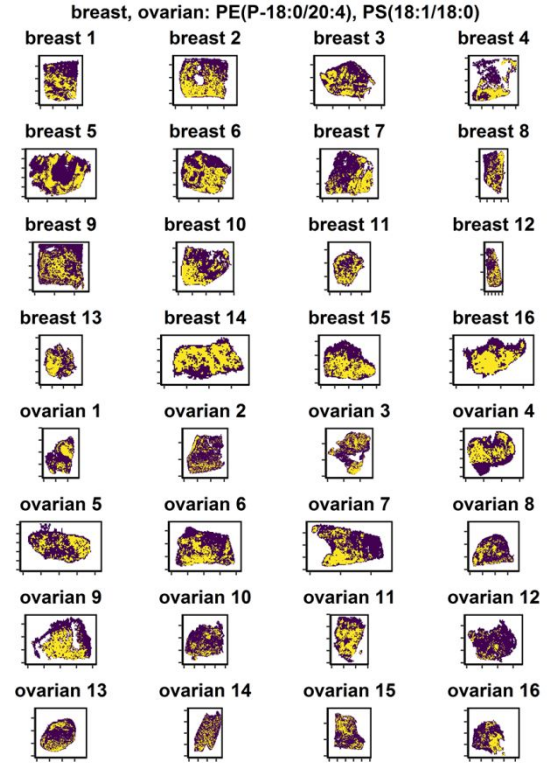
colorectal 8
colorectal 9
colorectal 10
colorectal 11
colorectal 12
colorectal 13
colorectal 14
colorectal 15
colorectal 16


breast, ovarian: PE(P-18:0/20:4), PS(18:1/18:0)
breast 1
breast 2
breast 3
breast 4
breast 5
breast 6
breast 7
breast 8
breast 9
breast 10
breast 11
breast 12
breast 13
breast 14
breast 15
breast 16
ovarian 1
ovarian 2
ovarian 3
ovarian 4
ovarian 5
ovarian 6
ovarian 7
ovarian 8
ovarian 9
ovarian 10
ovarian 11
ovarian 12
ovarian 13
ovarian 14
ovarian 15
ovarian 16

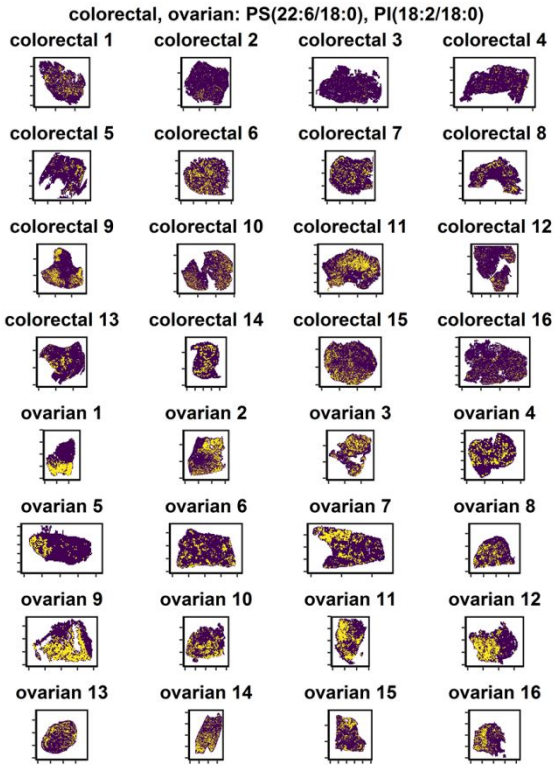


Figure S 6 – Clustering of the relative abundances of the ion pairs associated with the most significantly different correlations in the three contrasts. The yellow pixels represent the areas where the relative abundance of the two ions were higher than their average value.

Table S 1 – List of DESI-MS parameters used for the acquisition of the imaging data of the three sets of tissue sections. The columns represent the tissue section thickness, the lateral spatial resolution of the imaging data, the solvent flow rate and the nitrogen gas pressure used for running DESI-MS, respectively.

ID	Thickness (µm)	Resolution (µm)	Flow rate (µl/min)	Pressure (bar)
Breast Samples				
0502202A	15	100	1.5	7
502263	15	100	1.5	7
602115	15	100	1.5	7
602167	15	100	1.5	7
702006	15	100	1.5	7
702233A	15	100	1.5	7
702273	15	100	1.5	7
802086A	15	100	1.5	7
802157A	15	100	1.5	7
802183	15	100	1.5	7
802186	15	100	1.5	7
0902103A	15	100	1.5	7
1102011A	15	100	1.5	7
1102052A	15	100	1.5	7

1102116	15	100	1.5	7
1102138	15	100	1.5	7
BRB01S	15	100	1.5	7
BRB03N	15	90	1.5	7
BRB04N	15	65	1.5	7
BRB06E	15	100	1.5	7
BRB12S	15	100	1.5	7
BRB13D	15	100	1.5	7
CX12035	15	100	1.5	7
CX12068	15	100	1.5	7
CX12230	15	100	1.5	7
CXH12000200	15	100	1.5	7
CXH12000251	15	100	1.5	7
Colorectal Samples				
A15	10	100	1.5	7
A20	10	110	1.5	7
A22	10	100	1.5	7
A26	10	100	1.5	7
A28	10	100	1.5	7
A33	10	100	1.5	7
A37	10	85	2.5	7
A49	10	115	1.5	7
A52	10	80	1.5	7
A53	10	90	1.5	5
A56	10	125	1.5	5
A57	10	90	1.5	5
A62	10	100	1.5	7
A64	10	85	1.5	5
A74	10	100	1.5	7
Ovarian Samples				
All	10	100	1.5	7

Table S 2 – List of parameters used for MALDIquant peak matching, within the MS image and between MS images, respectively.

	Method	**Tolerance**	**Min. Freq. Threshold**
Within MS image	strict	10 ppm	0.5%
Between MS images	strict	20 ppm	100%

Table S 3 – List of SPUTNIK parameters used for filtering the tissue-unrelated peaks.

Global similarity filter	
Reference	Binary ROI (k-means2)
Method	Pearson's correlation
Threshold	0
Pixel count filter	
Reference	Binary ROI (k-means2)
Min. number of connected pixels	9
Aggressiveness	1

Table S 4 – Annotations of the common m/z values detected after peak matching. Annotation based on MS/MS are reported in the last column: YES, MS/MS was used to identify the molecule, YES (N/A), the molecule could not be identified by its MS/MS spectrum; NO, the molecule was identified using only the m/z value.

Obs. *m/z*	Theor. *m/z*	Error (ppm)	Sum formula	Adduct	Compound	MS/MS
259.2422	259.2431	3.4	C19H32	M-H-	Nonadecatetraene	YES (N/A)
279.2321	279.233	3.3	C18H32O2	M-H-	Linoleic acid (C18:2)	YES (N/A)
303.2321	303.233	2.9	C20H32O2	M-H-	Eicosatetraenoic acid (C20:4)	YES (N/A)
305.2478	305.2486	2.6	C20H34O2	M-H-	Eicosatrienoic acid (C20:3)	YES (N/A)
307.2635	307.2643	2.7	C20H36O2	M-H-	Eicosadienoic acid (C20:2)	YES (N/A)
309.2791	309.2799	2.5	C20H38O2	M-H-	Eicosenoic acid (C20:1)	YES (N/A)
327.2322	327.233	2.5	C22H32O2	M-H-	Docosatriynoic acid (C22:3)	NO
331.2635	331.2643	2.4	C22H36O2	M-H-	Docosatetraenoic acid (C22:4)	YES (N/A)
365.3417	365.3425	2.3	C24H46O2	M-H-	Nervonic acid (C24:1)	YES (N/A)
419.2562	419.2568	1.4	C21H41O6P	M-H-	Cyclic Phosphatidic acid (C18:0)	YES
437.2666	437.2674	1.7	C21H43O7P	M-H-	LysoPA(18:0)	YES
464.3139	464.3146	1.6	C23H48NO6P	M-H-	PE(P-18:0)	YES (N/A)
480.3089	480.3096	1.4	C23H48NO7P	M-H-	LysoPE(18:0)	YES
536.5043	536.5048	0.9	C34H67NO3	M-H-	N-Palmitoylsphingosine / Cer(d18:1/16:0)	YES
646.6142	646.6144	0.3	C42H81NO3	M-H-	Ceramide (d18:1/24:1)	YES
673.4812	673.4814	0.4	C37H71O8P	M-H-	PA(18:1/14:0) / PA(16:0/16:1)	YES
687.5444	687.5446	0.2	C38H77N2O6P	M-H-	SM(d33:1) / PE-Cer(d36:1)	YES (N/A)
698.513	698.513	0	C39H74NO7P	M-H-	PE(P-16:0/18:2)	YES
699.497	699.497	0	C39H73O8P	M-H-	PA(18:1/18:1) / PA(18:0/18:2)	YES
700.5286	700.5287	0.2	C39H76NO7P	M-H-	PE(P-14:0/18:1)	YES
701.5126	701.5127	0.1	C39H75O8P	M-H-	PA(36:1)	NO
714.5079	714.5079	0	C39H74NO8P	M-H-	PE(14:0/18:2)	YES
716.5236	716.5236	0	C39H76NO8P	M-H-	PE(14:0/18:1)	YES
718.5392	718.5392	0	C39H78NO8P	M-H-	PE(16:0/16:0)	YES
722.5131	722.513	-0.1	C41H74NO7P	M-H-	PE(P-16:0/20:4)	YES
723.4968	723.497	0.3	C41H73O8P	M-H-	PA(18:0/20:4) / PA(18:2/18:2)	YES
725.5128	725.5127	-0.1	C41H75O8P	M-H-	PA(18:0/20:3)	YES
726.5445	726.5443	-0.3	C41H78NO7P	M-H-	PE(O-18:2/18:1)	YES
728.5601	728.56	-0.1	C41H80NO7P	M-H-	PE(O-18:1/18:1)	YES
738.5082	738.5079	-0.4	C41H74NO8P	M-H-	PE(16:0/20:4)	YES
740.5240	740.5236	-0.5	C41H76NO8P	M-H-	PE(18:1/18:2)	YES
742.5394	742.5392	-0.3	C41H78NO8P	M-H-	PE(18:1/18:1)	YES
744.5551	744.5549	-0.3	C41H80NO8P	M-H-	PE(18:0/18:1)	YES
746.5139	746.513	-1.2	C43H74NO7P	M-H-	PE(P-16:0/22:6)	YES
748.5300	748.5287	-1.8	C43H76NO7P	M-H-	PE(P-18:1/20:4)	YES
750.5443	750.5443	0.1	C43H78NO7P	M-H-	PE(P-18:0/20:4)	YES

752.5603	752.56	-0.4	C43H80NO7P	M-H-	PE(P-18:0/20:3)	YES
762.5083	762.5079	-0.5	C43H74NO8P	M-H-	PE(16:0/22:6)	YES
764.5237	764.5236	-0.1	C43H76NO8P	M-H-	PE(18:1/20:4)	YES
766.5393	766.5392	-0.1	C43H78NO8P	M-H-	PE(18:0/20:4)	YES
768.5554	768.5549	-0.7	C43H80NO8P	M-H-	PE(18:0/20:3)	YES
769.5026	769.5025	-0.2	C42H75O10P	M-H-	PG(18:2/18:2)	YES
770.5708	770.5705	-0.4	C43H82NO8P	M-H-	PE(18:1/20:1) / PE(18:0/20:1)	YES
771.5183	771.5182	-0.2	C42H77O10P	M-H-	PG(18:1/18:2)	YES
772.5867	772.5862	-0.7	C43H84NO8P	M-H-	PE(38:1)	NO
773.5337	773.5338	0.2	C42H79O10P	M-H-	PG(18:1/18:1)	YES
774.5472	774.5443	-3.8	C45H78NO7P	M-H-	PE(P-18:0/22:6)	YES
776.5619	776.56	-2.4	C45H80NO7P	M-H-	PE(P-18:0/22:5)	YES
778.5761	778.5756	-0.6	C45H82NO7P	M-H-	PE(P-18:0/22:4) / PE(P-20:0/20:4)	YES
786.5291	786.5291	0	C42H78NO10P	M-H-	PS(18:2/18:0)	YES
788.5448	788.5447	-0.2	C42H80NO10P	M-H-	PS(18:1/18:0)	YES
792.5553	792.5549	-0.5	C45H80NO8P	M-H-	PE(18:0/22:5)	YES
794.5709	794.5705	-0.6	C45H82NO8P	M-H-	PE(18:0/22:4)	YES
810.5291	810.5291	0	C44H78NO10P	M-H-	PS(20:4:18:0)	YES
812.5454	812.5447	-0.8	C44H80NO10P	M-H-	PS(20:3/18:0)	YES
816.5768	816.576	-1	C44H84NO10P	M-H-	PS(18:1/20:0)	YES
820.5626	820.5629	0.4	C44H84NO8P	M+Cl-	PC(18:1/18:1)	YES
833.5188	833.5186	-0.2	C43H79O13P	M-H-	PI(18:2/16:0)	YES
834.5308	834.5291	-2	C46H78NO10P	M-H-	PS(22:6/18:0)	YES
857.5186	857.5186	0	C45H79O13P	M-H-	PI(20:4/16:0)	YES
859.5352	859.5342	-1.2	C45H81O13P	M-H-	PI(36:3)	NO
861.5501	861.5499	-0.3	C45H83O13P	M-H-	PI(18:2/18:0)	YES
883.5344	883.5342	-0.2	C47H81O13P	M-H-	PI(20:4/18:1)	YES
885.5501	885.5499	-0.2	C47H83O13P	M-H-	PI(20:4/18:0)	YES

Table S 5 – List of the five largest correlated pairs of ions in the three tissue types.

Ion 1	Ion 2	Correlation
Breast		
PA(36:1)	PS(18:1/18:0)	0.91964294
PG(18:1/18:2)	PG(18:1/18:1)	0.85387172
PE(P-18:0/20:4)	PE(P-18:1/20:4)	0.82841959
PE(P-18:1/20:4)	PE(P-18:0/20:4)	0.82781521
PE(P-16:0/20:4)	PE(P-18:0/20:4)	0.81827409
Colorectal		
PA(36:1)	PS(18:1/18:0)	0.89393631
PE(O-18:2/18:1)	PE(O-18:1/18:1)	0.84356314
PI(18:2/16:0)	PI(18:2/18:0)	0.83477204
PE(18:1/18:1)	PE(18:0/18:1)	0.79990005
PE(18:1/18:2)	PE(18:1/18:1)	0.79349843
Ovarian		
PA(36:1)	PS(18:1/18:0)	0.87004032
PG(18:1/18:2)	PG(18:1/18:1)	0.7705973
PE(18:1/18:1)	PE(18:0/18:1)	0.75112221
PE(18:0/18:1)	PE(18:1/20:1) / PE(18:0/20:1)	0.73529541
PE(18:1/20:1) / PE(18:0/20:1)	PC(18:1/18:1)	0.71865099

Table S 6 –Statistically significant different ion pair correlations obtained for the three contrasts are reported with the sum formulas corresponding to the m/z difference between the selected ions.

Molecule 1	Molecule 2	Sum formula	m/z 1	m/z 2	Δ m/z	Δ Sum formula
Breast - Colorectal						
PE(P-16:0/20:4)	PE(O-18:1/18:1)	C41H74NO7P , C41H80NO7P	722.513	728.56	6.047	(+H6 (3 double bonds))
PE(P-14:0/18:1)	PE(P-18:0/20:4)	C39H76NO7P, C43H78NO7P	700.5287	750.5443	50.0156	(+C4H2)
PE(18:0/18:1)	PE(18:0/22:5)	C41H80NO8P, C45H80NO8P	744.5549	792.5549	48	(+C4)
PE(18:0/22:4)	PC(18:1/18:1)	C45H82NO8P, C44H84NO8P	794.5705	820.5629	25.9924	(+CH2)
PS(18:1/20:0)	PS(22:6/18:0)	C44H84NO10P, C46H78NO10P	816.576	834.5291	17.9531	(+C2 –H6)
C20:4	PE(O-18:1/18:1)	C20H32O2, C41H80NO7P	303.233	728.56	425.327	(+C21H48NO5P)
PE(O-18:1/18:1)	PS(22:6/18:0)	C41H80NO7P, C46H78NO10P	728.56	834.5291	105.9691	(+C5O3 –H2)
PE(18:0/18:1)	PE(18:0/22:4)	C41H80NO8P, C45H82NO8P	744.5549	794.5705	50.0156	(+C4H2)
PE(18:0/18:1)	PE(18:0/20:4)	C41H80NO8P, C43H78NO8P	744.5549	766.5392	21.9843	(+C2 –H2)
PE(O-18:1/18:1)	PE(P-18:1/20:4)	C41H80NO7P, C43H76NO7P	728.56	748.5287	19.9687	(+C2 –H2)
Breast - Ovarian						
PE(P-16:0/20:4)	PS(18:1/18:0)	C41H74NO7P, C42H80NO10P	722.513	788.5447	66.0317	(+CH6O3)
PA(36:1)	PE(P-18:0/20:4)	C39H75O8P, C41H74NO7P	701.5127	722.513	21.0003	(+C2 -OH)
PE(P-18:0/20:4)	PE(P-18:0/20:3)	C41H74NO7P, C43H80NO7P	722.513	752.56	30.047	(+C2H6)
PE(P-14:0/18:1)	PE(P-18:1/20:4)	C39H76NO7P, C43H76NO7P	700.5287	748.5287	48	(+C4)
PE(P-18:0/20:4)	PE(P-18:1/20:4)	C41H74NO7P, C43H76NO7P	722.513	748.5287	26.0157	(+C2H2)
PS(18:1/18:0)	PI(20:4/18:0)	C42H80NO10P, C47H83O13P	788.5447	885.5499	97.0052	(+C5H3NO3)
PE(P-18:1/20:4)	PI(20:4/16:0)	C43H76NO7P, C45H79O13P	748.5287	857.5186	108.9899	(+C2H3NO6)
PE(P-16:0/22:6)	PS(18:1/18:0)	C43H74NO7P, C42H80NO10P	746.513	788.5447	42.0317	(-C +H6O3)
PA(36:1)	PI(20:4/18:0)	C39H75O8P, C47H83O13P	701.5127	885.5499	184.0372	(+C8H8O5)
PA(36:1)	PE(P-18:0/20:4)	C39H75O8P, C43H78NO7P	701.5127	750.5443	49.0316	(+C4H2 -O)
Colorectal – Ovarian						
PS(22:6/18:0)	PI(18:2/18:0)	C46H78NO10P, C45H83O13P	834.5291	861.5499	27.0208	(-CN /+H5O3)
C22:4	PE(P-18:0)	C22H36O2, C23H48NO7P	331.2643	464.3146	133.0503	(+CH12O5P)
PS(20:3/18:0)	PI(18:2/18:0)	C44H80NO10P, C45H83O13P	812.5447	861.5499	49.0052	(+CH3O3 /-N)
Cer(d18:1/16:1)	PE(18:0/22:4)	C34H67NO3, C45H82NO8P	536.5048	794.5705	258.0657	(+C11H15O5P)
PS(20:3/18:0)	PI(18:2/16:0)	C44H80NO10P, C43H79O13P	812.5447	833.5186	20.9739	(+NO3/ -CH)
PE(P-18:0)	PS(22:6/18:0)	C23H48NO6P, C46H78NO10P	464.3146	834.5291	370.2145	(+C23H30O4)

PE(16:0/16:0)	PI(20:4/18:0)	C39H78NO8P, C47H83O13P	718.5392	885.5499	167.0107	(+C8H5O5 / -N)
C22:4	PI(18:2/18:0)	C22H36O2, C45H83O13P	331.2643	861.5499	530.2856	(+C23H47O11P)
PE(18:0/20:4)	PI(18:2/16:0)	C43H78NO8P, C43H79O13P	766.5392	833.5186	66.9794	(+HO5 /-N)
PE(P-18:0)	PA(18:0/20:4) / PA(18:2/18:2)	C23H48NO6P, C41H73O8P	464.3146	723.497	259.1824	(+C18H25O2/ -N)