Electronic Supplementary Material (ESI) for Nanoscale Horizons.

Supplementary Text and Figures

Table S1. General information on patients and their cancer types whose plasma was used in this study.

PATIENT LABEL	AGE	GENDER	CANCER STAGE	CANCER STAGE CATEGORY
PANCREAS 1	81	M	TNM: cT4 N+M+	Advanced
PANCREAS 2	76	M	TNM: cT2 N+	Moderate
PANCREAS 3	60	F	TNM: cT4 N+	Advanced
PANCREAS 4	75	M	TNM: cT3 N+M+	Advanced
PANCREAS 5	61	F	TNM: cT3 N+M+	Advanced
LUNG 1	45	F	TNM: TIA N0 M0	Early
LUNG 2	24	M	TNM: TIA N0 M0	Early
LUNG 3	44	F	TNM: TIA N0 M0	Early
LUNG 4	48	F	TNM: TIA N0 M0	Early
LUNG 5	40	M	TNM: TIA N0 M0	Early
MYELOMA 1	41	F	Onset II (52% Plasmacells in Bone Marrow; Monoclonal IgG-k)	Moderate
MYELOMA 2	60	F	Onset I (28% Plasmacells in Bone Marrow; Monoclonal IgG-k)	Early
MYELOMA 3	57	F	Onset I (8% Plasmacells in Bone Marrow; Monoclonal IgG-k)	Early
MYELOMA 4	63	M	Onset I (9% Plasmacells in Bone Marrow; Monoclonal IgG-k)	Early
MYELOMA 5	75	M	Onset II (42% Plasmacells in Bone Marrow; Monoclonal IgG-k)	Moderate
GLIOBLASTOMA 1	58	M	WHO 4	Advanced
GLIOBLASTOMA 2	75	M	WHO 4	Advanced
GLIOBLASTOMA 3	76	M	WHO 4	Advanced
GLIOBLASTOMA 4	73	M	WHO 4	Advanced
GLIOBLASTOMA 5	76	F	WHO 4	Advanced
MENINGIOMA 1	82	F	WHO 2	Moderate
MENINGIOMA 2	50	F	WHO 2	Moderate
MENINGIOMA 3	80	M	WHO 2	Moderate
MENINGIOMA 4	67	M	WHO 2	Moderate
MENINGIOMA 5	64	M	WHO 2	Moderate

Table S2. Classification results obtained from two developed linear and nonlinear models for six groups of samples and also three groups of cohort samples (10-fold cross validation is repeated 10 times and the reported model performance statistics are averaged).

Models/ classification parameters		Fresh samples		Cohort samples				
		PLS-DA, LV=5	CPANN (8*8)		PLS-DA, 69 variables LV=3	CPANN 69 variables (8*8)	PLS-DA 8 variables LV=2	CPANN 8 variables (8*8)
Specificity	**1**	1.00	1.00	**1**	0.591	0.57	1.00	1.00
	2	0.96	0.984					
	3	1.00	0.996	**2**	0.760	0.86	1.00	1.00
	4	1.00	1.00					
	5	1.00	1.00	**3**	1.00	0.96	1.00	1.00
	6	1.00	1.00					
Sensitivity	**1**	1.00	0.98	**1**	0.751	0.72	1.00	1.00
	2	1.00	0.98					
	3	1.00	0.94	**2**	0.850	0.32	1.00	1.00
	4	1.00	1.00					
	5	1.00	1.00	**3**	0.540	0.74	1.00	1.00
	6	1.00	1.00					
Class error	**1**	0.00	0.033	**1**	0.329	0.38	0.00	0.00
	2	0.03	0.016					
	3	0.00	0.013	**2**	0.201	0.32	0.00	0.00
	4	0.00	0.00					
	5	0.00	0.00	**3**	0.131	0.11	0.00	0.00
	6	0.00	0.00					

1) Control 2) Glioblastoma 3) Meningioma 4) Myeloma 5) Pancreas 6) Lung cancers

Cohort samples: 1) Brain 2) Lung 3) Pancreas

LV (Latent variables): Partial Least Square defines a low dimensional space based on small number of orthogonal factor which called latent factors or variables. Each factor is a linear combination of the original variables. PLS extracts these latent factors, accounting for as much of the variation in data space as possible while modeling the responses properly.

CV (Cross Validation): Cross-validation is a method of assessing generalization performance of statistical models like classifiers. One of the most popular approaches to do CV is the holdout method. This could be done by splitting the original dataset into complementary subsets and performing the analysis using one subset (training set), and validating the results using the leaved-out subset (validation set).

Sensitivity= TP/(TP+FN),

Specificity= TN/(TN+FP)

Class error= 1-accuracy = (FP+FN)/(TP+TN+FP+FN)

TP: true positive; TN: true negative; FP: false positive; FN: false negative

Table S3. Correlation coefficient of CPANN weight map for each variable in the six classes.

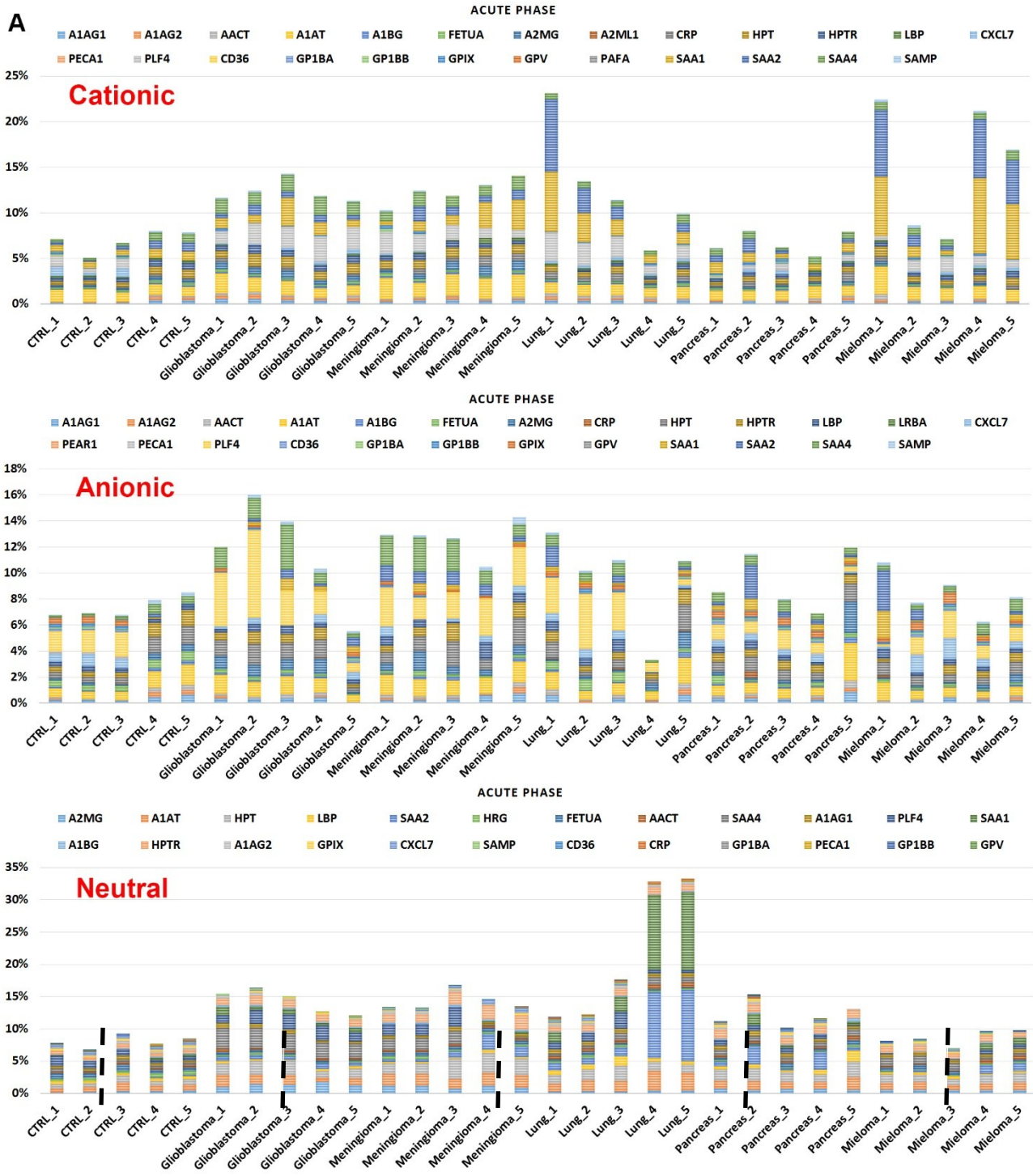
Variable ID	CC(1), Control	CC(2) Glioblastoma	CC(3) Meningioma	CC(4) Myeloma	CC(5) Pancreas	CC(6) Lung
1	0.60	-0.31	-0.39	-0.37	-0.13	0.46
2	-0.23	-0.13	-0.22	-0.18	0.92	-0.11
3	-0.20	-0.10	-0.27	-0.15	0.93	-0.16
4	0.38	-0.06	-0.21	-0.72	-0.03	0.58
5	-0.38	-0.23	-0.36	0.33	0.05	0.70
6	-0.01	-0.36	-0.58	0.42	0.01	0.56
7	-0.37	-0.32	-0.39	0.66	0.02	0.50
8	-0.15	-0.19	-0.32	0.02	0.94	-0.25
9	-0.45	0.11	-0.13	0.87	-0.22	-0.03
10	-0.51	-0.22	-0.15	0.55	0.57	-0.13
11	-0.33	-0.15	-0.24	0.65	-0.45	0.61
12	-0.34	-0.24	-0.43	0.36	0.02	0.72
13	0.18	0.40	0.38	-0.55	0.02	-0.47
14	-0.47	-0.04	-0.07	0.72	-0.49	0.47
15	-0.33	-0.18	-0.21	0.64	-0.43	0.60
16	0.15	-0.28	-0.44	0.16	-0.32	0.72
17	-0.09	-0.26	-0.32	0.85	-0.06	-0.08
18	-0.20	-0.03	-0.19	-0.21	0.89	-0.20
19	-0.12	-0.09	-0.30	-0.18	0.98	-0.24
20	-0.16	-0.16	-0.25	-0.13	-0.16	0.91
21	-0.37	-0.05	-0.33	0.55	-0.34	0.68
22	-0.03	-0.16	-0.32	-0.20	-0.13	0.87
24	-0.18	-0.13	-0.26	-0.14	0.96	-0.20
25	-0.19	-0.10	-0.25	-0.20	0.99	-0.20
26	-0.12	-0.15	-0.25	-0.09	-0.17	0.82
27	0.90	-0.24	-0.29	-0.21	-0.17	-0.18
28	-0.16	-0.13	-0.22	-0.12	-0.16	0.83
29	-0.29	-0.11	0.86	-0.08	-0.23	-0.23
30	-0.18	0.69	-0.02	-0.26	-0.05	-0.02
31	-0.37	0.33	0.67	-0.17	0.16	-0.59
32	0.56	-0.01	-0.01	-0.61	-0.44	0.39
33	-0.60	0.65	0.57	-0.08	-0.02	-0.37
34	-0.45	0.37	0.74	-0.17	0.07	-0.51
35	-0.45	0.50	-0.08	0.52	0.05	-0.34
36	-0.47	-0.04	0.11	0.90	-0.03	-0.37
37	-0.47	-0.01	0.42	0.74	-0.34	-0.28
38	0.41	-0.35	0.18	-0.31	-0.51	0.40
39	-0.23	-0.08	0.01	0.78	-0.08	-0.37
40	0.29	-0.32	-0.37	0.75	-0.20	-0.19
41	-0.57	0.14	0.78	0.14	-0.03	-0.43
42	0.42	0.26	-0.38	-0.45	-0.31	0.46
43	-0.28	0.03	0.59	-0.05	0.37	-0.68
44	-0.15	0.34	0.66	-0.62	0.08	-0.33
45	0.81	-0.04	-0.43	-0.35	-0.22	0.11
46	-0.16	0.04	0.80	-0.41	0.16	-0.51

47	0.68	-0.19	0.04	-0.41	-0.44	0.13
48	-0.40	-0.10	-0.32	0.03	0.69	0.22
49	-0.45	0.82	0.12	-0.07	-0.05	-0.16
50	0.87	-0.35	-0.21	-0.05	-0.36	-0.12
51	0.47	0.01	-0.56	-0.14	-0.30	0.50
52	-0.21	-0.11	-0.05	-0.23	0.87	-0.25
53	-0.63	0.12	0.81	-0.12	0.05	-0.19
54	-0.30	-0.13	-0.10	0.98	-0.21	-0.16
55	0.59	-0.05	-0.10	-0.61	-0.36	0.40
56	-0.19	-0.21	-0.32	-0.09	0.09	0.77
57	0.53	-0.35	0.42	-0.44	-0.24	-0.16
58	-0.14	-0.25	-0.17	-0.20	-0.12	0.89
59	-0.37	0.03	0.84	-0.31	0.14	-0.37
60	0.42	-0.11	-0.39	-0.32	-0.32	0.65
61	-0.33	0.66	0.07	-0.18	-0.26	0.22
62	-0.45	0.10	0.78	-0.20	0.03	-0.26
63	-0.18	-0.08	-0.24	-0.18	0.93	-0.19
64	-0.22	0.76	-0.12	-0.12	-0.15	0.05
65	-0.12	-0.05	-0.26	-0.15	-0.21	0.85
66	-0.18	-0.09	-0.25	-0.19	0.98	-0.20
67	0.95	-0.24	-0.35	-0.28	-0.10	-0.17
68	-0.16	0.58	-0.15	-0.16	0.18	-0.14
69	0.97	-0.23	-0.31	-0.26	-0.19	-0.19

For each variable, the correlation coefficient of corresponding weight map with the pattern of assignation map for each cancer class can be calculated:

CC(i) = 0: indicates no correlation between biomarker and the cancer class i;
1> CC(i) > 0: accordance between the biomarker intensity and cancer class i.
0<CC<-1: an inverse correlation between biomarker value and cancer class i.

The CC values >0.5 or <-0.5 are colored. For example, the weight map of biomarker 1282 is highly correlated with the pattern of cancer class 4 on the assignation map, and it may be an important biomarker for the samples from patients with myeloma.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ACUTE PHASE
A1AG1 A1AG2 AACT A1AT A1BG FETUA A2MG CRP HPT HPTR LBP LRBA CXCL7
PEAR1 PECA1 PLF4 CD36 GP1BA GP1BB GPIX GPV SAA1 SAA2 SAA4 SAMP
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ACUTE PHASE
A2MG A1AT HPT LBP SAA2 HRG FETUA AACT SAA4 A1AG1 PLF4 SAA1
A1BG HPTR A1AG2 GPIX CXCL7 SAMP CD36 CRP GP1BA PECA1 GP1BB GPV
Neutral
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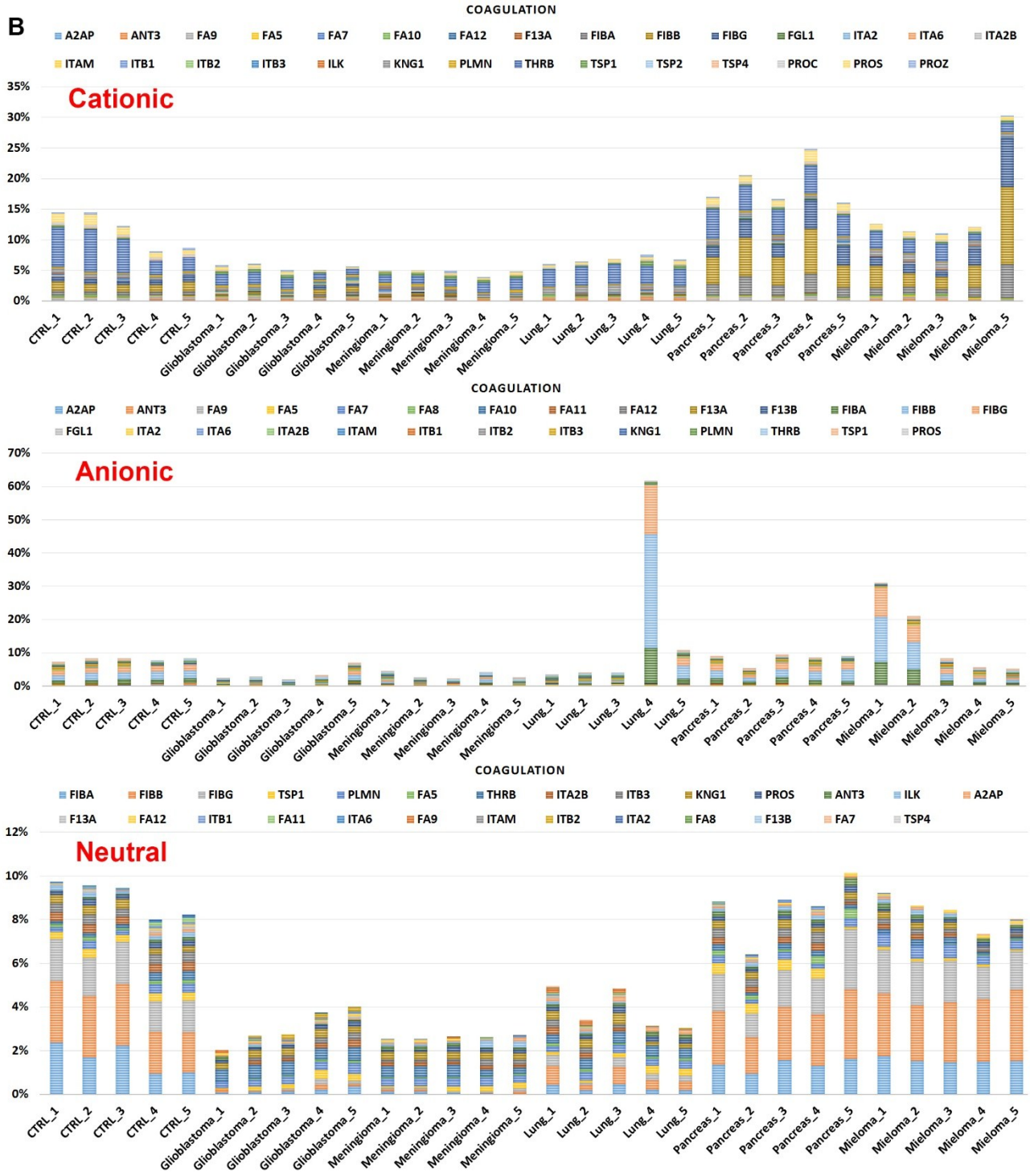
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COAGULATION
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COAGULATION
FIBA FIBB FIBG TSP1 PLMN FA5 THRB ITA2B ITB3 KNG1 PROS ANT3 ILK A2AP
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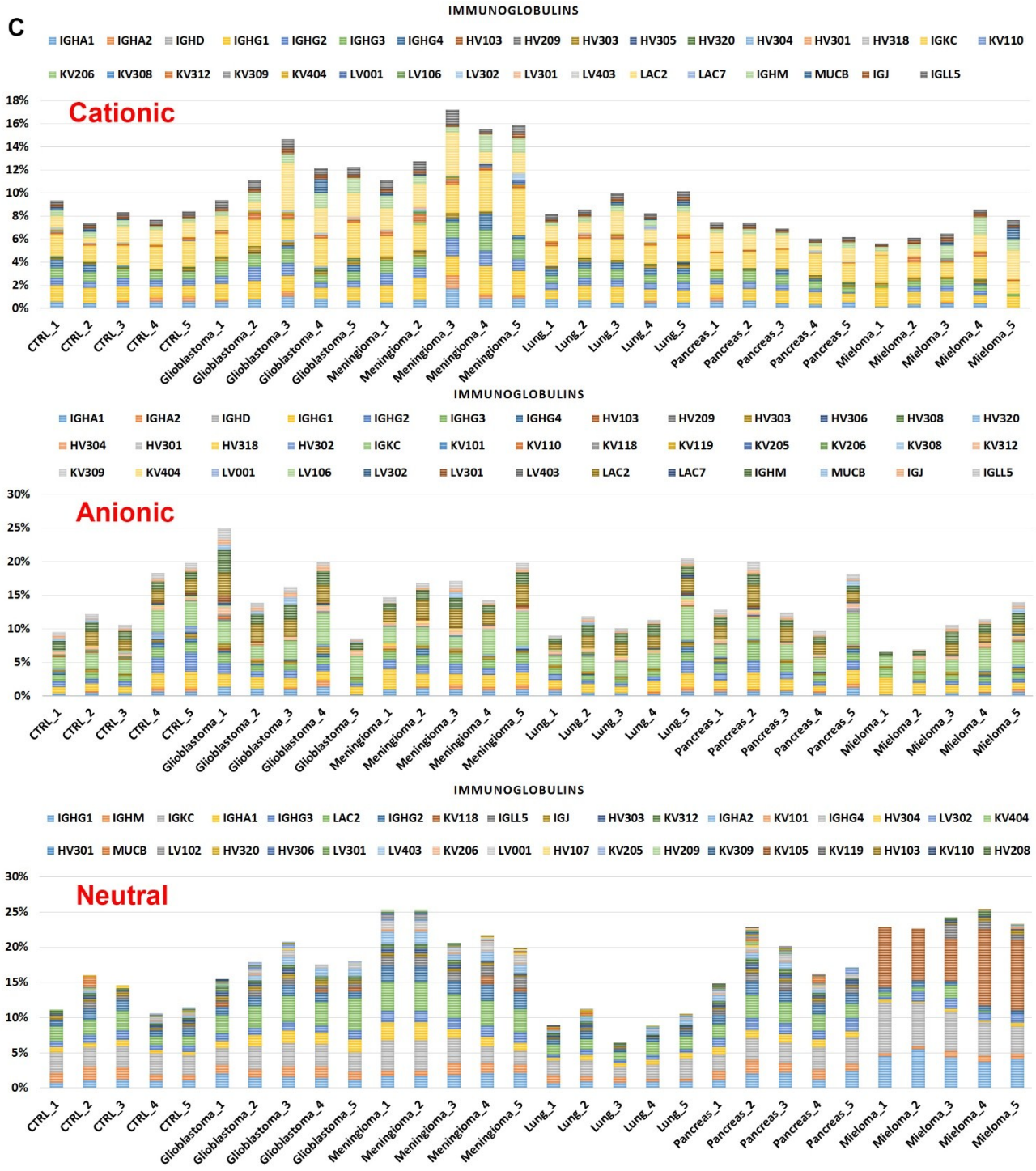
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HV304 HV301 HV318 HV302 IGKC KV101 KV110 KV118 KV119 KV205 KV206 KV308 KV312
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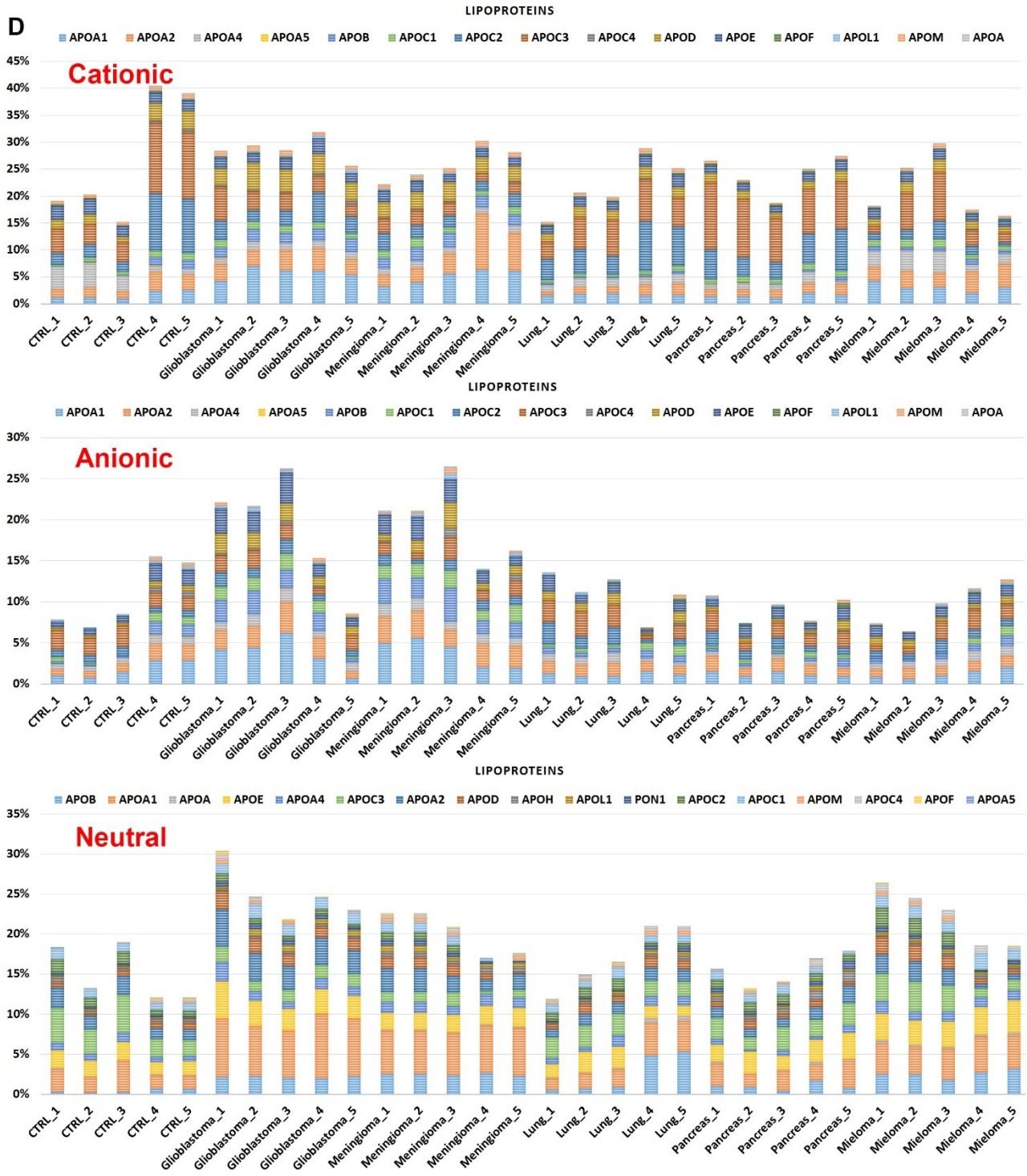
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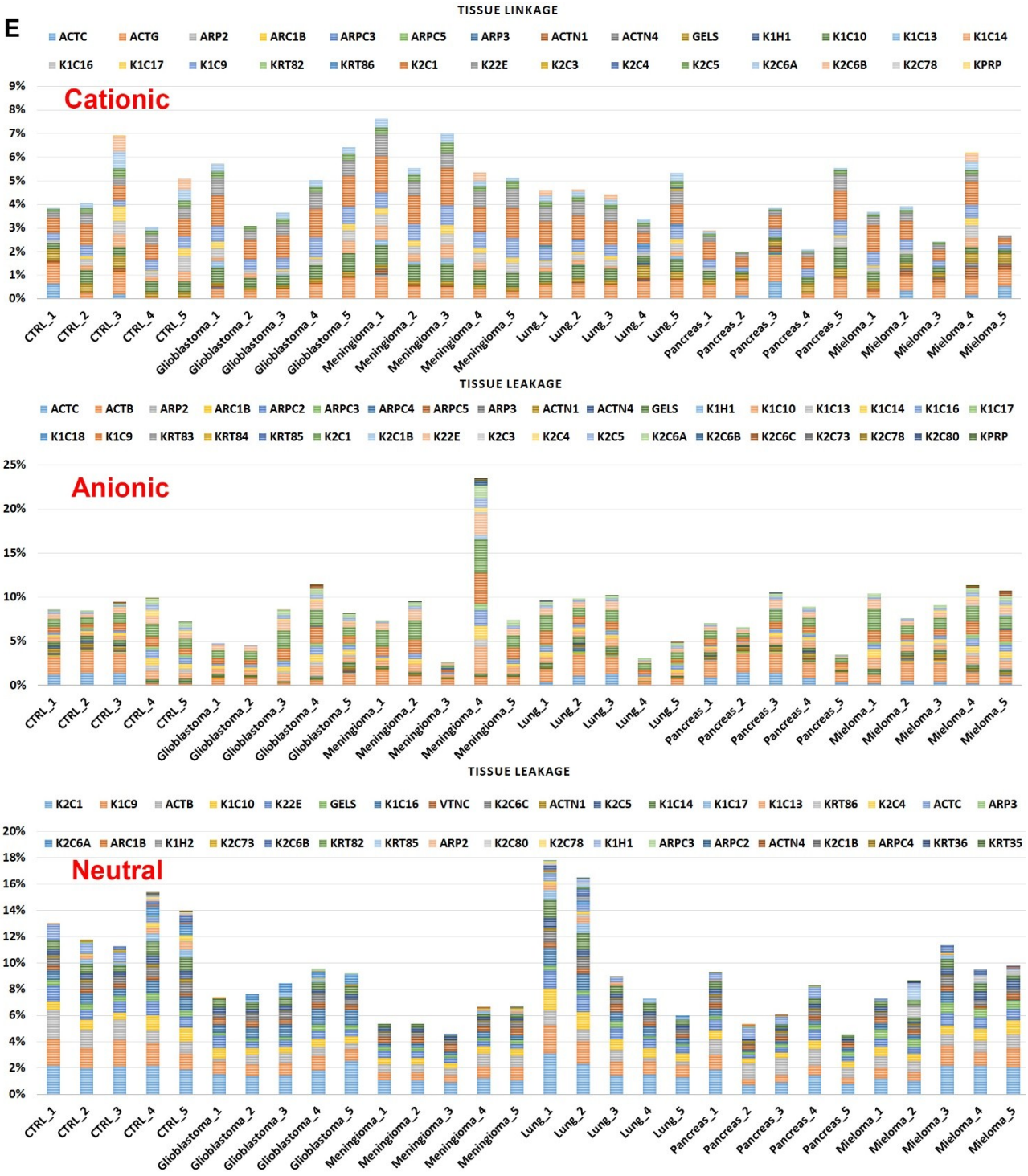
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TISSUE LINKAGE
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Cationic
9% 8% 7% 6% 5% 4% 3% 2% 1% 0%
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TISSUE LEAKAGE
ACTC ACTB ARP2 ARC1B ARPC2 ARPC3 ARPC4 ARPC5 ARP3 ACTN1 ACTN4 GELS K1H1 K1C10 K1C13 K1C14 K1C16 K1C17
K1C18 K1C9 KRT83 KRT84 KRT85 K2C1 K2C1B K22E K2C3 K2C4 K2C5 K2C6A K2C6B K2C6C K2C73 K2C78 K2C80 KPRP
Anionic
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TISSUE LEAKAGE
K2C1 K1C9 ACTB K1C10 K22E GELS K1C16 VTNC K2C6C ACTN1 K2C5 K1C14 K1C17 K1C13 KRT86 K2C4 ACTC ARP3
K2C6A ARC1B K1H2 K2C73 K2C6B KRT82 KRT85 ARP2 K2C80 K2C78 K1H1 ARPC3 ARPC2 ACTN4 K2C1B ARPC4 KRT36 KRT35
Neutral
20% 18% 16% 14% 12% 10% 8% 6% 4% 2% 0%
CTRL_1 CTRL_2 CTRL_3 CTRL_4 CTRL_5 Glioblastoma_1 Glioblastoma_2 Glioblastoma_3 Glioblastoma_4 Glioblastoma_5 Meningioma_1 Meningioma_2 Meningioma_3 Meningioma_4 Meningioma_5 Lung_1 Lung_2 Lung_3 Lung_4 Lung_5 Pancreas_1 Pancreas_2 Pancreas_3 Pancreas_4 Pancreas_5 Mieloma_1 Mieloma_2 Mieloma_3 Mieloma_4 Mieloma_5

F

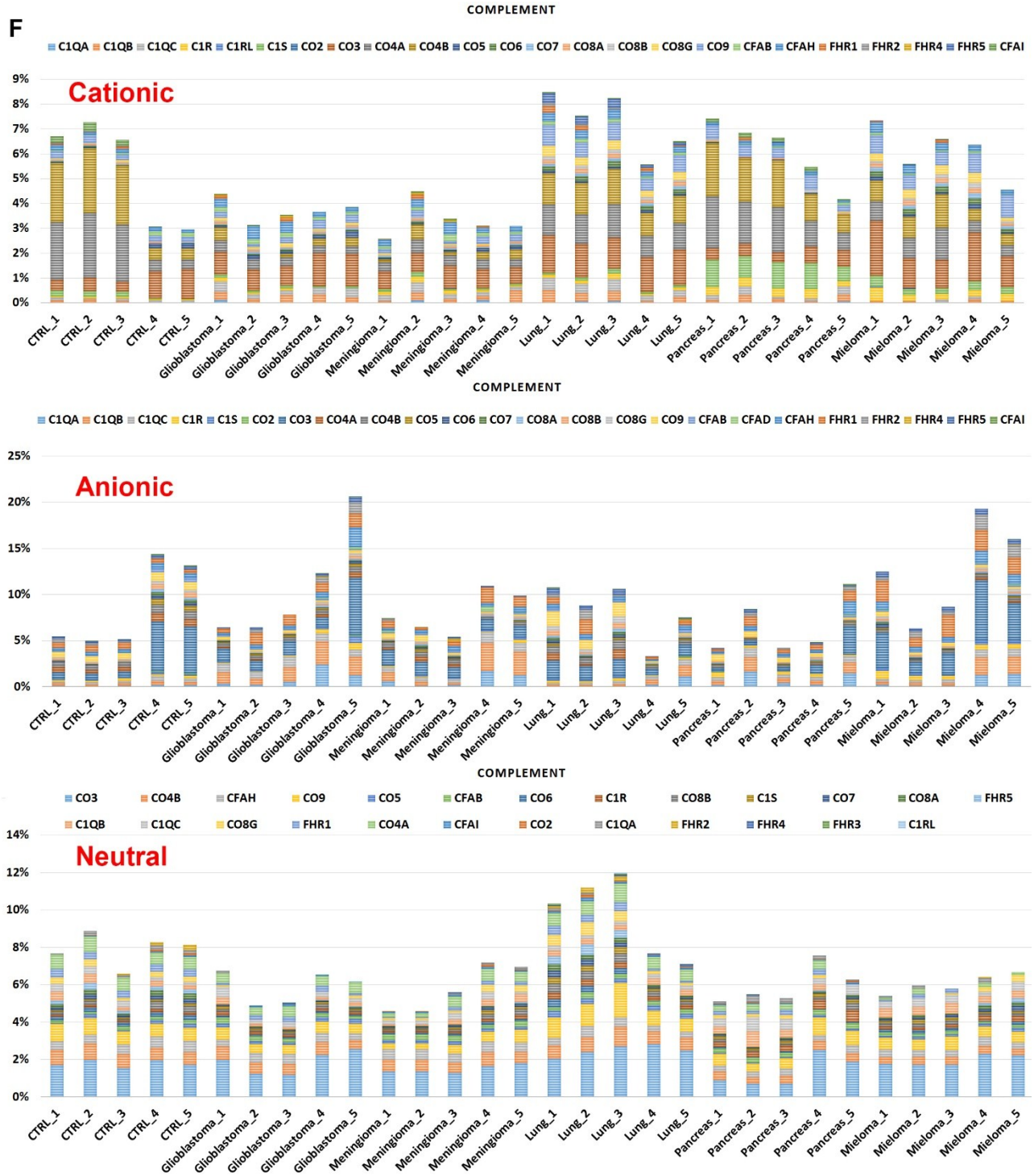
COMPLEMENT
C1QA C1QB C1QC C1R C1RL C1S CO2 CO3 CO4A CO4B CO5 CO6 CO7 CO8A CO8B CO8G CO9 CFAB CFAH FHR1 FHR2 FHR4 FHR5 CFAI
Cationic
9%
8%
7%
6%
5%
4%
3%
2%
1%
0%
CTRL_1 CTRL_2 CTRL_3 CTRL_4 CTRL_5 Glioblastoma_1 Glioblastoma_2 Glioblastoma_3 Glioblastoma_4 Glioblastoma_5 Meningioma_1 Meningioma_2 Meningioma_3 Meningioma_4 Meningioma_5 Lung_1 Lung_2 Lung_3 Lung_4 Lung_5 Pancreas_1 Pancreas_2 Pancreas_3 Pancreas_4 Pancreas_5 Mieloma_1 Mieloma_2 Mieloma_3 Mieloma_4 Mieloma_5
COMPLEMENT
C1QA C1QB C1QC C1R C1S CO2 CO3 CO4A CO4B CO5 CO6 CO7 CO8A CO8B CO8G CO9 CFAB CFAD CFAH FHR1 FHR2 FHR4 FHR5 CFAI
Anionic
25%
20%
15%
10%
5%
0%
CTRL_1 CTRL_2 CTRL_3 CTRL_4 CTRL_5 Glioblastoma_1 Glioblastoma_2 Glioblastoma_3 Glioblastoma_4 Glioblastoma_5 Meningioma_1 Meningioma_2 Meningioma_3 Meningioma_4 Meningioma_5 Lung_1 Lung_2 Lung_3 Lung_4 Lung_5 Pancreas_1 Pancreas_2 Pancreas_3 Pancreas_4 Pancreas_5 Mieloma_1 Mieloma_2 Mieloma_3 Mieloma_4 Mieloma_5
COMPLEMENT
CO3 CO4B CFAH CO9 CO5 CFAB CO6 C1R CO8B C1S CO7 CO8A FHR5
C1QB C1QC CO8G FHR1 CO4A CFAI CO2 C1QA FHR2 FHR4 FHR3 C1RL
Neutral
14%
12%
10%
8%
6%
4%
2%
0%
CTRL_1 CTRL_2 CTRL_3 CTRL_4 CTRL_5 Glioblastoma_1 Glioblastoma_2 Glioblastoma_3 Glioblastoma_4 Glioblastoma_5 Meningioma_1 Meningioma_2 Meningioma_3 Meningioma_4 Meningioma_5 Lung_1 Lung_2 Lung_3 Lung_4 Lung_5 Pancreas_1 Pancreas_2 Pancreas_3 Pancreas_4 Pancreas_5 Mieloma_1 Mieloma_2 Mieloma_3 Mieloma_4 Mieloma_5

Fig S1. Classification of identified coronas by sensor array elements according to their physiological functions, including (A) acute phase, (B) coagulation, (C) immunoglobulins, (D) lipoproteins, (E) tissue leakage, (F) complement, and (G) other plasma proteins, in human plasma of healthy subjects and patients having different types of cancers (y-axis is the percentage normalized spectral count for proteins).

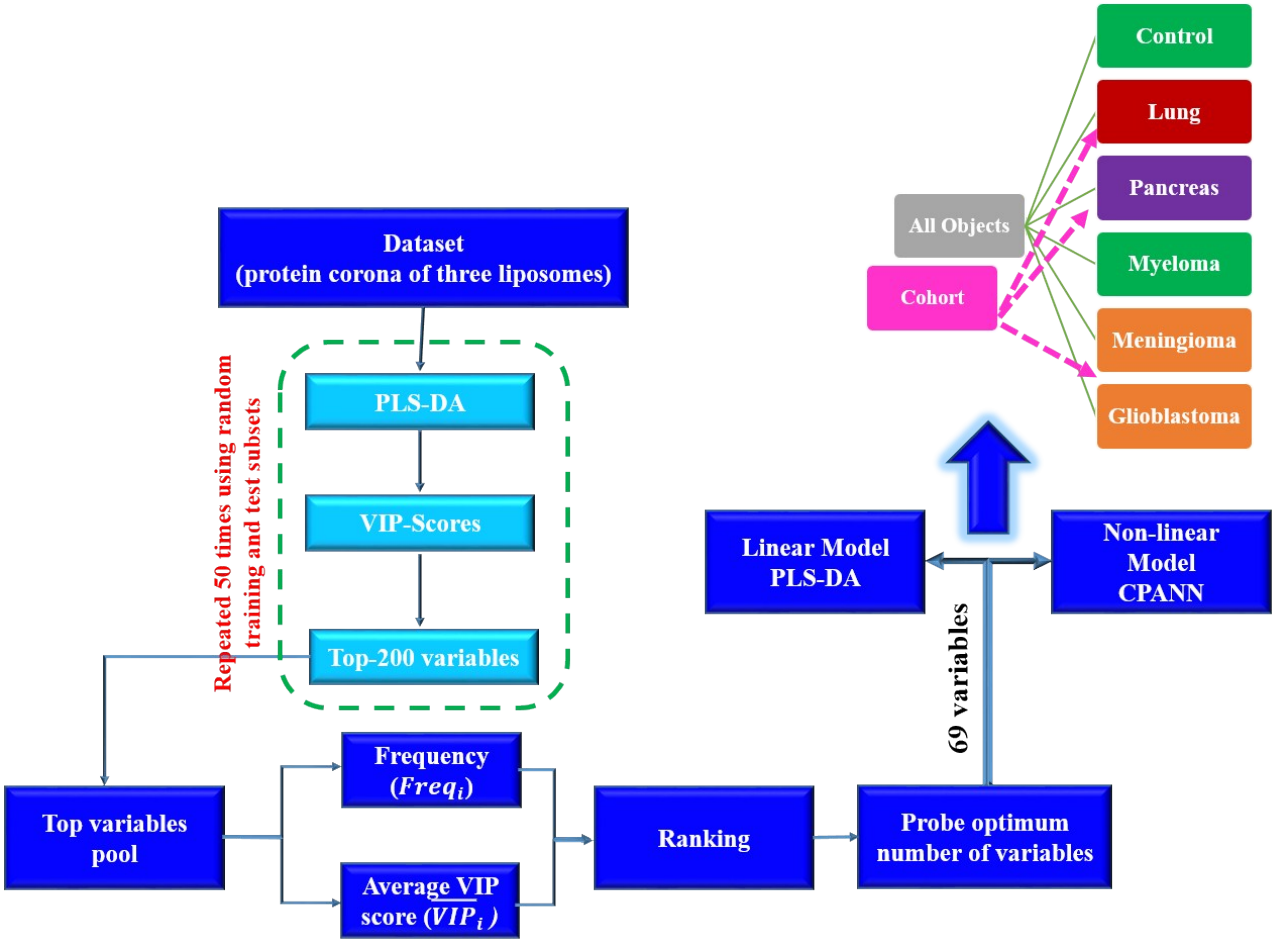

Fig. S2 Schematic representation of study outline. Informative variable selection and classification model building.

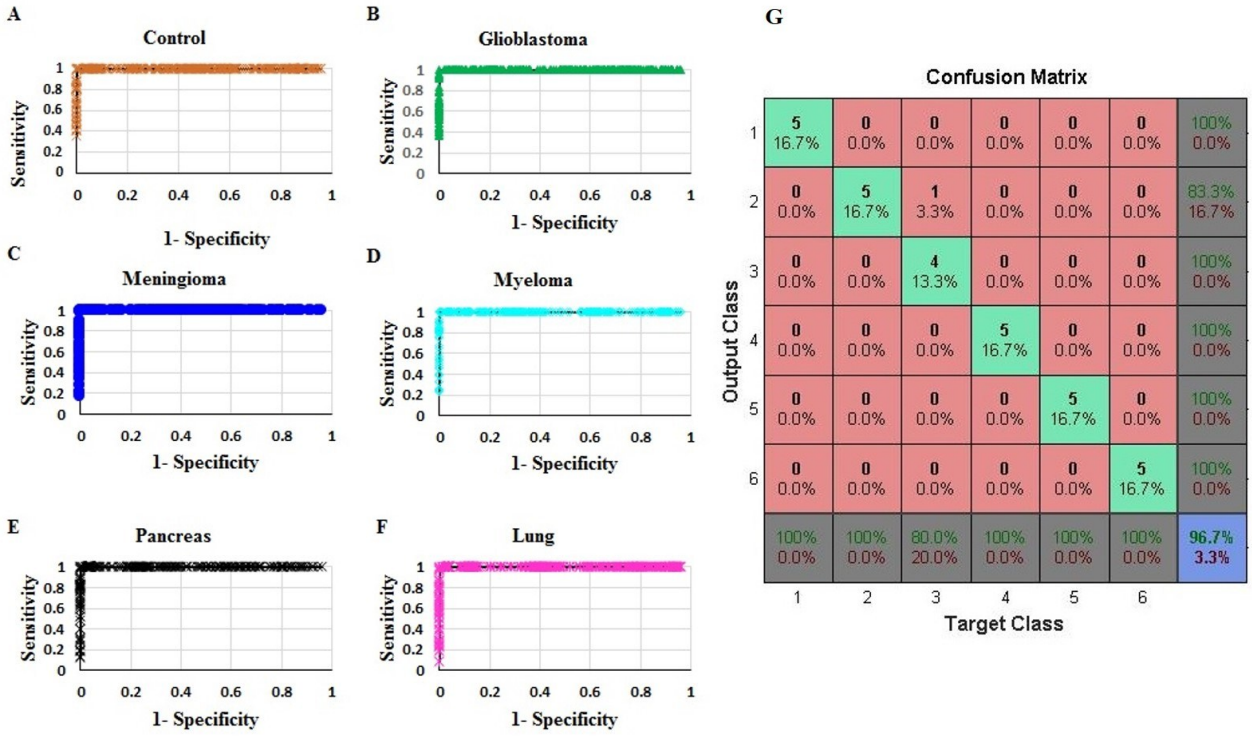

Fig. S3. ROC (receiver operating characteristic) plots derived from PLS-DA and confusion matrix drived from CPANN (counter propagation artificial neural network) based on the top 69 ranked variables for five cancer and control classes. (A-F) ROC plot of sensitivity (True Positive Rate, Y-axis) versus 1 – specificity (False Positive Rate, X-axis) based on a PLS-DA built upon the 69 markers with the highest contribution for six classes. (G) Obtained Confusion matrix by CPANN indicates good prediction for all classes.

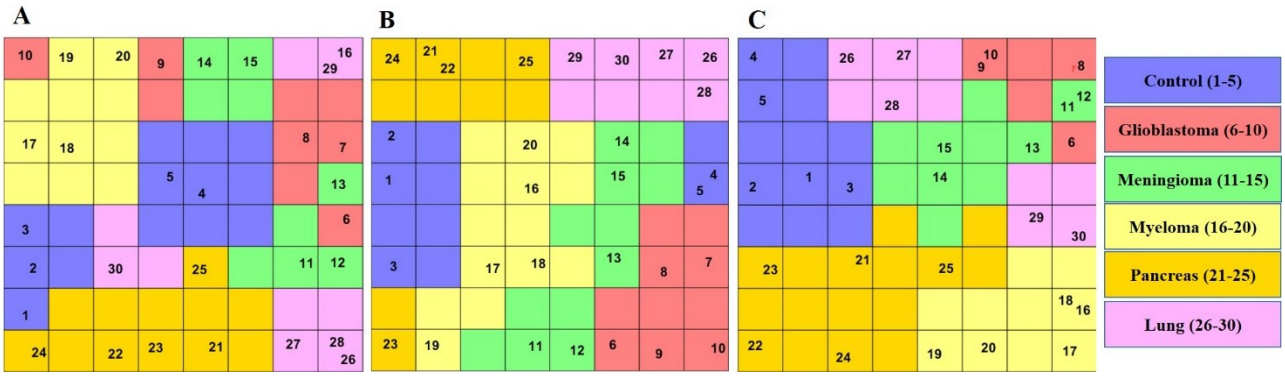

Fig. S4. Data analysis using CPANN on the data matrix corresponding to protein corona of each liposome separately. Assignation map obtained by all variables of protein corona of each liposome (anionic (A), cationic (B) and neutral (C)). As illustrated in these assignation maps, no one class of the liposomes could discriminate all 6 groups of samples as well as the composite response of the full array. The classification error for the model of anionic, cationic and neutral is 54%, 24% and 10%, respectively. Therefore, we used the protein corona profiles of all three liposomes to get the benefit of all sensor information probing cancer-specific signatures.