

Supporting Information

Upconversion fluorescent nanoparticles based-sensor array for discrimination of the same variety red grape wines

Kewei Wang,^a Yanli Li,^a Haijie Li,^a Mingyuan. Yin,^a Huilin Liu,^b Qiliang Deng,^{*a} and Shuo Wang^{*a,c}

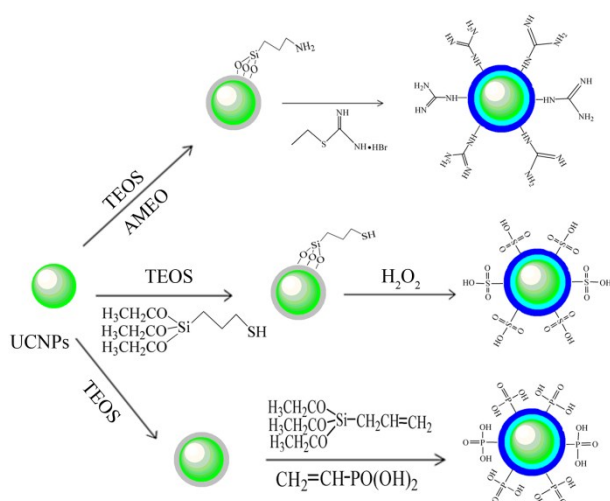
a. Key Laboratory of Food Nutrition and Safety, Ministry of Education, Tianjin Key Laboratory of Food Nutrition and Safety, College of Chemical Engineering and Materials Science, Tianjin University of Science and Technology, Tianjin, 300457, P. R. China. E-mail: yhdql@tust.edu.cn; s.wang@tust.edu.cn

b. Beijing Advanced Innovation Center for Food Nutrition and Human Health, Beijing Technology and Business University, 11 Fucheng Road, Beijing, 100048, China.

c. Tianjin Key Laboratory of Food Science and Health, School of Medicine, Nankai University, 94 Weijin Road, Tianjin, 300071, China.

Corresponding author: Qiliang Deng or Shuo Wang

E-mail: yhdql@tust.edu.cn or S. wang@tust.edu.cn



Scheme.S1 Schematic illustrations for the preparation of UCNPs@GDN, UCNPs@SO₃H and UCNPs@PO(OH)₂

Table S1. Details of the red grape wine ingredients used in this experiment

Type	Ingredient	Content
alcohols	Ethanol	12.0%
saccharides	Fructose	7 .0g L ⁻¹
	Glucose	3.0 g L ⁻¹
	Glycerol	8.0 g L ⁻¹
acids	malic acid	0.5 g L ⁻¹
	tartaric acid	4.0g L ⁻¹
	lactic acid	1.0 g L ⁻¹
	citric acid	0.5 g L ⁻¹
	gallic acid	0.5 g L ⁻¹
	acetic acid	1.0 g L ⁻¹
	tannic acid	0.085g L ⁻¹
vitamins	ascorbic acid	0.14 g L ⁻¹

Table S2. Details of the red grape wines used in this experiment.

Wine	Varietal	Origin	Suger	Alcohol Content (%)	Vintage	Brand
1	Cabernet Sauvignon	Xinjiang China	dry	12.0	2015	Changyu
2	Cabernet Sauvignon	Raman Spain	dry	12.0	2016	Detunda
3	Cabernet Sauvignon	Barossa Valley Australia	dry	14.5	2014	Penfolds BIN2
4	Cabernet Sauvignon	Medoc France	dry	13.0	2013	Comte Rossi
5	Cabernet Sauvignon	California USA	dry	13.5	2015	Beringer
6	Cabernet Sauvignon	Mendoza Argentina	dry	14.5	2016	Vinalba
7	Cabernet Sauvignon	Central Valley Chile	dry	13.5	2016	Concha y Toro
8	Cabernet Sauvignon	Parl South Africa	dry	14.5	2016	Nederburg
9	Cabernet Sauvignon	Puglia Italy	dry	12.5	2015	Saint Benitez

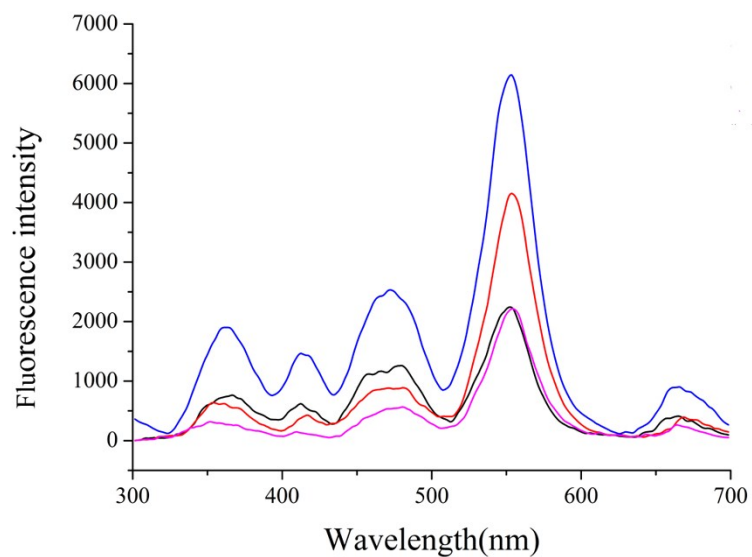


Fig.S1 Fluorescence spectra of UCNPs (pink), UCNPs@SO₃H (blue), UCNPs@PO(OH)₂ (red) and UCNPs@GDN (black)

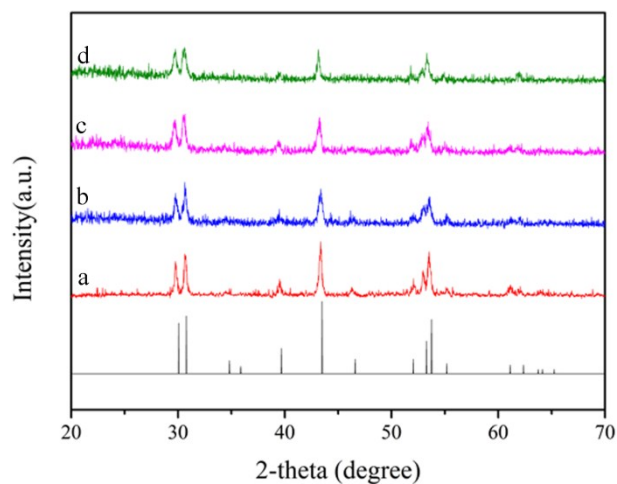


Fig.S2 XRD patterns of UCNPs (a), UCNPs@SO₃H (b), UCNPs@PO(OH)₂ (c) and UCNPs@GDN (d)

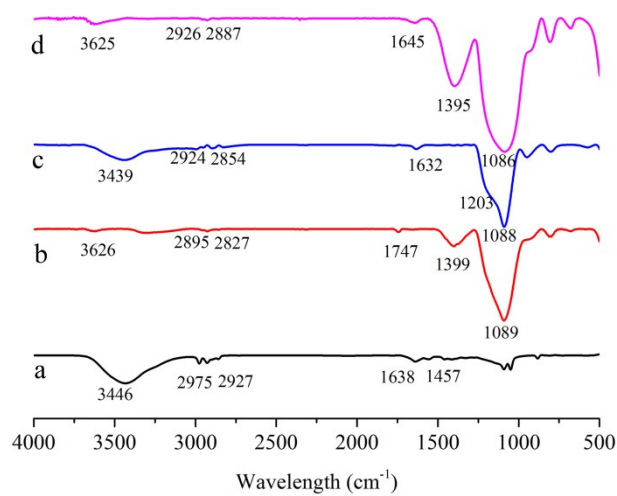


Fig.S3 FT-IR spectrum of UCNPs (a), UCNPs@SO₃H (b), UCNPs@PO(OH)₂ (c) and UCNPs@GDN (d)

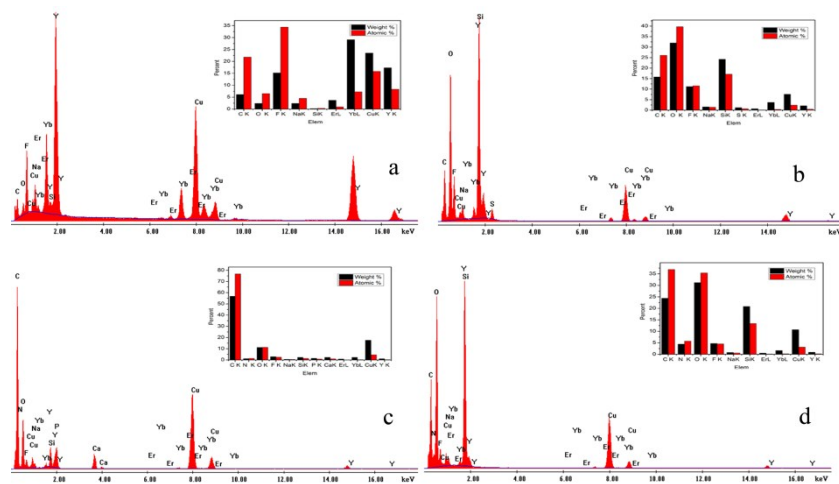


Fig.S4 EDS of UCNPs (a), UCNPs@SO₃H (b), UCNPs@PO(OH)₂ (c) and UCNPs@GDN (d)

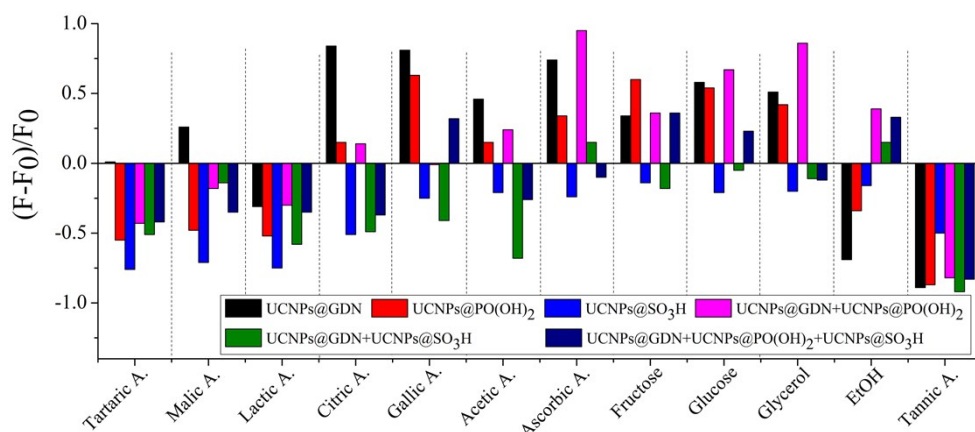


Fig.S5 Fluorescence response pattern $(F-F_0)/F_0$ obtained from the UCNPs materials treated with different wine ingredients.

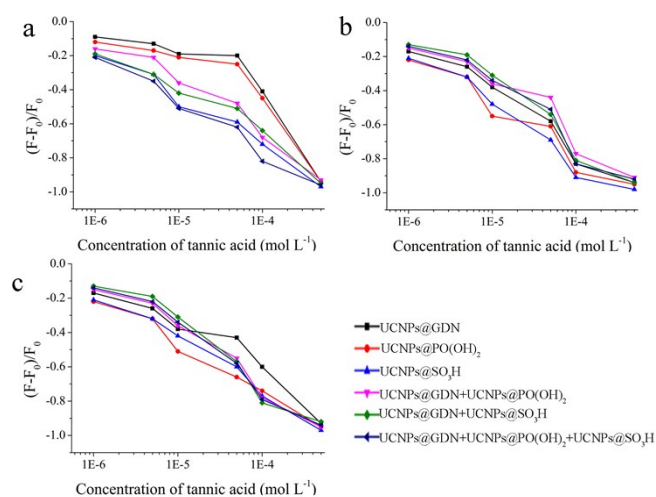


Fig.S6 Fluorescence response pattern $(F-F_0)/F_0$ obtained from the UCNPs materials treated with different concentrations tannic acid in PBS buffer (a), TRIS-HCl buffer (b) and HEPES buffer (c)

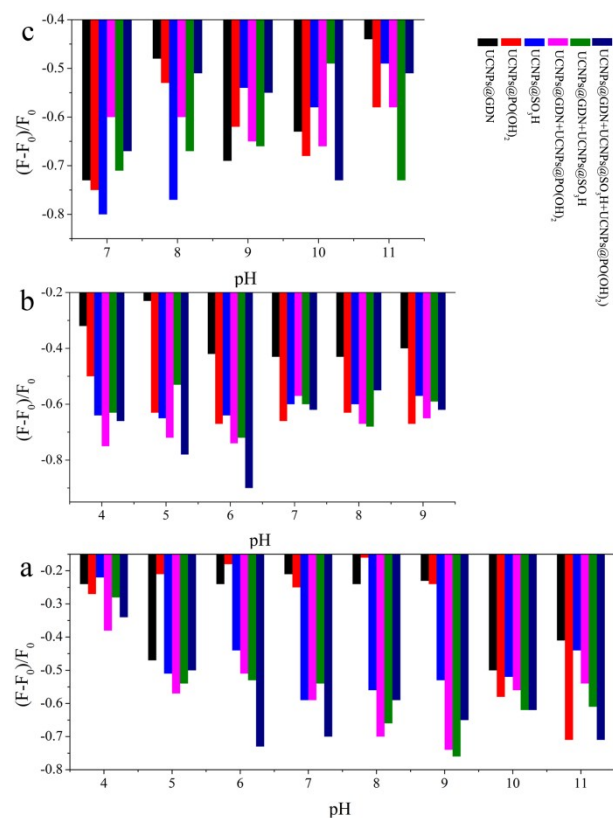


Fig.S7 Fluorescence response pattern $(F-F_0)/F_0$ obtained from the UCNPs materials treated with tannic acid in HEPES buffer (a), TRIS-HCl buffer (b) and PBS buffer (c)

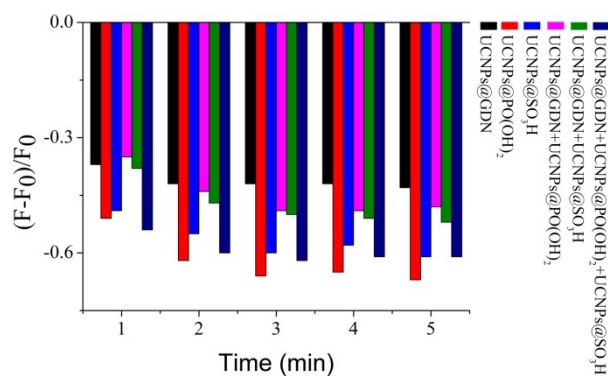


Fig.S8 Fluorescence response pattern $(F-F_0)/F_0$ obtained from the UCNPs materials treated with different times

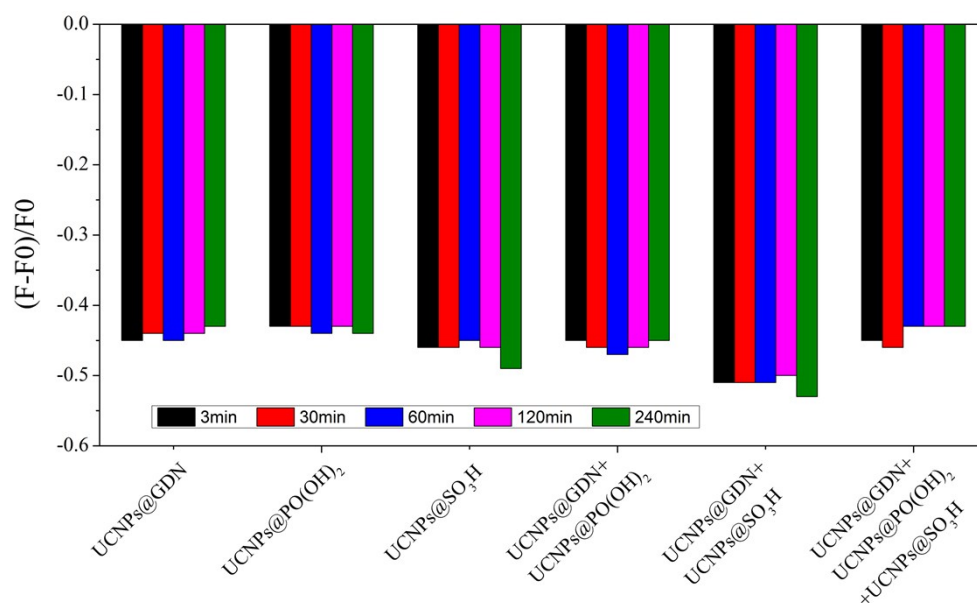


Fig.S9 The effect of different time (3min, 30min, 60min, 120min, 240min) on the quenching of tannic acid to sensor array resulted in differences in fluorescence $(F_0-F)/F_0$

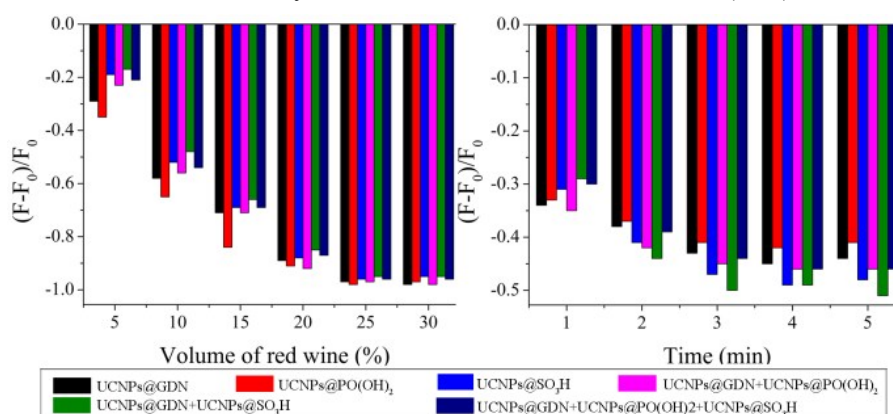


Fig.S10 Fluorescence response pattern $(F-F_0)/F_0$ obtained from the fluorescent materials treated with different concentrations red wine (left); different times (right) in PBS buffer, Tris-HCl buffer and HEPES buffer

Principal component analysis (PCA)

Table S3. Training matrix of fluorescence response pattern from an sensor array of UCNPs@GDN, UCNPs@SO₃H and UCNPs@PO(OH)₂ (each at pH=7.0 HEPES buffer) and the mixture 1 {UCNPs@GDN+UCNPs@SO₃H}, mixture 2 {UCNPs@GDN+UCNPs@PO(OH)₂} (at pH=9.0 PBS buffer), mixture 3 {UCNPs@GDN+UCNPs@SO₃H+UCNPs@PO(OH)₂} (at pH=6.0 TRIS-HCl buffer) against nine red grape wines. PCA was carried out as described above resulting in the six factors of the canonical scores and group generation.

Analyte		Fluorescence response pattern					Results PCA						
Wines	UCNPs@ GDN (pH7.0)	UCNPs@ PO(OH) ₂ (pH7.0)	UCNPs@ SO ₃ H (pH7.0)	mixture 1 (pH9.0)	mixture 2 (pH9.0)	mixture 3 (pH6.0)	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Group
Wine 1	-0.431	-0.393	-0.497	-0.445	-0.502	-0.441	1.5542	-1.376	-0.364	-1.1418	0.1661	-0.042	6
Wine 1	-0.439	-0.396	-0.498	-0.447	-0.507	-0.443	1.4771	-1.4064	-0.4368	-1.1929	0.1514	-0.0167	6
Wine 1	-0.441	-0.393	-0.504	-0.456	-0.509	-0.448	1.4201	-1.3922	-0.53	-1.1705	0.2997	-0.0202	6
Wine 1	-0.443	-0.394	-0.507	-0.459	-0.513	-0.454	1.3266	-1.42	-0.6153	-1.1414	0.3379	-0.0484	6
Wine 1	-0.448	-0.395	-0.511	-0.455	-0.515	-0.449	1.3629	-1.4747	-0.5601	-1.2211	0.293	0.0005	6
Wine 1	-0.449	-0.409	-0.514	-0.456	-0.516	-0.453	1.1433	-1.4919	-0.6063	-1.1645	0.1347	0.1056	6
Wine 2	-0.311	-0.399	-0.454	-0.405	-0.364	-0.432	2.3606	-0.7832	0.3365	0.2964	-0.528	0.011	7
Wine 2	-0.312	-0.405	-0.453	-0.403	-0.361	-0.431	2.3218	-0.7839	0.3439	0.3101	-0.6354	0.0764	7
Wine 2	-0.313	-0.397	-0.451	-0.406	-0.363	-0.436	2.3658	-0.7819	0.26	0.3242	-0.4991	-0.034	7
Wine 2	-0.315	-0.405	-0.453	-0.407	-0.369	-0.443	2.1754	-0.8246	0.1744	0.3633	-0.5692	-0.0318	7
Wine 2	-0.317	-0.406	-0.455	-0.413	-0.371	-0.439	2.1513	-0.7759	0.1997	0.2951	-0.5294	0.0465	7
Wine 2	-0.318	-0.407	-0.453	-0.409	-0.375	-0.445	2.0965	-0.8359	0.1353	0.3302	-0.568	-0.0393	7
Wine 3	-0.35	-0.399	-0.501	-0.483	-0.384	-0.522	1.2341	-0.8835	-1.2019	0.8764	0.466	-0.1412	8
Wine 3	-0.353	-0.407	-0.504	-0.483	-0.384	-0.525	1.113	-0.9148	-1.2507	0.9092	0.3731	-0.0755	8

Wine 3	-0.356	-0.409	-0.508	-0.485	-0.384	-0.527	1.0644	-0.9344	-1.2946	0.9209	0.3784	-0.0409	8
Wine 3	-0.359	-0.403	-0.513	-0.49	-0.393	-0.53	1.0422	-0.9617	-1.3445	0.8706	0.5346	-0.1096	8
Wine 3	-0.357	-0.406	-0.515	-0.488	-0.392	-0.532	0.9991	-0.9864	-1.3463	0.9197	0.4882	-0.1053	8
Wine 3	-0.358	-0.41	-0.515	-0.489	-0.391	-0.532	0.9509	-0.9758	-1.3566	0.9258	0.4416	-0.0563	8
Wine 4	-0.362	-0.494	-0.193	-0.645	-0.484	-0.543	-1.1304	1.849	-2.4391	-0.1565	-0.2324	-0.0681	9
Wine 4	-0.369	-0.5	-0.192	-0.644	-0.48	-0.54	-1.1423	1.8472	-2.4527	-0.1918	-0.3418	0.0431	9
Wine 4	-0.367	-0.497	-0.194	-0.644	-0.487	-0.541	-1.1583	1.823	-2.4348	-0.2165	-0.2809	-0.0252	9
Wine 4	-0.37	-0.497	-0.198	-0.647	-0.489	-0.543	-1.1979	1.8065	-2.4789	-0.2221	-0.2374	-0.0126	9
Wine 4	-0.368	-0.5	-0.203	-0.649	-0.49	-0.549	-1.3047	1.7808	-2.5349	-0.1439	-0.2286	-0.0198	9
Wine 4	-0.371	-0.498	-0.201	-0.647	-0.49	-0.549	-1.261	1.7619	-2.5534	-0.1646	-0.229	-0.0457	9
Wine 5	-0.139	-0.397	-0.201	-0.576	-0.383	-0.377	1.7786	2.7611	1.1765	-0.0777	0.2218	-0.1007	1
Wine 5	-0.137	-0.396	-0.202	-0.574	-0.38	-0.371	1.8566	2.7752	1.2681	-0.1079	0.2073	-0.0585	1
Wine 5	-0.14	-0.396	-0.209	-0.578	-0.387	-0.372	1.7864	2.746	1.2495	-0.1522	0.2756	-0.046	1
Wine 5	-0.139	-0.403	-0.211	-0.581	-0.387	-0.374	1.6636	2.7627	1.2281	-0.1147	0.2204	0.0193	1
Wine 5	-0.137	-0.405	-0.21	-0.579	-0.383	-0.379	1.6328	2.7462	1.1758	-0.0216	0.1779	-0.0061	1
Wine 5	-0.136	-0.405	-0.212	-0.578	-0.385	-0.377	1.6361	2.7348	1.2194	-0.0453	0.1763	-0.0007	1
Wine 6	-0.163	-0.49	-0.503	-0.55	-0.561	-0.451	-1.1534	0.3228	1.1606	0.1094	0.1724	0.201	2
Wine 6	-0.162	-0.483	-0.509	-0.547	-0.564	-0.458	-1.1258	0.2289	1.1032	0.1713	0.2771	0.0661	2
Wine 6	-0.162	-0.486	-0.502	-0.548	-0.565	-0.453	-1.1308	0.2943	1.1533	0.1047	0.2152	0.1121	2
Wine 6	-0.163	-0.488	-0.51	-0.55	-0.567	-0.457	-1.214	0.2487	1.1095	0.1423	0.2425	0.1274	2
Wine 6	-0.164	-0.488	-0.511	-0.549	-0.568	-0.46	-1.2355	0.2172	1.0721	0.1647	0.2432	0.0985	2
Wine 6	-0.165	-0.487	-0.508	-0.551	-0.569	-0.459	-1.2259	0.2479	1.067	0.1332	0.2626	0.0971	2
Wine 7	-0.273	-0.481	-0.522	-0.512	-0.511	-0.478	-0.5577	-0.4906	0.1263	0.1652	-0.105	0.3374	3
Wine 7	-0.274	-0.482	-0.522	-0.514	-0.513	-0.478	-0.5901	-0.4797	0.1173	0.1461	-0.0986	0.3507	3
Wine 7	-0.273	-0.482	-0.526	-0.515	-0.511	-0.47	-0.5329	-0.4512	0.2257	0.0859	-0.0931	0.4431	3
Wine 7	-0.273	-0.484	-0.524	-0.514	-0.518	-0.475	-0.6276	-0.4828	0.1774	0.0949	-0.1149	0.3734	3

Wine 7	-0.271	-0.485	-0.524	-0.514	-0.514	-0.478	-0.6424	-0.4795	0.1451	0.1645	-0.1272	0.3699	3
Wine 7	-0.276	-0.484	-0.524	-0.513	-0.517	-0.477	-0.6253	-0.5077	0.1329	0.1069	-0.1243	0.3654	3
Wine 8	-0.213	-0.522	-0.575	-0.542	-0.66	-0.508	-2.4931	-0.6906	0.4846	-0.0208	0.1342	-0.1115	4
Wine 8	-0.219	-0.522	-0.571	-0.54	-0.661	-0.508	-2.4726	-0.7097	0.4448	-0.0645	0.0994	-0.1216	4
Wine 8	-0.213	-0.523	-0.57	-0.544	-0.662	-0.511	-2.5431	-0.6632	0.4347	-0.0111	0.1301	-0.1433	4
Wine 8	-0.217	-0.525	-0.573	-0.545	-0.662	-0.511	-2.5692	-0.6816	0.4109	-0.0263	0.1188	-0.0975	4
Wine 8	-0.215	-0.527	-0.578	-0.546	-0.669	-0.508	-2.6289	-0.6919	0.4843	-0.0811	0.1252	-0.0721	4
Wine 8	-0.214	-0.529	-0.575	-0.545	-0.668	-0.511	-2.6635	-0.6907	0.4496	-0.0381	0.0848	-0.0932	4
Wine 9	-0.193	-0.486	-0.509	-0.482	-0.58	-0.458	-0.8734	-0.4717	1.1468	-0.0169	-0.3307	-0.2366	5
Wine 9	-0.191	-0.487	-0.508	-0.483	-0.582	-0.458	-0.9056	-0.4544	1.1598	-0.0199	-0.3336	-0.2415	5
Wine 9	-0.194	-0.485	-0.508	-0.482	-0.581	-0.461	-0.8848	-0.4851	1.1016	-0.0007	-0.3148	-0.2757	5
Wine 9	-0.197	-0.485	-0.51	-0.48	-0.579	-0.461	-0.8597	-0.5187	1.0877	0.0012	-0.3313	-0.2608	5
Wine 9	-0.195	-0.486	-0.515	-0.482	-0.58	-0.458	-0.8766	-0.509	1.1448	-0.0182	-0.3124	-0.2102	5
Wine 9	-0.195	-0.485	-0.513	-0.484	-0.579	-0.461	-0.887	-0.4925	1.0935	0.0122	-0.2847	-0.2355	5

Table S4. Detection and identification of unknown red wine samples using PCA. All unknown samples could be assigned to the corresponding group defined by the training matrix according to their shortest Mahalanobis distance. According to the verification, only 1 of 45 unknown wines was misclassified, representing an accuracy of 98%.

Sample	Fluorescence response pattern						Results PCA						Analyte		
#	UCNPs @GDN (pH7.0)	UCNPs@ SO ₃ H (pH7.0)	UCNPs@ PO(OH) ₂ (pH7.0)	mixture 1 (pH9.0)	mixture 2 (pH9.0)	mixture 3 (pH6.0)	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Group	Identification	Verification
1	-0.369	-0.498	-0.192	-0.646	-0.486	-0.544	-1.107	1.873	-2.538	-0.057	0.351	-0.021	9	wine 4	wine 4
2	-0.141	-0.397	-0.206	-0.581	-0.387	-0.373	1.686	2.756	1.286	-0.130	-0.218	0.073	1	wine 5	wine 5
3	-0.352	-0.393	-0.512	-0.486	-0.392	-0.531	1.182	-1.019	-1.309	0.815	-0.645	0.204	8	wine 3	wine 3
4	-0.268	-0.487	-0.528	-0.512	-0.521	-0.469	-0.674	-0.505	0.275	0.043	0.097	-0.449	3	wine 7	wine 7
5	-0.199	-0.483	-0.509	-0.478	-0.578	-0.462	-0.886	-0.572	1.030	0.090	0.340	0.247	5	wine 9	wine 9
6	-0.358	-0.401	-0.514	-0.487	-0.392	-0.53	1.095	-1.028	-1.334	0.793	-0.562	0.085	8	wine 3	wine 3
7	-0.364	-0.492	-0.197	-0.646	-0.488	-0.541	-1.041	1.866	-2.455	-0.089	0.265	0.029	9	wine 4	wine 4
8	-0.329	-0.402	-0.505	-0.473	-0.387	-0.528	1.142	-0.995	-1.081	1.006	-0.435	0.200	6	wine 1	wine 3
9	-0.358	-0.4	-0.513	-0.489	-0.391	-0.532	1.092	-1.013	-1.371	0.811	-0.592	0.100	8	wine 3	wine 3
10	-0.198	-0.482	-0.506	-0.477	-0.578	-0.462	-0.867	-0.563	1.035	0.094	0.349	0.273	5	wine 9	wine 9
11	-0.215	-0.523	-0.564	-0.544	-0.658	-0.507	-2.509	-0.627	0.352	-0.044	-0.113	0.132	4	wine 8	wine 8
12	-0.446	-0.391	-0.499	-0.445	-0.507	-0.448	1.467	-1.488	-0.550	-1.216	0.140	0.159	6	wine 9	wine 9
13	-0.144	-0.397	-0.21	-0.58	-0.389	-0.372	1.687	2.718	1.293	-0.165	-0.217	0.060	1	wine 5	wine 5
14	-0.271	-0.487	-0.527	-0.513	-0.52	-0.474	-0.698	-0.520	0.182	0.083	0.085	-0.427	3	wine 1	wine 1
15	-0.195	-0.482	-0.507	-0.483	-0.581	-0.462	-0.921	-0.512	1.040	0.075	0.286	0.271	5	wine 9	wine 9
16	-0.216	-0.529	-0.571	-0.542	-0.668	-0.507	-2.641	-0.698	0.385	-0.082	-0.055	0.108	4	wine 8	wine 8
17	-0.166	-0.486	-0.503	-0.549	-0.565	-0.452	-1.176	0.263	1.093	0.039	-0.281	-0.107	2	wine 6	wine 6
18	-0.133	-0.4	-0.204	-0.574	-0.387	-0.377	1.641	2.716	1.313	-0.012	-0.137	0.129	1	wine 5	wine 5

19	-0.313	-0.4	-0.451	-0.407	-0.367	-0.441	2.208	-0.850	0.265	0.430	0.538	-0.073	7	wine 2	wine 2
20	-0.357	-0.495	-0.2	-0.642	-0.484	-0.545	-1.080	1.831	-2.444	0.041	0.299	0.034	9	wine 4	wine 4
21	-0.189	-0.482	-0.506	-0.475	-0.577	-0.462	-0.871	-0.551	1.102	0.157	0.351	0.302	5	wine 9	wine 9
22	-0.355	-0.4	-0.505	-0.482	-0.387	-0.527	1.186	-0.995	-1.281	0.814	-0.490	0.101	8	wine 3	wine 3
23	-0.361	-0.496	-0.195	-0.645	-0.488	-0.542	-1.097	1.877	-2.445	-0.047	0.317	0.014	9	wine 4	wine 4
24	-0.317	-0.401	-0.453	-0.405	-0.367	-0.437	2.237	-0.873	0.301	0.374	0.575	-0.124	7	wine 2	wine 2
25	-0.143	-0.402	-0.21	-0.579	-0.387	-0.374	1.626	2.712	1.276	-0.107	-0.160	0.019	1	wine 5	wine 5
26	-0.268	-0.48	-0.527	-0.507	-0.501	-0.472	-0.461	-0.525	0.214	0.191	0.060	-0.440	3	wine 7	wine 7
27	-0.441	-0.392	-0.505	-0.473	-0.515	-0.445	1.276	-1.257	-0.555	-1.347	-0.127	0.052	6	wine 1	wine 1
28	-0.213	-0.521	-0.564	-0.538	-0.657	-0.506	-2.448	-0.669	0.398	-0.025	-0.082	0.170	4	wine 8	wine 8
29	-0.182	-0.483	-0.505	-0.475	-0.578	-0.452	-0.837	-0.483	1.280	0.084	0.378	0.244	5	wine 9	wine 9
30	-0.375	-0.499	-0.193	-0.647	-0.491	-0.545	-1.149	1.845	-2.585	-0.110	0.358	-0.021	9	wine 4	wine 4
31	-0.307	-0.394	-0.45	-0.408	-0.365	-0.436	2.311	-0.797	0.359	0.400	0.469	-0.044	7	wine 2	wine 2
32	-0.275	-0.487	-0.526	-0.513	-0.517	-0.473	-0.663	-0.517	0.160	0.067	0.098	-0.459	3	wine 7	wine 7
33	-0.15	-0.393	-0.209	-0.58	-0.387	-0.374	1.749	2.695	1.219	-0.177	-0.251	0.091	1	wine 5	wine 5
34	-0.215	-0.528	-0.571	-0.544	-0.668	-0.51	-2.660	-0.690	0.346	-0.053	-0.093	0.136	4	wine 8	wine 8
35	-0.443	-0.391	-0.499	-0.458	-0.511	-0.449	1.368	-1.375	-0.584	-1.252	0.015	0.144	6	wine 1	wine 1
36	-0.163	-0.488	-0.516	-0.553	-0.567	-0.456	-1.280	0.221	1.075	0.089	-0.363	-0.133	2	wine 6	wine 6
37	-0.311	-0.4	-0.452	-0.407	-0.373	-0.437	2.192	-0.843	0.343	0.362	0.541	-0.068	7	wine 2	wine 2
38	-0.211	-0.52	-0.553	-0.539	-0.657	-0.507	-2.438	-0.602	0.375	-0.015	-0.064	0.222	4	wine 8	wine 8
39	-0.163	-0.489	-0.515	-0.551	-0.568	-0.451	-1.255	0.229	1.147	0.037	-0.317	-0.165	2	wine 6	wine 6
40	-0.433	-0.394	-0.513	-0.457	-0.507	-0.449	1.322	-1.415	-0.492	-1.151	-0.018	0.082	6	wine 1	wine 1
41	-0.314	-0.401	-0.453	-0.406	-0.366	-0.438	2.226	-0.857	0.303	0.405	0.558	-0.117	7	wine 2	wine 2
42	-0.166	-0.487	-0.502	-0.548	-0.563	-0.452	-1.170	0.264	1.092	0.057	-0.257	-0.121	2	wine 6	wine 6
43	-0.274	-0.48	-0.529	-0.511	-0.511	-0.471	-0.523	-0.534	0.192	0.074	0.030	-0.432	3	wine 7	wine 7
44	-0.356	-0.403	-0.508	-0.486	-0.387	-0.531	1.101	-0.993	-1.348	0.851	-0.514	0.072	8	wine 3	wine 3

45	-0.166	-0.488	-0.5	-0.547	-0.565	-0.449	-1.167	0.275	1.135	0.017	-0.222	-0.134	2	wine 6	wine 6
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Colored 2D canonical score plot and 3D canonical score plot

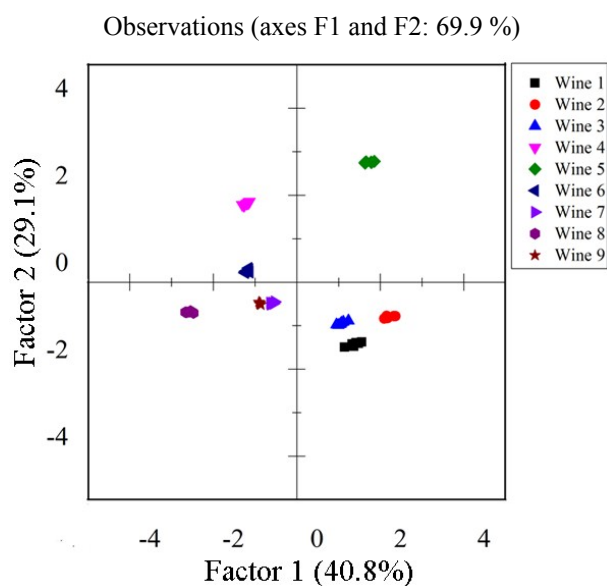


Fig.S11 2D canonical score plot for the first two factors of simplified fluorescence response patterns obtained with an sensor array of UCNPs@GDN, UCNPs@SO₃H, UCNPs@PO(OH)₂, mixture 1 UCNPs@GDN+UCNPs@SO₃H, mixture 2 UCNPs@GDN+UCNPs@PO(OH)₂ and mixture 3 UCNPs@GDN+UCNPs@SO₃H+UCNPs@PO(OH)₂ treated with different grape wines (10%).

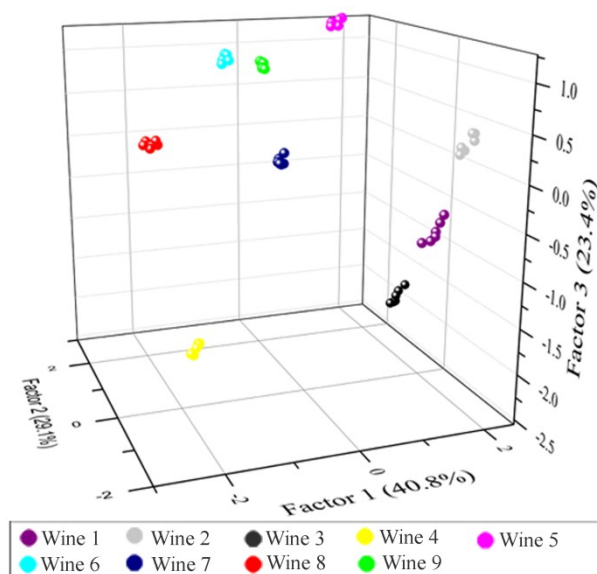


Fig.S12 3D canonical score plot obtained with the sensor array of six elements (UCNPs@GDN, UCNPs@SO₃H, UCNPs@PO(OH)₂, mixture 1 UCNPs@GDN+UCNPs@SO₃H, mixture 2 UCNPs@GDN+UCNPs@PO(OH)₂ and mixture 3 UCNPs@GDN+UCNPs@SO₃H+UCNPs@PO(OH)₂) treated with red grape wines (10%). Each point represents the response pattern for a single red grape wine to the sensor array.

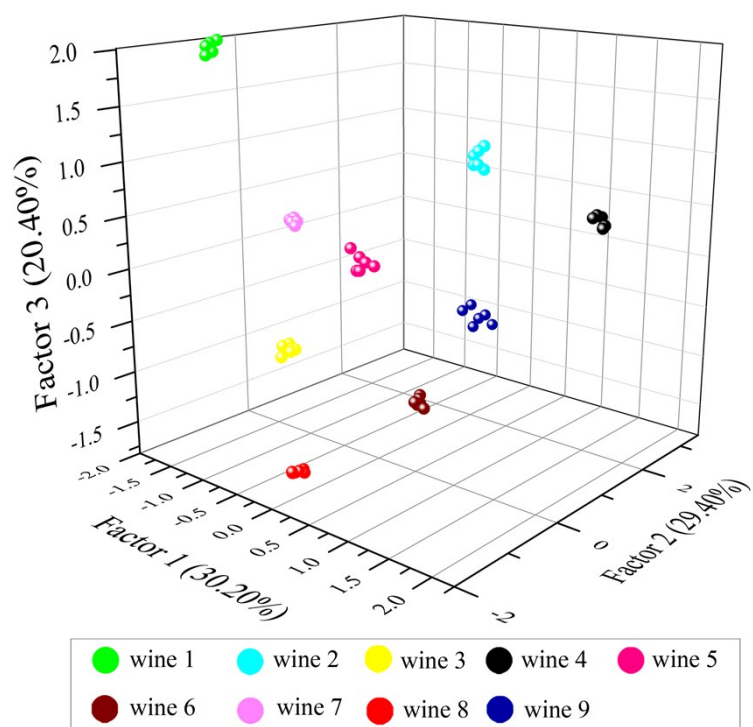


Fig.S13 3D canonical score plot obtained with the sensor array of six elements (UCNPs@GDN, UCNPs@SO₃H, UCNPs@PO(OH)₂), mixture 1 UCNPs@GDN+UCNPs@SO₃H, mixture 2 UCNPs@GDN+UCNPs@PO(OH)₂ and mixture 3 UCNPs@GDN+UCNPs@SO₃H+UCNPs@PO(OH)₂) treated with red grape wines (3%). Each point represents the response pattern for a single red grape wine to the sensor array.